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

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We report EPR and magnetic susceptibility measurements in single crystals of Cd$_{1-x}$Mn$_x$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]. Cd$_{1-x}$Mn$_x$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 ($\chi$) in a single crystals of Cd$_{1-x}$Mn$_x$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 Cd$_{1-x}$Mn$_x$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 $\chi$ 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 $\Theta$ are shown in table 1, where $C$ should be compared with the free ion value $C = 4.375$ K cm$^3$/mol Mn;

The EPR spectra of Cd$_{1-x}$Mn$_x$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

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Fig. 1. Magnetic susceptibility of Cd$_{1-x}$Mn$_x$Se 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 found that there is a small variation of linewidth $\approx 3\%$ independent of concentration. The observed broadening of the EPR linewidth with

<table>
<thead>
<tr>
<th>$x$ (± 1)</th>
<th>$-\Theta$ (± 10)</th>
<th>$C$ (±0.10)</th>
<th>$T_{c\Delta H}$ (K)</th>
<th>$T_{cg}$ (K)</th>
<th>$\alpha_{\Delta H}$ (±0.2)</th>
<th>$\alpha_{g}$ (±0.5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>10</td>
<td>3.65</td>
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<td>--</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td>$\leq 1$</td>
<td>--</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>80</td>
<td>4.05</td>
<td>$\leq 1$</td>
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<td>0.4</td>
<td>2.2</td>
</tr>
<tr>
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<td></td>
<td>4.10</td>
<td>$\approx 0.3$</td>
<td>$\approx 0.3$</td>
<td>0.5</td>
<td>2.9</td>
</tr>
<tr>
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<td></td>
<td></td>
<td>$\approx 1$</td>
<td>$\approx 0.4$</td>
<td>0.6</td>
<td>3.2</td>
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<tr>
<td>25</td>
<td>135</td>
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<td>$\approx 1$</td>
<td>$\approx 1$</td>
<td>1.0</td>
<td>5.4</td>
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<tr>
<td>30</td>
<td></td>
<td>3.5 ± 1</td>
<td></td>
<td>1.3</td>
<td></td>
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<tr>
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<td>4.35</td>
<td>6 ± 1</td>
<td>1.8</td>
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</tr>
<tr>
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<td>9 ± 1</td>
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<td>1.9</td>
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<tr>
<td>45</td>
<td>330</td>
<td>4.90</td>
<td>11.5 ± 1</td>
<td>2.0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The change of the g-factor with temperature can be described using a modified Huber expression:

$$\Delta H = A \left[ \frac{T_c}{(T - T_c)} \right]^\alpha + B(\Theta/T + 1),$$  

(1)

where $\Delta H$ denotes the EPR linewidth, $\alpha$ 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 $\alpha_g$ 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 infinite 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 Cd$_{1-x}$Mn$_x$Te [1].

Preliminary magnetization measurements at low dc magnetic field in Cd$_{1-x}$Mn$_x$Se and Cd$_{1-x}$Mn$_x$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.

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References