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SPIN-WAVE THEORY OF UNIAXIAL TETRAGONAL FERROMAGNETS WITH EXTERNAL MAGNETIC FIELD AND PSEUDO-DIPOLAR SPIN COUPLING. I. FREE-PARTICLE APPROXIMATION

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The spin-wave theory in the non-interacting-spin-waves approximation is applied to uniaxial ferromagnets with simple and body-centred tetragonal crystal lattices and pseudo-dipolar spin coupling. The influence of a homogeneous external magnetic field on the spin-wave energy spectrum and thermodynamic quantities (magnetization, magnetic susceptibility, specific heat) is studied. For the perpendicular-field case, the validity of the Tayblikov-Siklós (1959) long-wavelength approximation is reexamined and compared with a new approximation procedure which is shown to widen the restriction interval for the external-field-strength in the ferro- and paramagnetic phases. In Part II, the theory is extended to include spin-wave interactions in the lowest approximation, which leads to a temperature-dependent quantization direction and energy spectrum.

1. *Introduction*

The influence of the homogeneous external magnetic field on the thermodynamic behaviour of ferromagnets with uniaxial anisotropy was studied in several papers [1–18], using molecular field [1–7] and spin-wave theories [8–11], as well as Green's functions methods [12–17]. With a few exceptions, it was mainly the case of the longitudinal field (*i. e.*, parallel to the anisotropy axis) which was considered in those papers. Except for the papers [13–14, 18], only very recently became the case of the transversal field (*i. e.*, perpendicular to the anisotropy axis) again subject of extensive theoretical investigations [3–7, 17, 19], owing to the field-induced second-order phase transition that can occur in this case.

The apparent deficiency of the paper [18] where a microscopic approach (spin Hamiltonian) has been used, is the inconsistency between the uniaxial anisotropy and the cubic lattice symmetry for which the numerical results are presented. The aim of our paper is, therefore, to apply the spin-wave theory to a consistently uniaxial ferromagnet, and to examine the influence of an external field on its thermodynamical properties. Anisotropic

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interactions are purposely taken into account in the form of pseudo-dipolar coupling, as this form of anisotropy enables one automatically to distinguish uniaxial ferromagnets according to the crystal lattice and the degree of its deformation, and permits one to consider the anisotropic interactions in the approximation of successive coordination spheres [20]. In the first part of this paper the role of different approximations in the non-interacting-spin-waves energy spectrum, as well as their influence on thermodynamical quantities, are examined. In Part II we shall investigate the influence of the spin-wave interactions in the lowest approximation of the Holstein-Primakoff mapping showing, among others, that they lead to a temperature-dependent magnetization direction.

2. *The spin-deviation reference state*

The system of spins in consideration is described by the Hamiltonian

$$H = H_{EX} + H_Z + H_D. \tag{2.1}$$

The exchange energy is given by

$$H_{EX} = -\sum_{rr'} A_{rr'} S_r^\eta S_{r'}^\eta \tag{2.2}$$

$$A_{rr'} = A(|\boldsymbol{r} - \boldsymbol{r}'|)\,[1 - \delta_{rr'}] \geqslant 0$$

where the lower indices $r \equiv \boldsymbol{r}$, $r' \equiv \boldsymbol{r}'$ are lattice vectors, $A_{rr'}$ — the exchange integral between atoms at the sites $\boldsymbol{r}$ and $\boldsymbol{r}'$, and S_r^η denotes the components ($\eta = x, y, z$) of the spin vector assigned to the site $\boldsymbol{r}$. (To the vector indices $\eta, \nu, \ldots$ Einstein's summation rule is applied.) The energy of the system in a uniform external magnetic field $\vec{\mathscr{H}}$ perpendicular to the $0y$-axis has the form

$$H_Z = B \sum_r (S_r^x \sin\theta + S_r^z \cos\theta) \tag{2.3}$$

$$B = -\mu \mathscr{H} > 0$$

where θ is the angle between the $0z$-axis and the direction of the external field. The anisotropy energy is taken in the form proposed by Van Vleck [21]

$$H_D = \sum_{rr'} D_{rr'} \left[S_r^\eta S_{r'}^\eta - \frac{3(\varrho_{rr'}^\eta S_r^\eta)(\varrho_{rr'}^\nu S_{r'}^\nu)}{\varrho_{rr'}^2} \right] \tag{2.4}$$

where $\varrho_{rr'}^\eta$ are the components of the lattice vector between the sites $\boldsymbol{r}$ and $\boldsymbol{r}'$, and $D(|\boldsymbol{r} - \boldsymbol{r}'|) \geqslant 0$ is the pseudo-dipolar coupling function which at small distances can be approximated by the exchange integral as follows [22]:

$$|D(l)| \approx |A(l)|(g-2)^2. \tag{2.5}$$

Here, l is the nearest-neighbour distance and g is Lande's factor.

Let εa and a denote the lattice constants in the tetragonal direction (coinciding with the $0z$-axis) and tetragonal plane $x0y$, respectively. As was shown in [20] for the field-free case, by minimizing the energy of the spin system $H_{EX}+H_D$ in the saturation state and in the nearest-neighbour approximation, we get for tetragonal crystals the magnetically preferred directions presented in Table I.

TABLE I

Type of tetragonal lattice	Conditions for ε	Magnetically preferred directions
simple (s.t.)	$\frac{1}{2}<\varepsilon<1$	tetragonal axis
	$1<\varepsilon<\sqrt{2}$	tetragonal plane
body centred (b.c.t.)		tetragonal axis
	$\sqrt{\frac{2}{3}}<\varepsilon<1$	tetragonal plane

Let us denote by $|-S\rangle$ the ground state of the Hamiltonian H_{EX} in which the spins at all lattice sites have the lowest eigenvalue $-S$ in the same (but otherwise arbitrary) direction. In the representation where the components S_r^z are diagonal this state corresponds to total magnetic saturation and represents the spin deviation vacuum state $|0\rangle$ defined for all $\boldsymbol{r}$ in the following way:

$$S_r^z|0\rangle = -S|0\rangle, \qquad S_r^-|0\rangle = 0 \tag{2.6}$$

where $S_r^{\pm} = S_r^x \pm iS_r^y$.

