Hidden magnetic anisotropy in $Cd_{1-x}Mn_xTe$ spin glasses

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This paper investigates the effect of optically induced polarization of photoluminescence and the mechanisms of spin relaxation of localized excitons. Polarized luminescence spectra of spin glasses have been measured for excitation by circularly and linearly polarized light at various photon energies. Calculation of the luminescence spectra of a model medium with hidden anisotropy of the localized exciton states gives a satisfactory description of the physical phenomenon. Luminescence spectra for different polarizations of the exciting light, recorded under various experimental conditions (field strength, energy of the exciting quanta), have been obtained for the case of an external magnetic field. The role of self-absorption in the formation of the luminescence spectra is demonstrated. © *1996 American Institute of Physics*. [S1063-7761(96)01407-2]

1. INTRODUCTION

Semimagnetic semiconductors, which, as a rule, are solid solutions of semiconductors of the type $A^{II}_{1-x}Me_xB^{VI}$, where Me=Mn, Fe, Co, etc. are metals with nonzero atomic magnetic moment, have been grouped into a new class of semiconductor materials.[1] The study of this group of materials, and also of low-dimensional structures based on them, has aroused significant interest. The exchange interaction of the charge carriers with the magnetic ions in semimagnetic semiconductors leads to such striking magneto-optical phenomena as giant spin splitting and Faraday rotation, the formation of a magnetic polaron, etc.[1] Among the effects due to the exchange interaction inside the system of magnetic ions, we may single out for special attention the phase transition to the spin glass state.[2]

A number of remarkable phenomena have been discovered in low-dimensional structures with semimagnetic layers. It was only in recent years on the basis of semimagnetic semiconductors that spin superlattices have been created[3] and the magnetic-field-induced transition from type I to type II has been discovered.[4] All this stimulates further study of spin effects in these materials.

Spin phenomena in nonmagnetic semiconductors and low-dimensional structures have been effectively investigated by the method of optical orientation.[5,6] However, in comparison with nonmagnetic materials, in semimagnetic semiconductors there exists an additional mechanism of spin relaxation due to the exchange interaction of the carriers with the magnetic ions.[7] This mechanism is very effective and leads to the loss of spin memory of the free current carriers and excitons excited by circularly polarized light, as a result of which the luminescence turns out to be unpolarized. Nevertheless, the authors of Ref. 8 report the first observation of a high degree of circular polarization of photoluminescence in CdMnTe and CdMnSe crystals upon selective excitation of localized excitons by circularly polarized light. They also showed that polarization of luminescence results from the presence of magnetization fluctuations. In Refs. 9 and 10 optically induced polarization of luminescence was used to investigate the localization of excitons in CdMnTe crystals in the spin-glass phase. In particular, an examination of the luminescence and polarization spectra with selective excitation of excitons made it possible to determine the mobility threshold of the latter and the energy of the magnetic polaron.

The present paper examines the nature of optically induced polarization of luminescence and the mechanisms of spin relaxation of localized excitons. As will be shown, the degree of polarization of the luminescence of CdMnTe spin glasses with selective excitation of localized states is governed by the decay parameter of the tails of the density of states and by the local exchange fields generated by the magnetization fluctuations. In experiment the latter factor is revealed as a relative spectral shift of the emission lines at different polarizations. Preliminary results of these studies were reported in Ref. 11.

Section 2 describes the experimental technique. Section 3 presents results of an experimental study of the polarized luminescence spectra of CdMnTe spin glasses in the absence of an external magnetic field and describes a qualitative model of the appearance of optically induced polarization. Section 4 presents experimental results for a longitudinal magnetic field; mechanisms of spin relaxation of excitons are considered, taking place in strong and weak external fields. Finally, Sec. 5 presents a theoretical analysis of a model of the appearance of polarized luminescence in a medium with hidden magnetic anisotropy and compares the results of calculation with experiment. The good agreement that is obtained with experiment suggest that the model is a good one.

2. EXPERIMENTAL TECHNIQUE

The measurements were carried out on single crystals of $Cd_{1-x}Mn_xTe$ ($x=0.30$, 0.35, 0.40) with zinc-blende structure. The samples were grown by the Bridgman method and were specially undoped. Wafers of thickness 0.5–0.7 mm were cleaved from bulk single crystals along the (110) plane. For the luminescence measurements we used the cleavage

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surface. The samples were centered in a superconducting solenoid and immersed in liquid helium pumped down to $T=2$ K. Optical excitation of the crystals was realized by a tunable rhodamine 6G laser pumped by an argon laser. The laser spectrum was retuned in such a way that it was possible to realize both interband and selective excitation of localized excitons. The laser beam was focused onto the surface of the sample by a long-focal-length objective ($f=15$ cm). The radiation emitted by the sample at a moderate angle ($\leqslant 10°$) to the normal was collimated by an objective, passed through a circular polarization analyzer[12] and focused onto the entrance slit of a DFS-12 spectrometer. Taking into account the large index of refraction of CdMnTe crystals, it may be assumed that the direction of the rays of the exciting light and the luminescence in such a geometry are parallel. The degree of polarization of the emitted radiation was analyzed with the help of a quartz modulator working at the frequency $\nu \approx 26$ kHz, and a two-channel photon-counting circuit.[13] The accuracy of measurement of the degree of polarization depended on the magnitude of the signal and was at worst 0.5%.

