Nonequilibrium dynamics of hot carriers and hot phonons in CdSe and GaAs

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The dynamics of hot carriers has been investigated extensively for III-V compound semiconductors in the past but not as well for II-VI semiconductors. In this paper, we calculate the hot electron and hole energy-loss rates (ELR's) in CdSe taking into account all relevant phonon emission processes. Interband and intraband scattering in the two uppermost valence bands (A and B) in CdSe are included. We make a detailed comparison of these ELR's with those for GaAs. We then report our experimental measurements of time- and energy-resolved luminescence in CdSe performed using up-conversion luminescence spectroscopy with a time resolution of 2.5 ps. Using these results, we obtain the hot-carrier cooling rates in CdSe in the initial time domain up to 50 ps following photoexcitation at $t = 0$ ps. By varying the excited carrier density (n_0) within the range 10^{17}–10^{18} cm^{-3}, we find that the hot-carrier cooling behavior has a noticeable dependence on the excited carrier density n_0, the cooling getting slower as n_0 increases. We present a detailed comparison of the experimental cooling results with our calculations of the ELR's. The measured cooling rates are much smaller (by over two orders of magnitude) than expected in this theory. The slow cooling can be explained within the hot-phonon theory which takes into account the coupled dynamics of hot carriers and nonequilibrium optical phonons. Using this, we deduce that the lattice dynamical lifetime of the optical phonons in CdSe is about 5 ps (at 8 K, $n_0 = 2 \times 10^{17}$ cm^{-3}). We find that the energy-loss rates (ELR's) are initially dominated by the Fröhlich interaction of the electrons and holes but later, by transverse optical phonon emission by holes, as the carriers cool. It is shown that the reduction in the ELR's as a result of the hot-phonon effects is nearly ten times larger in CdSe than in GaAs. We also study the time dynamics of the nonequilibrium optical phonon occupancies in both CdSe and GaAs.

I. INTRODUCTION

Study of the dynamics of charge carriers in semiconductors under nonequilibrium conditions has been of great interest for some time.[1] These conditions are obtained when the carriers gain energy either under high electric fields or due to photoexcitation under steady or ultrafast pulsed laser illumination. Such carriers lose their excess energy to the lattice via various phonon interactions. Determination of their energy loss rates (ELR's) is of much relevance for understanding the physics of semiconductor devices. The development of ultrafast pulsed lasers has been of immense utility in these studies. It is generally recognized that carriers excited by ultrashort pulses in semiconductors at sufficiently large densities ($> 10^{16}$ cm^{-3}) form a thermalized energy distribution very rapidly, with a temperature which can be higher than the lattice temperature. Such a hot-carrier assembly subsequently cools towards equilibrium with the lattice by emitting optical and acoustic phonons in several picoseconds. Probing the energy distributions of these hot carriers as a function of delay after photoexcitation by ultrafast optical pulses provides direct information on the hot-carrier cooling dynamics. Time-resolved interband luminescence and pump-probe transmission are the two most widely used techniques for this.[2] Study of hot carrier cooling in a variety of semiconductors and their quantum wells, especially in III-V materials such as GaAs and $In_xGa_{1-x}As$, has been a topic of much interest in recent years.[3] This has led to many unexpected results. For example, it has been found that the measured energy-loss rates in these materials are much less than expected on the basis of longitudinal optical (LO) phonon emission via the Fröhlich coupling, the dominant carrier-lattice interaction in such polar semiconductors. Although several mechanisms were proposed to explain this, notably screening of the polar coupling by the carriers[4] and nonequilibrium phonon effects,[5] the latter has been recognized as the most effective mechanism for slowing down the hot-carrier cooling rates. The relative energy-loss rates of electrons and holes in GaAs quantum wells as against those in bulk have also been a topic of great interest and some controversy.[6]

Recent increased interest in the applications of II-VI semiconductors and their quantum wells[7] has made a similar investigation of hot-carrier energy relaxation dynamics in these materials of much relevance. However, detailed information on energy relaxation rates in II-VI semiconductors, such as CdSe and CdS, is not available. Although both III-V and II-VI types of materials have partly ionic bonds and many II-VI semiconductors have a direct band gap like GaAs, a typical III-V material,

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there are a few features which distinguish them from each other. To be specific, we briefly compare the properties of CdSe and GaAs. (The relevant material parameters[8–13] are listed in Table I, see also Fig. 1.) The lowest energy side valley of the conduction band in CdSe (at 2 K) is 1.7 eV (at A point) above the Γ band edge, but only 0.3 eV (at L point) in GaAs, making the complications due to side valley excitation avoidable in CdSe for a typical laser photon energy of ~ 2 eV, but not in GaAs. In GaAs, the valence band has a light and a heavy hole band, degenerate at $\vec{k} = \mathbf{0}$ and a spin-orbit split off band. The valence band at $\vec{k} = \mathbf{0}$ in CdSe splits into three bands, $A(\Gamma_9)$, $B(\Gamma_7)$, $C(\Gamma_7)$. (See Fig. 1.) The hole masses in the two uppermost valence bands (A and B) in CdSe are nearly the same ($\sim 0.8m_0$). The A and B valence band edges are separated by 25.3 meV.[14] For a thermalized hole assembly with a hole temperature (T) of 500 K ($k_BT \sim 40$ meV), about 36% of the holes, excited by 2 eV photons, will be in the B band and about 5% at 100 K ($k_BT \sim 8$ meV). In GaAs on the other hand, only 5% of the thermalized hot holes will be in the light hole band at any T. Since the zone center transverse optical (TO) phonon energy in CdSe is 21.1 meV (33.3 meV in GaAs) and the LO phonon energy is 26.5 meV in CdSe (36.5 meV in GaAs), the intravalence and intervalence band scattering of both A and B band hot holes, due to optical phonon emission can become an important mode of energy loss in CdSe. In comparison, the contribution of the thermalized light holes to the ELR's in GaAs is unimportant. (For the case of carrier excitation by ultrashort laser pulses, this assumes that the holes thermalize among themselves rapidly. In general, intervalence band scattering of holes can be important for III-V compounds, especially for their normal and strained layer quantum wells[15].)

The Fröhlich LO phonon coupling is about 3 [and 9 (Shah *et al.*, Ref. 3)] times stronger in CdSe than in GaAs (and $In_xGa_{1-x}As$). Furthermore, the smaller static dielectric constant in CdSe in comparison with GaAs suggests that the weakening, if any, of the Fröhlich coupling, due to static screening is smaller in CdSe. As shown later in Sec. III, the hole ELR in CdSe (in GaAs), due to nonpolar optical (NPO) deformation potential coupling is about 30 times (4.5 times) smaller than that due to Fröhlich coupling. The NPO phonon interaction of holes is about 0.5 times weaker in CdSe than in GaAs. The intraband scattering of electrons, due to NPO deformation potential scattering is much weaker than the corresponding scattering of holes in both GaAs and CdSe, transitions between the s-like electron states via the NPO deformation potential being forbidden. (Similarly, the Fröhlich coupling is about 3.3 times stronger in ZnSe than in GaAs, but the NPO deformation potential coupling of holes is nearly the same as in GaAs.) Since the ELR's in CdSe and GaAs are dominated by LO phonon emission by electrons and holes, due to Fröhlich mechanism at low excitation densities (ignoring possible reabsorption of the phonons by the carriers, included in the hot-phonon the-

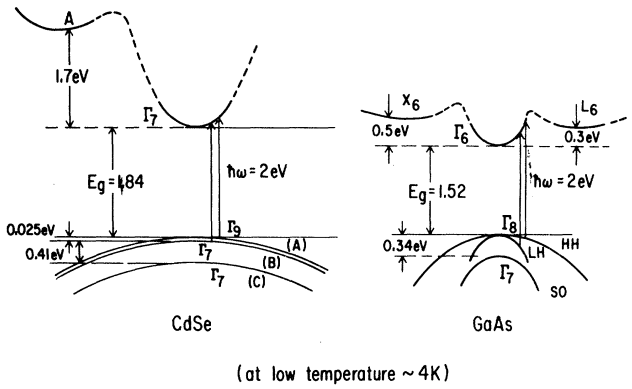


FIG. 1. Schematic band diagrams of CdSe and GaAs.

TABLE I. The material parameters for CdSe and GaAs. m_0 is the rest mass of electron.

Quantity	Symbol	CdSe	GaAs
LO phonon energy (Refs. 9 and 10)	$\hbar\omega_{LO}$ meV	26.5	36.5
TO phonon energy (Refs. 9 and 10)	$\hbar\omega_{TO}$ meV	21.1	33.3
Optical dielectric constant (Ref. 10)	ϵ_∞	6.25	10.63
Static dielectric constant (Ref. 10)	ϵ_o	9.75	12.56
NPO deformation			
Potential (holes) (Refs. 8 and 9)	D_{NPO} eV cm^{-1}	6×10^8	10×10^8
Piezoelectric potential (Refs. 9 and 11)	e^2_{pe} (LA) C^2 m^{-4}	0.0144	0.0043
	e^2_{pe} (TA) C^2 m^{-4}	0.0189	
Acoustic deformation	E_1(dp) hole eV	2.2	3.5
Potential (Refs. 11–13)	E_1(dp) electron eV	4.2	7.0
Effective hole mass (Refs. 9 and 12)	m^*_h	$0.8m_0$ (A)	$0.62m_0$ (HH)
		$0.8m_0$ (B)	$0.08m_0$ (LH)
Effective electron mass (Refs. 9 and 12)	m^*_e	$0.13m_0$	$0.067m_0$
Sound velocity (Refs. 9 and 10)	u cm sec^{-1}	2.391×10^5	3.57×10^5
Density (Refs. 9 and 10)	ρ gm cm^{-3}	5.81	5.36

ory), the carrier cooling rates are expected to be faster in CdSe than in GaAs.