Applying the operators S_r^+ to the vacuum state we get the complete and orthonormal set of eigenstates $|u\rangle$ of the operators S_r^z,

$$|u\rangle \equiv |\ldots u_r \ldots\rangle = \left\{\prod_r \left[\frac{(2S-u_r)!}{(2S)!(u_r)!}\right]^{1/2} (S_r^+)^{u_r}\right\}|0\rangle \tag{2.7a}$$

$$S_r^z|u\rangle = (u_r - S)|u\rangle \tag{2.7b}$$

$$u_r = 0, 1, 2, \ldots, 2S.$$

In the case of the isotropic ferromagnet, the choice of $|-S\rangle$ as the reference state (in the sense of [23]) is completely justified. However, the proper choice of the spin-wave reference state (approximate ground state) for a uniaxial ferromagnet in an external magnetic field is quite another problem if the spin-wave interactions are to remain sufficiently small to justify the standard long-wavelength low-temperature approximation. Different methods of determining the optimum quantization axis in the class of the so-called homogeneous reference states for a general ferromagnetic Hamiltonian have been analyzed in [24]. To find the homogeneous reference state for the Hamiltonian (2.1), the variational method (method A in [24]) will be employed. The method resides in minimizing the mean value of the Hamiltonian in the class of states $|0(\vartheta)\rangle$, generated from $|0\rangle$ by means of the unitary transformation $U(\vartheta)$, with respect to the uniform spin orientation given by the angle ϑ. (Due to the neglection of higher than dipolar interactions we have in the field-free case cylindrical symmetry with respect to the tetragonal axis. This permits us to restrict in our case the number of minimization parameters to a single one, ϑ, which is the angle between the quantization direction and the tetragonal axis.) This method is equivalent (in a limited sense) to eliminating from the spin Hamiltonian the terms which are linear with respect to the spin-deviation creation and destruction operators [24–26]. The transformation $U = U(\vartheta)$ converts the set of vectors $|u\rangle$ into the orthonormal set

$$|u(\vartheta)\rangle = U^{+}|u\rangle. \tag{2.8}$$

It is, however, convenient to use the representation (2.7a). Therefore, we transform the Hamiltonian (2.1) of our system as follows:

$$\tilde{H} = UHU^{+} \tag{2.9}$$

where [27]

$$US_r^{\eta}U^{+} = R_{\eta\nu}S_r^{\nu} = \tilde{S}_r^{\eta}. \tag{2.10}$$

In our case

$$R_{\eta\nu} = \begin{pmatrix} \cos\vartheta & 0 & \sin\vartheta \\ 0 & 1 & 0 \\ -\sin\vartheta & 0 & \cos\vartheta \end{pmatrix}. \tag{2.11}$$

The quantity to be minimized with respect to ϑ is then

$$E_0 \equiv \langle 0|\tilde{H}|\alpha\rangle. \tag{2.12}$$

Introducing the direction cosines α, β, γ of the lattice vector between the sites $\boldsymbol{r}$ and $\boldsymbol{r}'$

$$\boldsymbol{\varrho}_{rr'}\varrho_{rr'}^{-1} = (\alpha_{rr'}, \beta_{rr'}, \gamma_{rr'})$$

we get the following expression for E_0:

$$E_0 = -S^2\sum_{rr'} A_{rr'} + S^2\sum_{rr'} D_{rr'}(1-3\gamma_{rr'}^2) +$$
$$+3S^2\sum_{rr'} D_{rr'}\left[(\gamma_{rr'}^2-\alpha_{rr'}^2)\sin^2\vartheta - \alpha_{rr'}\gamma_{rr'}\sin 2\vartheta\right] - NSB\cos(\vartheta-\theta) \tag{2.13}$$

where N is the number of lattice sites in the crystal.

The minimum conditions for E_0 read

$$\frac{dE_0}{d\vartheta} = 0, \quad \frac{d^2E_0}{d\vartheta^2} > 0. \tag{2.14}$$

In the following, three cases will be considered, namely, when the external magnetic field forms the angles $\theta = 0, \pi/2, \pi/4$ with the $0z$ (tetragonal) axis of the coordinate system.

The solutions corresponding in these cases to a minimum of E_0 are presented in Table II. In thermodynamical calculations we shall restrict ourselves to the case when the tetragonal axis (0z-axis) is magnetically preferred.

TABLE II

External field direction	Solution	Minimum conditions for s.t.		Minimum conditions for b.c.t.	
		$^1/_2 < \varepsilon < 1$	$1 < \varepsilon < \sqrt{2}$	$\sqrt{^2/_3} < \varepsilon < 1$	$1 < \varepsilon < \sqrt{2}$
$\theta = 0$	$\sin\vartheta = 0$	B-arbitrary	$B \geqslant -B_s^a$	$B \geqslant -B_b^a$	B-arbitrary
	$\cos\vartheta = -B/B_j^a$	Not fulfilled	$B \leqslant -B_s^a$	$B \leqslant -B_b^a$	Not fulfilled
$\theta = \frac{\pi}{2}$	$\sin\vartheta = B/B_j^a$	$B \leqslant B_s^a$	Not fulfilled		$B \leqslant B_b^a$
	$\cos\vartheta = 0$	$B \geqslant B_s^a$	B-arbitrary		$B \geqslant B_b^a$
$\theta = \frac{\pi}{4}$	$\vartheta = \pi/4 + \arcsin\varphi^-$	B-arbitrary	Not fulfilled		B-arbitrary
	$\vartheta = 5\pi/4 - \arcsin\varphi^-$	Not fullfiled			
	$\vartheta = \pi/4 + \arcsin\varphi^+$	Not fulfilled	B-arbitrary		Not fulfilled
	$\vartheta = 5\pi/4 - \arcsin\varphi^+$	Not fulfilled			

