Optically Induced Magnetization in a Dilute Magnetic Semiconductor: $Hg_{1-x}Mn_xTe$

H. Krenn, W. Zawadzki,[a] and G. Bauer
Institut für Physik, Montanuniversität Leoben, A-8700 Leoben, Austria
(Received 19 February 1985)

This paper reports the observation of light-induced magnetization in $Hg_{1-x}Mn_xTe$ by a novel technique combining optical pumping and superconducting quantum interference detection. We show that the magnetization is due to orientation of Mn ions caused by spin transfer from polarized electrons and holes to Mn centers in the process of spin-flip exchange scattering. This interpretation goes beyond a mean-field approach. The effect furnishes direct information on the spin-lattice relaxation in dilute magnetic semiconductors.

PACS numbers: 78.90.+t, 71.70.Gm, 75.30.Et, 76.30.Fc

Dilute magnetic semiconductors (DMS) have become in recent years a subject of intensive experimental and theoretical investigations.[1] The exchange interaction between free carriers and paramagnetic ions which is responsible for most of the interesting features of DMS has usually been treated in the mean-field approach, in which the mobile carrier experiences an averaged effective field of the localized spin moments. For some phenomena, however, this approach has proved insufficient. Thus the recent theory of magnetic polarons in DMS has involved thermodynamic fluctuations of the spin field.[2,3] Other investigations have emphasized spin-flip scattering effects. Ryabchenko, Semenov, and Terletskii[4] have observed a change of the magnetization in $Cd_{0.95}Mn_{0.05}Te$ upon illumination of the sample with unpolarized laser light in the presence of an external magnetic field. The latest optical spin-pumping experiments[5,6] indicate that this spin exchange through the exchange interaction plays an important role in the spin relaxation of photocreated carriers.

In this Letter we report an optical pumping experiment with circularly polarized radiation in which a light-induced magnetization of $Hg_{1-x}Mn_xTe$ has been directly observed. We analyze the effect and show its connection with the spin-exchange and the spin-lattice-relaxation processes.

In order to detect an optically induced magnetization the sample was placed inside the flux transformer of a radio-frequency superconducting quantum interference device (rf SQUID),[7] which was encapsulated by a superconducting Nb shield in order to screen out all external magnetic fields. Optical interband transitions were excited by circularly polarized CO-laser radiation ($h\nu = 205$–240 meV; powers of 20–100 mW) with use of a photoelastic modulator (CdS) operating at 37 kHz. The experiments were carried out at a constant laser power impinging onto the sample, with modulation of the degree of polarization from linearly to circularly polarized light. Thus, parasitic effects originating from thermally induced excess currents, unavoidable when a chopped laser beam is used, were not present. The magnetization of the sample was detected by a pickup coil (inductance 2 μH) matched to the input impedance of the SQUID signal coil. To increase the magnetic coupling between the pickup coil and the SQUID a small coil with thirteen windings and a diameter of 2 mm was used, thus limiting the sample size. By use of the superconducting shield (with a pinhole of 2-mm diameter for the exciting laser radiation) no gradiometer configuration was necessary. Thereby a high efficiency for the coupling of the optically induced magnetic flux to the rf SQUID has been achieved. The coupling factor C was measured by replacing the sample by a coil with a diameter equivalent to the sample cross section. The obtained value of $C = \Phi_{\text{detected}}/\Phi_{\text{anticipated}}$ was 0.05. The magnetic-flux noise limit of the whole system is $(2\times10^{-4}\ \text{Hz}^{-1/2})\Phi_0$, Φ_0 denoting the flux quantum.

The entire system is placed in a variable-temperature cryostat. Despite the fact that the SQUID and the sample are in the same tail section of the cryostat, the temperature of the sample can be varied in the range of 4.2–15 K, by the laser illumination and by cooling with exchange helium gas, whereas the SQUID sensor remains at temperatures between 4.2 and 5 K.

