Anisotropy of magnetic linear dichroism in cubic magnetic substances

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A macroscopic and microscopic analysis is carried out of the second-order (with respect to magnetization) magneto-optical effects in cubic crystals containing magnetic ions in different crystal sublattices. The dipole-allowed optical transition $^6S(3d^5) \rightarrow {}^6P(3d^4 4p^1)$ in the Fe^{3+} ion at the octahedral and tetrahedral positions in the yttrium iron garnet is considered in detail. It is shown that in the case of cubic symmetry of the local surroundings of the magnetic ion the absorption does not depend on the direction of the magnetization **m** relative to the crystallographic axes, i.e., the magneto-optic effects are isotropic. Magneto-optic anisotropy arises if uniaxial deformations of the crystalline field around the magnetic ions are taken into account. The relation between the isotropic and anisotropic contributions is determined by the magnitudes of the absorption-band splitting due to noncubic distortions and to spin-orbit interaction. The relations between the macro- and microscopic parameters are obtained within the framework of the axial model for the case of an arbitrary direction of the local distortion axis. The results can explain a large number of "forbidden" effects observed in cubic crystals with various magnetic ions.

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1. INTRODUCTION

Recent investigations of magneto-optical effects of second order, such as magnetic linear birefringence and spontaneous and photoinduced magnetic linear dichroism, have shown (see the reviews[1,2] as well as Refs. 3–12) that in addition to the extraordinarily large magnitude, their characteristic feature is a strong anisotropy, i.e., dependence of the effects on the direction of the magnetization **m** in the crystal. Whereas the large magnitude of the magneto-optic effects can be sufficiently well explained as being due to the presence of large spontaneous magnetization, the nature of the magneto-optic anisotropy is at present less clear. To explain it, many models were proposed in the literature, based on the anisotropic interaction of polarized light with magnetic ions contained in a low-symmetry crystal field. These models were analyzed in greatest detail for the case of rare-earth iron garnets, where the magnetic ions are in octahedral, tetrahedral, and dodecahedral positions.[2,6,7] It was assumed that absorption of polarized light by magnetic ions can be described by the expression

$$W_r \sim (\mathbf{E}\mathbf{u}_r)^2 (\mathbf{m}\mathbf{u}_r)^2, \tag{1}$$

where **E**, **m**, and $\mathbf{u}_r$ are the unit vectors that characterize the directions of the polarization of the light, of the magnetization, and of the local high-symmetry axis of the crystal field around the considered ion. We shall hereafter call this the axial model. To describe the photoinduced linear dichroism, the following expression was proposed in Ref. 7

$$W_r = \{A[1+B(\mathbf{u}_r\mathbf{E})^2][1+C(\mathbf{u}_r\mathbf{m})^2]\}, \tag{2}$$

where W_r is the probability of photon absorption by a magnetic ion in the position r. It follows from (1) and (2) that the absorption is a maximum if the magnetization and polarization of the light are directed along the distortion axis of the local environment of the magnetic ion. It should be noted that Eqs. (1) and (2) are based only on intuitive ideas concerning the interaction of polarized light with a magnetic ion in a crystal field with axial distortion, but have so far not been corroborated microscopically in any way. Nonetheless, these formulas will be used to develop a theory of magneto-optic and photoinduced anisotropy, and the important result was here that the character of the anisotropy turned out to be substantially different for ions in the octahedral, tetrahedral, and dodecahedral surroundings.[2,6] Thus, for example, for ions in octahedra distorted along a threefold axis of the [111] type, the magnetic linear dichroism should vanish at an orientation of the magnetization along a fourfold axis of the type [100]. The same situation holds for the photoinduced linear dichroism when **m** is oriented along the [100] axis.[2,7] For ions in tetrahedra distorted along axis of the [100] type, in turn, the magnetic linear dichroism is a maximum at $\mathbf{m} \| [100]$. These and other conclusions have made it possible to deduce the distribution of various magnetic ions over the sublattices in the garnet structure from the character of the anisotropy of the linear dichroism.[8–10,12]

However, a careful examination of the experimental results points to systematic violation of the "ideal" picture of the magneto-optic or photoinduced anisotropy predicted by the axial model. Thus, in Ref. 11 the "forbidden" photoinduced dichroism at $\mathbf{m} \| [100]$ and $\mathbf{E} \| [100]$ was approximately one-quarter of the allowed one. The "forbidden" dichroism was observed also in many other studies.[7–10] To explain it, two mechanisms were proposed, namely the distribution of magnetic ions of the same type over different sublattices, and deviation of the local symmetry of the impurity ions from axial.[2] These mechanisms can explain in principle the appearance of forbidden dichroism, but so far there is no proof that this is in fact the situation. Indeed, in the case of pure yttrium iron garnet,[3] when these two mechanisms do not operate, there is still a clear-cut deviation from the axial model for the dichroism in the region of the first maximum

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of the electron absorption $\sim 0.9\ \mu$m. There is no doubt that this maximum is connected with the transition ${}^6A_{1g} \rightarrow {}^4T_{1g}$ for the octahedral ion of trivalent iron, but the "forbidden" dichroism at $\mathbf{m} \| [100]$ is approximately double the allowed.

The foregoing circumstances, namely the insufficient microscopic justification for Eqs. (1) and (2) and the presence of systematic discrepancies between the predictions of the axial model and the experimental data stimulated the present study, devoted to a macroscopic and microscopic analysis of the absorption of light by cubic magnets.

2. PRELIMINARY REMARKS

Since the overwhelming number of presently available experimental data on magneto-optic anisotropy were obtained for rare-earth iron garnets (REIG), we shall carry out the analyses for just these crystals. Nonetheless, many of the results obtained below are general in character and can be extended to other cubic magnets, for example those with spinel, perovskite, or rock-salt structures.

