

Spin reorientation in dysprosium yttrium iron garnets $Dy_x Y_{3-x} Fe_5 O_{12}$

G. A. Babushkin, V. A. Borodin, V. D. Doroshev, R. Z. Levitin, V. N. Orlov, A. I. Popov, and T. N. Tarasenko

Physicotechnical Institute, Academy of Sciences of the Ukrainian SSR, Donetsk

(Submitted 13 March 1984)

Zh. Eksp. Teor. Fiz. **87**, 989–998 (September 1984)

Measurements of the magnetization, initial susceptibility, torque, and ^{57}Fe NMR were combined with x-ray diffraction methods in an investigation of a spontaneous first-order spin-reorientation phase transition from high-symmetry $\langle 111 \rangle$ phases to canted $\langle uuv \rangle$ phases in iron garnets belonging to the $Dy_x Y_{3-x} Fe_5 O_{12}$ system ($0.2 \leq x \leq 3$). It was found that the transition temperature T_i is practically independent of the dysprosium concentration and it amounts to 13.5 ± 1.5 K. A theoretical model is developed in which an allowance is made for the isotropic rare-earth-iron exchange interaction and for the crystal field acting on rare-earth ions. An orientational phase diagram common to ferrites with the garnet structure is constructed on the basis of this model. It is shown that the model accounts satisfactorily for the experimentally observed characteristics of the $\langle 111 \rangle \leftrightarrow \langle uuv \rangle$ phase transition in dysprosium yttrium iron garnets.

Recent investigations have shown that rare-earth iron garnets with the formula $R_3 Fe_5 O_{12}$ (R is a rare earth or yttrium) exhibit spontaneous spin-reorientation transitions: variation of temperature causes the easy magnetization axis to change its orientation from one crystallographic direction to another.

In ferrimagnets such as rare-earth iron garnets the simplest mechanism which can give rise to spontaneous spin-reorientation phase transitions is a competition between the contributions made to the magnetic anisotropy by different magnetic subsystems (sublattices). This is true of ytterbium yttrium $Tb_x Y_{3-x} Fe_5 O_{12}$ (Refs. 1 and 2) and erbium yttrium $Er_x Y_{3-x} Fe_5 O_{12}$ (Ref. 3) iron garnets, for which the first magnetocrystalline anisotropy constants of the rare-earth and iron sublattices have opposite signs. In view of the different temperature dependences of these contributions, at some temperature there is a change in the sign of the net magnetic anisotropy constant of rare-earth iron garnets and a $\langle 100 \rangle \leftrightarrow \langle 111 \rangle$ spin-reorientation phase transition takes place. Since at low temperatures the anisotropy of the iron sublattice is considerably less than the anisotropy of the rare-earth sublattice, it follows that such transitions occur at relatively high temperatures.

However, we may also encounter physically more interesting situations when spin reorientation occurs because variation of temperature alters the state of the rare-earth subsystem and the influence of the magnetic anisotropy of the iron sublattice on this transition is not important. Spin-reorientation phase transitions of this type usually occur at lower temperatures. This is the nature of spontaneous reorientation transitions $\langle uv0 \rangle \leftrightarrow \langle 110 \rangle \leftrightarrow \langle 111 \rangle$ that occur in samarium iron garnet $Sm_3 Fe_5 O_{12}$ in which changes in the direction of the easy magnetization axis are due to filling of excited energy levels of the Sm^{3+} ion on increase in temperature.⁴ This mechanism makes it possible to describe the changes in the temperatures of spin-reorientation transitions in samarium yttrium iron garnets $Sm_x Y_{3-x} Fe_5 O_{12}$ on replacement of samarium with nonmagnetic yttrium.⁵

A similar situation should occur also in dysprosium yt-

trium iron garnets $Dy_x Y_{3-x} Fe_5 O_{12}$. It is known that at high temperatures ($T > 78$ K) the easy magnetization axes of dysprosium iron garnet are parallel to directions of the $\langle 111 \rangle$ type in a cubic crystal.⁶ However, Clark and Callen⁷ discovered that at 4.2 K none of the principal crystallographic directions $\langle 111 \rangle$, $\langle 110 \rangle$, or $\langle 100 \rangle$ is the easy magnetization axis. It therefore must follow that in $Dy_3 Fe_5 O_{12}$ there should be a spin reorientation at some temperature in the range 4.2–78 K. In fact, an orientational transition in this iron garnet was discovered in Ref. 9 and also independently in Refs. 9 and 10, the temperature obtained being 15 K. The published brief communications^{8–10} contain little information on the nature and characteristics of spin-reorientation phase transitions in $Dy_3 Fe_5 O_{12}$. Moreover, no studies have been made of spin reorientation in mixed dysprosium-yttrium iron garnets $Dy_3 Fe_5 O_{12}$. Therefore, we carried out a comprehensive experimental investigation of spin reorientation in these magnetic materials and developed a theoretical model which accounts satisfactorily for the certain characteristics of the observed phase transition.

