Magnetic linear birefringence of rare-earth garnets

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The magnetic linear birefringence (MLB) of rare-earth garnets has been investigated experimentally and theoretically. It was found that the field, temperature, and orientational dependence of the MLB depended strongly on the nature of the ground state of the rare-earth ion. In the case of $Gd_3Ga_5O_{12}$, where the Gd^{3+} ion has $L = 0$ in the ground state, the behavior of the MLB was basically described well by the formulas of the Akulov phenomenological theory of even effects. A microscopic theory predicted a specific dependence of the MLB on H and T, different from that proposed in the phenomenological theory. The MLB of garnets with non-Kramers ions, for which the ground state is an isolated singlet (Eu^{3+}, Tm^{3+}, Pr^{3+}), was found to depend quadratically on the magnetic field, was independent of temperature (in the limit $T \to 0$), and exhibited the orientational dependence predicted by the phenomenological theory. Experiments on $Tm_3Al_5O_{12}$ and $Tm_3Ga_5O_{12}$ confirmed satisfactorily the theoretical conclusions. The MLB of crystals with ions for which the ground state was a Kramers doublet (Dy^{3+}, Er^{3+}, Yb^{3+}, Sm^{3+}, or Nd^{3+}) or a quasidoublet (Tb^{3+}, Ho^{3+}) exhibited a strong field dependence in the magnetization saturation region. Generally speaking, these crystals did not obey the rules for even effects. Experiments on $Yb_3Ga_5O_{12}$, $Dy_3Al_5O_{12}$, and $Tb_3Al_5O_{12}$ confirmed fully these conclusions. It was found that the quadrupole moment of the electron shell of the rare-earth ion plays an important role in the even magnetooptic effects. In general, the symmetry of the magnetic corrections to the permittivity tensor is governed not by the point group of a crystal but by its space group O_h^{10}.

INTRODUCTION

An important task in magnetism is the determination of the relationship between the material tensors representing a crystal and its magnetic structure (in other words, the relationship with the magnetic order parameter). Knowing this relationship, we can investigate the behavior of thermodynamic and transport properties of a material when external parameters are varied, study the evolution of the properties in the course of phase transitions, consider the role of fluctuations, etc.

In the case of magnetooptic effects (particularly the magnetic linear birefringence, or MLB) it is necessary to determine the dependence of the permittivity tensor ε_{ik} on the magnetic structure of the applied magnetic field. This dependence has been investigated quite thoroughly for materials in which magnetic ions have zero or frozen orbital momentum. For example, detailed investigations have been made of the MLB of yttrium iron garnet.[1] This and other even magnetooptic effects in magnetic materials of this type can be described satisfactorily by a phenomenological theory originated by Akulov.[2,3] In this theory the tensor ε_{ik} is represented by an expansion in powers of a magnetic order parameter (magnetization, antiferromagnetic vector, etc.).

However, in the case of ions with an unfrozen orbital momentum the situation is more complex. This is confirmed, for example, by the results of an investigation of the MLB of terbium iron garnet.[1] In particular, the observed temperature dependence of the contribution of the rare-earth ion to the MLB of this ferrimagnet does not agree with the predictions of the phenomenological theory, the angular dependences of the MLB are more complex, etc.

When even magnetic effects in rare-earth magnetic materials are described, one is faced with two fundamental problems. The first is the degree of influence of the "unfrozen" state of the orbital momentum of the rare-earth ion on the even effects. The problem arises because in this case the main assumption of the Akulov theory of the even effects is not obeyed: the splitting of the energy levels of the magnetic ion in the crystal field is not small compared with the splitting in the external (or effective) magnetic field, so that the spectrum of the magnetic ion cannot be regarded as quasiequidistant. The second problem arises because in many rare-earth magnetic materials the magnetic ions occupy inequivalent crystallographic positions, so that even in the paramagnetic range of temperatures an external field induces a complex magnetic structure and it is not obvious whether the even magnetic effects can be described simply in terms of the total magnetization.

As pointed out above, many characteristic features of the MLB have been established by investigating rare-earth iron garnets. Compounds with the garnet structure are good model objects for the investigation of the even magnetic effects. Their crystal structure and the magnetic properties are known quite well.[4] Various methods have been used to determine the energy spectra of the rare-earth ions in garnets (see Ref. 4 and also the reviews in Refs. 5–8). All this makes it possible, at least in principle, to calculate quite accurately the contribution made by rare-earth ions to the MLB and to other even effects, and to compare such theoretical calculations with experimental data.

However, interpretation of the data obtained for iron garnets (and until recently measurements of the MLB of garnets have been made only for these compounds) is difficult because these substances are ferrimagnets, so that in addition to the rare-earth subsystem there is a second magnetic subsystem consisting of the iron ions.

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The contribution of rare-earth ions to the MLB was found in its pure form and the characteristics of this effect in magnetic subsystems containing ions with an unfrozen orbital momentum were determined in the present investigation by theoretical and experimental studies of the MLB in simpler paramagnetic systems containing only rare-earth magnetic ions such as garnet aluminates and gallates.

CRYSTAL STRUCTURE AND MAGNETIC PROPERTIES OF ALUMINATES AND GALLATES WITH THE GARNET STRUCTURE

Rare-earth compounds with the garnet structure can be described by a general formula $R_3M_5O_{12}$, where R is a rare earth or yttrium, and M = Fe, Al, Ga, etc. Garnets belong to the hexaoctahedral class of the cubic symmetry and are described by the space group O_h^{10}. A primitive cell of a garnet consists of four $R_3M_5O_{12}$ molecules. It is important to note that the rare-earth ions are distributed between six inequivalent c sites and the local symmetry of these sites is described by the point group D_2. The symmetry axes of all six sites (local e_α^r) are derived from the crystallographic coordinate system as a result of rotation by an angle $\pm\pi/4$ about the axes [100], [010], or [001], respectively,[9] and are listed in Table I.

Rare-earth gallates with the garnet structure $R_3Ga_5O_{12}$ (formed by all the rare earths) and rare-earth garnet aluminates $R_3Al_5O_{12}$ (formed only by heavy rare earths beginning from Gd) are antiferromagnets with very low Néel temperatures that usually do not exceed 1–2 K. Magnetic properties of gallates and aluminates are largely determined by the effect of the crystal field which is much stronger than the R–R exchange interaction (splitting of the ground-state multiplet of rare-earth ions by the crystal field in garnets is of the order of 10^2–10^3 cm^{-1}—see Refs. 5–8). It is important to note that the low symmetry of the crystal environment results in the maximum lifting of the degeneracy of the levels of the ground multiplet of the rare-earth ion (so that doublets are observed in the case of the Kramers rare-earth ions and singlets in the case of the non-Kramers ions).

SAMPLES AND MEASUREMENT METHODS

We determined the MLB of rare-earth garnet gallates (R = Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb) and garnet aluminates (R = Tb, Dy, Ho, Er, Tm, Yb) using single-crystal plates with surfaces parallel to the {110} or {100} planes of a crystal in the usual perpendicular geometry (an external field H oriented in the plane of the plate and the light propagation vector $\mathbf{k}$ perpendicular to the plane of the plate). Helium–neon laser radiation of wavelength 0.63 μm was used in an optical cryostat with a superconducting magnetic system of the Helmholtz type (where the variation of the field in a volume of $3\times3\times3$ mm dimensions was less than 1%) generating fields up to 50 kOe. A sample was cooled in a stream of gaseous helium to the required temperature in the range 4.2–50 K. The MLB was measured by the polarization modulation method[10] with a sensitivity of $\Delta n = 10^{-8}$ The experimental error was mainly due to the error in the setting of a sample (setting of the field direction relative to a given crystallographic direction in the plane of a plate to within 1.5° and setting of the plate at right-angles to the propagation vector $\mathbf{k}$ of the incident light to within 0.5°) and typically was less than 5–7%. The vibration magnetometer method was used in the same ranges of fields and temperatures to measure the magnetization of gallate and aluminate single crystals along various directions. The error in the magnetization measurements was 5%.

