PHYSICAL REVIEW B VOLUME 28, NUMBER 2 15 JULY 1983

Langevin equation for hot-electron problems

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(Received 23 February 1983)

In this paper, we build a Langevin equation for the macrovariables of an electronic system in contact with a lattice, when the entire system is in the hot-electron regime. A bridge is established between this equation and earlier ones which give the thermodynamic motion of these microvariables. Finally, by focusing on the Langevin equation for the electronic momentum of the system, we examine the causes of the failure of the first fluctuation-dissipation theorem in its usual form in the stationary (far-from-equilibrium) situation.

I. INTRODUCTION

The hot-electron problem in semiconductor transport is a classic problem in the area of nonequilibrium statistical mechanics. This problem has received considerable attention in the past two decades,[1] but the majority of this work has centered on the long-time limit applicable to the Boltzmann equation. With the advent of very-small semiconductor devices in which the drift length (in a high electric field) is of the same length as that necessary for momentum and energy relaxation to occur, it has become necessary to examine more fully the short-time limit, one which has received only limited attention to date. In this region, it is possible in fact to find and use a Langevin equation approach to treat the transport problem. The derivation of a Langevin equation for the problem of hot-electron transport is twofold: First, for a theoretical basis, it is required that one understand how fluctuations build up during the evolution of the carrier system toward the nonequilibrium steady state (the stationary state), and second, it is also necessary to examine the structure of the fluctuating forces themselves once the system reaches this stationary state.

In addition to the above problems, it is also necessary to study the resulting Langevin equation itself in the stationary state. In particular, this study should yield insight into the limits of application of the first fluctuation-dissipation theorem (the so-called Kubo fluctuation-dissipation theorem) that has previously been discussed in a somewhat different context by Proccacia *et al.*[2] Indeed, it has long been supposed that this theorem is invalid in the nonequilibrium, hot-carrier regime.[3] The presence, or lack, of this theorem is crucial, since for practical reasons it is necessary to be able to predict the noise characteristics of these very-small semiconductor devices.

From first principles, one can choose either a mechanical approach such as the Langevin equation[4,5] or a thermodynamic approach in which the time evolution of the relevant macroscopic variables (the macrovariables) is determined. In this work we shall choose the latter approach and build toward a Langevin equation by using the general principles described earlier by Zubarev.[6] This is done in Sec. II. Then, in Sec. III, our derivation of a conceptual framework for the Langevin equation is presented. This latter approach has several analogies with the earlier work of Grabert,[7] although the present point of view and approach differ from this latter work. Finally, in the remaining sections, we make the connection to the thermodynamic equations for the macrovariables previously treated by Kalashnikov[8] and Ferry[9] by using the Langevin equation obtained in Sec. IV. We end by discussing the problems which come into play with the first fluctuation-dissipation theorem. We find that this latter approach cannot be treated in a consistent manner, although the second fluctuation-dissipation theorem remains valid.

II. THE QUASIEQUILIBRIUM STATISTICAL DISTRIBUTION

In the approach to be used here, we treat an electronic system in a semiconductor lattice. One cannot decouple this sytem from the lattice if a complete and accurate representation is to be achieved. We therefore take the Hamiltonian of the system to be

$$H = H_e + H_{\text{ef}} + H_{e\text{-}L} + H_L \ , \tag{1}$$

where H_e is the complete electronic Hamiltonian (including the electron-electron interaction), $H_{e\text{-}L}$ is the electron-lattice interaction, H_{ef} is the time-independent electric field term, and H_L is the Hamiltonian of the lattice variables (usually limited to those terms representing the phonon field).

Following earlier work in this approach,[6–11] we introduce a set of thermodynamically relevant variables (the macrovariables) of the problem. There is no absolute rule governing such an approach. Rather, one must be led in this by physical intuition, and the aim of this approach is to be able to describe in a deterministic manner all the important thermodynamic properties of the system. We note also that the choice of the pertinent macrovariables used is critical, as this choice and the definition of the entropy operator for the far-from-equilibrium system are two aspects of the same problem.

In the problem at hand, we choose to use the electronic energy H_e, the total momentum of the electronic system $\vec{P}_e$, the energy of the lattice system H_L, and the momentum of the lattice system (which is identically zero). In order to maintain a grand canonical framework, we add to the previous set the number operator N_e for the electrons.

Many people approach the treatment of this system by the introduction of a "drifted Maxwellian" distribution function, in which the density matrix of the system is assumed to be not too far from[12]

$$\rho_{L,0}=\exp\left[-\phi_0-\sum_m F_m(t)P_m\right] , \tag{2}$$

where the individual P_m's are the previously defined macrovariables and the F_m's are the thermodynamically conjugate forces associated with the macrovariables. In addition, the quantity $\phi_0=\ln(\mathrm{Tr}\rho_{L,0})$ assures the proper normalization of the system by introducing a quantity similar to a partition function.

