GROUND STATE AND LOW-TEMPERATURE WEAK-FIELD THERMODYNAMICS OF A UNIAXIAL FERROMAGNET WITH TRANSVERSAL MAGNETIC FIELD

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The most suitable spin-deviation reference state (approximate ground state) for a uniaxial ferromagnet with an external magnetic field perpendicular to the anisotropy axis is determined by applying three different methods (A, B, C) which are shown to be equivalent for weak and strong fields. By utilizing these results the low-temperature weak-field expansion of the magnetization is carried through by means of Wallace's statistical perturbation method. In the isotropic case Dyson's T^4 — term is recovered. The anisotropy is shown to give contributions proportional to T^2 and T^3.

1. Introduction

A crucial problem in the application of the standard longwavelength low-temperature spin-wave approximation to the Heisenberg model of anisotropic ferromagnets is the choice of a suitable reference state, that is the state from which the system's elementary excitations are generated [1]. It is obvious that the adequacy of the single-particle spinwave energies and the convergence of the perturbation procedure depends decidedly on the proper choice of the reference state. For isotropic ferromagnets and certain special cases of (uniaxial) anisotropic ferromagnets with the external field parallel to the anisotropy axis the exact ground state is known to be the state of complete spin-alignment (spin-deviation vacuum), and its choice as reference state is in this case doubtlessly fully justified. In general, however, the spin-deviation vacuum is not the ground state of an anisotropic ferromagnet, particularly if the external magnetic field is not parallel to one of the magnetically easiest directions. What is more, the exact ground state in this case is unknown and its determination appears to be impossible. Nevertheless, even in this case one usually assumes a state of complete spin alignment as the spin-deviation reference state, apparently for reasons of mathematical simplicity [2, 3, 4]. In determining the favourable direction of spin alignment, three different

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methods have been examined in [5] for a quite general spin Hamiltonian: the method A which resides in minimizing the expectation value of the Hamiltonian in the class of states generated by spatial rotations from the state of complete spin alignment, the method B which is based on eliminating from the transformed Hamiltonian the terms linear with respect to spinwave creation and destruction operators, and the method C which consists of the minimization of the ground state energy of the diagonalized spinwave Hamiltonian in the non-interacting spin wave approximation.

In Sections 2 and 3 of the present paper these three methods are applied to a uniaxial ferromagnet with an external magnetic field perpendicular to the anisotropy axis, and the equivalence of methods A and B proven in [5] is here demonstrated. Moreover, the method C is shown to be approximately equivalent to methods A and B under the restriction to weak ($h \ll 2SG(0)$) and strong ($h \gg 2SG(0)$) fields.

Upon determining the reference state we use in Section 4 Wallace's statistical perturbation method [6] in the first approximation, in order to calculate the low-temperature weak-field expansion of the magnetization. In the nearest-neighbour approximation, explicit results are given which are correct to T^4 and h^2. In the limit case of an isotropic ferromagnet we obtain the correct low-temperature expansion of Dyson [7]. It is shown that due to the anisotropy there appear in the expansion terms proportional to T^2 and T^3.

2. *Methods A and B*

We consider the following Hamiltonian of a uniaxial ferromagnet in a homogeneous external magnetic field perpendicular to the anisotropy axis:

$$H = -h \sum_j \tilde{S}_j^x - \sum_{j,k} I_{jk} \tilde{\vec{S}}_j \cdot \tilde{\vec{S}}_k - \sum_{j,k} G_{jk} \tilde{S}_j^z \tilde{S}_k^z. \tag{2.1}$$

The anisotropy axis and the field direction are assumed to be parallel to the z- and x-axis of our co-ordinate system, respectively. According to the conditions (*i*) and (*ii*) of [5] we impose upon the isotropic I and anisotropic G exchange integrals translational invariance and exclude intra-atomic interactions, *i. e.*,

$$I_{jk} = I(j-k) = I(f),\ G_{jk} = G_{jk}(j-k) = G(f)$$
$$I_{jj} = G_{jj} = 0.$$

$\tilde{\vec{S}}_j = (\tilde{S}_j^x, \tilde{S}_j^y, \tilde{S}_j^z)$ denotes the spin vector operator ascribed to the lattice site j, and $h = \mu H_x$ where H_x is the field component in the x-direction and μ the magnetic moment per lattice atom.

Next we perform the following homogeneous rotation of the spins in the $x0z$-plane by the angle φ:

$$\begin{aligned} \tilde{S}_j^x &= \gamma_x S_j^z + \gamma_z S_j^x \\ \tilde{S}_j^y &= S_j^y \\ \tilde{S}_j^z &= \gamma_z S_j^z - \gamma_x S_j^x \end{aligned} \tag{2.2}$$

where $\gamma_x = \sin \varphi$ and $\gamma_z = \cos \varphi$.

