High-field magnetization of (Cd,Mn)Te

D. Heiman, E. D. Isaacs,* P. Becla, and S. Foner*
Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 7 August 1986)

The magnetization of the diluted antiferromagnetic system $Cd_{1-x}Mn_xTe$ with $0.1 \leq x \leq 0.5$ is examined up to fields of 40 T at liquid-helium temperature. The measured magnetization can be separated into the sum of two components: a paramagnetic Brillouin-type part that saturates by $B = 15$ T, and a part linear in B. This separation only becomes evident at fields above 20 T. For increasing x, the ratio of the linear part to the saturating part increases. These characteristics appear to be universal for similar dilute systems, and are a consequence of the spin-spin coupling in small isolated clusters and the random network.

The diluted antiferromagnetic system $Cd_{1-x}Mn_xTe$ exhibits a wide range of magnetic behavior: It shows paramagneticlike behavior at low x, a frustrated spin-glass-like phase, and short-range antiferromagnetism.[1-3] The static and dynamic magnetic properties have only recently been investigated[2] and are not totally understood. Although numerical computations of the magnetic properties have been applied to other random magnetic systems,[4] little effort has been devoted to the present systems.[5] The good optical properties of these semimagnetic semiconductors allow us to use Faraday rotation to probe these frustrated, diluted antiferromagnetic systems to very high fields. It is only at these fields above 20 T where the weakly interacting spins are completely saturated and the residual magnetic response becomes apparent.

In order to understand the effects of the s-d exchange interaction in semimagnetic (diluted-magnetic) semiconductors,[1] the magnetic properties must be known; the magnetization governs the enhanced Zeeman splittings of the semiconductor bands and magnetic polarons.[6] For example, at low temperatures the s-d exchange-enhanced field can be *1000 times larger* than the applied field. In addition, the magnetic response at high fields controls the properties of free and bound magnetic polarons, which can have self-induced internal exchange fields on the order of 100 T.

The low-temperature magnetization $M(B)$ of $Cd_{1-x}Mn_xTe$ for $x \leq 0.3$ was measured by Gaj, Planel, and Fishman in magnetic fields to $B = 15$ T.[7] They characterized the average z component of manganese spin by the empirical equation

$$\langle S_z \rangle = \bar{S} B_{5/2}[5\mu_B B / k_B(T + T_0)] , \quad (1)$$

where $B_{5/2}$ is the Brillouin function for spin $S = \frac{5}{2}$, μ_B is the Bohr magneton, k_B is the Boltzmann constant, and T is the temperature. $\bar{S}$ and T_0 are treated as fitting parameters which vary strongly with x and more weakly with T. $\bar{S}(x)$ is less than S due to strong nearest-neighbor antiferromagnetic coupling between Mn^{2+} ions, and $T_0(x) > 0$ arises from weaker, more-distant-neighbor coupling.

Subsequent measurements of $M(B)$ for $x > 0.2$ revealed significant deviations from the modified Brillouin function described above.[8] The present work at fields above 15 T shows even larger deviations. In this paper we show that $M(B)$ at liquid-helium temperatures can be described by the sum of two contributions: (i) a paramagnetic modified Brillouin function that saturates at low fields and (ii) a high-field susceptibility that is linear in B. The relative contributions of these two parts vary with manganese concentration: at low x the saturating part is largest, whereas at high x the linear contribution dominates. This trend is related to both the decreasing fraction of isolated clusters with increasing x, and to the random exchange fields experienced by each magnetic ion.[4]

Magnetic fields, furnished at the Francis Bitter National Magnet Laboratory pulsed-field facility, were produced by multilayer, steel-reinforced, copper solenoids operating at liquid-nitrogen temperature.[9] A field of 45 T with a pulse length of 10 msec is furnished by this facility. Minor heating of the sample was caused by the rapidly changing field, but for the data presented the sample temperature was within ± 1 K of the 4.2-K bath temperature as determined from low-field ($B \leq 15$ T) fits to Eq. (1). Faraday rotation measurements were made using a fiber-optical arrangement to transfer light from He-Ne and diode laser sources to the Dewar, as described previously.[10] The sample holder, immersed in liquid helium, held a "sandwich" consisting of a mirror, sample (~ 1 mm thick), and plastic linear polarizer. With the large rotations obtained using light nearly resonant with the band gap (within a few hundred meV), the output light signal was oscillatory with a period for every 180° of rotation, Fig. 1. Typically, 20 to 40 rotations were observed by 40 T. Relative magnetization measurements in a dc field to 20 T showed that $M(B)$ was proportional to the Faraday rotation to within about 10%. M was slightly larger at 20 T, which may be interpreted as a small decrease in the Verdet constant for increasing field. The assumption of a field-independent Verdet constant may produce an error in $M(B)$ of $+20/-10\%$ over the 40-T field range. The relative $M(B)$ up to 40 T was derived from the rotation angle as a function of field, assuming a field-independent Verdet constant, and was then scaled using absolute dc measurements of M at $B = 5$ T. The connection between

