

## Magnetic properties of diluted magnetic semi conductors and $\text{Cd}_{0.5}\text{Mn}_{0.5}\text{Te}$ nanoparticle

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**Abstract** - The magnetic properties of diluted magnetic semi conductors (DMS)  $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$  are investigated. We have evaluated the Néel temperature from the nearest neighbour  $J_1(L)$  interactions and the energy exchange for the different diameter of the  $\text{Cd}_{0.5}\text{Mn}_{0.5}\text{Te}$  nanoparticle by using the mean field theory. The shifts of the Néel temperatures  $T_N(L)$  from the bulk value  $\left[ \frac{T_N(\infty)}{T_N(L)} - 1 \right]$  can be described by a power law  $L^{-\lambda}$ , where  $\lambda = \frac{1}{\nu_b}$  is the inverse of the correlation length exponent. The obtained value is  $\nu_b = 1.1 \pm 0.1$ . The magnetic phase diagrams have been determined by the High-temperature series expansions. 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  $\text{Mn}$  magnetic ions arising from the topology of the DMS alloys. The critical exponents associated with the magnetic susceptibility ( $\gamma$ ) and with the correlation lengths ( $\nu$ ) are deduced.

**Keywords:** Diluted magnetic semi conductors; Néel temperature; Nanoparticle; Critical exponents; High-temperature series expansions; Padé approximants, Phase diagram.

### 1. Introduction

The diluted magnetic semiconductors (DMS) are compounds based on typical semiconductors (like  $\text{CdTe}$  or  $\text{InAs}$ ), for which a fraction of nonmagnetic cations has been replaced by magnetic ions (typically transition metal ions like  $\text{Mn}$ ,  $\text{Fe}$  or rare earth metal ions) [1] DMS bridge the physics of semiconductors and magnetic since they show typical semiconductor behaviour and they also reveal pronounced magnetic properties.  $\text{Cd}_{1-x}\text{Mn}_x\text{Te}$ , Zinc-blende structure DMS alloys are the most typical

representative of DMS. They can be considered as mixed crystal systems between two Zinc-blende phase materials,  $CdTe$  ( $ZnTe$ ) and  $MnTe$ . In particular magnetism of these materials is typical for magnetic ions possessing a spin momentum  $S = \frac{5}{2}$ , which would correspond to  $Mn^{2+}$  ( $d^5$ ) centers. The magnetic phase diagrams of the above two systems consist of two regions: a high-temperature, paramagnetic phase and low temperature frozen phase. The latter phase generally occurs when  $x > 0.2$ , but recent works has shown that spin freezing can also occurs for lower values of  $x$  at very low temperature [2,3]. The pure system  $MnTe$  presents a truly long-range, type III antiferromagnetic (AFM) ordering [4]. We have used the mean field theory, for calculated the Néel temperature from the nearest neighbour  $J_1$  [5] interactions and the energy exchange for the different diameter of the  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle. The shifts of the Néel temperatures  $T_N(L)$  from the bulk value  $\left[ \frac{T_N(\infty)}{T_N(L)} - 1 \right]$  can be described by a power law  $L^{-\lambda}$ , where  $\lambda$  is the inverse of the correlation length exponent.

Another part of this paper concerns the interesting topic of magnetic structure and spin glass behaviour in the diluted magnetic semiconductors  $Cd_{1-x}Mn_xTe$  with  $0 \leq x \leq 1$ . The Padé approximant (P.A) [6] 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 [7,8]. We have used this technique to determine the phase diagrams and the critical exponents  $\gamma$  and  $\nu$  associated with the magnetic susceptibility  $\chi$  and the correlation length  $\xi$  in the range  $0 \leq x \leq 1$ , respectively.

## 2. Néel temperatures and critical exponent's calculations

### a. Mean field approximation

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

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

where,  $J_{ij}$  is the exchange integral between the spins situated at sites  $i$  and  $j$ .  $\vec{S}_i$  is the atomic spin of the magnetic ion located on the  $i$ th site. The factor "2" in Eq. (1) arises from the fact that, when summing over all possible pairs  $\langle ij \rangle$  exchange interactions, we count each pair twice. The mean field approximation leads to a simple relations between the Néel temperature  $T_N$ , respectively, and the considered one exchange integral  $J_1$ .

Following, the method of Holland and Brown [9], the expression of  $T_N(K)$  is:

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

where  $k_B$  is the Boltzmann's constant and  $S = \frac{5}{2}$ .

Using the experimental values  $J_1(L)$  obtained by magnetic measurement [5] for the  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle. We have deduced the values of Néel

temperature  $T_N(L)$ . From these values, we have derived the energy exchange, for different diameter of nanoparticle. The obtained results are given in table 1. The temperature shift has been observed in numerous experimental studies and it has also been investigated in theory [10-14]. Specifically, the Néel temperature  $T_N$  must be regarded as a diameter-dependent parameter,  $T_N(L)$  which approaches the bulk Néel temperature  $T_N(\infty)$  as the scale factor  $L$

Table 1: The first exchange integrals, the Néel temperature  $T_N(K)$ , and the energy for different diameter of  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle.