MAGNETIZATION

We have mentioned already that the rare-earth ions occupy six inequivalent crystallographic sites in the garnet structure. Since the environment of a rare-earth ion is anisotropic, a magnetic field oriented along an arbitrary direction induces six different noncollinear and noncoplanar rare-earth magnetic moments $\mathbf{M}^{(r)}$. The magnetization of such a crystal per rare-earth ion is

$$M = {}^1/_6 H^{-1} \sum \mathbf{H}\mathbf{M}^{(r)}$$

and it depends on the direction of the magnetic field. In the subsequent analysis of the MLB we shall be interested in those cases when the field is directed along the symmetry axes of a crystal. We shall consider the types of magnetic structures which appear for $\mathbf{H}\|[111]$, $\mathbf{H}\|[110]$, and $\mathbf{H}\|[100]$.

$\mathbf{H}\|[111]$. In this case a double helical (spiral) magnetic structure is formed in the applied field. One helix represents the magnetic moments of ions at the sites 1, 3, and 5, whereas the other helix corresponds to the sites 2, 4, and 6. Inside each helix the moments are equal, and they lie in the $(1\bar{1}0)$, $(01\bar{1})$, and $(10\bar{1})$ planes at the same angles relative to the [111] axis.

$\mathbf{H}\|[110]$. The magnetic structure now consists of three magnetic subsystems. Two of them have different magnetic moments at the sites 1 and 2, which are directed along the [110] axis. The third subsystem is formed by equal magnetic moments at the sites 3, 4, 5, and 6. The moments at the sites 5 and 6 are mirror images, relative to the (001) plane, of the moments of the sites 3 and 4, respectively. The planes in which the moments at the sites 3 and 4 and also 5 and 6 are located pass through the [001] axis and are inclined relative to the (100) and (001) planes by the same angles measured in the direction toward the [110] axis.

TABLE 1. Orientations of local symmetry axes of c sites in garnet structure.

Axis	Site					
	1	2	3	4	5	6
e_x	110	$1\bar{1}0$	011	$01\bar{1}$	101	$\bar{1}01$
e_y	$\bar{1}10$	110	011	011	$10\bar{1}$	101
e_z	001	001	100	100	010	010

$\mathbf{H}\|[100]$. In this case there are two magnetic subsystems. One of them is formed by equal and collinear (with the direction of the field) moments at the sites 1 and 2. The other consists of the moments at the sites 3, 4, 5, and 6. The magnitudes of these moments are equal. The moments at the sites 3 and 4 lie in the (100) plane and are oriented at angles to the [001] axis which are the same in magnitude but have different signs. The moments at the sites 5 and 6, lying in the (010) plane, are oriented in a similar manner.

PERMITTIVITY TENSOR AND MAGNETIC LINEAR BIREFRINGENCE

In the interpretation of the experimental data on the MLB it is usual to employ the phenomonological approach based on the Akulov theory of even magnetic effects.[2,3] In this approach the permittivity tensor is represented by

$$\varepsilon_{ij}=\varepsilon_{ij}^{0}+\sum_{k}\beta_{ijk}M_{k}+\sum_{kl}\rho_{ijkl}M_{k}M_{l}, \qquad (1)$$

where M_k are the components of the magnetization vector. The observed phase shift due to the birefringence is defined as $\Phi = 2\pi\Delta n/\lambda$, where λ is the wavelength of light in vacuum and Δn is the difference between the refractive indices of light waves propagating at right-angles to the magnetic field (or magnetization) and polarized along and across the magnetic field. The relationship between the refractive indices (or refraction vectors) and the ε_{ij} tensor is governed in the optics of crystals by the Fresnel equation

$$\det|n^2\delta_{ij}-n_in_j-\varepsilon_{ij}|=0, \qquad (2)$$

which describes normal modes and the corresponding wave vectors $\mathbf{k} = \mathbf{n}\omega/c$ (or the refraction vectors $\mathbf{n}$).

This definition of the birefringence Φ suffers from certain limitations even in the case of cubic crystals.[3] It is valid only if the directions of the magnetic field $\mathbf{H}$ and of the wave vector $\mathbf{k}$ coincide with the principal axes of the optical indicatrix of a crystal. This situation occurs when the directions of the magnetic field (or of the magnetization) coincide with the symmetry axes of a crystal (i.e., with the [100], [111], and [110] axes).

In general, the principal axes of the optical indicatrix do not coincide with the crystal axes or with the direction of the magnetization, so that the quantity Δn in the formula for Φ could be understood to represent the difference between the refractive indices corresponding to two axes of the indicatrix when the wave vector is parallel to the third axis. It is this point that one has to bear in mind when speaking of the anisotropy of the MLB. It is understood to be the dependence of the difference Δn defined in this way on the direction of magnetization in a plane perpendicular to $\mathbf{k}$.

It should be pointed out that the MLB includes contributions from even (quadratic in $\mathbf{M}$) terms in the expansion of ε_{ij} and "gyrotropic" terms (linear in $\mathbf{M}$). Let us assume that, for example, $\mathbf{k}\|[001]$ and $\mathbf{M}\|[100]$. We have $\varepsilon_{xy} = \varepsilon_{xz} = 0$ and it follows from the Fresnel equation that there are two linearly polarized (along the x and y axes) modes with the refractive indices

$$n_x^2=\varepsilon^0+\delta\varepsilon_{xx}, \quad n_y^2=\varepsilon^0+\delta\varepsilon_{yy}+|\varepsilon_{yz}|^2/\varepsilon_{zz}.$$

In the majority of cases the "gyrotropic" contribution $|\varepsilon_{yz}|^2/\varepsilon_{zz}$ is small, so that $n_y^2 \approx \varepsilon^0 + \delta\varepsilon_{yy}$. This follows directly from a comparison of the experimental data for ε_{ii} and ε_{ij}, where $i \neq j$. The conclusion that the gyrotropic contribution to the birefringence can be ignored applies also in the case of more general situations. Therefore, in the birefringence studies we confine ourselves to the terms in the tensor ε_{ij} which are even in $\mathbf{M}$.

It follows from Eqs. (1) and (2) that the MLB depends only on the magnetization (or, more exactly, on its magnitude and orientation in a crystal) and it should remain constant if the magnetization is not changed. Moreover, application of Eqs. (1) and (2) to crystals of different symmetry allows us to derive certain relationships between the values of the MLB for various orientations of the vectors $\mathbf{H}$ and $\mathbf{k}$ (known as Akulov rules for the even effects). For example, in the case of cubic crystal it follows from Eqs. (1) and (2) that

$$\Delta n(\mathbf{H}\|[110],\ \mathbf{k}\|[001])=\Delta n(\mathbf{H}\|[111],\ \mathbf{k}\perp\mathbf{H}),$$
$$\Delta n(\mathbf{H}\|[110],\ \mathbf{k}\|[1\bar{1}0])=\tfrac{1}{2}\{\Delta n(\mathbf{H}\|[001],\ \mathbf{k}\perp\mathbf{H})+\Delta n(\mathbf{H}\|[111],\ \mathbf{k}\perp\mathbf{H})\}. \qquad (3)$$

However, this approach is not always valid and, strictly speaking, it should be applied only to S ions and even then the phenomenological theory based on the expansion of Eq. (1) describes correctly the field and temperature dependence of the MLB only at sufficiently high temperatures and in moderately strong fields]. In other cases, one should use different invariants permitted by the crystal symmetry in the description of the MLB; however, these invariants are ignored in the phenomenological theory because they are regarded as small.

We can show this by an analysis of the MLB for rare-earth paramagnets using a microscopic model which allows for the influence of the crystal and external magnetic fields on the electron structure and polarizability of the rare-earth ion. We shall allow for the real spatial symmetry of rare-earth garnets.

