Thermalization and cooling processes in a dense photogenerated plasma in polar semiconductors: Effects of screening and phonon heating

M. A. Rodríguez,* J. L. Carrillo, and J. Reyes
Departamento de Física del Instituto de Ciencias, Universidad Autónoma de Puebla, Apartado Postal J-48, 72570 Puebla, Puebla, Mexico
(Received 19 August 1986)

The relaxation process of a dense carrier population created by a light pulse in a polar semiconductor is examined. We analyze the influence of screening in the electron-electron and electron-phonon interactions as well as phonon heating effects upon the thermalization process of the carrier system and the subsequent cooling process. We perform our study by incorporating screening and phonon population effects into a phenomenological model previously proposed to explain some characteristics of the steady-state luminescence of GaAs under high density of excitation. Specifically, the time required for the carrier system to thermalize, the temperature at which this occurs, and the time evolution of the carrier temperature are examined as functions of the carrier density of excitation, the energy of excitation, and the light-pulse duration. We conclude that the thermalization process is mainly affected by a long-lived phonon population created by the relaxing carriers and that phonon heating and screening effects affect the cooling process at different densities of excitation and at different time intervals.

I. INTRODUCTION

The study of the relaxation process of a photogenerated hot-carrier population in polar semiconductors has become a field of growing interest in the last few years. It is currently believed that development of picosecond and sub picosecond optical techniques allows the testing of the relative importance of scattering and relaxation mechanisms.[1-3] The majority of the published studies refer to GaAs, in bulk form or in multiple quantum wells and heterostructures, and deal with the hot-carrier kinetics[4-7] or the related photon kinetics.[8-10] There are several important studies in other related semiconductors.[8-11]

Although the dominant scattering mechanisms, namely electron-electron (e-e) and electron-longitudinal optical phonon (e-ph) collisions, and recombination (rec), have been previously established and the basic theoretical ideas involved in the process have been formulated[12] for several years, our current understanding of the evolution of a dense carrier-distribution function (CDF) after excitation is by no means complete. The major difficulty is introduced by the long-range nature of the Coulomb interaction which makes an *ab initio* calculation of the CDF and its evolution a very involved problem. In order to describe the relaxation process, it is usually necessary to assume that a certain thermalized form, either degenerated or nondegenerated, for the CDF has already been reached. Then based on this thermalized CDF, by defining several parameters associated to it, one tries to characterize a rate of relaxation. As it is obvious, this procedure discards all the information about the thermalization process.[3,13]

In this paper we study the problem in some more general terms, and by generalizing a previously proposed kinetic model to include screening in the e-e and e-ph interaction and phonon population effects, we discuss the whole relaxation process, i.e., the thermalization process and the cooling process. With respect to the thermalization process, we discuss how several external variables such as excitation density, excitation energy, duration of the light pulse, etc., influence it.

In relation to the experimental observations of the cooling process, there has been in the last few years some discussion regarding the interpretation of the possible causes of the reduction of the cooling rate present at high excitation densities.[5,6,9,14] We discuss here the influence of the screening in the e-e and e-ph interaction and phonon population effects upon these processes.

Although the predictions of the model are of qualitative nature, i.e., the trends, not the absolute values, are to be compared with experimental data or other calculations, we think it is useful to have a simple framework to analyze the evolution of a carrier population. It may be added that it is precisely the simplicity of the model which allows us to deal with the CDF at all times, even prior to thermalization.

Briefly, we outline the problem as follows. At time $t=0$ in an empty conduction band of a polar semiconductor a laser pulse of duration t_p injects n_p electrons with an energy ε_p above the bottom of the band. This initial carrier population relaxes mainly due to e-e and e-ph collisions and the total number of carriers on the conduction band decreases due to recombination.

We wish to discuss the following questions: At what time, t^*, may the CDF be considered thermalized? What is the corresponding thermalization temperature T^*? How do t^* and T^* depend on the external parameters such as n_p, t_p, ε_p, etc.? How does $T(t)$ evolve for t greater than t^*?

It has been observed experimentally that the rate of

cooling of the CDF diminishes when the excitation density increases beyond certain critical values.[2,4,5] Such an effect has been attributed to several causes. One of the most analyzed is the possible influence of screening by free carriers which diminishes the effectivity of e-ph interaction as a relaxation channel.[8,15] Another possible cause is the influence of a long-lived phonon population which would act as a thermal reservoir impeding the CDF to cool down. The detailed form in which the phonon reservoir is created and acts on the CDF has been discussed by several authors.[6,9,16,17]

The procedure by which we take up the problem is the following. We incorporate in our original model[18,19] screening and phonon population effects and investigate their consequences on the CDF evolution. We address the problem in some more general terms than other authors; namely, we are considering in our analysis screening not only in the e-ph interaction but also in the e-e interaction. We think that the thermalization time must be affected by such a screening. To our knowledge nobody has pointed out this possibility although there might be some evidence of it.[4]

The paper is organized as follows. We present in Sec. II a brief description of our phenomenological model and the extensions required to incorporate screening and phonon population effects into it. In Sec. III we discuss some physical and mathematical details and the outline of the calculation is described. In Sec. IV we present and discuss our results, firstly those related to the thermalization process and secondly, those related to the cooling process. Finally we discuss in Sec. V some of our findings and present our conclusions as well as our suggestions for further studies.

II. THEORETICAL FRAMEWORK

The original form of the phenomenological kinetic model introduced in Refs. 18 and 19 is based in the following set of coupled nonlinear rate equations which govern the time evolution of carrier and phonon population and the carrier energy flux:

$$\frac{d\eta_0}{dt}=-w\eta_0+\nu\eta_1-\mu\eta_0+\frac{ZN}{2}(\eta_1-\eta_0)+\frac{Z\eta_0}{2}\eta_0\ , \quad (1a)$$

$$\frac{d\eta_{i\neq 0}}{dt}=-w\eta_i+\nu(\eta_{i+1}-\eta_i)-\mu(\eta_i-\eta_{i-1}) +\frac{ZN}{2}(\eta_{i+1}-2\eta_i+\eta_{i-1})+\frac{Z\eta_0}{2}(\eta_i-\eta_{i-1}) +\eta_p(t)\delta_{i,i_p}\ , \quad (1b)$$

$$\frac{dE_e}{dt}=-\Delta\varepsilon[(N-\eta_0)\nu-N\mu]-wE_e+\eta_p i_p\,\Delta\varepsilon\ , \quad (1c)$$

$$\frac{dN_\Omega}{dt}[(N-\eta_0)\nu-N\mu]-\xi[N_\Omega-N_\Omega(0)]\ . \quad (1d)$$

Very briefly, the basic ideas which support the model are the following. The continuum of electronic states on the conduction band is substituted by a discrete set of equally spaced levels of energy $\varepsilon_i=i\,\Delta\varepsilon$ and we choose, in order to have a point of reference, $\Delta\varepsilon=\hbar\Omega_{\rm LO}$, the longitudinal optical phonon energy. The quantities η_i, $i=0,1,2,\ldots$, and η_p are, respectively, the carrier populations associated to each level and the rate of generation of population by the laser pulse which can depend on time (in general it can depend on energy also, although in the present work it is assumed approximately monochromatic). $N=\sum\eta_i$ is the total carrier population and δ_{i,i_p} is a Kroneker delta function, indicating here that the electrons are injected by the laser pulse at the energy level i_p. The time evolution of the CDF is thus governed by the values of the phenomenological parameters w, ν, μ, and Z_N (to abbreviate the notation we shall define $Z_N\equiv ZN/2$) which are the effective collision frequencies (ECF's) associated with recombination, phonon emission and absorption, and electron-electron collisions, respectively. The energy flux into and out of the electronic system is given by Eq. (1c). In terms of these phenomenological parameters this equation can be derived from Eqs. (1a) and (1b) taking $E_e=\Delta\varepsilon\sum_i\eta_i$.[18] Finally the coupling between electronic and lattice vibration systems is given by Eq. (1d), where N_Ω is the longitudinal optical phonon (LOP) population, and ξ is a damping parameter for the LO phonon population, associated with decay via nonelectronic collisions. This equation is derived from Eq. (1c) as discussed in Ref. 19.

