Spin-flip transitions in cubic magnets. Magnetic phase diagram of terbium–yttrium iron garnets

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Spin-flip phase transitions have been detected experimentally in iron-garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$, in the temperature interval 80–200°K, by measurements of the susceptibility, of Young's modulus, and of the torque. With lowering of the temperature, in consequence of the change of sign of the first anisotropy constant K_1, the magnetization vector flips from the [111] axis to the [100] axis. A theoretical calculation has been made of the spin-flip phase diagram of a cubic magnet, and the effect of domain structure on the character of the phase transition has been investigated. Consideration is given to the character of the anomalies in physical properties that occur during spin flip. It is shown that for terbium–yttrium iron garnets the phase diagram constructed on the basis of the known values of K_1 and K_2 is in good agreement with the phase diagram determined by the experimental data on the susceptibility, of Young's modulus, and of the torque.

1. INTRODUCTION

In recent years there has been very intensive investigation of magnetic phase transitions of the spin-reorientation type, in which, under the action of temperature, magnetic field, or elastic stresses, the direction of the magnetic moments changes with respect to the crystallographic axes. Transitions of the spin-reorientation type are a new class of magnetic phase transitions. In contrast to magnetic phase transitions of the order-disorder type at points of magnetic ordering, these transitions are order-order transitions.

Spin-reorientation (SR) phase transitions are accompanied by anomalies of various magnetic, magnetoelastic, and other properties of the magnets, and study of them gives important new information both for the general theory of phase transformations and for a more complete understanding of the nature and peculiarities of magnetic ordering in various substances. We note that experimental and theoretical investigations of SR transitions have so far been made almost exclusively on uniaxial magnets. Such transitions have been studied especially comprehensively in rare-earth orthoferrites[1]. Meanwhile it is known that in a number of cubic ferro- and ferrimagnets there is a change of the axis of easy magnetization with change of temperature, but these phenomena have not been analyzed from the point of view of SR phase transitions.

The present paper considers theoretically the conditions for and the character of magnetic phase transitions of the spin-reorientation type in a single-domain cubic ferro- or ferrimagnet in zero magnetic field, and the effect of domain structure on the transition parameters. It considers the anomalies of certain magnetic and magnetoelastic properties during SR transitions in cubic ferro- and ferrimagnets. On the basis of theoretical relations and of the known magnetic anisotropy constants[2], a magnetic phase diagram is constructed for the ferrite-garnet system $Tb_xY_{3-x}Fe_5O_{12}$. A comparison is made between the phase diagram constructed on the basis of the anisotropy constants and the phase diagram obtained from measurements of the initial susceptibility, Young's modulus, and other properties of these ferrite-garnets.

2. THEORY OF SPIN REORIENTATION IN A SINGLE-DOMAIN CUBIC FERROMAGNET

It was first shown by Bozorth[3] 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 therefore if the signs and magnitudes of the anisotropy constants change on change of temperature, this may lead to a reorientation of the magnetization vector. Bozorth started from the assumption that in a cubic crystal only the high-symmetry axes, of the type {100}, {110}, and {111}, can be easiest directions.

We shall consider how the orientation of the magnetization vector depends on the relations between the anisotropy constants in a cubic crystal; in contrast to Bozorth, we shall not restrict ourselves to the distinguished directions alone, but shall also find, along with the equilibrium directions of the magnetization vector, the conditions for existence of metastable states.

In zero field, for a single-domain cubic crystal, the free energy contains only magnetic-anisotropy energy. If we retain only two terms in the expansion of the magnetic-anisotropy energy with respect to the direction cosines, we can write the free-energy density in the form

$$F_A=K_1(T)\,(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_1^2\alpha_3^2)+K_2(T)\,\alpha_1^2\alpha_2^2\alpha_3^2+\ldots, \qquad (1)$$

where $K_1(T)$ and $K_2(T)$ are the first and second constants of cubic magnetic anisotropy, and where α_i are the direction cosines of the magnetization vector. Going over to spherical coordinates (the polar axis is [100]), we express (1) as

$$F_A={}^1/_4K_1(T)\sin^2 2\theta+[K_1(T)+K_2(T)\cos^2\theta]\sin^4\theta\cos^2 2\varphi. \qquad (2)$$

On minimizing (2) with respect to θ and φ, we find that for all possible relations of K_1 and K_2, a minimum of the free energy is produced only by orientations of the magnetization vector along three different crystallographic directions: of the type {100}(cube edge), of the type {110} (face diagonal, and of the type {111} (cube diagonal. For each of these directions, minima of the free energy are attained for definite relations between the anisotropy constants. For orientation along a [100] axis,

$$K_1 \geqslant 0; \qquad (3a)$$

for orientation along a [110] axis,

$$0 \geqslant K_1 \geqslant -{}^1/{}_2 K_2; \qquad (3b)$$

for orientation along a [111] axis,

$$K_1 \leqslant -{}^1/{}_3 K_2. \qquad (3c)$$

We note that along with the stable directions of magnetization in a cubic crystal, there are saddle points, lying in ⟨110⟩ planes; their directions are determined by the relation

$$\sin^2\theta = -2K_1/K_2. \qquad (4)$$

Equalities in the relations (3) correspond to lines of loss of stability of one or another phase; and as follows from these relations (see also Fig. 1), there are regions in which different magnetic phases coexist.