$$B_j^a = \begin{cases} B_b^a = 12SDs(\lambda - 1) & \text{for s.t.} \quad D_s \equiv D_s(a) \\ B_b^a = 48SD_b \dfrac{\varepsilon^2 - 1}{\varepsilon^2 + 2} & \text{for b.c.t.} \quad D_b \equiv D_b\left(\dfrac{a}{2}\sqrt{\varepsilon^2+2}\right) \end{cases}$$

$$\lambda = \frac{D_s(\varepsilon a)}{D_s(a)} \qquad \varphi^{\pm} = \frac{1}{2}\left[\frac{B}{B_j^a} \pm \sqrt{\left(\frac{B}{B_j^a}\right)^2 + 2}\right]$$

3. *The spin wave energy spectrum*

Let us pass in the Hamiltonian (2.9) to Bose operators, using the Holstein-Primakoff mapping [28] in the lowest approximation:

$$S_r^+ = (S_r^-)^+ \to \sqrt{2S}\, a_r^+$$

$$S_r^z \to a_r^+ a_r - \delta$$

$$[a_r, a_{r'}^+] = \delta_{rr'} \qquad [a_r, a_{r'}] = 0 \tag{3.1}$$

Then we get

$$\tilde{H} = E_0 + \tilde{H}_1 + \tilde{H}_2 + \dots \tag{3.2}$$

$$\tilde{H}_1 = \sum_{rr'} (F_{rr'} a_r + F_{rr'}^* a_r^+) \tag{3.3a}$$

$$F_{rr'} = -3S(S/2)^{1/2}D_{rr'}[(\gamma^2_{rr'}-\alpha^2_{rr'})\sin 2\vartheta -$$
$$-2i\beta_{rr'}(\alpha_{rr'}\sin\vartheta+\gamma_{rr'}\cos\vartheta)-2\alpha_{rr'}\gamma_{rr'}\cos 2\vartheta]+$$
$$+(S/2)^{1/2}B\delta_{rr'}\sin(\theta-\vartheta) \tag{3.3b}$$

$$\tilde{H}_2 = \sum_{rr'} N_{rr'}a^+_r a_{r'} + \tfrac{1}{2}\sum_{rr'}(K_{rr'}a_r a_{r'} + K^*_{rr'}a^+_r a^+_{r'}) \tag{3.4a}$$

$$N_{rr'} = 3SD_{rr'}[-\alpha^2_{rr'}-\beta^2_{rr'}+(\alpha^2_{rr'}-\gamma^2_{rr'})\sin^2\vartheta+$$
$$+\alpha_{rr'}\gamma_{rr'}\sin 2\vartheta]+2S(D_{rr'}-A_{rr'})+\delta_{rr'}[B\cos(\vartheta-\theta)-$$
$$-2S\sum_{r''}M_{rr''}] \tag{3.4b}$$

$$M_{rr'} = -A_{rr'}+D_{rr'}(1-3\gamma^2_{rr'})+3D_{rr'}[(\gamma^2_{tr'}-\alpha^2_{rr'})\sin^2\vartheta-$$
$$-\alpha_{rr'}\gamma_{rr'}\sin 2\vartheta] \tag{3.4c}$$

$$K_{rr'} = 3SD_{rr'}[\beta^2_{rr'}-\alpha^2_{rr'}+(\alpha^2_{rr'}-\gamma^2_{rr'})\sin^2\vartheta+$$
$$+2i\beta_{rr'}\beta(\alpha_{rr'}\cos\vartheta-\gamma_{rr'}\sin\vartheta)+\alpha_{rr'}\gamma_{rr'}\sin 2\vartheta). \tag{3.4d}$$

It is easy to verify that the solutions of the equation $\sum_{r'} F_{rr'} = 0$ (which ensures the vanishing of the linear part of the Hamiltonian (3.2), *i. e.*, $H_1 = 0$) satisfy the necessary condition for the minimum of E_0.

Taking the Fourier transforms of the operators a_r, a^+_r in $\tilde{H}_2$,

$$a_r = \frac{1}{\sqrt{N}}\sum_k b_k e^{ikr}, \qquad a^+_r = \frac{1}{\sqrt{N}}\sum_k b^+_k e^{-ikr} \tag{3.5}$$

(where $k \equiv \boldsymbol{k}$ are reciprocal lattice vectors and $kr \equiv \boldsymbol{kr}$ scalar products) we get

$$\tilde{H}_2 = \sum_k [N_k b^+_k b_k + \tfrac{1}{2}(K_k b_k b_{-k} + K^*_k b^+_k b^+_{-k})] \tag{3.6}$$

where

$$N_k = \sum_r N_{rr'} e^{i(r-r')k}, \quad K_k = \sum_r K_{rr'} e^{i(r-r')k}. \tag{3.7}$$

