Magnetooptical investigation of the magnetization compensation point in gadolinium iron garnets

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The nature of rotation of the magnetization in cubic iron garnets near the magnetic compensation point is considered on the basis of energy considerations by taking anisotropy into account. The change in the polarization of light passing through a crystal during rotation of magnetization is considered on the basis of the Poincare-sphere technique. Rotation of the major axis of the polarization ellipse and the ellipticity of light passing through a gadolinium iron garnet crystal, $Gd_3Fe_5O_{12}$ ($T_k = 282\,^\circ K$) are studied experimentally at 250–310 °K and in a magnetic field up to 24 kOe oriented along the difficult magnetization axis [100] and easy magnetization axis [111]. The phase diagram in the (H, T) plane is found to be in qualitative agreement with the theoretical curve. It is shown that the existence of domains must be assumed to explain the rotation in the noncollinear region.

1. MOTION OF THE MAGNETIZATION OF A CUBIC FERRIMAGNET IN AN EXTERNAL MAGNETIC FIELD

Tyablikov[1] studied the magnetization of a ferrimagnet (a ferrite) and was the first to demonstrate that there is a range of magnetic fields in which the sublattice magnetizations form a noncollinear (canted) configuration. The theory of magnetization of ferrimagnets has subsequently been developed by several workers.[2–5] It has been found that canted configurations of ferrimagnets may be stable in a wide range of magnetic fields and temperatures. In particular, there is a range of temperatures ΔT near the magnetic compensation point T_c in which a canted phase can exist. This range increases with the applied field ($\Delta T \to 0$ as $H \to 0$). Transitions of the second kind take place at all the phase boundaries.

All the cited investigations have been concerned with an isotropic ferrimagnet. However, the anisotropy of the iron and rare-earth sublattices is important in rare-earth iron garnets.[6] In more recent papers[7,8] the phase diagram of ferrimagnets has been considered allowing for the cubic anisotropy. The main feature of the phase diagrams obtained by Zvezdin and Matveev[8] is a strong broadening of the range of temperatures in which the canted phase can exist in weak fields.

We shall now discuss the energy considerations which can be used to judge the nature of the motion of the magnetization. According to the theory of Clark and Callen,[4] the net magnetization of the canted phase in a crystal is directed along the field and constant in value. Therefore, the energy of such a crystal in an external field is independent of the orientations of the iron and rare-earth sublattice magnetizations. The energy is affected by this orientation if allowance is made for the anisotropy energy E_a. The expression for the anisotropy energy of a cubic crystal derived using the first anisotropy constant is

$$E_a = \sum_i K_{1i}(\alpha_{1i}^2\alpha_{2i}^2 + \alpha_{1i}^2\alpha_{3i}^2 + \alpha_{2i}^2\alpha_{3i}^2)M_i^2, \quad (1)$$

where α_{ji} are the direction cosines of the sublattice magnetizations M_i, measured from the crystallographic axes. Since the anisotropy constants of gadolinium iron garnet $Gd_3Fe_5O_{12}$ (GdIG) near the compensation temperature are close to the anisotropy constants of yttrium iron garnet $Y_3Fe_5O_{12}$ (YIG), we can ignore the anisotropy energy of the gadolinium sublattice and assume that the nature of the canting is governed by the anisotropy of the octahedral and tetrahedral iron sublattices.

Let us assume that an external field **H** is directed along a fourfold axis [100] of a cubic crystal (Fig. 1a). In the case of iron garnets the first anisotropy constant is $K_1 < 0$ and this direction is a difficult magnetization axis. The deviation of the net magnetization of the iron sublattice I_{Fe} from the direction of the field can be described by the polar angles θ and φ. An analysis of Eq. (1) shows that the rotation of M_{Fe} from the position 1 ($T < T_c$) to the position 4 ($T > T_c$) should take place in the (011) and (0$\bar{1}$1) planes. This is due to the fact that the maxima of the anisotropy energy are directed along the [010] and [001] axes, whereas the energy minima are directed along the [111] axes lying in the (011) and (0$\bar{1}$1) planes.

