Ladislaus Bányai:

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A volume dedicated to the
celebration of his 80th birthday

Editor: Victor Bârsan

Horia Hulubei Publishing House

Bucharest - Magurele, 2018

Cover designer: Tóth Erzsébet

Book designer: Radu Dragomir

Descrierea CIP a Bibliotecii Naţionale a României
BÁNYAI, LADISLAUS
Profile in motion: a volume dedicated to the celebration of his 80th birthday / Ladislaus Bányai; ed.: Victor Bârsan. - Măgurele: Editura Horia Hulubei, 2018
ISBN 978-606-94603-4-4

I. Bârsan, Victor (ed.)

94
929

This volume is dedicated to the 80th anniversary of Ladislaus Alexander Bányai - Laci, for his many friends -, (retired) extraordinary professor at the Johann Wolfgang Goethe Universität, Frankfurt am Main. This volume contains a biographic and a scientific part. The biographic one starts with a discussion with him. He recounts some significant events of his life including his remembrance about friends and coworkers.

The scientific part includes a selection of his papers with their short presentation, as well as a list of his about 140 scientific papers and 3 physics books. An English translation of his early (1968) but still relevant review on the quantum mechanical transport theory is also included.

Contents

Part I. The MAN

Part II. The WORK

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L. Bányai, A. Aldea, and P. Gartner: On the Nyquist noise. Z. Phys. B58,161 (1985)

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Part I. The MAN

Who are you, Mr. Bányai?

Conversations with Victor Barsan

VB: Dear Laci, I think, it would be interesting for those who want to learn about your activity, to get also some biographical marks. Life is strongly related to work and you went through many domains of science, got a wealth of personal experience in scientific centers, countries and political systems.

LB: I was born in Cluj, in a family of Hungarian intellectuals. My father, after his university studies in France, adhered in the thirties to the communist ideology and was for a while even jailed for communist propaganda. After the second world war, he had several important, but not political positions, for instance that of the Rector of the Bolyai Hungarian university in Cluj. My parents taught me internationalism, humanism, dignity and political courage. The latter contradicted their own political stances, however they accepted mine. By the end of his life my father became disillusioned with communism. His last words to me were: "I put my life on a wrong card!".

VB: Can you give a glimpse at the history of the today Babes-Bolyai University, which is Romanian, but has also a Hungarian section?

LB: Since the end of the 16th century Cluj is a university town. The Academies under the auspices of the catholic church educated in the German and Latin languages. In 1881 the Hungarian Royal Franz Josef University (the second in Hungary) was founded in Cluj. Among the students of this university were also famous Romanian intellectuals like Iuliu Maniu, Iuliu Hațieganu and George Coșbuc. After 1918 Transylvania became part of Romania and the Hungarian university was transferred to Debrecen. The Romanian King Ferdinand I University was founded on its place. After the Dictate of Vienna in 1940 the Romanian university was transferred to Sibiu and Timisoara, while in Cluj the Hungarian University was reopened. However, due to the 22-year long lack of continuity, with a heavy import of staff from Hungary. After the second world war and the return of North-Transylvania to Romania, two universities were simultaneously acting until 1959 in Cluj: the Romanian "Babes" -university and the Hungarian "Bolyai" -university. There was a cordial, good relationship between the two universities. My father for instance, during his time as a rector became life-long friends with Constantin Daicoviciu, the rector of the Babes University (well-known archeologist and historian of Dacian civilization).

VB: The multicultural character of Cluj always fascinated me, mostly for two of its aspects. On the one hand, two catholic clerics (roughly in the 15-th century) there firstly realized that the Romanian language belongs to the Romanic family. On the other hand here spent his youth Farkas Bolyai and here was born his son János, ("the unhappiest among the famous mathematicians"). It seems to me that this multicultural color of Cluj unfortunately has paled since your childhood.

LB: The name of the city in Hungarian is Kolozsvár, respectively Klausenburg in German (Claudiopolis in the middle-age Latin) and it was multi-ethnical along the centuries, though less today. During the middle-ages the population was mostly Hungarian and German. In the 18th century became mostly Hungarian, while in the 19th century it witnessed the development of the new Romanian bourgeoisie. Besides these three main groups there was an important French colony (refugees of the religious persecutions and of the French revolution). I should name also the neighboring Dutch colonist's village of Hochstadt. The last one supplied in abundance the city with first class vegetables and fruits (until Ceausescu!). The Jews played an important role both in the industrialization of the city, as well as in the cultural life, however most of them died in the Holocaust.

VB: The collective memory, at least in Romania, blames Horthy for the deportation of the Jews, but it seems, it is not correct.

LB: The anti-Jewish laws were introduced under Horthy and the deportation of the rural Jews began also under his rule. Therefore, it would be difficult not to blame him at all. It is also true, however, that after his anglophile youngest son presented him evidence about the real purpose of the deportations, Horthy stopped the deportations from Budapest. His son was kidnapped later by the Germans, Horthy's clumsy, slow efforts to get out Hungary from the war (he hoped the coming of the British instead of that of the Soviets) ended with the German occupation of Hungary and his arrest. The Germans brought the extremist "Nyilas" (Arrow Cross) party to ruling, The Nyilas government continued enthusiastically the deportation of Jews, even in the very last days of the war. The deportation of the Jews in Cluj occurred just three months before the Russians entered Cluj. It is worth mentioning, that the Nazis tried to hide the purpose of the deportations. They forced some of the deported Jews to send post-cards with nice landscapes about their well-being to their acquaintances in Cluj. On the other hand, in this period the Transylvanian catholic bishop of Alba-Iulia, Márton Áron (himself son of simple peasants), visited Cluj and held a sermon in the St. Michael cathedral against the persecution of the Jews. After the war, under the communist rule, he was arrested and kept under home-arrest until his death.

VB: The bright personality of bishop Márton Áron was subject of many reports in the Hungarian broadcast of the Romanian TV (with Romanian subtitles) in the early nineties. But let us come back to the story of your life.

LB: The first four classes I learned in Cluj in Hungarian, my mother tongue. The fifth class I continued also in Hungarian, but in Bucharest, after my father in 1949 had to move to the capital city as one of the leaders of the MNSz (Hungarian Popular Union) and after its forced dissolution as counselor in the ministry of education. In the 6th class I moved to a Romanian school for boys. Until that time, my knowledge of the Romanian language reached only a rudimentary level. I learned Romanian in a summer vacation camp. Nevertheless, I had to fight for years with the grammatical genders, lacking in Hungarian.

The 7th class I started in a special school for the children of the political elite, with special emphasis on the Russian language. This school had a lot of highly competent soviet teachers. We really loved them as good pedagogues. Contrary to the expectations of the regime, with a few exceptions, this school did not produce fervent adepts of the communism, or of the Soviet Union. However, I learned the

Russian language and got respect for the classical Russian literature, since we went over it according to the original Russian textbooks. Later I was able to read Solzhenitsyn Ulitskaya and others in original, as well as the rich amount of physics books in Russian.

My break with the political indoctrination occurred immediately after my arrival for study in the fatherland of socialism in the famous year 1956. "This is what we must build up?" I shall tell more about later, in describing my student years.

Strange enough, I discovered my skill in mathematics by a teacher with very poor formation (former sport-teacher). She run in a dead end by the proof of a geometry theorem at the blackboard an I helped her to get out of the catch.

Afterwards I became aware of my mathematical success and by the written tests I solved the problems of all my friends. To avoid cheating the teacher gave everyone a different problem but did not care to look at and I had enough time to do the whole job.

I had also artistic ambitions and painted in the Pioneers Palace under the guidance of Spiru Chintilă. Unfortunately, he used me for propagandistic paintings like Stalin's portrait and Tito's caricature and I lost my interest in painting. The last kick was my acquaintance with Eugen Mihăescu a true young artist, who later illustrated "The New Yorker". Now, at old age sometimes I again play around with painting.

I became a regular reader of the "Gazeta de Matematică si Fizică" (Journal for Mathematics and Physics, a publication for schoolchildren), solving a lot of the proposed mathematical problems just for fun. I remained until now just a passionate problem solver. Pure mathematics was not my domain. However, as I learned that mathematics plays an important role in physics, I have chosen physics for my future studies. A book just published at that time in Russian by Jungk: "Thousands time brighter than the sun" about the history of nuclear physics played a peculiar role in this decision.

Here it is the right place to relate an important aspect of my school years. My father was moved again to Cluj as rector of the Bolyai University from 1952 to 1956. However, I succeeded to convince my parents to let me further remain in Bucharest at the preferred school, getting full lodging in a room of an acquaintance of my parents. Thus, starting at the age of less than 14, I got my independence.

After graduating from high-school (that meant 10 years only in those times) I got a Diploma cum Laude and the opportunity to start my university studies in the Soviet Union. However, since there were no places in physics, I opted for the Physics Department of the Bucharest University.

VB: Were you not rather attracted by the home city of Cluj?

LB: Of course, I felt and still feel myself deeply connected to Cluj (where I spent also all my holydays), but the Bucharest University had a better fame, linked to the existence of the Institute of Atomic Physiscs (IFA - Institutul de Fizica Atomică in Romanian) and the names of Țiteica and Hulubei. That's why I opted for Bucharest.

My first impressions were not positive though. In the first year we had no good teachers. The one who read Analysis we called just “the convergent”, according to a known theorem, since he was monotonous and bounded (in Romanian it means also “narrow-minded”!). The Mechanics lecture was elementary and annoying. In one of the Heat Physics lectures the professor showed us different thermometers lying on the desk. “This is the thermometer with mercury, this one is with alcohol, and this one with sand …” Due to a mistake of his labor- assistant a clepsydra was laying on the desk. We as students were very malign and defined the unit of stupidity after his name, recommending for everyday use only its pico- variant. On the other hand, together with other colleagues I frequented on free basis the beautiful Calculus lectures of Miron Nicolescu for mathematicians.

During the first semester I was informed, that within a few months I could continue my physics studies in the Soviet Union. In this first semester I had to share a room with 24 colleagues in the students hostel with primitive beds on straw mattresses with bedbugs. The dishes in the refectory were hard to swallow. Under these conditions I decided to spend these last months at home in better conditions and moved to the Babes University in Cluj. I did not want to go to the Bolyai to avoid being the rector’s son. The Babes University I enjoyed for his nice atmosphere, but I cannot recall very much about it, since after two months I went to Leningrad.

VB: Tell me about your time as a student in the USSR.

LB: Of course, in Leningrad (today Sankt Petersburg), as a foreign student I got much better conditions. I shared a clean, well-heated room with a Chinese, a Czech and two Russians, all of them studying journalism. At least one of the Russians of course was a KGB stoolie. The student’s refectory was not good, but acceptable. Sometimes I went to a restaurant to get better dishes.

The Physics Department was less attractive, it was placed in the former stables of czar Peter the Great. The lavatories had seats in tandem with a low fence in between. One could conversate with his neighbor (student or teacher). A lot of primitive things shocked me. The housing was a big problem in the cities. Several families had to share a big apartment with only one bath-room and kitchen. The ugliest impressions came however from the stories heard from my colleagues about the situation outside the big cities. It was the time of the twentieth Conference of the Soviet Communist Party with Khrushchev demystifying Stalin and revealing his crimes. This was a very hot political period.

I found interesting as well as useful, that all physicists had to learn minimal technical skills (technical design, descriptive geometry, mechanical, glass and electric workshops etc.). The general physics and mathematics lectures were taught by lecturers according to the good standard books. The professors did not lecture at undergraduate level. Accordingly, one did not need to take notes or to read miserable heliographed texts, as the state of the art was in Romania. Strangely enough, we foreign students had to participate at the military training together with our Russian colleagues. We learned how to mount and unmount a Kalashnikov and similar things. (Later in Romania at the student’s military summer-trainings we learned almost nothing useful. It was just a useless tormenting.) Since I already spoke well Russian, I had no difficulties passing exams with good grades. Unfortunately, I did not reach the interesting lectures, compelled to leave soon Russia.

During the Hungarian revolution in the fall of 1956, we kept us informed about the events by listening Hungarian radio, as well as Free Europe and Voice of America. In Leningrad the Hungarian and Romanian broadcasts were not jammed. The jamming emitters had a finite radius of action and even in Romania there were some areas without jamming.

VB: Could You please explain for the younger readers something about the Hungarian revolution?

LB: After Stalin's death and the disclosure of his crimes in Hungary, agitations of left-wing intellectuals and students started to reform socialism. Due to the rigidity of the system finally a revolt emerged. In Hungary a new government was installed under the leadership of Nagy Imre (an old communist very popular for his aura of a reformer). Of course, the revolt was joined also by all anti-communists including also extreme right. Nagy demanded the depart of soviet troops and declared that Hungary leaves the Warsaw Pact. The answer was an armed soviet intervention, that after heavy fights on the streets of Budapest reinstalled the old regime and arrested the Nagy government. They were brought to Romania and lived some years detained at Snagov, a small town not far from Bucharest. Later Kádár, the new leader installed by the soviets, arranged a political trial and Nagy together with some members of his government were executed. Nevertheless, this revolution was not in vain. After a few years of terror, the Kádár government had to loosen the grip. In a well-defined frame in Hungary has been established a status that in the west one called "goulash communism", with a higher standard of life and a relative freedom of expression and travel. Looking from Romania, it appeared as a dreamland. The Hungarians themselves said that they are living in "the gayest shack of the camp".

But let me return to my own story:

Not only in Hungary, but even in the USSR there were student movements in 1956, with spontaneous readings of non-conventional poems at the statue of Mayakovski. Some unorthodox novels got published like "Not the bread alone". Our Marxism assistant wanted to learn from me (between four eyes) about the events in Hungary. Later I learned that the students were boiling also in Romania and this turned into many arrestations, especially in Transylvania. At the general meeting of the Romanian students in Leningrad, some students of philosophy required more democracy. Soon after the defeat of the Hungarian revolution, a special committee arrived from Bucharest inquiring the political behavior of the Romanian students in the USSR and at the same time lecturing us about the right policies of the Romanian communist party.

Many students and doctorands (called "aspirants") were immediately called back to Romania. Among them my friend from Cluj, Stephan Fischer. I was falling slowly in the trap. I had to face the committee's accusation that I am a Hungarian nationalist and, in this way, to harm also my father. Of course, I denied it, but they insisted "perhaps subconsciously?". What could I answer to such insinuations? Finally, I was asked if my level of political education deserves getting further a stipendium in the USSSR. As I was naïve and young I signed it, and that was it. My "unfriendly" comments regarding the soviet intervention in Hungary were transmitted by the KGB to this committee and in March 1957 I received a telegram ordering me back into Romania to continue my studies at the Bucharest University.

Totally I spent only a year in Leningrad. Before leaving the vice-dean Novozhilov, a theorist, advised me to continue in Romania theoretical and not experimental physics.

My departure caused lots of tears, since I had to leave behind my love (later my wife for already 60 years). We met in the Bucharest-Moscow train among my classmate girls.

VB: How long did this separation last?

LB: Until she got her engineer diploma in 1960. However, she came home in every semester break and we spent those times together. After her return we married, although I was still in my last year of studies. (l lost a year due to a punishment I am going to tell about later.)

VB: Thus, you returned to Bucharest …

LB: Indeed, however this time I was living with my parents, who were again transferred to Bucharest. In the physics department now, I attended interesting lectures and this time we had good teachers and many very good assistants.

I was lucky having gifted colleagues like Adam Schwimmer, who became later a very known and appreciated theorist after he left Romania and got with four years of retardation his degree in Israel. We competed in problem solving. The most important lecture was the electrodynamics of Prof. Valeriu Novacu. His lecturing itself was not good, but he had a good choice of the thematic. Often, he was replaced by his lecturers (Klarsfeld, Eftimiu) who gave a good performance. I took no notes but started to read the best textbooks available both in Russian as well as in English (already available in the library!). These lectures arose my passion for theoretical physics.

I have a nice recollection about Chaim Iusim (who left later for Israel), lecturing about statistical mechanics, as well as about Viorel Sergiescu (emigrated to France in 1972) lecturing thermodynamics, both from Prof. Șerban Titeica's chair. Prof. Radu Grigorovici's lectures on optics were also remarkable due to the many impressive experimental demonstrations.

VB: People lived with the illusion, that after Stalin's death, things should improve …

LB: Indeed, there was a hope until 1958 when the big cleansing started. That summer Gheorghiu-Dej, prime secretary of the PMR (Romanian Workers' Party) started a big cleansing of cadres overall. People were fired for any or no reason at all. Gheorghiu-Dej was afraid, that along the destalinization process, Khrushchev might want to eliminate him also. Therefore, he eliminated his opponents from the party leadership the "rightist deviationists" Miron Constantinescu, Iosif Chișinevschi and Gheorghe Apostol.) and eliminated all those suspected for loyalty to the soviets. (Before he killed already his competitor Pătrășcanu.) To hide the purpose of this operation he promoted chaotic reprisals. Afterwards he rehabilitated many of the victims.

That summer my parents were fired from the ministry of education and my later father-in-law from the ministry of commerce.

I was expelled from all the universities for “adverse attitude towards the RPR (Romanian People's Republic)”, due to my old sins in the USSR. Fortunately, thanks to the empathy of most of my colleagues I was not excluded from the UTM (Union of Young Workers) and this was very important in those times.

The only way for saving itself was to try rehabilitation by “work at the basis”. This rehabilitation was nothing more but a period of humiliation. I went as a lathe apprentice into a factory. In the beginning the workers looked at me with suspicion. They felt, I am different. After a very short time I got enough qualification to get respected also by the older workers. Of course, for me it was easy to learn the art from a textbook and I had also a natural skill for technics. I enjoyed solving not only theoretical, but technical problems alike. Later I repaired my car Dacia in our courtyard myself.

In the fall at the meeting of the Romanian students still returning to continue their study in the USSR one has exposed in a speech “the right deviationist clique of Fischer-Bányai”. Many years after indeed we both deviated to West-Germany.

VB: What happened to the “deviationist” Fischer in that summer of 1958?

LB: After the “work at the basis” as a frigorific technician and “rehabilitation” he got his diploma at the Bucharest Institute of Cinematography and worked as camera man and editor at the Hungarian TV in Bucharest. He left for West-Germany before me and worked for the German TV. He made a lot of very interesting documentaries about many countries. Even one about the Papuans in the jungle. He was a well-known and esteemed person among the Hungarian intellectuals in Romania as well as in Hungary. He was a charming and talkative person. We stayed good friends until his death two years ago in Aachen.

VB: What happened to you after the nightmare of “rehabilitation”?

LB: Next year the atmosphere got lighter and after my worker colleagues have lifted my UTM (Union of Worker Youth) sanction, I could continue my university studies. Nevertheless, at the Physics Department I was continuously victimized by the party secretary Smaranda, who treated me as a “spiteful element”. He forbad me a practicum at the IFA and any path to an academic career after Diploma.

During my work in the factory, as a headstrong I read many books on quantum mechanics and back in the university I felt myself above the level of my colleagues, even above some of the professors. I worked out my Thesis under the guidance of the lecturer Meinhard Mayer form Novacu’s chair about the renormalization group in the quantum field theory. He had a good reputation after signing a paper with Bogolyubov at Dubna and after publishing the first book on quantum field theory in Romanian.

VB: It would be interesting to explain the meaning of IFA and IFB. These acronyms look amazing, since the alphabetic order suggest a certain hierarchy, that incidentally was not fictitious.

LB: Indeed IFA (Institutul de Fizică Atomică) founded by Prof. Horia Hulubei (in Măgurele near Bucharest) had the first rang as importance (atomic reactor, betatron) and dimensions, but had very severe political criteria for the personnel. Prof. Eugen Bădărau founded another more modest institute in Bucharest

(Institutul de Fizica București) with research aims in gas discharges (a precursor of plasma physics) and solid state, with somewhat more liberal political personnel policy.

Let me return to my griefs: although I had excellent grades, due to my political case history, I got only a job in the radio-active logging by Ploesti, which I turned down. Six month I was hanging in the air, living on the salary of my wife, who worked as engineer at IOR.

However, I got the support of Prof. Valeriu Novacu deputy member of the Romanian Academy, head of one of the chairs of theoretical physics at the Bucharest University, member of the central committee of the communist party and governor at IAEA). He was an acquaintance of my father from the illegal times of the communist movement before the war. He was one of the idealistic communists and helped gifted young physicist without regard to their political sins. He stems also from Cluj and became, like my father, communist during his studies in France. After 6 months indeed, I succeeded to join the theoretical physics section of the IFB, that had been recently founded by Novacu.

VB: How did it look this theory section and what kind of activities were on in the IFB?

LB: This section at that time comprised only six researchers. The unofficial boss was Viorel Sergiescu, specialized in solid stat theory (noise in solids), a former lecturer at Țiteica's chair. During the 1958 cleansing he lost his faculty position, went first to IFA, but landed finally at the IFB. There were also other researchers who got transferred from IFA on political grounds: George Ciobanu worked on transport theory and Alexandru Glodeanu on the theory of impurity states in semiconductors. There was also Peter Handel, one year elder, working on the 1/f noise, that remained his life-obsession. And there were the two novices: Lucian Brînduș and me.

The orientation was determined by the experimental solid-state research in the institute, but without any direct link.

I met here again Prof. Grigorovici, who also lost his faculty position and led an experimental lab on semiconductors, something completely new for him.

VB: How did you integrate in this collective, and who guided you?

LB: Sergiescu showed me a list of solid-state theory books, but I interacted sparsely with him. I respected him for his solid-state book in Romanian, as well as for his courageous political mentality and his broad culture.

The two novices however cooperated from the beginning and we succeeded to publish already in the first year in science two papers on the quantum theory of transport in the Studii si Cercetări de Fizică (Studies and Research in Physics) and Revue Roumaine de Physique. The first one, although correct, was a typical beginners work. The next paper I wrote already with a senior researcher, George Ciobanu, on magneto-optical phenomena treated with the modern Green functions method. It was published in English in the just started Physica Satus Solidi. I was joined soon this time as a senior by Alexandru Aldea from Grigorovici's lab and we published a paper in the Physical Review about the Hall effect in impurity band conduction.

VB: Soon you started to interact with the experimentalists from the IFB.

LB: Seemingly, it was rumored that there is a new researcher on his own feet and the experimentalist, whose contact with the previous theorists failed, tried to contact me. First it was Cruceanu, but after some discussions with him I saw no common interests. Then one nice summer evening after some event in the institute we walked together with Grigorovici , Cruceanu (the scientific secretary) and Rodeanu (the party secretary) along the Dorobanți street on a common home way. The conversation was mainly a monologue of Grigorovici about his extremely interesting experimental findings with thin layers of amorphous Germanium. First at all he observed, that these had similar properties to the crystalline semiconductors. On the other hand, the activation energy of the conduction was much greater than the optical gap. He tried to convince me to solve theoretically this challenge.

VB: How did you react?

LB: The story of Grigorovici raised indeed my interest and I tried to build up myself an image about the amorphous semiconductors. I looked around at the literature but found nothing relevant. Unfortunately, I missed Anderson's paper. However, the Coster-Slater model seemed to me the right tool. I tried to imagine building an amorphous material out of a crystal by moving atoms out of their positions. Every such a displacement had to create a localized state eliminating in the same time one from the bands. In this way I concluded, that this leads to partially "eaten" nonlocalized (conducting) bands and a lot of localized states in the former gap. These states do not conduct, but still contribute to optical transitions. Therefore, the conduction gap should be greater than the optical one.

VB: How reacted Grigorovici to this theory of yours?

LB: I made a presentation in the seminar of Grigorovici's lab, preceded by a description of the Kubo formula. Until then everyone thought only in terms of Boltzmann' theory. Grigorovici was very excited and enthusiastic. He encouraged me to send a short paper about my theory to the international semiconductor conference in Paris. I did it, adding also another piece of theory to it. I extended the tight binding model to a disordered lattice of atoms for the estimation of the band conductivity using also the experimental X-ray results about the lattice structure. It came out surprisingly good, but I could not really justify the approximations I used.

Grigorovici was extremely interested to have me with him at the Paris conference, but with my background the chance to get a pass was just zero. Nevertheless, he made a desperate attempt. Without telling me about it, he visited my parents knowing, that they are old communists and convinced them about my scientific gift in the naïve hope they may move something through their connections. Anyway, my parents were very impressed.

Although I could not attend the conference, the paper appeared in the Proceedings and the later Nobel prize winner Sir Neville Mott read both the paper of Grigorovici and of mine. He made a lot of publicity to my theory and of course thereafter it was quoted in hundreds of other papers.

VB: As far as I know, the crowning of the success occurred at the first international conference on amorphous semiconductors held in Bucharest.

BL: Grigorovici personally met Mott and as an old spice intellectual got his respect. I do not know how it did happen, but Grigorovici succeeded to organize this first amorphous semiconductor conference in Bucharest and Mott itself attended it. At that time my mind was already on elementary particles.

What I do know however is that this project could not have occurred without the full rehabilitation of Grigorovici. There were some intrigues against him and at a given time he went to counter attack. He assembled all his publications in a nicely bound book and with this in the hand succeeded to obtain an audience by Stefan Voitec (one of the secretaries of the communist party, a former social- democrat, colleague of Radu's father, who died in prison). The time was probably rife and Voitec perhaps wanted to calm down his own conscience. Anyway, he succeeded to promote Radu Grigorovici to be a member of the Academy and nominated him as vice director of IFB. Without these preliminaries the conference had no chances at all.

Mott visited also the IFB and gave a talk speaking at a given moment about professor Bányai. He was shocked when he heard the audience laugh. Then, somebody explained him, that Bányai was just a young research associate, not a professor. He replied, that in America every young physicist is a professor, at least assistant professor.

VB: To my knowledge Mott invited you to Cambridge?

LB: Indeed, Mott invited me as a postdoc to Cambridge, but I politely declined it. It may look strange, but I had a clear motivation in my head. In the meantime, within the theory section Novacu initiated also a research group on elementary particles. I plunged myself into this new field having already several published papers at that time. Even more, I obtained by competition a six-month grant at the ICTP (Trieste), and I knew I had more chances to obtain a pass for that.

Besides this already important and passionate argument, there was another even one weighting more heavily. I could not enjoy being under the authority of the famous Mott dictating me what to do. Besides I did not like at all his style. Sure, it would have helped my career, but I would have lost my freedom in choosing my research topics and my coworkers. In Romania, under the conditions of a good connection to the external scientific word (what seemed plausible at that time) I felt myself in a more advantageous position.

Nevertheless. much later as I returned to solid state theory and worked together with Paul Gartner and Alexandru Aldea on hopping conduction, I had a fruitful correspondence with Mott.

VB: Please tell me more about that elementary particle theory group.

LB: Slowly Novacu brought a lot of new theorists into the IFB. First came Hans Raszillier (my very gifted former university colleague), then Vladimir Rittenberg, a former assistant at the chair of Novacu, who lost his university job due to the political purge. Together with Niky Marinescu (also a former colleague of mine) who came over from Grigorovici's experimental lab, we formed the nucleus of the elementary particle theory group, later joined us also the younger Petre Diță, Nucu Stamatescu and Luca Mezincescu . The solid-state theory group grew also. Mircea Bundaru came (a physicist with very bright mind), Tony Fazekas and Alex Friedmann. At these new dimensions of the theoretical physics section one could already organize regular seminars with well-prepared reports, published in a collection of volumes published by the Romanian Academy. The dynamic soul of the elementary particle group was my 4 four years older friend Vladimir Rittenberg. He was an enthusiast, full of energy and thirst for knowledge and he had a true gift for organizing. A person with rare intelligence and many human qualities. Around him assembled young physicists not only from Bucharest, but also from Cluj and Craiova. The two of us quarreled often about physics, but this did not at all affected our friendship, but just served for a better understanding. (Later I learned about the Securitate records of my phone conversations full, with mysterious terms like "Gamma function", "beta", "bootstrap" and so on).

Rittenberg organized a weekly informative seminar on particle physics about new papers in the Physical Review Letters. Each of us choose an article, worked it out and presented it the next week. He also organized the editing of preprints of our own works and that way also an exchange of preprints with important scientific centers.

After leaving Romania, Rittenberg spent 5 years in the USA as a postdoc and became a university professor at the Bonn University, having a wide international recognition. He died recently at the age of 84, but just a few months before, already having lost his mobility, he still worked on physics with his Brazilian and Russian coworkers.

Here I would like to tell a few words about a phenomenon, typical in those years for the science in socialist countries. I was already at the IFB, but Rittenberg grew crystals in a technical institute (ICET), while the excellent Adam Schwimmer was substitute teacher in Slobozia, a small town at about 100km from Bucharest. He was expelled also in 1958 from the university because his family applied for emigration to Israel. Together with other physicists interested in elementary particles, I organized a private seminar at my home, trying to follow the evolution in that field.

After Rittenberg, Marinescu and Stamatescu left for the West, the group became smaller and after the relocation of the institute, under the new name IFTM (Institute for Physics and Thechnology of Materials) to Măgurele it was even split, part of it went to the IFA (renamed as IFIN (Institute for Physiscs and Nuclear Engineering). I continued to work for a while with the 8 year younger Luca Mezincescu and we remained good friends till now even being at a big distance from each other.

VB: Can you tell me more about the Novacu's role in the theory section and in the elementary particle group at the IFB?

LB: As I already told, Novacu not only founded these collectives, but by his personal intervention he brought here young gifted physicists regardless of their "personal files". In a way he offered them a "political asylum". He was an idealist communist of the old guard, who rejected any kind of discrimination.

Novacu had very clear moral principles he always kept. He was against exploitation and never signed works of his coworkers. Therefore, had no scientific publications at all. His enemies in the Academy hated him because he was a communist and tried to mob him. Grigorovici heard about it and advised us to put his name at least on one of our papers, otherwise the very existence of the theory section was endangered. Novacu, like a virgin, held against, but in the end, we convinced him to do it for us the theorists of IFB.

Novacu was always present in our seminars but spoke rarely and was not nasty if we rebuked him. He gave us free hands in our activity.

Although he was not a remarkable physicist, his role in the development of theoretical physics in Romania was extremely important.

VB: What was the general ambiance in Romania, that allowed this evolution of the scientific activity?

LB: All this occurred in the frame of a slow opening under Georghiu-Dej regime. The collections of the physics libraries were filled with western books and journals. We succeeded to have in the IFTM even a better library than the one I found in Frankfurt. Of course, we alone chose the titles. Already in the university library one could read original books in English, not only the Russian translations. Nevertheless, the Russian physics literature (translated, or original Russian) played an important role for my generation. The Russian books (ignoring copy rights) were very cheap, covered almost all domains of physics and contrasted with the poor print quality of the Romanian books. I collected hundreds of physics books. Of these I succeeded to bring about a hundred to Germany.

Moreover, our generation had already the possibility to publish in western journals and I was among the first who published a paper in The Physical Review.

We got foreign visitors. At the beginning only from the socialist countries but later also from the USA. The doors opened a little bit for scientific visits in the west.

The previous after war generation of scientists had to work under extremely difficult circumstances. Especially those who did not managed the Russian language were restricted to a few poor translations. The libraries were empty. Publishing was restricted to local journals and of course any link to the west was blocked. No conferences, no visits and no visitors. Not to speak about the ideological dogmas. Quantum mechanics was hardly accepted because the indeterminacy relation. Blohintsev’s famous book, translated into Romanian had to include a chapter invoking Lenin. I do not have to recall the true political persecutions.

I would say: I was lucky not to be born earlier!

VB: You sustained Your PhD Thesis at the age of 29 with a subject concerning the theory of elementary particles. Tell me something about the most interesting part of this work.

LB: I shall not mention the content of the Thesis. Today it lost its importance. However, I will tell You about the work done on this Thesis, since it influenced my later evolution.

With Novacu’s help Rittenberg and me could work two weeks at the DACIC computer of the Mathematical Institute in Cluj. It was a self-made computer built up by enthusiastic local engineers and mathematicians out of occasional spare parts of different origin and electronic tubes. It took two big rooms of space and was noisy as a tractor. After several hours of work, it had to be shut down after having saved the data. The neon lamps showed only 0 or 1 (not lightening or lightening). It allowed programming in a primitive assembler language. The output was on punched bands or on an old telefax inherited form the railways. A mathematician explained us on the first day the meaning of programming and the use of the machine. Thereafter we had the monster in our own hands. This was the start of my flirt with the computers and they fascinate me up to this very time.

VB: In this period of opening of Romania you had the chance to "go west" frequently and you spent even there even longer periods. How did influence you this new experience?

LB: Of course, the visits in Trieste at the ICTP (the first one of 6 months, followed by shorter ones each of two months) impressed me very much and allowed to develop contacts with western physicists. I got also several invitations. One of them was by Ziman for a postdoc position at Bristol, but like by Mott I declined it, since I was at that time interested strictly in elementary particles. However, I visited several institutes in Italy giving seminar talks. I gave a talk also at CERN. While at the ICTP I published some papers, I worked out there. I lived with the hope that due to these contacts with the physics world I can further work in the field of elementary particle theory while still residing in Romania.

In 1974 I went to Marseille at the Centre de Physique Theorique du CNRS for a whole year. Luca Mezincescu at the same time went to Dubna for several years with my personal recommendation to Ogievetsky (during a short visit I had in Dubna).

When I got back from France, I decided to turn slowly back to solid-state physics. This change was motivated by many arguments. First of all, I understood that unfortunately, with the weak contacts we had, we cannot keep the pace with the rapid progresses in the field of elementary particles. Secondly, I felt already, that the relative opening in Romania is coming to its end (the "mini cultural revolution" just started). The third thought was also my disappointment regarding the quantum theory of fields. It made big progresses predicting and discovering a lot of new elementary particles, but there was no hope to compute anything, while the link to the rest of the physics was rather confuse.

VB: A change of the research orientation might be rather traumatic. It seems, with you it was quite different.

BL: It was not difficult. I enjoyed changing from time-to time my scientific interests. Otherwise I would get bored. On the other hand, on has a big advantage entering a new field. One is not yet impregnated with the prejudices that impede progresses forward and one might bring in methods and approaches from other domains. Changes of research objectives accompanied my whole life even in Germany.

Back in Romania I worked for a while on two fields. With Paul Gartner and Alexandru Aldea on solid-state theory (hopping conduction) and with Sorin Mărculescu from Țiteica's chair, on quantum field theory. According to my opinion, he was the most talented theorist from this chair. Of course, with the exception of the late Andrei Mezincescu, who choose to change soon to the IFTM, since he was a creative mind and had no people to talk with there and did not want to waste his time with teaching. Besides, he heard from his younger brother Luca about the pleasant, fertile atmosphere in our section. Previously he spent two years at the Moscow university by the famous Tyablikov, who died before Andrei could have had finished his PhD. Andrei was an encyclopedic mind with profound knowledge of physics and mathematics. He could immediately name the paper, where one may find the right answer to the given question. The only problem of communication with him was the enormous amount of links opened by him and one had to bring him back to the initial subject. Andrei spent two days in prison during the anti-Ceausescu upheaval and afterwards led the IFM for a while. I invited him to Frankfurt to give a talk, mostly for mathematicians. He spent also some time in the USA and the two brothers published their only common paper, which is a dear memory for Luca about brother who died so early.

During my collaboration with Mărculescu we had often chats with Țiteica, but he avoided recent aspects of physics, even those related to his famous thesis. I remarked a similar stand by other famous physicists in advanced age as they could not follow any more the evolution of science. Now I am also old, and I can understand very well their behavior.

In the same frame I recall the oft heard allegation that Țiteica did not create a Romanian school of theoretical physics. The truth is that, after losing the link to the world it was not any more possible. While his former colleagues by Heisenberg revolutionized physics in the USA, he sat in a corner completely isolated. Later maybe he could have compensated this, but he did it not. As I entered the IFB he was 53 old and for me it was already clear, he lost the link. On the other hand, he disposed of no such strong political position as Novacu, nor had he the energy to organize something. Țiteica was a disappointed man, looking for refuge in music and only on rare occasions showed the shining of his remarkable mind.

VB: Many of your colleauges left Romania and the institute underwent many changes, IFB became IFTM. The political atmosphere also changed drastically. How did that affect your scientific activity?

L.B. Indeed, after the "mini cultural revolution" of Ceausescu many things changed in Romania and our Institute moved to Măgurele near IFA. Despite the serious worsening of the situation, without being a party member I still enjoyed support from many persons in key positions. On local level I had to mention the late Florin Ciorăscu, a former director of the IFB a man of outstanding qualities and Alexandru Glodeanu the party secretary in the IFTM. My big Maecenas however was Ion Ursu the president of the CNST. I enjoyed direct access to him and he even kissed me at one of these encounters. I was also his intermediary connection to Abdus Salam (Nobel prize winner and director of ICTP). Among others I transmitted to Ursu Salam's initiative to invite Chinese physicists to Trieste through the Romanian channel. As a major action I got the support of the ICTP to organize in Bucharest the first international conference on hopping conduction.

The theoretical physics section and the IFTM itself still conserved a pleasant atmosphere of work. We lived in an ivory tower amid a gray society. In the IFTM I worked with Gartner and Aldea on hopping transport and we won the prize of the Academy. With Gartner I wrote also a series of papers about purely theoretical problems at the edge to mathematical physics like the quantum mechanical proof of the Clausius-Mosotti formula, the Meissner effect and the connection between the kinetic and hydrodynamic levels of description of semiconductors. Mircea Bundaru, Andrei Mezincescu and Paul Gartner established a bridge to the excellent mathematical physicists of the IFIN: the late Nae Angelescu, George Nenciu and Vlad Protopopescu. We hold several common seminars with them.

Otherwise, the connection to the external word was very reduced and we got less and less foreign journals in the library. For a xerox copy one needed a special approval. There was a shortage of writing paper. Instead coffee we drank a mixture of chickpea and rye coffee substitute. One mobbed us with meaningless “voluntary” works at the weekends. During the draught we had to irrigate the tomato fields with cups of water. The toughest torture were the winters without heating in the institute building. We worked with mantles and gloves on and hid small electric heaters under the desk.

VB: How it went the cooperation with the socialist countries?

LB: There were agreements between the academies including visits and cooperation. Although they did not compensate the lack of links to the West, they played an important role. I went for short visits to Czechoslovakia, Hungary and the USSR.

We had two important visitors from my point of view: Vladek Capek from Czechoslovakia and Harald Böttger from the GDR. With both I got soon a common language not only in physics, but also in politics. I really don’t know by what kind of miracle we had an immediate confidence to each other and remained good friends even after my emigration to Germany. With Capek, Aldea and Gartner we wrote also a paper to explain the very low temperature plateau of the Seebeck coefficient in amorphous semiconductors. This effect was observed also by Lili Vescan and I heard about during my morning coffee drinking by Tia Belu directly from Grigorovici.

At that time, I still had a close relationship to Grigorovici. He wanted to convince me to become his successor as the “virtual boss” of solid-state physics in Romania. I decidedly refused his proposal as I had no ambitions for any leadership. I wished to do research for my own pleasure. My wife always used to tell about me that I never worked but just played my whole life long. There is some truth in it.

VB: You had the chance after many years to visit again the USSR. What kind of impression you got this time?

LB: The one-week visits each in Moscow and Leningrad proved to be very interesting as well as from the scientific side as from the general view of the country. At the Moscow university I met Igor Zvyagin and we became friends. He was formerly a colleague with Ciobanu at de Nozières in Paris. Later he was one of the external referees for my Professor title in Frankfurt.

In Moscow I visited also FIAN (Physics Institute of the Academy of Sciences) and got an enthusiastic receival by Maximov and his coworkers. With them we had some common topics on the theory of dielectric response. The welcome resulted in a lot of vodka drinking. I managed to keep myself sober, but Maximov drunk out of his mind and one stopped him at the Metro entrance. After recovering a little bit, he took me against all my resistance to his home outside the city to show me as an outer space being to his family. Only before returning to Moscow he told me that the place he was living was forbidden for foreigners and I must keep my mouth closed during my travel. I felt a little bit cold in my back but succeeded to return without incidents.

In Leningrad I gave a seminar talk at the Joffe institute and got new friends Efros and Shklovsky. Both invited me to their homes. Their living conditions were decent. One of them had a renovated flat separated from a former huge apartment ("kommunalka"), the other one was the owner of two small ("koopertivnye") flats in a new building. One of them served as his office. The theorists worked mainly at home and met at the institute only for the seminars.

I made a visit also at a former colleague of my wife. They also lived on a decent standard. However, during the dinner her man looked often out of the windows to check if their car, they brought out of the garage for me, was not stolen. There were still some bizarre things, as the long years of wait for a car and its use only in the summer collecting mushrooms in the woods. The way they could spend their state organized summer vacations sounded also strange to me.

Compared to Ceaușescu's Romania, things looked still better. The people carried not any more the permanent fear with them, discussing freely. I was lodged in a new modern hotel built by the Finns on the Neva river-shore. (But the supervising ladies still sat at every floor at their small tables.) Both Moscow and Leningrad were brightly lightened in the night and everywhere I saw modern color TV-s.

Another funny experience I had as I went on my own money, out of curiosity to the amorphous semiconductor conference of the socialist countries in Chișinău. In the Ungheni border station the soviet officer could not conceive how one can combine two such things as a scientific visit and a private pass. As a private person I had to go through the Intourist offices and as a scientist I must have had an official pass. He led me to the commanding officer's office. I showed him my invitation to the conference, but at night around two o'clock he could reach nobody. I told him in an amused tone, that I am ready to take the next train back to Bucharest to relate about not being allowed to participate at an international conference. After that he got angry and greeted me with "idite k chortu" and I could continue my travel.

The Moldavians were very kind with us and again I met a lot of relevant soviet physicists like Dneprovskyi, with whom I kept also later the contact. The city itself was not very attractive. I had an interesting private visit by a Moldavian family and learned a lot about their fate and lives.

VB: Although you spent longer periods in the West, even together with you wife, you always returned to Romania. What determined you later to change your mind and how did you managed to emigrate?

LB: During my six months Trieste visit in 1970 my wife could visit me and with our Dacia 1100 car we travelled through Italy (Venice, Rome, Florence, Napoli, Capri, Amalfi, ...). On our way I gave also a seminar talk at FRASCATI and from the fees I bought new tires. I accompanied her by the return until Vienna and made a nice guided tour of the city. It was wonderful, but we never thought at all about remaining in the West. We believed, from now on everything would turn to be better.

In France where I stayed a whole year from March 1973 my wife got the permission to visit me for two months. I waited her in Switzerland at my old friend Victor Ionescu and we travelled through the beautiful landscapes of the Alps with my small, used Citroen “deux chevaux”. Later we travelled around in France (Paris, Vallée du Loire, Nizza, Monte Carlo, Avignon, Arles, …). It was wonderful in France, but in Romania the things were not any more looking pink. My wife had the first thoughts not to return. I was less pessimistic and still was for returning. Anyway, it was not a well-defined option. Our 10-year-old son and our parents would not understand such a step and I had no immediate job options.

After returning to Romania the changes I felt were so disastrous, that I was depressed for months. I succeeded however to change the direction of research and to adapt myself to the new conditions.

As I already mentioned, I was in the graces of Ursu and in August 1983 I was to travel to Trieste as lecturer at a summer school and in the same time to finalize the organization of the international conference on “hopping” I initiated with the support of the ICTP. I felt myself too sure in the “saddle” and convinced Ursu to let my coworkers (Gartner and Aldea) to accompany me. They already went to Trieste, but I still did not get a pass. I went to Ursu to complain about and he reassured me to solve the problem by phoning to the Securitate. I should call him the next day. Next day his secretary told me that Ursu was not in. It went on like this for days. My wife suggested me to clarify the situation and check the “absence” of Ursu. I placed in the early morning my Dacia 1300 at a corner with a nearby public phone and a good visibility towards the CNST entrance. I saw Ursu climbing the steps and ten minutes later I called his secretary. Although she covered the phone micro with her hands, I could overhear as she was asking, “What shall I say to this guy?”. Things got clear for us, I fell in disgrace due to something the “securitate” told Ursu. (Here I have to mention, that during the same summer I was summoned twice by the “securitate” to cooperate with them and I flatly refused it.)

You might understand how frightened and depressed we became. But things did not end that way. Since I applied for a private pass for this scientific visit, I applied for an audience at the passport office. The audience came fortunately at a time, when Ursu left Romania with an official delegation to South America. I brought with me all the correspondence regarding the future conference and bluffed saying: If they do not deliver my pass within two days, they must share the responsibility for the failure of this international event. Seemingly nobody wanted that, Ursu was not there to be asked and the next day I got a phone call to get my pass. The day Ursu landed at the Bucharest airport I was already in the train toward Budapest. Just before (under the blanket) my wife told me. “If you return I shall divorce you!”. She was ready to wait despite all the risks but did not want to remain any further in Romania.

I told my mother about our decision only on the way to the railway station and our son only as the train started. From Budapest, being at my uncle, I phoned home, that I am already outside.

A poignant detail expected me in Trieste. As Salam saw me, he got confused saying: “I just met Ursu a few days ago and he excused himself of not being able to support you anymore and nevertheless you are here!”

VB: You felt no remorse to leave your old mothers alone?

LB: Even, too much! However, my mother and later the mother of my wife were in good hands. From the West we were able to help them more efficiently with money, parcels of food, medicine, as well as all kind of gifts for those who took care of them. Both visited us in Germany and we tried to show to them form the western world, as much as they could enjoy. After the borders opened for us we went twice a year to visit them. My wife and me, we loved enormously our parents and always keep them in our heart and mind. Nevertheless, we think, it would have not been right to sacrifice our future and our mothers also encouraged us in our endeavor.

VB: Tell me about your first steps as a physicist in the West? How did you succeeded to get into the first lines of solid-state theory?

LB: After my arrival in Trieste, I called my old friend Vladimir Rittenberg, who was a professor at the Bonn University and told him, that I decided not to return to Romania. He sent me immediately an invitation for three weeks to give a series of lectures at his university. On this basis I got immediately a visa for West Germany. In the meantime, at the ICTP in the frame of the summer school Prof. Franz Wegener from the Heidelberg university gave a lecture about disorder and he wrote my name on the blackboard quoting my Paris paper. After that it I met him, introduced myself and told him, that I would be glad if he could offer me some months at Heidelberg to put my feet in Germany. He promised and indeed he offered me later a 6 months’ work contract in his theory group.

In Bonn I stayed more than three weeks. My friend succeeded to get some financial support to prolongate my stay until the Heidelberg invitation could start. He was extremely generous with me and I lived in his apartment for almost all this time. I have to mention here, that in the 14 years of separation our ways in Physics splat far away and he could not offer me any perspectives, besides the human solidarity.

In the meantime, I renewed my contacts with the German physicists I already met at the ICTP. Prof. Peter Thomas invited me to give a seminar talk at the Marburg University and I was a guest in his house for a couple of days. He recommended me a young coworker of him, who went to Heidelberg as a postdoc. In Heidelberg I wrote a paper together with him and a mathematician, that was published in Phys. Rev.

I had several invitations for seminar talks and in Köln even for a colloquium. Prof. Hajdu told me, that he knew two papers of mine: the one at the Paris Conference and my review paper about Kubo’s theory in Novacu’ s Seminars. Although it was in Romanian, he somehow managed to understand it.

Still being in Heidelberg I got an invitation to lecture at a summer school in Santa Fe (Arizona) about the scaling connection between the kinetic and hydrodynamic descriptions we published with Paul Gartner. I spent a month in the USA and got also a job proposal from Prof. Scully, which I declined, since I did not understand the American system. He told me to prepare a research proposal and I had no

Idea how to do it. In Europe you should adhere to the subjects of the group and the proposal for the financial support is forwarded by the inviting Professor not by the guest. Anyway, I was not very attracted by the American way of life and rather wanted to remain in Europe. In my eastern naivete I thought the farther one goes westward, the higher is the living standard and civilization, but it was not that way.

After returning to Germany I got soon two positive answers at my job applications. First came one from Prof. Götze at the Technical University in München. We met in Trieste and he worked on “hopping” like me. The position however was limited to two years. I visited him, gave a seminar talk and even got a lodging offer of a small furnished flat in the villa of another professor. A few days after I got by post the contract to sign. Before posting it, I got a phone call from the secretary of Prof. Hartmut Haug of the Frankfurt University inviting me to meet him the next day. We went to a small restaurant and had a pleasant conversation. At the end he offered me a position that could be extended up to 5 years. His condition was, that I must turn my activity toward his new, modern research orientation (non-linear optics of semiconductors). I decidedly preferred Haug’s variant, while the new research field attracted me. On the other hand, 5 years gave me the needed stability, since as I expected to get within a year or so my family out of Romania.

During the two and half years of waiting my wife could keep her job and my son continued his architecture studies. Before leaving Romania, my wife succeeded to change our spacious apartment, where we lived together with her mother into a small one not far away of her sister. She took over the burden of caring.

The choice of Haug was a very lucky one, although I knew almost nothing about him before. He was already an important person in the optics of semiconductors. A field with growing actuality due to the new lasers of high intensity with ultra-short pulses as well as the new low dimensional semiconductor structures. His field became soon one of the most important ones in solid state theory. I had luck also sharing the office with Stephan Koch preparing his habilitation. He was a bright ambitious young physicist and later he got to be one of the most important scientists and science organizers in Germany and the USA. We soon developed a friendship and published within a year together with him and Haug a successful paper. The cooperation with him went on over the years and I was his scientific guest at the Optical Science Center in Tucson for several months, while together with my wife we lived in his splendid villa in the Sonora desert.

By Haug I could benefit also of similar advantages I had in Romania. I got enough freedom in the research and I had several young gifted students and doctorands to supervise and work with me. Haug was an extremely active, stimulating physicist, being always up to date with the latest most important and interesting theoretical and experimental problems and I was glad to join him in solving these. We were complementary to each other. He also appreciated my experience with the quantum theory of fields as well as my mathematical skill. I participated at his broad cooperation, with many theoretical and experimental groups in the whole world and got also some appreciation. In the same time, he offered me and even encouraged me to have my own different, independent fields of research with own coworkers and invited scientists. He cared to equip the group with the most up-to-date computers, so we were the first to solve many new difficult problems.

As I came to Germany the prospects to get a professorship were just zero, due to formal aspects as well as to the lack of large contacts and knowledge of science organization. For a C2 or C3 professor I was too old, while for a C4 I had to follow a longer path. In Germany before getting the professor title one has to get the title of a “Privat-Dozent”, implying the publication of a scientific monography. Therefore, only a position of a “wissenschaftlicher Mitarbeiter” (scientific coworker) was available for me. It was well payed, but its duration is limited to 5 years. Haug got me on such a job but also pushed me forward to get the Privat- Dozent title followed with that of an extraordinary Professor. Moreover, in order to keep me in the Institute he succeeded to obtain the exceptional approval of the Ministry of Education to transform my position into a permanent one. I was not keen to be a globetrotter and I remained here.

I had various invitations for seminar talks at many universities. In Cambridge I met again Mott as a bronze statue. My most successful talk was at The Humboldt University in Berlin at the invitation of Prof. Zimmermann. The first day I had a Colloquium about quantum kinetics with many experimental illustrations and the next day I gave a supplementary seminar talk about irreversibility in a solvable classical polaron model, intended for a restricted auditory of theorists. To my greatest surprise, it turned out that so many were interested in my lecture, that they filled the room.

I held many years along the solid-state theory lecture in Frankfurt, 6 months also at the university of Strasbourg and I gave a series of lectures about low dimensional semiconductors at the University of Lausanne and about quantum kinetics at the university of Marburg. I had also an invited talk at the Spring meeting of the American Physical Society.

My old tradition to cooperate with experimentalist in the IFB continued also being in Germany with experimental groups in Frankfurt, Karlsruhe, Stuttgart, München, Strasbourg, Paris and Tucson. We used often to publish joint papers with them (experimental results and the theoretical interpretation). On the other hand, sometimes I worked on pure theoretical topics as Bose condensation in real time, biexciton in a quantum dot or motion of a classical polaron in an electric field.

I spent exceptionally interesting years with Hartmut Haug and we remained good friends, meeting often at our common office at the university.

VB: As far as I know, you kept along this time also a strong link with the old coworkers in Bucharest.

LB: After the regime change in Romania I invited Paul Gartner to Frankfurt and we worked again together for two and half years and published together with Haug and some of the doctorands 12 papers in Phys. Rev. and Phys. Rev. Letters. Thereafter Paul was already so known in Germany, that he got immediately a job at the University of Bremen, where he worked until his retirement. Just before my own retirement I invited also Mircea Bundaru for 6 months and we worked out together with Haug a nice paper on Bose condensation in a finite potential well. I invited also old friends to give seminar talks in Frankfurt like Luca and Andrei Mezincescu, Alexandru Aldea and some of my Russian acquaintances.

VB: Runs scientific research in Germany different compared to the one you experienced in Romania? What are the main differences?

LB: The most important difference is that scientific research occurs mostly in the universities and not in independent scientific centers. A professor most have had a research record and his main activity is still doing research. Pedagogy is merely a secondary task. His efficiency depends on the quality and number of coworkers (graduate students, doctorands and postdocs), on the technical equipment (by theorist -modern computers), on the good contacts with other research groups, the number and quality of visiting scientists and the possibilities to attend important conferences. To fulfill all these requirements, beyond the minimum offered by the finances of the university one has to apply for founding at the DFG (German Physical Society), EU or DAAG (Foundation for international cooperation) or other sources.

Generally, in Western Europe physicists prefer to work in large groups of theorists and experimentalists from different universities. In this way one forms clubs around a certain thematic. This collective activity is encouraged by getting easily financial support for their projects. Individual projects have less chance to get approved.

Of course, in the very last years in Romania we had to have research contracts, but for the theorists it was a formal game, while for the experimentalist it was detrimental guiding them toward industrial applications. The result was the drop of the scientific quality. Here the link to the industry is a spontaneous one. For example, Koch himself, together with some experimentalists is active as businessman trying to get money from their discoveries.

VB: Tell me something about the typical way of a young physicist in the West.

LB: This is a very relevant question since it differs essentially from the one used to be in the former socialist countries. As a young physicist at the IFB I envied the western colleagues moving around at various scientific centers in the world. Actually, there is not too much to envy. Stable jobs do not exist for the young scientists. These are always limited to a few years and may be prolongated only by looking for other sources of support, in the frame of some of the mentioned projects. One is compelled to look always for the next job and this mostly implies another country. To get any job one must have good recommendations from important people. If one does not succeed within a given time to get a professorship, one is lost for science. Then one has to look for a job in the industry or elsewhere. Here one must add, that it is forbidden to get a professorship at the university where one obtained the necessary titles (PHD, Privat-Dozent, extraordinary Professor). One has to be very talented and very self-conscious to follow this path. Another aspect not to be ignored is that under the above described conditions it is very difficult to get a stable partner and to ground a family. This complicates especially the possibilities of women for a scientific career.

Along the years in Frankfurt I had some gifted doctorands, but only two of them had the courage to try an academic career and one of them failed soon. The best of my doctorands had all the chances to make it to a professorship, but he chose industry, although he was fond of science. He wanted to have a quiet life with a family and not to migrate around the world. The Ericson company offered him a very good manager position already before he got his PhD. Recently Paul read in a newspaper, that he became the vice-president of German Wings. We congratulated him, and he replied, that it was more difficult get through a university examination, than to make this step.

I would like to add a few words about the number and quality of physics students. Under the conditions of socialism physics was one of the few domains of activity more or less shielded from political pressure. Science attracted many talented young people. As I arrived in Germany I met a completely different mentality. Due to the intense (almost hysterical) anti-atom energy propaganda of the greens the public opinion about physics was not at all positive. On the other hand, the youth chose careers promising more money (lawyers, medical doctors, …). Therefore, the number of students in physics was relatively low and they were not the most intelligent ones. In the same time the demands required from the students were rather low. On the other hand, the graduate students were already included in the scientific activity and got a desk and computer to work in the university.

Laci's years in Frankfurt starting in 1984

H. Haug[1]

*Institut f. Theoret. Physik, Goethe-Univ. Frankfurt

Laci Bányai came as a refugee in 1984 from Romania via short stays in Triest, Bonn and Heidelberg to my group at the Institut f. Theoretische Physik, at the Goethe-Uni. Frankfurt. Our main subject of interest was the theory of the optical properties of semiconductors excited by intensive laser pulses. The excitations are a non-equilibrium many-body system of electrons (e), holes (h), excitons (x) and bi-excitons interacting with the phonons and with each other by Coulomb forces. The optical response was in general nonlinear. The laser pulses have been in the 60ties in the nanosecond range, became picosecond pulses in the 80ties and became as short as femtoseconds at the end of the century. Many body theory in quasi-equilibrium and more and more in non-equilibrium were the tools required to understand the optical properties of laser excited semiconductors. Simultaneously the decreasing size of the samples made quantum confinement more and more important: Quasi-two-dimensional quantum well structures, quantum wires and quantum dots reduced the translational degrees of freedom of the excited carriers more and more.

Laci's interest turned first to the understanding of e,h pairs in quantum wires and dots. As an example he showed that two e,h pairs confined in a quantum dot lay energetically always lower the two noninteracting e,h pairs. The work on quantum dots had the nice advantage that Laci got invited for a few months by Stephan Koch, a former coworker in my group, a professor at the Optical Sciences Center in Tucson, Arizona with its beautifully saguaro cactus dessert. Obviously that scenery stimulated Laci and Stephan so much that they wrote a book on Semiconductor Quantum Dots, World Scientific, Singapore 1993. With these investigations Laci also got his Habilitation at the Goethe-Uni. in Frankfurt, which in 1997 was upgraded to an Extra-ordinary Professor position.

Naturally, the first years have been tough for Laci, because it took quite a long time until his family was allowed to join him. Anni, his wife also found a position in industry as an engineer, but because her company was outside of Frankfurt, they lived for many years about 50 km north of Frankfurt. Laci was quite polyglot, particularly his knowledge of the Russian language was very helpful for the conversation with our Russian guests in my group.

As already mentioned we had to use a non-equilibrium many-body theory in order to understand the femtosecond spectroscopy of semiconductors and semiconductor nano-structures. Such a theory existed and has been developed in the Russia and in the US, independently. Particularly, the real-time formulation of Leonid Keldysh could be used directly to describe the non-equilibrium time-development on a femtosecond time scale. Starting in the early 90ties, we calculated femtosecond four-wave mixing (FWM) and pump and probe signals.

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In non-equilibrium the Green functions depend truly on two-time arguments, and furthermore one has to calculate not only the spectral properties e.g. in terms of the retarded Green function G^r but also the kinetic ones in terms of $G^<$. One of the first important results in which Laci was directly involved was the appearance of the LO-phonon echo superimposed on the decaying four-wave mixing signal. Traditionally the FWM has been used to measure the polarization decay expressed by the T_2 time. A coherent oscillation on the decaying FWM signal was really something new. Fortunately, we cooperated with one of the leading experimental groups of Martin Wegener of the Uni. Karlsruhe. They observed the LO-phonon echo shortly after our prediction! See Fig. 1. Laci's excellent mathematical talent made him very valuable in our group when it came to reliable numerical evaluations of complex systems of integro-differential equations which one runs into in quantum kinetics.

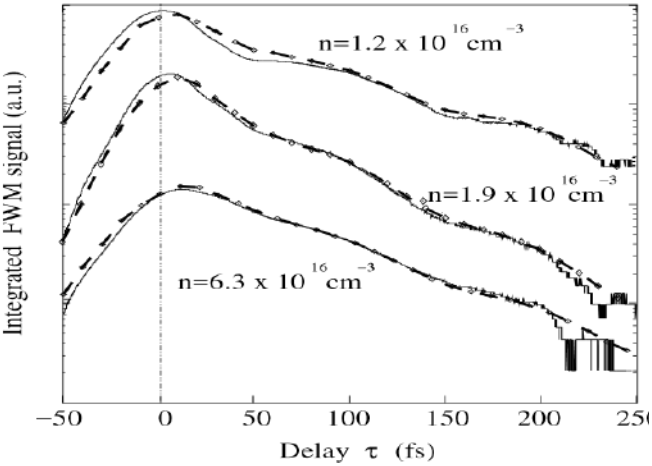

Figure 1: Measured (solid lines) and calculated (dashed lines) time-integrated four-wave mixing signals versus delay time τ showing the LO-phonon echoes for various excitation densities. (L. Banyai, D.B. Tran Thoai, E. Reitsamer, H. Haug, D. Steinbach, M.U. Wehner, M. Wegener, T. Marschner, W.Stolz, PRL 75, 2188 (1995))

Fortunately, we had Alex Ivanov as a Humboldt fellow and later as a scientific coworker in my group. A. Ivanov was a student of Keldysh, so Keldysh came with a Humboldt prize frequently to us, which obviously helped to develop our knowledge of the quantum kinetics quite a bit. And as already mentioned, Laci's ability to talk to Keldysh in his own language was also very helpful. Another approach to non-equilibrium many-body systems formulated by Karim El Sayed and Laci Bányai was the use of a Monte Carlo kinetics for a 2D electron gas starting from a given non-equilibrium distribution and following the loss of spatial correlation in time. As an initial 2D electron distribution we choose the letters QK, where each dot corresponds to one electron Very nicely Karim El Sayed and Laci could show that after a time interval in which each particle had on an average one collision the structure was lost (See Fig. 2).

We found out that if the pulse duration becomes shorter than the oscillation period say of an optical phonon or of a plasmon of an e-h gas, we are in a range where quantum kinetics is absolutely necessary in order to explain various observations of ultra-short time spectroscopy. Under high fs-excitation we studied in particular the temporal build-up of the screening of the Coulomb interaction again in cooperation with M. Wegener and another outstanding experimental group of A. Leitenstorfer and R. Huber who used ultrashort THz pulses. With these THz pulses one could study directly the build-up of the plasmon pole which were found in excellent agreement with our calculations (see Fig. 3) Similarly the temporal build-up of the polaron, i.e. the dressing of an excited carrier by LO-phonons have been studied successfully in these cooperations. Quite a few Physical Review Letters resulted from these fruitful cooperations. Most of these investigations were also supported by the collaboration with Paul Gartner, an old friend and colleague of Laci from Bucharest, where the two had been involved mainly in transport investigations. Fortunately, the DFG allowed me to finance the stay of Paul Gartner with us, which resulted in a fruitful collaboration with him for several years.

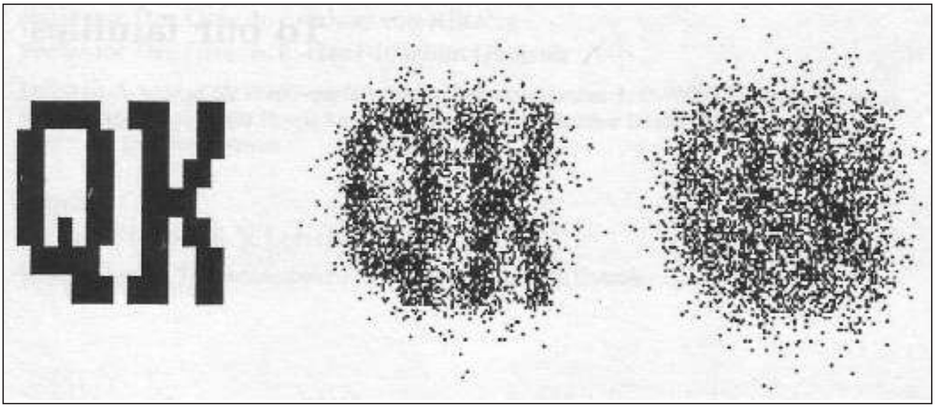

Figure 2: Monte-Carlo simulation of the Coulomb relaxation kinetics of a 2D electron gas, according to K. El Sayed and A. Banyai, taken from H. Haug and A. -P. Jauho, Quantum Kinetics for Transport and Optics of Semiconductors, Springer (1969), first ed. The initial distribution of the 3600 electrons form the initial letters of Quantum Kinetics (QK). This distribution relaxes under Coulomb scattering to a thermal distribution. The three snapshots are taken at t = 0,55 and 145 femtoseconds. In the last picture 3000 scattering events, i.e. roughly one scattering event per particle, produce a nearly thermal distribution.

In the last period before his retirement we studied the approach of a bosonic exciton gas coupled to a thermal bath towards Bose-Einstein condensation. Similarly, the kinetics of a dense interacting exciton gas towards the BEC was studied. While these studies showed that exciton condensation is in principle possible, the crucial question whether the critical density for exciton BEC is not higher than the Mott density of exciton ionization remained unanswered. Only later it turned out that the much smaller mass of an exciton polariton in a microcavity, which in turn causes an increase of the quantum-mechanical zero-point energy, makes a non-equilibrium BEC of microcavity exciton polaritons much easier compared to a BEC of the basic excitons.

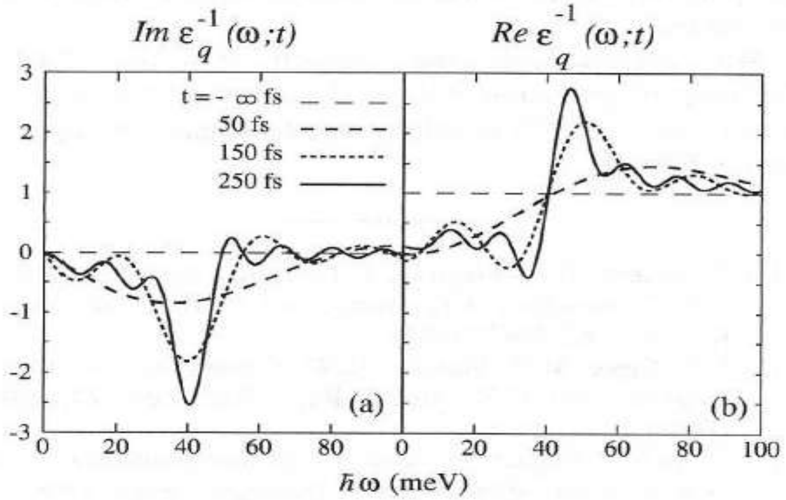

Figure 3: Spectra of the imaginary (left) and real (right) parts of the inverse dielectric function calculated for a wavenumber $qa_0 = 1$ and various times after the 15fs pulse at t = 0. The plasma frequency of the excited carriers is about 31meV the corresponding plasma period is about 100fs (L. Banyai, Q.T. Vu, B. Mieck, H. Haug, PRL 81, 882 (1998))

It remains my pleasure to thank Laci for the fruitful cooperation over many years, even in difficult times in which he struggled with an old Hepatitis C infection, which fortunately has been healed recently with a modern medical treatment. I wish him many more happy years with keen interest in many fields, such as science, computers, politics, arts and many more!

A Selfie with Laci

Paul Gartner

National Institute of Material Physics, Bucharest-Magurele

It is useful to start by declaring one's notations: Dr. Ladislaus Banyai is known to his inner and broader circle of friends and acquaintances as Laci (read approximately Lotzi), which is the Hungarian diminutive for Ladislaus.

In my life Laci's reputation preceded our actual encounter. After finishing my studies I learned that I could not get the security clearance for a position at the Institute for Atomic Physics (IFA), but I could have one at the Institute of Physics "Bucharest" (IFB). Presented like this, it sounded like a lower, less attractive option. I was hesitant, but then I was told that it cannot be that bad, since I would have Laci as a colleague. This was comforting and I took the position without second thoughts.

Indeed Laci was known as the 'enfant terrible' of the theory group at IFB. Yet the possibility to work with him had to wait for a while. In the early years of amorphous semiconductors Laci was in close contact with the experimental research group lead by Prof. Radu Grigorovici. They have reached paradoxical results: the optical gap was not matching the conductivity gap, contradicting the long-established intuition about semiconductors. But Laci went against the common wisdom and assumed the existence of localized states, optically active but not taking part in transport. These groundbreaking results were sent as a communication to the 7th Semiconductor Conference, Paris, 1964. Unfortunately, he was not allowed to attend the conference. The paper did appear in the conference proceedings ("On the theory of electric conduction in amorphous semiconductors", in "Physique des Semiconducteurs" p.417, M. Hulin Ed. Dunod, Paris,1964) and enjoyed a well-deserved popularity. Not presented by the author himself, the paper had instead the chance to be promoted by Sir Nevill Mott (Nobel prize, 1977). Today it is well-known that disorder brings localization (Anderson localization) but in the '60s this was new in the amorphous physics community. Banyai has succeeded to make Romanian physics known abroad, in spite of the semi-isolation conditions of the time.

The resulting frustration was endured discreetly: Laci abandoned for a while his interest in condensed matter in favor of quantum field theory. He was the only member of the group who was equally familiar and had contributions in both domains. When I joined the group I found him working on field theory with L. Mezincescu. (I remember that their collaboration involved a lot of friendly yelling at each other.) The topic was beyond my depth so that no collaboration was possible.

Laci returned to the condensed matter theory only in the mid '70s. The topic was again related to amorphous semiconductors. As mentioned above, the localized states do not contribute to conduction in the usual quasi-free, weakly-scattered carrier mechanism. But it turned out that they are active in another way, by phonon-assisted hopping, a new transport channel specific for disordered systems. This is when he invited me to work with him and A. Aldea on the subject. He was instrumental in establishing results concerning the hopping formalism, the role of Coulomb interaction and of the magnetic field, as well as the connection between the microscopic hopping parameters and the measurable macroscopic material constants like conductivity and magnetoresistance.

In 1983 Laci emigrated to Germany and he worked until his retirement in Prof. Hartmut Haug's group, at the Theoretical Physics Institute of the Frankfurt University. That was the time of new experiments in heterojunctions under extreme (strong, ultrashort) excitation conditions, for which new theoretical tools were required. Laci contributed decisively to the pioneering effort of the Frankfurt group in applying the methods of nonequilibrium Green's functions (Keldysh) to such novel optical and kinetic effects. His amply quoted results concern ultrafast (femtosecond) optical phenomena, which cannot be described by the usual Boltzmann equation. The methods allowed the understanding of many effects like screening, the buildup of plasmonic and polaronic modes, relaxation processes under interaction with phonons a.s.o. Several types of heterostructures, quantum wells, wires, dots, could be understood and modelled in a comprehensive framework. Also, Laci had a keen interest in a broad spectrum of problems, like the kinetics of the Bose-Einstein condensation, Coulomb correlations in semiconductors, the classical polaron theory, to name a few.

At his invitation I spent two and half years in the Frankfurt group. Professionally I could call them formative years, even though I was not a young researcher any more. I learned a lot from Laci, especially quantum kinetics in the Keldysh formalism, which was quite new for me. I had practically no previous experience with equilibrium Green's functions, let alone Keldysh ones, but Laci had a gift for clarity which stripped many so-called difficult problems of their mistique aura. First of all, he made sure to have a good contact with his audience. This meant establishing a common ground as a starting point. Then he was always checking that the contact is not lost. During his explanation he had a verbal tic "intelegi? intelegi?" (do you understand? do you understand?) while looking right into your eyes.

Also, when a problem captured his interest he was very focused and even obsessed. In Frankfurt we shared the same room, so we were within hearing distance. Once I had to tell him repeatedly that the coffee on his desk was getting cold. He was glued to his monitor, absorbed in some problem, and answered mechanically "yes" but paid no attention. I had to sent him an email from my desk to his desk, which popped up on his screen as the only mean of communication left.

I cannot overestimate his talent for finding and clearly formulating new problems. I think this is as important as finding the answers, if not more. Allways curious, he was very creative, his enthusiasms contagious and inspiring.

After all these years, remembering my musing on IFA versus IFB, I recall Robert Frost's poem:

> Two roads diverged in a wood, and I
> I took the one less traveled by,
> And that has made all the difference.

From Bucharest to Miami

Luca Mezincescu

University of Miami

I dedicate the rows below to Ladislaus Bányai on his 80th birthday. He and the late Vladimir Rittenberg where important pillars in my growth as a physicist.

At the end of the fourth year (1968) of study in the Physics Department of the University of Bucharest we were supposed to undergo a month long of so called “training “. I was specializing in Theoretical Physics and like many others was about to make a choice which will lead to important consequences for my future. Most of my colleagues were aiming for IFA (Institute of Atomic Physics), which was supposed to be the star choice. My schoolmate, friend, colleague, the beloved, recently deceased Iulian Uschersohn, and I, were thinking in a little different direction. As students we heard about the very promising Particle Theory group under the leadership of Valeriu Novacu at IFB (Institute of Physics - Bucharest) which at the time consisted of L. Bányai, P. Dita, N. Marinescu, H. Raszillier, V. Rittenberg and I.O- Stamatescu. We also learned that the duo compo- sed of Ladislaus Bányai and Vladimir Rittenberg were producing lots of interesting and successful papers. In fact, this Particle Physics group was very active: every week they held a seminar discussing the latest news from PRL, and beyond that there were also review sessions in which they were presenting talks at introductory level in order to enlarge their horizon. In the first few years of existence they also published a number of volumes in which they were contributing these original review talks. I mention only two such review articles, Bányai’s Veneziano amplitude and Rittenberg’s Effective Lagran-gians, both subjects which survived for 50 years, Veneziano amplitude being the basis of string theory, while effective Lagrangians being very important in QCD. Therefore, together with Iulian we opted to train at IFB. Beyond the above-mentioned plusses IFB was also located in the heart of Bucharest and as I was going to learn, at its ground floor was one of the best coffee shops in the Capital. As Laci was out of town, the trai-ning was directed by the late V. Rittenberg. We were supposed to go over some chap-ters of The Scattering Theory by Goldberger and Watson, and at least for me this was an extremely fruitful experience. Every day we were presenting different sections of the book and Vladimir was putting lots of questions. I was used to questions I did not know how to answer, and what I appreciated very much was, that Vladimir was formulating them in such a manner, that I got the feeling I had a hope of giving the answer in the right direction. With the training finished we decided to do our master thesis under the supervision of V. Rittenberg. In April of the following year, before we finished our thesis work, Vladimir left Romania for good, so that the completion of our work was done under the supervision of L. Bányai. This is how an interaction started which was going to become a lasting friendship for my whole life.

After the graduation I started to work at IFB under the supervision of L. Bányai and soon I was admitted for the graduate studies which were also supervised by Laci. It is from him that I understood what means to be correct and precise. I collaborated with him on a number of projects, the most long lived of which was an application of the Dirac method of quantization with constraints to the configuration space quantization of the relativistic spinor field, which in the literature was presented in a wishy-washy way. I was very lucky that for the second of my PhD exam Laci picked up the Yang Mills fields, their quantization and applications. This was around 1971 and I had the pleasure of learning the subject together with one of the examiners himself. I am not sure that I would have succeeded without his help. With some papers in the back and the exams finished together with Laci we decided that I should meet some other people. At a recent conference he met V.I. Ogievetsky with whom he established an instant contact, and he suggested that if I go to JINR-Dubna I should try to work in his group and he also appropriately recommended me. I spent a very fruitful two-year period, getting acquainted with supersymmetry and producing together with V. I. Ogievetsky one of the first reviews of supersymmetry (unfortunately my wish to extend my stay in JINR was turned down and I think this did a serious damage to my career!). As with Laci and Vladimir I formed a solid bond with the late Victor Isaakovich. I also had the pleasure of meeting Emery Sokatchev and getting in friendly relations with Zhenya Ivanov. On the return to Romania I learned with sadness that Laci was back in Solid State Physics his original passion. For me, there remained just to write a paper by myself which I did, using exhaustively the help of Mircea Bundaru. Then I wrote my PhD thesis and defended it in 1978, with Laci being one of the referees. When I left Romania at the beginning of 1982, I had the luck that Bruno Zumino knew my name and he suggested to contact (and called) in SLAC, Sydney Drell. I was also encouraged by Iulian, and by the fact that Laci knew Leonard Susskind, to ask him for help so that for a little more than a year I will be in Stanford University or SLAC with detours to UC Davis in Sacramento and University of Michigan in Ann Arbor. In Ann Arbor I met Marc Grisaru which was giving some lectures at a summer school, and I remember that we looked together at some papers related to the Harmonic Superspace, but nothing came out, however it was the beginning of some collaborations which spanned over few years. In Ann Arbor I also met Cosmas Zachos with whom latter I will collaborate. Then I was very lucky to be offered a Post Doc position by Steven Weinberg who recently left Harvard for UT Austin, Texas. This was basically the apotheosis of my career. Working in the Theory Group was an unbelievable pleasure. Steven Weinberg was giving class as usual to everything which he touched. Here I also met enormous number of very important physicists and I collaborated with D.R.T. Jones, M. Henneaux S de Alwis and last but by no means least Paul K. Townsend from University of Cambridge and through Paul with Peter van Nieuwenhuizen. While at a conference in Cambridge UK I had the pleasure of reestablishing contacts with Peter Freund and Adam Schwimmer. In the same year the much-regretted Peter Freund invited me to give a talk at University of Chicago, and this was one of the visits I enjoyed very much. Later with the help from Peter Freund I succeeded to get a tenure track appointment with University of Miami in the fall of 1986. From then on life went very fast between getting tenure, teaching, doing research, obtaining grants and waiting for the Sabbaticals. My first Sabbatical was spent at the University of Bonn as a guest of Vladimir Rittenberg during the 1995, it was an unbelievable nice sabbatical in the middle of which we spent two weeks at the Weizmann Institute in Israel, I was going to repeat this endeavor in the next two sabbaticals, thanks to Adam Schwimmer. Other very successful sabbatical leaves endeavors were spent in University of Cambridge working with Paul at different always exquisite projects. I should also mention the wonderful time I had in different occasions in JINR - Dubna as a guest of Zhenya Ivanov, always doing wonderful research. In University of Miami I collaborated with R. I. Nepomechie and T. L. Curtright.

To finish this text, I want to wish a Happy 80th Birthday to Laci and many more to come!!!

Part II. The WORK

Scientific papers

Elementary particles

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L. Bányai and S. W. Koch:

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Lectures on Non-Equilibrium Theory of Condensed Matter

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Ladislaus Alexander Bányai:

A Compendium of Solid State Theory.

Springer – International (2018)

Short introduction to my papers reproduced in this volume[1]

Out of my about 140 published scientific papers, mostly with coauthors I have chosen 28 to be reprinted in this volume. Since half of my scientific career occurred in Bucharest, while the other half in Frankfurt am Main, I selected 15 published in the Romanian period and 13 ones from my time in Germany. Some of these last ones were worked out together with my old colleagues from Bucharest, Paul Gartner and Mircea Bundaru. Obviously, I am a product of the Romanian physics with German improvements.

My scientific activity started in 1961, in Bucharest with papers dedicated to solid state theory and after a ten-year break of work on elementary particles and quantum field theory, it continued further on solid-state theory both in Bucharest, as well as in Frankfurt am Main. For sake of transparency I grouped the reprinted papers in these two categories, starting with those dedicated to the first field. Thus, the presentation is not strictly chronological.

In the IFB (The Physics Institute of the Academy in Bucharest) Prof. Valeriu Novacu organized in 1964 a research group of Elementary Particles Theory. The actual leader of the group was Vladimir Rittenberg (later professor at the University of Bonn) and its founding members were Hans Raszillier (later professor at the University of Erlangen) , Luca Mezincescu (still activ professor at the university of Miami) and Nicolae Marinescu (got his professor title at the Heidelberg University, but left physics). We published several papers of which here one finds one about the SU(3) group (all of us together) [2], one about an application of the non-linear effective Lagrangians (together with V.R.) [20] and another one on the null-plane quantization (together with L.M.) [26].

After the members of this group left Romania I continued to work on the same field together with Sorin Mărculescu from the chair of Professor Șerban Țițeica and chose for this volume two of our publications on the renormalization of gauge theories [29], [30]. This was the end of my activity on that field.

As I already mentioned, I started on solid state theory and my first publication abroad (about magneto-optical phenomena) was together with the elder theorist George Ciobanu [34]. It is followed here by my paper about the conductivity gap in amorphous semiconductors published in the Proceedings of the Paris Conference on Semiconductors in 1964 [37] and a paper published with my younger colleague Alexandru Aldea on the Hall effect in disordered semiconductors [38]. I included here also the English translation of my old (add notated) review on Kubo's theory of electric conduction [39].

Returning to solid-state after the afore mentioned break, I published together with my coworkers Alexandru Aldea and Paul Gartner several papers on hopping conduction of which I chose one written together with the late Vladek Capek of Prague on the very low temperature Seebeck effect in amorphous semiconductors

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[1] The quotation numbers refer to the "Scientific Publications" in this volume.

[41]. This period was followed by a series of papers on fundamental matters, close to mathematical physics written together with Paul Gartner on the scaling connection of the kinetic and hydrodynamic levels of which I chose two (one of them also with Vlad Protopopescu)[51], [53] . Of the papers published together with Paul Gartner I chose also a paper on the quantum mechanical derivation of the Clausius-Mosotti formula [55] , as well as one concerning the theory of the Meissner effect [57]. I included here also my last paper I wrote in Bucharest (on the Nyquist noise) together with Alexandru Aldea and Paul Gartner [58].

Of the many papers I published, while being at the Frankfurt University I chose first some related to quantum dots [78]. These resulted partly in collaboration with my friend Stephan Koch (first in Frankfurt, then in Tucson and Marburg) [66] and Pierre Gilliot from the University of Strasbourg [86].

I chose here also a paper of mine published on a classical solvable polaron model, where the dissipative motion of the electron results from the interaction with the phonon bath without any assumptions or approximations [93].

Another paper here was written together with our talented doctorand Karim El Sayed on the time reversal in many-body theories [94].

Then follow more papers of Hartmut Haug's group on the analysis of ultra-short laser spectroscopy of semiconductors in close cooperation with experimental groups in Karlsruhe [99], Paris (Anthony) [116] and München [129] within the theoretical approach of quantum kinetics. Another subject of the same period was on the THz emission of laser excited semiconductors [126], already joined by Paul Gartner, who spent two and a half years in Frankfurt. A very interesting topic of Haug's group was the real time description of the Bose-Einstein condensation [127]. Here our best doctorand Oliver Schmitt played an important role. Together with Paul Gartner we published also a paper [132] on a solvable model of Bose-condensation in real time. Then I quote a short paper of mine about the c-number approximation for the Bose condensate [134]. The papers from the Frankfurt period end with a paper together with Hartmut Haug and Mircea Bundaru on Bose-Einstein condensation in a finite potential well [136].

As one might have seen, the manifold of my papers had to do with the manifold of talented coworkers, most of them becoming also close friends of mine.

Reprints

Elementary particles

Commun. math. Phys. 2, 121—132 (1966)

Irreducible Tensors for the Group SU_3

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Abstract. The explicit determination of the matrix elements of the SU_3 irreducible tensors is carried out by a purely algebraic method. These expressions may be used to compute the Clebsch-Gordan coefficients by orthogonalisation. For the special case of $(0, q)$ tensors simple formulas are derived.

I. Introduction

Recently compact Lie groups of rank $\geqq 2$ have found wide applications in elementary particle physics. In view of concrete physical problems, for each group the following main problems have to be solved: (a) determination of the irreducible representations (I.R.) and the matrix elements of the group generators, (b) decomposition of the direct product of two I.R. and hence the computation of the Clebsch-Gordan (C.G.) coefficients. It is well known that the groups of rank $\geqq 2$ are not multiplicity-free (the same representation may occur in the direct product more than once) so that the C.G. coefficients are not completely specified by the basis vectors. The Wigner-Eckart theorem is also modified: the number of reduced matrix elements appearing there is equal to the multiplicity of the equivalent representations.

The simplest of the above groups is SU_3. In this case the problem (a) has already been solved by a number of authors [1, 2, 3, 4, 5], while problem (b) has received until now only an incomplete solution. MOSHINSKY [6] has derived a compact expression for the C.G. coefficients corresponding to the product $(p, q) \otimes (p', 0)$, which is multiplicity-free, while KURIAN, LUBIÉ and MACFARLANE [7] have tabulated the coefficients for the simple product $(p, q) \otimes (1, 1)$, BAIRD and BIEDENHARN [8] for the cases $(p, q) \otimes (1, 0)$, $(p, q) \otimes (0, 1)$, $(p, q) \otimes (1, 1)$ and PANDIT and MUKUNDA [9] for the case $(p, q) \otimes (3, 0)$. We must also mention the numerical tables of SU_3 C.G. coefficients [10, 11, 12, 13] for the products of lowest representations. However, the general problem of deriving a simple analytical formula analogous to the Wigner-Racah expression for SU_2 has not yet been solved and it is doubtful if such a task is really possible.

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* In partial fulfillment for the requierements of the doctoral degree at the Institute of Physics of the Academy.

In the present paper we establish an analytic expression for the matrix elements of an arbitrary irreducible tensor (I.T.). A method used by LURIÉ and MACFARLANE [15] for the (1, 1) tensor is generalised. The method consists in solving the commutation relations (C.R.) which define the I.T. We obtain equations with finite differences whose solutions contain the number of arbitrary constants corresponding to the equivalent representations which occur in the direct product. The constants are connected with the reduced matrix elements of the tensors. The C.G. coefficients are obtained by orthogonalisation.

It seems that the method used in this paper for SU_3 may be extended to other higher rank groups.

In the Sections II—V we establish the expression of the matrix elements of the I.T. in the general case while in Sec. VI, the formula for the $(0, q)$ irreducible tensors, which are multiplicity-free, is derived. These last expressions are obtained in a much easier way and are simpler than those of MOSHINSKY [6]. A brief version of the present paper has been published elsewhere [14].

II. Preliminary remarks

The irreducible tensors T^μ_ν corresponding to a representation $\mu = (p, q)$ of the SU_3 group and labelled by $\nu = (I, I_z, Y)$ are defined by their C.R. with the infinitesimal operators X:

$$[X, T^\mu_\nu] = (\mu, \nu' \,|X|\, \mu, \nu)\, T^\mu_{\nu'} \,. \tag{1}$$

The general structure of the eigenvalue diagram and hence the range of I, I_z and Y may be deduced from the paper of GINIBRE [16] and is represented in Fig. 1.

The matrix elements of the eight infinitesimal operators may be found in DE SWART's paper [17]. We shall mention only those which are used in the present paper,

$$\begin{aligned}&(\mu; I', I_z + 1/2, Y + 1 \,|K_+|\, \mu; I, I_z, Y)\\ &\qquad = C^{I,1/2,I'}_{I_z,1/2,I'_z}\left[\delta_{I',I+1/2}\frac{A_\mu(x)}{(2I+2)^{1/2}} - \delta_{I',I-1/2}\frac{B_\mu(y)}{(2I)^{1/2}}\right]\end{aligned} \tag{2a}$$

$$\begin{aligned}&(\mu; I', I_z + 1/2, Y - 1 \,|L_-|\, \mu; I, I_z, Y)\\ &\qquad = C^{I,1/2,I'}_{I_z,1/2,I'}\left[\delta_{I',I+1/2}\frac{B_\mu(y+1)}{(2I+2)^{1/2}} + \delta_{I',I-1/2}\frac{A_\mu(x-1)}{(2I)^{1/2}}\right]\end{aligned} \tag{2b}$$

where $C^{I',I'',I}_{I'_z,I''_z,I_z}$ are the C.G. coefficients of the SU_2 group and

$$A_\mu(x) = [(a - x)\,(b + x + 2)\,(c + x + 1)]^{1/2} \tag{3}$$

$$B_\mu(y) = [(a + y + 1)\,(b - y + 1)\,(-c + y)]^{1/2} \tag{4}$$

where

$$a = \frac{1}{3}(2p + q); \quad b = \frac{1}{3}(p + 2q); \quad c = \frac{1}{3}(p - q) \tag{5}$$

$$x = I + \frac{1}{2}Y; \quad y = I - \frac{1}{2}Y. \tag{6}$$

From Fig. 1 follows

$$-c \leqq x \leqq a; \quad c \leqq y \leqq b. \tag{7}$$

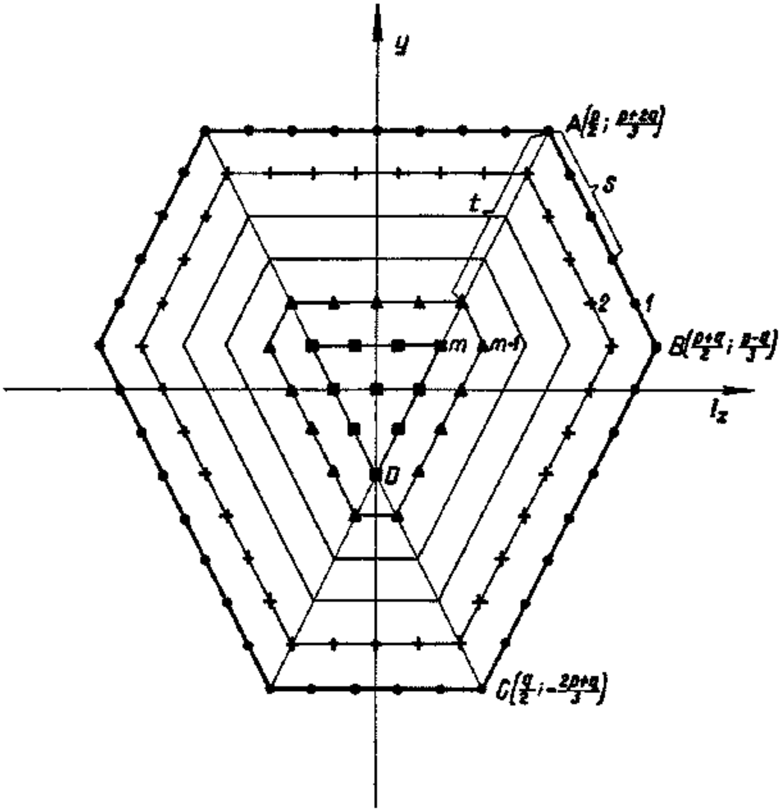

Fig. 1. Eigenvalue diagram for an irreducible representation (p, q). The numbers denote the eigenvalue multiplicity. The maximum multiplicity is $m = 1 + \min(p, q)$

The matrix elements (2) being expressed in the variables x, y instead of I, Y, we shall adopt the former to label the matrix elements of the I.T. Using the Wigner-Eckart theorem for SU_2 we find [17]:

$$\begin{aligned}&(\mu_3; I_3, I_{z_3}, Y_3 \,|T^{\mu_2}_{I_2, I_{z_2}, Y_2}|\, \mu_1, I_1, I_{z_1}, Y_1)\\ &= \delta_{Y_3, Y_1 + Y_2}\, C^{I_1, I_2, I_3}_{I_{z_1}, I_{z_2}, I_{z_3}}\, (\mu_1, x_1, y_1 \,|\mu_2, x_2, y_2|\, \mu_3, r),\end{aligned} \tag{8}$$

where we have used the well known triangle rule:

$$I_3 = I_1 + I_2 - r \quad (r = 0, 1, \ldots, 2\min(I_1, I_2)) \tag{9}$$

and the additivity of the hypercharge. The rather asymmetric notation of the new matrix element is more fashionable; we shall return to a symmetric notation in the final result.

We must also have in view the selection rules for μ which are given by Speiser's [17] graphical multiplication rules or by the rather intricate expressions in [18, 19, 20, 21, 22] and [9].

We shall mention only the following relations:

$$a_1 + a_2 - a_3 = \tau; \quad b_1 + b_2 - b_3 = \sigma \tag{10}$$

where τ and σ are nonnegative integers while the multiplicity of the equivalent representations is given by the expression:

$$M = N + 1 - n \tag{11}$$

where

$$N = b_2 - c_2 + \sum_{i=1}^{3} u_i \theta(-u_i) \tag{12}$$

with

$$u_1 = a_3 - a_1 + c_2; \quad u_2 = b_1 - b_3 + c_2; \quad u_3 = c_1 - c_3 + c_2 \tag{13}$$

and n is a nonnegative integer which vanishes for $p_1 \geqq p_2$, $q_1 \geqq q_2$ and whose concrete expression can be deduced from the above mentioned papers.

A careful examination of the commutation relations (1) suggests the following way of solving the system of equations which determine the matrix elements of the I.T.:

a) Find the matrix elements with $r = 0$ of the tensor $T^{\mu_2}_{\frac{1}{2}(p_2+q_2), \frac{1}{2}(p_2+q_2), c_2}$ (B in Fig. 1).

b) Determine by recurrence the matrix elements with $r = 0$ of the tensor $T^{\mu_2}_{\frac{p_2}{2}, \frac{p_2}{2}, b_2}$ (running from B to A).

c) Solve the finite difference equations which determine all the matrix elements of the tensor $T^{\mu_2}_{\frac{p_2}{2}, \frac{p_2}{2}, b_2}$ (A in Fig. 1).

d) Obtain by recurrence the tensors $T^{\mu_2}_{\frac{1}{2}(p_2+s), \frac{1}{2}(p_2+s), b_2-s}$ (running from A to B).

e) Determine by recurrence the matrix element of the tensors $T^{\mu_2}_{\frac{1}{2}(p_2+s-t), \frac{1}{2}(p_2+s-t), b_2-s-t}$ (running parallel to BC).

III. The matrix elements of the tensor $T^{\mu_2}_{\frac{p_2}{2}, \frac{p_2}{2}, b_2}$

Let us consider the matrix elements $\left(\mu_3, I_1 + \frac{1}{2}(p_2 + q_2 + 1), I_{z_1} + \frac{1}{2}(p_2 + q_2 + 1), Y_1 + c_2 \pm 1 | \ldots | \mu_1, I_1, I_{z_1}, Y_1\right)$ of the C.R.:

$$\left[K_+, T^{\mu_3}_{\frac{1}{2}(p_2+q_2), \frac{1}{2}(p_2+q_2), c_2}\right] = 0 \tag{14a}$$

$$\left[L_-, T^{\mu_3}_{\frac{1}{2}(p_2+q_2), \frac{1}{2}(p_2+q_2), c_2}\right] = 0 \tag{14b}$$

and the change of function

$$(\mu_1, x_1, y_1 \,|\mu_2, a_2, b_2|\, \mu_3, 0) =$$
$$\left[\frac{(x_1+y_1+1)!\, f(a_3, b_3, c_3; x_1+a_2)\, f(b_3, a_3, -c_3; y_1+b_2)}{(x_1+y_1+a_2+b_2+1)!\, f(a_1, b_1, c_1; x_1)\, f(b_1, a_1, -c_1; y_1)}\right]^{1/2} F(\mu, x_1, y_1) \tag{15}$$

where

$$f(a, b, c; x) = \frac{(b+x+1)!\,(c+x)!}{(a-x)!} \tag{16}$$

we obtain the system of finite difference equations

$$F(\mu, x_1+1, y_1) = F(\mu, x_1, y_1) \tag{17a}$$

$$F(\mu, x_1, y_1+1) = F(\mu, x_1, y_1) \tag{17b}$$

with the obvious solution

$$F(\mu, x_1, y_1) = F(\mu)\,. \tag{18}$$

In the last expressions μ stands for μ_1, μ_2, μ_3. Consider now the matrix elements $\left(\mu_3, I_1 + \frac{1}{2}(p_2+s+1), I_{z_1} + \frac{1}{2}(p_2+s+1),\ Y_1 + b_2 - s \pm \pm 1 \,|\, \ldots \,|\, \mu_1, I_1, I_{z_1}, Y_1\right)$ of the C.R.:

$$\left[K_+, T^{\mu_3}_{\frac{1}{2}(p_2+s), \frac{1}{2}(p_2+s), b_2-s}\right] = 0 \quad (0 \leqq s \leqq q_2) \tag{19a}$$

$$\left[L_-, T^{\mu_3}_{\frac{1}{2}(p_2+s), \frac{1}{2}(p_2+s), b_2-s}\right] = -$$
$$-\,[(s+1)(q_2-s)]^{1/2}\, T^{\mu_3}_{\frac{1}{2}(p_2+s+1), \frac{1}{2}(p_2+s+1), b_2-s-1} \tag{19b}$$

we make the change of function

$$(\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2+s|\, \mu_3, 0)$$
$$= \left[\frac{(b_2-c_2-s)!\,(x_1+y_1+1)!\, f(a_3, b_3, c_3; x_1+a_2)\, f(b_3, a_3, -c_3; y_1+c_2+1)}{s!\,(x_1+y_1+a_2+c_2+s+1)!\, f(a_1, b_1, c_1; x_1)\, f(b_1, a_1, -c_1; y_1)}\right]^{1/2} \times$$
$$\times\, G_s(\mu, x_1, y_1) \tag{20}$$

and find the system of finite difference equations

$$G_s(\mu, x_1+1, y_1) = G_s(\mu, x_1, y_1) \tag{21a}$$

$$G_s(\mu, x_1, y_1+1) - G_s(\mu, x_1, y_1) = -G_{s+1}(\mu, x_1, y_1)\,. \tag{21b}$$

Since from (15), (18) and (20) it results that the function $G_{b_2-c_2}(\mu, x_1, y_1)$ is independent of x_1 and y_1, for $s = 0$ we get:

$$G_0(\mu, x_1, y_1) = \sum_{\gamma=0}^{b_2-c_2} T'_\gamma(\mu)\, y_1^\gamma \tag{22}$$

where $T'_\gamma(\mu)$ are undetermined constants.

Let us take now the matrix elements $\left(\mu_3, I_1 + \frac{1}{2}(p_2+1) - r, I_{z_1} + \frac{1}{2}(p_2+1), Y_1 + b_2 + 1 \,|\, \ldots \,|\, \mu_1, I_1, I_{z_1}, Y_1\right)$ of the C.R.:

$$\left[K_+, T^{\mu_2}_{\frac{p_2}{2}, \frac{p_2}{2}, b_2}\right] = 0 \tag{23}$$

and make the change of function:

$$\begin{aligned}(\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2|\, \mu_3, r) &\\ = \left[\frac{(b_2-c_2)!\,(x_1+y_1+1)\,(x_1+y_1+a_2+c_2-r+1)!}{((x_1+y_1+a_2+c_2-2r+1)!)^2\,(x_1+y_1-r)!}\right]^{1/2} &\times \\ \times \left[\frac{f(a_3, b_3, c_3; x_1+a_2-r)\, f(b_1, a_1, -c_1; y_1)}{f(a_1, b_1, c_1; x_1)\, f(b_3, a_3, -c_3; y_1+c_2-r)}\right]^{1/2} f_r(\mu, x_1, y_1)&\end{aligned} \tag{24}$$

we obtain then the equations

$$\begin{aligned} r^{-1/2}(a_2+c_2+1-r)^{1/2}\,[f_r(\mu, x_1+1, y_1) - f_r(\mu, x_1, y_1)] &\\ = \frac{f_{r-1}(\mu, x_1, y_1-1)}{(x_1+y_1+1)\,(x_1+y_1)} - &\\ - \frac{f_{r-1}(\mu, x_1, y_1)}{(x_1+y_1+a_2+c_2-2r+3)\,(x_1+y_1+a_2+c_2-2r+2)}&. \end{aligned} \tag{25}$$

Equations (20) and (22) give

$$f_0(\mu, x_1, y_1) = f_0(\mu, y_1) = \frac{f(b_3, a_3, -c_3; y_1+c_2)}{f(b_1, a_1, -c_1; y_1)} \sum_{\gamma=0}^{b_2-c_2} T'_\gamma(\mu)\, y_1^\gamma\,. \tag{26}$$

One observes that for $r = a_2 + c_2 + 1$ the left side of (25) vanishes so that we obtain an identity which is easily verified. Consider the function

$$f_{a_2+c_2}(\mu, x_1, y_1) = \frac{(x_1+y_1-a_2-c_2+1)!\,(x_1+y_1-a_2-c_2)!}{(x_1+y_1+1)!\,(x_1+y_1)!}\, H(x_1, y_1)\,. \tag{27}$$

From (25) we then obtain for $r = a_2 + c_2 + 1$:

$$H(x_1, y_1+1) = H(x_1, y_1) = H(x_1)\,. \tag{28}$$

The general solution of the system of equations (25) is

$$\begin{aligned} f_r(x_1, y_1) = \sum_{l=0}^{r} \sum_{k=0}^{r-l} (-1)^k\, [(a_2+c_2-r+l)!\,(r-l)!]^{1/2} &\times \\ \times \frac{(x_1+y_1-k)!\,(x_1+y_1+a_2+c_2-2r+2l+1)!}{k!\,(r-k-l)!\,(x_1+y_1+a_2+c_2+l+1-r-k)!}\, \frac{1}{(x_1+y_1)!}\, \varphi_l(y_1-k)& \end{aligned}$$

where $\varphi_0 = f_0(\mu, y_1)$ and $\varphi_l(y_1)$ $(l = 1, 2, \ldots, r)$ are arbitrary functions. One can see by direct calculation that for the supplementary condition (28) we have $\varphi_l(y_1) \equiv 0$ if $l \neq 0$ and $f_0(\mu, y_1)$ must be a polinomial of degree p_2 in y_1. From the Speiser-Goldberg selection rules we then obtain some relations between the constants $T'_\gamma(\mu)$ so that instead of (26) we have

$$f_0(\mu, y_1) = \frac{f(b'_1, a'_1, -c'_1; y_1)}{f(b_1, a_1, -c_1, y_1)} \sum_{\gamma=0}^{N} T_\gamma(\mu)\, y_1^\gamma \tag{29}$$

where

$$a'_1 = a_1 + u_1\theta(u_1); \quad b'_1 = b_1 - u_2\theta(u_2); \quad c'_1 = c_1 - u_3\theta(u_3); \tag{30}$$

$T_\gamma(\mu)$ are arbitrary constants and N is given by (12).

