MOLECULAR-FIELD THEORY OF PHASE TRANSITIONS IN UNIAXIAL FERROMAGNETS WITH EXTERNAL MAGNETIC FIELD

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By applying Bogolubov's variational method in the molecular field approximation (MFA) to the Heisenberg ferromagnet with anisotropic exchange, the influence of an external magnetic field perpendicular or parallel to the easy axis (or plane) on the transition from the ferro- to the paramagnetic phase is examined. The respective magnetic states are proved to satisfy the conditions for the absolute minimum of the magnetic free energy with respect to the magnitude and the direction of the magnetization. The transition is shown to occur in the transverse field and to be destroyed by the parallel one. For the case of the transverse field, some of the critical indices corresponding to the field-dependent critical region are also derived.

1. Introduction

It is well known, from experimental as well as theoretical investigations, that the second-order phase transition in a ferromagnet is destroyed upon application of an external magnetic field (see, *e.g.*, [1, 2]). Recent experiments on ferromagnetic EuS [3], however, strongly indicate the possible existence of such a phase transition in the presence of an external field[1]. The specific magnetic properties of recently obtained ferromagnetic europium compounds (see, *e.g.*, [6—8,2]) make them particulary suitable a material for studying this rather subtle effect.

The theoretical explanation of this effect should apparently be looked for in the nature of the interactions to be taken into account. Following [8], it seems that particular attention should be paid to the long-range dipolar interactions, which causes the demagnetizing effect in a finite sample of a ferromagnetic material. As an example illustrating the qualitative considerations of [8] there has been presented the thin toroid of an isotropic ferromagnetic material, which in the presence of an external magnetic field perpendicular to its plane is capable of two magnetic states, the transition temperature being field-dependent. By applying the MFA method, in the case of spin one-half the transition between these two toroid's

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[1] See also the references given in [4, 5].

states has been shown to be of second-order [4]. This result has been generalized in [9] for the case of arbitrary spin; in this paper, however, it has also been stressed that the toroid sample of an isotropic ferromagnetic material considered in [8, 4] represents, in fact, a model of an anisotropic (uniaxial) ferromagnet, as the demagnetizing energy of the sample has been accounted for (shape anisotropy). This hypothesis has been fully confirmed in [10], by applying the MFA method to a uniaxial Heisenberg ferromagnet with an external magnetic field perpendicular to the easy axis. Thus it has been shown that a uniaxial anisotropy (in the case of the transverse field) can be responsible for that kind of effect. Moreover, in view of the results given in [10] the origin of the anisotropy seems to be here immaterial. Similar results as in [10] have been obtained in [5], however, in a rather phenomenological way.

The purpose of the present paper is to extend the considerations of [10], which are formulated here by using Bogolubov's variational principle (see, *e.g.*, [11, 12]), presenting not only the case of the external magnetic field with the components[2] $H_\perp \neq 0$, $H_{||} = 0$ [10], but also, for comparison, the cases $H_{||} \neq 0$, $H_\perp = 0$; $H_{||} = 0$, $H_\perp = 0$, and analogous results for the easy plane. The results presented here are apparently correct, in a qualitative sense, in the whole temperature region. In particular, these results as well as their (limited) correspondence to those obtained by well-founed low-temperature methods (*i.e.*, far below the ordinary Curie point; see, *e.g.*, [15–17]) indicate that the second-order phase transition should also occur at low temperatures, but under influence of considerable strong fields perpendicular to the easy axis.

2. *Formulation of the problem*[3]

We start with the Heisenberg Hamiltonian for a uniaxial magnetic crystal, having the form[4]

$$\mathscr{H} = -\frac{1}{2}\sum_{r,r'} A^{rr'} s_i^r s_i^{r'} - \frac{1}{2}\sum_{r,r'} K^{rr'} s_3^r s_3^{r'} - \mu H_i \sum_{r} s_i^r \tag{1}$$

where s_i^r are the components of the spin operator ascribed to the lattice site r, $A^{rr'}$ and K^{rr} are the isotropic and anisotropic parts of the exchange coupling, respectively, and H_i the components of the external magnetic field. If $A^{rr'} > 0$, $K^{rr'} > 0$ there is an easy axis in the direction x_3, whereas for $A^{rr'} > 0$, $-A^{rr'} < K^{rr'} < 0$ there is an easy plane, $x_1 0 x_2$. According to Bogolubov's variational principle (see, *e.g.*, [11, 12]) one can obtain the approximate free energy of a system by minimizing the so-called model free energy

$$F(\mathscr{H}) = F(\mathscr{H}_0) + \langle \mathscr{H} - \mathscr{H}_0 \rangle_0 \tag{2}$$

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[2] For clarity, we shall use occasionally the notation $X_{||}$, $X_\perp$ for the components of a vector $\boldsymbol{X}$ which are the projections of this vector on the easy axis (or plane) and the axis perpendicular to the easy directions, respectively.

[3] Our formulation is based on that proposed in [12].

[4] We use the summation convention for repeated lower indices.

where

$$\mathcal{H}_0 = \mathcal{H} - (\mathcal{H} - \mathcal{H}_0), \quad \langle \ldots \rangle_0 = \frac{\mathrm{Tr}(\ldots e^{-\beta \mathcal{H}_0})}{\mathrm{Tr} e^{-\beta \mathcal{H}_0}}$$

$$F(\mathcal{H}_0) = -\frac{1}{\beta} \ln \mathrm{Tr}\, e^{-\beta \mathcal{H}_0}, \quad \beta = \frac{1}{\mathrm{kT}}. \tag{3}$$

The Hamiltonian $\mathcal{H}$ describes the system, and the minimization of the model free energy should be carried out with respect to parameters introduced in $\mathcal{H}_0$. In our case (1) to obtain MFA we choose

$$\mathcal{H}_0 = -M_i \sum_r s_i^r \tag{4}$$

and thus the minimization parameters are the components M_i of the vector $\boldsymbol{M}$ or its length M and the direction cosines n_i, *i.e.*,

$$M = \sqrt{M_i M_i}, \quad n_i = M_i/M, \tag{5}$$

$$n_i n_i = 1. \tag{6}$$

We apply the above thermodynamical approach to the case of a uniformly magnetized crystal, it means we assume

$$\sigma_i^r \equiv \langle s_i^r \rangle_0 / s = \sigma_i \tag{7}$$

where s denotes the maximum eigenvalue of the spin operator at any of the N lattice sites of the crystal. To simplify further calculations we introduce the quantities

$$\varphi \equiv \frac{F(\mathcal{H})}{Ns^2 \Omega}, \quad \tau \equiv \frac{3\mathrm{kT}}{s(s+1)\Omega}, \quad h_i \equiv \frac{\mu H_i}{s\Omega},$$

$$m_i \equiv \frac{M_i}{s\Omega}, \quad \alpha \equiv \frac{\sum_l z_l A_l}{\Omega}, \quad \varkappa \equiv \frac{\sum_l z_l K_l}{\Omega}, \tag{8}$$

