Determinantal solution of the density matrix in quantum transport theory

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A new method previously elaborated on in time-dependent quantum mechanics is recalled and used to establish, on the basis of the unperturbed Hamiltonian, explicit and tractable expressions of the long-term density matrix, well suited to transport problems. A constant collision potential and a constant external field, switched on at $t=0$, are assumed. As an illustration, the general results are applied to the quantum theory of electrical conductivity in the case of quasifree electrons.

I. INTRODUCTION

The quantum treatment of transport processes in solid-state physics is well known to be dominated by the Kubo linear-response theory,[1] in terms of correlation functions of current density. In spite of its rather attractive form, dealing directly with the quantum-mechanical motion of microscopic quantities, and its widespread use in applications, this formalism is not free from severe criticisms. An important one, as discussed by Van Vliet,[2] is concerned with the lack of dissipation and relaxation, inherent to the starting von Neumann equation of the density matrix, which does not include the approach to equilibrium. On the other hand, the density matrix is extracted from the Schrödinger integral equation thanks to a preliminary linearization, in order to avoid a self-consistent resolution. This procedure, which is questionable when seeking the long-term behavior, has been shown to be equivalent to randomization by van Kempen,[3] and seems particularly dangerous in the vicinity of resonances. This is perhaps the reason why this formalism did not succeed in yielding convincing results when applied to the cyclotron-resonance linewidth, in the last decade.[4]

In common transport theory the external driving field is assumed to be adiabatically applied to the system from a remote past ($t=-\infty$), tending to support the idea that the knowledge of the adiabatic response is required to get a correct description of the transport process. This may look surprising because of the physical feeling that the way of applying the field is irrelevant, as long as the transport process is understood as a steady-state balance between the competitive effects of applied field and collisions. This state of things is, in fact, intimately connected with the lack of relaxation dissipation. Many theoretical descriptions, indeed, fail in taking properly into account the counterbalance between driving field and collisions.

The main concern of the present paper is to show that, to some extent, a trace-conserving theory previously developed[5,6] offers a simple tool to overcome the above-mentioned difficulties, at least in the cases of a constant or harmonic perturbation. In this method, denoted "determinantal," the time-dependent Schrödinger equation is rewritten in Laplace space and solved by means of Cramer's formulas, with some further arrangements to replace determinant expansions by more suitable ones. We are going to see that when applied to the linear-response theory the method yields a well-defined long-term response, independent of the way of applying the external field. In particular, there is no need to assume a very slow adiabatic application of the field and, therefore, use can be made of the Laplace transform which is well suited to the application of external disturbance from $t=0$, as in elementary circuit theory.

In Sec. II the starting von Neumann equation is presented with some useful arrangements, for notational convenience and discussion. The method of calculation is described in Sec. III, whereas Sec. IV is devoted to the derivation of more explicit expressions suited to current applications. A simplified treatment of the quantum theory of electrical conductivity will be given, as an illustration, in Sec. V.

II. THE von NEUMANN DENSITY-MATRIX EQUATION

The physical system is described by the unperturbed Hamiltonian H_0 with eigenstates $|a\rangle, |b\rangle, \ldots, |z\rangle, \ldots$, and related eigenvalues $\hbar\omega_a, \hbar\omega_b, \ldots, \hbar\omega_z, \ldots$. For present purposes it will be sufficient to describe the predominant contribution to collisions by means of a constant potential V. Then, other contributions, often involving inelastic collisions associated with the coupling of the system with the heat bath, correspond to much slower relaxation processes and are, therefore, sufficiently well represented by a simple phenomenological relaxation time τ. The consideration of such accessory relaxation processes is essential for the overall coherence of the theory, because they permit the equilibrium to be recovered, through relaxation with the bath, as the external field is switched off. But as far as V is strongly predominant, these processes should be eliminated from the final result which, thereby, is expected not to depend on τ, at least in the linear limit.

Also, for the sake of simplicity, the external field will be described with the constant operator F, assumed to be turned on at $t=0$. As a result, the von Neumann equa-

tion for the density matrix $\rho(t)$ will be written as

$$\frac{d\rho}{dt}=-i\hbar^{-1}[H_0+V+FY(t),\rho]-\frac{\rho-\rho_0}{\tau} . \quad (1)$$

$Y(t)$ denotes the Heaviside step function, and $\rho_0=\rho(0)$ is the initial value of $\rho(t)$.

