Ultrafast Phenomena in the Photoinjected Plasma in Semiconductors

Antonio C. S. Algarte, Aurea R. Vasconcellos, and Roberto Luzzi
Instituto de Física "Gleb Wataghin", Universidade Estadual de Campinas, Unicamp
13083-970 Campinas, São Paulo, Brazil

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We present general considerations and a summarized discussion of the question of the very rapid relaxation processes that follow in the photoinjected plasma in semiconductors, phenomena that can be experimentally studied via ultrafast laser spectroscopy.

I. Introduction

Studies of the optical and transport properties of semiconductors under high levels of excitation have shown a pleiad of novel and quite interesting features evidenced in ultrafast laser spectroscopy (UFLS). This powerful experimental technique involves the interaction of matter with radiation, which is one of the most studied areas of physics and has played a decisive role in the development of modern physics at the beginning of this century. However, until the second half of this century, all processes investigated have been associated with weak radiation fields for which the usual perturbation theory, and the accompanying linear response theory near equilibrium, is applicable. Although this approach has been remarkably successful in explaining a great variety of phenomena, in the last decades - and in the present one approaching the end of the twentieth century - the new and greatly improved technological situation involving the advent of lasers, providing us with sources of intense electromagnetic radiation, requires new and sophisticated theoretical approaches, that is, a response theory capable to deal with arbitrarily far-from-equilibrium systems. Moreover, the notable improvements in time-resolved laser spectroscopy have made it a very useful tool to be used with a high degree of confidence in the investigation of very rapid microscopic mechanisms in the physical and biological realms (see for example Refs [1] to [5] and, evidently, the other articles in this special Section of the Braz. J. Phys.).

In particular, ultrafast responses and functioning under far-from-equilibrium conditions in semiconductor systems pose new, interesting, and quite engaging problems in the physics of condensed matter. These systems, as we have already emphasized in several occasions, become an extremely useful testing ground for theoretical ideas in the domain of nonequilibrium statistical thermodynamics of many-body systems. Besides the interest in the comprehension of the basic physical principles underlying these significant situations, there exists a parallel relevant technological interest arising out of the fact that semiconductors working in nonequilibrium conditions have multiple practical applications in electronic devices. Picosecond and femtosecond laser spectroscopy allows to probe ultrafast nonlinear irreversible processes in matter, thus providing an extremely adequate and sophisticated experimental instrument for the study of the nonequilibrium thermodynamic evolution of highly excited semiconductor samples.

The theories appropriate for the treatement of these far-from-equilibrium many-body systems ought to make it possible to determine the detailed time evolution of the nonlinear irreversible processes that take place in the system while it is probed. This is a quite attractive and actual problem connected with the nonequilibrium nonlinear statistical mechanics and thermodynamics of dynamical processes. UFLS studies of the highly photoexcited plasma in semiconductors (HEPS, which consists of electron and hole pairs - as mobile carriers - created by an intense laser pulse which are moving in the background of lattice vibrations) have received particular attention along the last decades. These studies provide information on the kinetic of relaxation of the photoexcited carriers and of the nonequilibrium phonon field, as well as on ultrafast

transient transport. We present in this article a brief description of some aspects of the ultrafast phenomena in HESP that can be experimentally stdied by means of UFLS. In next section is presented a tentatively clarifying discussion of topics which have been reviewed with some detail in Refs. [6] to [9].

Before proceeding further, and closing this short Introduction, we recall our previous statement that we are dealing with open systems in far-from-equilibrium conditions, thus requiring a theoretical approach based on a nonlinear nonequilibrium thermodynamics, and an accompaning statistical mechanics, able to account for the rapid time evolution of the dissipative processes that unfold in these systems. In this context it is quite appropriate to quote Ryogo Kubo's statement in the oppening address of the Oji Seminar[10]: "statistical mechanics of nonlinear nonequilibrium phenomena is just in its infancy [and] further progress can only be hoped by close cooperation with experiment". UFLS is a precious tool to carry on Kubo's advice, and we may add that since 1978, the year the above sentence was expressed, a good deal of progress have been attained in the development of nonequilibrium statistical mechanics and irreversible thermodynamics. Concerning the first one we may mention the construction of a generalized Gibbs - Boltzmann - style algorithm in the context of Jaynes' Predictive Statistical Mechanics[11], the so-called Nonequilibrium Statistical Operator Method (NESOM), reviewed in Refs [12] and [13]. On the other hand, irreversible thermodynamics is dealt with at the phenomenological level in the framework of several approaches, a quite promissing one consisting in the so-called Extended Irreversible Thermodynamics (see for exemple Refs. [14] and [15]), and its microscopic foundation at the mechano-statistical level, namely the so-called Informational Statistical Thermodynamics (sometimes refered to as Information - theoretic Thermodynamics, briefly and partially reviewed in Refs [16] and [17], with applications of it in semiconductor physics reported in Refs [18] to [23]).

