Optical manifestation of spin-glass properties of semimagnetic semiconductors

B. P. Zakharchenya and Yu. G. Kusraev
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR

(Submitted 24 July 1989)
Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 4, 199–201 (25 August 1989)

The spectral dependence of circularly polarized luminescence of a semimagnetic semiconductor in the spin-glass region was studied. The frozen-in field, produced by cooling the sample in an external magnetic field, was optically recorded from the circular polarization of the luminescence.

Spin glasses are disordered magnetic materials in which the exchange-interaction energy varies randomly. Below a certain temperature T_f, which can be determined experimentally reasonably well, the magnetic moments are frozen in random directions. The curve of the magnetic susceptibility versus the temperature in this case exhibits a maximum at the temperature T_f. A distinctive feature of spin glasses is the dependence of the magnetic susceptibility of a sample on its history: Below T_f the susceptibility of a sample cooled in a magnetic field differs from the susceptibility of a sample cooled in the absence of a field. In the first case the sample retains a residual magnetization which decays to zero very slowly after the field is removed.[1] Similar properties were also observed in semimagnetic semiconductors (primarily in the solid solutions $A^2_{1-x}Mn_xTe$, in which the Mn atom, the substitutional impurity atom, is a magnetic atom).[2,3]

In the present letter we report the observation of optical magnetization produced by a circularly polarized light and we also report an optical recording of residual magnetic fields in spin glasses $Cd_{1-x}MN_xTe$. The optical magnetization of semimagnetic semiconductors was studied previously in Refs. 4–6. In particular, optical orientation in semimagnetic semiconductors was achieved for the first time in Ref. 6.

The experiment was carried out using $Cd_{1-x}Mn_xTe$ single crystals with $x = 0.25$ and 0.40 at liquid-helium temperatures. The luminescence was excited by a tunable Rhodamin-6G dye laser which was pumped by an argon laser. In the luminescence spectrum of samples with $x > 0.1$ the dominant line is the one attributable to the recombination of excitons trapped at the fluctuations of the composition and magnetization.[7] Figure 1 shows the spectra of the intensity I (curve 1) and of the degree of circular polarization ρ (curve 2) of the luminescence of $Cd_{0.6}Mn_{0.4}Te$, which is excited by circularly polarized photons with energy $\hbar\omega < E_0$, where E_0 is the energy approximately equal to the energy of a free exciton. A decrease in $\hbar\omega$ to an energy E_0 has no effect on the luminescence spectrum. This spectrum is represented by a dashed line in Fig. 1. Upon further decrease in $\hbar\omega$, however, the luminescence line shifts toward the long-wave side of the scale and attenuates. Consequently, the energy spacing $\Delta\hbar\omega = \hbar\omega - \hbar\omega_{max}$, where $\hbar\omega_{max}$ is the maximum of the luminescence line, remains constant, amounting to $\Delta\hbar\omega \approx 24.3$ meV. This shift is caused, in our view, by the magnetic trapping of an exciton and by the formation of a magnetic polaron. This

 0021-3640/89/160225-04$01.00

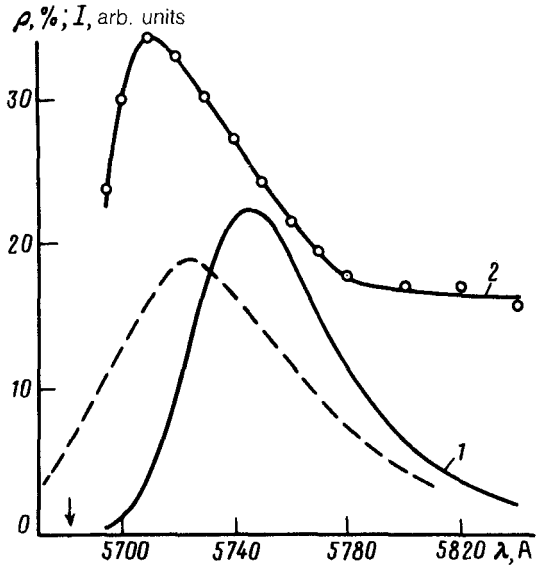

FIG. 1. The plot of $I(\lambda)$ (curve 1) and of $\rho(\lambda)$ (curve 2). Excitation—$\lambda = 5687$ Å ($\hbar\omega = 2.180$ eV $< E_0$). Dashed curve—Excitation—$\lambda = 5145$ Å ($\hbar\omega = 2.410$ eV $> E_0$); $T \approx 2$ K.

explanation is confirmed by the appreciable decrease in the energy of the shift, $\Delta\hbar\omega$, as the temperature is raised (see the discussion below). It would be reasonable to link the energy E_0 with the mobility threshold of excitons. In the case of excitation $\hbar\omega > E_0$, the luminescence is not polarized since the spin relaxation rate of the carriers and excitons is high because of the exchange scattering by magnetic Mn^{2+} ions. In contrast, the spin relaxation of the trapped excitons is hindered by the strong internal exchange fields.[6] This situation occurs in a circularly polarized luminescence of the trapped excitons (curve 2 in Fig. 1). A complex nonmonotonic behavior of the spectrum of

