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A LOW TEMPERATURE SPECIFIC HEAT INVESTIGATION OF $Cd_{1-x}Mn_xSe$ AND $Cd_{1-x}Mn_xS$

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Abstract

The specific heat of $Cd_{1-x}Mn_xSe$ ($x = 0.01$) and $Cd_{1-x}Mn_xS$ ($x = 0.026$) in the temperature range of 0.3 - 40K was measured. The measurements were carried out in different applied magnetic fields (H = 0,10, 20, 28KG). The results were analyzed with a model which includes long range interactions. Calculations based on on extended version of nearest neighbor pair approximation (including triplets) yielded a reasonable agreement with the experimental results. This approximation is seen to provide a good simultaneous description of the specific heat, susceptibility and magnetization results.

Introduction

Over the past few years, intensive investigations have been performed on the magnetic behavior of diluted magnetic semiconductors, i.e., *II-VI* or *II-V* compounds containing controlled quantities of randomly substituted magnetic ions such as $Hg_{1-x}Mn_xTe$, $Cd_{1-x}Mn_xTe$, $(Cd_{1-x}Mn_x)_3As_2$, $(Zn_{1-x}Mn_x)_3As_2$.

The properties of these materials have been extensively reviewed by several authors [1-3]. In an earlier publication [4], we attempted to interpret the susceptibility and specific heat experimental results simultaneously on the basis of a unique model. We reached the conclusion that by applying the ENNPA model based on incorporating short range exchange interactions (J_{nn}, J_{nnn}) as well as a long range interaction of the type $J/(R)^n$ the contradictory viewpoints on the magnetic behavior of DMS,s can be reconciled. We finally concluded with two remarks:

Firstly, as in our simplified proposed model clusters when more than two members are ignored, then, it is desirable to take into account the role of larger clusters such as triplets. Hence, in the present investigation we extended the model to include clusters with three ions (triplets).

Secondly, in the previous paper [4] we restricted ourselves to zero magnetic field results. In order to complete the investigation, the specific heat data obtained in a non-zero magnetic field should also be compared with the model based calculated results. In view of this, in this paper we present calorimetric (sepecific heat) data of $Cd_{1-x}Mn_xS$ ($x = 0.026$) and $Cd_{1-x}Mn_xSe$ ($x = 0.01$) in zero and non-zero magnetic fields.

Keywords: Condense matter; Specific heat

Experimental Results

The specific heat measurements were made between 0.3 and 50 K using the standard heat-pulse technique in a conventional He3 cryostat which has been described in detail in [5]. The magnetic contribution to the specific heat C_m was obtained by subtracting the lattice contribution of pure Cd S(or Cd Se) from the total specific heat. The results are shown in Figures 1 and 2.

Interpretation. In order to extend the model to include open triples, i.e. configurations where two spins are located at the same distance from the central spin (two spins at the same shell), the Hamiltonians and their eigenvalues for pair and open triangles are written as follows:

$$H^P = -2J\vec{S}_1.\vec{S}_2 - g\mu_B(S_1^z + S_2^z)B$$

$$E = -J[S(S+1) - 35/2] - g\mu_B m B$$

$$0 \leq S \leq 5 \quad , \quad m \leq 5$$

$$H^T = -2J[\vec{S}_1.\vec{S}_2 + \vec{S}_2.\vec{S}_3] - g\mu_B(S_1^z + S_2^z + S_3^z)B$$

$$\vec{S}_1 + \vec{S}_2 + \vec{S}_3 = \vec{S} \quad , \quad \vec{S}_1 + \vec{S}_3 = \vec{S}_a$$

$$E = -J[S(S+1) - S_a(S_a+1) - 35/2] - g\mu_B m B$$

$$0 \leq S_a \leq 5 \quad , \quad S_a - 5/2 \leq S \leq S_a + 5/2 \quad , \quad m \leq 5$$

