Magnetostriction of paramagnetic rare-earth garnets

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(Submitted 28 January 1988)
Zh. Eksp. Teor. Fiz. **94**, 358–370 (August 1988)

Experimental and theoretical investigations were made of the magnetic-field and temperature dependences of the magnetostriction of single crystals of paramagnetic rare-earth (R) garnets $R_3Ga_5O_{12}$ and $R_3Al_5O_{12}$ in magnetic fields up to 50 kOe at temperatures in the range 4.2–50 K. The field and temperature dependences of the magnetostriction were influenced greatly by the nature of the ground state (singlet, Kramers doublet, random non-Kramers doublet) of the rare-earth ion in the crystal field. Quantum-mechanical expressions were obtained for the magnetostriction and these described well the experimental results. An analysis was made of the behavior of the magnetostriction in the magnetically ordered state ($T < T_N$) of $Dy_3Al_5O_{12}$ experiencing metamagnetic phase transitions. The relationship between the magnetoelastic properties and the linear magnetic birefringence was investigated.

Many rare-earth compounds exhibit a giant magnetostriction ($\lambda = \Delta l \sim 10^{-3}$).[1] It has recently been found that the magnetostriction of rare-earth magnetics is interesting not only because of its magnitude, but also because of unusual magnetic-field, temperature, and orientational dependence which does not fit the framework of the usual representations. This behavior is quantum mechanical and is due to a strong spin–orbit interaction in rare-earth ions and a consequent partial quenching of the orbital momentum of these ions. A detailed quantitative theory of the magnetostriction of rare-earth compounds is far from complete. Serious difficulties in analyzing the experimental data on the magnetostriction of magnetically ordered rare-earth compounds (in addition to the difficulties associated with the complexity of the electron structure of rare-earth ions) are due to the existence of several (single-ion exchange, etc.) mechanisms of the magnetoelastic coupling.

We concentrate our attention here on the single-ion mechanism. We report a systematic experimental investigation of the magnetostriction of single crystals of several rare-earth gallium garnets $R_3Ga_5O_{12}$ and aluminum garnets $R_3Al_5O_{12}$ at temperatures in the range 4.2–50 K where these materials are paramagnetic; the Neel temperatures of these compounds are less than 2.5 K (Ref. 2). (The magnetostriction of rare-earth paramagnetic garnets had been investigated previously using mainly polycrystalline samples[3] and only some partial data on the magnetostriction of single crystals of $Dy_3Al_5O_{12}$ and $Tb_3Ga_5O_{12}$ are available.[4]) Our measurements were carried out using fairly strong magnetic fields (10^4–10^5 Oe) when the effects of the interaction of rare-earth ions with one another could be ignored. We were able to establish clearly the relationship between the behavior of the magnetostriction and the nature of the ground state of rare-earth ions in the crystal field.

One of the factors which stimulated the present investigation was the information we gained working on another magnetic effect which is even under time inversion, linear magnetic birefringence in paramagnetic garnets which we studied earlier.[5] We found that the magnetic-field and temperature dependences of the linear magnetic birefringence of the majority of rare-earth garnets could not be described by the relationship derived from the phenomenological theory of even magnetic Akulov–Callen effects[6]:

$$\Delta n \propto \hat{I}_{5/2}\{\mathscr{L}^{-1}[m(H,T)]\}, \qquad (1)$$

where $\hat{I}_{5/2}$ is the reduced Bessel function, $\mathscr{L}^{-1}$ is the inverse Langevin function, and m is the relative magnetization. As shown in Ref. 5, this is due to the fact that the main condition for the validity of the phenomenological theory, that the crystal field energy be small compared with the Zeeman energy, is not satisfied by rare-earth garnets. In the case of garnets containing rare-earth ions with a nonzero orbital momentum, the splitting of the ground-state multiplet of a rare-earth ion in the crystal field is approximately 10^2–10^3 cm^{-1} (Ref. 7), which is much larger than the Zeeman splitting amounting to 1–10 cm^{-1} in fields of 10^4–10^5 Oe. In this case the behavior of the linear magnetic birefringence and of other even magnetic effects is not described, contrary to the Akulov–Callen theory, by a universal magnetization function [Eq. (1)], but depends strongly on the nature of the splitting of the ground-state multiplet in the crystal field.

SAMPLES AND EXPERIMENTAL METHODS

We determined the magnetostriction of single crystals of paramagnetic garnets containing rare-earth ions with a nonzero orbital momentum: we studied gallates with the formula $R_3Ga_5O_{12}$ (R = Nd, Tb, Dy, Ho, Er, Tm, Yb) and aluminates $R_3Al_5O_{12}$ (R = Tb, Dy, Ho, Er, Tm, Yb). Our measurements were made on the same plates, with faces parallel to the {110} and {100} planes of a crystal, which were used earlier[5] in our study of the linear magnetic birefringence. We determined the difference between the longitudinal (field parallel to the direction of strain measurement) and transverse (field perpendicular to the direction of strain measurement) magnetostrictions $\Delta\lambda_{[pqt]}^{(ijk)}$ (here and below the lower index will be used for the direction of measurement of the longitudinal magnetostriction and the upper for the plane in which the transverse magnetostriction was measured). The field was oriented along the [100], [110], and [111] axes of the crystal.

