Nonstationary Quasiperiodic Energy Distribution of an Electron Gas upon Ultrafast Thermal Excitation

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We consider the evolution of an electronic system initially in equilibrium with a semiconductor crystal lattice upon a sudden rise in the lattice temperature. On a short time scale (~ 1 ps), electrons equilibrate with the optical phonons and establish a peculiar quasiperiodic energy distribution. Time evolution toward this distribution is discussed on the basis of an exact solution of the Boltzmann transport equation. While the quasiequilibrium regime persists (longer than ~ 1 ns), the electronic system possesses a number of unusual thermodynamic and transport properties. The specific heat is suppressed as if some of the electron degrees of freedom were frozen out; the low-field mobility is substantially enhanced.

Thermodynamic equilibrium between an electronic system and a crystal lattice is established by phonon scattering. At sufficiently high temperatures, the electron energy relaxation rate due to optical phonons ($\tau^{(op)}$) is higher than that due to acoustic phonons ($\tau^{(ac)}$) by several orders of magnitude. Typically, in semiconductors $\tau^{(op)} \lesssim 10^{-12}$ s and $\tau^{(ac)} \gtrsim 10^{-9}$ s.[1] This disparity of the inelastic relaxation times can lead to the formation of an electronic ensemble that is in equilibrium with the optical-phonon system but has not yet appreciably interacted with acoustic phonons. As will be shown below, such an ensemble possesses rather unusual properties. Manifestation of these properties in electronic transport can be conveniently referred to as the "classical mesoscopic effects" (drawing a parallel to and a distinction from the quantum mesoscopic effects that occur when the coherence length or time of an electronic wave function exceeds characteristic system dimensions[2]). In the present Letter, we discuss effects arising from abrupt changes in the lattice temperature. It will be shown that the electron energy distribution acquires a peculiar structure that affects a number of macroscopic properties.

Consider a semiconductor initially in equilibrium at a temperature T_0. Suppose the lattice temperature is suddenly raised to a value $T > T_0$. Let us discuss the evolution of the electron distribution over a relatively short time, when the only important source of inelastic scattering is due to optical phonons. For concreteness, we shall be considering the case of GaAs and hence the interaction with polar optic phonons.[3] In this case the Boltzmann kinetic equation can be written in the form

$$\tau_0 \frac{\partial f(\tilde{E})}{\partial t} = \frac{2\operatorname{cosech}(\beta)}{(\tilde{E})^{1/2}}(1 - e^{\partial/\partial\tilde{E}})[e^{-\beta}f(\tilde{E}-1) - e^{\beta}f(\tilde{E})]\ln[(\tilde{E}-1)^{1/2} + (\tilde{E})^{1/2}], \tag{1}$$

where $f(\tilde{E})$ is the electron distribution function, $\tilde{E} \equiv E/\hbar\omega_0$ is a dimensionless electron energy in units of optical-phonon energy $\hbar\omega_0$, and $\beta \equiv \hbar\omega_0/2kT$; the operator $e^{\partial/\partial\tilde{E}}\chi(\tilde{E}) \equiv \chi(\tilde{E}+1)$ effects displacements by unity. The characteristic time τ_0 is given by

$$\frac{1}{\tau_0} = \frac{e^2\sqrt{2m\hbar\omega_0}}{4\hbar^2}\left[\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_0}\right] \approx 2\times10^{12}\,\text{s}^{-1}, \tag{2}$$

where e is the electron charge, m the effective mass, and ε_∞ and ε_0 are, respectively, the optical and the static dielectric permittivities. Let us divide all electrons into energy ladders

$$\{E^{(\nu)}(\varepsilon)\} = (\varepsilon+\nu)\hbar\omega_0, \quad 0 \le \varepsilon \le 1, \quad \nu=0,1,2,\dots. \tag{3}$$

Here ε is the dimensionless electron energy *modulo* the optical-phonon energy $\hbar\omega_0$; for $E \le \hbar\omega_0$ both $\tilde{E}$ and ε have the same meaning. The total number of electrons on a given ladder will be denoted by $N_{\text{ext}}(\varepsilon)$; this number is a functional on the distribution $f(E)$.

