Surface morphology and magnetic anisotropy of Fe/MgO(001) films deposited at oblique incidence

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We have studied surface morphology and magnetic properties of Fe/MgO(001) films deposited at an angle varying between 0° and 60° with respect to the surface normal and with azimuth along the Fe[010] or the Fe[110] direction. Due to shadowing, elongated grains appear on the film surface for deposition at sufficiently large angle. X-ray reflectivity reveals that, depending on the azimuthal direction, films become either rougher or smoother for oblique deposition. For deposition along Fe[010] the pronounced uniaxial magnetic anisotropy (UMA) results in the occurrence of “reversed” two-step and of three-step hysteresis loops. For deposition along Fe[110] the growth-induced UMA is much weaker, causing a small rotation of the easy axes.

Magnetic anisotropy of epitaxial films and its relationship to surface morphology have attracted much attention in recent years [1]. Both film properties are intimately related to the molecular beam epitaxy (MBE) deposition process. Oblique incidence deposition results via a self-shadowing effect [2, 3] in the formation of grains in the plane of the film that are elongated perpendicular to the incident flux direction and with aspect ratio increasing at larger deposition angle with respect to the surface normal [3]. Consequently, an in-plane uniaxial magnetic anisotropy (UMA) with easy axis perpendicular to the incident flux direction is induced during growth of the magnetic films [1, 2, 4]. UMA was found to play an important role in determining the magnetization reversal in thin films of cubic systems [7, 8]. Depending on the strength and orientation of the UMA, hysteresis curves with one, two and three steps are observed in various films at different field orientation. The appearance of the steps can be explained in terms of nucleation and propagation of domain walls (DWs) [8, 9]. Consequently, understanding the influence of oblique incidence growth is very important because of its ability to control both magnetic anisotropy and surface morphology [10, 11, 12].

Although shadowing effects have been studied in many magnetic systems, including Fe on MgO(001) [13] and Co on Cu(001) [14], the plane of oblique incidence was always kept parallel to the in-plane cubic easy axes of the magnetic layers. Here, we report on the influence of oblique deposition on the surface morphology and magnetic properties of Fe/MgO(001) films for deposition azimuth both along the Fe[010] and Fe[110] in-plane directions. Deposition along [010] turns out to be more effective in producing elongated grains and introducing UMA than deposition along [110]. A considerable growth-induced UMA along [010] induces the occurrence of “reversed” two-step and of three-step hysteresis loops.

Fe (001) oriented films were grown on MgO(001) substrates in an MBE system with base pressure below 1 × 10^{-10} mbar. The substrates were annealed for one hour at 700°C and held at 150°C during deposition. The films were deposited using an e-beam gun at a rate of 0.1 Å/s, as monitored by a quartz crystal oscillator. The incident Fe beam was at an angle varying between 0° to 60° with respect to the surface normal and with azimuth either along the Fe[010] or the Fe[110] direction. In Figs. 1(a) and 1(b) we present the surface topography measured in situ by scanning tunneling microscopy (STM) for Fe films grown at an incidence angle of 30° and with azimuth along [010] and [110], respectively. The film surface consists of approximately square grains about 15 nm in size. The grain edges have a preferred in-plane orientation along (010). When the deposition angle is increased to 60°, Fe surface grains for deposition along [010] are obviously elongated perpendicular to the incident flux direction with typical length of 15 nm and typical width of 3 nm (see Fig. 1(c)). The elongated grains are believed to result from a redistribution of the incident flux due long-range attractive forces [15, 16]. During evaporation the incident Fe atoms arrive preferentially on top of already formed grains rather than behind these grains. This shadowing effect is also observed for film deposition at 60° with azimuth along [110]. The Fe grains on the surface are rhombic in shape with typical length of 15 nm and typical width of 6 nm (see Fig. 1(d)). Based on the STM images we conclude that the self-shadowing effect can be neglected for a deposition angle below 30°. Moreover, the self-shadowing effect is anisotropic, i.e., depends on the azimuth of the incident flux direction. The surface grains for growth along [010] have a larger aspect ratio than the grains for deposition along [110].

The anisotropy of the shadowing effect is confirmed by ex situ X-ray reflectivity (XRR). Before removing the Fe/MgO(001) layers from the vacuum chamber, the layers are capped with a 4 nm thick protective Au layer. In Fig. 2(a) we present the X-ray data collected on a PANalytical X'Pert Pro diffractometer. When increas-
ing the deposition angle from 0° to 60°, the oscillating X-ray intensity is rapidly damped for the samples grown along [010], while the oscillations are slightly enhanced for the samples grown along [110]. By fitting the X-ray curves, we quantitatively obtain the root-mean-square (rms) roughness for both the lower interface between the Fe layer and the MgO substrate (see Fig. 2(b)) and the upper interface between the Fe layer and the Au layer (not shown). The roughness of both interfaces rapidly increases when increasing the deposition angle for the samples deposited along [010], but slightly decreases for the samples deposited along [110]. Thus, the shadowing effect is able to not only roughen but also smoothen Fe/MgO(001) films, depending on the deposition geometry. Although we used the same evaporation rate and the same nominal thickness (15 nm) when growing the Fe layers, the actual thickness obtained from the X-ray etry. Although we used the same evaporation rate and the same nominal thickness (15 nm) when growing the Fe layers, the actual thickness obtained from the X-ray simulations decreases from 14 nm to 8 nm when the deposition angle increases from 14 nm to 8 nm when the deposition angle increases from 0° to 60° (see Fig. 2(b)).