The probability of finding the nearest spin in the v^{th} shell

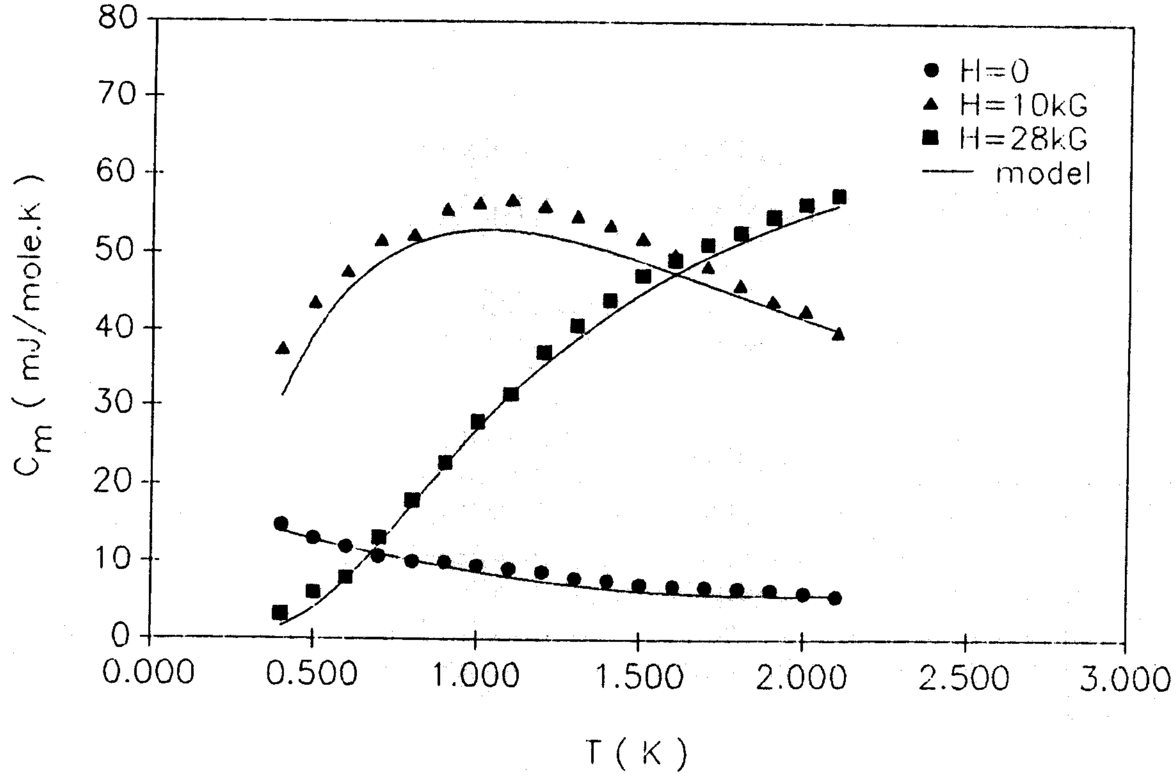

Fig.1· Magnetic specific heat versus temperature of $Cd_{1-x}Mn_xSe$ ($x = 0.01$). The solid curves are the result of model calculations using $x = 0.011$, $J_1/k_B = -8$ K, $J_2/k_B = -5.2$ K and $J/k_B = -14.7/(R)^{6.8}$ K.