In the visible range of wavelengths the magnetooptic properties of rare-earth crystals are governed by allowed $4f^x \rightarrow 4f^{x-1}5d$ transitions in the rare-earth ions. The energy of these transitions in free trivalent rare-earth ions is $\sim 10^5$ cm^{-1} (see, for example, Ref. 11). The electrostatic interaction of a $5d$ electron with the $4f^{x-1}$ core ($\sim 10^4$ cm^{-1}) is-of the same order of magnitnde as the interaction between the crystal field and a $5d$ electron. The spin–orbit interaction of the $4f$ electrons is somewhat weaker. The spin–orbit interaction of a $5d$ electron and the influence of the crystal field on the $4f$ electrons are even weaker effects. In the long-wavelength wing of an allowed optical transition we can ignore, in the first approximation, the splitting of the levels of the $4f^{x-1}$ $5d$ configuration (for ions with $L \neq 0$). In this case the actual corrections to the polarizability tensor of a rare-earth ion with $L \neq 0$ can be represented in the form[12]

$$\delta\alpha_{ij}=a\langle Q_{ij}(J)\rangle,\ a=\tfrac{9}{7}\alpha(r_{fd}e)^2\omega_0/\hbar(\omega_0^2-\omega^2), \qquad (4)$$

where $r_{fd} = \langle 4f|r|5d\rangle$ is the radial integral; α is the Stevens parameter (see, for example, Ref. 13) of the ground multiplet of the ion; ω_0 is the average frequency of f–d transitions ($\hbar\omega_0 = 10^5$ cm^{-1}):

$$Q_{ij}(J)=\tfrac{1}{2}[J_iJ_j+J_jJ_i-\tfrac{2}{3}\delta_{ij}J(J+1)]$$

is the quadrupole moment operator of the electron shell of the ion. Along the rare-earth ion series the value of r_{fd} ranges from 0.29 Å Tm^{3+} to 0.44 Å for Pr^{3+} (Ref. 14). We shall ignore the frequency dependences of the MLB and concen-

trate our attention on the field and temperature dependences of this effect. We simply note that if $(\omega/\omega_0)^2 \ll 1$ then the MLB is frequency-independent see Eq. (4)]. We shall give the values of a (obtained in the limit $\omega \to 0$) for some of the rare-earth ions:

$a_{Pr} = -6 \cdot 10^{-27}$ cm^3, $a_{Tb} = -2.1 \cdot 10^{-27}$ cm^3,

$a_{Tm} = 1.2 \cdot 10^{-27}$ cm^3.

In the case of S ions (Gd^{3+}, Eu^{2+}) the expression for $\delta\alpha_{ij}$ is (Ref. 12)[1)]

$$\delta\alpha_{ij} = a_S \langle Q_{ij}(S) \rangle, \quad a_S = \frac{2\omega_0(3\omega^2+\omega_0^2)}{[\hbar(\omega^2-\omega_0^2)]^3} \lambda_{SL}^2 (r_{fd}e)^2, \qquad (5)$$

where λ_{SL} is the spin–orbit coupling constant. In the case of Gd^{3+} at frequencies in the range $(\omega/\omega_0)^2 \ll 1$ when $r_{fd} = 0.4$ Å (Ref. 14) and $\lambda_{SL} = 300$–400 cm^{-1}, we find that Eq. (5) yields $a_S = -(3.2\text{–}5.8) \times 10^{-30}$ cm^3. The contribution of the terms (4) and (5) to the permittivity of a crystal obtained in the standard Lorenz–Lorentz approximation is

$$\delta\varepsilon_{ij} = A \sum_r \langle Q_{ij}^{(r)} \rangle, \qquad (6)$$

where $A = \frac{2}{3}\pi a N [(n^2+2)/3]^2$ (n is the refractive index and N is the number of the rare-earth ions per unit volume) and the summation is carried out over all six inequivalent c sites occupied by the rare-earth ions.

MAGNETIC LINEAR BIREFRINGENCE OF GARNETS: THEORETICAL RELATIONSHIPS AND EXPERIMENTAL RESULTS

The field and temperature dependences of the MLB can be determined provided we first calculate the field and temperature dependences of the quadrupole moment $\langle Q_{ij}^{(r)} \rangle$. Naturally, these dependences will be different for rare-earth ions in different states. We shall consider this problem in greater detail for typical rare-earth ion spectra of garnets, beginning from the simplest orbital singlet (S ion) and then going over to more complex spectra which are formed largely under the influence of the local crystal environment.

S ion (Gd^{3+}). The ground-state multiplet of Gd^{3+} is $^8S_{7/2}$. Since in this state we have $L = 0$, we can ignore the influence of the crystal field on the splitting of the levels of the multiplet. Therefore, the energy levels of Gd^{3+} in an external field are described by $2\mu_B mH$, where $m = -7.2$, $-5/2, \ldots, 5/2$, and $7/2$.

The quadrupole moment operators Q_{ij}. can be expressed conveniently in terms of irreducible tensor operators $Y_l^m(S)$, with $S = 7/2$, using the relationships

$$Q_{zz} = {}^4/_3 ({}^\pi/_5)^{1/2} Y_2^0, \quad Q_{xx} = ({}^{2\pi}/_{15})^{1/2} (Y_2^2 + Y_2^{-2}) - {}^2/_3 ({}^\pi/_5)^{1/2} Y_2^0,$$
$$Q_{yy} = -{}^2/_3 ({}^\pi/_5)^{1/2} Y_2^0 - ({}^{2\pi}/_{15})^{1/2} (Y_2^2 + Y_2^{-2}),$$
$$Q_{xy} = -i ({}^{2\pi}/_{15})^{1/2} (Y_2^2 - Y_2^{-2}),$$
$$Q_{xz} = -({}^{2\pi}/_{15})^{1/2} (Y_2^1 - Y_2^{-1}), \quad Q_{yz} = i ({}^{2\pi}/_{15})^{1/2} (Y_2^1 + Y_2^{-1}). \qquad (7)$$

The average values $\langle Y_2^m(S) \rangle$ are in this case given by (see, for example, Ref. 13)

$$\langle Y_2^m(S) \rangle = ({}^{4\pi}/_5)^{1/2} Y_2^m(\xi) \langle Y_2^0(S') \rangle, \qquad (8)$$

where $\xi = \mathbf{M}/M$, $\mathbf{M}$ is the magnetization, and $\langle Y_2^0(S') \rangle$ is calculated in a coordinate system in which the z axis is directed along the magnetization. Using Eqs. (7) and (8), we find from Eq. (6) that

$$\delta\varepsilon_{ij} = \sum_{kl} \rho_{ijkl} \xi_k \xi_l. \qquad (9)$$

Since the MLB is described by $\Delta n = \frac{1}{2}(\delta\varepsilon_\parallel - \delta\varepsilon_\perp)$, its field and temperature dependence are governed by the field and temperature dependence of $\rho_{ijkl}(H,T)$, defined by the ratio $\langle Y_2^0 \rangle_T / \langle Y_2^0 \rangle_0$. In the classical limit ($S \gg 1$), we have

$$\Delta n(H, T) = \Delta n^0 \hat{I}_{5/2} \{L^{-1}(M(H, T)/2\mu_B S)\}, \qquad (10)$$

where $\hat{I}_{5/2}(x) = I_{5/2}(x)/I_{1/2}(x)$ is a reduced Bessel function and L^{-1} is the reciprocal of the Langevin function. At low temperatures or in strong fields, when only the two lowest energy levels of the Gd^{3+} ion are populated, it follows from Eq. (10) that the MLB is proportional to $M(H,T)^3$, whereas at high temperatures when the magnetization is weak, we have $\Delta n \propto M(H,T)^2$. Saturation of the MLB (Δn_H) of $Gd_3Ga_5O_{12}$ occurs when the magnetization is sat-

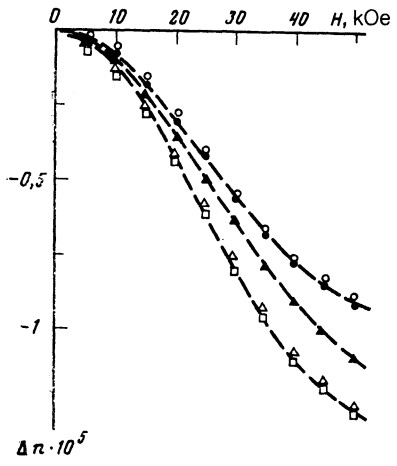

FIG. 1. Field dependence of the magnetic linear birefringence of $Gd_3Ga_5O_{12}$ at 4.2 K. The symbols are the experimental data obtained for $\mathbf{k} \perp (110)$: ●) $\mathbf{H} \| [001]$, ▲) $\mathbf{H} \| [\bar{1}10]$, □) $\mathbf{H} \| [\bar{1}11]$; $\mathbf{k} \perp (100)$; ○) $\mathbf{H} \| [001]$, △) $\mathbf{H} \| [011]$. The dashed curves are calculated using Eq. (10).

