

Anisotropic ferromagnet with biquadratic exchange

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Abstract. A formulation, using the double time Green's function method, is presented for the anisotropic spin-one ferromagnetic system in the presence of Ising part of the biquadratic exchange and an expression is given for the critical temperature T_c in terms of two parameters. One is associated with the uniaxial anisotropy (treated exactly) and the other one corresponds to the biquadratic interaction (treated in RPA). The bilinear Heisenberg exchange term is treated in Callen's approximation emphasizing the role of intersite correlation for small parameters. The prediction that T_c increases with the biquadratic exchange is found to be consistent with the recent calculation based on high temperature series expansion.

Keywords. Anisotropic ferromagnet; biquadratic exchange.

1. Introduction

It has come to be appreciated in recent years that in order to understand the magnetic behaviour of insulators containing ions, whose low lying states possess non-zero orbital angular momentum, it is generally incorrect to assume the Hamiltonian as a Heisenberg form coupling the effective spins isotropically. There are additional anisotropy terms that favour certain directions in the ordered state. One such term is the single-ion uniaxial anisotropy. The others are the non-bilinear terms, the origin of which can be traced in the mixing of orbital excited states with orbital singlet state *via* the spin-orbit coupling. When the orbital angular momentum is not quenched, the admixtures of orbital excited states can be much larger so that the effect of the nonbilinear terms, particularly the biquadratic one, in the Hamiltonian can be as large as the bilinear terms (Birgeneau *et al* 1969; Elliott and Thorpe 1968). A theoretical basis for the existence of such terms was given by Anderson (1959) and by Huang and Orbach (1964) within the framework of superexchange interaction. It was consistent with the result obtained by Schrödinger (1941) that the exchange interaction operator for $S = 1$ between the spins at the sites f and g , is

$$\rho_{fg} = (\mathbf{S}_f \cdot \mathbf{S}_g)^2 + (\mathbf{S}_f \cdot \mathbf{S}_g) - 1.$$

On the other hand, the necessity of including a small (negative) biquadratic term in the Hamiltonian was first pointed out by Harris and Owen (1963), and Rodbell *et al* (1963) to explain their data on paramagnetic resonance of Mn pairs in MgO. Nevertheless, these anisotropic terms are supposed to predict accurately the

observed magnon spectra, dipolar and quadrupolar ordering and the phase transitions (Pink and Tremblay 1972, Micnas 1974).

Thus a Hamiltonian of the form

$$H = -g\mu_B \tilde{H} \sum_g S_g^z - \sum_{i,\sigma} J_{i\sigma} S_i \cdot S_\sigma - \alpha \sum_{i,\sigma} J_{i\sigma} \left[S_i^* S_\sigma^* + \frac{\eta_i}{2} (S_i^+ S_\sigma^- + S_i^- S_\sigma^+) \right]^2 - D \sum_g (S_g^*)^2 \quad (1)$$

is representative of certain physically interesting systems (Brown 1971, Pink and Tremblay 1972). We shall use it to investigate the magnetic properties particularly the critical temperature T_c , as a function of α , of an anisotropic ferromagnetic spin-one system in reasonable approximations (for $\eta_i = 0$, and $\tilde{H} = 0$). The single-ion term will be evaluated exactly. Since random phase approximation (RPA) ignores correlations among spins located on different lattice sites, we shall incorporate these correlations within Callen's (1963) approximation in the bilinear exchange term. The present paper is an extension of the earlier work (Kumar and Sharma 1976 *a*) for $\alpha = 0$, and we will call it I. The biquadratic exchange will be treated in RPA. The formulation for a general spin, in particular for spin-one, will be presented in section 2 and the results will be discussed in section 3.

2. Formulation

To evaluate the single-ion anisotropy term exactly, we define the following general operators (Devlin 1971),

$$\begin{aligned} A_\sigma^1 &= S_\sigma^+; & A_\sigma^i &= -[A_\sigma^{i-1}, (S_\sigma^*)^2] \text{ for } 2S \geq i \geq 2, \\ B_\sigma^i &= [A_\sigma^i, S_\sigma^+]; & C_\sigma^i &= [A_\sigma^i, S_\sigma^-], \text{ for each } i, \\ [A_\sigma^i, S_\sigma^*] &= -A_\sigma^i \text{ for each } i. \end{aligned} \quad (2)$$

The particular commutation relationship that closes the set of anisotropy operators is

$$[A_\sigma^{2S}, (S_\sigma^*)^2] = \sum_{j=1}^{2S-1} a_j A_\sigma^j \quad (3)$$

where particular values for the c numbers a_j depends on the size of the spin of a system, (for the particular case $S = 1$, $a_1 = -1$).

