Spin polarization of localized magnetic moments in $Cd_{0.95}Mn_{0.05}Te$ in exchange scattering by photoexcited carriers

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An influence of the intensity of optical excitation on the giant spin splitting of the exciton luminescence in $Cd_{1-x}Mn_xTe$ was observed. The effect is attributed to the change of the magnetization of the system of localized magnetic moments of Mn^{2+}, as a result of the exchange scattering by them of the light-induced carriers that are nonequilibrium in spin (the analog of the Overhauser effect in magnetic resonance). It is shown that the experimental results can be explained both qualitatively and quantitatively if reasonable values are used for the parameters that enter in the theory of the effect.

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INTRODUCTION

Giant spin splitting (GSS) of exciton states when a magnetic field acts on magnetically mixed semiconducting crystals A^2B^6:Me($A^2_{1-x}Me_xB^6$) (here Me is a $3d$-metal ion) were revealed by exciton reflection spectra[1-5] and by exciton luminescence spectra.[6,7] They were shown to be caused by exchange interaction between the carriers contained in the excitons and the localized spin moments (LSM) of the magnetic ions, and to be proportional to the spin polarization $\langle S^z_M\rangle$ produced in the LSM system by the external field **H**. The effect is due to the diagonal matrix elements of the operator of the exchange scattering of the carriers (of the electrons, e, or of the holes, h)

$$\mathscr{H}_{ex}=-2\sum_j J_{e(h),M}(\mathbf{r}-\mathbf{r}_{M_j})\mathbf{S}_{e(h)}\mathbf{S}_{M_j} \tag{1}$$

by the localized spins S_M in quasifree motion of the carriers in the crystal. The diagonal part of (1) can lead to spontaneous polarization of the system of spins S_M and $S_e(S_h)$ at sufficient carrier density.[8]

The off-diagonal part of (1) should lead to indirect exchange interaction of the LSM (the Bloembergen-Rowland mechanism via virtual carrier states, or the RKKY mechanism via real carrier states). In addition, the interaction (1) may be an effective source of longitudinal relaxation of both the LSM and of the carriers.[9] Inasmuch as under GSS conditions the values of the spin splittings of the states of the carriers and of the LSM differ greatly, the mechanism of relaxation of spin-nonequilibrium carriers via exchange scattering should, in analogy with the Overhauser effect, lead to nonequilibrium polarizations of the system of spins S_{Mj}.

The conditions for the existence of this effect were considered theoretically in Ref. 10. The present paper is devoted to the first experimental observation of the influence, predicted in Ref. 10, of the exchange carrier scattering by the spin polarization of the LSM system, using the $Cd_{0.95}Mn_{0.05}Te$ crystal as an example.

Exciton luminescence data have demonstrated earlier[7] that under GSS conditions at least the hole spin system is in a strong nonequilibrium state when carriers are produced by light with energy greatly exceeding the band gap of the crystal. It was established that the spin polarization of both carriers bound into excitons[7] and carriers localized on donors and acceptors[11] are independent of the polarization of the exciting light and correspond to a situation wherein the carriers are first depolarized in spin on account of thermalization to a high kinetic temperature, after which they are partially or fully polarized during the lifetime in the state from which the recombination takes place.

In Refs. 7 and 11 the intensity of the optical pump that generated the carriers was low, so that the carrier spin relaxation via exchange scattering could not influence the spin polarization $\langle S^z_M\rangle$ of the LSM system. At the same time, as follows from Ref. 10, at sufficiently intense generation of carriers that are not spin-polarized the value of $\langle S^z_M\rangle$ should change, since relaxation via exchange scattering takes place without a change of the total spin of the system (the total spin moment of the carriers and the LSM). The sign of the change of $\langle S^z_M\rangle$ will depend on the signs of the exchange interactions.

