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2003 J. Phys.: Condens. Matter 15 2213

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Structure and magnetostriction of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds

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Received 11 September 2002, in final form 17 February 2003

Published 24 March 2003

Online at stacks.iop.org/JPhysCM/15/2213

Abstract

The crystal structure, Curie temperature and magnetostriction of compounds with the formula $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ were investigated by x-ray powder diffraction and magnetic measurements. It is revealed that the substitution of Fe for Co destabilizes the cubic Laves phase $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$. The maximum substitution of Fe for Co is about 0.57. The lattice constant of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$ phase increases with x increasing from $x = 0.0$ to 0.6, then remains almost unchanged from $x = 0.6$ to 0.9. The Curie temperature exhibits a variation similar to that of the lattice constant with the Fe concentration. A spin reorientation is observed, which can be understood on the basis of the two-sublattice model. The spontaneous magnetostriction constant λ_{111} shows a maximum value of 1.7×10^{-3} at $x = 0.4$. The temperature dependence of λ_{111} for $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$ exhibits an abrupt jump at 165 K, which corresponds to the temperature of the spin reorientation.

1. Introduction

The RFe_2 ($\text{R} = \text{rare earth}$) cubic Laves phase compounds are widely investigated due to their giant magnetostriction [1, 2]. TbFe_2 possesses the highest known room temperature anisotropic magnetostriction constant $\lambda_{111} = 2.4 \times 10^{-3}$ [3]. NdFe_2 has a large theoretical magnetostriction at absolute zero [2]. Little work has been done on this system because NdFe_2 can only be synthesized under high pressure [4]. The Fe sublattice contribution to the total anisotropic magnetostriction of RFe_2 is negligible compared to the R sublattice contribution. This follows from the fact that the magnetostriction in the compounds with nonmagnetic $\text{R} = \text{Y}$ or Lu does not exceed 8.0×10^{-5} [2], which is two orders lower than that in the cases with R ions having nonzero orbital moment.

Little work has been done on RCo_2 compounds due to their low magnetic ordering temperatures, although they have huge magnetostriction at low temperatures [5, 6]. The main difference in anisotropic magnetostriction between RCo_2 and RFe_2 is that the Co contribution is not negligible compared to the R contribution, albeit that the R sublattice plays a dominant role in the formation of λ_{111} -magnetostriction in both RCo_2 and RFe_2 . In addition, in the RCo_2 compounds the magnetic state of the Co sublattice is closely related to the rare-earth sublattice magnetization. For example, there are no local Co moments above the corresponding Curie temperatures in this RCo_2 Laves phases in contrast to the case for the RFe_2 Laves phases, where there are stable Fe moments below and above the Curie temperatures. The substitution of Fe for Co enhances the Curie temperatures of RCo_2 compounds, which is not only beneficial for applications purposes, but also helpful for studying the room temperature intrinsic properties of the Co-containing compounds.

For the Laves phase $\text{Nd}_{1-x}\text{Tb}_x\text{Co}_2$ compounds, the saturation moment M_S shows an anomaly at a critical concentration $x_c \approx 0.33$, where a minimum M_S was revealed, which can be explained by the two-sublattice model assuming antiferromagnetic coupling between the R sublattice moment and the Co sublattice moment [7]. The purpose of this study is to examine the effects of Fe substitution for Co on the structure and magnetostriction in the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds, which is expected to provide the possibility to tailor an improved class of Laves phase $\text{R}^{(1)}\text{R}^{(2)}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds for obtaining optimum magnetostriction properties over a wide concentration range.

2. Experimental procedure

Polycrystalline samples of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ ($x = 0.0\text{--}0.9$) were prepared by arc melting the constituent elements with a purity of 99.9% in an atmosphere of high-purity argon. The ingots were annealed at 800°C under vacuum for 14 days. X-ray powder diffraction (XRD) data were collected by a Rigaku Rint-2400 diffractometer with $\text{Cu K}\alpha$ radiation, in which the Bragg angle 2θ varies from 20° to 80° with a step of $\Delta 2\theta = 0.02^\circ$ and a scanning time of 0.2 s. The temperature dependence of the magnetization was measured by a vibrating sample magnetometer at a low field of 500 G. In order to study the intrinsic magnetostriction, the XRD patterns of some samples were deliberately recorded with a smaller step of $\Delta 2\theta = 0.01^\circ$ and a longer scanning time of 20 s at higher Bragg angles ranging from 71.5° to 74.5° .

