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DICHROISM AND MAGNETIC ANISOTROPY OF ERBIUM IRON GARNET

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The effect of magnetic fields up to 25 kOe on the spectrum of light absorption in a single-crystal plate of erbium iron garnet is investigated in detail at 4.2°K. It is shown that exchange splitting of the bands representing the optical transition $^4I_{15/2} \rightarrow {}^4S_{3/2}$ is anisotropic. The dichroism of the absorption bands is studied. It is found that the components of this transition exhibit intensities which depend on the orientation of the light-wave electric vector **E** with respect to the magnetization direction $\mathbf{M}_S$ of the iron garnet. Polarization measurements therefore yield the angles between the $\mathbf{M}_S$ vectors and the external magnetic field **H** for various field strengths and orientations. These data are used to calculate the magnetocrystalline anisotropy constants of erbium iron garnet: $K_1 = 9 \times 10^6$ erg/cm^3 and $K_2 = 5 \times 10^7$ erg/cm^3.

PRELIMINARY optical studies[1] have shown that the fine structure of $^4I_{15/2} \rightarrow {}^4S_{3/2}$ absorption in erbium iron garnet (ErIG(results from the splitting of Er^{3+} Kramers doublets that is induced by the exchange field of neighboring iron ions. Earlier studies have indicated that the structure of the exchange splitting changes when an external magnetic field alters the orientation of the magnetic moment of ytterbium iron garnet[2] or europium iron garnet[3] with respect to the crystallographic axes. Consequently the exchange interaction between rare-earth ions and iron ions cannot be isotropic. The anisotropy of the interaction depends on the orbital state of the rare-earth ion.[4,5]

In the present work we investigate details of the $^4I_{15/2} \rightarrow {}^4S_{3/2}$ fine-structure anisotropy of erbium iron garnet in magnetic fields ranging from one to 25 kOe at T = 4.2°K. Since the polarization exhibited by the individual optical transition components is associated with the orientation of the iron garnet magnetization vector $\mathbf{M}_S$,[6] our investigation of absorption dichroism enabled us to determine the direction of the magnetic moment for different external magnetic field directions. Measurements of the angle between $\mathbf{M}_S$ and the easy magnetization direction [100][7] yielded the crystal anisotropy constants of erbium iron garnet.

TECHNIQUE

Figure 1 shows the experimental geometry. The sample K was a single-crystal plate 60 μ thick with an area of about 20 mm^2, mounted between two lenses L_1 and L_2. The developed plate surface was parallel to the (110) plane. A special sample holder in the optical cryostat[8] enabled rotation of the crystal in its own plane to vary the angle φ between [100] and the external field **H**. An SII-47 electromagnet was used to apply fields up to 25 kOe. An Ahrens polarizer P was used; θ is the angle between the electric vector **E** of linearly polarized light and the [100] direction. The light source S was a 170-watt incandescent lamp. The absorption spectrum was photographed with a DFS-8 diffraction spectrograph having 6 Å/mm dispersion; Sp is the spectrograph slit.

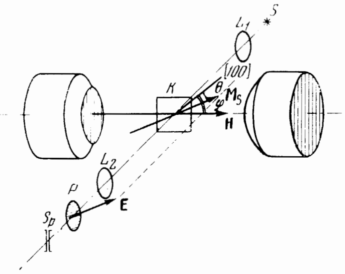

FIG. 1. Scheme of experimental apparatus.

EXPERIMENTAL RESULTS AND DISCUSSION

A. Characteristics of the Zeeman effect. Figure 2 shows light absorption spectra of erbium iron garnet for the $^4I_{15/2} \rightarrow {}^4S_{3/2}$ transition, showing differences depending on the orientation of the sample with respect to the external magnetic field **H**. The first spectra, obtained with **H** parallel to the easy magnetization direction [100], were completely identical with those obtained in the absence of an external field.[1] The sample was then rotated through successive intervals Δφ = 5° about

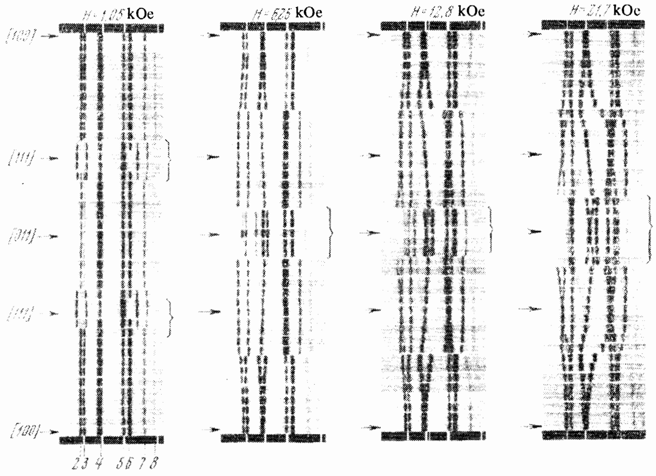

FIG. 2. Absorption spectra of $^4I_{15/2} \rightarrow {}^4S_{3/2}$ transition in erbium iron garnet for different orientations of the external magnetic fields **H** = 1.05, 6.25, 12.8, and 21.7 kOe, at 4.2°K.

