Magnetic anisotropy and magnetostriction in an Fe-rich amorphous film: Analysis of the cantilever method

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It is shown that the deflection of a cantilever consisting of a ferromagnetic film on an elastic substrate, arises from both anelastic and magnetoelastic contributions. With wide generality, the anelastic component of the film deformation, which is frozen in during the sample growth, is larger than its magnetoelastic deformations. The anelastic deformation is also field dependent. This must be taken into account when determining the magnetostriction constant from the deflection induced by applying a magnetic field. On the other hand we show that the experimental deflection, combined with domain observation and magnetization measurements, provides a coherent understanding of anisotropy and magnetostriction. [S0163-1829(98)01714-7]

In the last years a noticeable effort has been experimentally and theoretically carried out in order to control the perpendicular magnetic anisotropy in ferromagnetic films.[1] The first objective was to unravel the origin of such anisotropy. Different models have been proposed to account for the strength and direction of the magnetic anisotropy.[2,3] The origin of the anisotropy has been found to be related to the sample growth procedure and conditions.[4] Certainly, the cause of the anisotropic behavior does not seem to be directly dependent on whether the ferromagnetic film has an amorphous or crystalline structure. In particular rare-earth-transition metals amorphous films were found to exhibit anisotropy with strength as large as their crystalline counterparts.[5]

More recently a great deal of interest has been focused on the measurement and applications of magnetostriction in thin films and multilayers. Among the direct measuring methods, that one based on the determination of the film end deflection when magnetized, the so-called cantilever method, has been widely used and discussed.[6,7]

In this paper we report experimental results obtained, in a structurally soft Fe-rich amorphous alloy, by observing the domain structure and measuring magnetostriction by the cantilever method. The importance of the internal stresses for the anisotropy and magnetostriction of thin films has been emphasized. The analysis of the results points out the more important effects of the stresses frozen in during the sample growth and subsequent cooling: (i) the magnetic film, as obtained, is anelastically deformed into a shell that gives rise to a magnetoelastic energy and thereby an induced magnetic anisotropy and (ii) the magnetostatic energy density of the film also depends on this anelastic deformation. Therefore, a new effective magnetostriction associated with this dependence, which contributes to the experimental deflection should also be present and has to be superimposed on the structural magnetostriction. The influence of deformation on the magnetostatic energy has been previously analyzed when the deformation itself was purely magnetostrictive, and it was called pole effect.[6] The experimental results as well as the analysis reported here indicate that the pole effect associated with the anelastic deformation masks the pole effect originated by the magnetostrictive deformation.

In thin films and multilayers a complex internal stress distribution is generally observed. The origin of the stresses is the film-substrate interaction, through lattice mismatching, difference in thermal expansion coefficients or intrinsic growth factors. However, the complexity of the problem, as thoroughly analyzed by Lacheisserie,[6] is drastically reduced when the magnetic film thickness h_f is much smaller than the substrate thickness h_s. In this case, the in-plane stress tensor components, σ_{xx} and σ_{yy}, which are the only relevant ones, become nearly uniform along the magnetic film thickness. The measurement of the bending provides information about the strength of the internal stresses[8] through the relation that links internal stresses of the magnetic film with the curvature R^{-1} and the deflection D of the whole bimorph that becomes for isotropic in-plane stresses[6]

$$R^{-1}=-6\sigma_{\mathrm{int}}\frac{h_f}{h_s^2}\frac{(1-v_s)}{E_s}$$

and

$$D=L^2/2R=-A\sigma_{\mathrm{int}}\quad\text{where}\quad A=3L^2\frac{h_f}{h_s^2}\frac{(1-v_s)}{E_s},\qquad(1)$$

where E_s and v_s are, respectively, the Young's modulus and Poisson's ratio of the substrate; σ_{int} the value of the internal stresses $\sigma_{xx}=\sigma_{yy}$ in the magnetic film and L the cantilever length. The constant A was introduced for the sake of simplicity.

