Theoretical and experimental investigations of the magnetostriction of paramagnetic rare-earth garnets $Tb_3Ga_5O_{12}$ and $Dy_3Al_5O_{12}$

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The behavior of the magnetostriction of rare-earth paramagnets with garnet structure is investigated theoretically and experimentally. It is shown that, owing to the presence of low-symmetry nonequivalent sites occupied by the rare-earth ions in the garnet lattice, the set of invariants that describe the temperature, field, and angular dependences of the magnetostriction differ from the standard ones and do not reduce to them. The theoretical results are compared with the experimental data.

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In a phenomenological analysis of magnetoelastic phenomena in cubic systems, the magnetoelastic energy is usually represented, following Akulov,[1] as a sum of the following two magnetoelastic invariants:

$$U_{me}=-a_1(\varepsilon_{xx}m_x^2+\varepsilon_{yy}m_y^2+\varepsilon_{zz}m_z^2) \qquad (1)$$
$$-2a_2(\varepsilon_{xy}m_xm_y+\varepsilon_{xz}m_xm_z+\varepsilon_{yz}m_ym_z),$$

where ε_{ij} are the components of the strain tensor and m_i the components of the magnetization vector. We observed[2] certain peculiarities in the behavior of the magnetostriction of holmium iron garnet $Ho_3Fe_5O_{12}$, which are not covered by the classical theory of magnetostriction. Notice must be taken primarily of the unusually strong dependence of the magnetostriction of this garnet on the magnetic field, a dependence that cannot be attributed to the paraprocess of this material. A second important fact is that the temperature and the angular dependence of the magnetostriction do not correspond to those which follow from the classical theory[3] based on representing the magnetoelastic energy in the form (1). The reason is that the theory in Ref. 1 was developed for weakly anisotropic magnetic ions (strictly speaking, for ions in the S state), whereas the Ho^{3+} ion in iron garnets is strongly anisotropic.[2] The same situation obtains also for other rare-earth (RE) ions with nonzero orbital angular momenta.

The development of a theory of magnetostriction of magnetically ordered substances with strongly anisotropic ions, however, encounters serious difficulties. To understand the nature of the magnetostriction of strongly anisotropic magnetically ordered substances (and, in particular, iron garnets), we have turned to investigations of simpler magnets, namely RE paramagnetic garnets $R_3M_5O_{12}$, where M is a nonmagnetic ion (Al, Ga, and others). For these magnets the problem becomes simpler, since there is no exchange interaction of the RE ions with the iron ions in them.

Definite progress has been made recently in the investigation of magnetostriction of anisotropic paramagnets.[4–12] It has been shown, in particular, that the temperature and field dependences of the magnetostriction in compounds such as $LiTbF_4$ depends on the character of the ground state. The problem was solved for definite types of measurement generally by numerical means, and the angular dependence of the magnetostriction was not analyzed.

There is another approach, which we shall follow in the present paper. It is traditional in magnetism to pay principal attention in the theory to the invariants that determine the magnetoelastic energy (and hence also the magnetostriction) of the crystal. Garnets have a complicated structure with six nonequivalent RE-ion sites with low symmetry of the surrounding. The complicated spatial symmetry and the aforementioned appreciable anisotropy of the magnetic moment of the RE ion are the reason why the set of invariants in the magnetoelastic energy differs substantially from (1). It is determined by the space group and not by the point group of the crystal. This is also the reason why the magnetostriction has the unusual angular dependence that will be discussed below.

To determine the essential invariants we shall carry out a microscopic analysis of the magnetoelastic energy, based on the known premises concerning the ground state of RE ions in a garnet crystal. This analysis permits determination of the actual forms of the field, temperature, and orientation dependences of the magnetostriction, connect them with the character of the ground states of the investigated ions, and compare the theory with experiment using a small number of indeterminate magnetoelastic parameters.

THEORY

Crystalographic structure. RE garnets have a cubic symmetry described by the space group O_h^{10}. The unit cell of the garnet contains eight $R_3M_5O_{12}$ formula units, and the RE ions are located at sites with dodecahedral surrounding (c-sites). The symmetry of the surrounding of these sites is described by the point group D_2, which has one-dimensional irreducible representations. The basis functions of the irreducible representations of the guantities of interest to us (of the components of the strain tensor, of the operator **J**, of the external magnetic field strength vector **H**, and of the even powers of the components x', y', and z') are listed in the table. Here and below x', y', and z' pertain to the local coordinate system and x, y, and z to the crystallographic frame, whose unit vectors are [100], [010], and [001]. In addition, in our

TABLE I. Basis functions of irreducible group representations.

