Gigantic magnetic splitting of exciton reflection band in a ZnTe:Mn crystal

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Gigantic (~9 meV in fields 10–20 kOe) splittings of the exciton reflection spectrum were observed in a ZnTe crystal containing $(6\pm1.6)\times10^{19}$ cm^{-3} of Mn^{2+} ions. The effects are attributed to exchange interaction between the carriers that enter in the free exciton and the magnetic impurity ions.

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Qualitatively new magneto-optical effects connected with excitons in CdTe doped with Mn^{2+} were reported in[1]. The observed effects were attributed in[1] to exchange interaction between the carriers contained in the free exciton and the magnetic impurity ions. Although the model of[1] should be common to semiconducting crystals doped with magnetic ions, no observation of similar effects in other crystals have been reported so far.

The effects observed in[1] were revealed mainly by circular dichroism of the reflection spectrum, while the spectroscopically split components of the spectrum remained almost unresolved. In the present paper we report observation of a giant Zeeman splitting of the 1*s* exciton reflection line in ZnTe doped with Mn^{2+}, and the lines are spectroscopically resolved starting with fields of only 3–5 kOe. The obtained splittings are explained within the framework of the scheme developed in[1].

The investigations were carried out on single crystals with Mn^{2+} concentration $C=(6\pm1.6)\times10^{19}$ cm^{-3} at almost normal incidence of the light on the crystal in magnetic fields parallel and perpendicular to the light-incidence direction ($\mathbf{H}\|\mathbf{k}$ and $\mathbf{H}\perp\mathbf{k}$) and at temperatures 1.94 and 4.2 K, using a DFS-12 spectrometer with spectral slit width 0.15 Å.

At $\mathbf{H}\|\mathbf{k}$ the reflection spectra were observed in combined (σ) polarization as well as in σ^+ and σ^- polarizations separately, and at $\mathbf{H}\perp\mathbf{k}$ they were observed in σ and π polarizations. In the ZnTe crystal without Mn, the exciton-reflection spectrum re-

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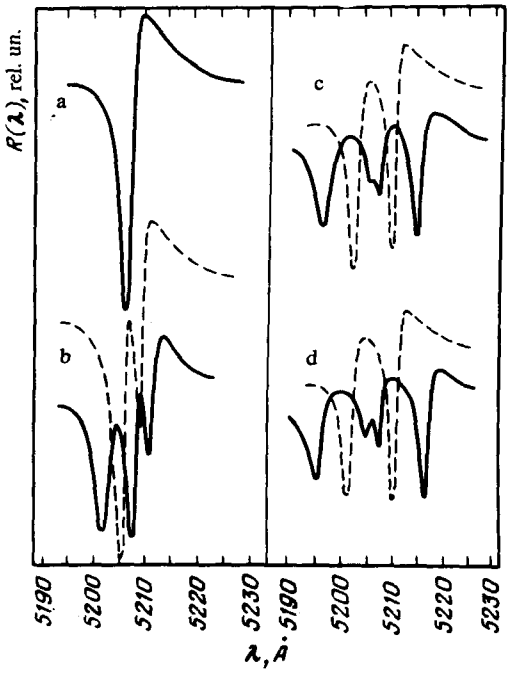

FIG. 1. Reflection spectra of 1*s* exciton in a magnetic field perpendicular to the direction of propagation of the light for different values of H: a—$H=0$, b—$H=5.72$ kOe, c—$H=17.15$ kOe; d—$H=28.55$ kOe. Solid curves—σ polarization, dashed—π polarization.

mained unchanged (within the limits of experimental accuracy) up to $H \approx 30$ kOe. The reflection spectra of the doped crystal for certain values of H are shown in Fig. 1(a–d). Figure 2 shows the positions of the minima of the σ and π reflection spectra as functions of H for the experiment with $\mathbf{H} \perp \mathbf{k}$. The signs of the σ^+ and σ^- polarizations are in correspondence with the measurements at $\mathbf{H} \| \mathbf{k}$ (the signs of the circular polarizations were obtained for one of the directions of the field $\mathbf{H}$; they are reversed when the sign of $\mathbf{H}$ is reversed).

It is seen from Fig. 1 that the 1*s* exciton reflection line splits into six differently polarized components almost symmetrically about the position at $H=0$. The splitting greatly exceeds $2\beta H$ and in fields 10–20 kOe it corresponds to an effective action of a field of several hundred kOe, becoming saturated with further increase of the field.

