# SPIN WAVE THEORY OF UNIAXIAL FINITE FERROMAGNETS. PART I. DETERMINATION OF THE GROUND STATE*

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For a uniaxial finite ferromagnet with nearest-neighboure isotropic exchange interactions, crystal-field anisotropy, and magnetostatic self-energy, the approximate ground state of the system is determined by examining several magnetically ordered spin-deviation reference states (spin-wave vacua) and their corresponding spin wave energy spectra. In Part I it is shown that a ferromagnetic spin configuration (saturation state) is inadmissible as reference state in the field-free case, and that the same holds for a magnetic field parallel to the anisotropy axis unless it exceeds a certain critical value and the spins are aligned along the field. It is also shown that, in the field-free case and in the first-order approximation of Wallace's Hamiltonian perturbation method, a simple 180° domain-structure configuration is a suitable reference state, as it ensures a real and non-negative spin wave energy spectrum. Moreover, the minimization of the corresponding ground state energy leads to the familiar Landau-Lifshitz half-power law for the thickness dependence of the domain width. In Part II of this paper this reference state is improved by considering a refined and more realistic 180° domain-structure configuration which ensures real and non-negative spin wave energies up to the second-order of perturbation procedure.

## *1. Introduction*

In the standard spin-wave approach to anisotropic Heisenberg ferromagnets, one usually assumes — quite like in isotropic ferromagnets — the spin-deviation reference state (or, equivalently, the spin wave vacuum) to be a state of parallel spin alignment [1, 2, 3] (saturation state). For uniaxial ferromagnets, this is certainly correct as long as surface effects (*i. e.*, the magnetostatic self-energy) or, correspondingly, long-range spin interactions are neglected, because in this case the only difference between the isotropic and the anisotropic ferromagnet lies in that that in the latter there exists (even in the absence of an external magnetic field) a magnetically preferred direction. Therefore, the saturation state remains the exact ground state (or a good approximation thereof — if the external field is not

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parallel to the anisotropy axis) of the system and its choice as homogeneous spin-deviation reference state is apparently justified.

However, the situation is quite different for multiaxial ferromagnets, or even for uniaxial ones if long-range interactions are to be taken into account. It has been emphasized in [4] that the spin-deviation reference state for anisotropic ferromagnets should actually correspond to the domain structure which apparently represents the equilibrium low-temperature state of true ferromagnets. This requires the use of inhomogeneous spin--deviation reference states as outlined in [4, 5]. The purpose of our paper is to apply the spin--wave theory as formulated in [4] to a finite uniaxial ferromagnet with the magnetostatic self-energy included. In Part I we examine several magnetically ordered spin-deviation reference states and show that in the absence of an external magnetic field saturation states are inadmissible as they lead to negative or imaginary spin wave energies. It is also shown that a reference state corresponding to a simplified 180° domain-structure configuration ensures up to the first order of Wallace's Hamiltonian perturbation method [6] real and non-negative spin wave energies. As expected (see, *e. g.*, Appendix VI in [7]), the minimization of the corresponding approximate ground state energy leads to a finite domain witdh whose dependence on the crystal thickness is in fair order-of-magnitude agreement with experiment. In Part II, the results obtained here are utilized in applying the spin wave theory to the same finite uniaxial ferromagnet with a more realistic 180° domain structure.

## 2. *Hamiltonian*

We consider a finite uniaxial ferromagnetic crystal of plate-like shape, cut perpendicularly to the anisotropy direction. We assume its thickness along the anisotropy axis to be very small as compared to its remaining dimensions, so that we can neglect the magnetostatic self-energy stemming from its lateral surfaces. For simplicity, we assume a simple cubic crystal lattice[1] with the lattice constant $a$.

