THE APPROXIMATE GROUND STATE OF TWO-SUBLATTICE UNIAXIAL FERRI- AND ANTIFERRIMAGNETS WITH TRANSVERSAL MAGNETIC FIELD

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The approximate ground state (spin wave reference state) of the two-sublattice uniaxial ferri- and antiferrimagnet with an external magnetic field perpendicular to the easy axis is determined. In the nearest-neighbours approximation, strict solutions for the field-dependence of the direction of spin-alignment in the sublattice reference states are obtained and discussed, and the critical fields for the transition to the paramagnetic state are determined. The ferri-para phase transition is shown to be of second order.

1. *Introduction*

When employing the spin wave formalism to the Heisenberg model of ferri- or antiferrimagnetism there is usually the problem if choosing a suitable reference state (spin wave vacuum) if the spin wave interactions are to be sufficiently small to justify the standard long-wavelength low-temperature approximations [1–4]. A typical example is the case when the (homogeneous) external magnetic field is not parallel to a direction of easiest magnetization, in which case the reference state depends on the field strength and direction. In [1–4], two different methods of determining the reference state for magnetic crystals have been studied quite generally: the method A, which is preferably used in the theory of ferromagnetism [5], aims at determining the approximate ground state of the spin Hamiltonian, by minimizing its expectation value in a class of trial states generated by spatial rotations from the state of complete spin alignment (saturation state); and the method B which resides in eliminating the terms linear with respect to the spin wave creation and annihilation

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operators appearing in the transformed Hamiltonian (see, *e.g.*, [6]). In [1–4], the methods A and B were shown to be equivalent in a limited sense.

Our purpose is to determine the reference state of the two-sublattice uniaxial ferri- or antiferrimagnet with an external magnetic field which is perpendicular to the easy axis, by applying the method A with the following restrictions:

a) the class of reference states is confined to "homogeneous" states corresponding to complete spin alignment in the sublattice (sublattice saturation state);

b) the ferri- or antiferrimagnet is assumed to be of Néel type, and the (exchange) interaction is restricted to nearest neighbours only;

c) uniaxial nearest-neighbours exchange anisotropy is assumed;

d) intra-atomic interaction (crystal field anisotropy) is excluded.

Within these limitations, strict solutions for the field-dependence of the direction of spin-alignment in the sublattice reference states are obtained and discussed, and the critical fields for the transition to the paramagnetic state are determined.

2. *Approximate ground state energy*

We consider a spin Hamiltonian of the form:

$$\mathscr{H} = -\varepsilon|J| \sum_{\langle f,g\rangle} (X\tilde{S}_f^x\tilde{S}_g^x+\tilde{S}_f^y\tilde{S}_g^y+Z\tilde{S}_f^z\tilde{S}_g^z)-\mu_1 H \sum_f \tilde{S}_f^x-\mu_2 H \sum_g \tilde{S}_g^x \tag{1}$$

where $X = 1+|K_x/J|$, $Z = 1+|K_z/J|$, $(Z > X)$. The subscripts f, g denote respectively the sites of the first and second sublattice (each of them having N sites); $\langle f, g\rangle$ denotes the summation over nearest neighbours; K_x, K_z are the exchange anisotropy constants in the x and z directions, and J is the nearest-neighbours exchange integral between atoms belonging to different sublattices; μ_1 and μ_2 are the effective magnetic moments per lattice atom in the first and second sublattice, respectively; H denotes the homogeneous external magnetic field which is parallel to the coordinate axis x (perpendicular to the anisotropy axis z); $\varepsilon = -1$ corresponds to the antiferrimagnetic, and $\varepsilon = 1$ to the ferrimagnetic case.