$$P_{ij} \equiv \alpha \delta_{ij} + \varkappa \delta_{i3} \delta_{j3}$$

where z_l, A_l and K_l denote respectively the number of the neighbours and the values of the (isotropic) exchange integral and the anisotropy constant for the l-th coordination sphere, and

$$\Omega \equiv \sum_l z_l (A_l + K_l). \tag{9}$$

Taking into account (3)–(8) one can derive the model free energy (2) for the Hamiltonian (1) (see, *e.g.*, [12]):

$$\varphi = -\frac{s+1}{3s} \tau \ln \frac{\mathrm{sh} \frac{2s+1}{2s} \tilde{B}_s(\sigma)}{\mathrm{sh} \frac{1}{2s} \tilde{B}_s(\sigma)} + \frac{s+1}{3s} \tau \sigma \tilde{B}_s(\sigma) - h_i n_i \sigma - \frac{1}{2} P_{ij} n_i n_j \sigma^2 \tag{10}$$

where in

$$\sigma = n_i\sigma_i = B_s(\beta sM) \tag{11}$$

the Brillouin function is defined

$$B_s(x) \equiv \frac{2s+1}{2s} \operatorname{cth} \frac{2s+1}{2s} x - \frac{1}{2s} \operatorname{cth} \frac{1}{2s} x, \tag{12}$$

and $\tilde{B}_s(x)$ in (10) denotes the inverse function of $B_s(x)$. Because of the condition (6), we ntroduce the Lagrange factor λ and minimize the function

$$\Phi = \varphi + \frac{1}{2} \lambda n_i n_i \tag{13}$$

with respect[5] to σ and n_i. Then the necessary conditions for an extremum of Φ to exist lead to the equations

$$\frac{s+1}{3s} \tau\tilde{B}_s(\sigma) - h_i n_i - P_{ij} n_i n_j \sigma = 0 \tag{14}$$

$$\lambda n_i - h_i\sigma - P_{ij} n_j \sigma^2 = 0. \tag{15}$$

As is well known, the vector of magnetization and that of the effective field ($M_i/\mu = m_i s\Omega/\mu$ in our case, see (4) and (8)) should be parallel if the molecular-field approach is to be consistent. We shall show that our calculation meets this condition automatically. Indeed, from (5), (6), (8), (11) and (14) we have

$$m_i = \frac{s+1}{3s} \tau\tilde{B}_s(\sigma) n_i = h_i + P_{ij}\sigma_j \tag{16}$$

and, upon eliminating λ from (15), one obtains

$$m_i\sigma_j = m_j\sigma_i \tag{17}$$

i.e., the parallelity and self-consistency condition. The equality (17), which is obtained here as a *result* of the minimization of the free energy, has been *assumed* in [4, 9, 10], in order to achieve the quoted results in the case of the transverse field.

Since the interactions described by the Hamiltonian (1) are isotropic in the plane $x_1 0 x_2$, without a loss of generality we may put, *e.g.*,

$$n_1 \equiv 0. \tag{18}$$

Thus (14) and (15) along with the condition (6) provide a system of four (nonlinear) equations for n_2, n_3, σ and λ. If $\Phi(\sigma, n_i)$ (given by (13)) is to have a minimum, it is sufficient that the form

$$d^2\Phi = \left(\frac{s+1}{3s} \tau\tilde{B}_s'(\sigma) - P_{ij} n_i n_j\right) d\sigma^2 - 2(h_i + P_{ij} n_j \sigma) d\sigma n_i + (\lambda\delta_{ij} - P_{ij}\sigma^2) dn_i dn_j \tag{19}$$

[5] According to (10) and (11) one may express φ by means of M or σ. Of course, the minimization of Φ with respect to M or σ is equivalent, and leads to the same results. For mathematical simplicity we choose σ (along with n_i) as minimization parameter.

$\left(\text{where } \tilde{B}_s'(x) \equiv \frac{d}{dx} \tilde{B}(x)\right)$ be positive for a solution of (6), (14),(15). Because of (6), the differentials dn_i are interdependent, as $n_i dn_i = 0$. For examining the sign, $d^2\Phi$ shoul-d be transformed to independent differentials. Due to (18), upon introducing the transfori mation

$$\tilde{n}_k = R_{ki}\, n_i$$

$$(R_{ki}) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos\vartheta & \sin\vartheta \\ 0 & -\sin\vartheta & \cos\vartheta \end{pmatrix} \tag{20}$$

and choosing the angle ϑ in such a way that

$$\tilde{n}_3 = 1, \tag{21}$$

one obtains $d^2\Phi$ as a quadratic form of the independent differentials $d\sigma, d\tilde{n}_2$

$$d^2\Phi = \left(\frac{s+1}{3s}\tau\tilde{B}_s'(\sigma) - \tilde{P}_{33}\right) d\sigma^2 - 2(\tilde{h}_2 + \tilde{P}_{23}\sigma) d\sigma d\tilde{n}_2 + (\lambda - \sigma^2\tilde{P}_{22}) d\tilde{n}_2^2$$
$$\equiv \Phi_{\sigma\sigma} d\sigma^2 + 2\Phi_{\sigma\xi} d\sigma d\xi + \Phi_{\xi\xi} d\xi^2 \tag{22}$$

where

$$\tilde{h}_k = R_{ki} h_i,\ \tilde{P}_{kl} = R_{ki} R_{lj} P_{ij}. \tag{23}$$

Hence, the sufficient conditions for a minimum of the free energy (10) to exist are

$$\Phi_{\sigma\sigma} > 0,\ \Delta = \Phi_{\sigma\sigma}\Phi_{\xi\xi} - \Phi_{\sigma\xi}^2 > 0. \tag{24}$$

3. Field perpendicular to the easy axis (or plane; $H_\perp \neq 0$, $H_{||} = 0$)

In accordance with the results of [4, 5, 8–10], a second-order phase transition occurs in the magnetic system described by the Hamiltonian (1) in the case of the transverse external field. Being dependent on the field strength and temperature, the direction of the magnetization lies between that of the easy axis and the external field in the first (ferromagnetic) phase, and is parallel to the field in the second (paramagnetic) one (see the interpretation of the toroid model given in [9]). Accordingly, in this case the system of equations (6), (14), (15) has the solutions (note (18))

$$n_\perp = \frac{h_\perp}{|\varkappa|\sigma},\quad n_{||} = \pm\sqrt{1 - n_\perp^2},\quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma\sigma, \tag{25}$$
$$\lambda = \gamma\sigma^2,$$

$$n_\perp = 1,\quad n_{||} = 0,\quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma'\sigma + h_\perp, \tag{26}$$
$$\lambda = \sigma(\gamma'\sigma + h_\perp),$$

$$n_\perp = -1,\quad n_{||} = 0,\quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma'\sigma - h_\perp, \tag{27}$$
$$\lambda = \sigma(\gamma'\sigma - h_\perp),$$

where

$$\gamma \equiv \begin{cases} 1 \\ \alpha = 1+|\varkappa| \end{cases} \gamma' \equiv \begin{cases} \alpha = 1-\varkappa \\ 1 \end{cases} \text{for} \begin{array}{l} 0 < \varkappa < 1 \\ \varkappa < 0, (\Omega > 0) \end{array} \tag{28}$$

