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Mössbauer study of spin reorientation in $Y_{1.8}Er_{0.2}Fe_{14}B$

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Fe^{57} Mössbauer spectra were taken from 300 K down to 4.2 K for two single-crystal samples of $Y_{1.8}Er_{0.2}Fe_{14}B$, one cut with its [001] axis normal to the surface, the other with its diagonal of [100], [010], and [001] axes normal to the surface. The former sample has shown that the angle between the Fe spin and the [001] increases continuously below ~ 90 K. This qualitatively agrees with the spin reorientation (i.e., the easy axis of magnetization rotates away from the [001] toward [100] on cooling below ~ 90 K) which have already been observed by torque magnetometry. The latter sample has shown that the Fe spin actually rotates continuously; abrupt change of Fe spin direction, which have been already suggested to occur in $Er_2Fe_{14}B$, does not occur in $Y_{1.8}Er_{0.2}Fe_{14}B$. The samples were so thick that the intensity of strong absorption lines were saturated. We have derived a formula with which the saturation effect can be corrected in order to determine the angle of the Fe spin direction correctly.

I. INTRODUCTION

Some of the $R_2Fe_{14}B$ (R = rare-earth elements) compounds exhibit spin reorientation transitions. In $Nd_2Fe_{14}B$, the easy axis of magnetization is directed along the [001] axis at room temperature, but rotates away from the [001] toward the [110] below ~ 135 K,¹ the rotation angle reaching $\sim 27^\circ$ at 4.2 K.² Similar spin reorientation has also been observed in $Ho_2Fe_{14}B$.³ In these compounds, not only the Fe but also the rare-earth (Nd, Ho) ion, having a negative Stevens factor α_j , prefer to have their spins point along the [001]. So, the spin reorientation cannot be attributed to the competing anisotropy of the Fe and the rare-earth ions. On the other hand, in $Er_2Fe_{14}B$ and $Tm_2Fe_{14}B$, spin reorientation occurs due to the competition between anisotropies of the Fe ion and the rare-earth ion, Er or Tm, which has a positive α_j , preferring the easy (001) basal plane. The easy axis of magnetization in $Er_2Fe_{14}B$ ($Tm_2Fe_{14}B$) changes from the [001] to the [100] on cooling around 316 K (310 K) in a quite narrow temperature range, suggesting that the spin reorientation transition is of first order.^{3,4} Now, in $Y_{1.8}Er_{0.2}Fe_{14}B$, where magnetic Er ion is diluted with the nonmagnetic Y ion, the easy axis rotates continuously in (010) plane from the [001] toward the [100]. The magnetic moment starts to rotate at around 90 K, stopping at an angle of 78° from the [001] below ~ 40 K.⁵ Here we can consider following two models for the spin reorientation in $Y_{1.8}Er_{0.2}Fe_{14}B$:

(1) The Fe spin rotates continuously (second-order transition). (2) The Fe spin jumps discontinuously from the [100] to the [001], the actual jump temperature of different spins varying widely due to disordered arrangement of Er and Y (first-order transition smeared). The purpose of this

study is to make it clear by Mössbauer measurement which of the two models apply to $Y_{1.8}Er_{0.2}Fe_{14}B$.

II. EXPERIMENT AND RESULT

A single crystal of $Y_{1.8}Er_{0.2}Fe_{14}B$ has been grown by the Czochralsky method. It was cut into two thin ($\sim 60 \mu m$) slices, one with the [001] normal to the surface, the other with the [100], [010], and [001] making equal angles with the normal. We refer to the former as a "[001]-sample" and to the latter as a "[111] sample." Since the crystal symmetry of $Y_{1.8}Er_{0.2}Fe_{14}B$ is not cubic but tetragonal, the normal of the [111] sample does not actually coincide with the [111], but we call it so for simplicity.

Fe^{57} Mössbauer spectra were taken for the two samples from 300 to 4.2 K with a 100 mCi Co^{57} source in a Rh matrix, using a constant acceleration spectrometer.

Figure 1 shows the spectra of the two samples taken at 300 and 4.2 K. The [001] sample spectrum at 300 K is composed of four strong peaks, 1st, 3rd, 4th, and 6th lines of Fe^{57} sextuplet, and additional weak peaks. We have assigned the peaks to the six sextuplet lines, respectively, as shown by arrows in Fig. 1(a). The weak 2nd and 5th peaks become very strong as temperature is lowered to 4.2 K, showing the spin reorientation. In the [111] sample spectra, the weak 1st and 6th peaks are masked by the strong 2nd and 5th peaks, which appreciably decreases in intensity as temperature is lowered.

