

metry, a second-order phase transition can occur only with an increase of symmetry at the transition point. Moreover, an increase of symmetry at the transition point will also occur for systems possessing a discrete covariance group if the system of RG equations has only one stable fixed point.

For such systems the same critical indices as for systems with the higher symmetry should be observed experimentally. The structure of the critical fluctuations and, consequently, the increase in the symmetry of the system as $T \rightarrow T_c$ can be investigated by means of electron paramagnetic resonance (EPR), light-scattering experiments, acoustic methods, etc. In particular, data which apparently indicate a change in the symmetry of the field of the critical fluctuations in the structural phase transition in the cubic crystal SrTiO_3 have recently been obtained by the method of EPR spectroscopy.^[11]

In conclusion I express my gratitude to A. I. Sokolov for numerous discussions and useful criticism, and also to S. L. Ginzburg and S. V. Maleev for a discussion of the results of the work. I am sincerely grateful to D. E. Khmel'nitskiĭ and A. A. Migdal. Discussions with them on the structure of the RG equations have been of great benefit to me.

¹⁾It is implied that the power-law asymptotic form has already been separated out from the expressions for the invariant charges.

²⁾The GML equation in^[4] was obtained in the framework of a certain self-consistent scheme and is not, strictly speaking, an exact equation for the renormalized coupling constants. It

is possible, however, to adduce arguments that the values of the critical indices calculated using such an equation will be close to the true values.

³⁾The behavior of such a model in the framework of the ϵ -expansion was investigated earlier in^[3,5,8]. It is used to describe structural phase transitions from tetragonal to rhombic symmetry and also applies to the formation of superstructures in alloys forming a body-centered cubic lattice.^[5,10]

⁴⁾The notation is the same as in^[5].

⁵⁾We note that for certain special values of the constants $\{\lambda_k\}$ the transformation K can lead to couplings between different λ'_k or can make some of the λ'_k vanish. In this case the symmetry of \mathscr{H} will not coincide with the symmetry of \mathscr{H} and our assertion does not apply to these special cases.

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Translated by P. J. Shepherd

Phase diagram and domain-boundary structure in a uniaxial ferrimagnet near the compensation point

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(Submitted February 20, 1976)

Zh. Eksp. Teor. Fiz. **71**, 1443-1452 (October 1976)

The phase diagram of a quasi-uniaxial mixed rare-earth iron garnet has been investigated by magneto-optical methods. The lines of stability loss were determined for the low- and high-temperature collinear phases and the noncollinear phase, together with their ranges of coexistence. Near the triple point of the phase diagram, there was observed a broadening of the domain boundary between collinear phases, with subsequent transformation of the boundary to the noncollinear phase. It was shown that phase segregation in the specimen occurs over a wide range of temperatures and of magnetic fields, located within the single-domain range for the "Weiss" domains that are due to the demagnetizing fields. The structure of the transition regions between coexisting magnetic phases was investigated.

PACS numbers: 75.50.Gg, 75.60.Fk, 78.20.Ls

The continued interest in investigation of the behavior of ferrimagnets in the vicinity of their magnetic compensation point has recently increased significantly because of the discovery, in this region, of the phenomenon of coexistence of several magnetic phases; that is, of a distinctive domain structure, which exists over a quite wide range of variation of the temperature and of

the external magnetic field, including fields that appreciably exceed the field for "technical" saturation of the material.^[1-10] In experiments on iron garnets, which have good optical transparency in the visible and infrared ranges of wavelength, broad use is made of visual methods of investigation, based on the use of the Faraday and Cotton-Mouton magneto-optic effects.^[2-11]

In particular, measurement of the Faraday rotation in each of the coexisting magnetic phases and observation during dynamic change of these phases enable us to obtain complete information about the phase diagram of the magnetic material.