As is well known, the microscopic von Neumann equation strictly applies to the complete isolated system, including the heat bath. In Eq. (1) this requirement is obeyed to within a very good approximation, through the introduction of the relaxation parameter τ. Since the coupling between the system and the heat bath is weak, the complete density matrix ρ_C can be written in the form of a Born-Oppenheimer–type product

$$\rho_C=\rho\rho_B \quad \text{with} \quad [\rho,\rho_B]\cong 0 . \quad (2)$$

ρ_B is the heat-bath density matrix (e.g., that of a phonon system). Let H_B be the bath Hamiltonian and V_B the coupling Hamiltonian between the system and the bath. We have the complete equation

$$\frac{d\rho_C}{dt}=-i\hbar^{-1}[H_0+H_B+V+V_B+FY(t),\rho_C] . \quad (3)$$

But the change of the bath motion due to the coupling with the system is completely negligible, so that

$$\frac{d\rho_B}{dt}=-i\hbar^{-1}[H_B,\rho_B] . \quad (4)$$

By taking Eqs. (2) and (4) into account, together with the evident commutation relations between the two quasi-independent subsystems,

$$[\rho,H_B]=0, \quad [\rho_B,H_0+V]=0 ,$$

Eq. (1) is recovered from Eq. (3), in the form

$$\frac{d\rho}{dt}=-i\hbar^{-1}[H_0+V+FY(t),\rho]-i\hbar^{-1}[V_B,\rho] ,$$

showing how the coupling with the bath on the right-hand side has been simplified through the introduction of the phenomenological parameter τ.

Finally, Eq. (1) is quite consistent with the partitioning of the complete isolated system into a small subsystem and a large reservoir. As discussed by Fano[7] and Balian *et al.*,[8] such a partitioning is the essential source of irreversibility and dissipation.

Assuming statistical equilibrium prior to the application of the perturbation, $\rho\rho_B$ is represented, at $t=0$, by the microcanonical density operator. But, as shown in elementary quantum statistical mechanics, the microcanonical density in the whole system leads to the canonical density in the small subsystem. The collision potential V contributes the initial thermal equilibrium, so that ρ_0 is, in fact, a function of H_0+V instead of H_0 alone. This entails the commutation relation

$$[\rho_0,H_0+V]=0 . \quad (5)$$

All previous considerations concern the system as a whole. In general, H_0, V, F, and ρ will refer to many-body operators. In some cases the reduction of the formalism to a one-particle problem will be possible. In such cases, ρ_0 can be expressed in terms of individual Fermi or Bose-Einstein occupation functions.

III. FORMAL SOLUTION OF THE DENSITY-MATRIX EQUATION

We shall proceed with a brief reformulation of the theory[5,6] more suited to the present transport problem. Returning to Eq. (1), upon performing the Laplace transform

$$R(\nu)=\int_0^{+\infty}\exp(-\nu t)\rho(t)dt ,$$

we obtain

$$(\nu+\tau^{-1})R(\nu)+i\hbar^{-1}[H_0+V+F,R(\nu)]=\rho_0(1+\nu^{-1}\tau^{-1}) . \quad (6)$$

Then, taking the cb matrix element of each side of this equation, we have

$$(\nu+\tau^{-1}+i\omega_{cb})R^c_b(\nu)+i\hbar^{-1}[V+F,R(\nu)]^c_b=\rho_{0b}^c(1+\nu^{-1}\tau^{-1}) ,$$

where $\omega_{cb}=\omega_c-\omega_b$. Summations over repeated indices are implicit throughout. The R operator will next be regarded as a column vector belonging to the Liouville space

$$\mathcal{E}_{H_0H_0}=\mathcal{E}_{H_0}\otimes\mathcal{E}^*_{H_0} ,$$

defined by tensorial multiplication of the unperturbed Hilbert space $\mathcal{E}_{H_0}$ by the dual space $\mathcal{E}^*_{H_0}$. In $\mathcal{E}_{H_0H_0}$, basis states will be specified by all pairs of indices (c,b). Superoperators involving commutation with V or F are conveniently defined by

$$\mathbb{K}_V=i\hbar^{-1}(V\delta-\delta V) , \quad (7)$$

where δ is the Kronecker delta. This means that any c_2b_2-c_1b_1 matrix element is given by

$$\mathbb{K}_{Vc_1b_2}^{c_2b_1}=K_{Vc_1}^{c_2}\delta_{b_2}^{b_1}-\delta_{c_1}^{c_2}K_{Vb_2}^{b_1} \quad \text{with} \quad K_V=i\hbar^{-1}V . \quad (8)$$

Diagonal matrix elements of V will be ignored without serious loss of generality. We shall also introduce the diagonal operator

$$\mathbb{d}=(\nu+\tau^{-1})\mathbb{I}+\mathbb{K}_{H_0} \quad \text{with} \quad \mathbb{K}_{H_0}=i\hbar^{-1}(H_0\delta-\delta H_0) . \quad (9)$$

$\mathbb{I}$ denotes the unity operator in $\mathcal{E}_{H_0H_0}$. Making use of the above definitions, Eq. (6) becomes

$$(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F)\mathbb{R}=\rho_0(1+\nu^{-1}\tau^{-1}) , \quad (10)$$

which can also be written as

$$[\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F)]\mathbb{R}=\mathbb{d}^{-1}\rho_0(1+\nu^{-1}\tau^{-1}) . \quad (11)$$

