

# Anomalies of the dispersion of single-crystal CdS in the region of exciton absorption

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Direct measurements of the dispersion curve inside the  $A$ -exciton band of the CdS crystal were performed at  $T=4.2^\circ\text{K}$ . An appreciable deviation of the shape of this curve from the classical shape is observed and ascribed to the appearance of spatial-dispersion effects.

It has been shown theoretically<sup>[1,2]</sup> that allowance for spatial dispersion leads to a distinct shape, different from the classical one, for the dispersion curves of the refractive index within the exciton bands. We present here the first results of a direct measurement of the dispersion within the exciton band of a CdS crystal at  $4.2^\circ\text{K}$ . The form and magnitude of the dispersion deviate strongly from the usual classical values, and in our opinion represent a direct manifestation of the singularities of the spatial dispersion of the crystal.

The measurements were performed on thin single-crystal strips, in a contactless mounting, with the aid of a Jamin interferometer crossed with a spectrograph. Figure 1a shows a spectral scan of the interference pattern in the region of band  $A$  (polarization  $\mathbf{E} \perp \mathbf{C}$ ), obtained for a crystal  $0.33 \mu$  thick. We see that the interference bands can be clearly seen over the extent of the entire absorption band (marked by arrows), and the refractive index increases everywhere in this section. The anomaly of the dispersion, predicted by classical optics, is not observed in this case.

Since the  $A$  band is resolved only at  $\mathbf{E} \perp \mathbf{C}$  polarization, the value of  $n$  in its region remains practically unchanged at  $\mathbf{E} \parallel \mathbf{C}$ . This has made it possible to verify and confirm the foregoing results by measuring the birefringence of the same single crystal (No. 1). To this end, the sample was placed between two polarizers, and the direction of the oscillations of the electric vector of the light wave, passed by each of them, made an angle of  $45^\circ$  with the optical axis of the crystal. Figures 1b and 1c show the obtained birefringence pictures for parallel and crossed mutual arrangements of the polarizers, respectively. As expected, they turned out to be complementary: the maxima of one occur at the minima of the other. This proves unambiguously that the observed extrema are indeed the result of interference of

birefringence waves and cannot have any other origin.

On the basis of the presented data, we used two methods to calculate the values of the refractive indices  $n(\lambda)$  shown in Fig. 2. Curve 1 pertains to  $\mathbf{E} \perp \mathbf{C}$  and curve 2 to  $\mathbf{E} \parallel \mathbf{C}$ . The points denote the values obtained from the interference pattern of Fig. 1a, while the crosses



FIG. 1. Interference pattern (a) and birefringence of CdS single crystal No. 1 in parallel (b) and crossed (c) polarizers at  $4.2^\circ\text{K}$ .

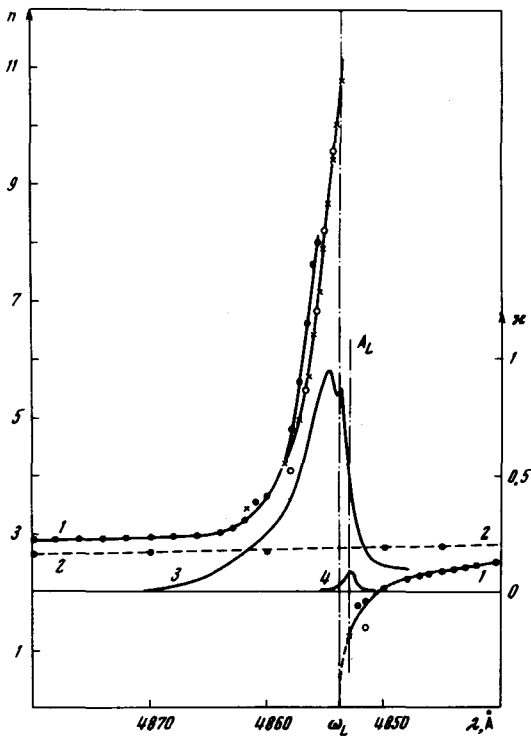


FIG. 2. Plots of the dispersion of  $n(\lambda)$  ( $E \perp C$ —1 and  $E \parallel C$ —2) and of the absorption  $\kappa(\lambda) = (\lambda/4\pi d) \ln(J_0/J)$  ( $E \perp C$ —3 and  $E \parallel C$ —4) in the region of the A-band of CdS at 4.2°K. The dash-dot lines denote  $\omega_L$  and  $A_L$ .

show the values calculated from the birefringence  $(n_1 - n_2)d = m\lambda$ . In addition, the same curve shows the values obtained from the birefringence of another single crystal (No. 2) 0.18  $\mu$  thick; these values are marked by circles. We see that all measurement runs agree well with one another and offer evidence of a monotonic and very steep increase of  $n$  up to 10.75.

Figure 2 shows also the A-band absorption curve measured by us earlier<sup>[3]</sup> at 4.2° for single-crystal No. 2. We see that at  $E \perp C$  (curve 3) the absorption band has a doublet character: there is a small narrow spike (4853.8 Å) on the short-wave side of the principal maximum (4855 Å). Its position does not coincide with the line of the longitudinal exciton  $A_L$  (4853 Å) observed with polarization  $E \parallel C$  (curve 4).