$L(nm)$	$-\frac{J_1}{K_B}(K)[5]$	$T_N(K)$	$\left \frac{E}{S^2}(K)\right $
bulk	8.1	189.0	97.12
8.0	7.8	182.0	93.6
7.2	6.9	161.0	82.72
6.7	8.9	207.6	106.72

(the diameter of nanoparticle) approach  $\infty$ . It has been shown that the approach of  $T_N(L)$  to  $T_N(\infty)$  can also be described by a simple power law [15] characterized by a shift exponent  $\lambda$  defined by:

$$\frac{T_N(L)}{T_N(\infty)} - 1 \propto L^{-\lambda} \quad (3)$$

The shift exponent  $\lambda$  is given by  $\lambda = \frac{1}{\nu_b}$  where  $\nu_b$  is the correlation length critical exponent.

In Figure 1, we exhibit the dependence of the shift  $\delta T = \frac{T_N(\infty) - T_N(L)}{T_N(L)}$  whit  $L(nm)$  in a Log- Log scale to determine the exponent  $\lambda$  by using equation (3) for Heisenberg model. The obtained values for the  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle is  $\nu_b = 1.1 \pm 0.1$ .

## b. High-temperature series expansions

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

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

where  $\beta = \frac{1}{k_B T}$  and  $N$  is the number of magnetic ion.  $\langle S_i S_j \rangle = \frac{Tr S_i S_j e^{-\beta H}}{Tr e^{-\beta H}}$  is the correlation function between spins at sites  $i$  and  $j$ . The expansion of this function in powers of  $\beta$  is obtained as follows [16]:

$$\langle \vec{S}_i \vec{S}_j \rangle = \sum_{l=0}^{\infty} \frac{(-1)^l}{l!} \alpha_l \beta^l \quad (5)$$

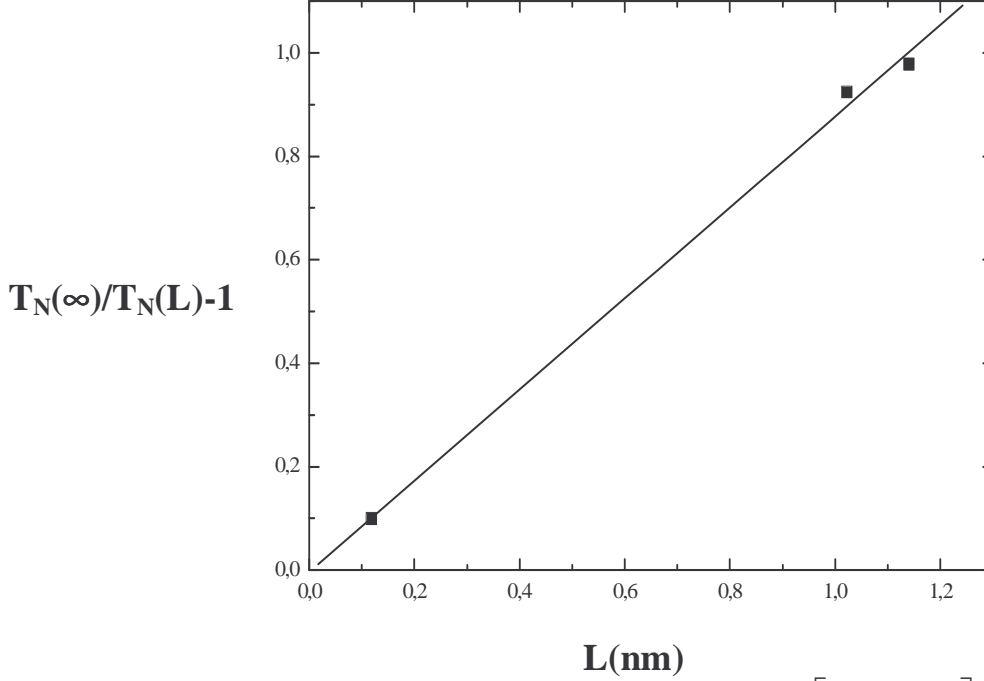


Fig. 1. Log-Log plot of the shift of reduced Néel temperature  $\left[ \frac{T_N(\infty)}{T_N(L)} - 1 \right]$  versus diameter  $L(nm)$  of  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle.

