Magnetic-field splitting of the $n = 2$ exciton state in ZnTe:Mn

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Giant spin splitting of the $n = 2$ exciton was observed in ZnTe doped with Mn^{2+}. The results confirm the exciton splitting model in the fields of the exchange interaction of the electron and hole with the magnetic impurities. The effective exchange fields for the $n = 2$ exciton exceed somewhat the analogous values for the $1s$ exciton.

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Giant magnetic-field splitting of the spin states of $1s$ excitons in magnetically doped crystals has been interpreted in[1,2] as the result of exchange interaction between the carriers bound into an exciton and the impurity magnetic ions polarized along the spin by an external magnetic field H. The splittings obtained in[1,2] are comparable with the exciton binding energy. Since the effective exchange fields act only on the spins of the electron and hole and do not enter in the vector potential that determines the carrier motion in the magnetic field, this circumstance should not limit the applicability of the model.[1,2] A check will be provided by an investigation of a situation wherein the splitting of the exciton in the exchange fields of the magnetic impurities exceeds the binding energy of the excitonic state, a situation that can be realized for states with $n > 1$. It is important also to investigate the influence of the character of the binding of the electron and hole into an exciton on their exchange interaction with the magnetic impurities. No magnetic splittings of excitonic states with $n > 1$ in magnetically doped crystals have been observed so far.

We report in this paper a giant spin splitting of an $n = 2$ exciton in ZnTe containing 6×10^{19} cm^{-3} Mn^{2+} ions. The splitting of the $1s$ excitonic state in the same crystal is described in[2]. At $H = 0$ the state of the $n = 2$ exciton manifests itself in the reflection spectrum at $T = 1.94$ K in the form of a peak that is 30 times weaker than the $1s$ state. The reflection spectra in the region of the $n = 2$ state are shown in Fig. 1 for different values of H in σ^+, σ^- and π polarizations. Figure 2 shows the positions of the minima of the $n = 2$ reflection components as a function of H. It follows from[3,4] that the split-

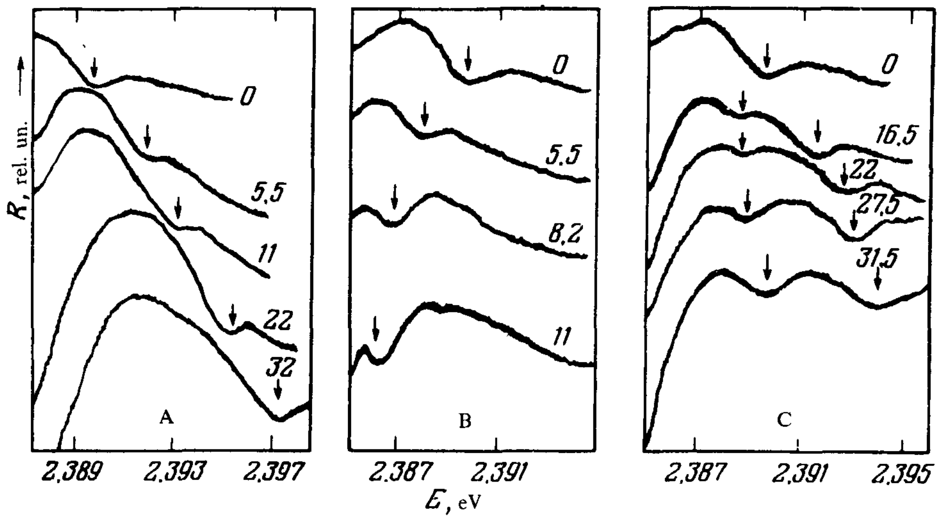

FIG. 1. Reflection spectra of ZnTe:Mn in the region of excitation of an exciton with $n=2$ in a magnetic field at 1.94 K. A,B,C—spectra in σ^+, σ^- (Faraday geometry) and π (Voigt geometry) polarizations. The numbers on the curves are the values of the magnetic field in kilooersteds.

tings of the reflection minima with the splitting of the excitonic states can lead to an incorrect estimate of the electron-hole exchange interaction in the exciton whenever the longitudinal—transverse splitting is larger than the damping constant Γ of the excitonic state.1) This circumstance is of no importance for the investigated $n=2$ state, in view of the small oscillator strength of this transition.

