

Nature of Coupled-Mode Contributions to Hot-Electron Relaxation in Semiconductors

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(Received 27 April 1990)

The energy flow in a system of hot electrons, hot LO phonons, and a bath of acoustic phonons is studied using nonequilibrium Green functions. In lowest order, Kogan's formula for the energy-loss rate is obtained. When coupled modes are included we find, in sharp contrast to Das Sarma and co-workers, that the energy-loss rate is significantly *suppressed* by coupled-mode formation and quasiparticlelike modes *do not* contribute to the energy-loss rate.

PACS numbers: 72.10.Di, 63.20.Ls, 71.38.+i, 72.20.Dp

Much experimental^{1,2} and theoretical³⁻⁵ effort has been directed at the important problem of hot-electron relaxation in semiconductors. The theory involves a difficult combination of many-body physics and non-equilibrium (NE) statistical mechanics. The possible role of coupled plasmon-phonon modes in the energy-loss rate (ELR) has to be addressed since light-scattering experiments reveal such coupled modes. Das Sarma and co-workers⁵ were the first to consider this question. They argued that the electron-hole-like phonons defining the modes of the coupled system (electrons plus optical phonons) are responsible for an "orders-of-magnitude" increase of the hot-electron ELR at low electron temperatures. The Fermi "golden rule" for the ELR by one-phonon processes can be written as a product of the spectral densities for the electron and phonon subsystems, and the Fröhlich matrix element M_q^2 . The spectral densities can be expressed as the imaginary parts of the electron-density response function $\chi(\mathbf{q}, \omega)$ and the phonon Green function $D(\mathbf{q}, \omega)$. In polar semiconductors the electron density fluctuations ρ_q and LO-phonon modes C_q become coupled modes ξ_q and the separate identities of the two systems with different excitations become lost. In Ref. 5 a coupled-mode (CM) form for ELR is proposed by replacing the uncoupled phonon spectral density by that of the coupled system while keeping the rest of the golden-rule formula essentially intact. The resulting formula gives an ELR enhanced by orders of magnitude over the Kogan formula, especially at low electron temperatures.

It is not evident that the Fermi golden rule remains valid when coupled-mode effects are introduced, and hence a more complete NE approach is necessary. The *objective* of this Letter is to show that (a) the quasiparticlelike CM phonons *do not* contribute to ELR as they are effectively at the electron temperature and hence the orders-of-magnitude enhancement does not occur; (b) the remaining CM phonons are effectively "hotter" than bare phonons and hence CM formation *suppresses* ELR, and may, in fact, be a source of the "hot-phonon" effect. The physical picture associated with the analysis given in this Letter can be clarified by noting that (i) the traditional Kogan formula predicts an ELR assuming that

electrons and phonons are independent systems with well-defined temperatures; (ii) Das Sarma and co-workers introduce the coupled-mode spectral densities but continue to use the unmodified distribution functions (Bose factors) and obtain an orders-of-magnitude enhancement; and (iii) we use the coupled-mode spectral densities *as well as* the coupled-mode distribution functions and find no enhancement of the sort obtained by Das Sarma and co-workers.

The NE problem can be tackled using either Zubarev's approach,⁶ or using a diagram method⁷ for Schwinger's closed-time-path Green functions conveniently evaluated on a Keldysh contour. In both methods the system at time $t = -\infty$ is in equilibrium. The NE system is constructed by building up the NE energy fluxes at time t , say $t=0$. In either method the ELR is expressed in terms of a Green function which has to be identified. Initially we follow Zubarev's approach. The lowest-order result leads to the Kogan formula⁸ usually derived from the Fermi golden rule. The next level of approximation is delicate, since (a) when coupled modes are formed the detailed CM-phonon distribution function is needed and (b) the modes are already damped by the ELR process which depends on NE conditions. Hence the random-phase approximation (RPA) should ideally use propagators containing self-energy insertions.

