Superoperator theory of high-field transport for an electron-phonon system

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We have developed a quantum-statistical theory of high-field transport based on Tani's theory of nonlinear response. An exact closed-form expression for a steady-state nonlinear current is obtained in Ohm's-law form, from which a field-dependent dc conductivity is defined. This conductivity formula is of a form similar to the Kubo formula in the linear-response theory, but is dependent on the field. We present a resolvent superoperator approach for the evaluation of this field-dependent conductivity (i.e., a field-dependent current-current correlation integral formula) of an interacting electron-phonon system. The nonlinear effects due to the presence of a high field are reflected directly in the self-energy superoperator. The field-dependent conductivity is expressed in terms of a self-energy superoperator and an expression is obtained for this temperature- and field-dependent quantity. In this approach "collisional broadening" and the "intra-collisional-field effect" appear quite naturally. These quantum effects are illustrated by the phenomena of the level broadening due to the relaxation and the accelerating effects.

I. INTRODUCTION

The transport of electrons in very large electric (and magnetic) fields has recently been one of the subjects of numerous theoretical and experimental investigations[1–20] due to the remarkable advance in the techniques of crystal growth and device processing which affords a new type of conduction mechanism in small solid-state structures, those having a submicrometer dimension. When the energy absorbed from the field is comparable to the characteristic phonon energies, the electrons become hot. Under such extremely nonequilibrium situations, the validity of the semiclassical Boltzmann equation, on which most theoretical studies[2] are based, is questionable. Since high fields alter basically the quantum states of carriers and their energy spectrum, a complete quantum description requires a theory of model based on quantum-transport theory, which differs from the Boltzmann type of model. Many theoretical attempts, both analytical[4–17] and numerical,[18–20] have been made toward developing a high-field–transport theory which would be capable of including a variety of high-field effects as well as bulk and interface properties in submicrometer structures. Through these intense theoretical studies, new phenomena of high-field transport are indeed predicted. Among these, "collisional broadening" (CB) and the "intracollisional field effect" (ICFE), originally predicted by Barker[3] in an effort to derive the steady-state Boltzmann high-field equation, have perhaps attracted most of the attention.

In this paper, we shall present a first-principles high-field quantum-transport theory based on the framework of Tani's theory[4–6] of nonlinear response, which could, in principle, account for a wide variety of high-field properties such as CB, ICFE, and for magnetophonon effects, and intervalley scatterings under intense electromagnetic fields. Based on the method of Tani, the closed-form formal expression for the steady-state current in Ohm's-law form with a field-dependent dc conductivity will be derived. The purpose of the present paper is to show how these effects (CB and ICFE) are formulated into the formal expression of the field-dependent dc conductivity without recourse to a relaxation approach[2,13] used in the quantum-transport theory when it is applied to an interacting electron-phonon system, and to show a detailed method of practical calculation of this field-dependent conductivity.

Our method is based on the resolvent formalism applied to quantum-statistical theory. Although one has to work in a superspace of operators, the use of superoperators in the theory of high-field transport does offer a distinct advantage in the fact that characteristic "gain-loss" relaxive structure associated with nonvanishing vertex corrections and the effect of the field within a collision process are built into formalism in a simple fashion. This gives "collisional broadening," which is normally included in a phenomenological way, and the spectral density is dependent on the electric field since CB is induced from the scattering processes in which the effect of the field within a collision is taken into account in the present theory. Therefore, the spectral density contains information about the "intracollisional field effect" and other transport properties of the carriers influenced by the strong electric (and magnetic) fields as well as by scatterers (phonons). We employ a factorization approximation for the equilibrium statistical operator [Eq. (2.20)] and calculate the field-dependent current correlation function [Eq. (2.16)] using the grand-canonical distribution for the electrons and the canonical distribution for phonons. Thus, any collision process between an electron and phonons is assumed to take place in the average field of the phonons. The effect of such a field is to induce perturbed single-particle energies and to introduce lifetimes (which have a dependence of the field) for the electron states. The lifetime broadening due to interactions and the field is, for example, responsible for the spectral broadening of the line shapes and can be studied by examining the real part of the conductivity tensor. The part of the broaden-

ing due to ICFE is, in practice, ignored in the low-field transport theory. We relate the field-dependent conductivity to a self-energy (or collision) superoperator. Hence it is important to provide a general expression for the temperature- and field-dependent self energy. In this paper we show how to do this in a self-consistent manner. The theory is developed independently of the single-particle representation, and hence can be applied irrespective of the system studied. In particular, it is valid for systems subject to a constant magnetic field and the differences due to **B** arise only at the final stage of calculation.

This paper is organized as follows. In Sec. II, we first derive the closed-form expression for the steady-state current in the Ohm's-law form with a field-dependent dc conductivity in the framework of Tani's theory of nonlinear response. Based on the expression, the field-dependent Kubo-type conductivity formula is obtained, and is applied to the special case of an electron-phonon system modeled by Hamiltonians given by Eqs. (2.1)–(2.3), and the general theoretical method of its evaluation is outlined. The one-electron field-dependent resolvent superoperator is given in terms of a simpler effective one-electron field-dependent resolvent R_F^D. This depends on the field-dependent self-energy (or collision) superoperator Σ_F^D, which is defined at the outset unlike previous theories[3,7] which are based on the perturbation expansion of the resolvent superoperators and hence depend heavily on the potential and the field strengths. Here we see the effects of the fields in the relaxation processes (ICFE) are included in Σ_F^D and so the collisional (lifetime) broadening (CB) due to the scatterings and the applied fields are formulated in the theory. In Sec. III, the general technique for dealing with superoperators is briefly presented. The general expression for the temperature- and the field-dependent self-energy is evaluated to lowest order in the electron-phonon interaction. Using this expression, we shall obtain the field-dependent conductivity formula for an electron-phonon system. In the last section, a summary and concluding remarks are given.

II. FORMAL PRELIMINARIES

A. The model Hamiltonian and the exact field-dependent static conductivity

We consider an electron-phonon system characterized by the following time-independent Hamiltonian:

$$H=\sum_l h^{(l)}+H_{\rm ph} \ , \tag{2.1}$$

$$h^{(l)}=h_e^{(l)}+\lambda V^{(l)}=h_e^{(l)}+\lambda\sum_{\mathbf{q}}(\gamma_{\mathbf{q}}^{(l)}b_{\mathbf{q}}+\gamma_{\mathbf{q}}^{\dagger(l)}b_{\mathbf{q}}^{\dagger}) \ , \tag{2.2}$$

$$H_{\rm ph}=\sum_{\mathbf{q}}\hbar\omega_{\mathbf{q}}(b_{\mathbf{q}}^{\dagger}b_{\mathbf{q}}+\tfrac{1}{2}) \ , \tag{2.3}$$

where h_e is the single-particle energy operator, $V=\sum(\gamma_{\mathbf{q}}b_{\mathbf{q}}+\gamma_{\mathbf{q}}^{\dagger}b_{\mathbf{q}}^{\dagger})$ is the interaction potential between an electron and a phonon with momentum $\mathbf{q}$, $b_{\mathbf{q}}$ and $b_{\mathbf{q}}^{\dagger}$ are, respectively, the phonon annihilation and creation operators, $\gamma_{\mathbf{q}}$ $[=C(q)\exp(i\mathbf{q}\cdot\mathbf{r})]$ is the *screened* interaction (one-electron) operator, and λ is a dimensionless expansion parameter which is set equal to 1 later on. It is noted that potential form $C(q)$ depends on the type of interaction. Here, $H_{\rm ph}$ stands for the phonon Hamiltonian and $\omega_{\mathbf{q}}$ is the frequency of a phonon. The energy operator h_e represents the electron Hamiltonian in the one-band approximation with an allowance for a magnetic field **B** $(=\nabla\times\mathbf{A})$.

