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# A study of the magnetocrystalline anisotropy of $RFe_{11-x}Co_xTi$ compounds with $R = Y$ and $Er$

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## Abstract

Structural and magnetic properties of the  $RFe_{11-x}Co_xTi$  compounds with  $R = Y$  and  $Er$  have been investigated. X-ray diffraction patterns and thermomagnetic curves show that all the synthesized compounds with  $x$  ranging from 0 to 11.0 are almost single phase and crystallize in the  $ThMn_{12}$ -type structure. Substitution of Co for Fe leads to a monotonic decrease of lattice constants and unit-cell volume and a clear increase of the Curie temperature. The saturation moments increase with increasing Co content, going through a maximum at around  $x = 2.5$ , and then decrease with further increasing Co content. The easy magnetization direction (EMD) at room temperature for  $ErFe_{11-x}Co_xTi$  compounds is along the  $c$ -axis for  $x \leq 4$ , perpendicular to the  $c$ -axis for  $6 \leq x \leq 9$  and then back to the  $c$ -axis for further increasing  $x$ . The anisotropy of the  $YFe_{11-x}Co_xTi$  compounds shows a similar behaviour. This concentration dependence of the magnetocrystalline anisotropy in the  $YFe_{11-x}Co_xTi$  and  $ErFe_{11-x}Co_xTi$  compounds results from the different contributions to the magnetocrystalline anisotropy from various transition-metal sites and the preferential occupation of Co atoms. A spin reorientation occurs below the Curie temperature for all  $ErFe_{11-x}Co_xTi$  compounds. Spin reorientation temperatures as a function of Co concentration were derived. A tentative spin phase diagram is given for  $ErFe_{11-x}Co_xTi$  compounds and can be understood in terms of crystal field theory.

## 1. Introduction

The rare earth (R)–transition metal (T) intermetallic compounds have been widely investigated since the Nd–Fe–B permanent magnet material was discovered [1–3]. Among the R–T compounds the study of structural and magnetic properties of  $RT_{12-x}M_x$  has been one of the major topics, where M represents a stabilizing element [4–9]. It is well accepted that in R–T compounds the easy magnetization direction (EMD) is determined by the competition between R-sublattice and T-sublattice anisotropy. In general, at low temperatures, sometimes even at room temperature the T-sublattice contribution to the total anisotropy is weaker than

the R-sublattice anisotropy and thus can be neglected. A recent study on the magnetic properties of  $R_2Fe_{17-x}Ga_x$  compounds shows that a substitution of Ga for Fe can lead to a room-temperature uniaxial anisotropy. This indicates that the T-sublattice contribution plays an important role in magnetocrystalline anisotropy at room temperature [10, 11]. On the  $RFe_{11-x}Co_xTi$  compounds a series of studies had been performed [12, 13], but the investigation of the magnetocrystalline anisotropy at low temperature has not been systematically reported. The singular point detection (SPD) [14] technique is well known as a quite effective and accurate method to measure the anisotropy field of compounds with easy-axis anisotropy. Recently, it was reported that this method can also be used to determine the anisotropy field of compounds with easy-plane anisotropy by using the rotation alignment technique [15].

In this paper, the influence of the substitution of Co for Fe in  $RFe_{11}Ti$  compounds with  $R = Y$  and Er on the structural and magnetic properties, especially low temperature anisotropy and exchange interaction, is presented.

## 2. Experimental process

The samples of  $RFe_{11-x}Co_xTi$  with  $R = Y$  and Er were prepared by arc-melting the constituent elements with a purity of at least 99.9% in argon atmosphere. In order to compensate for the loss of Y or Er during melting and annealing, an excess of 2–10 wt% of Y or Er was added. The ingots were remelted at least three times to ensure homogeneity. The alloys were annealed in high purity argon gas atmosphere at 1373 K for 24–48 hours for  $R = Y$  and at 1273 K for one week for  $R = Er$ , respectively, then quenched in water. The quality of the compounds was checked by both thermomagnetic analysis (TMA) and x-ray diffraction (XRD). The thermomagnetic curves were measured in a low field of 0.05 T using a vibrating sample magnetometer from 5 K up to temperatures above  $T_C$ . The Curie temperature was derived from  $M^2-T$  plots by extrapolating  $M^2$  to zero. The lattice parameters were determined from x-ray diffraction patterns with Cu  $K\alpha$  radiation of randomly oriented powder samples. XRD patterns of magnetically aligned powder samples were used to determine the easy magnetization direction. To prepare magnetically aligned samples of the compounds with easy-axis anisotropy, a mixture of fine powders (the average size of the powdered particles is less than 40  $\mu m$ ) of the compound and epoxy resin was put into a Teflon tube of cylindrical shape. Then the tube was put in a magnetic field of about 1 T with the axis of the tube parallel or perpendicular to the magnetic field direction until the epoxy resin hardened. In this way the EMD of the sample is either parallel or perpendicular to the axis of the tube. For the compounds with easy-plane anisotropy at room temperature, the fine powders were mixed with epoxy resin and embedded in a Teflon tube of cylindrical shape. The tube was rotated with the cylindrical axis perpendicular to a magnetic field of about 1 T until the epoxy resin solidified. In this case the cylinder axis is the hard magnetization direction (HMD) of the easy-plane compound [15].