We choose the Hamiltonian of the crystal in the form

$$H = \sum_{fg} J\mathbf{S}_f\mathbf{S}_{f+g} + \sum_f [K(S_f^z)^2 - \mu h_f S_f^z], \tag{1}$$

where $J$ denotes the (negative) nearest-neighbour exchange integral, $K$ the (negative) crystal--field anisotropy constant, and $\mu$ Bohr's magneton. The lower indices $f$ and $g$ represent respectively the ordinary and nearest-neighbour lattice *vectors*. (Note that in our simplified notation $f+g$ is a vector sum.) The coordinate system's axes coincide with the $\langle 100 \rangle$ crystallographic directions. The effective field $h_f$ consists of the internal $h_f^i$ (*i. e.*, demagnetizing) as well as the external magnetic field $h^e$, which are assumed to be always parallel to the anisotropy direction. From the phenomenological point of view the internal magnetic field re-

[1] The assumption of a uniaxial anisotropy to be consistent with the crystal symmetry necessitates actually a crystal lattice with uniaxial symmetry, *e. g.*, a tetragonal or hexagonal crystal structure. However, the assumption of a cubic crystal symmetry considerably simplifies the calculations, the result being qualitatively the same as for the tetragona lattice. This suits the purpose of our paper which resides in demonstrating the method rather than in obtaining precise quantitative results.

presents the long-range magnetostatic dipol-dipol interactions and can be calculated if the distribution of the magnetic poles on the crystal surface is known (see [8] for a detailed discussion of the problem).

According to [4] the approximate ground state of the Hamiltonian (1) is to be obtained by minimizing its expectation value in the class of trial states $U^+|0\rangle$ where the state $|0\rangle$ is defined by $S_f^z|0\rangle = -S|0\rangle$, $S_f^-|0\rangle = 0$, $S$ being the maximum spin eigenvalue, and the unitary transformation $U$ generates spatial rotations of the lattice spins. This, of course, is equivalent to transforming the Hamiltonian (1) and minimizing its expectation value in the state $|0\rangle$. As shown in [9, 10, 11] for quite general spin Hamiltonian, the necessary conditions of this minimizing procedure coincide with those obtained when eliminating from the Hamiltonian terms linear with respect to the Bose creation and annihilation operators, upon mapping the spin Hamiltonian according to the Holstein-Primakoff [12] or Dyson-Maleev [13] mapping rules (see also [14, 15]).

To simplify our problem we shall confine ourselves to a unitary transformation of the type

$$U = \prod_f \exp\,(i\varphi_f S_f^x), \tag{2}$$

which corresponds to (generally inhomogeneous) rotations of the lattice spins by the angles $\varphi_f$ around the $x$-axis. In replacing the spin operators in the Hamiltonian by Bose creation and annihilation operators we use the Holstein-Primakoff mapping in the lowest approximation, *i. e.*,

$$S_f^+ \to \sqrt{2S}\,\alpha_f^+, \quad S_f^- \to \sqrt{2S}\,\alpha_f,$$
$$S_f^z \to -S+\alpha_f^+\alpha_f. \tag{3}$$

By applying the transformation (2) and the mapping (3) to the Hamiltonian (1) one obtaines, upon writting the Bose operator products in normal order and neglecting higher h an second-order terms (non-interacting spin-waves approximation),

$$H = E+H_{\mathrm{I}}+H_{\mathrm{II}}, \tag{4}$$

where

$$E = \sum_{fg} JS^2 \cos\,(\varphi_f-\varphi_{f+g}) + \sum_f \{\tfrac{1}{2}KS[1+(2S-1)\cos^2\varphi_f]+\mu h_f S\cos\varphi_f\}, \tag{4a}$$

$$H_{\mathrm{I}} = \tfrac{1}{2}i\sqrt{2S}\,\{\sum_{fg} JS\sin\,(\varphi_f-\varphi_{f+g})\,(\alpha_f^+-\alpha_f-\alpha_{f+g}^+ +\alpha_{f+g})+$$
$$+\sum_f [K(2S-1)\sin\varphi_f\cos\varphi_f+\mu h_f\sin\varphi_f](\alpha_f^+-\alpha_f)\}, \tag{4b}$$