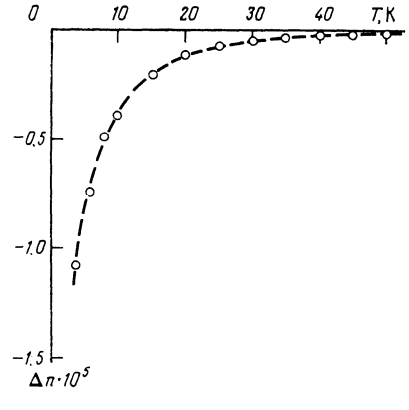

FIG. 2. Temperature dependence of the magnetic linear birefringence of $Gd_3Ga_5O_{12}$ in a field 40 kOe obtained for $\mathbf{k} \perp (110)$ and $\mathbf{H} \| [\bar{1}11]$. The circles are the experimental results and the dashed curve is calculated using Eq. (10).

urated paramagnetically. The value of Δn_H calculated using Eqs. (5) and (6) with the parameters $r_{fd} = 0.4$ Å (Ref. 14), $\lambda_{SL} = 300$–$400\ \text{cm}^{-1}$, $n \approx 2$, and $N = 1.27 \times 10^{22}\ \text{cm}^{-3}$, is $\Delta n_H = -(1.1$–$1.9) \times 10^{-5}$ and is of the same order of magnitude as the experimental data reported below.

It therefore follows from our calculations that the magnetic-field dependence of the MLB of compounds of gadolinium and other S ions is the same as that predicted by the phenomenological theory above-mentioned see [Eq. (9)]. A new feature of the microscopic theory is that it yields the field and temperature dependences very different from those proposed earlier and these agree with the earlier predictions only in the limit of weak magnetization. It should be pointed out that Eq. (10) is fully analogous to the formula which describes the field and temperature dependences of the magnetostriction obtained by Callen and Callen.[15]

Figure 1 shows the experimental field dependence of the MLB of $Gd_3Ga_5O_{12}$ obtained for various directions at 4.2 K and the theoretical dependence plotted on the basis of Eq. (10) using our own measured values of the magnetization of $Gd_3Ga_5O_{12}$. We can see that the agreement between the experimental and theoretical dependences is good. The experimental dependence $\Delta n(T)$ is plotted in Fig. 2 and once again it is described well by Eq. (10).

Therefore, our experiments and a microscopic theoretical analysis confirm that in the case of rare-earth compounds containing S ions the orientational dependence of the MLB can be described by the phenomenological theory in which the temperature and field dependences of the coefficients are given by Eq. (10).

Ions with a singlet ground state (Eu^{3+}, Tm^{3+}, Pr^{3+}). The Eu^{3+} ion has a singlet ground state with $J = 0$. The first excited multiplet $^7F_1$ is separated by an energy of $350\ \text{cm}^{-1}$ from the ground multiplet.[16] In the case of Tm^{3+} (ground multiplet $^3H_6$) the singlet ground state appears in a garnet because of lifting of the degeneracy of the ground multiplet in a crystal field of D_2 symmetry. The first excited level lies $35\ \text{cm}^{-1}$ above the ground state in the case of $Tm_3Al_4O_{12}$ and at $63\ \text{cm}^{-1}$ in the case of $Tm_3Ga_5O_{12}$ (Refs. 5 and 6). A similar structure of the splitting of the ground multiplet $^3H_4$ of Pr^{3+} by the crystal field is observed for $Pr_3Ga_5O_{12}$.

It is known that a nonzero magnetic moment of an ion with a singlet ground state appears only if excited states are admixed, in accordance with perturbation theory, to the ground state (Van Vleck mechanism). This also applies to the quadrupole moment. The only difference is that a field-dependent quadrupole moment appears only in perturbation theory which is of second order in the ratio of the Zeeman energy to the energy of the crystal field, so that

$$\delta\langle Q_{ij}\rangle \sim (\mu_B H/W)^2,$$

where W is a characteristic energy interval separating the ground singlet from excited levels.

A more detailed quantum-mechanical calculation based on perturbation theory gives

$$\delta\langle Q_{ij}\rangle = \sum b_{ijkl} H_k H_l, \qquad (11)$$

where the fourth-rank tensor b_{ijkl} can be called the quadrupole susceptibility tensor. The symmetry of this tensor is now governed by the point group D_2. The expressions for the components b_{ijkl} are given in the Appendix. At low-temperatures ($T \ll W$) the tensor b_{ijkl} is independent of temperature. Substituting Eq. (11) into the expression for the permittivity (6), we can now calculate the MLB. It is found that the orientational dependence of Δn is exactly the same as that predicted by the phenomenological theory. In particular, the rule of the even effects is obeyed. The expressions for Δn in the cases when $\mathbf{H} \| [001]$ and $\mathbf{H} \| [111]$ are as follows:

$$\Delta n(\mathbf{H} \| [001],\ \mathbf{k} \perp \mathbf{H}) = {}^3/_4 AH^2 (2b_{zzzz} + b_{zzxx} + b_{zzyy} + 4b_{xyxy}),$$

$$\Delta n(\mathbf{H} \| [111],\ \mathbf{k} \perp \mathbf{H}) = {}^1/_2 AH^2 (b_{zzxx} + b_{zzyy} + 2b_{xxxx} + 2b_{yyyy} + 4b_{zyzy} + 4b_{zxzx}). \qquad (12)$$

This is precisely the experimentally observed behavior of the MLB of $Tm_3Ga_5O_{12}$ (Figs. 3 and 4). In particular, in the investigated range of magnetic fields the value of Δn for this garnet is very accurately proportional to H^2 at 4.2 K. Below approximately 20 K the MLB of $Tm_3Ga_5O_{12}$ is independent of temperature. At higher temperatures the effects due to thermal filling of the excited levels become important and the MLB varies with temperature. This interpretation is supported by the results of our measurements of the MLB of thulium aluminate. Since in this compound the excited levels are much closer to the ground state than in thulium gallate, the temperature interval where $\Delta n(T) = \text{const}$ is much narrower and the value of Δn is larger (Fig. 4). It is clear from Fig. 4 that the MLB of thulium garnet varies with temperature but it is not proportional to the square of the magnetization; this is again in agreement with the theory.

The MLB of $Eu_3Ga_5O_{12}$ is very small (it is less than 2×10^{-6} in the highest field used in our study). This is probably due to the fact that the energy interval separating the ground singlet from excited levels is large.

