Journal of
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Magnetic phase diagram of semimagnetic semiconductor $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems

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Received 27 June 2008; accepted 22 November 2008

Abstract - The magnetic properties of the $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems in the range $0.75 \le x \le 1$ have been studied by mean field theory and high-temperature series expansions (HTSE). By using the first theory, we have evaluated the nearest neighbour and the next-nearest exchange interaction $J_1(x)$ and $J_2(x)$ respectively. The intra-planar and the inter-planar interactions are deduced. The corresponding classical exchange energy for magnetic structure is obtained for this system. The second theory combined with the Padé approximants (P.A) method are applied to the $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems; we have obtained the magnetic phase diagram (T_N versus dilution x) in the range $0.75 \le x \le 1$. The obtained theoretical results are in agreement with experimental results obtained by magnetic measurements. The critical exponents associated with the magnetic susceptibility (γ) and the correlation lengths (ν) are deduced in the range order $0.75 \le x \le 1$.

PACS: 64.60.Fr; 82.35.Jk; 75.40.Cx.
Keywords: Exchange interactions; High-temperature series expansions; Padé approximants, Critical exponents; Magnetic phase diagram.

1. Introduction

The semimagnetic semiconductors, with the formula AB_2X_4 are of continuing interest because of their wide variety of physical properties. The crystal structure of $MnGa_2Se_4$ has been found tetragonal, possibly isomorphous with $CdGa_2S_4$, space group $I\bar{4}$, with two chemical formulas per unit cell [1,2].

The semimagnetic semiconductors have received considerable attention for their interesting electrical and magnetic properties, which can vary greatly as a function of composition [3–8]. The compounds $(Hg,Cd)Cr_2S_4$ and $(Hg,Cd,Cu)Cr_2Se_2$ have been found [9-13] to be ferromagnetic semimagnetic semiconductor, whereas $ZnCr_2S(Se)_4$ and $MnGa_2Se_4$ have been found [14-18] to be antiferromagnetic semimagnetic semiconductor.

We have calculated the first and the second nearest neighbours exchange interactions $J_1(x)$ and $J_2(x)$, respectively on the basis of magnetic results of $Cu_{2(1-x)}Mn_xGa_2Se_4$ in ordered region $0.75 \le x \le 1$ [16]. The values of the intra-plane and inter-plane interactions J_{aa}, J_{ab} and J_{ac}; respectively, are deduced. The interaction energy of the magnetic structure is obtained for this system. In recent works [3,4,19], we have used the high-temperature series expansions (HTSE) to study the thermal and disorder variation of the short-range order (SRO) in the particular semimagnetic semiconductor systems. The *Padé* approximant (P.A) [20] analysis of the HTSE of the correlation length has been shown to be a useful method for the study of the critical region [21, 22]. We have used the HTSE technique to determine the Néel temperature T_N and the critical exponents γ and ν associated with the magnetic susceptibility χ and the correlation length ξ, respectively for $Cu_{2(1-x)}Mn_xGa_2Se_4$ in the range $0.75 \le x \le 1$.

2. Methods

2.1 Calculation of the values of the exchange integrals from mean field approximation

Starting from the well-known Heisenberg model, the Hamiltonian of the system is given by:

$$H = -2\sum_{i,j} J_{ij}\vec{S}_i\vec{S}_j \tag{1}$$

where, J_{ij} is the exchange integral between the spins situated at sites *i* and *j*. $\vec{S}_i$ is the spin operator of the spin localised at the site *i*. In this work we consider the nearest neighbour (nn) and next nearest neighbour (nnn) interactions J_1 and J_2 respectively.

In the case of semimagnetic semiconductor structure, the mean field approximation leads to a simple relations between the φ angle of helimagnetic ordering and the Néel temperature T_N, respectively, and the considered two exchange integrals J_1 and J_2, describing the $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems are given by [23]:

$$T_N = \frac{2}{3}S(S+1)\lambda(\varphi) \tag{2}$$

where $\lambda(\varphi)$ is the eigenvalue of the matrix formed by the Fourier transform of the exchange integral and $S = 5/2$.

$$\cos(\varphi) = -\frac{1}{4}[\frac{J_1 + 2J_2}{J_2}] \tag{3}$$

Using the experimental values of T_N obtained by magnetic measurement for the system $Cu_{2(1-x)}Mn_xGa_2Se_4$ [16]. We have deduced the values of exchange integrals $J_1(x)$ and $J_2(x)$ in the range

$0.75 \leq x \leq 1$. From these values, we have derived the variation of the intra-plane coupling and the coupling between nearest and next-nearest plane for $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems with $0.75 \leq x \leq 1$.

