

$$\varepsilon = \frac{1}{10} \left(1 - \frac{a^2}{b^2} \right), \quad f(\theta, \varphi) = 5 \left(\cos^2 \theta - \frac{1}{3} \right)$$

(the angle θ is measured from the direction of the axis of the spheroid). Then calculating the tensor f_{ik} with Eq. (5.4) we find

$$f_{ik} = k_i k_k - \frac{1}{3} \delta_{ik}, \quad (5.10)$$

where k_i is a unit vector in the direction of the axis of the spheroid. Now by substituting (5.10) into (5.9) (and assuming for simplicity that the axes of the two spheroids are parallel) we can easily see that the viscosity coefficient is proportional to $(\mathbf{k} \cdot \mathbf{e})^2 ((\mathbf{k} \times \mathbf{e}) \times \mathbf{e})^2$ and may vanish if the spheroid is magnetized either parallel or perpendicular to the axis.

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Magneto-optical investigations of the exciton band in CdTe:Mn²⁺

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A strong enhancement of magneto-optical effects (Faraday rotation of the plane of polarization of light, magnetic circular dichroism of the reflection) was observed when CdTe crystals were doped with Mn²⁺ ions. It was established that the effect reduces to splitting, of the band of the 1s exciton into five components in an external magnetic field. The splitting is proportional to the concentration and to the spin polarization of the system of the Mn²⁺ impurity ions. At the employed manganese concentrations ($\sim 8 \times 10^{18} \text{ cm}^{-3}$) the effective field in which the splitting of the exciton band takes place reaches several hundred kilooersteds in an external field of 30 kOe. The change of the spin polarization of the impurity system upon saturation of the microwave EPR transitions of the Mn²⁺ ions decreases the splitting. The magneto-optical effects observed in the exciton band of the CdTe:Mn²⁺ crystals, their dependence on the field H and on the microwave pump, and the polarization of the split components are all attributed to exchange interaction of the localized magnetic Mn²⁺ ions with the hole and electron contained in the free exciton.

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INTRODUCTION

Investigations of the exciton spectra of semiconducting CdTe crystals doped with certain ions (Fe, Mn) of the iron group^[1] have shown that when these ions are introduced into the crystal a short-wave shift of the exciton line takes place. The magnitude of the shift depends strongly on the impurity concentration, but the exciton line itself does not disappear. Investigations of the in-

fluence of the impurity on the character of the Zeeman splitting of the exciton band can yield important information on the singularities of the excitons in crystals doped in this manner. Taking into account the difficulty of observing the Zeeman effect of a sufficiently broad exciton band in the CdTe:Mn²⁺ crystals, we have decided to measure the Faraday rotation (FR) and the magnetic circular dichroism (MCD) of samples with different degrees of doping by Mn²⁺ ions.

EXPERIMENTS

The measurements were made at a temperature $T = 1.7$ K on CdTe cubic crystals doped with Mn^{2+} ions at concentrations from zero to $\sim 8 \times 10^{18} \text{ cm}^{-3}$. The samples were placed in a superconducting solenoid in such a way that the wave vector \mathbf{K} of the light wave was directed along the field and coincided with the $\langle 110 \rangle$ axis of the crystal, which was normal to the cleavage plane: $\mathbf{K} \parallel \mathbf{H}_0 \parallel \langle 110 \rangle$. In the optical spectral measurements we used a grating monochromator with a spectral slit 1.5 \AA wide. We registered the dispersion dependence of the following effects: a) the Faraday rotation of the plane of polarization of the light passing through thin crystal-line samples, b) the magnetic circular dichroism of the light reflected⁴⁾ from the cleavage plane (110). To measure the FR and the magnetic circular dichroism of the reflection (MCDR) we used sensitive modulation procedures, similar to those described in^[2]. The installation had also a microwave section intended for powerful ($P = 1$ W) microwave (10 GHz) pumping of samples with $\mathbf{H}_{\text{micro}} \perp \mathbf{H}_0$. This has made it possible, simultaneously with the optical measurements in the exciton line, to excite (and saturate) EPR transitions in the investigated samples, the EPR being detected by its optical effects (optico-magnetic double resonance—OMDR).

Finally, we observed spectroscopically the splitting of the exciton line in magnetic fields stronger than 20 kOe. The polarization measurements were made in light reflected from the cleavage plane (110) and propagating across the magnetic field: $\mathbf{K} \perp \mathbf{H}_0$. The electric field vector \mathbf{E} of the light wave was directed along ($\mathbf{E} \parallel \mathbf{H}_0$, π polarization) or across ($\mathbf{E} \perp \mathbf{H}_0$, σ polarization) the external field \mathbf{H}_0 . One of the axes ($\langle 111 \rangle$, $\langle 110 \rangle$, or $\langle 100 \rangle$) of the crystal was parallel to \mathbf{H}_0 .

