

Heating of photoexcited electron-hole plasma in II-VI compounds

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We investigate the heating of a nonequilibrium electron-hole plasma (EHP) that emits in the spectral region $h\nu > E_g$, in the crystals ZnTe, ZnSe, ZnS, CdTe, CdSe, and CdS in the case of high-power surface photoexcitation ($T = 77$ K). It is observed that with increasing excitation level the effective temperature of the nonequilibrium carriers saturates. A quantitative analysis of the power balance equation with allowance for the electron (hole)-phonon, electron-electron (hole), and electron-plasmon interactions, as well as the appearance of a characteristic anisotropic photoheating of the EHP in oriented CdSe and CdS crystals leads to the conclusion that at excitation densities on the order of 10^{24} cm $^{-2}$ ·sec $^{-1}$ the hot EHP is localized at the depth of penetration of the exciting light into the crystal.

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1. INTRODUCTION

It is known that excitation of semiconducting materials by light with quantum energy $h\nu_0$ greatly exceeding the band gap E_g leads to formation of a system of nonequilibrium carriers (NEC), whose effective temperature T_e can greatly exceed the lattice temperature T (Ref. 1). An investigation of the heating of photoexcited NEC is of great interest for the study of the mechanisms of energy exchange and of the spatial structure of an ensemble of nonequilibrium quasiparticles, and also for the determination of the regions of localization of the energy fluxes. One of the principal methods of investigating hot NEC is the study of luminescence in the spectral region $h\nu > E_g$. By now, hot photoexcited NEC were observed in a number of semiconducting materials.²⁻⁷ The dynamics of the heating of the NEC and the sensitivity of their effective temperature to the value of the quantum energy of the exciting light⁸⁻¹⁰ have made it possible to establish that the source of the heating of the NEC is the initial excess energy of the photoexcited electron-hole (e-h) pair ($h\nu_0 - E_g$), and the relaxation of the energy of the hot electron-hole plasma (EHP) is due to emission of longitudinal optical (LO) phonons. It is assumed here that the "hot" distribution of the NEC covers the entire excited region of the crystal (the region of ambipolar diffusion), and that the energy exchange is effected only via electron-phonon and electron-electron interactions.¹ A quantitative analysis of the power-balance equations of a hot EHP has shown, however, that in polar semiconductors the nonequilibrium e-h pair whose average energy corresponds to a Maxwellian distribution with effective temperature 200–400 K, transfers to the "cold" lattice an energy equal to the initial excess ($h\nu_0 - E_g$) within a time of the order of 10^{-12} sec, i.e., within a time much shorter than the lifetime (which for most II-VI semiconductors is of the order of 10^{-9} – 10^{-10} sec). In addition, in the case of CdS, which has an anisotropic absorption coefficient, the effective temperature of the NEC turned out to be sensitive to the polarization of the exciting light.⁵ Therefore the model of the photoexcited hot EHP uniformly heated over the entire ambipolar-diffusion length calls for a radical review.

In the present paper, to construct the power-balance equation for the hot EHP and to interpret the experimental data on the photo-heating of NEC in a number of II-VI compounds, we invoke, for the first time ever, to the model of localization of hot NEC at high excitation levels only in the region where the exciting light penetrates into the crystals.

2. THEORY

We assume that the hot EHP excited by strongly absorbing light is localized on the surface of the crystal on a small length l_{hot} , and the region of ambipolar diffusion is filled by partially ionized exciton gas. To calculate the hot length l_{hot} we turn to an analysis of the power-balance equation for a nondegenerate EHP, assuming satisfaction of the density criteria needed to make the intrinsic effective temperature T_e different from the lattice temperature.¹ A quantitative analysis of the power-balance equation calls for taking into account, besides the electron-phonon and electron-electron interactions, also of the hole-phonon energy losses, as well as of the role of the electron-hole and electron-plasmon energy exchange in the distribution of the excess energy in the EHP. In this case the temperature T_e is determined by the equation¹

$$\frac{I(h\nu_0 - E_g)}{l_{\text{hot}} h\nu_0 (n_{\text{hot}} + n_c^*)} = P_{eh}(T_e), \quad (1)$$

