Cooling of hot carriers in highly photoexcited semiconductors

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The rate of energy loss of high-density hot electrons and holes in highly photoexcited plasma in direct-gap polar semiconductors is investigated. We compare the efficiency of the several relaxation channels, and show the relevant role played by the TO phonons. Further, we reconfirm that the rapid mutual thermalization of carriers and LO phonons produces a long plateau in the evolution curve for the carrier's effective temperature. GaAs was selected for numerical computations.

In recent experiments, high-intensity laser pulses have been used to generate nonequilibrium concentrations of carriers in semiconductors. These experiments are motivated by the need to look into problems like laser annealing, "hot carriers," nonlinear conduction, ultrafast device operation, etc. To understand these phenomena, it is necessary to comprehend the manner in which energy is transferred from the laser to the semiconductor lattice. It is well known that in polar semiconductors the relevant electron-phonon scattering processes are due to polar-optical, piezoelectric, and deformation-potential interactions. However, the dependence of energy transfer on the concentration and energy of carriers is not well elucidated.[1]

In this work we address this problem by considering the relaxation of dense and highly excited carriers in GaAs. For that purpose we resort to the method developed by Zubarev,[2] based on Jaynes's maximum-entropy formalism,[3] that enables one to derive the nonlinear generalized transport equations that govern the irreversible evolution of the highly photoexcited plasma in direct-band-gap polar semiconductors.

We assume that energy is pumped to the sample by an intense monochromatic laser beam and direct absorption of one photon occurs, producing transitions from the valence band to the conduction band. These carriers have an excess of kinetic energy given by $\hbar\omega_L - E_G$, where ω_L is the laser frequency and E_G the semiconductor energy gap. Let us assume that the conditions are such that a double Fermi fluid of electrons and holes is formed instead of the exciton gas, i.e., that the system is on the metallic side of the Mott transition, which typically occurs for values of concentration of the order of 10^{16} cm^{-3} and larger.[4] This photoexcited plasma releases its excess energy to the lattice through the following relaxation channels: radiative-recombination, Fröhlich, deformation-potential, and piezoelectric interactions. For the phonons, we consider the relaxation effects mediated by anharmonic interaction and heat diffusion to a thermostat. The semiconductor sample is taken as an open system in contact with external ideal reservoirs, i.e., macroscopic states do not change as a result of the interactions with the sample, composed of the laser and a thermostat. For this model, the nonequilibrium statistical-operator method in Zubarev's approach allows one to derive a set of nonlinear integro-differential equations that describe the nonequilibrium thermodynamic state of this highly excited plasma in semiconductors (HEPS).

In this method, a basic set of macroscopic variables must be chosen. These variables must be appropriate for the description of the nonequilibrium state of the system during the characteristic time scale of the experiment. Following the notation of Ref. 5, we select as variables P_i, $i=1,\ldots,6$. $P_1=E_c(t)$, the energy of the carriers; $P_2=E_{\text{LO}}(t)$, the energy of the LO phonons; $P_3=E_{\text{TO}}(t)$, the energy of the TO phonons; $P_4=E_A(t)$, the energy of the A phonons; and $P_5=P_6=n(t)$, the concentration of electrons and holes (these are equal because electrons and holes are produced in pairs). P_j are the set of dynamical quantities whose mean values, in Zubarev's nonequilibrium ensemble, are the macrovariables $Q_j(t)$ which describe the macroscopic state of the system.[2] Finally, the set of intensive nonequilibrium variables thermodynamically conjugated to the above macrovariables is F_i, $i=1,\ldots,6$. The first four are reciprocal effective temperatures $\beta(t)\equiv 1/kT(t)$, which are analogous to the energy variables: $F_1=\beta_c(t)$, $F_2=\beta_{\text{LO}}(t)$, $F_3=\beta_{\text{TO}}(t)$, and $F_4=\beta_A(t)$. The other two are $F_5=-\beta_c(t)\mu_e(t)$ and $F_6=-\beta_c(t)\mu_h(t)$, in which $\mu_e(t)$ and $\mu_h(t)$ are the quasichemical potentials of the electrons and holes, respectively.