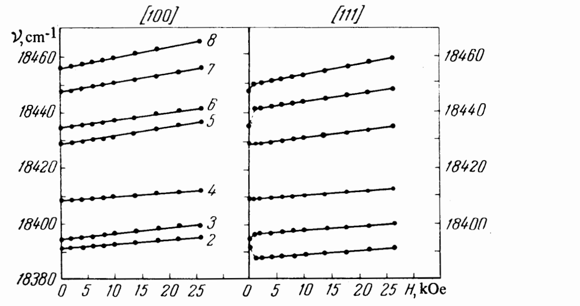

FIG. 3. Absorption band frequencies for the $^4I_{15/2} \to {}^4S_{3/2}$ transition in ErIG vs. external magnetic field strength, for **H** ∥ [100] and **H** ∥ [111], at 4.2°K.

the direction of the light beam (Fig. 1). The spectrum was photographed for each value of φ, which was thus varied from 0° to 180° with a fixed magnetic field strength. The anisotropy of the splitting was studied in greater detail, at intervals $\Delta\varphi = 2^\circ - 3^\circ$, in the spectra denoted by curly brackets in Fig. 2.

We shall now discuss certain characteristics of the Zeeman effect. The simplest spectra are recorded when the external field H is parallel to either the [100] or the [111] direction. We shall consider these two cases in greater detail.$^{1)}$

If the exchange-splitting structure depends on the direction of M_S in an iron garnet,[2] the marked difference between the two spectra recorded with H = 1.05 kOe (Fig. 2) is evidence of an abrupt change in the direction of M_S. Enhanced field strength when the field direction remains fixed along either [100] or [111] induces only a slight frequency shift of the absorption bands (Fig. 3) without altering the structure of the $^4I_{15/2} \to {}^4S_{3/2}$ transition. This result indicates saturation at 1.05 kOe for M_S parallel to [100] or [111] in a thin plate of ErIG.

With intermediate directions of the external magnetic field the spectrum is complicated by doublet splitting of bands 4 and 6. The behavior of the separation $\Delta\nu$ between the doublet components of band 4 (Fig. 2, at H = 21.7 kOe) is noteworthy. There is zero splitting when the field is parallel to [100]; the separation increases smoothly with the angle φ up to a maximum $\Delta\nu \sim 20$ cm^{-1} when H is along [111]. Still further rotation up to $\varphi = 80^\circ$ does not affect $\Delta\nu$. It is a reasonable hypothesis that the strong dependence on H (with a fixed direction) which is exhibited by the splitting of band 4 is associated with the fact that a field considerably exceeding 1.05 kOe is required to saturate M_S along each arbitrary direction in the plane of the sample. An external field H that is not along [100] and is also strong enough to surmount the anisotropy energy of a garnet will "entrain" M_S; however, the latter will lag behind H, forming an angle θ with the [100] direction (Fig. 1). A steep increase of $\Delta\nu$ for certain directions of a magnetic field exceeding 1.05 kOe (Fig. 2) indicates an abrupt change of θ. We shall show in the following section that this hypothesis can be verified and that it is possible to determine θ through a detailed study of dichroism in absorption.

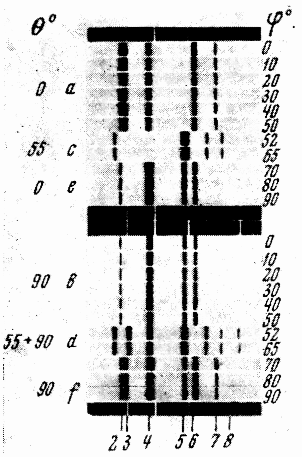

FIG. 4. Absorption spectrum of ErIG in polarized light. The angle θ between the vector E of linearly polarized light and the [100] direction corresponds to maximum intensity of the polarized absorption bands. $0^\circ \leqslant \varphi \leqslant 90^\circ$; H = 1.05 kOe; T = 4.2°K.

B. Dichroism of absorption. The relationship between the polarization of absorption bands and the orientation of the magnetization vector in an iron garnet was first noted in[6]. This investigation using polarized light showed that each direction of H corresponds to two mutually perpendicular directions, in the plane of an ErIG plate, for the electric vector E of linearly polarized light when the polarized bands of the $^4I_{15/2} \to {}^4S_{3/2}$ transition are of extremal intensity. For fixed directions of H = 1.05 and 21.7 kOe the angle θ between E and the [100] direction was varied at 5° intervals in the range 0°–180°. We selected the spectra with optimum polarization among the large number that were recorded. Figures 4 and 5 show these spectra and the corresponding directions of E; the field was 1.05 kOe in Fig. 4 and 21.7 kOe in Fig. 5.

