Diagram of magnetic orientational phase transitions in gadolinium iron garnet single crystals with internal stresses

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(Submitted 22 July 1983)
Zh. Eksp. Teor. Fiz. **86**, 1084–1091 (March 1984)

The method of polarization optics is used in a study of the magnetic orientational phase transitions in gadolinium iron garnet single crystals in an external magnetic field imposed along the $\langle 111 \rangle$ axis. The phase diagrams measured at different points in the samples exhibit previously unreported lines of phase transitions between canted phases. All the phase-transition lines are well described by a theory which incorporates the internal stresses in the crystals. For all the transitions which occur, the calculated phase-boundary configurations in the stress fields of the dislocations actually present in the samples practically coincide with those observed in experiment.

There has been considerable research interest of late in magnetic orientational (spin-flip) transitions—a particular class of order-order magnetic phase transitions due to the rotation of the spontaneous magnetization of the crystal upon a change in the temperature T or the external magnetic field H. Transitions of this type are observed, in particular, in rare earth iron garnets having a magnetic compensation point.[1]

Because thin wafers of these materials are transparent to visible light, one can use the method of polarization optics to study simultaneously the distribution of internal stresses (by the photoelastic effect) and the magnetic domain structure (by the Faraday and Cotton-Mouton effects).[2] This circumstance affords a unique possibility for direct experimental study of the influence of dislocations and other elementary lattice defects on the rotation of the magnetic moments, nucleation of domains of different phases, and motion of phase boundaries during spin-flip transitions. Such studies are of considerable interest in one of the current problems[3] of theoretical physics—the creation of a phase-transition theory which incorporates the real structure of crystals. In addition, this technique can elucidate the nature of the onset of inhomogeneous magnetic structure during the spin flip in a wide class of ordered magnets.

The method of polarization optics has already been used to make detailed studies of the existence regions of the magnetic phases in single crystals of gadolinium iron garnet $Gd_3Fe_5O_{12}$ and to investigate the temperature dependence of the direction of the net magnetization at various orientations of the external magnetic field with respect to the crystallographic axes.[4,5] The theoretical model of a many-sublattice ferromagnet with ideal cubic anisotropy,[1] although successfully describing the diversity of the phases which occur and the general behavior of the transitions between them, has been unable to explain many of the experimentally discovered features of these transitions—in particular, the shape of all the lines bounding the existence regions of the various phases on the H-T plane[4] and the sequence of transitions between equivalent canted phases.[5] It has also left unexplained the reasons for the coexistence of several magnetic phases at low magnetic fields over a wide temperature interval around the magnetic compensation point.[1,4,5] It is natural to suppose that these disagreements between theory and experiment are due to the real structure of the samples, i.e., to the presence of defects of the crystal structure. The problem of studying the influence of lattice defects on magnetic orientational transitions has thus moved to the forefront.

The observations of anomalies in the occurrence of the spin-flip phase transition near the leading sources of internal stresses in crystals—isolated dislocations—in gadolinium iron garnet wafers was first reported in Ref. 6. A detailed study of the orientational transitions in the interior of a real crystal was reported in Ref. 7. It was shown both theoretically and experimentally that the form of the H-T diagrams for first-order phase transitions is governed by the magnitude of the internal stresses, while the shape of the phase boundaries is governed by the distribution of the internal stresses over the sample. This distribution has a characteristic shape near dislocations. In the present article we consider transitions occurring at an orientation of the external field such that the number of phases realized is larger than in Ref. 7, and phase lines which were not predicted by the theory of Ref. 1 appear.

EXPERIMENTAL TECHNIQUES

The experiment was basically done by the techniques set forth in Ref. 7. Single crystals of gadolinium iron garnet ($Gd_3Fe_5O_{12}$) were cut parallel to the (110) plane into wafers whose thickness after mechanical and chemical polishing was 30–50 μm. These samples were placed in a cryostat on a copper cold finger through which heated nitrogen vapor was passed. The temperature of the cryostat was stabilized by an ESNT-1 unit feeding a Nichrome coil placed in a cooling gas jet. The unit was controlled by the out-of-balance signal of a dc bridge having a copper thermistor, placed alongside the sample, in one of its arms. The temperature was measured with the aid of a copper-constantan thermocouple to an accuracy of 0.02 K. The cryostat was mounted between the poles of an electromagnet which produced a static field of up to 22 kOe. The field was measured with a Hall transducer.