Irreducible representations	$J_iH_ix_i$	ε_{ij}	$(x)^{2n}$
Γ_1	—	ε_{ij}	$K_m^n(\Gamma_1)=2^{-1/2}\left(Y_{2n}^{2m}+Y_{2n}^{-2m}\right),\ m=0,\ldots,n$
Γ_2	J_zH_zz	ε_{xy}	$K_m^n(\Gamma_2)=2^{-1/2}/i\left(Y_{2n}^{2m}-Y_{2n}^{-2m}\right),\ m=1,\ldots,n$
Γ_3	J_yH_yy	ε_{xz}	$K_m^n(\Gamma_3)=2^{-1/2}\left(Y_{2n}^{2m-1}-Y_{2n}^{-2m+1}\right),\ m=1,\ldots,n$
Γ_4	J_xH_xx	ε_{yz}	$K_m^n(\Gamma_4)=2^{-1/2}/i\left(Y_{2n}^{2m-1}+Y_{2n}^{-2m+1}\right),\ m=1,\ldots,n$

convenition the Latin and Greek indices designate the local and crystallographic frames, respectively.

Hamiltonian of problem. The Hamiltonian of an RE ion at site r (in terms of the local symmetry axes of the given site) will be represented in the form

$$\mathscr{H}^{(r)}=\mathscr{H}_{cr}+\mathscr{H}_z+\mathscr{H}_{me}.$$

Here $\mathscr{H}_{cr}$ is the Hamiltonian of the RE ion interaction with the crystal field, and $\mathscr{H}_z$ is the Hamiltonian of the RE ion interaction with the external field and is conveniently represented in the form

$$\mathscr{H}_z=-\mu_B g_J\mathbf{JH}=-\sum_{j=2}^{4}\mu(\Gamma_j)H(\Gamma_j),\quad \mu(\Gamma_j)=g_J\mu_B J(\Gamma_j),$$

(μ_B is the Bohr magneton and g_J is the Lande factor of the basic multiplet of the RE ion); $\mathscr{H}_{me}$ is the single-frequency magnetoelastic Hamiltonian:

$$\mathscr{H}_{me}=\sum_{j=1}^{4}\varepsilon(\Gamma_j)\sum_{mn}C_{mn}{}^{j}K_m{}^{n}(\Gamma_j)\equiv\sum_{j}\varepsilon(\Gamma_j)N(\Gamma_j),$$

where C^j_{mn} are the magnetoelastic coefficients, and $\varepsilon(\Gamma_1)$ will be taken to mean an arbitrary linear combination of the diagonal components of the strain tensor: $\varepsilon(\Gamma_1)=a_1\varepsilon_{xx}+a_2\varepsilon_{yy}+a_3\varepsilon_{zz}$, $N(\Gamma_j)$ are quantities having the meaning of magnetoelastic stresses and transform in accord with the irreducible representations Γ_j of the point group D_2.

Effective spin-Hamiltonian. The energy spectrum of RE ions in garnet is split to the utmost: to doublets and singlets respectively in Kramers and non-Kramers ions. The distance between the doublets of Kramers ions reaches 50–100 cm^{-1}. The picture is similar also for certain non-Kramers ions (Tb^{3+}, Ho^{3+},...), whose lower levels are quasidoublets (in which the distance $2\Delta_0$ between the singlets is of the order of 1 cm^{-1}) sufficiently well separated from the upper levels.[13]

In a theoretical consideration of the magnetic and magnetoelastic properties of garnets with such ions at low temperatures $T \ll 100$ K it suffices to confine oneself to the lower doublet or quasidoublet, and take the interaction with the unpopulated excited levels into account by perturbation theory. It is convenient in such a physical situation to go over from the general Hamiltonian of the problem to the effective spin-Hamiltonian. To construct the spin-Hamiltonian we project $\mathscr{H}$ on the subspace of the functions generated by the two states of the lower doublet or quasidoublet. We shall use here the small parameter $\varkappa\sim\|\mathscr{H}_z\|W_3^{-1}$, $\|\mathscr{H}_{me}\|W_3^{-1}$, where W_3 is the characteristic energy that separates the excited levels from the ground doublet. In the analysis that follows we shall deal separately with the Kramers and non-Kramers ions.

Kramers ion. Using the Γ_i multiplication table for the D_2 group:

$$\Gamma_i\times\Gamma_i=\Gamma_1,\quad \Gamma_2\times\Gamma_3=\Gamma_3\times\Gamma_2=\Gamma_4,\quad \Gamma_2\times\Gamma_4=\Gamma_4\times\Gamma_2=\Gamma_3,$$

$$\Gamma_3\times\Gamma_4=\Gamma_4\times\Gamma_3=\Gamma_2,$$

we find that in the approximation linear in ε the effective spin Hamiltonian takes the form1)

$$\mathscr{H}_{eff}=-\sum_{j=2}^{4}\sigma(\Gamma_j)\left[g_j\mu_B H(\Gamma_j)+\sum_{i,k=2}^{4}\varepsilon(\Gamma_k)H(\Gamma_i)C_{ki}\delta(\Gamma_k\times\Gamma_i,\Gamma_j)\right],\tag{2}$$

where $\sigma(\Gamma_i)$ are Pauli matrices: $\sigma(\Gamma_2)=\sigma_{z'}$, $\sigma(\Gamma_3)=\sigma_{y'}$, $\sigma(\Gamma_4)=\sigma_{x'}$; g_j are the diagonal elements of the g-tensor of the ground doublet: $g_2=g_z$, $g_3=g_y$, $g_4=g_x$; C_{ki} are the real effective magnetoelastic coefficients having a W^{-1} dependence on the crystal field; there are nine of them.

