Spin-reorientation transitions in rare-earth magnets

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This review treats the little-studied magnetic phase transitions of the order-order, spin-reorientation type, in which a rotation of the magnetization vector (or of the antiferromagnetism vector) with respect to the crystallographic axes occurs upon change of temperature, magnetic field, or external pressure. It presents theoretical ideas about magnetic phase transitions of the spin-reorientation type; it considers the effect of domain structure on spin-reorientation phase transitions, and critical fluctuations during such transitions. Taking as an example rare-earth magnets (orthoferrites, iron-garnets, and rare-earth metals and intermetallic compounds), it considers the experimental data on various types of spin-reorientation transitions and on the anomalies of physical properties at such transitions.

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INTRODUCTION

Magnetic phase transitions of the order-disorder type, resulting from destruction of the ferromagnetic or antiferromagnetic ordering,[1] are now well known and are being actively investigated. Investigation of such transitions is important both for the theory of magnetism and for the general theory of the condensed state of matter. It is exerting an important influence on the development of the theory of phase transitions in solids and liquids. This is evidenced by the fact that there have recently appeared a number of monographs in which magnetic transitions of the order-disorder type and critical phenomena in a liquid-vapor system are treated together from a single point of view.

But in addition to the study of such magnetic transitions, there has recently developed great interest in the study of magnetic phase transitions of the order–order type, in which a change occurs in the type of magnetic structure. Such transitions include, for example, transformations between ferromagnetism and antiferromagnetism, between collinear antiferromagnetism and noncollinear antiferromagnetism, etc. Transitions of this type are very common, and they are very often responsible for anomalous behavior of the magnetic and other properties of magnetically ordered materials. The causes and the character of magnetic order–order phase transitions may be very diverse (several types of such transitions, particularly those caused by change of sign of the exchange integral and by change of the electronic energy spectrum, are considered in the review article[2]). But from a phenomenological point of view it is possible to treat them together, by taking into account that in such transitions one of the magnetic structures always loses its stability and another becomes stable, and thus the magnetic symmetry changes. From this point of view, magnetic phase transitions of the order–order type are very reminiscent of the well-studied structural transitions in crystals, which are characterized by the fact that during the transition

there occurs a deformation of the crystal, accompanied as a rule by a change of its crystalline symmetry. Therefore magnetic transitions of the order–order type are sometimes called magnetostructural phase transitions.

Among the diverse magnetostructural phase transitions, it is possible to separate out the spin-reorientation phase transitions. These transitions are characterized by the fact that upon change of the external parameters (temperature, magnetic field, pressure), a change occurs in the orientation of the magnetic moments with respect to the crystallographic axes. The simplest example of spin-reorientation transitions is a phenomenon often observed in various ferromagnets: upon change of temperature, the direction of easy magnetization changes—there occurs a rotation of the magnetization vector of the crystal as a whole from one direction to another. Spin-reorientation transitions are characterized by an ordering parameter θ, the angle of rotation of the magnetic moment with respect to the crystal axes. These transitions may be either phase transitions of the first kind, or of the second. In both cases the magnetic symmetry changes discontinuously at the transition point; but in spin-reorientation transitions of the first kind there occurs a discontinuous change of the ordering parameter θ, while in spin-reorientation transitions of the second kind θ changes gradually.

The present review summarizes and generalizes the information available in the literature on spin-reorientation phase transitions in rare-earth magnets. These materials have been chosen as object of investigation because in rare-earth magnets a large number of various types of transitions are observed, encompassing essentially all the diverse phase transitions of the spin-reorientation type. Principal attention is given to the spontaneous spin-reorientation transitions.

Also considered are spin-reorientation transitions in a magnetic field, principally in the example of the rare-earth orthoferrites. We do not discuss the field-induced transitions, observed in rare-earth ferrite-garnets, from a collinear ferromagnetic to a noncollinear state, since they have been recently elucidated in the reviews.[3]

1. MAGNETIC PHASE DIAGRAMS

We shall consider spin-reorientation transition on the basis of Landau's theory. Within the framework of this theory, for the description of magnetic structures one usually introduces a certain "state vector" $\mathbf{m}$: that is, the set of the magnetic moments that are necessary for a macroscopic description of the thermodynamic properties of the system. We shall not detail the form of $\mathbf{m}$, since the procedure for choice of a concrete form of $\mathbf{m}$ is well known.[4] We shall mention only that in the role of $\mathbf{m}$, in ferromagnets one usually chooses the magnetization, in antiferromagnets the magnetic moments of the sublattices or the magnetization and the antiferromagnetism vector, in ferrimagnets the magnetic moments of the sublattices, etc. Landau's theory assumes an expansion of the thermodynamic potential (TP) of the system, Φ, in components of the vector $\mathbf{m}$; the potential of the paramagnetic phase is taken as the zero-order approximation.

It is convenient to expand Φ in irreducible representations of the symmetry group of the paramagnetic phase. Minimization of the TP $\Phi(\mathbf{m}, H, T)$ with respect to $\mathbf{m}$ gives the possible set of states of the magnetic system (phases) $\mathbf{m}_n(H, T)$, which are stable in the corresponding ranges of the external parameters (H, T); that is, in the corresponding sections of the (H, T) phase diagram. The possible phases $\mathbf{m}_n(H, T)$ can be separated into two types. In phases of the first type, $\mathbf{m}(H, T)$ does not change upon change of H and T. Here the spins are usually directed along definite crystallographic directions. We shall call such phases "collinear." In phases of the second type, a continuous change of $\mathbf{m}_n$ occurs with change of the external parameters; we shall call them "angular" (canted).

Certain phases may be "degenerate"; this means that there is a set of solutions $m_n^\alpha(H, T)$ $(\alpha = 1, 2, \ldots)$ to which correspond identically equal values of the TP. To the presence of "degeneracy" are related certain symmetry elements of the paraphase that are absent in a phase $m_n^\alpha(H, T)$ but, acting on solutions with different α, convert them one to another (as one says in this case, "re-establish the symmetry"); "degeneracy" leads to a distinctive domain structure.

We shall discuss the basic features of the magnetic phase diagrams of magnetic materials that undergo spin-reorientation transitions. We shall restrict ourselves for simplicity to the case in which the state vector $\mathbf{m}$ can be described by two angles θ and φ.[1)]

We shall also assume that the modulus of the vector $\mathbf{m}$ does not change during the transition. The possible phases $\{\theta, \varphi\}$ are determined as the solutions of the equations

$$\frac{\partial\Phi}{\partial\theta}=0, \quad \frac{\partial\Phi}{\partial\varphi}=0. \tag{1.1}$$

Each phase has a definite region of existence (stability) on the H, T plane. The fundamental lines and points that separate them on the diagram are determined as follows (for characteristic examples of diagrams, see below).

a) Let there be a solution of the problem of minimization of the TP: $\theta = \theta^1_{\text{coll}}$, $\varphi = \varphi^1_{\text{coll}}$ (collinear phase). This phase is stable in a certain region on the H, T plane, in which the conditions

$$\frac{\partial^2\Phi}{\partial\theta^2}>0, \quad \frac{\partial^2\Phi}{\partial\theta^2}\frac{\partial^2\Phi}{\partial\varphi^2}-\left(\frac{\partial^2\Phi}{\partial\theta\,\partial\varphi}\right)^2>0. \tag{1.2}$$

are satisfied. Vanishing (with subsequent change of sign) of one of these expressions is the condition for loss of stability of the phase. It determines a critical line on the H, T plane, which may be a line of phase transition of the second kind or a line of loss of stability of a metastable phase. The type of transition can be established by analysis of the higher derivatives of

[1)] This restriction is not very severe. We shall see below that it is fulfilled in a broad class of magnetic materials.

the function $\Phi(\theta, \varphi)$ or by consideration of the region of existence of the neighboring phase.

b) It is obvious that a line separating two different collinear phases is a line of phase transition of the first kind. It is determined by the condition

$$\Phi(\theta_1, \varphi_1) = \Phi(\theta_2, \varphi_2). \quad (1.3)$$

c) The boundary between collinear and angular phases may be a line of phase transition either of the first or of the second kind. For analysis of the angular phase, it is sometimes convenient to consider the equation that determines it, $\theta = \theta(H, T)$,[2)] as the equation of a one-parameter family of curves with $\theta = \text{const}$. If the envelope of such a family, i.e., a certain boundary of the angular phase, is a line with $\theta = \theta_{\text{coll}}$ and $\varphi = \varphi_{\text{coll}}$, then it is a line of phase transition of the second kind. But if the envelope does not belong to the family of curves, then in this case the ranges of existence of the angular and collinear phases overlap; that is, between them a phase transition of the first kind occurs. The transition line is determined by an equation of the type (1.3) with appropriate values of the phases $\{\theta_{\text{coll}}, \varphi_{\text{coll}}\}$ and $\{\theta_{\text{ang}}(H, T), \varphi_{\text{ang}}\}$. The boundary of the angular phase in this case (envelope of the family) is the line of loss of stability of the angular phase and is determined by the following equations (by elimination of $\theta_{\text{ang}}(H, T)$):

$$\left.\frac{\partial \Phi}{\partial \theta}\right|_{\theta_{\text{ang}}, \varphi} = 0. \quad \left.\frac{\partial^2 \Phi}{\partial \theta^2}\right|_{\theta=\theta_{\text{ang}}, \varphi} = 0. \quad (1.4)$$

A line of second order phase transition between collinear and angular phases may at a certain point change to a line of the first order (such a point is sometimes called tricritical).

d) Within a region of existence of an angular phase, there may be a region where the function $\theta(H, T)$ becomes nonunique; that is, in this region Eq. (1.1) has two stable solutions (for given φ). Consequently, in this region there coexist two angular phases $\{\theta_{\text{ang}}^{(1)}(H, T), \varphi\}$ and $\{\theta_{\text{ang}}^{(2)}, \varphi\}$ with unequal values of the TP. It is obvious that within a region of coexistence there is a line of phase transition of the first kind between the phases $\{\theta_{\text{ang}}^{(1)}, \varphi\}$ and $\{\theta_{\text{ang}}^{(2)}, \varphi\}$, determined by an equation of the type (1.3) with appropriate values of the phases. The boundaries of the region of coexistence are the lines of loss of stability of the metastable angular phases. They are determined by equations of the type (1.4), where the appropriate metastable angular phase enters in the role of $\theta(T, H)$. The lines of loss of stability of two metastable angular phases may come together at some point (there, obviously, a line of phase transition of the first kind ends). This point is an ordinary critical point (the "vapor-liquid" type of critical point).

On orientational transition lines, the physical parameters experience characteristic singularities. As will be shown later (Chap. 3), orientational transitions are an example of transitions for which Landau's theory (or theories equivalent to it, for example molecular-field theory) is a very good approximation (it is inapplicable only in an extremely narrow range near a transition point of the second kind or other critical point). Within the framework of this theory, the singularities of thermodynamic quantities near orientational transition lines are completely determined by the TP introduced above. In practical calculations, it is convenient to expand the TP Φ near the transition line as a series in $\Delta\theta$, where $\Delta\theta$ is the deviation of the angle θ from its equilibrium value in the collinear phase. This is the expansion, typical of Landau's theory, in powers of the order parameter $\eta = \Delta\theta$:

$$\Phi = \alpha(\Delta\theta)^2 + \gamma(\Delta\theta)^3 + \beta(\Delta\theta)^4 + \ldots,$$

where $\alpha = 0$ on the critical line. It is obvious that for description of the behavior of thermodynamic quantities during the transition, the formulas of Landau's theory [4] may be used.

The role of order parameter in orientational transitions is played by the angle $\Delta\theta$. It is interesting to establish the relation of this "formal" order parameter to the order parameter that figures in Landau's general theory of phase transitions, and whose introduction is justified on the basis of the theory of symmetry. In passing, we shall consider certain symmetry aspects of orientational phase transitions.

We shall investigate this question in a simple example that reflects the basic features of many orientational transitions. We consider a ferromagnet of rhombic symmetry, in which the reorientation of the magnetization occurs in the x, z plane. The considerations advanced below are fully applicable also to a uniaxial or hexagonal magnet. In the role of state vector, it is natural to choose a unit vector directed along the magnetic moment, $\mathbf{m} = (m_x, m_y, m_z)$.[3)] The TP can be represented in the form

$$\Phi = \frac{A_1}{2} m_x^2 + \frac{A_2}{2} m_z^2 + \frac{B_1}{4} m_x^4 + \frac{B_2}{4} m_z^4 + \frac{B_4}{4} m_x^2 m_z^2. \quad (1.5)$$

We have set $m_y = 0$, since we are interested only in reorientation in the x, z plane. Minimization of this TP gives three possible phases: I) $m_x = 0$, $m_z = \pm 1$; II) $m_x = \pm 1$, $m_z = 0$; III) $m_x \neq 0$, $m_z \neq 0$. Phases I and II are collinear, whereas phase III is angular.

It is obvious that within the framework of the Landau theory, which starts from an expansion of the TP in invariant combinations of the components of the vector $\mathbf{m}$ with respect to the initial paramagnetic phase (paraphase), transitions of the type I – III and II – III must be described by the two-dimensional order parameter (m_x, m_z). We note that it transforms according to a two-dimensional reducible representation of the paraphase. The transition from the two-dimensional order to the "formal" angle θ is obvious; the substitution $m_x = \sin\theta$, $m_z = \cos\theta$ reduces the TP to the form[4)]

[2)] We set here $\varphi = \text{const}$, which is usually fulfilled (at least this is so in all the magnetic transitions considered below).

[3)] In the case of reorientation in orthoferrites or of a transition of the spin-flop type in antiferromagnets, the TP can be represented in the form (1.5), but in it $\mathbf{m}$ must be replaced by the antiferromagnetism vector.

[4)] Horner and Varma[5] first pointed out that such a TP describes two phase transitions of the second kind (for $K_2 > 0$), close together in temperature.

$$\Phi = K_1 \sin^2\theta + K_2 \sin^4\theta,$$

where K_1 and K_2 have the meaning of anisotropy constants. Two cases are possible, differing with respect to the character of the phase diagrams.