The spin-flip transitions were studied by a polarization-

 0038-5646/84/030633-05$04.00

optics technique. The orientation of the magnetization vectors of each of the phases was determined from analysis of the Faraday and Cotton-Mouton magnetooptic effects. The dislocations were revealed and identified from the birefringences rosettes[2] observed around them in crossed polarizers when the wafers were magnetized to saturation in the plane.

The instant at which the phase transition occurred at a chosen point in the sample was fixed by observing the passage of the corresponding phase boundary through it. By holding the boundary at a given point through coordinated changes in the magnetic field and temperature and feeding the signals from the Hall transducer and thermocouple (through a V2-11 unit) to an x-y recorder, we could automatically trace out the phase-transition line on the H-T plane.

In the present experiment the samples were oriented so that the magnetic field was directed along the [111] axis, which lay in the plane of the wafers. In this arrangement a greater number of phases was realized during the spin flip than in the earlier experiment[7] with the magnetization reversal along the other easy axis, $[11\bar{1}]$, which was a preferred direction in the investigated series of samples because of a small additional, growth-induced anisotropy.

The direction of the 180-degree boundaries separating domains with magnetization along $[11\bar{1}]$ (these were always present when the temperature was far from the compensation point at $H = 0$) and the boundaries of the growth sectors, which lie strictly along [111], [001], and [110], were convenient references for orienting the crystals.

EXPERIMENTAL RESULTS

When an external magnetic field was imposed along the easy axis [111], six phases were observed in a $(1\bar{1}0)$ $Gd_3Fe_5O_{12}$ single-crystal wafer as the field and temperature were changed. Two of the phases were collinear, with magnetization along [111] (high-temperature phase A and low-temperature phase B), and four were canted, with magnetizations close to the axes $[1\bar{1}1]$ and $[11\bar{1}]$ (high-temperature phases C_1 and C_2 and low-temperature phases D_1 and D_2, respectively). The high-temperature phases differ from the low-temperature phases in that the magnetization of the gadolinium sublattice in the former is directed counter to the external field. During the experiment the plane of the sample was perpendicular to the optic axis, and the polarizers were slightly crossed. This caused the color of the phases A and B to be slightly different from the color of C_2 and D_2 on account of the magnetic birefringence. Within each pair these (Cotton) phases could easily be distinguished from each other by the nature of the motion of their phase boundaries with the (Faraday) phases C_1 and D_1. Phases C_1 and D_1 were considerably brighter than the Cotton phases on account of the Faraday effect, and had different brightness and color when viewed in crossed Nicols.

As in Ref. 7, the transitions between phases corresponding to different easy axes occurred through a gradual displacement of the phase boundaries by an amount which, at a fixed change in temperature, grew with increasing magnetic field, while the transitions between phases corresponding to the same easy axis occurred through the motion of a 180-degree compensation boundary. The pattern of the low-temperature phase transitions is practically identical to that of the corresponding high-temperature transitions if T is replaced by $2T_c-T$ ($T_c \approx 283$ K is the magnetic compensation temperature of $Gd_3Fe_5O_{12}$).

To a large degree the kinetics of the spin-flip transition was governed by the presence of defects, primarily dislocations, in the crystals. Thus in the transition from A to C_1 the C_1 phase was nucleated at dislocations in the form of a bright, bean-shaped microscopic region, expanded with practically no change in shape, and then came to occupy the main volume of the crystal, compressing phase A into a microscopic region (the dark region in Fig. 1a) which was sym-

