Magnetic Properties of Layered Heisenberg Ferromagnets

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Abstract

The double-time-temperature spin Green's function method is used to study the magnetic properties of layered ferromagnets with arbitrary spin S, within Tyablikov's decoupling approximation. According to the extent to which interlayer coupling suppresses two-dimensional spin fluctuations, we divide the low-temperature region into two new ones, and give the asymptotic expressions for magnetisation and susceptibility over different temperature regions, including the low-temperature region, the vicinity of the Curie temperature and the high-temperature region. We also give the Curie temperature in an asymptotic form when interlayer coupling is weak.

1. Introduction

Over twenty years have been devoted to the study of layered magnetic materials (see e.g. De Jongh and Miedeman 1974). In particular, the discovery of high- T_c superconductors with layered structure in their parent materials has stimulated greatly the study of this field in connection with superconductivity. The physics of layered quasi-two-dimensional (quasi-2D) magnets is now a well developed branch of magnetism (De Jongh 1990).

Layered magnetic systems are usually described by the Heisenberg model. Several methods have been developed to deal with this model and numerous results have been achieved since it was first proposed. It is well known that long-range ordering (LRO) can only occur at zero temperature for the homogeneous 2D Heisenberg model, but finite interlayer coupling can realise LRO at finite temperatures no matter how small this coupling (Liu 1989). For a system with weak interlayer coupling, although it shows a three-dimensional magnetic-paramagnetic phase transition at temperature T_c , at low temperatures ($T \ll T_c$) it exhibits different temperature characteristics. When the temperature is much lower, the spin thermal fluctuation energy ($\sim k_B T$) may be much smaller than the interlayer coupling strength J_{\perp} , or $k_B T \ll J_{\perp}$, and thus spin fluctuations are suppressed by the interlayer coupling, and the system exhibits 3D temperature characteristics. On the other hand, for $k_B T \gg J_{\perp}$, spin fluctuations are not suppressed sufficiently and the system exhibits quasi-2D temperature characteristics (Singh *et al.* 1990; Kopietz 1992; Du and Wei 1992).

It is generally believed that spin-wave theory is useful at low temperatures. At sufficiently high temperatures, the spin fluctuations become nonlinear and the spin-wave description breaks down. At intermediate temperatures, a quantum Monte Carlo simulation can be used, and the Weiss molecular field theory is useful in the vicinity of the transition temperature. The high-temperature series expansion method is only suitable for high-temperature regions. The double-time-temperature spin Green's function method introduced by Bogolyubov and Tyablikov (1959), and applied to Heisenberg ferromagnets (FMs) with spin $\frac{1}{2}$ by Tyablikov (Tyablikov 1959; Zubarev 1960), proves a very popular theoretical approach to magnetic materials.

This approach agrees with the noninteracting spin-wave theory at very low temperatures and with statistical theory at very high temperatures, and the predicted Curie point in FMs is very close to the other theories. Since the extension of the theory to higher spin by Tahir-Kheli and ter Haar (1962) and improvement of the decoupling approximation (Callen 1963; Tahir-Kheli 1963), the theory has become a useful method to study the Heisenberg model. Using this method, Lines (1964) studied the transition temperature of layered FMs and antiferromagnets with spin $\frac{1}{2}$, while we (Wei and Du 1993) have studied layered antiferromagnets with arbitrary spin S. In this paper, we use this method to study analytically and numerically the suppression by interlayer coupling of 2D spin fluctuation in layered FMs with arbitrary spin S. The fundamental equations are given in Section 2; in Section 3 we discuss spontaneous magnetisation; in Section 4 we discuss the Curie temperature; in Section 5 we discuss susceptibility; and our conclusions are given in Section 6.

2. Fundamental Equations

The Heisenberg ferromagnetism model on a simple cubic lattice with intralayer and interlayer lattice parameters of a and c respectively is given by

$$H = -J \sum_{\langle ij \rangle} S_i \cdot S_j - J_{\perp} \sum_{\langle ij \rangle} S_i \cdot S_j - g\mu_{\rm B} h \sum_j S_j^z \,. \tag{1}$$

Here $\langle ij \rangle$ means the sum over nearest neighbours, J and J_{\perp} are intralayer and interlayer nearest neighbour ferromagnetic interactions respectively, and h is an external magnetic field which, for convenience, is always taken opposite to the positive axis z ($h_z = -h$).