In terms of $\mathbb{R}$-matrix matrix elements, say $\langle cb|\mathbb{R}\rangle=\mathbb{R}^c{}_b$, we have

$$\mathbb{R}^{c}{}_{b}+\mathbb{d}_{cb}^{-1}(\mathbb{K}_{V}{}^{cb}_{c_1 b}+\mathbb{K}_{F}{}^{cb}_{c_1 b})\mathbb{R}^{c_1}{}_{b_1}=\mathbb{d}_{cb}^{-1}\rho_{0b}^{c}(1+\nu^{-1}\tau^{-1}) . \tag{12}$$

Equation (12) turns out to represent a linear system in $\mathcal{E}_{H_0H_0}$ and as such it can be solved in terms of Cramer's minors and determinants (denoted by the symbol $\mathbb{D}$ with an appropriate set of indices) pertaining to the matrix $\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F)$. For the $\mu\lambda$ component of $\mathbb{R}$, we thus have

$$\mathbb{R}^{\mu}{}_{\lambda}=\frac{\mathbb{D}_{c\lambda}^{\mu b}}{\mathbb{d}_{cb}\mathbb{D}}\rho_{0b}^{c}(1+\nu^{-1}\tau^{-1}) . \tag{13}$$

Using Cayley's expansions of determinants,[9] Eq. (13) yields an explicit expression of the solution in increasing powers of K_V and K_F. It has been shown, however, that both numerator and denominator contain unexpected closed sequences of transitions (related to secular terms of perturbation series), which can be removed by means of a suitable "reduction procedure."[5] The latter turns out to come down to the division of the upper and lower determinants by the diagonal minor $\mathbb{D}_{cb}^{cb}$. It is convenient to reestablish this procedure straightforwardly, so as to introduce some appropriate minor changes. Equation (10) first gives

$$|\mathbb{R}\rangle=(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F)^{-1}(1+\nu^{-1}\tau^{-1})|\rho_0\rangle ,$$

or, equivalently,

$$|\mathbb{R}\rangle=\sum_{c,b}(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F)^{-1}\mathbb{P}_{cb}(1+\nu^{-1}\tau^{-1})|\rho_0\rangle . \tag{14}$$

$\mathbb{P}_{cb},\mathbb{Q}_{cb}$ denote the projector on the cb state and the complementary space, respectively, in $\mathcal{E}_{H_0H_0}$.

As pointed out above, in current-transport problems the proper "unperturbed states" are not eigenstates of H_0 but, instead, those of H_0+V, giving rise to the commutation relation (5). Using the commutation kernels, the latter can be written in $\mathcal{E}_{H_0H_0}$ as follows

$$(\mathbb{K}_{H_0}+\mathbb{K}_V)|\rho_0\rangle=0 . \tag{15}$$

which entails, from Eq. (9),

$$(\mathbb{d}+\mathbb{K}_V)|\rho_0\rangle=[(\nu+\tau^{-1})\mathbb{I}+\mathbb{K}_{H_0}+\mathbb{K}_V]|\rho_0\rangle$$
$$=(\nu+\tau^{-1})|\rho_0\rangle .$$

Consequently, Eq. (14) will be rewritten, without change, as

$$|\mathbb{R}\rangle=\sum_{c,b}(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F)^{-1}(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\mathbb{P}_{cb}$$
$$\times\frac{1+\nu^{-1}\tau^{-1}}{\nu+\tau^{-1}}|\rho_0\rangle .$$

Setting

$$|\mathbb{R}\rangle=\sum_{c,b}|\mathbb{R}^{(cb)}\rangle ,$$

every cb component will be considered separately,

$$|\mathbb{R}^{(cb)}\rangle=(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F)^{-1}(\mathbb{d}+\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\frac{\rho_{0b}^{c}}{\nu}|cb\rangle .$$

Factorizing out the $\mathbb{d}$'s and reversing the brackets back into the left-hand member yields

$$[\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})]^{-1}[\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F)]|\mathbb{R}^{(cb)}\rangle$$
$$=\nu^{-1}\rho_{0b}^{c}|cb\rangle . \tag{16}$$

We thus obtain a somewhat new form of the system (11), whose Cramer solution can again be written as

$$\mathbb{R}^{\mu}{}_{\lambda}=\frac{\mathbb{D}_{c\lambda}^{\mu b}(\mathbb{D}_{cb}^{cb})^{-1}}{\nu\mathbb{D}(\mathbb{D}_{cb}^{cb})^{-1}}\rho_{0b}^{c} . \tag{17}$$

$\mathbb{D}_{cb}^{cb}$ now denotes the determinant of the matrix

$$\mathbb{S}_{cb}=\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb}) . \tag{18}$$

This expression is slightly changed with regard to the initial reduction procedure,[6] in which the $\mathbb{S}_{cb}$ matrix was just $\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F)\mathbb{Q}_{cb}$, so that $\mathbb{D}_{cb}^{cb}$ exactly represented by cb-cb minor of the matrix $\mathbb{I}+\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F)$.