II. Ultrafast relaxation kinetics in heps

Let us now consider explicitly the case of highly excited plasma in semiconductors. These are quite interesting physical systems, among other reasons, because of the flexibility in the choice of a number of parameters such as Fermi energy, plasma frequency, energy dispersion relations, cyclotron frequency, different types of carriers and effective masses, etc.[24]. In this plasma in solid state the presence of the lattice introduces noticeable differences in comparison with a gaseous plasma. Typically it produces a background dielectric constant of the order of 10, and exciton effective masses one tenth the value of the free electron mass. Thus, the characteristic atomic units for length and energy, the Bohr radius and the Rydberg, become the excitonic radius, r_x, and excitonic Rydberg, R_y^x which are roughly 100 times larger and a hundreth times smaller than the corresponding atomic units respectively. Hence, the so-called metallic densities, namely, situations when the intercarrier spacing, r_s, measured in units of r_x, is in the range 1 to 5 [25 - 27], arise at quite accessible laboratory conditions for concentrations of roughly 10^{16} carriers per cm^3 and up.

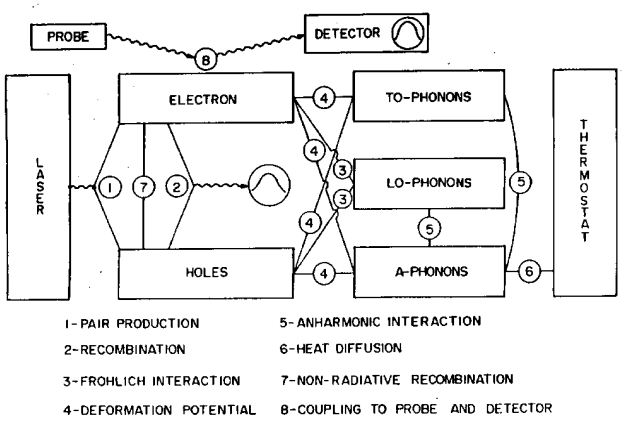

Figure 1. Schematic description of a pump-probe experiment in UFLS performed on a semiconductor sample. Several channels involving energy transfer are indicated.

In Fig. 1 we depict the situation to be expected in a typical pump-probe experiment. It describes a sample consisting of a direct-gap polar semiconductor where a concentration n of electron-hole pairs is generated by a pulse of intense laser light. Direct absorption of one photon occurs if $\hbar\omega_L > E_G$ where ω_L is the laser frequency and E_G the semiconductor energy gap. Excitation by means of nonlinear effects such as two-photon absorption or second harmonic generation, ($\hbar\omega_L < E_G$ but $2\hbar\omega_L > E_G$) allows for bulk excitation with a good degree of homogeneity. The sample is illuminated by a second laser (probe) of weak intensity, so as to avoid any noticeable modification of the nonequilibrium state of the system produced by the intense pulse from the pumping laser, and an optical response is recorded. Measurements of luminescence do not require a laser probe.

On absorption of the pumped laser light electrons make transitions from the valence band to the conduction band. These carriers (electrons and holes), with a concentration n, are initially narrowly distributed around the energy levels centered on, say, ϵ^e in the conduction band and ϵ^h in the valence band, with $\epsilon^e - \epsilon^h \simeq \hbar\omega_L$ (or $2\hbar\omega_L$). Next they are rapidly redistributed in energy space due to the strong long-range Coulomb interaction among them[28,29]. Fig. 2 provides a description of the nonequilibrium phase diagram of a photoexcited semiconductor.

Once the experiment is set, as described by Fig. 1, the connection with theory proceeds via a response function theory. For systems slightly deviated from equilibrium exact close expressions for their response functions to mechanical perturbations can be obtained in the form of correlation functions in equilibrium[30]. A practical way to calculate them is the double-time thermodynamic Green function formalism of Bogoliubov and Tyablikov, described in an already classic paper by Zubarev[31]. The actual calculation may be difficult for the case of interacting many-body systems but it is formally closed at this level. However, measurements can be performed on systems strongly departed from equilibrium, when the responses of the systems depend on their instantaneous and local nonequilibrium state, as it is the case of highly excited semiconductors when spectra obtained by means of ultrafast laser spectroscopy depend on the characteristics of the nonequilibrium distributions of the elementary excitations during the lapse of instrumental-resolution time.