The concentration $n(t)$ and the quasichemical potentials are connected, once the internal thermalization of carriers has occurred, by the relations

$$n(t)=n_e^0(t)F_{1/2}(\beta_c(t)\mu_e(t))$$
$$=n_h^0(t)F_{1/2}(\beta_c(t)\mu_h(t)) ,$$

where

$$n_\alpha^0(t)=2[2\pi m_\alpha kT_c(t)/\hbar^2]^{3/2} ,$$

and $F_{1/2}$ are Fermi functions of index one-half[6] and α is e or h.

In the case under consideration, a set of equations for the evolution of the conjugate intensive variables was ob-

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TABLE I. Initial conditions at t_i, and parameters used in calculations.

	t_i (ps)	$n \times 10^{15}$ cm^{-3}	T_c^* (K)	$T_{\rm LO}^*$ (K)	$T_{\rm TO}^*$ (K)	T_A^* (K)	T_B^* (K)
Case 1	0	5.5	8911	35	32	10	10
Case 2	−0.6	5.3	9166	34	31	10	10
Case 3	−0.6	10	9500	300	300	300	300
	t_L (ps)	I_L (W cm^{-2})	$\hbar\omega_L$ (eV)	$\tau_{{\rm LO},A}$ (ps)	$\tau_{{\rm TO},A}$ (ps)	$\tau_{A,B}$ (ps)	
Case 1	1	7.4×10^6	4	60	60	46	
Case 2	1	7.4×10^8	4	60	60	46	
Case 3	1	1.5×10^9	4	30	30	4.6×10^5	

tained. The equations are an extension of those derived in a previous article[5] once we remove the restriction of keeping TO and A phonons in equilibrium with the thermostat. The new equations are formally similar to those derived for LO phonons in Ref. 5, except for the replacement of the appropriate parameters and interaction strengths. It should be noted that in the calculations we have used a random-phase-approximation (RPA) screening for the carrier-phonon interactions. Also included are anharmonic interactions resulting from the decay of an optical phonon into two acoustic phonons. These are described in a relaxation-time approximation in terms of the relaxation times $\tau_{{\rm LO},A}$ and $\tau_{{\rm TO},A}$, which can be evaluated from linewidths of Raman lines.[7] Further, the heat diffusion from A phonons to the thermostat was also described introducing a relaxation time, $\tau_{A,B}$, which depends on the diffusion coefficients and the dimensions of the surface of the active volume of the sample.[8] This time must be estimated for each case when performing specific numerical calculations.

Next, we solve these equations for the case of a GaAs sample receiving a laser pulse with a Gaussian time profile of length t_L and peak intensity I_L. We are interested in high levels of excitation. For this purpose, we take a great excess of energy for the carriers ($\hbar\omega_L - E_G \simeq 2.5$ eV). We consider two cases: In one, the density attained is $n = 1\times10^{17}$ cm^{-3}, and in the other $n = 1\times10^{19}$ cm^{-3}. The values of the parameters and of the initial conditions at t_i (t_i is the instant when the carrier concentration attains the value $\sim 1\times10^{16}$ cm^{-3}) used