IV. PRACTICAL FORM OF THE SOLUTION

In practice, the determinant ratios arising in Eq. (17) can be given a convenient form in terms of geometrical series.[10] Making use of the splitting

$$\mathbb{d}^{-1}\mathbb{K}_F=\mathbb{d}^{-1}\mathbb{K}_F\mathbb{P}_{cb}+\mathbb{d}^{-1}\mathbb{K}_F\mathbb{Q}_{cb} ,$$

and the notation (18), Eq. (16) can be written in the new form

$$(\mathbb{I}+\mathbb{S}_{cb}^{-1}\mathbb{d}^{-1}\mathbb{K}_F\mathbb{P}_{cb})|\mathbb{R}^{(cb)}\rangle=\nu^{-1}\rho_{0b}^{c}|cb\rangle , \tag{19}$$

Due to the projector $\mathbb{P}_{cb}$, the operator on the left-hand side of (19) is represented by a column matrix, so that determinant and minors reduce to single matrix elements [see also Eqs. (A17) and (A18) in Ref. 10]. This gives the following equivalent operational form of the solution (17):

$$\mathbb{R}^{\mu}{}_{\lambda}=\frac{\langle\mu\lambda|(\mathbb{I}+\mathbb{Q}_{cb}\mathbb{S}_{cb}^{-1}\mathbb{d}^{-1}\mathbb{K}_F)|cb\rangle}{\nu(1+\langle cb|\mathbb{S}_{cb}^{-1}\mathbb{d}^{-1}\mathbb{K}_F|cb\rangle)}\rho_{0b}^{c} . \tag{20}$$

We thus arrive at a "once-reduced" result (the "reduction" is associated with the above division by $\mathbb{D}_{cb}^{cb}$), leading to a well-defined long-term limit by means of the elementary rule

$$\rho(t\to\infty)=\lim_{\nu\to 0+}[\nu\mathbb{R}(\nu)] .$$

Explicit expressions involving increasing orders of F and V will be finally obtained through expansion of $\mathbb{S}_{cb}^{-1}$ in Eq. (20). Thanks to definition (7) leading to rule (8) for the matrix elements of the commutator kernels $\mathbb{K}$, repeated applications of the Liouville operator can be performed in a mechanical manner, lending itself, if needed, to computational methods. In the case of many-body fermion systems, care must be taken of the chronological ordering which requires, first, to restore the order resulting from the action of commutators. This is easy to do by accounting that the symbol δV implies the permutation of

V with the operators on the right, whereas $V\delta$ does not. For any operator $\mathbb{A}$ of the Liouville space, we thus have

$$\langle nm|(\delta V)\mathbb{A}|cb\rangle=\delta_l^n V_m^k \mathbb{A}_{ck}^{lb}=\mathbb{A}_{ck}^{nb} V_m^k \ ,$$
$$\langle nm|(V\delta)\mathbb{A}|cb\rangle=V_l^n \delta_m^k \mathbb{A}_{ck}^{lb}=V_l^n \mathbb{A}_{cm}^{lb} \ . \tag{21}$$

The density-matrix solution is now given in a fractional form which can be shown to be trace conserving.[6] At first sight, it seems more attractive than a perturbation series. In fact, we are going to see that this is not so, to the first reduction stage. To lowest orders, the denominator in Eq. (20) can be written, upon expansion, as

$$1+\langle cb|\mathbb{S}_{cb}^{-1}\mathbb{d}^{-1}\mathbb{K}_F|cb\rangle=1+\frac{1}{\mathbb{d}_{cb}}\left[\frac{V_z^c F_c^z}{\mathbb{d}_{zb}}+\frac{F_z^b V_b^z}{\mathbb{d}_{cz}}\right]+\frac{1}{\mathbb{d}_{cb}}\left[\frac{|F_c^z|^2}{\mathbb{d}_{zb}}+\frac{|F_b^z|^2}{\mathbb{d}_{cz}}\right]+\cdots \ .$$

Except for the first one, all terms on right-hand side of this equation involve F to increasing orders. These terms represent the natural width of the transitions (collision assisted or not) induced by the applied field and, therefore, they are extremely small at vanishing values of the field. As a result, the fraction in Eq. (20) reduces to its numerator, which can be readily verified to be very close to a perturbation series of the density matrix, in increasing powers of both K_V and K_F. This is the reason why, to get a nontrivial result, we have to continue the same process one step further, by regarding, in turn, the matrix elements arising in the numerator of Eq. (20) as reduced solutions of the following system:

$$\begin{aligned}\mathbb{S}_{cb}|\mathbb{X}\rangle&=\mathbb{d}^{-1}\mathbb{K}_F|cb\rangle\rho_{0b}^c\\&=\mathbb{d}_{c_1b_1}^{-1}|c_1b_1\rangle\langle c_1b_1|\mathbb{K}_F\rangle\rho_{0b}^c\\&=\mathbb{d}_{zb}^{-1}K_{Fc}^z\rho_{0b}^c|zb\rangle\\&\quad-\mathbb{d}_{cz}^{-1}\rho_{0b}^c K_{Fz}^b|cz\rangle \ .\end{aligned} \tag{22}$$