The hot electrons lose energy mainly by emitting longitudinal-optical (LO) phonons which decay into acoustic (Ac) phonons via anharmonic interactions. The Ac phonons are at the temperature T of the heat bath. Within a time scale τ_{ee} much shorter than a typical LO-phonon emission time τ_{ep} , the electrons reach a quasi-equilibrium (QE) temperature T_e via electron-electron interactions H_{ee} . The time scale τ_{pp} for LO-phonon interactions is much longer than τ_{ee}, τ_{ep} , and the LO phonons do not equilibrate to an effective temperature T_p . However, as in Ref. 5 we assume a T_p and consider only single-phonon processes. The formal analysis given below is aimed at deriving Eq. (3) for the energy flux and some readers may prefer to go directly to Eq. (3). From Eq. (3) we obtain Eq. (5) which is the standard Kogan formula, while Eqs. (6) and (8) are our result inclusive

of coupled-mode effects.

The assumption of quasiconserved quantities β_e , β_p , and β leads to a simplified Zubarev NE density operator $\rho(t)$. It only involves the Hamiltonians H_e and H_p and the energy-flux operators \dot{H}_e, \dot{H}_p . These are given by $\dot{H}_e = -i[H_e, H]$ and $\dot{H}_p = -i[H_p, H]$. The total Hamiltonian H contains H_e, H_p, H_a , and the interactions H_{ep} and H_{ap} , i.e., electron-LO and Ac-LO, respectively. H_{ee} is included in H_e . We neglect the e -Ac coupling and treat the Ac-LO processes in a simplified manner. The simplified NE density operator at time $t=0$ can be written⁶ as $\rho = \rho_{NE} = Q^{-1} \exp[-A - B]$, where Q is the normalization and A is $\beta_e H_e + \beta_p H_p + \beta(H - H_e - H_p)$. Also B contains the fluxes. Thus

$$B = -\Delta\beta_e \int_{-\infty}^0 dt e^{\epsilon t} \dot{H}_e - \Delta\beta_p \int_{-\infty}^0 dt e^{\epsilon t} \dot{H}_p.$$

Here we have defined $\Delta\beta_e = \beta_e - \beta$ and $\Delta\beta_p = \beta_p - \beta$. For time scales $\tau < \tau_{ep}$ the whole system is in quasi-equilibrium, with the density operator given by $\rho_{QE} = Q_{QE}^{-1} \exp(-A)$. Since ρ_{QE} is stationary for time scales $\tau < \tau_{ep}$, it is not affected by small perturbations (to linear order) and, hence, how H_i , i.e., $H_{ep} + H_{ap}$, is treated in ρ_{QE} is not important. The NE operator ρ_{NE} may be pictured as built up from ρ_{QE} by switching on the energy fluxes using a positive infinitesimal ϵ (cf. scattering theory).⁶ The energy flux is now given in terms of diagonal and nondiagonal fluxes as

$$\langle \dot{H}_e \rangle = \Delta\beta_e J_{ee} + \Delta\beta_p (J_{e,ep} + J_{e,ap}), \quad (1)$$

$$J_{ee} = \left\langle \int_{-\infty}^0 dt e^{\epsilon t} \int_0^1 d\tau \dot{H}_e \dot{H}_e(t\tau) \right\rangle_{QE}.$$

Similarly $J_{e,ep}$ contains $\langle \dots \dot{H}_e K_{ep}(t\tau) \rangle_{QE}$, where $K_{ep} \equiv -i[H_p, H_{ep}]$, and $J_{e,ap}$ contains $K_{ap} \equiv -i[H_p, H_{ap}]$. In deriving (1) we used the property that $\langle \dot{H}_e \rangle_{QE}$ and $\langle \dot{H}_p \rangle_{QE}$ are zero, and simplified $\rho_{NE} = Q^{-1} \exp[-A - B]$ since the effect of B containing the fluxes is small compared to that of A . Similar expressions hold for the phonon energy flux $\langle \dot{H}_p \rangle$.