## 3. Results and discussion

From thermomagnetic curves and XRD patterns obtained it can be seen that all samples investigated are almost single phase except for a small amount of  $\alpha$ -Fe as second phase in some samples. All samples crystallize in the  $ThMn_{12}$ -type structure. The lattice parameters were derived from the XRD patterns of powder samples and are listed in tables 1 and 2 for  $YFe_{11-x}Co_xTi$  compounds and  $ErFe_{11-x}Co_xTi$ , respectively. It can be seen that substitution of Co for Fe leads to a decrease of lattice constants due to the smaller radius of Co compared with Fe.

**Table 1.** Structural and magnetic properties of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds. Error:  $a$  and  $c \pm 0.0005 \text{ \AA}$ ,  $T_C \pm 5 \text{ K}$ ,  $M_s$  at  $5 \text{ K} \pm 0.1 \mu_B$ ,  $B_a \pm 0.1 \text{ T}$ .

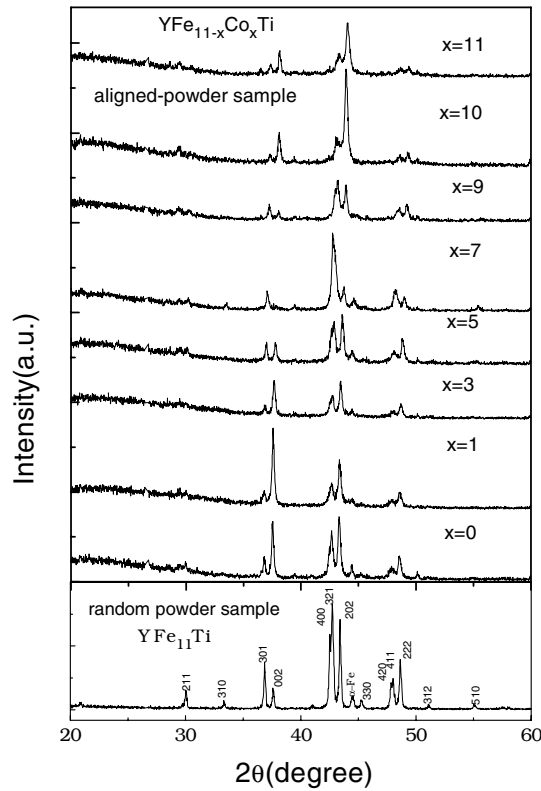
$x$	$a$ ( $\text{\AA}$ )	$c$ ( $\text{\AA}$ )	$V$ ( $\text{\AA}^3$ )	$T_C$ (K)	$M_s$ ( $\mu_B \text{ fu}^{-1}$ )	$B_a$ (5 K) (T)	$B_a$ (RT) (T)
0	8.4986	4.7755	344.9	528	20.6	4.1	2.2
1	8.5125	4.7840	346.7	638	21.2	3.7	2.3
3	8.5069	4.7762	345.6	796	21.9	2.9	2.0
5	8.4683	4.7567	341.1	908	21.0	0.8	0.6
7	8.4326	4.7363	336.3	981	19.5	-1.4	-0.8
9	8.4151	4.7300	335.0	1018	18.1	0.8	0.3
11	8.3694	4.7158	330.3	1020	15.7	2.8	1.7

**Table 2.** Structural and magnetic properties of  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  compounds. Error:  $a$  and  $c \pm 0.0005 \text{ \AA}$ ,  $T_C \pm 5 \text{ K}$ ,  $M_s \pm 0.1 \mu_B$ .