Ions with a doublet ground state (Sm^{3+}, Nd^{3+}, Dy^{3+}, Er^{3+}, and Yb^{3+}). The crystal field in garnets splits the

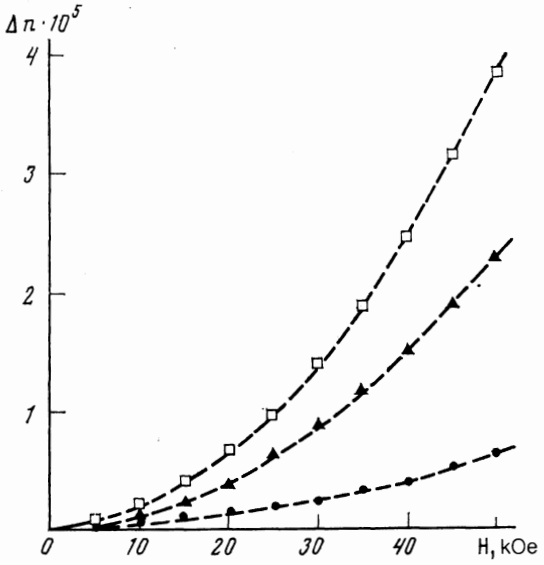

FIG. 3. Field dependence of the magnetic linear birefringence of $Tm_3Ga_5O_{12}$ at 4.2 K. The symbols are the experimental results (the notation is the same as in Fig. 1). The dashed curves represent $\Delta n \propto H^2$.

ground multiplet of the Kramers rare-earth ions into doublets. If we consider only the ground doublet (assuming that it is isolated), it then follows from the calculations of Ref. 12 that the average quadrupole moment of the rare-earth ion is independent of the field. Consequently, in this approximation the MLB vanishes. (The effective spin of an isolated doublet is $S_{\text{eff}} = 1/2$. We recall that in this state the single-ion magnetostriction and magnetic anisotropy also vanish.) This property of the MLB can be compared with vanishing of the magnetic moment of an isolated singlet: in the singlet state the magnetic moment, which is a first-rank tensor, vanishes; for a doublet the change in the quadrupole moment, which is a second-rank tensor with zero trace, again vanishes.

We can calculate the change in the quadrupole moment of a rare-earth ion with a doublet ground state if we use perturbation theory to allow for the admixture of excited doublets to the ground doublet (Van Vleck mechanism). As in the case of the singlet state, the perturbation is the Zeeman energy.

In the first approximation of perturbation theory we have

$$\delta\langle Q_{ij}\rangle \sim M(H, T)\ \mu_B H/W,$$

where M is the magnetization of the rare-earth ion. In this case the field dependence of the MLB differs qualitatively from the corresponding dependence for an S ion: Δn is governed by the product of the magnetization and the field (and not only by the magnetization alone), so that in strong fields when the magnetization ceases to depend on the field (paramagnetic saturation) the value of the MLB varies linearly with the field. The MLB of garnets with a Kramers rare-earth ion should be greater in the doublet ground state (other conditions being equal) than in the case of the singlet ground state, since it is governed by the first order of the small parameter $\mu_B H/W$.

More detailed calculations of the change in the quadrupole moment in a magnetic field yield

$$\delta\langle Q_{ij}\rangle = \sum_{kl} G_{ijkl} H_k M_l / g_l, \tag{13}$$

where M_l is the component of the magnetization of the rare-earth ion due to the splitting of the ground-doublet levels. In terms of the local symmetry axes of the rth site, we find that

$$M_l^{(r)} = {}^1/_2 \Delta_r^{-1} g_l^2 H_l^{(r)} \operatorname{th}(\Delta_r/2T), \quad \Delta_r = \left[\sum_k (g_k H_k^{(r)})^2\right]^{1/2}, \tag{14}$$

where g_l are the components of the g tensor of the ground doublet and G_{ijkl} are the effective coefficients, for which explicit expressions are given in Ref. 12. The tensor G_{ijk} is the main characteristic of a Kramers ion governing the appearance of the magnetic corrections to the quadrupole moment. It can be regarded as the quadrupole susceptibility tensor of a Kramers ion.

It is convenient to calculate the MLB using a coordinate system characterized by $\mathbf{z}\|\mathbf{k}$. We shall first consider the permittivity tensor. It follows from Eqs. (6) and (13) that in the laboratory coordinate system defined by orthonormalized triplet vectors γ_1, γ_2, and γ_3 (in the subsequent calculations of the MLB we shall assume that $\gamma_1\|\mathbf{H}$, $\gamma_3\|\mathbf{k}$, $\gamma_2 = [\gamma_3\gamma_1]$), the tensor $\delta\varepsilon_{ij}$ is

$$\delta\varepsilon_{ij} = A\sum_{rkq} T_{ik}^{(r)} T_{jq}^{r)}\, \delta\langle Q_{kq}^{(r)}\rangle = A\sum_{rkq} \{T_{ik}^{(r)} T_{jq}^{r)}\, g_{kkqq} H_q^{(r)} M_q^{(r)}$$
$$+ (T_{ik}^{(r)} T_{jq}^{(r)} + T_{jk}^{(r)} T_{iq}^{(r)})\, g_{kqkq} H_k^{(r)} M_q^{(r)} (1-\delta_{kq})\},$$
$$T_{ik}^{(r)} = \gamma_i e_k^r, \quad g_{ijkl} = G_{ijkl}/g_l. \tag{15}$$

It follows from Eq. (15) that the permittivity of rare-earth garnets is governed generally not by the resultant magnetization, but by a combination of the magnetizations of the rare-earth ions at various inequivalent sites. Using the system of equations (15) we find that the expressions for the MLB of garnets with rare-earth ions that have a doublet ground state can be represented as follows (for different orientations of the field and of the light propagation vector):

$$\Delta n(\mathbf{H}\|[001],\ \mathbf{k}\perp\mathbf{H}) = H\{C_1(M^{(1)}+M^{(2)})$$
$$+C_2(M^{(3)}+M^{(4)}+M^{(5)}+M^{(6)})\}_{001},$$
$$\Delta n(\mathbf{H}\|[111],\ \mathbf{k}\perp\mathbf{H}) = H\{B_1(M^{(1)}+M^{(3)}+M^{(5)})$$
$$+B_2(M^{(2)}+M^{(4)}+M^{(6)})\}_{111},$$
$$\Delta n(\mathbf{H}\|[110],\ p) = H\{D_1^p M^{(1)}+D_2^p M^{(2)}$$
$$+D_3^p(M^{(3)}+M^{(4)}+M^{(5)}+M^{(6)})\}_{110}, \tag{16}$$

where the coefficients $C_{1,2}$, $B_{1,2}$ and $D_{1,2,3}^p$ are given in the Appendix [see Eq. (A2)]; the index $p = 1$ corresponds to $\mathbf{k}\|[001]$, whereas the index $p = 2$ corresponds to $\mathbf{k}\|[110]$. The magnetic moment vectors of the rare-earth ions, defined by Eq. (14), occur in Eq. (16) and can be calculated for the appropriate directions of the magnetic field $\mathbf{H}$.

It follows from Eq. (16) that the field and temperature

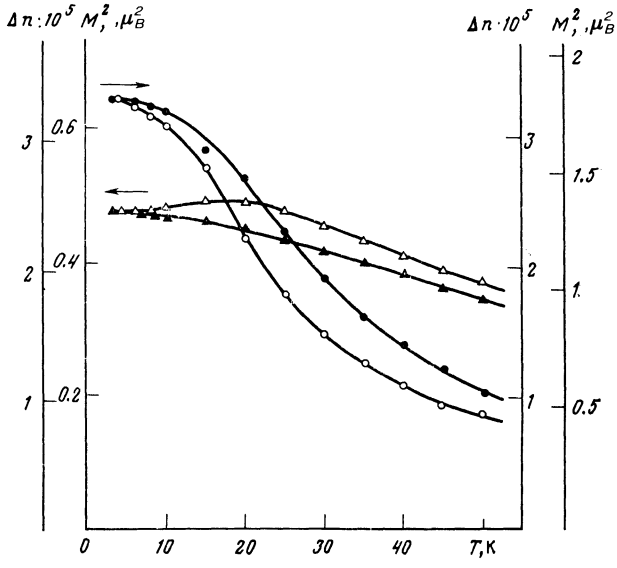

FIG. 4. Temperature dependence of the magnetic linear birefringence obtained for $\mathbf{k}\perp(110)$ and $\mathbf{H}\|[\bar{1}11]$ (open symbols) and of the square of the magnetization (black symbols) in a field of 40 kOe applied to $Tm_3Ga_5O_{12}$ (circles) and $Tm_3Al_5O_{12}$ (triangles).

dependences of the MLB are in this case very different from those proposed in the phenomenological theory. In particular, the MLB now exhibits a strong field dependence and it rises linearly with the field in strong fields, when the magnetization of the rare-earth sublattices (and, consequently, the total magnetization of the garnet) is completely or almost completely saturated. Moreover, the dependence of the MLB on the magnetic field orientation is more complicated than in the phenomenological approach. If we use Eqs. (14) and (A2), Eq. (16) shows that the even effects rule of Eq. (3) is now satisfied only in weak fields or at high temperatures (when $gH \ll W$) or in the case of a weak anisotropy of the g tensor of the ground doublet.

