Study of the spin correlation and the percolation threshold in the B-site spinel.

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High-temperature series expansion of the correlation function on the B- spinel lattice are computed up to order 6 in $\beta = \frac{1}{k_B T}$ for Heisenberg model including both the nearest and next-nearest-neighbor interactions J_1 and J_2 respectively.

The behavior with the temperature and the site dilution is presented. The approach is applied to the experimental results of the B-spinel $ZnCr_{2x}Al_{2-2x}S_4$.

The critical temperature and the critical exponents for the susceptibility and the correlation length are deduced by applying the Padé approximant methods. The following estimates are obtained for the familiar critical exponents: γ= 1.382±0.012 and ν = 0.691±0.011. These values are not sensitive to the dilution ratio x.

The bond percolation threshold x_p is determined by studying the disorder variation of the correlation length ξ. The x_p is considered as the concentration at which ξ vanishes. The obtained values are x_p=0.27 when only J_1 is considered and 0.23 when both J_1 and J_2 are considered.

I. INTRODUCTION

In the last several years, the theoretical and experimental studies of randomly diluted magnetic systems have been intensively investigated[1-6]. In these systems, non-magnetic ones randomly replace some magnetic atoms on the lattice and a bond connecting each pair of occupied atom first neighbor is modified. In this context, the diluted magnetic systems are associated with a percolation problem when the temperature goes to zero.

In this work, it is intended to extend the development of the high-temperature series expansion (H.T.S.E) of the spin correlation functions to the order 6 in β with nearest-neighbor (n-n) and next-nearest-neighbor (n-n-n) interactions J_1 and J_2 respectively, for the diluted B-spinel lattice $ZnCr_{2x}Al_{2-2x}S_4$.

To deduce the spin correlation functions $\gamma_{ij} = \frac{\langle S_i . S_j \rangle}{S(S+1)}$ between spins at site i and j we have used the diagrammatic representation, performed by Stanley and Kaplan (SK)[7,8]. Their method is general and can applied to any lattice. This semi-classical treatment is a simplification of the more complex procedure of Rushbrook and Wood[9] used for the calculation of the susceptibility in the quantum-mechanical case. In order to obtain more information about the magnetic properties in the B-spinel system, we have calculated the spin correlation functions between first, second and third n-n, spins γ_1, γ_2 and γ_3 respectively. We have applied the Padé approximants (P.A) method to the H.T.S.E of the spin correlation function, the magnetic susceptibility $\chi(q)$ and the correlation length $\xi(T)$. The critical temperature T_N, the critical exponents γ for the magnetic susceptibility and ν for the correlation length are deduced. In this whole range of concentration $0.85 \le x \le 1$, the exponents γ and ν are found to be equal to 1.382±0.12 and 0.691±0.011 respectively.

It is known that the measurement of the neutron scattering is a powerful tool for the investigation of spin correlation[10,11]. The magnetic ordering of the diluted system $ZnCr_{2x}Al_{2-2x}S_4$ was determined by neutron diffraction and reported in previous papers[5,12]. The main feature of this investigation can be briefly summarized. In the high concentration $(0.85 \le x \le 1)$ a long-range magnetic order builds up below the Neel temperature T_N. Below $x \le 0.80$ no range magnetic ordering was detected reflecting the spin glass (S.G) state. No anomaly or saturation was observed at the freezing temperature T_f below that the correlation length ξ still increases.

The inverse correlation length $\frac{1}{\xi}$ decreases linearly with temperature leading to a finite value at T=0 in the S.G state. The nature of the short-range order (S.R.O), which appears in the concentrated sample above T_N and in the S.G samples is of the same type. The S.R.O is fairly well developed at relatively high-temperature, T=100K[5]. The broad diffuse peak of the S.R.O becomes broader and weaker as the temperature is raised corresponding to the thermal disruption of the magnetic correlation. The variation of $S(\mathbf{q})$, obtained by the H.T.S.E technique with the disorder (variation with temperature T and magnetic site dilution x) is in good accordance with experimental neutron diffraction intensity obtained in the $ZnCr_{2x}Al_{2-2x}S_4$. For a given concentration the location of the broad diffuse peak is insensitive to the temperature[5,12,13]. In order to respect this result, the thermal variation of the set of J_1 and J_2 is obtained. Their absolute value decreases with the temperature.