On the other hand, at sufficiently high excitation densities, the strong Fröhlich coupling in CdSe and GaAs will lead to a buildup of nonequilibrium LO phonons in large numbers, provided that the hot LO phonon lifetime (τ_{LO}) is not very small.[5] One can then no longer ignore the rates of LO phonon reabsorption by the hot carriers in CdSe and GaAs. The hot phonon effects can considerably weaken the ELR's due to LO phonon emission in both GaAs and CdSe, even more so in CdSe, due to the stronger Fröhlich coupling in CdSe. The next most important mechanism of ELR is due to hole-NPO phonon interaction. This coupling, being relatively weaker, does not lead to strong hot-phonon effects such as those caused by the Fröhlich interaction. The hole-NPO phonon coupling is slightly weaker in CdSe than in GaAs, as noted before. In addition, smaller energy is lost in each optical phonon emission in CdSe, due to the smaller energies of the optical phonons in CdSe than in GaAs. Thus, the overall effect of the hot LO phonons may be to have a larger reduction in hot-carrier ELR in CdSe than in GaAs, when the total ELR in the "hot-phonon" case is compared with that obtained in the "no-hot-phonon" case.

To our knowledge, there is no detailed investigation of the above, either experimental or theoretical, performed so far, taking into account all the relevant effects. Previously, Pugnet *et al.*[9] reported their calculation of the ELR's in CdSe and GaAs. The presence of the two valence bands (A and B), however, was not fully accounted for in calculating the ELR's in CdSe. Later, we show that this can be important. There has been no attempt to compare the hot-phonon theory with experiments on CdSe, mainly because detailed experimental information with sufficient time resolution on the hot-carrier cooling rates in CdSe in the initial period up to 50 ps of optical phonon emission, following picosecond excitation at $t = 0$, is lacking. There have been only a few efforts in the past to study hot-carrier cooling in CdS and CdSe.[16,17] For example, Yoshida *et al.*[16] measured hot-carrier temperatures in CdSe using time resolved luminescence with a CS_2 Kerr shutter. However, these studies were performed with insufficient time resolution under the conditions of a large laser pulse width (20 to 30 ps). Recently, Masumoto and Sasaki[17] have studied hot-carrier cooling in thin CdSe (0.65 μm) with 0.4 ps time resolution at a very high density ($\sim 10^{19}$ cm^{-3}). Such results, being obtained using pump-probe transmission, have a relatively limited accuracy. Also, the effects of carrier excitation density on the hot-carrier cooling rates have not been studied.

In this paper, we report what is to our knowledge the first definitive study of hot-carrier cooling in CdSe. The results are presented as follows: We first write in Sec. II the basic equations and the various assumptions made in our calculations of the ELR's. Using this formalism, we calculate and compare the results of our calculations of the ELR's in CdSe and GaAs in Sec. III, obtained by taking into account all relevant phonon emission processes, but ignoring the phonon absorption processes (no hot phonons). The degenerate carrier statistics and the presence of the two uppermost valence bands (A and B) in CdSe is included in calculating the hole Fermi energy and intervalence and intravalence band hole-phonon scattering. In Sec. IV, we report our experimental results on hot-carrier cooling rates in CdSe, obtained using upconversion luminescence spectroscopy at 8 K with a 2.5 ps time resolution at different carrier excitation densities ($n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}). In Sec. V A, we compare the experimental hot-carrier cooling rates in CdSe with our theoretical calculations based on the ELR's of Sec. III performed without including the effects of nonequilibrium (hot) optical phonons emitted by the cooling e-h plasma. The theory without the hot-phonon effects predicts cooling rates much faster (by about two orders of magnitude) than the experimental rates. To explain the observed slow cooling, we next solve the equations describing the coupled dynamics of the carriers and the optical phonons to calculate the cooling rates within the hot-phonon theory (Sec. V B). By comparing these results with experiments, we deduce that the lattice dynamical optical phonon lifetime in CdSe (at 8 K) is 5 to 8 ps, depending upon n_0. We obtain the time evolution of the nonequilibrium longitudinal and transverse optical (LO and TO, respectively) phonon wave vector distributions. We find that the cooling occurs initially via the Fröhlich mechanism, but as the LO phonon population builds up, the LO phonons emitted by the hot carriers via the Fröhlich interaction feed energy back to the carrier system. The cooling then occurs mainly due to phonon emission via NPO deformation potential interaction of the holes. We make similar calculations of the ELR's in GaAs and compare the results with those obtained for CdSe to show that the reduction in the total ELR, due to hot-phonon effects is relatively larger in CdSe by a factor as large as 10. The main results are summarized in the concluding section (Sec. VI).

II. THEORY: BASIC FORMALISM

In this section, we write down the basic equations used in our calculations of the ELR's of the carriers. We assume that the generated electrons and holes are sufficiently large in number ($> 10^{16}$ cm^{-3}) to form thermalized energy distributions among themselves at an effective temperatures T_e (for electrons) and T_h (for holes), due to carrier-carrier scattering. (In general, T_e and T_h may not be equal for an initial time period, comparable to the characteristic e-h energy transfer time constant.)

In the case of CdSe, the ELR's due to holes in both A and B valence bands (and in the light and heavy hole bands for GaAs) should be included. The carrier energy distributions at the carrier temperature T_c ($c = e, h$) are given by Fermi-Dirac statistics. The quasi-Fermi energies E_e^F and E_h^F, measured from the conduction and valence band edge, respectively, for electrons and holes are determined by solving the following equations:

$$n_0 = N_e F_{1/2}(\eta_e), \tag{1}$$

$$N_e = 2[2\pi m_e^* k_B T_e/h^2]^{3/2}, \tag{2}$$

$$n_0 = N_h(1)F_{1/2}(\eta_h) + N_h(2)F_{1/2}(\eta_h - \delta), \quad (3)$$

$$N_h(i) = 2[2\pi m^*_{h(i)} k_B T_h/h^2]^{3/2}, \quad (4)$$

where $\eta_c = E_c^F/k_BT_c, c = e, h$. Here, n_0 is the carrier density, "F"s are the Fermi integrals, $\delta = \Delta/k_BT_h$, $\Delta(= 25.3$ meV) being the A-B band separation in the case of CdSe, i is the valence band index (i=1,2) and $N_h(1) = N_h(A) \approx N_h(2) = N_h(B)$. [In the case of GaAs, $\delta = 0$, but $N_h(1)$ and $N_h(2)$, respectively, correspond to the light and heavy holes.] Using the above equations, the electron and hole quasi-Fermi energies and hence the carrier Fermi-Dirac energy distribution functions can be obtained at T_c ($c = e, h$).

The hot carriers cool by losing energy to the lattice. The average rate of loss of energy per carrier in time is related to the average rate of phonon generation by the carrier. We have to consider phonon generation by both electrons and holes. The electrons and holes interact with LO phonons via the Fröhlich mechanism and with acoustic phonons via piezoelectric and deformation potential interactions. However, only holes emit optical phonons via the NPO deformation potential interaction. The average ELR for carrier of type c (= e or h), due to emission of phonons via s-type carrier-phonon coupling in a carrier scattering from band i to band j is given by[18]

$$\left\langle \frac{dE}{dt} \right\rangle_{c,s}^{\mathrm{ph},i,j} = -\frac{1}{n_0 V} \sum_{\vec{q}} \hbar\omega_q^s \left[\left(\frac{\partial N_q^s}{\partial t} \right)_c \right]_{i,j}, \quad (5)$$

where

V = crystal volume;

$\hbar\omega_q^s$ = energy of phonon emitted via s type of carrier-phonon coupling;

s = Fröhlich (LO phonon), nonpolar optical (NPO), and acoustic deformation potential (DP) and piezoelectric (PE) coupling, as appropriate for a carrier c;

$((\partial N_q^s/\partial t)_c)_{i,j}$ = net rate of the phonon generation by c type carriers in the i to j band scattering, via s type phonon coupling.

$\vec{q}$ = phonon wave vector.