In order to determine the lines of phase transitions, it is necessary to compare with each other the values of the free energy of magnetic phases with different orientations of the magnetization. The results obtained agree with those of Bozorth[3]: transition from a [100] axis to a [110] axis under the condition

$$K_1 = 0, \quad K_2 \geqslant 0; \qquad (5a)$$

transition from a [110] axis to a [111] axis under the condition

$$9K_1 + 4K_2 = 0, \quad K_1 \leqslant 0; \qquad (5b)$$

transition from a [111] axis to a [100] axis under the condition

$$9K_1 + K_2 = 0, \quad K_1 \geqslant 0. \qquad (5c)$$

We notice that within the range of existence of one or another magnetic phase, with change of the values of K_1 and K_2 there may be a change of the intermediate and hardest axes: for easiest axis [100], the hardest axis is

$$\begin{aligned} &[111] \quad \text{for} \quad 9K_1 + 4K_2 > 0, \quad K_1 \geqslant 0, \\ &[110] \quad \text{for} \quad 9K_1 + 4K_2 < 0, \quad K_1 \geqslant 0; \end{aligned} \qquad (6a)$$

for easiest axis [110], the hardest axis is

$$\begin{aligned} &[111] \quad \text{for} \quad 9K_1 + K_2 > 0, \quad K_1 \leqslant 0, \\ &[100] \quad \text{for} \quad 9K_1 + K_2 < 0, \quad K_1 \leqslant 0 \end{aligned} \qquad (6b)$$

for easiest axis [111], the hardest axis is

$$\begin{aligned} &[100] \quad \text{for} \quad K_1 < 0, \\ &[110] \quad \text{for} \quad K_1 > 0. \end{aligned} \qquad (6c)$$

We note that in zero field, a change of the intermediate and hardest axes does not constitute a phase transition, since under these conditions no reorganization of the magnetic structure occurs.

The complete magnetic phase diagram of a single-domain cubic ferromagnet in zero field, constructed on the basis of formulas (3), (5), and (6) in coordinates (K_1, K_2), is shown in Fig. 1. We notice the following characteristic features of this phase diagram.

With allowance only for the first and second magnetic-anisotropy constants, the easiest axes in a cubic ferromagnet can be only axes of the type {100}, {110}, and {111}; there are no equilibrium "angular" phases, i.e., states in which the direction of the magnetization vector fails to coincide with these axes. Hence, it follows that within the framework of this approximation, the SR phase transitions under consideration can be only first-

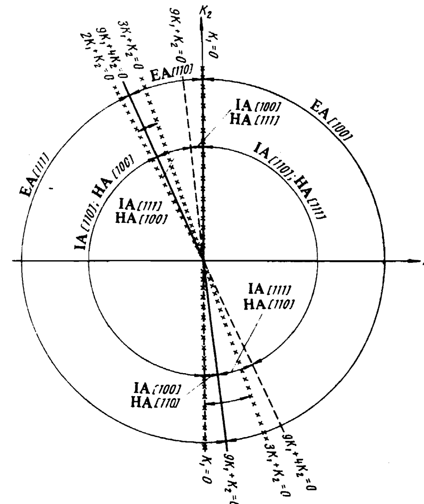


FIG. 1. Phase diagram of a single-domain cubic ferromagnet in zero magnetic field. Solid lines are phase-transition lines. Dashed lines are lines of interchange of the intermediate (IA) and hardest (HA) axes. Plotted with crosses are the lines of stability loss of the corresponding magnetic phases.

order transitions. In this respect SR transitions in a cubic ferromagnet differ from such transitions in a uniaxial ferromagnet, in which, depending on the magnitudes and signs of the first and second anisotropy constants, SR transitions may be phase transitions either of the first or of the second order.

Analysis of the phase diagram of a cubic ferromagnet shows also that reorientation of the magnetization vector from a [111] axis to a [110] axis or from a [111] axis to a [100] axis is accomplished with hysteresis, whereas reorientation from a [100] axis to a [110] axis (with allowance for two anisotropy constants) is a hysteresis-less phase transition of the first kind, since in the latter case the lines of stability loss coincide with lines of phase transition (see Fig. 1)[1)].

3. EFFECT OF DOMAIN STRUCTURE ON THE CHARACTER OF SPIN-REORIENTATION TRANSITIONS IN A CUBIC FERROMAGNET

The arguments presented above regarding the character and the hysteretic properties of SR transitions in a cubic ferromagnet relate to a single-domain specimen. In a many-domain specimen, the boundaries between domains can be regarded as "nuclei" of the new phase, since within the boundaries there are always sections in which the direction of the magnetization coincides with the direction of the magnetization in the new phase; therefore metastable states will not be realized here, and the SR transition in many-domain specimens will occur without hysteresis.

