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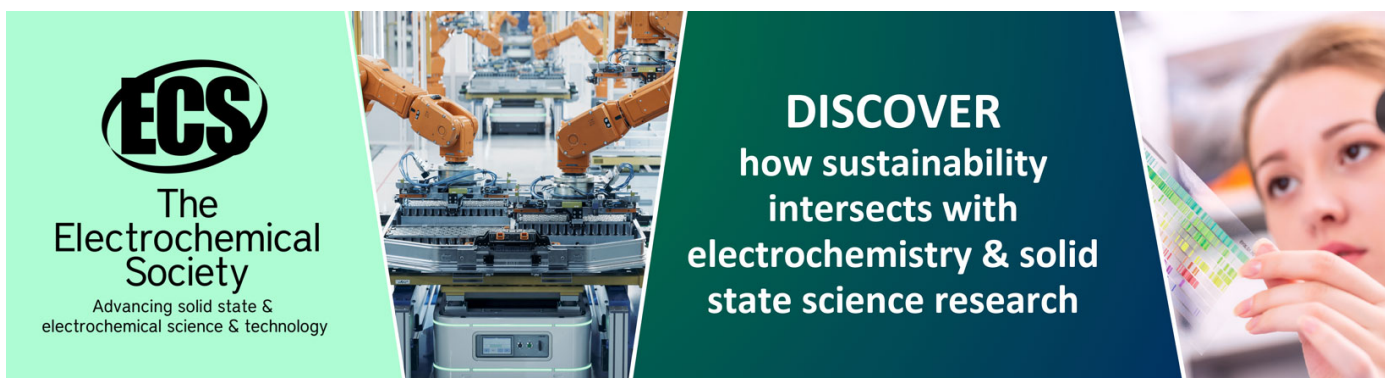
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Magnetoelastic effect in soft amorphous and nanocrystalline FeCuNbSiB thin films

Johan MOULIN¹, Medhi AMMAR¹, Iman SHAHOSSEINI¹, Francisco ALVES²,
Frédéric MAZALEYRAT³

¹ IEF, UMR 8622 Université Paris Sud XI - CNRS, Orsay, France

² LGEP, UMR 8507 Supelec-CNRS, Gif-Sur-Yvette, France

³ SATIE, UMR 8029 ENS Cachan-CNRS, Cachan, France

johan.moulin@u-psud.fr

Abstract. Thin films of FeCuNbSiB have been sputtered on SiO₂/Si substrates with thickness varying from 50 nm to 1.8 microns, then annealed at temperature ranging from 200 to 500 °C. The coercivity globally decreases with the film thickness down to 100 A m⁻¹ for as-deposited samples and 10 A m⁻¹ for annealed samples. However, it appears a local maximum for thickness close to 1 micron (i.e. the exchange length), regardless the annealing temperature. This behavior is supposed to be related to a modification of the magnetic domain structure, as the RAM applied to thin films and the dependence of the transitions points (glass state temperature T_g and crystallization temperature T_x) on the film dimension are monotonous. In addition, the films have been annealed under magnetic field and strong anisotropy energy has been reached, in the range of 150 J m⁻³.

1. Introduction

Since the discovery of the ultra-soft nanocrystalline Finemet-type alloys [1] and the explanation of their properties using the Random Anisotropy Model [2], these materials have been extensively used in electrical engineering or sensing fields. However, Finemet thin films have found few applications, except for GMI sensors fabrication [3] [4], because of an elaboration process –deposition then annealing for nanocrystallization- sometimes incompatible with MEMS technology. In addition, ultrasoft thin films are uneasy to obtain, as stress induced by the classical deposition techniques is high. This work presents the results of magnetic and mechanical investigations, in order to determine the relationship between the magnetic softness and the thermal behavior of amorphous and nanocrystalline film.