We shall now consider the motion of the magnetization in the (1$\bar{1}$0) plane. Two minima of E_a for $M_{Fe} \parallel [111], [1\bar{1}\bar{1}]$ (positions 2 and 3 in Fig. 1a) are separated by an intermediate maximum at $\theta = \pm\pi/2$. An external magnetic field deviates the minima of the ferrimagnet energy away from the [111] axes. In weak fields the minima are now located close to the [111] directions but are separated by an intermediate anisotropy energy maximum so that the transition from the position 2 to the position 3 is discontinuous. In stronger external fields the energy minima are shifted and the angle through which the magnetization jumps suddenly in the $2 \to 3$ transition decreases so that in fields higher than the critical value H' the rotation of the magnetization occurs without a discontinuity.

The nature of the canting rotation should be somewhat different for $H \parallel [111]$ (Fig. 1b). An analysis of Eq. (1) shows that the magnetization may deviate from the field direction in three symmetry planes ($\varphi = 0$, $2\pi/3$, $4\pi/3$) but on further rotation in the range $\theta > \pi/2$ the positions in other symmetry planes ($\varphi = \pi/3$, π, $5\pi/3$) become stable. It is clear from Fig. 1b that three jumps of the magnetization are possible during its rotation: $[111] \to [11\bar{1}] \to [1\bar{1}\bar{1}] \to [\bar{1}\bar{1}\bar{1}]$ (they correspond to the transitions $1 \to 2 \to 3 \to 4$).

Several methods have been suggested for the deter-

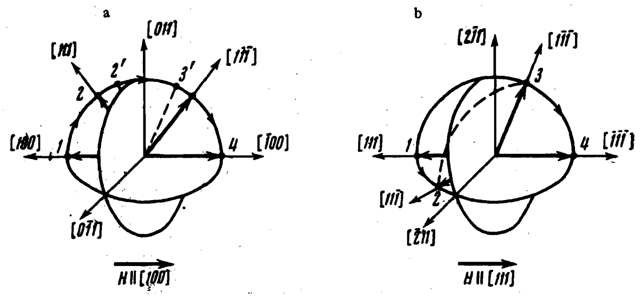

FIG. 1. Motion of the net magnetization of the iron sublattice in the region of the compensation point T_C. a—$H \parallel [100]$: $1(T < T_C)$ and $4 (T > T_C)$ are the orientations of the magnetization far from T_C in the canted phases; $2 \rightarrow 3$ is the magnetization jump in weak fields; $2' \rightarrow 3'$ is the jump in stronger fields. b—$H \parallel [111]$: $1 \rightarrow 2$ represents the first jump ($\Delta\theta \approx 70°$), $2 \rightarrow 3$ is the second jump ($\Delta\theta \approx 40°$), $3 \rightarrow 4$ is the third jump ($\Delta\theta \approx 70°$).

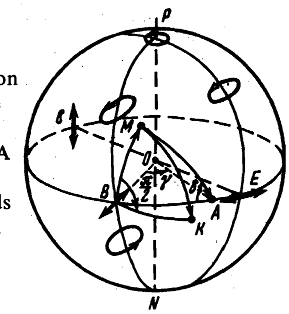

FIG. 2. Poincaré sphere. The transition from B to M represents the change in the state of polarization of light transmitted by a plate which is represented by the OA axis and which gives rise to a phase shift δ; the transition from M to K corresponds to a similar change in the polarization on passage through a quarter-wave plate.

mination of the critical field of the transition of a ferrimagnet to the canted configuration: they include measurements of the magnetization,[4,9,10] magnetostriction,[10,11] magnetocaloric effect,[12] absorption spectra,[13] Faraday effect,[5,14,15] and magnetic birefringence (Cotton-Mouton effect).[16] The results of experimental investigations confirm generally the theoretical predictions, i.e., it has been found that the canted phase does exist, that the range of temperatures of its existence near T_C increases with the external magnetic field, and the net magnetic moment of the canted phase is independent of temperature. However, no thorough experimental investigations have yet been made of the phase diagrams of ferrimagnets in weak fields in which the magnetic anisotropy exerts a strong influence. Moreover, the process occurring in the canted phase due to variation of the field, temperature, and other external factors have not yet been investigated at all.