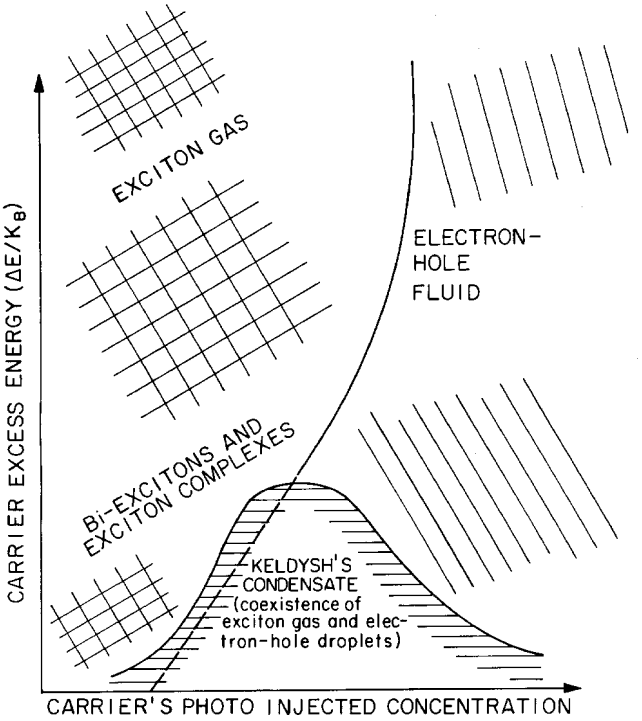

Figure 2. Schematic description of the nonequilibrium phase diagram of the carrier system in a photoexcited semiconductor. In the ordinate, the effective temperature amounts to a measure of the kinetic energy in excess of equilibrium pumped by the laser source.

Hence, the traditional response function theory needs be replaced by another capable to incorporate these features. A natural generalization rests on the use of scattering theory in conjunction with the nonequilibrium statistical operator method as described in [12].

The calculation is not difficult, and for scattering event, involving an energy transfer $\hbar\omega$, the rate of transition probability at time t is given by[12]

$$\mathcal{W}(\omega|t) = \frac{1}{\hbar^2}\int_{-\infty}^{t} dt' e^{-i\omega(t'-t)} Tr\{R^{\dagger}(t'-t)R(0)\rho_{\epsilon}(t) + C.C.\,, \tag{1}$$

where we have assumed adiabatic application of the perturbation in the remote past and $\rho_{\epsilon}(t)$ is the statistical distribution (nonequilibrium statistical operator) in Zubarev's approach to the NESOM[32-34]. Near equilibrium conditions, when $\rho_{\epsilon}(t)$ can be taken as the canonical distribution, eq. (1) goes over the well known results for the temperature-dependent rate of transition probability[35,36]. Moreover, $R(\tau)$ is the scattering operator defined by

$$R(\tau) = \bar{V}(\tau)\left[1 + \frac{1}{i\hbar}\int_{-\infty}^{\tau} dt' R(t') e^{-i\omega t'}\right]\,, \tag{2}$$

where $\bar{V}(\tau)$ is the interaction potential between the system and the probe, and all operators are given in the interaction representation. It may be noticed that for a practical way to calculate the correlation function in eq. (1) one may resort to the use of nonequilibrium thermodynamic retarded Green functions[12,37].

Differently to the case of experiments performed near equilibrium, eq. (1) is not closed in itself but is coupled to the set of nonlinear transport equations which describe the nonequilibrium macroscopic state of the system, which are of the form

$$\frac{d}{dt}Q_j(t) = \sum_{k=0}^{\infty} J_j^{(k)}(t) \ . \qquad (3)$$

Here $Q_1(t), Q_2(t), ..., Q_n(t)$ are the set of n variables which describe the macroscopic state of the system on the time-scale determined by the experimental conditions. The right hand side of eq. (3) consists into an expansion in a series of collision operators of increasing power in the interaction strengths as described in Ref. [38].

