Ultrafast kinetics of evolution of optical phonons in a photoinjected highly excited plasma in semiconductors

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We consider the ultrafast kinetics of evolution of optical phonons in a photoinjected highly excited plasma in semiconductors. The state of the nonequilibrium ("hot") phonon system is described in terms of the concept of a nonequilibrium temperature, referred to as quasitemperature, per mode, which can be experimentally characterized and measured. The phonon emission time shows that optical phonons are preferentially produced, well in excess of equilibrium, in a reduced off-center region of the Brillouin zone. The phonons in this region are responsible for the phenomenon referred to as "hot-phonon temperature overshoot." Most of the phonons, namely, those outside such a region, are only weakly to moderately excited, and mutual thermalization of the nonequilibrium carriers and optical phonons follows, typically, in the tenfold picosecond scale. All these results are influenced by the experimental conditions, which we discuss on the basis of calculations specialized for GaAs. Comparison with experimental data is presented. [S0163-1829(96)04840-0]

I. INTRODUCTION

The photogenerated highly excited plasma in semiconductors (HEPS) has in recent decades been the object of a vast analysis on both the theoretical and observational sides—the latter carried on mainly in terms of pump-probe experiments. It ought to be noted that, besides the technological relevance associated with this line of research because of the interest in the understanding of the functioning of electronic devices, the HEPS provides an excellent testing ground for ideas and methods in many-body theory and the frontier field of non-linear nonequilibrium thermodynamics and accompanying kinetic and mechanoestatistical theories.

Here we address the particular case of so-called "hot phonons" in the plasma in semiconductors. Resorting to a treatment based on the tenets of informational irreversible thermodynamics, and the generalized nonlinear quantum transport theory and response function for systems far from equilibrium, we study the irreversible evolution of carriers and phonons in the plasma in semiconductors. The formalism, the so-called nonequilibrium statistical operator method (NESOM, for short), is reviewed in Ref. 1, where applications to the HEPS are described. It allows for a determination of the nonequilibrium temperature (more appropriately called the quasitemperature, and sometimes referred to as the effective temperature) of carriers, and the derivation of the time evolution of the nonequilibrium population of the optical phonons in the different modes, the result reported here. From the equations of evolution for the optical phonons, we derive a nonequilibrium thermodynamic variable, namely, the phonon quasitemperature per mode. The theory accounts for the phenomenon of so-called "hot-phonon temperature overshoot"; that is, along the irreversible evolution of the system the quasitemperature of some optical-phonon modes becomes larger than the carrier quasitemperature. These particular modes are presented—they belong to a small off-center region of the Brillouin zone—and we discuss how the phenomenon is influenced by the experimental conditions.

We recall that the measurement of the phonon populations can be performed via Raman scattering. Experimental evidence of the phenomenon, together with a theoretical discussion, was provided by Kim and Yu.[2]

II. NONEQUILIBRIUM PHONON QUASITEMPERATURE

Let us consider the case of a direct-gap polar semiconductor where a concentration n of electron-hole pairs (carriers) is generated by a pulse of intense laser light. On absorption of the pumping laser light, electrons make transitions from the valence band to the conduction band. They are initially narrowly distributed around a set of energy levels in a way governed by energy conservation in the absorption process. Next they are very rapidly redistributed in energy space due to the strong long-range Coulomb interaction. As a result, on the picosecond-to-femtosecond time scale, depending on the experimental conditions, the carriers attain internal thermalization.[3] The carriers then compound a double Fermi fluid whose macroscopic state can be wholly characterized by an instantaneous quasitemperature $T_c^*(t)$, and the concentration $n(t)$, or better, by the quasichemical potentials for electrons, $\mu_e(t)$, and for holes, $\mu_h(t)$. It must be stressed that we are considering a fluid of conducting carriers, and then, that the concentration of photoinjected carriers should be high enough [say, typically of the order or larger than 10^{16} cm^{-3} (Ref. 4)] in order for the excitons to be ionized and the system to be on the metallic side of Mott's transition.