The transformation (2.2) is a special case ($\vartheta = 0$) of the general homogeneous spatial rotation (10) in [5]. It can be assumed without loss of generality because of the rotational symmetry of the Hamiltonian (2.1) around the z-axis. The Hamiltonian (2.1) in the new spin operators S_j^x, S_j^y, S_j^z reads:

$$H = -h\gamma_z \sum_j S_j^x - h\gamma_x \sum_j S_j^z - \sum_{j,k} I_{jk}\vec{S}_j \cdot \vec{S}_k - \sum_{j,k} G_{jk}(\gamma_z^2 S_j^z S_k^z - 2\gamma_x\gamma_z S_j^x S_k^z + \gamma_x^2 S_j^x S_k^x). \quad (2.3)$$

By using the Holstein-Primakoff mapping [8] from the spin operators $\vec{S}_j$ to Bose creation and annihilation operators a_j, a_j^+ in the second approximation, *i. e.*,

$$\begin{aligned} S_j^+ = (S_j^-)^+ = S_j^x + iS_j^y &\to \sqrt{2S}\,\sqrt{1 - a_j^+ a_j/2S}\; a_j \\ &= \sqrt{2S}\,(1 - a_j^+ a_j/4S - \ldots)\, a_j, \\ S_j^z &\to S - a_j^+ a_j \end{aligned} \quad (2.4)$$

and upon passing to ideal spin wave operators a_ν^+, a_ν corresponding to the wave vector ν according to the Fourier transformation[1]

$$a_j^{(+)} = \frac{1}{\sqrt{N}} \sum_\nu a_\nu^{(+)} e^{(-)ij\nu} \quad (2.5)$$

we obtain in the ideal spin wave representation the following Hamiltonian:

$$H = E_0 + H_1 + H_2 + H_3 + H_4 + \ldots \quad (2.6)$$

where

$$E_0 = -h\gamma_x NS - NS^2 J(0) - \gamma_z^2 NS^2 G(0) \quad (2.7)$$

$$H_1 = R(a_0^+ + a_0) \quad (2.8)$$

$$H_2 = \sum_\nu L_\nu a_\nu^+ a_\nu + \tfrac{1}{2} \sum_\nu M_\nu (a_\nu^+ a_{-\nu}^+ + a_\nu a_{-\nu}) \quad (2.9)$$

$$H_3 = \sum_{\nu\mu\lambda} \delta(\nu + \mu - \lambda)\, N_\mu a_\nu^+ a_\mu a_\lambda + \sum_{\nu\mu\lambda} \delta(\nu - \mu - \lambda)\, N_\mu a_\nu^+ a_\mu a_\lambda \quad (2.10)$$

$$\begin{aligned} H_4 = &\sum_{\nu\mu\lambda\varkappa} \delta(\nu + \mu - \lambda - \varkappa)\, F_{\nu\varkappa} a_\nu^+ a_\mu^+ a_\lambda a_\varkappa + \sum_{\nu\mu\lambda\varkappa} \delta(\nu - \mu - \lambda - \varkappa) k_\mu a_\nu^+ a_\mu a_\lambda a_\varkappa + \\ &+ \sum_{\nu\mu\lambda\varkappa} \delta(\nu + \mu + \lambda - \varkappa)\, k_\mu a_\nu^+ a_\mu^+ a_\lambda^+ a_\varkappa \end{aligned} \quad (2.11)$$

with the notation

$$R = \tfrac{1}{2}\sqrt{2NS}\,(2S\gamma_x\gamma_z G(0) - h\gamma_z) \quad (2.12)$$

$$L_\nu = h\gamma_z + 2S(J(0) - J(\nu) + 2S\gamma_z^2 G(0) - S\gamma_x^2 G(\nu) \quad (2.13)$$

$$M_\nu = -S\gamma_x G(\nu) \quad (2.14)$$

$$N_\nu = \sqrt{2S/N}(h\gamma_z/8S - \gamma_x\gamma_z G(\nu) - {}^1/_4\gamma_x\gamma_z G(0)) \quad (2.15)$$

$$F_{\nu\varkappa} = N^{-1}/2(J(\nu) + J(\varkappa) - 2J(\nu - \varkappa) + {}^1/_2\gamma_x^2 G(\nu) + {}^1/_2\gamma_x^2(\varkappa) - 2\gamma_z^2 G(\nu - \varkappa)) \quad (2.16)$$

$$k_\nu = N^{-1}/8\;\gamma_x^2 G(\nu). \quad (2.17)$$

[1] We use a simplified notation according to which $j\nu$ denotes the scalar product of the lattice vector j and the wave vector ν.

We neglect in the Hamiltonian (2.6) higher than fourth-order interaction terms. In the above formulae, $J(\nu)$ and $G(\nu)$ denote the Fourier transforms of the exchange integrals I_{jk} and G_{jk}:

$$J(\nu) = \sum_f I(f)\, e^{if\nu}, \tag{2.18a}$$

$$G(\nu) = \sum_f G(f)\, e^{if\nu}, \tag{2.18b}$$

N is the number of lattice sites, and S the maximum spin eigenvalue.

The method A of [5] resides here in minimizing the quantity E_0, Eq. (2.7), with respect to the angle φ. One easily proves that E_0 is equal to the expectation value of the Hamiltonian (2.1) in the saturation state of complete spin alignment along the direction given by φ, which in turn is equal to the classical counterpart of the spin Hamiltonian following from (2.3) by assuming the spin-vector operators to be classical spin vectors.