(compare (1) and (8)). Let us choose the coordinate system in such a way that $h_\perp > 0$. Then the solution (25) is real for

$$|\varkappa|\sigma > h_\perp, \tag{29}$$

and (27) requires

$$\gamma'\sigma > h_\perp \tag{30}$$

if negative temperatures are to be avoided. The quantities (24) read

$$\Phi_{\sigma\sigma} = \gamma\Psi(\sigma) + \frac{h_\perp^2}{|\varkappa|\sigma^2}$$

$$\Delta = \Phi_{\sigma\sigma}\left(|\varkappa|\sigma^2 - \frac{h_\perp^2}{|\varkappa|}\right) \tag{31}$$

for the solution (25),

$$\Phi_{\sigma\sigma} = \gamma\Psi(\sigma) + \frac{h_\perp \tilde{B}_s'(\sigma)}{\tilde{B}_s(\sigma)}$$

$$\Delta = \sigma\Phi_{\sigma\sigma}(h_\perp - |\varkappa|\sigma) \tag{32}$$

for (26), and

$$\Phi_{\sigma\sigma} = \gamma'\Psi(\sigma) - \frac{h_\perp \tilde{B}_s'(\sigma)}{\tilde{B}_s(\sigma)}$$

$$\Delta = -\sigma\Phi_{\sigma\sigma}(h_\perp + |\varkappa|\sigma) \tag{33}$$

for (27), where in

$$\Psi(\sigma) \equiv \frac{\sigma\tilde{B}_s'(\sigma)}{\tilde{B}(\sigma)} - 1 > 0 \tag{34}$$

the temperature τ has been eliminated (compare (22)) by utilizing the respective relations in Eqs (25)–(27). The proof of the inequality (34) is given in the Appendix. With the exception of (33), $\Phi_{\sigma\sigma}$ is positive in the whole interval $0 < \sigma < 1$, as it immediately follows from (34) and the properties of the function $\tilde{B}_s(x)$ (see Appendix). Accordingly, Δ is positive in the case (31) for (29), and in the case (32) for

$$|\varkappa|\sigma < h_\perp. \tag{35}$$

As regards (33), it is evident from the expression for Δ that $\Phi_{\sigma\sigma}$ and Δ have opposite signs unless they are equal to zero. Hence, only the solutions (25) for (29) and (26) for (35) satisfy the sufficient conditions for a minimum of the free energy (10).

As is well known, the equations for the dependence σ on τ, in the cases (25), (26), describe σ as single-valued[6] and decreasing functions of τ in the temperature regions which correspond to $0 < \sigma < 1$ (it means $0 < \tau < \gamma$ for (25) and $0 < \tau < \infty$ for (26)). This can be easy proved, since due to (31)–(34) we obtain, generally, for all the cases (25)–(27). that

$$\operatorname{sign} \frac{d\tau}{d\sigma} = -\operatorname{sign} \Phi_{\sigma\sigma} \tag{36}$$

and so $\frac{d\sigma}{d\tau}$ is negative for (25) and (26). If

$$h_\perp < |\varkappa| \tag{37}$$

the two minima of the free energy lie in temperature intervals which are separated by a field--dependent point τ_c,

$$\tau_c = \frac{3s}{s+1} \frac{\gamma h_\perp}{|\varkappa| \tilde{B}_s \left(\frac{h_\perp}{|\varkappa|}\right)}, \tag{38}$$

according to (25), (26) and (29), (35). Thus, as long as the condition (37) is satisfied, the solutions (25), (26), which describe the magnetization as decreasing function of temperature, minimize the free energy in the regions $0 < \tau < \tau_c$, $\tau_c < \tau < \infty$, respectively; otherwise, the solution (26) satisfies the minimum conditions in the whole temperature region.

As concerns the solution (27), the expression for Δ, (33) and (34), enable to discuss this case as well. As one sees from (33), (34), (36) and the properties of $\tilde{B}_s(x)$ given in the Appendix, in this case the function $\sigma(\tau)$ need not be monotonic. By a simple graphical analysis of the relation between σ and τ, (27), one can show that there can exist not more than two solutions of this equation with respect to σ. Hence, the function $\sigma(\tau)$ is double--valued if these solutions exist, and due to (33), (34), (36) one of its branches, $\sigma^{(in)}(\tau)$, is an increasing function of temperature, and the other, $\sigma^{(d)}(\tau)$, a decreasing one. If so, $\sigma^{(in)}(\tau)$ satisfies the conditions for a maximum of the free energy (10), while there is no extremum for the solution $\sigma^{(d)}(\tau)$ of (27). Both the solutions $\sigma^{(in)}(\tau)$, $\sigma^{(d)}(\tau)$ disappear above a threshold point τ_t (the term used in [16]) which can be determined from the condition $\frac{d\tau}{d\sigma} = \Phi_{\sigma\sigma} = 0$ (note (36)). Moreover, by a simple analysis of all the relations between σ and τ, (25)–(27), which temporarily we denote respectively by $\sigma^{(25)}(\tau)$, $\sigma^{(26)}(\tau)$ and $\sigma^{(in)}(\tau)$, $\sigma^{(d)}(\tau)$, one can find that

$$\lim_{\tau \to \infty} \sigma^{(26)}(\tau) = 0 \tag{39}$$

$$\begin{gathered} \tau_t < \tau_c \\ \sigma^{(25)}(\tau) \geqslant \sigma^{(26)}(\tau) \geqslant \sigma^{(d)}(\tau) \geqslant \sigma^{(in)}(\tau) \qquad 0 \leqslant \tau \leqslant \tau_t \\ \text{for} \\ \sigma^{(25)}(\tau) \geqslant \sigma^{(26)}(\tau) \qquad \tau_t \leqslant \tau \leqslant \tau_c \end{gathered} \tag{40}$$

[6] Since $\sigma_\perp = h_\perp / |\varkappa|$ and we assume $h_\perp \neq 0$, the solution $\sigma = \sqrt{\sigma_{||}^2 + \sigma_\perp^2} = 0$ of the equation $\frac{s+1}{3s} \tau \tilde{B}_s(\sigma) = \gamma\sigma$ is excluded.

where the equality sings in (40) hold only at the boundaries of the temperature intervals For illustration, the dependence of σ on τ for (25)–(27) is shown in Fig. 1.

In short, the (ferromagnetic) solution (25) exists in the temperature interval $0 < \tau < \tau_c$, satisfying the conditions for the minimum in this region; the (paramagnetic) solution (26) exists in the whole temperature region ($0 < \tau < \infty$) but satisfies the minimum conditions only for $\tau > \tau_c$. This is also marked in Fig. 1.