The properties of REIG have been well described in the handbook literature,[13] and we shall recall here only those of them which will be needed for the analysis. The REIG crystallized in a cubic structure with point group $m3m$ and space group O_h^{10}. The unit cell contains eight formula units $R_3Fe_5O_{12}$. The rare-earth ions occupied the dodecahedral positions 24*c*, while the ions of the trivalent iron occupies the octahedral positions 16*a* and the tetrahedral positions 24*d*. An essential element in the structure are the local distortions of the oxygen polyhedra. The octahedra are elongated along the threefold axes of type [111], and this leads to a lowering of the local symmetry of the 16*a* positions from the point group O_h to the group C_{3i}. The tetrahedra are elongated along one of the axes of the type [001], and this lowers their symmetry from T_d to S_4. The dodecahedra are elongated along one of the twofold axes of the type [110], their local symmetry is lowered to D_{2h}, and the crystal field can no longer be regarded as axial.

The distortions of the oxygen polyhedra depend on the types of the ions making up the garnet, and can reach appreciable magnitudes. Thus, for yttrium iron garnet the length of the edges of both the octahedra and the tetrahedra differ by approximately 10%. Since the parameters of the crystal field depend strongly on the distance r between the ions, for example $Dq \sim r^{-5}$ (Ref. 14), the axial component of the field should be sufficiently large. So far, however, there is no reliable quantitative proof of the appearance of noncubic distortions in the electronic spectra of the iron ions in the garnets, with the only exception of assumptions that a connection exists between the splitting of the first maximum of the absorption and the region of 0.9 μm and the lowering of the octahedra symmetry.[15] That the additional structure in the spectra is connected with the lowering of the local symmetry was assumed also for single sublattice garnets,[16] but no proof was presented in favor of this assumption as yet.

Absorption of light in an yttrium iron garnet in the region 10000 and 25000 cm^{-1} is due to electronic transitions between the $3d^5$ levels of the trivalent iron, split by the crystal field.[17] These transitions have relatively low intensity, inasmuch as in tetrahedra they are spin-forbidden, and in octahedra they are both spin and parity forbidden. Above 25000 cm^{-1} the absorption and the magneto-optic effects increase strongly, this being attributed to allowed dipole transitions, for example ${}^6S(3d^5) \rightarrow {}^6P(3d^4 4p^1)$. In Sec. 4 below we shall consider just this allowed transition, since the analysis becomes more complicated and is no longer clear in the case of forbidden transitions.

3. PHENOMENOLOGICAL DESCRIPTION OF QUADRATIC MAGNETO-OPTIC EFFECTS IN A CUBIC MAGNET

We consider the interaction of light with a cubic magnet within the framework of concepts of the dielectric tensor ε_{ij}. Let its magnetic ordering be describable by one magnetization vector $\mathbf{m}$. At high temperatures, in the parametric region and in the absence of magnetic field, the optical properties of such a cubic crystal are determined by the tensor $\varepsilon_{ij} = \varepsilon'_{ij} + i\varepsilon''_{ij}$, where ε'_{ij} describes the refraction and ε''_{ij} the absorption of the crystal. The transition of the crystal into the magnetically ordered state leads to the appearance of additions to the real and imaginary parts of the dielectric tensor. The increments linear in the magnetization $\mathbf{m}$ cause magnetic circular dichroism and magnetic circular birefringence of light (the Faraday effect), which we do not consider in the present paper since they are completely isotropic in a cubic magnet. The increments quadratic in the magnetization $\mathbf{m}$ lead to a change of the absorption, described by the anti-Hermitian part of the dielectric tensor $\varepsilon_{ij}^{\text{ah}}$.[17] The quadratic increments to $\varepsilon_{ij}^{\text{ah}}$ form an imaginary symmetrical tensor $i\varepsilon_{ij}^{\text{ah}}(\mathbf{m})$, where $\varepsilon_{ij}^{\text{ah}}(\mathbf{m})$ is a symmetric real tensor. For a cubic ferromagnet of symmetry $m3m$ the density of the light energy absorbed per second can be written, from symmetry considerations, in the form

$$Q/4\pi\omega = \varepsilon_0''\mathbf{E}^2 + \lambda_1''\mathbf{E}^2\mathbf{m}^2 + \lambda_2''(\mathbf{Em})^2 + \lambda_3''(E_x^2 m_x^2 + E_y^2 m_y^2 + E_z^2 m_z^2), \tag{3}$$

where E is the electric field of the light wave, λ_i'' are phenomenological constants, and ω is the frequency of the light. Differentiating (3) twice with respect to the components of $\mathbf{E}$, we can obtain an expression for the components $\varepsilon_{ij}^{\text{ah}}$ and $\varepsilon_{ij}^{\text{ah}}(\mathbf{m})$.[18] In the general case, the passage through a crystal is described by the complex refractive index $\tilde{n} = \text{Re}\tilde{n} + i\text{Im}\tilde{n}$. An analysis of the increments to the real part, which describes the influence of the magnetization on the magnetic linear (with respect to polarization) birefringence of light, was carried out earlier in Refs. 19 and 20. If the absorption is not too large, i.e., $\text{Re}\tilde{n} \gg \text{Im}\tilde{n}$, then the refraction and absorption coefficients $\text{Re}\tilde{n}$ and $\text{Im}\tilde{n}$ can be connected respectively with the Hermitian and anti-Hermitian parts of the tensor ε_{ij}.[21] In this case the different terms in (3) give rise to the following optical phenomena. The term with ε_0'' describes the isotropic, i.e., independent of polarization, light absorption which is not connected with the magnetization. The remaining terms with λ_1'' describe the isotropic absorption that depends neither on the orientation of the magnetization in the crystal nor on the polarization of the light, nor also on their relative orientation; the term with λ_2'' describes the