To record circular polarization spectra, we used two different experimental configurations. In the first, the sample was excited by circularly polarized light (σ^+ or σ^-) and we measured the intensity of the right-polarized (I^+) and left-polarized (I^-) luminescence. The degree of circular polarization ρ was determined from the relation $\rho=(I^+-I^-)/(I^++I^-)$. In the second, excitation was realized by light with modulated polarization ($\sigma^+ \leftrightarrow \sigma^-$) and the intensity of the luminescence was measured in one of the polarizations (I^+). For $\tilde{\rho}$ we have $\tilde{\rho}=(I_+^+-I_-^+)/(I_+^++I_-^+)$, where the subscript indicates the polarization of the exciting light. In the absence of an external magnetic field (and nonlinear phenomena in the semiconductor), ρ and $\tilde{\rho}$, generally speaking, coincide. In an external field, these two quantities take on a different meaning. In the study of polarized luminescence spectra in a magnetic field, measurements with modulation of the polarization of the exciting light are preferable, since they allow one to record photoluminescence spectra for σ^+- and σ^--excitation essentially simultaneously.

In the study of linear polarization, the samples were excited by light polarized along the Y axis, and the components of the photoluminescence polarized along the Y (I^Y) and X (I^X) axes were recorded.

3. EXPERIMENT. ZERO MAGNETIC FIELD

Mn^{++} ions, implanted in a CdTe crystal lattice, exist in a state with orbital angular momentum $L=0$ and spin $S=5/2$; the interaction between nearest neighbors has an antiferromagnetic character. The antiferromagnetic interaction of the manganese ions and the structural disorder of the implanted crystal have the result that at helium temperatures solid solutions of CdMnTe with high manganese content exist in the spin glass state.[2] As is well known, the magnetic moments in a spin glass freeze in random directions. Spatial fluctuations of the magnetization, in terms of their influence on charge carriers, are equivalent to randomly oriented local magnetic fields. Such a system is an example of a medium

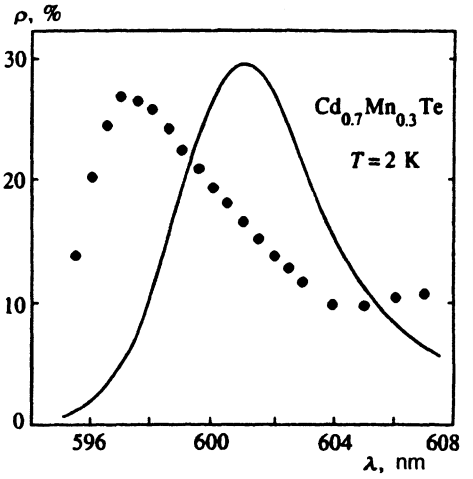

FIG. 1. Photoluminescence spectrum (solid curve) and degree of circular polarization of the luminescence (points) for selective excitation of localized excitons in a crystal of $Cd_{0.7}Mn_{0.3}Te$ by circularly polarized light: $T=2$ K, $\lambda_{ex}=594$ cm.

with hidden anisotropy. Hidden anisotropy can be manifested in luminescence only when the carriers are localized, where the dimension of the localization region does not exceed the spatial scale of the magnetization fluctuations. Otherwise, the action of the local fields on the carriers averages out and the anisotropy cannot be detected. The idea of the experiment is to use the anisotropic action of circularly polarized light on localized excitons to reveal the local magnetic anisotropy (another way of acting on the excitons, consisting of applying a magnetic field, was used in Ref. 14).

Figure 1 displays the luminescence spectrum and the circular polarization spectrum of the photoluminescence of a $Cd_{0.7}Mn_{0.3}Te$ crystal for selective excitation of the excitons in the region of the tails of the density of states. The observed line is due to recombination of the excitons localized at composition and magnetization fluctuations.[15] The shift E_p of the luminescence maximum relative to the energy of the exciting quanta ($\hbar\omega_{ex}=2.1$ eV) is due to the formation of a magnetic polaron as a result of the exchange interaction of a localized exciton with the spin system of manganese ions, and amounts to 23.5 meV (Ref. 9). In what follows, we will call this energy the polaron energy.

Under excitation by circularly polarized light into the region of delocalized exciton states, the emitted radiation is not polarized. For selective excitation, however, the luminescence turns out to be circularly polarized, and the degree of polarization varies in a complicated way within the line profile.[9] There exists a threshold energy E_0 such that for $\hbar\omega_{ex}>E_0$ the polarization of the luminescence disappears,[10] but the position of the photoluminescence line ceases to vary with increasing $\hbar\omega_{ex}$. The energy E_0 has been identified with the mobility threshold of the excitons.[9]

Figure 2 displays the photoluminescence polarization excitation spectrum, i.e., the dependence of ρ on the energy of the exciting photons. This same figure also plots the energy position of the maximum of the luminescence line as a function of $\hbar\omega_{ex}$. The decrease in ρ with increasing energy of the exciting photons is connected with the transition to the

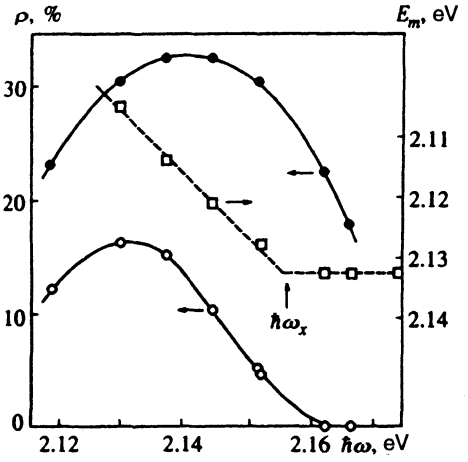

FIG. 2. Position of the maximum of the photoluminescence line of $Cd_{0.65}Mn_{0.35}Te$ as a function of $\hbar\omega_{ex}$ (squares), and photoluminescence polarization excitation spectra. The degree of polarization was recorded at the maximum of the polarization spectrum (filled circles) and in its long-wavelength wing (empty circles).