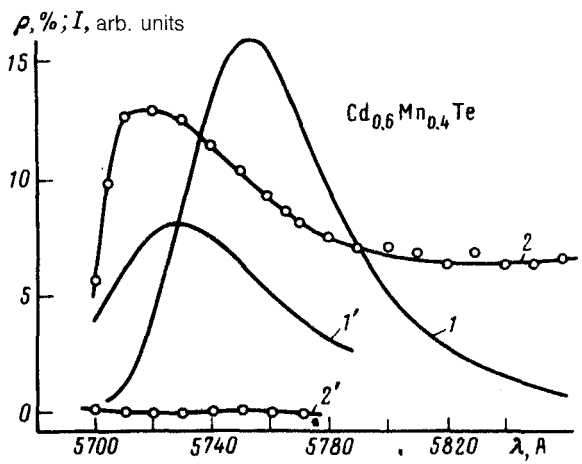

FIG. 2. The plots of $I(\lambda)$ (curves 1 and 1') and of $\rho(\lambda)$ (curves 2 and 2'); 1,2—$T \approx 2$ K; 1',2'—$T \approx 15$ K. Excitation—$\lambda = 5682$ Å ($\hbar\omega = 2.182$ eV).

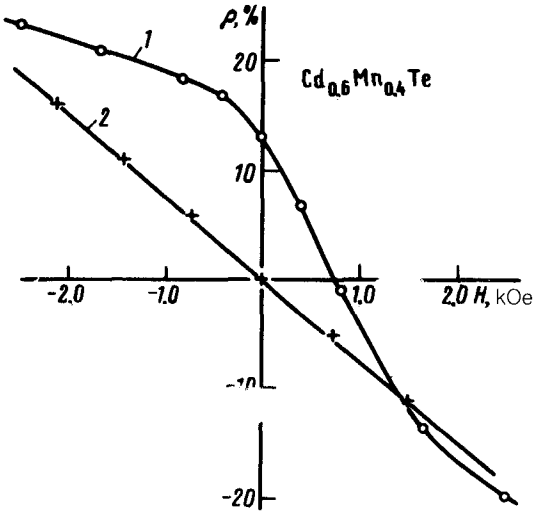

FIG. 3. Curves of $\rho(H)$ at $T \approx 2$ K. 1—The sample was cooled in a field $H \approx 2.5$ kOe; 2—the sample was cooled in zero field.

circular polarization $\rho(\lambda)$ mirrors the dynamics of the formation of a magnetic polaron (this problem will be discussed in greater detail in a separate paper).

Similar results were obtained with $Cd_{1-x}Mn_xTe$ samples with $x = 0.25$.

As subsequent experiments have shown, the polarization characteristics of luminescence depend on the conditions under which a crystal is cooled. Upon cooling a sample in a longitudinal (with respect to the direction of observation of the luminescence) magnetic field, for example, before the temperature $T < T_f$ is reached, the luminescence was found to be circularly polarized, even in the case of excitation by an unpolarized light in the absence of a magnetic field. Figure 2 shows the spectra of the intensity (I) and of the degree of circular polarization of the luminescence of the $Cd_{0.6}Mn_{0.4}Te$ crystal for two temperatures: $T_1 \approx 2$ K (curves 1 and 2) and $T_2 \approx 15$ K (curves 1′ and 2′); here $T_1 < T_f$ and $T_2 > T_f$; $T_f \approx 11$ K (Ref. 7). As can be seen from the figure, at $T > T_f$ light is not polarized. Furthermore, an increase in the temperature causes the luminescence line to shift toward shorter wavelengths, suggesting that the polaron energy decreases. The degree of circular polarization of the luminescence did not change appreciably at any time during the experiment (which lasted several hours) when the temperature was held at $T \approx 2$ K. The circularly polarized luminescence in the spin-glass region ($T < T_f$) can be attributed to the fact that the Mn^{2+} ion spins, which are polarized by the external magnetic field, become frozen when they pass through the freezing point. This process leads to the formation of local internal fields H_L, which affect the trapped exciton states. As the temperature is raised to $T > T_f$, these fields collapse, causing the polarization to vanish.

The local internal field can also be seen on the plot of the degree of circular polarization of the luminescence versus the external magnetic field when the excitation is unpolarized. Figure 3 shows this curve (curve 1), along with the $\rho(H)$ curve for the case in which the sample is cooled in zero magnetic field (curve 2). In the first case the $\rho(H)$ curve is nonlinear in nature and vanishes in a ≈ 0.8-kOe field. This means

that the external magnetic field, $H \approx 0.8$ kOe, cancels the effect produced by the frozen-in field H_L. This situation can be exploited to estimate H_L. To correctly measure the field H_L, however, it is necessary to independently measure the circular absorption dichroism.

We wish to thank V. F. Agekyan, I. A. Merkulov, Fan Zung, and K. V. Kavokin for useful discussions.

[1]M. Mezard *et al.*, *Spin Glass Theory and Beyond.* Singapore: World Scientific, 1988, p. 476 (World Scientific Lecture Notes in Physics, 9).
[2]M. A. Novak *et al.*, Phys. Rev. B **33**, 6391 (1986).
[3]M. Saint-Paul *et al.*, Solid State Commun. **60**, 621 (1986).
[4]H. Krenn *et al.*, Phys. Rev. Lett. **55**, 1510 (1985).
[5]D. D. Auschalom *et al.*, Phys. Rev. Lett. **58**, 812 (1987).
[6]J. Warnock *et al.*, Solid State Commun. **54**, 215 (1985).
[7]A. Golnic *et al.*, J. Phys. C **16**, 6073 (1983).
[8]M. A. Novak *et al.*, J. Appl. Phys. **57**, 3418 (1985).

Translated by S. J. Amoretty