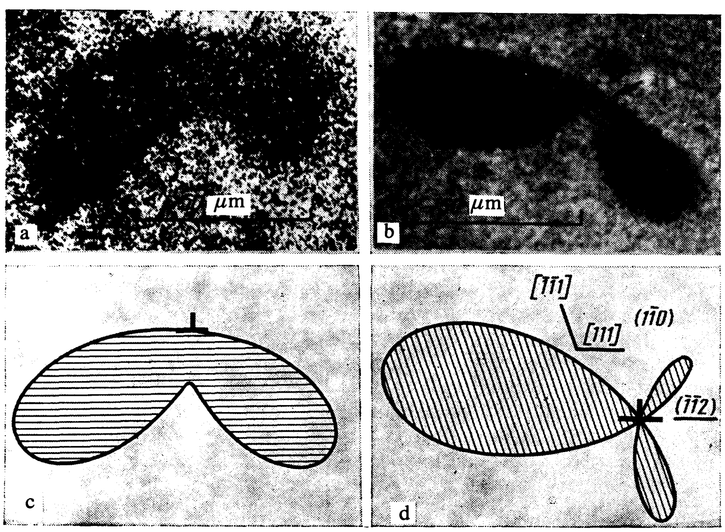

FIG. 1. Experimentally observed (a,b) and theoretically calculated (c,d) shape of the phase boundary near a dislocation with slip plane $(\bar{1}\bar{1}2)$ and Burgers vector $\mathbf{b}\|[111]$: a) the transition $B \leftrightarrow D_1$ at $H = 3.1$ kOe, $T = 281.8$ K; b) the transition $D_1 \leftrightarrow C_2$ at $H = 1.2$ kOe, $T = 282.9$ K (the axis of the dislocation is indicated by an arrow in a and b); c) the shape of the boundary between phases with magnetization along [111] (hatched along the magnetization direction) and $[1\bar{1}1]$; d) the same as in c but between $[11\bar{1}]$ (hatched along the magnetizaiton direction) and $[1\bar{1}1]$.

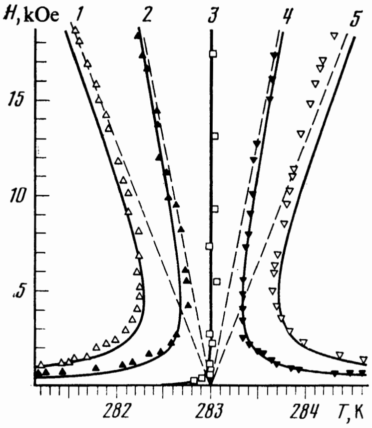

FIG. 2. Diagram of the magnetic orientational phase transitions at a single point in a $Gd_3F_5O_{12}$ crystal: 1) $B \leftrightarrow D_1$; 2) $D_1 \leftrightarrow D_2$; 3) $D_1 \leftrightarrow C_2$; 4) $C_2 \leftrightarrow C_1$; 5) $C_1 \leftrightarrow A$.

metric about the dislocation axis with the initial nucleus of phase C_1 (this is all analogous to the transition from the collinear to the canted phase as described in Ref. 7 except for the shape of the microscopic region which formed at the dislocation).

As H and T were changed, the boundary between canted phases belonging to different easy axes migrated, forming on the dislocation a rosette (Fig. 1b) which was identical to the rosette observed in Ref. 7. During these phase transitions there was a transformation, analogous to that described above, from the bright rosette of phase C_1 (or D_1) to a rosette of phase C_2 (or D_2) of the corresponding color, in a symmetric location with respect to the dislocation axis.

Let us consider the H-T phase diagram obtained at one of the points in the crystal under study (Fig. 2). As in Ref. 7, when the temperature was varied near the compensation temperature $T_c \approx 283.0$ K, phase transitions always occurred between collinear and canted phases ($A \leftrightarrow C_1$, curve 5; $B \leftrightarrow D_1$, curve 1). The corresponding experimental points in fields $H \gtrsim 10$ kOe asymptotically approach straight lines which converge at the point $H = 0$, $T = T_c$ with a slope $|dH/dT| = 13$ kOe/K, while for $H \to 0$ the points begin to deviate from these limits by a hyperbolic law. In the chosen experimental configuration, these lines of phase transitions in the investigated region of the crystal always diverged, and the width of the "throat" between them was characterized by the value of the induced anisotropy inhibiting the orientation of the magnetization along the [111] axis.

