Static magnetic susceptibility of $Zn_{1-x}Mn_xSe$

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We present results of an investigation of the static magnetic susceptibility of $Zn_{1-x}Mn_xSe$ for $x=0.05$, 0.15, 0.30, and 0.45. From the Curie-Weiss behavior of the susceptibility at high temperatures, we determine the effective Mn-Mn exchange constant J_1 to be -13.5 ± 0.95 K. By using the results of our study in conjunction with direct measurements of the nearest-neighbor exchange integral J_{NN} from inelastic neutron scattering by nearest-neighbor pairs, we obtain an estimate of the next-nearest-neighbor exchange constant $J_2=-2.4$ K.

I. INTRODUCTION

The classification and analysis of the static magnetic susceptibility of alloys belonging to the $A(II)_{1-x}Mn_x$-$B(VI)$ family of diluted magnetic semiconductors[1] (DMS) is critical for the understanding of magnetism in these systems. Fortunately, the behavior of the magnetic susceptibility in these materials is very systematic and has recently been placed on a sound theoretical footing,[2] at least in the high-temperature limit. Static susceptibility measurements are a useful source of information on the Mn-Mn exchange interaction in DMS, which has been the focus of extensive experimental and theoretical investigations.[2-10] Since the magnetic properties of DMS are largely determined by this interaction, the characterization of the Mn-Mn exchange is clearly of fundamental importance. In this report, we present the results of an investigation of the low-field static magnetic susceptibility of $Zn_{1-x}Mn_xSe$, and examine the Mn-Mn exchange interaction using its high-temperature behavior.

As shown in Ref. 2, the static magnetic susceptibility χ can be analytically derived in the high-temperature limit by assuming that the Mn^{2+} ions constitute a randomly diluted magnetic system interacting via antiferromagnetic Heisenberg exchange:

$$H=-2\sum_{\substack{i,j\\ i<j}} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \ . \tag{1}$$

The calculation of χ in the high-temperature limit then yields the Curie-Weiss behavior:

$$\chi=\frac{C(x)}{T-\Theta(x)} =\frac{C_0x}{T+\Theta_0x} \ . \tag{2}$$

In Eq. (2), the Curie constant per unit mass $C(x)$ is given by

$$C(x)=\frac{(g\mu_B)^2S(S+1)}{3k_B}\frac{N}{m}x \ , =C_0x \ , \tag{3}$$

where N is the total number of cation sites in a sample of mass m. The Curie-Weiss temperature $\Theta(x)$ is given by

$$\Theta(x)=\left[\tfrac{2}{3}S(S+1)\sum_p J_pz_p/k_B\right]x \ , =-\Theta_0x \ , \tag{4}$$

where J_p is the exchange integral between pth neighbors and z_p is the number of cations in the pth coordination sphere. (Note that our definition of Θ_0 differs from that in Ref. 2 by a sign change.) If *only* nearest-neighbor interactions are considered, the summation $\sum_p J_pz_p$ can be approximated by z_1J_1. The quantity J_1 can then be calculated from the experimental value of the Curie-Weiss temperature

$$J_1/k_B=-\frac{3}{2}\frac{\Theta_0}{S(S+1)z_1} \ , \tag{5}$$

where $z_1=12$ for wurtzite and zinc-blende DMS. (We shall refer to the value of J_1 obtained in this approximation as the "effective exchange constant," as opposed to the true nearest-neighbor exchange constant J_{NN}.) Similarly, the value of S can be obtained from $C(x)$. Typically, S turns out to be close to $\frac{5}{2}$, as would be expected for the Mn^{2+} ions.

II. EXPERIMENTAL METHOD

The static susceptibility measurements were carried out in the temperature range 8–300 K using a S.H.E. VTS-

905 SQUID magnetometer with the samples suspended by thin cotton threads. The temperature dependence of the magnetization M of each sample was measured at constant magnetic field H and the susceptibility was obtained from $\chi(T)=M(T)/H$. The applied fields were $H=10$ kOe for the $x=0.05$ sample and $H=1$ kOe for the samples with $x=0.15$, 0.3, and 0.45. (Note that these values of x are *nominal* concentrations; however, electron microprobe analysis of adjacent sections of the ingot showed that the variation in the actual composition was within $\pm 5\%$ of the nominal value of x.) The field dependence of the magnetization of each sample was measured at 100 and 300 K to check the linearity of $M(H)$. This ensured that there were no ferromagnetic inclusions in the samples. Note also that all the *quantitative* analysis in this paper is restricted to temperatures above 100 K, i.e., the Curie-Weiss regime. The diamagnetic susceptibility of ZnSe was measured at high temperatures ($\chi_d = -0.303\times10^{-6}$ emu/g) and was subtracted from the data as the diamagnetic contribution of the lattice.