However, the above choice of $\rho_{L,0}$ is not convenient, primarily because it does not give the proper equilibrium density matrix when $\vec{F}=0$ ($\langle\vec{P}_e\rangle=0$ as well) and when $\beta_e=\beta_L$ (β_e and β_L are the inverse temperatures of the electronic and lattice systems, respectively). In other words, if we write the Liouville operator equation with the total Hamiltonian of (1), but without the electric field, we would like any irreversibility to arise only from the fact that the trial density matrix does not obey the Liouville equation when the system is not in equilibrium. If we write the density matrix in the linear-response form $\rho=\rho_L+\Delta\rho$, the Liouville equation for the deviation term is ($\hbar=1$)

$$i\frac{\partial\Delta\rho}{\partial t}-[H,\Delta\rho]=[H,\rho_l]-i\frac{\partial\rho_L}{\partial t} . \tag{3}$$

Thus in order to fit the above requirements, we must introduce terms in $H_{e\text{-}L}$ into $\rho_{L,0}$.

To accomplish the above goals, we introduce the term $H_{e\text{-}L}$ as a thermodynamic variable as well. To be consistent, however, we assign it the conjugate force β_L. We can then define the quasiequilibrium statistical distribution (QESD) as

$$\rho_L=\exp\left[-\phi-\sum_m F_m(t)P_m\right] , \tag{4}$$

where

$$\{P_m\}=\{\vec{P}_e,H_e,N_e,H_L,H_{e\text{-}L}\}$$

and

$$\{F_m\}=\{-\beta_e\vec{V}_D,\beta_e,-\beta_e(\mu-\tfrac{1}{2}mV_D^2),\beta_L,\beta_L\} .$$

The values of the F_m's are such as to make $\langle P_m\rangle=\mathrm{Tr}\{\rho_L P_m\}$ for the first four of the macrovariables. In the following, we shall use the reduced chemical potential μ' for the quantity $\mu-mV_D^2/2$. We further define $\sigma(t)$ as

$$\sigma(t)=\phi+\sum_m P_m F_m(t) .$$

We will find it useful in the following first to derive two important properties of the mean values that are calculated with the help of ρ_L, the QESD. These are ($h=1$)

$$\langle\dot{P}_{m,L}\rangle_{\rho_L}=i\,\mathrm{Tr}(\rho_L[H_e,P_m])=0 , \tag{5}$$

$$\sum_m\langle\dot{P}_m\rangle_{\rho_L}F_m=i\sum_m F_m\mathrm{Tr}(\rho_L[H,P_m])=0 , \tag{6}$$

where the time to which ρ_L and F_m refer has been dropped. Here the notation $\dot{P}_{m,L}$ means the time variation introduced by $i[H_{e\text{-}L},P_m]$. We first note that the property (5) is trivial if $P_m=H_{e\text{-}L}$. To demonstrate (5) for the other four macrovariables, we note that

$$[\rho_L,H_{e\text{-}L}]=\frac{1}{\beta_L}[\rho_L,-\beta_eH_e+\beta_e\vec{V}_D\vec{P}_e+\beta_e\mu'N_e-\beta_LH_L] .$$

Now, by writing $\langle\dot{P}_{m,L}\rangle=i\,\mathrm{Tr}(P_m[\rho_L,H])$ and using the above expression for $[\rho_L,H_{e\text{-}L}]$, it becomes easy to demonstrate (5) by recognizing that the first four macrovariables mutually commute. The property (5) implies that we can prepare our system in a state defined by the QESD (4) so that the physical irreversibility which arises from $H_{e\text{-}L}$ has no effect on the motion, at least during the time in which the system resides in a stationary state. We will see below that (5) is essential for the derivation of a Langevin equation describing the motion of the first four macrovariables.

We can also easily establish (6). We first write the average of the time variation of the quantity

$$\sum_m\langle\dot{P}_m\rangle_{\rho_L}F_m=i\sum_m F_m\mathrm{Tr}(H[P_m,\rho_L])$$
$$=i\,\mathrm{Tr}\left[H\left[\sum_m F_mP_m,\rho_L\right]\right] .$$

The result (6) arises from the evident commutation of ρ_L and $\sum_m F_mP_m$.

We pointed out earlier that the macrovariables are defined through the averaging process $\langle P_m\rangle=\mathrm{Tr}(\rho P_m)$ $=\mathrm{Tr}(\rho_L P_m)$. It is therefore evident that

$$\delta\langle P_m\rangle=-\sum_n(P_m;P_n)^t\delta F_n \tag{7a}$$

and

$$\langle\dot{P}_m\rangle=-\sum_m(P_m;P_n)^t\dot{F}_n , \tag{7b}$$

where the correlation function $(P_m;P_n)^t$ is defined by

$$(P_m;P_n)^t=\int_0^1 d\tau\,\mathrm{Tr}[P_me^{-\tau\sigma(t)}(P_n-\langle P_n\rangle^t)\times e^{(\tau-1)\sigma(t)}] . \tag{8}$$

In the following, we will use this correlation function extensively, and will employ the same formal definition even if P_m and P_n are time-dependent quantities.

Finally to conclude this section, we note one more interesting property. If P_n and P_m are two of the macrovariables in the hot-electron problem, we obtain

$$(\dot{P}_{n,L};P_m)^t=0 . \tag{9}$$

This relation is readily obtained from (5), which we can apply both for ρ_L (associated with the near-equilibrium force F_m) and for $\rho_L+\delta\rho_L$ (associated with the modified force $F_m+\delta F_m$). Using this combination, we then obtain

$$\langle\dot{P}_{m,L}\rangle_{\rho_L+\delta\rho_L}=\langle\dot{P}_{m,L}\rangle_{\rho_L}+\sum_n(\dot{P}_{m,L};P_n)\delta F_n ,$$

from which (9) is an obvious result.

