Features of optical anisotropy of europium iron garnet

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A change in the nature of the anisotropy of the optical properties of a magnetically ordered crystal is observed experimentally upon variation of the relative directions of the magnetization vector **I** and of the light-wave vector **k**. It is shown that an $Eu_3Fe_5O_{12}$ crystal oriented along the [110] axis behaves at $\mathbf{k} \perp \mathbf{I}$ like an optically uniaxial crystal, whereas at $\mathbf{k} \parallel \mathbf{I}$ it resembles an optically biaxial crystal. This type of anisotropy cannot occur in the traditional crystal-optics theory based on expansion of the permittivity tensor ε in the direction cosines of the vector **I**. The feasibility of the aforementioned anomalous anisotropy in cubic crystals is demonstrated by expanding the linear-absorption coefficient $\varkappa$ in a series in the direction cosines of **I** and **k**.

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1. INTRODUCTION

The study of magnetic linear birefringence (MLB) of light, which includes also the effect of magnetic linear dichroism, in magnetically ordered crystals was started relatively recently both in the crystal transparency region[1] and in the region of the absorption bands of individual ions.[2] The interest on the part of the researchers was not accidental. It was due primarily to an unexpected result—the MLB, which depends quadratically on the magnetization, turned out to be anomalously large in magnetically ordered crystals and comparable with the magnetic circular birefringence, which is linear in the magnetization and is responsible for the Faraday effect. This uncovered a possibility of investigating the magnetic structures of magnetically ordered crystals and of practical application of MLB in devices such as light modulators, magneto-optical waveguides, and others. In addition, MLB is extensively used to study the electronic structure of magnetically ordered crystals, since the anisotropy of their optical and magneto-optical properties is determined by the action of the anisotropic exchange and crystal fields on the magnetoactive ions.

MLB reaches particularly large values in the region of the absorption bands of rare-earth ions in iron garnets (IG), has a strongly anisotropic character, and reveals a strong dependence on the wavelength of the incident light.[2,3] Interesting features of crystal-optics properties of magnetically ordered crystals were observed,[3] connected with the absence of a small parameter in the expansion of the dielectric tensor ε in powers of the magnetization.

We continued our study of the crystal-optics properties of europium IG in the region of the absorption band of the rare-earth ion. Principal attention was paid to observation of those crystal-optics anisotropy features which are connected with the change of the relative orientation of the magnetization vector I and the light wave vector. Investigation of the crystal-optics properties of europium IG in the region of the $^7F_0 \rightarrow {}^7F_6$ absorption band of the rare earth ion Eu^{3+} has shown[3] that when the light propagates perpendicular to the magnetization (Voight geometry, $\mathbf{k} \perp \mathbf{I}$) the crystal is optically uniaxial, with the absorption coefficient $\varkappa$ having the same value[1)] at $\mathbf{I} \parallel [110]$, $\mathbf{k} \parallel [110]$, $\mathbf{e} \parallel [001]$ and $\mathbf{I} \parallel [110]$, $\mathbf{k} \parallel [001]$, $\mathbf{e} \parallel [110]$ (here **e** is the vector of the electric component of the light-wave field, $\varkappa$ is the absorption coefficient, the imaginary part of the complex refractive index $N = n - i\varkappa$).

It will be shown below that when the linear dichroism is measured in the same sample and at the same orientation of the vector I, but in the Faraday geometry ($\mathbf{k} \parallel \mathbf{I}$), the crystal behaves like an optically biaxial one. The results attest to the inadequacy of the traditional description of the optical properties of a magnetically ordered crystal with the aid of a tensor ε whose components depend only on the light frequency and on the direction cosines of the magnetization vector.

EXPERIMENTAL RESULTS

The measurements were made on single-crystal samples in the form of plates 100 μm thick. The light incident on the sample was linearly polarized. The relative arrangements of the vectors **k**, **e**, and **I** for the Voigt geometry and for the Faraday geometry are shown in Figs. 1a and 1b. The absorption spectra were recorded at the temperatures $T = 82$ and 110 K in a field $H = 22$ kOe, using the magneto-optical apparatus described in Refs. 3 and 4. The dependence of the intensity $J(\omega)$ of the light transmitted through the sample, as well as of the intensity $J_0(\omega)$ of the light without the sample, was registered with an automatic recorder. The absorption coefficient $\varkappa$ was calculated from the equation

$$J = J_0(1 - R^2)e^{-2\alpha d}, \quad \alpha = 2\pi\varkappa/\lambda,$$

R is the reflection coefficient calculated under the assumption that the refractive index of the garnet is $n = 2.2$