A detailed discussion of the content of Eqs. (1) and the definition of the parameters may be found in Ref. 18.

We wish here only to remark that up to this point the parameters w, ν, μ, Z_N, and ξ are constants independent of carrier or phonon populations. Now we extend this model in two ways: (i) to include a LOP population dependence in the parameters ν and μ and (ii) to include screening effects through a dependence with carrier concentration of the coefficients Z_N, ν, and μ.

A. Phonon population effects

Our phenomenological approach is based on the central idea of trying to describe the evolution of the CDF by describing the macroscopic energy exchanges which appear in the system as a consequence of the different collision mechanisms; hence, it is reasonable in the steady-state case to assume (as in Ref. 18) that the phenomenological parameters ν and μ are constants. However this hypothesis requires modification in the dynamical case in which the rates of energy exchanges are not fixed.

In the absence of a sizable number of LOP's the relaxation of carriers is essentially governed by spontaneous emission of LOP's. In such a condition the effective collision frequencies ν and μ can be considered as constants and μ can be neglected compared with ν, since

$$\frac{\mu}{\nu}\sim\frac{N_\Omega}{(1+N_\Omega)}\sim e^{-\Delta\varepsilon/kT}\sim 10^{-7}$$

for $\Delta\varepsilon=38$ meV and $T=30$ K.

For high-carrier concentration, it is possible however that absorption and stimulated emission of LOP's becomes important, modifying in this way the energy exchange rates between the carrier distribution and the lattice.

In Ref. 19, we introduced for the rate of energy exchange due to e-ph processes, the first term in the right-hand side (RHS) of the quantity dE_e/dt given by Eq. (1c), and from it, we wrote the rate Eq. (1d) for the total population of LOP's. This approximation needs to be improved by giving some appropriate dependence of the phonomenological parameters ν and μ with the LOP population N_Ω. In order to do it, we consider by analogy with Eq. (1d), the rate equation governing the population P_α of LOP's in the mode α as follows

$$\frac{dP_\alpha}{dt}=-\xi[P_\alpha-P_\alpha(0)]+[(N-\eta_0)\nu_0(1+P_\alpha) -N\mu_0 P_\alpha] , \tag{2}$$

where the coefficients ξ, ν_0, and μ_0 can depend on the mode α, for example, through matrix elements. As is argued in Ref. 17, e-ph dispersion is predominant only for a small region near the Γ point in Brillouin zone. In order to keep our equations as simple as possible we shall assume that the coefficients ν_0 and μ_0 are of the form

$$\nu_0,\mu_0=\begin{cases}\text{constant for } q_\alpha \le Q_{\max}\\ 0 \text{ for } q>Q_{\max}\end{cases},$$

where q_α is the wave vector of mode α and $Q_{\max}\sim 10^7$ cm^{-1}.[17] We think these approximations are consistent with the phenomenological nature of the model even though we are aware that they might result in an underestimation of the effects of stimulated LOP emission at certain wave vector where e-ph matrix elements are strongly q-dependent.[6,17] For the total number of LOP's we write down the rate equation

$$\frac{d\mathcal{N}_\Omega(t)}{dt}=-\xi[\mathcal{N}_\Omega(t)-\mathcal{N}_\Omega(0)] +\frac{1}{U}\{[N(t)-\eta_0(t)]\nu[1+\mathcal{N}_\Omega(t)] -N(t)\mu\mathcal{N}_\Omega(t)\} , \tag{3}$$

where $N_\Omega=\sum'_\alpha P_\alpha$, $U=\sum'_\alpha(1)$, $\mathcal{N}_\Omega=N_\Omega/U$, $\nu=\nu_0 U$, and $\mu=\mu_0 U$. The primes in the summations indicate here that only accessible modes ($q_\alpha \le Q_{\max}$) are included, thus U is the number of accessible phonon modes. By comparison with Eq. (1d) we observe that in order to include a dependence on the phonon population in the parameters ν and μ we have to replace

$$\nu\to\nu[1+\mathcal{N}_\Omega(t)] ,$$
$$\mu\to\mu\mathcal{N}_\Omega(t) . \tag{4}$$

A similar reasoning can be performed with Eqs. (1a)—(1c). In this way we include the phonon population effects in our kinetic model.

Before we go on, a word is necessary in order to clarify the normalization of rate equation. It is computationally advantageous to normalize the carrier populations $\eta_i(t)$ and $N(t)$ appearing in Eqs. (1a)—(1d) in such a way that the total injected carrier density at the end of the light pulse, N_p, becomes equal to one. To do this, Eqs. (1a)—(1c) are divided by N_p while a term N_p is factored out of the square bracket in the right-hand side of Eq. (3). The ith and total normalized carrier populations are then written as fractions of the total carrier population at time $t=t_p, N_p$. It is also convenient to recall the definitions introduced in Ref. 19: $\sigma=(ZN_p/2)$ measures the strength of the unscreened e-e interaction at a given excitation density. Finally, by dividing by $\Delta\varepsilon$, Eq. (1c), it becomes a rate equation for $\varepsilon=E_e/(\Delta\varepsilon N_p)$, i.e., the carrier energy per excited electron in units of $\Delta\varepsilon$.

B. Screening effects

In the presence of a dense carrier population all the interactions of electromagnetic origin are modified due to screening effects. In our case the two most important interactions, i.e., e-e and e-ph, should be generalized to include screening effects. Consequently the effective collision frequencies ν, μ, and Z_N have to be modified. The screening depends on the total carrier concentration through the precise form of the frequency and wave-vector-dependent dielectric function which itself depends on the form of the CDF. Thus, the precise dependence of screening on the carrier concentration in an out-of-equilibrium CDF is an open problem. For our present purposes, however, it is enough to include in the model some sort of correction that simulates screening, that is, that which would produce a diminishing in the effective collision frequencies ν, μ, and Z_N at high values of carrier concentration.

1. Screening in the e-ph interaction

By following Yoffa[8] we assume the following form for the ECF for LOP emission in the presence of screening

$$\nu(N)=\bar{\nu}[1+(N/N^*)^2]^{-1} , \tag{5}$$

where $\bar{\nu}$ is the low carrier density value of $\nu(N)$ and N^* is a critical carrier concentration value at which screening effects begin to be important. In the same way, for the ECF for LOP absorption we assume the dependence

$$\mu(N)=\bar{\mu}[1+(N/N^*)^2]^{-1} . \tag{6}$$

Expressions (5) and (6) can also be justified as follows. Screening can be introduced by dividing the bare matrix element for e-e and e-ph interactions by the Thomas-Fermi (or Debye-Hückel) dielectric function $\epsilon(q)=[1+(q_d/q)^2]^{-1}$ characterized by the screening length q_d^{-1} which itself depends on the carrier density ($q_d^2\sim N$); since the ECF depends on the square of the screened matrix elements we replace the unscreened ECF for LOP emission and absorption by the screened expressions (5) and (6). From here on we shall drop the overbar in the expressions for $\nu(N)$ and $\mu(N)$.

