Giant magnetostriction of amorphous Tb_xFe_{1-x} ($0.10 < x < 0.45$) thin films and its correlation with perpendicular anisotropy

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The magnetostrictive behavior of amorphous Tb_xFe_{1-x} ($0.10 < x < 0.45$) thin films was studied by measuring the stress derivative of the perpendicular anisotropy. Large values of the magnetostriction were found, of order 10^{-3}, about the same as the value observed in their polycrystalline counterparts. Within this composition range, $Tb_{45}Fe_{55}$ shows a large magnetostriction with a relatively low anisotropy which makes it a good candidate for application as a magnetoelastic element. After annealing at low temperature (175 °C), relaxation of both perpendicular anisotropy and magnetostriction was observed. However, the relaxation strength was particularly remarkable in the range of low Tb content ($x < 0.30$). The relaxation behavior seems to indicate that local anelastic strain may be the origin of this bond anisotropy for low Tb content while additional structural anisotropy is likely to be responsible for the anisotropy in higher Tb content films.

INTRODUCTION

In 1972 huge room-temperature magnetostriction, of order 2.5×10^{-3}, was reported in the Laves phase $TbFe_2$ by Clark and Belson.[1] Since then the magnetostriction of single crystals and polycrystals of a wide range of rare-earth iron (R-Fe) compounds has been extensively studies.[2] In general these alloys also exhibit strong magnetocrystalline anisotropy which is a significant disadvantage for their application in magnetoelastic devices. Since the amorphous structure averages out local anisotropy, it was expected that amorphous R-Fe alloys might have a large magnetostriction and low magnetic anisotropy if stress free. Nevertheless it was found that amorphous thin films of these alloys generally exhibit a very high magnetic anisotropy with the easy direction perpendicular to the film plane.[3] The existence of this perpendicular easy axis in R-Fe films suggests potential for application as magneto-optic recording materials and various rare-earth transition metals (R-TM) alloys have been used as magneto-optic media. However, the origin of the perpendicular anisotropy remains to be elucidated.

It is well known that the huge values of anisotropy and magnetostriction exhibited by Tb-Fe crystals, polycrystals, and amorphous alloys are due to a combination of the aspherical cystalline field acting on the Tb sites (10^2 K) and the high spin-orbit coupling (10^4 K) characteristic of R atoms with an aspherical distribution of $4f$ charge ($L\neq0$). Spin-orbit coupling, between the spin and the oblate $4f$ electronic cloud of Tb atoms, is the source of the magnetic moment perpendicular to the orbit plane. In crystalline samples the magnetostriction is highly anisotropic and very sensitive to the crystalline structure. For cubic compounds such as $TbFe_2$ and Tb_6Fe_{23}, it is found that $|\lambda_{111}/\lambda_{100}| \gg 1$, where λ_{111} and λ_{100} are the magnetostriction when the sample is magnetized along the 111 or 100 crystalline direction. As a consequence of this remarkable anisotropy of magnetostriction, grain orientation is extremely important and texture in polycrystalline samples is a very relevant property determining the overall magnetostriction. The hexagonal phases $TbFe_3$ and Tb_2Fe_{17} exhibit magnetostriction values that differ from the cubic structures. Polycrystals of Tb_2Fe_{17} exhibit a value of λ of 1.3×10^{-4} in the as-cast state which after annealing becomes -1.4×10^{-5},[4] while the magnetostriction of polycrystalline $TbFe_3$ is 7×10^{-4} which is one half of that of corresponding polycrystalline $TbFe_2$.

Three models have been proposed to account for the origin of the perpendicular anisotropy in a-TbFe films. Gambino and Cuomo suggested that pair ordering was favored during sample growth.[5] Suzuki, Haimovich, and Egami claimed that anelastic distortion of the magnetic atom environment was the cause of the magnetic anisotropy.[6] Finally some recent models of the structure of amorphous TbFe (a-TbFe) films proposed that the amorphous structure consists of nanocrystalline structural

units with size too small to be detected by x-ray experiments. Analysis of pair distribution functions has shown short-range order which might be characteristic of nanocrystals of about 1 nm of bcc Fe with Tb on Fe-lattice sites.[7] More recently Mergel, Heitmann, and Hansen[8] have suggested that during sample growth hexagonal planar units with a preferred axis perpendicular to the film plane are formed.

A number of different terms are commonly used to discuss anisotropy in amorphous alloys. Since the relation between the different terms is not always unambiguous, particularly when discussing magnetic anisotropy, it is worthwhile summarizing the precise relation between the different measures of anisotropy used here and elsewhere. Since the major part of magnetic anisotropy in a-TbFe originates from the interaction of the local electric field with the $4f$ cloud of Tb atoms, its magnitude reflects the asphericity of the Tb atom environment. The $4f$ electronic cloud of Tb has the shape of an oblate ellipsoid with magnetic moment perpendicular to the equatorial plane. When the easy axis of the magnetic moment is perpendicular to the film plane, the minimum electrostatic energy of the $4f$ cloud occurs when its equatorial plane lies in the film plane. Such a configuration should be minimum energy as a result of angular dependence of the distance to the different nearest neighbors as well as any nonuniform charge distribution in the equidistant nearest-neighbor shell. This aspherical distribution of electric charge, which is the only fundamental physical phenomenon that can be inferred from macroscopic magnetic measurements, has been expressed in terms of different local anisotropies. For the sake of clarity four types of anisotropy commonly occurring, anelastic (AA), intrinsic (IA), pair ordering (POA), and magnetoelastic (MEA), are defined according to the microscopic origin of such asphericity.

