Vol. A41 (1972) *ACTA PHYSICA POLONICA* No 3

SPIN WAVE THEORY OF THE FIELD-INDUCED MAGNETIC PHASES OF A UNIAXIAL TWO-SUBLATTICE NÉEL-TYPE ANTIFERRIMAGNET. I. LONGITUDINAL MAGNETIC FIELD

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(Received July 12, 1971)

The linear spin-wave theory is applied to a two-sublattice uniaxial antiferrimagnet of Néel type with nearest-neighbour exchange anisotropy and external magnetic field parallel (Part I) or perpendicular (Part II) to the anisotropy axis, and the field-induced magnetic phase transitions are studied. It is shown that in some cases the spin-wave energy spectra are real and positive for fields far beyond the stability region of the system's approximate ground state. This proves that the reality and positiveness of the spin-wave energy spectrum cannot serve as sole criterion for the stability of a magnetic phase. The dependence of the spin-wave energy spectra on the field strength is determined and discussed for each magnetic phase, and the influence of the temperature and the magnetic field on the magnetization is qualitatively examined.

1. Introduction

In a previous paper [1] we examined the zero-temperature magnetic properties of a uniaxial two-sublattice Néel antiferrimagnet in an external magnetic field parallel to the easy axis. In particular, the critical field strengths for the phase transitions were determined and such thermodynamic quantities as the magnetization and susceptibility were studied. Since the exact ground state of an antiferrimagnet is unknown, various mathematical procedures are being used in order to determine it at least approximately (cp. [2]).

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In [1], the approximate ground state of the spin Hamiltonian was determined by minimizing its expectation value in a class of trial states corresponding to complete sublattice spin alignment, the directions of which were chosen as minimization parameters. Strict solutions of the minimization equations were given, and the critical field strengths were obtained from the stability conditions for the approximate ground state of the system. It was shown that there exist three stable magnetic phases, namely, the antiferrimagnetic (A), the canted-spin (CS), and the paramagnetic (P) phase.

A similar zero-temperature analysis for the perpendicular-field case was carried through in [3, 4]. It was shown that, depending on the magnitude of the anisotropy and the relative magnitude of the sublattice magnetic moments, there are in this case two [3] or four [4] stable magnetic phases, namely, the CS and P phases found in [3], and the additional quasi-antiferrimagnetic (Q) and A phases obtained in [4].

The purpose of the present paper is to derive and investigate the spin-wave energy spectra corresponding to the stable magnetic phases obtained in [1] and [4]. The physical properties of the system at low temperatures are examined in the limits of the linear spin-wave theory. Moreover, we re-examine the problem of field-induced phase transitions, considering again the cases when the external magnetic field is parallel (Part I) or perpendicular (Part II) to the anisotropy axis. While assuming nearest-neighbour interactions and uniaxial exchange anisotropy, we do not specify the crystal lattice. The critical fields for the phase transitions are now obtained from the standard reality and positiveness conditions for the spin-wave energy spectra, and are compared with those obtained in [1, 4]. It is shown that in some cases the energy spectra are real and positive for fields far beyond the stability region of the system's approximate ground state. Therefore, the investigation of the spin-wave spectra alone, without paying any attention to the (approximate) ground state of the system (as *e. g.* in [5]), can lead to erroneous results.

Furthermore, we discuss the dependence of the spin-wave energy spectrum in each magnetic phase on the strength of the external magnetic field. Also, we examine qualitatively the influence of the temperature and the magnetic field on the magnetization.

2. *The Hamiltonian*

We assume that the crystal is composed of two crystallographically equivalent sublattices interpenetrating each other, and that the nearest neighbours of an atom belong to the other sublattice. For simplicity, only nearest-neighbour interaction is considered. The Hamiltonian of the spin system is taken to be

$$\mathscr{H} = \sum_{j,a} \mathscr{H}^a_j, \qquad \mathscr{H}^a_j = \tfrac{1}{2} \sum_{m,n} P^a_{m_j n_l} \tilde{S}^a_{m_j} \tilde{S}^a_{n_l} - H^a_j \sum_{m} \tilde{S}^a_{m_j}, \tag{1}$$

where $j = 1, 2$ and $l \equiv j + (-1)^{j+1}$ are sublattice indices; m_j, n_l are sublattice vectors; $a = 1, 2, 3$ is a vector index; $\tilde{S}^a_{m_j}$ denotes the vector components of the spin operator ascribed to the lattice site m_j, and $P^a_{m_j n_l}$ is the interaction tensor between spins at sites m_j, n_l which has the form

$$P^a_{m_j n_l} = J_{m_j n_l} + K^a_{m_j n_l}(\delta_{a1} + \delta_{a3}), \tag{2}$$

where $J_{m_j n_l}$ describes isotropic and $K^a_{m_j n_l}$ anisotropic interactions. As we consider only nearest-neighbour interaction, we put

$$J_{m_j n_l} = \begin{cases} J \text{ for } n = m+\delta \\ 0 \text{ otherwise,} \end{cases} \qquad K^a_{m_j n_l} = \begin{cases} K_a \text{ for } n = m+\delta \\ 0 \text{ otherwise,} \end{cases} \tag{3}$$

where δ denotes the nearest-neighbour vector. We assume $J > 0$, $K_3 > K_1 \geqslant 0$ and

$$(H_j) = \mu_j H(\sin \varphi, 0, \cos \varphi), \; H \geqslant 0, \tag{4}$$

where μ_j is the effective magnetic moment per lattice atom in the j's sublattice; H is the magnitude of the (homogeneous) external magnetic field, and φ is the angle between the field and the x_3-axis (direction of easiest magnetization).

We now introduce the following transformations:

$$\begin{aligned} \tilde{S}^1_{m_j} &= S^1_{m_j} \cos \theta_j - e_j S^3_{m_j} \sin \theta_j, \\ \tilde{S}^2_{m_j} &= S^2_{m_j}, \\ \tilde{S}^3_{m_j} &= e_j S^1_{m_j} \sin \theta_j + S^3_{m_j} \cos \theta_j, \end{aligned} \tag{5}$$

where $e_j \equiv (-1)^j$. This transformation introduces a different co-ordinate system in each sublattice [1, 6].