$$H_{\mathrm{II}} = \sum_{fg} JS\{\tfrac{1}{2}[1-\cos\,(\varphi_f-\varphi_{f+g})]\,(\alpha_f^+\alpha_{f+g}^+ +\alpha_f\alpha_{f+g})+$$
$$+\tfrac{1}{2}[1+\cos\,(\varphi_f-\varphi_{f+g})](\alpha_f^+\alpha_{f+g}+\alpha_{f+g}^+\alpha_f)-\cos\,(\varphi_f-\varphi_{f+g})(\alpha_f^+\alpha_f+\alpha_{f+g}^+\alpha_f)\}+$$
$$+\sum_f \{[K(3S-1)\sin^2\varphi_f-K(2S-1)-\mu h_f\cos\varphi_f]\,\alpha_f^+\alpha_f-\tfrac{1}{2}KS\sin^2\varphi_f(\alpha_f^+\alpha_f^+ +\alpha_f\alpha_f)\}. \tag{4c}$$

We shall determine the spin-deviation reference state (approximate ground state) by equating the coefficients of the linear terms in $H_I$ to zero, *i. e.*,

$$2JS\sum_g \sin(\varphi_f-\varphi_{f+g})+K(2S-1)\sin\varphi_f\cos\varphi_f+\mu h_f\sin\varphi_f=0. \tag{5}$$

As already pointed out, this is equivalent to determining the minimizing parameters from the necessary conditions $\partial E/\partial\varphi_f=0$ of the minimization procedure, since $\langle 0|UHU^+|0\rangle = E$. Out of the numerous solutions of Eq. (5) we should select the stable ones by examining the sufficient conditions for a minimum of $E$ to exist, which in our case is practically impossible because of the large number of variables $\varphi_f$. We shall therefore select the proper solution by examining the spin-wave energy spectra corresponding to certain magnetically ordered reference states following from (5).

First of all let us further simplify the problem by assuming that for lattice sites lying in the same (100) crystal plane the angles $\varphi_f$ are identical, *i. e.*, that $\varphi_f$ depends only on the $x$-component $f_x$ of the lattice vector $f$; consistently, one must ensure that the demagnetizing field in $h_f$, which in this case is independent of $y$, does not depend on $z$, too. Secondly, we confine ourselves to studying only four solutions of Eq. (5) corresponding to those magnetically ordered reference states which are of foremost interest to our problem.

### 3. *Ferromagnetic configurations*

One easily verifies that the conditions (5) obtained for an external field parallel to the anisotropy axis admit in general two ferromagnetic spin configurations $\varphi_f=\text{const}\equiv\varphi$ (saturation states), namely,

$$\sin\varphi=0, \tag{6}$$

and

$$\cos\varphi=\frac{h^e}{h_0+K(2S-1)/\mu} \tag{7}$$

where $h_0=2\pi\mu Sa^{-3}$ is the demagnetizing field for the case of complete saturation along the anisotropy axis (*i. e.*, perpendicular to the crystal surface). Indeed, if the sample is saturated in the direction $\varphi+\pi$ we have[2]

$$h_f=h_f^i-h^e=h_0\cos\varphi-h^e \tag{8}$$

which upon inserting in (5) leads to the solutions (6) and (7). Note that the solution (7) is restricted to fields

$$|h^e|\leqslant h_0+K(2S-1)/\mu\equiv h_c. \tag{9}$$

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[2] Note that, according to our definition, the $z$-components on the spins are negative in the saturation state $|0>$ which corresponds to $\varphi_f=0$. Consequently, we assume the external magnetic field to be directed along the negative $z$-axis.

To find out whether the saturation states $|0\rangle$ and $\prod_f \exp(-i\varphi S_f^x)|0\rangle$ corresponding to the solutions (6) and (7) are admissible as reference states let us examine the spin-wave energy spectra they lead to.

