AIP Journal of Applied Physics

Magnetostriction and magnetomechanical coupling in Ho0.85Tb0.15Fe2−x Co x system

K. R. Dhilsha, G. Markandeyulu, and K. V. S. Rama Rao

Citation: Journal of Applied Physics **70**, 4450 (1991); doi: 10.1063/1.349101
View online: http://dx.doi.org/10.1063/1.349101
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/70/8?ver=pdfcov
Published by the AIP Publishing

Magnetostriction and magnetomechanical coupling in $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ system

K. R. Dhilsha, G. Markandeyulu,[a)] and K. V. S. Rama Rao[b)]
Magnetism and Magnetic Materials Laboratory, Department of Physics, Indian Institute of Technology, Madras-600 036, India

(Received 15 April 1991; accepted for publication 18 July 1991)

Magnetostriction and magnetomechanical coupling coefficient have been measured on $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ ($x = 0$, 0.5, and 1.2) alloys. The alloys were prepared by arc melting as well as by arc melting followed by zoning using induction furnace. Saturation magnetostriction (λ_s) and magnetomechanical coupling coefficient are found to decrease with the increase of cobalt concentration. The λ_s values of the zoned samples of $x = 0$ and 0.5 are seen to be more than those of arc melted samples whereas, it is otherwise in the case of $x = 1.2$. Magnetomechanical coupling coefficient values for all the samples are seen to increase with zoning. Change in the easy direction of magnetization in the case of $x = 1.2$ is reflected in the magnetostriction of zoned samples.

I. INTRODUCTION

A program has been undertaken in our laboratory to study the influence of rare earths (RE), transition metals (TM) and hydrogen on the magnetic and electrical properties of $REFe_2$ cubic Laves phase compounds, particularly highly magnetostrictive $Ho_{0.85}Tb_{0.15}Fe_2$ and $Dy_{0.73}Tb_{0.27}Fe_2$ systems. The effect of hydrogen on $Ho_{0.85}Tb_{0.15}Fe_2$ and $Dy_{0.73}Tb_{0.27}Fe_2$ systems investigated through magnetic,[1–3] and electrical measurements[4] gave interesting results.

Recently, the x-ray, magnetic, and electrical data on $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ ($0 \leqslant x \leqslant 2.0$) system have been reported.[5] The x-ray data revealed that all the compositions retained C-15 type cubic Laves phase structure. The lattice constant varied from a value of 7.30 to 7.17 Å, going through a minima at $x = 0.5$ and sharply decreasing at $x \approx 1.5$. The Curie temperature (T_C) was found to decrease from 630 K for $x = 0$ to 130 K for $x = 2.0$ going through a maxima (690 K) at $x = 0.5$. The saturation magnetization at room temperature (RT) was found to decrease with increasing cobalt (Co) concentration with the exception of $x = 0.5$ where the value was found to be approximately the same as that of $x = 0$. The electrical resistivity data revealed occurrence of several anomalies corresponding to the spin reorientations in the system which were also reflected in some concentrations in the magnetization data.

Several authors reported the magnetic and magnetoacoustic properties of $REFe_2$ Laves phase compounds prepared in the form of rods by two techniques, viz. (i) arc melted and (ii) arc melted and then zoned using induction furnace (contained float zone or free float zone).[6–10] The samples prepared by the second method were found to contain oriented grains unlike arc cast samples. These were reflected both in the values of saturation magnetostriction as well as in the fields required for saturation. The magnetomechanical coupling coefficients were also found to be different in the two types of samples.

In this paper, we report the results of magnetostriction and magnetomechanical coupling data obtained in the rods of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ system for $x = 0$, 0.5, and 1.2 prepared by arc and arc-induction methods.

II. EXPERIMENTAL DETAILS

A. Preparation of the samples

The rods of $Ho_{0.85}Tb_{0.15}Fe_2$, $Ho_{0.85}Tb_{0.15}Fe_{1.5}Co_{0.5}$, and $Ho_{0.85}Tb_{0.15}Fe_{0.8}Co_{1.2}$ used for measuring magnetostriction and magnetomechanical coupling coefficient were prepared by two different methods.