The next step resides in passing to Bose operators, which we shall do by employing the Holstein-Primakoff mapping [7] in the lowest approximation

$$\begin{aligned} S^1_{m_j} &\to (S_j/2)^{\frac{1}{2}}(a_{m_j} + a^+_{m_j}), \\ S^2_{m_j} &\to ie_j(S_j/2)^{\frac{1}{2}}(a_{m_j} - a^+_{m_j}), \\ S^3_{m_j} &\to e_j(a^+_{m_j} a_{m_j} - S_j). \end{aligned} \tag{6}$$

S_1 and S_2 are the intrinsic spins associated with the atoms of the sublattices 1 and 2, respectively. (We assume a unit system in which $\hbar = 1$.) The operators a_{m_j}, $a^+_{m_j}$ satisfy the standard boson commutation rules

$$[a_{m_j}, a^+_{n_l}] = \delta_{mn}\delta_{jl}, \quad [a_{m_j}, a_{n_l}] = 0. \tag{7}$$

Finally, the Hamiltonian can be expressed in the $\boldsymbol{k}$-representation by means of the Fourier transformation

$$a_{m_j} = N^{-\frac{1}{2}} \sum_k a_{k,j} \exp(ie_j k \cdot m_j), \tag{8a}$$

$$a_{k,j} = N^{-\frac{1}{2}} \sum_{m_j} a_{m_j} \exp(-ie_j k \cdot m_j), \tag{8b}$$

where N is the number of sublattice atoms.

When the transformations (5)–(8) are substituted in the spin Hamiltonian (1) and terms higher than bilinear (with respect to the Bose operators) rejected, the free-spin-wave Hamiltonian can be written in the following form:

$$\mathscr{H} = E_{oM}+\mathscr{H}_1+\mathscr{H}_2, \tag{9}$$

$$E_{oM} = S_1S_2NJ\gamma_o(X\sin\theta_1\sin\theta_2-Z\cos\theta_1\cos\theta_2)-NS_1(H_1^1\sin\theta_1+ +H_1^3\cos\theta_1)-NS_2(H_2^1\sin\theta_2-H_2^3\cos\theta_2), \tag{10}$$

$$\mathscr{H}_1 = \sum_{j,k} A_j(a_{k,j}^{+}+a_{k,j})\delta_{ko}, \tag{11}$$

$$A_j = (NS_j/2)^{1/2}\,[\gamma_oJS_l(Z\sin\theta_j\cos\theta_l+X\cos\theta_j\sin\theta_l)-H_j^1\cos\theta_j- -e_jH_j^3\sin\theta_j], \tag{12}$$

$$\mathscr{H}_2 = \sum_{j,k}\{B_ja_{k,j}^{+}a_{k,j}+\tfrac{1}{2}[C_k(a_{k,j}^{+}a_{-k,l}+a_{-k,l}^{+}a_{k,j})+ +D_k(a_{k,j}^{+}a_{k,l}^{+}+a_{k,j}a_{k,l})]\}, \tag{13}$$

$$B_j = \gamma_oJS_l(Z\cos\theta_j\cos\theta_l-X\sin\theta_j\sin\theta_l)+H_j^1\sin\theta_j-e_jH_j^3\cos\theta_j, \tag{14a}$$

$$C_k = \tfrac{1}{2}(S_1S_2)^{1/2}\,J\gamma_k(X\cos\theta_1\cos\theta_2-Z\sin\theta_1\sin\theta_2-1), \tag{14b}$$

$$D_k = \tfrac{1}{2}(S_1S_2)^{1/2}\,J\gamma_k(X\cos\theta_1\cos\theta_2-Z\sin\theta_1\sin\theta_2+1), \tag{14c}$$

where $X = 1+K_1/J, Z = 1+K_3/J$, and γ_o is the number of nearest neighbours. Here, we have introduced the structure factor

$$\gamma_k = \sum_{\delta}\exp(ik\cdot\delta). \tag{15}$$

If the crystal has inversion symmetry, the structure factor is real: $\gamma_k = \gamma_{-k} = \gamma_k^{*}$. (In the above formulae, the wave vector $\boldsymbol{k}$ is restricted to the first Brillouin zone.)

The linear terms in the Hamiltonian (9) can be eliminated by the transformation [8]

$$a_{k,j} = p_j(k)+c_{k,j} \tag{16}$$

which converts the Hamiltonian into the form

$$\mathscr{H} = E_{oM}+E_{oS}+\mathscr{H}_2, \tag{17}$$

where

$$E_{oS} = \sum_{j,k}[2A_jp_j\delta_{ko}+B_jp_j^2+(C_k+D_k)p_jp_l] = \sum_{j}A_jp_j(0), \tag{18}$$

$$\mathscr{H}_2 = \sum_{j,k}\{B_jc_{k,j}^{+}c_{k,j}+\tfrac{1}{2}[C_k(c_{k,j}^{+}c_{-k,l}+c_{-k,l}^{+}c_{k,j})+ +D_k(c_{k,j}^{+}c_{k,l}^{+}+c_{k,j}c_{k,l})]\}. \tag{19}$$

This leads to the following expressions for the (real) shifting parameters $p_j(k)$:

$$p_j(k) = \frac{A_l(C_k+D_k)-A_jB_l}{B_jB_l-(C_k+D_k)^2}\,\delta_{ko}. \tag{20}$$

The bilinear Hamiltonian (19) can be diagonalized by using a canonical transformation which leads to the result

$$\mathscr{H} = E_o + \sum_{j,k} E_{k,j} b^+_{k,j} b_{k,j}, \tag{21}$$

where the magnon operators $b_{k,j}$ are defined by

$$\begin{aligned} b_{k,1} &= u_{11}c_{k,1}+u_{21}c_{-k,2}-v_{11}c^+_{-k,1}-v_{21}c^+_{k,2}, \\ b_{k,2} &= u_{12}c_{-k,1}+u_{22}c_{k,2}-v_{12}c^+_{k,1}-v_{22}c^+_{-k,2}. \end{aligned} \tag{22a}$$

The expressions for the spin-wave operators (22a) can be inverted to give

$$\begin{aligned} c_{k,1} &= u_{11}b_{k,1}+u_{12}b_{-k,2}+v_{11}b^+_{-k,1}+v_{12}b^+_{k,2}, \\ c_{k,2} &= u_{21}b_{-k,1}+u_{22}b_{k,2}+v_{21}b^+_{k,1}+v_{22}b^+_{-k,2}, \end{aligned} \tag{22b}$$

where we assume the coefficients u and v to be real and even functions of $\boldsymbol{k}$.

The energy spectra $E_{k,j}$ follow in a standard way from the equation system

$$\begin{aligned} (B_1-E_{k,j})u_{1j}+C_ku_{2j}+D_kv_{2j} &= 0 \\ C_ku_{1j}+(B_2-E_{k,j})u_{2j}+D_kv_{1j} &= 0 \\ D_ku_{2j}+(B_1+E_{k,j})v_{1j}+C_kv_{2j} &= 0 \\ D_ku_{1j}+C_kv_{1j}+(B_2+E_{k,j})v_{2j} &= 0 \end{aligned} \tag{23}$$

for the transformation coefficients u and v [6, 9]. Thus, we obtain the non-trivial solutions

$$E^2_{k,j} = \tfrac{1}{2}\{B_1^2+B_2^2-2(D_k^2-C_k^2)\pm[(B_1^2-B_2^2)^2-4D_k^2(B_1-B_2)^2+4C_k^2(B_1+B_2)^2]^{\frac{1}{2}}\}. \tag{24}$$

This formula gives a general expression for the spin-wave energies in a two-sublattice antiferrimagnet of Néel type, in the approximation of non-interacting spin-waves.

