Effect of ambipolar diffusion on the hot-carrier relaxation in semiconductors

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The effect of the change in the carrier density due to ambipolar diffusion on the cooling of hot plasmas in semiconductors is investigated. It is shown that the diffusion contributes to a warm-up of the plasma.

In experiments for generation, by photoexcitation, of electron-hole plasmas in semiconductors, electron-hole pairs are produced with an excess of kinetic energy equal to the difference between the photon and the band-gap energy. Further, for the plasma state to be produced, the carrier concentration needs to be larger than $\sim 10^{16}$ cm^{-3}, so that the system is on the metallic side of the Mott transition and can be regarded as a two-component Fermi liquid. The investigation of the relaxation of these hot carriers is of scientific relevance in the study of the fundamental process in condensed matter, as well as of technological interest for the development of very fast operational devices.

It is acknowledged that, generally, the relaxation process can be broken down into three stages.[1] In the first stage, the energy and moment are redistributed among carriers owing to carrier-carrier scattering; as a result, the electrons and holes attain thermal distributions characterized by an effective temperature T_c greater than the lattice temperature T_l; simultaneously, in a very rapid process the carriers lose most of their excess energy due to optical-phonon emission. In the second stage, T_c decreases until optical phonons are no longer efficient at removing the excess energy. Finally, in the third stage the cooling of both carriers and optical phonons to the lattice temperature occurs through acoustic-phonon emission. Under the physical conditions relevant to our study there exists experimental evidence of a process that significantly reduces the plasma density;[2] in this experiment, the time-resolved luminescence of GaAs in air at room temperature, under laser surface excitation (wavelength $\simeq 0.53$ μm, duration 30 ps) is used to investigate the plasma energy and density relaxation. It was observed that the plasma density increases until the end of excitation pulse, with energy density equal to 10 mJ cm^{-2}, up to $\simeq 1.2\times 10^{19}$ cm^{-3}, then falls down by one order of magnitude in the following 100 ps (cf. Fig. 3 in Ref. 2). The aim of this work is to investigate the effect of this variation of the concentration in the plasma behavior.

Let us consider a direct-band-gap polar semiconductor, illuminated by an intense monochromatic laser beam, in which direct absorption of one photon occurs, producing transitions from the valence band to the conduction band. We assume that the conditions are such that a two-component Fermi liquid of electrons and holes is formed and that this photoexcited plasma releases its excess energy through the following relaxation channels. (i) Radiative recombination (we consider spontaneous emission only). (ii) Carrier–LO-phonon scattering with polar interaction (Fröhlich) and with deformation-potential interaction. (iii) Hole–TO-phonon scattering with deformation-potential interaction. (iv) Carrier–acoustic-phonon scattering with deformation-potential interaction. To take into account the variation of the concentration, we assume that the photoinjected carriers can depart from the active region via ambipolar diffusion. The semiconductor sample is taken as an open system in contact with external ideal reservoirs, composed of the laser and a thermostat. The LO and TO phonons are taken as free to depart from equilibrium, and we consider for them, in addition to the interaction with the carriers, the anharmonic interaction described in a relaxation-time approximation. The acoustic phonons are supposed to constantly remain in equilibrium with the thermostat. To study this system we resort to the method developed by Zubarev[3] that allows the investigation of nonequilibrium situations. The derivation of the nonlinear generalized transport equations that govern the irreversible evolution of the system has been given in a previous paper,[4] and here we will proceed directly to use those equations. Following Ref. 4, we must choose a set of dynamical quantities $\{P_1,P_2,\ldots,P_r\}$ whose nonequilibrium mean values in Zubarev's ensemble are the macrovariables $\{Q_1,Q_2,\ldots,Q_r\}$ that are observed and/or controlled in the experiment. In the situations we are going to consider, we choose for quantities Q_j the following: $E_c(t)$, the energy of the carriers: $n(t)=n_e(t)=n_h(t)$, the concentrations of electrons and holes (these are equal because production and annihilation occur in pairs): $\nu^{\mathrm{LO}}(q_i,t)$ and $\nu^{\mathrm{TO}}(q_i,t)$, the distribution functions for longitudinal and transverse optical phonons, respectively.