SAMPLES AND EXPERIMENTAL METHOD

Single crystals of $Dy_x Y_{3-x} Fe_5 O_{12}$ garnets ($x = 0.2, 0.4, 1.0, 1.5, 2.0, 2.5,$ and 3.0) were grown by spontaneous crystallization from a molten solution.¹¹ Polycrystalline samples were synthesized by the conventional ceramic technology. The magnetization was measured in the temperature range 4.2–300 K by a vibration magnetometer. The torque curves were determined in the same temperature range using a torsional anisometer. The magnetic susceptibility was measured at temperatures 1.8–300 K in weak alternating fields of frequencies 220 Hz and 20 KHz. Moreover, the ^{57}Fe NMR spectra were recorded for the Fe^{3+} ions at octahedral positions in samples with $x = 0.2$ and 0.4 at temperatures in the range 5–30 K. The two-pulse Hahn spin echo method was used and the signals originated from nuclei in the interior of domains. Dysprosium iron garnet $De_3 Fe_5 O_{12}$ was used to study distortions of the crystal structure by the x-ray diffraction method at temperatures 5–300 K.

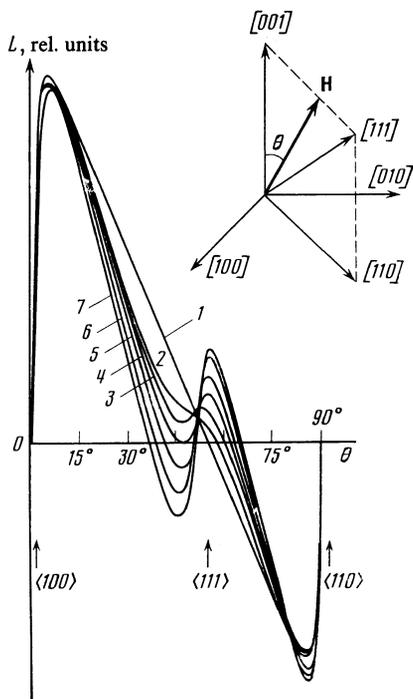


FIG. 1. Angular dependences of the torque obtained for a single-crystal disk of $\text{Dy}_{1.5}\text{Y}_{1.5}\text{Fe}_5\text{O}_{12}$ recorded in a $\{110\}$ plane in a field $H = 10$ kOe. Temperature of the sample (K): 1) 29.3; 2) 16.6; 3) 15.4; 4) 14.0; 5) 12.4; 6) 9.6; 7) 4.2.

EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows, by way of example, the dependences of the torque L for $\text{Dy}_{1.5}\text{Y}_{1.5}\text{Fe}_5\text{O}_{12}$ garnets on the angle of orientation θ of the magnetic field in a $\{110\}$ plane. [The $L(\theta)$ curves for the remaining compositions were similar.] It is clear from Fig. 1 that the conditions for an energy minimum

$$L(\theta) = 0, \quad \partial L / \partial \theta < 0 \quad (1)$$

are satisfied at high temperatures (curve 1, $T = 29.3$ K) for the $\langle 111 \rangle$ direction ($\theta = 55^\circ$), which is the easy magnetization axis. However, at low temperatures (for example at 4.2 K, curve 7) the $\langle 111 \rangle$ direction and other principal directions $\langle 100 \rangle$ and $\langle 110 \rangle$ are no longer easy magnetization axes and the conditions of Eq. (1) are satisfied for other directions characterized by $\theta_1 < 55^\circ$ and $\theta_2 > 55^\circ$. Therefore, it follows from measurements of the torque that in the case of $\text{Dy}_{x-x}\text{Y}_{3-x}\text{Fe}_5\text{O}_{12}$ garnets at low temperatures the easy magnetization axis deviates from the $\langle 111 \rangle$ direction, i.e., a spin-reorientation phase transition takes place from $\langle 111 \rangle$ high-symmetry phases to $\langle uuv \rangle$ canted phases. Measurements of the torques in a $\{110\}$ plane cannot be used alone to determine unambiguously which of the two angles θ_1 and θ_2 corresponds to the easy magnetization axis, unless we invoke other experimental data or model representations. It should be also pointed out that the dependences $L(\theta)$ in Fig. 1 recorded in a field $H_0 = 10$ kOe correspond to a polydomain unsaturated state of a sample (with the exception of the regions in the vicinities of the angle θ_1 and θ_2). This is demonstrated by a strong dependence of the absolute value of the torque on H_0 and by the finite value of the moment along the $\langle 111 \rangle$ axis at low temperatures. Saturation of a sample of

$\text{Dy}_3\text{Fe}_5\text{O}_{12}$ was not reached in an earlier study⁸ even in a field of 200 kOe.¹⁾

