

Relaxation of a hot-electron–two-mode-phonon system in highly excited $\text{CdS}_{1-x}\text{Se}_x$ crystals

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An investigation of the electron-hole-plasma effective-temperature relaxation in highly excited $\text{CdS}_{1-x}\text{Se}_x$ mixed crystals is presented. The slow (~ 100 -ps) relaxation stage, attributed to the depopulation of the fragments (decay products) of the initially produced nonequilibrium LO phonons, is examined with variation of the alloy composition. The relevant relaxation time dependence on x exhibiting a remarkable drop at small CdSe mole fractions is analyzed in terms of a two-route energy relaxation model considering hot-carrier plasma and two generations of nonequilibrium phonons each originating from both pure constituents of the alloy. The disorder-enhanced cross relaxation between two sublattices of the alloy is inferred to account for the experimental results.

Investigation of semiconductor crystals and structures with the electron and phonon systems far from thermal equilibrium reveals fundamental quasiparticle interactions and energy-transfer routes in the solid state. Much effort was made to understand a slowed carrier effective-temperature relaxation in pure group-III-V and -II-VI crystals.^{1–8} The nonequilibrium-phonon bottleneck in the energy transfer has been shown to hinder the relaxation.^{9,10} The role of the first-generation nonequilibrium phonons directly produced by hot carriers being elucidated unambiguously,^{11–13} a model of second-generation nonequilibrium phonons arising due to anharmonic decay of the first-generation ones was employed to account for late stages of the effective-temperature relaxation.^{14,15} An enhanced effect of nonequilibrium phonons on hot-carrier cooling in quantum wells is still argued (see Ref. 16 and references therein). In this view, semiconductor alloys may provide extended data on energy relaxation routes, since their phonon system is more complex and composition dependent.

The most important property of semiconductor alloys is the possibility of continuous band-gap tuning by varying the composition. This led to wide application of the alloys in manufacturing heterostructure devices, lasers, light-emitting diodes, low-pass filters, and selective radiation detectors. The vibration structure of pseudobinary mixed crystals ($AB_{1-x}C_x$ or $A_xB_{1-x}C$) is of two categories, in accordance with compositional dependence of Raman and IR reflection spectra.¹⁷ In amalgamated-type (one-mode) alloys ($\text{Zn}_x\text{Cd}_{1-x}\text{Se}$), the phonon energies vary continuously with x , and the number of phonon branches is the same as in a pure end-point crystal. In persistent-type (two-mode) alloys ($\text{CdS}_x\text{Se}_{1-x}$, $\text{Al}_x\text{Ga}_{1-x}\text{As}$), the optical-phonon modes of both pure constituents are present, their energies being weakly composition dependent. In both types of mixed crystals the electronic structure, lattice parameters, and other macroscopic physical properties vary continuously with composition parameter x , only deviations from linearity being different.

The amalgamation of the electronic structure can result in certain peculiarities of energy transfer due to band

alteration, for instance, the composition variation is capable of modifying the role of intervalley scattering in group-III-V mixed crystals.^{18,19}

The two-mode nature of small-wave-number optical phonons in persistent-type mixed crystals was shown to cause small changes in energy relaxation due to the phonon-lifetime insensitivity to the alloy composition.²⁰ Again, the anharmonic decay of nonequilibrium zone-center optical phonons produces a subsequent compact population of nonequilibrium large-wave-number phonons in the vicinity of a twice-less (subharmonic) frequency.^{21,22} Such phonons, capable of hindering the energy relaxation in pure crystals,^{14,15} are expected to be less efficient in alloys due to structural-disorder-induced lifetime decrease. However, up to now, no efforts to reveal the effect of large-wave-number fragments of nonequilibrium LO phonons on electron-energy conversion to heat in mixed crystals was made.

The present work aims at investigating the composition behavior of the electron effective-temperature relaxation in a persistent-type alloy $\text{CdS}_{1-x}\text{Se}_x$ excited with picosecond light pulses of an extreme intensity. The slow stage of the relaxation was analyzed both experimentally and theoretically in an attempt to obtain information about phonon-phonon interaction in the large-wave-number region.

