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Effect of biquadratic exchange and crystal field anisotropy on the Curie temperature of anisotropic ferromagnet

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Abstract

A spin-1 anisotropic ferromagnet with crystal field anisotropy parameter D and biquadratic exchange interaction parameter α ($0 \leqslant \alpha \leqslant 1$) has been investigated using the method of double time temperature dependent Green functions. Mixed Callen and RPA decoupling approximations have been utilized. The variations of Curie temperature T_C with α for different values of the exchange anisotropy parameter η ($0 \leqslant \eta \leqslant 1$) at $D/J = 1$ and for different values of D/J at $\eta = 1$ have been studied. Also the value of exchange anisotropy parameter for which dependence of T_C on α vanishes has been calculated at finite D. Drawbacks and limitations in the earlier calculations have been pointed out. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

The effect of biquadratic exchange on magnetic properties such as magnetization, Curie temperature, magnetic susceptibility, etc. have been investigated by Allan and Betts [1] Brown [2], Biegala [3], Chaddha and Singh [4] and Chaddha [5] using high-temperature series expansion method, constant coupling molecular field approximation and Green's function (GF) decoupling theories. The critical value of α ($= \alpha_c$ corresponds to the tricritical point) for which the transition becomes first order was also determined by Biegala [3].

In all these previous studies, the spin system has been confirmed as being isotropic with respect to both the bilinear and biquadratic exchange interactions. Due to the failure of this isotropic model to represent many real magnetic compounds, the inclusion of anisotropy in the exchange interaction in the presence of crystal field anisotropy has been proposed by many authors. The observed data from magnetic resonance experiments and theoretical studies of the crystal field, along with the perturbation calculation of the spin–orbit interaction [6,21] have clearly established the fact that the exchange anisotropy and the crystal field anisotropy

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are two fundamentally important features existing in a wide variety of magnetic substances and thus the inclusion of the corresponding terms in the Hamiltonian is fully justified.

The variation of Curie temperature T_C with the biquadratic exchange interaction for different values of the exchange anisotropy parameter η ($0 \leqslant \eta \leqslant 1$, the particular cases $\eta = 0$ and 1 describe Blume–Emery–Griffiths model [7] and Heisenberg model that contains in addition to the bilinear exchange, biquadratic exchange and single-ion anisotropy D, respectively) was studied by Iwashita and Uryu [8] using pair model approximation but the crystal field anisotropy parameter D was taken to be zero. At the same time only one ordering parameter, viz. $\sigma = \langle S^Z \rangle$, called dipolar ordering parameter was taken into account whereas due to the presence of the biquadratic exchange term, the present problem needs consideration of two ordering parameters, σ as well as $y = \langle C_g^2 \rangle = 6\langle (S_g^Z)^2 \rangle - 4$, called quadrupolar ordering parameter. Though the problem was later on studied by Chaddha and Kalsi [9] by taking both the said ordering parameters into account using simple GF technique but here too the calculations were restricted to $D = 0$

Tiwari and Srivastava [10] considered the combined effect of biquadratic exchange and crystal field anisotropy using a simplified form of the random phase approximation (RPA). Their calculations were, however, restricted to $\eta = 1$ case only and at the same time while linearizing the higher order Green functions they did not even pay attention to the fact that there exist the following spin-1 identities.

$$S_g^Z C_g^2 = S_g^Z \{6\,(S_g^Z)^2 - 4\} = 2S_g^Z \tag{1a}$$

and

$$A_g^2 S_g^Z + S_g^Z\, A_g^2 = A_g^1 = S_g^+ \tag{1b}$$

in which

$$A_g^2 = -\,[A_g^1, (S_g^Z)^2] = A_g^1 S_g^Z + S_g^Z A_g^1 \tag{1c}$$

and used the following linearization:

$$\langle\langle A_g^2\, S_g^Z + S_g^Z A_g^2; S_h^- \rangle\rangle = 2\langle S^Z \rangle \langle\langle A_g^2; S_h^- \rangle\rangle, \tag{2a}$$

$$\langle\langle S_g^Z C_g^2 A_f^2; S_h^- \rangle\rangle = \langle S_g^Z \rangle \langle C_g^2 \rangle \langle\langle A_f^2; S_h^- \rangle\rangle. \tag{2b}$$

The GF, $\langle\langle A_g^2; S_h^- \rangle\rangle$ arising from the biquadratic exchange and crystal field anisotropy terms was of course treated as such, that is without decoupling.