III. THE DISSIPATIONLESS MOTION

The Langevin equation describes the relaxation of mechanical variables of a given system that is near equilibrium. This result was first formally derived with the help of the projection operator technique by Mori.[4] Such an approach yields[13]

$$\begin{aligned}\dot{A} &= e^{iLt}DiLA + e^{(1-D)iLt}(1-D)iLA \\ &\quad + \int_0^t ds\, e^{iL(t-s)}DiLe^{i(1-D)Ls}(1-D)iLA \ , \end{aligned} \tag{10}$$

where A is taken to mean "the set of coupled mechanical variables of the system," L is the Liouville operator, and D is a projection operator.

Usually one constructs the operator D so that terms such as the second one on the right-hand side (rhs) of (10) represent the fluctuating force in the system. We would like at this point to emphasize a physical principle that leads one to also consider the first term on the rhs of (10) when constructing D. We consider this technique by applying the following treatment to our particular problem.

In the framework of the derivation of the Langevin equation (10), the system is generally assumed to be in equilibrium at the initial time $t=0$. Any initial value of A at this time is assumed to arise from a static field applied in the far distant past. Moreover, this field is turned off at $t=0$. At time $t=0$, the "speed' of the variables A is given only by the first and second terms on the right of (10). Since the second term is a fluctuation term by construction, the average speed of A is

$$\langle \dot{A} \rangle(0) = \langle iDLA \rangle(0) \ ,$$

and there is no dissipation associated with this particular motion. This simple, but physical, fact is clear if we recall that at $t=0$, when the applied field is switched off, the system still has time reversal symmetry and the fluctuations cannot, by themselves, induce any dissipation.

We now want to turn to the construction of the projection operator D. We first write

$$\langle \dot{A} \rangle(0) = \mathrm{Tr}[\rho(t=0)A] \ ,$$

and it now is easy to show that, in the framework of linear-response theory, we have

$$\langle \dot{A} \rangle(0) = \Omega\langle A \rangle(0) \ , \tag{11}$$

where $\Omega = (\dot{A};A)(A;A)^{-1}$. The correlation functions which appear in Ω correspond to the equilibrium situation, $\langle A \rangle = 0$. These formal expressions are the same as those given in (8), where σ is now recognized as the entropy of the equilibrium case. From the above, we can now define D through

$$DB = (B;A)(A;A)^{-1}A \ . \tag{12}$$

At the initial time $t=0$, the meaning of the first term of (9) is clear. It is this term that yields the mechanical state of the system that is being defined by the macrovariables A at time t, and these in turn represent the nondissipative components of the "speed" of these variables. If we use (12) to define D, we still must check to be sure that the second term on the right of (10) does indeed represent a fluctuation force. This, in fact, is trivial and will not be carried out here.

To construct the Langevin equation for the P_m's, and more precisely the projection operator to be utilized in our hot-electron problem, we are going to use mainly the above arguments. However, we introduce some slight differences due to the existence of the far-from-equilibrium stationary state in the present problem. We begin by preparing our system at $t=0$ in a thermodynamic state in which the density matrix is given by $\rho_L(t=0)$, i.e., $\rho(t=0)=\rho_L(t=0)$. We know from (5) that the source of the irreversibility in the system does not induce any dissipation into the motion of $\langle P_m \rangle$ at this initial time $t=0$. Further, a given P_m can be written ($\langle P_m \rangle$ is assumed to be $\langle P_m \rangle^0$)

$$P_m = \langle P_m \rangle + (P_m - \langle P_m \rangle) \ . \tag{13}$$

If we seek the particular evolution of P_m, for which there is no dissipation at $t=0$, we suppose that the evolution of the fluctuation $(P_m - \langle P_m \rangle)$ is the same as that for $\langle \delta P_m \rangle$ induced by some generalized force δF_m $(t=0)$. (This is a form of an Onsager hypothesis and is consistent with the fact that δP_m should be given by $\delta\rho_L$ at $t=0$ to ensure dissipationless motion.) We therefore introduce a dissipationless motion at $t=0$ as

$$\left.\frac{dP_m}{dt}\right|_{\mathrm{DL}} = \mathrm{Tr}(\rho_L \dot{P}_m) + \sum_n \Omega_{mn}(P_n - \langle P_n \rangle) \ , \tag{14}$$

where the matrix Ω is given by

$$\Omega = (\dot{P};P)(P;P)^{-1} \ . \tag{15}$$

As before, the correlation functions are given by (8). The thermodynamical parameters which enter these correlation functions are given by our initial condition $\rho_L(t=0)$. From (14), it is easy to see that the difference between dP_m/dt and $(dP_m/dt)|_{\mathrm{DL}}$ is just a fluctuation term.

We are now in a position to define the relevant projection operator, at least at $t=0$. This becomes

$$\begin{aligned}\Pi(0)X &= \mathrm{Tr}[\rho_L(t=0)X] \\ &\quad + \sum_{j,k} (X;P_j)(P;P)^{-1}_{jk}(P_k - \langle P_k \rangle) \ , \end{aligned} \tag{16}$$

and at $t=0$

$$\left.\frac{dP}{dt}\right|_{\mathrm{DL}} = \Pi(0)iLP \ . \tag{17}$$

Here, and in the following, we shall take P to be the column matrix whose elements are the individual P_m's. Thus all equations involving P (without an index) must be interpreted as having matrix products and sums.