On the other hand, the optical-phonon system is not internally thermalized at short-time delays after application of the laser pulse. Finally, concerning the acoustic phonons, we assume a good contact between the sample and the thermal reservoir in order to have a rapid diffusion of heat from the acoustic vibrations to the latter, such that the acoustic phonons can be considered—to a very good approximation—as remaining at the temperature T_0 of equilibrium with the reservoir.

Consequently, on the basis of these informations, following the fundamentals of the nonequilibrium statistical opera-

tor method (a detailed discussion of the different stages in the characterization of the HEPS is given in Ref. 5), for a description of the statistical thermodynamics of the system we choose a basic set of dynamical variables composed of

$$\{\hat{H}_c; \hat{N}_e; \hat{N}_h; \hat{\nu}_{\gamma\mathbf{q}}; \hat{H}_A\}. \tag{1}$$

In Eq. (1), H_c is the Hamiltonian of the carriers in Bloch bands, and with the Coulomb interaction treated in the random phase approximation; N_e and N_h are the number operators for electrons and for holes; H_A is the Hamiltonian of the acoustic phonons; and

$$\hat{\nu}_{\gamma\mathbf{q}} = a^{\dagger}_{\gamma\mathbf{q}} a_{\gamma\mathbf{q}}, \tag{2}$$

is the operator for the number of phonons in branch γ (TO or LO for transverse and longitudinal optical phonons, respectively) with wave vector $\mathbf{q}$ running over the Brillouin zone, and where, as usual $a^{\dagger}(a)$ are creation (annihilation) operators in mode $\gamma\mathbf{q}$. By

$$\{E_c(t); n(t); n(t); \nu_{\gamma\mathbf{q}}(t); E_A\}, \tag{3}$$

we indicate the basic set of macroscopic variables which characterizes the nonequilibrium thermodynamic state of the system, namely,

$$E_c(t) = \text{Tr}\{\hat{H}_c \rho(t)\}, \tag{4}$$

etc., and where $n(t)$ is the density of electrons, which is the same for holes since they are produced in pairs and we are considering an intrinsic semiconductor, and $\rho(t)$ is the statistical operator for the description of the macroscopic state of the system.[1]

In particular, the phonon population takes the form

$$\nu_{\gamma\mathbf{q}}(t) = \text{Tr}\{\hat{\nu}_{\gamma\mathbf{q}} \bar{\rho}(t,0)\} = [\exp\{F_{\gamma\mathbf{q}}(t)\} - 1]^{-1}, \tag{5}$$

where F is the time-dependent Lagrange multiplier introduced by the variational method NESOM. In the asymptotic limit, when the system attains equilibrium with a reservoir at temperature T, Eq. (6) goes over, as it should, a Planck distribution where F becomes a time-independent $F^{\text{equil}}_{\gamma\mathbf{q}} = \hbar\omega_{\gamma\mathbf{q}}/k_B T$, where $\omega_{\gamma\mathbf{q}}$ is the γ-type phonon frequency dispersion relation, and k_B the Boltzmann constant. As a consequence, a redefinition of this Lagrange multiplier written

$$F_{\gamma\mathbf{q}}(t) = \hbar\omega_{\gamma\mathbf{q}}/k_B T^*_{\gamma\mathbf{q}}(t), \tag{6}$$

is introduced, defining the so-called *γ-type phonon quasitemperature per mode* $T^*_{\gamma\mathbf{q}}(t)$. This is a measurable quantity in inelastic scattering of light experiments.[2] It is worth noting that the concepts of entropy and temperature out of equilibrium are quite difficult questions accompanied by lively controversy in the area of irreversible thermodynamics.[6]