We shall consider the effect of domain structure in more detail.

The free energy of a system with magnetic inhomogeneities is

$$F = \int_{-\infty}^{+\infty} \{F_A(\theta,\varphi) + A[(d\theta/dx)^2 + \sin^2\theta\,(d\varphi/dx)^2]\}\,dx. \qquad (7)$$

Here x is a coordinate, and A is an exchange constant.

A minimum of the free energy is attained when $\theta(x)$ and $\varphi(x)$ satisfy the Euler equations

$$2\frac{d^2\theta}{d\xi^2}=\frac{\partial \bar{F}_A}{\partial\theta}+\sin 2\theta\left(\frac{\partial\varphi}{\partial\xi}\right)^2,\qquad 2\frac{d}{d\xi}\left[\sin^2\theta\frac{d\varphi}{d\xi}\right]=\frac{\partial \bar{F}_A}{\partial\varphi}, \tag{8}$$

where the following new symbols have been introduced:

$$\bar{F}_A=\frac{F_A}{|K_2|},\qquad \xi=\frac{x}{(A/|K_2|)^{1/2}}.$$

The system of equations (8) has the first integral

$$\left(\frac{d\theta}{d\xi}\right)^2+\sin^2\theta\left(\frac{d\varphi}{d\xi}\right)^2-\bar{F}_A(\theta,\varphi)=E=\text{const}. \tag{9}$$

In general both unknown functions θ and φ of the system (8) depend on x. We shall be interested in solutions with φ = const, which corresponds to rotation of the magnetization in a single plane. For such solutions

$$\partial\bar{F}_A/\partial\varphi=0,\quad \partial^2\bar{F}_A/\partial\varphi^2>0. \tag{10}$$

Analysis of these equations shows that there exist two groups of solutions for φ. The first group corresponds to rotation of the magnetization in planes of type $\langle 100\rangle$ ($\varphi = 0, \pi/2, \pi, 3\pi/2$), the second to rotation of the magnetization in planes of type $\langle 110\rangle$ ($\varphi = \pi/4, 3\pi/4, 5\pi/4, 7\pi/4$). The stability of the solutions depends on the ratio $q = K_1/|K_2|$ (here and hereafter we consider the case most interesting from the point of view of our experiments, $K_2 < 0$).

The planes of type $\langle 100\rangle$ are stable for $q > 1/2$; near $q = 1/2$, directions lying in these planes become unstable if they are close to directions with $\theta = \pi/4 + n\pi/4$, and for $q = 0$ all directions in $\langle 100\rangle$ planes become unstable.

The planes of type $\langle 110\rangle$ are stable for $q < 0$; for $q \approx 0$, directions close to the directions with $\theta = n\pi/2$ lose their stability, and for $q = 1$ all directions in the plane lose their stability.

Let $q < 0$. Then, as was shown above, the magnetization lies in $\langle 110\rangle$ planes. We shall consider the behavior of a domain boundary in this case. Equation (9) transforms to the form

$$(d\theta/d\xi)^2=\bar{F}_A(\theta)+E, \tag{11}$$

where the constant E for an isolated domain wall is chosen from the following conditions (see above):

$$q<{}^1/_9,\quad \left.\frac{d\theta}{d\xi}\right|_{\xi=\pm\infty}=\left.\frac{d\theta}{d\xi}\right|_{\theta=\pm\arcsin\sqrt{2/3}}=0,\qquad q>{}^1/_9,\quad \left.\frac{d\theta}{d\xi}\right|_{\xi=\pm\infty}=\left.\frac{d\theta}{d\xi}\right|_{\theta=0,\,\pi/2}=0. \tag{12}$$

By use of (12), Eq. (11) is easily transformed to the form

$$q<{}^1/_9,\quad \frac{d\theta}{d\xi}=\pm\frac{1}{2}\left(\sin^2\theta-\frac{2}{3}\right)\left[\sin^2\theta-3\left(q-\frac{1}{9}\right)\right]^{1/2},\qquad q>{}^1/_9,\quad \frac{d\theta}{d\xi}=\pm\frac{1}{2}\sin\theta\left[\sin^4\theta-(1+3q)\sin^2\theta+4q\right]^{1/2}. \tag{13}$$

Figure 2 depicts the phase portrait of the system described by Eqs. (13), for various values of the parameter q (the figure shows only separatrices of the family of curves of (13), corresponding to isolated domain walls). From the figure it is evident that for $q < 0$ two types of domain walls are realized: the line ABC corresponds to a 110-degree neighborhood, the line CDE to a 70-degree neighborhood. Since, as has already been indicated, directions close to $\theta = \pm\pi/2$ lose their stability already for $q = 0$, while directions with $\theta = 0, \pi$ retain their stability up to $q = 1$, therefore we shall consider the most stable 110-degree neighborhoods, in which θ changes around 0 or π. In Fig. 2 such a neighborhood corresponds to the section ABC; the middle of the domain wall coincides with the point B (Fig. 2a). For $0 < q < 1/9$ (Fig. 2b), at the middle of the domain wall ($\theta = 0$) there is a kink, which may be considered a "nucleus" of the new magnetic phase, with magnetization direction along an axis of type $\{100\}$. With increase of q, the magnitude of the kink increases: the domain wall expands, and for $q = 1/9$ there appears a new domain with magnetization along a $\{100\}$ axis ($\theta = 0$, Fig. 2c).

