

## Nonlinear high-frequency conductivity in semiconductors

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We study the nonlinear effects of the electron conductivity in semiconductors when a strong high-frequency (HF) electric field is applied together with a direct-current (dc) electric field. The dynamic equation that we used is different from the Boltzmann equation by considering the memory effect for drift oscillating motion of electrons. Using the drifted temperature model, we derive a set of equations from which the amplitude and phase of each harmonic component of the electron drift velocity and the electron temperature can be obtained. In the weak-HF-field limit our approach reduces to the well-known memory-function method. We have calculated the conductivity of electrons in a bulk *n*-type GaAs sample. The nonlinear effects are shown. In particular, the dc conductivity decreases and definitely becomes negative with increase of the first- and second-harmonic components of the applied HF field. Comparison is made with experiments.

### I. INTRODUCTION

Study of the nonlinear behavior of the carrier conductivity in a strong high-frequency (HF) electric field is important, because microwave devices often operate in a region under a strong HF field. The free-carrier absorption of laser light is also related to the transport of electrons in a strong HF (possibly optical) field. This subject is also interesting in its own right as a fundamental problem. Experiments have shown some interesting nonlinear effects, for example, that the direct current (dc) conductivity decreases with increase of the strength of HF field.<sup>1,2</sup> However, there is a qualitative difference between the results of different experimental groups. The authors of Ref. 1 reported that dc current in an *n*-type GaAs sample placed in a strong microwave field decreases dramatically with an increase of HF field, and dc conductivity definitely becomes negative when the HF field exceeds 1 kV/cm. The results in Ref. 2 show a much slower decrease of dc conductivity with increasing HF field, declining only 40% from its static value even when HF electric field increases up to 3 kV/cm.

In theoretical study of HF conductivity, a good understanding has been achieved when only a weak HF electric field is applied.<sup>3-5</sup> There are several studies for the case of a strong dc field (hot electrons) together with a weak HF field.<sup>6-9</sup> The case of a strong HF electric field is a more difficult problem. Only a few papers deal with this subject using a dynamic theory,<sup>8,9</sup> and these calculations rely on Monte Carlo simulation to solve the time-dependent Boltzmann equation. While the Monte Carlo method is powerful in the treatment of dc transport, there are some difficulties in the management of HF transport. One of these is the difficulty in distinguishing the "steady state" from the initial transient. When the period of the applied field is comparable to the scattering

relaxation time, the applicability of the Boltzmann theory becomes questionable. A tractable and reliable approach is therefore still lacking.

In this paper we propose an analytical approach to study the nonlinear effects of electron transport when a strong HF electric field is applied (may together with a dc electric field). When a HF field is applied, the trajectory of an electron between scatterers is no longer straight. An oscillating drift motion of electrons is superposed upon the random motion of electron caused by the scattering processes. For determining the motion of electrons after time *t*, not only the condition at time *t* should be known, but information of the drift motion of electrons during a period before *t* is also needed. This memory effect is included in our approach. The dynamic equation that we used, therefore, is different from the Boltzmann equation. (If the memory effect for the drift motion is neglected, our formulas reduce to those obtained in the Boltzmann approach.) Using the above approach, we study the "steady-state" HF transport. We derive a set of evolution equations for the drift velocity of electrons. Assuming that the distribution function of electrons in relative coordinates is a Maxwellian, we also obtain a set of evolution equations for the energy of electrons, which is related to the electron temperature. Solving the group of equations, we can obtain dc component and each harmonic component of electron drift velocity and the electron temperature.

We have used this approach to calculate electron transport in an *n*-type GaAs sample. A microwave frequency  $\nu = 35$  GHz is chosen as the base frequency in the present calculation. The strength of the HF electric field is up to about 1 kV/cm. A weak dc electric field  $\mathcal{E}_0 = 10$  V/cm is also applied. We find that when only the electric field with base frequency  $\mathcal{E}_1$  is applied, the induced second harmonic of drift velocity is rather

small, while a large second harmonic of the electron temperature can be produced; the dc conductivity only reduces about 5% from its static value (without HF field), when the applied HF field is up to  $\mathcal{E}_1 = 1$  kV/cm. This result is in agreement with that of Ref. 2. Surprisingly, if a second harmonic electric field is applied together with the first harmonic (with the same phase), the dc conductivity decreases dramatically with increase of the HF field, and definitely becomes negative when  $\mathcal{E}_1$  and  $\mathcal{E}_2$  are near 1 kV/cm, namely, a weak positive dc potential drop can produce a negative dc current. This phenomenon was shown experimentally in Ref. 1.

The paper is organized as follows. In Sec. II we sketch the derivation of the kinetic equations for the harmonic components of electron drift velocity and the electron temperature. In Sec. III we briefly discuss the formulas and results for the case of a weak HF electric field. In Sec. IV we present the results of the numerical solution for nonlinear HF conductivity in an  $n$ -type GaAs bulk sample. Section V is devoted to discussion. Finally, in the Appendix we use a functional-integral approach to derive the formulas, proving an important statement in the text [Eq. (2.12)].

## II. FORMULATION

### A. Hamiltonian

For convenience in the theoretical description, we restrict our present study to a three-dimensional electron system. It is straightforward to generalize our approach to a quasi-two-dimensional electron system, such as electrons in a semiconductor quantum well. We consider  $N$  electrons, scattered by  $n_i$  randomly distributed impurities and phonons, applied by an spatially uniform electric field  $\mathcal{E}(t)$ . The following form of the applied electric field  $\mathcal{E}(t)$  is assumed:

$$\mathcal{E}(t) = \mathcal{E}_0 + \sum_n \mathcal{E}_n e^{in\omega t}, \quad n = \pm 1, \pm 2, \dots, \quad (2.1)$$

where we have  $\mathcal{E}_{-n} = \mathcal{E}_n^*$ , with  $\mathcal{E}^*$  the complex conjugate of  $\mathcal{E}$ , because  $\mathcal{E}(t)$  is a real quantity. In order to describe the dynamical properties in the presence of a spatially uniform electric field, it is convenient to separate the center-of-mass motion from the relative motion of the electrons.<sup>10</sup> The Hamiltonian of the free-electron system can then be written as

$$\hat{H}_e = \hat{H}_c + \hat{H}'_e + \hat{V}'_{ee}, \quad (2.2)$$

where

$$\hat{H}_c = \hat{\mathbf{P}}^2/2M - Ne\mathcal{E}(t) \cdot \hat{\mathbf{R}} \quad (2.3)$$

is the center-of-mass Hamiltonian, with  $\hat{\mathbf{P}}$  and  $\hat{\mathbf{R}}$  the

center-of-mass momentum and coordinate operators, respectively,  $N$  the total number of electrons, and  $M = Nm$ , with  $m$  the electron effective mass. The free-electron Hamiltonian in relative coordinates is given by

$$\hat{H}'_e = \sum_{\mathbf{k}} E_{\mathbf{k}} \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} \quad (2.4)$$

where  $E_{\mathbf{k}} = \hbar^2 \mathbf{k}^2 / 2m$  with  $\hbar \mathbf{k}$  the crystal momentum of electron in state  $\mathbf{k}$ . (We use parabolic-band approximation.)  $\hat{c}_{\mathbf{k}}^\dagger$  and  $\hat{c}_{\mathbf{k}}$  are electron creation and annihilation operators in relative coordinates; spin indices have been omitted (but not ignored).  $\hat{V}'_{ee}$  is the Coulomb interaction in relative coordinates. The latter has the standard form found in the literature.<sup>11</sup> The Hamiltonian for free phonons is given by

$$\hat{H}_p = \sum_{\mathbf{q}, \lambda} \hbar \Omega_{\mathbf{q}\lambda} \hat{b}_{\mathbf{q}\lambda}^\dagger \hat{b}_{\mathbf{q}\lambda}, \quad (2.5)$$

where  $\hat{b}_{\mathbf{q}\lambda}^\dagger$  and  $\hat{b}_{\mathbf{q}\lambda}$  are phonon creation and annihilation operators in static coordinates, with wave vector  $\mathbf{q}$  and in branch  $\lambda$ . The electron-impurity interaction is described by

$$\hat{V}_{ei} = \sum_{\mathbf{q}, a} u(\mathbf{q}) e^{i\mathbf{q} \cdot (\hat{\mathbf{R}} - \mathbf{R}_a)} \hat{\rho}_{\mathbf{q}}, \quad (2.6)$$

with  $\mathbf{R}_a$  the coordinates of impurity,  $u(\mathbf{q})$  the impurity potential, and  $\hat{\rho}_{\mathbf{q}} = \sum_{\mathbf{k}} \hat{c}_{\mathbf{k}+\mathbf{q}}^\dagger \hat{c}_{\mathbf{k}}$  the electron density operator. The electron-phonon interaction is described by

$$\hat{V}_{ep} = \sum_{\mathbf{q}, \lambda} M(\mathbf{q}, \lambda) (\hat{b}_{\mathbf{q}\lambda} + \hat{b}_{-\mathbf{q}\lambda}^\dagger) e^{i\mathbf{q} \cdot \hat{\mathbf{R}}} \hat{\rho}_{\mathbf{q}}, \quad (2.7)$$

with  $M(\mathbf{q}, \lambda)$  the electron-phonon matrix element.