Similarly as in [1], we perform the following rotations of the spins in the plane $x0z$

$$\begin{aligned}
\tilde{S}_f^x &= S_f^x \cos\theta_1 + S_f^z \sin\theta_1 & \tilde{S}_g^x &= S_g^x \cos\theta_2 + \varepsilon S_g^z \sin\theta_2 \\
\tilde{S}_f^y &= S_f^y & \tilde{S}_g^y &= S_g^y \\
\tilde{S}_f^z &= -S_f^x \sin\theta_1 + S_f^z \cos\theta_1 & \tilde{S}_g^z &= -\varepsilon S_g^x \sin\theta_2 + S_g^z \cos\theta_2
\end{aligned} \tag{2}$$

and define the sublattice saturation states $|0\rangle_f$ and $|0\rangle_g$ (homogeneous spin-deviation reference states; *cp.* [4]) as follows:

$$S_f^z|0\rangle = S_1|0\rangle, \quad S_f^+|0\rangle = 0$$

$$S_g^z|0\rangle = \varepsilon S_2|0\rangle, \quad S_g^\varepsilon|0\rangle = 0$$

where

$$S_{f,g}^+ = S_{f,g}^x + iS_{f,g}^y, \; |0\rangle = |0\rangle_f \cdot |0\rangle_g, \; {}_f\langle 0|0\rangle_f = {}_g\langle 0|0\rangle_g = 1 \tag{3}$$

(see Figs 1 and 2). Subsequently, the approximate ground state is determined by minimizing the mean value of the transformed Hamiltonian[1] in the state $|0\rangle$ with respect to the parameters θ_1 and θ_2,

$$\min \langle 0|\mathscr{H}|0\rangle \equiv \min E_0(\theta_1, \theta_2) \tag{4}$$

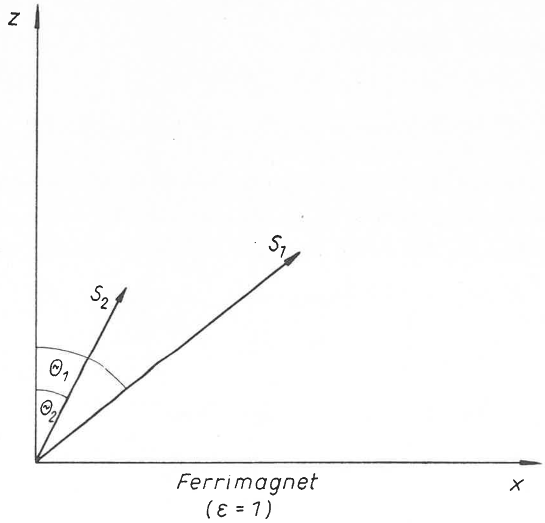


Fig. 1

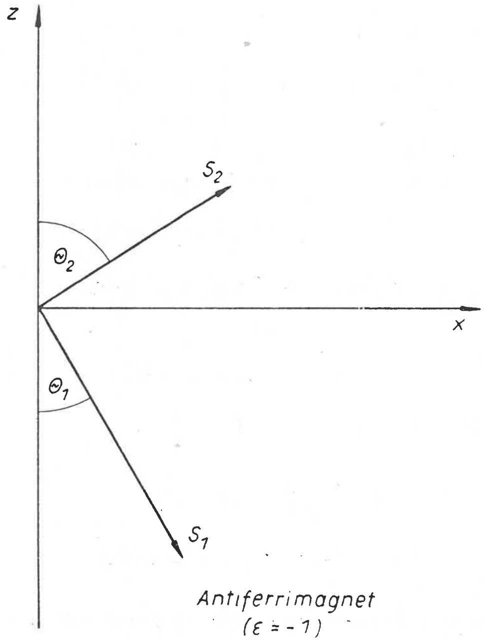


Fig. 2

[1] This is equivalent to minimizing the mean value of the Hamiltonian (1) in the class of reference states $|0(\theta_1, \theta_2)\rangle = U^+|0\rangle$ where the unitary transformation U corresponds to the transformation (2) (*cp.* [1]).

By taking into account Eqs (1)–(4) we obtain for E_0 the expression

$$E_0(\theta_1, \theta_2) = -\varepsilon d[X \sin\theta_1 \sin\theta_2 + \varepsilon Z \cos\theta_1 \cos\theta_2 + \varepsilon h \sin\theta_1 + \varepsilon\varkappa h \sin\theta_2] \qquad (5)$$

where

$$h = \mu_1 H/\gamma_0 S_2|J|, \qquad \varkappa = \mu_2 S_2/\mu_1 S_1, \qquad d = NS_1S_2\gamma_0|J|,$$

γ_0 — the number of nearest neighbours.