The magnetooptic methods have several advantages in investigations of the sublattice configurations of rare-earth iron garnets. For example, in studies of the magnetization we cannot say how the sublattices are reoriented in the canted phase (in which the projection of the moment along the field direction remains constant), whereas the magnetooptic effects depend only on one of the sublattices of a ferrimagnet and this makes it possible to determine not only the limits of existence of the canted phase but also to obtain information on the nature of the canting of the sublattices in this phase. In the particular case of GdIG near the compensation point the magnitude of the Cotton-Mouton effect is governed only by the iron ion sublattices.[17]

2. MEASUREMENT OF THE CANTING OF THE MAGNETIZATION OF A FERRIMAGNET BY A MAGNETOOPTIC METHOD

For an arbitrary orientation of the magnetization of a ferrimagnet with respect to the direction of propagation of light the polarization of the latter changes because of the rotation of the plane of polarization and because of the magnetic birefringence. These changes in the polarization of light can be described conveniently by the Poincaré sphere method,[18] according to which each polarization state can be represented by a point on a sphere and the action of a medium on light, by the rotation of this sphere (Fig. 2). The poles of the sphere correspond to the light with the right- and left-hand circular polarization; the points on the equator represent the linearly polarized light and the other points the light with the elliptic polarization.

When light travels at right-angles to an external magnetic field the canting occurs in such a way that the projection of the magnetization on the direction of propagation of light vanishes and, consequently, the Faraday effect is not observed. Therefore, the canting of the magnetization must be studied using the magnetic birefringence (Cotton-Mouton effect). Measurements can be carried out using a quarter-wave plate located near a crystal.

As shown earlier,[17] when the magnetization is oriented along fourfold and threefold axes of a magnetooptically isotropic crystal the magnetic birefringence converts an optically isotropic crystal to one which is optically uniaxial and whose optic axis is directed along the magnetization. In all other cases the crystal is optically biaxial. However, the magnetooptic anisotropy of GdIG is not very strong and we shall assume that the optic axis coincides with the magnetization.

If the magnetization is parallel to the magnetic field, i.e., if $\theta = 0$, and the light incident on the investigated crystal is polarized at an angle of 45° with respect to the external field (point B on the sphere in Fig. 2), the magnetic birefringence can be described by a rotation of the Poincaré sphere about the radius OE. The state of polarization of the transmitted light is then represented by points on the meridian BP. A quarter-wave plate with its axis parallel to the direction of polarization of the incident light transforms the elliptically polarized light emerging from a crystal to the linear polarization, i.e., it transfers the points on the meridian BP to the equator BE. The magnetic birefringence can be determined by measuring the direction of polarization of light with an analyzer. However, since the magnetization (and, consequently, the optic axis of the crystal) deviate from the field direction and since $\theta \neq 0$, the action of the crystal can be described by a rotation of the sphere about the radius OA ($\gamma = \pi/2 - 2\theta$) through an angle δ, i.e., the point B is transferred to M. The quarter-wave plate transfers the point M to K, i.e., the light becomes elliptically polarized. In this case, we can use an analyzer to measure the orientation of the major axis of the ellipse as well as the ellipticity, i.e., the ratio of the lowest and highest intensities of the light transmitted by the analyzer.

Thus, the deviation of the magnetization from the field direction can be represented by a change in the azimuth of the ellipse or by a change in the ellipticity. It should be noted that when the magnetization is oriented at angles $\theta = 0, \pm\pi/4, \pm\pi/2, \pm 3\pi/4$, and $\pm\pi$ with respect to the field the change in the azimuth corresponds to the magnetic birefringence (Cotton-Mouton effect) whereas for intermediate angles the measured azimuth differs somewhat from the true birefringence.

The apparatus for the measurement of the rotation of the major axis of the optical ellipse β and of the ellipticity Δ consisted of the following elements: a source of light emitting at $\lambda = 1.15\ \mu$, a polarizer, a crystal, a quarter-wave plate, an analyzer, and a radiation detector. The experimental values of β were obtained in units of the phase difference per unit length for light polarized parallel and perpendicular to the external magnetic field (deg/cm) and the ellipticity Δ was expressed in percent per unit length of the sample.