Figure 2 shows the measured refractive index $\varkappa(\hbar\omega)$, in the Faraday geometry, of an $Eu_3Fe_5O_{12}$ plate cut in the (110) plane, at $\mathbf{e} \parallel [\bar{1}10]$ and $\mathbf{e} \parallel [001]$, while Fig. 3 shows the $n'(\hbar\omega)$ spectra calculated from these results with the aid of the Kramers-Kronig relations [n' is the contribution made to the refractive index by the light absorption by the rare earth (RE) ions]. A difference between $\varkappa(\hbar\omega)$ and $n(\hbar\omega)$ at two **e** orientations is evidence that the crystal is optically biaxial. This dif-

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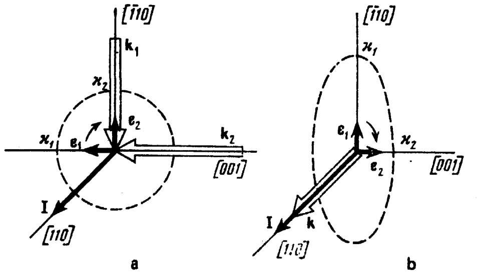

FIG. 1. Vector diagrams for the determination of the optical anisotropy of a crystal in the Voigt (a) and Faraday (b) geometry.

ference in $\varkappa$ is a maximum at the frequencies 4980 and 5100 cm^{-1} where it amounts to 1.2×10^{-3} and 1.5×10^{-3}, respectively, i.e., it reaches 60–70%. As seen from Fig. 3, $\Delta n = n'_{100} - n'_{110}$ reaches a value 1×10^{-3} and reverses sign several times in the region of the absorption band.

The character of the optical anisotropy of the crystal depends thus on the relative orientation of the vectors **k** and **I**. To our knowledge, no such effect was observed before.

We note that if the same $Eu_3Fe_5O_{12}$ crystal is magnetized along the [100] axis it behaves like a uniaxial one in both the Voigt geometry (Fig. 7 of Ref. 3) and in the Faraday geometry. The spectra obtained by us in the Faraday geometry for **k** ∥ [100], **I** ∥ [100], **e** ∥ [001] and **k** ∥ [100], **I** ∥ [100], **e** ∥ [010] are of the same form as the spectra of Fig. 7 of Ref. 3. Figure 4 shows the dependence of the refractive index of the light on the frequency in the Faraday geometry for a plate cut in the (111) plane at a magnetization-vector orientation along the [111] axis for **e** ∥ $[\bar{1}10]$ and **e** ∥ $[1\bar{1}0]$. The frequency dependences of $\varkappa(\hbar\omega)$ practically coincide, i.e., the crystal is uniaxial when magnetized along the [111] axis.

Having observed absorption anisotropy for the (110) plane in the Faraday geometry, we investigated the dependence of the transmitted light on the angle between the vector **e** of the light wave and the crystallographic axes at fixed wavelengths of the transmitted light. The angle between the direction of **e** and the crystallographic axes of the sample can be changed by rotating the polarizer or the sample around an axis that coincides with the propagation direction of the light. When the analyzer is rotated, the light intensity is changed because of the features of the polarizer design (Frank-Ritter prism), and also because of the displacement of the transmitted ray. We have therefore rotated the sample directly in the cryostat through angles up to 540°.

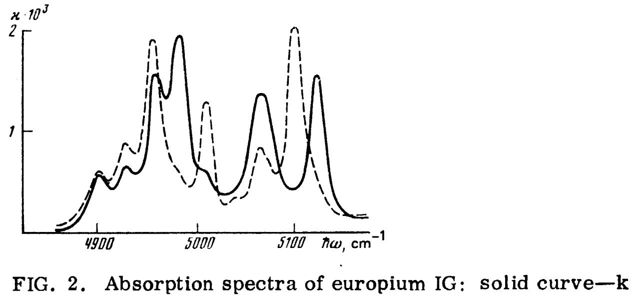

FIG. 2. Absorption spectra of europium IG: solid curve—**k** ∥[110], **I**∥[110], **e**∥[110]; dashed—**k** ∥[110], **I** ∥ [110], **e**∥[001].

