EFFECT OF DIPOLE-DIPOLE INTERACTION ON THE THERMODYNAMICS OF A FERROMAGNET NEAR THE CURIE TEMPERATURE

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The method of Vaks, Larkin, and Pikin is used to calculate, to the first order in $1/r_0^3$ (where r_0 is the radius of exchange interaction), the fluctuation corrections to thermodynamic quantities in a Heisenberg ferromagnet (cubic and uniaxial), with allowance for the dipole-dipole interaction of the spins. It is shown that in a cubic crystal, the dipole-dipole interaction does not change the character of the singularity of the fluctuation corrections; as before, they are proportional to $1/r_0^3 \sqrt{\tau}$, where $\tau = |T - T_C|/T_C$. In the case of uniaxial crystals, the anisotropy of the Lorentz field causes, at sufficiently small τ, a change from a root singularity to a logarithmic.

1. The method of the molecular field is appropriate for description of the properties of ferromagnets in the immediate neighborhood of the Curie temperature T_C (that is, when $\tau = |T - T_C|/T_C \ll 1$) if the radius of interaction of the atomic spins is sufficiently large in comparison with the lattice constant $d^{[1,2]}$. Vaks, Larkin and Pikin[2] showed that in a Heisenberg ferromagnet, the corrections to thermodynamic quantities that result from fluctuations form a power series in the parameter $1/r_0^3 \sqrt{\tau}$, where r_0 is the radius of exchange interaction of the spins in units of d. Since fluctuations of wavelengths $\lambda \gtrsim d/\sqrt{\tau}$ play a fundamental role in the correlation functions, it follows that at sufficiently small τ—such that $\lambda \gtrsim d v_0 T_C /4\pi \mu^2 S^2$, where v_0 is the volume of the elementary cell, μ is the Bohr magneton, and S is the spin of the atom—, the long-range dipole-dipole interaction will become important.

For this reason it has been suggested[1,3,4] that the dipole-dipole interaction may appreciably extend (on the small-τ side) the range of applicability of the molecular-field method.

In the present paper, the method developed in[2] is used to calculate the first-order fluctuation corrections to thermodynamic quantities in a Heisenberg ferromagnet (cubic and uniaxial) with allowance for the dipole-dipole interaction. We have taken account of the magnetocrystalline anisotropy only to the extent that it is determined by the Lorentz field. Also neglected are the effects of the crystal surface and of the domain structure, since the wavelengths important to the problem are considerably shorter than the dimensions of the domains.

2. The initial Hamiltonian has the following form:

$$\mathcal{H} = -\mu \sum_r S_r H - \frac{1}{2} \sum_{r \neq r'} V(r - r') S_r S_{r'} - 2 \sum_{r \neq r'} D_{\alpha\beta}(r - r') S_r^\alpha S_{r'}^\beta, \quad (1)$$

where

$$D_{\alpha\beta}(r - r') = \mu^2 \frac{\partial^2}{\partial r_\alpha \, \partial r_\beta} \frac{1}{|r - r'|}, \quad (2)$$

and where S_r is the spin of the atom on site r; the first term in (1) is the Zeeman energy of the spins in the external magnetic field H, the second describes the exchange interaction, and the third describes the dipole-dipole interaction.

It is convenient to divide the Hamiltonian (1) into $\mathcal{H}_0$, corresponding to the molecular-field approximation, and $\mathcal{H}_1$, describing the interaction of the fluctuations:

$$\mathcal{H}_0 = \frac{N \langle S_z \rangle^2}{2} W_0 - (\langle S_z \rangle W_0 + \mu H) \sum_r S_r^z; \quad (3)$$

$$\mathcal{H}_1 = -\frac{1}{2} \sum_{r \neq r'} W_{\alpha\beta}(r - r')(S_r^\alpha - \langle S_\alpha \rangle)(S_{r'}^\beta - \langle S_\beta \rangle). \quad (3')$$

Here

$$W_0 = \sum_r [V(r) + D_{zz}(r)] = V_0 + \frac{4\pi \mu^2}{v_0} l_z. \quad (4)$$

The second term in (4) is the energy of a dipole in the Lorentz field; $l_x + l_y + l_z = 1$. In a cubic crystal, $l_x = l_y = l_z = 1/3$;

$$W_{\alpha\beta}(r - r') = V(r - r') \delta_{\alpha\beta} + D_{\alpha\beta}(r - r'). \quad (5)$$

The magnetic field H is directed along the axis Oz. Hereafter we assume for simplicity that in a uniaxial crystal, Oz coincides with the axis and $l_z > l_x = l_y$.