The ground state energy E_o consists actually of the three parts: E_{oM}, E_{oS} and E_{oD}, corresponding respectively to the three transformations (6), (16) and (22b). Thus,

$$E_o = E_{oM}+E_{oS}+E_{oD}, \tag{25}$$

where

$$E_{oD} = -\sum_{j,k} E_{k,j}(v^2_{1j}+v^2_{2j}) \tag{26}$$

and E_{oM}, E_{oS} are given by Eqs (9), (18).

In determining the coefficients u and v from Eqs (23) we must take into account the canonical conditions of the Bogolyubov transformation [6, 9] which in our case read

$$\begin{gathered} \sum_j (u^2_{1j}-v^2_{1j}) = 1, \quad \sum_j (u^2_{2j}-v^2_{2j}) = 1, \\ \sum_j (u_{1j}v_{2j}-u_{2j}v_{1j}) = 0, \quad \sum_j (u_{1j}u_{2j}-v_{1j}v_{2j}) = 0, \\ \sum_j (u^2_{j1}-v^2_{j1}) = 1, \quad \sum_j (u^2_{j2}-v^2_{j2}) = 1, \\ \sum_j (u_{j1}u_{j2}-v_{j1}v_{j2}) = 0, \quad \sum_j (u_{j1}v_{j2}-u_{j2}v_{j1}) = 0. \end{gathered} \tag{27}$$

A general solution can be obtained if $\Delta_k \equiv 2B_2C_kD_k \neq 0$ (see Appendix A).

There are two particular cases in which $\Delta_k = 0$, namely,

(*i*) if $K_1 = 0$ in the A and P phase for $\varphi = 0$ (*i. e.*, when the field is parallel to the easy axis; these cases are considered separately in Appendix B), and

(*ii*) if $\gamma_k = 0$ which occurs for certain spin wave lengths $\boldsymbol{k}$ in each phase. The latter case, (*ii*), need not be examined any further, since for $\gamma_k = 0$ we have $C_k = D_k = 0$ and the non-diagonal terms in the Hamiltonian (19) vanish. Thus, the transformation (22b) becomes trivial, as the only non-zero coefficients are $u_{11} = u_{22} = 1$ and the contribution E_{0D} to the system's ground state energy, Eq. (26), is equal to zero. However, one easily verifies that the coefficients $v_{1j}, v_{2j}, u_{21}, u_{12}$ as given by Eqs (A.1)–(A.3) vanish automatically if $\gamma_k = 0$, that $u_{11}, u_{22} \to 1$ as $\gamma_k \to 0$, and that the general formula (24) for the two branches of the spin-wave spectrum reduces to B_1 and B_2 as required. Hence, despite the fact that $\Delta_k = 0$ the formulae (24) and (A.1)–(A.3) remain valid for $\gamma_k = 0$.

3. *Spin-wave energies and critical fields*

Henceforth we consider only the case when the external magnetic field is parallel to the easy axis, *i. e.*, $\varphi = 0$, and defer the case of the transversal magnetic field to the second part of this paper.

In [1] we examined the zero-temperature magnetic properties of a uniaxial two-sublattice Néel antiferrimagnet in an external field parallel to the easy axis. In particular, the stable magnetic phases were obtained and the critical field strengths for the phase transitions were determined from the minimum condition for the approximate ground state energy E_{oM}. It was shown that there exist three stable magnetic phases, namely, the antiferrimagnetic (A), the canted-spin (CS) and the paramagnetic (P) phase.

Here, we want to utilize the results of [1] in specifying the general formula (24) for those magnetic phases, and in determining again the critical field strengths from the standard condition for the reality and positiveness of the energy spectra. Finally, we shall compare these results with those obtained in [1].

As was shown in [1], in the antiferrimagnetic phase (A) one has to distinguish between the antiferrimagnetic configuration A_1 (with the larger spins pointing in the direction of the external magnetic field) and the opposite configuration A_2 (smaller spins pointing in the direction of the field). We consider at first the configuration A_1. Thus, we put $\theta_1 = \theta_2 = 0$. With the notation $\kappa = \mu_2 S_2/\mu_1 S_1$, $S = S_1/S_2$, $y_k = \gamma_k/\gamma_o$, $h = \mu_1 H/JS_2\gamma_o$ and upon specifying the expressions (14a)–(14c) we obtain from the general formula (24) the spin-wave energy spectrum in the A_1 phase:

$$E_{k,j} = \frac{1}{\sqrt{2}} S_2 J\gamma_o\{(Z+h)^2+S^2(Z-\kappa h)^2-2SXy_k^2\pm$$

$$\pm\sqrt{[(Z+h)^2-S^2(Z-\kappa h)^2]^2+4Sy_k^2[X(Z+h)-S(Z-\kappa h)]\,[SX(Z-\kappa h)-(Z+h)]}\}^{\frac{1}{2}}. \quad (28)$$

We adopt the standard limitation for the external field which ensures the lower branch in (28) to be real and non-negative at its minimum points $y_k = \pm 1$. This leads for $h > 0$ to the following restrictions for the external magnetic field:

$$h \leqslant h_l = (2\kappa)^{-1}\{Z(1-\kappa)+[Z^2(1-\kappa)^2+4\kappa w]^{1/2}\} \quad (29a)$$

and

$$h \geqslant h_l' = (2\kappa)^{-1}\{Z(1-\kappa)+[Z^2(1+\kappa)^2-4\kappa]^{1/2}\} > h_l, \tag{29b}$$

where h_l is the upper critical field for the A_1 phase [1] and $w = Z^2 - X^2$. It was shown in [1] that the approximate ground state for the antiferrimagnetic spin configuration A_1 is stable only for fields up to h_l. On the other hand, from (29b) it is seen that the spin-wave energy spectrum is real and positive for fields far beyond the stability region of the system's approximate ground state.

