# Relaxation of the momentum of free excitons in semiconductors

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The relaxation time of the momentum of free excitons in semiconductors (CdS) is measured for the first time over a wide temperature range 10–300 K. For $T < 60$ K, the relaxation is determined by scattering of the exciton by piezoacoustic phonons, while for $T > 100$ K, scattering by optical phonons dominates.

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The magnitude of the diffusion coefficient or mobility of nonequilibrium charge carriers or excitons is an important kinetic characteristic, which determines the transport processes in semiconductors. The problem of determining these quantities is solved easily only for charged particles. As far as excitons are concerned, we are aware of only a single work, in which an attempt was made to measure the mobility of excitons in a nonuniform strain field in a Si crystal over a narrow temperature range 1.5–20 K.[1]

In the present work, the magnitude of the diffusion coefficient of thermalized excitons $D$ and the value of the momentum relaxation time $\tau_p$ are directly measured for the first time over a wide temperature range $T = 10$–300 K in semiconductors (in particular, in CdS crystals). In order to measure $D$ and $\tau_p$, we studied nonstationary diffusion of excitons generated in the specimen by surface excitation of the semiconductor with short light pulses (inset in Fig. 1). The time dependence of the exciton radiation was recorded in the region of generation (point A) and in a region removed far from the excitation spot at a distance $x = L$ (point B). Since the coefficient of absorption of exciton radiation from the resonant state was $a_x \cong 10^5$ cm$^{-1}$, detection

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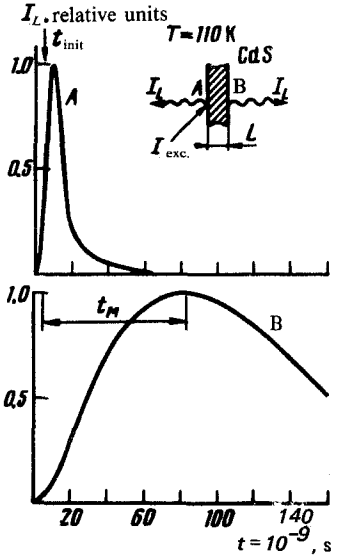


FIG. 1. The time dependence of exciton radiation in the region of generation (A) and in a region far from it at a distance $L$ (B) for $T = 110$ K.

of exciton radiation at point B was possible only if excitons diffused into this region. These CdS specimens, grown out of the gas phase, were plates with a thickness $L \cong 10$–$40\ \mu$m; the concentration of residual donors in them did not exceed $10^{15}\ \text{cm}^{-3}$.

The luminescence was excited by $N_2$ laser pulses with duration $(5\text{–}10)\times 10^{-9}$ s and intensity $I < 10^{23}\ \text{cm}^{-2}\,\text{s}^{-1}$. The specimens were placed into the chamber of a temperature-controlled cryostat, in which the temperature was maintained to within 0.1 K. The luminescence at the emission line of the exciton from the resonant state was analyzed with the help of a double DFS-24 monochromator with the spectral width of the slit not greater than 0.9 meV and a stroboscopic recording system with time resolution not worse than $10^{-9}$ s. When the lifetime of the excitons $\tau$ is long compared to $\tau_p$, they reach thermodynamic equilibrium with the lattice. On the other hand, when the mean free path of a photon, which is emitted as a result of recombination of an exciton, is short compared to the diffusion length of excitons, $L_D$ (the contribution of transport due to reradiation is small), the motion of excitons in the crystal in the absence of external fields can be described by the standard diffusion equation. The relation for determining $D$ follows from an analysis of the solution of this equation:

$$D = \frac{L^2 \tau}{4\left(t_m^2 + \frac{3}{2} t_m \tau\right)}, \qquad (1)$$

where $t_m$ is the time delay in attaining the maximum of the luminescence pulse of excitons that reach point B. In order to determine $D$ from (1), it is necessary to know the lifetime of the excitons $\tau$, which was determined by three independent methods: 1) from an analysis of damping of the luminescence pulse on the free-exciton phonon-repetition line, characterizing the relaxation of the integral concentration of excitons

in the specimen (the coefficient of absorption of light on this line is $a_{X-LO}L<1$, in contrast to the corresponding quantity for radiation of an exciton from the resonant state, where $a_X L \gg 1$); 2) from an analysis of damping of the exciton radiation signal at point B in the limit $t \to \infty$ [in this case $n(x,t) \sim \exp(-t/\tau)$]; 3) from an analysis of damping of luminescence as a result of homogeneous, volume excitation of the semiconductor (two-photon excitation). The numerical values of $\tau$ obtained by the methods indicated above differed in magnitude by not more than 10%. The quantity $t_m$ (Fig. 1) was determined from the experiment; then the magnitude of the diffusion coefficient of excitons was calculated from (1), taking into account the value found for $\tau$.[1)] The diffusion coefficient of particles in turn is related to the momentum relaxation time by the relation