The interest and relevance of perpendicular anisotropy in a-RFe films has three aspects, (i) the underlying physics, (ii) the tailoring of magneto-optic elements with precisely controlled anisotropy, and (iii) the tailoring of magnetoelastic devices with the lowest possible anisotropy.

Recent extended x-ray-absorption fine-structure (EXAFS) studies on a-TbFe films have shown the existence of a structural anisotropy strongly correlated to the magnetic anisotropy.[9] The analysis of these important observations has led the authors to suggest the existence of POA characterized by different probabilities for unlike bonds for in-plane and out-of-plane directions. In this work we report a systematic experimental study of the magnetostriction and perpendicular anisotropy in a-Tb_xFe_{1-x} $(0.10<x<0.45)$ thin films as deposited and after annealing. In relation to aspect (i) above, it is shown that the measurements of magnetostriction give information on the origin of the magnetic anisotropy. In particular it is shown that features of the anisotropic environment of the magnetic moment can be deduced from the sign of the stress derivative of the anisotropy. In relation to aspect (iii), it has been found that the magnetostriction of a-Tb_xFe_{1-x}, when measured by using magnetoelastic effects, exhibits giant values of the same order of their crystalline counterparts and that a-$Tb_{45}Fe_{55}$ exhibits the most suitable characteristics for application as a magnetoelastic element.

EXPERIMENT

Amorphous Tb_xFe_{1-x} thin films were deposited at room temperature in a UHV dc magnetron sputtering system. Pure argon was introduced into the vacuum chamber which had a base pressure of about 10^{-6} Pa, the Ar pressure was controlled at 1.95 ± 0.01 Pa during sputtering and the sputtering power was 40 W. Composite targets were used with pure Tb plates placed on a pure iron sheet, the composition of a film was controlled by adjusting the area and number of Tb plates. The target-substrate distance was 37 mm and the Ar pressure was chosen to give close to zero stress in the deposited films. The substrate material is Kapton. The thicknesses of the films were about 1.0 ± 0.1 mm.

Magnetization values were determined using an Oxford Instrument 3001 vibrating sample magnetometer. Magnetostriction and anisotropy values were measured using a magnetoelastic technique which is known as the initial susceptibility method. A detailed analysis of the method when it is applied to samples with uniaxial local magnetic symmetry is presented in the Appendix. In particular it is shown that in the experiments reported here the magnetostriction constant obtained by applying this method is the so-called λ_A in Mason notation.[10] According to the method a tensile stress was applied in the direction of the film plane and the change of the initial susceptibility produced by the stress was measured. Magnetostriction and anisotropy values were calculated from the initial susceptibility data through the expression[1]

$$\chi=\frac{\mu_0 M_s^2}{2K-3\lambda_s\sigma}\ , \tag{1}$$

where χ is the initial susceptibility, M_s is the saturation magnetization, K is the anisotropy, λ_s is saturation magnetostriction, and σ is the applied tensile stress. The inverse value of susceptibility should depend linearly on the stress, with positive or negative slope depending on the sign of λ_s. The value of the slope is proportional to λ_s. The anisotropy value was determined by extrapolating to zero stress the experimental values of inverse of the susceptibility and also from the hysteresis loop of the sample measured in the hard direction (the in-plane hysteresis loop), both of which are consistent with each other.

An ac magnetic field with 10 kHz frequency was applied to the sample by means of a solenoid. The magnitude of the field, about 400 Am^{-1}, was controlled to be in the linear range of the in-plane M-H curve. As a-TbFe thin films are magnetically hard, the values of initial susceptibility are quite low and very careful compensation of the sensing coils has to be achieved. For our experimental setup, the compensated signal was only 3 or 4 μv when no sample was in the sensing coil, which is low enough to detect the signal from the sample which is of the order of 20 μv. A sensing coil with about 1000 turns was used to improve the sensitivity. The signal is low due to both the low magnetization value and the small cross-

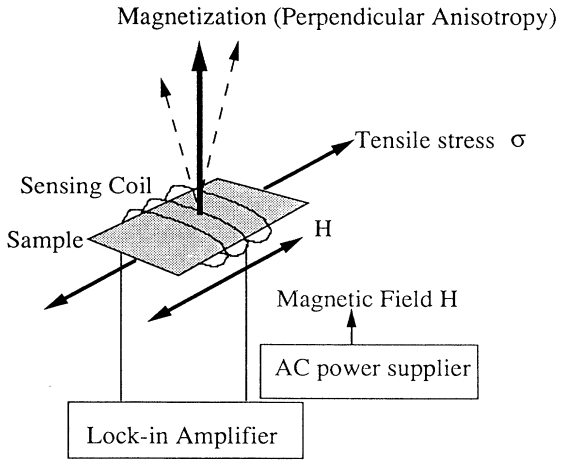

FIG. 1. Experimental setup of the measurement of magnetostriction and anisotropy.