The magnetostriction was determined by the method of quartz piezoelectric sensors, which were used first by Berezin[8] to measure the longitudinal magnetostriction and by Semenov[9] to find the difference between the longitudinal and transverse magnetostrictions. Thin y-cut quartz plates bonded suitably to a sample were used.[9] In the determination of

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the strain by the piezoelectric transducer method it was essential to ensure that the measurement time was much shorter than the time τ needed for the charge to leak away ($\tau = RC$, where C is the capacitance of the piezoelectric transducer and R is the input resistance of the measuring system), created in the transducer by the deformation of the sample, so that this method had been used earlier only in measurements employing pulsed magnetic fields.[8,9] However, it is not possible to use pulsed fields in studies of the magnetostriction of paramagnets at low temperatures because of the strong magnetocaloric effect (in a pulsed field the process of magnetization is adiabatic).

The magnetostriction was determined by the piezoelectric transducer method in a constant field of a superconducting solenoid using a modulation method we developed for this purpose: a sample was subjected to a magnetizing field H_0 and a weak (~ 100 Oe) alternating (77 Hz) field h. The signal recorded by a measuring circuit was proportional to the derivative $d\lambda/dh$ of the magnetostriction with respect to the field. The field dependence of the magnetostriction was found by integrating the field dependence of $d\lambda/dh$. The absolute sensitivity of the modulation method was deduced from the magnetostriction of standard samples measured by the strain gauge method. The error in the determination of the absolute value of the magnetostriction in our measurements was 10–15%, whereas the error in the relative measurements was only 5%.

EXPERIMENTAL RESULTS

The magnetostriction of cubic crystals can be divided into a bulk part, independent of the direction of measurement of the strain, and an anisotropic part which varies with the direction of measurement.[1] We shall be interested in the anisotropic magnetostriction, which in the case of garnets is of the single-ion type, is due to the crystal field, and—as demonstrated theoretically—is characterized by the difference between the longitudinal and transverse magnetostrictions $\Delta\lambda^{(ijk)}_{[pqt]}$.

Our garnets could be divided into three groups differing in the nature of the ground state of the rare-earth ion in the crystal field.

The ground state of the Tm^{3+} ion in $Tm_3Ga_5O_{12}$ and $Tm_3Al_5O_{12}$ garnets is a nonmagnetic singlet separated from the higher levels by an energy gap of 63 and 35 cm^{-1}, respectively.[7] Figure 1 shows the magnetostriction of $Tm_3Ga_5O_{12}$ at 4.2 K. We can assume that at this temperature only the ground state of the Tm^{3+} ion in thulium gallate is populated in the magnetic fields employed in our study. It is clear from Fig. 1 that in this case the magnetostriction varied quadratically with the field and, since the magnetization of $Tm_3Ga_5O_{12}$ was linear in the field in this range,[5] the results obtained demonstrated that the magnetostriction depended quadratically on the magnetization:

$$\lambda^{(ijk)}_{[pqt]} \propto m^2. \qquad (2)$$

The same dependence of the magnetostriction follows from the phenomenological equation (1) when the magnetization is low. Consequently, the Akulov–Callen theory describes the field dependence of the anisotropic magnetostriction of the garnets with a rare-earth ion in the singlet ground state. It should be stressed that the quadratic field dependence of the magnetostriction is observed only if the energy gap from the ground-state singlet excited levels is large. This is supported by our data on the magnetostriction of $Tm_3Al_5O_{12}$: in this paramagnet the magnetostriction at 4.2 K observed in strong fields (in excess of 30 kOe) did not obey Eq. (2). Thulium gallate and aluminate behaved differently because the excited levels were closer to the ground-state singlet in the aluminate than in the gallate.

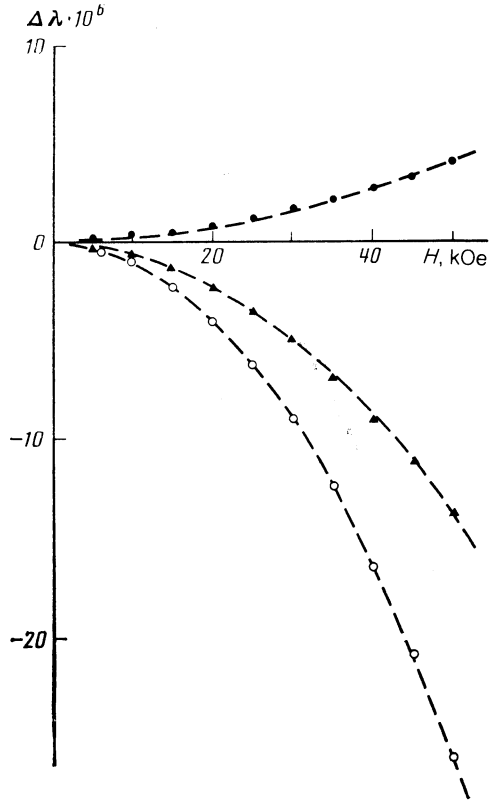

FIG. 1. Field dependence of the magnetostriction of $Tm_3Ga_5O_{12}$ at 4.2 K in the (110) plane: ○) H‖[111]; ▲) H‖[110]; ●) H‖[001]. The dashed curves represent dependences calculated on the basis of Eq. (2).