Using the Bogolyubov transformation [12]

$$b_k = u_k c_k + v^*_k c^+_{-k}$$
$$b^+_k = u^*_k c^+_k + v_k c_{-k} \tag{3.8}$$

we obtain $\tilde{H}_2$ in diagonal form,

$$\tilde{H}_2 = \Delta E_0 + \sum_k E_k c^+_k c_k \tag{3.9}$$

where

$$E_k = (N^2_k - |K_k|^2)^{1/2} \tag{3.9a}$$

is the energy a magnon with the wave vector $\boldsymbol{k}$, and

$$\Delta E_0 = -\tfrac{1}{2}\sum_k (N_k - E_k). \tag{3.10}$$

The coefficients u_k, v_k of the transformation (3.8) have the form [10]

$$u_k = \sqrt{\frac{N_k+E_k}{2E_k}}, \quad v_k = -\frac{K_k}{|K_k|}\sqrt{\frac{N_k-E_k}{2E_k}}. \tag{3.11}$$

For weakly excited states (low-temperature approximation) the terms of higher than second order in the operators a_r, a_r^+ (interactions) can be neglected in the Hamiltonian (3.2). When applying the above approximate second-quantization method, the following inequality must hold [12]:

$$\sum_k |v_k|^2 \ll 1. \tag{3.12}$$

In the low-temperature region the major contribution to thermodynamical quantities comes from the spin waves with small wave vectors. Therefore, N_k and K_k can be approximated as follows:

(*i*) for the simple tetragonal lattice:

$$N_k \approx B\cos(\vartheta-\theta) + \tfrac{1}{2}B_s^a(2-3\sin^2\vartheta) + Sa^2(2A_s+D_s)(k_x^2+ \\ + k_y^2 + \varepsilon^2\lambda k_z^2) - 3S\varepsilon^2 a^2\lambda D_s \left[k_z^2 - \left(k_z^2 - \frac{k_x^2}{\varepsilon^2\lambda}\right)\sin^2\vartheta\right] \tag{3.13}$$

$$K_k \approx 6SD_s\left\{\frac{a^2}{2}(k_x^2-k_y^2) + \left[1-\lambda+\frac{a^2}{2}(\lambda\varepsilon^2 k_z^2 - k_x^2)\right]\sin^2\vartheta\right\}; \tag{3.14}$$

(*ii*) for the body centred tetragonal lattice:

$$N_k \approx B\cos(\vartheta-\theta) + \tfrac{1}{2}B_b^a(2-3\sin^2\vartheta) + Sa^2 \times \\ \times \left\{2(A_b-D_b) - \frac{3D_b}{\varepsilon^2+2}[(1-\varepsilon^2)\sin^2\vartheta - 2]\right\}(k_x^2+k_y^2+\varepsilon^2 k_z^2) - \frac{6a^2\varepsilon SD_b}{\varepsilon^2+2}k_x k_z \sin 2\vartheta \tag{3.15}$$

$$K_k \approx \frac{24SD_b}{\varepsilon^2+2}\left\{(1-\varepsilon^2)\left[1-\frac{a^2}{8}(k_x^2+k_y^2+\varepsilon^2 k_z^2)\right]\sin^2\vartheta - \\ - \frac{1}{4}a^2\varepsilon^2 k_x k_z \sin 2\vartheta + i\frac{a^2 k_y}{2}(\varepsilon^2 k_z \sin\vartheta - k_x\cos\vartheta)\right\} \tag{3.16}$$

where B_s^a, B_b^a and λ are given in the explanations to Table II.

In deriving the above formulae we utilized the fact that the $0z$-axis of the coordinate system is magnetically preferred. Unfortunately, despite the simplifications (3.13)–(3.16) the spin wave energy spectrum in the form (3.9a) cannot be used for thermodynamical calculations and, therefore, we are forced to make further approximations.

Let us expand E_k in a power series with respect to the ratio $|K_k|/N_k$

$$E_k = N_k\{1-\tfrac{1}{2}(|K_k|/N_k)^2 - \ldots\}. \tag{3.17}$$

TABLE III

Solution	$X_{0,s}$	$X_{1,s}$	$X_{2,s}$	$X_{3,s}$
$\theta=0$	$B+B_s^a$	$Sa^2(2A_s+D_s)$		$2S\varepsilon^2a^2\lambda(A_s-D_s)$
$\theta=\pi/2$ $B>B_s^a$	$\frac{1}{2}Bg_2$	$2Sa^2(A_s-D_s)$	$Sa^2(2A_s+D_s)$	$Sa^2\varepsilon^2\lambda(2A_s+D_s)$
	Bg_1	$Sa^2(A_s-D_s)g_1^{-1}g_2$	$Sa^2(A_sg_2+D_sg_3)g_1^{-1}$	$Sa^2\varepsilon^2\lambda(g_2A_s+g_4D_s)g_1^{-1}$
$\theta=\pi/2$ $B<B_s^a$	$\frac{1}{2}B_s^aG_1$	$Sa^2(2A_s+D_sG_3)$	$Sa^2(2A_s+D_s)$	$2Sa^2\varepsilon^2\lambda(A_s-\frac{1}{2}D_sG_4)$
	$B_s^aG_0$	$Sa^2(A_sG_1+D_sG_2)G_0^{-1}$		$Sa^2\varepsilon^2\lambda(A_sG_1-2D_sG_2)G_0^{-1}$

