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## Magnetocrystalline anisotropy in $R(\text{Fe},\text{Co})_{11.3}\text{Nb}_{0.7}$ compounds with $R = \text{Dy}$ and $\text{Er}$

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**Abstract.** Magnetic properties, specially magnetocrystalline anisotropy, of the  $\text{Re}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds ( $R = \text{Dy}$  and  $\text{Er}$ ) have been investigated by x-ray diffraction (XRD) and magnetic measurements. All the compounds studied crystallize in the  $\text{ThMn}_{12}$ -type structure for  $x \leq 0.3$ . Substitution of Co for Fe leads to a monotonic increase of Curie temperature and an obvious decrease of  $a$  and  $c$  lattice parameters in both  $\text{Dy}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  compounds. The composition dependence of the saturation magnetization  $M_S$  at 5 K shows a maximum at  $x = 0.2$  and  $x = 0.15$  in  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds, respectively. The easy magnetization direction (EMD) at room temperature has been determined by XRD of magnetically aligned samples. Both ac susceptibility and magnetization measurements indicate that as the temperature decreases from 300 K to 5 K, the EMD of all the  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  compounds changes from  $c$ -axis to a cone whereas the EMD of the  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds goes from  $c$ -axis, passing through a cone, to a plane for  $x \leq 0.2$  and from  $c$ -axis to a plane for  $x \geq 0.25$ . A tentative spin phase diagram is constructed for  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with  $x \leq 0.3$ , respectively.

### 1. Introduction

The discovery of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  [1] not only led to the appearance of a new type of excellent permanent magnet, but also indicated that the rare-earth (R)–transition-metal (T) intermetallic compounds are very good objects to be used for investigation of the 3d–4f electron interaction. Since Coey and Sun [2] and Zhong *et al* [3] found that introduction of nitrogen and carbon led to a clear improvement of permanent-magnetic properties of the  $\text{Sm}_2\text{Fe}_{17}$  compound, much attention has been focused on the investigation of new nitrides and carbides of the R–T compounds. Quite recently, it has been found [4] that the  $\text{Sm}_3(\text{Fe}, \text{Ti})_{29}\text{N}_y$  compound is a novel candidate for permanent-magnet materials. On the other hand, some interest has been focused on the intermetallic compounds with  $\text{ThMn}_{12}$ -type structure for their potential application as permanent magnets, as well as for their variety of magnetic structures and magnetic phase transitions. The  $\text{ThMn}_{12}$ -type structure is a body centre tetragonal, with space group  $I4/mmm$ . In the  $\text{ThMn}_{12}$  structure there are one R site (2a), and three T sites (8i, 8j and 8f) in a unit cell. This structure can be derived from the basic  $\text{CaCu}_5$ -type structure by replacing some of the R atoms by dumb-bell pairs of transition-metal atoms. Although  $\text{RFe}_{12}$  compounds do not exist for any rare earth, the  $\text{ThMn}_{12}$  structure can be stabilized in the quasibinary

$RFe_{12-x}M_x$  compounds with  $M$  as a stabilizing element ( $M = Al, Ti, V, Cr, Mo, Si, Nb, W, Ga$  and  $Ta$ ). The  $R$  and  $Fe$  elements can be replaced by other rare-earth and transition elements partially or totally, respectively, while the  $M$  element can also be changed, which provides a comparable and abundant model to study the physical properties of  $R-T$  compounds and hence becomes an effective approach to improve magnetic performance of the materials. Recently, Hu *et al* [5] reported that a single phase 1:12 type of  $R-T$  compounds can be synthesized with a small amount of  $Nb$  as stabilizing element. In general, introduction of a stabilizing element leads to a decrease of saturation magnetization; therefore, a reduction of the amount of stabilizing element is favourable for permanent-magnet application. Earlier, Wang *et al* [6] reported the effect of the substitution of  $Ni$  for  $Fe$  on structure and magnetic properties of  $RFe_{11.3}Nb_{0.7}$  ( $R = Dy, Er$ ). Due to the different sign of the second order Steven factor  $\alpha_J$  of  $Dy$  and  $Er$ , the  $DyFe_{11.3}Nb_{0.7}$  and  $ErFe_{11.3}Nb_{0.7}$  compounds show the different magnetic properties, specially magnetocrystalline anisotropy. It is expected that the  $Co$  substitution for  $Fe$  will show the different influence on the magnetic properties of both  $Dy(Fe_{1-x}Co_x)_{11.3}Nb_{0.7}$  and  $Er(Fe_{1-x}Co_x)_{11.3}Nb_{0.7}$  compounds. In the present paper, a study of the influence of the substitution of  $Co$  for  $Fe$  on the magnetic properties, specially on the magnetocrystalline anisotropy of  $RFe_{11.3}Nb_{0.7}$  compounds ( $R = Dy$  and  $Er$ ), is presented.

