Field-induced spin-orientational phase transitions in the Néel ferrimagnets. II
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Field-Induced Spin-Orientational Phase Transitions in the Néel Ferrimagnets. II.

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The breakdown of the aligned state of rare-earth ferrite garnets in an applied magnetic field is considered. Comparison is made between experimental and theoretical results on the field- and temperature-dependences of the phase regions of the aligned and canted phases in the H–T–plane, the behaviour of magnetization, the specific heat, the magneto-caloric effect, and the orientation of magnetic sublattice moments in the canted phase. It is emphasized that even for a rough quantitative description of the phase diagram of rare-earth ferrite garnets with a high magnetic compensation temperature the approximation of rigid binding between the Fe-sublattices is inadequate, and it is necessary to take into account the field-induced weak noncollinearity of the Fe-sublattices.

1 INTRODUCTION

A change in the magnetic symmetry with break-up of the ferrimagnetic collinear structure must affect its different physical properties. Kinks, rapid changes, discontinuities in the magnetization (Clark and Callen, 1968; Clark and Alben, 1970; Fillon and Hug, 1970; Levitin, Ponomarev and Popov, 1970; Ferron, Fillon, Hug et al., 1971, 1972, 1973), sharp increases or jumps in the magnetocaloric effect (Clark and Callen, 1969; Clark and Alben, 1970; Belov et al., 1970), specific heat and entropy (Ferron et al., 1972; Kamilov and Shakhsaev, 1972), magnetostriction (Belov, Levitin, Ponomarev and Popov 1969; Levitin et al., 1970) and in the magneto-optical properties (Kharchenko, Eremenko et al., 1967, 1968, 1975, 1976; Grzhegorzhevski and Pisarev, 1973; Gnatchenko and Kharchenko, 1976; Gnatchenko, Kharchenko, Konovalov and Puzikov, 1977) are observed.
The majority of experimental work on the collinear structure disintegration process has been carried out on rare-earth iron garnets near their magnetic compensation temperatures. Rare-earth iron garnets, in which the ion distribution over the crystallographic sites is of a quite definite and unambiguous character, are the most suitable materials for studying this kind of magnetic transformation.

The magnetic compensation temperature, near which the values of the critical fields are suddenly reduced, allows experiments to be carried out under stationary conditions. The experimental results prove the applicability of the phenomenological treatment within the framework of the molecular field approximation to the description of the phase boundaries. However, the thermal properties (e.g. heat capacity, heat of transitions) are beyond the limits of this approximate theory.

The following sections deal with the major experimental results obtained with different techniques. We shall mainly consider magneto-optical methods and the results of studying the phase diagrams of cubic and uniaxial three-sublattice ferrimagnets.

2 MAGNETO-OPTICAL METHODS FOR THE STUDY OF CANTED FERRIMAGNETIC STRUCTURES

In a number of cases it is convenient to study magnetic phase diagrams by magneto-optical methods taking advantage of the fact that the optical properties of a magnetic crystal depend on the orientation of the sublattice magnetic moments relative to the light propagation vector. This dependence is especially strong in crystals with non-equivalent sublattices because partial contributions of individual sublattices to the magneto-optical properties of the crystal may be essentially different. This is, for example, possible in iron-garnets where, with proper selection of the rare-earth ion and the spectral region, a situation may be achieved where the contribution of one of the lattices to the resulting Faraday effect is predominant and it is possible to trace the motion of the magnetic moment of the chosen magneto-optically active sublattice. Figure 1 gives the relative contributions to the magneto-optical rotation of the polarization plane of the rare-earth sublattices ReIG depending on the light wavelength.

Another advantage of magneto-optical methods is the possibility of observing the sample macroscopic magnetic structure and of identifying the magnetic state in different domains by means of optical measurements in small sections of the sample.

The optical properties of a ferromagnet or aligned ferrimagnet crystal depend on the orientation of the magnetic moment relative to the crystal-
lographic axes. The optical indicatrix deforms and changes the orientation of its axes depending on the orientation of $\mathbf{M}$. In a canted ferrimagnet the form and position of the indicatrix depends also on the angle between the magnetic moments of the sublattices. The noncollinear ferrimagnet is always biaxial as there are no high-order rotation axes among its magnetic symmetry elements. The dependence of the orientation and deformation of the optical indicatrix of a ferrimagnet on the direction of the vectors $\mathbf{M}_i$ has been discussed by Pisarev, Sinii and Smolenskii (1969) and Zvezdin, Markarov, Popov and Red'ko (1972). The reconstruction of the

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**FIGURE 1** Ratio of proportionality factors between the Faraday rotation and magnetization of the rare-earth sublattice in ReIG as a function of the light wavelength.
directions of magnetic moments from the results of polarization studies is generally complicated and tedious even in the case of the two-sublattice structure; it can be substantially simplified in special situations, especially when the contribution of the rare-earth sublattice is insignificant, and the deformation of the indicatrix with the deviation of the sublattice magnetic moments from the high-order symmetry axes is also negligible. This kind of situation occurs in gadolinium iron garnet, GdIG. Here, because the ground state of the Gd$^{3+}$ ion is deprived of orbital moment and because of the remoteness of its first excited energy levels (the allowed transition in the electric dipole approximation), the contribution of the gadolinium sublattice to the magnetic circular birefringence is insignificant (Figure 1). Its contribution to linear birefringence is also small (Dillon, Remeika and Staton, 1969; Pisarev, Sinii, Kolpakova and Yakovlev, 1971. Moreover, the ratio of $\Delta n(M \parallel \{100\})$ and $\Delta n(M \parallel \{111\})$ in GdIG is close to unity and the angle between the optic axes appearing when $M$ deviates from the directions $[111]$ or $[100]$ does not exceed 15°. The additional change of the angle caused by the canting of the sublattices is basically determined by the very small deviation of the angle between the Fe sublattices from π and is insignificant for fields $H < H_F$. Thus, gadolinium iron garnet can be viewed as an optically uni-
axial crystal whose indicatrix is not deformed with the canting of the sub-
lattices. In this case, the phase shift of the linearly-polarized light is expressed by the simple relation

$$\delta = \frac{\Delta n_0 l 2\pi}{\lambda} \sin^2 \theta$$  \hspace{1cm} (2.1)

Here $\Delta n_0$ is the spontaneous magnetic birefringence in the direction perpendicular to the optic axis, $l$ is the sample thickness, $\lambda$ is the light wavelength, $\theta$ is the angle between the direction of light propagation in the crystal $K$ and the optic axis of the crystal. The direction of the latter is determined by the position of the vectors $M_1$ and $M_2$. Of course light waves propagating in the crystal in the general case have elliptical polarization owing to magnetic gyration and $\delta$ is the phase shift between the light components in the absence of the Faraday rotation. For the rotation of the polarization plane when $K \parallel H$ in ReIG in the absence of the linear birefringence effect we can write the expression

$$\phi = [D(\omega)\sigma_1 + A(\omega)\sigma_2] \cos \theta \cos \psi + [D(\omega)\sigma_1 - A(\omega)\sigma_2] \sin \theta \sin \psi + C(\omega)M_3 \cos \theta_3 + \phi_{\text{Res}} + F(\omega)H$$  \hspace{1cm} (2.2)

Here $\phi_{\text{Res}}$ is the contribution made to the rotation by the magnetic dipole resonance absorption in the radio-frequency and the far-infrared spectral regions; $A, D, C$ are frequency-dependent factors of proportionality between
the Faraday rotation and the magnetic moment of the corresponding sub-
lattice (Cooper, Crossley, Page and Pearson, 1968, Abulatya and Le Gall,
1972). The last term is due to the action of the magnetic field on the energy
states of the crystal (Kharchenko et al., 1968, 1969) (Figure 2) \( \theta = \frac{1}{2}(\theta_1 + \theta_2) \),
\( \psi = \frac{1}{2}(\theta_2 - \theta_1) \), \( \theta_i \) is the angle between the directions of the sublattice
moment \( M_i \) in the initial collinear and canted states. The second term may
become significant for fields \( H \ll H_z \) only at \( \theta \approx \frac{1}{2} \pi \), while remaining small
in absolute value because the canting angle of the Fe sublattices does not
exceed 1-2°. The contributions of individual terms in Eq. (2.2) to light
with wavelength 6328 Å may roughly be characterized by \( A = -330 \text{ deg/cm } \mu_B \),
\( D = -160 \text{ deg/cm } \mu_B \), \( C = -2.3 \text{ deg/cm } \mu_B \), \( F = -2 \text{ deg/cm kOe } \).
\( \phi_{\text{Res}}(M_o) = 4 \text{ deg/cm} \). Confining ourselves to the most important terms we can see
that, having measured \( \phi \) and knowing \( F \) and the spontaneous rotation \( \phi_0 \) of
the aligned ferrimagnet, it is also possible to find the rotation angle \( \theta \) of the
Fe sublattice magnetic moments.

In rare-earth iron garnets circular and linear magnetic birefringences are
comparable in order of magnitude (Dillon et al., 1969; Pisarev et al., 1971). This
may lead to certain difficulties in the interpretation of the results of polariza-
tion measurements. Simultaneous action of the effects cause a situation
where, after passing through the sample, light becomes elliptically polarized,
the azimuth of the principal axis of the polarization ellipse and the relation
between its axes being determined by both effects. The analysis of the action of
the gyrotropic birefringent sample on the polarization of incident light is

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**FIGURE 2** Variation of Faraday rotation of YIG in a magnetic field.
conveniently made with the help of the Poincaré sphere method (Ramachandran and Ramaseshan, 1952; Sherkliﬀ, 1962). If we assign the symbols \( \lambda \) and \( \lambda_1 \) to the azimuths of the principal axes of the polarization ellipses of the incident and outgoing waves relative to the principal plane of a crystal, \( \Delta = (\delta^2 + 4\rho^2)^{1/2} \) to the phase difference of two orthogonal elliptically polarized waves propagating in the crystal, \( \delta \) and \( 2\rho \) to analogous phase differences between linearly and circularly-polarized waves in the absence of circular and linear birefringence respectively, \( \tan 2\gamma = 2\rho/\delta \), and \( \tan \omega \) to the ellipticity of the light wave, we have the following expressions for the parameters characterizing polarization changes of linearly-polarized light. The azimuth of the ellipse axis of outgoing light:

\[
\tan 2\lambda_1 = \frac{\tan 2\lambda \cos \Delta + \sin 2\gamma \sin \Delta}{\cos^2 2\gamma + \sin^2 2\gamma \cos \Delta - \sin 2\gamma \sin \Delta \tan 2\lambda} \tag{2.3}
\]

the change of the azimuth (the rotation of the principal axis of the ellipse):

\[
\tan 2(\lambda_1 - \lambda) = \frac{\sin 2\gamma \sin \Delta(1 + \tan^2 2\lambda) + \cos^2 2\gamma \tan 2\lambda(\cos \Delta - 1)}{\cos^2 2\gamma + \sin^2 2\gamma \cos \Delta + \tan^2 2\gamma \cos \Delta}, \tag{2.4}
\]

ellipticity

\[
\varepsilon = \tan \omega,
\]

where

\[
\sin 2\omega = \cos 2\gamma \cos 2\lambda(\sin 2\gamma - \sin 2\gamma \cos \Delta + \tan 2\lambda \sin \Delta) \tag{2.5}
\]

When the incident light wave is polarized in the principal plane of the sample or at an angle of \( 45^\circ \) to it, the expressions for the changes in azimuth become simpler:

\[
\tan_{0^\circ} 2(\lambda_1 - \lambda) = \frac{\sin 2\gamma \sin \Delta}{\cos^2 2\gamma + \sin^2 2\gamma \cos \Delta} \tag{2.6}
\]

for \( \lambda = 0 \) and

\[
\tan_{45^\circ} 2(\lambda_1 - \lambda) = \frac{\sin 2\gamma \sin \Delta}{\cos \Delta} \tag{2.7}
\]

for \( \lambda = 45^\circ \).