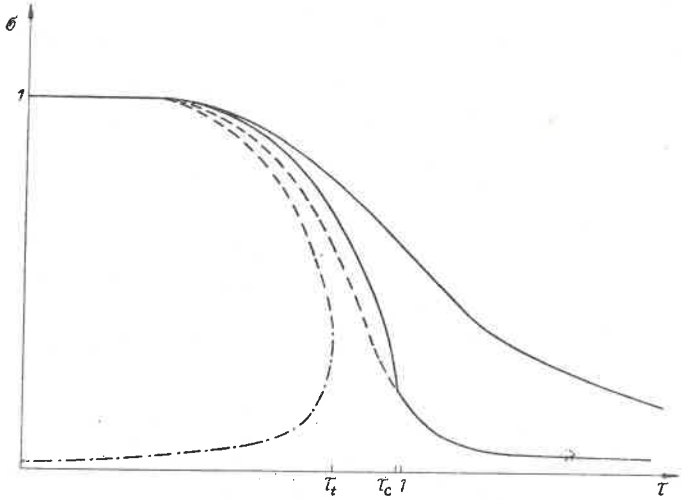

Fig. 1. Schematic curves $\sigma(\tau)$ corresponding to Eqs. (25)–(27). Solid, broken and dotted-broken lines represent respectively minima, maxima and absence of extrema of the free energy. The upper solid line corresponds to $h_\perp = 1.1\,\varkappa$, the remaining curves to $h_\perp = 0.2\varkappa$ ($\varkappa > 0$)

For examining the transition between the magnetic states described by the solutions (25), (26), the same procedure is used as in the case of the toroid model [4,9]. Accordingly, by inserting (25) and (26) into (10) (see also (8) and (28)) one obtains the free energy corresponding, respectively, to the ferro- and paramagnetic phases

$$\varphi_f = -\frac{s+1}{3s}\tau \ln \frac{\operatorname{sh}\dfrac{3(2s+1)}{2(s+1)}\dfrac{\gamma\sigma_f}{\tau}}{\operatorname{sh}\dfrac{3}{2(s+1)}\dfrac{\gamma\sigma_f}{\tau}} + \frac{1}{2}\gamma\sigma_f^2 - \frac{1}{2}\frac{h_\perp^2}{|\varkappa|} \tag{41}$$

(for $\tau < \tau_c$)

$$\varphi_p = -\frac{s+1}{3s}\tau \ln \frac{\operatorname{sh}\dfrac{3(2s+1)}{2(s+1)}\dfrac{\gamma'\sigma_p+h_\perp}{\tau}}{\operatorname{sh}\dfrac{3}{2(s+1)}\dfrac{\gamma'\sigma_p+h_\perp}{\tau}} + \frac{1}{2}\gamma'\sigma_p^2 \tag{42}$$

(for $\tau > \tau_c$)

where

$$\sigma_f = B_s\left(\frac{3s}{s+1}\frac{\gamma\sigma_f}{\tau}\right), \quad \sigma_p = B_s\left(\frac{3s}{s+1}\frac{\gamma'\sigma_p+h_\perp}{\tau}\right). \tag{43}$$

Differentiation of these free energies with respect to τ at constant $h_\perp$ yields the ferro- and paramagnetic entropies $S_{f,p}$ and the specific heats $C_{f,p}$ (all these quantities are given in units of Nk = gas constant)

$$S_f = \ln \frac{\operatorname{sh} \frac{3(2s+1)}{2(s+1)} \frac{\gamma\sigma_f}{\tau}}{\operatorname{sh} \frac{3}{2(s+1)} \frac{\gamma\sigma_f}{\tau}} - \frac{3s}{s+1} \frac{\gamma\sigma_f^2}{\tau} \tag{44}$$

$$S_p = \ln \frac{\operatorname{sh} \frac{3(2s+1)}{2(s+1)} \frac{\gamma'\sigma_p + h_\perp}{\tau}}{\operatorname{sh} \frac{3}{2(s+1)} \frac{\gamma'\sigma_p + h_\perp}{\tau}} - \frac{3s}{s+1} \frac{\sigma_g(\gamma'\sigma_p + h_\perp)}{\tau} \tag{45}$$

$$C_f = \frac{\sigma_f \tilde{B}_s^2(\sigma_f)}{\sigma_f \tilde{B}_s'(\sigma_f) - \tilde{B}_s(\sigma_f)} \tag{46}$$

$$C_p = \frac{(\gamma'\sigma_p + h_\perp)\tilde{B}_s^2(\sigma_p)}{(\gamma'\sigma_p + h_\perp)\tilde{B}_s'(\sigma_p) - \gamma' B_s(\sigma_p)}. \tag{47}$$

Since $\sigma_f = \sigma_p = h_\perp/|\varkappa|$ at τ_c, it is easy to notice that the entropy (and the free energy (41), (42)), contrary to the specific heat, is continuous through the transition. It proves the existence of a second-order phase transition at the field-dependent point τ_c. Thus, the formula (38) gives the dependence of the critical temperature τ_c on the external magnetic field, or rather on the ratio of the external field to the anisotropy constant. One easily sees that this formula leads to the ordinary Curie temperature in the case $\frac{h_\perp}{|\varkappa|} = 0$. The magnitude of the specific heat jump at τ_c, being given by

$$\Delta C \equiv C_f(\tau_c) - C_p(\tau_c) = \frac{\tilde{B}_s^2\left(\frac{h_\perp}{|\varkappa|}\right)}{\tilde{B}_s'\left(\frac{h_\perp}{|\varkappa|}\right) - \frac{|\varkappa|}{h_\perp}\tilde{B}_s\left(\frac{h_\perp}{|\varkappa|}\right)} - \frac{\gamma\tilde{B}_s^2\left(\frac{h_\perp}{|\varkappa|}\right)}{\gamma\tilde{B}_s'\left(\frac{h_\perp}{|\varkappa|}\right) - \gamma'\frac{|\varkappa|}{h_\perp}\tilde{B}_s\left(\frac{h_\perp}{|\varkappa|}\right)} \tag{48}$$

depends however explicitly $\left(\text{not only through } \frac{h_\perp}{|\varkappa|}\right)$ on the material constants (see also (8)). Its dependence on $\frac{h_\perp}{|\varkappa|}$ for two different values of $|\varkappa|$ is illustrated in Fig. 2, while the phase diagram $\left(\text{and the dependence of the critical temperature } \tau_c \text{ on } \frac{h_\perp}{|\varkappa|}\right)$ is shown in Fig. 3. According to (37), (48) and Figs 1–3, the phase transition vanishes if the external field $h_\perp$ exceeds the value of $|\varkappa|$. The approximate magnitude of this field is of the order 10^6–10^3 Oe for a ferromagnet for which the zero-field Curie point $T_c(0)$ is about 1000°K and $10^{-2} < |\varkappa| < 10^{-4}$, and 10^4–10^2Oe for $T_c(0) \approx 10$°K (see (8)).

