Stress and magnetoelastic properties control of amorphous Fe₈₀B₂₀ thin films during sputtering deposition

I. Fernández-Martínez,^{a)} J. L. Costa-Krämer, and F. Briones

Instituto de Microelectrónica de Madrid (CNM-CSIC), Isaac Newton 8 PTM, 28760 Tres Cantos, Madrid, Spain

(Received 14 November 2007; accepted 23 March 2008; published online 3 June 2008)

In situ stress measurements during sputtering deposition of amorphous Fe₈₀B₂₀ films are used to control their stress and magnetoelastic properties. The substrate curvature induced by the deposited film is measured optically during growth and quantitatively related to the deposition induced accumulated stress. The resulting magnetic properties are later correlated with the measured stress for a wide range of sputtering pressures $[(2-25) \times 10^{-3}$ mbar]. A significant tensile stress develops at the film-substrate interface during the early growth stages (initial 2–3 nm). At a critical thickness, a transition is observed from tensile to compressive stress, which is associated with amorphous island coalescence. By further increasing the thickness, a compressive stress follows, which is related to the local distortion induced by the ion peening effect. The Monte Carlo simulations of the sputtering process describe quantitatively the experimental results as a function of the Ar pressure and target bias voltage. © 2008 American Institute of Physics. [DOI: 10.1063/1.2931043]

I. INTRODUCTION

Amorphous magnetic alloys have been extendedly studied over the past decades.^{1,2} The materials are usually fabricated either by rapid quenching from the molten state or in thin film form by sputtering. They can exhibit unique mechanical properties,³ improved corrosion resistance,^{4,5} very soft magnetic response,⁶ and high magnetic moment and Curie temperatures. From a technological point of view, they are the best-known materials for stress sensing, having magnetomechanical coupling factors k^2 (ratio of energy transfer between the mechanical and magnetic subsystems) approaching unity.^{7,8}

Stress control in thin films during and after growth is of great importance both for the lifetime and performance in technological applications, in particular, for micro- and nano-electro-mechanical systems (MEMS and NEMS). Besides the technological interest, *in situ* measurements of mechanical stress are especially useful in obtaining additional information about nucleation and growth mechanisms.⁹ Amorphous metals, lacking long-range atomic order, might provide a good reference to study and discern different contributions to stress during growth in crystalline materials. The main parameter that characterizes the morphology and, consequently, the deposition stress of thin films prepared by sputtering at RT is the energy-effective temperature of the deposited atoms.¹⁰ For a given initial energy, the effective temperature depends on the energy lost by the sputtered atoms at collisions with the sputtering gas before they arrive at the substrate. In a triode sputtering system, the energy of the neutral atoms arriving at the substrate can then be varied by changing either the Ar working pressure or the target potential independently. In this study, the Ar working pressure is chosen as the parameter that controls the energy of the arriving sputtered atoms while the target potential is fixed.

Large tensile stresses often develop during growth of thin amorphous films.¹¹ It has been suggested that the large tensile stresses measured in amorphous binary films are caused by a growth related columnar structure, which develops when the surface mobility of adatoms is limited.¹² This kinetic restriction is the one that prevents crystallization of the amorphous alloys under standard deposition conditions. However, crystallization has been observed for high target potentials and long deposition times.¹³

This work presents *in situ* stress measurements during triode sputtering deposition of amorphous Fe₈₀B₂₀ films. The time evolution of the measured stress is used to discern the growth mechanism: an initial island formation and coalescence followed by a continuous thin film growth. For the continuous films (30–70 nm thick), the total accumulated stress is measured. The film final magnetic anisotropy is affected by two different contributions: a field induced anisotropy due to the presence of the field used to confine the plasma and a magnetoelastic anisotropy caused by the total accumulated stress. The final film magnetic properties are correlated with this stress for a wide range of sputtering pressures $[(2-25) \times 10^{-3}$ mbar]. A fine control of the final anisotropy direction due to the competition of field and stress induced anisotropies is achieved in this range of sputtering pressures. A fine control of stresses, together with a tunable anisotropy, high magnetostriction, excellent adhesion, and stability, makes these films ideal for magnetostrictive-based NEMS actuators.¹⁴