The study of the thermal variation of ξ obtained by the H.T.S.E combined with the experimental observations in the $ZnCr_{2x}Al_{2-2x}S_4$ system permits to

derive the variation of ξ with x at T=0. This in turn also gives us the bond percolation threshold x_p. The x_p is considered as the concentration at which ξ vanishes. The obtained values are x_p=0.27 when only J_1 is considered and 0.23 when both J_1 and J_2 are considered. These values are in accordance with those obtained by the Monte Carlo method and magnetic experimental results[14,15].

II. THEORY AND RESULTS

Starting with the zero-field Heisenberg Hamiltonian:

$$H = -2 \sum_{i,j} J_{ij} \mathbf{S_i}.\mathbf{S_j} \qquad (1)$$

Where the summation run over all pairs of n-n and n-n-n interactions J_1 and J_2 respectively. The expansion of the spin correlation function in powers of β is obtained as follow[7].

$$\langle \mathbf{S_i}.\mathbf{S_j} \rangle = \frac{Tr \mathbf{S_i}.\mathbf{S_j} e^{-\beta H}}{Tr e^{-\beta H}} = \sum_{=0} \frac{(-1)}{!} \alpha \beta \qquad (2)$$

With:

$$\alpha = \nu - \sum_{k=0}^{-1} C_k \alpha_k \mu_{-k}, \nu_m = \langle \mathbf{S_i}.\mathbf{S_j} H^m \rangle_{\beta=0} \quad (3)$$

and $\mu_m = \langle H^m \rangle_{\beta=0}$

This leads to a diagrammatic representation given in[8]. The calculation of the coefficients of the γ_i according to the diagrammatic method involves two separate phases.

(a) The finding of all the diagrams or graphs which can be constructed from dashed line connecting the site o and i and straight lines, and the determination of diagrams, in the case of the spinel lattice, whose contribution is non vanishing.

(b) Counting the number of times that a diagram can occur in the spinel lattice. Step (a) has already been accomplished in the S.K work. Step (b), however is very tedious.

In our case, we have to deal with the two Heisenberg constant couplings: J_1 and J_2 between first and second n-n in the spinel system.

For each topological form of a given diagram, a full line can either represent J_1 or J_2. We must, thus derive from each topological form a class of diagrams; each of them represents a term of the series as $J_1^m J_2^n (m,n = 0,1,..., \text{ and } m+n =)$ for the th order. This is especially the limiting factor in how far one can carry the expansion.

In this fashion, we calculated all of the coefficients required for the calculation of the spin correlation function $\gamma_i (i = 1-3)$ in the case of a diluted B-spinel lattice to order $= 6$, and the results of this calculation are given in ref.[25].

Equation (2) combined with the results in ref.[25] allows the computation of the spin correlation functions γ_i. The analytical expressions of γ_1, γ_2 *and* γ_3 are given in ref.[25].

Since we are interested in estimating critical points, we have used the P.A methods to study the dependence of the critical temperature T_N on the relative strength of J_1, J_2 and the ratio of dilution x. T_N is estimated as the temperature at which γ_1 diverges[12]. Using [3,4] P.A. we have calculated the ratio $\frac{T_N}{J_1}$ in the long range-ordering region of the system $ZnCr_2Al2_{-2x}S_4$. Figure 1 shows the obtained $\frac{T_N}{J_1}$ (solid line) against dilution x. In this figure we have included, for comparison, the experimental results obtained by neutron diffraction given in[5]. One can see the excellent agreement between the theoretical and experimental results.

