Faraday effect in semimagnetic semiconductor $Cd_{1-x}Mn_xTe$: anomalies of the spectral, magnetic-field, and temperature dependence

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The Faraday effect in crystals of the semimagnetic semiconductor $Cd_{1-x}Mn_xTe$ ($0 \leqslant x \leqslant 0.3$) was investigated in the spectral range 0.6–1.8 eV at temperatures 4.2–350 K using magnetic fields up to 250 kOe. Dispersion of the Faraday rotation was observed, due to a change in the rotation as a function of the composition and temperature. The results were analyzed using an approach in which the Faraday rotation was regarded as a sum of three contributions. The observed deviation from saturation of the Faraday rotation in strong magnetic fields was interpreted within the framework of an antiferromagnetic exchange interaction between Mn^{2+} ions. An analysis of the temperature dependence of the Faraday rotation indicated that the appearance of a spontaneous Faraday effect was typical of semimagnetic semiconductors in the spin glass phase.

INTRODUCTION

Crystals of semiconductor $Cd_{1-x}Mn_xTe$ solid solutions are typical representatives of a new class of materials known as semimagnetic semiconductors. In fact, the giant spin splitting of exciton states and the associated giant enhancement of the interband Faraday effect,[1,2] both due to an exchange interaction between the band carriers and the localized magnetic moments of Mn^{2+} ions, were first observed in CdTe:Mn and $Cd_{1-x}Mn_xTe$. The investigations reported in Refs. 1 and 2 and the later work on the Faraday effect in other semimagnetic semiconductors concentrated mainly on the strong rise of the absolute value of the Faraday rotation angle and largely ignored the direction of rotation. The first reports of a complex nature of the dispersion of the Faraday rotation accompanied by a change in the sign of the angle of rotation associated with the transition from binary compounds CdTe and ZnTe to their solid solutions $Cd_{1-x}Mn_xTe$ and $Zn_{1-x}Mn_xTe$ were reported only relatively recently.[3,4] Moreover, the previous investigations were limited primarily to helium temperatures and the region near the absorption edge, i.e., to the experimental conditions under which the exchange interaction effects were manifested more strongly and the characteristics of the temperature dependence of the Faraday rotation[5,6] were investigated in the range of temperatures where a transition to the spin glass state took place. Moreover, the range of magnetic fields in which the Faraday rotation measurements were made was also limited. The exception was the study reported in Ref. 7, where an increase in the Verdet constant of $Cd_{1-x}Mn_xTe$ solid solutions was observed in pulsed magnetic fields up to 1500 kOe. Theoretical analyses of the experimental data on the Faraday effect in semimagnetic $Cd_{1-x}Mn_xTe$ solid solutions were made in Refs. 3 and 8, but their common shortcoming was that they considered only the exciton mechanism.

The investigation reported below was concerned with various aspects of the Faraday effect in semimagnetic semiconductors and involved a detailed investigation of the characteristic features of the spectral, composition, magnetic-field, and temperature dependences of the Faraday rotation in $Cd_{1-x}Mn_xTe$ crystals. In the interpretation of the results an allowance was made for the fact that the Faraday effect includes contributions from at least three mechanisms and the observed anomalies are due to competition between these mechanisms.

EXPERIMENTAL METHOD

Measurements of the Faraday rotation angle were made using several variants of methods that depended on the nature of the source of a magnetic field. The range of intensities of magnetic fields used in the present study could be divided into three: a) weak (50–500 Oe) static fields created by small copper solenoids; b) moderate static fields (0.5–60 kOe) created by an SP-58B electromagnet and a superconducting solenoid; c) strong pulsed magnetic fields of intensities up to 250 kOe.

In the case of static magnetic fields we used a two-beam measurement method in which the light transmitted by a sample was split by a Rochon prism (which acted also as an analyzer) into two light fluxes which were then modulated by a phase shift of 180°. In the absence of a magnetic field a balance was achieved by equalizing the intensities of the two light fluxes, and when the field was applied the resultant unbalanced signal was recorded by an automatic plotter. The source of monochromatic light in the range 0.6–2 μm was a high-luminosity MDR-3 diffraction monochromator.