Structurally soft magnetic amorphous $Fe_{79.8}Cu_{0.25}Nb_{2.38}Si_{11.4}B_{6.23}$ films[9] of 1.3 μm thickness have been produced by ion-beam sputtering technique on glass substrates (thickness 150 μm). The as obtained samples exhibit a clear anelastic curvature. Sample compositions were monitored by wavelength dispersive x-ray analysis. The amorphous character has been tested by x-ray diffraction and transmission-electron microscopy. Magnetic domain observations were carried out by using magneto-optic Kerr effect. Hysteresis loops have been measured with a vibrating-sample magnetometer and a flux-gate magnetometer using an earth field compensated solenoid. Magnetostriction measurements were performed by the cantilever method; therefore

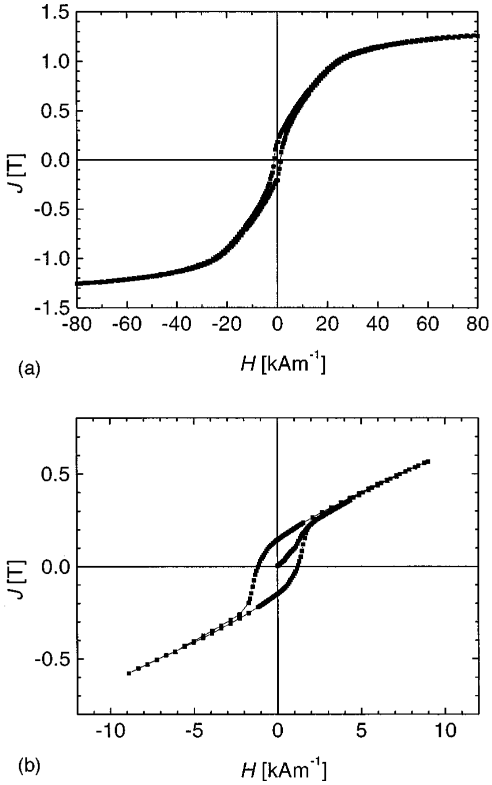

FIG. 1. (a) Magnetization curve corresponding to a magnetic field applied parallel to the film plane measured with a VSM. (b) Detail of the magnetization curve in the region of low fields in which hysteresis is observed measured with a flux-gate magnetometer.

the deflection of the film-substrate bimorph, with dimensions of about 20 mm in length and 2 mm in width, is measured by means of the deflection of the laser beam reflected on the free end of the bimorph. The magnetic field (max. 6.4×10^5 Am^{-1}) can be applied parallel and perpendicular to the beam direction, in both cases within the plane of the thin film.

After demagnetizing the sample, with an in-plane magnetic field, a domain structure formed by domains walls parallel to the previously applied demagnetizing field is observed. When a dc magnetizing field is applied parallel to film plane the domain walls grow, and the contrast gradually disappears. The room-temperature magnetization curve, plotted in Fig. 1, shows the typical behavior corresponding to the case of a magnetic field applied along the direction perpendicular to the easy axis. Therefore, the magnetization of the domains lies in perpendicular direction to the film plane which we call z axis. The hysteresis observed at low fields is due to rotations of the magnetization within the Bloch walls. The remanence appears because all the walls have the same in-plane magnetization component. As the field decreases, from the saturation, the magnetization rotates toward the z axis and domains are formed. Within the 180° Bloch wall the spins rotate in such a way that their in-plane components are parallel to the field. When the field is applied along the opposite direction the spin rotation within the walls changes its sign, process which gives rise to the small hysteresis. The influence of this component is also the origin of the orientation of the domains parallel to the in-plane demagnetizing field. The field required to reach technical saturation is around 2.5×10^4 Am^{-1}, therefore, after considering the value of the spontaneous magnetization at room temperature $\mu_0 M_s = 1.3$ T, the strength of the perpendicular anisotropy K_1 is found to be 1.6×10^4 Jm^{-3}. It is remarkable that the domain width deduced by minimizing the sum of the demagnetizing energy, and the domain-wall energy[10] is of the same order as the domain wall width, about 0.1 μm, when the following parameters are considered, $K_1 = 1.6 \times 10^4$ Jm^{-3}, the out of plane demagnetizing factor $N_z = 1$, and the exchange constant $A_{\mathrm{ex}} = 10^{-11}$ Jm^{-1}. In fact the domain width seems to be close to 1 μm making difficult its observation and measurement. The large number of walls may account for the relatively high value of the reduced remanence which is 0.23. The sample is magnetically isotropic in plane as evidenced by the domain orientation that is always that of the last field applied in plane. The in-plane domain reorientation is governed by the magnetic interaction between the in-plane field and the in-plane component of the magnetization that is within the domain walls.

