

Excitation spectroscopy of the electron-hole plasma in cadmium selenide single crystals: hot electrons and phonons

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(Submitted 20 May 1986)

Zh. Eksp. Teor. Fiz. **91**, 1909–1916 (November 1986)

The effects of concentration and energy factors on the broadening of the recombination emission band for a nonequilibrium electron-hole plasma can be studied separately by using a tunable laser to excite the semiconductor crystal. Luminescence spectra recorded at resonance (laser energy $h\nu_0 \approx E_g$, where E_g is the band gap) and at energies $h\nu_0 > E_g$ sufficient to heat the electron-hole plasma are compared for a CdSe crystal, and the broadening of the band found to be due not to multiparticle effects but rather to transfer of the excess photoexcitation energy $h\nu_0 - E_g$ in the plasma. The shortwave side of the band is broadened due to the laser heating of the plasma, while the longwave side is broadened because energy relaxation in the hot plasma causes the longitudinal-optical (LO) phonon system to deviate from equilibrium. The LO vibrational amplitude spectrum is calculated theoretically by solving the balance equations for the plasma energy and for the number of noninteracting LO phonons. The spectra are in good agreement with the experimentally determined temperature of the "hottest" LO phonon modes (400 K for an excitation density of $6 \cdot 10^{24}$ photons \cdot cm $^{-2}$ s $^{-1}$, $h\nu_0 - E_g = 0.4$ eV, and plasma and lattice temperatures equal to 450 and 295 K, respectively).

1. INTRODUCTION

Experimental studies of nonequilibrium electron-hole plasmas in strongly excited direct-gap semiconductors have customarily been based on analyzing the time behavior of the luminescence spectrum while the pump power is increased. Attempts to explain the change in the recombination emission spectra for strongly excited semiconductors have led to new ideas regarding the nature of the inhomogeneous band broadening for nonequilibrium electron-hole plasmas. In particular, we may mention models in which the excess carriers are heated due to the excess excitation energy,¹ models in which the phonon² and plasmon³ subsystems are excited, models in which the emission spectrum is broadened due to excess carrier degeneracy and superluminescence,⁴ models involving multiparticle interaction,^{5,6} and in particular models in which the quasimomentum selection rules are violated for interband transitions.^{7,8} It should be noted that the specific nature of the plasma processes in strongly excited semiconductors is essentially determined by two factors: 1) the rapid rate of carrier generation (concentration effects); 2) the large energy fluxes between the quasiparticle subsystems in the crystal (energy effects). The energy effects can be identified experimentally by using tunable lasers as the excitation sources.^{9–15} By contrast, resonant ("cold") excitation of the excess carriers is required in order to study pure concentration effects. Nevertheless, no systematic definitive method has yet been developed for analyzing the inhomogeneous broadening of the luminescence band from electron-hole plasmas which is based on comparing experimental lineshape data with results found by solving the balance equations for the energy and the number of nonequilibrium quasiparticles. The purpose of this paper is to develop such an approach which combines excitation

spectroscopy with a quantitative analysis of the energy transfer processes in CdSe, a polar semiconductor.

2. EXPERIMENT

A freshly cleaved CdSe single crystal grown from a melt was excited by pulses from a tunable dye laser at energies $h\nu_0 = 1.75$ – 2.15 eV (the line width was 0.3 meV, the pulse length was 10 ns, and the repetition rate was 12.5 Hz). Ethanol solutions of the dyes rhodamine 6G, rhodamine B, phenylamine, and nile blue + rhodamine B were pumped at the second harmonic of a YAG:Nd³⁺ laser, and the light incident on the crystal was polarized with E perpendicular to c, the optic axis of the crystal. We used a JY HRD1 double monochromator, an FEU-106 photomultiplier tube, and a strobing integrator to analyze the luminescence spectra. A personal computer monitored the measurement and correction of the distortions introduced by variations in the spectral characteristic of the monochromator-photomultiplier system.

