Exciton drag in CdS crystals during intense photoexcitation

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A kinetic study has been made of the exciton luminescence of CdS crystals, from the excited surface and from the opposite surface. The results yield spectra of the time of flight of excitons across the sample. Radical changes observed in these spectra indicate an exciton drag effect.

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The surface excitation of a semiconductor creates inhomogeneous concentrations of free excitons and free carriers, which decay exponentially with distance from the surface. The decay is characterized by the distance L_D, the diffusion length. With sufficiently intense excitation, however, some new effects may occur and lead

to changes in these distributions. As was first shown by Keldysh[1] and Bagaev *et al.*[2] in connection with a study of the motion of electron–hole drops, the nonequilibrium phonons, which result from the generation and recombination of free excitons and free carriers, can lead to a new effect: a drag of the free excitons and of electron–hole drops by the flux of these phonons. In contrast with the electron–hole drops, however, it has not been possible to detect the drag of free excitons.[2,3] In a material with a direct energy gap, on the other hand, it is necessary to consider the possibility of multiple re-emission and reabsorption of a photon,[4] which would lead to a photostimulated particle transport over distances greater than L_D.

In this letter we are reporting the observation of a transport of free excitons, and we shall discuss a possible mechanism for this effect. In the experiments we studied CdS single crystals grown from the gas phase with $N_D \leqslant 10^{15}$ cm^{-3} with a thickness $l = 2$–100 μm. The sample was excited with an N_2 laser with a stroboscopic system for measuring the luminescence spectrum.[5] The recombination radiation was measured in two geometric arrangements: with the wave vector of the exciting light, $\mathbf{k}_I$, parallel to the wave vector of the recombination radiation, $\mathbf{k}_L$($\mathbf{k}_I \| \mathbf{k}_L$), and with $\mathbf{k}_I$ antiparallel to $\mathbf{k}_L$($\mathbf{k}_I \| -\mathbf{k}_L$). Under these conditions we studied the luminescence kinetics and intensity, which reflect the temporal and spatial distribution of the excitons in the sample.[1)] It can be seen in Fig. 1 that there is no change in the kinetics of the emission at the free-exciton phonon-repetition line (curve 5) as the excitation intensity is varied over the range $I = 5 \times 10^{21} - 5 \times 10^{23}$ photons/cm^2 · s), and (within this intensity range) there is no change in the kinetics as the experimental geometry is varied.[5] In contrast, the emission at the free-exciton line undergoes

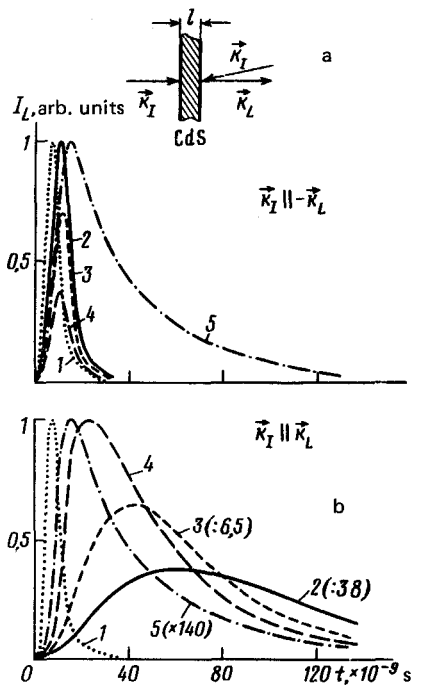

FIG. 1. Luminescence kinetics in the configurations with $\mathbf{k}_I \| -\mathbf{k}_L$ and $\mathbf{k}_I \| \mathbf{k}_L$ at $T = 77$ K. 1–Pulse of exciting light from the N_2 laser; 2–4–free-exciton emission pulse for various excitation intensities; 2–$I = 5 \times 10^{21}$ photons/(cm^2 · s); 3–5×10^{22} photons/(cm^2 · s); 4–5×10^{23} photons/(cm^2 · s); 5–emission pulse at the free-exciton phonon-repetition line for $I = 5 \times 10^{21} - 5 \times 10^{23}$ photons/(cm^2 · s).

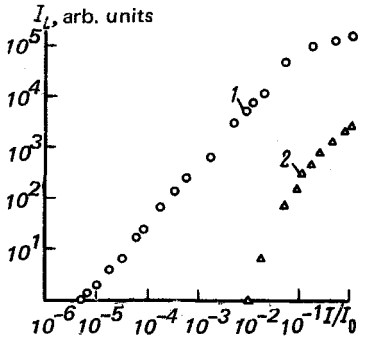

FIG. 2. Luminescence intensity at the maximum of the free-exciton emission line vs. the excitation intensity I at $T = 77$ K. $1 - \mathbf{k}_I \| -\mathbf{k}_L$; $2 - \mathbf{k}_I \| \mathbf{k}_L$.

some important changes: First, as I is increased the free-exciton emission intensifies faster than linearly in the configuration with $\mathbf{k}_I \| \mathbf{k}_L$, in contrast with the essentially linear behavior in the case $\mathbf{k}_I \| -\mathbf{k}_L$ (Fig. 2). Second, the half-width of the free-exciton emission in the case $\mathbf{k}_I \| \mathbf{k}_L$ decreases, and the maximum shifts toward the beginning of the excitation with increasing I. With $\mathbf{k}_I \| -\mathbf{k}_L$, on the other hand, the shape of the free-exciton pulse remains essentially constant (curves 2–4 in Fig. 1). This anomalous behavior of the free-exciton luminescence in the various experimental arrangements indicates an external force which depends on I and which causes free excitons to drift into the crystal. This drift leads, in turn, to the appearance of free excitons at the surface opposite the excitation surface, which means that we are observing exciton drag.

