Anisotropy of the heating of photoexcited electrons in CdS single crystals

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The temperature of the electrons heated during photoexcitation has been observed to depend on the orientation of the polarization plane of the exciting light with respect to the **c** optic axis in CdS single crystals. This is the first observation of such a dependence. The effect is shown to result from a localization of hot charge carriers at the depth to which the exciting light penetrates into the crystal. The time spent by a hot carrier in this layer has been determined: $\hat{\tau}_\perp = 0.81$ ps and $\hat{\tau}_\parallel = 0.58$ ps.

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The heating of nonequilibrium charge carriers during the photoexcitation of a semiconductor by photons with an energy $h\nu_0$ greater than the gap width E_g is of interest in connection with research on energy exchange in a system of nonequilibrium quasiparticles. Shah[1] has shown that nonequilibrium charge carriers are heated in polar semiconductors by the excess energy of an excited electron–hole pair, while the heating dynamics is determined by a competition between the electron–electron interaction, which causes heating, and the polar–optical scattering of nonequilibrium charge carriers, which causes cooling. Shah assumed that the hot nonequilibrium carriers were at a depth equal to the ambipolar diffusion length. In Ref. 2, however, we showed that a hot electron–hole pair dissipates its excess energy over an "active time" of the order of 10^{-12} s, i.e., over a time far shorter than its lifetime (which is of the order of 10^{-10}–10^{-9} s for most $A^{II}B^{VI}$ semiconductors). On this basis it may be assumed that the hot carriers are localized exclusively at the photon absorption depth.

In this letter we are reporting an effort to test this assumption on the basis of the anisotropy of the absorption of light in hexagonal CdS.

The heating of nonequilibrium carriers was studied in oriented CdS single crystals grown from a melt. A freshly cleaved sample was excited by the polarized third-harmonic beam from a YAG:Nd^{3+} laser ($h\nu_0 = 3.51$ eV, $\tau_p = 12$ ns, $f_{rep} = 12.5$ Hz, maximum excitation power density $I_0 = 5$ MW/cm^2). The polarization plane of the exciting light was oriented either perpendicular or parallel to the **c** optic axis ($\alpha_\perp = 1.6 \times 10^5$ cm^{-1}, $\alpha_\parallel = 2.5 \times 10^5$ cm^{-1}, Ref. 3). In both cases, the excited plane of the crystal was parallel to the optic axis. The luminescence was studied by the method of Ref. 4.

Figure 1 shows some typical spectra of the edge luminescence of CdS single crystals in the two excitation configurations ($\mathbf{E} \perp \mathbf{c}$ and $\mathbf{E} \parallel \mathbf{c}$) at a sample temperature of 77 K. The fundamental edge-luminescence band results from radiative processes which occur at the ambipolar-diffusion depth in a dense exciton gas.[5] In the present

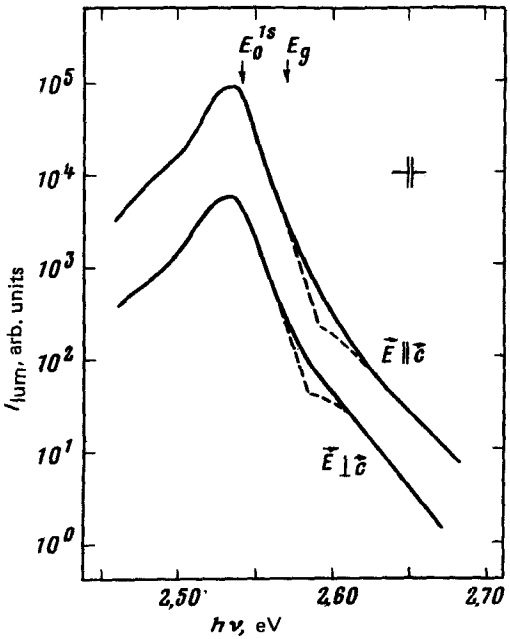

FIG. 1. Luminescence spectra of CdS single crystals at $T = 77$ K and at the maximum excitation level for two excitation configurations. The arrows show the energy positions of the exciton ground state (E_0^{1s}) and the width of the energy gap (E_g). The intensity scale is arbitrary for each spectrum.