The experiment was carried out at a temperature $T = 295$ K high enough to ensure that a large fraction of the excitons in the CdSe were ionized. Figure 1 shows some luminescence spectra for CdSe excited to various degrees by laser light with $h\nu_0 = 2.15$ eV. The exponential falloff in both directions from the peak is characteristic. As the excitation intensity I increases, both slopes become less curved and the halfwidth of the band increases [from 70 to 105 meV when I increases from $2 \cdot 10^{23}$ to $6 \cdot 10^{24}$ photons/(cm 2 ·s)]. This behavior is typical for emission from electron-hole plasmas in polar semiconductors and can be accounted for by various alternative models based on either the energy or the concentration approaches. However, there is a simple experimental technique that can be used to distinguish the en-

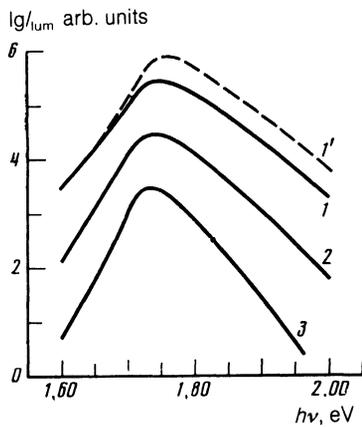


FIG. 1. Luminescence spectra for a CdSe single crystal (E1c) with $h\nu_0 = 2.15$ eV and pump intensity $I = I_0 = 6 \cdot 10^{24}$ photons/(cm²·s) (1), $I = 0.24I_0$ (2), $I = 0.03I_0$ (3); 1') spectrum 1 corrected for reabsorption. The intensity scale for each spectrum is shifted arbitrarily.

ergy and concentration effects in strongly excited crystals.⁹ This involves exciting the plasma resonantly (zero excess energy, $h\nu_0 \approx E_g$), changing the intensity so as to vary the excess carrier generation rate, and examining which changes in the luminescence spectrum are caused exclusively by the increase in the concentration of excess electrons and holes. Figure 2 shows luminescence spectra for CdSe recorded for "cold" excitation ($h\nu_0 = 1.75$ eV). We see that increasing the excess carrier concentration alone has almost no effect on the form of the spectrum—a 30-fold increase in I broadens the band from 62 to 65 meV (by just 5%); the slopes of the low- and high-energy sides are not changed and are equal to the corresponding slopes obtained at relatively low intensities I using light with $h\nu_0 > E_g$ (curve 3, Fig. 1). It follows that the spreading of the high- and low-energy portions of the line due to a transfer of excess photoexcitation energy in the crystal is primarily responsible for broadening the emis-

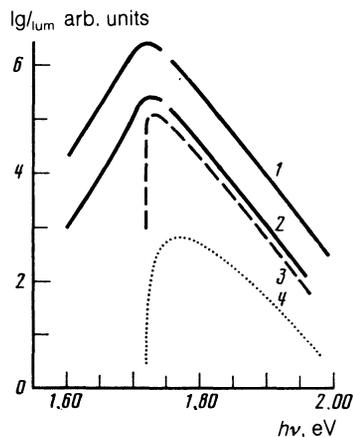


FIG. 2a,b. As in Fig. 1, but for $h\nu_0 = 1.75$ eV: $I = I_0$ (1), $I = 0.03I_0$ (2); (3, 4) show the theoretical luminescence spectra for an electron-hole plasma with $T_e = 295$ K and $\bar{E}_g = 1.72$ eV and the quasimomentum selection rules for interband transitions are (3) and are not obeyed (4).

sion band of the nonequilibrium electron-hole plasma at high levels of "heating" excitation.