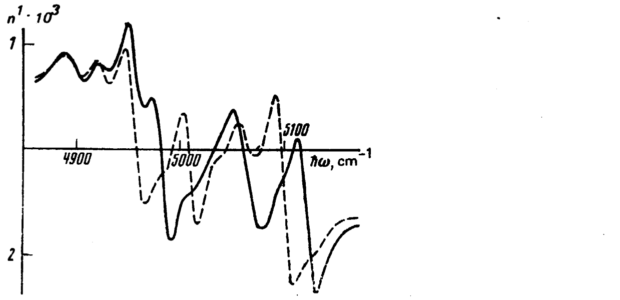

FIG. 3. Plot $n'(\hbar\omega)$ of the contribution made to the refractive index by the optical transition $^7F_0\to{}^7F_6$ of the Eu^{3+} ion of the europium IG: solid curve—**k**∥[110], **I**∥[110], **e**∥[110], dashed—**k**∥[110], **I**∥[110], **e**∥[001].

Figure 5 shows a recorder chart of the intensity of the transmitted light vs the rotation angle of the vector **e** relative to the crystallographic axes at two fixed frequencies, 5100 cm^{-1} (solid curve) and 5125 cm^{-1} (dashed curve). The distance between the extrema of the curves amounts to 90°. Figure 2 shows the spectra obtained at sample positions corresponding to the positions of the extrema of the curves of Fig. 5. We have assumed here that the orientation of the sample corresponds to a situation wherein the light-wavevector is aligned with the crystallographic axis [001] or [110] at the center of the sample on account of the Faraday effect. We have measured the rotation angle α_F of the principal axis of the polarization ellipse of the light wave emerging from the sample vs the light frequency for a plate 100 μm thick cut in the (111) plane. The maximum rotation reaches 30° at a frequency 5000 cm^{-1}. In the region of the maximum differences between the curves of Fig. 2, at the frequencies $\hbar\omega = 5100$ and 4890 cm^{-1} we have $\alpha_F = 8°$ and 24°, respectively.

DISCUSSION OF RESULTS

It follows from the presented experimental data on light absorption in $Eu_3Fe_5O_{12}$ in the region of the $^7F_0$

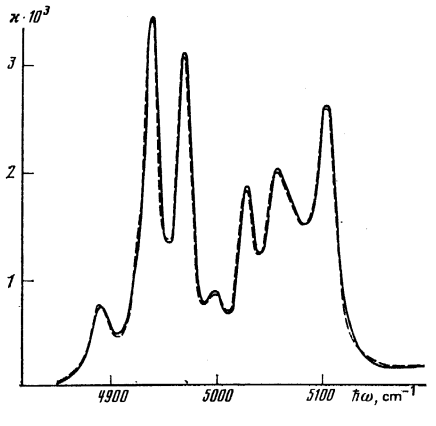

FIG. 4. Absorption spectra of europium IG: solid curve—**k**∥[111], **I** ∥[111], **e**∥ $[\bar{1}10]$; dashed—**k**∥[111], **I**∥[111], **e**⊥$[\bar{1}10]$.

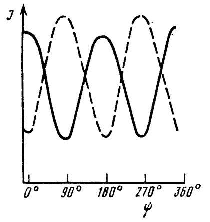

FIG. 5. Transmitted light intensity vs. the angle between the vector **e** and the crystallographic axes of the sample: $\mathbf{k}\parallel[110]$, $\psi = 0°$ corresponds to the [001] axis. Solid curve—$\hbar\omega = 5100$ cm^{-1}, dashed—$\hbar\omega = 5125$ cm^{-1}.

$\to {}^7F_6$ transition that the equatorial sections of the characteristic surfaces Im$\hat{\varepsilon}$ and Re$\hat{\varepsilon}$ determined by the method illustrated in Fig. 2a are circles ($\varkappa_1 = \varkappa_2, n_1 = n_2$), thus indicating that the crystal magnetized along the [110] axis is optically uniaxial. If, however, the anisotropy is investigated by the method illustrated in Fig. 1b (the direction of I remains the same as before), the crystal turns out to be optically biaxial ($\varkappa_1 \neq \varkappa_2$, $n_1 \neq n_2$). This situation seems incompatible with the traditional premises of the crystal optics of magnetically ordered crystals, which call for the anisotropy of the optical properties (say, uniaxiality or biaxiality) to be determined by the orientation of I and be independent of the orientation of the wave vector k of the light wave. Let us dwell on this question in greater detail.