Case 1: $K_2 > 0$. Phases I–III have the following ranges of existence:

$$\left.\begin{array}{lll} \text{Phase I:} & \theta = 0,\ \pi, & \text{stable for } K \geqslant 0, \\ \text{Phase II:} & \theta = \frac{\pi}{2},\ \frac{3\pi}{2}, & \text{stable for } K_1 + 2K_2 \leqslant 0, \\ \text{Phase III:} & \sin^2\theta = -\frac{K_1}{2K_2}, & \text{stable in the interval } K_1 \leqslant 0, \\ & & K_1 + 2K_2 \geqslant 0. \end{array}\right\} \quad (1.7)$$

Equalities in the relations (1.7) correspond to lines of loss of stability of the various phases. Usually such phase transitions are accomplished upon change of sign of the anisotropy constant $K_1(T)$. A monotonic change of temperature leads to a continuous reorientation of the vector **m** with two phase transitions of the second kind, I – III – II. The transition I – III occurs at $T = T_1$, where $K_1(T_1) = 0$; the order parameter is the angle, i.e., $\eta = \theta$ (or $\pi - \theta$). The transition II – III occurs at $T = T_2$, determined by the relation $K_1(T) + 2K_2 = 0$. Here the order parameter is $\eta = (\theta - \pi/2)$ or $\eta = (\theta - 3\pi/2)$. It is easy to see that near the transition point the expansion of the TP (1.6) with respect to η takes the standard form $\Phi = \alpha\eta^2 + \beta\eta^4$, where α vanishes at the appropriate transition point, whereas $\beta > 0$.

Case 2: $K_2 < 0$. Here phase III is unstable. The TP (1.6) describes a phase transition of the first kind I – II at $T = T_0$, where T_0 is determined by the condition for equality of the potentials of the two phases, $K_1(T_0) = -K_2$. The temperatures T_1 and T_2, determined by the equalities $K_1(T_1) = 0$ and $K_1(T_2) + 2K_2 = 0$, are the boundaries of the region of existence of the metastable states (on the assumption that the transition I – II occurs uniformly over the specimen).

The transitions considered are typical of transitions involving destruction of the magnetic symmetry of the crystal. In fact, in the first case, for example, during the transition I – III there is destruction of the symmetry with respect to rotation about the z axis through angle π (C_2^z). In phase III this symmetry element is absent, but there is a doubled number of solutions of the problem of minimization of the TP (1.5) as compared with phase I: (m_x, m_z) and $(-m_x, m_z)$, which are converted one to the other by the "destroyed" symmetry element. To these two solutions (two phases) corresponds a single energy ("degeneracy"). Similar arguments are applicable also to the transition II – III, where the role of "destroyed" symmetry element is played by rotation about the x axis. The picture considered, of destruction of symmetry and occurrence of degeneracy in the angular phase, leading to a domain structure, is typical for all continuous orientational transitions. We note that in continuous reorientation via two phase transitions, I – III – II, the angular phase III plays a "buffer" role: it enables the vector **m** to change continuously from phase I to phase II. From the symmetry point of view, a direct continuous transition I – II is impossible: in fact, for a continuous transition it is necessary that the magnetic symmetry group of one of the phases participating in the transition shall be a symmetry subgroup of the other phase[4]; it is obvious that for phases I and II this requirement is not fulfilled. A direct transition I – II can be only a phase transition of the first kind, as actually occurs when $K_2 < 0$.

2. SPIN-REORIENTATION TRANSITIONS AND DOMAIN STRUCTURE

All the considerations presented above are applicable to spatially homogeneous systems. The role of domain structure is especially important in magnetic materials, we shall consider the basic features of domain-structure behavior in transitions and shall analyze its influence on the character of a transition. It is convenient to investigate this question for the example analyzed above of a rhombic magnet. Here also it is necessary to consider two cases: $K_2 > 0$ and $K_2 < 0$.

Case 1: $K_2 < 0$. In a single-domain specimen this case, as we showed in Chap. 1, corresponds to a phase transition of the first kind with appropriate regions of existence of the metastable phases. In the presence of domain structure, the domain walls may serve as nuclei of the new phase, because in them there are always sections in which the direction of the magnetic moments coincides with the direction of the magnetic moments in the new phase. This leads to a change of character of the transition; in particular, to the result that the transition may occur without hysteresis.

We shall consider this question in more detail. For this purpose it is necessary to investigate the change of a domain wall during the transition. The TP with allowance for the energy of the magnetic inhomogeneities has the form

$$\Phi = \int \left[A \left(\frac{d\theta}{dy}\right)^2 + K_1(T)\sin^2\theta + K_2 \sin^4\theta \right] dy, \quad (2.1)$$

where A is the exchange constant. Euler's equation of the variational problem for this functional,

$$A\frac{d^2\theta}{dy^2} = K_1(T)\sin\theta\cos\theta + 2K_2\sin^3\theta\cos\theta + C \quad (2.2)$$

has the first integral

$$A\left(\frac{d\theta}{dy}\right)^2 = K_1(T)\sin^2\theta + K_2\sin^4\theta + C, \quad (2.3)$$

where C is an undetermined constant, determined by the boundary conditions.

For qualitative explanation of the behavior of the domain structure during a transition, it is sufficient to analyze the phase portrait of this equation: that is, the (θ', θ) plane, on which each solution corresponds to a certain trajectory determined by (2.3). Of all the solutions of (2.2), the most interesting are the solutions that describe a domain wall. To such solutions correspond the trajectories on the phase portrait that are called separatrices. They are shown in Fig. 1a. The character of the domain walls for a similar problem has been studied in more detail in[6,7]. From Fig. 1a it is evident that for $T > T_2$ the separatrix comes out of the saddle point $\theta' = 0$, $\theta = 0$ and goes into the saddle point $\theta' = 0$, $\theta = \pi$: it describes an ordinary 180° domain

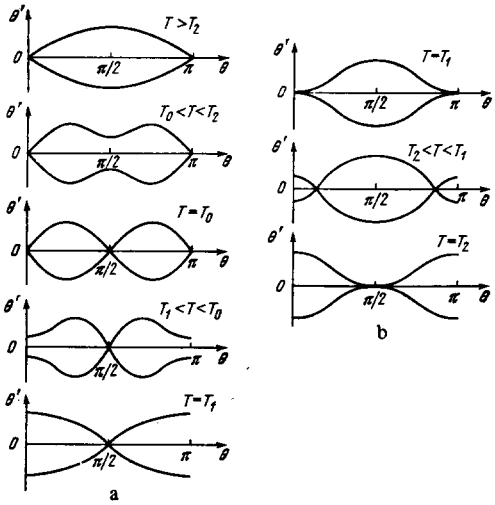

FIG. 1. Phase portraits of equation (1.9), illustrating the nature of the deformation of the domain structure during spin-reorientation phase transitions. a, phase transition of the first kind; b, continuous reorientation.

wall.[5)] For $T_2 < T < T_0$, extrema develop on the separatrix at the points $\theta = \pi/2$, $\theta = 3\pi/2$; this means that at the center of the domain walls kinks are developing, which "grow" with approach to $T = T_0$. For $T = T_0$ the kinks are transformed to domains of the new phase; that is, at this temperature a 90° domain structure originates. For $T_1 < T < T_2$, the separatrix "disengages" from the points $\theta = 0$, $\theta = \pi$. This means that the "old" domains (phase I) are transformed to kinks on 180° domain walls of the new phase; the kinks gradually decrease as $T \to T_1$ and disappear for $T < T_1$. From the mathematical point of view, the temperature T_0 of the phase transition is a point of bifurcation of the nonlinear differential equation (2.2). This model shows that an orientational transition of the first kind may be anhysteretic by virtue of a continuous restructuring of the domain walls, which play the role of nuclei of the new phase. Such a mechanism of formation of nuclei is very peculiar in that the nuclei begin to grow actually "within the depths" of the old phase; that is, before the system reaches the temperature of the phase transition. This fact is the reason for the anhystereticity. This type of anhysteretic transition was investigated experimentally and theoretically in[8] for the example of a transition of the Morin type in $DyFeO_3$ with Co^{2+} impurity. Similar transitions have also been detected in cubic ferrimagnets.[9]

Case 2: $K_2 > 0$, continuous reorientation. Certain peculiarities of behavior of the domain structure during the transition already follow from symmetry considerations. In phase I there are two energetically equivalent solutions $m_x = 0$, $m_z = \pm 1$ (or $\theta = 0, \pi$). This means that there can exist an "ordinary" domain structure with 180° domain walls. In phase III the number of solutions doubles: $\theta = 0$ is transformed to the two solutions $\theta_{1,2} = \pm\theta_0 = \pm \arcsin(\sqrt{-K_1/2K_2})$, and $\theta = \pi$ is transformed to $\theta_{1,2} = \pi \pm \theta_0$. This leads to the result that a domain of phase I with $\theta = 0$ divides into two domains with $\theta_1 = \theta_0$ and $\theta_2 = -\theta_0$; similarly, a domain with $\theta = \pi$ is transformed to domains $\theta_3 = \pi - \theta_0$ and $\theta_4 = \pi + \theta_0$: that is, the number of domains in phase III, as compared with phase I, is doubled. The new domains in phase III are separated by two forms of walls (not of 180° type). In the transition from phase III to II, a joining of solutions occurs, and consequently also a joining of domains θ_1, θ_3 into a domain of phase II $\theta = \pi/2$, and of domains θ_2, θ_4 into a domain $\theta = 3\pi/2$; that is, an "ordinary" domain structure is established, but one turned through $\pi/2$. Figure 1b shows the phase portraits of equation (2.2) for $K_2 > 0$. It is evident that they are in complete agreement with the picture presented above. Here also the temperatures of the phase transitions are points of bifurcation of the equation that determines the domain structure. We note that in the angular phase III the domain structure is still retained in a magnetic field sufficiently strong to suppress it in phases I and II. This also is a manifestation of the "degeneracy" inherent in the angular phase. All these processes have been treated in more detail in[10].

3. CRITICAL FLUCTUATIONS IN SPIN-REORIENTATION TRANSITIONS

A large portion of the theoretical work devoted to spin-reorientation transitions has been done within the framework of Landau's theory (or of approximations equivalent to it). Landau's theory (and also molecular-field theory), in constructing the TP of the system, neglects in a well-known sense the fluctuations of the order parameter. Near the transition temperature, i.e., near the point where the system loses its stability, the fluctuations increase greatly, and these theories become inadequate. A characteristic peculiarity of spin-reorientation transitions is the fact that for them the description by Landau's theory is applicable practically without limitation (the range of inapplicability becomes extremely narrow, $\Delta T \sim 10^{-4}$–10^{-7} °K). This is a consequence of the fact that the fluctuations that occur near the transition have a very large correlation radius.[11,12]

We shall consider this question for the example, investigated above, of an orientational transition in magnets of rhombic symmetry. Serving as a basis for a phenomenological study of critical fluctuations will be a thermodynamic potential of type (2.1), which allows for the energy of magnetic inhomogeneities. For small θ we have

$$\Phi = \int [K\tau\theta^2 + K_2\theta^4 + A\,(\mathrm{grad}\,\theta)^2]\,dV, \qquad (3.1)$$

where the anisotropy constant $K_1(T)$ is represented in the form $K_1(T) = K\tau$, $\tau = (T - T_1)/T_1$.

Let the fluctuation of the angle θ be $\delta\theta(\tau)$ and its Fourier transform $\delta\theta_q$. It is known that the mean square of the fluctuation $\delta\theta_q$ is[4]

$$\overline{|\delta\theta_q|^2} = \begin{cases} \dfrac{T}{V[K\tau + Aq^2]}, & \tau > 0, \\ \dfrac{T}{V[2K\tau + Aq^2]}, & \tau < 0. \end{cases} \qquad (3.2)$$

[5)] Saddle points of the separatrix (where $\theta' = 0$) describe domains, whereas the line between them describes a domain wall.

Inverse Fourier transformation of formula (3.2) gives the correlation function of the fluctuations of the order parameter,

$$g(r)=\overline{\delta\theta(0)\,\delta\theta(r)}=\frac{T}{4\pi A\tau}\,e^{-r/\rho}, \tag{3.3}$$

where

$$\rho=\begin{cases}\sqrt{\frac{A}{K\tau}}, & \tau>0,\\ \sqrt{\frac{A}{2K\tau}}, & \tau<0,\end{cases}$$

is the correlation radius, which approaches ∞ at the phase-transition point.

Unlimited growth of the homogeneous fluctuations on approach to the phase-transition point indicates inapplicability of Landau's theory in the immediate vicinity of that point. In[13,14] a criterion was proposed for determining the range of applicability of Landau's theory; it is conveniently expressed in the form

$$\sum_{q}^{q_{max}}\overline{|\delta\theta_q|^2}\ll\theta_0^2, \tag{3.4}$$

where

$$q_{max}=\frac{2\pi}{\rho},\quad \theta_0=\sqrt{\frac{2K|\tau|}{K_2}}.$$

On substituting the appropriate values in (3.4) and integrating, we get

$$\tau\gg\xi=\frac{T_1^2K_2^2}{(4\pi)^2A^3K}. \tag{3.5}$$

Estimation of the value of ξ according to this formula; for the typical parameter values $K_2/K_1\sim 0.1$, $T_1\sim 300$°K, $A\sim 10^{-7}$ erg/cm, $K\sim 10^4$ erg/cm^3, gives $\xi_K\sim 10^{-6}$. For comparison, we cite the value of the corresponding quantity for the transition at the Curie point: $\xi_K\sim 10^{-1}-10^{-2}$. We cite also characteristic values of the correlation radii for orientational transitions, ρ_{ot}, and for transitions at the Curie point, ρ_C (for the same parameters):

$$\rho_{ot}\sim 3\cdot 10^3\tau^{-1/2}\ \text{Å},\quad \rho_C\sim(3\text{—}5)\,\tau^{-1/2}\ \text{Å}.$$

Figure 2 shows the spectral density of the fluctuations $|\delta\theta_q|^2$ and, for comparison, the corresponding value of $|\Delta m_q|^2$ for critical fluctuations in the Curie-point region. It is evident that the "orientational" fluctuations are much longer-wave than the fluctuations in the Curie-point region. This explains, in particular, their small contribution to the thermodynamic potential of the system (the statistical weight of long-wave fluctuations is small, and the short-wave fluctuations are strongly suppressed).