As regards the configuration A_2 we have

$$E_{k,j} = \frac{1}{\sqrt{2}} S_2 J\gamma_o\{(Z-h)^2+S^2(Z+\kappa h)^2-2SX\gamma_k^2 \pm$$

$$\pm \sqrt{[(Z-h)^2-S^2(Z+\kappa h)^2]^2+4S\gamma_k^2[X(Z-h)-S(Z+\kappa h)]\,[SX(Z+\kappa h)-(Z-h)]}\}^{\frac{1}{2}}. \tag{30}$$

The spectra (30) are positive for

$$h \leqslant h_c = (2\kappa)^{-1}\{Z(\kappa-1)+[Z^2(1-\kappa)^2+4\kappa w]^{1/2}\} \tag{31a}$$

as well as for

$$h \geqslant h_c' = (2\kappa)^{-1}\{Z(\kappa-1)+[Z^2(1+\kappa)^2-4\kappa]^{1/2}\} > h_c. \tag{31b}$$

However, similarly as in the phase A_1, the approximate ground state for the configuration A_2 is stable only for fields up to h_c [1].

In the canted-spin phase, the sublattice spins form the angles θ_1 and θ_2 with the external magnetic field, which fixes the spin quantization axes in the two sublattices (or, in other words, the directions of parallel spin alignment in the sublattice reference states). Three methods are being used in determining θ_j, of which the first one resides in the minimization of E_{oM} as defined by Eq. (10), while in the second one the angles θ_j are obtained from the conditions $A_j = 0$ which eliminate the linear terms in the Hamiltonian (11). In [2] both methods were shown to be equivalent (in a limited sense). A third method was introduced in [6] and shown to be equivalent to the above methods in the non-interacting spin-waves approximation. In the latter method one determines θ_j from the condition that the quantization axes be parallel to the sublattice magnetizations.

In [1], the first method was applied, and the solutions which describe the field-dependence of the spin quantization directions (or, equivalently, of the sublattice magnetization directions) in the canted-spin phase were shown to have the form

$$\cos\theta_1 = hw^{-1}(\kappa Z - XR_1),\ \cos\theta_2 = -hw^{-1}(Z-\kappa XR_1^{-1}), \tag{32}$$

where $R_1^2 = (\kappa^2h^2-w)/(h^2-w)$. Accordingly, the quantities (14a)–(14b) assume the form

$$B_1 = S_2JXR_1^{-1}\gamma_0, \quad B_2 = S_1JXR_1\gamma_0,$$

$$C_k = \tfrac{1}{2}(S_1S_2)^{1/2}J\gamma_k[R_1Z-h^2w^{-1}(R_1Z-\kappa X)-1],$$

$$D_k = \tfrac{1}{2}(S_1S_2)^{1/2}J\gamma_k[R_1Z-h^2w^{-1}(R_1Z-\kappa X)+1], \tag{33}$$

and the general formula (24) for the spin-wave energy spectra specifies in the CS phase as follows:

$$E_{k,j} = \frac{1}{\sqrt{2}} S_2 J\gamma_o\{X^2(R_1^{-2}+S^2R_1^2)-2SR_1w^{-1}[h^2XR_1^{-1}\kappa-Z(h^2-w)]y_k^2\pm$$

$$\pm X[X^2(R_1^{-2}-S^2R_1^2)^2+4Sy_k^2\{w^{-1}[h^2XR_1^{-1}\kappa-Z(h^2-w)]-SR_1\}\times$$

$$\times\{SR_1^2w^{-1}[h^2XR_1^{-1}\kappa-Z(h^2-w)]-R_1^{-1}\}]^{\frac{1}{2}}\}^{\frac{1}{2}}. \tag{34}$$

The expressions under the square roots in (34) have minima at the points $y_k = \pm 1$. Thus, the energy spectra are real and positive if

$$h_l < h < h_u = (2\kappa)^{-1}\{Z(1+\kappa)+[Z^2(1+\kappa)^2-4\kappa w]^{1/2}\}. \tag{35}$$

We see that for the canted-spin phase the value of the lower (h_l) as well as the upper (h_u) critical field determined from the spectrum agrees with that obtained in [1] from the stability conditions for the approximate ground state. Therefore, in the CS phase the region of stability of the approximate ground state coincides with that of the positiveness of the energy spectra.

As regards the paramagnetic phase, all the spins become aligned in the direction of the external field, *i.e.*, $\theta_1 = 0$, $\theta_2 = \pi$, and (24) specifies as follows:

$$E_{k,j} = \frac{1}{\sqrt{2}} S_2 J\gamma_o\{(Z-h)^2+S^2(Z-\kappa h)^2+2SXy_k^2\pm$$

$$\pm\sqrt{[(Z-h)^2-S^2(Z-\kappa h)^2]^2+4Sy_k^2[X(Z-h)+S(Z-\kappa h)]\,[SX(Z-\kappa h)+(Z-h)]}\}^{\frac{1}{2}}. \tag{36}$$

The reality condition for the energy spectra (36) leads to the following restrictions for the external magnetic field: $0 < h < h_u'$, $h_u'' < h < h_u'''$ and $h > h_u$, where

$$h_u' = (2\kappa)^{-1}\{Z(1+\kappa)-[Z^2(1-\kappa)^2+4\kappa]^{1/2}\}, \tag{37a}$$

$$h_u'' = (2\kappa)^{-1}\{Z(1+\kappa)-[Z^2(1+\kappa)^2-4\kappa w]^{1/2}\}, \tag{37b}$$

$$h_u''' = (2\kappa)^{-1}\{Z(1+\kappa)+[Z^2(1-\kappa)^2+4\kappa]^{1/2}\} < h_u, \tag{37c}$$

and h_u is given by Eq. (35). However, we know from [1] that the approximate ground state in the paramagnetic phase is stable only for $h > h_u$. Therefore, in this case the examination of the reality of the energy spectra leads again to erroneous results.

4. Spin-wave energies as functions of the external magnetic field

The spin-wave energies in the A_1, A_2, CS and P phase are respectively given by Eqs (28), (30), (34) and (36). Thus, we may analyze the behaviour of these energies in each magnetic phase under the influence of the external magnetic field.

Let us start from the phase A_1 and first consider the simpler case $K_1 = 0$. From (B.6) results that

$$E_{k,1}(y_k = 0) = \gamma_0 JS_1(Z-\kappa h), \; E_{k,2}(y_k = 0) = \gamma_0 JS_2(Z+h), \tag{38a}$$

$$\Delta E_k \equiv E_{k,1} - E_{k,2} = \gamma_0 JS_2[Z(S-1)-(1+\kappa S)h]. \tag{38b}$$

From Eq. (38b) it is seen that the difference between the two branches of the energy spectrum does not depend upon the wave vector $\boldsymbol{k}$. Moreover, it is seen that ΔE_k is positive for $0 < h < h_d$ and negative for $h_d < h < h_l$, where

$$h_d = \frac{Z(S-1)}{1+\kappa S} \,. \tag{39}$$

Hence and from Eq. (38a) it is seen that the first spectrum branch lowers with increasing field while the second one rises. For $h = h_d$ both branches coincide (two-fold degeneracy of the spectrum).