The electron-LO interaction H_{ep} is $\sum M_q \langle n | \rho_q | m \rangle \times C_q a_m^\dagger a_n$, where the LO-phonon operator C_q is $b_q + b_{-q}^\dagger$, and m, n are electron states. The summations run over all indices. For plane-wave states H_{ep} becomes $\sum M_q (b_q + b_{-q}^\dagger) a_{k+q}^\dagger a_k$ or $\sum M_q C_q \rho(\mathbf{k}, \mathbf{q})$, where $\rho(\mathbf{k}, \mathbf{q})$ or $\rho(n, m)$ is the electron-hole (e - h) pair operator. The explicit form of the matrix elements $M_q \langle n | \rho_q | m \rangle$ is found in the literature.⁹ The electron Hamiltonian H_e is $H_e^0 + H_{ee}$ and H_e^0 is $\sum \epsilon_k a_k^\dagger a_k$. The e - e interaction is given by $\frac{1}{2} \sum V_q a_k^\dagger a_{k+q}^\dagger a_k a_{k+q}$, $\mathbf{q} \neq 0$, where V_q is the bare Coulomb interaction scaled by the background static dielectric constant. The LO-phonon Hamiltonian H_p is $\sum \omega_q (b_q^\dagger b_q + \frac{1}{2})$. From Eq. (1), the rate of change of the electron temperature is written as

$$\begin{aligned} d\beta_e/dt &= \langle \dot{H}_e \rangle [d\langle H_e \rangle / d\beta_e]^{-1} \\ &= F(J_{ee}, J_{e,ep}) + \Delta\beta_p / \tau_{e,ap}. \end{aligned} \quad (2)$$

Here only the LO-Ac term is treated via the relaxation time $\tau_{e,ap}$. This $\tau_{e,ap}$ relates to a nondiagonal Onsager kinetic coefficient if the QE average in $J_{e,ap}$ were replaced by an equilibrium average. The term $F(J_{ee}, J_{e,ep})$ contains the dependence on β_e, β_p, β and $J_{ee}, J_{e,ep}$ as implied by Eq. (1).

Using the explicit forms for H_{ep} , etc., and using m, n to stand for $\mathbf{k} + \mathbf{q}$ and \mathbf{k} (also ϵ_{mn} for $\epsilon_m - \epsilon_n$), the fluxes can be written in terms of quasiequilibrium Green functions $G(-t)$ defined as $G(t-t') = \langle \langle A, B \rangle \rangle$ which is $-i\theta(t-t') \langle \langle [A(t), B(t')] \rangle \rangle_{QE}$. The time integration in (1) picks up the $\omega + i\epsilon$ component at $\omega = 0$.

Finally, the energy flux can be written as

$$\langle \dot{H}_e \rangle = \sum M_q M_q \epsilon_{mn} (1/i) \{ \mathbb{G} \}_{\omega=0+i\epsilon}, \quad (3)$$

where the retarded quasiequilibrium Green function \mathbb{G} is given by $\langle \langle C_q^\dagger a_m^\dagger a_n, C_q a_m^\dagger a_n \rangle \rangle$. Here $C_q = b_q + b_{-q}^\dagger$ is the phonon operator and $a_m^\dagger a_n$ defines an electron-density fluctuation with energy ϵ_{mn} . The electron-phonon matrix element (for plane-wave states) is M_q . The same Green function arises for the phonon flux \dot{H}_p . Also, only the imaginary part of \mathbb{G} contributes to (3). In evaluating \mathbb{G} the diagram method⁷ can be used where time-ordered, anti-time-ordered, and distribution Green functions related to \mathbb{G} are needed. Or one may use the equations-of-motion method where Wick averages have to be explicitly worked out. However, a full NE treatment will not be very transparent.

Instead, let us consider the simpler *model problem* of evaluating \mathbb{G} using the Matsubara method for an *equilibrium* system at some temperature $1/\beta'$. Then we need to evaluate the frequency sum

$$-(1/\beta') \sum_{i\omega_n, k_n} D^0(i\omega_n, \mathbf{q}) g^0(ik_n + i\omega_n, \mathbf{k} + \mathbf{q}) g^0(ik_n, \mathbf{k}), \quad (4)$$