Summarising the results, we have

$$(\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2|\, \mu_3, r) = \sum_{\gamma=0}^{N} T_\gamma(\mu)\, (\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2|\, \mu_3, r)_\gamma \tag{31}$$

where:

$$\begin{aligned}(\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2|\, \mu_3, r)_\gamma \\ = \Bigg[(a_2 + c_2 - r)!\, r!\, (x_1 + y_1 + 1) \frac{(x_1 + y_1 + a_2 + c_2 + 1 - r)!}{(x_1 + y_1 - r)!} \times \\ \times \frac{f(a_3, b_3, c_3; x_1 + a_2 - r)\, f(b_1, a_1, -c_1, y_1)}{f(a_1, b_1, c_1; x_1)\, f(b_3, a_3, -c_3; y_1 + c_2 - r)}\Bigg]^{1/2} \times \\ \times \sum_{k=0}^{r} (-1)^k \frac{(x_1 + y_1 - k)!\, (y_1 - k)^\gamma}{k!\, (r-k)!\, (x_1 + y_1 + a_2 + c_2 + 1 - r - k)!} \times \\ \times \frac{f(b'_1, a'_1, -c'_1; y_1 - k)}{f(b_1, a_1, -c_1; y_1 - k)}\,.\end{aligned} \tag{32}$$

IV. The matrix elements for an arbitrary I. T.

Let us consider the matrix elements $\left(\mu_3, I_1 + \frac{1}{2}(p_2 + s + 1) - r,\; I_1 + \frac{1}{2}(p_2 + s + 1), Y_1 + b_2 - s - 1 \,|\, \ldots \,|\, \mu_1, I_1, I_{z_1}, Y_1\right)$ of the C.R (19b) and make the change of functions:

$$\begin{aligned}(\mu_1, x_1, y_1 \,|\mu_2, a_2, c_2 + s|\, \mu_3, r)_\gamma \\ = \Bigg[r!\, (a_2 + c_2 + s - r)!\, (a_2 + c_2 + s + 1)\, (x_1 + y_1 + 1) \times \\ \times \frac{(x_1 + y_1 + a_2 + c_2 + s + 1 - r)!}{(x_1 + y_1 - r)!} \frac{f(a_1, b_1, c_1; x_1)}{f(a_3, b_3, c_3; x_1 + a_2 - r)} \times \\ \times \frac{f(b_3, a_3, -c_3; y_1 + c_2 + s - r)}{f(b_2, a_2, -c_2; c_2 + s)\, f(b_1, a_1, -c_1; y_1)}\Bigg]^{1/2} g^\gamma_{s,r}(\mu, x_1, y_1)\,.\end{aligned} \tag{33}$$

We obtain the recurrence relation

$$
\begin{aligned}
&g^{\gamma}_{s+1,r}(\mu, x_1, y_1) \\
&= \frac{g^{\gamma}_{s,r}(\mu, x_1, y_1) + g^{\gamma}_{s,r-1}(\mu, x_1, y_1)}{x_1 + y_1 + a_2 + c_2 + s + 2 - 2r} - \frac{g^{\gamma}_{s,r}(\mu, x_1, y_1 + 1) + g^{\gamma}_{s,r-1}(\mu, x_1 - 1, y_1)}{x_1 + y_1 + 1}
\end{aligned}
\tag{34}
$$

in which $g^{\gamma}_{0,r}(\mu, x_1, y_1)$ is known from (32) and (33). The solution of the recurrence relation is:

$$
\begin{aligned}
&g^{\gamma}_{s,r}(\mu, x_1, y_1) \\
&= \sum_{m_{i,\alpha} \geqq 0} (-1)^{m_{0,2} + m_{0,4}} \delta_{\varepsilon_0, s}\, \theta(m_{0,2} + m_{0,4} + a_2 + c_2 - r) \prod_{i=1}^{s} \delta_{\varepsilon_i, s-i} \times \\
&\times (\delta_{m_{i-1,\alpha} - m_{i,\alpha}, 0} + \delta_{m_{i-1,\alpha}, 1}) \times \\
&\times \frac{(x_1 + y_1 + s + 1 - 2r + a_2 + c_2 + m_{i,2} - m_{i,1})^{m_{i,1} - m_{i-1,1} + m_{i,2} - m_{i-1,2}}}{(x_1 + y_1 + 1 + m_{i,3} - m_{i,4})^{m_{i-1,3} - m_{i,3} + m_{i-1,4} - m_{i,4}}} \times \\
&\times g^{\gamma}_{0, r - m_{0,2} - m_{0,4}}(\mu, x_1 - m_{0,4}, y_1 + m_{0,3}) \\
&\qquad (\alpha = 1, 2, 3, 4; \quad i = 0, 1, \ldots, s); \quad \varepsilon_i = \sum_{\alpha=1}^{4} m_{i,\alpha}\,.
\end{aligned}
\tag{35}
$$

Choosing $I_{z_1} = I_1 - r$ and taking the matrix elements

$$
\Bigl(\mu_3, I_1 + \frac{1}{2}(p_2 + s - t - 1) - r, I_1 +
+ \frac{1}{2}(p_2 + s - t - 1) - r, Y_1 + b_2 - s - t - 1 \,|\, \cdots \,|\, \mu_1, I_1\, I_1 - r, Y_1\Bigr)
$$

of the C.R.

$$
\begin{aligned}
&\Bigl[K_-, T^{\mu_2}_{\frac{1}{2}(p_2 + s - t), \frac{1}{2}(p_2 + s - t), b_2 - s - t}\Bigr] \\
&\qquad = \left[\frac{(s + 1)(q_2 - s)(p_2 + s + 2)}{(p_2 + s - t + 1)(p_2 + s - t + 2)}\right]^{1/2} \times \\
&\qquad \times T^{\mu_2}_{\frac{1}{2}(p_2 + s - t + 1), \frac{1}{2}(p_2 + s - t - 1), b_2 - s - t - 1} + \\
&\qquad + \left[\frac{(p_2 - t)(p_2 + q_2 - t + 1)(t + 1)}{p_2 + s - t + 1}\right]^{1/2} \times \\
&\qquad \times T^{\mu_2}_{\frac{1}{2}(p_2 + s - t - 1), \frac{1}{2}(p_2 + s - t - 1), b_2 - s - t - 1},
\end{aligned}
\tag{36}
$$

we make the change of function

$$
\begin{aligned}
&(\mu_1, x_1, y_1 \,|\, \mu_2, a_2 - t, c_2 + s \,|\, \mu_3, r)_{\gamma} \\
&\qquad = [(r!)^{-1}(a_2 + c_2 + s - t + 1)(x_1 + y_1 + 1)(x_1 + y_1 - r)!]^{1/2} \times \\
&\qquad \times \Bigl[(x_1 + y_1 + a_2 + c_2 + s - t - r + 1)!\,(a_2 + c_2 + s - t - r)! \times \\
&\qquad \times \frac{f(a_2, b_2, c_2; a_2 - t)\, f(a_1, b_1, c_1; x_1)}{f(b_2, a_2, -c_2; c_2 + s)\, f(a_3, b_3, c_3; x_1 + a_2 - r - t)} \times \\
&\qquad \times \frac{f(b_3, a_3, -c_3; y_1 + c_2 + s - r)}{f(b_1, a_1, -c_1; y_1)}\Bigr]^{1/2} h^{\gamma}_{t,s,r}(\mu, x_1, y_1)
\end{aligned}
\tag{37}
$$

and find the recurrence relation

$$\begin{aligned} h^{\gamma}_{t,s,r}(\mu, x_1, y_1) = {} & -h^{\gamma}_{t-1,s+1,r+1}(\mu, x_1, y_1) + (a_2 + c_2 + s - t + 2) \times \\ & \times [h^{\gamma}_{t-1,s,r}(\mu, x_1, y_1) + (x_1 + y_1 + 1)^{-1} \times \\ & \times (h^{\gamma}_{t-1,s,r+1}(\mu, x_1, y_1 + 1) - h^{\gamma}_{t-1,s,r}(\mu, x_1 - 1, y_1))] , \end{aligned} \tag{38}$$

where $h^{\gamma}_{0,s,r}(\mu, x_1, y_1)$ is known from (33)–(35) and (38).

The solution of the recurrence equation is:

$$\begin{aligned} h^{\gamma}_{t,s,r}(\mu, x_1, y_1) = {} & \sum_{n_{j,\beta} \geqq 0} (-1)^{n_{0,1}+n_{0,4}} \delta_{\omega_0,t}\, \theta(x_1 + y_1 - r - n_{0,1} - n_{0,4}) \times \\ & \times \theta(b_2 - c_2 - s - n_{0,1}) \prod_{j=1}^{t} \delta_{\omega_j, t-j} (\delta_{n_{j-1,\beta} - n_{j,\beta}, 0} + \delta_{n_{j-1,\beta} - n_{j,\beta}, 1}) \times \\ & \times \frac{(a_2 + c_2 + s + n_{j,1} - j + 2)^{1 + n_{j,1} - n_{j-1,1}}}{(x_1 + y_1 + 1 - n_{j,4} + n_{j,3})^{n_{j-1,3} + n_{j-1,4} - n_{j,3} - n_{j,4}}} \times \\ & \times h^{\gamma}_{0,s+n_{0,1},r+n_{0,1}+n_{0,3}}(\mu, x_1 - n_{0,4}, y_1 + n_{0,3}) \end{aligned} \tag{39}$$

$$(\beta = 1, 2, 3, 4;\ j = 0, 1, \ldots, t),\ \omega_j = \sum_{\beta=1}^{4} n_{j,\beta}$$

and so all the matrix elements are known.

V. Final result

Noting that $x_1 + x_2 - x_3 = y_1 + y_2 - y_3 = r$, we shall write the final result in a more symmetric manner:

$$\begin{aligned} & (\mu_3, I_3, I_{z,3}, Y_3 \,|T^{\mu_2}_{I_2, I_{z_2}, Y_2}|\, \mu_1; I_1, I_{z_1}, Y_1) \\ & \qquad = C^{I_1, I_2, I_3}_{I_{z_1}, I_{z_2}, I_{z_3}} (\mu_1, x_1, y_1 \,|\mu_2, x_2, y_2|\, \mu_3, x_3, y_3) . \end{aligned} \tag{40}$$

The expression derived for the I_z-independent matrix element is

$$\begin{aligned} & (\mu_1, x_1, y_1 \,|\mu_2, x_2, y_2|\, \mu_3, x_3, y_3) \\ & \qquad = \sum_{\gamma=0}^{N} T_{\gamma}(\mu_1, \mu_2, \mu_3)\, (\mu_1, x_1, y_1 \,|\mu_2, x_2, y_2\, |\mu_3, x_3, y_3)_{\gamma} \end{aligned} \tag{41}$$

The coefficients T_{γ} are related to the reduced matrix element. The formula for the (nonorthonormalized) isoscalar coefficients is:

$$\begin{aligned} & (\mu_1, x_1, y_1 \,|\mu_2, x_2, y_2|\, \mu_3, x_3, y_3)_{\gamma} \\ & \qquad = \Big[(x_1 + y_1 + 1)\, (x_2 + y_2 + 1)\, (y_1 - x_2 + x_3)!\, (-x_1 + y_2 + x_3)! \times \\ & \times \frac{(x_1 + x_2 + y_3 + 1)!\, f(a_1, b_1, c_1; x_1)\, f(a_2, b_2, c_2; x_2)\, f(b_3, a_3, -c_3; y_3)}{(x_1 + x_2 - x_3)!\, f(a_3, b_3, c_3; x_3)\, f(b_1, a_1, -c_1; y_1)\, f(b_2, a_2, -c_2; y_2)}\Big]^{1/2} F_{\gamma}(\mu, x, y) \end{aligned} \tag{42a}$$

where

$$F_{\gamma}(\mu, x, y) = \sum_{n_{0,\alpha}} \sum_{m_{0,\beta}} (-1)^{n_{0,1} + n_{0,4} + m_{0,3} + m_{0,4}}\, \delta_{\omega_0, a_2 - x_2}\, \delta_{\varepsilon_0, y_2 - c_2 + n_{0,3}} \times$$

$$\times\theta(b_2 - y_2 - n_{0,1})\,\theta(a_2 + c_2 - x_1 - x_2 + x_3 - n_{0,1} - n_{0,3} + m_{0,2} + m_{0,4}) \times$$

$$\times\varphi(n_{0,\alpha}; \mu, x, y)\,\psi(n_{0,\alpha}, m_{0,\beta}; \mu, x, y)\,\chi_\gamma(\mu; x_1 - n_{0,4} - m_{0,4}, \tag{42b}$$

$$y_1 + n_{0,3} + m_{0,3}, x_3 + n_{0,2} + m_{0,2}, y_3 - m_{0,1})\,\frac{(x_1 + x_2 - x_3 + n_{0,1} + n_{0,3})!}{(y_1 - x_2 + x_3 - n_{0,1} - n_{0,4})!}$$

and

$$\varphi(n_{0,\alpha}; \mu, x, y) = \sum_{n_{i,\alpha}} \prod_{i=1}^{a_3 - x_3} \delta_{\omega_i, a_3 - x_3 - i}(\delta_{n_{i-1,\alpha} - n_{i,\alpha}, 0} + \delta_{n_{i-1,\alpha} - n_{i,\alpha}, 1}) \times$$

$$\times \frac{(x_2 + y_2 + 2n_{i,1} + n_{i,2} + n_{i,3} + n_{i,4} + 2)^{1 + n_{i,1} - n_{i-1,1}}}{(x_1 + y_1 + n_{i,3} - n_{i,4} + 1)^{n_{i-1,3} + n_{i-1,4} - n_{i,3} - n_{i,4}}}\,; \tag{42c}$$

$$(\alpha = 1, 2, 3, 4)$$

$$\psi(n_{0,\alpha}, m_{0,\beta}; \mu, x, y) = \sum_{m_{j,\beta}} \prod_{j=1}^{y_2 - c_2 + n_{0,1}} \delta_{\varepsilon_j, y_2 - c_2 + n_{0,1} - j}\,(\delta_{m_{j-1,\beta} - m_{j,\beta}, 0} + \tag{42d}$$

$$+ \delta_{m_{j-1,\beta} - m_{j,\beta}, 1})\,\frac{(x_3 + y_3 + n_{0,2} - m_{j,2} - m_{j,1} + 1)^{m_{j,1} + m_{j,2} - m_{j-1,1} - m_{j-1,2}}}{(x_1 + y_1 + n_{0,3} - n_{0,4} + m_{j,3} - m_{j,4} + 1)^{m_{j-1,3} + m_{j-1,4} - m_{j,3} - m_{j,4}}}\,;$$

$$(\beta = 1, 2, 3, 4)$$

$$\chi_\gamma(\mu, x_1, y_1, x_3, y_3) = \frac{f(a_3, b_3, c_3; x_3)\,f(b_1, a_1, -c_1; y_1)}{f(a_1, b_1, c_1; x_1)\,f(b_3, a_3, -c_3; y_3)} \times \tag{42e}$$

$$\times \sum_k (-1)^k \frac{(x_1 + y_1 - k)!\,(y_1 - k)^\gamma\,f(b_1', a_1', -c_1'; y_1 - k)}{k!\,(x_1 - x_3 + a_2 - k)!\,(y_1 + x_2 + c_2 - k + 1)!\,f(b_1, a_1, -c_1; y_1 - k)}\,.$$

We note that if the selection rules correspond to a multiplicity smaller than $N + 1$, the functions appearing in (40) are no longer linearly independent.

The expression given above may be used to compute the C.G. coefficients if we use the orthonormality relations [17]. The computation of the general expression for the C.G. coefficients, a very difficult task, must be done for each concrete case individually.

VI. The special I. T. corresponding to the representation (0, q)

The matrix elements of these tensors which are multiplicity-free can be obtained directly without making use of the general formula given in Sec. V. This is preferable because it is difficult to observe the simplifications which occur in this case in the intricate expression (42). We shall return to Sec. III observing that in the present case the sum (31) contains a single term:

$$\left(\mu_1, x_1, y_1 \middle| \mu_2, \frac{q_2}{3}, \frac{-q_2}{3} \middle| \mu_3, r\right) = T_0(\mu)\left(\mu_1, x_1, y_1 \middle| \mu_2, \frac{q_2}{3}, \frac{-q_2}{3} \middle| \mu_3, r\right)_0 \tag{43}$$

while eq. (32) gives

$$\begin{aligned}&\left(\mu_1, x_1, y_1 \middle| \mu_2, \frac{q_2}{3}, \frac{-q_2}{3} \middle| \mu_3, r\right)_0 \\ &\qquad = \delta_{r,0}\left[\frac{f(a_3, b_3, c_3; a_2 + x_1)\,f(b_1, a_1, -c_1; y_1)}{f(a_1, b_1, c_1; x_1)\,f(b_3, a_3, -c_3; y_1 + c_2)}\right]^{1/2}.\end{aligned} \tag{44}$$

$T_0(\mu)$ is an undetermined constant. To determine the matrix elements of the other tensors we shall use a method other than that of Sec. IV. From the commutation relation (19b) and the change of functions (33) we shall determine the matrix elements $(\mu_1, x_1, y_1 \mid \mu_2, a_2, c_2 + s \mid \mu_3, 0)_0$.

The recurrence relation (34) with $r = 0$ is:

$$g_{s+1,0}(x_1, y_1) = \frac{g_{s,0}(x_1, y_1)}{x_1 + y_1 + s + 2} - \frac{g_{s,0}(x_1, y_1 + 1)}{x_1 + y_1 + 1} \,. \tag{45}$$

We make the change of function:

$$g_{s,0}(x_1, y) = \frac{(x_1 + y_1)!}{(x_1 + y_1 + s + 1)!} u_s(x_1, y_1) \tag{46}$$

and obtain the recurrence relation

$$u_{s+1}(x_1, y_1) = u_s(x_1, y_1) - u_s(x_1, y_1 + 1) \tag{47}$$

which has the solution:

$$u_s(x_1, y_1) = \sum_{k=0}^{s} (-1)^k \frac{s!}{k!(s-k)!} u_0(x_1, y_1 + k) \,. \tag{48}$$

From (46) and (48) we find finally:

$$\begin{aligned} g_{s,0} = \frac{(x_1 + y_1)!}{(x_1 + y_1 + s + 1)!} \sum_{k=0}^{s} (-1)^k \frac{s!}{k!(s-k)!} \times \\ \times (x_1 + y_1 + k + 1)\, g_{0,0}(x_1, y_1 + k) \,. \end{aligned} \tag{49}$$

The function $g_{0,0}(x_1, y_1)$ is derived from (33) and (44):

$$g_{0,0}(x_1, y_1) = \frac{1}{(q_2!)^{1/2}(x_1 + y_1 + 1)} \; \frac{f(a_3, b_3, c_3; a_2 + x_1)\, f(b_1, a_1, -c_1; y_1)}{f(a_1, b_1, c_1; x_1)\, f(b_3, a_3, -c_3; c_2 + y_1)} \,. \tag{50}$$

Using the relations (33), (49) and (50), the matrix elements $(\mu_1, x_1, y_1 \mid \mu_2, a_2, c_2 + s \mid \mu_3, 0)_0$ are completely determined. To find the other ones we consider the C.R.:

$$\left[K_+, T^{(0,q_2)}_{\frac{s}{2}, \frac{s}{2}, \frac{2q_2}{3} - s}\right] = 0 \tag{51}$$

and the same method as in Sec. III. We than obtain the matrix elements $\left(\mu_1, x_1, y_1 \middle| \mu_2, \frac{q_2}{3}, y_2 \middle| \mu_3, x_3, y_3\right)$ expressed in terms of $(\mu_1, x_1, y_1 \mid \mu_2, a_2, c_2 + s \mid \mu_3, 0)$ derived above.

Expressed in the variables x and y the final result is:

$$\begin{aligned} &\left(\mu_1, x_1, y_1 \middle| \mu_2, \frac{q_2}{3}, y_2 \middle| \mu_3, x_3, y_3\right) \\ &= T(\mu) \Big[(x_1 + y_1 + 1)\,(x_1 + x_2 + y_3 + 1)!\,(-x_1 + y_2 + x_3)! \times \\ &\quad \times (q_2 - x_2 - y_2)! \,\frac{(x_1 + x_2 - x_3)!\, f(a_3, b_3, c_3; x_3)\, f(b_1, a_1, -c_1; y_1)}{(y_1 - x_2 + x_3)!\, f(a_1, b_1, c_1; x_1)\, f(b_3, a_3, -c_3; y_3)}\Big]^{1/2} \times \\ &\quad \times \sum_{k,l} \frac{(-1)^{k+l}}{k!\,l!} [(x_1 + x_2 - x_3 - k)!\,(x_2 + y_2 - l)!]^{-1} \times \\ &\quad \times \frac{(x_1 + y_1 - k)!}{(x_1 + x_2 + y_3 - k)!} \, \frac{f(b_1, a_1, -c_1; y_1 - k + l)\, f(b_3, a_3, -c_3; y_1 + y_2 - k)}{f(b_3, a_3, -c_3; y_1 + c_2 - k + l)\, f(b_1, a_1, -c_1; y_1 - k)} \,. \end{aligned} \tag{52}$$

To compare the present result for the I.T. corresponding to a representation $(0, q)$ with MOSHINSKY's one [6] corresponding to a representation $(q, 0)$ we have to use the symmetry properties of the C.G. coefficients [17]. We then observe that the former is more convenient, containing fewer terms.

Acknowledgement. The authors would like to express their gratitude to Professor V. NOVACU for critical discussions.

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The $\rho \rightarrow 4\pi$ Vertex in Chiral Dynamics

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(Received 1 January 1969)

The $\rho \rightarrow 4\pi$ vertex has been computed assuming vector-meson dominance, and the resulting interaction is described by a chiral-symmetric nonlinear Lagrangian plus a symmetry-breaking term with tensorial properties. The vertex function is used to compute an approximative analytical expression for the $e^+e^- \rightarrow 4\pi$ cross section which is valid from the threshold up to 850 MeV. For the four-pion decay of the ρ meson, we have obtained the following branching ratios: $\Gamma_{\rho^0\rightarrow 2\pi^+2\pi^-}/\Gamma_{\rho^0\rightarrow\pi^+\pi^-} \approx 10^{-4}$; $\Gamma_{\rho^0\rightarrow 2\pi^0\pi^+\pi^-}/\Gamma_{\rho^0\rightarrow 2\pi^+2\pi^-} = \frac{1}{4}$; $\Gamma_{\rho^+\rightarrow 2\pi^+\pi^-\pi^0}/\Gamma_{\rho^0\rightarrow 2\pi^+2\pi^-} = \frac{3}{4}$. Experiment indicates upper limits of the order 1.5×10^{-3} for the four-pion decay-mode branching ratios.

ONE of the most interesting results of the nonlinear chiral $SU_2\times SU_2$ effective-Lagrangian model is the prediction of various many-pion vertices. Recently Olsson and Turner[1] have shown that chiral $SU_2\times SU_2$ symmetry with $(\frac{1}{2},\frac{1}{2})$ tensorial breaking (σ model) gives fairly good results for the production reaction $\pi^-p \rightarrow \pi^+\pi^-n$ at the threshold. In order to test the model further, we might consider many-pion production in pion-nucleon collisions. This type of calculation is, however, hard to perform since the contributions of different resonances are important and their couplings are unknown; thus supplementary assumptions would be necessary. This is why we have considered the $\rho \rightarrow 4\pi$ vertex, which does not imply new coupling constants and which can be directly measured in the $\rho \rightarrow 4\pi$ decay and (in the vector-meson-dominance model) in the reaction $e^+e^- \rightarrow 4\pi$.

Choosing the nonlinear transformations of the fields as in the σ model and assuming vector-meson dominance, the chiral-symmetric Lagrangian[2] is

$$\begin{aligned}\mathfrak{L}^{\text{sym}} = &-\tfrac{1}{4}(\mathbf{f}_{\mu\nu}+g\mathbf{a}_\mu\times\mathbf{a}_\nu)^2-\tfrac{1}{4}(D_\mu\mathbf{a}_\nu-D_\nu\mathbf{a}_\mu)^2\\ &-\tfrac{1}{2}m_\rho^2(\boldsymbol{\rho}_\mu^2+\mathbf{a}_\mu^2)-(\partial_\mu\sigma+g\boldsymbol{\pi}\mathbf{a}_\mu)^2-(D_\mu\boldsymbol{\pi}-g\sigma\mathbf{a}_\mu)^2\\ &+\frac{g\delta}{m_\rho^2}[(\mathbf{f}_{\mu\nu}+g\mathbf{a}_\mu\times\mathbf{a}_\nu)\cdot(D_\mu\boldsymbol{\pi}-g\sigma\mathbf{a}_\mu)\times(D_\nu\boldsymbol{\pi}-g\sigma\mathbf{a}_\nu)\\ &\qquad+2(D_\mu\mathbf{a}_\nu-D_\nu\mathbf{a}_\mu)\cdot(D_\mu\boldsymbol{\pi}-g\sigma\mathbf{a}_\mu)(\partial_\nu\sigma+g\boldsymbol{\pi}\cdot\mathbf{a}_\nu)],\end{aligned}$$

where

$$\mathbf{f}_{\mu\nu}=\boldsymbol{\rho}_{\mu\nu}+g\boldsymbol{\rho}_\mu\times\boldsymbol{\rho}_\nu,\quad \mathbf{a}_\mu=\mathbf{A}_\mu+\frac{1}{m_A}D_\mu\boldsymbol{\pi},\quad D_\mu=\partial_\mu+g\boldsymbol{\rho}_\mu\times,$$

$$\sigma=(f_\pi^2-\boldsymbol{\pi}^2)^{1/2}=f_\pi-\frac{1}{2f_\pi}\boldsymbol{\pi}^2-\frac{1}{8f_\pi^3}\boldsymbol{\pi}^4+\cdots,$$

$$f_\pi=\frac{m_A}{2g}\quad(m_A=m_\rho\sqrt{2}).$$

We consider that the symmetry-breaking part of the Lagrangian is composed of two parts: one depending only on the pion field, containing also the pion mass term, and the other containing the $\omega\rho\pi$ vertex. Both these parts can be written as $(N/2,N/2)$ tensors,[3-5] but the former will modify the partial conservation of axial-vector current (PCAC).[6] For our purpose it is enough to retain only the pion mass term, the four-pion interaction, and the $\omega\rho\pi$ interaction:

$$\mathfrak{L}^{\text{break}}=-\tfrac{1}{2}m_\pi^2\boldsymbol{\pi}^2-\xi\frac{m_\pi^2}{8f_\pi^2}\boldsymbol{\pi}^4+\frac{gh}{4m_\rho}\boldsymbol{\rho}_{\mu\nu}\boldsymbol{\pi}\omega_{\lambda\sigma}\epsilon_{\mu\nu\lambda\sigma}+\cdots,$$

where for an $(N/2,N/2)$ tensorial breaking

$$\xi=[8-N(N+2)]/5,$$

and for Schwinger's mass-term breaking $\xi=4$, while $h\approx2$ from the $\omega\rightarrow\pi\gamma$ decay width.

With these Lagrangians, we have in the tree-diagram approximation eight Feynman graphs (see Fig. 1) to describe the effective $\rho\rightarrow4\pi$ vertex. (However, the last two diagrams do not contribute to the most interesting $\rho^0\rightarrow2\pi^+2\pi^-$ vertex.)

We define an effective ρ-4π Lagrangian by summing the contributions of all these diagrams. We shall work in the momentum space, using the notation

$$\int dx\,\mathfrak{L}(x)=(2\pi)^4\int dp_1\cdots dp_n\delta(\sum^n p_i)\mathfrak{L}'(p_1,\cdots p_n),$$

$$\begin{aligned}\mathfrak{L}'_{\rho4\pi}=\frac{ig^2}{m_\rho^2}&\Gamma_\mu^{i\alpha\beta\gamma\delta}(q,p_1,p_2,p_3,p_4)\\ &\times\rho_\mu^i(q)\pi^\alpha(p_1)\pi^\beta(p_2)\pi^\gamma(p_3)\pi^\delta(p_4).\end{aligned}$$

Because we are not too far from the threshold (the mean kinetic energy per pion being 50 MeV), even for the physical mass of the ρ meson, we feel content to evaluate the form factors at the threshold. We believe that the error introduced in this way is not important, but it introduces essential simplifications especially when computing the phase-space integrals. At the threshold ($q^2=-16m_\pi^2$), we find

$$\Gamma_\mu^{i\alpha\beta\gamma\delta}=C\delta^{\gamma\delta}\epsilon_{i\alpha\beta}p_\mu^1,$$

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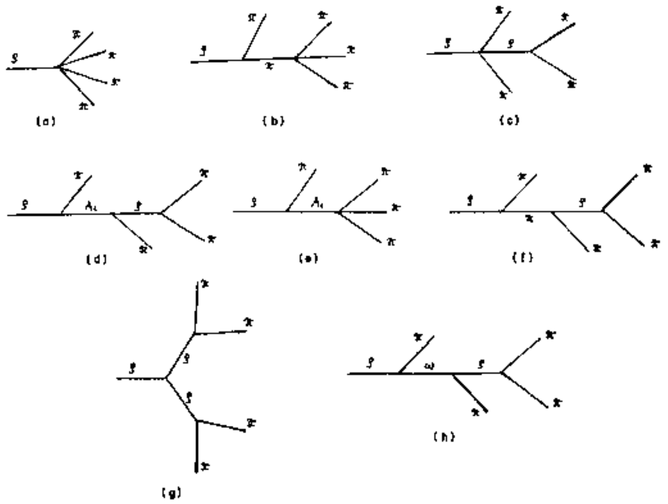


FIG. 1. Feynman graphs considered in the calculation of the effective $\rho \to 4\pi$ vertex.

where the constant C is equal to 1 for $m_\pi = 0$. For the actual pion mass its value is

$$C = 1.074 + 0.2162\xi + (0.1557 + 0.0333\xi)\delta - 0.0055\delta^2 - 0.0001\delta^3.$$

For $0 < \delta < 1$ and N not too high, C is close to its soft-pion value.

It is interesting to remark here that the contributions of diagrams f, g, and h vanish at the threshold. The most important diagrams are a, b, and c, all being of the same order of magnitude, but the last two are of opposite sign, so that the contribution of the contact graph is dominant (in the soft-pion limit it alone gives $C = 1$).

Now, according to the vector-meson-dominance graphs of Fig. 2 the ratio of the cross sections for the processes $e^+e^- \to 4\pi$ and $e^+e^- \to 2\pi$ is

$$\frac{\sigma_{e^+e^- \to 2\pi^+2\pi^-}(W)}{\sigma_{e^+e^- \to \pi^+\pi^-}(W)} = \left(\frac{g^2}{4\pi}\right)^2 C^2 \frac{W}{[1-(W^2/4m_\rho^2)(1-\delta)]^2 (W^2-4m_\pi^2)^{3/2}} \frac{2}{\pi^5 m_\rho^4} I(W)$$

$$\frac{\sigma_{e^+e^- \to 2\pi^0\pi^+\pi^-}(W)}{\sigma_{e^+e^- \to 2\pi^+2\pi^-}(W)} = \tfrac{1}{4},$$

where $W = \sqrt{(-q^2)}$ is the c.m. energy, and

$$I(W) = \frac{\pi^3}{4W^2} \int_{4m_\pi^2}^{\infty} ds_1 \int_{4m_\pi^2}^{\infty} ds_2 \left\{ \frac{(s_1 - 4m_\pi^2)(s_2 - 4m_\pi^2)[W^2 - (\sqrt{s_1} + \sqrt{s_2})^2][W^2 - (\sqrt{s_1} - \sqrt{s_2})^2]}{s_1 s_2} \right\}^{1/2}$$
$$\times (s_1 - 4m_\pi^2)\left(1 + \frac{s_1 - s_2}{W^2}\right) \times \theta(W - \sqrt{s_1} - \sqrt{s_2}).$$

In the vicinity of the threshold (first nonvanishing order in $W - 4m_\pi$) the integral can be computed exactly and

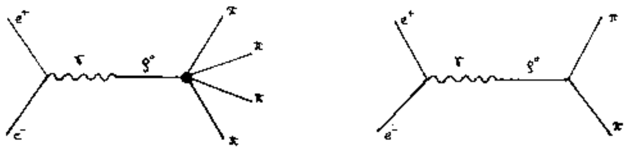
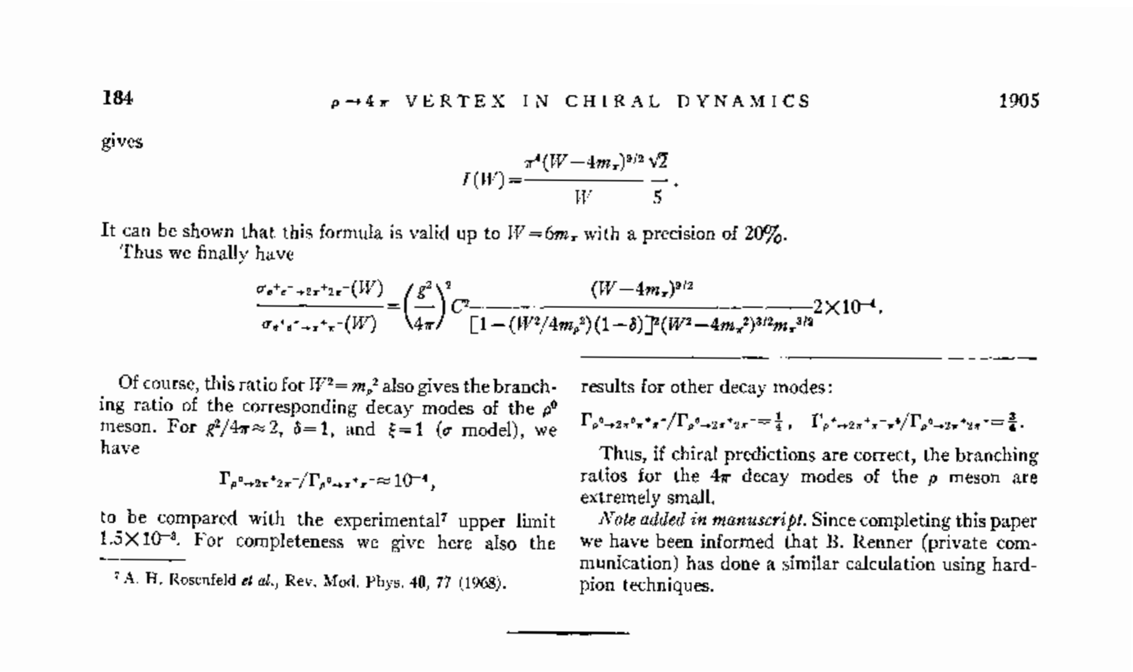


FIG. 2. ρ-meson dominance graphs for the processes $e^+e^- \to 4\pi$ and $e^+e^- \to 2\pi$.



gives

$$I(W)=\frac{\pi^4(W-4m_\pi)^{9/2}}{W}\frac{\sqrt{2}}{5}\,.$$

It can be shown that this formula is valid up to $W=6m_\pi$ with a precision of 20%.

Thus we finally have

$$\frac{\sigma_{e^+e^-\to 2\pi^+2\pi^-}(W)}{\sigma_{e^+e^-\to\pi^+\pi^-}(W)}=\left(\frac{g^2}{4\pi}\right)^2 C^2\frac{(W-4m_\pi)^{9/2}}{[1-(W^2/4m_\rho^2)(1-\delta)]^2(W^2-4m_\pi^2)^{3/2}m_\pi^{3/2}}2\times10^{-4}.$$

Of course, this ratio for $W^2=m_\rho^2$ also gives the branching ratio of the corresponding decay modes of the ρ^0 meson. For $g^2/4\pi\approx2$, $\delta=1$, and $\xi=1$ (σ model), we have

$$\Gamma_{\rho^0\to2\pi^+2\pi^-}/\Gamma_{\rho^0\to\pi^+\pi^-}\approx10^{-4},$$

to be compared with the experimental[7] upper limit 1.5×10^{-3}. For completeness we give here also the results for other decay modes:

$$\Gamma_{\rho^0\to2\pi^0\pi^+\pi^-}/\Gamma_{\rho^0\to2\pi^+2\pi^-}=\tfrac{1}{4}\,,\quad \Gamma_{\rho^+\to2\pi^+\pi^-\pi^0}/\Gamma_{\rho^0\to2\pi^+2\pi^-}=\tfrac{3}{4}.$$

Thus, if chiral predictions are correct, the branching ratios for the 4π decay modes of the ρ meson are extremely small.

Note added in manuscript. Since completing this paper we have been informed that B. Renner (private communication) has done a similar calculation using hard-pion techniques.

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PHYSICAL REVIEW D VOLUME 8, NUMBER 2 15 JULY 1973

Null-Plane Field Algebra*

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(Received 25 July 1972)

Field-algebra current commutators on the null plane are deduced using Dirac's canonical quantization method. The corresponding scaling relations are $F_2(\omega) = -2\omega F_5(\omega)$, $F_3(\omega) = 0$.

INTRODUCTION

In view of the recent interest in the study of the light-cone structure of current commutators, related to the scaling properties of deep-inelastic experiments, canonical quantization on the null plane of the field-theoretical models has proved very instructive. Along these lines the quark model has been extensively studied.[1]

In this paper we obtain the null-plane commutators of the field-algebra model of Lee, Weinberg, and Zumino.[2] Our results show the singularity structure needed for Bjorken[3] scaling.

For quantizing the theory we use a null-plane version of Dirac's[4] general method of quantizing the theories with constraints, which is based on the redefinition of the classical Poisson bracket and the correspondence principle.

In Sec. I we give a brief account of Dirac's canonical formalism. After presenting the field-algebra Lagrangian in Sec. II we analyze the second-class constraints that emerge. The redefinition of the Poisson bracket enables us to obtain the relevant current commutators.

Section III is devoted to the analysis of the singularity structures of the matrix elements of the commutator to deduce the scaling properties of deep-inelastic scattering.

I. CANONICAL FORMALISM

When passing from the Lagrangian form of a classical theory to the canonical one, it may happen that the equations that define the canonical momenta

$$p_n = \frac{\partial L}{\partial \dot{q}_n}\ , \quad n = 1, \ldots, N \tag{1.1}$$

are not all consistent with each other unless some constraints

$$\phi_r(p, q) = 0\ , \quad r = 1, \ldots, R \tag{1.2}$$

between the canonically conjugate variables are satisfied.

(This is generally the situation in the null-plane formulation of relativistic field theories.)

However, the usual Poisson bracket of ϕ_r with an arbitrary dynamical variable A,

$$\{\phi_r, A\} = \sum_{n=1}^{N} \left(\frac{\partial \phi_r}{\partial q_n}\frac{\partial A}{\partial p_n} - \frac{\partial \phi_r}{\partial p_n}\frac{\partial A}{\partial q_n} \right),$$

may not vanish. Therefore Eq. (1.2) must be considered only in a certain "weak" sense. Clearly, if we would establish a correspondence principle $(1/i)\{\ ,\ \} \to [\ ,\ ]$ as is usually done in canonical quantization, we would have inconsistencies with imposing (1.2) as an operator relation.

According to Dirac[4] we call the constraints (1.2) obtained from (1.1) *primary constraints*, whereas those obtained by imposing their conservation in time are called *secondary constraints*. The whole system of primary and secondary constraints may be divided into constraints of the first class and the second class. A constraint is said to be of the *first class* if

$$\{\phi_p, \phi_r\} = g^{p}_{rr'}\,\phi_{r'}$$

for any other constraint ϕ_r.

Let us suppose that we have succeeded in separating all the first-class constraints, so that no linear combinations of the other can be made of the first class. Then these remaining constraints are called *second-class constraints*.

The first-class constraints remain weak constraints, whereas the second-class ones can be made strong ones by defining a new Poisson bracket according to

$$\{A, B\}^* = \{A, B\} - \sum_{s,s'}^{S} \{A, \phi_s\} C_{ss'} \{\phi_{s'}, B\}\ , \tag{1.3}$$

where

$$\sum_{s''}^{S} C_{ss''} \{\phi_{s''}, \phi_{s'}\} = \delta_{ss'}\ , \tag{1.4}$$

$$C_{ss'} = -C_{s's}\ , \tag{1.5}$$

and ϕ_s $(s = 1, \ldots, S)$ are all the second-class constraints.

It can be shown that the new Poisson bracket has all the properties of a usual Poisson bracket and conserves the form of the equation of motion. Furthermore, as can be shown, for any second-class constraint ϕ_s,

$$\{\phi_s, A\}^* = 0 \ .$$

Therefore, when quantizing the theory, according to the correspondence principle

$$\frac{1}{i}\{,\}^* \rightarrow [\,,\,] \,,$$

the second-class constraints can be looked upon as operational relations while the first-class ones may be imposed only as constraints on the admissible states.

Fortunately, in our example, there will be no first-class constraints and the whole problem is reduced to the redefinition of the Poisson bracket.

An original aspect of our case is that we study a canonical formalism with the "null" time

$$x^+ = \frac{x^0 + x^3}{\sqrt{2}}$$

in the three-dimensional space defined by the "null" coordinates

$$x^- = \frac{x^0 - x^3}{\sqrt{2}} \ ,$$

$$x_\perp^a = x^a \qquad (a = 1, 2) \ .$$

The Dirac equations (1.4) for bosons here are first-order differential equations, but there are enough conditions to fix the solutions.

This procedure of quantization on the null plane gives the correct result for free field theories; however, its equivalence to ordinary (equal-time) quantization in the general case is not so obvious.

II. SECOND-CLASS CONSTRAINTS

We consider a field-algebra Lagrangian built from vector and axial-vector fields V_i^μ, A_i^μ ($i = 1, \ldots, 8$) transforming as the regular representation of the chiral $SU_3 \times SU_3$ group in minimal interaction with some massless spinor fields ψ_α ($\alpha = 1, \ldots, N$) that together with[5] $\gamma_5\psi_\alpha$ transform according to the $(N, 1) + (1, N)$ representation. That is, with the definitions

$$V_i^{(R,L)\mu} = \frac{1}{\sqrt{2}}\,(V_i^\mu \pm A_i^\mu) \ , \tag{2.1}$$

$$\psi_\alpha^{(R,L)} = \tfrac{1}{2}(1 + i\gamma_5)\psi_\alpha \tag{2.1'}$$

corresponding to the right and left SU_3 groups generated by the charges

$$Q_i^{(R,L)} = \tfrac{1}{2}(Q_i \pm Q_{5i}) \ ,$$

we have

$$[Q_i^{(R,L)}, V_j^{(R,L)\mu}] = i f_{ijk} V_k^{(R,L)\mu} \ ,$$
$$[Q_i^{(L,R)}, V_j^{(R,L)\mu}] = 0 \ , \tag{2.2}$$

$$[Q_i^{(R,L)}, \psi_\alpha^{(R,L)}] = -t^i_{\alpha\beta}\psi_\beta^{(R,L)} \ ,$$
$$[Q_i^{(L,R)}, \psi_\alpha^{(R,L)}] = 0 \ , \tag{2.2'}$$

$$\sum_{\gamma=1}^{N} (t^i_{\alpha\gamma}t^j_{\gamma\beta} - t^j_{\alpha\gamma}t^i_{\gamma\beta}) = i f_{ijk} t^k_{\alpha\beta} \ .$$

Introducing the notations

$$F_{\mu\nu}^{(R,L)i} = \partial_\mu V_\nu^{(R,L)i} - \partial_\nu V_\mu^{(R,L)i} - g\sqrt{2}\, f_{ijk} V_\mu^{(R,L)j} V_\nu^{(R,L)k} \ , \tag{2.3}$$

$$\overrightarrow{\mathfrak{D}}_\mu^{(R,L)}\psi^{(R,L)} = (\partial_\mu - ig\sqrt{2}\ t^i V_\mu^{(R,L)i})\psi^{(R,L)} \tag{2.3'}$$

for the covariant rotors and derivatives, our Lagrangian is the sum of two Lagrangians:

$$\mathfrak{L} = \mathfrak{L}^R + \mathfrak{L}^L \ , \tag{2.4}$$

where

$$\mathfrak{L}^{R,L} = -\tfrac{1}{4}F_{\mu\nu}^{(R,L)i}F_i^{(R,L)\mu\nu} + \tfrac{1}{2}m^2 V_\mu^{(R,L)i} V_i^{(R,L)\mu} + \tfrac{1}{2}i\bar{\psi}^{(R,L)}\overleftrightarrow{\mathfrak{D}}_\mu^{(R,L)}\gamma^\mu\psi^{(R,L)} \qquad (\overleftrightarrow{\mathfrak{D}} = \overrightarrow{\mathfrak{D}} - \overleftarrow{\mathfrak{D}}) \ . \tag{2.5}$$

This Lagrangian gives rise to the current-field identities

$$j_i^\mu = -\frac{m^2}{g}\,V_i^\mu \ ,$$
$$j_i^{5\mu} = -\frac{m^2}{g}\,A_i^\mu \ , \tag{2.6}$$

for the conserved currents. [Note that in our notation this corresponds to $j_i^{(R,L)\mu} = -(m^3/g\sqrt{2}\,)V_i^{(R,L)\mu}$.]

Since the variables of the two Lagrangians $\mathfrak{L}^R$ and $\mathfrak{L}^L$ are independent and uncoupled, our $SU_3 \times SU_3$ problem decouples into two independent SU_3 problems.

In what follows we shall omit therefore the R and L indices, working out the commutators inside one group of variables, keeping in mind that

$$j_i^\mu = \tfrac{1}{2}(j_i^{(R)\mu} + j_i^{(L)\mu}) \ ,$$
$$j_i^{5\mu} = \tfrac{1}{2}(j_i^{(R)\mu} - j_i^{(L)\mu}) \ , \tag{2.7}$$

and

$$[j_i^{(R)\mu}(x), j_j^{(L)\mu}(y)] = 0 \ . \tag{2.8}$$

It is useful for null-plane purposes to introduce[6]

$$\psi_\pm = P_\pm \psi \,, \qquad \bar{\psi}_\pm = \frac{1}{\sqrt{2}}\, \psi_\pm^* \gamma^\circ \,. \tag{2.9}$$

Then the canonical conjugate momenta (on the null plane) of our fields are

$$\Pi_i^\mu = \frac{\partial \mathfrak{L}}{\partial \partial_- V_i^\mu} \,, \qquad p_\pm^\alpha = \frac{\partial \mathfrak{L}}{\partial \partial_+ \psi_\pm^\alpha} \,, \qquad \bar{p}_\pm^\alpha = \frac{\partial \mathfrak{L}}{\partial \partial_+ \bar{\psi}_\pm^\alpha} \,, \tag{2.10}$$

satisfying the nonvanishing Poisson bracket relations

$$\{V_i^\mu(x), \Pi_j^\nu(y)\}_{x^+=y^+} = \delta^{\mu\nu}\delta_{ij}\delta^3(x-y) \,,$$
$$\{\psi_\pm^\alpha(x), p_\pm^\beta(y)\}_{x^-=y^+} = P_\pm \delta_{\alpha\beta}\delta^3(x-y) \,, \tag{2.11}$$
$$\{\bar{\psi}_\pm^\alpha(x), \bar{p}_\pm^\beta(y)\}_{x^+=y^-} = P_\pm \delta_{\alpha\beta}\delta^3(x-y) \,.$$

As is easy to see, Eqs. (2.10) give rise to the primary constraints

$$\begin{aligned}
\phi_i &\equiv \Pi_i^- = 0 \,,\\
\phi_i^a &\equiv \Pi_i^a - F_i^{+a} = 0 \,,\\
\phi_+^\alpha &\equiv p_+^\alpha - \tfrac{1}{2} i \bar{\psi}_+^\alpha \gamma^+ = 0 \,,\\
\bar{\phi}_+^\alpha &\equiv \bar{p}_+^\alpha + \tfrac{1}{2} i \gamma^+ \psi_+^\alpha = 0 \,,\\
\phi_- &\equiv p_- = 0 \,,\\
\bar{\phi}_- &\equiv \bar{p}_- = 0 \,.
\end{aligned} \tag{2.12}$$

The only nonvanishing Poisson brackets of these constraints are $\{\phi_i^a(x), \phi_j^b(y)\}$, $\{\phi_+^\alpha(x), \bar{\phi}_+^\beta(y)\}$. The Hamiltonian[7] is

$$\mathcal{H} = \int d^3x [\Pi_i^+(\tfrac{1}{2}\Pi_i^+ + D_{ij}^+ V_j^-) + \Pi_i^a D_{ij}^a V_j^- + \tfrac{1}{4}(F_i^{ab})^2 - \tfrac{1}{2} m^2 (2 V_i^- V_i^+ - V_i^a V_i^a)$$
$$+ ig\sqrt{2}\, V_i^-(p_+ t^i \psi_+ - \bar{\psi}_+ t^i \bar{p}_+) - \tfrac{1}{2} i(\bar{\psi}_- \gamma^- \bar{\mathfrak{D}}^+ \psi_- - \bar{\psi}_+ \gamma^a \bar{\mathfrak{D}}^a \psi_- - \bar{\psi}_- \gamma^a \bar{\mathfrak{D}}^a \psi_+)]$$
$$(D_{ij}^\mu = \delta_{ij}\partial^\mu + g\sqrt{2}\, f_{ikj} V_k^\mu) \,. \tag{2.13}$$

Now we have to impose the conservation condition for our primary constraints. From $\{\phi_i(x), \mathcal{H}\} = 0$ we get a new constraint

$$\chi_i \equiv D_{ij}^+ \Pi_j^+ + D_{ij}^a \Pi_j^a + m^2 V_i^+$$
$$- ig\sqrt{2}\,(p_+ t^i \psi_+ - \bar{\psi}_+ t^i \bar{p}_-) = 0 \tag{2.14}$$

and from $\{\chi_i(x), \mathcal{H}\} = 0$ another one

$$\tilde{\chi}_i \equiv 2\partial^+ V_i^- - \partial^a V_i^a + g\sqrt{2}\, f_{ijk} V_j^+ V_k^- + \Pi_i^+ = 0 \,. \tag{2.15}$$

Similarly we get two new constraints χ_-^α and $\bar{\chi}_-^\alpha$ from $\{\phi_-^\alpha, \mathcal{H}\} = 0$ and $\{\bar{\phi}_-^\alpha, \mathcal{H}\} = 0$. The remaining constraint conservation equations fix the arbitrariness of the Hamiltonian[4] or reduce to previous constraints. As can be seen, there are no first-class constraints in our problem. Furthermore, we do not have to work with all the constraints ϕ_i, ϕ_i^a, ϕ_+^α, $\bar{\phi}_+^\alpha$, ϕ_-^α, $\bar{\phi}_-^\alpha$, χ_i, $\tilde{\chi}_i$, χ_-^α, $\bar{\chi}_-^\alpha$. If a constraint just states the vanishing of a given canonical momentum then the redefinition of the Poisson bracket is equivalent with respect to this variable to express this canonical coordinate through the other canonical variables using another constraint equation.[4] In our case in this manner we get rid of three variables and six cooresponding constraints. (The fields V_i^-, ψ_-^α, $\bar{\psi}_-^\alpha$ whose canonical conjugate momenta vanish we shall consider as functions of the other fields[8] through the relations $\tilde{\chi}_i = 0$, $\chi_-^\alpha = 0$, $\bar{\chi}_-^\alpha = 0$.)

Thus we remain with the constraints ϕ_i^a, χ_i, ϕ_+^α, $\bar{\phi}_+^\alpha$ and their only nonvanishing Poisson brackets

$$\{\phi_i^a(x), \phi_j^b(y)\}_{x^+=y^+} = -2\delta_{ab} D_{ij}^+ \delta^3(x-y) \,,$$
$$\{\chi_i(x), \chi_j(y)\}_{x^+=y^-}$$
$$= -m^2(2D_{ij}^- + g\sqrt{2}\, f_{ijk} V_k^+(x))\delta^3(x-y) \,, \tag{2.16}$$
$$\{\phi_+^\alpha(x), \bar{\phi}_+^\beta(y)\}_{x^-=y^-} = -i\gamma^- \delta_{\alpha\beta}\delta^3(x-y)$$

to work out the redefinition of the Poisson bracket. Therefore the index s of the formulas (1.3)–(1.5) in our problem goes through values corresponding to ϕ_i^a, χ_i, ϕ_+^α, $\bar{\phi}_+^\alpha$ and to the continuum x^-, $\vec{x}_\perp$.

Since the spinor constraints ϕ_+^α, $\bar{\phi}_+^\alpha$ have vanishing Poisson brackets with the vector-meson constraints ϕ_i^a, χ_i, the quantization of the basic spinor fields ψ_+^α is completely decoupled from that of the basic boson fields (V_i^+, V_i^a). The result for the spinor fields ψ_+^α is the same as is given in Ref. 9. Here we are interested only in the commutator of the fields V_i^+, V_i^a; thus we consider the equations [cf. (1.4)]

$$\begin{aligned}
&\int d^3z\, C_{\chi_i,x;\chi_k,z}\{\chi_k(z),\chi_j(y)\} = \delta_{ij}\delta^3(x-y)\,,\\
&\int d^3z\, C_{\chi_i,x;\phi_k^b,z}\{\phi_k^b(z),\phi_j^a(y)\} = 0\,,\\
&\int d^3z\, C_{\phi_i^a,x;\chi_k,z}\{\chi_k(z),\chi_j(y)\} = 0\,,\\
&\int d^3z\, C_{\phi_i^a,x;\phi_k^c,z}\{\phi_k^c(z),\phi_j^b(y)\} = \delta_{ab}\delta_{ij}\delta^3(x-y)\,,
\end{aligned} \tag{2.17}$$

with

$$\begin{aligned}
&C_{\chi_i,x;\chi_j,y} = -C_{\chi_j,y;\chi_i,x}\,,\\
&C_{\chi_i,x;\phi_j^a,y} = -C_{\phi_j^a,y;\chi_i,x}\,,\\
&C_{\phi_i^a,x;\phi_j^b,y} = -C_{\phi_j^b,y;\phi_i^a,x}\,.
\end{aligned} \tag{2.18}$$

With the matrix notations

$$\begin{aligned}
&C_{\chi_i,x;\chi_j,y} = C(x,y)_{ij}\,,\\
&C_{\chi_i,x;\phi_j^a,y} = C^{\cdot a}(x,y)_{ij}\,,\\
&C_{\phi_i^a,x;\phi_j^b,y} = C^{ab}(x,y)_{ij}\,,
\end{aligned} \tag{2.19}$$

and

$$g\sqrt{2}\,f_{ikj}V_k^+(x) = \mathfrak{V}^+(x)_{ij}\,, \tag{2.20}$$

using (2.16), Eqs. (2.17) may be written as

$$\begin{aligned}
&[\partial_-^x + \tfrac{1}{2}\mathfrak{V}^-(x)]C(x,y) = -\frac{1}{2m^2}\delta^3(x-y)\,,\\
&[\partial_-^x + \mathfrak{V}^+(x)]C^{a\cdot}(x,y) = 0\,,\\
&[\partial_-^x + \tfrac{1}{2}\mathfrak{V}^-(x)]C^{\cdot a}(x,y) = 0\,,\\
&[\partial_-^x + \mathfrak{V}^+(x)]C^{ab}(x,y) = -\tfrac{1}{2}\delta^{ab}\delta^3(x-y)\,.
\end{aligned} \tag{2.21}$$

The solution of the first of these equations may be written as

$$C(x,y) = -\frac{1}{4m^2}\,[\epsilon(x^- - y^-)\delta(\vec{x}_\perp - \vec{y}_\perp)\Omega(x,y) + X(x,y)]\,, \tag{2.22}$$

where

$$\begin{aligned}
&[\partial_-^x + \tfrac{1}{2}\mathfrak{V}^-(x)]\Omega(x,y) = 0\,,\\
&\Omega(x,x) = 1\,,\\
&\Omega(x,y)^T = \Omega(y,x)
\end{aligned} \tag{2.23}$$

and

$$\begin{aligned}
&[\partial_-^x + \tfrac{1}{2}\mathfrak{V}^-(x)]X(x,y) = 0\,,\\
&X(x,y)^T = -X(y,x)\,.
\end{aligned} \tag{2.24}$$

The conditions (2.23) fix $\Omega(x,y)$ completely:

$$\begin{aligned}
\Omega(x,y) &= \theta(x^- - y^-)T\exp\left(-\tfrac{1}{2}\int_{y^-}^{x^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right) + \theta(y^- - x^-)\tilde{T}\exp\left(-\tfrac{1}{2}\int_{y^-}^{x^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right)\\
&= T\exp\left(-\tfrac{1}{2}\int_{-\infty}^{x^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right)\tilde{T}\exp\left(\tfrac{1}{2}\int_{-\infty}^{y^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right)
\end{aligned} \tag{2.25}$$

(T and $\tilde{T}$ being the chronological and antichronological product symbols with respect to the variable z^-), while the conditions (2.24) fix $X(x,y)$ up to an arbitrary antisymmetric matrix $\mathfrak{X}(x^+;\vec{x}_\perp,\vec{y}_\perp)$,

$$X(x,y) = T\exp\left(-\tfrac{1}{2}\int_{-\infty}^{x^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right)\mathfrak{X}(x^+;\vec{x}_\perp,\vec{y}_\perp)\tilde{T}\exp\left(\tfrac{1}{2}\int_{-\infty}^{y^-} dz^-\mathfrak{V}^-(x^+,z^-,\vec{x}_\perp)\right)\,. \tag{2.26}$$

On the other hand, due to causality[10]

$$\mathfrak{X}(x^+;\vec{x}_\perp,\vec{y}_\perp) = \delta(\vec{x}_\perp - \vec{y}_\perp)R(x^+,\vec{x}_\perp)\,, \tag{2.27}$$

with

$$R^T = -R\,.$$

Analogously we may solve all the equations (2.21) up to some arbitrary functions of x^+, $\vec{x}_\perp$ [due to the fact that Dirac's equations (1.4) for our system with continuous degrees of freedom become first-order differential equations]. We shall see that this arbitrariness may be eliminated, at least if we ignore our remark in Ref. 10.

Up to this uniqueness problem, the new Poisson brackets of the fields V_i^+, V_i^c are thus determined. To obtain the commutators containing also V_i^- we would have to solve Eq. (2.15). Its solution is again determined up to an arbitrary function of x^+, $\vec{x}_\perp$. However, on this function one has to impose some additional requirements assuring the locality of $V_i^-(x)$, which may determine it completely. Fortunately we do not need the commutators of V_i^- since all the relevant information for deep-inelastic scattering may be obtained from the commutators:

$$[j_i^+(x),j_j^+(y)]_{x^+=y^+} = if_{ijk}j_k^+\delta^3(x-y) + \frac{im^4}{2g^2}\,\partial_-^x\partial_-^y C(x,y)_{ij}\,, \tag{2.28}$$

$$[j_i^+(x), j_j^a(y)]_{x^+=y^+} = if_{ijk} j_k^a \delta^3(x-y) + \frac{m^2}{2g^2} \partial_a^x \delta^3(x-y)\delta_{ij} + \frac{im^4}{2g^2} \partial_-^x \{C(x,y)\mathcal{V}^a(y) - \partial_y^a C(x,y) - 3C'^a(x,y)\}_{ij} \,. \tag{2.29}$$

If we want to ensure the interpretation of the null-plane charges

$$\bar{Q}_i = \int d\vec{\mathbf{x}}_\perp \int dx^- j_i^+(x)$$

as generators of the SU_3 group, we have to impose the conditions[11]

$$\int d\vec{\mathbf{x}}_\perp \int dx^- \partial_-^x C(x,y) = 0 \,, \quad \int d\vec{\mathbf{x}}_\perp \int dx^- \partial_-^x C'^a(x,y) = 0 \,. \tag{2.30}$$

The first of these determines our arbitrary matrix R as

$$R(x^+, \vec{\mathbf{x}}_\perp) = \frac{1 + T\exp[-\frac{1}{2}\int_{-\infty}^{\infty} dz^- \mathcal{V}^+(x^-, z^-, \vec{\mathbf{x}}_\perp)]}{1 - T\exp[-\frac{1}{2}\int_{-\infty}^{\infty} dz^- \mathcal{V}^+(x^+, z^-, \vec{\mathbf{x}}_\perp)]} \tag{2.31}$$

while the second gives $C'^a(x,y) = 0$.

III. DEEP-INELASTIC SCATTERING

In this section we shall proceed to analyze the singularities of (2.28)–(2.29) from the point of view of deep-inelastic scattering. We are interested in the connected part of

$$t_{\mu\nu}^{ij}(x,p) = \langle p | [j_\mu^i(x), j_\nu^j(0)] | p \rangle \,, \tag{3.1}$$

where p is the 4-momentum of the one-particle state $|p\rangle$ of mass M (averaging over spin is understood).

The decomposition of $t_{\mu\nu}^{ij}$ into invariant amplitudes is

$$\begin{aligned} t_{\mu\nu}^{ij}(x,p) = {} & (\partial_\mu \partial_\nu - g_{\mu\nu}\partial_\lambda \partial^\lambda) f_1^{ij}(x^2, p\cdot x) \\ & - [p_\mu p_\nu \partial_\lambda \partial^\lambda - (p_\mu \partial_\nu - p_\nu \partial_\mu) p_\lambda \partial^\lambda + g_{\mu\nu} p_\lambda \partial^\lambda p_\sigma \partial^\sigma] f_2^{ij}(x^2, p\cdot x) + \epsilon_{\mu\nu\lambda\sigma} p^\lambda \partial^\sigma f_3^{ij}(x^2, p\cdot x) \,, \end{aligned} \tag{3.2}$$

and correspondingly for its Fourier transform

$$t_{\mu\nu}^{ij}(q,p) = \frac{1}{2\pi} \int dx\, e^{iq\cdot x}\, t_{\mu\nu}^{ij}(x,p) \tag{3.3}$$

we have

$$t_{\mu\nu}^{ij}(q,p) = (q_\mu q_\nu - g_{\mu\nu} q^2)\mathfrak{F}_1^{ij}(\nu, q^2) - [p_\mu p_\nu q^2 - (p_\mu q_\nu + p_\nu q_\mu)\nu + g_{\mu\nu}\nu^2]\mathfrak{F}_2^{ij}(\nu, q^2) + i\epsilon_{\mu\nu\lambda\sigma} p^\lambda q^\sigma \mathfrak{F}_3^{ij}(\nu, q^2) \,, \tag{3.4}$$

where

$$\mathfrak{F}_n^{ij}(\nu, q^2) = -\frac{1}{2\pi} \int dx\, e^{iq\cdot x} f_n^{ij}(x^2, p\cdot x) \tag{3.5}$$

with $\nu = p\cdot q$.

The general form of $f_n^{ij}(x^2, p\cdot x)$ due to causality and spectral conditions is (N is finite)

$$f_n^{ij}(x^2, p\cdot x) = \epsilon(p\cdot x)\left[\theta(x^2)\mathfrak{C}_n^{ij}(x^2, p\cdot x) + \sum_{s=0}^{N} \delta^{(s)}(x^2)\,\mathfrak{C}_n'^{(s)ij}(p\cdot x)\right] \tag{3.6}$$

(but f_2^{ij} contains also a noncausal term; see Ref. 12).

In the Bjorken limit ($\nu \to \infty$, $q^2 \to -\infty$ with $\omega = -q^2/2\nu$ fixed) the behavior of the structure functions $\mathfrak{F}_n^{ij}$ is determined by the light-cone behavior[13] of $f_n^{ij}(x^2, p\cdot x)$ (see also Ref. 14). The scaling behavior in the Bjorken limit,[3]

$$\begin{aligned} & \lim_{\mathrm{Bj}} q^2 \mathfrak{F}_1(\nu, q^2) = F_1(\omega) \,, \\ & \lim_{\mathrm{Bj}} q^2 \nu \mathfrak{F}_2(\nu, q^2) = F_2(\omega) \,, \\ & \lim_{\mathrm{Bj}} q^2 \mathfrak{F}_3(\nu, q^2) = F_3(\omega) \,, \end{aligned} \tag{3.7}$$

corresponds to

$$f_1^{ij}(x^2,p\cdot x)=\epsilon(p\cdot x)[\theta(x^2)\mathfrak{C}_1^{ij}(x^2,p\cdot x)+\delta(x^2)\mathfrak{C}_1'^{ij}(p\cdot x)]\,,$$

$$f_2^{ij}(x^2,p\cdot x)=\epsilon(p\cdot x)\theta(x^2)\mathfrak{C}_2^{ij}(x^2,p\cdot x)+(\text{noncausal term})\,, \tag{3.8}$$

$$f_3^{ij}(x^2,p\cdot x)=\epsilon(p\cdot x)[\theta(x^2)\mathfrak{C}_3^{ij}(x^2,p\cdot x)+\delta(x^2)\mathfrak{C}_3'^{ij}(p\cdot x)]\,,$$

with $\mathfrak{C}_2^{ij}(0,p\cdot x)$ finite, and

$$F_s^{ij}(\omega)=i2\pi\omega\,\tilde{\mathfrak{C}}_1'^{ij}(\omega)\,,$$

$$F_2^{ij}(\omega)=-i4\pi\omega\frac{d\tilde{\mathfrak{C}}_2^{ij}(\omega)}{d\omega}\,, \tag{3.9}$$

$$F_3^{ij}(\omega)=i2\pi\omega\,\tilde{\mathfrak{C}}_3'^{ij}(\omega)\,,$$

where $(\sigma\equiv p\cdot x)$

$$\tilde{\mathfrak{C}}_{1,3}'^{ij}(\omega)=\frac{1}{2\pi}\int d\sigma e^{-i\omega\sigma}\mathfrak{C}_{1,3}'^{ij}(\sigma)\,,\quad \tilde{\mathfrak{C}}_2^{ij}(\omega)=\frac{1}{2\pi}\int d\sigma e^{-i\omega\sigma}\mathfrak{C}_2^{ij}(0,\sigma)\,. \tag{3.10}$$

Using (3.7) and the relations

$$\epsilon(x^0)\delta(x^2)\mathfrak{C}'(p\cdot x)|_{x^+=0}=\tfrac{1}{2}\pi\epsilon(x^-)\delta(\vec{x}_\perp)\mathfrak{C}'(Mx^-/\sqrt{2})\,,$$

$$\partial_{a_1}\cdots\partial_{a_n}\partial_-^m(\epsilon(x^0)\theta(x^2)\mathfrak{C}(x^2,p\cdot x))|_{x^+=0}=0\,, \tag{3.11}$$

$$\partial_a\partial_+(\epsilon(x^0)\theta(x^2)\mathfrak{C}(x^2,p\cdot x))|_{x^+=0}=\partial_a(\epsilon(x^-)x^-\delta(\vec{x}_\perp)\pi\mathfrak{C}(0,Mx^-/\sqrt{2})$$

(for the singular functions their value at $x^+=0$ was defined as the semisum of their limits for $x^+\to\pm0$) one finds on the null plane (with $\vec{p}=0$)

$$t_{--}^{ij}(x,p)=\partial_-^2[\epsilon(x)\delta(\vec{x}_\perp)\tfrac{1}{2}\pi\mathfrak{C}_1'^{ij}(Mx^-/\sqrt{2})]+if_{ijk}\langle p|j_k^+(0)|p\rangle\delta^3(x) \tag{3.12}$$

(the last term is due to the noncausal contribution to f_2^{ij}),

$$\begin{aligned}t_{-a}^{ij}(x,p)=&\,\partial_a\partial_-[\epsilon(x^-)\delta(\vec{x}_\perp)\tfrac{1}{2}\pi\mathfrak{C}_1'^{ij}(Mx^-/\sqrt{2})]-\partial_a[\epsilon(x^-)x^-\delta(\vec{x}_\perp)\tfrac{1}{2}\pi M^2\mathfrak{C}_2^{ij}(0,Mx^-/\sqrt{2})]\\&+\epsilon_{ab}\partial_b[\epsilon(x^-)\delta(\vec{x}_\perp)(\pi M/2\sqrt{2})\mathfrak{C}_3'^{ij}(Mx^-/\sqrt{2})]\,.\end{aligned} \tag{3.13}$$

Now we have all the necessary elements for the identification of our structure functions.

Defining the matrix element of our bilocal operator

$$\frac{m^2}{2g^2}\langle p|\Omega(x,0)_{ij}|p\rangle|_{x^+=0;\,x_\perp=0}=\Omega^{ij}\left(\frac{Mx^-}{\sqrt{2}}\right) \tag{3.14}$$

and the form factors

$$\langle p|j_k^\mu(0)|p'\rangle=(p^\mu+p'^\mu)F_k((p-p')^2) \tag{3.15}$$

[with the covariant normalization $\langle p|p'\rangle=(2\pi)^3 2p^0\times\delta(\vec{p}-\vec{p}')$] and taking into account that

$$\langle p|C(x,0)\mathfrak{U}^a(0)|p\rangle=0$$

when $\vec{p}=0$ and that $\langle p|X(x,0)|p\rangle$ must vanish because otherwise it does not obey the spectral condition [compare with (3.6)], we identify

$$\mathfrak{C}_1'^{ij}(\sigma)=\frac{i}{2\pi}\Omega^{ij}(\sigma)\,,$$

$$\mathfrak{C}_2^{ij}(0,\sigma)=\frac{i}{2\pi\sigma}\frac{d}{d\sigma}\Omega^{ij}(\sigma)\,, \tag{3.16}$$

$$\mathfrak{C}_3'^{ij}(\sigma)=0$$

or

$$F_s^{ij}(\omega)=-\omega\tilde{\Omega}^{ij}(\omega)\,,$$

$$F_2^{ij}(\omega)=2\omega^2\tilde{\Omega}^{ij}(\omega)$$

$$\left[\tilde{\Omega}^{ij}(\omega)=\frac{1}{2\pi}\int d\sigma e^{-i\omega\sigma}\Omega^{ij}(\sigma)\right],$$

$$F_3^{ij}(\omega)=0\,. \tag{3.17}$$

Thus, the relation

$$F_2^{ij}(\omega)=-2\omega F_s^{ij}(\omega) \tag{3.18}$$

emerges. The same relation was obtained by Callan and Gross[15] considering the infinite-momentum frame ($|\vec{p}| \to \infty$) matrix element of the equal-time dotted current commutator of the field algebra model.

Recalling that due to (2.23)

$$\frac{d}{d\sigma}\,\Omega^{ij}(\sigma)\Big|_{\sigma=0} = -f_{ijk}F_k$$

and (3.19)

$$\Omega^{ij}(0) = 0\ ,$$

we find immediately the Adler sum rule

$$\int_{-1}^{1} d\omega\,\frac{1}{\omega}\,F_2^{ij}(\omega) = 2if_{ijk}F_k$$

and (3.20)

$$\int_{-1}^{1} d\omega\,\frac{1}{\omega^2}\,F_2^{ij}(\omega) = 0\ .$$

Here we assumed that $\Omega^{ij}(\pm\infty)$ is finite. Then it results also that

$$F_2^{ij}(\omega) \xrightarrow[\omega\to 0]{} \frac{i}{2\pi}\,[\Omega^{ij}(\infty) - \Omega^{ij}(-\infty)]\ ,$$

which does not correspond to the Regge behavior.

To conclude, we have seen that the formal manipulation of the classical Poisson brackets enabled us to derive the Bjorken scaling behavior given by the field-algebra Lagrangian. The results agree with those obtained by formal manipulations of the equal-time commutators.

Note added. After the completion of this paper, we learned about the work of Feinberg,[16] where the same results are obtained by using Schwinger's quantum action principle. We would like to thank him for sending us his preprint and for interesting comments.

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[*] Work performed under the contract with the State Committee for Nuclear Energy of Romania.

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LETTERE AL NUOVO CIMENTO VOL. 11, N. 2 14 Settembre 1974

Again on the Gauge Dependence of Renormalization Group Parameters (*).

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(ricevuto il 17 Luglio 1974)

Non-Abelian gauge theories have recently attracted attention as candidates for a field theory of strong interactions (1), due to their unique asymptotically free behaviour (2). The infra-red divergences inherent to these theories raise serious problems as well as certain hopes (2) regarding the nonobservability of massless vector bosons and quarks. For the moment we can discuss only the theory of Green's functions subtracted at an arbitrary Euclidean point. Hence, observability cannot be used as an argument in discussing the dependence on the gauge parameter of the various quantities. A peculiar importance has the understanding of the gauge dependence of renormalization group parameters.

Recently (3) Caswell and Wilczek derived certain relations for the gauge parameter derivatives of the renormalization group parameters in a pure Yang-Mills theory, and noticed that they lead to gauge independence in the 't Hooft (4) renormalization. However, the nature of their consistency relations is not transparent, and their argument about the 't Hooft renormalization is neither clear, nor complete.

Due to the importance of the problem we present in the following a new discussion using 't Hooft's renormalization procedure in the very convenient version of Collins and Macfarlane (5). We show that the consistency relations including massive fermions are actually contained in the very definitions of the renormalization group parameters and their gauge independence in 't Hooft's renormalization is connected with the gauge independence of certain renormalization constants.

(*) Work performed under contract with the State Committee for Nuclear Energy of Romania.

(1) D. Gross and F. Wilczek: *Phys. Rev. D*, **8**, 3633 (1973); S. Weinberg: *Phys. Rev. Lett.*, **31**, 494 (1973).

(2) D. Gross and F. Wilczek: *Phys. Rev. Lett.*, **26**, 1343 (1973); H. Politzer: *Phys. Rev. Lett.*, **26**, 1346 (1973); G. 't Hooft: unpublished.

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(5) J. C. Collins and A. J. Macfarlane: University of Cambridge preprint (Nov. 1973).

Let us consider a renormalized, proper Green function Γ_R obtained through multiplicative renormalization of the unrenormalized Green function Γ_u

$$\Gamma_R(p, \mu, m, g, \alpha) = Z_\Gamma \Gamma_u(p, m_0, g_0, \alpha_0) . \tag{1}$$

Here m, g, α (respectively m_0, g_0, α_0) are the renormalized (respectively unrenormalized) fermion mass, coupling constant and gauge parameter; p stands for the external momenta and μ is an independent mass parameter defining in some way the subtraction point ('t Hooft's unit of mass (4,5)). We shall assume all the unrenormalized quantities regularized through the dimensional procedure of 'T HOOFT and VELTMAN (6). Then from

$$\left.\frac{\partial \Gamma_u}{\partial \mu}\right|_{m_0, g_0, \alpha_0 \text{ fixed}} = 0 ,$$

follows (5) the renormalization group equation

$$\left(\mu \frac{\partial}{\partial \mu} + \beta \frac{\partial}{\partial g} - \gamma_m m \frac{\partial}{\partial m} - \gamma_\Gamma + \delta \frac{\partial}{\partial \alpha}\right) \Gamma_R = 0 . \tag{2}$$

Moreover, if Γ_u is gauge invariant, *i.e.*

$$\frac{\partial \Gamma_u}{\partial \alpha_0} = 0 ,$$

then also (1,3)

$$\left(\frac{\partial}{\partial \alpha} + \varrho \frac{\partial}{\partial g} - \varrho_m m \frac{\partial}{\partial m} - \varrho_\Gamma\right) \Gamma_R = 0 , \tag{3}$$

where the various coefficients defined by

$$\beta = \frac{\partial g}{\partial \ln \mu} , \qquad \gamma_m = \frac{\partial \ln m}{\partial \ln \mu} , \qquad \gamma_\Gamma = \frac{\partial \ln Z_\Gamma}{\partial \ln \mu} , \qquad \delta = \frac{\partial \alpha}{\partial \ln \mu} , \qquad (g_0, \alpha_0, m_0 \text{ fixed}) , \tag{4}$$

$$\varrho = \frac{\partial g}{\partial \alpha_0} \Big/ \frac{\partial \alpha}{\partial \alpha_0} , \qquad \varrho_m = \frac{\partial \ln m}{\partial \alpha_0} \Big/ \frac{\partial \alpha}{\partial \alpha_0} , \qquad \varrho_\Gamma = \frac{\partial \ln Z_\Gamma}{\partial \alpha_0} \Big/ \frac{\partial \alpha}{\partial \alpha_0} , \qquad (g_0, m_0, \mu \text{ fixed}) , \tag{5}$$

as well as the renormalization constants are dimensionless and may be expressed as functions of g, α and m/μ.

Using the second equation one may eliminate the term from the renormalization group equation. This gives rise to a redefinition of the coefficients

$$\begin{cases} \bar{\beta} = \beta - \varrho \delta , \\ \bar{\gamma}_m = \gamma_m - \varrho_m \delta , \\ \bar{\gamma}_\Gamma = \gamma_\Gamma - \varrho_\Gamma \delta . \end{cases} \tag{6}$$

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(6) G. 'T HOOFT and M. VELTMAN: *Nucl. Phys.*, **44** B, 189 (1972).

Some of these definitions may be expressed directly in terms of derivatives with respect to renormalized parameters (here $Z_m = m_0/m$)

$$(7a)\qquad \varrho_m = \left(1 + \frac{\partial \ln Z_m}{\partial \ln (m/\mu)}\right)^{-1} \left(\frac{\partial}{\partial\alpha} + \varrho\frac{\partial}{\partial g}\right)\ln Z_m\,,$$

$$(7b)\qquad \varrho_\Gamma = \left(\frac{\partial}{\partial\alpha} + \varrho\frac{\partial}{\partial g} - \varrho_m\frac{\partial}{\partial \ln (m/\mu)}\right)\ln Z_\Gamma\,,$$

$$(8a)\qquad \bar{\gamma}_m = \left(1 + \frac{\partial \ln Z_m}{\partial \ln (m/\mu)}\right)^{-1}\left(\bar{\beta}\frac{\partial}{\partial g} - \frac{\partial}{\partial \ln (m/\mu)}\right)\ln Z_m\,,$$

$$(8b)\qquad \bar{\gamma}_\Gamma = \left[\bar{\beta}\frac{\partial}{\partial g} - (1+\bar{\gamma}_m)\frac{\partial}{\partial \ln (m/\mu)}\right]\ln Z_\Gamma\,.$$

Let us now consider 't Hooft's mass-independent renormalization procedure (4,5), where

$$(9)\qquad g\frac{Z_1}{Z_3^{\frac{3}{2}}} = g_0\mu^{-\varepsilon/2}\,,$$

$$(10)\qquad \alpha Z_3 = \alpha_0\,,$$

with $\varepsilon = 4 - n$, n being the space-time dimension.

All the renormalization group parameters $\beta, \gamma_m, \ldots$ are supposed to be analytical functions of ε. Then, by differentiating (9) with respect to $\ln\mu$ and α_0, we get

$$(11)\qquad \bar{\beta} = \left(\frac{\partial f}{\partial g}\right)^{-1}, \qquad \varrho = -\bar{\beta}\frac{\partial f}{\partial\alpha}\,,$$

where

$$f(g, \alpha, \varepsilon) = -\frac{2}{\varepsilon}\ln g\frac{Z_1}{Z_3^{\frac{3}{2}}}\,.$$

Now, by using the ε-expansion of the mass-independent renormalization constants

$$(12)\qquad \frac{Z_1}{Z_3^{\frac{3}{2}}} = 1 + \sum_{\nu=1}^{\infty}\frac{1}{\varepsilon^\nu}a_\nu(g, \alpha)\,,$$

we obtain

$$(13)\qquad \bar{\beta}(\varepsilon) = -\frac{\varepsilon}{2}g\frac{1 + \sum\limits_{\nu=1}^{\infty}(1/\varepsilon^\nu)a_\nu}{1 + \sum\limits_{\nu=1}^{\infty}(1/\varepsilon^\nu)\partial g a_\nu/\partial g}\,,$$

$$(14)\qquad \varrho(\varepsilon) = \frac{2}{\varepsilon}\bar{\beta}(\varepsilon)\frac{\sum\limits_{\nu=1}^{\infty}(1/\varepsilon^\nu)\partial a_\nu/\partial\alpha}{1 + \sum\limits_{\nu=1}^{\infty}(1/\varepsilon^\nu)a_\nu}\,.$$

From (13) it results that $\bar{\beta}(t)$ behaves like ε at $\varepsilon \to \infty$, hence it is an entire function

$$(15) \qquad \bar{\beta}(\varepsilon) = -\frac{\varepsilon}{2} g + \bar{\beta}\,,$$

$$(15') \qquad \bar{\beta} \;= \frac{1}{2} g^2 \frac{\partial a_1}{\partial g}\,.$$

For cosistency certain relations between the coefficients a_ν have to be satisfied (5).

By combining eqs. (14) and (15) we see that $\varrho(\varepsilon)$ vanishes at $\varepsilon \to \infty$, hence it is zero everywhere. The consistency conditions here imply that $\partial a_\nu/\partial\alpha = 0$, *i.e.* $Z_1/Z_3^{\frac{3}{2}}$ is independent of α. (This is readily seen in the one-loop calculation, see for example ref. (7).) Similar arguments give

$$(16) \qquad \bar{\gamma}_m = -\frac{1}{2} g \frac{\partial a_{m1}}{\partial g}\,, \qquad \bar{\gamma}_\Gamma = -\frac{1}{2} g \frac{\partial a_{\Gamma 1}}{\partial g}\,,$$

$$(17) \qquad \varrho_m = \varrho_\Gamma = 0\,.$$

The last relation implies that neither Z_m nor Z_Γ depend on α (8).

Thus in this renormalization a gauge-independent unrenormalized Green function produces an α-independent renormalized Green function if multiplicatively renormalizable. According to (6)

$$\bar{\beta} = \beta\,, \quad \bar{\gamma}_m = \gamma_m\,, \quad \bar{\gamma}_\Gamma = \gamma_\Gamma$$

and since

$$\partial a_1/\partial\alpha = \partial a_{m1}/\partial\alpha = \partial a_{\Gamma 1}/\partial\alpha = 0\,,$$

they are α-independent.

In the Abelian case $Z_1/Z_3^{\frac{3}{2}}$ is to be replaced by $Z_3^{-\frac{1}{2}}$ and thus Z_3 and Z_m are gauge independent, a well-known result in the usual renormalization of the QED.

Changing the definition of the renormalized coupling constant, gauge parameter and fermion mass, we can pass to an arbitrary renormalization procedure, which is equivalent to a finite intermediate renormalization. Replacing g by $\vartheta(g, \alpha, m/\mu)$, α

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(7) P. S. Collecott: University of Cambridge preprint (Feb. 1974).

(8) The analiticity of the renormalization group parameters at $\varepsilon = 0$ to every order in g is a direct consequence of the corresponding analiticity of the renormalized Green functions. The resulting consistency relations between the coefficients a_ν, a_{m_ν}, a_{Γ_ν} give rise to

$$\frac{Z_1}{Z_3^{\frac{3}{2}}} = \exp\left[-\int\limits_0^g \frac{\mathrm{d}g'}{g'} \frac{\bar{\beta}(g')}{\bar{\beta}(g') - (\varepsilon/2)g'}\right], \qquad Z_{m,\Gamma} = \exp\left[-\int\limits_0^g \mathrm{d}g' \frac{\bar{\gamma}_{m,\Gamma}(g')}{\bar{\beta}(g') - (\varepsilon/2)g'}\right].$$

Taking into account the known (7) lowest-order expressions for $\bar{\beta}(g)$, $\bar{\gamma}_m(g)$, we find

$$\frac{Z_1}{Z_3^{\frac{3}{2}}} \underset{\varepsilon \to 0}{\simeq} \varepsilon^{\frac{1}{2}}\,, \qquad Z_m \underset{\varepsilon \to 0}{\simeq} \varepsilon^{9C_2(R)/[11C_2(G)-4T(R)]}\,.$$

It is interesting to observe that only for asymptotically free theories $(11C_2(G) - 4T(R) > 0)$ $Z_m \xrightarrow[\varepsilon \to 0]{} 0$, thus the bare-fermion mass vanishes. Similar results in the Landau gauge ($\alpha = 0$) can be derived for the other renormalization constants. Such relations in a momentum cut-off version have been found also in ref. (9).

(9) Ng Wing-Chiu and K. Young: Stony Brook preprint (Nov. 1973).

by $\hat{\alpha}(g, \alpha, m/\mu)$ and m by $m\psi(g, \alpha, m/\mu)$, we find

$$(18)\qquad \left|\begin{aligned} &\varrho = \frac{\partial\hat{g}}{\partial\alpha}\bigg/\frac{\partial\hat{\alpha}}{\partial\alpha}\,,\\ &\bar{\beta} = \left(\frac{\partial\hat{g}}{\partial g} - \varrho\,\frac{\partial\hat{\alpha}}{\partial g}\right)\beta^{\mathrm{H}} - (1+\gamma_m^{\mathrm{H}})\left(\frac{\partial\hat{g}}{\partial\ln(m/\mu)} - \varrho\,\frac{\partial\hat{\alpha}}{\partial\ln(m/\mu)}\right),\end{aligned}\right.$$

where β^{H}, γ_m^{H} are the values of these parameters in the 't Hooft renormalization (g, α, m here are also the renormalized parameters of the same procedure). Analogous relations express the other coefficients through their values in the 't Hooft renormalization.

Taking into account that

$$\varrho^{\mathrm{H}} = \varrho_m^{\mathrm{H}} = \varrho_\Gamma^{\mathrm{H}} = 0$$

and

$$\frac{\partial\bar{\beta}^{\mathrm{H}}}{\partial\alpha} = \frac{\partial\bar{\gamma}_m^{\mathrm{H}}}{\partial\alpha} = \frac{\partial\bar{\gamma}_\Gamma^{\mathrm{H}}}{\partial\alpha} = 0\,,$$

one then finds generally

$$(19)\qquad \left\{\begin{aligned} \frac{\partial\bar{\beta}}{\partial\alpha} &= \left[\bar{\beta}\,\frac{\partial}{\partial g} - (1+\bar{\gamma}_m)\,\frac{\partial}{\partial\ln(m/\mu)}\right]\varrho - \left(\varrho\,\frac{\partial}{\partial\varrho} - \varrho_m\,\frac{\partial}{\partial\ln(m/\mu)}\right)\bar{\beta}\,,\\ \frac{\partial\bar{\gamma}_m}{\partial\alpha} &= \left[\bar{\beta}\,\frac{\partial}{\partial g} - (1+\bar{\gamma}_m)\,\frac{\partial}{\partial\ln(m/\mu)}\right]\varrho_m - \left(\varrho\,\frac{\partial}{\partial g} - \varrho_m\,\frac{\partial}{\partial\ln(m/\mu)}\right)\bar{\gamma}_m\,,\\ \frac{\partial\bar{\gamma}_\Gamma}{\partial\alpha} &= \left[\bar{\beta}\,\frac{\partial}{\partial g} - (1+\bar{\gamma}_m)\,\frac{\partial}{\partial\ln(m/\mu)}\right]\varrho_\Gamma - \left(\varrho\,\frac{\partial}{\partial g} - \varrho_m\,\frac{\partial}{\partial\ln(m/\mu)}\right)\bar{\gamma}_\Gamma\,.\end{aligned}\right.$$

(Here g, α, m are the renormalized parameters of the given renormalization procedure.)

These equations are the generalization of the consistency relations of CASWELL and WILCZEK in the presence of massive fermions, and may be obtained also though their method, which consists in forbidding any more nontrivial equation to be satisfied by the Green functions.

It appears that even with fermions, if $\varrho = \varrho_m = \varrho_\Gamma = 0$, the normalization group parameters $\bar{\beta}, \bar{\gamma}_m, \bar{\gamma}_\Gamma$ remain gauge independent. This occurs in any renormalization obtainable from the 't Hooft one through a transformation with $\hat{g}$ and ψ independent of α.

Nuclear Physics B93 (1975) 355–364

ON 't HOOFT'S RENORMALIZATION PROCEDURE IN GAUGE THEORIES *

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Received 17 March 1975

Properties of renormalization constants (gauge dependence and the $\epsilon \to 0$ limit) are shown in 't Hooft's renormalization of Yang-Mills theories. The equivalence to a standard subtraction procedure is discussed at the one-loop level.

1. Introduction

Recently there have been proposed [1, 2] new methods of renormalization in quantum field theory which do not consist in subtractions of the divergent proper irreducible vertices at a given momentum. The main advantage of these new approaches is that the renormalization constants (including the fermion mass renormalization constant Z_m) do not depend on the mass [3]. Consequently, the renormalization group equations may be solved exactly at arbitrary momenta and asymptotic freedom in non-Abelian gauge theories was shown rigorously. Moreover, some of the renormalization constants and the related renormalization group coefficients are shown to be gauge independent [4, 5].

One purpose of this paper is to give a more detailed account of the remarkable properties of the renormalization constants and the renormalization group parameters in 't Hooft's scheme for a non-Abelian gauge theory. On the other hand we discuss, at least at the one-loop level, the eventual equivalence of this renormalization with a subtraction at an unconventional, mass and gauge dependent momentum. Conclusions are given at the end of each section.

We shall consider a gauge field theory, whose Feynman rules are defined by the Lagrangian.

$$L = -\tfrac{1}{4} F^a_{\mu\nu} F^{\mu\nu}_a + \bar{\psi}\,(i\gamma_\mu D^\mu - m_0)\,\psi - \frac{1}{2\alpha_0}\,\partial_\mu A^\mu_a\,\partial^\nu A^a_\nu - \bar{u}_a \partial^\mu \nabla^{ab}_\mu u_b\,, \tag{1.1}$$

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* Work performed under contract with the State Committee for Nuclear Energy of Romania.

where:

$$F^a_{\mu\nu} \equiv \partial_\mu A^a_\mu - \partial_\nu A^a_\mu + g_0 f_{abc} A^b_\mu A^c_\nu ,$$

$$D_\mu \equiv \partial_\mu - ig_0 t^a A^a_\mu , \qquad [t^a, t^b] = i f_{abc} t^c ,$$

$$\nabla^{ab}_\mu \equiv \delta_{ab} \partial_\mu - g_0 f_{abc} A^c_\mu ,$$

A^a_μ, and u_a being the unrenormalized Yang-Mills, fermion and Faddeev-Popov ghost fields respectively.

The theory is supposed to be gauge invariantly regularized through the dimensional regularization of 't Hooft and Veltman [6]. The divergences of the theory, when the dimension n of the space-time tends to 4, manifest themselves as poles at the origin (at most ν -uple at the ν-loop level) in the variable $\epsilon \equiv 4 - n$. 't Hooft's procedure consists in just dropping these poles, i.e. a multiplicative renormalization with dimensionsless renormalization constants of the form

$$Z_i = 1 + \sum_{\nu=1}^{\infty} \frac{1}{\epsilon^\nu} a_i^{(\nu)} , \tag{1.2}$$

where $a_i^{(\nu)}$ gets contribution starting with the ν-loop diagrams. The important feature is that $a_i^{(\nu)}$ does not depend on the renormalized mass [3]. The series in eq. (1.2) is supposed to converge in a domain of the ϵ-plane so as to define a unique $Z_i(\epsilon)$. Otherwise it may be understood as an asymptotic series for $\epsilon \to \infty$.

The renormalized coupling constant g, mass m and gauge parameter α are related to the unrenormalized ones through

$$g_0 \mu^{-\epsilon/2} = \frac{Z_1}{Z_3^{\frac{3}{2}}} g, \qquad m_0 = Z_m m, \qquad \alpha_0 = Z_3 \alpha . \tag{1.3}$$

Z_1 is the three-vector vertex renormalization constant, Z_3 is the vector field renormalization constant, while Z_m is related to the conventional fermion wave function and mass renormalization constants Z_2 and δm, through

$$Z_m = 1 + (\delta m/m) Z_2^{-1} ,$$

('tHooft's unit of mass μ was introduced everywhere to get the correct dimensions [1, 3].

The renormalized group equation à la Weinberg [2] is

$$\left(\mu \frac{\partial}{\partial\mu} + \beta \frac{\partial}{\partial g} - \gamma_m m \frac{\partial}{\partial m} - \gamma_\Gamma + \delta \frac{\partial}{\partial\alpha}\right) \Gamma_R(p,g,\alpha,m,\mu) = 0 , \tag{1.4}$$

where

$$\beta \equiv \frac{\partial g}{\partial \ln \mu} , \qquad \gamma_m \equiv \frac{\partial \ln m}{\partial \ln \mu} , \qquad \gamma_\Gamma \equiv \frac{\partial \ln Z_\Gamma}{\partial \ln \mu} , \qquad \delta \equiv \frac{\partial \alpha}{\partial \ln \mu} , \tag{1.5}$$

at fixed g_0, α_0 and m_0; obviously $\delta = -\alpha \gamma_3$. These coefficients have to be finite at $\epsilon = 0$ in every order in g. The same is true for the coefficients

$$\rho \equiv \frac{\partial g}{\partial \alpha_0} \bigg/ \frac{\partial \alpha}{\partial \alpha_0} , \qquad \rho_m \equiv \frac{\partial \ln m}{\partial \alpha_0} \bigg/ \frac{\partial \alpha}{\partial \alpha_0} , \qquad \rho_\Gamma \equiv \frac{\partial \ln Z_\Gamma}{\partial \alpha_0} \bigg/ \frac{\partial \alpha}{\partial \alpha_0} , \tag{1.6}$$

(at fixed g_0, m_0 and μ) of another equation [7]

$$\left(\frac{\partial}{\partial \alpha} + \rho \frac{\partial}{\partial g} - \rho_m \, m \frac{\partial}{\partial m} - \rho_\Gamma \right) \Gamma_R(p, g, \alpha, m, \mu) = 0 \tag{1.7}$$

to be satisfied by the Green functions obtained through multiplicative renormalization of gauge invariant unrenormalized Green functions. As we shall see later, the finiteness of these coefficients has far reaching consequences.

The solution of eq. (1.4), having a canonical dimension D, behaves like

$$\begin{aligned} &\Gamma_R(\lambda p, g, \alpha, m, \mu) \\ &\quad = \lambda^D \Gamma_R(p, g(t), m(t), \mu) \exp \left[- \int_0^{\ln \lambda} \mathrm{d}t' \, \gamma_\Gamma \left(g(t'), \frac{m(t')}{\mu} \right) \right] \end{aligned} \tag{1.8}$$

under the scaling of momenta. Here $t \equiv \ln \lambda$ and

$$\begin{aligned} &\frac{\mathrm{d}g(t)}{\mathrm{d}t} = \beta \left(g(t), \alpha(t), \frac{m(t)}{\mu} \right) , \qquad && g(0) = g , \\ &\frac{\mathrm{d}\alpha(t)}{\mathrm{d}t} = \delta \left(g(t), \alpha(t), \frac{m(t)}{\mu} \right) , \qquad && \alpha(0) = \alpha , \\ &\frac{\mathrm{d} \ln m(t)}{\mathrm{d}t} = -1 - \gamma_m \left(g(t), \frac{m(t)}{\mu} \right) , \qquad && m(0) = m . \end{aligned} \tag{1.9}$$

In 't Hooft's renormalization these equations are decoupled since β, δ and γ_m do not depend on m/μ and we shall see [4, 5] that β and γ_m and γ_m are gauge independent.

In the one-loop approximation the values of the various coefficient are

$$\begin{aligned} &\beta(g) = - \frac{g^3}{(4\pi)^2} \left[\tfrac{11}{3} \, C_2(G) - \tfrac{4}{3} T(R) \right] , \\ &\gamma_m(g) = \frac{g^2}{(4\pi)^2} \, 6 \, C_2(R) , \\ &\gamma_2(g, \alpha) = \frac{g^2}{(4\pi)^2} \, 2\alpha \, C_2(R) , \end{aligned} \tag{1.10}$$

$$\gamma_3(g,\alpha) = -\frac{g^2}{(4\pi)^2}\left[\frac{13-3\alpha}{3}\,C_2(G) - \tfrac{8}{3}\,T(R)\right],$$

$$\tilde{\gamma}_3(g,\alpha) = -\frac{g^2}{(4\pi)^2}\,\frac{3-\alpha}{2}\,C_2(G)\,,$$

where the values of the Casimir operator are $C_2(G)$ and $C_2(R)$ for the adjoint representation and the representation R of the gauge group G respectively, while $T(R)$ is defined by Tr $t^a\, t^b \equiv \delta_{ab}\, T(R)$. If the quantity $b \equiv \frac{11}{3}\, C_2(G) - \frac{4}{3}\, T(R)$ is positive, the origin is a ultraviolet attraction point for the first eq. (1.9) and one has "asymptotic freedom" [7, 8]:

$$g^2(t) \underset{t\to\infty}{\sim} \frac{(4\pi)^2}{2bt}\,. \tag{1.11}$$

For $t \to \infty$ it follows also

$$\frac{\mathrm{d}\ln m\,(t)}{\mathrm{d}t} \underset{t\to\infty}{\sim} -1\,,$$

and thus $m(\infty) = 0$. From the third eq. (1.9) we have for $t \to \infty$

$$\frac{\mathrm{d}\alpha(t)}{\mathrm{d}t} \underset{t\to\infty}{\sim} \frac{A}{t}\,\alpha(t)[\bar{\alpha} - \alpha(t)],$$

with $\bar{\alpha} \equiv \frac{13}{3} - \frac{8}{3}\, T(R)/C_2(G)$ and $A > 0$. Then 0 and $\bar{\alpha}$ are the fixed points. Which of them is ultravioleted stable depends on the sign of $\bar{\alpha}$: $\alpha(\infty) = \bar{\alpha}\,\theta(\bar{\alpha})$. However there is a special case $\alpha(0) = 0$, which implies $\alpha(t) = 0$ for all t. From $-3 < \bar{\alpha} < \frac{13}{3}$ it follows that $0 \leqslant \alpha(\infty) < \frac{13}{3}$ [9].

Therefore the Green function in the right-hand side of eq. (1.8) approaches asymptotically its free value for massless fermions in the gauge $\alpha(\infty)$.

As it has been shown by Weinberg [2] one can get in this way the first three terms in the asymptotic expansion of Green functions at arbitrary large momenta. Since different renormalization procedures are related by finite multiplicative renormalizations of the coupling constant, mass and gauge parameter, approaching the identity for $g \to 0$, the above results are valid in an arbitrary renormalization.

Unfortunately, the infrared (low momentum) limit of the Green functions cannot be understood through a similar analysis.

2. Renormalization constants and renormalization group parameters

Let us consider the ϵ-expansion of $Z \equiv Z_1/Z_3^{\frac{3}{2}}$:

$$Z = 1 + \sum_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}}\, a_{\nu}\,, \tag{2.1}$$

whose coefficients a_ν are functions of g and α. Then just from the definitions (1.5), (1.7) and (1.3) one has [5].

$$\bar{\beta}(\epsilon, g, \alpha) = -\tfrac{1}{2}\epsilon g \, \frac{1 + \sum\limits_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}}\, a_{\nu}}{1 + \sum\limits_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}} \frac{\partial}{\partial g}(g\, a_{\nu})}\,, \tag{2.2}$$

where $\bar{\beta} \equiv \beta - \rho\delta$ and

$$\rho(\epsilon, g, \alpha) = \frac{2}{\epsilon}\, \bar{\beta}(\epsilon, g, \alpha)\, \frac{\sum\limits_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}} \frac{\partial a_{\nu}}{\partial \alpha}}{1 + \sum\limits_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}}\, a_{\nu}}\,. \tag{2.3}$$

Since the ratio (and the product) of two asymptotic series is again an asymptotic series (or, the ratio of two Taylor series in $1/\epsilon$ is again a Taylor series in $1/\epsilon$), we have

$$\bar{\beta}(\epsilon, g, \alpha) = -\tfrac{1}{2}\epsilon g + \tfrac{1}{2} g^2 \frac{\partial a_1}{\partial g} + \sum_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}}\, b_{\nu}\,,$$

and

$$\rho(\epsilon, g, \alpha) = \sum_{\nu=1}^{\infty} \frac{1}{\epsilon^{\nu}}\, C_{\nu}\,.$$

Now, the important point, is that in every finite order in g one can see that there are a finite number of b_ν's and c_ν's different from zero. Since β, ρ and δ have to be finite at $\epsilon = 0$, in every order in g, the coefficients b_ν and c_ν have to vanish in every order. The vanishing of all the c_ν's implies the vanishing of ρ itself. Therefore $\bar{\beta} = \beta$ and $\partial a_\nu / \partial \alpha = 0$, thus

$$\bar{\beta}(\epsilon, g) = -\tfrac{1}{2} g + \beta(g)\,, \tag{2.4}$$

with $\beta(g) = \frac{1}{2} g^2\, \partial a_1 / \partial g$, being independent of α.

On the other hand, eq. (2.2) was nothing else than

$$\bar{\beta}(\epsilon, g) = -\tfrac{1}{2}\epsilon \left[\frac{\mathrm{d}}{\mathrm{d}g} \ln g\, Z(\epsilon, g)\right]^{-1}, \tag{2.5}$$

from where

$$Z(\epsilon, g) = \exp\left[-\int_0^g \frac{\mathrm{d}g'}{g'} \frac{\beta(g')}{\beta(g') - \frac{1}{2}\epsilon g'}\right] . \tag{2.6}$$

In the same way, one can easily show that $\rho_m = \rho_\Gamma = 0$, thus neither Z_m (and γ_m) nor Z_Γ (and γ) depend on α. Moreover, for an arbitrary γ_i one has

$$\gamma_i(g,\alpha) \equiv \left.\frac{\partial \ln Z_i}{\partial \ln \mu}\right|_{g_0,\alpha_0,m_0} = \beta \frac{\partial \ln Z_i}{\partial g} + \delta \frac{\partial \ln Z_i}{\partial \alpha} ,$$

thus

$$\gamma_i(\epsilon, g(t), \alpha(t)) = \frac{\mathrm{d}}{\mathrm{d}t} \ln Z_i(\epsilon, g(t), \alpha(t)) .$$

Since $g(t)$ is a monotonical function of t, we may define a function $\tilde{\alpha}(g)$ such that $\tilde{\alpha}(g(t)) \equiv \alpha(t)$. Then

$$\gamma_i(\epsilon, g, \tilde{\alpha}(g)) = \bar{\beta}(\epsilon, g) \frac{\mathrm{d}}{\mathrm{d}g} \ln Z_i(\epsilon, g, \tilde{\alpha}(g)) ,$$

from where (In fact γ_i cannot depend on ϵ due to its finiteness at $\epsilon = 0$)*

$$Z_i(\epsilon, g, \alpha) = \exp\left[\int_0^g \mathrm{d}g' \frac{\gamma_i(g', \tilde{\alpha}(g'))}{\beta(g') - \frac{1}{2}\epsilon g'}\right] . \tag{2.7}$$

As one can see from eqs. (2.6) and (2,7), the renormalization constants have a finite cut in the ϵ-plane.

Now we look for the behavior of the renormalization constants as ϵ tends to zero. The integrals in the exponent of eqs. (2.6) and (2.7) diverge at the integration end $g = 0$ as $\epsilon \to 0$. This divergent part can be thus extracted knowing only the lowest approximations for the renormalization group parameters. In this way we obtain

$$Z \underset{\epsilon\to 0}{\widetilde{\ \ }} \epsilon^{\frac{1}{2}} , \qquad Z_i \underset{\epsilon\to 0}{\widetilde{\ \ }} \epsilon^{C_i(\tilde{\alpha}(0))/2b} , \tag{2.8}$$

with c_i defined through

$$\gamma_i(g, \alpha) = (g^2/(4\pi)^2)\, C_i(\alpha) + O(g^4) .$$

Using their actual values from (1.10), we have (with $\tilde{\alpha}(0) = \alpha(\infty)$ in the case of "asymptotic freedom")

$$Z_m \underset{\epsilon\to 0}{\widetilde{\ \ }} \epsilon^{3C_2(R)/b} , \qquad Z_3 \underset{\epsilon\to 0}{\widetilde{\ \ }} \epsilon^{(\tilde{\alpha}[\theta(\tilde{\alpha})-1]/2b)\, C_2(G)} ,$$

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* There is a wrong sign for the integral at the exponent of Z_m as given in ref. [5].

$$Z_2 \underset{\epsilon\to 0}{\widetilde{\quad}} \epsilon^{(\bar{\alpha}[\theta(\bar{\alpha})-1]/2b)}\, C\,, \qquad \widetilde{Z}_3 \underset{\epsilon\to 0}{\widetilde{\quad}} \epsilon^{((\bar{\alpha}\,\theta(\bar{\alpha})-3)/4b)C_2(G)}\,. \qquad (2.9)$$

Thus Z_1 and Z_m always tend to zero (and so do the ratios Z_1/Z_3 and $Z_1/Z_3^{\frac{3}{2}}$) while Z_2 and Z_3 tend to zero or to a constant, depending on the sign of $\bar{\alpha}$. In either case there is no gauge dependence of the $\epsilon \to 0$ limit, except the actual value of the finite constants. Of course, as it was mentioned before, the transverse gauge is to be treated separately since for it $\widetilde{\alpha}(0) = \alpha(\infty)$ is always zero. Therefore, besides Z and Z_m, which are gauge independent, we have in this gauge

$$Z_2 \underset{\epsilon\to 0}{\widetilde{\quad}} \text{const}\,,$$

$$Z_3 \underset{\epsilon\to 0}{\widetilde{\quad}} \epsilon^{-(\bar{\alpha}/2b)C_2(G)}\,,$$

$$\widetilde{Z}_3 \underset{\epsilon\to 0}{\widetilde{\quad}} \epsilon^{-(3/4b)C_2(G)}\,,$$

so that Z_1/Z_3 vanishes again.