The present paper presents the results of an experimental investigation of the phase diagram of a quasi-uniaxial ferrimagnet and of the structure of the domain boundaries between coexisting phases near the magnetic compensation point. The results of the experiments agree qualitatively with the predictions of theory^[1]; the discrepancies observed can be explained by supposing that there is a composition gradient, which leads to a spatial variation of the compensation temperature in the specimen.

1. EXPERIMENT

The object of investigation was a film of iron garnet with bismuth content, of composition $(\text{YGdBiYb})_3(\text{FeAl})_5\text{O}_{12}$, with compensation temperature $T_c \approx 240$ K, thickness about $10 \mu\text{m}$, grown by the epitaxy method from the liquid phase on to a substrate of nonmagnetic gadolinium-gallium garnet, cut in a (111) plane.^[7] The film had anisotropy of the "easy-axis" type,^[1] with the axis of easy magnetization along a [111] direction, and had the following parameter values at room temperature: saturation magnetization $M_0 \approx 5.0$ G, uniaxial-anisotropy constant $K \approx 1.73 \cdot 10^4$ erg cm^{-3} , inhomogeneous-exchange constant $A \approx 6 \cdot 10^{-8}$ erg cm^{-1} ; the difference between the lattice constants of the film and the substrate was -0.01 \AA . The saturation magnetization and the inhomogeneous-exchange constant were determined by measurement of the characteristic diameters and bias fields for an isolated cylindrical magnetic domain^[12]; the value of the uniaxial-anisotropy constant was obtained by magnetizing the film by means of a field applied in its plane.^[13] The method used for measuring the anisotropy constant K enabled us to determine also the range of variation of K resulting from inhomogeneity of composition within the specimen. The values of K for different points of the specimen fluctuated between $1.67 \cdot 10^4$ erg cm^{-3} and $1.80 \cdot 10^4$ erg cm^{-3} ; that is, the relative variation $\Delta K/K$ was in order of magnitude ~ 0.1 . It may be assumed that the other film parameters (M_0 and A) at different points likewise varied within the same limits.

A significant contribution to the uniaxial anisotropy in the film under investigation came from the elastic stresses σ that resulted from mismatch of the lattice constants and coefficients of thermal expansion of the film and of the substrate. The magnitude of the elastic stresses was estimated by the formula^[14-16]

$$\sigma = \left[(1-\eta) \frac{a_s - a_f}{a_s} - \eta(\alpha_s - \alpha_f) \Delta T \right] \frac{E}{1-\mu} \quad (1)$$

where η is the coefficient of stress "relaxation" ($0 < \eta < 1$), a_s and a_f are the lattice constants of the substrate and of the film at room temperature, α_s and α_f are the coefficients of thermal expansion of the substrate and of the film, ΔT is the difference between the growth temperature and room temperature, and E is Young's modulus and μ Poisson's ratio for the film material. On

using the known values of the constants in formula (1),^[16-18] $a_s = 12.382 \text{ \AA}$, $\alpha_s = 9.2 \cdot 10^{-6} \text{ deg}^{-1}$, $\alpha_f = 10.4 \cdot 10^{-6} \text{ deg}^{-1}$, $\Delta T \approx 10^3 \text{ deg}$, $E \approx 2 \cdot 10^{12} \text{ dyn} \cdot \text{cm}^{-2}$, $\mu \approx 0.3$, and determining from the known composition the lattice parameter of the epitaxial film a_f (which was found to be 12.372 \AA), we find that $|\sigma_{\text{max}}| \approx 3 \cdot 10^9 \text{ dyn} \cdot \text{cm}^{-2}$. The potential contribution to the uniaxial anisotropy because of magnetostriction is determined by the expression

$$K_{ms} = \frac{3}{2} |\sigma_{\text{max}}| \lambda_{111}^{(\text{eff})} \quad (2)$$