The calculation of the coefficients  $\alpha_l$  leads to a diagrammatic representation [17], which involves two separate phases:

- 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.
- 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  $\alpha_l$  may be expressed for each topological graph as [16]:

$$\alpha_l = \bar{S}^2 (-2\bar{S}^2)^l (J_{ik_1}^{m_1} J_{k_2 k_3}^{m_2} \dots J_{k_w j}^{m_w}) [\alpha_l] \quad (6)$$

with the condition  $\sum_{r=1}^v m_r = l$  for  $m_r = 0, 1, \dots, l$ . The “weight”  $[\alpha_l]$  of each graph is tabulated and given in Ref. [18] and  $k_1, k_2, \dots, k_w$  represent the sites surrounding the sites  $i$  and  $j$ . In [8], 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 high temperature series expansion of  $\chi(T)$  and  $\xi^2$  are given by:

$$\chi(T) = \sum_{m=-n}^n \sum_{n=1}^6 a(m, n) y^m \tau^n \quad (7)$$

$$\xi^2(T) = \sum_{m=-n}^n \sum_{n=1}^6 b(m,n) y^m \tau^n \quad (8)$$

where  $y = \frac{J_2}{J_1}$  and  $\tau = \frac{2S(S+1)J_1}{k_B T}$ . The values of exchange interactions used are  $J_1 = -13k_B$  and  $J_2 = -4k_B$  given in [19]. The series coefficients  $a(m,n)$  and  $b(m,n)$  are given in [20]. In spin-glasses (S.G) critical behaviour near the  $T_{SG}$  SG transition, it is expected not in the linear part  $\chi_0$  of the dc susceptibility  $\chi$ , but in the nonlinear susceptibility  $\chi_s = \chi - \chi_0$ . This is due to the fact that the order parameter  $q$  in the spin glass state is not the magnetization but the quantity  $q = \frac{1}{N} \sum_i \left[ \langle S_i \rangle^2 \right]_{av}$ . As suggested by Edwards and Anderson [21], 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  $\left[ \langle S_i S_j \rangle \right]$  possibly diverges at  $T = T_{SG}$ . The behaviour of the nonlinear susceptibility has been already extensively studied theoretically and experimentally [22, 23]. We have used the expression of  $\chi_s$ , to determinate the freezing temperature  $T_{SG}$  in the region of spin glass for DMS  $Cd_{1-x}Mn_xTe$ . Figure 2 shows, the magnetic phase diagram of DMS  $Cd_{1-x}Mn_xTe$  nanoparticle. We can see the good agreement between the magnetic phase diagrams obtained by the HTSE technique and the experimental ones, in particular in the case of the last systems of which the phase diagrams have been established well by different methods [24-25]. The results given by the HTSE method are comparable with the experimental points that the results deals by the replica method [26].

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

$$\chi(T) \propto (T - T_N)^{-\gamma} \quad (9)$$

$$\xi^2(T) \propto (T_N - T)^{-2\nu} \quad (10)$$

Estimates of  $T_N$ ,  $\nu$  and  $\gamma$  for  $Cd_{1-x}Mn_xTe$  have been obtained using the Padé approximate method (P.A) [6] in the range  $0 \leq x \leq 1$ . The simple pole corresponds to  $T_N$  and the residues to the critical exponents  $\gamma$  and  $\nu$ . The obtained central values are  $\gamma = 1.4 \pm 0.1$  and  $\nu = 0.9 \pm 0.1$ .

#### 4. Discussions and conclusions

We have used the experimental value of  $J_1(x=0.5)$  [5] to derive the Néel temperature  $T_N$  and the energy exchange of the magnetic structure for different diameter of  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle. The results obtained are given in table 1. On the other hand, according to the universality hypothesis, critical phenomena can be described by quantities that do not depend on the microscopic details of the system, but only on global properties such as the dimensionality and the symmetry of the order parameter. It has been a point of interest to see the influence of exchange coupling on the behavior of the critical exponent  $\nu_b = \frac{1}{\lambda}$  associated with the magnetic  $\frac{T_N(L)}{T_N(\infty)} - 1 \propto L^{-\lambda}$ , of the  $Cd_{0.5}Mn_{0.5}Te$  nanoparticle. These values are nearest of the Heisenberg model. The high-temperature series expansion (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  $\chi(T)$  we have estimated the critical temperature  $T_N$  (or  $T_{SG}$ ) for each dilution  $x$ . The obtained magnetic phase diagram of the DMS  $Cd_{1-x}Mn_xTe$  system is presented in figure 2. Several thermodynamic phases may appear including the paramagnetic (PM), antiferromagnetic (AFM)  $0.6 \leq x \leq 1$  and spin-glass (SG) phase in range