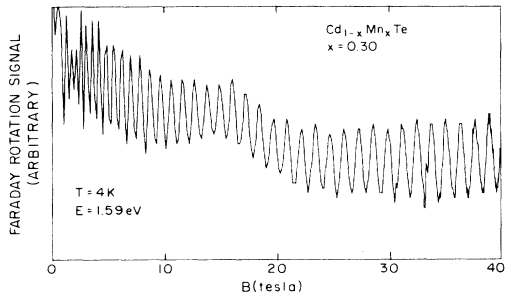

FIG 1. Faraday rotation versus applied magnetic field B for $Cd_{1-x}Mn_xTe$, $x=0.3$. Each peak represents 180° rotation for an optical path length of 0.114 cm, laser photon energy of 1.59 eV, and liquid-helium temperature.

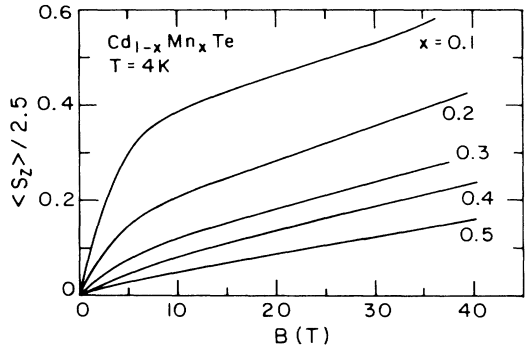

FIG. 3. Average z component of Mn^{2+} ion spin $\langle S_z \rangle$ normalized to $\frac{5}{2}$ versus applied magnetic field B, for $Cd_{1-x}Mn_xTe$, $x=0.11$, 0.20, 0.30, 0.38, and 0.49, at liquid-helium temperature.

$\langle S_z \rangle$ and M, in units of emu/g, is $M=(2\mu_B xA/W)\langle S_z \rangle$, where A is Avogadro's number and W is the molecular weight of $Cd_{1-x}Mn_xTe$.

Figure 2 shows $M(B)$ for samples with $x=0.1$ and 0.4 at $T=4$ K. Note that the overall magnitude of M is not a strong function of x. However, the low-field region is dominated by low x values, while in the high-field region the reverse is true. Although M does not change markedly with x, $\langle S_z \rangle$ is a strong function of the manganese concentration, as seen in Fig. 3. For increasing x there is a substantial decrease in the average spin, resulting from the increase in antiferromagnetic interactions. The most remarkable feature of these data is the exceedingly linear behavior (within a few percent) above 15 T. Linear-like behavior at high fields has been predicted by numerical Monte Carlo calculations.[5]

We now illustrate that $M(B)$ can be decomposed into a Brillouin-type part and a linear part, described by

$$M(B)=M_s B_{5/2}[5\mu_B B/k_B(T_{\text{eff}})]+\chi_{\text{HF}}B \ . \qquad (2)$$

T_{eff} is used instead of $T+T_0$ because T_0 itself depends on T. Good agreement between experiment and Eq. (2) was obtained for $0.1 \leq x \leq 0.5$. Figure 4 displays fits to the data for $x=0.2$ and 0.5 (the curves through the data points). The linear and nonlinear contributions are shown separately in the figures. The best-fit values for $\chi_{\text{HF}}(x)$, $M_s(x)$, and $T_{\text{eff}}(x)$ are compiled in Table I. Notice that as x increases, M_s decreases while χ_{HF} increases, but above $x=0.3$, χ_{HF} is constant at 0.24. These features are discussed below.

For materials with low manganese concentrations ($x<0.1$) the magnetization at low temperatures is well understood.[11] M_s is less than M_0 (obtained at full saturation) because the antiferromagnetic coupling energy of nearest neighbors is greater than both k_BT and $5\mu_BB$. When the Mn^{2+} ions are distributed randomly,[11] M_s is determined by (a) the probability of finding ions in singlets, pairs, triplets and larger clusters and (b) the total spin of the ground state for each of these clusters.[12] For $x=0.1$, agreement with the data is achieved using clus-

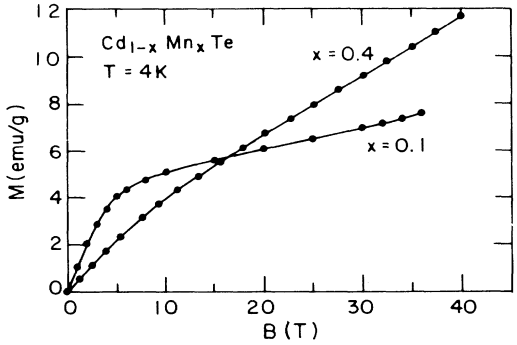

FIG. 2. Magnetization M vs applied magnetic field B for $Cd_{1-x}Mn_xTe$, $x=0.11$ and 0.38. Results were obtained from Faraday rotation at liquid-helium temperature. Solid lines were drawn through the data points for clarity.

