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MAGNETIZATION IN A FERROMAGNET WITH MAGNETIC AXIAL SYMMETRY

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Based on phenomenological free energy for a ferromagnet with axial symmetry, stable magnetic phases and their ranges of existence are determined as a function of the assumed material constants. The kind of phase transitions occuring at the stability boundaries are defined. The temperature is introduced according to Landau's assumption and the temperature dependence of the magnetization components calculated. It is shown that in the case under consideration the magnetization direction can change under the influence of temperature variations. Also, the magnetization length can increase with the temperature in a certain magnetic phase.

1. *Introduction*

Experimental investigations of recent years showed an intriguing dependence of the magnetization on temperature for certain ferromagnets. This unusual behaviour resides in the existence of the temperature region in which the magnetization increases with temperature [1]. The interpretation of this phenomenon was based on the spin-glas model [2] or by taking into account the domain structure of the sample [3].

The phenomenon in question occurs also in uranium compounds, in which, as is well known, the crystalline field plays a major role in forming the ordered state. It seems, however that the dependence of the magnetization on temperature, which is a manifestation of the possibility of different magnetic phases to occur, cannot only be a consequence of the single-ion interactions related to the crystalline field effects. It seems certain that anisotropic exchange interactions (super-exchange effects or the interaction via the conduction electrons in metalic compounds) should also play a vital role in some of, these compounds. The possibility of interplay of many types of interactions, and the arising problem of their coexistence, is very interesting from both the point of view of the general theory of magnetism and the particular properties discovered in these compounds.

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In the present paper we shall focus our attention on implications following from the coexistence of comparable exchange and single-ion interactions [4] or a special type crystallographic symmetry [5].

As shown in [4], a homogeneous deformation along the appropriate axis, when we have comparable exchange and single-ion interactions, might produce an easy magnetic axis and easy plane perpendicular to this axis.

Leaving behind microscopic considerations, one can ask how the phenomenon can be described qualitatively in terms of a phenomenological theory. To answer this question, appropriate symmetry conditions [4, 5] should be applied to a general form of the free energy and, then, physical conclusions drawn on a simple minimization procedure. The basic conclusion of the qualitative considerations of [4] and those presented here is a mere necessity of the symmetry axis to exist. We assume also the free energy to be of the fourth order in the magnetization components at most. In this way our model of axial ferromagnets is formulated on the physical and mathematical grounds.

Yet we should like to pay attention to the assumption that the lattice deformation plays a crucial role in our considerations. At the very least we suppose on this ground that the lattice deformation, which can apparently occur at the transition from the paramagnetic to ordered state, is of considerable importance. Therefore, the approches that neglect these effects should be treated with particular care. This concerns also the results of the present paper for the deformation, which leads to our initial-assumptions, is considered as constant and the relevant symmetry is not permitted to change at phase boundaries. This restriction should be born in mind when comparing our results with experimental data for a given magnetic compound. Such comparisons may be attempted, in particular, for rare earth and actinide compounds in which changes in the lattice parameters affects the occurence of magnetic phases and even the temperature dependence of the magnetization. Of course, the lattice transformations cannot be of such importance at some temperatures or for some compounds and then our results can be fully applicable. It is hard to find a general criterion for the validity of the above-mentioned restriction, which — in our opinion — should be taken into account when comparing our results with the given experimental data.

2. *Phase diagrams*

According to our assumption the generalized free energy of an axial ferromagnet can be written in the form [4]

$$F = k_1 m_1^2 + k_2 m_2^2 + d_1 m_1^4 + d_2 m_2^4 + 2c m_1^2 m_2^2, \tag{1}$$

where m_i denote the i-th components of the magnetization vector, k_i, d_i and c are constants ($i = 1, 2$). Principally, the same free energy was studied by Imry [6]. However, these investigations were restricted to studying phase diagrams when there exist a coupling of two order parameters. In particular, the influence of the coupling parameter (c — in our notation) on the occurence and kind of phase transitions, was examined while the functional dependence of the order parameters (magnetization components) on the temperature was neglected in [6]. As we emphasized at the beginning, the temperature dependence of the magnetization is of basic importance in our considerations. As concerns the phase diagrams, we shall construct them in another way, which is, in our opinion, better for interpretation. Moreover, these diagrams are independent as the temperature dependence of the coefficients k_1 and k_2 in the free energy (1) is assumed.