The extensive macrovariable Q_j are thermodynamically conjugated to one set of intensive variables $\{F_1,F_2,\ldots,F_r\}$ in the sense that

$$Q_j=-\frac{\partial\phi}{\partial F_j}\ , \tag{1}$$

where ϕ is the logarithm of the nonequilibrium partition

function, $\phi=\ln Z(F_1,\ldots,F_r)$. In our case the variables F_j are interpreted as $F_c=\beta_c(t)=1/k_B T_c(t)$, the reciprocal effective carrier temperature, $F_{\mu_e}=-\beta_c(t)\mu_e(t)$; $F_{\mu_n}=-\beta_c(t)\mu_h(t)$, where $\mu_e(t)$ and $\mu_h(t)$ are the quasichemical potentials of electrons and of holes, respectively; and $F_{\rm LO}(q_i,t)$ and $F_{\rm TO}(q_i,t)$ are the intensive variables related to the LO- and TO-phonon populations, respectively.

These variables satisfy the equations[3]

$$\sum_{l=1}^{3}(P_j;P_l|t)\dot{F}_i(t)=\sum_{m=0}^{\infty}J_j^{(m)}(t)\ , \tag{2a}$$

$$\dot{\nu}^{\gamma}(q_i,t)=\sum_{m=0}^{\infty}J_j^{(m)}(t)\ , \tag{2b}$$

where $(P_j;P_l|t)$ are nonequilibrium correlation functions and $J_{j(\gamma)}^{(m)}$ are collision operators of order m in the interaction strengths. Equations (2) are the nonlinear generalized transport equations that govern the irreversible evolution of the system; $\gamma=$LO or TO.

Let us now investigate how the change in the carrier concentration, owing to ambipolar diffusion, can influence the relaxation process. Equations (2a), explicitly written, are

$$\mathcal{C}_1\dot{\beta}_c-\mathcal{C}_2(\beta_c\dot{\mu}_e+\mu_e\dot{\beta}_c)-\mathcal{C}_3(\beta_c\dot{\mu}_h+\mu_h\dot{\beta}_c)=\sum_{m=0}^{\infty}J_1^{(m)}(t)=-\dot{E}\ , \tag{3a}$$

$$\mathcal{C}_2\dot{\beta}_c-\mathcal{C}_4(\beta_c\dot{\mu}_e+\mu_e\dot{\beta}_c)=\sum_{m=0}^{\infty}J_2^{(m)}(t)=-\dot{n}_e\ , \tag{3b}$$

$$\mathcal{C}_3\dot{\beta}_c-\mathcal{C}_5(\beta_c\dot{\mu}_h+\mu_h\dot{\beta}_c)=\sum_{m=0}^{\infty}J_3^{(m)}(t)=-\dot{n}_h\ , \tag{3c}$$

where

$$\mathcal{C}_1=(H;H|t)=\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{e2}f_{\mathbf{k}}^{e}(1-f_{\mathbf{k}}^{e})+\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{h2}f_{\mathbf{k}}^{h}(1-f_{\mathbf{k}}^{h})\ , \tag{4a}$$

$$\mathcal{C}_2=(H;n_e|t)=\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{e}f_{\mathbf{k}}^{e}(1-f_{\mathbf{k}}^{e})\ , \tag{4b}$$

$$\mathcal{C}_3=(H,n_h|t)=\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{h}f_{\mathbf{k}}^{h}(1-f_{\mathbf{k}}^{h})\ , \tag{4c}$$

$$\mathcal{C}_4=(n_e;n_e|t)=\sum_{\mathbf{k}}f_{\mathbf{k}}^{e}(1-f_{\mathbf{k}}^{e})\ , \tag{4d}$$

$$\mathcal{C}_5=(n_h;n_h|t)=\sum_{\mathbf{k}}f_{\mathbf{k}}^{h}(1-f_{\mathbf{k}}^{h})\ , \tag{4e}$$

H is the system Hamiltonian,

$$f_{\mathbf{k}}^{\alpha}(t)=(\exp\{\beta(t)[\varepsilon_{\mathbf{k}}^{\alpha}-\mu_{\alpha}(t)]\}+1)^{-1},\quad \alpha=e \text{ or } h\ ,$$

are the Fermi-Dirac functions, and

$$\varepsilon_{\mathbf{k}}^{e}=E_G+\frac{\hbar^2k^2}{2m_e},\quad \varepsilon_{\mathbf{k}}^{h}=\frac{\hbar^2k^2}{2m_h}\ .$$

In the case under consideration the energy and carrier concentration rates of change are given by

$$\dot{E}=\left[\frac{dE}{dt}\right]_L+\left[\frac{dE}{dt}\right]_f+\left[\frac{dE}{dt}\right]_R+\left[\frac{dE}{dt}\right]_a\ , \tag{5a}$$

$$\dot{n}_e=\dot{n}_h=\dot{n}=\left[\frac{dn}{dt}\right]_L+\left[\frac{dn}{dt}\right]_R+\left[\frac{dn}{dt}\right]_a\ , \tag{5b}$$

where L denotes the interaction with the laser, f with LO, TO, and acoustic phonons; R expresses the radiative recombination, and a the ambipolar diffusion.

