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Investigation of magnetic, magnetomechanical, and electrical properties of the $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ system

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X-ray-diffraction studies carried out on $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ ($x=0,0.5,1.2,2.0$) revealed that all compositions investigated retained the C-15 cubic Laves phase structure. Magnetization and electrical resistivity measurements were carried out in the temperature range 77–800 K. Anomalies observed in the resistivity measurements have been identified corresponding to spin reorientation transitions occurring in this system. Some of these transitions have also been observed in the magnetization data. Magnetostriction and magnetomechanical coupling coefficient were measured on samples of $x=0$, 0.5, and 1.2 prepared by arc melting as well as arc melting followed by zoning using an induction furnace. Magnetostriction and coupling coefficient were found to decrease with increase in cobalt concentration. Magnetostriction data on zoned samples revealed a change in the easy direction of magnetization from [111] to [100] in the case of $x=0.5$.

I. INTRODUCTION

Recent investigations of the magnetic, magnetomechanical, and electrical resistivity measurements carried out on the $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ system in our laboratory gave interesting results.[1,2] It was found that all compositions ($x=0,0.5,1.2$ and 2.0) investigated retained $MgCu_2$-type cubic Laves phase structure. The Curie temperature T_C increased from 630 K ($x=0$) to 690 K ($x=0.5$) and decreased to 130 K for $x=2.0$. The electrical resistivity data revealed the occurrence of several anomalies corresponding to spin reorientation transitions in the system which were also reflected in the magnetization data. An increase in the spin reorientation temperature toward high temperatures was also observed for low cobalt (Co) concentrations. Saturation magnetostriction λ_s and magnetomechanical coupling coefficient k_{33} measurements carried out on the samples of $x=0$, 0.5, and 1.2 prepared by arc melting (A samples) as well as arc melting followed by zoning using an induction furnace (I samples) were found to decrease with increase in Co concentration. A change in the easy direction of magnetization from [111] for $x=0$ to [110] for $x=1.2$ was reflected in the measurements carried out on I samples.

In continuation of such studies, we report here the investigations of the effect of Co on the magnetic, magnetomechanical, and electrical properties of the $Tb_{0.27}Dy_{0.73}Fe_2$ system.

II. EXPERIMENTAL DETAILS

The alloys were prepared by arc melting the constituent elements of 99.9% purity in an argon atmosphere at a pressure of 860 Torr and remelted several times for good homogeneity. The alloys were cast in the form of rods. These were annealed in vacuum at 1173 K for 10 days and the rods are identified as A samples.

The rods were also prepared by a different method. In this case, initially the alloys were cast in the form of rods in the arc furnace. Then these rods were remelted in evacuated quartz tubes using an induction furnace and the samples were lowered at a rate of 12 in. per hour. The rods which are designated as I samples were then annealed at 1173 K for 5 days.

X-ray-diffraction patterns taken at RT with Cu$K\alpha$ radiation showed the formation of a single phase with $MgCu_2$-type structure for all the samples prepared. The magnetization of these samples were measured from 77 K to above their T_C using a PAR-155 vibrating sample magnetometer (VSM). The temperatures were maintained to an accuracy of ± 1 K and the measurements were carried out in an applied magnetic field up to 10 kOe.

The samples for electrical resistivity measurements were cut from arc-melted and annealed rods using a diamond cutter. They were mechanically ground to get uniform thickness and then annealed at 1000 K for 24 h in order to remove any strain developed during the preparation. The dimensions of the samples were approximately of 7 mm diameter and 1 mm thickness.

Electrical resistivity measurements were carried out using a four-probe van der Pauw technique in the temperature range 77–800 K and the temperatures were maintained to an accuracy better than ± 1 K. A constant current of 90 mA was applied and the voltages were measured using a 181 Keithley nanovoltmeter.

The details of the measurements of magnetostriction and magnetomechanical coupling coefficient adopted were described earlier.[2] The magnetostriction measurements were carried out in an applied field up to 5.2 kOe.

