Direct observation of two polariton waves near the main exciton resonance in CdS crystals

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The dispersion of the index of refraction in the region of excitonic resonances at $T = 1.8$ K is investigated using the refraction of light by a thin CdS crystal with refraction angle $\sim 10^{-4}$ rad. The simultaneous transmission of two transverse $E \perp C$ waves is observed above the frequency of the longitudinal A_L exciton; the dispersion of the index of refraction and the spectral dependence of the transmission are measured for these waves. The basic parameters of the excitonic polaritons are determined.

One of the distinct manifestations of spatial dispersion in the region of excitonic resonances in CdS, predicted theoretically,[1,2] is the existence of two transverse waves with fixed frequency and polarization, distinguished by the dispersion of the index of refraction and by the transmission [according to Pekar—auxiliary light waves (ALW)]. Among the diverse and numerous experiments performed for the purpose of observing ALW, the spectroscopic investigations involving many-beam[3,4] and two-beam[5,6] interference accompanying transmission (reflection) of waves through thin crystal plates, measurements of group velocities of ALW with the help of picosecond laser technique,[7] and even the Mandel'shtam–Brillouin scattering associated with these waves[8] are all of great interest. A complete description of these and other experiments can be found in a monograph by Pekar.[2]

The most direct method for observing these waves and investigating the dispersive properties of the medium in the region of their propagation is the method of refraction of light by a prism-shaped crystal. This method was first used to investigate the dispersion of excitonic polaritons in CdS by Broser *et al.*,[9] and it was also used to study the dispersion properties of the mixed mode of excitonic polaritons in these crystals by Lebedev *et al.*[10] In these studies, however, it was not possible to observe two refracted waves simultaneously. In the present work, we observed the simultaneous transmission of two transverse waves through a thin (<1 μm) wedge-shaped CdS crystal and we spatially separated them.

For the investigations, we selected a plate-shaped CdS single crystal with a perimeter of 2×1 mm, grown from the gas phase, and shaped, in the transverse cross section, like a wedge with a refraction angle of α. The hexagonal C_6 axis lay in the plane of the plate parallel to the refracting edge. The plate thickness at the base of the wedge was 0.7 μm. The angle α was determined from the interference pattern—analyzed with the help of a microscope—formed by strips of equal thickness lying parallel to the edge of the wedge. The value of α was found to be 2.48×10^{-4} rad. The specimen, placed freely inside a specially prepared quartz cell, did not have any optical contacts with its supports, which eliminated stresses in the crystal. The specimen was submerged in superfluid helium ($T = 1.8$ K). The angle of incidence on the crystal θ could be varied by rotating the shaft to which the cell with the specimen was attached. To increase the accuracy of the angular measurements, the work was performed at large angles of incidence $\theta = 55.5°$.

A tunable organic dye laser, pumped by a pulsed N_2 laser (the spectral width of the laser was 0.3 Å), was used as the monochromatic source.

The crystal was oriented with a polarized monochromatic beam of light, which was collimated with the help of lenses and had a transverse cross section ~ 1 mm^2. The beam deflected by the crystal was focused onto the focal plane of the projecting lens ($f = 80$ mm) into an approximately 15-μm spot. Under the conditions corresponding to the maximum spatial separation of the $n^{(+)}$ and $n^{(-)}$ waves, the distance between the corresponding spots on the screen was about 740 μm. To achieve a higher degree of polarization, the crystal was placed between two polarizers. The displacement of the spot could be monitored visually with the help of a measuring microscope or photoelectrically by projecting the spot on the light-sensitive matrix of a OMA-2 optical multichannel analyzer. The use of photoelectric detection permitted monitoring the transmission of the waves through the crystal while measuring the refraction.