additional absorption that does not depend on the orientation of the magnetization in the crystal, but does depend on the relative orientation of the vectors **m** and **E**; the term with λ_3'' describes anisotropic absorption that depends on the orientation of **m** in the crystal and on the relative orientation of **m** and **E**. In other words, it can be stated that the term with λ_1'' leaves the cubic crystal optically isotropic, the term with λ_2'' makes it optically uniaxial with optical axis along the magnetization, while the term with λ_3'' transforms the crystal into an optically biaxial one, and the orientation of the axis is determined by the direction of the magnetization and by the value of the parameter of the magneto-optic anisotropy $a = \lambda_2''/(\lambda_2'' + \lambda_3'')$.[19-20] The coefficients λ_i'' can obviously be obtained from purposeful experiments on the change of absorption as a function of the orientation of the magnetization and of the polarization of the light in the crystal. To determine λ_1'' it is necessary here to measure the absolute absorption coefficient, while λ_2'' and λ_3'' can be obtained from the magnetic linear dichroism (MLD) spectra $\Delta\alpha = \alpha_\parallel - \alpha_\perp$, where $\alpha_\parallel$ and $\alpha_\perp$ are the absorption coefficients when the polarization of the light is oriented parallel and perpendicular to the magnetization. Since it is necessary to determine two parameter, λ_2'' and λ_3'', the experiment must be carried out with the magnetization oriented along two nonequivalent directions in the crystal.

The theoretical calculation of the parameters λ_1'', λ_2'', and λ_3'' entails the choice of a model that describes the anisotropic absorption of light by the magnetic centers. After summing over all the centers, definite relations are established between the microscopic and macroscopic parameters λ_i''. In Sec. 4 is considered the anisotropy of the absorption for an electrodynamic transition. In Sec. 5 are established relations in the microscopic and macroscopic parameters for the axial model at arbitrary directions of the local distortions.

4. MICROSCOPIC ANALYSIS OF THE MAGNETO-OPTIC ABSORPTION AND MLD FOR THE SINGLE-ION DIPOLE TRANSITION $^6S \to {}^6P$

The ground state of the free ion of trivalent iron $Fe^{3+}(3d^5)$ is an orbital singlet ^6S (see Fig. 1) with sixfold spin degeneracy ($S = 5/2$). The excited states correspond to several quartet and doublet terms inside the $3d^5$ shell, but dipole transitions for them are parity and spin forbidden. As noted above (see Sec. 2), the electric dipole transition between the states $^6S(3d^5) \to {}^6P(3d^5)4p^1)$ in the free ion Fe^{3+} is forbidden. Let us examine the mechanism whereby isotropic and anisotropic magneto-optic absorption sets in, using this transition as an example.

The splitting of the terms ^6S and ^6P, with consistent perturbation-theory allowance for the cubic crystalline field and for the exchange spin-orbit interaction, is shown in Fig. 1 (Ref. 22). The picture of the splitting is qualitatively the same for the Fe^{3+} ion in both the octahedral and the tetrahedral field, but the values of the crystal and exchange fields are different in these two ways. We consider for the sake of argument transitions in an octahedral iron ion.

A cubic crystal field does not split the terms ^6S and ^6P, but only shifts them in energy. The exchange field lifts the spin degeneracy, and the spin-obit interaction lifts the orbital degeneracy of the $^6T_{1u}(^6P)$ state. The electrodipole transitions are allowed between states with identical S, m_S, and $\Delta m_L = -1, 0, +1$. The exchange splitting of the ground state of the octahedral ion amounts to approximately $\Delta E = g\beta H_{exc}\Delta m_S \approx 100\ \text{cm}^{-1}$, therefore at low temperature we can confine ourselves only to consideration of transitions from the lower level of the ground state ($m_S = 5/2$). The transition $\Delta m_L = 0$ is allowed in π polarization ($\mathbf{E}\|\mathbf{m}$), while the transitions $\Delta m_L = -1, +1$ are allowed in σ^+ and σ^- polarization ($\mathbf{E}\perp\mathbf{m}$). The light energy absorbed by the crystal in electrodipole transitions can be written in the form

$$W(\omega) = \sum_i |\mathbf{d}_i\mathbf{E}|^2 f(\omega_i, \omega), \tag{4}$$

where $\mathbf{d}_i$ is the dipole-moment operator of the ith transition, $f(\omega_i,\omega)$ is the form function of the absorption band, ω_i is the resonant frequency, and ω is the frequency of the light. For the transition considered by us we can write

$$W(\omega) = f(\omega_0, \omega)(\mathbf{d}_\parallel\mathbf{E})^2 + {}^1/_2[f(\omega_0+\gamma/2, \omega) + f(\omega_0-\gamma/2, \omega)](\mathbf{d}_\perp\mathbf{E})^2, \tag{5}$$

where $\mathbf{d}_\parallel$ and $\mathbf{d}_\perp$ are dipole moments oriented respectively parallel and perpendicular to the magnetization $\mathbf{m}(\mathbf{m}\|\mathbf{H}_{exc})$, and $\gamma/2$ is the spin orbit splitting of the $^6T_{1u}$ state. Recognizing that

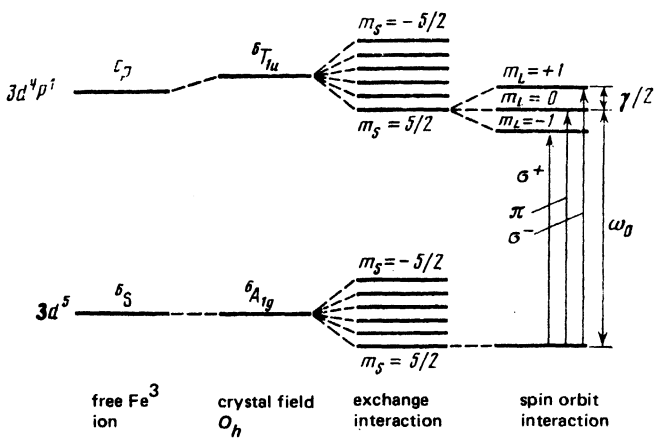

FIG. 1. Splitting of the $^6A_{1g}(^6S)$ and $^6T_{1u}(^6P)$ states in a cubic crystal field of symmetry O_h (or T_d) with allowance for the exchange and spin-orbit interactions.

