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1993 J. Phys.: Condens. Matter 5 L99

(http://iopscience.iop.org/0953-8984/5/7/004)

J. Phys.: Condens. Matter 5 (1993) L99–L102. Printed in the UK

LETTER TO THE EDITOR

New field-driven spin reorientations in $YbFeO_3$ at 4.2 K

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Received 21 December 1992

Abstract. Reorientation of the Fe spins in $YbFeO_3$ has been studied at 4.2 K using ^{57}Fe Mössbauer spectroscopy and applied magnetic fields up to 10 T. In addition to the expected reorientation (spin flop) induced by a field applied along the easy antiferromagnetic axis, unusual reorientations were observed for fields applied perpendicular to the easy axis. It is suggested that these are due to breaking of the Fe–Yb coupling.

The rare-earth orthoferrites $RFeO_3$, where R is a rare-earth ion, are antiferromagnets with ordering temperatures for the Fe spins typically in the region of 650 K. In some cases ordering of the rare-earth spins has also been observed at very low temperatures ($\lesssim$ 4 K). The Fe spins are normally canted at a small angle ($\lesssim$ 10 mrad) so that these materials exhibit weak ferromagnetism in a direction perpendicular to the easy antiferromagnetic axis. The spin configurations may be succinctly described in the notation of Bertaut [1]. At normal temperatures the Fe spins in most rare-earth orthoferrites have the $\Gamma_4(G_xA_yF_z)$ configuration, i.e. antiferromagnetic along the crystal a axis and weakly ferromagnetic along the crystal c axis. The compatible configuration of the paramagnetic rare-earth spins, polarized by the Fe–rare-earth interaction, is $\Gamma_4(O_x, O_y, F_z)$, i.e. net alignment along the crystal c axis.

A notable feature of the rare-earth orthoferrites is the phenomenon of spin reorientation (SR) in which the Fe spins rotate through 90° to lie along another crystal axis. The SR may be induced by temperature change or by application of a magnetic field. Several orthoferrites with magnetic rare-earth ions (e.g. R = Tb, Ho, Er, Yb) undergo a transition at low temperature wherein, over a temperature interval about a reorientation temperature T_R, the Fe spins rotate continuously from their high-temperature configuration Γ_4 to $\Gamma_2(F_xC_yG_z)$ so that the antiferromagnetic axis is then the crystal c axis. At a temperature above T_R the $\Gamma_4 \rightarrow \Gamma_2$ transition may be driven by applying a magnetic field along the magnetic easy axis (the crystal a axis) so that the antiferromagnetically coupled Fe spins 'flop' to being perpendicular to the applied field. Similarly, at a temperature below T_R, the $\Gamma_2 \rightarrow \Gamma_4$ transition may be induced by a field applied along the crystal c axis. Generally speaking, such field-driven transitions require higher fields, the higher the value of $|T - T_R|$. Yamaguchi [2] has given a detailed, microscopic theory of SR; [2] also contains a fuller summary of the various types of SR than is appropriate here.

0953-8984/93/070099+04$07.50

While SR has been studied with a number of techniques we focus here on its observation by Mössbauer spectroscopy, by which spin directions may be directly monitored. In a magnetically split (six-line) ^{57}Fe Mössbauer absorption spectrum the 1,6:2,5:3,4 line intensity ratio is $3:4\sin^2\theta/(1+\cos^2\theta):1$ where θ is the angle between the γ-ray direction and the magnetic hyperfine field $\boldsymbol{B}_{\rm hf}$, the direction of the latter being determined by the Fe ion spin. Thus the relative intensity of the 2,5 lines varies significantly from 3:0:1 to 3:4:1 as the spins rotate through 90°. In the case of an applied-field ($\boldsymbol{B}_{\rm app}$) experiment the nucleus senses an effective field

$$\boldsymbol{B}_{\rm eff} = \boldsymbol{B}_{\rm hf} + \boldsymbol{B}_{\rm app}$$