3. Results and discussion

Parts of the XRD patterns of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds are shown in figure 1. Metallographic examination and x-ray analysis confirm that the matrix of the homogenized $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds is cubic Laves phase with MgCu_2 -type structure when $x \leq 0.5$ (figures 1(a)–(c)). A small amount of a second phase, $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_3$ with PuNi_3 structure, appears when $x = 0.6$ (figure 1(d)). With increasing Fe concentration, the content of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_3$ phase increases and it becomes the major phase when $x = 0.9$ (figure 1(f)). It is also revealed in figure 1 that the solid solubility of Fe in $\text{Nd}_{0.67}\text{Tb}_{0.33}\text{Co}_2$ is between $x = 0.5$ and 0.6. Since all the samples are Nd rich, it is very difficult to obtain the single phase $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$ under ordinary conditions when $x \geq 0.6$. Like NdFe_2 , these compounds may only be synthesized under high pressure. According to the above results, Fe is not beneficial for the formation of the MgCu_2 -type cubic Laves phase.

The lattice constant a of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$ phase in $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds is shown in figure 2. It increases steadily with increasing Fe concentration

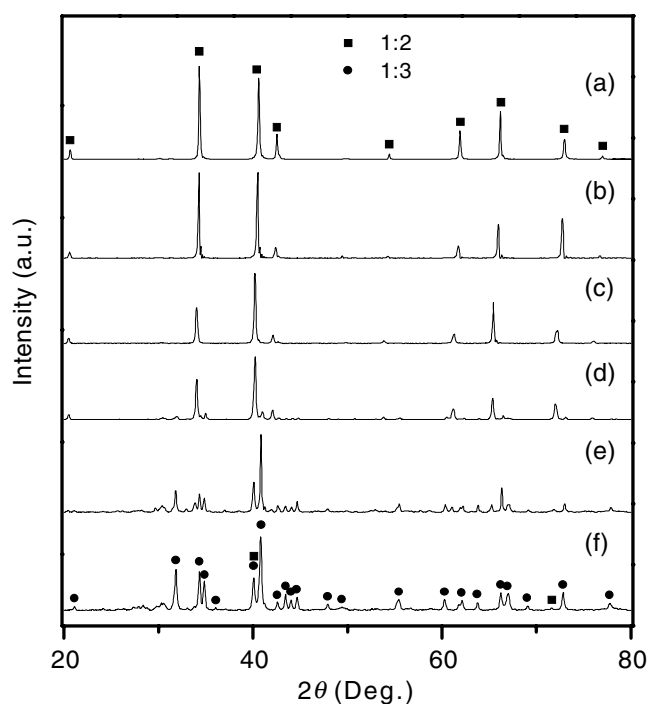


Figure 1. Parts of the x-ray diffraction patterns of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds: (a) $x = 0.1$, (b) $x = 0.3$, (c) $x = 0.5$, (d) $x = 0.6$, (e) $x = 0.7$, (f) $x = 0.9$.

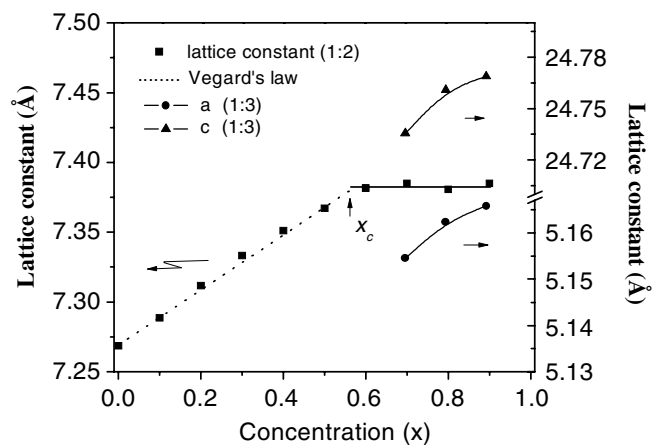


Figure 2. The concentration dependence of the lattice constant of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$. The arrow shows $x_c = 0.57$.

