# Magnetically induced spatial dispersion of crystals in the exciton region of the spectrum

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(Submitted 17 December 1982)
Pis'ma Zh. Eksp. Teor. Fiz. **37**, No. 3, 137–139 (5 February 1983)

A resonant increase in magnetically induced spatial dispersion of axes in the vicinity of exciton states in crystals without a center of inversion is predicted. It is found that this effect appears distinctly in regions where the dispersion branches of magnetopolaritons in CdS and CdSe crystals intersect.

PACS numbers: 71.36. + c

Magnetically induced spatial dispersion (MISD) of crystals, linear in the wave vector **k** and external magnetic field **H**, was analyzed by Agranovich and Ginzburg.[2] It is described by the pseudotensor **A** of rank 4:

$$\delta \epsilon_{\alpha\beta} ( \omega , \mathbf{k}, \mathbf{H} ) = A_{\alpha\beta\gamma\delta} (\omega) H_\gamma k_\delta \, . \tag{1}$$

Since the dielectric constant tensor in media without magnetic ordering satisfies the symmetry principles for kinetic coefficients

$$\epsilon_{\alpha\beta} ( \omega, \mathbf{k}, \mathbf{H}) = \epsilon_{\beta\alpha} ( \omega, -\mathbf{k}, -\mathbf{H} ), \tag{2}$$

the tensor $A_{\alpha\beta\gamma\delta}$ is symmetrical relative to interchange of the indices $\alpha$ and $\beta$. For this reason, in contrast to natural optical activity, electrogyration, and tensogyration, the contribution of (1) leads to birefringence,[1)] rather than to rotation of the plane of polarization of linearly polarized light.

MISD was observed experimentally in CdS crystals in well-known work on the effect of inversion of a magnetic field (see, for example, Ref. 3; other references are given in Ref. 1) and in $LiIO_3$ crystals.[4] In these papers, the effects stemming from the change in one of the diagonal components of the dielectric constant tensor $\varepsilon_{aa}$ in a magnetic field (its imaginary part in the case of CdS and real part in the case of $LiIO_3$) were studied.

  

In the present work, we observed the manifestation of the nondiagonal components of the tensor $\varepsilon_{\alpha\beta}$, induced by a magnetic field and odd with respect to **H** as well as **k** ($x_\alpha, x_\beta$ are the principal symmetry axes of the crystal). Thus, magnetically induced spatial dispersion of the axes, the dependence of the orientations of the principal axes of the tensor $\varepsilon_{\alpha\beta}(t-t', \mathbf{r}-\mathbf{r}', \mathbf{H})$ on the coordinate differences $(\mathbf{r}-\mathbf{r}')$ determined by (1), has been observed for the first time. Low-temperature ($T=1.6$ K) transmission spectra of thin (thickness 0.5–50 $\mu$m) CdS and CdSe crystals in the vicinity of $A_{n=1}$ and $B_{n=1}$ excitons were investigated. The magnetic field **H** was oriented parallel to the $C_6$ axis ($z$ axis); the light propagated along the axis $y \perp C_6$; the light incident on the crystal was polarized along the axis $x \perp C_6$; light polarized along the $z$ axis was analyzed at the output. In the geometry indicated, the output signal is proportional to the quantity

$$F = | \epsilon_{xz}^{(\perp)} / (\epsilon_{xx}^{(\perp)} - \epsilon_{zz}^{(\perp)}) |^2$$

(under the condition that $F \ll 1$). Here $\varepsilon_{\alpha\beta}^{(\perp)}$ is the transverse dielectric constant tensor. In the vicinity of the exciton resonance $A_{n=1}$ for $\mathbf{H} \| z$, $\mathbf{k} \| y$

$$\epsilon_{xz}^{(\perp)} = \epsilon_{xz} - \frac{\epsilon_{xy}\epsilon_{yz}}{\epsilon_{yy}} = \epsilon_{\perp}^{(0)} \frac{a k_y}{d_0} \frac{\omega_{LT}\Omega_{\|}/2}{(\omega-\omega_1)(\omega-\omega_2)}, \quad (3)$$

where $\varepsilon_{\perp}^{(0)} = \varepsilon_{xx}^{(0)} = \varepsilon_{yy}^{(0)}$ is the background dielectric constant; $\hbar\Omega_{\|} = g_{\|}\mu_B H_z$, $g_{\|}$ is the $g$ factor; $\mu_B$ is the Bohr magneton; $\omega_{LT}$ is the longitudinal-transverse splitting; $\omega_{1,2} = \omega_0(\mathbf{k}) \pm (\Omega_{\|}/2) - i\gamma$; $\omega_0(\mathbf{k})$ is the energy of the mechanical exciton including the diamagnetic shift; $d_0$ is the matrix element for optical excitation of the exciton $\Gamma_5$ in the dipole approximation; $ak_y$ is the correction, linear in **k**, to this matrix element; and $\gamma$ is the damping of the exciton. For brevity, we omit the expression for $\varepsilon_{\alpha\alpha}^{(\perp)}$.