The strong magnetic anisotropy of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ is used in Ref. 8 to account for the dependences $L(\theta)$ on the basis of a model in which the magnetization is "pinned" strongly along the easy magnetization axis. In the two alternative cases under discussion ($\langle uuv \rangle$ phases with $\theta_1 < 55^\circ$ and $\theta_2 > 55^\circ$) the three easy magnetization axes form an "umbrella" around each of the eight $\langle 111 \rangle$ directions, but in the former case the easy magnetization axes deviate from $\langle 111 \rangle$ to the nearest $\langle 100 \rangle$ direction, whereas in the latter case they deviate toward $\langle 110 \rangle$. If an allowance is made for the domains most favorable from the energy point of view, the positions of the zeros in the dependence $L(\theta)$ in the former case should satisfy the relationship

$$\text{tg}(55^\circ - \theta_1) = 2 \text{tg}(\theta_2 - 55^\circ), \quad (2)$$

where in the latter case they should satisfy the relationship

$$\text{tg}(55^\circ - \theta_1) = 1/2 \text{tg}(\theta_2 - 55^\circ). \quad (3)$$

The experimental data of Ref. 8 obey Eq. (2) but are in conflict with Eq. (3), so that the easy magnetization axis of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ was selected to be the direction characterized by the angle θ_1 (Ref. 8).

Our measurements of the magnetization of dysprosium iron garnet at 4.2 K made it possible to determine the directions of the easy magnetization axes without invoking any model representations. Figure 2 shows the magnetization curves determined for the principal directions and also along the directions corresponding to the angles θ_1 and θ_2 . We can see that the magnetization in a field oriented in a $\{110\}$ plane at an angle θ_1 relative to $\langle 100 \rangle$ is the strongest. Hence, it follows that this direction is the easy magnetization axis. The same conclusion follows from an analysis of the relationships between the values of the magnetization M_s along the principal crystallographic directions $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ in the limit $H \rightarrow 0$. It is clear from the data in Fig. 2 that $M_{s\langle 100 \rangle} / M_{s\langle 111 \rangle} = 0.82$ and $M_{s\langle 110 \rangle} / M_{s\langle 111 \rangle} = 0.91$. Simi-

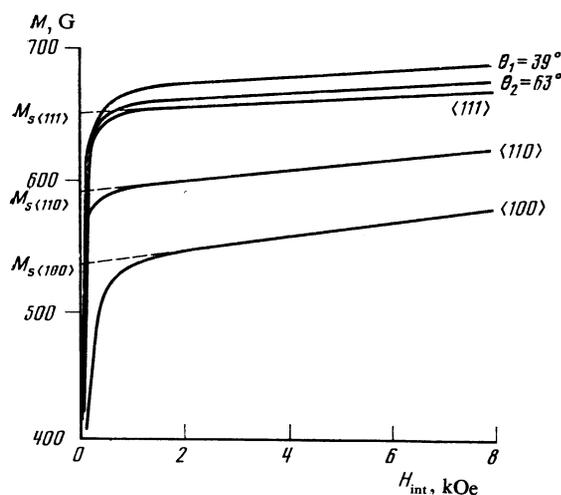


FIG. 2. Magnetization curves of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ at $T = 4.2$ K. The angles θ_1 and θ_2 were measured from a direction of the $\langle 100 \rangle$ type in a $\{110\}$ plane. The sample was spherical.

lar experimental values are obtained in Ref. 7 by extrapolation from much higher fields: $M_{s\langle 100 \rangle} / M_{s\langle 111 \rangle} = 0.79$ and $M_{s\langle 110 \rangle} / M_{s\langle 111 \rangle} = 0.88$. The corresponding calculated values for the easy magnetization axis along the direction governed by the angle $\theta_1 = 39^\circ$ are 0.81 and 0.90, whereas for the direction corresponding to the angle $\theta_2 = 63^\circ$ they are 0.64 and 0.90. (In the case of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ these angles were found by the torque method.) Consequently, the experimental magnetization ratios can be explained satisfactorily only for the easy magnetization axis governed by the angle θ_1 .

The angular dependences of the torque (Fig. 1) allow us to determine the nature of the $\langle 111 \rangle \leftrightarrow \langle uuv \rangle$ spin-reorientation phase transition. When temperature is increased, the value of the angle θ_1 for the $\langle uuv \rangle$ phases rises (curves 7–5) and at a certain temperature $T_i = 14$ K the angle reaches its critical value $\theta_{1cr} = 47^\circ$ (curve 4), which is smaller than $\theta = 55^\circ$ for the $\langle 111 \rangle$ phases. Such an abrupt change in the orientation of the easy magnetization axis corresponds to a first-order phase transition. Figure 3 shows the temperature dependences of θ_1 and θ_2 for the compositions with $x = 0.2, 1.5, 2,$ and 3 , obtained from the torque measurements. It is found that, in spite of the wide variation of the dysprosium concentration, the spin-reorientation transition temperature T_i , the critical angle θ_{1cr} and the limiting angle $\theta_1(0)$ (corresponding to $T \rightarrow 0$ K) change only slightly.²⁾