Experiments in weak magnetic fields were essential because only in such fields could we observe clearly a transition to the spin glass state. Moreover, from the methodological point of view it was much easier to find the temperature dependence of the Faraday rotation in weak fields because we could then use a controlled optical helium cryostat. We employed a UTREKS system in the cryostat part of which there was a combined holder and manipulator to which a copper-wire solenoid with an internal diameter 7 mm was attached. The UTREKS system ensured automatic temperature control to within $\pm$ 0.01 K.

The magnetic-field dependence of the Faraday rotation in the field of an SP-58B electromagnet and a superconducting solenoid was determined at fixed temperatures of 5, 80, and 300 K, and 4.2 K, respectively. The experiments in a pulsed magnetic field were made employing the conventional method.[9] The rise time of the field pulses was 7 ms and the amplitude of these pulses was up to 250 kOe. A comparison

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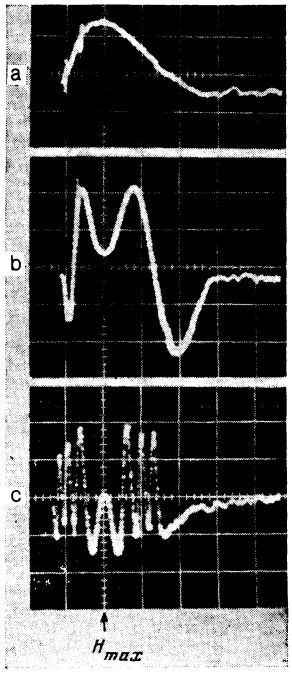

FIG. 1. Oscillograms of the Faraday rotation in $Cd_{1-x}Mn_xTe$ crystals subjected to pulsed magnetic fields: a) $x = 0$, $E = 1.44$ eV, $H_{max} = 45$ kOe, $T = 300$ K; b) $x = 0.2$, $E = 1.7$ eV, $H_{max} = 15$ kOe, $T = 300$ K; c) $x = 0.05$, $E = 1.44$ eV, $H_{max} = 190$ kOe, $T = 80$ K. The oscillatory pattern of the dependence of light transmitted by a polarizer–sample–analyzer system was due to an increase in the Faraday rotation angle and the change in the polarity of the signal in the region of the rise and fall of H indicated a change in the sign of the rotation from positive for CdTe (a) to negative for $Cd_{1-x}Mn_xTe$ (b,c).