Once a HEPS has been created, to describe its macroscopic state it is a prerequisite to define a basis set of macrovariables, the $Q_j(t)$ in eq. (3). As a general rule, according to the method, one chooses a set of variables to which one has direct or indirect access in the measurement procedure restricted by the experimental set up. In Fig. 1 the main energy relaxation channels between the sample subsystems and between these and external reservoirs (thermostat and pumping laser) are indicated. Since the recorded spectra is averaged over the finite volume observed by the measuring apparatus there is no experimental access to the local hydrodynamic properties of the HEPS. The characterization of the macroscopic state of the system is a crucial step in the theory. This question has been discussed elsewhere[12,16,39], and, for the particular case of the HEPS in Ref. [40]. Fig. 3 reproduces the chain of successive contracted descriptions appropriate for HEPS, and in what follows we concentrate the attention in the so-called *second kinetic stage*, the one present in typical pump-probe experiments, which we proceed to describe.

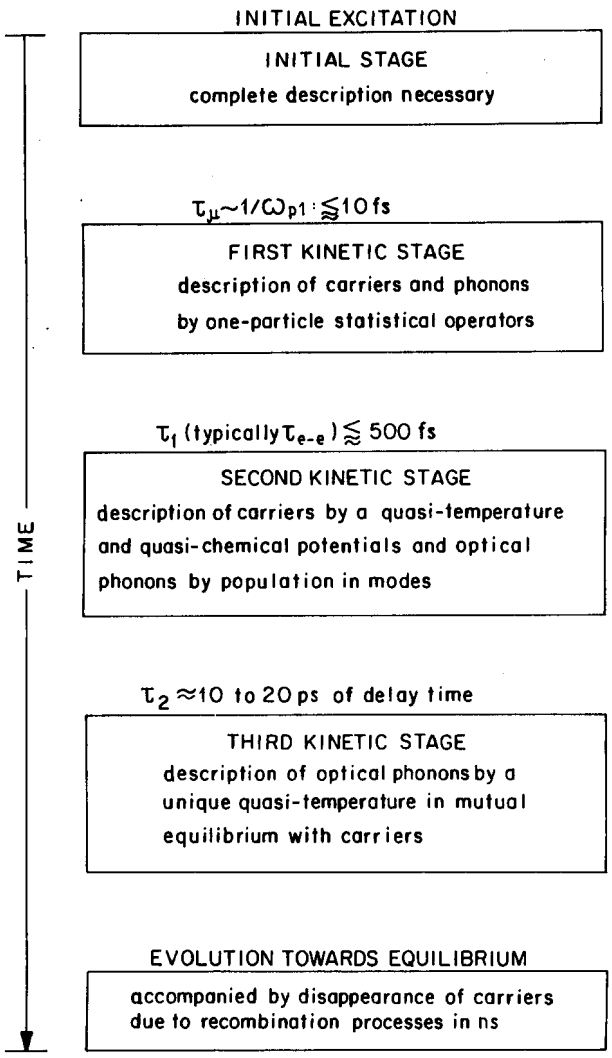

Figure 3. Schematic description of the successive kinetic stages in HEPS along its evolution in a pump-probe experiment (After Ref. [40]).

In this second kinetic stage (arising roughly in the pico- to sub-pico-second time scale after creation of the pairs) the energy and momentum in excess of equilibrium received by the carriers are redistributed among these carriers due to, mainly, carrier-carrier scattering - that is, the strong and long-range Coulomb interaction. As a consequence, in such time scale, there follows internal thermalization of the carrier system, whose statistical thermodynamic state can then be characterized by instantaneous distributions functions of the Fermi - Dirac type (which, however, can be very well approximated in the usual experimental conditions by a semiclassical Maxwell - Boltzmann - type one), depending on a nonequilibrium temperature - to be called quasitemperature - $T_c^*(t)$, and quasichemical potentials

$\mu_e(t)$ and $\mu_h(t)$ for electrons and holes respectively. Some additional comments are worth at this point. As described in Fig. 3 for ultrashort delay times the carrier system is to be described in terms of the nonequilibrium distribution function in energy space. The more mobile electrons acquire internal thermalization in the subpicosecond scale as shown in Refs [28] and [29], and what can also be estimated from the following expression for the electron - electron time between collisions[26]

$$\frac{1}{\tau_{ee}} = \frac{80}{r_s^3}(m^*/\epsilon_0^2)(T^*/T_F)^2 \times 10^{15} sec^{-1} , \qquad (4)$$

