

Effect of inversion of the optical axis of the CdS crystal

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We prove for the first time ever, with experiments on light reflection, the optical nonequivalence of the two opposite directions of the polar axis of a crystal of class C_{6v} . The experimental data on the reflection of light in CdS crystals demonstrate that the nonequivalence of the two orientations is due to gyrotropy of these crystals.

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Gyrotropy of crystals is usually taken to mean the ability of rotating the polarization plane of light propagating along a symmetry direction in the gyrotropic crystal. In fact, 15 out of 18 classes of gyrotropic crystals have this property; the direction of rotation makes it possible in this case to classify uniquely the crystals as either dextrorotary or levorotary.¹ At the same time, crystals with symmetry C_{3v} , C_{4v} , and C_{6v} have no rotating ability at any light-propagation direction, i.e., they occupy a special position among the gyrotropic crystals. The gyrotropy of such crystals leads to a “longitudinal” elliptical polarization of the extraordinary waves, wherein the wave vector \mathbf{k} lies in the plane of the polarization ellipse.²⁻⁴ The question is, what is the analog of dextro- or levorotational ability of these crystal classes, and with what observable effects is it connected?

In rotating gyrotropic crystals, the direction of rotation of the plane of polarization of the light is determined by the sign of the gyration tensor. In the case of crystals of symmetry C_{3v} , C_{4v} , and C_{6v} , the change of the sign of the gyration tensor corresponds to a change of the direction of rotation of the electric-field intensity vector \mathbf{E} of the extraordinary wave in a plane containing the optical axis \mathbf{c} and the vector \mathbf{k} . On the other hand, it is easily seen that a similar effect can be obtained by orienting the crystal axis in the opposite direction. In other words, the same crystal is simultaneous-

ly "dextrorotary" and "levorotary," depending on how it is oriented. This is in fact the essential difference between crystals of classes C_{3v} , C_{4v} , and C_{6v} , and all other gyrotropic crystals.

To identify the direction of the optical axis (or the sign of the gyration parameter), we can use the following light-reflection experiment (Fig. 1). Let the plane of

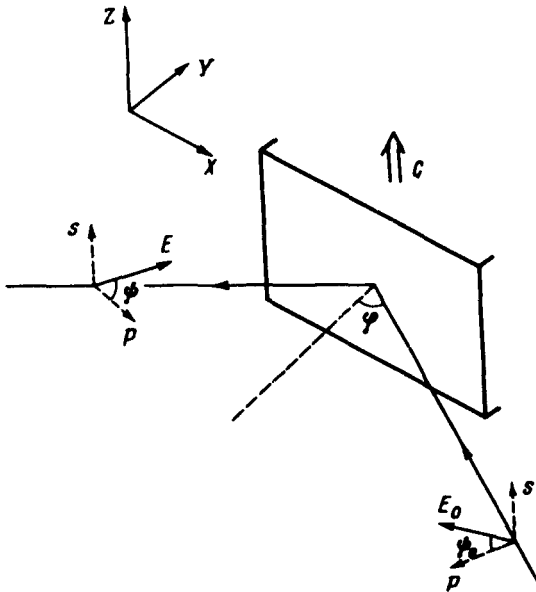


FIG. 1. Geometry of experiment.

incidence XY be perpendicular to the c axis, and let the light be obliquely incident on the reflecting boundary XZ of the crystal. If E_{0p} and E_{0s} are the p and s amplitudes of the incident wave, and if E_p and E_s are the analogous amplitudes of the reflected wave, then the general linear connection between them can be expressed in matrix form

$$\begin{pmatrix} E_p \\ E_s \end{pmatrix} = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} \begin{pmatrix} E_{0p} \\ E_{0s} \end{pmatrix}, \quad (1)$$

where the diagonal elements of the matrix \hat{r} are the amplitude coefficients of the reflection in terms of the p and s components ($p \rightarrow p$ and $s \rightarrow s$). The off-diagonal r_{ps} and r_{sp} describe the reflection in the orthogonal configurations $s \rightarrow p$ and $p \rightarrow s$, respectively.

From the invariance of the crystal to time reversal it follows that

$$(\tilde{\mathbf{e}}, \hat{r}(\tilde{\mathbf{k}}_0, \mathbf{k}_0) \mathbf{e}) = (\mathbf{e}^*, \hat{r}(-\mathbf{k}_0, -\tilde{\mathbf{k}}_0) \tilde{\mathbf{e}}^*),$$

where \mathbf{k}_0 and \mathbf{e} ($\tilde{\mathbf{k}}_0$ and $\tilde{\mathbf{e}}$) are the wave and polarization vectors of the incident (reflected) wave. Therefore in the case when one of the symmetry planes of the crystal is perpendicular to the reflecting face (case a) or the anisotropy in the plane of incidence is negligible (case b), the coefficients r_{ps} and r_{sp} should coincide. Within the limits of errors, experiments yield in fact $|r_{ps}| = |r_{sp}|$.³

