Nature of magnetic anisotropy and magnetostriction of orthoferrites and orthochromites

A. M. Kadomtseva, A. P. Agafonov, M. M. Lukina, V. N. Milov, A. S. Moskvin, V. A. Semenov, and E. V. Sinitsyn

Moscow State University
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It is shown theoretically and experimentally that, besides the antisymmetric exchange, one-ion anisotropy of d-ions makes a substantial contribution to the anisotropy constant of orthoferrites and orthochromites. The sign of the one-ion anisotropy constants of the ions Fe^{3+} (Cr^{3+}) is different for the orthoferrite (orthochromite) of yttrium and lutetium. It is also shown that the one-ion mechanism of the d ions in the orthoferrites and orthochromites makes the decisive contribution to the magnetoelastic coupling.

INTRODUCTION

The magnetic anisotropy and magnetostriction of rare-earth orthoferrites and orthochromites is determined both by the rare-earth ions and by the ions of the d-metals. To separate the contribution made by the d-ions, it is customary to investigate orthoferrites and orthochromites with nonmagnetic rare-earth ions, such as La^{3+}, Y^{3+}, and Lu^{3+}. The most thoroughly studied in this respect to date are $YFeO_3$ and $LuFeO_3$, for which the magnetic anisotropy stabilizes the spin configuration G_xF_z. It is customarily assumed[1] that the anisotropy constant of the Fe (Cr) sublattice of the whole series of rare-earth orthoferrites (orthochromites) is close to the value observed for $YFeO_3$ ($YCrO_3$) and is determined mainly by the antisymmetrical d-ion exchange.

We report here experimental results that cannot be explained within the framework of the foregoing concepts. In particular, we show that the d-ions make significantly different contributions to the anisotropy constants of the yttrium and lutetium orthoferrites (orthochromites). Replacement of the Y^{3+} ions in yttrium orthoferrite by Lu^{3+} causes a strong increase of the threshold field that initiates the spin reorientation $G_xF_z \rightarrow G_zF_x$, i.e., an increase of the anisotropy constant. A similar substitution in yttrium orthochromite leads to the opposite result—to a sharp decrease of the anisotropy constant. The results can be explained by taking into account the substantial contribution of the one-ion mechanism the anisotropy constants of the d-ions when the one-ion anisotropies of Fe^{3+} and Cr^{3+} are of opposite sign. It is also shown that the one-ion mechanism makes the decisive contribution to the magnetoelastic constants.

EXPERIMENTAL RESULTS AND THEIR DISCUSSION

To study the contributions of the d-ions to the anisotropy constants and the magnetostriction of orthoferrites and orthochromites, we investigated the reorientational transitions induced by an external magnetic field in $YFeO_3$ and $YCrO_3$ single crystals, as well as in $Y_{0.5}Lu_{0.5}FeO_3$ and $Y_{0.5}Lu_{0.5}CrO_3$, which are convenient model objects because Y^{3+} and Lu^{3+} are nonmagnetic ions. The single crystals were grown from the solution in a melt of lead compounds.

The isotherms of the longitudinal and transverse magnetostriction and the torque curves were measured in strong magnetic fields up to 200 kOe and in the temperature interval from 78 to 300 K. The magnetostriction and the torque were measured with a piezoelectric tensor. Below T_N, all the orthoferrites above are noncollinear antiferromagnets with a transverse weakly ferromagnetic moment oriented along the c axis of a rhombic crystal (spin configuration G_xF_z). When a sufficiently strong magnetic field H is applied along the antiferromagnetism axis (the a axis of the rhombic crystal), a spin reorientation from G_xF_z to G_zF_x takes place.

The threshold field can be determined by measuring the field dependence of the maximum torque $L_{max}(H)$ observed when the external magnetic field is perpendicular to the weak ferromagnetic moment.[1,2] Figure 1 shows plots of $L_{max}(H)$ obtained for $YFeO_3$ and $YCrO_3$ single crystals at 80 K. As seen from Fig. 1, with increasing field applied along the a axis of the crystal, the maximum torque first increases practically linearly, until the spins move away from the field direction, after which the torque decreases steeply because of the spin reorientation.