The strong diminution of the role of critical fluctuations in the thermodynamics of orientational transitions by no means implies that critical fluctuations are altogether unimportant in orientational transitions. Certain phenomena (for example, scattering of light) are determined by long-wave fluctuations; therefore they may have significant anomalies during orientational transitions. The intensity of scattered light is directly determined by the value of $|\delta\theta_q|^2$ (for details see[15]):

$$I\sim\overline{|\delta\theta_q|^2}\sim\frac{1}{\rho^{-2}(\tau)+4k^2\sin^2(\theta/2)}, \tag{3.6}$$

where $\rho(\tau)$ is the correlation radius according to (3.3), k is the wave vector of the incident light, and θ is the scattering angle. This formula clearly reflects the basic property of critical "orientational" fluctuations—the large sizes of their correlation radius. In light scattering this manifests itself in the narrow diagram of the direction of scattered light (large scattering intensity at small angles) and in the comparatively broad range of temperatures in which the critical fluctuations are "felt."[6)]

The "long-wave" nature of critical fluctuations in orientational transitions may be expressed also in scattering of waves of other nature, for example sound. For sufficiently intense scattering of sound it is necessary that the correlation radius ρ of the fluctuations shall be comparable with the wavelength of the sound. In the case of orientational transitions, this can be achieved by moderately close approach to the transition point.

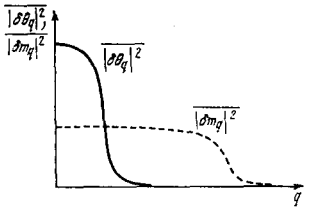

FIG. 2. Spectral density of critical fluctuations during orientational transitions and in the Curie-point region.

4. SPONTANEOUS SPIN-REORIENTATION TRANSITIONS IN RARE-EARTH ORTHOFERRITES

It is in rare-earth orthoferrites that a systematic and detailed investigation was first undertaken into the nature and character of the various phase transitions caused by spin reorientation.

The crystalline symmetry of the orthoferrites $RFeO_3$ (R is an ion of a rare-earth element) is described by the rhombic space group D_{2h}^{16}–$Pbnm$.[17] In the elementary cell there are four iron ions and four rare-earth ions. At high temperatures, an ordered antiferromagnetic structure is formed solely by the spins of the iron ions, which according to neutron-diffraction data[18] have a spin configuration of the G type, $+-+-$ (checkerboard order), with a transverse weak ferromagnetic moment F.

To find the possible orientations of the spins and of the weak ferromagnetic moment with respect to the crystallographic axes, we write the TP of the orthoferrites, in the absence of an external magnetic field, in the form

$$\Phi=\tfrac{1}{2}AF^2+\tfrac{1}{2}b_1G_x^2+\tfrac{1}{2}b_3G_z^2+\tfrac{1}{4}e_1G_x^4+\tfrac{1}{4}e_2G_x^2G_z^2+\tfrac{1}{4}e_3G_z^4+d_1G_zF_x-d_3G_xF_z, \tag{4.1}$$

where

$$\mathbf{F}=\frac{\mathbf{M}_1+\mathbf{M}_2+\mathbf{M}_3+\mathbf{M}_4}{4M_0},\qquad \mathbf{G}=\frac{\mathbf{M}_1-\mathbf{M}_2+\mathbf{M}_3-\mathbf{M}_4}{4M_0}$$

[6)]Reference[16] considers a microscopic theory of scattering near orientational transitions.

are the ferro- and antiferromagnetic vectors, respectively: $\mathbf{M}_1$, $\mathbf{M}_2$, $\mathbf{M}_3$, and $\mathbf{M}_4$ are the magnetic moments of the iron ions; M_0 is the value of the magnetic moment at absolute zero; A is an exchange-interaction constant; b_i and e_i ($i = 1, 2, 3$) are relativistic-interaction constants of the second and fourth order; and d_i is a constant of exchange-relativistic interaction.

By minimizing the TP (4.1) with respect to the vectors F and G, taking account of terms of the second order, we find, in accordance with[19], that in orthoferrites three types of spin configuration can be realized: 1) G_xF_z, 2) G_zF_x, 3) G_y; two of these permit weak ferromagnetism, whereas the third is purely antiferromagnetic.

At high temperatures, the weak ferromagnetic moment in orthoferrites is usually directed along the c axis of the rhombic crystal; that is, the spin configuration G_xF_z occurs. With lowering of temperature, a transition to the configuration G_zF_x becomes possible, and also to G_y, and this causes diversity in the reorientation transitions observed in orthoferrites. The phase transitions due to spin reorientation may be either of second or of first order, depending on the value and sign of the magnetic-anisotropy constants.

Introducing the angle θ between the antiferromagnetism vector G and the a axis of the rhombic crystal, we shall write the expression for the TP (4.1) after minimization with respect to F in the form

$$\Phi = \Phi_0 + K_1 \sin^2\theta + K_2 \sin^4\theta. \qquad (4.2)$$

where Φ_0 is the part of the energy that is independent of the spin orientation, and where K_1 and K_2 are the first and second anisotropy constants, which can be expressed in terms of the constants b_i, e_i, and d_i of the relation (4.1). The results of Chap. 1 may be applied to the TP (4.2).

For $K_2 > 0$, the reorientation of the spins occurs gradually via two phase transitions of second order; for $K_2 < 0$, the reorientation occurs discontinuously and is a phase transition of first order.

5. SPIN REORIENTATION ACCOMPLISHED BY TWO SECOND-ORDER PHASE TRANSITIONS

The most common and best studied reorientation transitions in orthoferrites are the transitions $G_xF_z \rightarrow G_zF_x$, which are associated with reorientation of the spins and consequently of the magnetic moment in the

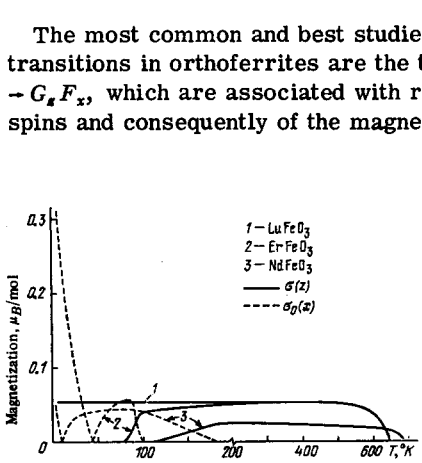

FIG. 3. Temperature dependence of the spontaneous magnetization of several orthoferrites.

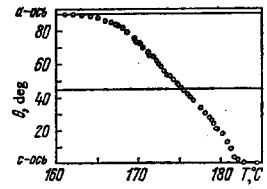

FIG. 4. Temperature dependence of the angle θ between the direction of the magnetic moment and the c axis of a rhombic crystal of samarium orthoferrite.[31]

(ac) plane of the crystal (Fig. 3). Reorientation of this type is observed in the orthoferrites $HoFeO_3$ (51–63 °K), $ErFeO_3$ (90–100 °K), $TuFeO_3$ (86–92 °K),[20–22,25] $YbFeO_3$ (6.5–7.8 °K),[23] $NdFeO_3$ (125–167 °K),[24] $SmFeO_3$ (470–490 °K),[20,21] and also in the substituted orthoferrites $Sm_{1-x}R_xFeO_3$, where the reorientation temperature varies within wide limits, depending on the concentration of the substituent rare-earth ion.[26,27] Reference[28] first pointed out that (ac) reorientation does not occur instantaneously but is spread over an interval of temperature ~ 10°. As was shown in[29–31], in (ac) reorientation in orthoferrites the first anisotropy constant changes sign, while the second constant is positive and only slightly temperature-dependent. In this case, on minimization of the TP (4.2), in addition to equilibrium states with stable orientation of the spins along the a axis ($T > T_2$) and c axis ($T < T_1$) of the crystal, in the temperature interval $T_1 < T < T_2$ a state is realized in which the spins are directed at an angle θ to the a axis of the crystal[30]:

$$\sin^2\theta = -\frac{K_1}{2K_2}. \qquad (5.1)$$

The temperature dependence of the angle θ for samarium orthoferrite in the spin-reorientation range, obtained experimentally, is shown in Fig. 4.[31]

As was shown in Chap. 1, at the temperatures T_1 and T_2, at the instant of beginning and ending of the reorientation process, two phase transitions of second order occur.

It is obvious that at the temperatures T_1 and T_2 anomalies should occur in the physical properties, such as the elastic moduli, the specific heat, etc.; and this is observed experimentally.[23,32–34] Figure 5 shows the temperature dependences of Young's modulus for the orthoferrites of thulium and holmium, along the c axis of the rhombic crystal, in the temperature range of reorientation.[32] It is evident that at the temperature of beginning (T_1) and ending (T_2) of the reorientation, two jumps of the modulus are observed; for

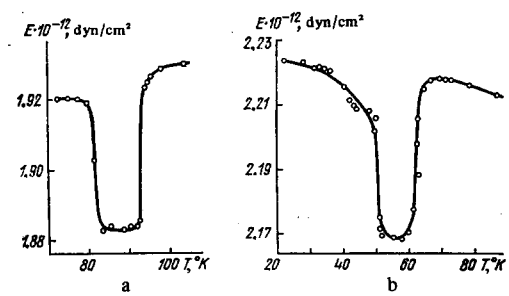

FIG. 5. Temperature dependence of Young's modulus along the c axis of a rhombic crystal of: a, $TuFeO_3$; b, $HoFeO_3$.

$TuFeO_3$ and $HoFeO_3$ they are, respectively, $\Delta E_c = 4.7 \cdot 10^{10}$ and $5 \cdot 10^{10}$ dyn/cm^2.

The value of the jump in Young's modulus can also be determined on the basis of a thermodynamic treatment.[32,35] We minimize the complete TP, taking into account, besides the magnetic energy, the elastic and magnetoelastic energies and the energies of the external stresses:

$$\Phi = \Phi_0 + K_1 \sin^2\theta + K_2 \sin^4\theta + \frac{E_i^2}{2}\xi_i^2 + L_i\xi_i \sin^2\theta + \frac{1}{2} M_i \xi_i^2 \sin^2\theta + P_i\xi_i, \quad (5.2)$$

where E_i is Young's modulus, L_i and M_i are magnetoelastic constants, P_i is the external stress, and ξ_i is the relative deformation ($i = x, y, z$). We find that at the temperatures T_1 and T_2 two jumps should be observed in Young's modulus:

$$\Delta E_i = \frac{L_i^2}{2K_2}, \quad (5.3)$$

corresponding to the two phase transitions of second order.

On substituting in formula (5.3) the second anisotropy constant $K_2 = 3 \cdot 10^4$ erg/cm^3, determined from magnetic measurements, and L_i, found from measurements of the deformation of the lattice on reorientation on the spins ($L_i = \xi_i E_i$)[35] ($L_c = 5.4 \pm 0.5) \cdot 10^7$ erg/cm^3 for $TuFeO_3$ and $L_c = (5.2 \pm 0.5) \cdot 10^7$ erg/cm^3 for $HoFeO_3$), we get $\Delta E_c = (4.8 \pm 1) \cdot 10^{10}$ erg/cm^3 and $\Delta E_c = (4.5 \pm 1) \cdot 10^{10}$ erg/cm^3 for $TuFeO_3$ and $HoFeO_3$ respectively; this agrees, within the limits of experimental error, with the directly observed jumps in Young's modulus. The good agreement in magnitude between the directly measured values of ΔE_c and those calculated by formula (5.3) indicates the correctness of the ideas adopted regarding the character of the reorientation transition under consideration. Another direct demonstration of the fact that (ac) reorientation occurs by two phase transitions of second order is provided by data on the change of heat capacity obtained for ytterbium orthoferrite (Fig. 6).[23] In this case also, at the temperatures of beginning (T_1) and ending (T_2) of the spin-reorientation process jumps were observed in the heat capacity; this points to the presence of phase transitions of second order.

In the presence of an external magnetic field, the character of the phase transitions in (ac) reorientation changes.[11,30] It can be shown that if without a field

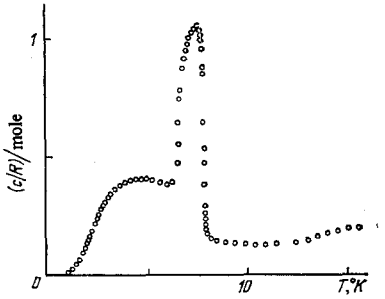

FIG. 6. Temperature dependence of the heat capacity of $YbFeO_3$.[23]

the transition $G_xF_z \rightarrow G_zF_x$ occurs by two phase transitions of second order, then upon application of a magnetic field along the a and c axes of the crystal, one phase transition shifts in temperature, while the other disappears. For this purpose we write the expansion of the TP in the presence of an external magnetic field H in the form

$$\Phi = \Phi_0 + K_1 \sin^2\theta + K_2 \sin^4\theta - \mathbf{FH}M_0. \quad (5.4)$$

In the case $\mathbf{H} \parallel c$ the field hinders deviation of the magnetic moment **F** from the c axis of the crystal on lowering of the temperature.

When the magnetic moment deviates from the c axis by an angle θ, the expansion of the TP (5.4) contains a term

$$FH\cos\theta M_0 = FH\left(1 - \sin^2\frac{\theta}{2}\right) M_0$$

and then the TP in the neighborhood of $\theta = 0$ can be written in the form

$$\Phi = \Phi_0 + K_1\theta^2 + K_2\theta^4 + BH\theta^2, \quad (5.5)$$

that is, just as in the absence of an external field, we shall have a phase transition of second order, which the field merely shifts in temperature. In the case $\mathbf{H} \parallel a$ with $\mathbf{F} \parallel c$, in the expansion of the energy there is an added term $\mathbf{FH}M_0 = FH\sin\theta M_0$, and the TP can be written in the form

$$\Phi = \Phi_0 + K_1\theta^2 + K_2\theta^4 + B'H\theta, \quad (5.6)$$

that is, on minimization of the TP the solution $\theta = 0$ drops out, and the second phase transition disappears.

When $\mathbf{H} \parallel c$, by minimization of the TP (5.4) we obtain the following two solutions:

$$\sin\theta = 0,$$
$$\cos^3\theta - \left(1 - \frac{K_1(T)}{2K_2}\right)\cos\theta - \frac{FHM_0}{4K_2} = 0. \quad (5.7)$$

The first solution corresponds to a minimum of the TP at an arbitrary value of the magnetic field H for temperatures above T_2, and also for $T < T_2$ at sufficiently large $H > H_{th}$, where H_{th} is the so-called threshold field, whose value can be found by means of (5.4) from the condition for existence of a minimum $(\partial^2\Phi/\partial\theta^2)_{\theta=0} \geq 0$. The second solution (the angular phase) is realized for $T < T_2$ when $H \leq H_{th}(T)$.

A similar situation occurs when $\mathbf{H} \parallel a$. The results discussed can be presented in the form of a phase diagram, shown in Fig. 7. The line AT_2A', which separates the angular phase from the phase $\theta = 0$ and determines the temperature dependence, is a line of phase transition of the second kind. The line $H = 0$, $T = T_1$ is a line of phase transition of first order (H parallel to the easy axis). The point T_1 is a critical point, in agreement with the fact that the phase transition at this point disappears when $H \neq 0$.

