Manifestation of optical activity in exciton reflection spectra of β-AgI crystals

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The optical activity typical of crystal classes C_{3v}, C_{4v}, and C_{6v} was observed in AgI crystals with wurtzite structure. The results of a theoretical calculation of the reflection spectra in the region of the exciton resonance B $n=1$ are compared with the experimental data.

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Among the crystals having natural optical activity (NOA), a special place is occupied by crystals belonging to the classes C_{3v}, C_{4v}, and C_{6v}.[1,2] The NOA in such crystals leads to elliptical polarization of the extraordinary wave, such that the polarization ellipse lies in the plane that contains the optical axis C of the crystal and the wave vector **k** of the normal wave.[2]

The first experimental observation of NOA of CdS of the Wurtzite type was recently reported.[3] In the present study we have investigated, for the first time ever, the NOA of AgI crystals (4H polytype, Wurtzite structure, C_{6v}^4) in the exciton region of the spectrum. In contrast to[3], the NOA was registered in the ordinary reflection spectrum with $\mathbf{E}\|\mathbf{C}$ polarization of the light.

The AgI single crystals were grown by diluting saturated aqueous KI–AgI solutions, and had natural faces parallel and perpendicular to the hexagonal axis **C**. The reflection spectra were investigated at $T=4.2$ K, both from the end faces and from the lateral faces.

The solid curves in Fig. 1 show the experimental reflection spectra obtained at normal incidence of the light on the lateral face of the crystal ($\mathbf{K}\perp\mathbf{C}$), at identical polarizations $\mathbf{E}\perp\mathbf{C}$ (a), and $\mathbf{E}\|\mathbf{C}$ (b) of the incident and reflected light relative to the **C** axis. The investigated section of the spectrum corresponds to the region of the exciton resonance B $n=1$.

The shape of the reflection spectrum in Fig. 1a is very close to that observed in the case of CdS crystals[3,4] in an analogous experimental geometry in the region of the resonance B $n=1$. It is therefore natural to connect the singularity in the region of the principal maximum of the reflection with the "flareup" of the optically inactive state Γ_2 on account of the terms linear in **K** in the energy spectrum of the exciton. When the light is reflected from an end face ($\mathbf{K}\|\mathbf{C}$), this singularity is not observed, just as in the case of CdS.

The experimental reflection spectrum shown in Fig. 1b ($\mathbf{E}\|\mathbf{C}$, $\mathbf{K}\perp\mathbf{C}$) differs substantially from the corresponding spectrum of the CdS crystals. First, the half-width of the reflection band in the polarization $\mathbf{E}\|\mathbf{C}$ greatly exceeds the half-width in the polarization $\mathbf{E}\perp\mathbf{C}$. Second, in the region of the principal maximum of the reflection one observes a dip located at the frequency of the principal minimum of the reflection in the polarization E⊥C. This correlation in the spectral arrangement of the singularities in the spectra (a) and (b) allows us to conclude that the dip in the spectrum (b) is due to the participation of the longitudinal excitons Γ_{5L}. The most probable cause of the appearance of longitudinal excitons in the **E**||**C** spectrum is the mixing of the logitudinal states Γ_{5L} with the transverse states Γ_1 on account of the terms linear in **K** in the energy spectrum of the exciton *B*.

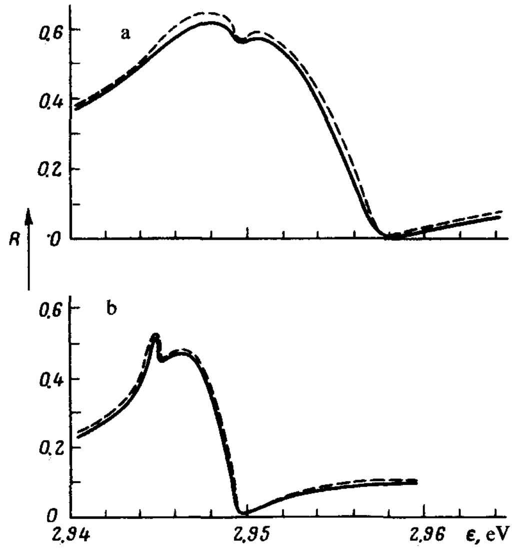

FIG. 1. Experimental (solid curves) and theoretical (dashed) reflection spectra for normal incidence of the light in the experimental geometry **K**⊥**C**, **E**⊥**C** (a) and **E**||**C** (b).

The symmetry of crystals of the Wurtzite type admits of the existence of two independent constants β_1 and β_2, which determine the contributions $\beta_1\mathbf{K}$ and $\beta_2\mathbf{K}$ of the terms linear in **K** in the energy spectrum of the excitons *B* $n=1$ and intermix the states Γ_1 and Γ_{5L}, as well as Γ_2 and Γ_{5T}, respectively.[5] Consequently, the additional singularity in the **E**||**C** spectrum can be due only to β_1, whereas the singularity in the **E**⊥**C** spectrum is due only to the constant β_2.[4]

To calculate the reflection spectra in the exciton region of the spectrum it is necessary, as is well known,[1] to have additional boundary conditions (ABC), and their number in our situation should be four in accordance with the 4-fold degeneracy of the exciton level *B* $n=1$. We can use as the ABC the condition that the wave functions of the four exciton states Γ_1,Γ_2,Γ_{5T} and Γ_{5L} vanish on the crystal boundary; this corresponds to an exciton potential in the form of an infinitely high wall at the boundary itself. These conditions are a generalization of the ABC used previously in the analysis of the reflection spectra of CdS crystal[4] and agree well with the experimental data. To take into account the difference between the model potential and the real one, we have also introduced, as was done in[4], a "dead" exciton-free layer of thickness *l* with a background permittivity ϵ_0, and specified the ABC on the internal surface of this layer.

The dashed curves in Fig. 1 show the results of the theoretical calculation of the reflection spectra for the experimental geometry corresponding to the experiment: **E**⊥**C** (a) and **E**||**C** (b).

We used in the calculations the following parameters of the exciton resonance B $n=1$. Resonant frequency $\omega_0=2945.2$ meV, logitudinal–transverse splittings $\omega_{LT}^{\perp}=4.2$ meV and $\omega_{LT}^{\|}=11.6$ meV for the states Γ_5(**E**⊥**C**) and Γ_1(**E**||**C**), respectively; $|\beta_1|\omega_0/c=|\beta_2|\omega_0/c=0.15$ meV; translational mass of the exciton $M=0.8\ m_0$ (m_0 is the mass of the free electron, **K**⊥**C**); background permittivities $\epsilon_0^{\perp}=5.0$ (for **E**⊥**C**) and $\epsilon_0^{\|}=6.5$ (for **E**||**C**), damping parameters $\gamma_{\perp}=0.3$ meV (for **E**⊥**C**) and $\gamma_{\|}=1.0$ meV (for **E**||**C**), $l=50$ Å.

We have also attempted to register a reflected signal in a geometry with a polarizer crossed with an analyzer at inclined incidence of the light, as was done in[3]. We did not succeed in realiably registering a signal. Our calculation of the reflection spectra for this experimental geometry shows that in the case of the parameters cited above the maximum value of the reflection coefficient is lower by one order of magnitude than the case of the parameters of CdS. On the other hand, calculation shows that at the parameters of the CdS crystal, in the geometry correspoding to Fig. 1b, there is no additional structure, and this agrees with the experimental data on CdS.[3]

Thus, the NOA of Wurtzite crystals is apparently a common phenomenon that can manifest itself under various experimental conditions, depending on the ratio of the parameters of the excitonic transition.

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