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ANISOTROPY OF MAGNETOSTRICTION IN THULIUM ORTHOFERRITE

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Longitudinal and transverse magnetostriction was investigated in a monocrystal of thulium orthoferrite along the a, b, and c crystal axes in the temperature range in which a spontaneous reorientation of spins is observed, and at liquid helium temperature. In the presence of a sufficiently strong magnetic field along the axis of antiferromagnetism, there was observed a magnetostriction resulting from inversion of the antiferromagnetic iron sublattices and having an anisotropic character. There was also observed a pronounced anisotropy of the magnetostriction caused by the paramagnetism of the rare-earth ions. The results obtained are discussed within the framework of a phenomenological treatment. An investigation was also made of the temperature dependence of the lattice parameters of a $Tu_{0.5}Sm_{0.5}FeO_3$ monocrystal. It is suggested that anisotropic change of the lattice parameters with temperature leads to a change of sign of the anisotropy constant and to reorientation of the spins in this compound.

THE magnetic properties of rare-earth orthoferrites above the ordering temperature of the rare-earth ions are determined by the weak ferromagnetism of the iron ions, on which is superposed the paramagnetism of the rare-earth ions.

At present the literature contains no data on the influence of the rare-earth and iron ions on the magnetoelastic properties of the orthoferrites. This paper reports an investigation of the longitudinal and transverse magnetostriction of a thulium orthoferrite monocrystal along the a, b, and c axes of the rhombic crystal near the temperature range in which spontaneous reorientation of the spins is observed, and at liquid helium temperature. Monocrystals of thulium orthoferrite were grown by the method of crucibleless zone fusion[1,2] in the Ferrite Problem Laboratory of the Moscow Power Institute. For measurement of the magnetostriction at helium temperatures, a low-temperature quartz dilatometer was adapted.

The curves of Figs. 1–3 show the dependence of the magnetostriction along the three principal crystallographic directions on a magnetic field applied along the c axis of the crystal, in the temperature interval from 78 to 130°K. It is seen that at temperatures below 95°K the magnetostriction has a complicated dependence on field. Thus the magnetostriction along the b axis of the crystal (Fig. 2) is negative in a weak field but then, with increase of field, changes sign and rapidly grows in size. At higher temperatures, the magnetostriction depends quadratically on the field; this attests to the fact that at these temperatures it is chiefly determined by the paramagnetism of the rare-earth ions. As is seen from Figs. 1–3, the magnetostriction caused by the paramagnetism of the rare-earth ions has a markedly anisotropic character, being different both in magnitude and in sign along different crystal axes. We observed anisotropy of the paramagnetic magnetostriction also when the field was applied along the a and b axes of the crystal.

The results obtained on magnetostriction of the rare-earth ions can be discussed within the framework of a phenomenological treatment. An expression for the magnetoelastic energy of orthoferrites[3] resulting from magnetization of the rare-earth ions can be written, on taking account of the symmetry of the crystal, in the following form:

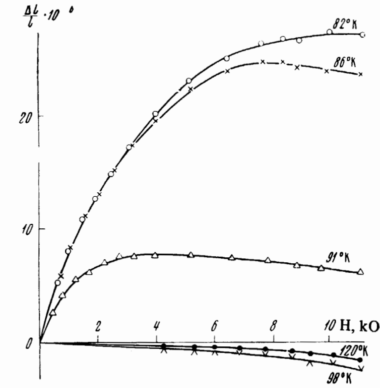

FIG. 1. Dependence of the longitudinal magnetostriction, along the c axis of a monocrystal of thulium orthoferrite, on the external magnetic field.

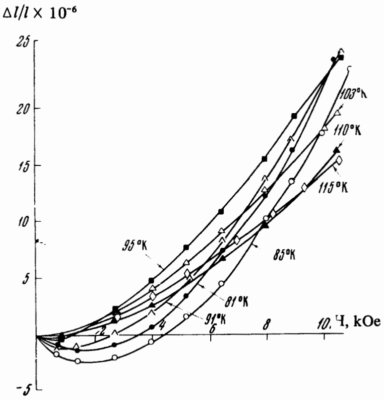

FIG. 2. Dependence of the magnetostriction along the b axis of a monocrystal of thulium orthoferrite on the field applied along the c axis of the crystal.

$$F_{\mathrm{me}} = [\Lambda_1(m_x^2 - m_z^2) + \Lambda_2(m_y^2 - m_z^2)]u_{xx}$$
$$+ [\Lambda_3(m_x^2 - m_z^2) + \Lambda_4(m_y^2 - m_z^2)]u_{yy}$$
$$+ [\Lambda_5(m_x^2 - m_z^2) + \Lambda_6(m_y^2 - m_z^2)]u_{zz}$$
$$+ 2\Lambda_7 m_x m_y u_{xy} + 2\Lambda_8 m_x m_z u_{xz} + 2\Lambda_9 m_y m_z u_{xz}.$$