$G_0=[1-(B/B_s^a)^2]^{1/2}$ $\quad G_3=3G_1-5$ $\quad g_2=2-B_s^a/B$

$G_1=2-(B/B_s^a)^2$ $\quad G_4=G_3+1$ $\quad g_3=2g_2-3$

$G_2=2G_1-3$ $\quad g_1=[1-B_s^a/B]^{1/2}$ $\quad g_4=3-g_2$

TABLE IV

Solution	$X_{0,b}$	$X_{1,b}$	$X_{2,b}$	$X_{3,b}$	$X_{4,b}$
$\theta=0$	$B+B_b^a$	$Sa^2\left[2(A_b-D_b)+\frac{6D_b}{\varepsilon^2+2}\right]$		$\varepsilon^2X_{1,b}$	
$\theta=\pi/2$ $B>B_b^a$	$B\Gamma_0$	$Sa^2\Gamma_4$			
	$B\Gamma_1$	$\left[Sa^2\Gamma_0\Gamma_4+\frac{a^2B}{32}(B_b^a/B)^2\right]\Gamma_1^{-1}$			
$\theta=\pi/2$ $B<B_b^a$	$B_b^a\Gamma_3$	$Sa^2\Gamma_5$			$\frac{\varepsilon a^2B}{4(1-\varepsilon^2)}\Gamma_2$
	$B_b^a\Gamma_2$	$\left[Sa^2\Gamma_3\Gamma_5+\frac{a^2B_b^a}{32}(B/B_b^a)^4\right]\Gamma_2^{-1}$			$\frac{\varepsilon a^2B}{4(1-\varepsilon^2)}\left[\frac{\varepsilon-1}{2}(B/B_b^a)^2+1\right]$

$\Gamma_0=1-B_b^a/2B$ $\quad \Gamma_2=[1-(B/B_b^a)^2]^{1/2}$ $\quad \Gamma_4=2(A_b-D_b)+3D_b\frac{\varepsilon^2+1}{\varepsilon^2+2}$

$\Gamma_1=(1-B_b^a/B)^{1/2}$ $\quad \Gamma_3=1-{}^1/_2(B/B_b^a)^2$ $\quad \Gamma_5=2(A_b-D_b)+\frac{3D_b}{\varepsilon^2+2}\left[(\varepsilon^2-1)(B/B_b^a)^2+2\right]$

The confinement to the first term of the above series is justified for appropriate field strengths depending on the field direction. A careful analysis shows that in the case $\theta = 0$ the approximation $E_k = N_k$ is valid for arbitrary B. In the case of a transversal field, $\theta = \pi/2$, this approximation requires for the solution $\cos \vartheta = 0$ the condition $B > B_j^a$ to be satisfied, and for the solution $\sin \vartheta = B/B_j^a$ the field must be small as compared with the critical field, *i. e.*, $B < B_j^a$. In the case $\theta = \pi/4$ there is no restriction whatever on the strength of the external magnetic field. For fields $B \approx B_j^a$, the approximation $E_k = N_k$ is inadmissible in the case $\theta = \pi/2$ and the condition (3.12) is violated.

Another way of approximating the energy $E_k = E(k_x, k_y, k_z)$ is the expansion of the square root in the formula (3.9a) in a power series with respect to small wave vectors [18]. From examining the energy spectrum we get field restrictions similar to those stemming from the approximation (3.17). In Tables III and IV the results of both approximations are presented respectively for the simple tetragonal and body-centred tetragonal lattice. The first row for each solution corresponds to the expansion (3.17) and the approximation $E_k = N_k$. In each case, the resulting (approximate) spin wave energy spectrum has the form

$$E_k = X_{0,j} + X_{1,j}k_x^2 + X_{2,j}k_y^2 + X_{3,j}k_z^2 + X_{4,j}k_xk_z \tag{3.18}$$

where $j = s, b$ for the simple and body-centred tetragonal lattice, respectively. Because of the complicated form of E_k for $\theta = \pi/4$ we do not present in Tables III and IV the respective formulae for this case (which was not considered in [18]).

4. *Magnetization, susceptibility and specific heat*

Let us examine, first of all, the magnetization and susceptibility at zero temperature as functions of the strength of the transversal magnetic field. The saturation magnetization for the simple tetragonal and body-centred tetragonal lattice, respectively, is equal to

$$M_0^s = \frac{\mu S}{\varepsilon a^3}, \quad M_0^b = \frac{2\mu S}{\varepsilon a^3}. \tag{4.1}$$

Considering the case $B < B_j^a$, we get for the transversal (*i. e.*, perpendicular to the anisotropy axis) component of the magnetization, in the approximation (3.12), the formula (*cf.* [24])

$$M_\perp^j = M_0^j \frac{B}{B_j^a} \qquad j = s, b \tag{4.2}$$

and for the susceptibility components

$$\chi_\perp^j = \frac{\mu M_0^j}{B_j^a} = \text{const} > 0, \quad \chi_\parallel^j = -\frac{\mu M_0^j B}{(B_j^a)^2}\left[1 - \left(\frac{B}{B_j^a}\right)^2\right]^{-1/2}. \tag{4.3}$$

The dependence of $\chi_\parallel^j$ on B/B_j^a is shown in Fig. 1. On the other hand, one easily verifies that $M_\perp^j = M_0^j$ and $\chi_\perp^j = 0$ for $B > B_j^a$. Hence, when extrapolating our results to the critical field region we find that the transversal component of the magnetization is a continuous function of the field strength, whereas $\chi_\perp^j$ has a jump for $B = B_j^a$ and $\chi_\parallel^j$ has a singularity at this point. This indicates a second-order phase transition.

For the case $\theta = 0$ (field parallel to the anisotropy axis), one easily proves that $M^j_\perp = \chi^j_{||} = 0$ and $M^j_{||} = M^j_0$, as defined by (4.1).

