Reorientation of magnetic anisotropy in obliquely sputtered metallic thin films

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Reorientation in the magnetic anisotropy as a function of film thickness has been observed in Co-Ni and Co thin films, obliquely sputtered on a polyethylene terephthalate substrate at a large incidence angle (70°). This effect is a consequence of the low magnetocrystalline anisotropy of the films (fcc structure of Co) and changes in microstructure from nuclei to columns according to the thickness. The critical thickness for this transition was estimated to be 30 nm for Co-Ni.

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Since the discovery of a large in-plane magnetic anisotropy in thin films of soft magnetic materials such as iron and Permalloy, grown by oblique deposition,[1–3] much attention has focused on this technique due to its potential application in high-density tape recording. It is well known that the morphology of obliquely deposited films consists of columnar type structures.[4] Depending on the incidence angle, a good separation between the columns can be achieved. Moreover, a large magnetic anisotropy can be induced due to the shape of the columns.

The orientation of the easy axis is the result of competition between various types of magnetic anisotropy in the material. In ultrathin films[5,6] it was shown that by reducing the film thickness, the easy axis could switch from the film plane to become perpendicular. The origin of this reorientation is due mainly to the surface anisotropy, which prevails at very low film thickness and can stabilize the easy axis out of plane. In obliquely deposited thin films, a reorientation of the anisotropy with the incidence angle was observed.[7,8] At growth angles larger than 60°, the easy axis stays confined to the incidence plane, whereas for lower angles the anisotropy switches to become parallel to the transverse direction. On the other hand, it was established that the structure of magnetic domains is strongly affected by the incidence angle and a transition from elongated to stripe domains may occur.[9]

In this paper, we report on the first observations showing a reorientation of magnetic anisotropy according to the film thickness in metallic films such as Co and Co-Ni, obliquely sputtered on a polymer substrate, polyethylene terephthalate (PET). Our samples have been prepared in a mini roll coater, which consists of a solid drum, covered with a polymer substrate. We have used a static configuration with continuous varying incidence (CVI). The films have been deposited at room temperature and at the same residual pressure of 4×10^{-3} mbar. By changing the sputtering time, films with different thicknesses have been grown at 70° of incidence angle. Because of similar effects observed in both materials (Co and Co-Ni), we have concentrated our studies in this paper to the Co-Ni films.

Figure 1 shows the magnetization loops of Co-Ni films with thicknesses varying between 13 and 132 nm, measured by a vibrating sample magnetometer (VSM). The solid and dashed curves correspond to in-plane loops, measured with field parallel to directions called longitudinal and transverse (parallel and perpendicular to the projection of incident beam in the film plane, respectively). At larger thickness, the longitudinal loops show a large coercivity H_c as well as high squareness S (for example, for $t = 132$ nm, $H_c = 48$ kA/m, $S = 0.81$), whereas those parameters are largely reduced in the transverse direction. Moreover, even by applying a large magnetic field (400 kA/m), it is hard to saturate the transverse loop. It is clear that thicker films exhibit the longitudinal direction as the preferred magnetization orientation. Upon reducing the film thickness, we see that the loop shape in both directions (longitudinal and transverse) varies continuously. It is a surprise to see such a drastic change in the

