Natural optical activity of CdS crystals in the exciton region of the spectrum

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We have investigated experimentally, for the first time ever, the natural optical activity (NOA) of CdS crystals. The reflection spectra have revealed a special type of NOA, which is typical of the crystal classes C_{3v}, C_{4v}, and C_{6v}.

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Natural optical activity (NOA) can be possessed by crystals whose symmetry admits of linear terms in the expansion of the dielectric tensor in powers of the wave vector **K**.[1] NOA is frequently identified with rotation of the plane of polarization of linearly polarized light when the light propagates through the crystal. At the same time, there are classes of crystals (C_{3v}, C_{4v}, C_{6v}), that admit of NOA but do not have the property of rotation of the plane of polarization for any direction of light propagation.[1,2] We report here, for the first time ever, experimental observation of NOA in crystals with symmetry C_{6v} (CdS).

In the crystals of the three classes indicated above, just as in uniaxial inactive crystals, "ordinary" (transverse) and "extraordinary" (mixed) waves can be excited. However, as the result of the NOA, the extraordinary wave in these crystals is electrically polarized. The polarization ellipse lies in the plane containing the optical axis **C** and the vector **K**.

NOA of this kind can be observed experimentally by investigating the reflection spectrum. In fact, let the crystal axis be perpendicular to the incidence plane and let the light be incident at a certain angle ϕ on the crystal face containing this axis (Fig. 1). Then, in the case of s polarization of the incident light (in the considered case $\mathbf{E}\|\mathbf{C}$) at $\phi\neq 0$ in reflected light, beside the s component, there will be observed a p component ($\mathbf{E}\perp\mathbf{C}$). The change of the light polarization upon reflection ($s\to p$) is due to the fact that the longitudinal component $E_l\|K$ of the wave-refraction polarization ellipse has a nonzero projection on the reflecting face. On the other hand, if $\phi=0$, then this projection is equal to zero and the polarization of the light is not altered upon reflection. Similar changes of the polarization can be observed also in the case $p\to s$, when the incident light has p polarization.

Far from the resonant-absorption bands, the intensity of the crossed component of the reflected light is proportional to $(a/\lambda)^2$, where a is quantity on the order of the lattice constant and λ is the wavelength of the light in the crystal. Under nonresonance conditions this quantity is small and it is difficult to record the NOA. In the resonant

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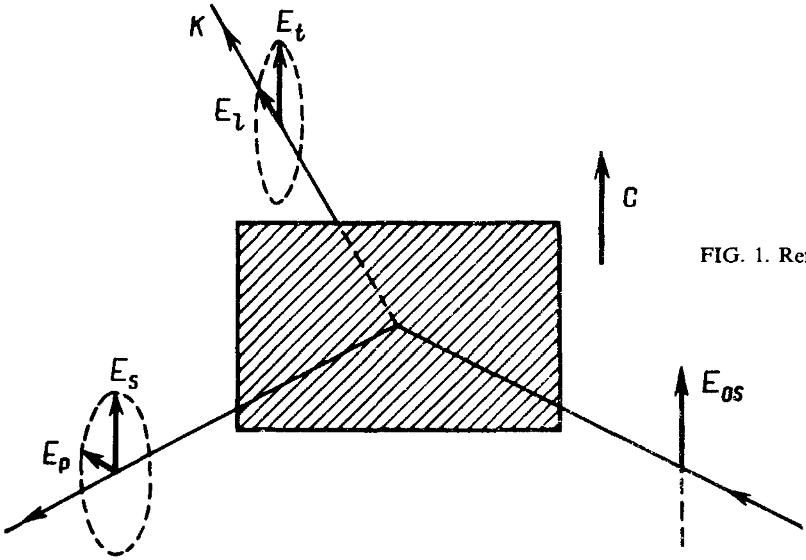

FIG. 1. Reflection geometry.

optical-exciton region, however, where the wavelength of the light in the crystal is substantially decreased, the effects due to the NOA should increase significantly (as well as other effects due to spatial dispersion).[1]

The reflection spectra in CdS crystals were measured at $T=2$ K in the geometry indicated above in the vicinity of the excitonic resonance $B_{n=1}$. The dashed line in Fig. 2(a) shows the reflection spectrum in the p-component $\mathbf{E}\perp\mathbf{C}$ at normal incidence $\phi\approx 0$