To derive a closed-form expression for the steady-state nonlinear current (and hence the nonlinear conductivity tensor), we consider the case where a uniform static electric field **F** of arbitrary strength is applied adiabatically from an initial time $t=-\infty$. The disturbance of the system due to the applied field is then represented by the additional Hamiltonian

$$H_F(t)=e\mathbf{F}\cdot\mathbf{R}\exp(\varepsilon t/\hbar)=H_F\exp(\varepsilon t/\hbar), \quad 0<\varepsilon\ll 1 \ , \tag{2.4}$$

where $-e\mathbf{R}$ is the many-electron polarization operator and H_F is given by

$$H_F=e\mathbf{R}\cdot\mathbf{F}=\sum_l e\mathbf{r}^{(l)}\cdot\mathbf{F}=\sum_l h_F^{(l)} \ . \tag{2.5}$$

The density matrix $\rho(t)$ of the disturbed system follows the equation of motion (Liouville equation):

$$i\hbar\frac{\partial\rho(t)}{\partial t}=[\hat{H}+\hat{H}_F(t)]\rho(t) \ . \tag{2.6}$$

Here carets denote the commutator-generating superoperators upon acting on ordinary quantum operators such that

$$\hat{O}A\equiv[O,A]=OA-AO \ . \tag{2.7}$$

In common with other approaches, the starting point is the solution of the Liouville equation (2.6) for the time evolution of the density matrix. Before we turned on the field, the system was initially in equilibrium state at a temperature T $[\equiv(k_B\beta)^{-1}]$:

$$\rho(-\infty)=\rho_{\rm eq}(H)=\exp[\beta(\zeta N-H)]/{\rm Tr}\{\exp[\beta(\zeta N-H)]\} \ , \tag{2.8}$$

where ζ is the chemical potential and N is the electron-number operator. At any later time, the time evolution of the density matrix may be described by

$$\rho(t)=\rho_{\rm eq}+\rho'(t) \ , \tag{2.9}$$

in which $\rho'(t)$ represents the deviation from equilibrium ensemble $\rho_{\rm eq}$ and is not necessarily small. Equation (2.6) is thus reduced to

$$d\rho'(t)/dt+(i/\hbar)(\hat{H}+\hat{H}_F)\rho'(t)=-(i/\hbar)\hat{H}_F\rho_{\rm eq} \ . \tag{2.10}$$

It should be noted that we keep the term $(i/\hbar)\hat{H}_F\rho'(t)$ since the electric field is not necessarily small and so Eq. (2.10) is exact. Noting that $d\rho_{\rm eq}/dt=0$ and $\rho'(-\infty)=0$, we have for the steady-state density matrix ρ_s at $t=0$ by switching adiabatically on the perturbation (2.4) from an infinite past a solution as

$$\rho_s=\rho_{\rm eq}+\frac{1}{i\hbar}\lim_{\varepsilon\to 0^+}\left[\int_{-\infty}^{0}dt[\hat{H}+\hat{H}_F(t)]\rho(t)\right]$$
$$=\rho_{\rm eq}+\frac{1}{i\hbar}\lim_{\varepsilon\to 0^+}\left[\int_{-\infty}^{0}dt\,\exp(\varepsilon t/\hbar)\exp[i(\hat{H}+\hat{H}_F)t/\hbar]\hat{H}_F\rho_{\rm eq}\right]. \tag{2.11}$$

The total steady current is then exactly obtained from

$$\langle\mathbf{J}\rangle={\rm Tr}(\rho_s\mathbf{J})$$
$$=\lim_{\varepsilon\to 0^+}\int_0^{\infty}dt\,\exp(-\varepsilon t/\hbar)\int_0^{\beta}d\beta_1\,{\rm Tr}[\rho_{\rm eq}\mathbf{J}(-i\hbar\beta_1|H)\mathbf{J}(t|H+H_F)]\mathbf{F}\,, \tag{2.12}$$

since ${\rm Tr}(\rho_{\rm eq}\mathbf{J})=\mathbf{0}$. Here the symbol Tr denotes a many-body trace. To derive Eq. (2.12), we have used the Kubo identity.[21]

$$\frac{1}{i\hbar}\hat{H}_F\rho_{\rm eq}=-\rho_{\rm eq}\int_0^{\beta}d\beta_1\exp(\beta_1 H)\dot{H}_F\exp(-\beta_1 H)$$
$$=\rho_{\rm eq}\int_0^{\beta}d\beta_1\,\mathbf{J}(-i\hbar\beta_1|H)\cdot\mathbf{F} \tag{2.13}$$

with the relation

$$\dot{H}_F=e\dot{\mathbf{R}}\cdot\mathbf{F}=-\mathbf{J}\cdot\mathbf{F}=-\sum_l\mathbf{j}^{(l)}\cdot\mathbf{F}\,. \tag{2.14}$$

Here the overhead dot denotes a time derivative. It should be noted that $\mathbf{J}(t|H)$ is a total current operator in the Heisenberg picture:

$$\mathbf{J}(t|H)\equiv\exp(iHt/\hbar)\mathbf{J}\exp(-iHt/\hbar),\quad \mathbf{J}=\sum_l\mathbf{j}^{(l)}\,, \tag{2.15}$$

where $\mathbf{j}$ is a single-electron current operator in the one-hand approximation and is defined by the velocity operator multiplied by the electric charge $-e$. It should be noted that Eq. (2.12) expresses an autocorrelation function of the field-dependent current operator $\mathbf{J}(t|H+H_F)$ and the zero-field form of the extended current operator $\mathbf{J}(-i\hbar\beta|H)$, which is obtained with substitution for $t=-i\hbar\beta$ in Eq. (2.15). From Eq. (2.12), the formal expression for the generalized (exact field-dependent) conductivity tensor $\sigma_{rs}(F_s)$ may be defined through[4] $\langle J_r\rangle/\Omega=\sigma_{rs}(F_s)F_s$ $(r,s=x,y,z)$ as

$$\sigma_{rs}(F_s)=\Omega^{-1}\lim_{\varepsilon\to 0^+}\left[\int_0^{\infty}dt\,\exp(-\varepsilon t/\hbar)\int_0^{\beta}d\beta_1\,{\rm Tr}[\rho_{\rm eq}J_s(-i\hbar\beta_1|H)J_r(t|H+H_F)]\right], \tag{2.16}$$

where Ω represents the volume. It should be noted that Eq. (2.16) is a functional of an applied field $\mathbf{F}$. The linear conductivity is readily found by taking the Ohmic limit $\mathbf{F}\to\mathbf{0}$ in Eq. (2.16) to give the Kubo formula[21] for the static conductivity. Equation (2.16) has an exact inclusion of the field—which has more physical relevance in a nonlinear region or for large fields, and hence can be applied to the study for high-field quantum-transport problems. Let us introduce the fictitious Hamiltonian $\tilde{H}\equiv H-\mathbf{u}\cdot\mathbf{J}$ ($\mathbf{u}$ is a c-number vector). The further simplification is effected by utilizing an identity:[22]

$$\int_0^{\beta}d\beta_1\rho_{\rm eq}(H)\mathbf{J}(-i\hbar\beta_1|H)=\lim_{\mathbf{u}\to\mathbf{0}}\left[\frac{\partial}{\partial u_s}\rho_{\rm eq}(\tilde{H})\right]. \tag{2.17}$$

Substituting Eq. (2.17) into (2.16), the field-dependent conductivity formula (2.16) is reduced to a more compact form:

$$\sigma_{rs}(F_s)=\lim_{\varepsilon\to 0^+}\lim_{\mathbf{u}\to\mathbf{0}}\frac{\partial}{\partial u_s}\left[\Omega^{-1}\int_0^{\infty}dt\,\exp(-\varepsilon t/\hbar){\rm Tr}[\rho_{\rm eq}(\tilde{H})J_r(t|H+H_F)]\right], \tag{2.18}$$

where $\rho_{\rm eq}(\tilde{H})$ is given by replacing H by $\tilde{H}$ in Eq. (2.8). So far we have made no approximation to derive Eqs. (2.16) and (2.18), which will be used in the study of high-field transport.