Investigations of the MBL[1] and of the orientational Faraday effect caused by it[5] have shown that the anisotropy of the optical properties of an IG magnetized far from the absorption lines can be satisfactorily described with the aid of a dielectric tensor expanded in powers of the magnetization I. In the general case, as shown in Ref. 1, a cubic crystal becomes optically biaxial upon magnetization, and the angle between the optical axes is determined by the orientation of the vector I in the crystal, namely, if I is directed along axes such as [111] and [100] the crystal is uniaxial, and at other directions of I it is in general biaxial.

When analyzing the properties of light propagation in an isotropic absorbing medium, and in particular in the region of the absorption bands, account must be taken of more complicated effects connected with magnetic linear and circular dichroism. It appears that this circumstance explains the lack of systematic studies of the anisotropy of the optical properties of absorbing crystals in the magnetized state. The crystal $Eu_3Fe_5O_{12}$ chosen by us is optically uniaxial in the transparency region.[6] We have shown earlier[3] that in the case of transverse magnetization ($\mathbf{k}\perp\mathbf{I}$) the crystal remains uniaxial also in the absorption lines. One remark must be made here concerning the employed term "uniaxial." The fact that the two experimental plots of the absorption coefficient $\varkappa$ coincide at $\mathbf{k}\parallel[001]$, $\mathbf{e}\parallel[\bar{1}10]$, $\mathbf{I}\parallel[110]$ and $\mathbf{k}\parallel[\bar{1}10]$, $\mathbf{e}\parallel[001]$, and $\mathbf{I}\parallel[110]$ gives grounds for assuming that in this case the cross sections of the two surfaces corresponding to Re$\hat{\varepsilon}$ and Im$\hat{\varepsilon}$ are circles, i.e., the characteristic surfaces of the complex tensor $\hat{\varepsilon}$ have axial symmetry with an axis parallel to I.

We shall show now that the traditional approach based on expanding $\hat{\varepsilon}$ in powers of I is inapplicable in absorption lines. If the symmetry of the Re$\hat{\varepsilon}$ and Im$\hat{\varepsilon}$ characteristic surfaces were to be determined only by the orientation of the vector I, then on going from the Voigt configuration ($\mathbf{k}\parallel\mathbf{I}$) to the Faraday configuration ($\mathbf{k}\perp\mathbf{I}$) a crystal uniaxial in the sense indicated above would remain uniaxial (with a fixed direction of the vector I relative to the crystallographic axes), and as a consequence all the absorption curves in the Faraday configuration at various orientations of the light-wave vector e should coincide. Experiment shows, however, that at $\mathbf{k}\parallel\mathbf{I}$ the $\varkappa(\hbar\omega)$ spectra for $\mathbf{e}\parallel[001]$ and $\mathbf{e}\parallel[110]$ differ greatly (Fig. 2).

Let us analyze now in succession the role of the following factors: the finite thickness of the plate, the presence of the Faraday effect, and the tensor character of μ. We shall show that allowance for the finite thickness of the plate does not change the conclusion. The intensity J of the light passing through the plate is connected with the components of the vector e of the light wave by the relation

$$J = F(N_1)e_\sigma^2 + F(N_2)e_\pi^2, \tag{1}$$

$$F(N_i) = 8|N_i|^2/\{[1+|N_i|^2)^2 + 4N_i'^2]\,\mathrm{ch}\,\beta_i'' + [(4|N_i|^2+1)N_i']\,\mathrm{sh}\,\beta_i'' + [(1+|N_i|^2)^2 + 4N_i''^2]\cos\beta_i' + [(4|N_i|^2+1)N_i'']\sin\beta_i'\},$$

where $N_i = N'_i + iN''_i$ are the complex refractive indices of the π and σ waves, and correspond to two mutually perpendicular polarizations of the waves (e_π to $\mathbf{e}\parallel\mathbf{I}$ and e_σ to $\mathbf{e}\perp\mathbf{I}$),

$$\beta_i = \pi N_i d/\lambda = \beta' + i\beta''.$$

If the tensor $\hat{\varepsilon}$ is represented in the form

$$\hat{\varepsilon} = \varepsilon_0 + \alpha_\varepsilon I^{\times 2} + \beta_\varepsilon I_i^2 \delta_{ij} + i\gamma I^\times, \tag{2}$$

where $I^\times$ is the tensor dual to the vector I, we obtain for $N_i (i = 1, 2)$ in the case $\mathbf{k}\parallel[\bar{1}10]$

$$N_1^{(1)} = [\varepsilon_0 - \alpha_\varepsilon - \gamma^2/(\varepsilon_0 - \alpha_\varepsilon + \beta_\varepsilon/2)]^{1/2}, \quad N_2^{(1)} = (\varepsilon_0 + \beta_\varepsilon/2)^{1/2}$$

and in the case $\mathbf{k}\parallel[001]$

$$N_1^{(2)} = [\varepsilon_0 - \alpha_\varepsilon + \beta_\varepsilon/2 - \gamma^2/(\varepsilon_0 - \alpha_\varepsilon)]^{1/2}, \quad N_2^{(2)} = (\varepsilon_0 + \beta_\varepsilon/2)^{1/2}.$$