MEASUREMENT RESULTS

The dispersion dependence of the FR effect, shown in Fig. 1a, was measured with a thin sample ($d = 0.2$ mm) of CdTe: Mn^{2+} ($C \sim 6 \cdot 10^{18} \text{ cm}^{-3}$) in a magnetic field $H_0 = 3.65$ kOe. Similar $\Phi(E)$ curves were obtained with other crystals containing Mn^{2+} , with a sharp increase of the FR angle with increasing energy of the light quantum $E = h\nu$. The region of the steep variation of $\Phi(E)$ coincided with the long-wave slope of the exciton band, characterized by a sharp increase of the absorption as the maximum was approached (the absorption coefficient at the maximum was $\approx 10^5 \text{ cm}^{-1}$). We could register the $\Phi(E)$ curves only up to an energy 1.569 eV, which was $\sim 32\text{--}38$ meV lower than the maximum, owing to the sharp increase of the optical density of the sample as $E_{\text{max}} = E_0$ was approached.

With increasing concentration C of the Mn^{2+} impurity, we observed a strong increase of the FR effect. For comparison, we show the FR angle measured for three samples differently doped with Mn^{2+} ions: $V_1 = 2.6 \times 10^{-3}$ deg/cm-Oe ($C_1 = 0$), $V_2 = 0.8$ deg/cm-Oe ($C_2 \approx 6 \times 10^{18} \text{ cm}^{-3}$), and $V_3 = 3.6$ deg/cm-Oe ($C_3 \approx 7.2 \times 10^{18} \text{ cm}^{-3}$). Here $V = \Phi/H_0 d$, Φ is the FR angle in degrees, d is the sample thickness, and $H_0 = 3.65$ kOe. The values of V_1 , V_2 , and V_3 were obtained at spectral points that were

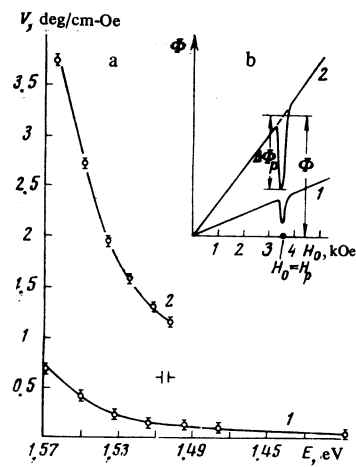


FIG. 1. a) Dispersion curves of FR, obtained on CdTe: Mn^{2+} samples with different degrees of doping: 1— $C \approx 6 \cdot 10^{18} \text{ cm}^{-3}$, 2— $C \approx 7.2 \cdot 10^{18} \text{ cm}^{-3}$. b) OMDR spectra, recorded on different sections of the dispersion curve of FR in CdTe: Mn^{2+} ($C \sim 6 \cdot 10^{18} \text{ cm}^{-3}$): 1— $h\nu_1 = 1500$ eV, 2— $h\nu_2 = 1.568$ eV.

closest to the maxima of the exciton bands of the indicated samples at $T = 1.7$ K.

In the case of microwave pumping of the samples, a strong decrease of the FR was observed in the region of the fields corresponding to EPR of Mn^{2+} in CdTe. Some of the OMDR spectra obtained with one of the CdTe: Mn^{2+} samples from the FR signal on the long-wave slope of the exciton absorption band are shown in Fig. 1b. The change $\Delta\Phi_r$ of the rotation angle reached a maximum value when the EPR transitions of Mn^{2+} were fully saturated. It was established that the relative depth $\Delta\Phi_r/\Phi$ of the resonance dip increases with increasing Mn^{2+} impurity concentration in the CdTe. In strongly doped samples, $\Delta\Phi_r/\Phi \approx 70\%$ and does not depend on the frequency of the incident light within the limits of the experimental error.

The results point directly to a connection between the optical effect of the FR and the degree of polarization of the spins of the Mn^{2+} impurity ions. This is confirmed also by the dependence of the FR angle on the field H_0 , a dependence characterized by saturation in the region of strong fields. The $\Phi(H)/\Phi(H_{\text{max}})$ curves are practically independent of either the wavelength of the light or the Mn^{2+} impurity concentration. On the basis of the presented data we can conclude that the effect of the FR of the exciton band in CdTe: Mn^{2+} is of paramagnetic origin and is determined by the difference between the populations of spin sublevels of the ground state of the Mn^{2+} ion. Since the spin multiplet of the ground state of Mn^{2+} is not the ground state of the exciton, the result seems unexpected.