The obtained values of $J_1(x)$ and $J_2(x)$ are given in Table (1). The values of corresponding classical exchange energy for the magnetic structure [23], the values of the intra-plane $J_{aa} = 2J_1$ and inter-plane interactions, $J_{ab} = 4J_1 + 8J_2$, $J_{ac} = 4J_2$ are given in the table 1 for the $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems in the range $0.75 \leq x \leq 1$.

Table 1. The Néel temperature $T_N(K)$, the values of the first, second, intra-plane, inter-plane exchange integrals and the energy of $Cu_{2(1-x)}Mn_xGa_2Se_4$ as a function of dilution x.

x	$T_N(K)$ [16]	$\frac{J_1}{K_B}(K)$	$\frac{J_2}{K_B}(K)$	$\frac{J_{aa}}{K_B}(K)$	$\frac{J_{ab}}{K_B}(K)$	$\frac{J_{ac}}{K_B}(K)$	$\frac{\lvert E\rvert}{K_B S^2}(K)$
1	7.50	-0.42	-0.21	-0.84	-3.36	-0.21	5.04
0.95	6.50	-0.37	-0.18	-0.74	-2.96	-0.74	4.44
0.93	6.25	-0.35	-0.17	-0.70	-2.80	-0.70	4.20
0.90	5.50	-0.31	-0.155	-0.62	-2.48	-0.62	3.72
0.85	5.25	-0.30	-0.15	-0.60	-2.40	-0.60	3.60
0.80	4.50	-0.25	-0.125	-0.50	-2.00	-0.50	3.00
0.75	4.30	-0.24	-0.12	-0.48	-1.92	-0.48	2.88

2.2. High temperature series expansions

In this section we shall derive the high-temperature series expansions (HTSE) for both the zero field magnetic susceptibility χ to order sixth in β. The relation ship between the magnetic susceptibility per spin and the correlation functions may be expressed as follows:

$$\chi(T) = \frac{\beta}{N}\sum_{ij}\left\langle \vec{S}_i\vec{S}_j\right\rangle \quad (4)$$

where $\beta = \frac{1}{k_B T}$ and N is the number of magnetic ion. $\left\langle S_i S_j\right\rangle = \frac{Tr S_i S_j e^{-\beta H}}{Tr e^{-\beta H}}$ is the correlation function between spins at sites *i* and *j*.

In the previous work [24], the HTSEs are developed for $\chi(T)$ and the correlation length $\xi(T)$ with arbitrary $y = \frac{J_2}{J_1}$ up to sixth order:

$$\chi(T) = \sum_{m=-n}^{n} \sum_{n=1}^{6} a(m,n) y^m \tau^n \tag{5}$$

$$\xi^2(T) = \sum_{m=-n}^{n} \sum_{n=1}^{6} b(m,n) y^m \tau^n \tag{6}$$

where $\tau = \frac{2S(S+1)J_1}{k_B T}$. The series coefficients $a(m,n)$ and $b(m,n)$ are given in Ref [26].

Fig. 1, show magnetic phase diagram of $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems. We can see the good agreement between the magnetic phase diagram obtained by the HTSE technique and the experimental results, in particular in the case of the last systems of which the phase diagrams have been established well by different methods [25-28].