The angles of deflection by the prism φ, refraction α, and incidence θ are related

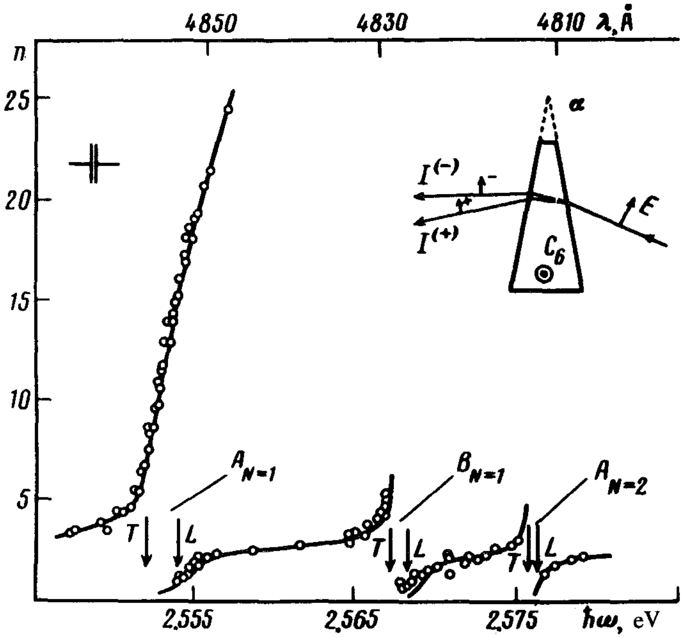

FIG. 1. Dispersion of the real part of the index of refraction in the region of the excitonic resonances $A_{N=1}$, $A_{N=2}$, $B_{N=1}$, measured at $T = 1.8$ K in $E \perp C$ polarization. The solid curves show the calculation for the values of the parameters indicated in the text. The inset in the upper right-hand corner is a schematic diagram of the experiment.

to the index of refraction n by the relation

$$\varphi = \alpha\left(\frac{\sqrt{n^2 - \sin^2\theta}}{\cos\theta} - 1\right).$$

This relation was used to calculate the index of refraction of the beam from the measured linear deflections of the collimated beam.

Figure 1 shows the dispersion of the real part of the index of refraction in the region of the excitonic resonances $A_{N=1}$, $A_{N=2}$, $B_{N=1}$, measured with polarization $E \perp C$ at $T = 1.8$ K. The arrows indicate the spectral positions of the longitudinal and transverse excitons in the corresponding regions. In the vicinity of the $A_{N=1}$ exciton, the effect of spatial dispersion is quite obvious. As is evident from Fig. 1, and also from Fig. 2, in which the region of the $A_{N=1}$ exciton is shown in greater detail, above the frequency of the longitudinal exciton there are two dispersion branches $n^{(+)}$ and $n^{(-)}$, corresponding to the simultaneous propagation in the crystal of two identically polarized waves which differ by their values of the index of refraction and transmission. The dispersion law of the $n^{(+)}$ wave approaches, as the energy is increased, the law of dispersion of the "mechanical" exciton. At the same time, the dispersion of the $n^{(-)}$ wave approaches a linear law as the distance from $n\omega_L$ increases, i.e., it becomes more light-like.