The studies were continued with magneto-optical investigations in reflected light. This made it possible to extend the spectral range (to 4000–10000 \AA) in which the magneto-optical effects were measured. At a fixed field H_0 and with smooth variation of the frequency $\nu = E/h$ of the incident light we registered a signal S proportional to the intensity difference ($I^{\sigma+} - I^{\sigma-}$) of the reflected light

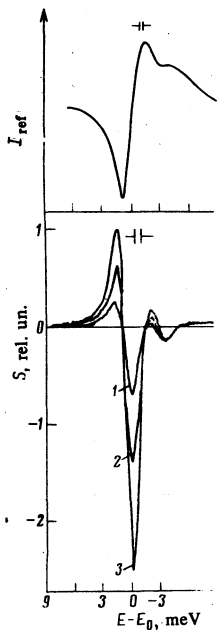


FIG. 2. Reflection in the region of the 1s-exciton band of CdTe: Mn²⁺ (I_{ref}) and MCDR spectra obtained with a crystal having a medium Mn²⁺ concentration ($C \sim 1.2 \times 10^{18} \text{ cm}^{-3}$) in magnetic fields 3.65 kOe (1), 8 kOe (2), and 18 kOe (3).

having circular polarizations σ^+ and σ^- and propagating along the field ($\mathbf{K} \parallel \mathbf{H}_0$). Characteristic plots of $S(E)$ obtained for samples with different Mn²⁺ concentrations are shown in Figs. 2 and 3. In the case of undoped CdTe crystals we were unable to register any MCDR spectra. It can be concluded from the presented curves (Figs. 2 and 3) that, first, the MCDR spectra are connected only with the exciton bands, and no additional bands of intracenter (Mn²⁺) origin were observed; second, the shape of curves 1–3 of Fig. 2 and curves 1 and 2 of Fig. 3a agree well with the curve for the derivative $\partial I_{\text{ref}}(E)/\partial E$ of the contour of the exciton reflection bands observed at $H_0 = 0$. It was also observed that in a strongly doped sample ($C \sim 8 \times 10^{18} \text{ cm}^{-3}$), in magnetic fields close to 20 kOe and higher, the effect reduces to an anomalously large splitting of the exciton band. We note for comparison that

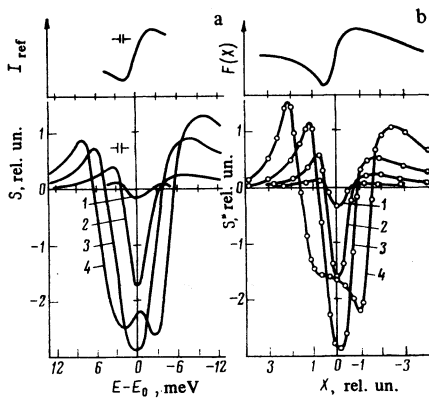


FIG. 3. a) Reflection in the region of the 1s exciton band of CdTe: Mn²⁺ (I_{ref}) and MCDR spectra obtained with a sample having a large concentration ($C \approx 8 \times 10^{18} \text{ cm}^{-3}$) at the following values of the magnetic field: 1—0.6; 2—4.2; 3—10; 4—40 kOe. b) Contour of tabulated reflection lines $F(X)$ and the calculated dependences of the quantity $S^*(X) = F(X + \Delta X/2) - F(X - \Delta X/2)$ at the following values of ΔX : 1—0.05; 2—0.2; 3—0.6; 4—1.5.

splitting of light magnitude ($\Delta E = 9 \text{ meV}$) can be expected for levels with a g -factor equal to two in a magnetic field $H_0 \approx 760 \text{ kOe}$. In the MCDR spectra of Fig. 3a (curve 4) one can see clearly two lines separated in frequency and observed at the two different circular polarizations.

By microwave pumping of the EPR of Mn²⁺ and observing the MCDR (OMDR) signal we obtained results (Fig. 4) analogous to those described above in the FR experiments: 1) the value of the MCDR decreased strongly upon saturation of the EPR of the ground state of the Mn²⁺ ions; 2) the degree of the decrease of the MCDR upon saturation of the EPR of Mn²⁺ increased with increasing impurity concentration; 3) the character of the dispersion relations without microwave pumping ($S(E)$) and in the case of saturated EPR transitions ($[S - \Delta S](E)$) remained the same, with the exception of the fact that in strongly doped samples a small narrowing of the $[S - \Delta S](E)$ spectrum was also observed. Figure 4 shows corresponding plots of the signals $S(E)$ (curve 1) and $[S - \Delta S](E)$ (curve 2).