where $\lambda_{111}^{(\text{eff})}$ is the effective magnetostriction constant, determined on the basis of the known values of magnetostriction constants λ_{111} of the "pure" rare-earth garnets $\text{Gd}_3\text{Fe}_5\text{O}_{12}$ ($-3.1 \cdot 10^{-6}$), $\text{Yb}_3\text{Fe}_5\text{O}_{12}$ ($-4.5 \cdot 10^{-6}$), and $\text{Y}_3\text{Fe}_5\text{O}_{12}$ ($-4.3 \cdot 10^{-6}$).^[19,20] Calculation of the effective magnetostriction constant, with allowance for dilution of the magnetic sublattices by diamagnetic ions, for a film of the chosen composition, gives the value $\lambda_{111}^{(\text{eff})} \sim -2.3 \cdot 10^{-6}$, whence we find by formula (2) that $K_{ms} \sim 1.0 \cdot 10^{-4}$ erg cm^{-3} . The surface of the film under investigation was smooth, without breaks or cracks; this indicates absence of significant inhomogeneity of the internal stresses.

The film was placed in a cryostat between the poles of an electromagnet, in such a way that the bias field was directed along the normal to the film surface (a [111] axis), and was illuminated with linearly polarized light at wavelength $0.63 \mu\text{m}$, propagated along the direction of the magnetic field (the geometry of the Faraday effect). Behind the film were placed an analyzer and a microscope, which made possible direct observation of the domain structure in the film and measurement of the Faraday rotation in each of the coexisting phases. The angle of rotation of the plane of polarization of light in the collinear phases was $\sim 10 \text{ deg}$; this made it possible to obtain a domain-structure image with sufficient contrast. The cryostat was provided with electronic apparatus for stabilization of the temperature; this made it possible to maintain the chosen value of T with accuracy 0.005 K over the range from 85 to 300 K . The temperature gradient in the specimen, when the system was stabilized at the chosen temperature over a period of several minutes, did not exceed 0.01 K mm^{-1} .

The temperature behavior of the ordinary "Weiss" domains,^[2] in the absence of a bias field ($H=0$), is illustrated by the photographs shown in Fig. 1, a-g. It is seen that with approach to the compensation point (either from lower or from higher temperatures), the domain size increases and reaches a certain critical value, after which the domains disappear. Thus near the compensation point there is a so-called single-domain range (Fig. 1d), the physical reason for whose existence is that the resultant magnetization of the specimen in this temperature range becomes so small that a subdivision into "Weiss" domains under the influence of the demagnetizing fields becomes energetically disadvantageous, and the specimen changes to a state of uniform magnetization.^[21,22] The temperature dependence of the size of the "Weiss" domains is shown in Fig. 2a.^[3] By doing similar experiments in the presence of a bias field, it is possible to determine the complete

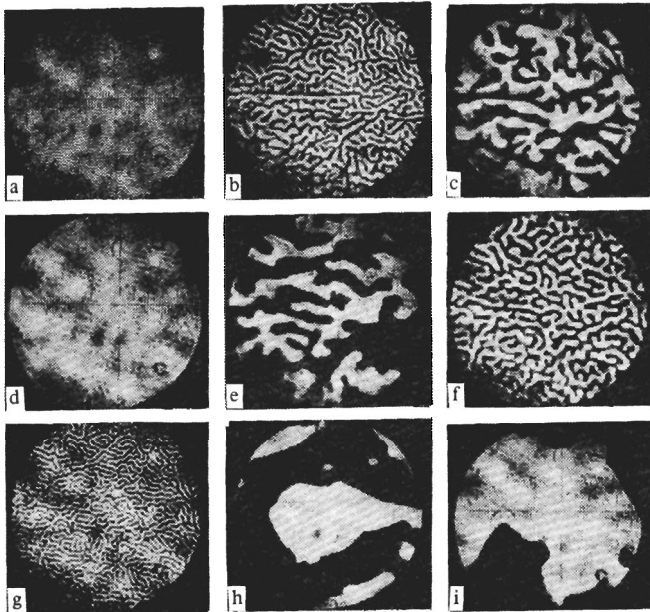


FIG. 1. Photographs of domain structure in absence of a bias field, with change of temperature: $T=195$ (a), 204 (b), 214 (c), 235 (d), 273 (e), 282 (f), 287 (g), 238.5 (h), 241.2 (i) K. Diameter of field of view about 3 mm.