As the film is an amorphous material with a negligible structural magnetic anisotropy and positive isotropic magnetostriction constant λ, the magnetoelastic anisotropy originated by σ_{int} can act as the main source of magnetic anisotropy different to the film shape. According to the magnetoelastic energy density corresponding to an isotropic sample, the estimated value of the anisotropy constant is $(\frac{3}{2})\lambda\sigma_{\mathrm{int}}$. The measurement of the magnetostriction, as is shown below, leads to $\lambda = 2 \times 10^{-5}$, hence, in order to account for the experimental anisotropy constant, the strength of the elastic stresses due to the anelastic deformation of the film should be of $\sigma_{\mathrm{int}} = -530$ MPa, where the negative character accounts for the direction of the easy axis, which is perpendicular to the stresses. The actual value of σ_{int} can be estimated experimentally through the measurement of the curvature of the sample by using the expression (1). Certainly the expressions given in Eq. (1) should be corrected to include the magnetostrictive strain produced by the magnetization oriented along the z axis, which yields a supplementary in-plane stress contribution of $\sigma_{\mathrm{me}} = -\varepsilon_{\mathrm{me}}[E_f/(1+\nu_f)] = -(\lambda_s/2)(E_f/1+\nu_f) = -B/3$; B, E_f, and ν_f denoting the magnetoelastic coupling coefficient, Young's modulus, and Poisson's ratio of the film, respectively. With this correction in the expression of R^{-1} and D, the resultant spontaneous deflection D_0 becomes[11]

$$D_0 = -A\left(\sigma_{\mathrm{int}} + \frac{B}{3}\right). \tag{2}$$

In our experiments the following values hold,[12] $L = 2$ cm, $\nu_s = 0.2$, $E_s = 7 \times 10^4$ MPa, $E_f = 2.5 \times 10^5$ MPa and therefore, $B = 6$ MPa and A as defined in Eq. (1) takes a value of about 10^{-12} mPa^{-1}. Notice that the initial deflection, in the absence of any applied field, consists of two terms, an anelastic one proportional to the internal stress and a magnetostrictive contribution proportional to the coupling coefficient. For $\sigma_{\mathrm{int}} = 530$ MPa, which is the value expected by assuming the internal stresses to be the origin of the experimental anisotropy, R can be estimated directly from Eq. (1), neglecting B, and leads to a value of 0.5 m. This is in perfect agreement with the experimental determination of equally 0.5

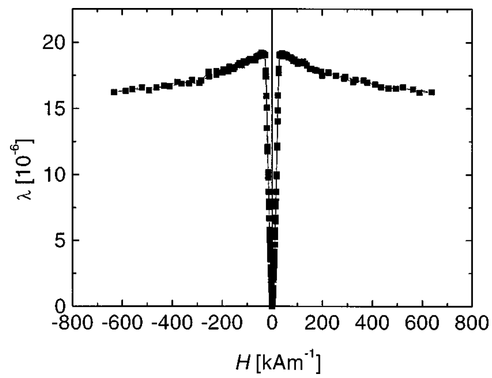

FIG. 2. The experimental deflection $D_{\parallel}-D_0$ plotted as a function of the applied field Hy along the cantilever direction.

m. Therefore it can be concluded that the magnetic anisotropy is governed by the magnetoelastic energy induced by the elastic stresses exerted by the substrate on the magnetic film.

Let us analyze the influence of the initial deformation of the bimorph in the field dependence of the deflections measured by the cantilever method. As a first approximation the field dependence of the anelastic deformation is disregarded. In particular when the sample saturates along the cantilever direction, y direction, the deflection of the cantilever becomes

$$D_{\parallel}=-A(\sigma_{\text{int}}-2B\alpha_{\parallel}) \tag{3}$$

with

$$\alpha_{\parallel}=\frac{(1-\nu_f\nu_s)-\frac{1}{2}(\nu_f-\nu_s)}{3(1-\nu_f)(1-\nu_s)}.$$

The coefficient $\alpha_{\parallel}$ accounts for the anisotropic character of the magnetostrictive strains when the magnetization lies in y direction[6] and replaces the constant $\frac{1}{3}$ for magnetization in z direction.