The equation of motion for the anisotropy Green function $\langle\langle A_\sigma^i; S_h^- \rangle\rangle$ is

$$\begin{aligned} \omega \langle\langle A_\sigma^i; S_h^- \rangle\rangle &= \frac{1}{2\pi} Z_i \delta_{\sigma,h} + \sum_j J_{j\sigma} \langle\langle 2A_\sigma^i S_j^* - B_\sigma^i S_j^- - C_\sigma^i S_j^+; S_h^- \rangle\rangle \\ &+ D \langle\langle A_\sigma^{i+1}; S_h^- \rangle\rangle + 2\alpha \sum_j J_{j\sigma} \langle\langle (S_j^*)^2 A_\sigma^i S_\sigma^* \\ &+ S_\sigma^* A_\sigma^i (S_j^*)^2; S_h^- \rangle\rangle. \end{aligned} \quad (4)$$

where $\langle C_\sigma^i \rangle = Z_i$. The operators C_σ^i are linear combinations of the powers of operators S_σ^i and hence correspond to longitudinal motion of spins.

The operators A_σ^i and B_σ^i are of the form of raising and lowering operators and therefore represent the transverse component of the spins. Further, only the operators C_σ^i have finite ensemble averages, therefore,

$$\langle\langle B_\sigma^i S_f^-; S_h^- \rangle\rangle \simeq 0. \quad (5)$$

Let us decouple the biquadratic exchange Green function in RPA as follows,

$$\begin{aligned} \langle\langle (S_f^z)^2 A_\sigma^i S_\sigma^z + S_\sigma^z A_\sigma^i (S_f^z)^2; S_h^- \rangle\rangle &= \langle (S_f^z)^2 \rangle \langle\langle A_\sigma^i S_\sigma^z + S_\sigma^z A_\sigma^i; S_h^- \rangle\rangle \\ &= \langle (S_f^z)^2 \rangle \langle\langle A_\sigma^{i+1}; S_h^- \rangle\rangle \end{aligned} \quad (6)$$

Due to eqs (5) and (6), eq. (4) becomes

$$\begin{aligned} \omega \langle\langle A_\sigma^i; S_h^- \rangle\rangle &= \frac{1}{2\pi} Z_i \delta_{\sigma,h} + \left(D + 2\alpha \sum_f J_{f\sigma} \langle (S_f^z)^2 \rangle \right) \\ &\cdot \langle\langle A_\sigma^{i+1}; S_h^- \rangle\rangle + \sum_f J_{f,\sigma} \langle\langle 2A_\sigma^i S_f^z - C_\sigma^i S_f^+; S_h^- \rangle\rangle. \end{aligned} \quad (7)$$

The system with spin one implies that $i = 1$ and 2 . Equation (7), for $i = 1$, can be expressed as

$$\begin{aligned} \omega \langle\langle A_\sigma^1; S_h^- \rangle\rangle &= \frac{1}{2\pi} Z_1 \delta_{\sigma,h} + \left(D + 2\alpha \sum_f J_{f\sigma} \langle (S_f^z)^2 \rangle \right) \\ &\langle\langle A_\sigma^2; S_h^- \rangle\rangle + 2 \sum_f J_{f\sigma} \langle\langle S_\sigma^+ S_f^z - S_\sigma^z S_f^+; S_h^- \rangle\rangle \end{aligned} \quad (8)$$

and for $i = 2$, as

$$\begin{aligned} \omega \langle\langle A_\sigma^2; S_h^- \rangle\rangle &= \frac{1}{2\pi} Z_2 \delta_{\sigma,h} + \left(D + 2\alpha \sum_f J_{f\sigma} \langle (S_f^z)^2 \rangle \right) \\ &\cdot \langle\langle A_\sigma^1; S_h^- \rangle\rangle + \sum_f J_{f\sigma} \langle\langle 2A_\sigma^2 S_f^z - C_\sigma^2 S_f^+; S_h^- \rangle\rangle. \end{aligned} \quad (9)$$

Here we decouple the various Green functions using Callen's approximation as follows,

$$\langle\langle A_\sigma^i S_f^z; S_h^- \rangle\rangle = \langle S_f^z \rangle \langle\langle A_\sigma^i; S_h^- \rangle\rangle - \gamma\eta \langle S_f^- A_\sigma^i \rangle \langle\langle S_f^+; S_h^- \rangle\rangle, \quad (10)$$

and

$$\begin{aligned} \langle\langle C_\sigma^2 S_f^+; S_h^- \rangle\rangle &= \langle C_\sigma^2 \rangle \langle\langle S_f^+; S_h^- \rangle\rangle - 2\eta\gamma \langle S_\sigma^- S_f^+ \rangle \langle\langle A_\sigma^2; S_h^- \rangle\rangle \\ &- 2\eta\gamma \langle S_\sigma^- S_f^+ \rangle \langle\langle S_\sigma^+; S_h^- \rangle\rangle. \end{aligned} \quad (11)$$

where we take

$$\gamma = \frac{1}{2S} \left(\frac{\langle S^z \rangle}{S} \right). \quad (12)$$