III. RESULTS

The lattice constants were determined from x-ray diffractograms. The variation of the lattice constant as a function of Co concentration is given in Fig. 1. The values of

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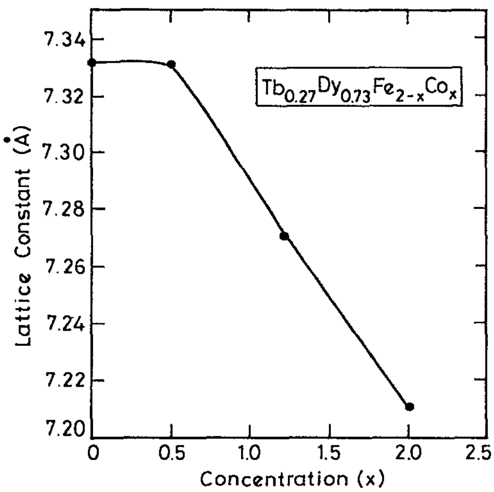

FIG. 1. Variation of lattice parameter of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ with x.

magnetic moments obtained from magnetization at 300 K (RT), 77 K, and T_C for different Co concentrations are given in Fig. 2. The RT magnetization values (magnetic moment) decreased with increase in Co concentration whereas the T_C increased from 660 K for $x=0$ to 700 K for $x=0.5$ and decreased to 195 K for $x=2.0$. The temperature variation of magnetization from 77 to 750 K is given in Fig. 3 and showed anomalies for $x=0$, 0.5, and $x=1.2$ in the low-temperature region.

The temperature variation of electrical resistivity ρ for $x=0$, 0.5, 1.2, and 2.0 is shown in Fig. 4. No thermal hysteresis was observed. ρ was found to increase with increase in Co content and several anomalies were observed in the temperature range investigated.

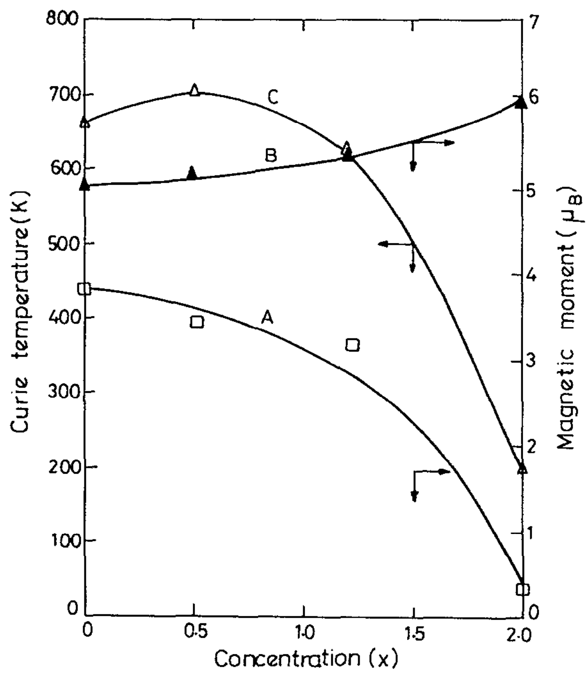

FIG. 2. Variation of (A) magnetic moment at 300 K (RT), (B) magnetic moment at 77 K, and (C) Curie temperature for different Co concentrations of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$.