In many garnets the ground state of the rare-earth ion is an isolated Kramers doublet separated by a fairly large energy gap from the higher levels. This is true of the Nd^{3+}, Dy^{3+}, Er^{3+}, and Yb^{3+} ions in garnets.[7] The field dependences of the magnetostriction of different garnets containing these ions are qualitatively similar. We give here the data on the magnetostriction of $Yb_3Al_5O_{12}$ (Fig. 2) and $Dy_3Al_5O_{12}$ (Fig. 3). These garnets were selected by way of illustration because in the aluminate an ion of Dy^{3+} is of the Ising type with the g tensor components: $g_x = 0.73$, $g_y = 0.4$, and $g_z = 18.2$ (Ref. 10), whereas the Yb^{3+} ion can be regarded approximately as isotropic: $g_x = 3.738$, $g_y = 2.594$ and $g_z = 3.842$ (Ref. 11) and, as shown below, the theoretical description of the magnetostriction is simpler in these cases. A characteristic feature of garnets with such ions is that the magnetostriction does not saturate in strong fields where the paramagnetic saturation of the magnetization is observed (the field dependence of the magnetization of garnets is given in Refs. 5 and 12). Consequently, in this case the magnetostriction is not described by the Akulov–Callen phenomenological model the theoretical field depen-

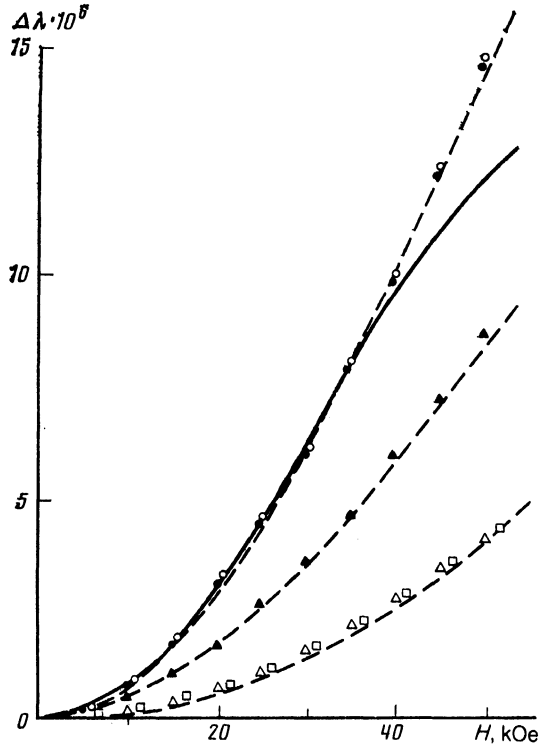

FIG. 2. Field dependence of the magnetostriction of $Yb_3Al_5O_{12}$ at 4.2 K in the (110) plane: □) $\mathbf{H}\|[\bar{1}11]$, ▲) $\mathbf{H}\|[\bar{1}10]$, ●) $\mathbf{H}\|[001]$ and the corresponding results for the (100) plane: △) $\mathbf{H}\|[011]$; ○) $\mathbf{H}\|[001]$. The dashed curves are calculated using Eq. (12) when the continuous curves are based on Eq. (1).

dence of the magnetostriction based on this model calculated from Eq. (1) is shown by the continuous curves in Figs. 2 and 3].

The third group are garnets with non-Kramers rare-earth ions (Tb^{3+}, Ho^{3+}), the ground state of which is an isolated quasidoublet (the separation between the quasidoublet levels is $\sim 5\ cm^{-1}$ and the gap from the quasidoublet to the first higher level is approximately 40–60 cm^{-1}).[7] In this case (the data for $Tb_3Al_5O_{12}$ are given by way of example, in Fig. 4) the field dependences of the magnetostriction are qualitatively similar to the field dependences discussed above for garnets with Kramers rare-earth ions in the doublet ground state: there is no saturation of the magnetostriction when the magnetization becomes saturated, i.e., the field dependence of the magnetostriction again disobeys the phenomenological model.

The magnetostriction of different paramagnetic garnets depends in different ways on temperature. This is demonstrated clearly in Fig. 5 which gives the temperature dependence of the magnetostriction of some of the investigated garnets. The magnetostriction of $Tm_3Ga_5O_{12}$ varies more slowly than the magnetostriction of the other compounds at low temperatures and in this case the Tm^{3+} ion is in the singlet ground state. The magnetostriction of the garnets with rare-earth ions in the doublet or quasidoublet ground state varies more rapidly with temperature, but even then the temperature dependence of the magnetostriction is weaker than that predicted by Eq. (1).

THEORY OF THE MAGNETOSTRICTION OF PARAMAGNETIC GARNETS; COMPARISON WITH EXPERIMENTAL RESULTS

As pointed out already, the main mechanism of the magnetoelastic coupling in paramagnetic garnets is of the single-ion type. This mechanism is due to the interaction of magnetic ions with the deformation-induced distortions of their environment, described by the effective deformation Hamiltonian $\mathscr{H}_d$. In general, the Hamiltonian $\mathscr{H}_d$ can be represented by a combination of invariants composed from

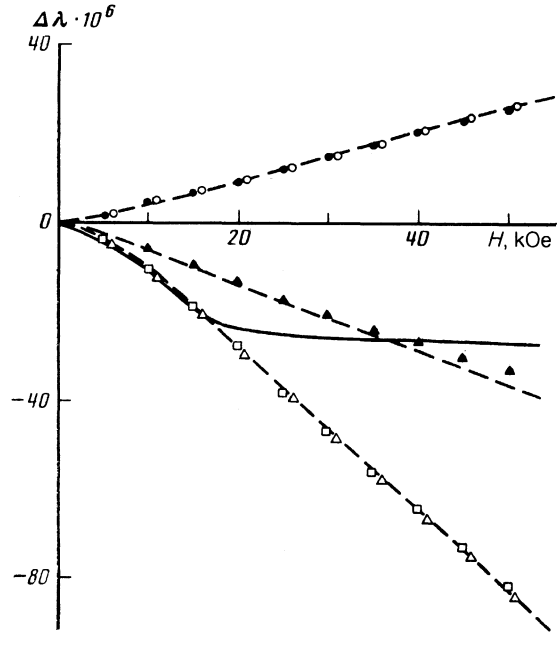

FIG. 3. Field dependence of the magnetostriction of $Dy_3Al_5O_{12}$ at 4.2 K in the (110) and (100) planes. The notation is the same as in Fig. 2.