The necessary conditions for an extremum of (1) with respect to m_1 and m_2 to exist lead to

$$m_1(k_1+2d_1m_1^2+2cm_2^2) = 0, \quad m_2(k_2+2d_2m_2^2+2cm_1^2) = 0. \tag{2}$$

By solving these equations and examining the sufficient conditions, we conclude that the free energy takes on minima if

$$m_1 = m_2 = 0 \quad \text{and } k_2 > 0,\ k_1 > 0 \tag{3}$$

$$m_1 = 0, \quad m_2^2 = -\frac{k_2}{2d_2} \quad \text{and } k_2 < 0, \quad d_2 > 0, \quad k_1 > \frac{c}{d_2}k_2 \tag{4}$$

$$m_1^2 = -\frac{k_1}{2d_1}, \quad m_2 = 0 \quad \text{and } k_1 < 0, \quad d_1 > 0, \quad k_2 > \frac{c}{d_1}k_1 \tag{5}$$

$$m_1^2 = \tfrac{1}{2}\frac{d_2k_1-ck_2}{c^2-d_1d_2}, \quad m_2^2 = \tfrac{1}{2}\frac{d_1k_2-ck_1}{c^2-d_1d_2} \quad \text{and } d_1 > 0, \quad d_2 > 0, \quad d_1d_2 > c^2. \tag{6}$$

In the last case, (6), the reality of m_1 and m_2 requires that the inequalities

$$k_1-\frac{c}{d_2}k_2 < 0, \quad k_2-\frac{c}{d_1} < 0 \quad \text{and } k_1 < 0, \quad k_2 < 0 \tag{7}$$

be satisfied. On the grounds of the relations (3)—(7) we can illustrate the regions of appropriate state in the plane (k_1, k_2). The states defined by (3)—(6) will be denoted: Eq. (3) — A, Eq. (4) — B, Eq. (5) — C, Eq. (6) — D. As seen from (3)—(7) we can distinguish two cases: I $d_1d_2-c^2 \leqslant 0$, which corresponds only to three phases — the paramagnetic A and two ferromagnetic ones B and C; II $d_1d_2-c^2 > 0$, when all the four phases — paramagnetic A, ferromagnetic B and C as well as deflection one D — can occur. The resulting phase diagrams for positive material constants d_1, d_2 and c are shown in Figs. 1 and 2. By a simple discussion one can show that in case II our system undergoes second-order phase transitions induced by certain changes of k_1 and k_2. In the opposite case I both second- and first-order phase transitions can occur because of an overlap of the states B and C on the plane (k_1k_2). It can easily be shown that the state B is stable and C metastable if

$$d_2k_1^2 > d_1k_2^2. \tag{8}$$

The oposite situation (B — metastable, C — stable state) corresponds to

$$d_2k_1^2 < d_1k_2^2. \tag{9}$$

Let us introduce temperature to our system in a standard way

$$k_1 = a_1 \frac{T-T_1}{T_1} \quad \text{and} \quad k_2 = a_2 \frac{T-T_2}{T_2}, \tag{10}$$

where $a_1 > 0$, $a_2 > 0$ (in our further considerations we shall still assume that d_1, d_2 and c are positive as well). The Landau, assumption (10), is obviously a rough approximation and, therefore, conclusions about the temperature dependence of the magnetization are of limited validity[1]. It is, however, reasonable to hope that gross features of this dependence