Moreover, an alternative interpretation of the Lagrange multiplier $F_{\gamma\mathbf{q}}(t)$ can be introduced. It consists of choosing, for the description of the phonon system, the whole energy E_γ in each branch, and the number of excitations in excess of equilibrium per mode $N_{\gamma\mathbf{q}}(t)$. Next, consistently, one introduces two associated thermodynamic variables in such a way that F of Eq. (6) is replaced by

$$F'_{\gamma\mathbf{q}}(t) = \beta_0[\hbar\omega_{\gamma\mathbf{q}} - \mu_{\gamma\mathbf{q}}(t)], \tag{7}$$

where now β_0 is the reciprocal temperature and appears, differently, $\mu_{\gamma\mathbf{q}}(t)$ playing the role of quasichemical potentials per mode. This interpretation was used by Landsberg[7] for the case of photons in a nonequilibrium steady state together with a fluid of carriers, and by Fröhlich[8] for the case of polar vibrations in excited biosystems, where it may follow a phenomenon reminiscent of a Bose-Einstein condensation in nonequilibrium conditions, called Fröhlich's effect. Lauck and two of the present authors[9] considered the possibility of Fröhlich's effect in polar semiconductors, on the basis of an excitation via free-carrier absorption, showing that such an effect, however possible, cannot follow through accessible experimental conditions.

Introducing Eq. (6) into Eq. (5), the population of γ-type phonons in mode $\mathbf{q}$ takes the form

$$\nu_{\gamma\mathbf{q}}(t) = [\exp\{\hbar\omega_{\gamma\mathbf{q}}/k_B T^*_{\gamma\mathbf{q}}(t)\} - 1]^{-1}, \tag{8}$$

which resembles a Planck distribution with "temperature" $T^*_{\gamma\mathbf{q}}(t)$, which we call the quasitemperature. Equation (8), which relates the quasitemperature per mode with the population, can be inverted to obtain

$$k_B T^*_{\gamma\mathbf{q}}(t) = \hbar\omega_{\gamma\mathbf{q}} \ln\left[1 + \frac{1}{\nu_{\gamma\mathbf{q}}(t)}\right], \tag{9}$$

a result to be used later on.

This Lagrange multiplier in mechanostatistical theory, and the related macrovariable in the accompanying statistical thermodynamics, can be characterized and measured in experiments. This is done by resorting to a description of the experimental data in terms of the response function theory consistently derived within the formalism.[10] Moreover, the time evolution of these quantities is obtained from the generalized nonlinear quantum transport theory that the formalism provides.[11,12] For the case of the HEPS we have recently reviewed the derivation of these kinetic equations,[1] and therefore we omit the details for the sake of brevity. Below we only rewrite the equation for the phonon populations, to be related to the quasitemperature per mode via Eq. (9), to facilitate the discussion of the results ensuing from its solution. It is given by

$$\frac{\partial}{\partial t}\nu_{\gamma\mathbf{q}}(t) = J^{(2)}_{\gamma\mathbf{q},c}(t) + J^{(2)}_{\gamma\mathbf{q},A_n}(t), \tag{10}$$

where the first term on the right stands for the collision operator due to the interaction with the carriers, via the deformation potential and, in the case of the LO phonons, also the Fröhlich potential [cf. Eq. (40) in Ref. 1], and the last term is the collision operator accounting for relaxation effects governed by the anharmonic interaction between phonons. We stress that while this contribution is a purely dissipative one, the first on the right is in fact a source, accounting for the transference of energy from the excited carrier system to the γ-type phonons, until mutual thermalization occurs.

Let us consider first the LO phonons, when the Fröhlich potential is the predominant interaction with the carriers. Assuming the internally thermalized carriers to be in a statistical nondegeneratelike state approximated by instantaneous

in-time Maxwell-like distribution functions characterized by the quasitemperature $T_c^*(t)$ and concentration $n(t)$, we obtain[1] that

$$J_{\mathrm{LOq},c}^{(2)}(t) = -\tau_{\mathrm{LOq}}^{-1}(t)[\nu_{\mathrm{LOq}}(t) - \bar{\nu}_{\mathrm{LOq}}(t)], \tag{11}$$

