Study of the Magnetic Property in the Systems $K_2Cu_xMn_{1-x}F_4$

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The exchange interactions are calculated by using the probability law. The high-temperature series expansions have been applied to the $K_2Cu_xMn_{1-x}F_4$ systems, combined with the Padé approximants method in order to determine the magnetic phase diagram, i.e., T_C versus dilution x. The critical exponent associated with the magnetic susceptibility (γ) is deduced. The obtained value of γ is insensitive to the dilution ratio x and may be compared with other theoretical results based on the XY model.

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I. INTRODUCTION

The magnetic properties of randomly mixed spin systems have been investigated extensively in the last few decades [1–10]. The two materials most extensively studied are K_2NiF_4 [11] and K_2MnF_4 [12]. In these materials there is a single-ion anisotropy term in the Hamiltonian with a magnitude of 0.2% and 0.4%, respectively, relative to the Heisenberg term. This term acts to align the spins during z behavior, so the materials would be expected to crossover to Ising behavior close to the point. The $K_2Cu_xMn_{1-x}F_4$ systems are mixed materials of K_2MnF_4 and K_2CuF_4. The magnetic structure of K_2NiF_4 is antiferromagnetic with the Néel temperature $T_N = 42.14$ K [13], denoting a body-centered tetragonal sub-lattice of magnetic Cu or Mn atoms. The magnetic structure of K_2CuF_4 is a two-dimensional ferromagnetic phase with $T_C = 6.2$ K [14] and K_2MnF_4, which is a two-dimensional antiferromagnetic with $T_N = 42.1$ K. The magnetic measurements for $K_2Cu_xMn_{1-x}F_4$ revealed the ferromagnetic ($0.8 \leq x \leq 1$), spin glass ($0.5 \leq x < 0.8$), and antiferromagnetic ($0 \leq x < 0.5$) phases [12]. The determination of the critical exponents is an important aspect of the theoretical description and experimental characterization of magnetic materials [15]. In this work, the values of the $J_{AB}(x)$ exchange interactions are calculated via a probability law for the systems $K_2Cu_xMn_{1-x}F_4$ in the range $0 \leq x \leq 1$. The Padé approximant (PA) [16] analysis of the high-temperature series expansions (HTSE) of these correlation functions has been shown to be a useful method for the study of the critical region [17, 18]. In the antiferromagnetic region we use this technique for the magnetic susceptibility to determine the Néel temperature, using the results given in Ref. [19], and in

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the ferromagnetic region we have also used the results regarding the magnetic susceptibility and the correlation length as given in Ref. [20]. The critical exponent associated with the magnetic susceptibility in the ferromagnetic region is given.

II. CALCULATION OF THE VALUES OF THE EXCHANGE INTEGRALS

In the previous work, the authors used the probability law to calculate the exchange integrals [21]. In this work, we have applied the same probability law in the diluted system $B_2A_xA'_{1-x}X_4$, the random placement of the ions A and B leads to the spatial fluctuations of the signs and the magnitudes of the super-exchange interaction between the magnetic ions A and B. Due to the nature of the dilution problem we choose a probability law permitting us to determine the exchange integral $J_{AB}(x)$ for each concentration of x. The exchange integrals of the opposite pure compound B_2AX_4 of the bound random body-centered tetragonal is denoted J_{AB}. The occupation probability $p(i)$ of the two ions A and B induced in the interaction is $p(i) = C_n^i x^{n-i}(1-x)^i$, where n is considered the number of cations situated in tetrahedral sites at the same distance from $n = 3$ while i varies from 0 to 3. The exchange integral for such an occupation is assumed to be $J_{AB}^i = (J_A^{n-i} J_B^i)^{1/n}$. The expression obtained is

$$J_{AB}\left(x\right) = \sum_{i=0}^{3} C_3^i x^{3-i}(1-x)^i (J_A^{3-i} J_B^i)^{1/3},$$

$$J_{AB}\left(x\right) = \left(x^3 + 3x^2\left(1-x\right) + 3x\left(1-x\right)^2 + \left(1-x\right)^3\right) J_{AB}. \tag{1}$$

J_{AB} corresponds to the exchange interactions of the opposite pure systems B_2AX_4.

$K_2Cu_xMn_{1-x}F_4$ are the diluted of K_2MnF_4 systems, where the different values of the exchange integral J_{AB} are given by Ref. [22].