The spectra $J(\mathbf{k}\parallel[\bar{1}10])$ and $J(\mathbf{k}\parallel[001])$ will coincide only in the case $N_1^{(1)} = N_1^{(2)}$, whence either : (a) $\gamma^2 = (\varepsilon_0 - \alpha_\varepsilon + \beta_\varepsilon/2)(\varepsilon_0 - \alpha_\varepsilon)$, or (b) $\beta_\varepsilon = 0$. In case (a) we have $N_1 = 0$ and, as follows from (1), $J = 0$, i.e., no waves are excited in the medium. From (b) follows isotropy in e in the Faraday configuration:

$$J(\mathbf{k}\|\mathbf{I}) = F(N_1) + F(N_2), \quad N_{1,2}^{(3)} = (\varepsilon_0 + \alpha_\varepsilon \pm \gamma)^{1/2},$$

which, as noted above, contradicts the experimental data (Fig. 2).

The character of light propagation in magnetically ordered crystals is substantially different in the Voigt and Faraday configurations. In the former case the polarization plane of the light propagating in the crystal remains unchanged. In the Faraday configuration the polarization plane is rotated. It is natural to assume that it is precisely the rotation of the polarization plane which leads to the difference between the two spectra in Fig. 1. This difference can be due to the existence of sites that are not equivalent with respect to the vector e and are occupied by the RE ions in IG.[7] The light intensity for each site having a surrounding with a symmetry group D_2 can be represented in the form

$$J=\sum a_i e_i'^2, \quad i=x, y, z,$$

where $a_i = a_i^0 + \Delta_i I'^2_i$, while the primes denote that the components of the vectors e and I were taken in the local coordinate frame. The total light-absorption intensity by the six nonequivalent RE ions at $\mathbf{I} \parallel [110]$ is written in the form

$$J_x=[2(a_x^0+a_y^0+a_z^0)+{}^3/_4(\Delta_x+\Delta_y)+\Delta_z](e_x^2+e_y^2)$$
$$+[2(a_x^0+a_y^0+a_z^0)+{}^1/_2(\Delta_x+\Delta_y)]e_z^2+(\Delta_x+\Delta_y)e_x e_y.$$

From the condition that the crystal be uniaxial we get in the Voigt geometry the relation

$$\Delta_z=(\Delta_x+\Delta_y)/4. \tag{3}$$

In the Faraday geometry

$$e_x=-e_y, \quad \sqrt{2}e_x=e_{x0}\cos\rho\xi+e_{z0}\sin\rho\xi,$$
$$e_z=-e_{x0}\sin\rho\xi+e_{z0}\cos\rho\xi,$$

where $\xi=(x+y)/\sqrt{2}$, ρ is the specific Faraday rotation and ξ is the path traversed by the light. Summation over the nonequivalent sites of the RE ions relative to e reduces to integration with respect to ξ. As a result we obtain for J_1 ($\mathbf{e} \parallel [001]$):

$$J_1=[2(a_x^0+a_y^0+a_z^0)+{}^3/_8(\Delta_x+\Delta_y)+{}^1/_2\Delta_z]+\left[\frac{1}{16}(\Delta_x+\Delta_y)-{}^1/_4\Delta_z\right]\frac{\sin\alpha}{\alpha},$$

where $\alpha=\rho d$, and an analogous relation for J_2 ($\mathbf{e} \parallel [\bar{1}10]$)

$$J_2=[2(a_x^0+a_y^0+a_z^0)+{}^1/_8(\Delta_x+\Delta_y)+{}^3/_2\Delta_z]+\left[\frac{1}{16}(\Delta_x+\Delta_y)-{}^1/_4\Delta_z\right]\frac{\sin\alpha}{\alpha}.$$

As a result we have

$$J_1-J_2=(\Delta_x+\Delta_y)/4-\Delta_z,$$

whence, taking (3) into consideration, we get $J_1=J_2$, i.e., allowance for the Faraday effect cannot explain the experimentally observed biaxiality of the crystal at $\mathbf{k} \parallel \mathbf{I}$.