We have for electrons (i.e., $c =$ e), $i = j =$ conduction band and for holes ($c = h$), $i,j = A$ or B valence band in CdSe and light or heavy hole band in GaAs. The rate of generation of phonons by a carrier of type c (= e or h) in intraband ($i \to i, i =$ conduction band and A and B valence bands in CdSe) or intervalence band ($A \leftrightarrow B$ in CdSe) transitions, due to interaction of type s (with both phonon emission and absorption included) is

$$\left[\left(\frac{\partial N_q^s}{\partial t} \right)_c \right]_{i,j} = \left(\frac{2\pi}{\hbar} \right) \sum_{\vec{k}} \Big\{ (N_q^s + 1)|M_{q,c}^s|^2 f_i(\vec{k} + \vec{q})[1 - f_j(\vec{k})]\delta(E^i_{\vec{k}+\vec{q}} - E^j_{\vec{k}} - \hbar\omega_q^s) - N_q^s|M_{q,c}^s|^2$$
$$\times f_j(\vec{k})[1 - f_i(\vec{k} + \vec{q})]\delta(E^j_{\vec{k}+\vec{q}} + \hbar\omega_q^s - E^i_{\vec{k}}) \Big\}, \quad (6)$$

where $E^l_{\vec{k}}$ is the energy of carriers with a wave vector $\vec{k}$ in band l (we assume $E^l_{\vec{k}} = \hbar^2k^2/2m_c^*$) and $f_l(\vec{k})$ is the (isotropic) momentum Fermi distribution function in band l. The matrix elements $|M|^2$ are given as

$$|M_{q,c}^s|^2 = (2\pi\hbar\omega_{\mathrm{LO}}e^2)\left[\left(\frac{1}{\epsilon_\infty} \right) - \left(\frac{1}{\epsilon_0} \right) \right] \Big/ Vq^2, \quad s = \text{Fröhlich coupling};$$

$$|M_{q,c}^s|^2 = (D^2\hbar/2\rho\omega_0 V), \text{where } D = \text{NPO deformation potential}, \quad s = \text{NPO deformation potential interaction};$$

$$|M_{q,c}^s|^2 = (\hbar E_1^2/2\rho u V)q, \quad s = \text{acoustic deformation potential interaction};$$

$$|M_{q,c}^s|^2 = (\hbar/2\rho u V)[4\pi e e_{pe}/\epsilon_0\sqrt{q}]^2, \quad s = \text{piezoelectric coupling}:$$

(The various symbols used are standard[18,19] and are also explained in Table I.)

The carrier energy distribution functions are time dependent, the time dependence coming from the carrier cooling, as well as due to possible changes in the Fermi energies as the carrier density, decreases due to recombination, diffusion, etc. Similarly, the nonequilibrium optical phonon occupancy may depend on time due to their rapid generation by the carriers and the optical phonon decay into acoustic phonons. At high carrier densities, the occupancy N_q of the generated phonons may considerably exceed $N_q(T_L)$, the phonon occupancy at equilibrium with the lattice at low lattice temperatures (T_L). The hot carriers initially cool rapidly predominantly by emitting optical phonons, as the coupling of the carriers to optical phonons is much stronger than to acoustic phonons. Thus, we need to consider a possible build up only of the nonequilibrium optical phonons with

$N_q > N_q(T_L)$. The dynamical equations describing the buildup and the subsequent decay of these phonons may be written as

$$\left(\frac{\partial N_q^{s'}}{\partial t}\right) = \sum_{i,j}\sum_{c}\left[\left(\frac{\partial N_q^{s'}}{\partial t}\right)_c\right]_{i,j} - \frac{[N_q^{s'} - N_q^{s'}(T_L)]}{\tau_{s'}}, \tag{7}$$

where $c = e(h)$ electron (hole); and s' is Fröhlich and NPO coupling. Note that for NPO coupling, we have contribution from holes only. Both LO and TO phonons can couple to the holes via the NPO deformation potential interaction. In comparison with the Fröhlich LO phonon coupling, however, the NPO interaction of LO phonons is of secondary importance. Thus, for LO phonons, we retain contributions of both electrons and holes via the Fröhlich coupling and only of holes for TO phonon generation via the NPO deformation potential interaction in Eq. (7). Here, $\tau_{s'}$ is the LO or the TO phonon lattice dynamical lifetime. When the nonequilibrium LO and TO phonon occupancies cannot be ignored in Eq. (6), we need to solve Eq. (7) to get the phonon occupancy $N_q^{s'}$, as a function of time and $\vec{q}$, the phonon wave vector. Note that the first term on the right hand side of Eq. (7), given by Eq. (6) (for $s = s'$), has factors depending on T_c, which is itself time dependent. Also, the Fermi-Dirac energy distributions of the carriers are T_c dependent. Thus, to calculate the time dependence of $N_q^{s'}$, T_c as a function of time should also be obtained. For this, we first write the average energy per carrier as

$$\langle E\rangle_c = \frac{3}{2}k_B[T_c\gamma_c(T_c)], \tag{8}$$

with $c = e, h$, $\gamma_e(T_e) = F_{3/2}(\eta_e)/F_{1/2}(\eta_e)$ and $\gamma_h(T_h) = [F_{3/2}(\eta_h) + F_{3/2}(\eta_h - \delta)]/[F_{1/2}(\eta_h) + F_{1/2}(\eta_h - \delta)]$. Then the rate of change in $\langle E\rangle$ in time *due to cooling* of carriers of type c is

$$\left[\frac{d\langle E\rangle}{dt}\right]_c = \frac{3}{2}k_B\Phi(T_c,\eta_c)\left(\frac{dT_c}{dt}\right), \tag{9}$$

where $c = e, h$ and $\Phi(T_c,\eta_c) = \{\gamma_c(T_c) + T_c[d\gamma_c(T_c)/dT_c]\}$ ($\Phi \to 1$ for single hole band and nondegenerate electrons and holes). Now, the total ELR per carrier (of type c) is given in terms of rates of phonon generation, using Eq. (5), as

$$\left\langle\frac{dE}{dt}\right\rangle^{\rm ph}_{{\rm total},c} = -\frac{1}{n_o V}\sum_{i,j}\sum_{s}\sum_{\vec{q}}\hbar\omega_q^s\left[\left(\frac{\partial N_q^s}{\partial t}\right)_c\right]_{i,j}. \tag{10}$$

This can be further simplified as,

$$\left\langle\frac{dE}{dt}\right\rangle^{\rm ph}_{{\rm total},c} = -\sum_{i,j}\sum_{s}\frac{1}{2\pi^2 n_0} \times \int_0^\infty \hbar\omega_q^s q^2\left[\left(\frac{\partial N_q^s}{\partial t}\right)_c\right]_{i,j} dq. \tag{11}$$

We can now write the cooling rate (i.e., the rate of change of T_c in time) as

$$\frac{3}{2}k_B\Phi(T_c,\eta_c)\left(\frac{dT_c}{dt}\right) = \left\langle\frac{dE}{dt}\right\rangle^{\rm ph}_{{\rm total},c} + \left\langle\frac{dE}{dt}\right\rangle^{c}_{e\text{-}h}, \tag{12}$$

where $c = e, h$ and $\langle dE/dt\rangle^c_{e\text{-}h}$ represents the ELR, due to energy transfer, due to e-h scattering. (In the general case in which holes in different valence bands are not assumed to be thermalized among themselves, similar relations will hold separately for holes in different valence bands and energy transfer rates, due to hole interband scattering should then be included). If $\langle dE/dt\rangle^c_{e\text{-}h}$ is known, the above equation can be solved to get the carrier temperature T_c as a function of time t using Eqs. (6), (7), and (11). We see that Eqs. (6), (7), and (12) are all coupled and need to be solved simultaneously as a function of time. Thus, in the hot-phonon case in which the dynamics of the nonequilibrium optical phonons is included, we solve Eqs. (6), (7), and (12) simultaneously to obtain $N_q^{s'}$ (s' = LO,TO) and T_c first and then using Eqs. (6) and (11) obtain the ELR's, due to LO and TO phonon emissions. The contributions to Eq. (11) of the ELR's, due to acoustic phonon interaction, are easily obtained for a given T_c by solving Eq. (6) and using Eq. (5) with $N_q^s \approx N_q^s(T_L)$. Similarly, in the no-hot-phonon case for which the possible buildup of nonequilibrium phonon occupancies is ignored, the ELR's can be obtained in a straightforward manner by evaluating $[(\partial N_q^s/\partial t)_c]_{i,j}$ [Eq. (6)] with $N_q^s \approx N_q^s(T_L)$ for all s. The average ELR of a carrier due to s type phonon scattering is given by

$$\left\langle\frac{dE}{dt}\right\rangle^{\rm ph}_{c,s} = \sum_{i,j}\left\langle\frac{dE}{dt}\right\rangle^{{\rm ph},i,j}_{c,s} \tag{13}$$

along with Eq. (5). In Sec. III, we first obtain the ELR's, due to various phonon emission mechanisms in the no-hot-phonon case, for both CdSe and GaAs. Later, in Sec. V, we obtain the ELR's in the hot-phonon case as well.

III. HOT-CARRIER ELR'S, DUE TO PHONON EMISSION IN CdSe AND GaAs (NO HOT PHONONS)

Using Fermi-Dirac statistical distributions for $f_i(\vec{k})$ in Eq. (6), we now evaluate the phonon generation rate $(\partial N_q/\partial t)^{s,c}_{i,j}$ for various scattering processes as follows. For electrons in CdSe or GaAs, we need to consider intraconduction band (Γ) scattering, due to Fröhlich coupling of electrons to LO phonons and due to deformation potential and piezoelectric coupling to acoustic phonons. For holes in CdSe, we have to consider, in addition to the above modes of scattering, hole scattering due to nonpolar optical deformation potential coupling to optical phonons and all the intravalence and intervalence

band scattering: $A \rightarrow A$, $B \rightarrow B$, $A \rightarrow B$, and $B \rightarrow A$. (For holes in GaAs, we may include the corresponding scattering in the light and heavy hole bands.) For CdSe, we assume parabolic conduction and valence bands, with $m_A^* \approx m_B^* (= m_h)$. The rate of generation of phonons in the scattering from band i to band j of carrier c via s type of interaction in CdSe can then be obtained analytically from Eq. (6) to be

$$\left(\frac{\partial N_q}{\partial t}\right)_{s,c}^{i,j} = |M_q|_s^2 \left(\frac{m_c^2 k_B T_c V}{\pi \hbar^5 q}\right) \ln\left[\frac{[1+\exp(P_{i,j}-Q_{i,j})]}{\{1+\exp[P_{i,j}-Q_{i,j}-(\hbar\omega_q/k_B T_c)]\}}\right] [N_q^s(T_c) - N_q^s], \tag{14}$$

where $N_q(T_c)$ is $1/[\exp(\hbar\omega_q^s/k_B T_c) - 1]$, $\hbar\omega_q^s$ is the energy of a s-type phonon at the wave vector q. Also, for CdSe,

$P_{i,j} = \eta_e$ for $i = j$ for electrons;
$P_{i,j} = \eta_h$ for $i, j = A$ and $i = B$, $j = A$
$= \eta_h - \delta$ for $i, j = B$ and $i = A$, $j = B$;
$Q_{i,j} = (\hbar^2/8m_c k_B T_c)[q - (2m_c\omega_q/\hbar q)]^2$ for $i, j = A$ and $i, j = B$ $(c = h)$ and for electrons $(c = e)$
$= (\hbar^2/8m_h k_B T_c)[q - (2m_h\omega_q/\hbar q) \pm (2\Delta m_h/\hbar^2 q)]^2$, with the negative sign for i
$= A$, $j = B$ and the positive sign for $i = B$, $j = A$.