$$(\mathbf{d}_\perp \mathbf{E})^2+(\mathbf{d}_\parallel \mathbf{E})^2=d^2E^2$$

(this follows from the fact that in perpendicular observation the intensity absorption of the π component is equal to the sum of the intensities of the σ components[23]) and assuming $\gamma/2$ to be less than the width of the absorption band, we have for the electrodipole absorption connected with the influence of the magnetization

$$W(\omega)=-\frac{1}{2}\left(\frac{\gamma}{2}\right)^2\frac{\partial^2 f}{\partial\omega_0^2}(\mathbf{d}_\parallel \mathbf{E})^2. \tag{6}$$

This absorption does not depend on the orientation $\mathbf{d}_\parallel$ (and meaning also on $\mathbf{m}$) relative to the crystallographic axes, i.e., the transition in a cubic field contributes to the isotropic term with λ_2'' in expression (3), and the anisotropic coefficient λ_3'' vanishes in this expression. This isotropy of the absorption and of the MLD is due to the fact that the orbital angular momentum is quantized relative to an axis directed along the magnetization $\mathbf{m}$ but not connected in any way with any crystallographic direction.

We proceed now to take into account the axial component of the crystal field. Assume that in the local coordinates (x',y',z') the distortion of the tetrahedron is directed along the z' axis. The noncubic crystal field lifts the orbital degeneracy of the ${}^6T_{1u}$ state (see Fig. 2) and partially freezes the orbital momentum. The remaining degeneracy is preserved at $\mathbf{m}\perp\mathbf{z}'$ and is lifted at $\mathbf{m}\|\mathbf{z}'$. The expression for the absorption in the case of the level scheme of Fig. 2 is of the form

$$W(\omega)=f(\omega_0-\delta,\omega)(\mathbf{d}_{z'}\mathbf{E})^2+\frac{1}{2}\left[f\left(\omega_0+\frac{\gamma}{2}(\mathbf{mz}'),\omega\right)+f\left(\omega_0-\frac{\gamma}{2}(\mathbf{mz}'),\omega\right)\right](\mathbf{d}_\perp\mathbf{E})^2, \tag{7}$$

where δ is the axial splitting of the level ${}^6T_{1u}$. Taking into account only terms that depend on the magnetization, we obtain

$$W(\omega)=-\frac{1}{2}\left(\frac{\gamma}{2}\right)^2\frac{\partial^2 f}{\partial\omega_0^2}(\mathbf{Ed}_{z'})^2(\mathbf{mz}')^2. \tag{8}$$

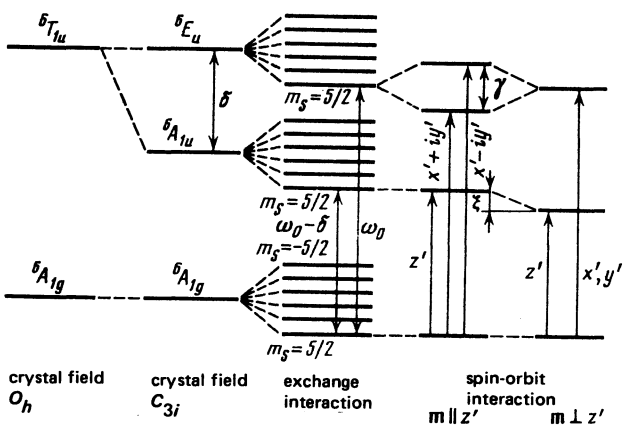

FIG. 2. Splittings of the ${}^6A_{1g}({}^6S)$ and ${}^6T_{1u}({}^6P)$ states in a crystal field of symmetry C_{3i} (octahedron elongated along the threefold axis [111]) with allowance for the exchange spin-orbit interactions.

The split-off level ${}^6A_{1u}$ can contribute to the dichroism only when account is taken of the anisotropy of the exchange field (due to the noncubic distortions of the local surrounding) acting on this state. Assuming the anisotropy of H_{exc} to be weak, we can write

$$W(\omega)=f(\omega_0-\delta+\xi(\mathbf{mz}')^2,\omega)(\mathbf{Ed}_{z'})^2 =f(\omega_0-\delta,\omega)(\mathbf{Ed}_{z'})^2+\xi\frac{\partial f}{\partial\omega_0}(\mathbf{Ed}_{z'})^2(\mathbf{mz}')^2, \tag{9}$$

where $\xi=4\beta^2S(g_\parallel^2H_{exc\parallel}^2-g_\perp^2H_{exc\perp}^2)$ is the anisotropy constant of the exchange splitting. We see that noncubic distortions of the local surrounding leads to a dependence of the absorption of the orientation of the magnetization relative to the distortion axis, and also to anisotropy of the MLD, i.e., in the general case, to a nonzero coefficient λ_3'' in (3).