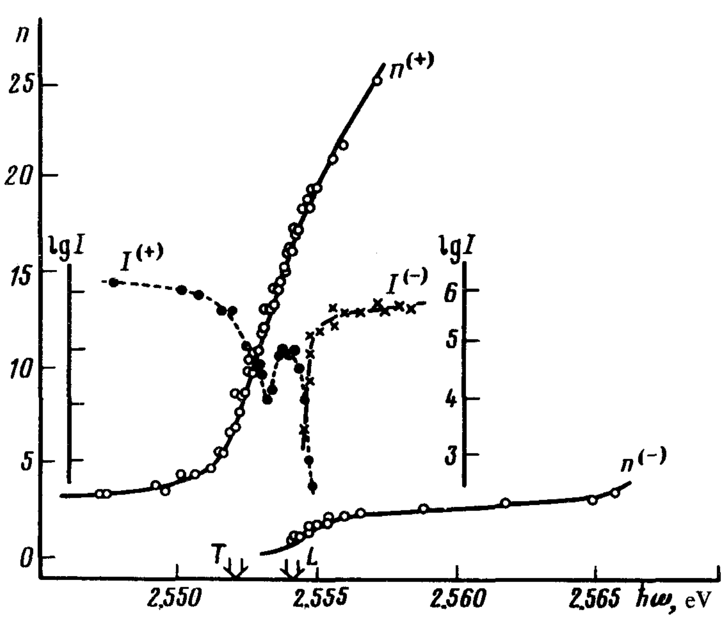

FIG. 2. Dispersion of the real part of the index of refraction and transmission of $n^{(+)}$ and $n^{(-)}$ waves in the region of the $A_{N=1}$ exciton. The solid curves show the calculation of dispersion of waves with the values of the parameters indicated in the text. The relative transmission for the $n^{(+)}$ wave (●) and for the $n^{(-)}$ wave (×) are shown on a logarithmic scale in arbitrary units. The dashed curves are drawn through the points corresponding to the observed transmission.

The solid curves in Figs. 1 and 2 show the dependence of the refractive indices of the $n^{(+)}$ and $n^{(-)}$ waves, calculated according to Pekar's formulas.[2] The effective spatial dispersion was included only for the $A_{N=1}$ exciton. The dielectric function of the medium in this approximation can be written in the form

$$\epsilon(\omega, k) = \epsilon_b \left[1 + \frac{\Delta^{LT}_{A(N=1)}}{\omega^0_{A(N=1)} + \dfrac{\hbar k^2}{2M_{ex}} - \omega - i\Gamma} + \frac{\Delta^{LT}_{B(N=1)}}{\omega^0_{B(N=1)} - \omega} + \frac{\Delta^{LT}_{A(N=2)}}{\omega^0_{A(N=2)} - \omega} \right].$$

In approximations making use of this expression, the resonance frequencies and the Δ^{LT} splittings for the $A_{N=2}$ and $B_{N=1}$ excitons were taken from Refs. 9 and 11. The best agreement with experiment was achieved for the following values of the adjustable parameters: background dielectric constant $\epsilon_b = 7.4$; translational mass of the $A_{N=1}$ exciton $M_{\text{ex}} = 0.8m_0$; L–T splitting $\Delta^{LT}_{A(N=1)} = 2.1$ meV. The resonance frequency $\omega^0_{A(N=1)}$ was found with the help of the expression $\omega^0_{A(N=1)} = \omega^L_{A(N=1)} - \Delta^{LT}$, where $\hbar\omega^L_{A(N=1)} = 2.554\,15$ eV is the energy of the longitudinal $A_{N=1}$ exciton, which was determined from the transmission spectra of the crystal measured in $\mathbf{E}\|\mathbf{C}$ polarization. The damping $\Gamma = 0.02$ meV was estimated from the transmission spectrum.[10]

As a result of the spatial separation of the $n^{(+)}$ and $n^{(-)}$ waves, it was possible to measure for the first time their transmission spectra (Fig. 2). In contrast to the monotonic transmission spectrum of the $n^{(-)}$ wave, the spectrum of the $n^{(+)}$ wave contains a narrow dip in the region of the L–T splitting. The transmission of the $n^{(+)}$ and $n^{(-)}$ waves are comparable near the energy of the longitudinal exciton. Far from this region the transmission of the waves differs by several orders of magnitude. The transmission curves, calculated from Pekar's theory,[12] with the additional boundary condition $P_{\text{ex}}|_{z=0} = 0$ (P_{ex} is the excitonic polarization) agree qualitatively with experiment. A detailed comparison of the theory and experiment for transmission curves of ALW, as well as an analysis of the dispersion of the index of refraction near $A_{N=2}$ and $B_{N=1}$, will be published elsewhere.

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