The necessary condition for the minimum of E_0 leads to the equation

$$dE_0/d\varphi = -hNS\cos\varphi + 2NS^2\cos\varphi\sin\varphi\; G(0) = 0 \tag{2.19}$$

or

$$\cos\varphi\,(2SG(0)\sin\varphi - h) = 0. \tag{2.20}$$

The solutions of Eq. (2.20) are

$$\begin{aligned} &\text{(a)} \quad \sin\varphi = 1 \\ &\text{(b)} \quad \sin\varphi = -1 \\ &\text{(c)} \quad \sin\varphi = h/2SG(0). \end{aligned} \tag{2.21}$$

The sufficient condition for the minimum of E_0,

$$d^2E_0/d\varphi^2 > 0, \tag{2.22}$$

leads to the inequality

$$h\sin\varphi - 2SG(0)\,(\sin^2\varphi - \cos^2\varphi) > 0. \tag{2.23}$$

If the inequality (2.23) is satisfied by more than one of the solutions (2.21), the one corresponding to the lowest energy E_0 must be chosen (when neglecting meta-stable states). Thus, depending on the sign of h and $G(0)$ we get the following solutions of Eq. (2.20) corresponding to the absolute minimum of E_0:

$$\begin{aligned} &(\alpha)\;\; G(0) > 0,\, h < 0;\;\; h/2SG(0) \geqslant 1: && \text{(a)} \\ &\qquad\qquad\qquad\qquad\quad h/2SG(0) \leqslant 1: && \text{(c)} \\ &(\beta)\;\; G(0) > 0,\, h > 0;\;\; |h/2SG(0)| \geqslant 1: && \text{(b)} \\ &\qquad\qquad\qquad\qquad\quad |h/2SG(0)| \leqslant 1: && \text{(c)} \\ &(\gamma)\;\; G(0) < 0,\, h > 0: && \text{(a)} \\ &(\delta)\;\; G(0) < 0,\, h < 0: && \text{(b)} \end{aligned} \tag{2.24}$$

The analysis of the third and fourthderivative of E_0 with respect to φ shows that for the equality sign in either case, (α) and (β), both solutions correspond to a minimum. At this point ($h = h_c = 2SG(0)$) a second-order field-induced phase transition takes place. In each case the first solution describes the paramagnetic phase, and the second one the ferromagnetic phase — in the sense of [9, 10]. For $G(0) < 0$ (the cases (γ) and (δ)) only the ferromagnetic phase exists, regardless of the field strength (magnetically preferred plane). This is in accordance with the conditions for the second-order phase transition in uniaxial ferromagnets derived in [9–11].

The method B of [5] resides in eliminating the terms linear with respect to the spin wave operators in the transformed Hamiltonian, that is in our case $R = 0$, Eq. (2.12). We see that this method leads to the necessary minimum condition (2.20) of method A, in agreement with the general proof given in [5] for the limited equivalence of both methods.

3. *Method C*

In [5] a further method for determining a suitable reference state has been proposed which consists in the minimization of the ground state energy in the non-interacting spin wave approximation. This procedure shall be applied, with a slight variation, to the Hamiltonian (2.6).

At first we remove the terms linear with respect to the Bose operators from (2.6), by means of the canonical transformation [1.5]

$$a_\nu^+ = \alpha\delta_{\nu 0} + b_\nu^+ \tag{3.1}$$

where α is a real c-number. Upon inserting (3.1) in (2.6) we neglect higher than second-order terms with respect to b_ν, b_ν^+ (non-interacting-spin-waves approximation). Furthermore, we drop terms $0(\alpha^2)$ generated by $H_3, H_4, \ldots$, whereas the quadratic contribution of H_2 to the ground state energy is included. This approximation is justified if $S^{-1} \ll 1$ and

$$|\alpha| \ll 1. \tag{3.2}$$

(Note that (2.6) represents a series of decreasing powers of S.)

Under the above assumptions the Hamiltonian (2.6) takes the form:

$$H = E_0 + \delta E_0 + \bar{R}(b_0^+ + b_0) + \sum_\nu \bar{L}_\nu b_\nu^+ b_\nu + {}^1/_2 \sum_\nu \bar{M}_\nu (b_\nu^+ b_{-\nu}^+ + b_\nu b_{-\nu}) \tag{3.3}$$

with

$$\delta E_0 = 2\alpha R + \alpha^2(L_0 + M_0), \tag{3.4}$$

$$\bar{R} = R + \alpha(L_0 + M_0), \tag{3.5}$$

$$\bar{L}_\nu = L_\nu + 2\alpha\,(N_\nu + N_0), \tag{3.6}$$

$$\bar{M}_\nu = M_\nu + 2\alpha N_\nu. \tag{3.7}$$

The parameter α is now determined by the condition that the linear terms in (3.3) vanish, *i. e.*,

$$\bar{R} = 0, \tag{3.8}$$

This leads to

$$\alpha = -\frac{R}{L_0+M_0} = -\frac{\sqrt{2NS}\gamma_z(2S\gamma_x G(0)-h)}{2(h\gamma_x+2S\gamma_z^2 G(0)-2S\gamma_x^2 G(0))}. \tag{3.9}$$

The remaining quadratic part of (3.3) can be diagonalized by the "local" Bogolyubov transformation [3]

$$b_\nu = u_\nu \eta_\nu + v_\nu \eta_{-\nu}^+ \tag{3.10}$$

with real coefficients u_ν, v_ν. The operators η_ν, η_ν^+ are of Bose type if

$$u_\nu^2 - v_\nu^2 = 1. \tag{3.11}$$

A standard calculation gives for these coefficients the result

$$u_\nu^2 = {}^1/_2\,[(1-(\bar{M}_\nu/\bar{L}_\nu)^2)^{-1/2}+1],\ v_\nu^2 = {}^1/_2\,[(1-\bar{M}_\nu/\bar{L}_\nu)^2)^{-1/2}-1]. \tag{3.12}$$