and, because of the equal and opposite $\boldsymbol{B}_{\rm hf}$ from the two antiferromagnetic sublattices, the Mössbauer spectrum shows two sextets with slightly different $\boldsymbol{B}_{\rm eff}$. This effective-field splitting is, of course, also sensitive to the orientation of the Fe spins, being greatest when these are parallel and antiparallel to the applied field and zero when they are perpendicular to the applied field. In practice, as has been done in the present work, the two effects of line intensity and of field splitting may be incorporated into one computational model for spectrum fitting. For this we have used a model developed from that of Kündig [3] in which line positions and intensities are computed as a function of the hyperfine interaction parameters and the relative orientations of $\boldsymbol{B}_{\rm hf}$, $\boldsymbol{B}_{\rm app}$, and γ-ray direction. [4] is an earlier example of the application of Mössbauer spectroscopy to SR in orthoferrites.

In $YbFeO_3$ T_R is quite low, the $\Gamma_2 \rightleftharpoons \Gamma_4$ SR taking place over 6.5–7.8 K [5]. Since this is just above liquid helium temperature it is experimentally convenient to study field-driven SR just below T_R; also at low temperature the effect of the strongly polarized Yb ions should be quite pronounced. Preliminary experiments with powder samples of $YbFeO_3$ gave unexpected spectra at high applied fields and this motivated the single-crystal experiments reported here. $YbFeO_3$ crystals prepared by R L White of Imperial College using the flux-growth technique were cut by diamond saw to 1 mm slices, and then thinned by abrasion to produce ab- and bc-plane absorbers set in low-temperature epoxy. Mössbauer spectra were taken at 4.2 K in applied fields up to 10 T, using ^{57}Co/Rh sources and a constant-acceleration spectrometer operated in double-ramp (flat-background) mode. The experiments were conducted in transverse geometry, i.e. applied field perpendicular to the γ-ray direction. Three absorber configurations were used and it is convenient to label them using an underline to signify the direction of the applied field. Thus, for example, $b\underline{c}$ means use of a bc-plane absorber (γ-rays along the crystal a axis) oriented such that the applied field is along the crystal c axis. The $b\underline{c}$ spectra showed the expected flop SR, the Fe spins rotating from the c axis (Γ_2) to the a axis (Γ_4).

Unusual results were, however, obtained with ab-plane absorbers, examples of spectra being shown in figure 1. At zero field the spectrum shows negligible 2,5 line intensity as expected. In $\underline{a}b$ geometry and at 5 T the spectrum was unchanged, showing the Fe spins still to be aligned along the c axis. At higher fields some 2,5 line intensity developed, showing the spins to have moved away from the c axis; there was an accompanying development of field splitting, showing that this movement was towards the axis of $\boldsymbol{B}_{\rm app}$, i.e. towards the a axis in this case. The $\underline{a}b$ 10 T spectrum is shown in figure 1, the fit giving a rotation of the Fe spins of $43 \pm 5°$ from the c axis towards the a axis. In $a\underline{b}$ geometry and for $B_{\rm app} \geqslant 5$ T there was also a development of 2,5 line intensity but this time with very little, if any, field splitting.

This implies spin rotation, this time around B_{app}, but again towards the a axis. The $a\underline{b}$ 10 T spectrum is shown also in figure 1, the fit giving a rotation of $73 \pm 5°$. The possible (unresolved) field splitting indicates the spins being out of the ac plane by no more than approximately 5°. These particular SR effects as observed by Mössbauer spectroscopy are unambiguous and straightforward but, to our knowledge, have not been recorded before.