The foregoing results do not conflict with the hypothesis, based on the analysis of the spectrum in Fig. 2, that the magnetization vector can lie along either [100] or [111] when H = 1.05 kOe. An abrupt change of the spectrum is accompanied by an alteration of polarization properties for the Er^{3+} bands; there is an abrupt change in the angle θ representing the direction of **E** at which the bands exhibit maximum intensity (Fig. 4).

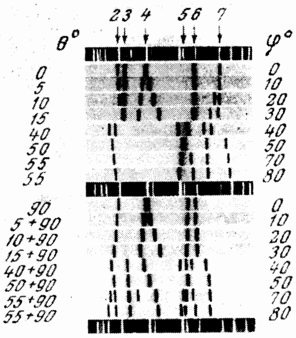

FIG. 5. Absorption spectrum of ErIG in polarized light. The angle θ between the vector E of linearly polarized light and the [100] direction corresponds to maximum intensity of the polarized absorption bands. $0^\circ \leqslant \varphi \leqslant 80^\circ$; H = 21.7 kOe; T = 4.2°K.

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1)The complex spectrum observed when **H** is parallel to [110] and its steep dependence on the field strength will be discussed in a separate article.

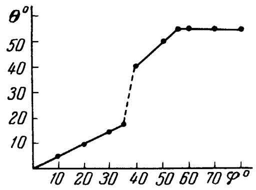

FIG. 6. Dependence of the angle θ between M_S and [100] on the angle φ between H and [100] at H = 21.7 kOe and T = 4.2°K.

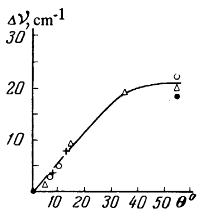

FIG. 7. Dependence of Zeeman splitting of band 4 on the angle θ between M_S and [100], for H = 1.05 kOe (●), 6.25 kOe (○), 12.8 kOe (+), 21.7 kOe (Δ), at T = 4.2°K.

Analogous measurements with H = 21.7 kOe (Fig. 5) enabled us to determine the value of θ between M_S and the [100] direction when H formed angles $\varphi = 0°-80°$ with this axis (Fig. 6).

The results shown in Figs. 2 and 6 enable us to determine the orientation of M_S in a thin ErIG plate for any direction of H. In the angular interval 0°—35° between **H** and [100] the moment M_S follows the field but is not parallel to the latter (Fig. 6). The separation $\Delta\nu$ between components of band 4 also increases uniformly. The field dependence of $\Delta\nu$ for fixed φ (Fig. 2) indicates that the angle θ between M_S and [100] increases with the field H. Still closer approach of H to [111] (with $\varphi > 35°$, and H = 6.25 or 12.8 kOe) induces shifting of M_S to the [111] direction in advance of the field. The maximum of $\Delta\nu$ is reached at $\varphi = 40°$. When the angle between H = 21.7 kOe and [100] exceeds 35°, M_S abruptly becomes parallel to the field ($\varphi = 40°$), and thereafter follows the field until the latter coincides with [111] ($\varphi = 55°$ in Figs. 5 and 6). Figure 2 indicates an abrupt change of $\Delta\nu$ in this angular region ($40° \leq \varphi \leq 55°$). At still higher field angles up to 80°, M_S remains parallel to [111].

The dependence of $\Delta\nu$ on θ can be plotted after determining the angle θ between M_S and [100] for H = 6.25 and 12.8 kOe. In Fig. 7 we observe a unique correspondence that does not depend on the external magnetic field. The separation of the Zeeman components of band 4 therefore depends on the relative orientation of M_S and the [100] axis.

C. Evaluation of the magnetocrystalline anisotropy constants. The investigation of Zeeman effect anisotropy in conjunction with the study of dichroism has enabled us to determine the equilibrium angle between M_S and [100] for each external magnetic field direction in the plane of a sample (Figs. 5 and 6). This angle is derived from the minimum free energy of the crystal. We write this energy as the sum of the anisotropy energy and the energy in the external magnetic field, and limit ourselves to the anisotropy constants K_1 and K_2:[9]

$$W = W_a + W_H = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2 - (\mathbf{HI}_s). \qquad (1)$$

For the (100) plane (Fig. 1) we have $\alpha_1 = \cos\theta$, $\alpha_2 = \alpha_3 = 2^{-1/2}\sin\theta$. The measured experimental equilibrium angle θ (Figs. 5 and 6) is obtained from the equation $\partial W/\partial\theta = 0$. Using the results shown in Figs. 5 and 6 for $0 < \varphi < 35°$, available data regarding the saturation magnetization $I_S = 20\ \mu_B$/molecule,[7] and the unit cubic cell parameter of the garnet (a = 12 Å), for H = 21.7 kOe we obtain $K_1 = 9 \times 10^6$ erg/cm³ and $K_2 = 5 \times 10^7$ erg/cm³. These values are consistent with the results obtained by Pearson,[10] who showed that at 4.2°K the first constant K_1 is positive and $K_1 \ll K_2$. It is confirmed, incidentally, that [100] is the easy direction of magnetization in erbium iron garnet.

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