They concluded that the effects due to the biquadratic exchange and crystal field anisotropy annul each other at $\alpha = 0.8$ and $D/J = 0.1$ (J being the exchange integral) and from then T_C is found to increase with the increase of α (up to $\alpha = 1$) instead of decreasing with the increase of α as expected. The expected behaviour could be interpreted as follows. Because of the squared form, that is $(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)^2$, $\boldsymbol{S}_i$ and $\boldsymbol{S}_j$ being the two spin operators at the lattice sites i and j, respectively, the biquadratic exchange interaction is able to stabilize the antiferromagnetic state ($\theta = 180^\circ$) as well as the ferromagnetic state ($\theta = 0^\circ$). Therefore, the spin-1 Heisenberg system with the biquadratic term does become disordered at a temperature lower than the pure Heisenberg model. However, only ferromagnetic state is stabilized in the Ising spin system.

Their [10] calculations were, however, based on the approximation $D/J \ll 1$ whereas the values of D/J used by them ranged from 0–1 in the text. Though the calculations were later on extended by them [11] by assuming the exchange interaction to be anisotropic but with the similar error.

The combined effect of biquadratic exchange and crystal field anisotropy was also considered by Zheng [12] using GF technique but with the following new type of Callen decoupling approximation:

$$\langle\langle S_f^Z A_g^2; S_h^- \rangle\rangle = \sigma \langle\langle A_g^2; S_h^- \rangle\rangle - \tfrac{1}{4} y \langle S_f^- S_g^+ \rangle \langle\langle A_f^1; S_h^- \rangle\rangle, \tag{3a}$$

$$\langle\langle C_f^2 A_g^i; S_h^- \rangle\rangle = \tfrac{1}{2} y \langle\langle A_g^i; S_h^- \rangle\rangle - \tfrac{1}{4} y \langle S_f^- A_g^i \rangle \langle\langle A_f^1; S_h^- \rangle\rangle, \quad (i = 1, 2) \tag{3b}$$

for which there was no physical justification. At the same time, the calculations were restricted to $\eta = 1$ case only. Though the problem was earlier studied by Zheng [13] in the case of anisotropic Heisenberg ferromagnet in the presence of crystal field anisotropy but with the same decoupling approximation and at the same time at $\alpha = 0$.

Later on, the problem at $\eta = 1$ was also studied by Chakraborty [14] using irreducible GF theory

which he claims, yields different results from those reported by GF equation of motion method employing various decoupling approximations, at α very small. But at the same time there were limitations on the calculations and it was not possible to have systematic quantitative estimates for the variation of dipolar and quadrupolar ordering parameters with temperature using the said theory. Chakraborty [15] applying the irreducible GF theory did study the case of anisotropic Heisenberg ferromagnet in the presence of crystal field anisotropy but the biquadratic exchange term was taken to be zero.

Keeping all these things in mind, we thought to re-study the problem. It was also thought to find the critical value of the exchange anisotropy parameter η for which biquadratic exchange interaction has no effect on the Curie temperature T_C. A decoupling procedure, the same as that used by Chaddha and Singh [4] in their earlier work has been utilized here. That is, the three spin Green functions are decoupled using Callen decoupling approximation [16] whereas for the still higher-order Green functions, RPA [17] has been utilized.