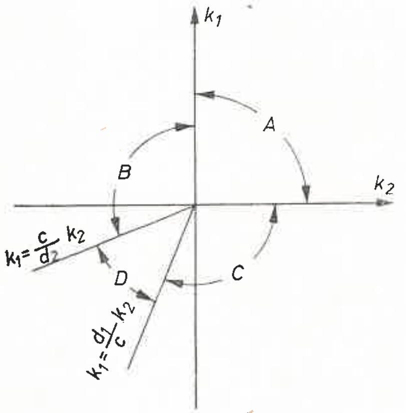

Fig. 1. Phase diagram if $d_1 d_2 > c^2$

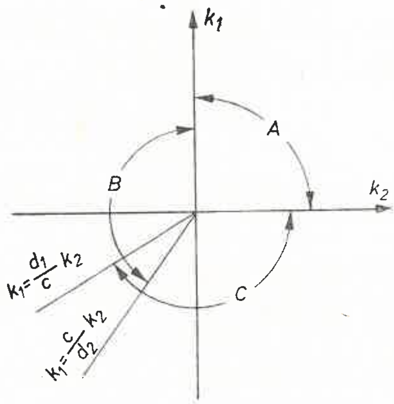

Fig. 2. Phase diagram if $d_1 d_2 \leqslant c^2$

are reproduced correctly. When treating T as a parameter from (10) we get the following linear relation

$$k_1 = \frac{a_1 T_2}{a_2 T_1} k_2 + a_1 \left(\frac{T_2}{T_1} - 1\right). \tag{11}$$

It should be noted that solely the region $k_1 > -a_1$, $k_2 > -a_2$ on the plane $(k_1 k_2)$ have a physical meaning ($T \geqslant 0$ in (10)) and an increase in T implies an appropriate shift on the straight line (11) in the positive direction of k_1 and k_2.

Upon introducing (10) to (4)–(6), we obtain the following temperature dependence of the magnetization in the respective states and intervals of the temperature in which they occur

A. $$m_1 = 0,\ m_2 = 0; \quad T > T_2 \quad \text{and} \quad T > T_2, \tag{12}$$

B. $$m_1 = 0, \quad m_2^2 = -\frac{a_2}{2d_2 T_2} T + \frac{a_2}{2d_2}; \quad T < T_2 \text{ and } \bar{\alpha} T > \alpha, \tag{13}$$

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[1] This approximation is strictly valid at T close to T_i, however, the critical fluctuations result in this way that it does not relate to the closest vicinity of T_c. It is obvious that obtained relations (e.g. magnetization versus temperature) give only qualitative suggestions for temperature far from T_i. Generally, accuracy of approximation applied here is also dependent on materials because material constant determine a range of the validity of Landau theory (see e.g. [7]).

$$\text{C.}\quad m_1^2 = -\frac{a_1}{2d_1T_1}T + \frac{a_1}{2d_1}, \qquad m_2 = 0; \qquad T < T_1 \text{ and } \bar{\beta}T > \beta, \tag{14}$$

$$\text{D.}\quad m_1^2 = \frac{\bar{\alpha}}{2\varphi}T - \frac{\alpha}{2\varphi}, \qquad m_2^2 = \frac{\bar{\beta}}{2\varphi}T - \frac{\beta}{2\varphi}; \qquad \bar{\alpha}T < \alpha \text{ and } \bar{\beta}T < \beta, \tag{15}$$

where we introduced the following, notation

$$\frac{a_1d_2}{T_1} - \frac{ca_2}{T_2} = \bar{\alpha}, \quad \frac{a_2d_1}{T_2} - \frac{ca_1}{T_1} = \bar{\beta}, \quad a_1d_2 - ca_2 = \alpha, \quad a_2d_1 - ca_1 = \beta,$$

$$c^2 - d_1d_2 = \varphi. \tag{16}$$

Phase transitions between different states can occur when the temperature line (11) crosses the phase boundaries in our phase diagrams. Thus, we obtain the following relations

$$T^{AB} = T_2, \quad T^{AC} = T_1, \quad T^{CD} = \frac{\beta}{\bar{\beta}}, \quad T^{BD} = \frac{\alpha}{\bar{\alpha}}, \tag{17}$$

