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Hot carrier relaxation and phonon dynarnics in GaAs

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Abstract We compare the rates of energy loss of hot electrons and hot holes in highly photoexcited plasma in GaAs and show the crucial role played by holes in plasma relaxation. Further, we determine the time evolution of the nonequilibrium distribution functíons of optical (longitudinal and transversal) phonons.

In order to understand problems like laser annealing, nonlinear conduction, ultrafast device operation etc., it is necessary to comprehend the manner in which the highly excited carriers transfer their energy excess to the lattice. It is known that, owing to this relaxation process, nonthermal phonon distributions are produced[1]. The dynamics of those nonequilibrium phonons plays a essential role in the hot carriers' relaxation, so that the two phenomena are correlated.

In this work we consider a GaAs sample excited by a short pulse of intense laser light and investigate the relaxation of the excited carriers by taking into account the rates of energy transfer between electrons and phonons. We also study the evolution of the distribution functions of LO and TO phonons.

Let us consider a GaAs sample under an intense monochromatic laser pulse with a Gaussian time profile of length $t_L = 6$ ps, peak intensity $I_L = 2.7 \times 10^9$ W cm^{-2}, and frequency $\omega_L = 3.4 \times 10^{15}\ s^{-1}$, which are typical experimental values[2]. Carriers are then produced with an excess kinetic energy given by $\hbar\omega_L - E_G$, where E_G is the semiconductor energy gap. We assume that the conditions are such that a double Fermi fluid of electrons and holes is formed, i.e., that the system is on the metallic side of the Mott transitions, which happens at a time

t_i when the carrier concentration attains the values $\sim 1 \times 10^{16}$ cm^{-3}, [3] and that this photoexcited plasma release its excess energy through the following relaxation channels: radiative-recombination, Frõhlich, deformation-potential, and piezoelectric interactions. For the phonons, we consider the relaxation effects mediated by an anharmonic interaction described in a relaxation time approximation. (See equations **28** and **29** of ref. 4). The relaxation times $\tau_{LO,A}$ and $\tau_{TO,A}$ can be evaluated from linewidths of Raman lines[5], and have the value 30 ps; this value allows a good fitting of experimental data at long delay times. We also take into account the heat diffusion to a thermostat (at a temperature $T_B = 300K$) introducting a relaxation time $\tau_{A,B}$ given by[6]

$$\tau_{A,B} = \frac{L^2 C(T_A)}{K(T_A)} \tag{1}$$

where C is the specific heat, K the thermal conductivity of the lattice, L an average linear dimension of the active volume of the sample, and T_A the effective temperature of A phonons. $C(T_A)$ was obtained from ref. *(6)* and $K(T_A)$ from ref. (7), and we take,

$$L^2 = \left(\frac{S}{\alpha}\right)^{2/3} \tag{2}$$

where S is the laser spot area and α the absorption coefficient. According to experimental conditions we have the spot size ~ 1 mm in diameter; a $= 9.4 \times 10^3$ cm^{-1}, and $T_A = 300K$.[2] Numerical calculations gives $\tau_{A,B} = 4.6 \times 10^5$ ps.

We note that whereas $\tau_{A,B}$ is related to a diffusion process, $\tau_{LO,A}$ and $\tau_{TO,A}$ are related to a local effect, which explains the difference in their values. The semiconductor sample is taken as an open system in contact with external ideal reservoirs, composed of the laser and a thermostat.

To investigate this system we resort to the method developed by Zubarev[8] that enables one to derive the time evolution of the distribution functions of carriers and optical phonons in the highly photoexcited semiconductor. The derivation of the nonlinear generalized transport equations (NGTE) that govern the irreversible evolution of the system once we remove the restriction of keeping TO and A

phonons in equilibrium with the thermostat was done in a previous paper[4], and here we will go directly to those equations. Following ref. 4 we have

$$\sum_{i=1}^{n}(P_j; P_i|t)\dot{F}_i(t) = \sum_{\ell=0}^{\infty} J_j^{(\ell)}(t) \tag{3}$$

where P_j are the set of dynamical quantities whose mean values, in Zubarev's nonequilibrium ensemble, are the macrovaribles $Q_j(t)$ which describe the macroscopic state of the system, F_j is the set of intensive nonequilibrium variables thermodynamically conjugated to the macrovariables $Q_j(t)$; $J_j^{(\ell)}$ is the collision operator of order ℓ; and $(P_j; P_i|t)$ are the nonequilibrium correlation functions. In the case we are now considering we select as variables P_j, $j = 1...6$, $P_1 = E_c(t)$, the energy of the carriers; $P_2 = E_{LO}(t)$, the energy of the LO phonons; $P_3 = E_{TO}(t)$, the energy of TO phonons; $P_4 = E_A(t)$, the energy of A phonons; and $P_5 = P_6 = n(t)$ the concentrations of electrons and holes (these are equal because carriers are produced in pairs). F_j, $j = 1...6$, the first four are reciproca1 effective temperatures $\beta(t) = 1/kT$ which are analogous to the energy variables, $F_1 = \beta_c(t)$, $F_2 = \beta_{LO}(t)$, $F_3 = \beta_{TO}(t)$, 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 potential are connected, once the internal thermalization of the carriers has occured, by the relations