where

$$\bar{\nu}_{\mathrm{LOq}}(t) = [\exp\{\hbar\omega_0/k_B T_c^*(t)\} - 1]^{-1} \tag{12}$$

and

$$\tau_{\mathrm{LOq}}^{-1}(t) = x^{-1/2}(t)[1 - \exp\{-x^{-1}(t)\}]\sum_a (\Omega_a(t)/y_a^3)$$

$$\times \exp\{-\tfrac{1}{4}x^{-1}(t)(y_a - y_a^{-1})^2\}, \tag{13}$$

with

$$x(t) = k_B T_c^*/\hbar\omega_0, \tag{14a}$$

$$y_a = \lambda_a q, \tag{14b}$$

$$\lambda_a = (2m_a^* \omega_0/\hbar)^{-1/2}, \tag{14c}$$

$$\Omega_a(t) = n(t)\pi^{3/2}\hbar\lambda_a\gamma_a/m_a^*, \tag{14d}$$

$$\gamma_a = (e^2/\hbar)(\epsilon_\infty^{-1} - \epsilon_0^{-1})(m_a^*/2\hbar\omega_0)^{1/2}, \tag{14e}$$

where γ_a is the Fröhlich coupling constant, and m_a^* is the a-type carrier's effective mass, with $a = e$ for electrons and h for holes, and we have taken a dispersionless frequency ω_0.

Equation (11), in view of Eq. (8), clearly tells us that the phonons in the given mode receive energy from the carrier system as long as $T_{\mathrm{LOq}}^* < T_c^*$, and that mutual equilibrium of carriers and phonons follows when the phonon quasitemperature in all modes coincides with the carrier quasitemperature. Hence, since at the start the phonons are in equilibrium while the carriers attain a large excess of energy from the laser pulse, the phonons are subsequently warmed up at a rate given by Eq. (13), which, it can be noticed, depends on the instantaneous nonequilibrium state of the system. This warming up of the phonons is a selective process depending on each mode: According to Eq. (13) the rate of warming up depends on the phonon wavelength $\lambda = 2\pi/q$ in the form $(\lambda/\lambda_a)^3$ times an exponential function depending on λ/λ_a and its reciprocal. Moreover, it depends on the nonequilibrium state of the system via the carrier quasitemperature and concentration. In Fig. 1 we show the dependence of the LO-phonon emission time on the wave number for several values of the carrier quasitemperature. It should be noted that it is given in terms of reduced variables, and then universal curves are valid for any polar semiconductor.

Inspection of Fig. 1 tells us that the phonon emission time diminishes with wave number as the latter keeps increasing from its zero value at the zone center, until a minimum is reached at an off-center value of the wave number, and it next increases as the wave number runs over intermediate to large values while approaching the Brillouin-zone end. This behavior is basically governed by the exponential in Eq. (13), which is the result of the interplay of energy and momentum conservation in the scattering events. The large diminution in the rate of production of LO phonons via scat-
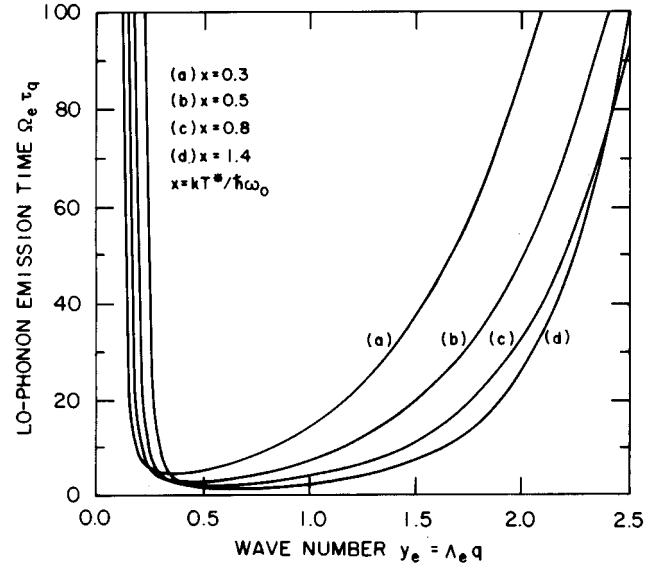