We can derive, from (3), the carrier effective temperature rate

$$\dot{T}_c=k_BT_c^2\frac{\dot{E}-\left[\frac{\mathcal{C}_2}{\mathcal{C}_4}+\frac{\mathcal{C}_3}{\mathcal{C}_5}\right]\dot{n}}{\mathcal{C}_1-\left[\frac{\mathcal{C}_2^2}{\mathcal{C}_4}+\frac{\mathcal{C}_3^2}{\mathcal{C}_5}\right]}\ . \tag{6}$$

Using Eqs. (4) it can be shown that the denominator is always positive. To deal with the ambipolar diffusion we introduce the relaxation time τ_a and write

$$\left[\frac{dE}{dt}\right]_a=-\frac{E}{\tau_a}=\frac{-1}{\tau_a}\left[\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{e}f_{\mathbf{k}}^{e}+\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}^{h}f_{\mathbf{k}}^{h}\right]\ , \tag{7a}$$

$$\left[\frac{dn}{dt}\right]_a=-\frac{n}{\tau_a}=\frac{-1}{\tau_a}\sum_{\mathbf{k}}f_{\mathbf{k}}^{e}=\frac{-1}{\tau_a}\sum_{\mathbf{k}}f_{\mathbf{k}}^{h}\ . \tag{7b}$$

Looking for the effect of the ambipolar diffusion on the rate of change of T_c we set

$$\dot{E}=\left[\frac{dE}{dt}\right]_a$$

and

$$\dot{n}=\left[\frac{dn}{dt}\right]_a$$

in Eq. (6) to verify, after some algebraic manipulation and remembering that f_k^{α} is a monotonic decreasing function of the energy, that the quantity

$$\dot{E}-\left[\frac{\mathcal{C}_2}{\mathcal{C}_4}+\frac{\mathcal{C}_3}{\mathcal{C}_5}\right]\dot{n}=\frac{1}{\tau_a}\left[\frac{\sum_{\mathbf{k},\alpha}\varepsilon_{\mathbf{k}}^{\alpha}f_{\mathbf{k}}^{\alpha}(1-f_{\mathbf{k}}^{\alpha})\sum_{\mathbf{k},\alpha}f_{\mathbf{k}}^{\alpha}-\sum_{\mathbf{k},\alpha}\varepsilon_{\mathbf{k}}^{\alpha}f_{\mathbf{k}}^{\alpha}\sum_{\mathbf{k},\alpha}f_{\mathbf{k}}^{\alpha}(1-f_{\mathbf{k}}^{\alpha})}{\sum_{\mathbf{k},\alpha}f_{\mathbf{k}}^{\alpha}(1-f_{\mathbf{k}}^{\alpha})}\right] \tag{8}$$

is always non-negative.

Since in Eq. (6), as noted, the denominator is always positive, and Eq. (8) tells us that the numerator is also positive, we find that

$$\left[\frac{dT_c}{dt}\right]_a \geq 0 \ . \tag{9}$$

Therefore we conclude that the ambipolar diffusion term contributes to a warm-up of the plasma.

In order to evaluate the effect quantitatively let us consider the experimental situation described in Ref. 2: a GaAs sample, in contact with a heat bath at room temperature, is illuminated by a laser pulse with exciting energy density equal to 10 mJ cm^{-2}, wavelength $\sim 0.53\ \mu m$, and a duration of 30 ps.

In our calculation we have used a laser pulse of rectangular time profile. To describe the anharmonic decay of different LO- and TO-phonon mode populations through intraband scattering toward their internal thermalization, we introduce "single-mode" relaxation times. We take these times to be constant and, guided by experimental observation, we choose for their values 7 ps.[5] The ambipolar diffusion time is written as $\tau_a = L^2/D$, where L is the diffusion length and D the ambipolar diffusion coefficient. The calculation of these figures, in nonequilibrium situations, is a difficult task; thus, instead of calculating their values, we take L equal to the inverse of the absorption coefficient, 10^{-4} cm, and $D = 2\times 10^{-10}$ $cm^2\ ps^{-1}$,[6] so that $\tau_a = 50$ ps. (It is in good agreement with experiment.[2]) The initial time is given by the condition $n(t_i) \sim 1\times 10^{16}$ cm^{-3}, i.e., the system is on the metallic side of the Mott transition. In the present case t_i is of the order of femtoseconds and we take $T(t_i) = (\hbar\omega_L - E_G)/3k_B = 3488$ K, and, for the phonon populations we take the equilibrium values.