single-domain range in the $(H-T)$ plane. The boundary of the single-domain range is plotted in Fig. 2b; the range of existence of "Weiss" domains is shaded. It is seen that near the compensation point there is in the $(H-T)$ plane a quite extensive region (the single-domain temperature range at $H=0$ is about 50 K), within which, according to the view prevailing until recently, the formation of a domain structure is energetically dis-

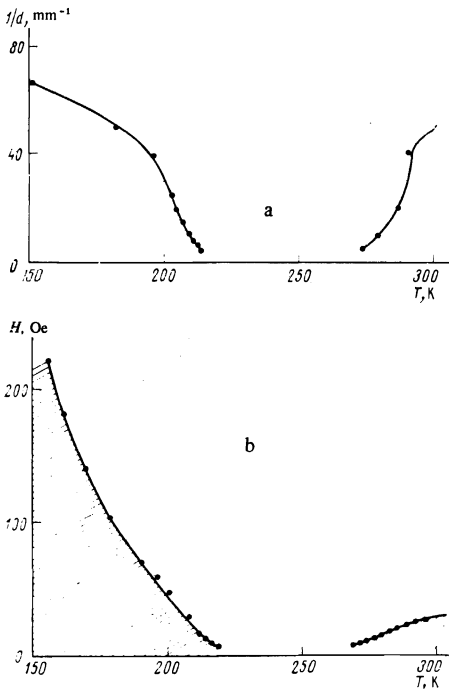


FIG. 2. a, Temperature dependence of domain size in the labyrinth structure, in absence of a bias field, b, Single-domain range of the specimen in the $(H-T)$ plane.

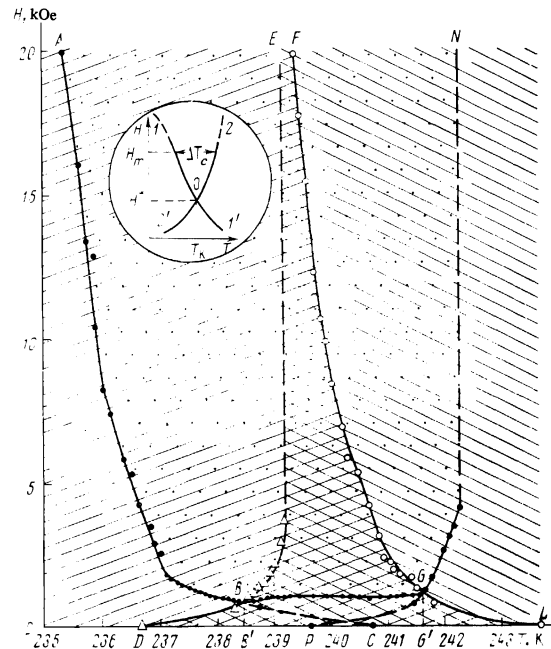


FIG. 3. Phase diagram of a quasiuniaxial ferrimagnet. In the insert, phase diagram of an "ideal" uniaxial ferrimagnet. Region $ABGN$ is the domain of existence of the noncollinear phase; the low-temperature collinear phase exists to the left of line FLG ; the high-temperature collinear phase, to the right of line EBD .

advantageous. But theoretical and experimental results presented in [1-10] show that, in actual fact, even inside this region there is a possibility for existence of domains, which differ radically from the "Weiss" domains in that they are not due to demagnetizing fields. In Fig. 3 the region of existence of domains of the new type in the $(H-T)$ plane, lying inside the single-domain range, is bounded by the lines $ABDLGN$. The form of the domains that exist in the region $DBGL$ is illustrated by the photographs in Fig. 1h, i and Fig. 4a.

