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Dynamic random fields in diluted magnetic semiconductors: $Cd_{1-x}Mn_xTe$

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We report electron-spin-resonance measurments in the diluted magnetic semiconductor $Cd_{1-x}Mn_xTe$, for $0.05 \leq x \leq 0.70$, $4 \leq T \leq 300$ K, and $3 \leq \nu \leq 50$ GHz. Over wide ranges of T and x the line broadens with increasing x and lowering T, but is Lorentzian and centered at $g \simeq 2$. A Lorentzian distribution of random fields, centered at zero, is introduced to account for the data. The x dependence of the width (Δ) of this distribution changes sharply at the site percolation threshold of an fcc lattice. Δ varies with T as $\exp(-T/T_0)$.

Electron paramagnetic resonance (EPR) studies[1–3] on diluted magnetic semiconductors (DMS), such as $Cd_{1-x}Mn_xTe$, have already revealed a number of interesting features. For instance, in Ref. 3 it has been reported from this laboratory that (i) at 300 K, although the observed linewidth increases from 80 Oe to 2.2 kOe as x is raised from 0.05 to 0.70, the line remains strictly symmetric, (ii) the g value is independent of x, and (iii) for $x=0.3$, and several other DMS, the linewidth increases with lowering T as

$$\Gamma=\Gamma_0+\Gamma_1\exp(-T/T_0) \ , \quad (1)$$

a form suggested by Bhagat, Spano, and Lloyd[4] several years ago to represent the temperature variation of the linewidth in a number of magnetic systems which show a transition to the spin-glass phase at low T. To understand (i) and (ii) it was suggested, in Ref. 3, that in DMS there exists a random field centered at zero. Such a field would vary in magnitude and direction from one Mn site to another, and would in general be a function of time. From the point of view of EPR the important quantity is the projection of the internal field, along the applied dc field direction, averaged over some characteristic time of the spin system. In Ref. 3 a Gaussian distribution was proposed to account for the variation of the linewidth with x at 300 K. In this note we present additional EPR measurments on $Cd_{1-x}Mn_xTe$ which, by and large, confirm the results of Ref. 3 but with some notable differences. For instance, detailed studies of the line shape show that a Gaussian distribution of random internal fields is inappropriate; rather, a Lorentzian distribution is required. The x variation of the half-width at half maximum (HWHM) Δ of the distribution shows a *sharp* transition at $x=0.2$; very close to the critical concentration for site percolation on an fcc lattice. When the temperature is lowered Δ increases as $\exp(-T/T_0)$.

Single crystals of $Cd_{1-x}Mn_xTe$ with nominal concentrations $0.05 \leq x \leq 0.7$ were grown at Purdue University by the Bridgman Technique. $Cd_{1-x}Mn_xTe$ has a homogeneous zinc-blende crystal structure for $x \leq 0.7$. The Mn ions occupy an fcc lattice as they substitute, at random, for Cd.

Standard EPR techniques were used at microwave frequencies of $\nu=3$ GHz for $77 \leq T \leq 300$ K, $\nu=10$, and 35 GHz for $1.5 \leq T \leq 300$ K, and $\nu=50$ GHz for $T=300$ K. Details of the high-frequency spectrometers can be found in earlier reports[5] from this laboratory. The 3-GHz system will be described in a separate paper.[6] In summary, using the field modulation technique, one traces out the derivative of the absorption curve and designates the linewidth Γ as the separation of the peaks and defines the resonance field H_R from the zero of the derivative curve (cf. Fig. 2).

Repeated measurements suggest that the Γ values are known to within 10% and the line center has a precision of about a percent.