Consider now the same phase A_1 in the more general case $K_1 > 0$. From (28) one easily obtains

$$E_{k,1}(y_k = 0) = \begin{cases} \gamma_o JS_1(Z-\kappa h) & \text{for} \quad 0 < h < h_d \\ \gamma_o JS_2(Z+h) & \text{for} \quad h_d < h < h_l, \end{cases} \tag{40a}$$

$$E_{k,2}(y_k = 0) = \begin{cases} \gamma_o JS_2(Z+h) & \text{for} \quad 0 < h < h_d \\ \gamma_o JS_1(Z-\kappa h) & \text{for} \quad h_d < h < h_l, \end{cases} \tag{40b}$$

$$\Delta E_k = \gamma_o JS_2\{(Z+h)^2+S^2(Z-\kappa h)^2-2SXy_k^2-2S[(Z+h)^2(Z-\kappa h)^2- -(X^2+1)(Z+h)(Z-\kappa h)y_k^2+X^2y_k^4]^{1/2}\}^{1/2} \geqslant 0. \tag{40c}$$

Hence, in this case the difference between the two branches depends upon the wave vector $\boldsymbol{k}$ and is always positive which means that the first branch lies always above the second one, except when $y_k = 0$ and $h = h_d$ as $\Delta E_k = 0$ (two-fold degeneracy point). Thus, the first spectrum branch lowers when the field increases from 0 to h_d, and rises again upon further increasing the field. For the second branch the opposite holds. The behaviour of the spin-wave energy spectrum in the phase A_1 for the cases $K_1 = 0$ and $K_1 > 0$ is illustrated in Figs 1 and 2, respectively.

The lack of correspondence in the shifting of the energy branches under the influence of the field in the case $K_1 = 0$ (Fig. 1) and for the limiting case $K_1 \to 0$ (Fig. 2) is merely apparent and results from the rather arbitrary labelling of the particles ascribed to the particular branch. Indeed, the spectra (B.6) may also be written in the form

$$E'_{k,1} = \tfrac{1}{2}S_2 J\gamma_o\{|(1+\kappa S)h-Z(S-1)|+\sqrt{[Z(1+S)+(1-\kappa S)h]^2-4Sy_k^2}\},$$

$$E'_{k,2} = \tfrac{1}{2}S_2 J\gamma_o\{-|(1+\kappa S)h-Z(S-1)|+\sqrt{[Z(1+S)+(1-\kappa S)h]^2-4Sy_k^2}\}, \tag{41}$$

which preserves in the whole interval $0 < h < h_l$ a uniform division of the particles in "heavy" ($E'_{k,1}$) and "light" ($E'_{k,2}$) ones and ensures the correspondence between the case $K_1 = 0$ from Fig. 1 and the limiting case $K_1 \to 0$ from Fig. 2 — according to Eq. (40c). Of course, this is a purely descriptive matter and has no influence whatever on the results.

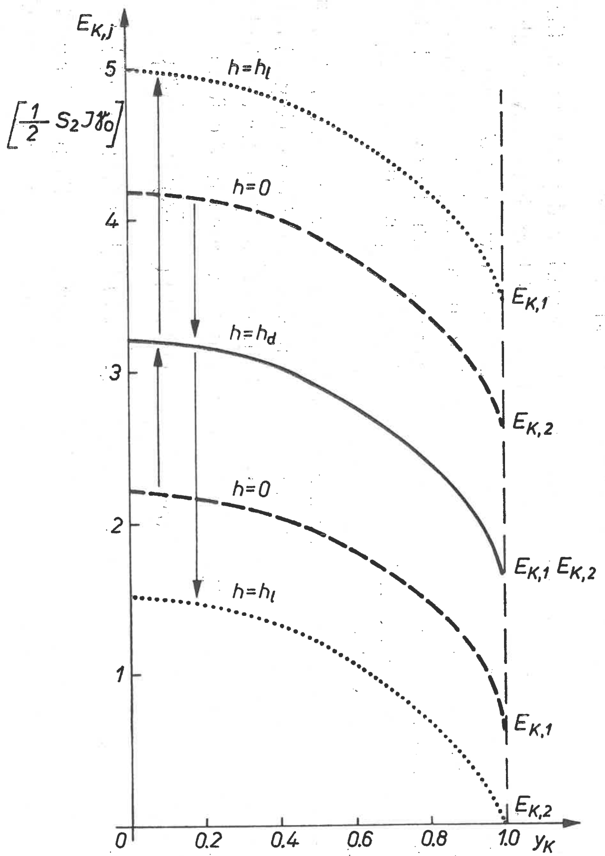

Fig. 1. Free-spin-wave energy spectra in the antiferrimagnetic phase A_1 for the case $K_1 = 0$

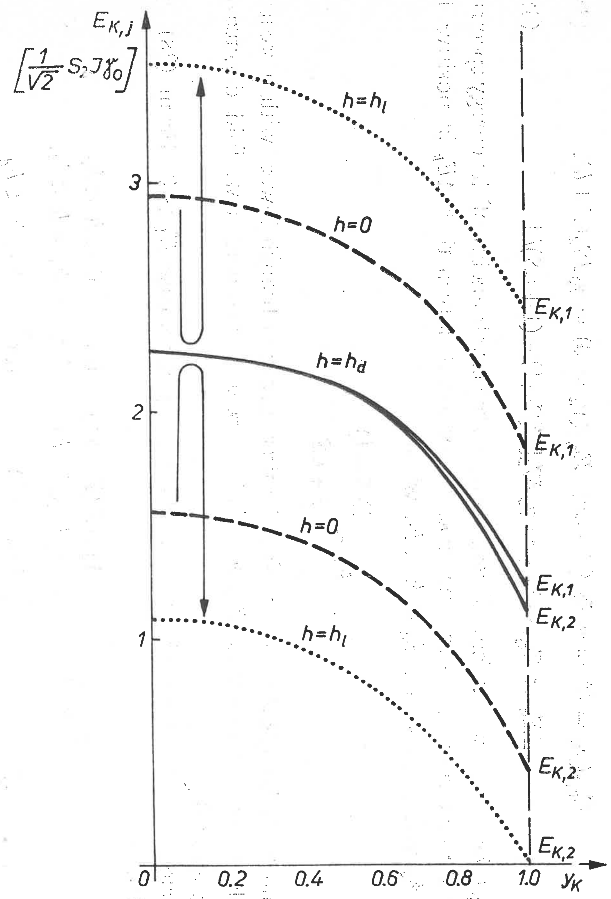

Fig. 2. Free-spin-wave energy spectra in the antiferrimagnetic phase A_1 for the case $K_1 > 0$