Under conditions when the exchange fields produced by the carriers at the spins S_{Mj} are not yet significant, and the LSM exchange fields at the carriers lead to GSS, the exchange relaxation will decrease $|\langle S^z_M\rangle|$ if the carrier-impurity exchange interaction is ferromagnetic, and increase it if the exchange is antiferromagnetic. From this point of view, the contributions to the change of $\langle S^z_M\rangle$ in CdTe:Mn from the exchange scattering of the conduction electrons and holes by S_{Mj} should be opposite ($J_{e,M}>0$ and $J_{h,M}<0$) for CdTe:Mn. If the changes of $\langle S^z_M\rangle$ due to the mechanism under consideration are small compared with S_M, the contributions from carriers of different types will, according to Ref. 10, be additive.

EXPERIMENT

The measurements were performed on $Cd_{0.95}Mn_{0.05}Te$ samples at $T=1.97$ K with the magnetic field applied in the Faraday configuration. The luminescence was excited by an He-Ne laser ($E_{ex}=1.959$ eV) with maximum

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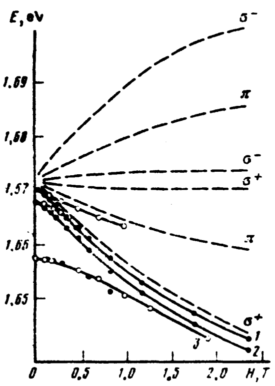

FIG. 1. Magnetic-field dependence of the spin splittings 1s $\Gamma_6-\Gamma_8$ of the exciton state at a temperature $T = 1.97$ K. Dashed lines—positions of maxima of luminescence bands of the free exciton (1), of the exciton bound to a neutral donor and to a neutral acceptor respectively (2, 3); ●) σ^+ radiation, ○) π radiation.

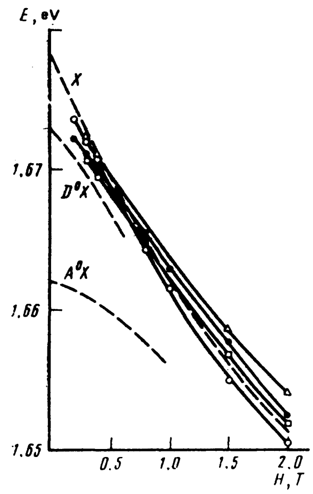

FIG. 3. Magnetic-field dependence of the exciton luminescence line in σ^+ polarization on the pump density: ○—1.5%; □—15%; ●—36%; △—100% of the maximum pump.

power ≈ 35 mW. The radiation could be focused on the sample down to a spot with diameter ≤0.5 mm. Only the emission from the irradiated section was registered.

The magnetic-field dependence of the σ^+ components of the radiation at defocused pumping, when further decrease of its density had no effect on the position of the luminescence line, is shown in Fig. 1. The spectrum includes the emission lines of the free exciton (X) and of the excitons bound to a neutral donor (D^0X) and to a neutral acceptor (A^0X). The intensity of the exciting light was varied with filters calibrated against the light transmission at 6328 Å wavelength. The absolute excitation density was not measured, but we estimate the maximum density to be ~10 W/cm^2. The σ^+ emission spectra in fields $H = 0.4$ and 1.5 T at various excitation densities are shown in Fig. 2, while Fig. 3 shows the magnetic-field dependences of the position of the σ^+ radiation of the free exciton for a number of pump level densities.

The dependence of the position of the exciton line on the pump density (the carrier generation rate) was plotted in greater detail for magnetic field values $H = 0.3$; 0.4; 1.5; 2.0 (Fig. 4). It is seen that the dependences of the positions of the exciton emission line on the pump density differ in the field regions up to 1 T from those above 1 T. In addition, two different pump-density sections are observed in which the dependence is different.

For pump densities <15% of the maximum attainable in fields 0.3 and 0.4 T one observes a line shift towards longer wavelengths with increasing pump, while in fields ≥1.5 T there is an intense shift towards shorter wavelengths. The possible spectrum changes due to heating of the crystal by the light cannot explain the observed effect, especially the emission line shift in weak fields. It must therefore be concluded that the observed line shifts in the magnetic field correspond to changes of $\langle S_M^z \rangle$, and the spin polarization of the system of localized moments increases in the weak-field region and decreases in strong fields with increasing pump.