$\text{CdS}_{1-x}\text{Se}_x$ is a typical representative of well-randomized alloys (solid solutions) with no inclination for clustering.²³ In addition, some unique properties of $\text{CdS}_{1-x}\text{Se}_x$, such as a highly efficient emission due to annihilation of localized excitons^{24,25} and a nontrivial composition dependence of structural-disorder fluctuations,²⁶ support continuous interest in this mixed crystal.

A series of $\text{CdS}_{1-x}\text{Se}_x$ crystals with $x = 0, 0.05, 0.09, 0.23, 0.63, 0.91$, and 1 was investigated. The crystals were grown by a method of resublimation in argon flow. The values of the mole fraction x were determined from the low-temperature exciton reflection spectra using the exciton-energy bowing function from Ref. 27 for calibration. The samples were excited using a passively mode-locked YAG: Nd^{3+} (yttrium aluminum garnet) laser [the full width at half maximum pulse duration is $\tau_L = 28$ ps,

the spectral linewidth is $\delta(h\nu_L)=0.15$ meV, and the repetition rate is 2.7 Hz]. The second harmonic of the laser irradiation ($h\nu_L=2.33$ eV, and the pump density $I_L=500$ MW/cm² for $x=0.63, 0.91$, and 1) and the third one ($h\nu_L=3.50$ eV, and $I_L=500$ MW/cm² for the rest) were used for excitation. The kinetics of the electron effective temperature was extracted from the time-resolved luminescence spectra taken by means of a computer-controlled spectrometer. The temporal resolution (≈ 30 ps) was provided by a CS₂ optical Kerr shutter. The luminescence spectra were dispersed by a 0.4-m grating monochromator and recorded photoelectrically by digital accumulation of the signal at each point, with discrimination of the pulses of unsuitable energy (the stability of the excitation intensity within 10% was preserved). The measurements were carried out at room temperature ($T=295$ K).

Figure 1 presents some typical time-resolved luminescence spectra of CdS_{1-x}Se_x crystals for a few values of the composition parameter x . The dominant emission band under experimental conditions used (high temperature and high excitation level) exhibits features characteristic of a dense electron-hole-plasma radiative recombination.^{6,28,29} The high-energy wing of the luminescence band in the region $h\nu > E_g + k_B T_c + \Gamma$ (E_g is the band gap, Γ is the sum of the collision and the inhomogeneous broadening) is shaped by the carrier distribution function, and reflects the effective carrier temperature³⁰ $T_c(t) = -\{k_B \partial[\ln I_{\text{LUM}}(h\nu, t)]/\partial(h\nu)\}^{-1}$. As the magnitude of the sum broadening is generally unknown, the values of $T_c(t)$ were obtained in the far high-energy region, where the spectral shape on a semilogarithmic plot exactly matched a linear slope. The accuracy of the procedure was proved by the relaxation of the measured effective temperature strictly to its initial value (295 K), independently of the alloy composition, and by the insensitivity of the T_c transient behavior to plasma density in samples with different carrier lifetimes. It should be noted that the observed carrier heating does not refer to a trivial heating of the crystal, as indicated by the spectral position of the luminescence band.

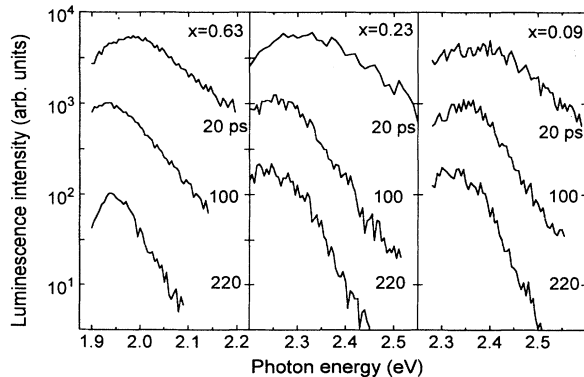


FIG. 1. Transient luminescence spectra of a few CdS_{1-x}Se_x mixed crystals. The delay time in respect of the exciting pulse is indicated. The spectra are arbitrarily shifted along the vertical axis.