The pattern of phase transitions in the region enclosed between these curves was significantly more complex than the pattern observed in Ref. 7. This complexity was manifested, in particular, in a different succession of canted phases for the same initial state if the spin flip was brought on at different values of the static field on account of a change in temperature. For example, as the temperature was increased from $T < T_c$ at $H \approx 3$ kOe the phase D_1 which appeared after phase B at the point under study gave way to a C_2 phase corresponding to another easy axis. Thus the actual succession of phases was that predicted by the theory for an ideal cubic crystal ($B \to D_1 \to C_2$), but with a change in the plane of rotation of the sublattice magnetizations from $(10\bar{1})$ to $(1\bar{1}0)$.[1] After this, however, the sequence of phases proposed in Ref. 1 was violated: instead of $C_2 \to A$ the transition which occurred was $C_2 \to C_1 \to A$. When the temperature was changed in the opposite direction the phase sequence was different: $A \to C_1 \to C_2 \to D_2 \to D_1 \to B$, in agreement with the experiment of Ref. 5. In other ranges of H the transition between canted phases could follow different patterns. The patterns were also different for different samples. Furthermore, the succession of phases depended strongly on the magnetization-switching history of the sample and the manner in which the temperature was changed over time. These features are due to structural defects in the crystals but were not investigated in detail in the present study, which was limited to the shape of the corresponding lines of phase transitions.

If a phase transition occurred at a chosen point in the sample (if the corresponding phase boundary passed through this point), then, by following along the phase-transition line by means of suitable changes in the field and temperature, we could track the transition throughout the field range $0 < H$ 20 kOe regardless of whether or not this transition was observed upon a change in temperature at the given value of the static field. When T and H deviated from the values corresponding to the phase transition and the phase boundary moved away from the chosen point, the phase equilibrium could easily be restored by changing one of these parameters; here, to within the accuracy of the measurements, hysteresis effects were not observed.

Of the previously[7] unobserved transitions between canted phases, only the transitions $D_1 \leftrightarrow D_2$, $D_1 \leftrightarrow C_2$, and $C_2 \leftrightarrow C_1$ are shown in Fig. 2 (curves 2, 3, and 4). The points corresponding to the transition $D_2 \leftrightarrow C_1$ and to the previously studied transition $D_2 \leftrightarrow C_2$, which is due to the motion of the compensation boundary,[7] are not shown, since for $H \gtrsim 1$ kOe they lie on the $T = T_c$ axis and coincide with the points of the transition $D_1 \leftrightarrow C_2$. The lines of transitions between canted phases are analogous to the lines (discussed above) of transitions between the collinear and canted phases, but the slope of the straight lines which they asymptotically approach at $H \gtrsim 5$ kOe (the dashed lines in Fig. 2) is twice as large for the transitions $D_1 \leftrightarrow D_2$ and $C_2 \leftrightarrow C_1$, viz. $|dH/dT| \approx 26$ kOe/K, while the points of the transition $D_1 \leftrightarrow C_2$ lie on the vertical line $T = T_c$. For $H \lesssim 1$ kOe the $D_1 \leftrightarrow C_2$ transition line deviates from the vertical on account of the induced anistropy at the investigated point in the crystal, while the $D_2 \leftrightarrow C_2$ transition line, by contrast, is insensitive to the induced anisotropy.[7]

ORIENTATIONAL PHASE TRANSITIONS IN INHOMOGENEOUSLY STRESSED RARE EARTH FERRITES WITH A MAGNETIC COMPENSATION POINT

The shape of the observed lines of phase transitions and the aforementioned features of the spin flip in the stress field of isolated dislocations can be described quantitatively in the framework of the phenomenological approach developed in Ref. 7.

In the region of weak magnetic fields the energy of a magnetic phase is written[7]

$$E=-\frac{1}{2}(\chi_{\perp}-\chi_{\parallel})H^2\sin^2\theta$$
$$-\frac{1}{2}\chi_{\parallel}H^2-\frac{T-T_K}{T}\chi_{\parallel}\lambda M_{Fe}H\cos\theta+\varepsilon. \quad (1)$$

Here θ is the angle between the magnetic field **H** and the easy axis along which the magnetic moments of the sublattices are oriented (the angle is measured from the magnetization direction of the iron sublattices), ε is the energy contribution from elastic stresses or from noncubic anistropy induced during the growth of the crystal, $\chi_{\perp}$ and $\chi_{\parallel}$ are the susceptibilities of the system of magnetic sublattices in the directions perpendicular to and parallel to the corresponding easy axis, λ is the effective homogeneous exchange constant between the rare earth and iron sublattices, and M_{Fe} is the net magnetization of the iron sublattices.