$$n(t) = n_\alpha^0(t)\ F_{1/2}(\beta_c(t)\mu_\alpha(t)) \tag{4}$$

where

$$n_\alpha^0(t) = 2[2\pi m_\alpha kT_c(t)\hbar^2]^{3/2}\ , \tag{5}$$

and $F_{1/2}$ is the Fermi function of index one-half and α is e or h.

To take into account the evolution of the populations of phonon modes we couple (3) with the equations

$$\frac{d}{dt}\nu_\gamma(\vec{q};t) = \sum_{\ell=0}^{\infty} J_j^{(\ell)}(\vec{q};t) \tag{6}$$

where $\nu_\gamma(\vec{q};t)$ is the phonon distribution function and 7 is LO, TO, *A*.

The initial time is given by the condition $n(t_1) = 1.0 \times 10^{16}$ cm^{-3}. In the present case $t_i = -5.5$ ps. ($t = 0$ corresponds to a laser intensity 1/e of the peak value). Estimating a production of ~ 5 $LO(TO)$ phonons per pair up to t_i and using energy conservation in the form $3\, k_B[T_{\text{excess}} - T(t_i)] = 5(\hbar\omega_{LO} + \hbar\omega_{TO})$ we find $T(t_i) \sim 1854K$. $T_{LO}(t_i)$ and $T_{LO}(t_i)$ are practically unaltered compared with equilibrium values, since the number of phonons produced in excess of equilibrium is much smaller than $N_{\text{eq.}} \sim 2 \times 10^{21}$ cm^{-3}.

It should be noted that the carrier-optical phonon interaction was obtained using a dielectric constant calculated in the random-phase approximation. This dielectric constant is given by

$$\epsilon(\vec{q},t) = 1 + \frac{q_0^2(t)}{q^2} \tag{7}$$

where

$$q_0^2(t) = \frac{4\pi e^2}{\epsilon_0 V} \sum_{k,\alpha} \left| \frac{\partial f_k^\alpha(t)}{\partial \epsilon_k^\alpha} \right| ; \tag{8}$$

$$f_k^\alpha(t) = \{1 + \exp[\beta_c(t)(\epsilon_k^\alpha - \mu_\alpha(t))]\}^{-1} . \tag{9}$$

Finally, the solution of the system NGTE up to terms of second order in the interactions is obtained resorting to numerical methods.

We investigated the relaxation of the carriers' excess energy and the evolution of the optical phonon populations. Fig. 1 shows the rates of energy transfer between carriers and phonons. We note that: a) the energy transferred to the lattice by the holes is significantly larger than that transferred by the electrons; b) the rate of energy transfer owing to the deformation-potential is equivalent to the rate owing to Fröhlich interaction; c) the energy transferred to TO-phonons is comparable with that transferred to the LO-phonons. Figures 2 and 3 show the time evolution of LO and TO phonon populations, respectively. It can be observed that nonequilibrium LO as well as TO phonons are produced in a small and off-center region of the Brillouin zone; this is a result of the fact that the rate of energy transfer is proportional to $1/q^3$ in the Fröhlich interaction and to $1/q$ in the

deformation-potential interaction, and the conservation of energy and momentum for the carrier-phonon scattering; the first factor inhibits the generation of large q modes and the second modes near band origin. Furthermore, in spite of the energy transferred to the TO phonons being comparable to that transferred to the LO phonons, LO modes are much farther from equilibrium than TO modes. This h'appens because the energy is transferred by Frõhlich interaction in a smaller region of q space.

