Full Length Research Paper

Magnetic properties of semimagnetic semiconductors materials

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The magnetic properties of semimagnetic semiconductor $A_{1-x}A'_x Te$ (A'= Mn, A = Cd, Zn) are studied. By using the mean field theory in ordered phase and a probability distribution in disordered phase, we have evaluated the nearest neighbouring and the next-neighbouring super-exchange interactions $J_1(x)$ and $J_2(x)$ for $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$. The magnetic phase diagram, that is, T_C versus dilution x, is obtained by the high-temperature series expansions combined with the Padé approximants for the $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems. A spin glass state has been obtained for $x < x_c$ $(x = x_c)$. This phase is attributed to the randomness and the frustration of the antiferromagnetic interactions between the Mn magnetic ions arising from the topology of the diluted magnetic semiconductor alloys.

Key words: Semimagnetic semiconductor, critical temperature, critical exponents, high-temperature series expansions, Padé approximants, phase diagram.

INTRODUCTION

The interest in semiconductors, which are spatially confined to a few tens of nanometers, has incremented in the latest years (Sebastian et al., 2010; Syed et al., 2010; Alsaad et al., 2010; Lu et al., 2011; Cao and Zhang, 2011; Diehl, 1997; Jacobs et al., 2002; Lemine et al., 2010; Gad et al., 2010). Mn-doped IIB-VI semimagnetic semiconductors have attracted considerable interest for several years (Mackowski et al., 2002; Brazis and Kossut 2002). Beside the well-known changes in the electronic structure of ordinary ternary semiconducting compounds due to the composition, IIB-VI compounds show novel magneto-optical and magnetotransport properties if the cation is partly substituted by Mn with its half-filled $3d$

shell (Mackowski et al., 2002). There are no previous reports on theoretical and experimental studies of Zn-doped $MnTe$ semiconductor. This material could be of special interest in the construction of room temperature antiferromagnetic alloys. One of the advantages of II-VI materials is that they can host magnetic ions (e.g., Mn^{2+}) which open the way for studying various spin-dependent phenomena in zero-dimensional geometries. The experimental results show a true order to long carry antiferromagnetic of III type in the system undiluted zinc $MnTe$ blende and the Néel temperature is $T_N = 67K$ (Santiago et al., 2002). The magnetic phase diagrams of the aforementioned two systems consist of two regions: A high-temperature, paramagnetic phase and low temperature spin glass (SG) phase. The latter phase generally occurs when $x > 2$, but recent works has shown that spin freezing can also occurs for lower values of x at very low temperature (Karaoulanis et al., 2000;

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Table 1. The Néel temperature $T_N(K)$, the Curie Weiss temperature $\theta_p(K)$ and the values of the first, second exchange integrals $Zn_{1-x}Mn_xTe$ as a function of dilution *x*.

x	$T_N(K)$ **(Moron, 1996)**	$\theta_p(K)$ **(Moron, 1996)**	$\dfrac{J_1}{k_B}(K)$	$\dfrac{J_2}{k_B}(K)$
0.66	38.94	-611.36	-5.20	-7.02
0.90	60.88	-947.22	-8.07	-10.92
1.00	66.70	-925.00	-8.00	-10.36

Karaoulanis et al., 1996). The magnetic and electronic properties of $Hg_{1-x}Mn_xTe$ and $Ca_{1-x}Mn_xO$ have been studies by Masrour and Hamedoun (2008); Masrour et al. (2010). The first principle studies the effect of Co substitution on the magnetic and electronic properties of wz-ZnO (Benam and Hezari, 2011).

The exchange interactions $J_1(x)$ and $J_2(x)$ of the diluted $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems have been obtained in the range $0.6 \le x \le 1$ by using the mean field theory and a probability distribution law adapting the nature of dilution problem in a disordered phase. Another part of this paper concerns the interesting topic of magnetic structure and spin glass behaviour in the diluted magnetic semiconductors $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ with $0 \le x \le 1$. The *Padé* approximant (P.A) (Baker and Graves-Morris, 1981) analysis of the high-temperature series expansion (HTSE) of the correlation length has been shown to be a useful method for the study of the critical region (Masrour et al., 2010). We used this technique to determine the magnetic phase diagrams of $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ in the range $0 \le x \le 1$.