## 2. Experiment

$Re(Fe_{1-x}Co_x)_{11.3}Nb_{0.7}$  ( $R = Dy, Er$ ) compounds were prepared by arc melting the constituent elements of 99.9% purity under an argon atmosphere with high purity. An additional amount of  $R$  was added to compensate the loss during the melting process. Thermomagnetic analysis (TMA) and XRD patterns with  $Cu K\alpha$  radiation were employed for identification of the single phase quality and the lattice parameters. XRD patterns of a magnetically aligned sample were used to determine the EMD in these compounds at room temperature, the sample were prepared by fixing the fine powder particles (diameter less than  $40 \mu m$ ) with epoxy resin at room temperature in a field of 1 T. The temperature dependence of the magnetization ( $M$ ) was measured in a low field of 0.05 T from room temperature to 1000 K in a vibrating sample magnetometer (VSM) and from 5 K to 300 K in a SQUID magnetometer, respectively. The Curie temperatures  $T_C$  were obtained from  $M^2-T$  plots by extrapolating  $M^2$  to zero. In order to confirm the transition of the magnetic phase the magnetization curves of aligned samples were measured in a field parallel and perpendicular to the aligned direction at different temperatures by a SQUID system when temperature below 300 K and by a pulsed magnetic field system (PMF) otherwise. The spin-reorientation temperatures ( $T_{sr}$ ) were obtained from the peak position in the temperature dependence of ac susceptibility. The singular point detection (SPD) technique [7] was used to determine the anisotropy field  $B_a$  at room temperature. To determine accurately the data of the saturation magnetic moment  $M_S$ , isothermal magnetization curves of fine powder were obtained by the SQUID system. The saturation magnetization  $M_S$  was determined from  $M-1/B$  curves of the powder by extrapolating  $1/B$  to zero using the saturated part.

## 3. Results and discussion

XRD patterns show that all the  $Re(Fe_{1-x}Co_x)_{11.3}Nb_{0.7}$  ( $R = Dy, Er$ ) compounds investigated with  $x \leq 0.3$  crystallize in the  $ThMn_{12}$  type of structure. For  $x > 0.3$ , the XRD patterns show existence of the impurity phase, i.e.  $\alpha-Fe$  and  $NbFe_2$ . The lattice parameters of  $R(Fe, Co)_{11.3}Nb_{0.7}$  ( $R = Dy, Er$ ) have been derived from XRD patterns of powder samples

**Table 1.** Structural and magnetic parameters of  $R(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  compounds with  $R = \text{Dy}, \text{Er}$ .

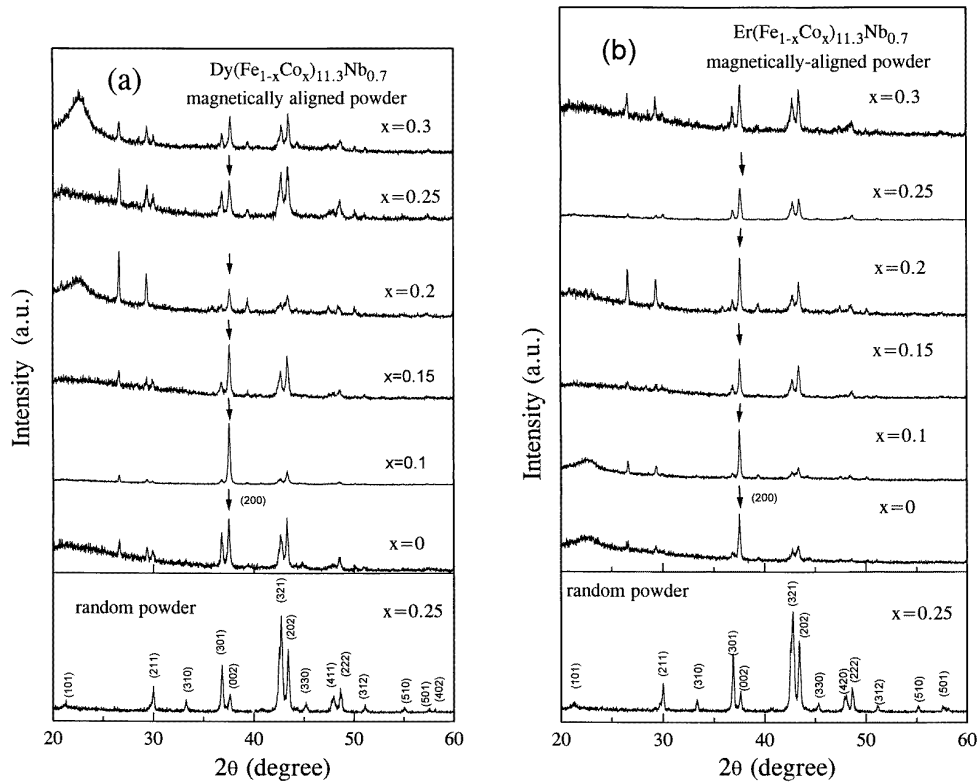
	$x$	$a$ (nm)	$c$ (nm)	$V$ (nm <sup>3</sup> )	$T_C$ (K)	$M_S$ (5 K) (10 <sup>3</sup> A m <sup>-1</sup> )	$B_a$ (300 K) (T)	$T_{sr1}$ (K)	$T_{sr2}$ (K)	EMD (RT)
R = Dy	0	0.8514	0.4795	0.3476	524	615	1.31	228	120	uniaxial
	0.10	0.8518	0.4788	0.3474	654	681	1.80	208	101	uniaxial
	0.15	0.8509	0.4785	0.3465	700	695	1.93	210	79	uniaxial
	0.20	0.8508	0.4780	0.3461	764	710	1.98	217	43	uniaxial
	0.25	0.8502	0.4779	0.3455	781	682	1.89	221	—	uniaxial
	0.30	0.8496	0.4769	0.3442	835	650	1.81	220	—	uniaxial
R = Er	0	0.8498	0.4793	0.3462	599	635	2.64	45	—	uniaxial
	0.10	0.8501	0.4778	0.3453	630	689	2.69	48	—	uniaxial
	0.15	0.8505	0.4793	0.3468	672	699	2.22	46	—	uniaxial
	0.20	0.8500	0.4786	0.3458	712	700	2.06	50	—	uniaxial
	0.25	0.8489	0.4780	0.3445	754	668	1.88	52	—	uniaxial
	0.30	0.8480	0.4773	0.3432	805	659	1.23	60	—	uniaxial

