Direct observation of a symmetry change induced in orthoferrite crystals by an external magnetic field

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Calculations predict monoclinic distortions of an orthoferrite lattice (a change in crystal class from D_{2h} to C_{2h}) in a spin-flip transition induced by an external magnetic field. These distortions have also been observed experimentally.

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Changes in the linear dimensions of crystals in the region of spin-flip transitions, both spontaneous changes and those induced by an external magnetic field, are familiar effects which have been studied thoroughly in the laboratory for a wide range of crystals, including orthoferrites.[1]

In addition to the ordinary magnetostrictive deformations, which do not involve a change in the crystal space group, spin-flip transitions in which the magnetic symmetry is lowered should be accompanied by lattice deformations that lower the crystal symmetry. In particular, for the rare-earth orthoferrites $RFeO_3$ or orthochrimites $RCrO_3$, the spin-flip transitions $\Gamma_4 \leftrightarrow \Gamma_2$, $\Gamma_2 \leftrightarrow \Gamma_1$, $\Gamma_1 \leftrightarrow \Gamma_4$, which are accompanied by smooth spin rotations in the *ac*, *bc*, and *ab* planes, respectively, should be accompanied by a lowering of the crystal class from D_{2h} to C_{2h} (Ref. 2), as a result of the appearance of shear strains ϵ_{ac}, ϵ_{bc}, and ϵ_{ab}, respectively.

Let us consider in more detail the spin-flip transition $\Gamma_4 \rightarrow \Gamma_2$, in which the spins of the Fe^{3+} ions rotate in the *ac* plane through two, second-order phase transitions.

In this case the magnetoelastic energy can be written as follows[3]:

$$\Phi_{\text{me}} = (L_a \epsilon_{aa} + L_b \epsilon_{bb} + L_c \epsilon_{cc}) \sin^2\theta + \frac{1}{2} \mu_2 \epsilon_{ac} \sin 2\theta, \tag{1}$$

where θ is the angle specifying the orientation of the weak, ferromagnetic moment, measured from the c axis, and $L_{a,b,c}$ and μ_2 are magnetoelastic constants. Using the standard expression for the elastic energy of orthoferrites,[3] we find the shear strain in the ac plane to be

$$\epsilon_{ac} = -\frac{\mu_2}{8C_{55}} \sin 2\theta = \epsilon_{ac}^{(o)} \sin 2\theta, \tag{2}$$

where C_{55} is the shear elastic constant. Analysis of (2) leads to three important conclusions.

(1) The shear strain, which is a measure of the extent to which the crystal is monoclinic, vanishes at the boundaries of the region of the spin-flip transition ($\theta = 0$, π, $\pm\pi/2$) and reaches a maximum at the center of this region ($\theta = \pm\pi/2, \pm 3\pi/4$).

(2) Near the phase-transition points, i. e., near the beginning or end of the spin rotation, the shear strain is linear in the order parameter, in contrast with the "ordinary" strains ϵ_{aa}, ϵ_{bb}, and ϵ_{cc}.

(3) The sign of the shear strain ϵ_{ac} is different for domains with different magnetization directions ($\pm\theta$).

The magnetoelastic anisotropy in orthoferrites is determined primarily by the effect of the strains on the noncubic crystal fields acting on the Fe^{3+} ions (a single-ion crystalline anisotropy is also implied here) and on the magnetic dipole interaction and antisymmetric exchange. Our analysis of various mechanisms for magnetoelastic coupling shows that the parameters $L_{a,b,c}$ for $YFeO_3$ are determined primarily by the mechanism involving the distortions of the octahedron Fe^{3+}-$6O^{2-}$, while the shear magnetoelastic constant μ_2 is also affected substantially by the magnetic dipole mechanism[4] and by the lattice, considered in the point-charge model. The theory predicts that $\mu_2 \approx 1.2 \times 10^7$ erg/cm^3 for $YFeO_3$. Using the experimental value $C_{55} = 0.87 \times 10^{12}$ erg/cm^3 from Ref. 5, we find a theoretical prediction of the maximum shear strain: $\epsilon_{ac}^{(0)} = -3.4 \times 10^{-6}$.

The simplest way to observe the shear strain is to measure the magnetostriction at an angle of 45° from the a and c axes in the ac plane in the spin-flip region. In this case,

$$\left(\frac{\Delta l}{l}\right)_{45^\circ} = \epsilon_{ac}^{(o)} \sin 2\theta + \frac{1}{2} (\epsilon_{aa}^{(o)} + \epsilon_{cc}^{(o)}) \sin^2\theta, \tag{3}$$

where $\epsilon_{aa}^{(0)}$, $\epsilon_{cc}^{(0)}$ are the maximum strains along the a and c axes during the spin flip.

In the orthoferrite $YFeO_3$, the spin-flip transition $\Gamma_4 \rightleftarrows \Gamma_2$ is caused in the ac plane by an external magnetic field H_a applied along the a axis, with a threshold value $H_t \approx 72$ kOe (Ref. 1).