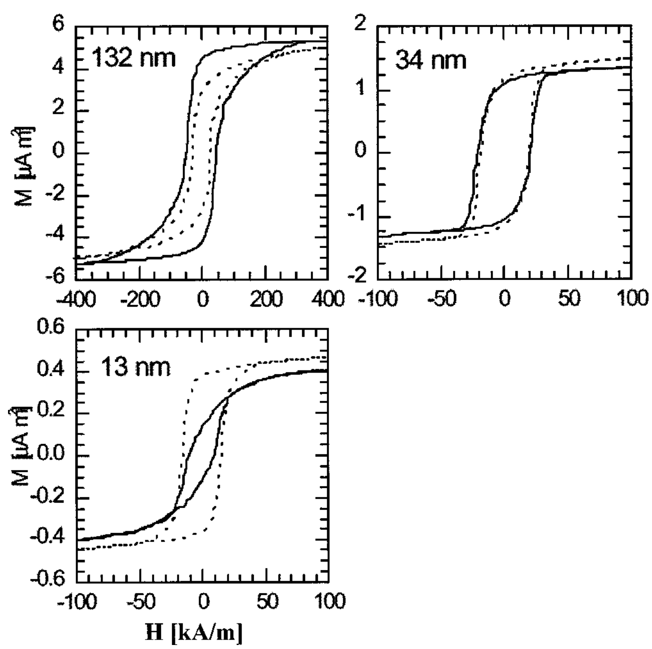

FIG. 1. Hysteresis loops of Co-Ni films with different thicknesses grown at 70° as incidence angle. The field was applied parallel to the longitudinal and transverse directions for solid and dashed curves, respectively. At large thickness the easy axis is confined to the incidence plane, whereas in thinner films the anisotropy switches to the transverse direction. Note the difference in the respective scales of the loops.

magnetization loops for thinner films ($t = 13$ nm). The transverse magnetization of very thin films ($t = 13$ nm) shows a square loop with a large remanence, whereas the hysteresis loss is strongly reduced in the longitudinal loop and a large field is required to saturate the magnetization following that direction. This result clearly indicates that after being confined in the incidence plane, the easy axis switches to be parallel to the transverse direction for low thicknesses. It is important to point out that close to a certain critical thickness ($t_c = 30$ nm), the two loops (longitudinal and transverse) exhibit similar behavior ($H_c = 20$ kA/m, $S = 0.8$), which indicates that both directions are magnetically equivalent. The critical thickness t_c can be considered as the crossover of the in-plane anisotropy transition. Additionally, Fig. 1 reveals many anomalous behaviors, which can be summarized as follows.

(i) At large thickness ($t = 132$ nm), although anisotropy in the incidence plane is confined, the transverse loop (hard axis) shows a large hysteresis with vertical switching, which clearly indicates the existence of a certain anisotropy oriented following in that direction. Moreover, the nonuniformity in the shape of the longitudinal loop reveals that more than one anisotropy form is active in the control of the magnetization during its reversal mechanism.

(ii) At the critical thickness t_c, both loops exhibit the same coercivity as well as a high remanence ($S = 0.8$). This squareness is too large and far exceeds the value typically known for a system with uniaxial anisotropy either oriented or random.

In order to establish the origin of the anisotropy orientation and to explain the anomalous effects revealed in Fig. 1, it is important to investigate different kinds of magnetic anisotropy in such a system. With the torque measurements, it was possible to establish the type and exact orientation of the easy axis in the films. Two kinds of measurement have been performed and consist of the following: (a) The magnetic field was rotated in the film plane in order to estimate the in-plane anisotropy. (b) The easy-axis tilt was determined by rotating the field from the normal to the parallel direction to the film plane. The in-plane torque curves reveal the existence of a uniaxial anisotropy with the easy axis depending on the film thickness. As is shown in Fig. 2, for large thicknesses, the anisotropy constant K_u is positive, indicating that the easy axis is aligned in the longitudinal direction. By reducing the film thickness, K_u decreases to vanish at the critical thickness t_c. Below t_c, K_u changes sign and becomes negative, which is a clear indication of a reorientation in anisotropy. In the latter range of thickness, the anisotropy chooses the transverse direction as the preferred orientation. From the second configuration of torque measurements, it was confirmed that the easy axis exhibits a tilt from the film plane. Figure 2 reveals the existence of a threshold thickness equal to t_c for the easy axis to be out of plane. Above t_c, the tilt of the easy axis increases and can reach 12° at a thickness of 132 nm. By combining both results of Fig. 2, we can establish that the anisotropy orientation consists of two regions of thickness: (a) Below t_c, the anisotropy is fully in plane and the easy axis lies along the transverse direction. (b) Above t_c, the anisotropy is confined to the incidence plane