EXCHANGE INTEGRALS CALCULATIONS

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*, and $\vec{S}_i$ is the atomic spin of the magnetic ion located on the *ith* site.

The factor ″2″ in Equation (1) arises from the fact that, when summing over all possible pairs $\langle ij \rangle$ exchange interactions, we count each pair twice. In this work, we consider the nearest-neighbouring (nn) and next-nearest neighbouring (nnn) interactions J_1 and J_2, respectively. In the case of semimagnetic semiconductor containing the magnetic moment only in the octahedral sublattice, the mean field approximation leads to a simple relations between the paramagnetic Curie-temperature θ_p and the critical temperature T_N, respectively, and the considered two exchange integrals J_1 and J_2.

Following the method of Holland and Brown (1972), the expressions of T_N and θ_p describing the $A_{1-x}A'_xTe$ (A'= Mn, A = Cd, Zn) systems is:

$$T_N(K) = \frac{2S(S+1)}{3k_B}[-4J_1 + 2J_2] \tag{2}$$

$$\theta_p(K) = \frac{2S(S+1)}{3k_B}[12J_1 + 6J_2] \tag{3}$$

where k_B is the Boltzmann's constant and $S = 5/2$ is the spin of Mn^{2+} ions.

To determine $J_1(x)$ and $J_2(x)$ in the whole range of concentration for the semimagnetic semiconductor $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems, we used the experimental values of T_N and θ_p obtained by magnetic measurement (Moron, 1996). We deduced the values of exchange integrals $J_1(x)$ and $J_2(x)$ in the range $0.6 \le x \le 1$. The obtained values of $J_1(x)$ and $J_2(x)$ for the first and the second systems are given in Tables 1 and 2, respectively.

Probability law

To determine $J_1(x)$ and $J_2(x)$ in the whole range of concentration, we used the similarly probability law given

Table 2. The Néel temperature $T_N(K)$, the Curie Weiss temperature $\theta_p(K)$ and the values of the first, second exchange integrals of $Cd_{1-x}Mn_xTe$ as a function of dilution x.

x	$T_N(K)$ (Moron, 1996)	$\theta_p(K)$ (Moron, 1996)	$\frac{J_1}{k_B}(K)$	$\frac{J_2}{k_B}(K)$
0.67	42	-548.75	-4.82	-6.04
0.71	48.81	-730	-6.26	-8.34
1.00	67	-925	-8.04	-10.34

by Stachow et al. (2000). In the semimagnetic semiconductors $A_x A'_{1-x} Te$ systems, only the random placement of the diamagnetic ions A and A' leads to the spatial fluctuations of the signs and magnitudes of the super-exchange interaction between the magnetic ions Mn. Indeed, the magnetic order is very sensitive to the distance between nearest neighbouring Mn ions and to the size of the anions, A and A'. Due to the nature of dilution problem, we choose a probability distribution permitting us to determine exchange integral $J_{AA'}(x)$ for each concentration x. The two exchange integrals of the opposite pure compound ATe and $A'Te$ of the bound random semimagnetic semiconductors are denoted J_A and $J_{A'}$, respectively. The occupation probability $p(i)$ of the two ions A or A' induced in the interaction is