B. Evaluation of $\sigma_{rs}(F_s)$

In the following, we will show a detailed method of practical calculation of the field-dependent conductivity (2.18) for an electron-phonon system modeled by the Hamiltonians (2.1)–(2.3). We assume that the phonons are distributed all the time according to the canonical distribution law:

$$\rho_{\rm ph}=\rho_{\rm ph}(H_{\rm ph})$$
$$\equiv\exp(-\beta H_{\rm ph})/{\rm Tr}^{\rm (ph)}[\exp(-\beta H_{\rm ph})]\,, \tag{2.19}$$

and further assume that the following factorization is allowed:[16–20,23,24]

$$\rho_{\rm eq}(\tilde{H})\approx\rho_{\rm ph}(H_{\rm ph})\otimes\rho\left[\sum_l h^{(l)}-\mathbf{u}\cdot\mathbf{J}\right]. \tag{2.20}$$

In Eq. (2.19), the symbol ${\rm Tr}^{\rm (ph)}$ denotes a trace over phonon coordinates. This is equivalent to the assumption that the phonons are part of the heat reservoir for the electron system and are in contact with another huge heat reservoir. Therefore the system we consider here is

one in which phonons are always in thermal equilibrium with the outer world, so that the phonon (lattice) temperature is being kept constant. This means that the amount of energy which is transferred to the lattice from the conduction electrons is quite small, so that the disturbance to the lattice through the interaction with the electrons is negligibly small. Therefore, this assumption is likely to be valid as long as the system has a low density of electrons, such as in a semiconductor. The approximation (2.20) and the assumption made for phonon distribution (2.19) are almost certainly invalid for a metal and for the extremely high-field regime where the effects of nonequilibrium heating of the phonon populations take place. In fact, experiments on hot electrons are usually performed under conditions that keep the lattice temperature practically equal to the original one during the time of measurement. Hence we will content ourselves with the ansatz[16–20,23,24] made for ρ_{eq} to obtain the approximate expression for the nonlinear conductivity (which, however, retains the field dependence exactly). Refinement of the theory and a pure many-body treatment will be left for future investigation. Substituting Eq. (2.20) into (2.18), the many-body trace Tr collapses into $\mathrm{tr}\cdot\mathrm{Tr}^{(\mathrm{ph})}$ and the exact field-dependent conductivity formula (2.18) can be expressed in terms of a single-electron trace (denoted by tr) as

$$\sigma_{rs}(F_s)=\lim_{\varepsilon\to 0^+}\lim_{\mathbf{u}\to 0}\frac{\partial}{\partial u_s}\left[\Omega^{-1}\int_0^\infty dt\,\exp(-\varepsilon t/\hbar)\mathrm{tr}\{j_r\langle\exp[-i(\hat{h}_{0F}+\lambda\hat{V}+\hat{H}_{\mathrm{ph}})t/\hbar]f\rangle_{\mathrm{ph}}\}\right], \tag{2.21}$$

where h_{0F} is defined by

$$h_{0F}\equiv h_e+h_F=h_e+e\mathbf{r}\cdot\mathbf{F}\ , \tag{2.22}$$

and the angular brackets $\langle\cdots\rangle_{\mathrm{ph}}$ denote the averaging over the phonon states, and f is the Fermi-Dirac operator given as

$$f\equiv\{\exp[\beta'(h_e+\lambda V-\mathbf{u}\cdot\mathbf{j}-\zeta)]+1\}^{-1}\ , \tag{2.23}$$

where $\beta'\equiv(k_B T_e)^{-1}$ and T_e is the electron temperature. It should be noted that the formula (2.21) is valid providing H does *not* include the Coulomb interaction. The integral in Eq. (2.21) can be easily performed and the nonlinear conductivity can be written as

$$\sigma_{rs}(F_s)=-i\hbar\lim_{\varepsilon\to 0^+}[\Omega^{-1}\mathrm{tr}(j_r\tilde{f}_F)]\ , \tag{2.24}$$

where

$$\begin{aligned}\tilde{f}_F&\equiv\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}\langle i\int_0^\infty dt\,\exp(-\varepsilon t)\exp[-i(\hat{h}_{0F}+\lambda\hat{V}+\hat{H}_{\mathrm{ph}})t]f\rangle_{\mathrm{ph}}\right]\\&=\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}\langle(\hat{h}_{0F}+\hat{H}_{\mathrm{ph}}+\lambda\hat{V}-i\varepsilon)^{-1}f\rangle_{\mathrm{ph}}\right].\end{aligned} \tag{2.24a}$$

As can be seen from Eqs. (2.24) and (2.24a), evaluation of $\sigma_{rs}(F_s)$ is reduced to a calculation of the phonon-averaged (one-electron) quantum-statistical operator $\tilde{f}_F$ defined in the Laplace-transformed space, viz.,

$$\tilde{f}_F=\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}\langle(\hat{h}_{0F}+\hat{H}_{\mathrm{ph}}+\lambda\hat{V}-i\varepsilon)^{-1}f\rangle_{\mathrm{ph}}\right]\equiv\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}\langle R_F f\rangle_{\mathrm{ph}}\right], \tag{2.25}$$

where the field-resolvent superoperator is defined by

$$R_F\equiv(\hat{h}_{0F}+\hat{H}_{\mathrm{ph}}+\lambda\hat{V}-i\varepsilon)^{-1}\ . \tag{2.26}$$

Here the complete transport kinetics of electrons under the influence of the field is embodied in $\tilde{f}_F$ given by Eq. (2.25).