Using the experimental results of Ref. 1 on the behavior $\epsilon_{aa}(H_a)$ and $\epsilon_{cc}(H_a)$

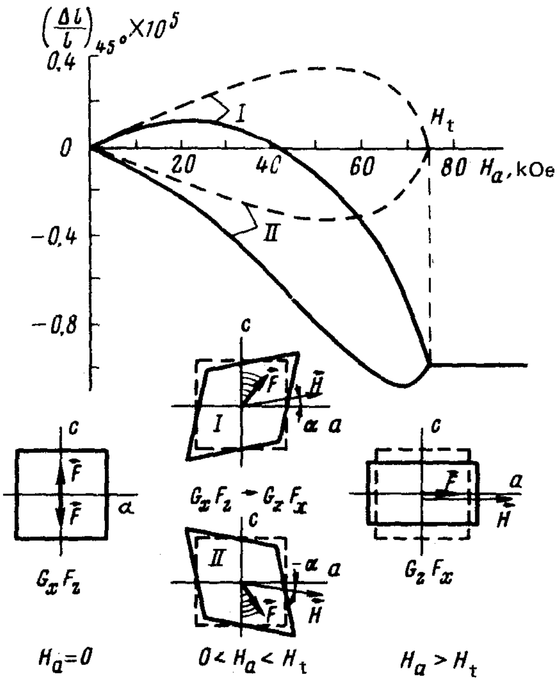

FIG. 1. Theoretical prediction of the magnetostriction $(\Delta l/l)_{45°}$ in the *ac* plane as a function of the external magnetic field H‖a. Curves I and II were found for single-domain samples differing in the original magnetization direction (the difference in the monoclinic distortions during the spin-flip transition is shown in the diagram). The dashed curve shows the *H* dependence of the relative shear strain.

and theoretical estimates of $\epsilon_{ac}^{(0)}$, we calculated $(\Delta l/l)_{45°}$ as a function of the field H_a in $YFeO_3$ (Fig. 1). Curves I and II in Fig. 1 were found for a reorientation of single-domain samples, differing in the original magnetization direction (that at $H_a = 0$). The situation is illustrated well by the inset in Fig. 1, which shows the difference between the monoclinic distortions in the spin-flip region for domains with different "past histories."

The difference in the sign of the shear strain for domains in different orientations makes it an exceedingly difficult matter to experimentally observe a change in crystal symmetry during a spontaneous spin flip.

In order to directly observe the monoclinic distortions which arise in the spin-flip transition $\Gamma_4 \rightleftarrows \Gamma_2$ induced by a field in an $YFeO_3$ single crystal, we measured the magnetostrictive strains $(\Delta l/l)_{45°}$ while a magnetic field was applied along the *a* axis of an orthorhombic crystal. As expected, the field dependence of the magnetostriction is complicated (Fig. 2), implying a superposition of two types of magnetostrictive strain: an ordinary strain, $\sim\sin^2\theta$, which reaches a maximum $\epsilon_{aa}^{(0)} + \epsilon_{cc}^{(0)}/2 \approx -1{,}2\cdot 10^{-5}$ upon the completion of the spin-flip transition, and a shear strain, which reaches a maximum at the center of the region of the spin-flip transition. The di-

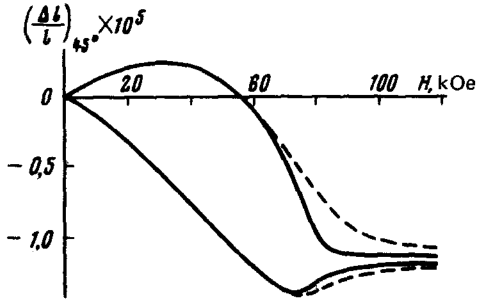

FIG. 2. Experimental dependence of the magnetostriction in the *ac* plane, at an angle of 45° from the axes, on the external magnetic field $H \parallel a$. Solid curves—$\alpha = \pm 0.5°$; dashed curves—$\alpha = \pm 2°$ (α is the angle between the field H and the a axis).

rection in which the Fe^{3+} spins rotate was changed (and thus the sign of the shear strain was changed) by applying the field H_a at an angle of +0.5° or -0.5° from the *a* axis; this measure had essentially no effect on the change in the ordinary magnetostriction, but it caused a radical change in the nature of the dependence of $(\Delta l/l)_{45^\circ}$ on the magnitude of the field H_a, in complete agreement with the theory. In this manner we have unambiguously established the existence of shear strains ϵ_{ac} and thus a lowering of the $YFeO_3$ symmetry from orthorhombic to monoclinic in the external magnetic field H_a. The experimental H_a dependence of $(\Delta l/l)_{45^\circ}$ agrees both qualitatively and quantitatively with the theoretical predictions. The monoclinic distortions reach a maximum magnitude $\sim 0.3 \times 10^{-5}$.

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