III. HIGH-TEMPERATURE SERIES EXPANSIONS

In order to deduce the expression of the susceptibility of the system with two sub-lattices, the Hamiltonian of the system with an external field h_{ex} may be put in the form

$$H = -2J_{AA}\sum_{\langle i,\grave{i}\rangle} \vec{S}_i\vec{S}_{\grave{i}} - 2J_{BB}\sum_{\langle j,\grave{j}\rangle} \vec{\sigma}_j\vec{\sigma}_{\grave{j}} - 2J_{AB}\sum_{\langle i,j\rangle} \vec{S}_i\vec{\sigma}_j - \mu_B h_{ex}\Big(g_A\sum_i S_i^z - g_B\sum_j \sigma_j^z\Big), \tag{2}$$

where $\vec{S}$ and $\vec{\sigma}$ are the spin operators of the ions in the sublattices A and B, respectively. g_A and g_B are the corresponding gyromagnetic factors. The symbol $\langle ...\rangle$ denotes summation over the nearest neighbors. J_{AA}, J_{BB}, and J_{AB} are the intra-and the inter-sublattice exchange interactions.

The magnetization of the antiferromagnetic system is

$$M = \mu_B\Big(g_A \sum_i \langle S_i^z\rangle - g_B \sum_j \langle \sigma_j^z\rangle\Big) \tag{3}$$

After computing the first derivation of the magnetization $\chi = \left(\frac{\partial M}{\partial h_{ex}}\right)_{h_{ex}\to 0}$, we obtained the general expression of the susceptibility for the collinear normal antiferromagnetic body-centred tetragonal as

$$\chi = \frac{\mu_B^2}{3k_BT}\left(N_A g_A^2 \bar{S}^2 + N_B g_B^2 \bar{\sigma}^2 - g_A^2 \sum_{i\neq \grave{i}} \left\langle \vec{S}_i \vec{S}_{\grave{i}}\right\rangle - g_B^2 \sum_{j\neq \grave{j}} \left\langle \vec{\sigma}_j \vec{\sigma}_{\grave{j}}\right\rangle - 2g_A g_B \sum_{i,j} \left\langle \vec{S}_i \vec{\sigma}_j\right\rangle\right) \tag{4}$$

where N_A, N_B, $S = \frac{5}{2}$, and $\sigma = \frac{3}{2}$ are respectively the number of ions and the spin value of each type of spin.

Finally, we obtain the simple form

$$\chi = \frac{\mu_B^2}{3k_BT}\left(N_A g_A^2 \bar{S}^2 + N_B g_B^2 \bar{\sigma}^2 - N_A g_A^2 \gamma_{AA} - N_B g_B^2 \gamma_{BB} - 2N_B g_A g_B \gamma_{BA}\right) \tag{5}$$

Following the procedure in [23–28], we compute the expressions for the spin correlation functions γ_{AA}, γ_{BB}, and γ_{AB} in terms of powers of β.

The correlations functionsγ_{AA}, γ_{BB}, and γ_{AB} may be expressed as follows:

$$\begin{aligned}
\gamma_{AA} &= \bar{S}^2 \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} a\,(m,n,p,q)\; J_1^m J_2^n J_3^p \beta^q,\\
\gamma_{BB} &= \bar{\sigma}^2 \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} b\,(m,n,p,q)\; J_1^m J_2^n J_3^p \beta^q,\\
\gamma_{BA} &= \bar{S}\bar{\sigma} \sum_{q=1}^{7} \sum_{m=0}^{q} \sum_{n=0}^{q-m} \sum_{p=0}^{q-m-n} c\,(m,n,p,q)\; J_1^m J_2^n J_3^p \beta^q,
\end{aligned} \tag{6}$$

with $J_1 = 2J_{BB}\,\bar{\sigma}^2$, $J_2 = 2J_{AB}\,\bar{S}\bar{\sigma}$, $J_3 = 2J_{AA}\,\bar{S}^2$, $\bar{S} = \sqrt{S(S+1)}$, and $\bar{\sigma} = \sqrt{\sigma(\sigma+1)}$.