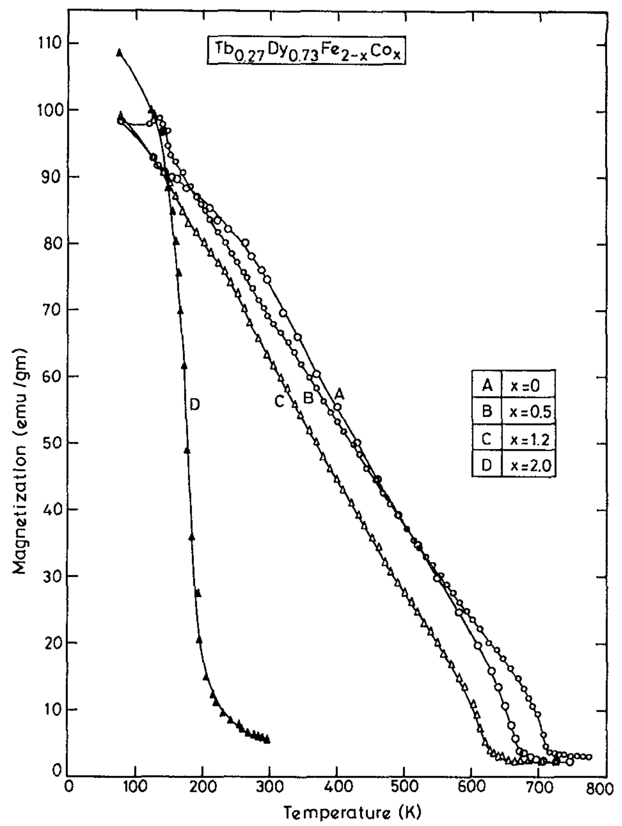

FIG. 3. Temperature variation of magnetization at an applied field of 10 kOe of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$.

The magnetostriction λ as a function of applied field for A and I samples is given in Fig. 5. As can be seen from the graph, λ could not be saturated within the field studied. The error in the measurements of λ is estimated to be $\sim$4%. The values of λ decreased with increase of Co concentration in both A and I samples.

The k_{33} values obtained for A and I samples as a function of bias field are found to decrease with increase of Co concentration as shown in Fig. 6.

IV. DISCUSSION

The value of the lattice constant of 7.33 Å obtained for $x=0$ agrees with the previously reported values.[3,4] This value remains almost the same for $x=0$ and 0.5, whereas it decreases sharply for $x=1.2$ and 2.0 suggesting a deviation from Vegard's law. Such variations were also reported in $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$,[1] $SmFe_{2-x}Co_x$,[5] and $Y(Fe_{1-x}Co_x)_2$ (Ref. 6) systems. Recently Eriksson *et al.*[7] from their band-structure calculations ascribed this behavior to magnetovolume effects. On the other hand, in $Ho_{0.85}Tb_{0.15}Fe_2H_x$ and $Tb_{0.27}Dy_{0.73}Fe_2H_x$ systems,[3] it was found that the lattice constant increased linearly with increasing hydrogen concentration.

A rhombohedral distortion of the cubic lattice (below the ordering temperature) was reported[8-10] for compounds

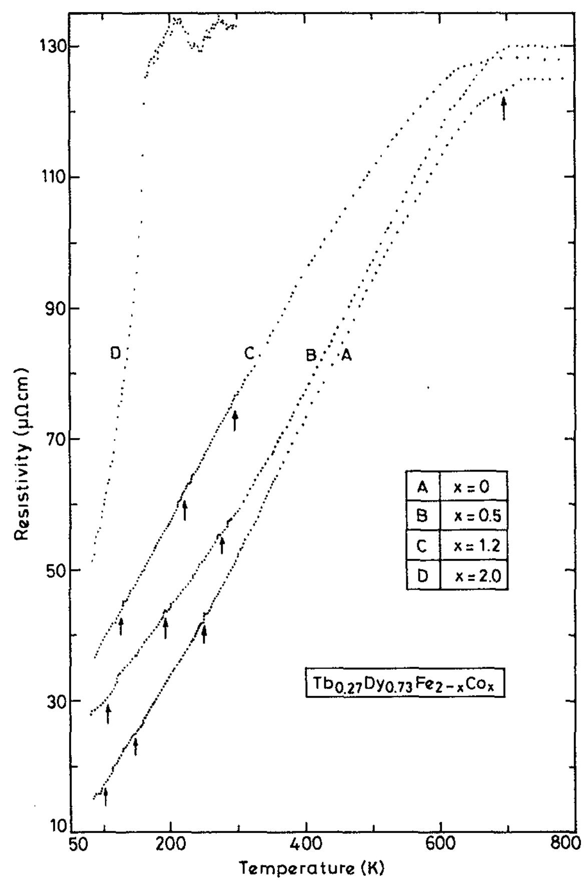

FIG. 4. Temperature variation of electrical resistivity of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$. Anomalies observed are marked by arrows.