The onset of MCD ($I^{\sigma^+} - I^{\sigma^-} = S$) in the optical band can be attributed to the following factors.

a) Splitting of the $I(E)$ line into Zeeman components that are separated in frequency by an amount ΔE . The contribution made to the MCD signal by this splitting is frequently called diamagnetic.^[3] Its value is proportional to the ratio $\Delta E/\Gamma$, where Γ is the width of the optical resonance line. S^{dia} is practically independent of temperature.

b) A difference between the intensities of the Zeeman components. In the presence of splitting in the ground level, which takes part in the transition, the intensities of the two circular components turn out to be unequal because of the difference between the populations of the magnetic sublevels. The contribution made to the MCD by this effect increases strongly when the temperature is lowered, and is frequently called paramagnetic.^[3]

In our experiments the character of the MCDR dispersion $S(E)$ points to a diamagnetic nature of the effect. A direct manifestation of this assumption is that $S(E) \sim \partial I_{\text{ref}}(E)/\partial E$, and that the MCDR spectrum splits into

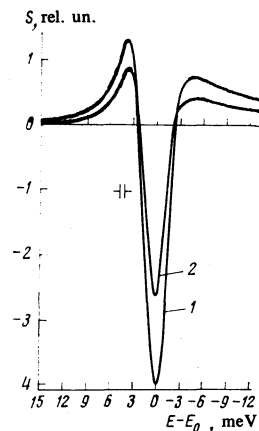


FIG. 4. MCDR spectra obtained with the crystal CdTe: Mn²⁺ ($C \approx 7 \cdot 10^{18} \text{ cm}^{-3}$): 1—without microwave pumping, 2—with microwave pumping of the EPR transitions of Mn²⁺; $H_0 = 3.65 \text{ kOe}$.

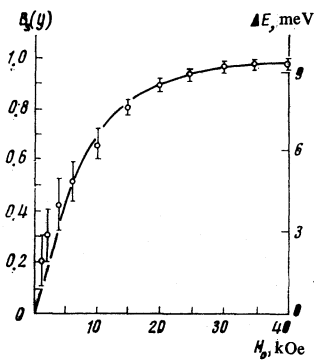


FIG. 5. Points—dependence of the splitting $\Delta E(H_0)$ for the σ^+ and σ^- components of the MCDR spectrum (Fig. 3a) obtained by comparison with calculation (Fig. 3b). The solid line is a plot of the Brillouin dependence $B_s(y)$ of the magnetization on the field H_0 at $s = 5/2$, obtained at $y = g\beta H_0 s / kT$, $g = 2$, $T = 1.7$ K.

two frequency-shifted reflection lines of equal intensity (Figs. 2 and 3a, curve 4).

As to the paramagnetic contribution to the MCDR, we point out that in the region of small splittings the plot of $S^{\text{par}}(E)$ should have the same shape as the reflection line $I_{\text{ref}}(E)$, and that this effect should manifest itself with increasing splitting in a difference between the intensities of the split components. Recognizing that $S^{\text{par}}(E)$ is proportional to the ratio $\Delta E/kT$, we get that at $T = 1.7$ K and at the observed splittings $\Delta E = 9$ meV the ratio of the intensities of the split components should be $I^{\sigma^+}(E)/I^{\sigma^-}(E) \approx 70$, a value not observed in the experiment. No paramagnetic effect could be observed in the MCDR spectra in the region of large (Fig. 3, curve 4), medium (Fig. 3, curves 3 and 2), or small (Fig. 2, curves 1–3) splittings of the exciton reflection band. It can therefore be stated that the ground state, which participates in the investigated optical transition, is not split in the field H_0 , in full agreement with the excitonic nature of the transition.

Thus, the magneto-optical effects (FR, MCDR) in CdTe: Mn^{2+} crystals are due to splitting of the exciton band into σ^+ and σ^- components. To obtain the values of the splittings $\Delta E(H_0)$ from the MCDR spectra (Fig. 3), the reflection line contour $I_{\text{ref}}(E)$ shown in Fig. 3a was calculated and fed in the form of the function $F(X)$ into a computer, where the values of $S^*(X) = F(X + \Delta X/2) - F(X - \Delta X/2)$ were determined for successive series of the shifts ΔX . Some of the calculated $S^*(X)$ curves are shown in Fig. 3b. Comparison of $S^*(X)$ and $S(E)$ made it possible to establish the character of the dependence of the splitting ΔE on the field H_0 , shown in Fig. 5. The obtained $\Delta E(H_0)$ dependence agrees well with the Brillouin curve $B_s(y)$ for the magnetization of a paramagnetic system with spin $s = 5/2$. This feature is apparently due to the presence in the crystal of Mn^{2+} ions with a spin $s_{\text{Mn}} = 5/2$ in the ground state ${}^6S_{5/2}$. In an identical magnetic field H_0 , the values of the splitting ΔE for different crystals are larger the greater the degree of doping with Mn^{2+} ions. Observation of OMDR of Mn^{2+} in MCDR and FR of the exciton line must be interpreted as a decrease of the value of the "Zeeman" splitting when the populations of the spin sublevels of Mn^{2+} become equalized. Thus, the results indicate that the splitting of the exciton band is determined not by the external field as such, but by the magnetization of the spin system of the paramagnetic ions Mn^{2+} in CdTe, which is produced in an external field H_0 .