Non-Kramers ion. Assume that at $\mathbf{H}=0$ the wave functions Ψ_i of the two levels of the ground quasidoublet, with energies $-\Delta_0$ and Δ_0, are transformed in accord with the representations Γ_A and Γ_B (Fig. 1), while the wave functions of the excited levels are transformed in accordance with Γ_m. All the wave functions are chosen to be real. The matrix elements of the operator $\mathscr{H}_{me}$ are then real, and those of $\mathscr{H}_z$ imaginary. We find in this case that the effective spin-Hamiltonian is of the form

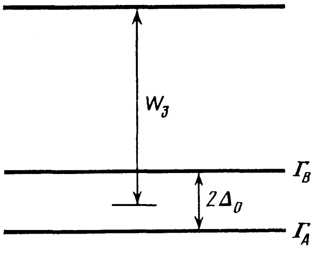

FIG. 1. Lower-level scheme of a non-Kramers RE ion in the garnet structure.

$$\mathcal{H}_{eff}$$

$$=-\sigma_{x'}\left[\sum_l \varkappa_0{}^l H(\Gamma_l)H(\Gamma_g\times\Gamma_l)+\varepsilon(\Gamma_g)N_0\right]-\sigma_{y'}\mu_g\left[H(\Gamma_g)\right. \tag{3}$$

$$\left.+\sum_l B_l\varepsilon(\Gamma_l)H(\Gamma_g\times\Gamma_l)\right]-\sigma_{z'}\left[\Delta_0+\sum_l \varkappa_-{}^l H^2(\Gamma_l)+N_1\varepsilon(\Gamma_1)\right],$$

where

$$\Gamma_g=\Gamma_A\times\Gamma_B,\quad \mu_g=-i\langle A|\mu(\Gamma_g)|B\rangle,\quad N_0=-\langle A|N(\Gamma_g)|B\rangle,$$

$$2N_-=\langle B|N(\Gamma_1)|B\rangle-\langle A|N(\Gamma_1)|A\rangle,$$

$$\varkappa_0{}^l=\sum_m \langle A|\mu(\Gamma_l)|m\rangle$$

$$\times\langle m|\mu(\Gamma_g\times\Gamma_l)|B\rangle W_m{}^{-1},\quad \varkappa_-{}^l=\sum_m[|\langle A|\mu(\Gamma_l)|m\rangle|^2$$

$$-|\langle B|\mu(\Gamma_l)|m\rangle|^2](2W_m)^{-1},$$

$$B_l=\frac{i}{\mu_g}\sum_m[\langle A|N(\Gamma_l)|m\rangle\langle m|\mu(\Gamma_l\times\Gamma_g)|B\rangle$$

$$+\langle A|\mu(\Gamma_l)|m\rangle\langle m|N(\Gamma_l\times\Gamma_g)|B\rangle]W_m{}^{-1}.$$

Thermodynamic potential. We calculate the contribution of the magnetoelastic interaction to the thermodynamic potential of the crystal. This calls for the following: a) Using the spin Hamiltonians obtained above, find the spectrum of the investigated RE ion as a function of the external magnetic field and of the strain. b) Calculate the thermodynamic potential of the r-th site, $\Phi_r = -T\ln Z_r$, where Z_r is the partition function. c) Separate from r the magnetoelastic part in accord with the formula

$$\Phi_{me}=\sum_{\alpha\beta}(\partial\Phi/\partial\varepsilon_{\alpha\beta})\varepsilon_{\alpha\beta};$$

d) Sum the result over r from 1 to 6. The transformation from the local coordinates of site r to the laboratory frame is by means of the formulas

$$\varepsilon_{\alpha\beta}^{(r)}=\sum_{\gamma\delta}T_{\alpha\gamma}{}^r\varepsilon_{\gamma\delta}(T_{\delta\beta}{}^r)^{-1},\quad T_{\alpha\beta}{}^r=(e_\alpha e_\beta{}^r), \tag{4}$$

where T is the rotation matrix, and e_α and e_β^r are unit vectors of the laboratory and local coordinate frames, respectively. We use for the latter the definition given in Ref. 2. We now present the calculation results.