3. *Minimization of E_0 and critical fields*

The approximate ground state energy corresponding to the minimum of (5) we obtain by studying the necessary and sufficient conditions for the existence of a minimum of a function of two variables θ_1, θ_2 which depends on the parameters $X, Z, h, \varkappa, \varepsilon$

$$\frac{\partial E_0}{\partial\theta_1} = 0, \qquad \frac{\partial E_0}{\partial\theta_2} = 0; \qquad \Delta = \frac{\partial^2 E_0}{\partial\theta_1^2}\frac{\partial^2 E_0}{\partial\theta_2^2} - \left(\frac{\partial^2 E_0}{\partial\theta_1\partial\theta_2}\right)^2 > 0, \qquad \frac{\partial^2 E_0}{\partial\theta_2^2} > 0. \qquad (6)$$

The necessary conditions can be written in the form:

$$\cos\theta_1 = \eta|R|\cos\theta_2 \qquad (7)$$

and

$$\cos\theta_2[w\sin\theta_1 - h(\eta Z|R| + \varepsilon\varkappa X)] = 0 \qquad (8)$$

or

$$\cos\theta_1\left[w\sin\theta_2 - h\left(\frac{\eta\varkappa Z}{|R|} + \varepsilon X\right)\right] = 0 \qquad (9)$$

where $R^2 = (\varkappa^2h^2 + w)/(h^2 + w)$, $w = Z^2 - X^2$, $\eta = \pm 1$.

From (7) and (8) or (9) we obtain the following solutions:

$$\sin\theta_1 = hw^{-1}(\eta Z|R| + \varepsilon\varkappa X) \qquad (10)$$

$$\sin\theta_2 = hw^{-1}(\eta\varkappa Z|R|^{-1} + \varepsilon X) \qquad (11)$$

or

$$\cos\theta_1 = \cos\theta_2 = 0. \qquad (12)$$

For the solution (10), (11) we have:

$$\frac{\partial^2 E_0}{\partial\theta_2^2} = \eta d Z|R| \qquad (13)$$

$$\Delta = \mathrm{d}^2R^2(h^2 + w)(1 - \sin^2\theta_2). \qquad (14)$$

It can easily be seen that in this case the necessary conditions (6) for the minimum of E_0 are satisfied for all values of the angles θ_1 and θ_2 (except for $|\sin\theta_1| = |\sin\theta_2| = 1$) if $\eta = +1$. In this case, Eq. (7) and the initial condition $\theta_1 = \theta_2 = 0$ for $h = 0$ permit without loss of generality to restrict the angles θ_1 and θ_2 to the interval $\langle 0, \pi/2\rangle$ for the ferrimagnet

($\varepsilon = +1$), and to the interval $\langle -\pi/2, \pi/2 \rangle$ in the antiferrimagneric case ($\varepsilon = -1$). On the other hand, the reality condition for the solution (10), (11) leads to the following limitation for the magnetic field:

$$0 \leqslant h < h_c = \frac{1}{2\varkappa}\left[-\varepsilon X(\varkappa+1)+\sqrt{X^2(\varkappa+1)^2+4\varkappa w}\right]. \tag{15}$$

It can easily be verified that in the interval (15) the solution (12) does not satisfy the sufficient minimum conditions (6). For values of the magnetic field above h_c, the situation is reversed, as only the solution (12) is real and satisfies the sufficient conditions (6). In this case, for the ferrimagnet ($\varepsilon = 1$) only the values $\theta_1 = \theta_2 = \pi/2$ for the angles are admitted, while for the antiferrimagnet ($\varepsilon = -1$) there are two solutions: $\theta_1 = \theta_2 = \pi/2$ and $\theta_1 = \pi/2$, $\theta_2 = -\pi/2$ (or *v.v.*); in the latter case, it can be easily shown that the solution $\theta_1 = \theta_2 = \pi/2$ represents the absolute minimum. So in both cases ($\varepsilon = \pm 1$) we have for $h > h_c$ the solution $\theta_1 = \theta_2 = \pi/2$, for which

$$\Delta = d^2[\varkappa h^2+\varepsilon X(\varkappa+1)h-w] > 0, \tag{16}$$

$$\frac{\partial^2 E_0}{\partial \theta_2^2} = d(\varepsilon X+\varkappa h) > 0. \tag{17}$$

Hence, the solution (10), (11) describes the "scissors phase" (S) in which the spins in the two sublattices form with the easy direction (z-axis) the angles θ_1 and θ_2, and the solutions (12) correspond to the so-called paramagnetic phase (P) in which the spins are aligned along the direction of the magnetic field ($\theta_1 = \theta_2 = \pi/2$). The transition between the first and second phase takes place for $h = h_c$.