It is easy to see that the kinetic equation (1) does not couple populations belonging to different energy ladders (3). Therefore, Eq. (1) can be split into a set of independent equations,

$$\frac{\partial \mathbf{f}(\varepsilon)}{\partial t} = \mathbf{M}\cdot\mathbf{f}(\varepsilon), \tag{4}$$

where we have introduced a multidimensional vector $\mathbf{f}(\varepsilon) \equiv \{f(\varepsilon), f(\varepsilon+1),\dots\}$. Equation (4) can be efficiently solved by numerically diagonalizing the matrix $\mathbf{M}$. It can be shown that one of the eigenvalues of $\mathbf{M}$ equals zero (this is required for the existence of a stationary solution) and all the other eigenvalues are real and negative. The initial condition for $\mathbf{f}$ corresponds to an equilibrium distribution at T_0.

Figure 1(a) shows a sequence of electron energy distributions [$\propto (\tilde{E})^{1/2}f(\tilde{E})$, normalized to unity] at different times, calculated for an electronic system initially at $T_0 = 77$ K interacting with the lattice at $T = 300$ K. We
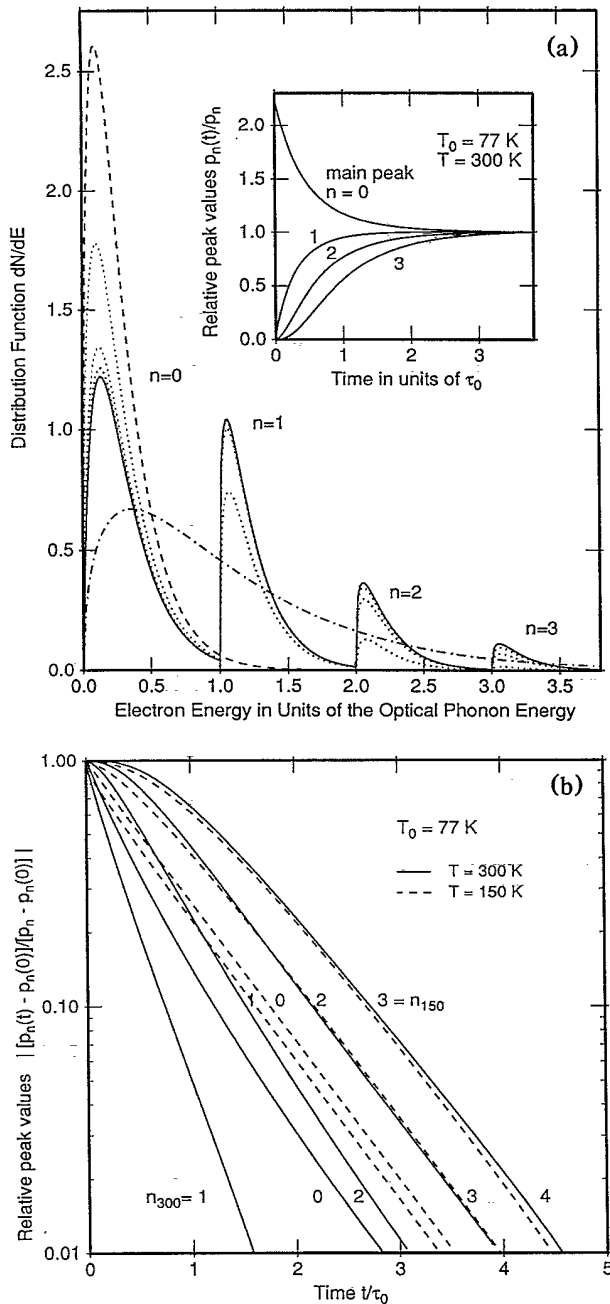
FIG. 1. Time evolution of the electron energy distribution, as electrons, initially at $T_0 = 77$ K are heated by phonons that are in equilibrium at $T = 300$ K. (a) Dotted curves show the normalized (to unity) distribution $\sqrt{E}f(t,E)$ at times $t = 0.4\tau_0$, $1.2\tau_0$, and $2.0\tau_0$. The initial distribution, corresponding to equilibrium at $T_0 = 77$ K, is shown by the dashed line. Solid curves describe the quasiequilibrium "mesoscopic" distribution $\sqrt{E}\tilde{f}(T_0 \rightarrow T,E)$, calculated from Eq. (8). The stipple curve corresponds to the equilibrium distribution $f(T,E)$ that will be established after a much longer time $t \sim \tau^{(ac)}$. Inset: Time dependence of the peak values $p_n(t)$ at $n = 0,1,2,3$ relative to their values p_n in quasiequilibrium. (b) Relative peak values, $|[p_n(t) - p_n(0)]/[p_n - p_n(0)]|$, plotted on a logarithmic scale. Dashed lines correspond to the phonon temperature $T = 150$ K.