### B. Equations for the drift velocity of electrons

Starting from the quantum-mechanical Liouville equation, one can derive the kinetic equations for a set of macroscopic observables,  $\{\gamma_i(t)\}$ . Here we choose  $\{\gamma_i(t)\} = \{\mathbf{v}(t), f_{\mathbf{k}}(t), n_{\mathbf{q}\lambda}(t)\}$ . The corresponding operators are  $\{\hat{\gamma}_i\} = \{\hat{\mathbf{P}}/M, \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}}, \hat{b}_{\mathbf{q}\lambda}^\dagger \hat{b}_{\mathbf{q}\lambda}\}$  connected with observables by

$$\gamma_i(t) = \text{Tr}[\hat{\gamma}_i \hat{\rho}(t)], \quad (2.8)$$

with  $\hat{\rho}(t)$  the statistical density matrix. The physical meaning of above observables is obvious:  $\mathbf{v}(t)$  is the drift velocity of the electron system,  $f_{\mathbf{k}}$  is the electron distribution function in the relative coordinate, and  $n_{\mathbf{q}\lambda}$  is the phonon occupation number. If we solve the Liouville equation to the lowest order, the rate of change of any function of  $\gamma(t)$ , denoted by  $F(\gamma(t))$ , is given by

$$\frac{\partial F(\gamma(t))}{\partial t} = \frac{i}{\hbar} \langle [\hat{H}_0(t), F(\hat{\gamma}(t))] \rangle + \frac{i}{\hbar} \langle [\hat{V}(t), F(\hat{\gamma}(t))] \rangle - \left[ \frac{i}{\hbar} \right] \lim_{\epsilon \rightarrow 0^+} \int_{-\infty}^t dt' e^{\epsilon(t'-t)} \langle [\dot{F}(\hat{\gamma}(t)), \hat{V}(t')] \rangle, \quad (2.9)$$

where  $\hat{F}(\hat{\varphi}(t)) = -(i/\hbar)[F(\hat{\varphi}(t)), \hat{V}(t)]$ , with  $[\hat{A}, \hat{B}]$  the commutator of operators  $\hat{A}$  and  $\hat{B}$ . For example, we define the frictional force operator,  $\hat{F}$ , as

$$\begin{aligned} \hat{F} &= \frac{-i}{\hbar} [\hat{P}, \hat{V}] \\ &= -i \sum_{q,a} \mathbf{q} u(q) e^{i\mathbf{q} \cdot (\hat{\mathbf{R}} - \mathbf{R}_a)} \hat{\rho}_q \\ &\quad - i \sum_{q,\lambda} \mathbf{q} M(q, \lambda) e^{i\mathbf{q} \cdot \hat{\mathbf{R}}} (\hat{b}_{q\lambda} + \hat{b}_{-q\lambda}^\dagger) \hat{\rho}_q. \end{aligned} \quad (2.10)$$

In Eq. (2.9),  $\hat{V} = \hat{V}'_{ee} + \hat{V}'_{ep} + \hat{V}'_{ei} + \hat{V}'_{pp}$ , and

$$\begin{aligned} \hat{V}(t') &= \exp \left[ (i/\hbar) \int_t^{t'} \hat{H}_0(s) ds \right] \hat{V}(t) \\ &\quad \times \exp \left[ (-i/\hbar) \int_t^{t'} \hat{H}_0(s) ds \right], \end{aligned} \quad (2.11)$$

with  $\hat{H}_0 = \hat{H}_c + \hat{H}'_e + \hat{H}'_p$ .  $\langle \cdots \rangle \equiv \text{Tr}[\hat{\rho}_0(\cdots)]$  with  $\hat{\rho}_0$  the unperturbed density matrix. The operators in Eq. (2.9) are in the interaction representation. In the interaction representation  $\hat{\mathbf{R}}$  changes with time as a "free" particle. Neglecting intracollision effects,<sup>12</sup> one should obtain

$$\begin{aligned} \langle \exp[i\mathbf{q} \cdot \hat{\mathbf{R}}(t)] \exp[-i\mathbf{q} \cdot \hat{\mathbf{R}}(t')] \rangle \\ = \exp[i\mathbf{q} \cdot \mathbf{v}(t)(t - t')], \end{aligned}$$

which leads to the usual Boltzmann transport theory.

However, in the following we use the idea proposed by Ting and Nee.<sup>13</sup> Since the drift motion of an electron, which is described by the center-of-mass motion of electrons,  $\mathbf{R}(t)$ , is superposed upon the random motion of electron in the scattering processes which changes the

trajectory of an electron between scatterers, the memory effect due to this drift motion should be taken into account. On the other hand, this motion could be considered as a classical motion due to the large mass,  $M$ . The factor related to the center-of-mass motion in Eq. (2.9) is then abstracted as

$$\langle \exp[i\mathbf{q} \cdot \hat{\mathbf{R}}(t)] \exp[-i\mathbf{q} \cdot \hat{\mathbf{R}}(t')] \rangle = \exp \left[ i\mathbf{q} \cdot \int_{t'}^t \mathbf{v}(s) ds \right], \quad (2.12)$$

with  $\mathbf{v}(s) = \mathbf{P}(s)/M$ , the drift velocity of the electron system. We emphasize here that this treatment implies a change of operator  $\hat{\mathbf{R}}$  from the interaction representation to the Heisenberg representation and, hence, the approach is to some extent correct beyond the lowest perturbative theory. If we assume that  $\mathbf{v}(s)$  in Eq. (2.12) is a variable weakly dependent on time and instead replace it by  $\mathbf{v}(t)$ , the approach presented here reduces to the usual Boltzmann transport theory. The parameters which describe the distribution of electrons in relative coordinates are still treated in the lowest perturbative theory. These parameters are obtained by using the following assumption for  $\hat{\rho}_0$ :<sup>14</sup>

$$\hat{\rho}_0(t) = \frac{1}{\Xi} \exp \left[ -\sum_i B_i[\gamma_i(t)] \hat{\gamma}_i \right], \quad (2.13)$$

where  $\Xi$  is a normalization constant and the functions  $B_i$  are determined by the requirement

$$\gamma_i(t) = \text{Tr}[\hat{\gamma}_i \hat{\rho}_0(t)]. \quad (2.14)$$

By substituting Eqs. (2.11)–(2.14) into Eq. (2.9), the following evolution equation for drift velocity of electrons,  $\mathbf{v}(t)$ , is obtained:

$$\begin{aligned} Nm \frac{\partial \mathbf{v}(t)}{\partial t} &= -Ne \mathcal{E}(t) - i \sum_{\mathbf{q}} \mathbf{q} \lim_{\epsilon \rightarrow 0^+} \int_{-\infty}^t dt' e^{\epsilon(t-t')} \exp \left[ i\mathbf{q} \cdot \int_{t'}^t \mathbf{v}(s) ds \right] \\ &\quad \times \left[ n_i |u(\mathbf{q})|^2 \Pi(\mathbf{q}, t - t') + \sum_{\lambda} |M(\mathbf{q}, \lambda)|^2 \Lambda^{(+)}(\mathbf{q}, \lambda, t - t') \right], \end{aligned} \quad (2.15)$$

where

$$\Pi(\mathbf{q}, t - t') \equiv \left\langle \frac{-i}{\hbar} [\hat{\rho}_q(t), \hat{\rho}_{-q}(t')] \right\rangle = \frac{-i}{\hbar} \sum_{\mathbf{k}} [f_{\mathbf{k}+\mathbf{q}}(t) - f_{\mathbf{k}}(t)] \exp \left[ \frac{i}{\hbar} (E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}})(t - t') \right], \quad (2.16)$$

$$\begin{aligned} \Lambda^{(\pm)}(\mathbf{q}, \lambda, t - t') &\equiv \left\langle \frac{-i}{\hbar} [\{\pm \hat{b}_{q\lambda}(t) + \hat{b}_{-q\lambda}^\dagger(t)\} \hat{\rho}_q(t), \{\hat{b}_{-q\lambda}(t') + \hat{b}_{q\lambda}^\dagger(t')\} \hat{\rho}_{-q}(t')] \right\rangle \\ &= \mp \frac{i}{\hbar} \sum_{\mathbf{k}} \{ [1 + n_{q\lambda}(t)] f_{\mathbf{k}+\mathbf{q}}(t) [1 - f_{\mathbf{k}}(t)] - n_{q\lambda}(t) f_{\mathbf{k}}(t) [1 - f_{\mathbf{k}+\mathbf{q}}(t)] \} \\ &\quad \times \exp \left[ \frac{i}{\hbar} (E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \hbar \Omega_{q\lambda})(t - t') \right] \\ &+ \frac{i}{\hbar} \sum_{\mathbf{k}} \{ [1 + n_{-q\lambda}(t)] f_{\mathbf{k}}(t) [1 - f_{\mathbf{k}+\mathbf{q}}(t)] - n_{-q\lambda}(t) f_{\mathbf{k}+\mathbf{q}}(t) [1 - f_{\mathbf{k}}(t)] \} \\ &\quad \times \exp \left[ \frac{i}{\hbar} (E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} + \hbar \Omega_{q\lambda})(t - t') \right]. \end{aligned} \quad (2.17)$$

Although the statement in Eq. (2.12) appears reasonable and natural, it is, however, nontrivial, as has been previously pointed out. A possible derivation of this form is obtained by the path-integral approach (see Appendix). In this approach we keep only the classical path for center-of-mass variables of electrons, but make a perturbative expansion only for the relative variables of electrons, so we can treat these two kinds of variables in different ways, which induce the present formulas.

We will study the high frequency "steady state" conductivity. What the "steady state" means is that the drift velocity of electrons can be described as

$$\mathbf{v}(t) = \mathbf{v}_0 + \sum_n \mathbf{v}_n e^{in\omega t}, \quad n = \pm 1, \pm 2, \dots, \quad (2.18a)$$

$$\begin{aligned} \exp \left[ i\mathbf{q} \cdot \int_{t'}^t \mathbf{v}(s) ds \right] &= \exp \left[ -i\mathbf{q} \cdot \mathbf{v}_0 \tau + \sum_n \frac{\mathbf{q} \cdot \mathbf{v}_n}{n\omega} e^{in\omega t} (1 - e^{in\omega \tau}) \right] \\ &= e^{-i\mathbf{q} \cdot \mathbf{v}_0 \tau} \left[ 1 + \sum_n \frac{\mathbf{q} \cdot \mathbf{v}_n}{n\omega} e^{in\omega t} (1 - e^{in\omega \tau}) + \dots \right. \\ &\quad \left. + \frac{1}{m!} \sum_{n_1} \dots \sum_{n_m} \frac{(\mathbf{q} \cdot \mathbf{v}_{n_1}) \dots (\mathbf{q} \cdot \mathbf{v}_{n_m})}{n_1 \dots n_m \omega^m} \prod_{k=1}^m (1 - e^{in_k \omega \tau}) \exp \left[ i\omega t \sum_{k=1}^m n_k \right] + \dots \right]. \end{aligned} \quad (2.19)$$

Since the exponential function is analytic in the whole complex plane of the variable, the above series expansion always converges. By substituting Eqs. (2.18) and (2.19) into Eq. (2.15), we can separate the evolution equation (15) for the drift velocity of electrons to a series of equations for harmonic components of drift velocity. We have

$$\begin{aligned} \mathbf{0} &= -Ne\mathcal{E}_0 + \mathbf{F}_0, \\ in\omega N m \mathbf{v}_n &= -Ne\mathcal{E}_n + \mathbf{F}_n. \end{aligned} \quad (2.20)$$