Measurement of the Faraday rotation in each of the domains shows that in the present case there is coexistence of the low-temperature ($M_R > M_{Fe}$) and high-temperature ($M_R < M_{Fe}$) collinear phases (M_R and M_{Fe} are, respectively, the magnetizations of the rare-earth and iron sublattices). The observed domains are essentially quasiantiferromagnetic, for in this range of temperatures and of magnetic fields $M_R \approx M_{Fe}$. For a fixed magnetic field less than 1 kOe, upon increase of the temperature from the region to the left of point D , where only the low-temperature (LT) collinear phase exists, at a temperature corresponding to the intersection with the line BC there appears at the edge of the plate a nucleus of the high-temperature (HT) collinear phase; then a clearly expressed domain boundary (see Fig. 4a) moves smoothly through the specimen, and a complete change of phase (exit of the domain boundary from the field of view) is observed at a temperature corresponding to the intersection with the line GL . Upon lowering of the temperature, similar phenomena occur on the lines GP and BD ; the domain boundary in this case moves in the opposite direction. In the temperature range $T_B < T < T_C$ (T_B and T_C are the temperatures

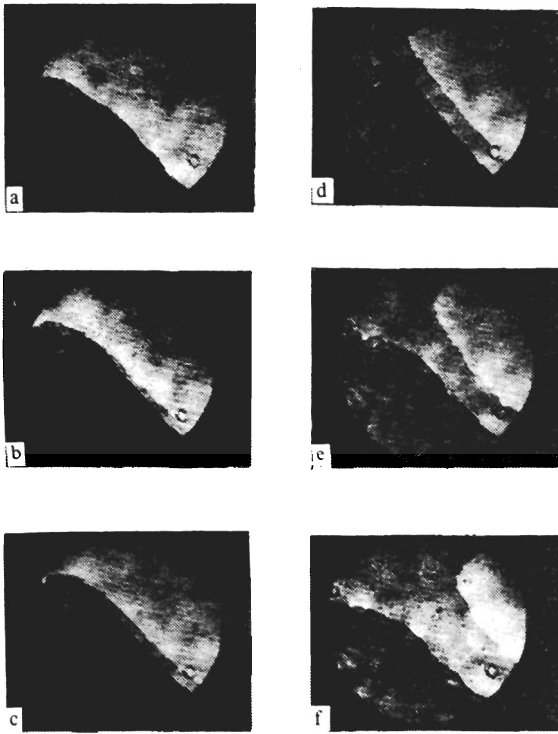


FIG. 4. Broadening of a domain boundary between collinear phases, with subsequent transformation of the boundary to the noncollinear phase. at $T = 240.8$ K. $H = 0.9$ (a), 1.2 (b), 1.5 (c), 2 (d), 2.5 (e), 3 (f) kOe.

corresponding to the points B' and G' in the phase plane), upon gradual increase of the bias field from zero, a new phase appears at fields corresponding to the lines BC and PG . By acting on the specimen with a weak (< 1 kOe) nonuniform bias field, let us say by bringing a permanent magnet near the specimen, it is possible to produce quasiantiferromagnetic domains over the whole region $DBGL$, without dependence on the previous history of the specimen. In this case the domains may have a most fantastic form (Figs. 1h, i). We remark that the points D and L do not lend themselves to accurate experimental determination.

If we start from the region $B'BGG'$ and gradually increase the bias field, at a certain threshold field H^* , whose value is practically independent of temperature and is ~ 1 kOe, there occurs a broadening of the domain boundary between the collinear phases, with subsequent transformation of the domain boundary to the noncollinear phase (Fig. 4). Noticeable is the persistence of quite clear domain boundaries between the noncollinear and both noncollinear phases. The coexistence of three magnetic phases is observed over a quite extensive range of temperatures and of magnetic fields ($EBGF$ in Fig. 3).