It can be seen from expressions (2.4)–(2.7) that the contribution of linear birefringence to the rotation of the axis of the polarization ellipse at small \( \delta \) is less than \( \delta^2/8 \), which in sufﬁciently thin samples can be signiﬁcantly smaller than the Faraday rotation. In gadolinium iron garnet the maximum additional rotation determined by the linear birefringence can be some hundredths of a degree if the sample is 50 \( \mu \)m thick. The Faraday rotation for this thickness is
close to $1^{\circ}$ and, consequently, at angles $\theta$ not very close to $\frac{1}{2}\pi$ the measured rotation of the polarization plane is almost entirely determined by circular birefringence.

If the incident light has circular polarization, the azimuthal angle of the axis of the ellipse of the outgoing wave is determined by the expression:

$$\tan 2\lambda_1 = \frac{\sin \Delta}{\sin 2\gamma (1 - \cos \Delta)} \quad \text{or} \quad \lambda_1 = \frac{1}{2} \arctan \left( \frac{\Delta}{2\rho} \cotan \frac{\Delta}{2} \right) \quad (2.8)$$

and the ellipticity by the expression:

$$\sin 2\omega_1 = \sin^2 2\gamma + \cos^2 2\gamma \cos \Delta \quad \text{or} \quad \varepsilon = \tan \left( \frac{\pi}{4} - \arcsin \left( \frac{\delta \sin \frac{\Delta}{2}}{2} \right) \right) \quad (2.9)$$

From Eq. (2.8) we can see that the azimuthal angle of the ellipse axis at small $\Delta$ is close to $45^{\circ}$

$$\tan \lambda_1 \approx 1 - \rho$$

and the deviation of ellipticity from unity is determined, as can be seen from Eq. (2.9), at small $\Delta$ only by the linear birefringence value

$$\varepsilon = 1 - \delta$$

Thus, measuring the rotation of the polarization plane of linearly-polarized light in gadolinium iron garnet, we can determine directly the cosine of the rotation angle of the magneto-active iron sublattices, measured from the direction $\mathbf{K}$. The measurement of ellipticity when using circularly-polarized incident light gives the sine squared of this angle, and the azimuthal angle of the polarization ellipse axis determines the rotation plane of the sublattice magnetic moments.

In the experiment it is useful to employ both kinds of light polarization as well as two geometries: longitudinal, in which the field and light propagation direction are collinear, and transverse, in which $\mathbf{K} \perp \mathbf{H}$. With $\mathbf{K} \parallel \mathbf{H}$ and linear polarization of the incident light, magnetic twins, differing in the azimuthal angle of the sublattice rotation plane, give equal contributions to the Faraday rotation. But they cause different orientations of the polarization ellipse when the incident light is polarized circularly and can be seen by eye. However, as a result of the small value of the ellipticity and insufficient quality of circular polarizers it is convenient to study magnetic twins in the transverse geometry. Null-methods can be conveniently used to register the angle of rotation of the polarization plane (or the principal axis of the polarizability ellipse). Figure 3 shows the compensation scheme for measuring the rotation of the polarization plane together with a device for visual control which
allows one to make measurements in small sections of the sample. The operating principle is clear from this figure. The modulator of the light beam over the polarization plane operates in a way analogous to a half-shade device and switches the polarization plane according to the law $\beta = \beta_0 \sin \Omega t$, and acts at the same time as an analyzer. The polarization plane of the light beam incident on the analyzer makes an angle $\alpha = \frac{1}{2} \pi + \beta_0 \sin \Omega t + \phi$ with the analyzer transmission axis. The intensity of the light passing through the system (ignoring losses) equals

$$I = I_0 \sin^2(\phi + \beta_0 \sin \Omega t)$$  \hspace{1cm} (2.10)

At small angles $\phi$ and $\beta_0$ this equals

$$I = I_0 \left[ \phi^2 + \frac{\beta_0}{2} + 2\phi \beta_0 \sin \Omega t - \frac{\beta_0}{2} \cos 2\Omega t \right]$$  \hspace{1cm} (2.11)

The harmonic of $\Omega$ occurs when the rotation angle of the polarization plane due to the sample is different from zero and can be used for measuring or as the input signal in the compensation circuit. In the circuit in Figure 3 a Faraday cell is the compensator and gives a current proportional to $\phi$. A mirror diaphragm (MD) is placed in the image plane of the objective where an enlarged image of the sample (S) can be seen. A fraction of the light which has passed through the diaphragm placed in a specially selected position of the sample image enters the measuring system. The size of the section studied depends on the enlarged sample image-to-diaphragm ratio. Most of the light passing through the sample is reflected by the mirror and is used for visual control. The position of the measured portion is found by moving the mirror diaphragm.
In pulsed magnetic fields a convenient analyzer is a Wollaston prism turned at an angle of 45° to the polarizer axis. The two beams coming out of the prism fall on different photomultipliers, and the difference in their signals is registered. The system has a reduced sensitivity to the light flux instability. The dependence of the polarization plane rotation on the magnetic field strength is recorded in pulse measurements on an oscilloscope in coordinates of rotation-field strength.

As was mentioned above, measurement of the linear birefringence is convenient using circularly-polarized light. To measure the ellipticity of the polarization of the outgoing light beam in stationary conditions a rotating linear analyzer (Figure 4) is used. The intensity of the light emerging from the sample and the analyzer is then

\[ I = \frac{1}{2} I_0 (1 + \sin 2r \cos 2\Omega t) \quad \text{where} \quad \sin r = \frac{\delta}{\Delta} \sin \frac{\Delta}{2} \quad (2.12) \]

At small \( \Delta \) the modulation depth \( I_-/I_- \) depends fully on the value of the linear birefringence

\[ I = \frac{1}{2} I_0 (1 + \delta \cos 2\Omega t) \quad (2.13) \]

At small \( \rho \) the phase of the modulated signal is determined by the principal sublattice rotation plane in the sample. If the rotation plane remains constant in the process of measurement, a synchronous detector can be used to increase the signal-to-noise ratio. The value of \( I_- \) can be easily measured by passing the linearly-polarized light through the rotating analyzer. A sufficient amount of polarization of the circularly-polarized light can be obtained with two birefringent plates.

The arrangement for visual study of the domains, where the moments of the magneto-optically active sublattice form various angles with the vector \( \mathbf{K} \) and
the field vector $\mathbf{H}$, is shown schematically in Figure 5a. Figure 5b illustrates the observation method, incorporating linearly-polarized light, which makes it possible to study magnetic twins in which the angle between $\mathbf{M}_i$ and $\mathbf{H}$ remains constant. But when the light propagation vector is perpendicular to $\mathbf{H}$, the projections of $\mathbf{M}_i$ on $\mathbf{K}$ are different in different twins, the Faraday rotation being also different. The scheme does not allow one to distinguish the states in which the vectors are in the plane of the sample (twins I and IV in

FIGURE 5 Scheme for visual observation of (a) coexistent magnetic phases and (b) energy-equivalent magnetic twins.
Figure 5b). However this can be done if the sample plate is deflected from its normal position towards the direction of light propagation.

In studying the domain structure it is more convenient to use white light, which has an advantage over monochromatic light owing to the difference in the absorption and Faraday rotation dispersion. The competition between these two properties causes the appearance of a colour domain picture which increases the visual contrast between the domains and helps to identify the domains and traces their changes during observation.

3 MAGNETIC PHASE DIAGRAMS OF CUBIC FERRIMAGNETS AND COEXISTENCE OF MAGNETIC STATES

As was mentioned above, GdIG is a convenient material for magneto-optical investigations. It was investigated by the Faraday rotation method by Bernasconi and Kuse (1971); Grzegorzhevskii and Pisarev (1973); Kharchenko et al. (1974, 1975); Gnatchenko and Kharchenko (1976). The purely spin origin of the magnetic moment and isotropic g-factor makes this ferrimagnet the simplest substance for magnetic studies. The latter greatly simplifies the calculation of its phase diagram. But the majority of exchange constants needed for the calculations and the great variability of their values, obtained by different methods (Belov, 1972) complicates the calculation of the diagram even for this well-studied ferrimagnet. It can be seen from part I of this review (Eremenko and Kharchenko, 1979) that to construct the magnetic phase diagram of the anisotropic ferrimagnet in the vicinity of \( K_{\text{comp}} \) and \( H \ll H_F \) it is sufficient to know the net anisotropy constant \( K = K_1 + K_2 + K_3 \) and the ratio \( \beta/\alpha \), which is a certain combination of exchange parameters determined directly from the slopes of the straight lines limiting the region of the canted structure in fields where the contribution of magnetic anisotropy is negligible. The appearance of the canted phase may be indicated by a decrease of the Faraday rotation when a certain field and temperature region have been achieved. Figures 6 and 7a show the rotation as a function of pulse and steady fields, and Figure 7b shows the rotation as a function of temperature. Fields \( H' \) and temperatures \( T_1 \) can be identified with the fields and temperatures of the transition of the crystal or its sections into the canted state provided the sample is sufficiently far from \( T_{\text{comp}} \) (when the rotation in weaker fields has achieved its spontaneous value). Other peculiarities in the \( \phi(H) \) and \( \phi(T) \) curves may be caused by more or less sharp changes of the content of magnetic phases characterized by the different values of rotation angle in the magnetically uniform sample. The appearance of the canted phase can also be shown by a decrease of the ellipticity of
the initially circularly-polarized light (Figure 8). The temperatures at which the linear birefringence appears (the ellipticity of the light decreases) can be determined but with a large error. Yet the values obtained agree with the temperatures $T_1$ from the data on the rotation of the polarization plane. The appearance of singularities in the curves $I_-/I_+ = f(T)$ agrees with the appearance of singularities in the $\phi(T)$ curves. The increase of the ratio $I_-/I_+$ (or the decrease of the ellipticity of the light wave) is connected with the rotation of the sublattices and the appearance of non-zero projections on
FIGURE 7 (a) Faraday rotation ($\lambda = 6328$ A) as a function of the stationary magnetic field strength in GdIG near $T_{\text{comp}}$; dashed line indicates the Faraday rotation of cryostat windows and optical lenses. (b) Temperature-dependence of magnetic rotation of the light polarization plane in GdIG near $T_{\text{comp}}$ at fixed magnetic field strength. $H||[111]$.