The decrease in the slope of the high-energy limb can be attributed to an increase in the effective temperature T_e of the excess carriers.¹ To deduce T_e accurately from the slope parameter $T^* = -[k_B \partial(\ln I_{lum}) / \partial(h\nu)]^{-1}$ ($h\nu > E_g$), one must analyze the spectral distortions caused by reabsorption and rigorously determine the functional dependence $T_e(T^*)$. As an illustration, curve 1' in Fig. 1 shows an emission line reconstructed with allowance for the absorption spectrum of the CdSe crystal¹⁶ (the excited region was assumed to extend to a depth of 1 μ m). We see that the spectral line is uniformly suppressed for $h\nu > E_g$; however, the reabsorption does not alter the slope of the high-energy limb. The theoretical lineshape for emission from an electron-hole plasma

$$I_{lum}(h\nu) \propto (h\nu - \bar{E}_g)^{\gamma} \exp(-(h\nu - \bar{E}_g)/k_B T_e), \quad (1)$$

can be used to relate T^* , which characterizes the decay, to the effective temperature T_e . Here \bar{E}_g is the renormalized band gap and $\gamma = 1/2$ in the extreme case when the interband transitions obey the quasimomentum selection rules ($\gamma = 2$ in the other extreme case when these rules are completely violated).⁸ Figure 2 compares the experimental spectra for a "cold" plasma ($T_e = T$, curves 1, 2) with the theoretical lineshapes for $\gamma = 1/2$ (curve 3) and $\gamma = 2$ (curve 4). We see that the k-selection rule is obeyed in this case. The effective temperature T_e in our experiment can thus be calculated by neglecting the reabsorption and setting $\gamma = 1/2$:

$$(T_e)^{-1} = (T^*)^{-1} + k_B [2(h\nu - \bar{E}_g)]^{-1}. \quad (2)$$

Figure 3 shows how T_e depends experimentally on the laser photon energy at the peak pump intensity [the value $\bar{E}_g = 1.72$ eV used in Eq. (2) was obtained by comparing the experimental and theoretical spectra].

The theoretical band (1) for the electron-hole plasma does not describe the low-energy side of the experimental spectra. Nevertheless, it is obvious that the broadening here is also due to energy effects—indeed, for resonant excitation

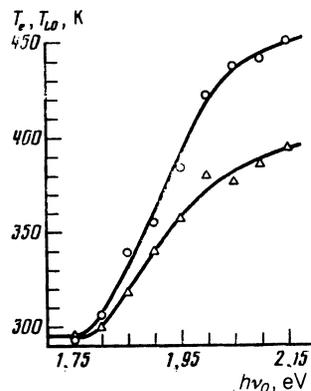


FIG. 3. Experimental dependence of the effective excess carrier (O) and LO-phonon (Δ) temperature as functions of the laser photon energy for CdSe, $I = I_0$.

($h\nu_0 = 1.75$ eV), the parameter $T^{**} = [k_B \partial(\ln I_{lum}) / \partial(h\nu)]^{-1}$ ($h\nu < 1.7$ eV) characterizing the exponential low-energy decay is equal to 260 K regardless of the excitation level, while for $h\nu_0 = 2.15$ eV it is sensitive to I and reaches 350 K when $I = 6 \cdot 10^{24}$ photon/(cm²·s) (curve 1, Fig. 1). The exponential decay at low energies $h\nu < E_g$ could be due to the interaction of the excess carriers with LO phonons. Indeed, it was shown in Ref. 17 that the Urbach rule¹⁸ is reversible and applies not only to light absorption but also to interband recombination of excess carriers. Calculations of the low-energy wing of the band for CdSe revealed that the expression

$$I_{lum}(h\nu) \sim \exp(\sigma(h\nu - E_g)/k_B T_{LO}) \quad (3)$$

is valid for $T_{LO} \lesssim T_e$; here the parameter σ is nearly equal to the ratio $T/T^{**} = 1.13$ for $T^{**} = 260$ K (when the electron-hole plasma was not heated). The "temperature" of the LO phonons is thus given by $T_{LO} = \sigma T^{**}$. Figure 3 plots the resulting dependence $T_{LO}(h\nu_0)$ at maximum excitation intensity. We see that T_{LO} lags behind the plasma temperature and reaches 400 K when $h\nu_0 = 2.15$ eV. However, this explanation for the broadening of the low-energy limb of the emission band requires additional support, such as could be provided by quantitatively calculating the occupation numbers for the LO lattice vibration modes which are amplified by relaxation of the excess energy $h\nu_0 - E_g$. A further discussion of the physical significance of the measurable temperature T_{LO} is also desirable.