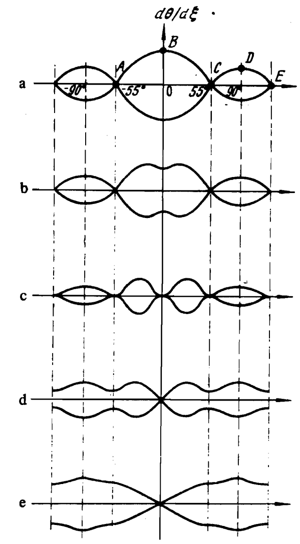


FIG. 2. Phase portrait of the system described by equations (13): a), $q < 0$; b), $0 < q < {}^1/_9$; c), $q = {}^1/_9$; d), ${}^1/_9 < q < {}^1/_3$; e), $q = {}^1/_3$.

On further increase of q above 1/9, the separatrix "disengages" from the axis of abscissas, and the ordinates of points A and C increase (Fig. 2d). This means that domains of phase $\{111\}$ decrease in size and are transformed to kinks of domain walls that separate domains of phase $\{100\}$. For $q = 1/3$, the kinks of domain walls vanish (Fig. 2e).

The analysis presented shows[2)] that a transition from one phase ($\{111\}$) to another ($\{100\}$) can occur by continuous growth of the new phase from a domain wall, which thus emerges as a "nucleus" of the new phase. We note that domain boundaries as "nuclei" of a new magnetic phase differ from the nuclei usually considered in the theory of first-order phase transitions, in that they are stable and grow in a region where the old phase is also still thermodynamically stable.

4. ANOMALIES OF PHYSICAL PROPERTIES DURING SPIN REORIENTATION IN CUBIC FERROMAGNETS

SR phase transitions are accompanied by anomalies of various physical properties of ferromagnets. For example, the latent heat of a SR transition can be determined by comparing the entropies of the different phases at the transition temperature T_t:

$$Q_{\{110\}\to\{100\}}=-{}^1/_4 T_t\, dK_1/dT|_{T_t},\qquad Q_{\{111\}\to\{100\}}=-{}^1/_3 T_t\,[dK_1/dT+{}^1/_9 dK_2/dT]|_{T_t},\qquad Q_{\{111\}\to\{110\}}=-{}^1/_3 T_t\,[{}^1/_4 dK_1/dT+{}^1/_9 dK_2/dT]|_{T_t}. \tag{14}$$

In many-domain specimens, because the transition is "smeared out", the evolution of heat will occur over a finite region near the transition.

In general, it must be mentioned that in cubic ferromagnets the domain structure significantly affects the anomalies of properties at the transition point. Thus it is easy to show that in specimens with an equilibrium domain structure (that is, in which the volumes of domains with magnetization along different easiest directions are equal), no anomalies of the thermal expansion will be observed during SR transitions, since, although such anomalies are present in each individual domain, for the specimen as a whole they compensate each other (when account is taken only of the first two magnetostriction constants λ_{111} and λ_{100}).

We shall consider the behavior of the initial susceptibility χ and of Young's modulus E of a cubic ferromagnet in a region of spin reorientation.

The value of χ is made up of two components: the susceptibility due to processes of rotation of the magnetization vector (χ_r), and the susceptibility due to displacement of domain boundaries (χ_d). Both these mechanisms also make a contribution to Young's modulus E, since they lead to additional magnetostrictive deformations. It is in the region of spin reorientation, where the domain structure becomes unstable and the anisotropy field decreases, that external magnetic fields and mechanical stresses will cause the most intensive reorganization of the domain structure and rotation of the magnetization vector. Therefore the initial susceptibility should go through a maximum at the spin-reorientation point, and Young's modulus, which determines the stiffness of the crystal, through a minimum.

We shall calculate the contribution to the initial susceptibility and to Young's modulus from rotation processes[3)].