Here  $\mathbf{F}_n$  is the  $n$ th component of frictional force:

$$\mathbf{F}(t) = \mathbf{F}_0 + \sum_n \mathbf{F}_n e^{in\omega t}, \quad (2.21)$$

with  $\mathbf{F}_{-n} = \mathbf{F}_n^*$ . In the case in which only a few low-harmonic components of electric field are applied we as-

sume the high-harmonic-current components are small enough and can be cut off to some order. By expanding Eq. (2.19) up to third order and keeping  $|n|$  and  $|j|$  up to 2, we obtain that

$$f_{\mathbf{k}}(t) = f_{\mathbf{k}}^{(0)} + \sum_j f_{\mathbf{k}}^{(j)} e^{ij\omega t}, \quad j = \pm 1, \pm 2, \dots, \quad (2.18b)$$

with  $f_{\mathbf{k}}^{(-j)} = f_{\mathbf{k}}^{(j)*}$ . Letting  $\tau = t' - t$ , we make a power expansion for following expression in Eq. (2.15):

sume the high-harmonic-current components are small enough and can be cut off to some order. By expanding Eq. (2.19) up to third order and keeping  $|n|$  and  $|j|$  up to 2, we obtain that

$$\begin{aligned} \mathbf{F}_0 &= \mathbf{F}_0^{(0)} + 2 \operatorname{Re} \mathbf{F}_1^{(-1)} + 2 \operatorname{Re} \mathbf{F}_2^{(-2)}, \\ \mathbf{F}_1 &= \mathbf{F}_1^{(0)} + \mathbf{F}_0^{(1)} + \mathbf{F}_2^{(-1)} + (\mathbf{F}_1^{(-2)})^*, \\ \mathbf{F}_2 &= \mathbf{F}_2^{(0)} + \mathbf{F}_1^{(1)} + \mathbf{F}_0^{(2)}. \end{aligned} \quad (2.22)$$

In  $\mathbf{F}_n^{(j)}$ , the lower index  $n$  is related to the harmonic component of  $\mathbf{v}(t)$ , obtained by expansion of Eq. (2.19), and the upper index  $j$  is related to the harmonic component of  $f_{\mathbf{k}}(t)$  in Eq. (2.18b). We have

$$\begin{aligned} \mathbf{F}_0^{(j)} &= \sum_{\mathbf{q}} \left[ \Gamma_{\mathbf{q}}^{(j)}(0) - \frac{(\mathbf{q} \cdot \mathbf{v}_1)(\mathbf{q} \cdot \mathbf{v}_1^*)}{\omega^2} [2\Gamma_{\mathbf{q}}^{(j)}(0) - \Gamma_{\mathbf{q}}^{(j)}(\omega) - \Gamma_{\mathbf{q}}^{(j)}(-\omega)] \right. \\ &\quad - \frac{(\mathbf{q} \cdot \mathbf{v}_2)(\mathbf{q} \cdot \mathbf{v}_2^*)}{4\omega^2} [2\Gamma_{\mathbf{q}}^{(j)}(0) - \Gamma_{\mathbf{q}}^{(j)}(2\omega) - \Gamma_{\mathbf{q}}^{(j)}(-2\omega)] \\ &\quad + \frac{(\mathbf{q} \cdot \mathbf{v}_2)(\mathbf{q} \cdot \mathbf{v}_1^*)^2}{4\omega^3} [2\Gamma_{\mathbf{q}}^{(j)}(\omega) - 2\Gamma_{\mathbf{q}}^{(j)}(-\omega) - \Gamma_{\mathbf{q}}^{(j)}(2\omega) + \Gamma_{\mathbf{q}}^{(j)}(-2\omega)] \\ &\quad \left. - \frac{(\mathbf{q} \cdot \mathbf{v}_2^*)(\mathbf{q} \cdot \mathbf{v}_1)^2}{4\omega^3} [2\Gamma_{\mathbf{q}}^{(j)}(-\omega) - 2\Gamma_{\mathbf{q}}^{(j)}(\omega) + \Gamma_{\mathbf{q}}^{(j)}(2\omega) - \Gamma_{\mathbf{q}}^{(j)}(-2\omega)] \right], \end{aligned} \quad (2.23)$$

$$\begin{aligned}
\mathbf{F}_1^{(j)} = \sum_{\mathbf{q}} \left[ \frac{\mathbf{q} \cdot \mathbf{v}_1}{\omega} [\Gamma_{\mathbf{q}}^{(j)}(0) - \Gamma_{\mathbf{q}}^{(j)}(\omega)] - \frac{(\mathbf{q} \cdot \mathbf{v}_2)(\mathbf{q} \cdot \mathbf{v}_1^*)}{2\omega^2} [\Gamma_{\mathbf{q}}^{(j)}(0) + \Gamma_{\mathbf{q}}^{(j)}(\omega) - \Gamma_{\mathbf{q}}^{(j)}(2\omega) - \Gamma_{\mathbf{q}}^{(j)}(-\omega)] \right. \\
- \frac{(\mathbf{q} \cdot \mathbf{v}_1)^2(\mathbf{q} \cdot \mathbf{v}_1^*)}{2\omega^3} [3\Gamma_{\mathbf{q}}^{(j)}(0) - 3\Gamma_{\mathbf{q}}^{(j)}(\omega) + \Gamma_{\mathbf{q}}^{(j)}(2\omega) - \Gamma_{\mathbf{q}}^{(j)}(-\omega)] \\
\left. - \frac{(\mathbf{q} \cdot \mathbf{v}_1)(\mathbf{q} \cdot \mathbf{v}_2)(\mathbf{q} \cdot \mathbf{v}_2^*)}{4\omega^3} [2\Gamma_{\mathbf{q}}^{(j)}(0) - \Gamma_{\mathbf{q}}^{(j)}(2\omega) - \Gamma_{\mathbf{q}}^{(j)}(-2\omega) - 2\Gamma_{\mathbf{q}}^{(j)}(\omega) + \Gamma_{\mathbf{q}}^{(j)}(3\omega) + \Gamma_{\mathbf{q}}^{(j)}(-\omega)] \right], \quad (2.24)
\end{aligned}$$

$$\begin{aligned}
\mathbf{F}_2^{(j)} = \sum_{\mathbf{q}} \left[ \frac{\mathbf{q} \cdot \mathbf{v}_2}{2\omega} [\Gamma_{\mathbf{q}}^{(j)}(0) - \Gamma_{\mathbf{q}}^{(j)}(2\omega)] + \frac{(\mathbf{q} \cdot \mathbf{v}_1)^2}{2\omega^2} [\Gamma_{\mathbf{q}}^{(j)}(0) - 2\Gamma_{\mathbf{q}}^{(j)}(\omega) + \Gamma_{\mathbf{q}}^{(j)}(2\omega)] \right. \\
- \frac{(\mathbf{q} \cdot \mathbf{v}_2)^2(\mathbf{q} \cdot \mathbf{v}_2^*)}{16\omega^3} [3\Gamma_{\mathbf{q}}^{(j)}(0) - 3\Gamma_{\mathbf{q}}^{(j)}(2\omega) + \Gamma_{\mathbf{q}}^{(j)}(4\omega) - \Gamma_{\mathbf{q}}^{(j)}(-2\omega)] \\
\left. - \frac{(\mathbf{q} \cdot \mathbf{v}_2)(\mathbf{q} \cdot \mathbf{v}_1)(\mathbf{q} \cdot \mathbf{v}_1^*)}{2\omega^3} [2\Gamma_{\mathbf{q}}^{(j)}(0) - 2\Gamma_{\mathbf{q}}^{(j)}(2\omega) + \Gamma_{\mathbf{q}}^{(j)}(3\omega) - \Gamma_{\mathbf{q}}^{(j)}(-\omega)] \right]. \quad (2.25)
\end{aligned}$$

$\Gamma_{\mathbf{q}}^{(j)}(n\omega)$  in Eqs. (2.23)–(2.25) is given by

$$\begin{aligned}
\Gamma_{\mathbf{q}}^{(j)}(n\omega) = \mathbf{q} \left[ n_i |u(q)|^2 \sum_{\mathbf{k}} (f_{\mathbf{k}+\mathbf{q}}^{(j)} - f_{\mathbf{k}}^{(j)}) \zeta(\mathbf{k}, \mathbf{q}, n\omega - \mathbf{q} \cdot \mathbf{v}_0) \right. \\
\left. + \sum_{\lambda} |M(q, \lambda)|^2 \right. \\
\left. \times \left[ \sum_{\mathbf{k}} [(1 + n_{\mathbf{q}\lambda}) f_{\mathbf{k}+\mathbf{q}}^{(j)} (1 - f_{\mathbf{k}}^{(0)}) - n_{\mathbf{q}\lambda} f_{\mathbf{k}}^{(j)} (1 - f_{\mathbf{k}+\mathbf{q}}^{(0)})] \zeta(\mathbf{k}, \mathbf{q}, \Omega_{\mathbf{q}\lambda} + n\omega - \mathbf{q} \cdot \mathbf{v}_0) \right. \right. \\
\left. \left. - \sum_{\mathbf{k}} [(1 + n_{-\mathbf{q}\lambda}) f_{\mathbf{k}}^{(j)} (1 - f_{\mathbf{k}+\mathbf{q}}^{(0)}) - n_{-\mathbf{q}\lambda} f_{\mathbf{k}+\mathbf{q}}^{(j)} (1 - f_{\mathbf{k}}^{(0)})] \zeta(\mathbf{k}, \mathbf{q}, -\Omega_{\mathbf{q}\lambda} + n\omega - \mathbf{q} \cdot \mathbf{v}_0) \right] \right], \quad (2.26)
\end{aligned}$$

where  $\zeta(\mathbf{k}, \mathbf{q}, x)$ , which is obtained by integrating over  $dt'$  (or  $d\tau$ ) in Eq. (2.15), has the following form:

$$\zeta(\mathbf{k}, \mathbf{q}, x) = \frac{-2i}{(E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \hbar x) + i\epsilon}, \quad (2.27)$$

where the factor of 2 comes from spin degeneracy. In order to obtain Eq. (2.26) we have neglected the oscillating terms in the factor  $[1 - f_{\mathbf{k}}(t)]$ , since we will use the nondegeneracy assumption later.  $\Gamma_{\mathbf{q}}^{(j)}(n\omega)$  has the following symmetry:

$$\Gamma_{-\mathbf{q}}^{(-j)}(-n\omega) = [\Gamma_{\mathbf{q}}^{(j)}(n\omega)]^*. \quad (2.28)$$

One can see from Eqs. (2.26) and (2.27) that the frequency of electric field enters into the expression of energy conservation, Eq. (2.27), in a form of a multiphoton emission or absorption. This quantum effect originates from Eq. (2.12), which includes the memory effect in the theory. Comparing Eq. (2.12) with the assumption, which reduces to the usual Boltzmann equation,  $\mathbf{R}(t) - \mathbf{R}(t') = \mathbf{v}(t)(t - t')$ , we can see that the latter is only approximately valid when  $t - t'$  is less than  $\frac{1}{4}$  of

the period of the applied HF field. This greatly limits the frequency region in which the Boltzmann equation is valid. It indicates the significance of introducing Eq. (2.12), especially in the study of the HF conductivity. Moreover, even in the region of low frequency, use of Eq. (2.12) still benefits us by automatically separating each harmonic component, as shown before.