Thus, consideration of the absorption for two splitting schemes shown in Fig. 1 and 2 demonstrates that if the axial splitting is substantially smaller than the spin-orbit splitting, i.e., $\delta\ll\gamma/2$, the magnetic absorption will be isotropic. In the other extreme case $\delta\gg\gamma/2$ the magnetic absorption for the split-off transition will be anisotropic. In those cases when these two splittings are comparable, mixing of the wave functions of the sublevels ${}^6E_u$ and ${}^6A_{1u}$ of the state ${}^6T_{1u}$ should lead to coexistence of the isotropic and anisotropic absorptions. For an exact solution of such a problem by perturbation theory it is necessary to take into account simultaneously the influence of the spin-orbit interaction and of the noncubic part of the crystal field on the wave functions of the electrons. To this day, however, we do not know the magnitude of these interactions. Nonetheless, for a qualitative examination we can use the analogy between the transitions from a singlet to a triplet and the classical harmonic oscillator in a magnetic field (Fig. 3). The influence of the noncubic distortions will be simulated here by the difference $\delta=\omega_1-\omega_0$ between the resonant oscillator frequencies, where ω_1 is the frequency of the oscillations along the distortion axis z', ω_0 is in the $x'y'$ plane (in the absence of the magnetic field $\mathbf{H}$), while the spin-orbit interaction will be simulated by the splitting of the states of an oscillator in the magnetic field $\mathbf{H}$. We shall assume that the field $\mathbf{H}$ lies in the $x'z'$ plane. It is easy to show that the eigenvalues of the energies and of the polarizations of the states of the oscillator

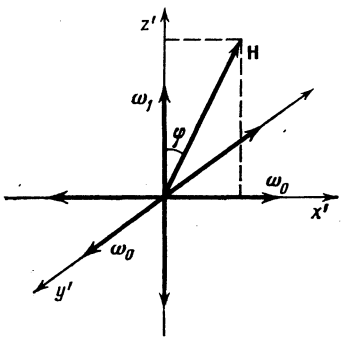

FIG. 3. Three-dimensional anisotropic harmonic oscillator in a magnetic field.

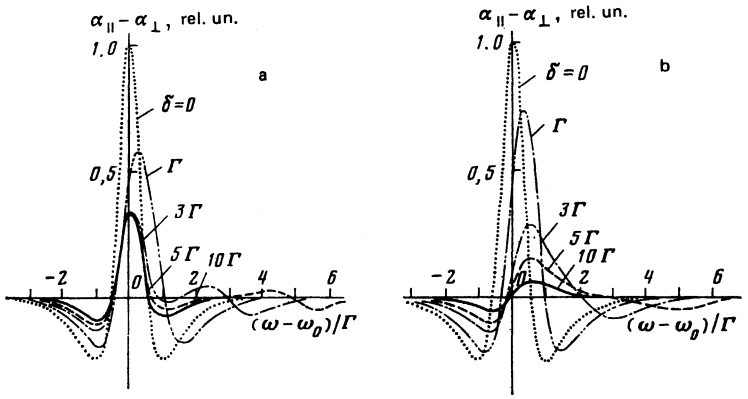

FIG. 4. Results of the calculation of the MLD for the $S\rightarrow P$ transition at different values of the axial splitting δ. The axes of the local distortions are directed along the axes of type [100], $\mathbf{k}\|[010]$; a) $\mathbf{m}\|[001]$; b) $\mathbf{m}\|[101]$.

correspond fully to the previously considered limiting values $\delta \ll \gamma/2$ and $\delta \gg \gamma/2$, if we put $\gamma/2 = eH/mc$, where e and m are the charge and mass of the electron and c is the speed of light. At arbitrary relations between $\gamma/2$ and δ (it is convenient here to introduce the parameter $\eta = 2\delta/\gamma$), the eigenstates of the harmonic oscillator are three ellipses whose directions of the principal axes, ellipticity, and resonant frequencies are complicated functions of the parameter η and of the angle φ between the directions of the distortion axis z' and of the magnetic field $\mathbf{H}$. Without going through the mathematical operations, we write down only the final expression for the contribution of an individual ith state ($i = 1,2,3$) to the MLD ($\Delta\alpha_i$):

$$\Delta\alpha_i = \frac{1}{\beta(p_i)}\left\{\frac{(k_{z'}m_{x'}-k_{x'}m_{z'})^2}{k^2m^2} + \frac{k_{y'}^2(p_i+\eta\cos^2\varphi)^2-k^2\eta^2\cos^2\varphi\sin^2\varphi}{p_i^2(p_i+\eta)^2k^2}\right\} f(\omega_i,\omega), \quad (10)$$

where $\mathbf{k}$ is the wave vector of the light, $\beta(p_i) = 1 + p_i^{-2}\times\cos^2\varphi + (p_i+\eta)^{-2}\sin^2\varphi$, p_i is one of the three solutions of the cubic equation $p^3 + \eta p^2 - p - \eta\cos^2\varphi = 0$, and $\omega_i = \omega_0 + \gamma p_i/2$. Equation (10) was obtained under the assumption that $\omega_0 \gg \gamma/2, \delta$, an assumption that holds for optical transitions.

To find the total MLD of a crystal, given η and $\mathbf{H}$, it is necessary to sum the contributions of the individual states of the harmonic oscillators $\Delta\alpha_i (i = 1,2,3)$ for all the positions that have different directions $\mathbf{u}_r(\mathbf{z}')$, i.e., different angles φ. The results of such a calculation, carried out wth a computer, for the distortion directions $\mathbf{u}_r$ and for the field $\mathbf{H}$ along the principal crystallographic axes at different values of the parameter δ are shown in Figs. 4 and 5. We have assumed in the calculation that $f(\omega_i,\omega)$ is a Lorentz function with resonant frequency ω_i and half-width Γ, while $\gamma/2\Gamma = 0.1$. Figure 6 shows the angular dependences of the MLD for octahedra and tetrahedra at different values of the axial splitting. In the absence of distortions, the MLD is isotropic and proportional to the second derivative $f''(\omega_0)$ in accordance with (6). Introduction of distortions $\delta \ll \Gamma$ changes somewhat the shape, decreases the amplitude, and leads to a small anisotropy of the MLD. Thus, at $\delta = \Gamma$, for the principal maximum of the MLD, the parameter of the magneto-optic anisotropy is $a = 0.84$ at $\mathbf{u}_r\|[111]$ and $a = 1.29$ at $\mathbf{u}_r\|[100]$. Figure 7 shows a plot of $a(\delta/\Gamma)$ at the frequency ω_0 for different distortion directions. It can be seen that substantial changes in the parameter a begin when the uniaxial splitting δ becomes larger than the half-width Γ. At $\delta > \Gamma$ the MLD line shape differs substantially from $f''(\omega_0)$ and depends on the direction of the magnetic field, and this gives rise to a frequency dependence of the parameter a. With further increase of δ ($\delta \gg \Gamma$) the line shape and the magnitude of the MLD tend to a limit described by Eq. (8) (summed over all the $\mathbf{u}_r$—see the next section). However, even at values δ /