C. Evaluation of $\tilde{f}_F$

In order to evaluate $\tilde{f}_F$, let us consider the following identity:

$$\begin{aligned}\tilde{f}_F&=\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}\langle R_F f\rangle_{\mathrm{ph}}\right]\\&=\lim_{\mathbf{u}\to 0}\left[\frac{\partial}{\partial u_s}[\langle R_F^D f\rangle_{\mathrm{ph}}+\langle(R_F-R_F^D)f\rangle_{\mathrm{ph}}]\right],\end{aligned} \tag{2.27}$$

where the effective one-electron field-resolvent superoperator R_F^D and the field-dependent collision (or self-energy) superoperator Σ_F^D are, respectively, defined by

$$R_F^D\equiv(\hat{h}_{0F}+\hat{H}_{\mathrm{ph}}+\Sigma_F^D-i\varepsilon)^{-1}\ , \tag{2.28}$$

$$\Sigma_F^D\equiv-\lambda^2\langle\hat{V}R_F\hat{V}\rangle_{\mathrm{ph}}\ , \tag{2.29}$$

where a superscript "D" means dressed. From Eqs. (2.26) and (2.28) we obtain

$$R_F-R_F^D=R_F^D(\Sigma_F^D-\lambda\hat{V})R_F\ , \tag{2.30}$$

so that the field-resolvent superoperator R_F can be expressed in terms of the effective one-electron field-resolvent superoperator R_F^D by applying the resolvent expansion method.[23] By successive iterations using Eqs. (2.29) and (2.30), we can express R_F as an infinite series in

terms of R_F^D. Thus, we can write R_F as

$$R_F = R_F^D \sum_{n=0}^{\infty} [(\Sigma_F^D - \lambda\hat{V})R_F^D]^n$$
$$\simeq R_F^D - \lambda R_F^D \hat{V} R_F^D + \lambda^2 (R_F^D \hat{V} R_F^D \hat{V} R_F^D - R_F^D \langle \hat{V} R_F^D \hat{V} \rangle_{\rm ph} R_F^D) + O(\lambda^3) . \quad (2.31)$$

Here and hereafter, the $\simeq$ sign means as asymptotic equality, such that the right-hand side is equal to the left-hand side in the bulk limit. By making use of this expression and Eq. (2.29) and noting that any term with an odd number of V's is zero in the phonon-averaging process since V is nondiagonal in the phonon quantum number (note that V is a bilinear function of $b_{\mathbf{q}}$ and $b_{\mathbf{q}}^{\dagger}$ [see Eq. (2.2)]), we obtain Σ_F^D as

$$\Sigma_F^D \equiv -\lambda^2 \langle \hat{V} R_F \hat{V} \rangle_{\rm ph} = -\lambda^2 \langle \hat{V} R_F^D \sum_{n=0}^{\infty} [(\Sigma_F^D - \lambda\hat{V})R_F^D]^n \hat{V} \rangle_{\rm ph}$$
$$\simeq -\lambda^2 \langle \hat{V} R_F^D \hat{V} \rangle_{\rm ph} - \lambda^4 [\langle \hat{V} R_F^D \hat{V} R_F^D \hat{V} R_F^D \hat{V} \rangle_{\rm ph} - \langle \hat{V} R_F^D \langle \hat{V} R_F^D \hat{V} \rangle_{\rm ph} R_F^D \hat{V} \rangle_{\rm ph}] + O(\lambda^6) . \quad (2.32)$$

We should emphasize here that these series for R_F and Σ_F^D are *not* the conventional perturbation expansions[3,7,22] and that, in general, we expect the convergence to be quite rapid even for strongly interacting systems. The reason for this is that the effects of the interaction are included in the denominator of R_F^D. This also has the effect that the term-by-term divergences experienced in the usual perturbation theory are circumvented. The operator Σ_F^D defined by Eq. (2.29) is similar but *not* identical to the corresponding operator defined in diagrammatic terms in the previous theories.[3,7,22] To second order in Eq. (2.32), these operators are the same but there are differences in fourth and higher orders. It should be noted that the collision (or self-energy) operator defined by Eq. (2.29) [and hence Eq. (2.32)] depends on the field. Physically the inclusion of the field in the collision operator accounts for the intracollisional field effect,[3] i.e., the accelerating effect of the electric field within a collision event. Substituting Eq. (2.30) into (2.27), Eq. (2.27) can be expressed as

$$\tilde{f}_F = (\hat{h}_{0F} + \Sigma_F^D - i\varepsilon)^{-1} M_F , \quad (2.33)$$

where an operator M_F is given by

$$M_F \equiv \lim_{u\to 0} \left\{ \frac{\partial}{\partial u_s} [\langle f \rangle_{\rm ph} + \langle (\Sigma_F^D - \lambda\hat{V}) R_F f \rangle_{\rm ph}] \right\}$$
$$\simeq \lim_{u\to 0} \left\{ \frac{\partial}{\partial u_s} [\langle f \rangle_{\rm ph} - \lambda \langle \hat{V} R_F^D f \rangle_{\rm ph} + \lambda^2 (\langle \hat{V} R_F^D \hat{V} R_F^D f \rangle_{\rm ph} - \langle \langle \hat{V} R_F^D \hat{V} \rangle_{\rm ph} R_F^D f \rangle_{\rm ph}) + O(\lambda^3)] \right\} . \quad (2.33a)$$

Here we have used Eqs. (2.31) and (2.32). Finally, we must take account of the initial correlations which arise originally from the presence of the scattering potential V in the equilibrium density matrix $\rho_{\rm eq}$ and hence from f. For weakly interacting systems, we can determine f by means of a perturbation expansion. By using the residue theorem, we can express f as

$$f = \frac{i}{2\pi} \int dz\, f(z)(h_e + \lambda V - \mathbf{u}\cdot\mathbf{j} - z)^{-1}$$
$$= \frac{i}{2\pi} \int dz\, f(z) \left[\sum_{n=0}^{\infty} G_z^0 [-\lambda V G_z^0]^n \right]$$
$$\simeq \frac{i}{2\pi} \int dz\, f(z)[G_z^0 - \lambda G_z^0 V G_z^0 + \lambda^2 G_z^0 V G_z^0 V G_z^0 + O(\lambda^3)] , \quad (2.34)$$

where $f(z)$ and G_z^0 are, respectively, defined by

$$f(z) \equiv \{\exp[\beta'(z-\zeta)] + 1\}^{-1} , \quad (2.34a)$$

$$G_z^0 \equiv (h_e - \mathbf{u}\cdot\mathbf{j} - z)^{-1} . \quad (2.34b)$$

It should be noted that the nondiagonal terms of nonadiabatic nature of the scattering potential arising from f are contracted after substituting Eq. (2.34) into Eq. (2.33a), whence only initially correlated terms with an even number of V's survive the phonon averaging process in M_F [in Eq. (2.33a)]. We have developed a prescription of a method for the practical evaluation of the nonlinear conductivity tensor for an electron-phonon system. We would like to emphasize that our results are exact in the bulk limit other than the assumption, which is justified for the statistical operator $\rho_{\rm eq}$ [Eq. (2.20)]. It should be stressed that the formulas obtained so far are developed independently of the single-particle representation (position, momentum, Landau, Stark ladder, or other) and can be applied for a system subjected to a static magnetic field. The operator equation (2.33) along with Eqs. (2.32) and (2.33a) forms the basic equation for further treatments of the high-field transport problems. Barker[3] also obtained a similar expression (2.33) but its internal structure is different in higher-order terms. So far we have not mentioned anything about the field strength. If the applied electric field is sufficiently weak, we may further expand $\sigma(\mathbf{F})$ in terms of the field. With increasing field strength, the nonlinear characteristics of the response become more important and an expansion in powers of the field becomes less suitable. Therefore it is desirable to derive the formulas available for computing $\sigma(\mathbf{F})$ which is valid for all values of the electric field. To evaluate the nonlinear conductivity tensor (2.33) without expanding in powers of an electric field, one must choose an appropriate representation for an electron state. We shall use a representation in which the Hamiltonian h_{0F} $(=h_e + h_F)$ is diagonal. In this representation, let us write $\mathrm{tr}(j_r \tilde{f}_F)$ in a matrix representation:

$$\mathrm{tr}(j_r \tilde{f}_F) = \sum_{\lambda_1} \sum_{\lambda_2} \langle \lambda_2 | j_r | \lambda_1 \rangle \langle \lambda_1 | \tilde{f}_F | \lambda_2 \rangle , \quad (2.35)$$