where I is the density of the power of the exciting light, n_{hot} is the density of the e-h pairs in the hot region, n_c^* is the critical density at which half of the initial excess energy is transferred to the Maxwellized EHP via interaction between the carriers (the remaining part of the excess energy is lost to cascade emission of LO phonons). The right-hand side of (1) is equal to the rate of the polar-optical energy losses for the e-h pair, the average energy of which corresponds to a distribution with effective temperature T_e (Refs. 11 and 12). Taking into account the properties of the hole-phonon interactions¹³, we have for the power of the polar-optical losses

$$P_{eh}(T_e) = \frac{q^2(k\theta)^{3/2} M_p^{1/2} (1/\kappa_\infty - 1/\kappa_0)}{2\pi^{3/2} \hbar^2 \epsilon_0} \left(\frac{\theta}{2T_e} \right)^{1/2} \times \frac{K_0(\theta/2T_e) \text{sh}(\theta/2T - \theta/2T_e)}{\text{sh}(\theta/2T)}, \quad (2)$$

where q is the electron charge, $k\theta$ is the energy of the LO phonon, κ_∞ and κ_0 is a modified Bessel function of order zero. Here

$$M_p^{1/2} = m_e^{1/2} + \frac{m_{hh}^2 + m_{lh}^2 e^{-\Delta/\hbar T_e}}{2(m_{hh}^{3/2} + m_{lh}^{3/2} e^{-\Delta/\hbar T_e})}, \quad (3)$$

where m_e , m_{hh} , and m_{lh} are respectively the masses of the electron and of the holes of the first and second subband; Δ is the splitting between the hole subbands at the point $k = 0$ (it is equal to zero for cubic compounds). The contribution of the third subband, which is usually greatly separated, has been omitted.

The critical density n_c^* will be obtained from the condition of equality of the powers of the polar-optical and plasma scattering by the initial excess energy of the photoexcited $e-h$ pair, the bulk of which is acquired at $m_h \gg m_e$ by the electron. The power of the plasma scattering, which heats the EHP, of the initial energy of the electron $E_e = (\hbar\nu_0 - E_g)(1 + m_e/m_h)^{-1}$ will be defined as the sum of the powers of the paired electron-hole (electron-electron) collisions and of the generation of plasmons. Leaving out the procedure of finding n_c^* , which is analogous to that described in Ref. 1, we have for the critical density

$$n_c^* = \frac{2\epsilon_0(k\theta)^2 (1/\kappa_\infty - 1/\kappa_0) \bar{\kappa}^2 m_e \ln[2(E_e/k\theta)^{1/2}]}{q^2 \hbar^2 [1 + 4m/(m_e + m_h) + \ln(E_e/\hbar\omega_p)^{1/2}]}, \quad (4)$$

where $\bar{\kappa}$ is the average dielectric constant of the material, m

is the reduced mass of the electron and hole, and $\omega_p = (q^2 n / \kappa \epsilon_0 m)^{1/2}$ is the plasma frequency.¹⁴ The first and second terms in the square brackets of the denominator of (4) take into account the paired electron-electron¹⁵ and the analogous electron-hole collisions, while the third term corresponds to plasmon generation.¹⁶ At low plasma frequency (in the case of II-VI compounds we have $\hbar\omega_p \approx 20$ meV at $n = 5 \times 10^{17} \text{ cm}^{-3}$). The plasmon-phonon interaction is negligible, and the plasmon energy is consumed in final analysis of heating of the EHP.¹⁷ The critical densities of the investigated II-VI compounds, calculated according to (4) are given in Table I.

To calculate I_{hot} from (1) it is necessary to determine in addition the dependence of the density of the hot NEC on the excitation level. This dependence has a complicated form for spatially and thermally inhomogeneous nonequilibrium system. The problem can be greatly simplified by taking the following into consideration. The NEC that are emitted from the hot region and to which no energy flows, cool rapidly (it follows from (2) that this occurs within $\sim 10^{-13}$ sec), and their diffusion coefficient, determined by the polar-optical scattering of the momentum, increases (see Eq. (7)). In addition, at low temperatures these NEC are bound into excitons, which are additionally dragged by the phonon wind directed from the hot region into the interior of the crystal.¹⁸ Consequently, a backward flow of nonequilibrium quasiparticles into the hot region is hindered not only by the density gradient, but also by the gradient of the diffusion coefficient and by the phonon wind. It can therefore be assumed that at $T_e \gg T$ the time that the $e-h$ pair remains in the hot region, $\hat{\tau}$ (the active time of the hot $e-h$ pair), is determined by the rate

TABLE I.