In the phase A_2 we have from Eq. (30)

$$E_{k,1}(y_k = 0) = \gamma_o JS_1(Z+\kappa h),\ E_{k,2}(y_k = 0) = \gamma_o JS_2(Z-h), \tag{42a}$$

$$\Delta E_k = \gamma_o JS_2\{(Z-h)^2+S^2(Z+\kappa h)^2-2SXy_k^2-2S[(Z-h)^2(Z+\kappa h)^2- -(X^2+1)(Z-h)(Z+\kappa h)y_k^2+X^2y_k^4]^{1/2}\}^{1/2} > 0. \tag{42b}$$

It is seen that with increasing field the first spectrum branch rises while the second one lowers. The only difference between the case $K_1 = 0$ and $K_1 > 0$ is that in the first case

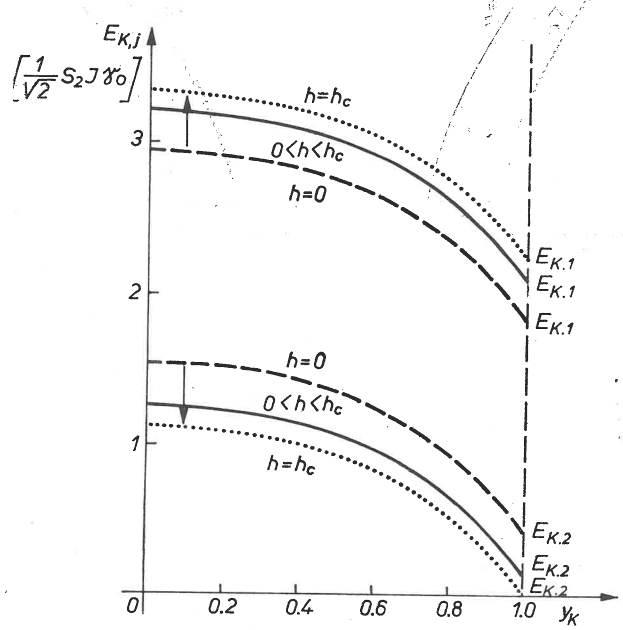

Fig. 3. Free-spin-wave energy spectra in the antiferrimagnetic phase A_2

ΔE_k does not depend on the wave vector $\boldsymbol{k}$. The spin-wave energy spectra in the phase A_2 are given in Fig. 3.

As regards the CS phase, we see from (34) that

$$E_{k,1}(y_k = 0) = \gamma_o JS_2XR_1^{-1},\ E_{k,2}(y_k = 0) = \gamma_o JS_1XR_1, \tag{43}$$

i.e., at the point $y_k = 0$ the first spectrum branch lowers with increasing field while the second one rises. A numerical analysis shows that in the CS phase the spin-wave energy spectrum changes under the influence of the field as shown in Fig. 4.

In the paramagnetic phase we have from (36)

$$E_{k,1}(y_k = 0) = \gamma_o JS_2(h-Z),\quad E_{k,2}(y_k = 0) = \gamma_o JS_1(\kappa h-Z), \tag{44a}$$

$$\Delta E_k = \gamma_o JS_2\{(Z-h)^2+S^2(Z-\kappa h)^2+2SXy_k^2-2S[(Z-h)^2(Z-\kappa h)^2- -(X^2+1)(Z-h)(Z-\kappa h)y_k^2+X^2y_k^4]^{\frac{1}{2}}\}^{\frac{1}{2}} > 0. \tag{44b}$$

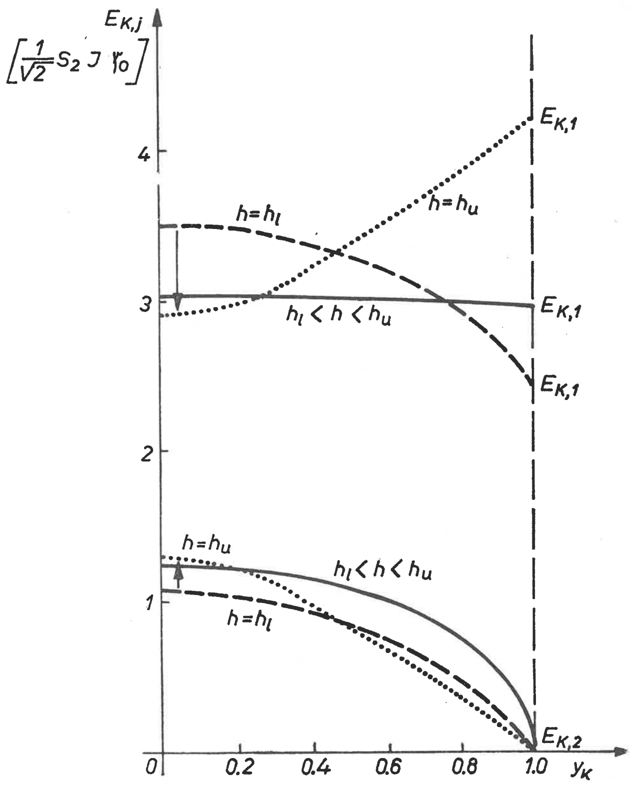

Fig. 4. Free-spin-wave energy spectra in the canted-spin phase CS

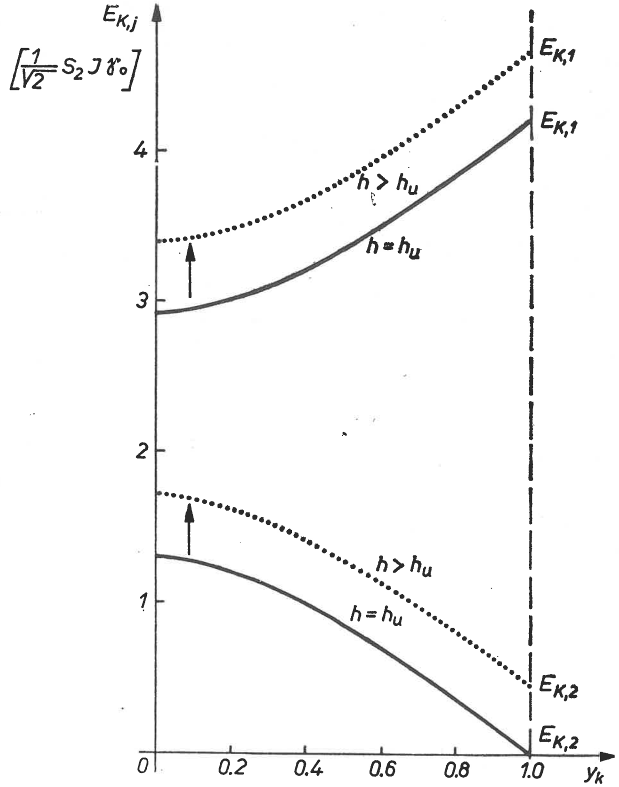

Fig. 5. Free-spin-wave energy spectra in the paramagnetic phase P

Thus, in this case both branches rise with increasing field, as illustrated in Fig. 5.