see that, to a good measure, the electronic system attains a quasistationary distribution already at $t \approx 2\tau_0$. It has a quasiperiodic structure with a period $\hbar\omega_0$; the ratio of neighboring peaks is approximately $e^{\hbar\omega_0/kT}$. Dynamics of the different peaks are illustrated in the inset of Fig. 1(a) and further in Fig. 1(b), where we compare the effects at two different temperatures. The equilibration processes are seen to be roughly exponential. The higher the energy, the longer the quasiequilibration time: Clearly, the equilibration of peaks with higher indices n requires a larger number of phonon absorption processes that move electrons up in the energy ladder. Since the high-energy peaks $n = 1,2,\ldots$ grow at the expense of the main peak with $n = 0$, the latter equilibrates slower than the $n = 1$ peak. The temperature dependence is due to the variation in the phonon number, $\propto e^{-\hbar\omega_0/kT}$.

The shape of the quasistationary distribution can be calculated analytically. It is clear that optical-phonon scattering cannot change the number N_{ext} of electrons on a given energy ladder,

$$N_{ext}(T_0,\varepsilon) = N_{ext}(T,\varepsilon). \qquad (5)$$

On the other hand, within a ladder, the distribution of populations on different rungs in quasiequilibrium at a temperature T is given by the Gibbs statistics. Therefore, Eq. (5) determines the quasi-Fermi-level $E_F(T,\varepsilon)$ with a ladder $E^{(\nu)}(\varepsilon)$ at quasiequilibrium:

$$\exp\left[\frac{E_F(T,\varepsilon)}{kT}\right] = \exp\left[\frac{E_F^{(0)}(T_0)}{kT_0}\right]\frac{Z(T_0,\varepsilon)}{Z(T,\varepsilon)}, \qquad (6)$$

where $Z(T,\varepsilon)$ is the partition function

$$Z(T,\varepsilon) = \sum_{\nu=0}^{\infty}(\varepsilon+\nu)^{1/2}e^{-(\nu+\varepsilon)\hbar\omega_0/kT}. \qquad (7)$$

The quasiequilibrium distribution function is thus given by

$$\tilde{f}(T_0 \rightarrow T,E) = \exp\left[\frac{E_F^{(0)}}{kT_0}\right]\exp\left[-\frac{E}{kT}\right]\frac{Z(T_0,\varepsilon)}{Z(T,\varepsilon)}. \qquad (8)$$

Note that the ratio $Z(T_0,\varepsilon)/Z(T,\varepsilon)$ in Eq. (8) is a periodic function of E, since ε is defined as a remainder in the division of E by $\hbar\omega_0$. The functions $\sqrt{E}\tilde{f}(T_0 \rightarrow T,E)$ calculated from (8) are plotted in Fig. 1(a) by solid lines. It can be seen that they are practically indistinguishable from the nonstationary electron energy distributions, calculated with the time-dependent Boltzmann equation for $t \gtrsim 2\tau_0$.

We see from Fig. 1(a) that the quasiequilibrium functions $\tilde{f}(T_0 \rightarrow T,E)$ are dramatically different from the equilibrium distribution $f(T,E)$. The latter is established by the acoustic-phonon scattering after a considerably longer time $t \sim \tau^{(ac)} \gg \tau_0$, that can be regarded as a time of the degradation of the mesoscopic order, embo-

died in Eq. (5). The rate at which the distribution $\bar{f}(T_0 \to T,E)$ degrades depends on the temperature and is different in different energy ranges. For $T = 300$ K, the degradation time $\tau^{(ac)}(\varepsilon) \gtrsim 10^{-7}$ s, at all energies.[4]