Let $\beta = (\beta_1, \beta_2, \beta_3)$ be the direction of application of the external field H, $\alpha(\theta, \phi) = (\alpha_1, \alpha_2, \alpha_3)$ the direction of the magnetization vector. The free-energy density of a cubic ferromagnet, which consists of the energy of the external field, the magnetoelastic energy, and the anisotropy energy, we shall write in the form

$$F=-I_sH\Phi(\theta,\varphi)-\lambda_s\sigma\Psi(\theta,\varphi)+F_A(\theta,\varphi). \qquad (15)$$

Here

$$\Phi(\theta,\varphi)=\alpha_1(\theta,\varphi)\beta_1+\alpha_2(\theta,\varphi)\beta_2+\alpha_3(\theta,\varphi)\beta_3,$$

$$\Psi(\theta,\varphi)=\frac{3}{2}\frac{\lambda_{100}}{\lambda_s}\left[\alpha_1^2(\theta,\varphi)\beta_1^2+\alpha_2^2(\theta,\varphi)\beta_2^2+\alpha_3^2(\theta,\varphi)\beta_3^2-\frac{1}{3}\right] \qquad (16)$$
$$+3\frac{\lambda_{111}}{\lambda_s}\left[\alpha_1(\theta,\varphi)\alpha_2(\theta,\varphi)\beta_1\beta_2+\alpha_2(\theta,\varphi)\alpha_3(\theta,\varphi)\beta_2\beta_3+\alpha_1(\theta,\varphi)\alpha_3(\theta,\varphi)\beta_1\beta_3\right],$$

σ is the elastic stress. By minimizing (15) with respect to θ and φ and differentiating the resulting equations with respect to σ and H, one can obtain

$$\chi_r=\left(\frac{\partial I}{\partial H}\right)_r=I_s^2\left[\frac{1}{(F_A)_{\theta\theta}}\left(\frac{\partial\Phi}{\partial\theta}\right)^2+\frac{1}{(F_A)_{\varphi\varphi}}\left(\frac{\partial\Phi}{\partial\varphi}\right)^2\right],$$
$$\Delta\left(\frac{1}{E}\right)_r=\left(\frac{\partial\lambda}{\partial\sigma}\right)_r=\lambda_s^2\left[\frac{1}{(F_A)_{\theta\theta}}\left(\frac{\partial\Psi}{\partial\theta}\right)^2+\frac{1}{(F_A)_{\varphi\varphi}}\left(\frac{\partial\Psi}{\partial\varphi}\right)^2\right], \qquad (17)$$

where $\Delta(1/E)_r$ is the magnetoelastic contribution to the Young's modulus of the ferromagnet, defined by the relation

$$\frac{1}{E}=\frac{1}{E_0}+\Delta\left(\frac{1}{E}\right), \qquad (18)$$

E_0 is Young's modulus without allowance for magnetoelastic interaction.

By use of the relations (16), one can obtain an explicit form of the expressions for the initial susceptibility χ_r and $\Delta(1/E)_r$ for each phase for various directions of the measurement. For many-domain specimens, it is necessary to average the expressions for χ_r and $\Delta(1/E)_r$ over the various domains. Then, as is easily shown, for the equilibrium domain structure the susceptibility is isotropic and is, for easiest axis [100],

$$\bar{\chi}_r={}^1/_3I_s^2/K_1, \qquad (19a)$$

and for easiest axis [111]

$$\bar{\chi}_r=\frac{1}{2}\frac{I_s^2}{|K_2|/3-K_1}. \qquad (19b)$$

Expressions for $\Delta(1/E)_r$ are obtained in analogous fashion. But in this case the value of $\Delta(1/E)_r$ averaged over all domains is not a scalar but depends on the directions of the applied elastic stresses: for easiest axis [100],

$$\sigma\|[100],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r=0;$$
$$\sigma\|[110],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r={}^3/_4\frac{\lambda_{111}^2}{K_1}; \qquad (20a)$$
$$\sigma\|[111],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r=\frac{\lambda_{111}^2}{K_1};$$

for easiest axis [111],

$$\sigma\|[100],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r=\frac{3}{2}\frac{\lambda_{100}^2}{{}^1/_3|K_2|-K_1};$$
$$\sigma\|[110],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r=\frac{3}{8}\frac{\lambda_{111}^2+\lambda_{100}^2}{{}^1/_3|K_2|-K_1}; \qquad (20b)$$
$$\sigma\|[111],\quad \Delta\left(\frac{\bar{1}}{E}\right)_r=\frac{\lambda_{111}^2}{{}^1/_3|K_2|-K_1};$$

Figure 3 shows the dependence of the initial susceptibility (a) and of Young's modulus (b) on q. The sharp minimum in E and maximum in χ_r at T_t are actually more spread out because of the contribution to the free energy from the domain wall energy, which was disregarded in the calculation performed.

5. SPECIMENS AND METHODS OF MEASUREMENT

Investigations of spin-reorientation phenomena were made on monocrystalline and polycrystalline iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$ ($0 \le x \le 3$). The methods of growing the monocrystals and of controlling their compositions were described in[2]. The polycrystalline specimens of mixed iron garnets were prepared by the usual ceramic technology.