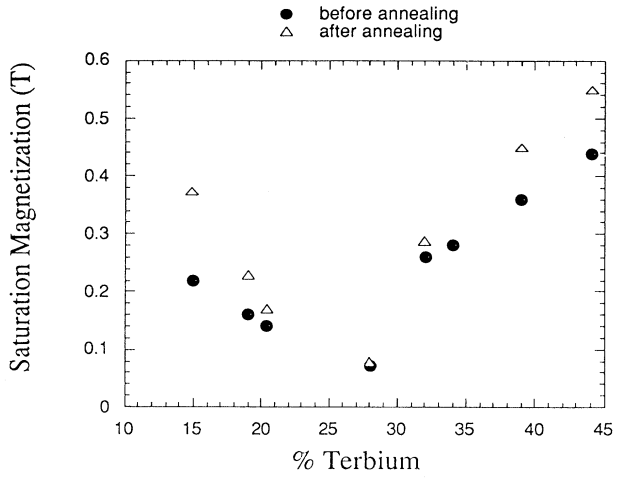

FIG. 2. The dependence of the saturation magnetization of a-Tb_xFe_{1-x} $(0.10<x<0.45)$ thin films on composition before and after annealing. The dots represent the values before annealing and the triangles represent the values after annealing.

sectional area of the sample. The sensing coil and stressing device in which the thin film samples were placed were also carefully designed and arranged to prevent the displacement of the sample during the application of tension, as the signal due to a very small movement of the sample may be of the order of the signal due to the change of the initial susceptibility. Since the method measures the saturation magnetostriction value without saturating the sample, it is particularly suitable for hard magnetic thin films. The experimental setup is shown in Fig. 1.

Annealing was carried out in a vacuum chamber at 10^{-4} Pa and 175±2 °C for about 30 min. The amorphous structure before and after annealing was confirmed by x-ray-diffraction (XRD) measurements.

RESULTS

The saturation magnetization of a-Tb_xFe_{1-x} thin-film samples is plotted as a function of composition in Fig. 2. We can see that at 25% Tb, the magnetization goes to zero. It is known that the saturation magnetization of R-TM alloys is determined by a strong R-TM exchange interaction which gives rise to an antiparallel coupling of the R and TM spins.[11] Figure 2 confirms this and indicates that the composition of 25% Tb is a compression point material. We can also see that the saturation magnetization of a-Tb_xFe_{1-x} $(0.10<x<0.45)$ thin films does not change significantly after low-temperature vacuum annealing, this supports the results from XRD that no crystallization or oxidation has occurred during annealing. The x-ray-diffraction patterns of samples as deposited and annealed at 175°C show that annealing does not give rise to any observable crystallization. The small broadening and shift of the main peak, are not enough to suggest any crystallization or texture development in the samples. The hysteresis loops of the a-$Tb_{45}Fe_{55}$ sample measured at room temperature along both the in-plane and perpendicular directions are illustrated in Fig. 3. We

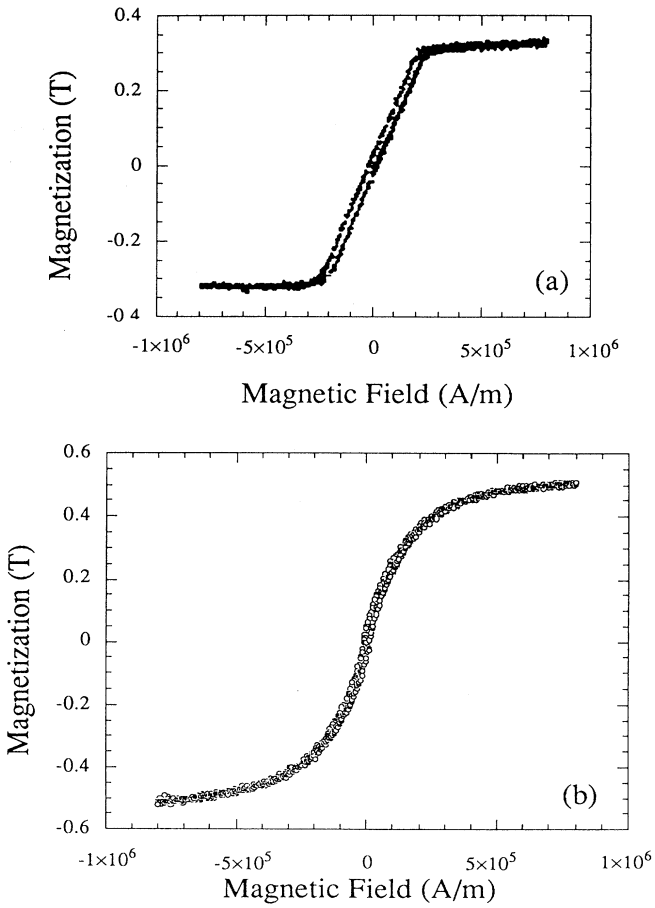

FIG. 3. Hysteresis loop for the a-$Tb_{45}Fe_{55}$ sample (a) applied field perpendicular to the film plane (b) applied field in the film plane.

can see from the shape of the hysteresis loops, after correcting demagnetizing effects which affect the curve shown in Fig. 3(b), that the sample shows strong perpendicular anisotropy. The shapes of the loops in the easy and hard directions indicate that the magnetization process is dominated by domain wall movement or by domain rotation for the field applied in easy or hard directions, respectively.