In eq. (11), we made use of the identity,

$$\frac{1}{2} C_\sigma^2 = 2 - (S_\sigma^z)^2 - S_\sigma^- A_\sigma^2 - S_\sigma^- S_\sigma^+.$$

In eqs (10) and (11), $\eta = 0$ corresponds to RPA and $\eta = 1$ corresponds to the additional terms arising from Callen's approximation. Here it may be mentioned that the above identity and hence the decoupling equations (10) and (11) are true for all the three eigenvalues* of S^z (i.e., 1, 0, -1) for $S = 1$ (Kumar and Sharma 1976 b).

We define the Fourier transforms,

$$\begin{aligned}
 G^1(\mathbf{k}, E) &= \sum_{\mathbf{R}_j - \mathbf{R}_k} \langle\langle A_j^+; S_k^- \rangle\rangle \exp i \mathbf{k} (\mathbf{R}_j - \mathbf{R}_k) \\
 J(\mathbf{k}) &= \sum_{\mathbf{R}_j - \mathbf{R}_k} J_{jk} \exp i \mathbf{k} (\mathbf{R}_j - \mathbf{R}_k) \quad \text{and} \\
 \psi^1(\mathbf{k}) &= \sum_{\mathbf{R}_j - \mathbf{R}_k} \langle S_k^- A_j^+ \rangle \exp i \mathbf{k} (\mathbf{R}_j - \mathbf{R}_k)
 \end{aligned}
 \tag{13}$$

Using eqs (10) to (13), eqs (8) and (9) can be written in a matrix form

$$\begin{bmatrix} \omega - 2\sigma J(o, k) - \sigma(\beta_1 - \gamma_1) & -\mathbf{D} \\ Z_2 J(k) - \mathbf{D} + \sigma(\gamma_2 - \beta_1) & \omega - 2\sigma J(o) - \sigma\beta_1 \end{bmatrix} \begin{bmatrix} G^1(\mathbf{k}, E) \\ G^2(\mathbf{k}, E) \end{bmatrix} \\
 = \frac{1}{2\pi} \begin{bmatrix} Z_1 \\ Z_2 \end{bmatrix} \quad \text{where}
 \tag{14}$$

$$\begin{aligned}
 \mathbf{D} &= D + 2\alpha J(o) \langle (S^z)^2 \rangle, \\
 J(k, k') &= J(k) - J(k'); \quad \sigma = \langle S^z \rangle \\
 \beta_i &= \eta \sum_{k'} J(k')^* \psi^1(k'); \quad \gamma_i = \eta \sum_{k'} J(k' - k) \psi^1(k').
 \end{aligned}
 \tag{15}$$

Equation (14) is solved for G^1 and G^2 , and then proceeding in exactly the similar manner as in I, we get the following expressions:

$$\mathbf{D} / \Gamma_0 = 2 J(o) + \frac{3}{4} \mathbf{D} + \beta_1$$

or

$$\langle (S^z)^2 \rangle = \frac{2}{3} + \frac{1}{3} \frac{D + 2\alpha J(o) \langle (S^z)^2 \rangle}{2J(o) + \frac{3}{4}D + \frac{3}{2}\alpha J(o) \langle (S^z)^2 \rangle + \beta_1} \tag{16}$$

The equation determining $k_B T_C / J(o)$ as a function of α and D , is given by

$$\begin{aligned}
 \frac{k_B T_C}{J(o)} &= \frac{4}{3W(1)} \cdot \left[\frac{1 + \frac{D}{4J(o)} + \frac{1}{2}P + \frac{1}{2}\alpha \langle (S^z)^2 \rangle}{1 + \frac{3}{8}\frac{D}{J(o)} + \frac{1}{2}P + \frac{3}{4}\alpha \langle (S^z)^2 \rangle} \right] \\
 &\cdot \left[1 + \frac{A}{W(1)} \left(\frac{\frac{3}{8}\frac{D}{J(o)} + \frac{3}{4}\alpha \langle (S^z)^2 \rangle + \frac{P}{2}}{1 + \frac{3}{8}\frac{D}{J(o)} + \frac{3}{4}\alpha \langle (S^z)^2 \rangle + \frac{P}{2}} \right)^{\frac{1}{2}} + \dots \right] \tag{17}
 \end{aligned}$$

where

$$W(1) = 1.51638 \quad \text{and} \quad A = \frac{3\sqrt{3}}{\sqrt{2}\pi}$$

* In I, the decoupling equations are little different and are true only for $S_z = 1$. However, the final expression of T_c remains unaffected.