Under the conditions of our problem $\cos\theta$ equals $+1$ for phase A, -1 for phase B, approximately $+1/3$ for phases C_1 and C_2, and approximately $-1/3$ for D_1 and D_2. The values of θ are slightly different for phases C_1 and C_2 (θ_1 and θ_2, respectively, with $|\theta_1-\theta_2|\ll 1$) as a consequence of the slight deviation of the axes from the $\langle 111\rangle$ directions in the induced-anisotropy field and the inexact ($\approx 2°$) orientation of the crystal with respect to the direction of the external magnetic field. In phases D_1 and D_2 the angle is equal to $\pi-\theta_1$ and $\pi-\theta_2$, respectively. Then the condition that the energies of the phases are equal at a first-order phase transition yields the H dependence of the phase transition temperatures T_t.

For the transitions $A\leftrightarrow C_1$, $B\leftrightarrow D_1$

$$\pm\frac{T_t-T_c}{T_c}=\frac{2}{3}\frac{\chi_{\perp}-\chi_{\parallel}}{\chi_{\parallel}}\frac{H}{\lambda M_{Fe}}+\frac{3(\varepsilon_0-\varepsilon_1)}{2\chi_{\parallel}\lambda M_{Fe}H}; \quad (2)$$

here ε_0, ε_1, and ε_2 are respectively the increments to the energy of phases A and B, C_1 and D_1, and C_2 and D_2; the plus sign is for $A\leftrightarrow C_1$, the minus sign for $B\leftrightarrow D_1$.

For the transitions $C_1\leftrightarrow C_2$ and $D_1\leftrightarrow D_2$

$$\pm\frac{T_t-T_c}{T_c}=\frac{1}{3}\frac{\chi_{\perp}-\chi_{\parallel}}{\chi_{\parallel}}\frac{H}{\lambda M_{Fe}}-\frac{3(\varepsilon_1-\varepsilon_2)}{2^{3/2}\chi_{\parallel}\lambda M_{Fe}H(\theta_1-\theta_2)}; \quad (3)$$

the plus sign is for $C_1\leftrightarrow C_2$, the minus sign for $D_1\leftrightarrow D_2$. We note that near T_c there should be a phase with a value of θ closer to $\pi/2$—in our case $\theta_1<\theta_2<\pi/2$—and this dictates the experimentally observed anomalies in the succession of phases during the spin flip.

Finally, for the transitions $D_1\leftrightarrow C_2$ and $C_1\leftrightarrow D_2$ we have

$$\pm\frac{T_t-T_c}{T_c}=\frac{2^{1/2}(\theta_2-\theta_1)}{3}\frac{\chi_{\perp}-\chi_{\parallel}}{\chi_{\parallel}}\frac{H}{\lambda M_{Fe}}+\frac{3(\varepsilon_1-\varepsilon_2)}{2\chi_{\parallel}\lambda M_{Fe}H}. \quad (4)$$

The plus sign is for $C_1\leftrightarrow D_2$, the minus sign for $D_1\leftrightarrow C_2$.

As in Ref. 7, in accordance with the measured slope of the phase-transition curves in the linear region, we have taken

$$\chi_{\parallel}(\chi_{\perp}-\chi_{\parallel})^{-1}\lambda M_{Fe}/T_c=8.5\ \text{kOe/K}$$

and, choosing the values of the differences in the energy increments $\varepsilon_1-\varepsilon_0$ and $\varepsilon_1-\varepsilon_2$, we have constructed the theoretical lines of phase transitions at the point under study (the solid curves in Fig. 2); these lines give a good description of the experimental trends. By comparing curve 3 with curves 2 and 4, one can conclude that $\theta_1-\theta_2\approx 1°$.