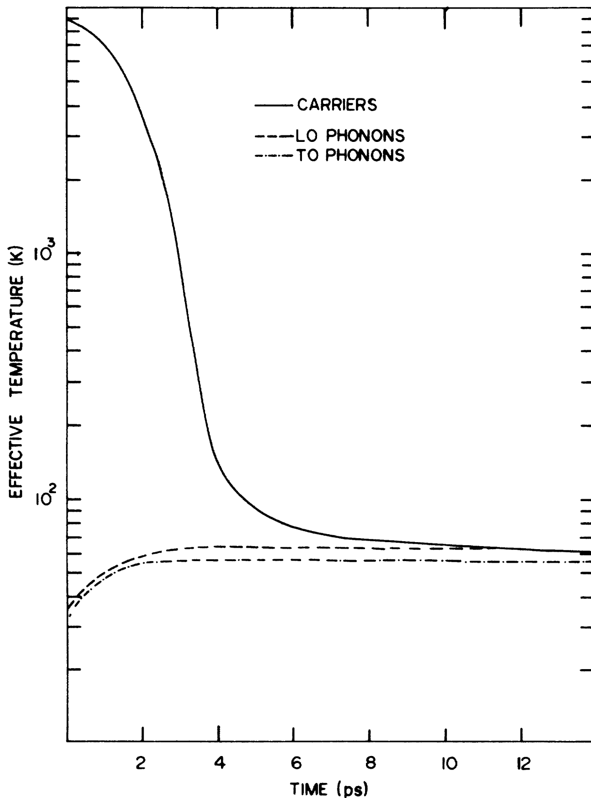

FIG. 1. Time evolution of the effective temperatures of carriers and TO and LO phonons in case 1.

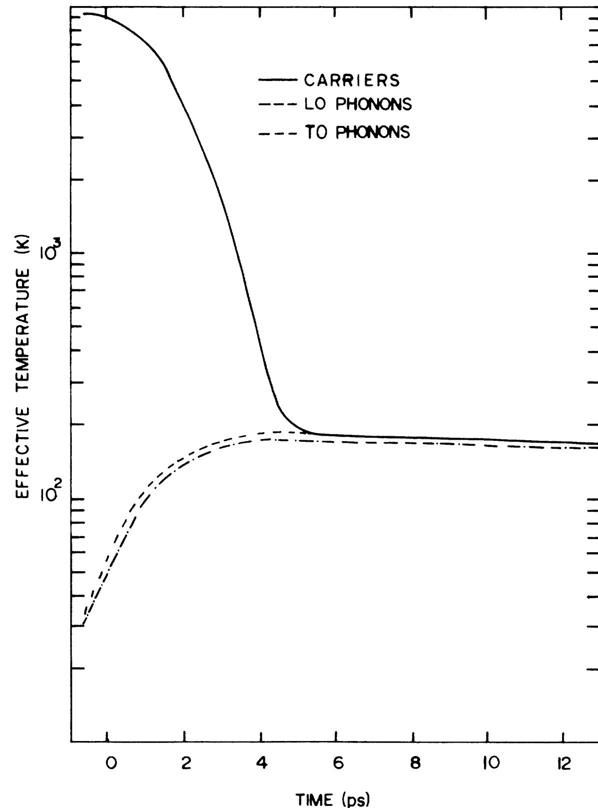

FIG. 2. Time evolution of the effective temperatures of carriers and TO and LO phonons in case 2.

in the calculation for those two different situations are given in Table I.

Figures 1 and 2 show the evolution of the effective temperatures of carriers and LO and TO phonons. It can be observed that the carrier effective temperature rapidly decreases until it nearly equals the effective temperature of LO phonons. Then, LO phonons are reabsorbed and the nonpolar TO interaction dominates the carrier's energy relaxation. Finally, in a third stage, the three subsystems proceed towards thermal equilibrium with the thermal bath in processes governed by relaxation of energy to the A phonons together with heat diffusion. The effective temperature of the A phonons remains practically equal to the thermal-bath temperature in the first case and is slightly larger in the second.

Figures 3 and 4 show the rates of energy transfer between carriers and phonons. We note that for high levels of carrier excitation (excess energy 0.8 and 0.3 eV, respectively) the relaxation to TO phonons is dominant. The rates of polar and nonpolar interaction for LO phonons show that for high levels of carrier excitation the nonpolar interaction is greater. The effect of the plasma density on the relaxation rate can be seen by comparing Fig. 3 ($n \sim 10^{17}$ cm^{-3}) with Fig. 4 ($n \sim 10^{19}$ cm^{-3}).