for SC system, and

$$P = \frac{\beta_1}{J(o)} = \sum_k \frac{J(k)}{J(o)} \langle S_k^- S_k^+ \rangle$$

$$= \sum_k \frac{J(k)}{J(o)} \cdot \frac{2k_B T}{2J(o) + \frac{3}{4} D + \beta_1 - 2J(k)}$$

which can be simplified so that we get

$$P = \frac{k_B T}{J(o)} \left[\left(1 + \frac{3}{8} \frac{D}{J(o)} + \frac{3}{4} \alpha \langle (S^*)^2 \rangle + \frac{P}{2} \right) \right. \\ \left. \cdot \left[W(1) - A \left(\frac{\frac{3}{8} \frac{D}{J(o)} + \frac{3}{4} \alpha \langle (S^*)^2 \rangle + \frac{P}{2}}{1 + \frac{3}{8} \frac{D}{J(o)} + \frac{3}{4} \alpha \langle (S^*)^2 \rangle + \frac{P}{2}} \right)^{\frac{1}{2}} \right] - 1 \right] \quad (18)$$

Equations (16) to (18) are the final equations which determine, in a self-consistent manner, $k_B T_c/J(o)$ as a function of various parameters.

3. Results and discussion

The variation of critical temperature T_c with the biquadratic exchange parameter α for different values of D , is given in figure 1. Here, we have treated the single-ion anisotropy term in the realistic Hamiltonian given by eq. (1), in an exact manner and the biquadratic exchange and the bilinear Heisenberg exchange has been considered respectively in RPA and in Callen's approximation. Within these approximations, we observe the effect of intersite correlations particularly for small values of parameters. Further, in all the cases T_c increases, initially, with the biquadratic exchange parameter and at larger α , it has the tendency of becoming constant. The increase of T_c vs α is consistent with the initial increase of T_c for small values of α , as predicted by Ditzian and Oitmaa (1974). Since these authors considered only the Ising part in the bilinear exchange, we do not expect any quantitative agreement. However, the qualitative agreement is certainly encouraging. In the limit when $\alpha = 0$ and also $D = 0$, we get $k_B T_c/J(o) = 1.06$ which is very close to the value of Callen (1963). Moreover, we do not expect to get the exact value because of the approximation expressed in eq. (5). It is worth mentioning that the RPA, according to Barma (1974) and Fittipaldi and Tahir Kheli (1975) can give physically incorrect prediction [e.g., $\langle (S^*)^2 \rangle = 0$] whereas in our self-consistent formulation for T_c no such ambiguity is expected to arise while using RPA in Ising part of the biquadratic exchange. Further, because the biquadratic exchange is an indirect one, it is expected to yield a dominant contribution from the intersite correlations. But the mathematics, using the

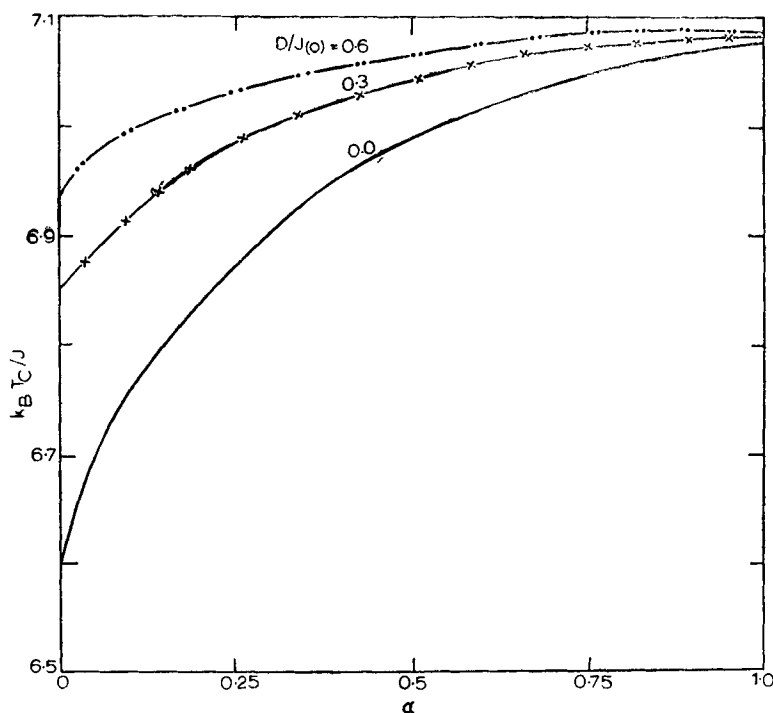


Figure 1. Calculated values of $k_B T_c / J$ as a function of α , for three values (0, 0.3 and 0.6) of $D/J(0)$ for a simple cubic ferromagnetic $S = 1$ system.

Callen's approximation becomes intractable. Still a few important features may be brought out and will be reported later.

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