Hence, this is an example of a quantum field theory where one can explicitely prove the finiteness of some of the renormalization constants, although they diverge in each finite order of perturbation theory. Strange enough in the same time, the bare coupling constant g_0 and the bare fermion mass m_0 are vanishing. This is valid also in the Abelian case (QED). However in what concerns the gauge dependent renormalization constants, "asymptotic freedom" was a necessary ingredient for obtaining the result.

Since due to infrared problems no on-shell S-matrix can be defined in either case, the physical interpretation of the above result is lacking *.

All our discussion refers to 'tHooft's renormalization procedure. However, the results on the $\epsilon \to 0$ limit of the renormalization constants are generally valid since a finite renormalization does not change anything.

Essentially, the content of this chapter is a more detailed account of results obtained in a previous work [5]. Similar results were obtained also in [11] in the frame of the Gell-Mann-Low renormalization scheme with ultraviolet cut-off. Whereas the results are correct their derivation is not satisfactory since they had to assume, as in the old renormalization group approach, that the m/μ and α dependence of β can be ignored.

Another class of renormalizations that give rise to gauge independence (at least for massless fermions) was proposed in [12]. There, the imposed normalization of the vertex part is chosen α dependent so as to give a gauge independent β.

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* See however ref. [10].

3. The equivalent subtraction point

In this section we put ourselves the question whether 'tHooft's renormalization procedure (where the mass parameter μ is just a unit of mass) is equivalent to a subtraction a la Gell-Mann-Low in a specially prescribed Euclidian point, i.e. dependent on m/μ, α and g.

Let us consider for example the renormalized meson self-energy part $\Pi(p^2/\mu^2, \alpha, m/\mu, g)$. The problem is then to find order by order in g the position of the zeroes for $p^2 < 0$. With the perturbative expansion of Π.

$$\Pi\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}, g\right) = \Pi_1\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) g^2 + \Pi_2\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) g^4 + \dots ,$$

and of its zero

$$\lambda^2 = \lambda^2_{(0)} + \lambda^2_{(1)} g^2 + \dots , \qquad \Pi\left(-\lambda^2, \alpha, \frac{m}{\mu}, g\right) = 0 ,$$

we have the following set of equations to determine successively the $\lambda^2_{(\nu)}$'s:

$$\Pi_1\left(-\lambda^2_{(0)}, \alpha, \frac{m}{\mu}\right) = 0 ,$$

$$\Pi_2\left(-\lambda^2_{(0)}, \alpha, \frac{m}{\mu}\right) + \lambda^2_{(1)} \Pi_1\left(-\lambda^2_{(0)}, \alpha, \frac{m}{\mu}\right) = 0 ,$$

....

Of course, there would remain serious convergence problems of this procedure.

We have studied the first step of this programme for the selfenergies of the vector meson, ghost and fermion fields. For that purpose we have computed in the one-loop approximation the finite part of the self-energy (which are ignored in renormalization group problems).

The renormalized vector meson self-energy Π, defined through the total vector meson propagator

$$D_{\mu\nu}(p) = \frac{1}{p^2[1 - \Pi(p^2)]} \left(g_{\mu\nu} - \frac{p_\mu g_\nu}{p^2}\right) + \alpha \frac{p_\mu p_\nu}{p^4} ,$$

is given in the one-loop approximation by

$$\begin{aligned} &\Pi\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) \\ &= \frac{g^2}{(4\pi)^2} \left\{ C_2(G) \left[\frac{3\alpha - 13}{6} \left(\ln\left(-\pi \frac{p^2}{\mu^2}\right) + \gamma \right) + \tfrac{22}{9} + \tfrac{1}{4}(\alpha + 1)^2 \right] \right. \\ &\left. + \tfrac{4}{3} T(R) \left[\ln\left(\pi \frac{m^2}{\mu^2}\right) + \gamma + 6 \int_0^1 \mathrm{d}\xi\, \xi(1 - \xi) \ln\left(1 - \frac{p^2}{m^2}\, \xi(1 - \xi)\right) \right] \right\} . \qquad (3.1) \end{aligned}$$

The renormalized ghost self-energy Σ_u, defined through the total ghost propagator

$$\Delta(p) = \frac{1}{p^2[1 + \Sigma_u(p^2)]}$$

is given in the one-loop approximation by

$$\Sigma_u\left(\frac{p^2}{\mu^2}, \alpha\right) = \frac{g^2}{(4\pi)^2}\, C_2(G)\left[\frac{\alpha - 3}{4}\, \left(\ln\left(-\pi \frac{p^2}{\mu^2}\right) + \gamma\right) + 1\right]. \tag{3.2}$$

The renormalized fermion self-energy Σ, defined through the total propagator of the fermion field

$$S(p) = \frac{1}{\gamma \cdot p - m - \Sigma(p)}$$

is given in the one-loop approximation by

$$\Sigma(p) = \gamma \cdot p\, a\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) + m\, b\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right),$$

where

$$\begin{aligned} a&\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) \\ &= \frac{g^2}{(4\pi)^2} C_2(R)\, \alpha \left[\ln\left(\pi \frac{m^2}{\mu^2}\right) + \gamma - 1 - \frac{m^2}{p^2} + \left(1 - \frac{m^4}{p^4}\right) \ln\left(1 - \frac{p^2}{m^2}\right)\right], \\ b&\left(\frac{p^2}{\mu^2}, \alpha, \frac{m}{\mu}\right) \\ &= \frac{g^2}{(4\pi)^2}\, C_2(R)\, (\alpha + 3) \left[\ln\left(\pi \frac{m^2}{\mu^2}\right) + \gamma - \frac{2(\alpha + 2)}{\alpha + 3} + \left(1 - \frac{m^2}{p^2}\right) \ln\left(1 - \frac{p^2}{m^2}\right)\right]. \end{aligned} \tag{3.3}$$

(In the above equations γ is Euler's constant $\gamma = 0.5772...$).

First, we consider the vanishing of the meson self-energy (3.1). For simplicity, let us take at the beginning a theory without fermions. Here there is a simple solution

$$\lambda^2_{(0)} = \frac{1}{\pi} \exp\left[\frac{\frac{44}{3} + \frac{3}{2}(\alpha + 1)^2}{13 - 3\alpha} - \gamma\right]$$

excepting the gauge $\alpha = \frac{13}{3}$, where there is no solution.

In the presence of fermions ($T(R) \neq 0$) no explicit solution can be found, but an analysis of (3.1) shows that for $\alpha < \bar{\alpha}$ and $\alpha > \frac{13}{3}$ the self-energy is a monotonical function of $-p^2/\mu^2$, diverging at the origin and the infinitey, while for $\overline{\alpha} < \alpha < \frac{13}{3}$ it has an absolute minimum (whose value depends on the ratio m/μ). Therefore, in the gauges $\alpha < \overline{\alpha}$ and $\alpha > \frac{13}{3}$ one has always a unique solution, but in the gauges $\overline{\alpha} < \alpha < \frac{13}{3}$ one may have a solution only for a given value of the ratio m/μ (above that value there is no solution at all and below there are more solutions).

Thus, at least in a class of gauges, characterized by $\overline{\alpha} < \alpha < \frac{13}{3}$, 't Hooft's renormalization is not equivalent to a subtraction of the meson self-energy for an arbitrary value of the mass parameter μ.

The ghost self-energy (3.2) has always a zero at ($\alpha \neq 3$)

$$\lambda^2_{(0)} = \frac{1}{\pi} \exp\left(\frac{4}{3-\alpha} - \gamma\right).$$

In what concerns the fermion self-energy (3.3), the invariant functions a and b do not vanish for any $p^2 < m^2$ if m/μ is sufficiently big.

To conclude, our one-loop level analysis shows that 't Hooft's renormalization scheme is not equivalent, generally speaking, to a subtraction at a special point.

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Reprints

Solid state and statistical physics

phys. stat. sol. 3, 2299 (1963)

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On the Kinetic Theory of Magneto-Optical Phenomena by Green Function Method

By

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General formulae for the frequency-dependent transverse electrical conductivity in a d.c. magnetic field are obtained, using the double-time temperature dependent Green's function method. The results obtained are applied to cyclotron resonance in high magnetic fields.

Es werden allgemeine Beziehungen für die frequenzabhängige transversale elektrische Leitfähigkeit in magnetischen Gleichfeldern unter Verwendung der temperaturabhängigen Greenschen Funktion erhalten. Die Ergebnisse werden auf die Untersuchung der Zyklotronenresonanz in hohen Magnetfeldern angewandt.

It is well known that the conductivity tensor may be related to the correlations of the equilibrium currents [1]. On the other hand, the correlation functions may be expressed in terms of the double-time temperature dependent Green functions, defined as

$$G_r(t-t') \equiv \langle\langle A(t);\ B(t')\rangle\rangle_r = -\,i\,\theta(t-t')\,\langle[A(t),\ B(t')]\rangle\,, \tag{1}$$

where $A(t)$ is an operator in the Heisenberg picture, $\theta(t)$ is the step function, and averaging is performed over the grand canonical ensemble.

According to [2], the electrical conductivity tensor is given by

$$\sigma_{ij}(\omega) = \frac{N\,e^2}{m}\,\frac{i}{\omega}\,\delta_{ij} + \frac{i}{\hbar\,\omega}\int\limits_{-\infty}^{\infty} dt\; e^{(i\omega-\eta)t}\,\langle\langle J_i(t);\ J_j\rangle\rangle_r;\quad \eta \to +0 \quad (i,j = 1,2,3) \tag{2}$$

where ω is the pulsation frequency of the applied electric field and J_i is the i-th component of the current operator. If we denote by $G_{ij}(\omega)$ the Fourier transform of the Green function, equation (2) becomes

$$\sigma_{ij}(\omega) = \frac{N\,e^2}{m}\,\frac{i}{\omega}\,\delta_{ij} + \frac{2\,\pi\,i}{\hbar\,\omega}\,G_{ij}(\omega + i\,\eta)\,. \tag{3}$$

1. The Green Function

In the second quantization representation the currents are

$$J_i = \sum_{\alpha,\beta} (\alpha|j_i|\beta)\,a^+_\alpha\,a_\beta\,, \tag{4}$$

$(\alpha|j_i|\beta)$ being the one-electron current matrix element. Therefore we are interested in the computation of the Green functions of the form

$$G^{\gamma\delta}_{\alpha\beta}(t-t') = \langle\langle a^+_\alpha(t)\,a_\beta(t);\ a^+_\gamma(t')\,a_\delta(t')\rangle\rangle\,. \tag{5}$$

This function obeys the equation

$$i\hbar \frac{\partial}{\partial t} G^{\gamma\delta}_{\alpha\beta}(t-t') = \delta(t-t')\,\hbar\,\langle[a^+_\alpha a_\beta, a^+_\gamma a_\delta]\rangle + \langle\langle[a^+_\alpha(t)\,a_\beta(t), H];\; a^+_\gamma(t')\,a_\delta(t')\rangle\rangle\,, \tag{6}$$

where, for elastic interactions the Hamiltonian is

$$H = H_0 + V\,, \qquad H_0 = \sum_\mu \varepsilon_\mu a^+_\mu a_\mu\,, \qquad V = \sum_{\mu,\nu} V_{\mu\nu} a^+_\mu a_\nu\,. \tag{7}$$

Performing the commutations, we have

$$\begin{aligned} i\hbar \frac{\partial}{\partial t} G^{\gamma\delta}_{\alpha\beta}(t-t') = {} & \delta(t-t')\,\hbar\,(\delta_{\beta\gamma}\langle a^+_\alpha a_\delta\rangle - \delta_{\alpha\delta}\langle a^+_\gamma a_\beta\rangle) + (\varepsilon_\beta - \varepsilon_\alpha)\,G^{\gamma\delta}_{\alpha\beta}(t-t') + \\ & + \sum_\nu \left(V_{\beta\nu}\,G^{\gamma\delta}_{\alpha\nu}(t-t') - V_{\nu\alpha}\,G^{\gamma\delta}_{\nu\beta}(t-t')\right). \end{aligned} \tag{8}$$

Rearranging the terms we obtain for the Fourier transforms

$$\begin{aligned} (\hbar\omega + \varepsilon_\alpha - \varepsilon_\beta + V_{\alpha\alpha} - V_{\beta\beta})\,G^{\gamma\delta}_{\alpha\beta}(\omega) = {} & \frac{\hbar}{2\pi}(\delta_{\beta\gamma}\langle a^+_\alpha a_\delta\rangle - \delta_{\alpha\delta}\langle a^+_\gamma a_\beta\rangle) + \\ & + \sum_{\nu(\neq\beta)} V_{\beta\nu}\,G^{\gamma\delta}_{\alpha\nu}(\omega) - \sum_{\nu(\neq\alpha)} V_{\nu\alpha}\,G^{\gamma\delta}_{\nu\beta}(\omega)\,. \end{aligned} \tag{9}$$

Similar equations can be written for the Green functions $G^{\gamma\delta}_{\alpha\nu}(\omega), G^{\gamma\delta}_{\nu\beta}(\omega)$. Substituting these latter functions in (9) and omitting all the terms containing Green functions different from $G^{\gamma\delta}_{\alpha\beta}(\omega)$ (which are multiplied by the second power of the interaction) and neglecting also the correction to the inhomogenous term, we have

$$\begin{aligned} \left\{\hbar\omega + \bar\varepsilon_\alpha - \bar\varepsilon_\beta - \sum_{\nu(\neq\beta)} |V_{\beta\nu}|^2 \frac{1}{\hbar\omega + \bar\varepsilon_\alpha - \bar\varepsilon_\nu} - \sum_{\nu(\neq\alpha)} |V_{\alpha\nu}|^2 \frac{1}{\hbar\omega + \bar\varepsilon_\nu - \bar\varepsilon_\beta}\right\} G^{\gamma\delta}_{\alpha\beta}(\omega) = \\ = \frac{\hbar}{2\pi}\,\delta_{\alpha\delta}\,\delta_{\beta\gamma}(f_\alpha - f_\beta)\,, \end{aligned} \tag{10}$$

where $\bar\varepsilon_\alpha = \varepsilon_\alpha + V_{\alpha\alpha}$, $f_\alpha = \langle a^+_\alpha a_\alpha\rangle$. As it may be seen, our method for solving (9) is an iteration method. We had stopped at the second step, which is sufficient for our problem. Neglecting the energy corrections we have

$$G^{\gamma\delta}_{\alpha\beta}(\omega + i\eta) = \frac{\dfrac{\hbar}{2\pi}\,\delta_{\alpha\delta}\,\delta_{\beta\gamma}(f_\alpha - f_\beta)}{\hbar\omega + \varepsilon_\alpha - \varepsilon_\beta + i\pi \sum'_\nu \{|V_{\beta\nu}|^2\,\delta(\hbar\omega + \varepsilon_\alpha - \varepsilon_\nu) + |V_{\alpha\nu}|^2\,\delta(\hbar\omega + \varepsilon_\nu - \varepsilon_\beta)\}}\,. \tag{11}$$

2. Transverse Components of $\sigma_{ij}(\omega)$ in Magnetic Field

Let us apply a d.c. magnetic field in the direction of OZ axis. In the isotropic effective mass approximation the electron state is described by a Landau wave function, characterized by the quantum numbers $\alpha = (n, k_z, k_y)$, and the energy

$$\varepsilon_{n,k_z} = \hbar\,\omega_0\left(n + \frac{1}{2}\right) + \frac{\hbar^2 k_z^2}{2m}\,.$$

In this case

$$\left.\begin{aligned} J_1 &= i\,e\left(\frac{\hbar\,\omega_0}{2m}\right)^{1/2} \sum_{n,k} \sqrt{n+1}\,(a^+_{n+1,k}\,a_{n,k} - a^+_{n,k}\,a_{n+1,k})\,, \\ J_2 &= e\left(\frac{\hbar\,\omega_0}{2m}\right)^{1/2} \sum_{n,k} \sqrt{n+1}\,(a^+_{n+1,k}\,a_{n,k} + a^+_{n,k}\,a_{n+1,k})\,. \end{aligned}\right\} \tag{12}$$

As a consequence we obtain for the Fourier transforms of the Green functions G_{11}, G_{12} the following expressions:

$$\left.\begin{aligned} G_{11}(\omega) &= -e^2 \frac{\hbar\,\omega_0}{2\,m} \sum_{n,k} \sum_{n',k'} \sqrt{(n+1)\,(n'+1)} \times \\ &\times \left\{G^{n'+1,k';n',k'}_{n+1,k;n,k}(\omega) + G^{n',k';n'+1,k'}_{n,k;n+1,k}(\omega) - G^{n'+1,k';n',k'}_{n,k;n+1,k}(\omega) - G^{n',k';n'+1,k'}_{n+1,k;n,k}(\omega)\right\}, \\ G_{12}(\omega) &= i\,e^2 \frac{\hbar\,\omega_0}{2\,m} \sum_{n,k}' \sum_{n',k'} \sqrt{(n+1)\,(n'+1)} \times \\ &\times \left\{G^{n'+1,k';n',k'}_{n+1,k;n,k}(\omega) - G^{n',k';n'+1,k'}_{n,k;n+1,k}(\omega) - G^{n'+1,k';n',k'}_{n,k;n+1,k}(\omega) + G^{n',k';n'+1,k'}_{n+1,k;n,k}(\omega)\right\}. \end{aligned}\right\} \tag{13}$$

On the other hand, according to (11) we have

$$\left.\begin{aligned} G^{n'+1,k';n',k'}_{n+1,k;n,k}(\omega) &= G^{n',k';n'+1,k'}_{n,k;n+1,k}(\omega) = 0\,, \\ G^{n',k';n'+1,k'}_{n+1,k;n,k}(\omega + i\,\eta) &= \frac{\frac{1}{2\,\pi}\,\delta_{n\,n'}\,\delta_{k\,k'}\,(f_{n+1,k} - f_{n,k})}{\omega + \omega_0 + i\,\zeta^{-1}_{n,k}(-\omega)}\,, \\ G^{n'+1,k';n',k'}_{n,k;n+1,k}(\omega + i\,\eta) &= \frac{\frac{1}{2\,\pi}\,\delta_{n\,n'}\,\delta_{k\,k'}\,(f_{n,k} - f_{n+1,k})}{\omega - \omega_0 + i\,\zeta^{-1}_{n,k}(\omega)}\,, \end{aligned}\right\} \tag{14}$$

where

$$\begin{aligned} \zeta^{-1}_{n,k}(\omega) = \frac{\pi}{\hbar} \sum_{n',k'}'' \{&|V_{n,k;n',k'}|^2\,\delta\,(\varepsilon_{n+1,k} - \varepsilon_{n',k'} - \hbar\,\omega) + \\ &+ |V_{n+1,k;n',k'}|^2\,\delta\,(\varepsilon_{n,k} - \varepsilon_{n',k'} + \hbar\,\omega)\}\,. \end{aligned} \tag{15}$$

Therefore,

$$\begin{aligned} \sigma_{11}(\omega) = \frac{N\,e^2}{m}\,\frac{i}{\omega} + i\,\frac{e^2}{2\,m}\,\frac{\omega_0}{\omega} &\sum_{n,k}' (n+1)\,(f_{n,k} - f_{n+1,k}) \times \\ &\times \left\{\frac{1}{\omega - \omega_0 + i\,\zeta^{-1}_{n,k}(\omega)} + \frac{1}{\omega + \omega_0 + i\,\zeta^{-1}_{n,k}(-\omega)}\right\}, \end{aligned} \tag{16}$$

$$\begin{aligned} \sigma_{12}(\omega) = \frac{e^2}{2\,m}\,\frac{\omega_0}{\omega} &\sum_{n,k}' (n+1)\,(f_{n,k} - f_{n+1,k}) \times \\ &\times \left\{\frac{1}{\omega - \omega_0 + i\,\zeta^{-1}_{n,k}(\omega)} - \frac{1}{\omega + \omega_0 + i\,\zeta^{-1}_{n,k}(-\omega)}\right\}. \end{aligned} \tag{17}$$

In the case of interaction with acoustical phonons in the deformation potential model, the Hamiltonian is

$$\left.\begin{aligned} H &= H_0 + U\,, \\ H_0 &= \sum_{\mu}' \varepsilon_\mu\,a^+_\mu\,a_\mu + \sum_{\boldsymbol{q}}' \omega_{\boldsymbol{q}}\,b^+_{\boldsymbol{q}}\,b_{\boldsymbol{q}}\,, \\ U &= \sum_{\mu,\nu} (\mu|\delta V|\nu)\,a^+_\mu\,a_\nu\,, \\ \delta V &= g \sum_{\boldsymbol{q}} \frac{|q|}{\sqrt{\omega_{\boldsymbol{q}}}} \{b_{\boldsymbol{q}}\,e^{i\,\boldsymbol{q}\boldsymbol{r}} - b^+_{\boldsymbol{q}}\,e^{-i\,\boldsymbol{q}\boldsymbol{r}}\}, \end{aligned}\right\} \tag{18}$$

where g is the electron-phonon coupling constant and $\omega_{\boldsymbol{q}}$ is the energy of the phonon with the momentum $\boldsymbol{q}$.

As it may be shown, in the elastic approach, the results are analogous to the precedent case, but the computations becomes more complicated owing to the apparition of the many-particle Green functions and to necessity of the cut-off of the chain of equations by the customary contraction procedure [2].

So the formulae (16, 17) permits in principle the computation of the magneto-optical effects for the types of interactions mentioned.

3. Cyclotron Resonance

Performing the calculations for the scattering on point defects $\left(V(\boldsymbol{r}) = \sum_{n=1}^{N} V_0 \times \times \delta(\boldsymbol{r} - \boldsymbol{r}_n)\right)$ and acoustical phonons in the elastic approach, we find

$$\zeta_{n,k}^{-1}(\omega) = \omega_0 K \sum_{n'} \left\{ \frac{\theta\left[(n - n' + 1)\,\hbar\,\omega_0 - \hbar\,\omega + \dfrac{\hbar^2 k_z^2}{2\,m}\right]}{\sqrt{(n - n' + 1)\,\hbar\,\omega_0 - \hbar\,\omega + \dfrac{\hbar^2 k_z^2}{2\,m}}} + \right.$$

$$\left. + \frac{\theta\left[(n - n')\,\hbar\,\omega_0 + \hbar\,\omega + \dfrac{\hbar^2 k_z^2}{2\,m}\right]}{\sqrt{(n - n')\,\hbar\,\omega_0 + \hbar\,\omega + \dfrac{\hbar^2 k_z^2}{2\,m}}} \right\} \tag{19}$$

where

$$K = \begin{cases} \dfrac{\pi\, 2^{1/2}\, m^{3/2}}{\hbar^3} \dfrac{N\, V_0^2}{(2\,\pi)^5} & \text{for point defects,} \\[2ex] \dfrac{2^{1/2}\, m^{3/2}}{\hbar^3} \dfrac{k_0\, T\, g^2}{v_0^2} & \text{for acoustical phonons.} \end{cases} \tag{20}$$

(k_0 is the Boltzmann constant and v_0 the velocity of sound.)

In semiconductors $f_{n,k}$ is the canonical distribution function and

$$\operatorname{Re}\{\sigma_{11}(\omega) + i\,\sigma_{12}(\omega)\} = -\operatorname{Im}\left\{\frac{4\,N\,e^2}{m}\,\hbar \sqrt{\frac{\beta}{2\,m\,\pi\,\omega}}\,\frac{\omega_0}{\omega}\,\operatorname{sh}^2\frac{\beta\,\hbar\,\omega_0}{2}\, e^{-\beta\,\hbar\,\omega_0} \times \right.$$

$$\left. \times \sum_n (n+1)\, e^{-n\,\beta\,\hbar\,\omega_0} \int dk_z \frac{e^{-\beta\frac{\hbar^2 k_z^2}{2\,m}}}{\omega - \omega_0 + i\,\zeta_{n,k}^{-1}(\omega)} \right\}. \tag{21}$$

For $\beta\,\hbar\,\omega_0 \gg 1$, which is the ultra quantum limit in the case of semiconductors, from (21) we should retain only the term with $n = 0$

$$\operatorname{Re}\{\sigma_{11}(\omega) + i\,\sigma_{12}(\omega)\} = -\frac{N\,e^2}{m}\,\hbar\,\frac{\omega_0}{\omega} \sqrt{\frac{\beta}{2\,m\,\pi}}\, \operatorname{Im} \int dk_z \frac{e^{-\beta\frac{\hbar^2 k_z^2}{2\,m}}}{\omega - \omega_0 + i\,\zeta_{0,k}^{-1}(\omega)}\,. \tag{22}$$

In the same approach

$$\zeta_{0,k}^{-1}(\omega_0) = \omega_0\, K \left\{ \frac{2}{\sqrt{\dfrac{\hbar^2 k_z^2}{2\,m}}} + \frac{1}{\sqrt{\dfrac{\hbar^2 k_z^2}{2\,m} + \hbar\,\omega_0}} \right\} \tag{23}$$

and therefore

$$\mathrm{Re}\,\{\sigma_{11}(\omega_0) + i\,\sigma_{12}(\omega_0)\} = \frac{N e^2}{m}\,\hbar \sqrt{\frac{\beta}{2\,m\,\pi}} \int dk_z \frac{e^{-\beta\frac{\hbar^2 k_z^2}{2m}}}{\zeta^{-1}_{0,k}(\omega_0)} \approx \frac{N e^2}{2\,m} \sqrt{\frac{1}{\pi\,\beta}}\,\frac{1}{\omega_0 K}\,. \tag{24}$$

In terms of the zero magnetic field relaxation time, we have for the maximum of the resonance curve

$$\mathrm{Re}\,\{\sigma_{11}(\omega_0) + i\,\sigma_{12}(\omega_0)\} \approx \frac{N e^2}{m}\,\frac{\tau_0}{\beta\,\hbar\,\omega_0}\,. \tag{25}$$

To compute the half width of the resonance curve in circularly polarized light, we should take $\zeta^{-1}_{0,k}(\omega) \approx \zeta^{-1}_{0,k}(\omega_0)$. As a consequence, in the vicinity of the maximum, we have

$$\begin{aligned}\mathrm{Re}\,\{\sigma_{11}(\omega) + i\,\sigma_{12}(\omega)\} &= -\frac{N e^2}{m}\,\hbar \sqrt{\frac{\beta}{2\,m\,\pi}}\,\frac{\omega_0}{\omega}\,\mathrm{Im} \int dk_z \frac{e^{-\beta\frac{\hbar^2 k_z^2}{2m}}}{\omega - \omega_0 + i\,\zeta^{-1}_{0,k}(\omega_0)} = \\ &= -\frac{N e^2}{m}\,\frac{\hbar\,\beta}{\tau_0}\,\frac{2\,\omega_0^2}{3\,\pi\,\omega\,\Delta\omega^2}\, e^{\left(\frac{2\,\beta\,\hbar\,\omega_0}{3\sqrt{\pi}\,\tau_0\,\Delta\omega}\right)^2} E_i\left\{-\left(\frac{2\,\beta\,\hbar\,\omega_0}{3\sqrt{\pi}\,\tau_0\,\Delta\omega}\right)^2\right\},\end{aligned} \tag{26}$$

$(\Delta\omega = \omega - \omega_0)$.
The half width is obtained from the equation

$$\left(\frac{2\,\beta\,\hbar\,\omega_0}{3\sqrt{\pi}\,\tau_0\,\Delta\omega}\right)^2 e^{\left(\frac{2\,\beta\,\hbar\,\omega_0}{3\sqrt{\pi}\,\tau_0\,\Delta\omega}\right)^2} E_i\left[-\left(\frac{2\,\beta\,\hbar\,\omega_0}{3\sqrt{\pi}\,\tau_0\,\Delta\omega}\right)^2\right] = \frac{1}{2}\left(1 + \frac{\Delta\omega}{\omega_0}\right). \tag{27}$$

For $\dfrac{\Delta\omega}{\omega_0} \ll 1$ the numerical computation of this equation gives the following expression for $\Delta\omega$:

$$\frac{\Delta\omega}{\omega_0} \approx \frac{\beta\,\hbar}{\tau_0} \tag{28}$$

or if we define, according to (25) $\tau = \dfrac{1}{\beta\,\hbar\,\omega_0}\,\tau_0$, then

$$\frac{\Delta\omega}{\omega_0} \approx \frac{1}{\tau\,\omega_0}\,. \tag{29}$$

In metals we take

$$f(\varepsilon_{n,k}) = \begin{cases} 0 & \text{for} \quad \varepsilon_{n,k} > \varepsilon_F, \\ 1 & \text{for} \quad \varepsilon_{n,k} \leqq \varepsilon_F, \end{cases}$$

therefore

$$\mathrm{Re}\,\{\sigma_{11}(\omega) + i\,\sigma_{12}(\omega)\} = -\frac{2\,e^2}{m}\,\frac{\omega_0}{\omega}\,\mathrm{Im}\sum_n (n+1) \int\limits_{\sqrt{\frac{2m}{\hbar^2}\left[\varepsilon_F - \left(n+\frac{3}{2}\right)\hbar\,\omega_0\right]}}^{\sqrt{\frac{2m}{\hbar^2}\left[\varepsilon_F - \left(n+\frac{1}{2}\right)\hbar\,\omega_0\right]}} dk_z \frac{1}{\omega - \omega_0 + i\zeta_{n,}^{-1k}(\omega)}\,. \tag{30}$$

In the limit of high magnetic fields $\left(\frac{1}{2}\,\hbar\,\omega_0 < \varepsilon_F < \frac{3}{2}\,\hbar\,\omega_0\right)$, which is the ultra quantum limit in the case of metals, we find for the maximum and half width of resonance curve the following expressions:

$$\operatorname{Re}\{\sigma_{11}(\omega_0) + i\,\sigma_{12}(\omega_0)\} = \frac{N\,e^2}{m}\,\frac{\sqrt{E_F\left(\varepsilon_F - \frac{1}{2}\,\hbar\,\omega_0\right)}}{\hbar\,\omega_0}\,\tau_0\,, \tag{31}$$

$$\frac{\Delta\omega}{\omega_0} \approx \frac{\hbar}{\sqrt{E_F\left(\varepsilon_F - \frac{1}{2}\,\hbar\,\omega_0\right)}\,\tau_0} = \frac{1}{\tau\,\omega_0}\,, \tag{32}$$

where E_F and τ_0 are the Fermi energy and the relaxation time in the absence of the magnetic field.

As it was seen, we had obtained the formulae for the transverse components of the frequency dependent conductivity tensor in a d. c. magnetic field using the Green functions method, in all cases of elastic interactions. These formulae could be a starting point for the computations of magneto-optical constants. Using these results we had shown that in the ultra quantum limit the relative half width of the cyclotron resonance curve in the case of interaction with acoustical phonons and point defects becomes independent from applied magnetic field for semiconductors, and has a poor field dependence for metals. (Partial results on this subject had also been reported in the works [3, 4].)

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(Received September 14, 1963)

T1-13 ON THE THEORY OF ELECTRIC CONDUCTION IN AMORPHOUS SEMICONDUCTORS

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In disordered structures a so called « structural resistance » exists and is probably predominant up to not too high temperatures. Thus the theoretical treatment of conduction phenomena rely on finding the eigen-values and functions of the movement of an electron in a non-periodic potential, dependent on a great number of aleatory variables (the positions of the atoms). No satisfactory way of solving such a problem in its general form is yet known. But recent studies [1] give some qualitative informations about the energy spectrum and the character of the one-electron states in a disordered structure.

Essentially, when the short range order is maintained during the disordering, the) changes consist in the narrowing of the energy gap by strongly perturbing the electronic states near the band edges, most of them becoming localized. The farther we go inward the bands, the more non-localized the states become, their density approaching the value it had in the crystal.

Unlike liquid metals [2], these changes play a very important role in amorphous semiconductors. We shall describe them in a simplified energy scheme (Fig. 1) introducing a parameter ε to define the energy limit of the non-localized states.

In this scheme there will be two types of electrons : localized and non-localized ones. Assuming a priori that in transport phenomena the essential role is played by the last ones we shall try to calculate the structural conductivity.

In a first approximation the non-localized electrons could be treated as free electrons scattered by the disordered lattice, as it has been done in the case of liquid metals [2]. It seems quite natural that such an approach will not be sufficient. Thus we shall develop another way of solving the problem by a more suitable tight binding method.

The quantum mechanical calculation of the conductivity [3] require the knowledge of the wave functions of the non-localized electrons. We shall build these wave functions as superpositions of atomic functions

$$\psi(\boldsymbol{r}) = \sum_n c_n \, \varphi(\boldsymbol{r} - \boldsymbol{r}_n)$$

where $\boldsymbol{r}_n$ indicates the positions of the atoms.

We shall assume that the probability for a non-localized electron to be found near any of the atoms will be the same, thus $|c_n|^2 = 1/N$, where N is the number of atoms. When restoring the long range order, the phases must become $\boldsymbol{k}.\boldsymbol{r}_n$.

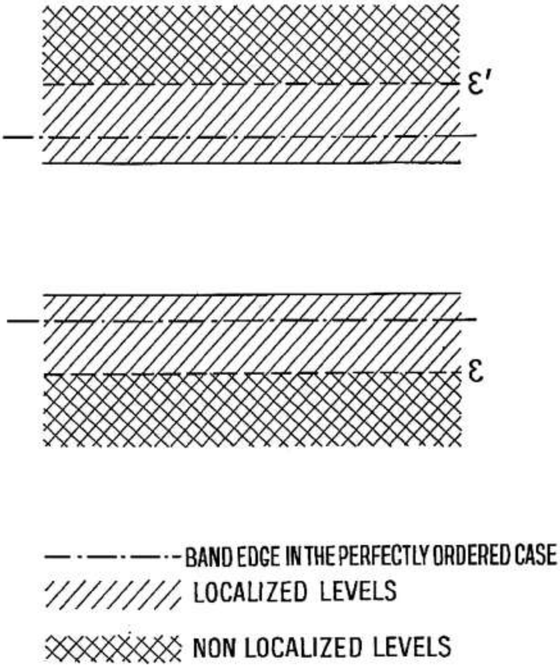

FIG. 1.

Therefore we shall take as an hypothesis that in amorphous elemental semiconductors with short range order the non-localized states of the electrons may be described by « quasi Bloch » wave functions

$$\psi_{\boldsymbol{k}}(\boldsymbol{r}) = \frac{1}{\sqrt{N}} \sum_n e^{i\boldsymbol{k}\boldsymbol{r}_n} \varphi(\boldsymbol{r} - \boldsymbol{r}_n) \,. \tag{1}$$

(For a chaotic distribution of the atoms this function has similar properties to those of the function taken by Kasuya in this treatment of impurity conduction [4].)

In the case of a disordered lattice such a function doesn't satisfy the ordinary Bloch condition and leads to a finite conductivity. Of course the ***k*** vector has no more the meaning of a wave vector, but as the short range order is maintained (and its role is predominant in the tight binding theory), the relation between the energy and the quantum number ***k*** in the case of a disordered lattice will be almost the same as the relation between the energy and the wave vector in the case of a perfectly ordered lattice, if the anisotropy is slight, so that an isotropic effective mass can be used.

Thus in the effective mass approximation

$$E_{\boldsymbol{k}} = \frac{\hbar^2}{2\, m^*} k^2 \tag{2}$$

and therefore the same density of states as in the unperturbed band will result (the energy origin is taken at the former band edge).

We shall now calculate the matrix element of the electric current between the non-localized states. We obtain

$$(\boldsymbol{k} \mid j_x \mid \boldsymbol{k}') = -\frac{ie}{\hbar N} \sum_{n,m} e^{i(\boldsymbol{k}'\boldsymbol{r}_m - \boldsymbol{k}\boldsymbol{r}_n)} \int d\boldsymbol{r} \varphi^*(\boldsymbol{r} - \boldsymbol{r}_n) \left[U(\boldsymbol{r} - \boldsymbol{r}_n) - U(\boldsymbol{r} - \boldsymbol{r}_m) \right] x \varphi(\boldsymbol{r} - \boldsymbol{r}_m)$$

where $U(\boldsymbol{r})$ is the potential of the free atom. Or, introducing the local effective potential $V(\boldsymbol{r})$

$$\begin{aligned} &\int d\boldsymbol{r} \varphi^*(\boldsymbol{r} - \boldsymbol{r}_n) \left[U(\boldsymbol{r} - \boldsymbol{r}_n) - V(\boldsymbol{r}) \right] x \varphi(\boldsymbol{r} - \boldsymbol{r}_m) - \\ &\quad - \int d\boldsymbol{r} \varphi^*(\boldsymbol{r} - \boldsymbol{r}_n) \left[U(\boldsymbol{r} - \boldsymbol{r}_m) - V(\boldsymbol{r}) \right] x \varphi(\boldsymbol{r} - \boldsymbol{r}_m) \approx \\ &\quad \approx x_m \int d\boldsymbol{r} \varphi^*(\boldsymbol{r} - \boldsymbol{r}_n) \left[U(\boldsymbol{r} - \boldsymbol{r}_n) - V(\boldsymbol{r}) \right] \varphi(\boldsymbol{r} - \boldsymbol{r}_m) - \\ &\quad - x_n \int d\boldsymbol{r} \varphi^*(\boldsymbol{r} - \boldsymbol{r}_n) \left[U(\boldsymbol{r} - \boldsymbol{r}_m) - V(\boldsymbol{r}) \right] \varphi(\boldsymbol{r} - \boldsymbol{r}_m) \approx (x_m - x_n) A(| \boldsymbol{r}_m - \boldsymbol{r}_n |) \end{aligned}$$

Thus

$$(\boldsymbol{k} \mid j_x \mid \boldsymbol{k}') = \frac{e}{\hbar N} \sum_m e^{i(\boldsymbol{k}' - \boldsymbol{k})\boldsymbol{r}_m} \frac{\partial}{\partial k_x} \left\{ \sum_n e^{-i\boldsymbol{k}(\boldsymbol{r}_n - \boldsymbol{r}_m)} A(| \boldsymbol{r}_m - \boldsymbol{r}_n |) \right\}.$$

The expression between the brackets is the very energy in the tight binding theory

$$E_{\boldsymbol{k}} = \sum_n e^{-i\boldsymbol{k}\boldsymbol{r}_n} A(| \boldsymbol{r}_n |) \approx \frac{\hbar^2 k^2}{2 m^*}.$$

Therefore

$$(\boldsymbol{k} \mid j_x \mid \boldsymbol{k}') = \frac{e\hbar k_x}{m^*} \cdot \frac{1}{N} \sum_m e^{i(\boldsymbol{k}' - \boldsymbol{k})\boldsymbol{r}_m}. \tag{3}$$

Substituting (3) in the quantum mechanical formula of the electric conductivity [3]

$$\sigma = -\pi\hbar \sum_{\boldsymbol{k},\boldsymbol{k}'} \frac{df(E_{\boldsymbol{k}})}{dE_{\boldsymbol{k}}} | (\boldsymbol{k} \mid j_x \mid \boldsymbol{k}') |^2 \delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}'}) \tag{4}$$

and taking into account the energetic position of the non-localized levels, we obtain

$$\sigma = -\frac{\pi e^2 \hbar^3}{(2\pi)^6 N m^{*2}} \int_{(E_{\boldsymbol{k}} > \varepsilon)} d\boldsymbol{k} \int d\boldsymbol{k}' \frac{df(E_k)}{dE_k} k_x^2 \, a(\boldsymbol{k} - \boldsymbol{k}') \, \delta(E_k - E_{k'}) \tag{5}$$

where

$$a(\boldsymbol{k}) = \frac{1}{N} \left| \sum_n e^{i\boldsymbol{k}\boldsymbol{r}_n} \right|^2$$

is the structure factor.

In the amorphous structures $a = a(|\boldsymbol{k}|)$ and the formula can be brought into the form

$$\sigma = -\frac{2\,e^2 m^*}{3(2\,\pi)^3\,\hbar^3\,N}\int_{\varepsilon}^{\infty} dE\,\frac{df(E)}{dE}\,E\int_0^{\sqrt{8m^*E/\hbar^2}} dx\,.\,xa(x)\,. \tag{6}$$

If $|\,\varepsilon - E_F\,|, \varepsilon \gg kT$, formula (6) can be approximately explicited and gives

$$\sigma = \frac{(em^*\,\varepsilon)^2\,\langle\, a\,\rangle}{3\,\pi^3\,\hbar^5\,N}\exp\left(-\frac{|\,\varepsilon - E_F\,|}{kT}\right) \tag{7}$$

where $\langle\, a\,\rangle$ is a mean structure factor.

Now we shall compare our theory with the experimental results of Grigorovici et al. [5] on amorphous Ge. In this case the large value of the activation energy of the electric conduction as well in the intrinsic as in the impurity range is easily explained by assuming a conduction by non-localized carriers in the band scheme drawn in Fig. 1. We shall put $\varepsilon = \varepsilon' \approx 0{,}2$ eV. Thus the conduction energy gap $\varDelta$ between the limits of the non-localized states will be $\varDelta = \varDelta^{\text{crystal}} + \varepsilon + \varepsilon' \approx 1{,}1$ eV, in agreement with the measured activation energy in the intrinsic conduction range of 0,55 eV.

This choice of the value of ε becomes even more plausible by evaluating the number of localized levels shifted from the valence band. Putting this number equal to the number of states between the valence band edge of the corresponding crystal and the ε level

$$\mathcal{N}_{\varepsilon} = \int_0^{\varepsilon} dEn(E)$$

one finds for $\varepsilon = 0{,}2$ eV a density of localized states $\mathcal{N}_{\varepsilon} \approx 10^{20}\ \text{cm}^{-3}$. This seems a fair value, as it exceeds noticeably all the measured acceptor densities.

The theory will be checked on the $\sigma(T)$ curve of heat-treated amorphous Ge, as in this case the curve is reversible and contains both intrinsic and impurity ranges, At 250 °K the conduction is p-type and $\sigma_{\text{exp}} = 4.10^{-5}\ \Omega^{-1}\ \text{cm}^{-1}$. Using the parameters of the valence band in crystalline Ge, putting the difference$|\,\varepsilon - E_F\,|$ constant and equal to the observed value of the activation energy of 0,18 eV, and $\varepsilon = 0{,}2$ eV, formula (7) gives $\sigma_{\text{theor}} = 4.10^{-4}\,\langle\, a\,\rangle\ \Omega^{-1}\ \text{cm}^{-1}$. An estimation of $\langle\, a\,\rangle$ leads to $\sim 0{,}1$ and therefore to a good agreement with σ_{exp}.

In view of the identity of the density of states beyond $E = \varepsilon$ both in the crystalline and amorphous substance, the density of the non-localized holes will be

$$p_n = \int_{\varepsilon}^{\infty} dEn(E)\,f(E)\,.$$

Therefore the mobility μ_n of the non-localized holes is given by the conductivity. Using the same parameters as above, we find $\rho_n \sim 10^{16}\ \text{cm}^{-3}$ and $\mu_n \sim 10^{-2}\ \text{cm}^2/\text{V sec}$. This low mobility of the non-localized carriers also explains the inapplicability, at least in the case of amorphous Ge, of the weak coupling method. Indeed a preliminary evaluation based on this method leads to a value of the conductivity exceeding the experimental one by a factor of about 10^3.

ACKNOWLEDGMENTS

The author would like to thank especially Dr R. Grigorovici for the proposal of the subject and fruitful discussions, and also V. Sergiescu and N. Croitoru for useful critical comments.

We are also grateful to Professor V. Novacu for encouragement of this work.

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PHYSICAL REVIEW VOLUME 143, NUMBER 2 MARCH 1966

Theory of the Hall Effect in Disordered Systems: Impurity-Band Conduction

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(Received 7 June 1965)

A general quantum-mechanical formula for the low-field Hall effect is derived in the density matrix approach, thus making it possible to treat this effect in disordered systems. With some assumptions about the one-electron matrix elements involved, the Hall and ordinary conductivities are correlated for the impurity-band conduction in the "metallic range." According to our results, the sign of the Hall effect depends on the degree of filling of the impurity band.

1. INTRODUCTION

THE theory of the Hall effect is an unclarified problem in disordered systems, such as impurity bands and amorphous semiconductors. In these systems the classical "scattering approach" to the transport problem fails, because in any approximation the electronic states can not be regarded as quasifree; consequently, such a basic concept as the effective mass can not be defined. But it is not clear what other parameter would then govern the sign of the Hall effect.

Some qualitative considerations, as yet unproved, can be found in a paper by Mott and Twose.[1] A rigorous basis for a future theoretical investigation of this problem is contained in R. Kubo's work[2] concerning the general theory of low-field Hall effect.

In this work, we derive the general formula for the low-field Hall conductivity in a manner different from that of Kubo, introducing adiabatically both the electric and magnetic fields (Sec. 2). Such an approach seems to be simpler. For a system of dynamically noninteracting electrons, using the second quantization formalism, we express the transverse conductivity in terms of products of certain one-electron matrix elements (involving momenta and the disordered potential), and Fermi distribution functions (Sec. 3).

The formula which has been obtained is applied to the case of high-concentration impurity-band conduction, where some simple assumptions about the matrix elements are possible (Sec. 4). According to our results, the sign of the Hall effect depends on the filling of the impurity band.

2. THE GENERAL FORMULA OF HALL CONDUCTIVITY

Let us consider the density-matrix equation

$$i\hbar d\rho/dt=[H+H'(t),\rho], \tag{1}$$

where H is the Hamiltonian of a dissipative system and $H'(t)$ is the adiabatically turned-on external-field Hamiltonian (electric and magnetic).

For a calculation of low-field Hall conductivity it is sufficient to consider the terms of the density matrix which are bilinear in the external electric and magnetic fields.

The second iterative solution of Eq. (1), with the initial condition

$$\rho|_{t\to-\infty}=\rho_0(H)$$

$[\rho_0(H)=$grand canonical equilibrium density matrix$]$ is

$$\rho^{(2)}=-\frac{1}{\hbar^2}\int_{-\infty}^{0}dt\int_{-\infty}^{0}dt'e^{s(t+t')}e^{iHt/\hbar}[H',e^{iHt'/\hbar}[H',\rho_0]e^{-iHt'/\hbar}]e^{-iHt/\hbar};\quad (s\to+0). \tag{2}$$

As we had mentioned before

$$H'=H_M+H_E, \tag{3}$$

where, in the second quantization formalism,

$$\begin{aligned}H_M&=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})\left(-\frac{e}{mc}\mathcal{H}xp_y+\frac{e^2}{2mc^2}\mathcal{H}^2x^2\right)\psi(\mathbf{r})\\ H_E&=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})(-e\mathcal{E}y)\psi(\mathbf{r}).\end{aligned} \tag{3a}$$

[We have chosen the dc magnetic and electric fields along Oz and Oy axes, respectively, with the potentials $\varphi=-\mathcal{E}y$ and $\mathbf{A}=(0,\mathcal{H}x,0)$.]

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Thus the average current, in which we are interested is

$$\langle j_x\rangle_{\mathrm{av}}=-\frac{e^3}{\Omega\hbar^2m^2c}\mathcal{E}\mathcal{H}\int_{-\infty}^{0}dt\int_{-\infty}^{0}dt'e^{s(t+t')}\,\mathrm{Tr}\{[Y,[M(t'),\rho_0]]P_x(-t)+[M,[Y(t'),\rho_0]]P_x(-t)\}\,, \tag{4}$$

where

$$Y=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})y\psi(\mathbf{r})\,;\quad M=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})x\hat{p}_y\psi(\mathbf{r})\,;\quad P_x=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})\hat{p}_x\psi(\mathbf{r})$$

and Ω is the volume of the system considered.

A step-by-step resolution of the commutators, using the well-known identity

$$[F,e^{zH}]=-e^{zH}\int_0^{z}d\lambda\,e^{\lambda H}[F,H]e^{-\lambda H}=-i\hbar e^{zH}\int_0^{z}d\lambda\,\dot{F}(-i\hbar\lambda)=i\hbar\int_0^{z}d\lambda\,\dot{F}(-i\hbar\lambda)e^{zH}\,,$$

gives for the transverse (Hall) conductivity

$$\sigma_H=-\frac{e^3}{\Omega\hbar m^2c}\mathcal{H}\int_{-\infty}^{0}dt\int_{-\infty}^{0}dt'e^{s(t+t')}\int_0^{\beta}d\lambda\bigg\langle\bigg\{i[\dot{M},Y]+i[\dot{Y},M]+\hbar\int_0^{(it'/\hbar+\lambda)}d\eta(\dot{M}\dot{Y}(-i\hbar\eta)+\dot{Y}\dot{M}(-i\hbar\eta))$$
$$-\hbar\int_0^{-(it'/\hbar+\lambda-\beta)}d\eta(\dot{M}(-i\hbar\eta)\dot{Y}+\dot{Y}(-i\hbar\eta)\dot{M})\bigg\}P_x(-t-t'+i\hbar\lambda)\bigg\rangle_0\,, \tag{5}$$

where the symbol $\langle\cdots\rangle_0$ means the average over the grand canonical distribution, and $\beta=1/kT$.

3. ONE-ELECTRON APPROACH

In what follows we shall make some suppositions which, of course, will restrict the generality of our results. We shall consider that our system of electrons can be described by a Hamiltonian of following type:

$$H=\int d\mathbf{r}\,\psi^\dagger(\mathbf{r})\bigg(-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r})\bigg)\psi(\mathbf{r})=\sum_\mu\epsilon_\mu a_\mu{}^\dagger a_\mu\,, \tag{6}$$

(that is, the electrons are dynamically independent).

Then, it can be easily shown that

$$\begin{aligned}&[Y,\dot{M}]=(\hbar/m)P_x\,;\quad[M,\dot{Y}]=0\,,\\&\dot{Y}=m^{-1}P_y=m^{-1}\textstyle\sum_{\mu,\nu}(\hat{p}_y)_{\mu\nu}a_\mu{}^\dagger a_\nu\,,\\&\dot{M}=\textstyle\sum_{\mu,\nu}(\hat{k})_{\mu\nu}a_\mu{}^\dagger a_\nu\,;\quad\hat{k}=m^{-1}(\hat{p}_x\hat{p}_y-x\partial V(\mathbf{r})/\partial_y)\,.\end{aligned} \tag{7}$$

Putting (7) into Eq. (5) and solving integrals over η, λ, t, and t' we have

$$\sigma_H=-\frac{e^3}{\Omega m^3c}\mathcal{H}\sum_{\substack{\mu_1,\mu_2,\mu_3,\\\nu_1,\nu_2,\nu_3}}\bigg\{\delta_{\mu_2\mu_1}\delta_{\nu_2\nu_1}\bigg(\frac{1}{(-i/\hbar)(\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\bigg)^2\frac{1-e^{-\beta(\epsilon_{\mu_3}-\epsilon_{\nu_3})}}{\epsilon_{\mu_3}-\epsilon_{\nu_3}}(\hat{p}_x)_{\mu_1\nu_1}(\hat{p}_x)_{\mu_3\nu_3}\langle a_{\mu_1}{}^\dagger a_{\nu_1}a_{\mu_3}{}^\dagger a_{\nu_3}\rangle_0$$
$$+\frac{((\hat{k})_{\mu_1\nu_1}(\hat{p}_y)_{\mu_2\nu_2}+(\hat{k})_{\mu_2\nu_2}(\hat{p}_y)_{\mu_1\nu_1})(\hat{p}_x)_{\mu_3\nu_3}}{(-i/\hbar)(\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\bigg[\frac{1}{\epsilon_{\mu_2}-\epsilon_{\nu_2}}\bigg(\frac{1}{(-i/\hbar)(\epsilon_{\mu_2}-\epsilon_{\nu_2}+\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\,\frac{1-e^{-\beta(\epsilon_{\mu_2}-\epsilon_{\nu_2}+\epsilon_{\mu_3}-\epsilon_{\nu_3})}}{\epsilon_{\mu_2}-\epsilon_{\nu_2}+\epsilon_{\mu_3}-\epsilon_{\nu_3}}$$
$$-\frac{1}{(-i/\hbar)(\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\,\frac{1-e^{-\beta(\epsilon_{\mu_3}-\epsilon_{\nu_3})}}{\epsilon_{\mu_3}-\epsilon_{\nu_3}}\bigg)-\frac{1}{\epsilon_{\mu_1}-\epsilon_{\nu_1}}\bigg(\frac{e^{\beta(\epsilon_{\mu_1}-\epsilon_{\nu_1})}}{(-i/\hbar)(\epsilon_{\mu_1}-\epsilon_{\nu_1}+\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\,\frac{1-e^{-\beta(\epsilon_{\mu_1}-\epsilon_{\nu_1}+\epsilon_{\mu_3}-\epsilon_{\nu_3})}}{\epsilon_{\mu_1}-\epsilon_{\nu_1}+\epsilon_{\mu_3}-\epsilon_{\nu_3}}$$
$$-\frac{1}{(-i/\hbar)(\epsilon_{\mu_3}-\epsilon_{\nu_3})+s}\,\frac{1-e^{-\beta(\epsilon_{\mu_3}-\epsilon_{\nu_3})}}{\epsilon_{\mu_3}-\epsilon_{\nu_3}}\bigg)\bigg]\langle a_{\mu_1}{}^\dagger a_{\nu_1}a_{\mu_2}{}^\dagger a_{\nu_2}a_{\mu_3}{}^\dagger a_{\nu_3}\rangle_0\bigg\}\,. \tag{8}$$

For the equilibrium average over the products of creation and annihilation operators we may use the formula

$$\langle a_{\mu_n}{}^\dagger\cdots a_{\mu_1}{}^\dagger a_{\nu_1}\cdots a_{\nu_n}\rangle_0=f(\epsilon_{\nu_1})\cdots f(\epsilon_{\nu_n})\,\det|\delta_{\mu_i\nu_k}|\,;\quad(i,k=1,\,\cdots,\,n)\,, \tag{9}$$

where $f(\epsilon)$ is the Fermi distribution function.

Taking into account that the equilibrium average of any of the involved one-electron operators $\hat{p}_x$, $\hat{p}_y$, $\hat{\kappa}$ vanishes, we need retain in Eq. (8) only the terms in which all the matrix indices are coupled. After a somewhat cumbersome regrouping of terms the calculation gives

$$\sigma_H=-\frac{e^3\hbar^2}{\Omega m^3c}\mathcal{H}\sum_{\mu_1,\mu_2,\mu}\Bigg\{\delta_{\mu_2\mu_1}\left(\frac{1}{\epsilon_{\mu_3}-\epsilon_{\mu_1}+i\hbar s}\right)^2\Delta f_{13}|(\hat{p}_x)_{\mu_1\mu_3}|^2$$
$$+\frac{((\hat{\kappa})_{\mu_2\mu_3}(\hat{p}_y)_{\mu_1\mu_2}+(\hat{\kappa})_{\mu_1\mu_2}(p_y)_{\mu_2\mu_3})(p_x)_{\mu_3\mu_1}}{\epsilon_{\mu_3}-\epsilon_{\mu_1}+i\hbar s}\Bigg[\frac{1}{\epsilon_{\mu_2}-\epsilon_{\mu_1}}\left(\frac{\Delta f_{32}}{\epsilon_{\mu_3}-\epsilon_{\mu_2}+i\hbar s}-\frac{\Delta f_{31}}{\epsilon_{\mu_3}-\epsilon_{\mu_1}+i\hbar s}\right)$$
$$+\frac{1}{\epsilon_{\mu_2}-\epsilon_{\mu_3}}\left(\frac{\Delta f_{21}}{\epsilon_{\mu_2}-\epsilon_{\mu_1}+i\hbar s}-\frac{\Delta f_{31}}{\epsilon_{\mu_3}-\epsilon_{\mu_1}+i\hbar s}\right)\Bigg]\Bigg\}, \quad (10)$$

where

$$\Delta f_{ij}=[f(\epsilon_{\mu_i})-f(\epsilon_{\mu_j})]/[\epsilon_{\mu_i}-\epsilon_{\mu_j}].$$

We mention that the same result may be obtained without the use of the second quantization formalism, by introducing into Eq. (4) a one-electron Hamiltonian and a Fermi-Dirac one-electron density matrix.

If we put into the Liouville equation (1) a term $i\hbar(\rho-\rho_0)/\tau$, which represents an ideal relaxation law, we obtain instead of Eq. (10) a similar expression, in which the adiabatic parameter s is changed to τ^{-1}. To verify our formula, it is easy to calculate σ_H for such an ideally relaxing free-electron system. Such a calculation gives the classical formula

$$\sigma_H=(Ne^3/\Omega m^2c)\mathcal{H}\tau^2. \quad (11)$$

Returning to our Eq. (10) for infinitesimal $\epsilon=\hbar s$, and using the identity

$$\frac{1}{\epsilon_j-\epsilon_i}\left(\frac{\Delta f_{kj}}{\epsilon_k-\epsilon_j+i\epsilon}-\frac{\Delta f_{ki}}{\epsilon_k-\epsilon_i+i\epsilon}\right)=\frac{\Delta f_{ik}+\Delta f_{jk}-\Delta f_{ij}}{(\epsilon_k-\epsilon_j+i\epsilon)(\epsilon_k-\epsilon_i+i\epsilon)},$$

we have

$$\sigma_H=-\frac{e^3\hbar^2}{\Omega m^3c}\mathcal{H}\sum_{\mu_1,\mu_2,\mu_3}\left(\frac{1}{\epsilon_{\mu_3}-\epsilon_{\mu_1}+i\epsilon}\right)^2\Bigg\{\delta_{\mu_2\mu_1}|(\hat{p}_x)_{\mu_1\mu_3}|^2\Delta f_{13}$$
$$+((\hat{\kappa})_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}+(\hat{\kappa})_{\mu_2\mu_3}(\hat{p}_y)_{\mu_1\mu_2})(\hat{p}_x)_{\mu_3\mu_1}\left[\frac{\Delta f_{13}+\Delta f_{23}-\Delta f_{12}}{\epsilon_{\mu_3}-\epsilon_{\mu_2}+i\epsilon}-\frac{\Delta f_{13}+\Delta f_{12}-\Delta f_{23}}{\epsilon_{\mu_2}-\epsilon_{\mu_1}+i\epsilon}\right]\Bigg\}. \quad (12)$$

(By interchanging the indices 1 and 3, it may be seen that this expression is real.)

With the notations

$$n(\epsilon_1)n(\epsilon_2)A(\epsilon_1,\epsilon_2)=\sum_{\substack{\mu_1,\mu_2\\(\epsilon_{\mu_1}=\epsilon_1,\ \epsilon_{\mu_2}=\epsilon_2)}}|(\hat{p}_x)_{\mu_1\mu_2}|^2, \quad (13)$$
$$n(\epsilon_1)n(\epsilon_2)n(\epsilon_3)B(\epsilon_1,\epsilon_2,\epsilon_3)=\sum_{\substack{\mu_1,\mu_2,\mu_3\\(\epsilon_{\mu_1}=\epsilon_1,\ \epsilon_{\mu_2}=\epsilon_2,\ \epsilon_{\mu_3}=\epsilon_3)}}((\hat{\kappa})_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}+(\hat{\kappa})_{\mu_2\mu_3}(\hat{p}_y)_{\mu_1\mu_2})(\hat{p}_x)_{\mu_3\mu_1},$$

and $n(\epsilon)$ for the density-of-states function, Eq. (12) reads

$$\sigma_H=-\frac{e^3\hbar^2}{\Omega m^3c}\mathcal{H}\Bigg\{\int d\epsilon_1\int d\epsilon_2\frac{d}{d\epsilon_1}\left(\frac{1}{\epsilon_2-\epsilon_1+i\epsilon}\right)n(\epsilon_1)n(\epsilon_2)\Delta f_{12}A(\epsilon_1,\epsilon_2)$$
$$+\int d\epsilon_1\int d\epsilon_2\int d\epsilon_3\frac{d}{d\epsilon_1}\left(\frac{1}{\epsilon_3-\epsilon_1+i\epsilon}\right)n(\epsilon_1)n(\epsilon_2)n(\epsilon_3)B(\epsilon_1,\epsilon_2,\epsilon_3)\left[\frac{\Delta f_{13}+\Delta f_{23}-\Delta f_{12}}{\epsilon_3-\epsilon_2+i\epsilon}-\frac{\Delta f_{13}+\Delta f_{12}-\Delta f_{23}}{\epsilon_2-\epsilon_1+i\epsilon}\right]\Bigg\}. \quad (14)$$

4. IMPURITY CONDUCTION

In disordered systems, of course, this expression must be averaged also over all the possible arrangements of the atoms. In the following, we shall assume, as is usually done in such problems, that after averaging, in Eq. (14), $n(\epsilon)$, $A(\epsilon_1,\epsilon_2)$, and $B(\epsilon_1,\epsilon_2,\epsilon_3)$ will appear with their mean values.

If the energy dependence of the averaged A and B is sufficiently smooth, they may be taken outside the integrals. Such a case probably occurs in impurity-band conduction, at least in the so-called "metallic range," as is shown in Kasuya's approach.[3] Thus, in the case considered, using the well-known identity

$$(x+i\epsilon)^{-1}=Px^{-1}-i\pi\delta(x),\quad \epsilon\rightarrow +0$$

we have

$$\sigma_H=-(e^3\hbar^2/\Omega m^3c)\mathcal{H}[\bar{A}I+\bar{B}(J_0+J_1)],\tag{15}$$

where

$$I=\int d\epsilon_1\int d\epsilon_2\frac{d}{d\epsilon_1}\left(P\frac{1}{\epsilon_2-\epsilon_1}\right)n(\epsilon_1)n(\epsilon_2)\Delta f_{12},$$

$$J_0=2\pi^2\int d\epsilon\, n^2(\epsilon)\frac{dn(\epsilon)}{d\epsilon}\frac{df(\epsilon)}{d\epsilon},\tag{15a}$$

$$J_1=\int d\epsilon_1\int d\epsilon_2\frac{d}{d\epsilon_1}\left(P\frac{1}{\epsilon_2-\epsilon_1}\right)n(\epsilon_1)n(\epsilon_2)\Delta f_{12}\int d\epsilon_3 n(\epsilon_3)\left(P\frac{1}{\epsilon_1-\epsilon_3}+P\frac{1}{\epsilon_2-\epsilon_3}\right).$$

Let us consider a density of states in the impurity band, of the type

$$n(\epsilon)=(N/\pi\Delta)[1+(\epsilon/\Delta)^2]^{-1},\tag{16}$$

where N is the total number of impurity states. With such a resonance-type function, the integrals (15) can be easily solved in the complex plane. For the integrals involving the symbol $d(Px^{-1})/dx$ we used the artifice

$$\int dx\,\varphi(x)\frac{d}{dx}\left(P\frac{1}{x}\right)=\lim_{a\rightarrow 0}\frac{d}{da}\int dx\,\varphi(x)P\frac{1}{x+a}.$$

The result is

$$\sigma_H=\frac{e^3\hbar^2}{\Omega m^3c}\mathcal{H}\frac{N^2\bar{A}}{\pi\Delta^3}\int_{-\infty}^{\infty}dx\left[f(x)\frac{2x(3-x^2)}{(1+x^2)^4}+\gamma\frac{df(x)}{dx}\frac{x(7-x^2)}{(1+x^2)^4}\right]\tag{17}$$

with

$$\gamma=\bar{B}N/\bar{A}\Delta\ \text{(dimensionless coefficient)}$$

and x being the energy in units of Δ.

If $kT\ll\Delta$, then $f(x)$ may be approximated by $\theta(\bar{\mu}-x)$ where $\bar{\mu}=\mu/\Delta$ ($\mu=$ chemical potential). Then, in the "metallic range," for low temperatures, the Hall conductivity reads explicitly

$$\sigma_H=-\frac{e^3\hbar^2}{\Omega m^3c}\mathcal{H}\frac{N^2\bar{A}}{\pi\Delta^3}\left[\frac{5-3\bar{\mu}^2}{6(1+\bar{\mu}^2)^3}+\gamma\frac{\bar{\mu}(7-\bar{\mu}^2)}{(1+\bar{\mu}^2)^4}\right].\tag{18}$$

On the other hand, concerning γ, we can observe that

$$\begin{aligned}(\hat{k})_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}(\hat{p}_x)_{\mu_3\mu_1}&=m^{-1}(\hat{p}_x\hat{p}_y)_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}(\hat{p}_x)_{\mu_3\mu_1}-m^{-1}(x\partial V/\partial y)_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}(\hat{p}_x)_{\mu_3\mu_1}\\&=m^{-1}|(\hat{p}_x)_{\mu_1\mu_3}|^2|(\hat{p}_y)_{\mu_2\mu_3}|^2+m^{-1}\sum_{\nu(\neq\mu_3)}(\hat{p}_x)_{\mu_1\nu}(\hat{p}_y)_{\nu\mu_2}(\hat{p}_y)_{\mu_2\mu_3}(\hat{p}_x)_{\mu_3\mu_1}\\&\quad-m^{-1}(x\partial V/\partial y)_{\mu_1\mu_2}(\hat{p}_y)_{\mu_2\mu_3}(\hat{p}_x)_{\mu_3\mu_1}.\end{aligned}$$

The first term of this expression is essentially real and positive, but the others have no definite sign. We may expect that their average value over a chaotic distribution of atoms will vanish. If this is correct, then

$$\bar{B}\approx 2\bar{A}^2/m.$$

Thus

$$\gamma\approx 2\bar{A}N/m\Delta.\tag{18a}$$

But we may easily show that the ordinary conductivity, with the same assumption about the averages, may be expressed also in terms of $\bar{A}$. Thus, according to the well-known quantum theory of electric conductivity,[4]

$$\sigma=-\frac{\pi e^2\hbar}{\Omega m^2}\sum_{\mu_1,\mu_2}\frac{df(\epsilon_{\mu_1})}{d\epsilon_{\mu_1}}|(\hat{p}_x)_{\mu_1\mu_2}|^2\delta(\epsilon_{\mu_1}-\epsilon_{\mu_2}).\tag{19}$$

After averaging over the impurities, we obtain

$$\sigma=-\frac{\pi e^2\hbar}{\Omega m^2}A\int d\epsilon\, n^2(\epsilon)\frac{df(\epsilon)}{d\epsilon}.\tag{20}$$

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[3] T. Kasuya, J. Phys. Soc. Japan **13**, 1096 (1958).

[4] R. Kubo, J. Phys. Soc. Japan **12**, 570 (1957).

Introducing into Eq. (20) our density of states (16), we get the following formula which is valid in the "metallic range" for low temperatures:

$$\sigma=(e^2\hbar/\Omega m^2)(N^2\bar{A}/\pi\Delta^2)(1+\bar{\mu}^2)^{-2}. \qquad (20a)$$

We may observe that the Hall and ordinary conductivity are correlated in a simple manner, through the phenomenological parameter $\bar{A}$:

$$\sigma_H=\frac{\hbar\omega_0}{\Delta}\left[\frac{5-3\bar{\mu}^2}{6(1+\bar{\mu}^2)}+\gamma\frac{\bar{\mu}(7-\bar{\mu}^2)}{(1+\bar{\mu}^2)^2}\right]\times\sigma; \quad \omega_0=\frac{|e|\mathcal{H}}{mc}, \qquad (21)$$

$$\gamma=(2\pi m/e^2\hbar)\Delta(1+\bar{\mu}^2)^2/(N/\Omega)\times\sigma. \qquad (21a)$$

By inspection of Eq. (21) and by comparison of its sign with that of the classical formula (11), we can conclude that the first term in (21) always gives a positive-charge contribution to the Hall effect, while the second gives a negative one for $\bar{\mu}<0$ and a positive one for $\bar{\mu}>0$. Therefore, for $\bar{\mu}>0$, that is for hole conduction in the impurity band—at least in the "metallic range"—we must have a positive-charge Hall effect. For low electron concentration our formula shows that a negative-charge Hall effect is very plausible; this seems, indeed, to be true because an estimate of γ [through Eq. (21a), using the data of the experiments of Fritzsche and Cuevas[5]] gives $\gamma\gg 1$. It is interesting that even so, there is a slight asymmetry in favor of the holes.

5. CONCLUSIONS

We have derived the general quantum-mechanical formula for low-field Hall conductivity. This is necessary for the discussion of the Hall effect in disordered systems, where such concepts as "effective mass" and "quasifree" approach are of doubtful value. Our formula was put in its one-electron form for a system of dynamically independent electrons. We think that such a formulation is possible for the majority of transport problems, particularly for disordered systems.

Assuming the constancy of certain matrix elements (averaged over all the possible arrangements of the atoms), we have correlated the Hall conductivity for an impurity band with its ordinary conductivity [Eqs. (17), (20)]. (Here we neglected the possible overlap of the impurity band with the nearest band of the host crystal, which occurs at too high impurity concentration.) Our assumption seems to be justified for the nonlocalized states, which are characteristic for the "metallic range" of impurity-band conduction. In this case, for low temperatures ($kT\ll\Delta$), according to our formula (18), both negative and positive Hall effects are possible (depending on the position of the Fermi level), in contradiction with Mott's arguments.[1] Nevertheless, the change of sign occurs when a symmetrical impurity band is less than half filled.

It seems that in the impurity-band case, as in the Bloch-band case, the sign of the Hall effect is governed by the sign of the first derivative of the density of states in the conduction region.

A thorough comparison of our results with the experiments could not be performed because only incomplete and uncertain experimental data are presently available.

We intend to apply our general formulas (5) or (14) to other interesting cases, such as the problem of the Hall conductivity of the amorphous semiconductors within the model used by one of the authors.[6]

ACKNOWLEDGMENTS

The authors are much indebted to Professor V. Novacu for his interest in this work and to V. Sergiescu, G. Ciobanu, and V. Rittenberg for very helpful discussions.

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[5] H. Fritzsche and M. Cuevas, Phys. Rev. 119, 1238 (1960).

[6] L. Bányai, *Proceedings of the Seventh International Conference on the Physics of Semiconductors* (Dunod Cie, Paris, 1964), p. 417.

Seminar de Fizică Teoretică 1, p.13 (1968)

Quantum theory of transport coefficients[1]

L. Bányai

1 Derivation of the electric conductivity formula (Kubo's formula)

The state of a quantum mechanical system (mixed ensemble) is defined by the statistical matrix ("density matrix") ρ. The average $\langle \mathbf{A} \rangle$ of a dynamic observable is given by

$$\langle \mathbf{A} \rangle = Tr\,(\rho \mathbf{A}) \tag{1}$$

The operator ρ in the Schrödinger picture is time-dependent and obeys the equation of motion (quantum mechanical Liouville equation)

$$\imath\hbar \frac{\partial \rho}{\partial t} = [\mathbf{H}, \rho] \tag{2}$$

with $\mathbf{H}$ being the Hamiltonian of the system.

The operator

$$\rho_0 = \exp\left(-\beta\left(\mathbf{H} - \mu\mathbf{N} - \boldsymbol{\Omega}\right)\right) \tag{3}$$

with

$$\exp\left(-\beta\Omega\right) = Tr\exp\beta\left(\mathbf{H} - \mu\mathbf{N}\right), \qquad \beta = \frac{1}{kT}$$

describes the thermodynamic equilibrium of a system in contact with a thermostat, having energy and particle exchange with. It is the analogue of the classical macro-canonical distribution.

Our task is to compute the average value of the electric current for the case of its deviation from equilibrium due to an applied external electric field.

We shall admit that at $t = -\infty$ the electric field was vanishing and the state of the system was described by the macro-canonical statistical matrix $\rho(\mathbf{H})$. Thereafter the external field is applied and the measurements are

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[1] See also a modern discussion of the same topic in recent books of the author. [18],[19].

made at some finite time t. (In this way one eliminates the transitory processes.) Other ways stated, we have to solve the equation

$$i\hbar\frac{\partial\rho}{\partial t} = \left[\mathbf{H} + \mathbf{H}_E\left(t\right), \rho\right] \quad . \tag{4}$$

where $\mathbf{H}_E$ is the interaction Hamiltonian with the external electrical field, while the initial conditions are

$$\mathbf{H}(-\infty) = 0, \qquad \rho\left(-\infty\right) = \rho_0 \tag{5}$$

In what concerns the time dependence of $\mathbf{H}_E\left(t\right)$ we take

$$\mathbf{H}_E\left(t\right) = \bar{\mathbf{H}}_E e^{i\omega t} e^{st}; \qquad \left(s \to 0^+\right) \quad . \tag{6}$$

Here s is the so called adiabatic parameter, that ensures the vanishing of the field at $t = -\infty$.(However, in the final stage we have to take the $s \to +0$ limit.)

Another way to formulate the problem would be to introduce the field suddenly at $t = 0$ and perform the measurement at $t = \infty$ (see Sec. 6). To give a proper mathematical formulation of the $t \to \infty$ limit, one has to introduce explicitly an interaction with the surrounding by a $\imath\hbar\frac{\rho-\rho_0}{\tau}$ $(\tau \to \infty)$ term in the Liouville equation.

These two formulations of the problem lead to identical results, since the role of the infinitesimal parameters s, $\frac{1}{\tau}$ was just to eliminate the oscillating solution due to a sudden coupling of the external field.

The linear differential equation equation Eq.5 is equivalent to the inhomogeneous integral equation

$$\rho\left(t\right) = \rho_0 + \frac{1}{i\hbar}\int_{-\infty}^{t} dt' e^{\frac{\imath}{\hbar}\mathbf{H}(t'-t)} \left[\mathbf{H}_E\left(t'\right), \rho\left(t'\right)\right] e^{-\frac{\imath}{\hbar}\mathbf{H}(t'-t)} \tag{7}$$

This integral equation may be solved in an iterative manner. Since we are interested only in the effects proportional (linear) in the external field, we may retain only the first term in this iteration

$$\rho_1\left(t\right) = \frac{1}{i\hbar}\int_{-\infty}^{t} dt' e^{\frac{\imath}{\hbar}\mathbf{H}(t'-t)} \left[\mathbf{H}_E\left(t'\right), \rho_0\left(t'\right)\right] e^{-\frac{\imath}{\hbar}\mathbf{H}(t'-t)} \quad . \tag{8}$$

Using Eq. 6 we get

$$\rho_1\left(t\right) = \frac{1}{i\hbar} e^{\imath(\omega-\imath s)t} \int_{-\infty}^{0} dt' e^{\frac{\imath}{\hbar}\mathbf{H}t'} \left[\overline{\mathbf{H}}_E, \rho_0\right] e^{-\frac{\imath}{\hbar}\mathbf{H}t'} \tag{9}$$

and therefore the average value of the electric current is

$$\begin{aligned} \left\langle \mathbf{j}_\mu\left(t\right)\right\rangle &= Tr\left\{\rho_0 \mathbf{j}_\mu\right\} + Tr\left\{\rho_1\left(t\right)\mathbf{j}_\mu\right\} \\ &= e^{i\omega t} \lim_{s\rightarrow+0} \frac{1}{i\hbar} \int_{-\infty}^{0} dt' e^{i(\omega-is)t'} Tr\left\{ e^{\frac{\imath}{\hbar}\mathbf{H}t'} \left[\overline{\mathbf{H}}_E, \rho_0\right] e^{-\frac{\imath}{\hbar}\mathbf{H}t'} \mathbf{j}_\mu \right\} \end{aligned}$$

On the other hand, using the operator identity

$$\left[\mathbf{A}, e^{-\beta\mathbf{H}}\right] = e^{-\beta\mathbf{H}} \int_0^\beta d\lambda e^{\lambda\mathbf{H}} \left[\mathbf{H}, \mathbf{A}\right] e^{-\lambda\mathbf{H}} \tag{10}$$

that my be easily checked by taking the matrix elements between the eigenstates of the Hamiltonian **H**, we have

$$\left[\overline{\mathbf{H}}_E, \rho_0\right] = -\imath\hbar\rho_0 \int_0^\beta d\lambda \dot{\overline{\mathbf{H}}}_E(-\imath\hbar\lambda) \quad ,$$

therefore

$$e^{-\imath\omega t}\langle \mathbf{j}_\mu(t)\rangle = -\lim_{s\rightarrow+0} \int_0^\infty dt' e^{-i(\omega-is)t'} \int_0^\beta d\lambda \langle \dot{\overline{\mathbf{H}}}_E(-i\hbar\lambda)\mathbf{j}_\mu(t')\rangle_0 \quad ,$$

where $\langle ... \rangle_0$ denotes averaging over the equilibrium state i.e. $Tr\left\{\rho_0\{...\}\right\}$. Thus

$$\left\langle \mathbf{j}_\mu\left(t\right)\right\rangle = V\sigma_{\mu\nu}E_\nu e^{i\omega t} \quad , \tag{11}$$

where

$$\sigma_{\mu\nu}\left(\omega\right) = \frac{1}{V} \lim_{s\rightarrow+0} \int_0^\infty dt' e^{-i(\omega-is)t'} \int_0^\beta d\lambda \left\langle \mathbf{j}_\mu\left(-i\hbar\lambda\right)\mathbf{j}_\mu\left(t\right)\right\rangle_0 \tag{12}$$

is the electric conductivity tensor (V being the normalization volume).

Eq.12 is the so called Kubo formula for the electric conductivity, defining this kinetic coefficient by the equilibrium characteristics of the system. It is exact in the whole domain of validity of Ohm's law. As we have seen, in its derivation no assumptions about the nature of the system played any role, but just about the external field. In the more general case of a time- and space- dependent field one may show a more general relation (for the average current density [1]

$$\langle \mathbf{j}_\mu (\mathbf{r}, t) \rangle = \int_{-\infty}^{t} dt' \int d\mathbf{r}' L_{\mu\nu} (\mathbf{r}, \mathbf{r}'; t - t') \, E_\nu (\mathbf{r}', \mathbf{t}') \tag{13}$$

with

$$L_{\mu\nu} (\mathbf{r}, \mathbf{r}', t) = \int_0^\beta d\lambda \, \langle \mathbf{j}_\nu (\mathbf{r}', 0) \, \mathbf{j}_\mu (\mathbf{r}, t + i\hbar\lambda) \rangle_0 \quad . \tag{14}$$

The connection between Eqs.12 and 13 is obvious.

Of course, these formulas need an explicit evaluation in each peculiar case, that as we shall see, is not at all trivial. However, from now on any computation of the conductivity relies only on the equilibrium properties of the considered system.

In the following we deal only with the case of homogeneous electric fields described by Eq. 12.

2 Symmetry relations and sum rules

The vanishing limit of the adiabatic parameter may be taken only after performing the time integral, since this defines the meaning of the improper integral. One may see this explicitly by expressing the average by insertion of the eigenfunctions of the operator **H**

$$\begin{aligned}
\sigma_{\mu\nu} (\omega) &= \frac{1}{V} \lim_{s \to +0} \int_0^\infty dt e^{-i(\omega - is)t} \int_0^\beta d\lambda \sum_{m,n} e^{-\beta(E_m - \mu N_m - \Omega)} \\
&\times \langle m | \mathbf{j}_\nu | n \rangle \langle n | \mathbf{j}_\mu | m \rangle \exp \left(\frac{i}{\hbar} (E_m - E_n) (t + i\hbar) \right) \\
&= \frac{1}{V} \lim_{s \to +0} \sum_{m,n} e^{-\beta(E_m - \mu N_m - \Omega)} \frac{e^{-\beta(E_n - E_m)} - 1}{E_m - E_n} \frac{\langle m | \mathbf{j}_\nu | n \rangle \langle n | \mathbf{j}_\mu | m \rangle}{\imath (\frac{E_n - E_m}{\hbar - \omega} - s)}
\end{aligned}$$

Now using the relation

$$\lim_{s \to +0} \frac{1}{x \pm is} = P \frac{1}{x} \mp i\pi\delta (x)$$

we get

$$\sigma_{\mu\nu}(\omega) = -\frac{\hbar}{V}\sum_{m,n}\frac{\rho_0(E_n)-\rho_0(E_m)}{E_n-E_m}\langle m|\mathbf{j}_\nu|n\rangle\langle n|\mathbf{j}_\mu|m\rangle \tag{15}$$

$$\times\left[\pi\delta(E_n-E_m-\hbar\omega)-iP\frac{1}{E_n-E_m-\hbar\omega}\right] \tag{16}$$

In this matrix form one may easily see the relations

$$\Re\sigma_{\mu\nu}(\omega)=\Re\sigma_{\nu\mu}(-\omega)\,,\ \Im\sigma_{\mu\nu}(\omega)=-\Im\sigma_{\nu\mu}(-\omega) \tag{17}$$

If one takes into account that

$$\langle|m\mathbf{j}_\mu|n\rangle=\langle n|\mathbf{j}_\mu|n\rangle*=-\langle\underline{n}|\underline{\mathbf{j}}_\mu|\underline{m}\rangle\ \ ,$$

where the state $|\underline{n}\rangle$ belongs to the same energy E_n and the operator $\underline{\mathbf{j}_\mu}$ belongs to the problem with reversed magnetic field $H\to -H$, then we get the relationship

$$\sigma_{\mu\nu}(\omega;H)=\sigma_{\nu\mu}(\omega;-H)\ \ . \tag{18}$$

On the ground of the above general Eq. 16 one may derive an interesting sum-rule for the diagonal elements of the conductivity tensor

$$\sigma_{\mu\mu}(\omega)=\lim_{s\to+0}\int_0^\infty dte^{-i(\omega-is)t}\varphi_{\mu\mu}(t)\,,$$

where

$$\begin{aligned}\varphi_{\mu\mu}(t) &= \int_0^\beta d\lambda\,\langle\mathbf{j}_\mu(-i\hbar\lambda)\,\mathbf{j}_\mu(t)\rangle_0\\ &= \sum_{m,n}\frac{\rho_0(E_n)-\rho_0(E_m)}{E_n-E_m}\left|\langle m|\mathbf{j}_\mu|n\rangle\right|^2\exp\left(-\frac{i}{\hbar}(E_m-E_n)\,t\right)\end{aligned}$$

is a real even function of time.

Thus

$$\begin{aligned}\int_{-\infty}^\infty d\omega\Re\sigma_{\mu\mu}(\omega) &= \int_0^\infty dt\varphi_{\mu\mu}(t)\cdot\Re\int_{-\infty}^\infty e^{-\imath(\omega-\imath s)t}\\ &= 2\pi\int_0^\infty dt\varphi_{\mu\mu}(t)\,\delta(t)=2\pi\varphi_{\mu\mu}(0)\,.\end{aligned}$$

On the other hand

$$\begin{aligned}
\varphi_{\mu\mu}(0) &= \int_0^\beta d\lambda \left\langle \mathbf{j}_\mu(-i\hbar\lambda)\,\mathbf{j}_\mu(t)\right\rangle_0 \\
&= \frac{1}{i\hbar} Tr\left\{\left[\rho_0, \sum_i e_i \mathbf{x}_\mu^i\right] \sum_j e_j \mathbf{x}_\mu^j\right\} \\
&= \frac{1}{i\hbar}\sum_{i,j} e_i e_j \left\langle \left[\mathbf{x}_\mu^i, \frac{d}{dt}\mathbf{x}_\mu^i\right]\right\rangle_0 = \sum_i \frac{e_i^2}{m_i}.
\end{aligned}$$

Then, using Eq. 17 we have also the relationship

$$\int_0^\infty d\omega \Re\sigma_{\mu\mu}(\omega) = \pi \sum_i \frac{e_i^2}{m_i}. \tag{19}$$

The cyclic permutation of the operators under the trace is not obviously an allowed manipulation in the case of unbounded operators. Therefore, this last relationship must be looked up with some caution.

3 The link to the Green function

We want to show now another useful form of the electric conductivity formula, that relates it to the statistical Green functions of the currents, respectively to the correlation functions of the currents [3]. We start from the equation

$$\sigma_{\mu\nu}(\omega) = -\frac{1}{i\hbar}\int_0^\infty dt e^{-(i\omega+s)t'} Tr \int_0^\beta \left\{\left[\sum_i e_i \mathbf{x}_\nu^i, \rho_0\right], \mathbf{j}_\mu(t)\right\} \quad (s \to +0)$$

proven in Sec. 1 (here we took the volume $V = 1$). Using the cyclic permutation of the operators under the trace (see also the remark at the end of the preceding Section) and introducing the step function

$$\theta(t) = \begin{cases} 1 & t > 0 \\ 0 & t < 0 \end{cases}$$

we get as it follows

$$
\begin{aligned}
\sigma_{\mu\nu}(\omega) &= \frac{1}{i\hbar}\int_{0}^{\infty} dt e^{-(i\omega+s)t}\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} \\
&= \frac{1}{i\hbar}\int_{-\infty}^{\infty} dt e^{-(i\omega+s)t}\theta(t)\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} \\
&= \frac{1}{\hbar\omega}\int_{-\infty}^{\infty} dt\frac{d}{dt}\left(e^{-(i\omega+s)t}\right)\theta(t)\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0}
\end{aligned}
$$

After a partial integration we have

$$
\begin{aligned}
\sigma_{\mu\nu}(\omega) &= -\frac{1}{\hbar\omega}e^{-(i\omega+s)t}\theta(t)\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0}\Bigg|_{-\infty}^{\infty} \\
&+ \frac{1}{\hbar\omega}\int_{-\infty}^{\infty} dt\delta(t)\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} \\
&- \frac{1}{\hbar\omega}\int_{-\infty}^{\infty} dt e^{-(\imath\omega+s)t}\theta(t)\left\langle\left[\frac{d}{dt}\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} \quad .
\end{aligned}
$$

The first term vanishes , the second gives rise to

$$
\frac{1}{\hbar\omega}\left\langle\left[\mathbf{j}_{\mu}(t), \sum_{i} e_{i}\mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} = \frac{1}{\hbar\omega}\sum_{i,j} e_{i}e_{j}\mathbf{x}_{\nu}^{i}\left\langle\left[\frac{d\mathbf{x}_{\mu}^{i}}{dt}, \mathbf{x}_{\nu}^{i}\right]\right\rangle_{0} = \frac{1}{i\omega}\sum_{i}\frac{e_{i}^{2}}{m_{i}}\delta_{\mu\nu}
$$

and if we have a single sort of carrier, it is $\frac{Ne^{2}}{m}\frac{1}{\imath\omega}\delta_{\mu\nu}$.

Using the Heisenberg equation of motion

$$
i\hbar\frac{\partial\mathbf{j}_{\mu}(t)}{\partial t} = \left[\mathbf{j}_{\mu}(t), \mathbf{H}\right]
$$

and the cyclic permutability under the trace one may overpass the time derivative with opposite sign on the coordinates. Thus we get

$$
\sigma_{\mu\nu}(\omega) = \frac{Ne^{2}}{m}\frac{1}{i\omega}\delta_{\mu\nu} + \frac{1}{\hbar\omega}\int_{-\infty}^{\infty} dt e^{-i(\omega+s)t}\theta(t)\left\langle\left[\mathbf{j}_{\mu}(t), \mathbf{j}_{\nu}\right]\right\rangle_{0} \qquad (20)
$$

or

$$\sigma_{\mu\nu}(\omega) = \frac{Ne^2}{m}\frac{1}{i\omega}\delta_{\mu\nu} + \frac{2\pi}{i\hbar\omega}\widetilde{G}_r^{\mu\nu}(\omega + is), \tag{21}$$

where $\widetilde{G}_r^{\mu\nu}(t)$ is the Fourier transform of the retarded statistical Green function of the currents

$$\widetilde{G}_r^{\mu\nu}(t) = -i\theta(t)\left\langle\left[\mathbf{j}_\mu(t), \mathbf{j}_\nu(0)\right]\right\rangle_0 \quad . \tag{22}$$

Farther, using the well-known dispersion relations of the Green functions , we may express thew real part of the electric conductivity tensor by the correlation function of the currents

$$\Re\sigma_{\mu\nu}(\omega) = \frac{2\pi}{\hbar\omega}\Im\widetilde{G}_r^{\mu\nu}(t) = \frac{e^{\beta\hbar\omega}-1}{2\hbar\omega}\int_{-\infty}^{\infty} dt\,\left\langle\mathbf{j}_\mu(t)\,\mathbf{j}_\nu(0)\right\rangle_0 e^{-i\omega t} \tag{23}$$

The link to the statistical Green functions is of utmost importance, since this offers a way for the calculation of the conductivity (mostly in *many body* problems). Thus we get two important tools to compute the Green functions. The first one consist in "cutting" the chain of the equations for the Green functions and solving the finite system of remaining equations. The other one is based on the Feynman diagram technique developed for the causal Matsubara Green functions (with imaginary time) and the analytic continuation of their Fourier coefficients in the whole upper complex plane until the real axis.

4 The one-electron version of the Kubo formula

When the system of carriers (electrons) may be described by an one electron Hamiltonian i.e. we have to do with dynamically independent particles being in an external potential, the conductivity formula, taking still into account the exclusion principle, takes a peculiar "one-electron" form.

$$H = \sum_{k,k'} h_{kk'}\mathbf{a}_k^+\mathbf{a}_{k'}, \tag{24}$$

where $\mathbf{a}_k^+$ and $\mathbf{a}_k$ are the creation and annihilation operators in an arbitrary basis denoted by the quantum numbers k, and $h_{kk'}$ are the matrix elements of the one-electron in this basis. The average value of the electric current is then

$$\langle \mathbf{j}_\mu (t)\rangle = Tr\left\{\rho (t)\, \mathbf{j}_\mu\right\} = \sum_{k,k'} \left(\mathbf{i}_\mu\right)_{kk'} Tr\left\{\rho (t)\, \mathbf{a}_k^+ \mathbf{a}_{k'}\right\} \quad , \qquad (25)$$

where $\mathbf{i}$ is the one-electron current.

Let us define an one-electron operator $\mathbf{f}$ ("one-electron density matrix") by

$$f_{kk'} = \left\langle \mathbf{a}_k^+ \mathbf{a}_{k'} \right\rangle \quad . \qquad (26)$$

Then

$$\langle \mathbf{j}_\mu (t)\rangle = tr \mathbf{f}\,(t)\, \mathbf{i}_\mu \qquad (27)$$

and as it is easily seen, the equation of motion of this operator is

$$i\hbar \frac{\partial \mathbf{f}}{\partial t} = [\mathbf{h}, \mathbf{f}] \qquad (28)$$

(Had we also not only one-electron terms in H, then we could have obtain at-most a chain of coupled equations for a series of operators like f.)

On the other hand, in the eigenstates of the energy (for a system having particle exchange with the thermostat)

$$f_{\alpha\alpha'} = \delta_{\alpha\alpha'} \frac{1}{\exp \frac{\varepsilon_\alpha - \mu}{kT} + 1} \quad ,$$

thus in equilibrium the operator f looks as

$$\mathbf{f}_0 = \frac{1}{e^{\beta(\mathbf{h}-\mu)}} \quad . \qquad (29)$$

We may formulate now the electric conductivity problem directly in terms of the one-electron density matrix f. A similar reasoning to that described in Section 1 leads to the equation

$$\sigma_{\mu\nu}(\omega) = -\frac{e}{i\hbar} \int_0^\infty dt e^{-(i\omega+s)t} tr\left\{[\mathbf{x}_\nu, \mathbf{f}_0]\, \mathbf{i}_\mu\right\} \quad . \qquad (30)$$

For the next step one has to use the peculiar form of the identity Eq. 10, namely

$$\left[\mathbf{a},\frac{1}{e^{\beta(\mathbf{h}-\mu)}}\right]=\frac{1}{e^{\beta(\mathbf{h}-\mu)}}\int_0^\beta d\lambda e^{\lambda\mathbf{h}}\left[\mathbf{h},\mathbf{a}\right]e^{-\lambda\mathbf{h}}\left(1-\frac{1}{e^{\beta(\mathbf{h}-\mu)}}\right)\quad . \tag{31}$$

Using this relation we get finally

$$\begin{aligned}\sigma_{\mu\nu}(\omega) &= \int_0^\infty dte^{-(i\omega+s)t}\int_0^\beta d\lambda tr\left\{\mathbf{f}_0\mathbf{i}_\nu(-i\hbar\lambda)(1-\mathbf{f}_0)\,\mathbf{i}_\mu(t)\right\} && (32)\\ &= \int_0^\infty dte^{-(i\omega+s)t}\int_0^\beta d\lambda\left\langle\mathbf{i}_\nu(-i\hbar\lambda)(1-\mathbf{f}_0)\,\mathbf{i}_\mu(t)\right\rangle_0 && (33)\end{aligned}$$

5 Explicit application of the one-electron formula to elastic scattering

The one-electron formula we derived in the preceding Section is especially useful to compare the quantum mechanical theory with the semi-classical Boltzmann one [2]. Let us consider a system of free electrons scattered on some static scatterer, described by the one-electron

$$h=-\frac{\hbar^2}{2m}\nabla^2+V(\mathbf{r})=\mathbf{h}_0+\mathbf{V} \tag{34}$$

If we make the approximation

$$\mathbf{f}_0(\mathbf{h})\simeq\mathbf{f}_0(\mathbf{h}_0) \tag{35}$$

in Eq. 30, then instead of the Eq. 33 we get

$$\begin{aligned}\sigma_{xx}(\omega) &= \beta\int_0^\infty dte^{-st}tr\left\{\mathbf{f}_0(\mathbf{h}_0)\,\mathbf{j}_x(1-\mathbf{f}_0(\mathbf{h}_0))\,\mathbf{j}_x(t)\right\} && (36)\\ &= -\beta\int_0^\infty dte^{-st}tr\left\{\frac{\partial\mathbf{f}_0(\mathbf{h}_0)}{\partial\mathbf{h}_0}\mathbf{f}_0(\mathbf{h}_0)\,\mathbf{j}_x\mathbf{j}_x(t)\right\} && (37)\end{aligned}$$

or, in the plane wave basis (after normalization in a box),

---

[2] Here as in he following the author meant the rate equation with inclusion of the Pauli principle. It was a frequent misuse of terminology in the early times.

$$\sigma_{xx} = -\frac{e^2\hbar^2}{m^2}\sum_{\mathbf{k}}\frac{\partial f\left(\varepsilon_k\right)}{\partial\varepsilon_k}\mathbf{k}_x\mathcal{K}_{\mathbf{k},\mathbf{k}}\left(s\right) \quad \left(s \to +0\right) \tag{38}$$

$$\mathbf{K}\left(s\right) = \int_0^\infty dt e^{-st}\mathbf{k}_x\left(t\right) \tag{39}$$

We shall compute the diagonal matrix element of $\mathbf{K}(s)$ using the equation of motion of $\mathbf{k}_x(t)$. One may see, that

$$\mathbf{K}\left(s\right) = -\frac{1}{s}\int_0^\infty dt\frac{d}{dt}\left(e^{-st}\right)\mathbf{k}_x\left(t\right) = -\frac{1}{s}\left\{-\mathbf{k}_x - \int_0^\infty dt e^{-st}\frac{d}{dt}\mathbf{k}_x\left(t\right)\right\} \ ,$$

giving rise to

$$s\mathbf{K}\left(s\right) = \mathbf{k}_x + \frac{1}{i\hbar}\left[\mathbf{K}\left(s\right), \mathbf{h}\right] \tag{40}$$

or, as matrix elements

$$\left(s + i\omega_{k'k}\right)\mathcal{K}_{\mathbf{k},\mathbf{k}'}\left(s\right) = k_x\delta_{k'k} + \frac{1}{i\hbar}\sum_{\mathbf{q}}\left(\mathcal{K}_{\mathbf{k},\mathbf{q}}\left(s\right)V_{\mathbf{q},\mathbf{k}'} - V_{\mathbf{k},\mathbf{q}}\mathcal{K}_{\mathbf{q},\mathbf{k}'}\left(s\right)\right) \tag{41}$$

with

$$\omega_{k'k} = \frac{\varepsilon_{k'} - \varepsilon_k}{\hbar} \ .$$

Let us admit, that the perturbation V is so weak, that the diagonal matrix elements of $\mathbf{K}(s)$ are mach bigger than the non-diagonal ones. Such a singular approximation becomes plausible if we observe, that the potential itself hat such singular character, at least after an averaging over the random distribution of the scattering centers. Indeed , if $V(\mathbf{r}) = \sum_i v(\mathbf{r} - \mathbf{r}_i)$, then $V_{\mathbf{kk}'} = v_{\mathbf{kk}'}\sum_i e^{\imath(\mathbf{k}-\mathbf{k}')\mathbf{r}_i}$ and due to the random oscillation of the phases the non-diagonal elements are proportional to $\sqrt{\mathcal{N}}$, while the diagonal ones are proportional to $\mathcal{N}$ (the number of scatterers). Under this seemingly self-consistent assumption, we get in a first approximation for the non-diagonal matrix elements

$$\mathcal{K}_{\mathbf{q},\mathbf{k}'}\left(s\right) \simeq \frac{V_{\mathbf{k},\mathbf{k}'}\left(s\right)}{\hbar\omega_{k'k} + i\hbar s}\left(\mathcal{K}_{\mathbf{k},\mathbf{k}}\left(s\right) - \mathcal{K}_{\mathbf{k}',\mathbf{k}'}\left(s\right)\right) \ . \tag{42}$$

Inserting this expression in the equation for the diagonal matrix elements leads to

$$s\mathcal{K}_{\mathbf{k},\mathbf{k}}\left(s\right) = k_x + \frac{1}{i\hbar}\sum_{\mathbf{q}}\left|V_{\mathbf{k},\mathbf{q}}\right|^2\left(\frac{1}{\hbar\omega_{\mathbf{qk}} + i\hbar s} - \frac{1}{\hbar\omega_{\mathbf{qk}} + i\hbar s}\right)\left(\mathcal{K}_{\mathbf{kk}}\left(s\right) - \mathcal{K}_{\mathbf{qq}}\left(s\right)\right). \tag{43}$$

After performing the $s \to +0$ limit, we get the equation

$$k_x = \sum_{\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'} \left(\mathcal{K}_{\mathbf{k},\mathbf{k}} (s) - \mathcal{K}_{\mathbf{k}',\mathbf{k}'} (s) \right) , \tag{44}$$

where

$$W_{\mathbf{k}\mathbf{k}'} = \frac{2\pi}{\hbar} \left| V_{\mathbf{k},\mathbf{k}'} \right|^2 \delta \left(\varepsilon_{k'} - \varepsilon_k \right) \tag{45}$$

is the $\mathbf{k} \to \mathbf{k}'$ transition rate. If this rate depends only on the energies of the initial and final states, we may look for the solution as

$$\mathcal{K}_{\mathbf{k},\mathbf{k}} = k_x \tau \left(\varepsilon_k \right) \tag{46}$$

and get finally

$$\tau^{-1} \left(\varepsilon_k \right) = \sum_{\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'} \frac{k_x - k'_x}{k_x} = \sum_{\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'} \left(1 - \cos \theta \right) \quad . \tag{47}$$

Therefore, under the above approximations, after passing from sums to integrals, we get for the conductivity

$$\sigma_{xx} = - \frac{e^2 \hbar^2}{m^2} \frac{1}{(2\pi)^2} \int dk \frac{d f_0 \left(\varepsilon_k \right)}{d \varepsilon_k} k_x^2 \tau \left(\varepsilon_k \right) \tag{48}$$

This expression coincides with the one given by the linearized Boltzmann equation and the evaluation of the transition rates through the quantum mechanical perturbation theory. As one may see from the proof given in the Appendix, the coincidence of the results is obvious, since under similar approximations The linearized Boltzmann equation may be derived from quantum statistics [5], [7].

These conclusions are general and obvious, although we got it for a particular case. Always, when a semi-classical Boltzmann equation is valid, the Kubo formula would lead to the same result. The exact formulas for the conductivity however offer the possibility to deal also with cases outside the availability of a Boltzmann formulation of the transport problem.

It is true however, that convenient methods of approximations in the Kubo formula are not available for all cases and the existent ones imply solving integral equations that are not simpler as the Boltzmann equation itself. Nevertheless, their validity extends that of the Boltzmann equation. For example, in the above example we could have continued the iteration with respect to the scattering potential (as formerly mentioned)and therefor obtain results valid also for higher concentration of the scattering centers.

Some of the cases, where the Boltzmann treatment fails due to strong quantum mechanical effects are : strong magnetic fields, high frequencies, strong couplings (very low mobilities).

For a general , more convenient discussion of these cases we shall derive in the next Section the general expression of the conductivity tensor for an *ideal relaxation.*

6 Ideal relaxation

Let us introduce an additional relaxation term

$$\imath\hbar\frac{\rho-\rho_0}{\tau}$$

in the right -hand side of the Eq. 2 of the statistical matrix, with a constant relaxation time τ. Such a term might replace in an oversimplified version the effects of a supplementary dissipative interaction. The solution of such an equation (in the linear approximation with respect to the external field) with the initial condition

$$\rho(0)=\rho_0$$

and sudden introduction of the filed at $t=0$ is

$$\rho=\rho_0+\frac{1}{i\hbar}e^{-\frac{t}{\tau}}\int_0^t dt' e^{t'/\tau}\exp\left(i\frac{\mathbf{H}\left(t'-t\right)}{\hbar}\right)\left[\mathbf{H}_E\left(t'\right),\rho_0\right]\exp\left(-i\frac{\mathbf{H}t'}{\hbar}\right) \quad (49)$$

or, after a sufficiently long time ($t\to\infty$) after the introduction of the field one has

$$\rho=\rho_0+\frac{1}{i\hbar}e^{i\omega t}\int_{-\infty}^0 dt' e^{\left(i\omega+\tau^{-1}\right)}\exp\left(i\frac{\mathbf{H}t'}{\hbar}\right)\left[\overline{\mathbf{H}}_E\left(t'\right),\rho_0\right]\exp\left(-i\frac{\mathbf{H}t'}{\hbar}\right) \quad (50)$$

and this leads to the formula

$$\sigma_{\mu\nu}\left(\omega\right)=\int_{-\infty}^0 dt' e^{\left(i\omega+\tau^{-1}\right)}\int_0^\beta d\lambda\left\langle \mathbf{j}_\nu\mathbf{j}_\mu\left(t+i\hbar\lambda\right)\right\rangle_0 \quad (51)$$

One may see, that for $\tau\to\infty$ Eq. 12 obtained by the adiabatic introduction of the electric field results.