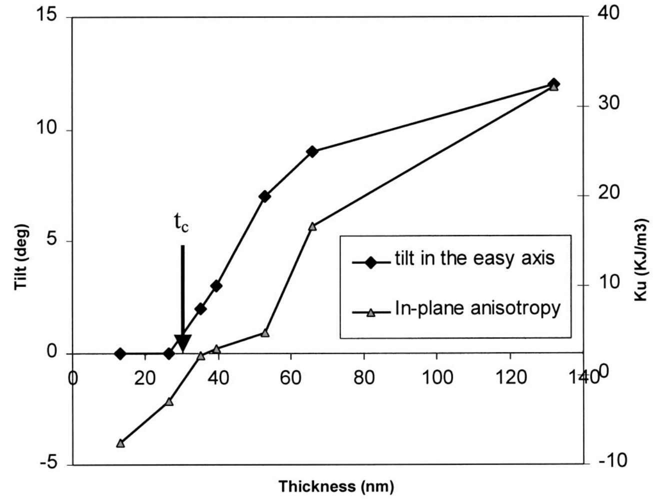

FIG. 2. Thickness dependence of the in-plane anisotropy and tilt of the easy axis. At a large thickness, K_u is positive, indicating that the easy axis is parallel to the incidence plane. However, above t_c, K_u becomes negative and confirms the reorientation of the anisotropy. Below t_c the anisotropy is fully in plane, whereas above t_c the easy axis exhibits a tilt, which increases with the film thickness.

and the easy axis is tilted from the film plane, depending on the film thickness. The crucial question arises as to what kinds of anisotropies are involved in controlling the magnetic properties and the reorientation of the easy axis. In order to establish the contribution of each magnetic anisotropy, further investigations have been performed and consist of microstructural analyses carried out by x-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and atomic force microscopy (AFM). At least two arguments suggest that magnetocrystalline anisotropy of hexagonal Co is not the prevailing form in the films: (a) The coercivity is too small, especially at low thickness compared to that usually induced by a hexagonal-close-packed (hcp) structure and varies strongly with the thickness. (b) If the magnetocrystalline anisotropy of the hcp structure is the main source of anisotropy in the film, reorientation of the easy axis should be related to a change in the *c*-axis orientation with thickness. Nevertheless, there is no valid reason to justify this brusque change in the texture with thickness. Additionally, XRD measurements (θ-2θ scan) reveal that the films are polycrystalline and cobalt exhibits a face-centered-cubic (fcc) rather than hcp structure. Clear evidence of the cubic structure of Co in the films is also confirmed by the magnetic loops around the critical thickness (Fig. 1). Both hysteresis loops show the same coercivity ($H_c = 20$ kA/m) as well as a high remanence ($S = 0.8$). Moreover, the torque analysis (Fig. 2) reveals the absence of any in-plane anisotropy at t_c. The existence of hysteresis in both directions with no in-plane anisotropy suggests a random distribution of the easy axes. If we consider that at t_c the anisotropy is uniaxial with a random distribution of the easy axes in the film plane, the maximum squareness which can be achieved is about $2/\pi \approx 0.636$. The large remanence (0.8) in both loops at t_c confirms that random uniaxial anisotropy is hardly conceivable in our films. The only way to

FIG. 3. TEM cross section made parallel to the incidence plane. The image shows tilted columns (55°) with strong shape anisotropy.