which define the temperature of the phase transitions. If $d_1d_2 - c^2 < 0$, T^{CD} and T^{BD} determine the temperature boundary for the existence of the states C and B, respectively. In this case we shall denote these temperature values by $T^C (= T^{CD})$ and $T^B (= T^{BD})$. They correspond to discontinuous changes of the free energy from metastable states to more favorable ones. As it follows from (8), (9) and (10) the temperature at which the state B (as well C) ceases to be stable (and becomes metastable) is determined by

$$T^{BC} = \frac{a_1d_2 - a_2d_1}{\dfrac{a_1d_2}{T_1} - \dfrac{a_2d_1}{T_2}}. \tag{18}$$

On the ground of (17) and (18) one can show that

$$T^B < T^{BC} < T^C \quad \text{when} \quad T_1 > T_2 \tag{19}$$

and

$$T^B > T^{BC} > T^C \quad \text{when} \quad T_1 < T_2. \tag{20}$$

3. *Temperature dependence of the magnetization components*

The presented results permit one to analyze changes in the magnetization and phases due to temperature variations. The straight line (11) determines, in fact, also changes in the magnetization depending on the mutual relations of a_1, T_1, a_2, T_2, besides the phase transformations. There change scan be considered in an analytical or graphical way. Assume, $T_1 > T_2$. The opposite case can be easily obtained by the simple renumeration ($1 \to 2, 2 \to 1$) of the material constants and, therefore, the results are quite similar. Consider, for instance, the case $c^2 > d_1d_2$, and $\dfrac{d_1}{c} < \dfrac{a_1T_2}{a_2T_1} < \dfrac{c}{d_2}$.

The temperature line runs as shown in Fig. 3 and crosses the stability ranges of the states A and C and that where both the states C and B satisfy the minimum conditions. In the first cases A and C the temperature dependence of the magnetization is described by (12) and (14), the second B or C corresponds to (13) or (14). On the grounds of (17) it

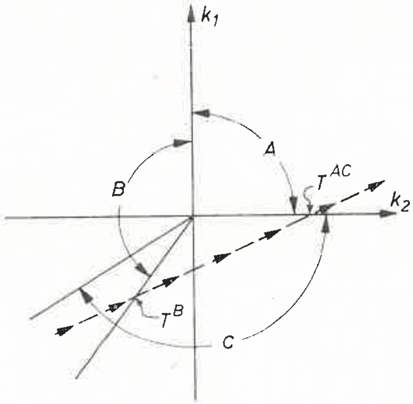

Fig. 3. Temperature line (dashed one) for $T_1 > T_2$, $\bar{\beta} < 0$, $\bar{\alpha} < 0$ in the phase diagram if $d_1 d_2 \leqslant c^2$. The arrows on the line denote the direction corresponding to an increase in temperature

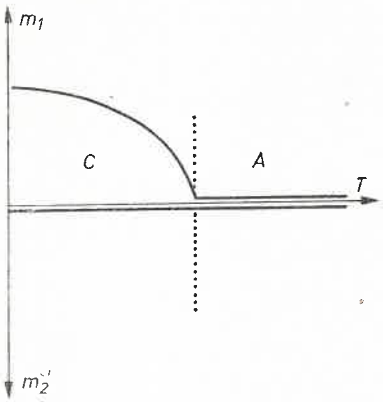

Fig. 4

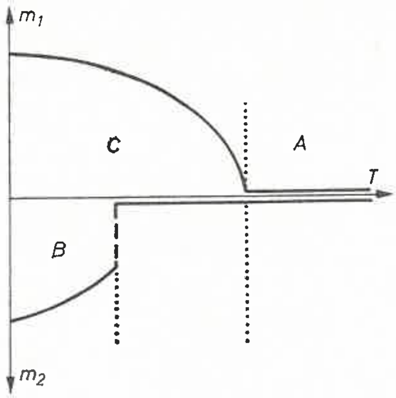

Fig. 5

Fig. 4. Temperature dependence of the magnetization components ($T_1 > T_2$) for $d_1 d_2 > c^2$ in the case of $\bar{\beta} < 0$ or $\bar{\beta} > 0, \beta < 0$ and for $d_1 d_2 < c^2$ in the cases: $\bar{\alpha} > 0$ or $\bar{\beta} < 0, \bar{\alpha} < 0, \alpha > 0$ or $\bar{\beta} > 0$, $\alpha > 0, \beta > 0$