2. Screening in the e-e interaction

There is some experimental evidence which indicates that the effectivity of e-e collisions as an energy exchange channel depends on the carrier concentration in such a way that for low concentrations, below a certain critical

value N_c which depends on the material, the effectivity increases with the carrier concentration. It reaches its maximum value at N_c and at higher concentrations the energy exchange by this mechanism tends to decrease. In order to simulate these screening effects in the e-e interaction, we assume the following dependence of the intensity of e-e interaction

$$Z_N(N)=(Z_{\max}N/N_c)(e/2)e^{-N/N_c} . \tag{7}$$

Here $Z_{\max}$ is the maximum value of the intensity of e-e interaction, which is reached at $N=N_c$. Notice that the ECF Z_N introduced in Refs. 18 and 19 is defined as a product of the intensity of interaction Z and the total carrier concentration N, thus containing already a dependence on N, however the intensity of interaction itself did not contain such dependence. Thus we incorporate screening effects in the e-e interaction by making the substitution indicated by Eq. (7).

The parameter $Z_{\max}$ and N_c can be chosen to represent conditions of low, medium, and high excitation density. Although there might be some relation between N_c and the N^* introduced in the preceding section, we keep them different to maintain the greatest generality in our calculations.

III. OUTLINE OF THE CALCULATION PROCEDURE

In what follows we shall adopt the modifications given by Eqs. (3)—(7) in the rate equations (1). Hence, these equations transform to

$$\frac{d\zeta_0}{dt}=-w\zeta_0+\nu(\mathcal{N}_\Omega,\Lambda)\zeta_1-\mu(\mathcal{N}_\Omega,\Lambda)\zeta_0+\mathcal{Z}(\Lambda)(\zeta_1-\zeta_0+\zeta_0^2) , \tag{8a}$$

$$\frac{d\zeta_{i\neq 0}}{dt}=-w\zeta_i-\nu(\mathcal{N}_\Omega,\Lambda)(\zeta_i-\zeta_{i+1})-\mu(\mathcal{N}_\Omega,\Lambda)(\zeta_i-\zeta_{i-1})+\mathcal{Z}(\Lambda)[\Lambda(\zeta_{i+1}-2\zeta_i+\zeta_{i-1})+\zeta_0(\zeta_i-\zeta_{i-1})]+\zeta_p(t)\delta_{i,i_p} , \tag{8b}$$

$$\frac{d\varepsilon}{dt}=-w\varepsilon-[(\Lambda-\zeta_0)\nu(\mathcal{N}_\Omega,\Lambda)-\Lambda\mu(\mathcal{N}_\Omega,\Lambda)]+i_p\zeta_p , \tag{8c}$$

$$\frac{d\mathcal{N}_\Omega}{dt}=-\xi[\mathcal{N}_\Omega-\mathcal{N}_\Omega(0)]+\frac{N_p}{U}[(\Lambda-\zeta_0)\nu(\mathcal{N}_\Omega,\Lambda)-\Lambda\mu(\mathcal{N}_\Omega,\Lambda)] , \tag{8d}$$

where we have defined $\zeta_i=\eta_i/N_p$, $\Lambda=N/N_p$, $\zeta_p=\eta_p/N_p$, $\nu(\mathcal{N}_\Omega,\Lambda)=\nu(1+\mathcal{N}_\Omega)/[1+(g_2\Lambda)^2]$, $\mu(\mathcal{N}_\Omega,\Lambda)=\mu\mathcal{N}_\Omega/[1+(g_2\Lambda)^2]$, $\mathcal{Z}(\Lambda)=(Z_{\max}e/2)g_1\Lambda e^{-g_1\Lambda}$, $g_1=N_p/N_c$ and $g_2=N_p/N^*$.

In order to carry out numerical calculations it is necessary to choose appropriate values for the parameters. The ECF's are linked by the relation

$$w+\nu(\mathcal{N}_\Omega,\Lambda)+\mu(\mathcal{N}_\Omega,\Lambda)+2\mathcal{Z}(\Lambda)<1 .$$

We recall here that, the relative values and not their absolute magnitudes are important, since by their definitions, the absolute magnitudes of the ECF's $w=\Delta t/\tau_{\text{rec}}$, $\nu=\Delta t/\tau_{e\text{-ph}}$, $2\mathcal{Z}=\Delta t/\tau_{e\text{-}e}$, $\xi=\Delta t/\tau_{\text{ph}}$ depend on the choice of the interval Δt (here τ_{ph} is the LOP characteristic decay time due to nonelectronic processes).

Thus we need to estimate the values of the characteristic times and critical densities, or else, obtain them from the literature.[20] We recall here that in our model we assume that the ECF's are energy-independent quantities, thus, the parameters w, ν, μ, Z, and ξ should be calculated by means of an energy average over the band of the probability per unit of time of the transition. In such a calculation one must incorporate in the respective matrix element for the transition, the information of the dynamical screening. Once the characteristic times τ_{rec}, $\tau_{e\text{-ph}}$, $\tau_{e\text{-}e}$, and τ_{ph} are evaluated, one must calculate Δt in a way consistent with the hypotheses that in this time Δt one electron in a given level can undergo transitions only to the immediate neighboring levels ($i\rightarrow i\pm 1$) and consistent with the restriction $w+\nu+\mu+2Z<1$.

For the purposes of this paper we assume that the time Δt does exist and adopt, as in Refs. 18 and 19, some characteristic times commonly accepted, i.e., 10^{-9} sec $\gtrsim\tau_{\text{rec}}\gtrsim 10^{-12}$ sec, 10^{-11} sec $\gtrsim\tau_{e\text{-ph}}\gtrsim 10^{-12}$ sec, and with this, we characterize these regimes of excitation: $2Z<\nu$ (low density), $2Z\sim\nu$ (medium density), and $2Z>\nu$ (high density).

Let us consider as an example the values $\tau_{\text{rec}}=10^{-9}$ sec, $\tau_{e\text{-ph}}=10^{-12}$ sec, and $\tau_{e\text{-}e}=2\times 10^{-12}$ sec, thus if we choose $\Delta t=10^{-13}$ sec as our unit of time we have the values $w=0.0001$, $\nu=0.1$ and $Z=0.2$ for our ECF.

We wish to remark here that in our results, presented in the next section, the time-dependent quantities are measured in Δt units and the energy-dependent quantities are measured in $\Delta\varepsilon=\hbar\Omega_{\text{LO}}$ units.

Notice that we have defined the excitation regimes paying attention to the relative importance of collision mechanisms in producing exchange of energy. It has been argued that there exists a certain carrier concentration, N_e, below (above) which the rate of energy loss by the carriers due to emission of LOP's exceeds (is less than) the rate of energy exchange among carriers due to Coulomb interaction. Shah and Leite have reported $N_e\approx 10^{14}$ cm^{-3} for GaAs.[21] In the presence of a dense plasma the relative importance of collision mechanisms might be modified by

screening effects. Yoffa[8] has estimated that the threshold value of carrier concentration, above which screening effects in the e-ph interaction become important, is $N^* \sim 5\times 10^{16}$ cm^{-3} for GaAs. It might be convenient to take $N^* = N_c$ in Eqs. (5)—(7), however, for clarity in testing out separate screening of e-e and e-ph interactions, we shall keep them different. The LOP decay time has been estimated from experimental results[3] to be of the order of $\tau_{\rm ph} \sim 5\times 10^{-12}$ sec.