	Material			
	ZnTe	ZnSe	ZnS	CdTe
$(\hbar\nu_0 - E_g)$, eV	1.12	0.69	0.84	0.75
T_{e0} , K	310	420	370	360
l_{hot}^{-1} , 10^5 cm^{-1}	1.1	2.5	2.5	0.72
$\alpha(\hbar\nu_0)$, 10^5 cm^{-1}	3.0[24]	{ 1.2[24] 1.2[25]	{ 4.7[24] 2.4[23]	0.4[24]
n_c^* , 10^{17} cm^{-3} (according to LTC)	1.7	3.4	14	0.80
n_c^* , 10^{17} cm^{-3} [theoretical value (4)]	3.2	5.0	14	1.7

	Material			
	CdSe		CdS	
	E \perp c	E \parallel c	E \perp c	E \parallel c
$(\hbar\nu_0 - E_g)$, eV	0.51	0.51	0.93	0.93
T_{e0} , K	300	310	270	345
l_{hot}^{-1} , 10^5 cm^{-1}	2.10	2.16	2.3	2.5
$\alpha(\hbar\nu_0)$, 10^5 cm^{-1}	{ 1.4[23] 0.5[24] 1.3[26]	{ 1.6[23] 0.9[24] 1.4[26]	{ 1.6[23] 0.9[24]	{ 2.5[23] 1.1[24]
n_c^* , 10^{17} cm^{-3} (according to LTC)	1.8	1.8	8.2	8.2
n_c^* , 10^{17} cm^{-3} [theoretical value (4)]	3.1	3.1	7.8	7.8

of the diffusion travel over the distance l_{hot} . In this case

$$\hat{\tau} = l_{\text{hot}}^2 / 2D_a(T_e), \quad (5)$$

$$n_{\text{hot}} = \hat{\tau} / h\nu_0 l_{\text{hot}} = l_{\text{hot}} / 2h\nu_0 D_a(T_e), \quad (6)$$

where $D_a(T_e)$ is the coefficient of ambipolar diffusion of the hot carriers, corresponding to polar-optical mobility^{12,13}:

$$D_a(T_e) = 6(2\pi)^{3/2} \epsilon_0 \hbar^2 (kT_e)^{3/2} \left[q^2 (k\theta)^2 \left(\frac{1}{\kappa_{\infty}} - \frac{1}{\kappa_0} \right) M_D^{3/2} \right]^{-1} \\ \times \text{sh} \left(\frac{\theta}{2T} \right) \left[\text{ch} \left(\frac{\theta}{2T} - \frac{\theta}{2T_e} \right) K_1 \left(\frac{\theta}{2T_e} \right) \right. \\ \left. + \text{sh} \left(\frac{\theta}{2T} - \frac{\theta}{2T_e} \right) K_0 \left(\frac{\theta}{2T_e} \right) \right]^{-1}, \quad (7)$$

where K_1 is a modified Bessel function of first order and

$$M_D^{3/2} = m_e^{3/2} +^{1/2} (m_{nh}^{3/2} + m_{vh}^{3/2} e^{-\Delta/kT_e}) (1 + e^{-\Delta/kT_e})^{-1}. \quad (8)$$

Substituting (6) in (1) and using the condition of saturation of the effective temperature ($T_e \rightarrow T_{e0}$ as $n_c^*/n \rightarrow 0$ Ref. 6), we obtain for the length of the hot region

$$l_{\text{hot}} = [2(h\nu_0 - E_g) D_a(T_{e0}) / P_{\text{eh}}(T_{e0})]^{1/2}. \quad (9)$$