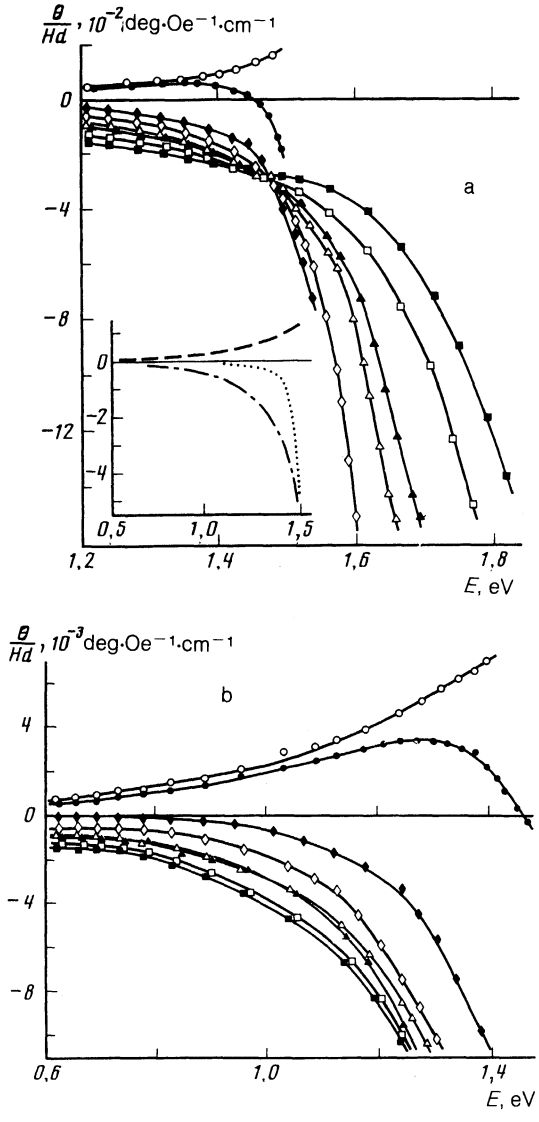

FIG. 2. Spectral dependence of the Faraday rotation obtained for different compositions of $Cd_{1-x}Mn_xTe$ crystals at 300 K in the region of the absorption edge (a) and at longer wavelengths (b): ○) $x = 0$; ●) 0.007; ◆) 0.05; ◇) 0.1; △) 0.15; ▲) 0.2; □) 0.25; ■) 0.3. In the inset the chain, dotted, and dashed curves represent qualitatively the spectral dependences of, respectively, the negative contributions $\theta_1(E)$ and $\theta_2(E)$ and the positive contribution $\theta_3(E)$ in the case when $x = 0.007$, $E_g = 1.52$ eV, $E_0 = 1.51$ eV, $E_{eff} = 2.43$ eV, and $T = 300$ K.

of the results obtained using pulsed fields with measurements of the Faraday effect (in the same samples) in a static magnetic field made it possible to reduce the errors typical of the pulsed method down to 5%. Examples of oscillograms illustrating the Faraday rotation in $Cd_{1-x}Mn_xTe$ at 80 and 300 K are shown in Fig. 1.

Single crystals of $Cd_{1-x}Mn_xTe$ ($x = 0$, 0.007, 0.05, 0.10, 0.15, 0.20, 0.25, 0.30) were grown by the Bridgman method. The composition (governed by the components of the initial charge) was determined by x-ray structure analysis. It was further checked by recording the position of the exciton structure in the reflection spectra of crystals at 4.2 K. We used the published data on the composition dependences of the lattice parameter[8] and on the energy position of the exciton line.[10] Samples used in our magnetooptic investigations were cleaved plane-parallel plates of thickness $d = 0.1$–2 mm.

EXPERIMENTAL RESULTS AND DISCUSSION

The very first experiments on $Cd_{1-x}Mn_xTe$ samples of different compositions indicated that the spectral dependence of the Faraday effect was somewhat different from that reported in earlier papers.[1,2] In addition to a strong increase in the absolute value of the Faraday rotation angle θ compared with "pure" CdTe, we found that at room temperature there were also changes in the direction of rotation (from positive for CdTe to negative for $Cd_{1-x}Mn_xTe$). Moreover, in the case of one of the samples with $x = 0.007$ we observed inversion of the sign of the angle of rotation in the Faraday effect spectrum (Fig. 2). Samples with $x > 0.05$ were characterized, up to the photon energy $E = 0.6$ eV, by a negative Faraday rotation, whereas at $x = 0.5$ there was practically no rotation in the photon energy range 0.6–0.9 eV. An increase in the concentration of Mn in the investigated solid solutions was accompanied also by a shift of the region of the strong rise of the Faraday rotation toward shorter wavelengths, which was associated with an increase in the band gap E_g as a result of such a change in the composition. It was interesting to note that, in the vicinity of $E = 1.47$ eV, mutually compensating effects of $E_g(x)$ and $\theta(x)$ kept the value of θ/Hd constant for compositions in the range $x = 0.5$–0.30.

The dispersion curves of the Faraday rotation also depended strongly on temperature. Figure 3 illustrates the influence of temperature in the case of the composition with $x = 0.007$ characterized by an inversion of the sign of the

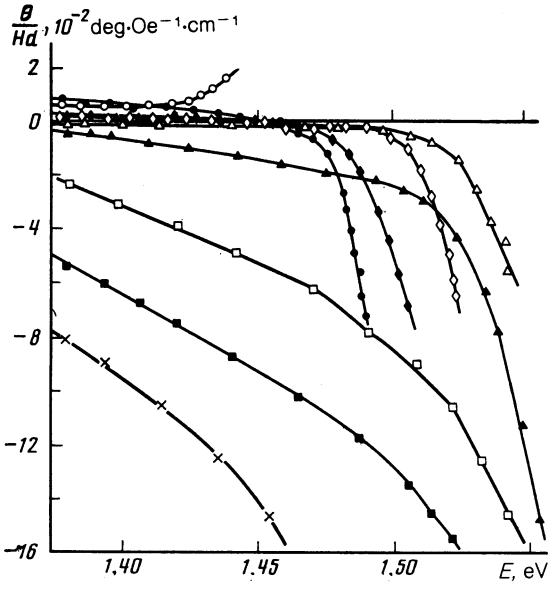

FIG. 3. Faraday rotation spectra of a $Cd_{0.993}Mn_{0.007}Te$ crystal recorded at different temperatures: ○) 350 K; ●) 270 K; ◆) 230 K; ◇) 200 K; △) 140 K; ▲) 85 K; □) 20 K; ■) 12 K; ×) 4.2 K.