where a single-electron state labeled λ is an eigenfunction of the Hamiltonian (2.22), satisfying

$$h_{0F} |\lambda\rangle = E_\lambda |\lambda\rangle . \quad (2.36)$$

The E_λ's are the eigenvalues of h_{0F} and depend on the field. It should be noted that for this choice of an electron state for an unperturbed Hamiltonian, the field can be treated exactly and hence thereby usual perturbation expansions in powers of the field (which assume a weak field) are avoided. Our problem is now reduced to the evaluation of the matrix elements of $\tilde{f}_F$ for averaging the electronic current operator in Eq. (2.35). Multiplying $(\hat{h}_{0F}+\Sigma_F^D-i\varepsilon)$ from the left on Eq. (2.33), we obtain

$$(\hat{h}_{0F}+\Sigma_F^D-i\varepsilon)\tilde{f}_F=M_F \ , \tag{2.37}$$

where operators Σ_F^D and M_F are, respectively, given by Eqs. (2.32) and (2.33a). The required matrix elements of $\tilde{f}_F$ are thus given by taking the λ_1,λ_2 element of Eq. (2.37):

$$(E_{\lambda_1}-E_{\lambda_2}-i\varepsilon)\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle+\langle\lambda_1|\Sigma_F^D\tilde{f}_F|\lambda_2\rangle = \langle\lambda_1|M_F|\lambda_2\rangle \ . \tag{2.38}$$

Equation (2.37) [or (2.38)] has a simple interpretation as a generalized high-field quantum-transport equation for the phonon-averaged one-electron quantum-statistical operator $\tilde{f}_F$ and it shows clearly where the influence of the reservoir (phonons) enters. In Eq. (2.38), if $\Sigma_F^D=0$ (no interaction), we observe the divergent nature of $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$ at $(E_{\lambda_1}-E_{\lambda_2}-i\varepsilon)=0$. It is to be expected as seen in Eq. (2.24) that $\mathrm{tr}(j_r\tilde{f}_F)$ represents the infinite current which must occur in the absence of scattering. Due to the interaction incorporated in Σ_F^D, $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$ is relaxed to finite values by scattering processes. It is noticed that the first term in Eq. (2.38) represents the response of the system to the electric field in the absence of collisions. On the other hand, the second term describes how electrons are scattered by phonons in the system and are influenced by the field during collisions. As will be shown in the next section, this collision term exhibits the gain-loss structure appearing in the quantum Boltzmann collision integral for an electron-phonon system but is relaxed by the inclusion of the effect of the field on the collision process (ICFE) through Σ_F^D. We note that those terms [Eqs. (2.32) and (2.33a)] which depend on the interaction between electrons and phonons may therefore give rise to dissipation of energies gained from the field. These terms play the most essential role in the analysis of transport phenomena. Solving Eq. (2.38) for $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$, one can, in principle, evaluate the field-dependent conductivity tensor which would be valid for an arbitrary strength of the electric field and the interaction potential.

III. APPROXIMATION PROCEDURE

The transport equation (2.37) [or (2.38)] along with Eqs. (2.28), (2.32), and (2.33a) represents the starting point for setting up tractable transport equations for $\tilde{f}_F$ (or $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$). An exact solution of transport equation (2.38) for $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$ is, however, not feasible, since the collision operator Σ_F^D is, in general, nondiagonal. In order to proceed further we therefore need an approximation scheme for the evaluation of $\langle\lambda_1|\Sigma_F^D\tilde{f}_F|\lambda_2\rangle$. We consider the simplest situation, assuming weak electron-phonon coupling. Firstly we approximate R_F^D of Eq. (2.28) by neglecting Σ_F^D, so that

$$R_F^D\approx(\hat{h}_{0F}+\hat{H}_{\mathrm{ph}}-i\varepsilon)^{-1}\equiv R_F^0 \ , \tag{3.1}$$

$$\Sigma_F^D\approx-\lambda^2\langle\hat{V}R_F^0\hat{V}\rangle_{\mathrm{ph}}\equiv-\Sigma_F^0 \ , \tag{3.2}$$

where we have kept only the lowest-order nonvanishing contribution to Σ_F^D from its series expansion in Eq. (2.32). It should be noted that the scattering vertex Σ_F^0 retains the field dependence, so that it includes the effect of the field on collisions (ICFE). To this order of approximation, the initial correlations vanish and the operator M_F is approximated by

$$M_F\approx M^0=\lim_{u\to 0}\left[\frac{\partial}{\partial u_s}\frac{i}{2\pi}\int dz\, f(z)G_z^0\right] \ , \tag{3.3}$$

where $f(z)$ and G_z^0 are given, respectively, by Eqs. (2.34a) and (2.34b). Using these, the right-hand-side term of Eq. (2.38) can be easily evaluated as

$$\langle\lambda_1|M^0|\lambda_2\rangle=\frac{f(\varepsilon_{\lambda_1}-f(\varepsilon_{\lambda_2})}{\varepsilon_{\lambda_1}-\varepsilon_{\lambda_2}}\langle\lambda_1|j_s|\lambda_2\rangle \ , \tag{3.4}$$

where ε_λ is the energy eigenvalue of h_e satisfying

$$h_e|\lambda\rangle=\varepsilon_\lambda|\lambda\rangle \ . \tag{3.5}$$

The second stage of approximation involves a scheme for evaluating $\langle\lambda_1|\Sigma_F^0\tilde{f}_F|\lambda_2\rangle$, which will be outlined in the following subsection.

A. Evaluation of the collision term

In order to see the structure of the collision term, we have to evaluate $\langle\lambda_1|\Sigma_F^0\tilde{f}_F|\lambda_2\rangle$ (or, more generally, $\langle\lambda_1|\Sigma_F^D\tilde{f}_F|\lambda_2\rangle$) explicitly. Since the collision operator is a function of superoperators, to evaluate the matrix element of such a function is a complicated matter. The practical calculation involving superoperators can be facilitated by using a general technique derived by Resibois.[22,25,26] In this method a four-leg structure (which should be used for calculating the matrix elements of superoperators) is reduced to familiar algebra. Let us specify the λ_1,λ_2 matrix element of an arbitrary operator A by a new matrix representation:

$$\langle\lambda_1|A|\lambda_2\rangle=A_{\lambda_1-\lambda_2}\left[\frac{\lambda_1+\lambda_2}{2}\right] \ . \tag{3.6}$$

If we introduce a pair of new variables (ν,μ) replacing the pair (λ_1,λ_2) such that

$$\nu\equiv(\lambda_1+\lambda_2)/2, \quad \mu\equiv\lambda_1-\lambda_2 \ , \tag{3.7}$$

then Eq. (3.6) is expressed by the new notation

$$\langle\lambda_1|A|\lambda_2\rangle=\langle\nu+\mu/2|A|\nu-\mu/2\rangle\equiv A_\mu(\nu) \ . \tag{3.8}$$