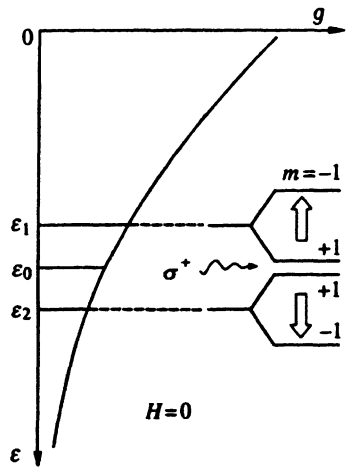

FIG. 3. Diagram of the creation of optically induced polarization of photoluminescence: $g(\varepsilon)$ is the density of exciton states, unperturbed by magnetic fluctuations. The exciting σ^+ quanta with energy $\hbar\omega_{ex}=E_0-\varepsilon_0$ are absorbed with the formation of excitons at the spin sublevels split by the local field. Since $g(\varepsilon_1)>G(\varepsilon_2)$, the excitons situated in the local field $+H_L$ are preferentially excited.

region of delocalized states and is due to rapid spin relaxation of the carriers.

For resonant excitation of excitons, optically induced circular polarization of the luminescence (optical orientation) is usually accompanied by linear polarization of the radiation (optical alignment).[16] However, for excitation by linearly polarized light under the same experimental conditions we were not able to detect linearly polarized luminescence. The reason for its absence is elucidated in Sec. 5. Here we simply note that the opposite situation has been observed in a solid solution CdSSe, in which circular polarization of the photoluminescence of the localized excitons was absent.[17]

The experimentally observed circular polarization of photoluminescence is due to splitting of the localized exciton state in the local exchange fields connected with the magnetization fluctuations. The mechanism for the generation of polarized luminescence can be elucidated in the following way (see Fig. 3). Let $g(\varepsilon)$ be the density of exciton states, unperturbed by magnetic fluctuations, in the region below the mobility threshold, and let the excitation be effected by circularly polarized light (for definiteness, σ^+) with photon energy $\hbar\omega_{ex}<\hbar\omega_0=E_0$, where $\hbar\omega_0$ corresponds to excitation of excitons at the mobility threshold. Next, for simplicity let the local field have a fixed value with the understanding that it can be directed either parallel or antiparallel to the ray (i.e., to the angular momentum of the photon). In addition, we assume that the state of the exciton is doubly degenerate in the projection of the angular momentum, $m=\pm 1$. In this case, the localized states of the excitons with energies ε_1 and ε_2 are split in the local fluctuation fields $+H_L$ and $-H_L$, respectively, so that the spin components of both states find themselves in resonance with the exciting light (Fig. 3). In addition, the optical transitions to these states are allowed by the angular momentum selection rules, while the transitions to the states $\varepsilon_1(-H_L)$ and $\varepsilon_2(+H_L)$ are forbidden. Thus, circularly polarized light will excite the spin sublevels $\varepsilon_1(+H_L)$ and $\varepsilon_2(-H_L)$ with probabilities proportional to $g(\varepsilon_1)$ and $g(\varepsilon_2)$, respectively. For a localized exciton in the local field $-H_L$ the state with spin projection $m=+1$ is not the ground state. As a result of spin flip, the exciton winds up in the state with spin projection -1, i.e., it aligns itself with the local field $-H_L$ and as a consequence (after the formation of a magnetic polaron) recombines with the emission of a σ^- photon (it is assumed that $\mu_B g H_L \gg kT$). As for a localized exciton in the local field $+H_L$, it finds itself in the spin ground state ($m=+1$) and recombines with the emission of a σ^+ photon after first forming a magnetic polaron.

The qualitative picture of the phenomenon laid out above has allowed us to draw the following conclusions:

a) the intensities of the right (σ^+) and left (σ^-) polarized luminescence are not equal; their ratio is equal to the ratio of the corresponding densities of states $g(\varepsilon_1)/g(\varepsilon_2)$;

b) the maximum of the σ^- line is shifted toward longer wavelengths relative to the maximum of the σ^+ line in the simple qualitative model by the magnitude of the Zeeman splitting $\mu_B g H_L$ in the local field H_L.

These conclusions allow us to qualitatively explain the occurrence of circular polarization of the luminescence and its spectral shape, as shown in Fig. 1.

A good illustration of the fidelity of the qualitative treatment developed above is given by Fig. 4, which presents spectra of the photoluminescence of localized excitons in both σ^+ and σ^- polarizations. It is apparent, first, that the intensities of the lines are not equal, $I^+>I^-$, and second, that there is a spectral shift between the two lines. The magnitude of this shift amounts to about 1.5 meV, although its measurement accuracy is not high, due to the large width of both lines. Note that thanks to the high-frequency quartz modulator, it was possible to measure the intensities I^+ and I^- almost simultaneously (the effect of slow signal drift was eliminated), which made it possible to accurately record the relative line shift.

A quantitative analysis of the model set forth above will be given below.

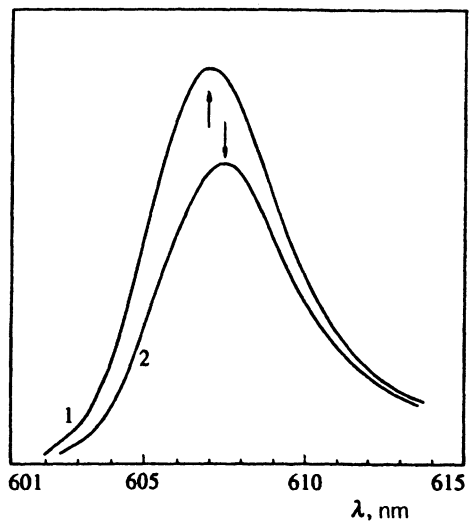

FIG. 4. Photoluminescence spectra of $Cd_{0.7}Mn_{0.3}Te$ in polarized σ^+ (curve 1) and σ^- (curve 2) for resonant excitation of localized excitons by light with σ^+ polarization: $T=2$ K, $\lambda_{ex}=600$ nm.