Since there are six Fe sites in $R_2Fe_{14}B$, to obtain a unique solution in decomposing the spectra into respective Fe site sextuplets is very difficult, especially in case of $Y_{1.8}Er_{0.2}Fe_{14}B$, where the disordered arrangement of Y and

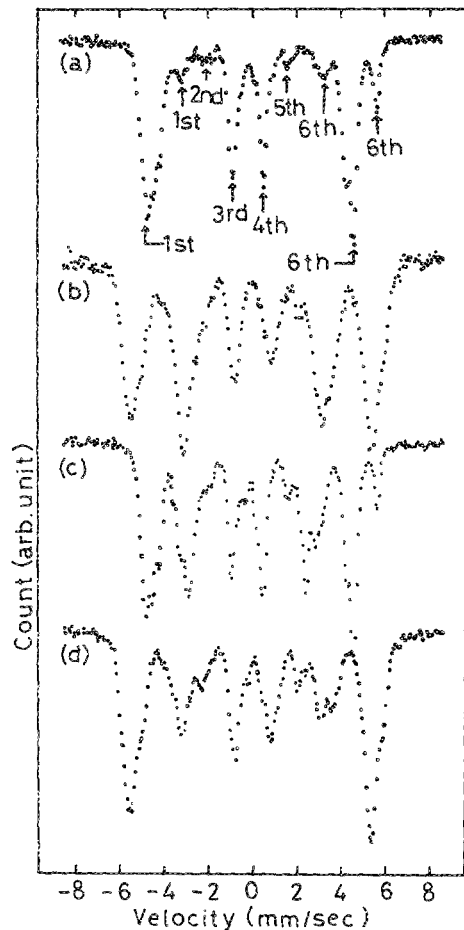


FIG. 1. Mössbauer spectra of Fe^{57} in $\text{Y}_{1.8}\text{Er}_{0.2}\text{Fe}_{14}\text{B}$ single crystal. (a) [001] sample at 300 K, (b) [001] sample at 4.2 K, (c) [111] sample at 300 K, and (d) [111] sample at 4.2 K. Arrows in (a) indicate the assignment of the peaks to Fe^{57} sextuplet lines.

Er might smear the spectrum shape. Thus, we have calculated intensities of the outermost pair (1st and 6th), middle pair (2nd and 5th), and innermost pair (3rd and 4th) peaks of the sextuplet, I_1 , I_2 , and I_3 , respectively, averaged over the six Fe sites. The intensities were calculated from the area of peaks which were graphically fitted to the observed peaks. For the [001] sample spectra above 70 K the spectra were assigned as shown in Fig. 1(a), while for those below 60 K, together with the [111] sample spectra at all temperatures, the weak 1st and 6th peaks seen in Fig. 1(a) were included in the strong 2nd and 5th peaks.

It is noted that the line intensity ratio I_3/I_1 is appreciably larger than $1/3$. This is because the samples are so thick that the intensity of a strong absorption line is saturated, being made weaker than that of a weak absorption line (so-called "saturation effect"). Correcting this effect by the way described in the Appendix, we have calculated the average spin direction angles θ and α , the former (latter) being the angle which the Fe spins on the six different sites in $\text{R}_2\text{Fe}_{14}\text{B}$, make on an average with the normal of the [001] sample ([111] sample).

Figure 2 shows θ and α as a function of temperature. The angle θ remains almost constant above 90 K, but on cooling below 90 K, the angle θ increases rapidly, reaching

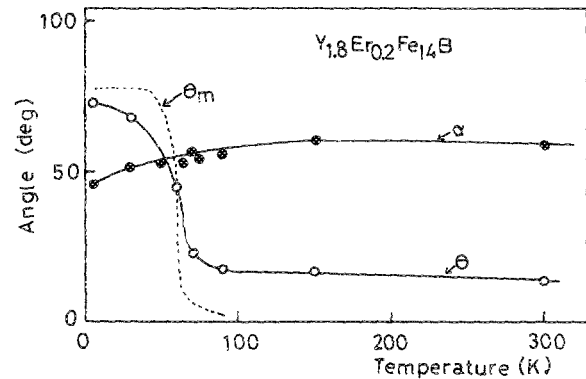


FIG. 2. Temperature dependence of the angles θ and α , which the Fe spins make with the normal of the [001] sample and the [111] sample, respectively, determined by Mössbauer spectroscopy. θ_m , which the easy axis of magnetization makes with the [001], determined by torque magnetometry,⁵ is also shown for comparison with θ .