FIG. 1. Reciprocal of the rate of LO-phonon production (in units of Ω_e) due to Fröhlich interaction with electron vs wave number (in units of Λ_e), for four different values of the carrier quasitemperature (in units of $\hbar\omega_0/k$) (e.g., in GaAs $\hbar\omega_0/k \simeq 437$ K, $\Lambda_e \simeq 36$ Å).

tering with carriers at low and at large wave numbers is a consequence of, in the first case, the low density of electron states at low energies, and, in the second, the small population of electron states at high energies.

A similar behavior is obtained in the case of TO phonons when only the deformation potential is present. The expression for the TO-phonon emission time takes a form similar to that of Eq. (13), but the denominator depends only on the first power of y_a (as a result of the different dependence on the wave number of the matrix elements for Fröhlich and deformation potential). Moreover, the effect is less intense, since this channel of phonon production is less efficient than the one arising out of Fröhlich interaction.

To proceed further we introduce numerical calculations appropriate to the case of a GaAs sample, with parameters characteristics of this polar III-V semiconductor: $m_e^* = 0.067m_0$, $m_h^* = 0.5m_0$, energy gap (room temperature) 1.42 eV, $\epsilon_0 = 12.5$, $\epsilon_\infty = 11.1$, and LO-phonon frequency $\omega_0 = 5.4 \times 10^{13}$ s^{-1}. Moreover, in the calculations we write the collision operator in Eq. (10), responsible for relaxation due to anharmonic effects, in the form

$$J_{\gamma\mathrm{q},\mathrm{An}}^{(2)}(t) = -\tau_{\mathrm{An}}^{-1}[\nu_{\gamma\mathrm{q}}(t) - \nu_{\gamma\mathrm{q}}^{\mathrm{equil}}], \tag{15}$$

where τ_{An} is the relaxation time associated with the anharmonic interaction. This relaxation time is a result of all possible scattering mechanism which the LO phonons are subjected to, namely, decay into acoustic phonons, decay into TO phonons via cubic anharmonic terms but involving modes at the the zone boundary, fourth-order anharmonicities, and so on. In the calculations we use the value obtained from the linewidth of Raman bands, taken from Ref. 13. Even though this experimental value includes the effect of all possible scattering mechanisms mediated by anharmonic effects, and also the residual contribution due to scattering with impurities, predominant at low temperatures, the dependence of the temperature on the linewidth at intermediate to
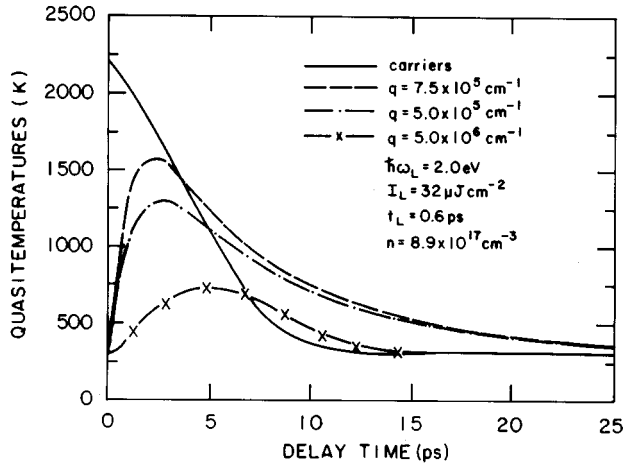
FIG. 2. Evolution of the quasitemperature of the carriers and three LO-phonon modes, for the conditions indicated in the upper right inset.

higher values of temperature (that is, comparable and large than Debye temperature in this material) indicates a predominance of decay into acoustic phonons.