and are listed in table 1. It can be seen that the lattice constants  $a$  and  $c$  as well as unit-cell volume  $V$  decrease with increasing Co content, which is due to the small radius of the Co atom compared with Fe.

XRD patterns at room temperature of a random powder sample of the  $R(\text{Fe}_{0.75}\text{Co}_{0.25})_{11.3}\text{Nb}_{0.7}$  and magnetically aligned samples of  $R(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  ( $R = \text{Dy}$  and  $\text{Er}$ ) are shown in figures 1(a) and 1(b), respectively. It can be seen that in all of the magnetically aligned  $R(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  ( $R = \text{Dy}$  and  $\text{Er}$ ) compounds the (002) reflection is greatly enhanced due to the alignment compared with the other peaks, which indicates that the EMD of all compounds investigated is along the  $c$ -axis at room temperature.

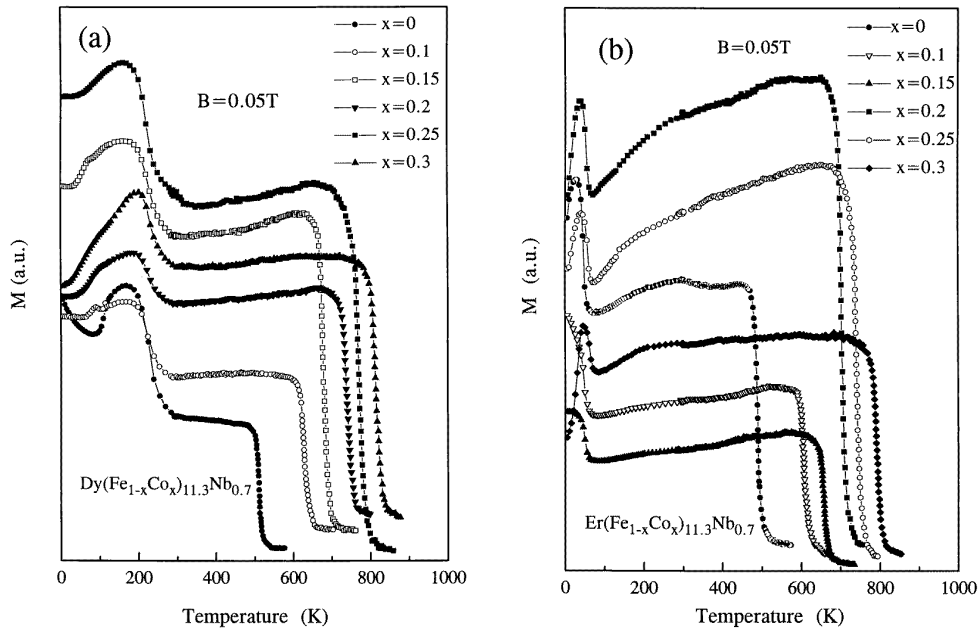
The values of saturation magnetization of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  ( $x \leq 0.3$ ) at  $T = 5$  K are listed in table 1. It can be seen that the concentration dependence of the saturation magnetization of both  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  has a maximum at  $x = 0.2$  and  $x = 0.15$ , respectively. Assuming that the  $R$  ( $\text{Er}$  and  $\text{Dy}$ ) magnetic moments are independent of the substitution of Co for Fe, the appearance of the maximum of the saturation magnetization in  $R(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  ( $R = \text{Dy}$  and  $\text{Er}$ ) compounds with increasing Co content can be understood in terms of the rigid band model. According to this model, in the Fe–Co binary system (Slater–Puling curves), with increasing substitution of Co for Fe the spin-up d band is filled further at first and hence the difference between the spin-up and spin-down states increases, which results in an increase of the saturation magnetization. As the Co content increases further the spin-up band is completely filled which results in strongest ferromagnetism and subsequently the spin-down band is filled further, thereby the difference between the spin-up and spin-down states decreases, which results in a decrease of the saturation magnetization.