The diagonalized Hamiltonian reads

$$H = E_0+\delta E_0+\Delta E_0+\sum_\nu \omega_\nu \eta_\nu^+ \eta_\nu \tag{3.13}$$

where

$$\omega_\nu = (\bar{L}_\nu^2-\bar{M}_\nu^2)^{1/2}, \tag{3.14}$$

$$\Delta E_0 = -\sum_\nu \omega_\nu v_\nu^2. \tag{3.15}$$

If the higher terms neglected in (3.3) are to give no large contributions to the ground state energy we must have

$$|v_\nu| \ll 1, \tag{3.16}$$

which corresponds to the (stronger) condition of the applicability of the approximate second-quantization method to spin-wave theory [3].

The expression $\tilde{E}_0 = E_0+\delta E_0+\Delta E_0$ represents the exact ground state energy of the bilinear Hamiltonian (3.3) and approximates reasonably the ground state energy of the original spin Hamiltonian if conditions (3.2) and (3.16) are satisfied.

The method *C* resides now in minimizing $\tilde{E}_0$ with respect to the angle φ, and the solutions have to meet the conditions (3.2) and (3.16). The difference between our approach and that in [5] consists in the different non-interacting-spin-waves approximation. In [5], this approximation has been carried through in the operators a_ν, a_ν^+ *i. e.*, before the transformation (3.1); here, we do it upon passing to the operators b_ν, b_ν^+ under certain restrictions.

For the sake of simplicity we confine ourselves now to the case

$$G(0) > 0, \quad h > 0. \tag{3.17}$$

It is instructive to start with the minimization of

$$E_0+\delta E_0 = NS\left[-h\sin\varphi - SJ(0) - SG(0)\cos^2\varphi - \tfrac{1}{2}\frac{\cos^2\varphi(2SG(0)\sin\varphi-h)}{h\sin\varphi+2SG(0)(\cos^2\varphi-\sin^2\varphi)}\right]. \tag{3.18}$$

The necessary condition for an extremum reads:

$$\frac{d}{d\varphi}(E_0+\delta E_0) = \frac{NS}{2}\,\frac{\cos^3\varphi(2SG(0)\sin\varphi-h)^2(h-8SG(0)\sin\varphi)}{[h\sin\varphi+2SG(0)(\cos^2\varphi-\sin^2\varphi)]^2} = 0. \tag{3.19}$$

From (3.19) and the examination of the higher derivatives we obtain the following extrema:

$$\sin\varphi = \pm 1 \qquad \text{minima} \tag{3.20}$$

$$\sin\varphi = h/2SG(0) \qquad \text{inflexion point} \tag{3.21}$$

$$\sin\varphi = h/8SG(0) \qquad \text{maximum} \tag{3.22}$$

where $\sin\varphi = 1$ is the lower minimum of (3.20).

A comparison with the results of methods A and B shows accordance between (3.20) and (2.21a, b). The minimum (2.21c) of method A, however, appears to correspond to the inflexion point (3.21). Moreover, we have the additional maximum (3.22).

The minimization of the total ground state energy $\tilde{E}_0$ of (3.13) cannot be carried through rigorously. It can be proved, however, that the solutions (2.21) are among those of the equation

$$\frac{d}{d\varphi}(E_0+\delta E_0+\Delta E_0) = 0. \tag{3.23}$$

To show this we note that

$$\frac{d}{d\varphi}\Delta E_0 = \cos\varphi\,\frac{d}{d\gamma_x}\Delta E_0 = \tfrac{1}{2}\cos\varphi\sum_\nu\left[\left(\bar{L}_\nu\frac{d\bar{L}_\nu}{d\gamma_x}-\bar{M}_\nu\frac{d\bar{M}_\nu}{d\gamma_x}\right)\omega_\nu^{-1}-\frac{d\bar{L}_\nu}{d\gamma_x}\right] \tag{3.24}$$

when utilizing Eqs (3.14) and (3.15). With (3.6) and (3.7) one easily verifies that

$$(d\bar{L}_\nu/d\gamma_x)_{\gamma_x=h/2SG(0)} = (d\bar{M}_\nu/d\gamma_x)_{\gamma_x=h/2SG(0)} = 0 \tag{3.25}$$

and, consequently,

$$(d\Delta E_0/d\varphi)_{\sin\varphi=\pm1} = (\mathrm{d}\Delta E_0/\mathrm{d}\varphi)_{\sin\varphi=h/2SG(0)} = 0. \tag{3.26}$$

From (3.26) and (3.24) follows (3.23).