2. Film fabrication

The films were deposited by RF sputtering in argon plasma. Previous studies focused on the influence of oxygen contamination in the magnetic properties of thin films sputtered by this technique [5]. In this way, a liquid nitrogen trap has been added to the sputtering equipment, so that the residual vacuum before the sputtering was always less than 10⁻⁵ Pa. The other experimental conditions (RF power 250 W and Ar working pressure 4 Pa) lead to a deposition rate close to 14 nm min⁻¹. The sample was not cooled during the deposition, its temperature increased up to a

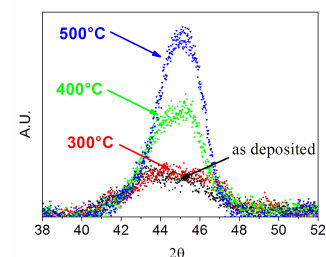


Fig 1. X-ray diagrams of as-deposited and annealed films

value ranging from 30 to 50°C depending on the sputtering time. The films composition, as determined by EDS, is close to $\text{Fe}_{72}\text{Cu}_3\text{Nb}_3\text{Si}_{15}\text{B}_7$. It differs from the target, i.e. $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{15.5}\text{B}_7$, in the sense that copper content is slightly higher. The films were processed on 2" silicon substrates covered by a 200 nm SiO_2 layer grown by dry oxidation and a 25 nm titanium adhesion layer deposited by RF sputtering. Samples were annealed for 1 h in a secondary vacuum between 200°C and 500°C with a heating and cooling rate of 10°C/min. The magnetic properties of the films were extracted from the hysteresis loops recorded at room temperature using a Princeton Measurement AGFM and a Lakeshore VSM for ultra-soft films. The microstructure has been investigated using a Philips X'Pert Pro diffractometer with a Cu anticathod ($\lambda = 1.541 \text{ \AA}$). The diagrams show that the as-deposited films are amorphous (see Fig 1) and remain in this state after annealing at 300°C. From 400°C, the films are crystallized, but the crystallites remain small, about 5 nm in diameter after Scherrer formula, and the Si-content in the grains remains lower (approximately 12%) compared to the Finemet ribbons (20%.) This difference can be explained by the shift in the composition, as copper is favorable to nucleation. The internal stress in the films has been measured as a function of the temperature using a FSM 500 TC. This scanning technique has already been described in a previous work [6].

3. Mechanical behaviour

The internal stress has been measured as a function of temperature with a heating and cooling rate of 2.5 K.min⁻¹. Two sets of measurements have been performed: samples with different thicknesses heated up to 500°C and then cooled (see Fig 2), and 500 nm samples heated at 200°C, 300°C 400°C or 500°C and then cooled.

The heating curve is characterized by four zones.

At low temperature, below 200°C, the decrease of the internal stress is linear and the phenomenon is reversible. The stress variations can be explained by the differential thermal expansion (DTE) effect between the substrate and the amorphous film, following the law:

$$\sigma_{\text{therm exp}} = (\mu_w - \mu_f) \frac{E_f}{1 - \nu_f} \Delta T = a \Delta T \quad (1)$$

with μ_w , μ_f , E_f , ν_f and ΔT respectively the thermal expansion coefficients of the substrate and the film, Young's modulus and Poisson's ratio of the film and the temperature variation involved in the thermal expansion process. The value of the coefficient a is independent of the film thickness and can be estimated to -0.75 MPa K^{-1} . As temperature rises above 200°C, a break in the slope is observed. Similar results have been obtained in Finemet or similar amorphous ribbons through the

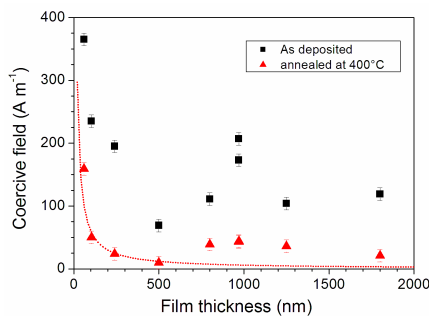


Fig 3. Coercive field as a function of the film thickness before and after annealing at 400°C. The red line corresponds to the RAM applied to thin films.