At the threshold fields $H_{thr} = 75 \pm 4$ kOe for $YFeO_3$ and $H_{thr} = 33 \pm 2$ kOe for $YCrO_3$ the value of L_{max} decreases almost to zero, owing to the completion of the $G_xF_z \rightarrow G_zF_x$ spin reorientation. The threshold fields were determined also from the field dependence of the magnetostriction (Figs. 2 and 3) which appears in
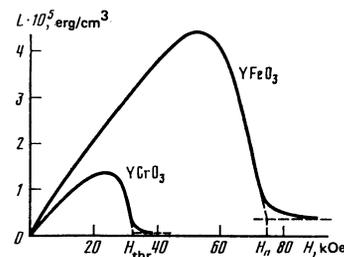
FIG. 1. Field dependence of torque ($H||a$) at 80 K for $YFeO_3$ and $YCrO_3$.

the spin-reorientation transition that is induced by the magnetic field $H\|a$.

As seen from Figs. 2 and 3, in the case of yttrium orthoferrite and orthochromite the threshold fields at which the spin rotation ends and the magnetostriction stops increasing are almost equal to the values obtained from the field dependence of the torque (Fig. 1). The magnetostriction strains accompanying the spin reorientation are strongly anisotropic and are different along the different crystallographic directions, both in magnitude and in sign.

Figures 2 and 3 show also the field dependences of the magnetostriction for the substituted-composition $Y_{0.5}Lu_{0.5}FeO_3$ and $Y_{0.5}Lu_{0.5}CrO_3$ signal crystals, obtained at 85 K. It is seen that the threshold field ($H_{thr} = 150 \pm 8$ kOe) for $Y_{0.5}Lu_{0.5}FeO_3$ increased greatly compared with the pure yttrium orthoferrite, whereas for $Y_{0.5}Lu_{0.5}CrO_3$ the threshold field decreased considerably to $H_{thr} = 17 \pm 1$ kOe when Y^{3+} ions were replaced by Lu^{3+}. The temperature dependence of the theshold fields, plotted in the temperature interval 80–300 K for all the investigated crystals, is shown in Fig. 4.

Knowing the threshold field, we can calculate the anisotropy field H_A from the equation[3]

$$H_{thr} = -H_D/2 + (H_D{}^2/4 + 2H_E H_A)^{1/2}. \tag{1}$$

Assuming, in accord with Ref. 4, exchange fields $H_E = 3.2 \times 10^6$ Oe and $H_E = 1.1 \times 10^6$ Oe for $YF3O_3$ and $YCrO_3$, respectively, as well as Dzyaloshinskiĭ fields $H_D^{Fe} = 1.4 \times 10^5$ Oe and $H_D^{Cr} = 6.1 \times 10^4$ Oe, we obtain $H_A(YFeO_3) = 2300$ Oe and $H_A(YCrO_3) = 1350$ Oe.

From the anistopy fields of the yttrium orthoferrite and orthochromite and from the angular dependences of the torques $L(\theta)$ (θ is the angle between the weakly ferromagnetic moment and the c axis of the crystal) we determined the anisotropy constants of the second and fourth order in the expansion of the thermodynamic potential

$$\Phi = \Phi_0 + K_1 \sin^2\theta + K_2 \sin^4\theta. \tag{2}$$

The $L(\theta)$ dependences were obtained by replotting the experimental torque curves $L(\varphi)$ (φ is the angle be-

FIG. 3. Field dependence, at $H\|a$ of the magetostriction of the orthochromites $YCrO_3$ [1] $\lambda\|a$; 2) $\lambda\|b,\ c$] and $Y_{0.5}Lu_{0.5}CrO_3$ [3] $\lambda\|c$].

tween the magnetic-field direction and the c axis of the crystal), measured in magnetic fields 2–3 times stronger than H_{thr}, at which the angle $\theta - \varphi$ was less than $10°$.[1]

At 80 K the second-order anisotropy constants K_1 per Fe^{3+} or Cr^{3+} ion in the yttrium orthoferrite and orthochromite are

$$K_1(YFeO_3) = 0.12\ \text{cm}^{-1}, \quad K_1(YCrO_3) = 0.043\ \text{cm}^{-1}.$$

The fourth-order anisotropy constant $K_2(YFeO_3)$ of yttrium orthoferrite did not exceed 12% of $K_1(YFeO_3)$, while $K_2(YCrO_3)$ is close to zero as expected.