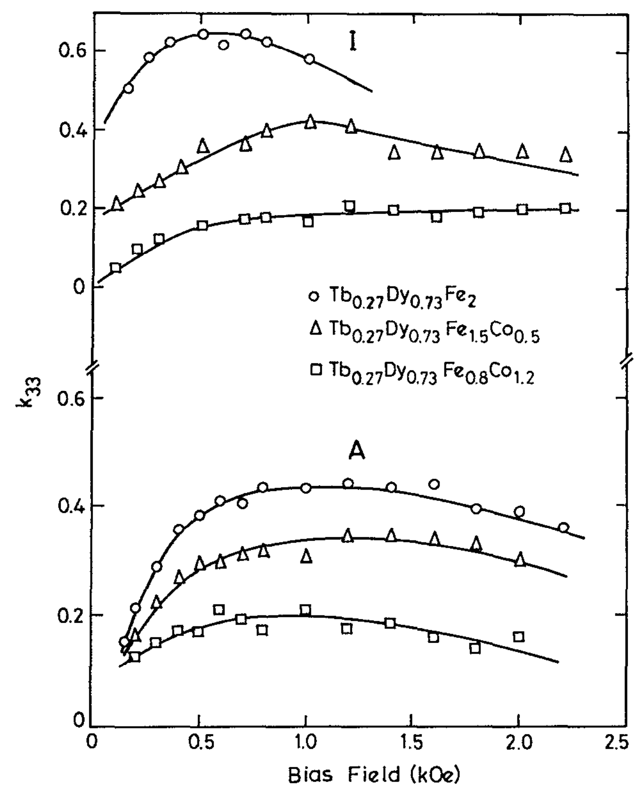

FIG. 6. Variation of k_{33} values for A and I samples of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ with dc bias field.

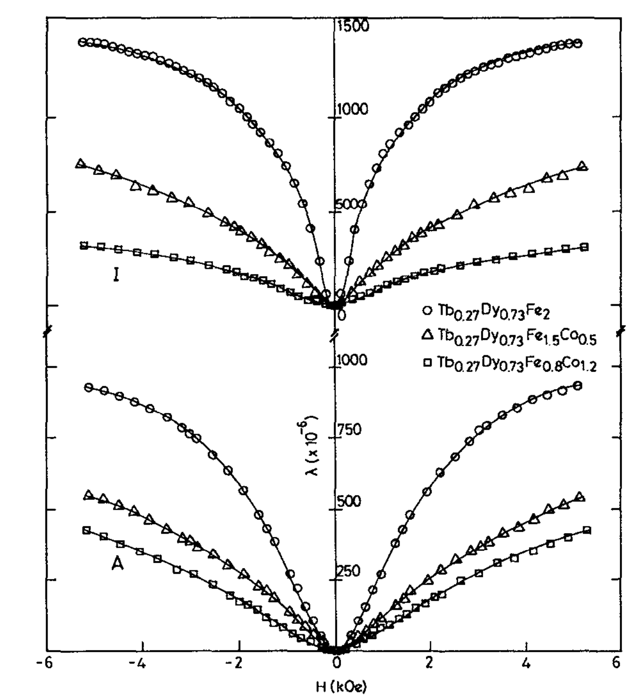

FIG. 5. Magnetostriction of A and I samples of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ as a function of applied field at RT.

containing Tb as well as Sm and this distortion was also found to be dependent on the easy axis of magnetization. As in the case of other $REFe_2$, $RECo_2$ compounds (where RE designates rare earth), it appears that the structural distortions are due to magnetoelastic interactions and the rhombohedral or orthorhombic distortions arise when the easy axis is in [111] or [110] direction, respectively. In the $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$ system,[2] at RT, the easy direction of magnetization was found to be along [111] for $x=0$ and 0.5 suggesting a rhombohedral distortion whereas in the case of $x=1.2$, the easy direction was along the [110] corresponding to a orthorhombic distortion.