when $x \leq 0.5$. This can be ascribed to Fe having a larger atomic radius than Co. It can also be seen that the concentration dependence of a approximately follows Vegard's law: $a = 2xa_5 + (1 - 2x)a_0$, where a_0 and a_5 are the lattice constants of $\text{Nd}_{0.67}\text{Tb}_{0.33}\text{Co}_2$ and $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.5}\text{Fe}_{0.5})_2$, respectively. With increasing Fe concentration, a remains almost unchanged when $0.6 \leq x \leq 0.9$, which again reveals that there is a limited solid solubility x_c

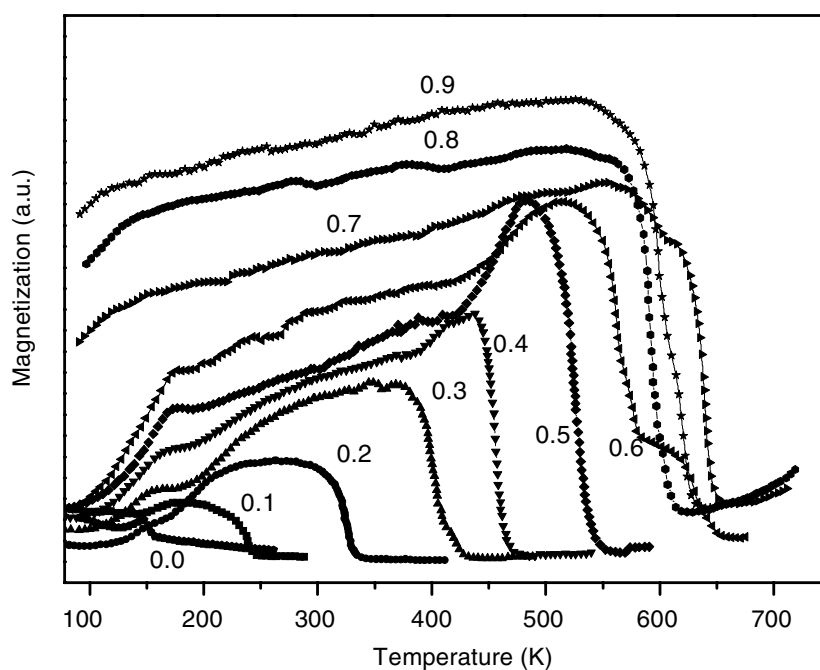


Figure 3. The temperature dependence of the magnetization of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ at a low field of 500 G.

of Fe in $\text{Nd}_{0.67}\text{Tb}_{0.33}\text{Co}_2$. Extrapolating the two linear curves to the point of intersection gives $x_c = 0.57$. The lattice constant of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_3$ phase in $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ is also shown in figure 2.

The temperature dependence of the magnetization, for all of the samples, is shown in figure 3. When the Fe concentration varies from 0.1 to 0.6, distinct kinks appear at 143, 151, 158, 165, 172 and 175 K. Such kinks may indicate a change of easy magnetization direction (EMD). Rosen *et al* [8] investigated the magnetoelasticity of SmFe_2 . A prominent peak at 195 K was observed for both the Young and shear elastic moduli at zero magnetic field. Guo *et al* [9] also found a distinct kink at 178 K in the magnetization curves of $\text{Sm}_{0.9}\text{Pr}_{0.1}\text{Fe}_2$. They claimed that the distinct kink marks the occurrence of a spin reorientation (SR). Usually SR occurs at low temperature. Within the two-sublattice model [10, 11], the R sublattice magnetocrystalline anisotropy and the transition metal (T) sublattice magnetocrystalline anisotropy may favour different EMD. Owing to the different temperature dependences of the anisotropies of the two sublattices, the competing R and T anisotropies could change the EMD of the compounds and lead to a characteristic temperature dependence of the magnetization.

