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APPLICATION OF PHOTOMEMORY EFFECT IN $Cd_{1-x}Mn_xTe_{1-y}Se_y$:In FOR DIRECT MEASUREMENTS OF MAGNETIZATION OF BOUND MAGNETIC POLARONS*

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The first direct measurement of the magnetization of donor bound magnetic polarons in diluted magnetic semiconductors is reported. The experiment has been performed taking advantage of photomemory effect found in n-type $Cd_{1-x}Mn_xTe_{1-y}Se_y$ crystals doped with In. Good agreement between experimental results and theory of bound magnetic polarons is observed.

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Studies of diluted magnetic semiconductors (DMS) [1] with large, direct band gaps and simple, well-known band structures have contributed significantly to current understanding of bound magnetic polarons (BMP) — ferromagnetic complexes formed by carriers localized at impurities [2].

However, the picture of BMP is not yet complete and there is still much to be done in both experiment and theory. In particular, not all of the thermodynamic properties of BMP have been experimentally studied to date. In this work we present the first direct measurements of magnetization of BMP.

The contribution of the magnetization of BMP to the total magnetization M of a paramagnetic DMS sample is small, and therefore extremely difficult to measure. Recent discovery of photomemory in In- and Ga-doped $Cd_{1-x}Mn_xTe_{1-y}Se_y$ [3, 4] has helped us to overcome this difficulty. It was found that illumination of the sample at a low temperature persistently increases (by orders of magnitude)the concentration N_D of occupied shallow-donors. The BMP formed on these

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additional (persistent) donors should produce a measurable contribution to the sample magnetization, easy to extract since the host magnetization is unchanged. We have indeed observed such a persistent change of M for the first time with the use of radio-frequency superconducting quantum interference device (SQUID) magnetometer.

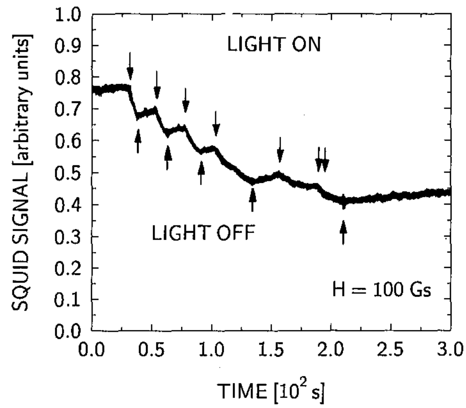

Fig. 1. SQUID signal vs. white light illumination time for In-doped $Cd_{0.9}Mn_{0.1}Te_{0.97}Se_{0.03}$. Data were taken at $T = 4.2$ K in a magnetic field of 100 Gs. The times of turning the light on and off are marked with ↓ and ↑, respectively. Decrease in the signal corresponds to the increase in the magnetization of the sample.

In our experiment the sample was cooled in dark very slowly down to helium temperature and placed inside the flux transformer of a SQUID. Then the sample was illuminated with white light and the SQUID signal for some magnetic field was recorded as a function of illumination time, as presented in Fig. 1. In this figure the decrease in the signal corresponds to the increase in the total sample magnetization M. As can be clearly seen, M starts to increase after turning the illumination on (the times marked with ↓) and stops after turning it off (the times marked with ↑). Between these two events the magnetization does not change due to the constant concentration of persistent shallow-donors. This procedure could be repeated several times, as presented in Fig. 1, until finally, after some longer total time of illumination, M saturates. From that moment on further illumination does not produce any measurable change of M. Magnetization was constant no matter whether the light was on or off. Total increase in the magnetization as high as 0.4% was observed at 4.2 K. This persistent change of magnetization ΔM was also in further experiments unambiguously correlated with the increase in N_D by simultaneous measurements of the resistance and magnetization for samples placed inside the flux transformer of a SQUID.

Observed in these experiments persistent change of sample magnetization ΔM is a straightforward measure of magnetization of BMP (M_P) formed on persistent donors ($\Delta M = M_P$ if the negative-U model of DX centers (Ref. [5]) is correct in DMS, because DX^- center has a diamagnetic ground state, and therefore no polaron can be formed on this center). This BMP magnetization was measured as a function of temperature between 4.2 and 2 K and in magnetic fields up to 200 Gs. Each point of experimental data was obtained by warming the sample up to the room temperature, then cooling it in the dark down to $T = 4.2$ K and finally illuminating with white light. The reproducibility of cooling procedure was obtained by allowing donors to reach the equilibrium state (as manifested by sample resistance) at each intermediate temperature. Some of the measurements results of ΔM for one of our samples are collected in Fig. 2, together with theoretical predictions for M_P. This figure shows polaron magnetization as a function of the magnetic field for two temperatures, $T = 4.2$ K and $T = 2.1$ K.