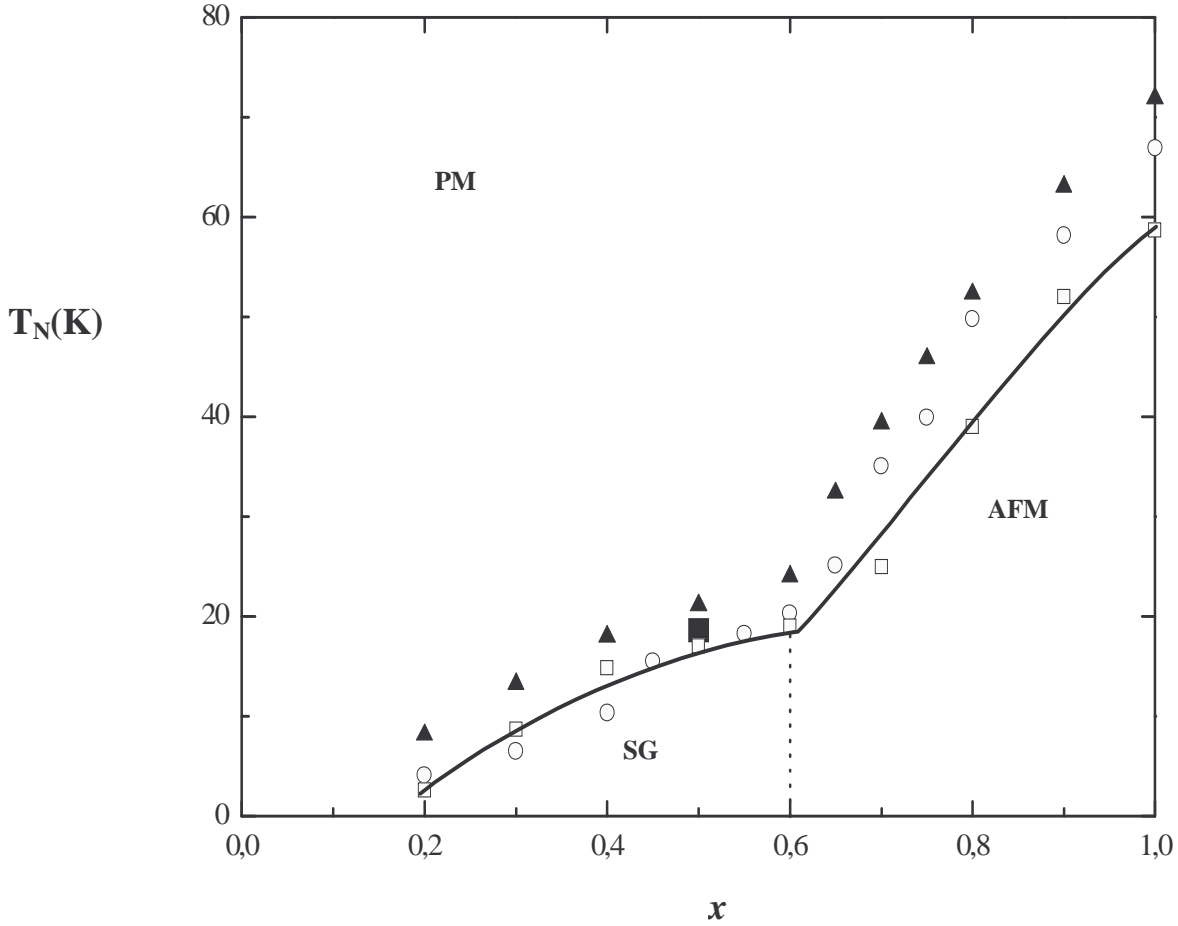


Figure 2. The magnetic phase diagram of  $Cd_{1-x}Mn_xTe$ . The various phases are the paramagnetic phase (PM), antiferromagnetic phase (AFM) ( $0.6 \leq x \leq 1$ ), and the spin glass phase (SG) ( $0.2 \leq x < 0.6$ ). The open squares are the theoretical results. The open circle, the solid square, the solid triangle represent the experimental points deduced by measurements magnetic and the by replica method [27], [5] and [26], respectively.

$0 \leq x < 0.6$ . The percolation threshold obtained  $x_c \approx 0.2$ , is the critical concentration for the appearance of an infinite percolative cluster produced by the first  $mn$  hops only. This value is comparable with  $x_c = 0.19$  obtained by [28]. In this figure we have included, for comparison, the experimental results obtained by magnetic measurement. From this figure one can see good agreement between the theoretical phase diagram and experimental results. In addition, we have determined the region spin glass while using the expression of the nonlinear susceptibility. In the other hand, the values of the critical exponents  $\gamma$  and  $\nu$  associated with the magnetic susceptibility  $\chi(T)$  and correlation length  $\xi(T)$ , respectively, have been estimated in the range of the composition  $0.6 \leq x \leq 1$ . 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 susceptibility and correlation length are found to be  $\gamma = 1.4 \pm 0.1$  and  $\nu = 0.9 \pm 0.1$ . These values are insensitive to dilution  $x$ .

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