It is worth noting that, generally speaking, the MLB depends on linear combinations of the sublattice magnetizations and not on their sum, which is the total magnetization of a crystal. The MLB can be expressed in terms of the total magnetization only in two limiting cases: rare-earth ions with an isotropic g tensor and very strongly anisotropic Ising rare-earth ions characterized by $g_z \ll g_x, g_y$.

In both these limiting cases the field and temperature dependence of the MLB is simple:

$$\Delta n(H, T) = cHM(H, T). \tag{17}$$

However, in the case of garnets with isotropic rare-earth ions the magnetization due to the ground doublet of the rare-earth ion is independent of the orientation of the field, so that in the case of garnets with these ions the even effects rule of Eq. (3) should be satisfied, whereas in the case of garnets with Ising ions the magnetization due to the ground doublet changes greatly as a result of a change in the orientation of the field and the even effects rule is not obeyed.

We demonstrated earlier[17] that the isotropic approximation can be used to describe the MLB of the gallate $Yb_3Ga_5O_{12}$ in which the components of the g tensor of the Yb^{3+} ion are similar ($g_x = 3.60$, $g_y = 3.78$, $g_z = 2.85$—see Ref. 18).

We shall consider $Dy_3Al_5O_{12}$ as an example of the MLB in garnets with Ising ions. The g tensor of Dy^{3+} in this garnet is strongly anisotropic ($g_x = 0.73$, $g_y = 0.40$, $g_z = 18.2$).[19] Then, the general formula (16) yields the MLB:

$$\begin{aligned}
&\Delta n(\mathbf{H}\|[001],\ \mathbf{k}\perp H) = b_1 H M_{001},\\
&\Delta n(\mathbf{H}\|[111],\ \mathbf{k}\perp\mathbf{H}) = b_2 H M_{111},\\
&\Delta n(\mathbf{H}\|[110],\ \mathbf{k}\|[001]) = b_2 H M_{110},\\
&\Delta n(\mathbf{H}\|[110],\ \mathbf{k}\|[1\bar{1}0]) = {}^1/_2(b_1+b_2) H M_{110},
\end{aligned} \tag{18}$$

where the magnetizations due to the ground doublet of the rare-earth ion are (per ion)

$$\begin{aligned}
&M_{001} = {}^1/_6 g_z \operatorname{th}(g_z H/2T), \quad M_{110} = (2^{1/2}/6)\, g_z \operatorname{th}(g_z H/2^{1/2}T),\\
&M_{111} = (g_z/2\cdot 3^{1/2}) \operatorname{th}(g_z H/2\cdot 3^{1/2}T),
\end{aligned} \tag{19}$$

and b_1 and b_2 are constants. Equations (18) and (19) clearly demonstrate breakdown of the even effects rule in the case of $Dy_3Al_5O_{12}$ when $g_z H \geqslant T$.

Figure 5 shows the experimental field dependences of the MLB obtained for $Dy_3Al_5O_{12}$ at 4.2 K. We can see that

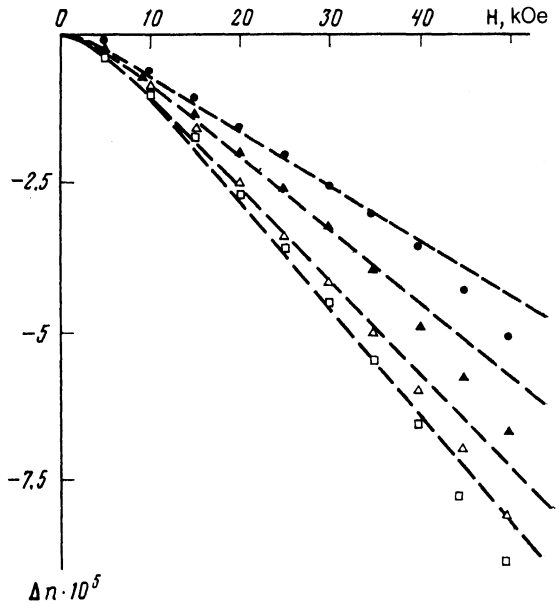

FIG. 5. Field dependence of the magnetic linear birefringence of $Dy_3Al_5O_{12}$ at 4.2 K. The symbols represent the experimental results (the notation is the same as in Fig. 1). The dashed curves are calculated using Eq. (17).

in the case of this garnet the application of strong fields such that the magnetization tends to saturation (Fig. 6) does not saturate the MLB (in contrast to $Gd_3Ga_5O_{12}$, see Fig. 1). Figure 5 includes also theoretical dependences $\Delta n(H)$ calculated on the basis of Eq. (17) using the experimental values of the magnetization the ground doublet magnetization was deduced from the experimental dependences $M(H)$ by subtracting the Van Vleck contribution deduced from the slope of $M(H)$ in strong fields]. We can see that Eq. (17) describes satisfactorily the experimental data obtained in fields below 35–40 kOe, whereas the experimental dependences $\Delta n(H)$ obtained in stronger fields vary more rapidly with the field than predicted theoretically. This is due to the fact that in strong fields the Zeeman splitting of the ground doublet is no longer small compared with the energy separa-

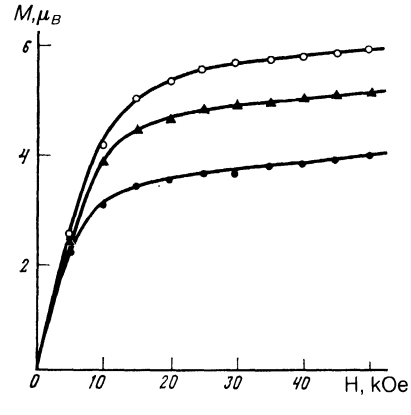

FIG. 6. Field dependence of the magnetization of $Dy_3Al_5O_{12}$ at 4.2 K: ●) $\mathbf{H}\|[100]$; ▲) $\mathbf{H}\|[110]$; ○) $\mathbf{H}\|[111]$.

tion between the ground and first excited doublets, so that Eq. (17) for the MLB must include the next term of the expansion proportional to H^2. Inclusion of this term makes it possible to describe the experimental results completely. Figure 7 gives the temperature dependence of the MLB for $Dy_3Al_5O_{12}$. We can see that it is described well by Eq. (17).

The MLBs of other garnets with Kramers rare-earth ions ($Nd_3Ga_5O_{12}$, $Er_3Al_5O_{12}$, etc.) exhibit field and temperature dependences which are qualitatively similar to those of the MLB of $Dy_3Al_5O_{12}$.

Ions with a quasidoublet ground state (Tb^{3+}, Ho^{3+}). The ground state of some non-Kramers rare-earth ions, such as Tb^{3+} and Ho^{3+}, in garnet crystals are two closely spaced singlets (quasidoublet) separated from higher levels by a large energy gap. For example, in the case of Tb^{3+} in $Tb_3Ga_5O_{12}$ and $Tb_3Al_5O_{12}$ the ground state is a quasidoublet and the separation between its levels is 2–3 cm^{-1}, whereas the nearest excited level is separated by 40 cm^{-1} in the gallate and 61 cm^{-1} in the aluminate.[6,20] Approximately the same parameters are exhibited by the ground quasidoublet of the Ho^{3+} ion in garnets.[6,21] We shall now consider the MLB for this case.