One easily proves that the solutions (2.21) of Eq. (3.23) correspond for sufficiently strong and weak fields to the following extrema of $\tilde{E}_0$:

$$\left.\begin{aligned}&\sin\varphi = 1 \qquad \text{minimum}\\ &\sin\varphi = -1 \qquad \text{maximum}\end{aligned}\right\} \quad \text{for } h \gg 2SG(0), \tag{3.27, 3.28}$$

$$\sin\varphi = h/2SG(0) \qquad \text{minimum} \qquad \text{for } h \ll 2SG(0). \tag{3.29}$$

Moreover, for the solutions (3.27) and (3.29) we have to check the conditions (3.2) and (3.16). In either case, the first one is automatically satisfied for arbitrary field-strengths as $\alpha = 0$, and condition (3.16) is also fulfilled under the above field restrictions as

$$|v_\nu(\sin\varphi = \pm 1)|^2 \ll 1 \qquad \text{if } h \gg 2SG(0),$$

$$|v_\nu(\sin\varphi = h/2SG(0)|\ll 1 \qquad \text{if } h \ll 2SG(0).$$

We see that the method C leads for weak and strong fields to the same results as the methods A and B, when disregarding the fact that the equation (3.23) may yet have other physically reasonable solutions corresponding to minima of $\tilde{E}_0$ besides (3.27) and (3.29). These, however, cannot be obtained analytically.

4. *Application of Wallace's statistical perturbation method*

In this section we shall calculate the low-temperature, weak-field expansion of the spontaneous magnetization by means of Wallace's statistical perturbation method [6]. For this purpose we use the results obtained in the preceding section, where the spinwave reference state and the spin quantization direction in the weak-field limit has been shown to correspond to the solution

$$\gamma \equiv \gamma_x = \sin\varphi = h/2SG(0) \ll 1. \tag{4.1}$$

For simplicity we assume again $G(0) > 0$, $h > 0$.

We start with the Hamiltonian (2.6) and confine ourselves to terms up to the fourth order in the spin wave operators. The linear terms of (2.6) vanish as $\bar{R} = R = 0$, and the bilinear part can be diagonalized by the Bogolyubov-transformation (3.10) with $\bar{L}_\nu = L_\nu$ and $\bar{M}_\nu = M_\nu$. Note that due to (3.9) and (4.1) we have $\alpha = 0$. Furthermore, we perform the transformation (3.10) in the interaction terms and confine ourselves to terms up to the fourth order with respect to γ, taking into account the relations

$$v_\nu = \tfrac{1}{2}\left(\frac{M_\nu}{L_\nu}\right)^2 + 0(\gamma^6),$$

$$\frac{M_\nu}{L_\nu} \sim \gamma^2, \quad u_\nu^2 = 1 + v_\nu^2 \tag{4.2}$$

following from Eqs (2.13), (2.14), (3.11) and (3.12). Thus, we obtain

$$H = E_0 + \sum (\omega_\nu + Q_\nu)\eta_\nu^+ \eta_\nu + \sum_{\nu\mu\lambda\varkappa} \delta(\nu+\mu-\lambda-\varkappa)\, P_{\nu\mu\lambda\varkappa}\eta_\nu^+ \eta_\mu^+ \eta_\lambda \eta_\varkappa +$$

$$+ \sum_{\nu\mu\lambda\varkappa} \delta(\nu-\mu-\lambda-\varkappa) T_{\nu\mu\varkappa}\eta_\nu^+ \eta_\mu \eta_\lambda \eta_\varkappa + \text{h. c.} \tag{4.3}$$

with the notation

$$Q_\nu = \sum_\alpha (F^0_{\nu\nu}v_\alpha^2 + 2F^0_{\nu\alpha}v_\alpha^2 + 4F^0_{\nu\alpha}v_\nu v_\alpha + F^0_{\alpha\alpha}v_\alpha^2 +$$

$$+ k_\nu v_\alpha + 2k_\alpha v_\nu) \sim \gamma^4 \tag{4.4}$$

$$P_{\nu\mu\lambda\varkappa} = F_{\nu\varkappa} + F^0_{\nu\varkappa}v_\nu^2 + F_{\nu\varkappa}v_\mu^2 + F^0_{\nu\varkappa}v_\lambda^2 + F^0_{\nu\varkappa}v_\varkappa^2 +$$

$$+ F^0_{\nu\varkappa}v_\mu v_\lambda + F^0_{\nu\lambda}v_\lambda^2 + F^0_{\mu\varkappa}v_\nu v_\lambda + F^0_{\mu\lambda}v_\lambda^2 +$$

$$+ 3k_\varkappa v_\mu + 3k_\mu v_\varkappa \tag{4.5}$$

$$T_{\nu\mu\varkappa} = F^0_{\nu\varkappa}v_\mu + F^0_{-\mu\varkappa}v_\mu + k_\varkappa \tag{4.6}$$

where $F^0_{\nu\mu}$ denotes the γ-independent part of $F_{\nu\mu}$ defined by Eq. (2.16). In (4.3) we neglected terms proportional to $\eta^+\eta\eta$, $\eta^+\eta^+\eta$, $\eta\eta$, $\eta^+\eta^+$, because they either play no role in the following perturbation method or contribute to the magnetization in the order γ^6.