where ik_n and $i\omega_n$ are Fermi and Bose Matsubara frequencies, and $g^0(ik_n, \mathbf{k})$ is a zeroth-order electron propagator [Fig. 1(a), dashed line is D^0]. Defining $\Delta n_{\mathbf{k}\mathbf{q}} = n_{\mathbf{k}} - n_{\mathbf{k}+\mathbf{q}}$, where $n_{\mathbf{k}}$ is a Fermi occupation number, and $\epsilon_{\mathbf{k}\mathbf{q}} = \epsilon_{\mathbf{k}+\mathbf{q}} - \epsilon_{\mathbf{k}}$, $D^\pm = \pm \omega_{-q} + \epsilon_{\mathbf{k}\mathbf{q}}$, and summing over $ik_n, i\omega_n$ we get $\Delta n_{\mathbf{k}\mathbf{q}} \{D^+ - D^-\}$ multiplied by $\Delta N = \bar{N}(\beta', \omega_q) - \bar{N}(\beta', \epsilon_{\mathbf{k}\mathbf{q}})$ containing Bose factors at the phonon energy and at the e - h excitation energy. In the Matsubara method both factors contain β' , but when \mathbb{G} is evaluated for the NE system using, say, the equations-of-motion method in lowest order they come out with β_p and β_e , respectively. When the imaginary part of \mathbb{G} is taken in Eq. (4) the \mathbf{k} summation can be carried out to yield the Kogan formula:⁸

$$\begin{aligned} \text{ELR} &= \sum_q \int (\omega/\pi) \omega M_q^2 \\ &\quad \times \{ -\text{Im} \chi_e(\mathbf{q}, \omega_q) \} \text{Im} D^0(\mathbf{q}, \omega) \Delta \bar{N}^0, \quad (5) \\ \Delta \bar{N}^0 &= \bar{N}(\beta_e, \omega) - \bar{N}(\beta_p, \omega). \end{aligned}$$

To study corrections to Kogan's formula we need to evaluate (5) with improved $D(i\omega_n, \mathbf{q})$ and $g(ik_n, \mathbf{k})$. If we introduce a spectral representation for these we obtain, for the *NE problem*,

$$\text{ELR} = \sum_q \int (d\omega/\pi) \omega M_q^2 \times \{-\text{Im}\chi_e(\mathbf{q}, \omega_q)\} \text{Im}D(\mathbf{q}, \omega) \Delta\bar{N}_{\text{CM}}, \quad (6)$$

$$\Delta\bar{N}_{\text{CM}} = \bar{N}(\beta_e, \omega_q) - \bar{N}_{\text{CM}}(\beta_p, \beta_e, \omega),$$

where the coupled-mode distribution \bar{N}_{CM} associated with the renormalized $D(\mathbf{q}, \omega)$ [Fig. 1(b), short-dashed line] occurs. $\chi(\mathbf{q}, \omega)$ is the electron response, preferably with self-energy insertions. If $\Delta\bar{N}_{\text{CM}}$ is replaced by $\Delta\bar{N}^0$, and $\chi(\mathbf{q}, \omega)$ by its RPA form we recover the result of Ref. 5. However, $\Delta\bar{N}_{\text{CM}}$ contains the CM-phonon distribution $\bar{N}_{\text{CM}}(\beta_p, \beta_e, \omega_q)$. To appreciate the structure of \bar{N}_{CM} we note that for an *equilibrium* system the dynamic structure factor (DSF), i.e., $S(\mathbf{q}, \omega)$, is related to its distribution function $\bar{N}(\beta, \omega)$ by $\bar{N}(\beta, \omega) = -\pi S(\mathbf{q}, \omega)/D_2(\mathbf{q}, \omega)$, where D_2 is $\text{Im}D(\mathbf{q}, \omega)$. The coupled-mode DSF is a linear combination of the separate phonon and electron DSFs which are proportional to D_2^0 and χ_2 . For an equilibrium CM system at $\beta = \beta_e = \beta_p$, we have $\bar{N}_{\text{CM}} = \bar{N}(\beta, \omega)$. That is, trivially,

$$\bar{N}_{\text{CM}}(\beta, \beta, \omega) = [D_2^0(\mathbf{q}, \omega)\bar{N}(\beta, \omega) + M_q^2 |D^0(\mathbf{q}, \omega)|^2 \chi_2(\mathbf{q}, \omega)\bar{N}(\beta, \omega)]/D_2(\mathbf{q}, \omega), \quad (7)$$

$$D_2(\mathbf{q}, \omega) = D_2^0(\mathbf{q}, \omega) + M_q^2 |D^0(\mathbf{q}, \omega)|^2 \chi_2(\mathbf{q}, \omega).$$