In this paper we are interested in the case of a high excitation density regime where screening and phonon population effects can modify the relaxation process, for instance, changing the cooling rate of the carrier system. However, our description of relaxation process neglecting screening and phonon population effects can be formally recovered from Eqs. (8a)—(8d) by taking

$$g_1\Lambda,\ g_2\Lambda \ll 1,\ \text{but}\ \mathscr{L}(\Lambda)\rightarrow Z_N\ , \tag{9}$$

$$\mathscr{N}_\Omega \ll 1\ . \tag{10}$$

There appears in the rate equation for the phonon population (8d) the number of excited particles per accessible phonon mode, N_p/U, which multiplies the last term. In our calculations we shall assume that such a quantity is of the order of unity although we think this hypothesis might result in a quantitatively wrong estimation of phonon population effects. It is, however, the simplest way of including LOP population effects. A precise description of the factor N_p/U requires, for given conditions of excitation, specification of the number of accessible modes ($q_\alpha \le Q_{\max}$). In a forthcoming work we shall present a detailed analysis of this term.

Very briefly, the numerical procedure is as follows. Once the carrier excitation density N_p is given, one fixes the appropriate values of the ECF by a suitable choice of Δt. This time interval is also used as the time step in the numerical integration of Eqs. (8a)—(8d) performed by the simple Euler procedure

$$\zeta_i(t+1)=\zeta_i(t)+\left[\frac{d\zeta_i}{dt}\right](1)\ , \tag{11a}$$

$$\mathscr{N}_\Omega(t+1)=\mathscr{N}_\Omega(t)+\left[\frac{d\mathscr{N}_\Omega}{dt}\right](1)\ . \tag{11b}$$

Recall that t is measured in units of Δt and all the populations ζ_i are now fractions of the total population at the end of the light pulse $\Lambda(t_p)$, which is normalized to the unity. The only quantities that have remained unspecified are η_p/N_p, t_p, and ε_p, which are, respectively, the carrier density excited per time step Δt, the pulse duration, and the energy $\varepsilon_p = i_p\Delta\varepsilon$ at which the pulse is injected. Once t_p is fixed η_p/N_p is determined by $\eta_p/N_p = \Delta t/t_p$ for $0 \le t \le t_p$. (In this way t_p has to be slightly redefined so that $t_p/\Delta t$ is an integer number.) Thus, we are dealing with a square pulse, however, slightly different shapes do not affect our results. The time evolution of the initial pulse together with the LOP population per accessible mode $\mathscr{N}_\Omega$, is followed as time increases. The thermalization time t^* which separates the thermalization process from the proper cooling process, is defined by adopting the experimental criterion to determine the temperature in a typical luminescence experiment[21,22] where only the carrier population near the bottom of the conduction band is amenable to observation. We demand that the lowest part of the CDF has already become a decreasing function of energy (in our case of i) and fit an exponential, $\exp[-i/k_BT(t)]$, to the lowest four points $i=0,1,2,3$. The thermalization time t^* is defined as the time at which a temperature $T^* = T(t^*)$ can be defined. These are the definitions of $T(t)$, T^*, and t^* that shall be used in the present paper, although we wish to point out other possibilities: one of these is to make more stringent the criterion to define the temperature by demanding that the fitting should be done with a greater number of levels. It turns out that such a modification does not affect the results in a significant way. Another possibility is to relax the criterion by demanding that it should be satisfied by an average of CDF's taken over an interval of time t_m, the time of measurement which usually should be $t_m > \Delta t$. We have not explored in detail these two possibilities. However in any case, the criterion we have chosen produces a monotonously decreasing behavior of $T(t)$ for all reasonable choices of the phenomenological parameters.

IV. RESULTS AND COMMENTS

Following the excitation pulse, the collision mechanisms act upon the carrier system thermalizing the CDF. Such a condition is reached after a characteristic time (in our case called t^*). The temperature at which the electronic system thermalizes $T(t^*)=T^*$ is commonly believed[4,6] to be a measure of the excess energy in the electronic system supplied by the laser pulse, and the evolution of $T(t)$ is usually accepted[4] to allow testing of the energy exchange between the carrier system and the lattice. Firstly, we would like to analyze the thermalization process. We shall analyze it by paying attention to influences on t^* and T^* from different experimental conditions represented in our model by different sets of our phenomenological parameters, as discussed before. In Fig. 1(a) we reproduce our Fig. 1 of Ref. 19. Here we present, for the unscreened case and neglecting phonon population effects, the CDF evolution for low (a), medium (b), and high (c) excitation density. The time interval represented there varies from $t=0$ up to times greater than t^*. One of the principal features is that the thermalization time t^* decreases when the excitation density increases, indicating that the effectivity of collision mechanisms to thermalize the CDF increases with the excitation density. Figure 1(b) reproduces our Fig. 2 of Ref. 19 where the behavior of $T(t \ge t^*)$ is shown. The curve (a) corresponds to a set of parameters representative of an intermediate excitation condition in which the rate of energy exchange among carriers is comparable to the rate of energy exchange between the carrier system and the lattice vibrations. In comparison curve (b) is representative of a low density of excitation condition in which the effectivity of e-ph collisions to exchange energy is greater than that corresponding to e-e collisions. In this case the CDF thermalizes sooner to a temperature T^* lower than in case (a). A high-excitation density condition is presented in

curve (c). Notice that in this case the CDF thermalizes faster and T^* is higher than in case (a).

We wish now to discuss the trends mentioned before in some more general terms, including screening and phonon population effects. Additionally, we analyze the effect on t^* and T^* due to the values of the excitation energy ε_p and the pulse duration t_p. In Fig. 2 the behavior of T^*

