Spin glass behavior of $Cd_{1-x}Mn_xTe$ below the nearest-neighbor percolation limit

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Magnetization measurements down to 10 mK of $Cd_{1-x}Mn_xTe$ for Mn concentrations $0.01 \leqslant x \leqslant 0.15$ show spin glass behavior. Such behavior is attibuted to short-range exchange and dipolar interactions. Both interactions are used to explain the concentration dependence of the spin freezing temperatures for $0.01 \leqslant x \leqslant 0.6$, the short-range exchange dominating at high concentrations, and the dipolar interaction at low concentrations.

INTRODUCTION

Dilute magnetic semiconductors have been the subject of considerable interest in recent years not only for their semiconductor behavior but also for their magnetic properties. The magnetic property of interest here is the spin glass behavior at low temperatures. $Cd_{1-x}Mn_xTe$, a member of this group of materials, has been studied extensively down to ~ 1.4 K. There was evidence[1-3] for a spin glass phase below a spin freezing temperature T_g for concentrations above the nearest-neighbor percolation limit, $x \approx 0.2$. Below this percolation limit, it was generally believed that CdMnTe remains in the paramagnetic phase. However, preliminary magnetization measurements[4] below 1 K for $0.0005 \leqslant x \leqslant 0.05$ concentrations have shown that for $x = 0.05$ there is spin glass behavior, even though it is below the percolation limit presumably due to more distant neighbors. The temperature dependence of EPR linewidth data suggests[3] spin freezing temperatures below 1 K. We report here an investigation of this problem for a wide range of concentrations. Contrary to general belief, spin glass behavior occurs for concentrations below the percolation limit, thus supporting our preliminary measurements. With this information a new magnetic phase diagram is presented extending the concentration range well below the percolation value and showing spin glass behavior down to the limit of our measurements, 0.01 K. To explain our results, a model based on short-range exchange and long-range dipolar interactions is presented. This model is also extended successfully to other studies of this system.

EXPERIMENTAL DETAILS

The samples came from the same source as in Ref. 3. Their composition and homogeneity were analyzed by atomic absorption and density measurements. The agreement between the measured and nominal concentrations was better than 2%. The single crystalline samples were crushed, ground, and then passed through a sieve #400. The fine powder was pressed inside an epoxy, Epibond cylinder, the sample size being 0.2 cm in diameter by 0.6 cm long. Magnetization measurements were taken with a SQUID magnetometer. Each sample was field cooled in 1 Oe down to 10 mK inside the mixing chamber of a ^3He-^4He dilution refrigerator. Temperatures were measured by a CMN thermometer also located inside the mixing chamber. Measurements were taken by heating the sample in small steps from 10 mK up to 4 K. Figure 1 shows the magnetization for $x = 0.10$ normalized to field and concentration.

At a certain temperature there is a kink in the magnetization and we call it the spin freezing temperature T_g. A similar characterization of T_g for this material was done by other groups for higher concentrations.[3] All the other samples, with $x = 0.01, 0.05, 0.125$, and 0.15, had similar behavior, each concentration showing its characteristic kink at T_g. As the concentration was reduced, the kinks were less pronounced. To show more clearly the spin freezing a plot of $\log M$ vs $\log T$ is made and is presented in Fig. 2. A similar kink is observed in the dc susceptibility measurements on $Eu_{0.1}Sr_{0.9}S$ by Maletta and Fesch.[5] In fact, their ac susceptibility cusp corresponds to a nearly invisible kink in the dc data. Below T_g, the magnetization still increases as the temperature is lowered due to free spins and groups of spins.

DISCUSSION AND CONCLUSIONS

Since the samples are field cooled, there is no cusp in the magnetization as a function of temperature. There is, however, a kink for each concentration as a result of a sudden change in slope of the magnetization. For every sample, we define the spin freezing temperature T_g as the temperature for the occurrence of the kink.

Such identification of the spin freezing temperature is justified by the fact that exactly the same method has been used for higher-concentration samples by other groups,[3] although the higher-x samples show a sharper kink than our lower-x samples.