If one may simulate the effect of dissipation alone by such a relaxation term, ensuring the finiteness of $\sigma_{\mu\nu}$ ("ideal relaxation"), then the time dependence of the currents is determined only by the Hamiltonian H_0 of the dynamically independent electrons without dissipation. The eigenfunctions of this Hamiltonian are mostly known and therefore Eq. 51 may be calculated explicitly. For electrons in the presence of a magnetic field,taking the one-electron version of Eq. 51 (see Eq. 33)using the well-known Landau functions and taking into account that

$$\mathbf{i} = e\frac{d\mathbf{x}}{dt} = \frac{e}{m}\left(\mathbf{p} - \frac{e}{c}\mathbf{A}\left(x\right)\right)$$

after relatively simple calculation we get

$$\sigma_{zz}\left(\omega\right) = \frac{Ne^2}{m}\frac{1}{i\omega + \tau^{-1}}$$

$$\sigma_{\mu\nu}\left(\omega\right) = \sigma_{\mu\nu}\left(\omega\right) = \frac{Ne^2}{m}\left(\frac{1}{i\left(\omega - \omega_0\right) + \tau^{-1}} + \frac{1}{i\left(\omega + \omega_0\right) + \tau^{-1}}\right) \tag{52}$$

$$\sigma_{xy}\left(\omega\right) = -\sigma_{yx}\left(\omega\right) = \frac{Ne^2}{m}\left(\frac{1}{i\left(\omega - \omega_0\right) + \tau^{-1}} - \frac{1}{i\left(\omega + \omega_0\right) + \tau^{-1}}\right)$$

This result coincides with the classical one.

Since in the absence of a dissipation ($\tau \to \infty$) one must conclude , that an expansion in powers of the coupling to the dissipative interaction is forbidden (see for example Eq. 47). To conclude, we may say, that generally speaking the conductivity cannot be calculated by a simple expansion in the powers of the coupling constant to the dissipation, not even in the case of a very weak coupling. Nevertheless there are some cases when such an expansion may work. As it may be seen, this happens for $\omega\tau \gg 1$ for the longitudinal conductivity and $|\omega \pm \omega_0|$ for the transverse ones. Also precisely in the cases of strong magnetic fields and high frequencies where the boltzmannian approach fails (except the case of the cyclotron resonance). In the next Section we shall give an example of such a perturbational calculus for the static magneto-resistance.

As a final remark: although the "ideal relaxation" is just a mathematical, non realistic model, the conclusions we derived from are valid for realistic systems, as it may be shown by direct application of perturbation theory.

7 The transverse magneto-resistance

Let us consider a system of electrons in a homogeneous magnetic field $\mathcal{H}$ oriented along the z axis, interacting with phonons. The Hamiltonian of the whole system is

$$\mathbf{H} = \mathbf{H}_e + \mathbf{H}_{ph} + \mathbf{H}_{e-ph} \ , \tag{53}$$

where the Hamiltonian of the independent electrons is

$$\mathbf{H}_e = \frac{1}{2m} \int d\mathbf{r} \psi^+ (\mathbf{r}) \left(-ih\nabla + \frac{e}{c} \vec{A} (\mathbf{r}) \right)^2 \quad (\mathbf{r}) \tag{54}$$

with

$$\vec{A} (\mathbf{r}) = (0, \mathcal{H} x, 0) \ \ .$$

Here we denoted by $\mathbf{H}_{ph}$ the Hamiltonian of the free phonons, while $\mathbf{H}_{e-ph}$ term represents the interaction between the electrons and phonons.

The Kubo formula for the static transverse conductivity is then

$$\sigma_{xx} = e^2 \int_0^{\infty} dt e^{-st} \int_0^{\beta} d\lambda \left\langle \dot{\mathbf{x}} (-ih\lambda) \, \dot{\mathbf{x}} (t) \right\rangle_0 \tag{55}$$

where we inserted

$$\mathbf{j}_x = e \int d\vec{r} \psi^+ (\vec{r}) \, \dot{x} \psi (\vec{r}) = e \dot{\mathbf{x}} \ \ .$$

On the other hand, we may separate the electron motion on the plane transverse to the magnetic field into the cyclotron center motion and the relative one

$$\dot{\mathbf{x}} = \dot{\mathbf{X}} + \dot{\xi} \tag{56}$$

where

$$\mathbf{X} = \frac{h}{m\omega_0} \frac{1}{i} \frac{\partial}{\partial y} = \frac{\hbar_y}{m\omega_0} \mathbf{k}; \qquad \omega_0 = \frac{e\mathcal{H}}{mc} \tag{57}$$

is the operator of the cyclotron motion along the x axis.

We recall here, that the eigenfunctions of the electron motion in a homogeneous magnetic field are the so called Landau functions

$$_{n,X,k_z} = \phi_n(x - X) e^{\imath(\frac{m\omega_0}{\hbar} X y + k_z z)} \ ,$$

with the eigenenergies

$$E_{n,X,k_z} = \hbar\omega_0 (n + \frac{1}{2}) + \frac{\hbar^2 k_z^2}{2m}$$

and the functions $\phi_n(x)$ being the eigenfunctions of the one-dimensional oscillator. The meaning of the operator $\mathbf{X}$ follows immediately.

We have

$$\sigma_{xx} = e^2 \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \left\langle \dot{\mathbf{X}} (-ih\lambda) \dot{\mathbf{X}} (t) \right\rangle_0 \tag{58}$$

$$+ \; e^2 \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \left\langle \dot{\mathbf{X}} (-ih\lambda) \dot{\xi} (t) \right\rangle_0 \tag{59}$$

$$+ \; e^2 \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \left\langle \dot{\xi} (-ih\lambda) \dot{\mathbf{X}} (t) \right\rangle_0 \tag{60}$$

$$+ \; e^2 \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \left\langle \dot{\xi} (-ih\lambda) \dot{\xi} (t) \right\rangle_0 \quad . \tag{61}$$

If we admit, that contrary to $\mathbf{X}$, the ξoperator is bounded, then it is meaningful to perform the time integration of $\dot{\xi}$. For example, the second term in Eq.58 thereafter looks as

$$\int_0^\beta d\lambda \left\langle \dot{\mathbf{X}} (-ih\lambda) \xi (\infty) - \dot{\mathbf{X}} (-ih\lambda) \xi (0) \right\rangle_0$$

and taking into account that correlations over infinite time have to vanish, we get further

$$\int_0^\beta d\lambda \left\langle \dot{\mathbf{X}} (-ih\lambda) \xi (\infty) \right\rangle_0 - \frac{e^2}{\imath\hbar} Tr \left\{ [\rho_{\mathbf{0}}, \mathbf{X}] \, \xi \right\}$$

$$e^2 \beta \langle \dot{\mathbf{X}} \rangle_0 \langle \xi(\infty) \rangle_0 - \frac{e^2}{\imath\hbar} \langle [\xi, \mathbf{X}] \rangle_0 = 0$$

since $\langle \dot{\mathbf{X}} \rangle_0 = 0$; $[\xi, \mathbf{X}] = \mathbf{0}$.

In the same way one may show that all the terms in Eq. 58 containing ξ vanish, if this is bounded. Thus, under this plausible assumption

$$\sigma_{xx} = e^2 \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \left\langle \dot{\mathbf{X}} (-ih\lambda) \dot{\mathbf{X}} (t) \right\rangle_0$$

emerges. Or in a more convenient form for applications (see Eq. 23)

$$\sigma_{xx} = \frac{1}{2} e^2 \beta \int_{-\infty}^\infty dt \left\langle \dot{\mathbf{X}} (0) \dot{\mathbf{X}} (t) \right\rangle_0 \quad .$$

On the other hand, since $\mathbf{X}$ commutes with $\mathbf{H}_e$and $\mathbf{H}_{ph}$, $\dot{\mathbf{X}}$ is of first order in the electron-phonon interaction potential. It follows, that an expansion in powers of this interaction starts directly with a second order term. If we are content with this first non-vanishing approximation, we have

$$\sigma_{xx} = \frac{1}{2} e^2 \beta \int_{-\infty}^{\infty} dt Tr \left\{ \dot{\mathbf{X}} e^{\frac{\imath}{\hbar}(\mathbf{H}_e + \mathbf{H}_{ph})t} \dot{\mathbf{X}} e^{-\frac{\imath}{\hbar}(\mathbf{H}_e + \mathbf{H}_{ph})t} \right\} \quad . \tag{62}$$

Using the occupation number representation of the Landau states (denoting the three quantum numbers n, k_z, X by ν) we have

$$\mathbf{X} = \sum_{\nu} X \mathbf{a}_{\nu}^{+} \mathbf{a}_{\nu}, \qquad \mathbf{H}_{e-ph} = \sum_{\nu,\nu'} U_{\nu,\nu'} \mathbf{a}_{\nu}^{+} \mathbf{a}_{\nu'} \quad ,$$

where $U_{\nu,\nu'}$ is an operator in the space of phonon states. Thus

$$\begin{aligned} \dot{\mathbf{X}} &= \frac{1}{\imath\hbar} [\mathbf{H}, \mathbf{X}] = \frac{1}{\imath\hbar} [\mathbf{H}_{e.ph}, \mathbf{X}] = \sum_{\nu,\nu',\nu''} U_{\nu',\nu''} X \left[\mathbf{a}_{\nu'}^{+} \mathbf{a}_{\nu''}, \mathbf{a}_{\nu}^{+} \mathbf{a}_{\nu} \right] \qquad (63) \\ &= \sum_{\nu,\nu',\nu''} U_{\nu',\nu''} X \left(\delta_{\nu,\nu''} \mathbf{a}_{\nu'}^{+} \mathbf{a}_{\nu} - \delta_{\nu',\nu} \mathbf{a}_{\nu}^{+} \mathbf{a}_{\nu''} \right) = \sum_{\nu\nu'} U_{\nu'\nu} (X - X') \mathbf{a}_{\nu'}^{+} \mathbf{a}_{\nu} \quad . \end{aligned}$$

After introducing it in Eq.62 end performing the trace in the space of the phonon states, characterized by the quantum number s and performing the time-integral we find

$$\begin{aligned} \sigma_{xx} = \frac{\pi e^2 \beta}{\hbar} & \sum_{\nu 1,\nu 2,\nu 3,\nu 4} \sum_{s,s'} \rho_0^{ph}(\varepsilon_s) \langle \mathbf{a}_{\nu_1}^{+} \mathbf{a}_{\nu_2} \mathbf{a}_{\nu_3}^{+} \mathbf{a}_{\nu_4} \rangle_0^0 \left(X_1 - X_2 \right) \left(X_3 - X_4 \right) \\ & \times U_{\nu_1,s;\nu_2,s'} U_{\nu_3,s';\nu_4,s} \delta \left(E_{\nu_4} + \varepsilon_s - E_{\nu_3} - \varepsilon_{s'} \right) \quad . \end{aligned}$$

However,

$$\begin{aligned} \langle \mathbf{a}_{\nu_1}^{+} \mathbf{a}_{\nu_2} \mathbf{a}_{\nu_3}^{+} \mathbf{a}_{\nu_4} \rangle_0^0 &= Tr \left\{ e^{-\beta\left(\sum_{\nu}(E_{\nu}-\mu)\mathbf{a}_{\nu}^{+}\mathbf{a}_{\nu} - \Omega\right)} \mathbf{a}_{\nu_1}^{+} \mathbf{a}_{\nu_2} \mathbf{a}_{\nu_3}^{+} \mathbf{a}_{\nu_4} \right\} \qquad (64) \\ &= \delta_{\nu_3\nu_2} \delta_{\nu_1,\nu_4} f_0(E_{\nu_1}) \left(1 - f_0(E_{\nu_2})\right) + \delta_{\nu_1\nu_2} \delta_{\nu_{31},\nu_4} f_0(E_{\nu_1}) f_0(E_{\nu_3}), \end{aligned}$$

with $f_0(E)$ being the Fermi function. Therefore

$$\begin{aligned} \sigma_{xx} &= \frac{\pi e^2 \beta}{\hbar} \sum_{\nu,\nu'} f_0(E_{\nu}) \left(1 - f_0(E_{\nu'})\right) \left(X - X'\right)^2 \qquad (65) \\ & \times \sum_{s,s'} \rho_0^{ph}(\varepsilon_s) |U_{\nu,s;\nu',s'}|^2 \delta \left(E_{\nu} + \varepsilon_s - E_{\nu'} - \varepsilon_{s'} \right) \quad . \qquad (66) \end{aligned}$$

or

$$\sigma_{xx} = e^2\beta \sum_{\nu,\nu'} f_0(E_\nu)\,(1 - f_0(E_{\nu'}))\,(X - X')^2\, W_{\nu;\nu'}$$

with $W_{\nu;\nu'}$being the average transition rate for the electron from state ν to state ν'due to the interaction with the phonons. If we neglect the phonon energies, or if we had instead of the phonons static defects, we had obtained

$$\sigma_{xx} = -e^2 \sum_{\nu,\nu'} \frac{\partial f_0(E_\nu)}{\partial E_\nu}\,(X - X')^2\, W_{\nu;\nu'}$$

These results [8]-[10] coincide also with those obtained by Titeica [11] in 1935 by a kinetic approach interpreting the transverse conductivity as the migration of the center of cyclotron motion.*An english version of this paper may be found in "Şerban Ţiţeica, Modificarea rezistenţei electrice a metalelor în camp magnetic". p. 49, Ed. Horia Hulubei (2018)*

We have seen, that in the exact formulation of the theory, this assumption could be formulated as the boundedness of the relative coordinate. A check of the above hypothesis has been verified on various definite cases by taking into account also the terms we neglected.

8 Strong coupling problems and strong electric fields

In the preceding Section we have dealt with a relatively simple example of calculating the electric conductivity by perturbation theory with regard to the dissipative potential. An important class of conductivity problems in (crystalline)solids allow such an approach , at least by an improved version of perturbation theory, even in cases where an expansion of the conductivity in powers of the dissipating potential is not allowed (like the one treated in Section 5). The application of such a method actually depends on the possibility to separate a part of the system Hamiltonian whose eigenstates and eigenvalues are known, but leading alone to a diverging static conductivity and a weak "dissipative" one, ensuring the existence of the stationary flow. The stronger the dissipation is, farther we have to go with improving the perturbation method. Obviously, there is a certain strength of this interaction, where such method does not work any more. This happens by amorphous substances, by impurity band conduction and always by very low mobilities.

Even in these cases the exact formula of the conductivity may be useful. The method of its evaluation consist in simple, plausible assumptions about the nature of the energy spectrum and the energy dependence of the matrix elements of the currents (see for example [12], [13]).

The elaboration of the conductivity theory has clarified the foundation and limits of the old kinetic theory, allowed the refinement of the old results and allowed as well completely news results. On the other hand, it has simplified and somewhat unified the approach to transport problems.

A next extension of the conductivity theory is the evaluation of higher order kinetic coefficients, namely of the higher order terms in the expression of the average current. For this sake we have just to consider the next iterative terms of the integral equation Eq. 7. As it is easy to see, we get

$$\rho(t) = \rho_0 + \sum_{n=1}^{\infty} e^{\imath n\omega t}\tilde{\rho}_n$$

where

$$\tilde{\rho}_n = \frac{1}{\imath\hbar}\int_{-\infty}^{0} dt e^{\imath n\omega+s)t} e^{\frac{\imath\mathbf{Ht}}{\hbar}}[\bar{\mathbf{H}}_E, \tilde{\rho}_{n-1}]e^{-\frac{\imath\mathbf{Ht}}{\hbar}}$$

With the help of this expression for the statistical matrix,one may compute the so called higher harmonics, i.e. those terms in the expression of the average current,that oscillate with multiples of the applied field frequency. Taking into account the availability of high intensity monochromatic sources (lasers), such computations got peculiar interest.

In the static case, the exact solution with coupling of the field at $t = 0$ (introducing a relaxation term as in Section 6) may be put in the form

$$\rho = \rho_0 + \frac{1}{\imath\hbar}\int_{-\infty}^{0} dt e^{\imath n\omega+s)t} e^{\frac{\imath(\mathbf{H}+\mathbf{H}_E)t}{\hbar}}[\mathbf{H}_E, \rho_0]e^{-\frac{\imath\mathbf{H}+\mathbf{H}_E)t}{\hbar}}$$

This equation however, needs to be brought in a usable form by an expansion in powers of the electric field.

A very special case is the one, where the intensity of the field is so strong, that no expansion in powers of the field is allowed. We have in mind the case of so called "hot electrons" for which $j \sim \sqrt{E}$. This problem got until now no adequate quantum mechanical treatment (although a formal procedure to improve the perturbation method with respect to the electric field [17].

9 Derivation of the "non-mechanical" transport coefficients

We have reviewed in the preceding Sections the proper formulation of the electric conduction problem. Kubo's exact formula relates the conductivity tensor to the correlation function of the currents in the equilibrium state. It is only to expect a generalization of this theory to other kinetic coefficients, characteristic for the linear response of a system to gradients of concentration and temperature. Based on the quasi-classical results, we may expect that these coefficients might be expressed by equilibrium correlations of the corresponding currents. The proof of such a statement, however is not at all simple, since in the case of these purely macroscopic "forces" a mechanical formulation of the problem is not possible. In other words, gradients of temperature or concentration cannot be introduced in a microscopical Hamiltonian. Therefore, the derivation of such formulas (although followed immediately after that of the electric conductivity) needed a lot of assumptions, which were actually equivalent the postulating the relationships. This situation throw a shadow of doubt to the very existence of a generalization of Kubo's theory. Only recently offered Luttinger [16] a simple, but convincing proof that confirms their validity. In what follows, we describe shortly his argumentation.

The diffusion coefficient usually may be computed using Einstein's relation relating it to the electric conductivity. On the other hand, it may be shown an analogous relation between the thermal conductivity and the response coefficient to a fictive gravitation field coupled to the energy density. This "mechanic" response however one may be calculated. In this way one may avoid the problems.

We define the coefficients $L^{(i)}_{\mu\nu}, \tilde{L}^{(i)}_{\mu\nu}, \quad (i = 1, \dots 4)$ by the phenomenological relationships for the average electric and energy currents:

$$\begin{aligned} \langle j_\mu \rangle &= L^{(1]}_{\mu\nu} \partial_\nu \phi - \tilde{L}^{(1)}_{\mu\nu} \frac{T}{e} \partial_\nu (\frac{\mu}{T}) - \tilde{L}^{(2)}_{\mu\nu} T \partial_\nu (\frac{1}{T}) + L^{(2]}_{\mu\nu} \partial_\nu && (67) \\ \langle j^E_\mu \rangle &= L^{(3]}_{\mu\nu} \partial_\nu \phi - \tilde{L}^{(3)}_{\mu\nu} \frac{T}{e} \partial_\nu (\frac{\mu}{T}) - \tilde{L}^{(4)}_{\mu\nu} T \partial_\nu (\frac{1}{T}) + L^{(4]}_{\mu\nu} \partial_\nu && (68) \end{aligned}$$

where ϕ and are the electric and the gravitational potentials. We have chosen as independent macroscopic variables the inverse temperature $\frac{1}{T}$ and the ratio $\frac{\mu}{T}$ of the chemical potential to the temperature.

The coefficients $L^{(i)}_{\mu\nu}, \quad (i = 1, \ldots 4)$ may be calculated by Kubo's method, considering the interaction with the applied external fields (within the second quantization description) as

$$\mathbf{F} = \int d\vec{x}\,\{\mathbf{n}(\vec{x})\phi(\vec{x}) + \mathbf{h}(\vec{x})\psi(\vec{x})\}\ ,$$

where the first term represents the electrostatic energy, with $\mathbf{n}(\vec{r})$ being the particle density operator, while the second one is the gravitational energy, with $\mathbf{h}(\vec{r})$ being the energy density. The similar computation by the method of statistical matrix of Section 1, gives rise to

$$\begin{aligned} L^{(1)}_{\mu\nu} &= \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \langle \mathbf{j}_\nu(t - \imath\hbar\lambda)\mathbf{j}_\mu(0)\rangle_0\,, \qquad (69)\\ L^{(2)}_{\mu\nu} &= \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \langle \mathbf{j}^E_\nu(t - \imath\hbar\lambda)\mathbf{j}_\mu(0)\rangle_0\,,\\ L^{(3)}_{\mu\nu} &= \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \langle \mathbf{j}_\nu(t - \imath\hbar\lambda)\mathbf{j}^E_\mu(0)\rangle_0\,,\\ L^{(4)}_{\mu\nu} &= \int_0^\infty dt e^{-st} \int_0^\beta d\lambda \langle \mathbf{j}^E_\nu(t - \imath\hbar\lambda)\mathbf{j}^E_\mu(0)\rangle_0\,. \end{aligned}$$

where $\mathbf{j}^E_\nu$ is the operator of the energy current (satisfying the continuity equation $\dot{\mathbf{h}}(\vec{r}) + \nabla\mathbf{j}^E(\vec{r}) = 0$).

We shall show, that for all $i = 1, \ldots 4$ we get $\tilde{L}^{(i)}_{\mu\nu} = L^{(i)}_{\mu\nu}$. To this end we consider the system in equilibrium ($\langle\mathbf{j}_\mu\rangle = \langle\mathbf{j}^E_\mu\rangle = 0$) in the presence of an electric , respectively gravitational field and we calculate the gradients $\partial_\mu(\frac{\mu}{T}, \quad \partial_\mu(\frac{1}{T})$ produced in order to compensate the gradients of the potentials. Therefore we require equilibrium in the presence of the fields. For the quantum statistical computation we express first these gradients by the gradients of the particle and energy densities:

$$\begin{aligned} \partial_\nu\left(\frac{\mu}{T}\right) &= \frac{\partial\left(\frac{\mu}{T}\right)}{\partial\langle n\rangle}\partial_\nu\langle n\rangle + \frac{\partial\left(\frac{\mu}{T}\right)}{\partial\langle h\rangle}\partial_\nu\langle h\rangle \qquad (70)\\ \partial_\nu\left(\frac{1}{T}\right) &= \frac{\partial\left(\frac{1}{T}\right)}{\partial\langle n\rangle}\partial_\nu\langle n\rangle + \frac{\partial\left(\frac{1}{T}\right)}{\partial\langle h\rangle}\partial_\nu\langle h\rangle\,. \end{aligned}$$

Let us consider first equilibrium in the presence of just an electric field

$$\mathbf{F} = \int d\vec{x}\,\{\mathbf{n}(\vec{x})\phi(\vec{x})\}\ ,$$

21

with the equilibrium particle density

$$\langle \mathbf{n}(\vec{r})\rangle = Tr\{e^{-\beta(\mathbf{H}+\mathbf{F}-\mu\mathbf{N}-\Omega)}\mathbf{n}(\vec{r})\} \quad .$$

Using the expansion

$$e^{-\beta(\mathbf{H}+\mathbf{F}-\mu\mathbf{N})} = e^{-\beta(\mathbf{H}-\mu\mathbf{N})}\left\{1-\int_0^\beta d\lambda \mathbf{F}(-\imath\hbar\lambda)+...\right\} ,$$

we get

$$\begin{aligned}&\langle \mathbf{n}(\vec{r})\rangle = \langle \mathbf{n}(\vec{r})\rangle_0 \\ &-\int_0^\beta d\lambda \int d\vec{r}'\left(\langle \mathbf{n}(\vec{r}', -\imath\hbar\lambda)\mathbf{n}(\vec{r})\rangle_0 - \langle \mathbf{n}(\vec{r}', -\imath\hbar\lambda)\rangle_0 \langle \mathbf{n}(\vec{r})\rangle_0\right)\phi(\vec{r}') \,.\end{aligned}$$

Where the symbol $\langle \ldots \rangle$ means $\frac{Tr\{e^{-\beta(\mathbf{H}-\mu\mathbf{N})}...\}}{Tr\{e^{-\beta(\mathbf{H}-\mu\mathbf{N})}\}}$.

Taking into account the macroscopic homogeneity in the absence of the field, by a partial integration we get

$$\partial_\nu \langle n(\vec{r})\rangle = \tag{71}$$

$$-e\int_0^\beta d\lambda \int d\vec{r}' \partial_\nu \left(\langle \mathbf{n}(\vec{r}', -\imath\hbar\lambda)\mathbf{n}(\vec{r})\rangle_0 - \langle \mathbf{n}(\vec{r}', -\imath\hbar\lambda)\rangle_0 \langle \mathbf{n}(\vec{r})\rangle_0\right)\phi(\vec{r}') \tag{72}$$

$$= -e\int_0^\beta d\lambda \int d\vec{r}'\left(\langle n(\vec{r}', -\imath\hbar\lambda)n(\vec{r})\rangle_0 - \langle n(0, -\imath\hbar\lambda)\rangle_0 \langle n(0)\rangle_0\right) e\partial_\nu\phi(\vec{r}'). \tag{73}$$

(Here by neglecting the surfaced integration terms the vanishing of long range correlations was admitted.) If the field does not vary too rapidly with the coordinate, taking into account that the operator of the total particle number commutes with the Hamiltonian, we have further

$$\partial_\nu \langle \mathbf{n}(\vec{r}\rangle = -\frac{e}{VkT}\left(\langle \mathbf{N}^2\rangle_0 - \langle \mathbf{N}\rangle_0^2\right)\partial_\nu\phi(\vec{r}). \tag{74}$$

In the same way one may get also

$$\partial_\nu \langle \mathbf{h}(\vec{r})\rangle = -\frac{e}{VkT}\left(\langle \mathbf{H}\mathbf{N}\rangle_0 - \langle \mathbf{H}\rangle_0\langle \mathbf{N}\rangle_0\right)\partial_\nu\phi(\vec{r}). \tag{75}$$

However, as it may be seen easily

$$\left(\langle \mathbf{N}^2\rangle_0 - \langle \mathbf{N}\rangle_0^2\right) = \frac{\partial\langle \mathbf{N}\rangle_0}{\partial\beta\mu} = V\frac{\partial\langle \mathbf{n}(\vec{r})\rangle_0}{\partial\beta\mu} \tag{76}$$

and

$$\left(\langle \mathbf{NH}\rangle_0 - \langle \mathbf{N}\rangle_0\langle \mathbf{H}\rangle_0\right) = \frac{\partial\langle \mathbf{H}\rangle_0}{\partial\beta\mu} = V\frac{\partial\langle \mathbf{h}(\vec{r})\rangle_0}{\partial\beta\mu}\ . \tag{77}$$

Therefore, from Eq. 70 and Eqs. 74 - 77 it follows

$$\begin{aligned} \partial_\nu\left(\frac{\mu}{T}\right) &= -\left[\frac{\partial\left(\frac{\mu}{T}\right)}{\partial\langle n\rangle}\frac{\partial\langle n\rangle}{\partial\left(\frac{\mu}{T}\right)} + \frac{\partial\left(\frac{\mu}{T}\right)}{\partial\langle h\rangle}\frac{\langle\partial\langle h\rangle}{\partial\left(\frac{\mu}{T}\right)}\right]\frac{e}{T}\partial_\nu\phi \\ &= -\frac{d\left(\frac{\mu}{T}\right)}{d\left(\frac{\mu}{T}\right)}\frac{e}{T}\partial_\nu\phi = -\frac{e}{T}\partial_\nu\phi \end{aligned} \tag{78}$$

and as it is to be expected

$$\begin{aligned} \partial_\nu\left(\frac{1}{T}\right) &= -\left[\frac{\partial\left(\frac{1}{T}\right)}{\partial\langle n\rangle}\frac{\partial\langle n\rangle}{\partial\left(\frac{\mu}{T}\right)} + \frac{\partial\left(\frac{1}{T}\right)}{\partial\langle h\rangle}\frac{\partial\langle h\rangle}{\partial\left(\frac{1}{T}\right)}\right]\frac{e}{T}\partial_\nu\phi \\ &= -\frac{d\left(\frac{1}{T}\right)}{d\left(\frac{\mu}{T}\right)}\frac{e}{T}\partial_\nu\phi = 0\ . \end{aligned} \tag{79}$$

From the conditions $\langle j_\mu\rangle = \langle j^E_\mu\rangle = 0$, together with Eqs.78, 79 we get immediately

$$\tilde{L}^{(1)}_{\mu\nu} = L^{(1)}_{\mu\nu}, \qquad \tilde{L}^{(3)}_{\mu\nu} = L^{(3)}_{\mu\nu}\,. \tag{80}$$

In the same manner one finds

$$\partial_\nu\left(\frac{\mu}{T}\right) = 0, \qquad \partial_\nu\left(\frac{1}{T}\right) = -\frac{1}{T}\partial_\nu\psi\ ,$$

and therefore also

$$\tilde{L}^{(2)}_{\mu\nu} = L^{(2)}_{\mu\nu} \qquad \tilde{L}^{(4)}_{\mu\nu} = L^{(4)}_{\mu\nu}\,. \tag{81}$$

The proof is over and we got finally the whole table of transport coefficients. One might also check Onsager's reciprocity relations.

The calculus of the coefficients $L^{(i)}_{\mu\nu}$, $(i = 1, \ldots 4)$ may be performed according to the same recipes as that of the electric conductivity $L^{(1)}_{\mu\nu}$. The only complication is that the expression of the energy current operator in the case of interacting particles is not that simple as that of the particle current.

10 Appendix. *Derivation of the Boltzmann equation for elastic scattering.*

Let us consider an dynamically independent electron system scattered on s static potential $V(\vec{r})$. After the application of a homogeneous electric field, according to Sec. 4, the equation of motion of the one-electron density matrix is

$$\imath\hbar\frac{\partial \mathbf{f}}{\partial t} = \left[-\frac{\hbar^2}{2m}\nabla^2 + V(\vec{r}) + exE_x e^{st}, \mathbf{f}\right] \tag{82}$$

or in matrix elements of the free electron states

$$\begin{aligned} \imath\hbar\frac{\partial f_{kk'}}{\partial t} &= (\varepsilon_{k'} - \varepsilon_k) f_{kk'} - \imath e E_x e^{st}\left(\frac{\partial}{\partial k_x} + \frac{\partial}{\partial k'_x}\right) f_{kk'} && (83) \\ &+ \sum_q [V_{kq} f_{qk'} - f_{kq} V_{qk'}) && (84) \end{aligned}$$

We consider, that the system at $t = -\infty$ was in equilibrium:

$$\mathbf{f} = \mathbf{f}^{(0)} + \mathbf{f}^{(1)} e^{st} + \ldots$$

with

$$\mathbf{f}^{(0)} = \frac{1}{e^{\beta(-\frac{\hbar^2}{2m}\nabla^2 + V(\vec{r}) - \mu)} + 1}$$

and $\mathbf{f}^{(1)}$ being of first order in the applied field. If in Eq. 83 we retain only terms linear in E_x, then

$$\begin{aligned} \imath\hbar s f^{(1)}_{kk'} &= (\varepsilon_{k'} - \varepsilon_k) f_{kk'} - \imath e E_x e^{st}\left(\frac{\partial}{\partial k_x} + \frac{\partial}{\partial k'_x}\right) f^{(0)}_{kk'} && (85) \\ &+ \sum_q \left[V_{kq} f^{(0)}_{qk'} - f^{(0)}_{kq} V_{qk'}\right) \end{aligned}$$

On the other hand, $f^{(0)}_{kk'}$ obeys the equation

$$\frac{\partial \mathbf{f}^{(0)}_{kk'}}{\partial t} = 0 \ .$$

or

$$(\varepsilon_{k'} - \varepsilon_k) f^{(0)}_{kk'} + \sum_q [V_{kq} f_{qk'} - f_{kq} V_{qk'}) = 0$$

Inserting into Eq. 85 we get

$$\begin{aligned} (\imath\hbar s + \varepsilon_k - \varepsilon_{k'}) f^{(1)}_{kk'} &= -\imath e E_x \left(\frac{\partial}{\partial k_x} + \frac{\partial}{\partial k'_x} \right) f^{(0)}_{kk'} \\ &+ \sum_q \left[V_{kq} f^{(1)}_{qk'} - f^{(1)}_{kq} V_{qk'} \right) \end{aligned} \qquad (85)$$

Approximating

$$f^{(0)}_{kk'} \approx f_0(\varepsilon_k) \delta_k k'$$

we get further

$$(\imath\hbar s + \varepsilon_k - \varepsilon_{k'}) f^{(1)}_{kk'} = -\imath e E_x \frac{\partial f_0(\varepsilon_k)}{\partial k_x} \delta_{kk'} + \sum_q \left[V_{kq} f^{(1)}_{qk'} - f^{(1)}_{kq} V_{qk'} \right)$$

We shall look for a solution of Eq. 10 under the assumption $f^{(}_{kk}1) \gg f^{(1)}_{kk'}$ for $k \neq k'$. Then we get as a first approximation for the non-diagonal matrix elements

$$f^{(1)}_{kk'} = \frac{V_{kk'}}{\imath\hbar s + \varepsilon_k - \varepsilon_{k'}} \left(f^{(1)}_{k'k'} - f^{(1)}_{kk} \right) \qquad (83)$$

and after inserting this into the equation for $k = k'$ we have finally

$$e E_x \frac{\partial f_0(\varepsilon_k)}{\partial k_x} = \sum_{k,k'} W_{k'k} \left(f^{(1)}_{kk} - f^{(1)}_{k'k'} \right) \quad , \qquad (83)$$

where

$$W_{k'k} = \frac{2\pi}{\hbar} \left| V_{kk'} \right|^2 \delta(\varepsilon_k - \varepsilon_{k'}) \qquad (83)$$

Equation 83. is nothing else as the linearized Boltzmann equation. Its solution one looks for in the form

$$f^{(1)}_{kk} = e E_x \frac{\partial f_0(\varepsilon_k)}{\partial k_x} \tau(\varepsilon_k) \qquad (83)$$

and finds

$$\tau(\varepsilon_k)^{-1} = \sum_{k'} W_{kk'} (1 - \cos\theta) \qquad (83)$$

Thereafter it follows for the conductivity

$$\sigma_{xx} = -\frac{e^2\hbar^2}{m^2}\frac{1}{(2\pi)^3}\int d\vec{k}\frac{df_0(\varepsilon_k}{d\varepsilon_k}k_x^2\tau(\varepsilon_k). \tag{83}$$

This is identical to Eq. 48. The derivation of the Boltzmann equation for the more general case of inelastic collisions one finds for example in [17].

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IMPORTANCE OF COULOMB EFFECTS IN TRANSPORT PHENOMENA ON LOCALIZED STATES

Although the importance of the Coulomb repulsion between charge carriers on localized states is generally accepted, as, for instance, in the case of statistical distribution of electrons and holes on impurity states in semiconductors, no attention was paid till now to the influence of this effect on the transport properties on localized states. We have in mind especially the very interesting case of hopping conduction in amorphous semiconductors.

Let us consider the following Hamiltonian

$$H = \sum_{j\sigma} \left[(E_j - \mu)\, n_{j\sigma} + \frac{1}{2} U_j\, n_{j\sigma} n_{j-\sigma} \right], \tag{1}$$

$n_{j\sigma}$ being the operator of the number of particles in the state $|j\sigma\rangle$, where σ stands for spin. The second term represents the Coulomb repulsion between two electrons of opposite spins in the same state

$$U_j = e^2 \int d^3r\, d^3r' \, \frac{|\Psi_j(r)|^2 \; |\Psi_j(r')|^2}{|r - r'|} . \tag{2}$$

In principle, in the interaction term, the dielectric properties of the medium should be also taken into consideration. Generally speaking, it is expected that U_j will decrease with delocalization, vanishing for the nonlocalized states.

With the Hamiltonian (1), instead of the usual Fermi distribution function f_o, one obtains for the average number of particles

$$f(\varepsilon_j) = \langle n_{j\sigma} \rangle = \langle n_{j-\sigma} \rangle = \frac{\exp(-\beta\varepsilon_j) + \exp(-\beta(\varepsilon_j + \tilde{\varepsilon}_j))}{1 + 2\exp(-\beta\varepsilon_j) + \exp(-\beta(\varepsilon_j + \tilde{\varepsilon}_j))} , \tag{3}$$

$$\varepsilon_j = E_j - \mu, \;\; \tilde{\varepsilon}_j = E_j + U_j - \mu .$$

At T = OK, the distribution f is a two-step function, vanishing at energies higher than the electron chemical potential μ, being 1/2 down to the hole chemical potential $\tilde{\mu}$, defined by

$$\tilde{\mu} = \mu - U(\tilde{\mu}) \tag{4}$$

and being 1 umder $\tilde{\mu}$. For energy levels which are in the vicinity of μ or $\tilde{\mu}$, so that $|\varepsilon| \ll U$ or $|\varepsilon + U| \ll U$, respectively, eq. (3) is well approximated by the well known functions

$$f \approx f_e = \frac{1}{2} \, \frac{1}{\exp[\beta(\varepsilon - kT \ln 2)] + 1} = \frac{1}{2} f_o \, (\varepsilon - kT \ln 2) \quad , \tag{5}$$

$$1 - f \approx f_h = \frac{1}{2} \, \frac{1}{\exp[\beta(-\tilde{\varepsilon} - kT \ln 2)] + 1} = \frac{1}{2} f_o \, (-\tilde{\varepsilon} - kT \ln 2)$$

which exclude the possibility of double occupancy, the first one for electrons and the second one for holes.

It is worthwhile to mention the importance of this effect for ESR. In such experiments, the response is proportional to the total number of "free" spins, i.e. to the quantity

$$\int dE \; \varrho(E) \left\langle (n_\uparrow - n_\downarrow)^2 \right\rangle = \int_{\tilde{\mu}}^{\mu} dE \; \varrho(E) \tag{6}$$

where $\varrho(E)$ is the density of one-electron states and zero temperature is considered. Eq.(6) indicates that the effect depends on the density of states between the two chemical potentials. Also we can see that a small effect can be due not only to a low density of states but also to a small $\mu - \tilde{\mu}$.

In what follows we shall discuss the influence of the Coulomb repulsion on the transport properties and namely, on the conductivity and thermoelectric power. Both these effects are proportional to the transition rate from an initial to a final state. This depends on the transition probability $\mathcal{P}_{if}$ and on the occupation of these states, i.e. it can be written as

$$\mathcal{R} = \left\langle \mathcal{P}_{if} \left[n^i_\uparrow \; (1 - n^f_\downarrow) + n^i_\downarrow \; (1 - n^f_\uparrow) \right] \right\rangle . \tag{7}$$

If the transport mechanism is the phonon-assisted hopping, then $\mathcal{P}_{if}$ contains, among other things, the Bose distribution function of phonons /1/. In the case of Seebeck effect, the expression (7) must be multiplied also by the mean energy (with respect to the chemical potential) of the states between which the absorption or emission of the phonon takes place.

In disordered systems, the configurational average must also be performed using the proper distribution of the coordinates and energies. In the present paper, we shall show that (excepting the case $U_j = 0$) there is always a low temperature domain in which the Seebeck coefficient is constant.

Depending on the occupancy of the initial and final states, four transitions are allowed which are outlined in Fig.1. For instance, in the case d), the initial and final state before the transition are doubly and singly occupied, respectively.

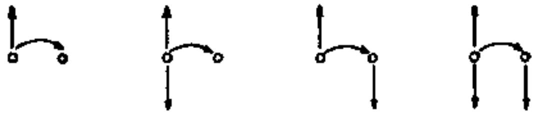

Fig.1a),b),c),d)

If $\mu - \tilde{\mu} \gg kT$, the transition rates, depending on the respective energies and distancies of the four processes are

$$\mathcal{R}_a = \mathcal{P}(\varepsilon_i, \varepsilon_f, r_{if}) \; f_o \; (\varepsilon_i - kT \ln 2) \; f_o(-\varepsilon_f + kT \ln 2), \tag{8}$$

$$\mathcal{R}_b = \mathcal{P}(\tilde{\varepsilon}_i, \; \varepsilon_f, \; r_{if}) \; f_o(\tilde{\varepsilon}_i + kT \ln 2) \; f_o(-\varepsilon_f + kT \ln 2),$$

$$\mathcal{Q}_c = \mathcal{P}(\varepsilon_i, \tilde{\varepsilon}_f, r_{if})\, f_o(\varepsilon_i - kT \ln 2)\, f_o(-\tilde{\varepsilon}_f - kT \ln 2),$$

$$\mathcal{Q}_d = \mathcal{P}(\tilde{\varepsilon}_i, \tilde{\varepsilon}_f, r_{if})\, f_o(\tilde{\varepsilon}_i + kT \ln 2)\, f_o(-\tilde{\varepsilon}_f - kT \ln 2)$$

the corresponding energy factors for the Seebeck coefficient being $\frac{1}{2}(\varepsilon_i + \varepsilon_f)$, $\frac{1}{2}(\tilde{\varepsilon}_i + \varepsilon_f)$, $\frac{1}{2}(\varepsilon_i + \tilde{\varepsilon}_f)$ and $\frac{1}{2}(\tilde{\varepsilon}_i + \tilde{\varepsilon}_f)$, respectively /2/. In these expressions, the occupation factors (the product of the two Fermi functions) are strongly peaked around small values of ε and respectively $\tilde{\varepsilon}$. The shift of the energy arguments by $\pm$ kT ln2 as compared with the case $U_J = 0$ is a very important effect, as we shall see immediately. Indeed, in the absence of the Coulomb repulsion, all four processes are identical and the thermoelectric power α is /3/

$$(9) \qquad \alpha = \frac{e}{kT^2\sigma} \int d\varepsilon_i\, d\varepsilon_f\, \frac{1}{2}\, (\varepsilon_i + \varepsilon_f)\, \tilde{\mathcal{P}}(\varepsilon_i, \varepsilon_f)\, f_o(\varepsilon_i)\, f_o(-\varepsilon_f)$$

while the conductivity is /1/

$$(10) \qquad \sigma = \frac{e^2}{kT} \int d\varepsilon_i\, d\varepsilon_f\, \tilde{\mathcal{P}}(\varepsilon_i, \varepsilon_f)\, f_o(\varepsilon_i)\, f_o(-\varepsilon_f).$$

The function $\tilde{\mathcal{P}}$ was defined as

$$(11) \qquad \tilde{\mathcal{P}}(\varepsilon, \varepsilon') = \int d^3r\, \mathcal{P}(\varepsilon, \varepsilon', r)\, \chi(\varepsilon, \varepsilon', r)\, r^2$$

where the function χ performs the configurational average. Considering that the function $\tilde{\mathcal{P}}(\varepsilon_i, \varepsilon_f)$ is independent of the argument $\varepsilon_i + \varepsilon_f$ within a range of a few kT, i.e. practically

$$(12) \qquad kT \ll \tilde{\mathcal{P}} \Big/ \left. \frac{\partial \tilde{\mathcal{P}}}{\partial(\varepsilon_i + \varepsilon_f)} \right|_{\varepsilon_i + \varepsilon_f = 0}$$

then the integrand in eq. (9) is an odd function of this variable and, consequently, the thermoelectric power vanishes. If the right hand side of (12) is different from zero, we arrive at a contradiction with most experimental results on e.g. amorphous Ge or amorphous Si that seem to indicate a constant value of α at low temperatures.

Coming back to the case $U_j \neq 0$, under the same assumption that $\tilde{\mathcal{P}}(\varepsilon, \varepsilon')$ depends only on $\varepsilon - \varepsilon'$ over a range of a few kT, one may see after a proper change of variables of the type $\varepsilon' = \varepsilon \pm kT \ln 2$, $\tilde{\varepsilon}' = \tilde{\varepsilon} \pm kT \ln 2$ that the processes b) and c) do not contribute to the thermoelectric power at low temperatures, while the contribution of the processes a) and d) is

$$(13) \qquad \alpha_{a,d} = \mp \frac{|e| \ln 2}{T\sigma} \int d\varepsilon\, d\varepsilon'\, \tilde{\mathcal{P}}_{a,d}(\varepsilon - \varepsilon')\, f_o(\varepsilon)\, f_o(-\varepsilon').$$

Therefore, we may write for the Seebeck coefficient at low temperatures

$$S(T) = \frac{\alpha|e|}{k} = \ln 2 \, \frac{\sigma_d(T) - \sigma_a(T)}{\sigma_a(T) + \sigma_b(T) + \sigma_c(T) + \sigma_d(T)} \, . \tag{14}$$

The partial conductivities σ_a, σ_b, σ_c and σ_d have a similar temperature dependence (for example, Mott's famous $T^{-1/4}$ law) but with different parameters, depending on the density of states and localization at μ and $\tilde{\mu}$. Therefore, at sufficiently low temperatures, either one of σ_a or σ_d prevails ($\sigma_{b,c}$ as describing transitions from μ to $\tilde{\mu}$ can never prevail) and then $S \to \pm \ln 2$, or their ratios are constant and

$$S = \zeta \ln 2, \quad -1 < \zeta < 1 \, . \tag{15}$$

The arguments presented here are valid not only for hopping mechanism but also for tunnelling, mechanism that prevails at extremely low temperatures. This means that if both mechanisms are included in σ and S, quite general asymptotic laws may be written as

$$\lim_{T \to 0} \sigma(T) = \sigma_0 \tag{15}$$
$$\lim_{T \to 0} S(T) = \zeta \ln 2$$

i.e. in usual units

$$\lim_{T \to 0} \alpha(T) \approx \zeta \times 0.06 \text{ mV grad}^{-1} . \tag{16}$$

It is interesting to observe that all low temperature plateaus in the hopping region of S in elemental covalent amorphous semiconductors lay in the range (-0.8, 0.25) / 4-9/, in a remarkable agreement with the prediction of our theory ($\ln 2 \approx 0.693$).

The authors would like to thank M.Bundaru for many enlightening discussions at the early stage of this work and Dr.N.Croitoru and Dr.L. Vescan for informations on the experimental results.

Received 25.11.1975.

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Physica 107A (1981) 166–178 *North-Holland Publishing Co.*

MACROSCOPIC BEHAVIOUR OF A CHARGED BOLTZMANN GAS

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Received 27 August 1980

We consider a classical charged gas (with self-consistent Coulomb interaction) described by a solvable linearized Boltzmann equation with thermalization on uniformly distributed scatterers. It is shown that if one scales the time t, the reciprocal space coordinate $\boldsymbol{k}$ and the Debye length l as $\lambda^2 t$, $(1/\lambda)\boldsymbol{k}$, λl, respectively, in the $\lambda \to \infty$ limit the charge density is equal to the solution of the corresponding diffusion-conduction (macroscopic) equation.

1. Introduction

The connection between the kinetic and the hydrodynamic description was illuminatingly discussed for the exactly soluble model of the Lorentz neutral gas[1–3]), for which the L_2-norm of the difference between the two solutions was shown to vanish with $t \to \infty$. On the other hand, a similar problem was tackled in a hopping model both for neutral[4]) and for charged particles with self-consistent interaction[5]). The technique used here was a scaling procedure suggested by the scale invariance of the macroscopic solutions.

This paper is an attempt to apply the latter approach for a charged Boltzmann gas. It seems rather obvious that the Lorentz model, lacking energy dissipation, cannot give rise to the desired macroscopical properties. This was recently discussed in ref.[6]). That is why we consider in this paper a Boltzmann model with thermalization.

The microscopical model is described in section 2 where also the macroscopical equations are shortly discussed. The main results concerning their relationship is stated as well. Some general properties of the model are discussed in detail in section 3. They are used in the proof of the limit theorems given in section 4.

2. Statement of the problem and the results

2.1. *The macroscopic equations*

We consider an infinite, isotropic and homogeneous medium, characterized by the phenomenological constants σ (electric conductivity), D (diffusion constant) and ϵ (electric permitivity). The evolution of the total (free and polarization) internal charge density $\rho_{\text{int}}(\boldsymbol{k}, t)$ in the presence of an external, static charge density $\rho_{\text{ext}}(\boldsymbol{r})$, if one neglects the magnetic field, is given[5]) (in Fourier transform) by

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}, t) = e^{-D(k^2+\epsilon_0/l^2\epsilon)t}[\hat{\rho}_{\text{int}}(\boldsymbol{k}, 0) - \hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty)] + \hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty), \tag{1}$$

where the $t \to \infty$ limit of $\hat{\rho}_{\text{int}}(\boldsymbol{k}, t)$ is

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty) = -\frac{1 + l^2k^2(\epsilon - \epsilon_0)/\epsilon_0}{1 + l^2k^2\epsilon/\epsilon_0}\,\hat{\rho}_{\text{ext}}(\boldsymbol{k}). \tag{2}$$

The characteristic length

$$l = \left(\frac{e^2}{\epsilon_0}\frac{\partial n_0}{\partial \mu}\right)^{-1/2} \tag{3}$$

appears in the Einstein relation

$$\sigma = \frac{\epsilon_0}{l^2} D \tag{4}$$

and is defined only in terms of equilibrium quantities (n_0 – the equilibrium carrier density; μ – the chemical potential).

This phenomenological description is suitable for a semi-conductor, not for a metal, where l is of microscopic size and the diffusion is completely negligible as compared with the conduction. For metals it would be important to consider a medium with boundary, with the external charges placed outside the medium.

If one considers the fundamental solution, defined by point-like initial and external conditions

$$\begin{aligned} &\hat{\rho}_{\text{int}}(\boldsymbol{k}, 0) = e, \\ &\hat{\rho}_{\text{ext}}(\boldsymbol{k}) = q\,e^{i\boldsymbol{k}\boldsymbol{r}_0}, \end{aligned} \tag{5}$$

then we have the following scale invariance property of the solution:

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}/\lambda, \lambda^2 t, \lambda l; \lambda \boldsymbol{r}_0) = \hat{\rho}_{\text{int}}(\boldsymbol{k}, t, l; \boldsymbol{r}_0), \tag{6}$$

which suggests to extract the macroscopic behaviour of the microscopic model through a scaling procedure.

2.2. *The microscopic model*

We consider a system of classical, charged particles in an infinite medium and treat their interaction in a self-consistent way. Their evolution in the presence of a static external charge $\hat{\rho}_{\mathrm{ext}}(\boldsymbol{r})$ is described by the following pair of equations for the distribution function $\varphi(\boldsymbol{r}, \boldsymbol{v}, t)$ and the self-consistent electric field $\boldsymbol{E}(\boldsymbol{r}, t)$:

$$\left(\frac{\partial}{\partial t} + \boldsymbol{v}\nabla_r + \frac{e}{m}\boldsymbol{E}(\boldsymbol{r}, t)\nabla_v\right)\varphi(\boldsymbol{r}, \boldsymbol{v}, t) = (B\varphi)(\boldsymbol{r}, \boldsymbol{v}, t),$$

$$\epsilon_0\nabla_r\boldsymbol{E}(\boldsymbol{r}, t) = en_0\int \mathrm{d}\boldsymbol{v}\varphi(\boldsymbol{r}, \boldsymbol{v}, t) + \rho_{\mathrm{ext}}(\boldsymbol{r}) + \rho_{\mathrm{bg}}. \tag{7}$$

The uniform compensating background charge is, of course,

$$\rho_{\mathrm{bg}} = -en_0. \tag{8}$$

The collision operator B describes a system of uniformly distributed scatterers at thermal equilibrium at temperature T.

$$(B\varphi)(\boldsymbol{r}, \boldsymbol{v}) = \int \mathrm{d}\boldsymbol{v}'[W_{v,v'}\varphi(\boldsymbol{r}, \boldsymbol{v}') - W_{v',v}\varphi(\boldsymbol{r}, \boldsymbol{v})], \tag{9}$$

with the transition rates obeying the detailed balance

$$W_{v,v'}\mathrm{e}^{-mv'^2/2k_{\mathrm{B}}T} = W_{v'v}\mathrm{e}^{-mv^2/2k_{\mathrm{B}}T}. \tag{10}$$

Our model is defined by the choice of the instantly thermalizing scattering cross-section:

$$W_{v,v'} = \frac{1}{\tau}M(v), \tag{11}$$

where $M(v)$ is the normalized Maxwell distribution

$$M(v) = \left(\frac{m}{2\pi k_{\mathrm{B}}T}\right)^{3/2}\mathrm{e}^{-mv^2/2k_{\mathrm{B}}T}, \tag{12}$$

and τ is a constant having dimensions of time.

In the homogeneous case (f and $\boldsymbol{E}^{\mathrm{ext}}$ independent of $\boldsymbol{r}$, implying $\boldsymbol{E} = \boldsymbol{E}^{\mathrm{ext}}$) due to the normalization condition $\int f\mathrm{d}\boldsymbol{v} = 1$, the collision term (9)–(11) reduces to the well known ideal relaxation form $(M - f)/\tau$ yielding

$$en_0\int \mathrm{d}\boldsymbol{v}\boldsymbol{v}f(\boldsymbol{v}, t) = \frac{e^2n_0}{m}\tau\boldsymbol{E} + \mathrm{e}^{-t/\tau}\left[en_0\int \mathrm{d}\boldsymbol{v}\boldsymbol{v}f(\boldsymbol{v}, 0) + \frac{n_0e^2}{m}\boldsymbol{E}(t - \tau)\right].$$

This is an exact linear response for the average velocity, giving the conductivity

$$\sigma = \frac{e^2 n_0}{m}\tau \quad \left(D = \frac{k_B T}{m}\tau\right).$$

It is clear that our system of equations is nonlinear, due to the self-consistent field term in the Boltzmann equation. Therefore, we shall consider only small deviations from the equilibrium solution in the absence of the external charge

$$\varphi_0(\boldsymbol{r}, \boldsymbol{v}) = M(v),$$
$$E_0(\boldsymbol{r}) = 0. \tag{13}$$

We emphasize that we cannot prove the uniqueness of the equilibrium solution, even in the absence of the external charge. Moreover, we perform a "double" linearization, with respect to the deviation from the actual equilibrium solution and with respect to the field as well (low field approximation). We denote

$$f(\boldsymbol{r}, \boldsymbol{v}) = \varphi(\boldsymbol{r}, \boldsymbol{v}) - M(v).$$

Taking the Fourier transform with respect to $\boldsymbol{r}$, we are left with

$$\left(\frac{\partial}{\partial t} + \mathrm{i}\boldsymbol{k}\boldsymbol{v}\right)\hat{f}(\boldsymbol{k}, \boldsymbol{v}, t) - \frac{e}{k_B T}\boldsymbol{v}\hat{\boldsymbol{E}}(\boldsymbol{k}, t)M(v) = B\hat{f}(\boldsymbol{k}, \boldsymbol{v}, t),$$
$$\hat{\boldsymbol{E}}(\boldsymbol{k}, t) = -\frac{\mathrm{i}\boldsymbol{k}}{k^2}\frac{e n_0}{\epsilon_0}\left[\int \mathrm{d}\boldsymbol{v}\hat{f}(\boldsymbol{k}, \boldsymbol{v}, t) + \frac{\hat{\rho}_{\text{ext}}(\boldsymbol{k})}{e n_0}\right]. \tag{15}$$

Now we eliminate $\hat{\boldsymbol{E}}$ and notice that, according to eq. (3)

$$\frac{1}{l^2} = \frac{e^2 n_0}{k_B T \epsilon_0}. \tag{16}$$

From now on, we use $\tau = 1$ and $m/2k_B T = 1$ units. Our equation reads

$$\frac{\partial}{\partial t}\hat{f}(\boldsymbol{k}, \boldsymbol{v}, t) = (-\mathrm{i}\boldsymbol{k}\boldsymbol{v} - 1)\hat{f}(\boldsymbol{k}, \boldsymbol{v}, t) + \left(1 - \frac{\mathrm{i}\boldsymbol{k}\boldsymbol{v}}{k^2 l^2}\right)M(v)\int \mathrm{d}\boldsymbol{v}'\hat{f}(\boldsymbol{k}, \boldsymbol{v}', t)$$
$$- \frac{\mathrm{i}\boldsymbol{k}\boldsymbol{v}}{k^2 l^2}M(v)\frac{\hat{\rho}_{\text{ext}}(\boldsymbol{k})}{e n_0} \tag{17}$$

We consider eq. (17) as an evolution problem in $L_1(\mathrm{R}^3)$ for fixed $\boldsymbol{k}$. The initial and external conditions are chosen to match the macroscopic ones, eqs. (5)

$$f(\boldsymbol{r}, \boldsymbol{v}, 0) = \frac{1}{n_0}\delta(\boldsymbol{r})f_0(\boldsymbol{v}); \quad f_0(\boldsymbol{v}) \in L_1,$$
$$\rho_{\text{ext}}(\boldsymbol{r}) = q\delta(\boldsymbol{r} - \boldsymbol{r}_0), \tag{18}$$

or, equivalently,

$$f(\boldsymbol{k}, \boldsymbol{v}, 0) = \frac{1}{n_0} f_0(\boldsymbol{v}), \quad \hat{\rho}_{\mathrm{ext}}(\boldsymbol{k}) = q e^{i\boldsymbol{k}\boldsymbol{r}_0}. \tag{19}$$

The case of neutral particles may be obtained from eq. (17) by taking $l \to \infty$, the corresponding macroscopic evolution being given by the well-known diffusion equation. Nevertheless, we shall treat it separately, since stronger results can be obtained there.

2.3. *The results*

Our main scaling result concerns the Fourier transform of the microscopical (actual) charge density

$$\rho_{\mathrm{micro}}(\boldsymbol{r}, t) = e n_0 \int \mathrm{d}\boldsymbol{v} f(\boldsymbol{r}, \boldsymbol{v}, t), \tag{20}$$

and reads

$$\lim_{\lambda \to \infty} \hat{\rho}_{\mathrm{micro}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t;\ \lambda l, \lambda \boldsymbol{r}_0\right) = \hat{\rho}_{\mathrm{int}}(\boldsymbol{k}, t;\ l, \boldsymbol{r}_0), \tag{21a}$$

or equivalently

$$\lim_{\lambda \to \infty}\left[\hat{\rho}_{\mathrm{micro}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t; \lambda l, \lambda \boldsymbol{r}_0\right) - \hat{\rho}_{\mathrm{int}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t; \lambda l, \lambda \boldsymbol{r}_0\right)\right] = 0 \tag{21b}$$

for any $f_0(\boldsymbol{v}) \in L_1$. Here the macroscopical charge density $\hat{\rho}_{\mathrm{int}}$ is given by eq. (1) with

$$D = \frac{1}{2}\left(= \frac{k_B T}{m}\tau = \frac{1}{3}\int \mathrm{d}\boldsymbol{v} M(\boldsymbol{v}) \boldsymbol{v}^2 \tau\right)$$

$$\epsilon = \epsilon_0. \tag{22}$$

In the neutral case it is convenient to consider the function

$$f' = \frac{f}{\sqrt{M}}, \tag{23}$$

obeying the equation

$$\frac{\partial}{\partial t} f'(\boldsymbol{r}, \boldsymbol{v}, t) = (-\boldsymbol{v}\nabla_r - 1) f'(\boldsymbol{r}, \boldsymbol{v}, t) + \sqrt{M(\boldsymbol{v})} \int \mathrm{d}\boldsymbol{v}' \sqrt{M(\boldsymbol{v}')} f'(\boldsymbol{r}, \boldsymbol{v}', t)$$

$$\equiv (T + J) f' \equiv A f. \tag{24}$$

Considering this problem in $L_2(\mathrm{R}^3 \times \mathrm{R}^3)$* with initial condition $f_0(\boldsymbol{r}, \boldsymbol{v}) \in$

---

* Considering the problem (24) in $L_2(\mathrm{R}^3 \times \mathrm{R}^3)$ is equivalent to consider the unsymmetrized evolution equation in the weighted Hilbert space $L_2(\mathrm{R}^3 \times \mathrm{R}^3; M(v)^{-1})$.

$L_2(\mathrm{R}^3\times\mathrm{R}^3)\cap L_1(\mathrm{R}^3\times\mathrm{R}^3)$, we obtain the following result

$$\frac{\|n_{\mathrm{micro}}-n\|}{\|n\|}\le C_{f_0}t^{-1/2}, \tag{25}$$

where norms are taken in $L_2(\mathrm{R}^3)$, the microscopical particle density is

$$n_{\mathrm{micro}}(r,t)=n_0\int \mathrm{d}v f(r,v,t), \tag{26}$$

while $n(r,t)$ is the solution of the diffusion equation with D given again by eq. (22) with the initial condition

$$n(r,0)=n_0\int \mathrm{d}v f_0(r,v), \tag{27}$$

C_{f_0} is a constant depending on the initial condition.

We must point out that from the physical point of view it would be desirable to have the scaling property in the coordinate space too and for the neutral case to prove a relation similar to (25) in the Banach space $L_1(\mathrm{R}^3\times\mathrm{R}^3)$.

3. Spectral properties

Let us denote the operators of eq. (17) as follows:

$$\begin{aligned}
&T(k)=-ikv-1,\\
&J(k,l)=\left(1-\frac{ikv}{k^2l^2}\right)M(v)\int \mathrm{d}v'=\psi(k,l;v)\int \mathrm{d}v',\\
&A(k,l)=T(k)+J(k,l).
\end{aligned} \tag{28}$$

For simplicity we shall occasionally omit the arguments k and l.

The free evolution operator $T(k)$ has only continuous spectrum on $\{z|\mathrm{Re}\, z=-1\}$ and generates the non-unitary group

$$U_0(t)=\mathrm{e}^{-(ikv+1)t}. \tag{29}$$

The perturbation $J(k,l)$ is a one-dimensional bounded projector on L_1, and therefore[7]) $A(k,l)$ is the generator of a strongly continuous semigroup.

The resolvent of A is easily seen to be

$$R(z)=(z-A)^{-1}=R_0(z)+\frac{1}{F(z)}R_0(z)JR_0(z), \tag{30}$$

where

$$R_0(z)=(z-T)^{-1} \tag{31}$$

and

$$F(\mathbf{k}, l; z) = 1 - \int \mathrm{d}\mathbf{v}\psi(\mathbf{k}, l; \mathbf{v}) \frac{1}{z + 1 + \mathrm{i}\mathbf{k}\mathbf{v}}. \tag{32}$$

$R(z)$ is bounded in $\mathbb{C}$, except the axis Re $z = -1$ and the zeroes of $F(z)$.

In what follows a detailed discussion of the function $F(z)$ is given. This function can be written as

$$F(z) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} \mathrm{d}x \, \mathrm{e}^{-x^2} \frac{z + \mathrm{i}(1 + 1/k^2l^2)kx}{z + 1 + \mathrm{i}kx}, \tag{33}$$

and is seen to be the weighted mean of the circle which is the image of the real axis through the fractional linear mapping

$$h(x) = \frac{z + \mathrm{i}(1 + 1/k^2l^2)kx}{z + 1 + \mathrm{i}kx}. \tag{34}$$

Thus the value of $F(z)$ lies inside this circle. If the origin is outside, or on the circumference of the circle we have clearly $F(z) \neq 0$. The position of this circle with respect to the origin results from the positions of

$$h^{-1}(\infty) = -\mathrm{i}\frac{z + 1}{k}$$

and

$$h^{-1}(0) = -\mathrm{i}\frac{z}{(1 + 1/k^2l^2)k},$$

with respect to the real axis. It is now obvious that for z outside the strip $\{z | -1 \leq \mathrm{Re}\, z < 0\}$, $F(z)$ cannot vanish.

In order to analyse the occurrence and properties of the zeroes in the strip it is useful to cast $F(z)$ in the form

$$F(z) = 1 + \frac{1}{k^2l^2} - \xi\left(1 + \frac{1}{k^2l^2}\xi\right)\frac{1}{k^2}\int_0^{\infty} \mathrm{d}x \, \frac{\rho(x)}{\xi^2/k^2 + x^2}, \tag{35}$$

where

$$\xi = z + 1$$

and

$$\rho(x) = \frac{2}{\sqrt{\pi}} \mathrm{e}^{-x^2}. \tag{36}$$

Now $F(z) = 0$ is equivalent to

$$\frac{\xi(1+(1/k^2l^2)\xi)}{k^2(1+1/k^2l^2)} = \left[\int_0^\infty \mathrm{d}x \, \frac{\rho(x)}{\xi^2/k^2+x^2}\right]^{-1}. \tag{37}$$

Noticing that the r.h.s. of eq. (37) is a Herglotz function with respect to ξ^2/k^2 we use the well-known representation^{8}) of such functions, as well as the rapid decrease of $\rho(x)$ and its first moments to get

$$\left[\int_0^\infty \mathrm{d}x \, \frac{\rho(x)}{\xi^2/k^2+x^2}\right]^{-1} = \frac{\xi^2}{k^2}+\frac{1}{2}-\int_0^\infty \frac{\mathrm{d}\sigma(u)}{\xi^2/k^2+u}, \tag{38}$$

where $\sigma(u)$ is a non decreasing function.

Therefore we are left with the equations

$$g(\xi) \equiv \xi(\xi-1)+\left(1+\frac{1}{k^2l^2}\right)\left[\frac{1}{2}k^2-k^4\int_0^\infty \frac{\mathrm{d}\sigma(u)}{\xi^2+uk^2}\right]=0. \tag{39}$$

Now we are in the position to prove the following statements concerning the zeroes of $F(z)$ in the right half ($-\frac{1}{2} < \operatorname{Re} z < 0$) of the allowed strip:

(i) for $\operatorname{Re} z > -\frac{1}{2}$ there is at most one solution of eq. (39) which is real;

(ii) there exist k_0, l_0 so that for $k < k_0$ and $l > l_0$ the solution (denoted by $z(k, l)$) exists;

(iii) for $k \to 0$ and $l \to \infty$ (with $kl = \text{const}$) this solution approaches the origin. More precisely

$$\lim_{\lambda\to\infty} x^2 z\left(\frac{1}{\lambda}k, \lambda l\right) = -\frac{1}{2}k^2\left(1+\frac{1}{k^2l^2}\right). \tag{40}$$

A direct inspection shows that $\operatorname{Im} g(\xi) \neq 0$ for $\operatorname{Im} \xi \neq 0$ if $\operatorname{Re} z > -\frac{1}{2}$ which rules out the existence of complex solutions. For ξ real and greater than $\frac{1}{2}$, $g(\xi)$ is strictly increasing, so (i) is proven. If $g(\frac{1}{2}) < 0$ the existence of the solution is obvious because $g(\xi)$ eventually becomes positive. But $g(\frac{1}{2})$ can be made arbitrarily close to $-\frac{1}{4}$ for small k and large l. The scaling result (40) follows immediately from eq. (39).

We remark, though it is not essential, that for the neutral case ($l = \infty$) these results can be extended in the whole strip with $k_0 = \sqrt{\pi}$. For finite l, on the contrary, we can have two solutions and even non-real solutions in the strip for suitable values of the parameters k, l.

4. The scaling limit

4.1. *The charged gas*

We begin by proving the inverse Laplace transform representation for the evolution operator

$$
\begin{aligned}
e^{tA(\mathbf{k},l)}f &= \frac{1}{2\pi \mathrm{i}} \lim_{\delta\to\infty} \int_{\gamma-\mathrm{i}\delta}^{\gamma+\mathrm{i}\delta} e^{zt} R(z) f \, dz \\
&= e^{tT(\mathbf{k})} f + \frac{1}{2\pi \mathrm{i}} \int_{\gamma-\mathrm{i}\infty}^{\gamma+\mathrm{i}\infty} e^{zt} \frac{1}{F(z)} R_0(z) J(\mathbf{k}, l) R_0(z) f \, dz, \qquad (41)
\end{aligned}
$$

for every $f \in L_1(\mathbb{R}^3)$, $\gamma > -\frac{1}{2}$ and greater than the pole of the resolvent. The existence of the integral in (41) is ensured by the Fubini theorem, alongside with

$$
\begin{aligned}
\int d\mathbf{v} \int_{-\infty}^{\infty} dz_2 |e^{zt} R_0 J R_0 f| &\le e^{\gamma t} \int d\mathbf{v} \int d\mathbf{v}' \int dz_2 \frac{|\psi(\mathbf{k}, l; \mathbf{v})|}{|z+1+\mathrm{i}\mathbf{k}\mathbf{v}|} \cdot \frac{|f(\mathbf{v}')|}{|z+1+\mathrm{i}\mathbf{k}\mathbf{v}'|} \\
&\le e^{\gamma t} \frac{\pi}{\gamma+1} \|\psi(\mathbf{k}, l)\| \cdot \|f\|, \qquad (42)
\end{aligned}
$$

where $z = z_1 + \mathrm{i}z_2$ and the last step is the Schwarz inequality for the integration over z_2. Use was made of the boundedness of $F^{-1}(z)$ on the integration path. The result follows now from the analicity of the integrand together with

$$\lim_{z\to\infty} \|R_0(z) J R_0(z) f\| = 0, \qquad (43)$$

which is straightforward.

Taking $\gamma < 0$ we can easily see that

$$\lim_{t\to\infty} \|e^{tA(\mathbf{k},l)} f\| = 0. \qquad (44)$$

In the presence of the zero $z(\mathbf{k}, l)$ of $F(z)$ in the strip $-\frac{1}{2} < \mathrm{Re}\, z < 0$ it is useful to move the integration contour to the left until $-\frac{1}{2} < \gamma < z(\mathbf{k}, l)$.

We have then

$$
\begin{aligned}
e^{tA(\mathbf{k},l)} f &= e^{tT(\mathbf{k})} f + \frac{1}{2\pi \mathrm{i}} \int_{\gamma-\mathrm{i}\infty}^{\gamma+\mathrm{i}\infty} e^{zt} R_0 J R_0 f \, dz \\
&\quad + e^{tz(\mathbf{k},l)} C_{\mathbf{k},l} \, R_0(z(\mathbf{k}, l)) J(\mathbf{k}, l) R_0(z(\mathbf{k}, l)) f, \qquad (45)
\end{aligned}
$$

where

$$C_{k,l} = \left(\int \mathrm{d}v \, \frac{\psi(k,l)}{(z(k,l) + 1 + \mathrm{i}kv)^2} \right)^{-1}, \tag{46}$$

is the residue of $F^{-1}(z)$ in $z(k, l)$.

In the scaling limit $k \to (1/\lambda)k$; $l \to \lambda l$; $\lambda \to \infty$, the first two terms of eq. (45) vanish exponentially. For the last one we have

$$\lim_{\lambda \to \infty} C_{k/\lambda, \lambda l} = 1. \tag{47}$$

$$s - \lim_{\lambda \to \infty} R_0\left(z\left(\frac{k}{\lambda}, \lambda l \right) \right) = 1, \tag{48}$$

$$s - \lim_{\lambda \to \infty} J\left(\frac{k}{\lambda}, \lambda l \right) = M(v) \int \mathrm{d}v' \equiv P, \tag{49}$$

and therefore

$$s - \lim_{\lambda \to \infty} \mathrm{e}^{\lambda^2 t A(k/\lambda, \lambda l)} = \mathrm{e}^{-1/2 k^2 t (1 + 1/k^2 l^2)} P. \tag{50}$$

This concludes the proof of our statement (21) for the particular case $\rho_{\text{ext}} = 0$.

In the presence of an external charge, the solution of eq. (17) is (with the notation justified by eq. (44))

$$\hat{f}(k, v, t) = \mathrm{e}^{tA(k)}[\hat{f}(k, v, 0) - \hat{f}(k, v, \infty)] + \hat{f}(k, v, \infty), \tag{51}$$

where $\hat{f}(k, v, \infty)$ is the time independent solution of the same equation:

$$\begin{aligned} \hat{f}(k, v, \infty) &= -R(0) \frac{\mathrm{i}kv}{k^2 l^2} M(v) \frac{\hat{\rho}_{\text{ext}}(k, r_0)}{en_0} \\ &= -\left[R_0(0) + \frac{1}{F(0)} R_0(0) J R_0(0) \right] \frac{\mathrm{i}kv}{k^2 l^2} M(v) \frac{\hat{\rho}_{\text{ext}}(k, r_0)}{en_0}. \end{aligned} \tag{52}$$

We remark that

$$\lim_{k \to 0} en_0 \int \mathrm{d}v \hat{f}(k, v, \infty) = -\hat{\rho}_{\text{ext}}(0),$$

i.e. the external charge gathers an induced total charge compensating it exactly. The non-conservation of the charge occurs at the expense of the charges and currents at the infinity, just like in the macroscopical description given by eq. (2). For a finite medium, conservation of charge would occur by properly taking into account the boundary conditions.

For proving the scaling it is sufficient to show that in $L_1(\mathbb{R}^3)$ we have

$$\lim_{\lambda\to\infty} \hat{f}\left(\frac{\boldsymbol{k}}{\lambda}, \boldsymbol{v}, \infty; \lambda l, \lambda \boldsymbol{r}_0\right) = -M(v)\frac{1}{1+k^2l^2}\frac{\hat{\rho}_{\text{ext}}(\boldsymbol{k}, \boldsymbol{r}_0)}{en_0}. \tag{53}$$

Indeed, our choice of $\hat{\rho}_{\text{ext}}$, eq. (19), gives

$$\hat{\rho}_{\text{ext}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda \boldsymbol{r}_0\right) = \hat{\rho}_{\text{ext}}(\boldsymbol{k}, \boldsymbol{r}_0), \tag{54}$$

and we have also

$$\lim_{\lambda\to\infty} \lambda^2 \int \mathrm{d}\boldsymbol{v}\, \frac{M(v)}{1+\mathrm{i}\boldsymbol{k}\boldsymbol{v}/\lambda}\frac{\mathrm{i}\boldsymbol{k}\boldsymbol{v}}{\lambda} = \tfrac{1}{2}k^2, \tag{55}$$

$$\lim_{\lambda\to\infty} \lambda^2 F\left(\frac{k}{\lambda}, \lambda l; 0\right) = \lim_{\lambda\to\infty} \lambda^2 \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} \mathrm{e}^{-x^2} \frac{\mathrm{i}\,\frac{k}{\lambda}(1+1/k^2l^2)x}{1+\mathrm{i}(k/\lambda)x}\,\mathrm{d}x = \tfrac{1}{2}k^2\left(1+\frac{1}{k^2l^2}\right). \tag{56}$$

Then, taking the scaling limit in eq. (52), the first term vanishes and the second gives the desired result. This proves the scaling result in the general case.

3.2. *The neutral gas*

It is easy to verify that the spectrum of the transport operator appearing in eq. (24) in only continuous and located on $\{z \mid \operatorname{Re} z = -1\} \cup \{z \mid -1 < z \leq 0\}$. The spectrum of each Fourier component of this transport operator, T_k, contains the continuum $\{z \mid \operatorname{Re} z = -1\}$ and, for $|k| < \sqrt{\pi}$, a real eigenvalue z_k which for small k can be written as

$$z_k \sim -\frac{k^2}{2} - \frac{k^4}{2}\ldots. \tag{57}$$

The corresponding eigenvector is

$$\psi_k(v) = \frac{c_k\sqrt{M(v)}}{z_k + 1 + \mathrm{i}\boldsymbol{k}\boldsymbol{v}}, \quad c_k \sim 1 + \frac{k^2}{4} + \ldots. \tag{58}$$

As $L_2(\mathbb{R}^3; M^{-1}(v))$ is contained in $L_1(\mathbb{R}^3)$ estimations similar to (42, 43) are valid in this case too and we assert that there exists $\epsilon > 0$, $-1 < \beta < 0$ such that for $|k| \geq \epsilon$

$$\mathrm{e}^{t\mathscr{A}_k}f = \lim_{\gamma\to\infty}\frac{1}{2\pi\mathrm{i}}\int_{\beta-\mathrm{i}\gamma}^{\beta+\mathrm{i}\gamma} \mathrm{e}^{tz}(z-\mathscr{A}_k)^{-1}f\,\mathrm{d}z = \mathrm{e}^{tT_k}f + \mathrm{e}^{\beta t}Z(\beta, t)f, \tag{59}$$

where $\|Z\| \leq C$ (independent of t).

For $|k| < \epsilon$, there exists β', $-1 < \beta' < 0$, such that

$$e^{t\mathscr{A}_k} = \lim_{\gamma\to\infty} \frac{1}{2\pi i} \int_{\beta'-i\gamma}^{\beta'+i\gamma} e^{tz}(z - \mathscr{A}_k)^{-1} f \, dz + e^{tz_k}(\bar{\psi}_k, f)\psi_k \tag{60}$$

If we estimate now the norm of the semigroup $\mathscr{U}(t)$ generated by $\mathscr{A}$, we obtain

$$\begin{aligned}\|\mathscr{U}(t)f\|^2 &= \int \|e^{t\mathscr{A}_k}\hat{f}_k\|^2_{L^2(R^3)}\, dk = \int_{|k|\leq\epsilon} \|e^{t\mathscr{A}_k}\hat{f}_k\|^2_{L^2(R^3)}\, dk + \int_{|k|>\epsilon} \|e^{t\mathscr{A}_k}\hat{f}_k\|^2_{L^2(R^3)}\, dk \\ &\leq C^2 e^{2\beta t} + C' e^{2\beta' t} \int \|\hat{f}_k\|^2 \, dk + \left\|\int_0^\epsilon dk\, e^{tz_k}(\bar{\psi}_k, f)\psi_k\right\|^2. \end{aligned} \tag{61}$$

Then,

$$\left\|\mathscr{U}(t)f - \int_0^\epsilon dk\, e^{tz_k}(\bar{\psi}_k, f)\psi_k\right\|^2 \leq \text{exponentially bounded terms.} \tag{62}$$

The integral in eq. (62) can be written $\mathscr{U}^H(t)Q_\epsilon f$, where $\mathscr{U}^H(t)$ is the "full hydrodynamic" semigroup giving the solution of the "full hydrodynamic" equation

$$\frac{\partial n^H}{\partial t} = \tfrac{1}{2}\nabla^2 n^H - \tfrac{1}{2}\nabla^4 n^H \dots \tag{63}$$

and Q_ϵ is the hydrodynamic projector

$$Q_\epsilon = \int_0^\epsilon (\bar{\psi}_k, \cdot)\psi_k \, dk. \tag{64}$$

From eq. (65) it results that

$$\|\mathscr{P}\mathscr{U}(t)f_0 - \mathscr{P}\mathscr{U}^H(t)Q_\epsilon f_0\| = \|\mathscr{P}\mathscr{U}(t)f_0 - \mathscr{U}^H(t)\mathscr{P}Q_\epsilon f_0\|$$

is exponentially bounded in time (here $\mathscr{P} \equiv \int\sqrt{M(v)}\, dv$ and $\mathscr{P}f \equiv n$ is the local density).

Therefore, denoting by $\mathscr{U}^D(t)$ the semigroup generated by $\frac{1}{2}\Delta$ we have

$$\begin{aligned}\|\mathscr{P}\mathscr{U}(t)f_0 - \mathscr{U}^D(t)\mathscr{P}f_0\|^2 &\leq \|\mathscr{P}\mathscr{U}(t)f_0 - \mathscr{U}^H(t)\mathscr{P}Q_\epsilon f_0\|^2 \\ &\quad + \|\mathscr{U}^H(t)\mathscr{P}Q_\epsilon f_0 - \mathscr{U}^H(t)\mathscr{P}f_0\|^2 + \|\mathscr{U}^H(t)\mathscr{P}f_0 - \mathscr{U}^D(t)\mathscr{P}f_0\|^2 \\ &\leq \text{exponentially bounded terms} + C_1 t^{-5/2} + C_2 t^{-7/2}. \end{aligned} \tag{65}$$

Collecting the results and taking into account that $\|\mathscr{U}^D(t)\mathscr{P}f_0\| \geq t^{-3/4}$ we obtain the result (25).

Acknowledgement

The authors are deeply indebted to G.A. Mezincescu for suggesting the Herglotz property argument used in section 3.

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Physica 115A (1982) 169–184 *North-Holland Publishing Co.*

THE MACROSCOPIC ELECTRODYNAMIC BEHAVIOUR OF A SOLUBLE HOPPING MODEL

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Received 11 January 1982

The scaling procedure we proposed in previous papers to obtain the macroscopic behaviour from the microscopic (kinetic) description is applied here to a soluble hopping model, on a periodic lattice, which exhibits both conductive and dielectric properties. The Kubo and the Clausius–Mossotti approaches are discussed in the light of our exact results.

1. Introduction

In refs. 1 and 2, hereafter referred to as I and II respectively, we have proposed a scaling procedure to obtain the macroscopic electrodynamic behaviour from the microscopic (kinetic) description. The approach was suggested by the scale invariance of the macroscopical equations.

This method was successfully applied to a soluble hopping model of charged particles, on an arbitrary periodical lattice (II), and to a charged Boltzmann gas[3]) (both in the self-consistent potential approximation). In both cases the macroscopic behaviour was recovered, describing a medium characterized by a diffusion constant D and a conductivity σ related by the Einstein relation

$$\sigma = \frac{\epsilon_0}{l^2} D, \tag{1.1}$$

where the length

$$l = \left(\frac{e^2}{\epsilon_0} \frac{\partial n_0}{\partial \mu} \right)^{-1/2} \tag{1.2}$$

(n_0 being the equilibrium carrier concentration, μ the chemical potential, e the electron charge and ϵ_0 the permitivity of the vacuum), played an important role in the scaling procedure.

However, neither of these models exhibited dielectric properties, i.e. they had $\epsilon = \epsilon_0$. This is not so surprising since in both cases all the particles were allowed to reach any point of the system. According to Lorentz's original ideas[4]) they should be considered as free or conduction "electrons", whereas dielectric properties arise from the polarisation or bound "electrons".

In this paper we modify the hopping model to provide for bound "electrons" too. Applying again the scaling procedure we confirm Lorentz's ideas in the sense that σ and D are determined by the free "electrons" only while $\epsilon - \epsilon_0 > 0$ by the bound "electrons".

We show also, that our dielectric constant satisfies, in the appropriate limit, the classical Clausius–Mossotti relation.

The outline of the paper is the following: in section 2 we summarize the macroscopic solution and its scale properties. In section 3 we give a brief description of the model. Section 4 is devoted to the study of the solution. Its scaling limit, which is the main result of the paper, is the object of section 5. The final section is left for the discussion of the results. It contains also a detailed comparison, on this example, with the standard dielectric response function theory and with the Clausius–Mossotti relation.

2. The macroscopic description

The macroscopic situation we aim at is that of an infinite homogeneous medium, characterized by the phenomenological tensors σ, D and ϵ, in which we consider at $t = 0$ a certain free charge distribution. We look at its evolution in the field of a static external charge ρ_{ext} (neglecting the internal magnetic field). The corresponding equations are given in II. Their solution is (in Fourier transform and using eq. (1.1))

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}, t) = \mathrm{e}^{-\boldsymbol{k}D\boldsymbol{k}(1+\epsilon_0/l^2\boldsymbol{k}\epsilon\boldsymbol{k})t}[\hat{\rho}_{\text{int}}(\boldsymbol{k}, 0) - \hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty)] + \hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty), \tag{2.1}$$

where

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}, \infty) = -\frac{1 + l^2(\boldsymbol{k}(\epsilon - \epsilon_0)\boldsymbol{k}/\epsilon_0)}{1 + l^2(\boldsymbol{k}\epsilon\boldsymbol{k}/\epsilon_0)}\hat{\rho}_{\text{ext}}(\boldsymbol{k}) \tag{2.2}$$

for the internal charge density and

$$\hat{V}(\boldsymbol{k}, t) = \frac{1}{\epsilon_0 \boldsymbol{k}^2}\hat{\rho}_{\text{tot}}(\boldsymbol{k}, t) = \frac{1}{\epsilon_0 \boldsymbol{k}^2}[\hat{\rho}_{\text{int}}(\boldsymbol{k}, t) + \hat{\rho}_{\text{ext}}(\boldsymbol{k})] \tag{2.3}$$

for the potential (ρ_{tot} being the total charge density).

We remind that in a semiconductor, the density ρ of free charges is defined as

$$\hat{\rho}(\boldsymbol{k}, t) = \hat{\rho}_{\text{int}}(\boldsymbol{k}, t) + \boldsymbol{k}(\epsilon - \epsilon_0)\boldsymbol{k}\hat{V}(\boldsymbol{k}, t). \tag{2.4}$$

In what follows it will be convenient to consider the evolution of the total density in terms of the initial value of the free charge density

$$\hat{\rho}_{\mathrm{tot}}(\boldsymbol{k}, t) = \frac{\epsilon_0 k^2}{\boldsymbol{k}\epsilon\boldsymbol{k}} \left\{ \mathrm{e}^{-\boldsymbol{k}D\boldsymbol{k}(1+\epsilon_0/l^2\boldsymbol{k}\epsilon\boldsymbol{k})t} \left[\hat{\rho}(\boldsymbol{k}, 0) + \frac{1}{1 + l^2(\boldsymbol{k}\epsilon\boldsymbol{k}/\epsilon_0)} \hat{\rho}_{\mathrm{ext}}(\boldsymbol{k}) \right] \right\}$$
$$+ \frac{l^2 k^2}{1 + l^2(\boldsymbol{k}\epsilon\boldsymbol{k}/\epsilon_0)} \hat{\rho}_{\mathrm{ext}}(\boldsymbol{k}). \tag{2.5}$$

Let us consider the fundamental solution, defined by an external point-like charge in $\boldsymbol{x}_0$ and a point-like initial free charge at the origin

$$\hat{\rho}_{\mathrm{ext}}(\boldsymbol{k}) = q^{\mathrm{ext}}\, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{x}_0}, \quad \hat{\rho}(\boldsymbol{k}, 0) = e. \tag{2.6}$$

It can be seen that

$$\hat{\rho}_{\mathrm{tot}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t; \lambda l, \lambda \boldsymbol{x}_0\right) = \hat{\rho}_{\mathrm{tot}}(\boldsymbol{k}, t; l, \boldsymbol{x}_0), \tag{2.7}$$

or, in the coordinate space

$$\lambda^3 \rho_{\mathrm{tot}}(\lambda \boldsymbol{x}, \lambda^2 t; \lambda l, \lambda \boldsymbol{x}_0) = \rho_{\mathrm{tot}}(\boldsymbol{x}, t; l, \boldsymbol{x}_0). \tag{2.8}$$

This scale invariance suggested the application of a "scale projection" on the microscopical charge density

$$\lim_{\lambda \to \infty} \hat{\rho}_{\mathrm{tot}}^{\mathrm{micro}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t; \lambda l, \lambda \boldsymbol{x}_0\right) \tag{2.9}$$

to obtain the macroscopical result.

3. The hopping model

Let us consider an array of sites i, characterized by the coordinates $\boldsymbol{x}_i$ and the energies ϵ_i, which can be occupied by a system of charged fermions, and let

$$W_{ij} = W_{ji}\, \mathrm{e}^{\beta(\epsilon_i - \epsilon_j)} \tag{3.1}$$

be the (equilibrium) transition rates between the sites i and j. Then, the linearized hopping rate equations for the deviation η_i of the average occupation number from its equilibrium value is (I, II)

$$\frac{\mathrm{d}}{\mathrm{d}t} \eta_i(t) = -\sum_j \Gamma_{ij} [\eta_j(t) + e\beta f_j(1 - f_j) \mathcal{V}_j(t)], \tag{3.2a}$$

$$\mathcal{V}_i(t) = \frac{e}{4\pi\epsilon_0} \sum_{j(\neq i)} \frac{\eta_j(t)}{|\boldsymbol{x}_i - \boldsymbol{x}_j|} + \mathcal{V}_i^{\mathrm{ext}}, \tag{3.2b}$$

where $\mathcal{V}_i(t)$ is the self-consistent potential, $\mathcal{V}_i^{\mathrm{ext}}$ is the static external potential f_i is the Fermi function of ϵ_i, and

$$\Gamma_{ij} = M_{ij}[f_j(1-f_j)]^{-1}; \quad M_{ij} = \delta_{ij}\sum_l \mathcal{W}_{il} - \mathcal{W}_{ij}, \quad \mathcal{W}_{ij} = f_i(1-f_j)W_{ij} = \mathcal{W}_{ji}. \qquad (3.3$$

We shall consider this hopping problem on a periodic lattice of sites with arbitrary symmetry and arbitrary elementary cell. The position of the site is given by

$$x_s(r) = r + \xi_s,$$

where r is a vector of the Bravais lattice and ξ_s specifies the position inside the elementary cell. Choosing

$$\epsilon_s(r) = \epsilon_s$$

and

$$W_{ss'}(r, r') = W_{ss'}(r - r')$$

the problem has the translational invariance of the Bravais lattice. It is then natural to consider the whole problem in discrete Fourier transform defined as

$$\tilde{f}(k) = \sum_r \mathrm{e}^{\mathrm{i}kr} f(r); \quad f(r) = \frac{v}{(2\pi)^3}\int_{\mathrm{BZ}} \mathrm{d}k\, \mathrm{e}^{-\mathrm{i}kr}\tilde{f}(k), \qquad (3.4)$$

where v is the volume of the elementary cell and the integration is performed over the Brillouin zone (BZ).

Combining the eqs. (3.2) we have, with matrix notation with respect to the cell indices s and in Fourier transform

$$\frac{\mathrm{d}}{\mathrm{d}t}|\tilde{\eta}(k, t)\rangle = -\tilde{A}(k)|\tilde{\eta}(k, t)\rangle - \frac{\epsilon_0 v}{e}\tilde{\Gamma}(k)\nu|\tilde{\mathcal{V}}^{\mathrm{ext}}(k)\rangle, \qquad (3.5$$

where

$$\nu_{ss'} = \frac{e^2\beta}{\epsilon_0 v} f_s(1-f_s)\delta_{ss'} = \nu_s\delta_{ss'},$$

$$\tilde{A}(k) = \tilde{\Gamma}(k)\tilde{X}(k), \quad \tilde{X}_{ss'}(k) = \delta_{ss'} + \nu_s\tilde{C}_{ss'}(k). \qquad (3.6$$

The matrix $\tilde{C}_{ss'}(k)$ is the Fourier transform of the Coulomb matrix

$$C_{ss'}(r) = \frac{v(1-\delta_{ss'}\delta_{r,0})}{4\pi|r + \xi_s - \xi_{s'}|} \qquad (3.7$$

given explicitly in II. It is important here to recall only that $\tilde{C}(k)$ can be split into a singular term and a well-behaved one,

$$\tilde{C}_{ss'}(k) = \frac{1}{k^2} e^{-ik(\xi_s - \xi_{s'})} + \tilde{R}_{ss'}(k) \tag{3.8}$$

($\tilde{R}(k)$ is indefinitely differentiable and $\tilde{R}(\epsilon k)$ is analytic in ϵ around $\epsilon = 0$ for any $k \in \mathrm{BZ}$ (see II)).

Among the properties of the model, an important part is played by the connectivity structure. Here lies the difference between the previous model (I, II) and the present one, as well as the explanation of their different macroscopic behaviour. A connected component of the system is defined as a maximal set of sites with the property that any pair of its points can be connected by a sequence of allowed (i.e. non zero transition rate) steps. In our previous papers the system consisted of only one connected component, the electrons being allowed to reach any point starting from any point. Now, besides such free electrons which move on an infinite "continent" we consider also bound particles, confined to "islands". Each cell contains one "island" and all "islands" are identical except for a lattice translation. No hop is allowed between different "islands" or between "islands" and the "continent". This way the sites are split into two types: The positions which belong to the free particles are denoted by $x_s(r)$, $s = 1, 2, \ldots, S$ and those corresponding to the bound particles $x_s(r)$, $s = S+1, S+2, \ldots, S+\Sigma$.

In virtue of our classification of the sites it is natural to consider the $S+\Sigma$-dimensional space in which the evolution (3.5) takes place as a (direct) sum of two sub-spaces $\mathcal{S}_f$ (corresponding to the first S values of the index s) and $\mathcal{S}_b$ (corresponding to the remaining Σ values). Therefore we shall use block forms for the matrices whenever appropriate.

In what follows, it is useful to introduce the notations

$$l^{-2} = \sum_{s=1}^{S} \nu_s; \quad L^{-2} = \sum_{s=S+1}^{S+\Sigma} \nu_s; \quad \Lambda^{-2} = l^{-2} + L^{-2}, \tag{3.9}$$

which explicitates eq. (1.2) for the free, bound and respectively total electron density. In the macroscopic picture only l appears.

We define also

$$p_s = \begin{cases} l^2 \nu_s, & \text{for } s \leqslant S, \\ 0, & \text{for } s > S, \end{cases} \quad \text{and} \quad q_s = \begin{cases} 0, & \text{for } s \leqslant S, \\ L^2 \nu_s, & \text{for } s > S, \end{cases} \tag{3.10}$$

which are the components of the vectors denoted by $|p\rangle$ and $|q\rangle$ respectively, as well as the diagonal elements of the diagonal matrices denoted by p and q. We have

$$\nu = \frac{1}{l^2} p + \frac{1}{L^2} q = \begin{pmatrix} p/l^2 & 0 \\ 0 & q/L^2 \end{pmatrix}, \tag{3.11}$$

where we identified the operator p (resp. q, with its restriction to $\mathcal{S}_\mathrm{f}$ (resp. $\mathcal{S}_\mathrm{b}$)). Such identifications will be currently used throughout the paper in order to avoid complicated notations.

It is easily seen that the solution of eq. (3.5) is

$$|\tilde{\eta}(\boldsymbol{k}, t)\rangle = \mathrm{e}^{-\tilde{A}(\boldsymbol{k})t}\left[|\tilde{\eta}(\boldsymbol{k}, 0)\rangle + \frac{v\epsilon_0}{e}\tilde{X}^{-1}\nu|\tilde{\mathcal{V}}^{\mathrm{ext}}(\boldsymbol{k})\rangle\right] - \frac{v\epsilon_0}{e}\tilde{X}^{-1}\nu|\tilde{\mathcal{V}}^{\mathrm{ext}}(\boldsymbol{k})\rangle, \tag{3.12}$$

where the existence of $\tilde{X}^{-1}$, which will be discussed in the next section, was admitted.

We choose again a point-like external charge q^{ext} located at

$$\boldsymbol{x}_0 = \boldsymbol{r}_0 + \boldsymbol{\xi}_0$$

not belonging to our lattice ($\boldsymbol{\xi}_0 \neq \boldsymbol{\xi}_s$ for every s) in order to avoid singularities. In Fourier transform this gives

$$|\tilde{\mathcal{V}}^{\mathrm{ext}}(\boldsymbol{k})\rangle = \frac{q^{\mathrm{ext}}}{v\epsilon_0}\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{r}_0}|\tilde{C}^0(\boldsymbol{k})\rangle, \tag{3.13}$$

where

$$C_s^0(\boldsymbol{r}) = \frac{v}{4\pi|\boldsymbol{r} + \boldsymbol{\xi}_s - \boldsymbol{\xi}_0|}. \tag{3.14}$$

4. The spectral properties of the evolution operator

As seen from eqs. (3.6) the matrix $\tilde{A}$, which governs the evolution, contains a hopping part $\tilde{\Gamma}$ and a coulombian interaction part $\tilde{X}$.

Because of the connectivity structure of our model, $\tilde{\Gamma}$ has the block diagonal form

$$\tilde{\Gamma} = \begin{pmatrix} \tilde{\Gamma}_\mathrm{f} & 0 \\ 0 & \tilde{\Gamma}_\mathrm{b} \end{pmatrix}, \tag{4.1}$$

$\tilde{\Gamma}_\mathrm{f}$ being the matrix $\tilde{\Gamma}$ of ref. I, where its properties are discussed at length, we give here only a brief sketch.

It is both reasonable and sufficient for our purpose to assume that $W_{ss'}(\boldsymbol{r})$ decays at least exponentially with $|\boldsymbol{r}|$ at infinity. This ensures the indefinite differentiability of $\tilde{\Gamma}_\mathrm{f}(\boldsymbol{k})$ as well as the analyticity of $\tilde{\Gamma}_\mathrm{f}(\epsilon\boldsymbol{k})$ in ϵ around $\epsilon = 0$. The fact that $\tilde{\Gamma}_\mathrm{f}p$ is a hermitian matrix (see eqs. (3.3)) shows that $\tilde{\Gamma}_\mathrm{f}$ is diagonalizable with real eigenvalues $\gamma_\alpha(\boldsymbol{k})$, $\alpha = 1, 2, \ldots, S$ its eigenvectors $|u_\alpha(\boldsymbol{k})\rangle$ $\alpha = 1, 2, \ldots, S$ being mutually orthogonal and normalized in the scalar product defined by p^{-1} in $\mathcal{S}_\mathrm{f}$. In other words the adjoint eigenvalue problem has the same eigenvalues and the eigenvectors are

$$|\bar{u}_\alpha(\boldsymbol{k})\rangle = p^{-1}|u_\alpha(\boldsymbol{k})\rangle. \tag{4.2}$$

The eigenvalues and eigenvectors have the differentiability and analyticity properties of $\tilde{\Gamma}_f(\mathbf{k})$.

The positivity of $\tilde{\Gamma}_f(\boldsymbol{k})$ is proved using its quadratic form in the scalar product defined by p^{-1}. Indeed, for any non-zero $|x\rangle \in \mathcal{S}_f$ with $|y\rangle = p^{-1}|x\rangle$ one has, using eqs. (3.3),

$$\langle x, p^{-1}\tilde{\Gamma}_f(\boldsymbol{k})x\rangle = \langle y, \tilde{\Gamma}_f(\boldsymbol{k})py\rangle = \frac{1}{2\sum_{s=1}^{S} f_s(1-f_s)} \sum_{\boldsymbol{r}} \sum_{s,s'=1}^{S} \mathcal{W}_{ss'}(\boldsymbol{r})|y_s - y_{s'}\,\mathrm{e}^{i\boldsymbol{k}\boldsymbol{r}}|^2 \geqslant 0. \tag{4.3}$$

We have equality in (4.3) for $\boldsymbol{k} = 0$ and $y_s = y_{s'}$ for any $s,s' \leqslant S$ and connectivity arguments show that this is the only case. Thus the only vanishing eigenvalue of $\tilde{\Gamma}_f(\boldsymbol{k})$ (labeled by $\alpha = 1$) occurs at $\boldsymbol{k} = 0$, is non degenerate and its eigenvector is

$$|u_1(0)\rangle = |p\rangle; \quad |\bar{u}_1(0)\rangle = p^{-1}|p\rangle = |1_f\rangle. \tag{4.4}$$

Moreover, by the reality of $\Gamma(\boldsymbol{r})$, $\gamma_1(\boldsymbol{k})$ has a quadratic behaviour around $\boldsymbol{k} = 0$

$$\gamma_1(\boldsymbol{k})_{k\to 0} \sim \boldsymbol{k}\mathcal{D}\boldsymbol{k} \tag{4.5}$$

and $\mathcal{D}$ can be shown to be a strictly positive definite tensor (I).

On the other hand, since

$$\Gamma_b(\boldsymbol{r}) = \Gamma_b\delta_{\boldsymbol{r},0} \tag{4.6a}$$

one gets

$$\tilde{\Gamma}_b(\boldsymbol{k}) = \Gamma_b \tag{4.6b}$$

and therefore γ_α and $|u_\alpha\rangle$ are independent of $\boldsymbol{k}$ for $\alpha = S+1, S+2, \ldots, S+\Sigma$. All the eigenvalues are strictly positive except

$$\gamma_{S+1} = 0; \quad |u_{S+1}\rangle = |q\rangle; \quad |\bar{u}_{S+1}\rangle = q^{-1}|q\rangle = |1_b\rangle. \tag{4.7}$$

It is clear that the spectrum of $\tilde{\Gamma}(\boldsymbol{k})$ is a superposition of the "bands" of $\tilde{\Gamma}_f(\boldsymbol{k})$ and those, perfectly flat, of $\tilde{\Gamma}_b = \Gamma_b$. All the branches are strictly positive except one which is identically zero and another which touches the origin at $\boldsymbol{k} = 0$ only.

We turn now to the interaction matrix $\tilde{X}$. Up to a similarity transformation, defined by $\nu^{1/2}$, it is hermitian

$$\nu^{-1/2}\tilde{X}(\boldsymbol{k})\nu^{1/2} = 1 + \nu^{1/2}\tilde{C}(\boldsymbol{k})\nu^{1/2}, \tag{4.8}$$

so that its spectrum is real. Let us see under what conditions it is positive. The Coulomb matrix is not positive definite, but is bounded from below. A lower bound is given by a well known result due to Onsager[5])

$$C \geqslant -\frac{v}{2\pi d}, \tag{4.9}$$

where d is the smallest distance in the lattice.

If L and l exceed a certain critical value $l_0 > 0$ the matrix ν becomes sufficiently small and the positivity of the unit operator in eq. (4.8) prevails. Using the bound (4.9) and $p, q \leqslant 1$ we get

$$l_0 \leqslant \sqrt{\frac{v}{2\pi d}}. \tag{4.10}$$

In its turn, the positivity of $\tilde{X}(\boldsymbol{k})$ ensures the positivity of the spectrum for the evolution operator $\tilde{A}(\boldsymbol{k})$, as will be shown below. This is essential for the convergence to equilibrium to take place for any solution eq. (3.12). Since the stability of the equilibrium is a characteristic feature of the master equation, which is the starting point[6]) in deriving the hopping rate equations (3.2), it is clear that, in the range of the parameters where negative eigenvalues occur, the instability is an artifact of the approximations used during the derivation. An example given in II shows the selfconsistent approximation to be responsible for the departure from the equilibrium in systems with too small distances between the sites. Therefore only the cases when $L, l > l_0$ and consequently

$$\nu^{-1/2}\tilde{X}(\boldsymbol{k})\nu^{1/2} > 0, \quad \text{for any } \boldsymbol{k} \in \mathrm{BZ} \tag{4.11}$$

will be dealt with.

We are now in the position to discuss the properties of the operator $\tilde{A}$. We begin by proving the following statement: $\tilde{A}(\boldsymbol{k})$ is diagonalizable, with real, positive eigenvalues, bounded as functions of $\boldsymbol{k}$ throughout the BZ. To this end let us consider the matrix

$$\begin{aligned} \mathscr{A}(\boldsymbol{k}) &= \nu^{-1/2}\tilde{\Gamma}(\boldsymbol{k})^{1/2}\tilde{X}(\boldsymbol{k})\tilde{\Gamma}(\boldsymbol{k})^{1/2}\nu^{1/2} \\ &= [\nu^{-1/2}\tilde{\Gamma}^{1/2}\nu^{1/2}][\nu^{-1/2}\tilde{X}\nu^{1/2}][\nu^{-1/2}\tilde{\Gamma}^{1/2}\nu^{1/2}]. \end{aligned} \tag{4.12}$$

Because $\tilde{\Gamma}(\boldsymbol{k})\nu$ is hermitian, so is $\nu^{-1/2}\tilde{\Gamma}\nu^{1/2}$, which makes the hermiticity and positivity of $\mathscr{A}(\boldsymbol{k})$ obvious. Zero belongs to the spectrum, and is nondegenerate for $\boldsymbol{k} \neq 0$ with the eigenvector $\nu^{-1/2}|q\rangle$. Moreover, since

$$\tilde{\Gamma}^{1/2}_{(\boldsymbol{k})}\nu^{1/2}\mathscr{A}(\boldsymbol{k}) = \tilde{A}(\boldsymbol{k})\tilde{\Gamma}(\boldsymbol{k})^{1/2}\nu^{1/2} \tag{4.13}$$

the following connection between the eigenvalue problem for $\mathscr{A}$ and $\tilde{A}$ holds:

if

$$\mathscr{A}(\boldsymbol{k})|x_\alpha\rangle = a_\alpha|x_\alpha\rangle$$

and

$$\tilde{\Gamma}(\boldsymbol{k})^{1/2}\nu^{1/2}|x_\alpha\rangle = |v_\alpha\rangle \neq 0 \tag{4.14}$$

then

$$\tilde{A}(\boldsymbol{k})|v_\alpha\rangle = a_\alpha|v_\alpha\rangle.$$

On the other hand, $\tilde{\Gamma}(\boldsymbol{k})^{1/2}\nu^{1/2}|x_\alpha\rangle = 0$ is equivalent to $a_\alpha = 0$. This way we proved that $\mathscr{A}(\boldsymbol{k})$ and $\tilde{A}(\boldsymbol{k})$ share all the non-vanishing eigenvalues, with the eigenvectors related by eq. (4.14). But zero belongs to the spectrum of $\tilde{A}(\boldsymbol{k})$ too, with the eigenvector

$$|v_{S+1}(\boldsymbol{k})\rangle = \tilde{X}(\boldsymbol{k})^{-1}|u_{S+1}\rangle, \tag{4.15}$$

so that $\mathscr{A}(\boldsymbol{k})$ and $\tilde{A}(\boldsymbol{k})$ have the same spectrum.