once it is adjudicated to the carriers an effective temperature T^* to measure their kinetic energy in excess of equilibrium. In eq. (4), r_s is the carrier spacing in units of the excitonic radius, T_F the Fermi temperature, m^* the effective mass in units of the free-electron mass, and ϵ_0 the static background dielectric constant. The internal thermalization of the more massive holes, and of them with electrons, follows in times which are a few multiples of the electron - electron time between collisions. For, say, $T_e^* \sim$ 1000K, $T_h^* \sim$ 400K, and $n \sim 10^{18}\text{cm}^{-3}$, we can estimate that $\tau_{ee} \sim 0.1$ ps, and $\tau_{hh} \sim 0.5$ ps, while for smaller excitation energy, say $T_e^* \sim$400K and $T_h^* \sim$ 200K, it follows that $\tau_{ee} \sim 0.5$ps and $\tau_{hh} \sim 2$ ps. We recall that the second kinetic stage corresponds to mutual themalization of both types of carriers at a unique quasitemperature $T_c^*(t)$, which, as time passes, keeps decreasing due to the rapid process in which the carriers lose large part of their excess kinetic energy due to optical - phonon emission. This decrease in T_c proceeds until optical phonons are no longer efficient at removing the carrier's excess energy, and the system enters the *third kinetic stage.*

It is relevant to notice that in the second kinetic stage, differently to the case of the carriers, the nonequilibrium statistical thermodynamic state of the optical phonons is described in terms of the phonon populations per mode, $\nu_{\vec{q}}(t)$. The kinetic of relaxation of the optical phonons shows interesting features. First, the optical phonons acquire populations in excess of equilibrium which are strongly peaked in a restricted off-center portion of the Brillouin zone, what is a result of the priviledged production of phonons in that region, as shown in Fig. 4. This particular characteristic leads to the phenomenon dubbed as "hot phonon temperature overshoot", namely, that the quasitemperatures adjudicated to each of a particular set of excited modes become larger than the carrier quasitemperature[41,42].

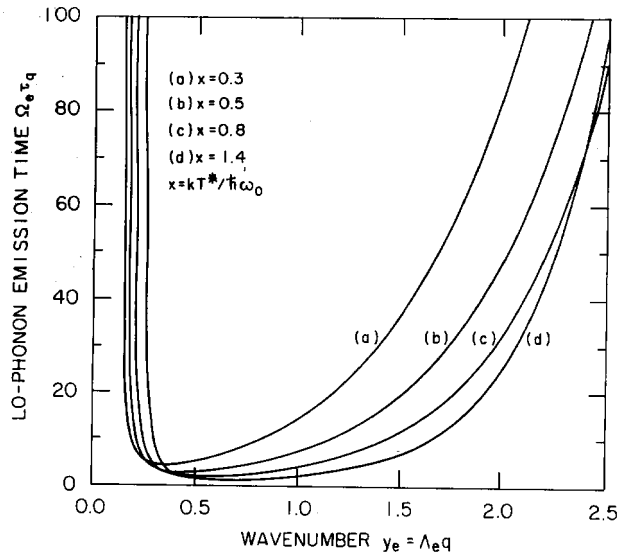

Figure 4. Reciprocal of the rate of LO-phonon production due to scattering with carriers via Fröhlich interaction as a function of phonon wavenumber, for direct gap polar semiconductors. All quantities are in reduced units, ω_α and Γ_α ($\alpha = e$ or h): For GaAs, $\Gamma_e \simeq 36\text{Å}$, $\hbar\omega_0/k_B \simeq 429$K (After Ref. [9]).

After the mutual thermalization of carriers and optical phonons in the ten-fold picosecond scale (entering the third kinetic stage), both carriers and optical phonons keep cooling througll emission of acoustic phonons, followed by heat diffusion from the latter to the thermal reservoir. It should be noticed that, once it has been set up a good thermal contact between the sample and the external reservoir, the acoustic phonons pratically remain in near equilibrium with the latter throughout the evolution of the relaxation processes that develop in the medium. Concerning the carriers' concentration, it ought to be noticed that it keeps decreasing due to recombination (in the nanosecond time scale), and diffusion out of the active volume of the sample (in the ten-fold picosecond time scale)[43,44]. After this brief revision on the kinetics of relaxation in HEPS, we proceed to present several illustrative examples showing experimental results and the accompanying theoretical calculations performed in the framework of the NESOM.