The spin-reorientation phase transition temperatures were determined by us also from measurements of the magnetic susceptibility in weak fields: they correspond to the maximum in the temperature dependence of the susceptibility.³ Measurements carried out on single-crystal and polycrystalline samples showed that the transition temperature is almost independent of the dysprosium concentration: it can be regarded as equal to $T_i = 13.5 \pm 1.5$ K for all the compositions with $x = 0.2-3$. The relatively large error in the determination of T_i is due to the considerable width of the susceptibility maximum. This error includes also the variations of the transition temperatures determined by different methods. More direct data on the nature of the spin-

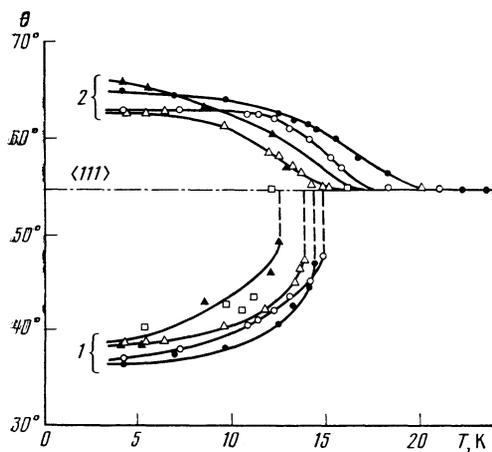


FIG. 3. Temperature dependences of the angles θ_1 (curves denoted by 1) and θ_2 (curves denoted by 2) obtained for $\text{Dy}_x\text{Y}_{3-x}\text{Fe}_5\text{O}_{12}$ by the torque method. Dysprosium concentration x : \blacktriangle 0.2; \bullet 1.5; \circ 2; \triangle 3. Here, the symbol \square represents the results obtained by the ^{57}Fe NMR method for a sample with $x = 0.2$.

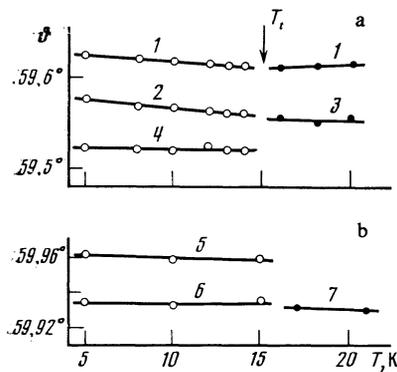


FIG. 4. Splitting of the cubic x-ray reflections (888) (a) and (1200) (b) in the $\langle 111 \rangle$ phases (black dots) and $\langle uuv \rangle$ phases (open circles): 1) (888); 2) (888/888); 3) (888/888/888); 4) (888); 5) (0012); 6) (1200/0120); 7) (1200/0120/0012). Here, ϑ is the angle of the position of the x-ray reflection.

reorientation phase transition in dysprosium yttrium iron garnets were obtained by us from x-ray and NMR measurements.

It is known that in the case of rare-earth iron garnets a transition to a magnetically ordered state is accompanied by distortions of the cubic crystal structure as a result of the magnetoelastic interaction. The nature of these distortions is governed by the direction of the easy magnetization axis. For example, in the case of the $\langle 111 \rangle$ phases the distortions should be rhombohedral, whereas in the case of the $\langle uuv \rangle$ phases they should be monoclinic. Figure 4 shows the positions of the x-ray reflections obtained for dysprosium iron garnet at various temperatures near the phase transition point. We can see that at temperatures $T > 15$ K the cubic x-ray reflection of the (888) type is split into two: (888) and (888/888/888), whereas the (1200) reflection is not split. This corresponds to rhombohedral distortions of the cubic crystal structure of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$. Below 15 K in the $\langle uuv \rangle$ phase the (888) reflection splits into three components: (888), (888/888), and (888), whereas the (1200) reflection splits into two: (0012) and (1200/0120), which is typical of monoclinic distortions of the crystal structure in the $\langle uuv \rangle$ phase. The splitting of the reflections is abrupt (it occurs in a temperature interval ≈ 1 K), indicating that the spin-reorientation transition to the $\langle uuv \rangle$ phase is a first-order phase transition.