In the limit of low frequency,  $\omega \rightarrow 0$ , expressions in Eqs. (2.23)–(2.25) are not divergent, since the terms with  $\omega^{-n}$  ( $n > 0$ ) in the Taylor expansion of  $\omega$  (divided by the denominator) completely cancel.

### C. Equation for distribution of hot electrons

In order to obtain a closed set of equations, we need equations for the electron distribution function in the relative coordinates,  $f_{\mathbf{k}}^{(j)}$ . We assume that due to the frequent electron-electron collisions the distribution function of electrons in the relative coordinates  $f_{\mathbf{k}}(t)$  is a Maxwellian distribution at the temperature  $T_e(t)$  (a typical relaxation time for electron-electron scattering is about 30–40 fsec):

$$f_{\mathbf{k}}(t) = \frac{N}{2} \left[ \frac{2\pi\hbar^2\beta(t)}{m} \right]^{3/2} \exp[-E_{\mathbf{k}}\beta(t)], \quad (2.29)$$

with  $\beta(t) = 1/k_B T_e(t)$ . We assume the electron temperature  $T_e(t)$ , or  $\beta(t)$ , oscillates in the following way:

$$\begin{aligned} T_e(t) &= T_0 + \sum_j T_j e^{in\omega t}, \\ \beta(t) &= \beta_0 + \sum_j \beta_j e^{in\omega t}. \end{aligned} \quad (2.30)$$

By expanding to third order and keeping  $|n|$  up to 2 in Eq. (2.30), the relation between amplitudes  $T_n$  and  $\beta_n$  can be obtained:

$$\begin{aligned} k_B T_0 &= \frac{1}{\beta_0} + \frac{1}{\beta_0^3} (2|\beta_1|^2 + 2|\beta_2|^2) \\ &\quad - \frac{1}{\beta_0^4} (3\beta_2\beta_1^{*2} + 3\beta_2^*\beta_1^2) + \dots, \\ k_B T_1 &= \frac{-\beta_1}{\beta_0^2} + \frac{2\beta_2\beta_1^*}{\beta_0^3} \\ &\quad - \frac{1}{\beta_0^4} (3|\beta_1|^2\beta_1 + 6|\beta_2|^2\beta_1) + \dots, \\ k_B T_2 &= \frac{-\beta_2}{\beta_0^2} + \frac{\beta_1^2}{\beta_0^3} \\ &\quad - \frac{1}{\beta_0^4} (3|\beta_2|^2\beta_2 + 6|\beta_1|^2\beta_2) + \dots. \end{aligned} \quad (2.31)$$

A similar expansion is made for  $f_{\mathbf{k}}(t)$  in Eq. (2.29), and we obtain expressions of amplitudes  $f_{\mathbf{k}}^{(j)}$  in terms of  $\beta_n$ 's:

$$\begin{aligned} f_{\mathbf{k}}^{(0)} &= A_{\mathbf{k}} [1 + C_{\mathbf{k}} (2|\beta_1|^2 + 2|\beta_2|^2) \\ &\quad + D_{\mathbf{k}} (3\beta_2\beta_1^{*2} + 3\beta_2^*\beta_1^2)] + \dots, \\ f_{\mathbf{k}}^{(1)} &= A_{\mathbf{k}} [B_{\mathbf{k}}\beta_1 + C_{\mathbf{k}} (2\beta_2\beta_1^*) \\ &\quad + D_{\mathbf{k}} (3|\beta_1|^2\beta_1 + 6|\beta_2|^2\beta_1)] + \dots, \end{aligned} \quad (2.32)$$

$$\begin{aligned} f_{\mathbf{k}}^{(2)} &= A_{\mathbf{k}} [B_{\mathbf{k}}\beta_2 + C_{\mathbf{k}}(\beta_1^2) \\ &\quad + D_{\mathbf{k}} (3|\beta_2|^2\beta_2 + 6\beta_2|\beta_1|^2)] + \dots, \end{aligned}$$

with

$$\begin{aligned} A_{\mathbf{k}} &= \frac{N}{2} \left[ \frac{2\pi\hbar^2\beta_0}{m} \right]^{3/2} \exp(-E_{\mathbf{k}}\beta_0), \\ B_{\mathbf{k}} &= \frac{3}{2\beta_0} - E_{\mathbf{k}}, \\ C_{\mathbf{k}} &= \frac{3}{8\beta_0^2} - \frac{3E_{\mathbf{k}}}{2\beta_0} + \frac{E_{\mathbf{k}}^2}{2}, \\ D_{\mathbf{k}} &= -\frac{1}{16\beta_0^3} - \frac{3E_{\mathbf{k}}}{8\beta_0^2} + \frac{3E_{\mathbf{k}}^2}{4\beta_0} - \frac{E_{\mathbf{k}}^3}{6}. \end{aligned} \quad (2.33)$$

The electron temperature  $T_e(t)$  is related to the energy of electrons in relative coordinates by  $E'_e \equiv \langle \hat{H}'_e \rangle = C_v T_e$ , with  $C_v = 3Nk_B/2$  the specific heat of the electrons. The evolution equation for energy of electrons,  $\partial E'_e(t)/\partial t$ , can be derived in a similar way as that for the drift velocity of electrons. We have

$$\begin{aligned} \frac{\partial E'_e(t)}{\partial t} &\equiv \langle \dot{H}'_e \rangle = -Ne\mathcal{E}(t) \cdot \mathbf{v}(t) - Nm\mathbf{v}(t) \cdot \frac{d\mathbf{v}(t)}{dt} - \langle \dot{H}_p(t) \rangle_{ep} \\ &= -\mathbf{F}(t) \cdot \mathbf{v}(t) - \langle \dot{H}_p(t) \rangle_{ep}. \end{aligned} \quad (2.34)$$

The operator of change of energy of phonons due to electron-phonon interaction is given by

$$\dot{H}_p = \frac{-i}{\hbar} [\hat{H}_p, \hat{V}_{ep}] = -i \sum_{\mathbf{q}, \lambda} M(\mathbf{q}, \lambda) \Omega_{\mathbf{q}\lambda} e^{i\mathbf{q} \cdot \hat{\mathbf{R}}} (-\hat{b}_{\mathbf{q}\lambda} + \hat{b}_{-\mathbf{q}\lambda}^\dagger) \hat{p}_{\mathbf{q}}. \quad (2.35)$$

The average energy change due to electron-phonon interaction can then be written as

$$\langle \dot{H}_p(t) \rangle_{ep} = -i \sum_{\mathbf{q}, \lambda} \Omega_{\mathbf{q}\lambda} \lim_{\epsilon \rightarrow 0^+} \int_{-\infty}^t dt' e^{\epsilon(t-t')} \exp \left[ i\mathbf{q} \cdot \int_{t'}^t \mathbf{v}(s) ds \right] |M(\mathbf{q}, \lambda)|^2 \Lambda^{(-)}(\mathbf{q}, \lambda, t-t'), \quad (2.36)$$

with  $\Lambda^{(-)}(\mathbf{q}, \lambda, t-t')$  is defined in Eq. (2.17). By use of the expansion in Eq. (2.19), and substituting Eq. (2.36) into Eq. (2.34), similar to Eq. (2.20), we obtain a set of equations of harmonic components for the energy of electrons  $E'_e(t)$  or the corresponding components for the electron temperature  $T_e(t)$ :

$$\begin{aligned}
& -(\mathbf{F}_0 \cdot \mathbf{v}_0 + \mathbf{F}_1 \cdot \mathbf{v}_1^* + \mathbf{F}_1^* \cdot \mathbf{v}_1 + \mathbf{F}_2 \cdot \mathbf{v}_2^* + \mathbf{F}_2^* \cdot \mathbf{v}_2) - \langle \dot{H}_p \rangle_0 = 0, \\
& -(\mathbf{F}_2 \cdot \mathbf{v}_1^* + \mathbf{F}_1 \cdot \mathbf{v}_0 + \mathbf{F}_0 \cdot \mathbf{v}_1 + \mathbf{F}_1^* \cdot \mathbf{v}_2) - \langle \dot{H}_p \rangle_1 = i\omega \left[ \frac{3N}{2} k_B T_1 \right], \\
& -(\mathbf{F}_2 \cdot \mathbf{v}_0 + \mathbf{F}_1 \cdot \mathbf{v}_1 + \mathbf{F}_0 \cdot \mathbf{v}_2) - \langle \dot{H}_p \rangle_2 = i2\omega \left[ \frac{3N}{2} k_B T_2 \right].
\end{aligned} \tag{2.37}$$

Here  $\mathbf{F}_0$ ,  $\mathbf{F}_1$ , and  $\mathbf{F}_2$  are given by Eqs. (2.22)–(2.26), and  $\langle \dot{H}_p \rangle_n$  is obtained from corresponding  $\mathbf{F}_n$  by changing  $\Gamma_q^{(j)}(n\omega)$  in Eqs. (2.23)–(2.25) to  $\Phi_q^{(j)}(n\omega)$ , with

$$\begin{aligned}
\Phi_q^{(j)}(n\omega) &= \sum_{\lambda} \Omega_{q\lambda} |M(q, \lambda)|^2 \\
&\times \left[ -\sum_{\mathbf{k}} [(1 + n_{q\lambda}) f_{\mathbf{k}+\mathbf{q}}^{(j)} (1 - f_{\mathbf{k}}^{(0)}) - n_{q\lambda} f_{\mathbf{k}}^{(j)} (1 - f_{\mathbf{k}+\mathbf{q}}^{(0)})] \zeta(\mathbf{k}, \mathbf{q}, \Omega_{q\lambda} + n\omega - \mathbf{q} \cdot \mathbf{v}_0) \right. \\
&\quad \left. - \sum_{\mathbf{k}} [(1 + n_{-q\lambda}) f_{\mathbf{k}}^{(j)} (1 - f_{\mathbf{k}+\mathbf{q}}^{(0)}) - n_{-q\lambda} f_{\mathbf{k}+\mathbf{q}}^{(j)} (1 - f_{\mathbf{k}}^{(0)})] \zeta(\mathbf{k}, \mathbf{q}, -\Omega_{q\lambda} + n\omega - \mathbf{q} \cdot \mathbf{v}_0) \right].
\end{aligned} \tag{2.38}$$

$\Phi_q^{(j)}(n\omega)$  also has the following symmetry:

$$\Phi_{-q}^{(-j)}(-n\omega) = [\Phi_q^{(j)}(n\omega)]^* . \tag{2.39}$$

In this paper we neglect hot-phonon effects and assume the phonons are in equilibrium with a heat bath; the occupation numbers of phonons are then determined by

$$n_{q\lambda} = \frac{1}{\exp(\hbar\Omega_{q\lambda}/k_B T_L) - 1} , \tag{2.40}$$

with  $T_L$  the temperature of the heat bath.