The experiments were performed with several p-type samples of the DMS $Hg_{1-x}Mn_xTe$ ($x = 0.07$–0.12). For comparison, $Hg_{1-x}Cd_xTe$ ($x = 0.23$) and InSb having the same open-gap band structure were also investigated. Figure 1 shows the observed magnetization signal (in units of Φ_0) versus the degree of light polarization. For circular polarization, the $Hg_{1-x}Mn_xTe$ open-gap sample shows a net optically induced magnetic flux, whereas for linearly polarized light no net flux is observed. The nonsemimagnetic semiconductors InSb and $Hg_{1-x}Cd_xTe$ do not yield any signal within the limits of the noise. The measured magnetic flux in $Hg_{1-x}Mn_xTe$ increased linearly with the laser power in the range of 0.2–1.25 W/cm^2.

The temperature dependence of the optically induced magnetization signal was measured for several open-gap $Hg_{1-x}Mn_xTe$ samples and it is shown for $x = 0.012$ in Fig. 2.

For the interband transitions between Γ_8 (valence) and Γ_6 (conduction) bands, one obtains a polarization

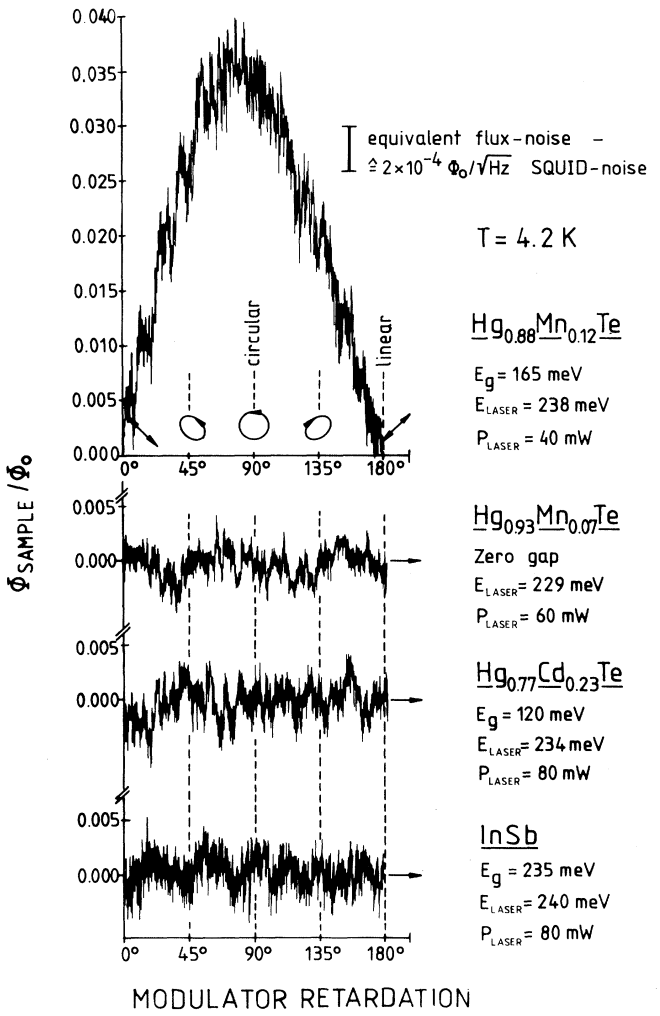

FIG. 1. Magnetic flux in the pickup coil proportional to the magnetization of various semiconductor samples vs the degree of circular polarization of the CO-laser radiation. Only in the open-gap diluted magnetic semiconductor $Hg_{0.88}Mn_{0.12}Te$ is the photomagnetization observed.

ratio of 3:1.[9] For the excitation between Γ_8 (valence) and Γ_8 (conduction) bands (zero-gap situation) the corresponding ratio is almost 1:1.[10] The nonexistence of observable magnetization in InSb and $Hg_{1-x}Cd_xTe$ indicates that we do not measure the magnetization of the spin-polarized electrons or holes. On the other hand, the applied laser radiation cannot directly polarize the Mn ions, since the photon energies are too high. This is confirmed additionally by the absence of magnetization in the zero-gap $Hg_{1-x}Mn_xTe$ sample.