2. Theory

We consider a generalized Heisenberg model expressed as

$$H = -\sum_{ij} J_{ij}[\{S_i^Z S_j^Z + \eta(S_i^X S_j^X + S_i^Y S_j^Y)\} + \alpha\{S_i^Z S_j^Z + \eta(S_i^X S_j^X + S_i^Y S_j^Y)\}^2] - D\sum_i (S_i^Z)^2, \tag{4}$$

where J_{ij} is a measure of the exchange force between the ith and jth lattice sites, $\boldsymbol{S}_i$ is the spin operator associated with the ith lattice site with components S_i^X, S_i^Y and S_i^Z. η measures the strength of the anisotropic exchange whereas D is the parameter measuring strength of the crystal field anisotropy. α the biquadratic exchange parameter is defined by the ratio of the biquadratic exchange to the bilinear exchange. The summation is assumed to extend over nearest neighbour pairs i and j. In the present paper, we restrict ourselves to the spin-1 case.

Using Devlin's [18] notation, we define the following two Green functions:

$$G_{gh}^i(t-t') = \langle\langle A_g^i(t); S_h^-(t')\rangle\rangle \quad (i = 1, 2). \tag{5}$$

Each A_g^i satisfies the relation

$$[A_g^i, S_g^Z] = -A_g^i \tag{6a}$$

and also

$$[A_g^2, (S_g^Z)^2] = -A_g^1, \qquad C_g^2 = [A_g^2, S_g^-]. \tag{6b}$$

The equations of motion of the Fourier transform of the above Green functions generated by the Hamiltonian [4] can be written as

$$\begin{aligned} wG_{gh}^1(w) = {} & (2\sigma/2\pi)\delta_{gh} \\ & + (2-\alpha\eta^2)\sum_f J_{gf}\langle\langle S_f^Z A_g^1; S_h^-\rangle\rangle \\ & - \eta(2-\alpha)\sum_f J_{gf}\langle\langle S_g^Z A_f^1; S_h^-\rangle\rangle \\ & - (\alpha\eta/2)\sum_f J_{gf}\langle\langle C_g^2 A_f^2; S_h^-\rangle\rangle \\ & + (\alpha/2)\sum_f J_{gf}\langle\langle\{(4+2\eta^2) \\ & \times (S_f^Z)^2 - 4\eta^2\}A_g^2; S_h^-\rangle\rangle \\ & + D\langle\langle A_g^2; S_h^-\rangle\rangle, \end{aligned} \tag{7}$$

$$\begin{aligned} wG_{gh}^2(w) = {} & (y/2\pi)\delta_{gh} + (2-\alpha\eta^2)\sum_f J_{gf}\langle\langle S_f^Z A_g^2; S_h^-\rangle\rangle \\ & - \tfrac{1}{2}\eta(2-\alpha)\sum_f J_{gf}\langle\langle C_g^2 A_f^1; S_h^-\rangle\rangle \\ & - \alpha\eta\sum_f J_{gf}\langle\langle S_g^z A_f^2; S_h^-\rangle\rangle \\ & + (\alpha/2)\sum_f J_{gf}\langle\langle\{(4+2\eta^2)(S_f^Z)^2 - 4\eta^2\} \\ & \times A_g^1; S_h^-\rangle\rangle \\ & + D\langle\langle A_g^1; S_h^-\rangle\rangle. \end{aligned} \tag{8}$$

The three spin Green functions are decoupled using Callen decoupling approximation [16]:

$$\langle\langle S_f^Z A_g^1; S_h^- \rangle\rangle_{f\neq g} = \sigma G_{gh}^1 - \sigma/2 \langle S_f^- A_g^1 \rangle\, G_{fh}^1 \quad (9a)$$

and for the still higher-order Green functions, we have chosen the following decoupling approximation:

$$\langle\langle S_f^Z A_g^2; S_h^- \rangle\rangle_{f\neq g} = \langle S_f^Z \rangle\, G_{gh}^2, \quad (9b)$$

$$\langle\langle C_f^2 A_g^2; S_h^- \rangle\rangle_{f\neq g} = \langle C_f^2 \rangle\, G_{gh}^2. \quad (9c)$$

The operators S_g^Z and C_g^2 correspond to the longitudinal and the operators A_g^i to the transverse motions of the spins, respectively. Since only the operators S_g^Z and C_g^2 have finite ensemble averages, the above decoupling represents the same type of factorization as in

$$\langle\langle S_f^Z A_g^1; S_h^- \rangle\rangle_{f\neq g} = \sigma G_{gh}^1, \quad (10)$$

which is nothing but RPA [17] and implies that the transverse motion of the spin at site g is completely uncorrelated with the longitudinal motion of the spin at site f. This is certainly a reasonable approximation as long as $f \neq g$.