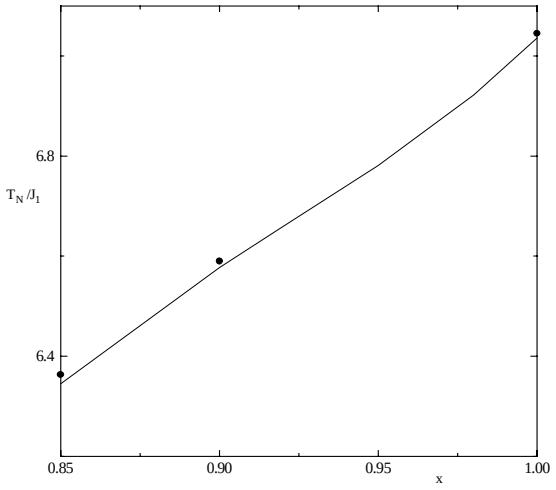

FIG. 1: Variation of the $\frac{T_N}{J_1}$ versus the magnetic concentration x in $ZnCr_{2x}Al_{2-2x}S_4$ for $0.85 \le x \le 1$. The circles are the experimental points reported in[5]. The line is the result obtained from the high-temperature series expansion extrapolated with the Padé approximant methods.

The wavelength dependent susceptibility $\chi(\mathbf{q})$ and correlation function $S(\mathbf{q})$ are defined as:

$$\chi(\mathbf{q}) = g\mu_B^2 \beta \sum_{i,j} \langle \mathbf{S_i}.\mathbf{S_j} \rangle e^{-i\mathbf{q}.\mathbf{R_{ij}}} \qquad (4)$$

and

$$S(\mathbf{q}) = \sum_{i,j} \langle \mathbf{S_i}.\mathbf{S_j} \rangle e^{-i\mathbf{q}.\mathbf{R_{ij}}} \qquad (5)$$

Where μ_B is the Bohr magneton, g the gyromagnetic ratio and $\mathbf{R_{ij}}$ is the separation between the spins i and j.

In order to obtain a qualitative measure of the correlation length $\xi(T)$ for a given temperature T we

have expanded the correlation function $S(\mathbf{q})$ in a Taylor expansion about the magnetic reciprocal lattice $\mathbf{Q}$ of the given system[16].

$$S(\mathbf{q}) = S(\mathbf{Q})[1 - \xi^2(\mathbf{q}-\mathbf{Q})^2 + 0(\mathbf{q}-\mathbf{Q})^4] \qquad (6)$$

Recasting this in the Ornstein-Zernike from, the following asymptotic form is obtained:

$$S(\mathbf{q}) = \frac{S(\mathbf{Q})\kappa^2(T)}{[\kappa^2(T) + (\mathbf{q}-\mathbf{Q})^2]} \qquad (7)$$

Where $\kappa(T) = \xi^{-1}(T)$

In the B-spinel lattice and for the particular case of the helimagnetic structure with wave vector $\mathbf{Q} = [0\,0\,q]$, we obtain:

$$S(\mathbf{q}) = 4\left[1 + \gamma_{aa} + \gamma_{ab}\cos\left(\frac{\pi q}{2}\right) + \gamma_{ac}\cos(\pi q)\right] \qquad (8)$$

$\gamma_{aa} = 2\gamma_1 + 4\gamma_3$ is the in-plane correlation, $\gamma_{ab} = 4\gamma_1 + 8\gamma_2$ is the correlation between neighbor planes and where $\gamma_{ac} = 4\gamma_2 + 8\gamma_3$ is the correlation between the second neighbor planes.

The maximization of $S(q)$ with respect to q=q_0 gives:

$$\cos\left(\frac{\pi q_0}{2}\right) = -\frac{\gamma_{ab}}{4\gamma_{ac}} \qquad (9)$$

q_0 is related to the helix angle by: $\theta = \frac{\pi q_0}{2}$.

Expanding the cosine of equation (8) in the Taylor expression about $q=q_0$ and using the equation (6), we obtain:

$$\left(\frac{\xi}{a}\right)^2 = \frac{1}{8S(q_0)}\left[-\gamma_{ac} + \frac{\gamma_{ab}^2}{16\gamma_{ac}}\right] \qquad (10)$$

where a is the lattice parameter.