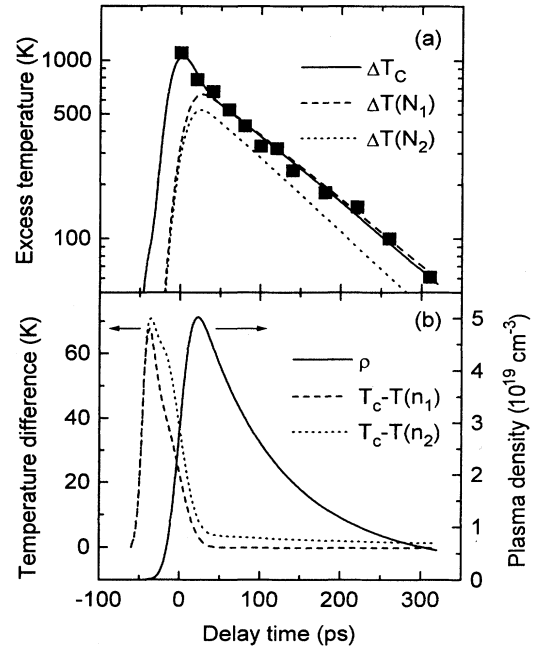


FIG. 2. (a) Temporal evolution of the excess electron effective temperature in a CdS_{0.77}Se_{0.23} crystal. Points, experiment; solid line, calculation. The dashed and dotted lines relate to the excess "temperature" of the second-generation nonequilibrium phonons of both constituents. (b) Calculated evolution of the electron-hole pair density (solid line) and the difference between the electron and the LO-phonon "temperatures" (dashed and dotted lines).

Points in Fig. 2(a) depict an experimental time evolution of the plasma excess temperature $\Delta T_c(t) = T_c(t) - T$ in a CdS_{0.77}Se_{0.23} sample. At zero delay, the excess temperature reaches a value of 1100 K due to redistribution of the photoexcitation excess energy $h\nu_L - E_g$ in plasma. Later, some part of the initial overheating decays rapidly in the first 20 ps following the shape of the pump pulse. This phase of the relaxation is apparently governed by decay of the nonequilibrium population of LO phonons. After initial cooling, a slow stage of the relaxation occurs, fitting an exponential law $\Delta T_c(t) = \Delta T_c(50 \text{ ps}) \times \exp(-t/\tau)$ with a time constant τ equal to 114 ps in the particular case of $x=0.23$. The slow stage of the relaxation is to be attributed to the depopulation of the second-generation nonequilibrium phonons¹⁴ produced during the rapid anharmonic decay of the nonequilibrium LO phonons, the latter considered to be that of the first generation.

The variation of the time constant τ with composition is shown by points in Fig. 3. With increasing x from 0 to 0.23, τ is seen to rapidly decrease from the initial value in pure CdS (320 ps) to 114 ps. Further, τ slowly increases up to the pure CdSe value of 130 ps. The asymmetric dip in the composition dependence of τ is apparently connected with the nature of phonon-phonon interaction in a two-mode vibration system of the alloy, and is to be carefully examined.

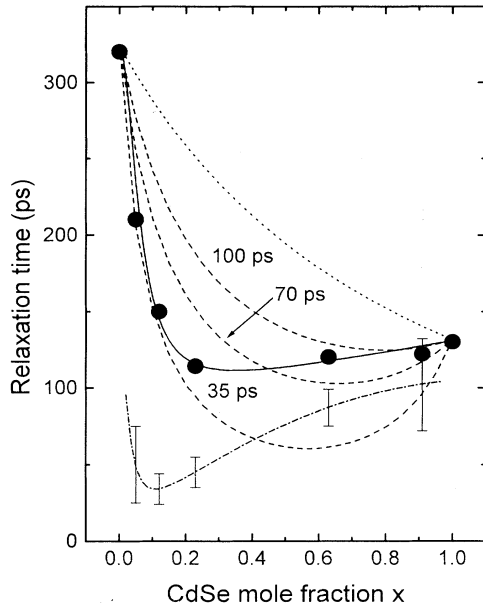


FIG. 3. Composition dependence of the time constant of the electron effective temperature slow relaxation in highly excited $\text{CdS}_{1-x}\text{Se}_x$ mixed crystals: points, experiment; solid line, approximation. The dotted line presents the results of calculation in case of the absence of cross relaxation; dashed lines show the same for the indicated constant values of the cross-relaxation time constant. The dash-dotted line shows the composition-dependent cross-relaxation time constant extracted from the approximated experimental curve.

