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Spin-one Heisenberg ferromagnet with three-atom exchange

S N Mitra and K G Chakraborty†
Department of Physics, Basirhat College, 24 Parganas (North), West Bengal-743412, India

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Abstract. We study the statistical mechanics of a spin-one ferromagnet with the nearest-neighbour bilinear Heisenberg exchange constant J and the three-atom coupling constant L using the equation-of-motion method for the two-time temperature-dependent Green function. It is seen that, as the three-atom coupling parameter $\alpha = L/J$ is increased positively, the Curie temperature T_C first increases steeply and then increases very slowly and ultimately approaches the limiting value $k_B T_C/Jz = \frac{4}{3}$ as α tends to infinity. The situation is much more complicated for negative α but, as $\alpha \to \infty$, T_C approaches the limiting value $\frac{4}{3}$. The temperature variation in spontaneous magnetization m and the quadrupolar ordering parameter λ for various values of α are studied. It is seen that there exists a critical value of α (which we shall call α_c) beyond which the phase transition is first order for SC, BCC and FCC lattices. α_c decreases as the number z of nearest neighbour increases. The discontinuity in m has been found to be extremely sensitive near α_c. The results are discussed with reference to those obtained by earlier workers.

1. Introduction

In this paper we present a study of the statistical mechanical properties of a three-dimensional spin-one Heisenberg ferromagnet with three-atom exchange in addition to the usual bilinear exchange, described by the following Hamiltonian (Iwashita and Uryu 1974, Munro and Girardeau 1976, Akasmit and Westwanski 1978, Adler and Oitmaa 1979):

$$H = -\sum_i \omega_0 S_i^z - \sum_{ij} J_{ij} S_i \cdot S_j - \sum_{ijk} L_{ijk} (S_i \cdot S_j)(S_j \cdot S_k) \qquad (1)$$

where $\omega_0 = \mu H_a$, μ being the magnetic moment per site and H_a the applied magnetic field. J_{ij} represents the usual bilinear exchange between two spins. S_i is the spin operator attached to the lattice site i. L_{ijk} represents coupling between three spins. The higher-order exchange term seems to arise from the process of double-electron exchange. In equation (1) the well known biquadratic exchange term is absent. In fact, the above Hamiltonian is a special case of the general Hamiltonian considered by Munro and Girardeau (1976). The statistical mechanics of the model were studied by Adler and Oitmaa (1979) using the Green function equation-of-motion method. Munro and Girardeau (1976) applied the molecular-field approximation (MFA) to an extended Hamiltonian, but in the MFA the effect of dimensionality of the lattice is not taken into account. In the Green function approach of Adler and Oitmaa the single-site Green functions were decoupled. The purpose of the present paper is to apply the random phase approximation (RPA) to decoupling the Green functions at a later stage. Two equations of motion—one for different sites and the other for the same sites—are developed and decoupled by the RPA, and the energy spectrum and other thermodynamic quantities are derived and computed subsequently.

† Author to whom correspondence should be addressed.

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2. Equations of motion, energy spectrum and ordering parameters

Using equation (1) the equation of motion for the two-time temperature-dependent two-spin Green function $\langle\langle S_f^+; S_m^-\rangle\rangle$ (f, m being the lattice sites) may be readily derived (Zubarev 1960) and can be written in the following form:

$$\begin{aligned}(\omega-\omega_0)\langle\langle S_f^+; S_m^-\rangle\rangle = \frac{\langle S_f^z\rangle}{\pi}\delta_{fm} + 2\sum_i J_{if}\langle\langle S_i^z S_f^+ - S_f^z S_i^+; S_m^-\rangle\rangle \\ - \sum_{jl} L_{fjl}\langle\langle S_f^z S_l^z \sigma_j^+ + \tfrac{4}{3}(S_f^z S_l^+ - S_l^z S_f^+) - \tfrac{1}{3}S_f^z S_l^+\lambda_j \\ - \tfrac{2}{3}S_f^+ S_l^z\lambda_j - \tfrac{1}{2}S_f^+ S_l^- \sigma_j^+ - \tfrac{1}{2}S_f^+ S_l^+\sigma_j^-; S_m^-\rangle\rangle \\ - \sum_{il} L_{ifl}\langle\langle -S_i^z S_l^z\sigma_f^+ + S_i^z S_l^+\lambda_f + \tfrac{1}{2}S_i^+ S_l^-\sigma_f^+ + \tfrac{1}{2}S_i^+ S_l^+\sigma_f^-; S_m^-\rangle\rangle \end{aligned} \tag{2}$$