Kramers ion.

$$\Phi_{me}=-\sum_{r=1}^{6}\sum_{ijk}\varepsilon^{(r)}(\Gamma_i)H^{(r)}(\Gamma_j)\frac{M^{(r)}(\Gamma_k)}{g_k}C_{ij}\delta(\Gamma_i\times\Gamma_j,\Gamma_k), \tag{5}$$

where $M^{(r)}(\Gamma_k)$ is the magnetic-moment component of the r-th ion and transforms in accord with the irreducible representation Γ_k:

$$M^{(r)}(\Gamma_k)=g_k{}^2\mu_B{}^2H^{(r)}(\Gamma_k)\chi_r,\qquad \chi_r=\mathrm{th}\,(\Delta_r/T)/\Delta_r. \tag{6}$$

Here $2\Delta_r$ is the splitting of the ground doublet of the r-th ion and is determined by the spin-Hamiltonian (2) at $\varepsilon = 0$:

$$2\Delta_r=\left\{\sum_{i=2}^{4}[g_i\mu_BH^{(r)}(\Gamma_i)]^2\right\}^{1/2}. \tag{7}$$

Non-Kramers ion.

$$\Phi_{me}=-\sum_{r,l}\chi_r[N_-\varepsilon^{(r)}(\Gamma_1)\varkappa_-{}^l(H^{(r)}(\Gamma_l))^2$$

$$+\mu_g{}^2B_l\varepsilon^{(r)}(\Gamma_l)H^{(r)}(\Gamma_g)H^{(r)}(\Gamma_g\times\Gamma_l) \tag{8}$$

$$+N_0\varkappa_0{}^l\varepsilon^{(r)}(\Gamma_g)H^{(r)}(\Gamma_l)H^{(r)}(\Gamma_g\times\Gamma_l)]$$

$$-N_-\Delta_0\sum_r\varepsilon^{(r)}(\Gamma_1)\left[\chi_r-\frac{2}{\Delta_0}\mathrm{th}\frac{\Delta_0}{2T}\right].$$

Here $2\Delta_r$ is the splitting of the ground quasidoublet and is determined by the spin-Hamiltonian (3) at $\varepsilon = 0$:

$$2\Delta_r=\left\{\left[\Delta_0+\sum_i\varkappa_-{}^lH^2(\Gamma_i)\right]^2+\mu_g{}^2H^2(\Gamma_g)\right\}^{1/2}$$

$$\approx\left[\Delta_0{}^2+\mu_g{}^2H^2(\Gamma_g)\right]^{1/2}. \tag{9}$$

We have left out of (9) a small term of order $\varkappa_0^2 H^4$, which can be easily accounted for if necessary.

Magnetoelastic variants of the space group O_h^{10}. From the symmetry viewpoint, expression (5) for Φ_{me} is a combination of invariants of the garnet-crystal space group O_h^{10}, which are made up of the components of the strain tensor, of the magnetic field, and of the magnetic moments of the six RE sublattices. We shall consider the structure of expression (5) from this point of view. The components of the vectors of the sublattice magnetic moments $\mathbf{M}(r)$, $r = 1,\dots,6$, make up an 18-dimensional irreducible representation of the O_h^{10} group, which we shall designate $\mathfrak{M}$. It can be resolved into magnetic modes, to each of which corresponds a definite irreducible representation τ_i. They are given, e.g., in Ref. 14. In that book are considered 12 sites for RE ions. For the magnetic properties of RE paramagnetic garnets it suffices to use the abbreviated 6-site description of the RE subsystem.

Of greatest interest to us are the following modes:

$$\tau_9:\ (00M_3^{(r)}),\quad (00H_3^{(r)}),\quad (00\varepsilon^{(r)}(\Gamma_2));$$

$$\tau_9{}':\ (M_1^{(r)}00),\quad (H_1^{(r)}00),\quad (\varepsilon^{(r)}(\Gamma_4)00);$$

$$\tau_7:\ (0M_2^{(r)}0),\quad (0H_2^{(r)}0),\quad (0\varepsilon^{(r)}(\Gamma_3)0);$$

$$\tau_1:\ \varepsilon^{(r)}(\Gamma_1),$$

where $r = 1,\dots,6$. These modes are expressed here in local coordinates, in which they take the simplest form. We do not present the rather lengthy expressions for the modes in the crystallographic coordinate frame, to which we can transform using Eqs. (4). The representations τ_9, τ_9', and τ_7 are three-dimensional. In abstract six-dimensional space, each of the points of which is a three-dimensional vector, these representations are specified by matrices of dimensionality

3×6. The unitary representation τ_1 is one-dimensional and is specified by a 1×6 matrix.