III. Illustrative examples

We consider exclusively the case of GaAs samples, and we begin with the case of experiments measuring ultrafast luminescence spectra. Typical results are shown in Fig. 5 [45]. According to the response function theory based on the NESOM, the time-resolved luminescence spectra is given by

$$I(\omega|t) = \alpha(\omega|t)\exp\{-(\hbar\omega - E_G)/k_B T_c^*(t)\} \ , \qquad (5)$$

viz., of the form of an instantaneous Shockley - Roshbroke relation[46], where α is an instantaneous absorption coefficient. Since the latter is weakly dependent on frequency on the high frequency side of the spectrum, in this region the angular coefficient of the slope of $\ln I(\omega|t)$ provides a measurement of the carrier quasitemperature T_c^*. Clearly, as inspection of Fig. 5 reveals, while time evolves this quasitemperature keeps decreasing as expected, consequence of the dissipative relaxation processes that develop in the medium as discussed in the previous section. Therefore, we are thus provided with a "thermometric device" to measure this nonequilibrium temperature (the carriers' quasitemperature in our nomenclature). This is specifically illustrated in Fig. 6, where the dots are from the experimental data of Ref [47], and the full line is a calculation of $T_c^*(t)$ in NESOM. Agreement of experiment and theory is very good.

We stress at this point that the overall behaviour of the system is strongly dependent on the experimental conditions. For example, at high levels of photoinjected carrier concentration it has been noticed a slowing down in the cooling of these carriers, accompanied by a large than expected duration of this cooling, viz. the observation in the curve displaying the evolution of the carrier quasitemperature of a nearly plateau (with a ~100 ps duration) instead of an otherwise rapid decay. Two possible mechanisms have been suggested to explain these effects. One is the screening of the electron-phonon interaction, and the other the already referred to phenomenon of "hot phonon temperature overshoot", that is, a one associated to the kinetic that governs the evolution of the distribution of phonons, which enhances phonon reabsorption by the carrier system thus reducing the rate of cooling of the latter[48,49]. It has been shown that both effects have in fact an influence of the kind, but that a third one also contributes for such slowing down in the carriers' cooling procedure, namely the influence of diffusion of carriers out of the active volume of the sample[43,44]. This is illustrated in Fig. 7, where dots are experimental points provided by the work of Ref. [50] and the full curve is a calculation in NESOM. Fig. 8 shows the evolution of the carriers' density clearly showing the influence of diffusion, which is the main responsible for the decrease in concentration at these short delay times when the recombination effect is weak.

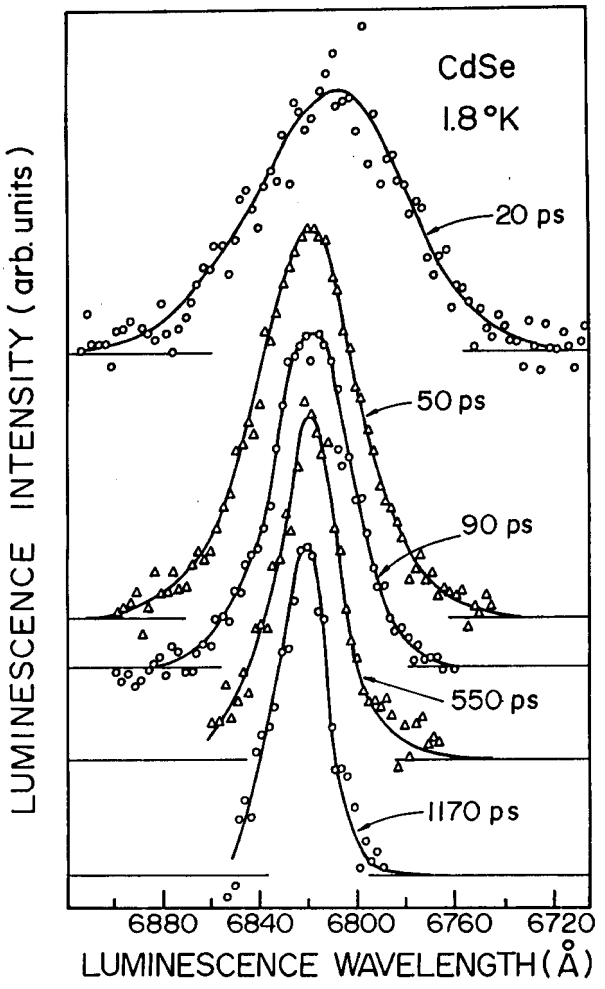

Figure 5. Time resolved luminescence spectra in CdSe (After Ref. [45]).