The nonzero coefficients $a(m,n,p,q)$, $b(m,n,p,q)$, and $c(m,n,p,q)$ up to order 7 in β are given in Ref. [19]. In the ordered region, we have used the expression and results of the magnetic susceptibility and the correlation length obtained by Ref. [20]. In the spin-glass (SG) region, critical behavior 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 $q = \frac{1}{N}\sum_i \left[\langle S_i\rangle^2\right]_{av}$, as was suggested by Edwards and Anderson [29], 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 $T = T_{SG}$. We use the powerful

PA method [16] to estimate the critical temperature. In this method, the Curie point is determined by locating the singularities in the PA method applied to the HTSEs of the magnetic susceptibility. The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(T)$ is that in the neighborhood of the critical point the following functions exhibit the asymptotic behavior

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

The usual approach is to compute the series in order to obtain the logarithmic derivative of $\chi\,(T)$,

$$\frac{d}{dT} \log\left[\chi\,(T)\right] \approx \frac{-\gamma}{T - T_c}, \tag{8}$$

as this function has a simple pole $T_C(T_N)$ or T_{SG} and should be well represented by the Padé approximants $[M,N]$. The exponent γ is then re-estimated from the approximates to be

$$(T - T_c)\,\frac{d}{dT} \log\left[\chi\,(T)\right] \tag{9}$$

evaluated at $T = T_N(T_C \text{ or } T_{SG})$.

Estimates of $T_C(T_N)$ or T_{SG}, and γ for $K_2Cu_xMn_{1-x}F_4$ have been obtained using the PA method [16]. The simple pole corresponds to $T_C(T_N)$ or T_{SG}, and the residues correspond to the critical exponents γ. The obtained central value of γ in the ferromagnetic region is $\gamma = 1.32 \pm 0.02$.

IV. DISCUSSIONS AND CONCLUSIONS

In this work, we have used a distribution of probability adapted from the nature of the dilution problem to determine the J_{AB} exchange interactions for the systems $K_2Cu_xMn_{1-x}F_4$. The obtained values are given in Table I in the range of $0 \leq x \leq 1$. From Table I one can see that the variation of $J_{AB}(x)$ depends on the phase region and the value of J_{AB} in the pure case.

The HTSE extrapolated via the Padé approximants method is shown to be a convenient method for providing valid estimations of the critical temperatures for a real system. By applying this method to the magnetic susceptibility $\chi\,(T)$, we have estimated the critical temperature $T_C(T_N)$ and the T_{SG} (in the spin glass region) for each dilution of x in the systems $K_2Cu_xMn_{1-x}F_4$. In Figs. 1 to 5, we have presented the magnetic phase diagrams of $K_2Cu_xMn_{1-x}F_4$, with different values for J_{AA}, J_{AB}, and J_{BB}. Several thermodynamic phases may appear including the paramagnetic (PM), ferromagnetic (FM), antiferromagnetic (AFM), and the spin glass phases (SG). For $J_{AA} > 0$ and $J_{BB} < 0$ all phases mentioned above may appear (see Figs. 1 and 2). The spin glass phase moves towards higher values of x when J_{AB} decreases. For $J_{AA} < 0$, $J_{BB} < 0$, and small J_{AB},

TABLE I: The value of exchange interaction $J_{AB}(x)$ for different values of J_{AB} for the systems $K_2Cu_xMn_{1-x}F_4$.

x	$J_{AB}(x)(J_{AB}=0.81)$	$J_{AB}(x)(J_{AB}=-0.81)$	$J_{AB}(x)(J_{AB}=0.17)$	$J_{AB}(x)(J_{AB}=0.24)$
1	0.81	-0.81	0.17	0.24
0.9	0.613	-0.613	0.128	0.181
0.8	0.498	-0.498	0.104	0.147
0.7	0.452	-0.452	0.095	0.134
0.6	0.460	-0.460	0.096	0.136
0.5	0.506	-0.506	0.106	0.150
0.4	0.576	-0.576	0.121	0.170
0.3	0.656	-0.656	0.137	0.194
0.2	0.732	-0.732	0.153	0.216
0.1	0.788	-0.788	0.165	0.233