The initial susceptibility in directions {100}, {110}, and {111} was measured on single-crystal disks cut in a (110) plane, by the change of resonance frequency of an LC circuit when the specimen under study was introduced into the inductance coil. The susceptibility measurements were made at frequency ~200 kHz. Since the space factor of the coil was unknown, only the relative value of the susceptibility was determined.

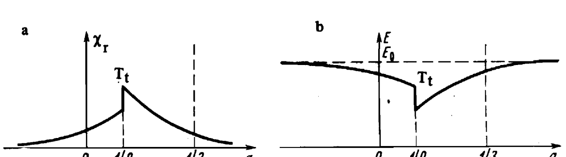


FIG. 3. Dependence of initial susceptibility (a) and Young's modulus (b) of a cubic ferromagnet on $q = K_1/|K_2|$. T_t is the temperature of the spin-reorientation phase transition.

Measurements of the elastic modulus and of the internal friction were made on polycrystalline specimens by the composite oscillator method[6] at frequency ~150 kHz.

We also measured the temperatures of loss of "stability" of crystallographic directions {100} and {111} in a magnetic field. For this purpose a single-crystal disk, cut in a (110) plane, was hung pseudo-freely (on a very thin thread) in an electromagnet so that the field was parallel to one of these directions, and it was determined how the orientation of the disk changed with change of temperature. Measures were taken to eliminate parasitic torques that arise from the elasticity of the thermocouple fastened to the specimen and from other causes.

6. EXPERIMENTAL RESULTS

As has been shown by measurements of the magnetic anisotropy[2], in mixed terbium-yttrium iron garnets the second anisotropy constant K_2 is negative, while the first constant K_1 changes sign at a certain temperature. This is due to the fact that the contribution to K_1 from the terbium ions is positive, while the contribution from the iron (a–d)-sublattice is negative. Since at low temperatures the anisotropy of the ferrite is determined chiefly by the terbium sublattice, the constant K_1 is positive at low temperatures; but at high temperatures, when the contribution from the terbium sublattice decreases sharply, it changes sign. This leads to a SR phase transition in these ferrites with change of temperature.

Figure 4 shows the magnetic phase diagram of iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$, constructed according to formulas (3), (5), and (6) with use of experimental anisotropy data from[2].[4)]

Figure 5 shows the temperature dependence of the susceptibility along various crystallographic directions for a single crystal of $Tb_{0.26}Y_{2.74}Fe_5O_{12}$. It is evident that at high temperatures the susceptibility depends little on temperature, that it goes through a maximum at 134 K, and that it drops abruptly on further lowering of the temperature (in consequence of the sharp increase of the anisotropy constant). In accordance with what was set forth above, we suppose that the temperature of the maximum corresponds to the SR transition temperature T_t. As is shown by a comparison of Figs. 4 and 5, for this specimen the SR transition temperature T_t determined from susceptibility measurements agrees, within the limits of error, with the T_t calculated from data on the anisotropy constant.

Figure 6 shows the temperature dependence of Young's modulus and of the internal friction for a polycrystalline specimen of approximate composition $Tb_{0.2}Y_{2.8}Fe_5O_{12}$. It is evident that at temperature 130.5 K a minimum occurs in Young's modulus; the magnetic field removes this anomaly, an indication of its magnetic nature. The temperature of the minimum is the SR transition temperature and also agrees well with the value of T_t calculated from measurements of the anisotropy constant (Fig. 4). As is evident from Fig. 6, the SR transition is accompanied by a maximum of the internal friction, caused by losses during reorganization of the domain structure and rotation of the magnetization under the influence of elastic stresses.

Our measurements showed that hysteresis of the transition is absent in many-domain specimens: the temperatures of the anomaly of the susceptibility and

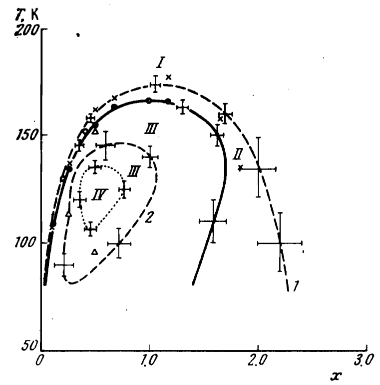


FIG. 4. Magnetic phase diagram of iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$: I, EA [111], IA [110], HA [100]; II, EA [111], IA [100], HA [110]; III, EA [100], IA [111], HA [110]; IV, EA [100], IA [110], HA [111]. The solid curve is the spin-reorientation phase-transition line; the dashed curves are the lines of stability loss of the magnetic phase {100} (1) and of the magnetic phase {111} (2); the dotted curve is the line of interchange of the intermediate and hardest axes [110] and [111]. The errors of the determination from the experimental data on K_1 and K_2 [2] are shown for the lines of phase transition, of stability loss, and of interchange of intermediate and hardest axes. The points show the phase-transition temperatures obtained from measurements of the susceptibility (●) and from measurements of Young's modulus (○), and the temperatures of stability loss of phase {100} (X) and of phase {111} (Δ).