An investigation of the ^{57}Fe NMR in single crystals near the phase transition temperature could be carried out only for the compositions with $x = 0.2$ and 0.4 , because a further increase in the dysprosium concentration broadened considerably the resonance lines and the intensities of the echo signal fell strongly because of a reduction in the NMR gain. A study was made of the behavior of the spectra of the octahedral Fe^{3+} ions located in the interior of the domains. It is known (see, for example, Ref. 2) that the anisotropic contributions to the NMR frequency and the spatial distribution of the local symmetry axes of the positions occupied by the Fe^{3+} ions in the garnet structure makes it possible to use the number of lines in the spectrum, their relative intensities, and relative positions to determine at the microscopic level the directions of the moments of the iron sublattices. For example, for a specific octahedral position the local sym-

metry axis is the threefold axis which has the same direction as one of the four axes of the $\langle 111 \rangle$ type. Therefore, the anisotropic components of the resonance frequency for the octahedral Fe^{3+} ions can be expressed in the form

$$f_a^{(k)} = -(3 \cos^2 \alpha_k - 1) F_a, \quad (4)$$

where α_k are the angles between the direction of the magnetization in a domain and the local symmetry axes of the octahedral positions ($k = 1, \dots, 4$) and F_a is the amplitude of the anisotropic component of the resonance frequency dependent on the anisotropic hyperfine and dipole interactions.² For an arbitrary rotation of the magnetization of the antiferromagnetically ordered iron sublattices we should observe four branches of the resonance frequencies with equal-intensity lines, but if the magnetization rotates in a $\{110\}$ plane, there is a partial degeneracy and the three branches are observed with the line intensity ratio 1:2:1 for $\theta < 55^\circ$ and 1:1:2 for $\theta > 55^\circ$. If $\theta = 55^\circ$ ($M_{\text{Fe}} \parallel \langle 111 \rangle$) the spectrum consists solely of two lines with intensities in the ratio 1:3.

Figure 5a shows the temperature dependence of the ^{57}Fe NMR frequencies of the octahedral Fe^{3+} ions in $\text{Dy}_{0.2}\text{Y}_{2.8}\text{Fe}_5\text{O}_{12}$. Above ≈ 12 K the spectrum consists of two lines with the intensity ratio 1:3, which confirms unambiguously the presence of the $\langle 111 \rangle$ phases. At low temperatures the spectrum consists of three lines with the intensity ratio 1:2:1, which corresponds to the $\langle uuv \rangle$ canted phases with $\theta_1 < 55^\circ$. We can thus see that the NMR data allow us to identify the $\langle uuv \rangle$ canted phases in which the magnetization is rotated from $\langle 111 \rangle$ toward $\langle 100 \rangle$ in $\{110\}$ -type planes, which confirms the results of our magnetic measurements. The transition between the spectra of the $\langle 111 \rangle$ and $\langle uuv \rangle$ phases is abrupt (it occurs within an interval of 0.7 K), as expected for a first-order phase transition.

An analysis of the NMR spectra in accordance with Eq. (4) gives the following results. The temperature dependence of the angle θ deduced from the NMR measurements is in satisfactory agreement with the dependence obtained by the torque method (Fig. 3). The temperature dependences of the isotropic and anisotropic components of the resonance frequency (Fig. 5) exhibit discontinuities at $T \approx 12$ K, providing additional evidence of the occurrence of a first-order spin-reorientation phase transition. These abrupt changes are clearly due to an abrupt change in the nature of the crystallographic distortions.

THEORETICAL DESCRIPTION OF A SPIN-REORIENTATION PHASE TRANSITION IN $\text{Dy}_x\text{Y}_{3-x}\text{Fe}_5\text{O}_{12}$ GARNETS

All our experimental data indicate that dysprosium yttrium iron garnets exhibit a low-temperature first-order spin-reorientation phase transition from high-symmetry phases of the $\langle 111 \rangle$ type to canted phases of the $\langle uuv \rangle$ type in which the magnetization is distributed in $\{110\}$ planes between the $\langle 100 \rangle$ and $\langle 111 \rangle$ directions (24 degenerate phases). Since in the temperature range in which this transition takes place the anisotropy of the iron subsystem is much less than the anisotropy of the dysprosium subsystem (even in the case of a strongly diluted iron garnet with $x = 0.2$ we have $E_a^{\text{Dy}} \sim 10^2 E_a^{\text{Fe}}$), it follows that this transition involves a

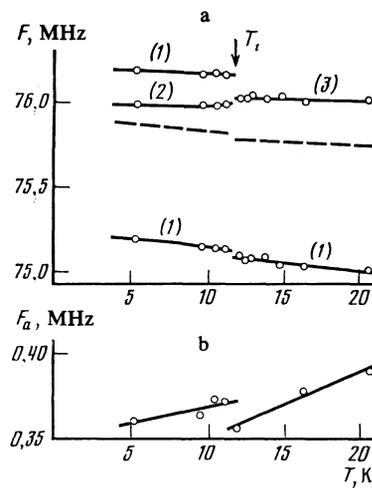


FIG. 5. Temperature dependences of the ^{57}Fe NMR frequencies (a) of the octahedral Fe^{3+} ions in $\text{Dy}_{0.2}\text{Y}_{2.8}\text{Fe}_5\text{O}_{12}$ (the line intensities are given in parentheses and the dashed line is the isotropic component of the NMR signal) and of the anisotropic component of the NMR frequency (b).

change in the state of the rare-earth subsystem because of a change in temperature.