3. THEORY

We will now calculate the spectrum of the excess LO phonons under the assumption that there is no interaction within the phonon branch, i.e., that the wave vector \mathbf{q} of an excess phonon remains constant until the moment of decay. The calculation will be carried out for a nondegenerate, isotropic hot electron-hole plasma in which the excess phonon generation rate depends only on the magnitude q of the wave vector. We neglect the dispersion of the LO phonons and the plasmon-phonon interaction; the energy of the coupled plasmon-phonon mode is assumed to differ little from the energy of an LO phonon, while the plasmon component of the energy is ultimately converted into kinetic energy of the excess carriers.¹⁹ The probability for Frölich scattering of carriers of type i by an LO phonon with wave vector \mathbf{q} is²⁰⁻²²

$$p_i^{\pm} = \frac{\alpha_i K_i (\hbar\omega_{LO})^{3/2}}{\hbar q E^{3/2}} \left[\frac{q^2}{q^2 + q_D^2} \right]^2 \left(N_q + \frac{1}{2} \pm \frac{1}{2} \right), \quad (4)$$

where $i = e, h$ for electrons and holes, respectively, and the $+$ ($-$) sign corresponds to emission (absorption) of a phonon; E is the kinetic energy of the carrier, K_i is the overlap function ($K_e = 1, K_h = 0.5$),

$$\alpha_i = e^2 m_i^{1/2} (\kappa_{\infty}^{-1} - \kappa_0^{-1}) / 2^{3/2} \pi \epsilon_0 \hbar^{3/2} \omega_{LO}^{1/2}$$

is the polar constant, and κ_{∞} and κ_0 are the high-frequency and static values of the relative dielectric permittivity; $\hbar\omega_{LO}$ is the energy of the LO phonon, m_i is the effective carrier mass, and ϵ_0 is the permittivity of free space. The factor in square brackets reflects the screening of the electron (hole)-

phonon interaction in the static limit of the random phase approximation,¹¹ and $q_D = e(2n/\epsilon_0 \kappa_{\infty} k_B T_e)^{1/2}$ is the reciprocal of the Debye screening length. The occupation number for phonons q is

$$N_q = [\exp(\hbar\omega_{LO}/k_B T_q) - 1]^{-1}, \quad (5)$$

and the problem is to find the mode "temperature" T_q . In the quasi-stationary case, the generation rate G_q for LO phonons in a hot electron-hole plasma is equal to the relaxation rate of their nonequilibrium population. Per unit volume of crystal, the latter is equal to²⁴

$$R_q = 4\pi q^2 (N_q - N^0) / (2\pi)^3 \tau', \quad (6)$$

where τ' is the relaxation time for the excess population and N^0 is the equilibrium occupation number (for $T_q \equiv T$). The rate G_q is equal to the difference between the rates of phonon emission and absorption by the electron-hole pairs:

$$G_q = \sum_i \left[\int_{\epsilon_i + \hbar\omega_{LO}}^{\infty} p_i^+ g_i(E) f_i(E) dE - \int_{\epsilon_i}^{\infty} p_i^- g_i(E) f_i(E) dE \right], \quad (7)$$

where $\epsilon_i = (\hbar^2 q^2 - 2m_i \hbar\omega_{LO})^2 / (8\hbar^2 q^2 m_i)$ is the minimum kinetic energy for a carrier capable of absorbing a phonon q , and $g_i(E)$ and $f_i(E)$ are the state density and the excess carrier distribution function, respectively. If we assume a Boltzmann energy spectrum, the condition $G_q = R_q$ gives the following spectrum for the noninteracting excess LO phonons:

$$N = \frac{N^0 + n\tau' F_q \exp(-\hbar\omega_{LO}/k_B T_e)}{1 + n\tau' F_q [1 - \exp(-\hbar\omega_{LO}/k_B T_e)]}, \quad (8)$$

where

$$F_q = \frac{4(\pi\hbar\omega_{LO})^{3/2} q}{(q^2 + q_D^2)^2 \hbar (k_B T_e)^{3/2}} \sum_i \alpha_i K_i \exp\left(-\frac{\epsilon_i}{k_B T_e}\right). \quad (9)$$