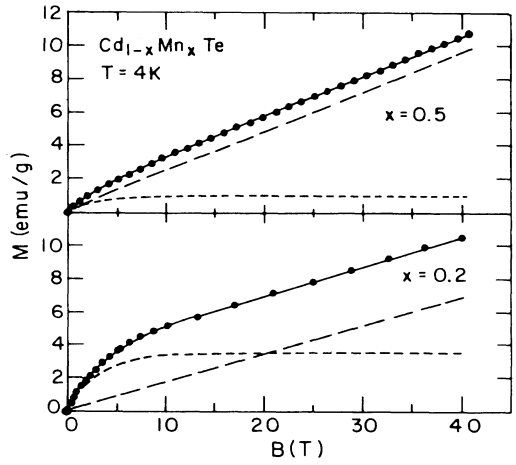

FIG. 4. Components of Eq. (2) for magnetization M vs applied magnetic field B, for $Cd_{1-x}Mn_xTe$, $x=0.20$ and 0.49, at liquid-helium temperature. The solid circles show the experimentally measured data, and the solid lines represent fits of Eq. (2). The dotted and dashed lines represent the saturating and linear contributions of Eq. (2), respectively.

TABLE I. Parameters fit to Eq. (2). Uncertainties are estimated at 5%, except for M_s and χ_{HF}, which are 20%.

Manganese mole fraction x	T_{eff} (K)	M_s (emu/g)	χ_{HF} (emu/g T)
0.11	6.5	4.5	0.08
0.20	7.3	3.5	0.17
0.30	9.2	2.4	0.19
0.38	12.7	1.8	0.25
0.49	6.2	1.8	0.24

ters of up to three spins. However, for $x > 0.1$, M_s cannot be determined by this simple model because the majority of spins are in clusters larger than triplets. For example, at $x = 0.2$, 85% of the spins are in these larger clusters, and the clusters of three Mn^{2+} ions or less contribute only one-half of the measured M_s. Similarly, the high-field slope χ_{HF} can be predicted only for $x \leq 0.05$. The slope is derived from steps in $M(B)$ resulting from internal transitions of clusters with two or more Mn^{2+} ions.[11,13] In this regime a near-linear dependence arises from thermal broadening of the steps observed in $M(B)$. Unfortunately, this prescription for χ_{HF} does not apply for the x values in present study.

Although there is no simple theory of $M(B)$ for $x > 0.1$, numerical calculations have been made for a microscopic model of a diluted isotropic Heisenberg antiferromagnet.[4] This model of a random network of coupled spins was applied to $M(B)$ data for $KMn_xMg_{1-x}F_3$ which show a linear susceptibility in the large x limit and substantial nonlinear component at smaller x.[14] For $x = 0.39$ the calculations suggest that the small isolated clusters cannot account for the majority of the *nonlinear* $M(B)$ response. Additional contributions arise from the gradual saturation of local ferrimagnetic variations, where each Mn^{2+} spin feels an exchange field of a different magnitude and direction.

We emphasize, on the other hand, that the *linear* contribution to $M(B)$ for $x > 0.2$ arises mainly from changes in the *internal* magnetization of large clusters and the infinite chain. The exceedingly linear behavior of our data is qualitatively similar to the perpendicular component in antiferromagnets,[15] dilute antiferromagnets,[14] and dilute metallic glasses.[16,17] This feature seems to be a common property of diluted magnetic systems.