In absence of the B band, Eq. (14) reduces to a well known expression for the phonon generation rates [Eq. (13) in Ref. 19]. The quasi-Fermi energies, η_e and η_h and the A to B band separation parameter δ are defined in Sec. II. In the case of GaAs, $\delta = 0$. Further, we assume that the contribution of light holes to the total ELR is small, as an approximation. Thus for GaAs, we retain only the intraband scattering contributions from the $P_{i,j}$ and $Q_{i,j}$ terms. In the general case, the time dependence of $N_q^{s'}$ (s' = LO, TO) is given by Eq. (7). However, in the no-hot-phonon case, $N_q^{s'}(\approx N_q^{s'}(T_L))$ can be ignored in comparison with $N_q^{s'}(T_c)$ in Eq. (14). (In CdSe at 8 K, for example, $N_{\rm LO} \sim 5 \times 10^{-18}$ cm^{-3}.) Using Eqs. (5), (13), and (14), we can then separately calculate the various contributions of phonon emission processes to the total ELR, which in turn is given by Eq. (11).

In our calculations, we multiply the ELR, due to holes by a phenomenological factor of 0.5 to account for the reduced hole scattering, due to the p-like states of the valence band.[20] Also, we ignore the q dependence of $\hbar\omega_{\rm LO}$ and $\hbar\omega_{\rm TO}$ in our calculations. For acoustic phonons, we write $\hbar\omega_q \approx \hbar uq$, u = sound velocity. We now describe the results of our calculations of the ELR's for each of the various phonon scattering processes in CdSe. In Sec. III B, we will compare the ELR's in CdSe with those in GaAs. The material parameters used to calculate all the rates are already given in Table I.

A. ELR's in CdSe

Figure 2 shows the ELR's in CdSe originating from various types of scattering processes (s =LO,NPO,DP,PE) as a function of T_c. We clearly see that for $T_c > 25$ K, the ELR, due to Fröhlich (LO) interaction, is the most dominant, followed by ELR, due to the NPO interaction. The ELR due to LO emission is over 30 times larger than the ELR due to NPO interaction. The ELR's, due to DP and PE interactions, are almost four orders of magnitude smaller than the ELR, due to LO phonons for $T_c > 100$ K. At low temperatures, the LO and the NPO contributions decrease and for $T_c \leq 20$ K, the DP and PE interactions dominate. Figure 3 shows the total ELR's, due to holes and the ELR's, due to various interband and intraband scattering. The ELR, due to

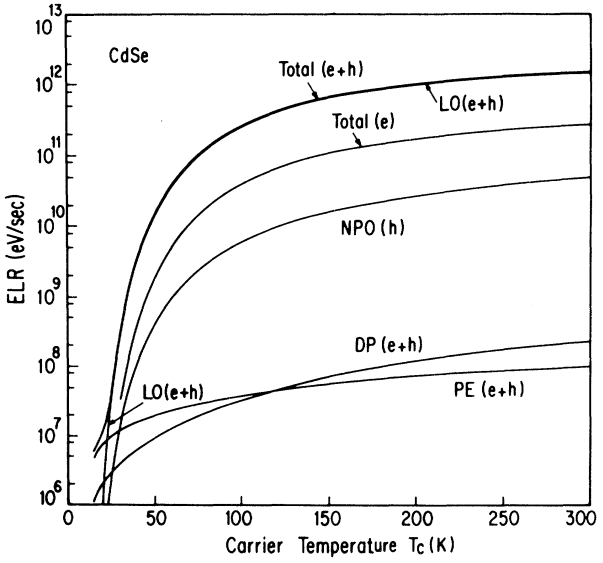


FIG. 2. The ELR's due to various phonon interactions (LO, NPO, DP, and PE) and the total ELR in CdSe at 8 K in the no-hot-phonon case at a carrier density $n_0 = 8 \times 10^{17}$ cm^{-3}. The total ELR only of electrons is also shown.

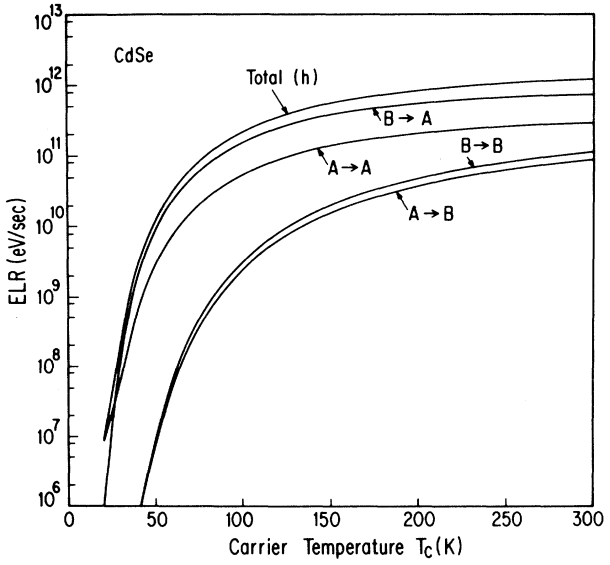


FIG. 3. The ELR's of holes in intravalence and intervalence band phonon and the total ELR (due to holes only) in CdSe at 8 K in no-hot-phonon case at carrier density $n_0 = 8 \times 10^{17}$ cm^{-3}.

holes, is about $\sim$ 4 to 5 times larger than that due to electrons in the temperature range $T_c > 100$ K. Another interesting and important feature of the ELR's in CdSe is the role of the intervalence band scattering. Previously, Pugnet *et al.*[9] calculated the ELR's in CdSe by approximating the two hole bands by a single effective hole band. We have considered here all possible intervalence band scattering mechanisms to determine the hole ELR's. Figure 3 shows the hole ELR's, due to various intraband and intervalence band scattering in CdSe. We find that intervalence band scattering makes an important contribution to the total ELR. In fact, the ELR due to $B \rightarrow A$ is the most dominant for $T_c \geq 30$ K, followed by $A \rightarrow A$, $B \rightarrow B$, and $A \rightarrow B$ processes in that order. This is so because the LO phonons emitted in the $B \rightarrow A$ transitions can have "q"s smaller than the those emitted in $A \rightarrow A$ scattering (for example). This leads to an enhanced ELR as the Fröhlich mechanism has a $1/q$ dependence in the matrix element.[8] Although the population of holes in B band decreases with decreasing hole temperature, [it is merely 5% (0.01%) of the total number of excited holes at 100 K (30 K)], the total ELR is seen to be still dominated by the $B \rightarrow A$ scattering. This, of course, is due to the dominance of the LO phonon emission rates (Fig. 2) for $T_c > 25$ K and the fact that only a small percentage of the total number of holes in the A band (comparable to the number of holes in the B band) have energy large enough to emit LO phonons at these temperatures. Thus, the $B \rightarrow A$ scattering rates are important at all hole temperatures above 25 K. The ELR, due to $B \rightarrow A$ transitions, is almost 5 times larger than that due to $A \rightarrow B$ and about twice as high as $A \rightarrow A$ at $T_c \sim 500$ K. The magnitude of the ELR's in $B \rightarrow B$ scattering is of the same order as that due to $A \rightarrow B$. In summary, we find that approximating the two valence bands in CdSe by a single effective hole band may not be adequate for determining the correct hole ELR's in CdSe. In the next section, we obtain the ELR's in GaAs under the simplification made here and compare them with those in CdSe. (As mentioned earlier, evaluating the relative contributions to the ELR's of the holes in different valence bands in III-V compounds and their quantum wells may require separate consideration in the general case, similar to the above.)