It follows then that the detected magnetization in

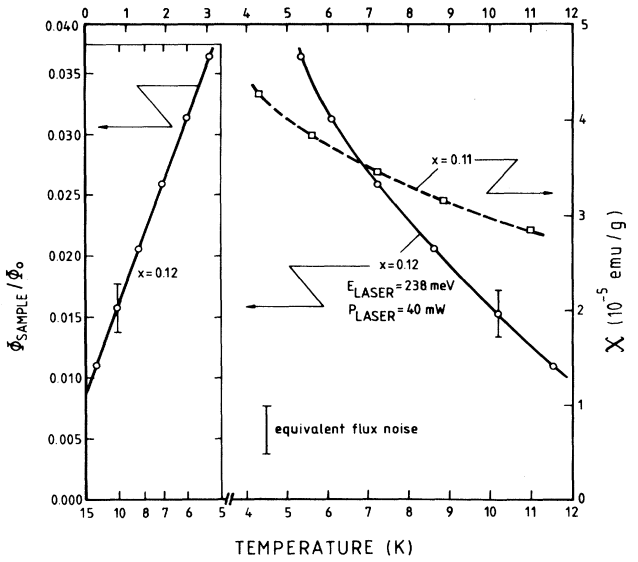

FIG. 2. Photomagnetization in an open-gap $Hg_{1-x}Mn_xTe$ sample vs temperature. For comparison, the low-field magnetic suscpetibility (Ref. 8) of $Hg_{0.89}Mn_{0.11}Te$ with similar chemical composition is shown. The photomagnetization effect is inversely proportional to the temperature (see inset).

open-gap $Hg_{1-x}Mn_xTe$ is due to an orientation of the paramagnetic Mn ions by the polarized mobile carriers. The electron-ion interaction is described by the exchange Hamiltonian[11]

$$H = \sum_i J(r - R_i)(S_+ s_- + S_- s_+ + S_z s_z) \qquad (1)$$

in the standard notation. In the mean-field approach to DMS, one takes into account only the $S_z s_z$ term, calculating a macroscopic average magnetization $\langle S_z \rangle$ in an external magnetic field, and then considers its influence on the free-carrier properties. Within the same framework one could imagine that the observed photomagnetization is due to an inverse mechanism: The stream of free carriers with their spins polarized in the direction of light propagation creates an effective magnetic field, causing a corresponding partial orientation of the Mn ions. However, it is known that in $Hg_{1-x}Mn_xTe$ the induced internal field due to Mn ions is comparable to the external magnetic field.[11] In consequence, such a magnetization effect would be comparable in magnitude to the initial free-carrier magnetization, i.e., not measureable in our experiments.

Thus, we are led to the conclusion that the orientation of Mn ions caused by the spin-polarized free carriers is due to the nondiagonal terms in the exchange Hamiltonian, Eq. (1). The nonvanishing matrix elements of this interaction are

$$\langle M, m+1 | S_- s_+ | M+1, m \rangle = \langle M+1, m | S_+ s_- | M, m+1 \rangle$$
$$= \tfrac{1}{2}[(J-M)(J+M-1)]^{-1/2}[(j-m)(j+m+1)]^{1/2}, \qquad (2)$$

where $J=\frac{5}{2}$ is the total spin value of the Mn^{2+} ion, j is that of the free carriers [$j=\frac{1}{2}$ for electrons (Γ_6 conduction band) and $j=\frac{3}{2}$ for holes (Γ_8 valence band)], and M and m are their quantized projections on the direction of the light propagation, respectively. It is seen from Eq. (2) that in the exchange-interaction processes the sum of projections $M+m$ is conserved, so that the free carriers will partially polarize the Mn ions in the course of spin-flip scattering. The matrix elements of the Hamiltonian involve also the exchange integrals[11] $\langle S|J|S\rangle=\alpha$ or $\langle P|J|P\rangle=\beta$ for the conduction and the valence bands, respectively. The contributions of electrons and holes to the Mn orientation are additive: For example, a hole with $s=-\frac{1}{2}$ and an electron with $s=+\frac{1}{2}$, created by a circularly polarized photon, polarize the Mn ions in the same direction. The electron-ion and the hole-ion processes are independent.