4. *Discussion of results*

In order to determine what kind of phase transition takes place at the critical point h_c, we examine the energy E_0, the magnetization M and the susceptibility χ of the system in the approximate ground state (3).

Upon inserting (10), (11) or (12) into (5) we obtain for the phase S and P, respectively,

$$E_0^S = -d[Z|R|+h^2w^{-1}(Z|R|+\varepsilon\varkappa X)], \tag{18}$$

$$E_0^P = -d[\varepsilon X+(\varkappa+1)h]. \tag{19}$$

With the components of the sublattice magnetizations in the state (3) defined as follows:

$$M_{\mathrm{I}}^z = \mu_1 \sum_f \langle 0|\tilde{S}_f^z|0\rangle, \qquad M_{\mathrm{I}}^x = \mu_1 \sum_f \langle 0|\tilde{S}_f^x|0\rangle \tag{20}$$

$$M_{\mathrm{II}}^z = \mu_2 \sum_g \langle 0|\tilde{S}_g^z|0\rangle, \qquad M_{\mathrm{II}}^x = \mu_2 \sum_g \langle 0|\tilde{S}_g^x|0\rangle. \tag{21}$$

the total magnetization M has the form

$$M = |\boldsymbol{M}| = \sqrt{M_z^2+M_x^2} = \sqrt{(M_{\mathrm{I}}^z+M_{\mathrm{II}}^z)^2+(M_{\mathrm{I}}^x+M_{\mathrm{II}}^x)^2} \equiv$$
$$\equiv \sqrt{M_\perp^2+M_\parallel^2} = \mu_1 S_1 N \sqrt{(\cos\theta_1+\varepsilon\varkappa\cos\theta_2)^2+(\sin\theta_1+\varkappa\sin\theta_2)^2}, \tag{22}$$

where $M_\perp$ and $M_{||}$ denote the components of the magnetization in the direction perpendicular and parallel to the external magnetic field (transversal and longitudinal magnetization), respectively. For the corresponding components of the sublattice magnetizations we have

$$M_\perp^{\mathrm{I}} = \mu_1 S_1 N \cos\theta_1 \qquad M_\perp^{\mathrm{II}} = \mu_2 S_2 N \varepsilon \cos\theta_2, \tag{23}$$

$$M_{||}^{\mathrm{I}} = \mu_1 S_1 N \sin\theta_1, \qquad M_{||}^{\mathrm{II}} = \mu_2 S_2 N \sin\theta_2. \tag{24}$$

For the transition point between the phase S and P we obtain

$$E_0^S|_{h=hc} = E_0^P|_{h=hc} = -\frac{d}{2\varkappa}\left[-\varepsilon X(\varkappa^2+1)+(X^2(\varkappa+1)^2+4\varkappa w)^{1/2}(\varkappa+1)\right] \tag{25}$$

$$M^S|_{h=hc} = M_{||}^S|_{h=hc} = N(\mu_2 S_1+\mu_2 S_2) = M_{||}^P|_{h=hc} = M^P|_{h=hc}. \tag{26}$$

From (25) and (26) we see that for $h = h_c$ the (ground state) energy of the system and its first derivative with respect to the magnetic field (*i.e.*, $M_{||}$) are continuous. However, one easily proves that the susceptibility has a jump (phase transition of second order), as

$$\chi^S|_{h=hc} = \chi_{||}^S|_{h=hc} = \frac{\varkappa+1}{h_c}\left\{1-\left(1-\frac{X^2}{Z^2}\right)\left(\frac{\varkappa-1}{\varkappa+1}\right)^2\right\}, \tag{27}$$

$$\chi^P|_{h=hc} = \chi_{||}^P|_{h=hc} = 0. \tag{28}$$

To illustrate the influence of the external magnetic field on the approximate ground state energy and on the magnetization, schematic numerical curves for the case $\varkappa = 1/2$, $X=1(\tilde{K}_x = 0)$, $Z = 1.1(\tilde{K}_z = 0.1)$ are given in Figs 3–8. The dependence of the angles θ_1, θ_2 on the (reduced) external field h is shown in Figs 3 and 4, respectively for the ferri- and antiferrimagnetic case. The curves in Fig. 5. demonstrate the field-dependence of the ground state energy, the critical fields for the $S \leftrightarrow P$ transition being denoted by h_c^f and h_c^{af} for the