3. We consider the correlator

$$K_{\alpha\gamma}(k, \omega_n) = \frac{1}{2\beta} \int_{-\beta}^{\beta} e^{i\lambda\omega_n} \, d\lambda \sum_r e^{ikr} \quad (6)$$
$$\times \langle (S_r^\alpha(\lambda) - \langle S_\alpha \rangle), (S_{r'}^\gamma(0) - \langle S_\gamma \rangle) \sigma(\beta) \rangle,$$

where

$$S_r^\alpha(\lambda) = e^{\mathcal{H}_0 \lambda} S_r^\alpha e^{-\mathcal{H}_0 \lambda},$$

$$\sigma(\beta) = T_\lambda \exp \left\{ -\int_0^\beta \mathcal{H}_1(\lambda) \, d\lambda \right\},$$

$$\langle \cdots \rangle = \mathrm{Sp} \{ e^{-\beta \mathcal{H}_0} \cdots \} \{ \mathrm{Sp} \, e^{-\beta \mathcal{H}_0} \}^{-1},$$

T_λ is the ordering symbol, $\beta = 1/T$, $\omega_n = 2\pi n T$, and α and $\gamma = +, -, z$.

In the approximation of first order in $1/r_0^3$ for $K_{\alpha\gamma}(k, \omega_n)$, we get the three systems of equations

$$G_{\alpha\gamma}(\omega_n) + G_{\alpha\nu}(\omega_n) W_{\nu\mu}(k) K_{\mu\gamma}(k, \omega_n) = K_{\alpha\gamma}(k, \omega_n), \quad (7)$$

where

$$y = \beta(W_0 \langle S_z \rangle + \mu H), \quad (8)$$

$$W_{\alpha\gamma}(k) = \sum_r e^{ikr} W_{\alpha\gamma}(r), \quad (9)$$

and the nonvanishing components $G_{\alpha\beta}(\omega_n)$ are[2]

$$G_{+-}(\omega_n) = G_{-+}^*(\omega_n) = b/(y - i\beta\omega_n),$$

$$G_{zz}(\omega_n) = \delta_{n0}b' = \delta_{n0}db/dy. \tag{10}$$

The value of y is determined by the condition for self-consistency

$$\langle S_z \rangle = (y - \mu\beta H)/\beta W_0 = b(y). \tag{11}$$

The determinant of each of the systems (7) has the form

$$\Delta(\mathbf{k}, \omega_n) = [|1 - \beta(V_k - \tfrac{1}{2}D_{zz}(\mathbf{k}))G_{+-}(\omega_n)|^2 \tag{12}$$
$$- |G_+ B_k|^2][1 - \beta(V_k + D_{zz}(\mathbf{k}))G_{zz}(\omega_n)]$$
$$- 2G_{zz}(\omega_n)\,\mathrm{Re}\{G_{+-}[1 - \beta(V_k - \tfrac{1}{2}D_{zz}(\mathbf{k}))G_{-+}(\omega_n)]|C_k|^2$$
$$+ C_k^2 B_k^*|G_{+-}(\omega_n)|^2\},$$

where

$$B_k = \tfrac{1}{2}\beta[D_{xx}(\mathbf{k}) - D_{yy}(\mathbf{k}) - 2iD_{xy}(\mathbf{k})], \tag{13}$$
$$C_k = 2^{-1/2}\beta[D_{xz}(\mathbf{k}) - iD_{yz}(\mathbf{k})],$$
$$V_k = \sum_r e^{ikr}V(\mathbf{r}).$$