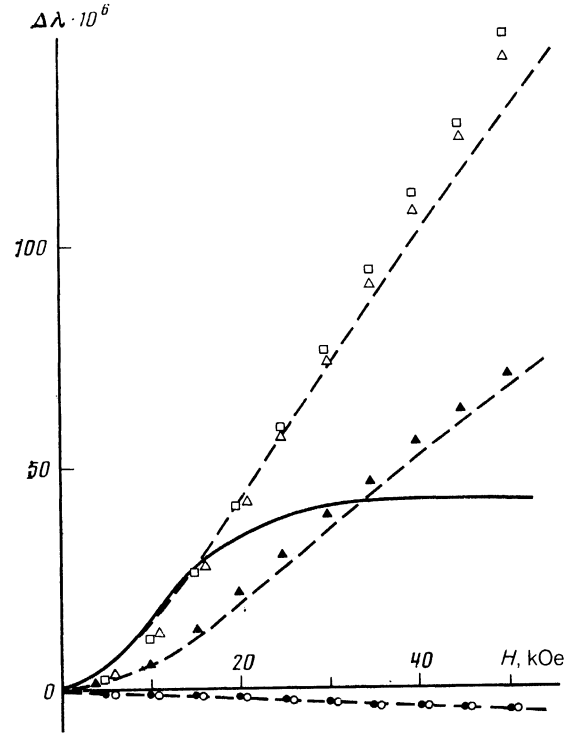

FIG. 4. Field dependence of the magnetostriction of $Tb_3Al_5O_{12}$ at 4.2 K in the (110) and (100) planes. The notation is the same as in Fig. 2.

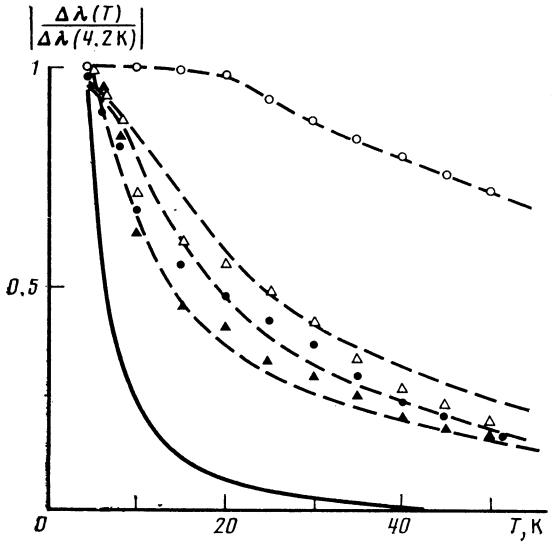

FIG. 5. Reduced temperature dependence of the magnetostriction in a 40-kOe magnetic field. The dashed curves are calculated on the basis of Eq. (12). The notation is as follows: ○) $Tm_3Ga_5O_{12}$, $\mathbf{H}\|[111]$; △) $Dy_3Al_5O_{12}$, $\mathbf{H}\|[111]$; ▲) $Tb_3Al_5O_{12}$, $\mathbf{H}\|[111]$; ●) $Yb_3Al_5O_{12}$, $\mathbf{H}\|[100]$. The continuous curve represents a calculation based on Eq. (1) for $Yb_3Al_5O_{12}$.

products of the spherical harmonics $Y_n^m(\mathbf{r})$ and of the components of the strain tensor $\varepsilon_{\alpha\beta}$ (Ref. 13):

$$\mathcal{H}_d=\sum_i \sum_{\alpha\beta nm} c_{n\alpha\beta}{}^m Y_n{}^m(\mathbf{r}_i) r^n \varepsilon_{\alpha\beta}, \tag{3}$$

where the summation over i applies to all the electrons in a partly filled shell of the magnetic ion. The coefficients $c^m_{n\alpha\beta}$ are governed by the characteristics of the electron–phonon interaction and reflect the symmetry of the environment of the magnetic ion. A microscopic calculation of the coefficients $c^m_{n\alpha\beta}$ is a very difficult task,[14] especially for garnets with a complex crystal structure, so that we shall consider them only as phenomenological parameters.

The appearance of magnetoelastic coupling is different for rare-earth ions with zero (or quenched) orbital momentum ($L=0$) and for those with ions characterized by $L\neq 0$. In the case of the ions with $L=0$ the magnetoelastic coupling appears when we allow for the levels lying above the orbital singlet in the third and higher orders of perturbation theory with respect to the operator $V=\mathcal{H}_{SL}+\mathcal{H}_d$ ($\mathcal{H}_{SL}=\lambda\,\mathbf{SL}$ is the spin–orbit interaction operator).[15] The main contribution to the magnetoelastic spin Hamiltonian comes from the third-order terms of perturbation theory. This contribution is of the form

$$\mathcal{H}_{me}=\sum b_{\alpha\beta}{}^m \varepsilon_{\alpha\beta} Y_2{}^m(\mathbf{S}), \tag{4}$$

where $b^m_{\alpha\beta}$ are the effective magnetoelastic coefficients. We note that these coefficients exhibit a dependence of the $b\propto(\lambda/W)^2$ type on the ratio of the magnitudes of the spin–orbit interaction and the energy gap W separating the excited states from the orbital singlet.[1)] Averaging Eq. (4) with an equilibrium density matrix, we obtain Eq. (1) for the magnetostriction derived in the phenomenological approach.