For a more complete explanation of the structure of the "Zeeman" splitting of the exciton band we investigated a strongly doped sample of CdTe: Mn^{2+} ($C \approx 8.2 \cdot 10^{18} \text{ cm}^{-3}$), in which we were able to register spectroscopically, in magnetic fields stronger than 20 kOe, the splitting of the exciton reflection band in σ and π polarization with the light propagating transverse to the field H_0 . As seen from Fig. 6, five Zeeman components are observed at $H_0 = 30$ kOe, two in π polarization ($E \parallel H_0$), separated by $\Delta E \approx 5 \pm 0.5$ meV, and three in σ polarization ($E \perp H_0$)—two separated by $\Delta E \approx 10 \pm 0.5$ meV and a weak one at the center near E_0 . (The intensities of the lines in π polarization are approximately double those of the outer lines in the σ polarization.) Consequently, the $1s$ excitonic state splits into five equidistant lines observed in different polarizations. Measurements of the dependence of the splitting on the field H_0 yields results that agreed well with those described above and obtained by investigation of FR and MCDR, while the character of the "Zeeman" splitting was independent of the orientation of the crystal axes relative to the magnetic field. Thus, a qualitatively new type of optico-magnetic effect was observed, the splitting of exciton bands in a magnetic field, having no analog for either exciton states or local magnetic centers in crystals. Let us examine its possible causes.

THEORY

Magneto-optical effects in the exciton region of the spectrum are usually interpreted on the basis of Zeeman splitting (with respect to the magnetic quantum number) and the diamagnetic shifts of the exciton lines with allowance for conservation of the angular momentum in the system of the interacting excitons and photons. In cubic straight-band crystals of the II-VI group, the excitons are produced by binding of holes from the upper valence subband Γ_6 , which is split by spin-orbit interaction, with electrons from the conduction band Γ_6 . The lower states of the relative motion of a $1s$ -type electron and a hole in an exciton correspond to the crystal terms Γ_3 , Γ_4 , and Γ_5 . The triplet Γ_5 is optically allowed, while the doublet Γ_3 and triplet Γ_4 can be observed only in strong magnetic fields, and their center of gravity is located at a distance $\delta E_{J_{he}}$ from the level Γ_5 , owing to the electron-hole exchange interaction (according to the data of^[4], $\delta E_{J_{he}}$ amounts to -0.6 meV).

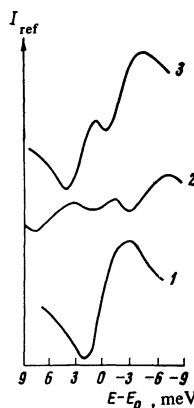


FIG. 6. Spectra of $1s$ exciton reflection band obtained with strongly doped CdTe: Mn^{2+} ($C \approx 8 \cdot 10^{18} \text{ cm}^{-3}$) in measurements across the field ($H_0 \perp K$) at the following magnetic field values: 1— $H_0 = 0$, 2— $H_0 = 30$ kOe, σ polarization ($E \perp H_0$); 3— $H_0 = 30$ kOe, π polarization ($E \parallel H_0$).

The described energy structure of the exciton was confirmed by an investigation of the Zeeman effect of the exciton line in undoped CdTe crystals.^[4] The picture of the splitting of the exciton spectrum observed in^[4] does not correspond at all to the "Zeeman effect" observed by us in the CdTe : Mn²⁺ crystals. Nonetheless, our results can be understood if it is assumed that the doping does not lead to localization of the excitons, and the free exciton (its individual electron and hole) experience exchange interaction with the ions of the 3d-metal impurity. The appearance of exchange interaction at such small concentrations of the magnetic impurity (C ~ 0.1 mol.%) is due to the large radius and motion of the exciton in the crystal.