Figure 4 shows the magnetostriction values as a function of Tb content before and after annealing. The values of magnetostriction of a-Tb_xFe_{1-x} thin films, before annealing, are very similar to the values observed for polycrystalline Tb-Fe compounds. It is to be noted that previously reported magnetostriction values for a-Tb Fe before annealing films were typically one order of magnitude lower[12] than those presented in this work. Before annealing the sputtered thin films exhibit giant magnetostriction values, after annealing the values decrease by about one order for the samples with Tb contents below 35%. As illustrated in Fig. 4, the relaxation of the magnetostriction is much smaller for alloys with higher Tb content. It is worth noting that a very similar decrease of the magnetostriction after annealing was observed in polycrystalline Tb_2Fe_{17} by Clark and Abbundi.[4]

Figure 5 gives the magnetic anisotropy value as a function of composition before and after annealing. Before annealing, anisotropy is strong and perpendicular to the film plane. After annealing, the anisotropy values of all the samples generally decreased. The decrease of the anisotropy can be due to different relaxation processes which must be correlated to the origin of the anisotropy itself. Contributions coming from internal stress relaxation would decrease the possible magnetoelastic component of the total anisotropy. However, relaxation may also be a consequence of the decrease of anelastic anisotropy AA, pair ordering POA, or intrinsic anisotropy IA. As stressed in the discussion the overall results reported here provide insight into the origin of the perpendicular anisotropy in a-TbFe films.

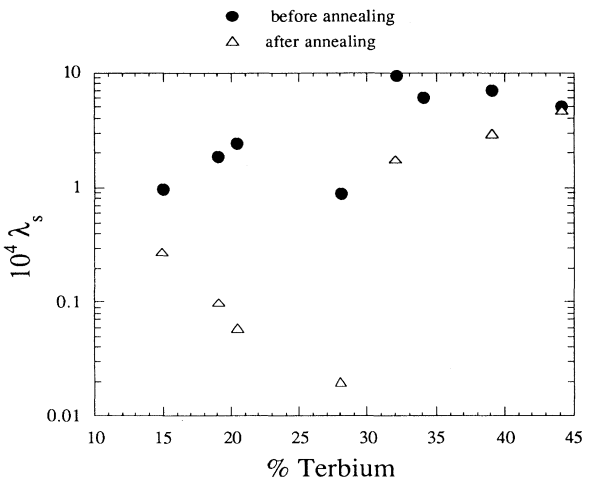

FIG. 4. The magnetostriction of amorphous Tb_xFe_{1-x} $(0.10 < x < 0.45)$ thin films.

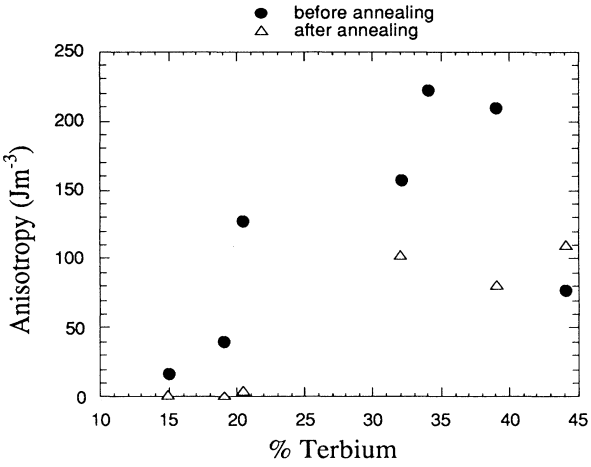

FIG. 5. The value of anisotropy as a function of composition before and after annealing.

DISCUSSION

The experimental results relating to saturation magnetization and magnetic anisotropy reported in Figs. 2 and 5 show good agreement with the data obtained in different laboratories on films deposited by different systems (Ref. 9 and references therein). The x-ray-diffraction patterns indicate that the structure of the samples can be considered as amorphous. The magnetostriction values, shown in Fig. 4, are larger than the values obtained from the direct determination of the strain as a function of the applied field which were reported by Clark[12] and Forester *et al.*[13] (Here, samples with the same thickness were grown by electron-beam coevaporation, but they were also amorphous.) This difference is probably due to the difference of the methods used for measuring magnetostriction. In the strain method, it is very difficult to reach saturation, while in the stress derivative method, the slope of the curve is straightforward related with saturation magnetostriction. In what follows we will discuss the origins of the perpendicular anisotropy and magnetostriction. First it is shown that the experimental results from the relaxation of anisotropy and magnetostriction exhibit clear evidence that different mechanisms are dominant in the low and high Tb content range, mainly anelastic anisotropy (AA) in the low Tb range, and anelastic anisotropy and pair ordering (POA) in the high Tb range.

Note that as the samples with different compositions have different Curie temperatures, the anisotropy values obtained at a constant temperature only provide an approximate measure of the compositional dependence. Nevertheless the anisotropy strength as illustrated in Fig. 5 shows a tendency to increase with Tb content, at least up to $x = 0.35$, indicating that the anisotropy of R-TM materials is dominated by the rare-earth element. Thus it

is reasonable to suppose that the anisotropy has its origin in the Tb atom environment. As stated in the Introduction it can be inferred from the perpendicular easy axis that the oblate electron clouds tend to lie parallel to the film plane. Such an arrangement should be due to an aspherical charge distribution around the Tb sites. Although due to the metallic nature of Tb and Fe, which should mean electrical neutrality, Coehoorn[14] shows that within the metallic model, there can be differences in the electron density at the cell of the atom. Then, the perpendicular anisotropy in these Tb-Fe thin films could be due to a bigger negative charge density along the out-of-plane direction or a smaller density along the in-plane direction. Finally the stresses produced on the film by the substrate might contribute to the magnetic anisotropy via magnetoelastic interactions.