We are interested in the magnetostriction of crystals with rare-earth ions that have a nonzero orbital momentum. We shall discuss the deformation potential in this case. For a majority of rare-earth ions it is sufficient to consider the space of functions of the ground-state multiplet.

Projecting the Hamiltonian of Eq. (3) onto the space functions, we obtain

$$\mathcal{H}_{me}=\sum c_{n\alpha\beta}{}^m \varepsilon_{\alpha\beta} Y_n{}^m(\mathbf{J})\, a_n\langle r^n\rangle, \tag{5}$$

where a_n is the Stevens coefficients and $\mathbf{J}$ is the total mechanical momentum. The corrections to the free energy of the rare-earth ions dependent on the magnetic field and strains appear directly as a result of averaging of the Hamiltonian (5) over the states of the ground-state multiplet split by the crystal field.

The single-ion magnetoelastic energy of a crystal is an additive sum of the contributions of the individual magnetic ions:

$$E_{me}=\sum_k E^{(k)}_{me}, \qquad E^{(k)}_{me}=\sum c_{n\alpha\beta}{}^m a_n\langle r^n\rangle \varepsilon_{\alpha\beta}\delta\langle Y_n{}^m(\mathbf{J})\rangle^{(k)}, \tag{6}$$

where

$$\delta\langle Y_n{}^m(\mathbf{J})\rangle^{(k)}=\langle Y_n{}^m(\mathbf{J})\rangle_H{}^{(k)}-\langle Y_n{}^m(\mathbf{J})\rangle_0{}^{(k)} \tag{7}$$

describes the change in the field of the multiple moment of the k th rare-earth ion. The summation in Eq. (6) is carried out over all the rare-earth ions in a crystal. In particular, in the case of garnets we must allow for the fact that these ions are distributed between six inequivalent c sites (the local symmetry of these is governed by the point group D_2), which differ in the orientations of the local symmetry axes.[13]

Minimization of the sum of the magnetoelastic energy E_{me} and the elastic energy E_e with respect to the components of the strain tensor $\varepsilon_{\alpha\beta}$ yields the following expression for the anisotropic magnetostriction:

$$\lambda=-\sum B_{n\alpha\beta}{}^m \delta\langle Y_n{}^m(\mathbf{J})\rangle^{(k)} l_\alpha l_\beta,$$
$$B_{n\alpha\beta}{}^m=a_n\langle r^n\rangle \sum S_{\alpha\beta\gamma\delta} c_{n\gamma\delta}{}^m, \tag{8}$$

where $S_{\alpha\beta\gamma\delta}$ is the tensor of the elastic (compliance) constants and $l_{\alpha,\beta}$ are the direction cosines of the directions of measurement.

The field and temperature dependences of the magnetostriction are governed by the multipole moments of the rare-earth ions $\delta\langle Y^m_n(\mathbf{J})\rangle$. A rigorous calculation of these moments can be carried out if we know the energy levels and the wave functions of the ground-state multiplet of a rare-earth ion in the crystal and magnetic fields. At present complete data on these characteristics of the majority of rare-earth ions in garnets are still lacking. We shall therefore identify the main contribution to the temperature and field dependence of the magnetostriction, which can be calculated knowing only the nature of the splitting of the ground state of a rare-earth ion in the crystal field.

In the case of rare-earth ions with the singlet ground state (Eu^{3+}, Pr^{3+}, Tm^{3+}) we can expect nonzero corrections to the multipole moments $\delta\langle Y^m_n(\mathbf{J})\rangle$ only in the second order of perturbation theory,[13] described by

$$\delta\langle Y_n^m\rangle = \sum Q_{npq}^m(T) H_p H_q,$$

$$Q_{npq}^m = (\mu_B g_J)^2 \sum \{[(E_k - E_g)(E_e - E_g)]^{-1} \times (2\langle g|Y_n^m|e\rangle\langle e|J_p|k\rangle\langle k|J_q|g\rangle + \langle g|J_p|e\rangle\langle e|Y_n^m|k\rangle\langle k|J_q|g\rangle) - (E_k - E_g)^{-2}\langle g|Y_n^m|g\rangle|\langle g|J_p|k\rangle|^2\delta_{qp}\delta_{ek}\}\rho_g, \quad (9)$$

$$\rho_g = \left[\sum_n \exp(-E_n/T)\right]^{-1} \exp(-E_g/T),$$

where E_k is the level of a singlet $|k\rangle$ and g_J is the Landé factor.

The components of the magnetic moment of a rare-earth ion with the singlet ground state are given by

$$M_\alpha = (\mu_B g_J)^2 H_\alpha \chi_\alpha(T),$$

$$\chi_\alpha(T) = \sum_{kg} (E_k - E_g)^{-1} |\langle g|J_\alpha|k\rangle|^2 \rho_g.$$

Therefore, in this case at low temperatures ($T \ll W$, where W is the gap between the ground and the first excited singlet of a rare-earth ion) the magnetization is a linear function of the field and the magnetostriction is a quadratic function, which does not change with temperature. Summation of Eq. (8), subject to Eq. (9), over all the inequivalent sites of rare-earth ions yields expressions for the orientational dependence of the magnetostriction obtained earlier on the basis of the Akulov–Callen phenomenological theory.[6] In particular, in the case of garnets containing such rare-earth ions we can expect that the rules of the even Akulov effects will be obeyed:

$$\Delta\lambda_{[011]}^{(100)} = \Delta\lambda_{[\bar{1}11]}^{(110)}, \quad \Delta\lambda_{[\bar{1}10]}^{(110)} = {}^1/_2(\Delta\lambda_{[001]}^{(100)} + \Delta\lambda_{[\bar{1}11]}^{(110)}). \quad (10)$$

All the theoretical predictions are confirmed by the experimental data on garnets with Tm^{3+} ions: we can see from Fig. 1 that the magnetostriction of $Tm_3Ga_5O_{12}$ at 4.2 K depends quadratically on the field and that it satisfies the rules for even effects. Moreover, at low temperatures the magnetostriction of this garnet is independent of temperature (Fig. 5).