Fig. 5. Temperature dependence of the magnetization vector ($T_1 > T_2$) for $d_1 d_2 < c^2$ in the cases: $\bar{\beta} < 0$, $\bar{\alpha} < 0$, $\alpha < 0$ or $\bar{\beta} > 0$, $\alpha < 0$, $\beta < 0$

can easily be noticed that $T^{B} > 0$ if $a_1 d_2 < c a_2$, and $T^{B} < 0$ if $a_1 d_2 > c a_2$. The latter result is not, of course, physical, and leads to the conclusion that the state B does not occur. Similar considerations and discussions concerning the relations between the temperatures which determine the stability ranges of the admissible states enable us to find the temperature dependence of the magnetization in all cases.

The results obtained this way are shown in Fig. 4—10. The corresponding analytical expressions are given by (12)—(15) when assigning the appropiate formula to the states marked in the figures. Attention should be paid to that for $c^2 > d_1 d_2$ there exists the range of coexistence of two different phases with the magnetization $(m_1, 0)$ or $(0, m_2)$.

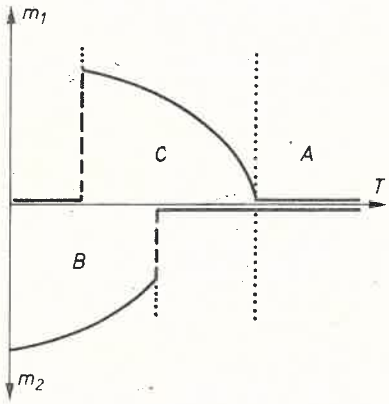

Fig. 6

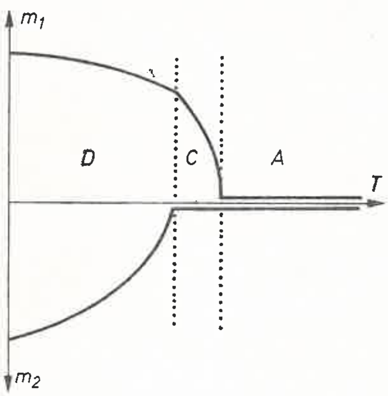

Fig. 7

Fig. 6. Temperature dependence of the magnetization vectors ($T_1 > T_2$) for $d_1 d_2 < c^2$ and $\bar{\beta} > 0$, $\alpha < 0$, $\beta > 0$

Fig. 7. Temperature dependence of the magnetization components ($T_1 > T_2$) for $d_1 d_2 > c^2$ and $\bar{\alpha} > 0$, $\bar{\beta} > 0$, $\beta > 0$

This is why the diagrams present the results for m_1 and m_2. On the other hand, if $c^2 < d_1 d_2$, m_1 and m_2 denote the components of the magnetization vector since the state with declined magnetization satisfies the minimum conditions.

It is worth noticing that a change in temperature at a given relation between the material constants can lead to an effective change in the direction of the easy axis. At