III. ULTRAFAST LO-PHONON KINETICS IN GaAs

Let us consider a sample of intrinsic GaAs illuminated by a laser pulse of duration t_L, energy flux I_L, and photon frequency ω_L. As described in Sec. II, a photoinjected plasma is produced, provided the pumped energy flux is high enough to produce mobile electrons and holes (carriers); that is, the carrier system is on the metallic side of Mott's transition. The macroscopic state of this HEPS in GaAs is described in terms of the nonequilibrium thermodynamic variables of Eq. (3). For given experimental conditions their equations of evolution are computationally solved, and from them we obtain the evolution for the carrier quasitemperature and the LO-phonon quasitemperatures per mode; the former and a few of the latter are displayed in Fig. 2.

We consider an experimental situation characterized by the laser parameters $\hbar\omega_L = 2$ eV, $I_L = 32$ μJ cm^{-2}, and $t_L = 600$ fs, when the carrier concentration at the end of the pulse is $n = 8.9 \times 10^{17}$ cm^{-3}. In Fig. 2 the evolution of the carriers' quasitemperature and that of three LO-phonon modes is shown. These modes are contained in the privileged region of large production of excited phonons evidenced in Fig. 1 (in this case roughly contained in the interval $3 \times 10^5 < q < 5 \times 10^6$ cm^{-1}), and they display temperature overshoot. For the modes outside that region the overshoot phenomenon is absent and their quasitemperatures, as well as those of the modes displaying overshoot, tend to near coincidence with the value of the carrier quasitemperature after a delay time of 30–40 ps. The temperature overshoot follows for a delay time of roughly 10 ps. Additional calculations show that maintaining the given values ω_L and I_L, with a decreasing length of the pump pulse t_L, the quasitemperature overshoot occurs at earlier time delays and becomes more pronounced. We stress that it must clearly be kept in mind that only a small portion of modes (roughly in a volume one one-thousandth of the volume of the Brillouin zone) display the overshoot phenomenon. Most modes attain quasitem-
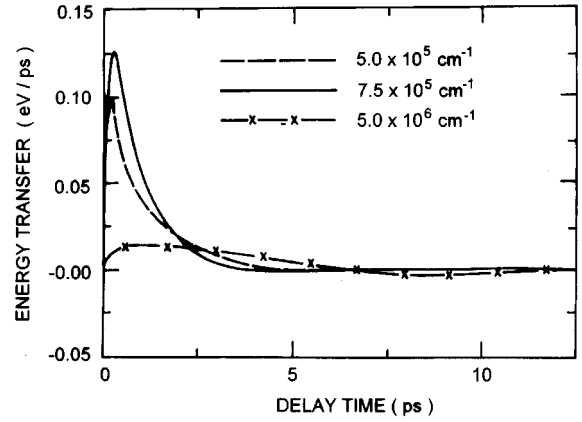

FIG. 3. Rates of energy transfer between the carrier system and LO-phonon modes, for the same situation as in Fig. 2.

peratures smaller than the carrier's quasitemperature: The carriers as a whole are always energetically more excited than the phonons as a whole, as expected.

Let us further analyze these results. For that purpose, we first consider the case in the experimental conditions that lead to the result of Fig. 2, and in Fig. 3 we show the evolution of energy transfer between the carrier system and the LO-phonon modes $q = 5 \times 10^5$, 7.5×10^5, and 5×10^6 cm^{-1}. The first two modes are contained within the region of the Brillouin zone, where there follows a large production of excited phonons [cf. Eq. (1)], and the third outside it. Figure 3 gives the clear indication that, as expected, the two privileged modes are largely excited during the first two or so picoseconds, temperature overshoot follows around a 5-ps delay time, and next they slowly return the excess energy to the carriers to attain thermalization with them in a larger-than-20-ps time scale. The mode corresponding to the wave number 5×10^6 cm^{-1} acquires energy at a rate one order of magnitude smaller than that for the other two, and the overshoot is very weak.