$x$	$a$ ( $\text{\AA}$ )	$c$ ( $\text{\AA}$ )	$V$ ( $\text{\AA}^3$ )	$T_C$ (K)	$T_{sr}$ (K)	$M_s$ (5 K) ( $\mu_B \text{ fu}^{-1}$ )	EMD room temperature
0	4.770	8.460	341.4	98	39	10.5	easy-axis
1	4.757	8.454	339.9	611	41	11.8	easy-axis
2	4.741	8.436	337.4	733	38	13.0	easy-axis
3	4.742	8.432	337.1	828	27	13.2	easy-axis
4	4.735	8.415	335.3	918	35	12.1	easy-axis
6	4.734	8.412	335.0	940	489	8.5	easy-plane
7	4.731	8.391	333.1	980	604	7.8	easy-plane
9	4.721	8.397	332.9	1001	627	6.9	easy-plane
11	4.708	8.368	329.7	1008	133	6.0	easy-axis

As an example, XRD patterns of the magnetically aligned powder samples of plate-shape of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds with the plane of sample perpendicular to the direction of the external field are shown in figure 1. In order to give a good comparison, the XRD pattern of the unaligned sample of  $\text{YFe}_{11}\text{Ti}$  is also drawn in the same figure. It can be seen that, compared with the XRD pattern of the randomly oriented  $\text{YFe}_{11}\text{Ti}$  powder sample, the peak of (002) for the  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds with  $x \leq 3$  and  $x \geq 10$  is clearly enhanced and the ( $hk0$ ) peaks are weakened. This indicates that the EMD of these compounds is along the  $c$ -axis. For  $x = 9$  and 5 the intensity of (002) is also enhanced but not clearly. When  $x = 7$ , the peak of (002) is weakened and peaks of ( $hk0$ ), such as (400) and (420), are enhanced, which means that the EMD has changed from the  $c$ -axis to the  $c$ -plane. To confirm this, we measured the XRD pattern of the rotation-aligned powder sample with the plane of the plate-shape sample parallel to the direction of the external magnetic field. The XRD patterns of 'normal' aligned (a) and rotation aligned (b) samples are shown in figure 2 together with that of  $\text{YFe}_{11}\text{Ti}$  compound for comparison. It can be seen that the (002) peak of the rotation-aligned sample is enhanced after magnetic alignment. This proved further that the EMD of the  $\text{YFe}_4\text{Co}_7\text{Ti}$  compound is the  $c$ -plane. For  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  compounds the case is similar. The EMD at room temperature for  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  compounds is along the  $c$ -axis for  $x \leq 4$ , perpendicular to the  $c$ -axis for  $6 \leq x \leq 9$  and then reverts to the  $c$ -axis with increasing  $x$ . The composition dependence of the EMD at room temperature is in good agreement with the results reported by Sinha *et al* [12].

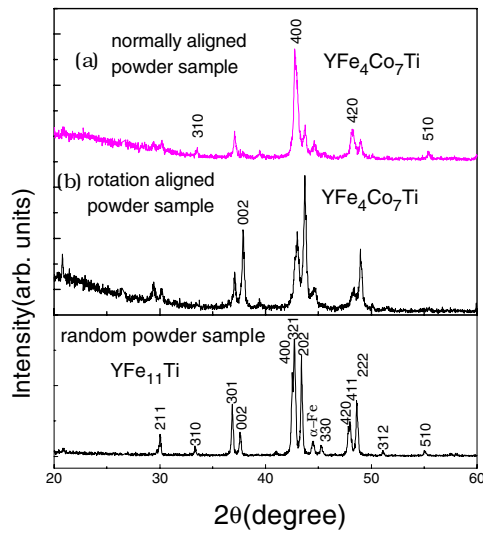
The values of  $T_C$  of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  and  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  compounds are listed in tables 1 and 2, respectively. It can be seen that  $T_C$  increases monotonically with increasing  $x$ .



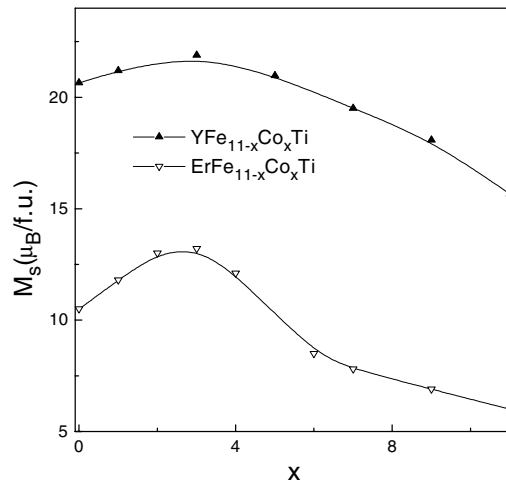
**Figure 1.** XRD patterns of normally aligned powder samples for  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  compounds.