So long as the mesoscopic order persists it affects many properties of the electronic system. Consider, for example, the average energy of the electron gas

$$\langle E \rangle = \frac{1}{N} \int E^{3/2} \bar{f}(T_0 \to T,E) dE , \qquad (9)$$

where

$$N \equiv \int_0^\infty E^{1/2} \bar{f}(T_0 \to T,E) dE$$

$$= \int_0^\infty E^{1/2} f_{t=0}(E,T_0) dE . \qquad (10)$$

In equilibrium, of course, one has $\langle E \rangle = 3kT/2$. Figure 2 shows the ratio $2\langle E \rangle/3kT$ as a function of the lattice temperature for several initial electron distributions, characterized by different values of T_0. We see that the lower the T_0 the more suppressed is the mean kinetic energy in the mesoscopic regime. This effect can be viewed in analogy to the suppression of degrees of freedom at low temperatures in a quantum system with an energy gap.[5] Since the environment allows electrons to change energy only by discrete portions $\hbar\omega_0$, at low $T \ll \hbar\omega_0$ most electrons remain at the lowest rung $v = 0$ of the energy ladder $E^{(v)}(\varepsilon)$. The dashed lines in Fig. 2 show the temperature dependence of the electronic specific heat C in the mesoscopic regime. Suppression of the specific
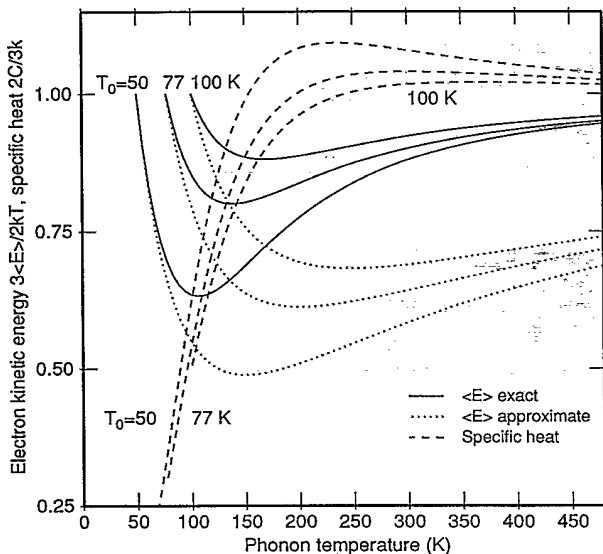

FIG. 2. Thermodynamic properties of the electron system in the mesoscopic regime. Solid lines show the mean kinetic energy of electrons distributed according to $\bar{f}(T_0 \to T,E)$ as a function of the phonon temperature T for three different initial electron temperatures T_0. Dotted lines correspond to the crude model of Eq. (11). Dashed lines represent the specific heat per electron relative to the equilibrium value $3k/2$.

heat at low T and its behavior at higher temperatures is quite analogous to the deviation of the lattice specific heat from the classical Dulong-Petit behavior in the Einstein model of a solid. The slight difference, manifested in the overshoot by C of the classical value of $3k/2$ per electron and the existence of a maximum, originates from the concrete shape of the electron density of states.

Qualitatively, the behavior of $\langle E \rangle$ can be reproduced in a simple model in which the initial electron distribution is assumed by the form $\delta(E - kT_0)$ and the density of states is assumed independent of the energy. The former assumption is reasonable, since $kT_0 \ll \hbar\omega_0$; the latter is crude, since the $(\tilde{E})^{1/2}$ factor is non-negligible. This analytical model should be more accurate for a two-dimensional electron gas. The result

$$\frac{\langle E \rangle\{\tilde{f};T\}}{\langle E \rangle\{f;T\}} = \frac{\hbar\omega_0 + T_0(e^{\hbar\omega_0/kT} - 1)}{\hbar\omega_0 + T(e^{\hbar\omega_0/kT} - 1)} \qquad (11)$$

is plotted in Fig. 2 by dotted lines.

Perhaps the simplest way of experimentally observing the mesoscopic order in the electron distribution would be to measure the electron mobility as a function of time upon a rapid increase in the lattice temperature. In a weak electric field, the mobility μ is given by the following expression:[6]

$$\mu = -\frac{2e}{3m} \frac{1}{N} \int_0^\infty E^{3/2} \tau_p(E) \frac{\partial f(E,T)}{\partial E} dE , \qquad (12)$$

where $\tau_p(E)$ is the momentum relaxation time. In the time range $2\tau_0 \lesssim \tau^{(ac)}$, the mesoscopic mobility $\bar{\mu}$ is given by Eq. (12) with f replaced by $\bar{f}(T_0 \to T,E)$.