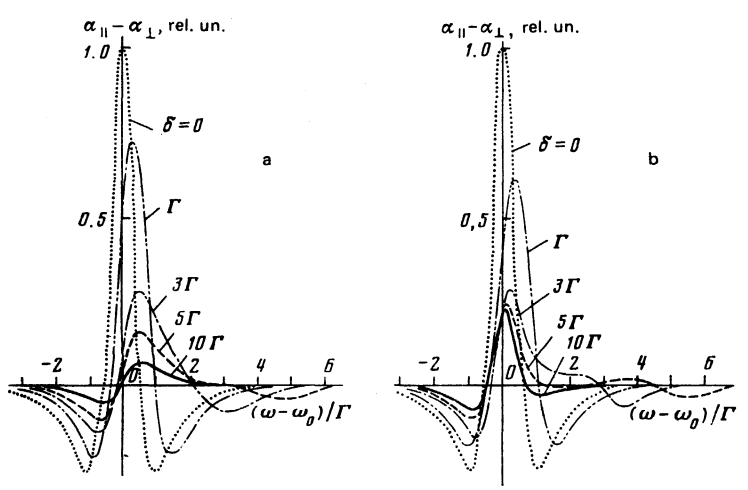

FIG. 5. Results of calculation of the MLD for the $S\rightarrow P$ transition at different values of the axial splitting δ. The axes of the local distortions are directed along axes of type [111], $\mathbf{k}\|[110]$; a) $\mathbf{m}\|[001]$; b) $\mathbf{m}\|[111]$.

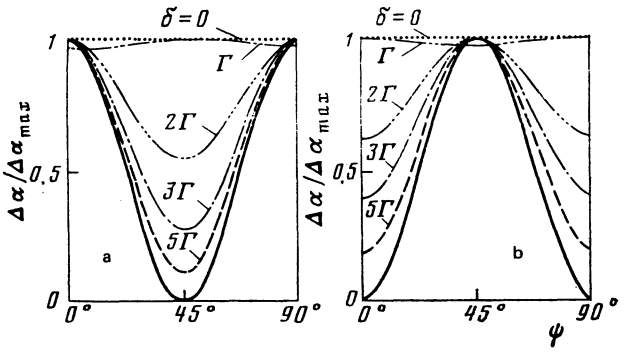

FIG. 6. Angular dependences of the MLD at $\mathbf{k}\|[100]$ and $\mathbf{m}\|[100]$ for different values of the axial splitting δ; a) $\mathbf{u}_r\|[100]$; b) $\mathbf{u}_r\|[111]$. The solid curve indicates the axial model.

$\Gamma \geqslant 5$ there are deviations from the purely axial model—the MLD line shape at $\mathbf{u}_r\|[100]$, $\mathbf{H}\|[100]$, and $\mathbf{u}_r\|[111]$, $\mathbf{H}\|[111]$ does not duplicate $f''(\omega_0)$ exactly, and in the remaining cases one observes near ω_0 an MLD proportional to $f'(\omega_0)$. The form, amplitude, and anisotropy of the MLD depend thus substantially on the relations between the parameters $\gamma/2$, δ, and Γ, with an isotropic MLD realized at $\delta \ll \gamma/2, \Gamma$; the coexistence of a "locally" isotropic and a "locally" anisotropic MLD is observed at $\delta > \gamma/2$; the purely axial model is observed at $\delta \gg \gamma/2$ and $\delta \gg \Gamma$. In the general case absorption of an individual center at a given frequency can be represented as a sum of two contributions: locally isotropic $\sim(\mathbf{E}\cdot\mathbf{m})^2$ and locally anisotropic $\sim(\mathbf{E}\cdot\mathbf{u}_r)^2(\mathbf{m}\cdot\mathbf{u}_r)^2$, in contrast to Eqs. (1) and (2), where only a locally anisotropic term is present.

The noted peculiarities of the MLD spectra of the electodipole S–P transition for the cases $\gamma/2 < \delta < \Gamma$ and $\delta > \Gamma, \gamma/2$ make it possible in principle to determine uniquely the directions of the distortions for the investigated center or, if the distortion directions $\mathbf{u}_r$ are known, to relate the transition to a particular center. There exists, however, a certain region $\delta \approx (2-3)\Gamma$ where such an identification is difficult and it is necessary to use more subtle characteristics, such as the distances between the zeros of the MLD, etc.

An interesting question is that of the influence of the distortions on the anisotropy of the MLD for the P–S transition, i.e., for the transition from a triplet to a singlet. If it is assumed that the transitions go only from a lower energy level, the MLD line shape should in this case be proportional to $f(\omega_0)$.

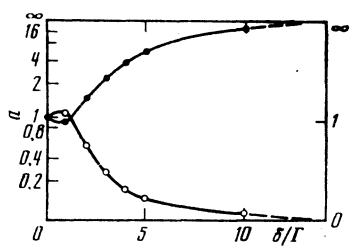

FIG. 7. Dependence of the parameter of the magneto-optic anisotropy on δ/Γ: ●) $\mathbf{u}_r\|[111]$; ○) $\mathbf{u}_r\|[100]$.