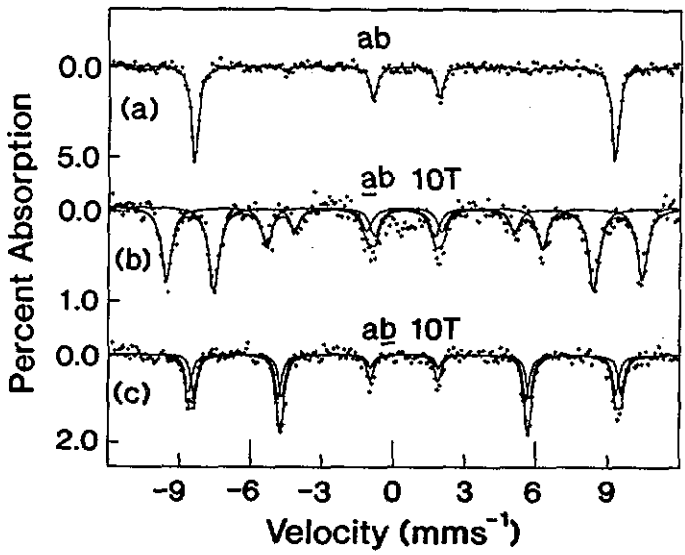

Figure 1. Mössbauer spectra from $a\underline{b}$-plane absorbers at 4.2 K (*a*) zero applied field (*b*) 10 T applied field along the a axis (*c*) 10 T applied field along the b axis.

It seems unlikely that these effects are an artefact of some absorber defect. The Mössbauer parameters deduced from our spectra are in good agreement at 4.2 K and room temperature with those previously reported [6] for $YbFeO_3$; we found the temperature-driven SR to occur at $T_R \approx 7$ K, consistent with previous work [5]; and the observation of 3:0:1 relative line intensities in e.g. figure 1(*a*) demonstrates the homogeneity of spin alignment in the crystal. Furthermore, the unusual SR reported here were also observed with a second ab-plane absorber.

In seeking an explanation of these effects we note that the common feature of the two SR described above is that of the Fe spins reorienting towards the a axis. Now the high-temperature (Γ_4) phase has the Fe spins along the a axis and we may recall that the temperature-driven $\Gamma_4 \rightarrow \Gamma_2$ SR is believed [2] to be assisted by polarization of the rare-earth spins through a complicated combination of symmetric and antisymmetric exchange interactions. It seems possible then that an applied field may weaken the Fe–rare-earth coupling and, if the temperature is not far below T_R, the Fe spins recover their high-temperature configuration. In support of this suggestion we recall also the compatible configurations of the rare-earth spins: $\Gamma_4(F_z)$ for the high-temperature phase and $\Gamma_2(F_xC_y)$ for the low-temperature phase. According to Yamaguchi [2], and as verified by Davidson *et al* [7] for $YbFeO_3$, the rare-earth spins are as shown in figure 2. In the low-temperature phase, the rare-earth spins are not all collinear so that it is possible that an applied field directed anywhere in the ab plane would

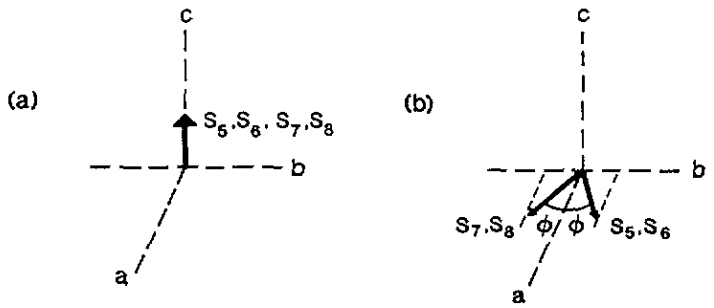

Figure 2. Rare-earth spin configurations (*a*) Γ_4 (high-temperature phase) (*b*) Γ_2 (low-temperature phase). There are four Fe ions (spins S_1–S_4 not shown) per unit cell and four *R* ions (spins S_5–S_8). *In* $YbFeO_3$ *the angle* $\phi = 27°$ at 4.2 K [7].

weaken the Fe–rare-earth coupling. Furthermore, the magnitude of field required might vary with field direction and this may explain why the SR effect was greater in the $a\underline{b}$ case than in the $\underline{a}b$ case. An alternative explanation is that in the latter case the Fe spins were rotating *towards* the applied-field axis so that the SR mechanism would have to overcome the normal tendency of antiferromagnetically coupled spins to align perpendicular to an applied field.

This work will be reported more fully elsewhere. We acknowledge discussions with C E Johnson and the support of the UK Science and Engineering Research Council.

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