Dispersion of the relaxation of $A_{n=1}$ excitons in CdS crystals

V. Ya. Reznichenko, M. I. Strashnikova, and V. V. Chernyĭ
Institute of Physics, Academy of Sciences of the Ukrainian SSR

(Submitted 3 April 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **49**, 554–556 (25 May 1989)

An effect of a mixing of states of allowed and forbidden transitions on the exciton relaxation spectrum near the $A_{n=1}$ resonance in a CdS crystal has been observed for the first time.

The relaxation of excitons in the resonant part of the spectrum is a topic of current interest in solid state physics, but we are still far from a clear understanding because this characteristic cannot be measured directly, and indirect data must be used. In this letter we construct the frequency dependence of the exciton relaxation constant $\Gamma(\omega)$ through a comparison of the measured transmission of a wedge-shaped CdS crystal[1] with theoretical predictions incorporating additional Pekar waves.[2]

Figure 1a shows the transmission measured at $T = 1.8$ K of a wedge-shaped crystal with an average thickness of 0.5 μm and an angle of refraction $\approx 50''$ in the vicinity of the lowest $A_{n=1}$ exciton. The filled circles refer to the "+ Pekar wave," i.e., the lower optical-exciton branch, and the open circles to the "− wave," i.e., the upper branch. The solid (+ wave) and dashed (− wave) lines are theoretical predictions for a CdS crystal with the same dimensions; the spatial dispersion and the additional Pekar boundary conditions are taken into account in these lines. All of the theoretical parameter values used here were the same as in Ref. 1 except Γ, which we assigned a discrete series of values: for curve 1, 1×10^{-5} eV; 2, 2×10^{-5} eV; 3, 4×10^{-5} eV; 4, 8×10^{-5} eV; 5, 1.4×10^{-4} eV; 6, 2.2×10^{-4} eV. Within each curve, Γ is independent of the frequency. A comparison shows that none of the calculated curves coincide with the experimental results. We accordingly assumed that for each frequency ω the relaxation Γ has the value for which the experimental point falls on the corresponding theoretical curve.

The dashed line in Fig. 1b shows the functional dependence $\Gamma(\omega)$ constructed in this way. On the long-wave side of a transverse exciton, ω_T, the relaxation $\Gamma(\omega)$ asymptotically approaches the very small value 1.5×10^{-5} eV. In the region of the longitudinal-transverse splitting Δ_{LT}, at a wavelength of 4855 Å, the dependence $\Gamma(\omega)$ is described by a fairly narrow delta-shaped curve. As the frequency ω_L is approached, there is a sharp increase, to a shelf with a value $\approx 2\times10^{-4}$ eV. This behavior of $\Gamma(\omega)$ was completely unexpected.

Figure 1c shows the frequency dependence of the group velocity of optical excitons in the crystal, V_{gr}, and the corresponding refractive index $n_{gr} = c/V_{gr}$, where c is the velocity of light in vacuum. Thanks to the submicron thickness of the crystals used here, we have been able, for the first time, to measure the behavior $V_{gr}(\omega)$ of a + wave over the entire absorption band and above ω_L. The circles were found from the

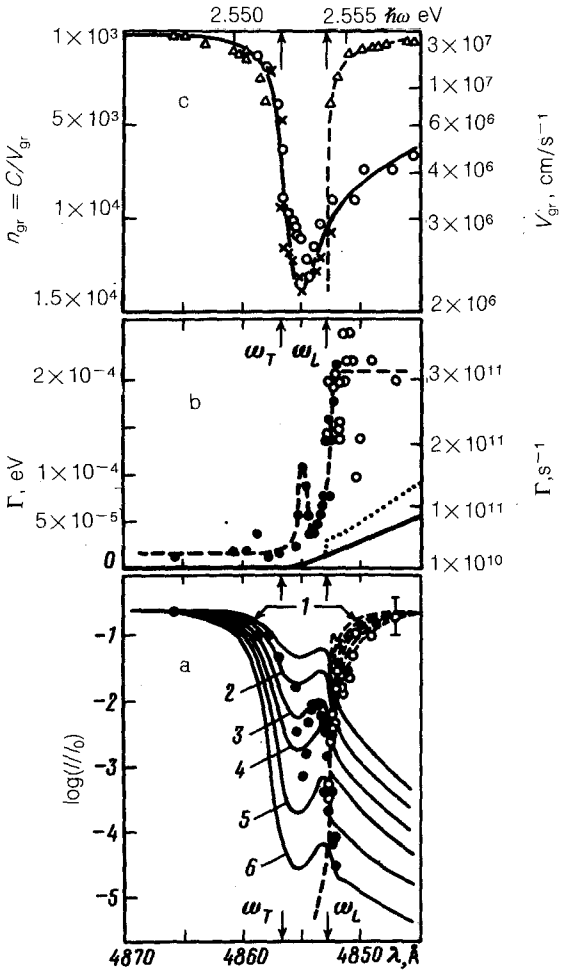

FIG. 1.