3. EXPERIMENTAL PROCEDURE

We investigated ZnTe, ZnSe, ZnS, CdTe, CdSe, and CdS single crystals, which were not specially doped and had a high quantum luminescence yield and a small luminescent background.¹ Freshly cleaved samples of the single crystals were excited by harmonics of an AYG:Nd³⁺ laser operating in the intermittent regime ($\tau_p = 10$ nsec, $f_{\text{rep}} = 12.5$ Hz). The energy of the exciting photon for each material was chosen such that the excess energy was in the range 0.5–1 eV. The maximum excitation levels I_0 were determined by the sample damage threshold and amounted to the following: 1.7 MW/cm² ($h\nu_0 = 3.50$ eV) for ZnTe 2.7 ($h\nu_0 = 3.50$) for ZnSe; 3.0 ($h\nu_0 = 4.67$) for ZnS; 1.9, ($h\nu_0 = 2.34$) for CdTe; 2.5 ($h\nu_0 = 2.34$) for CdSe and 5.0 ($h\nu_0 = 3.50$) for CdS. The oriented CdSe and CdS crystals were investigated in two excitation configurations relative to the optical **c** axis: **E**⊥**c** and **E**∥**c** (here **E** is the electric-field vector of the exciting light). The samples were placed in a nitrogen cryostat ($T = 77$ K). The edge-luminescence spectra were analyzed with a grating monochromator and registered photoelectrically in a wide dynamic range, followed by synchronous detection and analog determination of the logarithm of the signal.

4. RESULTS AND THEIR DISCUSSION

Figure 1 shows by the way of example the luminescence spectrum of ZnTe at a maximum excitation level. Similar spectra were obtained also for other investigated materials. The fundamental emission band of ZnTe with a maximum of 2.36 eV is due to radiative processes in the dense partially ionized exciton gas.¹⁹ The short-wave edge of this band usually has an exponential shape, which reflects the temperature of the system of excitons²⁰ localized at the diffusion depth. The exciton temperature T_x in our experiment, just as in Refs. 3, 21, and 22, exceeded the lattice temperature by several dozen degrees K. In the region $h\nu > E_g$, at high excitation levels, there appears in the luminescence spectra one more exponential wing, which can be easily separated by

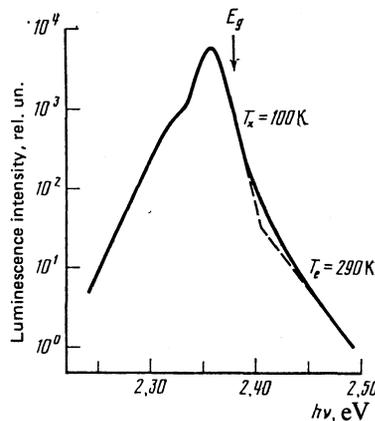


FIG. 1. Luminescence spectrum of ZnTe single crystal at the maximum excitation level (the luminescent background has been subtracted); $T = 77$ K, $h\nu_0 = 3.50$ eV, $I = 1.7$ MW·cm⁻². The arrow marks the energy position of the band gap.

subtracting the exponential edge of the fundamental band and the luminescence background from the spectrum. The characteristic temperature T_e of this wing greatly exceeded the lattice temperature and was saturated under maximum excitation levels. Figure 2 shows in a $[I^{-1}, (T_e - T)^{-1}]$ scale the dependences of the effective NEC temperature determined from the luminescence spectra on the excitation level (the lux-temperature characteristics LTC) for the investigated cubic crystals, and CdTe. The scale employed is convenient for the determination of the saturation temperature T_{e0} by a graphic method, using extrapolation of the LTC to the point $I^{-1} = 0$. Figure 3 shows the LTC for the hexagonal crystals CdSe and CdS, investigated at two excitation configurations (**E**⊥**c** and **E**∥**c**). The saturation temperature for CdS [Fig. 3(b)] depends noticeably on the polarization of the exciting light relative to the optical **c** axis. A less pronounced anisotropy of the heating of the NEC was observed in CdSe [Fig. 3(a)]. The effective NEC temperature was higher in the **E**∥**c** polarization than in **E**⊥**c**, on the average by 70 K for CdS and by 10 K for CdSe. It is known that oriented CdSe and CdS crystals have an anisotropic absorption for the employed light-quantum energies. The absorption coefficient in the polarization **E**∥**c** is larger in this case than for **E**⊥**c** (Ref. 23 and 24). Inasmuch as in our experiments the diffusion conditions did not change when the excitation polarizations were interchanged (the excited plane of the crystal remained parallel to the **c** axis in both cases), the anisotropic heating of the EHP can be attributed only to localization of hot NEC at the absorption depth of the exciting light.⁵