Before applying Wallace's statistical perturbation method to the Hamiltonian (4.3) we shall briefly outline the method itself. Let H be the Hamiltonian constructed of the basic operators Θ_i, Θ_i^+ that obey the Hamiltonian commutator equations

$$[H, \Theta_i^+]_- = \omega_i \Theta_i^+ + R_i^+. \tag{4.7}$$

The operators Θ_i, Θ_i^+ and the (real) c-numbers ω_i are to be chosen in such a way that the remainder operator R_i^+ be small, in the sense that statistical averages involving R_i^+ are to be small. Using (4.8), a formula for the average $\langle \Theta_i^+ \Theta_i \rangle$ has been derived in [6], which is correct to the first order in the operators R_i^+:

$$\langle \Theta_i^+ \Theta_i \rangle_1 = \Phi_i^{\pm} \langle [\Theta_i, \Theta_i^+]_{\pm} \rangle_0 \pm \beta(\Phi_i^{\pm} \pm 1) \langle R_i^+ \Theta_i \rangle_0 \tag{4.8}$$

where

$$\Phi_i^{\pm} = (e^{\beta\omega_i} \pm 1)^{-1}, \quad \beta = \frac{1}{kT}. \tag{4.9}$$

Here, $\langle\ \rangle_0$ means the zeroth-order average ($R_i^+ = 0$) and $[\ ]_{\pm}$ denotes respectively the anticommutator or commutator. In generalizing [6], an expression for the average $\langle \Theta_i^+ \Theta_{-1}^+ \rangle$ has been derived in [12]:

$$\langle \Theta_i^+ \Theta_{-i}^+ \rangle_1 = \Phi_i^{\pm} \langle [\Theta_{-i}^+, \Theta_i^+]_{\pm} \rangle_0 \pm \frac{1}{2\omega_i} (\Phi_i^{\mp})^{-1} \langle \Theta_{-i}^+ R_i^+ \rangle_0. \tag{4.10}$$

The operators Θ_i, Θ_i^+ need not necessarily be of Bose or Fermi type. The smallness of R_i^+ in the above sense is to be checked *a posteriori*. We prefer to apply Wallace's method to our problem because, firstly, it is easy to work with and, secondly, for isotropic ferromagnets it gives the correct low-temperature expansion for the magnetization obtained by Dyson [7].

For our case we specify in Eqs (4.7), (4.8) and (4.10) $\Theta_i = \eta_\alpha$ and choose in (4.8), (4.10) the commutator. For the Hamiltonian (4.3) the commutator equations (4.7) read

$$[H, \eta_\alpha^+]_- = (\omega_\alpha + Q_\alpha) \eta_\alpha^+ + R_\alpha^+ \tag{4.11}$$

where

$$R_\alpha^+ = \sum_{\nu\mu\lambda} \delta(\nu + \mu - \lambda - \alpha) (P_{\nu\mu\lambda\alpha} + R_{\nu\mu\alpha\lambda}) \eta_\nu^+ \eta_\mu^+ \eta_\lambda + \tag{4.12}$$

$$+ \sum_{\nu\mu\lambda} \delta(\nu - \mu - \lambda - \alpha) (T_{\nu\mu\alpha} + T_{\nu\mu\lambda} + T_{\nu\alpha\mu})\ \eta_\nu^+ \eta_\mu \eta_\lambda.$$

Since

$$\langle \eta_\nu^+ \eta_\mu^+ \eta_\lambda \eta_\alpha \rangle_0 = \Phi_\nu \Phi_\mu (\delta_{\nu\lambda} \delta_{\mu\alpha} + \delta_{\nu\alpha} \delta_{\mu\lambda}) \tag{4.13}$$

one obtains

$$\langle R_\alpha^+ \eta_\alpha \rangle_0 = \Phi_\alpha \sum_\nu \Phi_\nu S_{\nu\alpha}, \tag{4.14}$$

$$\langle \eta_{-\alpha}^+ R_\alpha^+ \rangle_0 = \Phi_\alpha \sum_\nu \Phi_\nu W_{\nu\alpha} \tag{4.15}$$

where

$$S_{\nu\alpha} = P_{\nu\alpha\nu\alpha} + P_{\nu\alpha\alpha\nu} + P_{\alpha\nu\nu\alpha} + P_{\alpha\nu\alpha\nu}, \tag{4.16}$$

$$W_{\nu\alpha} = T_{\nu,-\alpha,\alpha} + T_{\nu,-\alpha,\nu} + T_{\nu,\alpha,-\alpha} + T_{\nu,\nu,\alpha} + T_{\nu,\nu,-\alpha} + T_{\nu,\alpha,\nu} \tag{4.17}$$

and Φ_ν denotes the Bose distribution

$$\Phi_\nu = [e^{\beta(\omega_\nu + Q_\nu)} - 1]^{-1}. \tag{4.18}$$

Thus, with (4.8), (4.10) we arrive at the following averages in the first-order statistical perturbation approach:

$$\langle \eta_\alpha^+ \eta_\alpha \rangle_1 = \Phi_\alpha - \beta(\Phi_\alpha + 1)\, \Phi_\alpha \sum_\nu \Phi_\nu S_{\nu\alpha}, \tag{4.19}$$

$$\langle \eta_\alpha^+ \eta_{-\alpha}^+ \rangle_1 = -\frac{1}{2(\omega_\alpha + Q_\alpha)} (2\Phi_\alpha + 1) \sum_\nu \Phi_\nu W_{\nu\alpha}. \tag{4.20}$$