In the first method, the alloys were prepared by arc melting the constituent elements of 99.9% purity in an argon atmosphere. The alloys were cast in the form of bars and were annealed in vacuum at 1173 K for 10 days. The bars thus obtained are designated as *A* samples.

In the second method, the constituent elements were initially melted in arc furnace. The bars obtained after several meltings were remelted in evacuated quartz tubes, using an induction furnace. The melt temperature was lowered at a rate of 12″/h and the rods thus obtained were annealed in vacuum at 1173 K for 5 days. The rods were then carefully removed from the quartz tubes and polished. These samples which are arc melted and then zoned are designated as *I* samples.

All the samples prepared were characterized by x-ray and magnetization and the results were found to agree with our earlier results.[5]

B. Magnetostriction

Magnetostriction measurements were carried out on rods using strain gauges employing Wheatstone's bridge circuit. Strain gauges (KARMA-WK-06-125BS-120) obtained from Micro-Measurements Group, USA were made use of for the measurements. The sample was held between the pole pieces of an electromagnet with the help of very thin cushions. A constant voltage of 3 V was used for the bridge excitation and the bridge output was amplified by a

[a)]CSIR Research Associate.
[b)]To whom all correspondence should be addressed.

factor of 100 using an instrumentation amplifier. The magnetostriction (λ) was determined using the formula[11]

$$\lambda=\frac{\Delta l}{l}=\frac{4\Delta E}{VK}, \tag{1}$$

where ΔE is the unbalanced bridge voltage, V is the bridge excitation voltage and, K is the gauge factor. The measurements were carried out up to a maximum magnetic field of 5 kOe.

C. Magnetomechanical coupling coefficient

Magnetomechanical coupling coefficient (k_{33}) has been determined by the three parameter method.[7] k_{33} is given by

$$[k_{33}]^2=\frac{(d_{33}^2)}{(\mu_0\mu_r S_{33}^B + d_{33}^2)}, \tag{2}$$

where d_{33} is the dynamic strain coefficient, μ_r is the incremental relative permeability, and S_{33}^B is the elastic compliance constant at constant induction (B).

The dynamic strain measurements (d_{33}) were carried out at a frequency of 70 Hz with an excitation field (H_{ac}) of 2 Oe which was provided by a primary coil kept around the sample. The samples were subjected to both ac and dc magnetic fields simultaneously along the axis of rod and the strain induced voltages were measured using a PAR Lock-in amplifier. Vector subtraction method[7] was used for separating the stray pickup. d_{33} was determined using the formula

$$d_{33}=\frac{(\delta l/l)_{ac}}{H_{ac}}. \tag{3}$$

Measurements were carried out for different bias fields up to 1.6 kOe.

To measure the incremental relative permeability (μ_r), in addition to the primary coil, a secondary coil was also wound on the sample rod for detecting the pickup voltage. An ac field of 2 Oe operating at 70 Hz was employed for excitation. The ac pickup voltage was measured using a lock-in-amplifier for various bias fields. The experiment was repeated using a perspex rod in place of the sample rod and μ_r was calculated.

The elastic compliance constant (S_{33}) of the sample was measured with the same arrangement as employed for measuring μ_r. Here the induced pickup voltage was plotted as a function of frequency using a lock-in-amplifier in phase with the reference drive current and for various bias fields. S_{33} was calculated using the formula

$$S_{33}^B=\frac{1}{4L^2\rho f_A^2}, \tag{4}$$

where L is the length, ρ the density of the rod, and f_A the antiresonance frequency. The density of the rods were measured by determining apparent weight loss of the sample in water using a Mettler balance. Using the values of d_{33}, μ_r, μ_0, and S_{33}^B, k_{33} was determined from Eq. (2).

TABLE I. Saturation magnetostriction (λ_s) values of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$.

