Site-bond correlations in the three-dimensional Heisenberg model: Application to $KNi_pMg_{1-p}F_3$

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Site-bond-correlated dilution pictures have been successfully proposed to account for the striking differences experimentally observed by nuclear-magnetic-resonance (NMR) measurements between random-diluted $KNi_pMg_{1-p}F_3$ and the isostructural system $KMn_pMg_{1-p}F_3$. The present work makes use of a variational principle for the free energy to treat the critical behavior of the system $KNi_pMg_{1-p}F_3$ in the three-dimensional Heisenberg model by considering the interaction of the nearest neighbor and next-nearest neighbor in the spin system. The calculated phase diagram in the T-p plane for the Heisenberg model is in good accordance with experimental data obtained by NMR, and the value of the correlation parameter is estimated. [S0163-1829(97)04842-X]

For many years physicists tried to understand the collective behavior of magnets and other condensed matter systems by studying the pure materials, such as crystals with perfect structure and full translational symmetry. However, all materials have some structural disorder in their atomic constitution.[1–3] Therefore, it is natural that an increased understanding of the collective phenomena that occur in magnets materials has arisen from the study of the structurally disordered magnetic systems. Also, disordered systems have intrinsic properties that cannot be treated by employing oversimplified methods. Thus, the literature is filled with different models and their variants. In this paper, we consider the technique usually applied to describe disordered magnetic materials, namely, the dilution picture.

The effect of diluting a magnetic system with nonmagnetic impurities has attracted the attention of many researchers and, in the last several years, the theoretical and experimental study of randomly diluted magnetic systems has been intensively investigated such that an enormous increase of work, in the literature, has arisen from these studies.[1–10]

The uncorrelated diluted model is well known and suited to describe the critical properties of certain magnets.[4–10] In this model, some magnetic atoms on the lattice are randomly replaced by nonmagnetic atoms and a bond connecting each pair of occupied magnetic first neighbors is modified. In this context, the diluted magnetic systems are usually associated with an uncorrelated percolation problem when the temperature goes to zero. Particular interest has been given to the phase diagram for the critical temperature vs concentration. However, recent experiments have shown that the introduction of nonmagnetic species in some magnetic materials displays effects not predicted by the usual dilution model (i.e., neither bond nor site dilution). In these materials the correlation is relevant to describe the system and therefore such effects must be included. Thus, other models have been proposed to take into account the correlation effects. Examples of this approach to alternative site-dilution models are the bootstrap percolation[11,12] and dilution, high-density percolation,[11,13] and, more recently, the site-bond-correlated (SBC) dilution model.[14]

SBC is a recent dilution model proposed by de Aguiar and co-workers[14] to explain the differences, observed by nuclear magnetic resonance, in the phase diagram T_c vs p between the diluted Heisenberg antiferromagnet $KMn_pMg_{1-p}F_3$ and the isostructural system $KNi_pMg_{1-p}F_3$. The experiments have shown that for the randomly diluted Heisenberg magnetic compound $KNi_pMg_{1-p}F_3$, T_c decreases faster than for isostructural $KMn_pMg_{1-p}F_3$ when the concentration decreases. This happens because the latter system is more adequately described by an uncorrelated site-dilution picture. Furthermore, the phase diagram in the T-p plane for $KNi_pMg_{1-p}F_3$ displays an upward curvature in contrast to the compound $KMn_pMg_{1-p}F_3$. In the pure materials mentioned above, the Ni^{+2} ions can form only σ bonds, while the Mn^{+2} ions can form both σ and π bonds. The symmetry of the σ bonds suggests that the substitution of a Ni^{+2} ion in $KNi_pMg_{1-p}F_3$ by a nonmagnetic one has a stronger effect on the exchange interaction of a nearest-neighbor magnetic pair situated along the line joining the three atoms than the same effect induced by substitution of a Mn^{+2} ion in $KMn_pMg_{1-p}F_3$. In the SBC model, the coupling between two nearest-neighbor magnetic atoms is assumed to be dependent upon the occupancy of the other nearest-neighbor sites, where a parameter α measures the correlation between the spins.

The SBC model has been studied by means of various methods.[6,7,11,12,14–21] At first, de Aguiar and co-workers[14] studied the SBC model by employing the effective-field theory for the diluted spin Ising model on a square lattice in a cluster with one spin. They found qualitative trends, such