Fabry-Perot interference pattern of a plane-parallel CdS crystal 0.18 μm thick; the crosses were found from an interference pattern of polarized beams. The triangles are data from the measurements of Ref. 3, based on thicker crystals, in the wings of the absorption band. The solid and dashed lines are theoretical for the + and − waves, respectively.

A comparison of parts b and c of Fig. 1 shows that the minimum group velocity, which corresponds to the maximum value $n_{gr} \approx 1.3 \times 10^4$, occurs at $\lambda = 4855$ Å; this wavelength agrees with that at which $\Gamma(\omega)$ reaches its maximum, within Δ_{LT}. We are thus observing a qualitative anticorrelation between the relaxation spectrum of an optical exciton and its group velocity. The overall $V_{gr}(\omega)$ curve, however, is broader than the spike on the $\Gamma(\omega)$ curve, so we cannot say that there is a quantitative anticorrelation.

If the relaxation constant is assumed to be dominated by the exciton-phonon interaction with longitudinal acoustic phonons through the strain energy,[1)] a calculation from the relation which was used in Ref. 4 at $T = 0$ leads to a frequency dependence $\Gamma(\omega)$, as shown by the solid line in the lower part of Fig. 1b. We see that the calculated curve lies well below the experimental curve. Consequently, at very low temperatures, exciton-phonon coupling cannot explain the relaxation observed experimentally.

A scattering of excitons by impurities (charged donors and acceptors) in a compensated semiconductor was studied theoretically in Ref. 5. A comparison of the results of that study with the experimental results is possible only in the spectral interval Δ_{LT}. Taking the concentration of donors and acceptors in the compensated crystal to be 10^{15}–10^{16} cm^{-3}, we find the value $\Gamma \approx (0.4\text{–}4) \times 10^{-5}$ eV. This value is comparable to our values for $\omega < \omega_T$ and in the region Δ_{LT} under the δ-shaped peak. It can thus be assumed that this value of Γ is indeed a consequence of scattering by impurities.

We link the δ-shaped peak with an "anticrossing" of the dispersion branches of optical excitons of symmetry Γ_5 and Γ_6, i.e., with a mixing of states of allowed and forbidden transitions. The greatest mixing should occur in a very narrow spectral interval, which would be at the point at which the dispersion curves cross in the absence of their interaction ($\lambda = 4855$ Å). This effect of a forbidden transition on the dispersion of the refractive index, $n(\omega)$, was observed and interpreted in Ref. 6, and the results were confirmed in Ref. 1. In the present study, we have seen this effect for the first time in the exciton relaxation spectrum—in the form of a sharp increase in Γ, the reciprocal time of the "mean free path" of an exciton. Impurities in a crystal should intensify this effect.

Beginning at ω_L, the component of Γ, which stems from intraband scattering involving the lower optical-exciton branch, is supplemented with a component due to interband transitions between the upper and lower branches. The curve of $\Gamma(\omega)$ calculated for the case in which phonons alone participate in these processes is shown by the dashed line in Fig. 1b. We see that it lies far from the experimental results.

The sharp increase in $\Gamma(\omega)$ at the frequency ω_L and the onset of a constant value are apparently due to an additional contribution to Γ from interband transitions involving impurities, defects, etc. The actual meaning here is that a $-$ wave can convert into a $+$ wave in a collision with any obstacle. Furthermore, the wave can be scattered by the branch of longitudinal excitons. The mutual conversions of $+$ waves, $-$ waves, and longitudinal waves at spherical inclusions were studied theoretically in Ref. 7. It follows from that study that $\Gamma(\omega)$ can reach a constant value above ω_L. We therefore believe that at very low temperatures, even in high-quality CdS crystals, the predominant mechanism for the relaxation of excitons is their scattering by impurities. This conclusion applies to both intraband scattering ($\omega < \omega_L$), and interband scattering ($\omega > \omega_L$).

[1)]Incorporating scattering through the piezoelectric interaction cannot substantially change either the magnitude or shape of the Γ curve.[4]

[1]A. A. Demidenko, M. V. Lebedev, S. I. Pekar *et al.*, Zh. Eksp. Teor. Fiz. **89**, 330 (1985) [Sov. Phys. JETP **62**, 185 (1985)].
[2]A. A. Demidenko, S. I. Pekar, and B. E. Tsekvava, Fiz. Tverd. Tela (Leningrad) **27**, 741 (1985) [Sov. Phys. Solid State **27**, 457 (1985)].
[3]G. K. Blasov, A. V. Kritskiĭ, and Yu. A. Kupchenko, Zh. Eksp. Teor. Fiz. **72**, 2063 (1977) (Sic.).
[4]V. V. Travnikov and V. V. Krivolapchuk, Zh. Eksp. Teor. Fiz. **85**, 2087 (1983) [Sov. Phys. JETP **58**, 1210 (1983)].
[5]O. S. Zinets and É. Skaĭstis, Litovskii Fiz. Sb. **16**, 871 (1976).
[6]V. A. Kiselev, B. S. Razbirin, and J. N. Uraltsev, Phys. Status Solidi B **72**, 161 (1975).
[7]Yu. V. Kryuchenko and V. J. Sugakov, Phys. Status Solidi B **111**, 177 (1982).

Translated by Dave Parsons