D_2^0 and χ_2 are the imaginary parts of propagators D^0 and χ , while D_2 is the numerator of $\text{Im}D(\mathbf{q}, \omega)$. The CM-phonon distribution function for the *NE system* is given by

$$\bar{N}_{\text{CM}}(\beta_p, \beta_e, \omega) = [D_2^0(\mathbf{q}, \omega)\bar{N}(\beta_p, \omega) + M_q^2 |D^0(\mathbf{q}, \omega)|^2 \chi_2(\mathbf{q}, \omega)\bar{N}(\beta_e, \omega)]/D_2(\mathbf{q}, \omega) \quad (8)$$

and is the obvious generalization of (7). In Fig. 2, top panel, we show $\bar{N}_{\text{CM}}(\beta_p, \beta_e, \omega)$ for electrons at 50 K, the lattice at 4 K, and for a wave vector $q = 0.22 \times 10^6 \text{ cm}^{-1}$, as a function of ω . These parameters correspond to those of Ref. 5. It is clear that in the quasiparticlelike region ($\omega < 0.3\omega_{\text{LO}}$) the CM distribution \bar{N}_{CM} is almost exactly $\bar{N}(\beta_e, \omega_q)$ and hence $\Delta\bar{N}_{\text{CM}}$ occurring in the ELR formula is zero, and hence there is no contribution to energy loss from the QP-like CM phonons. If $\Delta\bar{N}^0$ were used instead of $\Delta\bar{N}_{\text{CM}}$ as in Ref. 5, one would incorrectly conclude that an orders-of-magnitude increase in ELR would result from the QP-like phonons. The lower panel of Fig. 2 shows the CM-phonon spectral function at two densities. In Fig. 3 we show the ELR spectrum for three electron temperatures, where the solid lines correspond to the theory of Ref. 5 [cf. Fig. 6 of Ref. 5(a)], using $\Delta\bar{N}^0$, while the dashed line is from the present theory, using $\Delta\bar{N}_{\text{CM}}$. The 4 orders-of-magnitude difference in the two calculations arises mainly from the $\bar{N}(\beta_p, \omega)$ factor contained in $\Delta\bar{N}^0$ which is dominant, since the lattice temperature is 4 K for all three cases considered.

The underlying physical picture is that although the coupled-mode phonon spectral density has some weight in the quasiparticlelike (i.e., electron-hole-like) region of energies, these coupled-phonon modes are effectively at the electron temperature and not, as assumed in Ref. 5, at the lattice temperature. Hence there is no enhance-

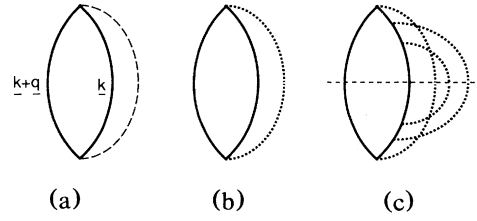


FIG. 1. (a) Structure of G , see Eqs. (2) and (3); (b) with renormalized phonon propagator; (c) with a self-energy insertion having three phonons propagating with an electron.