Even though $\tilde{A}(\boldsymbol{k})$ is singular in $\boldsymbol{k} = 0$, it is easy to check that $\mathscr{A}(\boldsymbol{k})$ is not, wherefrom the boundedness of the spectrum follows.

5. The scaling limit

This section is devoted to the scaling properties of $\tilde{A}(\boldsymbol{k})$ and of the solution $|\tilde{\eta}(\boldsymbol{k}, t)\rangle$. $\tilde{A}(\boldsymbol{k})$ depends on the scaling parameter λ through $\boldsymbol{k}$ and l, and the solution through $\boldsymbol{k}$, l, t and $\boldsymbol{x}_0$. We shall use sometimes the subscript λ to indicate the scaling of these arguments.

The behaviour of the spectrum and eigenprojectors of $\tilde{A}_\lambda(\boldsymbol{k}, l) = \tilde{A}(\boldsymbol{k}/\lambda, \lambda l)$ for $\lambda \to \infty$ is an analytic perturbation problem^{7}) in $1/\lambda$ as expansion parameter. The results we are going to prove are summarized below.

Using the notations

$$\tilde{X}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right)^{-1} = Y\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right) = Y_0 + \frac{1}{\lambda}Y_1 + \frac{1}{\lambda^2}Y_2 + \cdots, \tag{5.1}$$

$$|1\rangle = |1_f\rangle + |1_b\rangle, \tag{5.2}$$

and $|f(\boldsymbol{\xi})\rangle$ being the vector with components $f(\boldsymbol{\xi}_s)$, $s = 1, 2, \ldots, S + \Sigma$ for any function f, we have

(i) $\lim_{\lambda\to\infty} a_\alpha(\boldsymbol{k}/\lambda, \lambda l)$ exists and is strictly positive for all α except two

$$a_{S+1}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right) = 0, \tag{5.3}$$

$$\lim_{\lambda\to\infty} \lambda^2 a_1\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right) = \boldsymbol{k}\mathscr{D}\boldsymbol{k}\left(1 + \frac{\epsilon_0}{l^2 \boldsymbol{k}\epsilon\boldsymbol{k}}\right), \tag{5.4}$$

where the tensor ϵ is defined by

$$\frac{\boldsymbol{k}(\epsilon - \epsilon_0)\boldsymbol{k}}{\epsilon_0} = \frac{1}{L^2}\langle \boldsymbol{k}\boldsymbol{\xi}, Y_0 q \boldsymbol{k}\boldsymbol{\xi}\rangle \geqslant 0 \tag{5.5}$$

and will be later identified as the dielectric tensor (hence the notation)

(ii) the eigenprojectors P_1, P_{S+1} corresponding to the vanishing eigenvalues have poles in $1/\lambda = 0$, but

$$\lim_{\lambda\to\infty} \langle \mathrm{e}^{-i(\boldsymbol{k}\boldsymbol{\xi}/\lambda)} | P_{S+1}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right) = \frac{k^2 l^2}{1 + l^2(\boldsymbol{k}\epsilon\boldsymbol{k}/\epsilon_0)} \langle 1_b|, \tag{5.6}$$

$$\lim_{\lambda\to\infty} \langle \mathrm{e}^{-i(\boldsymbol{k}\boldsymbol{\xi}/\lambda)} | P_1\left(\frac{\boldsymbol{k}}{\lambda}, \lambda l\right) = \frac{k^2 \epsilon_0}{\boldsymbol{k}\epsilon\boldsymbol{k}}\left[\langle 1_f| + \frac{1}{1 + l^2(\boldsymbol{k}\epsilon\boldsymbol{k}/\epsilon_0)}\langle 1_b|\right]; \tag{5.7}$$

(iii) defining the microscopical internal charge by

$$\rho_{\text{int}}^{\text{micro}}(\boldsymbol{x}, t) = e\sum_{\boldsymbol{r}} \sum_{s=1}^{S+\Sigma} \eta_s(\boldsymbol{r}, t)\delta(\boldsymbol{x} - \boldsymbol{r} - \boldsymbol{\xi}_s), \tag{5.8a}$$

i.e.

$$\hat{\rho}_{\text{int}}^{\text{micro}}(\boldsymbol{k}, t) = e\langle \mathrm{e}^{-i\boldsymbol{k}\boldsymbol{\xi}}, \tilde{\eta}(\boldsymbol{k}, t)\rangle, \tag{5.8b}$$

and the total microscopical charge

$$\hat{\rho}_{\text{tot}}^{\text{micro}}(\boldsymbol{k}, t) = \hat{\rho}_{\text{int}}^{\text{micro}}(\boldsymbol{k}, t) + q^{\text{ext}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{x}_0}, \tag{5.9}$$

the following scaling limit holds:

$$\lim_{\lambda\to\infty} \hat{\rho}_{\text{tot}}^{\text{micro}}\left(\frac{\boldsymbol{k}}{\lambda}, \lambda^2 t; \lambda l, \lambda \boldsymbol{x}_0\right) = \text{r.h.s. of eq. (2.5)}, \tag{5.10}$$

in which the dielectric constant tensor is given by eq. (5.5), and

$$\begin{gathered} D = \mathscr{D}, \\ \hat{\rho}(\boldsymbol{k}, 0) = e\langle 1_f, \tilde{\eta}(0, 0)\rangle = \sum_{\boldsymbol{r}} \sum_{s=1}^{S} \eta_s(\boldsymbol{r}, 0), \\ \hat{\rho}_{\text{ext}}(\boldsymbol{k}) = e\langle 1_b, \tilde{\eta}(0, 0)\rangle + q^{\text{ext}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{x}_0} = e\sum_{\boldsymbol{r}} \sum_{s=S+1}^{S+\Sigma} \eta_s(\boldsymbol{r}, 0) + q^{\text{ext}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\boldsymbol{x}_0}. \end{gathered} \tag{5.11}$$

The last relation shows that if the "islands" are not electrically neutral their total charge eventually plays the role of an external fixed charge, located at the origin.

The proof makes use again of the fact that $\tilde{A}_\lambda(\boldsymbol{k}, l)$ is related by eqs. (4.13) and (4.14) to the selfadjoint operator $\mathcal{A}_\lambda(\boldsymbol{k}, l)$ which is analytic around $1/\lambda = 0$. This is easily seen, using the fact that $\tilde{\Gamma}(\boldsymbol{k}/\lambda)^{1/2}$ is analytic (see II). This already proves$^{7)}$ that the eigenvalues are analytic too. The relationship between the eigenprojectors of $\mathcal{A}_\lambda$ which are analytic and those of $\tilde{A}_\lambda$ is more complicated, and the analyticity is in general lost. The eigenprojectors of $\tilde{A}_\lambda$ may have poles in $1/\lambda = 0$, but it is important that $1/\lambda = 0$ is not a branching point for them because it is not for the eigenvalues$^{7)}$.

The existence and the positivity of the limit for the eigenvalues is now obvious, but we are interested in the detailed behaviour of the eigenvalues which vanish in the scaling limit and of their eigenprojectors. To this end we found convenient to transform the eigenvalue problem

$$\tilde{A}_\lambda|v_\lambda\rangle = a_\lambda|v_\lambda\rangle \tag{5.12}$$

into the eigenvalue problem in the generalized form$^{7)}$

$$\tilde{\Gamma}_\lambda|x_\lambda\rangle = a_\lambda Y_\lambda|x_\lambda\rangle; \quad |v_\lambda\rangle = Y_\lambda|x_\lambda\rangle. \tag{5.13}$$

They are equivalent, but the latter has the advantage that in perturbing the vanishing eigenvalue, the unperturbed problem contains only $\tilde{\Gamma}(0)$.

Since

$$\tilde{\Gamma}^+ = \nu^{-1}\tilde{\Gamma}\nu; \quad Y^+ = \nu^{-1}Y\nu, \tag{5.14}$$

the solution of eq. (5.13) answers the adjoint eigenvalue problem too and gives

$$|\bar{v}_\lambda\rangle = \nu_\lambda^{-1}|x_\lambda\rangle, \tag{5.15}$$

so that the eigenprojector of $\bar{A}_\lambda$ is

$$P_\lambda = \frac{Y_\lambda|x_\lambda\rangle\langle x_\lambda|\nu_\lambda^{-1}}{\langle x_\lambda, \nu_\lambda^{-1} Y_\lambda x_\lambda\rangle}. \tag{5.16}$$

The order by order analysis of eq. (5.13) is a straightforward matter and we shall skip the details. We merely point out the important ingredients. Perhaps the most important is the fact that the singular part of the Coulomb matrix is of one-dimensional range

$$\tilde{X}(\boldsymbol{k}) = 1 + \frac{1}{\boldsymbol{k}^2\Lambda^2}Q(\boldsymbol{k}, l) + \nu\tilde{R}(\boldsymbol{k}), \tag{5.17a}$$

$$Q(\boldsymbol{k}, l) = \Lambda^2\nu|\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\xi}\rangle\langle\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\xi}| = Q^2(\boldsymbol{k}, l) \tag{5.17b}$$

and for $\boldsymbol{k} = 0$, Q projects on the hull subspace of $\tilde{\Gamma}$. It is useful to write

$Y(\mathbf{k}, l)$ in the form

$$\begin{aligned} Y(\mathbf{k}, l) &= \left[1 + \left(1 - \frac{1}{1 + k^2 \Lambda^2} \mathrm{Q}\right) \nu \bar{R}\right]^{-1} \left(1 - \frac{1}{1 + k^2 \Lambda^2} \mathrm{Q}\right) \\ &= B^{-1} \left(1 - \frac{1}{1 + k^2 \Lambda^2} \mathrm{Q}\right) \end{aligned} \tag{5.18}$$

on which it is easy to check that

$$\begin{aligned} &\langle 1_{\mathrm{f}} | Y_0 = \langle 1_{\mathrm{f}} |; \quad \langle 1_{\mathrm{b}} | Y_0 = -\langle 1_{\mathrm{f}} |; \quad Y_0 | q \rangle = 0, \\ &\langle 1_{\mathrm{f}}, Y_1 q \rangle = \langle 1_{\mathrm{b}}, Y_1 q \rangle = 0, \\ &\langle 1_{\mathrm{f}}, Y_2 q \rangle = -\frac{L^2}{l^2}, \\ &\langle 1, Y_2 q \rangle = k^2 L^2 + \langle \mathbf{k} \boldsymbol{\xi}, Y_0 q \mathbf{k} \boldsymbol{\xi} \rangle \geqslant k^2 L^2. \end{aligned} \tag{5.19}$$

The last inequality stems from the positivity of $Y\nu$ whose zeroth order in $1/\lambda$ is $L^{-2} Y_0 q$. It is also important to observe that in $\mathscr{S}_{\mathrm{b}}$ only the zeroth order of $\bar{\Gamma}_\lambda$ appears.

The perturbation of the doubly degenerate eigenvalue $\gamma_1(0) = \gamma_{S+1}(0) = 0$ of $\bar{\Gamma}(0)$ gives rise to

$$a_{S+1} \equiv 0; \quad |x_{S+1}\rangle \equiv |q\rangle \quad \text{and} \quad a_1\left(\frac{\mathbf{k}}{\lambda}, \lambda l\right) = \frac{1}{\lambda^2} \mathbf{k} \mathscr{D} \mathbf{k} \frac{\langle 1_{\mathrm{b}}, Y_2 q \rangle}{\langle 1, Y_2 q \rangle} + \cdots, \tag{5.20}$$

$$\left| x_1\left(\frac{\mathbf{k}}{\lambda}, \lambda l\right) \right\rangle = |q\rangle + \frac{1}{\lambda^2} |p\rangle \langle 1_{\mathrm{b}}, Y_2 q \rangle,$$

wherefrom (i) and (ii) follow. In order to derive (iii) we use the spectral decomposition

$$\mathrm{e}^{-\bar{A}_\lambda \lambda^2 t} = \sum_{\alpha=1}^{S+\Sigma} \mathrm{e}^{-a_\alpha(\mathbf{k}/\lambda,\, \lambda l) \lambda^2 t} \mathrm{P}_\alpha\left(\frac{\mathbf{k}}{\lambda}, \lambda l\right). \tag{5.21}$$

It is obvious that the terms corresponding to the nonvanishing eigenvalues ($\alpha \neq$, 1, $S + 1$) decay exponentially when $\lambda \to \infty$. The terms we are left with were described in (i) and (ii). We use also

$$|\bar{C}^0(\mathbf{k})\rangle = \frac{1}{k^2} |\mathrm{e}^{-\mathrm{i}\mathbf{k}\boldsymbol{\xi}}\rangle \mathrm{e}^{\mathrm{i}\mathbf{k}\boldsymbol{\xi}_0} + |\bar{R}^0(\mathbf{k})\rangle, \tag{5.22}$$

which is the analogue of eq. (3.8) and

$$\lim_{\lambda \to \infty} Y\nu |\bar{C}^0(\mathbf{k})\rangle \mathrm{e}^{\mathrm{i}\mathbf{k}\mathbf{r}_0} = \left[B_0^{-1} |q\rangle + \frac{1}{L^2} Y_0 q |\bar{R}^0(0)\rangle\right] \mathrm{e}^{\mathrm{i}\mathbf{k}\mathbf{x}_0}. \tag{5.23}$$

6. Discussion of the results

The problem of the macroscopic behavior of our system is answered by eqs. (5.10) and (5.11) which form the central result of the paper. The asymptotic domain in which the microscopic evolution approaches the macroscopic fundamental solution is suggested by the scale invariance of the latter.

A slight generalization of the procedure should be used in order to produce non-fundamental solutions. Since the scale invariance eq. (2.8) does not hold in this form for the general solution, one recovers it only by scaling all the relevant geometric parameters of the initial and external charge densities. This prevents them from shrinking to point-like distributions in the scaling limit, exactly as we had to scale $\boldsymbol{x}_0$ in order to keep the initial and external conditions macroscopically far apart. The phenomenon is illustrated by the fact that no matter what (summable) initial conditions we considered, it got delta-form ($\boldsymbol{k}$-independent Fourier components) in the end, as seen in eqs. (5.11).

It is interesting to point out that the scaling procedure we employed is entirely equivalent to keeping $\boldsymbol{x}$, t, l, $\boldsymbol{x}_0$ fixed and scaling instead the lattice constant as $1/\lambda$, the transition rates as λ^2, and the charge of the bound electrons as λ, while keeping the average density of particles constant. (The scaling of the bound charge alongside with the diminishing of the space between the sites is the natural way to produce a distribution of point dipoles.) In more physical terms this means that the macroscopical behavior is observed for distances which are much larger than the lattice constant and times much larger than the inverse of some typical value of the transition rates.

The model is reasonable only for l and L greater than a critical value l_0. Otherwise the equilibrium solution becomes unstable, which is both unphysical and in contradiction with the starting point of the model—the master equation. As far as l is concerned this is not so restrictive since the macroscopical behaviour we have in mind is typical for semiconductors, where l has macroscopic values.

Another feature of the model is the fact that the scaling limit and the $t \to 0$ limit do not commute

$$\lim_{\lambda\to\infty}\lim_{t\to 0}\hat{\rho}_{\text{tot}}^{\text{micro}} = e(1, \tilde{\eta}(0,0)) + q^{\text{ext}}\, e^{i\boldsymbol{k}\boldsymbol{x}_0} = \hat{\rho}(\boldsymbol{k},0) + \hat{\rho}_{\text{ext}}(\boldsymbol{k}), \tag{6.1}$$

$$\lim_{t\to 0}\lim_{\lambda\to\infty}\hat{\rho}_{\text{tot}}^{\text{micro}} = \frac{k^2\epsilon_0}{\boldsymbol{k}\epsilon\boldsymbol{k}}[\hat{\rho}(\boldsymbol{k},0) + \hat{\rho}_{\text{ext}}(\boldsymbol{k})], \tag{6.2}$$

the difference being exactly the polarization charge. At a microscopic time scale the polarization sets in with a certain delay, but the characteristic time is too short to be observed macroscopically. Eq. (6.2) shows that indeed, the macroscopic polarization charge follows instantaneously the free and external charge.

Even though the free and bound electrons move in a strongly interdependent way, the phenomenological constants governing the macroscopic evolution are the same as for uncoupled systems. Indeed, the diffusion constant (and hence the conductivity) is expressed in terms of the free electrons parameters only, as seen from eqs. (4.5) and (5.11). In its turn, the expression (5.5) of the dielectric tensor is dependent only on quantities pertaining to the bound electrons, because

$$Y_0 q = \begin{pmatrix} 0 & 0 \\ 0 & [1+(1/L^2)(1-|q)\langle 1_\mathrm{b}|)q\bar{R}_{\mathrm{bb}}(0)]^{-1}(1-|q)\langle 1_\mathrm{b}|)q \end{pmatrix}, \tag{6.3}$$

where $\bar{R}_{\mathrm{bb}}$ is the diagonal block of $\tilde{R}$ corresponding to $\mathscr{S}_\mathrm{b}$, and the inverse in eq. (6.3) is taken in $\mathscr{S}_\mathrm{b}$.

This is why, in the subsequent discussion of the dielectric constant formula, we shall omit the free electron system altogether, without loss of generality.

On the one hand, we are interested in the relationship with the well-known Clausius–Mossotti formula

$$\frac{\epsilon-\epsilon_0}{\epsilon_0} = \frac{\kappa}{1-\frac{1}{3}\kappa} \tag{6.4}$$

which links the dielectric constant to the polarizability $\epsilon_0\kappa$ for a system of point dipoles, in a lattice with cubic symmetry. The point dipoles may be obtained in our model by taking the limit $\xi_s \to 0$ and at the same time increasing the elementary charge ($e \to \infty$ or, equivalently, $L \to 0$) in the same proportion, in order to prevent a vanishing results. More precisely, we have to perform the limit $\xi_s \to 0$; $L \to 0$; $\xi_s/L = \text{const.}$ in eq. (5.5). The delicate point here is the fact that for small ξ_s's the Coulomb matrix is singular. For cubic lattices we have (using the explicit expression of $\tilde{R}(\boldsymbol{k})$ given in II)

$$\hat{R}_{ss'}(0) = \frac{v}{4\pi}\frac{1-\delta_{ss'}}{|\boldsymbol{\xi}_s-\boldsymbol{\xi}_{s'}|} + C_0 + \frac{1}{6}(\boldsymbol{\xi}_s-\boldsymbol{\xi}_{s'})^2 + \cdots. \tag{6.5}$$

It is clear that a more reasonable treatment of the Coulomb interaction between the sites of a given cell is necessary if the sites come too close to one another. Nevertheless it can be proved that no matter what "softened" interaction replaces the singular term in (6.5), the dipolar limit of ϵ satisfies eq. (6.4) with κ given by the polarizability of the isolated unit cell

$$\boldsymbol{p} = \frac{1}{v}\sum_s e\boldsymbol{\xi}_s\eta_s = \epsilon_0\kappa\boldsymbol{E}^{\mathrm{ext}}. \tag{6.6}$$

Of course, the dipolar response to the external field is computed using the same intracellular interaction.

On the other hand, it is instructive to discuss Kubo's linear response theory on this model, where an exact solution is available. The linear response of the charge with respect to the field, regardless of the source of the latter, is given by eq. (3.2a). In the spirit of the standard dielectric response theory one expects to find here all the relevant information.

Let us consider the equilibrium value of the charge density in the presence of the field:

$$\begin{aligned}\hat{\rho}_{\text{int}}^{\text{micro}}(\boldsymbol{k}) = e\langle \mathrm{e}^{-i\boldsymbol{k}\xi}, \tilde{\eta}(\boldsymbol{k},\infty)\rangle = -\frac{\epsilon_0 v}{L^2}\langle \mathrm{e}^{-i\boldsymbol{k}\xi}, (1-|q\rangle\langle 1|)q\tilde{\mathcal{V}}(\boldsymbol{k},\infty)\rangle \\ = \langle \tilde{\chi}(\boldsymbol{k}), \tilde{\mathcal{V}}(\boldsymbol{k},\infty)\rangle. \qquad (6.7)\end{aligned}$$

The notation emphasizes the fact that the equilibrium values correspond to taking $t \to \infty$ in the solutions $|\tilde{\eta}(\boldsymbol{k},t)\rangle$, $|\tilde{\mathcal{V}}(\boldsymbol{k},t)\rangle$.

The usual approach is to obtain the permitivity by comparing eq. (6.7) with the macroscopical relation

$$\hat{\rho}_{\text{int}}(\boldsymbol{k}) = -\boldsymbol{k}(\epsilon-\epsilon_0)\boldsymbol{k}\hat{V}(\boldsymbol{k}), \qquad (6.8)$$

using only the information contained in $\tilde{\chi}_s(\boldsymbol{k})$. The procedure involves some space averaging on $\chi_s(\boldsymbol{r})$ (see for instance II). However, it is obvious that the Coulomb interaction which appears in the expression of the dielectric constant eqs. (5.5) and (6.3) is absent in $\tilde{\chi}_s(\boldsymbol{k})$ (and therefore one may recover at most the polarizability of the isolated cell without Coulomb interactions).

Our result may be recovered from eq. (6.7) by taking its scaling limit, which implies the knowledge of the actual dependence of $|\tilde{\eta}\rangle$ and $|\mathcal{V}\rangle$ on the scaling parameter.

This information is given only by the complete solution of eqs. (3.2a) and (3.2b). The convergence of the left-hand side of eq. (6.7) to the macroscopical charge density being already established, we concentrate upon the potential. Combining the eqs. (3.2b), (3.8), (3.13) and (5.22) one finds the following scaling behaviour:

$$\begin{aligned}v|\tilde{\mathcal{V}}_\lambda(\boldsymbol{k},t)\rangle = \lambda^2|1\rangle\hat{V}(\boldsymbol{k},t) + \lambda\Big[|-i\boldsymbol{k}\boldsymbol{\xi}\rangle\hat{V}(\boldsymbol{k},t) + \frac{e}{\epsilon_0}\bar{R}(0)|\tilde{\eta}_\lambda(\boldsymbol{k},t)\rangle_{-1} + \frac{1}{\epsilon_0 k^2}|1\rangle \\ \times(\langle \mathrm{e}^{-i(\boldsymbol{k}\xi/\lambda)}, \tilde{\eta}_\lambda(\boldsymbol{k},t)\rangle)_1\Big] + \cdots. \qquad (6.9)\end{aligned}$$

Here $\hat{V}(\boldsymbol{k},t)$ is the macroscopic potential eq. (2.3) and the subindices of the λ-dependent quantities show the order of the $1/\lambda$ expansion. For instance

$$|\tilde{\eta}_\lambda(\boldsymbol{k},t)\rangle_{-1} = -\frac{Y_0 q}{L^2}|-i\boldsymbol{k}\boldsymbol{\xi}\rangle\epsilon_0\hat{V}(\boldsymbol{k},t) \qquad (6.10)$$

is the singular part of $|\tilde{\eta}_\lambda(\boldsymbol{k}, t)\rangle$. Introducing eq. (6.9) in (6.7) one observes the rather surprising fact that the dominant term, which is used in the identification of the macroscopic potential (see also II)

$$\lim_{\lambda\to\infty}\frac{1}{\lambda^2}v|\tilde{\mathcal{V}}_\lambda(\boldsymbol{k}, t)\rangle = |1\rangle\hat{V}(\boldsymbol{k}, t) \tag{6.11}$$

is cancelled out by the operator $(1-|q\rangle\langle q|)q$, because it is proportional to the vector $|1\rangle$. Fortunately the next term also contains $\hat{V}(\boldsymbol{k}, t)$ alongside with $\tilde{R}(0)$ and $\boldsymbol{\xi}$ dependent quantities and leads to the desired result.

The whole discussion remains unchanged if we start from the dynamical response function and take the zero frequency limit in order to obtain the static dielectric constant.

Acknowledgment

The authors are gratefully acknowledging illuminating discussions with G.A. Mezincescu.

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PHYSICAL REVIEW B VOLUME 29, NUMBER 2 15 JANUARY 1984

Clausius-Mosotti limit of the quantum theory of the electronic dielectric constant

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(Received 4 February 1983; revised manuscript received 25 May 1983)

It is shown that an unambiguous discussion of the limit of independent atoms in the theory of the dielectric constant has to be formulated in terms of asymptotic series in the ratio of the Bohr radius to the lattice spacing. In this sense, a derivation of the classical Clausius-Mosotti (or Lorentz-Lorenz) formula is given, starting from the quantum-mechanical theory including "local-field" corrections, at the self-consistent Hartree-Fock level. Our approach clarifies the origin of the many diverging results on the subject and eliminates most of the unnecessary approximations and/or assumptions.

I. INTRODUCTION

The classical Clausius-Mosotti (or Lorentz-Lorenz, for nonzero frequency) relation for cubic crystals (and gases)

$$\frac{\epsilon}{\epsilon_0}-1=\frac{\kappa^{\mathrm{at}}}{1-\frac{1}{3}\kappa^{\mathrm{at}}} \tag{1.1}$$

between the dielectric constant ϵ and the atomic polarizability κ^{at} (ϵ_0 is the permittivity of the vacuum) was derived before the advent of quantum mechanics, from the model of independent neutral atoms assimilated to point dipoles whose magnitude is proportional to the local electric field. This field is given by the sum of the external field and the field of all the other atomic dipoles. In the textbook derivations the latter is evaluated in a hybrid manner, i.e., it is done microscopically within a finite sphere while the rest of the system is treated as a continuum. More recent treatments[1,2] perform the discrete summation over the whole lattice of atoms and compute the macroscopic field by taking the long-wavelength limit. These derivations contain again the essential ingredient of uncorrelated atoms, i.e., there is no overlap between the wave functions of electrons on different atoms.

On intuitive grounds one expects that as the ratio r_0/a of Bohr radius to the lattice spacing tends to zero, one should approach the limit of uncorrelated atoms. On the other hand, it was known for a long time that the well-known quantum-mechanical linear-response Kubo formula, which for crystals in the one-electron frame is equivalent to that derived in Refs. 3 and 4, leaves no hopes of recovering (1.1) for very large lattice spacing. Only after the refining of the quantum theory of the dielectric constant of crystals, due to Adler[5] and Wiser[6] the possibility of a deeper understanding of this problem evolved. They remarked that since the translational invariance is only discrete (and this is precisely the way the atomic structure enter the theory), specific "local-field" corrections appear.

Nevertheless, until now there has been a wide controversy in the literature with arguments both for and against the Clausius-Mosotti (Lorentz-Lorenz) limit. Some of these papers[7,8] use unnecessarily restrictive assumptions and approximations, being at the same time mathematically nonconvincing. Other works,[9–12] under similar conditions obtain alternative results disagreeing with (1.1). A typical source of misunderstanding was the ambiguous definition of the atomic polarizability, as it was pointed out recently in Ref. 13. Generally speaking, most of the discussion was within the frame of self-consistent theories with respect to Coulomb interactions, however, it was felt that self-interaction is dangerous and therefore exchange and perhaps correlation effects should be essential. Another ingredient which is often used to simplify the calculations is some variant of a factorization assumption. Whereas there is a general agreement as to the necessity of taking the atoms sufficiently far apart, this idea is implemented in a mathematically imprecise manner, differing from author to author.

In this paper we will show that the Clausius-Mosotti problem must be formulated as an asymptotic series problem in the parameter r_0/a. This is the only mathematically reasonable formulation. It means that one should neglect only terms that vanish faster than any power. We restrict the discussion to the self-consistent potential or Hartree-Fock approximations and will prove that, in the above-mentioned sense, the Clausius-Mosotti formula is recovered in both cases. The only ingredients are the inclusion of local-field effects in the manner of Adler-Wiser[5,6] and certain assumptions about the asymptotic behavior (in r_0/a) of the wave functions and spectrum. These plausible assumptions coincide with the commonly used ones.

In Sec. II the quantum theory of the macroscopic dielectric constant is described together with the basic approximation schemes. The quantum theory of the atomic polarizability is outlined in Sec. III in a way that facilitates our purposes. Our basic statement and its proof on the asymptotic validity of the Clausius-Mosotti relation, are contained in Sec. IV. The last section is devoted to the discussion of the results.

II. QUANTUM THEORY OF THE MACROSCOPIC DIELECTRIC CONSTANT

With the use of linear-response theory with respect to an external potential V^{ext} of frequency ω, in the quantum-mechanical problem of the motion of electrons in a periodic potential at $T=0$ K one may define the dielectric matrix $[\hat\epsilon(\vec k,\omega)]_{\vec K\vec K'}$ [$\vec k$ belongs to the first Brillouin zone (BZ), while $\vec K$ and $\vec K'$ run through the reciprocal lattice] as relating the Fourier transforms of the total potential (created by the external and internal nonequilibrium charges) to the external one

$$\sum_{\vec K'}[\hat\epsilon(\vec k,\omega)]_{\vec K\vec K'}V^{\text{tot}}(\vec k+\vec K',\omega)=\epsilon_0 V^{\text{ext}}(\vec k+\vec K,\omega)\ . \tag{2.1}$$

In terms of the perturbation theory diagrams (see, for example, Ref. 14) with respect to the electron-electron Coulomb interaction it can be shown that $[\hat\epsilon(\vec k,\omega)]_{\vec K\vec K'}$ is related to the so-called polarization part Π:

$$\begin{aligned}[\hat\epsilon(\vec k,\omega)]_{\vec K\vec K'}&=\epsilon_0\delta_{\vec K,\vec K'}-\frac{e^2}{v|\vec k+\vec K|^2}\\&\quad\times\sum_{\Gamma,\Gamma'}\langle\vec K|u_\Gamma(\vec k)\rangle\Pi_{\Gamma\Gamma'}(\omega,\vec k)\langle u_{\Gamma'}(\vec k)|\vec K'\rangle\ ,\end{aligned} \tag{2.2}$$

where

$$\langle\vec K|u_\Gamma(\vec k)\rangle=\int_v d\vec x\, e^{i(\vec k+\vec K)\cdot\vec x}\varphi^*_{n\vec p}(\vec x)\varphi_{m\vec p-\vec k}(\vec x) \tag{2.3}$$

with $\varphi_{n\vec p}(\vec x)$ being the Bloch functions [of band index n and quasimomentum $\vec p\in$ BZ (Brillouin zone)] of the self-consistent Hartree-Fock (HF) problem in the absence of the external potential. (The self-consistency being achieved for the ground-state.) Here Γ denotes the ensemble of indexes $(n,m,\vec p)$ and v is the volume of the elementary cell.

According to Alder[5] and Wiser[6] the macroscopic dielectric constant $\epsilon(\vec k,\omega)$ is defined as

$$\epsilon(\vec k,\omega)=\frac{1}{[\hat\epsilon^{-1}(\vec k,\omega)]_{00}}\ . \tag{2.4}$$

Use of Eq. (2.2) here gives

$$\frac{\epsilon(\vec k,\omega)}{\epsilon_0}-1=-\frac{e^2}{\epsilon_0 v\vec k^2}\sum_{\Gamma,\Gamma'}\langle 0|u_\Gamma(\vec k)\rangle\left[\frac{1}{1-\Pi\zeta}\Pi\right]\times\langle u_{\Gamma'}(\vec k)|0\rangle\ , \tag{2.5}$$

where a matrix notation is used with respect to the indexes Γ,Γ' with

$$\zeta_{\Gamma\Gamma'}\equiv\frac{e^2}{\epsilon_0 v}\sum_{\vec K(\neq\vec 0)}\frac{\langle u_\Gamma(\vec k)|\vec K\rangle\langle\vec K|u_{\Gamma'}(\vec k)\rangle}{(\vec k+\vec K)^2}\ . \tag{2.6}$$

We are interested in the $\vec k\to\vec 0$ limit of Eq. (2.5).

The existence of this limit is assured due to the fact that the polarization part has no singularity at $\vec k=\vec 0$ and obeys the identities

$$\sum_{n,\vec p}[\Pi(\omega,0)]_{nn\vec p,n'm'\vec p'}=\sum_{n',\vec p'}[\Pi(\omega,0)]_{nm\vec p,n'n'\vec p'}=0\ , \tag{2.7a}$$

$$\sum_{\substack{n,\vec p,\\ n',\vec p'}}\left[\frac{\partial}{\partial k_\mu}[\Pi(\omega,\vec k)]_{nn\vec p,n'n'\vec p'}\right]_{\vec k=\vec 0}=0\ , \tag{2.7b}$$

which are a consequence of the charge conservation. Using these identities, the expansion in powers of $\vec k$

$$\begin{aligned}\Pi(\omega,\vec k)&=\Pi^{(0)}(\omega)+k^\mu\Pi^{(1)}_\mu(\omega)\\&\quad+k^\mu k^\nu\Pi^{(2)}_{\mu\nu}(\omega)+\cdots\ ,\end{aligned}$$

$$\langle 0|u(\vec k)\rangle\equiv v(\vec k)=v^{(0)}+k^\mu v^{(1)}_\mu+\cdots\ ,$$

$$\zeta(\vec k)=\zeta^{(0)}+\cdots\ ,$$

and also that

$$v^{(0)}_{nm\vec p}=\delta_{n,m}\ ,$$

one can derive

$$\begin{aligned}\frac{\epsilon(0,\omega)}{\epsilon_0}-1=-\frac{e^2}{\epsilon_0 v}\sum_{\Gamma,\Gamma'}\Bigg[&\vec e\cdot\vec v^{(1)}_\Gamma\left[\frac{1}{1-\Pi^{(0)}(\omega)\zeta^{(0)}}\Pi^{(0)}(\omega)\right]_{\Gamma\Gamma'}\vec e\cdot\vec v^{(1)*}_{\Gamma'}+\vec e\cdot\vec v^{(1)}_\Gamma\left[\frac{1}{1-\Pi^{(0)}(\omega)\zeta^{(0)}}\vec\Pi^{(1)}(\omega)\cdot\vec e\right]_{\Gamma\Gamma'}v^{(0)*}_{\Gamma'}\\&+v^{(0)}_\Gamma\left[\vec\Pi^{(1)}(\omega)\cdot\vec e\,\frac{1}{1-\zeta^{(0)}\Pi^{(0)}(\omega)}\right]_{\Gamma\Gamma'}\vec e\cdot\vec v^{(1)*}_{\Gamma'}\\&+v^{(0)}_\Gamma\left[\vec e\cdot\hat\Pi^{(2)}(\omega)\cdot\vec e+\vec e\cdot\hat\Pi^{(1)}(\omega)\zeta^{(0)}\frac{1}{1-\Pi^{(0)}(\omega)\zeta^{(0)}}\zeta^{(0)}\vec e\cdot\hat\Pi^{(1)}(\omega)\right]_{\Gamma\Gamma'}v^{(0)*}_{\Gamma'}\Bigg]\ ,\end{aligned} \tag{2.8}$$

where

$$\vec e=\frac{\vec k}{|\vec k|}\ .$$

In the following sections we will restrain the generality of the discussion by resorting to approximations for the polarization part Π; nevertheless some comments will be made also with regard to these general formulas.

The time-dependent, self-consistent Hartree-Fock approximation in the presence of the external potential is equivalent to the summation of all ladder diagrams for Π giving

$$[\Pi^{\mathrm{HF}}(\vec{k},\omega)]_{\Gamma\Gamma'} = -2\left[\frac{1}{1-\mathscr{K}(\vec{k},\omega)\mathscr{C}^{e}(\vec{k})}\mathscr{K}(\vec{k},\omega)\right]_{\Gamma\Gamma'} \tag{2.9}$$

with

$$[\mathscr{K}(\vec{k},\omega)]_{\Gamma\Gamma'} = -\delta_{\Gamma\Gamma'}\frac{\Theta(\mu-E_{n\vec{p}})-\Theta(\mu-E_{m,\vec{p}-\vec{k}})}{E_{n\vec{p}}-E_{m,\vec{p}-\vec{k}}+\hbar\omega-i0}\,, \tag{2.10}$$

where $E_{n\vec{p}}$ is the band-energy, μ is the chemical potential and

$$[\mathscr{C}^{e}(\vec{k})]_{\Gamma\Gamma'} = \frac{e^2}{4\pi\epsilon_0}\int d\vec{x}\int_v d\vec{x}\,'\varphi_{n\vec{p}}(\vec{x})\varphi^{*}_{m\vec{p}-\vec{k}}(\vec{x}\,')\frac{1}{|\vec{x}-\vec{x}\,'|}\varphi^{*}_{n'\vec{p}'}(\vec{x})\varphi_{m',\vec{p}'-\vec{k}}(\vec{x}\,') \tag{2.11}$$

is a type of exchange Coulomb matrix. [The factor of 2 in Eq. (2.9) comes from the spin.]

Introducing Eq. (2.9) in Eq. (2.5), one finds

$$\frac{\epsilon^{\mathrm{HF}}(\vec{k},\omega)}{\epsilon_0}-1 = \frac{2e^2}{\epsilon_0 v k^2}\sum_{\Gamma,\Gamma'}(0|u_\Gamma(\vec{k}))\left[\frac{1}{1+\mathscr{K}(\vec{k},\omega)[2\zeta(\vec{k})-\mathscr{C}^{e}(\vec{k})]}\mathscr{K}(\vec{k},\omega)\right]_{\Gamma\Gamma'}(u_{\Gamma'}(\vec{k})|0)\,. \tag{2.12}$$

The time-dependent self-consistent potential approximation is equivalent to omitting all the exchange effects, i.e., ignoring $\mathscr{C}^{e}$ in Eq. (2.9) and considering the Bloch functions $\varphi_{n\vec{p}}$ as solutions of the unperturbed self-consistent potential problem. This is nothing but the random-phase approximation (RPA) for the polarization diagram, not to be confused with the RPA approximation for the dielectric constant itself, which implies ignoring also ζ. The appearance of ζ, due to the inversion procedure (2.4), is often referred to as the "local-field" correction.

Within the HF approximation we are interested again in the $\vec{k}\rightarrow 0$ limit. It can be shown that Eq. (2.8) becomes

$$\begin{aligned}\frac{\epsilon^{\mathrm{HF}}(0,\omega)}{\epsilon_0}-1 = \frac{2e^2}{v\epsilon_0}\sum_{\Gamma,\Gamma'}\Bigg[&\vec{e}\cdot\vec{v}^{(1)}_{\Gamma}\left[\frac{1}{1+\mathscr{K}^{(0)}(\omega)(2\zeta^{(0)}-\mathscr{C}^{e(0)})}\mathscr{K}^{(0)}(\omega)\right]_{\Gamma\Gamma'}\vec{e}\cdot\vec{v}^{(1)*}_{\Gamma'}\\ &+\vec{e}\cdot\vec{v}^{(1)}_{\Gamma}\left[\frac{1}{1+\mathscr{K}^{(0)}(\omega)(2\zeta^{(0)}-\mathscr{C}^{e(0)})}\vec{\mathscr{K}}^{(1)}(\omega)\cdot\vec{e}\right]_{\Gamma\Gamma'}v^{(0)*}_{\Gamma'}\\ &+v^{(0)}_{\Gamma}\left[\vec{\mathscr{K}}^{(1)}(\omega)\cdot\vec{e}\frac{1}{1-\mathscr{K}^{(0)}(\omega)(2\zeta^{(0)}-\mathscr{C}^{e(0)})}\right]\vec{e}\cdot\vec{v}^{(1)*}_{\Gamma'}\\ &+v^{(0)}_{\Gamma}\left[\vec{e}\cdot\hat{\mathscr{K}}^{(2)}(\omega)\cdot\vec{e}-2\vec{e}\cdot\vec{\mathscr{K}}^{(1)}(\omega)\zeta^{(0)}\frac{1}{1+\mathscr{K}^{(0)}(\omega)(2\zeta^{(0)}-\mathscr{C}^{e(0)})}\vec{e}\cdot\vec{\mathbf{K}}^{(1)}(\omega)\right]_{\Gamma\Gamma'}v^{(0)*}_{\Gamma'}\Bigg]\,,\end{aligned} \tag{2.13}$$

where

$$\mathscr{C}^{e(0)} \equiv \mathscr{C}^{e}(\vec{k})|_{\vec{k}=\vec{0}}\,,$$

$\mathscr{K}^{(0)}$, $\mathscr{K}^{(1)}_{\mu}$, $\mathscr{K}^{(2)}_{\mu\nu}$ are the corresponding terms from the $\vec{k}$ expansion of $\mathscr{K}(\omega,\vec{k})$, or explicitly

$$[\mathscr{K}^{(0)}(\omega)]_{\Gamma\Gamma'} = \delta_{\Gamma\Gamma'}[\mathscr{K}^{(0)}(\omega)]_{\Gamma}\,,$$

$$[\mathscr{K}^{(0)}(\omega)]_{nm\vec{p}} = -\frac{\Theta(\mu-E_{n\vec{p}})-\Theta(\mu-E_{m\vec{p}})}{E_{n\vec{p}}-E_{m\vec{p}}+\hbar\omega-i0}\,,$$

$$\sum_{\Gamma'}\vec{e}\cdot[\vec{\mathscr{K}}^{(1)}(\omega)]_{\Gamma\Gamma'}v^{(0)*}_{\Gamma'} = \sum_{\Gamma'}v^{(0)}_{\Gamma'}\vec{e}\cdot[\vec{\mathscr{K}}^{(1)}(\omega)]_{\Gamma'\Gamma} \tag{2.14}$$

$$= \frac{1}{\hbar\omega}\vec{e}\frac{\partial E_{n\vec{p}}}{\partial\vec{p}}\delta(\mu-E_{n\vec{p}})\,,$$

$$\sum_{\Gamma,\Gamma'}v^{(0)}_{\Gamma}\vec{e}\hat{\mathscr{K}}^{(2)}_{\Gamma\Gamma'}\vec{e}v^{(0)*}_{\Gamma'} = -\frac{1}{(\hbar\omega)^2}\left[\vec{e}\frac{\partial E_{n\vec{p}}}{\partial\vec{p}}\right]^2\delta(\mu-E_{n\vec{p}})\,. \tag{2.14}$$

In the case of the insulators, where the chemical potential μ is situated in an energy gap, only the first term of Eq. (2.13) survives; the others appear only in the case of metals, carrying a typical $1/\omega^2$ singularity at $\omega=0$.

Although we are interested mostly in the insulator problem, where a static dielectric constant also exists, we cannot *a priori* ignore these spurious terms. Indeed, the gap of most real dielectrics is a so-called Bloch gap, i.e., it is not related to the atomic gap. When the lattice constant is increased, the gap disappears, reappearing later as an atomic gap. Correspondingly, the system may undergo several metal-insulator transitions (the chemical potential being in the band or the gap). Since we are interested in the study of the limit $a\rightarrow\infty$, we must keep all the terms and consider $\omega\neq 0$. Nevertheless, as we shall see later, other arguments help to eliminate these complications.

In the following sections we shall base our discussion on Eq. (2.13). On the other hand, since as we shall see, no specific role is played by the exchange terms, all the results hold also in the simple self-consistent potential approximation.

III. QUANTUM THEORY OF THE ATOMIC POLARIZABILITY

If one considers the linear response of the system of interacting electrons belonging to a given atom with respect to a homogeneous monochromatic external field $\vec{\mathscr{E}}e^{i\omega t}$ at $T=0$ K, one may compute the induced dipole moment

$$p_\mu = e\int d\vec{x}\, \vec{x}_\mu \langle n(\vec{x},t)\rangle \ .$$

One defines then the atomic polarizability κ^{at} through the coefficient of proportionality between the density of induced dipole moments $\vec{P}=\vec{p}/v$, where v is the volume attributed to one atom (in the crystalline arrangement of the atoms in a Bravais lattice it would correspond to the volume of the cell), and the external field

$$\vec{P}(t)=\epsilon_0\kappa(\omega)\vec{\mathscr{E}}e^{i\omega t} \ . \tag{3.1}$$

The resulting formula is

$$\kappa(\omega) = -\frac{e^2}{\epsilon_0 v}\int d\vec{x}\int d\vec{x}\,'(\vec{e}\cdot\vec{x})(\vec{e}\cdot\vec{x}\,')\frac{1}{i\hbar} \times\int_0^\infty dt\, e^{i(\omega - i0)t} \times\langle[n(\vec{x},t)_H, n(\vec{x}\,')]\rangle_0 \ , \tag{3.2}$$

where $\vec{e}$ is an arbitrary unit vector ($\vec{e}\,^2=1$). The retarded Green function in Eq. (3.2) of course can be related to the Coulomb operator and the polarization diagram, giving rise to

$$\kappa^{at}(\omega) = -\frac{e^2}{v\epsilon_0}\sum_{\gamma,\gamma'}\vec{e}\cdot\vec{v}_\gamma^{(1)}\left[\frac{1}{1-\Pi^{at}(\omega)\mathscr{C}^{at\,d}}\Pi^{at}(\omega)\right]_{\gamma\gamma'} \times(\vec{e}\cdot\vec{v}_{\gamma'}^{(1)})^* \ , \tag{3.3}$$

where

$$\vec{v}_\gamma^{(1)} \equiv i\int d\vec{x}\,\varphi_i^*(\vec{x})\vec{x}\varphi_j(\vec{x}) \tag{3.4}$$

with $\varphi_i(\vec{x})$ being the atomic wave functions of the self-consistent Hartree-Fock problem in the absence of the external potential (the self-consistency being achieved for the ground state). Here γ denotes the ensemble of atomic indexes (i,j), and

$$\mathscr{C}^{at\,d}_{ij;i'j'} = \frac{e^2}{4\pi\epsilon_0}\int d\vec{x}\int d\vec{x}\,'\varphi_i(\vec{x})\varphi_j^*(\vec{x})\frac{1}{|\vec{x}-\vec{x}\,'|} \times\varphi_{i'}^*(\vec{x}\,')\varphi_{j'}(\vec{x}\,') \tag{3.5}$$

is the direct Coulomb matrix. (Actually the Feynman graph technique applies only for the non-degenerate ground state—in the atomic case it means full shell occupation.)

The time-dependent, self-consistent Hartree-Fock approximation in the presence of the external potential is given, similarly to the crystalline case, by

$$[\Pi^{at\,HF}(\omega)]_{\gamma\gamma'} = -2\left[\frac{1}{1-\mathscr{K}^{at}(\omega)\mathscr{C}^{at\,e}}\mathscr{K}^{at}(\omega)\right]_{\gamma\gamma'} \tag{3.6}$$

with

$$[\mathscr{K}^{at}(\omega)]_{\gamma\gamma'} \equiv -\delta_{\gamma\gamma'}\frac{\Theta(\mu-\epsilon_i)-\Theta(\mu-\epsilon_j)}{\epsilon_i-\epsilon_j+\hbar\omega-i0} \ , \tag{3.7}$$

where ϵ_i is the atomic energy and

$$\mathscr{C}^{at\,e}_{\gamma\gamma'} = \frac{e^2}{4\pi\epsilon_0}\int d\vec{x}\int d\vec{x}\,'\varphi_i(\vec{x})\varphi_j^*(\vec{x}\,')\frac{1}{|\vec{x}-\vec{x}\,'|} \times\varphi_{i'}^*(\vec{x})\varphi_{j'}(x') \tag{3.8}$$

is the Coulomb exchange matrix.

Introducing Eq. (3.6) into Eq. (3.3) gives

$$\kappa^{at\,HF}(\omega) = \frac{2e^2}{\epsilon_0 v}\sum_{\gamma,\gamma'}\vec{e}\cdot\vec{v}_\gamma^{(1)}\left[\frac{1}{1+\mathscr{K}^{at}(\omega)(2\mathscr{C}^{at\,d}-\mathscr{C}^{at\,e})}\mathscr{K}^{at}(\omega)\right]_{\gamma\gamma'}\vec{e}\cdot\vec{v}_{\gamma'}^{(1)*} \ . \tag{3.9}$$

A comparison of the formulas of this section with those of the preceding one shows much similitude underlined by our choice of the notations. We were careful of course to introduce similar entities with similar (HF) approximations. This is a very important point to be emphasized.

In our opinion, in comparing crystalline entities with atomic ones, one must stick to the same approximation. A striking example is given by the case of the monoelectronic atom. In this case we know exactly the atomic polarizability as given by the dipole formula

$$\frac{e^2}{\epsilon_0 v}\sum_{\gamma,\gamma'}\vec{e}\cdot\vec{v}_\gamma^{(1)}[\mathscr{K}^{at}(\omega)]_{\gamma\gamma'}\vec{e}\cdot\vec{v}_{\gamma'}^{(1)}$$

(where, however, the true one-electronic wave functions and energies appear). The time-dependent HF approximation Eq. (3.9) for this problem as can be shown, would give the same result for spinless electrons (in spite of the formal differences), but it introduces a spurious Coulomb interaction for spinning electrons. The same approximation may become quantitatively a good one in the crystal (where no exact solution is available), but from our point of view these qualities are irrelevant for our discussion. It would be meaningful, of course, to compare the exact results, but it is meaningless to compare entities from different approximation schemes and levels.

An important feature of the atomic polarizability is that its dependence on the average distance between the atoms is only through the factor $1/v$ in front of Eqs. (3.3) or (3.9). It can be shown through a dimensional analysis that

$$\kappa^{at}(\omega) = \left[\frac{r_0}{a}\right]^3\bar{\kappa}^{at}\left[\frac{\hbar\omega}{e^2/4\pi\epsilon_0 r_0}\right] \ , \tag{3.10}$$

where $\bar{\kappa}^{at}(x)$ is a universal function (not depending on any physical constant except Z—the number of electrons per atom).

Here the case of a simple cubic arrangement of atoms was considered where $v=a^3$ and r_0 denotes the Bohr radius. Equation (3.10) shows that increasing a or decreasing r_0 are equivalent only for the zero-frequency case. In what follows, we shall vary the parameter r_0/a only through the increase of the lattice constant a.

IV. CLAUSIUS-MOSOTTI LIMIT

With the use of Eq. (3.10), the Clausius-Mosotti formula for crystals of cubic symmetry can be written as

$$\frac{\epsilon^{\text{CM}}}{\epsilon_0}-1=\frac{(r_0/a)^3\bar{\kappa}^{\text{at}}}{1-\frac{1}{3}(r_0/a)^3\bar{\kappa}^{\text{at}}} \ . \tag{4.1}$$

Then one is confronted with the dilemma of how to reconcile the idea that this nonlinear relation supposedly holds for $r_0/a \to 0$, with the obvious relation

$$\lim_{r_0/a\to 0}\left[\frac{\epsilon^{\text{CM}}}{\epsilon_0}-1\right]\left[\frac{a}{r_0}\right]^3=\bar{\kappa}^{\text{at}}$$

that forbids the discernibility of the nonlinearity in the same limit.

The only conceivable conjecture is that the quantum-mechanical expression of the dielectric constant of a simple cubic crystal has the same asymptotic series in the parameter r_0/a as that given by Eq. (4.1). Otherwise stated

$$\epsilon \sim \epsilon^{\text{CM}} ,$$

where $\sim$ stands for equality up to terms that decrease faster than any power of r_0/a. This is a meaningful mathematical statement that we shall try to prove in the frame of the approximations described in the previous sections.

Unfortunately, the straightforward proof must be supplemented with some assumptions about the properties of the unperturbed (equilibrium) Hartree-Fock solution, which is not explicitly known. These assumptions (generally accepted in the literature) are the following:

(a) The bands tend to the atomic levels faster than a power law.

(b) The restrictions of the Bloch functions on the cell tend to the atomic functions faster than a power law. Under this form they are acceptable for those bands that correspond to the negative part of the atomic spectrum (bound states). The analysis of simple Kronig-Penney models shows that they hold for the negative spectrum, while for the positive part of the atomic spectrum these statements are meaningless. For these states a much more sophisticated mathematical property should hold, which we are unable to formulate. We are compelled therefore to ignore that part of the spectrum. In this respect, however, our state of the art is identical with that of all the other approaches, where these assumptions are brought in under the form of the tight-binding scheme.

Under the assumptions (a) and (b), for our asymptotic purpose we may consider $\partial\varphi_{n\vec p}/\partial\vec p$ and $\partial E_{n\vec p}/\partial\vec p$ as vanishing. As a consequence Eq. (2.13) reduces to the first term. Therefore, irrespective of the position of the Fermi level, for nonvanishing frequencies, we remain only with the expression that is typical for insulators.

We may also replace the crystal energies $E_{n\vec p}$ and wave functions $\varphi_{n\vec p}(\vec x)$ by the corresponding atomic energies and wave functions ϵ_n and $\varphi_n^{\text{at}}(\vec x)$. (It is understood that the lattice constant is already so big that we encounter no more band crossings.) In applying this procedure we must be careful, however, to put the expression under a form in which only restrictions of the wave functions on the elementary cell appear. Therefore we must rewrite the definition of $\mathcal{C}^{e(0)}$ [see Eq. (2.11)] in the form

$$\mathcal{C}^{e(0)}_{\Gamma\Gamma'}\equiv\frac{e^2}{4\pi\epsilon_0}\sum_{\vec r}\int_v d\vec x\int_v d\vec x\,'\varphi_{n\vec p}(\vec x)\varphi^*_{m\vec p}(\vec x\,')\frac{e^{i(\vec p-\vec p\,')\cdot\vec r}}{|\vec x-\vec x\,'+\vec r\,|}\varphi^*_{n'\vec p\,'}(\vec x)\varphi_{m'\vec p\,'}(\vec x\,') \ . \tag{4.2}$$

Then

$$\mathcal{C}^{e(0)}_{\Gamma\Gamma'}\sim\frac{e^2}{4\pi\epsilon_0}\sum_{\vec r}\int d\vec x\int d\vec x\,'\varphi^{\text{at}}_n(\vec x)\varphi^{\text{at}*}_m(\vec x\,')\frac{e^{i(\vec p-\vec p\,')\cdot\vec r}}{|\vec x-\vec x\,'+\vec r\,|}\varphi^{\text{at}*}_{n'}(\vec x)\varphi^{\text{at}}_{m'}(\vec x\,') \tag{4.3}$$

and also

$$\zeta^{(0)}_{\Gamma\Gamma'}\sim\frac{e^2}{\epsilon_0 v}\sum_{\vec K\neq 0}\frac{1}{\vec K^2}\int d\vec x\, e^{i\vec K\cdot\vec x}\varphi^{\text{at}}_n(\vec x)\varphi^{\text{at}*}_m(\vec x)\int d\vec x\,'e^{-i\vec K\cdot\vec x\,'}\varphi^{\text{at}}_{n'}(\vec x\,')^*\varphi^{\text{at}}_{m'}(\vec x\,')\equiv\zeta^{\text{at}}_{\gamma\gamma'} \ , \tag{4.4}$$

$$\vec v^{\,(1)}_{\Gamma}\sim i\int d\vec x\,\varphi^{\text{at}*}_n(\vec x)\vec x\varphi^{\text{at}}_m(\vec x)\equiv\vec v^{\,(1)}_{\gamma} \ . \tag{4.5}$$

Here after the introduction of the atomic wave functions the integration over the cell has been extended to the whole space. Such an extension is allowed due to the faster-than-power-law decay of the atomic wave functions.

Now we see that the dependence on the quasimomenta survives only in $\mathcal{C}^{e(0)}$ according to Eq. (4.3). On the other hand, since the summation symbol is

$$\sum_{\Gamma}\cdots\equiv\sum_{n,m}\frac{v}{(2\pi)^3}\int_{\text{BZ}}d\vec p\cdots \tag{4.6}$$

while

$$\frac{v}{(2\pi)^3}\int_{\text{BZ}}d\vec p=1 \ ,$$
$$\frac{v}{(2\pi)^3}\int_{\text{BZ}}d\vec p\,e^{i\vec p\cdot\vec r}=\delta_{\vec r,\vec 0} \ , \tag{4.7}$$

we may replace the summation symbol (in all the matrix manipulations) with

$$\sum_{\gamma}\equiv\sum_{n,m}$$

simultaneously with the replacement of $\mathcal{C}^{e(0)}_{\Gamma\Gamma'}$ with

$$\mathscr{C}^{\mathrm{at}\,e}_{\gamma\gamma'} \equiv \frac{e^2}{4\pi\epsilon_0}\int d\vec{x}\int d\vec{x}\,'\varphi^{\mathrm{at}}_n(\vec{x})\varphi^{\mathrm{at}\,*}_m(\vec{x}\,')\frac{1}{|\vec{x}-\vec{x}\,'|}\varphi^{\mathrm{at}\,*}_{n'}(\vec{x})\varphi^{\mathrm{at}}_{m'}(\vec{x}\,') \,. \tag{4.8}$$

Therefore we arrive at the formula

$$\frac{\epsilon^{\mathrm{HF}}(0,\omega)}{\epsilon_0}-1\sim\frac{2e^2}{v\epsilon_0}\sum_{\gamma,\gamma'}\vec{e}\cdot\vec{v}^{(1)}_{\gamma}\left[\frac{1}{1+\mathscr{K}^{\mathrm{at}}(\omega)(2\zeta^{\mathrm{at}}-\mathscr{C}^{\mathrm{at}\,e})}\mathscr{K}^{\mathrm{at}}(\omega)\right]_{\gamma\gamma'}\vec{e}\cdot\vec{v}^{(1)*}_{\gamma'} \,, \tag{4.9}$$

which resembles Eq. (3.9) for the atomic polarizability. The only difference is the appearance of ζ^{at} instead of the direct Coulomb matrix $\mathscr{C}^{\mathrm{at}\,d}$. On the other hand, it is obvious from the inspection of its definition Eq. (4.4) that

$$\lim_{a\to\infty}\zeta^{\mathrm{at}}=\mathscr{C}^{\mathrm{at}\,d} \,. \tag{4.10}$$

A hasty superficial conclusion would be then, that actually we obtained a linear relationship between the dielectric constant and the atomic polarizability. However, this is not so. Equation (4.10) says nothing about how fast is the limit achieved and this is crucial for the discussion of the Clausius-Mosotti limit. Actually we shall prove the following lemma,

$$\zeta^{\mathrm{at}}_{\gamma\gamma'}\sim\mathscr{C}^{\mathrm{at}\,d}_{\gamma\gamma'}-\frac{e^2}{3v\epsilon_0}\vec{v}^{(1)*}_{\gamma}\vec{v}^{(1)}_{\gamma'} \,, \tag{4.11}$$

which says that ζ^{at} tends toward $\mathscr{C}^{\mathrm{at}\,d}$ faster than any power of r_0/a only after the subtraction of a term that falls as $(r_0/a)^3$.

From the Poisson formula

$$\frac{1}{a^3}\sum_{\vec{K}}^{1/a}\hat{g}(\vec{K})=\frac{1}{(2\pi)^3}\sum_{\vec{r}}^{a}g(\vec{r}) \tag{4.12}$$

(where

$$g(\vec{x})\equiv\int d\vec{k}\,e^{-i\vec{k}\cdot\vec{x}}\hat{g}(\vec{k})$$

and the limits on the summation signs mean that the summations are performed over the lattice of constant a and the reciprocal lattice of constant $1/a$) we may conclude that

$$\frac{1}{a^3}\sum_{\vec{K}}^{1/a}\hat{g}(\vec{K})\sim\frac{1}{(2\pi)^3}\int d\vec{k}\,\hat{g}(\vec{k}) \tag{4.13}$$

if $g(\vec{x})$ decreases faster than any power law, or equivalently if $\hat{g}(\vec{k})$ is continuous, indefinitely differentiable, decreasing together with its derivatives faster than any power. On the other hand, we have for $\zeta^{\mathrm{at}}_{\gamma\gamma'}$ an expression of the form

$$\frac{1}{a^3}\sum_{\vec{K}(\neq\vec{0})}^{1/a}\hat{F}(\vec{K})\frac{1}{\vec{K}^2} \,,$$

where $\hat{F}(\vec{k})$ is continuous, indefinitely differentiable in $\vec{k}$, decreasing together with its derivatives faster than any power and vanishing quadratically at the origin. Owing to the singularity of the summand in the origin, we cannot apply directly Eq. (4.13). Let us subtract the behavior $\vec{k}\hat{A}\vec{k}$ of $\hat{F}(\vec{k})$ around the origin in the following way:

$$\frac{\hat{F}(\vec{k})}{\vec{k}^2}=\frac{\vec{k}\cdot\hat{A}\cdot\vec{k}}{\vec{k}^2}e^{-\alpha\vec{k}^2}+\frac{[\hat{F}(\vec{k})-\vec{k}\cdot\hat{A}\cdot\vec{k}]e^{-\alpha\vec{k}^2}}{\vec{k}^2}$$
$$+\frac{\hat{F}(\vec{k})}{\vec{k}^2}(1-e^{-\alpha\vec{k}^2}) \,,$$

where α is an arbitrary positive number. Only the first term does not satisfy the required conditions. However, due to the cubic symmetry,

$$\frac{1}{a^3}\sum_{\vec{K}(\neq\vec{0})}\frac{\vec{K}\cdot\hat{A}\cdot\vec{K}}{\vec{K}^2}e^{-\alpha\vec{K}^2}=\frac{1}{3a^3}(\mathrm{tr}\hat{A})\sum_{\vec{K}(\neq\vec{0})}e^{-\alpha\vec{K}^2}$$
$$=\frac{1}{3a^3}(\mathrm{tr}\hat{A})\sum_{\vec{K}}e^{-\alpha\vec{K}^2}$$
$$-\frac{1}{3a^3}\mathrm{tr}\hat{A} \,,$$

where the function under the sum again satisfies all the conditions.

Therefore

$$\frac{1}{a^3}\sum_{\vec{K}(\neq\vec{0})}^{1/a}\frac{\hat{F}(\vec{K})}{\vec{K}^2}\sim\frac{1}{(2\pi)^3}\int d\vec{k}\left[\frac{\hat{F}(\vec{k})-\vec{k}\cdot\hat{A}\cdot\vec{k}}{\vec{k}^2}e^{-\alpha\vec{k}^2}+\frac{\hat{F}(\vec{k})}{\vec{k}^2}(1-e^{-\alpha\vec{k}^2})+\tfrac{1}{3}(\mathrm{tr}\hat{A})e^{-\alpha\vec{k}^2}\right]-\frac{1}{3a^3}\mathrm{tr}\hat{A} \,.$$

But due to the spherical symmetry of integration

$$\int d\vec{k}\,\frac{\vec{k}\cdot\hat{A}\cdot\vec{k}}{\vec{k}^2}e^{-\alpha\vec{k}^2}=\tfrac{1}{3}(\mathrm{tr}\hat{A})\int d\vec{k}\,e^{-\alpha\vec{k}^2} \,,$$

we may regroup the terms and we have finally

$$\frac{1}{a^3}\sum_{\vec{K}(\neq\vec{0})}^{1/a}\frac{\hat{F}(\vec{K})}{\vec{K}^2}\sim\frac{1}{(2\pi)^3}\int d\vec{k}\,\frac{\hat{F}(\vec{k})}{\vec{k}^2}-\frac{1}{3a^3}\mathrm{tr}\hat{A} \,, \tag{4.14}$$

which gives immediately Eq. (4.11).

Now let us use Eq. (4.11) in Eq. (4.9). To this end it is important to observe that the asymptotic difference between ζ^{at} and $\zeta^{\mathrm{at}\,d}$ is a factorized matrix, therefore the formula expanded in formal powers of this matrix gives rise to products of the 3×3 matrix

$$\kappa_{\mu\nu}^{\mathrm{at\,HF}}=\frac{2e^2}{v\epsilon_0}\sum_{\gamma,\gamma'} v_{\gamma}^{(1)\mu}\left[\frac{1}{1+\mathscr{K}^{\mathrm{at}}(\omega)(2\zeta^{\mathrm{at}\,d}-\zeta^{\mathrm{at}\,e})}\times\mathscr{K}^{\mathrm{at}}(\omega)\right]_{\gamma\gamma'} v_{\gamma'}^{(1)*\nu}$$

$$(\mu,\nu=1,2,3) .$$

But, owing to the spherical symmetry of the atom

$$\kappa_{\mu\nu}^{\mathrm{at\,HF}}=\delta_{\mu\nu}\kappa^{\mathrm{at\,HF}} ,$$

and therefore we have after resummation

$$\frac{\epsilon^{\mathrm{HF}}(0,\omega)}{\epsilon_0}-1\sim\frac{\kappa^{\mathrm{at\,HF}}}{1-\frac{1}{3}\kappa^{\mathrm{at\,HF}}} , \tag{4.15}$$

which is exactly the proof of our conjecture.

V. DISCUSSION

We have seen how the formulation of the problem in terms of asymptotic series in r_0/a leads in a natural way to the Clausius-Mosotti formula. While the plausible assumptions about the asymptotic behavior of the Bloch functions and band widths are the same as generally accepted in the literature, a crucial point of our proof is the lemma [contained in Eq. (4.11)] about the asymptotic behavior of a certain lattice sum. In the absence of the guiding criterion of asymptotic equivalence, one could derive the most contradictory results, since ignoring terms that vanish only as a power law would modify completely the results. Therefore, those papers that accidentally ignored only terms that according to our lemma decrease faster than a power law have obtained the Clausius-Mosotti relation, while the others have not. Of course, in the identification of the Clausius-Mosotti relation it was important also to use the appropriate definition of the atomic polarizability. As we have shown, one must compare similar approximations for the crystal dielectric constant and atomic polarizability.

Our discussion was limited to the self-consistent, time-dependent Hartree-Fock and the simple self-consistent potential approximations. All the results hold in both approximations, irrespective to the presence or absence (in the spinless HF scheme) of a spurious self-interaction. We think that this is natural since only for pointlike charge distributions is self-interaction dangerous.

We expect that the Clausius-Mosotti relation in the above described asymptotic sense holds also between the exact crystal dielectric constant and the exact atomic polarizability, however no proof of such a general statement seems available. Moreover, an order by order perturbative argument for the irreducible polarization diagram Π leads to immediate difficulties. Indeed, a succesion of second-order photon self-energy diagrams (electron loop with two external potential lines) within will give rise to crystal convolutions of the exchange Coulomb terms and not products of atomic exchange Coulomb terms.

A weak point of our discussion is that it ignores the part of the spectrum that tends toward the positive atomic spectrum (not bound states). This is a serious shortcoming, but again it is common for all approaches in the literature. The inclusion of this part of the spectrum would require an adequate formulation of the asymptotic properties, which obviously will be less intuitive and even harder to prove.

We would like to mention here also our results on the analysis of the Clausius-Mosotti problem within an exactly solvable hopping model with self-consistent potential.[15] In that lattice problem the role of the atoms was played by the elementary cell to which were confined the "bound electrons." The Clausius-Mosotti relation (for zero frequency) was obtained in the limit, when the dimension of the cell goes to zero, while the total electronic charge on the cell increases, in such a way that their product is constant. (The existence of this limit unfortunately implied a regularization of the Coulomb interaction at the origin.)

It can be shown however, that the result of the present paper about the asymptotic validity of the Clausius-Mosotti relation holds exactly also within the above-mentioned model, eliminating the need of any artificial regularization procedure.

ACKNOWLEDGMENTS

One of the authors (L.B.) would like to thank Professor N. H. March, E. G. Maximov, and O. V. Dolgov for useful discussions.

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PHYSICAL REVIEW B VOLUME 29, NUMBER 9 1 MAY 1984

Meissner effect in gauge-invariant, self-consistent pairing theories

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(Received 30 September 1983)

Within the frame of a self-consistent pairing theory, with potential interelectron interaction, a gauge-invariant discussion of the Meissner effect is given. It is shown that the terms that are usually ignored, proportional to the variation of the order parameter due to the magnetic field, are indeed vanishing in the Coulomb gauge, provided that the pairing phase is locally stable. An instructive example is given, where due to the degeneracy of the phase, although a pairing gap exists, there is no Meissner effect at all.

I. INTRODUCTION

In the discussion of the Meissner effect within pairing theories, the usual choice of the gauge for the vector potential is the transverse (Coulomb) gauge. It is supposed that in this gauge (for vanishing wave vector) one may ignore the terms coming from the vector-potential dependence of the order parameter, while, otherwise, the contribution of these terms is admittedly essential for the gauge invariance of the result. This neglect is due to technical difficulties, rather than a clear physical or mathematical motivation. Within the frame of the self-consistent (SC) pairing theory, Rickayzen[1] tried to estimate the neglected terms. He concluded that for certain "potentials" the error is small. However, his analysis actually does not include at all true potentials that only depend on the distance between the electrons; therefore, the coordinate-space description is inapplicable.

Owing to the principal importance of the problem, we undertook an investigation within the frame of a SC gauge-invariant pairing theory with arbitrary translational-invariant potential. Our conclusion is that the ignored terms are rigorously vanishing, provided the pairing phase satisfies a certain local stability condition. On the contrary, if this is not satisfied, these terms may acquire importance and even totally compensate the Meissner effect of the naive theory. This is illustrated on an exactly solvable model that exhibits a pairing gap, but has no Meissner effect.

II. DESCRIPTION OF THE SC PAIRING MODEL

We consider an interelectron potential interaction (in the second-quantization formalism)

$$\int d\vec{x} \int d\vec{x}' V(|\vec{x}-\vec{x}'|)\psi^{\dagger}(\vec{x})\psi^{\dagger}(\vec{x}')\psi(\vec{x}')\psi(\vec{x}) \tag{2.1}$$

as a prototype model for superconductivity. Although perhaps too crude, it is the simplest model that might show superconductive properties. To the study of the equilibrium properties of such a pure electron-electron interaction, one may apply the Hartree-Fock-Bogoliubov (HFB) approximation scheme, which is the best one-electron approximation for the grand-canonical potential. As is well known, the HFB solution corresponds to taking all the simple contractions (including the anomalous ones) in Eq. (2.1) and subtracting a constant term obtained by taking all the double contractions.

For the sake of simplicity, as it is customary, we shall ignore all the normal contractions. Then our SC Hamiltonian in the presence of a magnetic field is

$$\begin{aligned}\mathscr{H}=&\sum_{\sigma}\int d\vec{x}\,\psi_{\sigma}^{\dagger}(\vec{x})\left[\frac{1}{2m}\left[-i\hbar\vec{\nabla}-\frac{e}{c}\vec{A}(\vec{x})\right]^{2}-\mu\right]\psi_{\sigma}(\vec{x})\\&+\int d\vec{x}\int d\vec{x}' V(|\vec{x}-x'|)[\langle\psi_{\uparrow}^{\dagger}(\vec{x})\psi_{\downarrow}^{\dagger}(\vec{x}')\rangle\psi_{\downarrow}(\vec{x}')\psi_{\uparrow}(\vec{x})+\langle\psi_{\downarrow}(\vec{x}')\psi_{\uparrow}(\vec{x})\rangle\psi_{\uparrow}^{\dagger}(\vec{x})\psi_{\downarrow}^{\dagger}(\vec{x}')\\&-\langle\psi_{\uparrow}^{\dagger}(\vec{x})\psi_{\downarrow}(\vec{x}')\rangle\langle\psi_{\downarrow}(\vec{x}')\psi_{\uparrow}(\vec{x})\rangle]\,,\end{aligned} \tag{2.2}$$

where the SC nature of $\mathscr{H}$ is determined by the presence of the Gibbs average

$$\langle\,\rangle=\mathrm{Tr}(e^{-\beta\mathscr{H}}\cdots)/\mathrm{Tr}e^{-\beta\mathscr{H}}\,. \tag{2.3}$$

Actually, the magnetic field must be interpreted here as the total SC field (the sum of the external and internal ones). It is considered, as usual, that the omission of the coupling of the magnetic field to the spin is not very important in the discussion of the Meissner effect.

Owing to the existence of the anomalous contractions, this Hamiltonian does not commute with the charge operator, and consequently, the current density,

$$\vec{j}_A(\vec{x})=\frac{e}{2m}\left[\psi^\dagger(\vec{x})\left(-i\hbar\vec{\nabla}-\frac{e}{c}\vec{A}(\vec{x})\right)\psi(x)+\text{H.c.}\right], \tag{2.4}$$

is not conserved. Nevertheless, it can be shown that the average current density is conserved,

$$\vec{\nabla}\langle\vec{j}_A(\vec{x})\rangle=\vec{0}\,. \tag{2.5}$$

This model is also gauge invariant (see Rickayzen[2]) in the sense that if

$$\langle\psi_\downarrow(\vec{x}')\psi_\uparrow(\vec{x})\rangle_{A_\mu}$$

is the anomalous contraction for the vector potential A_μ, then

$$\langle\psi_\downarrow(\vec{x}')\psi_\uparrow(\vec{x})\rangle_{A_\mu+\partial_\mu\chi}=e^{i(e/\hbar c)[\chi(\vec{x})+\chi(\vec{x}')]}\times\langle\psi_\downarrow(\vec{x}')\psi_\uparrow(\vec{x})\rangle_{A_\mu}\,. \tag{2.6}$$

This relation, in conjunction with

$$\mathscr{H}_{A_\mu+\partial_\mu\chi}[e^{i(e/\hbar c)\chi(\vec{x})}\psi(\vec{x})]=\mathscr{H}_{A_\mu}[\psi(\vec{x})]\,, \tag{2.7}$$

gives rise to the gauge independence of the observables.

Since the effect of the magnetic field will be studied only within first-order perturbation theory, we outline, here, the main features of the model in the absence of the magnetic field. Here one usually looks for solutions that are invariant under space translations, rotations, and spin rotations. Then (using the Bogoliubov transformation), one may diagonalize the Hamiltonian

$$\mathscr{H}_0=\sum_{\vec{k}}\left[E_{\vec{k}}(C^\dagger_{\vec{k}\uparrow}C_{\vec{k}\uparrow}+C^\dagger_{\vec{k}\downarrow}C_{\vec{k}\downarrow})+\epsilon_{\vec{k}}-E_{\vec{k}}+\frac{|\Delta_{\vec{k}}|^2}{2E_{\vec{k}}}[1-2f(E_{\vec{k}})]\right], \tag{2.8}$$

where

$$\epsilon_{\vec{k}}=\frac{\hbar^2\vec{k}^2}{2m}-\mu\,,$$

$$E_{\vec{k}}=+(\epsilon^2_{\vec{k}}+|\Delta_{\vec{k}}|^2)^{1/2},\quad f(E_{\vec{k}})=\frac{1}{e^{\beta E_k}+1}\,,$$

and the order parameter (or gap parameter) $\Delta_{\vec{k}}$ is the solution of the self-consistency equation

$$\Delta_{\vec{k}}=-\frac{1}{(2\pi)^3}\int d\vec{k}'\,\hat{V}(\vec{k}-\vec{k}')\frac{\Delta_{\vec{k}'}}{2E_{\vec{k}'}}[1-2f(E_{\vec{k}'})]\,, \tag{2.9}$$

with

$$\hat{V}(\vec{k})=\int d\vec{x}\,e^{i\vec{k}\cdot\vec{x}}V(\vec{x})\,.$$

The chemical potential has to be determined from the equation

$$\frac{1}{(2\pi)^3}\int d\vec{k}\left[1-\frac{\epsilon_{\vec{k}}}{E_{\vec{k}}}[1-2f(E_{\vec{k}})]\right]=\frac{\langle N\rangle}{\Omega}\,, \tag{2.10}$$

which fixes the average number of electrons in the volume Ω.

These results may actually be obtained as the solution of the variational problem

$$\delta\mathscr{F}^\Delta/\delta\Delta_{\vec{k}}=0 \tag{2.11}$$

for the grand-canonical potential

$$\mathscr{F}^\Delta=-\frac{1}{\Omega\beta}\ln\text{Tr}e^{-\beta\mathscr{H}_0^\Delta}\,, \tag{2.12}$$

with

$$\mathscr{H}_0^\Delta=\sum_{\vec{k}}[\epsilon_{\vec{k}}(a^*_{\vec{k}\uparrow}a_{\vec{k}\uparrow}+a^*_{-\vec{k}\downarrow}a_{-\vec{k}\downarrow})+\Delta_{\vec{k}}a_{-\vec{k}\downarrow}a_{\vec{k}\uparrow}+\Delta^*_{\vec{k}}a^\dagger_{\vec{k}\uparrow}a^\dagger_{-\vec{k}\downarrow}]$$
$$+\frac{1}{\Omega}\sum_{\vec{k},\vec{k}'}[\hat{V}(\vec{k}-\vec{k}')\langle a^\dagger_{\vec{k}'\uparrow}a^\dagger_{-\vec{k}'\downarrow}\rangle_0\langle a_{-\vec{k}\downarrow}a_{\vec{k}\uparrow}\rangle_0-\Delta_{\vec{k}'}\langle a_{-\vec{k}\downarrow}a_{\vec{k}\uparrow}\rangle_0-\Delta^*_{\vec{k}'}\langle a^\dagger_{\vec{k}\uparrow}a^\dagger_{-\vec{k}\downarrow}\rangle_0]\,. \tag{2.13}$$

This variational problem replaces the full HFB variational problem when normal propagators are ignored in the scheme.

Actually, one looks for a minimum of $\mathscr{F}^\Delta$, while Eq. (2.11) [or Eq. (2.9)] assures only stationarity, but we shall return later to this important aspect.

For $\Delta_{\vec{k}}$ satisfying Eq. (2.9), the grand-canonical potential takes the simple form

$$\mathscr{F}=-\frac{1}{\Omega\beta}\ln\text{Tr}e^{-\beta\mathscr{H}}$$
$$=\frac{1}{(2\pi)^3}\int d\vec{k}\left[\epsilon_{\vec{k}}-E_{\vec{k}}+\frac{2}{\beta}\ln[1-f(E_{\vec{k}})]+\frac{\Delta_{\vec{k}}}{2E_{\vec{k}}}[1-2f(E_{\vec{k}})]\right]. \tag{2.14}$$

[It is supposed that the potential $\hat{V}(\vec{k})$ vanishes sufficiently fast for $|\vec{k}| \to \infty$, and therefore $\Delta_{\vec{k}}$ also vanishes to assure the convergence of the integral.]

No exact solutions are known for the integral equation (2.9) (except a case that will be analyzed in Sec. V). However, for separable kernels, where instead of $\hat{V}(\vec{k}-\vec{k}')$ (depending on the single variable $|\vec{k}-\vec{k}'|$) we have had $U(\vec{k})U(\vec{k}')$, the equation is easily solvable. Such factorizable potentials are largely used in the literature, being a useful ingredient in most phenomenological applications by pointing out the essential features (even quantitative), that for the most part, depend only on the existence of a pairing gap around the Fermi level. On the other hand, through such an "approximation" the link with the initial problem in coordinate space is lost.

For a rigorous discussion of coordinate-space behavior, only true translation-invariant potentials should be considered. In what follows we will consider only such potentials, without sticking to any given candidate (with the exception of Sec. V), but only assuming that Eq. (2.9) has anomalous solutions (with $\Delta_{\vec{k}}$ not vanishing everywhere). Moreover, we shall assume that the minimum of the grand-canonical potential is attained with a $\Delta_{\vec{k}}$ which can be chosen to be real. (Otherwise there would actually be two equivalent minima.)

III. EQUILIBRIUM LINEAR RESPONSE TO THE MAGNETIC FIELD

We shall consider a small vector potential $\vec{A}(\vec{x})$ which is sufficiently well localized in space (it falls off sufficiently fast). Then, we may use the equilibrium linear-response formula

$$\langle X\rangle = \langle X\rangle_0 + \int_0^\beta d\lambda \langle [\mathscr{H}'(-i\hbar\lambda) - \langle \mathscr{H}'\rangle_0](X - \langle X\rangle_0)\rangle_0 \tag{3.1}$$

for the equilibrium average of an operator X, where $\mathscr{H}'$ is the small perturbation of the Hamiltonian. Keeping only terms linear in $\vec{A}$, we have, from (2.2),

$$\mathscr{H}' = -\frac{1}{c}\int d\vec{x}\, \vec{j}(\vec{x})\cdot\vec{A}(\vec{x}) + \int d\vec{x} \int d\vec{x}'\, V(\vec{x}-\vec{x}')[\eta(\vec{x},\vec{x}')\psi_\downarrow(\vec{x}') \times \psi_\uparrow(\vec{x}) + \text{H.c.}]\,, \tag{3.2}$$

where η is the variation of the anomalous propagator

$$\eta(\vec{x},\vec{x}') = \langle \psi_\uparrow^\dagger(\vec{x})\psi_\downarrow^\dagger(\vec{x}')\rangle - \langle \psi_\uparrow^\dagger(\vec{x})\psi_\downarrow^\dagger(\vec{x}')\rangle_0 \tag{3.3}$$

(actually, here also we keep only terms of first order in $\vec{A}$).

Also taking into account that the current-density operator of Eq. (2.4) has a term linear in $\vec{A}$, one finds for the Fourier transform of the average current density,

$$\langle \hat{j}^\mu_A(\vec{k})\rangle = \frac{e^2}{mc}\left\{\left[-\frac{\langle N\rangle}{\Omega}\delta_{\mu\nu} + \frac{2\hbar^2}{m}\frac{1}{(2\pi)^3}\int d\vec{p}\, p_\mu p_\nu S(\vec{k},\vec{p})\right]\hat{A}_\nu(\vec{k}) - \frac{2\hbar^2}{m}\frac{1}{(2\pi)^6}\int d\vec{q}\,\hat{V}(\vec{q})\int d\vec{p}\, p_\mu a(\vec{k},\vec{p})\nu(\vec{k},\vec{p}+\vec{q})\right\}, \tag{3.4}$$

where

$$S(\vec{k},\vec{p}) = \frac{1}{2}\left[\frac{1-f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}+E_{\vec{l}'}}\left[1-\frac{\epsilon_{\vec{l}}\epsilon_{\vec{l}'}+\Delta_{\vec{l}}\Delta_{\vec{l}'}}{E_{\vec{l}}E_{\vec{l}'}}\right] - \frac{f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}-E_{\vec{l}'}}\left[1+\frac{\epsilon_{\vec{l}}\epsilon_{\vec{l}'}+\Delta_{\vec{l}}\Delta_{\vec{l}'}}{E_{\vec{l}}E_{\vec{l}'}}\right]\right], \tag{3.5}$$

$$a(\vec{k},\vec{p}) = \frac{\epsilon_{\vec{l}'}\Delta_{\vec{l}}-\epsilon_{\vec{l}}\Delta_{\vec{l}'}}{2E_{\vec{l}}E_{\vec{l}'}}\left[\frac{1-f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}+E_{\vec{l}'}} + \frac{f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}-E_{\vec{l}'}}\right]$$

(with $\vec{l} = \vec{p}+\vec{k}/2$, $\vec{l}' = -\vec{p}+\vec{k}/2$), and

$$\nu(\vec{k},\vec{p}) = \frac{m}{2\hbar c}\left[\hat{\eta}\left[\frac{\vec{k}}{2}+\vec{p}, \frac{\vec{k}}{2}-\vec{p}\right] - \hat{\eta}^*\left[-\frac{\vec{k}}{2}+\vec{p}, -\frac{\vec{k}}{2}-\vec{p}\right]\right]. \tag{3.6}$$

We now also have to write the linear-response formula for $\nu(\vec{k},\vec{p})$, which is a self-consistency equation, to have an explicit relation between the average current density and the vector potential. This SC equation for ν appears as

$$\int d\vec{p}'[M(\vec{k})]_{\vec{p},\vec{p}'} d^{-1}(\vec{k},\vec{p}')\nu(\vec{k},\vec{p}') = \vec{p}\hat{\vec{A}}(\vec{k})a(\vec{k},\vec{p})\,, \tag{3.7}$$

where

$$[M(\vec{k})]_{\vec{p},\vec{p}'} = d(\vec{k},\vec{p})\delta(\vec{p}-\vec{p}') + \frac{1}{(2\pi)^3}d(\vec{k},\vec{p})\hat{V}(\vec{p}-\vec{p}')d(\vec{k},\vec{p}') \tag{3.8}$$

and

$$d(\vec{k},\vec{p})=\frac{1}{2}\left[\frac{1-f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}+E_{\vec{l}'}}\left(1+\frac{\epsilon_{\vec{l}}\epsilon_{\vec{l}'}+\Delta_{\vec{l}}\Delta_{\vec{l}'}}{E_{\vec{l}}E_{\vec{l}'}}\right)-\frac{f(E_{\vec{l}})-f(E_{\vec{l}'})}{E_{\vec{l}}-E_{\vec{l}'}}\left(1-\frac{\epsilon_{\vec{l}}\epsilon_{\vec{l}'}+\Delta_{\vec{l}}\Delta_{\vec{l}'}}{E_{\vec{l}}E_{\vec{l}'}}\right)\right] \tag{3.9}$$

(with $\vec{l}=\vec{p}+\vec{k}/2$, $\vec{l}'=-\vec{p}+\vec{k}/2$).

If the matrix $M(\vec{k})$ has no vanishing eigenvalues for the $\vec{k}$'s of interest, one may determine (formally) $\nu(\vec{k},\vec{p})$ and introduce its expression [containing the inverse of the matrix $M(\vec{k})$] in Eq. (3.4). One then finds

$$\langle \hat{j}^{\mu}_{A}(\vec{k})\rangle=K_{\mu\nu}(\vec{k})\hat{A}_{\nu}(\vec{k}) , \tag{3.10}$$

with

$$K_{\mu\nu}(\vec{k})=\frac{e^2}{mc}\left\{-\frac{\langle N\rangle}{\Omega}\delta_{\mu\nu}+\frac{2\hbar^2}{m}\frac{1}{(2\pi)^3}\left[\int d\vec{p}\,p_\mu p_\nu\left(S(\vec{k},\vec{p})-\frac{a(\vec{k},\vec{p})^2}{d(\vec{k},\vec{p})}\right)\right.\right.$$
$$\left.\left.+\int d\vec{p}\int d\vec{p}'p_\mu a(\vec{k},\vec{p})M^{-1}(\vec{k})_{\vec{p}\vec{p}'}a(\vec{k},\vec{p}')p'_\nu\right]\right\} . \tag{3.11}$$

From the conservation of the average current, gauge invariance, and invariance with respect to rotations, it follows that $K_{\mu\nu}(\vec{k})$ has the structure

$$K_{\mu\nu}(\vec{k})=(\delta_{\mu\nu}-k_\mu k_\nu/\vec{k}^2)\kappa(\vec{k}^2) . \tag{3.12}$$

This can be shown explicitly by straightforward, but lengthy algebra, that makes use of the useful identity

$$\frac{\hbar^2}{m}\vec{p}\cdot\vec{k}a(\vec{k},\vec{p})=\int d\vec{p}'[M(\vec{k})]_{\vec{p}\vec{p}'}d^{-1}(\vec{k},\vec{p}')\left(\frac{\Delta_{\vec{k}/2+\vec{p}'}}{E_{\vec{k}/2+\vec{p}'}}[f(E_{\vec{k}/2+\vec{p}'})-\tfrac{1}{2}]+\frac{\Delta_{\vec{k}/2-\vec{p}'}}{E_{\vec{k}/2-\vec{p}'}}[f(E_{\vec{k}/2-\vec{p}'})-\tfrac{1}{2}]\right) . \tag{3.13}$$

For the normal phase ($\Delta_{\vec{k}}=0$) it can be seen that $\kappa(0)=0$. The existence of a Meissner effect is assimilated with a nonvanishing $\kappa(0)$, actually

$$\kappa(0)=-c/\Lambda^2 , \tag{3.14}$$

where c is the velocity of light and Λ is the so-called London length. Taking the proper zero wave-vector limit in Eq. (3.11), we immediately have

$$\lim_{\epsilon\to 0}K_{\mu\nu}(\epsilon\vec{k})=-\frac{c}{\Lambda^2}\delta_{\mu\nu}+\frac{2e^2\hbar^2}{mc}\frac{1}{(2\pi)^3}\lim_{\epsilon\to 0}\int d\vec{p}\int d\vec{p}'p_\mu a(\epsilon\vec{k},\vec{p})M^{-1}(\epsilon\vec{k})_{\vec{p}\vec{p}'}a(\epsilon\vec{k},\vec{p}')p'_\nu , \tag{3.15}$$

where

$$\Lambda^{-2}=\frac{e^2}{mc}\left[\frac{\langle N\rangle}{\Omega}+\frac{4}{3}\frac{1}{(2\pi)^3}\int d\vec{p}(\epsilon_{\vec{p}}+\mu)\frac{\partial f(E_{\vec{p}})}{\partial E_{\vec{p}}}\right] . \tag{3.16}$$

The difficulty lies in the calculation of the second term in Eq. (3.15) (coming from η due to the field-induced variation of the order parameter). It is generally admitted without proof that its contribution is purely longitudinal; therefore, Eq. (3.14) results with Λ given by Eq. (3.16). Our aim is to analyze exactly under which conditions this is indeed true.

IV. STABILITY AND THE MEISSNER EFFECT

Let us now examine the properties of the matrix $M(\vec{k})$, which is the central piece of our problem. From its definition, Eq. (3.8), it is obvious that it is Hermitian, real, and an even function of $\vec{k}$,

$$M^{\dagger}(\vec{k})=M(\vec{k})=M(-\vec{k}) . \tag{4.1}$$

If the potential $\hat{V}(\vec{k})$ is a rapidly decreasing continuous function with $|\vec{k}|\to\infty$, it can be shown that $\mathrm{Tr}M(\vec{k})^2<\infty$ [i.e., $M(\vec{k})$ is a Hilbert-Schmidt operator]. Such an operator has only a discrete spectrum.[3]

It can also be seen immediately that $M(\vec{k})$ has a zero eigenvalue for $\vec{k}=0$,

$$\int d\vec{p}'[M(0)]_{\vec{p}\vec{p}'}\Delta_{\vec{p}'}=0 , \tag{4.2}$$

as a consequence of Eq. (2.9). Since $\Delta_{\vec{p}}$ is an eigenfunction, it is square integrable, and this assures the convergence of the grand-canonical potential, Eq. (2.14).

If we examine the local stability of the grand-canonical potential, Eq. (2.12), around the assumed solution of the self-consistency equation (which assures only its stationarity), we are lead to consider its second derivatives with respect to $\Delta_{\vec{k}}$ and $\Delta^{*}_{\vec{k}}$. For our purposes it is sufficient to look at variations with respect to the phase $\varphi_{\vec{k}}$ of $\Delta_{\vec{k}}$

($\Delta_{\vec{k}} = |\Delta_{\vec{k}}| e^{i\varphi_{\vec{k}}}$). A simple calculation shows that

$$\int d\vec{p} \int d\vec{p}\,' \frac{\delta^2 \mathcal{F}^{\Delta}}{\delta\varphi_{\vec{p}}\,\delta\varphi_{\vec{p}\,'}}\Bigg|_{\text{stat pt}} \delta\varphi_{\vec{p}}\,\delta\varphi_{\vec{p}\,'}$$

$$=(2\pi)^3 \int d\vec{p} \int d\vec{p}\,'(\delta\varphi_{\vec{p}}\Delta_{\vec{p}})^*[M(0)]_{\vec{p}\,\vec{p}\,'}(\delta\varphi_{\vec{p}\,'}\Delta_{\vec{p}\,'}) \tag{4.3}$$

(where stat pt denotes stationary point). Local stability (local minimum) requires non-negativity of this entity; therefore,

$$M(0) \geq 0 \ . \tag{4.4}$$

One cannot avoid the vanishing eigenvalue corresponding to variations with a constant phase [giving rise to Eq. (4.2)], since the wave functions are determined only up to a constant phase.

In principle the zero eigenvalue may be degenerate. Then, the minimum condition should be checked on higher variations along the directions given by the corresponding eigenfunctions. In this section we shall admit that there is no such degeneracy.

Had we studied variations that are not translational invariant we would have obtained a similar condition for arbitrary $\vec{k}$. This thermodynamical stability corresponds also to the stability of $\nu(\vec{k},\vec{p})$ (which is proportional to the field-induced phase of the order parameter) with respect to small magnetic fields. Indeed, Eq. (3.7) shows that a small vector potential induces a small ν (through linear-response theory) only if $M(\vec{k})$ has no vanishing eigenvalues (except the one that induces an arbitrary constant phase).

For the forthcoming discussion of the Meissner effect we shall assume that such a strict local stability holds, i.e.,

$$M(\vec{k}) > 0 \ , \tag{4.5}$$

for $\vec{k} \neq 0$, and there is only one nondegenerate, isolated zero eigenvalue for $\vec{k}=0$ corresponding to Eq. (4.2).

Now we may return to performing the $\epsilon \to 0$ limit in Eq. (3.15). From the definitions, Eqs. (3.5), (3.8), and (3.9), it follows that $a(\epsilon\vec{k},\vec{p})$ and $M(\epsilon\vec{k})$ can be developed in a power series in ϵ, containing only odd and even powers, respectively, as follows:

$$a(\epsilon\vec{k},\vec{p}) = \epsilon a^{(1)}(\vec{k},\vec{p}) + \cdots \ , \tag{4.6}$$

$$M(\epsilon\vec{k}) = M(0) + \epsilon^2 M^{(2)}(\vec{k}) + \cdots \ . \tag{4.7}$$

Therefore, the finite contribution of

$$I_{\mu\nu} = \lim_{\epsilon\to 0} \int d\vec{p} \int d\vec{p}\,' p_\mu \times a(\epsilon\vec{k},\vec{p})[M^{-1}(\epsilon\vec{k})]_{\vec{p}\,\vec{p}\,'} a(\epsilon\vec{k},\vec{p}\,')p'_\nu \tag{4.8}$$

to Eq. (3.15) results only because of the vanishing eigenvalue of $M(0)$. Using bra and ket notation, one may therefore write

$$I_{\mu\nu} = \lim_{\epsilon\to 0} \left\langle p_\mu a(\epsilon\vec{k}) \left| \frac{1}{m(\epsilon\vec{k})} P(\epsilon\vec{k}) \right| p_\nu a(\epsilon\vec{k}) \right\rangle \ , \tag{4.9}$$

where $m(\epsilon\vec{k})$ is that eigenvalue of $M(\epsilon\vec{k})$ which goes to zero as $\epsilon \to 0$, and $P(\epsilon\vec{k})$ is the corresponding eigenprojector,

$$\lim_{\epsilon\to 0} m(\epsilon\vec{k}) = 0, \quad \lim_{\epsilon\to 0} P(\epsilon\vec{k}) = \frac{|\Delta\rangle\langle\Delta|}{\langle\Delta|\Delta\rangle} \ . \tag{4.10}$$

First-order nondegenerate perturbation theory (allowed for an isolated eigenvalue) applied to Eq. (4.7) [where the perturbation is $\epsilon^2 M^{(2)}(\vec{k})$] gives

$$m(\epsilon\vec{k}) = \epsilon^2 \frac{\langle\Delta|M^{(2)}(\vec{k})|\Delta\rangle}{\langle\Delta|\Delta\rangle} \ . \tag{4.11}$$

Therefore, the limit can be easily performed, giving

$$I_{\mu\nu} = \frac{\langle p_\mu a^{(1)}(\vec{k})|\Delta\rangle\langle\Delta|p_\nu a^{(1)}(\vec{k})\rangle}{\langle\Delta|M^{(2)}(\vec{k})|\Delta\rangle} \ . \tag{4.12}$$

On the other hand, from the ϵ expansion of the identity Eq. (3.13), one immediately obtains

$$\frac{\hbar^2}{m} \sum_\mu k_\mu \langle\Delta|p_\mu a^{(1)}(\vec{k})\rangle = -2\langle\Delta|M^{(2)}(\vec{k})|\Delta\rangle \ , \tag{4.13}$$

or using the rotational invariance ($\Delta_{\vec{p}}$ being a function of $|\vec{p}|$ in the absence of degeneracy),

$$\langle\Delta|p_\mu a^{(1)}(\vec{k})\rangle = k_\mu \mathcal{C} \ , \tag{4.14}$$

with

$$(\hbar^2/m)\vec{k}^2 \mathcal{C} = -2\langle\Delta|M^{(2)}(\vec{k})|\Delta\rangle \ . \tag{4.15}$$

Thus

$$I_{\mu\nu} = -(k_\mu k_\nu/\vec{k}^2)\mathcal{C}(2m/\hbar^2) \ . \tag{4.16}$$

The constant $\mathcal{C}$ can be computed directly from the ϵ expansion of $a(\epsilon\vec{k},\vec{p})$, but since we know that $K_{\mu\nu}$ has the structure given by Eq. (3.12), the pure longitudinal structure of $I_{\mu\nu}$ also predicts its coefficient, giving

$$\lim_{\epsilon\to 0} K_{\mu\nu}(\epsilon\vec{k}) = -\left(\delta_{\mu\nu} - \frac{k_\mu k_\nu}{\vec{k}^2}\right)\frac{c}{\Lambda^2} \ . \tag{4.17}$$

We may conclude, therefore, that the stability of the pairing phase we have required leads to the Meissner effect, with the London length given by the naive theory. A by-product of our discussion is the proof of the positivity of the square of the London length ($\Lambda^2 \geq 0$) if the stability conditions are satisfied. This results from the positivity of the matrix $M(\vec{k})$ that gives rise to the positivity of the 3×3 matrix $I_{\mu\nu}$. A good illustration of the troubles one faces when this stability condition is not met is given by an exactly solvable model described in the next section.

V. EXACTLY SOLVABLE EXAMPLE WITH UNSTABLE PAIRING SOLUTION

Let us consider the case of a constant attractive potential. Its Fourier transform is

$$\hat{V}(\vec{k}) = -(2\pi)^3 |v| \delta(\vec{k}) \,. \tag{5.1}$$

The gap equation (2.9) is then just a simple transcendental equation,

$$\Delta_{\vec{k}} = |v| (\Delta_{\vec{k}}/2E_{\vec{k}})[1-2f(E_{\vec{k}})] \,. \tag{5.2}$$

It has an anomalous (pairing) solution,

$$\Delta_{\vec{k}} = \begin{cases} 0, & |\epsilon_{\vec{k}}| > E(T) \\ [E(T)^2-\epsilon_{\vec{k}}^2]^{1/2}, & |\epsilon_{\vec{k}}| < E(T) \end{cases} \tag{5.3}$$

where $E(T)$ is the solution of the equation

$$1 = |v| [1/2E(T)][1-2f(E(T))] \,, \tag{5.4}$$

or

$$\frac{E(T)}{E(0)} = \tanh\left[\frac{E(T)}{E(0)}\frac{T_c}{T}\right] , \tag{5.5}$$

with

$$E(0) = 2kT_c \,, \tag{5.6}$$

and the critical temperature T_c is given by

$$kT_c = |v|/4 \,. \tag{5.7}$$

In this model $E(T)$ is the actual gap above the Fermi level, and Eqs. (5.5) and (5.6) are very close to the similar relations of the phenomenology. In spite of this, we will show that this model does not give rise to a Meissner effect. The profound reason for this perhaps surprising result is the local instability of the solution. Indeed, along with the solution $\Delta_{\vec{k}}$ given by Eq. (5.3), $\Delta_{\vec{k}} e^{i\varphi_{\vec{k}}}$ is also a solution for arbitrary phase $\varphi_{\vec{k}}$. Therefore, the conditions we required in the preceding section are not met. $M(0)$ has an infinitely degenerate zero eigenvalue.

To avoid any confusion about the kind of instability that exists, let us remark that for the difference of the grand-canonical potentials of the anomalous and normal phases, one has

$$\begin{aligned} \frac{1}{\Omega}(\mathcal{F}_s - \mathcal{F}_N) = \frac{1}{(2\pi)^3} \int d\vec{k}\, \Theta(E(T)-|\epsilon_{\vec{k}}|) \\ \times \left[|\epsilon_{\vec{k}}| - E(T) + \frac{E(T)^2-\epsilon_{\vec{k}}^2}{2E(0)} \right. \\ \left. + \frac{2}{\beta}\ln\frac{1-f(E(T))}{1-f(\epsilon_{\vec{k}})} \right] . \end{aligned} \tag{5.8}$$

Below the critical temperature this is always negative; therefore, the anomalous solution is preferred. The instability refers to the variations of the phase of the order parameter. Actually we have an infinity of anomalous solutions that are unstable against perturbation.

The linear response of this model to a magnetic field is facilitated by the fact that here the matrix $M(\vec{k})$ is diagonal

$$\begin{aligned} M(\vec{k})_{\vec{p}\vec{p}'} &= \delta(\vec{p}-\vec{p}')d(\vec{k},\vec{p})[1-|v|d(\vec{k},\vec{p})] \\ &= \delta(\vec{p}-\vec{p}')M(\vec{k},\vec{p}) \,, \end{aligned} \tag{5.9}$$

allowing an explicit form for $K_{\mu\nu}(\vec{k})$ for any $\vec{k}$.