Let us define a matrix operator $(\mu|\check{O}(\nu)|\mu')$ in ν space associated with superoperator $\hat{O}$ such that

$$(\mu|\check{O}(\nu)|\mu')\equiv\eta^{\mu'}O_{\mu-\mu'}(\nu)\eta^{-\mu}-\eta^{-\mu'}O_{\mu-\mu'}(\nu)\eta^{\mu} \ , \tag{3.9}$$

where we have introduced the shift operators $\eta^{\pm\mu}$ which replaces a function of ν by the same function of $\nu\pm\frac{1}{2}\mu$:

$$\eta^{\pm\mu}f(\nu)=f(\nu\pm\mu/2)\ . \tag{3.10}$$

Notice that an operator $\check{O}(\nu)$ is just another way of writing the superoperator $\hat{O}$, and a matrix operator $(\mu|\check{O}(\nu)|\mu')$ is a matrix with respect to one set of quantum numbers and an operator with respect to the other. It is then easy to verify that the matrix elements of a commutator can be expressed in terms of $(\mu|\check{O}(\nu)|\mu')$:

$$\langle\lambda_1|\hat{O}A|\lambda_2\rangle\equiv(\hat{O}A)_\mu(\nu)=\langle\nu+\mu/2|\hat{O}A|\nu-\mu/2\rangle$$
$$=\sum_{\mu'}(\mu|\check{O}(\nu)|\mu')A_{\mu'}(\nu)\ . \tag{3.11}$$

In the present case, from Eqs. (2.36), (3.8), and (3.9), the explicit form of the unperturbed and the perturbed parts ($\hat{h}_{0F}$ and $\hat{V}$) are given, respectively, by

$$(\mu|\check{h}_{0F}(\nu)|\mu')=(E_{\nu+\mu/2}-E_{\nu-\mu/2})\delta_{\mu'-\mu}\ , \tag{3.12}$$

$$(\mu|\check{V}(\nu)|\mu')=\langle\nu+\mu/2|V|\nu-\mu/2+\mu'\rangle\eta^{\mu'-\mu}$$
$$-\langle\nu+\mu/2-\mu'|V|\nu-\mu/2\rangle\eta^{-\mu'+\mu}\ , \tag{3.13}$$

where in Eq. (3.12), δ denotes the Kronecker symbol. It should be noted that the unperturbed superoperator $\hat{h}_{0F}$ is diagonal in the μ variable.

Since we know the mathematical device outlined above, the evaluation of the collision term is straightforward. Applying the ν-μ representation to the present problem, one obtains

$$\langle\lambda_1|\Sigma_F^0\tilde{f}_F|\lambda_2\rangle=\lambda^2\langle 1|\mathrm{Tr}^{(\mathrm{ph})}(\rho_{\mathrm{ph}}\hat{V}R_F^0\hat{V}\tilde{f}_F)|2\rangle\equiv(\Sigma_F^0\tilde{f}_F)_\mu(\nu)$$
$$=\sum_{\mu'}(\mu|\check{\Sigma}_F^0(\nu)|\mu')\tilde{f}_{F_{\mu'}}(\nu)$$
$$=\lambda^2\sum_{\mu'}\sum_{\mu''}\langle(\mu|\check{V}(\nu)|\mu')[h_{0F}(\nu,\mu')+\hat{H}_{\mathrm{ph}}-i\varepsilon]^{-1}(\mu'|\check{V}(\nu)|\mu'')\rangle_{\mathrm{ph}}\tilde{f}_{F_{\mu''}}(\nu)$$
$$=\lambda^2\sum_{\mathbf{q}}\sum_{\mu',\mu''}\{(N_{\mathbf{q}}+1)[\langle\nu+\mu/2|\gamma_{\mathbf{q}}|\nu-\mu/2+\mu'\rangle(E_{\nu-\mu/2+\mu'}-E_{\nu-\mu/2}+\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu-\mu/2+\mu'|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2+\mu''\rangle\tilde{f}_{F_{\mu''}}(\nu-\mu/2+\mu''/2)$$
$$+\langle\nu+\mu/2-\mu''|\gamma_{\mathbf{q}}|\nu+\mu/2-\mu'\rangle(E_{\nu+\mu/2}+E_{\nu+\mu/2-\mu'}-\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu+\mu/2-\mu'|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu+\mu/2-\mu''/2)]$$
$$+N_{\mathbf{q}}[\langle\nu+\mu/2|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2+\mu'\rangle(E_{\nu-\mu/2+\mu'}-E_{\nu-\mu/2}-\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu-\mu/2+\mu'|\gamma_{\mathbf{q}}|\nu-\mu/2+\mu''\rangle\tilde{f}_{F_{\mu''}}(\nu-\mu/2+\mu''/2)$$
$$+\langle\nu+\mu/2-\mu''|\gamma_{\mathbf{q}}^\dagger|\nu+\mu/2-\mu'\rangle(E_{\nu+\mu/2}-E_{\nu+\mu/2-\mu'}+\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu+\mu/2-\mu'|\gamma_{\mathbf{q}}|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu+\mu/2-\mu''/2)]\}$$
$$-\lambda^2\sum_{\mathbf{q}}\sum_{\mu',\mu''}\{(N_{\mathbf{q}}+1)[\langle\nu+\mu/2|\gamma_{\mathbf{q}}|\nu-\mu/2+\mu'\rangle(E_{\nu-\mu/2+\mu'}-E_{\nu-\mu/2}+\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu-\mu/2+\mu'-\mu''|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu-\mu/2+\mu'-\mu''/2)$$
$$+\langle\nu+\mu/2|\gamma_{\mathbf{q}}|\nu+\mu/2-\mu'+\mu''\rangle(E_{\nu+\mu/2}-E_{\nu+\mu/2-\mu'}-\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu+\mu/2-\mu'|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu+\mu/2-\mu'+\mu''/2)]$$
$$+N_{\mathbf{q}}[\langle\nu+\mu/2|\gamma_{\mathbf{q}}^\dagger|\nu-\mu/2+\mu'\rangle(E_{\nu-\mu/2+\mu'}-E_{\nu-\mu/2}-\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu-\mu/2+\mu'-\mu''|\gamma_{\mathbf{q}}|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu-\mu/2+\mu'-\mu''/2)$$
$$+\langle\nu+\mu/2|\gamma_{\mathbf{q}}^\dagger|\nu+\mu/2-\mu'+\mu''\rangle(E_{\nu+\mu/2}-E_{\nu+\mu/2-\mu'}+\hbar\omega_{\mathbf{q}}-i\varepsilon)^{-1}$$
$$\times\langle\nu+\mu/2-\mu'|\gamma_{\mathbf{q}}|\nu-\mu/2\rangle\tilde{f}_{F_{\mu''}}(\nu+\mu/2-\mu'+\mu''/2)]\}\ , \tag{3.14}$$

where $N_{\mathbf{q}}$ is the Planck distribution for phonons which is given by

$$N_{\mathbf{q}}=\mathrm{Tr}^{(\mathrm{ph})}(\rho_{\mathrm{ph}}b_{\mathbf{q}}^\dagger b_{\mathbf{q}})=[\exp(\beta\hbar\omega_{\mathbf{q}})-1]^{-1}\ . \tag{3.15}$$