$p(i) = C_n^i x^{n-i}(1-x)^i$. Where n is the total number of lattice sites inside a sphere with the volume $\frac{4}{3}\pi R_i^3$ (R_i denotes the distance between the sites i and j, n is the number of cations, the *Pth* coordination sphere around a given cation chosen as the central one, for the zinc blende structure $n = 12$), while i varies from 0 to 12. The exchange integral for such an occupation is assumed to be: $J_{AA'}^i = (J_A^{n-i} J_{A'}^i)^{1/n}$. We obtain in the case of the zinc blende structure:

$$J_{AA'}(x) = \sum_{i=0}^{12} C_n^i x^{n-i}(1-x)^i (J_A^{n-i} J_{A'}^i)^{1/n}$$

$$J_{AA'}(x) = x^{12}J_A + 12x^{11}(1-x)(J_A^{11}J_{A'})^{(1/12)} + 66x^{10}(1-x)^2(J_A^{10}J_{A'}^2)^{(1/12)} + 220x^9(1-x)^3(J_A^9J_{A'}^3)^{(1/12)} + 495x^8(1-x)^4(J_A^8J_{A'}^4)^{(1/12)} +$$
$$792x^7(1-x)^5(J_A^7J_{A'}^5)^{(1/12)} + 924x^6(1-x)^6(J_A^6J_{A'}^6)^{(1/12)} + 792x^5(1-x)^7(J_A^5J_{A'}^7)^{(1/12)} + 495x^4(1-x)^8(J_A^4J_{A'}^8)^{(1/12)} +$$
$$220x^3(1-x)^9(J_A^3J_{A'}^9)^{(1/12)} + 66x^2(1-x)^{10}(J_A^2J_{A'}^{10})^{(1/12)} + 12x(1-x)^{11}(J_A J_{A'}^{11})^{(1/12)} + (1-x)^{12}J_{A'} \tag{4}$$

If J_A ($J_{A'}$) corresponds to the *nn* interactions of the opposite pure systems A(Mn)Te (A'(Cd or Zn)Te), respectively, $J_{AA'} = J_1(x)$. If J_A ($J_{A'}$) corresponds to the *nnn* super-exchange of the opposite pure systems, $J_{AA'} = J_2(x)$. The using coupling variables $J_1(x)$ and $J_2(x)$ in the pure case discussed later on are given by (Moron, 1996) for $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems. The obtained values of $J_1(x)$ and $J_2(x)$ are used later on for two systems.

HIGH-TEMPERATURE SERIES EXPANSIONS

Here, we shall derive the high-temperature series expansions (HTSEs) for both the zero field magnetic susceptibility χ to order six in β. The relationship between the magnetic susceptibility per spin and the correlation functions may be expressed as follows:

$$\chi(T) = \frac{\beta}{N}\sum_{ij}\left\langle \vec{S}_i \vec{S}_j \right\rangle \tag{5}$$

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. The expansion of this function in powers of β is obtained as follows (Hachimi et al., 2003):

$$\left\langle \vec{S}_i \vec{S}_j \right\rangle = \sum_{l=0}^{\infty}\frac{(-1)^l}{l!}\alpha_l \beta^l \tag{6}$$

With $\alpha_l = v_l - \sum_{k=0}^{l-1} C_k^l \alpha_k \mu_{l-k}$, $v_m = \left\langle \vec{S}_i \vec{S}_j H^m \right\rangle_{T=\infty}$ and

$$\mu_m = \left\langle H^m \right\rangle_{T=\infty}$$

The calculation of the coefficients α_l leads to a diagrammatic representation (Tawardowski et al., 1987), which involves two separate phases:

1. The finding and cataloguing of all diagrams or graphs which can be constructed from one dashed line connecting the site i and j, and l straight lines, and the determination of diagrams whose contribution is nonvanishing. This step has already been accomplished in the Stanley work (Stanley and Kaplan, 1966).
2. Counting the number of times that each diagram can occur in the magnetic system.

In our case, we have to deal with nearest-neighbour coupling J_{ij}.

The coefficient α_l may be expressed for each topological graph as (Hachimi et al., 2003):

$$\alpha_l = \overline{S}^2 (-2\overline{S}^2)^l (J_{ik_1}^{m_1} J_{k_2 k_3}^{m_2} ... J_{k_w j}^{m_v})[\alpha_l] \qquad (7)$$

With the condition $\sum_{r=1}^{v} m_r = l$ for $m_r = 0,1,....,l$. The "weight" $[\alpha_l]$ of each graph is tabulated and given in Stanley and Kaplan (1966) and $k_1, k_2,..., k_w$ represent the sites surrounding the sites i and j.