It follows from (5) that the magnetoelastic energy is determined by the product of three modes: $\tau(\varepsilon_{ik})$, $\tau(\mathfrak{M})$, and $\tau(\mathbf{H})$. For this product to be an invariant of the group it is necessary that it transform in accord with the unitary representation τ_1. To find all the invariants that are products of the components $\varepsilon_{\alpha\beta}$, H_α, and $M_\alpha^{(r)}$ it is necessary to find all the triple products of the representations τ_9, τ_9', τ_7, and τ_1 which contain a single τ_1. Such products are $\tau_1\times\tau_9\times\tau_9$, $\tau_1\times\tau_9'\times\tau_9'$, $\tau_1\times\tau_7\times\tau_7$ as well as products of the type $\tau_7\times\tau_9\times\tau_9'$, with different permutations of the indices. We assume in these products that the first, second, and third factors pertain to the strains $\tau(\varepsilon_{ik})$, $\tau(\mathfrak{M})$, and $\tau(\mathbf{H})$, respectively. It can be easily seen by direct comparison that these invariants (of which there are nine) coincide with those contained in (5). Thus, the general expression (5) for the magnetoelastic energy can be represented in the form

$$\Phi_{me}=\sum_{k=1}^{9} G_k J_k(\hat{\varepsilon},\mathfrak{M},\mathbf{H}),$$

where J_k are invariants written in abstract form,

$$J=\tau(\varepsilon)\oplus\tau(\mathfrak{M})\times\tau(\mathbf{H}).$$

The indices of the representations and the number of the invariant have been left out here for brevity. In accordance with the foregoing, a representation that coincides with $\tau(\varepsilon)$ is singled out in the direct product $\tau(\mathfrak{M})\times\tau(\mathbf{H})$. The symbol $\oplus$ denotes a scalar product in the space introduced above, i.e., a scalar product of three-dimensional vectors (or one-dimensional in the case of τ_1) and summation over r. A symmetry analysis of the potential (8) for a non-Kramers ion can be carried out in by the same procedure.

Magnetostriction. Let us calculate, starting from expression (5) for the magnetoelastic energy, the crystal magnetostriction along the [111] direction. To this end it is necessary to add to Φ_{me} the energy of the elastic stresses and minimize the sum over the components of the tensor ε_{ik}. We present the results of such a calculation of the quantity $(\delta l/l)_{[111]}=\varepsilon_{xy}+\varepsilon_{xx}+\varepsilon_{yz}$, where the components of the tensor ε_{ik} are calculated in the crystallographic system of coordinates:

$$\left(\frac{\delta l}{l}\right)_{[111]}=c_{44}{}^{-1}\Big\{\sum_{i=2}^{4} C_{1i}g_i(a_2-a_1)\sum_{r=1}^{6}(-1)^r(H^{(r)}(\Gamma_i))^2\chi_r$$
$$+(g_4C_{32}+g_2C_{34})\sum_{r=1,3,5} H^{(r)}(\Gamma_4)H^{(r)}(\Gamma_2)\chi_r$$
$$+(g_3C_{42}+g_2C_{43})\sum_{r=2,4,6} H^{(r)}(\Gamma_3)H^{(r)}(\Gamma_2)\chi_r\Big\}, \tag{10}$$

where c_{44} is the component of the elastic-constants tensor (in the Voigt notation). The quantity (10) is thus determined in the general case by five effective parameters.

MAGNETOSTRICTION OF $Tb_3Ga_5O_{12}$ AND $Dy_3Al_5O_{12}$. THEORY AND EXPERIMENT

We have presented above general relations for the magnetostriction of paramagnetic garnets with anisotropic RE ions. To use these relations for specific garnets we must know the energy structure of their RE ions. The simplest are the expressions for the magnetostriction in the case of Ising RE ions. These can be taken to be the Tb^{3+} ion in the gallate garnet[13] and the Dy^{3+} ion in the aluminum garnet.[15] We have calculated theoretically the magnetostrictions of these garnets and compared them with the experimental data.

For the Kramers ion Dy^{3+} in $Dy_3Al_5O_{12}$ we have[15]

$$g_{z'}\equiv g_2=18.2,\quad g_{y'}\equiv g_3=0.4,\quad g_{x'}\equiv g_4=0.75.$$

In this case $\chi_1\approx\chi_2$, $\chi_3\approx\chi_4$, $\chi_5\approx\chi_6$. The validity of this statement, with allowance for the relation

$$H^{(1)}(\Gamma_2)=H^{(2)}(\Gamma_2),\quad H^{(3)}(\Gamma_2)=H^{(4)}(\Gamma_2),$$
$$H^{(5)}(\Gamma_2)=H^{(6)}(\Gamma_2)$$

follows obviously from (13) and (14) for field directions such that

$$g_2H^{(r)}(\Gamma_2)\gg g_3H^{(r)}(\Gamma_3),\quad g_4H^{(r)}(\Gamma_4).$$

For field orientations in planes of the type {100}, however, the component $H(\Gamma_2)=0$ for any pair of sites $r=1$ and 2, 3 and 4, or 5 and 6. Thus for example, for a vector $\mathbf{H}$ located in the (001) plane we have $H^{(1)}(\Gamma_2)=H^{(2)}(\Gamma_2)=0$. In this case, by virtue of the smallness of g_3 and g_4 and the following relation is valid at helium temperatures and higher even in strong fields,

$$(\Delta_{1,\,2}/2T)\,|_{H^{(1,2)}(\Gamma_2)=0}\lesssim 1.$$

Then

$$\chi_{1,\,2}=\text{th}\,(\Delta_{1,\,2}/2T)/\Delta_{1,\,2}\approx[1-{}^1/_3(\Delta_{1,\,2}/2T)^2]/2T.$$