IV. THE LANGEVIN EQUATION

Let us now turn to the $t \neq 0$ case. With the previous considerations in mind, we will assume that the dissipationless motion of the P_m is given by an expression analogous to (14), as

$$\left.\frac{dP_m}{dt}\right|_{\mathrm{DL}} = \mathrm{Tr}[\rho_L(t)P_m] + \sum_m \Omega_{mn}[P_n(t) - \langle P_n \rangle^t] \ , \tag{18}$$

where the matrix Ω has the definition (15), in which the correlation functions are calculated with the appropriate thermodynamical parameters at time t. By defining $\Pi(t)$ as

$$\Pi(t)X = \mathrm{Tr}[\rho_L(t)X] + \sum_{j,k} (X;P_j)(P;P)^{-1}_{jk}(P_k - \langle P_k \rangle^t) \ , \qquad (19)$$

we can write

$$\left.\frac{dP}{dt}\right|_{\mathrm{DL}} = e^{iLt} i\Pi(t)LP \ . \qquad (20)$$

It is obvious from (19) that $\Pi(t)$ is a projection operator. This operator corresponds to one introduced in a somewhat different manner by Grabert.[7] In Appendix A, we list several important properties of this operator. The derivation of a Langevin equation, with the help of (20), is now just a matter of technical algebra. We first write

$$\frac{dP}{dt} = e^{iLt} i\Pi(t)LP + e^{iLt} i[1-\Pi(t)]LP \ . \qquad (21)$$

Introducing the operator $G(s,t)$ as a two-time Green's function given by[7,14]

$$G(s,t) = \mathrm{Tr}\exp\left[i \int_s^t du \, L[1-\Pi(u)] \right] ,$$

and satisfying the differential equations

$$\frac{dG(s,t)}{dt} = iG(s,t)L[1-\Pi(t)] \ , \qquad (22a)$$

$$\frac{dG(s,t)}{ds} = -iL[1-\Pi(s)]G(s,t) \ , \qquad (22b)$$

it is easy to establish the following identity:

$$G(s,t) = e^{iL(t-s)} - \int_s^t ds' e^{-iL(s-s')} iL\Pi(s')G(s',t) \ . \qquad (23)$$

The property given by (23) can now be used to express the operator e^{iLt} in the second term of (21) within the framework usually utilized for the derivation of a Langevin equation, and[13,14]

$$e^{iLt}[1-\Pi(t)]LP = G(0,t)[1-\Pi(t)]LP + \int_0^t ds' e^{iLs'} iL\Pi(s') \times G(s',t)[1-\Pi(t)]LP . \qquad (24)$$

In order to examine the characteristic features of (24), we have to define precisely our initial condition. For this, we take $\rho(t=0)=\rho_0$, where ρ_0 is the density matrix corresponding to the equilibrium situation where the field is zero. We therefore assume that the field is applied at $t=0$ (when $\beta_e = \beta_L$, $\langle \vec{\mathrm{V}}_e \rangle = 0$). This initial condition corresponds to a particular ρ_L. Now it appears that the first term of (24) is not a fluctuating force for this particular initial condition. Nevertheless, we can always write it as

$$G(0,t)[1-\Pi(t)]LP = [1-\Pi(0)]G(0,t)[1-\Pi(t)]LP + \Pi(0)G(0,t)[1-\Pi(t)]LP \ , \qquad (25)$$

where the first term on the right is a fluctuating force for our initial condition, as is pointed out in Appendix A.

The second term on the right of (24) is now integrated by parts, and the first term of (24) is replaced using (25), so that we arrive at

$$\begin{aligned}\dot{P}(t) = {} & e^{iLt}\Pi(t)\dot{P} \\ & + \int_0^t ds' e^{iLs'}\Pi(s') iL[1-\Pi(s')]G(s',t)\dot{P} \\ & + [1-\Pi(0)]G(0,t)\dot{P} \\ & + \int_0^t ds' e^{iLs'}\dot{\Pi}(s')G(s',t)[1-\Pi(t)]\dot{P} \ , \end{aligned} \qquad (26)$$

where the first term gives the dissipationless motion (which may be compared to the $t=0$ situation). The second term on the right of (26) can now be divided into two parts, one of which characterizes the dissipation and the other of which leads to a fluctuating force caused by the nonequilibrium nature of the system (we return to the latter term in Sec. VI below). The third and fourth terms are two additional fluctuating forces, the former of which is Mori-type in nature and the latter characterizes the fluctuations induced by the "speed" of macrovariables during the transient regime.

Equation (26) is now a Langevin equation for the macrovariables, and its derivation does not include any approximations. In the next section we focus our attention on the first two terms on the rhs of (26) and establish that the evolution of the $\langle P_m \rangle$ given by (26) is the same as that obtained by beginning with the framework introduced by Zubarev[6] for nonequilibrium thermodynamics (and used subsequently for the hot-electron problem by Kalashnikov[8] and Ferry[9]). For this we use an expansion to second order in $H_{e\text{-}L}$, which is the relevant approximation level of the electron-phonon interaction in the Kalashnikov-Ferry results.