$$D = \frac{1}{3}\tau_p \bar{v}^2, \tag{2}$$

where $\bar{v} = (3kTm_x)^{1/2}$ is the thermal velocity of an exciton, and $m_x$ is the effective mass of an exciton. Figure 2 illustrates the temperature dependence of $\tau_p$, obtained from an analysis of the experimental data in Fig. 1 using Eqs. (1) and (2). Figure 3 shows the results of a theoretical analysis of the scattering of excitons by acoustical and optical phonons. The experimental dependence $\tau_p = f(T)$ exhibits two sections with differing temperature behavior of $\tau_p$ (Fig. 2). Thus, for $T > 100$ K, the experimental dependence is nearly exponential, which is characteristic for scattering by optical phonons. As the results of the analysis of scattering of an exciton by optical phonons[3] and comparison with the data from our experiment show, the scattering of the exciton as a whole and the interaction with the *LO* phonons, which leads to its decay, are both very important. The results of the theory, illustrated in Fig. 3, show that for $T > 100$ K the contribution to scattering from both mechanisms is nearly identical. As far as the temperature range $T < 60$ K is concerned, as the results of the theoretical analysis

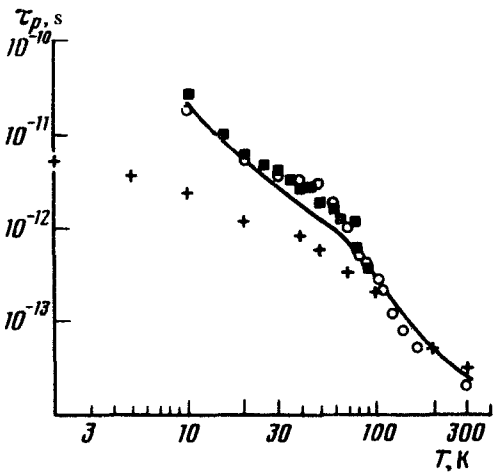


FIG. 2. The experimental values $\tau_p = f(T)$: ■ are for specimen No. 1; ○ are for specimen No. 2; + indicates data from Ref. 4 for electrons in *n*-CdS; and the solid curve represents the net theoretical dependence $\tau_p = f(T)$.

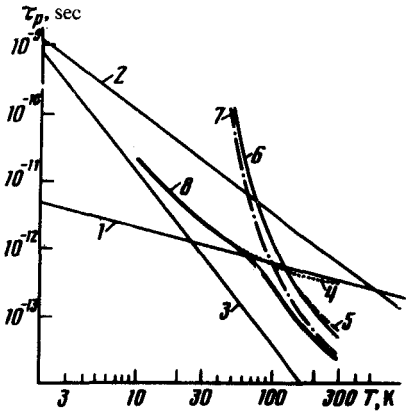


FIG. 3. The results of a theoretical analysis of the relaxation of the exciton momentum: 1) Scattering of electrons by acoustical phonons (piezoelectric interaction); 2) scattering of excitons by acoustical phonons (deformation interaction); 3) asymptotic dependence $\tau_p = f(T)$, acoustical phonons (piezoelectric interaction); 4) exact dependence $\tau_p = f(T)$ for scattering by acoustical phonons (piezoelectric interaction); 5) dependence $\tau_p = f(T)$ for scattering of excitons by optical phonons; 6) $\tau_p = f(T)$ for scattering by optical phonons, owing to decay; 7) total dependence $\tau_p = f(T)$ for scattering by optical phonons; 8) the net dependence $\tau_p = f(T)$ for exciton scattering.

show, it turned out that the most effective relaxation mechanism is scattering by acoustical phonons as a result of piezoelectric interaction of excitons and phonons. For temperatures 60 K $< T <$ 100 K, there is a characteristic region in which a mixed mechanism involving scattering by piezoacoustic and optical phonons is realized. A calculation of the contribution of the deformation interaction to the probability of scattering of excitons by acoustical phonons shows that this contribution, as compared with the piezoelectric interaction over the temperature range from 10 to 300 K, is small.

Thus, we were able to measure directly the momentum relaxation time of free excitons, to understand the mechanism for dissipation of the momentum of excitons and, using the quantities determined directly in the experiment, to obtain a good agreement between theory and experiment without any adjustable parameters.

[1)]It should be noted that in order to determine $D$ correctly, it is necessary that the maximum excitation intensity not exceed the pumping at which exciton drag by phonons begins to appear.[2]

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