It will be shown that relaxation experiments supply information on the microscopic origin of the anisotropy. As indicated by the results plotted in Figs. 4 and 5, the changes produced by annealing on magnetostriction and anisotropy are strongly correlated and depend on composition. The magnetic anisotropy of samples with low Tb content decreases to a low value after annealing. If this decrease were due only to stress relaxation the magnetostriction should not change significantly. However, the magnetostriction shows a strong relaxation of about one order of magnitude. This decrease of magnetostriction indicates that the decrease of both the anisotropy and magnetostriction is due to a decrease in the asphericity of the Tb atom environment. The fact that a short low-temperature annealing treatment is sufficient to eliminate the anisotropy suggests that for low Tb content the main source contributing to the magnetic anisotropy is AA. Thus, those microscopic processes which remove the local anisotropy are of such low activation energy that they should correspond to small local rearrangement and therefore to processes for which the interchange of atomic positions can be disregarded.

In the range of high Tb content, $x > 0.3$, the same thermal treatment leads to a much smaller decrease of anisotropy and magnetostriction. A reasonable explanation of this behavior can be given by considering that the perpendicular anisotropy generally consists of AA and POA. The POA component cannot be eliminated by the activation of only low-energy processes and therefore would remain after annealing. AA is dominant in the samples with low Tb content, at least with the sputtering conditions reported here, but the POA contribution should increase with Tb content. The remanent anisotropy after annealing could also be a consequence of some IA arising from uniaxial crystalline symmetry around Tb atoms in this composition range. Note that in the presence of IA, POA should not generally follow the $x(1-x)$ dependence and therefore the decrease of anisotropy exhibited by the samples with $x=0.39$ and 0.44 in Fig. 5 suggests that some type of IA similar to that proposed by Mergel, Heitmann, and Hansen[8] probably exists. However, the most important deduction is that when the Tb nearest-neighbor shell contains two types of atoms the magnetic anisotropy acquires some sort of POA independently of the isotropic or uniaxial character of the nearest-neighbor shell geometry. Hence the relaxation experiments show that, in the range of high Tb content, POA contributes to the magnetic susceptibility.

Just as the relaxation results allow us to distinguish between AA and POA (or IA) contributions, the sign of the magnetostriction provides information about the preferred electronic charge density around Tb atoms with respect to the film geometry. As it has been mentioned above, either more negative charge density in the out-of-plane direction or less negative density in the in-plane direction can account for the observed perpendicular anisotropy. However, the expected change of the anisotropy with the stress applied in-plane, is opposite according to whether the more negative electronic distribution is out or in the film plane. Magnetostriction experiments have shown that the perpendicular anisotropy decreases with the in-plane tensile stress, so, it may be concluded that the anisotropy is due to lower electronic density along the in-plane direction, because the elastic elongation of this electronic cloud would decrease the anisotropy. As it has been mentioned in the Introduction, and in good agreement with our conclusions for the high Tb content range, EXAFS studies in Ref. 9 show the existence of a structural anisotropy (POA), specifically, bigger probability of Tb-Fe bonds in the out-of-plane direction. Nevertheless, it is not clear the correlation between this bond distribution and the perpendicular magnetic anisotropy. On the contrary, if it would be valid the point charge approximation,[2,15,16] and taking into account that in this approximation the $4f$ electronic cloud in Tb should be negative and the Fe atoms, when they lose the conduction electrons, should be considered as positive, the perpendicular anisotropy of these films would mean more probability of Tb-Tb bonds out of plane or of Tb-Fe bonds in plane, and finally, the sign of the stress derivative of the anisotropy would indicate the preference of Tb-Fe bonds in plane.

In our discussion MEA has been neglected as a possible source of anisotropy. It is clear that stress relaxation of the amorphous structure cannot account by itself for the remarkable correlation between the relaxation behavior of anisotropy and magnetostriction and in particular for the drastic decrease of magnetostriction observed in the range $x < 0.3$. The stresses present in the sample are either those originating from the elastic interaction between substrate and film, which cannot be removed even for annealing at 300 °C for 1 h,[9] or those produced in the unstrained regions by regions anelastically strained during sample growth. The last component is partially relaxed during anelastic strain relief. However, the experimental magnetostriction results for $x < 0.3$ confirm that the decrease of anisotropy is not a consequence of described elastic stress relaxation, as usually happens for Fe- and Co-based amorphous alloys, but of the relaxation of the anelastic strain in the neighborhood of Tb atoms. The experiments reveal that giant magnetostriction in a-Tb-Fe is a consequence of the high anisotropy. This effect is opposite to that exhibited by Fe- and Co-based amorphous ferromagnets in which the anisotropy is a consequence of the magnetostriction through the coupling to the internal stresses.

CONCLUSIONS

From the experimental results reports here and from the discussion the following conclusions can be drawn.

(i) Amorphous Tb-Fe alloys show giant magnetostriction when measured through magnetoelastic effects. Lower magnetic anisotropy was found for $Tb_{45}Fe_{55}$, which makes it a good candidate for magnetostrictive sensor applications.