a. Parallel ferromagnetic configuration

For the solution (6) of Eq. (5) the Hamiltonian (4) takes the form

$$H = 6NJS^2 + NKS^2 + N\mu(h_0 - h^e)S + \\ + \sum_{fg} JS(\alpha_f^+ \alpha_{f+g} + \alpha_{f+g}^+ \alpha_f - \alpha_f^+ \alpha_f - \alpha_{f+g}^+ \alpha_{f+g}) - \\ - \sum_f [K(2S-1) + \mu(h_0 - h^e)] \alpha_f^+ \alpha_f, \tag{10}$$

where $N$ is the number of lattice sites. In diagonalizing the above Hamiltonian we apply the Fourier transformation

$$\alpha_f^+ = N^{-1/2} \sum_k \exp(-ikf)\, \beta_k^+ \tag{11}$$

to the Hamiltonian (10). (We use a simplified notation, according to which $k$ denotes a reciprocal-lattice *vector*, and $kf$ is a *scalar* product.) Here and in the following we impose the Born-Karman periodicity conditions, which is justified for not too thin crystals (see, *e. g.*, [16]). Inserting (11) into (10) we have

$$H = E_0 + \sum_k \omega_k \beta_k^+ \beta_k \tag{12}$$

where

$$E_0 = NJS^2\gamma_0 + NKS^2 + N\mu(h_0 - h^e)\, S, \tag{13}$$

$$\omega_k = 2JS(\gamma_k - \gamma_0) - K(2S-1) - \mu(h_0 - h^e) \\ = 2JS(\gamma_k - \gamma_0) - K(2S-1) - 2\pi\mu^2 S a^{-3} + \mu h^e, \tag{14}$$

$$\gamma_k = \sum_g \exp(ikg). \tag{15}$$

As $-\gamma_0 \leqslant \gamma_k \leqslant \gamma_0$ and $J < 0$, it is seen that the spin wave energies $\omega_k$ are non-negative for all wave vectors $k$ if

$$h^e \geqslant 2\pi\mu S a^{-3} + K(2S-1)\mu^{-1} = h_c. \tag{16}$$

For example, assuming for the constants the values

$$J = -10^{-13}, \quad K = -10^{-17}, \quad S = 1; \\ a = 3 \cdot 10^{-8}, \quad \mu = 10^{-20}, \tag{17}$$

(in CGSM units) one gets from (16) that the external magnetic field must be stronger than $7 \cdot 10^3$ [Oe] if the ferromagnetic configuration parallel to the anisotropy axis is to be admissible as reference state.

b. Canted ferromagnetic configuration

Upon substituting the solution (7) of the condition (5) into the Hamiltonian (4) we have

$$H = 6NJS^2+\tfrac{1}{2}NKS-\tfrac{1}{2}NKS(2S-1)\,(h^e/h_c)^2+$$
$$+\sum_{fg} JS(\alpha_f^+\alpha_{f+g}+\alpha_{f+g}^+\alpha_f-\alpha_f^+\alpha_f-\alpha_{f+g}^+\alpha_{f+g})+$$
$$+\sum_{f} KS[1-(h^e/h_c)^2](\alpha_f^+\alpha_f-\tfrac{1}{2}\alpha_f^+\alpha_f^+-\tfrac{1}{2}\alpha_f\alpha_f), \tag{18}$$

or, by applying the Fourier transformation (11)

$$H = 6NJS^2+\tfrac{1}{2}NKS-\tfrac{1}{2}NKS(2S-1)(h^e/h_c)^2+$$
$$+\sum_{k} \{2JS(\gamma_k-\gamma_0)+KS[1-(h^e/h_c)^2]\}\beta_k^+\beta_k-$$
$$-\sum_{k} \tfrac{1}{2}KS[1-(h^e/h_c)^2](\beta_k^+\beta_{-k}^++\beta_k\beta_{-k}). \tag{19}$$

The Hamiltonian (19) can be diagonalized by the transformation

$$\beta_k^+ = u_k\xi_k+v_{-k}\xi_{-k}^+ \tag{20}$$

which leads to

$$H = E_0+\sum_{k} \omega_k\xi_k^+\xi_k \tag{21}$$

where

$$E_0 = NJS^2\gamma_0+\tfrac{1}{2}NKS-\tfrac{1}{2}NKS(2S-1)\,(h^e/h_c)^2-$$
$$-\tfrac{1}{2}K^2S^2[1-(h^e/h_c)^2]^2\sum_{k}\{2JS(\gamma_k-\gamma_0)+KS[1-(h^e/h_c)^2]+\omega_k\}^{-1}, \tag{22}$$

$$\omega_k = \sqrt{\{2JS(\gamma_k-\gamma_0)+KS[1-(h^e/h_c)^2]\}^2-K^2S^2\,[1-(h^e/h_c)^2]^2}\,. \tag{23}$$