To obtain more details about the relaxation process, the whole energy-transfer scheme is to be considered for a two-mode crystal. The proposed route of the electron excess energy transfer to the lattice is as follows. The laser pulse being in action, intercarrier collisions rapidly (within ~ 0.01 ps) redistribute the initial excess energy of photoexcited carriers in the quasithermalized system of electrons and holes considered as a plasma with a collective parameter T_c . The plasma is regarded as shared by both sublattices. As soon as T_c is brought above the equilibrium value T , the rate of the LO-phonon emission by carriers overcomes the rate of the absorption giving rise to nonequilibrium populations of phonons in both uncoupled branches of LO modes. The probability of exciting a certain branch is assumed to be weighted by the mole fraction of the corresponding constituent. The intravalley phonon emission characteristic of direct-gap semiconductors and the long-range nature of the polar electron-phonon interaction result in the concentration of nonequilibrium LO phonons in the vicinity of $\mathbf{k}=0$ for both LO branches. Further, the three-phonon anharmonic decay³¹ of a zone-center phonon with a small wave number q produces a pair of phonons with a nearly subharmonic frequency and almost opposite wave vectors \mathbf{Q}' and \mathbf{Q}'' , the condition $|\mathbf{Q}' - \mathbf{Q}''| \leq q$ holding.³² Hence the fragments of the nonequilibrium LO phonons are concentrated in a quasispherical layer of \mathbf{k} space with a

thickness q and an average radius Q , a second-generation nonequilibrium phonon population being built. The subharmonic frequencies of zone-center LO phonons in wurtzite-type CdS and CdSe crystals belong to the large-wave-number region of the B_1 optical-phonon branch.^{33,34} The optical modes in $\text{CdS}_{1-x}\text{Se}_x$ mixed crystals generally being treated as uncoupled oscillators throughout the whole Brillouin zone,³⁵ the two sorts of long-wavelength nonequilibrium LO phonons are expected to place their fragments only in the corresponding host subharmonic region. Due to large wave vectors, the second-generation nonequilibrium phonons relax to a wide spectrum of lower-frequency vibration modes of partially amalgamated³⁵ sublattices (the relaxation can be mediated by fusion of the second-generation nonequilibrium phonons to large-wave-vector LO phonons^{15,36} to a certain extent). Thus both host- and alien-induced relaxation is to be taken into account. Eventually, the energy of the second-generation nonequilibrium phonons being widely scattered in \mathbf{k} space, a small increase in lattice temperature is produced.

The most hazy stage of the relaxation, apparently responsible for the observed slow phase of effective-temperature decay, is the depopulation of the second-generation nonequilibrium phonons in the alloy. To quantitatively estimate the influence of alloying on the rate of establishing the thermodynamic equilibrium in highly excited crystals, a set of equations describing transient behavior of the electron and hole distribution functions and the occupation numbers of phonon modes is to be solved and fitted with the experimental data. An exact solution of the problem can be performed;¹⁵ however, it fits the experimental data with difficulty because of a large number of phonon modes involved with the exact form of the wave-vector and frequency dependencies of the anharmonic coupling coefficients still being unknown. Below, a simplified analysis on the basis of rate equations for the carrier effective temperature, plasma density, and averaged phonon-mode occupation numbers in the relevant regions of \mathbf{k} space is presented.

Supposing the effective temperature is high enough to preserve carrier degeneracy, its evolution is described by a rate equation containing the energy gain due to photo-generation of electron-hole pairs with an excess energy $h\nu_L - E_g(x)$ (the latter is assumed to be instantaneously redistributed in plasma by intercarrier collisions) and energy-loss rates which at elevated temperatures originate only from the polar LO-phonon emission

$$\frac{dT_c}{dt} = \frac{1}{3k_B} \left[G(t) \frac{h\nu_L - E_g(x) - 3k_B T_c}{\rho} - f_1 P_1(T_c, n_1) - f_2 P_2(T_c, n_2) \right]. \quad (1)$$

Here $G(t)$ is the volume carrier generation rate, ρ is the density of electron-hole pairs, and $f_1 = 1 - x$ and $f_2 = x$ are the mole fractions for CdS and CdSe, respectively. The electron-phonon energy-loss rates P_i ($i=1,2$) referred to one electron-hole pair depend generally on the carrier effective temperature and occupation numbers of

phonons interacting with electrons and holes. Here the occupation numbers are approximated by averaged values n_1 and n_2 corresponding to both branches of LO phonons in an appropriately estimated region of \mathbf{k} space. In this case the energy-loss rates for undegenerated carriers can be expressed in the usual way,³⁷ substituting the equilibrium LO-phonon occupation numbers by averaged nonequilibrium ones:

$$P_i(T_c, n_i) = \frac{2^{3/2}[\alpha_e(x) + \frac{1}{2}\alpha_h(x)](\hbar\omega_i)^{5/2}}{\hbar(\pi k_B T_c)^{1/2}} \times K_0(z_i)[(n_i + 1)y_i^{-1} - n_i y_i], \quad i = 1, 2. \quad (2)$$