If the field forms the angle $\theta = \pi/4$ with the easy axis, the parallel to the field component of the magnetization has the form

$$M^j_{\pi/4} = M^j_0(1-\varphi^2)^{1/2} \tag{4.4}$$

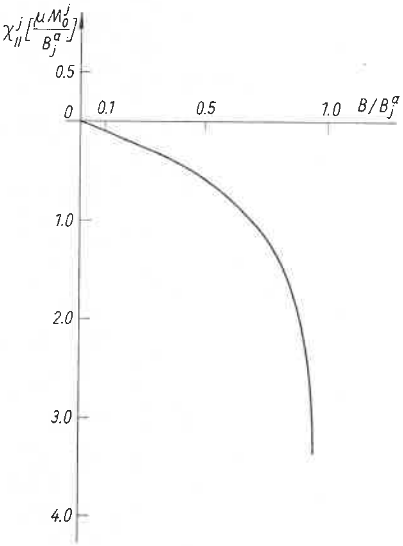

Fig. 1

where

$$\varphi = \frac{1}{2}\left\{\frac{B}{B^a_j} - \left[\left(\frac{B}{B^a_j}\right)^2 + 2\right]^{1/2}\right\}. \tag{4.5}$$

Let us calculate now the magnetization for $T > 0$. By virtue of (2.13), (3.2) and (3.9) we have for the free energy

$$F = E_0 + \Delta E_0 + k_B T \sum_k \ln\,(1 - e^{-E_k/k_B T}). \tag{4.6}$$

The parallel to the field component of the magnetization is defined as

$$M[T, B] = -\frac{\mu}{V}\frac{\partial F}{\partial B} = M[0, B] - \Delta M[T, B], \tag{4.7}$$

$$M[0, B] = -\frac{\mu}{V}\frac{\partial}{\partial B}(E_0 + \Delta E_0), \tag{4.8}$$

$$\Delta M[T, B] = \frac{\mu}{V}\sum_k \frac{\partial E_k}{\partial B}\langle n_k \rangle, \tag{4.9}$$

where

$$\langle n_k \rangle = (e^{E_k/k_BT}-1)^{-1} \tag{4.10}$$

and V is the volume of the crystal. Restricting ourselves to low temperatures and to E_k in the form (3.18), we get for $\Delta M[T, B]$ the expression (A.1) of Appendix A. Similarly, the spin specific heat $C = \dfrac{dU}{dT}$ can be calculated from the internal energy

$$U = \sum_k E_k \langle n_k \rangle \tag{4.11}$$

which leads in the case of the energy spectrum (3.18) to the formula (A.2) of Appendix A.

For the final thermodynamical calculations we shall use our approximation $E_k = N_k$ based on the expansion (3.17). As the structure of the formulae for $M^j[T, B]$, the magnetic susceptibility $\chi^j = \mu \dfrac{dM^j[T, B]}{dB}$ and the spin specific heat C are identical when $\theta = 0$ and $\theta = \pi/2$ for $B > B_j^a$, both cases can be described by the same formulae

$$M^j[T, B] = M_0^j\left\{1 - \frac{1}{2S}\left(\frac{k_BT}{4\pi r_j}\right)^{3/2} Z_{3/2}\left[\frac{h_j}{k_BT}\right]\right\}, \tag{4.12}$$

$$\chi^j[T, B] = \frac{\mu M_0^j}{8\pi S r_j}\left(\frac{k_BT}{4\pi r_j}\right)^{1/2} Z_{1/2}\left[\frac{h_j}{k_BT}\right], \tag{4.13}$$

$$C = \frac{1}{\varepsilon a^3 e_j}\left(\frac{k_B}{4\pi}\right)^{3/2}\left\{\frac{h_j^2}{k_B} T^{-1/2} Z_{1/2}\left[\frac{h_j}{k_BT}\right] + \right.$$

$$\left. +3h_j T^{1/2} Z_{3/2}\left[\frac{h_j}{k_BT}\right] + \frac{15}{4} k_B T^{3/2} Z_{5/2}\left[\frac{h_j}{k_BT}\right]\right\} \tag{4.14}$$

where

$$Z_p\left[\frac{h_j}{k_BT}\right] = \sum_{n=1}^{\infty} n^{-p} e^{-nh_j/k_BT}. \tag{4.15}$$

In the case of the longitudinal field ($\theta = 0$) we have in the formulae (4.12)–(4.14)

$$h_j = B + B_j^a, \tag{4.16}$$

$$r_s \equiv r_s^{||} = S(2A_s + D_s)^{2/3}\left[\frac{\lambda}{2}(A_s - D_s)\right]^{\frac{1}{3}}, \tag{4.17a}$$

$$r_b \equiv r_b^{||} = 2S\left(A_b - D_b\frac{\varepsilon^2-1}{\varepsilon^2+2}\right), \tag{4.17b}$$

$$e_j = r_j^{3/2}. \tag{4.18}$$

From (4.12) and (4.16) it follows that the decrease of the spontaneous magnetization with increasing temperature is slower in anisotropic than in isotropic ($B_j^a = 0$) ferromagnets.

For the transversal field $\theta = \pi/2$ (and $B > B_j^a$) we have in the formulae (4.12)-(4.14)

$$h_j = B - \tfrac{1}{2} B_j^a \tag{4.19}$$

$$r_s \equiv r_s^{\perp} = r_s^{\parallel}, \quad r_b \equiv r_b^{\perp} = S\left(2A_b + D_b^{\perp} \frac{\varepsilon^2 - 1}{\varepsilon^2 + 2}\right), \quad e_j = r_j^{3/2}. \tag{4.20}$$

Taking $h_j \approx B$ (*i. e.*, $B \gg B_j^a$) we get from (4.12) a formula for the magnetization similar to that obtained in [18].

