

Natural birefringence of cubic CuBr crystals in the exciton part of the spectrum

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A birefringence of cubic crystals in the exciton part of the spectrum resulting from a difference in the translational masses of the transverse excitons has been detected experimentally for the first time.

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The optical anisotropy of cubic crystals which results from the spatial dispersion of the dielectric constant leads to a birefringence in the transparency region. The effect is proportional to $(a/\lambda)^2$, where a is a distance of the order of the lattice constant, and λ is the wavelength of the light in the crystal, and it should intensify near absorption bands.¹

In this letter we are reporting measurements of the birefringence Δn in cubic CuBr crystals. The values of Δn were measured by the standard interference method with light propagating along the [110] direction in the crystal. Figure 1 shows the dependence of Δn on the photon energy. The filled and open circles show the values of Δn measured with a spectrometer and with a tunable dye laser, respectively, for two different samples. The inset shows the intensity of the light passing through crossed polarizers as a function of the angle between the oscillation direction in the polarizer and the $[\bar{1}10]$ axis of the crystal.

These results are quite different from those found in Refs. 2 and 3. In those previous studies, birefringence was observed in Ge, Si, GaAs, GaP, ZnTe, and ZnSe crystals at room temperature. All of these crystals have a low exciton binding energy, and for this reason the birefringence observed in these cases is determined by interband transitions. The Z_{12} exciton state in the CuBr crystal has a large binding energy (~ 110 meV), and the exciton state may be predominant in the birefringence even at room temperature.

The CuBr crystal is an $A^I B^{VII}$ crystal with the zinc blende structure. Its upper valence band is fourfold degenerate and corresponds to the Γ_8 representation; the conduction band is twofold degenerate and corresponds to the Γ_6 representation. Corresponding to these bands is an exciton state $\Gamma_6 \times \Gamma_8$ (Z_{12}), which is eightfold degenerate at $q=0$ (the exchange interaction is neglected); this exciton state transforms in accordance with the following irreducible representations:

$$\Gamma_6 \times \Gamma_8 = \Gamma_3 + \Gamma_4 + \Gamma_5 \quad (\equiv \Gamma_{12} + \Gamma_{15} + \Gamma_{25}),$$

where the representation Γ_3 is twofold degenerate, and Γ_4 and Γ_5 are threefold degenerate. The states Γ_4 (Γ_{15}) are dipole-allowed, while the states Γ_3 and Γ_5 are di-

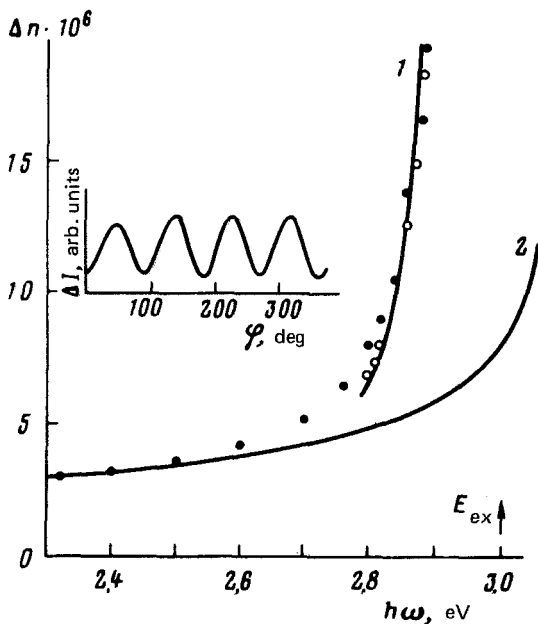


FIG. 1. Frequency dependence of Δn . Points—experimental; curves—theoretical.

pole-forbidden.

We believe that the natural birefringence observed by us at energies near the Z_{12} exciton is caused primarily by the state Γ_{15} . In the case $q \parallel [110]$ there may be an excitation of a longitudinal exciton polarized along \mathbf{q} and of transverse excitons polarized in the directions $[\bar{1}10]$ and $[001]$, which are perpendicular to \mathbf{q} . For light frequencies near the ground state of the Z_{12} exciton the dielectric tensor can be written as follows¹:

$$\epsilon_{\alpha\beta}(\omega, \mathbf{q}) = \epsilon_0 \delta_{\alpha\beta} + \frac{\pi e^2}{\hbar\omega^2 m_0^2} \sum_i \frac{P_i^\alpha(\mathbf{q}) P_i^\beta(\mathbf{q})}{\hbar\omega - E_i(\mathbf{q})} \quad (1)$$

In (1), the index "i" indicates the exciton state Γ_{15} with the energy $E_i(\mathbf{q})$; $P_i^\alpha(\mathbf{q})$ is the α component of the matrix element for the optical excitation of the i th exciton (we assume that this component depends on only the direction of \mathbf{q} ; i.e., we ignore the quadrupole corrections); and ϵ_0 is the background dielectric constant, which incorporates the other excitations and nonresonant terms. If we write the dielectric constant $\epsilon_{\alpha\beta}$ in a coordinate system whose three axes (X, Y, Z) are the directions $[\bar{1}10]$, $[001]$, and $[110] \parallel q$, then the birefringence is determined by a difference:

$$n_o \Delta n = \epsilon_{XX} - \epsilon_{YY} \quad (2)$$

At small values of \mathbf{q} , such that the kinetic energy of the exciton is small in comparison with the energy "deficit" ($\hbar\omega - E_q - E_0$), we find from (1) and (2)

$$n_o \Delta n = \frac{Ax^2}{(1-x)^2} \left(\frac{1}{\mu_1} - \frac{1}{\mu_2} \right). \quad (3)$$

Here $x = E_0 / (E_g - \hbar\omega)$; E_0 is the binding energy of the Z_{12} exciton, $A = (\pi e^2 \hbar n_0^2 P^2 / 2m_0^2 c^2 E_0^2)$, and μ_1 and μ_2 serve as the masses of the transverse excitons.⁴

The part of the birefringence attributable to the dipole-allowed exciton state Γ_{15} results from a splitting of the exciton bands at $\mathbf{q} \neq 0$ and is determined by the difference between the translational masses μ_1 and μ_2 of the transverse excitons. Pekar and Tsekava⁵ have previously mentioned the possibility of a birefringence in cubic crystals in the case of different effective masses in two transverse exciton bands.

