Magnetic behaviour of $^{57}\text{Fe}/\text{Co}/\text{Al}$ multilayers deposited on a glass substrate

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Abstract. Multilayers of $^{57}\text{Fe}/\text{Co}/\text{Al}$ 20 and 40 trilayers (TLs) prepared on a glass substrate using ion beam sputtering deposition techniques have been studied. Magnetization curves obtained at different angles show the anisotropic nature of both the samples. Both samples show strong in-plane uniaxial anisotropy. Negative δm values suggest that dipolar type interactions between grains contribute to the in-plane uniaxial anisotropy. Angular dependence of coercivity indicates that the magnetization reversal mechanism is different for both samples. The variation in coercivity with azimuthal angle is well reproduced in the 20 TL sample using the modified Kondorsky model and by the Stoner–Wohlfarth model in the 40 TL sample.

Keywords. Magnetic properties; magnetic multilayers; ion beam sputtering; magnetocrystalline anisotropy; uniaxial anisotropy; magnetization reversal.

1. Introduction

Fe–Co based nanostructures/multilayers (MLs) have been intensively studied because of their wealth of diverse properties, such as the presence of square loops, exchange bias, high magnetic moment, high Curie temperature and magnetocrystalline anisotropies which make them desirable for read/write head cores, recording/memory, sensors, magnetic amplifiers, pulse transformers, generators, motors, dynamos and switching circuits [1–4]. Such intrinsic magnetic properties are optimized by controlling the microstructure through appropriate thin film deposition techniques.

Magneto-crystalline anisotropy is also an important determining factor in the case of thin films and MLs. For parallel recording, in-plane uniaxial magneto-crystalline anisotropy is required and perpendicular magneto-crystalline anisotropy (PMA) is required for perpendicular recording. Low switching currents and large thermal stability can also be achieved in materials with PMA [5] and the perpendicular alignment of magnetic moments increases the storage areal density. While several reports over the last decades show that strong in-plane uniaxial magneto-crystalline anisotropy and PMA can be realized in Fe/Co-based MLs through application of field during deposition, high interface roughness and compressive stresses induced by the lattice mismatch between Fe and Co [6,7], there is not much agreement on the origin of these anisotropies. Moulas [8] has pointed out that although the magnetic anisotropy energy in bulk Fe–Co alloys is small

due to cubic symmetry, it is possible to achieve high magnetic anisotropy energy by breaking the cubic symmetry through the design of artificial structures, such as superlattices and MLs.

A detailed study on the MLs of the $^{57}\text{Fe}/\text{Co}/\text{Al}$ trilayer (TL) system with variation of modulation periods and substrates is still not available in the literature. We had recently reported the presence of strong uniaxial anisotropy in $^{57}\text{Fe}/\text{Co}/\text{Al}$ MLs deposited on the Si (100) substrate [9]. Continuing this study, in this paper, we report the results of structural and magnetic studies on the $^{57}\text{Fe}/\text{Co}/\text{Al}$ ML system, which exhibit strong in-plane uniaxial magneto-crystalline anisotropy with different layer thicknesses deposited on the glass substrate.

2. Experimental

ML films were prepared in an ion beam sputtering system. Two samples, $[^{57}\text{Fe}_{17\text{\AA}}/\text{Co}_{8\text{\AA}}/\text{Al}_{10\text{\AA}}]_{\times 20}$, 20 TLs and $[^{57}\text{Fe}_{8.5\text{\AA}}/\text{Co}_{4\text{\AA}}/\text{Al}_{5\text{\AA}}]_{\times 40}$, 40 TLs with different individual layer thicknesses were prepared on a glass substrate by using a broad-beam Kaufman types ion source with Ar ions with an energy of 800 eV and a beam current of 25 mA. The base pressure inside the chamber was 1.5×10^{-7} Torr, while Ar gas pressure during deposition was $\sim 2 \times 10^{-4}$ Torr. The rate of deposition was calibrated by individually sputtering Fe, Co and Al for 10 min under identical conditions. Each target used in the system has purity > 99.99%.