We give here, however, only the discussion for $\vec{k}\to 0$. Now we have, instead of Eq. (4.8),

$$I_{\mu\nu} = \lim_{\epsilon\to 0} \int d\vec{p}\, p_\mu p_\nu a(\epsilon\vec{k},\vec{p})^2 [M(\epsilon\vec{k},\vec{p})]^{-1} \,, \tag{5.10}$$

or, with the help of the identity Equation (3.13) that now takes the form

$$\begin{aligned} \frac{\hbar^2}{m}\vec{p}\vec{k}a(\vec{k},\vec{p}) = M(\vec{k},\vec{p})d^{-1}(\vec{k},\vec{p}) \\ \times \left[\frac{\Delta_{\vec{k}/2+\vec{p}}}{E_{\vec{k}/2+\vec{p}}}[f(E_{\vec{k}/2+\vec{p}})-\tfrac{1}{2}] \right. \\ \left. + \frac{\Delta_{\vec{k}/2-\vec{p}}}{E_{\vec{k}/2-\vec{p}}}[f(E_{\vec{k}/2-\vec{p}})-\tfrac{1}{2}] \right] , \end{aligned}$$

one may eliminate M^{-1} and perform the limit

$$I_{\mu\nu} = -\frac{2m}{\hbar^2} \int d\vec{p}\, p_\mu p_\nu \frac{\Delta\vec{p}}{\vec{p}\vec{k}} a^{(1)}(\vec{k},\vec{p}) \,. \tag{5.11}$$

Using, again, the rotational invariance (that again holds)

$$a^{(1)}(\vec{k},\vec{p}) = C(\vec{p}^{\,2})\vec{p}\cdot\vec{k} \,,$$

we get

$$\begin{aligned} I_{\mu\nu} &= -\frac{2m}{\hbar^2} \int d\vec{p}\, p_\mu p_\nu \Delta_{\vec{p}} C(\vec{p}^{\,2}) \\ &= -\frac{2m}{3\hbar^2}\delta_{\mu\nu} \int d\vec{p}\, \vec{p}^{\,2}\Delta_{\vec{p}} C(\vec{p}^{\,2}) \,, \end{aligned}$$

or denoting

$$\tfrac{1}{3}\int d\vec{p}\, \vec{p}^{\,2}\Delta_{\vec{p}} C(\vec{p}) = \mathscr{C}$$

similar to Eq. (4.14),

$$I_{\mu\nu} = -\delta_{\mu\nu}\mathscr{C}(2m/\hbar^2) \tag{5.12}$$

The constant $\mathscr{C}$ is again completely determined by the gauge-invariance requirement and expressible through the "London length" of this model. However, now the structure of $I_{\mu\nu}$ being $\delta_{\mu\nu}$ and not $k_\mu k_\nu/\vec{k}^2$, the result is identically zero,

$$\lim_{\epsilon\to 0} K_{\mu\nu}(\epsilon\vec{k}) = 0 \,. \tag{5.13}$$

Therefore, there is no Meissner effect in this case.

VI. DISCUSSIONS AND CONCLUSIONS

In this paper we have performed a gauge-invariant analysis of the Meissner effect in SC pairing theories with potentials depending only on the distance between the electrons. We have shown that if the second variation of the grand-canonical potential around the pairing solution is strictly positive (with the exception of a single variation that corresponds to a change with a constant phase factor), implying that it is stable against small external perturbations, then the field-induced variation of the order parameter does not change the textbook expression of the London length. If, however, the phase is unstable, the expression is modified. In the constant-potential model discussed in Sec. V, this modification is so important that it actually cancels out the Meissner effect. It is interesting to note that this example actually has only a third-order phase transition. This is due to the fact that in this model, near the critical temperature, not only the maximum value of the order parameter decreases to zero, but simultaneously the range where it is nonvanishing diminishes: $\Delta_{\vec{k}}=[E(T)^2-\epsilon_{\vec{k}}^2]^{1/2}\Theta(E(T)-|\epsilon_{\vec{k}}|)$ with $E(T) \sim 2\sqrt{3}kT_c(1-T/T_c)^{1/2}$.

All the above results refer to the linear-response kernel $\kappa(\vec{k}^{\,2})$ at $\vec{k}=\vec{0}$. It can be seen that if, for example, one would like to compute $\partial\kappa(\vec{k}^{\,2})/\partial\vec{k}^{\,2}|_{\vec{k}=\vec{0}}$ that determines the so called "correlation length," then the stability arguments do not contain enough information about the inverse of the matrix $M(\vec{k})$, and therefore one cannot avoid the calculation of the contribution of the field-induced variation of the order parameter.

ACKNOWLEDGMENTS

The authors would like to thank Dr. A. Aldea and A. Mezincescu for fruitful discussions.

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Z. Phys. B – Condensed Matter 58, 161–164 (1985)

Condensed
Zeitschrift
für Physik B Matter
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On the Nyquist Noise

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Received September 18, 1984

A quantum-statistical derivation of the general frequency dependent Nyquist theorem is given, that has also a simple circuit-theoretical interpretation. Attention is brought on its application to some very low conductivity materials, where already at 100 Hz the equilibrium noise should be attenuated with a factor of 10^{-7}, with respect to its zero frequency value.

0. Introduction

In his 1928 paper, Nyquist [1] gave his famous theorem, explaining the electrical noise produced by equilibrium fluctuations, first observed experimentally by Johnson [2]. Using an equivalent electric circuit argument, in which the resistor is placed in series with a voltage source (identifyed with the square root of the voltage noise spectral density δV_ω^2) Nyquist obtained

$$(\delta V_\omega^2)^N = 4 k_B T R$$

where T is the temperature, k_B – Boltzmann's constant and R is the resistance of the sample. This is the famous thermal white noise law. Given its importance, besides the original argument, the need was felt for a direct proof, valid for arbitrary frequencies, based only on the statistical mechanics of an isolated solid in thermal equilibrium. This step was achieved first by Callen and Wellton in 1951 [3], as a peculiar example to their "fluctuation-dissipation theorem". The corresponding formula is

$$(\delta V_\omega^2)^{CW} = 4 E_T(\omega) R(\omega)$$

where $R(\omega)$ is the resistance at frequency ω and

$$E_T(\omega) = \frac{\hbar\omega}{2} \operatorname{cth} \frac{\hbar\omega}{2 k_B T}.$$

For not too high frequencies $\hbar\omega \ll k_B T$ the function $E_T(\omega)$ can be approximated by $k_B T$, while as it is usually argued [4, 5], for most conductors $R(\omega) - R(0)$ for $\omega < 10^{10}$ Hz. Therefore, in the usual frequency range of noise measurements one recovers Nyquist's white noise law.
An essential step in the Callen-Wellton proof is, that the current is assumed to be the "cause" and the voltage the "effect".
Later in 1957 Kubo [6] elaborated his linear response theory and gave his famous formula for the conductivity, which actually relates it to the current fluctuations

$$(\delta I_\omega^2)^K = 4 E_T(\omega) \frac{1}{R(\omega)}.$$

In Kubo's theory the voltage (identified with the external one) is the "cause", while the current is the "effect". His formula actually holds in the self-consistent potential approximation.
Izuyama in 1961 [7] was the first to bring the attention to the fact, that in a Coulomb system, while the external electric field is the mechanical "cause", the actual field is to be identified with the summ of the external and internal fields. Therefore, the relationship between the current fluctuations and conductivity is a non-linear one. (Otherwise stated: The

inverse of the complex dielectric constant satisfies the dispersion relations [8].)

In what follows, we will exploit this argument, but we complete the discussion with a definition of the "potential drop on the sample", to compute its fluctuation and relate it (in the thermodynamic limit) to the current fluctuations. Our result (2.11) is a generalized, frequency dependent Nyquist theorem, that can be interpreted also in the equivalent circuit language. Instead of a simple resistor, one has to consider an RC (resistor-capacitor) circuit, to represent a material with complex conductivity.

A peculiar class of very low conductivity materials is brought to attention, where a very strong damping of the equilibrium voltage noise occurs already at 100 Hz.

1. Equilibrium Current Fluctuations and the Complex Conductivity

The spectral density of the equlibrium noise of a dynamical variable X is defined in quantum statistical mechanics as [9]

$$\delta X_\omega^2 = 4\int_0^\infty dt\cos\omega t\,\langle\{X, X(t)\}\rangle_0 \tag{1.1}$$

where

$$\{A, B\} = \tfrac{1}{2}(AB+BA)$$

and

$$\langle AB(t)\rangle_0 = \mathrm{Tr}\left\{\frac{e^{-\beta H}}{Z}Ae^{\frac{iHt}{\hbar}}Be^{-i\frac{Ht}{\hbar}}\right\} \tag{1.2}$$

Using a well-known identity one may express δX_ω^2 through the quantum-mechanical correlator, that appears in linear response theory

$$\delta X_\omega^2 = 4E_T(\omega)\,\mathrm{Re}\int_0^\infty dt\,e^{-i\omega t}\int_0^{\frac{1}{k_BT}} d\lambda\langle XX(t+i\hbar\lambda)\rangle_0. \tag{1.3}$$

Let us now consider a sample of volume $\Omega = LS$, where L is its length (along the OZ axis) and S its transverse section. The current j_z is usually defined as the average z-component of the current density

$$j_z = \frac{1}{\Omega}\int d\mathbf{x}\,j_z(\mathbf{x}) = \frac{1}{SL}\int_0^L dz\int_S dx\,dy\,j_z(x,y,z) = \frac{1}{S}I \tag{1.4}$$

where I is the average flux over all cross sections. The noise of the flux is therefore

$$\delta I_\omega^2 = 4\frac{S}{L}E_T(\omega)\,\mathrm{Re}\,\varphi(\omega) \tag{1.5}$$

where

$$\varphi(\omega) = \Omega\int_0^\infty dt\,e^{-i\omega t}\int_0^{\frac{1}{k_BT}} d\lambda\langle j_z j_z(t-i\hbar\lambda)\rangle_0. \tag{1.6}$$

On the other hand, $\varphi(\omega)$ appears within linear response theory as the coefficient between the average current density and an applied homogenous external electric field of frequency ω [6].

If one does not notice the difference between the external field and the actual field in a Coulomb system (described by the complex dielectric constant $\varepsilon(\omega)$ one is tempted to identify $\varphi(\omega)$, in the thermodynamic limit, with the complex conductivity $\sigma(\omega)$. Taking into account the internal field, it can be shown [7] (see also [10]) that actually

$$\varphi(\omega) = \sigma(\omega)\frac{\varepsilon_0}{\varepsilon(\omega)} = \frac{\sigma(\omega)}{1-\frac{i}{\omega\varepsilon_0}\sigma(\omega)}. \tag{1.7}$$

(The Kubo formula gives reasonable estimates of $\sigma(\omega)$ within the self-consistent potential approximation, but then the current correlator is that of the s.c. theory. It is obvious then, that at low frequencies, this turns out to be a poor approximation for the correlator itself.)

Therefore,

$$\delta I_\omega^2 = 4\frac{S}{L}E_T(\omega)\frac{\mathrm{Re}\,\sigma(\omega)}{\left|1-\frac{i}{\omega\varepsilon_0}\sigma(\omega)\right|^2}. \tag{1.8}$$

Since it is supposed, that for low frequencies

$$\sigma(\omega)\approx\sigma_{dc}+i\omega(\varepsilon-\varepsilon_0) \tag{1.9}$$

it follows that

$$\varphi(\omega)\approx i\omega\varepsilon_0+\frac{(\varepsilon_0\omega)^2}{\sigma_{dc}}. \tag{1.10}$$

(Compare with the s.c. approximation i.e. the Kubo formula, where $\varphi(\omega)^{sc}\approx\sigma_{dc}+i\omega(\varepsilon-\varepsilon_0)$!)

Then

$$\delta I_\omega^2\approx\frac{4k_BT}{R}\left(\frac{\omega\varepsilon_0}{\sigma_{dc}}\right)^2;\qquad\left(R\equiv\frac{L}{S}\frac{1}{\sigma_{dc}}\right) \tag{1.11}$$

i.e. the flux noise (in an isolated sample) is vanishing quadratically at zero frequency.

At a first glance, the vanishing as ω^2 of the current fluctuations at $\omega=0$ seems to disagree with the Nyquist formula. However, this is not so. According to the usual circuit-theoretical arguments, our resistor being isolated, its equivalent circuit is rather closed on a capacitor and correspondingly, the current noise has to vanish quadratically. In this open circuit, however, we may compute the voltage noise, and we will show, that by virtue of (1.8), (1.9) the Nyquist theorem is indeed obeyed at zero frequency in the classical form.

2. The Voltage

The potential $V(\mathbf{x})$ at a given point $\mathbf{x}$ is determined by the charge density $\rho(\mathbf{x})$:

$$V(\mathbf{x})=\frac{1}{4\pi\varepsilon_0}\int_{\Omega} d\mathbf{x}\,\frac{\rho(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}. \tag{2.1}$$

We shall define the "voltage", or "potential drop along the sample" as the average potential difference between the end sections of our sample

$$V\equiv\frac{1}{S}\int_{S} dx\int dy\,[V(x,y,L)-V(x,y,0)] \tag{2.2}$$

or

$$V=\frac{1}{S}\int_{\Sigma} dS_z\,V(\mathbf{x}) \tag{2.3}$$

where Σ is the whole surface and dS_z is the z-component of the surface element $d\mathbf{S}$.

This is a reasonable definition, although no proof can be given, that a voltmeter actually measures this entity.

Using also the electro-neutrality of the system under consideration, one may write

$$V=\frac{1}{S}\int_{\Omega} d\mathbf{x}'\,\rho(\mathbf{x}')\int_{\Sigma} dS_z\,\frac{1}{4\pi\varepsilon_0}\left[\frac{1}{|\mathbf{x}-\mathbf{x}'|}-\frac{1}{|\mathbf{x}|}\right]. \tag{2.4}$$

The last integral represents the potential difference between the points $\mathbf{x}'$ and 0 created by the plates of a plane capacitor with uniformly distributed charge of opposite signs. In the limit $S\to\infty$, this difference tends to

$$\frac{1}{\varepsilon_0}z'.$$

Therefore, the large S behaviour of V is

$$V=\frac{1}{\varepsilon_0 S}\int_{\Omega} d\mathbf{x}\,\rho(\mathbf{x})z. \tag{2.5}$$

Using now the equation of continuity

$$\frac{\partial\rho(\mathbf{x},t)}{\partial t}+\nabla\mathbf{j}(\mathbf{x},t)=0 \tag{2.6}$$

and the vanishing of the normal current density at the surface, one has

$$\frac{\partial V(t)}{\partial t}=\frac{L}{S\varepsilon_0}I(t). \tag{2.7}$$

Correspondingly, the fluctuations of these entities are related through

$$\delta V_\omega^2=\frac{L^2}{S^2(\varepsilon_0\omega)^2}\,\delta I_\omega^2. \tag{2.8}$$

Using now (1.8) we get

$$\delta V_\omega^2=4\frac{L}{S}E_T(\omega)\frac{1}{(\omega\varepsilon_0)^2}\frac{\operatorname{Re}\sigma(\omega)}{\left|1-\frac{i}{\omega\varepsilon_0}\sigma(\omega)\right|^2}. \tag{2.9}$$

This result may also be written as

$$\delta V_\omega^2=\frac{4L}{S}\frac{E_T(\omega)}{\omega}\operatorname{Im}\frac{1}{\varepsilon(\omega)} \tag{2.10}$$

or

$$\delta V_\omega^2=2\hbar\omega\operatorname{cth}\frac{\hbar\omega}{2k_BT}\cdot R(\omega)\frac{1}{1+\left(\frac{\omega\operatorname{Re}\varepsilon(\omega)}{\operatorname{Re}\sigma(\omega)}\right)^2} \tag{2.11}$$

where

$$R(\omega)=\frac{L}{S}\frac{1}{\operatorname{Re}\sigma(\omega)}$$

is the frequency dependent resistance of the sample.

3. Conclusions and Discussions

Equation (2.1) is the general form of the Nyquist theorem. Indeed, Nyquist's equivalent circuit formula for the equilibrium voltage noise due to an impedance $Z(\omega)$ gives

$$\delta V_\omega^2=2\hbar\omega\operatorname{cth}\frac{\hbar\omega}{2kT_B}\cdot\operatorname{Re}\frac{1}{Z(\omega)}. \tag{3.1}$$

On the other hand, a solid described by a complex conductivity (whose imaginary part is given by the dielectric constant) is better represented by an equivalent circuit made up of a parallely linked resistor $R(\omega)$ and capacitor $C(\omega)$. The capacitor should be chosen so, that the time constant RC of the circuit be the same as the time constant of the material ε/σ. Therefore

$$\frac{1}{Z(\omega)}=\frac{1}{R(\omega)}+i\omega C(\omega) \tag{3.2}$$

with

$$C(\omega)=\frac{1}{R(\omega)}\cdot\frac{\operatorname{Re}\varepsilon(\omega)}{\operatorname{Re}\sigma(\omega)}=\frac{S}{L}\operatorname{Re}\varepsilon(\omega). \tag{3.3}$$

Then from (3.1) our result (2.11) follows.

For ordinary conductors $\operatorname{Re}\sigma(\omega)/\operatorname{Re}\varepsilon(\omega)>10^{15}$ Hz and therefore in the classical noise measurement range still we recover the ordinary white noise law. In the case of semiconductors however, the correction due to the dielectric constant may be important.

A special class of materials is worth of mentioning in this context, because of their peculiar behaviour.

Many very low d.c. conductivity semiconductors ($\sigma_{dc} \approx 10^{-14}\,(\mathrm{ohm \cdot cm})^{-1}$) (doped crystals at very low temperatures, glassy and amorphous solids, molecular solids at room temperatures) show up a characteristic frequency dependence of the conductivity [11–15]

$$\mathrm{Re}\,\sigma(\omega) \sim \omega^s$$

with s close to 1, in the low frequency range 10^2–10^7 Hz. This unusual increase of the conductivity is atributed to electronic conduction through hopping near the Fermi level. In the same time, the dielectric constant is practically frequency-independent, being determined by other electronic states. Then the supplementary factor

$$\frac{1}{1+\left(\frac{\omega\,\mathrm{Re}\,\varepsilon(\omega)}{\mathrm{Re}\,\sigma(\omega)}\right)^2} \tag{3.4}$$

becomes very important, but practically constant in the above mentioned frequency range, while the voltage noise will have an approximate $1/\omega$ behaviour. (Not to be confused with the usual $1/\omega$ current-noise, which is a non-equilibrium noise.)

Let us give a few numerical examples of this factor, taken from different experiences. In the n-type Si experiments of Pollak and Geballe [11] at $T=3$ K, it is 10^{-4}. In the experiments of Lakatos and Abkovitz [16] on amorphous Se at $T=300$ K it is 10^{-5}. In the experiments of Mansingh and Dhawan [17] on V_2O_5–TeO_2 glasses at $T=80$ K, it is 2×10^{-6}.

Together with the rapid diminishing of the resistance itself, we have here a dramatic drop of the equilibrium voltage noise to 10 [illegible] of its zero frequency value already at 100 Hz.

Although besides the intuitive circuit theoretical argument, we gave in this paper a proof of the general Nyquist formula (2.11) within pure statistical mechanics, we do think that experimental verification on materials of the above mentioned type is important. Intuitive arguments are not always correct, while our proof also implied several delicate points, which are debatable. The most important ones are: *a)* The Quantum-Mechanical definition of the equilibrium noise (1.1). *b)* Our definition of the "voltage". *c)* The peculiar order of the limits ($S \to \infty$ and then $L \to \infty$). *d)* The implicit assumption of configurational averages, that assure macroscopical homogeneity for disordered systems.

The authors would like to thank Dr. A. Belu for bringing the noise subject in disordered materials to our attention.

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At the beginning of Section **3** *(Conclusions and Discussions) the first paragraph and Eq. (3.1) shuld read as:*

Equation (2.11) is the general form of the Nyquist theorem. Indeed, Nyquist's equivalent circuit formula for the equilibrium voltage noise due to an impedance $Z(\omega)$ gives

$$\delta V_{\omega}^{2} = 2\hbar\omega \coth\left(\frac{\hbar\omega}{2kT_B}\right) Re Z(\omega) \tag{3.1}$$

Absorption Blue Shift in Laser-Excited Semiconductor Microspheres

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(Received 18 August 1986)

The energy of an electron-hole pair in laser-excited semiconductor microcrystallites is computed with plasma screening and dielectric polarization taken into account. A strong excitation-induced blue shift of the absorption is predicted which causes a large optical nonlinearity for crystallite sizes exceeding the bulk-exciton Bohr radius.

PACS numbers: 71.35.+z, 42.65.−k, 78.20.−e

Quantum confinement effects in semiconductor systems with reduced dimensions have attracted considerable attention within the last few years. Besides the well-known multiple-quantum-well structures which provide confinement in one dimension, quite recently also semiconductor microcrystallites were investigated which confine the laser-excited electron-hole pairs in all three space dimensions.[1-5] Such systems are readily available in the form of colloids[4] or semiconductor microcrystallite-doped glasses. The nonlinear optical properties of the glasses are presently being studied extensively[2,3,6-11] and experimental results on four-wave mixing, phase conjugation, luminescence, and femtosecond carrier dynamics have been reported.

It is now well established that the finite size of the microcrystallites causes an increase in the kinetic energy of the confined quasiparticles. Efros and Efros have shown[1] that quantitatively different confinement effects occur depending on the ratio of the crystallite radius R and the Bohr radius of the electron-hole pairs, of the holes and of the electrons, respectively. For the different regimes of crystallite sizes one has either quantization of the center of mass motion of the electron-hole pair, of the hole motion, or of the electron motion. Besides these kinetic energy effects, quite recently Brus[4] has also calculated the important modification of the electrostatic energy of an electron-hole pair due to dielectric polarization at the boundary of the crystallites.

In the present Letter, we predict strong excitation-dependent quantum-size effects for the regime in which the crystallite radius is larger than the Bohr radius a_0^B of the exciton in the corresponding bulk material. Under these conditions, without laser excitation one has only a relatively small blue shift of the exciton ground-state energy in comparison to the bulk material. The exciting light, however, leads to the creation of additional electron-hole pairs, which screen the Coulomb potential causing an increase of the exciton Bohr radius a_0 and therefore an excitation-dependent ratio R/a_0. For larger Bohr radii confinement effects become increasingly important, leading to a pronounced *excitation-induced* blue shift of the exciton absorption and, hence, to strong optical nonlinearities in the spectral regime of the semiconductor band gap.

In detail, we consider a system of semiconductor microspheres with $R > a_0^B$ embedded in a host material with dielectric constant ε_2. For semiconductors such as CdS, CdSe, or GaAs, this implies crystallite sizes around 100 Å up to several hundred angstroms, respectively, for which it is justified to assume the bulk semiconductor band structure and to apply the effective-mass approximation. In fact, it has been shown[12] that the effective-mass approximation holds even for very small crystallites, containing as little as 95 atoms. In our computations, finite-size effects enter in two ways: (i) The electrostatic energy of an electron-hole pair is modified because of polarization effects and (ii) the quantum-mechanical motion of the quasiparticles is influenced through the condition that the wave functions vanish at the crystallite boundaries.

As usual, we compute the electrostatic energy as $W=\frac{1}{2}\int d^3r\, V(\mathbf{r})\rho(\mathbf{r})$, where ρ is the charge density, which vanishes for $r > R$, and the potential V obeys Poisson's equation. The charge density inside the sphere is composed of the external charge ρ^{ext} (two point charges $+e$ at $\mathbf{r}_1$ and $-e$ at $\mathbf{r}_2$, respectively) and of the induced plasma charge ρ^{ind}, which is related to V through $\rho^{\text{ind}} = -(\varepsilon_1/4\pi)\kappa^2 V$, where the inverse screening length is given by $\kappa = [(4\pi e^2/\varepsilon_1)(\partial n/\partial\mu)]^{1/2}$. The quantity ε_1 is the background dielectric constant of the crystalline, n is the average free-carrier concentration, and μ is the quasi chemical potential. The Poisson equation for V has to be solved by use of appropriate boundary conditions for a sphere.

Since the electrostatic energy diverges for pointlike external charges (self-interaction), we compute only the variation $\delta W = W(R) - W(\infty)$. The properly subtracted bulk electrostatic energy is the screened Coulomb potential

$$-(e^2/|\mathbf{r}_1-\mathbf{r}_2|)\exp(-\kappa|\mathbf{r}_1-\mathbf{r}_2|).$$

Because of linear superposition, one may solve the problem by calculating the potential U for a single point charge,

$$V(\mathbf{r};\mathbf{r}_1,\mathbf{r}_2) = U(\mathbf{r};\mathbf{r}_1) - U(\mathbf{r};\mathbf{r}_2).$$

Expanding U in terms of Legendre polynomials, applying the conditions of total internal charge neutrality, of finiteness in the origin and vanishing at infinity, one obtains an explicit expression for U, and hence δW, in terms of Bessel functions.[13] For vanishing κ our result reduces to that of Ref. 4 (although it is not explicitly given there).

As a second step, we have to solve the quantum-mechanical two-body problem with the Hamiltonian

$$H = -\frac{(\hbar\nabla_1)^2}{2m_1} - \frac{(\hbar\nabla_2)^2}{2m_2} - \frac{e^2 e^{-\kappa|\mathbf{r}_1-\mathbf{r}_2|}}{\varepsilon_1|\mathbf{r}_1-\mathbf{r}_2|}$$
$$+\delta W(\kappa r_1, \kappa r_2, \mathbf{r}_1\cdot\mathbf{r}_2/r_1 r_2, \kappa R, \varepsilon_1/\varepsilon_2)$$

taking into account the boundary conditions for the wave functions at the surface of the sphere. In bulk semiconductors ($R\rightarrow\infty$), one can separate the center of mass and the relative motions of the electron and hole in the presence of a screened Coulomb potential. Although the Schrödinger equation cannot be solved exactly even in this relatively simple case, one has a very good approximation if one replaces the screened Coulomb potential by the Hulthén potential,[14,15] for which the s wave functions Ψ and the energy eigenvalues E are explicitly known. Details of the calculation for bulk semiconductors are given in Ref. 15. Here, we only mention that within the discussed approximations, the electron-hole ground-state energy is $E = -E_R(1-1/g)^2$, where E_R is the Rydberg energy. The exciton Bohr radius in the presence of screening is $a_0 = a_0^B g/(g-1)$ with $g = 12/(\pi^2 a_0^B \kappa)$. Without screening, $g=\infty$, and one obtains the known Coulomb results, while for $g=1$ no bound states exist.

If the radius R of the semiconductor microspheres considerably exceeds a_0^B, one expects only small size-dependent energy corrections. However, through the screening effects caused by the laser-generated electron-hole plasma of density n, the exciton radius a_0 increases and approaches R. For the situation $a_0 \simeq R$, one can approximate the pair wave function Ψ as a product of the electron and hole wave functions in a spherical potential well representing the crystallite and compute the lowest energy level of the electron-hole pair as $E_x = E_g + \langle\Psi|H|\Psi\rangle$. Here, E_g is the bulk band-gap energy in the presence of the electron-hole plasma.[15] In the regime $R \simeq a_0$, one obtains the total shift $\delta E_x = E_x(\kappa, R) - E_x(0,\infty)$ of the bound-state energy due to finite-size effects as

$$\frac{\delta E_x}{E_R} = \left(1-\frac{1}{g}\right)^2 + \left(\frac{\pi a_0^B}{R}\right)^2 + \frac{2a_0^B}{R}\bar{V}\left(\kappa R;\frac{\varepsilon_1}{\varepsilon_2}\right).$$

The first term on the right-hand side of this equation describes the reduction of the exciton binding energy in a bulk semiconductor due to screening,[15] the second term is the kinetic energy contribution due to quantum confinement,[1,4] and the third term represents the average electrostatic energy of the electron-hole pair inside the microcrystallite in the presence of plasma screening. The function $\bar{V}$ has been computed numerically and is plotted in Fig. 1 for two ratios of $\varepsilon_1/\varepsilon_2$. For $\varepsilon_1 = \varepsilon_2$ no dielectric polarization occurs at the interface between microcrystallite and bulk material and only the screening effects are present. For large κR the function $\bar{V}$ becomes independent of $\varepsilon_1/\varepsilon_2$ since for strong screening the Coulomb potential essentially decreases to zero within

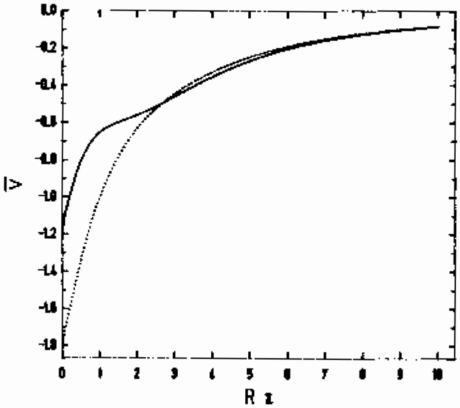

FIG. 1. Comparison of the average electrostatic energy $\bar{V}$ of an electron-hole pair in a microcrystallite (in units of $2E_R a_0^B/R$) for two ratios of the background dielectric constants of the crystallite, ε_1, and of the host material, ε_2. Solid line, $\varepsilon_1/\varepsilon_2 = 10$; dotted line, $\varepsilon_1/\varepsilon_2 = 1$. R is the radius of the microcrystallites, κ is the inverse screening length, E_R is the bulk-exciton Rydberg, and a_0^B is the bulk-exciton Bohr radius (see text).

TABLE I. Shift of the bound-state energy for different crystallite sizes. $g = 12/(\pi^2 a_0^B \kappa)$, where κ is the inverse screening length and a_0^B is the exciton Bohr radius in the unexcited bulk semiconductor.

R/a_0^B	g	$\delta E_x/E_R$
3	1.5	0.86
5	1.25	0.323
10	1.1111	0.0967

TABLE II. Shift of the bound-state energy for different crystallite sizes $R > a_0$ without screening.

R/a_0^3	$\delta E_x/E_R$
3	0.158
5	0.055
10	0.0131

the sphere, eliminating surface polarization effects. In Table I we give some typical results for different ratios of R/a_0^3 and screenings such that the excitation-dependent exciton Bohr radius a_0 equals the radius R of the semiconductor microsphere. For stronger screenings $\bar{V}$ approaches zero (see Fig. 1) and δE_x becomes practically excitation independent. To determine the excitation-dependent part of the energy shifts which give rise to nonlinear optical properties, we give in Table II the corresponding energy variations without screening ($g = \infty$, weak excitation), which have to be computed for the regime $R > a_0$ and which can already be obtained from Ref. 1 taking into account the dielectric corrections.[4] The results are presented in Table II for the example of CdS. The comparison shows that the screening induces a strong *additional* blue shift of the exciton energy, which reaches $\simeq 70\%$ of the exciton binding energy for $R = 3a_0^3$. For CdS, this large shift should therefore be observable for materials with microcrystallites of the order of $R \simeq 84$ Å, a value which is easily obtainable by use of semiconductor-doped glasses or colloids. A very promising method for the manufacture of appropriate microcrystallites also for materials such as GaAs seems to be anisotropic reactive-ion etching procedure[16] applied to multiple-quantum-well structures having the desired layer thickness.

In conclusion, we have presented a simple calculation which predicts a strong excitation-induced blue shift of the exciton resonance in semiconductor microcrystallites. The physical origin of this shift is the plasma screening which causes an increasing exciton Bohr radius and hence a plasma density-dependent increase of the confinement energy. The static equivalent of this energy shift is well established[1-4] and is obtained by a decrease of the microcrystallite radius for fixed exciton Bohr radius. Based on the same physical picture of increasing confinement effects for increasing ratio of exciton Bohr radius and crystallite radius, we predict an excitation-induced absorption blue shift which is of the order of the exciton binding energy and which leads to a decreasing absorption in the spectral region below the semiconductor band gap. This shift therefore causes an excitation-dependent absorption in these semiconductor microcrystallites and hence a significant optical nonlinearity. The magnitude of the predicted absorptive changes are equivalent to those obtained by almost complete ionization of the exciton in the case without quantum confinement. The electron-hole density required to observe the size-induced shift is only about 60% of the Mott density in the corresponding bulk material. These properties make the semiconductor microcrystallites an extremely interesting system in which to study nonlinear optical effects, such as optical bistability, nonlinear wave interaction, wave mixing, to name only a few examples, as well as for device applications, such as optical logic gates.

We thank H. M. Gibbs and E. M. Wright for a critical reading of the manuscript. One of us (S.W.K.) acknowledges financial support from the Deutsche Forschungsgemeinschaft (Bonn, Germany) and from the Optical Circuitry Cooperative of the University of Arizona.

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RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 39, NUMBER 11 15 APRIL 1989-I

Asymptotic biexciton "binding energy" in quantum dots

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(Received 24 January 1989)

The biexciton "binding energy" in a microsphere is calculated in the limit of vanishing sphere radius by use of ordinary second-order perturbation theory. This "binding energy" is found to be always positive and may exceed one excitonic Rydberg, depending on the ratio of the dielectric constants of the semiconductor and the cladding material, as well as on the electron-hole mass ratio.

Quantum confinement effects in semiconductor microcrystallites are the subject of many recent experimental and theoretical studies[1-22] Efros and Efros[1] have developed a theory of the lowest electron-hole pair state (exciton) based on the successive quantization of the center of mass, the electron, and the hole motions. Brus[2] implemented the important dielectric effects due to the difference in the dielectric constants of the semiconductor and the cladding material.

For nonlinear optics two-pair states are relevant. In recent investigations[20-22] the lowest two-pair state (biexciton) problem in a sphere of finite radius was analyzed using the phenomenological theory of Efros-Efros[1] in the so-called intermediate quantization regime. In this regime the lighter electron is frozen in its well ground state, while the heavier hole moves in the average electronic charge distribution. This picture is expected to apply whenever the electron-hole ratio is very small and the radius of the sphere lies between the Bohr radii of the electron and hole. The rather interesting result of this theory is that, unlike in bulk, quantum wells or wires, the energy of the biexciton in microcrystals may be bigger than twice the energy of the exciton. Such a negative biexciton "binding energy" does not destabilize the biexciton because in a finite sphere one cannot take the excitons far apart. Whether this theory is well justified for the experimental mass ratios, however remains an open problem. Unfortunately, variational calculations are not very helpful for the delicate energy balance because the exciton and biexciton problem have different Hamiltonians and, therefore, the well-known inequalities do not work. One has to control the errors of each approximation very accurately since the biexciton "binding energy" is only a very small fraction of the exciton energy.

An alternative method to study the confinement problem is to apply standard perturbation theory for the smallness parameter $\lambda = R/a_B$ (where R is the sphere radius and a_B the exciton Bohr radius). The peculiarity of this series expansion is that it starts with λ^{-2} (kinetic energy) followed by λ^{-1} (average of potential energy), while the first regular term in the second-order perturbation theory starts with λ^0. Within the first two approximations the biexciton energy is just twice the exciton energy. This has led to the assumption, that in the extreme quantum confinement limit the biexciton "binding energy" vanishes. However, this is not true. We will show, that second-order perturbation theory gives a radius-independent asymptotic "binding energy" which is always positive. Its numerical evaluation gives unexpectedly big values.

Let us consider the quantum-mechanical motion of electrons and holes with effective masses m_e and m_h, respectively, in an infinite potential well of radius R. The dielectric constants of the medium ϵ_1 and of the cladding ϵ_2 determine the effective Coulomb interaction between the particles (surface polarization effect). For a system of N particles of charge $q_i (i=1,\dots,N)$, the additional Coulomb energy in the sphere is[2,22]

$$\frac{1}{2}\sum_{i,j=1}^{N} q_i q_j \frac{\epsilon-1}{\epsilon_1 R}\sum_{l=0}^{\infty}\left(\frac{r_i r_j}{R^2}\right)^l P_l\left(\frac{\mathbf{r}_j\mathbf{r}}{r_j r_i}\right)\frac{1}{1+\epsilon l/(l+1)}\,, \tag{1}$$

where $\epsilon=\epsilon_1/\epsilon_2$ and $\mathbf{r}_i$ is the position of the ith particle.

For a neutral system of particles of the same absolute charge e, this potential energy can be written as a sum of positive (repulsive) and negative (attractive) two-body interaction energies $V(\mathbf{r}_i,\mathbf{r}_j)$.

We give the explicit form of the potential energy U using units appropriate for our problem. In what follows, we shall use coordinates (lengths) in units of the sphere radius R and energies in units of excitonic Rydberg energy

$$E_R=\frac{\hbar^2}{2\mu a_0^2}\left(a_0=\frac{\epsilon_1\hbar^2}{\mu e^2};\ \ \frac{1}{\mu}=\frac{1}{m_e}+\frac{1}{m_h}\right).$$

Then

$$V(\mathbf{r}_1,\mathbf{r}_2)=\frac{2}{\lambda}U(\mathbf{r}_1,\mathbf{r}_2)\,. \tag{2}$$

The partial-wave decomposition of U is given by

$$U(\mathbf{r}_1,\mathbf{r}_2)=\sum_{l=0}^{\infty}U_l(r_1,r_2,\epsilon)P_l\left(\frac{\mathbf{r}_1\mathbf{r}_2}{r_1 r_2}\right), \tag{3}$$

where

$$U_l(r_1,r_2,\epsilon)=\frac{1}{r_1}\left[\frac{r_1}{r_2}\right]^{l+1}\theta(r_2-r_1)+\frac{1}{r_2}\left[\frac{r_2}{r_1}\right]^{l+1}\theta(r_1-r_2)$$
$$+(l-\delta_{l,0})\frac{\epsilon-1}{1+\epsilon l/(l+1)}(r_1r_2)^l-\delta_{l,0}\frac{\epsilon-1}{2}\sum_{k=1}^{\infty}\frac{r_1^{2k}+r_2^{2k}}{1+\epsilon l/(k+1)}\,. \tag{4}$$

The exciton Hamiltonian is

$$H_{\text{exc}}=-\frac{1}{\lambda^2}\left[\frac{\mu}{m_e}\nabla_e^2+\frac{\mu}{m_h}\nabla_h^2\right]-\frac{2}{\lambda}U(\mathbf{r}_e,\mathbf{r}_h)\,, \tag{5}$$

while the biexciton Hamiltonian is

$$H_{\text{biexc}}=-\frac{1}{\lambda^2}\left[\frac{\mu}{m_e}(\nabla_{e_1}^2+\nabla_{e_2}^2)+\frac{\mu}{m_h}(\nabla_{h_1}^2+\nabla_{h_2}^2)\right]$$
$$-\frac{2}{\lambda}[U(\mathbf{r}_{e_1},\mathbf{r}_{h_1})+U(\mathbf{r}_{e_2},\mathbf{r}_{h_2})+U(\mathbf{r}_{e_1},\mathbf{r}_{h_2})+U(\mathbf{r}_{e_2},\mathbf{r}_{h_1})-U(\mathbf{r}_{e_1},\mathbf{r}_{e_2})-U(\mathbf{r}_{h_1},\mathbf{r}_{h_2})]\,, \tag{6}$$

The wave functions have to vanish for each particle coordinate on the sphere boundary ($r=1$).

We are interested in the biexciton spin state where the electrons and the holes are in the spin-singlet state; therefore, the exclusion principle does not apply. (This state of course will be the lowest.)

The ratio of the potential energy to the kinetic energy in both Hamiltonians is proportional to $\lambda=R/a_0$. This parameter will be considered small in what follows (quantum dots). Then one may apply standard perturbation theory with respect to λ (actually to the Hamiltonian rescaled with λ^2). The zero-order wave functions are products of the one-particle kinetic energy eigenfunctions in the sphere

$$-\nabla^2\psi(\mathbf{r})=e\psi(\mathbf{r})\text{ with }\psi(\mathbf{r})|_{r=1}=0\,. \tag{7}$$

Explicitly, they are given by

$$\psi_{l,m,n_r}=\frac{u_{l,n_r}}{r}P_l(\cos\theta)e^{im\phi}\,, \tag{8}$$

with

$$u_{l,n_r}=\text{const}\times j_l(\kappa_{l,n_r}r)\,, \tag{9}$$

where j_l are Bessel functions and κ_{l,n_r} are their zeros, i.e.,

$$j_l(\kappa_{l,n_r})=0\,. \tag{10}$$

The corresponding eigenenergies are

$$e_{l,n_r}=\kappa_{l,n_r}^2\,. \tag{11}$$

Standard perturbation theory for the ground-state eigenenergy up to second order gives

$$E=E_0+\langle 0|H'|0\rangle+\sum_a{}'\frac{|\langle 0|H'|a\rangle|^2}{E_0-E_a}\,, \tag{12}$$

where $|0\rangle$ and E_0 are the unperturbed state vector and energy E_0, H' is the perturbation, while $|a\rangle$ and E_a are the unperturbed state vectors and energies of all the other states. In our case, $|0\rangle$ will be the ground state of the kinetic energy operator for an electron-hole pair, respective for two electron-hole pairs in the sphere.

As mentioned in the Introduction, in its orthodox form perturbation theory should be applied to the rescaled Hamiltonians λ^2H_{ex} and $\lambda^2H_{\text{biexc}}$ and the correspondingly rescaled energies, then the perturbational expansion contains only positive powers of the expansion parameter λ. This is however only a formal aspect. The same result is obtained if one explicitly considers E_a of order $1/\lambda^2$ and H' of order $1/\lambda$.

The relevant matrix element in the one-pair problem is

$$\langle 0,0|H'_{\text{exc}}|\alpha,\beta\rangle=-2\frac{1}{\lambda}\int d\mathbf{x}_e\int d\mathbf{x}_h\,\psi_0(\mathbf{x}_e)\psi_0(\mathbf{x}_h)U(\mathbf{x}_e,\mathbf{x}_h)\psi_\alpha(\mathbf{x}_e)\psi_\beta(\mathbf{x}_h)=-V_{\alpha\beta}=-V_{\beta\alpha}\,, \tag{13}$$

where α (or β) stands for a given set of one-particle-state quantum numbers (l,m,n_r) in the sphere. [0 stands for the set (0,0,1) describing the one-particle ground state.]

The relevant matrix element in the two-pair problem can be expressed through the same matrix elements $V_{\alpha\beta}$:

$$\langle 00,00|H'_{\text{biexc}}|\alpha\alpha',\beta\beta'\rangle=V_{\alpha\alpha'}\delta_{\beta0}\delta_{\beta'0}+V_{\beta\beta'}\delta_{\alpha0}\delta_{\alpha'0}-V_{\alpha\beta}\delta_{\alpha'0}\delta_{\beta'0}-V_{\alpha'\beta'}\delta_{\alpha0}\delta_{\beta0}-V_{\alpha\beta'}\delta_{\alpha'0}\delta_{\beta0}-V_{\alpha'\beta}\delta_{\alpha0}\delta_{\beta'0}\,. \tag{14}$$

It can be seen immediately, that the "molecular binding energy," defined as

$$E_{\text{mol}}=2E_{\text{exc}}-E_{\text{biexc}}\,, \tag{15}$$

vanishes in zero- and first-order perturbation theory and has a finite (λ-independent), strictly positive value in second-order perturbation theory:

$$E_{\text{mol}}^{(2)}=4\sum_{\alpha,\beta>0}|U_{\alpha\beta}|^2\left[\frac{2}{(\mu/m_e)e_{\alpha0}+(\mu/m_h)e_{\beta0}}+\frac{m_e+m_h}{\mu}\frac{1}{e_{\alpha0}+e_{\beta0}}\right]\,, \tag{16}$$

where $e_{\alpha0}=e_\alpha-e_0$.

This perturbative result, unfortunately, is still difficult enough to evaluate analytically or even numerically. However, one can easily obtain lower and upper bounds for

$$\lim_{\lambda\to 0} E_{\mathrm{mol}} = E^{(2)}_{\mathrm{mol}}$$

The lowest contributing excited state to the summation in Eq. (16) is the degenerate state corresponding to $\alpha=\beta=1=(l=1, m=0, \pm 1, n_r=1)$ having the one-particle energy increment $\epsilon_{10}=\kappa_{11}^2-\kappa_{01}^2=10.3174$.

Therefore,

$$\frac{4}{\epsilon_{10}}\left[2+\frac{m_e+m_h}{2\mu}\right]\sum_{m,m'=0,\pm 1}|U_{1m1,1m'1}|^2 < \lim_{\lambda\to 0} E_{\mathrm{mol}} < \frac{4}{\epsilon_{10}}\left[2+\frac{m_e+m_h}{2\mu}\right]\sum_{\alpha,\beta>0}|U_{\alpha\beta}|^2 . \tag{17}$$

The lower bound is just the contribution of the above-mentioned state, while the higher bound comes from the replacement of all the higher-energy denominators through the lowest one.

The sum over all the excited states can be performed using the completeness of the one-particle eigenfunctions:

$$\sum_{\alpha,\beta>0}|U_{\alpha\beta}|^2=\int dx_1\int dx_2[\psi_0(x_1)\psi_0(x_2)U(x_1,x_2)]^2+\left(\int dx_1\int dx_2\psi_0(x_1)^2\psi_0(x_2)^2U(x_1,x_2)\right\}^2$$
$$-2\int dx_1\int dx_2\int dx_3[\psi_0(x_1)\psi_0(x_2)\psi_0(x_3)]^2U(x_1,x_2)U(x_2,x_3) , \tag{18}$$

where $\psi_0(x)=\sin(\pi r)/r\sqrt{2\pi}$ and the vector notation for the coordinates x was omitted.

Our result thus can be written as

$$\left[2+\frac{m_e+m_h}{2\mu}\right]C_l(\epsilon) < E_{\mathrm{mol}} < \left[2+\frac{m_e+m_h}{2\mu}\right]C_u(\epsilon) , \tag{19}$$

where the constants $C_l(\epsilon)$ and $C_u(\epsilon)$ depend only on the dielectric constant ratio ϵ. A numerical evaluation of these constants for three representative values of $\epsilon=10$, 4, and 1 gives

$$C_l(10)=0.104 \quad C_l(4)=0.076 \quad C_l(1)=0.052$$

and

$$C_u(10)=0.71 \quad C_u(4)=0.79 \quad C_u(1)=0.55 .$$

The distance between the lower and the upper bounds is still very big; however, the price to pay for a better prediction is too high. Tests of including some other peculiar states showed that the lower bound can be fairly enhanced with 50%. However, it is relevant that already this modest lower bound gives a huge asymptotic "molecular binding energy" exceeding 0.84 exciton Rydberg (for an electron-hole mass ratio of 0.1 and a dielectric ratio of $\epsilon=10$). The corresponding upper bound is 5.7 exciton Rydberg.

Our asymptotical results about the huge biexciton "binding energy" are mathematically exact. They may be helpful in the understanding of qualitative trends as well as in checking correctness of other theoretical approaches like the variational ones. It is not obvious however, that they may be really checked experimentally. The problem lies in the inherently wide inhomogeneous as well as homogeneous width of the microcrystal levels. Another source of bias is the deviation from parabolicity at high k vectors, that must be taken into account for very small sphere radii.

I would like to thank Professor H. Haug for many stimulating discussions and a critical reading of the manuscript. This work was supported by the Deutsche Forschungsgemeinschaft through the Sonderforschungsbereich 185 Frankfurt-Darmstadt.

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PHYSICAL REVIEW B VOLUME 45, NUMBER 24 15 JUNE 1992-II

Surface-polarization instabilities of electron-hole pairs in semiconductor quantum dots

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(Received 12 November 1991)

The surface polarization instabilities of a Coulomb-interacting electron-hole pair in a spherical semiconductor quantum dot inside a dielectric medium are studied. Two independent numerical solutions for the ground state are presented which are based on a direct integration of the pair Schrödinger equation or on a diagonalization of the Hamiltonian matrix. For decreasing confinement potential at fixed dot radius, and for decreasing dot radius at fixed confinement potential, it is found that the electron-hole-pair state changes from a volume state, in which both particles are mostly inside the dot, to a surface trapped state, in which the surface polarization causes the carriers to be self-trapped at the surface of the dot. The transition from volume to surface trapped states occurs for parameters which are very close to those of II-VI semiconductors in a glass matrix or in a liquid.

I. INTRODUCTION

The lowest-lying energy levels of electron-hole pairs in semiconductor quantum dots are a subject of recent theoretical investigations. The general approach of handling the attractive electron-hole Coulomb interaction was described in an early paper of Efros and Efros.[1] The influence of the surface dielectric polarization was introduced by Brus.[2] This surface polarization is especially strong for dots inside a glass matrix or in liquid solution, where the background dielectric constants of the two media are substantially different, with a typical ratio of the order of 10. On the other hand, in these systems the confinement potential barrier for the electron-hole excitations is also quite high, at least in comparison to the band offsets in epitaxially grown structures. Therefore, most numerical calculations of the last few years[3-10] have been performed within the approximation of an infinite potential well, for which the wave functions of the lowest-lying states vanish on the dot boundaries. Finite-well effects have been considered in Refs. 11 and 12, but without a simultaneous inclusion of the surface polarization.

Surface dielectric effects have been taken into account also in quantum wells[13] and quantum-well wires.[14] Recently, in the contest of quantum wells,[15] it has been shown that finite barriers lead to substantial and conceptual difficulties in the treatment of the dielectric surface polarization. The classical potential energy (self-energy) of a charged particle facing a separating surface to another medium with higher dielectric constant becomes infinite negative when approaching this surface. This singularity is not integrable and allows no normalizable ground-state solution of the Schrödinger equation. The particle "falls" onto the surface.

These unphysical results clearly show that the classical electrostatic description of the interface between dielectric media fails at distances comparable to the interatomic distance. One is forced to introduce a phenomenological cutoff distance (of the same order of magnitude as the interatomic distance) that regularizes the potential. With such a cutoff it was shown[15] for GaAs-$Ga_{1-x}Al_xAs$ quantum wells that, although the confinement potential barriers are low, the corrections due to the dielectric surface polarization are not too important due to the small difference of the dielectric constants.

In this paper we present an analysis of the interacting-electron-hole-pair ground state (exciton ground state) in a spherical quantum dot, including a finite confinement potential barrier and a cutoff dielectric self-energy. For simplicity we assume the same effective masses inside and outside the dot. In Sec. II we discuss the effective Coulomb potential energy of charged particles inside and outside a dielectric sphere. In Sec. III we then present the Schrödinger problem of an electron-hole pair inside such a sphere. In Sec. IV we show examples of our numerical solutions and in Sec. V we discuss possible consequences of the results presented.

II. EFFECTIVE COULOMB ENERGY

A charged particle in the neighborhood of a dielectric interface induces a surface polarization charge, and consequently its potential energy depends strongly on the distance to this interface. This induced surface charge acts also on other particles and therefore renormalizes the Coulomb interaction energy. The total effective Coulomb energy for two oppositely charged particles of absolute charge e in a dielectric sphere of radius R and dielectric constant ϵ_1 embedded in an infinite dielectric medium of dielectric constant ϵ_2 may be written (see the Appendix) as

$$W = \tfrac{1}{2}[V(\mathbf{r}_1/R, \mathbf{r}_2/R) + V(\mathbf{r}_2/R, \mathbf{r}_1/R)] + \Sigma(r_1/R) + \Sigma(r_2/R) , \qquad (2.1)$$

where the self-energy is given by

$$\Sigma(\rho)=E_R\frac{a_B}{R}(\epsilon-1)\sum_{l=0}^{\infty}\frac{1}{1+l(\epsilon+1)} \times\begin{cases}(l+1)\rho^{2l} & \text{for } \rho<1\\ -\epsilon l\rho^{-2(l+1)} & \text{for } \rho>1\end{cases} \tag{2.2}$$

and the interaction energy between the particles is

$$V(\rho_1,\rho_2)=-2E_R\frac{a_B}{R}\left[\frac{C(\rho_2)}{|\boldsymbol{\rho}_1-\boldsymbol{\rho}_2|} +(\epsilon-1)\sum_{l=0}^{\infty}\frac{1}{1+l(\epsilon+1)}P_l\left(\frac{\boldsymbol{\rho}_1\cdot\boldsymbol{\rho}_2}{\rho_1\rho_2}\right) \times d_l(\rho_1,\rho_2)\right] \tag{2.3}$$

with

$$C(\rho)=\begin{cases}1 & \text{for } \rho<1\\ \epsilon & \text{for } \rho>1\end{cases},$$

$$d_l(\rho_1,\rho_2)=\begin{cases}(l+1)\rho_1^l\rho_2^l & \text{for } \rho_1<1 \text{ and } \rho_2<1\\ -\epsilon l\rho_1^l\rho_2^{-l-1} & \text{for } \rho_1<1 \text{ and } \rho_2>1\\ (l+1)\rho_1^{-l-1}\rho_2^l & \text{for } \rho_1>1 \text{ and } \rho_2<1\\ -\epsilon l\rho_1^{-l-1}\rho_2^{-l-1} & \text{for } \rho_1>1 \text{ and } \rho_2>1\,.\end{cases}$$

$\epsilon\equiv\epsilon_1/\epsilon_2$ is the ratio of the dielectric constants inside and outside the dot, a_B and E_R are the exciton Bohr radius and binding energy (rydberg) of the exciton in the bulk semiconductor material of the dot.

For the case when both particles are inside the sphere, this interaction energy coincides with that discussed by Brus.[2] The other cases are obtained analogously (see the Appendix) by solving the Poisson equation with the appropriate dielectric boundary conditions.

The general term of the series defining the self-energy behaves for $l\to\infty$ like the geometrical series, and therefore the self-energy is diverging as $r/R\to 1$. Subtracting known series, one may put the self-energy into the form

$$\Sigma(\rho)=E_R\frac{a_B}{R}\frac{\epsilon-1}{\epsilon+1}\left[\frac{1}{1-\rho^2}-\frac{1}{\rho^2}\frac{\epsilon}{\epsilon+1}\ln(1-\rho^2) +\frac{\epsilon^2}{\epsilon+1}\sum_{l=0}^{\infty}\frac{\rho^{2l}}{(l+1)[1+l(\epsilon+1)]}\right]$$

for $\rho<1$ and

$$\Sigma(\rho)=E_R\frac{a_B}{R}\frac{\epsilon-1}{\epsilon+1}\epsilon\left[\frac{1}{1-\rho^2}-\frac{1}{\epsilon+1}\ln(1-\rho^{-2}) +\frac{1}{\rho^2}\frac{\epsilon}{\epsilon+1} \times\sum_{l=0}^{\infty}\frac{\rho^{-2l}}{(l+1)[1+l(\epsilon+1)]}\right]$$

for $\rho>1$.

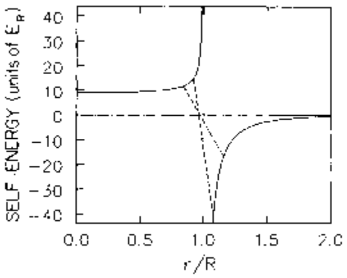

FIG. 1. Self-energy of a charged particle (in units of the Rydberg energy E_R) at the radial position ρ (in units of the sphere radius R), inside and outside the dielectric sphere having a relative dielectric constant to the surrounding $\epsilon\simeq 10$. (The dashed lines correspond to regularizations with $\delta=0.08$ and 0.16.)

After this separation of the singularities, the remaining series are converging everywhere, including the boundaries. In Fig. 1 the self-energy is represented in a radial domain ranging from the center of the sphere to twice its radius. The divergence at the surface of the sphere is not integrable and is pathological for the Schrödinger equation. The attractive potential just outside the sphere captures (traps) all particles at $r=R+0$. In what follows we shall use a regularized self-energy $\Sigma^r(\rho)$, which is finite everywhere. It coincides with $\Sigma(\rho)$ for $|\rho-1|>\delta a_B/R$ and in the interval $|\rho-1|<\delta a_B/R$ the regularized self-energy is just a linear interpolation between its values at $\rho=1\pm\delta a_B/R$, respectively. The cutoff parameter δ represents the exclusion of a layer whose thickness δa_B should be of the order of the lattice constant, a domain in which electrodynamics of continuous media breaks down. In Ref. 15 another regularization is introduced which gives a smooth interpolation. The numerical results, however, are sensitive only to the range of the interpolation δ.

The series defining the interaction energy are also diverging for $r_1\to R$ and simultaneously for $r_2\to R$, however, the divergencies are integrable and thus do not pose too severe a difficulty for the Schrödinger equation.

III. SCHRÖDINGER PROBLEM OF AN ELECTRON-HOLE PAIR IN A QUANTUM DOT

Besides their Coulomb interactions, the electron and hole in a quantum dot feel a confinement potential, chosen as a finite potential barrier of height U_0. For simplicity we take the same barrier height for both particles. We are interested to find the lowest-lying eigenenergies and wave functions of the Hamiltonian operator

$$H=-\frac{\hbar^2}{2m_e}\nabla_{\mathbf{r}_e}^2-\frac{\hbar^2}{2m_h}\nabla_{\mathbf{r}_h}^2+U_0[\Theta(r_e-R)+\Theta(r_h-R)] +W^r(\mathbf{r}_e,\mathbf{r}_h) \tag{3.1}$$

for an electron-hole pair with effective electron and hole masses m_e and m_h, respectively. The superscript r at the Coulomb energy indicates that the self-energy was replaced by the regularized one. [Actually, it is convenient to work with a continuous potential barrier which also is linearly interpolated between 0 and U_0 in the interval $R-\delta a_B<r<R+\delta a_B$ and not with the usual step function barrier $U_0\Theta(r-R)$.]

On general grounds one may expect that if the depth of the dielectric Coulomb trap just outside the dot is much bigger than the height of the confinement barrier, then the particles may be trapped in this potential minimum. The first one to be trapped would be the heavier particle, i.e., the hole in an electron-hole pair. To investigate this scenario and to find the range of physical parameters where the surface trapping occurs, we solved the Schrödinger equation with the two-particle Hamiltonian (3.1) using two different numerical methods.

Our first approach is a Hartree or self-consistent potential approximation. This means that we factorize the two-body ground-state wave function into a simple product of spherically symmetrical one-particle wave functions:

$$\Psi(\mathbf{r}_e,\mathbf{r}_h)=\psi_e(r_e)\psi_h(r_h) \ . \tag{3.2}$$

The variational principle then yields the self-consistent, coupled one-particle Schrödinger equation

$$\left[-\frac{\mu}{m_e}a_B^2E_R\nabla_{r_e}^2+U_0\Theta(r_e-R)+\Sigma^{4}(r_e/R)-U_e(r_e)-e_e\right]\psi_e(r_e)=0 \tag{3.3}$$

for the electron and analogously for the hole. The potential energy of the electron in the average field of the hole is given by

$$U_e(r_e)=\int d\mathbf{r}_h V(\mathbf{r}_h/R,\mathbf{r}_e/R)|\psi_h(r_h)|^2 \ . \tag{3.4}$$

Due to the s symmetry of the one-particle ground-state functions only the $l=0$ partial wave of the interaction potential (2.3) contributes here. The total ground-state energy E differs from the sum of the one-particle Hartree ground-state energies through the Hartree constant

$$E=e_e+e_h+\int d\mathbf{r}_e\int d\mathbf{r}_h V(\mathbf{r}_e/R,\mathbf{r}_h/R)\times|\psi_e(r_e)|^2|\psi_h(r_h)|^2 \ . \tag{3.5}$$

This system of one-dimensional Schrödinger equations was solved iteratively for the two particles using the Runge-Kutta bisection method for one-dimensional Schrödinger equations.[16] The iteration starts by solving the Schrödinger equation of the lighter particle (electron) without interaction with the heavier particle (hole). This wave function is used in the full equation of the heavier particle and so on. The convergence is achieved typically within five steps.

Another approach to solving the two-body ground-state problem is a modification of the matrix diagonalization method of Refs. 9–11, which enables us to achieve a high degree of precision. This method consists in developing the two-particle wave function in sums of products of a complete system of one-particle eigenfunctions.

$$\Psi(\mathbf{r}_e,\mathbf{r}_h)=\sum_{\nu,\mu}C_{\nu\mu}\psi_\nu^e(\mathbf{r}_e)\psi_\mu^h(\mathbf{r}_h) \ , \tag{3.6}$$

where ν and μ are running through the quantum numbers of a complete basis of one-particle kinetic-energy eigenfunctions for electrons and holes, respectively. Taking into account the spherical symmetry of the problem, the angular parts may be separated and each of the indices ν,μ summarizes the angular momentum basis indices l,m and a radial quantum number n_r:

$$\psi_{l,m,n_r}(r)=\phi_{l,m,n_r}(r)Y_l^m(\Omega) \ .$$

Taking any finite number of terms in the expansion (3.6) reduces the Schrödinger problem to a matrix diagonalization. Supercomputers permit one to achieve a high degree of convergence, successively increasing the number of components. Of course, angular momentum conservation allows an efficient reduction of the dimensionality of the matrix. A peculiar aspect of this method, when applied to the problem in this paper, is the choice of the radial part of the one-particle basis. Since the potential barrier is finite and the ground-state wave function is not to be expected to vanish everywhere outside the sphere boundary, we choose the basis of the wave functions of a single particle moving freely in a fictive sphere whose radius was chosen twice as big as that of the actual dot. This size of the fictive sphere was found to be large enough to not influence the results.

Actually the Hartree approximation may be considered as a peculiar case of the general matrix diagonalization method. Indeed, if only s-wave one-particle functions are retained and the coefficients are looked upon only in a factorized form, one recovers Eq. (3.2). Although these restrictions are not negligible, we shall see that qualitatively, and to a great extent also quantitatively, the Hartree solution gives predictions in good agreement with those of the numerical diagonalization.

IV. NUMERICAL RESULTS

We performed numerical calculations with the described methods using identical physical parameters. We took an electron-hole effective mass ratio $m_e/m_h=0.1$, a dielectric-constant ratio $\epsilon=10$, a dot radius identical with the exciton Bohr radius $R=a_B$, and a cutoff parameter $\delta=0.08$. The height of the potential barrier U_0 was varied in discrete steps in a wide domain to catch the whole scenario of surface trapping.

In Fig. 2 the radial distribution of electrons and holes for the case of an infinite potential barrier ($U_0\rightarrow\infty$) is shown both for the Hartree approximation and the matrix-diagonalization method. Even though we see some quantitative differences, both methods show that the electron and hole distributions are displaced from another, and the heavier hole is cushioned away from the surface and pushed toward the center of the sphere. For later reference we call a state with such an electron-hole distribution a "volume state" of the quantum dot.

In Fig. 3 we show within the Hartree approximation

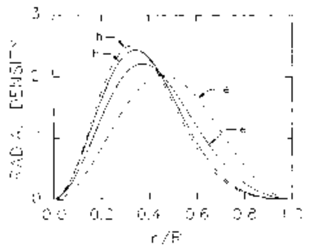

FIG. 2. Radial distribution of electrons and holes in an infinitely high potential well with $R=a_B$, $\epsilon=10$, and $m_e/m_h=0.1$ (full line, matrix diagonalization; dashed line, Hartree approximation).

the changes of the radial distributions of the electrons and holes introduced by a finite potential barrier. At $U_0=40E_R$ the barrier is still equivalent to an infinite one, but a further reduction down to $30E_R$ causes gradual changes of the hole position. The hole gradually moves to the surface of the sphere, while the electron still remains delocalized inside the sphere. This state is denoted as "surface state." A following decrease of the potential barrier brings the electron also to the surface, but still with a wider distribution than that of the hole. Qualitatively this picture survives even after the complete elimination of confinement barrier ($U_0=0$) The particles remain confined in the minimum of the potential energy given by the self-energy represented in Fig. 1.

Essentially the same scenario is described by the matrix-diagonalization calculations, but Fig. 4 shows that some quantitative differences occur. The hole radial distribution on the surface is less sharp and the electron radial distribution follows more closely that of the holes. As in the Hartree approximation, we observe a gradual transition of the electron-hole pair from volume to surface state.

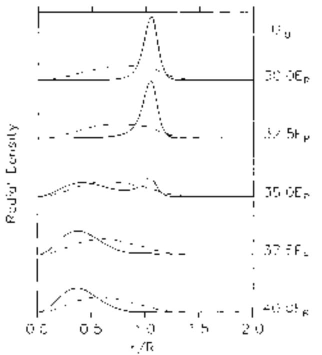

FIG. 3. Radial distributions of electrons (dashed curves) and holes (full curves) by different potential barrier heights U_0 (in units of E_R) with $R=a_B$, $\epsilon=10$, $\delta=0.08$, and $m_e/m_h=0.1$, within the Hartree approximation.

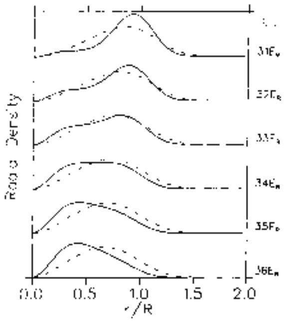

FIG. 4. Radial distributions of electrons (dashed curves) and holes (full curves) by different potential barrier heights U_0 (in units of E_R) with $R=a_B$, $\epsilon=10$, $\delta=0.08$, and $m_e/m_h=0.1$, within the matrix-diagonalization method.

In Fig. 5 we show the energies of the electron-hole pair ground state as a function of the confinement potential height. It can be seen that parallel to the localization of hole and electron on the surface, a strong drop of the energy follows, which soon becomes negative. Negative energies here mean a reduction of the electron-hole-pair ground state in the quantum dot below the band gap of the corresponding bulk semiconductor material. This energy reduction would amount to a redshifted absorption onset in the quantum dot in comparison to the continuum absorption in bulk. The polarization energy compensates and even overcompensates the confinement energy. Before localization of both particles on the surface sets in, the matrix-diagonalization results are lower than the Hartree results. For smaller confinement potential values the Hartree calculations give lower energy values.

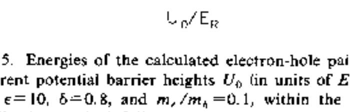

FIG. 5. Energies of the calculated electron-hole pair states for different potential barrier heights U_0 (in units of E_R) with $R=a_B$, $\epsilon=10$, $\delta=0.8$, and $m_e/m_h=0.1$, within the matrix-diagonalization method (full curve) and the Hartree approximation (dashed curve).

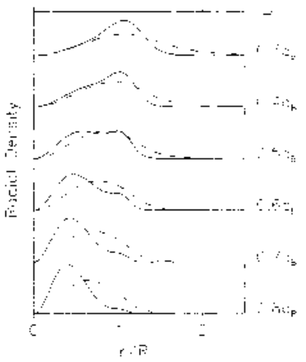

FIG. 6. Radial distributions of electrons (dashed curves) and holes (full curves) as functions of the normalized radius r/R for dots of different radii R for the same parameters $U_0=40E_R$, $\epsilon=10$, $\delta=0.08$, and $m_e/m_h=01$ within the Hartree approximation.

In principle the matrix-diagonalization energies can always be reduced to less than or equal to the Hartree calculation values simply by increasing the expansion basis. In the present case we did not attempt this numerically increasingly expensive task, since we do not believe that the domain of negative energies has physical significance.

In an experimentally realized quantum dot system it is not trivial to change the confinement potential, since this is basically determined by the chosen semiconductor and host materials. However, a reduction of the dot size at fixed confinement potential also leads to a decreasing influence of the quantum confinement and to a more pronounced penetration of the hole and electron into the host material. To investigate this experimentally relevant situation we show in Fig. 6 computed electron and hole distribution functions (within the Hartree approximation) for various dot sizes at fixed potential barrier ($U_0=40E_R$ and $\delta=0.08$). For large dot radii ($R/a_B \geq 0.8$) we see the electron-hole-pair volume state.

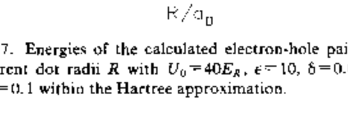

FIG. 7. Energies of the calculated electron-hole pair states for different dot radii R with $U_0=40E_R$, $\epsilon=10$, $\delta=0.08$, and $m_e/m_h=0.1$ within the Hartree approximation.

A transition to the surface state occurs around $R/a_B \simeq 0.4$. In Fig. 7 we represent the corresponding energy of the electron-hole pair as a function of the dot radius R. It is always much smaller than the ideal kinetic confinement energy $(\pi a_B/R)^2 E_R$.

V. DISCUSSION AND CONCLUSIONS

Our numerical results show that the interplay between the attractive surface polarization and the repulsive quantum confinement potential leads to a scenario where the electron-hole excitations may be confined inside the quantum dot or trapped close to the surface. For typical CdS parameters ($a_B=30$ Å and $E_R=30$ MeV) and regularization of the surface potential at a reasonable cutoff distance (about 5 Å), surface trapping of the optically generated electron-hole pair occurs at a confinement potential height of the order of $30E_R$. Alternatively, for fixed confinement potential ($40E_R$) the transition from the volume to surface state occurs around $R/a_B \simeq 0.4$.

The described scenario is a direct consequence of the use of the continuum theory of the dielectric boundary effects and the resulting divergence of the particle self-energies is mainly a difficulty of the theory itself. The simplification of using the same effective masses outside the sphere may influence quantitatively the results, however, the strong attractive potential just outside the dots remains.

Even though the actual numbers resulting from our simplified study may have to be considered with skepticism, the qualitative aspects of our theory should be relevant for the understanding of experimental observations. Here we think in terms of a scenario with a possible dielectric self-trapping in small quantum dots, mainly of the hole, close to the surface of the dot, whereas the electron is still mainly delocalized inside the dot. In this way the confinement energy roughly corresponds to the elementary kinetic confinement energy, while the charge distribution of the two particles are significantly separated and the probability for the hole to tunnel out of the dot into the surrounding material is increased.

A stronger charge separation might be an explanation for the big Huang-Rhys factors suggested by recent absorption, luminescence, and Raman scattering experiments.[17,18] The localization of the hole on the surface, on the other hand, might agree with some experimental conclusions about possible surface states of quantum dots.[19-21]

An interesting possibility emerges if one takes into account that the background dielectric constant that should be used on a short time scale is only determined by contributions from the valence electrons (ϵ_∞), while on the long time scale one should use the dielectric constant ϵ_0, which also includes the slow ionic response. Since according to our results a variation of the ratio ϵ of the dielectric constants inside (ϵ_1) and outside (ϵ_2) the quantum dot may result in a drastic change of the pair states, it is possible in principle to have an effect similar to the standard Huang-Rhys effect, in the sense of having strongly different electron-hole-pair wave functions in the early and late state of the presence of the pair in the dot.

Hence, the states into which absorption occurs and out of which the luminescence takes place could be substantially different. Since $\epsilon_0 > \epsilon_\infty$ the luminescence could occur from the surface trapped state, whereas the absorption takes place into the volume state.

In conclusion, our present work shows that surface effects, even without defects or surface irregularities, may play a very important role in the understanding of the properties of semiconductor quantum dots.

ACKNOWLEDGMENTS

One of the authors (P.G.) gratefully acknowledges partial support by the French Ministère des Affaires Etrangères during his stay in Frankfurt. The work of two of us (Y.Z.H. and S. W. Koch) has been supported by grants from NSF, ARO, and AFOSR (JSOP), the Optical Circuitry Cooperative at the University of Arizona, and the Pittsburgh Supercomputer Center. Furthermore, we acknowledge NATO which also provided partial support for our work.

APPENDIX

We give here the derivation of the classical effective Coulomb energy [Eq. (2.1)] due to the dielectric polarization effects at the surface of separation between a dielectric sphere embedded in another dielectric medium. Let us calculate first the potential created by setting a unit positive charge in an arbitrary point s inside or outside the sphere.[22] To this effect one has to solve the Poisson equation inside

$$\epsilon_1 \nabla^2 V = -4\pi\rho \quad (r < R) , \tag{A1}$$

and outside the sphere

$$\epsilon_2 \nabla^2 V = -4\pi\rho \quad (r > R) \tag{A2}$$

with the charge density

$$\rho(\mathbf{r}) = \delta(\mathbf{r}-\mathbf{s}) , \tag{A3}$$

and match the solutions at the surface of the sphere according to

$$\mathbf{r}\times\nabla V|_{r=R-0} = \mathbf{r}\times\nabla V|_{r=R+0} , \tag{A4}$$

$$\epsilon_1 \mathbf{r}\cdot\nabla V|_{r=R-0} = \epsilon_2 \mathbf{r}\cdot\nabla V|_{r=R+0} . \tag{A5}$$

The standard method is to look for the solution as a superposition of a peculiar solution of the inhomogeneous equation with the general solution of the homogeneous one. For the former we chose the bulk solution

$$V_c(\mathbf{r}) \equiv \frac{1}{|\mathbf{r}-\mathbf{s}|} \times \begin{cases} \dfrac{1}{\epsilon_1} & \text{for } s < R \\ \dfrac{1}{\epsilon_2} & \text{for } s > R . \end{cases} \tag{A6}$$

Then

$$V = V_c + \delta V , \tag{A7}$$

where δV is the general solution of the Laplace equation.

It follows from the rotational symmetry of the problem, that the potential V may depend only on the lengths s, r of the two vectors $\mathbf{s}, \mathbf{r}$ and the angle θ between these two vectors. Then an expansion in Legendre polynomials is the right way to approach the problem:

$$V(\mathbf{r}) = \sum_{l=0}^{\infty} V_l(r) P_l(\cos\theta) . \tag{A8}$$

The lth partial waves then satisfy the equation

$$\frac{1}{4}\frac{\partial^2}{\partial r^2}[r\,\delta V_l(r)] - \frac{l(l+1)}{r^2}\delta V_l(r) = 0 \tag{A9}$$

and the behavior in the origin and at infinity allows only the solutions

$$\delta V_l(r) = \begin{cases} A_l r^{l+1} & \text{for } r < R \\ B_l r^{-l} & \text{for } r > R . \end{cases} \tag{A10}$$

Using also the well-known partial-wave expansion

$$\frac{1}{|\mathbf{r}-\mathbf{s}|} = \frac{1}{\epsilon_1}\sum_{l=0}^{\infty}\left[\frac{1}{r}\left[\frac{r}{s}\right]^{l+1}\Theta(s-r) + \frac{1}{s}\left[\frac{s}{r}\right]^{l+1}\Theta(r-s)\right] P_l(\cos\Theta) \tag{A11}$$

one may fix the arbitrary constants by imposing the boundary conditions Eqs. (A4) and (A5), which for the partial waves look as

$$\epsilon_1 \frac{\partial V_l}{\partial r}\bigg|_{r=R-0} = \epsilon_2 \frac{\partial V_l}{\partial r}\bigg|_{r=R+0} , \tag{A12}$$

$$V_l|_{r=R-0} = V_l|_{r=R+0} . \tag{A13}$$

The complete solution is then

$$V(\mathbf{r};\mathbf{s}) = \frac{1}{\epsilon_1}\left[\frac{1}{|\mathbf{r}-\mathbf{s}|} + \frac{\epsilon-1}{R}\sum_{l=0}^{\infty}\left[\frac{s}{R}\right]^{l}\left[\frac{r}{R}\right]^{\alpha_l} \times P_l\left[\frac{\mathbf{r}\cdot\mathbf{s}}{rs}\right]\frac{l+1}{1-l(\epsilon+1)}\right] \tag{A14}$$

for $s < R$ and

$$V(\mathbf{r};\mathbf{s}) = \frac{1}{\epsilon_2}\left[\frac{1}{|\mathbf{r}-\mathbf{s}|} - \frac{\epsilon-1}{R}\sum_{l=0}^{\infty}\left[\frac{s}{R}\right]^{-l-1}\left[\frac{r}{R}\right]^{\alpha_l} \times P_l\left[\frac{\mathbf{r}\cdot\mathbf{s}}{rs}\right]\frac{l}{1+l(\epsilon+1)}\right] \tag{A15}$$

for $s > R$. Here we did introduce the notations

$$\alpha_l = \begin{cases} l & \text{for } r < R \\ -l-1 & \text{for } r > R \end{cases} \tag{A16}$$

and

$$\epsilon = \frac{\epsilon_1}{\epsilon_2} . \tag{A17}$$

Now we discuss the electrostatic energy of a system of point charges q_i $(i=1,2,\ldots,N)$ described by the charge density

$$\rho(\mathbf{r})=\sum_{i=1}^{N} q_i \delta(\mathbf{r}-\mathbf{s}_i) . \tag{A18}$$

With a continuous potential that vanishes at infinity the electrostatic energy may be written as

$$W=\frac{1}{2}\int d\mathbf{r}\, V(\mathbf{r})\rho(\mathbf{r}) . \tag{A19}$$

On the other side, the potential V, according to the superposition principle, may be expressed as the sum of the potentials created by each of the particles

$$V(\mathbf{r})=\sum_{i=1}^{N} q_i V(\mathbf{r};\mathbf{s}_i) . \tag{A20}$$

Because of the known divergence of the self-energy of classical pointlike particles the electrostatic energy is infinite. Nevertheless, after the subtraction of the diverging contributions

$$\lim_{\mathbf{r}\to 0}\frac{q_i^2}{\epsilon_1|\mathbf{r}|} \quad \text{and} \quad \lim_{\mathbf{r}\to 0}\frac{q_i^2}{\epsilon_2|\mathbf{r}|}$$

that are identical to those in an infinite medium,[2] we may define the effective electrostatic interaction energy

$$W_{\text{eff}}=\frac{1}{2}\sum_{\substack{i,j=1\\(i\neq j)}} q_i q_j V(\mathbf{s}_i;\mathbf{s}_j)+\frac{1}{2}\sum_{i=1}^{N} q_i^2\,\delta V(\mathbf{s}_1;\mathbf{s}_i) . \tag{A21}$$

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Motion of a Classical Polaron in a dc Electric Field

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(Received 24 August 1992)

The motion of the classical polaron in a dc electric field is investigated numerically. In a limited range of parameters (field and coupling constant) a stable stationary asymptotic drift of the electron with a constant velocity is shown to exist. Outside this range of parameters the electron is asymptotically accelerated by the field, like a free charge. This model is an illustration of the dissipative behavior of a classical mechanical subsystem coupled to a mechanical system with an infinite number of degrees of freedom (here the classical LO phonon field).

PACS numbers: 71.38.+i

The model of an electron interacting with a longitudinal optical (LO) continuum phonon field through the Fröhlich coupling [1] is one of the basic quantum-mechanical models used in the description of polar solids. Many years ago Feynman [2] introduced a path integral formulation of this polaron model, in which the integration over the phonon variables is formally performed. The action functional of the polaron appearing under this path integral corresponds to a retarded self-interaction of the electron with itself. In spite of the many applications of the quantum-mechanical polaron model, its truly classical version got very little attention. About a decade ago some analytical asymptotic solutions with finite orbits [3–5] were found.

In this work the evolution of the system from an initial state at $t=0$ without polarization (no phonons) and an applied dc electric field collinear with the initial velocity of the particle is investigated. Therefore the solutions describe a one-dimensional collinear motion and do not include for vanishing dc field the finite orbit solutions of Refs. [3–5].

The main result of this paper is the analytical and numerical proof of the existence of stationary-flow asymptotic solutions in the presence of the dc electric field. This is an example of a dissipative asymptotic motion of a particle interacting with a system having infinitely many degrees of freedom (the phonon field). In recent years dissipative asymptotic results on a mathematically rigorous level have been obtained concerning the motion of a classical particle in a Rayleigh gas (see Ref. [6] for a comprehensive review). The peculiarity of the dissipative behavior described in this paper is that it is not of statistical nature, but refers to the trajectory of a single particle and describes a state with stationary flow.

Let us define briefly the model (although it may be found in many textbooks). It describes a polarization (dipole) density $\mathbf{P}(\mathbf{x})$, which in the absence of interaction $\mathbf{u}$ obeys an oscillator equation with the mass density μ and a single frequency ω_{LO}. This phonon field interacts with an electron of mass m and charge e through the Coulomb energy

$$-\frac{1}{\epsilon_\infty}\int d\mathbf{x}\int d\mathbf{y}\,\frac{\nabla\cdot\mathbf{P}(\mathbf{x},t)\rho(\mathbf{y},t)}{|\mathbf{x}-\mathbf{y}|},$$

where $\rho(\mathbf{x},t)=e\delta(\mathbf{x}-\mathbf{r}(t))$ is the charge density of the electron, $\mathbf{r}$ is the current coordinate of the electron, and ϵ_∞ is the background dielectric constant.

Unfortunately the model with Coulomb potential is mathematically ill defined due to the singularity at the origin. This is due to the idealization to an oscillator continuum contained in the above formulation. The phonons of solid-state physics are actually oscillations of a discrete lattice and the dangerous continuum idealization is usually repaired by restricting the wave vectors of the phonon (polarization) field to the first Brillouin zone, therefore assuring the conservation of the correct number of degrees of freedom (Debye trick). This is equivalent to the replacement of the Coulomb potential through a nonsingular cutoff potential $v(\mathbf{x};a)$ depending on a cutoff length a. The modification of the potential is such that

$$\lim_{a\to 0} v(\mathbf{x},a)=\frac{1}{|x|}\,.$$

Besides the interaction with the phonons we consider also an external dc electric field $\mathbf{E}$ acting on the electron.

The prototype of the classical polaron model we consider in the following is then described by the Lagrangian function

$$L=\frac{1}{2}\dot{\mathbf{r}}^2+\mathcal{E}\mathbf{r}+\int d\mathbf{x}\left\{\dot{\mathbf{u}}(\mathbf{x})^2-\mathbf{u}(\mathbf{x})^2\right.$$
$$\left.+\left(\frac{C}{4\pi}\right)^{1/2}\nabla\cdot\mathbf{u}(\mathbf{x})v(\mathbf{x}-\mathbf{r})\right\}, \quad (1)$$

where the inverse of the phonon frequency ω_{LO} was chosen as a unit of time, the cutoff length a as a unit of length, the energy unit is $m(\omega_{\mathrm{LO}}a)^2$, and the phonon field was rescaled to a dimensionless vector field $\mathbf{u}(\mathbf{x},t)$. After this rescaling it becomes obvious that the theory depends only on two dimensionless parameters, the coupling constant and the rescaled dc field,

$$C=\frac{e^2}{\epsilon^* m\omega_{\mathrm{LO}}^2a^3},\quad \frac{1}{\epsilon^*}\equiv\frac{4\pi e^2}{\mu\epsilon_\infty^2\omega_{\mathrm{LO}}^2},\quad \mathcal{E}=\frac{e}{ma\omega_{\mathrm{LO}}^2}E\,.$$

(For illustrational purposes, with a cutoff length $a=16$ Å and GaAs parameters $C\simeq3.2$.)

Since only the longitudinal part of the phonon field is coupled to the electron, it is sufficient to concentrate on these longitudinal degrees of freedom. The coupled equations of motion follow as

$$\nabla\cdot\ddot{\mathbf{u}}(\mathbf{x},t)+\nabla\cdot\mathbf{u}(\mathbf{x},t)=-\left(\frac{C}{4\pi}\right)^{1/2}\nabla^2v(\mathbf{x}-\mathbf{r}(t))\,, \quad (2)$$

$$\ddot{\mathbf{r}}(t)=\mathcal{E}-\left(\frac{C}{4\pi}\right)^{1/2}\int d\mathbf{x}\,\nabla v(\mathbf{x}-\mathbf{r}(t))\nabla\cdot\mathbf{u}(\mathbf{x},t)\,. \quad (3)$$

One may eliminate the polarization charge in favor of the electronic variable through the formal solution of Eq. (3). We shall do this by choosing a special solution of the inhomogeneous equation adequate for defining an initial value problem at $t=0$. We chose a vanishing polarization in the absence of the electron charge (introduced at $t=0$); then

$$\nabla\cdot\mathbf{u}(\mathbf{x},t)=-\left(\frac{C}{4\pi}\right)^{1/2}\int_0^t dt'\sin(t-t')\nabla^2v(\mathbf{x}-\mathbf{r}(t'))\,. \quad (4)$$

This polarization charge density induced by the electron is nonvanishing only along the path of the electron within a tube whose transverse dimension is given by the cutoff length. Introducing this result into the Newton equation of the electron one gets the closed equation for the electron (from now on to be called polaron)

$$\ddot{\mathbf{r}}(t)=\mathcal{E}+C\int_0^t dt'\sin(t-t')\frac{\partial}{\partial\mathbf{r}(t)}V(\mathbf{r}(t)-\mathbf{r}'(t))\,, \quad (5)$$

where a new potential V was introduced according to the definition

$$V(\mathbf{r})\equiv-\frac{1}{4\pi}\int d\mathbf{x}\,v(\mathbf{r}-\mathbf{x})\nabla^2v(\mathbf{x})\,. \quad (6)$$

If one chooses for $v(\mathbf{x})$ to be the Coulomb potential, one gets for $V(\mathbf{r})$ again the Coulomb potential, but otherwise the two are different. Nevertheless, if one chooses a cutoff procedure in which the "smoothed point charge" $\rho(\mathbf{x})\equiv-\frac{1}{4}\pi\nabla^2v(\mathbf{x})$ falls off sufficiently rapidly away from $\mathbf{x}=0$, the potential $V(\mathbf{r})$ will be also Coulomb-like.

One sees that it is convenient to consider the potential $V(\mathbf{r})$ as the primary quantity instead of $v(\mathbf{r})$. We chose for our calculations a simple analytical form for this potential which is regular in the origin, and Coulomb-like at large distances

$$V(\mathbf{r})\equiv(r^2+1)^{-1/2}\,. \quad (7)$$

In what follows we shall consider only collinear motions, which are the only solutions if the initial velocity of the electron is collinear with the field, and arrive at the one-dimensional equation

$$\frac{d^2x(t)}{dt^2}=-C\int_0^t dt'\sin(t-t')\frac{x(t)-x(t')}{\{[x(t)-x(t')]^2+1\}^{3/2}}+\mathcal{E}\,. \quad (8)$$

The solution is completely determined by giving the coordinate and the velocity of the polaron at $t=0$.

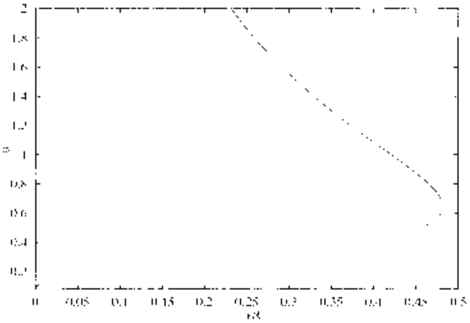

FIG. 1. Possible (asymptotical) stationary-flow velocities of the polaron u as a function of the ratio of the field $\mathcal{E}$ to the coupling constant C.

Equation (11) is a nonlinear integrodifferential equation with infinite memory and therefore finding its general solution analytically is hopeless.

Let us assume that there is a solution in the presence of the field, which "very rapidly" develops into a stationary motion with constant velocity v:

$$x(t)=ut\,.$$

A necessary condition for this velocity is

$$0=-C\int_0^\infty dt\,\sin t\,\frac{ut}{[(ut)^2+1]^{3/2}}+\mathcal{E}\,, \quad (9)$$

which leads to the transcendental equation (in terms of a Bessel function)

$$\mathcal{E}=\frac{C}{u^2}K_0\left(\frac{1}{u}\right)\,. \quad (10)$$

The dependence of the asymptotically stationary velocity u on $\mathcal{E}/C$ is represented in Fig. 1. It is clear that above a certain field ($\mathcal{E}_{\max}\simeq0.483C$), which is the upper bound of the momentum transfer rate to the phonons in a uniform motion, no asymptotically stationary-flow solutions are possible. It is worth mentioning that in order to sustain a stationary flow with a finite (unscaled) velocity au as $u\to0$, according to Eq. (8) one needs an infinite (unscaled) field E.

Of course the existence of such an asymptotically stationary solution is not yet shown, but just a necessary criterion for its existence was found. Unfortunately a standard stability analysis is not possible. First of all, we do not know the exact solution but just its asymptotically leading term. Second, any linearized version of the theory, due to the memory effects looks even more complicated than the original nonlinear equation.

The very existence and stability (against variations of the initial velocity) of the stationary drift solutions will be shown only numerically. Equation (8) has indeed a very

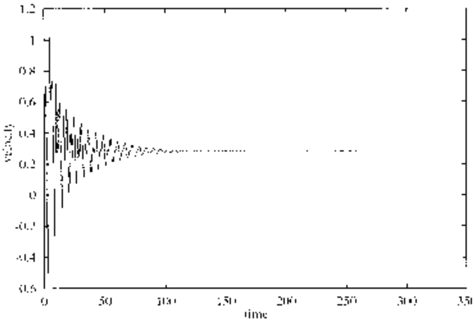

FIG. 2. Velocity of the polaron as a function of time in the presence of a field $\mathcal{E}=0.75$ at a coupling constant $C=3.2$. The initial velocity was taken to be zero.

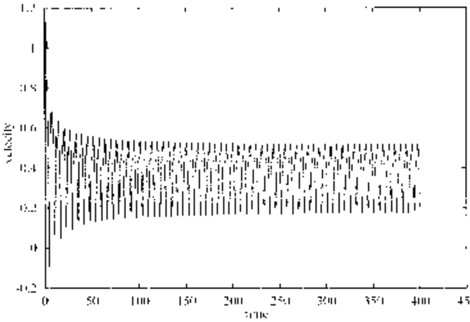

FIG. 4. Velocity of the polaron as a function of time in the presence of a field $\mathcal{E}=1.25$ at a coupling constant $C=3.2$. The initial velocity was taken to be -2.0.

simple structure, which is easy to translate into a rapidly converging discrete numerical algorithm. In what follows, numerical solutions of this equation obtained on a work station are reported.

All the solutions found for various coupling constants, fields, and initial velocities may be classified in one of two categories: (a) paths which asymptotically tend to a uniform drift, whose velocity (within some error) lies on the lower branch of the curve of Fig. 1; and (b) paths which asymptotically tend to the uniformly accelerated motion of the noninteracting electron in the external dc field.

The example given in Fig. 2 illustrates a trajectory of the first category. At a coupling constant $C=3.2$ in the presence of a field $\mathcal{E}=0.75$, after starting with an initial velocity $\dot{x}(0)=0$, one very rapidly obtains a steady motion, whose velocity corresponds to the asymptotically predicted value. Under the same parameters, but an initial velocity $\dot{x}(0)=1.5$, the trajectory suddenly changes its nature and becomes uniformly accelerated as is shown in Fig. 3. The same kind of transition to accelerated motion occurs if the initial velocity $\dot{x}(0)=0$ of the electrical field is increased to $\mathcal{E}=1.25$, although this is still smaller than the maximally allowed momentum transfer rate given for this coupling constant by $\mathcal{E}_{\max}\simeq 1.5456$. Nevertheless, an asymptotic motion with a constant drift may be again realized if the initial velocity is taken opposite to the direction of the field $\dot{x}(0)=-2.0$. Above the maximal field of 1.5456 the asymptotical motion is always uniformly accelerated.

It can be easily seen from Eq. (4) that in the asymptotically steady drift motion the induced polarization charge density closely follows the electron, and it is well approximated by a running wave with the phase velocity v and the phonon frequency along the electronic path. At the same time it can be shown that the energy of the phonon system increases linearly with the time, while the interac-

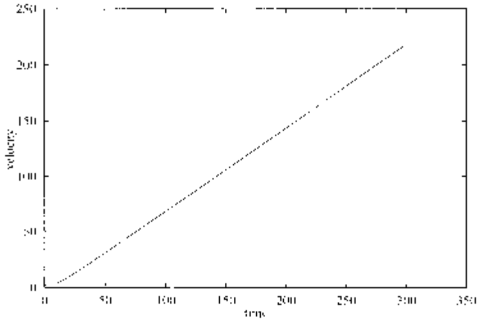

FIG. 3. Velocity of the polaron as a function of time in the presence of a field $\mathcal{E}=0.75$ at a coupling constant $C=3.2$. The initial velocity was taken to be 1.5.

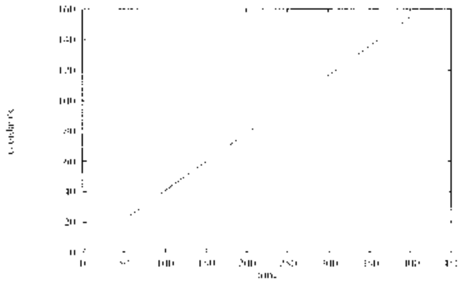

FIG. 5. Coordinate of the polaron as a function of time in the presence of a field $\mathcal{E}=1.25$ at a coupling constant $C=3.2$. The initial velocity was taken to be -2.0.

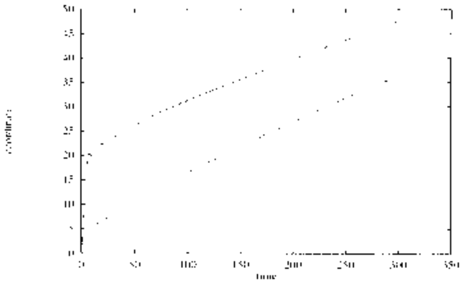

FIG. 6. Coordinate of the polaron as a function of time in the absence of a field for a coupling constant $C=3.2$. The initial velocity for the two trajectories was taken to be 2.0 and 4.0, respectively.

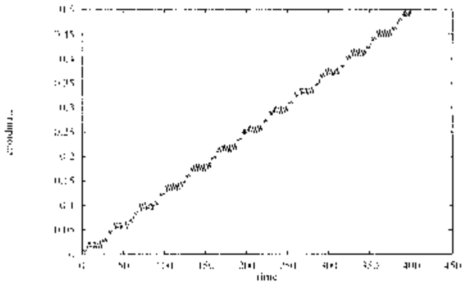

FIG. 7. Coordinate of the polaron as a function of time in the absence of a field at a coupling constant $C=3.2$, for an initial velocity of 0.01.

tion energy remains asymptotically constant. If during the initial stage of the motion, which is mainly determined by the initial velocity of the electron and the applied field (ballistic motion), the electron does not lose its contact to the polarization charge, then steady motion follows asymptotically, if in the same time the field strength does not exceed the maximal momentum transfer rate $\mathcal{E}_{\max}$.

On the contrary, in the asymptotically accelerated motion the polarization charge density decreases as $1/t$ at any finite distance behind the electron. The electron loses its polarization cloud and the interaction energy vanishes as $1/t$. This kind of asymptotic behavior follows whenever, either due to the high initial velocity or high field already in the initial (ballistic) state of the motion, the electron leaves its polarization charge far behind.

On the grounds of the discussion above it is also understandable why no drift solutions on the upper branch of the stationary curve, having high velocities, were found.

A closer inspection (blowup) of Fig. 2 actually shows small amplitude oscillations, which decay very slowly, if at all. These oscillations are well pronounced in the case of the drift motion at $C=3.2$ and $\mathcal{E}=1.25$ and $\dot{x}(0)=-2.0$ shown in Fig. 4. Nevertheless, the path of this polaron in Fig. 5 shows a clear constant average drift velocity of 0.455, slightly above the expected ideal value of 0.41. The ground frequency of the oscillations is always 1 (the phonon frequency); however, it has many higher harmonics. The deviation of the drift velocity from its ideal value might be attributed either to the fact that the true asymptotic regime was not yet achieved, or rather to the rough asymptotical analysis, which took only the leading asymptotic term into account.

The above described scenario has been checked by various coupling constant strengths, fields, and initial velocities.

Strong asymptotic oscillations are also typical for very low fields. Therefore, although in the absence of the electric field according to Fig. 1 the asymptotical drift velocity should vanish, we cannot exclude oscillating slow asymptotic drift solutions. According to the numerical experience, the motion of the electron in the absence of a dc field first suffers a rapid slowdown and afterward a very slow drift regime sets in. In Fig. 6, two trajectories are represented for $C=3.2$ having two different initial velocities [$\dot{x}(0)=2.0$ and 4.0]. The drift velocity of the slow motion is, however, not constant. One of the trajectories was followed over a long time duration ($t=1000$) and we found that the average drift coordinate increases sublinearly approximately as $t^{0.85}$. We cannot decide, however, on the basis of our numerical results, whether the motion is asymptotically very slowly damped, or a steady asymptotic drift regime with a very small velocity will be achieved. It is also relevant that for very small initial velocities the oscillatory component of the motion has a very complicated structure like that shown in Fig. 7 for $\dot{x}(0)=0.01$ and $C=3.2$ and no damping could be put into evidence.

The author would like to thank H. Haug for many useful discussions and K. El Sayed for discussions and methodical advices.

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Reprinted from ANNALS OF PHYSICS
Vol. 233, No. 2, August 1, 1994
Printed in Belgium

Time Reversal and Many-Body Non-equilibrium Green Functions

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Received June 30, 1993

A detailed discussion of the relationships following from time reversal invariance or covariance of many-body theories is given. The discussion includes equilibrium and non-equilibrium Keldysh Green functions. It is shown that self-consistent RPA approximations conserve the above deduced relationships. © 1994 Academic Press, Inc.

I. INTRODUCTION

Time reversal invariance of mechanical theories is one of the most important symmetries. Its fundamental relevance for relativistic quantum field theories has been recognized very early. Within ordinary quantum mechanics it help to understand important relationships like Onsager relations, detailed balance, etc. A detailed technical understanding of this basic symmetry is particularly important to understand the origins of irreversibility observed in nature. Most obviously time reversal symmetry may be broken for a system in contact with a bath (open system). If a system is treated in the thermodynamic limit the symmetry may also be broken, because the system obtains infinitely many degrees of freedom it may act as its own bath. But the evolution of some systems, e.g., a semiconductor excited by a laser pulse with a duration of just a few femtoseconds or a small electronic device with very high internal electric fields, cannot be described as either coherent or dissipative. The so-called quantum kinetics, developed in the last years [1–3], tries to combine these two aspects of the evoluotion within a unified formulation. In this context the time reversal properties of the many-body theories, as well as those of the usual approximation schemes, may be very useful. With all the progress of the last decades in the field of many-body theories and the abundance of good textbooks and reviews, a detailed discussion of the time-reversal symmetry in the context of non-equilibrium, to our knownledge, is still lacking.

In this paper we follow the book of Lee [4] in the formulation of time-reversal in the second quantization formalism, but we use the simplifications of the non-relativistic limit. We derive first relations following from time reversal invariance for the equilibrium Green function. Afterwards we discuss the case of general

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time-dependent Hamiltonians describing the interaction with external time-dependent electromagnetic fields. In this case we treat the non-equilibrium Green functions introduced by Keldysh [5] and speak more correctly about time reversal covariance, since also changes of the external sources are implied. We show that the derived relationships are obeyed also within the self-consistent RPA approximation both for Coulomb interacting particles and interacting particle–phonon systems.

II. TIME REVERSAL

Time reversal invariance in classical mechanics may be understood as the possibility to reverse the trajectories if the velocities of the particles are inverted at a certain moment. This simple formulation is true only in the absence of velocity-dependent forces and of external forces explicitly depending on time. In the following we restrict the discussing to interactions via electromagnetic forces which are the relevant for macroscopic systems (solid state, liquid, and gases). Time-reversal covariance then reads: *If* $\mathbf{r}(t)$, $\mathbf{E}(\mathbf{r}, t)$, $\mathbf{B}(\mathbf{r}, t)$ *are the solutions for the particle coordinates, electric and magnetic fields in the presence of the external charge current densities* $\rho_{\text{ext}}(\mathbf{r}, t)$ *and* $\mathbf{j}_{\text{ext}}(\mathbf{r}, t)$, *then* $\mathbf{r}(-t)$, $\mathbf{E}(\mathbf{r}, -t)$, $-\mathbf{B}(\mathbf{r}, -t)$ *are also solutions in the presence of the external charge and current densities* $\rho_{\text{ext}}(\mathbf{r}, -t)$ *and* $-\mathbf{j}_{\text{ext}}(\mathbf{r}, -t)$.

In quantum mechanics the formulation is a little bit more complicated [6], because the state of the system is described by a wave function. In the simplest case of a particle in a time-independent external electromagnetic field characterized by the potentials $\mathscr{V}(\mathbf{r})$, $\mathscr{A}(\mathbf{r})$, the Hamiltonian

$$\mathscr{H} = \frac{1}{2m}(-i\hbar\nabla - e\mathscr{A})^2 + e\mathscr{V} \tag{2.1}$$

has the property

$$\mathscr{H} = \mathscr{H}[\mathscr{V}, \mathscr{A}] = (\mathscr{H}[\mathscr{V}, -\mathscr{A}])^*, \tag{2.2}$$

where the square brackets indicate a functional dependence and the star indicates complex conjugation. It follows, that if $\psi(\mathbf{r}, t; [\mathscr{V}, \mathscr{A}])$ is a solution of the Schrödinger equation,

$$i\hbar\frac{\partial\psi}{\partial t} = \mathscr{H}\psi \tag{2.3}$$

with $\psi(\mathbf{r}, 0) = \psi_0(\mathbf{r})$, then $\psi'(\mathbf{r}, t) \equiv \psi^*(\mathbf{r}, -t; [\mathscr{V}, -\mathscr{A}])$ is again a solution of the same equation with $\psi'(\mathbf{r}, 0) = \psi_0^*(\mathbf{r})$.

The charge and current densities are defined as

$$\rho(\mathbf{r}, t) = e\psi^*(\mathbf{r}, t)\,\psi(\mathbf{r}, t) \tag{2.4}$$

and

$$\mathbf{j}(\mathbf{r}, t) = \frac{e}{2m} [\psi^*(\mathbf{r}, t)(-i\hbar\nabla - e\mathscr{A}(\mathbf{r}))\,\psi(\mathbf{r}, t) + \text{compl. conj.}]. \tag{2.5}$$

Therefore,

$$\rho'(\mathbf{r}, t; [\mathscr{V}, \mathscr{A}]) = \rho(\mathbf{r}, -t; [\mathscr{V}, -\mathscr{A}]) \tag{2.6}$$

and

$$\mathbf{j}'(\mathbf{r}, t; [\mathscr{V}, \mathscr{A}]) = -\mathbf{j}(\mathbf{r}, -t; [\mathscr{V}, -\mathscr{A}]), \tag{2.7}$$

where the prime indicates that the densities are calculated with the wave function ψ'.

The time-reversal invariance of the Hamiltonian (2.1) implies that if $\psi_n(\mathbf{r}; [\mathscr{V}, \mathscr{A}])$ is an eigenfunction of $\mathscr{H}$,

$$\mathscr{H}\psi_n = E_n\,\psi_n$$

with the eigenvalue E_n, then $\psi_n^*(\mathbf{r}; [\mathscr{V}, -\mathscr{A}])$ is again an eigenfunction belonging to the same eigenvalue.

In the following we shall make precise the formal aspects of the time-reversal operation in the more abstract formalism of second quantization in the Fock space, which is very important for many-body physics.

Let H be the Hamiltonian of a non-relativistic many-particle system which may consist of charged fermions and photons. For the moment we consider a closed system without external fields. Then H is *time reversal invariant* (T-invariant) [4] if there is a unitary operator U_T in the Fock space such that

$$U_T H^* U_T^+ = H. \tag{2.8}$$

Here the star means complex conjugation of any complex number. The star does not act on the creation and annihilation operators (a^+, a, b^+, b), nor on the vacuum state $|0\rangle$.

If H is T-invariant and the state vector $|\Phi(t)\rangle$ satisfies the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}|\Phi(t)\rangle = H\,|\Phi(t)\rangle, \tag{2.9}$$

then $|\Phi'(t)\rangle = U_T\,|\Phi(-t)\rangle^*$ satisfies the Schrödinger equation, too,

$$i\hbar\frac{\partial}{\partial t}|\Phi'(t)\rangle = H\,|\phi'(t)\rangle. \tag{2.10}$$

Frequently used shorthand notation for the time reversal operation on any operator $\mathcal{O}$ and any state $|\phi\rangle$ is

$$T\,\mathcal{O}\,T^{-1} \equiv U_T\,\mathcal{O}^*\,U_T^+ \tag{2.11}$$

$$T\,|\phi\rangle \equiv U_T\,|\phi\rangle^*. \tag{2.12}$$

With this definition, the T-operator is anti-unitary, i.e., unitary, but anitilinear, and has to be handled with some mathematical care.