Let us dwell also on one possibility of explaining the results without going outside the framework of classical crystal optics. We have in mind introduction of the magnetic permeability tensor $\hat{\mu}$. When the spectra are measured for the σ components the vector h remains unchanged, and in the Faraday configuration the orientation of h changes when the orientation of e is changed. We can thus attempt to attribute the experimental facts to the anisotropy of the crystal response to h, i.e., assume the tensor $\hat{\mu}$ biaxial but $\hat{\varepsilon}$ uniaxial:

$$\mu=\mu_0+\alpha_\mu I^{\times 2}+\beta_\mu I_i\delta_{ij}, \quad \beta_\varepsilon=0.$$

Actually, as shown by calculation, the complex refractive indices $N_{1,2}$ of the waves with polarizations $\mathbf{e} \parallel [\bar{1}10]$, $\mathbf{k} \parallel [001]$ and $\mathbf{e} \parallel [001]$, $\mathbf{k} \parallel [110]$ coincide in this case and are equal to

$$N_1^{(1)}=N_1^{(2)}=[(\varepsilon_0-\alpha_\varepsilon)(\mu_0+{}^1/_2\beta_\mu)]^{1/2},$$

while in the Faraday configuration they are different[2)]:

$$N_1^{(3)}=[(\varepsilon_0-\alpha_\varepsilon)(\mu_0-\alpha_\mu)]^{1/2}, \quad \mathbf{e}\parallel[\bar{1}10],$$

$$N_2^{(3)}=[(\varepsilon_0-\alpha_\varepsilon)(\mu_0-\alpha_\mu+{}^1/_2\beta_\mu)^{1/2}, \quad \mathbf{e}\parallel[001].$$

Here, however, the difference between the Faraday spectra should lead also to a difference between the π spectra at $\mathbf{k} \parallel [001]$ and $\mathbf{k} \parallel [\bar{1}10]$:

$$N_2^{(1)}=[\varepsilon_0(\mu_0-\alpha_\mu+{}^1/_2\beta_\mu]^{1/2}, \quad \mathbf{k}\parallel[001],$$
$$N_2^{(2)}=[\varepsilon_0(\mu_0-\alpha_\mu)]^{1/2}, \quad \mathbf{k}\parallel[\bar{1}10].$$

No such difference is observed in experiment, so that introduction of the tensor μ does not explain all the available experimental data. We conclude thus that the optical anisotropy of the crystal undergoes a substantial change of nontrivial origin when the relative orientation of the vectors k and I changes.

Without specifying the microscopic mechanism of this change, we shall show below that simultaneous uniaxiality of the crystal in the Voigt configuration with biaxiality in the Faraday configuration does not contradict in principle the general requirements that follow from the symmetry of the magnetized cubic crystal, if consistent account is taken of the dependences of the crystal optical properties on the direction cosines of the light-wave vector.

The absorption coefficient of linearly polarized light, just as any true scalar, can be expanded in the direction cosines I_i and k_i of the vectors I and k. In a cubic crystal (symmetry group O_h) we have accurate to terms quadratic in I_i and k_i:

$$\varkappa=\sum_{i=1}^{3}e_i^2[\alpha_1(\omega)(k_j^2I_j^2+k_k^2I_k^2)+\alpha_2(\omega)k_i^2I_i^2+\alpha_3(\omega)(k_j^2I_k^2+k_k^2I_j^2)$$
$$+\alpha_4(\omega)I_i^2(k_j^2+k_k^2)+\alpha_5(\omega)k_i^2(I_j^2+I_k^2)]$$
$$+\sum_{i,j=1}^{3}e_ie_j[\beta_1(\omega)k_k^2I_iI_j+\beta_2(\omega)I_k^2k_ik_j+\beta_3(\omega)I_iI_j(k_i^2+k_j^2)+\beta_4(\omega)k_ik_j(I_i^2+I_j^2)], \tag{4}$$

where $\alpha_i(\omega)$ and $\beta_i(\omega)$ are certain coefficients that depend in the general case on the frequency ω of the light.