This behaviour is different from the case of the  $\text{YFe}_{10-x}\text{Co}_x\text{Si}_2$  system [16], where a maximum occurs at about  $x = 6$ . It is well known that in R–T compounds  $T_C$  is mainly determined by the T–T exchange interaction and that this is sensitive to the distance between the T atoms. In the  $\text{ThMn}_{12}$ -type structure there exist three non-equivalent transition-metal sites 8i, 8j and 8f. The T–T nearest-neighbour distances are different for the three different T sites. It has been reported that in the 1:12 compounds Si and Ti have different preferential occupations [17]. The different composition dependence of  $T_C$  between  $\text{Y}(\text{Fe}, \text{Co})_{11}\text{Ti}$  and  $\text{Y}(\text{Fe}, \text{Co})_{10}\text{Si}_2$  systems may be associated with the atom preferential occupations.

The composition dependences of the saturation moments  $M_s$  at 5 K of  $\text{RFe}_{11-x}\text{Co}_x\text{Ti}$  compounds with  $\text{R} = \text{Y}$  and Er are listed in tables 1 and 2, respectively, and shown in figure 3. It can be seen that a maximum is observed at about  $x = 2.5$ . Compared with the Pauling–Slater curve of the binary Fe–Co alloy, the position of the maximum shifts towards a smaller Co content. This may be due to the contribution of the Er or Y ions, as seen in the  $\text{Y}(\text{Fe}_{1-x}\text{Co}_x)_{11}\text{Ti}$  compounds [13]. The appearance of a maximum in the saturation magnetization in  $\text{R}(\text{Fe}_{1-x}\text{Co}_x)_{11}\text{Ti}$  ( $\text{R} = \text{Y}$  and Er) at an intermediate Co content can be understood in terms of a rigid band model. In the Fe–Co binary system (Slater–Pauling curves), with increasing substitution of Co for Fe the spin-up d band is filled at first. Hence the difference between the spin-up and spin-down states increases, and this results in an increase of the saturation magnetization. As the Co content increases the spin-up band is completely filled and this results in strong ferromagnetism and subsequently the spin-down band is filled.



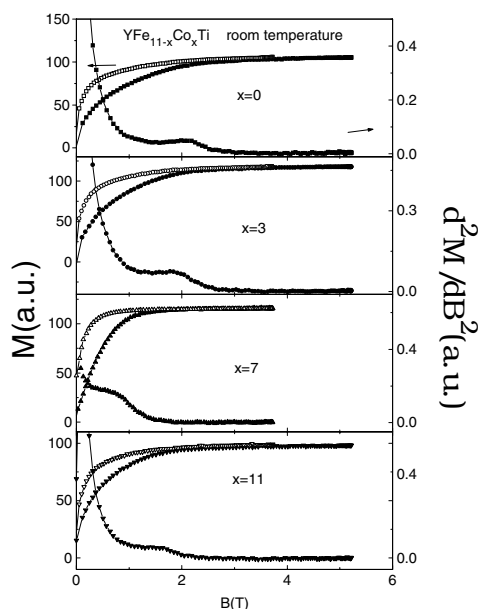
**Figure 2.** XRD patterns of normally aligned (a) and rotation aligned (b) samples.



**Figure 3.** Composition dependence of the saturation magnetization  $M_s$  at 5 K for  $R\text{Fe}_{11-x}\text{Co}_x\text{Ti}$  compounds with  $R = \text{Y}$  and  $\text{Er}$ .

Thereby the difference between the spin-up and spin-down states decreases, which results in a decrease of the saturation magnetization.