We have shown earlier^{4,12,13} that the magnetic properties of samarium and holmium yttrium iron garnets can be described satisfactorily if we allow for the isotropic exchange interaction between the rare-earth and iron ions and for the crystal field acting on the rare-earth ions. It is assumed in our earlier treatments that the splitting of the levels in the crystal field is such that the ground-state doublet (quasidoublet) of the rare-earth ions is separated considerably from higher energy levels, so that the influence of the latter can be allowed for by regarding them as a perturbation (in the case of samarium iron garnets such a splitting scheme for the Sm^{3+} ions was recently confirmed by a microscopic calculation¹⁴).

The magnetic properties of iron garnets considered on the basis of this model and particularly the occurrence of spin-reorientation phase transitions of certain type depend on the ratio of the components of the g tensor of the ground-state doublet (quasidoublet) of the rare-earth ions and generally on the influence of the excited levels. For example, the properties of holmium yttrium iron garnets are described well by this model if only the diagonal g_z component of the g tensor of the ground-state quasidoublet of the Ho^{3+} ions remains finite.^{12,13} A sequence of spin-reorientation phase transitions in samarium iron garnets can be accounted for satisfactorily if $g_x \gg g_y$ and it is necessary to allow for the influence of excited levels on the shift of the center of gravity of the ground-state doublet.⁴

A similar model was used by us to describe spontaneous spin-reorientation phase transitions in dysprosium yttrium iron garnets. It was found that the $\langle 111 \rangle \leftrightarrow \langle uuv \rangle$ transition observed in these ferrimagnets can occur only if the ground-state doublet of the Dy^{3+} ions has at least two nonzero diagonal components of the g tensor.

In the general case of this model the thermodynamic potential of iron garnets (calculated per one rare-earth ion) can be represented in the form

$$F = -\frac{T}{6} \sum_{r=1}^6 \ln \left(2 \operatorname{ch} \frac{\Delta_r}{T} \right) + K (\gamma_x^2 \gamma_y^2 + \gamma_y^2 \gamma_z^2 + \gamma_z^2 \gamma_x^2), \quad (5)$$

where

$$\Delta_r = \frac{1}{2} \lambda M_{\text{Fe}} \left[\sum_{\alpha} g_{\alpha}^2 (\gamma \mathbf{e}_{\alpha}^r)^2 \right]^{1/2} \quad (6)$$

represents splitting of the ground-state doublet of the rare-earth ion at the r th site because of the exchange interaction of this ion with the iron ions; λM_{Fe} is the molecular field exerted on the rare-earth ion by the rare-earth sublattice; $\gamma = \mathbf{M}_{\text{Fe}} / M_{\text{Fe}}$ is a unit vector which defines the direction of the magnetization of the iron sublattice; \mathbf{e}_{α}^r are unit vectors along the local symmetry axes of the r th site ($\alpha = x, y, z$; $r = 1, \dots, 6$); g_{α} are the components of the g tensor. Here and later the temperature is in energy units. In writing down Eqs. (5) and (6) an allowance is made for the fact that the rare-earth ions in the garnet structure occupy six inequivalent dodecahedral sites (and the symmetry of their environment is described by the point group D_2). The orientations of the local symmetry axes can be found in Table II of Ref. 15.

In Eq. (5) the first term describes the contribution to the thermodynamic potential made by the splitting of the ground-state doublet of the rare-earth ions by the molecular field. The second term represents the anisotropic component of the sum of the displacements of the centers of gravity of the doublets of the rare-earth ions in different inequivalent positions because of the influence of the excited levels.³ At low temperatures ($T < W$, where W is the separation between the ground-state doublet and the excited levels) the quantity K can be regarded as temperature-independent.

In terms of dimensionless variables

$$\Phi = \frac{2F}{g\lambda M_{\text{Fe}}}, \quad k = \frac{2K}{g\lambda M_{\text{Fe}}}, \quad \tau = \frac{2T}{g\lambda M_{\text{Fe}}},$$

$$\sigma_{\alpha} = \frac{g_{\alpha}^2}{g^2}, \quad g = \left(\sum_{\alpha} g_{\alpha}^2 \right)^{1/2}$$

the thermodynamic potential of Eq. (5) becomes

$$\Phi = -\frac{\tau}{6} \sum_r \ln \left\{ 2 \operatorname{ch} \left(\frac{1}{\tau} \left[\sum_{\alpha} \sigma_{\alpha} (\gamma \mathbf{e}_{\alpha}^r)^2 \right]^{1/2} \right) \right\} + k (\gamma_x^2 \gamma_y^2 + \gamma_y^2 \gamma_z^2 + \gamma_z^2 \gamma_x^2). \quad (7)$$