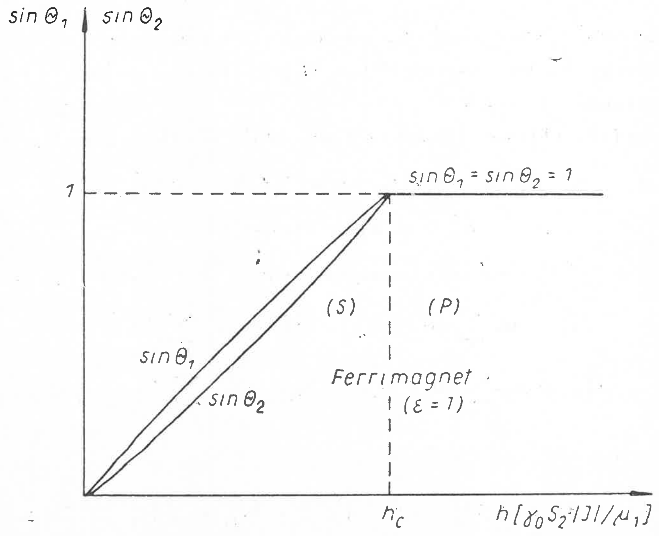


Fig. 3

ferri- and antiferrimagnetic case, respectively. The (reduced) absolute value of the sublattice magnetizations m^{I}, m^{II} as well as their longitudinal ($m^{I}_{||}$, $m^{II}_{||}$) and transversal ($m^{I}_{\perp}$, $m^{II}_{\perp}$) components are for the antiferrimagnetic case plotted in Fig. 6, while the respective quantities corresponding to the total magnetization are given in Fig. 7. The absolute value and the longitudinal and transversal components of the total magnetizations for the ferrimagnetic

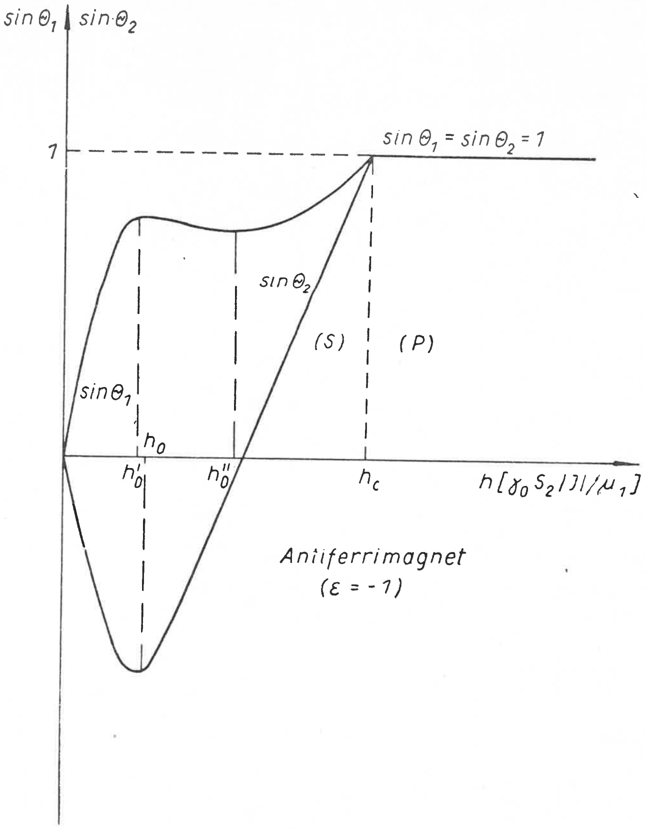


Fig. 4

case are presented in Fig. 8. In the latter case, the field-dependence of the respective quantities corresponding to the sublattice magnetizations is qualitatively the same as in Fig. 8, except that the absolute values in the S-phase are field-independent and equal to 1, much like in the antiferrimagnetic case (*cp.* Fig. 6).