Because CdMnTe has a large energy gap (1.6 eV and increasing with Mn concentrations) it behaves like an insulator, there being no RKKY interactions. Actually the interaction between spins is a short-range antiferromagnetic exchange $J(R)$ depending strongly on the distance R between spins. It is usually written in the form

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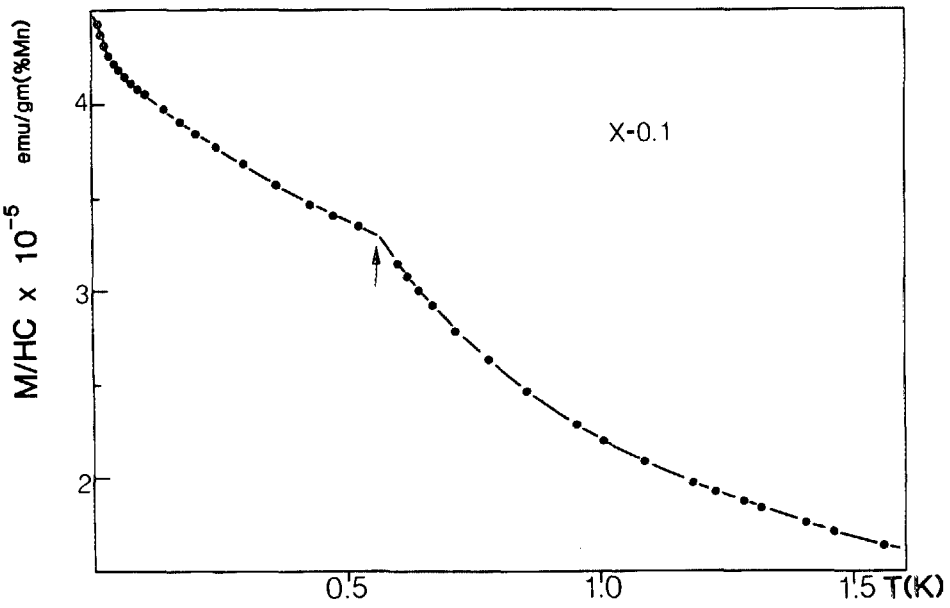

FIG. 1. Magnetization of $Cd_{0.9}Mn_{0.1}Te$ as a function of temperature. Arrow indicates spin freezing temperature, T_g.

$$J(R) = J_0(R)\exp(-\alpha R) \tag{1}$$

and was first proposed by Bloembergen and Rowland,[6] with subsequent analysis and modification by Abrikosov,[7] and Bastard and Lewiner[8] as to the spatial dependence of $J_0(R)$. The damping factor α is equal to $[2E_g(m_e + m_h)/\hbar]^{1/2}$ for semiconductor with a band gap E_g and electron and hole masses m_e and m_h.

When the damping term α is large, as in CdMnTe, the exchange interaction is short ranged. Thus it was reasonable to take as a model a nearest-neighbor percolation limit corresponding to $x \approx 0.2$, the system remaining paramagnetic below this limit. However, other internal interactions must eventually cause ordering in the system as it cannot remain paramagnetic down to 0 K. This is exactly what our results show: the spin freezing at low concentrations occurring mainly because of dipolar interactions. It is a long-range interaction varying as $1/R^3$ and hence it will be effective in coupling spins at low concentrations. Since $1/R^3$ is proportional to x, the dipolar interaction will depend on x. Also, since $J_0(R)$ has a $1/R^3$ dependence, combining the dipolar and exchange interaction leads to

$$T_g = Ax + Bx\exp[-\alpha(x)x^{-1/3}]. \tag{2}$$

We assume T_g is proportional to the mean effective field at the Mn site. The damping term in the exchange depends on x because the energy gap depends[9] on x (we neglect the concentration dependence of the effective masses). We fit this expression to our data, which are presented on the new magnetic phase diagram in Fig. 3, with the following values:

$$T_g(x) = (2.4\text{ K})x + (7200\text{ K})x \times \exp[-2.8(1.6 + 1.59x)^{1/2}x^{-1/3}]. \tag{3}$$

The solid line in Fig. 3 shows the best fit to the data. Agreement for the concentration range $0 \leqslant x \leqslant 0.6$ is excellent. Ear-