(ii) The strong correlation between anisotropy and magnetostriction relaxation seems to indicate that, in contrast to the 3*d*-based amorphous ferromagnets, the anisotropy is the cause of the huge values of a magnetostriction in *a*-TbFe.

(iii) According to the relaxation experiments, two mechanisms leading to structural anisotropy contribute to enhance the perpendicular magnetic anisotropy, one recoverable, AA, and the other more stable, POA. These two mechanisms also are present at low Tb contents but the anelastic one dominates in this range of compositions. The tendency to decrease shown by the anisotropy for $x > 0.35$ suggests that the POA contribution at high Tb content might depend on some IA, as reported in Refs. 7 and 8.

(iv) The metallic nature of these alloys, which implies electrical neutrality, does not allow the use of the point charge approximation, but, the arising of the perpendicular magnetic anisotropy points out to the existence of an aspherical electrical Tb atom environment. According to the studies about the differences is the electron density in the neighborhood of the metallic atoms,[14] and taking into account the sign of the stress derivative of the anisotropy, seems to make clear the existence of a lower negative charge density around Tb atoms in the film plane.

(v) It has been shown that the magnetostriction is a quantitative tool for the elucidation of anisotropy environment at the atomic scale and that it can be a suitable complement to the EXAFS analysis recently carried out and reported in Ref. 9.

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APPENDIX: LOCAL ANISOTROPY AND MAGNETOSTRICTION

The expression (1) is valid for samples with isotropic magnetostriction, uniform anisotropy strength and perfect alignment of easy axis. Let us consider now a sample composed of uniaxial local structures with anisotropy constant K. We assume that this local anisotropy is stronger than the exchange interactions between the magnetic moments of different oriented structural units. This approximation is reasonable in the case of rare-earth magnetic atoms in amorphous structures for which the exchange correlation length is of the order of the correlation length of the orientational fluctuations of the local easy axis. Under these conditions the existence of a macroscopic anisotropy reflects the anisotropic distribution of the orientation of local axes. Let us call z the macroscopic easy axis which in the case of the Tb-Fe amorphous films coincides with the direction perpendicular to the film plane and let y be the in-plane direction along which both stress, σ, and an ac magnetic field, ΔH, much smaller than the anisotropy field, are applied (Fig. 6) (the longitudinal direction). The x axis (transverse direction) lies in plane and is perpendicular to the y axis. Two cases should be distinguished. The first case corresponds to a perfect alignment of the local easy axes respective to the sample axis z and the second case occurs when the orientation of the local easy axes fluctuates around the macroscopic easy axis, z. In what follows we analyze the relation existing between the values of λ and K obtained through the initial susceptibility method and the local anisotropy strength and magnetostriction coefficient's characteristics in the structure of uniaxial symmetry. The magnetoelastic energy referred to the local uniaxial symmetry axes can be written as

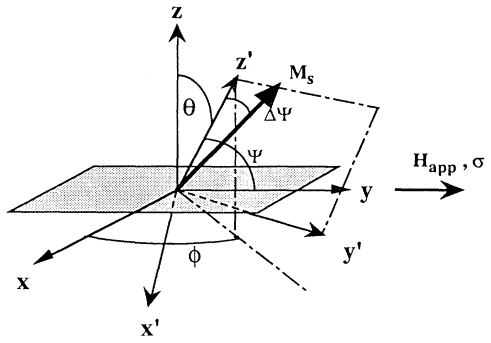

FIG. 6. The configuration of local vectors and axes.

$$\begin{aligned} F = -\sigma\{&\lambda_A[(\alpha_1\beta_1+\alpha_2\beta_2)^2-(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3] \\ &+\lambda_B[(1-\alpha_3^2)(1-\beta_3^2)-(\alpha_1\beta_1+\alpha_2\beta_2)^2] \\ &+\lambda_C[(1-\alpha_3^2)\beta_3^2(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3] \\ &+\lambda_D[(\alpha_1\beta_1+\alpha_2\beta_2)\alpha_3\beta_3]\}\ , \end{aligned} \tag{A1}$$

where σ is the applied tensile stress, α_i and β_i are the components of the magnetization and stress direction, respectively, λ_A, λ_B, λ_C, and λ_D are magnetostriction coefficients.

1. Local uniaxial magnetic symmetry perfectly aligned macroscopically

Assume first that all the structural units are aligned in the sense that the easy axis z' lies along the z axis. For this case $\beta_1=\beta_3=0$ and $\beta_2=1$. The magnetoelastic energy (A1) becomes

$$F=-\sigma\lambda_A(1-\alpha_3^2) \tag{A2}$$

and the macroscopic anisotropy, K^{eff}, and experimental saturation magnetostriction, λ_s, obtained from the inverse of the susceptibility are given by

$$K^{\text{eff}}=K \quad \text{and} \quad \lambda_s=\lambda_A \ . \tag{A3}$$

2. Local uniaxial magnetic symmetry with orientational fluctuation around the macroscopic easy axis

Consider now the case when the easy axes z' axis makes an angle θ with the z axis. By considering the uniaxial character of the local symmetry the local axis y' is considered to be in the plane defined by the macroscopic y axis and the local easy axis z'. Let Ψ be the angle between z' and the direction of the applied field and stress, y ($\cos\Psi=\sin\theta\sin\phi$). If domain wall motion is neglected, the angle $\Delta\Psi$, rotated by the magnetization from the z' axis toward the y axis when a magnetic field ΔH and a stress σ are applied along the y axis can be found through the minimization of the Zeeman energy F_Z, anisotropy energy F_k, and magnetoelastic energy F_m, which can be written as

$$\begin{aligned} &F_Z=-\mu_0 M_s\Delta H\cos(\Psi-\Delta\Psi), \quad F_k=K\sin 2\Delta\Psi \ , \\ &F_m=-\sigma(\lambda_A\sin^2\Psi+\lambda_C\cos^2\Psi)\sin^2\Delta\Psi \\ &\quad -(\sigma/4)(\lambda_A+\lambda_C-\lambda_D)\sin 2\Delta\Psi\sin 2\Psi \ . \end{aligned} \tag{A4}$$