It is seen from Fig. 2 that the character of the spin splitting of the $n=2$ state is similar to the splitting of the 1*s* state, with account taken of the fact that the weak "internal" σ^+ and σ^- lines of the spin multiplet, observed for the 1*s* exciton, cannot be observed with sufficient reliability for $n=2$. On the other hand, the behavior of strong "external" σ^+ and σ^- and π lines in a magnetic field differs from the corresponding lines of the 1*s* exciton by an additional general shift towards shorter wavelengths. The change of the energy of the short-wave components of the spin excitonic multiplet with changing H exceeds the binding energy for the $n=2$ excitonic state, namely $E_b/4$, where E_b is the exciton binding energy in the lowest 1*s* state. For ZnTe we have $E_b \cong 10.6$ meV.[3]

The results can be explained within the framework of the model of[1,2]. According to this model, neglecting anisotropy, we have

$$E_j(\mathbf{K},H)_{nl}=E_0(\mathbf{K},H)_{nl}+E_j^{\mathrm{sp}}(H), \tag{1}$$

where $E_j(\mathbf{K},H)_{nl}$ is the energy of the jth component of the spin multiplet of the nl state of the exciton and depends on the wave vector $\mathbf{K}$, $E_0(\mathbf{K},H)_{nl}$ is the energy of the given excitonic state without allowance for the spin part of the Hamiltonian,[2] and E_j^{sp} is the

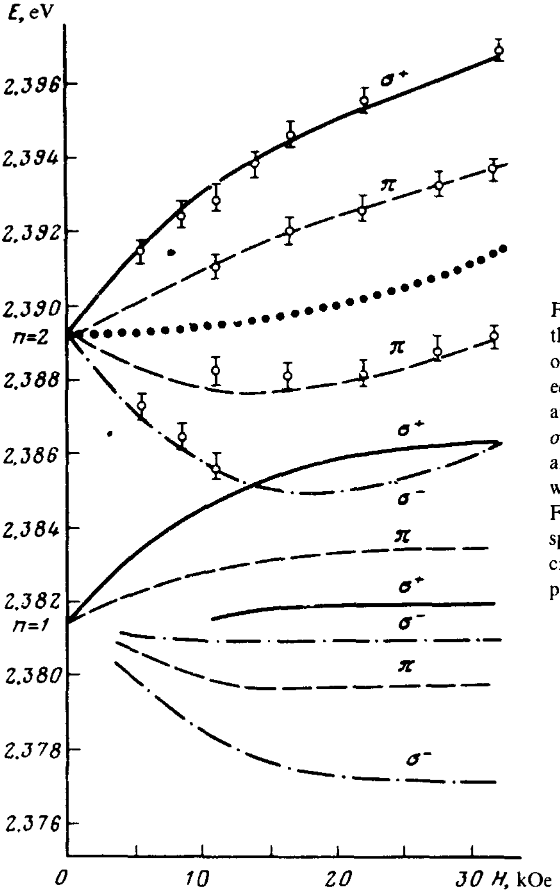

FIG. 2. Points dependence of the positions of the reflection minima of the spin components of the $n = 2$ exciton. Dotted curves—calculated position of $E_0(K = 0,H)_{2s}$; solid, dash-dot, and dashed curves—calculated positions of the σ^+, σ^-, and π components of 2*s* for $\pi_{2s} = -0.5$ and $|I_e|_{2s} = 0.651$ meV (the positions of the weak σ^+ and σ^- components are not shown). For comparison, the results are shown[2] of the spin splitting of the 1*s* exciton in the same crystal by the same lines for the corresponding polarizations.