II. EXPERIMENTAL

Amorphous Fe₈₀B₂₀ films were deposited on glass substrates at ambient temperature in a triode sputtering system by using a target with a nominal composition of Fe₈₀B₂₀ (at. %). The deposition was carried out for a fixed bias voltage of –2 kV by using ultrahigh purity Ar in the range of $(2-25) \times 10^{-3}$ mbar. The target to substrate distance was 8

^{a)}Electronic mail: ivan@imm.cnm.csic.es.

FIG. 1. Schematic diagram of the experimental setup dedicated to the *in situ* stress measurement during triode sputtering deposition. Notice the filament, the anode and target, the laser beam, and the position-sensitive detector. The laser incidence plane together with the geometry of the substrate/cantilever is rotated 90° in the picture for clarity reasons. Notice that the short edge of the cantilever is parallel to the plasma confining field.

cm and the base pressure was in the low 10^{-8} mbar range. The angle of deposition was chosen at 45° with respect to normal incidence. A plasma confining magnetic field of a few oersteds was applied parallel to the film plane during deposition. The obtained $\text{Fe}_{80}\text{B}_{20}$ films were found to be x-ray amorphous. The deposition time for all of them was 300 s, producing thicknesses that varied from 30 to 70 nm depending on the Ar pressure. The saturation magnetostriction constant of the films was measured to be $\lambda_s = 26 \times 10^{-6}$, which is in agreement with published data.¹³

The development of film stress during deposition was monitored by using an optical beam deflection method on substrates cut in the shape of a cantilever. Changes in the substrate curvature can be directly related to the accumulated stress σ by the well-known Stoney equation revised for biaxial stress:¹⁵

$$\sigma = \frac{1}{6} \frac{E_s h_s^2}{(1 - \nu_s) h_f} \left[\frac{1}{r} - \frac{1}{r_0} \right], \quad (1)$$

where $E_s/(1 - \nu_s)$ is the biaxial elastic modulus of the substrate, h_s and h_f are the thicknesses of the substrate and film, respectively, and r_0 and r denote the curvature radii before and after the film deposition, respectively. The substrate curvature was determined by measuring the free end bending by using

$$\left(\frac{1}{r} - \frac{1}{r_0} \right) = \frac{1}{2} \frac{\Delta x}{lL}, \quad (2)$$

where Δx , l , and L are the laser deflection measured in the photodiode, the length of the cantilever/substrate, and the distance between the substrate and the photodiode respectively, as schematically shown in Fig. 1. The initial substrate curvature r_0 was negligible. The glass substrate cantilevers were 4 mm wide, 16 mm long, and 0.125 mm thick. A scheme of the deposition chamber geometry and the *in situ*

optical measurement is shown in Fig. 1. For clarity reasons, the optical detection system has been rotated 90° in the figure with respect to its real orientation.

The hysteresis loops were measured by using a conventional transverse magneto-optical Kerr effect setup at a $\lambda = 634$ nm wavelength of a HeNe laser. It is well known that in the transversal Kerr configuration, polarized light reflectivity changes are linear with the magnetization. For $\text{Fe}_{80}\text{B}_{20}$ films, reflectivity changes at magnetic saturation are of the order of $\Delta R/R = 0.5\% - 1\%$. A related magneto-optical torque technique¹⁶ has been used to characterize the magnetic anisotropy of the films.

III. RESULTS AND DISCUSSION

A. *In situ* stress measurements

The evolution of the accumulated stress for $\text{Fe}_{80}\text{B}_{20}$ films as a function of the film thickness for different Ar gas pressures and at a constant target potential of -2 kV is shown in Fig. 2(a). Three regimes are clearly distinguished in Fig. 2(a): I–III.