To analyse FMs with arbitrary spin S we introduce, according to Callen (1963), the Green's function

$$G_t^b(i,j) = \left\langle \left\langle S_i^+(t) \,|\, \exp(bS_j^z)S_j^- \right\rangle \right\rangle,\tag{2}$$

where b is a parameter. Using the equation of motion technique for Green's function and the Tyablikov decoupling approximation (Tyabilkov 1959; Zubarev 1960)

$$\langle\langle S_i^+ S_j^z | \exp(bS_j^z) S_j^- \rangle\rangle \to \langle S^z \rangle \langle\langle S_i^+ | \exp(bS_j^z) S_j^- \rangle\rangle, \qquad (3)$$

we have the following expression for the Fourier component of Green's function:

$$G_E^b(k) = \frac{\Sigma}{2\pi [E - E(k)]}; \qquad \Sigma = \langle [S^+, \exp(bS^z)S^-] \rangle, \qquad (4)$$

$$E(k) = g\mu_{\rm B} h + 4J \langle S^z \rangle (2+\delta) [1-\eta(k)], \qquad (5)$$

where

$$\delta = J_{\perp}/J \,, \tag{6}$$

$$\eta(k) = \frac{\cos(k_x a) + \cos(k_y a) + \delta \cos(k_z c)}{2 + \delta}.$$
(7)

Callen's method allows us to derive a self-consistent equation for $\langle S^z \rangle$:

$$\langle S^z \rangle = \frac{(S-n)(1+n)^{2S+1} + (S+1+n)n^{2S+1}}{(1+n)^{2S+1} - n^{2S+1}},$$
(8)

where

$$n = \frac{1}{N} \sum_{k} \frac{1}{\exp[\beta E(k)] - 1}.$$
(9)

With these equations, we can calculate and discuss spontaneous magnetisation (h = 0) and magnetic susceptibility for different interlayer coupling strengths δ .

3. Spontaneous Magnetisation

(3a) Low-temperature Region

When h = 0, the summation over k in equation (9) has been carried out by many authors (Tyabilikov 1959; Zubarev 1960; Tahir-Kheli and ter Haar 1962; Callen 1963; Tahir-Kheli 1963) for the homogeneous case ($\delta = 1$). For the layered structure, we do it by using expansions of Bessel functions instead of the usual long-wavelength approximation, which is very tedious when calculating higher-order terms of temperature. Expanding the Bose distribution function in (9) into a series of $\exp[\beta E(k)]$ and finishing the integral over k, we have

$$n = \sum_{m=1}^{\infty} \exp[-4m(2+\delta)J\beta\langle S^{z}\rangle] \left[I_{0}(4mJ\beta\langle S^{z}\rangle)\right]^{2} I_{0}(4m\delta J\beta\langle S^{z}\rangle), \quad (10)$$

where I_0 and I_1 are the zeroth- and first-order Bessel functions of imaginary argument respectively.

For low-temperature regions we have $T \ll T_1 = 4JS/k_B \sim T_c$. When the interlayer coupling $J_{\perp} (= \delta J)$ is very small, $k_B T$ may be much greater than J_{\perp} , in which case interlayer coupling may not suppress the 2D spin fluctuations sufficiently. We therefore define a characteristic temperature T_0 to distinguish this case: $T_0 = 4J_{\perp} S/k_B$. The low-temperature region is now divided into two new ones: $T \ll T_0 \leq T$, and $T_0 \ll T \ll T_1$.

For $T \ll T_0 \leq T_1$, using the asymptotic expressions for the Bessel functions I_0 and I_1 in (10), we may easily obtain

$$n = \frac{1}{(2\pi R)^{3/2}} \frac{T}{T_1} \left(\frac{T}{T_0}\right)^{1/2} \left[\zeta(\frac{3}{2}) + \frac{2\delta + 1}{8R} \zeta(\frac{5}{2}) \frac{T}{T_0} + \frac{20\delta^2 + 4\delta + 9}{128R^2} \zeta(\frac{7}{2}) \left(\frac{T}{T_0}\right)^2 + \dots\right],$$
(11)

with $R = \langle S^z \rangle / S$ and ζ is the Riemann zeta function.