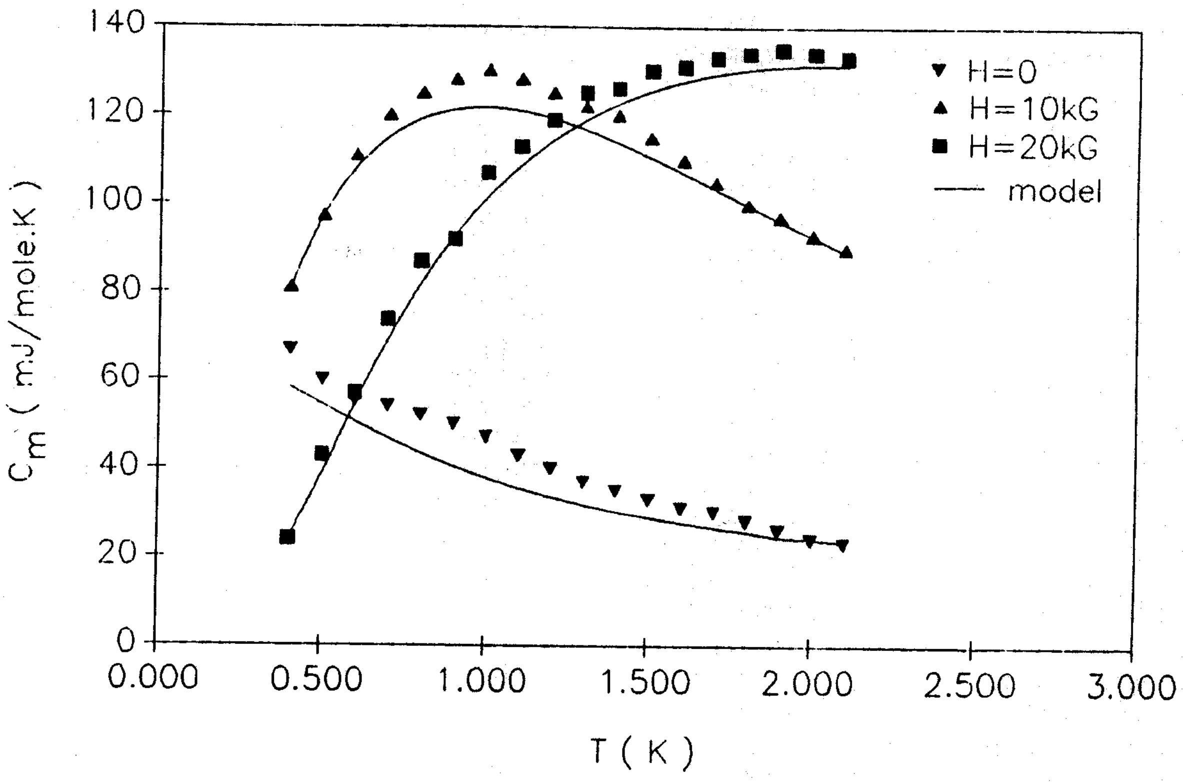

Fig.2· Magnetic specific heat versus temperature of $Cd_{1-x}Mn_xS$ ($x = 0.026$). The solid curves are the result of model calculations using $x = 0.03$, $J_1k_B = -10.6$ K, $J_2/k_B = -0.4$ K and $J/k_B = -10$ K.

for a random distribution of spins is:

$$P_v(x) = (1-x)_v^{n-1} - (1-x)^{n}{}_v$$

where, $n_v = \sum_{1}^{v} N_v$, N_v is the number of lattice sites in shell_v.

The probability for finding two neighbors in the same shell (triple) is:

$$P_v^T(x) = 1/2\, N_v (N_v - 1)\, x^2 (1-x)_v^{n-2}$$

Then, the probability for a pair is approximated by:

$$P_v{}^P(x) = P_v(x) - P_v{}^T(x)$$

Figures 3 and 4 compare the probability of finding a neighbor in shell v (pair) with the probability of finding two neighbors in shell v (triple). The total specific heat can be obtained by summing the respective contributions over the shells according to the probability of the pairs and triples in them.

$$C_m = \sum_{v=1}^{\infty} [P_v{}^P(x) C^P{}_{m.v}/2 + P_v{}^T(x) C^T{}_{m.v}/3]$$

Summation over the shells is carried out up to shell $\bar{v}$ for which

$$\sum_{v=1}^{\bar{v}} [P_v^P(x) + P_v{}^T(x) \geq \%\, 99$$

For x = 0.026 we obtained: $\sum_{v=1}^{18} P_v{}^P = 0.87$, $\sum_{v=1}^{18} P_v{}^T = 0.12$.