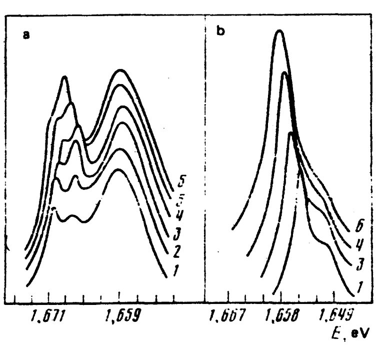

FIG. 2. Spectra of exciton radiation in fields $H = 0.4$ (a) and $H = 1.5$ T (b) at various excitation densities: 1—1.5%; 2—4%; 3—7%; 4—12%; 5—36%; 6—100% of the maximum pump I_{max}.

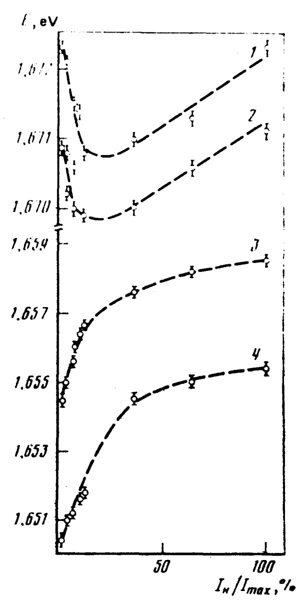

FIG. 4. Positions of the σ^+ components of the free exciton vs the pump density in magnetic fields: 1—0.3; 2—0.4; 3—1.5; 4—2.0 T.

At densities higher than ~15% of the maximum, the increase of the pump leads to weak short-wave shifts, corresponding to a decrease of $|\langle S_M^z\rangle|$ compared with the polarization at optimal pumping.

DISCUSSION

The change of the positions of the exciton-luminescence lines as a function of the pump density can be due to exciton-exciton interaction, or else to a change of $\langle S_M^z\rangle$. The employed pump densities are too small to produce exciton densities such that the exciton-exciton interaction is substantial. In addition, exciton-exciton interaction cannot lead to opposite signs of the line shifts in weak and strong fields. The change of $\langle S_M^z\rangle$ can be due to spin ordering,[8] exchange scattering,[10] and finally heating of the sample by the pump radiation. The first of these effects should lead to an increase of $|\langle S_M^z\rangle|$ and should increase with increasing GSR as the spin thermalization of the carriers increases, something not observed. Heating should decrease $|\langle S_M^z\rangle|$ and can be responsible for the behavior of the luminescence-line positions as functions of the pump only at densities >15% of the maximum. At the same time, at pump densities <15% the observed effects can, from our point of view, be due only to exchange relaxation.

Indeed, in this case the spin polarizations of the carriers and the LSM should satisfy the equation for the balance of the spin fluxes due to spin-lattice relaxation (SLR) (with times τ_{ML}, τ_{eL} and τ_{hL} respectively for S_M, S_e and S_h) and recombination (τ_r):

$$k_S(x)n_M\tau_{ML}^{-1}(x-x_0)+n_e(\tau_{eL}^{-1}+\tau_r^{-1})(y-\bar{y})+n_h(\tau_{hL}^{-1}+\tau_r^{-1})(z-\bar{z})=0, \tag{2}$$

obtained[10] from the kinetic equations for the spin polarizations of the LSM, electrons, and holes. Here x, y, and z are the normalized differences of the populations $(n_{p-1}-n_p)/(n_{p-1}+n_p)$ of neighboring spin levels $p-1$ and p for the spins S_M, S_e and S_h (p is the spin projection on the Z ∥ H direction), x_0 is the equilibrium value of x;

$$x_0=\text{th}(\omega_0/2k_BT_{\text{eff}}),\quad \omega_0=g\beta H,\quad T_{\text{eff}}=T_0+\Theta,$$

T_0 is the lattice temperature; Θ is the Curie-Weiss constant for the S_M magnetic system; $k_S(x)$ determines the connection of x with $\langle S_M^z\rangle$ ($\langle S_M^z\rangle=-k_S(x)x/2$). Since the spin splittings of the electrons and holes greatly exceed the electron-hole exchange interaction under GSS conditions, it is necessary to take into account in (2) both the free carriers and those bound into excitons.