The spontaneous magnetization in the quantization direction given by the angle φ can be written as

$$\sigma = 1 - \frac{1}{NS} \sum_\alpha \langle a_\alpha^+ a_\alpha \rangle = 1 - \frac{1}{NS} \sum_\alpha [v_\alpha^2 + (u_\alpha^2 + v_\alpha^2) \langle \eta_\alpha^+ \eta_\alpha \rangle + 2u_\alpha v_\alpha \langle \eta_\alpha^+ \eta_{-\alpha}^+ \rangle]. \tag{4.21}$$

(Note that in our case $\langle \eta_\alpha^+ \eta_{-\alpha}^+ \rangle = \langle \eta_\alpha \eta_{-\alpha} \rangle$.) By virtue of (4.19), (4.20), (4.21) and taking into account (4.2) one finally obtains the following formula for the magnetization correct to the order γ^4:

$$\sigma = 1 - \frac{1}{NS} \sum_\alpha v_\alpha^2 - \frac{1}{NS} \Big[\sum_\alpha \Phi_\alpha + \sum_\alpha v_\alpha^2 \Phi_\alpha - \beta \sum_{\alpha,\nu} (\Phi_\alpha + 1) \Phi_\alpha \Phi_\nu S_{\nu\alpha} - 2\beta \sum_{\alpha,\nu} v_\alpha^2 (\Phi_\alpha + 1) \Phi_\alpha \Phi_\nu S_{\nu\alpha}^0 - \sum_{\alpha,\nu} \frac{v_\alpha}{\omega_\alpha} (2\Phi_\alpha + 1) \Phi_\nu W_{\nu\alpha} \Big] \tag{4.22}$$

where

$$S_{\nu\alpha}^0 = 2F_{\nu\alpha}^0 + F_{\nu\nu}^0 + F_{\alpha\alpha}^0 \tag{4.23}$$

denotes the γ-independent part of $S_{\nu\alpha}$. In the denominator of the last term of (4.22) we have omitted the quantity Q_α because $Q_\alpha \sim \gamma^4$ and, therefore, $v_\alpha(\omega_\alpha + Q_\alpha)^{-1} = v_\alpha \omega_\alpha^{-1} + O(\gamma^6)$.

In the following we shall explicitly evaluate Eq. (4.19) up to the fourth order in the temperature T but only to the second order in the reduced field γ. The latter restriction simplifies markedly the calculations. Accordingly, Eq. (4.22) turns into

$$\sigma = 1 - \frac{1}{NS} \Big[\sum_\alpha \Phi_\alpha - \beta \sum_{\alpha,\nu} (\Phi_\alpha + 1)\, \Phi_\alpha \Phi_\nu S_{\nu\alpha} \Big]. \tag{4.24}$$

Further, the quantity Q_α in (4.18) can be dropped. With (2.13) the spin-wave energy spectrum reads

$$\omega_\alpha = \sqrt{L_\alpha^2 - M_\alpha^2} = L_\alpha(1 - O(\gamma^4)) \approx L_\alpha. \tag{4.25}$$

In order to analyze the formula (4.24) we expand at first the function (4.18) into a series and pass from sums to integrals:

$$\Phi_\alpha = \sum_{h=1}^{\infty} e^{-n\beta\omega_\alpha}, \quad (\Phi+1)\Phi_\alpha = \sum_{h=1}^{\infty} n e^{-n\beta\omega_\alpha}, \tag{4.26}$$

$$\frac{1}{N}\sum_{\alpha} \to \frac{V}{N}\cdot\frac{1}{(2\pi)^3}\int d^3\alpha. \tag{4.27}$$

For simplicity, we assume a simple cubic lattice and use the nearest-neighbour approximation. The calculations can be carried through as well for a tetragonal lattice, but are in this case considerably lengthier. With these assumptions Eqs (2.18a) and (2.18b) become

$$J(\nu) = 2I(\cos a\nu_x + \cos a\nu_y + \cos a\nu_z), \tag{4.28a}$$

$$G(\nu) = 2G(\cos a\nu_x + \cos a\nu_y + \cos a\nu_z) \tag{4.28b}$$

where a denotes the lattice constant, and I and G are the isotropic and anisotropic nearest-neighbour exchange integrals.

To obtain the correct low-temperature expansion of σ up to the order T^4 within the long-wavelength approximation, the cosine functions in (4.28a), (4.28b) are to be expanded to the sixth order:

$$\cos x \approx 1 - \frac{x^2}{2!} + \frac{x^4}{4!} - \frac{x^6}{6!}. \tag{4.29}$$

The spin wave energy ω_α can now be written as

$$\omega_\alpha = \omega^{(0)} + \omega_\alpha^{(2)} + \omega_\alpha^{(4)} + \omega_\alpha^{(6)} \tag{4.30}$$

where $\omega^{(n)}$ involves products of n wavevector components $\alpha_x, \alpha_y, \alpha_z$. With (4.30) we rewrite and expand the exponential functions in (4.26) as follows:

$$e^{-n\beta\omega_\alpha} = e^{-n\beta\omega_0} e^{-n\beta\omega_\alpha^{(2)}} e^{-n\beta(\omega_\alpha^{(4)}+\omega_\alpha^{(6)})}$$

$$= e^{-n\beta\omega_0} e^{-n\beta\omega_\alpha^{(2)}} \left[1 - n\beta(\omega_\alpha^{(4)} + \omega_\alpha^{(6)}) + \frac{n^2\beta^2}{2!}(\omega_\alpha^{(4)})^2 + \ldots\right]. \tag{4.31}$$