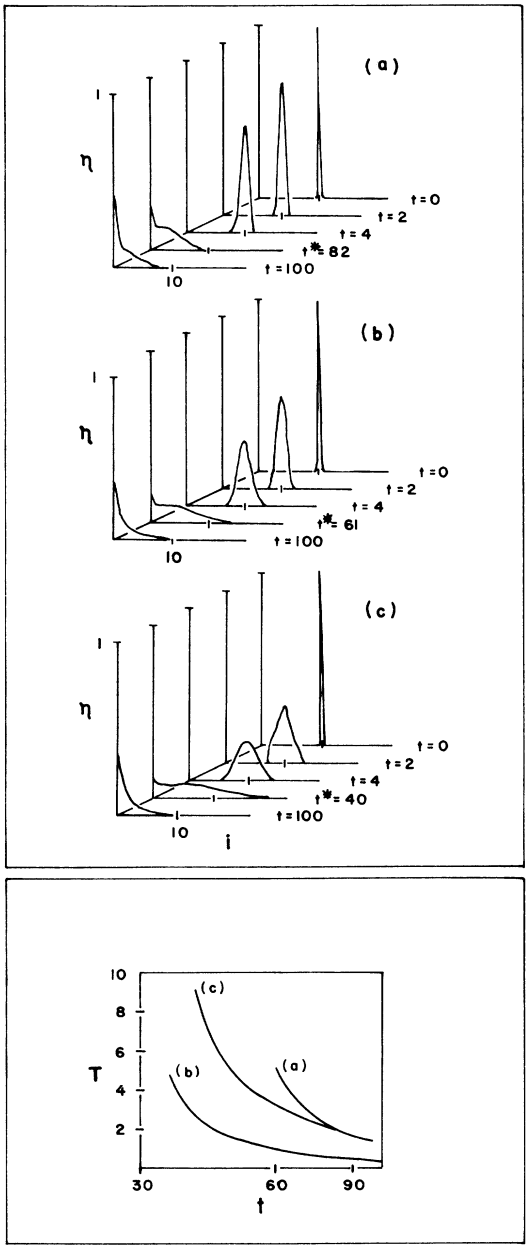

FIG. 1. (a) The time evolution of a CDF calculated with the original form of the model, Eqs. (1a) and (1b), for the cases: (a) low density of excitation ($Z_N < \nu$), (b) intermediate density of excitation ($Z_N \sim \nu$). (b) The evolution of the temperature of the CDF, $T(t > t^*)$, for the cases (a) $\nu = Z_N = 0.1$, (b) $\nu = 0.2 > Z_N = 0.1$, and (c) $\nu = 0.1 < Z_N = 0.3$. The time is measured in Δt units and the energy and Te are measured in $\Delta\varepsilon$ units. For discussion see the text.

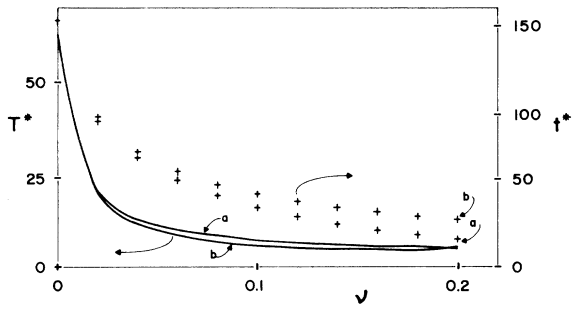

FIG. 2. Behavior of the temperature, T^* solid line, and the time, t^*, crosses, of thermalization as function of the electron-phonon effective collision frequency ν. The range of values of ν depicted covers the interval $0 \leq \nu/Z_N \leq 1.0$ thus the leftmost (rightmost) part of the graph is the region where electron-electron collisions predominate (are dominated by) electron-phonon collisions. The results obtained neglecting (including) phonon population effects are labeled by a (b.)

and t^* is presented as function of ν, the ECF associated to e-ph processes. The values of the parameters are as follows: $w = 0.0001$, $\mu = 0.0001$, $Z_{\max} = 0.4$, $i_p = 10$, $\xi = 0.3$, $t_p = 0$, $g_1 = 0$, $g_2 = 0$. This set of values shall be used in all our figures, except those which are specifically varied in each figure. Notice that an instantaneous pulse is numerically represented by $t_p = 0$. The scales for T^* and t^* are seen on left-hand and right-hand sides, respectively. In the figure, crosses represent the t^* values and the solid lines represent the T^* values. In this figure we attempt to represent qualitatively the influence on T^* and t^* for a series of experimental conditions in which the rate of energy exchange by e-ph processes varies from a condition in which it is negligible compared with the exchange by e-e collision to a condition in which the exchange by e-ph and e-e processes are comparable. The results obtained neglecting phonon population effects for t^* are the lower crosses and for T^* the upper line. Notice that the qualitative behavior for T^* and t^* is not affected by the inclusion of phonon population effects.

As we have discussed in Ref. 19, e-ph collisions contribute to the thermalization of the CDF, thus reducing t^*, however comparatively higher values of ν produce a comparatively higher rate of energy dissipation for the electronic system, reducing the thermalization temperature. In addition to phonon population effects, we wish to consider the screening of the e-ph interaction by calculating ν by the expression

$$\nu(\mathcal{N}_\Omega, \Lambda) = \nu(t) = \frac{\nu(1+\mathcal{N}_\Omega)}{1+(g_2\Lambda)^2}\,, \tag{12}$$

and choose the g_2 value in order to reproduce the same initial condition in ν as in the unscreened case. It turns out that the results for T^* and t^* are not modified by inclusion of the screening whether or not there is creation of phonons, (thus they superimpose to the plots of Fig. 2). This result is reasonable since in the time interval in which the electronic system thermalizes the total carrier concentration does not change significantly, in such a way

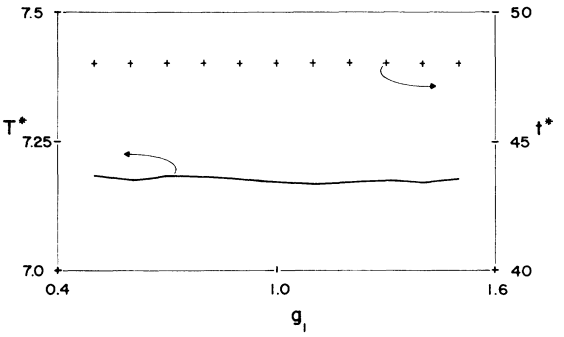

FIG. 3. Same as Fig. 2 but varying the screening parameter $g_1=N_p/N_c$, see Sec. II B, but choosing $Z_{\max}$ in Eq. (7) so that the initial conditions are always preserved.

as to produce variations in the ν value due to screening effects.

For the same reason changes in the screening parameter for the e-e interaction, g_1, does not produce significant variations on T^* and t^*, as it can be seen in Fig. 3 where the e-e ECF is calculated by Eq. (7), for the same set of parameters as in Fig. 2 and for an initial value of $\nu=0.1$. In Fig. 3 we are choosing the $Z_{\max}$ value in order to conserve the same initial value of $\mathscr{Z}(1)=0.2$ for different values of g_1, this is $Z_{\max}(g_1)=0.4\exp(g_1)/g_1$, since at $t=0$ $\Lambda=1$ (recall $t_p=0$), varying from $g_1=0.5$ up to $g_1=1.5$. The purpose of this calculation, by the use of Eq. (7), is to test the effect on T^* and t^* of various strengths of screening, represented here by g_1. Notice that the behavior of T^* (solid line) and t^* (crosses) are essentially constant indicating that in the thermalization time interval, the variation of the total carrier concentration is not enough to produce noticeable changes in the ECF $\mathscr{Z}(\Lambda)$.

If we fix in Eq. (7) the $Z_{\max}$ value and vary the strength of screening g_1, the initial value of ECF $\mathscr{Z}(1)$ changes with g_1. Thus, we are simulating different regimes of excitation density. In Fig. 4 we present the behavior of T^* and t^* for different initial values of $\mathscr{Z}(1)$ from values of carrier density of excitation representing a regime, in which the effectivity of e-e collisions to exchange energy increases with the carrier concentration, up

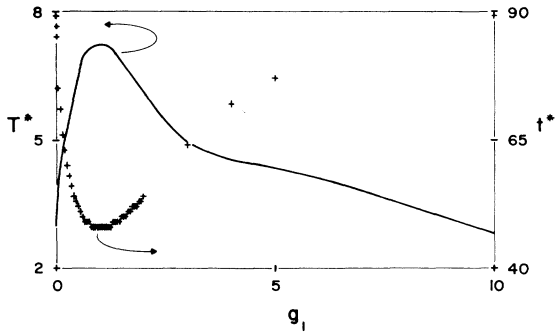

FIG. 4. Same as Fig. 3 but now $Z_{\max}$ is fixed throughout the figure. Recall that at $g_1=1$ the (e-e) collision frequency attains its maximum value. The actual results for T^* were slightly smoothed.

