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Spin reorientation in $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ compounds

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Résumé. — L'effet Mössbauer et la diffraction des rayons X sur des échantillons de ErFe_3 alignés magnétiquement ont montré qu'à température croissante la direction de facile aimantation tourne de l'axe cristallographique c à l'axe b à 50 K. Dans la série des composés $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ la température de réorientation des spins reste pratiquement la même de $x = 0$ à $x = 0,4$. Aux concentrations de cobalt plus élevées la température de réorientation augmente considérablement.

Abstract. — With Mössbauer effect and X-ray diffraction on magnetically aligned ErFe_3 it has been shown that the easy axis of magnetization turns from the crystallographic c -axis to the b -axis at 50 K with increasing temperature. In the $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ series from $x = 0$ up till $x = 0.4$ almost the same spin reorientation temperature is found. For higher Co concentration the spin reorientation temperature increases drastically.

The $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ compounds crystallize in the rhombohedral PuNi_3 structure with three non equivalent iron sites (1 b site, 2 c sites, and 6 h sites). From the literature [1, 2, 3] it follows that below 50 K ErFe_3 is a simple uniaxial ferrimagnet, though above 50 K the various authors seem to disagree with respect to the moment arrangement proposed. Therefore we have performed X-ray diffraction and ^{57}Fe Mössbauer effect measurements on magnetically aligned samples (i). As it is likely that this magnetic transition finds its origin in a competition between the Er single ion crystal field anisotropy (c -axis) and the Fe sublattice anisotropy (b -axis) [1] we have extended this investigation to the $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ pseudobinary compounds (ii), in which the c -axis anisotropy probably becomes more dominant with increasing Co concentration (YCo_3 is uniaxial).

(i) Previous investigations [1] have shown that the magnetization of a magnetically aligned powder sample of ErFe_3 at $T = 295$ K strongly decreases when it is cooled down below 50 K. This has been interpreted in terms of a 90° moment reorientation occurring at this temperature which has been confirmed with ^{57}Fe Mössbauer effect investigation. However, according to another ^{57}Fe Mössbauer effect study [2] the direction of magnetization is canted over an angle of 15° from the c -axis at $T = 50$ K and further it turns to the basal plane at $T = 133$ K. On the other hand from a magnetic neutron diffraction study [3] it is concluded that the Fe moments only reorient 56° from the c -axis at $T = 50$ K. In order to determine the easy axis of magnetization at $T = 295$ K in an independent way we have made

a Debye Scherrer diagram of a magnetically aligned powdered sample of ErFe_3 (needle shaped). From the observed nearly point reflections of the $(0, 0, l)$ planes and of the (101) plane in particular, it can be concluded that the easy axis of magnetization at room temperature lies parallel to the b -axis in the basal plane. In addition, we have carried out Mössbauer effect measurements at $T = 4.2, 80$ and 295 K on a sample, which has been magnetically aligned at $T = 295$ K parallel to the γ -ray beam. From the change in the relative intensities of the second and fifth lines it is possible to determine the angle of the spin reorientation at these temperatures with regard to each other. This relative intensity Z ($0 \leq Z \leq 4$) is given by the formula

$$Z(\varphi, q) = \frac{(4 - 4q) \sin^2 \varphi}{1 + \cos^2 \varphi} + 2q. \quad (1)$$

with φ being the angle between the γ -ray beam and the easy axis of magnetization, while q ($0 \leq q \leq 1$) gives the fraction of the sample which is not magnetically aligned. Table I gives the results of these measurements.