The thermomagnetic curves of the  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds are shown in figure 2(a) and 2(b), respectively. The Curie temperatures  $T_C$  derived from the thermomagnetic curves are listed in table 1. From figure 2(a), 2(b) and table 1, it can be seen that in both  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds,  $T_C$  increases monotonically with increasing Co content. Further, the increase is faster than that expected from a dilution model. It is well known that there are three types of exchange interaction in  $R\text{Fe}_{12-x}\text{M}_x$  compounds, namely, the R–R interaction between the R atom magnetic moments, the T–T interaction between the T atomic magnetic moments and R–T interaction between the R and T atomic magnetic moments. Among them the T–T interaction is the strongest and dominates the magnetic ordering temperature. According to Wang *et al*



**Figure 1.** X-ray diffraction patterns at room temperature of random powder sample of  $R\text{Fe}_{8.475}\text{Co}_{2.825}\text{Nb}_{0.7}$  and magnetically aligned samples of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with (a)  $R = \text{Dy}$  and (b)  $R = \text{Er}$ .

[8], in  $\text{Y}(\text{Fe}, \text{Co})_{10}\text{Si}_2$ , the exchange interaction between the Co–Co pairs and Co–Fe pairs is larger than that of previous Fe–Fe pairs. This may be also true in the  $\text{Dy}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  and  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  compounds. Therefore it is expected that the substitution of Co for Fe would lead to an increase of  $T_C$ . The faster increase of  $T_C$  can be interpreted in terms of preferential occupation of Co atoms. It has been reported [9] that in the  $\text{R}_2\text{Fe}_{14-x}\text{Co}_x\text{B}$  compounds the Co atoms preferentially occupy the  $16k_2$  sites and contribute a positive exchange interaction whereas the Fe atoms in  $16k_2$  contribute a negative exchange interaction. Thus the substitution of Co for Fe leads to a decrease of the negative exchange interaction and therefore the total exchange interaction increases. In the  $\text{RFe}_{12-x}\text{M}_x$  compounds Fe atoms occupy three non-equivalent T sites and the dominant Fe–Fe exchange interaction is very sensitive to the distance between the Fe atoms. The average distance of Fe–Fe pairs at different sites shows the following relationship:  $d\text{Fe}(8i) > d\text{Fe}(8j) > d\text{Fe}(8f)$ . It is well known [10] that in Fe-based  $\text{ThMn}_{12}$ -type compounds, when the distance of Fe–Fe pairs is smaller than a certain critical value, the Fe–Fe interaction is negative. Therefore maybe the Fe–Fe interaction in 8f or 8j sites is negative. Neutron-diffraction studies of the  $\text{YFe}_{10-x}\text{Co}_x\text{Mo}_2$  compounds [11] and  $\text{Y}(\text{Co}_{0.5}\text{Fe}_{0.5})_{11}\text{Ti}$  compound [12] show that the Co atoms prefer to occupy the 8f and 8j sites. This may be true in the  $\text{RFe}_{11.3-c}\text{Co}_c\text{Nb}_{0.7}$  compounds. So the substitution of Co for Fe leads to a decrease of the negative exchange interaction and a faster increase of  $T_C$  with increasing Co content.