Let us assume that $|A\rangle$ and $|B\rangle$ are the wave functions of the state of a rare-earth ion forming a quasidoublet in zero external field. Since these functions are not Kramers-conjugates of one another, a change in the average quadrupole moment of the rare-earth ion in the applied magnetic field is then predicted in the zeroth approximation of perturbation theory. Therefore, the MLB of a non-Kramers quasidoublet appears in principle even if we make no allowance for an admixture of higher levels.

It is known[22] that rare-earth ions with a quasidoublet ground state are of the Ising type. In garnets (when the symmetry of the environment of the rare-earth ion is D_2) the magnetization axis of the rare-earth ion coincides with some local symmetry axis e_α^r. In the case of Tb^{3+} and Ho^{3+} in garnet gallates and aluminates, the magnetization axis is oriented along e_z^r. In this case the contribution made to the MLB by the zeroth-order terms is

$$\Delta n_0(\mathbf{H}\|[001],\mathbf{k}\perp\mathbf{H}) = {}^3/_4 A\Delta_0(\chi(H)-\chi(0))C_{zz},$$
$$\Delta n_0(\mathbf{H}\|[111],\ \mathbf{k}\perp\mathbf{H}) = \Delta n_0(\mathbf{H}\|[110],\ \mathbf{k}\|[001]) = 0,$$
$$\Delta n_0(\mathbf{H}\|[110],\ \mathbf{k}\|[1\bar{1}0]) = {}^3/_4 A\Delta_0(\chi(H/\sqrt{2})-\chi(0))C_{zz}, \quad (20)$$

where χ represents the magnetic susceptibility due to the ground quasidoublet:

$$\chi(H) = \Delta^{-1}(H)\,\mathrm{th}(\Delta(H)/2T),\quad \Delta(H) = (\Delta_0^2+\mu^2H^2)^{1/2},$$
$$\mu = 2\mu_B g_J|\langle A|J_z|B\rangle|,\quad C_{zz} = \langle A|Q_{zz}|A\rangle - \langle B|Q_{zz}|B\rangle,$$

where Δ_0 is the separation between the quasidoublet levels in zero field. It should be noted that this contribution to the MLB has a characteristic field dependence: n_0 tends to saturation in strong fields when $\mu H \gg T$ and $\Delta n_0 \approx -\frac{3}{4}AC_{zz}\tanh(\Delta_0/2T]$.

The zeroth-order contribution is frequently insufficient to account for the experimental data on the MLB of crystals with rare-earth ions characterized by a quasidoublet ground state. Clearly, this is due to the similarity of the wave functions of the quasidoublet levels and due to the smallness of Δ_0, so that a non-Kramers quasidoublet can be regarded approximately as a doublet. We need then to allow for the influence of excited states on the quadrupole moment of the rare-earth ion in accordance with perturbation theory. Inclusion of corrections allowing for this influence makes a contribution to the MLB which is analogous to that calculated for garnets with Kramers rare-earth ions and a doublet ground state.

By way of example, we shall consider the MLB of $Tb_3Al_5O_{12}$. Using the data on the wave functions of the Tb^{3+} ion in this garnet,[23] we find that $|C_{zz}/\langle A|Q_{zz}|A\rangle| \approx 4\times10^{-3}$, i.e., the main contribution to the MLB comes from the mechanism of mixing of the ground states. This is confirmed by the experimental data on the field dependence of the MLB of terbium aluminate at 4.2 K (Fig. 8), which is at least qualitatively described by Eq. (17). This is demonstrated particularly by the characteristic rise of the MLB with the field when the field is sufficiently strong for paramagnetic saturation of the magnetization. As in the case of $Dy_3Al_5O_{12}$, in strong fields the contribution

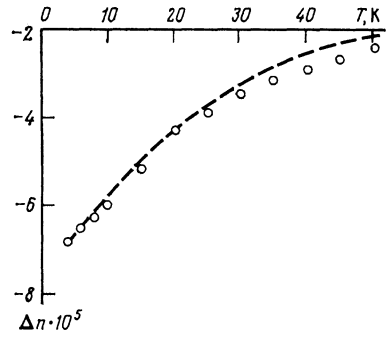

FIG. 7. Temperature dependence of the magnetic linear birefringence of $Dy_3Al_5O_{12}$ obtained in a field 40 keV for $\mathbf{k}\perp(110)$ and $\mathbf{H}\|[\bar{1}11]$. The circles are the experimental results and the dashed curve is calculated using Eq. (17).

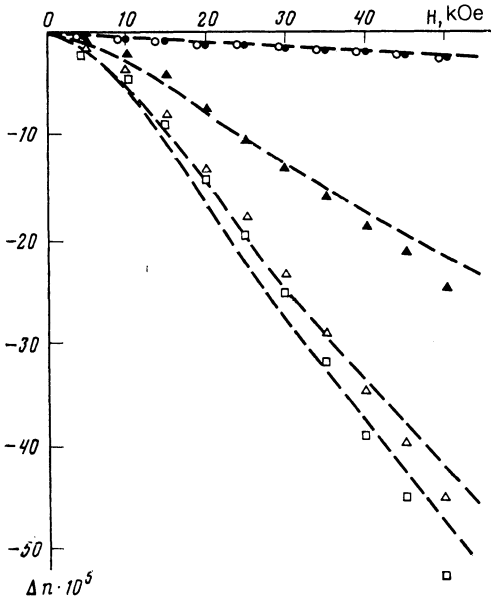

FIG. 8. Field dependence of the magnetic linear birefringence of $Tb_3Al_5O_{12}$ at 4.2 K. The symbols are the experimental data (the notation is the same as in Fig. 1). The dashed curves are calculated using Eq. (17).

made to the MLB by the term proportional to H^2 becomes important. We obtained similar results for $Tb_3Ga_5O_{12}$, $Ho_3Ga_5O_{12}$, and $Ho_3Al_5O_{12}$.

SPACE GROUP O_h^{10}, MAGNETIC MODES, AND SYMMETRY OF THE PERMITTIVITY TENSOR

A rare-earth garnet differs from a classical paramagnet because it contains six types of magnetic ions (six sublattices) which are magnetized in different ways by an external field. A complete description of the magnetic state of such a paramagnet cannot be provided by just the magnetization of the material, but it is necessary to determine the magnetic moments of the six sublattices, i.e., to find a vector in a space with 18 dimensions. This generalized vector transforms in accordance with the reducible representation of the group O_h^{10}, which can be expanded in terms of magnetic modes $\mathfrak{M}_v^{\mu j}$, each of which transforms in accordance with one of the irreducible representations τ^3, τ^5, τ_1^7, τ_2^7, τ_1^9, τ_2^9, and τ_3^9 (the notation is the same as in Kovalev's handbook[24]). A magnetic field of arbitrary orientation generally induces all these modes. Each of the modes normally contributes to the MLB and to other even magnetooptic effects.