The anisotropy fields at room temperature and 5 K of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  were obtained by the SPD method and by linearly extrapolating the  $\Delta M (= M_{\parallel} - M_{\perp}) - B$  curve to  $\Delta M = 0$  respectively. For example, in figure 4 the SPD and magnetization curves at room temperature of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  with  $x = 0, 3, 7$  and  $11$  are displayed. The peaks in the SPD patterns correspond to the anisotropy field. A singularity, indicating an anisotropy field  $B_a$ , is clearly detectable in the  $d^2M/dB^2 - B$  curves. The composition dependences of  $B_a$  at room temperature and at 5 K are listed in table 1 for  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$ . It can be seen that the composition dependence of the anisotropy field has a minimum at  $x = 7$ , which is similar to the case of  $\text{Pr}_2(\text{Fe},\text{Co})_{14}\text{B}$  [2]. The value of  $B_a$  of the compound  $\text{YFe}_4\text{Co}_7\text{Ti}$  is negative because its anisotropy is planar.



**Figure 4.** The SPD patterns and magnetization curves along the EMD (open symbols) and HMD (solid symbols) of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  with  $x = 0, 3, 7$  and  $11$ .

Because Y is nonmagnetic and thus has no contribution to the magnetocrystalline anisotropy, in  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  the anisotropy is determined by the interplay of the Fe- and Co-sublattice contributions. Based on the anisotropy of  $\text{YFe}_{11}\text{Ti}$  and  $\text{YCo}_{11}\text{Ti}$ , it can be concluded that the anisotropies of the Fe and Co sublattices are uniaxial from low temperatures up to room temperature. In the  $\text{ThMn}_{12}$ -type structure there are three non-equivalent transition-metal sites 8i, 8j and 8f. Thus it is reasonable to assume that in the 1:12-type compounds the transition-metal atoms at the different sites give different contributions to the anisotropy. In fact, in the  $\text{RFe}_{11}\text{Ti}$  compounds the uniaxial anisotropy is mainly due to the contribution of Fe at the 8i site [18]. Moreover, it has been found [19] that a spin reorientation occurs in  $\text{YCo}_{11}\text{Ti}$ , which suggests that different Co sites may have different contributions to the anisotropy just as in  $\text{R}_2\text{Co}_{17}$  compounds [20]. Therefore, the unusual concentration dependence of the anisotropy in  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$  may be associated with the preferential substitution of Co for Fe [21].

The temperature dependence of the magnetization of the  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  compounds at a low field of  $B = 0.05$  T from 5 K up to the Curie temperature is shown in figure 5. For the samples with  $x < 6$  anomalies occur below room temperature, which corresponds to a spin reorientation while for  $x = 6, 7$  and  $9$  the corresponding anomalies occur above room temperature. Moreover, when  $x$  is equal to 4, a second anomaly emerges at about 270 K (as shown in figure 5). In order to examine what happens at these critical temperatures, the dependence of the magnetization  $M$  on the angle  $\theta$  between the alignment direction and the external field direction at various temperatures was measured. It is clear that at a given temperature, if the anisotropy of the compound is of the easy-axis or the easy-plane type, there is only one maximum in the  $M-\theta$  curve between  $0^\circ$  and  $180^\circ$ . There are two maxima for a compound with an easy-cone type anisotropy. If the EMD changes in the temperature range investigated, the position of the maximum in the  $M-\theta$  curves will change. As an example, we show in figures 6(a) and (b) the angular dependence of the magnetization measured in a field of 0.1 T at various temperatures for  $\text{ErCo}_{11}\text{Ti}$  and  $\text{ErFe}_7\text{Co}_4\text{Ti}$ , respectively. It can