The simplest assumption that one can make concerning the nature of the singularity of the magnetic susceptibility $\chi(\mathbf{q})$ and the correlation length $\xi(T)$ is that in the neighborhood of the critical points the above two functions exhibit an asymptotic behavior:

$$\chi(\mathbf{q}) \alpha (T_N - T)^{-\gamma} \text{ and } \xi^2(T) \alpha (T_N - T)^{-2\nu} \qquad (11)$$

T_N represents the critical temperature, deduced by the P.A. methods, γ and ν the critical exponents.

Representation of the series expansion of $\chi(\mathbf{q})$ and $\xi^2(T)$ by [3,4] P.A. would enable us to find γ and 2ν by finding: $\lim_{y \to y_N} (y - y_N)\frac{d}{dy} LogF(y), (F(y) = \chi, \xi^2)$ with $y = \frac{J_1}{T}$ and $y_N = \frac{J_1}{T_N}$. We have obtained the values of the above two critical exponents in the case of the B-spinel $ZnCr_{2x}Al_{2-2x}S_4$. Using the helix angle θ deduced from the position of the magnetic satellite of the neutron diffraction pattern [17], θ(x=1)=71^0, θ(x=0.90)=68^0 and θ(x=0.85)=64^0, we have obtained the central values of γ and ν for three concentrations γ=1.382±0.012 and ν=0.691±0.011. These values may be compared with those of the 3D Heisenberg model, namely, 1.3866±0.0012 and 0.7054±0.0011[18-19]. The agreement is excellent.

On the other hand in figure 2 we show the temperature dependence of the $S(\mathbf{q})$ in the case of $ZnCr_2S_4$ (x=1). We observed a strongly developed S.R.O hump peaking at the wave vector $\mathbf{q_0} = [0\,0\,0.79]$. The broad diffuse peak becomes broader and weaker as the temperature is raised, corresponding to the thermal disruption of the magnetic correlation. The accordance with the experimental result obtained by the neutron diffraction is excellent. We note that the S.R.O is fairly well developed at relatively high temperatures, T≈100K, and may even be present at room temperature. The thermal variation of set of absolute values of J_1 and J_2 keeping the maximum of $S(\mathbf{q})$ at the particular wave vector $\mathbf{q_0}$ is obtained. This variation is presented in figure 3.

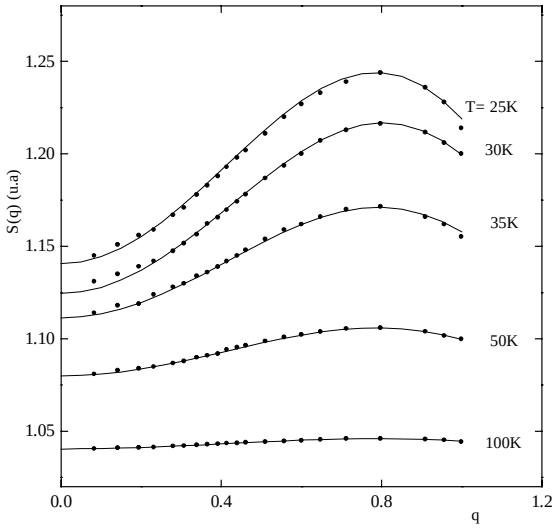

FIG. 2: Static structure factor $S(\mathbf{q})$ of the system $ZnCr_2S_4$ for various temperatures along the wave vector $\mathbf{q} = [0\,0\,q]$. Lines are theoretical results obtained from equation 2 (see text) and the dots are the experimental data obtained in [5].

In figure 4 we present the variation of $S(\mathbf{q})$ with the ratio of dilution x in the paramagnetic phase of the system $ZnCr_{2x}Al_{2-2x}S_4$. This variation is similar to that with the temperature. The only difference is that the position of the peak shifts to lower values when x decreases. This result is in agreement with the experimental neutron diffraction[5]. The variation of the

peak position of the S.R.O with the ratio of dilution and not with the temperature can be linked to the fact that the lattice parameter in the thiospinel varies with x according to the Vegard's rule[20] but it is independent of temperature[21].