Theoretical calculation of the critical field H^* , corresponding to the beginning of the broadening of the domain boundary between the two collinear phases, leads to the following expression^[23]:

$$H^* \approx (KH_{cd}/M_{Fe})^{1/2}, \quad (3)$$

where H_{cd} is the effective field of exchange interaction

between the tetrahedral (d) and dodecahedral (c) magnetic sublattices. In undiluted rare-earth iron garnets the value of the exchange field H_{cd} is $\sim 10^5$ Oe^[24]; for our specimen this value must be cut in half, because each sublattice is diluted by diamagnetic ions to the extent of about one third. As a result we find from formula (3) that $H^* \approx 10^3$ Oe; this agrees with the value of the field observed in the experiment.

The visual monitoring of the motion of domain boundaries made it possible to obtain the complete phase diagram of the film in the $(H-T)$ plane, which is shown in Fig. 3. Change of the relative volumes of two or three coexisting phases in the region $ABGN$ of the phase diagram occurred by motion of the interfaces between the phases (domain boundaries), just as in the region $B'BGG'$. But the lines AB , BE , FG , and GN differ qualitatively from the lines DB , BC , PG , and GL . Thus, for example, upon crossing of line AB during a transition from region ABD (the region of existence of the LT collinear phase alone) into region ABE , the noncollinear phase appears; upon return from region ABE to region ABD , the noncollinear phase disappears approximately on the same curve AB . The observed hysteresis (about 0.1° for fixed field in the range $1 \text{ kOe} < H < 20 \text{ kOe}$; not shown on Fig. 3) is significantly less than that that exists in weak fields ($H < 1$ kOe) and may be caused by interaction of the domain boundary with defects.

On the right boundary for existence of the noncollinear phase (line GN), in fields ~ 6 kOe, there was observed near defects a discontinuous appearance of cylindrical magnetic domains (CMD) of size $\sim 100 \mu\text{m}$. With further increase of field, the CMD were stable over the whole available range of fields (up to 20 kOe). With decrease of field, the CMD persisted down to fields ~ 1 kOe, after which they collapsed at $H < 1$ kOe. The size of the CMD within the range of stability was practically independent both of the field H and of the temperature. Annihilation of a CMD within the stability range was observed only when a domain wall between the noncollinear and the HT collinear phases crept up to it during a change of field or of temperature. The observed CMD were isolated cylindrical regions of the noncollinear phase within the HT collinear phase.

The structure of the domain boundaries between phases is determined by the number of coexisting phases, the bias field, and the temperature. When three magnetic phases coexist, the noncollinear phase plays the role of a 180-degree domain boundary between the collinear phases. But the structure of such a "domain boundary" differs essentially from that of a simple Bloch wall.^[25] By way of example, Fig. 5 shows, for the case of three coexisting phases and at three values of the bias field H , the variation of the angle $\theta = \arccos(\Phi/\Phi_{\max})$, where Φ and Φ_{\max} are the angles of rotation of the plane of polarization in the noncollinear and the collinear phases respectively, with the coordinate in the film plane in the direction perpendicular to the interface between the phases. Since in garnets that contain bismuth the Faraday rotation is determined principally by the diamagnetic rotation of the bismuth ions in the exchange field of the iron ions,^[26] θ represents the angle between the bias

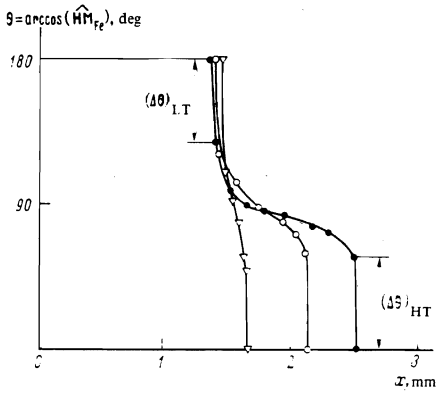


FIG. 5. Variation with coordinate x of the angle between the resultant magnetization of the iron sublattices and the magnetic field, in coexisting phases.