The projector $\sum_{c_1,b_1}|c_1b_1\rangle\langle c_1b_1|$ has been inserted and use has been made of the definition (8) and the rule (21). The solution is obtained the same way as before,

$$\mathbb{X}^{\mu}{}_{\lambda}(cb)=\frac{\mathbb{D}_{z\lambda}^{\mu b}(\mathbb{D}_{zb}^{zb})^{-1}}{\mathbb{d}_{zb}\mathbb{D}(\mathbb{D}_{zb}^{zb})^{-1}}K_{Fc}^z\rho_{0b}^c-\frac{\mathbb{D}_{c\lambda}^{\mu z}(\mathbb{D}_{cz}^{cz})^{-1}}{\mathbb{d}_{cz}\mathbb{D}(\mathbb{D}_{cz}^{cz})^{-1}}\rho_{0b}^c K_{Fz}^b \ . \tag{23}$$

The bracket (cb) reminds us that this $\mathbb{X}$ solution is written assuming cb to be the starting state. Determinants and minors pertain to the $\mathbb{S}_{cb}$ matrix, and can be written again in terms of single matrix elements involving geometrical series,[10]

$$\mathbb{X}^{\mu}{}_{\lambda}(cb)=-\frac{\langle\mu\lambda|\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\mathbb{T}_{zb}^{-1}|zb\rangle}{\mathbb{d}_{zb}+\langle zb|(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\mathbb{T}_{zb}^{-1}|zb\rangle}K_{Fc}^z\rho_{0b}^c+\frac{\langle\mu\lambda|\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\mathbb{T}_{cz}^{-1}|cz\rangle}{\mathbb{d}_{cz}+\langle cz|(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb})\mathbb{T}_{cz}^{-1}|cz\rangle}\rho_{0b}^c K_{Fz}^b \ , \tag{24}$$

with

$$\mathbb{T}_{zb}=\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}(\mathbb{K}_V+\mathbb{K}_F\mathbb{Q}_{cb}), \quad \cdots \ . \tag{25}$$

The result (24) holds for $\mu\lambda\neq zb,cz$. For $\mu\lambda=zb$ or cz, the related numerator reduces to 1, as immediately seen in Eq. (23). Finally, making use of the $\mathbb{X}$'s as given by Eq. (24), the solution (20) becomes

$$\mathbb{R}^{\mu}{}_{\lambda}(\nu)=\sum_{c,b\neq\mu,\lambda}\frac{\mathbb{X}^{\mu}{}_{\lambda}(cb)}{\nu[1+\mathbb{X}^c{}_b(cb)]}\cong\nu^{-1}\sum_{c,b\neq\mu,\lambda}\mathbb{X}^{\mu}{}_{\lambda}(cb) \ , \tag{26}$$

which again leads to a well defined steady-state value of ρ_{λ}^{μ}.

It is worth noticing that although both the collision potential and the external field are similarly regarded as perturbations on the state basis of H_0, all final transition rates are direct functions of the external field F (through the kernel $\mathbb{K}_F$), i.e., they vanish if $F=0$, as shown in Eq. (24). This, of course, results from the inclusion of the commutation relation (5) [or (15)] in the formalism, which gives rise to the particular form of the S operator as given by Eqs. (18), which $\mathbb{K}_F$ and $\mathbb{K}_V$ does not enter symmetrically.

In the linear-response theory the field can be dropped inside the brackets ($\langle\ |\ |\ \rangle$) of Eq. (24). We are thus left with the final twice-reduced result of the steady-state density matrix ($t\to\infty$):

$$\rho_{\lambda}^{\mu}=-\frac{\langle\mu\lambda|\mathbb{d}^{-1}\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}{\mathbb{d}_{zb}+\langle zb|\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}K_{Fc}^z\rho_{0b}^c+\frac{\langle\mu\lambda|\mathbb{d}^{-1}\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{cz}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|cz\rangle}{\mathbb{d}_{cz}+\langle cz|\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{cz}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|cz\rangle}\rho_{0b}^c K_{Fz}^b \ . \tag{27}$$

Let us recall that this expression must be understood in limit where $\nu\to 0+$, with the index restriction $\mu\lambda\neq zb,cz$. We note that, because of the conjugation relation in Liouville space,[6]

$$\langle\mu\lambda|\mathbb{A}|cb\rangle=\langle\lambda\mu|\mathbb{A}|bc\rangle^* \ ,$$

the second term on the right-hand side in Eq. (27) is conjugate to the first one through permutation between λ and μ. This is readily seen upon exchanging the dummy indices b and c in the second term.

As observed, the calculation of ρ_{λ}^{μ} requires knowledge of nondiagonal matrix elements ρ_{0b}^c of ρ_0. This, of course, is a consequence of dealing with the unperturbed basis of H_0 instead of that of H_0+V, which is not known. In the widespread random phase approximation, nondiagonal matrix elements of ρ_0 are ignored. Then, upon interchanging the dummy indices b,z in the second term on the right-hand side, Eq. (27) can be given the more compact form

$$\rho_{\lambda}^{\mu}=-\frac{\langle\mu\lambda|\mathbb{d}^{-1}\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}{d_{zb}+\langle zb|\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}\times(K_{Fb}^z\rho_{0b}^b-\rho_{0z}^z K_{Fb}^z) \ . \tag{28}$$

An alternative way which seems physically valid consists of assuming that the correct long-term limit of the density matrix can be obtained as well if V and F are *both* applied to the system at $t=0$. This point of view, which was adopted, for instance, by Dumke[11] in his early work on the intraband infrared absorption in semiconductors, has, of course, the great advantage of allowing us to use directly the unperturbed density matrix, at $t=0$. Care must be taken, however, because in doing so we are not quite sure how to avoid introducing spurious contributions into the solution.