The equilibrium orientations of the vector γ can be found by minimization of Eq. (7). A numerical analysis on a computer made it possible to determine the orientational phase diagram which is common to all rare-earth iron garnets considered on the basis of the assumptions given above. This diagram is shown in Fig. 6 in terms of dimensionless coordinates (k, σ_x, σ_y) for the case when $T = 0$. We can see that for $k = 0$ the $\langle uuv \rangle$ phases do not appear for any values of the components of the g tensor. However, there is a range of values of the components of the g tensor corresponding to $k < 0$ in which the $\langle uuv \rangle$ phases are absolutely stable at 0 K. This range is shaded in Fig. 6.

A calculation demonstrates that in this case an increase in temperature should result in a first-order spin-reorienta-

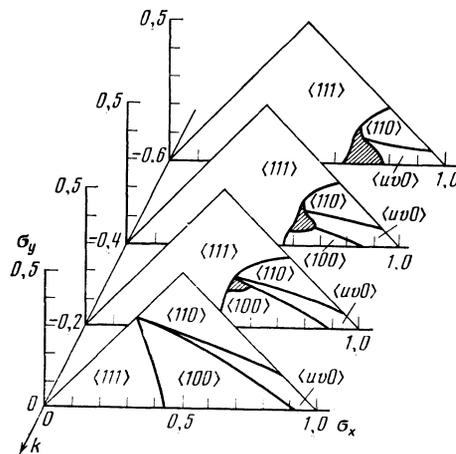


FIG. 6. Theoretical orientational phase diagram of rare-earth iron garnets at $T = 0$. The shaded region corresponds to the $\langle uuv \rangle$ phases.

tion phase transition $\langle uuv \rangle \leftrightarrow \langle 111 \rangle$. This transition is physically due to the fact that an increase in temperature reduces the first term in the thermodynamic potential (7) due to the ground-state doublet, whereas the second term corresponding to $k < 0$ stabilizes the $\langle 111 \rangle$ phases.

The orientation angle θ of the easy magnetization axes of the $\langle uuv \rangle$ phases and the transition temperature T_i depend in this model on the values of the g factors and of K . For example, if we assume that $g = 16.1$, $\sigma_x = 0.62$, $\sigma_y = 0.0915$, and $\sigma_z = 0.2885$, which are values similar to those reported for dysprosium gallate garnet,¹⁶ and if we also assume that $\lambda M_{\text{Fe}} = 158$ kOe (Ref. 7) and $k = -0.432$, we can then describe satisfactorily the temperature dependence of θ observed for dysprosium iron garnet.

In this model the properties of mixed iron garnets (such as the transition temperature T_i and the angle θ_1) should be independent of the concentration of the rare-earth ions (at least to such strong dilutions that the anisotropy of the iron sublattice begins to play a significant role), provided there is no change in the splitting of the energy levels of the rare-earth ion in the crystal field of the lattice. The experimental results indicate that such a situation does indeed occur in the system of dysprosium yttrium iron garnets. This is the qualitative difference between these garnets and samarium yttrium iron garnets in which spin-orientation phase transitions occur at temperatures that depend strongly on the composition because of a change in the splitting of the energy levels of the Sm^{3+} ion on replacement of samarium with yttrium.⁵

The difference between the behavior of the $\text{Dy}_x \text{Y}_{3-x} \text{Fe}_5 \text{O}_{12}$ and $\text{Sm}_x \text{Y}_{3-x} \text{Fe}_5 \text{O}_{12}$ systems is clearly due to the following circumstance. In the former case the crystal lattice constant a of the terminal compositions is almost the same: ($a_Y = 12.376$ Å, $a_{\text{Dy}} = 12.405$ Å). Therefore, variation of the dysprosium concentration does not alter the parameters of the dodecahedral environment of the Dy^{3+} ions and, consequently, it does not alter the crystal fields acting on these ions. In the case of samarium yttrium iron garnets the lattice constant depends strongly on the composition ($a_{\text{Sm}} = 12.529$ Å). This alters the parameters of the dodeca-

hedra and, consequently, changes the crystal field acting on the Sm^{3+} ion.

We shall conclude by noting that the "umbrella" (helical) magnetic structure of $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ with the resultant magnetic moment of a crystal along $\langle 111 \rangle$, established in Ref. 17 by the neutron diffraction method, is incorrect and must be redetermined. The same conclusion was reached in Ref. 8.

We regard it as our pleasant duty to thank K. P. Belov and N. M. Kovtun for valuable discussions of the results.