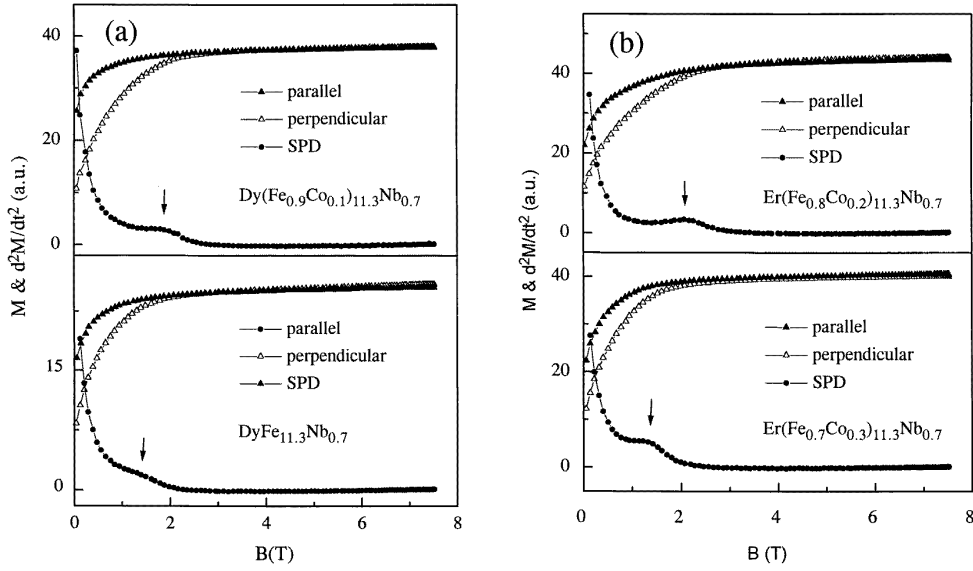


**Figure 2.** Temperature dependence of the magnetization of bulk sample in a field of 0.05 T of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with (a)  $R = \text{Dy}$  and (b)  $R = \text{Er}$ .

Figures 3(a) and (b) show magnetization curves of several  $\text{Re}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  ( $R = \text{Dy}, \text{Er}$ ) compounds together with their SPD signals as a function of magnetic field at room temperature. The values of magnetocrystalline anisotropy field  $B_a$  were obtained from the corresponding peak positions in the SPD curves and are listed in table 1.

It can be seen from figure 2(a) that the temperature dependence of the magnetization  $M$  of bulk samples of the  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds measured in a field of 0.05 T shows two anomalies for the compounds with  $x \leq 0.2$ , while only one appears in the compounds with  $x = 0.25$  and 0.3 above 5 K. In order to confirm these anomalies, the temperature dependence of the ac susceptibility  $\chi$  of  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  was measured and some of the experimental results are shown in figure 4(a). We find the temperature of the anomalies in  $\chi-T$  curves are in good agreement with that in the  $dM/dT-T$  curves below Curie temperatures. According to the measurement of angular dependence of magnetization of magnetically aligned samples of the  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  compound [13], it can be concluded that with decreasing temperature the EMD of  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  changes from uniaxial at room temperature, going through a cone type, then to planar at low temperature. Magnetization curves of the magnetically aligned  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  compounds measured at different temperatures shown in figure 5(a) have supported the conclusion mentioned above. It can be seen from figure 5(a) that the magnetization curve measured at room temperature in a field parallel to the alignment direction is higher than that in the perpendicular direction. At 125 K two magnetization curves almost coincide, which shows that a cone-type spin phase exists. At 5 K the magnetization curve measured in a field parallel to the alignment direction becomes lower than that with field perpendicular, which shows that a planar spin phase exists.

Therefore all the anomalies appearing in  $M-T$  and  $\chi-T$  curves below  $T_C$  are associated with the spin reorientation. The spin-reorientation phase diagrams of  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$