To determine the effect of screening on the evolution of the relaxation of energy to the lattice, we consider the situation of case (C) in Ref. 9, with the initial conditions listed in Table I as case 3. Figure 5 shows the rate of energy transfer from carriers to LO phonons (upper part) via (A) Fröhlich interaction screened by nonequilibrium carriers calculated in the RPA, and (B) due to the bare interaction. In the lower part is shown the time evolution of the carrier effective temperature. The carrier density is 2×10^{19} cm^{-3}. It can be seen that even at this high concentration screening does not sensibly alter the nonequilibrium thermodynamic behavior of the excited plasma. This result reinforces our previous conclusions.[9] The carriers thermalize with optical phonons at $\sim$380 K and the A phonon's effective temperature at 9 ps is $\sim$336 K.

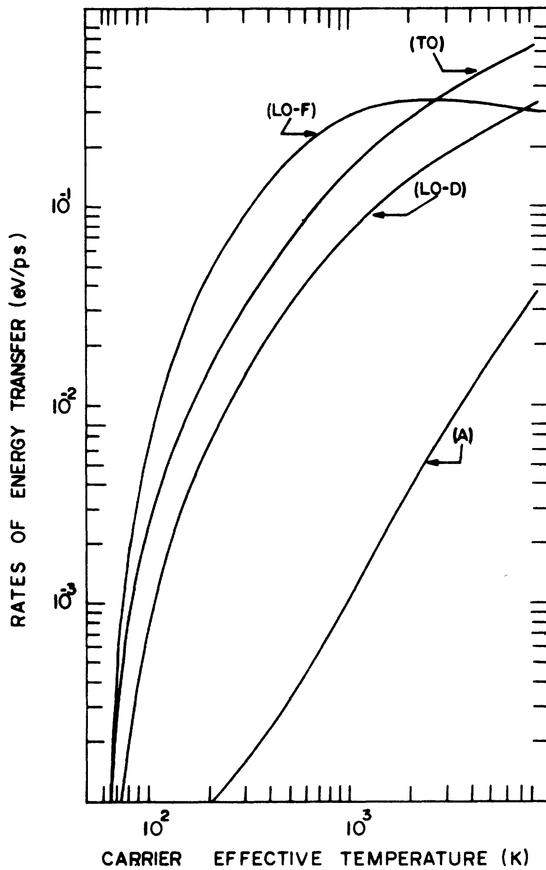

FIG. 3. Rates of energy transfer per pair vs carrier effective temperature in case 1: (LO-F) denotes carriers to LO phonons via Fröhlich interaction, (LO-D) carriers to LO phonons via deformation-potential interaction, (TO) carriers to TO phonons, and (A) carriers to A phonons via deformation-potential interaction.

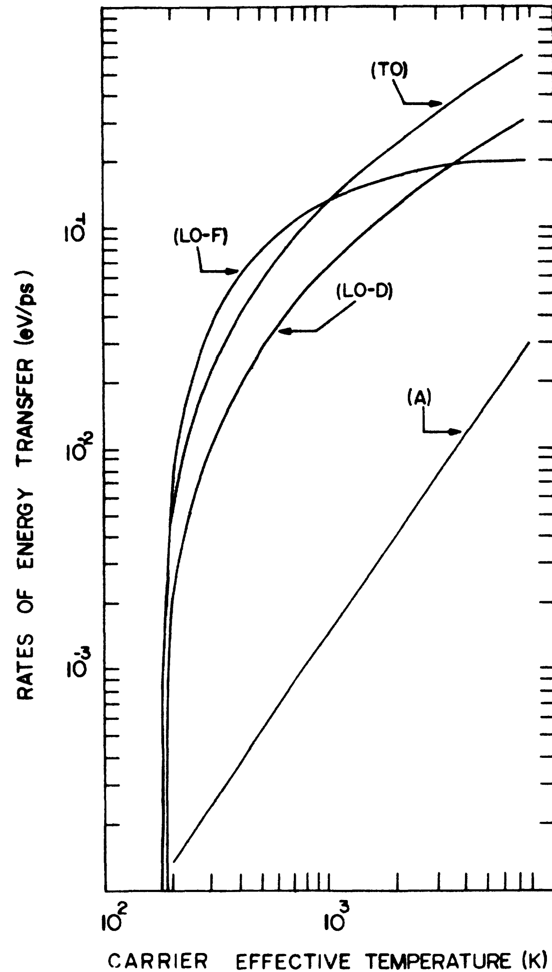

FIG. 4. Rates of energy transfer per pair vs carrier effective temperature in case 2: (LO-F) denotes carriers to LO phonons via Fröhlich interaction, (LO-D) carriers to LO phonons via deformation-potential interaction, (TO) carriers to TO phonons, and (A) carriers to A phonons via deformation-potential interaction.