Indeed, the spin-dependent (we have in mind the effective hole spin $s_h^{eff} = 3/2$, the electron spin $s_e = 1/2$, and the spin $s_{Mn} = 5/2$) part of the interaction energy can be written in the form

$$\hat{\mathcal{H}} = g_h \beta \mathbf{H}_0 s_h^{eff} + g_e \beta \mathbf{H}_0 s_e + g_{Mn} \beta \mathbf{H}_0 \sum_i s_{Mn i} + J_{he} s_h^{eff} s_e + \sum_i J_{hMn} s_h^{eff} s_{Mn i} + \sum_i J_{eMn} s_e s_{Mn i} + \sum_{ij} J_{ij} s_{Mn i} s_{Mn j}. \quad (1)$$

Here J are the exchange coefficients for the different interactions, while i and j number the Mn²⁺ impurity ions. Since the EPR of the Mn²⁺ ions is observed in the investigated CdTe crystals in the same magnetic field as for the isolated ions (low concentrations), the terms containing J_{ij} is of no importance in what follows.

A free exciton interacts simultaneously with a large number of Mn²⁺ ions, so that in the term containing the exchange interaction of the electron and the hole with the Mn²⁺ ions it is possible to change over to the "molecular-field" model. We can then introduce in the indicated terms, in place of summation over the Mn²⁺ ions with their true spin, the average value of the spin of Mn²⁺. The term that describes the Zeeman interaction of the Mn²⁺ ions with the external field leads to

$$\langle s_{Mn i} \rangle_{H_0, T} = s_{Mn} B_{s_{Mn}}^{-1} (g \beta H_0 / kT), \quad (2)$$

i. e., to a Brillouin dependence of the magnetization of the Mn²⁺ ions, while the exchange terms h -Mn and e -Mn determine the magneto-optical effects in the exciton band (the splittings observed by us are much larger than $\delta E_{J_{he}}$). The spin of the hole (electron) is then acted upon by an effective field

$$\frac{5}{2} g_{h(e)} \beta^{-1} \sum_i J_{h(e)Mn} s_{Mn i} \left(\frac{g \beta H_0}{kT} \right),$$

whereas the direct action of the magnetic field H_0 on the hole and on the electron is much less. In this case the Hamiltonian (1) can be approximately rewritten in the form

$$\hat{\mathcal{H}} = G_h s_h^{eff} + G_e s_e + \hat{\mathcal{H}}_{Ze,h} + J_{he} s_h^{eff} s_e, \quad (3)$$

$$G_{h(e)} = n_{h(e)} J_{h(e)Mn} \langle s_{Mn} \rangle_{H_0, T},$$

where $\hat{\mathcal{H}}_{Ze,h}$ includes the first two terms of (1); n_h and n_e are the average numbers of the Mn²⁺ ions with which exchange interaction is effective for the hole and elec-

tron contained in the free exciton. This number increases with increasing concentration of the impurity ions, and can apparently be reasonably approximated by the number of Mn²⁺ ions in a volume $4\pi a_{ex}^3$, where a_{ex} is the effective Bohr radius of the exciton (for the 1s state of the exciton in CdTe their radius is $a_{ex} = 60 \text{ \AA}$). Since $\langle s^x \rangle_{Mn} = \langle s^y \rangle_{Mn} = 0$, it follows that

$$G_{zh(e)} = |G_{h(e)}|, \quad G_{zh(e)} = G_{zh(e)} = 0 \quad (4)$$

by virtue of the definition of G_h and G_e in terms of the mean values of the spin components of the impurity in the external field \mathbf{H}_0 .

The eigenvalues of the Hamiltonian (3) are

$$\begin{aligned} E_{1,s} &= 1/2 \{ 3/2 J \mp [3G_h + G_e + (3g_h + g_e) \beta H_0] \}, \\ E_{2,s} &= 1/2 \{ -2(G_h + g_h \beta H_0) - 1/2 J \mp (\Delta_1^2 + 3J^2)^{1/2} \}, \\ E_{3,s} &= 1/2 \{ -1/2 J \mp (\Delta^2 + 4J^2)^{1/2} \}, \\ E_{4,\tau} &= 1/2 \{ 2(G_h + g_h \beta H_0) - 1/2 J \mp (\Delta_2^2 + 3J^2)^{1/2} \}, \\ \Delta &= (G_e - G_h) + (g_e - g_h) \beta H_0, \quad \Delta_{1,2} = \Delta \mp J, \quad J = J_{he}. \end{aligned} \quad (5)$$

For the values $J = -0.3 \text{ meV}$ (the external integral J is determined from the relation $\delta E_{J_{he}} = 2J$) we have $G_h = 2.2 \text{ meV}$ and, with the Zeeman terms for the electron and hole neglected, these states (as functions of $\eta = G_e/G_h$) are shown in Fig. 7a. It follows from the experimental data (see Figs. 2, 3, and 6) that the widths of the reflection lines in the MCDR and in the Zeeman effect amount to 1–2 meV. At $\eta = 0$ we would therefore observe in the spectra four equidistant lines separated from one another by an amount G_h , while at $\eta = 1$ we would observe five equidistant lines (splittings smaller than the width of the observed lines cannot be observed in experiment). For the spectra shown in Fig. 6, five equidistant lines are observed with different polarizations. It can be assumed that this corresponds to the case $\eta = 1$ at $G_{h(e)} > J$.