where $\lambda_i = 3(S_i^z)^2 - 2$, $\sigma_i^\pm = S_i^z S_l^\pm + S_l^\pm S_l^z$, and the angular brackets denote the usual averages. σ_i will now be treated as a single operator. The equation of motion for the Green function $\langle\langle \sigma_f^+; S_m^-\rangle\rangle$ can be derived in the form

$$\begin{aligned}(\omega-\omega_0)\langle\langle \sigma_f^+; S_m^-\rangle\rangle = \frac{\langle\lambda_f\rangle}{\pi}\delta_{fm} + 2\sum_i J_{if}\langle\langle S_i^z\sigma_f^+ - \lambda_f S_i^+; S_m^-\rangle\rangle \\ - \sum_{jl} L_{fjl}\langle\langle \lambda_f S_l^z\sigma_j^+ + \tfrac{4}{3}(\lambda_f S_l^+ - S_l^z\sigma_f^+) - \tfrac{1}{3}\lambda_f S_l^+\lambda_j \\ - \tfrac{2}{3}\sigma_f^+ S_l^z\lambda_j - \tfrac{1}{2}\sigma_f^+ S_l^-\sigma_j^+ - \tfrac{1}{2}\sigma_f^+ S_l^+\sigma_j^-; S_m^-\rangle\rangle \\ - \sum_{il} L_{ifl}\langle\langle -S_i^z S_l^z S_f^+ + S_i^z S_l^+ S_f^z + \tfrac{1}{2}S_i^+ S_l^- S_f^+ - \tfrac{1}{2}S_i^+ S_l^+ S_f^-; S_m^-\rangle\rangle. \end{aligned} \tag{3}$$

Neglecting the terms S^+S^+, Fourier-transforming to momentum space, decoupling the higher-order Green functions by the RPA and solving two linearized equations of motion, we finally arrive at the result

$$\langle\langle S_k^+; S_q^-\rangle\rangle = (m/\pi)\delta_{k-q}(\omega-\omega_k)^{-1} \tag{4}$$

$$\langle\langle \sigma_k^+; S_q^-\rangle\rangle = (\lambda/\pi)\delta_{k-q}(\omega-\omega_k)^{-1} \tag{5}$$

where k and q are the momentum indices, and m and λ are the ordering parameters defined as $m = \langle S^z\rangle$ and $\lambda = 3(\langle S^z)^2\rangle - 2$. ω_k represents the energy spectrum given by

$$\omega_k = \omega_k^{(1)} \pm M(k) \tag{6}$$

with

$$\omega_k^{(1)} = \tfrac{1}{2}[4mJ_0 - 2mJ_k - \tfrac{1}{3}m(4-\lambda)L_{kk} + \tfrac{4}{3}m(2+\lambda)L_{00} - 2\lambda mL_{0k}] \tag{6a}$$

$$\begin{aligned}M(k) = \tfrac{1}{2}\{[2mJ_k + \tfrac{1}{3}m(4-\lambda)L_{kk} + 2m^2L_{0k} - 2m^2L_{00}]^2 - 8m^2(\lambda-m)J_k(L_{00}-L_{0k}) \\ - \tfrac{4}{3}m^2(4-\lambda)(\lambda-m)L_{kk}(L_{00}-L_{0k})\}^{1/2}\end{aligned} \tag{6b}$$

where J_k, L_{kk}, etc, are the Fourier transforms of J_{ij}, L_{ijk}, etc (Akasmit and Westwanski 1978, Adler and Oitmaa 1979). Equation (6) represents two branches of the energy spectrum in contrast with the single branch obtained by Adler and Oitmaa. However, these two branches do not coincide, at $T = 0$, with the exact result. Hence we shall consider that branch which leads to the exact result at $T = 0$. It is seen that the negative sign satisfies this requirement and so we reject the positive sign. At $T = 0$, equation (6) thus leads to

$$\omega_k = \omega_0 + 2(J_0 - J_k) + 3L_{00} - 2L_{0k} - L_{kk} \tag{7}$$

which is exact.