B. Comparison with GaAs

The valence band of GaAs has a light hole, a heavy hole, and a split off band. For simplicity, we consider only that case of excitation in which the thermalized hole occupancy of the split off band can be ignored. As noted earlier, the ratio of the number of thermalized holes in the light and heavy hole valence bands (degenerate at $\vec{k} = \mathbf{0}$), at any hole temperature, is about 5%. It is, therefore, quite adequate to replace the two bands by a single effective valence band for calculating the ELR's of holes in GaAs. We calculate the ELR's for different types of scattering mechanisms in GaAs, using Eq. (5) and Eq. (13) in the no-hot-phonon case. They are shown in Fig. 4. Once again, the ELR, due to LO phonon emission, dominates at all T_c, except at low temperatures ($<$ 40 K), at which the DP and PE ELR's become relatively more important. The ELR, due to NPO interaction, is almost one fourth of that due to LO phonons in the temperature range $T_c > 200$ K. For $T_c > 100$ K, the ELR's due to LO or NPO interactions are almost two and four

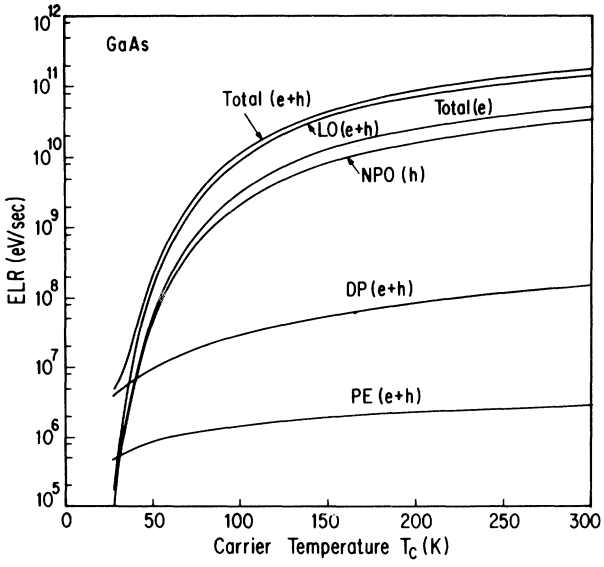


FIG. 4. The ELR's in the case of GaAs are shown, to be compared with those for CdSe (Fig. 2).

orders of magnitude higher than the ELR's, due to DP and PE couplings, respectively. The NPO-ELR and DP-ELR in both GaAs and CdSe are each of nearly the same magnitude. We find that the ELR, due to PE coupling, is one order of magnitude smaller than the ELR, due to DP coupling in GaAs, while in CdSe the ELR's, due to DP and PE interactions, are roughly of the same order. In fact, the PE-ELR in GaAs dominates over DP-ELR below 100 K. We see that the PE-ELR in GaAs is smaller by more than one order of magnitude than in CdSe, the PE interaction in GaAs being weaker than in CdSe. The LO-ELR in GaAs is about five times smaller than the LO-ELR in CdSe in the temperature range $T_c > 150$ K. The ratio (R) of the total ELR's in CdSe and GaAs is about 5 at $T_c = 1000$ K. As T_c decreases below 500 K, R begins to increase and is about 20 at $T_c = 100$ K. This is so because the ELR, due to LO phonon emission in GaAs, decreases more rapidly below 50 K than that in CdSe. We now describe our experiments to determine the ELR's in CdSe. In Sec. V, we compare the experimental ELR's with those obtained in this section (Sec. III). We also consider the hot-phonon effects in the hot-carrier cooling behavior and compare the calculated ELR's within the hot-phonon theory for both CdSe and GaAs.

IV. EXPERIMENTAL RESULTS FOR CdSe

In our experiments, pulses from a Nd-YAG pumped Rhodamine 6G dye laser at 612 nm are used for carrier excitation in CdSe at a repetition rate of 76 MHz. The pulse width of 1.8 ps is determined using an autocorrelator trace, while the spectral half width at full maximum is 7 Å, as measured using the laser light scattered from the sample. The beam is focused to a spot of about 50 μm in diameter on the CdSe crystal, maintained at 8 K on a cold finger in a cryostat. The beam is vertically polarized and is perpendicular to the c axis of CdSe. The exciting pulses cause transitions from both the Γ_9 like A and Γ_7 like B valence bands into the Γ_7 like conduction band, separated by ~ 1.84 eV from the A band (at 8 K) (see Fig. 1).[10] The luminescence from the sample is collected and focused onto a $LiIO_3$ nonlinear optical crystal for upconversion. The upconverted signal is dispersed with a 0.35 m monochromator and detected using a standard photon counting set up and a cooled GaAs photomultiplier tube. The energy resolution is about 2.5 meV. The time resolution in these measurements is 2.5 ps, as determined by upconverting the laser light scattered from the sample. The luminescence energy spectrum covers the range of 1.83 to 1.88 eV. The carrier densities excited in the sample are $2\times, 4\times$, and 8×10^{17} cm^{-3}. These are estimated on the basis of the number of photons absorbed in the sample per unit area per pulse, taking an absorption depth of 0.2 μm and a reflectivity of 0.2.[21]

For the moderately large carrier densities used in our experiments, we assume the carrier-carrier interactions to be fast enough for the electrons and holes to rapidly thermalize among themselves to form Fermi energy distributions at a common temperature T_c (> lattice temperature) as they are generated. The luminescence spectrum, due to spontaneous recombination of electrons and holes with energies in the tail distributions, is expected to have an approximate $(\hbar\omega)^2 \exp(-E/k_B T_c)$ behavior in the photon energy $\hbar\omega$. Comparing this with the data, the carrier temperatures are obtained and are shown in Fig. 5 (discrete points), as a function of various delays following photoexcitation centered at $t = 0$ ps for three carrier excitation densities. Note that the carrier excitation density n_0 has a significant influence on the cooling behavior, the energy-loss rates decreasing as n_0 increases. A similar behavior has been noticed for GaAs, but not in $In_xGa_{1-x}As$, where the cooling rates appear to be insensitive to a carrier density variation in the range $n_0 \approx 10^{17} - 10^{18}$ cm^{-3}. In the next section, we compare the experimental results with the theory of carrier energy relaxation in detail.

V. COMPARISON WITH THEORY

Using the framework of Secs. II and III, we can make a detailed comparison of the experimental cooling rates obtained for CdSe in Sec. IV with theory, first in the "no-hot-phonon" case and then including the "hot phonons."

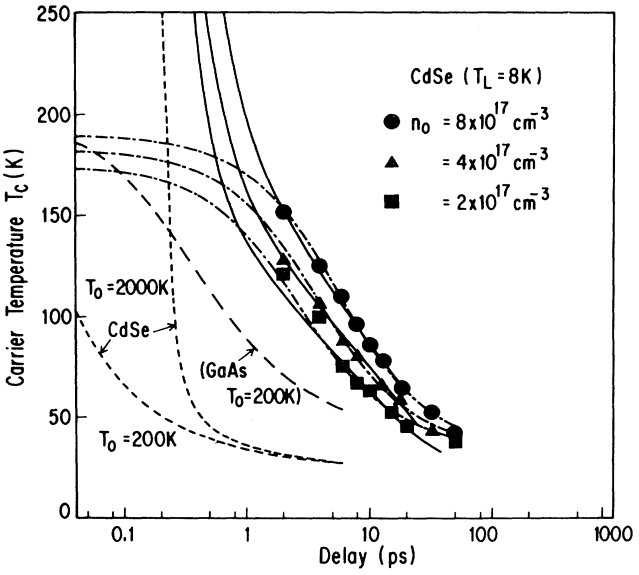


FIG. 5. The time evolution of T_c obtained experimentally is shown for three different carrier excitation densities (discrete points). Cooling rates calculated for $C_0 = 1$ [Ref. Eq. (15)], with no hot phonons (dashed curve) are shown for $T_0 = 200$ K and 2000 K. For comparison, cooling rate in GaAs for $C_0 = 1$ in the no-hot-phonon case with $T_0 = 200$ K is shown. Also shown (dash-dotted curves) are the no-hot-phonon calculations (for CdSe), with C_0 chosen to fit the data ($T_0 = 175$, 183, and 190 K for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respectively). Calculations including hot-phonon effects (solid curves) are also seen to fit the data well with $T_0 = 2050$, 2150, and 2250 K for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respectively.

A. Cooling behavior with no hot phonons

We have already obtained the total ELR per e-h pair at the effective carrier temperature (T_c) in Sec. III with no hot phonons. We now use this to obtain the rate of change of T_c in time. In what follows, we make the usual assumption that the electrons and holes attain a common effective temperature, T_c ($= T_e = T_h$) in a very short time after their generation. This is a reasonable assumption for conditions of strong and frequent e-h collisions and may be valid for high densities ($\sim 10^{17}$ cm^{-3} and higher) for most of the time, except for very early times after excitation by very short pulses.[22] Under these conditions, the second term on the right hand side of Eq. (12) (ELR, due to e-h scattering) can be removed, the role of carrier-carrier interactions being only to maintain a common T_c for all carriers. To simplify the calculations further, we assume that the carriers are generated instantaneously at t = 0 ps by the excitation pulse, and that they immediately attain Fermi distribution at an effective temperature, T_c. This is not unreasonable for ultrashort ps pulses in view of the time duration of several tens of psecs over which the cooling occurs. The average energy ($\langle E \rangle$) per e-h pair is obtained using Eq. (8) and the rate of change in $\langle E \rangle$ in time, *due to cooling*, is given by Eq. (9). The average rate of change in the energy of the carriers, due to the generation of phonons, is already given in Sec. II [Eq. (10)] and in Sec. III. The rate of change of T_c with time is given by Eq. (12), which is now rewritten as

$$\left(\frac{dT_c}{dt}\right) = C_0 \frac{\sum_{c=e,h} \left\langle \frac{dE}{dt} \right\rangle^{\rm ph}_{{\rm total},c}}{\frac{3}{2} k_B \sum_{c=e,h} \Phi(T_c, \eta_c)}. \qquad (15)$$