A sample of GdIG ($T_C = 282^\circ$K) was bonded to a copper heat sink in a cryostat in which the temperature was monitored with a copper-constantan thermocouple. The measurements were carried out during continuous variation of the temperature at a rate ranging from 2×10^{-4} to 1×10^{-2} deg K/sec. The measurements were carried out in the temperature range 250–310°K. Two series of measurements were carried out in a magnetic field up to 24 kOe directed along the difficult [100] and easy [111] magnetization axes. In these experiments light traveled along the [110] axis. A second sample of GdIG ($T_C = 290^\circ$C) was used in a study of the Faraday effect in the 240–310°K range in fields up to 16 kOe. In this case, light traveled along the magnetic field which was parallel to the [111] axis.

3. EXPERIMENTAL RESULTS

Some of the temperature dependences of β and Δ obtained in a field directed along the [100] axis are plotted in Fig. 3. We shall now consider the most important features of these dependences. In all magnetic fields far from T_C the birefringence varies little with temperature except near the compensation point, where it varies very rapidly and passes through a minimum at this point. In weak magnetic fields the value of β far from T_C increases with increasing field and reaches saturation in H = 0.8 kOe. This is due to the magnetization of the investigated crystal. The minimum of β in weak fields is reached by a discontinuous change, whereas in strong fields the change is continuous. Between 0.1 and 0.8 kOe the discontinuity and the smooth minimum coexist, the magnitude of the discontinuity decreasing and the depth of the smooth minimum increasing with increasing field. Consequently, the net depth of the minimum decreases up to 0.8 kOe and then the minimum becomes symmetric and its depth increases with the field (passing through zero in ~2 kOe) right up to 15 kOe. Moreover, the negative azimuth β at the minimum remains equal to approximately half the value of the same azimuth far from T_C; when the field is increased the minimum becomes more pronounced. Random jumps of β, which disappear in stronger fields, are observed in very weak fields of the order of several tens of oersteds. Steps of β occur in stronger fields.

The changes in the ellipticity Δ begin at the same temperatures as the changes in β. In weak fields there is a broad maximum of Δ whose magnitude increases in fields up to about 0.8 kOe reaching 6–7%; in stronger fields the amplitude decreases with the field. Beginning from fields of 4 kOe, the maximum of Δ splits into two or more peaks which diverge with increasing field. Control experiments have indicated that the amplitude Δ may vary slightly from one experiment to another but the positions of the maxima remain fixed along the

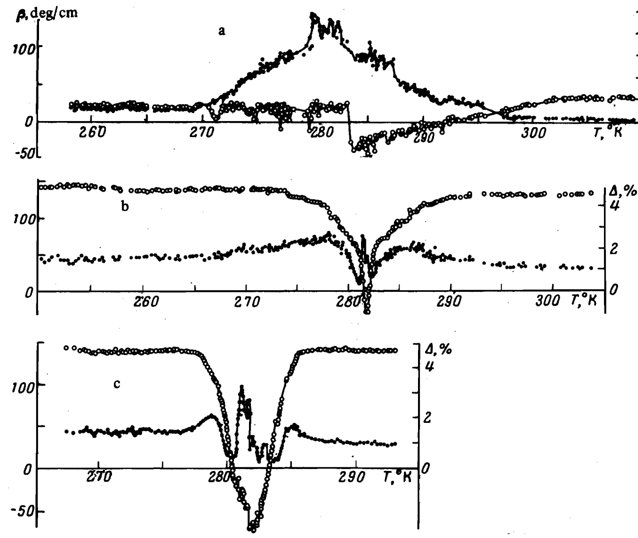

FIG. 3. Temperature dependences of the azimuth β of the major axis of the polarization ellipse (O) and of the ellipticity Δ (●) of the light transmitted by a crystal 0.26 cm thick subjected to a field **H** ∥ [100]: a–H = 0.043 kOe; b–H = 4.0 kOe; c–H = 21.4 kOe.

temperature scale to within 0.1 deg K. In fields above 4 kOe there are two minima of Δ in the region where β passes through zero and a third minimum of Δ in the region of minimum of β.