The first one is a nearly instantaneous rise in an apparently compressive stress when the target potential is switched on [regime I in Fig. 2(a)]. This effect is reversible, which disappears when the target potential is switched off. This is shown in Fig. 2(b), where the stress evolution is plotted as a function of the deposition time. At 300 s, which is the deposition time for all of the samples, an abrupt apparent tensile relaxation is observed when the target potential is switched off. The magnitude is of the order of the initial apparent compressive stress (regime I). A similar effect that develops during the Volmer–Weber growth of polycrystalline low melting point films¹⁷ has been associated with adatom-surface interactions. However, in the case of amorphous metallic films, surface diffusion is strongly limited and no large adatom population is expected. Therefore, in our case, this effect should rather originate on an effective surface heating due to thermalization of the impinging atom kinetic energy and chemical bond formation. The difference between the effective kinetic surface temperature and the bulk gives rise to a substrate bending equivalent to a surface stress of compressive character. The amplitude of this effect is proportional to the instantaneous deposited power on the substrate. Indeed, it depends strongly on the target bias potential, deposition rate, and Ar gas pressure and disappears reversibly when growth is interrupted [Fig. 2(b)]. It does not contribute to the total accumulated stress.

Therefore, the accumulated stress during the initial few nanometer growth (regime II) is clearly tensile, which corresponds to the interface stress increase that accompanies the nucleation of very small islands strongly interacting with the glass substrate surface. The measured tensile stress is found to be lower for higher Ar working pressures, as shown in Fig. 2(a), i.e., for lower energy of the arriving atoms and, correspondingly, a lower surface diffusion. This tensile stress reaches a saturation value of about 18×10^{-3} mbar. Taking into account that amorphous films usually exhibit a columnar structure due to self-shadowing effects, a lower density $\text{Fe}_{80}\text{B}_{20}$ films grown at higher Ar pressures is expected,¹¹

FIG. 2. (Color online) (a) Accumulated stress evolution as a function of the film thickness for four different pressures during the early stages of deposition. Three stress regimes are clearly distinguished. Regime I: initial instantaneous compression; thermal induced curvature. Regime II: precoalescence tensile regime related to the interaction between the discontinuous film and substrate. Regime III: compressive stress due to ion peening. (b) Accumulated stress evolution as a function of the deposition time during growth interruption. The initial instantaneous compression is relaxed reversibly at a 300 s deposition time. (c) Dependence of the critical thickness on the deposition pressure. The dots are the measurements of the minimum of the total accumulated stress curve, i.e., the points at which the instantaneous stress changes sign.

with a higher density of voids, defects, etc. The interface stress between the glass substrate and deposited atoms seems to be the dominant source of the large component of tensile stress measured because it depends on the interaction between the growing film and the substrate. Moreover, the maximum tensile value shifts to higher nominal thickness with increasing sputtering pressure. Confirming the above arguments, it has also been found experimentally that the

stress sign and magnitude in regime II depend strongly on the type of substrate, changing as well if a buffer layer is present.

A critical thickness can be defined between this initial tensile (II) and the following compressive regime (III). This would be the thickness value at which the film becomes continuous and coalescence or building of a columnar structure takes over.⁹ Figure 2(c) shows the dependence of this critical thickness on the Ar pressure, i.e., the minimum of the accumulated stress versus the thickness curve. The critical thickness seems to increase in steps of about 0.7 nm for increasing Ar pressure.

There is general agreement on the origin of this compressive stress being ion or energetic neutral atoms peening.¹⁸ It accumulates proportionally with the film thickness and is strictly dependent on the material that is being deposited¹⁹ and obviously on the kinetic energy of the impinging atoms on the growing film. This energy is strongly dependent on the mass of the atoms, the target potential, and the thermalization capacity of the sputtering gas filling the space between the target and the substrate.

