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Internal stress influence on the coercivity of FeCuNbSiB thin films

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ABSTRACT

Thin films of Finemet-type alloy with thickness varying from 50 to 1000 nm have been deposited by RF sputtering and annealed at temperature ranging from 150 to 450 °C. Their magnetic and structural properties have been characterized using alternating gradient field magnetometry and X-ray diffraction. In addition, the stress in the films has been measured as a function of temperature from the curvature of the wafers using a laser scanning technique.

The coercive field of the films first decreases with annealing temperature due to stress relaxation, and then increases again when crystallisation begins. The optimal annealing conditions comprises between the glass transition and the crystallisation temperature.

Its is observed that the coercivity of the as-deposited material is continuously decreasing as the thickness increases, following an inverse square root dependence, in relation with the stress-induced magneto-elastic contribution to the total anisotropy. By opposition, it has been found that the coercive field of devitrified and totally relaxed films is inversely proportional to film thickness. In order to explain this evolution, a model is proposed, based on random anisotropy considerations applied to thin films in which the anisotropy was considered localised in the dimension of thickness.

1. Introduction

The efficiency of electromagnetic microdevices involving soft magnetic materials like actuators, sensors or transformers is most of the time driven by the magnetic losses in the constitutive ferromagnetic layer. In this frame, the research in this domain is oriented to the fabrication of thin films with the softest magnetic properties.

However, magnetic softness is highly dependent on the internal stress induced both by the deposition technique and the differential thermal expansion with the substrate:

$$\sigma = \sigma_{sput} + \sigma_{therm\ \ exp} = \sigma_{sput} + (\mu_w - \mu_f)\frac{E_f}{1-\upsilon_f}\Delta T \quad (0.1)$$

with μ_w, μ_f, E_f, υ_f and ΔT, respectively, the thermal expansion coefficients of the substrate and the film, the Young modulus and Poisson's ratio of the film and the temperature variation involved in the thermal expansion process. The sputtering technique, despite its convenience, is known for inducing high stress in the deposited films. Nevertheless, the contribution of stress to the magnetic anisotropy can be limited by the using of a non-magnetostrictive material or by a specific sample preparation or heat treatment.

The permalloy is often used as a soft magnetic material in microsystems, because it is easy to produce by sputtering or electrodeposition. However, this material suffers from a low electrical resistivity, which is detrimental to its high frequency use. Since its discovery in 1988 by Yoshizawa and al. [1], a new family of soft magnetic materials, namely Finemet, is available for MEMS fabrication. These amorphous Fe-based alloys show after heat treatment both soft magnetic properties, high polarization, thermal stability and higher resistivity (around 130 μΩ) than permalloy [2].

In this work, we present the correlation between magnetic properties and internal stress in Finemet thin films, as a function of the film thickness and annealing temperature. A model based on the random anisotropy model (RAM) will be proposed in order to explain the thickness dependence of the coercive films, taking into account to the internal stress.

2. Experimental procedure

The amorphous Finemet films were deposited by RF sputtering in argon plasma. The experimental conditions (RF power 250 W, residual vacuum 3×10^{-5} Pa and working pressure 3 Pa) lead to a deposition rate close to 9.2 nm min^{-1}. The composition of the film has been measured by EDS and is close to $Fe_{72}Cu_3Nb_3Si_{15}B_7$. It differs sensitively from that of the target, i.e. $Fe_{73.5}Cu_1Nb_3Si_{15.5}B_7$, in the sense that copper content is slightly higher. The films were

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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 at temperature ranged between 150 and 450 °C with a heating and cooling rate of 10 °C/min.

The magnetic properties of the films were extracted from the hysteresis cycles drawn at room temperature using a Princeton Measurement AGFM. The microstructure has been investigated using a Philips XRD diffractometer with a Co anticathod ($\lambda = 1.7889$ Å).

The internal stress in the films has been measured as a function of the temperature using a FSM 500 TC. This scanning technique detects the deflection of a laser beam on the surface of the wafer, which allows to extract its curvature and finally the internal stress of the film. Indeed, under some conditions (film thickness negligible compared to the wafer thickness, working in elastic domain), the wafer can be considered as a sphere portion and the stress in the film can be expressed as

$$\sigma = \frac{E_w}{6(1-\upsilon_w)}\frac{t_w^2}{t}\left(\frac{1}{R}-\frac{1}{R_0}\right) \tag{0.2}$$

where $E_w = 150$ GPa, $\upsilon_w = 0.17$, $t_w = 280\,\mu$m, respectively, Young's modulus, Poisson's ratio and thickness of the substrate are the constants and t, R_0 and R are the thicknesses, the curvature radius before and after film deposition, respectively. In order to avoid sample oxidation, the measurements were performed in a forming gas (95% Ar, 5% H_2).

For reference, a wafer covered with silica and titanium only has been tested using this technique and no evidence of wafer curvature was observed.