	$\lambda_s(\times 10^{-6})$		
Composition	A sample	I sample	Calculated
$x=0$	320 ± 10	335 ± 10	330
$x=0.5$	280 ± 10	315 ± 10	283
$x=1.2$	230 ± 10	200 ± 10	216

III. RESULTS

The λ_s values for A and I samples are given in Table I. The error in the magnetostriction measurements is estimated to be around 4%. It is seen that the λ_s values decrease with the increase of x in both the cases. In the case of $x=0$ and $x=0.5$, λ_s[I] is seen to be larger than $\lambda_s[A]$, whereas it is otherwise in the case of $x=1.2$. Figure 1 shows the λ-H plots for A and I samples. No hysteresis was observed in the magnetostriction measurements.

Resonance curves were obtained for A and I samples. Figure 2 shows typical curves in the case of I sample of $Ho_{0.85}Tb_{0.15}Fe_2$ with different applied bias fields. The magnetomechanical coupling coefficient (k_{33}) obtained for A and I samples as a function of bias fields are given in Fig. 3. The k_{33} value is found to decrease with the increase of Co concentration.

IV. DISCUSSION

The value of $\lambda_s(320\times 10^{-6})$ for $x=0$ of A samples lies close to the reported value of 325×10^{-6} by Koon *et al.*[12] for arc melted sample. The λ_s value is seen to decrease with the increase of Co content for both A and I samples.

In the case of (Ho,Tb)Fe_2 system, the value of λ_s is reported to vary linearly from 80×10^{-6} for $HoFe_2$ to 1750×10^{-6} for $TbFe_2$ in arc cast samples.[12] Using this fact, λ_s for $Ho_{1-y}Tb_yFe_2$ can be expressed as

$$\lambda_s[Ho_{1-y}Tb_yFe_2]=(1-y)\lambda_s[HoFe_2]+y\lambda_s[TbFe_2]. \tag{5}$$

λ_s value for $Ho_{0.85}Tb_{0.15}Fe_2$ evaluated from this equation agrees well with the experimentally determined value as seen from Table I.

The above empirical calculation has been extended to (Ho,Tb)(Fe,Co)$_2$ system. Here while doing so, the single-ion model for magnetocrystalline anisotropy of (Ho,Tb)Fe_2 system[12] has been assumed to be valid for Co substituted systems as well. In this case, λ_s can be expressed as

$$\begin{aligned}\lambda_s[Ho_{1-y}Tb_yFe_{2-x}Co_x] &=(1-y)\{\{(2-x)/2\}\lambda_s[HoFe_2] \\ &\quad + x/2\,\lambda_s[HoCo_2]\}+y\{\{(2-x)/2\}\lambda_s[TbFe_2] \\ &\quad + x/2\,\lambda_s[TbCo_2]\}.\end{aligned} \tag{6}$$

The extension of Eq. (5) for Co substituted systems is justified on the basis that the single-ion model for anisot-