the plane orthogonal to the direction of the light propagation. Dips in the curves may be caused by magnetic twins formed which compensate the phase difference between the optical modes appearing in an individual domain. Inaccuracy in orientation of $H$ relative to the axis $[111]$ or the presence of local stresses may lead to the initial predominant appearance of one of the three possible magnetic twins (visual control is difficult because of the weakness of the effect). Predominantly two canted structures with the azimuths of the rotation planes differing by $\pi$ should be found in the immediate vicinity of
FIGURE 8  Variation of ellipticity of circularly-polarized light in a magnetic field $\mathbf{H}||[111]$ for a noncollinear GdIG near $T_{\text{comp}}$, and change in azimuth of magnetic sublattice rotation plane:
(a) temperature dependence of amplitude of alternating signal $I_+/I_-$ recorded using a phase detector set at the constant phase of the reference signal.
The most complete magnetization data for GdIG have been published by Fillon and Hug (1970). Figure 30a shows a representative curve of $M(H)$, illustrating the kinks which appear when the ferrimagnet goes into the canted state; their positions were used for constructing the phase field of the canted state. By plotting in the $(H-T)$ plane the field strength and the temperature at which the singularities are observed on different curves, we obtain lines approximating to the stability boundaries of the collinear state. The result is given in Figure 9. The points are plotted for several representative curves with good agreement between the points obtained by different methods. In magnetic fields weaker than 30 kOe the points are grouped near two types of lines. With increase of magnetic field strength the lines approach each other and in fields exceeding 30–40 kOe they become indistinguishable, the points being placed nearly on a straight line. The slopes of the straight lines at $T > T_{\text{comp}}$ and $T < T_{\text{comp}}$ are somewhat different. The mean value of the slope $H_1/(T - T_{\text{comp}})$ is close to 7.8 kOe/K.

From the slope using (3.30b) of part I we can determine the ratio

$$\frac{\beta}{\alpha} = \frac{\mu_3}{m_3 k B_{S_3}^{-1} (M_{3\text{comp}}/M_{30})} \cdot \frac{H_1}{|T - T_{\text{comp}}|}$$

(3.1)

assuming that the resultant magnetization of the Fe sublattices in GdIG is the same as in YIG (Anderson, 1964) (at $T = T_{\text{comp}}$ it is 22.2 emu/g, and $M_{30} = 124.4$ emu/g). The value obtained $\beta/\alpha = 8.9$ may be used for constructing stability lines of various magnetic phases. It may also be used for determining the exchange constants $\lambda_{13}$ and $\lambda_{23}$ at a given $\lambda_{12}$. For this we must additionally determine the effective exchange constant $\lambda = (\lambda_{13} \sigma_1 + \lambda_{23} \sigma_2)(\sigma_1 + \sigma_2)^{-1}$. This can be obtained from the value $T_{\text{comp}} = 285.5$ K and the magnetization of the gadolinium sublattice in $T_{\text{comp}}$ equal to 22.2 emu/g:

$$\lambda = B_{S_3}^{-1} \left( M_{3\text{comp}}/M_{30} \right) \cdot \frac{kT_{\text{comp}}}{\mu_3 (M_1 - M_2)} = -11.570 \text{ Oe} \cdot \text{g/emu}$$

(3.2)
FIGURE 9 Stability boundaries of the aligned phases in GdIG with \( H\parallel[111] \). Solid thick lines—three-sublattice model, calculated with the aid of Eq. (3.46 of part 1) at \( \beta/\alpha = 8.9 \); thin lines—two-sublattice model (\( \kappa_{1,2} \to \infty, \beta/\alpha = 33 \)). Experimental points were obtained from pulse (\( \bigcirc \)) and stationary (\( \times \)) measurements of Faraday rotation (field \( H' \)) and from temperature dependence measurements: \( \nabla - T_1, \Delta - T_2 \). Points \( \bullet \) — the data from the work of Fillon and Hug (1970).
and $-\lambda(M_1 - M_2) = 3.56 \cdot 10^5 \text{kOe}$. To determine $M_1$ and $M_2$ we have used the temperature dependences of the sublattice magnetizations $a$ and $d$ given by Litster and Benedek (1966). The values $\lambda_{13}$ and $\lambda_{23}$ (in Oe·g·emu$^{-1}$) obtained for various $\lambda_{12}$ are given below (Table I). The value of the magnetic anisotropy constant needed for the construction of the phase diagram lines has been taken from the work of Podrique, Meyer and Jones (1960) to be equal to $K = 6.7 \cdot 10^3 \text{erg/cm}^3 = -1.04 \cdot 10^3 \text{erg/g}$ ($\kappa = 1.85 \cdot 10^{-4}$). The solid line in Figure 9 shows the phase boundaries of the aligned phases calculated on the basis of these data. The thin solid lines outline the boundaries of aligned phases at $\lambda_{12} \rightarrow \infty$. The slope of the latter in the linear region is close to $29 \text{kOe/K}$ and exceeds the experimental value $7.8 \text{kOe/K}$ by more than a factor of 4. This comparison shows the necessity to take into account noncollinearity of the Fe sublattices in a quantitative treatment of the sublattice canting in rare-earth iron garnets. The quoted experimental results indirectly prove the existence of orientational first-order magnetic phase transitions induced by the magnetic field and coexistence of canted configurations. But these experimental results are not enough for a complete reconstruction of the magnetic phase diagram of GdIG in the region of the fields $H \leq (H_{\text{eff}})_{1/2}$, where the phase diagram is rather complicated. Direct visual observations of the sample in this region close to $T_{\text{comp}}$ allow this to be done.

Nonuniformity of the magnetic structure in the region of existence of canted magnetic structures hampers a detailed study of magnetic phase diagrams of a cubic ferrimagnet by traditional methods. Visual polarization methods allow one to record the appearance of a new magnetic state and together with the measurements of the rotation angle of the light-polarization plane, allow one to construct with good accuracy the phase diagrams at different field orientations. The longitudinal geometry of the experiment with the field $\mathbf{H}$ parallel to the light propagation vector $\mathbf{K}$ is better suited for their construction. This geometry makes the magnetic twins, differing only in azimuthal angles of the sublattice magnetic moments but having identical projections of the sublattice moments on the $H$ direction, invisible in observations based on the Faraday rotation of the polarization plane. The problem of constructing the magnetic phase regions is then greatly simplified.

**Table I**

<table>
<thead>
<tr>
<th>$\lambda_{12}$</th>
<th>$\lambda_{13}$</th>
<th>$\lambda_{23}$</th>
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<tbody>
<tr>
<td>$\lambda_{ad}$</td>
<td>$\lambda_{bc}$</td>
<td>$\lambda_{ac}$</td>
</tr>
<tr>
<td>$-91,000$ (Anderson, 1964 b)</td>
<td>$-3,800$</td>
<td>$-540$</td>
</tr>
<tr>
<td>$-80,800$ (Lister and Benedek, 1966)</td>
<td>$-4,250$</td>
<td>$-1,160$</td>
</tr>
</tbody>
</table>

*The molecular field coefficients are given in Oe·g·emu$^{-1}$.\)
Visual observations have shown that the mechanism for the transition of the gadolinium iron garnet from the aligned to the canted state qualitatively differs when the magnetic field is parallel to the easy \( \mathbf{H} \parallel [111] \) and difficult \( \mathbf{H} \parallel [100] \) magnetization axes. In the former case the canted phase appears in the form of individual growing regions and in the latter there is a smooth uniform transition from the aligned to the canted state. Abrupt orientational
transitions between the different canted structures have been observed in both cases.

Let us consider \( H || [111] \) and \( H || [100] \) in greater detail. If the field is oriented along [111], as the sample temperature approaches the magnetic compensation temperature \( T_{\text{comp}} \) clearly outlined sections appear in it, with optical density (at fixed polarizer positions) different from that of the crystal. With further approach to \( T_{\text{comp}} \) and then with increasing difference from it, new regions with different optical densities appear and replace one another. By varying the position of the analyzer and by measuring the Faraday rotation in individual domains, it is possible to distinguish and identify four types of domain. Figure 10 illustrates the successive alternation of the sample magnetic states with changing temperature: the low-temperature aligned state (grey sections) gives way to the low-temperature canted state (dark sections). Then there appear domains in the high-temperature canted state (new grey regions) which are replaced in turn by the high-temperature aligned states (light sections). The contrast between the neighbouring domains changes little with temperature but decreases noticeably when the field strength is increased. In fields of strength higher than 9kOe it is possible to observe simultaneously only two types of domain. After one state is replaced by another, the sample becomes homogeneous and then the next alternation of states takes place.

In weak fields the boundaries between the domains are frequently localized near scratches or other surface defects. In strong fields the arrangement of the boundaries is not affected by surface defects. The boundaries are frequently not perpendicular to the sample surface and during observation different states project onto each other. Nevertheless, by tracing the positions of the phase boundaries it is possible to determine reliably the moments when domains of various types appear and vanish in weak fields. A diagram of the sample magnetic states, obtained visually, is shown in Figure 11. Here lines \( 1_{\text{exp}}, 4_{\text{exp}} \) are the phase boundaries of the high- and low-temperature collinear phases. The phase regions of the high- and low-temperature canted phases are outlined by lines \( 2_{\text{exp}}-2_{\text{exp}} \) and \( 3_{\text{exp}}-3_{\text{exp}} \), respectively. At field strengths higher than 9kOe the coexisting magnetic phases are separated by a region corresponding to the single-phase state of the ferrimagnet. In fields \( H < 4kOe \) simultaneous existence of all four types of magnetic phase is possible. The figure also shows the phase boundaries calculated with equations (3.46–3.48) of part I. In the calculations we used the experimentally found value \( \beta/\alpha = 8.9 \) as well as the exchange and anisotropy constants given above. The figures near the curves correspond to the same ferrimagnetic states as the ones near the experimental curves. Comparison of the boundaries obtained shows agreement between the calculated and experimental results in the sense that observed phase regions are entirely within the corresponding calculated ones.
The sublattice rotation angle in the coexisting phases could be determined by measuring the Faraday rotation in a magnetically uniform section of the sample using a diaphragm placed in the plane of the magnified image of the crystal. By moving the diaphragm over the sample image or by shifting the domain boundaries by slightly changing the temperature it was possible to determine the law governing the variation of the Faraday rotation and the sublattice rotation angle in canted phases with changing temperature or field. Figure 12 shows the characteristic changes of the Faraday rotation by a section of a sample of 75 μm diameter with changing temperature and fixed magnetic
Rotation of sublattices in GdIG at a fixed magnetic field of 14 kOe parallel to [111], and at variable temperature; the noisy curve—the experimental dependence of the Faraday rotation by a sample section of 75 μm diameter; solid lines—calculated dependence of the rotation angle; dashed lines—regions of metastable states; dash-dot lines—temperature ranges in which the coexistence of magnetic phases was observed.

