PHYSICAL REVIEW B VOLUME 49, NUMBER 11 15 MARCH 1994-I

Mechanism for LO-phonon temperature overshoot in GaAs

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(Received 28 June 1993)

Carrier relaxation and LO-phonon dynamics are investigated in GaAs crystals illuminated by picosecond laser pulses. It is evidenced that an overshoot of the LO-phonon quasitemperature above the electron quasitemperature can occur in the absence of intervalley scattering. This effect is ascribed to the screening of the polar optical interaction.

I. INTRODUCTION

The physical properties of hot photoinjected carriers in semiconductors, in spite of the large interest that generated a vast bibliography (see for example the reviews in Refs. 1–4), are not entirely understood. The first studies of relaxation of these carriers[5,6] showed that, at high concentration levels, their cooling was slower than expected on the basis of a simple model. Several suggestions were advanced to improve the model, namely the inclusion of the screening of the electron-phonon interaction,[7,8] hot-phonon effects,[9–11] intervalley scattering,[12] and ambipolar diffusion of hot carriers.[13,14] All these effects, depending on the features of the system, i.e., carrier concentration and excess excitation energy, are more or less significant for the relaxation processes. In addition, they are correlated among each other, making the problem quite complex. So further work, experimental as well as theoretical, seems to be necessary for their complete elucidation.

In a recent paper, Kim and Yu[15,16] reported the use of Raman scattering to study the relaxation process in a sample of GaAs excited by subpicosecond laser pules. At electron densities of about 10^{18} cm^{-3}, they observed a transient quasitemperature overshoot of longitudinal-optical (LO) phonons (with wave vector $q=7.5\times10^5$ cm^{-1}) above the electron quasitemperature. They explained this overshoot effect by the fast production of zone-centered LO phonons by hot electrons, combined with the slowed reabsorption of the emitted phonons due to a rapid cooling of Γ electrons due to intervalley scattering. The overshoot phenomenon is a very interesting one, and in the present paper we describe the possibility of its occurrence in the absence of intervalley scattering.

II. THEORETICAL MODEL

Let us consider an intrinsic GaAs sample at room temperature excited by a laser pulse with a Gaussian time profile of 1-ps full width at half maximum, excitation power of 30 μJ/cm^2, and photon energy $\hbar\omega=1.735$ eV. Under these conditions the excess energy per carrier is about 300 meV; since the energy difference between Γ and X valleys is 460 meV and between Γ and L valleys is 300 meV, the intervalley transition should be a minor effect. We assume that the conditions are such that a two-component Fermi liquid (i.e., electrons and holes photoproduced in pairs) is formed and that they release their excess energy through the following relaxation channels: (i) carrier-LO-phonon scattering via polar interaction and deformation potential interaction; (ii) hole-TO-phonon scattering via deformation potential interaction; and (iii) carrier-acoustic-phonon scattering via deformation-potential interaction. To take into account the variation of concentration, we assume that the photoinjected carriers can depart from the active region via ambipolar diffusion. To account for the ambipolar diffusion we consider the relaxation-time approximation, with a relaxation time $\tau_a=50$ ps.[17] The sample is taken to be an open system, in contact with external ideal reservoirs, the latter composed of the laser and a thermostat. The LO phonons are taken as free to depart from equilibrium, and for them we consider, in addition to the interaction with the carriers, the anharmonic interaction described in a relaxation-time approximation. The TO and acoustic phonons are supposed to remain constantly in equilibrium with the thermostat at 300 K.