In view of (11), (18) and (25), the components of the magnetization in the ferromagnetic phase are

$$\sigma_{f,\perp} = \frac{h_\perp}{|\varkappa|}, \quad \sigma_{f,\parallel} = \pm\sqrt{\sigma_f^2 - \left(\frac{h_\perp}{\varkappa}\right)^2}. \tag{49}$$

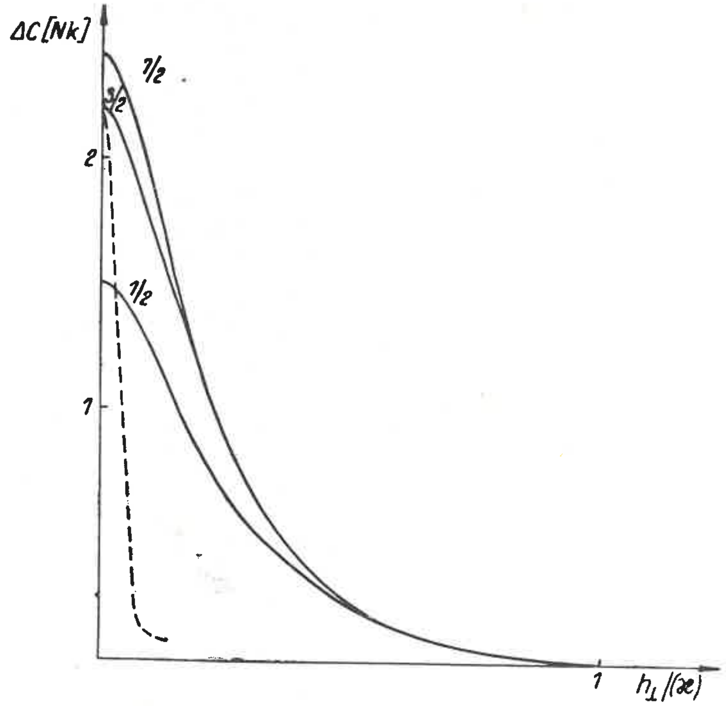

Fig. 2. The dependence of the specific-heat jump ΔC on $h_\perp/|\varkappa|$ for $|\varkappa| = 4 \cdot 10^{-2}$ and $s = 1/2$, 3/2, 7/2, and for $|\varkappa| = 4 \cdot 10^{-4}$, s = 3/2 (broken line)

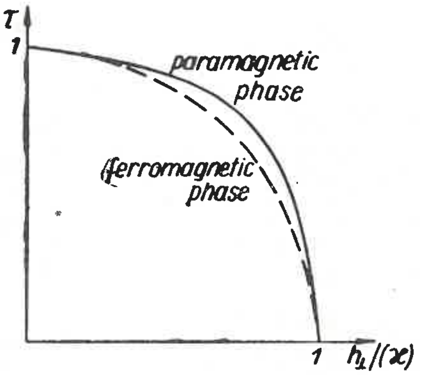

Fig. 3

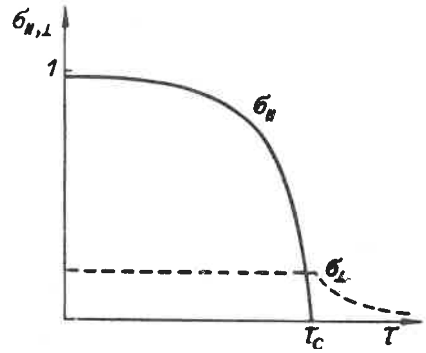

Fig. 4

Fig. 3. The $H-T_c$ diagram for the ferro- and paramagnetic phases: $s = 1/2$ — solid line, $s = 7/2$ — broken line

Fig. 4. The temperature-dependence of the magnetization components $\sigma_{\parallel}$ and $\sigma_\perp$ as given by (49) for $\tau < \tau_c$, and (26) for $\tau > \tau_c$ ($h_\perp = 0.2\varkappa$, $\varkappa > 0$)

As it is seen, only one of them, namely $\sigma_{f,||}$, depends on the temperature and vanishes above τ_c, due to (29) and (43). To illustrate, we present the dependence of the magnetization components on the temperature in Fig. 4.

4. *External field parallel to the easy axis (or plane; $H_{||} \neq 0$, $H_{\perp} = 0$) and the field-free case ($H_{||} = 0$, $H_{\perp} = 0$)*

For comparison with the results of the previous Section, we present here also a rigorous mathematical treatment of the longitudinal- and zero-field cases. As previously, we start with the solutions of (6), (14) and (15) (see also (18) and (28)).

Provided that $h_{||} \neq 0$, $h_{\perp} = 0$ we obtain

$$n_{||} = -\frac{h_{||}}{|\varkappa|\sigma}, \quad n_{\perp} = \pm\sqrt{1-n_{||}^2}, \quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma'\sigma, \qquad (50)$$
$$\lambda = \gamma\sigma^2,$$

$$n_{||} = 1, \quad n_{\perp} = 0, \quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma\sigma + h_{||}, \qquad (51)$$
$$\lambda = \sigma(\gamma\sigma + h_{||}),$$

$$n_{||} = -1, \quad n_{\perp} = 0, \quad \frac{s+1}{3s}\tau\tilde{B}_s(\sigma) = \gamma\sigma - h_{||}, \qquad (52)$$
$$\lambda = \sigma(\gamma\sigma - h_{||}).$$

Due to (19)–(24), the sufficient conditions for a minimum of the free energy (10) to exist are

$$\Phi_{\sigma\sigma} = \gamma'\Psi(\sigma) - \frac{h_{||}^2}{|\varkappa|\sigma^2} > 0$$
$$\Delta = \Phi_{\sigma\sigma}\left(\frac{h_{||}^2}{|\varkappa|} - |\varkappa|\sigma^2\right) > 0 \qquad (53)$$

for (50),

$$\Phi_{\sigma\sigma} = \gamma\Psi(\sigma) + \frac{h_{||}\tilde{B}_s'(\sigma)}{\tilde{B}_s(\sigma)} > 0,$$
$$\Delta = \sigma\Phi_{\sigma\sigma}(|\varkappa|\sigma + h_{||}) > 0 \qquad (54)$$

for (51), and

$$\Phi_{\sigma\sigma} = \gamma'\Psi(\sigma) - \frac{h_{||}\tilde{B}_s'(\sigma)}{\tilde{B}_s(\sigma)} > 0,$$
$$\Delta = \sigma\Phi_{\sigma\sigma}(|\varkappa|\sigma - h_{||}) > 0 \qquad (55)$$

for (52), upon eliminating τ from $\Phi_{\sigma\sigma}$ according to (50)–(52). Here, $\psi(\sigma)$ is given by (34). If we assume $h_{||} > 0$, the solution (50) is real for