The Curie temperature T_C , which is derived by extrapolating M^2 to zero on the basis of the linear part of the M^2 versus T curve, is shown in figure 4. T_C exhibits a similar variation to the lattice constant, which again reflects the limited solid solubility of Fe in $\text{Nd}_{0.67}\text{Tb}_{0.33}\text{Co}_2$. The two T_C , when $x = 0.6$ and 0.7 , correspond to those of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$ phase and the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_3$ phase, respectively, as can be clearly seen in figure 3.

The step-scanned XRD patterns at room temperature for the (440) peak of the compounds $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ ($x = 0.0-0.6$) are shown in figure 5. The effect of the $K\alpha_2$ radiation has been removed by a standard method. No splitting is found when $x = 0.0$ and 0.1 due to their T_C being below room temperature. However, the splitting is clearly seen when $x = 0.2-0.6$

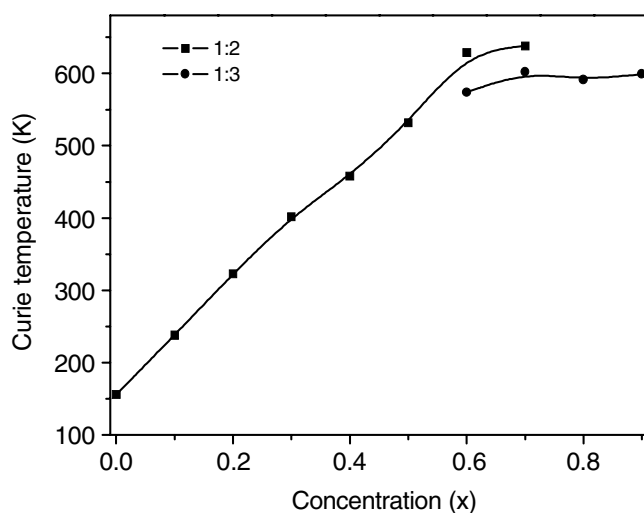


Figure 4. The concentration dependence of the Curie temperature T_C of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds.

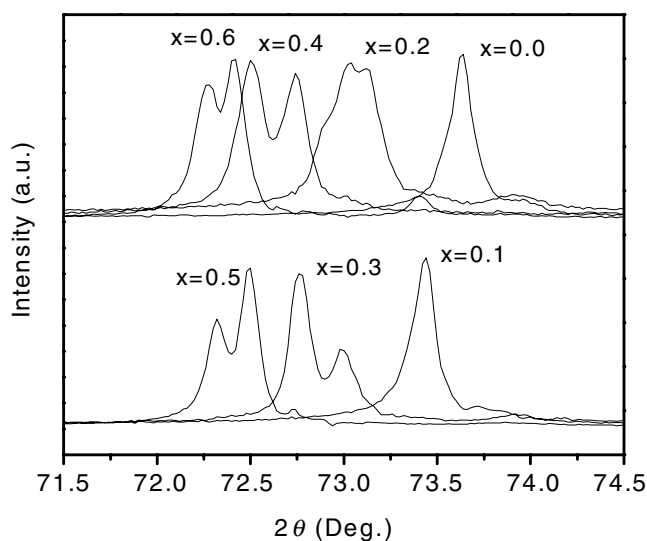


Figure 5. The step-scanned x-ray diffraction patterns of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ ($x = 0.0-0.6$) at room temperature.

and exhibits a maximum when $x = 0.4$. The splitting of the (440) peak is related to the rhombohedral distortion of the cubic lattice by $\lambda_{111} = \Delta\alpha$, where $\Delta\alpha$ is a deviation of the angle between neighbouring edges of the distorted cube from $\pi/2$ [12]. It has been shown that the splitting of some of the cubic reflections into doublets can be used conveniently to determine λ_{111} [13]. In particular, for (hhh)- and ($hh0$)-type cubic reflections, the splitting $\Delta d/d$ equals $(4/3)\lambda_{111}$ and λ_{111} , respectively. The derived spontaneous magnetostriction λ_{111} is shown in figure 6. It reveals that λ_{111} increases with the Fe concentration and then decreases. The maximum value of λ_{111} is 1.7×10^{-3} ($x = 0.4$). Guo *et al* [9] also observed a similar variation of λ_{111} in the $(\text{Sm}_{0.9}\text{Pr}_{0.1})(\text{Fe}_{1-x}\text{Co}_x)_2$ compounds.