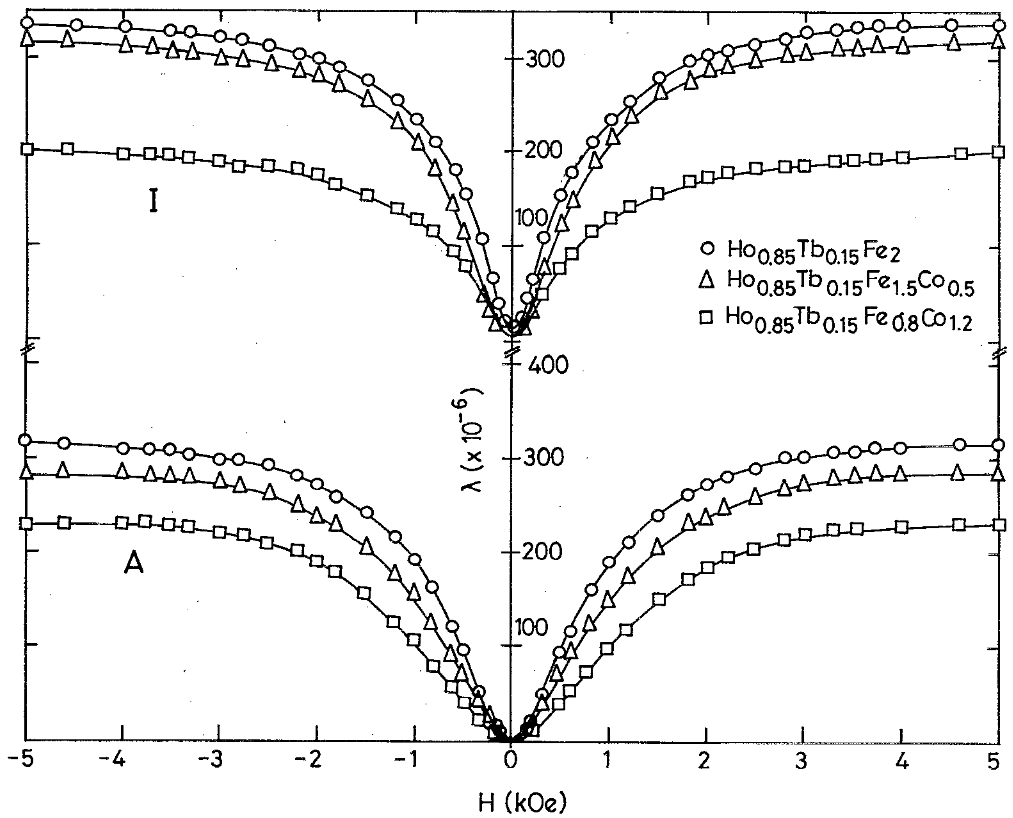

FIG. 1. Magnetostriction of A and I samples of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ as a function of applied field at RT.

ropy as well as magnetostriction is also applicable for these systems.[13,14] The λ_s value of $HoCo_2$ and $TbCo_2$ taken are those at 77 K.[15] It can be seen from Table I that the experimentally determined λ_s value for $x = 0.5$ and 1.2 of A samples are in agreement with the values calculated using Eq. (6).

The normalized magnetostriction (λ/λ_s) versus applied field for A and I samples are given in Figs. 4 and 5, respectively. The aspect ratio of the rods was greater than 5 and therefore the demagnetizing effects could be neglected.[16] From a comparison of the (λ/λ_s) versus field curves for three concentrations of A and I samples, it can be seen that the I samples tend to saturate at lower fields compared to the A samples. Thus an enhancement in the λ_s values and a low anisotropy in I samples compared to A

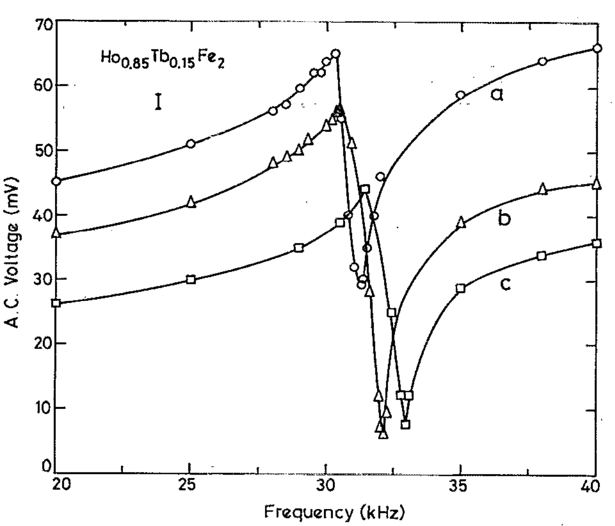

FIG. 2. Typical variation of resonant peaks with frequency for an I sample of $Ho_{0.85}Tb_{0.15}Fe_2$ of 5.39-cm long. (a) 100 Oe, (b) 400 Oe, and (c) 1000 Oe.