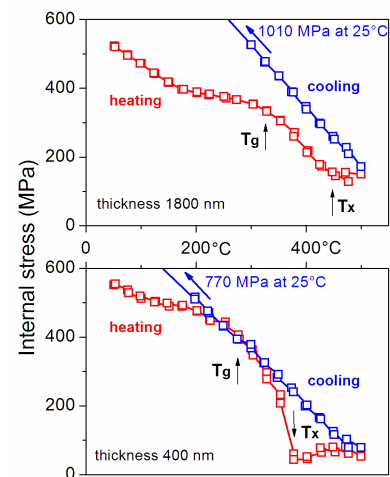


Fig 2. Internal stress measurement during heating up to 500°C then cooling for 400 nm and 1800 nm thick films.

direct measurement of the Young modulus [7] or the strain [8 9]. This stage is associated to a topological ordering and rearrangement. Then the decrease of the stress gradually accelerates, as the amorphous material enters in the supercooled liquid state. The temperature corresponding to the beginning of this zone is the glass transition T_g . It is to notice that T_g values are far below that of bulk material, from 275°C for the 400 nm film to 325°C for the 1.8 μm film (see table 1). These low values are explained both by the slow heating rate and by the presence of stress applied to the film during the measurements. Above this stage, the crystallization begins. Young's modulus of the material increases and compensates

the DTE effect, which leads to a stabilization of the internal stresses. The dependence of the crystallization temperature T_x on the film thickness is similar to that of T_g . The extension of the supercooled liquid region depends on the film dimensions, increasing from 100°C to 125°C for film thickness increasing from 400 nm to 1.8 μm .

During the cooling, the curve is linear because of the DTE between the substrate and the film. This reflects the irreversibility of the phenomenon happened during the heating. It is to notice that after annealing, the slope is increasing, which correspond to an enhancement of Young's modulus (see Eq. 1).

Table 1. Influence of the films thickness and the annealing temperature on the characteristic parameters.

Film thickness	Final temperature	T_g	T_x	Cooling slope
400 nm	500°C	275°C	375 °C	-1.52 MPa·K ⁻¹
500 nm	200°C			- 0.75 MPa·K ⁻¹
500 nm	300°C			- 0.76 MPa·K ⁻¹
500 nm	400°C			- 0.78 MPa·K ⁻¹
500 nm	500°C	300°C	400 °C	- 1.63 MPa·K ⁻¹
1000 nm	500°C	310°C	425 °C	
1800 nm	500°C	325°C	450 °C	- 1.77 MPa·K ⁻¹

4. Magnetic behaviour

Films with varying thickness and patterned with 2 mm $\times$ 2 mm squares have been deposited by shadow masking on a SiO₂/Si substrate. After separation, the samples have been annealed for 1h at various temperatures in secondary vacuum ($P_{\text{res}} < 10^{-7}$ mbar) in order to avoid surface oxidation. The effect of annealing is obvious (see Fig. 3), as coercivity is lowered by several units independently of the film thickness. As expected, the coercivity of annealed films decreases with thickness. Indeed, it has been already shown that the Random Anisotropy Model (RAM) applied to thin film leads to an effective anisotropy energy inversely proportional to the film thickness [6]. This model fits properly measurements up to 500 nm. At this point, the coercive field is minimal (10 A m⁻¹ for the 500 nm film). Above this point, the coercivity increases and reaches a local maximum at 1 μm , which corresponds approximately to the wall thickness in such materials. It is notable that $T_x = 400^\circ\text{C}$ for a 500 nm thick film, i.e. thinner films annealed at 400°C are crystallized and thicker ones are amorphous. Nevertheless, the same evolution can be observed for non annealed samples, all amorphous. This latter result shows that the magnetocrystalline anisotropy is not the only source of the increasing of the coercive field near 1 μm . It is also supposed that this behavior is linked to a modification of the domain structure.