Figure 3 shows the dependences $\bar{\mu}(T)$ for three initial temperatures T_0 of the electron gas. The curves are plotted in the range $T \geq T_0$. In the assumed expression for $\tau_p(E)$ we included three scattering mechanisms: by acoustic phonons, by polar optic phonons, and by ionized impurities of concentration $N_D = 10^{16}$ cm^{-3}. The solid curve in Fig. 3 corresponds to the equilibrium mobility calculated from Eq. (12) for the same scattering mechanism. The shape of this curve indicates that at such a low N_D the phonon scattering dominates, except at lowest temperatures; in the nonstationary mobility, the effect of impurity scattering is more pronounced. In general, we see that $\bar{\mu}(T) > \mu(T)$. This has a simple explanation in light of the data in Fig. 2: A lower kinetic energy implies less phonon scattering. Relative suppression of the mean kinetic energy is larger at lower T and the mobility enhancement is therefore stronger in that range. At still lower temperature, the impurity scattering becomes more important and the suppression of kinetic energy leads to lower mobility.

It should be clear that, in addition to the mobility, a number of other macroscopic properties will be affected by the mesoscopic order in the electron distribution. In particular, one can expect a quasiperiodic structure in thermionic emission or tunneling across heterostructure
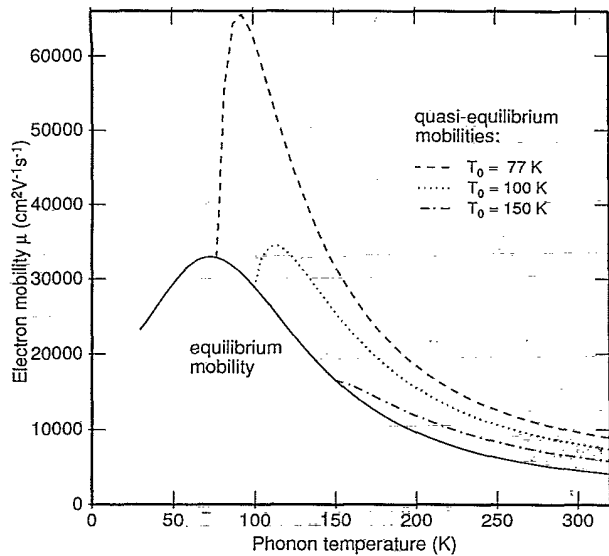
FIG. 3. Temperature dependence of the mobility of an electron gas, distributed according to $\tilde{f}(T_0 \rightarrow T, E)$, for three different initial electron temperatures T_0. The solid line shows the equilibrium mobility, calculated with the same scattering mechanisms (acoustic and optic phonons plus ionized impurities of concentration 10^{16} cm^{-3}).

barriers, in the free-electron light absorption, etc. A sudden increase of the lattice temperature in the vicinity of an electronic system can be achieved by supplying heat from a bath. In the heat bath itself, the temperature can be varied rapidly by a variety of means. Inasmuch as the heat is transferred mainly by acoustic phonons, an important factor is the time of thermal equilibration between the acoustic- and the optic-phonon fields. In GaAs, this time is approximately 7 ps,[7,8] which

is longer than the time $t \sim 2\tau_0 \approx 1$ ps required to establish the mesoscopic distribution $\tilde{f}(T, E)$. This implies that the distribution will be faithfully following the variation of the optical-phonon number, offering a way for experimentally studying kinetics of the optical-phonon generation by acoustic phonons.

Finally, it should be noted that, although we have only considered here a spatially uniform electronic system, in the absence of currents, similar effects can be expected in situations when electrons flow across regions of high-temperature gradient. These situations can be analyzed by generalizing the mesoscopic "conservation law" (5) to a continuity equation, appropriate for a nonuniform system with a current flow.[4,9] The typical length scale of classical mesoscopic effects is of the order of tens of microns which exceeds the size of most modern semiconductor devices.

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