The temperature dependence of the angle θ for a field H parallel to the c and a axes of the crystal, calculated for samarium orthoferrite,[11] is shown in Fig. 8. It is evident that instead of two phase transitions of second order, in the presence of a field there remains a sin-

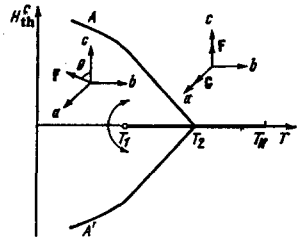

FIG. 7. Magnetic phase diagram of rare-earth orthoferrites corresponding to the case of continuous reorientation of the spin spins in the a, c, plane. T_2A and T_2A' are lines of phase transition of the second kind, $T_1T_2T_N$ is a line of phase transition of the first kind; T_1 is a critical point.

gle phase transition, shifted in temperature.

In[32-36] an investigation was made of the influence of a magnetic field on the anomalies of Young's modulus and of the velocity of sound in orthoferrites of thulium and erbium in (ac) reorientation of the spins. It was shown that with a field parallel to the a and c axes of the crystal, instead of two jumps of Young's modulus, corresponding to two phase transitions of second order, there remains a single one, shifted in temperature (Fig. 9). Similarly, in measurement of the heat capacity of ytterbium orthoferrite in the region of the reorientation temperature under the influence of a magnetic field, there remained only one phase transition, while the second became blurred and disappeared.[23]

The shift of the reorientation temperatures T_1 and T_2 under the influence of a field is determined by the formula

$$\Delta T_{1,2}(H)=\frac{H(T_2-T_1)F}{4K_2}. \qquad (5.8)$$

In the derivation of the relation (5.8) it was taken into account that according to experimental data,[29,30] near the reorientation temperature the first anisotropy constant varies practically linearly with temperature, and it may be supposed that

$$K_1(T)=\frac{2K_2(T-T_2)}{T_2-T_1}. \qquad (5.9)$$

The shift of the reorientation temperature of orthoferrites in an external magnetic field has been observed experimentally in measurement of magnetization curves,[37] of torque curves,[29] of the differential susceptibility,[38] and of the magnetostriction that occurs during field-induced reorientation of the spins.[35,36,39]

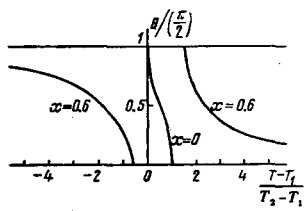

FIG. 8. Temperature dependence of the angle θ, calculated for samarium orthoferrite, with a field parallel to the a and c axes of the crystal ($x=\chi_\perp H/\sigma$)[11].

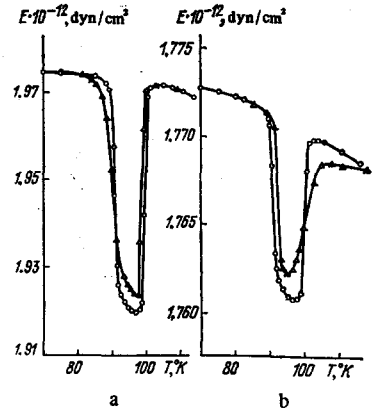

FIG. 9. Temperature dependence of Young's modulus of $ErFeO_3$ in the absence of a field ( ) and with an external field ($H=2\cdot10^3$ Oe) parallel to the c axis (a) and to the a axis (b) of the crystal (Δ).

From the data of these measurements, temperature dependences have been obtained for the threshold fields that produce reorientational phase transitions (phase diagrams; Figs. 10 and 11). From Figs. 10 and 11 it is evident that the threshold field decreases with approach to the reorientation temperature, as a result of the decrease of the anisotropy constant, which vanishes inside the reorientation region.

As was shown in[29,39-41], an external magnetic field applied along the antiferromagnetism axis may induce the reorientation transition $G_xF_z-G_zF_x$ also for orthoferrites for which no spontaneous reorientation of spins is observed. Thus for the orthoferrites $YFeO_3$ and $EuFeO_3$, the threshold fields that produce reorientation of the spins were, according to[29,41], 70 and 75 kOe respectively and remained practically unchanged over the temperature interval 78–300 °K.

By using the expression obtained in[37] for the threshold field,

$$H_{th}=-\frac{H_D}{2}+\sqrt{\frac{H_D^2}{4}+H_0H_A}, \qquad (5.10)$$

and taking, according to[42], the exchange field $H_0=6.4\cdot10^6$ Oe and the Dzyaloshinskii field $H_D=1.4\cdot10^5$ Oe, one can determine from the value of the threshold field of yttrium orthoferrite (the yttrium ions are nonmagnetic) the anisotropy field H_A Oe and, correspondingly, the anisotropy constant of the iron ions, $K=H_AM_0/2=5\cdot10^5$ erg/cm^3.

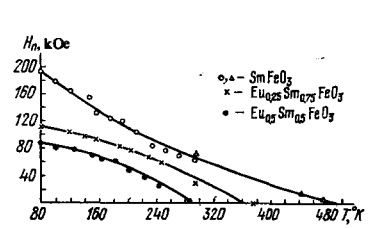

FIG. 10. Phase diagram, $H_{th}(T)$, of the orthoferrites $SmFeO_3$ and $Eu_xSm_{1-x}FeO_3$ accordng to the data of references[29,39].

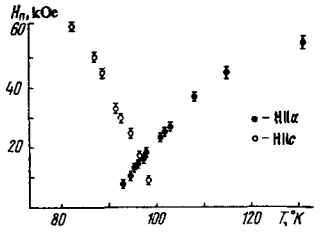

FIG. 11. Phase diagram, $H_{th}(T)$, of $ErFeO_3$.

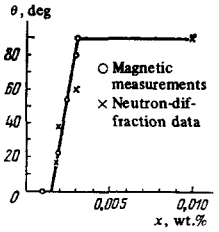

FIG. 13. Concentration dependence of the angle θ for the system $YFe_{1-x}Co_xO_3$.

6. SPIN REORIENTATION OCCURRING AS A SINGLE SECOND-ORDER PHASE TRANSITION

Besides the reorientation transitions considered above, which are accomplished by two second-order phase transitions, and in which the magnetic moment is reoriented from the c to the a axis of the crystal, in certain substituted orthoferrites there is observed a partial spin reorientation, in which the magnetic moment departs from the c axis of the crystal but does not reach the a axis; instead, it sets itself at a certain angle to it. This kind of reorientation transition is observed, for example, in the system $YFe_{1-x}Cr_xO_3$.[43] Figure 12 shows the temperature dependence of the angle between the direction of the magnetic moment and the c axis of the crystal, obtained for several compositions of this system from torque curves. For all the compositions presented, with lowering of temperature there was observed a reorientation of the magnetic moment from the c axis of the crystal to a direction at an angle to it; the angular orientation of the magnetic moment remained stable and constant during subsequent diminution of the temperature. Similar spin-reorientation transitions have been observed also at small replacement, in yttrium orthoferrite, of Fe^{3+} ions by Co^{2+} ions.[44] In[45] a critical concentration of Co^{2+} ions was found, above which the complete spin reorientation $G_xF_z \rightarrow G_zF_x$ is observed; its occurrence is due to the large single-ion anisotropy of the Co^{2+} ions. At Co^{2+} concentrations below 0.3%, there is observed a reorientation transition of the type $G_x \rightarrow G_{xz}$, in which the magnetic moment sets itself, with lowering of temperature, in the (ac) plane at an angle θ to the c axis (Fig. 13).

The presence of orientational transitions of the type $G_x \rightarrow G_{xz}$ in orthoferrites of yttrium replaced by Co^{2+} and Cr^{3+} has been substantiated by neutron-diffraction data.[46]

In contrast to the usual orientational transitions in the ac plane, of the type $G_xF_z \rightarrow G_zF_x$, in the spin reorientation $G_x \rightarrow G_{xz}$ only one phase transition of second order is observed, at temperature T_2, where the magnetic moment departs from the c axis of the crystal. A magnetic field applied along the c axis of the crystal should shift the phase transition $G_x \rightarrow G_{xz}$ toward the lower-temperature region in accordance with the relation (5.8). For yttrium orthoferrite containing 0.24% Co^{2+}, measurements have been made of the magnetostriction that occurs during the reorganization of the magnetic structure in a magnetic field applied along the c axis of the crystal. From these measurements was constructed the temperature dependence of the threshold field at which the spin-rotation process $G_{xz} \rightarrow G_x$ was completed (Fig. 14). The shift of the phase-transition temperature in a field $H \| c$ amounted to $\Delta T_R/\Delta H \sim 3 \cdot 10^{-3}$ deg/Oe.

7. SPONTANEOUS REORIENTATION TRANSITIONS OF FIRST-ORDER (TRANSITIONS OF THE MORIN TYPE)

There has been considerably less investigation of the spin reorientation in orthoferrites that is associated with a transition from the weakly ferromagnetic to the antiferromagnetic state (a transition of the Morin type). Such a reorientation transition $G_xF_z \rightarrow G_y$ is observed, for example, in dysprosium orthoferrite upon lowering of the temperature to ~ 40 °K[20,47] (Fig. 15). To explain the nature of the Morin-type transition in dysprosium orthoferrite, measurement of the magnetic, elastic, and magnetoelastic properties and also of the thermal expansion was undertaken in the transition-temperature region.[48] It was shown that the dimensions of the crystal change discontinuously at the Morin point; the deformations of the crystal that occurred during the transition displayed strong anisotropy, differing along the various crystallographic directions both in magnitude and in sign. Thus the spontaneous deformations along the c and a axes of a $DyFeO_3$ crystal during the transition were, respectively, $(\Delta l/l)_c = -(1.0 \pm 0.1) \cdot 10^{-5}$ and $(\Delta l/l)_a = (9.5 \pm 0.9) \cdot 10^{-5}$. The

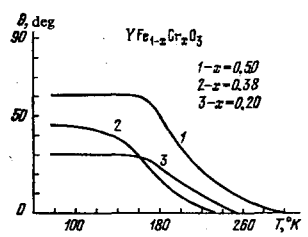

FIG. 12. Temperature dependence of the angle θ for the system $YFe_{1-x}Cr_xO_3$.

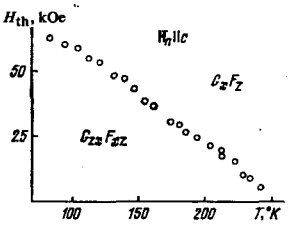

FIG. 14. Phase diagram, $H_{th}(T)$, for $YFe_{1-x}Co_xO_3$ ($x = 0.0024$)

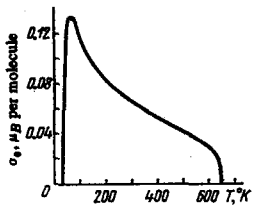

FIG. 15. Temperature dependence of the spontaneous magnetization of $DyFeO_3$[47].

abrupt change of dimensions of the crystal at the Morin point is evidence that the reorientation transition in $DyFeO_3$ proceeds not by smooth rotation of the spins, as was observed for (*ac*) reorientation, but by a jump. In the Morin-type transition there is also observed a form qualitatively different from that considered above in the anomaly of the temperature dependence of Young's modulus (Fig. 16). In the $E(T)$ dependence near 42 °K, where the transition from the weakly ferromagnetic to the antiferromagnetic state occurs, there is observed no jump of Young's modulus, but only a slight change of the modulus, $\Delta E/E \approx 10^{-2}\%$, which is two orders of magnitude smaller than during a smooth spin rotation (see Chap. 4). The absence of the two jumps of Young's modulus, which provide evidence of the existence of "soft" modes of the antiferromagnetic sublattices of the iron ions when $K_2 > 0$, indicate that in the present case apparently $K_2 < 0$, and the spin reorientation occurs discontinuously, by a phase transition of first order.[49]

The measurements of the magnetic and elastic properties in the spin-reorientation range did not display the hysteresis, on heating and cooling of the specimen, that is characteristic of a phase transition of first order. The absence of hysteresis is evidence of the reversible nature of the reorganization of the domain structure in the transition range; this seems entirely natural, since the 180° domain walls, upon approach to the reorientation temperature, serve as nuclei of the new ($\theta = \pi/2$) phase (for more details, see Chap. 2). Upon application of external stresses, the decrease of Young's modulus in the transition-temperature range is obviously caused in the present case by reorganization of the domain structure, and not by rotation of the spins within the domains as was observed in the smooth transition.

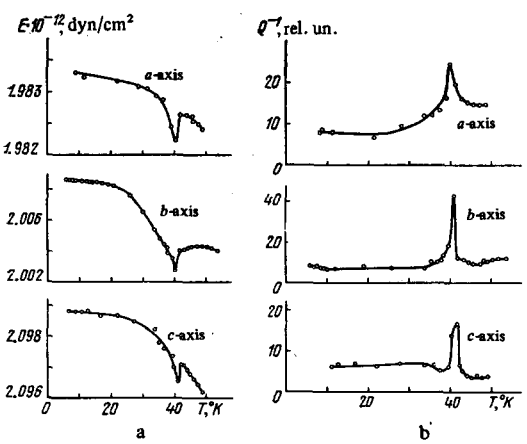

FIG. 16. Temperature dependence of Young's modulus (a) and of the internal friction (b) along the *a*, *b*, and *c* axes of $DyFeO_3$.

The slight difference of moduli observed to the left and to the right of the transition temperature is apparently due to the different stiffness of the crystal in the weakly ferromagnetic and antiferromagnetic states. In the transition-temperature range, appreciable increase of the internal friction is observed (see Fig. 16); this is apparently due to irreversible energy losses during the displacement of the interdomain walls under the influence of the external stresses.