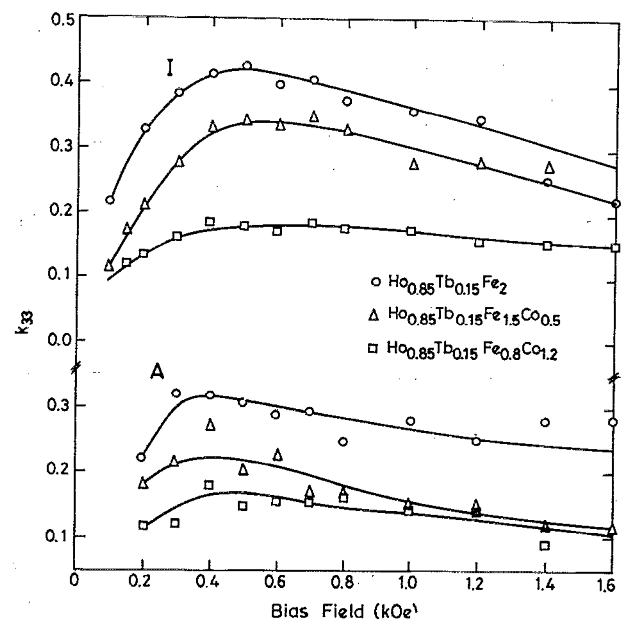

FIG. 3. Variation of k_{33} values for A and I samples of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ with dc bias field.

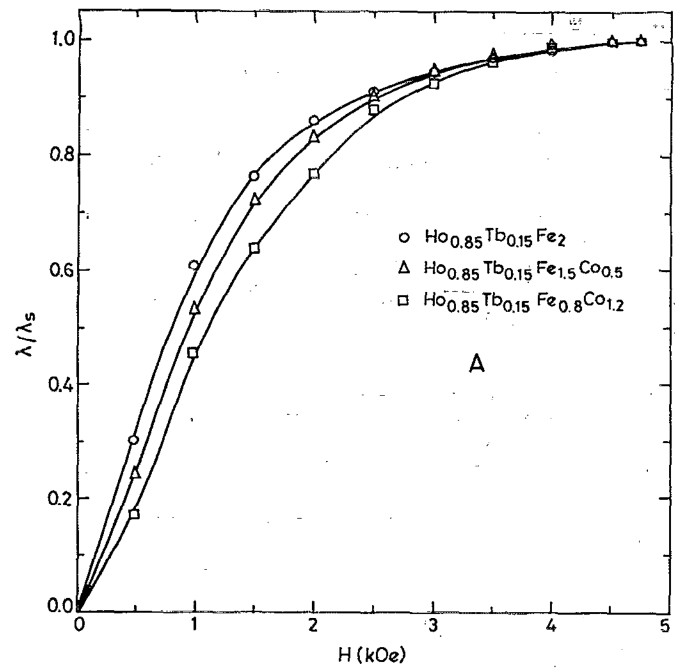

FIG. 4. Normalized magnetostriction as a function of applied bias field for *A* samples of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$.

samples would probably be because of a preferred grain orientation in *I* samples. The orientation of grains in *I* samples is also reflected in the density values (Table II) wherein they are seen to be smaller than those of *A* samples. This is probably due to large volume of voids in *I* samples compared to *A* samples. Savage *et al.*[6] reported

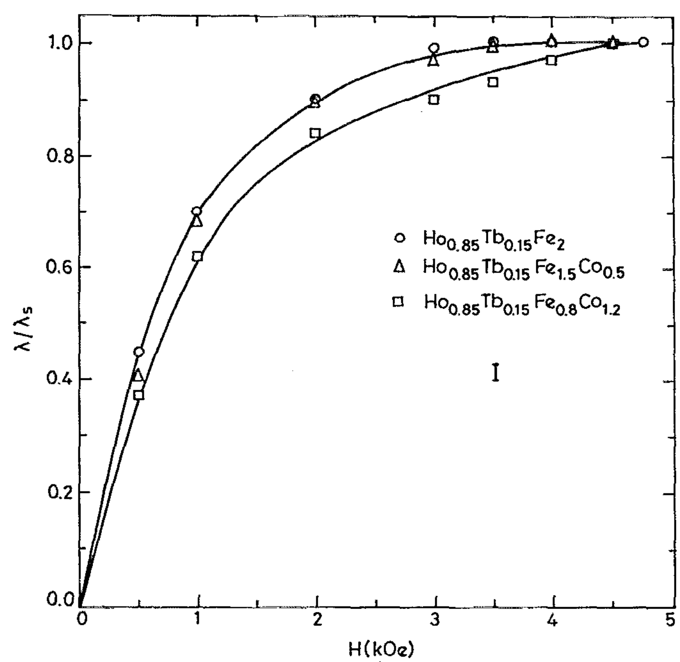

FIG. 5. Normalized magnetostriction as a function of applied bias field for *I* samples of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$.