As $J$ and $K$ are negative it is seen that in the field interval (9) where the solution (7) is valid, there is always a subset of wave vectors $k$ for which the spin-wave energies $\omega_k$ are imaginary. Hence, the canted ferromagnetic configurations described by the solution (7) are inadmissible as spin-deviation reference states, except at the boundary of the interval (9) where the solutions (7) and (6) coincide.

It should be emphasized that the particular choice of the rotation axis in the transformation (2) and of the simple cubic crystal lattice is immaterial for the conclusion of this section (except for the value of $h_c$).

The main conclusion to be drawn from the above considerations is that, if the magnetostatic self-energy is taken into account, the ferromagnetic configuration is inadmissible as spin-deviation reference state for $h^e < h_c$. This includes the field-free case to which for simplicity our further considerations shall be confined.

## *4. Domain structure configuration*

It may seem that in the field-free case the next best choice as reference state is an antiferromagnetic configuration along the anisotropy axis, as in that case the magnetostatic self-energy is negligible. One can prove, however, that this configuration, too, leads to spin-

-wave energies which are imaginary for certain wave vectors $k$. The same holds for the antiferromagnetic configuration perpendicular to the anisotropy axis whose anisotropy energy is even higher. No other antiferromagnetic configurations are admitted by the condition (5) in the case $h_f = 0$.

Thus, the above results lead quite naturally to the supposition that a configuration of domain-structure type might be a suitable reference state for our problem. We shall therefore examine a simple 180° domain structure configuration in which the crystal is divided into plate-like regions (domains) perpendicular to the rotation axis. The spins are ordered ferromagnetically along the anisotropy axis, being antiparallel in neighbouring domains. In this model the thickness of the interdomain walls is equal to the lattice constant $a$, and the width $\Delta$ of the domains is a parameter to be determined by minimizing the corresponding approximate ground state energy $E_0$.

The simple domain structure described above can be defined as follows:

$$\varphi_f = 0 \text{ for } 0 \leqslant f_x < \Delta,$$

$$\varphi_f = \pi \text{ for } \Delta \leqslant f_x < 2\Delta,$$

and

$$\varphi_{f+2\Delta} = \varphi_f, \tag{24}$$

where the symbol $f+2\Delta$ denotes a lattice vector whose $x$-component is equal to $f_x + 2\Delta$. One easily proves that the spin configuration defined by (24) satisfies the conditions (5) and is therefore admissible as reference state provided it leads to real and positive spin-wave energy spectrum.

By averaging the respective $z$-dependent demagnetizing field following from phenomenological considerations (see *e. g.*, [17]), the internal magnetic field $h_f^i$ in the case of the simple domain structure can be expressed, approximately, by the formula

$$h_f^i = \frac{2\mu S\Delta}{a^3 L} \cos\varphi_f = h_f \tag{25}$$

where $L$ denotes the thickness of the crystal along the anisotropy direction.

The Hamiltonian (4) for the crystal with the simple domain structure becomes

$$H = E_0 + \sum_{fg} JS(\alpha_f^+\alpha_{f+g} + \alpha_{f+g}^+\alpha_f - \alpha_f^+\alpha_f - \alpha_{f+g}^+\alpha_{f+g}) -$$

$$- \sum_f [K(2S-1)+\mu h]\,\alpha_f^+\alpha_f + \sum_{fn} 2JS(\alpha_f^+\alpha_{f-a}^+ + \alpha_f\alpha_{f-a} - \alpha_f^+\alpha_{f-a} - \alpha_{f-a}^+\alpha_f +$$

$$+ 2\alpha_f^+\alpha_f + 2\alpha_{f-a}^+\alpha_{f-a})\,\delta(f_x, n\Delta), \tag{26}$$

where

$$n = 0, \pm 1, \pm 2, \ldots,$$

$$E_0 = 6NJS^2 + NKS^2 + N\mu hS - 4NJS^2 a/\Delta, \tag{27}$$

$$h = \frac{2\mu S\Delta}{a^3 L}, \tag{28}$$

and

$$\delta(x, y) \equiv \delta_{x,y},$$

*i. e.*, the usual Kronecker delta.