contribution made to the exciton energy by the spin part of the exciton Hamiltonian. The dependence of $E_j^{\rm sp}$ on H was considered in[1,2] together with the selection rules and the probabilities of the transitions to different spin states.2) The splittings of the possible spin states are determined by the effective fields of the exchange interaction of the electron and hole ($\mathbf{G}_e$ and $\mathbf{G}_h$) with the impurity magnetic ions and by the electron-hole exchange, which is small enough in this case to be negelected. In this case we have[2]

$$\mathbf{G}_{e(h)} = g_{e(h)}\beta\mathbf{H} + \sum_i J_{e(h)\,\mathrm{Mn}\,i}\,\mathbf{S}_{\mathrm{Mn}\,i} = g_{e(h)}\mathrm{B}\mathbf{H} + I_{e(h)} < \mathbf{S}_{\mathrm{Mn}} >, \qquad (2)$$

where $g_{e(h)}$ is the electron (hole) g factor, $J_{e(h),\mathrm{Mn}i}$ is the constant of the exchange interaction of the electron (hole) with the *i*th Mn^{2+} impurity ion. The contribution $g_{e(h)}\beta\mathbf{H}$ can in this case be neglected. The values obtained for the 1*s* exciton[2] are $|I_e|_{1s} = 0.566 \pm 0.008$ meV; $\eta_{1s} = G_e/G_h = -0.5 \pm 0.02$. We have neglected in[2] the dependence of $E_0(\mathbf{K})_{1s}$ on H at $H < 40$ kOe because of the small diamagnetic shift of the 1*s* exciton. For $n = 2$, the diamagnetic shift is much larger (14 times larger for the 2*s* exciton than for the 1*s* exciton), and it must now be taken into account. In the hydrogenlike model, assuming that the observed transitions are due to the $2s$ exciton state, we obtain

$$E_0(K=0,\, H)_{2s} = E_0(K=0,\, H=0)_{2S} + \frac{e^2H^2}{12\mu c^2}\overline{r^2_{2s}} = E_0(K=0,\, H=0)_{2s} + H^2\frac{7}{2}\,\frac{\epsilon^2}{\mu^3}\,\frac{\hbar^4}{m^3c^2e^2}\,. \tag{3}$$

Here μ is the reduced effective mass of the electron and hole in units of the electron mass m, and ϵ is the dielectric constant. At $H \leqslant 30$ kOe this approximation should be acceptable for the $2s$ exciton in ZnTe.

Taking into account the strong dependence of (3) on ϵ and μ, we used for the coefficient of H^2 in the expression for the the diamagnetic shift of the $2s$ exciton a value 14 times larger than the coefficient obtained from the experimental diamagnetic shift of the $1s$ exciton.[3] Its value corresponds to $\epsilon^2/\mu^3 = 128 \times 10^3$, which is in acceptable agreement with ϵ and μ as known from the literature. The $E_0(K=0,\, H)_{2s}$ dependence obtained in this manner is shown in Fig. 2.

Using (1) and the expressions for E_j^{sp} from[1], we have plotted the expected H-dependences of the energies of the spin components of the $2s$ excitonic state. It turned out that the calculated values of $E_j(K=0,\, H)_{2s}$, obtained by using the values of η_{1s} and $|I_e|_{1s}$, are close to the experimental points, but the experimental values of the spacings between the individual components of the split $n = 2$ state are somewhat larger. Better agreement with experiment was obtained at $\eta_{2s} = \eta_{1s} = -0.5$, but with $|I_e|_{2s}$ approximately 15% larger than $|I_e|_{1s}$, i.e., with $|I_e|_{2s} = 0.651 \pm 0.05$ meV.

The difference between $|I_e|_{1s}$ and $|I_e|_{2s}$ may be a reflection of the approximate character of the model, or else a consequence of the fact that the exchange interaction of the electron (hole) with the magnetic impurities changes somewhat, depending on whether the carrier is free or bound into an exciton. The cause of the change may be the screening of the exchange interaction of each of the carriers with the magnetic impurities by the second carrier present in the exciton. This screening should decrease with increasing n, and this can explain the observed differences.

In all other respects the experimental results confirm the applicability of the model of[1,2] to excitons in magnetically doped semiconductors.

[1)] This circumstance was not taken into account in[2], where the electron-hole exchange constant $J_{e,h}$ is patently overestimated.

[2)] The expressions for the transition probabilities to separate spin systems in[1] are incorrect, but the correct expressions are given in[2].

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