The parameters T_e , n , and τ' must be known before (8) can be used to calculate the excess phonon spectrum. We showed above that T_e can be determined experimentally. We will use the power balance equation for a hot electron-hole plasma to calculate the excess carrier concentration n in the "hot" region of the crystal (extending to a depth $1/\mu$ equal to the penetration distance of the laser light,²⁵ where μ is the refractive index). The specific excitation power (per unit volume of plasma) for light with $h\nu_0 > E_g$ is equal to $\mu I / (h\nu_0 - \bar{E}_g - 3k_B T_e)$, where the factor in parentheses is the initial excess energy, including the average kinetic energy of a thermalized electron-hole pair.²⁾ Under quasi-stationary conditions, this power is equal to the power of the electron (hole)-phonon losses, which can be calculated by integrating the function $\hbar\omega_{LO} G_q$ over the entire Brillouin zone. In view of the foregoing, the excess carrier concentration is determined by the power balance equation

$$\mu I (h\nu_0 - \bar{E}_g - 3k_B T_e) = n \hbar\omega_{LO} \int_0^{\pi/4} F_q \frac{q^2}{2\pi^2} \left[(N_q + 1) \exp\left(-\frac{\hbar\omega_{LO}}{k_B T_e}\right) - N_q \right] dq, \quad (10)$$

where a is the lattice constant.

Since direct measurements of τ' for CdSe are unavailable, we will calculate τ' from the known dephasing time τ for the LO phonons. Measurements of the damping rate for an incoherent system of excess LO phonons in polar semiconductors show that $\tau' \approx \tau$ (Ref. 26). We thus conclude that the dephasing and population relaxation rates for LO phonons are governed by the same processes, namely three-phonon interactions $LO \leftrightarrow LA' + LA''$ (Ref. 27). The time τ' depends on the difference between the generation and annihilation rates for the LO phonons and is equal to²⁸

$$\tau' = [C_3(N' + N'' + 1)]^{-1}, \quad (11)$$

where N' and N'' are the occupation numbers for the first and second LA phonons, respectively, and C_3 is the interaction constant for interaction of an LO phonon with two longitudinal acoustic phonons. The dephasing time for the LO phonon also depends on C_3 ,

$$\tau = [C_3(N' + 1)(N'' + 1)]^{-1}. \quad (12)$$

If we substitute (12) into (11) and recall that longwave LO phonons ($q \approx 0$) interact with LA phonons whose frequency is close to $\omega_{LO}/2$, we have

$$\tau' = \tau(N^0 + 1) \quad (13)$$

for the population relaxation time. For a CdSe crystal at room temperature $\tau = 0.7$ ps (Ref. 29), and (13) yields $\tau' = 1.1$ ps.

The excess LO phonon spectrum found by solving the system (8), (10) is shown in Fig. 4. The calculations assumed $I = 6 \cdot 10^{24}$ photons/(cm²·s), $h\nu_0 = 2.15$ eV, and $T_e = 450$ K, and the following parameter values for the CdSe crystal were used: $\hbar\omega_{LO} = 26$ meV, $\kappa_0 = 9.6$, $\kappa_\infty = 6.2$ (Ref. 29), $m_e = 0.13m_0$, $m_h = 0.8m_0$ (Ref. 30),