The equilibrium domain width $\Delta$ as function of the crystal thickness $L$ can be determined by minimizing $E_0$, *i. e.*,

$$\frac{dE_0}{d\Delta} = NS^2\left(\frac{2\mu^2}{a^3L} + \frac{4Ja}{\Delta^2}\right) = 0, \tag{29}$$

$$\Delta = \frac{a^2}{\mu}\sqrt{-2JL}. \tag{30}$$

This is the well-known Landau-Lifshitz half-power law for thickness dependence of the domain width. For example, with the constants as in (17) and $L = 1$ cm we have $\Delta = 4 \cdot 10^{-2}$ cm, in fair order-of-magnitude agreement with experiment [18].

In diagonalizing the Hamiltonian (26), let us first note that the only coefficient which depends on $f$ is $\sum_n \delta(f_x, n\Delta)$. As it is a periodic function with the period $\Delta$, it can be expanded in a Fourier series as

$$\sum_n \delta(f_x, n\Delta) = \frac{a}{\Delta}\sum_m \exp(imqf) \tag{31}$$

where the reciprocal-lattice vector $q$ is defined by

$$q = \left(\frac{2\pi}{\Delta}, 0, 0\right)$$

and

$$m = 0, \pm 1, \ldots, \pm\left(\frac{\Delta}{2a} - 1\right), \frac{\Delta}{2a}. \tag{32}$$

By utilizing (31) and applying the Fourier transformation (11) the Hamiltonian (26) takes the form

$$H = E_0 + \sum_k \{a_k\beta_k^+\beta_k + \sum_m [d_k^m\beta_k^+\beta_{k-mq} + c_k\beta_k^+\beta_{-k+mq}^+ + c_k^*\beta_k\beta_{-k-mq}]\}, \tag{33}$$

where

$$\begin{aligned} a_k &= 2JS(\gamma_k - \gamma_0) - K(2S-1) - \mu h, \\ c_k &= 2JSa/\Delta \exp(iak), \\ d_k^m &= 2JSa/\Delta[2 + 2\exp(iamq) - \exp(-ia[k-mq]) - \exp(iak)]. \end{aligned} \tag{34}$$

The diagonalization of the Hamiltonian (33) by the use of the general Bogolubov transformation is a rather hopeless mathematical task of solving $2\Delta/a$ homogeneous linear equations. Therefore, in calculating the spin wave energy spectrum we shall use Wallace's Hamiltonian perturbation method [6] (equation-of-motion perturbation method). This method

resides in renormalizing the single-particle creation and annihilation operators and energies, so as to remove the interactions to successively higher orders of perturbation, for a system composed of many weakly interacting particles.

We choose the zeroth-order Hamiltonian as follows:

$$H_0 = E_0 + \sum_k \Big\{ \{a_k + d_k^0)\beta_k^+ \beta_k + \sum_m d_k^m \beta_k^+ \beta_{\bar{k}} \Big[ \delta\Big(k_x, \frac{m}{2} q\Big) + \\ + \delta\Big(k_x, \frac{m}{2} q + \frac{\Delta}{2a} q\Big)\Big] (1 - \delta_{m,0}) \Big\} \tag{35}$$

where

$$\bar{k} = k - 2k_x = -k_x + k_y + k_z. \tag{36}$$

(Note that (36) are vector sums.)