72° at 4.2 K. The result agrees qualitatively with the torque magnetometry evidence.⁵ However, our result obtained on the [001] sample does not necessarily mean that the Fe spin rotates continuously with temperature: If the Fe spins on different sites jump abruptly from the [001] to the [100] at different temperatures, we might still observe a continuous θ vs temperature curve, because of averaging and "smearing" in bulk measurements.

For the [111] sample, on the other hand, we can distinguish whether the Fe spin rotates continuously or jumps abruptly. If the former is the case, the angle α should change continuously with temperature as the magnetic moment rotates from the [001] toward the [100]. If the latter is the case, α should remain constant, since the normal of the [111] sample makes an equal angle with the two axes [100] and [001]. As seen in Fig. 2, α changes appreciably with temperature, the Fe spins rotating continuously. Thus, the above mentioned model (1) applies to $\text{Y}_{1.8}\text{Er}_{0.2}\text{Fe}_{14}\text{B}$.

In Fig. 2 we show for comparison the angle θ_m , which the easy axis of the magnetization of $\text{Y}_{1.8}\text{Er}_{0.2}\text{Fe}_{14}\text{B}$ makes with the [001], as determined by torque magnetometry.⁵ θ deviates considerably from θ_m , especially above the spin reorientation temperature, ~ 90 K (i.e., at 300 K $\theta = 13^\circ$, while $\theta = 0^\circ$ is expressed⁵). This suggests a noncollinear (canted) alignment of the Fe sublattice moments. To reach a conclusion, however, we must examine the following:

(1) The demagnetizing field ($4\pi M_s \sim 13$ kG) in $\text{Y}_{1.8}\text{Er}_{0.2}\text{Fe}_{14}\text{B}$, which is of the same size as the anisotropy field (~ 10 kOe),⁵ may cause the magnetization to deviate from the easy axis.

(2) The Fe spins on the different six sites in this compound may have different half-widths of γ -ray energy level, whereas we have assumed the half-widths of the lines from Fe atoms on all six sites to be the same, and equal to that in the source (cf. Appendix). This may result in an error in the calculated value of θ .

(3) Misalignment in the crystal cutting and the sample mounting may give an error of $\sim 7^\circ$ in θ , as in our former case of $\text{Nd}_2\text{Fe}_{14}\text{B}$.²

(4) Fe spins in domain walls may result in an error in

the calculated value of θ . The error is estimated to be about 5° when 5% volume fraction of the domain wall is assumed.

(5) The assignment of the spectrum peaks to the sextuplet, shown in Fig. 1(a), is not appropriate.

(6) Impurity phase in the sample, though not found by x-ray diffraction, contributes to the spectrum.

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APPENDIX

Here we describe how to correct for saturation of a strong absorption line in the Mössbauer spectrum of a thick sample, and how to determine the spin direction angle θ with respect to the normal to the sample surface.

Assuming that the source is nonresonant absorbing⁶ and both the emission (source) and the absorption (sample) lines of the γ -ray have the same half-width Γ , the normalized transmission probability at a Doppler shift energy ϵ is expressed by the formula⁷ [Eqs. (11)–(13) in Ref. 7],

$$P(\epsilon) = [(1-f) + fI(\epsilon)]P_{\text{nonres.}} \quad (\text{A1})$$

Here f is the recoil-free fraction, $P_{\text{nonres.}}$ the normalized probability of the nonresonant absorption in the source, and $I(\epsilon)$ an integral function of the form

$$I(\epsilon) = \frac{2}{\pi\Gamma} \int_{-\infty}^{\infty} \frac{\Gamma^2/4}{(E+\epsilon)^2 + \Gamma^2/4} \times \exp\left(-\frac{T_A\Gamma^2/4}{E^2 + \Gamma^2/4}\right) dE. \quad (\text{A2})$$