5. ANISOTROPY OF MLD OF A CUBIC CRYSTAL IN THE CASE OF THE AXIAL MODEL

In the preceding sections it was shown that noncubic distortions of the local surrounding of a magnetic field, at sufficiently large axial splitting δ, lead to a local anisotropy of the absorption, described by an expression of the type $A(\mathbf{E}\cdot\mathbf{u}_r)^2(\mathbf{m}\cdot\mathbf{u}_r)^2$, where $\mathbf{u}_r$ is the distortion axis. Locally isotropic absorption $(\mathbf{E}\cdot\mathbf{m})^2$ obviously contributes only to the term with λ_2'' in (3); we therefore consider here the possible contributions of the locally anisotropic term to the total absorption. We shall assume that the local surrounding of an individual magnetic ion has noncubic uniaxial distortions along an arbitrary direction $\mathbf{u} = u_x\mathbf{x} + u_y\mathbf{y} + u_z\mathbf{z}$($x$, y, and z are fourfold axes in the crystal), $\mathbf{u}^2 = 1$. For the crystal to be cubic, it is necessary in the general case that there exist $24n$($n = 1,2,3,\ldots$) positions of ions with different directions of the distortion $\mathbf{u}_r = g_r\mathbf{u}$ (g_r is a symmetry element of the O_h group). In the particular cases when $\mathbf{u}$ is parallel to the symmetry axis, this number decreases. In order to obtain an expression for the absorption coefficient connected with the locally anisotropic term, it is necessary sum its contribution from all the positions in the unit cell of the crystal:

$$\alpha = A\sum_{i=1}^{24}(\mathbf{E}\mathbf{u}_r)^2(\mathbf{m}\mathbf{u}_r)^2. \tag{11}$$

Without loss of generality, it suffices to consider the propagation of light along the fourfold axis $\mathbf{k}\|\mathbf{x}$ with the magnetization located in the yz plane. The expression for the MLD takes then the form

$$\Delta\alpha = 8A\sum_{i<j}[(u_i^2-u_j^2)^2\cos^2 2\psi + 4u_i^2u_j^2\sin^2 2\psi], \tag{12}$$

where ψ is the angle between $\mathbf{m}$ and the z axis; $i,j = x,y,z$. Analysis of expression (3) shows that in the case $\mathbf{k}\|[100]$ and $\mathbf{m}\|[110]$ one observes an isotropic MLD $\sim\lambda_2''m^2$, while at $\mathbf{m}\|[001]$ one observes a sum of an isotropic and anisotropic MLD $\sim(\lambda_2'' + \lambda_3'')m^2$. Comparing these results with (12) we have

$$\lambda_2''m^2 = 32A\sum_{i<j}u_i^2u_j^2, \tag{13a}$$

$$\lambda_3''m^2 = 8A\sum_{i<j}[(u_i^2-u_j^2)^2 - 4u_i^2u_j^2]. \tag{13b}$$

Thus, in the general case, the locally anisotropic term contributes to the isotropic and anisotropic MLD. The isotropic part vanishes ($\lambda_2'' = 0$) only under the condition $\mathbf{u}_r\|[001]$, etc., for tetrahedral magnetic ions. Also possible is a "accidental" vanishing of the anisotropic MLD ($\lambda_3'' = 0$) if the right-hand side of (13b) is equal to zero. If, however, $\mathbf{u}_r\|[111]$, as is the case for octahedral ions, then $\lambda_3'' = -\lambda_2''$. The anisotropy of the MLD when the local distortions are oriented along fourfold and threefold axes is shown in Fig. 6. We note that the MLD anisotropy considered in Ref. 6 is a particular case of the general formula (12).

6. COMPARISON WITH EXPERIMENT

The experimental results on the angular and spectral dependences of MLD in $Y_3Fe_5O_{12}$ show[3] that in the Voigt geometry it is perfectly correct to use the phenomenology considered above without resorting to invariants of other types, proposed for example in Ref. 24. The MLD spectra in pure $Y_3Fe_5O_{12}$ (Ref. 3) at different directions are characterized by isotropic and anisotropic MLD that are comparable in magnitude and whose relations with each other are not described by the axial model. Indeed, for the first absorption band in the 0.9-μm region, a band corresponding to the octahedral transition ${}^6A_{1g}\rightarrow{}^4T_{1g}$, the parameter $a\approx 0.5$, whereas in accordance with the purely axial model $a=\infty$ for $\mathbf{u}_r\|[111]$. This points to the coexistence of a locally isotropic and locally anisotropic MLD. Since the $a(\omega)$ dependence is very weak for this transition, one can expect realization of the case $\gamma/2<\delta\leqslant\Gamma$. We recall that for the case $\delta=\Gamma$ and $\mathbf{u}_r\|[111]$ we have $a=0.84$ for the principal maximum of the MLD (Fig. 5). Thus (disregarding the line shape of the MLD), the model considered gives the correct tendency of the deviation of the parameter a from the purely axial model. The shorter-wavelength part of the MLD spectrum is characterized by a strong $a(\omega)$ dependence that can be due either to satisfaction of the condition $\delta>\Gamma$ or to the overlap of transitions having different anisotropies.

A number of recent papers are devoted to the anisotropy of MLD in the region of the absorption bands of various ions implanted as impurities in iron garnets. In Refs. 7–11 were investigated the angular dependences of MLD in the transitions of Fe^{2+} ions, produced when $Y_3Fe_5O_{12}$ is doped with Si, Sn, and Nb. Just as for Fe^{3+}, the MLD are characterized in this case by the presence of an isotropic (λ_2'') and an anisotropic (λ_3'') part, the ratios between which depend on the type of the implanted ion. When $Y_3Fe_5O_{12}$ is doped with Nb ions[8] the angular dependences are close to the ones predicted by the purely axial model with $\mathbf{u}_r\|[100]$ (Fig. 6), and in the case of doping with Sn and Si ions[7–9]—to the model with $\mathbf{u}_r\|[111]$. On this basis the authors of the papers deduce various degrees of population of the $\mathbf{u}_r\|[111]$ octahedra and of the $\mathbf{u}_r\|[100]$ tetrahedra by the Fe^{2+} ions. In light of the concepts developed here, we can propose also another explanation of this behavior of MLD, by assuming that introduction of different ions into the garnet lattice influences the relaxation between the locally isotropic and locally anisotropic terms in the absorption. In this case the Fe^{2+} ions can occupy only octahedral sites, but can have different axial splittings when doped by different ions, causing thereby different anisotropy of the MLD.