To describe the macroscopic state of the system, we take into account that the photoinjected carriers attain a very rapid (subpicosecond) internal thermalization due to strong Coulomb interaction. Hence, for times in the picosecond scale, it is possible to introduce a contracted macroscopic description of the carriers in terms of a quasitemperature $T_c^*(t)$, and quasichemical potentials $\mu_e(t)$ and $\mu_h(t)$. On the other hand, this is not the case for the LO phonons, which require a (mesoscopic) description in terms of the populations $\nu_{\mathbf{q}}^{\mathrm{LO}}(t)$ of the different modes.[18] Therefore the distribution function of the internally thermalized carriers is an instantaneous Fermi-Dirac distribution:

$$f_{\mathbf{k}}^{\alpha}(t)=\frac{1}{e^{[E_{\mathbf{k}}^{\alpha}-\mu_\alpha(t)]/k_B T_c^*(t)}+1}\,, \tag{1}$$

and for the LO phonons it is a Planckian-like distribution,

$$\nu_{\mathbf{q}}^{\mathrm{LO}}(t)=\frac{1}{e^{\hbar\omega_{\mathbf{q}}^{\mathrm{LO}}/k_b T_{\mathbf{q}}^*(t)}-1}\,, \tag{2}$$

which defines a quasitemperature $T_{\mathbf{q}}^*$ per mode. $E_{\mathbf{k}}^{\alpha}$ are the carrier energies (we take the effective-mass approximation), α stand for electrons and holes, and $\omega_{\mathbf{q}}^{\mathrm{LO}}$ is the LO-phonon dispersion relation. The carrier quasitemperature and quasichemical potentials are derived from the expressions for the energy density and particle density, namely

$$E_c(t)=\frac{1}{V}\sum_{\mathbf{k},\alpha} E_{\mathbf{k}}^{\alpha} f_{\mathbf{k}}^{\alpha}(t) , \tag{3a}$$

$$N^{\alpha}(t)=\frac{1}{V}\sum_{\mathbf{k}} f_{\mathbf{k}}^{\alpha}(t) . \tag{3b}$$

The concentration of electrons and holes is the same since they are photoproduced in pairs in an intrinsic sample.

The equations that govern the time evolution of this system are given by

$$\frac{df_{\mathbf{k}}^{\alpha}(t)}{dt}=\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{L}+\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{\mathrm{FR}}+\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{\mathrm{OPT,DF}}+\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{\mathrm{AC}}+\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{\mathrm{DIF}} , \tag{4a}$$

$$\dot{\nu}_{\mathbf{q}}^{\mathrm{LO}}(t)=\dot{\nu}_{\mathbf{q}}^{\mathrm{FR}}(t)+\dot{\nu}_{\mathbf{q}}^{\mathrm{LO,DF}}(t)+\dot{\nu}_{\mathbf{q}}^{B}(t) , \tag{4b}$$

where index L stands for the optical generation of electron-hole pairs due to direct absorption; FR for the optical interaction between carriers and LO phonons through the screened Fröhlich potential; OPT,DF for nonpolar optical interaction via the deformation potential; AC for the carrier interaction with acoustic phonons through the deformation potential; DIF for the ambipolar diffusion of carriers; and B for the LO phonon interactions with the thermal bath.

Each of the interaction terms introduced in Eq. (4), except the one due to ambipolar diffusion of carriers, is written in terms of rate equations of the type[19]

$$\left.\frac{\partial f_{\mathbf{k}}(t)}{\partial t}\right|_{INT}=\sum_{\mathbf{k}'}\{f_{\mathbf{k}'}(t)[1-f_{\mathbf{k}}(t)]W(\mathbf{k}',\mathbf{k})-f_{\mathbf{k}}(t)[1-f_{\mathbf{k}'}(t)]W(\mathbf{k},\mathbf{k}')\} , \tag{5}$$

where index INT stands for the different type of mechanisms present in Eq. (4a), and W is the quantum transition rate given by Fermi's golden rule averaged over the phonon (or photon) ensemble, namely

$$W(\mathbf{k},\mathbf{k}\pm\mathbf{q})=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}|\langle\mathbf{k}\pm\mathbf{q}|H'|\mathbf{k}\rangle|^2\times\{\nu\delta(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega_{\mathbf{q}})+(\nu+1)\delta(E_{\mathbf{k}-\mathbf{q}}-E_{\mathbf{k}}+\hbar\omega_{\mathbf{q}})\} , \tag{6}$$

where ν is the phonon population (or photon, depending on the interaction considered) and $\langle\mathbf{k}\pm\mathbf{q}|H'|\mathbf{k}\rangle$ the matrix element associated to the given transition mechanism. Similar expressions to the one in Eq. (5) follow for the different terms in Eq. (4b), except for the term of interaction with the thermal bath.