The above expansion is again necessary to get all temperature powers up to T^4. The insertion of (4.31) and (4.26) in (4.24) and the use of (4.27) leads to integrals of the type

$$I = \int_{-\infty}^{\infty}\int\int e^{-A[(a\alpha_x)^2+(a\alpha_y)^2+(a\alpha_z)^2]}(a\alpha_x)^p(a\alpha_y)^q(a\alpha_z)^r d^3\alpha, \tag{4.32}$$

where p, q, r are even integers. Therein, the integration has been extended beyond the first Brillouin zone over the whole wavevector space (see, *e. g.*, [2] for justification). The integral (4.32) can easily be calculated:

$$I = \frac{1}{a^3} A^{-\frac{1}{2}(3+p+q+r)} \Gamma\left(\frac{p+1}{2}\right)\Gamma\left(\frac{q+1}{2}\right)\Gamma\left(\frac{r+1}{2}\right) \tag{4.33}$$

where $\Gamma(x)$ denotes the gamma-function. After somewhat tedious but straightforward calculations we finally obtain

$$\sigma = 1 - S^{-1}[P_{3/2} Z_{3/2}(d/\tau)\tau^{3/2} + P_{5/2} Z_{5/2}(d/\tau)\tau^{5/2} + P_{7/2} Z_{7/2}(d/\tau)\tau^{7/2} + Q_2 Z_{3/2}(d/\tau) Z_{5/2}(d/\tau)\tau^2 + Q_3(Z_{3/2}(d/\tau) Z_{7/2}(d/\tau) + [Z_{5/2}(d/\tau)]^2)\tau^3 + (Q_4 Z_{5/2}(d/\tau) Z_{7/2}(d/\tau) + Q_4' Z_{3/2}(d/\tau) Z_{9/2}(d/\tau))\tau^4] \tag{4.34}$$

with the dimensionless quantities

$$\tau = \frac{kT}{8\pi SI}, \tag{4.35}$$

$$P_{3/2} = 1 - \frac{3}{4} g\gamma^2, \quad P_{5/2} = \frac{3\pi}{4}\left(1 - \frac{5}{4} g\gamma^2\right), \quad P_{7/2} = \frac{33}{32}\pi^2\left(1 - \frac{7}{4} g\gamma^2\right)$$

$$Q_2 = \frac{4g}{\pi S}\left(1 - \frac{3}{2}\gamma^2 - \frac{3}{2} g\gamma^2\right), \quad Q_3 = \frac{3}{4S} g\left(1 - \frac{3}{8}\gamma^2 - 2g\gamma^2\right)$$

$$Q_4 = \frac{3\pi}{2S}\left(1 + \frac{g}{8}\left[1 - \frac{35}{2}\gamma^2 - \frac{5}{2} g\gamma^2\right]\right)$$

$$Q_4 = -\frac{21\pi}{8S} g\left(1 - \frac{3}{2}\gamma^2 - \frac{5}{2} g\gamma^2\right), \quad d = \frac{3}{2\pi} g\left(1 - \frac{1}{2}\gamma^2\right) \tag{4.36}$$

$$g = G/I, \gamma = h/12SG, \tag{4.37}$$

$$Z_p(x) = \sum_{n=1}^{\infty} n^{-p} e^{-nx}. \tag{4.38}$$

Let us briefly discuss the magnetization formula (4.34). To begin with, we consider $h = 0$, $G = 0$ (isotropic field-free ferromagnet). In this case we obtain Dyson's [7] low-temperature expansion to the order T^4, apart from a slight discrepancy in the coefficient of the T^4 term due to the different approximation methods. Note that the spurious T^3-term of the first order Green's function theories [3,13] does not appear in (4.34). For $G > 0$ and $h = 0$ additional contributions proportional to T^2 and T^3 emerge. The presence of a weak field perpendicular to the anisotropy axis does not change the powers of T but merely changes the coefficients according to (4.36).

5. *Concluding remarks*

The method C, as employed in Section 3 to the uniaxial ferromagnet with transversal field described by the approximate spin-wave Hamiltonian (3.3), was shown to lead for sufficiently weak and strong fields to the same result as the methods A and B. We may therefore conclude that the methods A and B — which are by far simpler — are considerably more effective and sufficiently adequate in determining the suitable spin-deviation or spin-wave reference state which reasonably approximates the spin system's ground state, provided the class of trial states is properly chosen.

It is to be noted that the field-restrictions in Eqs (3.27–3.29) actually result from the Bogolyubov transformation (3.10) and the condition (3.16). When using the method A or B, they also turn up (though in a weaker form) upon applying the method of approximate second quantization in determining the spin-wave energy spectrum [14, 15], without restricting in any way the reference state itself. It may also be noted that there have been suggested other methods of determining the spin-wave reference state which are yet to be explored [1, 16].

Finally, it should be emphasized that the low-temperature weak-field expansion of the magnetization (4.34), obtained on the basis of the reference state corresponding to (4.1) by means of Wallace's statistical perturbation method, presents a generalization of Dyson's results [7]. It can easily be seen that the perturbation term in Eq. (4.24) is much smaller than the unperturbed term if $\tau \ll 1$, $h \ll h_c \equiv 2SG(0)$, $g = G/I \ll 1$, which constitutes in our case the criterion for the applicability of the statistical perturbation method.

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