In the spherical-symmetry approximation, the Hamiltonian of the exciton in a magnetic field, for a doped crystal, is of the form

$$\mathcal{H} = \mathcal{H}_{\text{exc}}^{\text{orb}}(\mathbf{K}, \mathbf{H}) + \mathbf{G}_e \mathbf{S}_e + \mathbf{G}_h \mathbf{S}_h^{\text{eff}} + J_{eh} \mathbf{S}_e \mathbf{S}_h^{\text{eff}} \qquad (1)$$

where $\mathcal{H}_{\text{exc}}^{\text{orb}}(\mathbf{K},\mathbf{H})$ is the Hamiltonian of the orbital motion of the exciton and its constituent carriers in a field $\mathbf{H}$; $\mathbf{K}$ is the wave vector of the exciton; $\mathbf{G}_{e(h)} = g_{e(h)} \beta \mathbf{H} + \Sigma_i J_{e(h)\text{Mn}_i} \mathbf{S}_{\text{Mn}_i} = g_{e(h)} \beta \mathbf{H} + I_{e(h)} \langle \mathbf{S}_{\text{Mn}} \rangle$ (i numbers the impurity ions of Mn^{2+}); $J_{e,h}$ is the electron-hole exchange constant; $\mathbf{S}_e$ is the electron spin; $\mathbf{S}_h^{\text{eff}}$ is the effective spin of the hole ($S_h^{\text{eff}} = 3/2$).

According to[2,3], the diamagnetic shifts determined by $\mathcal{H}_{\text{exc}}^{\text{orb}}(K,H)$ are negligi-

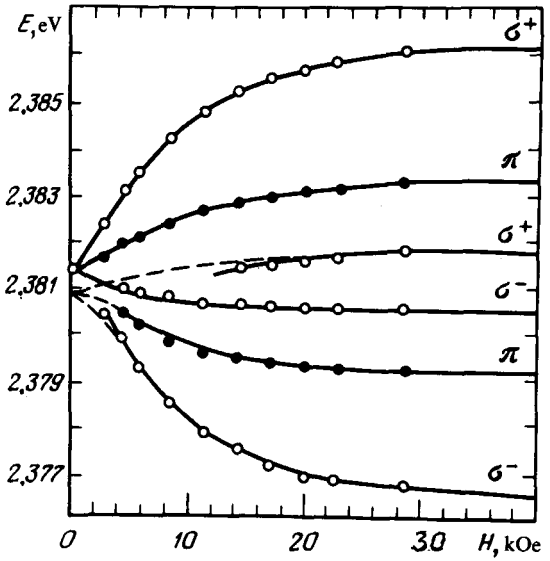

FIG. 2. Comparison of the experimental and calculated positions of the minima of the reflection spectra for H⊥k as functions of the field. The experimental values for the σ and π polarized light are designated by the points (○) and (+), respectively. The solid lines show the results of the calculation for the σ and π spectra. The dashed line shows the calculated positions of the minima of the individual components of the spectrum in the field region where these components have low intensity.

bly small for II-VI crystals in fields $\lesssim 40$ kOw. Then the energies of the eight possible excitonic states in the field **H** will be equal to $E_j(\mathbf{K}) = E_0(\mathbf{K}) + E_j^{sp}$. The values of E_j^{sp} were calculated in[1] with allowance for the fact that $\langle S_{x\,\mathrm{Mn}}\rangle = \langle S_{y\,\mathrm{Mn}}\rangle = 0$, and $\langle S_{z\,\mathrm{Mn}}\rangle = -\frac{5}{2}B_S = \frac{5}{2}(g_{\mathrm{Mn}}\beta HS/kT)$, where $B_S(X)$ is the Brillouin function. The polarizations in which the spin-split excitonic states E_j and the probabilities of the optical transitions will be active were also considered in[1]. The schematic arrangement of $E_j(K=0)$ as a function of $\eta = G_e/G_h$ is shown schematically for $|G_h(H\to\infty)| > |J_{eh}|$ in Fig. 3. The experimental results of[1] correspond to $\eta = -1$, although the relatively large widths of the spin-split components of the spec-

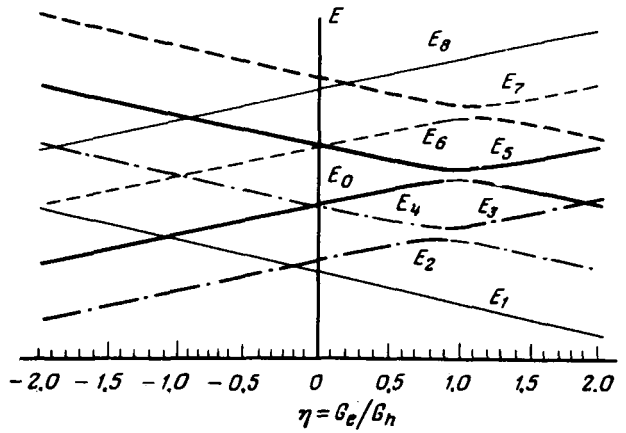

FIG. 3. Position of $E_j(K=0)$ as a function of η for $J_{eh}<0, G_h>0$, dotted lines—dipole-forbidden states; dashed—allowed in σ^+; dashed-dot—in σ^- polarization; solid—in π polarization. The line thicknesses reflect schematically the transition probabilities.

trum in CdTe : Mn do not make it possible to determine in that references this quantity with high accuracy. As seen from a comparison of Figs. 1(a–d) and Fig. 2 with the scheme of Fig. 3 for ZnTe : Mn, we have $-1<\eta<0$.