The classical analogy suggests that quantum numbers like momentum and angular momentum should be inverted by time reversal. We define accordingly the unitary transformation U_T acting on a fermion annihilation of wavevector **k** and spin projection $\sigma_z = \sigma/2$ $(\sigma = \pm 1)$ by ?

$$U_T\,a_{\mathbf{k},\,\sigma}\,U_T^+ = \boxed{\sigma}\,a_{-\mathbf{k},\,-\sigma}, \quad ✓ \tag{2.13}$$

and on a photon annihilation operator of wavevector **k** and helicity $s = \pm 1$ by

$$U_T\,b_{\mathbf{k},\,s}\,U_T^+ = -b_{-\mathbf{k},\,-s}. \quad ✓ \tag{2.14}$$

These linear transformations conserve the equal-time commutators and therefore the existence of a unitary transformation U_T is ensured.

With this choice of the time-reversal operator, it follows that the field operator in the interaction picture,

$$\psi_\sigma(\mathbf{r}, t) = \sum_{\mathbf{k}} e^{i(\mathbf{kr} - \hbar^2 k^2/2m)t} a_{\mathbf{k},\,\sigma}, \tag{2.15}$$

transforms under time reversal as

$$T\,\psi_\sigma(\mathbf{r}, t)\,T^{-1} = \sigma\psi_{-\sigma}(\mathbf{r}, -t), \tag{2.16}$$

whereas the electromagnetic (EM) vector potential operator **A** in the coloumb gauge transforms as

$$T\,\mathbf{A}(\mathbf{r}, t)\,T^{-1} = -\mathbf{A}(\mathbf{r}, -t). \quad ✓ \tag{2.17}$$

The transformation law, Eq. (2.16), implies that the charge density, current density, and spin density operators transform as

$$T\,\rho(\mathbf{r}, t)\,T^{-1} = \rho(\mathbf{r}, t), \tag{2.18}$$

$$T\,\mathbf{j}(\mathbf{r}, t)\,T^{-1} = -\mathbf{j}(\mathbf{r}, -t), \tag{2.19}$$

and

$$T\,\mathbf{s}(\mathbf{r}, t)\,T^{-1} = -s(\mathbf{r}, -t). \tag{2.20}$$

Under these transformations indeed the Hamiltonian H describing the interaction between charged fermions with spin and photons is invariant,

$$T\,H\,T^{-1} = H. \tag{2.21}$$

The vacuum state is expected to be non-degenerate and, therefore,

$$T\,|0\rangle = \eta\,|0\rangle \qquad (|\eta| = 1). \tag{2.22}$$

if $|n\rangle$ is an eigenstate of H with the eigenvalue E_n,

$$H\,|n\rangle = E_n\,|n\rangle, \tag{2.23}$$

then $T\,|n\rangle \equiv U_T\,|n\rangle^*$ is also an eigenstate of H belonging to the same eigenvalue:

$$H\,T\,|n\rangle = E_n\,T\,|n\rangle. \tag{2.24}$$

We gave a formulation suited for electrons, ions, and photons with EM coupling, but other elementary objects, e.g., phonons, can be included easily with a suitable choice of the unitary transformation U_T acting on these objects.

III. EQUILIBRIUM PROPERTIES

Equilibrium properties of many-body system are determined by the grand canonical density matrix,

$$\rho = \frac{e^{-\beta(H-\mu N)}}{Z}, \tag{3.1}$$

where N is the total number of fermions, β is the inverse temperature, and μ is their chemical potential. In the absence of magnetic fields the T-invariance of the Hamiltonian implies for the evolution operator,

$$T\,e^{-iHt/\hbar}\,T^{-1} = e^{iHt/\hbar}, \tag{3.2}$$

and for the density matrix,

$$T\,\rho\,T^{-1} = \rho. \tag{3.3}$$

By calculating equilibrium averages

$$\langle \mathcal{O} \rangle \equiv \mathrm{Tr}\{\rho\mathcal{O}\}, \tag{3.4}$$

one is tempted to use the T-invariance property Eq. (3.3) under the trace, and thereafter to transfer the action of the unitary operator T on the operator $\mathcal{O}$, a trick one usually applies by standard invariances under a group of transformations described by a unitary transformation. In the case of an anti-unitary operator this is not possible. However, any complications can be avoided by using the properties of the eigenstates under time reversal. Indeed, in terms of the eigenstates of H Eq. (3.4) can be written as

$$\langle \mathcal{O} \rangle = \sum_n \rho_n \langle n | \, \mathcal{O} \, | n \rangle,$$

where

$$\rho_n = \frac{e^{-\beta(E_n - \mu N_n)}}{Z}.$$

The degenerated states $U_T |n\rangle$ form also a complete system of eigenstates and, therefore,

$$\langle \mathcal{O} \rangle = \sum_n \rho_n {}^* \langle n | \, U_T^+ \, \mathcal{O} \, U_T \, | n \rangle^* = \langle (U_T^+ \, \mathcal{O} \, U_T)^* \rangle^*. \tag{3.5}$$

Now for the operator $\mathcal{O}$ we take products of the field operators of the electrons,

$$\psi_\sigma(\mathbf{r}, t)^H \equiv e^{iHt/\hbar} \psi_\sigma(\mathbf{r}, 0) \, e^{-iHt/\hbar}, \tag{3.6}$$

in the Heisenberg picture. With our choice Eqs. (2.13)–(2.14) of the unitary operator U_T we obtain

$$(U_T^+ \, \psi_\sigma(\mathbf{r}, t)^H \, U_T)^* = -\sigma \psi_{-\sigma}(\mathbf{r}, -t)^H. \tag{3.7}$$

Using Eqs. (3.5) and (3.7) we find

$$\langle \psi_\sigma(\mathbf{r}, t)^H \, \psi_{\sigma'}^+(\mathbf{r}', t')^H \rangle = \sigma\sigma' \langle \psi_{-\sigma}(\mathbf{r}, -t)^H \, \psi_{-\sigma'}^+(\mathbf{r}', -t')^H \rangle^* \tag{3.8}$$

for the one-particle correlation functions as a direct consequence of the time reversal invariance of the hamiltonian H.

If only spin-symmetrical systems are considered, then

$$\begin{aligned} \langle \psi_\sigma(\mathbf{r}, t)^H \, \psi_{\sigma'}^+(\mathbf{r}', t')^H \rangle &= \delta_{\sigma, \sigma'} \langle \psi_\sigma(\mathbf{r}, t)^H \, \psi_\sigma^+(\mathbf{r}', t')^H \rangle \\ &= \langle \psi_{-\sigma}(\mathbf{r}, t)^H \, \psi_{-\sigma}^+(\mathbf{r}', t')^H \rangle. \end{aligned} \tag{3.9}$$

To simplify the notations we shall drop the spin from now on.

The time-reversal property of the correlation functions gives immediately the corresponding properties of the equilibrium Green functions. However, we postpone their discussion until the general framework of the non-equilibrium is established.

IV. TIME REVERSAL OF KELDYSH–GREEN FUNCTIONS

For the treatment of non-equilibrium problems Keldysh [5] has introduced a very elegant diagram technique, which is a natural extension of the usual Feynman diagrams of the zero temperature equilibrium theory. Even for equilibrium at finite temperatures this technique tends to replace the standard Matsubara formalism, which deals with imaginary times.

Let us assume as usual that all interactions are absent in the remote past and future, that some of them decoupled only adiabatically, and that the system was in equilibrium in the past, described by the density matrix ρ_0. The Heisenberg and interaction pictures are chosen to coincide at $t=-\infty$. The unitary evolution operator from time t_1 to the time t_2, where $t_2>t_1$, $S(t_2, t_1)$ connects the operators in the two pictures.

$$\psi(t)=S(t, -\infty)^+ \psi_0(t) S(t, -\infty). \tag{4.1}$$

With $S(t_2, t_1) S(t_2, t_1)^+=1$. For notational simplification we omit whenever possible the coordinate **r** in the arguments. This relationship may be rewritten also under the alternative forms

$$\psi(t)=S^+ T\{\psi_0(t) S\}=\tilde{T}\{S^+\psi_0(t)\} S \tag{4.2}$$

with the help of the chronological ($T\{\ \}$) and antichronological ($\tilde{T}\{\ \}$) products of operators and of the full S operator acting between $-\infty$ and ∞:

$$S \equiv S(\infty, -\infty)=T\left\{\exp\left(-\frac{i}{\hbar}\int_{-\infty}^{\infty} d\tau\, H'(\tau)\right)\right\}. \tag{4.3}$$

Here $H'(\tau)$ denotes the interaction Hamiltonian in the interaction picture.

The basic idea of Keldysh is to introduce a time contour that runs from $-\infty$ to ∞ (upper branch) and back (lower branch, see full time contour C on Fig. 1). One introduces a chronological product $T_c\{\ \}$ on this contour as well as a full S-matrix on the same contour:

$$S_c=T_c\left\{\exp\left(-\frac{i}{\hbar}\int_C d\tau\, H'(\tau)\right)\right\}. \tag{4.4}$$

A supplementary time index $\eta=\pm 1$ indicates the upper or lower branch on the contour C and correspondingly the operators carry also this time index.

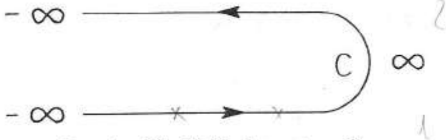

FIG. 1. The Keldysh contour C.

In close analogy to the zero-temperature theory one has a generalized Gell-Mann–Low formula connecting perturbed to unperturbed entities,

$$\langle T_c\{\psi(t,\eta)\,\psi^+(t',\eta')\}\rangle_0=\langle T_c\{\psi_0(t,\eta)\,\psi_0^+(t',\eta')\,S_c\}\rangle_0, \tag{4.5}$$

and the usual Feynman rules are valid with the unperturbed matrix propagator (causal Green functions):

$$g^{0c}_{\eta\eta'}(t,t')\equiv\frac{1}{i}\langle T_c\{\psi_0(t,\eta)\,\psi_0^+(t',\eta')\}\rangle_0. \tag{4.6}$$

(See Ref. [7–9] for a detailed description of the formalism.)

Instead of these Green functions one often uses also

$$G^c_{\eta\eta'}(t,t')\equiv g^c_{\eta\eta'}(t,t')\eta' \tag{4.7}$$

because after this redefinition some relations of the Green functions and of the higher correlators have a higher symmetry. Through a unitary transformation they might be brought to the very convenient form

$$\hat{R}\,\hat{G}\,\hat{R}^{-1}\equiv\begin{pmatrix} G_R & G_\Delta \\ 0 & G_A\end{pmatrix},\qquad \hat{R}=\frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 1 \\ -1 & 1\end{pmatrix}, \tag{4.8}$$

showing only the three linearly independent components.

We assume that the unperturbed Hamiltonian H_0 is time-reversal invariant,

$$U_T\,H_0^*\,U_T^+=H_0, \tag{4.9}$$

whereas the perturbation part H', including the external, time-dependent EM potentials $\mathscr{V}(\mathbf{r},t)$, $\mathscr{A}(\mathbf{r},t)$ transforms as

$$U_T\,H'[\mathscr{V},\mathscr{A}]^*\,U_T^+=H'[\mathscr{V},-\mathscr{A}]. \tag{4.10}$$

In the interaction picture

$$H'(t;[\mathscr{V},\mathscr{A}])=e^{iH_0t/\hbar}H'[\mathscr{V}(t),\mathscr{A}(t)]\,e^{-iH_0t/\hbar}, \tag{4.11}$$

the transformation reads

$$U_T\,H'(t;[\mathscr{V},\mathscr{A}])^*\,U_T^+=H'(-t;[\mathscr{V}_T,\mathscr{A}_T]), \tag{4.12}$$

with the transformed external potentials

$$\mathscr{V}_T(t)\equiv\mathscr{V}(-t),\qquad \mathscr{A}_T(t)\equiv-\mathscr{A}(-t). \tag{4.13}$$

From Eqs. (4.7)–(4.11) the transformation law of the S-matrix follows:

$$\begin{aligned} U_T\, S^*[\mathscr{V}, \mathscr{A}]\, U_T^+ &= T\left\{\exp\left(\frac{i}{\hbar}\int_{-\infty}^{\infty} d\tau\, H'(-\tau; [\mathscr{V}_T, \mathscr{A}_T])\right)\right\} \\ &= \tilde{T}\left\{\exp\left(\frac{i}{\hbar}\int_{-\infty}^{\infty} d\tau\, H'(\tau; [\mathscr{V}_T, \mathscr{A}_T])\right)\right\} \qquad (4.14) \\ &= S^+[\mathscr{V}_T, \mathscr{A}_T]. \end{aligned}$$

We can now apply this relationship to derive the time-reversal transformation property of the Keldysh–Green functions.

Let us now consider, for example,

$$g^c_{-+}(\mathbf{r}, t; \mathbf{r}', t') \equiv \frac{1}{i}\langle \psi(\mathbf{r}, t)\, \psi^+(\mathbf{r}', t')\rangle_0$$

which according to Eq. (4.2) may be written as

$$\frac{1}{i}\langle \tilde{T}\{S^+ \psi_0(\mathbf{r}, t)\}\; T\{\psi_0^+(\mathbf{r}', t') S\}\rangle_0.$$

Now using Eqs. (3.5) and (2.16) as in the previous section we may show that it equals also

$$\begin{aligned} &\frac{1}{i}\langle (T\{\psi_0(\mathbf{r}, -t) S\}\; \tilde{T}\{\psi_0^+(\mathbf{r}', -t')\, S^+\})^+\rangle_0\Big|_{\mathscr{V}, \mathscr{A} \to \mathscr{V}_T, \mathscr{A}_T} \\ &\quad = \frac{1}{i}\langle T\{\psi_0(\mathbf{r}', -t') S\}\; \tilde{T}\{\psi_0^+(\mathbf{r}, -t) S^+\}\rangle_0\Big|_{\mathscr{V}, \mathscr{A} \to \mathscr{V}_T, \mathscr{A}_T} \\ &\quad = \frac{1}{i}\langle S\, \psi(\mathbf{r}', -t')\, \psi^+(\mathbf{r}, -t) S^+\rangle_0\Big|_{\mathscr{V}, \mathscr{A} \to \mathscr{V}_T, \mathscr{A}_T.} \end{aligned}$$

The S matrices in the last line may be reinterpreted as performing the average on a new density matrix,

$$\tilde{\rho}_0 \equiv S^+\, \rho_0\, S, \qquad (4.15)$$

instead of ρ_0. This density matrix describes the state of the system in the remote future.

The result may be rewritten then as

$$g^c(\mathbf{r}, t; \mathbf{r}', t'; [\mathscr{V}, \mathscr{A}])_{\rho_0} = g^c(\mathbf{r}', -t'; \mathbf{r}, -t; [\mathscr{V}_T, \mathscr{A}_T])_{\tilde{\rho}_0} \qquad (4.16)$$

which is actually valid for all the components of the Keldysh–Green function matrix. This is the time-reversal covariance in terms of the one-particle Keldysh–Green functions. It is a sort of generalized Onsager relation. In the absence of time-dependent fields and no static magnetic fields, the S matrix describes the adiabatic introduction and elimination of the internal interactions in the remote past and future. Then the density matrix in the remote future equals that in the remote past $\tilde{\rho}_0 = \rho_0$; the extra notation of the functional dependence on $\mathscr{V}$ and $\mathscr{A}$ may be omitted and we obtain, for example, choosing the component $(-, +)$ in the Keldysh indices,

$$\langle \psi(\mathbf{r}, t)\, \psi^+(\mathbf{r}', t') \rangle = \langle \psi(\mathbf{r}', -t')\, \psi^+(\mathbf{r}, -t) \rangle = \langle \psi(\mathbf{r}, -t)\, \psi^+(\mathbf{r}', -t') \rangle^*;$$

i.e., we recover the relationship Eq. (3.8) that is typical for equilibrium.

Proporties analogous to Eq. (4.16) may be derived also for the many-particle Green functions. Under this form, however, this basic property is not yet very useful, because we need also recipes to calculate the averages on $\tilde{\rho}_0$. This is the next step on which we shall elaborate.

V. GREEN FUNCTIONS ON THE KELDYSH ANTI-CONTOUR

Let us now define another set of Green functions according the same rules, but on the contour $\tilde{C}$ shown in Fig. 2,

$$g^{\tilde{c}}_{\eta\eta'}(\mathbf{r}, t; r', t') \equiv \frac{1}{i} \langle T_{\tilde{c}} \{ \psi(x, t, \eta)\, \psi^+(x', t', \eta') \} \rangle_0. \tag{5.1}$$

According to the time ordering on the contour $\tilde{C}$ we have

$$i\, g^{\tilde{c}}(\mathbf{r}, t, \mathbf{r}'t') = \begin{pmatrix} \langle T\{\psi_0(\mathbf{r}, t)\, \psi_0^+(\mathbf{r}', t') S\} S^+ \rangle_0 & \langle T\{\psi_0(\mathbf{r}, t) S\} \tilde{T}\{\psi_0^+(\mathbf{r}', t') S^+\} \rangle_0 \\ -\langle \tilde{T}\{\psi_0^+(\mathbf{r}', t') S^+\} T\{\psi_0(\mathbf{r}, t) S\} \rangle_0 & \langle S \tilde{T}\{\psi_0(\mathbf{r}, t)\, \psi_0^+(\mathbf{r}', t') S^+\} \rangle_0 \end{pmatrix}. \tag{5.2}$$

This form shows that the matrix elements consists of the Green functions on the old contour C, but with the average taken for the final density matrix $\tilde{\rho}_0$ instead of the initial one ρ_0.

$$g^{\tilde{c}}|_{\rho_0} = \begin{pmatrix} g^c_{++} & -g^c_{-+} \\ -g^c_{+-} & g^c_{--} \end{pmatrix}_{\tilde{\rho}_0}. \tag{5.3}$$

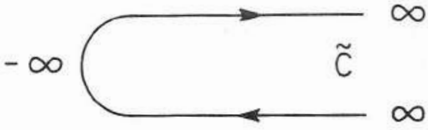

FIG. 2. The Keldysh anti-contour $\tilde{C}$.

In terms of the above G_R, G_A, and G_Δ Green functions (see Eq. (4.8)) this relationship reads

$$G^{\tilde{C}}_{R,A}|_{\rho_0} = G^{C}_{A,R}|_{\tilde{\rho}_0} \tag{5.4a}$$

$$G^{\tilde{C}}_{\Delta}|_{\rho_0} = G^{C}_{\Delta}|_{\tilde{\rho}_0}. \tag{5.4b}$$

In the non-interacting case ρ_0 equals $\tilde{\rho}_0$ and this index can be dropped. Therefore

$$G^{0\tilde{C}}_{R,A} = G^{0C}_{A,R} \tag{5.5a}$$

$$G^{0\tilde{C}}_{\Delta} = G^{0C}_{\Delta}. \tag{5.5b}$$

These last relationships Eqs. (5.5a), (5.5b) mean that the diagrammatic rules with ρ_0 on the new contour $\tilde{C}$ can be obtained from the old rules with ρ_0 on the contour C by just exchanging $G^0{}_R$ with $G^0{}_A$. On the other hand, according to Eqs. (5.4a), (5.4b) one obtains the Green functions on the contour C, but with the density matrix $\tilde{\rho}_0$, by exchanging the indices R with A of the Green functions on the contour $\tilde{C}$, but with the density matrix ρ_0. Therefore, if the one-particle Green functions with ρ_0 on the contour C are known as a given functional of the free propagators,

$$G_{R,A,\Delta}^{\;C}|_{\tilde{\rho}_0} = \mathcal{G}_{R,A,\Delta}[G^{0C}_{R}, G^{0C}_{A}, G^{0C}_{\Delta}], \tag{5.6}$$

the Green functions on the old contour C, but with the new density matrix $\tilde{\rho}_0$, which appear in the time-reversal relation, Eq. (4.16), are given by

$$G_{R,A,\Delta}^{\;C}|_{\tilde{\rho}_0} = \mathcal{G}_{A,R,\Delta}[G^{0C}_{A}, G^{0C}_{R}, G^{0C}_{\Delta}], \tag{5.7}$$

i.e., by a double change of the retarded–advanced indices.

Now we may reformulate the basic time-reversal relation, Eq. (4.16), as

$$\begin{aligned}&\mathcal{G}_{R,A,\Delta}(\mathbf{r}, t; \mathbf{r}', t'; [G^0{}_R, G^0{}_A, G^0{}_\Delta, \mathscr{V}, \mathscr{A}])\\ &\quad = \mathcal{G}_{A,R,\Delta}(\mathbf{r}', -t'; \mathbf{r}, -t; [G^0{}_A, G^0{}_R, G^0{}_\Delta, \mathscr{V}_T, \mathscr{A}_T]),\end{aligned} \tag{5.8}$$

where also the contour index has been omitted, since everything is on the standard Keldysh contour C. This is an explicit functional relationship which the Keldysh–Green functions satisfy in virtue of the time-reversal covariance of the Hamiltonian (see Eqs. (4.9)–(4.10)).

VI. TIME REVERSAL OF THE SELF-ENERGY

Most of the approximation schemes of many-body theories are formulated within the frame of the Dyson equations for the one-particle propagators. In a symbolic notations they read

$$G = G^0 + G^0\,\Sigma\,G = G^0 + G\,\Sigma\,G^0, \tag{6.1}$$

where Σ is the self-energy. The self-energy itself is related to higher Green functions which again are coupled to even higher Green functions. In that way an infinite set of coupled equations with rising complexity is formed. The standard approximations consist in choosing a simplified form for Σ which decouples the equations. We shall assume that we are working within such an approximation; i.e., the self-energy is a given functional of the Green functions G,

$$\Sigma = \Sigma[G].$$

In the case of the Keldysh formalism it is convenient to write the Dyson equations (on the contour C, with ρ_0) for the three independent Green functions G_R, G_A, and G_Δ,

$$\begin{aligned} G_R &= G^0{}_R + G^0{}_R\,\Sigma_R[G_R, G_A, G_\Delta]\,G_R \\ &= G^0{}_R + G_R\,\Sigma_R[G_R, G_A, G_\Delta]\,G^0{}_R \end{aligned} \tag{6.2a}$$

$$\begin{aligned} G_A &= G^0{}_A + G^0{}_A\,\Sigma_A[G_R, G_A, G_\Delta]\,G_A \\ &= G^0{}_A + G_A\,\Sigma_A[G_R, G_A, G_\Delta]\,G^0{}_A \end{aligned} \tag{6.2b}$$

$$\begin{aligned} G_\Delta &= G^0{}_\Delta + G^0{}_R(\Sigma_R[G_R, G_A, G_\Delta]\,G_\Delta + \Sigma_\Delta[G_R, G_A, G_\Delta]\,G_A) \\ &\quad + G^0{}_\Delta\,\Sigma_A[G_R, G_A, G_\Delta]\,G_A \\ &= G^0{}_\Delta + (G_R\,\Sigma_R[G_R, G_A, G_\Delta] + G_\Delta\,\Sigma_A[G_R, G_A, G_\Delta])\,G^0{}_A \\ &\quad + G_R\,\Sigma_R[G_R, G_A, G_\Delta]\,G^0{}_\Delta. \end{aligned} \tag{6.2c}$$

Since these equations now implicitly define the functional dependence of the Green functions G_R, G_A, and G_Δ on the unperturbed ones $G^0{}_R$, $G^0{}_A$, and $G^0{}_\Delta$, according to the given proves at the end of the previous section, we may write down immediately also the Dyson equations for the Green functions with the density matrix $\tilde{\rho}_0$,

$$\tilde{G}_{R,A,\Delta} \equiv G_{R,A,\Delta}\,|_{\tilde{\rho}_0}.$$

Following the recipe of Eqs. (5.6), (5.7) we obtain from Eqs. (6.2a)–(6.2c)

$$\begin{aligned}\tilde{G}_R &= G^0{}_R + G^0{}_R\, \Sigma_A[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, \tilde{G}_R \\ &= G^0{}_R + \tilde{G}_R\, \Sigma_A[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, G^0{}_R\end{aligned} \tag{6.3a}$$

$$\begin{aligned}\tilde{G}_A &= G^0{}_A + G^0{}_A\, \Sigma_R[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, \tilde{G}_A \\ &= G^0{}_A + \tilde{G}_A\, \Sigma_R[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, G^0{}_A\end{aligned} \tag{6.3b}$$

$$\begin{aligned}\tilde{G}_\Delta &= G^0{}_\Delta + G^0{}_A(\Sigma_R[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, \tilde{G}_\Delta + \Sigma_\Delta[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, \tilde{G}_A) \\ &\quad + G^0{}_\Delta\, \Sigma_A[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, \tilde{G}_R \\ &= G^0{}_\Delta + \{\tilde{G}_A\, \Sigma_R[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta] + \tilde{G}_\Delta\, \Sigma_A[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta])\, G^0{}_R \\ &\quad + \tilde{G}_A\, \Sigma_R[\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]\, G^0{}_\Delta.\end{aligned} \tag{6.3c}$$

The time-reversal relationship Eq. (4.16) implies, however, besides the replacement of ρ_0 through $\tilde{\rho}_0$, also the replacements of the arguments,

$$\mathbf{r}, t; \mathbf{r}', t' \Rightarrow \mathbf{r}', -t'; \mathbf{r}, -t,$$

any of the external fields

$$\mathscr{V}(\mathbf{r}, t), \mathscr{A}(\mathbf{r}, t) \Rightarrow \mathscr{V}(\mathbf{r}, -t) = \mathscr{V}_T(\mathbf{r}, t), -\mathscr{A}(\mathbf{r}, -t) = \mathscr{A}_T(\mathbf{r}, t).$$

We introduce again a simplifying notation for this operation, namely

$$G^X(\mathbf{r}, t; \mathbf{r}', t'; [\mathscr{V}, \mathscr{A}]) \equiv G(\mathbf{r}', -t'; \mathbf{r}, -t; [\mathscr{V}_T, \mathscr{A}_T]) \tag{6.4}$$

and

$$G^T \equiv \tilde{G}^X \tag{6.5}$$

for the complete time-reversal operation. If time-reversal covariance holds, then according to Eq. (4.16),

$$G^T = G; \tag{6.6}$$

i.e., G^T fulfills the same equations as G.

The $\sim$ operation is defined through Eqs. (6.3a)–(6.3c). Now we have to perform the now operation ^{X}. First we remark that

$$G^{0X} = G^0 \tag{6.7}$$

and that for matrix products one has

$$(G_1\, \Sigma\, G_2)^X = G_2{}^X\, \Sigma^X\, G_1{}^X$$

where the matrix product does not run over the Keldysh indices. From Eqs. (6.3a)–(6.3b) we obtain

$$G_R^T = G^0{}_R + G_R^T \Sigma_A [\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]^X G^0{}_R, \tag{6.8a}$$

$$G_A^T = G^0{}_A + G_A^T \Sigma_R [\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]^X G^0{}_A, \tag{6.8b}$$

$$\begin{aligned} G_\Delta^T = G^0{}_\Delta &+ (G_\Delta^T \Sigma_R [\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]^X + G_R^T \Sigma_\Delta [\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]^X) G^0{}_A \\ &+ G_R^T \Sigma_A [\tilde{G}_A, \tilde{G}_R, \tilde{G}_\Delta]^X G^0{}_\Delta. \end{aligned} \tag{6.8c}$$

Obviously to recover the original Eqs. (6.2),

$$\Sigma_{R,A,\Delta}[\tilde{G}_R, \tilde{G}_A, \tilde{G}_\Delta]^X = \Sigma_{A,R,\Delta}[G_A^T, G_R^T, G_\Delta^T] \tag{6.9}$$

is required. If this relationship is satisfied, then also Eq. (6.6) or, equivalently, Eq. (4.16) holds.

After these preliminaries, we may analyze the time reversal properties of different approximation schemes of the many-body theory.

VII. TIME REVERSAL OF THE RPA APPROXIMATION

The simplest interaction to be considered is that with some external scalar and vector potentials $\mathscr{V}(t)$, $\mathscr{A}(t)$. In this case the self-energy is local in time and space and it does not depend on any Green function. Its time reversal property is obvious. If further interactions are added, then still this term will be the only one that depends on the external fields and the time.

A more complicated structure of the self-energy occurs within the self-consistent RPA approximation for the Coulomb interactions between the fermions described by the Feynman–Keldysh diagram of Fig. 3. The full straight line represents the renormalized self-consistent fermion propagator, while wavy line represents the self-consistent screened Coulomb propagator (or two-time screened potential) the sum of all Coulomb self-energy corrections is pictured in Fig. 4. Accordingly,

$$\Sigma_{\eta,\eta'}(x, x') = i\, V_{\eta,\eta'}(x, x')\, G_{\eta,\eta'}(x, x')\, \eta' \tag{7.1}$$

and

$$V = v + v\, \Pi\, V \tag{7.2}$$

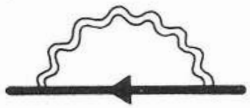

FIG. 3. RPA self-energy diagram.

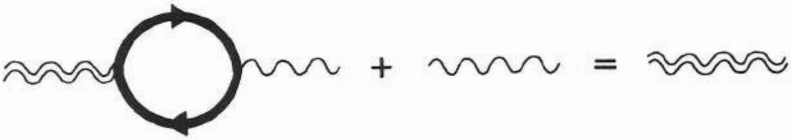

FIG. 4. Polarization diagrams in the RPA approximation.

with the polarization diagram given by

$$\Pi_{\eta,\eta'}(x,x') = -2i\,G_{\eta,\eta'}(x,x')\,G_{\eta',\eta}(x',x)\eta. \tag{7.3}$$

In Eq. (7.2) a symbolical matrix notation is used. The entities are matrices with respect to the space-time coordinates $x \equiv \mathbf{r}, t$ and Keldysh indices η. The bare Coulomb propagator is diagonal in time,

$$v_{\eta,\eta'}(x,x') \equiv v(|\mathbf{r}-\mathbf{r}'|)\,\delta(t-t')\,\delta_{\eta,\eta'}, \tag{7.4}$$

with the bare Coulomb potential,

$$v(\mathbf{r}) = e^2/|\mathbf{r}|. \tag{7.5}$$

Separating the singular part from the Coulomb propagator and introducing its nonsingular part by

$$\delta V = V - v,$$

we obtain

$$\Sigma_R(x,x') = -\frac{i}{2}\{\delta V_R(x,x')\,G_\Delta(x,x') + \delta V_\Delta(x,x')\,G_R(x,x'),$$

$$+\,v(x,x')[G_A(x,x') + G_R(x,x') - G_\Delta(x,x')]\} \tag{7.6a}$$

$$\Sigma_A(x,x') = -\frac{i}{2}\{\delta V_A(x,x')\,G_\Delta(x,x') + \delta V_\Delta(x,x')\,G_A(x,x')$$

$$+\,v(x,x')[G_A(x,x') + G_R(x,x') - G_\Delta(x,x')]\}, \tag{7.6b}$$

$$\Sigma_\Delta(x,x') = -\frac{i}{2}\{\delta V_R(x,x')\,G_R(x,x')$$

$$+\,\delta V_A(x,x')\,G_A(x,x') + \delta V_\Delta(x,x')\,G_\Delta(x,x')\}. \tag{7.6c}$$

and

$$V_{R,A} = v + v\,\Pi_{R,A}\,V_{R,A}, \tag{7.7a}$$

$$V_\Delta = V_R\,\Pi_\Delta\,V_A, \tag{7.7b}$$

together with

$$\Pi_R(x, x') = \frac{i}{2}\{G_R(x, x')\, G_\Delta(x', x) + G_\Delta(x, x')\, G_R(x', x)\}, \tag{7.8a}$$

$$\Pi_A(x, x') = \frac{i}{2}\{G_A(x, x')\, G_\Delta(x', x) + G_\Delta(x, x')\, G_A(x', x)\}, \tag{7.8b}$$

$$\Pi_\Delta(x, x') = \frac{i}{2}\{G_R(x, x')\, G_A(x', x) + G_A(x, x')\, G_R(x', x) + G_\Delta(x, x')\, G_\Delta(x', x)\}, \tag{7.8c}$$

The time-reversal covariance of the self-energy, Eq. (6.9), requires

$$\delta V_{R, A, \Delta}[\tilde{G}_R, \tilde{G}_A, \tilde{G}_\Delta]^X = \delta V_{A, R, \Delta}[G_A^T, G_R^T, G_\Delta^T], \tag{7.9}$$

which in turn implies

$$\Pi_{R, A, \Delta}[\tilde{G}_R, \tilde{G}_A, \tilde{G}_\Delta]^X = \Pi_{A, R, \Delta}[G_A^T, G_R^T, G_\Delta^T]. \tag{7.10}$$

In virtue of Eqs. (7.8a)–(7.8c) this is obviously satisfied.

This concludes the proof that the self-consistent RPA approximation for the Coulomb interaction is time-reserval covariant in the sense of Eq. (5.8).

The proof of this covariance in the case of the self-consistent RPA approximation for the electron–phonon interaction follows analogously. The self-consistency here includes also the phonon propagators. The diagrams are the same, but one has a non-trivial, non-singular unperturbed phonon propagator. One should also take into account the specific aspects of the boson theory in the Keldysh–Feynman rules.

VIII. CONCLUSIONS

The time-reversal property of a many-body Hamiltoninan determines a certain relationship, Eq. (4.16), between Keldysh–Green functions with different density matrices, showing that the same state may be achieved in evolving from the remote past to a certain time t as evolving in the negative time direction from the remote future state to the time $-t$, but with the time-reversed external electromagnetic fields. No direct statements concerning the existence and nature of the state in the remote future can be made a priori on this ground.

It has been shown that for exact solution there is an equivalent, but more explicit form of this relationship, Eq. (5.8), involving only the Keldysh–Green functions with the same density matrix. This last relationship looks like an invariance of the Keldysh–Green functions, defined as functionals of the free propagators, against a simultaneous permutation of the retarded and advanced indices, time-reversal of the EM field, interchange of the coordinate-time arguments and time mirroring.

This functional property is shown to hold also within the self-consistent RPA approximation, although its interpretation in the sense of Eq. (4.16) is not granted. In deriving Eq. (5.8), the introduction of a second Keldysh contour served as an important ingredient.

Although in most of our treatment we considered no spin and a single type of fermions (no bands), the results may easily be extended also to more general cases.

ACKNOWLEDGMENT

We thank H. Haug for suggesting these investigations and for helpful discussions.

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Printed by Catherine Press, Ltd., Tempelhof 41, B-8000 Brugge, Belgium

Exciton–KLO-Phonon Quantum Kinetics: Evidence of Memory Effects in Bulk GaAs

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(Received 4 April 1995; revised manuscript received 19 June 1995)

Oscillations of the transient four-wave-mixing signal with a period of about 100 fs are observed in bulk GaAs using 14 fs pulses tuned to the exciton resonance at low temperatures. The measurements are explained in terms of the non-Markovian quantum kinetics for electron-hole pairs due to LO-phonon scattering. It is shown that the observed oscillations are evidence for memory effects. The experiments provide a first test of the central ideas of quantum kinetics, in which the effects of quantum coherence and of dissipation are intrinsically connected.

PACS numbers: 72.20.Jv, 42.50.Md, 42.65.Re, 78.47.+p

Ultrafast relaxation in liquids and solids can directly be investigated in the time domain by femtosecond spectroscopy. The coupling of electronic excitations to a vibrational mode is a particularly interesting example. Semiconductor quantum dots, for which this coupling has recently been investigated [1,2], can be considered as an inhomogeneously broadened ensemble of two-level systems with a diagonal coupling to longitudinal optical (LO) phonons. This coupling through the Franck-Condon mechanism gives rise to quantum beats in the four-wave-mixing signal with exactly the phonon frequency [2], which are not related to non-Markovian relaxation. In bulk (polar) semiconductors, where the electronic excitations have a continuous band spectrum, the coupling to the optical phonons results in real intraband scattering transitions and provides the fastest relaxation and dephasing mechanism at low to moderate excitation densities. The ultrafast scattering kinetics in band states is thus distinctly different from the dynamics of discrete electronic states coupled to a lattice vibration mode. Particularly for time intervals, which are short compared to the period of an optical lattice oscillation period ($\simeq$ 115 fs in GaAs), the kinetics can no longer be described by the classical Boltzmann kinetics with its completed energy-conserving collisions. Instead, quantum kinetics has to be used in order to account for the partially coherent nature of electronic states in the band. *This quantum coherence gives rise to memory effects* [3–5]. Earlier solutions of the non-Markovian quantum kinetic equations for single-pulse excitation in spectral vicinity of the band edge indicated that the coupling to optical phonons gives rise to a periodic modulation superimposed on the polarization decay [6,7].

In order to test the theoretical prediction for bulk semiconductors we employ transient four-wave-mixing (FWM) experiments which are directly compared with solutions of the quantum kinetic equations under identical conditions. As usual, two pulses (of equal linear polarization) with wave vectors $\vec{q}_1$ and $\vec{q}_2$, respectively, are delayed in time and focused onto the sample. The self-diffracted signal in direction $2\vec{q}_2 - \vec{q}_1$ is detected as a function of time delay τ. The pulses are derived from a laser system similar to that of Ref. [8]. The sech^2-shaped pulses have a temporal full width at half maximum (FWHM) of 14.2 fs and a spectral FWHM of 87 meV, resulting in a bandwidth product of 0.30 close to the theoretical limit of 0.315. The autocorrelation has been taken under identical conditions as the FWM data. Every component introduced into the beams (like, e.g., neutral density filters) has been precompensated in a four-prism sequence. The sample is a high quality GaAs/Al$_x$Ga$_{1-x}$As ($x = 0.3$) double heterostructure grown by metal-organic vapor phase epitaxy with a thickness of the bulk GaAs layer of 0.6 μm, resulting in a small optical density of 0.3 for continuum states at a lattice temperature of 77 K. The sample is glued to a sapphire substrate; its front side is antireflection coated. Electron-hole densities quoted are determined via the measured total incident flux and the spot radius of 35 μm, measured with a knife edge technique. For the effective absorption coefficient we have used half of the measured unsaturated continuum value of 1.1×10^4 cm^{-1}, since about half of the laser spectrum is below the band gap. Figure 1 exhibits typical signals as a function of time delay for three different excited electron-hole pair densities. The resonant excitation conditions can be seen from comparison of the laser spectrum with the FWM spectra in Fig. 2. The FWM signal (Fig. 1) exhibits a quantum beat [9] behavior, the period of which is density independent. For increasing density the modulation becomes less pronounced. We have fitted

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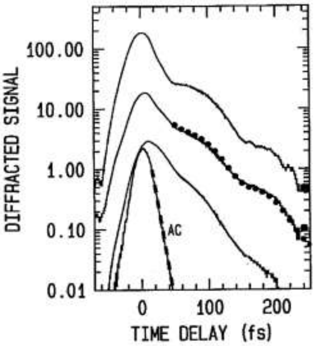

FIG. 1. Experiment: transient four-wave mixing on GaAs at 77 K. The diffracted signal is shown as a function of time delay for three different carrier densities (from top to bottom: 1.2×10^{16}, 1.9×10^{16}, and 6.3×10^{16} cm^{-3}). The curves are vertically displaced for clarity. The dots are the result of the quantum kinetic theory. The curve labeled *AC* is the autocorrelation of the laser pulses, the exponential wings of which are well fitted by a 6.5 fs time constant (dashed line).

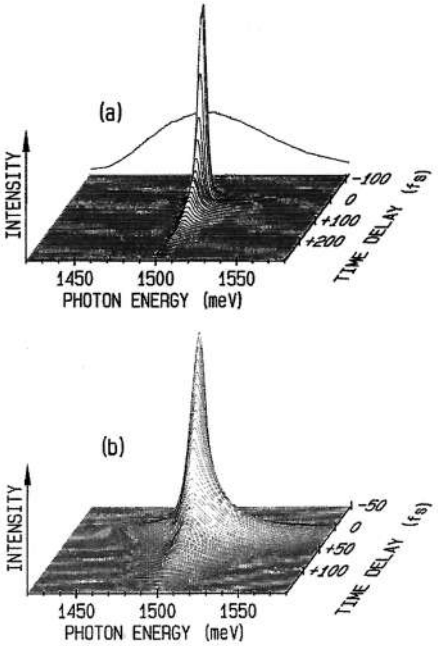

FIG. 2. Experiment: spectrally resolved four-wave-mixing signal as a function of time delay for a density of (a) 1.9×10^{16} and (b) 6.3×10^{16} cm^{-3}; other parameters as in Fig. 1. The resonant excitation condition can be seen from the laser spectrum depicted in the background of (a).

the decays in Fig. 1 with a function of the form $\sim [1 + a\sin(\omega_{\rm osc}\tau - \alpha)]\exp(-\tau/T_{\rm eff})$, obtaining best fits with periods $T_{\rm osc} = 2\pi/\omega_{\rm osc}$ of 100, 98, and 98 fs for the densities 1.2×10^{16}, 1.9×10^{16}, and 6.3×10^{16} cm^{-3}. The decay time constants $T_{\rm eff}$ are 44.1, 46.7, and 32.5 fs and the modulation amplitudes a 0.26, 0.17, and 0.17, respectively. For yet higher densities the modulation is less pronounced; for lower densities the worse signal-to-noise ratio inhibits detailed analysis. Interestingly, we observe only one peak in the FWM spectra as a function of time delay (Fig. 2) and no satellite related to the LO-phonon energy. Via the Fourier theorem this is equivalent to the absence of oscillatory structures in the signal as a function of time for any given time delay. From the density-independent modulation period we can clearly exclude any interpretation along the lines of Rabi or plasmon oscillations. Furthermore, the magnitude of the expected period is completely off for either mechanism. Propagation effects would also lead to a much longer oscillation period. *Surprisingly, the observed period of about* 100 *fs is smaller than expected from the well-established LO-phonon energy in GaAs of* $\hbar\omega_{\rm LO} = 36$ *meV equivalent to an oscillation period of* 115 *fs.*

For the theoretical analysis we use the semiconductor Bloch equations [10] combined with the retarded collision integrals for the LO-phonon scattering. This accounts for the important fact that the lattice cannot react on a time scale shorter than a lattice vibration period. The scattering rates of this delayed, partially coherent early time regime can be derived either with nonequilibrium Green functions (GF's) [3–6,11,12] or, alternatively, in a density matrix theory [13–15]. We use the nonequilibrium GF theory which allows the inclusion of nonperturbative effects by partial summations. It results in coupled nonlinear integrodifferential equations for the density matrix $\rho_{\mu\nu,k}(t) = \langle a^{\dagger}_{\nu,k}(t) a_{\mu,k}(t)\rangle$ (μ, ν are band indices, k is the momentum), which is just the equal time limit of the two-time particle propagator $G^{<}_{\mu\nu,k}(t,t') = i\langle a^{\dagger}_{\nu,k}(t) a_{\mu,k}(t')\rangle$ with $\hbar = 1$. The resulting collision rates for the equal time density matrix contain integrals over the history of the system. A typical term has the form $\sum_{\sigma}\int_{-\infty}^{t} dt'\, \Sigma^{>}_{\mu\sigma,k}(t,t') G^{<}_{\sigma\nu,k}(t',t)$, where $\Sigma^{\lessgtr}_{\mu\nu,k}(t,t')$ are the scattering self-energies. These self-energies are taken in the simple loop approximation $\Sigma^{\lessgtr}_{\mu\nu,k}(t,t') = i\sum_{q} g_q^2 D_q^{\lessgtr}(t,t') G^{\lessgtr}_{\mu\nu,k-q}(t,t')$, where for simplicity the phonons are considered as a thermal bath, and their propagators $D^{\lessgtr}$ are taken in the free-particle approximation. g_q is the Fröhlich interaction matrix element. The quantum kinetic equation is finally closed by the generalized Kadanoff-Baym ansatz (GKBA)

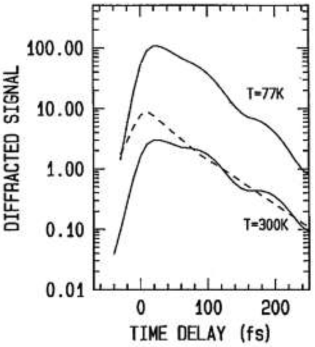

FIG. 3. Quantum kinetic theory: time-integrated four-wave-mixing signal as a function of time delay for GaAs at two different temperatures. $T = 77$ K, full line: theory with an additional dephasing mechanism with $T_2 = 143$ fs (same as dots in Fig. 1); $T = 300$ K, full line: quantum kinetic theory for phonon scattering only; dashed line: Markovian limit.

[3,4] which relates the two-time particle propagator to the density matrix: $G^{<}_{\mu\nu,k}(t,t') = -\sum_\sigma G^r_{\mu\sigma,k}(t,t')\rho_{\sigma\nu,k}(t')$ for $t > t'$. For $t < t'$ the advanced GF G^a enters in this relation. The GKBA is exact for a mean-field Hamiltonian [7] because the correction term to the GKBA is proportional to a scattering self-energy. With the GKBA one typical term of the scattering rates becomes, e.g., $\sum_{\sigma,q}\int_{-\infty}^{t} dt'\, \mathrm{K}(t,t')\rho_{\mu\sigma,k-q}(t')[1 - \rho_{\sigma\nu,k}(t')]$, where the memory kernel is given by $\mathrm{K}(t,t') = g_q^2 e^{i\omega_{\mathrm{LO}}(t-t')} G^r_{\mu\sigma,k-q}(t,t') G^a_{\sigma\mu,k}(t',t) n_q$, with the phonon distribution n_q. Previously [3,4,6], diagonal free-particle Wigner-Weisskopf approximations for the spectral functions have been used. For long times the scattering rate approaches asymptotically the Boltzmann limit with a broadened energy-conserving delta function. With the diagonal approximation Thoai and Haug [16] showed that oscillations with the LO-phonon frequency [contained in $D^{\lessgtr}(t,t')$] are superimposed on the decay of the interband polarization induced by a short laser pulse. These oscillations are due to interference of the various LO-phonon sideband polarizations and can therefore be interpreted as LO-phonon quantum beats. Here, we have improved the description of the spectral functions by a mean-field approximation [7] for which the GKBA is still exact. In this description—combined again with a Wigner-Weisskopf collision damping—the spectral functions are calculated under the influence of the coherent light pulses and the Coulomb Hartree-Fock interaction consistently with the density matrix equations. Thus all optical band mixing effects and the important excitonic effects are now contained consistently in both the semiconductor Bloch equations and the spectral functions. We solve the quantum kinetic equations for two delayed excitation pulses $E_0(t)e^{i\vec{q}_1\cdot\vec{x}}$ and $E_0(t-\tau)e^{i\vec{q}_2\cdot\vec{x}}$, with $E_0(t) = E_0\, e^{-i\omega_0 t}/\cosh(t/\Delta t)$ and a FWHM of the intensity of 15 fs, corresponding to the experiment. The diffracted signal in direction $2\vec{q}_2 - \vec{q}_1$ is calculated in the following manner. The interference pattern with wave vector $\Delta\vec{q} = \vec{q}_2 - \vec{q}_1$ can transfer multiples of $\Delta\vec{q}$ to the two beams. We therefore calculate the time dependence of the total polarization $P(t,\tau,\phi)$ as a function of the phase $\phi = \vec{x}\cdot\Delta\vec{q}$ and time delay τ. The various diffracted orders $P_n(t,\tau)$ can then be projected out of the calculated function $P(t,\tau,\phi)$. As in the experiment, we choose the direction $2\vec{q}_2 - \vec{q}_1$ corresponding to $n = 2$. This sequential procedure is essential for the iteration of the huge set of coupled equations. Furthermore, we neglect propagation effects which are of minor importance at low optical densities. In Fig. 3 we show the calculated time-integrated diffracted signal $\int_{-\infty}^{+\infty} dt|P_2(t,\tau)|^2$ as a function of time delay for GaAs parameters at 77 K: heavy-hole-electron mass ratio $m_h/m_e = 6.67$, exciton Rydberg $R_y = 4.15$ meV, and Bohr radius $a_B = 12.5$ nm. The collisional damping is taken to be $\gamma_c = \gamma_v = 1$ meV. The excited electron-hole density is $n = 1.6\times10^{16}$ cm^{-3}, comparable to the experiment. In order to model the experiment at 77 K we have added an additional, phenomenological dephasing mechanism with $T_2 = 143$ fs. This additional dephasing mechanism is most likely the Coulomb scattering among carriers, consistent with the fact that the oscillations vanish at higher excitation densities. Except for very short delay times, the agreement between theory and experiment is very good. From the same fitting procedure as in the experiment we obtain $T_{\mathrm{osc}} = 98$ fs, $T_{\mathrm{eff}} = 45$ fs, and $a = 0.17$. The oscillation period of about 100 fs can be interpreted as a simple beating of interband-polarization components with frequencies ω and ω' which are connected by coherent LO-phonon scattering. These frequency components are resonant with the band states k and k': $\hbar\omega = \hbar^2k^2/2\mu + E'_g$ and $\hbar\omega' = \hbar^2k'^2/2\mu + E'_g$, where μ is the reduced electron-heavy-hole mass $\mu = m_e m_h/(m_e + m_h)$ and E'_g is the Hartree-Fock renormalized band gap. The two interfering momentum states are coupled by an LO-phonon scattering event in the conduction band $\hbar^2(k'^2 - k^2)/2m_e = \hbar\omega_{\mathrm{LO}}$, from which one gets $\omega_{\mathrm{osc}} = \omega' - \omega = (1 + m_e/m_h)\,\omega_{\mathrm{LO}}$, which yields a period of 100 fs, close to the experiment. We have checked that this interpretation holds indeed for various mass ratios up to $m_h/m_e = 1$. The contribution of the scattering in the flat valence band is much smaller because the Fröhlich coupling is weak for large momentum transfer. This interpretation clearly shows that the observed effect is connected to band-to-band transitions; however, excitonic effects are important because they increase the necessary coherence considerably. In fact, the oscillation amplitude becomes very small if the

electron-hole attraction is switched off in our calculations. Note that we have treated the LO phonons as a thermal bath. In reality a certain number of phonons is generated by the hot carriers excited by the spectrally rather broad 15 fs pulse. It is known for the numerically much simpler diagonal approximation for the spectral functions that the combined quantum kinetics of the electronic excitations and the LO phonons [15,16] provide both a faster relaxation and a larger amplitude of the quantum beats. Therefore we also present calculations at 300 K with no additional dephasing time and damping constants which are determined self-consistently using Fermi's golden rule. The actual phonon populations in the experiment are somewhere in between these two curves which, however, exhibit no qualitative difference. We also show *that the observed oscillations as a function of time delay are absent in the Markovian limit of the theory* (dashed curve in Fig. 3). Figure 4 finally depicts the calculated FWM spectra at 77 K versus delay time. They are in good qualitative agreement with the corresponding experimental FWM spectra (Fig. 2), even though the theoretical spectra tend to be somewhat broader. In particular, the FWM signal exhibits no sign of oscillations in real time t, showing that the dynamics in the delay time τ and in real time are not simply connected.

Because of the good agreement between theory and experiment, the reported measurements provide a first direct experimental verification of the basic ideas of quantum kinetics. According to quantum kinetics, quantum coherence—here observed in the form of LO-phonon related quantum beats—cannot be separated from dephasing and relaxation on time scales which are shorter or comparable to the inverse frequency of characteristic resonances in the system.

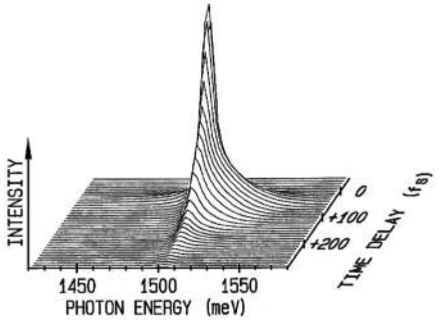

FIG. 4. Quantum kinetic theory: spectrally resolved diffracted signal versus time delay. Parameters as in Fig. 3 for $T = 77$ K.

We acknowledge support by the DFG and the Volkswagen-Stiftung. The research of M. W. has been supported by the Krupp-Stiftung.

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Ultrafast Electron Redistribution through Coulomb Scattering in Undoped GaAs: Experiment and Theory

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(Received 9 July 1996)

We report the observation of spectral hole burning exclusively due to the nonequilibrium electron population in a nondegenerate pump-test configuration. The rapid redistribution of electrons as well as the other features of the differential absorption spectra are well described by a theory using quantum-kinetic bare Coulomb collisions in the framework of the semiconductor Bloch equations. [S0031-9007(96)01740-1]

PACS numbers: 78.47.+p, 42.65.Re, 71.10.-w, 78.20.Bh

The redistribution of nonequilibrium carrier populations in semiconductors has attracted considerable interest in the last two decades. The tremendous progress of femtosecond lasers in terms of pulse duration and stability has rendered possible the observation of the initial stages of carrier relaxation [1–6] and the study of very low carrier densities [6]. However, studying the contributions of different scattering mechanisms such as LO-phonon and carrier-carrier scattering remains a difficult task, because most experiments measure a combination of electron and hole dynamics and the signals in ultrashort-pulse experiments contain coherence effects [7] and are not solely population dependent. Indeed, standard pump-test experiments [1,3,5,6] measure the absorption saturation due to the Pauli exclusion principle and are sensitive to the sum of the electron and hole distribution functions (f_e and f_h, respectively) while time-resolved luminescence experiments [2] measure the product $f_e f_h$. A selective investigation of the hole dynamics has been used in Ref. [8] to measure the heavy-hole thermalization time. However, this method cannot measure the complete hole distribution and the initially injected hole population.

Here we have used a modified pump-test scheme in order to isolate the electron dynamics [4]: the pump pulse excites electrons from the heavy-hole (HH) and light-hole (LH) valence bands while the test pulse probes the absorption saturation of the interband transition from the split-off (SO) valence band to the conduction band C (see inset of Fig. 1). Because of the large spin-orbit splitting in GaAs (340 meV), no holes are present in the SO band and the differential absorption signal $-\Delta\alpha = \alpha_{\text{without-pump}} - \alpha_{\text{with-pump}}$ depends on the electron distribution only. This method has a further important advantage: pump and test are at different wavelengths which allows the observation of spectral hole burning due to the initially injected electron population without any contribution from the induced-grating coherence effect [7,9] which considerably complicates the interpretation of standard pump-test experiments. Moreover, due to the isotropic matrix element of the SO-C transition, the measured signal is equally sensitive to the presence of electrons with all possible wave vector directions.

We report the first observation of hole burning which can be attributed exclusively to the electron population. While in previous experiments hole burning was not discernible [4], recent ameliorations of the experimental setup have permitted the observation of hole-burning signals for carrier densities ranging from a few 10^{15} to a few 10^{18} cm^{-3} and for excess photon energies ranging from 50 to 110 meV. The ensemble of these results will be discussed elsewhere. In this Letter, we concentrate on the very short pump-test delay times at moderate densities, the rapid redistribution of electrons causing the

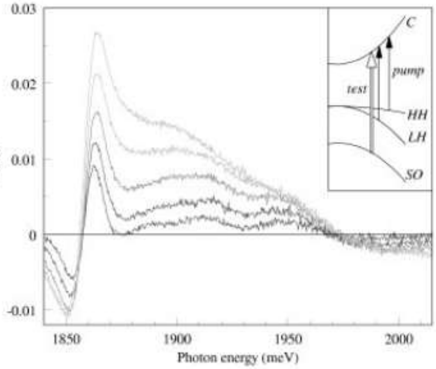

FIG. 1. Differential absorption spectra for the following pump-test delay times: −80, −40, 0, 40, and 80 fs. The inset shows the pump-test configuration.

disappearance of hole burning, and the comparison of this early-time behavior with theory.

We used a Ti:sapphire mode-locked oscillator (Coherent Mira) and regenerative amplifier system (Coherent RegA) both pumped by an argon-ion laser. Part of the output generates a spectral continuum and is used as the test pulse. After chirp compensation of the continuum with a combination of prisms and gratings we obtain nearly Fourier-transform-limited pulses with a duration of 30 and 130 fs for the test and pump pulses, respectively. The pulses were focused down to 50 and 150 μm for the test and pump, respectively, on the sample which was an intrinsic GaAs layer of thickness $d = 0.65$ μm antireflection coated on both sides and held at 15 K. In order to minimize the noise, a shutter is used in the optical path of the pump at an 8-Hz rate. In addition, a reference beam is simultaneously detected on a different track of the CCD detector and is used to normalize the transmitted test beam.

The differential absorption spectra for a pump width of 15 meV and a pump energy of 1.589 eV (excess energy of 70 meV with respect to the band gap) and for various pump-test delay times are shown in Fig. 1. The carrier density was estimated to be 6×10^{16} cm^{-3}. The zero delay is defined as the coincidence of the pump and test maxima and is taken at the middle of the integrated-signal rise time. The spectra show two broad peaks at about 1.913 and 1.950 eV due to spectral hole burning associated with the electron populations photoexcited from the LH and HH bands, respectively. Note that the two peaks disappear already before the end of the pump pulse. While it is clear that the signal in the spectral region from 1.88 to 1.96 eV is dominated by the induced transmission due to the electron population, the induced absorption above 1.97 eV and the oscillatory structure around 1.86 eV cannot be easily explained. Furthermore, even in the hole-burning region the differential absorption spectra do not directly reflect $f_e(t)$ due to energy-time uncertainty and excitonic Coulomb effects. Therefore, a theoretical analysis in terms of a quantum-kinetic approach is necessary, since the commonly used theories based on the golden-rule long-time limit are not applicable at such ultrashort times.

Quantum kinetics is a generic name for the theory describing kinetics with memory on very short time scales (see Ref. [10] for a review). The Markovian rate equation which has been so successful in the description of picosecond and nanosecond phenomena should be regarded as a limiting case of the quantum kinetics. In the experimental results described in this Letter, many scattering mechanisms are involved. It is most interesting to look at the limited short-time regime where Coulomb scattering dominates because, although the quantum kinetics of the electron-LO-phonon interaction (at low carrier densities) has already received attention in the past few years [11–15] and some observed quantum-kinetic effects have been explained [16], no treatment of the quantum-kinetic Coulomb scattering for real experiments has been attempted yet.

Coulomb scattering presents peculiar features which require imperatively a quantum-kinetic formulation. It is well known already from the equilibrium theory of screening that the screened Coulomb potential has an $\frac{1}{q^2}$ singularity as $q \to 0$ at any finite frequency ω. The singularity is absent only at $\omega = 0$. However, a vanishing frequency implies an infinite time. Therefore, the singularity is always present and plays an important role at short time scales. This singularity which corresponds to that of the bare Coulomb potential is fatal for the Boltzmann equation since the argument of the energy-conserving δ function also vanishes at $q = 0$ and the collision integral diverges. The energy-time uncertainty which is taken into account by quantum kinetics automatically eliminates the divergence [17].

For times less than a typical plasma period, screening is negligible [17] and thus the relatively complicated theory of time-dependent screening [18–20] can be avoided. We may also simplify the theoretical task by restricting our calculations to times less than or comparable with the effective interband polarization decay time. In our configuration, where there is no interference between the pump and test polarizations, one can use a simple phenomenological description of the polarization collision term and concentrate only on the quantum-kinetic collision terms of the electron and hole populations excited by the pump.

The semiconductor Bloch equations [21] for the populations and polarizations in the case of the pump field are

$$\frac{\partial f_{e,\vec{k}}(t)}{\partial t} = \sum_{\alpha} \Im\{\Omega^{P*}_{\alpha,\vec{k}}(t) p_{\alpha,\vec{k}}(t)\} + \frac{\partial f_{e,\vec{k}}(t)}{\partial t}\bigg|_{\mathrm{coll}} \qquad (1)$$

$$\frac{\partial f_{\alpha,\vec{k}}(t)}{\partial t} = \Im\{\Omega^{P*}_{\alpha,\vec{k}}(t) p_{\alpha,\vec{k}}(t)\} + \frac{\partial f_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\mathrm{coll}} \qquad (2)$$

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\epsilon_{e,\vec{k}} + \epsilon_{\alpha,\vec{k}} - \hbar\omega_P)\right] p_{\alpha,\vec{k}}(t) = \frac{i}{2}\Omega^{P}_{\alpha,\vec{k}}(t)(1 - f_{e,\vec{k}} - f_{\alpha,\vec{k}}) + \frac{\partial p_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\mathrm{coll}}. \qquad (3)$$

Here $\alpha = \mathrm{HH}, \mathrm{LH}$ and the renormalized energies ϵ and Rabi frequencies Ω are given by

$$\epsilon_{i,\vec{k}}(t) = \epsilon^0_{i,\vec{k}} - \sum_{\vec{k}'} V_{\vec{k}-\vec{k}'} f_{i,\vec{k}'}(t), \qquad i = e, \mathrm{HH}, \mathrm{LH}, \qquad (4)$$

$$\hbar\Omega^{P}_{\alpha,\vec{k}}(t) = d_{\alpha,\vec{k}} \mathcal{E}_P(t) + 2\sum_{\vec{k}'} V_{\vec{k}-\vec{k}'} p_{\alpha,\vec{k}'}(t). \qquad (5)$$

In the above equations, $\mathcal{E}_P(t)$ is the envelope of the pump field with frequency ω_P, $d_{\alpha,\vec{k}}$ are the respective interband dipole matrix elements, and $V_{\vec{q}}$ is the Fourier transform of the Coulomb potential. Since we consider an isotropic model with dipole matrix elements independent of the field polarization and $\vec{k}$, we take them to be equal for heavy and light holes.

The quantum-kinetic collision terms for the populations are $(i, i' = e, \mathrm{HH}, \mathrm{LH})$:

$$\frac{\partial f_{i,\vec{k}}(t)}{\partial t}\bigg|_{\mathrm{coll}} = -\frac{4}{\hbar^2}\sum_{i'}\int_{-\infty}^{t} dt' \int \frac{d\vec{q}}{(2\pi)^3}\int \frac{d\vec{k}'}{(2\pi)^3}\,|V(q)|^2 \cos\left(\frac{(t-t')}{\hbar}(\epsilon^0_{i,\vec{k}} + \epsilon^0_{i',\vec{k}'} - \epsilon^0_{i,\vec{k}-q} - \epsilon^0_{i',\vec{k}'+q})\right) \times \{f_{i,\vec{k}}(t')f_{i',\vec{k}'}(t')[1 - f_{i,\vec{k}-\vec{q}}(t')][1 - f_{i',\vec{k}'+\vec{q}}(t')] - f_{i,\vec{k}-\vec{q}}(t')f_{i',\vec{k}'+\vec{q}}(t')[1 - f_{i,\vec{k}}(t')][1 - f_{i',\vec{k}'}(t')]\}. \tag{6}$$

In the Markovian limit one gets from this equation the usual golden-rule rate equation.

The phenomenological collision term of the polarization is

$$\frac{\partial p_{\alpha,\vec{k}}(t)}{\partial t}\bigg|_{\mathrm{coll}} = -\frac{1}{T_2}\,p_{\alpha,\vec{k}}(t), \qquad \alpha = \mathrm{HH}, \mathrm{LH}. \tag{7}$$

In the case of the test pulse, one may retain only the electron population created by the pump and, therefore, we have to consider only the test polarization equation

$$\left[\frac{\partial}{\partial t} + \frac{i}{\hbar}(\epsilon_{e,\vec{k}} + \epsilon^0_{\mathrm{SO},\vec{k}} - \hbar\omega_T)\right]p_{\mathrm{SO},\vec{k}}(t) = \frac{i}{2}\,\Omega^T_{\mathrm{SO},\vec{k}}(t)(1 - f_{e,\vec{k}}) - \frac{p_{\mathrm{SO},\vec{k}}(t)}{T_2}, \tag{8}$$

where the unrenormalized energy of the SO holes $\epsilon^0_{\mathrm{SO},\vec{k}}$ and the renormalized SO Rabi frequency were introduced,

$$\hbar\Omega^T_{\mathrm{SO},\vec{k}}(t) = d_{\mathrm{SO},\vec{k}}\,\mathcal{E}_T(t) + 2\sum_{\vec{k}'} V_{\vec{k}-\vec{k}'}\,p_{\mathrm{SO},\vec{k}'}(t). \tag{9}$$

Here $\mathcal{E}_T$ is the envelope of the test field having the carrier frequency ω_T and $d_{\mathrm{SO},\vec{k}}$ the SO dipole matrix element. To obtain the absorption spectrum, one has to perform a Fourier transform of the test polarization summed over all $\vec{k}$.

The electron population excited by the pump acts first as a final-state blocking factor on the right hand side of Eq. (8) and second as a band shift through the Fock energy of the renormalized electron energies [Eq. (4)]. These effects are all mixed up, vary in time, and get Fourier transformed and therefore it is very difficult to discuss them separately. In addition, specific Coulomb spectral effects of the Wannier operator in the polarization equation (exciton and Coulomb enhancement) impede a simple additive interpretation.

Using a 130-fs pump pulse we performed calculations of the excited populations up to 300 fs which corresponds roughly to the plasma period at our pair density of 6×10^{16} cm^{-3}. We took $T_2 = 130$ fs. The effective mass ratios were taken to be integer ($m_{\mathrm{HH}}/m_e = 6$, $m_{\mathrm{LH}}/m_e = 1$, $m_{\mathrm{SO}}/m_e = 2$) for convenience of the numerical algorithm. The numerical calculation on a discrete lattice of k-space points neglects low-momentum-transfer contributions which in the Coulomb case are important. Nevertheless, we take into account low-momentum-transfer collisions within a Landau approximation through a Taylor expansion around $\vec{q} = 0$. The complete quantum-kinetic calculation gives rise to an electron population as shown in Fig. 2. The electron-population peaks are rapidly smeared out and already at about 300 fs after the pump maximum the distribution is very close to a nondegenerate Fermi distribution. The calculated differential absorption spectra with a 30-fs test pulse are shown in Fig. 3. We did not consider delay times longer than 80 fs since their calculation involves information on the electron population for times above 300 fs due to the Fourier transform.

The agreement with the experiment is surprisingly good, although in many details quantitatively rough. The most remarkable achievement is the prediction that at about 80 fs after the pump maximum the induced hole burning is smeared out. This is related to the fact that the electron population is almost in equilibrium already at about 300 fs after the pump maximum. The only fit parameter was the phenomenological polarization relaxation time T_2. However, if T_2 is taken comparable or larger than the pump duration, it affects only the negative parts of the spectra slightly. In the comparison of theory and experiment, one has to take into account that the exact energy positions of the various features are affected by the roughness of the electron energy discretization of about 5 meV, by the slightly modified effective masses as well as by the inaccuracy of the numerical Fourier transform. The more pronounced valley above the band threshold (at about 15 meV) as compared to the experiment may be due to the neglect of LO-phonon emission, which provides the cooling of the electron system.

Both the experimental and the theoretical differential absorptions show a final-state occupation effect (hole burning) due to our narrow-band excitation and an oscillation

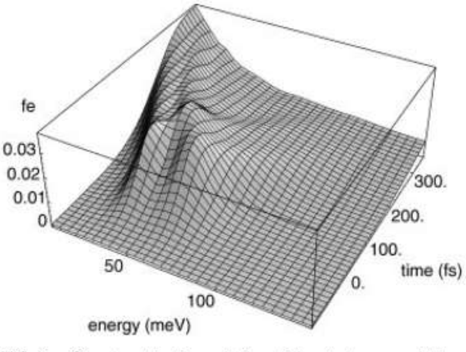

FIG. 2. Quantum-kinetic evolution of the electron population.

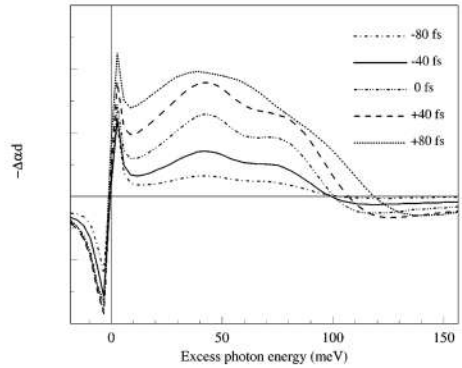

FIG. 3. Calculated differential absorption spectra for the same delay times as in Fig. 1.

that looks like an energy shift in the excitonic region, while the negative signal on the high-energy side of the excitation is mainly due to the nonlinearity introduced by the product of the occupation factor with the Coulomb force term

$$\frac{i}{\hbar}\sum_{\vec{k}'} V_{\vec{k}-\vec{k}'}\, p_{\mathrm{SO},\vec{k}'}(t)\,[1 - f_{e,\vec{k}}(t)], \qquad (10)$$

which stems from the replacement of the Rabi frequency by the renormalized one in the presence of the Coulomb interaction [see Eq. (9)]. Actually, this excitonic enhancement term plays an important role also in the oscillatory structure on the low-energy side in addition to the true Coulomb band shift of Eq. (4).

In conclusion, we have observed hole burning in a pump-test configuration free from coherence effects and succeeded in giving a satisfactory description of the differential absorption spectra for ultrashort delay times with the semiconductor Bloch equations using the quantum-kinetic bare Coulomb collision term for the populations. We stress that the use of quantum kinetics is mandatory due to the Coulomb singularity. The main success of the theory is determined by the structure of the semiconductor Bloch equations but the numerical prediction of the effective intraband relaxation time (smearing out of the hole burning) is due entirely to quantum kinetics. Our theoretical approach was highly simplified due to the specific experimental configuration implying a differential signal determined only by the electron population.

An improved version of the theory should include the quantum-kinetic polarization collision term, the buildup of screening, LO-phonon collisions, as well as the transition to the Markovian behavior in order to extend its applicability to higher densities and longer times. To incorporate such improvements in the theory for the test beam will still be an insurmountable task due to the Fourier transform, which requires an enormous time interval.

We are grateful to R. Planel and V. Thierry-Mieg (Laboratoire de Microstructures et de Microélectronique, Bagneux, France) for providing us with the GaAs sample and to M. Joffre and J.-P. Likforman for helpful discussions. We acknowledge partial support by the European Training and Mobility for Researchers program under Grant No. ERBFMGECT950019. The theoretical work has been supported by the Deutsche Forschungsgemeinschaft.

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PHYSICAL REVIEW B VOLUME 62, NUMBER 8 15 AUGUST 2000-II

Theory of THz emission from optically excited semiconductors in crossed electric and magnetic fields

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(Received 1 February 2000)

The coherent transients generated by femtosecond interband photoexcitation of a semiconductor in crossed electric and magnetic fields are calculated. The scattering with LO phonons is considered and the complex interplay between excitation and dephasing is analyzed. While below the LO-phonon threshold the signal is not effectively damped, above the threshold damping takes place on a picosecond time scale, in qualitative agreement with corresponding experiments.

I. INTRODUCTION

Within the last decade considerable efforts have been devoted to the search for terahertz (THz) emitters as well as to the use of this radiation as a method of condensed matter spectroscopy. In this context a new research topic known as the dynamical Franz-Keldysh effect[1–5] developed. Several mechanisms have been proven to generate THz signals: coherent phonons,[6,7] optical rectification and instantaneous polarization[8–12] in magnetic fields,[13–15] cold plasma oscillations,[16] asymmetric double quantum wells,[17–19] heavy–light-hole beatings in quantum wells,[17,20] and Bloch oscillations in superlattices.[21–23] As different as these mechanisms are in detail, they all are footed on common ground: In a system with broken symmetry it is possible to create coherent wave packets by short laser pulses. When the wave packets are charged, the motion of the wave packets gives rise to a time-dependent dipole moment and a corresponding emission of electromagnetic radiation in the form of THz oscillations. With respect to emitter applications, the tunability and the amplitudes of the signals, as well as the damping of the oscillations, are of crucial interest.

In the present paper we show how THz radiation can be generated in bulk material exposed to perpendicular electric and magnetic fields, known as the Voigt geometry. Here the electric field serves to break the symmetry so that the optical excitation leads to a coherent generation of electrons and holes, while the frequency of the oscillations is determined by the magnetic field, which can be tuned readily. As the dominant damping process we investigate the interaction with LO phonons by calculating quantum-number-dependent transverse relaxation times. A main feature of our analysis is that these relaxation times are not taken as phenomenological fit parameters but are calculated microscopically. It has been found[27–29] that the relaxation time is considerably larger than that without applied fields. Our calculated relaxation times yield an explanation for the observed THz signals on a picosecond time scale. Furthermore, the complex interplay of excitation and the various transverse relaxation times could not be described by a simple classical model with a constant damping term.

The paper is organized as follows: First we diagonalize the one-particle Hamiltonian in the crossed fields and formulate the optical excitation and the scattering due to LO phonons in this representation. Then we derive the kinetic equations on a reduced subset of quantities, sufficient to express the current connected with the THz radiation. Finally we present numerical results and discuss them with respect to experimental observations.

II. THE REPRESENTATION OF A SHIFTED OSCILLATOR

For a magnetic field in the z direction and an electric field in the x direction, the Hamiltonian for an electron or hole $(i=e,h)$ is in the asymmetric Landau gauge $\vec{A}=x\mathcal{B}\vec{e}_y$, where $\vec{e}_y$ is a unit vector in the y direction, given by

$$H^i=\frac{1}{2m_i}\left(\frac{\hbar}{i}\vec{\nabla}-e^i\vec{e}_y x\mathcal{B}\right)^2-e^i\mathcal{E}x. \tag{2.1}$$

Because the potential energy depends only on the x coordinate, one can use plane waves in the y and z directions:

$$\psi(\vec{r})=\frac{e^{i(k_z z+k_y y)}}{\sqrt{L_z L_y}}\phi(x). \tag{2.2}$$

The resulting Hamiltonian for $\phi(x)$ is bilinear in x and can be put into the form

$$H^i=\frac{\hbar^2 k_z^2}{2m_i}-\frac{\hbar^2 d^2}{2m_i dx^2}+\frac{1}{2}m_i\omega_c^{i2}(x-X^i)^2-e^i\mathcal{E}X^i+\frac{(e\mathcal{E})^2}{2m_i\omega_c^{i2}}, \tag{2.3}$$

where the spatial shift of the oscillator origin is

$$X^i=\frac{e^i}{e}l^2 k_y+\frac{e^i\mathcal{E}l^2}{\hbar\omega_c^i}. \tag{2.4}$$

The cyclotron frequency ω_c^i and the magnetic length l are given by

$$\omega_c^i=\frac{e\mathcal{B}}{m_i},\quad l^2=\frac{\hbar}{e\mathcal{B}}. \tag{2.5}$$

In these formulas the electron and hole charges are $e^e=-e$ and $e^h=e$, respectively. The eigenfunctions are given by shifted oscillator eigenfunctions

0163-1829/2000/62(8)/5003(7)/$15.00 PRB 62 5003

$$\psi^i(\vec{r})=\frac{e^{i(k_z z+k_y y)}}{\sqrt{L_z L_y}}\phi_n(x-X^i), \tag{2.6}$$

where

$$\phi_n(x)=\frac{1}{\sqrt{\sqrt{\pi}l2^n n!}}e^{-x^2/2l^2}H_n(x/l),$$

$$H_n(x)=(-1)^n e^{x^2}\frac{d^n}{dx^n}e^{-x^2}, \tag{2.7}$$

and the spectrum is

$$\epsilon^i_{n,X,k_z}=\frac{\hbar^2k_z^2}{2m_i}+\hbar\omega^i_c(n+\tfrac{1}{2})-e^iX^i\mathcal{E}+\frac{e^2\mathcal{E}^2}{2m_i\omega_c^{i2}}, \tag{2.8}$$

which is composed of the kinetic free-particle energy in the z direction, the Landau energies, and two corrections due to the electric field. Note that the effect of the electric field is twofold: First the degeneracy of the energies with respect to X is lifted and second the shift in the wave functions becomes mass dependent.

Using this shifted oscillator basis we now treat the coupling to the light field. Because the optical wavelength is typically larger than l, the interband polarization has to be averaged spatially. Within the slowly varying envelope approximation, the averaged polarization is

$$P(t)=\frac{d}{V}\int d^3r\langle\hat{\Psi}_h(\vec{r},t)\hat{\Psi}_e(\vec{r},t)\rangle+\text{H.c.}, \tag{2.9}$$

where d is the matrix element between the Bloch states of the conduction and the valence band close to the band edge. For the optical transitions the momentum components are conserved, i.e., $k_z=-k'_z$, $k_y=-k'_y$, where the $\vec{k}$, $\vec{k}'$ wave vectors refer to the electron and hole, respectively. a^i_{n,X,k_z} are the annihilation operators for a particle i in the state n,X,k_z. Because the excitation is diagonal in k_y, the shift quantum numbers X differ for electrons and holes due to the field term,

$$X_e=-l^2k_y-\frac{|e|\mathcal{E}l^2}{\hbar\omega^e_c},\quad X_h=-l^2k_y+\frac{|e|\mathcal{E}l^2}{\hbar\omega^h_c}. \tag{2.10}$$

Thus the field operators for the electron and the hole $\hat{\Psi}_i(\vec{r},t)$ in the overlap integral are shifted differently. Expanding the field operators into the eigenfunctions (2.6) one gets

$$P(t)=\frac{d}{V}\sum_{n,n',X,k_z}C(\xi)_{nn'}\langle a^h_{n',X+\xi,-k_z}(t)a^e_{nXk_z}(t)\rangle+\text{H.c.}, \tag{2.11}$$

with $C(\xi)_{nn'}=\int dx\,\phi_n(x)\phi_{n'}(x-\xi)$, where $\xi=|e|\mathcal{E}l^2/\hbar\omega_c$ with $1/\omega_c=1/\omega^e_c+1/\omega^h_c=(m_e+m_h)/|e|B$. Again the effect of the electric field is twofold: Equation (2.11) shows that the creation and annihilation of the electron-hole pair is nonlocal (a hole is created at $X+\xi$ while the electron is created at X). This effect has been well examined in the purely electric case ($\mathcal{B}=0$), where it is known as optical rectification.[8] Moreover, the selection rules known from the purely magnetic case[24] ($\mathcal{E}=0$) are destroyed: the optical matrix element may be nondiagonal in the Landau-level quantum numbers. From the overlap integral one sees that the orthogonality of the shifted oscillator functions no longer applies, because the shift of the wave functions is different for electrons and holes. This point is crucial for our analysis: Only due to the symmetry-breaking effect of the electrical field does it become possible to excite wave packets with short pulses, which would otherwise be forbidden by the selection rules. As will be discussed later these wave packets and the corresponding intersubband polarization give rise to THz signals.

At low densities, where the electron-electron interaction can be neglected, the interaction with LO phonons is the dominant scattering mechanism. This interaction is usually modeled in the basis of plane waves by the Fröhlich coupling. To adapt this coupling to the Voigt geometry the shifted oscillator functions are expanded in terms of plane waves in order to calculate the matrix elements. Fermi's golden rule yields the transition rates

$$W^{\vec{q},i}_{n,X,k_z;n',X',k'_z}=\frac{2\pi C^i}{\hbar L_xL_yL_z}[1+N(\hbar\omega_{LO})]\delta(\epsilon^i_{nXk_z}-\epsilon^i_{n'X'k'_z}-\hbar\omega_{LO})\times\delta_{q_y,k_y-k'_y}\delta_{q_z,k_z-k'_z}\frac{\left|\int dx\,e^{iq_xx}\phi_n(x)\phi_{n'}(x-(X-X'))\right|^2}{q_x^2+(1/l^4)(X-X')^2+(k_z-k'_z)^2}, \tag{2.12}$$

with

$$C^i=\alpha_i\frac{4\pi\hbar(\hbar\omega_{LO})^{3/2}}{(2m_i)^{1/2}},$$

where

$$\alpha_i=\frac{e^2}{\hbar}\left(\frac{m_i}{2\hbar\omega_0}\right)^{1/2}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_0}\right)$$

is the dimensionless Fröhlich polaron coupling constant and m_i the effective mass of the electrons and holes; $N(\hbar\omega_{LO})=1/(e^{\beta\hbar\omega_{LO}}-1)$ is the thermal phonon distribution. The transition rates for the phonon absorption can be obtained through the ''detailed balance'' relation from the given rate due to the phonon emission process.

These transition rates can now be used to write the full collision terms for a Boltzmann scattering. Instead of treating the full kinetics of the density matrix elements, we will take only the dephasing of the off-diagonal elements by relaxation times into account. For this purpose we calculate the probability (per time) to scatter from a given state into all other states (neglecting the fact that this state may be occupied already). Summing over all possible scattering channels the result is a quantum-number-dependent inverse transverse relaxation time

$$\frac{1}{\tau^{i}_{nXk_z}}=\frac{1}{\tau^{i}_{nk_z}}=\sum_{n'X'k_z'}\sum_{\vec{q}} W^{\vec{q},i}_{n,X,k_z;n',X',k_z'}. \qquad (2.13)$$

Thus all scattering processes give rise to exponential decays. The corresponding characteristic times are individually calculated for any set of quantum numbers.

III. DERIVATION OF THE KINETICS ON A REDUCED SUBSET

Postponing the scattering kinetics for a moment and putting all quantum numbers n,X,k_z in one multi-index ν, the Hamiltonian for the electron-hole system interacting with a coherent light pulse is

$$H=\sum_{\nu}\left(\epsilon^{e}_{\nu}a^{e\dagger}_{\nu}a^{e}_{\nu}+\epsilon^{h}_{\nu}a^{h\dagger}_{\nu}a^{h}_{\nu}\right)$$
$$-\frac{1}{2}\left[dE_0(t)e^{i\omega t}\sum_{\nu\nu'}C_{\nu\nu'}a^{h}_{\nu'}a^{e}_{\nu}+\text{H.c.}\right], \qquad (3.1)$$

where $E_o(t)$ is the amplitude of the femtosecond pulse and ω is the central frequency. For the electron and hole subband density matrix elements we define $f^{i}_{\nu\nu'}\equiv\langle a^{i\dagger}_{\nu}a^{i}_{\nu'}\rangle$ with $i=e,h$. One has to distinguish between populations $(\nu=\nu')$ and intersubband polarizations $(\nu\neq\nu')$ by the index combination. The interband polarization components are defined as $P_{\nu\nu'}\equiv e^{-i\omega t}\langle a^{h}_{\nu}a^{e}_{\nu'}\rangle$. In the rotating-wave approximation the equations of motion are

$$\left(\frac{\partial}{\partial t}-\frac{i}{\hbar}(\epsilon^{e}_{\nu}-\epsilon^{e}_{\nu'})\right)f^{e}_{\nu\nu'}=-\frac{i}{2}\sum_{\mu\mu'}C_{\mu\mu'}(\Omega_R\delta_{\mu\nu}P_{\mu'\nu'}$$
$$-\Omega_R\delta_{\mu\nu'}P^{*}_{\mu'\nu}), \qquad (3.2)$$

$$\left(\frac{\partial}{\partial t}-\frac{i}{\hbar}(\epsilon^{h}_{\nu'}-\epsilon^{h}_{\nu})\right)f^{h}_{\nu\nu'}=-\frac{i}{2}\sum_{\mu\mu'}C_{\mu\mu'}(\Omega_R\delta_{\mu'\nu}P_{\nu'\mu}$$
$$-\Omega_R\delta_{\nu'\mu'}P^{*}_{\nu\mu}), \qquad (3.3)$$

$$\left(\frac{\partial}{\partial t}+\frac{i}{\hbar}(\epsilon^{e}_{\nu'}+\epsilon^{h}_{\nu}-\omega)\right)P_{\nu\nu'}=\frac{i}{2}\Omega_R\sum_{\mu\mu'}C_{\mu\mu'}(\delta_{\nu'\mu}\delta_{\nu\mu'}$$
$$-\delta_{\mu'\nu}f^{e}_{\mu\nu'}-\delta_{\nu'\mu}f^{h}_{\mu'\nu}), \qquad (3.4)$$

where $\hbar\Omega_R(t)=dE_0(t)$ is the Rabi frequency. In these equations two consequences of the symmetry-breaking effect of the electric field are easily observed. While the equations for the intraband matrix elements of electrons and holes are normally equal, they differ here by the coupling to the interband polarization via the optical field. Even more important is the observation that the intersubband polarization components $f^{i}_{\nu\nu'}$ are excited at all.[25,26] In the next paragraph it will be shown that this intersubband polarization is closely related to the THz signal. Thus it is the electric field in the Voigt geometry that allows the THz emission. In order to handle the large number of density matrix elements caused by the appearance of the intersubband polarizations $f^{i}_{\nu\nu'}$, an approximation scheme will be introduced, which is based on the properties of the optical matrix element. We showed that $C_{\nu,\nu'}=C_{n,n'}\delta_{X',X-\xi}\delta_{k_z,-k_z'}$. Although there is no longer a selection rule in the Landau levels, still the conservation of momentum enforces $\delta_{k_z,-k_z'}$ and the shift in the quantum number X to be fixed through $\delta_{X',X-\xi}$. Applying these rules to the equations one ends with a closed subset of equations. It has to be emphasized that this subset is closed as long as only the coherent part (3.2) to (3.4) is considered and scattering processes are neglected. On the other hand, the collision terms couple to quantities not initially induced by the light field. In this approximation these quantities are considered to be of minor importance. Into this reduced set, we insert finally in the quantum-number-dependent transverse relaxation times.

With the definitions $f^{i}_{nXkn'Xk}=f^{i}_{nn'Xk}, P_{nXkn',X-\xi,-k}=P_{nn',X-\xi/2,k}$, the equations become

$$\left(\frac{\partial}{\partial t}-i\nu^{e}\right)f^{e}_{nn',X-\xi/2,k}=-\frac{i}{2}\sum_{m}(C_{nm}\Omega_R P_{mn',X,-k}$$
$$-C_{n'm}\Omega_R P^{*}_{mn,X,-k})$$
$$+\left.\frac{\partial}{\partial t}\right|_{coll}f^{e}_{nn'Xk}, \qquad (3.5)$$

$$\left(\frac{\partial}{\partial t}-i\nu^{h}\right)f^{h}_{nn',X+\xi/2,k}=-\frac{i}{2}\sum_{m}(C_{mn}\Omega_R P_{n'm,X,k}$$
$$-C_{mn'}\Omega_R P^{*}_{nm,X,k})+\left.\frac{\partial}{\partial t}\right|_{coll}f^{h}_{nn'Xk}, \qquad (3.6)$$

$$\left(\frac{\partial}{\partial t}+i\nu^{P}\right)P_{nn'Xk}=\frac{i}{2}\Omega_R\Bigg(C_{n'n}-\sum_{m}(C_{mn}f^{e}_{mn',X-\xi/2,-k}$$
$$+C_{n'm}f^{h}_{mn,X+\xi/2,k})\Bigg)+\left.\frac{\partial}{\partial t}\right|_{coll}P_{nn'Xk}, \qquad (3.7)$$

$$\left.\frac{\partial}{\partial t}\right|_{coll}f^{i}_{nn'X\mp(\xi/2)k}=-\frac{f^{i}_{nn'X\mp(\xi/2)k}}{\frac{1}{2}(\tau^{i}_{nk_z}+\tau^{i}_{n'k_z})}\quad(n\neq n'), \qquad (3.8)$$

$$\left.\frac{\partial}{\partial t}\right|_{coll}P_{nn'Xk}=-\frac{P_{nn'Xk}}{\frac{1}{2}(\tau^{h}_{nk_z}+\tau^{e}_{n'k_z})}. \qquad (3.9)$$

Here ν^e, ν^h, and ν^P are defined as

$$\nu^i = \hbar \omega_c^i (n - n'), \quad i = e,h \tag{3.10}$$

$$\nu^P = e^e_{\nu'} + e^h_{h\nu} - \omega = \frac{\hbar^2 k_z^2}{2m_e} + \frac{\hbar^2 k_z^2}{2m_h} + \hbar\omega_c^e (n' + \tfrac{1}{2}) + \hbar\omega_c^h (n + \tfrac{1}{2}) - \frac{E^2}{B^2}\frac{m_c^e + m_c^h}{2} - \Delta, \tag{3.11}$$

where $\Delta = \omega - E_g$ is the detuning, i.e., the energy surplus of the light field with respect to the unrenormalized band gap. Note that there is no thermalization term in the equations with the populations $n = n'$. Due to the electric field the spectrum is not bounded from below if the spectrum is not limited by imposing boundary conditions. Therefore the particles do not relax toward a Fermi function. Populations that do not relax might seem to be a highly unphysical feature on the picosecond time scale we are interested in, but this matches our approximation scheme: The THz signal is generated by the intersubband polarization. With only transverse relaxation times the equations of populations and intersubband polarizations are only coupled on the short time scale of the pulse (on which thermalization is not very effective). After the pulse they decouple and the damping of the intersubband polarization is (in the model) perfectly described by the inverse lifetimes applied to these quantities. The longitudinal relaxation time is of the same order (up to a factor of 2) as the transverse relaxation time. Our calculations show that the transverse relaxation times are in the picosecond range, while the excitation pulses are taken to be 141 fs (full widths at half maximum of the intensity). Because the longitudinal relaxation time is much larger than the duration of the pulse, the approximation is well justified. Izumida *et al.*[13] did experiments with chirped pulses varying the pulse duration. While they observe differences for longer pulses between positive and negative chirp, which suggests occupation effects, there are no discrepancies for pulses of the short duration that we used. This is a confirmation that our approximation without thermalization is well applicable.

IV. THz CURRENT OSCILLATIONS

Charge oscillations and thus current oscillations lead to the emission of electromagnetic dipole radiation. For the inspected THz radiation only intraband contributions are taken into account, i.e., the contributions from the interband polarization are neglected. These contributions are governed by frequencies related to the band gap and are therefore in the optical range.

In the literature on THz radiation without a magnetic field the creation of nonlocal electron-hole pairs has been studied as ''instantaneous polarization'' and a distinction has been made between displacement and transport contribution.[8,10,11,9] All these low-frequency parts are included in the present model, although the displacement current is hardly visible in the range of the considered parameters.

Starting from the definition of the current density operator

$$\vec{J}^i = \frac{1}{V}\int d\vec{r}\,\frac{e^i}{2m_i}\hat{\Psi}_i^\dagger(\vec{r},t)\left[\frac{\hbar}{i}\nabla - e^i\vec{e}_y x \mathcal{B}\right]\hat{\Psi}_i(\vec{r},t) + \text{H.c.} \tag{4.1}$$

one gets by expanding the field operators in the shifted oscillator basis for the current components (again with $i = e,h$):

$$\langle J_z^i \rangle = \frac{e^i}{m_i V}\sum_{n,X,k_z} \hbar k_z f^i_{n,n,X,k_z}, \tag{4.2}$$

$$\langle J_y^i \rangle = -\frac{e^i \mathcal{E}}{\mathcal{B}}\frac{N^i}{V} - \frac{|e|\hbar\sqrt{2}}{m_i l V}\sum_{n,X,k_z}\sqrt{n+1}\,\text{Re}(f^i_{n+1,n,X,k_z}), \tag{4.3}$$

$$\langle J_x^i \rangle = \frac{-e^i\hbar\sqrt{2}}{m_i l V}\sum_{n,X,k_z}\sqrt{n+1}\,\text{Im}(f^i_{n+1,n,X,k_z}). \tag{4.4}$$

Obviously the current in the z direction vanishes, as the effect of the fields is confined to the x-y plane. The first term in the y direction describes a net current proportional to the population. It is mass independent and dependent on the sign of the charges; in the case of charge neutrality (the optical field creates as many electrons as holes) it vanishes. It is of conceptual interest to see that in spite of the electrical field, the coherent motion does not separate electrons and holes on a macroscopic scale. Only the asymmetry in their scattering behavior leads finally to a separation. The influence of this scattering is weak and can be neglected for very low densities. Therefore the electric field does not have to be calculated self-consistently. From Eqs. (4.2) to (4.4) it is obvious that all contributions to the current and to the radiated signal stem from the intersubband polarization. More exactly, the intersubband polarizations for which the Landau-level quantum numbers differ by one are contributing. Reexamining the equations with respect to these quantities, it is immediately clear that they oscillate with nothing but the cyclotron frequency after the femtosecond pulse.

V. NUMERICAL RESULTS AND DISCUSSION

The numerical evaluations are given for GaAs parameters with an LO-phonon energy $\hbar\omega_{LO} = 36$ meV. We consider here the contribution of the light holes only, which have been studied also in the corresponding experiments. As all experiments[27–29] were done at low temperatures, in our calculations $T = 0$ K is taken for the phonon bath, i.e., only phonon emission is considered. In all results the time integral over the Rabi frequency is taken to be 0.01π, and the full widths at half maximum of the intensity (field squared) is 141 fs (Gaussian pulse shape). Figure 1 shows the inverse lifetime for electrons for the lowest Landau levels as functions of the kinetic energy ($E_{kin} = \hbar^2 k_z^2 / 2m_e$) for a magnetic field $\mathcal{B} = 6$ T, where the electron and the light-hole cyclotron energies are $\hbar\omega_c^e = 10.5$ meV and $\hbar\omega_c^h = 8.5$ meV. In the lowest Landau subband with $n = 0$ one sees a smeared-out one-phonon threshold. For the scattering within this subband the threshold becomes flatter with increasing electric field. For the higher Landau subbands the damping increases already at lower kinetic energies because of the transitions to lower subbands. For $n = 1$ and $E = 6$ kV/cm, one can see

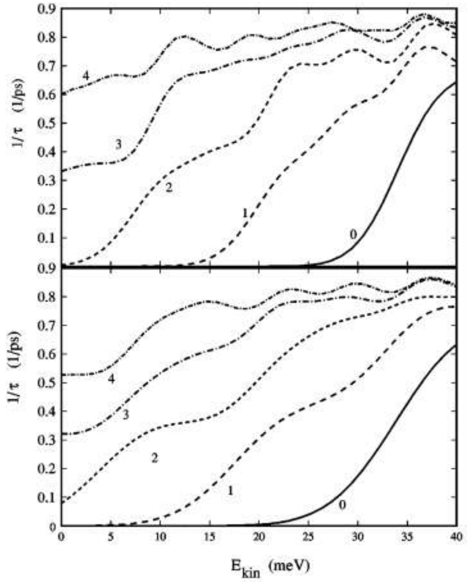

FIG. 1. Inverse of the transverse relaxation times τ^e_{n,k_z} for the electrons in various Landau levels versus $E_{kin}=\hbar^2k_z^2/2m_e$ for a magnetic field $B=6$ T and an electric field $E=4$ kV/cm (upper figure) and $E=6$ kV/cm (lower figure).

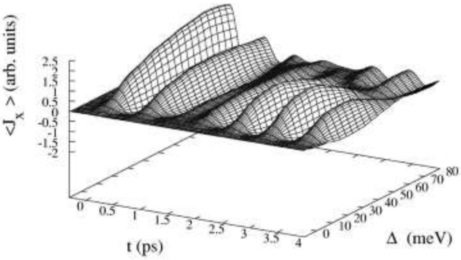

FIG. 2. The time evolution of the x component of the current versus detuning Δ for fixed fields: $E=4$ kV/cm, $B=4$ T.

clearly first the contributions due to the intersubband scattering by phonon emission to $n=0$, followed by the contributions due to the intrasubband scattering. Because the spectra of the subbands are not bounded from below due to the term $-e^i\mathcal{E}X^i$ intrasubband scattering is at least in principle also possible even at very small kinetic energies $\hbar^2k_z^2/2m_e$. But due to the localization of the wave function in the magnetic field, scattering over a distance, which is considerably larger than a few magnetic lengths (details are dependent on the shift due to the electric field and the quantum numbers of the wave functions), becomes extremely weak. Because the distance in the scattering process is limited, the energy gained by the nonlocal process in the electric field is restricted also. For the parameters we studied ($4\ \mathrm{T}<B<8$ T, 4 kV/cm $<\mathcal{E}<6$ kV/cm) this energy is by far to small to provide the 36 meV necessary for the emission of a LO phonon. Thus for intrasubband scattering the main contribution has to come from the kinetic energy.

From the calculated relaxation times one expects an approximately undamped motion for excitations below the threshold and damping on a picosecond time scale for above threshold excitations. This result is in agreement with the experimental observations[27–29] of a few resolved cycles of cyclotron radiation on a picosecond time scale.

In Fig. 2 the mean current in the x direction is shown as a function of time and detuning. We limit ourselves to the x component, for its contributions of electrons and holes add up, while in the y direction they work against each other, so that in a model with equal electron and hole masses the y component of the current would vanish. The two different regimes below and above the threshold are easily recognized. Below threshold there is little damping and so there is a strong beating of electron and hole cyclotron frequencies, but above threshold the situation is more subtle. There is not only a strong damping, but due to the optical excitation of different intersubband polarization components, which differ in sign, partial cancellation occurs. Together with the quantum-number dependence of the transverse relaxation times, the damping of the resulting current is a rather complex interplay of these two effects. One sees, e.g., a revival phenomena: At a certain time total cancellation of all contributions occurs; after the strongly damped parts have died out, only the weakly damped contributions (which are below the one-phonon threshold) survive. By these means the cancellation is lost, and therefore some current reoccurs. The reason for these weakly damped contributions that still exist even for large detunings is the lack of a selection rule in the Landau levels for the optical excitation. For large detuning, a particle (e.g., an electron) can be created high above the threshold, while its counterpart (e.g., a hole) is created in a state below threshold, or vice versa. This explains also why the damping effectively sets in already with a detuning of about one-LO-phonon energy. A similar one-phonon threshold behavior has been seen in the case of Bloch oscillations as well.[30]

All these features are more clearly seen in the Fourier representation of the current (see Fig. 3). The electron and hole cyclotron frequencies appear as separate peaks for low detuning. At about a detuning of an LO-phonon energy,

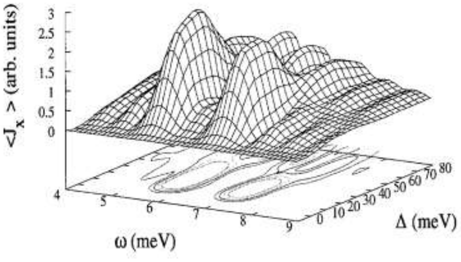

FIG. 3. Spectra of the x component of the current versus detuning Δ for fixed fields: $E=4$ kV/cm, $B=4$ T.

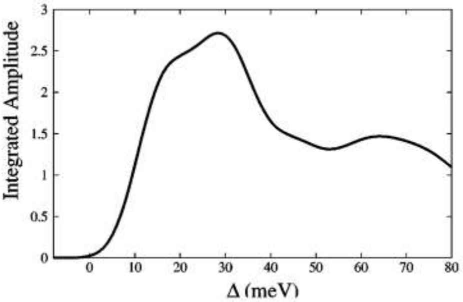

FIG. 4. Integrated square amplitude of the derivative of the current as function of the detuning Δ for fixed fields: $E=4$ kV/cm, $B=4$ T.

damping broadens the peaks, and the competition of different interband polarization contributions causes a crossover from a simple peak structure to a peak-valley-peak structure around both cyclotron frequencies. This can be understood in terms of a superposition of two differently broadened Lorentzians with different signs. Only the additional satellites have no physical meaning, but are numerical artifacts. Although the parameters of our theoretical analysis do not match those of the experiments of Ref. 28, in which only an electric depletion field has been used qualitatively the same features are observed: decay on a picosecond time scale and oscillations with the cyclotron frequencies of electrons and light holes. In Fig. 4 the integrated square amplitude of the time derivative of the current, which is proportional to the radiated signal, is shown. Again this result resembles that of the experiment.[28] A peak for small detunings of about 25–30 meV is followed by a dip. This dip is due to the strong increase of damping for excitations above the LO-phonon energy. In the experimental results, however, the minimum of the dip is reached for twice the LO-phonon energy. This has been explained in terms of the magnetophonon resonance, with the argument that due to the magnetic selection rules both electrons and holes had to be excited above threshold. As shown before, it is a crucial point of our analysis that the selection rules do not hold for $E\neq 0$. Thus for a magnetophonon resonance at $2\hbar\omega_{LO}$ one has to assume the limit of small electric fields, but then the signals would be very weak as well. Because the intrinsic depletion field in the experiment of Ref. 28 has not been under control, further experimental examinations of the field dependence are needed.

VI. CONCLUSION

The mechanism of terahertz generation in a bulk semiconductor in perpendicular electric and magnetic fields has been studied in terms of Bloch equations. We emphasized the importance of the electrical field in breaking the symmetry and inducing the polarization components between various electron or hole Landau subbands. These inter-Landau-subband polarization components are responsible for generating the oscillating current and thus the terahertz signal. Furthermore, the damping of these signals, respectively the current, is analyzed in terms of LO-phonon scattering. As a characteristic quantity, transverse quantum-number-dependent inverse relaxation times have been calculated. A smeared-out one-phonon threshold is obtained, which leads to two different damping regimes: below this threshold regime only a weak damping exists, while above the threshold regime damping on a picosecond time scale is calculated in qualitative agreement with the few available experimental data in this geometry.

ACKNOWLEDGMENTS

The preliminary experimental results of Bauer and Roskos initiated these investigations. We appreciate stimulating discussions with them. This work was supported by the DFG in the framework of the Schwerpunkt *Quantenkohärenz in Halbleitern*.

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Bose-Einstein Condensation Quantum Kinetics for a Gas of Interacting Excitons

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(Received 2 October 2000)

A quantum kinetics of the Bose-Einstein condensation in the self-consistent (s.c.) Hartree-Fock-Bogoliubov (HFB) model of the interacting Bose gas is formulated and numerically solved for the example of excitons scattering with a thermal bath of acoustic phonons. The theory describes the condensation in real time starting from a nonequilibrium initial state towards the equilibrium HFB solution. The s.c. changes of the spectrum are automatically incorporated in the scattering terms.

DOI: 10.1103/PhysRevLett.86.3839 PACS numbers: 71.35.Lk, 03.75.Fi

A nonequilibrium many-body description of the condensation of atomic or excitonic bosons starting with experimentally specified initial conditions and ending in the stationary Bose-Einstein condensation (BEC) state is an urgent problem since the experimental realizations in atomic traps [1–3] and experimental indications in semiconductors [4–7] of such a phase transition appeared. In such a phase transition, quantum coherence occurs spontaneously once critical conditions are reached. For the description of such an effect, a helpful analogy is the laser theory in which the spontaneous creation of a coherent photon amplitude in a driven nonequilibrium many-body system is treated [8]. As demonstrated for lasers, rate equations are the simplest approach to treat the self-organization of collective coherence in terms of a macroscopically populated laser mode. For a nonequilibrium boson system, semiclassical Boltzmann kinetics has indeed been shown to be able to describe a transition into a state with macroscopically populated condensate, provided the thermodynamic limit is treated with care [9]. However, a spatially homogeneous interacting boson system is known to change its spectrum from a quadratic one in the normal phase to a nonquadratic one in the condensed phase. Therefore, a semiclassical kinetics with scattering between the free-particle states cannot reach the stationary solution of the condensed state.