The dynamical screening effect can be included by replacing the  $\Gamma_q^{(j)}(n\omega)$  in Eq. (2.25) and  $\Phi_q^{(j)}(n\omega)$  in Eq. (2.38) by the screened quantities  $\tilde{\Gamma}_q^{(j)}(n\omega)$  and  $\tilde{\Phi}_q^{(j)}(n\omega)$ .

$$\text{Re} \left[ \sum_{\mathbf{k}} E_{\mathbf{k}}^n \exp(-\beta_0 E_{\mathbf{k}}) \zeta(\mathbf{k}, \mathbf{q}, x) \right] = \frac{-m^2}{2\pi\hbar^4 q} (-1)^n \frac{\partial^n}{\partial \beta_0^n} \left[ \frac{1}{\beta_0} \exp(-\eta^2) \right] \tag{2.41}$$

with

$$\eta = \hbar \left[ \frac{\beta_0}{2m} \right]^{1/2} \left[ \frac{q}{2} - \frac{mx}{\hbar q} \right] \tag{2.42}$$

and

$$\begin{aligned}
& \text{Im} \left[ \sum_{\mathbf{k}} E_{\mathbf{k}}^n \exp(-\beta_0 E_{\mathbf{k}}) \zeta(\mathbf{k}, \mathbf{q}, x) \right] \\
&= \frac{-4\pi^{1/2} m^2}{(2\pi)^2 \hbar^4 q} (-1)^n \frac{\partial^n}{\partial \beta_0^n} \left[ \frac{1}{\beta_0} e^{-\eta^2} \int_0^{\eta} e^{x^2} dx \right].
\end{aligned} \tag{2.43}$$

The integral in above equation is related to Dawson's integral.<sup>15</sup>

The latter are obtained by dividing each term in Eqs. (2.25) and (2.38) by the corresponding dielectric function  $\epsilon(\mathbf{q}, \bar{\omega})$ , where  $\bar{\omega}$  is the frequency argument of the  $\zeta$  function in each term. The dielectric function,  $\epsilon(\mathbf{q}, \bar{\omega})$ , can be calculated, for example, in the random-phase approximation.

Equations (2.20) and (2.37), together with Eqs. (2.31) and (2.32), consist of a set of equations for determining the magnitudes and phases of the harmonic components of the drift velocity,  $\mathbf{v}_n$ , and the electron temperature  $T_j$  (or  $\beta_j$ ).

If degeneracy is neglected, namely,  $(1 - f_{\mathbf{k}}^{(0)})$  is replaced by 1, the summation over  $\mathbf{k}$  in Eqs. (2.26) and (2.38), then, can be worked out. Typical terms in  $\sum_{\mathbf{k}} f_{\mathbf{k}}^{(j)} \zeta(\mathbf{k}, \mathbf{q}, x)$  in Eq. (2.26) are obtained as follows:

### III. LINEAR HIGH-FREQUENCY CONDUCTIVITY

We first briefly discuss the case in which a weak HF electric field is applied, without or with a strong dc electric field.

#### A. Cooling-electron case

If we apply only a weak HF field (along the  $x$  direction) on the sample,

$$\mathcal{E}(t) = \mathcal{E}_1 (e^{i\omega t} + e^{-i\omega t}) , \tag{3.1}$$

the electron distribution in the relative coordinates is assumed to be in equilibrium with the heat bath, and has only the component  $f_{\mathbf{k}}^{(0)} = A_{\mathbf{k}}$  in Eq. (2.33), with  $\beta_0 = 1/k_B T_L$ . By taking only the linear term in Eq.

(2.24), our formulas reduce to a simple equation given by Lei, Horing, and Zhang,<sup>5</sup>

$$i\omega Nm \mathbf{v}_1 = -Ne \mathcal{E}_1 + \sum_{\mathbf{q}} \frac{\mathbf{q} \cdot \mathbf{v}_1}{\omega} [\Gamma_{\mathbf{q}}^{(0)}(0) - \Gamma_{\mathbf{q}}^{(0)}(\omega)]. \quad (3.2)$$

From Eq. (3.2) it leads directly to the well-known expression of linear conductivity,  $\sigma(\omega)$ , in terms of the memory function (see, for example, Götze and Wölfle<sup>4</sup>):

$$\sigma(\omega) = i \frac{Ne^2}{m} \frac{1}{\omega + M(\omega)}, \quad (3.3)$$

with the memory function  $M(\omega)$  given by

$$M(\omega) = \sum_{\mathbf{q}} \frac{-iq_x}{Nm\omega} [\Gamma_{q_x}^{(0)}(0) - \Gamma_{q_x}^{(0)}(\omega)]. \quad (3.4)$$

The calculations of HF conductivity based on the memory-function method or Kubo's formula have been worked out.<sup>3-5</sup>

### B. Hot-electron case

If a weak ac field is applied together with a strong dc field (both are along the  $x$  direction), the electrons are heated by the dc field. The hot-electron effect on the high-frequency conductivity has previously been studied based on the Boltzmann transport equation.<sup>6-9</sup> We keep only the zero- and first-harmonic components and cut off the expansion up to linear order in the case of weak HF field. Two sets of equations then are obtained. One set of equations for the zero-harmonic components is given by

$$\begin{aligned} -Ne \mathcal{E}_0 + \mathbf{F}_0^{(0)} &= 0, \\ -\mathbf{F}_0^{(0)} \cdot \mathbf{v}_0 - \langle \dot{H}_p \rangle_0^{(0)} &= 0, \end{aligned} \quad (3.5)$$

with  $\mathbf{F}_0^{(0)} \approx \sum_{\mathbf{q}} \Gamma_{\mathbf{q}}^{(0)}(0)$  and  $\langle \dot{H}_p \rangle_0^{(0)} \approx \sum_{\mathbf{q}} \Phi_{\mathbf{q}}^{(0)}(0)$ . This set of equations is independent of the first-harmonic components, and can be solved as in the case in which only a static field exists. The solutions of  $\mathbf{v}_0$  and  $\beta_0$  provide a bias for calculation of the linear differential conductivity for a weak HF field. Another set of equations for  $\mathbf{v}_1$  and  $T_1$  (or  $\beta_1$ ) is given by

$$\begin{aligned} i\omega Nm \mathbf{v}_1 &= -Ne \mathcal{E}_1 + \mathbf{F}_1^{(0)} + \mathbf{F}_1^{(1)}, \\ -i\omega \frac{3\beta_1 N}{2\beta_0^2} &= -[(\mathbf{F}_1^{(0)} + \mathbf{F}_1^{(1)}) \cdot \mathbf{v}_0 + \mathbf{F}_0^{(0)} \cdot \mathbf{v}_1] \\ &\quad - \langle \dot{H}_p \rangle_1^{(0)} - \langle \dot{H}_p \rangle_0^{(1)}, \end{aligned} \quad (3.6)$$

with

$$\begin{aligned} \mathbf{F}_1^{(0)} &\approx \frac{\mathbf{v}_1}{\omega} \cdot \sum_{\mathbf{q}} \mathbf{q} [\Gamma_{\mathbf{q}}^{(0)}(0) - \Gamma_{\mathbf{q}}^{(0)}(\omega)], \\ \langle \dot{H}_p \rangle_1^{(0)} &\approx \frac{\mathbf{v}_1}{\omega} \cdot \sum_{\mathbf{q}} \mathbf{q} [\Phi_{\mathbf{q}}^{(0)}(0) - \Phi_{\mathbf{q}}^{(0)}(\omega)], \end{aligned} \quad (3.7)$$

$$\begin{aligned} \mathbf{F}_0^{(1)} &\approx \sum_{\mathbf{q}} \Gamma_{\mathbf{q}}^{(1)}(0), \\ \langle \dot{H}_p \rangle_0^{(1)} &\approx \sum_{\mathbf{q}} \Phi_{\mathbf{q}}^{(1)}(0). \end{aligned} \quad (3.8)$$

The superscript (1) in Eq. (3.8) is related to the first-harmonic component of the distribution function,  $f_{\mathbf{k}}^{(1)} \approx A_{\mathbf{k}} B_{\mathbf{k}} \beta_1$  in Eq. (2.32), with  $A_{\mathbf{k}}$  and  $B_{\mathbf{k}}$  given by Eq. (2.33). Equation (3.6), therefore, is a set of linear equations for  $\mathbf{v}_1$  and  $\beta_1$  provided by the bias parameters  $\mathbf{v}_0$  and  $\beta_0$ . The structure of these sets of equations is similar to that obtained by Das and Ferry.<sup>7</sup> Since they derived their formulas from the Boltzmann transport equation, the coefficients in Eq. (3.6) of the present paper have an expression different from that of Das and Ferry. In Ref. 7, the coefficients in the linear equations corresponding to our Eq. (3.6) are obtained based on the analytical expressions of the derivative of the relaxation times. Our formulas do not require a special analytical expression, and, therefore, it is easy to generalize them to more complicated cases, such as two-dimensional electrons in a quantum well.

We have calculated the linear HF conductivity of hot electrons in bulk  $n$ -type GaAs and in a GaAs-Ga<sub>1-x</sub>Al<sub>x</sub>As quantum well. Detailed numerical results are planned to be published elsewhere.<sup>16</sup> The total trend of HF mobility of electrons as a function of  $\omega$  is in agreement with that of Ref. 7. We found, however, that resonance peaks appear in the curves of the real part of HF mobility and the amplitude of oscillating electron temperature,  $T_1$ , near  $\omega = \Omega_{\text{LO}}$ , with  $\Omega_{\text{LO}}$  the frequency of the longitudinal-optical phonon. This effect originates from the appearance of the frequency  $\omega$  of the electric field in the energy-conservation  $\delta$  function, and this resonance could not be obtained from the Boltzmann theory.