5. Magnetoelastic interactions

The magnetic and mechanical properties of a 500 nm thick film are reported in the Fig 4. The internal stress was measured on continuous films and to the coercive field with 2 mm $\times$ 2 mm samples, after annealing between 200°C and 500°C. The magnetostrictive effect is obvious, as the coercive field variations follows that of residual stress. However, values for H_c are much lower than expected. Indeed, starting from the magnetostriction coefficient $\lambda_s = 23$ ppm for the studied alloy, the magnetostrictive energy associated to the stress induced by

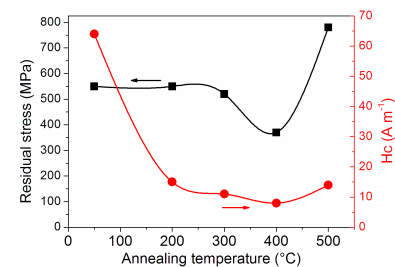


Fig 4. Residual stress and coercive field of a 500 nm thick film annealed a various temperatures

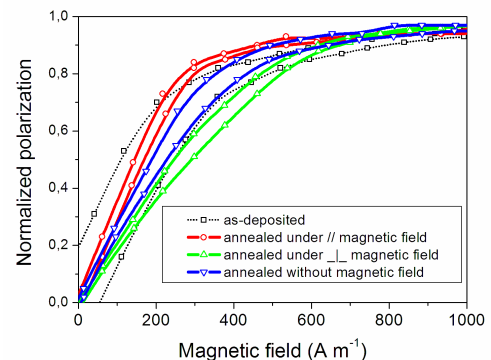


Fig 5. Magnetic annealing applied to 500 nm thick films

the deposition is:

$$E_s = \lambda_s \sigma \quad (2)$$

which corresponds to a value close to 12 kJ m^{-3} . These weak values of H_c are explained by the fact that the stress inside the discontinuous film (patterned with 4 mm^2 squares separated by 2 mm) is much lower than that inside a continuous film on a whole wafer. Considering that the magnetostrictive energy contribution is predominant, the equation 2 allows estimating the effective internal stress to 4 MPa only. Taking into account this correction, one can estimate the magnetostriction coefficient of the annealed samples to 4 to 5 ppm. It is to notice that the coercive field decreases strongly after annealing at 200°C , despite no evidence of a stress relief. This can be explained by the atomic rearrangement occurring at very low temperature (80-90 % of T_g , i.e. $\sim 500 \text{ K}$) due to both the high free volume in amorphous materials and high Fe-content.

In order to induce anisotropy, 500 nm thick films have been annealed at 300°C under a 50 kA m^{-1} magnetic field (see Fig 5). The estimated anisotropy energy induced in the film is in the range of 150 J m^{-3} . This value is much larger than energies induced in ribbons using magnetic annealing [10], which confirms a weak but non-zero magnetostriction effect in the films. These measurements have been validated by the visualization of the magnetic structure using longitudinal Kerr-effect (see Fig. 6). In the remanent state, the domains are randomly oriented in the stressed amorphous film (to the left) and aligned in the direction of the applied magnetic field in the film annealed at 300°C (to the right).

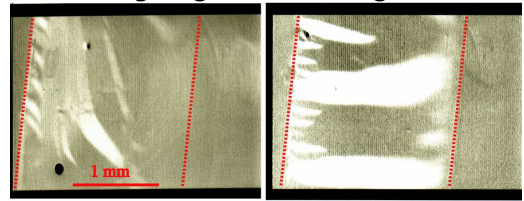


Fig 6. Magnetic structure of 500 nm films in the as-deposited state (left) and annealed at 300°C under a horizontal magnetic field (right).

6. Conclusion

Finemet thin films deposited using RF sputtering in high quality secondary vacuum exhibit ultrasoft magnetic properties. This behaviour is related to the stress relief in the material that is optimal for an annealing just below the crystallization temperature. Indeed, the crystallization increases the Young's modulus and consequently the residual stress after cooling. The weak but non-null magnetostriction effect in the films allows inducing high anisotropy energy, in the range of 150 J m^{-3} for the sample annealed at 300°C .

Furthermore, a particular point has been found in the dependence of the coercive field and the film thickness. Indeed, H_c strongly increases around $1 \mu\text{m}$. As this parameter depends on neither the film microstructure nor the mechanical properties, this phenomenon is associated to a modification in the magnetic structure.

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