Using the above decoupling approximations and assuming the translational invariance, we can solve for the spatial Fourier transforms of Green functions $G_w^1(K)$ and $G_w^2(K)$, i.e.

$$G_w^1(K) = \frac{1}{2\pi}\left[\frac{a_+}{w - w_+} + \frac{a_-}{w - w_-}\right],$$

$$G_w^2(K) = \frac{1}{2\pi}\left[\frac{b_+}{w - w_+} + \frac{b_-}{w - w_-}\right], \quad (11)$$

where the poles of the Green functions are given by

$$w_\pm = \sigma(A + B) \pm \sqrt{\sigma^2(C + B)^2 + w_0^2(K)}$$

and

$$w_0^2(K) = (D + F)(D + G).$$

Making use of spectral theorem and setting $g = h$ and $t = t'$, one obtains the following correlation functions:

$$\langle S_g^- A_g^1 \rangle = \frac{8 - y}{6} - \sigma = \frac{1}{N}\sum_K \left[\frac{a_+}{e^{\beta w_+} - 1} + \frac{a_-}{e^{\beta w_-} - 1}\right], \quad (12)$$

$$\langle S_g^- A_g^2 \rangle = \sigma - y/2 = \frac{1}{N}\sum_K \left[\frac{b_+}{e^{\beta w_+} - 1} + \frac{b_-}{e^{\beta w_-} - 1}\right] \quad (13)$$

with the renormalization function,

$$f = \frac{1}{NJ_0}\sum_K J_K \psi(K) \quad (14)$$

and

$$a_\pm = \sigma \pm \frac{\sigma^2(C + B) + y(D + F)/2}{\sqrt{\sigma^2(C + B)^2 + w_0^2(K)}},$$

$$b_\pm = y/2 \pm \frac{\sigma(D + G) - \sigma y(C + B)/2}{\sqrt{\sigma^2(C + B)^2 + w_0^2(K)}},$$

$$A = (2 - \alpha\eta^2)J_0 - \eta J_K,$$

$$B = f\{\eta(2 - \alpha)J_0 - (2 - \alpha\eta^2)J_K\}/4,$$

$$C = -\eta(1 - \alpha)J_K,$$

$$F = \alpha[\{(4 + 2\eta^2)m - 4\eta^2\}J_0 - \eta y J_K]/2,$$

$$G = \alpha\{(4 + 2\eta^2)m - 4\eta^2\}J_0/2 + Cy,$$

$$G_w^n(K) = \sum_{g-h} G_{gh}^n(w) e^{-iK\cdot(g-h)}, \qquad n = 1, 2,$$

$$J_K = \sum_{g-h} J_{gh} e^{-iK\cdot(g-h)},$$

$$\beta = 1/kT \quad \text{and} \quad m = \langle (S^Z)^2 \rangle.$$

It may be noted that the above result reduces to that of Callen [16] for $\eta = 1$ and $\alpha = D = 0$, that is in the case of the Heisenberg model. Also the result reduces to that of Chaddha and Singh [4] obtained earlier in the case of isotropic ferromagnets at $D = 0$.

Following Devlin [18] for the accurate determination of T_C, Eqs. (12) and (13) are expanded in the power series of σ about the point $\sigma = 0$. The coefficients of each power of σ must be the same on each side of each equation. A comparison of the zeroth power of σ will yield the paramagnetic equation valid for all $T > T_C$. The coefficients of the first power of σ in each equation will yield one additional equation. Accordingly, we get the following two coupled equations,

$$\frac{8 - y}{3y} = \frac{1}{N}\sum_K \frac{(D + F)}{w_0(K)} \coth \beta_C\, w_0(K)/2, \quad (15)$$

$$\frac{4(2-y)}{3y^2} = \frac{1}{2N}\sum_K \frac{(B-C)}{w_0(K)} \coth \beta_C w_0(K)/2 + \frac{1}{4N}\sum_K \beta_C(A+B)\text{cosech}^2 \beta_C w_0(K)/2 \quad (16)$$

with

$$f = \frac{y}{2N}\sum_K \frac{J_K}{J_0}\frac{(D+F)}{w_0(K)} \coth \beta_C w_0(K)/2. \quad (17)$$

The reason why T_C now appears as an unknown is that the comparison of the coefficients of the first power of σ gives equations which are not valid for $T > T_C$, and hence they are valid only at the single temperature T_C, which is as yet undetermined.