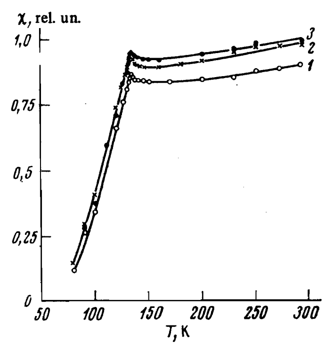


FIG. 5

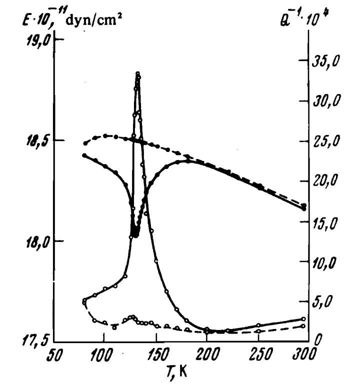


FIG. 6

FIG. 5. Temperature dependence of the susceptibility of single-crystal iron garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$: curve 1, along axis [111]; 2, along axis [110]; 3, along axis [100].

FIG. 6. Temperature dependence of Young's modulus (●) and of the internal friction (○) of polycrystalline iron garnet $Tb_{0.2}Y_{2.8}Fe_5O_{12}$. Solid lines are measurements at H = 0, dashed lines are measurements at H = 2.0 kOe.

of Young's modulus obtained on heating and on cooling are the same. This agrees with the theoretical considerations presented above.

We obtained similar $\chi(T)$ and $E(T)$ curves for compositions with $0.1 \le x \le 1.17$; this enabled us to construct lines of SR phase transition according to these measurements and to compare them with the lines determined from anisotropy measurements, see Fig. 4. As is evident from this figure, the temperatures of the SR phase transition determined by the different methods agree for compositions with $x \le 1.17$. But for compositions with x = 1.65 (single crystal) and x = 1.5 (polycrystal), we observed no anomaly in the temperature variation of the susceptibility and of Young's modulus, although according to the anisotropy measurements there should be a transition of the spin-reorientation type in these also.

Two possible explanations can be given for this. First, in compounds with large x the magnetostriction increases abruptly[2], and the determining factor impeding motion of domain boundaries under the action of external influences becomes the internal stresses. Since the energy of these stresses ($\sim\lambda\sigma$) does not experience a singularity at the point of spin reorientation, in such specimens there is no anomaly of χ and E at the transition point. This point of view agrees with our experiments, which showed that prolonged annealing of the specimens increases the value of the susceptibility and renders more abrupt its maximum at the spin-reorientation point. The other possible reason is the following. The phase-transition line was constructed on the assumption that in the ferrite-garnets studied there is only cubic anisotropy. In actuality, in mixed iron garnets there is present, besides, an additional uniaxial growth-induced anisotropy[2, 7]. Uniaxial anisotropy may lead to a difference of the measured phase diagram from the diagram constructed theoretically. Its influence will show up especially strongly in specimens with $1 \lesssim x \lesssim 2$, since in them the uniaxial anisotropy reaches its highest value, while the first cubic-anisotropy constant is small over a broad temperature interval, so that the uniaxial growth-induced anisotropy shifts strongly the point of spin reorientation.

We also investigated the temperature dependence of the orientation of a free single-crystal disk placed in a magnetic field. Since the measurements are made in a field, the specimen is single-domain, and therefore the rotation of the magnetization vector from one direction to another (which in this case corresponds to a rotation of the specimen in the field) occurs at the temperature at which a given phase loses its stability.

Figure 7 shows the temperature dependence of the angle of rotation of a free single-crystal disk of iron garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ in various fields. At high temperatures the [111] direction is stable; therefore in this temperature range, the specimen is oriented with direction [111] along the field. Cooling leads to the result that at a certain temperature $T_{\{111\}}$ there occurs a rotation of the specimen, with direction [100] along the field: at this temperature, the phase {111} loses its stability. If we now raise the temperature of the specimen, then rotation of it from axis [100] to axis [111] will occur at the temperature $T_{\{100\}}$ at which phase {100} loses its stability; and $T_{\{100\}} > T_{\{111\}}$.

As is evident from Fig. 7, the temperature at which one or another phase loses its stability depends on the field. This is due to the fact that, as has already been mentioned, the anisotropy constants of the ferrite-garnets investigated depend on the field[2]. The field-dependence of $T_{\{100\}}$ and $T_{\{111\}}$ for the ferrite-garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ is shown in Fig. 8. The values of $T_{\{100\}}$ and $T_{\{111\}}$ extrapolated to zero field are, respectively, 136.4 K and 114.6 K. The temperatures of stability loss for other ferrite-garnets were determined by a similar method. As is evident from Fig. 4, at small x the temperatures of stability loss determined by this method for phases {111} and {100} agree with those calculated from the anisotropy constants. On increase of the concentration of terbium in the garnet, $T_{\{100\}}$ and $T_{\{111\}}$ as determined by different methods begin to differ from one another. Apparently this is due to the influence of uniaxial growth-induced anisotropy.