¹Spin reorientation from the high-symmetry $\langle 111 \rangle$ phases to the canted $\langle uuv \rangle$ phases exhibited by dysprosium yttrium iron garnets can be described phenomenologically only if we allow for three magnetic anisotropy constants of a cubic magnetic material: K_1 , K_2 , and K_3 (Refs. 3 and 9). Estimates of these constants for $\text{Dy}_3\text{Fe}_5\text{O}_{12}$ at 4.2 K yield the following results: $|K_i| \sim 10^8 \text{ erg/cm}^3$; $K_1 < 0$; $K_2, K_3 > 0$.

²It should be pointed out that some features of the torque curves cannot be explained by the polydomain model of Ref. 8 with the magnetization pinned strongly along the easy magnetization axis or by assuming that the sample is homogeneously magnetized. For example, at $T > T_i$ the conditions $\theta_1 = \theta_2 = 55^\circ$ and $L(55^\circ) = 0$ should be obeyed, whereas the experimental results indicate that θ_2 still differs from 55° at temperatures exceeding T_i by 3–5 K (see Figs. 1 and 2 in the present paper and also Ref. 8); moreover, we have $L(55^\circ) \neq 0$. This may be due to the coercivity of the samples or due to the existence of a low-temperature uniaxial anisotropy originating from magnetostrictive distortions of the cubic crystal structure.

³This term describes also the anisotropy of the iron sublattice. However, as pointed out earlier, this anisotropy is negligible at low temperatures.

¹K. P. Belov, A. K. Zvezdin, R. Z. Levitin, A. S. Markosyan, B. V. Mill', A. A. Mukhin, and A. P. Perov, Zh. Eksp. Teor. Fiz. **68**, 1189 (1975) [Sov. Phys. JETP **41**, 590 (1975)].

²V. A. Borodin, V. D. Doroshev, V. A. Klochan, N. M. Kovtun, R. Z. Levitin, and A. S. Markosyan, Zh. Eksp. Teor. Fiz. **70**, 1363 (1976) [Sov. Phys. JETP **43**, 711 (1976)].

³K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, Orientatsionnye, perekhody v redkozemel'nykh magnetikakh (Orientational Transitions in Rare-Earth Magnetic Materials), Nauka, M., 1979.

⁴G. A. Babushkin, V. A. Borodin, V. D. Doroshev, A. K. Zvezdin, R. Z. Litvin, and A. I. Popov, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 28 (1982) [JETP Lett. **35**, 34 (1982)].

⁵V. A. Borodin, V. D. Doroshev, R. Z. Levitin, V. Nekvasil, V. N. Orlov, and T. N. Tarasenko, Zh. Eksp. Teor. Fiz. **86**, 2255 (1984) [Sov. Phys. JETP **59**, 1312 (1984)].

⁶R. F. Pearson, J. Appl. Phys. **33**, Suppl., 1236 (1962).

⁷A. E. Clark and E. Callen, J. Appl. Phys. **39**, 5972 (1968).

⁸G. Aubert and B. Michelutti, J. Magn. Magn. Mater. **31-34**, 811 (1983).

⁹V. A. Borodin, V. D. Doroshev, and T. N. Tarasenko, Fiz. Met. Metalloved. **56**, 220 (1983).

¹⁰V. A. Borodin, V. D. Doroshev, V. N. Orlov, and T. N. Tarasenko, Tezisy dokladov XVI Vsesoyuznoi konferentsii po fizike magnitnykh yavlenii (Abstracts of Papers presented at Sixteenth All-Union Conf. on Physics of Magnetic Phenomena), Tula, 1983, Part I, p. 185.

¹¹K. P. Belov, A. K. Gapeev, R. Z. Levitin, A. S. Markosyan, and Yu. F. Popov, Zh. Eksp. Teor. Fiz. **68**, 241 (1975) [Sov. Phys. JETP **41**, 117 (1975)].

¹²V. I. Silant'ev, A. I. Popov, R. Z. Levitin, and A. K. Zvezdin, Zh. Eksp. Teor. Fiz. **78**, 640 (1980) [Sov. Phys. JETP **51**, 323 (1980)].

¹³G. A. Babushkin, A. K. Zvezdin, R. Z. Levitin, A. I. Popov, and V. I. Silant'ev, Zh. Eksp. Teor. Fiz. **80**, 1952 (1981) [Sov. Phys. JETP **53**, 1015 (1981)].

¹⁴M. Guillot, A. Marchand, V. Nekvasil, and F. Tcheou, J. Phys. C. (in press).

¹⁵A. K. Zvezdin, R. Z. Levitin, A. I. Popov, and V. I. Silant'ev, Zh. Eksp. Teor. Fiz. **80**, 1504 (1981) [Sov. Phys. JETP **53**, 771 (1981)].

¹⁶J. Filippi, F. Tcheou, and J. Rossat-Mignod, Solid State Commun. **33**, 827 (1980).

¹⁷F. Tcheou, E. F. Bertaut, and H. Fuess, Solid State Commun. **8**, 1751 (1970).

Translated by A. Tybulewicz