The second-order anisotropy constant of the substituted orthoferrtie $Y_{0.5}Lu_{0.5}FeO_3$ increased to 0.44 cm^{-1}, while an anlogous substitution in the orthochormite $Y_{0.5}Lu_{0.5}CrO_3$ decreased the anisotropy constant to 0.022 cm^{-1}. The increase of the anisotropy constant in the yttrium orthoferrite and the decrease in the orthochromite following replacement of the Y^{3+} ions by Lu^{3+} is evidence that, despite the prevailing opinion, the anisotropy constants of the d sublattices differ greatly in the orthoferrite and orthochromite series.

To interpret the experimental results it is necessary to consider the principal mechanisms of the anisotropy and of the magnetostriction. In accordance with the prevailing notions, contributions to the anisotropy constant and to the magnetoelastic coupling are made by antisymmetric exchange, by magneto-dipole interaction, and by one-ion crystallographic anisotropy. It is known[5-8] that antisymmetric exchange and magneto-

FIG. 2. Field dependence, at $H\|a$ of the magetostriction of the orthoferrites $YFeO_3$ [1.3] $\lambda\|a,\ b$; 2) $\lambda\|c$] and $Y_{0.5}Lu_{0.5}FeO_3$ 4) $\lambda\|a$).
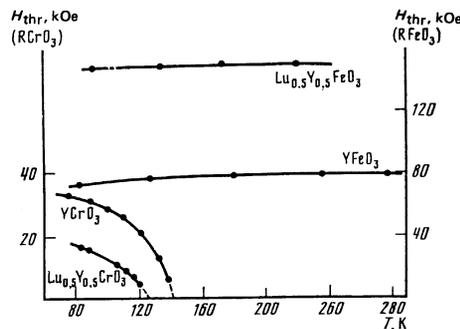
FIG. 4. Temperature dependence of threshold fields that induce the spin reorientation $G_x F_z \rightarrow G_z F_x$.

dipole anisotropy stablize the G_xF_z spin configuration in the entire orthoferrite series. The contribution of the antisymmetric exchange to the anisotropy constant K_1 in the ac plane can be represented in the form[9]

$$K_1 = 8IS^2(C_y^2 - 2A_y^2), \qquad (3)$$

where I is the exchange integral. C_y and A_y are the parameters of the "latent" noncollinearity of the spins[1] in the respective configurations $\Gamma_2(F_xC_yG_z)$ and $\Gamma_4(G_xA_yF_z)$. Recognizing that $I < 0$ and $C_y < A_y$ in orthoferrites,[9] we find that $K_1 > 0$ and calculate its numerical value using the theoretical values of the ratios C_y/F_z and A_y/F_z (Ref. 9) and the z component F_z of the ferromagnetism vector, which is known from the magnetization data (in fact, the cant angles of the magnetic sublattices):

$$K_1 = \begin{cases} 0.19 \text{ cm}^{-1}, & \text{YFeO}_3 \\ 0.24 \text{ cm}^{-1}, & \text{LuFeO}_3 \end{cases}$$

The magnetodipole contribution to K_1 for orthoferrites was calculated numerically earlier in Ref. 7. These calculations show that the contribution of the magnetodipole interaction to the magnetoelastic constants is on the whole small. The experimental data on the effect of pressure on the weakly ferromagnetic moment[9] indicate that the contribution of the antisymmetrical exchange to the magnetoelastic coupling is also small.

The nature of the one-ion second-order crystallographic anisotropy in orthoferrites is connected with the noncubic crystal field and with the spin-orbit interaction for the Fe^{3+} ions. The noncubic field can be represented as a sum of two contributions, a lattice contribution considered in the point-charge approximation ("point" lattice) and the contribution of the nearest environment (six O^2-ions) considered within the framework of the deformation model. In this model, the noncubic-field parameters, meaning also the anisotropy constants in the $Fe^{3+}-6O^2$ complexes, are assumed to be proportional to the deformation of the complex relative to an ideal octahedron.[5,8] The proportionality coefficients have the meaning of the electron-lattice interaction constants (the Jahn-Teller coupling constants) or of the magnetoelastic constants. The advantage of this approach to the analysis of noncubic fields is the possibility of determining the relation between the contributions to the anisotropy and to the magnetostriction.