On substituting (10) in (12), we get for $\omega_n \neq 0$

$$\Delta(\mathbf{k}, \omega_n) = \frac{A_k^2 - b^2|B_k|^2 - (i\beta\omega_n)^2}{y^2 - (i\beta\omega_n)^2}, \tag{14}$$

where

$$A_k = y - \beta[V_k - \tfrac{1}{2}D_{zz}(\mathbf{k})]b. \tag{15}$$

From (14) follows the expression for the energy Ω_k of the spin waves,

$$\beta^2\Omega_k^2 = A_k^2 - b^2|B_k|^2. \tag{16}$$

The expression (16) describes the spectrum of spin waves whose dispersion is dependent both on exchange and on dipole-dipole interactions. In the absence of exchange interaction and at T = 0 this spectrum coincides with the spectrum of a dipole-dipole ferromagnet, investigated by Cohen and Keffer[5]. The dispersion dependent on dipole-dipole interaction is important if $V_0 \lesssim \mu^2/v_0$. When $V_0 \gg \mu^2/v_0$, it is negligible, and then[5,6]

$$D_{\alpha\beta}(\mathbf{k}) = \frac{4\pi\mu^2}{v_0}\left(l_\alpha\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2}(1 - \delta_{k0})\right), \tag{17}$$

so that for a uniaxial crystal ($l_z > l_x = l_y$) the spin-wave spectrum for $k \neq 0$ takes the form

$$\Omega_k^2 = \left[b(V_0 - V_k) + \mu H + 4\pi\frac{\mu^2}{v_0}b\sin^2\vartheta_k + 2\pi\frac{\mu^2}{v_0}b\delta\right] \tag{18}$$
$$\times\left[b(V_0 - V_k) + \mu H + 2\pi\frac{\mu^2}{v_0}b\delta\right],$$

where

$$\delta = 3l_z - 1, \qquad \cos\vartheta_k = k_z/k. \tag{19}$$

Thus anisotropy of the Lorentz field produces a gap in the spectrum. This gap is proportional to b(y), and consequently it decreases as $\langle S_z \rangle$ near the Curie temperature. In a cubic crystal, $\delta = 0$, and the expression (18) at $T \ll T_C$ (then b(y) = S) reduces to the usual spin-wave spectrum. In uniaxial crystals, the value of δ depends on the specific structure. For a hexagonal close-packed lattice, δ is small ($\approx 10^{-3}$)[7]; in simple hexagonal and tetragonal lattices, δ depends on the ratio between the lattice constants and can be ~ 1[8,9]. We shall see later that the character of the singularities of the fluctuation corrections to thermodynamic quantities near the transition depends significantly on the value of δ.

4. To calculate the fluctuation correction $F^{(1)}$ to the free energy, we use the procedure explained in[10]; namely, we replace $\mathscr{H}_1$ in the interaction Hamiltonian by $g\mathscr{H}_1$, where $0 \leq g \leq 1$; then

$$\frac{dF^{(1)}}{dg} = -\frac{1}{2}\sum_{k,n}\mathrm{Sp}\,W_{\alpha\beta}(\mathbf{k})K_{\beta\alpha}(\mathbf{k}, \omega_n, g). \tag{20}$$

The equation for $K_{\alpha\beta}(\mathbf{k}, \omega_n, g)$ is obtained from (7) by replacing $W_{\nu\mu}(\mathbf{k})$ in it by $gW_{\nu\mu}(\mathbf{k})$. It is easy to show that the determinant $\Delta(\mathbf{k}, \omega_n, g)$ of this modified equation is related to the right side of (20) as follows:

$$-\sum_{k,n}'\mathrm{Sp}\,W_{\alpha\beta}(\mathbf{k})K_{\beta\alpha}(\mathbf{k}, \omega_n, g) = \frac{d}{dg}\ln\Delta(\mathbf{k}, \omega_n, g), \tag{21}$$

so that on integrating with respect to g, we get

$$\beta F^{(1)} = \frac{1}{2}\sum_{k,n}\ln\Delta(\mathbf{k}, \omega_n). \tag{22}$$

On performing the summation over discrete frequencies, we find finally that

$$\beta F^{(1)} = \sum_k\ln\left[\frac{y}{\Omega_k}\frac{\mathrm{sh}(\Omega_k/2)}{\mathrm{sh}(y/2)}\right]$$
$$+ \frac{1}{2}\sum_k\ln\frac{1}{y^2}\{\beta^2\Omega_k^2[1 - \beta(V_k + D_{zz}(\mathbf{k}))b'] \tag{23}$$
$$- 2bb'(A_k|C_k|^2 + b\,\mathrm{Re}\,B_k^*C_k^2)\}.$$

The first term in this expression is obtained by summation of n from ± 1 to $\pm\infty$; the second is the contribution of $\Delta(\mathbf{k}, 0)$, that is of the denominator of $K_{zz}(\mathbf{k}, 0)$. The structure of this particular term is important for later purposes.