Ambipolar diffusion and LO-phonon decay are treated phenomenologically in the relaxation-time approximation, which we express as

$$\left.\frac{\partial f_{\mathbf{k}}^{\alpha}(t)}{\partial t}\right|_{\mathrm{DIF}}=-\frac{f_{\mathbf{k}}^{\alpha}(t)-f_{\mathbf{k}}^{\alpha}(\mathrm{equil})}{\tau_D} , \tag{7a}$$

where τ_D is the parameter that determines the rate of the process, and

$$\left.\frac{\partial \nu_{\mathbf{q}}^{\mathrm{LO}}(t)}{\partial t}\right|_{B}=-\frac{\nu_{\mathbf{q}}^{\mathrm{LO}}-\nu_{\mathbf{q}}^{\mathrm{LO}}(\mathrm{equil})}{\tau_B} , \tag{7b}$$

with τ_B being the relaxation time.

Two aspects lead us to consider the LO phonons as out of internal equilibrium. One is that the excitation rate of the LO phonons in the first stages of the relaxation process is larger than their decay rate; that is the cause of the hot phonons' appearance. The other is the fact that there is a narrow region of $\mathbf{q}$-reciprocal space that couples carriers and LO phonons very effectively.

The LO-phonon time evolution is governed by the equation

$$\dot{\nu}_{\mathbf{q}}^{\mathrm{LO}}(t)=\frac{2\pi}{\hbar}\sum_{\alpha}\sum_{\mathbf{k}}|M_q|^2\{f_{\mathbf{k}}^{\alpha}(t)[1-f_{\mathbf{k}-\mathbf{q}}^{\alpha}(t)][\nu_{\mathbf{q}}^{\mathrm{LO}}(t)+1]-f_{\mathbf{k}-\mathbf{q}}^{\alpha}(t)[1-f_{\mathbf{k}}^{\alpha}(t)]\nu_{\mathbf{q}}^{\mathrm{LO}}(t)\}\delta(E_{\mathbf{k}-\mathbf{q}}^{\alpha}-E_{\mathbf{k}}^{\alpha}+\hbar\omega_{\mathrm{lo}}) , \tag{8}$$

where ω_{lo} is the dispersionless LO-phonon frequency, and M_q is the matrix element of the Fröhlich interaction.[20]

Transforming the sum into an integration, we obtain

$$\dot{\nu}_{q}^{\mathrm{LO}}(t)=\sum_{\alpha}\frac{Vm_{\alpha}^{*}}{\hbar^3\pi q}|M_q|^2[\nu_{q}^{\mathrm{LO}}(t)-(e^{\beta^{*}\hbar\omega_{\mathrm{lo}}}-1)^{-1}]\times\int_{k_1}^{k_2}k[f^{\alpha}(E_k^{\alpha},t)-f^{\alpha}(E_k^{\alpha}-\hbar\omega_{\mathrm{lo}},t)]dk , \tag{9}$$

where $k_2=\infty$, $k_1=(q/2)+(m_{\alpha}^{*}\omega_{\mathrm{lo}}/\hbar q)$, and $\beta^{*}=1/k_B T_c^{*}$.

These limits of integration determine those carriers that can interact with the LO phonons of wave vector $\mathbf{q}$. Inspection of Fig. 1 shows that there is a minimum for the k_1 parameter, hence the phonons with a wave vector in the neighborhood of this point are expected to be largely excited, i.e., there must be a large production of phonons with these wave vectors. We also note that q depends on the carrier effective mass (m_e^{*} or m_h^{*}); however, due to the smaller effective mass of the electrons, the majority of the carrier excess energy goes to them, so that there must be a large generation of LO phonons in mode $q=(2m_e^{*}\omega_{\mathrm{lo}}/\hbar)^{1/2}$.