angle of rotation at 300 K. Clearly, the inversion disappeared both on increase in temperature and also as a result of cooling. A shift of the steep part of the rise of the Faraday rotation toward shorter wavelengths was observed up to 140 K, but then in the range 85–4.2 K the enhancement of the Faraday rotation was the dominant effect. It was remarkable that at temperatures 140–200 K the rotation was practically zero in a wide range of photon energies.

The experimental results indicated that in explaining the anomalies of the spectral dependence of the Faraday rotation of $Cd_{1-x}Mn_xTe$ we should begin with the hypothesis of several contributions to the Faraday effect with different signs. The first contribution was the interband Faraday effect which in the case of direct-gap semiconductors was characterized by a frequency dependence $\theta_1(E) \propto A_1(E_g - E)^{-1/2}$. It was supplemented by an exciton contribution $\theta_2(E) \propto A_2(E_0 - E)^{-2}$ (Ref. 8), where E_0 is the energy position of the maximum of the exciton absorption band. The constants A_1 and A_2 in these expressions governed the signs of these contributions and the signs of A_1 and A_2 depended in the final analysis on the sign of the spin splitting of the exciton term ΔE, which was described by

$$\Delta E = x\langle S_M\rangle N_0(J_h - J_e) + (g_h + g_e)\mu_B H,$$

i.e., it included the exchange interaction and the direct influence of the applied magnetic field on the carrier spins. We used the approach of Ref. 11, according to which the interband and exciton transitions observed in the Faraday configuration should be characterized by the same spin splitting ΔE. The notation used in the above expression for ΔE is as follows: $\langle S_M\rangle$ is the average value of the magnetization of the magnetic impurity subsystem; N_0 is the number of cation states per unit volume; $J_{e,h}$ are the exchange integrals of the interaction of Mn ions with electrons and holes, respectively; $g_{e,h}$ are the effective g factors of an electron and a hole, respectively. In the case of $Cd_{1-x}Mn_xTe$ crystals we have $N_0(J_h - J_e) = -1.12$ eV (Ref. 12), so that at low temperatures and for high values of x the sign of $\theta_1(E)$ and $\theta_2(E)$ was negative. These contributions could also be positive under conditions such that the second term in the expression for ΔE had an absolute value higher than that of the first term (for low x and high T). Nevertheless, the contributions $\theta_1(E)$ and $\theta_2(E)$ were insufficient to account for the more complex dependence $\theta(E)$ characterized by an inversion of the sign of the angle of rotation, i.e., by the existence of the positive and negative Faraday rotations in different parts of the spectrum. The third positive contribution $\theta_3(E)$, governing the dispersion of the Faraday rotation at long wavelengths and the characteristic inversion of the sign in the overall dependence $\theta(E)$, can be represented by

$$\theta_3(E) \propto A_3E^2/(E_{\text{eff}}^2 - E^2),$$

where E_{eff} is the effective energy of transitions in the short-wavelength part of the spectrum. The above dependence obtained for $\theta_3(E)$ could describe the Faraday rotation due to intracenter transitions and the contribution made to the Faraday effect by interband transitions to higher energy bands.

Figure 4 shows the magnetic-field dependences of the Faraday rotation for a sample with the composition $Cd_{0.95}Mn_{0.05}Te$, which were determined using static and pulsed magnetic fields. It should be noted that in addition to saturation of the Faraday rotation in the range $H > 30$ kOe, typical of helium temperatures and obeying the Brillouin function, we also observed a monotonic rise right up to 250 kOe. This rise could be explained by an abrupt increase in the magnetization $\langle S_M\rangle$. This rise was possible in the case of an antiferromagnetic exchange interaction between the neighboring Mn^{2+} ions. In fact, the experimental results reported in Ref. 12 on the spin splitting of the exciton line of ΔE of $Cd_{0.95}Mn_{0.05}Te$ demonstrated that the dependence $\Delta E(H)$ had steps, so that we could expect similar steps in the dependence $\theta(H)$. We were unable to resolve these steps (Fig. 4) because the pulsed method for recording the Faraday rotation was too crude, but an estimate $\Delta[\theta/Hd] = 0.018$ deg·Oe^{-1}·cm^{-1} obtained for 150 kOe was comparable with the height of the first step $\Delta E_1 = 5$ meV (Ref. 12).