The steep sections correspond to the passage of phase boundaries through the working region of the sample and to replacement of one state by another. Knowing the spontaneous rotation $\phi$ at $M_i \parallel H$ found in the aligned state of GdIG it is possible to find the cosine of the rotation angle of the magnetic moment of the active sublattice and to construct a diagram of its motion (insert, Figure 12). Dependence of the cosine of the rotation angle of the Fe sublattice magnetic moments on temperature at fixed magnetic field strength $H = 14$ kOe was calculated by Eq. (3.48). Dotted lines show temperature boundaries of the calculated metastable state regions, and dash-dot lines show the experimentally observed phase coexistence regions. There is good agreement between experimental and calculated curves in fields $H > 10$ kOe. At weaker field strength the central jump in rotation angle is smaller than the calculated one. This decrease is probably connected with domains of different phases overlapping in weak fields. In fields $H < 5$ kOe it is impossible to distinguish the three steep sections of the $\phi(T)$ curve because of the overlapping of domains.
FIGURE 13 Domain structure appearing upon reorientation of magnetic moments of the GdI2G sublattices for $\mathbf{H}||[100]$: (a) $H = 7$ kOe, $T = 284.75$ K (1); 284.86 K (2); 284.91 K (3); 284.98 K (4); 285.16 K (5); 285.25 K (6). (b) A sample with many surface defects, temperature is constant, $T = 284.9$ K, $H = 0.2$ kOe (7); 5.8 kOe (8); 13.3 kOe (9).

When the external field is directed along the difficult magnetization axis [100], visual observations through crossed polarizers show that the appearance of the domains is preceded by uniform bleaching of the sample as its temperature approaches $T_{\text{comp}}$; this bleaching corresponds to a smooth rotation of the sublattices. Close to $T_{\text{comp}}$, in a temperature interval of approximate width 1°K, the magnetic uniformity of the sample breaks down. The sample breaks into domains of two types differing in sign and value of Faraday rotation. Figure 13 shows the character of the appearance and growth of the new phase with increasing temperature. The motion of the boundaries in fields weaker than 10 kOe is frequently discontinuous. The irreversibility of the boundary motion manifests itself particularly strongly at
low field strength (up to 5–6 kOe), when the boundaries are localized near surface defects. In fields stronger than 9–10 kOe the surface defects have a smaller influence on the boundary position, and the shape of the domains changes appreciably (Figure 13). With increasing field the contrast between the domains decreases as well as the average size of the domain, and the form of the domain also changes. The decrease in the size of the domains is probably related to the fact that with increasing field the rotation angle of the magnetic domains within the domain wall decreases, as well as its energy. Figure 14 shows the temperature dependences of the Faraday rotation measured in a sample of 75 μm diameter. The smooth change of the angle of the polarization plane corresponds to smooth transition of the ferrimagnet from the aligned to the canted state, in agreement with the prediction of the theory that, in this case, the aligned-canted state transition is of second-order. Near the compensation point in fields less than 10 kOe there is a distinct kink in the Faraday rotation which corresponds to the passage of the interphase boundary through the studied section of the sample. Simultaneously with the change in the Faraday effect we carried out visual observations of the domain structure, which allowed us to record the value of the Faraday rotation at the moment of appearance and disappearance of the nonuniform magnetic state. During the time the domain boundary travelled through the measured section the crystal the temperature remained practically unchanged, and the measured values of the Faraday rotation angles of the polarization plane at the instant of appearance of the new magnetic state and vanishing of the old one give the rotation angles of the sublattices in the coexisting phases. The inset in Figure 14 shows the rotation of the resultant magnetic moment of the Fe sublattices with changing temperature, reconstructed from the temperature-dependence of the Faraday rotation, and the solid line is the calculated dependence of the cosine of the sublattice rotation angle on temperature. Near $T_{\text{comp}}$ in sufficiently weak fields there can be observed a small (about a few tenths of a degree) thermal hysteresis of the Faraday rotation.

With increase of field the jump in Faraday rotation $\Delta \phi$ on the boundary decreases. This behaviour is in qualitative agreement with the idea that the transition is analogous to the liquid-vapour phase transition and the phase transition line must end with a critical point, where the differences between low- and high-temperature canted phases disappear. The jump $\Delta \phi$ must go to zero in the critical field $H_{\text{cool}}$. Using equation (3.43) of part I we can calculate the region of possible values of $\Delta \phi/2\phi_0 = f(H)$. The region has an upper limit given by the curve of the difference of cosines of the sublattice rotation angles in the coexisting phases (Figure 15)

$$\frac{1}{2} (\cos \theta_1 - \cos \theta_2) = \frac{1}{\sqrt{3}} \left(1 - \frac{h^2 x}{\beta k}\right)^{1/2}$$

(3.3a)
FIGURE 14 Rotation of sublattices in GdIG at fixed magnetic field $H \| [100]$ and at variable temperature. Solid smooth lines show the calculated dependence of the cosine of the sublattice rotation angle. Experimental curves are the dependence, $\phi/\phi_0(T)$ at $H = 3.2$ kOe (a), 10.5 kOe (b).
FIGURE 15  Magnetic strength dependence of differences of Faraday rotation angles and differences of sublattice rotation angles in uniform states of GdIG closest to $T_{\text{comp}}$. 1, 2—calculated plots of the difference of the cosines of the sublattice rotation angles in stable and metastable canted states; 3, 4—a plot of $\Delta \cos \theta$ vs. $H$ at $\delta T_{\text{comp}}$ equal to 0.1 and 0.2 K, respectively; 5—width of thermal hysteresis as a function of magnetic field strength.
shown in Figure 15 by line 1. The lower limit is given by the curve of the difference of cosines of the sublattice rotation angles in the metastable and stable phases most remote from $T_{\text{comp}}$ (line 2):

$$\frac{1}{2} (\cos \theta_1' - \cos \theta_2') = \frac{1}{2} (\cos \theta_1'' - \cos \theta_2'') = \frac{1}{2} \left( 1 - \frac{h^2 \pi}{\beta \kappa} \right)^{1/2}$$  \hspace{2cm} (3.3b)

The experimentally-found difference of the angles of the Faraday rotation at the instant of appearance and disappearance of the non-homogeneous structure $\Delta \phi/2 \phi_0 = f(H)$ is also shown in Figure 15. The calculation of the critical field from Eq. (3.44) in part I gives the value $H_{\text{cool}} = 10.5$ kOe. In fields above this value the magnetic state of the sample should be uniform but the sample is magnetically nonuniform even in the field $H_{\text{cool}} = 15$ kOe, though the domains in these fields are small and the boundaries between them are indistinct. The existence of the nonuniform magnetic structure in fields $H > H_{\text{cool}}$ is probably connected with the variation of the compensation temperature over the sample. The change in $T_{\text{comp}}$ may be caused by lattice defects and associated microstresses. With increasing distance from the structural defect the associated stresses decrease. This results in a diffuse magnetic nonuniformity with unclear boundaries. Figure 15 schematically shows the temperature-dependences of the Faraday rotation at $H > H_{\text{cool}}$ in the two uniform sections of the crystal whose compensation temperatures $T_{\text{comp}}^{(1)}$ and $T_{\text{comp}}^{(2)}$ differ by a small value $\delta T_{\text{comp}}$. Far from the compensation points the difference of the Faraday angles for these sections is not great and they are indistinguishable. But within the interval $(T_{\text{comp}}^{(1)}, T_{\text{comp}}^{(2)})$ the difference of the Faraday rotation in the sections is much greater and sections with different $T_{\text{comp}}$ are visually observable. It is possible to compare the dependence of $\Delta_{\text{comp}}(\cos \theta)$ characterizing the contrast between nonuniform sections on field strength with the experimental dependence $\Delta \phi/2 \phi_0(H)$. Calculated curves for $\delta T_{\text{comp}} = 0.1$ and 0.2 K are given in Figure 15 and agree satisfactorily with experiment. Using the results of the work of Bloch, Chaisse and Pauthenet (1967) it is possible to estimate roughly the elastic stresses necessary to shift $T_{\text{comp}}$ by 0.2 K. The necessary hydrostatic pressure could be obtained in the crystal near dislocations and point defects which appeared as the crystal grew. Thus, we may arrive at the conclusion that the existence of the nonuniform magnetic structure in fields greater than 10 kOe is connected with imperfections of the monocrystal GdIG. This conclusion is also borne out by the dependence of the width of the thermal hysteresis on field strength (Figure 15). The absence of thermal hysteresis in fields $H > 10$ kOe may be connected with vanishing of the domain walls in a field stronger than the critical one. However, it is necessary to point out that the nature of the magnetic nonuniformity may be different. A decrease of wall energy close to $H_{\text{cool}}$ can bring about a situation where the domain boundary
becomes thermodynamically favoured owing to the increasing relative contribution of its entropy. We shall discuss this question at the end of this section.

The measured dependences of the Faraday rotation angle on temperature and the visual observations of the magnetic state of the sample allow one to construct the phase diagram of GdIG for $H || [100]$ (Figure 16). The temperature at which the canted structure appears was assumed to be the temperature at which the sample decreases its Faraday rotation angle by a value comparable to the noise (Figure 14). The coexistence region of the canted states was determined from visual observation data. The lines shown in the figure which delimit the region of the canted structures have been calculated from Eqs. (3.40) of part I and the boundaries of the region of metastable states from Eq. (3.45). Despite the approximate nature of the method employed to determine the temperature of the transition to the canted state, the agreement between the calculation and experiment should be considered as good.

In conclusion we shall discuss the possible causes of the appearance of the nonuniform magnetic state in the vicinity of first order-phase transitions.
First of all it is necessary to note that the demagnetization fields cannot be responsible for the observed coexistence of different magnetic phases because the magnetization jumps occurring during the orientational transitions in GdIG do not exceed, at the least, several gauss. The estimates of changes of magnetization occurring during the transition of the ferrite from the aligned to the canted state of \( \mathbf{H} \parallel [111] \) in a field of 14 kOe give the value of the jump to be 0.2 gauss. The domain structure period due to the action of the demagnetization fields should exceed the dimensions of the sample (of the order 1 cm). In addition, the temperature interval over which the domain structure exists, if the mechanism of its appearance is magneto-static, should not exceed
\[
4\pi \Delta M \left( \frac{dH_i}{d(T - T_{\text{comp}})} \right)^{-1} \sim 10^{-2} \text{ K}
\]
Moreover, with the magnetic moments of the neighbouring phases differing so slightly, the formation of the domains is energetically unfavoured as the decrease of the magnetostatic energy does not compensate for the increase of energy due to the formation of the domain boundaries. Equating the energies of a plane-parallel plate broken into Kittel domains with a magnetically uniform one we find that the domain structure is not favoured at
\[
\Delta M < (1.7E_w/l_3)
\]
(here \( E_w \) is the energy of the domain wall, \( l_3 \) is its thickness). In the case of gadolinium iron garnet, 90° wall and \( l_3 = 5 \cdot 10^{-3} \text{ cm} \)
\[
(1.7E_w/l_3)^{1/2} \approx 10 \text{ gauss}
\]
The causes of magnetic stratification of the sample should be analogous to those leading to the appearance of the domain structure in antiferromagnets (Farztdinov, 1964, Hubert, 1974). In particular, the domain wall may become energetically favoured in a real sample with lattice defects, owing to the lowering of the elastic energy of the crystal at the plane where the wall is localized as a result of magnetoelastic interaction. Boundaries of this kind can stabilize the non-uniform state of the sample with sections of thermodynamically stable and metastable phase. Moreover, the appearing domain structure can reduce the elastic energy of the stressed macroscopic sections of the sample. This decrease of elastic energy can compensate for the loss of energy when domain boundaries and metastable states are formed. In this case the role of demagnetization fields is played by long-range elastic stresses, and the formation of the domain structure is a result of the interaction of this field with magnetostrictive stresses.