Table 1. Structural parameters obtained from XRD and XRR measurements.

Sample	Lattice parameter (Å) ± 0.002 (Å)	Crystallite size (nm) ± 1 nm	Simulation thickness (Å)	Average interface roughness (σ) (Å)
20 TLs	2.885	10	~ 700	~ 15.12
40 TLs	2.894	9	~ 690	~ 10.33

Figure 1. High angle X-ray patterns for 20 and 40 TL ML samples.

After the growth, the samples were subjected to multiple characterization techniques. Structural characterization of the MLs, such as thickness, interfacial roughness, phase formation and crystallite size, were obtained by X-ray reflectivity (XRR) and X-ray diffraction (XRD) measurements using a Bruker D8 Advance X-ray diffractometer with $\text{CuK}\alpha$ radiation. Analysis of the reflectivity data to obtain layer thickness and roughness was performed using Parratt's formalism (table 1) [10]. The surface morphology was studied using atomic force microscopy (AFM) and the magnetic properties of the samples at room temperature were characterized using a Lake Shore 7304 vibrating sample magnetometer (VSM). Angular dependence on magnetization was studied by recording the rotation magnetization curve in the in-plane directions.

3. Results and discussion

Figure 1 shows the XRD pattern of the MLs of 20 and 40 TL systems, both of which possess only one broad peak with low intensity around $2\theta = 44^\circ$, which can be attributed to Fe/Co/Al (110). Other Fe/Co/Al peaks are not visible in the XRD spectra and this may be due to the relative weak intensity of other Fe/Co/Al peaks. The crystallite size of the samples was obtained by first fitting

Figure 2. XRR patterns of 20 and 40 TL ML samples. Experimental data are given by red open symbols and the blue solid line shows the fitted data.

the diffraction peaks with a pseudo-Voigt profile function and extracting the line width and position. Subsequently, the Scherrer equation was used to calculate the crystallite size. The value of lattice parameter for Al (2.89 Å) is greater than that reported for Fe/Co (2.82 Å) because the atomic size of Al is much bigger than Co (1.67 Å) or Fe (1.72 Å); thus any substitution of Fe or Co by Al would lead to a considerable enlargement of the lattice size. The crystallite size is about ~ 10 nm in both MLs.

Figure 2 shows the spectra of XRR measurements. An independent layer model based on the iterative Parratt recursive method has been used for fitting the XRR spectrum to elucidate the ML structure of the films. XRR patterns of both samples show a ML structure with very low signal intensity at high angles because of the low thickness of the MLs and diffusion of aluminium. Also, the average roughness for the 40 TL sample is more due to the much smaller thickness of individual layers as compared to the 20 TL sample. Overall high interface roughness due to interdiffusion is observed in both systems, which leads to unusual magnetic properties in these samples.

Figure 3. (a and b) are EA and HA curves of 20 and 40 TL samples. (c–f) are polar curves as a function of squareness and coercivity. All figures are for in-plane magnetization.

Room temperature hysteresis loops have been recorded for 20 and 40 TL samples with respect to the direction of the applied magnetic field in two configurations: (1) magnetic field along the in-plane (parallel) direction of the sample and (2) magnetic field along the out-of-plane (perpendicular) direction of the sample. Hysteresis loops have also been obtained in the in-plane/out-of-plane configurations by orienting the film at different angles (azimuthal angle θ) with

respect to the thin film plane. Figure 3a and b shows the in-plane, easy axis (EA) and hard axis (HA) curves at room temperature. Parameters obtained from magnetization measurements are listed in table 2.

The maximum of saturation values (M_s) in the in-plane direction along the HA are 1038 and 1105 emu cm⁻³, respectively, for the 20 and 40 TL samples, while the thickness of nonmagnetic Al is ~28% of the total thickness in both

Table 2. Experimental and calculated magnetization parameters at room temperature.