When $\theta = \pi/2$ and $B < B_j^a$, the temperature-dependent part of the magnetization (4.7) has the following form

$$\Delta M^j[T, B] = \frac{\mu}{\varepsilon a^3} \mathscr{L}_j^{-1/2} \left(\frac{k_B T}{4\pi}\right)^{3/2} \mathscr{P}_j Z_{3/2}\left[\frac{h_j}{k_B T}\right] \tag{4.21}$$

where

$$h_j = B_j^a - \frac{B^2}{2B_j^a}, \quad \mathscr{P}_j = -\frac{B}{B_j^a} \tag{4.22}$$

and the coefficients $\mathscr{L}_j$ are given in Appendix B. The temperature-independent part $M^j[0, B] = M^j_{\perp}$ in this case is given by Eq. (4.2). Eq. (4.21) leads, for the approximation $B \ll B_j^a$, to the following formulae for the component of the magnetization (4.7) for both crystal lattices:

$$M_{\perp}^j[T, B] = M_0^j \frac{B}{B_j^a} \left\{1 + \frac{1}{2S}\left(\frac{k_B T}{4\pi r_j^{\parallel}}\right)^{3/2} Z_{3/2}\left[\frac{B_j^a}{k_B T}\right]\right\}. \tag{4.23}$$

Thus, for $B < B_j^a$ the transversal component of the magnetization decreases with increasing temperature (at constant field), quite like in the case $B > B_j^a$ (*cf.* Eqs (4.12), (4.19) and (4.20)).

The formulae (4.23) differ essentially from those obtained in [18] by the minus sign of the temperature term.

The dependence of the susceptibility on temperature is in the case $\theta = \pi/2$, $B < B_j^a$ given by the formula

$$\Delta\chi^j[T, B] = \chi^j[0, B] - \chi^j[T, B] = \frac{\mu^2}{\varepsilon a^3}\left(\frac{k_B}{4\pi}\right)^{3/2}\left\{W_{1,j}\frac{T^{1/2}}{k_B} Z_{1/2}\left[\frac{h_j}{k_B T}\right] + \right.$$

$$\left. + W_{2,j} T^{3/2} Z_{3/2}\left[\frac{h_j}{k_B T}\right]\right\} \tag{4.24}$$

where $\chi^j[0, B] = \chi^j_{\perp}$, as defined in Eq. (4.3).

The coefficients W depend only on the external field and on material constants and are given in Appendix C for the simple and body-centred tetragonal lattices. The spin specific heat is still given by the formula (4.14), where h_j is defined by Eq. (4.22) and

$$e_j^2 = \mathscr{L}_j \tag{4.25}$$

(*cf.* Appendix B).

For the case $\theta = \pi/4$ and $T > 0$, the magnetization along the field and the specific heat are given again by Eqs (4.7), (4.21), (4.14) and (4.25), except that $M^j[0, B] = M^j_{\pi/4}$ as defined by Eq. (4.4), and the coefficients $\mathscr{L}_j$ and $\mathscr{P}_j$ as well as h_j are (for the simple tetragonal lattice only) given in Appendix D. The corresponding susceptibility can easily be calculated according to Eq. (4.13).

5. *Concluding remarks*

In approximating the spin-wave energy spectrum (3.9a) according to (3.17) and (3.18), we restricted ourselves to small spin-wave vectors and utilized formulae (3.13) and (3.15). For the case $\theta = \pi/2$ this leads to restrictions for the field strength (*cf.* text following Eq. (3.17) and Tables III, IV) which automatically ensure the condition (3.12) to be satisfied. It is instructive to examine the accuracy of this approximation, which we shall demonstrate briefly.

One easily proves from Table II and Eqs (3.13)–(3.16) that in either case, $B > B_j^a$ and $B > B_j^a$, the approximation $E_k \approx N_k$ appears to be least accurate for $k = 0$, and that for the transversal-field case ($\theta = \pi/2$) one has from Eqs (3.13)–(3.17)

$$E_{k=0} = B\left(1 - \frac{1}{2}\frac{B_j^a}{B}\right)\left[1 - \frac{1}{8}\left(\frac{B_j^a}{B}\right)^2\left(1 - \frac{1}{2}\frac{B_j^a}{B}\right)^{-2} - \dots\right] \tag{5.1}$$

for the paramagnetic solution $\cos\vartheta = 0$ ($B > B_j^a$), and

$$E_{k=0} = B_j^a\left[1 - \frac{1}{2}\left(\frac{B}{B_j^a}\right)^2\right]\left\{1 - \frac{1}{8}\left(\frac{B}{B_j^a}\right)^4\left[1 - \frac{1}{2}\left(\frac{B}{B_j^a}\right)^2\right]^{-2} - \dots\right\} \tag{5.2}$$

for the ferromagnetic solution $\sin\vartheta = B/B_j^a$ ($B < B_j^a$). Now, one easily verifies that the second term of the series in the square brackets (which corresponds to $|K_k|^2/2N_k^2$ in Eq. (3.17)) amounts to 0.1 and 0.01 respectively for $B_j^a/B = 0.6$ and 0.25 in (5.1), and for $B/B_j^a = 0.7$ and 0.5 in (5.2). This shows that our approximation based on the expansion (3.17) is actually quite reasonable. Unfortunately, although there apparently do exist uniaxial ferromagnets with tetragonal symmetry (*cf.*, *e. g.*, [29–31]), no reliable experimental measurements of the magnetization or susceptibility on single-crystalline samples are so far available.

The main theoretical results of the present paper reside in showing that (*i*) pseudo-dipolar spin coupling can effectively be used in describing uniaxial magnetic anisotropy in ferromagnetic materials with tetragonal symmetry-including (*ii*) field-induced magnetic phase transitions, and that (*iii*) the mathematically inconvenient spin-wave energy spectrum (3.9a) can be satisfactorily approximated by using the power-series expansion (3.17) combined with the long-wavelength approximation (3.13)–(3.16). In contrast, the corresponding approximations applied in [18] are not only more laborious but also more restrictive and less accurate for small wave-vectors to which, in fact, the thermodynamical calculations are usually confined (low-temperature approximation). On the other hand, in comparison with the Green's-function approach given in [17] our method has clear advantage that it permits one to obtain effective formulae for the magnetization, susceptibility and specific heat.

In Part II, we shall show that our approach permits us to take into account spin-wave interactions of lowest order and leads in the transversal-field case to a temperature-dependent magnetization direction in the ferromagnetic phase (*i. e.*, ϑ depends on temperature).