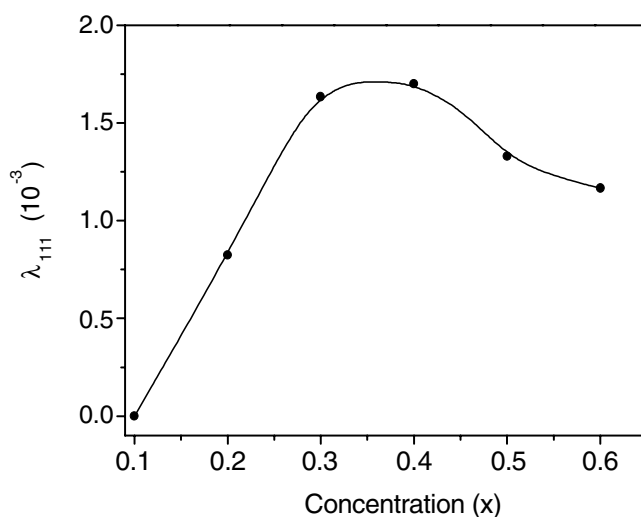


Figure 6. The spontaneous magnetostriction constant λ_{111} as a function of concentration x for $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ ($x = 0.1\text{--}0.6$) compounds.

It is accepted that the magnetic natures of the Co and Fe atoms in the compounds RCO_2 or RFe_2 are different. One has an induced moment, whereas the other has an intrinsic moment. On the one hand, the Co makes a considerable contribution to the anisotropic magnetostriction as compared to the R contribution, whereas this is not the case for Fe. On the other hand, in the compound $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$, the substitution of Fe dilutes the contribution of Co and leads to an increase of the ordering temperature accompanied by the appearance of room temperature magnetostriction. Thus the competition of the Fe and Co sublattice anisotropies changes the anisotropy of the compounds and leads to the complex concentration dependence of the spontaneous magnetostriction λ_{111} . In addition, the magnetoelastic anisotropy constant $\Delta k_1 = -(9/2)c_{44}\lambda_{111}^2$ [14], where $c_{44} = 4.87 \times 10^{11} \text{ dyn cm}^{-2}$, reflects the fact that there is a maximum magnetoelastic anisotropy when $x = 0.4$.

The sample of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$ with maximum λ_{111} was step-scanned from 123 to 543 K in order to study the temperature dependence of λ_{111} . The x-ray diffraction patterns, from which the $K\alpha_2$ radiation has been removed, are shown in figure 7. The reflection of the compounds moves to higher Bragg angle with decreasing temperature because of the shrinkage of the lattice. The two split peaks of the (440) reflection are clearly seen from 123 to 390 K. The nonsymmetric peak in the range $390 \text{ K} < T < 454 \text{ K}$ ($=T_C$) implies the existence of splitting, although it is hard to resolve the twofold peaks. No splitting is found above T_C . The temperature dependence of the magnetostriction constant λ_{111} of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$ is shown in figure 8. It exhibits an almost linear increase of λ_{111} with decreasing temperature. An abrupt jump appears in the vicinity of 165 K, which corresponds to the characteristic temperature of SR. With continuing decrease in temperature, λ_{111} increases again. According to the single-ion model [15], the temperature dependence of λ_{111} depends on the moment of the R sublattice: $\lambda_{111}(T)/\lambda_{111}(0) = I_{5/2}\{L^{-1}[\mu_R(T)\mu_R(0)]\}$, where $I_{5/2}(x)$ is the normalized hyperbolic Bessel function $(3x^{-2} + 1) - (3x^{-1}) \coth(x)$; $L^{-1}(x)$ is the function inverse to the Langevin function $\coth(x) - x^{-1}$. Callen and Callen [15] found that λ_{111} decreases monotonically with increasing temperature in RFe_2 and exhibits very good agreement with the single-ion temperature dependence. Perhaps our experimental data could also be used to examine the validity of the single-ion model. Work on this is in progress.