Using the obtained values of the saturation temperature, we can calculate the reciprocal length l_{hot}^{-1} of the hot region in accordance with (9) and to compare it with the absorption coefficient. The saturation temperature, the reciprocal length of the hot region, and the absorption coefficients α obtained in Refs. 23–26 for the corresponding wavelengths are all listed in Table I. As seen from Table I, the reciprocal length of the hot region is close in magnitude, for all the investigated crystals, to the absorption coefficient, thus confirming the model of localization of the hot NEC at

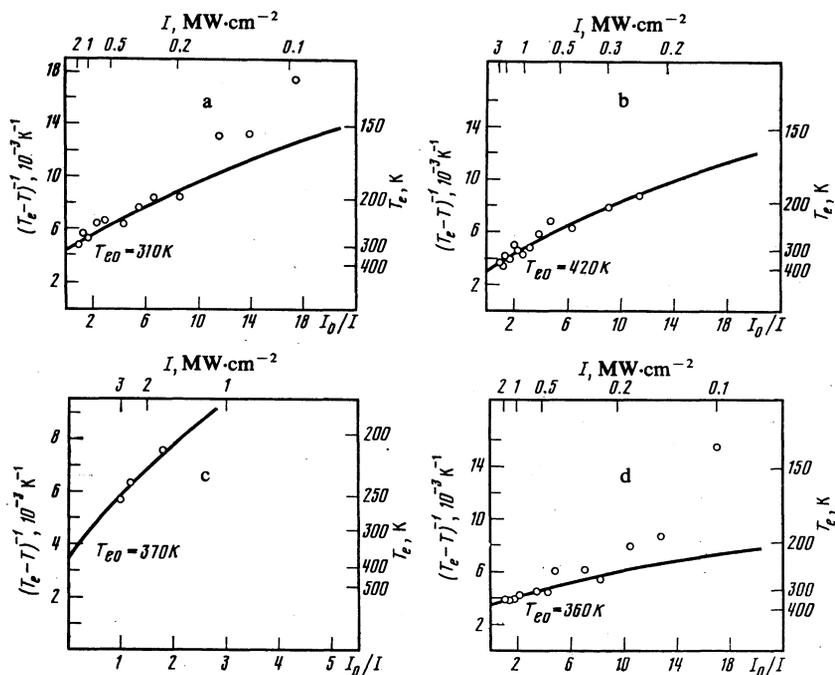


FIG. 2. Lux-temperature characteristic of cubic single crystals at $T = 77$ K: a - ZnTe ($h\nu_0 = 3.50$ eV); b - ZnSe ($h\nu_0 = 3.50$ eV) c - ZnS ($h\nu_0 = 4.67$); d - CdTe ($h\nu_0 = 2.34$). The solid lines correspond to the theoretical LTC (10).

the depth of penetration of the exciting radiation into the crystal. The stratification of the system of nonequilibrium quasiparticles ensures effective heating of the surface layer of the EHP, since the active time of the e - h pair is small in this layer [$\hat{\tau} \sim 10^{-12}$ sec according to (5)].

The obtained LTC make is also possible to estimate the critical density n_c^* and to compare it with the theoretical one calculated in accordance with (4). We used for this purpose the power-balance equation (1), which can be reduced, taking (6) and (9) into account, to the form

$$\frac{P_{eh}(T_{e0})}{P_{eh}(T_e)} = \frac{D_a(T_{e0})}{D_a(T_e)} + \frac{2h\nu_0 D_a(T_{e0}) n_c^*}{Il_{hot}} \quad (10)$$