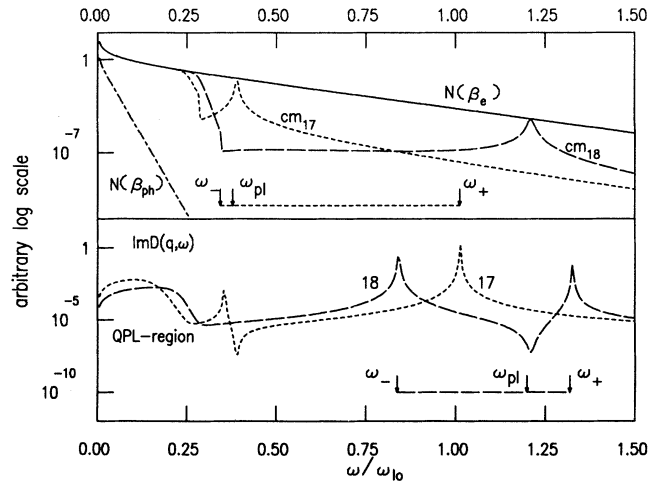


FIG. 2. Top panel: $N(\beta_e)$ and $N(\beta_p)$ are "bare" electron and phonon (Bose) distributions, with $T_e = 50 \text{ K}$ and $T_p = 4 \text{ K}$. The coupled-mode phonon distribution N_{CM} for wave vector $q = 0.22 \text{ nm}^{-1}$ and electron density $n_e = 10^{17}$ and 10^{18} electrons/cm³ (marked cm_{17} and cm_{18}). Positions of coupled modes ω_{\pm} and the plasma frequency ω_{pl} are shown in the arrowed sketch. Bottom panel: Coupled-mode phonon spectral functions at the two densities, marked 17 and 18. QPL denotes quasiparticlelike.

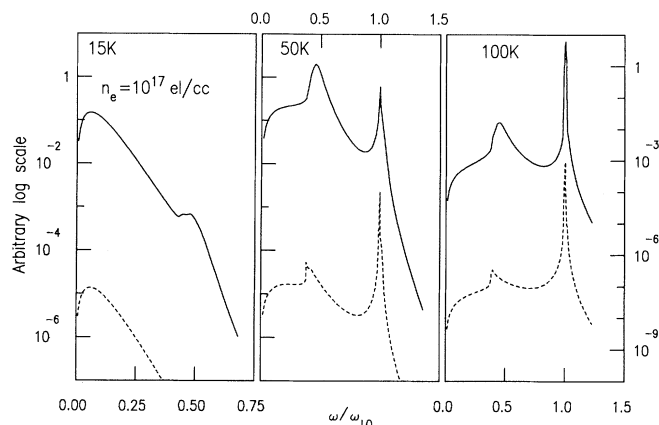


FIG. 3. The ELR spectrum calculated with $\Delta\bar{N}^0$ (solid line) and with $\Delta\bar{N}_{CM}$ (dashed line). Y axis has arbitrary scale factors. The solid line is similar to Fig. 6 of Ref. 5(a).

ment. The results of Fig. 2 are for $T=50$ K, while the LO-phonon mechanism is most appropriate for higher temperatures. Nevertheless, as seen from Fig. 2 CM phonons are hotter than bare phonons and this leads to a *suppression of ELR*. This should be considered in all discussions or simulations of the hot-phonon effect.

For ω comparable to QP energies the CM time scale $\tau_{CM} \sim 1/\omega$ is $\gg \tau_{ep} \sim 1/\omega_{LO}$. This slow CM process has to compete with fast ($\sim \tau_{ep}$) ELR-type self-energy processes where the electron loses energy via a series of phonon emissions before reabsorption of a phonon. One such process is shown in Fig. 1(c) where three hot phonons propagate with the electron at the time instant corresponding to the horizontal dashed line. Many diagrams which are zero for equilibrium become nonzero in the NE picture, and the propagators should include self-energies prior to RPA summation. A consistent calculation is a formidable task.¹⁰ A crude evaluation shows that the contribution of *e-h*-like phonons to the phonon spectral density is approximately halved when these effects are included for, say, $T_e=50$ K and $n \sim 10^{17}$ electrons/cm³.

In conclusion, we have studied the ELR for hot-electron cooling by emission of LO phonons using a NE many-body approach. The Green functions in the energy current are identified. Their lowest-order evaluation leads to the Kogan formula. A higher-order evaluation

requires the CM *distribution functions*, the renormalized propagators, screening functions, etc. The CM-phonon distribution function shows that the low-energy tails are effectively at the electron temperature and hence QP-like phonons *do not* contribute to ELR. The higher-energy CM phonons are hotter than "bare" phonons and contribute to a *decrease* in ELR and may explain some of the effects currently attributed to hot phonons.

We are grateful to Geof Aers, H.-Y. Chu, and Pawel Hawrylak for their comments on the manuscript and for many useful discussions. We thank Jeff Young for many valuable discussions and for bringing this problem to our attention in the first place.

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