to values of $\mathscr{Z}(1)$ representative of an excitation regime in which it has been reduced by screening effects. (In this figure LOP effects are included but e-ph screening is neglected.) It can be observed in Fig. 4 that both T^* and t^* are strongly affected by the value of $\mathscr{Z}(1)$ and the behavior of the curves indicates the importance of e-e collisions to thermalize the CDF. We notice that t^* decreases when $\mathscr{Z}(1)$ increases until it reaches its maximum value $Z_{\max}$ at $g_1=1$. For higher carrier concentrations the effectivity of e-e collisions to thermalize the CDF diminishes, increasing consequently t^*. It has been experimentally observed that for a given observation time after the pulse injection the carrier temperature depends nonlinearly on the carrier concentration.[4,21] We observe in Fig. 4 that for relatively low density of excitation, i.e., $g_1<1$, T^* increases nonlinearly with $\mathscr{Z}(1)$. For higher carrier concentrations ($g_1>1$) T^* shows a clear tendency to decrease by effects of screening.

In Fig. 5, t^* and T^* are shown for different excitation energies, ε_p, from 0 to 15 (in units of $\Delta\varepsilon$, that is $\varepsilon_p=i_p\Delta\varepsilon$). The scale on the left-hand side corresponds to T^* and the scale for t^* is on the right-hand side. We are incorporating here all the effects mentioned before. Notice that t^* increases monotonously with ε_p, while T^* increases drastically reaching its maximum value at $\varepsilon_p=4$. (Recall that our level width is $\Delta\varepsilon=\hbar\Omega_{\mathrm{LO}}$ approximately a few tens of meV for a typical polar semiconductor. Thus excitation to the zeroth level means that the electrons are within an energy $\Delta\varepsilon$ of the bottom of the band.) This seems to indicate that there is a competition between two effects: first as ε_p is increased one expects T^* to follow it since it is a measure of the energy per particle injected into the system, second as the particles are injected higher into the conduction band the time to reach the bottom and to thermalize increases and so do the losses. Thus one expects a decrease in T^*. The maximum in the curve corresponds to the crossover. We see in the same figure that for values of excitation energy $\varepsilon_p>4$, the temperature T^* shows an obvious tendency to decrease. For $\varepsilon_p>7$, T^* diminishes in a very mild way. In Fig. 6 we present our results for t^* and T^* as functions of the duration of the light pulse, t_p, which varies from zero to ten (mea-

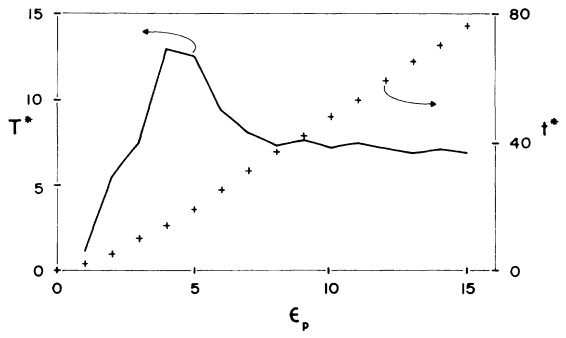

FIG. 5. The temperature and time of thermalization as a function of the energy, of excitation level ε_p. The solid line representing T^* was obtained by joining the points representing the results of the calculation by straight lines, thus explaining its broken nature.

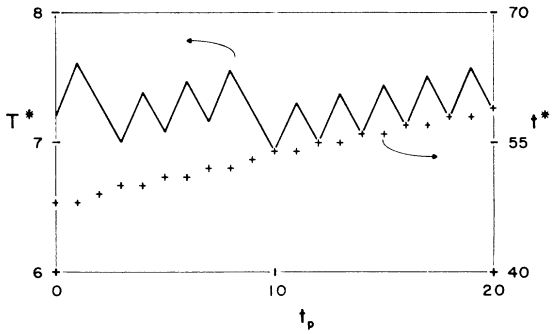

FIG. 6. Same as Fig. 5 but varying instead the duration of the exciting pulse t_p. The solid line representing T^* was obtained by joining the points representing the results of the actual calculation.

sured in units of Δt). We observe again a clear tendency of t^* to increase with t_p, although it does not present a smooth variation. Notice that T^* is strongly affected by the pulse duration, producing significant variations in the T^* values. We wish to point out here that these variations are not caused by the operational definition of T^*, i.e., if in the criterion to define T^*, we use a number of levels greater than four, one gets quantitatively a similar result. Rather, we think this is an effect attributable to the fact that the CDF is really an out-of-equilibrium distribution, as we discussed in Ref. 19 and can also be seen in Fig. 1(a), and thus there are fluctuations in the lowest level populations which produce the results shown in Fig. 6.

We have mentioned before that one of the possible factors which affect the thermalization and cooling processes is a long-lived phonon population generated by relaxing excited carriers. Physically this phonon population in excess respect to the equilibrium number results from the difference between the number of emitted phonons and the number of phonons absorbed by the carriers and by the thermal bath. In Eq. (8d), the rate of creation of phonons by the carriers appears multiplied by the factor N_p/U, and the rate of phonon population damping by the thermal bath is described by the term $-\xi[\mathcal{N}_\Omega(t)-\mathcal{N}_\Omega(0)]$. This last process is driven predominantly by the decay of LOP's into acoustical phonons. In Fig. 7 we present the influence on T^* and t^* of the effective frequency associated to LOP damping, ξ. We observe that for increasing values of ξ, which would show that the excess phonon population diminishes, both, T^* and t^* increases. This result is consistent with those discussed in Fig. 2 since, as we pointed out before, phonon population contributes to thermalizing the CDF, by changing the effective value of ν. Thus the effect of increasing ξ is analogous to that of decreasing ν. We would like to remark on this point by discussing Fig. 8(a) in which the rate of energy exchange between electronic and phonon systems is plotted. The curves (a), (b), and (c) correspond to $\xi=0.2$, 0.3, and 0.4, respectively. The arrows on the time axis indicate here the thermalization times. We observe in this figure that the maximum value of the energy exchange is reached in the system before the thermalization occurs. Up to this, time differences in the energy exchange in the three cases might be significant and its effect must be noticed in the temperature. In fact, as it can be seen in Fig. 7 and with the help of Eq. (8d), small values of ξ cause the phonon population to increase, increasing therefore the e-ph ECF and consequently increasing the rate of energy exchange by e-ph processes. This fact causes thermalization temperature to diminish in comparison with other cases for greater values of ξ. However an increment in the ν frequency implies a

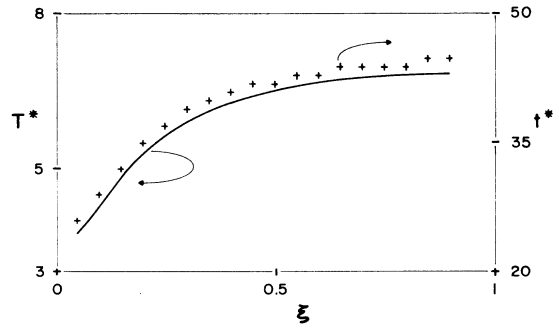

FIG. 7. Dependence with the decay effective frequency, ξ, of longitudinal optical phonons via nonelectronic channels on this temperature and time of thermalizations of the CDF. Recall that smaller values of ξ increase the peak value attained by the LOP population and thus increase the carrier energy loss due to stimulated LOP emission. The curve for T^* has been slightly smoothed.