In an early Tawardowski et al. (1987) work, the coefficient α_l required for the calculation of the three first correlation functions in the case of the f.c.c lattice are given. The topological diagram type, as in SK notation (τ) and the corresponding α_l which are needed to extend the high-temperature series to order $l = 6$ in the case of a zinc, blends structure with n.n.n and n.n.n. interactions.

In Hamedoun et al. (1998), a relation between the susceptibility, correlation length and the three first correlation functions is given in the case face centred cubic lattice with a particular ordering vector $Q = (0,0,k)$. In the ferromagnetic case, we get $k = 0$. The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(T)$ and $\xi(T)$ is that at the neighbourhood of the critical point, the aforementioned two functions exhibit the asymptotic behaviour:

$$\chi(T) = \sum_{m=-n}^{n} \sum_{n=1}^{6} a(m,n) y^m \tau^n \qquad (8)$$

$$\xi^2(T) = \sum_{m=-n}^{n} \sum_{n=1}^{6} b(m,n) y^m \tau^n \qquad (9)$$

where $y = \dfrac{J_2}{J_1}$ and $\tau = \dfrac{2S(S+1)J_1}{k_B T}$. The series coefficients $a(m,n)$ and $b(m,n)$ are available on request. In the spin-glass (SG) region, critical behaviour near the freezing temperature J_{SG} is

expected in the nonlinear susceptibility $\chi_s = \chi - \chi_0$ rather than in the linear part χ_0 of the dc susceptibility χ. This is due to the fact that the order parameter q in the SG state is not the magnetization but the quantity $q = \dfrac{1}{N} \sum_i \left[\langle S_i \rangle^2 \right]_{av}$. As was suggested by Edwards and Anderson, (Hamedoun et al., 2001), leading to an associated susceptibility

$$\chi_s = \frac{1}{NT^3} \sum_{ij} \left[\langle s_i s_j \rangle^2 \right]_{av}$$, where the correlation length of

the correlation function $\left[\langle S_i S_j \rangle^2 \right]$ possibly diverges at $T = T_{SG}$. The behaviour of the nonlinear susceptibility has been already extensively studied theoretically and experimentally (Moron, 1996). We used the expression of χ_s, to determine the critical temperature in the region of spin glass for semimagnetic semiconductors $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$. Figures 1 and 2 shows magnetic phase diagrams of $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$, respectively.

Estimates of T_N and T_{SG}, for $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ have been obtained using the *Padé* approximate method (P.A) (Baker and Gravis-Morris, 1981). A P.A [M,N] to a

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

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

P_M / Q_N, with P_M and Q_N, polynomials of order M and N in

$$\beta = \frac{1}{k_B T},$$ such that:

$$\chi(\beta) \, or \, \xi^2(\beta) \approx P_M / Q_N + O\left(\beta^{M+N+1}\right).$$ The sequence

of [M,N] P.A to be both $\log(\chi(T))$ and $\log(\xi^2(T))$ are found to be convergent; this method is more detailed in Afif et al. (2000). The simple pole corresponds to T_N or T_{SG}.