3. Results and discussion

Eqs. (15)–(17) have been solved numerically in the case of spin-1 anisotropic BCC ferromagnet to study the variation of Curie temperature T_C with $\alpha(0 \leqslant \alpha \leqslant 1)$ for different values of exchange anisotropy parameter η and for different D/J values. The variation of T_C with α for $D/J = 0$, 1 and 4 at $\eta = 1$ that is, in the case of a Heisenberg model, has been displayed in Fig. 1. T_C is found to be a decreasing function of α for all values of D/J. Thus the two effects one due to the crystal field anisotropy and another due to the biquadratic exchange never annul each other as far as $0 \leqslant \alpha \leqslant 1$ is concerned which is contrary to the results earlier reported by Tiwari and Srivastava [10]. Their wrong conclusions are probably because of the invalid approximations used by them during the course of calculations. The slopes of our T_C versus α curves for finite values of the crystal field anisotropy parameter D are significantly different from those reported by Zheng [12]. This could be due to the fact that we have used a decoupling procedure which is physically very sound. However, for $D/J \to \infty$, the renormalization factor $f \to 0$ and the results reduce to those in RPA. Also at $D/J \to \infty$, one gets $y = 2$, implying $\langle (S^Z)^2 \rangle = 1$, which is an expected result because in this limit S^Z is effectively restricted to take only the values $\pm S$ so that $\langle (S^Z)^2 \rangle \to S^2$.

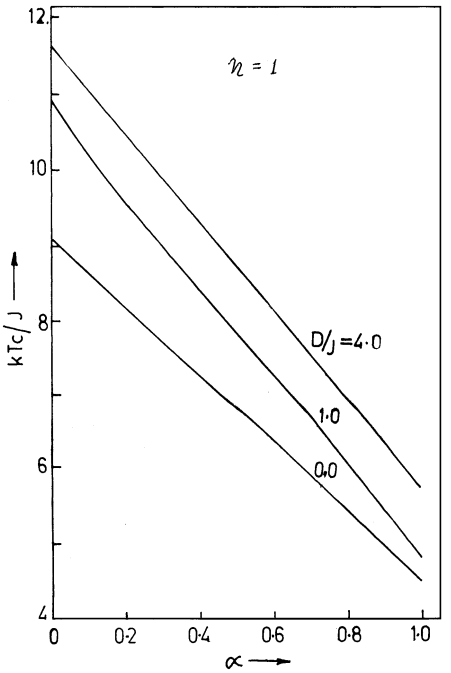

Fig. 1. Variation of kT_C/J with α on a BCC lattice for $D/J = 0$, 1 and 4 at $\eta = 1$.

Fig. 2 shows the variation of T_C with α for different values of the exchange anisotropy parameter $\eta = 0$, 0.5, 0.65, 0.8 and 1.0 at $D/J = 1$. The results indicate a decrease in T_C with an increase of α for $\eta = 1$ spin system whereas T_C is found to increase with increasing α for $\eta = 0$ spin system. This trend is consistent with the results reported by Iwashita and Uryu [8] in pair model approximation and by Chaddha and Kalsi [9] using simple GF technique. However, the results for the intermediate values of η differ significantly. The departure could be probably due to the fact that Iwashita and Uryu [8] considered only the dipolar ordering parameter whereas the present problem needs consideration of two ordering parameters viz. dipolar and as well as the quadrupolar ordering parameters. The omission of the quadrupolar ordering parameter by Brown [19] resulted in two values of T_C for the same value of α which is quite unphysical and also the existence of a tricritical point at $\alpha > 1$ which contradicted the results reported in molecular field approximation by Chen and Levy [20] and also by Biegala [3] in RPA.