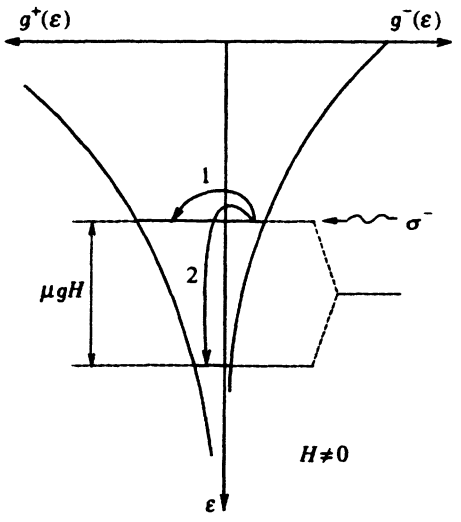

FIG. 5. Schematic depiction of spin relaxation of excitons excited on non-ground-state branches of a spin multiplet in an external magnetic field: $g^+(\varepsilon)$ and $g^-(\varepsilon)$ are the tails of the density of states for $m=+1$ and $m=-1$, split by the external field. Possible paths of the transition of an exciton to the ground spin state are shown: an elastic process with spatial transfer of the exciton (channel 1) and spin flip of the exciton within the limits of one localization region (channel 2).

4. EXPERIMENT. LONGITUDINAL MAGNETIC FIELD

The interpretation of polarized photoluminescence spectra proposed in the preceding section, in which the spin relaxation of the localized excitons is due to inelastic spin-flip processes, should find confirmation in experiments on selective photoexcitation of localized states in an external magnetic field. Indeed, under such conditions, spin flip of the exciton will take place not in the local field of a magnetic fluctuation, the magnitude of which is difficult to influence from outside, but in the total field (''local''+external). The corresponding spectral shift of the emission line will now depend on the magnitude of the applied magnetic field. Having the ability to vary the external field, we can realize those experimental conditions under which the efficiency of the inelastic spin-flip processes is clearly revealed. On the other hand, the application of an external field leads, as will be shown, to a number of effects bearing no relation to spin relaxation of excitons. The influence of these effects, associated with reabsorption of luminescence and with the finite width of the sample, must be taken into account in an analysis of the experimental results.

Before going on to a presentation of the experimental results, we indulge in some preliminary remarks. When a magnetic field H is applied, the crystal becomes magnetically anisotropic: there is a preferred direction. In the region of strong external fields, $H \gg H_L$, quantization of the exciton spin sublevels takes place along the direction of H. For one sign of the polarization of the exciting light (σ^+) the ground state of the spin multiplet is excited, that is, the state of the heavy exciton with spin projection $m=+1$. But another state can also be excited, namely the light exciton with spin projection $m=+1$. For the other sign of the polarization of the exciting light (σ^-) the transition to the ground state is forbidden by the selection rules, so two non-ground-state components of the spin multiplet, having the spin projection $m=-1$, are excited. Nevertheless, the luminescence for both signs of the polarization of the exciting light is strongly polarized, and the sign of its polarization is determined by the direction of the field. This circumstance indicates the presence of highly efficient spin relaxation of the excitons.

When a non-ground spin state of the localized exciton is excited, the spin relaxation can be either elastic or inelastic (Fig. 5). In the region of localized states the process of elastic spin relaxation in a magnetic field should be accompanied by spatial transfer of the exciton (channel 1); inelastic relaxation can also take place without transfer of an exciton (channel 2). Both type-I relaxation (resonant tunneling with spin flip) and type-II relaxation (spin flip without tunneling) are possible in principle. The relative efficiency of these processes depends, as we shall see, on the magnitude of the magnetic field.

Figure 6 presents characteristic photoluminescence spectra of CdMnTe in a strong magnetic field $H=4.4$ T. The pairs of spectra (a, b, c) correspond to different photon energies of the exciting light. For each $\hbar\omega_{ex}$ photoluminescence spectra are shown for σ^+ and σ^- excitation (I_+ and I_-, respectively), and in Fig. 6b also their relative difference $\tilde{\rho}$. In all cases the photoluminescence signal in the σ^+ polarization was recorded. At all $\hbar\omega_{ex}$ smaller than $E_0(H)$ a significant Stokes shift of the luminescence line is observed, which is due to the formation of a magnetic polaron. (A detailed analysis of the dependence of this shift on $\hbar\omega_{ex}$ and H is given in Ref. 10.)

It is evident from the spectra that over a certain range of $\hbar\omega_{ex}$ the maximum of the I_- line is shifted toward longer wavelengths relative to the I_+ maximum (Fig. 6b). Superficially, such behavior is reminiscent of the effect described above in zero external field. However, to interpret it in a similar way is disallowed by the following circumstance. In a number of cases, under different experimental conditions (H,$\hbar\omega_{ex}$) the long-wavelength limbs of the I_+ and I_- lines are observed to coincide, with their signal intensities being