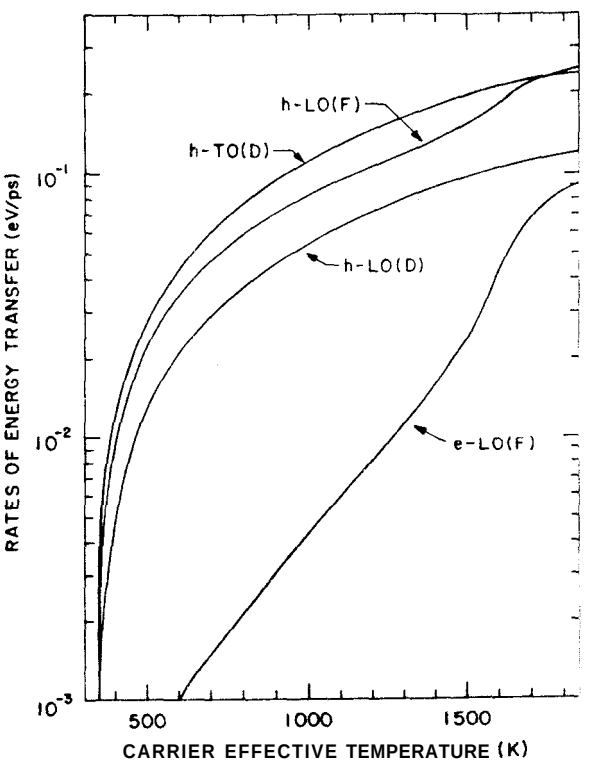

Fig. 1 - Rates of energy transfer per pair vs. carrier effective temperature: $h-LO(F)$ denotes holes to LO phonons via Frõhlich interaction, $h-TO(D)$ holes to TO phonons via deformation potential interaction, $h-LO(D)$ holes to LO phonons via deformation potential interaction, $e-LO(F)$ electrons to LO phonons via Frõhlich interaction.

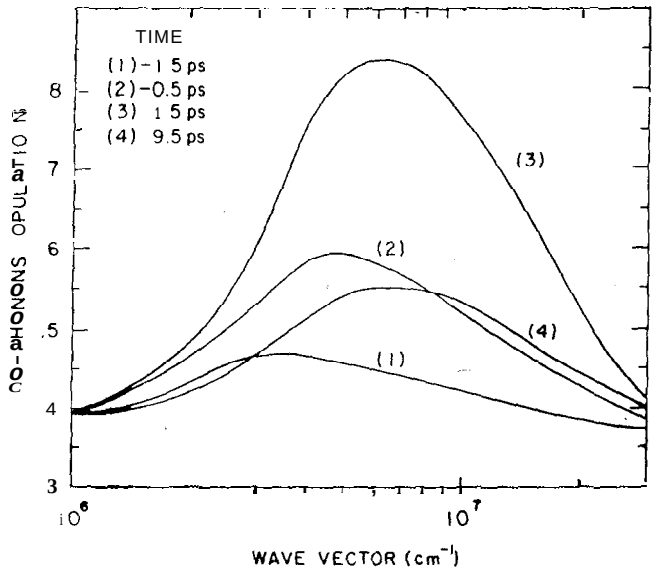

Fig. 2 - Time evolution of LO phonon population vs. wave vector.

In conclusion, the main results of this study are: (i) owing to the large difference between electron and hole rates of energy transfer and depending of experimental conditions, (carrier concentration and carrier excess energy) it is necessary to assume different electron and hole effective temperatures in the early stage of the relaxation[g]. (ii) In order to have a satisfactory description of the relaxation process it is necessary to take into account the effects of the deformation-potential and TO-phonons. (iii) The peak in the LO phonon population is due to the Frõhlich interaction. (iv) After a first stage, when a great generation of LO-phonons occurs, the nonequilibrium LO-phonon population inhibits the carrier-lattice energy transfer via Fröhlich interaction. As a consequence the carriers' relaxation time increases.

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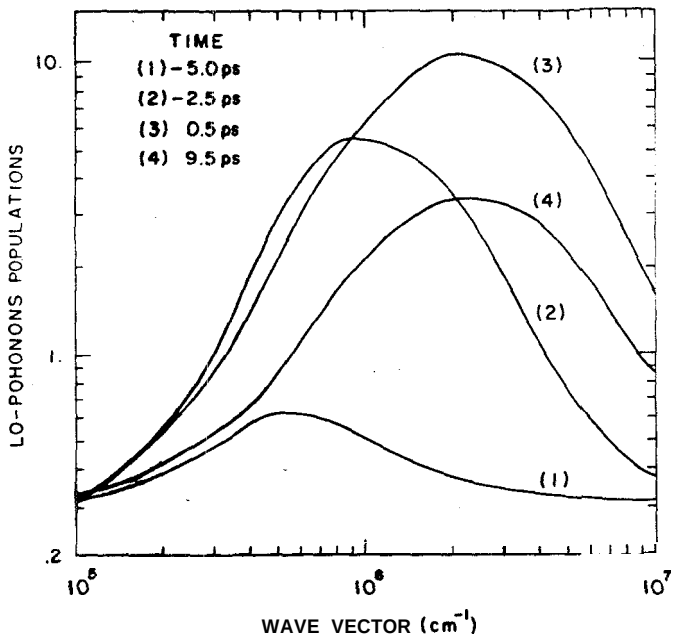

Fig. 3 - Time evolution of TO phonon population vs. wave vector.

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Resumo

Comparamos as **taxas** de transferência de energia de um plasma de elétrons e buracos gerado por fotoexcitação em GaAs, mostrando o importante papel desempenhado pelos buracos no processo de relaxação. Determinamos também a evolução temporal de distribuições não equilibradas de fonons óticos.