In the ideal crystal domain walls can also be thermodynamically favoured if the entropy of interphase boundaries, whose position is not fixed by the demagnetization fields, is taken into account (Li, 1956; Farztdinov, 1964). As a result of thermal motion the domain walls can shift from the equilibrium position a distance of several lattice constants which causes an increase of the transition-layer entropy. The increment of energy of the domain boundary \( E_w \) can be compensated by the gain in entropy \((-TS_w)\). The entropy contribution can be especially important in the vicinity of the critical point in the case of \( \mathbf{H} \parallel [100] \), where the energy of the domain boundary tends to zero.
High symmetry of the crystal should also facilitate an increase in the role of the boundary entropy. We cannot, of course, strictly speak about a domain wall here, as the sample in this case broke into layers whose sizes were comparable with the width of the transition layer.

4 MAGNETIC TWINS IN THE CANTED STATE OF A CUBIC FERRIMAGNET

The magnetic field along one of the crystallographic symmetry axes reduces the symmetry of a crystal at the transition to the canted state; it eliminates the rotation elements of symmetry and the reflection operation in the plane normal to the magnetic moment plane. The canted state is characterized by the group of remaining symmetry elements, which is a subgroup of the space group of the collinear state. Therefore, a more symmetric aligned magnetic configuration may change to a less symmetric canted one in \( n \) ways. Thus, the crystal may break into \( n \) magnetic domains (magnetic twins), within each domain the transition being realized by one of the \( n \) ways. The symmetry operations lost are rotatory, and when they act, the magnetic twins transform from one to another. A crystal of sufficiently large size (or rather a set of crystals) retains its high initial symmetry according to the Curie symmetry principle. In our case, with \( H \parallel [001] \) the fourfold axis is lost and there is a possibility of four types of magnetic twins. As the field increases and the temperature changes the rotation of the sublattices is continuous in the (110), (110), (1 \( \overline{1} \) 0) or (\( \overline{1} \overline{1} 0 \))-planes. Similarly, for \( H \parallel [111] \) three types of twin can exist. The twinned structure may be formed because of either first- or second-order phase transitions. Since interdomain boundaries increase the crystal energy, the break up into thermodynamically equivalent domains is not energetically favoured as much as in the case of a domain structure with coexisting stable and metastable phases. This break-up can result from the action of the mechanisms given in the previous section. Furthermore, in the course of growth of the new phase with a randomly-oriented sublattice rotation plane when there are three or more types of twins, the formation of optimum angles at line and point boundaries of these regions is probable; the latter impede the subsequent boundary displacement and may result in the formation of a domain structure, which, though thermodynamically unfavoured, is stable kinetically (Lifshitz, 1962).

In observations of thermodynamically-equivalent domains in a canted sample, it is convenient to use a transverse experimental geometry where \( k \perp H \) (Figure 5b). Visual observation in polarized light shows that the GdIG sample in the magnetic field becomes magnetically nonuniform as its
FIGURE 17  Typical domain pictures observed in GdIG-plates at temperatures near $T_{\text{comp}}$. The magnetic field is directed along the plate surface. (a) a sample with mechanically-polished surface; $T$: (1) 278.2 K, (2) 278.8 K, (3) 279.9 K; $H = 14$ kOe, $T_{\text{comp}} = 279.5$ K; (b) a sample cut from the crystal with marked growth tracks, surface mechanically-polished, $T$: (1) 284.0 K; (2) 283.7 K, $T_{\text{comp}} = 282$ K, $H = 14.3$ kOe; (c) a thermally-treated sample with chemically-polished surfaces, $T$: (1) 278.4 K, (2) 280.3 K, (3) 280.7 K, $H = 13.8$ kOe, $T_{\text{comp}} = 279.5$ K.

temperature approaches the magnetic compensation temperature (Eremenko, Kharchenko and Gnatchenko, 1974; Lisovskii and Shapovalov, 1974; Gnatchenko and Kharchenko, 1976). The type of domain structure is very dependent on the state of the sample surface, conditions of growth, and thermal and magnetic histories. Figure 17 presents photographs of typical domain structures observed in some samples. In plates of mechanically-polished surfaces (Figure 17a) domains appear, usually near surface defects. For samples cut from crystals with pronounced directions of growth, a banded domain structure with bands parallel to traces of growth is typically found.
PHASE TRANSITIONS IN NEEL FERRIMAGNETS

(Figure 17b). The domain structure of chemically polished samples (Figure 17a) is characterized by closed drop-like domains with fine, hardly noticeable bands which seem also to be due to defects appearing in the crystal growth. The domain structure of chemically-polished specimens varies with time.

In spite of the great diversity of the domain structures observed, the temperature ranges in which the magnetic state of the sample is inhomogeneous, varied hardly from sample to sample. This fact, along with the high sensitivity of the domain structure type to the magnetic field orientation and the exchange of well-defined movable domain walls, indicates that the domain formation is due to the appearance of some energetically stable magnetic states near \( T_c \). The domain structures observed are difficult to identify because of the domain boundaries parallel to the developed sample surface. A slight deviation of the vector \( \mathbf{H} \) from the axis of high-order symmetry decreases the number of boundaries parallel to the surface, and even at an angle of deviation of \( 3^\circ \) domains penetrate through the sample. By rotating the sample through a small angle about the axis perpendicular to the vectors \( \mathbf{K} \) and \( \mathbf{H} \) and by measuring the Faraday rotation in a separate domain, the polar and azimuthal angles of the vector of the magneto-active sublattice, \( \mathbf{M}_a \), can be determined, taking into account the cubic symmetry of the crystal.

Figure 18 illustrates variations of the Faraday rotation of a small section of the sample (75 \( \mu \)m dia), when domains of different types pass through it at variable temperature for the two orientations of \( \mathbf{H} \). In the inset (see also the insets in Figure 21 and 25) figures 1–8 denote directions of easy magnetization of the \( \langle 111 \rangle \) type. Small rotations of the sample about its vertical axis cause an opposite change in the Faraday rotation of different domains which rotate the polarization plane equally when \( \mathbf{H} \) is parallel to the \[ 100 \] or \[ 111 \] axes, and result in a more distinct differentiation in domains, in particular, when \( \mathbf{H} \) is close to \[ 100 \] (Figure 18). The directions of sublattice magnetic moments in a selected domain are not constant with temperature or field. In the domain of one type different orientations of \( \mathbf{M}_i \) correspond to different \( H \) and \( T \), \( M_1 \) and \( M_2 \) being closer to one of the easy directions but not coinciding with it. To emphasize this fact, different types of domains are denoted by figures \( 1' \cdots 8' \). Thus, in the domain of type \( 1' \) the easy direction closest to \( M_2 \) is the direction \( 1 \), and the angle between \( M_2 \) and the direction \( 1 \) varies with \( H \) and \( T \).

When \( \mathbf{H} \) makes a certain angle \( \alpha \) with the axes \[ 100 \] or \[ 111 \], the directions of \( M_2 \) will differ (at the same values of \( H \) and \( T \)) from the directions of \( M_2 \) in the case of strict orientation. Emphasizing this fact, the domains are denoted by figures \( 1'' \cdots 8'' \) for \( \alpha \neq 0 \). In the domain of type \( 1'M_2 \) makes a minimum angle with the easy direction \( 1 \), but the orientation of \( M_2 \) with unchanged \( H \) and \( T \) differs from the orientation of \( M_2 \) in domains of type \( 1' \) for strict field orientation.
FIGURE 18. Faraday rotation in a sample section of 75 μm diameter: (a) H close to the [100] axis; (b) H close to [111] axis; in both cases \( H = 13.8 \text{ kOe} \).
By identifying all types of domains, the positions of their boundaries can be observed visually owing to distinctions in tints of the separate domains and their overlap sections. By determining the instant of appearance and disappearance of domains of a certain type, the existence region of a given magnetic state can be constructed in the H–T-plane. Below, we consider results for H along [100] and [111].

1) H || [100]. In this case the aligned-canted state transition is smooth, but because of the existence of four equivalent planes, a nonuniform state can be formed. In fields lower than the critical one, there are first-order phase transitions between the canted states in a narrow temperature range; an additional stratification of the sample is also possible. A typical feature of this is the existence of a uniform smooth rotation of the sublattice magnetic moments preceding the appearance of domains. If the field is made to lie as close as possible to the (110)-plane of the sample, the smooth rotation is not observed up to the temperature where the domains appear. Therefore it seems to occur in the sample plane. A small angular deviation (less than 0.5°) of the field direction from the sample plane transfers the magnetic moments to another plane and makes it possible to observe the rotation. The rotation sign is dependent on the direction of deviation of the (110)-plane from the vector H. Though the Faraday rotation due to the sublattice rotation is seen as $T_{\text{comp}}$ is approached, beginning from $T_1$ and $T'_1$ (Figure 19) the magnetic twins are only observed in a temperature range considerably smaller than $T_1T'_1$. Figure 19 shows the parts of the temperature curves of the Faraday rotation which correspond to the appearance of canted states in GdIG. $T_2$ and $T'_2$ are the temperatures at which the magnetic twins are observed. The types of domains are illustrated in Figure 20. Here, the angle between the analyzer and the polarizer is somewhat different from 90°. The formation of twins is seen in photographs 1 and 2: smeared, poorly defined patches in photograph 1 and twins formed with a distinct boundary in photograph 2. As the temperature approaches closer to $T_c$, a temperature can be distinguished at which a distinct domain structure appears. Photographs 3 to 5 show large twins penetrating through the sample. As the temperature rises further, the domains vanish and a smooth uniform rotation of the moments towards the H-direction takes place. Many photographs show the upper part of the sample with a section of unmoved boundaries—a block of somewhat different compensation temperature. The moments the domains appear and disappear are shifted on the temperature scale compared with the remaining part of the sample.