Later, we focus attention on (26) as a Langevin equation for the electronic momentum $\vec{\mathrm{P}}_e$. In so doing, we will neglect the coupling between the other macrovariables.[15] However, we will examine the reason for the failure of the so-called fluctuation-dissipation theorem[3] in its usual form.

V. THE THERMODYNAMICAL MOTION

From the form of (26), it is easy to determine the average motion of the macrovariables. This can be written as

$$\langle \dot{P}(t) \rangle = \langle e^{iLt}\Pi(t)\dot{P} \rangle + \left\langle \int_0^t ds' e^{iLs'}\Pi(s') iL[1-\Pi(s')]G(s',t)\dot{P} \right\rangle , \qquad (27)$$

where

$$\langle X \rangle = \mathrm{Tr}(\rho_0 X) \ .$$

Equation (27), in which $\langle \dot{P}(t) \rangle$ is given by (7b), gives a closed set of equations describing the evolution of the parameters F_n. Up to second order in the interaction term $H_{e\text{-}L}$, the Zubarev form has been shown earlier to yield[9]

$$\langle \dot{P}_m(t)\rangle = \sum_n \alpha_{mn}\langle P_n(t)\rangle + \sum_n \int_0^t dt'(\dot{P}_{m,L};\dot{P}_{n,L}(-t))^t F_n(t-t') , \tag{28}$$

where $\dot{P}_{m,L}$ was defined in Sec. II and corresponds to the effect of $H_{e\text{-}L}$ on the time derivative of P_m. In (28), the α_{mn} arise from the fact that the individual macrovariables are coupled in the hot-electron case. Here, the Liouville equation

$$\dot{P}_m = iL_0 P_m ,$$

where L_0 is the Liouville operator in the absence of the interaction between the electrons and the phonons, can be written

$$\dot{P}_m = \sum_n \alpha_{mn} P_n . \tag{29}$$

More precisely, if $H_{e\text{-}L}=0$, the definition of α_{mn} is given by

$$\begin{aligned} &\dot{\vec{\mathrm{P}}}_e = eN_e\vec{\mathrm{F}}, \quad \dot{N}_e = 0 \\ &\dot{H}_e = \frac{e\vec{\mathrm{F}}}{m}\vec{\mathrm{P}}_e, \quad \dot{H}_L = 0 . \end{aligned} \tag{30}$$

Here $\vec{\mathrm{F}}$ is the applied electric field.

To prove the equivalence of (27) and (28), we first look at the first two terms. The equality of these terms is easily established through (5). Let us now turn to the dissipation terms, which we shall examine only up to second order in the interaction $H_{e\text{-}L}$. As a starting point, we take the term from (27) as

$$\begin{aligned} T &= \left\langle \int_0^t ds' e^{iLs'}\Pi(s')iL[1-\Pi(s')]G(s',t)\dot{P}\right\rangle \\ &= \int_0^t ds'\mathrm{Tr}\{\rho_L(s')iL[1-\Pi(s')]G(s',t)\dot{P}\} , \end{aligned} \tag{31}$$

where the last equality comes from Appendix A. To establish the connection with the equivalent form in (28), we proceed in three steps. We first show that

$$T = \sum_n \int_0^t ds' F_n(s')([1-\Pi(s')]G(s',t)\dot{P};\dot{P}_n)^{s'} . \tag{32}$$

This can be established by recognizing that the properties of the operator L under a trace operation allow us to write (31) as

$$T = \int_0^t ds'\mathrm{Tr}\{[-iL\rho_L(s')][1-\Pi(s')]G(s',t)\dot{P}\} . \tag{33}$$

Using the Kubo formula for the commutator, T takes the form

$$T = \sum_n \int_0^t ds' F_n(s') \int_0^1 d\alpha\, \mathrm{Tr}\left[\exp\left[-\alpha\sum_j F_j(s')P_j\right]\dot{P}_n \exp\left[\alpha\sum_k F_k(s')P_k\right]\cdots\rho_L(s')[1-\Pi(s')]G(s',t)\dot{P}\right] , \tag{34}$$

which can be written as (32) by using (6) and the definition (8) for the correlation functions.

For the second part of the proof, we want to show that (32) can be written in the form

$$T = \sum_n \int_0^t ds' F_n(s')(G(s',t)[1-\Pi(t)]\dot{P};[1-\Pi(s')]\dot{P}_n)^{s'} . \tag{35}$$

This can be established from (32) by recognizing that the operator $G(s',t)$ has the general property (which can be established by its expansion and the properties of Appendix A)

$$[1-\Pi(s')]G(s',t)\dot{P} = [1-\Pi(s')]G(s',t)[1-\Pi(t)]\dot{P} . \tag{36}$$

We then note that since $\Pi(s')$ is a projection operator associated with the scalar product $(A;B)^{s'}$, defined by (8), the result (35) follows immediately.