We have calculated the energy distributions of the sputtered Fe and B atoms ejected from the target and the residual energy distribution as a function of the distance traveled through the Ar sputtering gas for different working pressures by using a Monte Carlo simulation (SRIM 2006-02 by Ziegler), as in earlier publications.²⁰ From these results, we can extract the thermalization distance. This distance is strongly dependent on the initial energy and the chamber pressure. For our conditions (initial energy of Ar⁺=2 keV, cathode-substrate distance=8 cm), iron and boron atoms reach the substrate thermalized at 1.5 × 10⁻² mbar, while Ar⁺ atoms do not arrive thermalized at the substrate in our pressure range [(2–25) × 10⁻³ mbar]. The thermalization distance of the Ar⁺ ions for a working pressure of 1.5 × 10⁻² mbar is 15 cm.

The onset of compressive stress is attributed to the bombardment of sputtered atoms together with the energetic backreflected inert gas atoms striking the film with energies of hundreds of eV ions during deposition (ion peening). A slightly positive potential of the Ar⁺ plasma (of the order of 35 V relative to the ground in our system) probably contributes substantially to this ion peening effect.

The information about the distortion created by energetic arriving atoms can be extracted from the linear part of the substrate curvature evolution (regime III). With a target potential of -2 kV, even when the iron and boron atoms are thermalized (above 1.5 × 10⁻² mbar), the ion peening effect still appears due to Ar. Although these Ar energetic particles arrive at the substrate attenuated by gas scattering, they are still not at thermal equilibrium with the substrate. The total accumulated stress in the films after a 5 min deposition time at -2 kV for different Ar working pressures is shown in Fig. 3. Under these conditions, note that the residual stress cannot be compensated, varying only the pressure at a fixed target potential of -2 kV. To obtain near-zero stress amorphous Fe₈₀B₂₀ films, lower target potentials combined with lower Ar⁺ working pressures are required.

FIG. 3. Total internal accumulated stress as a function of the working pressure for a fixed target potential of -2 kV. This accumulated stress is compressive for all of the ranges due to the ion peening effect.

B. Magnetic properties

The magnetic properties of a metallic glass, such as magnetic anisotropy, coercivity, permeability, etc., are extremely sensitive to the structure and can be tailored within certain ranges continuously maintaining the amorphous structure. Anisotropies can be induced during growth or post-growth by annealing below the crystallization temperature under an external directional agent such as field, stress, or current.

All of the amorphous $\text{Fe}_{80}\text{B}_{20}$ films grown on the glass cantilevers exhibit a uniaxial anisotropy (Fig. 4). The films were grown under a small external applied magnetic field (applied parallel to the shorter edge of the glass substrate) that is used to confine the plasma. For lower Ar pressures, the induced magnetic uniaxial anisotropy points in the direction of the plasma confinement field (see Fig. 1), i.e., parallel to the short edge of the glass substrate/cantilever. This uniaxial anisotropy results from local atomic-pair ordering in the amorphous matrix.²¹

At pressures above 8×10^{-3} mbar, the anisotropy direction starts deviating from the plasma confining field direction and turns perpendicular at about 14×10^{-3} mbar, as shown in Fig. 5 at a -2 kV target potential. The field induced mag-

FIG. 4. (Color online) Angular dependence of the remanence for a $\text{Fe}_{80}\text{B}_{20}$ film deposited at -2 kV and 8×10^{-3} mbar of Ar working pressure. The insets show the hard and easy axes loops. DR/R are the normalized reflectivity changes as a function of the applied magnetic field.

FIG. 5. Anisotropy angle direction as a function of the Ar working pressure for a fixed target potential of -2 kV.

netic anisotropy is then dominant in the case of lower Ar working pressures, where the effective temperature of the deposited atoms is higher. As the pressure increases, stress induced anisotropy overcomes the effect of the field induced anisotropy. This continuous easy axis rotation is then due to the fact that compressive stress is not isotropic in the substrate/cantilever.

