Optical Orientation of Free and Bound Excitons in Hexagonal Crystals

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It was shown recently [1−3] that interband transitions in semiconductors, produced by absorption of circularly polarized light, can cause optical orientation of the magnetic moments of the carriers relative to the direction of propagation of the exciting light. The application of the method of optical orientation to semiconductor optics is very useful and makes it possible in particular to determine such parameters as the lifetime and the spin-relaxation time of the free electrons [4], to investigate the features of spin relaxation of "hot" electrons [5−7], etc. So far, however, investigations of the optical orientation were carried out only on crystals with cubic symmetry. In these crystals, owing to the degeneracy of the valence band at the point $k = 0$, there occurs a strong spin relaxation of the holes [6] and it was therefore possible to observe only the orientation of the electrons. We report in this paper observation of optical orientation of electrons and holes bound into excitons in hexagonal crystals of cadmium selenide. Unlike in cubic crystals, we observed anisotropy of the optical orientation and the absence of depolarization in a transverse magnetic field.

Compared with cubic crystals, the anisotropic crystal field in hexagonal crystals leads to lifting of the degeneracy at the point $k = 0$ and to splitting of the upper valence band Γ_8 into two subbands Γ_9 and Γ_7 [8] (see Fig. 1). An examination of the selection rules shows that the optical orientation of the magnetic moments of the carriers in transitions from the upper valence subband Γ_9 to the conduction band can be realized only by excitation with circularly polarized light along the hexagonal axis C of the crystal. The degree of orientation of the carriers is $P = |(n_+ - n_-)/(n_+ + n_-)|$ (where n_+ and n_- are the numbers of the carriers with magnetic moments directed parallel and antiparallel to the propagation direction of the light) in such transitions will amount to $P = 1$. When excited light propagates perpendicular to the crystal axis, the transitions $\Gamma_9 - \Gamma_7$ are allowed only for linear polarization of the light with $E \perp C$, and absorption of the light will not lead to orientation of the moments of the carriers.

Transitions from both valence subbands Γ_9 and Γ_7 to the conduction band will be realized when light is absorbed with a quantum energy $E_g^B < h\nu < E_g^C$. In the case when $\Delta_{cr} \ll \Delta_{so}$ (where Δ_{cr} and Δ_{so} are the crystal and spin−orbit splitting of the valence band) and the excitation is from the interior of the valence subbands Γ_9 and Γ_7, the degree of orientation of the carriers produced by the circularly polarized light propagating along the crystal axis will be 0.5, just as for cubic crystals. At low temperatures, however, owing to the interband thermalization of the holes, the luminescence will be due to transitions from the conduction band to the upper valence subband Γ_9. The circular polarization of the luminescence will take place only for observation along the crystal axis. Then the degree of polarization $S = |(I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})|$ (where I_{σ^+} and I_{σ^-} are the intensities of the right- and left-circularly polarized light) will be equal to the degree of orientation of the carriers, $S = P$ (unlike in cubic crystals,
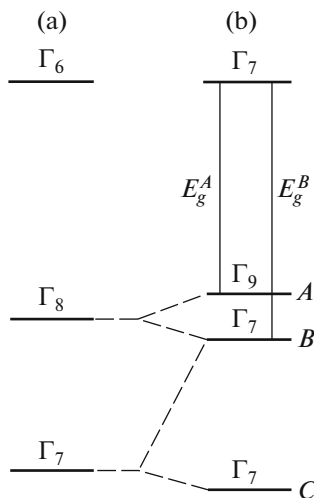

Fig. 1. Splitting of the valence band in (a) cubic and (b) hexagonal crystals at $k = 0$.
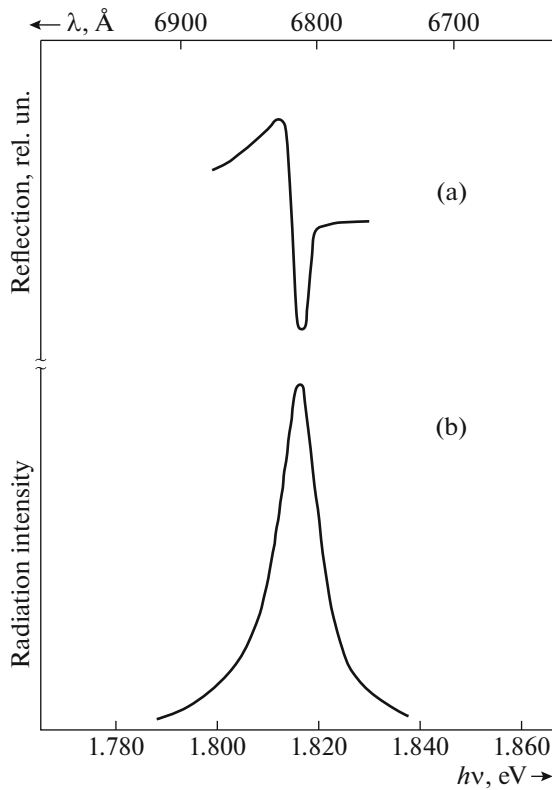
Fig. 2. (a) Reflection spectrum and (b) luminescence spectrum of the crystals investigated in the present work at $T = 77$ K.