Expanding the right-hand side of (8) in powers of the small quantity n, we obtain an iterative solution for the small quantity $\langle S^z \rangle - S$,

$$\langle S^{z} \rangle_{1/2} = \frac{1}{2} - \frac{1}{(2\pi)^{3/2}} \frac{T}{T_{1}} \left(\frac{T}{T_{0}} \right)^{1/2} \left[\zeta(\frac{3}{2}) + \frac{2\delta + 1}{8} \zeta(\frac{5}{2}) \frac{T}{T_{0}} \right] + \frac{20\delta^{2} + 4\delta + 9}{128} \zeta(\frac{7}{2}) \left(\frac{T}{T_{0}} \right)^{2} - \frac{1}{(2\pi)_{3}} \zeta(\frac{3}{2}) \left(\frac{T}{T_{1}} \right)^{2} \frac{T}{T_{0}} \left[\zeta(\frac{3}{2}) \right] + \frac{2\delta + 1}{2} \zeta(\frac{5}{2}) \frac{T}{T_{0}} - \dots,$$

$$(12)$$

$$\langle S^{z} \rangle_{S>1} = S - \frac{1}{2} \frac{T}{2} \left(\frac{T}{2} \right)^{1/2} \left[\zeta(\frac{3}{2}) + \frac{2\delta + 1}{2} \zeta(\frac{5}{2}) \frac{T}{2} \right]$$

$$= \frac{20\delta^{2} + 4\delta + 9}{128} \zeta(\frac{7}{2}) \left(\frac{T}{T_{0}}\right)^{2} \left[-\frac{3}{2S(2\pi)^{3}} \zeta(\frac{3}{2}) \left(\frac{T}{T_{1}}\right)^{2} \frac{T}{T_{0}} \right]$$

$$\times \left[\zeta(\frac{3}{2}) + \frac{2\delta + 1}{3} \zeta(\frac{5}{2}) \frac{T}{T_{0}}\right] - \dots$$

$$(13)$$

It is easy to show that the $T^{3/2}$ law is satisfied and these are just the results of a homogeneous simple cubic lattice as $\delta = 1$. Therefore 3D characteristics are reflected in this temperature region. A term T^3 appears in the magnetisation, which does not exist in the spin-wave theory (Dyson 1959), and it can be eliminated by improving the simple decoupling approximation (Callen 1963). For $T_0 \ll T \ll T_1$, transferring $I_0(4m\delta J\beta \langle S^z \rangle)$ in (10) into an integral, and after a lengthy integration, we have

$$n = \frac{1}{2\pi R} \frac{T}{T_1} \left[\ln \left(\frac{2T}{RT_0} \right) + \frac{\zeta(2)}{4R} \frac{T}{T_1} \right] + \dots$$
(14)

We must keep in mind that $T_0 \ll T \ll T_1$ is in the low-temperature region, or $T \ll T_c$, which means $\langle S^z \rangle \to S$, and so *n* is a small quantity, that is $(T/T_1)\ln(2T/RT_0) \ll 1$ for any interlayer coupling strength. In the same way, we can obtain the spontaneous magnetisation

$$\langle S^{z} \rangle_{1/2} = \frac{1}{2} - \frac{1}{2\pi} \frac{T}{T_{1}} \left[\ln \left(\frac{2T}{T_{0}} \right) + \frac{\zeta(2)}{4} \frac{T}{T_{1}} \right] - \frac{2}{(2\pi)^{2}} \left(\frac{T}{T_{1}} \right)^{2} \left[\ln \left(\frac{2T}{T_{0}} \right) + \frac{\zeta(2)}{4} \frac{T}{T_{1}} \ln \left(\frac{2eT}{T_{0}} \right) \right] - \dots,$$
(15)
$$\langle S^{z} \rangle_{s \ge 1} = S - \frac{1}{2\pi} \frac{T}{T_{1}} \left[\ln \left(\frac{2T}{T_{0}} \right) + \frac{\zeta(2)}{4} \frac{T}{T_{1}} \right] \times \left\{ 1 + \frac{1}{2\pi S} \frac{T}{T_{1}} \left[\ln \left(\frac{2eT}{T_{0}} \right) + \frac{\zeta(2)}{2} \frac{T}{T_{1}} \right] \right\} - \dots,$$
(16)