We now show that

$$T = \sum_n \int_0^t dt'(\dot{P}_{,L};\dot{P}_{n,L}(-t))^t F_n(t-t') , \tag{37}$$

where $\dot{P}_{,L}$ is a general notation for the $\dot{P}_{m,L}$'s. The value of T given by (37) is the expected result that ensures the equivalent of (27) and (28). This final result can be established by making use of an expansion in $H_{e\text{-}L}$ up to second order, an approximation not made up to now. We start by noticing that

$$[1-\Pi(s')]\dot{P}_n = \dot{P}_{n,L} , \tag{38}$$

which is easily stated with the help of (9), so that

$$T = \sum_n \int_0^t ds' F_n(s')(G(s',t)\dot{P}_{,L};\dot{P}_{n,L})^{s'} . \tag{39}$$

In order to determine T up to second order in $H_{e\text{-}L}$, it is adequate to obtain the expansion of $G(s',t)\dot{P}_{,L}$ only up to zero order in $H_{e\text{-}L}$. This follows from the examination of the form of (39). In Appendix B we establish that this may be achieved by writing

$$G(s',t)\dot{P}_{,L} = \dot{P}_{,L}(t-s') .$$

It therefore turns out that T can now be written as

$$T = \sum_n \int_0^t ds'(\dot{P}_{,L}(t-s');\dot{P}_{n,L})^{s'}F_n(s') ,$$

which is not exactly the form (37). This is because the correlation functions are not evaluated at the same time. However, we can write $t-s'=u$, so that the correlation function we must evaluate is

$$(\dot{P}_{,L}(u);\dot{P}_{n,L})^{t-u} = \int_0^1 d\alpha\,\mathrm{Tr}[\dot{P}_{,L}(u)e^{-\alpha\sigma(t-u)}\dot{P}_{n,L}e^{\alpha\sigma(t-u)}\rho_L(t-u)]\;, \tag{40}$$

where the product $\langle \dot{P}_{,L}\rangle\langle \dot{P}_{n,L}\rangle$ has been omitted since it is of fourth order in the interaction. The evaluation of (40) to second order in the interaction requires that our knowledge of ρ_L and $e^{\alpha\sigma}$ only has to be up to zero order. Up to this order, we can write (here u is always limited to a characteristic time)

$$\rho_L(t)=e^{-iHu}\rho_L(t-u)e^{iHu}\;, \tag{41a}$$

$$e^{\alpha\sigma(t)}=e^{-iHu}e^{\alpha\sigma(t-u)}e^{iHu}\;. \tag{41b}$$

We can now write (40) as

$$(\dot{P}_{,L}(u);\dot{P}_{n,L})^{t-u}=(\dot{P}_{,L};\dot{P}_{n,L}(-u))^{t}\;,$$

which gives (37) for T.

Having shown the equivalence of the two formulations (namely, the Langevin equation and the Zubarev approach of nonequilibrium thermodynamics), at least for the evaluation of the average motion of the macrovariables, we would like to now look more carefully at the fluctuation forces. To do so, we will neglect the cross-correlation functions in the dissipation terms of (26) [which has been shown earlier to be an acceptable procedure if $mV_e^2 \ll \langle H_e\rangle$ (Ref. 8)]. We will also neglect these cross correlations in the fluctuating forces in order to be consistent. Finally, we will limit our study to the Langevin equation of the momentum $\vec{\mathbf{P}}_e$ of the carriers.

VI. DIFFICULTIES WITH THE FIRST FLUCTUATION-DISSIPATION THEOREM

To proceed, let us now write the equation for the carrier momentum, noting that we are interested only in that component P_e along the electric field. By using the results of Sec. V, and the definition of the parameter that is thermodynamically conjugate to P_e, i.e., $-\beta_e V_D$, we can write the equation as

$$\begin{aligned}\frac{dP_e}{dt}={}&eFN_e-\int_0^t \beta_e(t-t')V_D(t-t')(\dot{P}_{e,L};\dot{P}_{e,L}(-t'))^t dt'\\&+\int_0^t ds'[P_e(s')-mN_eV_D(s')](iL[1-\Pi(s')]G(s',t)\dot{P}_e;P_e)^{s'}((P_e;P_e)^{s'})^{-1}\\&+[1-\Pi(0)]G(0,t)\dot{P}_e+\frac{1}{N_em}\int_0^t ds'[P_e(s')-mN_eV_D(s')]\dot{V}_D(s')\mathrm{Tr}\left[\frac{\partial^2\rho_L}{\partial V_D^2}(s')G(s',t)[1-\Pi(t)]\dot{P}_e\right]\;.\end{aligned} \tag{42}$$

At this time, we wish to group the terms which represent random forces into two groups:

$$\phi(s',t)=(iL[1-\Pi(s')]G(s',t)\dot{P}_e;P_e)^{s'}[(P_e;P_e)^{s'}]^{-1}\;, \tag{43}$$

$$\Psi(s',t)=\frac{V_D(s')}{N_em}\mathrm{Tr}\left[\frac{\partial^2\rho_L}{\partial V_D^2}(s')G(s',t)[1-\Pi(t)]\dot{P}_e\right]\;. \tag{44}$$

The last kernel, (44), is only different from zero in the case of a nonstationary situation. The first kernel, (43), can be examined by expanding the terms up to second order in the interaction, as

$$\phi(s',t)=\frac{1}{(P_e;P_e)^{s'}}\int_0^1 d\alpha\,\mathrm{Tr}\{\rho_L(s')[H,[1-\Pi(s')]G(s',t)\dot{P}_e]e^{-\alpha\sigma(s')}P_e e^{\alpha\sigma(s')}\}\;.$$

Now if H commutes with $\rho_L(s')$ (and hence with $e^{\pm\alpha\sigma(s')}$), then $\phi(s',t)$ reduces to

$$\begin{aligned}\phi(s',t)&=\frac{-1}{(P_e;P_e)^{s'}}([1-\Pi(s')]G(s',t)\dot{P}_e;\dot{P}_e)^{s'}\\&=\frac{-1}{(P_e;P_e)^{s'}}(\dot{P}_{e,L};\dot{P}_{e,L}(-t'))^t\;,\end{aligned}$$

where the last equality is valid up to second order in the interaction.