A comparison of curves 1 and 3 shows that the left-hand branch of the dispersion curve increases over the entire extent of the absorption band, and, reaches the supplementary maximum after passing through its principal maximum. The right-hand branch could be measured starting with values of  $n$  close to unity, and the spectral position of this start coincides with the position of  $A_L$  in the absorption spectrum. The obtained spread of the dispersion curve turns out to be eight times larger than expected on the basis of the maximum value of  $\kappa$  if the Kramers-Kronig relations are satisfied. In fact, the discrepancy between these relations in the case of CdS at 20°K was indicated by us already in<sup>[4]</sup>, where, however, we were unable to measure the  $n(\lambda)$  curve inside the absorption band.

The obtained dispersion curve and the contour of the

absorption band can be explained in general outline by assuming the presence of spatial dispersion in the crystal. In fact, the left-hand branch of the  $n(\lambda)$  curve, with its decrease beyond the absorption maximum, recall the calculated course of one of the branches of the normal waves ( $n'_+$  in<sup>[11]</sup> and  $n_2$  in<sup>[21]</sup>). On the whole, the  $n(\lambda)$  curve, if its left-branch is continued upward and the right-hand branch downward to the intersection with the abscissa (this section is shown dashed in Fig. 2), agrees qualitatively to the dispersion branches of polaritons without allowance for absorption (see, e.g.,<sup>[5]</sup>).

As to the absorption band, its shape for CdS with allowance for polariton-phonon interaction was calculated theoretically in<sup>[8]</sup>. It was shown that the absorption maximum should occur not at the frequency  $\omega_T$  of the transverse excitons, but should approach the longitudinal frequency  $\omega_L$ , which coincides with the point of intersection of the upper polariton branch with the energy axis at  $K=0$ . The measured curve with large negative asymmetry agrees well with the calculated one if  $\omega_L$  is taken to be the intercept of the right-hand branch of  $n(\lambda)$  on the abscissa axis, which coincides in frequency with the additional absorption maximum. In this case it can be assumed that the complicated shape of the absorption curve is a result of interaction of both polariton branches with the phonons, and the additional maximum on the curve reflects the contribution made to the absorption by processes of scattering from the upper branch.

The position of the point  $n=1$  on the dispersion curve is probably due to satisfaction of the momentum conservation law  $K_{\text{light}} = Q_{\text{polariton}}$ . The maximum momentum at which phononless excitation of the dispersion branch is possible corresponds to the wave vector of the light wave in vacuum.

Attention should be called also to one more peculiarity that follows from the results. According to the values of  $\kappa$  measured with crystal No. 2 (0.18  $\mu$ ), the light in the region of the A-band should be completely absorbed in the case of sample No. 1 (0.33  $\mu$ ). Yet we see inside the band a distinct set of interference fringes and a distinct birefringence picture, i.e., the wave carriers phase information in both measurement runs. This suggests that the effective  $\kappa$  is smaller for crystal No. 1 than for No. 2, i.e.,  $\kappa$  is thickness-dependent. As follows from calculations<sup>[9]</sup> in which spatial dispersion is taken into account, a certain "saturation" of the effective absorption coefficient can indeed take place with increasing thickness. We note that under the same experimental conditions, the dispersion in the B band, which has the same intensity at the maximum (according to measurements on the crystal No. 2), could not be measured.

In conclusion, the authors are sincerely grateful to A.S. Davydov for a useful discussion of the results.

<sup>1)</sup> We note that in<sup>[6,7]</sup> is given the lower polariton branch of CdS and both branches of the CdSe crystal, obtained indirectly from the luminescence spectrum of CdS<sup>[6]</sup> and the reflection spectrum of CdSe.<sup>[7]</sup>

<sup>5)</sup> S.I. Pekar, Zh. Eksp. Teor. Fiz. 33, 1022 (1957) [Sov.

Phys. -JETP 6, 785 (1958)]; Usp. Fiz. Nauk 77, 309 (1962) [Sov. Phys. -Usp. 5, 515 (1962)].

<sup>2</sup>A. S. Davydov and E. N. Myasnikov, Preprint ITF-74-4R, Kiev (1974).

<sup>3</sup>M. I. Strashnikova and A. T. Rudchik, Fiz. Tverd. Tela. 14, 984 (1972) [Sov. Phys-Solid State 14, 842 (1972)].

<sup>4</sup>M. S. Brodin and M. I. Strashnikova, Fiz. Tverd. Tela 4, 2454 (1962) [Sov. Phys. -Solid State 4, 1798 (1963)].

<sup>5</sup>R. S. Knox, Theory of Excitons, Suppl. 5 of Solid State

Physics, F. Seitz and D. Turnbull, eds., Academic, 1963.

<sup>6</sup>S. A. Permogorov and V. V. Travnikov, Fiz. Tverd. Tela 13, 709 (1971) [Sov. Phys. -Solid State 13, 586 (1971)].

<sup>7</sup>V. A. Kiselev, B. S. Razbirin, and I. N. Ural'tsev, ZhETF Pis. Red. 18, 504 (1973) [JETP Lett. 18, 296 (1973)].

<sup>8</sup>W. C. Tait and R. L. Weiher, Phys. Rev. 166, 769 (1968).

<sup>9</sup>A. S. Davydov and A. A. Eremko, Ukr. Fiz. Zh. 18, 1863 (1973).