The right hand side of Eq. (6) includes all the scattering mechanisms via which the electrons and holes lose energy to the lattice. Note that we have introduced the factor C_0 in Eq. (15) as a multiplier. [C_0 equals unity according to Eq. (12). However, we may need to adjust C_0 to simulate a possible reduction in the ELR's, so as to fit the data.] We now obtain T_c as a function of time using Eqs. (11) and (12), under the assumed initial condition: $T_c = T_0$ at $t = 0$. Also, we treat n_0 as time independent, in the time domain of a few tens of psecs, for the present.[23]

The solution $T_c = T_c(t)$, thus obtained using the material parameters for CdSe (Table I), is shown in Fig. 5 for two cases: $T_0 = 200$ K and 2000 K for $C_0 = 1$ (dashed curves). The calculations are unable to satisfy the data. Obviously, the experimental cooling rates are much slower than those expected theoretically on the basis of Eqs. (11), (12), and (15), with $C_0 = 1$. It is possible to obtain a satisfactory fit to the data by appropriately reducing the factor C_0 in Eq. (15). As shown in Fig. 5, the solutions (dash-dotted curves) fit the data for $C_0^{-1} = 80$, 120, and 180 for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respectively. These reduction factors may be compared with the corresponding factors of $C_0^{-1} = 100$ and 10 previously obtained for GaAs (Leo *et al.*[3]) and $In_{0.53}Ga_{0.47}As$ (Kash and Shah[3]) for $n_0 \sim 10^{18}$ cm^{-3}, respectively. (See, however, Lobentanzer *et al.*,[3] who observe slower cooling in $In_{0.53}Ga_{0.47}As$.) The initial temperatures $T_0 = 175$, 183, and 190 K for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respectively, were used in these calculations. The small (<4%) increase in T_0, with n_0 required to fit the data, presumably arises because the model does not take into account the carrier and phonon dynamics, during the finite time (1.8 ps) of carrier generation. Although the excess energy of the carriers at the time of injection is the same in all these cases of n_0, the maximum temperature attained by the carriers, averaged over the pulse width, may be determined by the carrier ELR's, during the finite time of generation, as modified by screening. Thus, the larger the n_0, the larger may be the effects of reduced ELR's, due to screening. This has to be simulated by a small increase in T_0.

Comparison with GaAs cooling rate

The theoretical cooling rates with $C_0 = 1$, obtained for CdSe (dashed curves in Fig. 5) can be compared with those obtained for GaAs under identical initial conditions, namely $T_0 = 200$ K. The results for GaAs are shown in Fig. 5 and, as expected (Secs. I and III), show that the theoretical ELR's (no hot phonons) lead to slower cooling in GaAs than in CdSe.

B. Hot phonon effects

There is no simple explanation within the no-hot-phonon theory, for the reduced cooling rates obtained experimentally in CdSe (Fig. 5). From previous such studies in the case of GaAs, it is known that screening of the Fröhlich interaction due to carriers plays only a secondary role. Since we had ignored the effects of the optical phonon occupancies on the carrier dynamics in Sec. V A above, we now investigate the role of the hot-phonon effects in slowing down the hot-carrier cooling in CdSe. We no more assume that $N_q^s \approx N_q^s(T_L) (\ll 1)$, for s = LO and TO in Eqs. (6) and (7). We now solve Eq. (7) in addition to Eq. (15) (with $C_0 = 1$) to obtain $T_c(t)$ and $N_q^{\rm LO}(t)$ and $N_q^{\rm TO}(t)$ simultaneously. We assume that, the LO and TO phonon energies $\hbar\omega_{\rm LO}$ and $\hbar\omega_{\rm TO}$, respectively, are independent of q, so that $N_q^s(T_L) = 1/[\exp(\hbar\omega^s/k_B T_L) - 1]$ (s= LO,TO) is also independent of q. The range of q values chosen in these calculations is from $q = 1 \times 10^4$ to 1×10^8 cm^{-1} for CdSe and from $q = 5 \times 10^4$ to 1×10^8 cm^{-1} for GaAs, with 50 points in each decade of q. We use Rünge-Kutta fourth order method to solve the dN_q/dt and dT_c/dt equations and the Simpson integration method to calculate dE/dt. All these calculations are performed on DEC-Alpha AXP3000/400 machines running under OSF/1 operating system. For these calculations, we take into account hole occupancy and scattering in both the upper valence bands of CdSe (A and B). We vary T_0 and $\tau_{\rm LO} (= \tau_{\rm TO})$ to obtain a satisfactory fit to the data. The results for these calculations are shown (continuous curves) in Fig. 5 for three excitation densities. We obtain $T_0 = 2100$, 2150, and 2250 K for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respectively. The calculations are seen to satisfy the data well and lead to values of $\tau_{\rm LO}$, the lattice dynamical life time of optical phonons, of 5, 6.5, and 8 ps for $n_0 = 2\times, 4\times$, and 8×10^{17} cm^{-3}, respec-

tively. This may be compared with the values of τ_{LO} = 7 ps obtained earlier for GaAs.[24] The optical phonon lifetime is mainly determined by its decay into acoustic phonons via the anharmonic part in the lattice bonding within the constraints of phonon selection rules. There is no reliable theoretical or experimental estimate of τ_{LO} for CdSe available at present. However, a value of about 1 ps at 300 K is quoted by Baltramiejunas and Zukauskas,[25] deduced from a fit to the dielectric function obtained by Geick *et al.*,[26] using infrared reflectivity measurement on CdSe. [A calculation in which the intravalence and intervalence band hole scattering is replaced by intraband scattering in a single effective valence band as a simplification, gives marginally larger values of τ_{LO} = 6, 7.5, and 9 ps, respectively, for the three excitation densities. The corresponding values of T_0 of = 1850, 1900, and 2000 K, respectively, required to fit the data for the calculation with a single effective hole band are relatively smaller (by about 12%), as the hole ELR's then are smaller in the initial stages of hot-carrier cooling (before the hot phonons build up), than when both A and B hole bands are included. A summary of these results was published recently[27].] The phonon lifetime τ_{LO} deduced in these calculations shows a weak dependence on n_0, the carrier density. Understanding this weak dependence of τ_{LO} on n_0 obtained in these calculations requires further investigation. It is not clear at present whether the rate of decay of the LO phonons into acoustic phonons gets modified as the acoustic phonon density builds up,[25] making the probability of the reverse reaction finite (although small). The larger the excited carrier density, the larger then may be this effect. The other effects which we have not considered in our simplified model are screening of carriers and phonon renormalization,[28] plasma diffusion and recombination and the small but finite time (of a few ps) taken by the electrons and holes to attain a common temperature.

C. ELR's in the hot-phonon case

Figure 6 shows the energy-loss rates in CdSe obtained in case A (no hot phonons C_0 = 1) and case B (hot phonons) both for $n_0 = 8 \times 10^{17}$ cm^{-3}. The rates in case B fit the cooling rate data (for $n_0 = 8 \times 10^{17}$ cm^{-3}, Fig. 5). The reduction in the effective ELR's, due to hot-phonon effects, is clearly seen in Fig. 6. Figure 6 also shows the total ELR's, due to LO and NPO scattering interactions, in the hot-phonon case. We note that the LO-ELR rapidly reduces for $T_c < 300$ K and even becomes negative after about 200 K (corresponding to > 1 ps as in Fig. 5), while the NPO-ELR is positive throughout. This shows that the LO phonons in effect feed energy back to the carrier system, thus reducing the carrier temperature cooling rate. It thus appears that the carrier cooling occurs mainly due to emission of TO phonons by holes and via the NPO interaction for $T_c < 250$ K. This is similar to the case of GaAs, as shown earlier.[5] After a few tens of psecs, these hot-phonon effects diminish as the LO phonons disintegrate into acoustic phonons and the LO-ELR is again positive. In the next section, we compare the ELR's in CdSe with GaAs under a similar choice of n_0, τ_{LO}, and T_0.

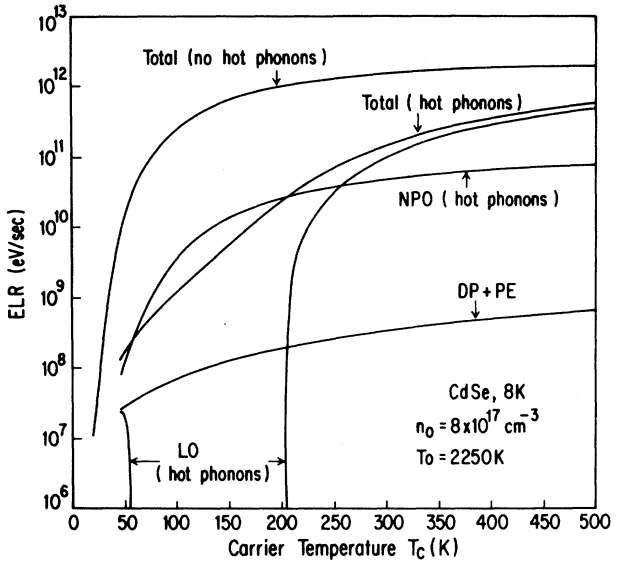


FIG. 6. The ELR's due to different phonon interactions (LO, NPO, DP, and PE) and the total ELR in CdSe at 8 K in the hot-phonon case at a carrier density n_0 of 8×10^{17} cm^{-3} and initial carrier temperature T_0 = 2250 K are shown. For comparison, the total ELR in the no-hot-phonon case is also shown for the same n_0.