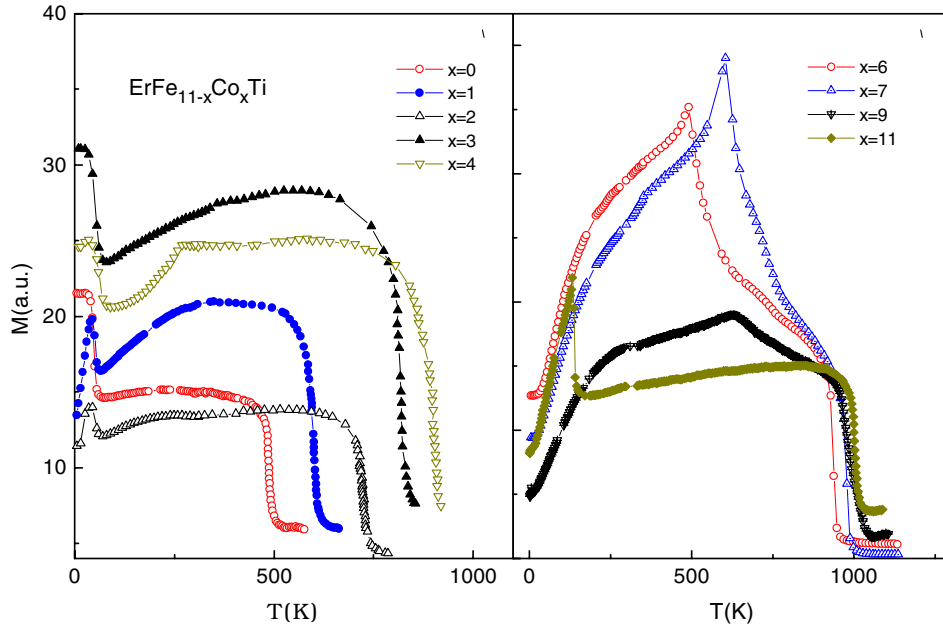


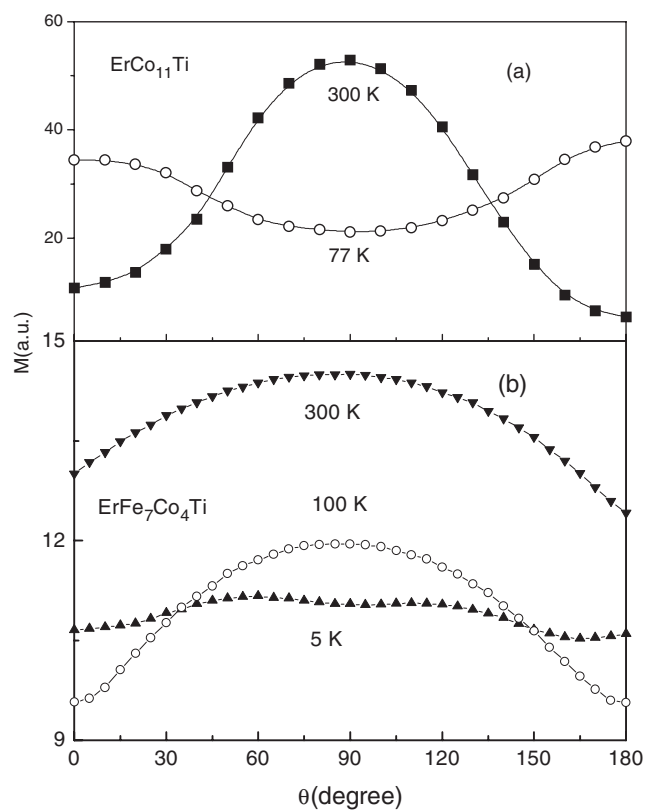
Figure 5. Temperature dependence of magnetization at a low field for  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$ .

be seen that for  $\text{ErCo}_{11}\text{Ti}$ , with increasing temperature from low temperature to 300 K, the number of maxima does not change but the position of the maximum shifts from  $0^\circ$  (or  $180^\circ$ ) at 77 K to  $90^\circ$  at 300 K, which means that the anisotropy of  $\text{ErCo}_{11}\text{Ti}$  changes from easy plane at 77 K to easy axis at 300 K. For  $\text{ErFe}_7\text{Co}_4\text{Ti}$ , however, with increasing temperature from 5 K to 300 K, the number of maxima changes from two at 5 K to one at 100 K and 300 K. Moreover, the position of the maximum does not shift when the temperature increases from 100 K to 300 K. It can be concluded that for  $\text{ErFe}_7\text{Co}_4\text{Ti}$  the first anomaly (at 35 K) in the  $M-T$  curves corresponds to a spin reorientation from easy-cone anisotropy at 5 K to easy-axis anisotropy at high temperature while the second anomaly at about 270 K does not represent a spin reorientation and is maybe associated with domain wall movement. The spin phase diagram for the system  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  is shown in figure 7. It is well accepted that in R-T compounds the anisotropy is determined by the competition between the rare-earth sublattice and the transition-metal sublattice. In these compounds the total anisotropy is the sum of the T-sublattice and the Er-sublattice anisotropy. Because both  $\text{YFe}_{11}\text{Ti}$  and  $\text{YCo}_{11}\text{Ti}$  exhibit easy-axis anisotropy at room temperature, so the T-sublattice anisotropy constant  $K_1^T$  is positive. From the spin reorientation of  $\text{RCo}_{11}\text{Ti}$  Buschow *et al* [9] have deduced that  $A_{20}$  is positive in the Co-based compounds whereas  $A_{20}$  is negative in the  $\text{RFe}_{11}\text{Ti}$  compounds [5]. It is well accepted that if one only takes a first-order approximation, the rare-earth-sublattice anisotropy constant  $K_1^{\text{Er}}$  is determined by the product of the second-order crystal-field parameter  $A_{20}$  and the second-order Stevens coefficient  $\alpha_J$ . Specifically