RESULTS AND DISCUSSION

We have used the experimental values of the Néel temperature $T_N(K)$ and the Curie Weiss temperature $\theta_p(K)$ of $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ (Hamedoun et al., 1998) to derive the values J_1 and J_2, in the range $0.6 \le x \le 1$. The obtained results are given in Tables 1 and 2. In disordered phase, we have used a distribution function of probability adapted of the nature of dilution problem to determine $J_1(x)$ and $J_2(x)$. The obtained
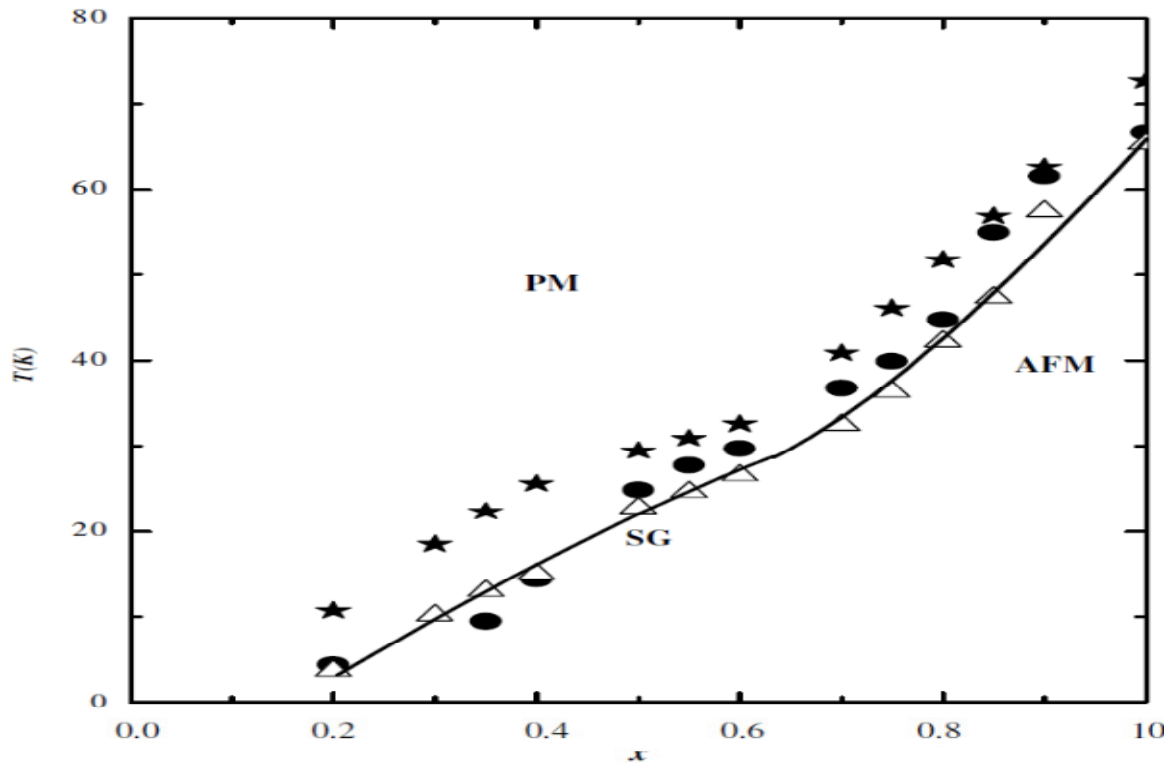
Figure 1. The magnetic phase diagram of $Zn_{1-x}Mn_xTe$. The circles, the stars, represent the experimental data deduced by measurements magnetic and the by replica methods of Moron (1996) and Alba et al. (1982), respectively. The solid line is a guide to the eye.
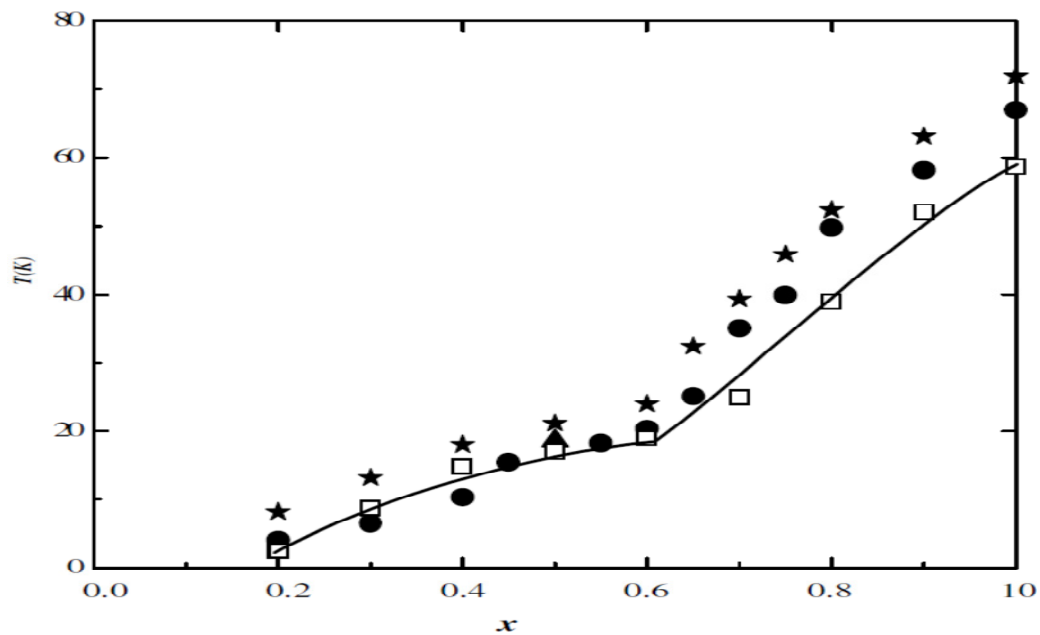