### IV. NONLINEAR HF CONDUCTIVITY

We have used the approach shown in Sec. II to calculate the nonlinear conductivity in an  $n$ -type GaAs bulk sample when a strong HF electric field is applied. The HF field is applied up to 1 kV/cm, and the effect of higher valleys, which is important when the strength of the field reaches 3–4 kV/cm, is neglected in the present calculation. A weak dc electric field  $\mathcal{E}_0 = 10$  V/cm is also applied. The base frequency of the HF electric field is chosen in the microwave region,  $\nu = 35$  GHz. The lattice temperature  $T_L = 300$  K. The electron–longitudinal-optical (LO)-phonon scattering through polar interaction is the dominant scattering mechanism at this temperature, and we neglect electron scatterings with impurities and other kinds of phonons. The electron–LO-phonon interaction is described by the Fröhlich model:  $M(\mathbf{q}) = \alpha/q^2$ , with the Fröhlich coupling constant,  $\alpha = [2\pi e^2 \hbar \Omega_{\text{LO}} (1/\epsilon_{\infty} - 1/\epsilon_0)]^{1/2}$ ; here the energy of the LO phonon  $\hbar \Omega_{\text{LO}} = 36.2$  meV, the static dielectric constant  $\epsilon_0 = 12.91$ , and the high-frequency dielectric constant  $\epsilon_{\infty} = 10.91$ . The effective mass of the electron  $m = 0.067m_0$ , with  $m_0$  the free-electron mass. The density of electrons is chosen as  $N = 10^{17}/\text{cm}^3$ , which, in fact, is irrelevant in the calcu-



lation as long as a Maxwell distribution is assumed. The screening effect is neglected in present calculation, since the dynamic screening is weak for electron-LO-phonon scattering.

We have calculated the dc velocity  $v_0$  and the harmonic components of velocity  $v_n(t) = |v_n| \cos(n\omega + \phi_{v_n})$  ( $n = 1, 2$ ), and corresponding components of the electron temperature  $T(t)$ , as functions of the strength of the applied HF electric field,  $\mathcal{E}_1 \cos(\omega t)$  and  $\mathcal{E}_2 \cos(2\omega t)$ . This calculation requires us to solve a set of 10 coupled nonlinear equations. We use an optimization program of the AT&T Bell laboratories Mathematical Subroutine Library Port<sup>17</sup> based on the algorithm of an improved Newton method. The accuracy generally is  $10^{-8}$ – $10^{-9}$ . The results are shown in Figs. 1–6. Three cases are considered: (1)  $\mathcal{E}_2 : \mathcal{E}_1 = 0 : 1$  (dotted curves), (2)  $\mathcal{E}_2 : \mathcal{E}_1 = 0.4 : 1$  (cross curves), and (3)  $\mathcal{E}_2 : \mathcal{E}_1 = 1 : 1$  (solid curve). Figure 1 shows that the dc conductivity is only reduced by about 5% from its static value, up to  $\mathcal{E}_1 = 1$  kV/cm if only  $\mathcal{E}_1$  is applied. Surprisingly, if a second-harmonic electric field is applied together with the first-harmonic one (with the same phase), the dc conductivity decreases dramatically with increasing the HF field. As shown by the solid curve in Fig. 1, if  $\mathcal{E}_2 : \mathcal{E}_1 = 1 : 1$ , the dc drift velocity definitely becomes negative when  $\mathcal{E}_1$  and  $\mathcal{E}_2$  are near 1 kV/cm, namely, a weak positive dc potential drop can produce a negative dc current. Figures 2 and 3 show that the HF components of the electron drift velocity increase approximately linearly with an increase of the HF electric field in the region of HF field  $\mathcal{E}_1 < 1$  kV/cm. The second-harmonic components  $|v_2|$  induced by the pure  $\mathcal{E}_1$  field are less than  $10^{-3}$  of value of  $|v_1|$ , as shown in Fig. 3

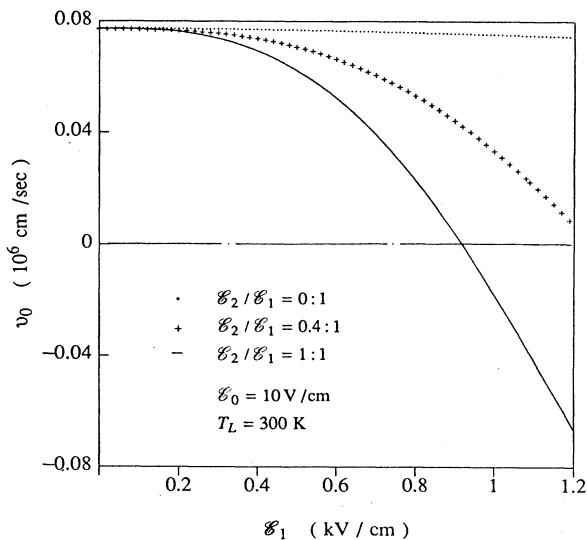


FIG. 1. The direct current component of the electron drift velocity,  $v_0$ , as a function of the strength of high-frequency electric field,  $\mathcal{E}_1$ .  $\mathcal{E}_1 \cos(\omega t)$  and  $\mathcal{E}_2 \cos(2\omega t)$  are applied together with (1)  $\mathcal{E}_2 : \mathcal{E}_1 = 0$  (dotted curve), (2)  $\mathcal{E}_2 : \mathcal{E}_1 = 0.4 : 1$  (cross curve), and (3)  $\mathcal{E}_2 : \mathcal{E}_1 = 1 : 1$  (solid curve). The frequency  $\nu = \omega/2\pi = 35$  GHz.

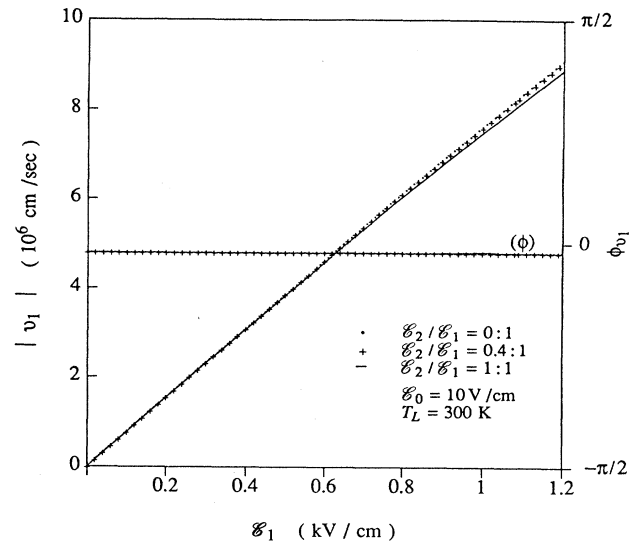


FIG. 2. The first-harmonic component of the electron drift velocity,  $|v_1| \cos(\omega t + \phi_{v_1})$ , as a function of  $\mathcal{E}_1$ . The lines labeled by  $(\phi)$  are those for phase  $\phi_{v_1}$ .

(dotted curve). The retarded phases, respectively,  $\phi_{v_1} \approx -3.7^\circ$  and  $\phi_{v_2} \approx -7.3^\circ$ , remain constant with increasing HF field. Figure 4 shows the heating effect of the HF electric field. The average temperature  $T_0$  increases with increasing HF field, roughly proportional to  $\mathcal{E}_1^2$  as anticipated. Figures 5 and 6 show the HF components of the electron temperature as functions of  $\mathcal{E}_1$  and  $\mathcal{E}_2$ . We see that the second-harmonic component  $T_2$  induced by  $\mathcal{E}_1$  is large, but the first-harmonic component  $T_1$  is small (see dotted curves). A large  $T_2$  appears be-

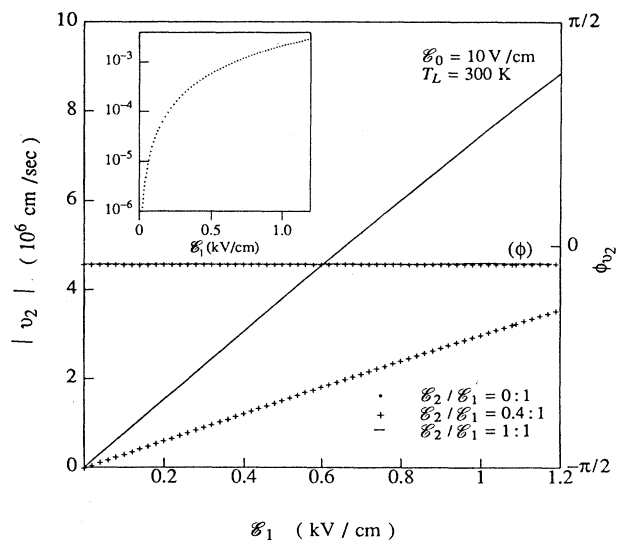


FIG. 3. The second-harmonic component of the electron drift velocity,  $|v_2| \cos(2\omega t + \phi_{v_2})$ , as a function of  $\mathcal{E}_1$ . The lines labeled by  $(\phi)$  are those for phase  $\phi_{v_2}$ . Inset:  $|v_2|$  as a function of  $\mathcal{E}_1$  for the case that  $\mathcal{E}_2 : \mathcal{E}_1 = 0 : 1$ .