3. Results

3.1. Magnetic properties of finemet thin films

A set of films with different thicknesses has been deposited and annealed. The coercive field of these films has been measured as a function of the annealing temperature (see Fig. 1). It has been found that the optimal annealing temperature T_{opt}, resulting in the softest magnetic properties, depends on the film thickness. T_{opt} increases from 225 °C for a 50 nm thick layer to 300 °C for a 1 μm thick layer. Such behaviours have been already observed in previous works, for films of similar composition [3,4] as well as for other Fe-based amorphous alloys [5].

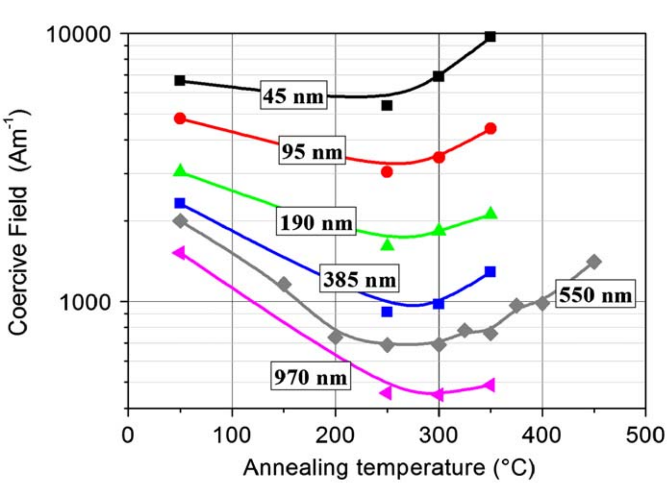

Fig. 1. Coercive field of thin films with various thicknesses as a function of annealing temperature.

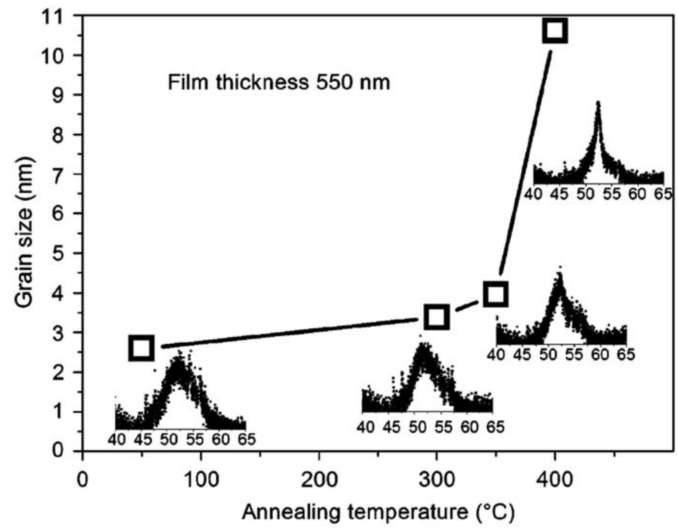

Fig. 2. Grain size and corresponding X-ray diagrams of the 550 nm film annealed at different temperature.

In a previous work [3], it has been supposed that T_{opt} is the result of two competitive phenomena: a softening of the material due to the relaxation of the stress induced by the deposition technique and a hardening due to the crystallisation. Indeed, it has been shown that the crystallisation does not lead to magnetic softening in thin films [4], which can be explained on the basis of random anisotropy model first applied by Herzer to soft nanocrystalline alloys [6]. However, the increase of the coercive field for annealing temperature above T_{opt} cannot be explained by an increasing of the magnetocrystalline energy only. Indeed, T_{opt} values are far below the crystallisation temperature for thin films [3,4], which has been confirmed by microstructural investigations. X-rays diffraction performed on a 550 nm thick film (see Fig. 2) reveals that α-Fe nucleation begin above 300 °C and that changes in microstructure is limited between 300 and 350 °C. The crystallites size remains smaller than that of bulk material, probably in relation with a relatively higher copper content in the films, increasing the density of nucleation sites. For sample annealed at 400 °C, the crystallites contain 5 at% of Si only, their size reaches 10 nm and crystallised volume is about 30%.

For a given annealing temperature, the coercive field H_c depends on the film thickness t. The Fig. 3 shows that analytical relation between H_c and t is allotropic

$$H_c = \frac{C}{t^n} \tag{0.3}$$

with C a constant.

The power n has been computed and the results are presented in Fig. 3. It has been found that the coercive field varies from $1/\sqrt{t}$ (as-deposited films) to a $1/t$ (films annealed at 400 °C) dependence.

In nanocrystalline films, local magnetocrystalline and magnetostatic energies only are supposed to be involved in the magnetization process since the stress due to the sputtering has been relaxed by annealing.

Following the RAM, the effective magnetocrystalline energy depends on the number of grains N included in the exchange volume

$$K_{eff} = \frac{xK_1}{\sqrt{N}} \tag{0.4}$$

where x and K_1 are the volumic ratio and the magnetocrystalline energy constant of the crystallites, respectively. N is defined by the