First, the linewidths are independent of frequency. Next, for a given T they increase monotonically with x (cf. Fig. 1, Ref. 3). Finally, the T dependence, shown in Fig. 1, can be summarized as follows. For $x \leq 0.2$, Γ remains relatively constant as T is lowered below 300 K, except at very low T where Γ increases with reducing T in every case. Although not obvious in Fig. 1, even the 0.05 compound has a slight increase for $T \leq 30$ K. For $x \geq 0.25$, Γ begins to increase, with reducing T, around room temperature. The results shown in Fig. 1 are in good agreement with earlier[1,2] reports, as is the observation that H_R is independent of T except at very low T. What was not noted before is that, although there is a sizable ($\sim$1–4 kOe) increase in Γ, *the line shape remains Lorentzian* over wide ranges (column 5 of Table I) of temperature. At temperatures below those given in Table I the line becomes asymmetric and there is also a downshift in H_R. In the present paper we will discuss only the regime in which the lines broaden with increasing x and lowering T but neither change their shape nor show any

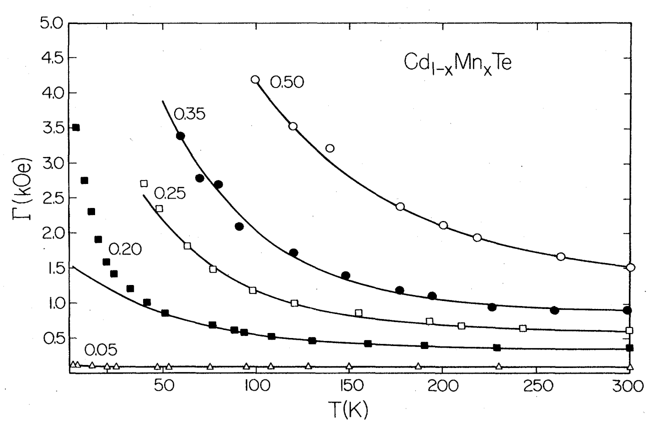

FIG. 1. Temperature dependence of the peak-to-peak linewidth Γ for different values of Mn concentration. The full lines were obtained using Eq. (1) and the parameters given in Table I (see text).

TABLE I. Linewidth parameters [Eq. (1)] for $Cd_{1-x}Mn_xTe$.

x	Γ_0 (Oe)	Γ_1 (Oe)	T_0 (K)	Temperature range (K)
0.05	80			
0.1	200	450	35	110–4
0.15	250	700	45	150–25
0.175	300	1200	55	180–40
0.2	350	1200	55	230–40
0.25	600	4000	50	240–40
0.30	650	4500	50	220–40
0.35	900	8000	55	250–70
0.45	1100	8500	55	250–100
0.5	1200	10000	80	300–120
0.6	1400	11000	80	300–125
0.7	1500	12000	105	300–135

shift away from an H_R value consistent wth $g=2.01\pm0.02$. It is notable that the temperature range of interest here is well above any spin-glass transition temperature for these compounds.[7]

Figure 2 shows the widest resonance observed at 300 K. It is centered at 3500 Oe and has $\Gamma=2200$ Oe. Using these parameters, dP/dH was calculated for Lorentzian and Gaussian line shapes, and these values are shown as squares and circles, respectively, in Fig. 2. Clearly, the observed line shape is very close to being Lorentzian. This is surprising since the HWHM of this resonance is of the same order as H_R.

It is well known that for a strongly damped oscillator the resonance is not only shifted with respect to its position for low damping but is also asymmetric with respect to the center. Since in the present case the line broadens without changing its position or shape, it seems unlikely that the increase in width comes from a rise in the relaxation rate of the Mn spin. Following Ref. 3, it is suggested that the increase in Γ arises from a distribution of internal fields (or, rather, their projection along the applied field). Obviously, to account for the x and T dependences one has to allow the width of the distribution to increase with rising x and lowering T. In Ref. 3 this distribution was taken to be Gaussian as that seemed to be the most natural choice for a random system. Once it is realized that the observed lines are strictly Lorentzian, it is clear that a Gaussian distribution is inappropriate. It is proposed that in the DMS there exist random internal fields whose average over some characteristic time of the system varies from site to site according to a Lorentzian distribution such that the derivative of the power absorbed in an EPR experiment can be written as