F_m is obtained from Eq. (A1) by taking into account that $\alpha_3=\cos\Delta\Psi$, $\alpha_1=0$, $\beta_3=\cos\Psi$, and $\beta_1=0$. By minimizing the sum of the three energy terms it is found for $\Delta\Psi$ small, as corresponds to the initial susceptibility range of applied fields

$$\Delta\Psi=[h(\sigma)\sin\Psi-\sigma S\cos\Psi\sin\Psi]/[1+h(\sigma)\cos\Psi] \ , \tag{A5}$$

where

$$\begin{aligned} &h(\sigma)=\mu_0 M_S\Delta H/T\ll 1, \\ &T=2K-\sigma(\lambda_A\sin^2\Psi+\lambda_C\cos^2\Psi) \ , \\ &S=(\sigma/2)(\lambda_A+\lambda_C-\lambda_D)\sin 2\Psi \ . \end{aligned} \tag{A6}$$

The variation of the magnetization along the y axis is given by

$$\Delta M_y=M_s\Delta\Psi\sin\Psi \ . \tag{A7}$$

Provided $h(\sigma)\ll 1$, the following approximations can be made:

$$\begin{aligned} &1/[1+h(\sigma)\cos\Psi]=[1-h(\sigma)\cos\Psi] \ , \\ &h(\sigma)=h(0)+\sigma(\lambda_A\sin^2\Psi+\lambda_C\cos^2\Psi)/2K \ . \end{aligned} \tag{A8}$$

By taking into consideration the relations (A8) in Eq. (A5), ΔM_y as given by (A7) becomes the following sum:

$$\begin{aligned} \Delta M_y=M_s\{&A_1 h(0)+[h(0)/2K](A_3\lambda_A+A_4\lambda_C) \\ &-(\sigma/2K)^2(\lambda_A+\lambda_C-\lambda_D)(A_5\lambda_A-A_6\lambda_C) \\ &-A_2[(\sigma/2K)(\lambda_A+\lambda_C-\lambda_D)-h^2(0)]\} \ , \end{aligned} \tag{A9}$$

where

$$\begin{aligned} &A_1=\sin^2\Psi, \quad A_2=\sin^2\Psi\cos\Psi, \quad A_3=\sin^4\Psi \ , \\ &A_4=\sin^2\Psi\cos^2\Psi, \quad A_5=\sin^5\Psi\cos\Psi \ , \\ &A_6=\sin^3\Psi\cos^3\Psi \ . \end{aligned} \tag{A10}$$

Consider now that the sample is composed of an assembly of magnetically uncoupled uniaxial structures whose easy axis z' are symmetrically distributed around the z axis. The average ΔMy can be easily obtained by finding the average values of A_i but referred to the symmetry axes through the relationship,

$$\cos\Psi=\sin\theta\sin\phi \ . \tag{A11}$$

If the angular distribution of z' is given by $L(\theta)$ the average of A_i is given by

$$\langle A_i\rangle=(1/4\pi)L(\theta)A_i(\theta,\phi)\sin\theta\, d\theta\, d\phi \ . \tag{A12}$$

It can be immediately seen that as a consequence of the rotational invariance of L around z

$$\langle A_2\rangle=\langle A_5\rangle=\langle A_6\rangle=0 \ . \tag{A13}$$

Hence the average ΔM_y becomes

$$\langle\Delta M_y\rangle=\mu_0 M_s^2\Delta H/(2K^{\text{eff}}-\lambda_s^{\text{eff}}\sigma) \ , \tag{A14}$$

where

$$K^{\text{eff}}=(K/A_1) \quad \text{and} \quad \lambda_s^{\text{eff}}=(A_3/A_1)\lambda_A+(A_4/A_1)\lambda_C \ . \tag{A15}$$

Figure 7 shows A_1 and $L_A=(A_3/A_1)$ and $L_B=(A_4/A_1)$, for a uniform distribution of the easy axis around the z axis with $0<\theta<\theta_0$, as a function of θ_0.