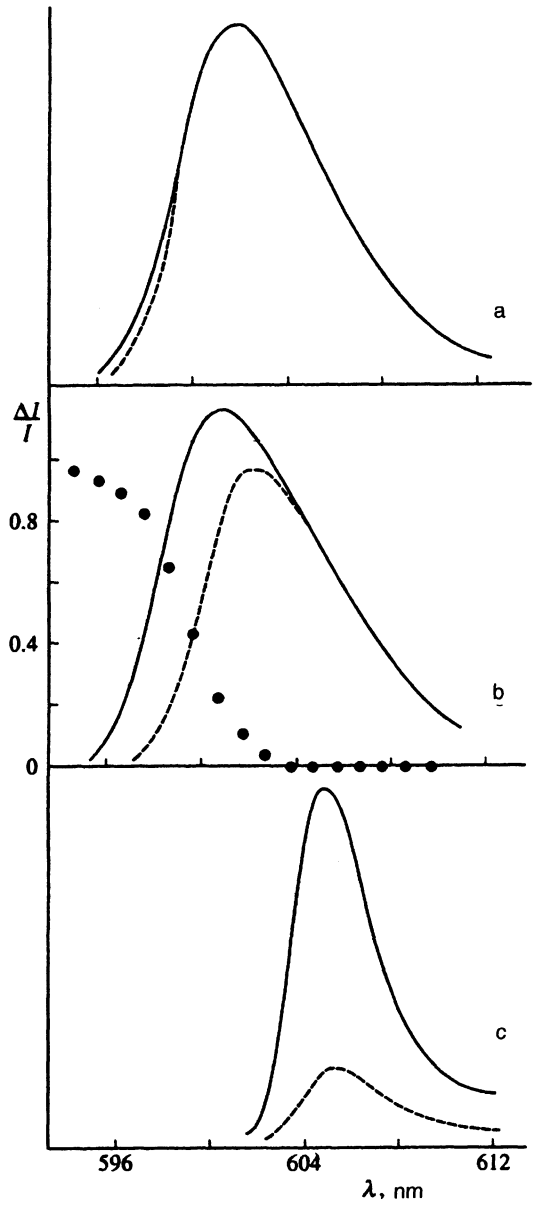

FIG. 6. Photoluminescence spectra I_+^+ and I_-^+ (for the notation, see Sec. 2) of a crystal of $Cd_{0.7}Mn_{0.3}Te$ in an external magnetic field $H=4.4$ T for modulation of the polarization of the exciting light; $T=2$ K, excitation wavelength: a) 588 nm, b) 592 nm, c) 602 nm. Points depict the spectrum $\tilde{\rho}$.

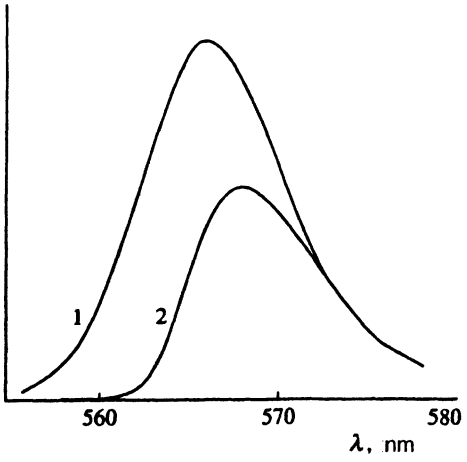

FIG. 7. Photoluminescence spectra of $Cd_{0.6}Mn_{0.4}Te$ in the "transillumination" geometry (curve 2) and in the "reflection" geometry (curve 1); $T=2$ K, $H=0$, interband excitation $\lambda_{ex}=514.5$ nm.

equal to high accuracy ($\leqslant 0.5\%$), which compels us to search for a general reason for this coincidence. Such a reason, in our opinion, is self-absorption, that is, the well-known absorption of the luminescence in the sample itself.[18]

In the vicinity of the fundamental absorption edge, where the density of states varies abruptly within the photoluminescence line profile, self-absorption leads to a substantial narrowing of the photoluminescence spectrum. The short-wavelength edge of the spectrum is absorbed most strongly; therefore the maximum of the recorded spectrum turns out to be shifted toward longer wavelengths in comparison with the maximum of the true exciton luminescence spectrum. In the absence of an external field or in the case of interband excitation in the presence of a field, the shapes of the I_+ and I_- lines are identically distorted (or not distorted) by self-absorption, and one needs to take special measures to detect this effect. (For example, by recording the photoluminescence spectra in "transillumination" and "reflection" geometries.[19])

Another situation develops for selective excitation in a field. In this case, the absorption depths of σ^+ and σ^- light, respectively α_+^{-1} and α_-^{-1}, turn out to be, first of all, different and, second, comparable with the absorption depth of the light at the luminescence frequencies $\alpha_L^{-1}(\omega)$. To the extent that $\alpha_+^{-1}<\alpha_-^{-1}$ and the self-absorption effect more strongly distorts the I_- line, its maximum is that much further shifted toward longer wavelengths than is the maximum of the I_+ line. The magnitude of this effect grows as the field increases.

The coincidence of the long-wavelength wings of the lines thus finds a natural explanation: in this wavelength region self-absorption is negligibly small; however, the exciting light is completely absorbed in the sample, and therefore the intensities are exactly equal. This indicates that the true luminescence spectra I_+ and I_- coincide, but the observed difference in the short-wavelength region of the spectrum is due to self-absorption.

A control measurement which we performed of photoluminescence spectra in the "transillumination" and "reflection" geometries confirmed the important role of self-absorption in the formation of the photoluminescence spectra of the investigated crystals. Corresponding spectra of $Cd_{0.6}Mn_{0.4}Te$ for $\hbar\omega_{ex}>E_g$, when the exciting light is absorbed in the near-surface region, are shown in Fig. 7. The luminescence spectra in the "transillumination" geometry is indeed shifted toward longer wavelengths relative to the spectrum obtained in the "reflection" geometry. At the same time, the long-wavelength wings of the lines essentially coincide. The reason for the shift is the same here: when the luminescence radiation passes through the sample, the spectrum is strongly distorted as a result of self-absorption.

Note that with a further decrease in $\hbar\omega_{ex}$ the long-

wavelength wings of the I_+ and I_- lines cease to coincide (Fig. 6c) while the maxima of the lines approach each other. This happens when the exciting light in the σ^- polarization (and with a further decrease in $\hbar\omega_{ex}$, in both polarizations) ceases to be completely absorbed in the sample with thickness d: α_-^{-1}, $\alpha_+^{-1} \geqslant d$.

Thus, although a shift between the maxima of the I_+ and I_- lines is observed in a strong field, it is not attributable to type-II relaxation. The dominant mechanism of spin relaxation is the type-I elastic process. As for type-II relaxation, its low efficiency in comparison with type-I relaxation can be explained by the great difference in the density of final exciton states for these processes (see Fig. 5). The stronger the field, the greater the difference and consequently the greater the splitting of the exciton states.