The spontaneous magnetization m and the quadrupolar ordering parameter λ are obtained from the following equations:

$$m = (1 + 2y)F(y) \tag{8a}$$

$$\lambda = F(y) \tag{8b}$$

where the function $F(y)$ is given by

$$F(y) = (1 + 3y + 3y^2)^{-1} \tag{8c}$$

with

$$y = \frac{1}{N}\sum_k [\exp(\beta\omega_k) - 1]^{-1}. \tag{9}$$

3. The Curie temperature

Following Akasmit and Westwanski (1978) we use the results $L_{00} = zL_1$, $L_{0k} = zL_1\gamma_k$, $L_{kk} = zL_1(z\gamma_k^2 - 1)/(z - 1)$, $L_1 = (z - 1)L$, $J_k = zJ\gamma_k$, $J_0 = zJ$ with

$$\gamma_k = \frac{1}{z}\sum_k \exp(\mathrm{i}k \cdot \delta) \tag{10}$$

where z is the number of nearest neighbours, δ is the nearest-neighbour vector, and J and L represent the nearest-neighbour bilinear and three-atom exchange constants, respectively. The energy spectrum (equation (6)) can then be readily written as

$$\omega_k = \omega_0 + 2mzJR_k(1 - \gamma_k) \tag{11}$$

where R_k is a renormalization factor given by the following expression:

$$R_k = 1 + \tfrac{1}{6}\alpha z(4 - \lambda) + \tfrac{1}{2}\alpha(z - 1)(m + \lambda) + \tfrac{1}{6}\alpha z\gamma_k(4 - \lambda) + \alpha(z - 1)(\lambda - m)f(\gamma_k) \tag{12}$$

where $\alpha = L/J$ and

$$f(\gamma_k) = N/(N + N') \tag{13}$$

with

$$N = \gamma_k + \tfrac{1}{6}\alpha(4 - \lambda)(z\gamma_k^2 - 1) \tag{14a}$$

$$N' = -\alpha(z - 1)m(1 - \gamma_k). \tag{14b}$$

Expanding $f(\gamma_k)$ in powers of γ_k and substituting in the energy spectrum, one can include terms up to any desired order and obtain the computed values of m, λ and T_C. Instead of this, we consider only the leading term of the expansion of $f(\gamma_k)$ so that we can write

$$R_k = R(1 + \mu\gamma_k) \tag{15}$$

R being given by

$$R = 1 + a\alpha z + b\alpha + \alpha P \tag{15a}$$

with

$$\mu = a\alpha z/R \tag{15b}$$

$$a = \tfrac{1}{6}(4 - \lambda) \tag{15c}$$

$$b = \tfrac{1}{2}(m + \lambda)(z - 1) \tag{15d}$$

$$P = \tfrac{1}{2}(\lambda - m)(z - 1)a[a + m(z - 1)]^{-1}. \tag{15e}$$

Using the above simplifications, one can easily write the explicit expressions for m and λ. We do not present these expressions here; rather we shall concentrate on the Curie temperature. Taking the limit $\omega_0 \to 0$, $m \to 0$, we arrive at the following expression for the Curie temperature T_C:

$$T_C = \tfrac{4}{3}z(J/k_B)(R_0/F_0) \tag{16}$$

where R_0 and F_0 are the values of R and F at T_C, F being defined by

$$F = (1 - \mu)^{-1}[I - \mu(I - 1)] \tag{17}$$

where I is the usual Watson sum given by

$$I = \frac{1}{N}\sum_k (1 - \gamma_k)^{-1}. \tag{17a}$$

In computing F_0, the parameter μ is to be replaced by μ_0, the latter being obtained from equations (15*a*)–(15*e*) in the limit $\omega_0 \to 0$, $m \to 0$.

4. Results and discussion

For computations of m, λ and T_C we shall use the symbols $\alpha = L/J$, $\alpha' = \alpha/(z-1)$, $t_C = k_B T_C/4Jz$ and $t = k_B T/4Jz$. The values of t_C for the SC, BCC and FCC lattices have been calculated for both positive and negative values of α. Firstly the results show that, for α approaching infinity, t_C for all lattices converges to the interesting limiting value $\frac{1}{3}$. This value of t_C is exactly equal to that of a simple spin-one Heisenberg ferromagnet in the MFA. In addition to this, it has been found that, as α increases from zero, t_C first increases sharply, then increases slowly and ultimately approaches the limiting value. For negative α, the nature of the variation in t_C is complicated. This complication arises because, for negative α, the ferromagnetic state ceases to be stable, at least for α less than a certain limiting value. In fact, we have seen that, as α increases negatively, the critical values α_0 at which t_C vanishes are -0.30, -0.208 and -0.132 for SC, BCC and FCC lattices, respectively. The vanishing of t_C at a certain negative value of α agrees qualitatively with that obtained by Munro and Girardeau.