It follows hence that $|\chi_1-\chi_2|\ll\chi_1$, i.e., $\chi_1\approx\chi_2$. Thus, at all field orientations the pure Ising approximation for χ_r is sufficient:

$$\chi_r\approx[g_2\mu_BH^{(r)}(\Gamma_2)]^{-1}\,\text{th}\,[g_2\mu_BH^{(r)}(\Gamma_2)/2T].$$

Expression (10) takes in this case the form (in the crystallographic coordinate system)

$$(\delta l/l)_{[111]}=C_1I_1(\mathbf{H})+C_2I_2(\mathbf{H}), \tag{11}$$

where

$$C_1=C_{34}+C_{43},\qquad C_2=2(a_2-a_1)(C_{13}g_3-C_{14}g_4)/g_2;$$

$$I_1(\mathbf{H})=\sum_{\alpha\beta} H_\alpha m_\beta(1-\delta_{\alpha\beta}), \tag{12}$$

$$I_2(\mathbf{H})=H_xH_ym_z/H_z+H_xH_zm_y/H_y+H_yH_zm_x/H_x, \tag{13}$$

$$m_\beta=\text{th}\,[g_2\mu_BH_\beta/2T]. \tag{14}$$

The quantity m_β has the meaning of the reduced components of the summary magnetization of the Dy^{3+} ions in the pure Ising approximation. Despite the fact that $|C_2|\ll|C_1|$ ($g_2\gg g_3, g_4$) in the general case, the invariant $I_2(\mathbf{H})$ can nonetheless make a noticeable contribution to $(\delta l/l)_{[111]}$ at low temperatures in strong fields when the external field is directed along an axis such as ⟨110⟩. At such an orientation of the vector $\mathbf{H}$ the invariant $I_2(\mathbf{H})$ is a quadratic function of the field and takes the form

$$I_2(\mathbf{H})=g_2\mu_BH^2/4T.$$

According to Ref. 13, the Tb^{3+} ion in $Tb_3Ga_5O_{12}$ has $\Gamma_g = \Gamma_A \times \Gamma_B = \Gamma_2$ and the Ising axis coincides with the local z axis. In this case we have, just as for the Dy^{3+} ion, $\chi_1 \approx \chi_2, \chi_3 \approx \chi_4, \chi_5 \approx \chi_6$ [see (9)]. In addition, since the splitting for the Tb^{3+} ion is $\Delta_0 \approx 1\ \text{cm}^{-1}$,[13] at helium temperatures and higher we have $\Delta_0/2T \ll 1$. Calculation of $(\delta l/l)_{[111]}$ using expression (8) in which $\Gamma_g = \Gamma_2$, with account taken of the facts described above, leads to an expression similar to (11):

$$(\delta l/l)_{[111]}=D_1I_1(\mathbf{H})+D_2I_2(\mathbf{H}), \tag{15}$$

where

$$D_1=(B_3+B_4)\mu_g, \qquad D_2=2(a_2-a_1)N_-(\varkappa_-^{(3)}-\chi_-^{(4)})/\mu_g,$$

and the quantities $I_{1,2}(\mathbf{H})$ are defined by Eqs. (12)–(14), in which g_2 must be replaced by μ_g/μ_B.

It must be noted that in contrast to $(\delta l/l)_{[111]}$ the quantities $(\delta l/l)_{[100]}$ for Kramers and non-Kramers RE ions will be described by different invariants whose field and angular dependences are not the same; this is a consequence of the difference between the magnetoelastic energies (5) and (8).

We emphasize once more that the representation of the magnetostriction $(\delta l/l)_{[111]}$ by a sum of two relatively simple invariants I_1 and I_2 defined by Eqs. (12)–(14) is valid only under the cited restrictions on the values of H and T, and is applicable only to strongly anisotropic (almost-Ising) ions. To describe the magnetostriction of RE garnets in the general case we can use Eqs. (5), (8), and (10).