It is noted that expression (3.14) is exact and includes the field dependence within a collision. The collision term (3.14) is rather complicated. In order to see the mathematical structure more clearly, let us write Eq. (3.14) in the λ representation:

$$\langle\lambda_1|\Sigma_F^0\tilde{f}_F|\lambda_2\rangle\equiv\langle 1|\Sigma_F^0\tilde{f}_F|2\rangle=\sum_{3,4}\Sigma^0_{F12;34}\langle 3|\tilde{f}_F|4\rangle=T_{\rm in}-T_{\rm out}\ , \tag{3.16}$$

where $T_{\rm in}$ and $T_{\rm out}$ are given, respectively, by

$$T_{\rm in}=\lambda^2\sum_{\mathbf{q}}\sum_{3,4}\left[(N_{\mathbf{q}}+1)\left[\frac{\langle 1|\gamma_{\mathbf{q}}|3\rangle\langle 3|\gamma_{\mathbf{q}}^\dagger|4\rangle\langle 4|\tilde{f}_F|2\rangle-\langle 1|\gamma_{\mathbf{q}}|3\rangle\langle 3|\tilde{f}_F|4\rangle\langle 4|\gamma_{\mathbf{q}}^\dagger|2\rangle}{E_3-E_2+\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right.$$
$$\left.+N_{\mathbf{q}}\left[\frac{\langle 1|\gamma_{\mathbf{q}}^\dagger|3\rangle\langle 3|\gamma_{\mathbf{q}}|4\rangle\langle 4|\tilde{f}_F|2\rangle-\langle 1|\gamma_{\mathbf{q}}^\dagger|3\rangle\langle 3|\tilde{f}_F|4\rangle\langle 4|\gamma_{\mathbf{q}}|2\rangle}{E_3-E_2-\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right], \tag{3.16a}$$

$$T_{\rm out}=\lambda^2\sum_{\mathbf{q}}\sum_{3,4}\left[(N_{\mathbf{q}}+1)\left[\frac{\langle 1|\gamma_{\mathbf{q}}|4\rangle\langle 4|\tilde{f}_F|3\rangle\langle 3|\gamma_{\mathbf{q}}^\dagger|2\rangle-\langle 1|\tilde{f}_F|4\rangle\langle 4|\gamma_{\mathbf{q}}|3\rangle\langle 3|\gamma_{\mathbf{q}}^\dagger|2\rangle}{E_1-E_3-\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right.$$
$$\left.+N_{\mathbf{q}}\left[\frac{\langle 1|\gamma_{\mathbf{q}}^\dagger|4\rangle\langle 4|\tilde{f}_F|3\rangle\langle 3|\gamma_{\mathbf{q}}|2\rangle-\langle 1|\tilde{f}_F|4\rangle\langle 4|\gamma_{\mathbf{q}}^\dagger|3\rangle\langle 3|\gamma_{\mathbf{q}}|^2\rangle}{E_1-E_3+\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right]. \tag{3.16b}$$

It should be note that the $|\lambda_i\rangle$ ($\equiv|i\rangle$) and the E_i ($i=1,2,3,\ldots$), are, respectively, the eigenstates and eigenvalues of h_{0F} satisfying Eq. (2.36). For the weak-field limit ($\mathbf{F}\rightarrow\mathbf{0}$), the collision terms (3.16) reduce to those obtained by the Green's-function approach [see Eq. (7.40) of Ref. 27]. Equation (2.38) along with Eqs. (3.4), (3.16a), and (3.16b) represents a set of matrix equations which specify the elements of $\langle 1|\tilde{f}_F|2\rangle$ for the lowest order in λ. It is noticed that $T_{\rm in}$ and $T_{\rm out}$ correspond to the gain (scattering in) and the loss (scattering out) structures of terms, respectively. The first two terms in $T_{\rm in}$ physically represent the terms that account for the rate at which the electrons gain the energy from the field and arrive at the state of interest $|\lambda\rangle$ by emitting a phonon of frequency $\omega_{\mathbf{q}}$ and the wave vector $\mathbf{q}$. On the other hand, the last two terms in $T_{\rm in}$ similarly account for the rate at which the electrons gain the energy from the field and leave the state $|\lambda\rangle$ by absorbing a phonon of frequency $\omega_{\mathbf{q}}$ and the wave vector $\mathbf{q}$. Although the first two and the last two terms in $T_{\rm out}$ have the same physical meaning as mentioned above, the only difference is that these terms arise because of the microscopic collisions in which the electrons gain the energy from the field but leave from the state $|\lambda\rangle$ by emitting and absorbing a phonon of frequency $\omega_{\mathbf{q}}$ and the wave vector $\mathbf{q}$. It should be noted that in contrast to the treatment of collisional broadening (CB) discussed for weak fields (where collisions are treated as point events),[23] Eq. (3.16) includes the field effect during collisions (ICFE). Equation (2.38), along with Eqs. (3.4) and (3.16), represents the generalized quantum-transport equation to be solved for $\langle\lambda_1|\tilde{f}_F|\lambda_2\rangle$. With only the diagonal terms retained, Eq. (3.16) becomes

$$\langle 1|\Sigma_F^0\tilde{f}_F|2\rangle\approx\Sigma^0_{F12;12}\langle 1|\tilde{f}_F|2\rangle\equiv\Sigma_{1,2}(F_s)\langle 1|\tilde{f}_F|2\rangle\ , \tag{3.17}$$

where the complex irreducible self-energy $\Sigma_{1,2}(F_s)$ is given by

$$\Sigma_{1,2}(F_s)\approx\lambda^2\sum_{\mathbf{q}}\sum_{3}\left[(N_{\mathbf{q}}+1)\left[\frac{|\langle 2|\gamma_{\mathbf{q}}|3\rangle|^2}{E_1-E_3-\hbar\omega_{\mathbf{q}}-i\varepsilon}+\frac{|\langle 3|\gamma_{\mathbf{q}}^\dagger|1\rangle|^2}{E_3-E_2+\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right.$$
$$\left.+N_{\mathbf{q}}\left[\frac{|\langle 2|\gamma_{\mathbf{q}}^\dagger|3\rangle|^2}{E_1-E_3+\hbar\omega_{\mathbf{q}}-i\varepsilon}+\frac{|\langle 3|\gamma_{\mathbf{q}}|1\rangle|^2}{E_3-E_2-\hbar\omega_{\mathbf{q}}-i\varepsilon}\right]\right], \tag{3.17a}$$

where vertex correction terms involving $\langle i|\gamma_{\mathbf{q}}|i\rangle$ have been dropped[27] since they lead to contributions of order $n_0^{-1/3}$ compared to those which are kept (n_0 is the number of unit cells in the crystal). It should be noted that the matrix elements of the interaction term depend generally on the electric field, and the field dependence on the electronic transition rates is introduced through the electron states and the energy denominators. Real and imaginary parts in expression (3.17a) are of basic interest and are related to the quantities measured experimentally. Utilizing Eqs. (2.38) (3.4), and (3.17a), the steady-state nonlinear conductivity (2.24) is then given by

$$\mathrm{Re}\{\sigma_{rs}(F_s)\}=\frac{\hbar}{\Omega}\sum_{1,2}\langle 1|j_s|2\rangle\langle 2|j_r|1\rangle\frac{f(\varepsilon_1)-f(\varepsilon_2)}{\varepsilon_1-\varepsilon_2}\frac{\Gamma_{1,2}(F_s)}{[E_1-E_2-\Delta_{1,2}(F_s)]^2+\Gamma_{1,2}^2(F_s)}\ , \tag{3.18}$$

where $\Gamma_{1,2}(F_s)$ and $\Delta_{1,2}(F_s)$ can be calculated from Eq. (3.17a):

$$\Gamma_{1,2}(F_s)\equiv\mathrm{Im}\{\Sigma_{1,2}(F_s)\}$$
$$=\pi\sum_{\mathbf{q}}\sum_{3}\{(N_{\mathbf{q}}+1)[|\langle 2|\gamma_{\mathbf{q}}|3\rangle|^2\delta(E_1-E_3-\hbar\omega_{\mathbf{q}})+|\langle 3|\gamma_{\mathbf{q}}^\dagger|1\rangle|^2\delta(E_3-E_2+\hbar\omega_{\mathbf{q}})]$$
$$+N_{\mathbf{q}}[|\langle 2|\gamma_{\mathbf{q}}^\dagger|3\rangle|^2\delta(E_1-E_3+\hbar\omega_{\mathbf{q}})+|\langle 3|\gamma_{\mathbf{q}}|1\rangle|^2\delta(E_3-E_2-\hbar\omega_{\mathbf{q}})]\}\ , \tag{3.19a}$$