We stress that the optical-phonon modes that are produced well in excess of equilibrium, and that display the phenomenon of quasitemperature overshoot, are contained in a small fraction (typically one one-thousandth) of the whole extension of the Brillouin zone. The other modes over most of the zone are moderately, and most weakly, excited. Therefore, we consider, for study of the carrier system macrostate, how reasonable the approximation used by several authors maybe, of considering a unique quasitemperature for all the phonon modes; that is, taking

$$\nu_{\gamma\mathbf{q}}(t) = [\exp\{\hbar\omega_{\gamma 0}/k_B T_\gamma^*\} - 1]^{-1} \qquad (16)$$

for the dispersionless LO- and TO-phonon populations. In the experimental conditions that led to the results shown in Fig. 2, we recalculate, resorting to the approximation of Eq. (16), the evolution of the carrier quasitemperature. Comparison of these results with those already obtained using the general expression given by Eq. (8) is shown in Fig. 4. It may be noticed that the cooling down of the carriers in curve (a), corresponding to the complete calculation also displayed in Fig. 2, follows at a slower pace than the one showed in curve (b), corresponding to a calculation using Eq. (16); that is, a unique quasitemperature for the phonons.
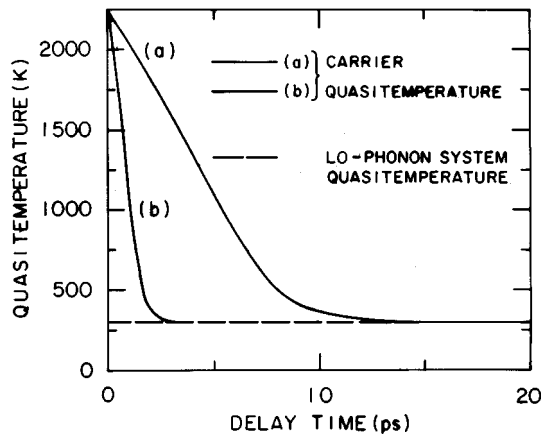
FIG. 4. Evolution of the quasitemperature of the carriers for the same situation as in Fig. 2. Curve (a), complete calculation. Curve (b), unique quasitemperature for the phonons.

The evolution of the latter is described by the dashed line, showing a small increase of roughly 1%.

Finally we proceed to compare theory and experiment with the, to our knowledge, only report with detailed experimental data, namely the one of Ref. 16. The laser intensity is 3.6 μJ cm^{-2}, the duration of the pulse is 2.5 ps, and the photon energy is 2.16 eV. From the intensity of the lines in Raman scattering we can derive the evolution of the LO-phonon population for the mode with wave number 7.7×10^5 cm^{-1}. In Fig. 5 we show the experimental data from Ref. 16—the dots—and the full line is the theoretical result. The agreement is very good, with the differences in the first stages to be ascribed to the fact that in the calculation we used a rectangular laser pulse profile, different from the peaked one in the actual experiment;[14] zero in the time axis
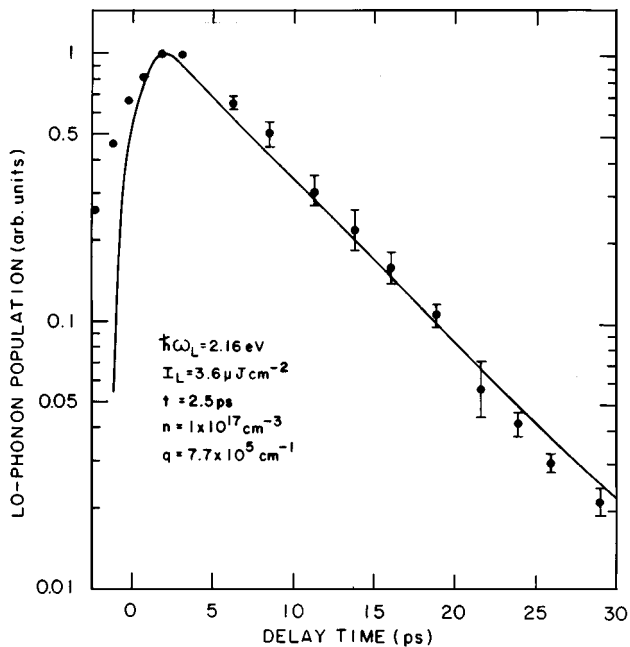