$$\frac{dP}{dH}\sim\int dH_i\frac{(H-H_R-H_i)}{[(H-H_R-H_i)^2+\lambda^2]^2}\cdot\frac{1}{[H_i^2+\Delta^2]^2}\ , \qquad (2)$$

where λ has the significance of the "intrinsic" HWHM for the Mn spin system. To fix λ, one notices that for $x=0.05$, $\Gamma\simeq 80$ Oe, independent of T from 300 to 30 K. It was decided to put $\lambda=80(\sqrt{3}/2)$ which may, in some sense, be regarded as an upper limit. In any case, the quantity of interest here is Δ, the HWHM of the Lorentz distribution of internal fields, and the resultant values of Δ will be only affected slightly if λ is actually somewhat smaller. The broadened line represented by the integral in (2) is itself a Lorentzian, centered at H_R and with a width $\Gamma=80+(2/\sqrt{3})\Delta$.

The temperature and concentration dependences of the width Δ are the main results of the present investigation. First, consider the T dependence. Since the observed resonance remains a Lorentzian and does not shift (in the T ranges shown in Table I) the T dependence of Δ can be inferred directly from that of Γ. The temperature dependence of Γ can be summarized by saying that at high T, Γ is a constant $(=\Gamma_0)$, while at low T it follows Eq. (1). Figure 3 illustrates how the parameters listed in Table I were obtained. It is estimated that Γ_1 is good to about 10% and T_0 to roughly ±10 K. Substituting in Eq. (1) we obtain the full lines shown in Fig. 1 to demonstrate how well Eq. (1) describes the data in the relevant temperature intervals. Thus, it is concluded that in DMS the width of the distribution of internal fields, introduced to account for the large EPR linewidths, varies (over the ranges of T listed in Table I) with T as

$$\Delta=\Delta_0+\Delta_1\exp(-T/T_0) \qquad (3)$$

with T_0 values given in Table I. Also at high T, $\Delta\simeq\Delta_0$, in-

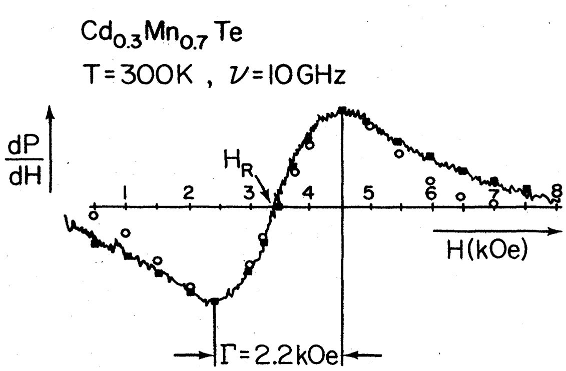

FIG. 2. Derivative of the power absorbed as a function of the applied field for $x=0.70$ at 300 K. The squares and circles indicate values calculated for a Lorentzian and a Gaussian line shape, respectively.

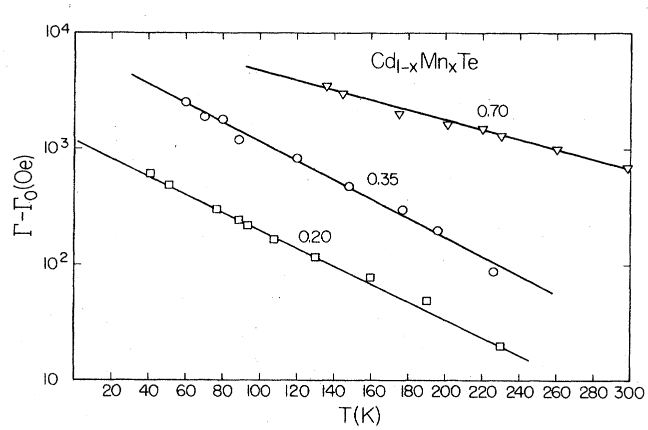

FIG. 3. $(\Gamma-\Gamma_0)$ as a function of T for $x=0.20$, $x=0.35$, and 0.70. Least-squares fits (straight lines) yield the parameters in Table I.

dependent of T. In the absence of a microscopic model one can merely speculate that Δ_1 (or Γ_1) will be determined by the variation in the magnetic environment of a Mn ion while T_0 is a measure of the potential barrier separating two neighboring ground states of a disordered spin system.