Supposing that ErFe_3 is ferrimagnetically ordered parallel to the b -axis in the basal plane (as it is proven by the Debye Scherrer diagram) it follows from the measurement at $T = 295$ K in an external field of 1 kOe parallel to the γ -ray beam that $q = 0.5$. When the field is removed from the sample, the spins are redistributed over the six b -axes two of which have an angle of 0° and four an angle of 60° with the γ -ray beam direction. As the same spectrum in zero field was obtained at $T = 80$ K no spin reorientation apparently has taken place between $T = 80$ and

Table I. — The relative intensities Z of second and fifth lines in ErFe_3

T (K)	Measured Z	Calculated $Z(\phi)$ for $q = 0.5$
4.2	3.0 ± 0.1	$Z(90^\circ) = 3.0$
80	1.8 ± 0.1	$\frac{1}{3}Z(0^\circ) + \frac{2}{3}Z(60^\circ) = 1.8$
295	1.8 ± 0.1	$\frac{1}{3}Z(0^\circ) + \frac{2}{3}Z(60^\circ) = 1.8$
295 ($H_{\text{ext}} = 1 \text{ kOe}$ $\parallel \gamma\text{-beam}$)	1.0 ± 0.1	$Z(0^\circ) = 1.0$

295 K. However, at $T = 4.2$ K we found $Z = 3.0$ from which it follows that all spins are turned over 90° to a plane perpendicular to the γ -ray beam. In summary we conclude from both the magnetization and ^{57}Fe Mössbauer effect measurements on ErFe_3 published earlier [1] and the results presented in this paper that the easy axis of magnetization turns from c -axis to b -axis at $T = 50$ K with increasing temperature. The results found by Bowden and Day [2] using ^{57}Fe Mössbauer effect and those by Davis *et al.* [3] derived from magnetic neutron diffraction measurements are not consistent with our conclusion.

(ii) In addition the ^{57}Fe Mössbauer effect was studied in the compounds $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ in order to investigate how the anisotropy is influenced by substituting iron by cobalt in ErFe_3 . Like in the case of ErFe_3 a spin reorientation from c -axis to b -axis is found as is shown for $\text{Er}(\text{Fe}_{0.90}\text{Co}_{0.10})_3$ in figure 1. In figure 2a the transition temperature (T_M) is plotted as a function of the Co concentration. Until 40 % Co, T_M is almost constant between $T = 40$ and 50 K. However with 45 % Co, T_M increases to $T = 158$ K, while the compound $\text{Er}(\text{Fe}_{0.35}\text{Co}_{0.65})_3$ is uniaxial over the entire temperature range below T_c . As the magnetic moment reorientation most likely is the result of a competition between the Er single ion crystal field anisotropy (c -axis) and the Fe-Co sublattice anisotropy [4], the size of the latter in the basal plane is almost constant up till 40 % Co, but decreases rapidly for higher Co concentrations. In figure 3 we give the Mössbauer spectra of $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ at $T = 4.2$ K with $x = 0.05, 0.25$ and 0.45 , respectively, each of which being decomposed into three different hyperfine field spectra. It appears that the Mössbauer

Fig. 1. — Mössbauer spectra of $\text{Er}(\text{Fe}_{0.9}\text{Co}_{0.1})_3$ at $T = 4.2$ K and 50 K from which the magnetic moment reorientation can be deduced.

Fig. 2. — a) The dependence of the spin reorientation temperature T_M in $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ compounds with composition. b) The hyperfine fields at three different iron sites in $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ at $T = 4.2$ K.

hyperfine lines of these spectra are slightly broadened with increasing Co concentration up to $x = 0.45$ similar as it has been found before in the $\text{R}(\text{Fe}_{1-x}\text{Co}_x)_2$ compounds [5]. In spite of this line broadening a detailed analysis could be made. The line width broadening observed in the compound with $x = 0.65$ is such that only an estimate can be made of the sublattice hyperfine fields. Apparently the influence of the local environment of an iron atom at this concentration has become large. The hyperfine fields at the three different iron sites are given in figure 2b. The observed changes in hyperfine fields at T_M in ErFe_3 [1] are also found in the $\text{Er}(\text{Fe}, \text{Co})_3$ compounds. Furthermore we note that iron substitution by cobalt occurs preferentially at the b and c sites as can be deduced from the relative peak intensities. No preference between b and c sites was found.

Fig. 3. — Decomposed Mössbauer spectra of $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_3$ at $T = 4.2$ K.

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