If the field is oriented along the [111] axis (Fig. 4) the temperature dependences of β and Δ are generally similar to those in the case H ∥ [100], but there are several differences. Far from T_C the birefringence is approximately 1.2 times stronger than in the preceding case. A reversal of the sign of β is observed in fields of ~7 kOe. In fields up to 2.4 kOe several maxima of the ellipticity Δ are observed; in the range 2.4 kOe $< H <$ 10 kOe one weak maximum ($\Delta < 1\%$) is observed but in stronger fields all the three or four maxima of Δ can be seen and these maxima grow up to 3% in the strongest field employed.

The temperature dependences of the Faraday effect in H ∥ [111] obtained for the second sample of GdIG are plotted in Fig. 5. In this case, the ellipticity ranges up to 10% and there is only one maximum of Δ.

4. DISCUSSION OF RESULTS

The large changes in β and Δ in the region of T_C are evidence of deviation (canting) of the magnetizations of the iron and gadolinium sublattices from the magnetic field. The onset of the canting can be deduced from the beginning of the changes in β and Δ. The points of inflection of the temperature dependences obtained for H ∥ [100] can be used to plot a phase diagram in the (H, T) plane with the boundaries set by the transitions to the canted phase (Fig. 6). It is clear from Fig. 6 that in weak fields the range of existence of the canted phase ΔT broadens considerably and is in qualitative agreement with the theory of Zvezdin and Matveev.[8] The smallest value of $\Delta T \approx 9$ deg K corresponds to 20–22 kOe; in stronger fields this range becomes wider. Measurements of the Faraday effect in the second sample of GdIG have demonstrated the existence of a narrower range $\Delta T \sim 4$ deg K. The narrowness of this range depends strongly on the number of defects in a crystal (the value of T_C for the second sample was considerably higher than for the first).

Earlier investigations of GdIG in the region of the compensation point T_C include those reported in[5] and and[15]. However, in the first of them[5] no broadening of the temperature range ΔT has been observed and the interpretation has been based on the model which ignores the anisotropy field. In the second case,[15] the measurements of the Faraday effect in fields up to 10 kOe have established that the narrowest range ΔT corresponds to 3–5 kOe. These discrepancies obviously require further experimental studies which would establish reliably the boundaries in the phase diagram of GdIG.

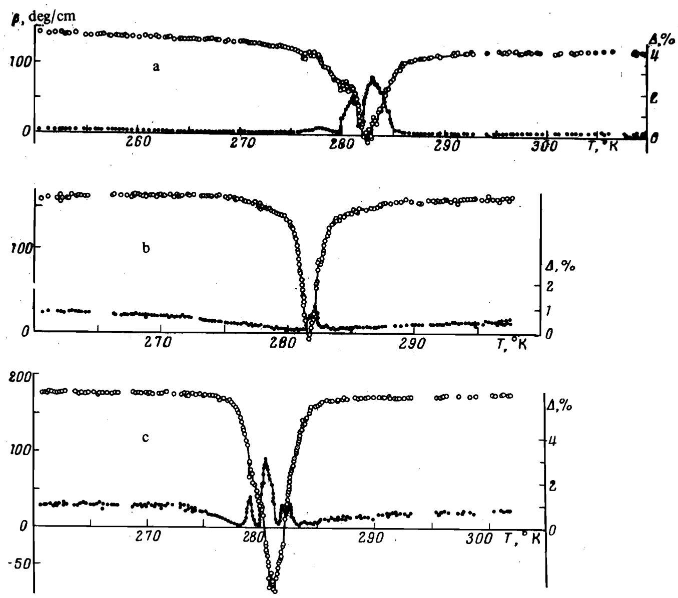

FIG. 4. Temperature dependences of β (O) and Δ (●) for H ∥ [111]: a–H = 0.25 kOe; b–H = 7.7 kOe; c–H = 23.7 kOe (crystal 0.26 cm thick).