only an antiferromagnetic phase takes place at low temperature (see Fig. 3), the spin glass phase (SG) appears above a critical value of the exchange integral J^*_{AB} (see Fig. 4, in the case of $J_{AA} = -1$ and $J_{BB} = -0.66$, J^*_{AB} is closer to 0.179). These results are obtained with the approximated mean field analysis [1]. It is known that the occurrence of the SG phase depends on a kind of quenched randomness, in addition to the symmetry of spin and the range of the interactions [10]. Fig. 5 confirms that the range of concentration x where SG exists increases with J_{AB}. These results are in rough qualitative agreement with those deduced from phase diagrams of a number of real systems; the competition between different interactions contributes to the appearance of a SG phase. The range of concentrations where this phase appears depends on the degree of frustration. For fixed values of J_{AA} and J_{BB} the nature of the phase diagrams depends on the spin number and on the sign and the strength of J_{AB}. It is worth noting that if $J_{AA} > 0$ and $J_{BB} > 0$, the AFM and FM phases exchange their role, and the SG phase appears for a negative threshold of $-J^*_{AB}$. It can be observed that in our model we deal with classical spins. The results obtained by the HTSE method are comparable with those given by the replica method [22]. In Fig. 6, we presented the magnetic phase diagram of a planar mixed system $K_2Cu_xMn_{1-x}F_4$. In this system, we cross the gap between the opposite pure compounds K_2MnF_4 and K_2CuF_4. We took values for the exchange interactions ($J_{Cu-Cu} = 11.4$ K, $J_{Mn-Mn} = -4.2$ K, and $J_{Cu-Mn} = -5.05$ K) [30–32]. The spin glass phase limits are situated on Cu-concentrations $x^*_c \approx 0.5$ and $x^*_c \approx 0.85$. These values are predicted by magnetic measurements [33] and by the replica method [22]. On the other hand, the value of the critical exponent γ associated with the magnetic susceptibility $\chi(T)$, have been estimated in the range order. The sequence of [M,

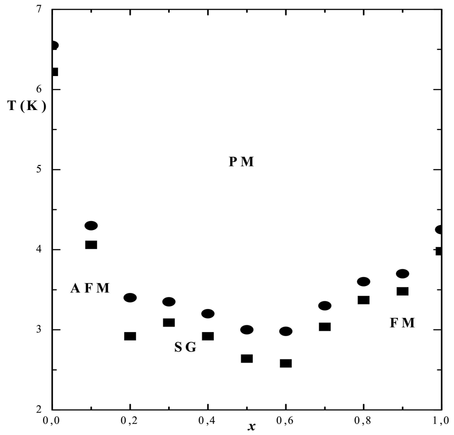

FIG. 1: Magnetic phase diagram with $J_{AA} = 1$ K, $J_{BB} = -0.66$, and $J_{AB} = 0.81$ K. The circles represent the results given by the HTSE method, and the squares represent the results given by the replica method [22].

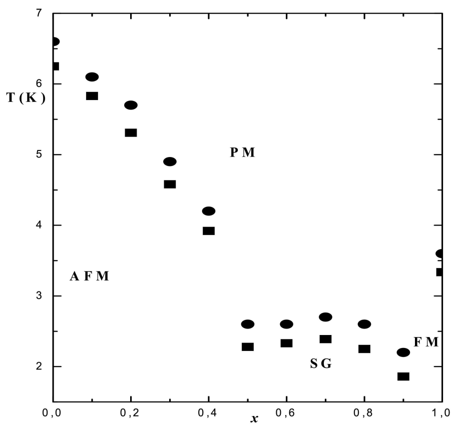

FIG. 2: Magnetic phase diagram with $J_{AA} = 1$ K, $J_{BB} = -0.66$, and $J_{AB} = -0.81$K. The circles represent the results given by the HTSE method, and the squares represent the results given by the replica method [22].

N] PA to the series of $\chi(T)$ has been evaluated. By examining the behavior of these PA values, the convergence was found to be quite rapid. Estimates of the critical exponents associated with the magnetic susceptibility for the $K_2Cu_xMn_{1-x}F_4$ systems are found to be $\gamma = 1.32 \pm 0.02$. On the other hand, the values of the critical exponents γ associated with the magnetic susceptibility $\chi(T)$ have been estimated in the ordered region and for several [M, N] PA. The convergence is extremely good, and from the elements near to and on the diagonal of PA [M, N], we were able to estimate the central value of the critical exponents

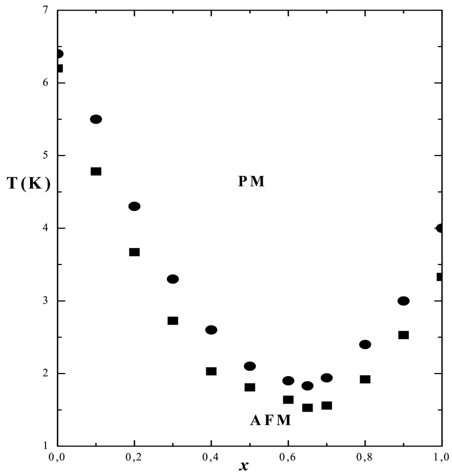

FIG. 3: Magnetic phase diagram with $J_{AA} = -1$ K, $J_{BB} = -0.66$, and $J_{AB} = 0.17$ K. The circles represent the results given by the HTSE method, and the squares represent the results given by the replica method [22].