$$|\varkappa|\sigma < h_{||},$$

and, as a consequence of (34) and due to the properties of $\tilde{B}_s(x)$ (see Appendix), this solution does not satisfy the conditions for an extremum if $\Phi_{\sigma\sigma} > 0$ $(\Delta < 0)$, or corresponds to a maximum if $\Phi_{\sigma\sigma} < 0$, $(\Delta > 0)$. On the other hand, one obtains that (51) satisfies the conditions (54) in the whole interval $0 < \sigma < 1$. As regards (52) this solution is restricted to (cp. (30))

$$\gamma\sigma > h_{||}, \tag{57}$$

and since the identity (36) is also valid for (50)–(55), we can discuss the solution (52) in the same way as (27) in the previous Section. Thus, we can show that the implicit dependence $\sigma(\tau)$, (52), describes two functions, $\sigma^{(in)}(\tau)$ and $\sigma^{(d)}(\tau)$, respectively increasing and decreasing with τ. Due to (36) and (53) if the condition

$$|\varkappa|\sigma > h_{||} \tag{58}$$

is satisfied, $\sigma^{(d)}(\tau)$ corresponds to a minimum as well, otherwise to the absence of an extremum. In the same manner we obtain a maximum or the absence of an extremum for $\sigma^{(in)}(\tau)$, depending on the sign of $|\varkappa|\sigma - h_{||}$. Similarly as in the previous Section, when denoting here the dependence $\sigma(\tau)$ by $\sigma^{(50)}(\tau)$, $\sigma^{(51)}(\tau)$ and $\sigma^{(in)}(\tau)$, $\sigma^{(d)}(\tau)$ for (50)–(52) respectively, one can prove the validity of the inequalities

$$\begin{gathered} \sigma^{(51)}(\tau) \geqslant \sigma^{(d)}(\tau) \geqslant \sigma^{(in)}(\tau) \\ \sigma^{(51)}(\tau) \geqslant \sigma^{(50)}(\tau) \end{gathered} \tag{59}$$

in the temperature region where the respective solutions exist, and that

$$\lim_{\tau\to\infty} \sigma^{(51)}(\tau) = 0. \tag{60}$$

The dependence of σ on τ for the solutions (50)–(52) is illustrated in Fig. 5. If we denote the free energies which correspond to the solutions (51), (52) by $\varphi_{(51)}(\tau)$ and $\varphi_{(52)}(\tau)$, respectively, it can be proved that

$$\lim_{\tau\to 0} \varphi_{(51)}(\tau) < \lim_{\tau\to 0} \varphi_{(52)}(\tau), \qquad \frac{d\varphi_{(51)}}{d\tau} < \frac{d\varphi_{(52)}}{d\tau} < 0, \tag{61}$$

which implies

$$\varphi_{(51)}(\tau) < \varphi_{(52)}(\tau). \tag{62}$$

Hence, as one would expect, the solution (51) satisfies the conditions for the absolute minimum of the free energy (10). The substitution of this solution in (10) gives the free energy $\varphi_{(51)}(\tau) \equiv \varphi(\tau, h_{||})$

$$\varphi(\tau, h_{||}) = -\frac{s+1}{3s}\,\tau \ln \frac{\operatorname{sh} \dfrac{3(2s+1)}{2(s+1)} \dfrac{\gamma\sigma + h_{||}}{\tau}}{\operatorname{sh} \dfrac{3}{2(s+1)} \dfrac{\gamma\sigma + h_{||}}{\tau}} + \frac{1}{2}\gamma\sigma^2, \tag{63}$$

where

$$\sigma = B_s\left(\frac{3s}{s+1}\,\frac{\gamma\sigma + h_{||}}{\tau}\right), \tag{64}$$

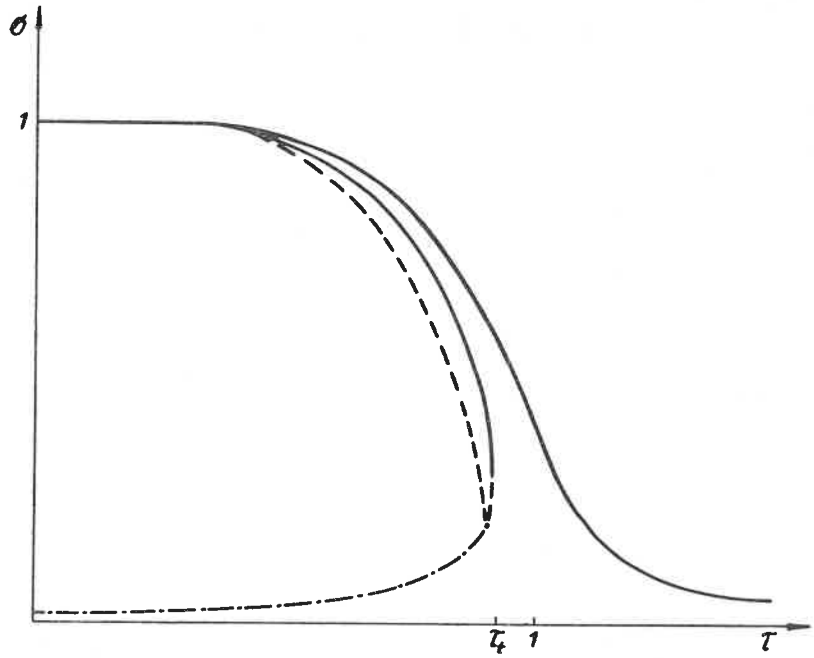

Fig. 5. Schematic curves $\sigma(\tau)$ according to (50)–(52) (denotation of lines as in Fig. 1), for $h_{||} = 0.2\varkappa$ ($\varkappa > 0$)

applying to the whole temperature region. Since $\varphi(\tau, h_{||})$ and its derivatives are continuous functions of τ and $h_{||}$, the phase transition cannot occur in this case.

We have shown that the solution $\sigma^{(d)}(\tau)$ (magnetization vector antiparallel to the external field and parallel to the easy direction) satisfies the conditions for a relative minimum of the magnetic free energy (see, however, the restriction (58)). This solution is apparently relevant in the case of domain structure and its existence supports the considerations of [16]. Similarly, our considerations exclude the solution $\sigma^{(in)}(\tau)$ which had to be rejected in [16] by a rather physical argument.

