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Magnetic anisotropy in iron thin films evaporated under ultra-high vacuum

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ABSTRACT

α -iron thin films with thickness ranging between 20 and 1500 nm have been evaporated using an electron gun under ultra-high vacuum conditions ($5 \cdot 10^{-7}$ Pa). The columnar structure observed in cross-sectional TEM is related to the large surface diffusion. From Mössbauer spectra the spin orientation is deduced and found to be influenced by the column axis. Spins can be obtained perpendicularly to the film plane by rotating the substrate during the deposition. The magnetization of the samples is reduced by about 30 % and the reduction attributed to the interstitial space which increases with the incident angle. The substrate rotation also decreases $K_u(//)$ by a factor 2 and increases $K_u(\perp)$.

I. INTRODUCTION

Evaporated thin films are usually considered as the most appropriate materials for consumer magnetic products. Due to their high saturation magnetization and coercive force, iron films deposited at oblique incidence angle are promising. Hara et al. [1] have shown that magnetic anisotropy can be analysed using two components. The first one is in the incident plane and attributable to the columnar growth ; the second one is in the direction perpendicular to the incident plane and due to the bundle formation.

In this study, the films were structurally analysed using a combination of planar TEM, cross-sectional TEM and high resolution TEM (HREM). Magnetic characterisations were carried out using ^{57}Fe Mössbauer spectroscopy and magnetometry. Special attention will be paid to the influence of the substrate rotation on the columnar structure.

II. EXPERIMENTAL PROCEDURE

Iron films were electron-gun evaporated on various substrates (kapton, quartz, silicon, boron nitride) at room temperature at an incidence angle of 45° . The ultra-high

vacuum system consisted of ionic and cryogenic pumps so the residual pressure before deposition was about $2\text{--}4\cdot 10^{-7}$ Pa and $5\text{--}8\cdot 10^{-7}$ Pa during the film deposition. Active gas like carbon, oxygen, hydrogen and nitrogen were believed to be trapped by means of titanium sublimator and films were obtained in better conditions than the limit value ($2\cdot 10^{-3}$ Pa) beyond which magnetic properties can be affected [2, 3].

The evaporation rate was monitored by a calibrated quartz oscillator and fixed at a standard value of 0.5 nm/s. The substrate temperature (300 K before the deposition) was not water-cooled, and the possible temperature increase could not be measured during the deposition. These two conditions were similar to those chosen by Hara et al [1].

The peculiar influence of the incident plane inducing magnetic anisotropy was investigated by using a rotating device in order to avoid preferential incident plane, in this way the film growth must be homogeneous and cannot promote any geometrical direction.

Planar TEM experiments were performed on samples deposited either on microscope grids covered with carbon or freshly cleaved NaCl substrates. Cross-sectionnal specimens were obtained by the microtome technique slicing both film and substrate perpendicularly to the film plane. ^{57}Fe Mössbauer measurements were carried out using mainly an electron conversion counter working at RT ; more usual transmission spectra were also achieved through piled specimens deposited on kapton or boron nitride substrates.

Both magnetization (M) and M-H hysteresis loops were obtained using a vibrating sample magnetometer, under external fields (up to 15 kOe) applied in the film plane or normal to it. In-plane and out of plane anisotropy were also measured using a torque magnetometer.

III. RESULTS

Fig.1 shows both bright field (a) and diffraction rings (b) observed in a 60 nm thick film which correspond to a bcc structure with a mean grain size of about 7-9 nm. HREM (fig.2) reveals more details and disoriented grains of much smaller size can be detected. CE-EXAFS experiments [4] give us more informations ; the bcc structure is confirmed and the reduction observed in the EXAFS oscillations is analysed as a decrease of the cristallinity.

Fig.1. Bright field (x150000)

Fig.2. HREM (x 3.10⁶)

TEM observations in the cross-section of a 1000 nm thick film obtained with a rotating substrate are resumed in fig.3. Very marked columnar grains, almost perpendicular to the film plane, are formed and the diffraction rings near the substrate tend towards more dotted diffraction patterns at the top of the film. A noticeable grain growth then appears when the film becomes thicker ; moreover the upper part of a column becomes singlecrystalline. In addition, the columns very often present remarkable twins in their middle and the free surface consists of facets geometrically well defined.