The cantilever shape and placement render the long dimension edge free to rotate and the short dimension edge constrained. As calculated in earlier works,²² if the long dimension edge of the substrate is four times larger than that of the short dimension, the effect of clamping is to restrict the bending to a purely one dimensional deformation along the long edge. Accordingly, as growth develops, there is a stress imbalance, and thus, the net stress is larger in magnitude along the short direction, which in case of a net accumulated compressive stress and a positive saturation magnetostriction constant results in an anisotropy along the long axis direction.

As expected from the above considerations, the anisotropy rotation was not found for thicker substrates/cantilevers in which the deformation that causes stress relaxation along the long axis is not possible.

The value of the anisotropy constant K_u has been measured by using a magneto-optical torque technique¹⁶ and the results are shown in Fig. 6. The anisotropy constant has the same behavior as the accumulated stress up to the compensation pressure ($\sim 15 \times 10^{-3}$ mbar), where the field and magnetoelastic anisotropies compensate each other. Figure 7 shows how the values of the coercive field H_c change with

FIG. 6. Anisotropy constant K_u as a function of the Ar working pressure measured by using the magneto-optical torque technique.

FIG. 7. Coercive field H_c of the easy axis loop as a function of the Ar working pressure for a fixed target potential of -2 kV.

sputtering working pressure at a -2 kV target potential. Notice a low ~ 4 Oe coercivity at pressures above 20×10^{-3} mbar. In a high magnetostrictive material, usually residual inhomogeneous stress inherent to the fabrication process hinders domain wall motion.²³ The fact that the coercivity is not a minimum at the position where the anisotropy is minimum (see Fig. 6) points to a more homogeneous material in the case of larger deposition pressures and, consequently, a smaller domain wall propagation field.

A parameter of importance for applications, such as magnetostrictive actuators, is the rate of change in magnetostriction with field, $d\lambda/dH$, which is termed as D . Ideally, it should be as large as possible and occur at the lowest field strengths possible. The largest reported values of D are of the order of 10^{-5} Oe $^{-1}$ for Tb_xDy_{1-x}Fe_y films.²⁴ The value of our films is 4×10^{-6} Oe $^{-1}$ for the lower value of H_k , which corresponds to a 1.5×10^{-2} mbar working pressure. The high value of D for Fe₈₀B₂₀ thin films is a remarkable technical aspect, which results from the combination of a very low structural anisotropy and a large magnetostriction.

Finally, we have a few words about the oblique incidence anisotropy. Our substrate/cantilever is placed at 45° with respect to the incoming flux due to geometrical constraints of our deposition chamber and the optical detection system. The measurements at a -2 kV target potential show that there is no anisotropy due to the oblique incidence of up to a 60° angle for any Ar pressure. Measurements of the anisotropy induced as a function of the substrate/cantilever angle with respect to the incoming flux point to an oblique incidence anisotropy that starts to develop above -1 kV. This is in agreement with all of the above arguments about a limited surface diffusion at this target bias.

The magnetic anisotropy of Fe₈₀B₂₀ films grown on cantilever shaped glass substrates by triode sputtering is the balance of three contributions: stress induced anisotropy, field induced anisotropy, and oblique incidence anisotropy. This balance is determined by the energy of the incoming flux. At a fixed deposition angle, the stress induced anisotropy competes with the field induced anisotropy at target potentials below -1 kV and above this value, the detailed balance between the three anisotropy sources has to be considered.

IV. CONCLUSIONS

In situ stress measurements during sputtering of amorphous Fe₈₀B₂₀ films are presented. The glass substrate curvature is measured optically during growth and the resulting film magnetic properties are correlated with the measured stress for a wide range of sputtering pressures. The field and magnetoelastic induced anisotropy balance can be controlled by varying the Ar gas pressure, which, in turn, controls the degree of energetic particle bombardment during sputtering. Three markedly different regions of stress have been distinguished. An initial apparent compressive stress is related to the heat transfer of energetic arriving atoms as the target bias is turned on. This apparent stress disappears after switching the target potential off. The second regime is where a tensile stress develops as islands nucleate and grow. A maximum of tensile stress occurs as the islands coalesce. Finally, a regime of compressive stress develops due to an “atomic peening” mechanism. The control of the final stress state of Fe₈₀B₂₀ films grown on cantilever shaped substrates allows tailoring of the magnetic anisotropy, which is crucial for magnetomechanical actuators.