We consider next the polarization properties of the optical transitions in the excitonic states $E_1 - E_8$. The operator of the interaction of the electrons and holes of the crystal with the light in the dipole approximation,

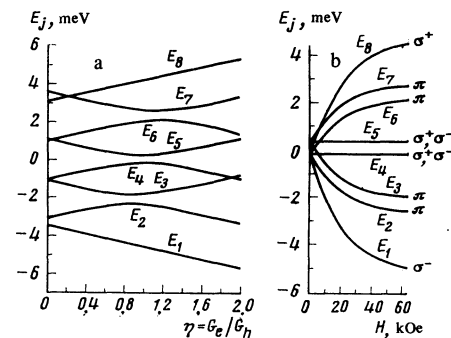


FIG. 7. Dependences of the 1s-exciton energy levels split by exciton-impurity exchange interaction (formulas (5)): a—on the ratio G_e/G_h at $J = -0.3 \text{ meV}$, $G_h = 2.2 \text{ meV}$; b—on the magnetic field at $G_h = G_e = 2.5 B_5/2 (g_{Mn} \beta H_0 / kT)$ [meV], $J = -0.3 \text{ meV}$, $T = 1.7 \text{ K}$. The value G_h for (a) and the factor 2.5 in the Brillouin function for (b) were chosen to obtain agreement with the results of the experiments with the strongly doped crystal (Figs. 3 and 6).

with allowance for the group symmetry T_d of the crystal, is given by

$$\begin{aligned} \hat{\mathcal{H}}_i = & -\frac{e}{m} \sum_{\mathbf{q}} \left(\frac{2\pi\hbar}{\varepsilon_{\mathbf{q}} \omega_{\mathbf{q}}} \right)^{1/2} c_{\mathbf{q}} \sum_{\mathbf{k}} \left\{ \frac{1}{\sqrt{2}} P_{c,v}^{(+)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ \right. \\ & - \frac{2i}{\sqrt{6}} P_{c,v}^{(+)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ + \frac{1}{\sqrt{6}} P_{c,v}^{(-)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ \\ & + \frac{i}{\sqrt{6}} P_{c,v}^{(+)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ + \frac{2}{\sqrt{6}} P_{c,v}^{(+)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ \\ & \left. + \frac{i}{\sqrt{2}} P_{c,v}^{(-)} a_{\mathbf{k}+\mathbf{q},\uparrow} b_{\mathbf{k},\uparrow}^+ \right\} c_{\mathbf{q},j} + \text{h.c.}, \end{aligned} \quad (6)$$

where $a_{\mathbf{k}\sigma}^+$ ($b_{\mathbf{k}\sigma}^+$) is the operator of the production of an electron (hole) with wave vector \mathbf{k} and spin projection $\sigma(\Sigma)$, $c_{\mathbf{q}j}$ is the operator of the annihilation of a photon of frequency $\omega_{\mathbf{q}}$, wave vector \mathbf{q} and polarization j , $\mathbf{e}_{\mathbf{q}j}$ is the unit vector of the photon polarization, e and m are the charge and mass of the free electron, and $\varepsilon_{\mathbf{q}}$ is the dielectric constant of the crystal. (We shall henceforth take the volume of the system to be $v = \hat{1}$.) The matrix elements of the momentum operator $\hat{\mathbf{p}}$ in (6) are made up of periodic parts of the Bloch functions of the conduction band and the valence band (with allowance for the orbital degeneracy of the latter) and are of the form

$$P_{c,v}^{(\pm)} = \int_{(v_0)} u_{k,c}^*(\mathbf{r}) \hat{\mathbf{p}} u_{0,v}^{\pm}(\mathbf{r}) d\mathbf{r}, \quad (7)$$

$$P_{c,v}^{(\pm)} = \int_{(v_0)} u_{k,c}^*(\mathbf{r}) \hat{\mathbf{p}} \{ u_{0,v}^{\pm}(\mathbf{r}) \pm i u_{0,v}^{\mp}(\mathbf{r}) \} d\mathbf{r}.$$

Here v_0 is the volume of the unit cell of the crystal. The functions $u_{0,v}^X$, $u_{0,v}^Y$, $u_{0,v}^Z$, transform under symmetry operations in analogy with the functions X , Y , Z .