The magnetostriction was measured with wire strain gauges at 4.2 K in fields up to 65 kOe. The samples were thin single-crystal $Tb_3Ga_5O_{12}$ and $Dy_3Al_5O_{12}$ cylindrical plates cut in the (110) plane with diameter 5 mm and thickness 1 mm. Special strain gauges with small galvanomagnetic effect were used. We measured the dependences of the magnetostriction strains along the [111] direction on the field oriented along the measurement direction [111] as well as along other directions in the (110) plane: [001], [110], $[11\bar{1}]$ and perpendicular to [111]. In addition, we measured the angular dependences of the magnetostriction along the [111] axis in a given field on the field orientation in the (110) plane.

Our measurements have shown that the magnetostriction along the [111] direction with the field oriented along the [001] axis is close to zero. The experimental data for the dependence of the magnetostriction along the [111] axis for other field directions are shown in Figs. 2a and 2b for $Tb_3Ga_5O_{12}$ and $Dy_3Al_5O_{12}$, respectively. Attention is called to the following: in most cases the magnetostriction does not saturate in the investigated field range, whereas the plots of the field dependences of these compounds show a pronounced tendency to saturation at $H \geqslant 30$ kOe. This can be easily seen in Fig. 2, which shows the magnetization along the [111] axis as obtained in Ref. 16 for $Tb_3Ga_5O_{12}$ and from our data for $Dy_3Al_5O_{12}$. Thus, even a qualitative consideration leads to the conclusion that, in agreement with the theoretical estimates, the magnetostriction is not a simple function of the magnetization. The solid lines in Fig. 2 show the theoretical magnetostriction field dependences plotted using the parameters given above for the ground states of Tb^{3+} and Dy^{3+} in the garnets. The calculations were in accord with Eqs. (11) and (15). To determine the two magnetoelastic coefficients contained in these equations, the theoretical curves were matched to the experimental plots of the magnetostriction at two points: in a field 60 kOe oriented along the axes [111] and [110]. The magnetoelastic coefficients are

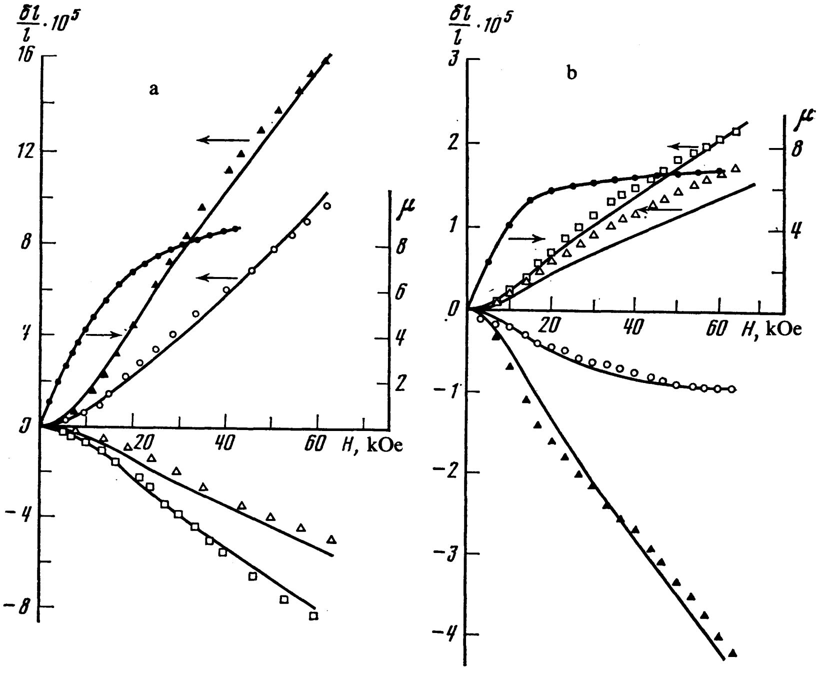

FIG. 2. Field dependences of the magnetostrictions of the single crystals $Tb_3Ga_5O_{12}$ (a) and $Dy_3Al_5O_{12}$ (b) at different field orientations in the (110) plane. Solid lines—theory. Symbols—experiment: ▲—**H**∥[111]; ○—**H**∥[110]; □—**H**⊥[111]; △—**H**∥$[11\bar{1}]$; ●—magnetization along [111] measured in units of μ_B per Tb^{3+} ion in (a) or Dy^{3+} ion in (b).