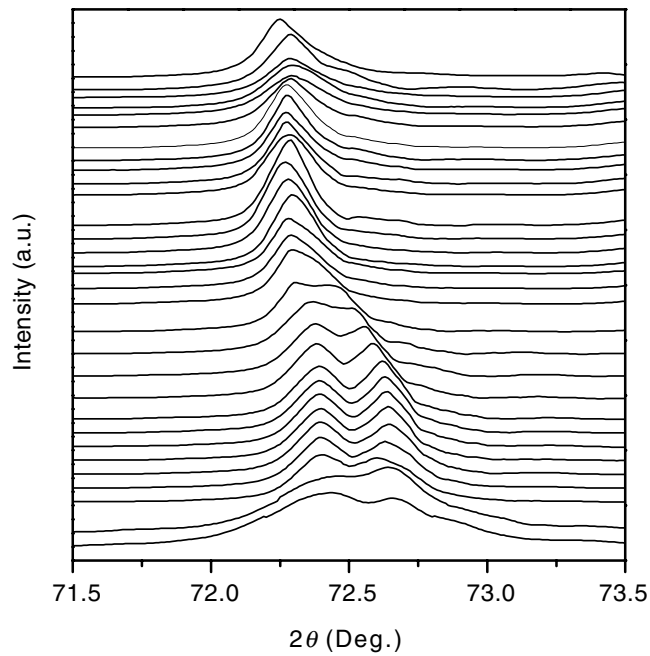


Figure 7. The step-scanned x-ray diffraction patterns of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$: from bottom to top, $T = 123, 143, 155\text{--}180$ (step = 5 K), 189, 223, 253, 298, 343, 390, 398, 403–433 (step = 10 K), 438–466 (step = 4 K), 471, 476, 503, 543 K, respectively.

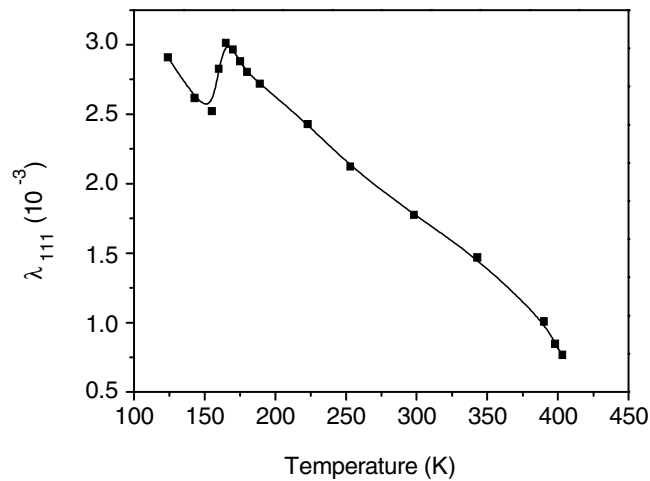


Figure 8. The temperature dependence of the magnetostriction λ_{111} of $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$.

4. Conclusions

The present investigation of the $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{1-x}\text{Fe}_x)_2$ compounds reveals that the substitution of Fe for Co has a strong effect on the formation of cubic Laves phase $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}, \text{Fe})_2$. The maximum substitution of Fe for Co is about 0.57. The observed SR

can be understood on the basis of a two-sublattice model. The spontaneous magnetostriction constant λ_{111} is obtained. A maximum value of 1.70×10^{-3} , when $x = 0.4$, is observed. The temperature dependence of λ_{111} for $\text{Nd}_{0.67}\text{Tb}_{0.33}(\text{Co}_{0.6}\text{Fe}_{0.4})_2$ exhibits an abrupt jump at 165 K, which corresponds to the temperature of the SR.

Acknowledgments

The work was supported by National Natural Science Foundation of China and the State Key Project of Fundamental Research.

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