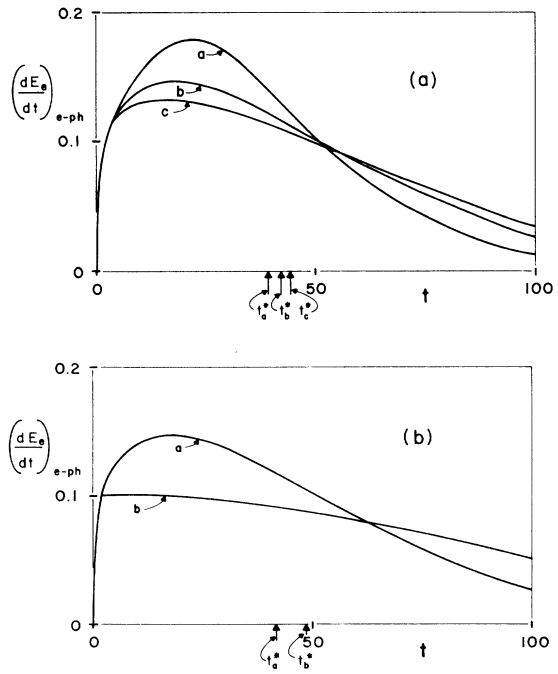

FIG. 8. The rate of energy loss by the electrons to the phonons. (a) The three curves correspond to different choices of the LOP decay frequency $\xi=0.2$, (a); $=0.3$, (b); and $=0.4$, (c). On the time axis the respective thermalization times are depicted. (b) Results obtained with $\xi=0.2$: (a), including LOP effects into the model and, (b), neglecting them.

greater effectivity of the e-ph collisions as a relaxation channel, increasing the speed at which the carriers reach the bottom of conduction band to thermalize. Thus, this results in a decrement of t^*.

In Fig. 8(b) we present a comparison of the time evolution of the rate of energy exchange by e-ph process given by Eq. (8c). The curve (a) is the same curve (a) as in Fig. 8(a) and the curve (b) is obtained by neglecting phonon population effects ($\mathcal{N}_\Omega \equiv 0$). Again here the arrows on this axis indicate the thermalization times. We observe that there is a notorious difference in the energy exchange, this difference in the case of curve (a) must produce an increment in the effective value of the parameter ν which according to the results of Fig. 2 must produce a faster thermalization process and a greater energy dissipation, consequently this reduces the values of T^* and t^* compared with the corresponding values of curve (b).

There has been in the last few years some discussion regarding the relaxation process of a dense hot electron plasma.[23] In particular it has been argued that screening by free carriers and phonon population effects might be the main cause by which the rate of cooling of the carrier system diminishes at high densities of excitation. We wish now to test how the time evolution of carrier temperature is affected by the inclusion of screening in the e-e and e-ph interaction and phonon creation.

In Fig. 9(a), we show the time evolution of carrier temperature $T(t)$ for the time interval $t^* \leq t \leq 100$ (in units of Δt). The curve (a) corresponds to the unscreened case, and neglecting phonon creation, here $Z_N = 0.2$, $\nu = 0.1$, $\mu = 0.0001$, this curve is the same as curve (a) of Fig. 1(b). In comparison curve (b) corresponds to the case in which we start with an initial value of $\nu = 0.1$ but this value changes in time due to phonon creation accordingly to the rate equation (8d). We observe that for equal initial conditions the effect of phonon population is to decrease t^* and T^* as we have discussed before. The qualitative behavior of $T(t)$ is modified by the inclusion of phonon creation effects. Such a change can be discussed based on Fig. 8(b). We observe that for times lower than 60 the energy exchange between carrier and phonon systems is greater in the case in which one is considering phonon creation. In the temperature behavior, it can be noticed in the relative lower values of $T(t)$, in addition to a decrement in T^* and t^*. For times greater than 60 in Fig. 8(b) the energy exchange of curve (b) is lower than in curve (a) indicating that phonon population at this time begins to act as a thermal reservoir diminishing the rate of cooling. In Fig. 9(a) this is noticed in the smaller slope of curve (b) compared to curve (a) for times greater than 60. If in curves (a) and (b) in Fig. 9(a), in addition to phonon population effects, we include screening in e-e and e-ph interactions, choosing the g_1, $Z_{\max}$, and g_2 values in order to reproduce the same initial conditions as in the unscreened case [that is $Z_{\max} = 2Z_N \exp(g_1)/g_1$ and $\nu = \nu^{(\mathrm{US})}(1+g_2^2)$ where (US) represents unscreened, and the reader may recall that initially $\Lambda = 1$ and $\mathcal{N}_\Omega = 0$], quantitatively the results do not change significantly as can be seen in the figure where the screened results appear superimposed to the unscreened ones.

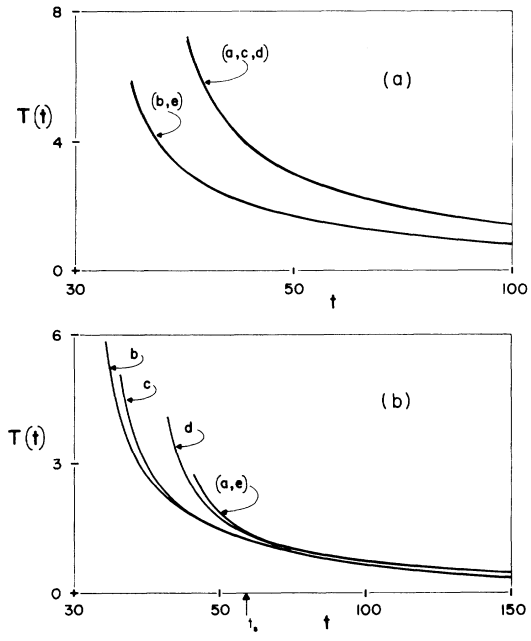

FIG. 9. The time evolutions of the temperature of the CDF for times greater than the thermalization time. (a) Comparison between results neglecting, curve labeled (a), and including LOP effects, curve labeled (b). If screening is included in such a way that the initial value of the parameter $\mathcal{Z}(\Lambda)$, see Eq. (7), is kept constant, the resulting curves (c), (d), and (e) superimpose to (a) and (b), see the text. (b) Same results obtained fixing $Z_{\max}$ in Eq. (7) and varying g_1. When observed at a fixed time, t_s, long enough so that in all cases the CDF is thermalized this plot presents the same trends observed experimentally. See the text for a fuller discussion.

Of course, in reality different densities of excitation must set a given system to different initial conditions. In order to compare for various regimes of excitation, how the competition of interactions affects the time evolution of carrier temperature, we present in Fig. 9(b), the behavior of $T(t)$ for several values of $g_1 = N_p/N_c$, representative of excitation regimes varying from low excitation density $N_p/N_c = 0.001$ to high excitation density $N_p/N_c = 10$. We observe that increasing the value of N_p/N_c first makes the curve move toward smaller t^* and higher T^*. For densities bigger than N_c (i.e., $N_p/N_c > 1.0$) the opposite is true. We would like to interpret this last behavior by arguing that if the CDF temperature is measured at a fixed time, denoted by t_s in the figure, the trends observed experimentally upon increasing the density of excitation by Shah[4] and others[2] is reproduced. (See, for example, Fig. 8 of Shah's paper, Ref. 4.) However this interpretation is tentative and we would have to wait until we have a precise definition of our units of time. Presently, work in this direction is in progress, also notice that the relative change of a given curve with respect to another one depends clearly on the time interval at which it is measured, thus speaking of a variation in the rate of cooling requires a precise definition of this quantity.