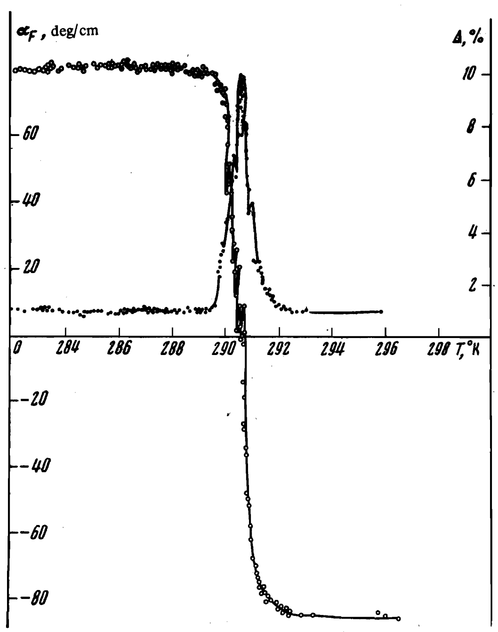

FIG. 5. Temperature dependences of the Faraday angle α_F and of the ellipticity Δ for H ∥ [111] = 2.6 kOe applied to a 0.49 cm thick sample of $Gd_3Fe_5O_{12}$.

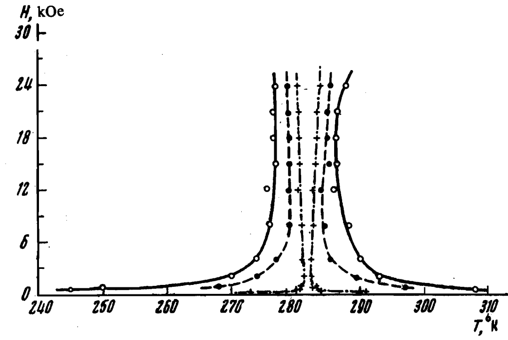

FIG. 6. Phase diagrams of GdIG in the (H, T) plane for H ∥ [100]. The curves are plotted on the basis of the kinks in the temperature dependences of the azimuth β (O), outer maxima of the ellipticity Δ (●), and ellipticity minima (+).

Let us now consider the processes which occur in the canted phase of GdIG. The nature of the curves obtained indicates that the canting is not a uniform process. The behavior of β and Δ expected on the basis of the Poincaré sphere method is different from that found experimentally. Thus, it is predicted that the value of Δ should decrease when the magnetization passes through the angles $\theta = \pm\pi/4, \pm\pi/2, \pm3\pi/4$ and between these directions it should rise. The experimental $\Delta(T)$ curves actually exhibit rises and falls of the effect but not as strong as those predicted theoretically. For the crystal under investigation the value of Δ at a maximum should be about 25%, which is considerably greater than the experimental values of 6–7%. Figure 6 also includes the points corresponding to the maxima and minima of the ellipticity of light. It is interesting to note that the points corresponding to the ellipticity minima are close to the points in the phase diagrams given in[5,15].

It follows from our study that the observed temperature dependences of β and Δ cannot be explained by the rotation of the magnetization of the crystal as a whole. On the basis of the considerations put forward in Sec. 1, we may suggest that at the boundary of the collinear and noncollinear (canted) phases a crystal splits into several regions with different orientations of the magnetization. Thus, in the H ∥ [100] case a crystal may split into four types of domain with deviations of the magnetization equally likely in the (011) and $(0\bar{1}1)$ planes, as shown in Fig. 1a. If we assume that these domains are small compared with the dimensions of the crystal, we can easily show that β should vanish not at $\theta = 45^\circ$ but at $\theta = \cos^{-1}3^{-1/2}$, when the magnetizations of the iron sublattices are oriented along the [111] easy axes. The experimentally determined β passes through zero in fields of 1–2 kOe. Consequently, in these fields the magnetization of the iron sublattice switches from one easy direction to the other. In stronger fields the depth of the minimum of β increases. This means that the discontinuity $\Delta\theta$ decreases. The minimum value of β is reached in fields of $H \approx 15$ kOe and is approximately equal to half the value of β far from T_C. Therefore, we may assume that in fields in excess of 15 kOe the magnetization rotates without a jump.

In the H ∥ [111] case, the onset of the fall of β is not so strongly marked. The width of the temperature range ΔT varies little with the field and some broadening is observed only in very weak fields. If we assume that a crystal splits into three types of domain (Fig. 1b) and the magnetization deviates in three jumps, as suggested by the energy considerations, the temperature dependence of β should be in the form of a rectangular well. This does not agree with the experimentally obtained dependences and we must therefore conclude that, in this case, the sublattice magnetizations rotate smoothly and the discontinuities are less marked than in the H ∥ [100] configuration.

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