Equation (2) was obtained in Ref. 10 for the case of a simple valence band at $S_h=\frac{1}{2}$. Recognizing that in CdTe:Mn the valence band is characterized at the Γ point of the Brillouin zone by an effective spin $S_h=\frac{3}{2}$, as well as that only transitions to the upper spin sublevel of the valence band with $S_h^z=-\frac{3}{2}$ are considered in the experiment, for comparison with experiment it is necessary to define z as equal to

$$\Big(n_{-3/2}-\sum_{p\neq-3/2}n_p\Big)\Big/\sum_p n_p.$$

Total absence of spin polarization of the valence-band electrons corresponds then to $z=0.5$.

In Eq. (2), $\bar{y}$ and $\bar{z}$ are the carrier spin polarizations that would be obtained in the absence of exchange scattering on account of the aggregated of the remaining spin-relaxation mechanisms:

$$\bar{y}=\frac{y_0\tau_{eL}^{-1}+y^*\tau_r^{-1}}{\tau_{eL}^{-1}+\tau_r^{-1}},\quad \bar{z}=\frac{z_0\tau_{hL}^{-1}+z^*\tau_r^{-1}}{\tau_{hL}^{-1}+\tau_r^{-1}}. \tag{3}$$

Here y_0 and z_0 are the equilibrium values of y and z at a given GSS, while y^* and z^* are the spin polarizations of the electrons and holes at the instant of their production. When account is taken of the values and signs of the GSS for e and h, we can replace y_0 and z_0 with sufficient accuracy by 1 and −1, corresponding to total thermal polarization of the spins at GSS values $|G^{e(h)}|\gg k_BT_0$. At the experimentally realizable unpolarized optical pumping $y^*=0$ and $z^*\approx 0.5$.

The observable deviations of $\langle S_M^z\rangle$ from the equilibrium value (of x from x_0) are much less than $S_M=\frac{5}{2}$. Therefore, according to Ref. 10, to analyze the experimental results we can substitute in Eq. (2) the values of $y(x)$ and $z(x)$ calculated at $x\to x_0$ and in the approximation in which the contributions of the electrons and holes are additive.

The expression for $y(x_0)$ [and the analogous one for $z(x_0)$] takes according to Ref. 10 the form

$$y(x_0)=\frac{x_0+\text{th}(G^e/2k_BT_h)+\bar{y}(\tau_{eL}^{-1}+\tau_r^{-1})/k_S(x_0)\tau_{eM}^{-1}}{1+x_0\,\text{th}(G^e/2k_BT_h)+(\tau_{eL}^{-1}+\tau_r^{-1})/k_S(x_0)\tau_{eM}^{-1}}. \tag{4}$$

It is seen that $y(x_0)[z(x_0)]$ depends on the ratio of the rate of exchange relaxation τ_{eM}^{-1} of the electron spins on the localized moments to τ_{eL}^{-1} and τ_r^{-1}.

It follows from Ref. 10 that

$$\begin{gathered}\tau_{eM}^{-1}=n_MU[F(G^e/k_BT_h)+F(-G^e/k_BT_h)]/2;\\ U=m^2J_e^2\bar{v}/\pi\hbar^4,\qquad \bar{v}=(8k_BT_h/\pi m)^{1/2},\\ F(x)\approx\begin{cases}1, & |x|<1;\\ \tfrac{1}{2}(\pi|x|)^{1/2}\exp[-(x+|x|)/2], & |x|>1.\end{cases}\end{gathered} \tag{5}$$

Whence, using the estimates[1-4] of J_e and J_h for CdTe:Mn and substituting the LSM density $n_M\approx 5\times 10^{20}$ cm^{-3}, we obtain at $m\lesssim 0.5m_e$ the values $\tau_{eM}^{-1}\lesssim 10^{12}$ sec^{-1} and $\tau_{hM}^{-1}\gtrsim 6\tau_{eM}^{-1}$. Not one of the known SLR mechanisms leads to such short times τ_{eL} in the absence of GSS at $T_0\approx 2$ K. Assuming that in the case of GSS, too, we have $\tau_{eL}^{-1}\ll\tau_{eM}^{-1}$, as well as $\tau_r^{-1}\ll\tau_{eM}^{-1}$, we can simplify expression (4) for the electron spin polarization y (and the analogous one for z), by leaving out the terms that contain the ratios of τ_{eL}^{-1}, τ_{hL}^{-1}, τ_r^{-1} to τ_{eM}^{-1}, τ_{hM}^{-1}. As a result we have