The theoretical curve 1 in Fig. 1 was plotted from Eq. (3) for $A (1/\mu_1 - 1/\mu_2) = 2.6 \times 10^{-3}$ in the exciton part of the spectrum ($\hbar\omega \approx 2.8 + 2.9$ eV). The good agreement with experiment indicates that the Γ_{15} exciton state plays a key role in this part of the spectrum.

The theoretical curve 2, shown for comparison, is plotted from an equation in Ref. 6 and describes the part of Δn which results from interband transitions:

$$|\Delta n| = a \left\{ \frac{1}{\sqrt{1 - \frac{\hbar\omega}{E_g}}} + \frac{1}{\sqrt{1 + \frac{\hbar\omega}{E_g}}} \right\}. \quad (4)$$

Curve 2 corresponds to the value $a = 1.3 \times 10^{-6}$, which was determined by fitting the calculated and experimental values of Δn in the long-wave part of the spectrum, where the exciton states are of minor importance. The theory which ignores the Coulomb interaction obviously does not give an accurate description of the experimental data in the exciton part of the spectrum. As was pointed out in Ref. 3, the interband birefringence results from an anisotropy of a degenerate valence band. This circumstance can be exploited to determine the difference in Luttinger constants $|\gamma_3 - \gamma_2|$ (Ref. 7). The expression for the constant a is

$$a = \frac{3\sqrt{2}}{2} \frac{e^2 s^2 \mathbf{q}^2 \sqrt{m_c}}{\sqrt{E_g} \hbar\omega^2 \left(1 + \frac{m_c}{m_o} \gamma_1 \right)^{5/2}} \frac{m_c}{m_o} |\gamma_3 - \gamma_2|, \quad (5)$$

where s is the matrix element of the velocity operator.

The difference $|\gamma_3 - \gamma_2|$ calculated from (5) for the CuBr crystal is ~ 0.06 .

This value could apparently be in error only on the high side. Optical inhomogeneities resulting from the crystal growth conditions might also contribute to the measured value. The situation is illustrated by Fig. 2, where curves 2 (for $q \parallel [110]$) and 3 ($q \parallel [100]$) show measurements for a sample having important optical inhomogeneities. Curve 1 corresponds to the sample with the best optical homogeneity.

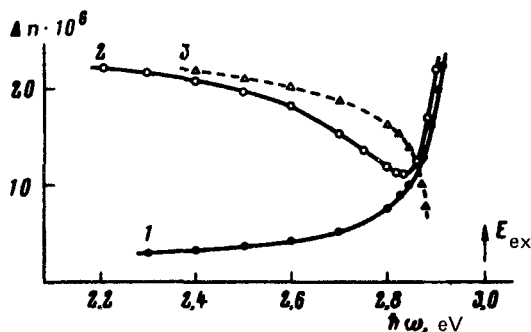


FIG. 2. Dispersion of Δn for various samples.

Analysis of the dispersion of Δn in the CuBr crystals suggests that the actual value of the difference $(\gamma_3 - \gamma_2)$ should be less than that determined in the present experiments. This value is approximately equal to the theoretical value from Ref. 8 ($\gamma_3 - \gamma_2 = 0.02$), but it is markedly different from the value measured in Ref. 9 ($\gamma_3 - \gamma_2 = 0.3$).

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1. V. M. Agranovich and V. L. Ginzburg, *Kristallogoptika s uchetom prostranstvennoy dispersii i teoriya eksitonov* (Optics of Crystals with Spatial Dispersion and Theory of Excitons), Nauka, Moscow, 1979.
2. J. Pastrnak and K. Vedam, *Phys. Rev. B* **3**, 2567 (1971).
3. P. Y. Yu and M. Cardona, *Solid State Commun.* **9**, 1421 (1971); M. Bettini and M. Cardona, *Proceedings of the Eleventh International Conference on the Physics of Semiconductors, Warsaw, 1072, 1972.*
4. E. O. Kane, *Phys. Rev. B* **11**, 3850 (1975).
5. S. I. Pekar and B. E. Tsekvava, *Fiz. Tverd. Tela (Leningrad)* **2**, 261 (1960) [*Sov. Phys. Solid State* **2**, 242 (1960)].
6. E. G. Tsitsishvili, *Fiz. Tekh. Poluprovodn.* (in press).
7. Yu. N. Berozashvili, S. Z. Machavariani, A. G. Natsvlishvili, E. G. Tsitsishvili, and V. Sh. Ėdilashvili, *Fiz. Tekh. Poluprovodn.* (1981).
8. M. A. Khan, *Phys. Status Solidi* **60**, 641 (1973).
9. H. J. Mattausch and Ch. Uihlein, *Solid State Commun.* **25**, 447 (1978).

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