TABLE II. Density values of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$.

	Density (g/cc)		
		Measured	
Composition	Calculated[a]	*A* sample	*I* sample
$x=0$	9.41	9.27	9.14
$x=0.5$	9.54	9.33	9.29
$x=1.2$	9.58	9.53	9.44

[a]Using lattice parameters, Ref. 5.

larger values of magnetostriction and magnetomechanical coupling in grain oriented samples of $Tb_{0.27}Dy_{0.73}Fe_2$ prepared by free-float zoning, compared to arc melted samples.

In *I* samples, the field dependence of λ (Fig. 5) also indicates an increase in magnetocrystalline anisotropy of the system when the Co concentration is increased to $x = 1.2$. Hirosawa[17] has reported the presence of a large and anisotropic orbital moment associated with the Co atoms in RE-Co intermetallics leading to the presence of magnetocrystalline anisotropy in the Co sublattice through the spin-orbit coupling. This may be adding up to the anisotropy in the RE sublattice in the case of $x = 1.2$.

Another important feature of (λ/λ_s) versus field curves for *I* samples is that the $x=0$ and $x=0.5$ samples behave in a very similar way, whereas it is not so in the case of $x=1.2$. The saturation magnetostriction as well as the field dependence of λ are dependent on the easy direction of magnetization and the preferred orientation. In $REFe_2$ system,[18] the magnetostriction is highly anisotropic with $\lambda_{111} \gg \lambda_{100}$. The smaller value of λ_s observed for *I* sample compared to *A* sample in the case of $x = 1.2$ may be due to the change in the easy direction of magnetization compared to $x=0$ and 0.5. Mössbauer[2,19] and Torque magnetometry[20] measurements on $Ho_{0.85}Tb_{0.15}Fe_2$ system showed [111] as easy direction of magnetization at RT. Dhilsha and Rama Rao[5] from resistivity measurements suggested that the easy direction lies along [111] for $x=0$ and 0.5 and changes to [110] for $x = 1.2$, at RT.

The k_{33} values for *A* and *I* samples are given in Fig. 3. The peak k_{33} obtained for $x=0$ of *A* samples, 0.32, is agreeing with the value 0.34 reported by Timme[21] on arc cast samples. The k_{33} values are found to decrease with the increase of Co concentration for both *A* and *I* samples. In the case of $x=0$ and 0.5, k_{33} values for *I* samples are seen to be clearly higher than those for *A* samples, whereas in the case of $x = 1.2$ the increase is marginal. Branwood *et al.*[7] have reported marked improvements in magnetomechanical coupling accompanying increased grain size, for zoned samples of $Tb_{0.27}Dy_{0.73}Fe_2$.

A slight shift of the k_{33} peak to the higher bias field also is indicative of an increase in the anisotropy of the system with the Co substitution. The k_{33} curves show a peak for $x=0$ and 0.5 compositions, whereas almost a constant value of k_{33} is seen in the case of $x = 1.2$.

Measurements of the angular variation of magnetization were performed to observe the effect of grain orientation on the magnetization in the I samples. Similar experiments were also carried out on A samples for comparison. For this purpose cylinders of 3-mm diameter and 3-mm height were prepared in such a way that the direction of preferred orientation is perpendicular to the cylindrical axis. These were annealed at 623 K for 12 h and cooled slowly.