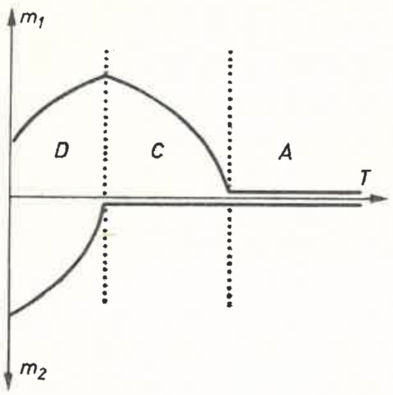

Fig. 8

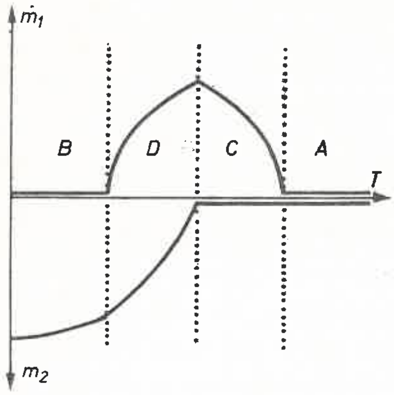

Fig. 9

Fig. 8. Temperature dependence of the magnetization components ($T_1 > T_2$) for $d_1 d_2 > c^2$ and $\bar{\alpha} < 0$, $\alpha > 0$, $\beta > 0$

Fig. 9. Temperature dependence of the magnetization components ($T_1 > T_2$) for $d_1 d_2 > c^2$ and $\bar{\alpha} < 0$, $\alpha < 0$, $\beta > 0$

a certain relation between the material constants metastable (overheated or supercooled) states can arise (see Fig. 5 and Fig. 6). In other cases continuous change takes place. Thus, both discontinuous and continuous phase transitions are admissible. They correspond to the first- and second-order phase transitions, respectively, since the Landau theory is applied here. As pertains to the continuous transitions the jump of the specific heat occurs in accordance with the old classification.

The results concerning the change in the direction of the effective easy axis correspond to those of [8]. The starting point of this paper was the free energy

$$F = F_0 + K_1 \sin^2 \psi + K_2 \sin^4 \psi,$$

minimized with respect to the angle ψ. It is of some importance that the minimization was carried out with respect to the single parameter solely and the temperature dependence of the anisotropy constants — being quite different from that of our paper — was assumed from experimental data.

4. Temperature dependence of the length of the magnetization vector

Based on the results of the previous sections it seems interesting to consider the change of the length of the magnetization vector in the deflection state D. For other states such results are, of course, presented previously ($m = m_1$ or $m = m_2$ for the states C or B, respectively). The state D occurs if $c^2 < d_1 d_2$ and $\bar{\alpha}T < \alpha$, $\bar{\beta}T < \beta$. For simplicity, we shall still assume $T_1 > T_2$. When introducing the notation (16) and taking advantage of (15), we obtain the following expressions for the magnetization length and cosines of the angle ψ that the magnetization vector forms with the axis parallel to the vector $(0, m_2)$.

$$2\varphi M^2 = (\bar{\alpha}+\bar{\beta})T - (\alpha+\beta), \tag{21}$$

$$\cos^2 \psi = \frac{\bar{\beta}T - \beta}{(\bar{\alpha}+\bar{\beta})T - (\alpha+\beta)}. \tag{22}$$

The first dependence is parabolic while the second one is a hyperbola in the coordinate frame $(\cos^2 \psi, T)$. The asymptotes of the latter curve are given by

$$(\cos^2 \psi)_{\rm as} = \frac{\bar{\beta}}{\bar{\alpha}+\bar{\beta}}, \qquad T_{\rm as} = \frac{\alpha+\beta}{\bar{\alpha}+\bar{\beta}}. \tag{23}$$

As seen from (21) at $T = T_{as}$ the magnetization length is equal to zero. By a simple analysis of (21) and (22) one can show that both the magnetization length and $\cos^2 \psi$ are decreasing function of T if $\bar{\beta} > 0$ and $\bar{\alpha} > 0$ or $\bar{\beta} > 0$, $\bar{\alpha} < 0$ and $|\bar{\alpha}| < \bar{\beta}$. Otherwise, the magnitude of the magnetization increases with temperature (if $\bar{\beta} > 0$, $\bar{\alpha} < 0$ and $|\bar{\alpha}| > \bar{\beta}$) or independent of T (if $\bar{\beta} > 0$, $\bar{\alpha} < 0$ and $|\bar{\alpha}| = \bar{\beta}$) while $\cos^2 \psi$ is still decreasing (linearly if $|\bar{\alpha}| = \bar{\beta}$). In the first case $\cos^2 \psi$ is a convex function of T while in the second case it is a concave function. The obtained results are illustrated in Figs 10—11, which include all the possible cases following from our free energy. The results shows in Figs 10—11 seem particularly interesting and unexpected for they correspond to an increasing dependence of the magnetization length on T. Therefore, we also examined the behaviour of the entropy in the state under consideration. By inserting (10) and (15) to (1) and taking the temperature derivative we have