The foregoing expression is based on the complete rational basis of the O_h group with account taken of in-

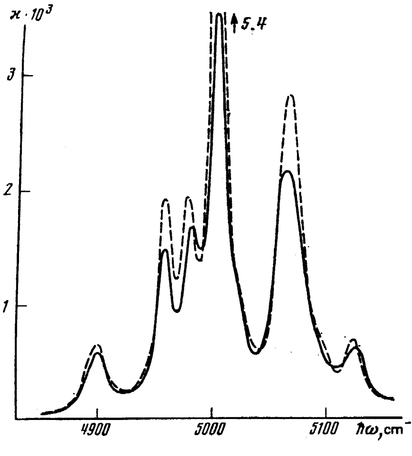

FIG. 6. Absorption spectra of europium IG: solid curve—$\mathbf{k}\parallel[001]$, $\mathbf{I}\parallel[\bar{1}10]$, $\mathbf{e}\parallel[\bar{1}10]$; dashed—$\mathbf{k}\parallel[110]$, $\mathbf{I}\parallel[\bar{1}10]$, $\mathbf{e}\parallel[\bar{1}10]$.

variance to time reversal. Let us consider some particular consequences of the expansion (4). We direct the magnetization first along the fourfold axis:

1. $\mathbf{k}\parallel[001]$, $\mathbf{I}\parallel[001]$,

$$\varkappa=\alpha_1(e_x^2+e_y^2)=\alpha_1,$$

i. e., there is no isotropy of $\varkappa$, as is indeed observed in experiment.

2. $\mathbf{k}\parallel[110]$, $\mathbf{I}\parallel[001]$,
   a) $\mathbf{e}\parallel[001]$, $\varkappa=\alpha_4$,
   b) $\mathbf{e}\parallel[\bar{1}10]$, $\varkappa={}^1/_2(\alpha_3+\alpha_5-2\beta_2)$.
3. $\mathbf{k}\parallel[100]$, $\mathbf{I}\parallel[001]$,
   a) $\mathbf{e}\parallel[010]$, $\varkappa=\alpha_3$,
   b) $\mathbf{e}\parallel[001]$, $\varkappa=\alpha_4$.

We find from 2a and 3b that the spectra $\varkappa(2.a)$ and $\varkappa(3.b)$ should coincide. This is indeed the case (see Fig. 2 of Ref. 3). For a threefold axis:

4. $\mathbf{k}\parallel[111]$, $\mathbf{I}\parallel[111]$,

$$\varkappa={}^1/_{18}(4\alpha_1+2\alpha_2+4\alpha_3+4\alpha_4+4\alpha_5-\beta_1-\beta_2-2\beta_3-2\beta_4).$$

The isotropy in **e** in this experimental geometry agrees with experiment (Fig. 4).

We consider now the most interesting case, when the magnetization is directed along a twofold axis.

5. $\mathbf{k}\parallel[001]$, $\mathbf{I}\parallel[110]$,
   a) $\mathbf{e}\parallel[110]$, $\varkappa={}^1/_2(\alpha_3+\alpha_4)+{}^1/_4\beta_1$,
   b) $\mathbf{e}\parallel[\bar{1}10]$, $\varkappa={}^1/_2(\alpha_3+\alpha_4)-{}^1/_4\beta_1$.
6. $\mathbf{I}\parallel[110]$, $\mathbf{k}\parallel[\bar{1}10]$,
   a) $\mathbf{e}\parallel[110]$, $\varkappa={}^1/_4(\alpha_1+\alpha_2+\alpha_4+\alpha_5+\beta_3-\beta_4)$,
   b) $\mathbf{e}\parallel[001]$, $\varkappa={}^1/_2(\alpha_1+\alpha_3)$.
7. $\mathbf{k}\parallel[110]$, $\mathbf{I}\parallel[110]$,
   a) $\mathbf{e}\parallel[001]$, $\varkappa={}^1/_2(\alpha_1+\alpha_3)$,
   b) $\mathbf{e}\parallel[\bar{1}10]$, $\varkappa={}^1/_4(\alpha_1+\alpha_2+\alpha_4+\alpha_5-\beta_3-\beta_4)$.

The condition that the crystal magnetized along the [110] axis be uniaxial in the Voigt geometry is obtained by equating (6.b) and (5.b).