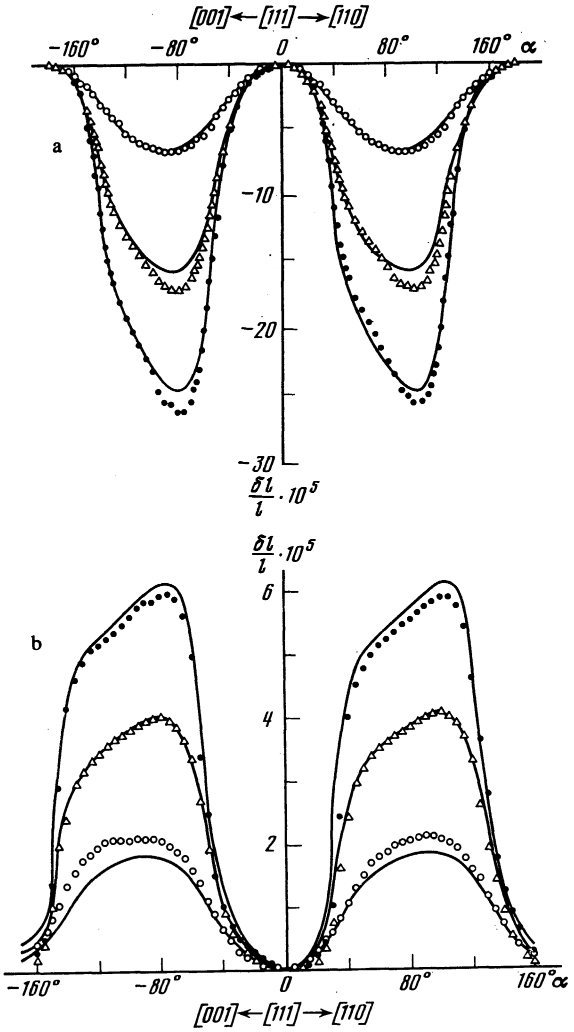

FIG. 3. Angular dependences of the magnetostriction of $Tb_3Ga_5O_{12}$ (a) and $Dy_3Al_5O_{12}$ (b) along the [111] axis at field orientation in the (110) plane, in fields: ○—H = 20 kOe; △—H = 40 kOe; ●—H = 60 kOe. The magnetostriction at H||[111] is taken equal to zero.

$$C_1\approx-4.2\cdot10^{-7}\,\mathrm{kOe}^{-1},\quad C_2\approx3.3\cdot10^{-8}\,\mathrm{kOe}^{-1},$$

$$D_1\approx14\cdot10^{-7}\,\mathrm{kOe}^{-1},\quad D_2\approx1.1\cdot10^{-7}\,\mathrm{kOe}^{-1}.$$

The experimental dependences of the magnetostriction on the angle at various field values are compared with the theoretical ones in Fig. 3. No adjustment parameters were used in this case. It can be seen that the theoretical dependences on the field and on its orientation describe subtle features of the experimental data, and in particular the asymmetry of angular dependence of $\delta l/l$, the almost linear field dependence of the magnetostriction in strong fields for most directions and, at the same time, the saturation of the dysprosium-aluminate magnetostriction in a field parallel to [110], and others, although there is no quantitative agreement in a number of cases. Better agreement with experiment can be obtained by taking into account in the theory invariants of higher order in the field, exchange interactions [this is particularly important for $Dy_3Al_5O_{12}$, whose Néel temperature is 2.5 K, Ref. 15], and others.

CONCLUSION

Thus, the constructed general theory of the magnetostriction of paramagnetic garnets with anisotropic RE ions permits a satisfactory description of the experimental dependences of the magnetostriction, along the [111] axis, of the garnets $Tb_3Ga_5O_{12}$ and $Dy_3Al_5O_{12}$ with Ising ions.

We emphasize once more that the experimental data do not agree with the theoretical relations that follow from the classical expression (1) for the magnetoelastic energy. This manifests itself most strongly in the fact that the magnetostriction continues to vary with the field in that field region where the magnetization saturates and, in addition, the angular dependence of the magnetostriction is much more complicated than predicted by this theory.

There are two physical causes of this difference. First, as shown above, the magnetoelastic-coupling constants are determined not only by the lower levels but also by the influence, primarily, of the excited states. In other words, the magnetoelastic coupling appears in the considered cases because of mixing, in the field, of states belonging to different doublets. This is so to speak an analog of the van Vleck susceptibility in the magnetoelasticity, and determines the field dependence of the magnetostriction. Second, in magnets with weakly anisotropic ions the saturation magnetic moment does not depend on the field orientation. In magnets with strongly anisotropic RE ions the saturation magnetic moment changes with change of field orientation relative to the crystallographic axes. This fact together with the presence of nonequivalent sites occupied by the RE ions is indeed the cause of the unusual dependence of the magnetostriction on the field direction.

The angular and field dependences of the magnetoelastic interaction and of the magnetostrictions are determined by the space group of the crystal, and the point symmetry of the crystal suffices for their description only as $H\rightarrow0$.

We note that the theory developed above deals with states having an effective spin 1/2 and contradicts to some degree the classical theory of Akulov[1] and of the Callens,[17] which predicts the vanishing of the magnetostriction in the case of ions with $S=1/2$.

[1)] Here and elsewhere we have left out of $\mathscr{H}_{\mathrm{eff}}$ the terms proportional to the unit matrix, since they are irrelevant to the subsequent analysis.

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