If the appearance and disappearance temperatures of different types of domains are taken, we can plot the phase diagram shown in Fig. 21. The dotted lines $A$ and $A'$ are the boundaries delineating the region of the canted
FIGURE 19 Temperature dependences of Faraday rotation in the vicinity of transitions to the canted magnetic structures. The [100]-direction makes an angle of about 0.5° with the vector $\mathbf{H}$; $T_1$ and $T_1'$ are temperatures at which the Faraday rotation is comparable to noise signals, $T_2$ and $T_2'$ are temperatures at which magnetic twins are observed.
FIGURE 20 Field-induced magnetic twins in GdIG near $T_{comp}$ at $H = 14.1$ kOe; the direction of H is close to [100]; temperature is expressed in °K: (1) 275.21; (2) 276.45; (3) 277.92; (4) 278.26; (5) 278.82; (6) 279.13; (7) 279.62; (8) 279.75; (9) 279.92; (10) 280.05; (11) 280.29; (12) 280.65; (13) 280.73; (14) 280.87; (15) 282.25.
states. The regions of existence of well-defined domains of types 1', 2', 3', 4' and 5', 6', 7', 8' frequently penetrating through the sample are bounded by the lines C', D and C, D' respectively. The lines D and D' delineate the regions of nonequivalent magnetic states of types 1', 2', 3', 4' and 5', 6', 7', 8'. Triangles indicate the temperatures $T_2'$ at which the magnetic twins just formed are seen. Solid lines B and B' near these points are the calculated curves of the aligned canted-state phase transitions of second order.

2) $\mathbf{H} \parallel [100]$. Unlike the previous case, for $\mathbf{H} \parallel [111]$ there is no smooth rotation of sublattices. The process of magnetic moment reorientation starts as domains appear in which the direction of magnetic sublattices is close to the directions of equivalent easy axes 2, 3, 4. There is practically no temperature range in which interdomain walls are formed. The development of domains as the temperature changes is shown in Figure 22. In photograph 1 the sample is still homogeneous: photographs 2 and 3 show both light (the magnetic moments are close to direction 2) and dark (the moments are close to direction 3) noncollinear twins and the section of aligned magnetic moments (grey

![FIGURE 21](image-url)
FIGURE 22 Field-induced magnetic twins in GdIG near $T_{comp}$ at $H = 14.1$ kOe; the direction of $H$ is close to [111]; temperature is expressed in °K: (1) 277.47; (2) 277.98; (3) 278.28; (4) 278.54; (5) 278.67; (6) 279.03; (7) 279.51; (8) 279.72; (9) 280.11; (10) 280.37; (11) 280.56; (12) 280.69; (13) 280.85; (14) 281.03; (15) 281.98.
The canted phases are seen as narrow bands and wedges oriented along [111]. Similarly, the low-temperature canted structures are replaced by the high-temperature canted ones and then these by the high-temperature aligned structure. Figure 23 illustrates the appearance of a nonuniform magnetic structure in a magnetic field at a constant temperature. The magnetic field produces first a uniform magnetization of the sample (photograph 2) and then (photographs 3, 4), induces the canted structure, to cause the formation of magnetic twins and coexisting stable and metastable magnetic states.

The relative volume of coexisting phases and equivalent structures changes by both a smooth and jumplike motion of the boundaries. It should be mentioned that twins with magnetic moments in the sample plane are rarely encountered. The formation of such domains is not energetically favoured as a result of increasing magnetostrictive energy. The importance of the magnetoelastic interaction is shown by the high sensitivity of the domain structure to the stresses in the crystal. The domain boundaries observed between the equivalent structures are often distinct and of 1 μm in width. The elastically-stressed regions available in the crystal result in boundaries
Figure 24 Formation of a 6'-type magnetic twin from the boundary between twins of types 5' and 7'. $H = 14.1$ kOe the temperature varies uniformly from 280.21 to 280.37 K.

between certain twins sometimes forming an interlayer of variable thickness and these can act as nucleation centers for the new phase. Figure 24 shows the transformation of the domain boundary to the interlayer and then to a domain of the new phase. Dark areas in photographs 3 and 4 are domains in which the magnetic moments are in the sample plane.

The phase diagram plotted by visual observations is given in Figure 25. The figures by the curves correspond to types of appearing and disappearing domains. The dotted lines are the calculated phase boundaries of the canted structure. They are in good agreement with the experimentally observed phase boundaries. As for when $H \parallel [100]$, if the transition to the canted phase is of second order, the calculated boundaries (Figure 25) lay close to the experimental points corresponding to the appearance of magnetic twins.

The good fit of the phase boundaries to the experimental observation of twin appearance should be regarded as accidental, since the temperature, at which rather sharp boundaries between twins appear, is determined somewhat conditionally. Nevertheless, the formation of noncollinear twins near the stability boundaries of the aligned phase is quite natural. In a wide temperature range between the lines $A$ and $B$, $A'$ and $B'$, where according to the calculation only the aligned state should exist, the Faraday rotation reaches a value of more than 0.1 of the maximum value, indicating that sublattice rotation takes place here. In this temperature range the rotation of sublattices seems to be due to the existence of microscopic and lattice defects. It is
similar to the process whereby a magnetization curve approaches the saturation limit (Akulov, 1931; Brown, 1951). The approach of the $\phi(T)$ curve to zero with growing distance from $T_{\text{comp}}$ involves the formation of microscopic crystalline defects in the sample. In this respect the comprehensive studies of regularities of the tail of the $\phi(T)$ curve in the longitudinal and transverse geometries are of great interest. It should be noted that the transverse experimental geometry is sensitive to the deviation of the vectors from the direction of $H$, since here $\phi \sim \sin \theta$. As for the longitudinal geometry $\phi$ is proportional to $\cos \theta$.

5 MAGNETIC PHASE DIAGRAM OF A UNIAXIAL EPITAXIAL FERRIMAGNET

The previous sections presented experimental results on the magnetic field break up of the aligned structure and the behaviour of the canted structure for a cubic ferrimagnet. It is particularly interesting to consider phase diagram experiments on ferrimagnets with different anisotropies. Experiments with
axial ferrimagnets, the phase diagrams of which are already available for calculation purposes, are the most useful. Of the known ferrimagnets with an "easy-axis"-type of anisotropy, single-crystal films of multicomponent garnets with a magnetic compensation point, are of considerable interest. Their structural properties are quite perfect, the magnetic properties are well described with the two- or three-sublattice collinear model, and their high transparency allows visual checks to be made. However, because of its complex composition, the presence of non-uniform internal stresses can result in a variation of $T_{\text{comp}}$ with thickness and within the film plane. Furthermore, the magnetic anisotropy of the films may be more complicated than in a simple axial material.

This makes it difficult to use single-crystal epitaxial films as objects for studying the magnetic phase diagram of an axial Néel ferrimagnet, in particular, near its critical points.

Given below are the results of magnetooptical and visual studies of an epitaxial film, with composition near to $Y_{2.6}\text{Gd}_{0.4}\text{Fe}_{3.9}\text{Ga}_{1.1}\text{O}_{12}$ (Gnatychenko and Kharchenko, 1977). The film was grown from solution in the melt on a (111) GdGaG-substrate, by liquid-phase epitaxy. The film thickness was about 6 μm. The magnetic compensation temperature was measured magnetooptically in sections of about 80 μm. The $T_{\text{comp}}$ obtained are the averages over the film thickness. For the largest selected specimen (1.5 × 0.5 mm²) $T_{\text{comp}}$ varied from 182.0 to 183.5 K, the maximum gradient being about 1 K/mm. For the smallest sections studied (80 μm dia) the temperature variations did not exceed 0.1 K. It should be noted that variations of the compensation temperature along the film thickness were considerably larger, and according to studies of a film in the canted state, were about 13 K with an average gradient of $2 \times 10^3$ K/mm. Observations of the domain structure behaviour in a magnetic field far from $T_{\text{comp}}$ confirmed the existence of distinct uniaxial magnetic anisotropy.

We used a longitudinal experimental geometry, $\mathbf{K} \parallel \mathbf{H} \parallel E.A.$, and the measurements were carried out at $\lambda = 5730$ Å. Visual methods were used to find magnetic phases at weak fields.

A magnetically homogeneous state with interdomain boundaries perpendicular to the plane, is observed near $T_{\text{comp}}$, when field intensities are smaller than 7 kOe. The temperature range, within which the inhomogeneous state is realized, is about 12 K at a magnetic field of 2 kOe. Observations of the domain structure in a variable magnetic field show the typical changes of domain boundaries. In a range of field from 0 to a certain threshold value (the largest $H_{\text{th}}$ approaches 0.85 kOe and 0.7 kOe with increasing and decreasing the field, respectively), the domain boundaries are perpendicular to the film plane, and cut through it, their thickness being of several microns; they can only shift along the sample when the field changes. On increasing the field
FIGURE 26 Behaviour of domain boundaries in a film with increasing magnetic field strength: (a) crossed polarisers: (1) $H = 0.2$; (2) 0.6; (3) 0.8 kOe; (b) almost crossed polarizers: (1) $H = 0.3$; (2) 0.6; (3) 0.9; (4) 1.4 kOe; $T = 178.5$ K.

$H \gtrsim H_{th}$, the boundary “broadens” and then splits into two almost parallel boundaries between which a region of optical density different from that of the coexistent phase domain appears (Figure 26a). As the field increases still further, the distance between the boundaries increases and then there occur new “splittings” of the boundaries. A multistage structure in the optical density arises which indicates the development of domain boundaries parallel to the film plane (Figure 26b). The boundaries that are perpendicular to the film plane disappear in a field of about 7 kOe, and the sample becomes diffusively inhomogeneous. Smeared regions of different optical densities can be observed in higher fields.

The appearance and growth of the new phase with temperature change was always similar in the range 0 to 4 kOe. Domains of the new phase appeared in a jump-like manner and did not always cut through the film initially. Subsequent growth of domains was mainly due to the jump-like motion of the boundaries and to the development of new nuclei. The contrast between domains at a field strength up to 5 kOe was good enough to allow us to record the temperatures of the appearance and disappearance of magnetic phases. The results obtained with heating and cooling the sample are shown in Figure 27. The figure also shows the fields $H_{th}$ corresponding to the appearance (or the disappearance) of plane boundaries. The distances between pairs of points at a given field in Figure 27 denote the thermal hysteresis for
FIGURE 27 Diagram of magnetic states of the epitaxial film Y_{2.6}Gd_{0.4}Fe_{3.3}Ga_{1.1}O_{12} near the compensation temperature at weak magnetic fields. Points \( \bullet \) and \( \bigcirc \) are found from visual observations and correspond to: \( \bullet \)—the onset (from the left) and the completion (from the right) of the disappearance of the low-temperature phase upon heating, \( \bigcirc \)—the onset (from the right) and the completion (from the left) of the disappearance of the high-temperature phase with cooling. Rectangular points correspond to \( H_{\text{th}} \) with increasing (\( \square \)) and decreasing (\( \blacksquare \)) field. Points \( \bigcirc \) (\( H > 5 \text{ kOe} \)) are found from the \( \phi(T) \) relation. Solid curves—the calculated boundaries of the aligned phases.
the two sample sections most widely spaced in compensation temperature. Figure 27 shows that for these sections the fields at which hysteresis vanishes differ by a factor of two and approach $H_{c1} = 1.5$ kOe and $H_{c2} = 3.5$ kOe. Visual observations reveal that, at least at fields smaller than $H_{th}$, the sample may only be in two states according to the uniaxial model. The boundary “broadening,” observed in fields $H \geq H_{th}$, can be treated at first sight as the appearance of a canted phase due to a first-order spin-flip type of phase transition. But the fact, that a further increase in field results in some new boundary “splittings” and in the development of stepwise domain structure, indicates that in the field the plane compensation boundaries between aligned phases become energetically most advantageous. The apparent boundary broadening is associated with the development of the first step. The behaviour of the domain structure may be due to the change in $T_{comp}$ across the film thickness.