Here ω_i is the LO-phonon frequency, $K_0(z)$ is the modified Bessel function for zero order, $z_i = \hbar\omega_i/k_B T_c$, $y_i = \exp(z_i/2)$, and $\alpha_e(x)$ and $\alpha_h(x)$ are the amalgamated polar interaction constants for electrons and holes, respectively. The hole contribution is multiplied by $\frac{1}{2}$ to take into account the overlap integral for p -like hole wave function.³⁸

The carrier density, contained in Eqs. (1) and (4) is given by a kinetic equation

$$\frac{d\rho}{dt} = G(t) - \frac{\rho}{\tau_c} - \gamma(T_c)\rho^2, \quad (3)$$

where τ_c is the carrier lifetime, and $\gamma(T_c)$ is the effective-temperature-dependent bimolecular recombination coefficient. In the absence of degeneration, the latter can be approximated as $\gamma(T_c) = \gamma(T)(T_c/T)^{-3/2}$, where $\gamma(T)$ refers to the lattice temperature.

Each of the two rate equations for LO phonons comprises a generation term proportional to the electron-energy-loss rate related to the relevant phonon branch and a three-phonon relaxation term³⁹

$$\frac{dn_i}{dt} = \frac{\rho P_i(T_c, n_i)}{\hbar\omega_i \xi_i} - \frac{n_i(N_i + 1)^2 - (n_i + 1)N_i^2}{\vartheta_i}, \quad i = 1, 2, \quad (4)$$

where ξ_i is the number of LO modes active in energy transfer per unit volume of the i th component, N_i is the averaged occupation number of the second-generation phonons, and ϑ_i is the zero-temperature LO-phonon lifetime with regard to the anharmonic decay.

The second-generation nonequilibrium phonons are assumed to concentrate within the regions with mode densities Ξ_i , thus the generation term in a rate equation is derived from the decay term of Eq. (4):

$$\frac{dN_i}{dt} = \frac{\omega_i \xi_i}{\Omega_i \Xi_i} \frac{n_i(N_i + 1)^2 - (n_i + 1)N_i^2}{\vartheta_i} - \frac{N_i - N_i^{(0)}}{\Theta_i(x)}, \quad i = 1, 2, \quad (5)$$

where $\Omega_i = \omega_i/2$ is the subharmonic frequency. The relaxation to an equilibrium occupation value $N_i^{(0)}$ is controlled by a composition-dependent depopulation time constant $\Theta_i(x)$. The shape of the dependence conceals the origin of the phonon-phonon interaction in a mixed

crystal.

The LO-phonon modes active in the energy transfer are contained in a quasispherical region centered at $\mathbf{k}=0$. The radius of the sphere is determined by the characteristic wave number q_i of LO phonons emitted by the heaviest carriers. Thus

$$\xi_i = (2\pi)^{-3} \frac{4\pi}{3} q_i^3, \quad (6)$$

where q_i is supposed to equal $(2m_{hi}\omega_i/\hbar)^{1/2}$, m_{hi} denoting the effective hole mass.

The considered LO-phonon decay route implies the second-generation nonequilibrium phonons to be accumulated in a quasispherical layer of \mathbf{k} space with the density of modes

$$\Xi_i = (2\pi)^{-3} 4\pi Q_i^2 q_i, \quad (7)$$

where q_i now is the thickness of the layer, i.e., the largest difference of wave vectors of LO-phonon fragments, and Q_i is the wave number of the subharmonic phonons averaged over directions.