We present now the calculated one-ion anisotropy and magnetostriction for the ac planes of $YFeO_3$ and $LuFeO_3$ within the framework of the indicated model. We represent the magnetoelastic energy in the form

$$\Phi_{me} = (L_a\varepsilon_{aa} + L_b\varepsilon_{bb} + L_c\varepsilon_{cc})\sin^2\theta + {}^1\!/_2\mu\varepsilon_{ac}\sin 2\theta, \qquad (4)$$

where $L_{a,b,c}$ and μ are the magnetoelastic constants and ε_{ij} are the components of the strain tensor. Table I lists the values of K_1, L_a, L_b, L_c, and μ calculated by us. The data in the first line (for both $YFeO_3$ and $LuFeO_3$ are the contributions of E-type strains (tension and compression of the axes of the $Fe^{3+}-6O^2$ octahedron), and the data of the third line are the contributions of the point lattice. Naturally, all that our model establishes is the relation between K_1, L_a, L_b, L_c, and μ for each of the considered contributions, but provides no "measurement unit" for each of the mechanisms.

The contribution made to K_1 in all cases is smaller by approximately two orders than that of the corresponding magnetoelastic parameters. The parameters $L_{a,b,c}$ and μ for $YFeO_3$ and $LuFO_3$ are close in value, but there is no correlation between the corresponding contributions to K_1. This fact can be understood if it is recognized that the magnetoelastic constants reflect the essentially perovskite character of the structure of the orthoferrites and respond weakly to relatively small orthorhombic distortions in whereas the very existence of K_1 is due only to the presence of the orthorhombic distortions in the orthoferrites.

We note that for parameters L_a, L_b, and L_c in Table I obey the relation $L'a + L'b + L'c = 0$, the reason being that we have not considered the influence of isotropic strains.

By using our present data on the magnetostriction of $YF3O_3$ in a field-induced spin-reorientation transition, as well as the data of Ref. 10 on the elastic constants of $TmFeO_3$, we can obtain the values of the parameters

$$L_a = -3.8 \cdot 10^7 \text{ erg/cm}^3; \quad L_b = -2.9 \cdot 10^7 \text{ erg/cm}^3; \quad L_c = 5.4 \cdot 10^7 \text{ erg/cm}^3.$$

For comparison with the data of Table I we need separate only the anisotropic parts of $L_{a,b,c}$. The constants $L_{a,b,c}$ renormalized in manner are equal to

$$L_i' = L_i - (L_a + L_b + L_c)/3;$$

$$L_a' = -3.4 \cdot 10^7 \text{ erg/cm}^3; \quad L_b' = -2.5 \cdot 10^7 \text{ erg/cm}^3; \quad L_c' = 5.9 \cdot 10^7 \text{ erg/cm}^3,$$

and the relation between them is surprisingly close to that predicted by the model that takes into account the contribution of the E strains of the $Fe^{3+}-6O^2$ octahedron. This allows us to suggest that the E-strain mechanism plays the predominant role in the magnetoelastic interactions, meaning also in the anisotropy. Assuming that the magnetostriction is due only to the E contribution, we obtain the measurement unit for the quantities in Table I:

$$A_E \approx 13.6 \cdot 10^7 \text{ erg/cm}^3,$$

i.e., for the contribution to K_1 we obtain (in erg/cm³)

$$K_1 = \begin{cases} -4.0 \cdot 10^5, & \text{YFeO}_3; \\ +1.6 \cdot 10^5, & \text{LuFeO}_3, \end{cases}$$

meaning that the one-ion anisotropies in the ac plane are of opposite sign in $YFeO_3$ and $LuFeO_3$. In $YFeO_3$ the one-ion anisotropy, in constrast to anitsymmetric exchange and magnetodipole interaction, favors the configuration G_xF_x, whereas in $LuFeO_3$ the G_xF_z configuration is energywise more profitable for all three mechanisms. This fact explains fully the growth of

TABLE I.