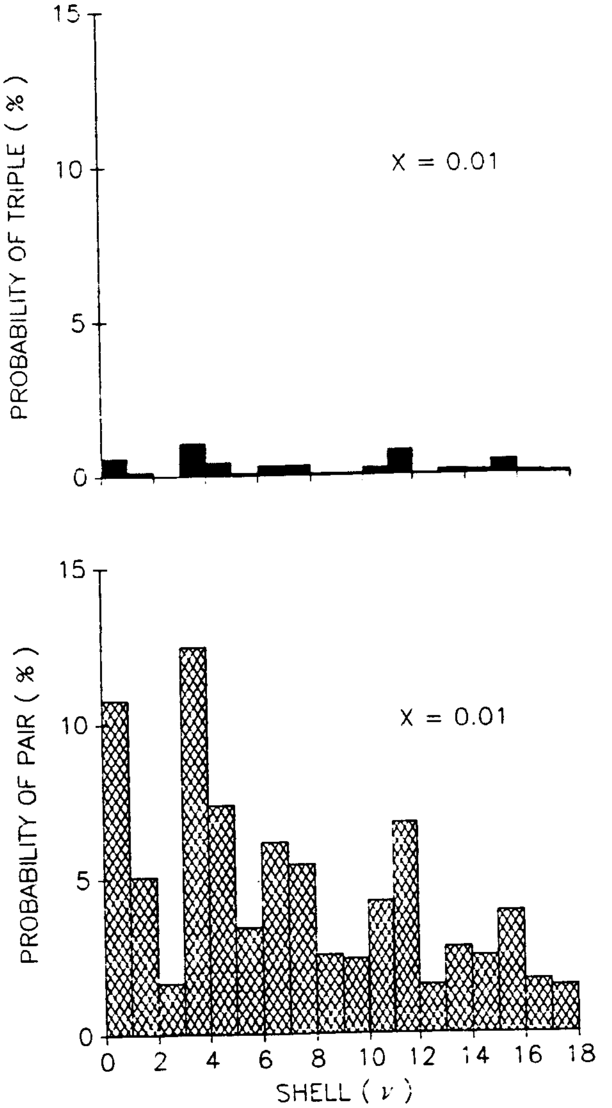

Fig.3 Probability of finding a neighbor in shell v (pair) represented by the crosshatch histogram and probability of finding two neighbors in shell v (triple) represented by the filled histogram.