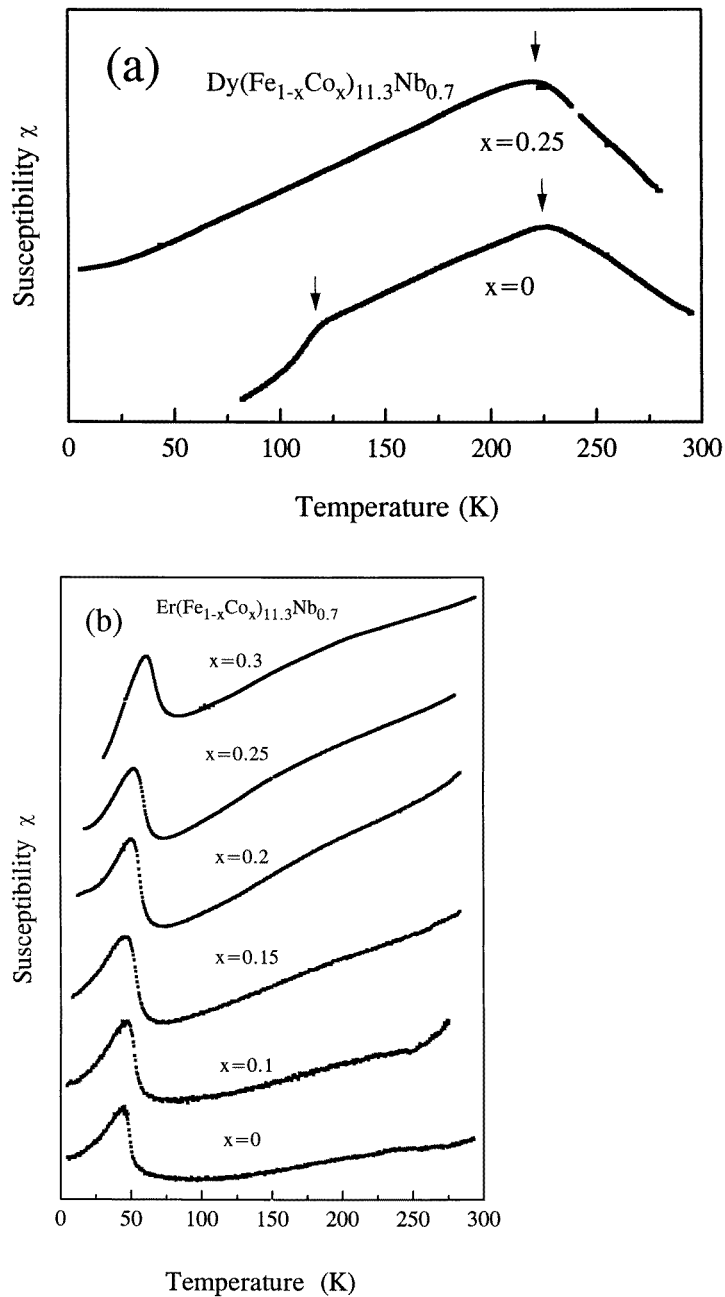
**Figure 3.** SPD signals and magnetization curves at room temperature of magnetically aligned samples of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with the field parallel and perpendicular to the  $c$ -axis for (a)  $R = \text{Dy}$ ,  $x = 0, 0.1$  and (b)  $R = \text{Er}$ ,  $x = 0.2, 0.3$ .

compounds with  $x \leq 0.2$  are regarded as analogues. However, for the  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with  $x = 0.25$  and  $0.3$ , only one anomaly appeared in  $M$ - $T$  and  $\chi$ - $T$  curves above 5 K. The type of anisotropy below the spin-reorientation temperature  $T_{sr}$  would be one of the two cases: planar or cone type. Assuming that the former is true, then the magnetization curves measured in a field perpendicular to the alignment direction would be above that of the same sample with field parallel at 5 K. The magnetization curves of the aligned sample of  $\text{DyFe}_{7.91}\text{Co}_{3.39}\text{Nb}_{0.7}$  compounds shown in figure 5(a) disprove the hypothesis mentioned above, which suggests that the anisotropy below  $T_{sr}$  is a cone type. The values of spin-reorientation temperature  $T_{sr1}$  at which the spin reorientation from uniaxial to cone occurs and  $T_{sr2}$  from cone to planar are listed in table 1. A tentative magnetic phase diagram of  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  is summarized in figure 6(a). It is clear that as temperature decreases from room temperature, the EMD changes from uniaxial, going through a cone type, then to planar for  $x \leq 0.2$ , while only one spin reorientation occurs from uniaxial to cone type above 5 K for  $x = 0.25$  and  $0.3$ .

A spin-reorientation transition is associated with a change of the EMD with respect to the crystallographic direction and results from competition of transition-metal and rare-earth anisotropies as a function of temperature. In the lowest-order approximation [14], the contribution of the R atoms to the magnetocrystalline anisotropy can be expressed by the second-order anisotropy constant

$$K_1^R = -\frac{3}{2}\alpha_J \langle r^2 \rangle \langle \bar{O}_2^0 \rangle A_2^0 \quad (1)$$