The existence of a large temperature gradient across the film thickness is indicated by the fact that the width of the temperature range within which the inhomogeneous state is observed, $\Delta T_{comp} \approx 12$ K (Figure 27), is much more than the maximum difference in effective compensation temperatures $\Delta T_{comp} \approx 2$ K, in different sample sections.

Besides the changes in $T_{comp}$, the magnetic anisotropy constant is also found to be variable across the film thickness. From Figure 27 we can see that the thermal hysteresis values for regions most distant from $T_c$ vary in different ways as the field increases. The phase diagram lines calculated by Eq. (3.32) of part I are fitted to experimental points at the critical fields $H_{c1} = 1.5$ kOe and $H_{c2} = 3.5$ kOe. It is natural to assume that the field $H_{c2}$ corresponds to the film layers closest to the substrate. In this case the magnetic compensation temperature decreases from 188.5 to 178.0 K as the distance from the substrate increases, and the energy of the uniaxial anisotropy, which is proportional to $H_{c2}$, reduces by more than four times.

Temperature dependences of the Faraday rotation were measured at a constant field $H$ in film sections of different sizes. Besides a small temperature difference in $T_{comp}$, the dependences scarcely differ from one another. A typical feature of the Faraday rotation in the aligned state region is its well pronounced linear dependence on temperature, deviations from which are observed only near boundaries of the canted region (Figure 28). The derivative $d\phi/dT$ for linear parts of $\phi(T)$ is inversely proportional to the field strength from 15 kOe upwards. In weaker fields $d\phi/dT$ is hardly affected by the inverse field strength. (Figure 28, inset). The distinct linear dependences confirm the validity of Eq. (3.33) of part I for the cosine of the rotation angle of ferrite sublattices in the uniaxial ferrimagnet with $(H_{c2}/H)^2 \ll 1$.

To describe the behaviour of Faraday rotation in the canted state regions in detail, we use as a model a film in which $T_{comp}$ remains constant in the film plane.
and changes across the thickness according to the linear law $T_{\text{comp}} = T_c + \delta T_{\text{c}}/l_3$. For simplicity the anisotropy constant is assumed to be constant. In an inhomogeneous film the sublattice rotation is inhomogeneous too. It is convenient to distinguish three fields which characterize the behaviour of the magnetic structure in inhomogeneous films: (a) the field $H_{\text{th}}$ at which the stability of a single collinear phase is lost if only in one layer; it reaches its maximum at $T = T_{\text{comp}}$; (2) the critical field $H_{\text{c}}(z)$ above which only a non-collinear state is stable at the temperature $T = T_c(z)$ for a layer which is at a distance $z$ from the substrate; when the anisotropy constant is variable, the critical field also changes across the film thickness; (3) the minimum field $H^*$ at which the stability of both collinear phases is lost for the whole film thickness.

Similarly, we can distinguish temperatures characterizing the film behaviour with temperature: $T'_1$ (and $T'_2$)-temperatures at which the canted
layer appears, and \( T'_1 \) (and \( T''_2 \))-temperatures at which the stability of a collinear phase is lost for the whole film thickness (Figure 27).

Various ranges of temperatures and magnetic fields correspond to different temperature dependences of the Faraday rotation. For fields \( H > H^* \), in the temperature range \( T'_1 \) to \( T''_2 \) where the film has the canted magnetic structure with sublattice rotation angle variable across the thickness, the Faraday rotation is linear in temperature, the slope of the linear portions being equal to the slope of \( \phi(T) \) for a homogeneous sample with \( T_{\text{comp}} = \bar{T}_{\text{comp}} \). Furthermore, it is independent of the distribution of \( T_{\text{comp}} \) across the film thickness.

In the temperature ranges \( T'_1 \) to \( T''_2 \) and \( \bar{T}_1 \) to \( \bar{T}_2 \) in which the aligned and canted states smoothly transformed from one to the other, the relation is nonlinear as given by:

\[
\phi(T) = \mp \phi_0 \left\{ \frac{1}{2} \left( \frac{A \delta T_c}{2} - 1 \right) \left( 1 - \frac{1}{A \delta T_c} \right) - \left( \frac{A}{2} + \frac{1}{A \delta T_c} \right) |T - T_c| \right. \\
+ \frac{A}{2 \delta T_c} (T - T_c)^2 \}
\]

where

\[
A = \left[ \frac{d}{dT} \frac{\phi}{\phi_0} \right]_{T_1 < T < T_2, H > H^*}
\]

For fields \( H_{\text{\parallel}} < H < H^* \) the rotation is a linear function, but is independent of the field strength:

\[
\phi = 2 \phi_0 \frac{T - T_c}{T_c}
\]  

(5.2)

Using the experimental value of the coefficient, \( A \), we can calculate the nonlinear portions of the \( \phi(T) \) curves. The calculated results indicate that a better fit to the experimental data is found at \( \delta T_c = 13 \) K. This value of \( \delta T_c \) is in agreement with the field \( H^* = 15 \) kOe, and is consistent with the behaviour of \( d\phi/dT \) as a function of \( 1/H \) (Figure 28).

In the range of small fields, \( H_c > H \geq H_{\text{th}} \) one of the aligned phases becomes unstable, and a plane compensation wall appears near \( H_{\text{th}} \). The field \( H_{\text{th}} \) should be correlated with the field at which apparent broadening of the domain boundary occurs. The calculated and experimental values for the maximum threshold field (0.5 and 0.8 kOe, respectively) agree within an order of magnitude. The temperature dependences of calculated and experimental \( H_{\text{th}}(T) \) are similar—both values decrease away from a certain temperature. The appearance of a stepwise domain structure for \( H > H_{\text{th}} \) is due to the
contribution of domain-wall energies to the thermodynamic potential and to the existence of a gradient in $T_{\text{comp}}$ in the film plane. For the model in question, domains should not be observed at fields $H > H_{c\parallel}$, but experimentally we failed to observe them only when the field was about 7 kOe. This seems to be affected by the inhomogeneous exchange energy, the effect of which, in a film with sublattice rotation angle varying from 0 to $\pi$ within a small distance $d(\approx 10^{-4} \text{ cm})$, increases in the vicinity of $H_{c\parallel}$ (where $d \to 0$). From this it follows that the above model of the film with compensation temperature linear across its thickness well describes the experimental results.

Recent magneto-optical studies on films with thickness reduced in stages by etching (Avaeva, Lisovskii, Mansvetova and Shapovalov, 1977) confirmed the conclusion about the strong change of the magnetic compensation temperature across the thickness of the epitaxial layer and showed that the dependence of $T_c$ on the distance to the substrate is of a stepwise type, the gradient of $T_c$ averaged over the steps being almost constant.

Since, in the model given, the slope of the linear part of $\phi(T)$ is independent of changes in $T_{\text{comp}}$ at $H > H^*$ and equal to the slope for the homogeneous film with $T_{\text{comp}} = \bar{T}_{\text{c}}$, the phase diagram of the homogeneous ferrimagnet can be constructed. The temperatures $T_1$ and $T_2$, at which the sublattice rotation begins and ends, are found by extrapolation of the linear parts of $\phi(T)$ to the values $\phi_0$ and $-\phi_0$. This is shown in Figure 29. Using the values of the slopes $(dH_r/dT) = 2.33 \text{ kOe K}^{-1}$ at $H > H_{c\parallel}$ and choosing values of $H_{c\parallel}$, we can obtain a better fit between the experimental results for $H < H_{c\parallel}$ and the metastability lines of the aligned phases (Figure 27).

Now let us try to estimate the slope of the phase diagram lines using Eq. (3.30) of part I. Unfortunately, the values of sublattice magnetizations and molecular constants are not known to high accuracy, and therefore only rough estimates of the slope can be made using the following molecular constants of GdIG: $\lambda_{12} = -80800$, $\lambda_{13} = -4250$, $\lambda_{23} = -1110 \text{ Oe} \cdot \text{g} \cdot \text{emu}^{-1}$. The magnetization of a rare-earth sublattice in the film is found from the data on gadolinium concentration. For $T = 0 \text{ K}$ the magnetic moment $M_{30}$ is equal to 16.6 $\text{emu/g}$. Knowing the compensation temperature $T_c$ and $M_{30}$, we obtain the effective exchange constant $\lambda = -3S_3kT_c/(S_3 + 1)\mu_3 M_{30} = -54000 \text{ Oe} \cdot \text{g/emu}$. The resultant moment of the Fe sublattices, $M_1 - M_2$, is dependent on the thermal history of the sample and may vary within a range of several $\text{emu/g}$ units. Taking account of the fact that after its growth the film is cooled rapidly ($10^2 \text{ K/min}$), and using the results from Czerlinsky (1969) and Visnovski, Prosser, Zvara and Polivka (1974), we can find the distribution of Ga$^{3+}$ ions over tetrahedral and octahedral sites and then calculate the value of $M_1 - M_2$. The calculations show that at a Ca$^{3+}$ ion concentration of $y = 1.10 \pm 0.05$ the resultant moment may be from 2 to 4.5 $\text{emu/g}$. The latter value is in closest agreement with experiment. In this
Reconstructed magnetic phase diagram for the uniform uniaxial film $Y_{2.6}Gd_{0.4}Fe_{2.9}Ga_{1.1}$ with $T_{\text{comp}} = 183$ K and $H_i = 2.5$ kOe. Dashed lines: (1) three-sublattice model, (2) two-sublattice model.

Using the experimental value of $(dH_i/dT)_{H \parallel H_{\|}}$ and the estimate $\lambda(M_1 - M_2) = 240$ kOe, we can find from the values of $H^{(1)}_i$ and $H^{(2)}_i$ (Figure 27) the limits of variation of the magnetic anisotropy constant, $K = (1/2)(\alpha/\beta)$.
PHASE TRANSITIONS IN NÉEL FERRIMAGNETS

(1/\lambda)H^2, in the film. The values found are \(K^{(1)} = -380\ \text{erg/cm}^3\) close to the substrate and \(K^{(2)} = -70\ \text{erg/cm}^3\) close to the free film surface. The average value of anisotropy constant is in satisfactory agreement with the data from the work of Moody, Shaw, Sandfort and Stermer (1973).

In conclusion, it should be mentioned that the gradients of \(T_c\) and the gradients of the anisotropy constant found in epitaxial films prevent detailed experimental studies of the phase diagram near the critical points, since the ranges of temperature and field intervals, where the spin-type first-order transitions in such films occur, are much smaller than variations of \(T_{\text{comp}}\) and \(H_{c||}\) in film sections of about \(10^{-4}\ \text{cm}\). Furthermore, the structural defects found impede such studies even in a massive single-crystal. Ferrimagnets with a high axial magnetic anisotropy constant may be more favourable for studies of this kind.