In the case of Kramers rare-earth ions with the doublet ground state (Sm^{3+}, Nd^{3+}, Dy^{3+}, Er^{3+}, Yb^{3+}) we can expect field-dependent multipole moments $\delta\langle Y_n^m\rangle$ in the first order of perturbation theory[13]:

$$\delta\langle Y_n^m\rangle = \sum G_{n\alpha\beta}^m H_\alpha M_\beta / g_\beta \mu_B, \quad (11)$$

where $G_{n\alpha\beta}^m$ is the material tensor, where symmetry is governed by the point group D_2 characterizing the symmetry of the environment of the rare-earth ion, and M_β are the components of the magnetic moment of this ion:

$$M_\beta = {}^1/_2(g_\beta\mu_B)^2 H_\beta \Delta^{-1} \tanh(\Delta/2T), \quad \Delta = \left[\sum_\alpha (\mu_B g_\alpha H_\alpha)^2\right]^{1/2}$$

(g_α are the components of the g tensor of the ground-state doublet of a rare-earth ion).

It is clear from Eq. (11) that the magnetostriction of the garnets with rare-earth ions in the doublet ground state does not saturate in strong fields in spite of paramagnetic saturation when $M \to$ const. In this range of fields the magnetostriction is a linear function of the field (small corrections quadratic in the field appear in the second-order perturbations).

It is more difficult to describe the behavior of the magnetostriction in weaker fields before the magnetization reaches saturation. Substituting Eq. (11) into Eq. (8), we find that the magnetostriction of the garnets containing rare-earth ions in the doublet state generally depends on a linear combination of the magnetizations of the rare-earth ions at different inequivalent c sites, and not on the sum of these magnetizations which determines the total magnetization of a crystal.

The expression for the magnetostriction is simplest in two limiting cases: for rare-earth ions with an isotropic magnetic moment ($g_x = g_y = g_z = g$) and extremely anisotropic Ising rare-earth ions for which only the g_z component of the g tensor differs from zero.

In the former (isotropic) case which applies to, for example, garnets with Yb^{3+} ions, the magnetostriction is described by

$$\Delta\lambda \propto HM(H, T), \quad (12)$$

where M is the magnetization of the rare-earth ion due to the lower doublet (without allowance for the Van Vleck contribution) and equal to

$$M(H, T) = {}^1/_2\mu_B g \tanh(\mu_B g H/2T).$$

In this case the magnetostriction for the field orientation along one of the principal crystallographic directions satisfies the rules for even effects.[10] All these features (proportionality of the magnetostriction to the product HM, the rules governing even effects) follow from the experimental data for garnets with the nearly isotropic Yb^{3+} ion (Figs. 2 and 5).

In the latter (Ising) case which applies to, for example, the garnet $Dy_3Al_5O_{12}$ for which the g_z component of the g tensor of the ground-state multiplet of the Dy^{3+} ion is considerably greater than the other two components, we find that the magnetostriction depends on the intensity and orientation of the magnetic field according to

$$\Delta\lambda = \lambda_1 \sum H_\alpha M_\alpha (l_\alpha^2 - {}^1/_3) + \lambda_2 \sum H_\alpha M_\beta l_\alpha l_\beta (1 - \delta_{\alpha\beta}), \quad (13)$$

where λ_i are the magnetostriction coefficients, $l_{\alpha,\beta}$ are the direction cosines of the directions of measurements, and M_α is the component of the magnetization of a crystal given by

$$M_\alpha = \tfrac{1}{2}\mu_B g_z \tanh(\mu_B g_z H_\alpha/2T).$$

In this case the magnetostriction varies also with the field as the product HM, but the rules for even effects are not obeyed. This is due to the fact that the magnetization is anisotropic, i.e., that it varies as a result of a change in the orientation of the magnetic field in a crystal. The magnetostriction of $Dy_3Al_5O_{12}$ behaves precisely in this way, as demonstrated by the experimental data presented in Figs. 3 and 5.

In the case of non-Kramers rare-earth ions with the ground state in the form of a quasidoublet (Tb^{3+}, Ho^{3+}),

the magnetostriction appears in principle even without participation of admixtures of higher levels.[13] This is due to the fact that the wave functions $|A\rangle$ and $|B\rangle$ of the quasidoublet state are not Kramers-conjugate with one another in zero field. Moreover, in these calculations we must allow for the fact that in accordance with the Griffith theorem, these ions are of the Ising type and in the case of the Tb^{3+} and Ho^{3+} ions in garnets the Ising axis is oriented along the local z axis.[13] In this case the contribution made to the magnetostriction by the zeroth-order terms is

$$\Delta\lambda=\lambda\sum_{\alpha}\chi(H_\alpha)\,(l_\alpha^2-1/3),$$

where

$$\lambda=1/12(c_{11}-c_{12})^{-1}\Delta_0\sum a_n\langle r^n\rangle(2c_{nzz}^{m}-c_{nxx}^{m}-c_{nyy}^{m})$$
$$\cdot(\langle A|Y_n^m|A\rangle-\langle B|Y_n^m|B\rangle),$$

and $\chi(H_\alpha)$ is the magnetic susceptibility due to the ground-state quasidoublet when the magnetic field has the appropriate direction:

$$\chi(H_\alpha)=\Delta^{-1}(H_\alpha)\,\mathrm{th}\,[\Delta(H_\alpha)/2T],$$
$$\Delta(H_\alpha)=[\Delta_0^2+(\mu H_\alpha)^2]^{1/2},\qquad \mu=2\mu_B g_J|\langle A|J_z|B\rangle|,$$

c_{11} and c_{12} are the elastic moduli, and Δ_0 is the separation between the quasidoublet levels.