The magnitude of the transition interval ΔT in which a gradual reorganization of the domain structure should occur can be estimated from the relation

$$\Delta T \frac{dK_1}{dT} \sim 4\pi I^2, \qquad (7.1)$$

where I is the magnetization. On substituting in this formula the experimentally determined values for dysprosium orthoferrite, $I = 20$ G and $dK_1/dT = 2 \cdot 10^3$ erg cm^{-3} deg^{-1}, we get $\Delta T \approx 2°$, which is close to the observed temperature interval where the anomaly of the elastic moduli and of the internal friction appears. Application of an external magnetic field parallel to the *c* axis of the crystal shifts the Morin point toward the lower-temperature region and slightly changes the character of the observed anomalies of Young's modulus and of the internal friction (Fig. 17). From field ~ 1 kOe up, the "spike," which is due to motion of domain boundaries, disappears, because the specimen apparently becomes single-domain, and there is observed only a slight jump, caused by the different stiffness of the crystal in the weakly ferromagnetic and antiferromagnetic states. With application of a magnetic field, the internal-friction peak observed in the Morin-point

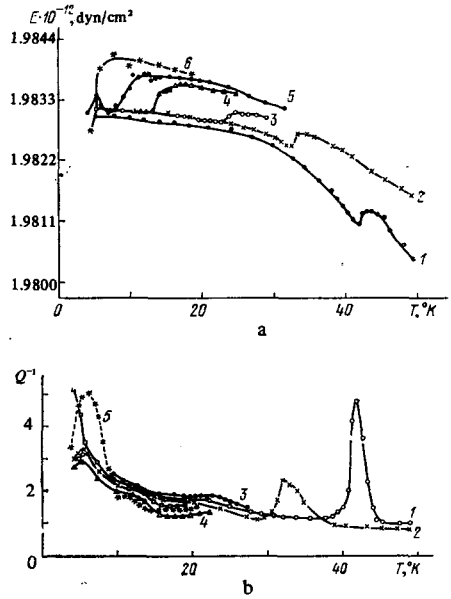

FIG. 17. Temperature dependence of Young's modulus (a) and of the internal friction (b) of $DyFeO_3$ in an external magnetic field parallel to the *c* axis of the crystal. Curve 1, $H = 0$; 2, $H = 1$ kOe; 3, $H = 2$ kOE; 4, $H = 3$ kOe; 5, $H = 4.4$ kOe; 6, $H = 6$ kOe.

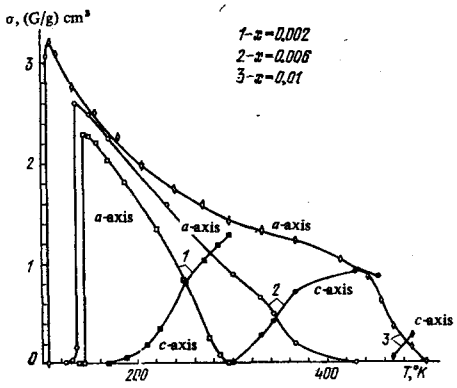

FIG. 18. Temperature dependence of the spontaneous magnetization of $DyFe_{1-x}Co_xO_3$.

region disappeared; this also is obviously due to a transition of the specimen to a single-domain state.

The strong influence of reorganization of the domain structure on the character of the observed anomalies once more corroborates the correctness of the supposition that the reorientation of spins at the Morin point in $DyFeO_3$ occurs discontinuously. The low-temperature transition observed in the orthoferrite $DyFe_{1-x}Co_xO_3$ ($x<1\%$) has a similar character. A small replacement of iron ions by cobalt ions in dysprosium orthoferrite leads to an appreciable rise of the Morin temperature, and also to the occurrence of still another reorientation transition in the ac plane (Fig. 18); in consequence, $DyFe_{1-x}Co_xO_3$ monocrystals are very suitable objects for study of the character of spin-reorientation transitions.[50,51] As is evident from Fig. 18, at high temperatures the weak ferromagnetic moment in these crystals is oriented along the c axis of the crystal, and with lowering of the temperature it reorients to the a axis of the crystal (the transition $G_xF_z \rightarrow G_zF_x$). With further lowering of the temperature, a transition is observed from the weakly ferromagnetic to the purely antiferromagnetic state, because of reorientation of the spins to the rhombic b axis. In contrast to the high-temperature transition, this transition occurs practically discontinuously, within a very narrow temperature interval. In order to resolve unambiguously the question of the nature of the observed transitions in $DyFe_{1-x}Co_xO_3$, a determination was made of the spin of the second anisotropy constant K_2, whose influence becomes important near the reorientation temperature, where the first anisotropy constant K_1 changes sign. To determine the sign of the second anisotropy constant, torque curves were taken in various crystallographic planes. Analysis of rotation curves taken in the ac plane showed that the second anisotropy constant for the transition $G_xF_z \rightarrow G_zF_x$ is positive; consequently the spin reorientation occurs here by continuous rotation of the spins, and there are two phase transitions of second order, at the instants of beginning and of ending of the reorientation process.

We shall now consider in more detail the less-studied transition from the weakly ferromagnetic to the antiferromagnetic state (Morin-type transition), which is accompanied by reorientation of the spins in the ab plane. On the torque curves taken below the Morin point ($T_M \approx 140$ °K) there were observed pronounced anomalies, due to transition of the crystal from the antiferromagnetic to the weakly ferromagnetic state when the projection of the magnetic field on the a axis of the crystal was sufficiently large (Fig. 19). It can be shown that near the Morin point, for small values of the angle φ (φ is the angle between the b axis and the magnetic field H), the torque due to rotation of the magnetic moment of the iron ions by the external field is expressed by the relation

$$M = \frac{\sigma_0^2 H^2}{4(K_1' - K_2')} \sin 2\varphi. \qquad (7.2)$$

where K_1' and K_2' are the first and second anisotropy constants in the expansion of the TP

$$\Phi = K_1' \sin^2\theta - K_2' \sin^2\theta \cos^2\theta - \sigma_0 H \sin\theta \sin\varphi, \qquad (7.3)$$

where σ_0 is the specific spontaneous magnetization, and where θ is the angle between the b axis and the antiferromagnetism vector.[51]

The abrupt transition, observed experimentally in Fig. 19 (curve 2), from the rotation curve described by the relation (7.2) to the curve $M = \sigma_0 H \cos\varphi$, when the crystal becomes weakly ferromagnetic, is possible only when the second anisotropy constant has a negative sign. The second anisotropy constant determined from the torque curves had the value $K_2' = -(7 \pm 2) \cdot 10^4$ erg/cm³. The negative sign of K_2 is evidence of the fact that the transition $G_xF_z \rightarrow G_y$, in contrast to the transition $G_xF_z \rightarrow G_zF_x$, occurs in $DyFe_{1-x}Co_xO_3$ by a phase transition of first order. For dysprosium orthoferrite, because of the low reorientation temperature, the anisotropy of the susceptibility of the rare-earth ions is large near the transition temperature, and this prevents the direct determination of the value of K_2 from the torque curves.

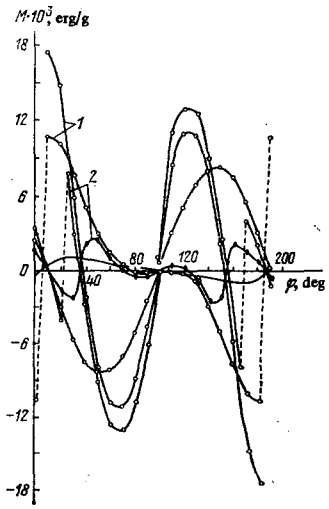

FIG. 19. Torque curves of a monocrystal of $DyFe_{0.998}Co_{0.002}O_3$ in the a, b plane, in field 6.25 and 12.3 kOe. Curve 1, T = 145 °K; 2, T = 135 °K; 3, T = 118 °K. φ is the angle between the direction of the magnetic field and the b axis of the rhombic crystal.

8. REORIENTATION TRANSITIONS FROM THE ANTIFERROMAGNETIC TO THE WEAKLY FERROMAGNETIC STATE INDUCED BY AN EXTERNAL MAGNETIC FIELD

In orthoferrites that exhibit a spontaneous transition of the Morin type, application of an external magnetic field at low temperatures, when the crystal is in an antiferromagnetic state, may produce unusual orientational transitions. We shall consider in its general features the phase diagram for such orthoferrites with a negative second anisotropy constant.[48,52] We write the expansion of the TP for $DyFe_{1-x}Co_xO_3$, in which transitions $G_xF_z - G_zF_x - G_y$ are observed, when a field h is directed along the a axis of the crystal below the Morin point:

$$\Phi = \Phi_0 - mh\cos\theta + K_1\cos^2\theta + K_2\cos^4\theta \qquad (8.1)$$

(θ is the angle between the direction of the spins and the c axis of the crystal). In accordance with experimental data we set

$$K_1 = K\frac{T-T_1}{T}, \quad K_2 < 0. \qquad (8.2)$$

By minimizing the free energy, we obtain the equilibrium states of the antiferromagnetism vector G and the corresponding regions on the H, T phase diagram where these states are stable (Fig. 20).

a) *The phase* $\theta = 0$ *is stable* in the region located *above* the curve T_2BA, whose equation is

$$K_1(T) + K_2 - mh = 0. \qquad (8.3)$$

By use of (8.2) we get

$$T = T_1\left(1 + \frac{2K_2}{K} - \frac{mh}{2K}\right) \qquad (8.4)$$

and

$$T_2 = T_1\left(1 - \frac{2K_2}{K}\right).$$

b) *Angular phase.* The equation for the angle θ is

$$\cos^3\theta + 3p\cos\theta + 2q = 0. \qquad (8.5)$$

The angular phase is stable in the region below T_1BA. The line AB is determined by Eq. (8.3); the line BT is determined by the equation

$$h = \frac{2|K_2|}{m}\left(\frac{K_1(T)}{3|K_2|}\right)^{3/2}. \qquad (8.6)$$

At the point B the lines AB and BT have a common tangent.

We consider transitions between the phases. Obviously the line AB is a line of phase transition of second order (a continuous transition). In the region contained within the curve T_2BT_1, the angular phase and the collinear phases ($\theta = 0, \pi$) coexist. The line of phase transition of first order (BT_0) is determined by the equation $\Phi(\theta = 0) = \Phi[\theta(HT)]$ (the section BT_0). The temperature T_0 is equal to $T_0 = (T_2 + T_1)/2$; the point B is the critical point at which the line of phase transition of second order changes to a line of first order, and its coordinates are

$$T_B = T_1\left(1 - \frac{3|K_2|}{K}\right), \quad H_B = \frac{2|K_2|}{m}. \qquad (8.7)$$

It is experimentally convenient to investigate the phase diagram of the orthoferrite under consideration by measuring the field dependence of the magnetostriction produced by an external magnetic field applied along the a axis of the crystal below the Morin point.[48] It is easily shown that the value of the magnetostriction upon reorientation of the Fe^{3+} spins is determined by the relation

$$\lambda_i = \frac{L_i}{E_i}\cos^2\theta, \qquad (8.8)$$

where L_i and E_i ($i = x, y, z$) are the magnetoelastic constant and Young's modulus along the corresponding axis of the crystal, and where θ varies with field in accordance with (8.5). Figure 21 shows the dependence of the transverse magnetostriction along the c axis of the crystal on a field applied along the c axis of the crystal in order to induce, below T_M, a transition from the antiferromagnetic to the weakly ferromagnetic state. The magnetostriction that occurred during reorientation of the spins reached the value $-7.2\cdot10^{-5}$. Knowing the value of Young's modulus ($E_c = 2\cdot10^{12}$ dyn/cm^2), one can determine for the orthoferrite under study the value of the magnetoelastic constant $L_c = \lambda_c E_c = -1.4\cdot10^8$ dyn/cm^2. From the field dependence of the magnetostriction it is possible to determine the values of the threshold fields, which produce reorientation of the spins from the b axis to the c axis of the rhombic crystal ($G_y - G_zF_x$), at various temperatures, and to construct the (H, T) phase diagram (Fig. 22).

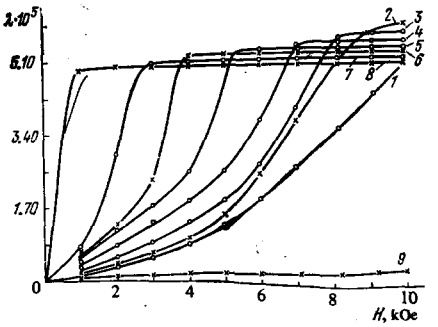

FIG. 21. Experimental field dependence of the longitudinal magnetostriction along the a axis of a crystal of $DyFeO_{.994}Co_{0.006}O_3$, *at various temperatures.* Curve 1, T = 86 °K; 2, T = 90 °K; 3, T = 96 °K; 4, T = 100 °K; 5, T = 106 °K; 6, T = 111 °K; 7, T = 115 °K; 8, T = 120 °K; 9, T = 125 °K.

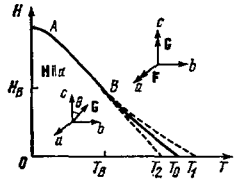

FIG. 20. Theoretical phase diagram of $DyFe_{1-x}Co_xO_3$. AB, line of phase transition of the second kind; BT_0, line of phase transition of the first kind; BT_1 and BT_2, lines of loss of stability of the homogeneous phases.

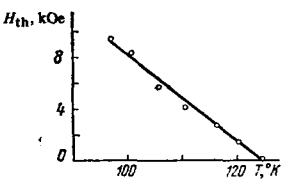

FIG. 22. Experimental phase diagram, $H_{th}(T)$, along the a axis of a crystal of $DyFe_{0.994}Co_{0.006}O_3$.

From the experimental phase diagram one can determine $dK_1/dt = m = dh/dT = 5 \cdot 10^3$ erg cm deg^{-1} and, knowing $|K_2| = (7 \pm 2) \cdot 10^4$ erg/cm^3, estimate the coordinates of the tricritical point B:

$$H_B \approx 10^4 \text{ Oe}, \quad T_B \approx 100 \text{ °K}.$$

In contrast to the theoretical phase diagram presented above (see Fig. 20), experimentally no temperature hysteresis is observed, such as would correspond to the presence of a metastable phase near the transition temperature; this can be explained on the basis of the mechanism described in Chap. 1, in which an anhysteretic transition occurs by virtue of a continuous growth of the new phase from domain boundaries.

A similar transition from the antiferromagnetic to the weakly ferromagnetic state should be observed also for hysprosium orthoferrite with a field applied along the c axis of the crystal (the transition $G_y - G_xF_z$). Since the Morin point of dysprosium orthoferrite lies in a region of lower temperatures ($T_M \approx 40$ °K) than for the substituted compounds, it is necessary in the expansion of the free energy for this case to take into account the paramagnetic system of rare-earth ions, as was done in[53]. According to[53] the equilibrium value of the TP of dysprosium orthoferrite in an external field H can be written in the form

$$\Phi = -\frac{1}{2A}[\mathbf{H}_t^2 - (\mathbf{H}_t\mathbf{G})^2] + \tilde{a}_1 G_x^2 + \tilde{a}_2 G_z^2, \qquad (8.9)$$

where $\mathbf{H}_t = \mathbf{H} + \mathbf{H}_D + \mathbf{H}_R$; here H_D is the Dzyaloshinskiĭ field, with components $(d_1G_z, 0, d_3G_x)$, $H_R = (\hat{\lambda}_1 M_R^+ + \hat{\lambda}_2 M_R^-)/2$ is the field exerted by the rare-earth ions on the Fe^{3+} ions, and M_R^+ is the magnetization of the rare-earth ions.