as an increase in the initial slope $\Delta \equiv 1/T_c dT\Delta/dp$, at $p=1$, and an upward curvature in the T-p plane, which is consistent with the experimental data for $KNi_pMg_{1-p}F_3$.[22] They also found three different percolation thresholds at zero temperature. This result, as noted by the authors of Ref. 14, is not consistent with their model Hamiltonian, from which one should obtain only two different thresholds: one for the correlation case and the other for the uncorrelated case. Furthermore, de Aguiar and co-workers[14] obtained values for the critical temperature that is dependent on the correlation parameter. This unphysical result was also not expected. However, da Silva and Moreira[17] studied the critical properties of the SBC model, on a square lattice employing the same scheme of Ref. 14, and obtained good qualitative and quantitative results, for example, finding only two critical concentrations. In 1987, Coutinho *et al.*[18] studied the SBC model, on the Bethe lattice, in a version of an n-vector model obtaining an exact solution. In this work, the behavior of phase diagrams T_c vs p exhibited most of the expected physical properties. The exceptions were in the $T=0$ axis, where only one value of p_c was found for $0 \leqslant \alpha \leqslant 1$, and an upward curvature in the T_c vs p curve even for $\alpha=0$ and the Ising model, which is in contradiction to both effective-field theory[17] and Monte Carlo results.[16,21] These undesired aspects seem to be due to the pathological geometry of the Bethe lattice. Particularly, in the SBC model, where directional effects are important, the fact that it is only in the ''radial'' direction that is uniquely defined for the Bethe lattice may introduce an undesirable bias. More recently, the SBC model has been applied to the three-dimensional Heisenberg model by employing the Migdal-Kadanoff approximation[20] and it has also been studied by the Monte Carlo method for two- and three-dimensional systems.[16,21] In these works, the authors of Refs. 16, 20, and 21 suggest that the value of the phenomenological parameter α for the compound $KNi_pMg_{1-p}F_3$ is above zero. However, neither value has been estimated. The SBC model has also been revisited by the mean-field renormalization-group[23] scheme.[6,7,15] In these works various interesting aspects were observed. For example, a universal relation for the initial slope at $p=1$ between the correlated case and uncorrelated case was obtained.[6] Furthermore, the phase diagrams for this model, using two different exchange interactions in two- and three-dimensional lattices, was presented.[7,15] In both the exchange interactions used, we obtained only two critical concentrations. On the other hand, we recall that in neither of these works mentioned above, the experimental contact with the compound $KNi_pMg_{1-p}F_3$ has been presented.

In the present work, it is our main interest to study the randomly diluted site-bond-correlated Heisenberg model, including both the contribution of the nearest and next-nearest neighbor (NNN) sites in the Hamiltonian that describe the system. The introduction of the effect of the NNN sites resides in the fact that experimental observations for $KNi_pMg_{1-p}F_3$ suggest an apperfeccioned version that correctly describes the spin-lattice relaxation nuclear mechanism in high temperatures.[24–26] The experimental estimative for the value of the exchange interaction between the NNN sites is 0.009 K,[24] where K is the exchange interaction of the nearest-neighbor sites. This same value would be used in the present work.

The Hamiltonian considered is given by

$$H = -\sum_{\langle i,\delta\rangle} J_{i,i+\delta}\mathbf{S}_i\cdot\mathbf{S}_{i+\delta} - \sum_{(i,j)} J^*_{ij}\mathbf{S}_i\cdot\mathbf{S}_j, \tag{1}$$

where $\mathbf{S}_k$ $(k=i,j,\ i+\delta)$ is a spin-1/2 vector operator, with component $\mathbf{S}_k$ along the direction z, $J_{i,i+\delta}$ is the exchange interaction between the nearest-neighbors spins for the ions at sites i and $i+\delta$ on a d-dimensional lattice, defined by

$$J_{i,i+\delta} = J\epsilon_i\epsilon_{i+\delta}[(1-\alpha)\epsilon_{i+2\delta}\epsilon_{i-\delta}+\alpha]. \tag{2}$$

δ is an elementary vector of the lattice and ϵ_i is a random variable that obeys the probability distribution

$$P(\epsilon_i) = p\,\delta(\epsilon_i-1)+(1-p)\,\delta(\epsilon_i), \tag{3}$$

where p is the concentration of magnetic atoms. $J(J/k_BT \equiv K>0)$ is the ferromagnetic coupling constant and α is a parameter governing the correlation. The cases $\alpha=1$ and $\alpha=0$ correspond to the standard uncorrelated and completely correlated site dilution, respectively. On the other hand, J^*_{ij} (with $J^*\equiv\lambda J$, $\lambda>0$) are the next-nearest-neighbor exchange interactions with the probability distribution given by

$$P(J^*_{ij}) = p\,\delta(J^*_{ij}-J^*)+(1-p)\,\delta(J^*_{ij}). \tag{4}$$

In the present work, we present the results of a systematic calculation of the SBC model with first- and second-neighbor interactions based on a variational principle for the free energy.[27]

The critical line obtained is given by

$$p^3\tanh_{1/2}(3K_c)+p(1-p^3)\tanh_{1/2}(3\alpha K_c) = \frac{1}{Z-1-Z^*\lambda}, \tag{5}$$

where $\tanh_{1/2}(x)$ denotes the generalized hyperbolic tangent represented by modified Bessel functions of the first kind defined by

$$\tanh_{1/2}(x) = \frac{I_{3/2}(x)}{I_{1/2}(x)}, \tag{6}$$

where Z is the coordination number and Z^* is the number of NNN on a d-dimensional lattice. Here, we shall use $\lambda=0.009$, $Z=6$ (simple cubic lattice), $Z^*=12$, and $0\leqslant\alpha\leqslant1$.