This question of diffusion belong to the field of the thermo-hydrodynamics of HEPS. We do not pursue this topic further in this communication, and simply refer the reader that may be interested in the subject, to

forthcoming papers dealing with propagation of matter and of energy (diffusion, second sound, etc) in the carrier system[51,52] and in the phonon field[53].

Moreover, an interesting topic is the case of a HEPS in the presence of electric fields, leading to the subject of nonlinear charge transport in these systems. A study of this question including a derivation of a generalized Einstein relation (that is, one connecting the diffusion coefficient and mobility) is presented elsewhere[54,55].

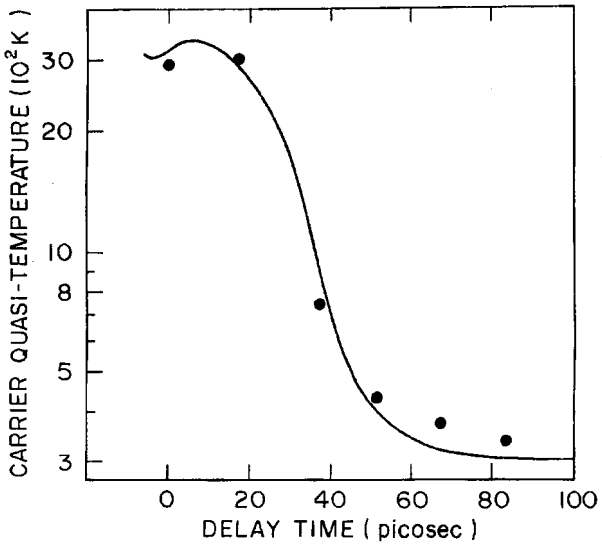

Figure 6. Evolution of the carrier quasitemperature. Dots are experimental data from Ref. [47].

Finally, we consider the kinetic of evolution of the optical-phonon system. As already noticed, the optical phonons, initialy in a equilibrium state, depart from this condition as a result of the gain of energy which they are receiving from the nonequilibrated photoinjected carriers. The energy transfer proceeds via deformation and Fröhlich potential interactions, and it has already been noted that this transfer is highly asymmetrical in the Brillouin zone, as described in Fig. 4 for the case of LO phonons when Frohlich interaction predominates. Fig. 4 tells us that the rate of production of LO phonons increases with diminishing wavenumber until a maximum is reached (a minimum of its inverse in the figure) and next it again decreases as the wavenumber approaches the zone center. Hence, as already pointed out, there is an off-center (small) region of reciprocal space where the rate of production of LO phonons in excess of equilibrium is large, whereas it is small near the zone center and for intermediate to large wavenumbers. Similar behavior is obtained in the case of TO phonons, when the deformation potential is in action.

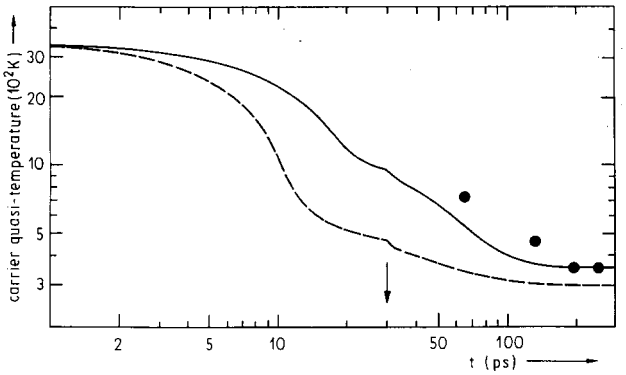

Figure 7. Evolution of the carrier quasitemperature. The upper curve is calculated with the effect of ambipolar diffusion taken into account, while in the lower curve (dashed line) this effect is neglected. The arrow indicates the end of the laser pulse, and the dots are experimental data from Ref. [50].

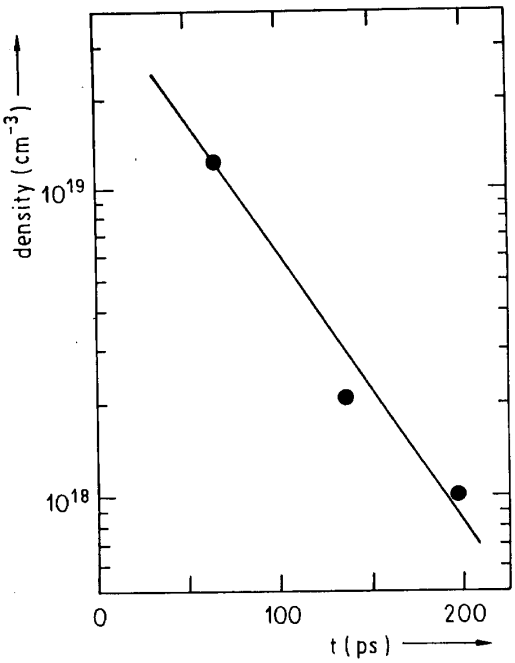

Figure 8. Evolution of the photoinjected carrier density. Dots are experimental data from Ref. [50].