If one considers more negative values, there is a range of α (we call this range 'forbidden') where t_C is negative, but, after this range, t_C decreases from an infinitely large value as α increases to more negative values; eventually t_C approaches the limit $\frac{1}{3}$ in the limit $\alpha \to -\infty$. All these results are shown in figures 1 and 2.

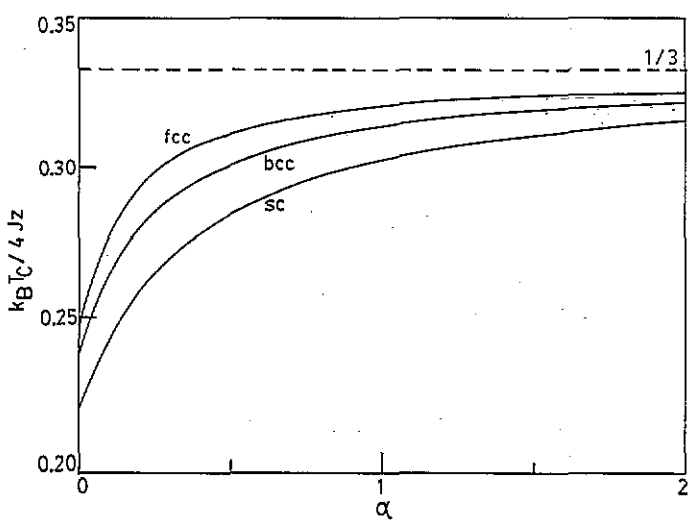

Figure 1. The variation in $k_B T_C/4Jz$ with α for SC, BCC and FCC lattices: ----, limiting value of $k_B T_C/4Jz = \frac{1}{3}$ for $\alpha \to \infty$.

Some interesting aspects have been observed in the studies of thermal variation in m and λ. The results for SC, BCC and FCC lattices for a wide range of values of α have been computed and analysed. The results are summarized in figures 3–7. The results of computation show that there exists a critical value α_c beyond which m and λ become double valued at and beyond T_C, forming a bulge near T_C. It is seen that the bulge is more pronounced for large α. Only the bulge for $\alpha = 0.2$, $z = 6$, is shown. It appears that both m and λ jump discontinuously at T_C, and the first-order phase transition occurs for all lattices. The value of α_c was estimated roughly by Adler and Oitmaa (1979) for a

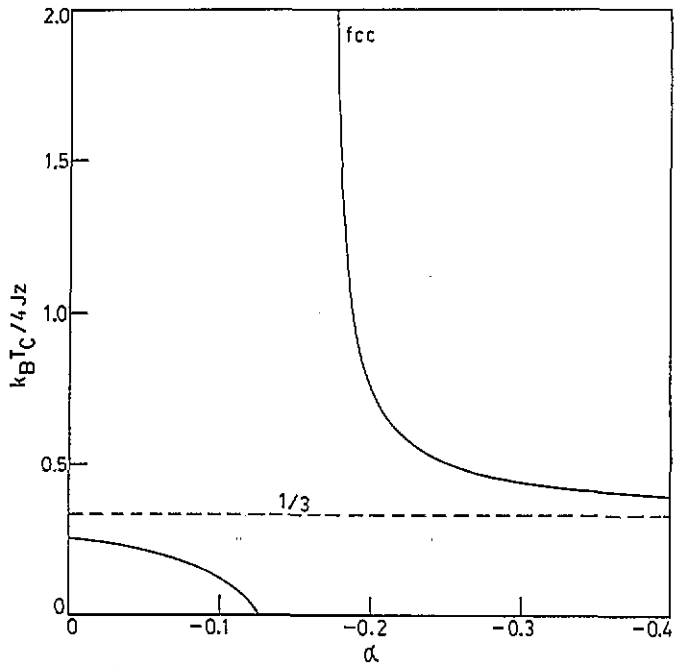

Figure 2. The variation in $k_BT_C/4Jz$ with negative values of α for the FCC lattice: ----, limiting value of $k_BT_C/4Jz = \frac{1}{3}$.