We consider now the case when the vectors \mathbf{e} and $\bar{\mathbf{e}}$ correspond to linear polarization of the polarizer and analyzer. Let ψ_0 and ψ be the azimuths of the polarizer and analyzer, respectively, and let ϕ be the incidence angle (Fig. 1). We can then obtain from (1), taking into account the symmetry to time reversal and the equality $r_{ps} = r_{sp}$, the energy reflection coefficients $R_+(\psi_0, \phi, \psi)$ and $R_-(\psi_0, \phi, \psi)$ corresponding to opposite orientations of the polar axis:

$$R_{\pm}(\psi_0, \phi, \psi) = \left| (r_{pp} \cos \psi \cos \psi_0 + r_{ss} \sin \psi \sin \psi_0) \pm r_{ps} \sin(\psi_0 + \psi) \right|^2. \quad (2)$$

It follows therefore that when the crystal gyrotropy is taken into account ($r_{ps}, r_{sp} \neq 0$) the coefficients R_+ and R_- are unequal if the expression in the round parentheses under the absolute-value sign or $\sin(\psi_0 + \psi)$ is not equal to zero. We note that $R_-(\psi_0, \phi, \psi) = R_+(180^\circ - \psi_0, \phi, 180^\circ - \psi)$. Measurement of R_- therefore does not call for rotating the crystal through 180° , and it suffices to replace, at a fixed crystal position, the azimuths ψ_0 and ψ by $(180^\circ - \psi_0)$ and $(180^\circ - \psi)$ respectively. In the particular case $\psi_0 = \psi = 45^\circ$ we get from (2)

$$R_+(45^\circ, \phi, 45^\circ) - R_-(45^\circ, \phi, 45^\circ) = 2 \operatorname{Re} [r_{ps}^* (r_{pp} + r_{ss})]. \quad (3)$$

Figure 2 shows the reflection spectra of CdS crystals ($T = 2$ K) in the region of

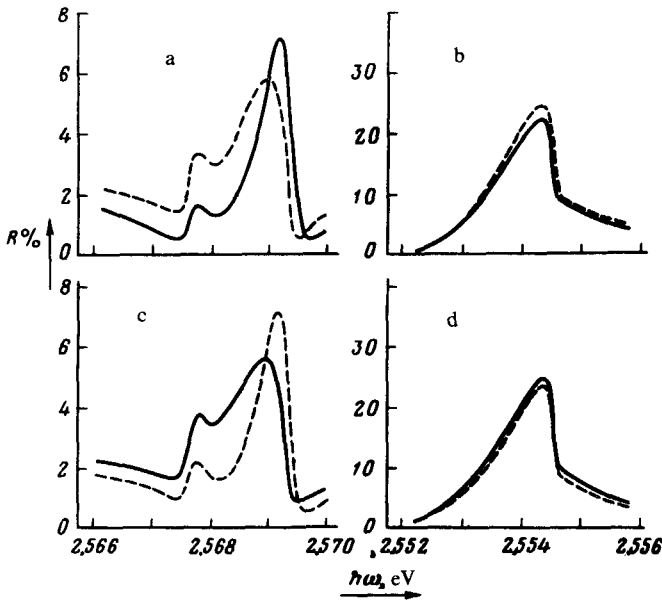


FIG. 2. Reflection spectra of CdS crystals ($T = 2$ K) in the region of excitonic resonances A_{n-1} (b,d) and B_{n-1} (a,c) for the configurations $(45,45,45^\circ)$ (a,b) and $(135,45,135^\circ)$ (c,d). The solid and the dashed curves correspond to opposite orientations of the optical axis.

the A_{n-1} and B_{n-1} exciton resonances in the configurations $(\psi_0, \phi, \psi) = (45,45,45^\circ)$ (a,b) and $(135,45,135^\circ)$ (c,d). The solid and dashed curves correspond to mutually

opposite orientations of the optical axis of the sample in space. Figures 2a and 2c show clearly the effect of the inversion of the polar axis, which attests to optical activity (gyrotropy) of the CdS crystals in the region of the $B_{n=1}$ resonance. At the same time, in the resonance region of the A exciton (Figs. 2b and 2d) the effect is practically nonexistent ($R_+ \approx R_-$), as should be the case for an A exciton.³

In the vicinity of the $B_{n=1}$ exciton resonance, as is noted in Ref. 3, the gyrotropy of the CdS crystals is due to the contribution of the terms linear in the wave vector \mathbf{K} to the energy spectrum of the excitons. Consequently, from the spectra measured in the present paper we can determine the sign of the terms linear in \mathbf{K} in the exciton Hamiltonian.

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