where the terms in the first square brackets in equations (12), (13), (15) and (16) are just the results of linear spin-wave theory (Du and Wei 1992). The temperature dependence of the magnetisation in logarithmic form reflects the quasi-2D magnetic characteristics of the system (Singh *et al.* 1990; Kopietz 1992). Thus, at low temperatures ($T \ll T_c$), for a system with weak interlayer coupling ($J_{\perp} \ll J$), with an increase of temperature above zero the deviation of the magnetisation from its zero-temperature value changes from a $T^{3/2}$ to $T \ln T$ behaviour. When the interlayer coupling is not very weak ($J_{\perp} \rightarrow J$), the temperature region $T_0 \ll T \ll T_1$ does not exist and only the $T^{3/2}$ behaviour is retained.

(3b) Temperatures Just Below the Curie Temperature

Just below the Curie temperature (assuming h = 0), the average magnetisation $\langle S^z \rangle$ is small and n is large. Equation (8) can, therefore, be expanded in inverse powers of n (Tahir-Kheli 1963), the result being

$$\langle S^z \rangle = \frac{2S(S+1)}{3} \left(2n+1 + \frac{C_1}{n} - \frac{C_1}{2n^2} - \dots \right)^{-1},$$
 (17)

where

$$C_1 = \frac{1}{5} \left[\frac{2}{3} S(S+1) - \frac{1}{2} \right].$$
(18)

Furthermore, E(k) is proportional to $\langle S^z \rangle$, and the exponential in the Bose distribution can be expanded, giving

$$1 + 2n = \frac{F(-1)}{2(2+\delta)J\langle S^z \rangle\beta} + \frac{2(2+\delta)J\langle S^z \rangle\beta}{3} - \frac{[2(2+\delta)J\langle S^z \rangle\beta]^3F(3)}{45} + \dots,$$
(19)

where

$$F(m) = \frac{1}{N} \sum_{k} [1 - \eta(k)]^{m}, \qquad (20)$$

$$F(1) = 1$$
, $F(2) = \frac{3\delta^2 + 8\delta + 10}{2(2+\delta)^2}$, $F(3) = \frac{5\delta^2 + 8\delta + 14}{2(2+\delta)^2}$

Using equations (19) and (17), we obtain the magnetisation just below the Curie temperature,

$$\langle S^{z} \rangle = \frac{2S(S+1)}{\{3[F(-1)+6C_{1}]\}^{1/2}} \left[\frac{T}{T_{c}} \left(1 - \frac{T}{T_{c}} \right) \right]^{1/2},$$
 (21)

where the Curie temperature $T_{\rm c}$ is defined by

$$k_{\rm B} T_{\rm c} = \frac{4(2+\delta)JS(S+1)}{3F(-1)} \,. \tag{22}$$

This result is the same as that of Lines (1964) for $S = \frac{1}{2}$; the critical exponent of magnetisation is $\frac{1}{2}$, as in layered antiferromagnets (Wei and Du 1993).

For a layered ferromagnet with $S = \frac{1}{2}$, we have calculated numerically the magnetisation as a function of temperature for $J_{\perp}/J = 0.5$ and 0.0005. The results are shown in Fig. 1, the dotted and dashed lines representing the asymptotic formulae (12) and (15) respectively. The low-temperature characteristics of the magnetisation can be seen clearly as stated above: $J_{\perp}/J = 0.5$ is not very small, so only the $T^{3/2}$ law is retained; for $J_{\perp}/J = 0.0005 \ll 1$, the $T^{3/2}$ law is only valid over a very narrow temperature region, and when the temperature increases $(T \ll T_c)$, the TlnT law agrees with the numerical result very well. This feature is similar to that of a quasi-2D antiferromagnet (Kopietz 1992).

4. Curie Temperature

When the interlayer coupling strength $\delta (=J_{\perp}/J)$ is much smaller than unity, the Curie temperature can be expressed in the asymptotic form (see equation 20)

$$F(-1) = \frac{2+\delta}{N} \sum_{k} \frac{1}{2 - \cos(k_x a) - \cos(k_y a) + \delta[1 - \cos(k_z c)]}.$$
 (23)



Fig. 1. Temperature dependence of magnetisation for two interlayer coupling strengths, $J_{\perp}/J = 0.5$ and 0.0005, corresponding to the Curie temperatures $k_{\rm B} T_{\rm c}/4JS = 0.801$ and 0.285. The dotted and dashed curves represent the asymptotic formulae (12) and (15) respectively. We take $S = \frac{1}{2}$.