The RT magnetization values (magnetic moment) were found to decrease with increasing Co concentration and this variation is shown in Fig. 2(A). The magnetic moments from the magnetization data at 77 K remained constant around 5.0 μ_B until $x=1.2$ and then increases to $5.9\mu_B$ for $x=2.0$. Since the T_C values lie around 660 ± 40 K for $x=0$, 0.5, and 1.2, the observed value of $5.3\mu_B$ corresponds to a near-saturation value. However, since T_C is well below (~ 195 K) RT for $x=2.0$, the observed value of $5.9\mu_B$ at 77 K may not be a saturated value.

The values of ρ increased with the increase in Co content in $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$. The T_C values observed from the resistivity measurements agree well with the magnetization data for $x=0$, 0.5, and 1.2; however, in the case of $Tb_{0.27}Dy_{0.73}Co_2$, a first-order transition corresponding to T_C was observed at 166 K. On the other hand, the magnetization measurements carried out in an applied field of

TABLE I. Anomalies observed in $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ system.

	Magnetization	Electrical resistivity data			
x	data (T_S) (K)	T_{sp} (K)	T_{S1} (K)	T_{S2} (K)	T_{S3} (K)
0.0	135	640–720	110	160	250
0.5	135	···	110	190	285
1.2	160	···	125	225	290

10 kOe gave a value of 195 K. This value decreased to 175 K when measurements were made at 0.5 kOe.

In $RECo_2$ compounds,[11] the RE - Co exchange interaction is dominant and the magnetization at the ordering point arises in a jumplike fashion for $HoCo_2$ and $DyCo_2$. This first-order transition has been attributed to the metamagnetism of Co. The reason for the difference in T_C values observed from resistivity measurements and magnetization could probably be attributed to the induced magnetic moment on the Co when external field is applied while doing magnetization measurements. In $Ho_{0.85}Tb_{0.15}Co_2$ (Ref. 1) also, the T_C values observed from magnetization measurements gave a higher value compared to the value obtained from resistivity data. Similar observations were also reported by Steiner *et al.* in (Ho,Y) Co_2.[12]

Another important observation in the resistivity measurements is the anomaly observed just above T_C (a minimum in the resistivity measurements) for $Tb_{0.27}Dy_{0.73}Co_2$ and was not observed in the case of $Ho_{0.85}Tb_{0.15}Co_2$. This can be attributed to the role of RE atoms affecting the spin fluctuation scattering in $RECo_2$ systems.[12,13]

The measurement of the temperature variation of magnetization and resistivity in $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ showed anomalies as listed in Table I.

Several authors[14–18] have carried out various measurements on single crystals to identify the spin reorientation transition temperatures occurring in $Tb_{0.27}Dy_{0.73}Fe_2$. From the torque magnetometry and thermal-expansion measurements by Sato *et al.*,[14] it was found that the easy direction of magnetization is along [110] below 23 K above which it changes to [100]. It lies along [100] up to 285 K above which it changes to [111]. On the other hand, measurements carried out by Methasiri and Tang[15] suggested [100] as the easy direction in the temperature range 300–130 K and changing to [110] easy axis through a [*uv*0] phase in the range 130–80 K. Earlier measurements by Mori and co-workers[16] suggested [111] as the easy direction of magnetization in the range 298–280 K, [100] in the range 280–65 K, and along [110] below 65 K through a [*uuw*] phase. However, recent x-ray measurements by Al-Jiboory and Lord[17] confirmed the easy axis as [111] in the range 373–270 K and [100] in the range 270–173 K.