In recent years a quantum kinetic theory has been developed on the basis of Keldysh nonequilibrium Green functions and the equation of motion technique in which such time-dependent renormalizations of the spectra are contained [10,11]. Particularly for the description of femtosecond semiconductor spectroscopy, a quantum kinetics in terms of the single-time density matrix has been developed with non-Markovian scattering integrals. The scattering integral kernels are determined by the spectral (i.e., retarded and advanced) nonequilibrium Green functions which have to be calculated self-consistently together with the density matrix. This theory has been successful in describing fine details of recent phase sensitive femtosecond four-wave mixing experiments [10].

A first approach for the BEC kinetics in a dilute atomic Bose gas was proposed using a Fokker-Planck equation [12]. In an atomic trap Markovian rate equation approaches were used [13] to simulate numerically the occupation of the lowest levels without touching the problem of spontaneous symmetry breaking. A more recent formal treatment [14] starts from quantum kinetics and includes anomalous averages, but finally a Markovian approximation is made and the nature of the solutions is not discussed.

We apply a purely quantum kinetic approach to the nonequilibrium phase transition of a BEC of a weakly interacting Bose gas within deterministic equations. While in an atomic system the particle-particle interaction causes the irreversibility, in a low-density exciton (x) system the x-phonon scattering dominates the relaxation kinetics. Therefore we treat weakly interacting x's with x-phonon scattering as a relatively simple and thus sufficiently transparent model system for a BEC phase transition. For the discussion of possible obstructions to a BEC, e.g., in the form of nonideal boson commutator relation of x's, the polariton effect, too short lifetimes, or Auger processes we refer to the literature [15,16].

We consider x's in a spatially homogeneous system as ideal bosons with creation and annihilation operators in momentum states $a^\dagger_{\vec{k}}, a_{\vec{k}}$. The nonequilibrium x system is described approximately by the self-consistent (s.c.) time-dependent Hartree-Fock-Bogoliubov (HFB) Hamiltonian [17–19]

$$\begin{aligned} H_{\mathrm{HFB}}(t) = {} & \sum_{\vec{k}} (e_k + wn) a^\dagger_{\vec{k}} a_{\vec{k}} \\ & + \frac{1}{2} \sum_{\vec{k}} [c(t) a^\dagger_{\vec{k}} a^\dagger_{-\vec{k}} + \mathrm{H.c.}] \\ & - \sqrt{V} [w |p(t)|^2 p(t) a^\dagger_0 + \mathrm{H.c.}], \end{aligned} \qquad (1)$$

where w is the x-x contact potential, n is the total density, V is the volume, and e_k is the kinetic energy. The s.c. parameter $c(t) = \frac{w}{2}[\frac{1}{V} \sum_{\vec{q}} F_q(t) + p(t)^2]$ is given by the pair (or anomalous) function F_k and the coherent amplitude p,

$$\begin{aligned} & F_k(t) = \langle \tilde{a}_{\vec{k}}(t) \tilde{a}_{-\vec{k}}(t) \rangle, \qquad n_k(t) = \langle \tilde{a}^+_{\vec{k}}(t) \tilde{a}_{\vec{k}}(t) \rangle, \\ & p(t) = \frac{1}{\sqrt{V}} \langle a_0(t) \rangle, \end{aligned} \qquad (2)$$

with $\tilde{a}_{\vec{k}}(t) = a_{\vec{k}}(t) - \langle a_{\vec{k}}(t)\rangle$. The order parameter in an x system is also its polarization and therefore named p. The total density is the sum of the noncondensate and the condensate $n = \frac{1}{V}\sum_k n_k(t) + |p(t)|^2$. This approximation does not lead to the gapless linear spectrum of the much simpler Bogoliubov approximation. However, for a proper definition of, e.g., superfluidity, not the single particle spectrum but the density response function has to be gapless [20] and these are indeed well behaved in the HFB approximation.

We use a "spinor" notation for the boson operators $a_{\vec{k},1} = a_{\vec{k}}$ and $a_{\vec{k},2} = a^{+}_{-\vec{k}}$ with the commutator $[a_{\vec{k},i}, a_{\vec{k}',j}]_- = i\sigma^y_{ij}\delta_{\vec{k},-\vec{k}'}$, where σ^y_{ij} is a Pauli matrix. The condensate amplitude is given by $p_i = \frac{1}{\sqrt{V}}\langle a_{0,i}\rangle$, the reduced density matrix for the noncondensate by $\rho_{q,ij} = \langle \tilde{a}_{-q,i}\tilde{a}_{q,j}\rangle$, and the phonon expectation values by $N_{q,ij} = \langle b_{-q,i}b_{q,j}\rangle$.

Splitting the time development $\frac{\partial}{\partial t} = \frac{\partial}{\partial t}|_{\text{coh}} + \frac{\partial}{\partial t}|_{\text{coll}}$ into a coherent part due to the HFB Hamiltonian and a part due to the collisions with phonons we find

$$\frac{\partial}{\partial t}\, p_i(t)|_{\text{coh}} = -i\sum_j \mathcal{H}^0_{0,ij}(t)p_j(t) - \frac{i}{\sqrt{V}}\, s_i(t)\,, \tag{3}$$

$$\frac{\partial}{\partial t}\, \rho_{k,ij}|_{\text{coh}} = -i\sum_l [\mathcal{H}^0_{k,il}(t)\rho_{k,lj} + \mathcal{H}^0_{k,jl}(t)\rho_{k,il}], \tag{4}$$

where $s_i(t) = -\sqrt{V}\, w|p(t)|^2\sigma^z_{ii}p_i(t)$ is the cubic Gross-Pitaevskii term and

$$\mathcal{H}^0_{k,ij}(t) = (e_k + wn)\sigma^z_{ij} + [c(t)\sigma^+_{ij} - \text{H.c.}] \tag{5}$$

is the k-dependent HFB Hamiltonian matrix, where $\sigma^+ = (\sigma^x + i\sigma^y)/2$.

Furthermore, the x's are coupled to a thermal bath of acoustic phonons with the boson operators $b_{\vec{q}}, b^{\dagger}_{\vec{q}}$, the linear spectrum $\omega_q = c|\vec{q}|$, and the inverse temperature β through $H_{x-\text{ph}} = \frac{1}{\sqrt{V}}\sum_{\vec{k},\vec{q}} g_{\vec{q}}a^{\dagger}_{\vec{k}+\vec{q}}a_{\vec{k}}(b_{\vec{q}} + b^{\dagger}_{-\vec{q}})$, where $g_{\vec{q}} = G\sqrt{\omega_{\vec{q}}}$ is the long-wavelength deformation potential coupling. The dissipative evolution due to the phonon coupling is treated in the framework of quantum kinetics. The hierarchy of equations of motion for the boson density matrix is truncated on the second level by factorizing the averages in the equations for the phonon-assisted density matrices which contain also the HFB motion. After a formal integration of these equations, one finds the following non-Markovian quantum kinetic collision integrals:

$$\frac{\partial}{\partial t}\, p_i(t)|_{\text{coll}} = -\int_0^t dt' \sum_{\vec{k},j,l} g^2_k\mathcal{D}_{k,jj}(t,t') \times \sigma^z_{ii}\mathcal{T}_{k,il}(t,t')\mathcal{R}_{k,lj}(t')\,, \tag{6}$$

$$\begin{aligned}\frac{\partial}{\partial t}\, \rho_{k,ij}|_{\text{coll}} = &-\sum_{l,m} g^2_k\int_0^t dt'\, \mathcal{D}_{k,ll}(t,t')\,(\sigma^z_{jj}p_j(t)\mathcal{T}_{k,im}(t,t') + (i \rightleftharpoons j))\mathcal{R}_{k,ml}(t') - \sum_{\vec{q},l,m,n}\frac{g^2_q}{V}\int_0^t dt'\, \mathcal{D}_{q,ll}(t,t') \\ &\times (\sigma^z_{jj}\mathcal{T}_{k,im}(t,t')\mathcal{T}_{|\vec{k}-\vec{q}|,jn}(t,t')S_{\substack{-\vec{k},\vec{k}-\vec{q},\vec{q}\\ mnl}}(t') + \sigma^z_{ii}\mathcal{T}_{|\vec{k}+\vec{q}|,im}(t,t')\mathcal{T}_{k,jn}(t,t')S_{\substack{-\vec{k}-\vec{q},k,q\\ m,n,l}}(t'))\,.\end{aligned} \tag{7}$$

The functions $\mathcal{R}$ and S are combinations of the generalized initial and final state population factors. They are taken at the earlier time t', therefore demonstrating the quantum-kinetic memory structure of the equations, which is caused by the elimination of higher correlations.

$$\begin{aligned}\mathcal{R}_{k,ij}(t) &= \sigma^z_{ii}N_{k,\bar{j}j}(t)p_i(t) + \sigma^z_{jj}(\rho_{k,i2}(t)p_1(t) + \rho_{k,i1}(t)p_2(t))\,, \\ S_{\substack{k_1,k_2,k_3\\ i,j,l}}(t) &= N_{k_3,\bar{l}l}(\sigma^z_{jj}\rho_{k_1,ij}(t) + \sigma^z_{ii}\rho_{k_2,ij}(t)) + \sigma^z_{ll}(\rho_{k_1,i1}(t)\rho_{k_2,j2}(t) + \rho_{k_2,j1}(t)\rho_{k_1,i2}(t))\,.\end{aligned}$$

For all indices $\bar{n} \neq n$ is used. The memory kernel is determined by the phonon propagator $\mathcal{D}_k(t,t') = e^{-i\omega_k\sigma^z(t-t')}$ and the particle propagator function $\mathcal{T}_k(t,t')$. The latter describes the HFB evolution, generated by $\mathcal{H}^0_k(t)$,

$$i\,\frac{\partial\mathcal{T}_k(t,0)}{\partial t} = \mathcal{H}^0_k(t)\mathcal{T}_k(t,0); \qquad \mathcal{T}_k(0,0) = 1\,. \tag{8}$$

$\mathcal{T}_k(t,t')$ replaces the usual free propagation factors $e^{ie_k(t-t')}$ in the scattering terms by the coherent HFB dynamics without any additional adiabatic approximations. So the important s.c. temporal evolution of the renormalized particle energies is fully taken into account. For $t > t'$, the evolution matrix $\mathcal{T}_k(t,t')$ is up to a phase the retarded HFB Green function. The system of Eqs. (3),(4),(8) have to be numerically solved simultaneously.

One expects that the solution approaches the grand-canonical equilibrium averages of the equilibrium HFB, at least for weak phonon coupling. We indeed show numerically in what sense this conjecture holds. Note that these equations do not break the gauge invariance of the fundamental Hamiltonian with respect to a constant phase (particle number conservation). Therefore, if the anomalous quantities p and F_k are zero initially, they will be zero for all times. However, a finite, but very small symmetry breaking initial condensate population will blow up for supercritical conditions ($n > n_c$) but decays immediately for subcritical conditions. This behavior has been demonstrated also in the framework of rate equations before [9]. However, it is unique to our model that the condensate amplitude $|p(t)|$, the pair function $|F_k(t)|$, the total anomaly $|c(t)|$, and the noncondensate population $n_k(t)$ converge to stationary values given by the equilibrium theory of the interacting HFB gas with the proper thermodynamic quasiparticle spectrum.

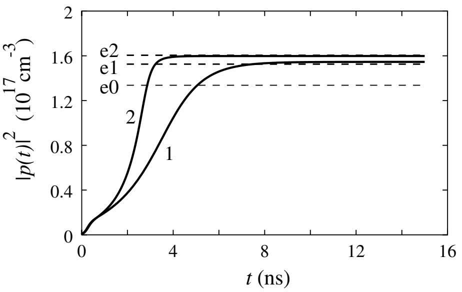

FIG. 1. Kinetics of the order parameter p. Curves 1 and 2: $|p(t)|^2$ for $w = w_1$ and for $w = w_2$, respectively. The dashed lines e1 and e2 correspond to the solution of the HFB equilibrium theory and e0 to the equilibrium of the ideal Bose gas.

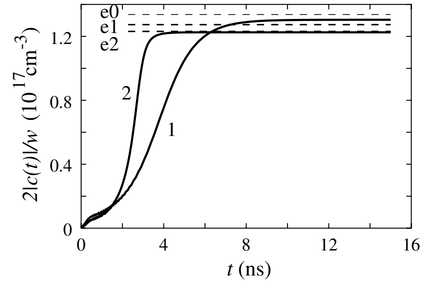

FIG. 2. Kinetics of the anomalous density $2|c(t)|/w$. Curves 1 and 2: $2|c(t)|/w$ for $w = w_1$ and $w = w_2$, respectively. The dashed lines e1 and e2 are the HFB equilibrium solutions and e0 is the equilibrium solution of the ideal Bose gas.

In order to illustrate these statements, the full set of the kinetic equations Eqs. (3),(4),(8) is solved numerically. We take the material parameters of Cu_2O, a phonon temperature $T = 1.5$ K, and a supercritical density of $n = 1.76 \times 10^{17}$ cm^{-3}, typical for related experiments [4]. In order to speed up the numerics, the coupling constant to the phonons is chosen to be twice the value reported in the literature. The interaction strength w is varied around the estimated x-x coupling.

In Cu_2O and the results for two coupling constants $w_1 = 0.5 \times 10^6$ μeV nm^3 and $w_2 = 2w_1 = 10^6$ μeV nm^3 and are shown. The scattering length corresponding to w_2 is an exciton Bohr radius. Furthermore, a small initial condensate seed of less than 1% of the density is introduced. A Gaussian initial noncondensed x distribution is assumed centered at $k_0 = 0.2$ nm^{-1} (excess energy 0.5 meV) and a width $\Delta k = 0.1$ nm^{-1}.

For the comparison of the asymptotics of the time-dependent theory with the result of the equilibrium theory, the s.c. equations for the HFB equilibrium parameters are solved numerically, too. Figure 1 shows how the order parameter $|p|$ converges in the full kinetics towards the value of the equilibrium theory for both interaction constants w_1 and w_2. The still existing differences stem from the nonvanishing energy corrections due to the interaction with phonons. For the actual x-phonon coupling G in Cu_2O they are negligibly small. An additional small phenomenological damping of the memory kernel $\mathcal{T}_k(t, t')$ stabilizes the numerics on a finite grid of k points. Surprisingly, the speed of the condensation is strongly affected by the HFB corrections in the scattering terms. Future estimates of condensation time scales should take into account the buildup of the quasiparticle spectrum. A quasiclassical Boltzmann equation with phonon scattering does not yield an accurate description of the x kinetics for typical high-density experiments in Cu_2O. In Fig. 2 the time dependence of the anomalous density [19] $2|c(t)|/w$ is shown. The anomalous density approaches the equilibrium asymptotes quite well. The resulting asymptotic distributions n_k and the pair function $|F_k|$ are compared with the equilibrium solution in Fig. 3. It can be seen that for Cu_2O parameters both results are nearly identical and far away from the trivial condensation of an ideal Bose gas, which predicts $F_k = 0$. For a truly quantitative calculation, one should take for large k values the k dependence of interaction potential into account. However, already our contact potential results demonstrate clearly the importance of interaction effects for the resulting x distributions. In particular, they show that experimentally determined distributions n_k for condensed x's in Cu_2O cannot be fitted with an ideal gas distribution with $\mu = 0$ as has been tried quite often; see, e.g., [21,22].

The derived quantum-kinetic equations bring all real quantities and absolute values to the grand-canonical equilibrium state of the interacting Bose gas. However, the equilibrium theory makes no prediction for the phase of the complex quantities. In Fig. 4 the real parts of the order parameter p and of the sum of the anomalous function $\frac{1}{V}\sum_k F_k$ are plotted versus the time. Note that both oscillate with multiples of the chemical potential

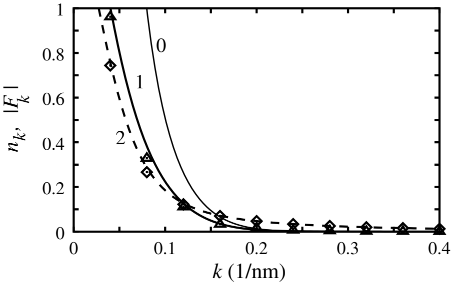

FIG. 3. Asymptotic distribution functions n_k (△) and $|F_k|$ (◇) of the kinetics and the corresponding equilibrium solutions (curves 1 and 2) for $w = w_2$. Equilibrium solution of the ideal Bose gas for n_k (curve 0) (here $F_k = 0$).

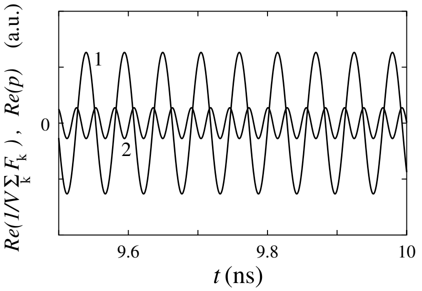

FIG. 4. Asymptotic time dependence of the real parts of the polarization p (curve 1) and of the sum of all anomalous functions $\frac{1}{V}\sum_k F_k$ (curve 2) for $w = w_2$.

$\mu = 76.531\ \mu$eV of the equilibrium theory. For the evolution under $H_{\mathrm{HFB}}(t) - \mu N$, the equilibrium values are stationary. For the real evolution the second term in the grand-canonical Hamiltonian is missing, which gives rise to oscillations for quantities which do not commute with N.

Our quantum kinetic results can be summarized by the following asymptotic statement for $\rho(t)$:

$$\rho(t) \longrightarrow \frac{1}{Z}\, e^{-i\mu N t} e^{-\beta(H_{\mathrm{HFB}}^{\mathrm{eq}} - \mu N)} e^{i\mu N t}, \qquad (9)$$

where Z is the partition function. Observables which commute with N relax to their equilibrium value, while the noncommuting ones oscillate with multiples of μ but have absolute values given by the equilibrium theory. A quasi-adiabatic approach would not be sufficient for our problem, because $H_{\mathrm{HFB}}(t)$ rotates very fast even asymptotically. Therefore, quantum kinetics with integral kernels determined by the HFB evolution is the appropriate way to treat the buildup of the quasiparticle and thermodynamic properties. Equation (9) is not an artifact of the HFB approximation but a special case of a more general statement. In Eq. (9) the essential point is that $H_{\mathrm{HFB}}^{\mathrm{eq}}$ does not commute with N. The situation is common in systems with spontaneous particle number symmetry breaking. In the Van Hove limit of weak coupling to the bath, the "rotated" density matrix $\tilde{\rho}(t) = e^{i\mu N t}\rho(t)e^{-i\mu N t}$ relaxes to a macrocanonical distribution. With infinitesimal symmetry breaking initial conditions one reaches (as in Ref. [9]) a Bogoliubov quasiaveraging macrocanonical density matrix which also does not commute with N, as in Eq. (9).

In conclusion, a quantum-kinetic theory for the BEC of an interacting Bose gas has been developed with far-ranging possible extensions also to atomic systems. It has been applied successfully to x's scattering with a thermal phonon bath. A better insight in the kinetics of a x-BEC in semiconductors has been obtained. This example also shows that quantum kinetics with its time-dependent memory kernels is the appropriate formalism to treat systems, where a quasiparticle spectrum builds up in time as in a BEC.

This work has been supported by the DFG Schwerpunktprogramm *Quantenkohärenz in Halbleitern*.

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Physica B 314 (2002) 76–80

PHYSICA B

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Virtual carrier–LO phonon interaction in the intermediate coupling region: the quantum dynamical formation of polarons

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Abstract

The relaxation of nonequilibrium heavy-hole distributions in highly polar CdTe is studied via femtosecond transmission spectroscopy and compared to more covalent GaAs. Heavy holes in CdTe show ultrafast energy redistribution via the Fröhlich mechanism even if photoexcited below the LO phonon energy. This sub-threshold dynamics is a genuine quantum kinetic effect relevant whenever the polaron self-energy is comparable to the LO phonon energy. © 2002 Elsevier Science B.V. All rights reserved.

PACS: 71.38.+i; 72.10.Di; 78.47.+p

Keywords: Quantum kinetics; Polarons; Femtosecond spectroscopy

1. Introduction

In recent years, materials such as III–V nitrides and semimagnetic II–VI semiconductors have obtained special attention for applications in optoelectronics and future spintronics [1,2]. A common feature of these materials is their relatively high degree of ionicity as compared to the well established AlGaAs/InP family. Accordingly, the polar-optical Fröhlich interaction of charge carriers with longitudinal-optical (LO) phonons is significantly stronger in the nitrides and II–VI compounds. This scattering mechanism is of central importance for the transport and optical properties of direct-gap semiconductors. The coupling strength of LO phonon scattering for a carrier with effective mass m^* may be characterized by the dimensionless polaron coupling constant

$$\alpha = \frac{e^2}{\hbar}\left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right)\left(\frac{m^*}{2\hbar\omega_{\mathrm{LO}}}\right)^{1/2},$$

which determines the ratio between the polaron self-energy and the LO phonon energy $\hbar\omega_{\mathrm{LO}}$. Electrons in the Γ-valley of GaAs represent a typical example for weak Fröhlich interaction with $\alpha \ll 1$. In contrast, charge carriers in CdTe fall into the so-called intermediate coupling regime where α is in the order of unity.

The most direct access to electron–phonon interaction is provided by ultrafast optical spectro-

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PII: S0921-4526(01)01456-9

scopy. Situations have been found, where the semiclassical Boltzmann picture with instantaneous scattering events is insufficient for the treatment of femtosecond kinetics. As an example, LO phonon quantum beats in the decay of the excitonic interband polarization [3–6] call for a quantum kinetic description. Information on the time dependence of the energy distribution of nonequilibrium carriers interacting with phonons is gained analyzing femtosecond transmission measurements [7]. In GaAs, scattering events without energy conservation and with memory effects have been observed for highly energetic electrons which emit LO phonons [8]. Quantum kinetic calculations agree with the experiment [9–11].

In this article, we demonstrate that the dynamics of nonequilibrium carriers for intermediate electron–phonon coupling exhibits qualitatively new and important features which are completely unexpected in semiclassical physics and may be understood only on the most sophisticated level of quantum kinetic theories. The most striking result is that carriers injected below the one-LO-phonon threshold, still experience significant relaxation if the Fröhlich coupling is strong enough [12].

2. The experiment

We choose the direct II–VI material CdTe as a model substance for intermediate Fröhlich interaction: electrons at the minimum of the conduction band ($m_e^* = 0.09 m_0$) exhibit a polaron coupling constant of $\alpha_e = 0.33$. Due to their larger effective mass (as in GaAs, $m_{hh}^*/m_e^* \approx 10$ in CdTe), the heavy holes are even more strongly coupled with $\alpha_{hh} \approx 1$. The band gap energy of CdTe is $E_g = 1.60\,\mathrm{eV}$ at low temperatures and $\hbar\omega_{LO} = 21\,\mathrm{meV}$. The results are directly compared to analogous investigations in GaAs where $E_g = 1.52\,\mathrm{eV}$, $m_e^* = 0.067 m_0$, $\alpha_e = 0.06$ and $\alpha_{hh} \approx 0.15$.

In the experiments, we create unbound electron–hole pairs with Gaussian light pulses of a duration of 80 fs and a central photon energy above E_g. The samples are epitaxial layers of high-purity CdTe and GaAs of a thickness of $d = 370$ and 500 nm, respectively. They are anti-reflection coated on both sides, glued to transparent substrates and mounted inside a He cryostate. In order to gain insight into the dynamics of the photoexcited carrier distributions, we measure the pump induced transmission changes with a time delayed probe pulse of a duration of 15 fs and a bandwidth of 100 meV. Perfectly synchronized pulses for excitation and probing are provided by a special two-color femtosecond Ti:sapphire laser system [13]. The test pulse is spectrally dispersed with a double monochromator (spectral resolution set to 4 meV) after transmission through the sample.

3. Theoretical simulations

In the theoretical treatment, we consider a quantum kinetic approach based on the Keldysh Green functions with two-time arguments. The photoexcited electrons and holes interact with LO phonons. Coulomb collisions between carriers are neglected in the low density regime of the experiment, but the exciton and excitonic enhancement are included through the Hartree–Fock approximation. The phonons are taken to be in equilibrium.

In a previous publication [14], we have developed a straightforward procedure to directly solve the Dyson equation for the nonequilibrium charge carriers numerically. Our solution for the intermediate coupling regime ($0.1 < \alpha < 1$) predicted important deviations from the earlier one-time approximations.

Numerical simulations are performed for the material parameters of CdTe and GaAs taking into account time delayed optical probe and pump pulses of the experiment.

4. Results and discussion

Differential transmission spectra for various delay times t_D are measured at a photoexcited electron–hole density of $4 \times 10^{14}\,\mathrm{cm}^{-3}$ with cross-linearly polarized pump and probe beams in GaAs (left column of Fig. 1) and CdTe (right column of Fig. 1) [12]. The excitation density is maintained

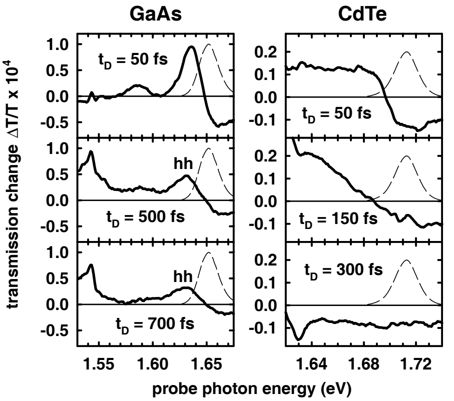

Fig. 1. Spectrally resolved transmission changes $\Delta T/T$ in GaAs (left column) and CdTe (right column) for various delay times t_D at a carrier density of $4 \times 10^{14}\,\text{cm}^{-3}$ and $T_L = 4.5\,\text{K}$. The excitation spectra are shown as dashed lines.

extremely low to suppress carrier–carrier scattering [15]. A lattice temperature of 4.5 K ensures very slow scattering of carriers with acoustic phonons on a time scale of 10 ps. Consequently, the polar-optical interaction with LO phonons is by far the dominant relaxation mechanism in the sub-picosecond regime.

Exciting GaAs with an 80 fs pulse at 1.65 eV (see dashed line) results in a heavy-hole (hh) distribution centered at a kinetic energy of 18 meV which is smaller than $\hbar\omega_{LO}$, i.e. no scattering with LO phonons is expected. In contrast, the electrons are created with an excess energy of 112 meV and allowed to transfer energy to the crystal lattice via rapid emission of LO phonons [7,8]. At $t_D = 50\,\text{fs}$ in GaAs, a signature of the nonthermal carrier distribution appears near the excitation energy. For a delay of $t_D = 500\,\text{fs}$, i.e. after twice the electron–LO phonon emission time of approximately 240 fs in GaAs [7,15], most of the electrons have relaxed towards the minimum of the Γ-valley, inducing a transmission increase below a probe photon energy of 1.55 eV. A well resolved bleaching peak due to the generated heavy holes remains at 1.63 eV (indicated by hh in Fig. 1). As late as 700 fs after excitation, the increased transmission associated with the hh distribution is still clearly visible in GaAs.

In strong contrast to GaAs, no analogous signature of a hh distribution is found in CdTe (right column of Fig. 1): excitation with an 80 fs pulse centered at 1.71 eV (see dashed line) generates heavy holes with an average kinetic energy of 12 meV. In the semiclassical picture of carrier relaxation, the hh distribution should, therefore, behave similarly as in GaAs. However, at a delay time of $t_D = 150\,\text{fs}$, approximately twice the electron–LO phonon emission time of 70 fs in CdTe [15], no bleaching peak is observed close to the excitation energy. Apparently, the distribution of heavy holes in CdTe relaxes on a time scale comparable to the electrons even though real emission of LO phonons should be energetically impossible. This surprisingly fast dynamics can only be related to the increased polaron coupling in CdTe since all other parameters are very similar in GaAs.

To elucidate the physical origin of the missing bleaching peak in more polar CdTe, we have performed extensive theoretical studies [12]. The hh energy distributions computed with the two-time quantum kinetics (thick lines) and the semiclassical Boltzmann kinetics (thin lines) are depicted in Fig. 2 for GaAs (left column) and CdTe (right column). In GaAs, both models result in rather similar hh populations. A strongly peaked distribution is conserved on a sub-picosecond time scale. In contrast, the populations obtained with the Dyson equation for CdTe show a significant femtosecond relaxation of the holes. A very broad continuous background resembling a quasi-thermal distribution has formed already after a delay time t_D as short as 60 fs. Even at a delay time of $t_D = 200\,\text{fs}$, the background component experiences further relaxation indicating that the phenomenon is not linked exclusively to the energy uncertainty during the ultrafast carrier generation process. This effect is purely quantum kinetic in nature: if the sub-threshold hole dynamics in CdTe is simulated semiclassically (thin lines in Fig. 2), the distribution functions undergo no relaxation. Upon closer inspection the background component is discernible also in the quantum kinetic simulation of GaAs, but the

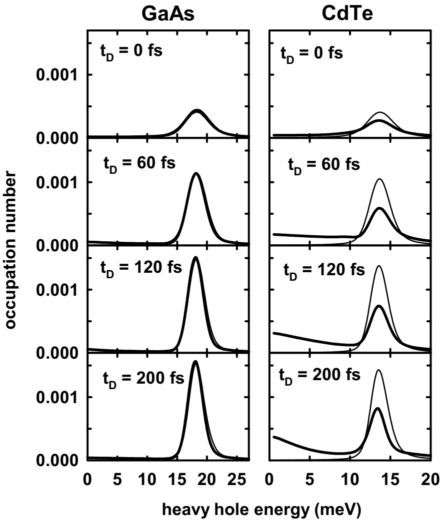

Fig. 2. Heavy-hole energy distributions in GaAs (left column) and CdTe (right column), as calculated in the two-time quantum kinetic simulation based on the Dyson equation (thick lines) and with the semiconductor Bloch equations including Boltzmann scattering terms (thin lines) for various delay times t_D.

effects are much smaller than in CdTe. In the experimental result for CdTe, the relaxation of the heavy holes is even more pronounced than expected from the calculated hole distribution. This finding suggests an additional broadening of the bleaching signal due to valence band warping that is not included in the simulation.

To understand the unexpected dynamics of the heavy holes, one has to take into account the fact that within the two-time quantum kinetics with a stronger coupling constant the energy of the free particles is no longer conserved. The interaction energy plays an important role and allows transitions that are forbidden in the Boltzmann picture.

The dynamics of the hole distribution is a result of a dynamical polaron formation: the laser pulse creates bare electrons and holes. These quasiparticles get dressed via deformation of the surrounding polar crystal lattice. The buildup of this virtual LO phonon wave packet leads to a new class of carrier dynamics. Such phenomena are relevant whenever the polaron renormalization energy is comparable to the LO phonon energy. The approximate time scale for the polaron formation is linked to the duration of the LO phonon oscillation period which is $\omega_{LO}^{-1} = 200\,\mathrm{fs}$ for the case of CdTe.

5. Conclusion

In conclusion, we have found an ultrafast dynamics of low-energy heavy holes interacting with unoccupied polar-optical modes in CdTe. A theoretical description of the sub-threshold scattering calls for a sophisticated quantum kinetic treatment beyond the Kadanoff–Baym ansatz. This phenomenon represents a typical many-body effect: the free-particle energy ceases to be a constant of motion in systems where the coupling between electronic and lattice degrees of freedom can no longer be regarded as a weak perturbation.

Acknowledgements

We wish to thank K. Ortner and C.R. Becker for growing the high-quality CdTe sample. The high-purity GaAs specimen has been provided by G. Böhm.

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Real-Time Bose-Einstein Condensation in a Finite Volume with a Discrete Spectrum

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(Received 11 February 2002; published 13 May 2002)

We show that Bose condensation in real time occurs in a finite system not only as an accumulation of the bosons in the ground state below a critical temperature, but also as a rapid enhancement of an arbitrary small symmetry breaking, followed by a very slow decay of the symmetry breaking order parameter from the almost ideal value to the vanishing equilibrium value. We show this analytically on an exactly soluble model and numerically on a model of noninteracting bosons in an oscillator potential.

DOI: 10.1103/PhysRevLett.88.210404 PACS numbers: 03.75.Fi, 05.20.Dd

Extraordinary experimental achievements have recently proven Bose-Einstein condensation (BEC) of atoms in magnetic traps [1,2]. While the original idea about BEC was formulated only in the thermodynamic limit of a free Bose gas, one may easily conceive that, with a finite number of atoms in a finite volume, below a certain temperature the number of bosons in the ground state will increase proportionally to their total number (or volume). *A more unexpected aspect for a theorist is the observation of the order parameter (interference) implying a spontaneous symmetry breaking (of phase transformations corresponding to the particle number conservation) in a finite system.* We show in this paper, by a study of the evolution in real time, that even with a finite number of bosons in a finite volume below a "critical temperature" one gets, besides the accumulation of the bosons in the ground state, also a strong and rapid enhancement of the order parameter, which later decays very slowly. The more particles one has, the slower this decay will be, and of course in the thermodynamic limit it will not decay at all. In other words, what is forbidden in equilibrium in a finite volume (a nonzero order parameter) is allowed and therefore experimentally accessible in the kinetics, for a certain time interval which grows with the number of particles.

The treatment is based on the Markovian equations for the particle occupation numbers and the order parameter as they may be derived through the equation of motion techniques for a system of noninteracting bosons in contact with a thermostat. A treatment of interacting bosons already would require going beyond the Markov approximation [3]. The Markovian equations were derived and used in earlier publications [4,5] in the context of Bose condensation, however, without calculation of the order parameter in finite systems. For completeness these equations are first discussed. Then, an exactly soluble model is discussed [4], to prove analytically our statements about BEC. On the other hand, in order to be closer to experiments, we consider also bosons in an oscillator potential. We make the assumption of very simple degenerate transition rates due to a thermostat and get numerical solutions. The behavior obtained in this numerical study is in perfect agreement with the conclusions of the soluble model. Already with 100 bosons one gets, at ($\frac{k_B T}{\hbar\omega_0} = 0.5$), very rapidly all the atoms in the ground state, and the order parameter almost achieves its ideal thermodynamic value before its very slow asymptotic decay.

Let us consider the interaction between bosons and a thermal bath (phonons) described by the Hamiltonian $H = \sum_\alpha e_\alpha a_\alpha^+ a_\alpha + \sum_q \hbar\omega_q b_q^+ b_q + \sum_{\alpha,\alpha',q}(g_{q,\alpha\alpha'} a_\alpha^+ a_{\alpha'} b_q + \text{H.c.})$. Starting from the Heisenberg equations for the creation and annihilation operators of the bosons a_α^+, a_α, taking averages with the bath being in thermal equilibrium, decoupling the higher correlations and taking the long-time Markovian limit, one gets the equations describing the time evolution of the average boson occupation numbers $\langle a_\alpha^+ a_\alpha\rangle$ and of the (square modulus) of the average ground state annihilation operator $\langle a_0\rangle$ (anomalous average related to the order parameter):

$$\frac{\partial}{\partial t}\langle a_\alpha^+ a_\alpha\rangle_t = \sum_{\alpha'}\{W_{\alpha'\alpha}\langle a_{\alpha'}^+ a_{\alpha'}\rangle_t(1 + \langle a_\alpha^+ a_\alpha\rangle_t) - (\alpha \rightleftharpoons \alpha')\},$$

$$\frac{\partial}{\partial t}|\langle a_0\rangle_t|^2 = |\langle a_0\rangle_t|^2 \sum_{\alpha\neq 0}[\langle a_\alpha^+ a_\alpha\rangle_t W_{\alpha 0} - (1 + \langle a_\alpha^+ a_\alpha\rangle_t)W_{0\alpha}].$$

The transition rates due to the interaction with the thermal bath $W_{\alpha,\alpha'}$ satisfy the detailed balance relation $W_{\alpha\alpha'} = W_{\alpha'\alpha} e^{\beta(e_\alpha - e_{\alpha'})}$. The explicit expression of the transition rates is given by the golden rule, but it is not relevant for our further discussion.

It is important to remark that the equation for $|\langle a_0\rangle_t|^2$ resembles very much the equation of $\langle a_0^+ a_0\rangle_t$, but they are not identical and, as we shall see, this fact has far reaching consequences. The stationary solution of these equations is the Bose distribution for the occupation numbers and a vanishing anomalous average. However, if one starts with an arbitrarily small (but extensive) symmetry breaking initial condition $\langle a_0^+ a_0\rangle_{t=0} \neq 0$, one may show [4] that, below a critical temperature, in the thermodynamic limit the symmetry breaking survives as a spontaneous symmetry breaking as it is given by Bogolyubov's quasiaverages theory.

The equation for the phase ϕ of $\langle a_0\rangle$, however, is not governed by the transition rates,

$$\frac{\partial}{\partial t}\phi(t) = -e_0 - \sum_{q,\alpha\neq 0} |g_{q,\alpha 0}|^2 \times \left\{ \frac{1}{e_\alpha - e_0 - \hbar\omega_q}\left[\frac{\langle a_\alpha^+ a_\alpha\rangle}{e^{-\beta\hbar\omega_q}-1} - \frac{\langle a_\alpha^+ a_\alpha\rangle + 1}{e^{\beta\hbar\omega_q}-1}\right] + P\,\frac{1}{e_\alpha - e_0 + \hbar\omega_q}\left[\frac{a_\alpha^+ a_\alpha\rangle}{e^{\beta\hbar\omega_q}-1} - \frac{\langle a_\alpha^+ a_\alpha\rangle + 1}{e^{-\beta\hbar\omega_q}-1}\right]\right\}.$$

The phase behaves asymptotically as $\phi(t) \to -(e_0 + \Delta e_0)t$, where Δe_0 is the lowest order correction to the ground state energy due to the interaction with the thermostat. A derivation of these equations is given in Ref. [4] for free massive bosons interacting with acoustical phonons in thermal equilibrium, but it does not depend on these specific details. In what follows, we shall discuss only the first two equations considering different models for the energies and transition rates.

All the essential features of the BEC are contained also in an exactly soluble boson model consisting of a ground state (particle energy ϵ_0) and an excited state (particle energy ϵ_1). The lowest state is taken nondegenerate, while the higher state is taken to be macroscopically degenerate (i.e., the degeneracy is Vn_1, proportional to the volume V). It simulates a continuum. The transition rate between the two states is $W_{10} = \frac{1}{V}we^{-\beta\epsilon_0}$, respectively $W_{01} = \frac{1}{V}we^{-\beta\epsilon_1}$, and obeys the detailed balance. This volume dependence is typical for transition rates between a bound state and a state in the part of the spectrum; that in the infinite volume limit goes to a continuous one [5]. Within this model, the real-time evolution can be studied analytically.

We are looking for degenerate solutions having identical occupation of the degenerate states. The rate equations for $f_\alpha(t) \equiv \langle a_\alpha^+ a_\alpha\rangle_t$; $(\alpha = 0, 1)$ are then

$$\frac{\partial}{\partial t} f_1(t) = \frac{w}{V}\{f_0(t)[1 + f_1(t)]e^{-\beta\epsilon_1} - f_1(t)[1 + f_0(t)]e^{-\beta\epsilon_0}\},$$

$$\frac{\partial}{\partial t} f_0(t) = n_1 w\{f_1(t)[1 + f_0(t)]e^{-\beta\epsilon_0} - f_0(t)[1 + f_1(t)]e^{-\beta\epsilon_1}\},$$

and the equation for the square modulus of the order parameter $p(t) \equiv (1/\sqrt{V})\langle a_0\rangle_t$ is

$$\frac{\partial}{\partial t}|p(t)|^2 = |p(t)|^2 n_1 w\{f_1(t)e^{-\beta\epsilon_0} - [1 + f_1(t)]e^{-\beta\epsilon_1}\}.$$

These equations conserve the average particle density $n_{\text{tot}} \equiv n_1 f_1 + \frac{1}{V} f_0$. Since the chemical potential in equilibrium has an upper bound, $\mu \leq \epsilon_0$, the particle occupation in the state 1 is bounded by $1/(\epsilon^{\beta(\epsilon_0-\epsilon_1)-1})$. Therefore, above a critical density at fixed temperature (or below a critical temperature at fixed n_{tot}), the equilibrium occupation of the state (0) has to increase with the volume. The critical density is given by $n_c \equiv n_1/(e^{\beta(\epsilon_1-\epsilon_0)} - 1)$. These equations are exactly soluble for any set of parameters. For the sake of simplicity, we take $\epsilon_0 = 0$, $w = 1$ (which amounts to choosing the time unit $\frac{1}{w}$), and denote $e^{-\beta(\epsilon_1-\epsilon_0)} \equiv \xi < 1$. Then $n_c \equiv n_1\frac{\xi}{1-\xi}$. We eliminate f_1 in favor of f_0 through the conservation equation and get a closed equation for f_0:

$$\frac{\partial}{\partial t} f_0(t) = -\frac{1-\xi}{V} f_0(t)^2 + f_0(t)(1-\xi)\left(n_{\text{tot}} - n_c - \frac{1}{V(1-\xi)}\right) + n_{\text{tot}}\,.$$

The discriminant of the polynomial in f_0 on the right side is positive, the roots (x_1, x_2) are real with opposite signs, and we choose x_1 as the positive one. Then the solution of the differential equation [with $f_0(0) \geq 0$] is

$$f_0(t) = \frac{x_1 + x_2\frac{x_1 - f_0(0)}{f_0(0)-x_2}e^{-a(x_1-x_2)t}}{1 + \frac{x_1 - f_0(0)}{f_0(0)-x_2}e^{-a(x_1-x_2)t}},$$

with $a \equiv \frac{1-\xi}{V}$. Now, as it is easy to see, the roots behave for large volumes as $V(n_{\text{tot}} - n_c) + n_c/[(1-\xi)(n_{\text{tot}} - n_c)]$ and $-n_c/[(1-\xi)(n_{\text{tot}} - n_c)]$, respectively. Then since $\lim_{t\to\infty} f_0(t) = x_1 \geq 0$, one gets the following for $n_0(t) \equiv \frac{1}{V} f_0(t)$:

$$\lim_{V\to\infty}\lim_{t\to\infty} n_0(t) = \begin{cases} 0 & \text{for } n_{\text{tot}} < n_c \\ n_{\text{tot}} - n_c & \text{for } n_{\text{tot}} \geq n_c. \end{cases}$$

Thus, BEC for the population occurs above the critical density. By taking the opposite order of the limits, one has to be careful regarding the initial condition. Let us now consider the infinite volume limit of the condensate particle density. For $n_{\text{tot}} < n_c$ (subcritical density),

$$n_0(t) = \frac{n_0(0)(n_c - n_{\text{tot}})e^{-(1-\xi)(n_c - n_{\text{tot}})t}}{n_c - n_{\text{tot}} + n_0(0)(1 - e^{-(1-\xi)(n_c-n_{\text{tot}})t})},$$

for $n_{\text{tot}} > n_c$ (supercritical density),

$$n_0(t) = \frac{n_0(0)(n_{\text{tot}} - n_c)}{n_0(0) + [n_{\text{tot}} - n_c - n_0(0)]e^{-(1-\xi)(n_{\text{tot}}-n_c)t}},$$

and for $n_{\text{tot}} = n_c$ one gets a "critical slowing down," i.e., powers of t instead of exponentials:

$$n_0(t) = \frac{n_0(0)}{1 + n_0(0)(1-\xi)t}.$$

Therefore, if no condensate is present at the initial time $[(n_0(0) = 0]$, then there is no evolution of the condensate at all, while for any finite initial condensate $[(n_0(0) > 0]$ one gets condensation at $t \to \infty$ above the critical density, while the condensate disappears below the critical density. One gets the square modulus of the order parameter by simple integration using the previous solution:

$$\frac{|p(t)|^2}{|p(0)|^2} = \frac{1 + \frac{x_1 - f_0(0)}{f_0(0)-x_2}}{1 + \frac{x_1 - f_0(0)}{f_0(0)-x_2}e^{-|a(x_1-x_2)|t}}\,e^{-(1-\xi)(x_1/V - n_{\text{tot}} + n_c)t}.$$

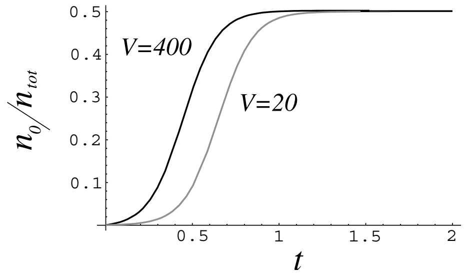

FIG. 1. Evolution of the average occupation of the lowest level in the soluble model for the volumes $V = 20$ and 400.

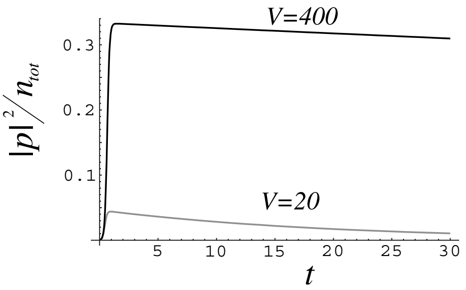

FIG. 2. Evolution of the square of the order parameter in the soluble model for the volumes $V = 20$ and 400.

The last exponential behaves for $V \to \infty$ as $e^{-(1/V)[n_c/n_{\rm tot}-n_c]t}$ in the supercritical regime, and as $e^{-(1-\xi)(n_c-n_{tot})t}$ in the subcritical one. It is clear that one encounters here two time scales: a fast or "microscopic" one, i.e., weakly affected by the volume, and a slow or "macroscopic" one, describing evolutions which get slower as the volume increases. The relaxation of the population to the equilibrium BE distribution is in this sense fast, as is the decay of the order parameter to zero under subcritical conditions. On the contrary, in the supercritical regime the order parameter decays slowly. A further consequence of this is the observation that the time constant of this slow decay can be readily obtained directly from the equation of $|p(t)|^2$. Indeed, the populations are quickly thermalized and therefore one may replace $f_1(t)$ with $f_1(\infty)$. Using the properties of the BE functions, together with the detailed balance, one gets the following for large volumes:

$$\frac{1}{|p(t)|^2}\frac{\partial}{\partial t}|p(t)|^2 \sim n_c(1 - e^{-\beta\mu}) = -\frac{1}{V}\frac{n_c}{n_{\rm tot} - n_c},$$

in agreement with the discussion above.

It is clear that for any large but finite volume at $t \to \infty$ the order parameter disappears. However, if the infinite volume limit is performed first, then the order parameter reaches the stationary value,

$$\frac{|p(\infty)|^2}{|p(0)|^2} = \frac{n_0(\infty)}{n_0(0)},$$

and the Bose condensation is perfect also in the sense of the order parameter if it was different from 0 at $t = 0$. If in the initial state one had $|p(0)|^2 = n_0(0)$, then also in the final state $|p(\infty)|^2 = n_0(\infty)$. In a finite but sufficiently big volume, under supercritical conditions, the order parameter will be first enhanced from any small initial value and then disappears very slowly. Figures 1 and 2 illustrate the time evolution of n_0 and $\frac{|p(t)|^2}{|p(0)|^2}$ above the critical density for finite volumes $V = 20$ and 400. [We took here $n_{\rm tot} = 20; n_c = 10; n_0(0) = 0.01; \xi = 0.001$.]

The features shown above are actually rather general and not pertinent only to this soluble model. The same kind of scenario for the order parameter can be shown through numerical simulation to occur also in a more complicated model of noninteracting bosons in a finite volume in contact with a thermal bath of acoustical phonons, but we give here no numerical details of it. Instead we shall analyze a quite different model with discrete spectrum, where no useful concept of a volume may be defined, but the spectrum is discrete.

We consider a three-dimensional isotropic oscillator whose states may be characterized by three integers $n_1, n_1, n_3 = 0, 1, 2, \ldots,$ but its spectrum is determined only by $n = n_1 + n_2 + n_3$, $\epsilon_n = \hbar\omega_0(n + \frac{3}{2})$; $n = 0, 1, \ldots$ and therefore each energy level has the degeneracy $\frac{1}{2}(n + 1)(n + 2)$.

This model has an appropriate thermodynamic limit (see Ref. [6]) by letting the distance between the levels go to zero ($\hbar\omega_0 \to 0$) and at the same time the average number of particles $\langle N\rangle$ go to infinity in such a way that $\nu \equiv \langle N\rangle(\hbar\omega_0)^3$ remains constant. In this limit, Bose condensation with a critical temperature defined by $k_BT_c = \hbar\omega_0(\frac{\langle N\rangle}{\zeta(3)})^{1/3}$ occurs, with $\zeta(3) = 1.202\,06$ the Riemann $\zeta(n)$ function for $n = 3$.

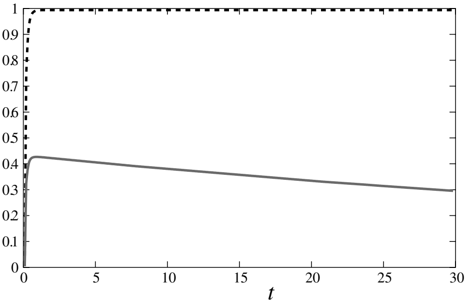

FIG. 3. Time evolution of the relative occupation of the lowest level $|\langle a_0\rangle|^2/\langle N\rangle$ (dashed line) and the square of the order parameter $|P|^2$ (solid line) in the oscillator model. ($\langle N\rangle = 100$, $\frac{k_BT}{\hbar\omega_0} = 0.5$, $|\langle a_0\rangle|^2_{t=0} = \langle a_0^+ a_0\rangle_{t=0} = 1$.)

We will treat now the kinetics of a finite average number $\langle N\rangle$ of bosons in the oscillator potential with a finite distance ($\hbar\xi_0$) between the levels. We consider very simple transition rates depending only on the energies of the initial and the final states: $W_{n_1,n_2,n_3;n_{1'},n_{2'},n_{3'}} = W_{n,n'}$, and we look only for degenerate solutions $\langle a^+_{n_1,n_2,n_3} a_{n_1,n_2,n_3}\rangle \equiv f_{n_1,n_2,n_3} = f_n(n = n_1 + n_2 + n_3)$. Therefore the equations for the populations f_n and the square modulus of the "order parameter" $P \equiv \langle a_{0,0,0}\rangle/\sqrt{\langle N\rangle}$ are

$$\frac{\partial}{\partial t} f_n = \sum_{n'=0}^{\infty} \frac{1}{2}(n' + 1)(n' + 2)\{W_{n'n} f_{n'}(1 + f_n) - W_{nn'} f_n(1 + f_{n'})\}; \qquad (n = 0, 1, 2, \ldots),$$

$$\frac{\partial}{\partial t}|P|^2 = |P|^2 \sum_{n=1}^{\infty} \frac{1}{2}(n + 1)(n + 2)\{W_{n0} f_n - W_{0n}(1 + f_n)\}.$$

We suppose again that the transition rates satisfy the detailed balance relation: $W_{nn'} = W_{n'n} e^{\beta\hbar\omega_0(n-n')}$. These equations conserve the total number of bosons: $\sum_{n=0}^{\infty} \frac{1}{2}(n + 1)(n + 2) f_n = \langle N\rangle$ and the stationary (equilibrium) solution is the Bose distribution for the populations and a vanishing order parameter: $f_n^0 = 1/(e^{\beta[\hbar\omega_0(n+3/2)-\mu]} - 1)$; $P^0 = 0$. We shall illustrate the kinetics of evolution to equilibrium for finite numbers of bosons below the formerly discussed critical temperature by numerical calculations with transition rates only to nearest states $W_{nn'} = w(e^{(1/2)\beta\hbar\omega_0}\delta_{n,n'+1} + e^{-(1/2)\beta\hbar\omega_0}\delta_{n',n+1})$.

For the numerics, we choose the temperature as $\beta\hbar\omega_0 = 2.0$ far below the "critical" one. (Of course, in the discrete spectrum one has no true critical temperature, and we borrow the value given above in the specific thermodynamic limit.) We choose initial conditions satisfying $|\langle a_0\rangle|^2_{t=0} = \langle a_0^+ a_0\rangle$ at ($t = 0$) and put very few particles in the lowest state as a "seed" for $|P|^2$ and the rest in a high lying excited state (20th level).

Surprisingly enough (see Fig. 3), one gets practically the complete enhancement scenario of the order parameter as described before within the soluble model at a finite volume already at $\langle N\rangle = 100$. The dotted curve in Fig. 3 shows the population condensation ($\langle a_0^+ a_0\rangle/\langle N\rangle$), and one may see that the square of the order parameter reaches very rapidly almost half of the ideal value and thereafter slowly decays. The maximally enhanced value of the order parameter is still strongly dependent on the initial condition. For an initial condition with an initial condensate of 0.1 instead of 1, one gets a 4 times smaller order parameter.

The slow decay of the order parameter after its enhancement may be understood along the same lines as in the soluble model. By introducing in the right-hand side of the equation for the order parameter the equilibrium population given by the Bose function, and using again the detailed balance, one gets a decay constant proportional to $\frac{3}{2}\hbar\omega_0 - \mu$. As $\langle N\rangle \rightarrow \infty$, $\mu \rightarrow \frac{3}{2}\hbar\omega_0$ and the decay constant goes to zero.

Indeed, as is shown in Fig. 4, where the time evolution of the order parameter is shown on a longer time scale for $\langle N\rangle = 100$ and $\langle N\rangle = 1000$ with the same initial condition as before, the decay slope changes drastically with the increase of the average number of bosons. In the same time, the maximal value of the order parameter increases slowly with $\langle N\rangle$. On the same figure one may see also explicitly the exponential character of the decay (at least for $\langle N\rangle = 100$).

In conclusion, we have shown, in a Markovian approach within a soluble model, how in a finite system the full scenario of the BEC develops in real time with a strong enhancement of any small initial symmetry breaking. However, asymptotically the symmetry breaking order parameter very slowly disappears. We obtained an analogous numerical result also for bosons in an oscillator potential with as few as 100 atoms. The more atoms one has, the more slowly the order parameter disappears.

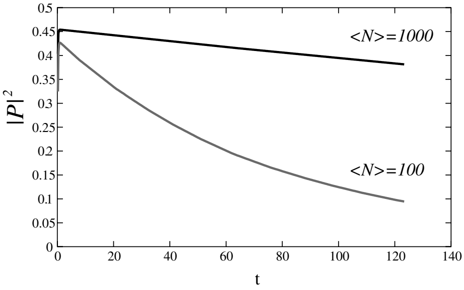

FIG. 4. Long-time behavior of the order parameter $|P|^2$ for $\langle N\rangle = 100$ and $\langle N\rangle = 1000$. The rest of the parameters are as in Fig. 3.

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About the c-Number Approximation of the Macroscopical Boson Degrees of Freedom within a Solvable Model

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(Received July 17, 2002; accepted August 1, 2002)

PACS: 71.35.Lk

Dedicated to Professor Dr. Roland Zimmermann on the occasion of his 60th birthday

We show on a solvable model in equilibrium and non-equilibrium, that in the thermodynamic limit the c-number approximation of the macroscopically occupied lowest state is exact for all averages of products of these operators.

Introduction One of the basic ideas of the theory of condensation of weakly interacting bosons is the replacement of the creation and annihilation operators of the condensate states by their averages (c-numbers) in the many-body Hamilton operator. The argumentation goes over the macroscopic occupation of these lowest states. This step has allowed the developement of a specific diagramm technique [1]. Although very successful, this theory is mathematically still not very well understood (see Ref. [2]). Among others, it is not clear whether this is a self-consistent approximation, or in some limiting sense equivalent to the exact theory. In this paper we describe a simple solvable model, in which the infinite volume limit of the averages of the exact theory coincides with the predictions of the c-number version. We describe first a non-equilibrium version in which the symmetry breaking and the macroscopic occupation result from the coupling to an external field. This model emerges from solid state theoretical models for excitons interacting with optical fields. Then we describe the equilibrium condensation, in which the symmetry breaking in the Hamiltonian remains infinitesimal and one gets spontaneous symmetry breaking in the sense of Bogolyubov's quasi-averages.

Non-Equilibrium Model in an External Field Let us consider the time-dependent Hamiltonian for a single boson mode interacting with itself and with an external field $\mathcal{E}(t)$

$$H(t) = \epsilon a^{+} a + \frac{w}{V}\, a^{+2} a^{2} + \sqrt{V}\, \mathcal{E}(t)\, (a + a^{+})\,. \tag{1}$$

Here V is the volume and the creation and annihilation operators satisfy

$$[a, a^{+}] = 1\,. \tag{2}$$

The field term is typical for bosonic models of excitons in a semiconductor interacting with an optical field. We may omit the one-particle energy term $\epsilon a^{+} a$, since it can be eliminated from the Heisenberg equations by a simple phase transformation of the operators $a \to a\, \mathrm{e}^{-\imath\epsilon t}$.

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We shall make use of the commutation relations

$$\begin{aligned}
&\left[a^{+n}a^{m},\, a^{+2}a^{2}\right] = 2(m-n)\, a^{+n+1}a^{m+1} + (m(m-1) - n(n-1)\, a^{+n}a^{m}\,,\\
&[a^{+n}a^{m},\, a] = -na^{+n-1}a^{m}\,,\\
&[a^{+n}a^{m},\, a^{+}] = ma^{+n}a^{m-1}
\end{aligned}$$

to obtain the Heisenberg equation of motion for the operators $B_{n,m}$ defined as

$$B_{n,m} \equiv a^{+n}a^{m} \quad (n,\, m = 0, 1, \ldots)\,, \tag{3}$$

and we get

$$\begin{aligned}
\imath\hbar\, \frac{\partial}{\partial t}\, B_{n,m} = {} & \frac{w}{V}\, (2(m-n)\, B_{n+1,m+1} + (m(m-1) - (n-1))\, B_{n,m})\\
& + \sqrt{V}\, \mathcal{E}(t)\, (mB_{n,m-1} - nB_{n-1,m})\,.
\end{aligned}$$

Obviously the undefined symbols with $n = -1$ or $m = -1$ have vanishing coefficients and therefore do not appear.

Now let us define the infinite volume limits (in the thermodynamical sense) of the averages

$$b_{n,m} \equiv \lim_{V\to\infty} \frac{1}{V^{\frac{n+m}{2}}}\, \langle B_{n,m}\rangle < \infty\,. \tag{4}$$

We anticipated here, that the so defined objects have a finite thermodynamic limit.

For these limiting objects one gets

$$\imath\hbar\, \frac{\partial}{\partial t}\, b_{n,m} = 2w(m-n)\, b_{n+1,m+1} + \mathcal{E}(t)\, (mb_{n,m-1} - nb_{n-1,m})\,. \tag{5}$$

The obvious solution is

$$b_{n,m} = b_{1,0}^{n} b_{0,1}^{m}\,, \tag{6}$$

with

$$\imath\hbar\, \frac{\partial}{\partial t}\, b_{0,1} = 2wb_{1,0}b_{0,1}^{2} + \mathcal{E}(t)\,; \qquad b_{1,0} = b_{0,1}^{*}\,. \tag{7}$$

This shows, that in the thermodynamic limit the weighted averages all factorize in products of averages of $\frac{1}{\sqrt{V}}\, \langle a\rangle)$ and its conjugate.

Equilibrium Model for Bose Condensation Now let us consider the Hamiltonian

$$H = \frac{w}{V}\, a_0^{+2}a_0^{2} + \sum_{k\neq 0} \frac{\hbar^2 k^2}{2m}\, a_k^{+}a_k + \sqrt{V}\, \eta(a_0 + a_0^{+}) \tag{8}$$

describing bosons of wave-vectors k. The $k = 0$ mode interacts with itself and an infinitesimal symmetry breaking parameter η was introduced, according to the definition of the quasi-averages of Bogolyubov and it will vanish after performing the infinite volume limit (in the thermodynamic sense) of the averages over the macro-canonical density matrix

$$\langle \ldots \rangle \equiv \mathrm{Tr}\left(\frac{1}{Z}\, \mathrm{e}^{-\beta(H-\mu N)} \ldots\right). \tag{9}$$

The degrees $k = 0$ and $k \neq 0$ are coupled only through the average particle number

$$\frac{1}{V} \sum_k \langle a_k^+ a_k \rangle = n \,. \tag{10}$$

Let us define as usual for any operator A

$$A(\tau) \equiv \mathrm{e}^{\tau(H-\mu N)} \, A \, \mathrm{e}^{-\tau(H-\mu N)}; \quad (\tau < \beta) \,. \tag{11}$$

Of course $\frac{\mathrm{d}}{\mathrm{d}\tau} \langle A(\tau) \rangle = 0$, however, we get along the same scheme the factorization of the averages at $V \to \infty$,

$$\lim_{V\to\infty} \frac{1}{V^{\frac{n+m}{2}}} \langle a_0^{+n} a_0^m \rangle = \left(\lim_{V\to\infty} \frac{1}{\sqrt{V}} \langle a_0 \rangle \right)^{*n} \left(\lim_{V\to\infty} \frac{1}{\sqrt{V}} \langle a_0 \rangle \right)^m \,. \tag{12}$$

With the notation $P \equiv \lim_{V\to\infty} \frac{1}{\sqrt{V}} \langle a_0 \rangle$ we obtain the relation:

$$-\mu P + 2w \, |P|^2 \, P + \eta = 0 \,. \tag{13}$$

Now one can go over to the limit $\eta \to 0$ and get besides the "normal" solution $P = 0$ also the symmetry-breaking one

$$\mu = 2w \, |P|^2 \,. \tag{14}$$

Above the critical temperature of the free Bose gas one gets the normal solution, while below it one gets the symmetry breaking one. In both cases we recovered the results of the c-number approximation to be exact for the averages of products of operators in the thermodynamic limit.

Unfortunately, we cannot generalize our proofs to the case of a true coupling between the $k = 0$ and $k \neq 0$ modes, therefore we cannot proof the Bogolyubov model itself, which includes as an important feature also a modified quasiparticle spectrum.

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PHYSICAL REVIEW B **70**, 045201 (2004)

Quasiclassical approach to Bose condensation in a finite potential well

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(Received 26 June 2003; revised manuscript received 25 February 2004; published 6 July 2004)

We treat the problem of self-consistently interacting bosons in the presence of a finite (but macroscopic) potential well within a quasiclassical approximation for the normal component and the order parameter. We solve the equilibrium problem and show that condensation actually occurs in two steps: one already at low densities with Bose condensation only in the well, and another one corresponding to the usual condensation in bulk. The peak and width of the distribution of trapped particles in the well display a distinct signature of the local condensation. A possible connection to recent experiments with excitons is discussed.

DOI: 10.1103/PhysRevB.70.045201 PACS number(s): 71.35.Lk, 03.75.Kk

I. INTRODUCTION

Since Bose-Einstein condensation (BEC) of trapped atoms was experimentally put into evidence,[1,2] much theoretical work was devoted to the discussion of the various aspects of BEC (see Refs. 3–5 for recent reviews). The theoretical description of the real-time evolution of Bose condensing systems has also received much attention in the last few years.[6–21] While condensation and spontaneous symmetry breaking in real time may be well understood for a homogeneous system of noninteracting bosons coupled to a thermal reservoir in the frame of the rate equation formalism[14] as an amplification of the however small initial condensate, the problem of weakly interacting bosons remains a problem of high complexity. The fact that the quasiparticle spectrum outside equilibrium is itself time dependent necessitates a quantum kinetic approach. This leads, however (even after various approximations), to very complicated systems of nonlinear integro-differential equations which cannot be analyzed with the usual methods of nonlinear dynamics. Nevertheless, numerical solutions illustrating BEC in real time are available.[18] Additional complications arise in inhomogeneous situations like in an external trap due to the fact that, strictly speaking, one cannot expect a true phase transition in a discreet spectrum.[22]

In order to describe Bose-Einstein condensation of particles in a potential well which contains many closely spaced energy levels, a quasiclassical approach may be appropriate, because the detailed knowledge of the population of higher lying energy levels is not of much interest and may cause an enormous numerical effort for its determination. Our attention is devoted here mainly to finite potential wells, for which the trapping in the well is controlled by the bulk population.

Quasiclassical approximations have been used already in the literature either for the description of the equilibrium[3] or of nonequilibrium.[10,12] In both cases this approach was used only for the noncondensate. On the other hand, the quasiclassical equilibrium distribution is not the stable solution of the quasiclassical kinetic equation, which is not quite satisfactory. In this paper we develop a consistent quasiclassical approach for the description of weakly interacting bosons with condensate and noncondensate in equilibrium and nonequilibrium in an external potential. The clue of our approach is to apply overall the quasiclassical limit. We consider the bosons as coupled to a thermostat by Markovian collision terms. This is a situation typical for excitons in solids. Within this frame one obtains a very simple and intuitive system of local equations (without memory effects) describing interacting bosons via a self-consistent (Hartree) potential. These equations conserve the average number of bosons and under critical conditions have a unique stable equilibrium solution with condensation and a quasiclassically distributed noncondensate. Of course this description, although mathematically consistent, is far from being satisfactory for all purposes, since pure quantum-mechanical effects beyond the quasiclassics such as the Fock terms were neglected.

In a previous publication[23] the evolution of a noninteracting Bose gas coupled to a thermal reservoir towards equilibrium in the presence of a finite potential well with many energy levels was discussed. It was shown that above a critical density the lowest state will be macroscopically populated, i.e., the local density will go to infinity. In this paper we take into account the repulsion between the bosons which eliminates this unphysical behavior. We develop this theory within a quasiclassical approach and a self-consistent treatment of the repulsion. We discuss in detail the exact equilibrium solution of these equations on a finite potential trap. The resulting scenario is a Bose condensation in two stages: At a first (rather low) critical overall density a locally confined condensate appears in the well; its distribution broadens with increasing overall particle density and at a second (higher) critical density one gets the usual bulk condensate. The spatial distribution of the trapped particles changes abruptly as the local condensation in the well occurs. The width of the distribution displays as a function of the overall particle density a minimum in the form of a cusp, while the slope of the peak changes discontinuously.

The described scenario may have a connection to recent experiments[24] on excitons in a Cu_2O semiconductor under mechanical stress, where a strong, narrow luminescence from the center of the well was observed.

II. QUASI-CLASSICAL DESCRIPTION OF A BOSE SYSTEM

The Wigner function depending on the momentum $\vec{p}$ and the coordinate $\vec{x}$ is defined as

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$$f(\vec{p},\vec{x}) = \int d\vec{y}\, e^{i\vec{p}\vec{y}/\hbar} \left\langle \psi\left(\vec{x}+\tfrac{1}{2}\vec{y},t\right)^{+} \psi\left(\vec{x}-\tfrac{1}{2}\vec{y},t\right)\right\rangle, \qquad (1)$$

where $\psi(\vec{x},t)$ is the second quantized wave function and $\langle \ldots \rangle$ means averaging over a given ensemble. This implies the normalization

$$\int d\vec{x} \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f(\vec{p},\vec{x},t) = \langle N \rangle. \qquad (2)$$

All the averages of operators which are a sum of two operators $O(\vec{x},\hbar/i\nabla) \equiv A(\vec{x}) + B(\hbar/i\nabla)$ depending on coordinate and momentum, respectively, may be expressed as integrals $\langle O(\vec{x},\hbar/i\nabla)\rangle = \int d\vec{x} \int d\vec{p}/(2\pi\hbar)^3 f(\vec{p},\vec{x},t) O(\vec{x},\vec{p})$. In the quasiclassical limit $f(\vec{p},\vec{x})$ is real and positive. For the case of an interaction with a thermal bath and slowly varying potentials[25,26] the following bosonic kinetic equation holds:

$$\begin{aligned}\left(\frac{\partial}{\partial t} + \frac{1}{m}\vec{p}\nabla_{\vec{x}} - \nabla_{\vec{x}} U(\vec{x},t)\nabla_{\vec{p}}\right) f(\vec{p},\vec{x},t) \\ = -\int \frac{d\vec{p}'}{\hbar^3}\left(W(\vec{p},\vec{p}') f(\vec{p},\vec{x},t)(1+f(\vec{p}',\vec{x},t)) - (\vec{p} \leftrightarrow \vec{p}')\right),\end{aligned} \qquad (3)$$

where the left-right arrow indicates an analogous term with $\vec{p}$ exchanged with $\vec{p}'$ and the positive transition rates $W(\vec{p},\vec{p}')$ satisfy the detailed balance relation

$$W(\vec{p},\vec{p}') = W(\vec{p}',\vec{p})\, e^{(\beta/2m)(p^2-p'^2)}, \qquad (4)$$

and $U(\vec{x},t)$ is a given potential. Generally speaking, one has an external potential $U_0(\vec{x})$, which we choose to be time independent (the given potential well) and an internal one, due to the interaction between the bosons through a potential $v(\vec{x})$ (where $\vec{x}$ is the relative coordinate of the two particles) related to the particle density $n(\vec{x})$ (Hartree self-energy)

$$\begin{aligned}U(\vec{x},t) &\equiv U_0(\vec{x}) + \int d\vec{x}'\, v(\vec{x}-\vec{x}')\, n(\vec{x}') \\ &= U_0(\vec{x}) + \int d\vec{x}'\, v(\vec{x}-\vec{x}') \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f(\vec{p},\vec{x}',t).\end{aligned} \qquad (5)$$

The Fock term, which in the case of a contact interaction[27] cannot be distinguished from the Hartree one has no classical counterpart and will be ignored in our treatment. The kinetic equation Eq.(3) has a single stable solution, corresponding to equilibrium

$$f_{eq}(\vec{p},\vec{x}) = \frac{1}{e^{\beta[(p^2/2m)+U_{eq}(\vec{x})-\mu]} - 1}, \qquad (6)$$

where μ is the chemical potential and $U_{eq}(\vec{x})$ is determined by the self-consistency equation

$$\begin{aligned}U_{eq}(\vec{x}) &= U_0(\vec{x}) + \int d\vec{x}'\, v(\vec{x}-\vec{x}')\, n(\vec{x}') \\ &= U_0(\vec{x}) + \int d\vec{x}'\, v(\vec{x}-\vec{x}') \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f_{eq}(\vec{p},\vec{x}').\end{aligned} \qquad (7)$$

The above-described equations are valid only in the absence of a condensation. As it is easy to understand, above a critical overall density $\langle N\rangle / V$ the Bose distribution in 3D cannot provide a description of the whole particle density, and this is in conflict with the conservation of the particle number. It can be shown rigorously[14] that for supercritical conditions, where a condensation occurs, Eq.(3) has no solutions with ordinary functions but only with distributions.

An appropriate approach to avoid this problem is to consider a two-component treatment where $f(\vec{p},\vec{x},t)$ describes only the noncondensate and a condensate is taken into account explicitly.

The existence of such a phase transition with spontaneous symmetry breaking in the presence of a potential is of course an idealization, which admits that the distance between the energy levels is negligible. This admition is implicitly made by the quasiclassical approximation. A discussion of the meaning of BEC in systems with discrete spectrum is given in Ref. 22.

In the presence of a condensate the self-consistent potential is then given by the total density (noncondensate and condensate)

$$n(\vec{x},t) = \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f(\vec{p},\vec{x},t) + |\langle \psi(\vec{x},t)\rangle|^2, \qquad (8)$$

where $\langle \psi(\vec{x},t)\rangle$ is the condensate amplitude (average of the second quantized wave function) and thus the two components are coupled. A further coupling will result from irreversible transitions from and to the condensate.

In order to include the condensate one needs an equation for its amplitude. For the mathematical consistency of the whole model, however, the quasiclassical approximation for the condensate also has to be formulated. One may start from the average of the exact equation of motion for $\psi(x)$

$$\begin{aligned}i\hbar\frac{\partial}{\partial t}\langle \psi(x,t)\rangle = \left(-\frac{\hbar^2}{2m}\nabla^2 + U_0(x)\right)\langle \psi(x,t)\rangle + \int dx'\, v(x-x') \\ \times \langle \psi(x')^{+} \psi(x') \psi(x,t)\rangle,\end{aligned}$$

and again retaining only the Hartree term we get in terms of the self-consistent (s.c.) potential $U(\vec{x},t)$ the equation for the condensate ampitude

$$i\hbar\frac{\partial}{\partial t}\langle \psi(\vec{x},t)\rangle = \left(-\frac{\hbar^2}{2m}\nabla^2 + U(\vec{x},t)\right)\langle \psi(\vec{x},t)\rangle. \qquad (9)$$

Neglecting the contribution of the noncondensate to the s.c. potential, this equation coincides with the Gross-Pitaevskii equation, but we do not neglect this contribution.

One has to recall here that the quasiclassical limit means $\hbar\to 0$, and therefore in this limit the kinetic energy term disappears (but as usual, the $\hbar$ from the time derivation survives) and we get

$$i\hbar\frac{\partial}{\partial t}\langle\psi(\vec{x},t)\rangle = U(\vec{x},t)\langle\psi(\vec{x},t)\rangle. \tag{10}$$

Next, as already mentioned, we have to include the transitions between the normal component and condensate due to the irreversible interaction with the thermostat in the Eqs.(3) and (10).

In the kinetic equation Eq. (3) we have to add the scattering rate due to the transitions from the state $\vec{p}$ to the $\vec{p}=0$ condensate

$$-W(\vec{p},0)f(\vec{p},\vec{x},t)|\langle\psi(\vec{x},t)\rangle|^2. \tag{11}$$

The initial state has to be populated, hence the factor $f(\vec{p},\vec{x},t)$. The final state factor contains the term due to stimulated transitions $\propto|\langle\psi(\vec{x},t)\rangle|^2$, because the term describing spontaneous transition is lost in the thermodynamic limit (see also Ref. 14). Similarly, the scattering rate due to transitions from the condensate to a state $\vec{p}$ is given by

$$W(0,\vec{p})(1+f(\vec{p},\vec{x},t))|\langle\psi(\vec{x},t)\rangle|^2. \tag{12}$$

Formally, these terms result by adding to the distribution function $f(\vec{p}\,',\vec{x},t)$ in the collision term of Eq. (3) a condensate term $(2\pi\hbar)^3\delta(\vec{p}\,')|\langle\psi(\vec{x},t)\rangle|^2$.

The gain and loss rates for the order parameter amplitude have to be calculated by taking the initial and final state factors for the condensate to be 1

$$\frac{i\hbar}{2}\langle\psi(\vec{x},t)\rangle\int\frac{d\vec{p}}{(2\pi\hbar)^3}(W(\vec{p},0)f(\vec{p},\vec{x},t) - W(0,\vec{p})(1+f(\vec{p},\vec{x},t))). \tag{13}$$

Such collision terms have been obtained[14] in the frame of the equation of motion method for a system of bosons weakly interacting only with a thermostat, decoupling higher order correlations and taking the long-time Markovian limit (see also Ref. 22).

The resulting system of coupled kinetic equations describes the quasiclassical evolution of interacting Bosons within the self-consistent approximation as

$$\begin{aligned}\left(\frac{\partial}{\partial t}+\frac{1}{m}\vec{p}\nabla_{\vec{x}}-\nabla_{\vec{x}}U(\vec{x},t)\nabla_{\vec{p}}\right)f(\vec{p},\vec{x},t)\\ = -\int\frac{d\vec{p}\,'}{(2\pi\hbar)^3}(W(\vec{p},\vec{p}\,')f(\vec{p},\vec{x},t)(1+f(\vec{p}\,',\vec{x},t))\\ -(\vec{p}\leftrightarrow\vec{p}\,'))-(W(\vec{p},0)f(\vec{p},\vec{x},t)-W(0,\vec{p})(1+f(\vec{p},\vec{x},t)))\\ \times|\langle\psi(\vec{x},t)\rangle|^2,\end{aligned} \tag{14}$$

together with

$$\begin{aligned}i\hbar\frac{\partial}{\partial t}\langle\psi(\vec{x},t)\rangle = U(\vec{x},t)\langle\psi(\vec{x},t)\rangle\\ +\frac{i\hbar}{2}\langle\psi(\vec{x},t)\rangle\int\frac{d\vec{p}}{(2\pi\hbar)^3}(W(\vec{p},0)f(\vec{p},\vec{x},t)\\ -W(0,\vec{p})(1+f(\vec{p},\vec{x},t))),\end{aligned} \tag{15}$$

and of course with the self-consistency condition

$$\begin{aligned}U(\vec{x},t) = U_0(\vec{x})+\int d\vec{x}\,'v(\vec{x}-\vec{x}\,')\biggl(\int\frac{d\vec{p}}{(2\pi\hbar)^3}f(\vec{p},\vec{x}\,',t)\\ +|\langle\psi(\vec{x}\,',t)\rangle|^2\biggr).\end{aligned} \tag{16}$$

The equation for the condensate density $|\langle\psi(\vec{x},t)\rangle|$ emerges as

$$\begin{aligned}\frac{\partial}{\partial t}|\langle\psi(\vec{x},t)\rangle|^2 = |\langle\psi(\vec{x},t)\rangle|^2\int\frac{d\vec{p}}{(2\pi\hbar)^3}(W(\vec{p},0)f(\vec{p},\vec{x},t)\\ -W(0,\vec{p})(1+f(\vec{p},\vec{x},t))).\end{aligned} \tag{17}$$

One sees that the equations conserve the total average number of bosons

$$\int d\vec{x}|\langle\psi(\vec{x},t)\rangle|^2+\int d\vec{x}\int\frac{d\vec{p}}{(2\pi\hbar)^3}f(\vec{p},\vec{x},t) = \langle N\rangle. \tag{18}$$

These equations describe an irreversible evolution. Above a certain overall particle density (at a given thermostat temperature) any arbitrarily small, but nonvanishing initial condensate in all points $\vec{x}$ will evolve to its finite stable equilibrium (nonhomogeneous) value, while below this density it will disappear.

Because the $\hbar\to 0$ limit for the Schrödinger equation is equivalent to the infinite mass limit, in this quasiclassical approximation the condensate does not propagate, but will be created or destroyed locally.

A theory, in which the quasiclassical approximation is used only for the noncondensate, runs into mathematical inconsistencies. Other inconsistencies would arise if one would try to develop a quasiclassical theory starting from the s.c. Bogolyubov-Popov theory[27] with a contact interaction and retaining automatically also the Fock terms. Although in this last frame one may introduce an "effective potential," this one is not the same for the noncondensate and condensate.

The theory we described takes into account the effect of the particle repulsion for the spatial distribution only. Anomalous pair correlations giving rise to a modified quasiparticle (Bogolyubov-) spectrum were ignored. This is also the price to be paid in order to get a local Markovian description. Since the quasiparticle spectrum in nonequilibrium is time dependent, only a quantum kinetic approach with memory effects would be adequate for its incorporation.

As for any quasiclassical approach, we expect the validity of the above-described theory for slowly varying potentials where the distance between the energy levels is very small. A detailed discussion of these conditions is given in the last section, where the theory is tentatively applied to interpretation of experimental data.

For a local interaction $v(\vec{x})=w\delta(\vec{x})$ the self-consistency equation also becomes local

$$U(\vec{x},t) = U_0(\vec{x}) + w\left(|\langle\psi(\vec{x},t)\rangle|^2 + \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f(\vec{p},\vec{x},t)\right), \tag{19}$$

which facilitates its treatment. In what follows we shall restrict the discussion to the equilibrium problem with such a local interaction.

III. BOSE CONDENSATION

We discuss here the equilibrium solution of Eqs. (14)–(16) considering a local interaction with $w>0$ (repulsion) at a given temperature for various overall densities $\bar{n} \equiv \langle N\rangle / V$. Here, the thermodynamic limit is implicitly understood.

In the absence of an equilibrium condensate $|\langle\psi(\vec{x})\rangle_{eq}|^2 = 0$, the equilibrium solution for the normal component is the Bose distribution, Eq.(6). Above a certain overall density $\bar{n}$ [see Eq.(18) divided by the volume V] a local condensate appears. The normal component should be further distributed according to the Bose distribution, which is a stationary solution of the left-hand side of the kinetic equation, Eq.(14), and of the part of the collision term which does not involve the condensate. The equilibrium condensate must be such as to cancel the additional collision term

$$|\langle\psi(\vec{x},t)\rangle|^2 \int \frac{d\vec{p}}{(2\pi\hbar)^3}(W(\vec{p},0)f(\vec{p},\vec{x},t) - W(0,\vec{p})(1+f(\vec{p},\vec{x},t))), \tag{20}$$

of both Eq. (14) and Eq. (17). Inserting the Bose function, the equilibrium condensate density $n_0(\vec{x})$, together with the detailed balance relation Eq. (4), this term becomes

$$n_0(\vec{x}) \int \frac{d\vec{p}}{(2\pi\hbar)^3} W(\vec{p},0) f_{eq}(\vec{p},\vec{x})(1 - e^{\beta(U_{eq}(\vec{x})-\mu)}). \tag{21}$$

As a result, the equilibrium condensate density $n_0(\vec{x})$ may be different from zero only where $U_{eq}(\vec{x})-\mu=0$. Due to the positivity of $f_{eq}(\vec{p},\vec{x})$, the chemical potential always must lie below the minimum of the potential $\mu \leq \min U_{eq}$. Therefore, the condensate can exist only in the minimum of the equilibrium self-consistent potential. This again corresponds to the behavior of particles with infinite mass.

The order parameter itself cannot be constant in time, but according to Eq.(15) oscillates as $\langle\psi(\vec{x},t)\rangle_{eq} = e^{-i\mu t}\sqrt{n_0(\vec{x})}$ and is actually determined only up to an arbitrary coordinate-dependent phase factor, which is determined from the initial condition. In general, the order parameter in equilibrium always oscillates with the chemical potential because the particle number operator does not commute with the macrocanonical density matrix in the case of the spontaneous symmetry breaking (see Ref. 18).

Without repulsion, the chemical potential can increase with the overall density up to the minimum of the potential $U_0(\vec{x})$. Further increase of the overall density $\bar{n}$ is possible only if the local condensate density $n_0(\vec{x})$ itself increases as the volume. This means that the assumed slow spatial variation which was required in the derivation of the quasiclassical description breaks down. However, we shall see that the situation changes substantially by the existence of a repulsion between the bosons, allowing a quasiclassical description of BEC.

Let us discuss what happens in the presence of a repulsion when one increases the chemical potential starting form below the minimum of the external potential, $U_0(\vec{x})$. This is of course equivalent to increasing the overall density $\bar{n}$.

The self-consistency condition for equilibrium with a local interaction is

$$U_{eq}(\vec{x}) = U_0(\vec{x}) + w\left(n_0(\vec{x}) + \int \frac{d\vec{p}}{(2\pi\hbar)^3}\, f_{eq}(\vec{p},\vec{x})\right). \tag{22}$$

This equation has been extensively used in the equilibrium theory of Bose-Einstein condensation of atoms trapped in an oscillator potential (see Ref. 3 and references therein), where similar considerations apply, although the nature of the trapping kinetics is different.

We choose an attractive external potential $U_0(\vec{x}) \leq 0$ which vanishes outside a finite domain around $\vec{x}=0$, and has a minimum in $\vec{x}=0$. One may expect that the potential $U_{eq}(\vec{x})$ also will have a point of minimum or a minimum surface and will be constant outside the range of U_0. Generally speaking, $\min U_{eq} > \min U_0$, since the repulsion opposes the attractive external potential.

For a repulsive interaction the increasing density increases also the minimum of $U_{eq}(\vec{x})$ and eventually flattens the bottom of the potential. The number of locally condensed particles will still remain finite and therefore does not contribute to the overall density. Only for an overall density $\bar{n}$ greater than the critical density

$$n_c = \int \frac{d\vec{p}}{(2\pi\hbar)^3} \frac{1}{e^{\beta(p^2/2m)} - 1}, \tag{23}$$

an overall condensate appears.

Because outside the range of $U_0(\vec{x})$ both the local density and the self-consistent potential are constant, we have for $\bar{n} \leq n_c$ the following equation for the determination of the chemical potential:

$$\int \frac{d\vec{p}}{(2\pi\hbar)^3} \frac{1}{e^{\beta[(p^2/2m)+w\bar{n}-\mu]} - 1} = \bar{n}. \tag{24}$$

This equation has a solution only for $\mu \leq wn_c$. Above this value an overall condensate has to appear.

For simplicity, let us consider a spherically symmetric situation [$U_0(r)$ with $U_0(r)=0$ for $r>R$ and a negative minimum in $r=0$]. We consider the case where μ is above the minimum of U_0, but below wn_c. Let us look at the shape of $U_{eq}(r)$ coming from large r. Here, one has a spatially constant self-consistent potential

$$U_{eq}(r) = w\bar{n};\ (r>R). \tag{25}$$

For $r<R$ one gets a radius-dependent density and the self-consistency equation

$$U_{eq}(r) = U_0(r) + w \int \frac{d\vec{p}}{(2\pi\hbar)^3} \times \frac{1}{e^{\beta[(p^2/2m)+U_{eq}(r)-\mu]}-1}; \; (r_0 < r < R) \tag{26}$$

for $U_{eq}(\vec{x})-\mu>0$ giving rise to a negative $U_{eq}(r)$ varying monotonously. At a certain $r=r_0$ one may get $U_{eq}(r_0)=\mu$ and the integral achieves its maximal value n_c. Due to the condition $U_{eq}(\vec{x})-\mu \geqslant 0$ all the points $r<r_0$ must belong to the minimum of a monotonous $U_{eq}(r)$ and a condensate $n_0(r)$ must emerge

$$U_{eq}(r) = U_0(r) + w(n_c + n_0(r)); \; (r < r_0), \tag{27}$$

in order to ensure that $U_{eq}(r)$ remains constant for $r<r_0$, i.e.,

$$U_{eq}(r) = U_{eq}(r_0+0) = \mu (r < r_0), \tag{28}$$

implying

$$n_0(r) = \frac{1}{w}(U_0(r_0) - U_0(r))(r < r_0). \tag{29}$$

When the overall density $\bar{n}$ reaches the critical value n_c, i.e., $\mu=wn_c$, the self-consistent potential is completely flat, r_0 approaches R, while the local condensate density approaches $-(1/w)U_0(r)$.

For overall densities above the critical one, one gets an overall condensate, while the local condensate density is $\bar{n}-n_c-(1/w)U_0(r)$.

This scenario is complete and self-consistent!

However, local condensation shows nonanalyticities only of local entities, but does not affect the analyticity of the total thermodynamic entities. Therefore, according to the usual terminology it does not define a phase transition.

We expect the following scenario in a self-consistent but quantum-mechanical picture: In the presence of a repulsive interaction between the particles the chemical potential approaches the lowest s.c. level and pushes it up without actually touching it until the bound states disappear completely and the true bulk phase transition occurs. In this way a finite local density appears first, which can be higher than the critical density of the bulk.

IV. NUMERICAL ILLUSTRATION

For the following, we choose a spherically symmetric, attractive ($v<0$) external potential

$$U_0(r) = v\left(1 - \left(\frac{r}{R}\right)^2\right). \tag{30}$$

For further discussions it is convenient to measure densities in units of the critical density n_c, the energies in units of k_BT ($\mathcal{U} \equiv \beta U_{eq}$, $\mathcal{U}_0 \equiv \beta U_0$, $\mathcal{M} \equiv \beta\mu$, $\mathcal{V} \equiv \beta v$), the radius in units of $R(\rho \equiv r/R)$, and use the dimensionless interaction constant $\mathcal{W} \equiv \beta w n_c$.

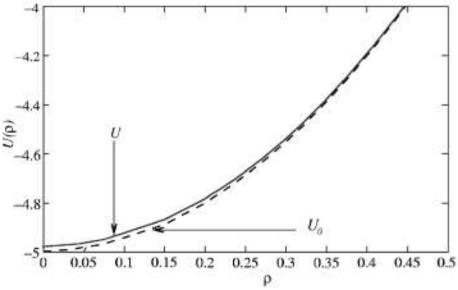

FIG. 1. External potential $\mathcal{U}_0(\rho)$ (dashed line) and self-consistent potential $\mathcal{U}(\rho)$ (full line) for a subcritical condition $\mathcal{M}=-5.2(\bar{n}=0.0021n_c)$ with a potential depth $\mathcal{V}=-5$, and an interaction constant $\mathcal{W}=0.05$.

Then

$$\mathcal{U}_0(\rho) = \mathcal{V}(1-\rho^2)(\mathcal{V}<0), \tag{31}$$

and one has to solve the equation

$$\mathcal{U}(\rho) = \mathcal{U}_0(\rho) + \mathcal{W}\frac{1}{\int_0^\infty ds \frac{\sqrt{s}}{e^s-1}} \int_0^\infty ds \frac{\sqrt{s}}{e^{s+\mathcal{U}(\rho)-\mathcal{M}}-1}, \tag{32}$$

in every point $\rho_0<\rho<1$ in order to reconstruct the condensate density as described above. Obviously the radius $\rho_0=r_0/R$ for which the self-consistent potential touches the chemical potential is defined by the solution of the equation

$$\mathcal{M} = \mathcal{V}(1-\rho_0^2) + \mathcal{W}, \tag{33}$$

which exists in the interval $0<\rho_0<1$ only for $\mathcal{M}>\mathcal{V}+\mathcal{W}$.

Since the transcendental equation Eq. (32) is local, for a numerical solution it is convenient to solve it in favor of $\mathcal{U}_0$ at a given $\mathcal{U}$, thus performing a simple integration. The association with a certain radius is given by the explicit definition of $\mathcal{U}_0(\rho)$.

We illustrate the above-described scenario with some numerical examples. We choose $\mathcal{V}=-5$ and $\mathcal{W}=0.05$, and discuss later what happens for stronger coupling interaction potentials.

In Fig. 1, the potentials $\mathcal{U}_0(\rho)$ (dotted line) and $\mathcal{U}(\rho)$ (full line) are shown for a chemical potential $\mathcal{M}=-5.2$ just below the minimum $\mathcal{V}$ of the external potential, i.e., slightly below the onset of the local condensation. One sees that the self-consistent potential $\mathcal{U}(\rho)$ is pushed up. For the chemical potential of $\mathcal{M}=-4.8$ (corresponding to a supercritical density) just above the minimum $\mathcal{V}$ of the external potential, Fig. 2 shows that the self-consistent potential obtains a flat bottom at the chemical potential. This is a typical signature for the appearance of a condensate here in the range $0<\rho<0.2$. Figure 3 shows the resulting peak in the density. It is remarkable that the overall densities for the two cases $\bar{n}=0.0021n_c$ and $\bar{n}=0.0031n_c$ do not differ very much, both being much lower than the critical one, while the condensate density in the second case exceeds the critical density by a factor of 4.

A further increase of the chemical potential $\mathcal{M}$ (or equivalently of the overall density $\bar{n}$) will lift the bottom of

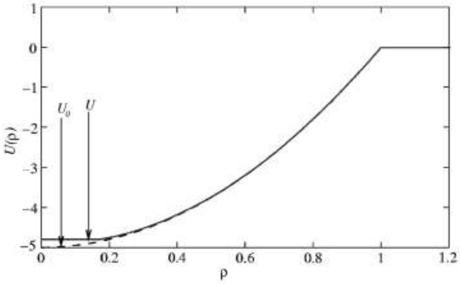

FIG. 2. External potential $\mathcal{U}_0(\rho)$ (dashed line) and self-consistent potential $\mathcal{U}(\rho)$ (full line) for a supercritical condition $\mathcal{M}=-4.8(\bar{n}=0.0031n_c)$ with a potential depth $\mathcal{V}=-5$, and an interaction constant $\mathcal{W}=0.05$.

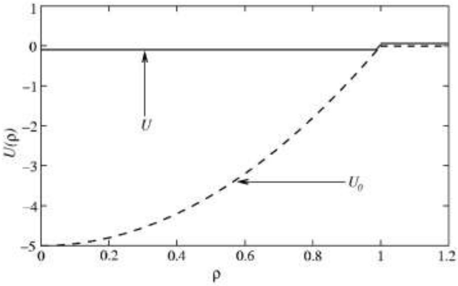

FIG. 4. The external potential $\mathcal{U}_0(\rho)$ (dashed line) and the renormalized potential $\mathcal{U}(\rho)$ (full line) for the normalized chemical potential $\mathcal{M}=-0.1$, and the corresponding density ($\bar{n}=0.584n_c$) with a potential depth $\mathcal{V}=-5$, and an interaction constant $\mathcal{W}=0.05$.

the self-consistent potential further as it may be seen in Fig. 4, while the corresponding condensate density in the minimum of the self-consistent potential reaches very high values and the spatial density distribution (shown in Fig. 5) widens out. Actually, it is already very close to $-(1/\mathcal{W})\mathcal{U}_0$, which will be its value at $\bar{n}=n_c$.

The general feature of the condensation scenario is seen most clearly in Fig. 6, where we represented the density peak of the trapped particles and its half-width as functions of the overall density $\bar{n}$ (here, with the choice $\mathcal{W}=0.05$ and $\mathcal{V}=-5$). The height of the density peak increases with the overall density, but its slope increases abruptly at the overall density of $0.0027n_c$. The half-width of the local distribution at the same overall density has a minimum resembling a cusp.

For a stronger interaction, corresponding to $\mathcal{W}=0.5$, the role of the local condensate is not so striking. It can enhance the density in the well maximally by a factor of 10, resulting in a less pronounced peak. Nevertheless, the density inside the well may reach very high values compared to the overall density. This is illustrated in Fig. 7 for two positions of the chemical potential $\mathcal{M}=-4.7$ and $\mathcal{M}=-4.3$, corresponding to situations without and with condensate. In both cases the overall density is much lower than the critical one ($\bar{n}=0.0035n_c$ and $0.0052n_c$).

The densities described above, with condensate only in the local potential well, are all below the critical density of the bulk. In terms of the temperature it corresponds to being much higher than the critical temperature of the bulk $k_BT_c^{\text{bulk}}=6.028\,85(\hbar^2/m)\bar{n}^{2/3}$, however below the critical temperature of the ideal oscillator potential $k_BT_c^{\text{well}}=0.831\,907\hbar\omega_0 N^{1/3}$ (in terms of the energy distance $\hbar\omega_0$ between the oscillator levels and the number of particles $\langle N\rangle$ in the well).[3] At the same time, for the validity of the quasiclassical approach, the temperature must be much higher than the distance between the levels. It is, however, possible that just around the critical temperature of the well T_c^{well} our approximations lose their validity due to critical fluctuations.

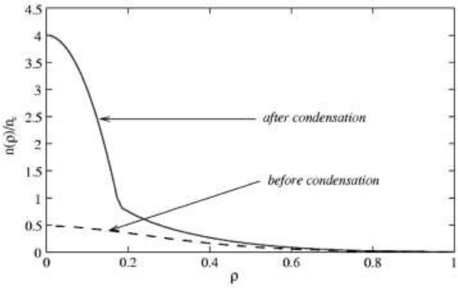

FIG. 3. The local density $n(\rho)/n_c$ for the sub- and supercritical overall densities $\bar{n}=0.0021n_c$ (dashed line) and $\bar{n}=0.0031n_c$ (full line), respectively with a potential depth $\mathcal{V}=-5$, and an interaction constant $\mathcal{W}=0.05$.

V. EXCITONS IN A LOCAL POTENTIAL

In recent experiments[24] Naka and Nagasawa have observed after uniform illumination a strong luminescence in the center of a strain-induced potential well in a Cu_2O sample at $T=2$ K. This luminescence may be due to the con-

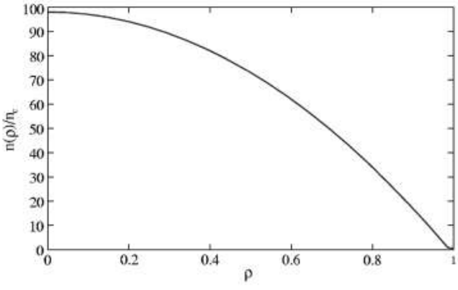

FIG. 5. The local density $n(\rho)/n_c$ for the chemical potential $\mathcal{M}=-0.1$ (corresponding density $\bar{n}=0.584n_c$) with a potential depth $\mathcal{V}=-5$, and an interaction constant $\mathcal{W}=0.05$.

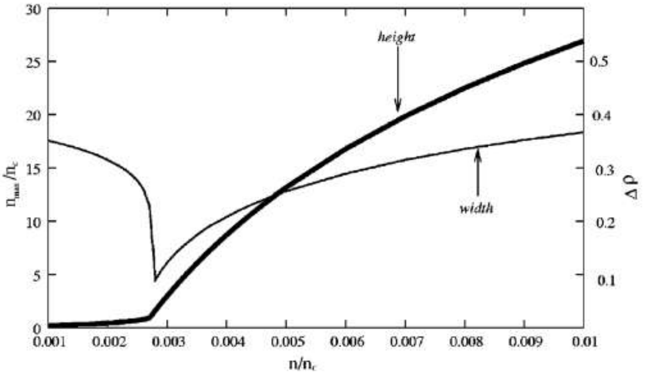

FIG. 6. The dependence of the height (thick line) and half width (thin line) of the density distribution of the trapped particles on the overall density in the vicinity of the condensation point.

densation of para-excitons (having a Bohr radius $a_{ex} = 0.710^{-7}$ cm). The luminescence peak was concentrated in a small vicinity around the potential well minimum. The strain induced potential in their experiment is asymmetric, but may be approximated qualitatively by a parabolic potential well like the one we choose having a depth of $v \approx -1$ meV and a radius of $r_0 \approx 10^{-2}$ cm. This would correspond in our dimensionless parameters to $\mathcal{V} = -5$. In an ideal oscillator potential with the same curvature one gets an energy spacing of the quantum-mechanical levels of $\hbar\omega \approx 10^{-4}$ meV and a very small radial extension of the ground-state oscillator wave function of 510^{-5} cm for the Cu_2O exciton mass of $m = 2.7 m_0$. Since $v/\hbar\omega \gg 1$ many energy levels are contained in the potential well and one may expect the validity of the quasiclassical description. At this temperature one has in Cu_2O a critical density of $n_c \approx 10^{17}$ cm^{-3}. The density of the excitons created by laser illumination has not been measured directly.

On the other hand, the interaction constant w is not directly experimentally accessible. Tentatively one may consider the well-known potential between hydrogen atoms obtained by the Heitler-London theory of the hydrogen molecule[29] and replace the Rydberg energy and the electron mass, with the corresponding entities of the exciton (exciton Rydberg E_R and exciton Bohr radius a_B

$$v(r) = \frac{E_R}{S(r)^2} \int d\vec{y}_1 \int d\vec{y}_2 \Bigg((\phi(\vec{y}_1)\phi(\vec{y}_2)\phi(\vec{y}_1 + \vec{r})\phi(\vec{y}_2 - \vec{r})) \times \left(\frac{1}{|\vec{y}_1 - \vec{y}_2 + \vec{r}|} + \frac{1}{|\vec{r}|} - \frac{1}{|\vec{y}_1 + \vec{r}|} - \frac{1}{|\vec{y}_2 - \vec{r}|} \right) \times \phi(\vec{y}_1)\phi(\vec{y}_2) \Bigg), \tag{34}$$

where the normalization factor is

$$S(r)^2 = 1 \int d\vec{y}_1 \int d\vec{y}_2 \phi(\vec{y}_1)\phi(\vec{y}_1 + \vec{r})\phi(\vec{y}_2)\phi(\vec{y}_2 - \vec{r}). \tag{35}$$

The 1s wave functions $\phi(\vec{x})$ are

$$\phi(\vec{x}) = \frac{1}{\sqrt{\pi a_B^3}} e^{-|\vec{x}'/a_B}. \tag{36}$$

Actually, Sugiura[29] evaluated analytically all the involved integrals. The positive sign corresponds to the symmetrical state, while the negative one corresponds to the antisymmetrical state in the coordinates. There are also more sophisticated variational calculations for the bound molecular state (symmetrical state), however not for the repulsive (antisymmetrical) case, where a variational approach is not justified.

The para-excitons (singlet spin state) in Cu_2O are the lowest exciton state. Then, antisymmetric wave functions in the coordinates for the identical particles should be constructed and the resulting force between the excitons will be repulsive as shown in Fig. 8. The Heitler-London potential decreases with the distance between the excitons exponentially as e^{-2r/a_B} and the local approximation should hold. The calculated value of the interaction constant w is $49.26 E_R a_B^3$, respectively. For Cu_2O this value leads to $\mathcal{W} \approx 1$. This means that in Cu_2O the strong coupling case is realized.

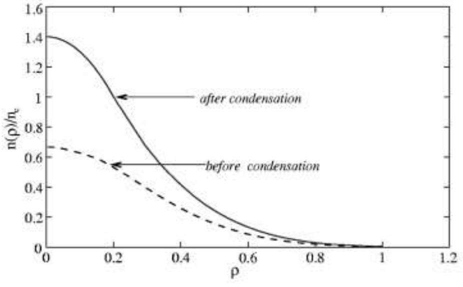

FIG. 7. The local density $n(\rho)/n_c$ for the sub- and supercritical overall densities at $\bar{n} = 0.0035 n_c$ (dashed line) and $\bar{n} = 0.0052 n_c$ (full line) for the strong interaction case with a potential depth $\mathcal{V} = -5$, and an interaction constant $\mathcal{W} = 0.5$.

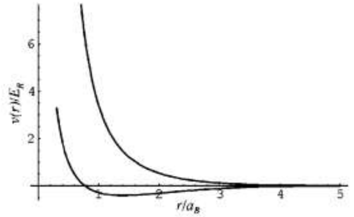

FIG. 8. The Heitler-London potential for the symmetrical and antisymmetrical configurations (lower, respectively, upper curve).

Because we had to identify excitons with point-like bosons, it is questionable to consider a Heitler-London potential between the excitons at distances smaller than $2a_B$. Actually, one should consider such quantitative estimates with some reservations. Therefore, one cannot make a reliable quantitative prediction for the trapping wells in Cu_2O. However, the photoluminescence of the trapped excitons should display the characteristics of Fig. 6 if a local Bose condensation occurs.

Finally, we want to draw attention to similar observations[28] with excitons confined to an indirect GaAs/AlGaAl quantum well layer. Here also a strong localized luminescence has been observed due to trapping, but the system is rather two-dimensional.

Actually, all the considerations relating to Bose-Einstein condensation of excitons are subject to the condition, that the electron-hole pairs are mainly bound into excitons, which under experimental conditions may be too restrictive.

In conclusion, we have formulated a quasiclassical condensation kinetics of interacting bosons which are in contact with a thermostat and trapped in a finite macroscopic potential well. The theory is based on the coupled equations Eqs. (14)–(16) for the noncondensate and condensate. The equilibrium solution has been discussed in detail. Due to the repulsive interaction, the trapping potential becomes renormalized after the condensation has occurred, allowing the chemical potential to increase over the minimum of the well for an increasing overall particle density. In particular, it has been shown that the spatial distribution of the trapped particles shows distinct changes as a local Bose Einstein condensation occurs. The possible relation of these results to recent experiments with trapped excitons has been discussed.

ACKNOWLEDGMENTS

One of the authors (A.M.B) thanks the Deutsche Forschungsgemeinschaft for the generous support allowing his stay at the University of Frankfurt. We appreciate private communications on their experiments by N. Nagasawa and D.S. Chemla.

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