field H and the resultant magnetization vector M_{Fe} of the iron sublattices. It is seen that there are narrow transition sections between the collinear and the noncollinear phases, with rapid change of the angle θ , which in Fig. 5 are provisionally shown as jumps through $(\Delta\theta)_{LT}$ and $(\Delta\theta)_{HT}$; inside the noncollinear phase, the rotation of the magnetization proceeds more smoothly. For $H \approx H^*$, the magnitude of the jumps $|(\Delta\theta)_{LT,HT}|$ approaches 90° ; then in the noncollinear phase the ferrimagnet is essentially in a state like the state of an antiferromagnet with anisotropy of the "easy plane" type. With increase of the field, the magnitude of the jumps $\Delta\theta$ decreases. The variation of the jumps $(\Delta\theta)_{LT}$ and $(\Delta\theta)_{HT}$ with the magnetic field, at several typical temperatures, is plotted in Fig. 6; it is shown not only for the case in which three phases coexist (Fig. 6b, c) but also for the two-phase state (Fig. 6a, d).

We remark that in the film investigated, an appreciable paraprocess of the Faraday rotation was observed in the collinear phase.⁵ In the LT collinear phase far from the compensation point, on change of the field from 1 to 20 kOe the Faraday rotation changed from 10.8° to 12.5° ; in the HT collinear phase, from -10.6° to -7.7° . With approach to the compensation point, the paraprocess in the LT collinear phase changes little; in the HT collinear phase, at temperatures slightly above the compensation point the paraprocess is practically absent, but it then slowly increases to approximately the same value as in the LT collinear phase.⁶ The observed change of the Faraday rotation in the paraprocess range enables us to estimate the magnitude of the effective field acting on the magneto-optically active ions. It is found to be $\sim 10^5$ Oe, which corresponds in order of magnitude to the effective exchange field exerted by the iron sublattices (with allowance for their dilution by diamagnetic ions) on ions located in the dodecahedral interstices. Since the rare-earth ions in bismuth-containing garnets make no appreciable contribution to the rotation, the magnitude of the paraprocess of the Faraday rotation in the film under investigation indirectly supports the hypothesis that the huge Faraday rotation in bismuth-containing films is due to diamagnetic rotation of the bismuth ions in the exchange field of the iron sublattices.

2. DISCUSSION OF RESULTS

The results obtained, in particular the form of the phase diagram for the magnetic material presented in Fig. 3, can be explained on the basis of the assumption that there exists in the specimen a composition gradient, which leads to a spatial variation of the compensation temperature. In fact, let us assume that the compensation temperature in the film is nonconstant; then the form and character of motion of the domain boundaries justify us in stating that the compensation temperature varies monotonically in the direction perpendicular to the direction of preferred orientation of the domain boundaries (see Fig. 4a). Then on one edge of the film (we shall by convention call it the "left") the compensation temperature will be lowest, and on the other (the "right") highest. Sufficiently small regions of the specimen may be considered homogeneous in composition; therefore the "local" phase diagram should have the usual form for a uniaxial ferrimagnet (see¹¹), shown schematically in the insert in Fig. 3. Curves 101 and 202 are the lines of stability loss for the LT and HT collinear phases, respectively; OT_c is a line of phase transitions of the first kind; 10 and 20 are lines of phase transitions of the second kind; O is a triple point. The peculiarities of the phase diagram near the triple point are not pictured in the insert, because they relate to a very small region of the phase space and can hardly show up in specimens with an inhomogeneity of properties.