The zeroth-order contribution to the magnetostriction differs from zero in fields with $\mathbf{H}\|\langle 001\rangle$ and $\mathbf{H}\|\langle 110\rangle$, but it vanishes when the field is parallel to $\langle 111\rangle$. Moreover, it should saturate in strong fields (under paramagnetic saturation conditions). It follows from our results that this behavior is not confirmed by the experimental result for our garnets (see Fig. 4 for $Tb_3Al_5O_{12}$): in strong fields the magnetostriction increases with the field and $\Delta\lambda_{\langle 111\rangle}$ is larger than $\Delta\lambda_{[110]}$ and $\Delta\lambda_{[100]}$. This shows that the contribution of the zeroth order of perturbation theory to the magnetostriction is small (clearly because the wave functions describing the ground-state quasidoublet of the Tb^{3+} and Ho^{3+} ions are nearly Kramers-conjugate). Therefore, for garnets with such rare-earth ions we have to include corrections representing higher (first and second) orders of perturbation theory, and these corrections are fully analogous to those discussed above in the case of Kramers "Ising" ions. The experimental field and temperature dependences of the magnetostriction of garnets containing non-Kramers rare-earth ions (Tb^{3+} and Ho^{3+}) are practically identical with those made from garnets with Kramers rare-earth ions (Figs. 2–5).

MAGNETOSTRICTION IN A MAGNETICALLY ORDERED STATE OF $Dy_3Al_5O_{12}$

As pointed out already, at temperatures below 4.2 K the majority of the rare-earth garnets we investigated go over to the antiferromagnetic state. We know only of one experimental investigation[16] of the magnetostriction of $Dy_3Al_5O_{12}$ below the Néel temperature $T_N=2.5$ K (Fig. 6). The theory developed above can be obtained in a natural manner from the experimental results of Ref. 16. The compound $Dy_3Al_5O_{12}$ is a six-sublattice noncollinear Ising antiferromagnet: in zero field there are three pairs of orthogonal sublattices oriented along the $\pm[100]$, $\pm[001]$, and $\pm[001]$ crystallographic axes, respectively. At temperatures below 1.6 K in a field $H_c=4$ kOe, parallel to the $\langle 111\rangle$ axis, this compound undergoes a metamagnetic transition in which the directions of the magnetic moments of the sublattices making an obtuse angle with the direction of the field are reversed.[17]

According to Ref. 18, magnetic ordering in $Dy_3Al_5O_{12}$ can be described with the aid of two order parameters, one of which is ferromagnetic m and the other antiferromagnetic η:

$$m=\sum_{i=1}^{6}M_{z_i},\qquad \eta=\sum_{i=1}^{6}(-1)^i M_{z_i},$$

where M_{z_i} is the component of the magnetic moment of Dy^{3+} along z_i which is the local axis at the ith position. Generalization of the above theory involves replacement, in Eqs. (11)–(13), of the external field $\mathbf{H}$ with an effective field $\mathbf{H}+\mathbf{H}_{\mathrm{exch}}$, where $\mathbf{H}_{\mathrm{exch}}$ is the exchange field exerted on a given Dy^{3+} ion by other magnetic ions in a crystal. It depends on m and η. A direct calculation using Eq. (11) gives the following expression for the anisotropic magnetostriction $Dy_3Al_5O_{12}$ at temperatures less than the Néel temperature in a field directed along the [111] axis:

$$\Delta\lambda_{[111]}=AHm(H,T)+BH\eta(H,T). \tag{14}$$

The first term in Eq. (14) is the contribution of the ferromagnetic order parameter to the magnetostriction of a crystal. It follows naturally, from Eq. (13) describing the magnetostriction of $Dy_3Al_5O_{12}$ in the paramagnetic range of temperatures. On the other hand, the second term in Eq. (14) is due to the contribution of the antiferromagnetic order parameter η. Its appearance can be explained as follows. It is known[13,18] that the antiferromagnet order parameter of $Dy_3Al_5O_{12}$ interacts directly with the field $H_S=H_xH_yH_z$, which is called the "staggered field," i.e., the quantity $H_S\eta$ is an invariant of the space group of the crystal O_h^{10}. Obviously, the product $\eta(\varepsilon_{xy}H_z+\varepsilon_{xz}H_y+\varepsilon_{yz}H_x)$ is also an invariant. It is this product that determines the contribution of the parameter η to the magnetoelastic energy of a crystal and the corresponding contribution to the magnetostriction of $Dy_3Al_5O_{12}$.

In strong magnetic fields ($H>H_c$), we have $\eta\approx 0$ and

$$\Delta\lambda_{[111]}=AHm, \tag{15}$$

i.e., in this range of fields the expression for the magneto-

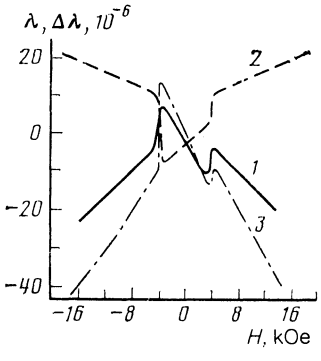

FIG. 6. Field dependence of the magnetostriction of $Dy_3Al_5O_{12}$ at 1.35 K in $\mathbf{H}\|[111]$: 1) longitudinal magnetostriction; 2) transverse magnetostriction; 3) difference between the longitudinal and transverse magnetostrictions.