D. Comparison with GaAs

Figure 7 shows that the corresponding total ELR in the no-hot-phonon case (C_0=1) and the various rates in the

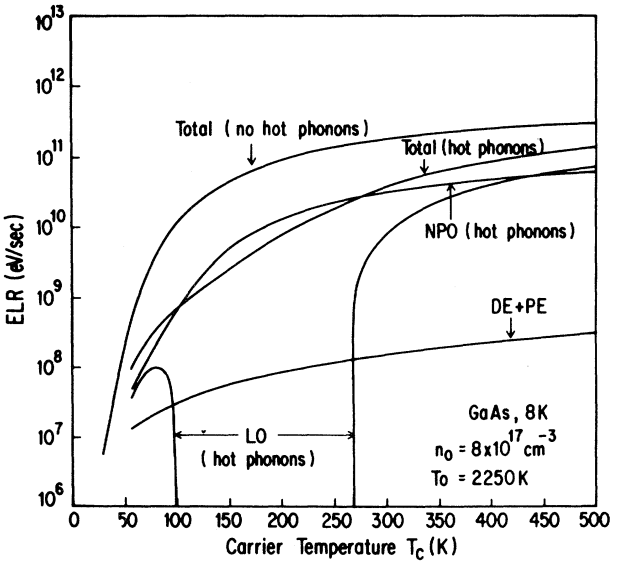


FIG. 7. The different ELR's in GaAs are shown, with n_0 and T_0 as in Fig. 6.

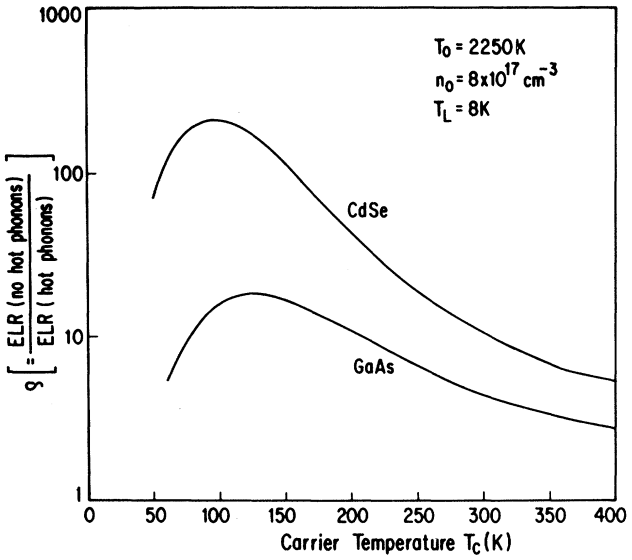


FIG. 8. The reduction in the ELR, due to hot phonons in CdSe is compared with that in GaAs.

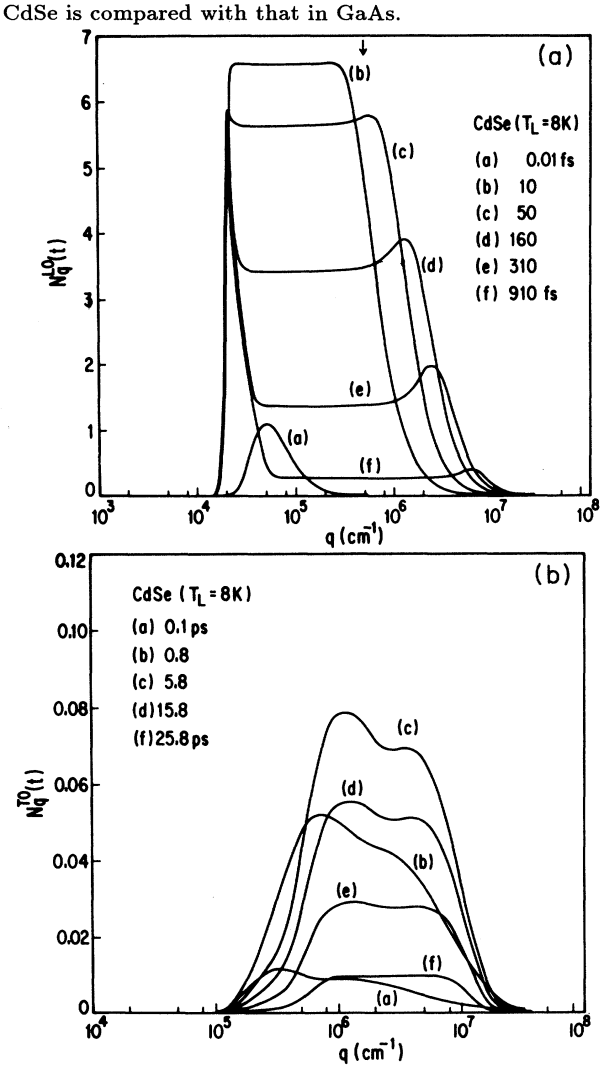
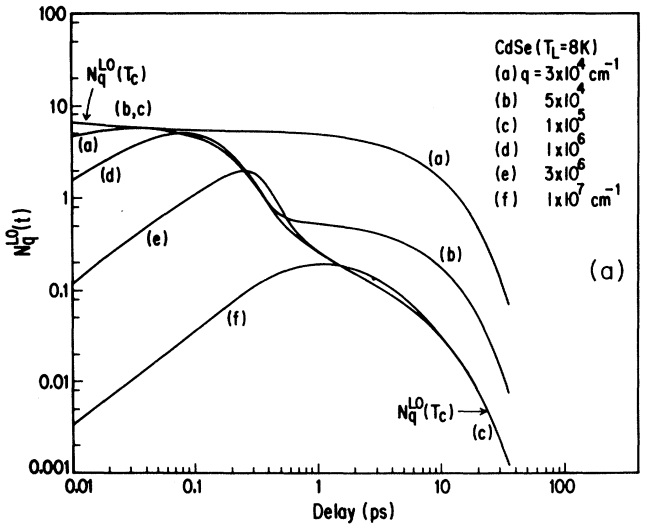


FIG. 9. The q distribution of the LO (a) and TO (b) phonon occupancies in CdSe at $n_0 = 8 \times 10^{17}$ cm^{-3} and $T_0 = 2250$ K, obtained from the fit to the data in Fig. 5.

hot-phonon case with $n_0 = 8 \times 10^{17}$ cm^{-3}, $\tau_{LO} = 7$ ps, and $T_0 = 2250$ K in GaAs. Since $\hbar\omega_{LO}$ is larger in GaAs (=36.5 meV) than that in CdSe (=26.5 meV), $N_q^{LO}(T_c)\{= 1/[\exp(\hbar\omega^{LO}/k_B T_c) - 1]\}$ is smaller in GaAs than in CdSe at a given T_c. Under similar conditions of n_0, T_0, and τ_{LO}, the nonequilibrium LO phonon occupancy, therefore, exceeds $N_q^{LO}(T_c)$ at a larger T_c in GaAs, than in CdSe, thus causing the LO-ELR to become negative at $\sim$ 250 K in GaAs to be compared with the corresponding T_c of $\sim$ 200 K in CdSe.

As discussed in Sec. I, the size of the reduction in the ELR's due to hot-phonon effects is expected to be larger in CdSe than in GaAs. This can be examined by com-



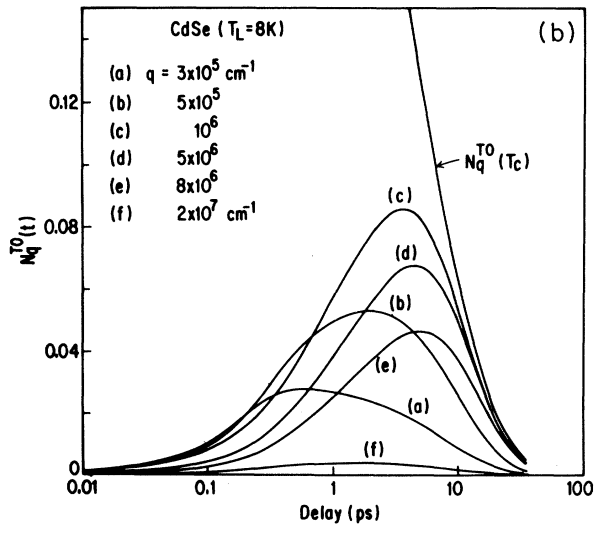


FIG. 10. The time evolution of the LO (a) and TO (b) phonon occupancies in CdSe at $n_0 = 8 \times 10^{17}$ cm^{-3} and $T_0 = 2250$ K, obtained from the fit to the data in Fig. 5.

paring the ratios of the ELR's in the no hot-phonon case with those in the hot-phonon case for CdSe and GaAs, obtained under similar conditions of T_0, n_0, and τ_{LO}. It is clear from Fig. 8 that the reduction in the ELR's due to hot phonons, as represented by ρ, is on the whole larger in CdSe than in GaAs. The ratio ρ for CdSe is about 3 – 10 times larger than that for GaAs in the temperature range of 50 to 400 K. We now study the q and time dependence of the nonequilibrium LO and TO phonons, generated by the hot carriers in both CdSe and GaAs, as obtained in our calculations.