From this point of view, we can represent Eq. (15) for the tensor $\delta\varepsilon_{ij}$ as follows:

$$\delta\varepsilon_i^{\mu} = \sum K_{\lambda j}^{\mu} (\tau(H) \otimes \tau(\mathfrak{M}))_{ij}^{\mu\lambda}, \qquad (21)$$

where $(\tau(H) \otimes \tau(\mathfrak{M}))_{ij}^{\mu\lambda}$ are the basis functions of the tensor representation, representing linear combinations of the products $H_k \mathfrak{M}_l^{\lambda j}$. For example, we have

$$(\tau(H) \otimes \tau(\mathfrak{M}))_{ij}^{19} = \sum_{\nu=1}^{3} H_\nu \mathfrak{M}_\nu^{9j}, \quad j=1,2,3.$$

In Eq. (21), $\mu = 1, 6, 7$; $\lambda = 3, 6, 7, 9$; i is the dimensionality of the representation τ^μ; j is the multiplicity of the representation τ^λ; $K_{\lambda j}^{\mu}$ are linear combinations of the constants G_{ijkl} (there is a total of 15 such combinations);

$$\varepsilon_1^1 = \varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz}, \quad \varepsilon_1^5 = {}^1/_2(\varepsilon_{xx}-\varepsilon_{yy}), \quad \varepsilon_2^5 = (3^{1/2}/2)(\varepsilon_{zz}-{}^1/_3\varepsilon_1^1),$$
$$\varepsilon_1^7 = \varepsilon_{yz}, \quad \varepsilon_2^7 = \varepsilon_{xz}, \quad \varepsilon_3^7 = \varepsilon_{xy}.$$

It therefore follows that the permittivity tensor is generally determined by the space (not point) symmetry of a crystal and it includes a larger number of the effective constants $k_{\lambda_j}^{\mu}$ than that obtained from the phenomenological theory utilizing the point symmetry of the crystal. Transition to the classical description of the even magnetooptic effects occurs at high temperatures T (or in weak fields) when $gH \ll T$. Then, in the approximation linear in $gH/T \ll 1$, there are only three nonzero modes $\mathfrak{M}^{9j}$:

$$\mathfrak{M}_i^{91} = g_z H_i/T, \quad \mathfrak{M}_i^{92} = {}^1/_2(g_x+g_y)H_i/T, \quad \mathfrak{M}_i^{93} = {}^1/_2(g_y-g_x)H_i/T.$$

Then, Eq. (21) assumes the following simple form (it is described by three constants):

$$\varepsilon_1^1 = c^{(1)}H^2/T, \quad \varepsilon_1^5 = {}^1/_2 c^{(5)}(H_x^2-H_y^2)/T,$$

$$\varepsilon_2^5 = (3^{1/2}/2)\,c^{(5)}(H_z^2-{}^1/_3H^2)T,$$

$$\varepsilon_1^7 = c^{(7)}H_zH_y/T, \quad \varepsilon_2^7 = c^{(7)}H_xH_z/T, \quad \varepsilon_3^7 = c^{(7)}H_xH_y/T,$$

where

$$c^{(\mu)} = g_z K_{91}^{\mu} + {}^1/_2(g_x+g_y)K_{92}^{\mu} + {}^1/_2(g_y-g_x)K_{93}^{\mu}, \quad \mu = 1, 5, 7.$$

CONCLUSIONS

We have thus demonstrated theoretically and experimentally that the field, temperature, and angular dependences of the MLB are influenced strongly by the nature of the orbital degeneracy of the magnetic ion. When the orbital momentum of the ground state of the ion vanishes (or is frozen) the behavior of the MLB is generally described satisfactorily by the formulas of the phenomenological theory put forward by Akulov. A new result of the present treatment is a different dependence of the MLB on the applied magnetic field and temperature.

If the orbital momentum in the ground does not vanish, the behavior of the MLB is very different. The magnitude of the MLB is then greater than for $L = 0$ (when other conditions are the same).

In the case of ions with a singlet ground the MLB depends quadratically on the field, is not affected by variation of temperature (in the limit $T \to 0$), and obeys the same orientational dependence predicted by the phenomenological theory. There is a remarkably strong dependence of the magnitude of the effect on the energy W separating the ground state from excited levels. This confirms the general conclusion that the MLB of rare-earth compounds depends more strongly on the nature of the excited states if $L \neq 0$.

The MLB of crystals containing Kramers ions with a doublet ground state exhibits a strong field dependence in the magnetization saturation region. This is due to the fact that a change in the polarizability of these ions in a magnetic field is due to mixing of excited states with the ground doublet, representing a characteristic analog of the Van Vleck susceptibility for the even magnetic effects.

Crystals with non-Kramers ions with two closely spaced singlets (quasidoublet) in the ground state, separated by a large energy gap from excited levels, exhibit similar behavior of the MLB.

When the degree of degeneracy is higher, i.e., when the effective spin of the ground state is $S_{\text{eff}} > 1/2$, the field dependences of the MLB (and of other even effects) should become saturated in strong fields in the range where the magnetization is saturated. This does not apply to paramagnetic garnets.

There is a close analogy between the behavior of the MLB and magnetostriction. This is due to the fact that both effects are governed by the same quantity, namely by the field-induced change of the quadrupole moments of the rare-earth ions. However, in describing the magnetostriction (and other magnetoelastic effects) it is generally essential to have a larger number of phenomenological parameters than the number used to describe the MLB (at least in the approximation adopted in the present study).

It should be stressed that the symmetry of the magnetic corrections to the permittivity tensor of rare-earth garnets is not determined by the point symmetry group of a crystal, but by the space group, i.e., it is very important to allow for the inequivalent positions of the rare-earth ions in the garnet crystal structure. There is a complete analogy with the symmetry properties of the magnetoelastic energy.[13] Naturally, this conclusion does not apply just to rare-earth garnets but is more general.

We considered the MLB in paramagnetic rare-earth garnets. Generalization of the theory to magnetically or-

dered rare-earth ion garnets can be made as usual employing the self-consistent field approximation: the external field in all the formulas should be replaced by the effective field which includes the external and exchange fields created by the iron sublattices.

The main conclusions reached above can be extended in a natural manner to the magnetic linear dichroism. This can be done using the Kramers–Kronig dispersion formulas or calculating directly the polarizability of a rare-earth ion from the Kramers–Heisenberg formula, as is done in Ref. 12.

The authors are grateful to B.V. Mill' who grew the majority of the investigated crystals from a molten solution, and to A.A. Kaminskiĭ, who supplied garnet crystals synthesized by the Czochralski method.

APPENDIX

I. A singlet is described by

$$b_{ijkl}=(\mu_B g_J)^2\Big\{\sum_{mpn}[2\langle n|Q_{ij}|m\rangle\langle m|J_k|p\rangle\langle p|J_l|n\rangle + \langle n|J_k|m\rangle\langle m|Q_{ij}|p\rangle\langle p|J_l|n\rangle][(E_m-E_n)(E_p-E_n)]^{-1}\rho_n - \sum_{mn}\langle n|Q_{ij}|n\rangle|\langle n|J_k|m\rangle|^2(E_m-E_n)^{-2}\rho_n\delta_{kl}\Big\},$$

$$\rho_n=\Big[\sum_k \exp(-E_k/T)\Big]^{-1}\exp(-E_n/T), \qquad \text{(A1)}$$

where E_m is an energy level of a singlet $|m\rangle$.

II. A doublet is described by

$$C_1={}^3/_4 g_{zzzz}A,$$

$$C_2={}^3/_8 A(g_x^2+g_y^2)^{-1}[g_x^2(2g_{yxyx}-g_{zxxx})+(x\rightleftarrows y)],$$

$$B_1={}^1/_4 A[(g_{xxzz}-g_{yyzz}+4g_{xzxz})g_z^2 + 2(g_{xxxx}-g_{yyxx}+2g_{zxzx})g_x^2](g_z^2+2g_x^2)^{-1},$$

$$B_2=B_1(x\rightleftarrows y),$$

$$D_1^1={}^1/_2 A(g_{xxxx}-g_{yyxx}), \quad D_2^1=D_1^1(x\rightleftarrows y),$$

$$D_3^1=A[(g_{yzyz}+g_{xzxz})g_z^2+g_{zyzy}g_y^2+g_{zxzx}g_x^2](2g_z^2+g_x^2+g_y^2)^{-1},$$

$$D_1^2={}^1/_2 A(g_{xxxx}-g_{zzxx}), \quad D_2^2=D_1^2(x\rightleftarrows y),$$

$$D_3^2={}^1/_8 A\{[(2g_{yzyz}+2g_{xzxz}+3g_{zzzz})g_z^2+(4g_{zyzy}+6g_{xyxy}+3g_{zzyy})g_y^2](2g_z^2+g_x^2+g_y^2)^{-1}+(x\rightleftarrows y)\}. \qquad \text{(A2)}$$

[1)]In the case of the Fe^{3+} ions (d–p transitions) the quantity a_S in Eq. (5) should be replaced with $a_S' = (2/3)a_S$.

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