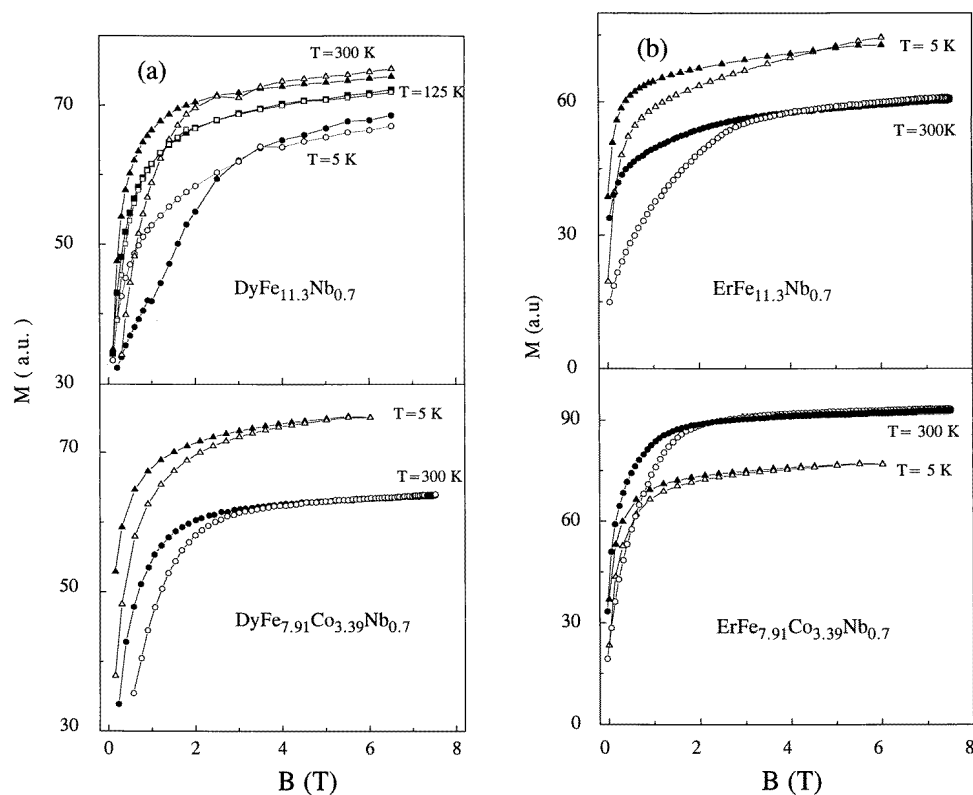
where the  $\alpha_J$  is the second-order Stevens factor,  $A_2^0$  the crystal-field parameter which depends on the crystal structure and composition of a given compound and  $\langle \bar{O}_2^0 \rangle$  the quantum-mechanical expectation value of  $\langle 3J_z^2 - J(J+1) \rangle$ . Taking  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  as an example, since  $A_2^0 < 0$  [5] and for Dy  $\alpha_J$  is negative, the second-order anisotropy constant  $K_1^{\text{Dy}} < 0$ . The total anisotropy of  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  is mainly determined by the competition of  $K_1^{\text{Dy}} (< 0)$  and  $K_1^{\text{Fe}}$



**Figure 4.** Temperature dependence of ac susceptibility of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with (a)  $R = \text{Dy}$ ,  $x = 0, 0.25$  and (b)  $R = \text{Er}$ ,  $x = 0, 0.1, 0.15, 0.2, 0.25, 0.3$ .

(>0). It is well known that the rare-earth anisotropy is dominant at low temperature while at high temperature the transition-metal anisotropy is dictatorial. Therefore with decreasing temperature, a spin reorientation of  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  compounds would be expected which



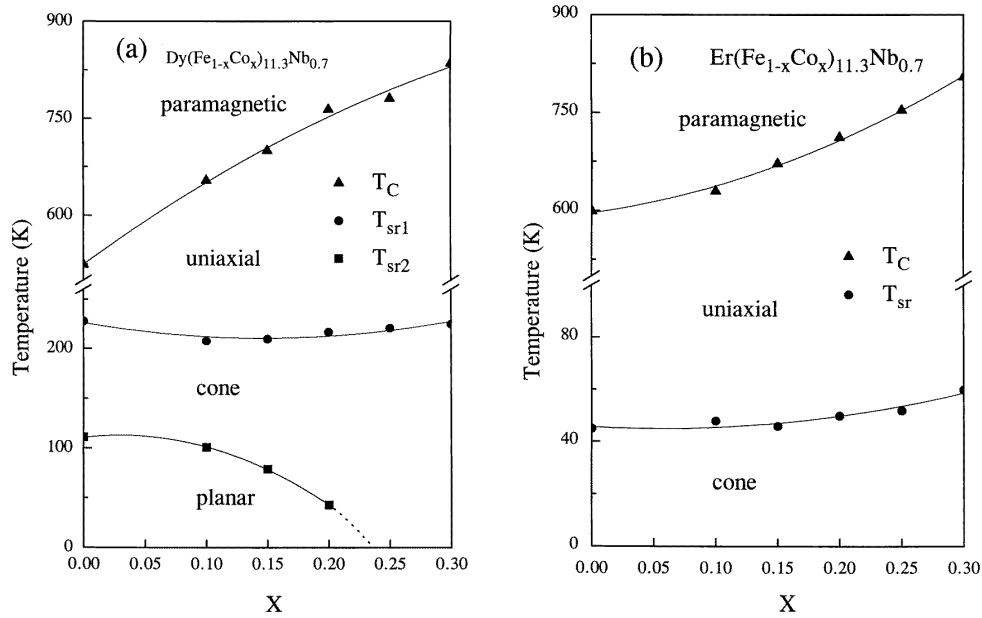


**Figure 5.** Magnetization curves of magnetically aligned samples of  $\text{R}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with  $x = 0$  and  $x = 0.3$  in an external field parallel and perpendicular to the  $c$ -axis for (a)  $\text{R} = \text{Dy}$  and (b)  $\text{R} = \text{Er}$ . The solid symbols represent the parallel direction and the open represent the perpendicular direction.