The angular dependences and the MLD spectra, investigated in Ref. 10 for transitions on Ru^{4+} ions implanted in octahedral positions of $Y_3Fe_5O_{12}$, also offer evidence of the existence of locally isotropic and locally anisotropic MLD. For the absorption band in the region of 1.35 μm, the magnetooptic-anisotropy parameter a depends on the frequency and has in the region of the maxima a value ~ 3, i.e., in this case the anisotropic contribution exceeds the isotropic one.

Thus, all the available experimental results point to the coexistence of a locally anisotropic and a locally isotropic MLD. The relation between them, as we have shown above, depends on the concrete characteristics of the transition, such as the axial splitting, the spin-orbit splitting, the line width, etc. As a result, the anisotropy of the MLD in cubic crystals is determined not only by the direction of the axis of the local distortions of the magnetic centers, a fact taken into account in the axial model, but also by the properties of the transition itself. An analysis of the experimental results and of the theory considered here indicates that realization of the axial model is possible only when the axial splitting greatly exceeds the line width and the splitting due to the remaining interactions.

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APPENDIX

To find the eigenfrequencies and the polarizations of an anisotropic harmonic oscillator in a magnetic field it is necessary to solve the equation of motion, which can be written in the form

$$\begin{pmatrix} \omega_0^2-\omega^2 & ih_z\omega & 0 \\ -ih_z\omega & \omega_0^2-\omega^2 & ih_x\omega \\ 0 & -ih_x\omega & \omega_1^2-\omega^2 \end{pmatrix}\begin{pmatrix} a_x \\ a_y \\ a_z \end{pmatrix}=0, \tag{A1}$$

where $h_i=eH_i/mc=\gamma m_i/2$, and a_i is the electron-displacement component. The solution of this problem for the case $\omega_1=\omega_0$ can be found, for example, in Ref. 23. Equating to zero the determinant in (A1), we obtain an equation for the eigenfrequencies

$$(\omega_0^2-\omega^2)^2(\omega_1^2-\omega^2)-(\omega_0^2-\omega^2)h_x^2\omega^2-(\omega_1^2-\omega^2)h_z^2\omega^2=0. \tag{A2}$$

Introducing the notation $\Delta=2\delta/\omega_0, s^2=h^2/\omega_0^2, \eta=2\delta/\gamma$, $p=(\omega_0^2-\omega^2)/\omega_0^2 s$, and assuming that $\Delta, s\ll 1$ (in this case $\omega_1^2\approx\omega_0^2+2\omega_0\delta$), we rewrite (A2) in the form

$$p^3+p^2(\eta+s)-p(1-\Delta\cos^2\varphi)-\eta\cos^2\varphi=0.$$

Recognizing that $\Delta\ll 1$ and $\eta\gg s$ at $\eta\geqslant 1$, we obtain

$$p^3+p^2\eta-p-\eta\cos^2\varphi=0. \tag{A3}$$

The solutions p_1, p_2, and p_3 of this cubic equation determine the sought eigenfrequencies $\omega_i=\omega_0+\gamma p_i/2$ of the oscillator. The polarization of the oscillations is obtained by substituting ω_i in (A1) and by the normalization relation

$$|a_x^i|^2+|a_y^i|^2+|a_z^i|^2=d^2/e^2.$$

We have assumed in the calculation that the value of the damping, which determines the absorption of the light, is the same for all the normal oscillations, i.e., the absorption is described by one and the same form function of the absorption band, $f(\omega_i,\omega)$. In this case the polarization dependences of the absorption are determined only by the state of the polarization and by the resonant frequencies of the natural oscillations of the oscillator. Calculation of the absorption is carried out in accordance with Eq. (4), in which the components of the dipole moments $\mathbf{d}$ are determined by the components of the displacements $\mathbf{d}=e\mathbf{a}$. The expression for $\mathbf{d}^i$, as follows from (A1), is of the form

$$d_x^i = -\frac{ih_z\omega_i}{\omega_0^2-\omega_i^2}\frac{d}{\alpha}, \quad d_y^i = \frac{d}{\alpha}, \quad d_z^i = \frac{ih_x\omega_i}{\omega_1^2-\omega_i^2}\frac{d}{\alpha};$$

$$\alpha = 1+\left(\frac{h_z\omega_i}{\omega_0^2-\omega_i^2}\right)^2+\left(\frac{h_x\omega_i^2}{\omega_1^2-\omega_i^2}\right)^2, \quad i=1,2,3. \qquad \text{(A4)}$$

The MLD for the ith natural oscillations of the oscillator is given by the formula

$$\Delta\alpha_i = (|\mathbf{d}_i\mathbf{E}_\parallel|^2-|\mathbf{d}_i\mathbf{E}_\perp|^2)f(\omega_i,\omega). \qquad \text{(A5)}$$

In the subsequent transformations it is convenient to use the substitutions $E = E\,\mathbf{h}/h$ and $\mathbf{E}_\perp = E\,[\mathbf{k}\times\mathbf{h}]/kh$, where $\mathbf{k}$ is the wave vector of the light ($\mathbf{k}\perp\mathbf{h}$). Substituting these expressions in (A5), taking d_i from (A4), and expressing ω_i in terms of p_i and $\gamma/2$, we obtain Eq. (10) which was used in the calculations.

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