$$-2(c^2-d_1d_2)S = \left[d_2\left(\frac{a_1}{T_1}\right)^2 - 2c\left(\frac{a_1}{T_1}\right)\left(\frac{a_2}{T_2}\right) + d_1\left(\frac{a_2}{T_2}\right)^2\right]T$$

$$-\left(\frac{a_1}{T_1}\right)(a_1d_2-ca_2)-\left(\frac{a_2}{T_2}\right)(a_2d_2-ca_1) \qquad (24)$$

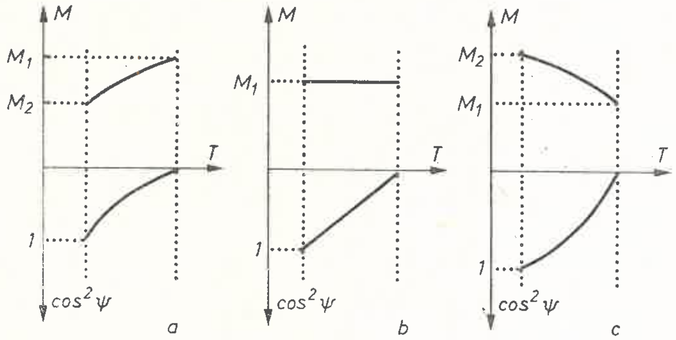

Fig. 10. Temperature dependence of the magnetization length M and angle ψ that the magnetization forms with the axis $(0, m_2)$ in the state D if $\bar{\beta} > 0, \alpha > 0, \beta > 0$ in the cases: a. $\bar{\alpha} < 0, \bar{\beta} < |\bar{\alpha}|$; b. $\bar{\alpha} < 0, \bar{\beta} = |\bar{\alpha}|$; c. $\bar{\alpha} < 0$, $\bar{\beta} > |\bar{\alpha}|$ or $\bar{\alpha} > 0$. We use the notation $\gamma = \frac{\beta}{\alpha+\beta}$, $M_1^2 = \frac{\bar{\alpha}\beta-\bar{\beta}\alpha}{2\varphi\bar{\beta}}$, $M_2^2 = -\frac{\alpha+\beta}{2\varphi}$

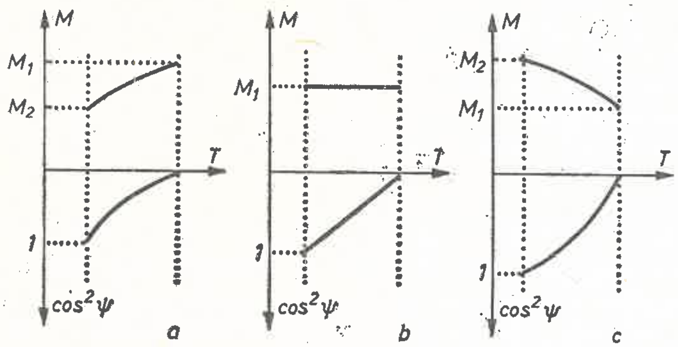

Fig. 11. Temperature dependence of the magnetization length M and angle ψ that the magnetization forms with the axis $(0, m_2)$ in the state D if $\bar{\beta} > 0, \bar{\alpha} < 0, \alpha < 0, \beta > 0$ in the cases: a. $|\bar{\alpha}| > \bar{\beta}$; b. $\bar{\beta} = |\bar{\alpha}|$; c. $|\bar{\alpha}| < \bar{\beta}$. We denote $M_1^2 = \frac{\bar{\alpha}\beta-\bar{\beta}\alpha}{2\varphi\bar{\beta}}$, $M_2^2 = \frac{\bar{\beta}\alpha-\bar{\alpha}\beta}{2\varphi\bar{\alpha}}$