5. We turn to the presentation of the results of the calculation of the mean moment $\langle S_z \rangle$ and of the heat capacity C_H near the Curie temperature. The dipole-dipole interaction plays a role if $\tau \ll \eta$, where $\eta = 4\pi\mu^2/v_0V_0$. For a cubic crystal, by use of the expression for the free energy $\beta F^{(0)}$ in the molecular-field approximation[2] and of the fluctuation correction (23), we get

a) $T > T_C$, $ch^2/3a\tau^3 \ll 1$:

$$\langle S_z \rangle = \frac{ha}{\tau}\left(1 + \frac{10}{9}\frac{\gamma}{\sqrt{\tau}}\right), \tag{24}$$

$$C_H = \frac{a^2}{c}\left(\frac{1}{2}\frac{\gamma}{\sqrt{\tau}} + \frac{ch^2}{a\tau^3} + \frac{\gamma}{\tau^{1/2}}\frac{5}{3}\frac{ch^2}{a}\right);$$

b) $T < T_C$, $ch^2/3a\tau^3 \ll 1$:

$$\langle S_z \rangle = \sqrt{\frac{3a^3\tau}{c}}\left[1 + \frac{1}{2}\left(\frac{ch^2}{3a\tau^3}\right)^{1/2} + \frac{5\pi}{24}\frac{\gamma}{\sqrt{2\tau}} + \frac{1}{6}\frac{\gamma}{\sqrt{\tau}}\left(\frac{ch^2}{3a\tau^3}\right)^{1/4}\right],$$

$$C_H = \frac{a^2}{c}\left[\frac{3}{2} + \frac{3\pi}{16}\frac{\gamma}{\sqrt{2\tau}} + \frac{7}{16}\frac{\gamma}{\sqrt{\tau}}\left(\frac{ch^2}{3a\tau^3}\right)^{1/4}\right]. \tag{25}$$

Here the following abbreviations are used: a = S(S + 1)/3, $c = \tfrac{3}{5}a^2 + \tfrac{1}{10}a$, $\gamma = 3c\sqrt{6}/2\pi r_0^3 a^2$, $h = \mu H/T_C$. The Curie temperature is $T_C = aW_0 + O(\gamma)$. We shall not write out the terms $O(\gamma)$, since they are cumbersome and since we shall not need them hereafter.

From a comparison of (24) and (25) with the corresponding formulas from paper[2], it is seen that in a cubic crystal the dipole-dipole interaction does not change either the character of the singularity in the fluctuation corrections or their dependence on the magnetic field. It leads only to a certain decrease of the

numerical factor that multiplies $\gamma/\sqrt{\tau}$. Thus the criterion for applicability of the molecular-field approximation remains, as before, the inequality $\gamma/\sqrt{\tau} \ll 1$.

A different situation arises in the case of uniaxial crystals at temperatures $\tau \ll \eta\delta$. Here the root singularity of the fluctuation corrections is replaced by a logarithmic one. The following expressions for $\langle S_z \rangle$ and C_H are obtained, to within terms small in comparison with $\ln(\eta\delta/\tau)$:

a) $T > T_c$, $ch^2/3a\tau^3 \ll 1$:

$$\langle S_z \rangle = \frac{ah}{\tau}\left(1 - \frac{ch^2}{3a\tau^3}\right)\left[1 + \frac{\gamma}{4}\sqrt{\frac{2+\delta}{\eta\delta}}\ln\frac{\eta\delta}{\tau}\right], \quad (26)$$

$$C_H = \frac{a^2}{8c}\gamma\sqrt{\frac{2+\delta}{\eta\delta}}\left(1 + 2\frac{ch^2}{a\tau^3}\right)\ln\frac{\eta\delta}{\tau},$$

b) $T < T_c$, $ch^2/3a\tau^3 \ll 1$:

$$\langle S_z \rangle = \sqrt{\frac{3a^3\tau}{c}}\left(1 + \frac{h}{2}\sqrt{\frac{c}{3a\tau^3}}\right)\left(1 + \frac{\gamma}{4}\sqrt{\frac{2+\delta}{\eta\delta}}\ln\frac{\eta\delta}{\tau}\right), \quad (27)$$

$$C_H = \frac{a^2}{c}\left[\frac{3}{2} - \frac{\sqrt{3}}{4}\left(\frac{ch^2}{a\tau^3}\right)^{1/2} + \frac{\gamma}{8}\sqrt{\frac{2+\delta}{\eta\delta}}\left(4 - \frac{1}{2}\sqrt{\frac{3ch^2}{a\tau^3}}\right)\ln\frac{\eta\delta}{\tau}\right].$$

The condition for applicability of these expansions is the inequality

$$\frac{\gamma}{\sqrt{\eta\delta}}\ln\frac{\eta\delta}{\tau} \ll 1, \quad (28)$$

which in the temperature range considered, $\tau \ll \eta\delta$, is considerably weaker than the inequality $\gamma \ll \sqrt{\tau}$. When $\tau \gg \eta\delta$, in a uniaxial crystal as in a cubic, the expansion is carried out in powers of $\gamma/\sqrt{\tau}$.

The expansions (26) and (27) have a range of applicability if γ is sufficiently small, since then the inequality $\gamma \ll \sqrt{\tau}$ is satisfied until $\tau \approx \eta\delta \ll 1$. In what ferromagnets these conditions may be satisfied is in general not apparent. Favorable in this respect are ferromagnets in which a large role is played by indirect exchange, in particular via the conduction electrons (in this case it is to be expected that the correlation radius will be sufficiently large), and also ferromagnets with small T_c and consequently not too small η.

Our results for ferromagnets are quite similar to the results of paper[10] for ferroelectrics. In ferroelectrics as in ferromagnets, the dipole-dipole interaction changes the character of the singularity in the fluctuation terms only in noncubic crystals. But whereas in ferroelectrics this result does not depend on the anisotropy of the Lorentz field, in ferromagnets precisely this anisotropy plays a fundamental role. For ferromagnets in which $\delta \ll 1$, the range of applicability of formulas (26) and (27) is greatly narrowed, giving place to an expansion in powers of $\gamma/\sqrt{\tau}$.

6. In closing, we make a few qualitative remarks.

We shall suppose that the interaction consists of two isotropic parts V_k and V'_k with radii r_0 and R_0 respec-

tively, and with $R_0 \gg r_0$. In this case a Fourier component of the correlator $K_{zz}(k, 0)$ near the Curie temperature has the form

$$\frac{a}{(W_0 - W_k)/W_0 + 2\tau}$$

where $W_k = V_k + V'_k$. Analysis of the expression for $K_{zz}(k, 0)$, which basically determines the character of the singularity of the fluctuation terms in thermodynamic quantities, shows that in the interval $\eta(r_0/R_0)^6 \ll \tau \ll \eta = V'_0/W_0$, an expansion can be carried out in the parameter $1/r_0^3\eta^{1/2}$. This result follows immediately from the form of the denominator of $K_{zz}(k, 0)$ for wave vectors $1/R_0 \ll k \ll 1/r_0$:

$$(kr_0)^2 + \eta + 2\tau.$$

As is evident, in this case $K_{zz}(k, 0)$ differs from the Ornstein-Zernike correlator by the presence in the denominator of the constant gap η. Since the dipole-dipole interaction has an infinite radius of influence, it might be expected that the expansion parameter $1/r_0^3\eta^{1/2}$ would apply for all $\tau \ll \eta$. Actually, as we have seen, this is not so. This is explained by the fact that in a cubic crystal, the contributions to the gap from the diagonal components $D_{\alpha\alpha}(k)$ and the nondiagonal components $D_{\alpha\beta}(k)$ of the dipole-dipole interaction tensor compensate each other. In a uniaxial crystal, however, there remains a contribution proportional to $\eta\delta\cos^2\theta_k$. The angular dependence of the size of the gap leads to the appearance of $\ln\tau$.

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