The magnetic correlation length ξ is given by equation 3. In figure 5 the variation of the inverse correlation length $\frac{1}{\xi}$ with the temperature

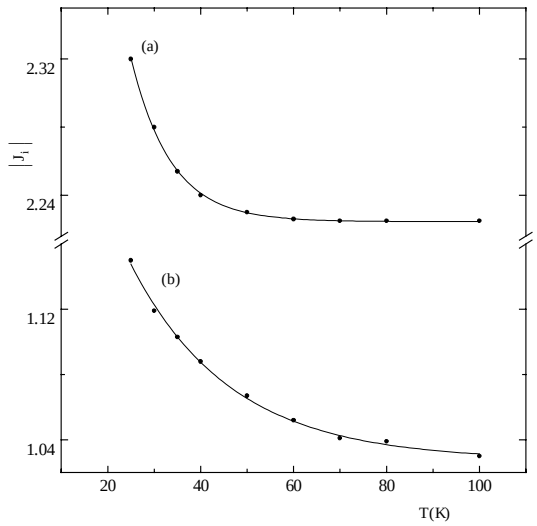

FIG. 3: Temperature dependence of the absolute values of exchange integrals in the system $ZnCr_2S_4$. (a) nearest neighbor interactions J_1. (b) next nearest neighbor interactions J_2.

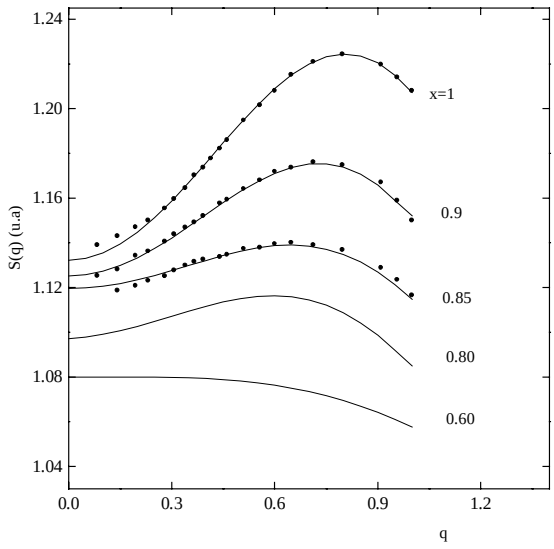

FIG. 4: Structure factor $S(\mathbf{q})$ for several ratio of dilution in $ZnCr_{2x}Al_{2-2x}S_4$ in the paramagnetic phase. The lines are the theoretical results and the dots are experimental data obtained by neutron diffraction obtained in [5].

for different x is presented. The variation is linear with the same slope of different composition in the S.G. region.

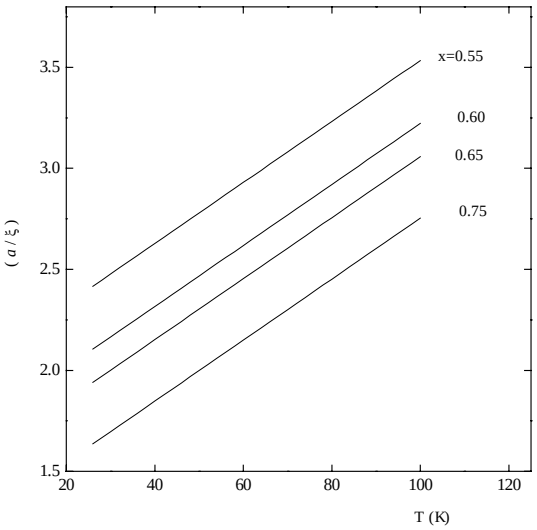

FIG. 5: Temperature dependence of the inverse correlation length for several values of x situated in the spin glass region of the system $ZnCr_{2x}Al_{2-2x}S_4$.

An important result exhibited by neutron diffraction experiments in the thiospinel systems is the non existence of any anomaly in the correlation length at the freezing temperature T_f and the extrapolated values of $\frac{1}{\xi}$ at T=0 are finite in the S.G region[12,13,22]. We extrapolate the correlation length presented in figure 5 to T=0. In figure 6 the variation of $\xi(T=0)$ with x is presented for two sets of parameters, (i) n-n interactions only and (ii) n-n and n-n-n interactions. We know, on physical grounds, that if the magnetic concentration fell below the percolation concentration (for a given lattice), the macroscopic connectivity of the magnetic lattice would break down such that the spin correlation length would reduce to zero. This critical behavior is deduced from figure 5. The percolation threshold is considered as the concentration at which $\xi = 0$. It is equivalent to the concentration at which there is no magnetic connection between spins. The obtained values are x_p=0.27 when only J_1 is considered and 0.23 when both J_1 and J_2 are considered. These values are in agreement with those obtained by the Monte Carlo method[14] and the magnetic measurements[15].