Due to the elongated shape of the Fe grains in the Fe/MgO(001) films an additional UMA is superimposed upon the intrinsic cubic anisotropy $K_1$ of Fe. Depending on the azimuthal angle, the growth-imposed UMA may have a component $K_{u1}$ along [010], a component $K_{u2}$ along [110], or both. When $K_{u1} < \angle K_1$ and $K_{u2} < K_1$, the component $K_{u2}$ rotates the position of the overall easy axes backwards with respect to the uniaxial hard axis over an angle $\delta$ that is given by \[
\delta = \frac{1}{2} \sin^2(K_{u2}/K_1) \]
[17], as illustrated in Fig. 2(c). The magnetic properties of the Fe/MgO(001) films were characterized \textit{ex situ} by the longitudinal and transverse magneto-optical Kerr effect (MOKE) for different field orientation $\phi$ as defined in Fig. 2(c).

For the category of samples grown with deposition angle $\leq 30^\circ$ and along [010] and for the samples with deposition angle $\leq 49^\circ$ and along [110], one-step and two-step hysteresis loops are measured at different $\phi$. For the typical loops presented in Figs. 3(a) and 3(b), which are obtained at $\phi = 15^\circ$ and $125^\circ$ for the film deposited at normal incidence, the switching routes that occur for increasing field are [100]→[010]→[010] and [010]→[100]→[010], respectively. The corresponding spin orientations are given by the arrows that are enclosed in a square in Fig. 3. For this category of samples, the switching fields for one-step ($H_{c1}$) and two-step ($H_{c1}, H_{c2}$) loops as a function of $\phi$ are all similar to the fields of the film deposited at normal incidence, which are presented in Fig. 4(a). From the symmetries of the coercive fields and the angles where one-step and two-step loops are observed, we infer that a weak uniaxial anisotropy is present along [010].

For the two samples deposited at an angle exceeding $49^\circ$ and along [010], we observe, apart from one step loops, “reversed” two-step, and three-step loops as well. The typical loops presented in Figs. 3(c) and 3(d) are obtained at $\phi = 95^\circ$ and $65^\circ$, respectively, for the sample deposited at $60^\circ$ and along [010]. The switching routes for increasing field are [010]→[100]→[010] and [010]→[100]→[010]→[010], respectively. Obviously, the magnetization $M$ for the “reversed” two-step loops rotates in the opposite direction (i.e., the switching route changes from clockwise to counter-clockwise or vice versa) when compared to the normal two-step loops observed in the same half quadrant of $\phi$. The $\phi$ dependence of the experimental switching fields of one-step ($H_{c1}$), “reversed” two-step ($H_{c3}, H_{c4}$), and three-step ($H_{c3}, H_{c4}, H_{c5}$) loops is symmetric about [010] for the two samples, as illustrated in Fig. 4(b) for the sample grown at $60^\circ$ and along [010]. The symmetries of the coercive fields and the angles where different loops occur, suggest that the extra UMA is oriented along [010].

For the sample deposited at $60^\circ$ and along [110], only one-step and two-step loops are observed at different $\phi$. The easy axes are found to slightly deviate from the cubic easy axes by $\delta = 3^\circ$. The magnetization does not switch exactly over $90^\circ$, but over $90^\circ \pm 25^\circ$. We link the presence of a component $K_{u2} \neq 0$ to the oblique deposition along [110]. The influence of $K_{u2} \neq 0$ also clearly emerges in the $\phi$ dependence of the experimental switching fields ($H_{c1}, H_{c2}$) presented in Fig. 4(c), which is symmetric about [110]. However, the range of angles $\phi$ where one-step loops occur, is not symmetric about either [110] or [010], which indicates that $K_{u1}$ cannot be neglected.