$$\begin{gathered}y=\text{th}\Big[\frac{1}{2}\Big(\frac{\omega_0}{k_BT_0}+\frac{G^e}{k_BT_h}\Big)\Big],\\ z=-\text{th}\Big[\frac{1}{2}\Big(\frac{|G^h|}{k_BT_h}-\frac{\omega_0}{k_BT_0}\Big)\Big],\end{gathered} \tag{6}$$

where account is taken of the signs of the GSS for the electron and hole bands, $G^e=-2J_en_M\langle S_M^z\rangle>0$ and $G^h=2J_hn_M\langle S_M^z\rangle<0$.

In Ref. 10, T_h is defined as the kinetic temperature of the carriers. In our experiment the energy of the exciting light exceeds considerably the band gap, so that the distribution function is determined by the particle flux in energy space with emission of LO and acoustic phonons. In this situation the concept of a kinetic temperature of the carriers can hardly be correct. We must therefore regard T_h hereafter as a parameter chosen to obtain the best fit to the true carrier energy distribution function to a Boltzmann distribution.

Substituting (3) and (6) in (2), we obtain ultimately

$$x-x_0=\frac{I}{k_sN\tau_{ML}^{-1}}\left\{0.5+\frac{2(1+\tau_{eL}^{-1}/\tau_r^{-1})}{1+\mathcal{E}_1}-\frac{2(1+\tau_{hL}^{-1}/\tau_r^{-1})}{1+\mathcal{E}_2}\right\},$$
$$\mathcal{E}_1=\exp\left(\frac{\eta|G^h|}{k_BT_h}+\frac{\omega_0}{k_BT_0}\right),$$
$$\mathcal{E}_2=\exp\left(\frac{|G^h|}{k_BT_h}-\frac{\omega_0}{k_BT_0}\right). \quad (7)$$

Here $I=n_{e(h)}\tau_r^{-1}$ is the pump intensity, and $\eta\equiv G^e/G^h=-0.75$ for CdTe:Mn (Ref. 4).

To compare (7) with experiment we must know the dependences of τ_{eL} and τ_{hL} (of y_0 and z_0) on the value of the GSS (in this case τ_r^{-1} and T_h can be regarded as independent of the GSS, and τ_{ML} as independent of H). The dependences of τ_{eL} and τ_{hL} on the spin splittings have not been investigated in the literature for the known SLR mechanisms. In additions, under GSS conditions there can appear new relaxation mechanisms. It is clear only that the effectiveness of the electron hole SLR should increase with increasing spin splitting. Without loss of generality we can approximate τ_{eL}^{-1} and τ_{hL}^{-1} by power-law functions with a certain unknown exponent α. We represent the time ratio in the form

$$\tau_{eL}^{-1}/\tau_r^{-1}=(G^e/G_0^e)^\alpha, \quad \tau_{hL}^{-1}/\tau_r^{-1}=(G^h/G_0^h)^\alpha, \quad (8)$$

where G_0^e and G_0^h are coefficients with the dimension of energy.

One should expect the SLR for holes to be at least several times more effective than for electrons ($G_0^h < G_0^e$). Even this argument is sufficient to cause (7), with allowance for (8), to lead to an effect that agrees qualitatively with experiment.