To understand the observed magnetization we have to solve the rate equations for the numbers of polarized free carriers and Mn ions. The equation for electrons reads

$$\frac{\partial(n_+-n_-)}{\partial t}=G_+-G_--\frac{n_+-n_-}{\tau}-\frac{n_+-n_-}{T_{\mathrm{eph}}}-\frac{n_+-n_-}{T_{\mathrm{eMn}}}, \tag{3}$$

where n_+,n_- are the numbers of spin-up and spin-down electrons, G_+,G_- are their creation rates; τ, T_{eph}, and T_{eMn} are recombination, electron-phonon relaxation, and electron-Mn relaxation times, respectively. The stationary solution is

$$n_+-n_-=\frac{G_+-G_-}{1/\tau+1/T_{\mathrm{eph}}+1/T_{\mathrm{eMn}}}. \tag{4}$$

If we assume that $T_{\mathrm{eMn}}\ll\tau,T_{\mathrm{eph}}$ (which are of the order of 10^{-9} s, cf. Bichard, Guldner, and Lavallard[12]), then

$$n_+-n_-\cong(G_+-G_-)T_{\mathrm{eMn}}, \tag{5}$$

which is a small number for short times T_{eMn}. The electron spin-flip rate due to the exchange interaction with Mn ions can be estimated as[13]

$$1/T_{\mathrm{eMn}}\approx(N_M/\pi\hbar^3)(\alpha\Omega)^2m^*k,$$

where k is the electron wave vector [in our experiments $\epsilon(k)\approx E_g$] and Ω is the volume of unit cell. Setting $\alpha\Omega=0.4\times10^{-22}$ eV cm^3 and using other appropriate parameters we get $T_{\mathrm{eMn}}\approx10^{-12}$ s, so that really $T_{\mathrm{eMn}}\ll\tau,T_{\mathrm{eph}}$.

The oriented Mn ions can relax their spin through the interaction with (i) each other, (ii) free carriers, and (iii) acoustic phonons. The first interaction, although important (see Oseroff[14] and Kremer and Furdyna[15]) conserves the spin in the Mn system. The second mechanism is of no importance since the number of Mn ions is much larger than that of the free carriers. Thus the rate equation for the ions reads

$$\frac{\partial(N_+-N_-)}{\partial t}=\frac{n_+-n_-}{T_{\mathrm{eMn}}}-\frac{N_+-N_-}{T_{\mathrm{Mnph}}}, \tag{6}$$

where T_{Mnph} characterizes the rate of Mn-spin relaxation due to phonons. In the stationary state

$$N_+-N_-=(G_+-G_-)\frac{T_{\mathrm{Mnph}}}{T_{\mathrm{eMn}}}\frac{1}{1/\tau+1/T_{\mathrm{eph}}+1/T_{\mathrm{eMn}}}. \tag{7}$$

If $T_{\mathrm{eMn}}\ll\tau,T_{\mathrm{eph}}$ we obtain

$$N_+-N_-\cong(G_+-G_-)T_{\mathrm{Mnph}}. \tag{8}$$

Comparing Eqs. (5) and (8) we see that

$$N_+-N_-\cong(n_+-n_-)T_{\mathrm{Mnph}}/T_{\mathrm{eMn}}.$$

This explains the possibility of observing the magnetization of Mn ions: It is much larger than that of the spin-polarized free carriers if $T_{\mathrm{Mnph}}\gg T_{\mathrm{eMn}}$. The creation rate of polarized Mn ions is almost equal to the creation rate of spin-polarized electron-hole pairs, but the ions "memorize" their spin orientation as a result of the slow spin relaxation with phonons. The above derivation is valid provided that the resulting number N_+-N_- is much smaller than the total number of Mn ions N_{Mn}. If this is not the case, the first term in Eq. (6) should include the restriction expressed in Eq. (2) that the spin-oriented electrons may polarize only unpolarized ions.