On the other hand, the initial slope $\Delta \equiv K_c dK_c^{-1}/dp$, at $p=1$, is given by

$$\frac{\Delta(\alpha)}{\Delta(0)} = 1-\frac{2}{3}\frac{\tanh_{1/2}(3\alpha K)}{\tanh_{3/2}(3K)}. \tag{7}$$

This equation presents an interesting relation between $\Delta(\alpha=1)$ and $\Delta(\alpha=0)$, i.e.,[28]

$$\frac{\Delta(1)}{\Delta(0)} = \frac{1}{3}. \tag{8}$$

The relation (8) show a universality for the SBC model in the limiting for $\alpha=1$ and $\alpha=0$ and is independent of the dimensionality of model as well of the lattice structure. Therefore, with this result, it is possible to estimate the exact values of the initial slope for the case of complete correlation. For

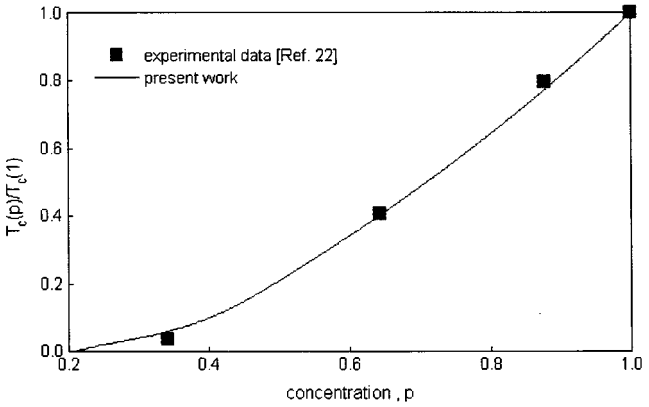

FIG. 1. Phase diagram in the randomly site-bond-correlated diluted Heisenberg model for the compoud $KNi_pMg_{1-p}F_3$ with interaction between *NNN* sites. The correlation parameter estimated by the SBC model in the present work is $\overline{\alpha}=0.145$.

example, for a simple cubic lattice the initial slope for the uncorrelated case is $\Delta(1)=1.04$.[29] By using the relation (8) we find $\Delta(0)=3.12$.[30]

In this part we are interested in evaluating the value of the correlation parameter α that better adjusts to the experimental data obtained by de Aguiar and co-workers.[22] It was mentioned previously, that all works that studied the SBC model do not make experimental contact with the compound $KNi_pMg_{1-p}F_3$. Furthermore, the value estimated for the correlation parameter was not obtained. However, in the present work we aim to obtain an estimate for the correlation parameter α for the Heisenberg model that better adjusts to the SBC model. This value, by experimental expectation, must be in the range $0<\alpha<0.20$.[24] A computational-analytical evaluation can provide a value for α that adequately describes $KNi_pMg_{1-p}F_3$. In the present work, we use the experimental data and assume that it satisfies Eq. (5). In this approach, for each experimental point (p_E, T_E),[22] we find one value for the correlation parameter. All these values are such that $0<\alpha_i<0.2$ $(i=1,2,3,4)$. In order to obtain the curve fitting that adjusts itself better to the experimental data, we evaluated the arithmetic mean of the values found for $\overline{\alpha_1}$. Therefore, our estimate for the correlation parameter is $\alpha=0.145$. The phase diagram in the T-p plane for the Heisenberg model when $\alpha=\overline{\alpha}=0.145$ is presented in Fig. 1 together with the experimental data. We observe that the value obtained for the correlation parameter α shows good adjustment with the experimental data as well as exhibiting the behavior of the experimental evidence for $KNi_pMg_{1-p}F_3$. On the other hand, we also observe that P_c $(0\leqslant\alpha<1)=0.205$, while p_c $(\alpha=1)=0.589$. Therefore, the Heisenberg adequately describes the compound $KNi_pMg_{1-p}F_3$ with the value of the correlation parameter within experimental expectation. The reduced critical temperature and critical concentration for the three-dimensional Heisenberg model in the SBC model is quoted in Table I.

TABLE I. Values for reduced critical temperature and critical concentration for the Heisenberg model for the SBC dilution model are compared with exact results or by expansion series.

	Critical temperature	
k_BT_c/J	4.767	4.330[a]
	Critical concentration	
p_c $(0\leqslant\alpha<1)$	0.205	0.31[a]
p_c $(\alpha=1)$	0.589	

[a]Reference 31.

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