Here Λ_k are magnetoelastic constants, $m_{x,y,z} = \chi_{x,y,z}H$ ($\chi_{x,y,z}$ are the susceptibilities of the rare-earth ions along the crystal axes a, b, c), and u_{ij} are the components of the deformation tensor of the crystal. On application of a field H along the c axis (the z direction) of the crystal, we have

$$m_z = \chi_z H, \quad m_x = m_y = 0$$

and the expression for the magnetoelastic energy will have the form

$$F_{\mathrm{me}} = -[\Lambda_1 + \Lambda_2]m_z^2 u_{xx} - [\Lambda_3 + \Lambda_4]m_z^2 u_{yy} - [\Lambda_5 + \Lambda_6]m_z^2 u_{zz}.$$

By tracing the analogy between the expression obtained and the expression for the energy of external stresses,

$$F_{\mathrm{ext.\,sw}} = -\Sigma\sigma_{ij}u_{ij},$$

one can calculate

$$u_{xx} = \left[\frac{\Lambda_1 + \Lambda_2}{E} - \frac{\mu}{E}(\Lambda_3 + \Lambda_4) - \frac{\mu}{E}(\Lambda_5 + \Lambda_6)\right] m_z^2,$$

$$u_{yy} = \left[-\frac{\mu}{E}(\Lambda_1 + \Lambda_2) + \frac{\Lambda_3 + \Lambda_4}{E} - \frac{\mu}{E}(\Lambda_5 + \Lambda_6)\right] m_z^2,$$

$$u_{zz} = \left[-\frac{\mu}{E}(\Lambda_1 + \Lambda_2) - \frac{\mu}{E}(\Lambda_3 + \Lambda_4) + \frac{\Lambda_5 + \Lambda_6}{E}\right] m_z^2,$$

where u_{xx}, u_{yy}, and u_{zz} may be considered the magnetostrictive deformations along the axes a, b, and c on application of a field along the c axis of the crystal. Here, in considering the magnetostrictive deformations, we have neglected the anisotropy of Poisson's ratio μ and Young's modulus E, since according to our experimental results on measurement of Young's modulus in longitudinal and torsional oscillations[5], this anisotropy is small (~ 10%), whereas the magnetostriction due to the paramagnetism of the rare-earth ions is markedly anisotropic.

Similarly, for a field applied along the a axis of the crystal,

$$F_{\mathrm{me}} = \Lambda_1 m_x^2 u_{xx} + \Lambda_3 m_x^2 u_{yy} + \Lambda_5 m_x^2 u_{zz},$$

from which the magnetostrictive deformations along the crystal axes a, b, and c can be calculated.

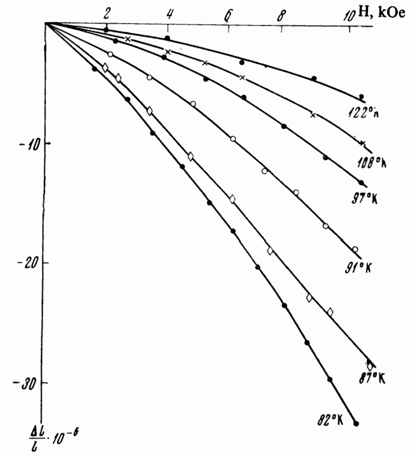

FIG. 3. Dependence of the magnetostriction along the a axis of a monocrystal of thulium orthoferrite on the field applied along the c axis of the crystal.

By substituting in these relations our measured values of the magnetostriction along axes a, b, and c, with fields applied along the c axis and along the a axis, one can calculate, at a definite temperature, the values of the magnetoelastic constants. For temperature 120°K, the following values are obtained:

$$\Lambda_1 = (6 \pm 1)\cdot 10^6; \quad \Lambda_2 = (-8{,}5 \pm 2)\cdot 10^6; \quad \Lambda_3 = (-14 \pm 2)\cdot 10^6;$$
$$\Lambda_4 = (15 \pm 2)\cdot 10^6; \quad \Lambda_5 = (0{,}5 \pm 0{,}5)\cdot 10^6; \quad \Lambda_6 = (-0{,}5 \pm 0{,}5)\cdot 10^6;$$

(Λ_i are given in erg $g^2/\mathrm{gauss}^2\ \mathrm{cm}^9$).

We take for the value of Young's modulus, in accordance with [5], $E = 1.9 \times 10^{12}$ erg/cm^3; and for the value of Poisson's ratio, $\mu = 0.4$.