As is seen from Fig. 3, our exact solutions clearly show that the customary approximation $\theta_1 \approx \theta_2$ (see e.g. [7]) in the "scissors phase" S of the ferrimagnet is justified for small $(0 \leqslant h \ll h_c)$ as well as large $(0 \ll h \leqslant h_c)$ fields, as in these cases the spins in the two sublattices are nearly parallel to each other, the deviation from parallelity being largest for intermediate field strengths and depending on the magnitude of the anisotropy and the effective spin ratio $\varkappa = \mu_2 S_2/\mu_1 S_1$. Since $\theta_1 > \theta_2$ for $0 < h < h_c$, it is the larger spin S_1

which is more inclined toward the external magnetic field. The same is true for the anti-ferrimagnetic case (Fig. 4), though in this case the smaller spins S_2 rotate at first toward the negative field direction, and only upon exceeding a certain field strength h_0 swing gradually back to the positive field direction. As for the larger spins S_1, there is also clearly visible a swing-back motion for intermediate field strengths $h_0' < h < h_0''$. One easily verifies that no phase transitions take place at h_0, h_0', h_0''. This somewhat strange motion of the spins leads to the peculiar field-dependence of the longitudinal and transversal sublattice

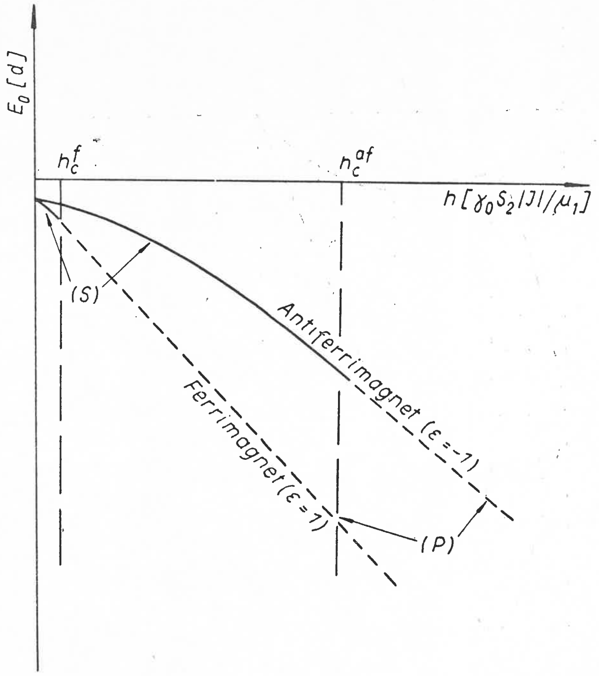


Fig. 5

magnetizations shown in Fig. 6. It is evidently seen from Fig. 7 that the customary assumption $\vec{m}\|\vec{h}$ in the molecular-field approach (*cp.* [8]; see also the case of the S-phase considered in [9, 10] is not justified unless $h > h_c/2$.

It is interesting to determine the field h_1 for which the smaller spins S_2 pass again through the anisotropy direction (*i.e.*, for $h \neq 0$; *cp.* Figs 4 and 6):

$$h_1^2 = \varkappa^{-2}(X^2 - \varkappa^2 Z^2). \tag{29}$$

One easily concludes from this formula that there is no swing-back motion of the spins if $Z/X \leqslant \varkappa$, in which case the S-phase in antiferrimagnets resembles the spin-flop phase of antiferromagnets. Finally, we may point out that for $\varkappa = 1$ our results correspond respectively to those for ferro- and antiferromagnets (*cp.* [5, 11]), with the critical fields

$$\begin{aligned} h_c &= Z - X \quad \text{for} \quad \varepsilon = 1 \\ h_c &= Z + X \quad \text{for} \quad \varepsilon = -1. \end{aligned} \tag{30}$$

