

model taking spin-lattice relaxation effects approximately into account. Of particular interest is the result that for fields applied in the easy direction in a temperature interval  $T_1 < T < T_C$ , the instability occurs with respect to a change of magnetization magnitude. Associated with this transition are a soft parallel relaxation mode and critical fluctuations of magnetization magnitude.

For experimental observation of such behavior a very hard ferromagnet is required, because the temperature interval  $T_C - T_1$  is of order  $(K/J)T_C$ . It has further to be noticed that the transition is of first order, and the ideal stability limit usually cannot be reached because of local nucleation of the reversed phase at imperfections, etc. It will, therefore, be necessary to infer the

type of transition by an extrapolation procedure. If a suitable system can be found, it should be possible to study also the dynamic properties, because all relevant frequencies are in the microwave range or below.

The isotropic ferromagnet represents a singular case because  $T_1 \rightarrow T_C$  for  $K \rightarrow 0$ , and both the perpendicular and the parallel mode become soft at  $T = T_C$ . This may be the origin for some of the difficulties encountered in theoretical treatments of the phase transition of this system.

#### ACKNOWLEDGMENT

I am indebted to Dr. H. Rohrer for help with the numerical calculations.

PHYSICAL REVIEW

VOLUME 187, NUMBER 2

10 NOVEMBER 1969

### Direct Observation of Spin Rotation in $\text{ErFeO}_3$ †

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(Received 6 May 1969)

We present conclusive evidence that the magnetic-spin reorientation occurring in  $\text{ErFeO}_3$  at about 95°K proceeds by spin rotation.

**I**N a number of the weak ferromagnetic rare-earth orthoferrites (formula  $R\text{FeO}_3$ , where  $R$  is a rare earth), the direction of the magnetically ordered iron spins changes from the  $c$  to the  $a$  crystal axis on heating.<sup>1-5</sup> We report, in this article, a direct observation of the spin reorientation occurring in  $\text{ErFeO}_3$  using the Mössbauer effect. In agreement with previous studies<sup>4,5</sup> our data provide conclusive evidence that the spins rotate continuously from the  $c$  to the  $a$  axis on heating.

Microscopic<sup>2</sup> and macroscopic<sup>3</sup> descriptions of the

spin reorientation have indicated the possibility of two types of reorientation process:

(a) The spin magnetic moment rotates continuously from the  $c$  axis to the  $a$  axis, the rotation beginning at  $T = T_1$ , and ending at  $T = T_2$ , say.

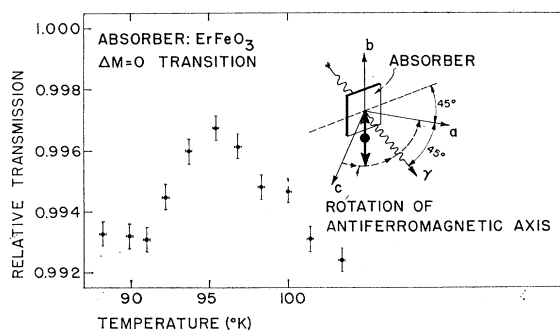


FIG. 1. Relative Mössbauer transmission of  $\text{Fe}^{57}$  in  $\text{ErFeO}_3$  as a function of temperature. The source-absorber velocity was kept constant at the value corresponding to the peak of the  $\Delta m = 0$  absorption line. Below about 90°K, the spins lie along the  $c$  axis. On heating, the spins rotate towards the  $a$  axis. For  $T \approx 95^\circ\text{K}$ , the spins lie approximately parallel to the  $\gamma$ -ray direction, and the intensity of the  $\Delta m = 0$  line is reduced. A relative transmission of 1.000 corresponds to the transmission off resonance.

† Sponsored in part by the Air Force Materials Laboratory Research and Technology Division AFSC through the European Office of Aerospace Research, United States Air Force, under Contract No. F 61052-67C-0040.

\* Part of a Ph.D. thesis to be submitted to the Feinberg Graduate School of the Weizmann Institute of Science.