Our resistivity measurements for $x=0$ show three anomalies corresponding to spin reorientation transitions T_{S1}, T_{S2}, and T_{S3} in the temperature range investigated. Comparing our results with the data available in the literature, the anomaly (T_{S1}) occurring at 110 K corresponds to a transition from [110] to [*uv*0] and the anomaly occurring at 160 K corresponding to T_{S2} could be from [*uv*0] to [100]. The one corresponding to T_{S3} at 250 K can be attributed to a change from a [100] phase to [111]. In $x=0.5$ and 1.2 also, three transitions for each composition corresponding to T_{S1}, T_{S2}, and T_{S3} were observed. These probably, correspond to similar type of spin reorientations occurring for $x=0$. Our magnetization data gave indication of a single anomaly occurring at 135 K for $x=0$ and 0.5 and at 160 K for $x=1.2$. Comparing this data with the literature, it can be suggested that these correspond to a change in the easy direction from [110] as one goes from low to high temperature.

The resistivity data gave indication of all possible spin reorientation transitions. This can be understood if we consider the various contributions to the resistivity. The resistivity ρ_{total} arises from residual resistivity ρ_0, lattice contribution ρ_{ph}, and magnetic contribution ρ_{mag}.[19] Of these, if the magnetic contribution (spin disordered) predominates, the resistivity almost remains constant above T_C. Furthermore, Ozhogin and Preobrazhenskii[20,21] have reported giant magnetoacoustic nonlinearity in the vicinity of spin reorientation transitions giving rise to an anomalous growth in the magnon-phonon coupling owing to a decrease in the activation energy of one of the branches of the magnon spectrum, due to magnetostrictive coupling of the elastic and spin subsystems. If there is a change in the easy direction of magnetization, ρ_{mag} gets affected thereby causing anomalies in the resistivity data.

Another anomaly T_{sp} occurs very close to T_C as a dip for $x=0$ and is a broad one extending in the range 640–720 K. Such anomalies were also observed by us near T_C in the resistivity measurements of $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$.[1] This can be attributed to a strong magnon-phonon coupling (due to magnetoelastic coupling) and an uneven softening of the phonon vibrational modes due to spin fluctuations near T_C.[1,22]

Magnetostriction measurements were carried out at RT for A and I samples of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ ($x=0$, 0.5, and 1.2) in an applied field up to a maximum available of 5.2 kOe. Saturation could not be attained with this field, but the value of 930×10^{-6} for $x=0$ for A samples lies very close to the reported λ_s value of 1000×10^{-6} by Savage *et al.*[23] for arc-melted samples. The magnetostriction values were found to decrease with increase in Co concentration in both A and I samples. In $REFe_2$ systems,[24] the magnetostriction is highly anisotropic with $\lambda_{111} \gg \lambda_{100}$. In $RECo_2$,[25] the sign of λ_{100} is reported to be negative with λ_s values comparable to or greater than that of $REFe_2$. The reduction in magnetostriction values can be due to a change in the easy axis as well as a negative contribution of magnetostriction from Co sublattice.

The magnetization and magnetostriction values of I samples of $Tb_{0.27}Dy_{0.73}Fe_2$ were found to be low compared to the A sample. It was observed by many authors,[26,27] that while preparing $Tb_{0.27}Dy_{0.73}Fe_2$, the composition of Fe has to lie between 1.9 and 1.95 for obtaining a single phase. Any excess amount of Fe beyond 1.95 normally does not yield a single phase when prepared using the zone-melting technique. This was a reoccurring problem and could be