Samples	M_s (emu cm $^{-3}$) (EA–HA)	H_c (Oe) (EA–HA)	M_r/M_s (max.)	H_k (Oe)	K_u (10 4 erg cm $^{-3}$)
20 TLs	930–1038	~1–4	99%	~47	2.18–2.43
40 TLs	1036–1105	~3–11	95%	~38	1.96–2.09

samples. Almost all the magnetic moments in these TLs originate from the ferromagnetic Fe/Co layers and thus can be estimated using the following expression [11]:

$$M_{\text{FeCo}} = M_s \left(\frac{\sigma}{t_{\text{FeCo}}} \right). \quad (1)$$

The total thickness of MLs is given by σ and t_{FeCo} is the total thickness of the ferromagnetic layers in the sample. The M_s values thus estimated are 1414 and 1547 emu cm $^{-3}$ for the 20 and 40 TL samples, respectively, while the saturation magnetization values reported for FeCo (1910 emu cm $^{-3}$) are more than these values. This is due to the formation of nonmagnetic phases at the interfaces because of which a lower value of saturation magnetization is obtained in the TL samples as compared to FeCo.

The in-plane anisotropy field (H_k) was calculated by taking the intersection point of EA and HA, and is above 35 Oe for both samples. The anisotropy field H_k is the one at which the system will saturate if a perfect HA behaviour occurs. The M_s and H_k were deduced from the magnetization curves enabled the determination of the uniaxial anisotropy constant (K_u) which ranges from ~2.18 to ~2.43 $\times 10^4$ erg cm $^{-3}$ for the 20 TL sample and from ~1.96 to ~2.09 $\times 10^4$ erg cm $^{-3}$ for the 40 TL sample.

Although both samples are magnetically soft ($H_c \leq 11$ Oe), the coercivity of the 40 TL sample is more than that of the 20 TL sample. However, as pointed out earlier, the coercivities are still very small and can be explained by the random anisotropy model (RAM). According to RAM, if the crystal-lite/grain size is less than the ferromagnetic exchange length, the anisotropies are averaged out over several crystals and the sample exhibits good soft magnetic properties as is the case in the present samples. The ferromagnetic exchange length for Fe–Co is ~20 nm [12], while the average particle size in both MLs, as reported in table 1, is ~10 nm and the inter-layer separation is much < 20 nm. Both samples also have perfect square-shaped hysteresis loops showing high squareness ratios (M_r/M_s) (0.99 for the 20 TL and 0.95 for 40 TL samples along the EA). As is evident from figure 4, the mechanism of angular dependence of coercivity is different for both samples. Two types of magnetization reversal mechanisms are possible in highly uniaxial anisotropy systems, namely (a) coherent rotation and (b) domain–wall nucleation and/or depinning [13,14]. In the case of the 40 TL sample, the variation in coercivity is the same as that normally observed in ML systems, where it decreases from EA to HA and is explained by the Stoner–Wohlfarth (SW) model. According

Figure 4. Variation of H_c as a function of azimuthal angle (θ). The blue stars are experimental data and red open circle curves have been obtained using theoretical simulations.

to the SW model, the variation in coercivity from 0 to 45° (0° with respect to the EA) is given by the following relation:

$$\frac{H_c(\theta)}{H_c(0)} = (\cos^{2/3}\theta + \sin^{2/3}\theta)^{-3/2}. \quad (2)$$

Here, the angle along the EA is fixed to be 0° and $H_c(0)$ is the coercivity for measurement along the EA. $H_c(\theta)$ is then the coercivity for the M – H loop recorded at an angle θ with respect to the EA and $H_c(\theta)/H_c(0)$ is the reduced coercivity. For angles 45–90°, the reduced coercivity is given by:

$$\frac{H_c(\theta)}{H_c(0)} = \sin(\theta) \cos(\theta). \quad (3)$$

But in the case of the 20 TL sample, the coercivity along the HA is higher than that along the EA and can be explained by the modified Kondorsky relation, according to which the pinning sites have a predominant influence on magnetization

Figure 5. δm curves for in-plane magnetization.