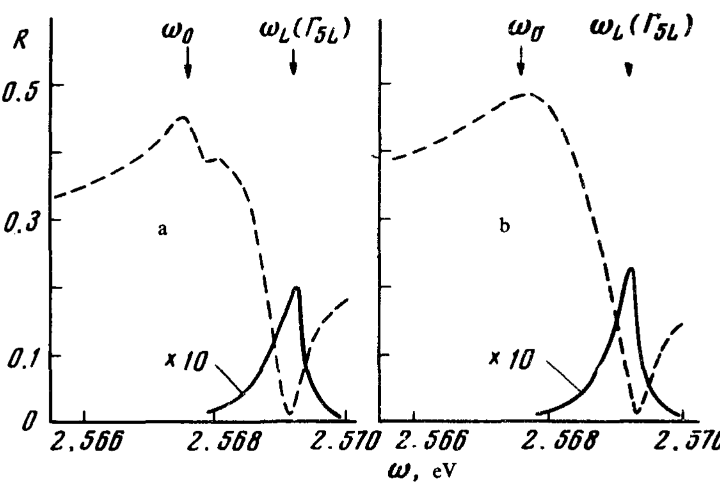

FIG. 2. Reflection spectra: a) $p\to(\mathbf{E}\perp\mathbf{C}\to\mathbf{E}\perp\mathbf{C})$—dashed curve, $p\to s$ $(\mathbf{E}\perp\mathbf{C}\to\mathbf{E}\|\mathbf{C})$—solid curve; b) $s\to s$ $(\mathbf{E}\|\mathbf{C}\to\mathbf{E}\|\mathbf{C})$—dashed curve, $s\to p$ $(\mathbf{E}\|\mathbf{C}\to\mathbf{E}\perp\mathbf{C})$—solid curve.

($p \to p$). The solid lines shows the reflection spectrum in crossed polarizations $p \to s$ at $\phi = 45°$. The dashed line in Fig. 2(b) is the spectrum in the s component $s \to s$ at $\phi \approx 0$ ($\mathbf{E} \| \mathbf{C}$), while the solid line corresponds to crossed polarizations $s \to p$ at $\phi = 45°$. As $\phi \to 0$ the crossed component of the reflected light is practically absent both for the case $s \to p$ and for the case $p \to s$. In the exciton region $A_{n=1}$ the effect of the change of the polarization is observed only at the sensitivity limit of the appratus.

The appearance of the signal in crossed polarizations (Fig. 2) is unexpected and inexplicable from the point of view of ordinary Fresnel crystal optics in the considered high-symmetry experimental geometry. However, the obtained experimental data can be explained in main outline within the framework of the theory of optical excitons, which takes into account terms linear in **K** in the energy spectrum of the excitons.

The symmetry of crystals with wurtzite structure admits of the existence of corrections, linear in **K**, to the exciton Hamiltonian; these corrections are determined by two constants β_1 and β_2.[3] At $\mathbf{K} \perp \mathbf{C}$ allowance for the constant β_1 leads to a mixing of the polaritons β_1 with the longitudinal excitons Γ_{5L}, while allowance for the constant β_2 leads to a mixing of the transverse polaritons Γ_{5T} with the dipole-forbidden excitons Γ_2. The corrections connected with β_2 were taken into account by Mahan and Hopfield[4] in an analysis of the reflection spectra of CdS in the $B_{n=1}$ region at normal incidence of the light. They have shown, in particular, that in the vicinity of the resonant frequency of the exciton $B_{n=1}$ the constant β_2 gives rise to an additional structure (see Fig. 2(a), $p \to p$). This structure is not connected with NOA, since the mixing of the states Γ_2 and Γ_{5T} leads only to "flareup" of the dipole-forbidden state Γ_2, but the normal waves remain purely transverse. The NOA, and consequently also the appearance of a signal in crossed polarizations, is connected with the constant β_1. Namely, at $\beta_1 \neq 0$ the mixing of the transverse excition Γ_1 with the longitudinal excition Γ_{5L} leads to the existence of elliptically polarized waves of the type described above. In fact, the reflection spectra in crossed polarizations $s \to p$ and $p \to s$ have a sharp maximum at the frequency of the longitudinal excition Γ_{5L}, corresponding to the principal minimum of reflection in the $p \to p$ spectrum (Fig. 2a). The existence of the constant β_1 might lead also to the appearance of an additional singularity in the $s \to s$ reflection spectrum at the frequency of the longitudinal excition Γ_{5L}. In CdS crystals, however, in polarization $s \to s$, the principal minimum of the spectrum falls in this region, and this apparently hinders the observation of this structure.

We note that the contribution of terms linear in **K** to the energy spectrum of the excitons $A_{n=1}$ should not lead to NOA. This explains why the cross-polarization signal is small in experiments performed in the region of the excition $A_{n=1}$.

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