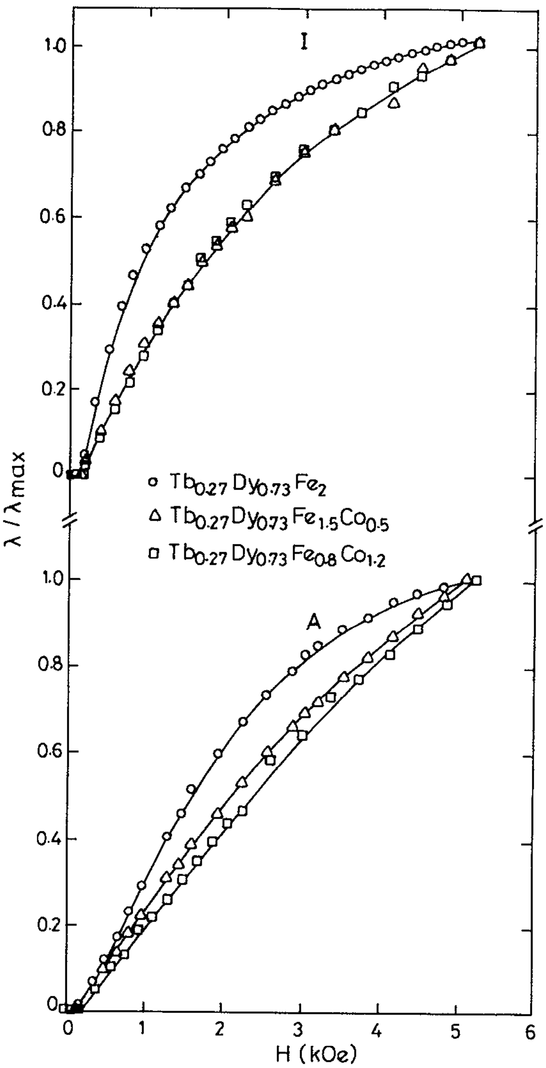

FIG. 7. Normalized magnetostriction as a function of applied bias field for A and I samples of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$.

avoided by preparing a nonstoichiometry compound. Therefore, the actual composition for $x=0$ for I samples was $Tb_{0.27}Dy_{0.73}Fe_{1.93}$.

The normalized magnetostriction (λ/λ_{max}) versus applied field for A and I samples is given in Fig. 7. The aspect ratio of the samples was greater than 5 and therefore the demagnetizing effects could be neglected.[28] The λ values for I samples of $x=0$ and 0.5 tend to increase at lower fields compared to the arc-melted samples. A significant increase in the λ values and a low anisotropy in I samples suggest the formation of a certain degree of grain orientation. This is also reflected by an increase in the k_{33} values of the I samples compared to A samples. The variation of k_{33} as a function of the bias field for different Co concentrations is given in Fig. 6. k_{33} was also found to decrease with increasing Co concentrations in both A and I samples. Branwood *et al.*[29] reported marked improvements in k_{33} accompanying increased grain size for zoned samples. The size and shape of the grains may probably be affected with the increase in Co concentration.

An important feature of λ/λ_{max} versus field curves for I samples (Fig. 7) is that for $x=0$, the field required to saturate is less than that required for $x=0.5$ and 1.2. The $x=0.5$ and 1.2 behave in a similar way and show a strong dependence of λ on the applied field compared to $x=0$. This indicates a change in the easy direction of magnetization from [111] (for $x=0$) even for a Co concentration of $x=0.5$. The strong field dependence and low λ values are typical when the easy axis is [100].[15,16,18] The cubic lattice undergoes a tetragonal distortion when [100] is the easy axis and is small compared to the other distortions. It is also interesting to notice that the lattice constants also show a linear variation beyond the concentration $x=0.5$.

The change in the easy direction of magnetization is also dependent on the magnitude of anisotropy constants K_1 and K_2. The condition for [111] to be the easy axis is that $K_1<0$, and [100] to be the easy axis is $K_1>0$. In $Ho_{0.85}Tb_{0.15}Fe_{2-x}Co_x$, the easy axis remains the same [111] for $x=0$ and 0.5 and changes to [110] for $x=1.2$. Since the changes in the easy axis in these two systems for the same Co concentrations are different, the origin of magnetocrystalline anisotropy which may also be due to an anisotropic unquenched orbital moment[30] in Co may probably be affected by the rare-earth partner. Since the magnetostriction in $Tb_{0.27}Dy_{0.73}Fe_2$ is large compared to that of $Ho_{0.85}Tb_{0.15}Fe_2$, a significant anisotropy due to magnetoelastic interactions would also probably be contributing to the total anisotropy of $Tb_{0.27}Dy_{0.73}Fe_{2-x}Co_x$ system.

The field at which k_{33} maximizes is a measure of the anisotropy of the system. As can be seen from Fig. 6, the field at which the maximum value of k_{33} occurs shifted toward higher-field values with an increase in Co content. This suggests an increase in the anisotropy with increase in Co concentration.