E. Optical phonon dynamics

Figures 9(a) and 9(b) show the q distribution of LO and TO phonons generated by the hot carriers. We expect that due to a smaller effective mass of electrons, as compared to that of holes in both GaAs and CdSe, the q value of the optical phonons emitted by electrons is smaller than by holes. In addition, the LO and TO phonons emitted in the $B \rightarrow A$ scattering in CdSe (especially the LO phonons due to the $1/q$ dependence of the Fröhlich interaction) are emitted with even smaller q's. It can be shown that the smallest q possible for hot LO phonons in CdSe can be approximately estimated using $q_{\min}[\approx \sqrt{(m^*/3k_BT_c)}(|\Delta - \hbar\omega^s|)/\hbar]$ to be $\approx 5\times10^4$ cm^{-1} at $T_c = 2000$ K and the largest by $q_{\max}(\approx 2(\sqrt{3k_BT_cm^*}/\hbar))$ to be $\approx 5 \times 10^7$ cm^{-1} at $T_c = 2000$ K. These values are in good agreement with Fig. 9(a). (Similar estimates can be made for TO phonons.) As the time progresses, the nonequilibrium optical phonon distribution approaches the assumed flat distribution of hot

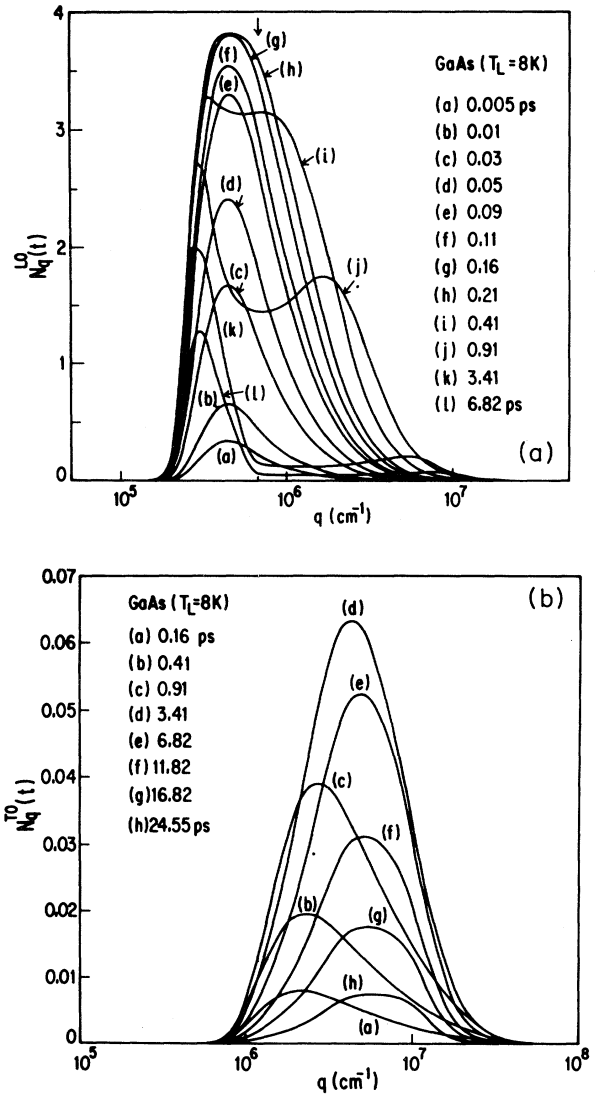


FIG. 11. The q distribution of the LO (a) and TO (b) phonon occupancies in GaAs with $n_0 = 8 \times 10^{17}$ cm^{-3}, $T_0 = 2250$ K, and $\tau_{LO} = 7$ ps is shown, to be compared with Fig. 9.

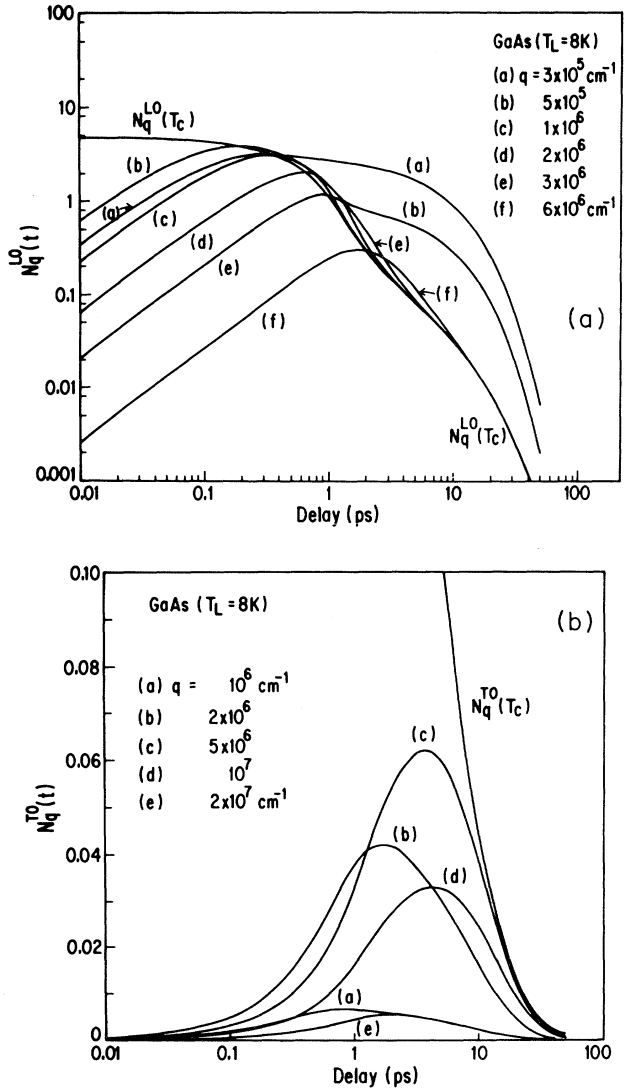


FIG. 12. The time evolution of the LO (a) and TO (b) phonon occupancies in GaAs under the conditions of Fig. 11, to be compared with Fig. 10.

phonons $\{N_q^s(T_c) = 1/[\exp(\hbar\omega^s/k_BT_c) - 1]\}$ in equilibrium with the carriers [see Eq. (7)]. However, LO and TO phonons emitted with very small q wave vectors in the initial stages are not reabsorbed efficiently by the rapidly cooling carriers. These phonons remain in a highly nonequilibrium state and decay with a time constant τ_{LO} [see Figs. 10(a) and 10(b)]. As the carriers cool and approach energies close to the band edges, the optical phonons emitted have increasingly larger q's. The q distributions of Figs. 9(a) and 9(b), therefore, shift to the right on the q axis with time. Thus, Figs. 10(a) and 10(b) show that optical phonons with small q are rapidly generated and those with large q rise relatively more slowly in time. It may also be noted [Fig. 10(b)] that the occupancy of the TO phonons generated by the hot holes via the NPO interaction never exceeds the distribution $(N_q^{TO}(T_c))$ in equilibrium with the carriers. On the other hand, Fig. 10(a) shows that the strong Fröhlich coupling of the carriers with LO phonons leads to their rapid and large generation, reaching occupancies in excess of $N_q^{LO}(T_c)$, the equilibrium distribution at T_c.

For comparison, we show the q distribution of the nonequilibrium LO and TO phonons in GaAs in Fig. 11. The smallest q possible for hot LO phonons in GaAs in this case is due to emission by electrons and is given for example by 4.7×10^5 cm^{-1} at $T_c = 2000$ K, while the LO phonons with a large q value are emitted by holes with $q \approx 3.8 \times 10^6$ cm^{-1} at $T_c = 2000$ K. These values are in agreement with Fig. 11(a). The arrows in Figs. 9(a) and 11(a) indicate the q values of LO phonons detected in a Raman experiment in the backscattering geometry. It is seen that it may be possible to obtain the LO phonon population lifetime in CdSe just as in GaAs in Raman experiments. We show the time evolution of the LO and TO phonon occupancies in GaAs, for a few values of q for comparison (Fig. 12).

VI. CONCLUSION

In this paper, we first calculate hot-carrier energy-loss rates (ELR's) in CdSe and GaAs, using a theory of phonon emission by hot carriers via all relevant carrier-phonon coupling mechanisms in CdSe. We show that intraband and interband hole scattering in both the upper valence bands (A and B) in CdSe make important contributions to the total ELR in CdSe. The total ELR in CdSe obtained in this theory is about 2 to 4 times larger than in GaAs for carrier temperatures in the range 50 K to 300 K. We also report our picosecond time-resolved measurements of the luminescence energy spectra in CdSe and use these to obtain hot-carrier cooling rates in CdSe. The rates are found to be very sensitive to the excited carrier density. These rates are found to be much smaller than those expected in the above simple theory based on phonon emission by hot carriers. We then study the role of nonequilibrium dynamics of optical phonons in reducing the hot-carrier ELR's in CdSe and GaAs. It is found that the stronger carrier-LO phonon coupling in CdSe compared to that in GaAs, in fact, leads to a larger reduction in the ELR's in CdSe in the hot-phonon theory. The experimental hot-carrier cooling rates in CdSe are consistent with this theory provided that the lattice dynamical lifetime of the optical phonons in CdSe is between 5 and 8 ps (depending upon the injected carrier density). As in the case of GaAs, it turns out that the contribution of the LO phonon emission to the hot-carrier energy relaxation in CdSe diminishes (and even becomes negative), as the nonequilibrium LO phonon occupancy overshoots during the carrier cooling process. The cooling then is governed mainly by emission of TO phonons by holes. We also study the nonequilibrium optical phonon wave-vector distribution and its rise and decay with time in both CdSe and GaAs.

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