It should be noted that a high excitation power is supposed to enable the exciting light to penetrate a sample deeper than in the case of usual absorption ($\sim 0.1 \mu\text{m}$), the electron states responsible for incident photon absorption not being capable of depleting within the electron-electron relaxation time ($\tau_{ee} \sim 10$ fs). The penetration depth can be estimated as $d = I_L \tau_{ee} / [h\nu_L g(h\nu_L - E_g)\delta(h\nu_L)]$, where $g(E)$ is the density of states (e.g., $d \sim 1 \mu\text{m}$, in the case of CdS). Under such conditions, diffusion of second-generation nonequilibrium phonons can be neglected on the relevant time scale.¹⁴ In addition, the first-generation nonequilibrium LO phonons are immobilized due to zero group velocity. This allows only the anharmonic decay to be taken into account when describing phonon relaxation in Eqs. (4) and (5).

The system of Eqs. (1) and (3)–(5) was solved numerically, using the following parameters for CdS ($i=1$) and CdSe ($i=2$): $E_{g1,2} = 2.49$ and 1.75 eV; $\hbar\omega_{1,2} = 38$ and 26 meV; $m_{h1,2} = 1.14m_0$ and $0.47m_0$; $\alpha_{e1,2} = 0.63$ and 0.48 ; and $\alpha_{h1,2} = 1.5$ and 0.90 . The amalgamated values of parameters $b(x) = E_g(x)$, $\alpha_e(x)$, and $\alpha_h(x)$ were fixed as $b(x) = (1-x)b_1 + xb_2$. The recombination parameters τ_c and $\gamma(T)$ were estimated from the luminescence-intensity decay kinetics. The values of 10^{-9} s and 10^{-9} cm³s⁻¹, respectively, were found to be appropriate in the whole range of x . The values of subharmonic-phonon wave numbers were obtained from the available data on the lattice dynamics of CdS and CdSe. In CdS,³³ Q_1 equals 2×10^7 cm⁻¹ in the A direction, 7×10^7 cm⁻¹ in the K direction, and there are no subharmonic phonons in the third principal (M) direction. Thus the averaged value of Q_1 was estimated to equal 4×10^7 cm⁻¹. In CdSe,³⁴ the B_1 optical branch is investigated only in the A direction, yielding a value of 2×10^7 cm⁻¹ at the subharmonic frequency. As the phonon-dispersion curves in CdS and CdSe are considered to be very similar,³⁵ Q_2 was assumed also to be equal to 4×10^7 cm⁻¹.

The carrier generation rate was taken in the Gaussian

form

$$G(t) = \frac{I_L}{h\nu_L d} \left[\frac{\beta}{\pi} \right]^{1/2} \exp \left[-\beta \left[\frac{t}{\tau_L} \right]^2 \right], \quad (8)$$

where d is the thickness of the excited region, and $\beta = 4 \ln(2) \approx 2.77$.

The second-generation phonon lifetime was assumed to have a form of

$$\Theta_i(x) = \left[\frac{f_i}{\Theta_i} + \frac{f_j}{\Theta_{ij}} \right]^{-1}, \quad i, j = 1, 2, \quad j \neq i. \quad (9)$$

The first term in parentheses on the right-hand side of Eq. (9) accounts for the relaxation in the host sublattice, while the second occurs due to relaxation in the alien sublattice or in the amalgamated acoustical-phonon modes. Below, the cross-relaxation time constants Θ_{12} and Θ_{21} are assumed to be equal.

The solid line in Fig. 2(a) depicts the result of the calculation for T_c in a sample with $x = 0.23$. The fitting parameters $d = 4.5 \mu\text{m}$, $\vartheta_1 = \vartheta_2 = 3.5 \text{ ps}$, and $\Theta_{12} = \Theta_{21} = 45 \text{ ps}$ were used (the values of $\Theta_1 = 300 \text{ ps}$ and $\Theta_2 = 115 \text{ ps}$ were taken from the fitting the calculated kinetics with the experimental ones in end-point crystals). The slope of the calculated curve in the slow-relaxation region is sensitive basically to the alien-induced relaxation time, while the peak temperature and the vertical position on a semi-logarithmic plot were adjusted with d and ϑ parameters. The carrier-recombination parameters τ_c and $\gamma(T)$, as well as the polar-interaction constants, weakly affect the calculated effective-temperature kinetics. The obtained values of the excitation depth ($d \sim 1 \mu\text{m}$ in various samples) corroborate the assumption that the nonequilibrium phonon population is built into the surface layer enhanced over the conventional exciting-light penetration depth.