III. ANALYSIS AND DISCUSSION

The temperature dependence of χ^{-1} for the various samples is shown in Figs. 1 and 2. At high temperatures, the susceptibility shows a good fit to the Curie-Weiss form in Eq. (2), while there is a systematic deviation from the Curie-Weiss law at low temperatures. This behavior is common to all DMS. For the highest concentration $x=0.45$, we observe a very clear cusp at $T=20$ K, which is characteristic of the spin-glass transition.[1]

The Curie constant $C(x)$ and the Curie-Weiss temperature $\Theta(x)$ can be determined from the linear high-temperature part of the data shown in Figs. 1 and 2. They are given in Table I. Note that it is important to subtract the diamagnetic contribution of the ZnSe lattice, particularly for the lowest Mn concentrations, to avoid erroneous values of $C(x)$ and $\Theta(x)$. This is illustrated in Fig. 1. Note also that the validity of the Curie-Weiss law goes far beyond the asymptotic regime $T \gg |\Theta(x)|$ for which it has been derived.[2] Using the results given in Table I in conjunction with Eq. (2), the high-temperature value of χ for $Zn_{1-x}Mn_xSe$ can be expressed as

$$\chi = \frac{0.033x}{T+944x} , \tag{6}$$

where χ is in units of emu/gK. The effective exchange constant J_1 can be determined from Θ_0, as described in Eq. (5). We find an average of $J_1 = -13.5 \pm 0.95$ K. The values of S have also been calculated, yielding an average $S = 2.59 \pm 0.15$.

The value of J_1 obtained above is higher than the value of the nearest-neighbor exchange constant J_{NN} as measured by more direct methods involving inelastic neutron scattering by nearest-neighbor Mn^{2+} pairs[3] ($J_{NN} = -12.3$ K) and high-field magnetization steps[4] ($J_{NN} = -12.6$ K). This is to be expected, since our calculations have ignored the next-nearest-neighbor integral J_2, as well as higher-order terms, in the summation $\sum_p J_p z_p$. It is reasonable to assume that the $p=2$ contribution is the leading term after $p=1$. We can then obtain an estimate of J_2 by assuming the neutron scattering value of J_{NN} to be correct. Using Eq. (4), and neglecting terms in the sum for $p>2$, we then have

$$\frac{J_2}{k_B} = \frac{1}{z_2}\left[-\frac{3}{2}\frac{\Theta_0}{S(S+1)} - \frac{J_{NN}}{k_B}z_1\right] , \tag{7}$$

where $z_2=6$ for the wurtzite $Zn_{1-x}Mn_xSe$. This yields a value of $J_2 = -2.4$ K, or $J_2/J_{NN} \simeq 0.2$ which is comparable to the situation in $Cd_{1-x}Mn_xTe$ (Ref. 5) ($J_2/J_{NN} \simeq 0.2$), and $Cd_{1-x}Mn_xSe$ (Ref. 5) ($J_2/J_{NN} \simeq 0.3$).

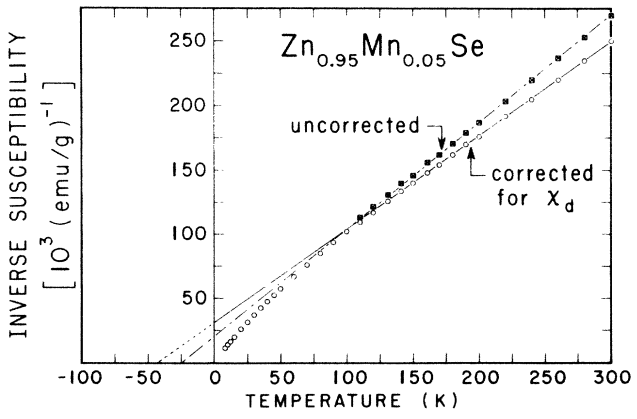

FIG. 1. Inverse susceptibility vs temperature for $Zn_{0.95}Mn_{0.05}Se$. The best fit of the high-temperature data to the Curie-Weiss law is shown, and the fitting parameters $C(x)$ and $\Theta(x)$ are given in Table I. We also show the data uncorrected for the diamagnetic contribution of ZnSe to indicate the degree of error which can result when the diamagnetic contribution is not taken into account.

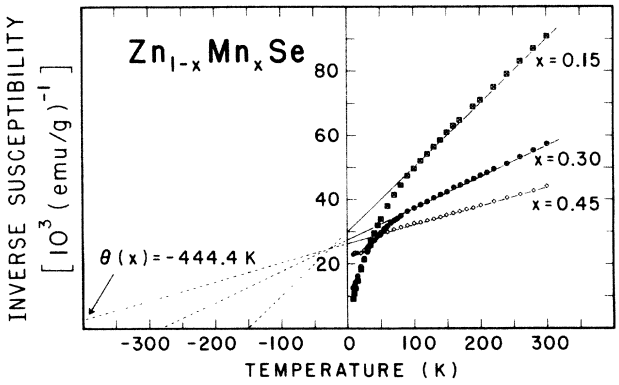

FIG. 2. Inverse susceptibility vs temperature for $Zn_{1-x}Mn_xSe$ with $x=0.15$, 0.3, 0.45. The diamagnetic contribution of ZnSe has already been subtracted. The best fit of the high-temperature data to the Curie-Weiss law is shown, and the fitting parameters $C(x)$ and $\Theta(x)$ are given in Table I.

TABLE I. Parameters used in fitting high-temperature susceptibility data to the Curie-Weiss law.

x	$C(x)$ (10^3 emu/g K)	C_0 (10^3 emu/g K)	S	$\Theta(x)$ (K)	Θ_0 (K)	J_1 (K)
0.05	1.37	27.40	2.35	−42.0	840.0	−12.0
0.15	4.96	33.20	2.61	−152.1	1014.0	−14.5
0.30	10.11	33.70	2.63	−280.3	934.3	−13.3
0.45	16.90	37.55	2.78	−444.4	987.5	−14.1
Average		32.96 ± 3.6	2.59 ± 0.15		944 ± 66	-13.5 ± 0.95

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