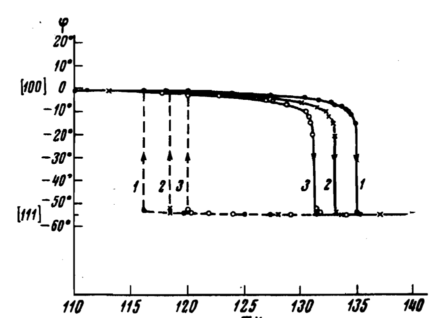


FIG. 7. Temperature dependence of the orientation of a single-crystal disk of $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ freely suspended in a magnetic field. The solid lines correspond to raising and the dashed to lowering of the temperature: 1, H = 5.0 kOe; 2, H = 10.0 kOe; 3, H = 16.1 kOe.

7. CONCLUSION

The results of the present work can be summarized as follows.

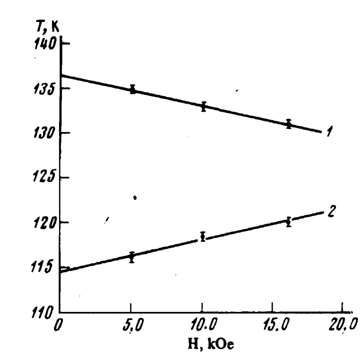
The theoretical calculation presented for the magnetic phase diagram of a single-domain cubic ferromagnet, with allowance for the first and second magnetic-anisotropy constants, shows that, because the easiest axes of such a magnet can be only high-symmetry axes of the type {100}, {110}, and {111}, therefore the transitions of spin-reorientation type resulting from change of the anisotropy constants are first-order phase transitions.

Allowance for the domain structure of a cubic crystal leads to removal of the hysteresis of the transition, which then occurs in continuous fashion; the domain boundaries emerge in the role of "nuclei" of the new magnetic phase. Near the transition point, both the new and the old magnetic phases simultaneously coexist.

On the basis of the theoretically obtained relations, with use of the known values of the magnetic-anisotropy constants K_1 and K_2 of terbium-yttrium iron garnets, a magnetic SR phase diagram has been constructed for these ferrimagnets at H = 0, in the (x–T) plane.

In the temperature interval 80–300 K, Young's modulus and the susceptibility have been measured for the system $Tb_xY_{3-x}Fe_5O_{12}$. Anomalies of Young's modulus and of the susceptibility were detected at the temperature of the transition of spin-reorientation type; their character is in agreement with the theoretical calculations presented in the paper: Young's modulus goes through a minimum at the spin-reorientation temperature, the susceptibility through a maximum. The SR transition in this system, in accordance with theory, occurs in continuous fashion without hysteresis. The phase-transition points determined from these experiments agree with the theoretical curve constructed on the basis of the known values of K_1 and K_2.

FIG. 8. Dependence on external magnetic field of the temperature of stability loss of axis: $T_{\{100\}}$(1) and $T_{\{111\}}$(2), for iron garnet $Tb_{0.26}Y_{2.74}Fe_5O_{12}$.



The lines of stability loss have been determined for the phases with easiest axis [100] and with easiest axis [111]. The agreement with the theoretically constructed phase diagram is completely satisfactory.

The experiments done to investigate SR phase transitions in iron garnets of the system $Tb_xY_{3-x}Fe_5O_{12}$ show good agreement with the theory of these transitions developed for cubic ferro- and ferrimagnets.

1)When a third term is included in the expansion of the energy of cubic magnetic anisotropy, the nature of the transition will depend on the sign of the third anisotropy constant K_3. For $K_3 < 0$, the transition between phases {110} and {100} will be a first-order transition with hysteresis, whose width is determined by the magnitude of K_3; whereas for $K_3 > 0$, this transition will occur smoothly, by formation of an "angular" phase within a certain interval of values of the anisotropy constants. The nature of the phase transitions between other phases, with allowance for K_3, is practically unchanged.

2)A similar phenomenon of growth of a "nucleus" of a new phase from a domain wall during reorientation of spins in uniaxial magnetic materials was considered theoetically in [4].

3)Quantitative estimates of the effect of displacement processes on χ and E are difficult, since at present there is no complete theory, but only approximate calculations of displacement processes for various special models. For example, in Kersten's model [5], in which bending under the influence of a field is considered for a domain wall clamped at the ends, the initial susceptibility from displacement satisfies the relation $\chi_d \sim I_s^{3/2}/K_{eff}^{3/4}$; consequently, at a SR transition point, where K_{eff} is minimal, χ_d should go through a maximum.

4)In [2] it was shown that the anisotropy constants of terbium-yttrium iron garnets depend on the field. The phase diagram in Fig. 4 was constructed on the basis of values of K_1 and K_2 extrapolated to zero field.

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