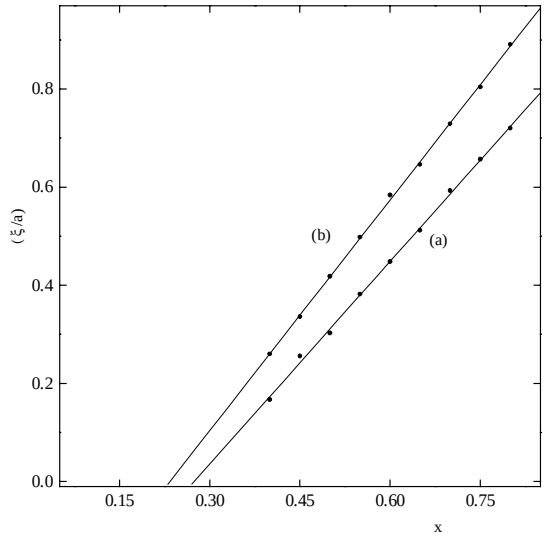

FIG. 6: Variation of the correlation length with x in $ZnCr_{2x}Al_{2-2x}S_4$ at T=0. (a) Only J_1 is considered. (b): Both J_1 and J_2 are considered.

III. CONCLUSIONS:

The first three spin correlation functions for the diluted B-spinel lattice $ZnCr_{2x}Al_{2-2x}S_4$ where determined with the nearest and next- nearest neighbor interactions to order 6 in β by the high-temperature series expansion.

High-temperature series expansion extrapolated with Padé approximants is shown to be a convenient method to provide valid estimations of critical temperatures for magnetic system[23]. By applying this method to the γ_1, we have estimated the long-range order region of $ZnCr_{2x}Al_{2-2x}S_4$. The obtained results are in good agreement with those obtained by experimental neutron diffraction. The critical exponents of the magnetic susceptibility γ and the correlation length ν are found equals to 1.382±0.012 and 0.691±0.011 in the whole range of concentration $0.85 \le x \le 1$. These values are compatible with those of 3D Heisenberg model. According to Harris criterion[24], since $ZnCr_2S_4$ is a good 3D Heisenberg system, the critical exponent α of the specific heat is less than zero[18], hence, the critical exponents are unaffected by dilution.

On the other hand the variation of $S(\mathbf{q})$ and ξ with temperature and magnetic site dilution x is derived. We find that for a given ratio of dilution the thermal variation of the magnetic structure remains peaked at the wave-vector corresponding to the magnetic structure. The set of the nearest-neighbor and next-nearest neighbor interactions, J_1 and J_2 respectively, respecting this behavior are deduced. For the pure system (x=1), the absolute values of J_1 and J_2 exhibit an exponential decay with the temperature. The position of the peak in $S(\mathbf{q})$ shifts to lower $\mathbf{q}$ with increasing dilution. These features are in agreement with neutron diffraction experiments on the system $ZnCr_{2x}Al_{2-2x}S_4$.

The obtained inverse correlation length $\frac{1}{\xi}$ decreases linearly with the temperature and the extrapolated values at T=0 are finite for the concentration in the spin glass region $x \le 0.80$. The variation of the correlation length with x at T=0 gives us a method for the estimation of the threshold percolation x_p. The x_p is considered as the concentration at which ξ vanishes. The obtained values are x_p=0.27 when only J_1 is considered and 0.23 when both J_1 and J_2 are considered. These values are comparable with those obtained by the Monte Carlo method and the experimental phase diagram in the T-x plane of $ZnCr_{2x}Al_{2-2x}S_4$ obtained by the a.c and d.c susceptibility.

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