If $h_{||} = 0$, $h_{\perp} = 0$, the system of equations (6), (14) and (15) has the solutions (see also (18) and (28))

$$\sigma = 0, \quad \lambda = 0 \tag{65}$$

$$n_{||} = \pm 1, \quad n_{\perp} = 0, \quad \frac{s+1}{3s}\,\tau \tilde{B}_s(\sigma) = \gamma\sigma, \quad \lambda = \gamma\sigma^2, \quad \sigma \neq 0 \tag{66}$$

$$n_{||} = 0, \quad n_{\perp} = \pm 1, \quad \frac{s+1}{3s}\,\tau \tilde{B}_s(\sigma) = \gamma'\sigma, \quad \lambda = \gamma'\sigma^2, \quad \sigma \neq 0. \tag{67}$$

In the case of (65), in which the magnetization direction is obviously meaningless, the free energy (10) does not depend on the direction cosines and (21), (22) reduces to

$$d^2\Phi = \left(\frac{s+1}{3s}\,\tilde{B}_s'(0) - \tilde{P}_{33}\right) d\sigma^2. \tag{68}$$

According to (20), (21), the angle of the transformation ϑ can be easily found in the remaining two cases (66), (67). Having eliminated τ with the help of (66), (67), respectively, from (22) one obtains

$$\Phi = \gamma\Psi(\sigma), \quad \Delta = |\varkappa|\sigma^2\Phi_{\sigma\sigma} \tag{69}$$

for the solution (66), and

$$\Phi = \gamma'\Psi(\sigma), \quad \Delta = -|\varkappa|\sigma^2\Phi_{\sigma\sigma} \tag{70}$$

for (67), where $\Psi(\sigma)$ is defined by (34). Due to (34) (see also 28)), one has a minimum in the case (66) ($\Phi_{\sigma\sigma} > 0$, $\Delta > 0$) and the absence of an extremum in the case (67) ($\Phi_{\sigma\sigma} > 0$, $\Delta < 0$). As regards the solution (65), due to the fact that $\tilde{B}_s'(0) = \dfrac{3s}{s+1}$ and according to (20), (23), (68), we have

$$d^2\Phi > 0 \text{ for } \tau > \alpha + \varkappa \cos^2 \vartheta. \tag{71}$$

Hence, the point above which σ vanishes depends on the magnetization direction below that point, *i. e.*, on the direction of the quantization axis (see, *e. g.*, [12]). In other words, if there were any factors which would force the magnetization to assume another direction than that of the easy axis or plane, according to (65), (71) the point above which σ vanishes would depend on this direction. Such a result has been obtained in [17], while examining the influence of the domain structure on the transition temperature of a ferromagnet.

The solution (65), (66), correspond, of course, to the para- and ferromagnetic phases, respectively. Upon substituting them into (10) (see also (28)) one has the free energy for the ferromagnetic phase

$$\varphi_f = -\frac{s+1}{3s}\tau \ln \frac{\operatorname{sh} \dfrac{3(2s+1)}{2(s+1)} \dfrac{\gamma\sigma_f}{\tau}}{\operatorname{sh} \dfrac{3}{2(s+1)} \dfrac{\gamma\sigma_f}{\tau}} + \frac{1}{2}\gamma\sigma_f^2 \tag{72}$$

where $\sigma_f \neq 0$ is determined by

$$\sigma_f = B_s\left(\frac{3s}{s+1}\frac{\gamma\sigma_f}{\tau}\right), \tag{73}$$

and respectively for the paramagnetic one

$$\varphi_p = \lim_{\sigma \to 0} \varphi = -\frac{s+1}{3s}\tau \ln (2s+1). \tag{74}$$

As one can easily prove making use of (73), the condition $\sigma_f \neq 0$ can only be satisfied for $\tau < \gamma$ and the limit temperature for $\sigma_f \to 0$ is $\tau = \gamma$. Thus, it follows from (66) and (71) that in the temperature intervals

$$\begin{aligned} \alpha < \tau < 1 \qquad & \varkappa > 0 \\ \alpha + \varkappa < \tau < \alpha \quad \text{for} \quad & \varkappa < 0 \end{aligned} \tag{75}$$

the sufficient conditions for a relative minimum of φ are satisfied by (65) as well as by (66). However, by expanding the function $\varphi_f(\sigma_f)$ into a Maclaurin series with respect to σ_f, it can be found that $\varphi_f < \varphi_p$, and so φ_f satisfies the condition of the absolute minimum φ in those temperature intervals. The differentiation of the above formulae for the free energy (72), (74) with respect to τ yields the ferro- and paramagnetic entropies $S_{f,p}$ and specific heats $C_{f,p}$, respectively. From these formulae one can easily find that $S_f(\tau = \gamma) = S_p(\tau = \gamma)$ and $C_f(\tau = \sigma) \neq C_p(\tau = \gamma)$, with jump of the specific heat given (in units of Nk = gas constant) by

$$\Delta C = \frac{5s(s+1)}{2s(s+1)+1}. \tag{76}$$

5. *Critical parameters*

For the most part recent theories of phase transitions (see, *e. g.*, [1, 2]) are based on Landau's assumption that the free energy is expandable into a power series with respect to an order parameter [18]. Such approach enables, in particular, to describe the behaviour of a system near the critical point by means of a set of critical indices. The apparent relations between these parameters can be established on the basis of the scaling hypothesis (see, *e. g.*, [1]). If the phase transition of a uniaxial ferromagnet is to be described by means of these theories, one should answer the questions what is the appropriate order parameter in this case, and whether the application of the scaling approach to this problem yields satisfactory results.

As regards the first question, it is easy to notice that the longitudinal component of the magnetization (parallel to the easy axis or plane — $\sigma_{f,||}$) has all the properties which are required of an order parameter (cp. (49) and [1,19]). It is rather obvious that this critical parameter should be chosen in such a way as to correspond to the case $H_\perp = 0$, *i. e.*, to the absolute value of the magnetization in this case. One sees from (49) that $\sigma_{f,||}$ satisfies this demand as well. Furthermore, one can easily show, by expanding the implicit function $\sigma^2_{f,||}$, (49), into a Taylor series, that in the neighbourhood of τ_c

$$\sigma^2_{f,||} \approx a\left(1 - \frac{\tau}{\tau_c}\right) \tag{77}$$

where

$$a = \frac{2\frac{h_\perp}{|\varkappa|}\tilde{B}_s\left(\frac{h_\perp}{|\varkappa|}\right)}{\tilde{B}_s\left(\frac{h_\perp}{|\varkappa|}\right) - \frac{|\varkappa|}{h_\perp}\tilde{B}_s\left(\frac{h_\perp}{|\varkappa|}\right)}, \quad \lim_{\substack{h_\perp/|\varkappa|\to 0\\(\tau_c\to\gamma)}} a = \frac{10}{3}\,\frac{(s+1)^2}{2s(s+1)+1}. \tag{78}$$

It means that the behaviour of the component $\sigma_{f,||}$ near the critical field-dependent point resembles that of the total magnetization in the neighbourhood of the ordinary (field-free) Curie temperature.

The answer to the second question was given in [12], where the relation between the critical parameters, which determine the field-dependence of the critical temperature, the transversal magnetization ($\sigma_{f,\perp}$ in our notation — (49)) [4, 5, 8–10] and the temperature dependence of the magnetization for $H = 0$ (see, *e. g.*, (77), (78) and [1]), has been established.