In this form, the free energy is a function of G (or of the polar and azimuthal angles θ and φ). The expression (8.9) coincides in form with the free energy of an antiferromagnet in a magnetic field, whose role in the present case is filled by H_t, the total effective field acting on the Fe^{3+} ions. The external field here plays a double role. Besides its direct action on the iron sublattice, it induces magnetization of the rare-earth ions and thus amplifies the molecular field exerted on the Fe^{3+} ions by the rare-earth ions. At low temperatures the induced molecular field may far exceed the external field, so that the orientational transition will actually occur in the internal molecular field. Since the induced molecular field depends strongly on temperature, the phase diagrams in this case have a different character from that considered above.

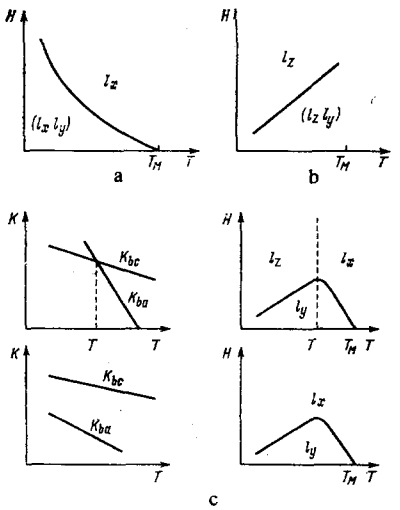

FIG. 23. Theoretical phase diagrams of $DyFeO_3$ for H ∥ c (Fig. 23a), H ∥ a (Fig. 23b), and H ∥ b (Fig. 23c), and for various ratios of the anisotropy constants K_{ba} and K_{bc} in the plane b, a and b, c.

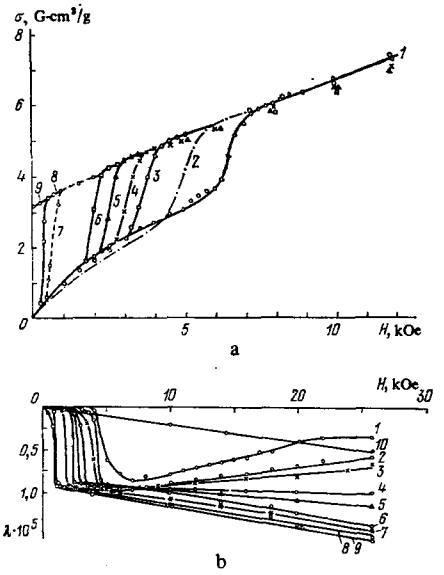

FIG. 24. Isotherms of the magnetization (a) and of the longitudinal magnetostriction (b) of $DyFeO_3$ along the c axis of the crystal, at various temperatures. a) T(°K) = 4.2° (1), 6.7° (2), 10.9° (3), 14.9° (4), 19.7° (5), 27.7° (6), 34° (7), 40.3° (8) and 41.7° (9); b) T(°K) = 6.1° (1), 7.5° (2), 8.7° (3), 12.9° (4), 17° (5), 21.7° (6), 25.6° (7), 30.5° (8), 34° (9) and 42° (10).

Figure 23 shows theoretical phase diagrams constructed from dysprosium orthoferrite according to the data of[53], with the external magnetic field parallel to the c, a, and b axes of the crystal. In order to obtain experimental phase diagrams, measurements were made of the magnetization curves and of the magnetostriction for various orientations of the magnetic field. When the magnetic field was applied along the c axis of the crystal in the low-temperature region ($T < T_M$), a field-induced transition $G_y - G_xF_z$ was observed. Figure 24 shows magnetization curves taken along the c axis of a $DyFeO_3$ monocrystal. It is evident that below the Morin point ($T_M = 42$ °K), when the crystal is antiferromagnetic (G_y), on application of a sufficiently large magnetic field H an abrupt transition to the weakly ferromagnetic state is observed. The value of the threshold field that produces this transition decreases with increase of temperature and vanished at the Morin point, above which the crystal becomes a weak ferromagnet (G_xF_z). The field-induced transition $G_y - G_xF_z$ was accompanied also by the occurrence of magnetostrictive deformations (see Fig. 24b). Figure 25a shows the temperature dependence of the threshold field obtained experimentally on the basis of measurements of the magnetization curves, the magnetostriction, and

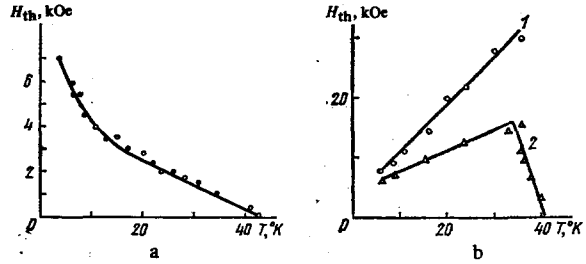

FIG. 25. Experimental phase diagram of $DyFeO_3$. a, $H \parallel c$ (●, according to magnetization measurements; ⊙, from measurements of longitudinal magnetostriction). b, $H \parallel b$ (Curve 2).

the field dependence of Young's modulus (Fig. 17). Near the Morin point, the $H_{th}(T)$ dependence is practically linear; the values of $\Delta T_M/\Delta H_{th}$ obtained from different measurements agree well with each other and are ~ 12 deg/kOe. Far from T_M, in the low-temperature range, $H_{th}(T)$ departs from the linear dependence and shows a rapid rise with lowering of the temperature. A distinctive peculiarity of the transition under consideration is the fact that the reorientation of the spins occurs here in the a, b plane, perpendicular to the direction of the external magnetic field.

In agreement with theoretical ideas, reorientation transitions were also induced in $DyFeO_3$ on application of a magnetic field along the a and b axes of the crystal and were detected in measurement of the magnetostriction.[53]. Figure 25b shows the temperature dependence of the threshold field that produces the reorientation transition, determined from the isotherms of the magnetostriction along the a and b axes. On the experimental phase diagram for $H \parallel a$, the threshold field decreases with lowering of the temperature, in proportion to the distance from the Morin point. In the case $H \parallel b$, the temperature dependence of the threshold field has a more complicated character. Comparison of the experimental and theoretical phase diagrams shows good qualitative agreement. The threshold field along the a axis is larger than the field along the b axis; this is obviously due to the large value of the g factor along the b axis of the crystal, since, according to[53], the value of the threshold field is inversely proportional to the value of the susceptibility along the corresponding axis of the crystal. A specific peculiarity of the reorientation transitions in the case $H \parallel a$ and $H \parallel b$ is the fact that the reorientation occurs here under the influence of the molecular field of the rare-earth ions, amplified by the action of the external field.

9. ORTHOFERRITES WITH TWO DIFFERENT TRANSITIONS OF THE MORIN TYPE

As was shown in the preceding paragraphs, the transition from the weakly ferromagnetic to the antiferromagnetic state occurs in the orthoferrites $DyFeO_3$ and $DyFe_{1-x}Co_xO_3$ by a phase transition of the first kind. In the mixed orthoferrite $Ho_{0.5}Dy_{0.5}FeO_3$, two Morin-type transitions, different in character, are observed.[54] At high temperatures, the weak ferromagnetic moment in this orthoferrite is oriented along the c axis of the

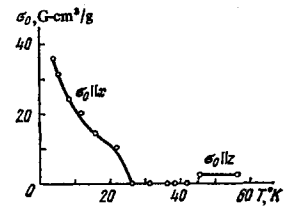

FIG. 26. Temperature dependence of the spontaneous magnetization of a monocrystal of $Ho_{0.5}Dy_{0.5}FeO_3$.

crystal (Fig. 26). When the temperature is lowered to 4.6 °K, the weak ferromagnetism disappears, and the crystal changes to a purely antiferromagnetic state ($G_xF_z \rightarrow G_y$), in which it remains until 25 °K; there a new reorientation transition occurs to a state with a weak ferromagnetic moment along the a axis of the crystal (the transition $G_y \rightarrow G_zF_x$). For the purpose of determining the character of the two Morin-type reorientation transitions observed in $Ho_{0.5}Dy_{0.5}FeO_3$, in addition to magnetic measurements, measurements were made of the temperature dependence of the elastic moduli, the internal friction, and the thermal expansion (Fig. 27). As is seen from Fig. 27a, the anomalies of Young's modulus on reorientation of the spins near 45 and 21 °K differ significantly from each other. In the low-temperature transition $G_zF_x \rightarrow G_y$, there are two jumps of Young's modulus, at temperatures $T_1 = 18.8$ °K and $T_2 = 24.2$ °K; this is similar to what was observed for the transition $G_xF_z \rightarrow G_zF_x$ (see Chap. 4) and indicates a gradual rotation of the spins in the reorientation process. The appreciable decrease of Young's modulus in the spin reorientation-temperature region is due to the fact that in this temperature interval, the spins rotate spontaneously with change of temperature, and this facilitates their rotation by an external stress and makes the crystal, as it were, less stiff.

As was shown in Chap. 4, in this case two phase transitions of the second kind take place, at temperatures $T_1 = 18$ °K and $T_2 = 24$ °K. A qualitatively different

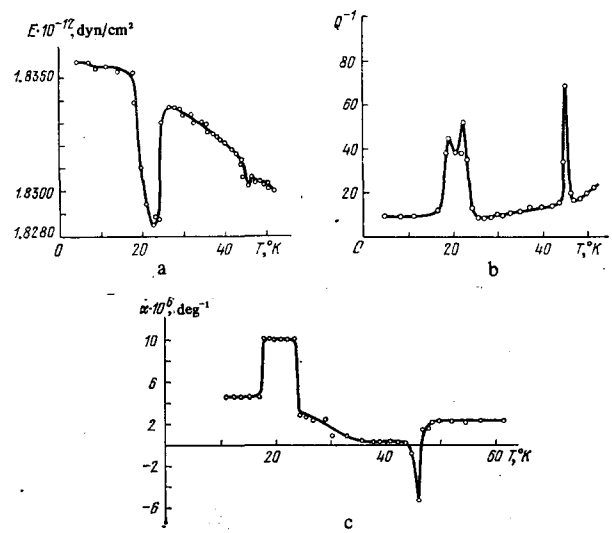

FIG. 27. Temperature dependence of Young's modulus (a), the internal friction (b), and the coefficient of thermal expansion (c) of a monocrystal of $Ho_{0.5}Dy_{0.5}FeO_3$.

anomaly of Young's modulus occurs near 45 °K with the transition $G_xF_z \rightarrow G_y$. In the temperature dependence of Young's modulus there is observed no jump of the modulus, but only a small negative peak due, as was noted in Chap. 3, to reorganization of the domain structure. Application of external stresses does not promote rotation of the spins, as was the case in the low-temperature transition, but changes the ratio between the phases G_xF_z and G_y.

In the temperature dependence of the internal friction, maxima of the damping were observed near 21 and 45 °K; these are obviously due to irreversible energy losses during reorientation of the spins (Fig. 27b). In the low-temperature transition, the damping maximum obtained was, as it were, bifurcated, indicating an increase of the damping at the instant of beginning T_1 and of ending T_2 of the reorientation process. The difference in character of the two observed transitions is especially clearly evident from the curve showing the temperature dependence of the coefficient of thermal expansion (Fig. 27c). It is evident that in the low-temperature transition, two jumps are observed in the coefficient of thermal expansion, at temperatures $T_1 = 18$ °K and $T_2 \approx 24$ °K, while in the second Morin-type transition, near 46 °K, there is an abrupt anomaly, corresponding to a discontinuous decrease of the crystal dimensions at the instant of the transition. The observed difference of the anomalies of thermal expansion in the transitions $G_xF_z \rightarrow G_y$ and $G_y \rightarrow G_zF_x$ is evidence of the different character of these reorientation transitions in $Ho_{0.5}Dy_{0.5}FeO_3$. The presence of two transitions of Morin type in this compound has enabled us to establish that, in contrast to the generally accepted point of view, transitions from a weakly ferromagnetic to an antiferromagnetic state in orthoferrites can occur not only jumpwise, but also continuously, via two phase transitions of second order.

10. SPONTANEOUS SPIN-REORIENTATION TRANSITION IN GADOLINIUM

Besides orthoferrites, spin-reorientation phase transitions have been observed in a whole series of uniaxial rare-earth ferro- and ferrimagnets, for example in gadolinium.

We consider a uniaxial ferromagnet. In the absence of a field, its TP, which depends on the orientation of the magnetization, contains only the magnetic-anisotropy energy; consequently, when two magnetic-anisotropy constants are taken into account, it can be represented in the form

$$\Phi = K_1 \sin^2\theta + K_2 \sin^4\theta. \qquad (10.1)$$

where K_1 and K_2 are the first and second magnetic-anisotropy constants, and where θ is the angle between the hexagonal axis and the spontaneous magnetization. The relation (10.1) for a uniaxial ferromagnet coincides with the expression given above for the TP of orthoferrites (see (4.2)); therefore for uniaxial ferromagnets the conditions for occurrence of spin-reorientation phase transitions and their character for various relations between the constants are completely analogous

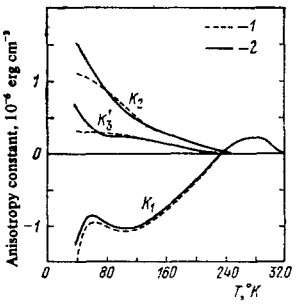

FIG. 28. Temperature dependence of the anisotropy constants of gadolinium in zero field (1) and in field 30 kOe (2).

to those presented above for orthoferrites (for uniaxial ferromagnets, these relations were obtained in[55-57]). The difference between uniaxial crystals and orthoferrites consists solely of the fact that in orthoferrites the reorientation occurs in a single plane (for example *ac*), whereas in uniaxial magnets (when there is no anisotropy in the basal plane) there emerges a cone of directions of easy magnetization. This leads to the occurrence in uniaxial ferromagnets, in the spin-reorientation range, of an additional gapless branch of spin waves, in which the magnetization vector moves along the surface of the cone of easy magnetizations.