for the entropy S of the deflection state D. The coefficient of T determines their behaviour under the change of temperature. The coefficient is the quadratic form with respect to a_1/T_1 and a_2/T_2, and its determinant (equal to $d_1d_2-c^2$) is positive for the state D (see (6)). Thus, the entropy is an increasing function of T without respect to the particular dependence of the magnetization length on temperature.

5. *Concluding remarks*

As seen from our results, the assumed free energy [4, 5] leads to various behaviour of the temperature dependence of the magnetization depending on mutual relations of the material constants. The fact that variations of the temperature cause changes in the direction of the effective easy axis is characteristic of this behaviour. This change manifests itself as a jump (first-order phase transition) or undergoes in a continuous way (second--order phase transition). It seems interesting to consider again the origin of the assumed free energy, which is a consequence of the compatibility between the exchange and crystal-

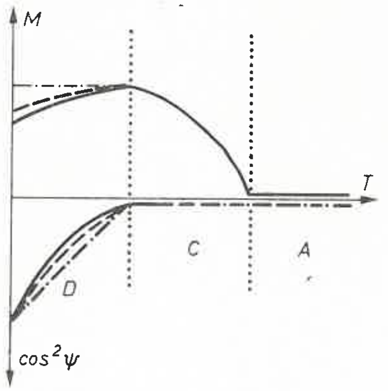

Fig. 11

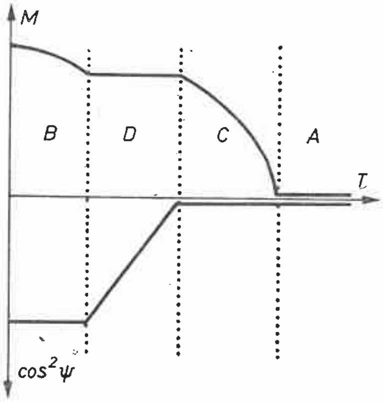

Fig. 12

Fig. 12. Temperature dependence of the magnetization length M and angle ψ that the magnetization forms with the axis $(0, m_2)$ if $d_1d_2 > c^2$, $\bar{\alpha} < 0$, $\bar{\beta} > 0$, $\alpha > 0$, $\beta > 0$ and $\bar{\beta} \leqslant |\bar{\alpha}|$. The three cases are marked with: full line (δ_1), dashed one (δ_2) and dash-dotted line (δ_3, $\beta = |\alpha|$). where $\delta_i = \alpha_i + \beta_i$ and $\delta_1 > \delta_2 > \delta_3$

Fig. 13. Temperature dependence of the magnetization length M and angle ψ that the magnetization forms with the axis $(0, m_2)$ if $d_1d_2 > c^2$, $\bar{\alpha} < 0$, $\alpha < 0$, $\beta > 0$ and $\bar{\beta} = |\bar{\alpha}|$

-field interactions. One of these interactions create the easy axis while the second causes the perpendicular easy plane to arise. Thus, as evidently seen from our results, these interactions depend in an effectively different way on the temperature. Upon assuming such a physical conclusion, the results concerning the temperature change of the effective easy direction and an increase of the magnetization length with temperature do not seem unexpected. It seems also that this is how one can interpret experimentally found cases of an increase of the magnetization in certain magnetic materials.

At appropriate choice of the material constants c, d_i, T_i, a_i the presented results permit one to obtain a large variety of the dependence of the magnetization length on the temperature, Figs 12 and 13 illustrate two of these possibilities, which seem particularly interesting to us. The experimental results of [1], for instance, are in an eventual correspondence to the case shown in Fig. 12.

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