To show the explicit derivation of the formula for the field-dependence of the critical temperature in the neighbourhood of the ordinary Curie point ($H_\perp = 0$) [10], we expand the right-hand side of (38) into a power series of $h_\perp/|\varkappa|$. From the expansion of the Brillouin function, which due to (12) reads

$$B_s(x) = \sum_{l=1}^{\infty} \frac{(-1)^{l+1} 2^{2l} B_l}{(2l)!} A_{2l} x^{2l-2}, \tag{79}$$

we obtain for its inverse function

$$\tilde{B}_s(x) = b_1 x + b_3 x^3 + b_5 x^5 + O(x^7), \tag{80}$$

where in (79) B_l denotes the Bernoulli numbers and

$$A_{2l} \equiv \left(\frac{2s+1}{2s}\right)^{2l} - \left(\frac{1}{2s}\right)^{2l}, \tag{81}$$

while the coefficients in (80) have the form

$$b_1 = 3A_2^{-1}, \quad b_3 = \frac{9}{5} A_2^{-4} A_4,$$
$$b_5 = \frac{81}{5} A_2^{-7} \left(\frac{1}{5} A_4^2 - \frac{2}{21} A_2 A_6\right). \tag{82}$$

When applying (80) to (38), we obtain

$$\tau_c = \gamma \left\{1 - \frac{3}{10} \frac{2s(s+1)+1}{(s+1)^2} \left(\frac{h_\perp}{\varkappa}\right)^2\right\} + O\left\{\left(\frac{h_\perp}{\varkappa}\right)^4\right\}. \tag{83}$$

It is worth-while to note that a similar dependence of the critical temperature on the external field (with field in second power) has been found experimentally [3] (in the power 1.6±0.4) as well as theoretically [4, 5, 8–10].

Since according to (43) $\sigma_f = \sigma_p = \dfrac{h_\perp}{|\varkappa|}$ at τ_c, the application of (80) to (46) and (47) yields

$$C_f(\tau_c) = c_0 - c_2 \left(\frac{h_\perp}{\varkappa}\right)^2 + O\left\{\left(\frac{h_\perp}{\varkappa}\right)^4\right\} \tag{84}$$

$$C_p(\tau_c) = \frac{3s}{s+1} \frac{\gamma}{|\varkappa|} \left(\frac{h_\perp}{\varkappa}\right)^2 + O\left\{\left(\frac{h_\perp}{\varkappa}\right)^4\right\}, \tag{85}$$

where in (84)

$$c_0 = \frac{5s(s+1)}{2s(s+1)+1}, \quad c_2 = \frac{3s}{14(s+1)} \frac{32s^4 + 64s^3 + 84s^2 + 52s + 13}{[2s(s+1)+1]^2}. \tag{86}$$

When comparing with (76), from the above formulae one sees that the jump of the specific heat (48) corresponds to that for $h_{||} = 0$, $h_\perp = 0$.

6. *Concluding remarks*

In the cases being here under consideration, the conditions for a minimum of the magnetic free energy (10) gave the possible magnetization directions illustrated in Fig. 6. Such directions, qualitatively, are also an immediate consequence of symmetry conditions, *i. e.*, of the fact that the anisotropy and the external field distinguish certain directions in the crystal [19]. These results are usually obtained when considering a uniaxial ferromagnet in the presence of an external magnetic field. With the exception of the dependence $\sigma(\tau)$,

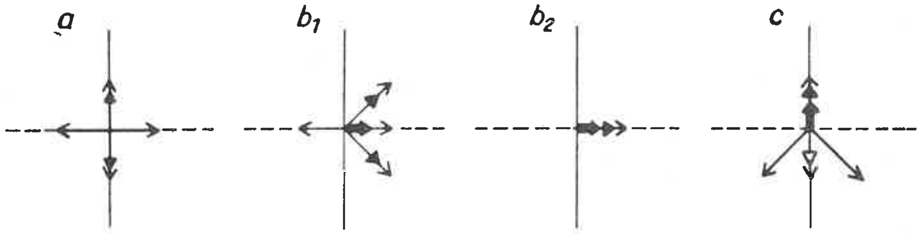

Fig. 6. The magnetization directions obtained from the necessary conditions for extrema of the model free energy in the cases: *a)* $h_{||} = 0$, $h_{\perp} = 0$; *b*$_1$) $h_{||} = 0$, $h_{\perp} \neq 0$, $\tau < \tau_c$; *b*$_2$) $h_{||} = 0$, $h \neq 0$, $\tau > \tau_c$; *c)* $h_{||} \neq 0$, $h_{\perp} = 0$. The solid lines mark the easy axis (plane); the arrows denote: ➡ the external magnetic field, → the directions obtained from the necessary conditions, —▶ the directions satisfying the sufficient conditions for the absolute minimum, —▷ the direction satisfying the sufficient conditions for a relative minimum

the direction cosines of the magnetization in the case $h_{\perp} \neq 0$, $h_{||} = 0$ (formulae (25)–(27)) are much the same as those obtained by the Green function method [14, 15] and the approximate second quantization [13]. Moreover, our results in that case correspond precisly to those given in [4, 9] when replacing the reduced demagnetization factor d with $\varkappa > 0$ (see also [10]).

The obtained results raise the question whether a second-order phase transition occurs in the case of an arbitrarily directed external magnetic field. As shown in [19] basing on the results of [9, 10, 5], the answer to this question is negative. It is impossible to confirm it in a rigorous way as presented here, for the necessary conditions (14), (15) for the minimum are in this case analytically unsolvable. However, there is a further argument supporting the results of [19] if we assume that — according to [19] and the previous Section — $\sigma_{f,||}$ is to be the order parameter. Namely, an infinitesimal external magnetic field along the easy axis should create $\sigma_{||} \neq 0$ for arbitrary temperature, which suffices to destroy the phase transition. Moreover, this would explain why it is experimentally difficult to detect the phase transition in a transversal magnetic field.

APPENDIX

In order to prove the inequality (34) we take advantage of the following properties of the function $\tilde{B}_s(x)$:

(*i*) $\tilde{B}_s(x)$ is defined, continuous and differentiable in the interval $-1 < x < 1$, (*ii*) $\tilde{B}_s(0) = 0$, (*iii*) $B_s'(x) > 0$, and in the interval $0 < x < 1$, (*iv*) $\tilde{B}_s(x) > 0$, (*v*) $\tilde{B}_s''(x) > 0$, resulting from (12). Due to (*i*), (*ii*), it follows from the mean value theorem that for any x from the interval (0,1) there exists an x_0 in the interval $(0, x)$ such that

$$x\tilde{B}_s(x_0) = \tilde{B}_s(x). \tag{A1}$$

Since $\tilde{B}_s'(x)$ is an increasing function (*v*), we have

$$x\tilde{B}_s'(x) > x\tilde{B}_s'(x_0). \tag{A2}$$

By combining (A1) with (A2) and taking into account (*iv*), one immediatly proves (34).

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