Comparison of the ideal phase diagram with that actually observed shows that the latter can be obtained by simple superposition of "local" phase diagrams corresponding to different points of the specimen. Thus curves ABC and DBE may be considered lines of stability loss of the collinear phases on the phase diagram for the left edge of the specimen, where the compensation temperature is 238.45 K, while curves FGL and NGP are the analogous lines for the right side of the specimen, with compensation temperature 241.6 K. Thus the change ΔT_c of compensation temperature across the specimen is about 3° . The line BG corresponds to the

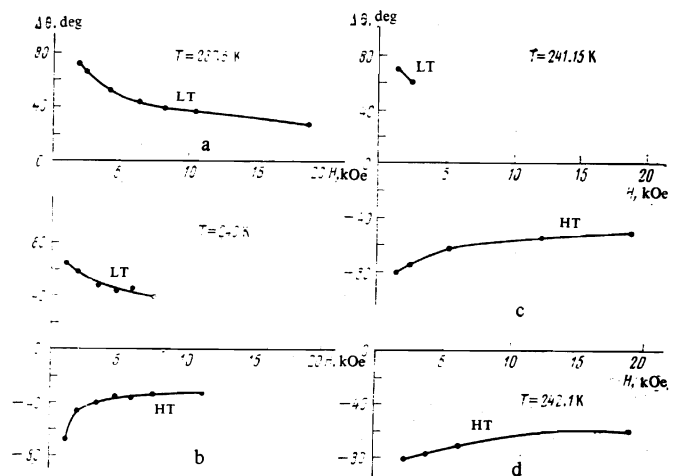


FIG. 6. Variation of $(\Delta\theta)_{LT}$ and $(\Delta\theta)_{HT}$ with magnetic field for the three-phase (b, c) and the two-phase (a, d) states.

triple points existing in various regions of the specimen. At the critical point all orientations of the quasicollinear phases become energetically equivalent; therefore with approach to this point from below (with respect to the magnetic field), there occurs a broadening of the domain boundary between the collinear phases. In an ideal crystal, above the triple point, in the region 102, only the noncollinear phase should exist; but in our experiments, in the region *EBGF* simultaneous coexistence of three magnetic phases was observed. Analysis of the "ideal" phase diagram shows that in specimens with a variation of the compensation temperature, such a phenomenon is actually possible in the field interval from H^* to a certain field H_m (see insert in Fig. 3), at which the temperature width of the region of existence of the noncollinear phase becomes equal to ΔT_c ; for $H > H_m$, only existence of two magnetic phases is possible. Nonuniformity of composition may also be the reason for the existence of sharp interfaces between the collinear and the noncollinear phases. The presence of a composition gradient in the quasiuniaxial film investigated, which leads to a variation of compensation temperature across the specimen, permits easy explanation also of other experimentally observed effects, namely: induction by a magnetic field $H < H^*$ of separation into LT and HT collinear phases in the region *B'BGG'*; the difference of the magnetization distribution from the simple Bloch type in a domain boundary between the LT and HT collinear phases; etc.

All the phenomena described above and a phase diagram of the form shown in Fig. 3 will occur in any real crystal, with appropriate change of scale along the temperature axis. Strictly speaking, for a specimen with inhomogeneity of composition the concepts "phase" and "phase transition" lose all meaning; but since inhomogeneities are always present even in the most perfect specimens, the use of these concepts is in some degree justified.

The authors express their gratitude to A. K. Zvezdin, B. A. Ivanov, A. A. Mukhin, E. P. Stefanovskii, V. V. Tarasenko, and D. A. Yablonskii for discussion of the research, to I. G. Avaeva for providing the specimens, and to O. E. Lisitska for help in the conduct of the experiments.

¹Strictly speaking, epitaxial iron-garnet films should be considered quasi-uniaxial, for a residual cubic anisotropy is always present in them.

²By "Weiss" domains we shall understand domains that owe their existence to the stray fields produced by magnetic charges on the specimen surface.

³To eliminate the effect of defects, whose coercive force increases greatly near the compensation point, the domains were "shaken" before each measurement by an alternating magnetic field orientated in the plane of the film.

⁴Evidence in favor of this assumption is provided by results of an investigation of the paraprocess in the collinear phases (see later).

⁵The change of the Faraday rotation because of the paraprocess was taken into consideration in the plotting of the graphs in

Fig. 6.

⁶The reasons for this phenomenon are not fully understood.

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Translated by W. F. Brown, Jr.