In the case of $x=0$ of A samples, no anisotropy was observed whereas, for I sample, a small anisotropy of the order of 2–3 emu/gm was observed, suggesting the orientation of grains. In the case of $x=0.5$, no appreciable anisotropy was observed in both A and I samples. This may be due to the fact that the spin reorientation in this system occurs just above 300 K (Ref. 5), thereby leading to the realization of an unperceptible anisotropy.

However, in the case of $x=1.2$, for both the I and A samples, anisotropy of the order of 3–4 emu/gm was observed even though one would not expect such a behavior in A sample. In order to understand the unusual behavior in A sample, one should consider the origin of anisotropy arising out of shape of grains and their physical alignment in respect to each other and the magnetocrystalline anisotropy due to Co. One can rule out the first possibility since the A samples are cooled fast which may not lead to oriented grains. On the other hand, ^{59}Co nuclear magnetic resonance data available on $RECo_2$ samples[17] suggest the presence of large magnetocrystalline anisotropy arising due to the anisotropic orbital moment associated with the Co atom. Therefore, in the present case ($x=1.2$), the magnetocrystalline anisotropy arising out of the introduction of Co is probably responsible for unusual behavior in A sample.

ACKNOWLEDGMENTS

The authors thank the Department of Electronics, Government of India for supporting this research program. One of the authors (KRD) is grateful to the Indian Institute of Technology, Madras for financial support.

[1] S. Annapoorni, G. Markandeyulu, and K. V. S. Rama Rao, J. Appl. Phys. **65**, 4955 (1989).
[2] S. Annapoorni and K. V. S. Rama Rao, J. Appl. Phys. **67**, 424 (1990).
[3] S. Annapoorni, G. Markandeyulu, and K. V. S. Rama Rao, J. Appl. Phys. **68**, 1394 (1990).
[4] S. Annapoorni, G. Markandeyulu, and K. V. S. Rama Rao, J. Phys. Soc. Jpn. **59**, 3014 (1990).
[5] K. R. Dhilsha and K. V. S. Rama Rao, J. Appl. Phys. **68**, 259 (1990).
[6] H. T. Savage, R. Abbundi, A. E. Clark, and O. D. McMasters, J. Magn. Magn. Mater. **15–18**, 609 (1980).
[7] A. Branwood, R. Dudley, A. L. Janio, and A. R. Piercy, IEE Bulletin **60**, 411 (1986).
[8] A. E. Clark, J. P. Teter, and O. D. McMasters, IEEE Trans. Mag. **MAG-23**, 3526 (1987).
[9] A. G. I. Jenner, D. G. Lord, and C. A. Faunce, IEEE Trans. Mag. **24**, 1865 (1988).
[10] J. S. Abell, D. Butler, R. D. Greenough, V. Joyce, and K. C. Pitman, J. Magn. Magn. Mater. **62**, 6 (1986).
[11] M. A. Chaudhri, A. A. Joraide, and W. D. Corner, J. Phys. E **17**, 287 (1984).
[12] N. C. Koon, A. I. Schindler, C. M. Williams, and F. L. Carter, J. Appl. Phys. **45**, 5389 (1974).
[13] F. Pourarian, Phys. Lett. **72A**, 175 (1979).
[14] R. Z. Levitin and A. S. Markosyan, J. Magn. Magn. Mater. **84**, 247 (1990).
[15] E. W. Lee and F. Pourarian, Phys. Status Solidi (A) **34**, 383 (1976).
[16] B. D. Cullity, *Introduction to Magnetic Materials* (Addison Wesley, Reading, MA, 1972).
[17] Satoshi Hirosawa, Ph. D. thesis, Kyoto University, Japan, 1981.
[18] A. E. Clark, *Ferromagnetic Materials, Vol. I*, edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980).
[19] U. Atzmony, M. P. Dariel, E. R. Bauminger, D. Lebenbaum, I. Nowik, and S. Ofer, Phys. Rev. Lett. **28**, 244 (1972).
[20] C. M. Williams and N. C. Koon, Solid State Commun. **27**, 81 (1978).
[21] R. W. Timme, J. Acoust. Soc. Am. **59**, 459 (1976).