Figure 2. The magnetic phase diagram of $Cd_{1-x}Mn_xTe$. The open squares are the theoretical results. The circles, the triangles, the stars represent the experimental data deduced by measurements magnetic and the by replica method (Moron, 1996; Averous and Balkanski, 1991; Alba et al., 1982), respectively. The solid line is a guide to the eye.

values of J_1 for $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ are comparable to those given by (Hamedoun et al., 2003; Roger, 1998) (J_1= -6.3 K for $Cd_{1-x}Mn_xTe$ and -9.5 K for $Zn_{1-x}Mn_xTe$) and (J_1 = -6.9 ± 0.15 K for $Cd_{1-x}Mn_xTe$ and -11.85 ± 0.25 K for $Zn_{1-x}Mn_xTe$) (Furdyna, 1988). The sign of $J_1(x)$ and $J_2(x)$ are negative in the range $0.6 \leq x \leq 1$ and are antiferromagnetic in the range $0.6 \leq x \leq 1$ (Tables 1 and 2). The high-temperature series expansions (HTSEs) 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 $\chi(T)$, we have estimated the Néel temperature $T_N(K)$ for each dilution x. The obtained magnetic phase diagrams of $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems are presented in Figures 1 and 2, respectively. Several thermodynamic phases may appear including the paramagnetic (PM), antiferromagnetic (AFM) $0.6 \leq x \leq 1$ and spin-glass (SG) phase $x < 0.6$. The percolation threshold obtained for two systems is $x_c \approx 0.2$, is the critical concentration for the appearance of an infinite percolative cluster produced by the first nn hops only. This value of percolation threshold is comparable with those given by Roger (1998) ($x_c \approx 0.19$). In the $0 \leq x \leq 0.2$ region, $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ have behaviour paramagnetic for all values of the critical temperature. In these figures, we have included, for comparison, the experimental results obtained by magnetic measurement. From these figures, one can see good agreement between the theoretical phase diagram and experimental results. In addition, we have determined the region spin glace while using the expression of the nonlinear susceptibility.

Conclusions

The obtained values of exchange interactions J_1 in $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ are comparable to those given by Hamedoun et al. (2003) and Roger (1998). The sign of $J_1(x)$ and $J_2(x)$ is negative the range $0.6 \leq x \leq 1$. The high-temperature series expansions and *Padé* approximants are applied to the magnetic susceptibility $\chi(T)$, to find the Néel temperature $T_N(K)$ for each dilution x. The magnetic phase diagrams of $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ systems are obtained. The appearance of the SG state in the intermediate region

of dilution in $Zn_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ is due to the strong competition between the FM and the AFM interactions (Galazka et al., 1980). The theoretical phase diagram and experimental results are comparable (Figures 1 and 2).

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