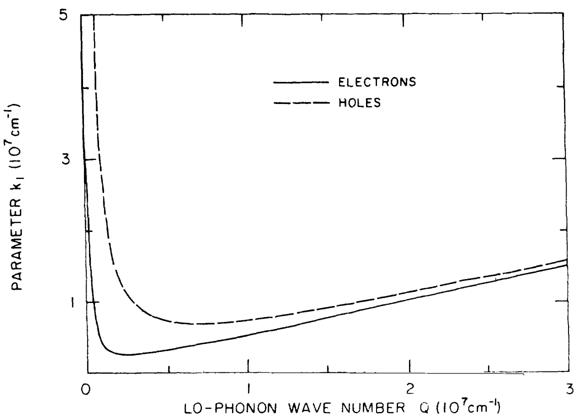

FIG. 1. Parameter k_1, in Eq. (9), as a function of q.

It should be noticed that **q** runs over the Brillouin zone in reciprocal space in a quasicontinuous manner, so to obtain the evolution of the population ν_q^{LO} for different values of wave vector **q** we resort to an approximate treatment. An appropriate partition of the q axis in a finite number of intervals is introduced, and Eq. (9) for these values of q is used, in conjunction with an interpolation procedure, to complete the whole set of populations in reciprocal space. The number of intervals is increased until a good convergence is achieved. 20 q values proved to be sufficient for the case.

III. RESULTS AND CONCLUSIONS

The solution of this system of nonlinear integrodifferential equations is obtained by resorting to a numerical method capable of solving ordinary differential equations,[21] and the results are shown in the following figures. Figure 2 shows the temporal evolution of the quasitemperatures of carriers and LO phonons, the latter defined for each mode by Eq. (2).

A quasitemperature overshoot can be observed (that is, the quasitemperature of phonons above the quasitemperature of carriers) in a small and off-centered region of the Brillouin zone. The peak value of the carriers' quasitemperature is in near accordance with the expression $\hbar\omega - E_g = 3kT_c^*$, where E_g is the band-gap energy (a result close to the one that can be derived from the equipartition of energy law). Due to collisions with LO phonons, as well as with TO and acoustic phonons, the carriers lose their excess energy and their quasitemperature decreases. As a consequence of this energy relaxation, LO phonons are produced at very large rates. It is interesting to note, still considering Fig. 2, that the LO-phonon quasitemperatures do not increase immediately after the carrier generation; the reason is that as soon as the laser pulse creates the first electron-hole pairs there is a low carrier density, and few LO phonons are produced in scattering events. The sudden rise of the phonons' quasitemperatures occurs for a carrier concentration of approximately 10^{17} cm^{-3}. After a time delay of roughly 10 ps all the modes thermalize, acquiring a single quasitemperature, because of its interactions with the carriers and the LO-phonon decay processes; at this point mutual internal thermalization of the LO phonons and the carriers has followed. For Figs. 2–6 we have taken a characteristic time of $\tau_B = 5$ ps for the LO-phonon decay to the thermal bath,[16] although it must be emphasized that this value should be a function of temperature.

Other aspects of the LO-phonon quasitemperature overshoot can be noticed if we look at their distribution in q space. Figure 3 depicts the dynamics of the phonon modes. For a time delay of -1.8 ps the phonon distribution has an equilibrium profile. It can be seen that there is a preferential region where the LO phonons are produced fast and at large rates; this region is in accordance with the q wave vector that minimizes k_1. In these curves we observe remaining peaks centered around 2×10^6 cm^{-1} (the phonons in the neighborhood of this wave number thermalize more slowly than the others); this is an indication of a phonon quasitemperature

