UC Irvine

UC Irvine Previously Published Works

Title

Magnetic behavior of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$

Permalink

<https://escholarship.org/uc/item/6qd4r90h>

Journal

Physics Letters A, 80(4)

ISSN

0375-9601

Authors

Oseroff, SB

Calvo, R

Fisk, Z

et al.

Publication Date

1980-12-08

DOI

10.1016/0375-9601(80)90030-4

Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at

<https://creativecommons.org/licenses/by/4.0/>

Peer reviewed

MAGNETIC BEHAVIOR OF $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$

S.B. OSEROFF¹ and R. CALVO¹

Centro de Física, Instituto Venezolano de Investigaciones Científicas (IVIC), Caracas 1010A, Venezuela

and

Z. FISK² and F. ACKER³

Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, CA 92093, USA

Received 12 August 1980

We report EPR and magnetic susceptibility measurements in single crystals of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ as a function of concentration and temperature. The data indicate that there is a critical concentration $x \approx 0.22$ which we identify with the percolation critical point x_c .

Semiconducting systems with controlled quantity of magnetic elements have recently received considerable attention [1,2]. $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ is a new semiconductor with a large gap and wurtzite structure within a wide range of concentration ($0 \leq x \leq 0.5$).

Electron paramagnetic resonance (EPR) has been found to be useful technique to study spin-spin interactions as well as effects related to magnetic phase transitions [3]. Broadening of the EPR linewidths and changes of the gyromagnetic factors (g) with decreasing temperatures have been observed in systems undergoing ferromagnetic, antiferromagnetic and spin glass phase transitions [4-6].

In this work we have measured the EPR spectra and magnetic susceptibility (χ) in single crystals of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ for different concentrations ($0.0005 \leq x \leq 0.45$). By fitting the broadening of the resonance linewidth and the shift of the resonance field as a function of temperature to a modified Huber expression [1-3] we have deduced the concentration dependence of the order-disorder transition temperature (T_c). At

$0.2 \leq x \leq 0.25$, we find a discontinuity in T_c suggestive of a percolation critical point.

Solid solutions of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ were grown using a modified Bridgman technique; good quality crystals were obtained for x up to 0.45. The concentration was measured by atomic absorption. The EPR measurements have been made at 9 GHz using a Varian spectrometer as a function of temperature ($1.5 \leq T \leq 300$ K). The Faraday method was used to obtain the χ data between $90 \leq T \leq 300$ K.

The reciprocal susceptibility is shown as a function of temperature in fig. 1. In the range of temperature measured, the data obey a Curie-Weiss law $\chi_{\text{mol Mn}}^{-1} = T - \Theta/C$. Values for the Curie constant C and the asymptotic Curie temperature Θ are shown in table 1, where C should be compared with the free ion value $C = 4.375 \text{ K cm}^3/\text{mol Mn}$;

The EPR spectra of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ have been studied for low concentration ($x \leq 0.005$), and a well separated hyperfine structure is observed [7]. As the concentration is increased beyond $x \approx 0.005$, the hyperfine lines initially broaden due to the dipole-dipole interaction, eventually becoming a single broad line for $x \approx 0.015$. Then, the line narrows with a further increase of x due to exchange narrowing, with the minimum linewidth corresponding to $x \approx 0.03$. For larger concentration we observed a single, symmetrical resonance

¹ Work supported in part by CONICIT, Venezuela.

² Work supported by National Science Foundation Grant, no. DMR77-08469.

³ Work supported in part by the Swiss National Science Foundation.

Fig. 1. Magnetic susceptibility of Cd_{1-x}Mn_xSe as a function of temperature and concentration for $H = 8.5$ kOe.

line which broadens monotonically with increasing concentration. The line shape is qualitatively lorentzian characteristic of exchange narrowing.

In fig. 2 we present our measurements of the EPR linewidth as a function of temperature for $0.05 \leq x \leq 0.45$. A significant increase in linewidth as a function of decreasing temperature is observed for all samples. In fig. 3 the values of the gyromagnetic factors are shown as a function of temperature for $0.15 \leq x \leq 0.25$; for other values of x , changes of g were not observed since we were limited, either because the linewidth becomes comparable to the resonance field at 9 GHz (the case for $x > 0.25$), or our lowest available temperature of 1.5 K (the case for $x < 0.15$). We have measured the angular dependence of the EPR line and

Fig. 2. Temperature dependence of the EPR linewidth for $0.05 \leq x \leq 0.45$. Solid lines are least squares fitting with eq. (1).

found that there is a small variation of linewidth $\approx 3\%$ independent of concentration.