reversal. Accordingly, the variation in coercivity from 0 to 60° can be expressed as:

$$\frac{H_c(\theta)}{H_c(0)} = h + \frac{(1-h)}{\cos(\theta)}, \quad (4)$$

where h is the reduced coercivity constant [15]. Figure 4 shows the experimental and simulated curves using different magnetization reversal mechanisms for both samples. While the 40 TL sample can be satisfactorily simulated using the SW model, the experimental data for the 20 TL sample cannot be simulated by a single model from 0 to 90° . In the 20 TL sample, the curve of reduced coercivity vs. the azimuthal

angle can be reproduced from 0 to 60° using the Kondorsky model by which the coercivity increases as the azimuthal angle increases. However, on approaching the HA, this model no longer holds good since the coercivity decreases from 75° to 90° and follows a complex system in which the domain wall motion as well as coherent rotation seem to play a role in the magnetization reversal process. Other workers have also observed such behaviour in various systems. For example, previously, Xi *et al* [16] and Pawlik *et al* [17] found complex magnetization reversal mechanisms in Sm-doped FeCo and Sm–Fe thin film systems.

Figure 3c–f represents the polar magnetization curves of the samples. Both samples show almost similar $\cos \theta$ behaviour with 180° periodicity. These results suggest a perfect in-plane, two-fold magnetic uniaxial anisotropy for both samples. Johnson *et al* [18] have explained the origin of magnetic anisotropy in terms of magnetic spin orbital and dipolar interaction with shape contribution to be zero for in-plane anisotropy. According to the SW model, the type of interaction between particles with uniaxial anisotropy can be determined from the remanence curves and Wohlfarth relation [13,19]. Isothermal remanent magnetization (IRM) curve and direct current demagnetization (DCD) measurements were therefore undertaken using the VSM to clarify the types of exchange interactions.

Figure 5 shows the δm (Kelly–Henkel plot) obtained from the IRM–DCD curves recorded for both samples at room temperature using the equation (Wohlfarth relation) given below:

$$\delta m = M_d(H) - [1 - 2M_r(H)]. \quad (5)$$

Here, $M_d(H)$ is the normalized DCD remanence as a function of the reversal field and $M_r(H)$ is the normalized IRM curve. A positive δm in the Kelly–Henkel plot indicates ferromagnetic exchange interactions and dipolar interactions for negative δm values. For both samples, δm curves suggest the presence of dipolar type interactions to be the dominant type. Due to its long-range character, the dipolar interactions

Figure 6. 3D topographic AFM images of 20 and 40 TL ML samples.

between spins of Fe–Fe, Fe–Co and Co–Co are also responsible for the in-plane magnetization observed in these samples.

To throw further light on the type of microstructure formed, three-dimensional (3D) topographic AFM images of the samples have been recorded and are displayed in figure 6 ($500 \times 500 \text{ nm}^2$). These images evidence the formation of irregular island-like (crests and troughs) structures in both samples which lead to strain and high surface roughness. For films, three types of deposition growth are possible: (1) layer by layer growth (two-dimensional), (2) island like growth (3D) and (3) layered growth followed by island growth (equilibrium growth) [20]. The type of growth depends on the thickness of the layers and the substrate. Generally, for very low thickness, island-like growth is observed since the direction of energy minimization is easily reversed and layers breakdown into small clusters on growing. An intermediate state is also possible, where the first few atomic layers grow as a layer, followed by a roughening transition that results in the formation of island-like structures. These structural distortions lead to stacking faults, twin boundaries, interstitials and vacancies, resulting in greater roughness. The surface/interface roughness (revealed by XRR and AFM), thus plays an important role in the magnetic anisotropic properties of these MLs.

4. Conclusion

Magnetically very soft anisotropic $^{57}\text{Fe}/\text{Co}/\text{Al}$ TL thin films on a glass substrate were successfully produced with high magnetic moment and high squareness. Soft magnetic properties are the result of a significant reduction in grain size, consistent with RAM. δm curves suggest that the dipolar interactions between grains lead to strong in-plane anisotropy.

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