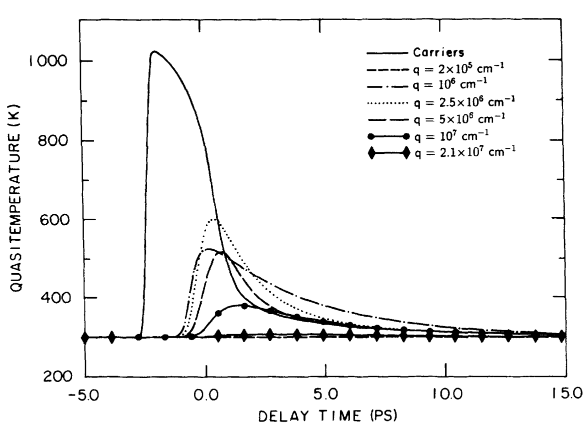

FIG. 2. Carrier quasitemperature (solid curve) and LO-phonon effective temperatures, as functions of the delay time. The phonon modes considered are indicated in the upper right corner. The LO-phonon decay time to the thermal bath is 5 ps. The laser pulse time profile is Gaussian with a length of 1 ps; the origin (0.0 ps) of the delay time stands at the peak of the pulse.

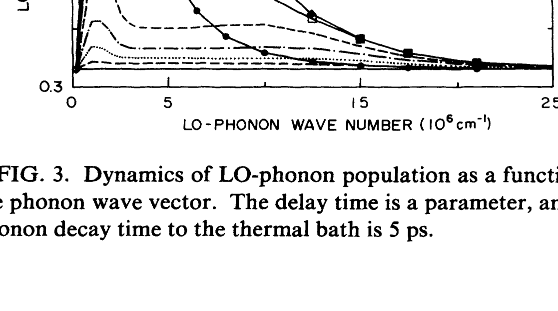

FIG. 3. Dynamics of LO-phonon population as a function of the phonon wave vector. The delay time is a parameter, and the phonon decay time to the thermal bath is 5 ps.

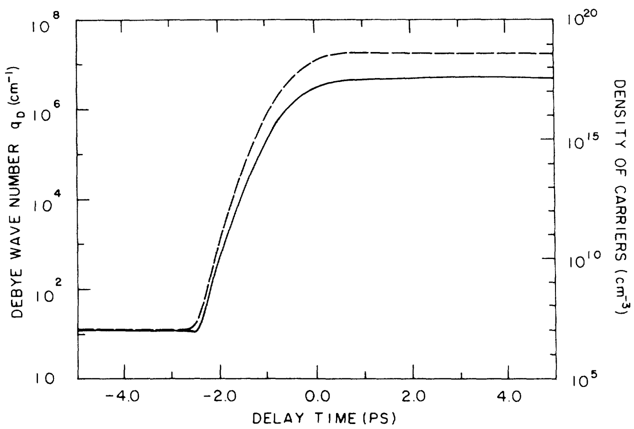

FIG. 4. Left vertical axis (solid line): Time evolution of the Debye-Hückel wave number. Right vertical axis (dashed line): Time evolution of the carrier concentration.

overshoot, as shown below. After 10 ps the distribution of phonons approaches its equilibrium profile, but with a temperature slightly larger than 300 K, corresponding, as can be seen from Fig. 2, to mutual thermalization with the carrier system, as has already been stressed.

Since under the stated conditions intervalley transitions are negligible, we ascribe the overshoot to carrier screening of the polar interaction. In fact, when Fröhlich scattering is screened, using a dielectric constant calculated in the random-phase approximation (RPA), the matrix of the interaction is corrected by the presence, in the denominator, of the square of the RPA static dielectric function, i.e.,

$$|M_q^{\mathrm{FR}}|^2 \propto \frac{1}{\left[1+\left[\dfrac{q_D}{q}\right]^2\right]^2}\,, \tag{10}$$

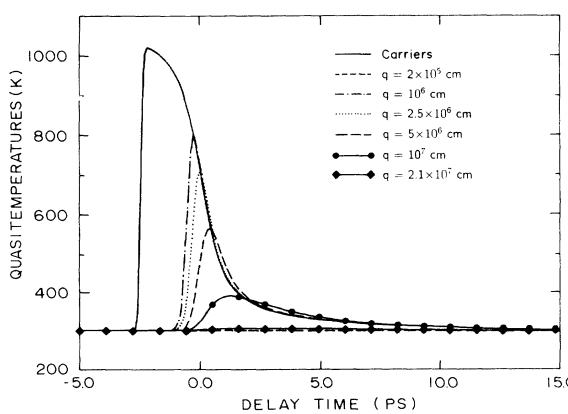

FIG. 5. Carrier quasitemperature (solid curve) and LO-phonon effective temperatures, as a function of delay time, in the absence of screening effects. The phonon modes considered are indicated in the upper right corner. The LO-phonon decay time to the thermal bath is 5 ps.