The observed broadening of the EPR linewidth with

Table 1
Values measured as a function of x in Cd_{1-x}Mn_xSe of the Curie temperature Θ , and the Curie constant C ; temperatures $T_{c\Delta H}$ and T_{cg} and critical exponents $\alpha_{\Delta H}$ and α_g as obtained from the temperature dependences of ΔH and g .

$x (\pm 1)$ (%)	$-\Theta (\pm 10)$ (K)	$C (\pm 0.10)$ K cm ³ /mol Mn	$T_{c\Delta H}$ (K)	T_{cg} (K)	$\alpha_{\Delta H} (\pm 0.2)$	$\alpha_g (\pm 0.5)$
5	10	3.65	$\ll 1$	—	0.2	
11			$\ll 1$	—	0.3	
15	80	4.05	$\ll 1$	$\ll 1$	0.4	2.2
20			≈ 0.3	≈ 0.3	0.5	2.9
23			≈ 1	≈ 0.4	0.6	3.2
25	135	4.10	≈ 1	≈ 1	1.0	5.4
30			3.5 ± 1		1.3	
35	220	4.35	6 ± 1		1.8	
41			9 ± 1		1.9	
45	330	4.90	11.5 ± 1		2.0

Fig. 3. Temperature dependence of the g -factor for $0.15 \leq x \leq 0.25$. Solid lines are least squares fitting with eq. (1).

decreasing temperature can be described using a modified Huber expression:

$$\Delta H = A [T_c / (T - T_c)]^\alpha + B(\Theta / T + 1), \quad (1)$$

where ΔH denotes the EPR linewidth, α the critical exponent, T_c the temperature of the order-disorder transition and $B(\Theta / T + 1)$ is the high temperature linewidth. The first term on the right accounts for the dynamical contribution and is valid for T close to T_c , and the second explains the decrease of the linewidth over a region of temperature where $T \gg T_c$.

The change of the g -factor with temperature can be analyzed with an expression similar to eq. (1), where the second term on the right is replaced by the value of the gyromagnetic factor measured at high temperature ($g \approx 2.00$). Since this formula is expected to be valid near T_c , the values of T_c , $\alpha_{\Delta H}$ and α_g listed in table 1, were obtained using only low temperature data. The solid lines shown in figs. 2 and 3 are a least squares fitting of the experimental points with eq. (1).

From the parameters listed in table 1, we can separate the data into three regions of concentration: $x < 0.20$, $0.20 \leq x \leq 0.25$, and $x > 0.25$. In theories of

the site percolation problem, it is found that the mean cluster becomes infinitive above a certain critical concentration x_c . Long range magnetic ordering does not take place for $x < x_c$ where only finite clusters of magnetic ions exist. In this region short range magnetic ordering occurs and the behavior can be analyzed in terms of a cluster model [8]. This model could explain, in our case, the rising linewidth and shift of the resonance field in the region of $x < 0.25$, as originated from the growth of short-range magnetic correlations. For $x > 0.25$, T_c is larger and increases linearly with concentration, as is shown in table 1. The rapid change in T_c occurs at $x \approx 0.22$ which we identify with a percolation critical point x_c . A similar result is obtained for $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ [1].

Preliminary magnetization measurements at low dc magnetic field in $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ and $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ indicate that these systems have micromagnetic behavior above $x = 0.2$ [9].

As a conclusion, our EPR data indicate that there is a value of critical concentration in the region of $0.2 \leq x_c \leq 0.25$. This value agrees with the results of calculations based on a series expansion of the mean cluster size, considering the nearest neighbors [10]. However, more data (ac and dc magnetic susceptibility, specific heat, etc.) are needed to understand the magnetic behavior of this system.

We would like to thank Professor W. Girit for supplying the crystals.

References

- [1] S.B. Oseroff, R. Calvo, W. Girit and Z. Fisk, *Solid State Commun.* 35 (1980) 539.
- [2] S.B. Oseroff, R. Calvo, W. Girit and Z. Fisk, *Bull. Am. Phys. Soc.* 25 (1980) 548.
- [3] D.L. Huber, *Phys. Rev.* 6 (1972) 3180.
- [4] E. Dormann and V. Jaccarino, *Phys. Lett.* 48A (1974) 81.
- [5] R.H. Taylor and B.R. Coles, *J. Phys.* F5 (1975) 121.
- [6] B.S. Salomon and R.M. Herman, *Phys. Rev. Lett.* 41 (1978) 1506.
- [7] J. Schneider, S.R. Sirear and A. Rauber, *Z. Naturforsch.* 18a (1963) 980.
- [8] A.W. Simpson, *Phys. Stat. Sol.* B40 (1970) 207.
- [9] F. Acker, unpublished results (1980).
- [10] C. Domb and N.W. Dalton, *Proc. Phys. Soc.* 89 (1966) 859.