Since the lines of Fig. 1 are partially superimposed, for a more accurate determination of the parameter from the experimental data we have fed into the computer the contour $F(E)$ of the exciton reflection line at $H=0$ and then calculated the contours $\Phi^{\sigma}(E)_{H,T}=(1/N^{\sigma})\Sigma_j M_j^{\sigma} F\{E-[E_j^{sp}(H,T)-E_{H=0}]\}$ and $\Phi^{\pi}(E)_{H,T}=(1/N^{\pi})\Sigma_j M_j^{\pi} F\{E-[E_j^{sp}(H,T)-E_{H=0}]\}$, where N^{σ} and N^{π} are normalizations and $E_{H=0}$ is the calculated position of the energy of the allowed transition at $H=0$. Using the same numbering sequence as in[1] for the spin-split exciton states (it is shown in Fig. 3), we have

$$M^{\sigma\pm}_{1,4,5,8}=M^{\sigma+}_{2,3}=M^{\sigma-}_{6,7}=M^{\pi}_{1,2,3,6,7,8}=0;\quad M^{\pi}_{4,5}=\frac{2}{3}Ae_z^2|\pm c_{4,5}+c_{5,4}|^2,$$

$$M^{\sigma-}_{2,3}=\frac{1}{2}A(1-e_z^2)|\pm\frac{1}{\sqrt{3}}c_{2,3}+c_{3,2}|^2;\quad M^{\sigma+}_{6,7}=M^{\sigma-}_{2,3}(c_2\to c_7;\ c_3\to c_6),\quad (2)$$

where A is the constant for the given substance, e_z is the projection of the light-polarization unit vector on the direction of **H**,

$$c_{2,3}=p_{2,3}\left(\frac{1}{2}\pm\frac{\Delta_1}{2\sqrt{\Delta_1^2+3J^2}}\right)^{1/2};$$

$$c_{4,5}=p_{4,5}\left(\frac{1}{2}\pm\frac{\Delta}{2\sqrt{\Delta^2+4J^2}}\right)^{1/2};\quad c_{6,7}=c_{2,3}(\Delta_1\to\Delta_2);\quad J\equiv J_{e,h});$$

$$\Delta=G_e-G_h;\ \Delta_{1,2}=\Delta+J;\ G_{e(h)}\equiv G_{ze(h)};\ p_{2,4}=J/|J|;\quad p_{3,5}=1.$$

The calculations were performed for different values of η and $J_{e,h}$ at values of $|I_e|$ such that ensured for each η agreement between the calculated and measured positions of the minima of the reflection in the maximum field. The positions of the minima of the reflection spectra calculated in this manner for $\eta=-0.5$; $J_{eh}=-0.23$ meV; $|I_e|=0.566$ meV ($|G_e(H\to\infty)|=1.412$ meV are shown in Fig. 2 by solid lines. In the calculation of $E_j^{sp}(H,T)$ we neglected the direct action of the magnetic field on the spins of the electron and holes (the terms $g_{e(h)}\beta H$ in the expressions for $G_{e(h)}$). The calculated spectra are less sensitive to J_{eh} than to η, and agree quite well with the experimental ones. With the measurement accuracy taken into account, it is necessary to assume for the investigated crystal $\eta=-0.5\pm0.02$; $J_{eh}=-0.23\pm0.05$ meV; $|I_e|=0.566\pm0.008$ meV. The values of $J_{e,h}$ practically agree with the data of[4]. The measurement results for $T=4.2$ K also agree with the calculation at the given values of the parameters, if account is taken of the fact that $G_{e(h)}\sim B_{S=5/2}(g\beta HS)/kT$.

Thus, the measurements confirm the general character of the effects of the ex-

change interaction of the carriers with the impurity magnetic ions and make it possible to compare in greater detail the theory of these effects with experiment.

In CdTe crystals, doping with Fe^{2+} or Mn^{2+} ions leads, besides the effects investigated in[1], to shifts of the exciton energy in the absence of a magnetic field.[5] These shifts do not follow from the model of[1]. The absence of such shifts in ZnTe : Mn in the presence of a rather large exchange interaction between the carriers and the magnetic impurity confirms that the shifts observed in[5] are not of exchange origin.

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