As a possible mechanism[2)] for the free-exciton drag, we consider the "phonon wind" which was proposed by Bagaev *et al.*[2] The free-exciton luminescence intensity is proportional to the concentration of these excitons, $n(x,t)$, which in our case should satisfy a diffusion equation with a drift term $-j_d = Df/k_BTn$, where D is the diffusion coefficient, $\mathbf{f}$ is the drag force, and T is the temperature. Assuming, for simplicity, that free excitons are generated at the surface of the sample by a source $g = \delta(x)\delta(t)$, ignoring surface recombination, and assuming that $n(x,t)_{\substack{x\to\infty \\ t\to\infty}} \to 0$, we find the following expression for $n(x,t)$:

$$n(x,t) = \frac{1}{2\pi^{1/2}} (Dt)^{-1/2} \exp\left(-\frac{t}{\tau}\right) \exp\left[-\frac{\left(x - \frac{Df}{k_BT}t\right)^2}{4Dt}\right] \qquad (1)$$

$$t \geqslant 0 .$$

Figure 3 compares the results calculated from (1) for $x = l = 20$ μm and for various exciton velocities $v = Df/k_BT$. For these calculations we assumed that the drag force was constant over the time required for the free exciton to cross the sample.[6] It follows from Fig. 3 that over the intensity range studied [$I = 5 \times 10^{21} - 5 \times 10^{23}$ photons/(cm^2 · s)] the free-exciton velocity is $v = (0.8-8) \times 10^4$ cm/s $< s$, where s is the sound velocity. The drag force is $f = (0.8\text{–}8.5) \times 10^{-11}$ dyn for $I = 5 \times 10^{21} - 5 \times 10^{23}$ photons/(cm^{-2} · s). From the experimental data we can estimate τ_r, the relaxation time for the free-exciton momentum. Assuming that the damping time of

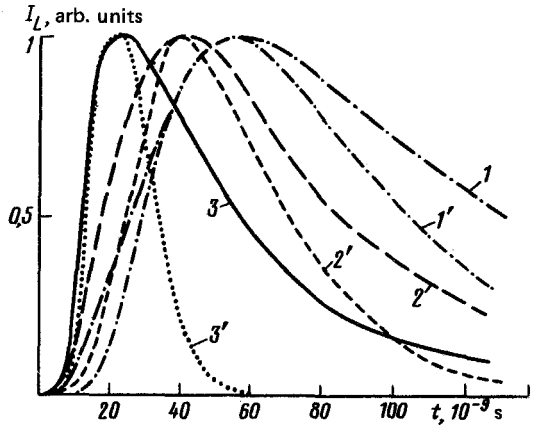

FIG. 3. Comparison of the observed shape of the free-exciton emission pulse in the arrangement $\mathbf{k}_I \| \mathbf{k}_L$ with the results calculated from (1) for $T = 77$ K. $1-I = 5 \times 10^{21}$ photon/(cm^2 · s); $2-5 \times 10^{22}$ photons/(cm^2 · s); $3-5 \times 10^{23}$ photons/(cm^2 · s). 1'–3'–Calculations from (1); 1'–$v = 8 \times 10^3$ cm/s; 2'–3×10^4 cm/s; 3'–8×10^4 cm/s.

the drag force is long in comparison with τ_r in our case, we have

$$\tau_r = \frac{v m_x}{f} .$$

In our case we have $\tau_r = 8 \times 10^{-13}$ s at $T = 77$ K. It follows from (1) that a decrease in the exciton lifetime causes a contraction of the light pulse in the configuration with $\mathbf{k}_I \| \mathbf{k}_L$. For the samples with $\tau \leqslant 2 \times 10^{-9}$ s it is much more difficult to observe the free-exciton drag. It should be noted that the effect falls off with increasing temperature, possibly because of a decrease in the lifetime of nonequilibrium phonons. At low temperatures the effect again falls off, in this case because of a decrease in the free-exciton lifetime caused by radiative recombination through a bound-exciton state with $\tau \leqslant 10^{-9}$ s (Ref. 7 and 8).

[1]It should be noted that the intensity of the free-exciton line is proportional to the number of excitons in a thin surface region of thickness $\sim \alpha_x^{-1}$, where $\alpha_x \sim 10^5$ cm^{-1} is the absorption coefficient for the light at the free-exciton line. The intensity of the phonon-repetition line of the free excitons is determined by the total number of free excitons in the volume which they occupy.

[2]Estimates show that attempts to explain the experimental results on the basis of the reradiation effect[4] run into difficulties because it becomes necessary to appeal to a nonlinear particle-recombination mechanism. The nonlinear recombination processes, which are possible in the exciton system, should lead to emission in a spectral region where the absorption coefficients are low (the mean free path of the corresponding photons is greater than the dimensions of the sample), so that the exciton diffusion coefficient cannot increase. The significant effective duration of the drag force observed experimentally can be explained by the "hot-spot" effect.[6]

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