ACKNOWLEDGMENTS

I.F.M. would like to thank MEC for the FPI financial support. J. P. Silveira is gratefully acknowledged for stimulating support.

- ¹T. Egami, *Rep. Prog. Phys.* **47**, 1601 (1984).
- ²R. C. O’Handley, *J. Appl. Phys.* **62**, R15 (1987).
- ³A. Inoue, T. Zhang, and A. Takeuchi, *Appl. Phys. Lett.* **71**, 464 (1997).
- ⁴H. Karamon, T. Masumoto, and Y. Makino, *J. Appl. Phys.* **57**, 3527 (1985).
- ⁵K. Hashimoto, N. Kumagai, H. Yoshioka, J. H. Kim, E. Akiyama, H. Habazaki, S. Mrowec, A. Kawashima, and K. Asami, *Corros. Sci.* **35**, 363 (1993).
- ⁶A. Inoue, T. Zhang, H. Koshiba, and A. Makino, *J. Appl. Phys.* **83**, 6326 (1998).
- ⁷M. L. Spano, K. B. Hataway, and H. T. Savage, *J. Appl. Phys.* **53**, 2667 (1982).
- ⁸A. Hernando, M. Liniers, and V. Madurga, *An. Fis., Ser. A* **79**, 136 (1983).
- ⁹R. Koch, *J. Phys.: Condens. Matter* **6**, 9519 (1994).
- ¹⁰J. A. Thornton, *J. Vac. Sci. Technol.* **11**, 666 (1974).
- ¹¹S. Dina, U. Geyer, and G. V. Minnigerode, *Ann. Phys.* **4**, 623 (1995).
- ¹²H. J. Leamy and A. G. Dirks, *J. Appl. Phys.* **49**, 3430 (1978).
- ¹³I. Fernández-Martínez, M. S. Martín-González, R. González-Arrabal, R. Álvarez-Sánchez, F. Briones, and J. L. Costa-Krämer, *J. Magn. Magn. Mater.* **320**, 68 (2008).
- ¹⁴M. R. J. Gibbs, E. W. Hill, and P. J. Wright, *J. Phys. D* **37**, R237 (2004).
- ¹⁵H. Ibach, *Surf. Sci. Rep.* **29**, 195 (1997).
- ¹⁶G. Armelles, J. L. Costa-Krämer, J. I. Martín, J. V. Anguita, and J. L. Vicent, *Appl. Phys. Lett.* **77**, 2039 (2000).
- ¹⁷C. Friesen and C. V. Thompson, *Phys. Rev. Lett.* **89**, 126103 (2002).
- ¹⁸J. A. Thornton and D. W. Hoffman, *Thin Solid Films* **171**, 5 (1989).
- ¹⁹I. Fernández-Martínez, J. L. Costa-Krämer, and F. Briones (unpublished).
- ²⁰K. Meyer, I. K. Schuller, and C. M. Falco, *J. Appl. Phys.* **52**, 5803 (1981).
- ²¹J. C. Slonczewski, in *Magnetic Annealing*, edited by G. Rado and H. Suhl (Academic, New York, 1963), Vol. I, p. 205.
- ²²K. Dahmen, S. Lehwald, and H. Ibach, *Surf. Sci.* **446**, 161 (2000).
- ²³B. D. Cullity, *Introduction to Magnetic Materials* (Addison-Wesley, Reading, MA, 1971), p. 330.
- ²⁴J. D. Verhoeven, E. D. Gibson, and O. D. MacMasters, *J. Appl. Phys.* **66**, 772 (1989).