Knowing the magnetoelastic constants, we can calculate the paramagnetic magnetostriction for application of the magnetic field along the b axis; the magnetoelastic energy in this case has the form

$$F_{\mathrm{me}} = \Lambda_2 m_y^2 u_{xx} + \Lambda_4 m_y^2 u_{yy} + \Lambda_6 m_y^2 u_{zz}.$$

The calculated and directly measured values of the magnetostriction agree with each other within the limits of experimental error; this indicates the correctness of our treatment. Thus we have found, in agreement with experimental results, that even on the assumption of isotropy of the elastic energy, the magnetoelastic energy of orthoferrites resulting from the paramagnetism of the rare-earth ions is markedly anisotropic.

We shall now consider the behavior of orthoferrites near the temperature of reorientation of the spins, where, as was noted above, the dependence of the magnetostriction on the field has a more complicated character (Figs. 1–3). It is known that in thulium orthoferrite, the spontaneous reorientation of the spins of the iron ions is observed in the temperature range 80–92°K. Because of this, on application of a sufficiently large magnetic field along the c axis of the crystal, which is the axis of antiferromagnetism of the iron ions at low temperatures, besides the magnetostriction due to the paramagnetism of the rare-earth ions there should be observed a magnetostriction resulting from inversion of the antiferromagnetic iron sublattices[4]. The "inversion" magnetostriction is best examined along the c axis of the crystal (Fig. 1), since along this axis the magnetostriction due to the paramagnetism of the rare-earth ions is small. The presence of a weakly ferromagnetic moment along the c axis of the crystal at high temperatures likewise does not make an appreciable contribution to the amount of the magnetostriction, since magnetostriction along the c axis occurs chiefly upon rotation of the antiferromagnetism vector of the iron ions. To determine the value of the "inversion" magnetostriction along the various axes of the crystal, it is necessary to subtract from the total magnetostriction the part due to the paramagnetism of the rare-earth ions. As a result, the following values can be obtained for the magnetostriction caused by inversion of the sublattices, along the a, b, and c axes of the crystal:

$$\lambda_a = (-11 \pm 2)\cdot 10^{-6}, \quad \lambda_b = (-11 \pm 2)\cdot 10^{-6}, \quad \lambda_c = (28 \pm 3)\cdot 10^{-6}.$$

It should be noted that for monocrystals grown from a melt of lead compounds, the magnetostriction along the c axis had a slightly smaller value: $\lambda_c = 20 \cdot 10^{-6}$[4]. By equating the values obtained for the magnetostriction along the a, b, and c axes to the deformations of the lattice along the corresponding axes upon spontaneous reorientation of the spins, one can determine, in accordance with[5], the values of the three magnetoelastic constants in thulium orthoferrite:

$$L_x = \lambda_a E_x = (-2.2 \pm 0.4) \cdot 10^7 \text{ erg/cm}^3,$$
$$L_y = \lambda_b E_y = (-2.2 \pm 0.4) \cdot 10^7 \text{ erg/cm}^3,$$
$$L_z = \lambda_c E_z = (5.6 \pm 0.6) \cdot 10^7 \text{ erg/cm}^3.$$

The values found for the magnetoelastic constants agree well with those obtained in[5] from the formula $L_i = \sqrt{K_2 \Delta E_i}$, where ΔE_i is the discontinuity of Young's modulus along the a, b, or c axis of the crystal upon reorientation of the spins, and K_2 is the second anisotropy constant:

$$L_x = (-2.2 \pm 0.2) \cdot 10^7 \text{erg/cm}^3; L_y = (-2.8 \pm 0.3) \cdot 10^7 \text{erg/cm}^3;$$
$$L_z = (5 \pm 0.5) \cdot 10^7 \text{ erg/cm}^3.$$

Determination of the magnetoelastic constants from measurements of magnetostriction has this advantage over the method described in[5], that it permits determination both of the magnitude and of the sign of the magnetoelastic constants.

Knowing the magnetoelastic and elastic constants, one can estimate the contribution of the magnetoelastic and elastic energies to the anisotropy energy. Far from the temperature where reorientation of spins is observed, the anisotropy energy ($\sim 10^5$ erg/cm^3) considerably exceeds the magnetoelastic and elastic energies due to the iron ions ($\sim 10^2$ erg/cm^3). But near the reorientation temperature, the anisotropy constant goes through zero, and the influence of the magnetoelastic and elastic energies becomes important.

We also made measurements of the longitudinal magnetostriction along the a, b, and c axes of the crystal at liquid helium temperature, in the field of a superconducting solenoid, up to 50 kOe (see Fig. 4). It is seen from Fig. 4 that at 4.2°K the magnitude of the magnetostriction has greatly increased. The largest value of the magnetostriction was observed along the c axis of the crystal, where at field ~45 kOe a value up to $-4 \cdot 10^{-4}$ was reached. The magnitude of the longitudinal magnetostriction along the a, b, and c axes is negative and has an approximately linear dependence on $(\chi H)^2$ (see Fig. 5); this indicates that it is chiefly due to the paramagnetism of the rare-earth ions. The magnetostriction along the c axis of the crystal in a strong magnetic field disclosed an abrupt break, apparently due to the fact that the c axis is, at low temperatures, the axis of antiferromagnetism of the spins of the Fe^{3+} ions, and a field ~30 kOe is sufficient to cause inversion of the iron sublattices. This hypothesis is supported by the fact that the magnitude of the magnetostriction observed by us earlier along the c axis upon inversion of the iron sublattices, in the temperature range in which reorientation of the spins occurs (Fig. 1), agrees with the amount of the jump of the magnetostriction at 4.2°K at the location of the break in the curve $\Delta l/l = f(H)$, and has the same sign.