It can be seen from Eqs (28), (34) and (36) that the corresponding energy branches of the neighbouring phases are continuous at the transition points, *i.e.*,

$$E^{A_1}_{k,j}(h_l) = E^{CS}_{k,j}(h_l), \quad E^{CS}_{k,j}(h_u) = E^{P}_{k,j}(h_u), \tag{45a}$$

except for the transition $A_2 \to A_1$ for which we have

$$E^{A_2}_{k,j}(h_c) \neq E^{A_1}_{k,j}(h_c). \tag{45b}$$

5. The magnetization as function of the external field and temperature

In [1] the approximate ground state was determined by minimizing the expectation value of the spin Hamiltonian (1) in a class of trial states representing sublattice saturation states. This corresponds actually to minimizing E_{oM} and assuming complete sublattice spin alignment. It was shown that the energy of the approximate ground state E_{oM} is a continuous function of the external field, except for h_c if the phase transition $A_2 \to A_1$ occurs. The same is true for the approximate ground state energy E_o given by Eq. (25). (Note that E_o is the exact ground state energy of the Hamiltonian (9) which is an approximation of the Hamiltonian (1).) From Eqs (25) and (26) it is seen that $E_o < E_{oM}$ (as E_{oS} vanishes in all three phases), but the continuity of the energy at the critical points h_l, h_u is preserved. One easily verifies that there is no complete sublattice spin alignment in the direction θ_j in the ground state (25).

The components of the sublattice magnetization vectors $M_{j,a}$ are defined as follows:

$$M_{j,a} = \mu_j \sum_m \langle \tilde{S}^a_{mj} \rangle, \quad (j = 1, 2;\ a = 1, 2, 3). \tag{46}$$

Noting the fact that $p_j(k) = 0$ for all phases we obtain from (5), (6), (8a), (16) and (22b)

$$M_{j,1} \equiv M_{j\perp} \to \mu_j N S_j \sin\theta_j - \mu_j \sin\theta_j \sum_k \sum_{r=1}^{2} [(u^2_{jr} + v^2_{jr})\bar{n}_{kr} + v^2_{jr}],$$

$$M_{j,3} \equiv M_{j\parallel} \to -e_j \mu_j N S_j \cos\theta_j + e_j \mu_j \cos\theta_j \sum_k \sum_{r=1}^{2} [(u^2_{jr} + v^2_{jr})\bar{n}_{kr} + v^2_{jr}],$$

$$M_{j,2} = 0 \tag{47}$$

where

$$\bar{n}_{kr} \equiv \langle b^+_{k,r} b_{k,r} \rangle = [\exp(\beta E_{k,r}) - 1]^{-1}, \quad \beta = (k_B T)^{-1} \tag{48}$$

is the average number of r-type particles (spin waves) at temperature T.

Let us write the transversal $M_{j\perp}$ and longitudinal $M_{j\parallel}$ components of the sublattice magnetizations in the form

$$M_{j\parallel} \equiv M_{oMj\parallel} + M_{oDj\parallel} + M_{Tj\parallel} \equiv M_{oj\parallel} + M_{Tj\parallel},$$

$$M_{j\perp} \equiv M_{oMj\perp} + M_{oDj\perp} + M_{Tj\perp} \equiv M_{oj\perp} + M_{Tj\perp}, \tag{49}$$

where, according to (47),

$$M_{oMj||} = -e_j\mu_j NS_j \cos\theta_j,$$

$$M_{oDj||} = e_j\mu_j \cos\theta_j \sum_k \sum_{r=1}^{2} v_{jr}^2,$$

$$M_{Tj||} = e_j\mu_j \cos\theta_j \sum_k \sum_{r=1}^{2} (u_{jr}^2+v_{jr}^2)\bar{n}_{kr}, \tag{50a}$$

$$M_{oMj\perp} = \mu_j NS_j \sin\theta_j,$$

$$M_{oDj\perp} = -\mu_j \sin\theta_j \sum_k \sum_{r=1}^{2} v_{jr}^2,$$

$$M_{Tj\perp} = -\mu_j \sin\theta_j \sum_k \sum_{r=1}^{2} (u_{jr}^2+v_{jr}^2)\bar{n}_{kr}. \tag{50b}$$

The lower indices "o" and "T" denote respectively the parts independent and dependent on the temperature, and the index "M" denotes the value of the corresponding quantity in the approximate ground state E_{oM} (which corresponds to complete sublattice spin

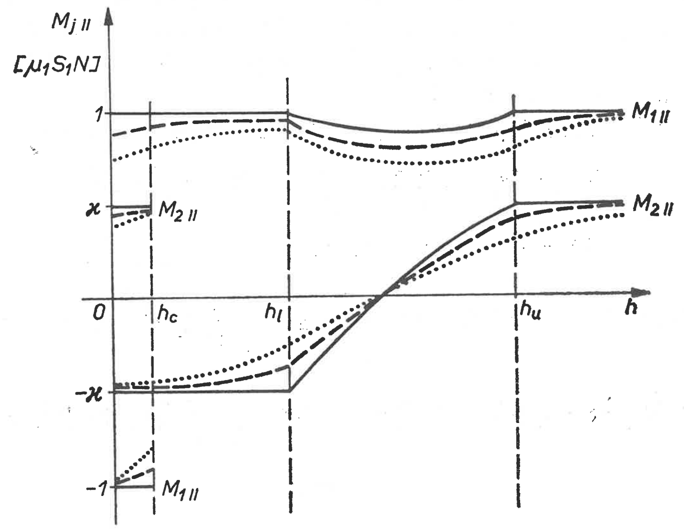

Fig. 6. Schematic curves for the longitudinal components of the sublattice magnetizations as functions of the external magnetic field. The solid line corresponds to the approximate ground state E_{oM}, the dashed line — to the approximate ground state E_o, and the dotted line — to non-zero temperature

alignment). The index "D" denotes the contribution originating from the diagonalizing transformation (22b). Upon specifying θ_j and the coefficients u and v in the formulae (47)–(50), we obtain the expressions for the sublattice magnetizations in each phase.

From the explicit formulae for the magnetizations in all the phases it is easy to see that the magnetizations are continuous at the transitions $A_1 \leftrightarrow$ CS and CS $\leftrightarrow$ P, except for the transition $A_2 \rightarrow A_1$. To illustrate the influence of the temperature and magnetic field on the sublattice magnetizations, schematic curves for $T = 0°K$ and $T > 0°K$ are given in Fig. 6 for the longitudinal components of the sublattice magnetizations.