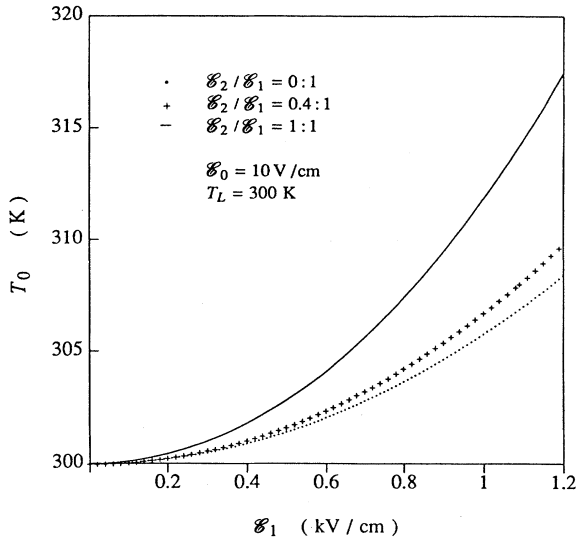


FIG. 4. The average electron temperature  $T_0$  as a function of  $\mathcal{E}_1$ .

cause the input power to the electron system in relative coordinates is  $-Ne\mathcal{E}(t)\cdot\mathbf{v}(t) - M\mathbf{v}(t)\cdot d\mathbf{v}(t)/dt$ , and a large  $\mathcal{E}_1$  and  $v_1$  should produce a large second-harmonic component in the power input to the energy of electrons in the relative coordinates. On the other hand, terms such as  $\mathcal{E}_1v_0, \mathcal{E}_1v_2, \dots$ , which contribute to the first-harmonic component  $T_1$ , are relatively small. Figures 5 and 6 also show that the amplitudes of oscillating components,  $|T_1|$  and  $|T_2|$ , usually have the same order of magnitude as that of the average increase of electron temperature  $T_0 - T_L$  in the microwave region, and

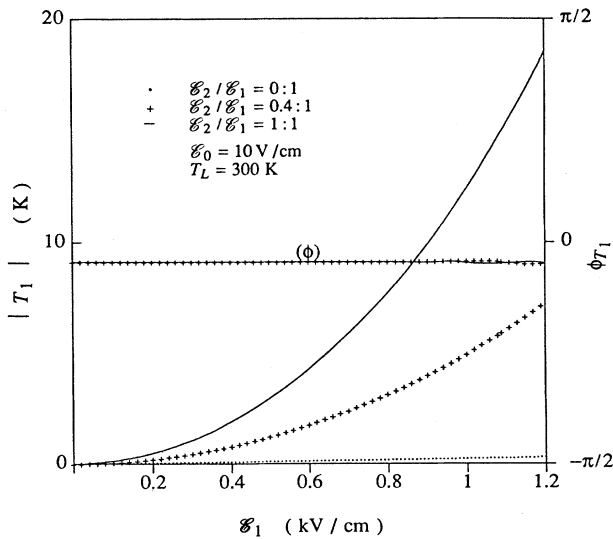


FIG. 5. The first-harmonic component of the electron temperature,  $|T_1|\cos(\omega t + \phi_{T_1})$ , as a function of  $\mathcal{E}_1$ . The lines labeled by  $(\phi)$  are those for phase  $\phi_{T_1}$ .

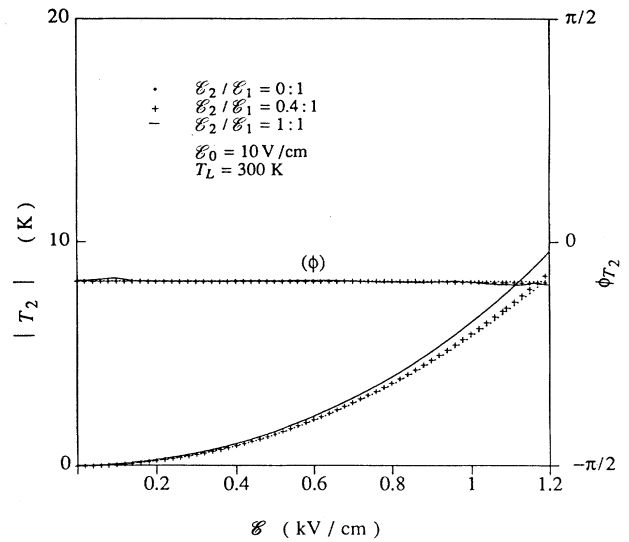


FIG. 6. The second-harmonic component of the electron temperature,  $|T_2|\cos(2\omega t + \phi_{T_2})$ , as a function of  $\mathcal{E}_1$ . The lines labeled by  $(\phi)$  are those for phase  $\phi_{T_2}$ .

should not be neglected. The retarded phases are  $\phi_{T_1} \approx -7.8^\circ$  and  $\phi_{T_2} \approx -17^\circ$ .

The appearance of the negative dc conductivity was explained by Pozhela<sup>1</sup> as due to formation of the high-field domain in the sample. This induced electric field exceeds the threshold of the negative differential mobility, which is about 3–4 kV/cm for GaAs. They also showed, in a qualitative argument, that the HF rectification effect in this region of negative differential mobility may induce a negative dc conductivity. Our calculation indicates another possible origin of the negative dc conductivity; that is, production and amplification of higher-harmonic components of the field and current (may be applied or induced). Since this possibility does not require formation of a high-field domain, it may be easier to develop. For understanding the origin of this negative dc conductivity induced by a HF field, further experiments and theoretical study are needed. Our calculation also warns that, in the measurement of the drift velocity of electrons by use of the integral microwave technique, it is important to avoid the appearance of the high-harmonic components.

## V. DISCUSSION

In summary, we have developed a new analytical method, which is tractable, though not easy, to study the carrier transport in a strong HF electric field. In view of the derivation of our formulas, our theory may be also valid in the higher-frequency region where the Boltzmann theory is not available. We have calculated the electron transport in an *n*-type GaAs sample in a microwave field. Our results show an important mixing frequency effect which produces a negative dc conductivity. Finally, we conclude this paper with a few remarks.

(1) It is obvious that our approach is a balance-

equation (BE) method. Recently, there have been some arguments about the validity of the balance-equation method.<sup>18,19</sup> It has been shown that in the weak-field limit the conductivity based on the balance-equation method disagrees with that obtained from the Boltzmann equation or the Kubo formula, when the lattice temperature is nonzero. This is a so-called “ $\langle 1/\tau \rangle \neq 1/\langle \tau \rangle$ ” paradox.<sup>18–20</sup> We notice that this paradox appears because both the BE method and the Boltzmann theory are compared in the realm of a noninteracting-electron theory. In order to simplify the infinite-degrees-of-freedom problem of the dynamics of carriers, people often introduce a finite number of parameters to describe the carrier system, and use a finite number of balance equations to evaluate these parameters. One of the BE methods is based on the assumption that the strong electron-electron interaction equilibrates electrons, so that the term  $(\partial f_{\mathbf{k}}/\partial t)_{ee}$  in the dynamical equation becomes zero. This BE method, from our understanding, is an approximate approach for treating interacting electrons. In fact, we have indicated<sup>21</sup> that the BE approach<sup>22</sup> can be equivalent to the Boltzmann-equation method provided the distribution function of electrons is assumed to be a drifted Fermi-Dirac distribution. In the weak-field limit, if only electron-impurity scattering is considered, we know that from the Boltzmann equation for noninteracting electrons the distribution function of electrons can be obtained as  $f_{\mathbf{k}} = f_{\mathbf{k}}^0 + f_{\mathbf{k}}^1$ , with  $f_{\mathbf{k}}^0$  the equilibrium distribution of electrons without an electric field and

$$f_{\mathbf{k}}^1 = f_{\mathbf{k}}^0 \beta \frac{e\hbar}{m} \mathbf{k} \cdot \mathcal{E} \tau_k. \quad (5.1)$$

In Eq. (5.1)  $\tau_k$  is the well known energy dependent scattering time, the expression of which is given, for example, in Eq. (5) of Ref. 20;  $\beta = 1/(k_B T_L)$ . On the other hand, if we assume the distribution function of electrons is a drift Maxwellian distribution due to strong electron-electron interaction (the change of electron temperature is proportional to  $\mathcal{E}^2$ , and is zero in the weak-field limit), by expanding the distribution function to linear term of velocity we have

$$f_{\mathbf{k}}^1 = f_{\mathbf{k}}^0 \beta \frac{e\hbar}{m} \mathbf{k} \cdot \mathcal{E} \bar{\tau}, \quad (5.2)$$

with  $\bar{\tau} = \langle 1/\tau_k \rangle^{-1}$ . We see that due to electron-electron interaction the distribution function of electrons changes by taking a constant  $\bar{\tau}$  instead of  $\tau_k$  and is more isotropic. This difference between Eqs. (5.1) and (5.2) immediately shows that the conductivity of electrons obtained by this BE method is different from that deduced by the Boltzmann equation and the Kubo formula for noninteracting electrons, as shown in Refs. 18–20. This is reasonable and acceptable. In fact, comparing theories which derive this kind of balance equation, we find that there are some assumptions. For example, the distribution of electrons in the relative coordinates at  $t = -\infty$  is assumed to be of Fermi-Dirac type at a temperature  $T_e$ ,<sup>22</sup> also the Welton-Callen-Kubo thermal fluctuation-dissipation theorem with temperature  $T_e$  is used.<sup>23</sup> These assumptions reduce the complexity of solv-

ing a problem for the whole distribution function by solving two (momentum and energy) balance equations, and, according to our opinion, also leads to the conclusion that this BE approach is equivalent to the drifted-temperature model. It is interesting that the temperature model used in the realm of the Boltzmann theory<sup>21,22</sup> can also be inserted into a non-Boltzmann approach, so as to include the memory effect for drift motion<sup>13</sup> or high orders of electron-impurity and electron-phonon interaction.<sup>23</sup> There are other BE methods, such as the classical moment-expansion method of Chapman and Cowling, to solve the Boltzmann equation,<sup>24</sup> or recent work by Su *et al.*,<sup>25</sup> in which a Gaussian approximation is used. This type of BE method, of course, is for noninteracting carriers, and should obtain the same result as that from the Kubo formula in the weak-field limit.

(2) In our calculation, we keep terms up to second-harmonic components for drift velocity and electron temperature. We also make the expansion up to third order. Although the series expansions in Eqs. (2.19), (2.31), and (2.32) are convergent, the higher-harmonic components and the higher terms of expansions may be important with increasing applied electric field. There is no difficulty, in principle, to include higher-order terms; however, it will require more computing time. It is also important to include the intervalley scattering in the high-field region, since it induces the Gunn effect. A multicarrier model<sup>26</sup> of the BE method is available; however, this will double the number of equations. Our theory could be easily extended to the quasi-two-dimensional electron system, as in a heterostructure. It is also interesting to study the nonlinear transport of carriers in a higher-frequency field. These shall constitute a future subject for study.

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#### APPENDIX

As we pointed before, the formulas in the text, especially Eq. (2.12), imply a generalization beyond the lowest perturbation theory. This generalization has not been proven. In this Appendix we use the path-integral formulation for the closed-time-path Green's function<sup>23,25,27</sup> (CTPGF) to derive the formulas in the text. The clue to our derivation is similar to that presented by Su *et al.*,<sup>25</sup> but the system we deal with is a system of many electrons rather than the single electron system in Ref. 25.