Transferring the sum over k into an integral in the first Brillouin zone, and integrating over the two variables k_x and k_y , we have

$$F(-1) = \frac{2+\delta}{\pi^2} \int_0^{\pi} \mathrm{d}k_z \ t \ K(t) \,.$$

where $t = 2/(2 + \delta - \delta \cos k_z)$, and K(t) is the complete elliptic integral of the first kind. In the limit of a small interlayer coupling strength δ , using the asymptotic expression $K(t) = \frac{1}{2} \ln[16/\delta(1 - \cos k_z)]$, then after integrating over k_z we have

$$F(-1) = \frac{2+\delta}{2\pi} \ln\left(\frac{32}{\delta}\right).$$

In the weak coupling limit, we therefore get

$$k_{\rm B} T_{\rm c} = \frac{8\pi J S(S+1)}{3} \bigg/ \ln\left(\frac{32}{\delta}\right),\tag{24}$$

which shows that interlayer coupling J_{\perp} (= δJ) is essential to keep LRO in layered Heisenberg FMs. This formula is slightly different from the one given by Liu (1989), with $32/\delta$ instead of his $8/\delta$. We have also calculated numerically the Curie temperature as a function of interlayer coupling strength based on (22). The result is plotted in Fig. 2, where the dashed curve represents the asymptotic result from (24). It is found that the asymptotic result is very close to the numerical result when $\delta \leq 0.02$.



Fig. 2. Curie temperature T_c as a function of the interlayer coupling strength $\delta = J_{\perp}/J$. The dashed curve represents the asymptotic formula (24).

A pure 2D ferromagnet does not have LRO at finite temperatures. A small interlayer coupling can lead to LRO at nonzero temperatures, and the critical exponent of magetisation is $\frac{1}{2}$, as in 3D homogeneous ferromagnets. Although $J_{\perp} = 10^{-4}J$, the Curie temperature is as large as about 25% of the value for the 3D homogeneous ferromagnet, and such a $T_{\rm c}$ can be detected experimentally (Kopietz 1992).

5. Susceptibility

At a temperature above the Curie temperature, spontaneous magnetisation vanishes and the susceptibility χ is of interest. Considering the linear response of magnetisation to the applied magnetic field, we define χ by

$$\chi = \lim_{h \to 0} \frac{g\mu_{\rm B} \langle S^z \rangle}{h} \,. \tag{25}$$

For small h existing in E(k), $\langle S^z \rangle$ is small and equation (17) is still valid. Expanding n and using (17) and (25), we may obtain the susceptibility χ implicitly in the equation

$$\frac{4J\beta S(S+1)}{3} = \frac{1}{N} \sum_{k} \frac{1}{\lambda + (2+\delta)[1-\eta(k)]},$$
(26)

where

$$\chi = (g\mu_{\rm B})^2 / 4J\lambda \,. \tag{27}$$

Employing a similar relation, Dalton and Wood (1967) studied the susceptibility of the 2D and 3D cases for an Isling-like ferromagnetic model. Yablonskiy (1991) studied the 1D and 2D cases for the Heisenberg model. Tahir-Kheli and ter Haar (1962), Callen (1963) and Tahir-Kheli (1963) also studied the 3D ferromagnet. We shall discuss the susceptibility between the 2D and 3D cases ($0 \le \delta \le 1$) in terms of the above equation.