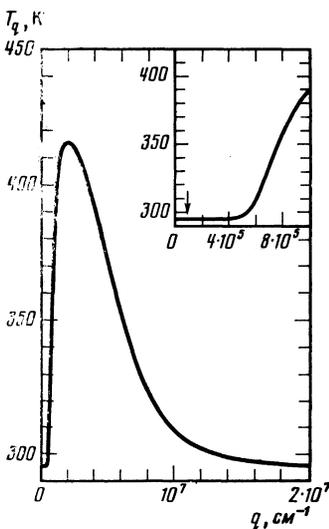


FIG. 4. Calculated spectrum for the excess LO phonons, $h\nu_0 = 2.15$ eV, $I = I_0$, $T_e = 450$ K. In the insert showing the small- q region, the arrow indicates the maximum wave number for a phonon capable of participating in first-order Raman light scattering for a probing light beam with $h\nu_0 = 2.15$ eV.

$\mu = 4 \cdot 10^4$ cm⁻¹ (Ref. 16). The excess carrier concentration $n = 3.8 \cdot 10^{17}$ cm⁻³ was also calculated.

4. DISCUSSION

Figure 4 shows that the excess LO phonons are concentrated in a narrow interval at the center of the Brillouin zone ($q_{\max} = \pi/a \approx 6 \cdot 10^7$ cm⁻¹). The maximum calculated mode temperature was 415 K for $q = 2 \cdot 10^6$ cm⁻¹, in good agreement with the value $T_{LO} = 400$ K deduced from the low-energy wing of the luminescence band for the electron-hole plasma (Fig. 1, curve 1). We note that this wing receives contributions from the entire ensemble of LO modes, whose populations are nonuniform both in momentum space and when considered as a function of depth in a crystal subjected to single-photon excitation. However, by adding the contributions from all the modes to the emission spectrum for energies $h\nu > E_g$, one can show that if $h\nu$ is large enough, the shape of the low-energy limb is determined by the temperature (occupation number) of the hottest modes—the luminescence component from the interaction with these modes decays the most slowly as $h\nu$ decreases. The observed broadening of the low-energy wing of the emission band thus reflects the amplification of a subgroup of the LO vibrations in a strongly excited crystal, and the measured temperature T_{LO} is in fact close to the maximum mode temperature. The agreement between the experimental and theoretical results lends support to the method proposed in Ref. 17 for analyzing excess LO phonons in terms of the luminescence spectra. This method can be used to study the heating of the optical phonons in crystals for which the Raman light scattering technique is unsuitable, and in many cases it may be indispensable. The insert to Fig. 4 illustrates such a situation for CdSe which is characteristic for most II–VI compounds. The arrow indicates the maximum wave number of an LO phonon capable of participating in first-order Raman light scattering. We see that the Raman-active LO phonon modes are not heated in this case.

The rate of energy loss from the electron-hole plasma drops appreciably when the temperature gap between the plasma and the LO phonons becomes narrower, even when the narrowing is confined to a small wavelength interval in momentum space corresponding to lower energies. Indeed, Fig. 4 shows that in this case the power loss per electron-hole pair is just 66% of the value calculated by neglecting the phonon heating. One should also bear in mind that excess LO phonons can be generated in a hot electron-hole plasma only if the gap $T_e - T_q$ is positive, as was the case in the experiment (Fig. 3). In this connection we note that further and more precise work (using excitation spectroscopy, in particular) is needed in order to analyze the situation observed previously in CdS (Ref. 2) and ZnS (Ref. 31), where the form of the low-energy edge of the luminescence spectrum for the electron-hole plasma corresponded formally to the case $T_{LO} > T_e$.

To summarize, excitation spectroscopy has enabled us to obtain for the first time experimental data both on excess carrier heating and on the deviation of the LO phonon system from equilibrium. The results are found to be mutually

consistent when interpreted using a model for energy transfer in a strongly excited polar semiconductor.

¹⁾This is justified because $\hbar\omega_{LO} < k_B T_e$ (Ref. 23).

²⁾We assume here that all of the initial excess photoexcitation energy is used to heat the electron-hole plasma, i.e., the excess carrier concentration is quite large.

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Translated by A. Mason