It follows from (7) that the holes contained in the free excitons are strongly nonequilibrium in spin at $H<1.0$ T. An analysis of the data of Ref. 7 on the basis of Eq. (4) shows that, to explain this result, T_h should amount to several dozen degrees Kelvin. In this case, at the values G^e, G^h and ω_0 prevailing in experiment, neglecting τ_{eL}^{-1} compared with τ_{hL}^{-1}, we can reduce (7) to the form

$$x-x_0\approx\frac{I}{k_sN\tau_{ML}^{-1}}\left\{0.5-\frac{(G^h/G_0^h)^\alpha}{1+\mathcal{E}_2}\right\}. \quad (9)$$

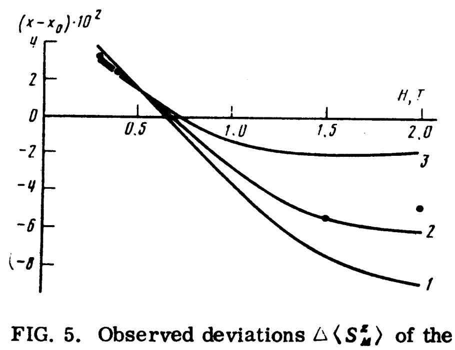

FIG. 5. Observed deviations $\Delta\langle S_M^z\rangle$ of the LSM magnetization from the equilibrium value at a pump 8% of I_{max}; ●—experimental values of $(x-x)$; 1, 2, 3) theoretical curves for $T_h = 51$ K, $i = 0.1$ for various values of the parameter α:1—$\alpha = 2.25$; 2—$\alpha = 2$; 3—$\alpha = 1.75$.

Comparison of (9) with experiment at the optimally chosen parameters T_h, α, G_0^h and $i=I/k_sN\tau_{ML}^{-1}$ is shown in Fig. 5, where the magnetic-field dependence of $x-x_0$, obtained in experiment[1)] at $I\approx 8\%$ of I_{max} are compared with the field dependences of $x-x_0$ calculated from (9) for several sets of parameters. The obtained value $i\approx 0.1$ agrees satisfactorily with experiment under the assumption $\tau_{ML}\sim 10^{-3}-10^{-4}$ sec and recognizing that the light is absorbed over a thickness $10^{-4}-10^{-5}$ cm. The values $\alpha\approx 2$ and $T_h\approx 51$ K seem reasonable. The linear dependence of $x-x_0$ on the pump density agrees with (7) and (9) in the weak-pump region.

The result can be qualitatively understood in the following manner. In the region of relatively low values of the GSS, the recombination rate predominates compared with the SLR τ_{eL} and τ_{hL}, so that the polarizations of the carriers y and z do not manage to deviate noticeably from y^* and z^* during the lifetime τ_r. Since $z^*>y^*$, the contribution of the holes to the exchange scattering predominates and determines the positive sign of the change (the enhancement) of the polarization of the localized moments. With increasing H (and correspondingly G^e and $|G^h|$) the role of the SLR increases and in view of the faster growth of their relaxation, the holes manage to become thermalized, causing a weakening of their contribution to the exchange scattering and even a change in the sign of the $x-x_0$ because of the electron contribution. The arguments presented show that exchange scattering of carriers by Mn in CdTe is indeed responsible for the observed change of the polarization $\langle S_M^z\rangle$ in the region of not too intense pumping $I<15\%\ I_{max}$.

With further increase of the pump density the crystal seems to become locally heated, so that τ_{ML}, τ_{eL} and τ_{hL} become shorter and the influence of the exchange scattering as $x-x_0$ is therefore decreased and eventually vanishes. At the same time, the rise in the lattice temperature leads to a decrease of x_0, as is indeed observed in experiment.

We note in conclusion that the possibility, considered in Ref. 10, of a strong change of $\langle S_M^z\rangle$ due to the exchange mechanism of the nonequilibrium carrier relaxation turns out to be limited, since the effect is determined in final analysis by the ratio of the optically produced carriers during the SLR time of the spins S_M to the number of localized spins in the irradiated volume. The heating of the crystal by the excited seems to start before this ratio reaches unity. One can expect, however, the relative value of the effect to be larger if the density of the localized magnetic moments and the energy of the exciting light are optimally chosen.

1) The quantity $x-x_0$ was defined as the ratio of the change of the energy of the σ^+ exciton luminescence line under the influence of the pump to the obtained value of ΔE_{max}, where ΔE_{max} was obtained by approximating the magnetic-field dependence of the position of the σ^+ component of the luminescence by the function $E=E(0)-\Delta E_{max}B_{5/2}(g\beta H/k_BT_{eff})$.

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