SC lattice. The value obtained in the present treatment for the SC lattice is slightly lower than their value. The values of α_c for the SC, BCC and FCC lattices obtained in the present paper are 0.489, 0.324 and 0.194, respectively. Thus α_c decreases as z increases. It may also be noted that only one phase transition exists—the transition which occurs from the ferroquadrupolar phase ($m \neq 0; \lambda \neq 0$) to the paramagnetic phase ($m = 0; \lambda = 0$)—and that no separate quadrupolar phase exists.

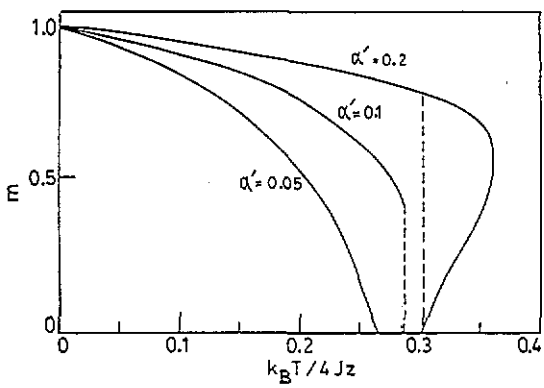

Figure 3. The variation in m with $k_BT/4Jz$ for the SC lattice and for several values of the reduced coupling parameter $\alpha' = \alpha/(z-1)$, where only the bulge for $\alpha' = 0.2$ beyond T_C is shown: ----, discontinuities.

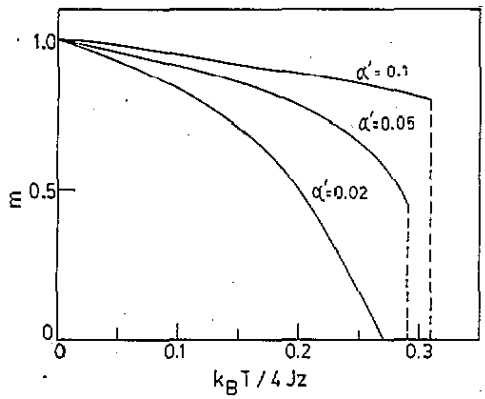

Figure 4. The variation in m with $k_BT/4Jz$ for the BCC lattice for $\alpha' = 0.02$, 0.05 and 0.1: ----, discontinuities.

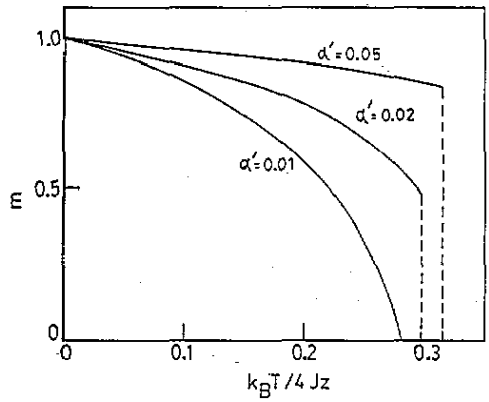

Figure 5. The magnetization curves for the FCC lattice for $\alpha' = 0.01$, 0.02 and 0.05: ----, discontinuities.

We have also carried out computations for m for small values of α for all the three lattices. The results are presented in table 1. Single-valued m and λ are denoted by 1 and double-valued m and λ by 2. The values of α_c are also presented.

Finally, we have computed the magnitude of the discontinuous change Δm_s in the spontaneous magnetization at T_C. The variation in Δm_s with α for the SC, BCC and FCC lattices is represented in figure 7. It should be observed that Δm_s is extremely sensitive near α_c as demonstrated by the almost vertical nature of the curve. However, away from α_c the value of Δm_s increases very slowly and remains almost constant at large α, reaching the value $\Delta m_s = 1$ for $\alpha \to \infty$. We also note that, for the same value of α, Δm_s is larger for larger z.

It is worth noting in this connection that, using the generalized mean-field approximation, Munro and Girardeau (1976) estimated Δm_s for the case of biquadratic exchange only. They observed that the sensitivity mentioned above is much less than that occurring in the present case of three-atom exchange. However, the entire problem should be re-examined in the

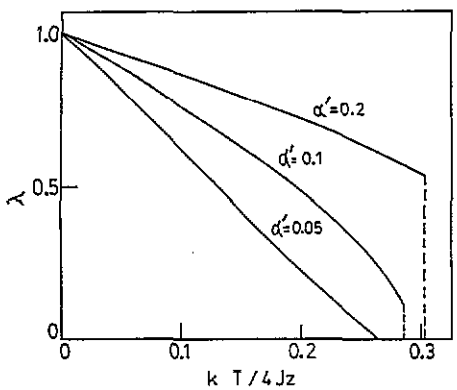

Figure 6. The temperature variation in the quadrupolar ordering parameter λ for the SC lattice corresponding to $\alpha' = 0.05$, 0.1 and 0.2: ----, discontinuities.