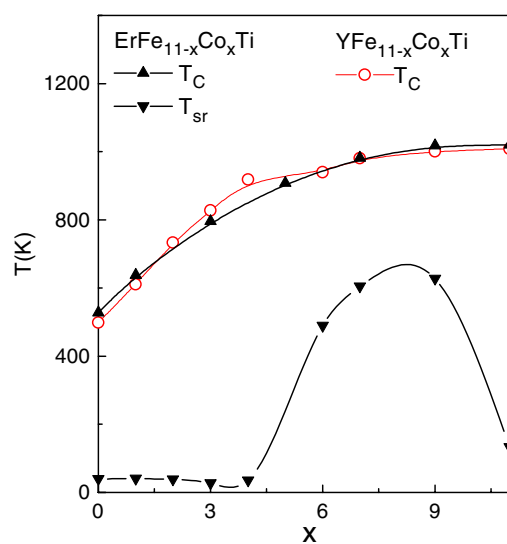
$$K_1^{\text{Er}} = -\frac{3}{2}\alpha_J\langle r^2 \rangle \langle 3J^2 - J(J+1) \rangle A_{20}. \quad (1)$$

Here the quantities in angular brackets represent the expectation values. Since  $\alpha_J$  is positive for Er,  $K_1^{\text{Er}}$  in  $\text{ErFe}_{11}\text{Ti}$  is positive, which indicates that the Er sublattice should favour easy-axis anisotropy. Hu *et al* [5] reported that a spin reorientation in  $\text{ErFe}_{11}\text{Ti}$  may result from the higher-order term  $B_{60}\langle O_{60} \rangle$ , since Er has a large and positive  $\gamma_J$ . It is known that the sign





**Figure 6.** Dependence of the magnetization of  $\text{ErCo}_{11}\text{Ti}$  (a) and  $\text{ErFe}_7\text{Co}_4\text{Ti}$  (b) on the angle between the alignment direction and the direction of the external field of 0.1 T.



**Figure 7.** Composition dependence of spin reorientation temperature  $T_{sr}$ , Curie temperature  $T_C$  of  $\text{ErFe}_{11-x}\text{Co}_x\text{Ti}$  and  $T_C$  of  $\text{YFe}_{11-x}\text{Co}_x\text{Ti}$ .

reversal of the second-order crystal field parameter occurs when going from R–Fe compounds to R–Co compounds [9]. In ErCo<sub>11</sub>Ti the negative  $K_1^{Er}$  of the Er sublattice is expected to be responsible for an easy-plane magnetization. Thus, the spin reorientation in ErCo<sub>11</sub>Ti can be understood in terms of a competition between the opposite contributions from the Er sublattice and the Co sublattice.

When Co replaces Fe,  $A_{20}$  increases with increasing Co content, which results in a decrease of  $K_1^{Er}$ . Combined with the composition dependence of the anisotropy in YFe<sub>11-x</sub>Co<sub>x</sub>Ti the composition dependence of the spin reorientation temperature for ErFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds (as shown in figure 7) can be understood.

#### 4. Conclusion

The substitution of Co for Fe in YFe<sub>11</sub>Ti and ErFe<sub>11</sub>Ti compounds does not change the crystal structure but results in a decrease of the unit cell volume. A clear increase of the Curie temperature for both systems, YFe<sub>11-x</sub>Co<sub>x</sub>Ti and ErFe<sub>11-x</sub>Co<sub>x</sub>Ti, was observed by substituting Fe by Co. The composition dependence of the saturation moments exhibits a maximum at about  $x = 0.25$ . It was also found that the composition dependence of the anisotropy field of YFe<sub>11-x</sub>Co<sub>x</sub>Ti has a minimum at  $x = 7$ . For all ErFe<sub>11-x</sub>Co<sub>x</sub>Ti compounds, a spin reorientation occurs below the Curie temperature. A tentative spin phase diagram has been constructed for the system ErFe<sub>11-x</sub>Co<sub>x</sub>Ti.

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