At the known values of T_{e0} and l_{hot} , the theoretical LTC (10) contains one unknown parameter n_c^* , selection of which makes it possible to reconcile the theoretical LTC with the experimental points in the region of maximum excitation levels. The solid curves in Figs. 2 and 3 show the theoretical dependences obtained in this manner for the effective temperature of the NEC on the excitation level, and the corresponding values of n_c^* are listed in Table I. The semiconductor-material parameters needed for the calculations were

taken from Refs. 27-30. We note that the deviations of the experimental points from the theoretical curves, towards lower temperatures with decreasing excitation level, observed for certain materials [Figs. 2(a,d); 3(b)] are apparently due to the turning on of the NEC momentum scattering mechanism of the NEC by the impurities at the lower EHP density. This leads to a decrease of the diffusion coefficient, causing a sharper decrease of the temperature T_e in the hot region according to (9).

As seen from the table, the obtained values of n_c^* agree on the whole well with the theoretical ones, thus confirming the applicability of the approximation of diffuse passage through the hot region, an approximation used in the derivation of (9). A certain disparity for materials with low effective electron mass (in particular, for CdTe) can be attributed to the weak screening of the polar-optical interaction, which decreases the critical density. When dealing with the screening of the electron-phonon interaction, it must be noted that the observed characteristic saturation of the effective temperature does not confirm the conclusions of the theoretical papers^{31,32} from which it follows that the power of the polar-optical losses of a hot e - h pair decreases noticeably already at

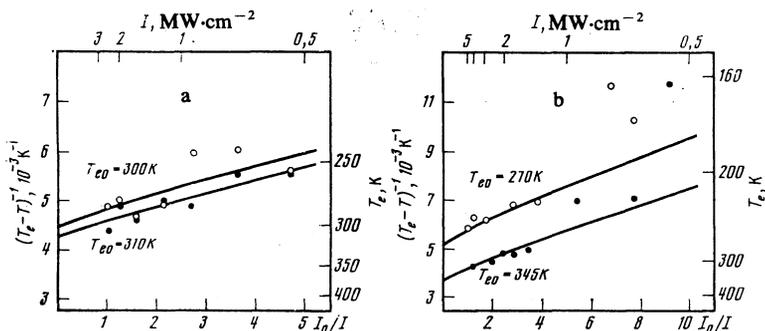


FIG. 3. Lux-temperature characteristics of oriented single crystals at $T = 77$ K: a - CdSe ($h\nu_0 = 2.34$ eV); for b - CdS ($h\nu_0 = 3.50$ eV) and $E||c$ (●) and $E\perp c$ (○). Solid lines—theoretical LTC (10).

a density 10^{17} cm^{-3} , and subsequently falls abruptly with increasing density. It appears that the threshold NEC density corresponding to the turning on of the screening is in fact somewhat higher (more than 10^{18} cm^{-3}).

We note that localization of a hot EHP at the penetration depth of the exciting light does not always take place. Thus, at low excitation levels (i.e., at a weak electro-plasma interaction), in materials with a less pronounced electron-phonon coupling than in II-VI compounds, the photoexcited e-h pair loses its initial excess energy at a depth considerably exceeding the reciprocal absorption coefficient.³³ In this case the nonequilibrium quasiparticles are heated practically in the entire excited volume, but the heating effect itself is small, since the active time of the hot e-h pair is close in value to the lifetime. On the other hand, when the EHP is degenerate, the power of the polar-optical losses decreases, and the diffusion coefficient increases. Under these conditions, the hot NEC that leave the light-diffuse absorption region to a considerable depth during the cooling time.¹⁷

Thus, study of the photo-heating of NEC in a number of compounds of the II-VI group has shown that high-power surface excitation leads to formation of a layer of hot EHP in the region where the exciting light penetrates. A characteristic feature of the dynamic of the NEC heating in this layer is the saturation of the effective temperature. The surface EHP layer plays an important role in the process of energy exchange between the crystal and the light source: The processes of thermalization of the photoexcited NEC and generation of nonequilibrium phonons are localized in this layer.

¹By luminescent background is meant the "white" structureless emission whose intensity, as already noted, depends strongly on the surface state of the sample, on the lattice temperature, and on the crystal quality. In better samples, the background intensity amounted to 10^{-4} of the intensity at the emission maximum. The nature of this phenomenon is not completely clear.

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