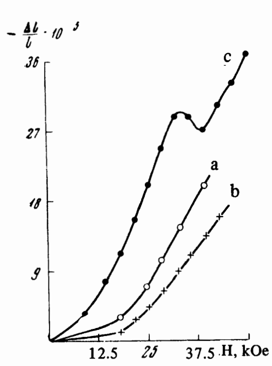

FIG. 4

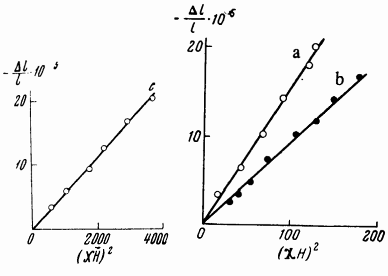

FIG. 5

FIG. 4. Dependence of the absolute value of the longitudinal magnetostriction along the a, b, and c axes of a monocrystal of thulium orthoferrite on the field at 4.2°K. ●, a axis; ○, b axis; ×, c axis.

FIG. 5. Dependence of the longitudinal magnetostriction of thulium orthoferrite on the square of the magnetization at 4.2°K: left plot, along the c axis of the crystal; right plot, along the b and a axes of the crystal.

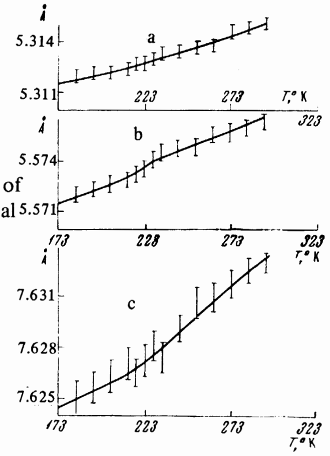

FIG. 6. Temperature dependence of the lattice parameters of a monocrystal of $Tm_{0.5}Sm_{0.5}FeO_3$.

The deformation of the lattice that is observed on application of the threshold field along the axis of antiferromagnetism of the iron ions should occur also, even in the absence of a field, in the temperature range in which spontaneous reorientation of the spins occurs. In order to measure directly the lattice deformation that occurs on reorientation of the spins, a measurement was made of the temperature dependence of the lattice parameters in the orthoferrite $Tu_{0.5}Sm_{0.5}FeO_3$, for which the temperature range for reorientation of the spins has its center at 223°K. It should be noted that for thulium and samarium orthoferrite and for the mixed composition $Tu_xSm_{1-x}FeO_3$, the causes that lead to the reorientation of the spins are so far unknown. From magnetic measurements it follows that in these compounds the reorientation of the spins is not caused by interaction of the rare-earth and iron ions, as is the case, for example, in the orthoferrites of holmium, erbium, and ytterbium[6].

In the measurements of the temperature dependence of the lattice parameters of $Tu_{0.5}Sm_{0.5}FeO_3$ in the temperature interval 173–300°K, it was ascertained that the lattice parameters along the a, b, and c axes change with temperature anisotropically (Fig. 6). It is possible that the anisotropic change of the lattice parameters with temperature leads to a change of sign of the anisotropy constant and is the reason for the reorientation of the spins in this compound; the more so, because according to published data[7], for lanthanum orthoferrite, for which the phenomenon of spin reorientation is not observed, the lattice parameters change with temperature practically isotropically. As regards the jump in the lattice parameters on reorientation of the spins, its value, as is seen from Fig. 6, lies within the limits of experimental error, and consequently cannot be determined from our measurements. Thus it is apparently safest to determine the amount of the deformation on reorientation of the spins from measurements of the magnetostriction that occurs on inversion of the antiferromagnetic iron sublattices, especially for those orthoferrites in which the magnetostriction due to the paramagnetism of the rare-earth ions is small.

In closing, we should like to express our deep appreciation to Professor M. M. Umanskiĭ, under whose direct supervision the x-ray measurements were made; to Senior Scientist V. A. Timofeeva, of the Institute of Crystallography, Academy of Sciences, USSR, for providing the monocrystal of $Tu_{0.5}Sm_{0.5}FeO_3$ for our measurements; and also to A. S. Pakhomov, for his participation in the discussion of the results and for valuable counsel.

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