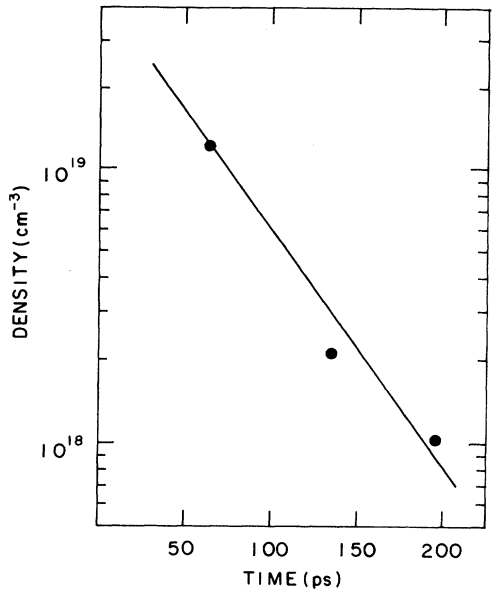

FIG. 2. Carrier-density kinetics calculated with an ambipolar diffusion relaxation-time constant set equal to 50 ps. Dots are experimental data from Ref. 2.

Finally, the solution of Eqs. (2) up to terms of second order in the interactions is obtained resorting to numerical methods. The results are shown in Figs. 1 and 2. Figure 1 shows the evolution of the effective temperature of carriers with (solid line) and without (dashed line) the ambipolar diffusion term. Dots are experimental data from Ref. 2. The significant effect of diffusion can be observed; this result shows that ambipolar diffusion ought to be taken into account in the study of the semiconductor

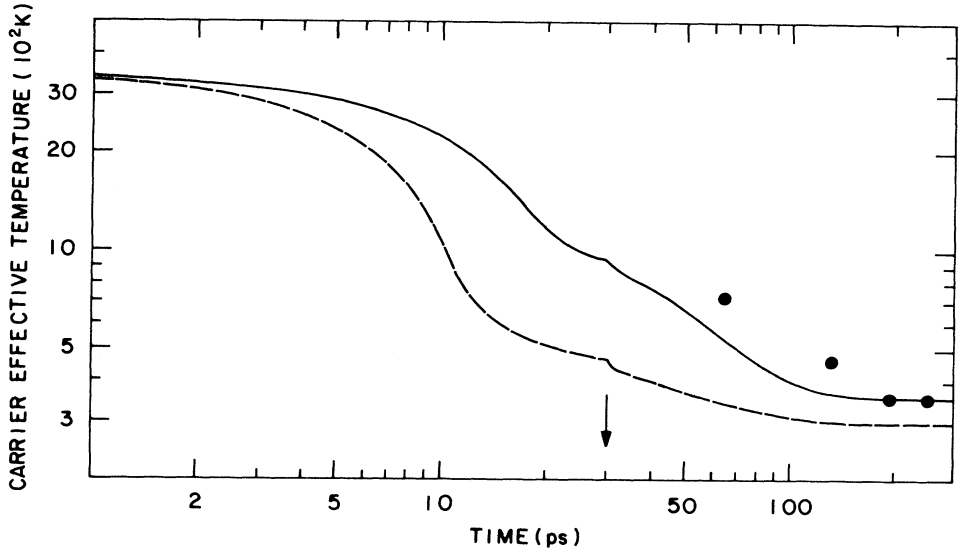

FIG. 1. Evolution of the carrier effective temperature. Dots are experimental data from Ref. 2. The upper curve is calculated with the effect of the ambipolar diffusion taken into account; in the lower curve this effect is neglected. The arrow indicates the end of the laser pulse.

hot-carrier relaxation and that it helps to explain the lower than expected effectivity observed in the cooling of the carriers at high concentration levels. Figure 2 depicts the plasma density kinetics. Dots are experimental data from Ref. 2. The rate of change of concentration owing to radiative recombination, in this stage of the process, is negligible compared with the rate of change due to ambipolar diffusion, so the calculated result is one straight line with a 50-ps slope. We note that the experimental relaxation time is not constant; it is smaller than 50 ps at the onset of the process and larger than 50 ps later on. This shows that with a more realistic value for τ_a, i.e., if we take into account its time dependence, a better agreement with expeimental values, in Fig. 1, should be obtained.

In conclusion, (i) it has been shown that the effect of the ambipolar diffusion term on the carrier temperature is to increase it and (ii) the significance of this term, in the study of the relaxation of photoexcited hot carriers in semiconductors, to explain the unexpectedly slow cooling of the hot-carrier distribution, is pointed out.

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