APPENDIX A

Following the procedure applied in [6] one easily proves that for $\Delta_k \neq 0$ the coefficients u, v of the transformation (22a, b) have the form

$$v_{11}^2 = \frac{\Delta_k^2}{\Delta}(\Delta_{12}\Delta_{22}-\Delta_{32}\Delta_k), \tag{A.1}$$

$$v_{12}^2 = \frac{\Delta_k^2}{\Delta}(\Delta_{31}\Delta_k-\Delta_{21}\Delta_{11}), \tag{A.2}$$

$$u_{1j} = \frac{\Delta_{1j}}{\Delta_k}v_{1j}, \quad u_{2j} = \frac{\Delta_{2j}}{\Delta_k}v_{1j}, \quad v_{2j} = \frac{\Delta_{3j}}{\Delta_k}v_{1j}, \tag{A.3}$$

where

$$\Delta = (\Delta_{11}^2-\Delta_k^2)(\Delta_{12}\Delta_{22}-\Delta_{32}\Delta_k)-(\Delta_{12}^2-\Delta_k^2)(\Delta_{11}\Delta_{21}-\Delta_{31}\Delta_k), \tag{A.4}$$

$$\Delta_{1j} = -\tfrac{1}{2}(B_1+E_{k,j})(B_1^2-B_2^2-e_jG_k)+D_k^2(B_1-B_2)-C_k^2(B_1+B_2), \tag{A.5}$$

$$\Delta_{2j} = -C_k\{(B_1+B_2)E_{k,j}+\tfrac{1}{2}[(B_1+B_2)^2-e_jG_k]\}, \tag{A.6}$$

$$\Delta_{3j} = D_k\{(B_1-B_2)E_{k,j}+\tfrac{1}{2}[(B_1-B_2)^2-e_jG_k]\}, \tag{A.7}$$

$$G_k = [(B_1^2-B_2^2)^2-4D_k^2(B_1-B_2)^2+4C_k^2(B_1+B_2)^2]^{\frac{1}{2}}. \tag{A.8}$$

A straightforward calculation shows that these coefficients fulfil the canonical conditions (27).

APPENDIX B

There are two special cases in which due to $\Delta_k = 0$ the general formulae (A.1)–(A.3) are no longer valid, namely, when $K_1 = 0$ and $\varphi = 0$ in the A and P phases. Let us consider briefly these cases.

First note that $K_1 = \varphi = 0$ implies $C_k = 0$ in the antiferrimagnetic phase, in which case the set of equations (23) splits in two subsets:

$$\begin{aligned} (B_1-E_{k,j})u_{1j}+D_kv_{2j} &= 0, \\ D_ku_{1j}+(B_2+E_{k,j})v_{2j} &= 0; \end{aligned} \tag{B.1}$$

$$\begin{aligned} (B_2-E_{k,j})u_{2j}+D_kv_{1j} &= 0, \\ D_ku_{2j}+(B_1+E_{k,j})v_{1j} &= 0. \end{aligned} \tag{B.2}$$

The corresponding secular equations lead to the energy spectra

$$E_{k,j}^{(B.1)} = \tfrac{1}{2}\{B_1-B_2\pm[(B_1+B_2)^2-4D_k^2]^{\frac{1}{2}}\},$$
$$E_{k,j}^{(B.2)} = \tfrac{1}{2}\{B_2-B_1\pm[(B_1+B_2)^2-4D_k^2]^{\frac{1}{2}}\}. \quad \text{(B.3)}$$

Similarly as in [6], one easily verifies that a careful analysis of the solutions and the canonical conditions leads unequivocally to the two different energy branches, namely,

$$E_{k,1} = \tfrac{1}{2}\{B_2-B_1+[(B_1+B_2)^2-4D_k^2]^{1/2}\},$$
$$E_{k,2} = \tfrac{1}{2}\{B_1-B_2+[(B_1+B_2)^2-4D_k^2]^{1/2}\}. \quad \text{(B.4)}$$

The coefficients u and v have the form

$$u_{11}^2 = u_{22}^2 = \frac{D_k^2}{D_k^2-(B_1-E_{k,1})^2},$$

$$v_{12}^2 = v_{21}^2 = \frac{(B_1-E_{k,1})^2}{D_k^2-(B_1-E_{k,1})^2},$$

$$u_{12} = u_{21} = v_{11} = v_{22} = 0. \quad \text{(B.5)}$$

As for the A_1 phase, we have

$$E_{k,1} = \tfrac{1}{2} S_2 J\gamma_o\{(1+\kappa S)h - Z(S-1) + \sqrt{[Z(S+1)+(1-\kappa S)h]^2-4S\gamma_k^2}\},$$
$$E_{k,2} = \tfrac{1}{2} S_2 J\gamma_o\{Z(S-1)-(1+\kappa S)h + \sqrt{[Z(S+1)+(1-\kappa S)h]^2-4S\gamma_k^2}\}. \quad \text{(B.6)}$$

In the paramagnetic phase we have $D_k = 0$ (as $K_1 = \varphi = 0$). In this case the equation system (23) splits again in two subsets,

$$(B_1-E_{k,j})u_{1j}+C_k u_{2j} = 0,$$
$$C_k u_{1j}+(B_2-E_{k,j})u_{2j} = 0; \quad \text{(B.7)}$$
$$(B_1+E_{k,j})v_{1j}+C_k v_{2j} = 0,$$
$$C_k v_{1j}+(B_2+E_{k,j})v_{2j} = 0, \quad \text{(B.8)}$$

of which only one has non-trivial solutions if $E_{k,j} \neq 0$. Choosing the set (B.7) and taking into account the canonical conditions (27) one easily obtains

$$E_{k,1} = \tfrac{1}{2} S_2 J\gamma_o\{-Z(S+1)+(1+\kappa S)h+\sqrt{[Z(S+1)+(1-\kappa S)h]^2+4S\gamma_k^2}\},$$
$$E_{k,2} = \tfrac{1}{2} S_2 J\gamma_o\{-Z(S+1)+(1+\kappa S)h-\sqrt{[Z(S+1)+(1-\kappa S)h]^2+4S\gamma_k^2}\}, \quad \text{(B.9)}$$

$$u_{11}^2 = u_{22}^2 = \frac{C_k^2}{C_k^2+(B_1-E_{k,1})^2},$$

$$u_{12}^2 = u_{21}^2 = \frac{C_k^2}{C_k^2+(B_1-E_{k,2})^2},$$

$$v_{11} = v_{12} = v_{21} = v_{22} = 0. \quad \text{(B.10)}$$

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