In a weak field the situation changes. The densities of final states for type-I and type-II processes now differ only slightly, and the type-II process can become dominant since a type-I process requires transfer of excitation between neighboring localization regions. This is illustrated by Fig. 8, which displays luminescence spectra for σ^+ and σ^- excitation in a field $H=0.75$ T. Here we also observe a shift between the maxima of the I_+ and I_- lines; however, the "departure" of the wing of the I_- line beyond the profile of the I_+ line (Fig. 8b) can in no way be explained by self-absorption since self-absorption leads only to a decrease in I_- in comparison with I_+. This forces us to give preference to the type-II mechanism, which, in contrast to the type-I mechanism, leads to a shift in the maximum of the I_- line in the long-wavelength direction from the I_+ line. At the same time, the influence of self-absorption on the shift between the maxima is less pronounced in this case than in a strong field, since the absorption coefficients of σ^+ and σ^- light are closer in magnitude.

With a decrease in the energy of the exciting quanta, the profile of the I_- line "subsides" (Fig. 8c). As in the case of a strong field, this results from the finite thickness of the sample, $\alpha_-^{-1}(\hbar\omega_{ex}) > d$.

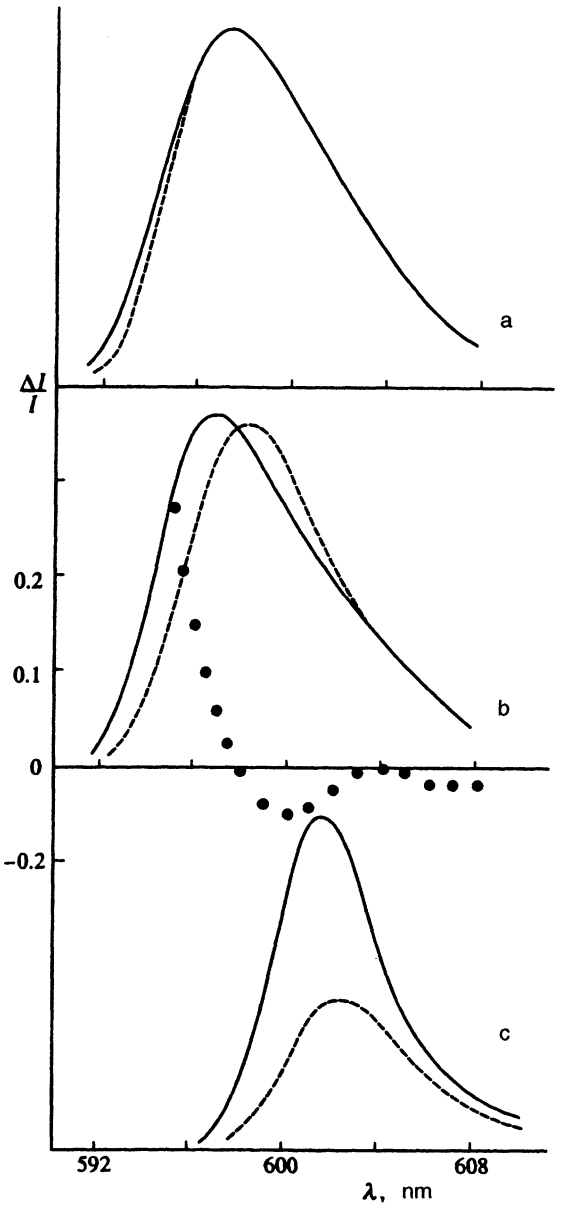

FIG. 8. The same as in Fig. 6, but for $H=0.73$ T. Wavelength of the exciting light: a) 588 nm, b) 590 nm, c) 596 nm.

5. CALCULATION OF POLARIZED LUMINESCENCE SPECTRA

Following the method proposed in Ref. 8, we calculate the polarized luminescence spectra for resonant circularly polarized and resonant linearly polarized excitation of excitons in a medium with hidden magnetic anisotropy. To start with, let selective photoexcitation of localized exciton states be realized by means of circularly polarized light. We assume that with the localization region of each exchange state there is associated in a random way an oriented local exchange field causing spin splitting of the exciton state. Thus, the magnitude of the spin splitting Δ can be considered a characteristic of the hidden anisotropy. We assume that $\Delta \gg kT$.

The magnitudes and signs of the exchange constants of the electrons ($\alpha N_0 = 0.22$ eV) and holes ($\beta N_0 = -0.88$ eV) are such that they lead to the indicated ordering of the six optically active spin states of the exciton. As a consequence of the random orientation of the local fields, all six states can be excited by circularly polarized light. However, the probability of excitation turns out to be different for different spin branches, and this difference is governed by two factors: the form of the density of states and the angular momentum selection rules. Ignoring for the moment the first of these factors, let us consider the evolution of a localized exciton from its time of creation to recombination.

The probability of excitation by circularly polarized light of any of the spin sublevels depends on the angle θ between the local field and the wave vector of the incident light. In the dipole approximation it is not hard to obtain the following expressions for the transition probabilities to each sublevel:

$$I_1 \propto \frac{3}{4}(1+\cos\theta)^2, \quad I_2 \propto \sin^2\theta, \quad I_3 \propto \frac{1}{4}(1+\cos\theta)^2,$$

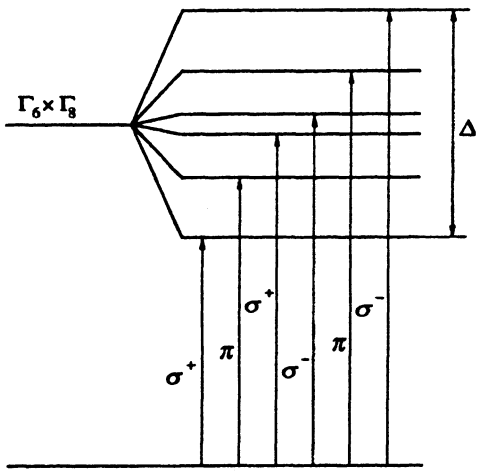

FIG. 9. Spin splitting of an exciton state. Two optically inactive sublevels are excluded from the multiplet. Optical transitions allowed by the selection rules are shown.

$$I_4 \propto \frac{1}{4}(1-\cos\theta)^2, \quad I_5 \propto \sin^2\theta, \quad I_6 \propto \frac{3}{4}(1-\cos\theta)^2; \tag{1}$$

where I_k is the excitation rate of the kth sublevel, and the subscript k labels the spin sublevels.