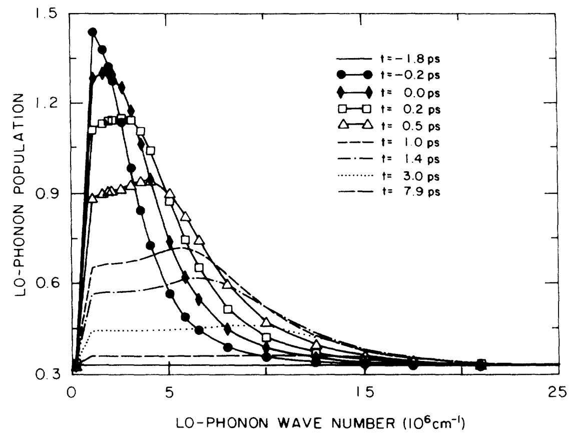

FIG. 6. Dynamics of LO-phonon population as a function of the phonon wave number, in the absence of screening effects. The time delay is a parameter, and the phonon decay time to the thermal bath is 5 ps.

where

$$q_D^2 = \frac{4\pi e^2}{\epsilon_\infty V}\sum_{\mathbf{k},\alpha}\left|\frac{df^\alpha(E_{\mathbf{k}}^\alpha)}{dE_{\mathbf{k}}^\alpha}\right|\,, \tag{11}$$

with e being the electron charge, and ϵ_∞ the high-frequency dielectric constant. Expression (11) is the so-called Debye-Hückel wave number obtained in the static limit of the Lindhard expression for the dynamic dielectric function.[22]

Since the screening is a many-body phenomenon, q_D is a function of the carrier density, as we can see in Fig. 4. With the fast enhancement of the density, q_D quickly takes values of the order of 4.5×10^6 cm^{-1}, reducing the polar optical interaction for low values of q and preventing the reabsorption of phonon energy by the carriers. As a consequence the quasitemperatures of those modes remain high and overshoot the carrier quasitemperature.

In order to test this assumption we repeat the calculation, now without screening, with the results shown in Figs. 5 and 6. Clearly, there is almost no quasitemperature overshoot in Fig. 5, and in Fig. 6 we observe no peaks of LO-phonon population in the neighborhood of 10^6 cm^{-1} after 3 ps (as can be seen in Fig. 3).

As we remarked above, the lifetime of phonons is a function of temperature. To test the sensitivity of the overshoot phenomenon upon changes in τ_B, we considered $\tau_B = 3$ ps, obtaining essentially the same results as in Figs. 2 and 3. For $\tau_B = 1$ ps the overshoot disappears, because the rate of decay of these LO phonons becomes similar to their rate of creation, and few hot LO phonons are produced.

In conclusion it has been shown that phonon quasitemperature overshoot can occur in the absence of intervalley scattering, and that, in this case, the effect is due to the screening of the polar optical interaction. Kim *et al.*[23] have considered that the majority of hot phonons accessible by Raman experiments are produced through intravalley relaxation of hot carriers; our work shows that the screening is also an important mechanism responsible for the phonon quasitemperature overshoot in GaAs, and the main one at low values of excess kinetic energy.

ACKNOWLEDGMENTS

We thank R. Luzzi, A. R. Vasconcelos, and L. H. Aciole for useful discussions, and also R. Luzzi for a critical reading of the manuscript. One of us (L.G.C.R.) acknowledges partial financial support from the Brazilian National Research Council (CNPq).

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