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[1] K. R. Dhilsha and K. V. S. Rama Rao, J. Appl. Phys. **68**, 259 (1990).
[2] K. R. Dhilsha, G. Markandeyulu, and K. V. S. Rama Rao, J. Appl. Phys. **70**, 4450 (1991).
[3] S. Annapoorni, G. Markandeyulu, and K. V. S. Rama Rao, J. Appl. Phys. **65**, 4955 (1989).
[4] P. Westwood, J. S. Abell, and K. C. Pitman, IEEE Trans. Magn. **MAG-24**, 1873 (1988).
[5] A. V. Korolev, N. V. Mushnikov, A. V. Andreyev, and V. S. Gaviko, Phys. Met. Metall. **69**, 88 (1990).
[6] A. R. Piercy and K. N. R. Taylor, J. Phys. C **1**, 1112 (1968).
[7] O. Eriksson, B. Johansson, M. S. S. Brooks, and H. L. Skriver, Phys. Rev. B **40**, 9519 (1989).
[8] A. E. Dwight and C. W. Kimbal, Acta. Cryst. B **30**, 2791 (1974).
[9] A. S. Markosyan, Sov. Phys. Solid State **23**, 965 (1981).
[10] V. V. Aleksandryan, R. Z. Levitin, and A. S. Markosyan, Sov. Phys. Solid State **26**, 1165 (1984).
[11] Hans. R. Kirchmayer and C. A. Poldy, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner and L. Eyring (North-Holland, Amsterdam, 1982), Vol. 2.

[12] W. Steiner, E. Gratz, H. Ortbauer, and H. W. Camen, J. Phys. F **8**, 1525 (1978).
[13] Nguyen Huu Duc, Than Duc Hien, Phuong Mai, and P. E. Brommer, Physica B **172**, 399 (1991).
[14] K. Sato, Y. Isikawa, K. Mori, A. E. Clark, and E. Callen, J. Magn. Magn. Mater. **54**, 875 (1986).
[15] T. Methasiri and I. M. Tang, J. Magn. Magn. Mater. **67**, 190 (1987).
[16] K. Mori, J. Cullen, and A. Clark, IEEE Trans. Magn. **MAG-19**, 1967 (1983).
[17] M. Al-Jiboory and D. G. Lord, IEEE Trans. Magn. **26**, 2583 (1990).
[18] R. Abbundi and A. E. Clark, IEEE Trans. Magn. **MAG-13**, 1519 (1977).
[19] E. Gratz and M. J. Zuckermann, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneider and L. Eyring (North-Holland, Amsterdam, 1982), Vol. 5.
[20] V. I. Ozhogin and V. L. Preobrazhenskii, Sov. Phys. Usp. **31**, 713 (1988).
[21] V. I. Ozhogin and V. L. Preobrazhenskii, J. Magn. Magn. Mater. **90–91**, 191 (1990).
[22] D. Feder and I. Nowik, J. Magn. Magn. Mater. **12**, 149 (1979).
[23] H. T. Savage, R. Abbundi, A. E. Clark, and O. D. McMasters, J. Magn. Magn. Mater. **15–18**, 609 (1980).
[24] A. E. Clark, in *Ferromagnetic Materials*, edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980), Vol. 1.
[25] E. W. Lee and F. Pourarian, Phys. Status Solidi A **34**, 383 (1976).
[26] H. T. Savage, R. Abbundi, and A. E. Clark, J. Appl. Phys. **50**, 1674 (1979).
[27] J. S. Abell and D. G. Lord, J. Less-Common Met. **126**, 107 (1986).
[28] B. D. Cullity, *Introduction to Magnetic Materials* (Addison-Wesley, Reading, MA, 1972).
[29] A. Branwood, R. Dudley, A. L. Janio, and A. R. Piercy, IEE Bull. **60**, 411 (1986).
[30] S. Hirosawa, Ph.D. thesis, Kyoto University, Japan, 1981.