V. CONCLUSIONS

In order to establish our conclusions, firstly we would like to discuss some physical aspects and hypotheses con-

tained in this paper. In the numerical calculation procedure we have adopted a value for the phonon absorption frequency, μ, lower by 3 orders of magnitude than the phonon emission frequency, ν. We justify such a great difference in Ref. 19 by comparing the probabilities for phonon emission and absorption in thermal equilibrium. In the present case, in order to introduce in a phenomenological way the out-of-equilibrium phonon population effects, we assumed the dependence $\nu \propto (1+\mathcal{N}_\Omega)$, $\mu \propto (\mathcal{N}_\Omega)$ and adopt for the phonon absorption frequency an initial value comparable to the recombination frequency. This last assumption requires a detailed analysis, which is in process and shall be reported elsewhere. Preliminary results indicate that comparable initial values of μ and ν strongly enhance the phonon population effects.[24]

The expression which we have adopted in order to simulate screening in the *e-e* interaction is motivated by trying to describe the observed increment in the rate of energy exchange by *e-e* collisions when the excitation density grows from a regime of low excitation density to a high excitation one. That expression has the convenience of allowing the testing of different values for the strength of screening. From the comparison presented in Fig. 9(b) we conclude that for the time intervals considered here the variation of the total carrier concentration is not enough to bring out significant differences in the time evolution of the carrier temperature. However, with respect to the thermalization process, we think that T^* and t^* can be sensitive quantities to test phonon population and screening effects, since the arguments in which our discussion is based, mainly, of energy exchange, are to some extent, independent of our model.

We wish to recall here that in the model the description of the CDF as a Boltzmann factor characterized by an effective temperature $T(t)$ is in itself an approximation.[19] Formally a more consistent definition of $T(t)$ might be $T(t) \equiv U(N)$, where $U(N)$ is the average energy per particle. Such a definition would require improvement of the definition of the average energy, by including terms which take into account the fact that carriers are linked by *e-e* interaction, and thus an independent particle type of expression, $\varepsilon = \Delta\varepsilon \sum_i \eta_i / N$, as we have been using must be modified.[24]

Finally, from the results shown in Fig. 9(b) we conclude that the cooling process of the CDF is driven by the phonon creation at the time interval that immediately follows the thermalization condition as can be noticed observing Fig. 8(a). For longer times the cooling process is affected by screening and mainly by the existence of a long-lived phonon population that acts as a thermal reservoir.

ACKNOWLEDGMENTS

The partial financial support of Consejo Nacional de Ciencia y Tecnología (CONACyT) (México) is acknowledged. The authors would like to acknowledge the critical reading of the manuscript by German Luna. During the development of this work one of the authors M.A. Rodríguez received CONACyT support.

*Permanent address: Escuela de Ciencias Físico Matemáticas, Universidad Autónoma de Sinaloa, Apartado Postal 1872, 80000 Culiacán, Sinaloa, Mexico.

[1]R. Ulbrich, Phys. Rev. B **8**, 5719 (1973).

[2]R. J. Seymour, M. R. Junnarkar, and R. R. Alfano, Solid State Commun. **41**, 657 (1982).

[3]D. von der Linde and R. Lambrich, Phys. Rev. Lett. **42**, 1090 (1979).

[4]J. Shah, J. Phys. (Paris), Colloq. **42**, C7-445 (1981).

[5]C. V. Shank, R. L. Fork, R. Yen, J. Shah, B, I. Greene, A. C. Gossard, and C. Weisbuch, Solid State Commun. **47**, 981 (1983).

[6]J. Shah, A. Pinczuk, A. C. Gossard, and W. Wiegmann, Phys. Rev. Lett. **54**, 2045 (1985).

[7]C. H. Yang, J. M. Carlson-Swindle, S. A. Lyon, and J. M. Worlock, Phys. Rev. Lett. **55**, 2359 (1985).

[8]E. J. Yoffa, Phys. Rev. B **21**, 2415 (1980); **23**, 1909 (1981).

[9]W. Pötz and P. Kocevar, Phys. Rev. B **28**, 7040 (1983).

[10]J. A. Kash, J. C. Tsang, and J. M. Hvam, Phys. Rev. Lett. **54**, 2151 (1985); T. A. Perry, R. Merlin, B. V. Shanabrook, and J. Comas, *ibid.* **54**, 2623 (1985).

[11]H. Münzel, A. Steckenborn, and D. Bimberg, J. Lumin. **24/25**, 569 (1981); J. G. Fujimoto, S. G. Shevel, and E. P. Ippen, Solid State Commun. **49**, 605 (1984); D. Hulin, M. Combescot, J. Bok, A. Migus, J. Y. Vinet, and A. Antonetti, Phys. Rev. Lett. **52**, 1998 (1984). T. E. Orlowski and H. Scher, *ibid.* **54**, 220 (1985).

[12]G. Bauer, Solid State Electron. **21**, 17 (1978), and references therein.

[13]A. C. S. Algarte and R. Luzzi, Phys. Rev. B **27**, 7563 (1983).

[14]J. F. Ryan, R. A. Taylor, A. J. Turberfield, A. Maciel, J. M. Worlock, A. C. Gossard, and W. Weigmann, Phys. Rev. Lett. **54**, 1841 (1984); Z. Y. Xu and C. L. Tang, Appl. Phys. Lett. **44**, 692 (1984).

[15]A. C. S. Algarte, Phys. Rev. B **32**, 2388 (1985).

[16]H. M. van Driel, Phys. Rev. B **19**, 5928 (1979).

[17]A. Elci, M. Scully, A. L. Smirl, and J. C. Matter, Phys. Rev. B **16**, 191 (1977).

[18]J. L. Carrillo, J. Reyes, and A. Zehe, Solid State Commun. **45**, 655 (1983); J. L. Carrillo and J. Reyes, Phys. Rev. B **29**, 3172 (1984).

[19]M. A. Rodríguez, J. L. Carrillo, and J. Reyes, Solid State Commun. **53**, 789 (1985).

[20]*Semiconductors Probed by Ultrafast Laser Spectroscopy*, edited by R. R. Alfano (Academic, New York, 1984), Vol. I.

[21]J. Shah and R. C. C. Leite, Phys. Rev. Lett. **22**, 1304 (1969).

[22]R. F. Leheny, J. Shah, R. L. Fork, C. V. Shank, and A. Migus, Solid State Commun. **31**, 809 (1979); C. V. Shank, R. L. Fork, R. F. Leheny, and J. Shah, Phys. Rev. Lett. **42**, 112 (1979).

[23]See for instance in the issue dedicated to the V International Conference of Hot Electrons in Semiconductors, in Physica **134B**, 1 (1985); P. Bordone, C. Jacoboni, P. Lugli, L. Reggiani, and P. Kocevar, *ibid.* **134B**, 169 (1985); S. Das Sarma and B. A. Mason, *ibid.* **134B**, 301 (1985); J. Collet, and T. Amand, *ibid.* **134B**, 394 (1985); J. Collet and T. Ammand, J. Phys. Chem. Solids **47**, 153 (1986).

[24]M. A. Rodríguez and J. L. Carrillo (unpublished).