In this formula the origin of the energy (E) axis is put at the center of an excited level in the absorber. T_A is the effective absorber thickness, which is expressed, for respective line of the Fe⁵⁷ sextuplet, as a function of the spin angle θ as follows:

for the 1st and 6th (outermost) lines:

$$T_A = T_1 = c3(1 + \cos^2 \theta); \quad (\text{A3a})$$

for the 2nd and 5th (middle) line:

$$T_A = T_2 = c4(1 - \cos^2 \theta); \quad (\text{A3b})$$

for the 3rd and 4th (innermost) lines:

$$T_A = T_3 = c(1 + \cos^2 \theta); \quad (\text{A3c})$$

c is a constant proportional to the thickness of the sample.

In the usual case $T_A \ll 1$ holds, and the exponential function in Eq. (A2) is expanded keeping only first-order term in thickness (T_A). Now we include the 2nd order term in the expansion in order to correct for the "saturation effect." Thus, we get

$$I(\epsilon) = 1 - \frac{T_A}{2} \frac{\Gamma^2}{\epsilon^2 + \Gamma^2} + \frac{T_A^2}{16} \left[\frac{\Gamma^2}{\epsilon^2 + \Gamma^2} + 2 \left(\frac{\Gamma^2}{\epsilon^2 + \Gamma^2} \right)^2 \right] \quad (\text{A4})$$

after performing the integration. From Eq. (A4) we see that

the absorption line shape is no longer Lorentzian. The area intensity I of the absorption line is given by integrating the ϵ -dependent term in Eq. (A1), or Eq. (A2), with respect to ϵ from $-\infty$ to $+\infty$, as follows:

$$I = fP_{\text{nonres.}} \left\{ -\frac{T_A}{2} \int_{-\infty}^{\infty} \frac{\Gamma^2}{\epsilon^2 + \Gamma^2} d\epsilon + \frac{T_A^2}{16} \times \int \left[\frac{\Gamma^2}{\epsilon^2 + \Gamma^2} + 2 \left(\frac{\Gamma^2}{\epsilon^2 + \Gamma^2} \right)^2 \right] d\epsilon \right\} = fP_{\text{nonres.}} \frac{T_A + T_A^2/4}{4}. \quad (\text{A5})$$

Since T_A in Eq. (A5) should be replaced by $T_1, T_2,$ and T_3 according to Eqs. (A3), the intensity ratio between the middle lines and the outermost lines is expressed

$$\frac{I_3}{I_1} = \frac{T_3(1 - T_3/4)}{T_1(1 - T_1/4)} \approx \frac{T_3}{T_1} \left[1 + \frac{1}{4}(T_1 - T_3) \right]. \quad (\text{A6})$$

Here we have neglected the second-order term of T_A in the bracket of the last equation. This means that the correction for the saturation effect is performed to first-order in thickness, which is adequate for our experimental data (i.e., $T_A \approx 0.2$ – 0.3). Substituting Eqs. (A3a) and (A3c) to Eq. (A6), we get

$$I_3/I_1 = \frac{1}{2} \left[1 + \frac{1}{2}c(1 + \cos^2 \theta) \right] \quad (\text{A7})$$

In a similar way we obtain

$$\frac{I_2}{I_1} = \frac{4(1 - \cos^2 \theta)}{3(1 + \cos^2 \theta)} \left[1 + \frac{1}{4}c(-1 + 7\cos^2 \theta) \right]. \quad (\text{A8})$$

Eliminating c from eqs. (A7) and (A8) gives

$$\frac{I_2}{I_1} = \frac{4(1 - \cos^2 \theta)}{3(1 + \cos^2 \theta)} \left[1 + \frac{1}{2} \left(3 \frac{I_3}{I_1} - 1 \right) \frac{-1 + 7\cos^2 \theta}{1 + \cos^2 \theta} \right]. \quad (\text{A9})$$

This is a quadratic equation of $\cos^2 \theta$, so that we get an analytical solution:

$$\cos^2 \theta = \frac{4a - b + \sqrt{9a^2 - 16ab + 6a + 1}}{1 + 7a + b}, \quad (\text{A10a})$$

where we have put

$$a = \frac{1}{2} \left[3(I_3/I_1) - 1 \right], \quad (\text{A10b})$$

$$b = \frac{1}{2} (I_2/I_1). \quad (\text{A10c})$$

We can determine θ using Eq. (A10).

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