Recently, we have shown that both the $180^\circ$ and the $90^\circ \pm 25^\circ$ magnetic transitions, which occur in the hysteresis loops with different number of steps, are mediated by successive or separate $90^\circ \pm 25^\circ$ DW nucleations [18]. The UMA component $K_{u1}$ and the DW nucleation energies $\epsilon_{90^\circ \pm 25^\circ}$ can be evaluated by fitting the $\phi$ dependence of the experimental switching fields $[8, 17, 18]$. In Figs. 4(a) to 4(c) we present the fitting curves for three typical samples. The fitting parameters for all our Fe/MgO(001) films are plotted in Fig. 4(d). For films with $K_{u2} = 0$, i.e., $\delta = 0$, $\epsilon_{90^\circ \pm 25^\circ}$ remains constant at about 0.35 mT for a deposition angle $\leq 30^\circ$ (along [010] as well as along [110]), but increases to 0.79 mT for the deposition at $60^\circ$ and along [010]. This indicates that the DW nucleation energy is sensitive to the Fe layer thickness, but not the deposition geometry. The small enhancement of $\epsilon_{90^\circ}$ can be accounted for by the thickness reduction of Fe layers deposited at larger angle. For the sample grown at $60^\circ$ and along [110], the DW nucleation energy can take the two values $\epsilon_{90^\circ \pm 25^\circ}$ due to the shift of the easy axes resulting from $K_{u2} \neq 0$. For the set of samples deposited with azimuthal angle along [110], $K_{u1}/M$ is smaller than 0.1 mT. Such weak UMA cannot be avoided even for films deposited at normal incidence. We therefore assume that this small $K_{u1}$ results from the growth imposed by the MgO(001) substrate and not from the oblique deposition. The influence of the shadowing effect on $K_{u2}$ is also small. Only for the sample deposited at $60^\circ$ and along [110], a small $K_{u2}$, which is about 0.1 $K_1$, is observed experimentally. Changes in deposition geometry are clearly more effective for deposition along [010]. $K_{u1}/M$ slowly increases from 0.07 mT to 0.21 mT when increasing the deposition angle from $0^\circ$ to $30^\circ$, and then quickly rises to
2.70 mT at 49° and 7.45 mT at 60°. The stronger UMA obviously results from changes in the surface morphology, where clearly elongated grains start to be formed at a deposition angle between 30° and 49° for the growth along [010]. It is more difficult to impose an elongated grain structure for oblique deposition along [110] because of the preferential directions [100] and [010] for the growth of Fe grains on MgO(001).

For different ϕ the switching fields are predicted to be proportional to |K_u1 ± 90ϕ| [8, 9]. Moreover, the field orientation where different kinds of hysteresis loops can be observed, is determined by the condition tan ϕ < K_u1/ε90ϕ, where 0° < ϕ < 45° for the one-step loops and 45° < ϕ < 90° for the three-step loops, respectively [18]. Consequently, for samples deposited along [010] the coercive fields also rapidly increase due to the enhancement of K_u1 resulting from the oblique deposition. On the other hand, the range of ϕ for the occurrence of one-step and three-step loops becomes wider as predicted by theory. Since the observation of the “reversed” two-step and of the three-step switching events requires K_u1 > ε90ϕ, we conclude that the critical angle for deposition of Fe/MgO(001) films for which we can observe these two kinds of loops is between 30° and 49° for deposition along [010]. For deposition along Fe[110], the growth-induced UMA is much weaker, causing a small rotation of the easy axes.

FIGURE CAPTIONS

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FIG. 1. (Color online) STM images for Fe films deposited (a) at 30° and along [010], (b) at 30° and along [110], (c) at 60° and along [010], and (d) at 60° and along [110]. The size of the images is 200 nm x 200 nm for (a) and (b), and 100 nm x 100 nm for (c) and (d).

FIG. 2. (Color online) (a) XRR spectra for films deposited at 0° (black), 11° (red), 30° (green), 49° (blue), and 60° (magenta). The azimuthal angle for deposition is parallel to [010] (lower curves) and [110] (upper curves), respectively. (b) The rms roughness and the thickness for Fe layers deposited along [010] (squares) and along [110] (dots), as inferred from simulations of the XRR spectra. (c) Definition of the angles that are used to describe a film with in-plane cubic anisotropy and an additional UMA.

FIG. 3. (Color online) Longitudinal (||) and transverse (⊥) MOKE loops with (a) one step and (b) two steps, obtained at ϕ = 15° and 125° for the film deposited at normal incidence, and (c) “reverse” two-step and (d) three-step loops obtained at ϕ = 95° and 65° for the sample deposited at 60° and along [010]. The blue (red) curves are for applied fields varying from negative (positive) to positive (negative) saturation. The orientation of the Fe spins in the switching processes is represented by the arrows enclosed in a square.

FIG. 4. (Color online) The experimental switching fields H_{c1} (Ο), H_{c2} (□), H_{c3} (▲), H_{c4} (Ο), and H_{c5} (△) as a function of ϕ, and the corresponding theoretical curves for the samples deposited (a) at normal incidence, (b) at 60° and along [010], and (c) at 60° and along [110]. (d) The fitting parameters K_u1 (Ο), ε_{90ϕ} (□), ε_{90ϕ−25} (▲), and ε_{90ϕ+25} (△) for samples grown along [010] (upper panel), and along [110] (lower panel).
(a) Intensity (a. u.) vs. Growth angle (deg) with various thicknesses.

(b) Roughness (nm) and Thickness (nm) vs. Growth angle (deg).

(c) Diagram showing the c-axis orientation and growth angle (θ) with respect to the [010] and [100] directions.