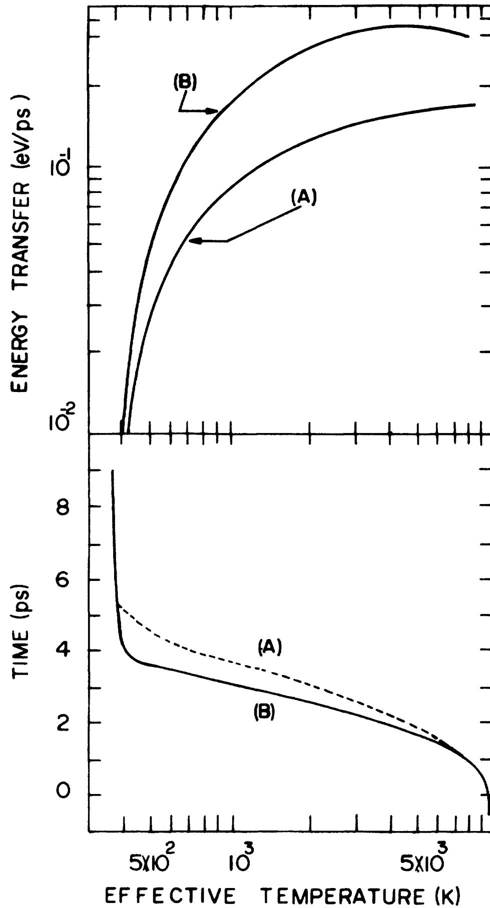

FIG. 5. Dependence of the rate of energy transfer per pair from carriers to LO phonons in case 3. Upper part: via (A) RPA-screened Fröhlich interaction and (B) bare Fröhlich interaction. In the lower part is shown the time evolution of the carrier effective temperature.

It should be noted that we have applied the concept of effective temperatures to the optical-phonon branches in a stage of the relaxation processes when their state should be better described by the complete set of occupation numbers, ν_q, and, consequently, our results are valid within this approximation. Preliminary results showing the evolution of the optical-phonon distribution functions suggest that mutual thermalization of the whole set of phonon modes and the carriers follows in a 10-ps time scale.[10]

In conclusion, we have used the nonequilibrium statistical-operator method in Zubarev's approach to study the behavior of direct-gap polar semiconductors illuminated by a very short intense pulse of laser light. GaAs was selected for numerical computations. The main results are (i) a long plateau in carrier effective temperatures versus delay time because of the rapid mutual thermalization of the carriers and optical phonons (this long plateau agrees with experimental observation); (ii) negligible heating of the acoustical phonons, when the thermal bath is at room temperature, except for very high levels of excitation; (iii) deformation-potential interactions become a stronger source for carrier excess-energy relaxation than the Fröhlich interaction (this depends on excitation conditions and is a result of the fact that for highly excited carriers the total scattering rate decreases with increasing energy owing to the electrostatic nature of the polar-optical interaction); (iv) after a very fast initial state (after the carriers have lost most of their excess energy), the main relaxation mechanism is the emission of TO phonons by the holes; (v) the effect of the piezoelectric interaction is negligible in this stage of the carrier relaxation, owing to the thermalization of the carriers with the optical phonons at effective temperatures greater than the acoustical-phonon effective temperature; and (vi) screening effects seem to be relevant only in the very early stages of the relaxation processes and at high concentrations of carriers ($n > 10^{20}$ cm^{-3}).

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