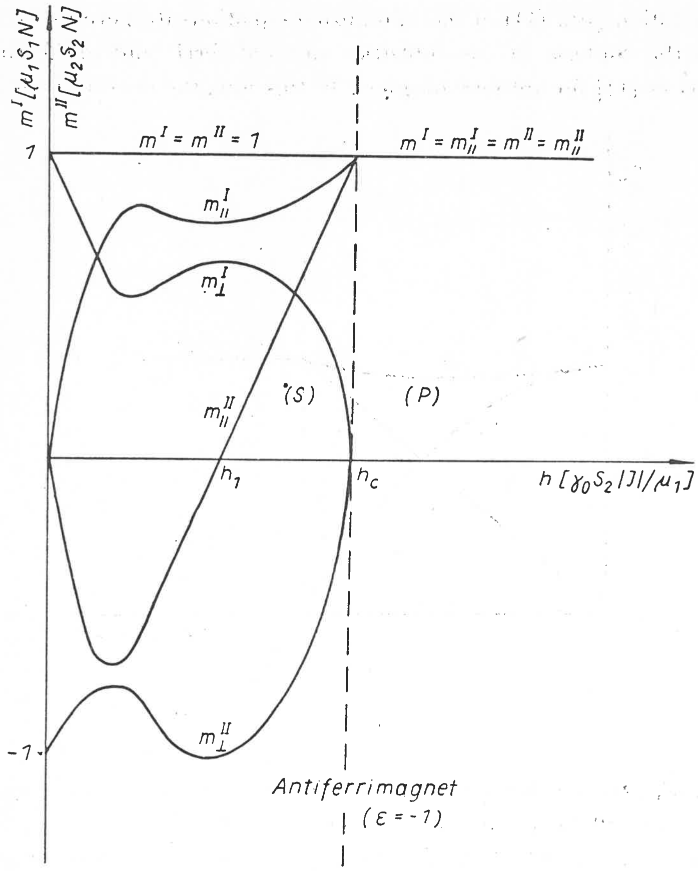

$m^I[\mu_1 S_1 N]$
$m^{II}[\mu_2 S_2 N]$
1
-1
$m^I = m^{II} = 1$
$m^I = m^I_{\parallel} = m^{II} = m^{II}_{\parallel}$
$m^I_{\parallel}$
$m^I_{\perp}$
$m^{II}_{\parallel}$
$m^{II}_{\perp}$
(S)
(P)
h_1
h_c
$h\,[\gamma_0 S_2 |J|/\mu_1]$
Antiferrimagnet
$(\varepsilon = -1)$

Fig. 6

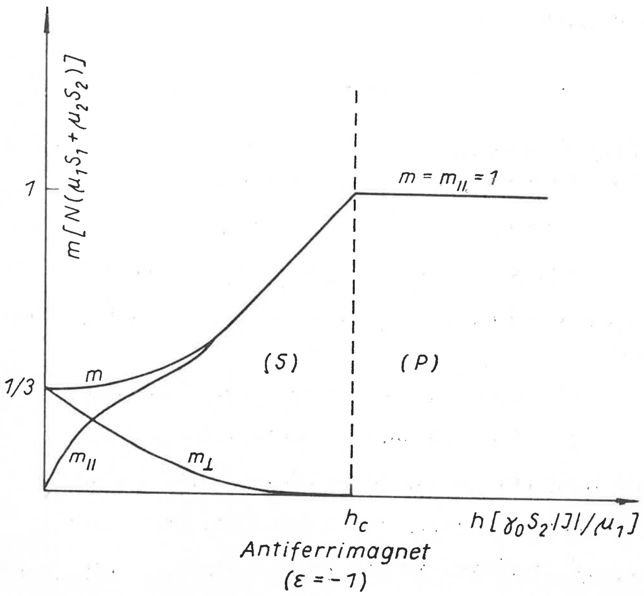
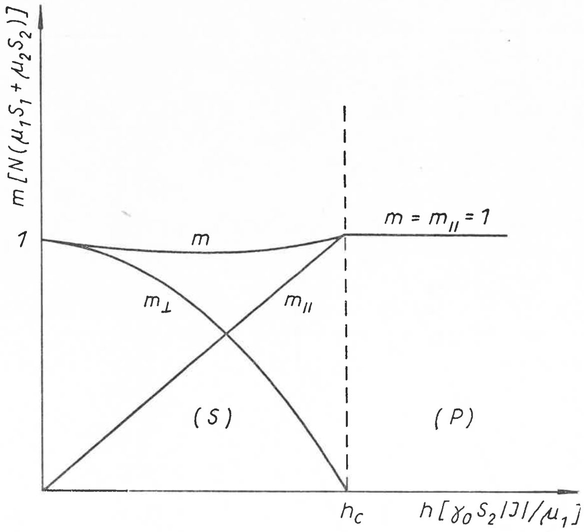