The evolution in time of the phonon populations is obtained in time-resolved Raman scattering experiments, through the ratio of the intensity of anti-Stokes to Stokes bands, namely

$$\frac{I_S}{I_{AS}} = \frac{\nu_{\vec{q}}(t)+1}{\nu_{\vec{q}}(t)} \ . \qquad (6)$$

A quasitemperature for mode $T^*_{\vec{q}}(t)$ is defined by the relation

$$\nu_{\vec{q}}(t) = [\exp\{\hbar\omega_{\vec{q}}/k_B T^*_{\vec{q}}(t)\} - 1]^{-1} \qquad (7)$$

and Fig. 9 illustrates a particular case, where dots are experimental points from Ref. [56], and the full line is the corresponding calculation in NESOM. Finally, in Fig. 10, it is evidenced the phenomenon of "hot phonon temperature overshoot" in a calculation in HEPS[41], a phenomenon observed as reported in Ref. [42]. It is worth noticing that these results also shown that mutual thermalization of carriers and optical phonons follows in the ten-fold picosecond time scale, as already anticipated in an earlier section.

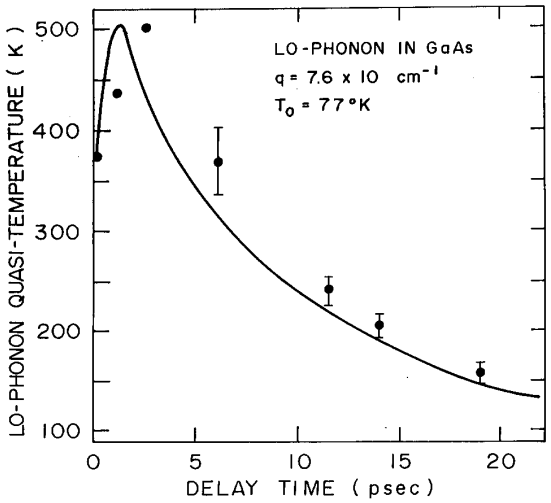

Figure 9. Evolution of the LO-phonon quasitemperature corresponding to the mode $q = 7.6 \times 10^5 \mathrm{cm}^{-1}$ in GaAs. Dots are experimental data from Ref. [56].

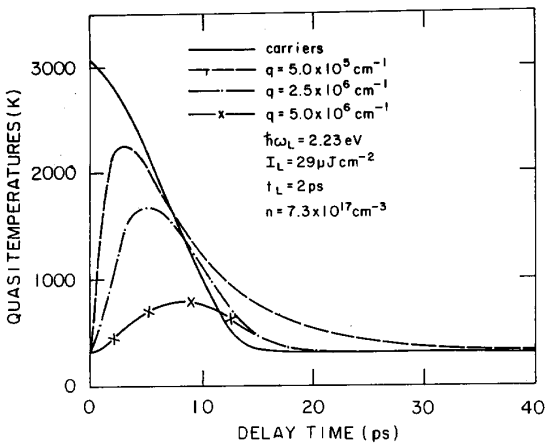

Figure 10. Evolution of the quasitemperature of several modes in GaAs, illustrating the phenomenon of "hot phonon temperature overshoot", and the mutual thermalization of carriers and phonons in the ten-fold picosecond scale (After Ref. [41]).

IV. Concluding remarks

As already stated in the Abstract, we are contributing to this special Section of the Braz. J. Phys. on Ultrafast Phenomena, with a general presentation - free of technical details which can be consulted in the references indicated in each case - partially describing the main aspects of the significant problem of ultrafast relaxation phenomena in HEPS, which can be experimentally evidenced and studied resorting to UFLS. We once again stress the quite important fact that this kind of research is of great relevance, on the one side to the technological area of semiconductor devices, and on the other for the understanding of basic concepts, and to test and apprise powerful theoretical approaches, in the physics of many-body systems in arbitrary nonequilibrium conditions and subject to nonlinear kinetic laws ror the description of their statistical thermodynamical state.

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