In Wallace's method the zeroth-order Hamiltonian should describe a system of non-interacting particles. Therefore, before starting the perturbation calculations we must diagonalize $H_0$. This can be done by means of the canonical transformation defined as follows:

$$\beta_k = \frac{1}{\sqrt{2}} (\xi_k + \xi_{\bar{k}}),$$

$$\beta_{\bar{k}} = \frac{1}{\sqrt{2}} \frac{d_k^{m*}}{|d_k^m|} (\xi_k - \xi_{\bar{k}}) \tag{37}$$

if

$$k_x = \frac{m}{2} q \quad \text{or} \quad k_x = \frac{m}{2} q + \frac{\Delta}{2a} q \quad \text{and} \quad m \neq 0, \tag{38}$$

and

$$\beta_k = \xi_k \tag{39}$$

otherwise. Then $H_0$ takes the form

$$H_0 = E_0 + \sum_k \omega_{0,k} \xi_k^+ \xi_k \tag{40}$$

with the zeroth-order spin wawe energy spectrum

$$\omega_{0,k} = a_k + d_k^0 + |d_k^m|$$
$$\omega_{0,\bar{k}} = a_k + d_k^0 - |d_k^m| \tag{41}$$

if (38) applies, and

$$\omega_{0,k} = a_k + d_k^0 \tag{42}$$

otherwise.

As is seen from (34) and (28), the zeroth-order energy spectrum (41), (42) is real and positive for all wave vectors $k$ provided

$$\frac{\Delta}{L} < \frac{|K|(2S-1)a^3}{6\mu^2 S} \sim 1 \tag{43}$$

or, due to (30),

$$L > \frac{72\mu^2 S^2 |J|}{K^2(2S-1)^2 a^2} \sim 10^{-3}\,\text{cm}, \tag{44}$$

with the estimation based on the values (17). The above reastrictions are reasonable in view of the phenomenologically derivable critical size for single-domain particles (see *e.g.*, [19]). Thus, in the zeroth-order approximation the domain-structure configuration (24) turns out to be admissible as spin-deviation reference state, with the restriction (44) for the crystal thickness along the anisotropy direction. However, as this is a perturbation procedure the question arises whether the first- or higher-order corrections to the spin wave energies (41), (42) do not alter this result. Let us therefore consider the first-order energy corrections.

The perturbation part of the Hamiltonian,

$$H_1 = H - H_0 = \sum_{k,m} \Big\{ c_k \beta_k^+ \beta_{-k+mq}^+ + c_k^* \beta_k \beta_{-k-mq} + d_k^m \beta_k^+ \beta_{k-mq} [1 - \delta(k_x, 0)] \times$$

$$\times \left[1 - \delta\left(k_x, \frac{m}{2} q\right)\right]\left[1 - \delta\left(k_x, \frac{m}{2} q + \frac{\Delta}{2a} q\right)\right]\Big\}, \tag{45}$$

is used to calculate the first-order energy corrections from the equation (see [6])

$$\langle \ldots n_k+1, \ldots n_l \ldots (0) | [H_1, \xi_{0,k}^+] | \ldots n_k, \ldots n_l \ldots (0) \rangle = (n_k+1)^{1/2} \omega_{1,k}. \tag{46}$$

The first suffix of Bose operators and energies denotes the approximation order of those quantities. For example: $\omega_{1,k}$ means the first-order energy corrections, and $\xi_{0,k}^+$ the zeroth--order creation operator. Note that $\xi_k^+$ as used in Eq. (40) is tantamount to $\xi_{0,k}^+$ in the present notation. The symbol (0) in the state vector indicates its zeroth-order, *i.e.*, it is an eigenvector of $H_0$.