Figure 2(a) also depicts the calculated temporal behavior of the second-generation-phonon temperatures. The temperatures are derived from the Bose functions of the nonequilibrium occupation numbers as $T(N_i) = \hbar\Omega_i / [k_B \ln(1 + N_i^{-1})]$. In this notation, the energy-transfer direction can easily be determined. In the particular case of $x = 0.23$, the condition $T(N_1) > T_c > T(N_2)$ is held in the slow-relaxation region. This means that the excitation pulse being terminated, the energy is concentrated in CdS-like second-generation nonequilibrium phonons as they employ more modes in comparison with CdSe-like ones. These phonons relax directly through an anharmonic process, as well as through LO-phonon and plasma-mediated excitation of CdSe-like subharmonic phonons. Figure 2(b) show that the temperatures of both branches of LO phonons $T(n_i) = \hbar\omega_i / [k_B \ln(1 + n_i^{-1})]$ differ significantly from the plasma temperature only at the onset of the excitation, when the energy flow to the electron system and the carrier density are increasing with time. In the slow-relaxation region the differences between the electron effective temperature and the LO-phonon temperatures are negligible, the condition $T(n_1) > T_c > T(n_2)$ holding.

The solid line in Fig. 2(b) depicts the calculated temporal behavior of the electron-hole pair density ρ . The densities of the order 10^{19} cm^{-3} should cause degeneration of electrons, the holes still being undegenerated. However, the carrier diffusion being neglected, the calculated densities are to be considered as overestimated, and the applicability of the Maxwell-Boltzmann carrier distribution functions implied in Eqs. (1) and (2) seems to be fairly good.

Dashed lines in Fig. 3 present the calculated dependences of the effective-temperature relaxation time on composition for constant values of the cross-relaxation time Θ_{12} . The dotted line illustrates the case of the absence of cross relaxation, the second-generation phonons being assumed to reside and relax only in the host sublattice [$\Theta_i(x) = \Theta_i$ in Eq. (5)]. None of the calculated curves is seen to account for the experimental data. Meanwhile, the intersections of the experimental curve with the calculated ones enable us to simulate a composition dependence of Θ_{12} (dash-dotted line in Fig. 3). This dependence implies a minimum near $x = 0.1$ which corresponds to an asymmetric composition dependence of structure-fluctuation parameters in $\text{CdS}_{1-x}\text{Se}_x$, e.g., the exciton linewidth.²⁶ Apparently, the second-generation nonequilibrium phonons, their wavelengths being of a few interatomic distances, decay more effectively in a disordered lattice. In such a case, Fig. 3 provides indications that the disorder is capable of doubling the cross-relaxation rate in a ternary alloy. This is in contrast to plasma-produced LO phonons, which are less sensitive to an alloy disorder²⁰ due to large wavelength.

In conclusion, an investigation of the electron-hole plasma effective-temperature relaxation in highly excited $\text{CdS}_{1-x}\text{Se}_x$ mixed crystals revealed a composition-dependent slow cooling stage. A two-route energy relaxation scheme was proposed, assuming that both first- and second-generation nonequilibrium phonons are uncoupled in a persistent-type $\text{CdS}_{1-x}\text{Se}_x$ alloy. In accordance with this scheme, the observed transient behavior of the effective temperature can be described by solving a system of six rate equations for the effective temperature, the carrier density, and the occupation numbers for two branches of LO phonons and two branches of subharmonic phonons. Soon after terminating the excitation pulse, the effective-temperature relaxation is governed by the decay of two competing populations of the second-generation phonons that are thermally linked through the LO phonons and the electron-hole plasma. The analysis of the composition dependence of the electron effective-temperature relaxation time indicated an enhancement of the depopulation rate of the second-generation nonequilibrium phonons near $x = 0.1$. The obtained asymmetric composition dependence of the relevant cross-relaxation time is supposed to originate from the composition-fluctuation-induced depopulation of the short-wavelength second-generation nonequilibrium phonons.

A concept of second-generation nonequilibrium phonons, their relaxation time being sensitive to inhomogeneity and, possibly, to reduced dimensionality of a semiconductor object, may support the understanding of

hot-carrier cooling in quantum wells and related structures. Hindering the electron-energy relaxation by the second-generation phonons was observed in a GaAs-Al_xGa_{1-x}As superlattice under steady-state conditions,⁴⁰ and is to be tested in a transient experiment.

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