Formulas (20) and (24)–(27) are quite general. In many applications, many-body states must be considered. In the case of an independent-particle system in which, in addition, the correlation between individual motion resulting from the external applied field and the collision potential can be neglected, individual states entering the collective initial states can be treated separately. This amounts to ignoring coupling between the particle under consideration and the other particles that will give rise, necessarily, to subsequent factorizations in the starting solution (13). This is completely equivalent to saying that the system (16) is broken into one-particle–independent systems, to every one of which the above reduction procedure can be applied identically.

V. APPLICATION OF THE THEORY OF ELECTRICAL dc CONDUCTIVITY

Since the early work by Kohn and Luttinger,[12] and Lax,[13] there have been a great many theoretical studies on the quantum theory of electrical conductivity, a large part of which is based on the Kubo formalism. The purpose of this section is not to give a critical discussion of these previous approaches, as compared with the present one, but simply to test how the above general formulas work, in a simplified case.

We shall consider a system of quasifree electrons of mass m, and with an individual energy $\varepsilon(\mathbf{k})=\hbar^2k^2/2m$, corresponding to an isotropic and parabolic band. The unperturbed one-electron eigenstates are plane waves, normalized to the volume $\mathcal{V}$ of the system,

$$\langle \mathbf{r}|\mathbf{k}\rangle=\mathcal{V}^{-1/2}\exp(i\mathbf{k}\cdot\mathbf{r}) .$$

The electrons are subjected to the collision potential V of zero diagonal elements. Spin is ignored. An external electric field E_x is applied along the x axis, from $t=0$.

In the momentum representation the current-density operator is diagonal and given, in a second-quantization scheme, by

$$\mathbf{J}=-(e\hbar/\mathcal{V}m)\mathbf{k}^{(c)}=-(e\hbar/\mathcal{V}m)\sum_k \mathbf{k}c_k^\dagger c_k . \tag{29}$$

The superscript (c) is used to distinguish the collective from the individual $\mathbf{k}$. Similarly, the field kernel K_F is given by

$$K_F=i\hbar^{-1}eE_x x^{(c)}=i\hbar^{-1}eE_x\sum_{k,k'} x_{k'}^k c_k^\dagger c_{k'} . \tag{30}$$

The individual matrix elements of x between plane waves,

$$x_{k'}^k=\mathcal{V}^{-1}\int \exp(-i\mathbf{k}\cdot\mathbf{r})x\exp(i\mathbf{k}\cdot\mathbf{r})d\mathbf{r}$$
$$=-i\mathcal{V}^{-1}\nabla_{k_x'}\int\exp[-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{r}]d\mathbf{r} ,$$

are conveniently written with a δ function,

$$x_{k'}^k=-i(8\pi^3/\mathcal{V})\nabla_{k_x'}\delta(\mathbf{k}-\mathbf{k}') . \tag{31}$$

From Eq. (27), it is straightforward to write the σ_{xx} component of the conductivity:

$$\sigma_{xx}=\mathrm{Tr}(J_x\rho)/E_x=J_{x\lambda}^{\lambda}\frac{\delta_z^\lambda\delta_\lambda^b-\langle\lambda\lambda|\mathbb{d}^{-1}\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}{\mathbb{d}_{zb}+\langle zb|\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}\frac{K_{F_c^z}}{E_x}\rho_{0b}^c+\text{c.c.}$$

Substituting, next, J_x and K_F from Eqs. (29) and (30), we have

$$\sigma_{xx}=-\frac{ie^2k_{x\lambda}^{\lambda}}{\mathcal{V}m}\frac{\delta_z^\lambda\delta_\lambda^b-\langle\lambda\lambda|\mathbb{d}^{-1}\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}{\mathbb{d}_{zb}+\langle zb|\mathbb{K}_V(\mathbb{I}+\mathbb{Q}_{zb}\mathbb{d}^{-1}\mathbb{K}_V)^{-1}|zb\rangle}x_c^z\rho_{0b}^c+\text{c.c.} \tag{32}$$

The particular zero-order term, $\lambda\lambda=zb$, is separated out in the numerator [which then reduces to 1 as shown in Eq. (23)], whereas the restriction $\lambda\lambda\neq zb$ is implied in the brackets ($\langle\,|\,|\,\rangle$). Since, on the other hand, the brackets ($\langle\,|\,|\,\rangle$) in the denominator starts from second order, to lowest order Eq. (32) reduces to

$$\sigma_{xx}=-\frac{ie^2}{\mathcal{V}m}\frac{[k_x^{(c)}]_b^b x_c^b\rho_{0b}^c}{\tau^{-1}-\langle bb|\mathbb{K}_V\mathbb{Q}_{bb}\mathbb{d}^{-1}\mathbb{K}_V|bb\rangle}+\text{c.c.} \tag{33}$$