It is seen that the condition that the condition for the quality of the $\varkappa(\hbar\omega)$ spectra in the case shown in Fig. 2 does not exclude birefringence in the Faraday configuration, i.e., generally speaking

$$\varkappa(7.a)-\varkappa(7.b)\neq0.$$

It follows from relations 7.a and 6.b that the $\varkappa(\hbar\omega)$ spectra at $\mathbf{I}\parallel[110]$, $\mathbf{k}\parallel[110]$, $\mathbf{e}\parallel[001]$ and $\mathbf{I}\parallel[110]$, $\mathbf{k}\parallel[\bar{1}10]$, $\mathbf{e}\parallel[001]$ should coincide. Indeed the $\varkappa(\hbar\omega)$ spectrum shown in Fig. 2 in the Faraday geometry (solid curve) coincides with the $\varkappa(\hbar\omega)$ spectrum in the Voigt geometry (solid curve of Fig. 5 of Ref. 3). The following relation is valid:

$$\begin{gathered}{}^1/_2[\varkappa(\mathbf{k}\parallel[100],\ \mathbf{I}\parallel[001],\ \mathbf{e}\parallel[010])+\varkappa(\mathbf{k}\parallel[001],\ \mathbf{I}\parallel[001])]\\ =\varkappa(\mathbf{k}\parallel[110],\ \mathbf{I}\parallel[110],\ \mathbf{e}\parallel[001]).\end{gathered}\tag{5}$$

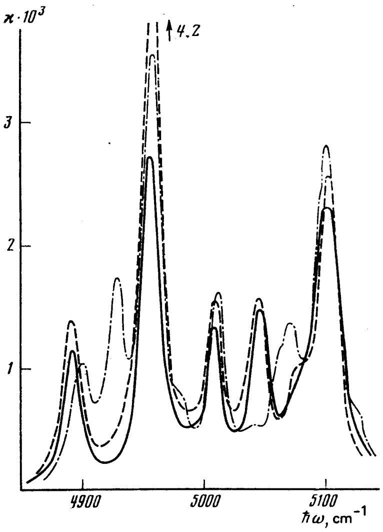

FIG. 7. Plot of Eq. (5). Solid curve ($\mathbf{I}\parallel[001]$)—left-hand side of (5), dashed ($\mathbf{I}\parallel[110]$)—right-hand side of (5); dashed curve—$\varkappa(\hbar\omega)$ spectrum from Ref. 3 ($\mathbf{k}\parallel[110]$, $\mathbf{I}\parallel[001]$, $\mathbf{e}\parallel[110]$).

Figure 7 shows the frequency dependences of the right- and left-hand sides of (5), calculated from the curves of Fig. 2 and from the absorption spectra in the Faraday geometry for the (100) plane. The $\varkappa(\hbar\omega)$ spectrum at $\mathbf{k}\parallel[\bar{1}10]$, $\mathbf{I}\parallel[001]$, $\mathbf{e}\parallel[\bar{1}10]$ is shown for comparison. The two spectra (solid and dashed lines) coincide when the vector **I** is directed along the same axis. The solid and dashed lines deviate in those regions where the expansion (2) of ε in terms of **I** is not valid (see Ref. 3).

The expansion (4) makes it possible thus to explain, from a unified point of view, all the observed features of the optical anisotropy of europium IG. At definite relations between the coefficients α_i and β_i the expansion (4) is reduced to a form that determines the usual optical anisotropy of a magnetized cubic crystal, and the character of the anisotropy does not depend on the relative orientation of the vectors **k** and **I**.

The relations

$$\alpha_1=\alpha_3,\quad \alpha_2={}^1/_2(2\alpha_3+\beta_1+\beta_2),\quad \alpha_5={}^1/_2(2\alpha_3+\beta_2),$$
$$\beta_2=\beta_4,\quad \beta_1=\beta_3$$

define an optically biaxial crystal, and if we introduce the additional relation

$$\alpha_4={}^1/_2(2\alpha_3+\beta_1),$$

then the crystal is uniaxial at any orientation of the vector **I**.

By changing to a local coordinate frame connected with the vectors **k** and **e**, one can separate in (4) the terms that are essentially anisotropic in the wave vector **k**. These terms, when account taken of the obtained data, do not vanish and it can therefore be proposed that the anomalous crystal optics of the europium IG, observed by us, is due to spatial dispersion, which begins to influence particularly strongly the optical properties of the crystal[8] in the absorption-line region.

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[1] Identical spectra are hereafter taken to be $\varkappa(\hbar\omega)$ curves all of whose peaks coincide in position, and whose amplitude ratios correlate with one another.

[2] For simplicity we neglect the $\hat{\varepsilon}$ and $\hat{\mu}$ terms that are odd in **I**, since allowance for them does not affect the main conclusions.

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