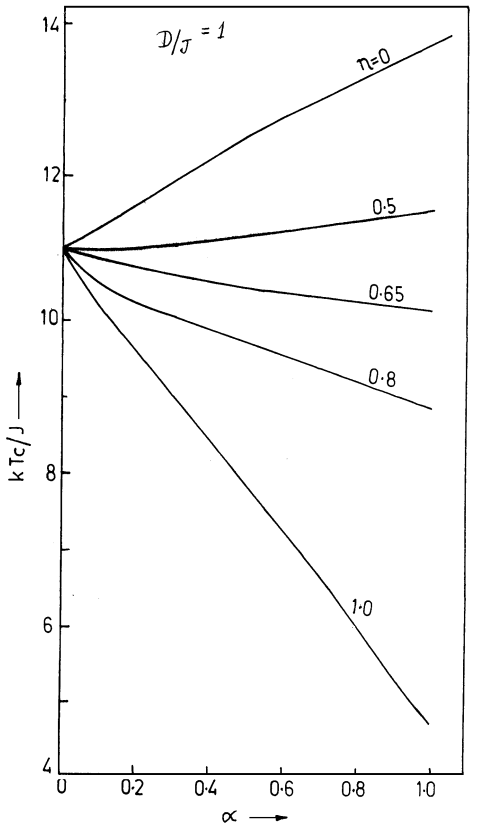

Fig. 2. Variation of kT_C/J with α on a BCC lattice for $\eta = 0.0$, 0.5, 0.65, 0.8 and 1.0 at $D/J = 1$.

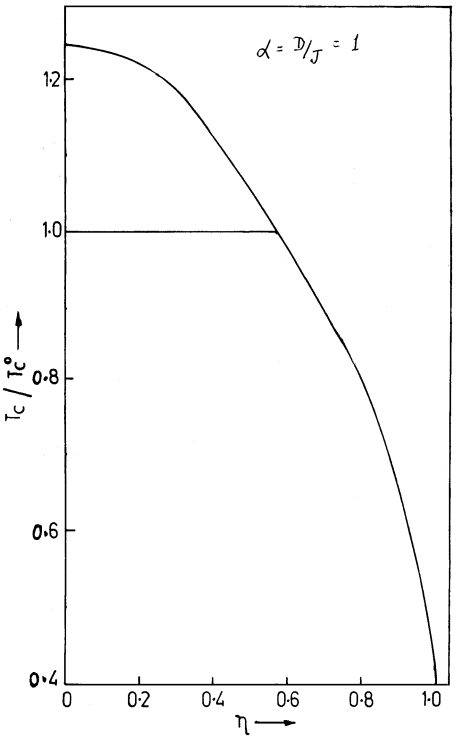

Fig. 3. Variation of T_C/T_C^0 with η on a BCC lattice at $\alpha = D/J = 1$.

Denoting the Curie temperature in the case of vanishing α as T_C^0 and in order to estimate η_C, the value of η at which biquadratic exchange interaction has no effect on the Curie temperature, we have studied the variation of T_C/T_C^0 with η at $\alpha = D/J = 1$. The results so obtained are displayed in Fig. 3. The value of η_C determined from these observations comes out to be 0.57 in the case of spin-1 BCC ferromagnet. Though the value of η_C has been determined earlier by Iwashita and Uryu [8] using pair model approximation and by Chaddha and Kalsi [9] using simple GF technique they were both determined at $D/J = 0$. To the best of our knowledge it is for the first time that the value of η_C has been found at the finite value of D.

The present results are thought to be certainly more reliable and at the same time more realistic compared with the earlier results as they have been obtained for the finite value of D in the presence of anisotropic exchange and by assuming that there are two ordering parameters σ and y in the biquadratic problem and also by using a decoupling procedure which is physically very sound. The formalism, of course eliminates completely the need to decouple the higher order GF, $\langle\langle A_g^2; S_h^- \rangle\rangle$ arising from the biquadratic exchange and crystal field anisotropy terms.

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