The limiting value of $\chi_{HF} = 0.24$ can be compared to the isotropic susceptibility, χ_{AF}, in the mean-field approximation for an antiferromagnet below the Néel temperature ($T_N \geq 36$ K, from $Cd_{1-x}Mn_xTe$ at $x = 0.7$).[12] Assuming nearest-neighbor interactions only, a type-II face-centered cubic array of Mn^{2+} ions has four independent, interpenetrating simple cubic sublattices, giving[18]

$$\chi_{AF} = \frac{A(g\mu_B)^2}{12WzJ} ,$$

where $z = 4$ is the number of sublattices and J is the nearest-neighbor exchange constant. Using $J/k_B = 6.1$ K,[19] and $x = 0.5$, then $\chi_{AF} = 0.25$ emu/g T. The good agreement with $\chi_{HF} = 0.24$ is fortuitous, since we have neglected higher-neighbor interactions and one only expects qualitative agreement (factor of 2) with such mean-field calculations. We further point out that the formula for χ_{AF} also holds in the diluted case ($x < 1$); although the susceptibility *per ion* is proportional to $1/xz$, the susceptibility *per volume* is independent of x. This supports the limiting behavior for χ_{HF} as x increases.

If we extrapolate the high-field behavior of $\langle S_z \rangle$ of Fig. 3, it suggests that full saturation ($S = 5/2$) should occur at 100, 120, and 160 T, for $x = 0.1$, 0.2, and 0.3, respectively. We plan to extend these measurements to this higher field range and to higher temperatures.

In summary, for fields above 20 T we clearly see two components of the low-temperature magnetization of (Cd,Mn)Te for $x \geq 0.1$. The first component is described phenomenologically by a paramagnetic Brillouin-type function, and arises from isolated clusters at low x, and from gradual saturation of ferrimagnetic fluctuations at higher x. At fields above about $B = 15$ T this part saturates, and a contribution linear in field dominates. This linear susceptibility arises from changes in the *internal* magnetic moment of large clusters and the infinite network of coupled spins, and appears to be a general property of dilute systems.

We thank Y. Shapira, P. A. Wolff, and J. J. Zayhowski for stimulating and helpful discussions, and J. Conlon for technical assistance. This work was supported by the National Science Foundation DMR-8504366, and the Office of Naval Research N00014-83-K-0454. The Francis Bitter National Magnet Laboratory is supported by the National Science Foundation through its Division of Materials Research. E. D. Isaacs is supported by AT&T Bell Laboratories.

*Also at Department of Physics.

[1]N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 193 (1984); J. K. Furdyna, J. Appl. Phys. **53**, 7637 (1982); and references cited therein.

[2]S. Oseroff and P. H. Keesom, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1986).

[3]R. R. Galazka, S. Nagata, and P. H. Keesom, Phys. Rev. B **22**, 3344 (1980).

[4]A. Brooks-Harris and S. Kirkpatrick, Phys. Rev. B **16**, 542 (1977).

[5]J. J. Zayhowski, Ph.D. thesis, Massachusetts Institute of Technology, 1986.

[6]P. A. Wolff, in *Semiconductors and Semimetals*, edited by R. K. Willardson and A. C. Beer (Academic, New York, 1987).

[7]J. A. Gaj, R. Planel, and G. Fishman, Solid State Commun. **29**, 435 (1979).

[8]D. Heiman, Y. Shapira, S. Foner, B. Khazai, R. Kershaw, K.

Dwight, and A. Wold, Phys. Rev. B **29**, 5634 (1983).
[9]S. Foner, Appl. Phys. Lett. **49**, 982 (1986).
[10]D. Heiman, Rev. Sci. Instrum. **56**, 684 (1985).
[11]Y. Shapira, S. Foner, D. Ridgley, K. Dwight, and A. Wold, Phys. Rev. B **30**, 4021 (1984); Y. Shapira, S. Foner, P. Becla, D. N. Domingues, M. J. Naughton, and J. S. Brooks, *ibid.* **33**, 356 (1986).
[12]M. M. Kreitman, F. J. Milford, R. P. Kenan, and J. G. Daunt, Phys. Rev. **144**, 367 (1986); S. Nagata, R. R. Galazka, D. P. Mullin, H. Akbaradeh, G. D. Khattak, J. K. Furdyna, and P. H. Keesom, Phys. Rev. B **22**, 3331 (1980).
[13]R. L. Aggarwal, S. N. Jasperson, P. Becla, and R. R. Galazka, Phys. Rev. B **32**, 5132 (1985).
[14]D. J. Breed, K. Gilijamse, J. W. E. Sterkenburg, and A. K. Miedema, Physica **68**, 303 (1973).
[15]J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966).
[16]R. Boucher, Phys. Status Solidi A **10**, 197 (1977); R. Ferrer, R. Harris, D. Zobin, and M. J. Zuckermann, Solid State Commun. **26**, 451 (1978).
[17]G. R. Gruzalshi and D. J. Sellmyer, Phys. Rev. B **20**, 184 (1979).
[18]P. W. Anderson, Phys. Rev. **79**, 705 (1950).
[19]Y. Shapira and N. F. Oliveira, Jr. (unpublished).