Spin reorientation in gadolinium has recently been investigated in detail. Even the first measurements, on polycrystalline specimens of gadolinium, revealed anomalies of the magnetization in weak fields[58] near temperature 220 °K. At first they were interpreted (by analogy with other heavy rare-earth metals) as caused by formation of a helical magnetic structure.[58] But subsequent investigations of the magnetic anisotropy[59,60] and neutron-diffraction measurements made on Gd monocrystals[61-63] showed that in this temperature range the first magnetic-anisotropy constant of gadolinium changes sign, while the second anisotropy constant is positive (Fig. 28); hence, with lowering of temperature, there occurs in gadolinium a transition from a phase with the magnetization vector parallel to the hexagonal axis of the crystal to an angular phase, with the magnetization vector at an angle to the hexagonal axis (Fig. 29). The unusual temperature dependence of the angle of the cone is caused by the complicated temperature dependence of the anisotropy constants (see Fig. 28).[7)]

The transition to the angular phase in gadolinium is, as was shown above, a spin-reorientation phase transition of the second kind and is accompanied by anomalies of many properties. Thus, for example, in[58] it was discovered that at this temperature there is a minimum of the coercive force, and according to reference[64] the magnetoresistance passes through a maximum in the transition region.

The properties of gadolinium that have been most

[7)]As is seen from Fig. 28; in gadolinium at low temperatures the value of the third anisotropy constant K_3' is appreciable. Near the spin-reorientation point, however, $K_3' = 0$; therefore in this temperature range the thermodynamic potential (10.1), which takes account only of K_1 and K_2, is correct.

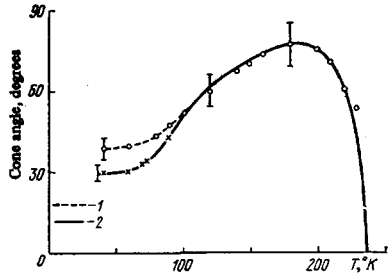

FIG. 29. Temperature dependence of the direction of easy magnetization of gadolinium in zero field (1) and in field 30 kOe (2).

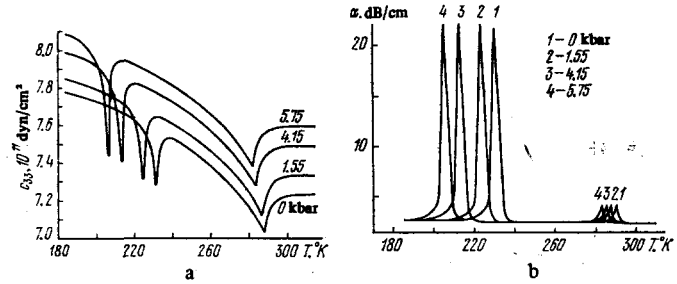

FIG. 31. Temperature dependence of the elastic constant c_{33} of gadolinium at various pressures (a) and of the absorption of longitudinal ultrasonic waves along the c axis of a monocrystal of gadolinium at various pressures (b).

thoroughly investigated in the spin-reorientation range are the magnetoelastic. Measurements of the thermal expansion of a gadolinium monocrystal[65] have shown that in the transition-temperature range (~ 220 °K) there is a maximum of the coefficient of thermal expansion along the hexagonal axis of the crystal, whereas the anomaly of the thermal expansion in the basal plane is considerably smaller (Fig. 30).

The anomalies of the elastic moduli of gadolinium in the spin-reorientation range have been thoroughly investigated, not only at atmospheric pressure, but also when an external hydrostatic pressure is applied to a gadolinium crystal.[66–69] It was discovered that the spin-reorientation transition is accompanied by anomalies of the elastic constant c_{33}, which describes the velocity of propagation of longitudinal elastic waves along the hexagonal axis of the crystal (Fig. 31a), whereas the other constants experience no anomalies in this temperature range. The spin-reorientation transition is also accompanied by a maximum of the absorption of longitudinal ultrasonic waves along the hexagonal axis (Fig. 31b).

The anomalies of the elastic properties in the spin-reorientation range are due to the fact that upon application of elastic stresses, because of magnetoelastic interaction, a change of the anisotropy energy occurs. This leads to the result that the orientation of the magnetization vector changes, and this causes additional magnetostrictive deformations, which lead to a change of the elastic constants. Since $K_1 \approx 0$ in the spin-reorientation range, the effect of elastic stresses will be greatest here; consequently the anomalies of the elastic properties will be greatest in this same temperature range. As was shown in[69], with allowance for magnetoelastic interaction the TP of a hexagonal ferromagnet can be represented in the form

$$\Phi = \Phi_0 + \frac{1}{2}\sum c^0_{ij}\xi_i\xi_j + K_1(T)\sin^2\theta + K_2(T)\sin^4\theta + (\xi_{xx}+\xi_{yy})L^\theta_{xx}\sin^2\theta + \xi_{zz}L^\theta_{zz}\sin^2\theta; \tag{10.2}$$

here c^0_{ij} are the elastic constants without allowance for magnetoelastic interactions, and L^θ_{ii} are the magnetoelastic-interaction constants connected with orientation of the magnetization vector with respect to the crystallographic axes and determined by the anisotropic magnetostriction. (In formula (10.2) no allowance is made for the magnetoelastic interaction connected with paraprocess magnetostriction.) On minimizing (10.2) with respect to θ, we find for the region below the spin-reorientation temperature

$$\sin^2\theta = -\frac{K_{1\,\mathrm{eff}}}{2K_2} = -\frac{K_1 + (\xi_{xx}+\xi_{yy})L^\theta_{xx} + \xi_{zz}L^\theta_{zz}}{2K_2}. \tag{10.3}$$

Thus the orientation angle of the magnetization vector depends on the deformation of the crystal lattice. (It is necessary to take into account that the ξ_{ii} contain not only the spontaneous magnetostrictive deformations but also the deformations produced by the external stresses.) From (10.2) we obtain for the elastic constant c_{11}

$$c_{11} = \frac{\partial^2\Phi}{\partial\xi^2_{xx}} = c^0_{11} + L^\theta_{xx}\frac{d(\sin^2\theta)}{d\xi_{xx}}.$$

Above the spin-reorientation point, where $\theta = 0$, $c_{11} \equiv c^0_{11}$; but in the angular phase, we find from (10.3)

$$c_{11} = c^0_{11} - \frac{(L^\theta_{xx})^2}{2K_2}. \tag{10.4}$$

Analogous expressions are easily obtained for the other elastic constants; as was shown in[69], in the transition to the angular phase the elastic constants characterizing longitudinal deformations should decrease, and no anomalies should be observed in the elastic constants for shear waves. The values of L^0_{ii} can be calculated from the known elastic constants of gadolinium in the paramagnetic range and the magnetostriction constants of gadolinium.[70] The results of the calculation are shown in Fig. 32. It is seen that in the reorientation range, $L^\theta_{xx} \ll L^\theta_{zz}$. This explains the fact that in the spin-reorientation transition in gadolinium, anomalies are observed only in the elastic constant c_{33}.

From the expression for the angle θ (see (10.3)) it follows that when magnetoelastic interaction is taken into account, the first effective anisotropy constant $K_{1\,eff}$ depends on the deformation. On taking into ac-

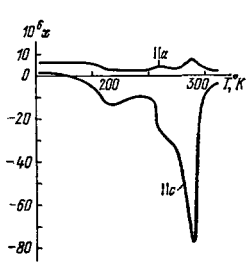

FIG. 30. Temperature dependence of the coefficient of thermal expansion of a monocrystal of gadolinium.

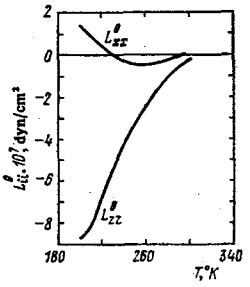

FIG. 32. Temperature dependence of the magnetoelastic constants of gadolinium.

count that under the action of hydrostatic pressure p

$$\xi_{xx} = \xi_{yy} = -\beta_\perp p, \quad \xi_{zz} = -\beta_\parallel p,$$

where $\beta_\perp$ and $\beta_\parallel$ are the perpendicular and parallel compressibilities, and also remembering that $K_{1\,eff} = 0$ at the reorientation point, we get

$$\frac{dT_{th}}{dp} = -(2\beta_\perp L^0_{xx} + \beta_\parallel L^0_{zz}) \left(\frac{dK_1}{dT}\right)^{-1}_{T_{th}}. \qquad (10.5)$$

Figure 33 shows the experimental and theoretical $\Delta T_{th}(p)$ dependences for gadolinium. It is seen that there is good agreement of the theoretical calculations with experiment.

In[68,71] the effect of a magnetic field on the elastic properties, the specific heat, and the electrical resistivity of gadolinium was investigated near the spin-reorientation temperature. Without going into this question in detail, we note that a field parallel to the hexagonal axis shifts the spin-reorientation point toward the low-temperature region, since it stabilizes the magnetic structure with the direction of the magnetization vector along the hexagonal axis; but in a field perpendicular to this direction, the spin-reorientation phase transition disappears, since any finite field of this orientation prevents the magnetic moment from orienting along the c axis. We note that in sufficiently strong fields $H \parallel c$ the spin-reorientation phase transition disappears, since in fields larger than the magnetic-saturation field no rotation of the magnetization vector occurs when the temperature is changed. For theoretical calculations devoted to this question, see[72]; there the effect of domain structure on the properties in the spin-reorientation range is also treated.

11. SPIN-REORIENTATION TRANSITIONS IN RARE-EARTH UNIAXIAL INTERMETALLIC COMPOUNDS

Recently, magnetic anisotropy has been investigated in hexagonal intermetallic compounds of the type

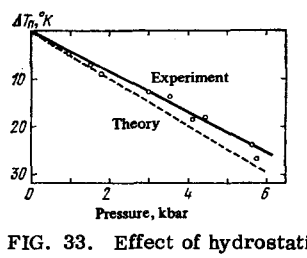

FIG. 33. Effect of hydrostatic pressure on the spin-reorientation temperature of gadolinium.

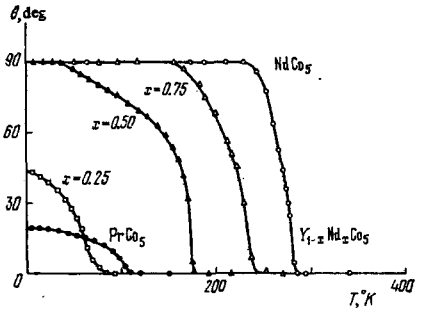

FIG. 34. Temperature dependence of the orientation of the magnetization in compounds RCo_5.

RCo_5[73] (where R is a rare-earth ion); it was shown that in a number of these compounds, the first magnetic-anisotropy constant changes sign at a certain temperature. This leads to spin-reorientation phase transitions; and since $K_2 > 0$ in the compounds studied, these transitions are phase transitions of second order.

Figure 34 shows the temperature dependence of the angle θ of rotation of the magnetization for several RCo_5 compounds. It is seen that in the compounds $Y_{1-x}Nd_xCo_5$ with $x \geq 0.5$ there is a reorientation of the spins from the hexagonal axis to the basal plane; but for the composition $x = 0.25$, as well as for the compound $PrCo_5$, the magnetic moment with lowering of the temperature becomes deflected from the hexagonal axis and takes a stable orientation at an angle less than $\pi/2$. But an unambiguous deduction about the character of these transitions cannot be made, since the anomalies of the physical properties in the reorientation range have not yet been studied in these compounds.

12. SPONTANEOUS SPIN-REORIENTATION TRANSITIONS IN CUBIC MAGNETS

Until recently, spin-reorientation transitions have been investigated principally in uniaxial magnets, although it was known that in a number of cubic ferro- and ferrimagnets a change of the axis of easy magnetization occurs with change of temperature.[74,75] This phenomenon, however, was not analyzed earlier from the point of view of the theory of spin-reorientation phase transitions.

It was first shown in a paper of Bozorth[76] that the direction of the magnetization vector in a cubic crystal at $H = 0$ depends on the signs and magnitudes of the first and second magnetic-anisotropy constants, and consequently a change of the anisotropy constants with temperature can lead to reorientation of the magnetization vector. We shall consider this question in greater detail, following[9].

In zero field, the TP for a single-domain cubic crystal can be written in the form

$$\Phi = \Phi_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2, \qquad (12.1)$$

where $K_1(T)$ and $K_2(T)$ are the first and second constants of cubic magnetic anisotroyy, and where α_i are the direction cosines of the magnetization vector. By going over to spherical coordinates (polar axis along

[100]), we can put (12.1) into the form

$$\Phi=\Phi_0+\frac{1}{4}K_1\sin^2 2\theta+\frac{1}{4}(K_1+K_2\cos^2\theta)\sin^4\theta\cos^2 2\varphi. \qquad (12.2)$$

On minimizing (12.2) with respect to θ and φ, we find that for all possible ratios of K_1 and K_2 minimization of the TP is achieved by orientation of the magnetization vector along only three different crystallographic directions:

$$\left.\begin{array}{lll} 1) & M \parallel \text{axis } \langle 100\rangle \text{ at } & K_1\geqslant 0, \\ 2) & M \parallel \text{axis } \langle 110\rangle \text{ at } & 0\geqslant K_1\geqslant -\frac{1}{2}K_2, \\ 3) & M \parallel \text{axis } \langle 111\rangle \text{ at } & K_1\leqslant -\frac{1}{3}K_2. \end{array}\right\} \qquad (12.3)$$

Equalities in the relations (12.3) correspond to lines of loss of stability of one or another phase; and, as follows from these relations (see also Fig. 35), there are regions in which different magnetic phases coexist.

Comparison of the energies of the various phases leads to the following relations for the phase-transition lines:

1) Rotation of M from axis $\langle 100\rangle$ to $\langle 110\rangle$ along the line $K_1=0,\ K_2\geqslant 0$,

2) Rotation of M from axis $\langle 110\rangle$ to $\langle 111\rangle$ along the line $9K_1+4K_2=0,\ K_1\leqslant 0$,

3) Rotation of M from axis $\langle 111\rangle$ to $\langle 100\rangle$ along the line $K_1+9K_2=0,\ K_1\geqslant 0$.

(12.4)

In a region of existence of one or another magnetic phase, change of the values of K_1 and K_2 may lead to interchange of the intermediate and hardest axes.

The complete magnetic phase diagram of a single-domain cubic ferromagnet in zero field, in coordinates K_1 and K_2, is shown in Fig. 35.

We note the following characteristic peculiarities of this phase diagram.