Next, consider the x dependence of Δ. Figure 4 shows the x variation of $(\Delta_1+\Delta_0)$, i.e., Δ at $T\rightarrow 0$. Δ_0, i.e., Δ for $T\rightarrow\infty$, has a very similar x dependence. In short, for constant T,

$$\Delta\propto\begin{cases}(x-0.05) & \text{for } x\leq 0.2\\ (x-0.2)^{1/2} & \text{for } x>0.2\ .\end{cases} \tag{4}$$

Considering that for site percolation on an fcc lattice, with only nearest-neighbor interactions, the critical concentration is about 0.2, it is very tempting to suggest that we are observing a percolation transition in the Δ-x plane. The exponent is also not far from that expected for the percolation probability, i.e., the probability that a given site is incorporated in the infinite cluster. However, it is important to note that the percolation probability saturates at $x\simeq 0.24$ whereas Δ obeys Eq. (4) up to $x=0.70$. In this connection it may be useful to point out that recent neutron scattering data[8] on $Cd_{1-x}Mn_xTe$ with $0.6\leq x\leq 0.7$ do not show any evidence of a percolating magnetic cluster. Rather the system seems to have only short-range magnetic order even for these high x values.

The problem of an EPR line arising from a spin system in which there are random internal fluctuating fields attracted considerable attention many years ago. On very general stochastic grounds it was shown[9] that if $\delta\tau_s << 1$, where δ is a measure of the magnitude of the fluctuation and τ_s of its "speed," one can obtain a line which will closely approximate a Lorentzian whose width is $\delta^2\tau_s$. This will, of course, be correct, only as long as the field region of the observation is such that $(H-H_R)\leq\delta$. Farther away from H_R the Lorentzian must give way to a Gaussian. It would be fruitful to account for the observed x and T variations of Γ by appeal to the behavior of δ and τ_s. Indeed, one can write $\Delta=(\delta^2\tau_s-\text{const})$. However, this merely replaces one macroscopic parameter by two unknowns without any guidance, at this time, as to how δ and τ_s may be derived from microscopic theory or other experiments.

In conclusion, on the basis of EPR linewidth and lineshape data, extending over wide ranges of temperature, it is suggested that in DMS, such as $Cd_{1-x}Mn_xTe$, there exists a Lorentzian distribution of random internal fields whose time average can be modeled by a Lorentzian centered at zero. The width of the model distribution is independent of frequency but increases either when x is raised [Eq. (4)] or T is lowered [Eq. (3)]. It does not seem fruitful to quantitatively correlate the present results with stochastic models of EPR.

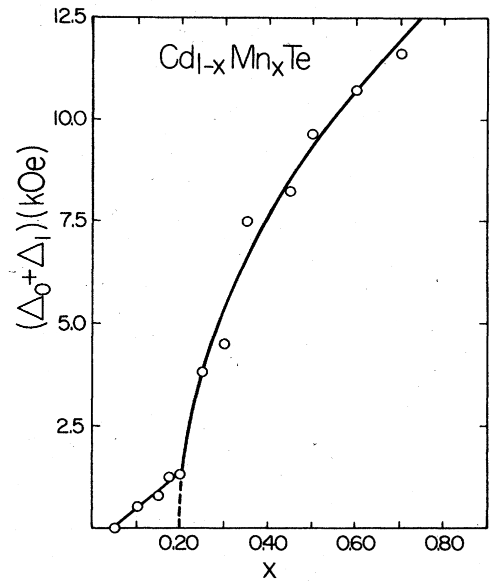

FIG. 4. HWHM Δ of the Lorentzian distribution of internal fields at $T=0$ as a function of Mn concentration.

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