where $S = 0.5P$). The presence of orientation of the magnetic moments of the free carriers under certain conditions can lead to orientation of the magnetic moment of the exciton.

Investigations of the optical orientation of the excitons in crystals of cadmium selenide were carried out at $T = 4.2$ and 77 K. The luminescence was excited by circularly polarized light from an He−Ne laser with energy $h\nu = 1.96$ eV. At the same time, carriers were excited from the interior of both valence subbands Γ_9 and Γ_7 ($E_g^A = 1.841$ eV and $E_g^B = 1.868$ eV in CdSe crystals at $T = 4.2$ K).

At $T = 77$ K the luminescence of the CdSe crystals is due principally to annihilation of the free excitons connected with the upper valence subband Γ_9. The reflection spectra and the luminescence spectra of the crystals investigated by us are shown in Fig. 2, respectively. Upon excitation with circularly polarized light along the crystal axis, we observed circular polarization of the exciton luminescence. The degree of polarization was $S = 0.09 \pm 0.02$. As expected, no circular polarization of the luminescence was observed for excitation perpendicular to the crystal axis.

An external magnetic field perpendicular to the direction of propagation of the exciting light leads in cubic crystals to depolarization of the luminescence,

owing to the spin precession (the analog of the Hanle effect) [2, 3]. The magnetic field needed for the depolarization is determined by the lifetime of the spin and by the g-factor of the electron, and is of the order of 1−1000 G for cubic crystals [2, 3]. In our experiments with hexagonal crystals, however, a magnetic field of intensity up to 10 kG did not lead to any noticeable luminescence depolarization. This is apparently due to the fact that in this case it is the excitons that are oriented, and not the free electrons. Indeed, the magnetic moments of the electron and of the hole in the exciton are coupled in direction,[1] and what is oriented is the total magnetic moment of the exciton. In hexagonal crystals, however, as in deformed cubic crystals [9], the g-factor of the hole, and consequently also of the exciton (in the presence of strong exchange interaction) has a strong anisotropy and is equal to zero in the direction perpendicular to the crystal axis. Since in our experiments the excitation and observation of the luminescence were along the crystal axis, and the magnetic field was directed perpendicular to the axis, it did not lead to precession of the magnetic moment of the exciton, since $g_\perp^e = 0$. This explains the absence of the Hanle effect on excitons in hexagonal crystals.

Let us dwell briefly on the results obtained for bound excitons. At $T = 4.2$ K there dominates in the luminescence the line due to annihilation of excitons bound on a neutral donor (J_2 line). We also observed circular polarization of this line upon excitation with circularly polarized light. The degree of polarization was $S = 0.14 \pm 0.02$. Since the magnetic moment for an exciton bound on a neutral donor is due to the uncompensated magnetic moment of the hole, optical orientation of the holes is realized in this case. This is confirmed by the absence of the Hanle effect on this line. Thus, it turned out to be possible to observe optical orientation of the holes in hexagonal crystals. No hole orientation is observed in cubic crystals, owing to the rapid spin relaxation due to the degeneracy of the valence band at the point $k = 0$ [6]. However, the lifting of the degeneracy of the valence band upon uniaxial deformation of the cubic crystals leads to a considerable slowing down of the spin relaxation of the holes [10]. In hexagonal crystals, on the other hand, the degeneracy of the valence band is lifted by the anisotropic crystal field. Thus, observation of optical orientation of holes in hexagonal crystals became possible apparently because of the slow spin relaxation of the holes, resulting from the absence of degeneracy of the valence band at the point $k = 0$.

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[1] Only *para*-excitons are allowed in optical transitions.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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