explain this result is to consider the cubic anisotropy of cobalt. In fact, for a random distribution of the easy axes in a cubic anisotropy, the remanence can be as large as 0.832.[10] In addition to the XRD measurements, the latter result reveals that the origin of magnetocrystalline anisotropy in the films is due to the fcc rather than the hcp structure. It is clear that the reorientation of the in-plane anisotropy assumes the existence of an additional uniaxial anisotropy, which is strongly dependent on the film thickness as revealed by the torque analyses (Fig. 2). In a recent work[11] we established from SEM investigations that at large incidence angle, the film microstructure consists of columns with an ellipsoidal shape, well separated from each other. The columns are oriented following the longitudinal direction. The isolation between the columns can be understood from the shadow effect, which increases enormously with the incidence angle. The low exchange coupling in such microstructure suggests that each column behaves like a single magnetic entity. Moreover, depending on the dimensions of the columns (length-to-diameter ratio), their shape anisotropy may be strong enough to play a crucial role in the control of the magnetic properties. In Fig. 3, a TEM image of Co-Ni film, 200 nm thick, is shown. This cross section is prepared parallel to the incidence plane and reveals the following information: (a) The interface between the metallic layer and polymer substrate is sharp. (b) The morphology of the film consists of columns tilted from the normal to the film plane. The tilt angle is estimated to be 55° and satisfies the tangent rule $\tan(70°) \approx 2\tan(55°)$.[12,13] (c) The length-to-diameter ratio of columns can be as high as 10, inducing a strong magnetic anisotropy estimated at 1.5×10^5 J/m^3. By considering the low random magnetocrystalline anisotropy of fcc Co and both anisotropies related to the shape of the columns and film, it is possible to interpret the results of films with large thickness (Figs. 1 and 2). Because of the large anisotropy of the columns and their alignment in the incidence plane, the easy axis will be confined to this plane. Moreover, due to competition between the two shape anisotropies (film and columns), the easy axis will be oriented between the columnar direction and the film plane. The loops of thicker films ($t=132$ nm) in Fig. 1 can be well interpreted by assuming the contribution of two different anisotropies (cubic and uniaxial) to the magnetization process. The cubic anisotropy (magnetocrystalline of fcc Co) is randomly oriented and can induce a large hysteresis with vertical switching in both directions (longitudinal and transverse). However, the uniaxial anisotropy (shape of the columns) stabilizes the easy axis in the incidence plane. The superposition of both anisotropies can explain the anomalous behaviors mentioned before in both loops. The dependence of the easy-axis tilt on the film thickness can be explained by the change in the shape anisotropy of the columns. In fact, variation of the film thickness can strongly affect the dimensions of the columns, especially in the elongated axis, which is supposed to be the fast direction of growth in such a microstructure. The reorientation of the anisotropy at low thickness suggests that a drastic change occurs in the microstructures. By reducing the film thickness, it is expected that the column length becomes smaller. Consequently, the shape anisotropy of the columns decreases, to disappear at the critical thickness where the length-to-diameter ratio is expected to be close to 1. Reduction of the shape anisotropy of the columns can explain the decrease in the tilt of the easy axis well (Fig. 2) due to the domination of the shape anisotropy of the film, which will try to confine the anisotropy as close as possible to the film plane. It is clear that in-plane uniaxial anisotropy is responsible for the orientation of the easy axis for thicker films and corresponds to the shape anisotropy of the columns. But what is the possible origin for the switching of the easy axis to the transverse direction at low thickness? Figure 4 shows an AFM image of a very thin film (13 nm) in which the magnetic anisotropy is fully in plane and parallel to the transverse direction. In contrast to the thicker films with a columnar structure (Fig. 3), at an early stage in the growth, the morphology of the thin film consists of small nuclei (Fig. 4), which can be isolated as well as in the form of clusters. Moreover, the nuclei are not circular, but exhibit an elongated shape following the transverse direction. Due to the low random magnetocrystalline anisotropy, the elongation of the nucleus can explain well the transverse orientation of the magnetic anisotropy in such films. On the other hand, in contrast to the tilted columns in the thicker film (Fig. 3), at low thickness the nuclei are parallel to the film plane. Consequently, no tilt is expected in the easy axis of thinner films as revealed by the torque measurements (Fig. 2). In the first stage of growth in oblique deposition, elongation of the shape of nuclei is one of the most important characteristics of the film microstructure.[3,7,14,15] For most metals such as Co, Ni, and Fe, elongation of the nuclei was observed with