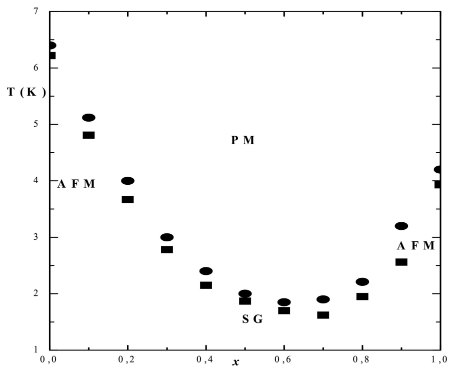

FIG. 4: Magnetic phase diagram with $J_{AA} = -1$ K, $J_{BB} = -0.66$, and $J_{AB} = 0.24$ K. The circles represent the results given by the HTSE method, and the squares represent the results given by the replica method [22].

in the ferromagnetic region: $\gamma = 1.32 \pm 0.02$. This value is comparable with those given by [34] and may be compared with other theoretical results based on the XY model.

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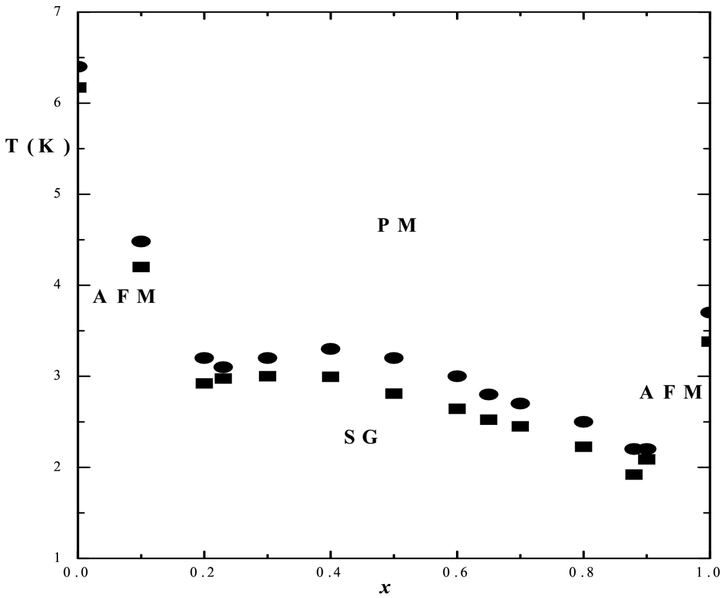

FIG. 5: Magnetic phase diagram with $J_{AA} = -1$ K, $J_{BB} = -0.66$, and $J_{AB} = 0.81$ K. The circles represent the results given by the HTSE method, and the squares represent the results given by the replica method [22].

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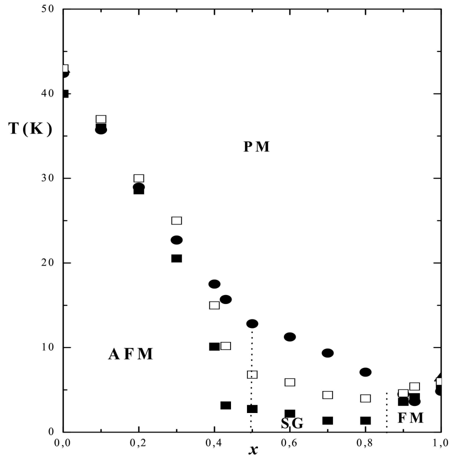

FIG. 6: The magnetic phase diagram of the $K_2Cu_xMn_{1-x}F_4$ systems. The circles, the triangles, the solids squares and the opens solids represents the results given by: replica method [22], experiment results [13,35], the results given by Monte Carlo simulations [36] and the results given by HTSE method, respectively.

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