	$K_1 \cdot 10^2$	L'_a	L'_b	L'_c	μ
YFeO₃	−0.29	−0.24	−0.19	0.43	−0.30
	0.23	−0.38	0.19	0.19	−0.92
	0.07	−0.50	0.30	0.20	−1.21
LuFeO₃	0.12	−0.23	−0.18	0.41	−0.33
	0 09	−0.38	0.18	0.21	−0.90
	−0,17	−0.52	0.29	0.23	−1.10

TABLE II. Various contributions ot the first anisotropy constant in the ac plane for $YFeO_3$ and $LuFeO_3$ (in cm-1/ion).

	Antisymmetric exchange[6,9]	Magnetodipole interaction[7]	One-ion crystallographic anisotropy (E-contribution)	Total contribution	Experiment
$YFeO_3$	$19 \cdot 10^{-2}$	$5 \cdot 10^{-2}$	$-11 \cdot 10^{-2}$	$13 \cdot 10^{-2}$	$12 \cdot 10^{-2}$
$LuFeO_3$	$24 \cdot 10^{-2}$	$5 \cdot 10^{-2}$	$4 \cdot 10^{-2}$	$33 \cdot 10^{-2}$	–

the anisotropy of $Y_{1-x}Lu_x FeO_3$ in the ac plane with increasing concentration of the Lu^{3+} ions.

By way of illustration, Table II gives the contribution calculated on the basis of various data[7,8] and in the present paper, of the principal anisotropy mechanisms of the constant K_1 for $YFeO_3$ and $LuFeO_3$. Despite the model character of the calculation of the contributions of the antisymmetric exchange[6,8] and of the crystallographic anisotropy, the data of Table II show clearly the relative role of the various anisotropy mechanisms in the ac plane and point definitely to a much larger K_1 in $LuFeO_3$ than in $YFeO_3$. The dominant mechanism of the anisotropy in the ac plane for $LuFeO_3$ is antisymmetric exhange, whereas in $YFeO_3$ the crystallographic anisotropy cancels out the greater part of the contribution of the antisymmetric exchange.

No such analysis of the one-ion anisotropy was carried out for the orthochromite, in view of the lack of data on the crystal-structure parameters. However, the experimental data on the magnetostriction of $YCrO_3$ along the axes a, b, and c for a field-induced spin-reorientation transition (Fig. 3) are evidence in favor of the a magnetostriction mechanism that is connected wither with the contribution T_2 of the deformation model, or with the contribution of the point lattice. Indeed, the relation

$$\lambda_a \sim -{}^1/_2 \lambda_b \sim -{}^1/_2 \lambda_c$$

observed in $YCrO_3$ (Fig. 3) agrees with that predicted for the indicated mechanisms for isostructural orthoferrites (Tables II and III).

Data on EPR of the Fe^{3+} and Cr^{3+} ions[11] in La ortho-aluminate, which is isostructural to the orthoferrites, show that the one-ion anisotropy constants of the Fe^{3+} ions are close to those observed for the Cr_{3+} ions, but are of opposite sign. In particular, it was found in the axial-anisotropy approximation[11] ($V_{an} = DS_z^2$) that

$$LaAlO_3 : Fe^{3+}, D = 4.90 \cdot 10^{-2} \text{ cm}^{-1};$$

$$LaAlO_3 : Cr^{3+}, D = -5.55 \cdot 10^{-2} \text{ cm}^{-1}.$$

It is thus natural to assume that the one-ion contribution to the anisotropy constants of orthoferrites and orthochromites can differ in sign. There are grounds[7,8] for assuming that the contribution of the antisymmetrical exchange and of the magnetodipole interaction in the orthochromites, just as in orthoferrites, stabilizes the $G_x F_z$ spin configuration. The decrease of the anisotropy which we observed in experiment upon replacement of Y^{3+} by Lu^{3+} in orthochromites can then be attributed to the fact that the one-ion contribution increases the anisotropy constant in $LuCrO_3$ and decreases it in $LuCrO_3$. This is indirectly confirmed by the fact that the spin configuration $G_x F_x$ ($K_1 < 0$) is observed in $LuCrO_3$ but not in $YCrO_3$.

The observed regularities can be used for purposeful changes in the anisotropy constants in orthoferrites and orthochromites when new materials are synthesized for magnetically controlled optics and for computer technology. We note in particular, the interesting possibility of developing a composition with zero anisotropy constant in the ac plane on the basis of $Y_{1-x}Lu_xCrO_3$.

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