FIG. 5. Evolution of the LO-phonon population for the mode $q = 7.7 \times 10^5$ cm^{-1}. Dots are experimental points from Ref. 16, and the full line is the theoretical result. The zero in the time axis corresponds to the middle of the pulse.
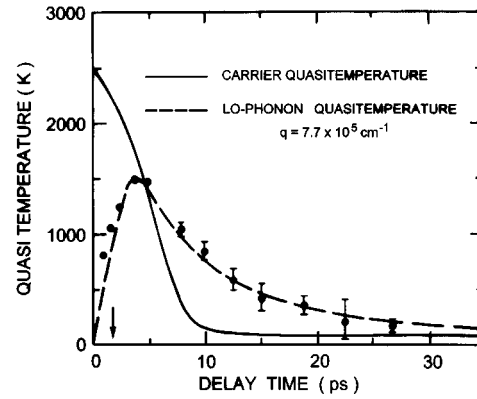

FIG. 6. Evolution of the quasitemperature of the carriers and the LO-phonon mode $q = 7.7 \times 10^5$ cm^{-1} for the conditions indicated in Fig. 5. In the calculation was used a rectangular time profile for the laser pulse starting at the origin of the horizontal axis, with duration $t_L = 2.5$ ps. The arrow indicates the positioning of the middle (peak) of the Gaussian-like experimental time profile of the laser pulse. Dots are values derived from the experimental data shown in Fig. 5.

corresponds to the middle of the pulse. Finally, in Fig. 6 we show how proceeds the evolution of the carrier quasitemperature and the quasitemperature of the given mode. Phonon quasitemperature overshoot follows around 5 ps after initiation of the pulse, and near mutual thermalization follows in roughly 40 ps.

IV. DISCUSSION AND CONCLUDING REMARKS

We have presented a study of the evolution of the nonequilibrium macroscopic state of optical phonons in a highly excited plasma in semiconductors, expected in pump-probe experiments in ultrafast laser spectroscopy. We have resorted to the seemingly powerful, concise, and practical formalism in informational statistical thermodynamics, referred to Sec. I. The general theory appropriate to deal with the photoinjected plasma in semiconductors was briefly described in Sec. II. This general theory was applied to the case of GaAs samples, with numerical results derived for different possible experimental conditions, as reported in Sec. III. When possible, comparison with experimental results were performed. On the basis of the results thus obtained, we can summarize the main conclusions to be drawn as follows:

(i) Hot-phonon quasitemperature overshoot follows under general conditions of photoexcitation, but it involves only a small fraction of the phonon modes, namely, those that are contained in a reduced off-center region of Brilouin zone.

(ii) A large majority of phonon modes present only a low-to-moderate increase in quasitemperature above the equilibrium temperature.

(iii) Mutual thermalization of phonon modes with the carrier system (i.e., equalization of the quasitemperature of all phonon modes with the carrier quasitemperature) follows in the tenfold picosecond scale.

(iv) The hot-phonon temperature overshoot associated with the privileged modes follows for delay times on the picosecond time scale, with an earlier and earlier occurrence of the phenomenon with shorter and shorter laser pulse durations.

(v) At high laser intensities, producing a carrier photoinjected concentration near the optical saturation value of nearly 2×10^{19} cm^{-1}, the overshoot phenomenon is very weak. (This is an additional result of our calculations, not presented in previous sections.)

(vi) Although the phonon modes involved in the temperature overshoot phenomenon are contained in a small region of the Brillouin zone, they affect the rate of cooling of the carrier system in the earlier stages (less than 10 ps) of evolution.

As already stated, these are the main findings that can be listed as derived from the study here presented.

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