The fastest of the processes following excitation of the exciton is probably spin relaxation.[7] Scattering off manganese ions and clusters, the exciton winds up on the lower σ^+-branch of the spin multiplet (Fig. 9). This is followed by the formation of a magnetic polaron, leading to a lowering of the energy of the exciton, but preserving inviolate the anisotropy axis. The formation of a polaron is manifested in the photoluminescence spectrum as a Stokes line shift by the polaron energy E_p.

Finally, recombination of the exciton takes place, and the probabilities of emission of a photon with spin projection $+1$ or -1 in the look direction also turn out to depend on θ. Taking into account the recording geometry, it is necessary to append a factor $[1+\cos(\pi-\theta)]^2$ or $[1-\cos(\pi-\theta)]^2$ to the expressions for the intensity of the circular components of the photoluminescence. The final expressions look like this:

$$I_1^+ \propto \frac{3}{4}(1+\cos\theta)^2(1-\cos\theta)^2, \quad I_1^- \propto \frac{3}{4}(1+\cos\theta)^4,$$

$$I_2^+ \propto \sin^2\theta(1-\cos\theta)^2, \quad I_2^- \propto \sin^2\theta(1+\cos\theta)^2,$$

$$I_3^+ \propto \frac{1}{4}(1+\cos\theta)^2(1-\cos\theta)^2, \quad I_3^- \propto \frac{1}{4}(1+\cos\theta)^4,$$

$$I_4^+ \propto \frac{1}{4}(1-\cos\theta)^4, \quad I_4^- \propto \frac{1}{4}(1-\cos\theta)^2(1+\cos\theta)^2,$$

$$I_5^+ \propto \sin^2\theta(1-\cos\theta)^2, \quad I_5^- \propto \sin^2\theta(1+\cos\theta)^2,$$

$$I_6^+ \propto \frac{3}{4}(1-\cos\theta)^4, \quad I_6^- \propto \frac{3}{4}(1-\cos\theta)^2(1+\cos\theta)^2. \tag{2}$$

TABLE I. Stokes shifts S_n of the components of the photoluminescence corresponding to excitation of various spin sublevels, and their relative intensities under conditions of polarized excitation.

n	S_n/Δ	I_+^+	I_+^-	I_Y^Y	I_Y^X
1	0	3	18	12	9
2	4/15	6	6	4	8
3	7/15	1	6	4	3
4	8/15	6	1	4	3
5	11/15	6	6	4	8
6	1	18	3	12	9

The subscript here, as before, indicates the excited spin sublevel, and the superscript indicates the polarization of the exciting light. All that remains is to average expressions (2) over all orientations of the local field.

The results of averaging are given in Table I in the form of numbers characterizing the relative intensity of the spectral components. If inhomogeneous line broadening is small in comparison with spin splitting in the local field, $\Gamma \ll \Delta$, then the photoluminescence spectrum will consist of a set of six lines of different intensity, with the Stokes shift in the interval from E_p to $E_p+\Delta$. Of these lines, the second and fifth, reckoning from the exciting line, are unpolarized, and the rest are approximately 70% polarized, where the sign of the circular polarization for the first and third lines is opposite that for the fourth and sixth lines.

In our case, the spectrum consists of a single line, from which we may conclude that the opposite case, $\Gamma \geqslant \Delta$, is realized here. In this case, inhomogeneous broadening "absorbs" the line structure of the photoluminescence, but peculiarities associated with spin flip of the exciton in the local field are all manifested in the polarization spectra.

Finally, it is easy to see from the table that even for $\Gamma \geqslant \Delta$ the polarization spectrum should be symmetric (or antisymmetric) about $\varepsilon = E_p + \Delta/2$. This, as we have seen, is not experimentally observed. The asymmetry in the polarization spectrum comes from the density of exciton states. Indeed, if in our mind's eye we separate out the excitation of the upper components of the exciton multiplet, then it turns out that the states that are in resonance with $\hbar\omega_{ex}$ are those states that in the absence of a local field would be located "below" $\hbar\omega_{ex}$ in the tail of the density of states. Conversely, for the excitation of the lower components of the multiplet the states that are in resonance with $\hbar\omega_{ex}$ turn out to be the states located "above" $\hbar\omega_{ex}$ on the unperturbed tail (unperturbed by the local fields). Since the density of states decreases with decreasing energy, the lower components of the exciton splitting always make a larger contribution to the photoluminescence spectrum than the upper states, which then leads to asymmetry of the polarization spectrum.