The commutator $[H_1, \xi_{0,k}^+]$ is equal to

$$[H_1, \xi_{0,k}^+] = \frac{1}{\sqrt{2}} \sum_m \Big\{ (c_k^* + c_{k-mq}^*) \beta_{-k-mq} + \frac{d_k^{m*}}{|d_k^m|} (c_{\bar{k}}^* + c_{\bar{k}-mq}^*) \beta_{-\bar{k}-mq} +$$

$$+ \frac{d_k^{m*}}{|d_k^m|} d_{\bar{k}+mq}^m \beta_{\bar{k}+mq}^+ (1-\delta_{m,0}) \left[1 - \delta\left(k_x, \frac{m}{2} q\right)\right]\left[1 - \delta\left(k_x, \frac{m}{2} q + \frac{\Delta}{2a} q\right)\right] +$$

$$+ d_{k+mq}^m \beta_{k+mq}^+ (1-\delta_{m,0}) \left[1 - \delta\left(k_x, -\frac{m}{2} q\right)\right]\left[1 - \delta\left(k_x, -\frac{m}{2} q + \frac{\Delta}{2a} q\right)\right]\Big\}, \tag{47}$$

if (38) applies, and

$$[H_1, \xi_{0,k}^+] = \sum_m \{ (c_k^* + c_{k+mq}^*) \beta_{-k-mq} + d_{k+mq}^m \beta_{k+mq}^+ (1-\delta_{m,5}) \}, \tag{48}$$

otherwise.

By applying the transformation (37), (39) to Eqs (47) and (48) it is easily seen that the left-hand side of Eq. (46) is always zero and, consequently,

$$\omega_{1,k} = 0 \tag{49}$$

for all $k$. Hence, the first-order approximation does not change the conclusions drawn from the zeroth-order result.

To calculate the second-order energy corrections one has first to calculate the first-order operator corrections from the following equation (see [6]):

$$[H_0, \xi^+_{1,k}]+[H_1, \xi^+_{0,k}] = \omega_{0,k}\xi^+_{1,k}. \tag{50}$$

Then, the second-order energy corrections can be calculated from the formula

$$\langle \dots n_k+1, \dots n_l \dots (0)|[H_1, \xi^+_{1,k}]|\dots n_k \dots n_l \dots (0)\rangle = (n_k+1)^{1/2}\omega_{2,k}. \tag{51}$$

The calculations are simple but rather lengthy, and the formulae for $\omega_{2,k}$ quite complicated for wave vectors conforming to conditions (38) and those having $k_x = 0$ or $k_x = \Delta q/2a$. For the remaining wave vectors the formula is rather simple and reads

$$\omega_{2,k} = \sum_m \frac{|d^m_{k+mq}|^2(1-\delta_{m,0})}{\omega_{0,k}-\omega_{0,k+mq}} - \sum_m \frac{|c^*_k+c^*_{k+mq}|^2}{\omega_{0,k}+\omega_{0,k+mq}}. \tag{52}$$

From (34) we see that for small wave vectors $k$ the first sum in Eq. (52) is negative and constitutes the leading term of the second-order energy correction. A rough estimation based on the values (17) shows that in this case

$$|\omega_{2,k}|/\omega_{0,k} > 1, \tag{53}$$

which means that there is a subset of wave vectors $k$ for which the second-order spin-wave energies $\omega_{0,k}+\omega_{2,k}$ are negative. Hence, in the second-order approximation the simple domain-structure configuration (24) appears to be inadmissible as spin-deviation reference state. This, however, does not mean that higher-order approximation would not reverse the conclusion, apart from the fact that inequality (53) makes the reliability of our perturbation procedure questionable. For one thing, it is impossible to prove its convergence, *i.e.*, to show whether or not

$$|\sum_n \omega_{n,k}| < \infty. \tag{54}$$

On the other hand, despite the inequality (53) it is possible that

$$\sum \omega_{n,k} \geqslant 0 \tag{55}$$

in which case the configuration (24) would be strictly admitted as reference state. Finally it is conceivable that a different choice of $H_0$ and/or a different perturbation method could uphold the zeroth-order conclusion in the second perturbation step. In any case with $H_0$ chosen as in Eq. (35) the simple domain-structure configuration (24) is admitted as reference state in the first approximation.

In Part II of this paper we shall demonstrate a refined approach to this problem, by considering a more realistic 180° domain-structure configuration described by inhomogeneous rotations of the spins. We shall show that this configuration leads, up to the *second*-order

of perturbation method, to real and positive spin wave energies for all wave vectors $k$.

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