TABLE I. Magnetostriction and linear magnetic birefringence of aluminate and gallate garnets at 4.2 K in a field of 50 kOe.

Rare-earth ion	Type of garnet	Sign of Stevens parameter a_2	H ∥ [111], plane (110)			H ∥ [100], plane (110)		
			$\Delta\lambda\cdot10^6$	$\Delta n\cdot10^6$	$\Delta n/\Delta\lambda$	$\Delta\lambda\cdot10^6$	$\Delta n\cdot10^6$	$\Delta n/\Delta\lambda$
Nd^{3+}	$Nd_3Ga_5O_{12}$	−	−6.5	22.5	−3.4	−3.4	1.8	−0.5
Tb^{3+}	$Tb_3Al_5O_{12}$	−	146	−525	−3.6	−7	−25	3.6
	$Tb_3Ga_5O_{12}$		330	−980	−3	−15	−49	3.2
Dy^{3+}	$Dy_3Al_5O_{12}$	−	−82	−91	1.1	24	−51	−2.1
	$Dy_3Ga_5O_{12}$		−274	−331	1.2	−77	−139	1.8
Ho^{3+}	$Ho_3Al_5O_{12}$	−	−148	269	−1.8	9.8	−29	−3
	$Ho_3Ga_5O_{12}$		−208	257	−1.2	14	−33	−2.4
Er^{3+}	$Er_3Al_5O_{12}$	+	−48	496	-10	11.3	343	30
	$Er_3Ga_5O_{12}$		−20	175	−8.5	−35	257	−7.3
Tm^{3+}	$Tm_3Al_5O_{12}$	+	−37	49	−1.3	3.6	15	4
	$Tm_3Ga_5O_{12}$		−25	38	−1.5	3.2	6.4	2
Yb^{3+}	$Yb_3Al_5O_{12}$	+	3.8	−1.8	−0.5	14.5	−15	−1.1
	$Yb_3Ga_5O_{12}$		7.5	−2.9	−0.5	24	−25.4	−1.1

striction is analogous to that valid in the paramagnetic region. Since at low temperatures the state of paramagnetic saturation is reached in fields of a few kilo-oersted, the magnetostriction varies linearly with the field (Fig. 6). A reduction of the field to H_c results in a metamagnetic transition from a ferromagnetic to an antiferromagnetic phase and the antiferromagnetic order parameter η increases strongly, whereas the ferromagnetic order parameter m practically vanishes. Therefore, a discontinuity of the magnetostriction is observed in the field H_c (Fig. 6). In the range of fields $-H_c<H<H_c$ the magnetostriction is described by

$$\Delta\lambda_{[111]}=BH_\eta. \tag{16}$$

Since the order parameter η for the range $|H|<H_c$ can be regarded as constant, in this range of fields the magnetostriction should depend linearly on the field and its sign should be reversed as a result of reversal of the sign of the field (Fig. 6). It should be noted that the linearity of the magnetostriction along the field (in cases when $|H|<H_c$ or $|H|>H_c$) cannot be explained by the phenomenological theory postulating that the magnetostriction is proportional to even powers of the actual order parameters.

MAGNETOSTRICTION AND LINEAR MAGNETIC BIREFRINGENCE

We discussed above the field and temperature dependences of the magnetostriction of garnets. We shall now consider the change in the magnetostriction along a series of rare-earth ions and compare this change with the corresponding effect in the case of the linear magnetic birefringence we investigated earlier.[5] It is clear from Table I that the magnetostriction and the linear magnetic birefringence vary quite differently when we pass from one rare-earth ion to another. This circumstance is due to the fact that the description of these formally similar effects exhibits quite definite differences.

According to Ref. 5, the linear magnetic birefringence is due to a change in the quadrupole moments of rare-earth ions in a magnetic field. The sign of the linear magnetic birefringence is governed by the sign of the product of second-order Stevens parameter and the quadrupole susceptibility of a rare-earth ion. A calculation of the quadrupole susceptibilities requires a detailed knowledge of the energy levels and wave functions of rare-earth ions which are not at present available. We can only conclude that the actual combinations of the quadrupole susceptibilities of the majority of rare-earth ions in garnets are positive since the sign of the linear magnetic birefringence of the majority of the investigated garnets is the same as the sign of the Stevens parameter a_2 (Table I). The most difficult to explain is the linear magnetic birefringence of garnets containing Ho^{3+}, when the sign of the effect varies with the direction. It would therefore be very desirable to investigate this topic and particularly the quadrupole susceptibilities of rare-earth ions in garnets.

In the magnetostriction case this situation is more complex. According to the above theory, the magnitude and the sign of the magnetostriction are governed by a linear combination of field-dependent multipole moments of rare-earth ions of the second (quadrupole moments) and fourth and sixth ranks, and are described by a large number of phenomenological parameters. Therefore, the magnetostriction changes in a complicated manner when we pass from one rare-earth ion to another (Table I), and we cannot predict the magnitude or even the sign of the magnetostriction.

On the other hand, the different behavior of the linear magnetic birefringence and the magnetostriction of garnets on transition from one rare-earth ion to another shows that the linear magnetic birefringence is not, contrary to the assumptions made—for example—in Ref. 19, a simple consequence of the magnetoelastic deformation of the cubic structure of a garnet and cannot be reduced to the elastooptic effect.

[1)] This explains the relatively small value ($\lambda\sim10^{-6}$) of the magnetostriction of crystals in the S state.

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Translated by A. Tybulewicz