$m[N(\mu_1 S_1 + \mu_2 S_2)]$
1
1/3
$m = m_{\parallel} = 1$
m
$m_{\parallel}$
$m_{\perp}$
(S)
(P)
h_c
$h\,[\gamma_0 S_2 |J|/\mu_1]$
Antiferrimagnet
$(\varepsilon = -1)$

Fig. 7

The exact solution (10), (11) of the minimizing equations (6) permits to apply the spin wave theory to the S-phase in two-sublattice uniaxial ferri- and antiferrimagnets, with the same rigor as in [11] for antiferromagnets. In this way, the critical fields obtained here



Ferrimagnet
($\varepsilon = 1$)
Fig. 8

can be checked with those following from the spin wave energy spectra. Such investigations are under way and will be published in a separate paper [12].

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APPENDIX

In order to get Eqs (7–9) from (6) we write down the necessary conditions for the existence of a minimum of a function E_0 in the following form:

$$\varepsilon Z \sin\theta_1 \cos\theta_2 - X \cos\theta_1 \sin\theta_2 = \varepsilon h \cos\theta_1 \tag{A}$$

$$\varepsilon Z \cos\theta_1 \sin\theta_2 - X \sin\theta_1 \cos\theta_2 = \varepsilon\varkappa h \cos\theta_2 \tag{B}$$

Multiplying Eqs (A) and (B) by εZ and X respectively, and *v.v.*, and subtracting (A) from (B) we obtain

$$(Z^2 - X^2)\sin\theta_1 \cos\theta_2 = hZ\cos\theta_1 + \varepsilon X\varkappa h \cos\theta_2 \tag{C}$$

$$(Z^2 - X^2)\cos\theta_1 \sin\theta_2 = h\varkappa Z\cos\theta_2 + \varepsilon hX \cos\theta_1 \tag{D}$$

Upon squaring and subtracting again the above equations one gets

$$(Z^2 - X^2 + h^2)\cos^2\theta_1 = (Z^2 - X^2 + h^2\varkappa^2)\cos^2\theta_2 \tag{F}$$

With the notation $\varkappa^2 h^2 + Z^2 - X^2/h^2 + Z^2 - X^2 \equiv R^2$ we easily arrive at Eq. (7).

REFERENCES

[1] H. Pfeiffer, W. J. Ziętek, *Acta Phys. Polon.*, **A38**, 176 (1970).
[2] W. J. Ziętek, *Acta Phys. Polon.*, **31** 799 (1969); **36**, 145 (1969).
[3] A. Pękalski, W. J. Ziętek, *Acta Phys. Polon.*, **31**, 131 (1967).
[4] L. Biegała, *Acta Phys. Polon.*, **A38**, 773 (1970).
[5] S. V. Tyablikov, *Methods of the Quatum Theory of Magnetism*, Moscow 1964 (in Russian).
[6] J. Feder, E. Pytte, *Phys. Rev.*, **168**, 640 (1968).
[7] E. H. Yakovlev, *FMM*, **6**, 976 (1958).
[8] A. A. Gusev, *Sov. Phys.-Crystallog.*, **4**, 655 (1960).
[9] A. E. Clark, E. Callen, *J. Appl. Phys.*, **39**, 5972 (1968).
[10] S. V. Tyablikov, *FMM*, **3**, 3 (1956).
[11] A. Pękalski, W. J. Ziętek, *Acta Phys. Polon.*, **A39** 327 (1971).
[12] L. Biegała, S. Krzemiński, *Acta Phys. Polon.*, — to be published.

Note added in proof: Only recently came to our attention the short note by M. J. Besnus *et al.* (*J. Appl. Phys.*, **39**, 903 (1968)), in which similar qualitative results for the field-induced phase transition in a two-sublattice uniaxial antiferrimagnet have been obtained, in a phenomenological way, and in the weak and strong anisotropy approximations.