$$\Delta_{1,2}(F_s)\equiv \mathrm{Re}\{\Sigma_{1,2}(F_s)\}$$
$$=\sum_{\mathbf{q}}\sum_{3}\left[(N_{\mathbf{q}}+1)\left[|\langle 2|\gamma_{\mathbf{q}}|3\rangle|^2\mathrm{P}\frac{1}{E_1-E_3-\hbar\omega_{\mathbf{q}}}+|\langle 3|\gamma_{\mathbf{q}}^{\dagger}|1\rangle|^2\mathrm{P}\frac{1}{E_3-E_2+\hbar\omega_{\mathbf{q}}}\right]\right.$$
$$\left.+N_{\mathbf{q}}\left[|\langle 2|\gamma_{\mathbf{q}}^{\dagger}|1\rangle|^2\mathrm{P}\frac{1}{E_1-E_3+\hbar\omega_{\mathbf{q}}}+|\langle 3|\gamma_{\mathbf{q}}|1\rangle|^2\mathrm{P}\frac{1}{E_3-E_2-\hbar\omega_{\mathbf{q}}}\right]\right]. \tag{3.19b}$$

To obtain Eqs. (3.19a) and (3.19b), we have passed to the limit $\varepsilon\to 0^+$ as is required by Eq. (2.24). The symbols Re and Im denote, respectively, the real and the imaginary parts of the quantity which follows are to be taken, and the symbol P in Eq. (3.19b) denotes Cauchy's principal-value integral. The expression (3.18) is just the nonlinear version of the Drude formula for the static conductivity, which is associated with electronic transition between states $|\lambda_1\rangle$ and $|\lambda_2\rangle$ in the presence of the electron-phonon interaction. We see that the field-dependent conductivity (3.18) exhibits a Lorentz-like line shape. The quantity $\Delta_{1,2}(F_s)$ plays the role of the line shift, and $\Gamma_{1,2}(F_s)$ plays the role of the half-width. $\Gamma_{1,2}(F_s)/\hbar$ gives the reciprocal of the relaxation time. It should be noted that both of these quantities are functions of temperature and the external electric field. The δ functions in the terms for $\Gamma_{1,2}(F_s)$ state the conservation of energy in those processes where an electron is excited from $|\lambda_1\rangle$ to $|\lambda_2\rangle$ by gaining energy from the field during collision events and is then scattered by the electron-phonon interaction to state $|\lambda_3\rangle$ with the emission or absorption of a phonon. For weak-field limit ($F_s\to 0$), Eq. (3.18) reduces to the Drude formula for the static conductivity[23,27] (linear response to the field), which is given by replacing the energies E_{λ_i}'s and eigenfunctions $|\lambda_i\rangle$'s in Eq. (3.18) with Eqs. (3.19a) and (3.19b) by corresponding energies ε_{ν_i}'s and eigenfunctions $|\nu_i\rangle$'s since the eigenvalue equation $h_{0f}|\lambda_i\rangle=E_{\lambda_i}|\lambda_i\rangle$ becomes $h_e|\nu_i\rangle=\varepsilon_{\nu_i}|\nu_i\rangle$ as $F_s\to 0$.

Although we have formulated the theory for an electron-phonon system, relaxation due to impurities can be also included in the theory; in the lowest approximation, the relaxation effect due to phonons and impurities are obtained by the sum of relaxations due to the phonons and the impurity scatterings. The formula for the relaxation due to the impurity scattering can be obtained for a low-impurity-density case simply by the following replacement in Eq. (3.19a): $N_{\mathbf{q}}+1\to\Omega N_{\mathrm{imp}}$ (N_{imp} is the impurity density), $N_{\mathbf{q}}\to 0$, $\omega_{\mathbf{q}}\to 0$.

IV. SUMMARY AND CONCLUDING REMARKS

We have developed a quantum-statistical theory of high-field transport based on Tani's theory of nonlinear response.[4-6] A general expression for an exact steady-state nonlinear current in the Ohm's law form is obtained and a field-dependent dc conductivity (2.12) is defined. We have shown a detailed method of evaluation of this field-dependent conductivity (and hence a steady-state nonlinear current) for an electron-phonon system, applying a representation-independent resolvent superoperator method.[23]

In our formulation, two restrictions have been made: one is the neglect of electron-electron interactions, and can be partly included in $\gamma_{\mathbf{q}}$ through the screened interaction potential; the other is that the lattice is assumed to be in thermal equilibrium and that the so-called factorization approximation (2.20) is employed for the equilibrium statistical operator ρ_{eq}. Therefore, the present theory can be applied to low-density nondegenerate semiconductors when the effects of nonequilibrium heating of phonon population is negligible.

Adopting these assumptions, the evaluation of the field-dependent conductivity is then reduced to the problem of finding the solution of the tractable operator transport equation (2.37) for the phonon-averaged one-electron statistical operator $\tilde{f}_F$. The statistical operator $\tilde{f}_F$ [and hence the field-dependent conductivity $\sigma_{rs}(F_s)$] is determined by a field-dependent one-electron resolvent R_F, which is given in terms of a simpler effective one-electron resolvent R_F^D, (2.28). This depends on the self-energy (or collision) superoperator Σ_F^D which is defined at the outset [see (2.29)]. Hence, any collision process between an electron and phonons is thereby assumed to take place in the averaged field of phonons. The effect of such a field is to induce perturbed single-particle energies and to introduce lifetimes for the electron states by virtual collision processes. In other words, within each scattering event, an electron will propagate in a perturbed state which is controlled by virtual scattering on the entire scattering medium. We have shown how this self-energy effect, which takes ICFE into account, is formulated into the theory without recourse to a relaxation approximation used in the Boltzmann approach.[2] The self-energy results in, e.g., the lifetime broadening, which is responsible for the spectral broadening of line shapes. Therefore, the effects of the interaction, ICFE in each collision event, and scatterings are studied theoretically by examining the real part of the conductivity tensor. We have obtained the expression for the field-dependent conductivity [(3.18) with (3.19a) and (3.19b)] for the lowest-order nonvanishing contribution of an electron-phonon interaction. As can be seen in Eq. (3.18), these self-energy effects lead to level shift Δ and energy broadening Γ of otherwise sharp energy conservation in the collision process and so the field-dependent conductivity is of the Lorentz-like line shape. It should be noted that Δ and Γ depend on the lattice temperatures as well as on the field $\mathbf{F}$ since the influence of the field within a collision (ICFE) is taken into account. If we take the limit, $\mathbf{F}\to\mathbf{0}$, to Δ and Γ, these quantities are reduced to the usual formulas obtained from Boltzmann-transport theory (or the linear-transport theory), which assumes scatterings as the point collisions so that the effect of the field within a collision (ICFE) is not taken into considera-

tion in the scattering events. The field-dependent self-energy effect is conjectured to be crucial in the transport physics of ultrasmall devices for which the mean free time between collisions is comparable to the transit time through the semiconductor. Although the present formulation is based on a simple one-band spin-free model, the extension of the present theory to a multiband model[28] can be done straightforwardly, since our formulation is made in an operator form so that it is valid irrespective of the system studied. The differences arise only in the final stage of calculation. We will extend the present theory to the study of hot-electron problems in future publications.

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