Fig.3. Cross-sectional TEM - Thickness = 1500 nm
with rotation (x 100000)

Fig.4. Without rotation
(x 37000)

In this study, the very low pressure during deposition induces a great surface diffusion and the evaporation rate (0.5 nm/s) is sufficiently small to make the momentum of adatoms ineffective ; hence surface diffusion must be the predominant parameter and as a result an isotropic grain growth will be expected instead of the observed columnar structure. It seems that the role of surface diffusion of atoms had been overestimated in the competition with the geometrical movement of adatoms due to their momenta. Hara et al. [1] suggested that an active surface diffusion would point the column axis towards the vapour beam direction and produce a close dense-packed columnar structure. The surface density measured by grazing X-rays is decreasing gradually when the film thickness increases, showing a more and more weak package in the columns.

In the present experiments, the columnar structure normal to the film plane can be explained if the rotation of the substrate during the deposition is taken into account. Previous conclusions of Keitoku et al [5], when two symmetrically arranged sources are simultaneously used, suggest that the direction of the columnar grains is induced by the vector sum of the two vapour beams. With a rotating substrate (about 1 rps) around the normal of the film, the mean momentum lies on a cone axis and the vector sum is perpendicular to the substrate. As a result, the columnar grain growth will be directed along the normal of the film.

A confirmation of this explanation is easily obtained if the substrate is not rotating. Fig.4 shows a TEM cross-section of a 1500 nm film deposited onto a static substrate. The columnar axis then inclines towards the vapour beam direction. The

mean angle between the film normal and the columnar axis is about $30\text{--}35^\circ$ which is to be compared to the empirical value (26°) deduced from the tangent rule.

Mössbauer spectra enable us to calculate the average θ of the Fe spins with the film normal : $b = 4 \sin^2 \theta / (1 + \cos^2 \theta)$ where b is the relative intensities of the $\Delta m = 0$ lines. From Fig. 5, it can be seen that when the substrate is rotating the spins lie in the film plane and tend towards the film normal when the thickness increases. By comparison, when the deposition is realised onto a static substrate the mean θ angle tilts away from the film normal. As an example fig.5 shows a typical CEMS spectrum of a 20 nm film and also transmission spectra with (b) and without (c) rotating substrate for two 1500 nm films (see table 1). The transmission measurements take into account the crystal near the substrate where spins are in the film plane, so that in thick film the angle must be corrected and the true values of θ are in fact smaller ; hence the spins nearly lie along the columnar axis.

From Fig. 5c, an average angle $\theta = 36^\circ$ is deduced showing that spins and columnar axis make the same angle.

Fig.5. Mössbauer Spectra

- a) 20 nm film
- b) 1500 nm film with rotation
- c) 1500 nm without rotation.

Table 1 - θ angle versus thickness

Thickness (nm)	20	1500 rotating substrate	1500 static substrate
Geometry	reflection	transm.	transm.
θ angle ($^{\circ}$)	88	15	36

The magnetization is found to be 1200 ± 10 % emu/cm³, the main cause of error arising from the uncertainty in the film thickness. Therefore one observes a reduction of about 30 % with respect to bulk α -iron. A similar reduction in the magnetization of Fe films prepared by oblique incidence was also reported by Ozawa et al [6]. The authors found a strong dependence between the magnetisation and the angle of incidence. This apparent reduction in the magnetization has been attributed to the interstitial space which increases with the incident angle. Fig.6 shows the in-plane loop of a sample 500 nm thick prepared with substrate rotation. The loop shape and the coercive field of 140 Oe are in general agreement with the results of Ozawa et al. [6].

The values of the anisotropy measured by torque magnetometer for the samples prepared with and without substrate rotation are also shown in Table 2.

Table 2 - Anisotropy constants as a function of evaporation technique

Sample	(nm)	$K_u(\perp)$ (erg/cm ³)	$K_u(//)$ (erg/cm ³)
1. Fe (rotating)	500	7.2×10^6	4.4×10^5
2. Fe (static)	500	4.6×10^6	9.0×10^5

The above results are in agreement with the structural studies. For instance when the substrate is rotated, the columns tend to be normal to the film plane and hence due to dipolar effects an easy direction is induced along this direction leading to the higher value of $7.2 \cdot 10^6$ erg/cm³. Also, when the substrate is static, probably due to bundle effect, the in-plane anisotropy is enhanced and hence we find $K_u(//) = 9 \cdot 10^5$ erg/cm³.

As a conclusion it has been clearly shown that, under ultra-high vacuum conditions, the films present a columnar grain growth. The columnar axis tends towards the direction perpendicular to the film plane when the substrate is rotating. The spins lie along the column axis and the out of plane anisotropy is enhanced.

Fig.6. In-plane hysteresis loop

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