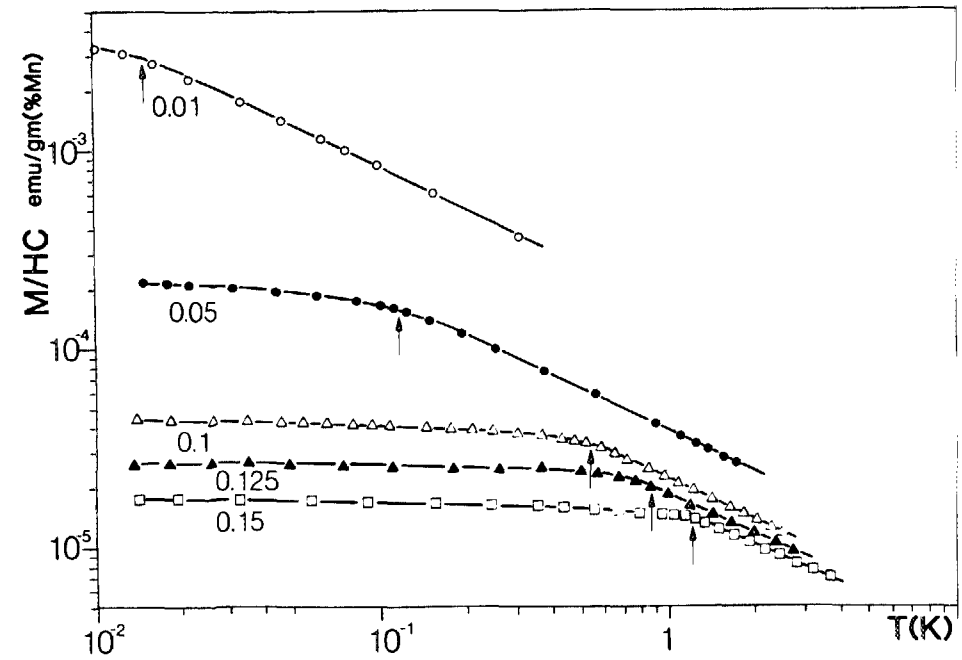

FIG. 2. Log of magnetization as a function of Log T for $x = 0.15$ to 0.01 samples. Arrows indicate spin freezing temperatures T_g.

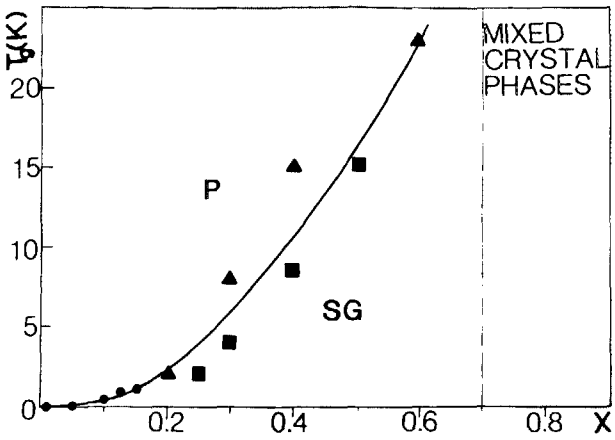

FIG. 3. Magnetic phase diagram of $Cd_{1-x}Mn_xTe$. The squares are data from Ref. 2 and the triangles from Ref. 3; the circles are our data. The solid line is a fit to all data using Eq. (3).

lier we had fit our data to only an x^2 dependence; this was valid only for a small concentration range and had no physical significance.

Our model, based on exchange and dipolar spin glass behavior, is used to explain the magnetic phase diagram of CdMnTe alloys and the results of other groups, without the need for percolation theory.

The anomaly in the specific heat in $Cd_{0.9}Mn_{0.1}Te$ observed by Galazka *et al.*[1] at $T = 0.5$ K corresponds to our $T_g = 0.54$ for the $x = 0.1$ sample. Their specific heat is that of a spin glass.

At low x, susceptibility measurements of Oseroff[3] and specific-heat data[1] were analyzed in terms of clusters larger than expected from a random distribution of spins. Our model with the onset of a dipolar spin glass at low x explains these results.

Although our model on which Eq. (3) is based is a phenomenological one, the order of magnitude of the parameters is reasonable. Further work is necessary to relate these parameters to the more detailed characteristics of the system.

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