(5a) Temperatures Just Above the Curie Temperature

When approaching the Curie temperature, $\chi \gg 1$ and accordingly $\lambda \ll 1$. The dominant contribution to the summation in (26) therefore comes from small values of k. Using the long-wavelength approximation (Tahir-Kheli 1963), we have

$$\chi = \frac{(g\mu_{\rm B})^2}{2\delta J} \left(\frac{(2+\delta)T T_{\rm c}}{2\pi T_{\rm M}}\right)^2 \frac{1}{(T-T_{\rm c})^2},$$
(28)

where

$$k_{\rm B} T_{\rm M} = 4(2+\delta) J S(S+1)/3, \qquad (29)$$

indicating that the critical exponent of susceptibility for Heisenberg ferromagnets between the quasi-2D and 3D cases is -2. In the quasi-2D cases ($\delta \rightarrow 0$), using an elliptic integral like (23), equation (26) is changed into the following form:

$$\frac{4J\beta S(S+1)}{3} = \frac{1}{\pi^2} \int_0^{\pi} t \ K(t) \ \mathrm{d}\theta \,, \tag{30}$$

where $t = 2/[2 + \lambda + \delta(1 - \cos\theta)]$.

Using the asymptotic conditions $\lambda \ll 1$ and $\delta \ll 1$, we obtain

$$\chi = \frac{(g\mu_{\rm B})^2}{8\delta J} \left[\sinh\left(\frac{\pi T_{\rm M}(T-T_{\rm c})}{(2+\delta)T T_{\rm c}}\right) \right]^{-2}.$$
(31)

The asymptotic form of this equation for $\delta \neq 0$ is just (28), in which case the coefficient of $(T - T_c)^{-2}$ is much larger.

In the pure 2D cases ($\delta = 0$), $T_c = 0$, and equation (31) becomes

$$\chi = \frac{(g\mu_{\rm B})^2}{64J} \exp\left(\frac{8\pi J S(S+1)}{3k_{\rm B} T}\right).$$
 (32)

This result is same as that of Yablonskiy (1991) and in agreement with that of Dalton and Wood (1967). In the 2D case, the susceptibility is in the form of an exponential with temperature $T \rightarrow 0$.



Fig. 3. Temperature dependence of the inverse susceptibility for several interlayer coupling strengths, where χ is normalised by $(g\mu_{\rm B})^2/J$.

(5b) Temperatures Much Beyond the Curie Temperature

In this temperature region, $\chi \ll 1$, and accordingly $\lambda \gg 1$ and $t \rightarrow 0$. The complete elliptic integral in (30) is approximately $K(t) = \pi (1+t^2/4)/2$. Thus, a high-temperature expansion for the susceptibility is obtained:

$$\chi = \frac{(g\mu_{\rm B})^2 S(S+1)}{3k_{\rm B} T} \left[1 + \frac{T_{\rm M}}{T} + \frac{\delta^2 + 8\delta + 6}{2(2+\delta)^2} \left(\frac{T_{\rm M}}{T}\right)^2 + \frac{4\delta + 2}{(2+\delta)^2} \left(\frac{T_{\rm M}}{T}\right)^3 + \dots \right].$$
(33)

In the pure 2D case ($\delta = 0$), if we take $K(t) = \pi/2$, the Curie–Weiss law for susceptibility is obtained (Yablonskiy 1991).

The numerical result for the inverse susceptibility above T_c as a function of temperature for several interlayer coupling strengths is shown in Fig. 3. Just above the Curie temperature, the inverse susceptibility increases slowly; the weaker the interlayer coupling, the more slowly the inverse susceptibility increases (see equation 28). When the temperature is well beyond the Curie temperature, the inverse susceptibility approximates a straight line, and thus the Curie–Weiss law is satisfied.

6. Conclusion

The effect of interlayer coupling on the magnetic properties of layered ferromagnets with arbitrary spin S has been studied with the use of the double-time-temperature spin Green's function method. According to the extent to which

interlayer coupling suppresses 2D spin fluctuations, we define a characteristic temperature T_0 and divide the low-temperature region $(T \ll T_1)$ into two new ones. With an increase of temperature from one region $(T \ll T_0 \leq T_1)$, to another $(T_0 \ll T \ll T_1)$, the temperature dependence of the magnetisation changes from a $T^{3/2}$ to a TlnT behaviour for small interlayer coupling ($\delta \ll 1$). For a finite interlayer coupling, the critical exponents of magnetisation and susceptibility in the vicinity of the Curie temperature are $\frac{1}{2}$ and -2 respectively. For weak interlayer coupling, the critical exponents of approaches zero for the pure 2D case. At the same time, the susceptibility varies exponentially with temperature when the temperature approaches zero. The high-temperature expansion form for the susceptibility is given in the high-temperature region for arbitrary interlayer coupling strength.

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