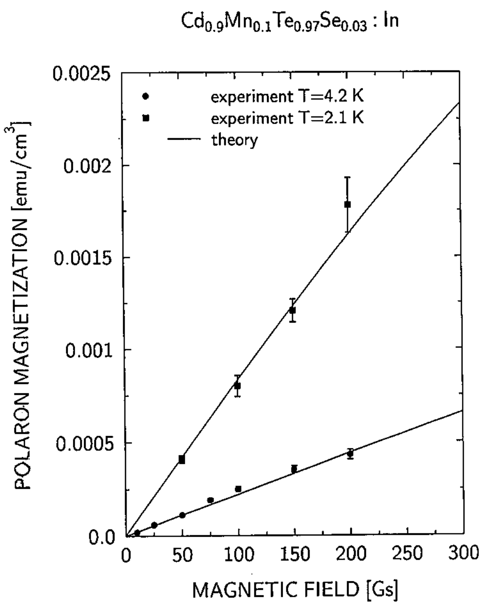

Fig. 2. Experimental (squares and circles) and theoretical (lines) magnetization of BMP as a function of the magnetic field for two temperatures. Theory is calculated using $N_D = 1\times10^{16}$ cm^{-3}, $a_B^* = 40$ Å [8], $N_0\alpha = 220$ meV [8] and the experimentally measured sample magnetic susceptibility.

The theory of BMP in DMS was developed by Dietl and Spałek [6] and independently by Heiman, Wolff and Warnock [7] with the use of a different formalism. The magnetization of BMP can be obtained from Eq. (4.2) of Ref. [6] by taking the partial derivative of the contribution ΔF to the free energy of the system, introduced by the presence of the donor electron which had formed the BMP, over magnetic field. The general formula is quite complicated and describes the gradual saturation of BMP magnetization as magnetic field increases. However, in the range of field used in our experiments M is nearly linear, as can be seen in Fig. 2. The theoretical curves have been obtained using the experimentally measured sample magnetization, described well by the relation $\chi = A/(T + 2.2)$, and other parameters as listed in the caption of Fig. 2. The only fitting parameter used in our calculations was N_D. One can see that theory describes experimental data fairly well if we assume $N_D = 1 \times 10^{16}$ cm^{-3}. This value of N_D is slightly less than the value estimated from Hall data on another sample cut from the same crystal. However, for the concentration of donors close to Mott's critical concentration for the metal-nonmetal transition N_C, considerable donor–donor antiferromagnetic exchange interaction takes place [9]. The concentration of donors required to obtain the observed M_P within the framework of the theory which does not include this effect [6, 7] is lower than the real N_D in the crystal.

In conclusion, we have measured for the first time the magnetization of the donor BMP taking advantage of recently discovered photomemory effect. The experimental results are in good agreement with the theory of BMP in diluted magnetic semiconductors. Further experiments at lower temperatures and for higher magnetic fields could provide a sensitive test of the theoretical models for both BMP and DX centers in DMS.

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References

[1] See, e.g. J.K. Furdyna, *J. Appl. Phys.* **64**, R29 (1988) and references cited therein.

[2] For a recent review, see P.A. Wolff, in: *Semimagnetic Semiconductors and Diluted Magnetic Semiconductors*, Eds. M. Averous, M. Balkanski, Plenum Press, New York 1991, p. 147.

[3] I. Terry, P. Becla, A. Torressen, H. Ohno, S. von Molnar, *Bull. Am. Phys. Soc.* **35**, 601 (1990).

[4] T. Wojtowicz, G.L. Yang, P. Kłosowski, M. Dobrowolska, J.K. Furdyna, I. Miotkowski, *Bull. Am. Phys. Soc.* **35**, 663 (1990).

[5] D.J. Chadi, K.J. Chang, *Phys. Rev. B* **39**, 10366 (1989).

[6] T. Dietl, J. Spałek, *Phys. Rev. B* **28**, 1548 (1983).

[7] D. Heiman, P.A. Wolff, J. Warnock, *Phys. Rev. B* **27**, 4848 (1983).

[8] D.L. Peterson, D.U. Bartholomew, U. Debska, A.K. Ramdas, S. Rodriguez, *Phys. Rev. B* **32**, 323 (1985).

[9] For a recent review, see M.A. Paalanen, R.N. Bhatt, *Physica B* **169**, 223 (1991).