distinguish the easy-axis and easy-plane phases. The existence of the transitional easy-cone-type magnetic phase of  $\text{DyFe}_{11.3}\text{Nb}_{0.7}$  between 111 and 226 K (see table 1 and figure 6(a)) suggests that at low temperature the higher order anisotropy constants play an important role.

It can also be seen from figure 6(a) that  $T_{sr2}$  of the  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds decreases with increasing Co content and reaches zero between  $x = 0.2$  and 0.25. This may be due to the preferential occupation of Co atoms on 8f sites, where the Fe atom has almost no contribution to anisotropy while Co atom contributes an uniaxial anisotropy [15]. Therefore the substitution of Co for Fe leads to an increase of uniaxial anisotropy.

In the case of  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$ , only one anomaly can be observed in the  $M$ - $T$  (figure 2(b)) and  $\chi$ - $T$  (figure 4(b)) curves above 5 K. It has been reported [9] that there exists spin reorientation in  $\text{ErFe}_{12-x}\text{Ti}_x$  and  $\text{ErFe}_{12-x}\text{V}_x$  compounds. Similarly, the anomaly in  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds may be associated with spin reorientation. The magnetization curves shown in figure 5(b) of magnetically aligned samples of these compounds were measured at temperatures below and above  $T_{sr1}$  in a field applied perpendicular or parallel to the alignment direction to check the EMD below  $T_{sr1}$ . It can be seen that all the  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds studied show an easy-cone-type anisotropy below  $T_{sr1}$ , similar to what has been discussed for the  $\text{DyFe}_{7.91}\text{Co}_{3.39}\text{Nb}_{0.7}$  compound above. The values of  $T_{sr1}$  are listed in table 1 and the magnetic phase diagram of  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  is



**Figure 6.** Magnetic phase diagrams of  $R(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds with  $x \leq 0.3$  for (a)  $R = \text{Dy}$  and (b)  $R = \text{Er}$ . The dashed line is a guide to eye.

summarized in figure 6(b). It can be seen that  $T_{sr1}$  of  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds is almost a constant which is independent of Co content, similar to the case reported by Wang *et al* [6] for  $\text{Er}(\text{Fe}_{1-x}\text{Ni}_x)_{11.3}\text{Nb}_{0.7}$  compounds. Take  $\text{ErFe}_{11.3}\text{Nb}_{0.7}$  as an example. It is well known that in  $\text{ThMn}_{12}$ -type Fe-based compounds  $A_2^0$  is negative and  $\alpha_J$  is positive for Er. According to equation (1),  $K_1^{\text{Er}} > 0$ . Therefore, the second-order anisotropy constants of both Er and Fe sublattices contribute an easy-axis type of anisotropy. The occurrence of the spin reorientation in  $\text{ErFe}_{11.3}\text{Nb}_{0.7}$  may be related to the contribution resulting from the higher order anisotropy energy, as discussed in [6]. That the value of  $T_{sr}$  in  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  is almost independent of Co content suggests that the higher order anisotropy constant is nearly independent of Co content.

#### 4. Conclusion

The  $\text{Er}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  and  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  compounds have been synthesized in the  $\text{ThMn}_{12}$  type of structure for  $x \leq 0.3$ . With increasing Co content, the lattice constants and unit-cell volume of both  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  and  $\text{Dy}(\text{Fe}_{1-x}\text{Co}_x)_{11.3}\text{Nb}_{0.7}$  decrease while the Curie temperature increase monotonically. A maximum appears in the saturation magnetization of both compound series. All the compounds studied have EMD along the  $c$ -axis at room temperature. As temperature decreases the EMD changes from uniaxial, going through a cone type, then to the planar in  $\text{Dy}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$ , while in  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  the EMD changes from uniaxial to a cone-type. The  $T_{sr}$  of  $\text{Er}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$  compounds is nearly independent of Co content, while in the case of  $\text{Dy}(\text{Fe}, \text{Co})_{11.3}\text{Nb}_{0.7}$   $T_{sr2}$  decreases with Co content for  $x \leq 0.2$ , but for  $x \geq 0.25$ ,  $T_{sr2}$  cannot be detected above 5 K.

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