When saturated in plane but perpendicular to the cantilever direction, x direction, the cantilever deflection is

$$D_{\perp}=-A(\sigma_{\text{int}}+B\alpha_{\perp}) \tag{4}$$

with

$$\alpha_{\perp}=\frac{(1-\nu_f\nu_s)-2(\nu_f-\nu_s)}{3(1-\nu_f)(1-\nu_s)}.$$

The magnetostriction can be obtained through the difference $D_0-D_{\parallel}$, after saturating along y direction, or through $D_{\parallel}-D_{\perp}$, after saturating along x direction, the latter leading to the known formula elaborated by Trémolet.[6] However, it is important to know that the three terms (2), (3), and (4) contain an anelastic term 90 times larger than the magnetoelastic contributions, and this anelastic term also depends on the applied field, as illustrated by the experimental results in the following.

Figure 2 shows the experimental cantilever deflection $D_{\parallel}-D_0$ undergone by the sample as a function of the applied field along the y direction. The deflection does not saturate, but, after a maximum, decreases continuously to fields much higher than that in which saturation is observed in the magnetization curve. This anomaly can be well understood as a consequence of the combination of film geometry and its initial bending D_0, which leads to the field dependence of the effective elastic constant E_s.

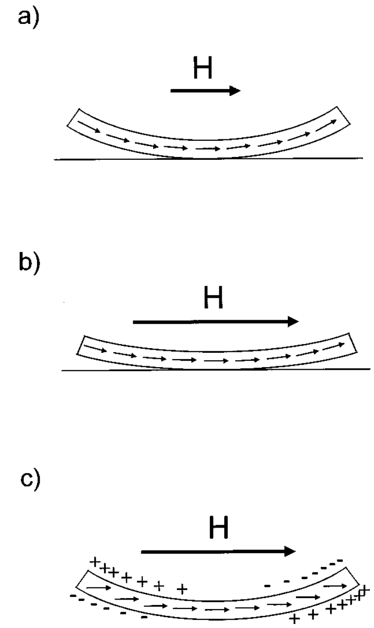

FIG. 3. Scheme of the origin of the pole effect. As indicated in the text, the in plane field is higher than the effective field corresponding to the initial perpendicular anisotropy. The case (b) corresponds to a shape anisotropy energy much larger than the elastic energy. This assumption was made to obtain relation (5). The case (c) corresponds to the case of comparable demagnetizing and elastic energies and has been disregarded in the present calculations.

Consider that once the magnetization saturates under the action of the in-plane field it is not parallel to the field but contained in the deformed sample to eliminate poles and decrease the demagnetizing energy, according to the scheme shown in Fig. 3. As the field rises, the magnetization tends to rotate toward the field direction so exerting an effective bending torque opposite to that due to the internal stresses. This rearrangement of both magnetization and sample deformation are not observed by the pickup coils which measures the magnetization, however, it turns out to be perfectly detected by the extremely more sensitive cantilever. The dependence of the magnetostatic energy with the elastic strain is known as pole effect[13] and produces an effective increase of E_s that has been calculated in Ref. 6 as

$$\Delta E_s=3\mu_0 M_s H L^2(h_f/h_s^3), \tag{5}$$

which in our case becomes $\Delta E_s=600H$ $[\text{Am}^{-1}]$ Pa. This magnetic field dependence of the elastic constant reduces the total deflection of the cantilever expressed in relation (3) where instead of E_s it should be written $E_s+\Delta E_s$; and augments the deflection expressed in Eq. (4) where E_s has to be replaced by $E_s-v_s\Delta E_s$. After introducing this substitution, the experimental deflection obtained by applying a saturating field H_y along the cantilever direction is expected to be

$$D_{\parallel}-D_0=AB(2\alpha_{\parallel}+\tfrac{1}{3})-D_0\Delta E_s/E_s, \tag{6}$$

or, when the saturating field is applied along the x direction, the experimental deflection along the cantilever direction is

$$D_{\perp}-D_0=AB(\tfrac{1}{3}-\alpha_{\perp})+\nu_s D_0 \Delta E_s/E_s. \tag{7}$$

It was considered that $\Delta E_s/E_s \ll 1$; relation that, for the case of our sample and according to Eq. (5), holds well for fields below 10^6 Am^{-1}.