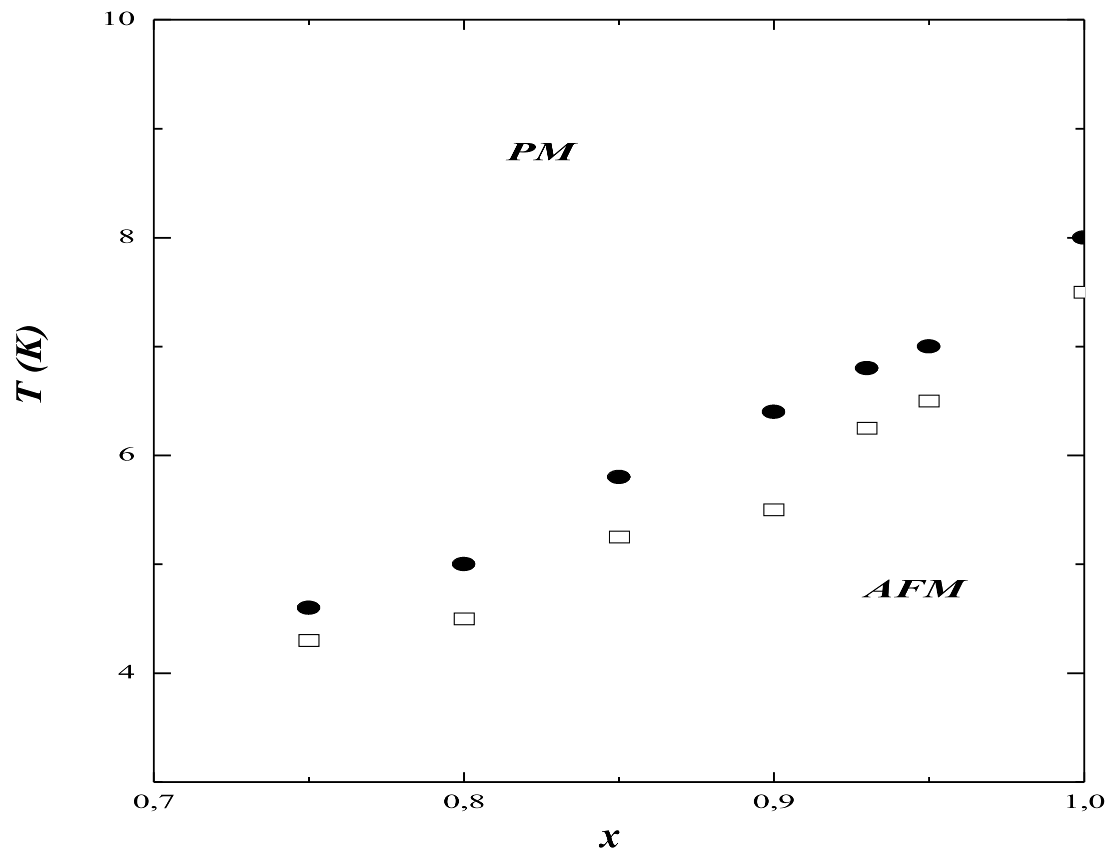

Fig. 1. Magnetic phase diagram of $Cu_{2(1-x)}Mn_xGa_2Se_4$. The various phases are the paramagnetic phase (PM) and antiferromagnetic phase (AFM)$(0.75 \leq x \leq 1)$. The solid circles are the present results. The open squares are deduced by magnetic measurement [16].

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(T)$ and the correlation length $\xi(T)$ are that at the neighbour hood of the critical point the above two functions exhibit an asymptotic behaviour:

$$\chi(T) \propto (T - T_N)^{-\gamma} \tag{7}$$

$$\xi^2(T) \propto (T_N - T)^{-2\nu} \tag{8}$$

Estimates of T_N, γ and ν for $Cu_{2(1-x)}Mn_xGa_2Se_4$, have been obtained by using the HTSE method and the Padé approximate method (P.A) [20]. The simple pole corresponds to T_N and the residues to the critical exponents γ and ν. The obtained central values are $\gamma = 1.37 \pm 0.02$ and $\nu = 0.67 \pm 0.01$. These values of γ and ν are nearest to the one of Heisenberg model and insensitive to the dilution.

3. Discussions and Conclusions

The values of $J_1(x)$ and $J_2(x)$ have been determined from mean field theory, using the experimental data of T_N given in Ref [16], for each dilution (see Table 1). From these values, we have deduced the values of the intra-plane and inter-plane interactions J_{aa}, J_{bb} and J_{ac}, respectively, the energy of the magnetic structure is given in the same table for the $Cu_{2(1-x)}Mn_xGa_2Se_4$ system with $0.75 \le x \le 1$. The values of $J_1(x)$ and $J_2(x)$ decreases with the absolute value when x decreases. The sign of $J_1(x)$ and $J_2(x)$ are negative in the whole range of concentration.

The HTSE extrapolated with Padé approximants method is shown to be a convenient method to provide valid estimations of the critical temperatures for real system. By applying this method to the magnetic susceptibility $\chi(T)$ combined with Padé approximants method we have estimated the critical temperature T_N for each dilution x. The obtained magnetic phase diagram of $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems is presented in Fig.1. Several thermodynamic phases may appear including the paramagnetic (PM) and antiferromagnetic phase (AFM) $(0.75 \le x \le 1)$ for the $Cu_{2(1-x)}Mn_xGa_2Se_4$ systems. In this figure, we have included, for comparison, the theoretical results and the experimental results obtained by magnetic measurement. From this figure one can see good agreement between the theoretical phase diagram and experimental results.

In the other hand, the values of critical exponents γ and ν associated with the magnetic susceptibility $\chi(T)$ and with the correlation length $\xi(T)$, have been estimated in the range of the composition $0.75 \le x \le 1$. The sequence of [M, N] PA to series of $\chi(T)$ and $\xi(T)$ has been evaluated. By examining the behaviour of these PA, the convergence was found to be quite rapid. Estimates of the critical exponents associated with magnetic susceptibility and with the correlation length are found to be $\gamma = 1.37 \pm 0.02$ and $\nu = 0.67 \pm 0.01$, respectively. In the magnetic order the values of γ and ν are nearest to the one of Heisenberg model and insensitive to the dilution.

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