The temperature dependence of the Faraday rotation obtained for some $Cd_{1-x}Mn_xTe$ compositions at fixed values of the photon energy is plotted in Fig. 5. In the case of compositions in the range $x < 0.2$ we found that in a narrow

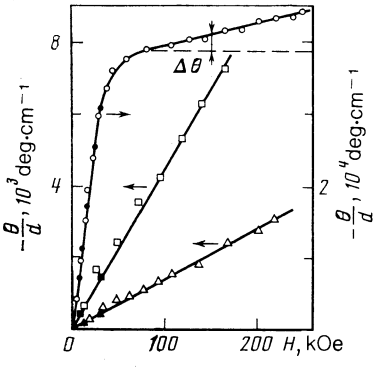

FIG. 4. Dependence of the Faraday rotation of a $Cd_{0.95}Mn_{0.05}Te$ crystal on the magnetic field: ▲), ■), ●) results of measurements in a static magnetic field; △), □), ○) measurements in pulsed magnetic fields at temperatures 300, 80, and 5 K, respectively.

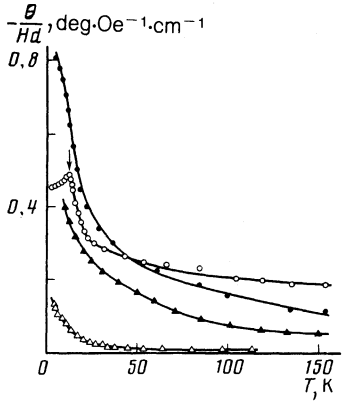

FIG. 5. Temperature dependence of the Faraday rotation in $Cd_{1-x}Mn_xTe$ samples of different compositions: △) x = 0.007, $E = 1.51$ eV; ▲) $x = 0.1$, $E = 1.55$ eV; ●) $x = 0.2$, $E = 1.65$ eV; ○) $x = 0.3$, $E = 1.84$ eV.

temperature interval 4.2–50 K there was a steep drop of the Faraday rotation, which corresponded to the temperature-induced drop in the magnetization

$$\langle S_M \rangle = S_0 B_{5/2}[{}^5/_2 g \mu_B H/k(T+T_0)],$$

where $g = 2$; μ_B is the Bohr magneton; $B_{5/2}(a)$ is the Brillouin function; and T_0 and S_0 are the fitting parameters. In the case of a $Cd_{0.7}Mn_{0.3}Te$ crystal the dependence $\theta(T)$ recorded in weak magnetic fields had a peak typical of a transition to the spin glass state. An estimate of the transition temperature $T_c = 11 \pm 0.5$ K was in satisfactory agreement with the magnetic phase diagram of semimagnetic $Cd_{1-x}Mn_xTe$ semiconductors reported in Ref. 13. The anomalies of the Faraday rotation associated with this transition had been discussed earlier in Refs. 5 and 6; however, we should make here some comments about the nature of the Faraday effects at temperatures in this range. The observed differences between the rotation values measured in weak and strong magnetic fields in the spin glass state, i.e., at temperatures $T < 11$ K, should obviously be interpreted as a manifestation of the spontaneous Faraday effect by analogy with the recently discovered spontaneous Hall effect in semimagnetic $Hg_{1-x}Mn_xTe$ semiconductors.[14]

We thus established experimentally the existence of a number of anomalies of the spectral, magnetic-field, and temperature dependences of the Faraday rotation in $Cd_{1-x}Mn_xTe$ crystals which were clearly typical of the whole class of semimagnetic semiconductors. One should stress the fact that the method of Faraday spectroscopy is very informative in the case of semimagnetic semiconductors. The main value of this method is that it is more sensitive than the direct recording of the Zeeman spin splitting of exciton and band states, so that the method could be more effective also in determination of the constants of the exchange interaction of magnetic ions both with one another and with the band carriers. The Faraday rotation method should also have advantages in studies of the spin glass state and its characteristics in the case of semimagnetic semiconductors.

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