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APPENDIX A

By utilizing Eqs (4.9), (4.10) and the standard approximation [9]

$$\sum_{\boldsymbol{k}} f(\boldsymbol{k}) \to \frac{V}{(2\pi)^3} \int f(\boldsymbol{k}) d^3k$$

we get for the approximate energy spectrum (3.18)

$$\begin{aligned} \Delta M[T, B] = \mu \left(\frac{k_B T}{4\pi}\right)^{3/2} [X_{1,j} X_{2,j} (X_{3,j} - \tfrac{1}{4} X_{4,j}^2 X_{1,j}^{-1})]^{-1/2} \times \\ \times \Bigg\{ Y_{0,j} Z_{3/2} \left[\frac{X_{0,j}}{k_B T}\right] + \frac{k_B T}{2} \Bigg[Y_{1,j} X_{1,j}^{-1} + Y_{2,j} X_{2,j}^{-1} + \\ + \frac{Y_{3,j} - \tfrac{1}{2} X_{4,j} X_{1,j}^{-1} (Y_{4,j} - \tfrac{1}{2} X_{4,j} X_{1,j}^{-1} Y_{1,j})}{X_{3,j} - \tfrac{1}{4} X_{4,j}^2 X_{1,j}^{-1}} \Bigg] Z_{5/2} \left[\frac{X_{0,j}}{k_B T}\right] \Bigg\} \end{aligned} \tag{A.1}$$

where

$$Y_{m,j} = \frac{\partial}{\partial B} X_{m,j}; \quad m = 0, 1, 2, 3, 4; \quad j = s, b.$$

Analogously, we calculate the specific heat C from the internal energy (4.11)

$$\begin{aligned} C = \left(\frac{k_B}{4\pi}\right)^{3/2} X_{1,j} X_{2,j} (X_{3,j} - \tfrac{1}{4} X_{4,j}^2 X_{1,j}^{-1})]^{-1/2} \times \\ \times \left\{ X_{0,j}^2 k_B^{-1} T^{-1/2} Z_{1/2} \left[\frac{X_{0,j}}{k_B T}\right] + 3 X_{0,j} T^{1/2} Z_{3/2} \left[\frac{X_{0,j}}{k_B T}\right] + \frac{15}{4} k_B T^{3/2} Z_{5/2} \left[\frac{X_{0,j}}{k_B T}\right] \right\}. \end{aligned} \tag{A.2}$$

APPENDIX B

The coefficients $\mathscr{L}_j$ appearing in Eq. (4.21) are as follows for the simple tetragonal lattice

$$\mathscr{L}_s = S^3 \lambda (2A_s + D_s) \left\{ 2A_s + D_s \left[1 - 3 \left(\frac{B}{B_s^a}\right)^2 \right] \right\} \left\{ 2A_s - D_s \left[2 - 3 \left(\frac{B}{B_s^a}\right)^2 \right] \right\}; \tag{B.1}$$

for the body-centred tetragonal lattice

$$\mathscr{L}_b = \mathscr{T} \left\{ \mathscr{T}^2 - \left[\frac{B}{8(\varepsilon^2 - 1)}\right]^2 \left[1 - \left(\frac{B}{B_b^a}\right)^2 \right] \right\} \tag{B.2}$$

where

$$\mathscr{T} = 2S(A_b - D_b) + \frac{B_b^a}{16} \left[\left(\frac{B}{B_b^a}\right)^2 + \frac{2}{\varepsilon^2 - 1} \right] \tag{B.3}$$

APPENDIX C

The coefficients W appearing in Eq. (4.24) are as follows

$$W_{1,j} = \mathscr{L}_j^{-1/2}\left(\frac{B}{B_j^a}\right)^2 \tag{C.1}$$

$$W_{2,s} = \frac{1}{B_s^a}\mathscr{L}_s^{-1/2} - \frac{3}{4}\frac{\lambda}{\lambda-1}S^2(2A_s+D_s)D_s\mathscr{L}_s^{3/2}\left(\frac{B}{B_s^a}\right)^2\left[1-2\left(\frac{B}{B_s^a}\right)^2\right] \tag{C.2}$$

$$W_{2,b} = \frac{1}{B_b^a}\mathscr{L}_b^{-1/2} - \frac{1}{16}\mathscr{L}_b^{-3/2}\left(\frac{B}{B_b^a}\right)^2\Bigg\{2\mathscr{T}^2+\mathscr{L}_b\mathscr{T}^{-1} -$$

$$-\frac{1}{4}\mathscr{T}\frac{B_b^a}{(\varepsilon^2-1)^2}\left[1-2\left(\frac{B}{B_b^a}\right)^2\right]\Bigg\} \tag{C.3}$$

where the coefficients $\mathscr{L}_j$, $\mathscr{T}$ are given in Appendix B.

APPENDIX D

The field-variable h_s and the coefficients $\mathscr{L}_s$, $\mathscr{P}_s$ for the case $\theta = \pi/4$ are as follows

$$h_s = \frac{1}{2}B_s^a\left[2-3\sin^2\left(\frac{\pi}{4}+\arcsin\varphi\right)\right]+B(1-\varphi^2)^{1/2} \tag{D.1}$$

$$\mathscr{L}_s = S^3\lambda(2A_s+D_s)\left[2A_s+D_s\left[1-3\sin^2\left(\frac{\pi}{4}+\arcsin\varphi\right)\right]\right]\times$$

$$\times\left[2A_s-D_s\left[2-3\sin^2\left(\frac{\pi}{4}+\arcsin\varphi\right)\right]\right] \tag{D.2}$$

$$\mathscr{P}_s = (1-\varphi^2)^{-1/2} \tag{D.3}$$

where φ is given in (4.5).

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