Figure 1b shows the dispersion curves for normal light waves (polaritons) in CdSe, calculated in a magnetic field including terms linear in **k**,[5] but ignoring spatial dispersion induced by the magnetic field, i.e., for $\varepsilon_{xz}^{(\perp)} = 0$. It is evident that in the

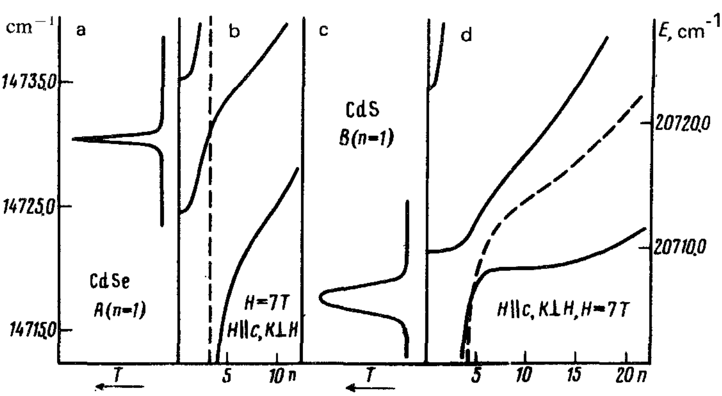


FIG. 1. Dispersion of magnetopolaritons (b, d) and transmission spectra (a, c) in crossed fields [1a] in a magnetic field $H=7$ $T$ in the geometry [1b] in the region of exciton states: $A_{n=1}$ in CdSe (a, b) and $B_{n=1}$ in CdS (c, d).

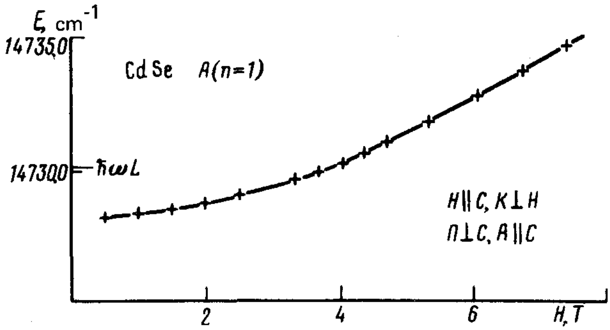


FIG. 2. Dependence of the energy of the transmitted signal on the magnitude of the magnetic field in the region of the exciton state $A_{n=1}$ in CdSe.

region of longitudinal-transverse splitting there is an isotropic point $\omega_{\text{meas}} \approx \omega_0(0) + \omega_{LT}$, at which the branches of two normal waves intersect (branch 2 and the dashed straight line). As the isotropic point is approached, the modulus of the difference $(\varepsilon_{xx}^{(\perp)} - \varepsilon_{zz}^{(\perp)})$ greatly decreases, which must lead to a sharp increase in the quantity $F$, and therfore to resonant behavior of the signal near $\omega_{\text{meas}}$ with crossed polarizer and analyzer (with half-width of the order of the exciton damping).

In accordance with the prediction of the theory, when an external magnetic field is switched on in CdSe, a transmitted signal is observed in crossed polaroids in the region of the longitudinal-transverse splitting of the $A_{n=1}$ state (Fig. 1a). The signal was reliably recorded starting with fields $H = 0.5$ T (at $H = 0$ the signal was completely missing); as the field increased to 7 T, intensity of the signal increased by more than an order of magnitude, while the half-width remained small ($\sim 0.7$ cm$^{-1}$). Investigation of the magnetopolaritonic dispersion[6] and the dependence of the spectral position of the signal on the magnitude of the magnetic field (Fig. 2) permit correlating this transmission uniquely with the isotropic point. The effect is related, first, to mixing of the polariton $\Gamma_{5x}$ with the longitudinal exciton $\Gamma_{5y}$ in the magnetic field $\mathbf{H} \parallel z$ and, second, to the polarization of the medium induced by the field $E_z$ along the $y$ axis due to the contribution, linear in $k_y$, to the matrix element for the optical excitation of the exciton $\Gamma_5$.

It was also possible to observe MISD in CdS crystal near the ground state of the $B$ exciton. The transmission signal arising in the magnetic field is shifted by 8 cm$^{-1}$ toward lower energies from the resonance $B_{n=1}$ (see Figs. 1c and 1d). In this case, the effect arises as a result of mixing of the states $\Gamma_{5x}$ and $\Gamma_{5y}$ in the magnetic field and mixing, due to terms linear in **k**, of the longitudinal exciton $\Gamma_{5y}$ and the polariton $\Gamma_1$ (in the notation used in Ref. 7 these terms are proportional to the constant $\beta_1$). The effect also results from mixing of the states $\Gamma_1$ and $\Gamma_2$ in a magnetic field and mixing, linear in **k** and proportional to the constant $\beta_2$, of the exciton states $\Gamma_2$ and $\Gamma_{5x}$.

A detailed calculation of the MISD signal and comparison with experimental data will be given in a separate paper.

The main feature of the phenomenon observed—resonant behavior in the region of intersection of the dispersion curves—serves as a reliable and accurate method for determining the isotropic points of magnetopolaritons in crystals without an inversion center. The study of this phenomenon could provide a sensitive method for observing restructuring of the energy spectrum in the field of an intense light wave.

[1)]Onsager's principle (2) was ignored in Ref. 2, and as a result it was concluded incorrectly that additional optical rotation proportional to $H_z k_z$ ($z$ is the principal optical axis of crystals with symmetry $C_3$ and $C_6$) can arise.


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Translated by M. E. Alferieff
Edited by S. J. Amoretty