case, we are primarily interested in the spectral region $h\nu > E_g$, where there is an exponentially decaying wing caused by the radiative recombination of hot nonequilibrium carriers. From the slope of this wing in a semilogarithmic plot we can determine the temperature of the hot electron–hole pairs,[1] T_e. To improve the reliability of the results, we used a graphical method to separate the hot-plasma emission bands from the fundamental band; furthermore, we studied only light with the $\mathbf{E} \perp \mathbf{c}$ polarization. In this manner we obtain the intensity–temperature characteristics shown in Fig. 2 for the system of hot nonequilibrium carriers through a study of the lumine-

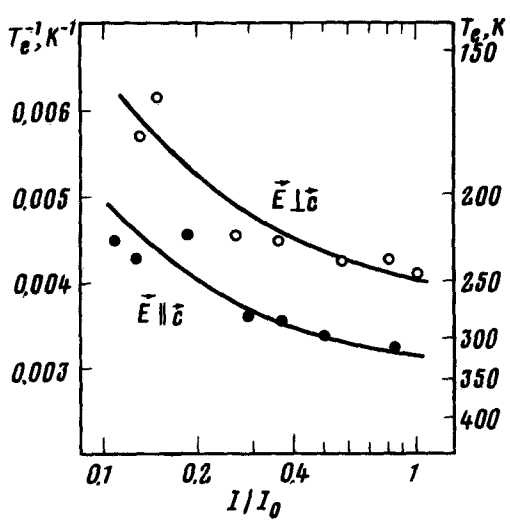

FIG. 2. Intensity–temperature characteristics for the two excitation configurations. Points–Experimental; curves–theoretical.

scence spectra for various excitation levels and with the two excitation polarizations. We see from this figure that the temperature T_e for the polarization $\mathbf{E} \parallel \mathbf{c}$ averages 70 K higher than for the $\mathbf{E} \perp \mathbf{c}$ polarization; in other words, there is an anisotropy in the heating of the nonequilibrium carriers.

For a quantitative analysis of the effect, let us calculate the saturation temperature T_{e0}, which is a parameter of the heating of the system of nonequilibrium carriers.[2] This saturation temperature can be determined by comparing the experimental intensity–temperature characteristic with the theoretical characteristic found from

$$\frac{n}{n + n_c^*} = \frac{P(T_e)}{P(T_{e0})}, \tag{1}$$

in which T_{e0} appears as a parameter. Here n is the concentration of hot electrons, n_c^* is the critical concentration, at which the rate of the polaroptical loss for a photoexcited electron is equal to the rate at which energy is transferred to the system of hot nonequilibrium carriers with the temperature T_e, and $P(T_e)$ is the polar–optical loss power for a single electron of this system.[1,6] Working in this manner, we found $T_{e0\perp} = 270$ K for the perpendicular excitation polarization and $T_{e0\parallel} = 345$ K for the parallel polarization (the solid curves in Fig. 2 show the theoretical intensity–temperature characteristics corresponding to these temperatures). Using T_{e0}, we can determine $\hat{\tau}$, the active time over which the hot electron–hole pair dissipates its excess energy:

$$\frac{h\nu_0 - E_g - 2kT_{e0}}{\hat{\tau}} = P(T_{e0})\left[1 + \frac{1}{2}\left(\frac{m_h}{m_e}\right)^{1/2}\right]. \tag{2}$$

The factor in the brackets reflects the contribution to the polar-optical loss of holes which have been thermalized with electrons, in accordance with Ref. 7. The active times calculated from Eq. (2) are $\hat{\tau}_\perp = 0.81 \pm 0.1$ ps and $\hat{\tau}_\parallel = 0.58 \pm 0.1$ ps. The ratio $\hat{\tau}_\perp/\hat{\tau}_\parallel = 1.4$ is approximately equal to the ratio of absorption lengths in the corresponding excitation configurations, $\alpha_\parallel/\alpha_\perp = 1.5$. This approximate equality suggests that the emission by the hot electron–hole plasma occurs from the light absorption region, rather than from the ambipolar-diffusion depth of the nonequilibrium carriers; in the present experiments, this depth did not change when the excitation configuration was changed.

In summary, the anisotropy which we have detected in the heating of photoexcited nonequilibrium carriers suggests that these carriers are localized exclusively at the absorption depth of the exciting light. The entire system of quasiparticles is "stratified" in terms of both coordinate and temperature, and this circumstance must be taken into account in experiments involving intense surface excitation of crystals.

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