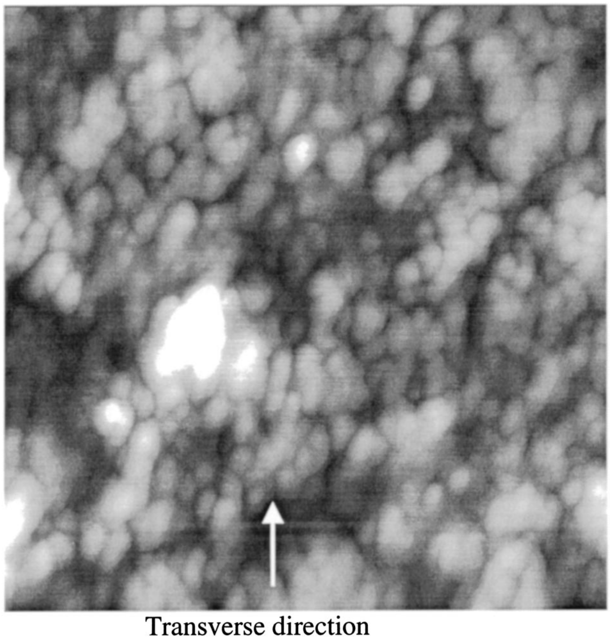

FIG. 4. $0.5 \times 0.5\ \mu m^2$ AFM picture of a very thin film (13 nm) showing a structure with nuclei at the first stage of growth. The nuclei exhibit an elongated shape parallel to the transverse direction.

the exception of Au, in which the nuclei adopt a circular shape rather than elongated. Because of the low affinity of Au for oxygen, an explanation based on the mobility of the adatoms was proposed for the elongation of the nuclei.[16] Due to the anisotropic supply of atoms (projection of the incidence beam in the film plane) and the isotropic supply of oxygen from the background gas, a mobility of adatoms can be induced around the nucleus.[16] The adatoms arriving at the nucleus will travel along its contour until the relative oxygen concentration exceeds a critical value where they will be trapped. Nevertheless, affinity to oxygen cannot explain recent observations on ultrathin films (few monolayers) of Co obliquely evaporated in ultrahigh vacuum ($<10^{-10}$ mbar).[17] The latter result reveals an elongated shape of the adatom structures with uniaxial anisotropy perpendicular to the incidence plane. This phenomenon was explained by steering,[18] which consists of a nonuniform distribution of the flux of the arriving atoms due to protrusions on the surface. To illustrate the importance of this effect in our case where the energy of the sputtered atoms is much higher (3 eV) than that of evaporation (0.15 eV),[17,18] we calculated the atom trajectories of

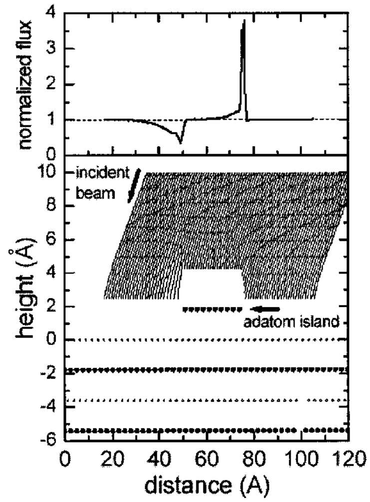

FIG. 5. Simulated trajectories of atoms in oblique incident beam (70°) with energy of 3 eV. The substrate surface consists of fcc Cu with a monolayer island on the top. The relative flux on the top of the island shows the steering effect with high deposition rate at the front of the island.

the energetic particles just before adsorption. The conditions of the simulation consist of a Cu surface with fcc structure (close to that of fcc Co) and a one-monolayer-high island on top. The interaction between the atoms of the incident beam (70°) and the surface is modeled by a pairwise Lennard-Jones potential.[18] As we can see in Fig. 5, the atoms approaching the surface with island structures are influenced by long-range attractive forces. Island structures induce a local distortion in the attractive potential, which creates a heterogeneous distribution of the incoming atoms close to the surface. An increase in flux at the front side and a decrease in flux at the backside is observed. This could be explained due to geometric shadowing. However, this would not lead to elongation. The decreased flux region behind the island is about 2 nm, which is 4 times larger than the 0.5 nm expected from geometric considerations. Also, on the island a similar flux increase is observed as noticed for the 0.15-eV particles. Exactly this latter flux increase is responsible for elongated structures[17,18] as the redistribution of these atoms will also be in the transverse direction. In most metals grown by oblique deposition, the elongated axis of the nuclei was found to be perpendicular to the incidence plane.[16,17] These results are in complete agreement with our microstructural analyses (Fig. 4) and support our magnetic measurements (Fig. 1).

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