However, because irreversibility is involved in the problem at hand, ρ_L does not commute with the Hamiltonian H, and we must write

$$\phi(s',t)=\frac{1}{(P_e;P_e)^{s'}}[-(\dot{P}_{e,L};\dot{P}_{e,L}(-t'))^t+I_R(s',t)]\;, \tag{45}$$

where I_R can be evaluated in the classical limit (in which quantum fluctuations are ignored) up to second order as

$$I_R(s',t)=i\,\mathrm{Tr}([\rho_L(s'),H]\{[1-\Pi(s')]\dot{P}_e(t-s')\}P_e)\;. \tag{46}$$

The correlation function $(P_e;P_e)^{s'}$ in (45) can be evaluated with (7a) and we get

$$(P_e;P_e)^{s'}=Nm\beta_e^{-1}(s') .$$

Our Langevin equation for P_e is now

$$\frac{dP_e}{dt}=eFN_e-\frac{1}{mN_e}\int_0^t dt'\beta_e(t-t')P_e(t-t')(\dot{P}_{e,L};\dot{P}_{e,L}(-t'))^t+[1-\Pi(0)]G(0,t)\dot{P}_e + \int_0^t dt'[P_e(t-t')-N_e m V_D(t-t')][I_R(t-t',t)+\psi(t-t',t)] . \tag{47}$$

The first terms in (47) are the expected ones in view of Sec. V. The third term is a Mori-type fluctuating force, and the fourth term comes from the irreversibility and the nonstationary part of the dynamics. This last random force term still bears some memory of the past states of the system. From (47), we can show that the second fluctuation-dissipation theorem holds for the kernel of the dissipation term, at least up to second order in the electron-phonon interaction $H_{e\text{-}L}$, which appears to be the correlation function of the random force.

We would now like to examine the situation for the first fluctuation-dissipation theorem in the stationary state. The general form of (47) is then

$$\frac{dP_e(t)}{dt}=F-\int_0^t dt'K(t-t')P_e(t')+R(t) . \tag{48}$$

Since we consider the stationary situation, the time $t=0$ has to be such that $\rho(t=0)=\rho_L$ (steady state). In (48), $R(t)$ is the random force.

If we add to F a small field $\delta F(t)$, the resulting $\langle \delta P(t)\rangle$ is given by

$$\frac{d\langle \delta P(t)\rangle}{dt}=\delta F(t)-\int_0^t dt'K(t-t')\langle \delta P(t')\rangle , \tag{49}$$

where the effect of $\delta F(t)$ on $K(t-t')$ has been neglected (this may be a questionable assumption for far-from-equilibrium systems,[16] but adding this effect would cause large deviations). The resulting differential mobility is given by

$$\mu(\omega)=\frac{1}{i\omega+K(\omega)} . \tag{50}$$

The aim of the first fluctuation-dissipation theorem is to obtain $\mu(\omega)$ from the correlation function of P_e.

We now define a correlation function $C(P_e;P_e(\tau))$, such that $C(P_e;P_e(\tau))=0$ if $\tau<0$. From (48), we can write

$$\frac{d}{d\tau}C(P_e;P_e(\tau)) =\delta(\tau)C(P_e;P_e)-\int_0^\tau K(\tau-\tau')C(P_e;P_e(\tau')) +C(P_e;R(\tau)) . \tag{51}$$

The second term in (51) has this simple form essentially because the onset of the correlation function corresponds to the initial time in (48). If $C(P_e;R(\tau))=0$, we can obtain the quantity

$$\frac{1}{i\omega+K(\omega)}$$

from (51), and hence the first fluctuation-dissipation theorem becomes

$$\mu(\omega)=\frac{C(P_e;P_e(\tau))(\omega)}{C(P_e;P_e)} . \tag{52}$$

In the framework of the approximations of this section, the critical point in deriving such a fluctuation-dissipation theorem from (48) is in being able to find a correlation function which satisfies

$$C(P_e;R(\tau))=0 .$$

By considering the $R(\tau)$ that arises from (47), it appears that it will be extremely difficult to cancel the two components of the random force at the same time. In fact, the trivial definition

$$C(P_e;R(\tau))=\Theta(\tau)(P_e;R(\tau)) ,$$

where $\Theta(\tau)$ is the Heavyside step function, succeeds in canceling the Mori-type fluctuating force but does not give a zero result for the fluctuating force coming from the irreversible features of the steady state. In fact, our initial state evolves toward the steady state even though the thermodynamic parameters stay constant and this irreversible evolution creates the second fluctuating force.