The observed temperature dependence of the photomagnetization shown in Fig. 2 is determined mostly by the temperature dependence of the Mn-spin–phonon relaxation, since the Mn–free-carrier spin exchange rate does not vary much with the temperature. Spin relaxation of Mn ions due to phonons may occur because of the spin-orbit interaction. For Mn^{++} this interaction is very weak since the five outer electrons are in the S orbital state. This is further confirmed by the value of the Landé factor, $g\cong2$ (see Leibler *et al.*[8]). Thus one expects long spin-relaxation times. At low temperatures the relaxation is usually governed by one-phonon direct processes. Such processes lead to a $1/T$ dependence of the T_{Mnph}. It can be seen from the inset in Fig. 2 that the photomagnetization has, in fact, the $1/T$ dependence. For rare-earth paramagnetic ions the calculated and measured spin-relaxation times are in the range of 10^{-2}–10^{-3} s.[16] For the iron-group ions they should be somewhat shorter and we estimate $T_{\mathrm{Mnph}}\cong10^{-4}$ s. In our experiments $G_+-G_-\cong10^{21}$ s^{-1} cm^{-3}, which gives for the above relaxation times

T_{eMn} and T_{Mnph} $n_+ - n_- \cong 10^9$ cm^{-3} and $N_+ - N_- \cong 10^{17}$ cm^{-3} (as compared to $N_{Mn} \cong 10^{22}$ cm^{-3}). Thus in equilibrium one ion in 10^5 is polarized as a result of the spin-flip scattering. The maximum measured magnetization of the sample was $M \cong 2\times 10^{-4}$ A/m. Since this magnetization comes only from the polarized ions, we have $M = g\mu_B(N_+ - N_-)S_z$, from which we calculate $S_z \cong 10^{-4}$ for the polarized ion. This gives the local Zeeman splitting of conduction electrons as $\Delta E = \alpha S_z \cong 5\times 10^{-2}$ meV, so that at our temperatures the $S_z s_z$ term is really negligible. The different temperature variation of the photomagnetization effect from that of the measured $\langle S_z \rangle$ magnetization[17] in Fig. 2 confirms that this effect goes beyond the framework of the mean-field picture.

We have not included in Eqs. (3)–(5) the depolarization effects due to electron-hole collisions.[18] For open-gap p-$Hg_{1-x}Mn_xTe$ ($x \approx 0.12$) and $N_A - N_D \approx 10^{16}$ cm^{-3} the free-hole concentration[19] due to thermal activation of $T \approx 4$ K is negligibly small compared to the light-induced excess hole concentration which is equal to the excess electron density and of the order of 10^{14} cm^{-3} for our experimental conditions.

The initial polarization ratio of 3:1 for excitation close to $k \approx 0$ is reduced because of band-mixing effects for our experimental conditions, $h\nu (= 200 - 240$ meV$) > E_g (\approx 165$ meV). Also the spin-relaxation time T_{eph} obviously changes with the excitation energy. However, in our case these changes do not alter the relevant relaxation times in Eqs. (1)–(8) to such an extent as to violate at all the essential inequalities and thus our conclusions.

To summarize, we argue that the observed photomagnetization effect in $Hg_{1-x}Mn_xTe$ is due to the orientation of Mn ion magnetic moments by the spin-flip scattering of spin-polarized free carriers via the nondiagonal part of the exchange interaction. The effect is directly related to the Mn spin-lattice relaxation, which brings new physical information since the spin resonance of the Mn ions in $Hg_{1-x}Mn_xTe$ is dominated by the Mn-Mn interactions.[14,15]

We thank R. R. Galazka for growing the $Hg_{1-x}Mn_xTe$ samples and B. Kaufmann for his help in the initial stages of this work. Elucidating discussions with J. Kossut, T. Dietl, and G. Lampel are gratefully acknowledged. This work was supported by the Fonds zur Förderung der wissenschaftlichen Forschung, Vienna, Austria.

Note added.—The optical polarization of nuclei in ^{29}Si has been recently detected by use of a superconducting magnetic flux meter.[20]

[a]On leave from the Institute of Physics, Polish Academy of Sciences, Warsaw, Poland.

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