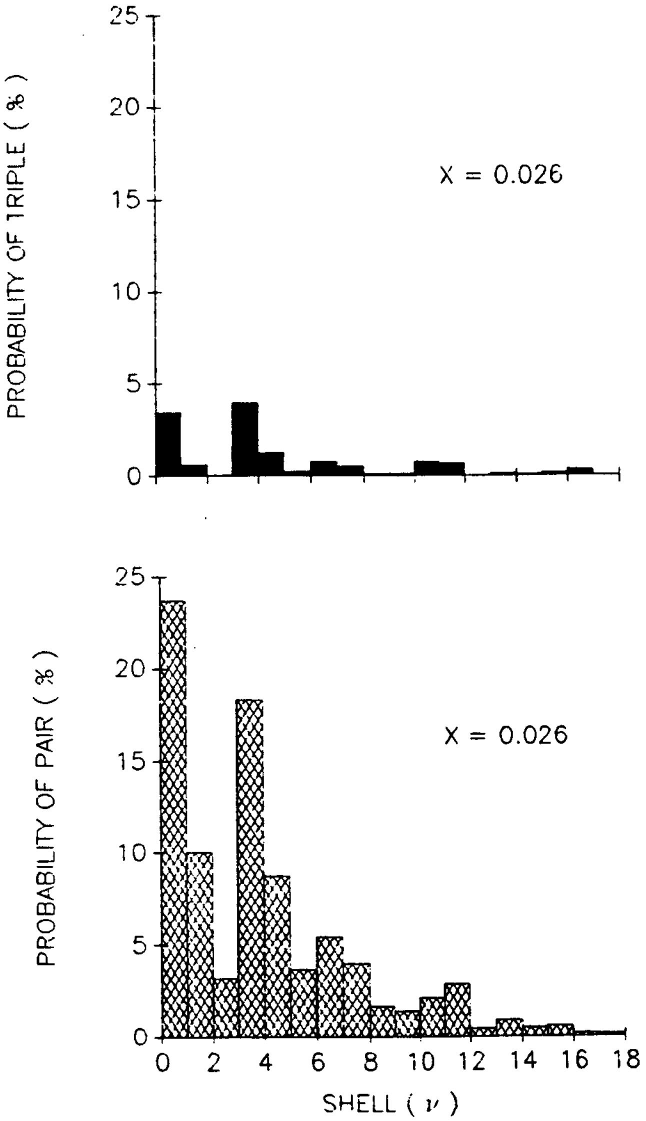

Fig.4 Probability of finding a neighbor in shell v (pair) represented by the crosshatch histogram and probability of finding two neighbors in shell v (triple) represented by the filled histogram.

For a lower concentration ($x = 0.01$) a higher $\bar{v}$ was needed.

As was stressed in the previous publication [4] the experimental specific heat and susceptibility data can be understood on the basis of the following assumptions:
(i) Relatively strong nearest and moderate next-nearest neighbor antiferromagnetic interactions (J_1, J_2).
(ii) An AF long-range interaction of the type $J(R)^{-n}$ for ions beyond next-nearest neighbor ($n = 6.8$ for wide gap samples [7] such as $Cd_{1-x}Mn_xS$ and $Cd_{1-x}Mn_xSe$).
(iii) A random distribution of Mn ions.
(iv) $g = 2$, $S = 5/2$ based on susceptibility results. The J_1 values obtained from independent experimental and theoretical sources are shown in Table I. The J_2 and J parameters were chosen such that the best overall agreement for specific heat experimental results are obtained.

Table I. Nearest and next-nearest neighbor exchange interaction (K)

Material	This paper J_1/k_B	Others J_1/k_B	This paper J_2/k_B	Others J_2/k_B
$Cd_{1-x}Mn_xS$	-10.6	-10.6±0.2[ab]	-0.4	-0.3[c] -4.7[d]
$Cd_{1-x}Mn_xSe$	- 8	-7.9[e] -8.3[f] -8±0.5[g]	-5.2	-5.2±0.3[a]

a reference 8 , b reference 12 , c reference 9
d reference 10 , e reference 13 , f reference 14
g reference 15

The calculated results for C_m are compared with experimental data in Figures 1 and 2. It is shown that the overall agreement between calculated results and the experimental data for both samples (specially low X one) are fair. Furthermore, the value of next-nearest neighbor integral J_2 for $Cd_{1-x}Mn_xSe$ obtained by our calculation ($J_2/k_B = 5.2$ K) is in excellent agreement with the results of Bartholomew *et al.* [8] ($J_{nnn}/k_B = -5.2 \pm 0.3$). For $Cd_{1-x}Mn_xS$ we obtained $J_2/k_B = 0.4$ K which is in good agreement with Keesom's experimental results [9] ($J_2/k_B = 0.3$ K), but far from the estimated value ($J_2/k_B = -4.7$ K) of Chen *et al.* [10].

We also apply the model to obtain the value of the Curie-Weiss temperature for both compounds. As the calculations were presented in the previous paper [4], we will not repeat them here. The results are shown in Table II. In $Cd_{1-x}Mn_xS$ for best fitting we assumed $x = 0.03$ which is well within the limits of concentration for a boule with a nominal concentration of 0.026. The calculated value for Curie temperature is $\theta = 790$ K which is in fair agreement with that of other workers [10] $\theta = 907 \pm 80$ K). In $Cd_{1-x}Mn_xSe$ we obtained $\theta = 790$ K which is in reasonable agreement with the results of Spalek *et al.* [11] ($\theta = 743 \pm 15$K).

Table II. Curie-Weiss temperature θ (K)

Material	This paper	Other sources
$Cd_{1-x}Mn_xS$	790	907±80[d]
$Cd_{1-x}Mn_xSe$	790	743±15[h]

d reference 10 , h reference 11

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