By taking into account the connection between the induced anisotropy and the internal stresses, $\varepsilon=\bar{\varepsilon}-\Lambda_{ijmn}\sigma_{ij}\times\alpha_m\alpha_n$ ($\bar{\varepsilon}$ is the induced-anistropy energy from other sources, Λ_{ijmn} and σ_{ij} are the components of the magnetostriction and stress tensors, and α_m are the direction cosines of the magnetization) and also the form of the tensor σ_{ij} around the dislocations (for the dislocation in Fig. 1 the slip plane is $(\bar{1}\bar{1}2)$ and the Burgers vector is $\mathbf{b}=1/2a$ [111]; a is the lattice constant of $Gd_3Fe_5O_{12}$) in the elastically isotropic approximation, we calculated the shape of the microscopic phase regions arising at the dislocations for a transition between sublattice magnetization directions along [111] and $[1\bar{1}1]$ (Fig. 1c) and between $[11\bar{1}]$ and $[1\bar{1}1]$ (Fig. 1d) in precisely the same way as was done in Ref. 7.

The results of these calculations are in good agreement with the experimentally observed domain configurations for the different phases at the dislocation. The size of the microscopic phase region realized at a dislocation under specified conditions is governed by the difference in the value of ε in the neighboring phases; this energy difference is calculated as a function of T and H using formulas (2)–(4), depending on the type of transition.

DISCUSSION OF RESULTS

We have set that the experimentally measured lines of phase transitions agree well with the theoretical lines calculated with allowance for the induced anisotropy. In addition, there is practically complete agreement between the observed and calculated domain shapes for the different phases in the internal stress field created by isolated dislocations. This indicates that the induced magnetic anisotropy, especially the magnetoelastic anisotropy due to structural defects, plays the governing role in shaping the anomalies of the spin-flip processes in a real crystal in weak magnetic fields. Furthermore, recognizing that the domain boundaries between the corresponding phases at fixed values of T and H are described by lines of constant ε, one can measure the induced anisotropy and study the inhomogenity of its distribution in crystals by investigating the topology of the domain boundaries at the spin-flip phase transitions.

Generally speaking, it must be remembered that the calculations reported above do not take into account the contributions to the energy from the domain-boundary surface tensions, the magnetic fringing fields, and the inhomogeneous intrinsic magnetostrictive strains, which can exert an important infuence on an orientational phase transition. Let us estimate the size of these effects for the case of a domain rosette around a dislocation, assuming the characteristic dimension of the rosette is $R\sim 10\,\mu$m. At a distance R from the dislocation the internal stress is $\sigma\sim 10^7$ erg/cm^3 and the magnetoelastic-anisotropy energy density is $\sigma\Lambda_{111}\sim 30$ erg/cm^3 (the magnetostriction constant is given in Ref. 8 as $\Lambda_{111}=-2.93\ 10^{-6}$). For the elastic energy of the inhomogeneous magnetostrictive strains one can estimate an upper bound of $G\Lambda_{111}^2 \sim 7$ erg/cm^3 ($G = 7.6\ 10^{11}$ erg/cm^3 is the elastic shear modulus). Finally, the surface-tension contribution is $\tau/R \sim 10$ erg/cm^3 (the surface energy of the phase boundary is $\tau \sim 10^{-2}$ erg/cm^2). The saturation magnetization at a temperature close to T_c is small:

$$M_s \approx \lambda\chi_{\parallel} M_{Fe}(T-T_c)/T_c,$$

and the magnetostatic energy density (at $T - T_c \approx 1$ K) can be assumed approximately equal to

$$2\pi M_s^2 \approx 5\cdot 10^{-5}\ M_{Fe}^2 \approx 1\ \text{erg/cm}^3$$

($\lambda\chi_{\parallel} \approx 1$, $M_{Fe} = 22$ emu/g).[9]

These estimates show that the magnetoelastic interaction is dominant, and in this sense one is justified in neglecting the remaining contributions. Nevertheless, for an exact quantitative calculation of the behavior of the domains of different phases during the spin flip it is necessary to include the energy of the intrinsic magnetostrictive strains and the surface tension of the phase boundaries.

We note in conclusion that detection and analysis of the domain rosettes arising around dislocations during spin-flip phase transitions is a convenient way to search for dislocations in a crystal and to determine rather accurately their characteristics.

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Translated by Steve Torstveit