Before going on to a comparison of the results with experiment, we present the solution of the problem of optically induced linear polarization. The model of the phenomenon is analogous to that considered at the beginning of this section, the only difference being that this time the excitation is brought about by linearly polarized light and, consequently, we record the linearly polarized components of the photolu-

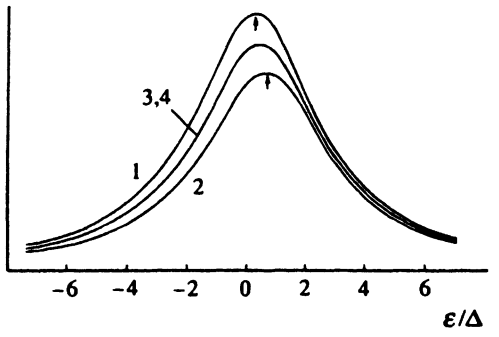

FIG. 10. Calculated spectra for polarized luminescence in zero external field. The numerals indicate the following spectra: 1) I_+^-, 2) I_+^+, 3) I_Y^Y, 4) I_Y^X. The calculation was performed for the parameters $\Gamma_0/\Delta=2.5$ and $U/\Delta=1.5$.

minescence. For example, let the excitation be brought about by light polarized along the Y axis. Following the procedure described above, this time instead of Eqs. (2) we obtain

$$I_1^X=\frac{3}{4}(\cos^2\theta+\sin^2\theta\sin^2\varphi\cos^2\varphi),$$

$$I_1^Y=\frac{3}{4}(\cos^2\theta+\sin^2\theta\cos^2\varphi)^2,$$

$$I_2^X=\sin^2\theta\cos^2\varphi(\cos^2\theta+\sin^2\theta\cos^2\varphi),$$

$$I_2^Y=\sin^2\theta\cos^2\varphi(\cos^2\theta+\sin^2\theta\cos^2\varphi),\ldots \qquad (3)$$

We did not write out all 12 expressions, since the component I_3 differs from the component I_1 only by the factor 1/3, and the components I_4, I_5, and I_6 coincide with the components I_3, I_2, and I_1, respectively.

From the results of angular averaging of the intensities (3), the reason for the lack of an "alignment" effect in the experiment is strikingly evident—it is unfavorable selection rules (see the table). Indeed, taking into account that due to the spectral shape of the density of states the contribution of the components of the spin multiplet to the intensity of the photoluminescence falls off with increasing index, it is reasonable for the sake of an estimate to take only the first two components. As is clear from the table, the corresponding intensities are in the ratio 16:17, i.e., the polarization is very small.

Figure 10 presents results of a model calculation of the polarized luminescence spectra in the system under consideration, with hidden magnetic anisotropy. Each of the lines in the figure is the sum of six "elementary" lines corresponding to excitation of the six spin components. The summation took account of the corresponding selection rules. As an "elementary" line we chose a Lorentz profile of width $2\Gamma_0$. The exponential falloff in the density of states is characterized by the parameter U.

Lines 1 and 2 correspond to the two circular components of the photoluminescence spectrum for excitation by σ light. As in the experiment, there is a notable shift between their maxima (see Fig. 4). Lines 3 and 4, which essentially coincide, correspond to the two linear components of the photoluminescence spectrum for excitation by π light. The coincidence of these lines reflects the experimentally verified lack of alignment.

In comparing the results of calculation with experiment it is necessary to take into account that the line width Γ greatly exceeds the experimentally observed interval between the maxima of I_+ and I_-. On the one hand, under such conditions it is difficult to ensure high accuracy of measurement of the shift; on the other, to calculate the polarized photoluminescence spectra correctly, one needs to know the shape of the inhomogeneously broadened "elementary" line, i.e., the photoluminescence line for excitation of only one component of the exciton multiplet. In addition, we have found that the results of our model calculation (Fig. 10) accurately reproduce the overall qualitative picture of the phenomenon (the "smooth" shape of the total lines, the ratio of the intensities I_+ and I_- of the components, the shift between their maxima, the spectral shape of the degree of circular polarization, and the lack of an alignment effect) when varying the model parameters (Γ_0/Δ and U/Δ) over rather wide limits. On the one hand, this latter circumstance can be considered an additional indication of the faithfulness of the model. On the other, a comparison of experimental results with calculation allows us only to estimate the parameters Δ, U, and Δ_0, for ignoring such factors as the spread in magnitude of the local fields or the asymmetry of the "elementary" line could lead to a loss of accuracy. Nevertheless, it is possible, quite reliably, to estimate both the scale of the model parameters and their hierarchy. For example, for the most studied solid solution with $x=0.3$, the magnitudes of all three parameters lie in the several meV range. The smallest is clearly the parameter Δ, whose magnitude we estimate at 3 meV. The parameter U has roughly the same magnitude; the ratio U/Δ is most probably somewhat greater than unity. The largest is the parameter Γ_0; we estimate the ratio Γ_0/Δ to be 2–3. The above estimate of the parameter U is in reasonable agreement with values of this parameter known from the literature[17,20] in other solid solutions (e.g., in CdSSe and CdZnTe with composition $x=0.3$, the values of U are roughly 5 and 2.5 meV, respectively).

6. CONCLUSIONS

The experimental results in this work on optically induced polarization of luminescence of CdMnTe spin glasses (Sec. 3) are in good agreement with theory (Sec. 5). The calculation, carried out within the model of a medium with hidden anisotropy, has allowed us to explain the appearance of circularly polarized luminescence and the shape of the spectral dependence of its degree of circular polarization, and also to identify the reason for the lack of an "optical alignment" effect. The calculated spectra of polarized luminescence reflect even such a nuance of the experimental picture as the only moderately noticeable shift of the maxima of the circular components of the photoluminescence relative to each other. The agreement between model and experiment leads us to believe that this model is a faithful one. Within the framework of the model we were able to estimate the scale and hierarchy of the three main parameters responsible

for the formation of the polarized luminescence spectra—the anisotropy parameter Δ, the Urbach parameter U, and the "elementary" line width Γ_0.

The experimental results of Sec. 4 allow us to judge the merits of candidate mechanisms of spin relaxation of localized excitons in an external magnetic field. While resonant tunneling of excitons with spin flip predominates in a strong external field, the dominant mechanism in a weak field is an inelastic process without spatial transfer of an exciton, related to the spin relaxation mechanism for the case of zero field. In addition, Sec. 4 underscores the necessity of taking self-absorption into account in any interpretation of the photoluminescence spectra.

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