Since, from expression (30), x is a sum of one-electron operators, there only remains in the numerator of the fraction (33) matrix elements of ρ_0 between b and c states which differ from each other by a one-electron transition, say $\mathbf{k}\rightarrow\mathbf{k}'$. This can be expressed by

$$|c\rangle=c_{k'}^\dagger c_k|b\rangle .$$

ρ_{0b}^c is, otherwise, necessarily diagonal with respect to any individual state distinct from $\mathbf{k},\mathbf{k}'$. Thus, one can write in a simplified form

$$\rho_{0b}^{c}=\rho_0(\mathbf{k}',\mathbf{k}) . \tag{34}$$

In the present case, assuming a large number of empty individual states in the band, electrons can be regarded as undergoing V transitions independent from one another. Correlation effects in electron motion are negligible and, according to the remark at the end of the preceding section, the one-electron point of view can be adopted. Thus, the initial state will be broken up into individual contributions involving only one active electron, say that occupying a given $\mathbf{k}$ state at $t=0$, and every other one remains "quenched" in its initial state. Let us first write out, accordingly, the resulting expression of the denominator in Eq. (33). Making use again of the definition (8) and the ordering rule (21), we obtain

$$\begin{aligned}-\langle bb|\mathbb{K}_V\mathbb{Q}_{bb}\mathbb{d}^{-1}\mathbb{K}_V|bb\rangle&=\hbar^{-2}(V_{c_1}^{b}\delta_{b}^{b_1}-\delta_{c_1}^{b}V_{b}^{b_1})\mathbb{d}_{c_1b_1}^{-1}(V_{b}^{c_1}\delta_{b_1}^{b}-\delta_{b}^{c_1}V_{b}^{b})\\&=\hbar^{-2}(V_{c_1}^{b}V_{b}^{c_1}/\mathbb{d}_{c_1b}+V_{b_1}^{b}V_{b}^{b_1}/\mathbb{d}_{bb_1}) .\end{aligned}$$

The index restriction $c_1b_1\neq bb$ is automatically satisfied by the assumption that diagonal matrix elements are missing. Since only the $\mathbf{k}$ state is relevant, this can be more explicitly written as follows:

$$-\langle bb|\mathbb{K}_V\mathbb{Q}_{bb}\mathbb{d}^{-1}\mathbb{K}_V|bb\rangle=\hbar^{-2}\sum_{k_1}|V_k^{k_1}|^2/(\tau^{-1}+i\omega_{k_1k})+\text{c.c.} \tag{35}$$

The detailed value of (35), which represents twice the relaxation frequency, depends on both the collision potential and the dispersion law in the band. As pointed out above, as far as the electron relaxation is strongly dominated by the V processes, the τ^{-1} frequency can be regarded as an infinitely small quantity, thereby giving the summation in Eq. (35) the form of a Cauchy-type integral.[9]

Now, from Eqs. (29)–(31) and the definition (34), expression (33) of the σ_{xx} conductivity becomes

$$\sigma_{xx}=-\frac{e^2}{\mathcal{V}m}\frac{8\pi^3}{\mathcal{V}}\sum_{k,k'}\frac{k_x[\nabla_{k_x'}\delta(\mathbf{k}-\mathbf{k}')]\rho_0(\mathbf{k}',\mathbf{k})+\text{c.c.}}{\tau^{-1}+2\hbar^{-2}\mathrm{Re}\sum_{k_1}|V_k^{k_1}|^2/(\tau^{-1}+i\omega_{kk_1})} . \tag{36}$$

The integration over $\mathbf{k}'$ is easily carried out by parts

$$\frac{8\pi^3}{\mathcal{V}}\sum_{k'}[\nabla_{k_x'}\delta(\mathbf{k}-\mathbf{k}')]\rho_0(\mathbf{k}',\mathbf{k})=-\int[\nabla_{k_x'}\rho_0(\mathbf{k}',\mathbf{k})]\delta(\mathbf{k}-\mathbf{k}')d\mathbf{k}'=-[\nabla_{k_x'}\rho_0(\mathbf{k}',\mathbf{k})]_{k'=k} .$$

Hence, ignoring again τ^{-1} as compared with the V-collision relaxation frequency defined by

$$\tau_V^{-1}(\mathbf{k})=\hbar^{-2}\mathrm{Re}\sum_{k_1}|V_k^{k_1}|^2/(\tau^{-1}+i\omega_{k_1k}) , \tag{37}$$

we found the Boltzmann-like result

$$\sigma_{xx}=(e^2/\mathcal{V}m)\sum_{k}k_x\tau_V(\mathbf{k})[\nabla_{k_x'}\rho_0(\mathbf{k}',\mathbf{k})]_{k'=k} . \tag{38}$$