6 CHANGES IN PHYSICAL PROPERTIES AT FIELD-INDUCED TRANSITIONS OF THE NÉEL FERRIMAGNET TO THE CANTED STATE

A change in the magnetic symmetry with breakup of the ferrimagnetic collinear structure must affect its different physical properties. The behaviour of thermodynamic quantities at the transition can be treated using the mean-field approximation to the free energy and omitting the contribution from order-parameter fluctuations to the thermodynamic potential. The temperature range, for which such a treatment holds, can be estimated if the free energy of the system is added to the energy associated with non-uniform fluctuations. According to Volkov, Goranskii and Zvezdin (1970) this is given by (order of magnitude)

\[
\frac{\Delta T}{T_c} = \frac{k^2 b^2}{\delta a/\delta T \delta^3} \sim (0.1 - 1) \cdot \left( \frac{H_1}{H_E} \right)^3
\]

where \(k\) is the Boltzmann constant, \(a\) and \(b\) are coefficients of the \(\theta^2\) and \(\theta^4\) terms in the expansion of free energy in \(\theta\), and \(\delta\) and \(H_E\) are the constant and the exchange field interaction respectively between the Re and Fe sublattices. Unlike the phase transitions at the Curie point, orientational transitions in ReIG are much more pronounced, and the mean-field approximation must also be appropriate near the transition temperature. The reduction of the temperature range, in which the fluctuations have a pronounced effect, is due to the large fluctuation correlation radius for such transitions:

\[
r \approx \left( \frac{H_E}{H_1} \right)^{1/2} r_0
\]
FIGURE 30 Magnetization of some rare-earth ferrite-garnets near $T_{\text{comp}}$, as a function of magnetic field strength: (a) YbIG, $T = 4.2$ K (Clark and Callen, 1968); (b) HoIG, $T$: (1) 76.7; (2) 96.3; (3) 112.9 K (Ferronera et al., 1971); (c) DyIG, $T$: (1) 200 K, (2) 210 K (Levitine et al., 1970); (d) CdIG, $T = 255$ K (Fillon and Hug, 1970).

which is $(H_E/H_c)^{1/2}$ times more than the correlation radius for the magnetic transition at the Curie point. All the field-induced orientational transitions in ferrite-garnets can be divided into four groups: first-order transitions with no change in symmetry, liquid-vapour-like critical transitions and first- and second-order transitions with changes in symmetry. Changes in entropy, heat capacity, magnetic moment and magnetic susceptibility at these transitions have been considered in detail by Alben (1970) for the two-sublattice model of YbIG.
FIGURE 31  Temperature dependence of magnetization of YbIG at fixed magnetic strength. $H || [111]$: (a) theory (Alben, 1970b); (b) experiment (Ferron et al., 1973).
Using the approximate expressions for the free energy and the equation for \( \cos \theta \), the temperature and field-dependences of these quantities can be calculated. For some cases approximate analytical expressions can be obtained.

In the vicinity of the phase transitions anomalous behaviour of the properties is observed: rapid changes and discontinuities in the magnetization and entropy, sharp increases or jumps in the specific heat and the susceptibility, and in the magnetocaloric effect, Young's modulus, etc. Consider now some of them in detail.

The magnetic moment of ferrite-garnets was measured during their transition to the canted state in HoIG (Levitin et al., 1970, Ferron et al., 1971), GdIG (Fillon and Hug, 1970), DyIG (Levitin et al., 1970), YbIG (Clark and Callen, 1968, 1969; Clark and Alben, 1970; Ferron et al., 1972, 1973). Typical curves are shown in Figure 30. The kink in the \( M(H) \) curves made it possible to determine the field for transition to the canted state and to find the phase

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**FIGURE 32** Temperature dependences of magnetization of YbIG at fixed magnetic field strength, \( H \|[100] \): (a) theory (Alben, 1970b); (b) experiment (Ferron et al., 1973).
The experimental and calculated phase diagram of YbIG at $H\parallel[111]$ (Ferron et al., 1973 and Alben, 1970b).

Figures 31 and 32 show the experimental dependences of the garnet magnetization at $H\parallel[111]$ and $H\parallel[100]$ compared with the calculated ones from the works of Alben (1970a,b). Jumps in the magnetic moment at the transition are close to those calculated both in magnitude and position. The reconstructed phase diagram for $H\parallel[111]$ is given in Figure 33. The experimental lines are somewhat different from the calculated ones. Thus, the temperature $T^*_3$ approaches $16^\circ$K, according to the calculations, but its experimental value is about $28^\circ$K. Similar discrepancies are also observed for $H\parallel[100]$. The discrepancies seem to be due to the fact that the exchange parameters and the tensor components of the g-factor used in the calculations, are still not clearly understood. It should be mentioned that the effect of the Fe sublattice noncollinearity in YbIG is much smaller than that in GdIG, because of a lower compensation temperature. According to our estimates, the break up of the Fe sublattices may give a correction of no more than $30\%$ to the values of $T^*_3$.

The results of numerical calculation of the isentropes in the phase diagram of YbIG are shown in Figure 37. At points where first-order phase transitions occur the entropy changes discontinuously and a latent heat of transition is involved (absorbed). As the field rises, the heat of transition to a canted state for $H\parallel[111]$ increases, reaches a maximum at a field of about 80 kOe, decreases and then reduces to zero at a second-order transition point at $H = 197$ kOe, where $M_3 = 0$. Data on the entropy change in a magnetic
field can be obtained by magnetocaloric measurements. The variation of the crystal temperature with magnetization is as follows:

$$\Delta T = -\frac{T}{C_h} \left( \frac{\partial M}{\partial T} \right)_H \Delta H$$

where $C_h$ is the total heat capacity and $M$ — the resultant magnetic moment of the ferrimagnet. In the canted state the magnetic moment of the isotropic two-sublattice ferrimagnet is temperature-independent and $\Delta T = 0$. Taking into account the anisotropy causes a small change in the resultant magnetization with temperature and the appearance of the magnetocaloric effect in the canted state. The resultant moment of the canted phase in the three-sublattice
magnetic system is dependent on temperature in the isotropic case, too. However, in rare-earth ferrite garnets this dependence is weak, and in measurements of the integral magnetocaloric effect, after the applied field has reached the field where the transition to the canted phase takes place, \( \Delta T(H) \) should show only minor variation. Figure 35 shows the experimental data on the magnetocaloric effect in GdIG, HoIG (Belov et al., 1970) and YbIG (Clark and Alben, 1970) which give a good fit to theoretical results. Comparison between

**FIGURE 35** Adiabatic cooling of YbIG upon magnetization (Clark and Alben, 1970) (a) and magnetic field dependence of magnetocaloric effect in GdIG (b) and HoIG (c) (Belov et al., 1970) near \( T_{\text{comp}} \).
experimental and calculated isentropic curves is given in Figure 34 for YbIG. Values of the entropy jumps and latent heats of the transitions were found experimentally from heat capacity measurements for $H||[111]$ (Ferron et al., 1972). Figure 36 shows $C_H(T)$ for various values of the field strength. Presented here are also the results of the calculation of $C_H(T)$ for the same field orientation at $H = 100$ kOe. In accordance with the diagram the curve should display two singularities corresponding to two first-order phase transitions. Three distinct peaks were observed experimentally, one of which increased particularly fast with field strength. It may be that peaks in the heat capacity near the phase transitions, rather than its discontinuities are connected with the appearance of interphase boundaries and fluctuations in $\theta$ at phase transitions which are disregarded in the theory. The appearance of an additional peak 2 is possibly due to the orientation of the field not being strictly along the $[111]$ direction and to an increase of the number of phase transitions at this orientation. From these data the entropy jumps and the values of the latent heat of transition were found. They are equal to 2.5 and 6.1 joule/mole at $H = 17.25$ kOe for transitions 1 and 3, respectively. They increase with field and at $H = 31.6$ kOe reach the values 4.4 and 18.5 joule/mole.

Studies of heat capacity near the critical point with $H\parallel[100]$ are of particular interest. Figure 37 shows curves of $C_H(T)$ calculated in the work of Alben (1970a,b), with fields and temperatures close to critical ones. For $H||H_{c01}$ the heat capacity should vary with temperature as $(T - T_{comp})^{-2/3}$. Unfortunately, there are no comprehensive experimental studies in this region. In the works of Kamilov and Shakhsaev (1972) measurements on the heat capacity of GdIG at $H\parallel[100]$ at some values of the field strength are described. As one would expect, at high fields two peaks are observed in the heat capacity which are connected with second-order transitions. At lower fields the number of peaks increases. But in the region of the canted state there are observed two additional peaks instead of a single one. In this case the increase in the number of peaks may also be due to the errors in orientation of the field.

Other physical properties sensitive to the ferrimagnetic structure, such as the magnetostriction studied in the work of Levitin et al. (1970) and elastic properties, should be mentioned. In particular, ultrasonic attenuation should be sensitive to the magnetic twinning in the noncollinear state and to the magnetic phase splitting at the first-order transitions. In conclusion, it should be emphasized that in the experiments the crystal is required to be finely adjusted relative to the magnetic field and the sample should have no regions differing in $T_{comp}$. As the magneto-optical studies show, the latter are common, even in samples of small size, in studies of the orientational transitions and the canted state. Magneto-optical visual examination combined with investigation of other physical properties of ferrite-garnets is very useful.
FIGURE 36 Specific heat of YbIG with fixed magnetic field $H||[111]$ (Ferron et al., 1972). Solid lines in 1–3 correspond to $H = 0$, lines in 4 are calculated lines for $H = 100$ kOe (Alben, 1970b).
FIGURE 37 Specific heat of YbIG with fixed magnetic field $H \parallel [100]$, theory (Alben, 1970a).
7 CONCLUDING REMARKS

The aim of this paper has been to present results of theoretical and experimental investigations of field-induced phase-transitions in two- and three-sublattice Néel ferrimagnets. The behaviour of isotropic ferrimagnets has been treated in the framework of the molecular-field theory. The mean-field approximation was used to analyse the behaviour of anisotropic ferrimagnets. Particular emphasis has been placed on the vicinity of the magnetic compensation temperature, where influences of anisotropy are most significant. Whereas in the isotropic ferrimagnet the aligned and canted phases are separated by second-order transition curves, in anisotropic ferrimagnets the transitions from aligned to canted states can become symmetry-affected first-order transitions and new first-order transitions between the canted states can appear. Approximate analytical expressions which are valid near the compensation point have been given for phase boundary positions, typical magnetic fields and temperature ranges in the phase diagrams of two- and three-sublattice ferrimagnets. We have presented the most important results of the experimental investigations of field-induced transitions in the rare-earth iron garnets obtained through different techniques. Most attention has been given to the magneto-optical methods which allowed a visual check of the sample to be made. Most of the experiments were in satisfactory agreement with the phenomenological mean-field theory with allowance for the three-sublattice magnetic structures of rare-earth iron-garnets.

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