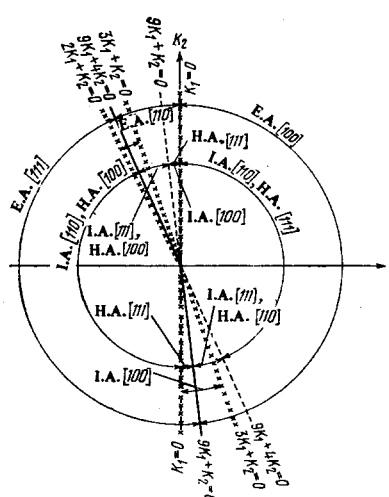

FIG. 35. Phase diagram of a single-domain cubic ferromagnet in zero magnetic field. Soldi lines are lines of phase transition; dashed lines are lines of interchange of intermediate and hardest axes; the crosses denote lines of loss of stability of the corresponding magnetic phases. Axes of magnetization; E. A., easy axis; I. A., intermediate axis; H. A., hard axis.

Since the easiest axes in a cubic ferromagnet can be only axes of type $\langle 100\rangle$, $\langle 110\rangle$, $\langle 111\rangle$, therefore, when only the first two constants of magnetic anisotropy are taken into account, spontaneous spin-reorientation transitions always occur discontinuously; that is, they are always phase transitions of the first kind. In this respect spin-reorientation transitions in a cubic magnet differ from such transitions in uniaxial magnets, in which, depending on the ratio of the first and second anisotropy constants, the spin-reorientation transitions may be either phase transitions of first or second order.

Transitions from the easiest axis $\langle 111\rangle$ to axis $\langle 110\rangle$ and from axis $\langle 111\rangle$ to axis $\langle 100\rangle$ occur with hysteresis, but a transition from axis $\langle 100\rangle$ to axis $\langle 110\rangle$ is (when two anisotropy constants are taken into account) an anhysteretic phase transition of first order, since in the last case the lines of loss of stability coincide with the phase-transition lines (see Fig. 35).

The discussion given above regarding the character and the hysteretic properties of spin-reorientation transitions in cubic ferromagnets relate to a single-domain specimen. In a many-domain specimen, the boundaries between domains may be regarded as "nuclei" of the new phase; therefore metastable states will not be realized, and a spin-reorientation transition in many-domain specimens will occur without hysteresis (for more details see Chap. 1 and[9]).

Spin-reorientation phase transitions are accompanied by anomalies of various physical properties of cubic ferromagnets. In a spin-reorientation region, where the domain structure becomes unstable and the anisotropy field decreases, external magnetic fields and mechanical stresses will produce the most intense reorganization of the domain structure and rotation of the magnetization vector. Therefore the initial susceptibility should pass through a maximum at the spin-reorientation point, and Young's modulus, which determines the stiffness of the crystal, through a minimum. The initial susceptibility and the magnetic part of Young's modulus, due to rotation processes, for a cubic ferromagnet with an equilibrium domain structure (that is, with a structure in which the volumes of domains along various easy directions are the same) are given, according to[9], by the following expressions:

a) Easiest axis $\langle 100\rangle$:

$$\bar{\chi}_B=\frac{1}{3}\frac{M_s^2}{K_1}\begin{cases}\sigma\parallel\langle 100\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=0,\\ \sigma\parallel\langle 110\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=\frac{3}{4}\frac{\lambda_{111}^2}{K_1},\\ \sigma\parallel\langle 111\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=\frac{\lambda_{111}^2}{K_1};\end{cases} \qquad (12.5)$$

b) easiest axis $\langle 111\rangle$:

$$\bar{\chi}_B=\frac{1}{2}\frac{M_s^2}{(1/3)|K_2|-K_1}\begin{cases}\sigma\parallel\langle 100\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=\frac{3}{2}\frac{\lambda_{100}^2}{(1/3)|K_2|-K_1},\\ \sigma\parallel\langle 110\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=\frac{3}{8}\frac{\lambda_{100}^2+\lambda_{111}^2}{(1/3)|K_2|-K_1},\\ \sigma\parallel\langle 111\rangle, & \Delta\overline{\left(\frac{1}{E}\right)}=\frac{\lambda_{111}^2}{(1/3)|K_2|-K_1};\end{cases} \qquad (12.6)$$

here M_s is the spontaneous magnetization, and λ_{111} and λ_{100} are the magnetostriction constants. It is evident

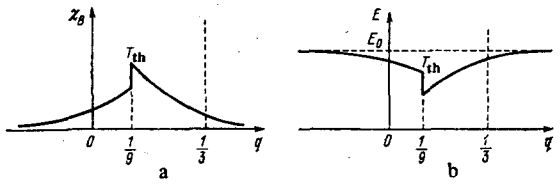

FIG. 36. Dependence of initial susceptibility (a) and of Young's modulus (b) of a cubic ferromagnet on $q = K_1/K_2$.

from the formulas that the size of the ΔE-effect depends on the direction of application of the external stresses σ. The theoretically calculated behavior of the susceptibility and of the ΔE-effect during spin reorientation in cubic ferromagnets is shown in Fig. 36.

13. SPIN REORIENTATION IN TERBIUM-YTTRIUM IRON GARNETS

As has been shown by magnetic-anisotropy measurements,[77] in certain mixed terbium-yttrium iron garnets ($Tb_xY_{3-x}Fe_5O_{12}$) the second magnetic-anisotropy constant K_2 is negative, and the first constant K_1 changes sign at a definite temperature. This is due to the fact that the contribution to K_1 from the terbium ions is positive, and from the iron a-d-sublattice negative. Accordingly, the constant K_1 at low temperatures is positive but at high temperatures, where the contribution from the terbium sublattice decreases sharply, becomes negative. This behavior of $K_1(T)$ leads to a spin-reorientation phase transition in these ferrites:

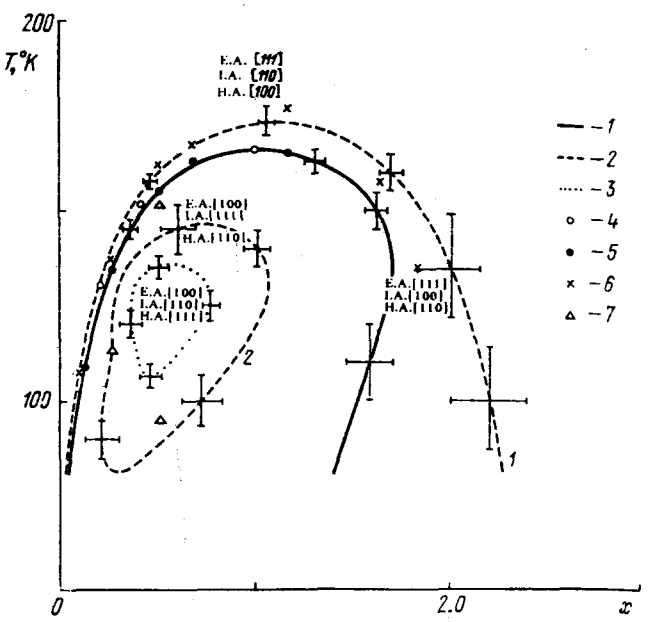

FIG. 37. Magnetic phase diagram of iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$. 1, line of spin-reorientation phase transition; 2, line of loss of stability of magnetic phase ⟨100⟩ (Curve 1) and of magnetic phase ⟨111⟩ (Curve 2); 3, line of interchange of intermediate and hard axes ⟨110⟩ and ⟨111⟩; 4, phase-transition temperatures obtained from measurements of susceptibility; 5, phase-transition temperatures obtained from measurements of Young's modulus; 6, temperatures of loss of stability of axis ⟨100⟩; 7, temperatures of loss of stability of axis ⟨111⟩.

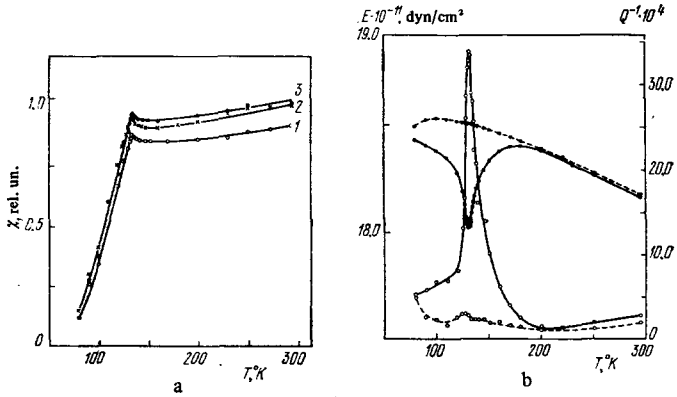

FIG. 38. Temperature dependence of the susceptibility of a monocrystal of ferrite-garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ (a) (Curve 1, along axis ⟨111⟩; Curve 2, along axis ⟨110⟩; Curve 3, along axis ⟨100⟩) and of Young's modulus (dark circles) and the internal friction (light circles) of polycrystalline ferrite-garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ (b).

with lowering of temperature, a reorientation occurs from the axis ⟨111⟩ to the axis ⟨100⟩.[9]

Figure 37 shows the magnetic phase diagram of the iron garnets $Tb_xY_{3-x}Fe_5O_{12}$ obtained in[9] and constructed according to the theoretical formulas (12.3)–(12.4), with use of experimental data on the temperature dependence of the magnetic-anisotropy constants from[77]. It was found that, in accordance with (12.5) and (12.6), at the spin-reorientation point the susceptibility passes through a maximum (Fig. 38a) and Young's modulus through a minimum (Fig. 38b); this also follows from the relations considered above, where it was shown that in many-domain specimens this transition occurs without hysteresis.

In[9] the temperature dependence of the orientation of a free monocrystal, located in a magnetic field, was also investigated; and from these data the temperatures of loss of stability of the various phases were determined.

As is seen from Fig. 37, at small x (up to 1.17) the elements of the phase diagram obtained by different methods agree, but at large terbium concentrations such agreement is lacking. This is explained by the influence of uniaxial anisotropy and of internal elastic stresses.[9]

14. SPIN-REORIENTATION TRANSITIONS IN INTERMETALLIC COMPOUNDS RFe_2

It was shown above that in cubic magnets, when the first two magnetic-anisotropy constants are taken into account, spin-reorientation transitions are phase transitions of the first kind, and the "angular" (canted) (in the sense defined above) phase is absent.

References[9,78] considered the influence of the third anisotropy constant on the magnetic phase diagram of a cubic ferromagnet. In[9] it was shown that if $K_3 \ll K_1$, K_2, then it has essentially no effect on the phase transitions ⟨111⟩⇄⟨100⟩ and ⟨111⟩⇄⟨110⟩, but in the transition ⟨110⟩⇄⟨100⟩ allowance for the third anisotropy con-

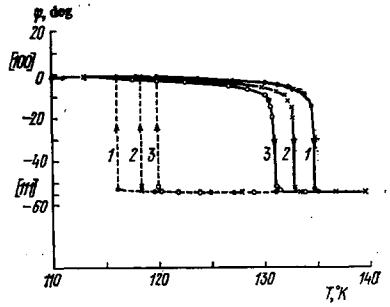

FIG. 39. Temperature dependence of the orientation of a monocrystalline disk of $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ freely suspended in a magnetic field. The solid lines correspond to rising temperature, the dashed lines to falling. Curve 1, $H = 5.0$ kOe; 2, $H = 10.0$ kOe; 3, $H = 16.1$ kOe.

stant leads to the occurrence of hysteresis in the transition (when $K_3 < 0$) or to the appearance of an "angular" phase (when $K_3 > 0$), in which the magnetic moments rotate in the (110) plane. In[78] the phase diagram of a cubic magnet was calculated numerically for values of all three magnetic-anisotropy constants that were comparable in magnitude; and it was shown that under these conditions a transition is also possible from phase ⟨111⟩ to phase ⟨110⟩ and from phase ⟨111⟩ to phase ⟨100⟩ through an intermediate angular phase, in which the magnetic moments rotate in the (110) plane.

Recently the presence of such a phase has been detected in measurements of the Mössbauer effect in cubic intermetallic compounds of the type RFe_2.[78-80] For example, in $CeFe_2$ below 150 °K the magnetization is parallel to the ⟨100⟩ axis, but above this temperature an angular structure develops and is retained up to the magnetic-ordering temperature of this compound[78] (Fig. 40); in $SmFe_2$ the easy axis in the temperature interval 140–240 °K turns from the axis ⟨110⟩ to the axis ⟨111⟩[79]; in $HoFe_2$ an angular structure develops below about 20 °K.[80] The transitions to the angular phase in these compounds, as was shown in[80], are accompanied by the occurrence of anomalies in the elastic moduli (Fig. 41).

Unfortunately, the magnetic anisotropy of these compounds has been studied very little, so that it is not possible to compare the phase diagrams obtained experimentally with those calculated theoretically with allowance for three magnetic-anisotropy constants.

CONCLUSION

From the experimental data presented in this review it follows that in rare-earth magnets there exists a great diversity of spin-reorientation transitions. Up to the present time there have been investigations, in the region of these transitions, of the magnetic and magnetoelastic characteristics, and this enables us to explain the basic thermodynamic properties of the indicated transitions.

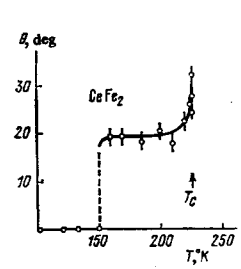

FIG. 40. Temperature dependence of the direction of easy magnetization in $CeFe_2$.

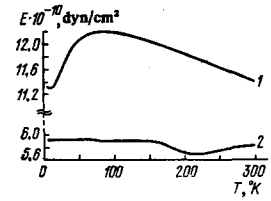

FIG. 41. Temperature dependence of Young's modulus for $HoFe_2$ (1) and for $CeFe_2$ (2).

It is of interest to study, in the vicinity of spin-reorientation transitions, the various kinetic characteristics (electrical and thermal conductivity, galvanomagnetic effects, etc.), which, as is well known, are sensitive to fluctuations of the magnetization during the transition. The obtaining of such data would make possible a substantial advance in the investigations of spin-reorientation transitions.

The greatest expectations lie in the wider application of nuclear methods of investigation (neutron diffraction, Mössbauer effect, NMR, etc.), which give information about the microscopic mechanisms of spin-reorientation transitions.

Undoubtedly there will be obtained, in the next few years, new data on spin-reorientation transitions in various magnetic materials. This will promote the further development of the theory of magnetism and of phase transitions and will also reveal the possibilities of technical use of materials possessing spin-reorientation transitions.

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