For applied fields larger than the saturating field, 2.5×10^4 Am^{-1}, which also produce the magnetostrictive strain saturation, relation (6) describes the experimental curve shown in Fig. 2. The second term in Eq. (6) is responsible for the monotonic decrease of the experimental deflection and can be estimated from the value of D_0 ($=4\times 10^{-4}$ m) as a function of the field, which yields

$$D_0 \Delta E_s/E_s = 3\times 10^{-12} H_y (\mathrm{Am}^{-1}). \tag{8}$$

The curve shown in Fig. 2 is in good agreement with the one predicted by relation (6). The maximum component of the magnetostrictive deflection, which saturates at $H_y=2.5\times 10^4$ Am^{-1}, corresponds to $D_{\parallel}-D_0$. According to Eq. (6), the maximum magnetostrictive deflection is of the order of $AB=10^{-6}$ m, as experimentally found (see Fig. 2) whereas according to Eq. (8) the deflection due to the pole effect takes this same value for fields of the order of 10^6 Am^{-1}.

In the present case, the experimental magnetostriction constant is underestimated by the pole effect, but for $H_y=3\times 10^4$ Am^{-1} this last contribution gives rise to a deflection of 10^{-7} m, hence, one order of magnitude smaller than the magnetostrictive deflection. Therefore, the experimental value $\lambda=2\times 10^{-5}$, obtained from the experimental $D_{\parallel}-D_0$ at 2.5×10^4 Am^{-1}, through relation (6), can be considered a good value within 10%. From the experimental point of view, the resolution to distinguish between the two contributions, anelastic and magnetoelastic, is larger as magnetically softer is the material and lower is the slope of ΔE_s with respect to H.

As the pole effect produces an effective increase of E_s, it always acts to decrease the total bending of the films. Notice that in the present case the magnetoelastic coupling coefficient and the initial deformation of the sample D_0 are positive. As shown above, the anelastic strains producing this initial bending are presumed to induce the magnetic anisotropy in our samples. The case of a pole effect decreasing the measured deflection at high fields is therefore characteristic for a strain induced perpendicular anisotropy in as-sputtered thin films.

However, when the anisotropy has not any relation to the initial deformation of the magnetic film, the magnetostrictive deflection induced by the applied field can be either of the same or of different sign than that one due to pole effect. In particular, in ideal initially undeformed samples both contributions should be opposite in sign since in this circumstance the magnetostrictive deflection is the only one to be eliminated by the pole effect. It is concluded that the measurement of the deflection up to fields high enough to make evident the pole effect can elucidate the correlation between the internal stresses and the magnetic anisotropy.

In summary, our experiments have evidenced that the elastic stresses exerted by the substrate on the magnetic film, due to the anelastic deformation, are the cause of the perpendicular anisotropy observed in the amorphous film studied here. The magnetic field produces two types of modifications in the initial deformation. First, those associated with the magnetostriction and second, those originated from the initial deformation itself and which tend to eliminate it. Care should be taken when measuring magnetostriction constants, since the pole effect is field dependent. On the other hand, the relation between the signs of both components supplies information on the origin of the magnetic anisotropy.

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[10] The calculation of the domain width can be seen in *Introduction to Magnetic Materials*, edited by B. D. Cullity (Addison-Wesley, Reading, MA, 1972), p. 301.

[11] This expression can be easily obtained by considering the magnetoelastic energy and the initial deflection. The magnetoelastic energy is that given in Ref. 6. However, in this reference the initial deformation was not considered.

[12] The values corresponding to the substrate were obtained from the supplier, and the Young modulus of the film is that measured for this composition in ribbon shape.

[13] Notice that in Ref. 6 this effect was calculated for the limit of very large shape anisotropy. Here the same approximation was considered. Therefore we disregarded the possibility shown in Fig. 3 as case c.