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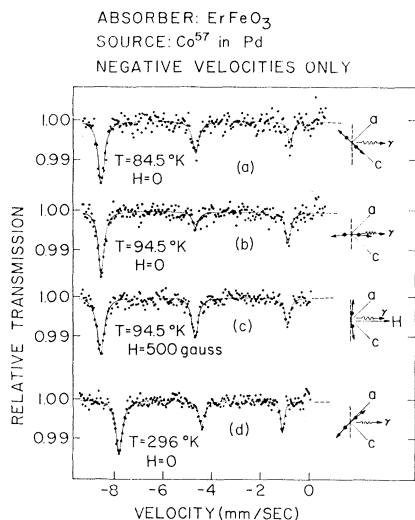


FIG. 2. Mössbauer absorption spectra of  $\text{Fe}^{57}$  in  $\text{ErFeO}_3$ . Only that portion of the spectrum corresponding to negative velocities is given (the complete spectra are closely symmetrical about  $V = +0.1$  mm/sec). The central peak of the spectra shown corresponds to the  $\Delta m = 0$  absorption line. The full line is a least-squares fit of three Lorentzian lines to the experimental data points. For each temperature and applied magnetic field  $H$ , the approximate orientation of the spins relative to the  $\gamma$  ray and crystal axes is indicated. The dotted line in the sketches gives the plane of the absorber.

(b) The spin magnetic moment jumps discontinuously from the  $c$  to the  $a$  axis, the actual jump temperature of different spins varying over the range  $T_1 < T < T_2$ , due to local departures of the specimen from perfection.

Previous measurements have supported<sup>4,5</sup> the occurrence of process (a) in  $\text{SmFeO}_3$  and in  $\text{TmFeO}_3$ . Our Mössbauer study provides direct evidence for process (a) in  $\text{ErFeO}_3$ .

A single crystal of  $\text{ErFeO}_3$  was cut into  $50\text{-}\mu$  sections along a plane whose normal lies in the  $ac$  plane, making an angle of  $45^\circ$  with respect to both the  $a$  and  $c$  axes. A mosaic absorber was constructed and oriented with respect to the  $\gamma$ -ray direction as shown in Fig. 1. If the spins rotate as indicated, at some temperature during the rotation process the spins will lie parallel to the  $\gamma$  ray. The intensity of the  $\Delta m = 0$  Mössbauer peak will then be zero.<sup>6</sup> If process (b) occurs, the  $\Delta m = 0$  line will show no intensity change.

<sup>6</sup> G. K. Wertheim, *The Mössbauer Effect* (Academic Press Inc., New York, 1964).

Figure 1 gives the relative transmission as a function of temperature, the source-absorber velocity being held fixed at the value corresponding to the peak of the  $\Delta m = 0$  absorption line. The clear drop in absorption intensity in this line indicates the occurrence of process (a). The residual absorption intensity at  $T \approx 95^\circ\text{K}$  probably arises partly from crystalline inhomogeneity and small misalignments of the crystal sections comprising the mosaic absorber. More importantly, we believe that slight crystal imperfection causes different spins to rotate over somewhat different temperature ranges.

In Figs. 2(a), 2(b), and 2(d) we display the Mössbauer spectra (for negative velocities only) at the indicated temperatures. The decrease in intensity of the  $\Delta m = 0$  peak at  $T \approx 95^\circ\text{K}$  is evident.

The effect of a 500-G external magnetic field [Fig. 2(c)] applied parallel to the  $\gamma$ -ray direction is noteworthy. At first sight there would appear to be no reason for spin rotation to occur in an anticlockwise direction on heating, as indicated in Figs. 2(a), 2(b), and 2(d), rather than say in a clockwise direction. If the latter were the case, then the  $\Delta m = 0$  line would attain a maximum<sup>6</sup> at  $T \approx 95^\circ\text{K}$ . Clockwise rotation, however, turns the spontaneous weak ferromagnetic moment perpendicular to the plane of the crystal section, and thereby increases the demagnetizing field; thus, anticlockwise rotation is preferred in general. On the other hand, the application of a magnetic field perpendicular to the plane of the absorber will favor clockwise spin rotation [Fig. 2(c)]. The qualitative increase in intensity in the  $\Delta m = 0$  absorption line is apparent. The nonattainment of the theoretical<sup>6</sup> 3:4:1 absorption line intensity ratio for the spin configuration depicted in Fig. 2(c) is probably due to the same sort of effects giving rise to the residual absorption noted in Figs. 1 and 2(b), as discussed previously.

In conclusion, we wish to mention that Ozhogin *et al.*<sup>7</sup> have used the Mössbauer effect to observe spin rotation in  $\alpha\text{-Fe}_2\text{O}_3$  upon the application of an external field. He concludes that the sense of spin rotation is an intrinsic property of  $\alpha\text{-Fe}_2\text{O}_3$ . By contrast, our measurements indicate that the direction of spin rotation is, in fact, determined by the details of the particular experimental set up.

One of the authors (B. M. W.) acknowledges the support of the Science Research Council, England.

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