We must consider the action of the operator (6) on the basis of exciton wave functions corresponding to E_1-E_8 .

$$\begin{aligned} \Psi_1 &= |\mathbf{K}; \downarrow, -3/2\rangle, & \Psi_8 &= |\mathbf{K}; \uparrow, 3/2\rangle, \\ \Psi_{2,3} &= (-c_2, c_3) |\mathbf{K}; \downarrow, -1/2\rangle + (c_3, c_2) |\mathbf{K}; \uparrow, -3/2\rangle, \\ \Psi_{4,5} &= (-c_4, c_5) |\mathbf{K}; \downarrow, 1/2\rangle + (c_5, c_4) |\mathbf{K}; \uparrow, -1/2\rangle, \\ \Psi_{6,7} &= (-c_6, c_7) |\mathbf{K}; \downarrow, 3/2\rangle + (c_7, c_6) |\mathbf{K}; \uparrow, 1/2\rangle, \end{aligned} \quad (8)$$

where

$$|\mathbf{K}; \sigma, \Sigma\rangle = \sum_{\mathbf{k}} \varphi(\mathbf{k} - \beta^* \mathbf{K}) a_{\mathbf{k}-\mathbf{K},\sigma}^+ b_{\mathbf{k},\Sigma} |0\rangle,$$

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

$$c_{4,5} = \left[\frac{\Delta (\Delta \pm (\Delta^2 + 4J^2)^{1/2}) + 2J^2}{\Delta (\Delta \pm (\Delta^2 + 4J^2)^{1/2}) + 4J^2} \right]^{1/2}$$

$$c_{6,7} = c_{2,3} (\Delta_1 \rightarrow \Delta_2).$$

Here $\varphi(\mathbf{k} - \beta^* \mathbf{K})$ is the Fourier transform of the function of the relative motion of the electron and hole in the 1s exciton, $\beta^* = m_h^*/m_e^*$, and \mathbf{K} is the wave vector of the exciton. The obtained squares of the matrix elements $M_{0j} = M_j$ ($j = 1, \dots, 8$), which determine the probabilities of the optical transitions from the ground state of the crystal to the excitonic state E_j (and the singularities in the reflection), are equal to

$$M_1^- = M_8^+ = 1/2 A(\mathbf{K}) (1 - e_{\mathbf{K}z}^2), \quad M_1^+ = M_8^- = 1/3 c_2^2 M_1^-, \quad (9)$$

$$M_4^- = M_5^+ = 1/2 c_2^2 M_1^-, \quad M_{2,3} = c_{2,3}^2 B(\mathbf{K}), \quad M_{4,5} = c_{4,5}^2 B(\mathbf{K}),$$

$$M_1^+ = M_2^{\pm} = M_3^{\pm} = M_6^{\pm} = M_7^{\pm} = M_8^- = M_1^{\pm} = M_4^{\pm} = M_5^{\pm} = M_6^{\pm} = 0,$$

where

$$A(\mathbf{K}) = \frac{2\pi\hbar e^2}{m^2 \varepsilon_{\mathbf{K}} \omega_{\mathbf{K}}} \left| \int_{(v_0)} u_{c,k}(\mathbf{r}) \beta_k u_{v,0}^{\pm}(\mathbf{r}) d\mathbf{r} \right|^2, \quad (10)$$

$$B(\mathbf{K}) = 2/3 A(\mathbf{K}) e_{\mathbf{K}z}, \quad \xi = X, Y, Z, \quad \xi' = x, y, z.$$

As seen from (9) and (5), the MCDR spectrum, if the assumptions made are correct, should be determined by the splitting of the levels E_1 and E_8 , transitions to each of which are allowed only in one circular polarization. At the same time, in the case of the "Zeeman effect" one should observe five equidistant lines, the outermost of which and the central line should be active in σ light, and the two inner ones, which are symmetrical about the central σ line, should be active in π light. The dependence of ΔE on the field H_0 at $G_h = G_e$, and the polarizations in which the lines corresponding to the transitions to the state E_j are active, are shown in Fig. 7b, where the value $G_h(H_{\max}) = 2.5$ meV was used to reconcile the calculation with the experimental data. The relative values of $M_j^{\pm(n)}$ from (9) do not contradict the experimentally observed intensities of the reflection lines (see Fig. 6). This theoretical result agrees fully with the experimental data reported above.

It can thus be stated that the observed magneto-optical effects can be qualitatively attributed fully to exchange interaction of the carriers bound into the free exciton with the isovalent Mn^{2+} impurity. We note that in this mechanism the diamagnetic shifts of the exciton bands should be determined only by the direct action of the external magnetic field on the electron and the hole, and should be insignificant in comparison with the spin splittings due to the effective exchange field.

¹Experiment

²Theory

³Crystals

⁴The absolute sign of the circular dichroism was not determined in the OMDR measurements.

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