The anisotropy and magnetostriction given by Eq. (A15) are those which are measured through the inverse susceptibility method. It is easy to see that when all the easy axes are aligned along the z axis, $\Psi=\pi/2$ in all the structural units and it is evident

$$A_1=\langle A_1\rangle=1, \quad A_3=\langle A_3\rangle=1, \quad \text{and} \quad A_4=\langle A_4\rangle=0 \tag{A16}$$

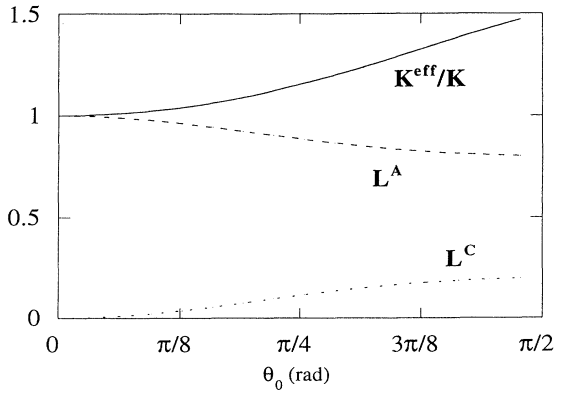

FIG. 7. The dependence of L^c, L^A, and K^{eff}/K.

and therefore the relations Eq. (A3) hold.

Consider now that the easy axes are isotropically distributed, then it can be shown that

$$A_1=\tfrac{1}{3},\quad A_3=\tfrac{4}{15},\quad A_4=\tfrac{1}{15}\ . \tag{A17}$$

The anisotropy and magnetostriction determined by the initial susceptibility method should then be

$$K^{\text{eff}}=3K \quad \text{and} \quad \lambda_s^{\text{eff}}=(\tfrac{4}{5}\lambda_A+(\tfrac{1}{5})\lambda_C\ . \tag{A18}$$

As concerns the experimental method used in this work a quite important conclusion is obtained from the relationships given by Eq. (A18). In our experiments it was generally observed from the shape of the hysteresis loops (Fig. 3) that there is a decrease of the perpendicular anisotropy after annealing. The experimental anisotropy measured from the difference between in-plane and out-of-plane magnetization curves would decrease for either misalignment of easy axes or decrease of the anisotropy constant. However, Eq. (A18) gives a method to distinguish the cause of the anisotropy decrease. If K^{eff} obtained from the initial susceptibility method has increased, the cause of the anisotropy decrease has been a decrease in the degree of alignments of the easy axes, but if K^{eff} has decreased it is because the anisotropy constant has decreased. Therefore the decrease of the experimental K after annealing, plotted in Fig. 5, corresponds to a decrease of asphericity of the Tb environment produced by the annealing.

Finally the evidence of the decrease of the local anisotropy strength, with constant preferential orientation of the easy axis leads to the conclusion that the experimental magnetostriction constant measured in this work is λ_A.

[1]A. E. Clark and H. Belson in *Magnetism and Magnetic Materials—1971, Chicago,* Proceedings of the 17th Annual Conference on Magnetism and Magnetic Materials, edited by D. C. Grahm and J. J. Rhyne, AIP Conf. Proc. No. 5 (AIP, New York, 1972), p. 1498; Phys. Rev. B **5**, 3642 (1972); IEEE Trans. Mag. **MAG-8**, 477 (1972).

[2]A. E. Clark, *Ferromagnetic Materials,* edited by E. P. Wohlfarth (North-Holland, Amsterdam, 1980), Vol. 1, and references therein.

[3]P. Chaudhari, J. J. Cuomo, and R. J. Gambino, IBM J. Res. Rev. **11**, 66 (1973).

[4]R. Abbundi and A. F. Clark, J. Appl. Phys. **49**, 1969 (1978).

[5]R. J. Gambino and J. J. Cuomo, J. Vac. Sci. Technol. **15**, 296 (1978).

[6]Y. Suzuki, J. Haimovich, and T. Egami, Phys. Rev. B **35**, 2162 (1987).

[7]M. Tewes, J. Zweck, and H. Hoffmann, J. Magn. Magn. Mater. **95**, 43 (1991).

[8]D. Mergel, H. Heitmann, and P. Hansen, Phys. Rev. B **47**, 882 (1993); J. M. Riveiro, V. Madurga, and A. Hernando, *ibid.* **39**, 11 950 (1989); G. Suran, H. Ouahame, and R. Zuberek, IEEE Trans. Magn. **20**, 723 (1994).

[9]V. G. Harris, K. D. Aylesworth, W. T. Elam, and N. C. Koon, J. Alloys Compounds **181**, 431 (1992); V. G. Harris, K. D. Aylesworth, B. N. Das, W. T. Elam, and N. C. Koon, Phys. Rev. Lett. **69**, 1939 (1992); V. G. Harris, W. T. Elam, N. C. Koon, and F. Hellman, Phys. Rev. B **49**, 3637 (1994).

[10]*Magnetostriction Theory and Applications of Magnetoelasticity,* edited by E. du Tremolet de Lacheisserie (CRC, Boca Raton, 1993), p. 200.

[11]I. A. Campbell, J. Phys. F **2**, L47 (1972).

[12]A. E. Clark, in *Magnetism and Magnetic Materials—1973, Boston,* Proceedings of the 19th Annual Conference on Magnetism and Magnetic Materials, edited by C. D. Grahm and J. J. Rhyne, AIP Conf. Proc. No. 18 (AIP, New York, 1974), p. 1015.

[13]D. W. Forester, C. Vittoria, J. Schelleng, and P. Lubitz, J. Appl. Phys. **49**, 1966 (1978).

[14]R. Cohehoorn, J. Magn. Magn. Mater. **99**, 55 (1991).

[15]Y. Suzuki, S. Takayama, F. Kirino, and N. Ohta, IEEE Trans. Magn. **MAG-23**, 2275 (1987).

[16]Y. Suzuki and N. Ohta, J. Appl. Phys. **63**, 3633 (1988).