To proceed further, the knowledge of nondiagonal matrix elements of the equilibrium density matrix is needed. Second-quantization expressions can be derived in the form of expansion with respect to the commutator $[H_0,V]$. It is physically intuitive that the result of such a study,[14] which is out of the scope of the present illustration, involves to lowest order the unperturbed collective density matrix which $\mathbf{k}$ and $\mathbf{k}'$ enter through the Fermi occupation factors, say $f(\mathbf{k})[1-f(\mathbf{k}')]$. The predominant contribution to the gradient in Eq. (38) is thus expected to be

$$-\nabla_{k_x'}f(\mathbf{k}')|_{k'=k}=-\hbar v_{k_x}\partial f(\varepsilon_k)/\partial\varepsilon_k ,$$

leading to the usual result

$$\sigma_{xx}=-(\hbar e^2/\mathcal{V}m)\sum_{k}K_xv_{kx}\tau_V(\mathbf{k})\frac{\partial f(\varepsilon_k)}{\partial\varepsilon_k} . \tag{39}$$

$\mathbf{v}_k=\hbar^{-1}\nabla_k\varepsilon(\mathbf{k})$ stands for the electron group velocity.

Thus, the first-principles quantum theory directly yields a Boltzmann-type expression without using the

random-phase approximation, which is a basic assumption in the elementary derivation. The reasons for which the random-phase approximation is superfluous in a quantum treatment have been already discussed,[15,16] in connection with the Van Hove fundamental analysis.[17]

The question remains as to what occurs to higher orders of the collision potential in Eq. (32). Upon examination of the possible transitions to second, third order in V, . . . in the numerator, one finds a net trend of complete cancellation of all terms, as in the trace of a commutator. Thus the only change in the result (36) is likely to replace the second-order relaxation frequency τ_V^{-1} by a new expression of increasing order.

VI. CONCLUSIONS

The general theory previously elaborated has been applied to work out a steady-state solution of the Schrödinger equation, in transport problems. General expressions involving the applied field and the collision potential to increasing orders have been derived. Substantial simplifications are obtained in the case of linear response, and also by taking carefully into account that natural broadening is often completely negligible compared to collision broadening. To some extent, the contribution of the collision potential to the initial thermodynamical equilibrium is also taken into account.

The theory is capable, as well as Kubo's formalism, to handle with the relevant physical quantities taken in their quantum nature. It has, however, the advantage of straightfowardly yielding a trace-conserving form of the response from first principles, using quite elementary and simple mathematics. This is the main reason that relaxation effects arise in a simple and natural way. The fractional form permits one, in addition, to overcome difficulties associated with divergences such as those occurring in the old problem of the cyclotron-resonance linewidth in the quantum limit. A preliminary report about it has been published.[18] A treatment of the quantum Hall effect should likely constitute another fairly attractive test of the theory. Finally, higher orders of the collision potential together with nonlinear effects in the applied field could be included.

In this paper some of the above capabilities of the method have been examplified in a simplified illustration dealing with the quantum theory of dc conductivity, in which the expression derived from the elementary Boltzmann theory can be recovered.

[1]R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
[2]K. M. van Vliet, J. Math. Phys. **19**, 1345 (1978).
[3]N. G. Kempen, Phys. Norv. **5**, 279 (1971).
[4]See, for example, the criticisms of theories based on the Kubo formalism, in P. N. Argyres and J. L. Sigel, Phys. Rev. B **10**, 1139 (1974); and P. N. Argyres, Solid State Commun. **50**, 279 (1984).
[5]A. Fortini, Phys. Rev. Lett. **53**, 1125 (1984).
[6]A. Fortini, J. Phys. A **16**, 3987 (1983); **17**, 2641 (1984).
[7]U. Fano, Rev. Mod. Phys. **29**, 74 (1957).
[8]B. Balian, Y. Alhassid, and H. Reinhart, Phys. Rep. **131**, 1 (1986).
[9]V. I. Smirnov, *A Course on Higher Mathematics,* edited by I. N. Sneddon (Pergamon, New York, 1964), Vol. III.
[10]A. Fortini, Phys. Rev. A **23**, 395 (1981).
[11]W. P. Dumke, Phys. Rev. **124**, 1813 (1961).
[12]W. Kohn and J. M. Luttinger, Phys. Rev. **108**, 590 (1957).
[13]M. Lax, Phys. Rev. **109**, 1921 (1958).
[14]J. Des Cloizeaux, in *Many-Body Physics,* edited by C. De Witt and R. Balian (Gordon and Breach, New York, 1968).
[15]D. A. Greenwood, Proc. Phys. Soc. London **71**, 585 (1958).
[16]G. V. Chester and A. Tellung, Proc. Phys. Soc. London **73**, 745 (1959).
[17]L. Van Hove, Physica **21**, 517 (1955).
[18]A. Fortini, in *Proceedings of the 18th International Conference on the Physics of Semiconductors,* edited by Olof Engström (World Scientific, Singapore, 1987), Vol. 2, p. 1157.