Nevertheless, to overcome this difficulty we can try a correlation function such as $C(P_e(T);P_e(T+\tau))$, with $\tau>0$, and where T is such that the steady state is well established. This would correspond to the fluctuation dissipation in its usual form. However, this definition creates a further difficulty at the level of the convolution integral of (51) which does not retain its simple form. Furthermore, there remains little hope to obtain a zero correlation between $P_e(T)$ and the two components of the random force.

It appears, therefore, that because of the breaking of the time-reversal symmetry, the correlation between P_e and the random force is always different from zero and it is therefore not possible to obtain a fluctuation-dissipation theorem in the usual form for systems which are in a far-from-equilibrium, stationary situation. The failure of the usual form of the fluctuation-dissipation theorem was already recognized by Price[3] and by Procaccia *et al.*[2] However, our method of approaching the Langevin equation gives new insight into this physical phenomenon.

VII. CONCLUSIONS

In this paper we have derived a Langevin equation for the relevant variables of an electronic system that is coupled to a phonon bath in a semiconductor for the case of a high electric field. The presence of this high electric field case leads to a far-from-equilibrium situation. By limiting our investigations to the Langevin equation for the momentum in the system, we have shown that it contains three sources of random forces, two of which have memory of the past due to irreversibility and nonstationary behavior. In fact, it is the presence of these terms which causes a failure in the existence of the first fluctuation-dissipation theorem.

ACKNOWLEDGMENT

This work was supported in part by the U.S. Office of Naval Research.

APPENDIX A: PROPERTIES OF $\Pi(t)$

The definition of $\Pi(t)$ is as follows:

$$\Pi(t)X = \mathrm{Tr}[\rho_L(t)X] + \sum_{j,k} (X;P_j)^t [(P;P)^t]^{-1}_{jk} \times (P_k - \langle P_k \rangle^2) .$$

Properties of $\Pi(t)$:

$$\Pi(t)X = \mathrm{Tr}[\rho_L(t)X] + \sum_k \mathrm{Tr}\left[\frac{\partial \rho_L(t)}{\partial \langle P_k \rangle} X\right] \times (P_k - \langle P_k \rangle^t) , \tag{A1}$$

$$\dot{\Pi}(t)X = \sum_{j,k} \mathrm{Tr}\left[\frac{\partial^2 \rho_L(t)}{\partial \langle P_k \rangle \partial \langle P_j \rangle} X\right] \langle \dot{P}_j \rangle (P_k - \langle P_k \rangle^t) , \tag{A2}$$

$$\Pi(t)\Pi(t') = \Pi(t') , \tag{A3a}$$

$$[1-\Pi(t)][1-\Pi(t')] = [1-\Pi(t)] , \tag{A3b}$$

$$\mathrm{Tr}[\rho(t)\Pi(t)X] = \mathrm{Tr}[\rho_L(t)X] , \tag{A4a}$$

$$\mathrm{Tr}[\rho_L(t)\Pi(t)X] = \mathrm{Tr}[\rho_L(t)X] , \tag{A4b}$$

$$\mathrm{Tr}\{\rho_L(t)[1-\Pi(t)]X\} = 0 , \tag{A4c}$$

$$(X;\Pi(t)Y)^t = (\Pi(t)X;Y)^t . \tag{A5}$$

APPENDIX B

We want to show that the equality

$$G(s,t)\dot{P}_{n,L} = \dot{P}_{n,L}(t-s) \tag{B1}$$

is valid to first order in $H_{e\text{-}L}$, and the higher orders are insignificant in the present context. From the definition (22), we can write

$$G(s,t)\dot{P}_{n,L} = \left[1 + \sum_{m=1}^{\infty} \int \cdots \int L[1-\Pi(t_1)] \cdots \times L[1-\Pi(t_m)] dt_1 \cdots dt_m \right] \dot{P}_{n,L} , \tag{B2}$$

where $t_1 \cdots t_m$ are time ordered so that $t_1 < t_2 < \cdots < t_m$. If we want to evaluate $G(s,t)\dot{P}_{n,L}$ to first order in $H_{e\text{-}L}$, we need to replace L by L_0 in (B2). We notice that

$$[1-\Pi(t_m)]\dot{P}_{n,L} = \dot{P}_{n,L} ,$$

thanks to (38). We have now to study the terms $[1-\Pi(t_{m-1})]L_0\dot{P}_{n,L}$ in order to evaluate $G(s,t)\dot{P}_{n,L}$ from (B2). It appears that $\Pi(t)L_0\dot{P}_{n,L}$ is of second order in $H_{e\text{-}L}$ because $[\rho_L, H_0]$ is of first order in $H_{e\text{-}L}$ and also $(\dot{P}_{n,L};L_0P_m)$ can be shown to be zero, by (30) and (9). Therefore

$$[1-\Pi(t_{m-1})]L_0\dot{P}_{n,L} = L_0\dot{P}_{n,L} . \tag{B3}$$

This proof can be extended to all orders in L_0 so that

$$[1-\Pi(t_j)]L_0^k\dot{P}_{n,L} = L_0^k\dot{P}_{n,L} \tag{B4}$$

to first order in $H_{e\text{-}L}$. Consequently, up to this same order, it is trivial to write

$$G(s,t)\dot{P}_{n,L} = e^{iL_0(t-s)}\dot{P}_{n,L} , \tag{B5}$$

from which (B1) is a natural extension if only the first order in $H_{e\text{-}L}$ is considered.

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