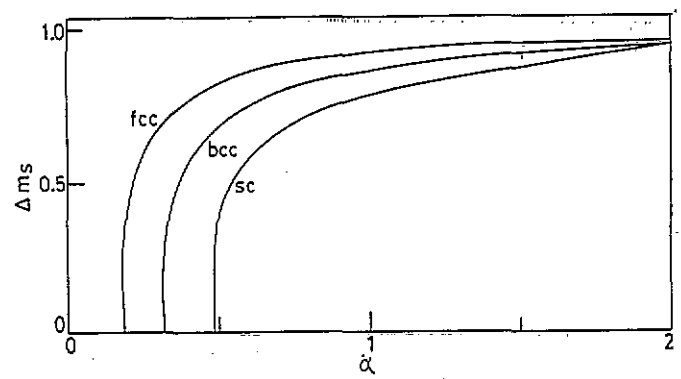

Figure 7. The magnitude Δm_s of the discontinuous jump in the spontaneous magnetizaiton plotted against α for SC, BCC and FCC lattices.

light of the Green function approximation.

5. Concluding remarks

The Green function study of a spin-one Heisenberg ferromagnet with three-atom exchange presented in the preceding sections yields several special features which may be summarized as follows.

(1) A first-order transition appears at T_C, the Curie temperature, above $\alpha > \alpha_c$ for SC, BCC and FCC lattices, α_c decreasing as z increases. Below α_c the transition is second order.

(2) As the three-atom coupling parameter α increases positively, T_C increases sharply at first and then slowly, gradually approaching the limiting value $k_B T_C/zJ = \frac{4}{3}$, which is identical with the molecular-field value for a Heisenberg ferromagnet without the three-atom exchange.

(3) As α increases negatively, it has been found that there exists a critical value α_0 at which T_C vanishes.

Table 1. Multiplicity of the roots of m and λ at $t_C + 0.0001$ for SC, BCC and FCC lattices for several values of α. The superscript c in the first column denotes the critical value of α, corresponding to that lattice for which m and λ are double valued. Single valued is denoted by 1 and double valued by 2.

	Multiplicity of roots of m and λ		
α	SC	BCC	FCC
0.0001	1	1	1
0.001	1	1	1
0.01	1	1	1
0.1	1	1	1
0.194^c	1	1	2
0.2	1	1	2
0.3	1	1	2
0.324^c	1	2	2
0.4	1	2	2
0.489^c	2	2	2
0.5	2	2	2
0.6	2	2	2

The disappearance of T_C at a certain negative value of α, however, agrees qualitatively with the work of Munro and Girardeau; on further negative increase in α, t_C becomes negative and, as α increases still further negatively, t_C decreases from an infinitely large positive value, ultimately approaching the same molecular-field limit.

As it has been observed by Munro and Girardeau that the $S = 1$ model does not favour partially ordered states and also since, for large negative α, the ferromagnetic spin alignment is presumably not favoured, we may conjecture that the phase transition for large negative α, as shown in figure 2, might correspond to the antiferromagnetic-to-paramagnetic phase transition. It should be noted that figure 2 corresponds only to the FCC lattice and that the same type of variation has also been found for SC and BCC lattices.

Finally, it is necessary to point out that the expansion of $f(\gamma_k)$ has been found to be extremely sensitive and one should be careful about the expansion and about the consideration of terms in the expansion. In addition to the leading term, which has been considered here, other higher-order terms should also be included for improved results. Unfortunately the higher-order terms are so complicated that at present we are not able to include the contribution from these terms. These aspects will be the subject of future investigations where we shall also include the effects of biquadratic exchange. For quantitatively more accurate formulation we need to develop a more rigorous procedure and in this respect the irreducible Green function theory (Chakraborty 1988, 1989) is important and work in this direction is in progress. Some initial work has already been completed and we have noted some similarities to the phase transitions in the Ising model with multi-spin coupling (Chakraborty 1992).

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