For simplifying the description, we consider an electron-LO-phonon system. The total Lagrangian is given by

$$\begin{aligned}
L(t) &= \frac{1}{2}M\dot{\mathbf{R}}(t)^2 - eN\mathcal{E}(t)\cdot\mathbf{R}(t) \\
&+ \sum_{\mathbf{k}} c_{\mathbf{k}}^\dagger(t)S^{-1}(\mathbf{k})c_{\mathbf{k}} + \frac{1}{2}\sum_{\mathbf{q}} B_{-\mathbf{q}}(t)\Delta^{-1}(\mathbf{q})B_{\mathbf{q}} \\
&+ \sum_{\mathbf{q}} M(\mathbf{q})B_{\mathbf{q}}(t)e^{i\mathbf{q}\cdot\mathbf{R}(t)}\rho_{\mathbf{q}}(t), \tag{A1}
\end{aligned}$$

where  $B_{\mathbf{q}} = b_{\mathbf{q}} + b_{-\mathbf{q}}^\dagger$  and  $\rho_{\mathbf{q}} = \sum_{\mathbf{k}} c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}$ ;  $S(\mathbf{k})$  and  $\Delta(\mathbf{q})$  are, respectively, the free propagators of the electron and phonon. Their expressions can be found, for example, in Eqs. (2.21) and (2.26) of Ref. 27. The meanings of other notations are the same as that in the text.

The expectation of an arbitrary observable  $O$  can be expressed as the following functional integral which is defined on the closed time path,

$$\langle\langle O \rangle\rangle = \int [d\mathbf{R}][dB_{\mathbf{q}}][dc_{\mathbf{k}}^\dagger][dc_{\mathbf{k}}] O[\mathbf{R}, B_{\mathbf{q}}, c_{\mathbf{k}}^\dagger, c_{\mathbf{k}}] \exp(iI_p[\mathbf{R}, B_{\mathbf{q}}, c_{\mathbf{k}}^\dagger, c_{\mathbf{k}}]). \tag{A2}$$

The action defined on the closed time path is given by

$$\begin{aligned}
I_p &= \int_p dt L(t) = \int_p dt \left[ \frac{1}{2}M\dot{\mathbf{R}}(t)^2 - eN\mathcal{E}(t)\cdot\mathbf{R}(t) + \sum_{\mathbf{q}} M(\mathbf{q})B_{\mathbf{q}}(t)e^{i\mathbf{q}\cdot\mathbf{R}(t)}\rho_{\mathbf{q}}(t) \right. \\
&\quad \left. + \frac{1}{2}\int_p dt' \sum_{\mathbf{q}} B_{-\mathbf{q}}(t)\Delta^{-1}(\mathbf{q}; t, t')B_{\mathbf{q}}(t') + \int_p dt' \sum_{\mathbf{k}} C_{\mathbf{k}}^\dagger(t)S^{-1}(\mathbf{k}; t, t')C_{\mathbf{k}}(t') \right], \tag{A3}
\end{aligned}$$

with

$$\int_p dt = \int_{-\infty}^{+\infty} dt_+ + \int_{+\infty}^{-\infty} dt_- = \int_{-\infty}^{+\infty} dt_+ - \int_{-\infty}^{+\infty} dt_-. \tag{A4}$$

Since the  $B_{\mathbf{k}}$  dependence of  $I_p$  is quadratic, the functional integration over the phonon variable  $\int [dB_{\mathbf{q}}]$  in Eq. (A2) can be exactly worked out. We obtain an effective action  $I_{\text{eff}}[\mathbf{R}, C_{\mathbf{k}}^\dagger, c_{\mathbf{k}}]$ , which is defined by

$$\exp(iI_{\text{eff}}[\mathbf{R}, c_{\mathbf{k}}^\dagger, c_{\mathbf{k}}]) = \int [dB_{\mathbf{q}}] \exp(iI_p[\mathbf{R}, B_{\mathbf{q}}, c_{\mathbf{k}}^\dagger, c_{\mathbf{k}}]). \tag{A5}$$

We have

$$\begin{aligned}
I_{\text{eff}}[\mathbf{R}, c_{\mathbf{k}}^\dagger, c_{\mathbf{k}}] &= \int_p dt \left[ \frac{1}{2}M\dot{\mathbf{R}}(t)^2 - eN\mathcal{E}(t)\cdot\mathbf{R}(t) \right] + \int_p dt \int_p dt' \sum_{\mathbf{k}} c_{\mathbf{k}}^\dagger(t)S^{-1}(\mathbf{k}; t, t')c_{\mathbf{k}}(t') \\
&\quad + \frac{1}{2}\sum_{\mathbf{q}} |M(\mathbf{q})|^2 \int_p dt \int_p dt' \Delta(\mathbf{q}; t, t') e^{i\mathbf{q}\cdot[\mathbf{R}(t) - \mathbf{R}(t')]} \rho_{\mathbf{q}}(t)\rho_{-\mathbf{q}}(t'). \tag{A6}
\end{aligned}$$

Also, the following formula is useful for eliminating the phonon variables:

$$\langle\langle (\dots) B_{\mathbf{q}}(t) \rangle\rangle = - \int_p ds M^*(\mathbf{q})\Delta(\mathbf{q}; t-s) \langle\langle (\dots) e^{-i\mathbf{q}\cdot\mathbf{R}(s)} \rho_{-\mathbf{q}}(s) \rangle\rangle, \tag{A7}$$

where

$$\begin{aligned}
\langle\langle (\dots) \rangle\rangle &= \int [d\mathbf{R}][dB_{\mathbf{q}}][dc_{\mathbf{k}}^\dagger][dc_{\mathbf{k}}] (\dots) \exp(iI_p), \\
\langle (\dots) \rangle &= \int [d\mathbf{R}][dc_{\mathbf{k}}^\dagger][dc_{\mathbf{k}}] (\dots) \exp(iI_{\text{eff}}). \tag{A8}
\end{aligned}$$

From the Lagrangian in Eq. (A1), the operator equation for the center-of-mass motion of electrons is obtained as

$$\dot{\mathbf{P}}(t) = -eN\mathcal{E}(t) + i\sum_{\mathbf{q}} M(\mathbf{q})\mathbf{q}B_{\mathbf{q}}(t)\exp[i\mathbf{q}\cdot\mathbf{R}(t)]\rho_{\mathbf{q}}(t).$$

Taking the expectation value of the nonequilibrium ensemble, we have

$$\begin{aligned}
\langle \dot{\mathbf{P}}(t) \rangle &= M \frac{d\mathbf{v}(t)}{dt} = -eN\mathcal{E}(t) + i\sum_{\mathbf{q}} \langle\langle M(\mathbf{q})\mathbf{q} \exp[i\mathbf{q}\cdot\mathbf{R}(t)] \rho_{\mathbf{q}}(t) B_{\mathbf{q}}(t) \rangle\rangle \\
&= -eN\mathcal{E}(t) - i\sum_{\mathbf{q}} |M(\mathbf{q})|^2 \mathbf{q} \int_p ds \langle e^{i\mathbf{q}\cdot\mathbf{R}(t)} \rho_{\mathbf{q}}(t) e^{-i\mathbf{q}\cdot\mathbf{R}(s)} \rho_{-\mathbf{q}}(s) \rangle \Delta(\mathbf{q}; t-s). \tag{A9}
\end{aligned}$$

Here  $\mathbf{v}(t)$  is the drift velocity of electrons. Until now we did not make any approximation. In the following we assume

that the center-of-mass motion of electrons is a classical motion due to its huge mass; therefore, we neglect the functional integration  $\int [d\mathbf{R}]$  and take only the classical path:

$$\exp\{i\mathbf{q}\cdot[\mathbf{R}(t) - \mathbf{R}(s)]\} \approx \exp[i\mathbf{q}\cdot\langle\mathbf{R}(t) - \mathbf{R}(s)\rangle] = \exp\left[i\mathbf{q}\cdot\int_s^t \mathbf{v}(\tau)d\tau\right]. \quad (\text{A10})$$

Equation (A9) is now written as

$$M\frac{d\mathbf{v}(t)}{dt} = -eN\mathcal{E}(t) - i\sum_{\mathbf{q}} |M(\mathbf{q})|^2 \mathbf{q} \int_p ds \exp\left[i\mathbf{q}\cdot\int_s^t \mathbf{v}(\tau)d\tau\right] \Delta(\mathbf{q};t-s)\Pi(\mathbf{q};t,s), \quad (\text{A11})$$

with

$$\Pi(\mathbf{q};t,s) = \langle T_p[\rho_{\mathbf{q}}(t)\rho_{-\mathbf{q}}(s)] \rangle, \quad (\text{A12})$$

where  $T_p$  is the time-ordering operator along the closed time path.  $\langle \dots \rangle$  is defined in Eq. (A8) without integration  $\int [d\mathbf{R}]$ .  $\Pi(\mathbf{q};t,s)$  is related to the density-density correlation function in relative coordinates. It is difficult to calculate it completely (note:  $c_{\mathbf{k}}^\dagger$  and  $c_{\mathbf{k}}$  are Grassman numbers). However, its lowest-order (free) term is easily obtained by making a perturbative expansion of  $\exp(iI_{\text{eff}})$  and taking only the term of zero order of  $[M(\mathbf{q})]$ . Then, following the standard closed-time-path integral method (see Sec. V and Appendix in Ref. 23), we directly obtain Eqs. (2.15) and (2.17) in the text.

It is easy to include the electron-impurity interaction in our formulas. The electron-electron interaction term can be introduced without difficulty in our derivation, since this term is only related to the electron variable in relative coordinates. In the limit of very strong electron-

electron scattering, electrons are in equilibrium with each other and the term reflecting electron-electron scattering in the dynamical equation disappears. The procedure of derivation of the dynamical equation in the strong electron-electron interaction limit is the same as that without the electron-electron interaction; however, the distribution functions for these two cases are different from each other. The former in relative coordinates should be Fermi-Dirac (or Maxwellian) distribution, described by temperature; the latter is non-equilibrium and is described by infinite parameters.

The difference between this derivation and the usual perturbative expansion is that we first take the classical approximation for the center-of-mass motion of electrons and make the perturbation expansion only for the electron variable in relative coordinates. The usual perturbation theory, on the other hand, makes perturbative expansion for both center-of-mass and relative-variables of electrons to the lowest order; therefore,  $\langle\mathbf{R}(t) - \mathbf{R}(s)\rangle = \mathbf{v}(t)(t-s)$  in that case, which induces to the Boltzmann theory.

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