Magnetic anisotropies of epitaxial Fe/MgO(001) films with varying thickness and grown under different conditions

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\textbf{Abstract.} Fe films with thickness varying between 5 and 100 nm were grown on flat MgO(001) substrates while rotating the substrates. Hysteresis loops with one step and two steps were observed and interpreted in terms of a magnetization reversal mechanism with either two successive or two separate 90° domain wall (DW) nucleations, respectively. This recently introduced, novel mechanism for 180° magnetic transitions was used to quantitatively evaluate both the uniaxial magnetic anisotropy (UMA), which accompanies the intrinsic fourfold in-plane magnetic anisotropy, and the DW nucleation energy. The strength of both the UMA and the DW nucleation energy turns out to be inversely proportional to the thickness of the Fe layers for Fe/MgO(001). This suggests that the extra UMA and the DW nucleation/propagation for Fe layers are pure interface related effects. By comparing six 15 nm thick Fe/MgO(001) films deposited under different conditions, it is found that these interface related effects originate from the presence of atomic steps due to the substrate miscut and from the presence of strain relaxation resulting from lattice mismatch.
1. Introduction

Epitaxial growth of Fe layers on MgO substrates continues to be of great interest from an application as well as a fundamental point of view [1]. The Fe/MgO(001) system is an excellent candidate to investigate the correlation between atomic structure and magnetism in low-dimensional heterostructures [2]–[4]. Moreover, Fe/MgO/Fe magnetic tunnel junctions have been extensively studied since this type of junction was predicted to have a very high tunneling magnetoresistance [5].

The magnetic anisotropies of Fe/MgO(001) are crucial when using such Fe films for spintronic applications. Due to the crystalline symmetry of bcc Fe, the epitaxial Fe/MgO(001) films usually exhibit an intrinsic fourfold in-plane magnetic anisotropy. The fourfold anisotropy is accompanied by an additional uniaxial magnetic anisotropy (UMA) [6]. The latter component of the anisotropy is a common feature for ferromagnetic films grown on various substrates and has been attributed to different origins. For example, the extra UMA observed in Fe/GaAs(001) was attributed to the uniaxial nature of dangling bonds at the substrate surface [7]. The UMA for Fe films grown on a stepped substrate was explained in terms of the Néel surface effect [8, 9]. A growth-induced UMA was observed for Fe/MgO(001) prepared by means of oblique-incidence deposition [10]. Recently, we successfully relied on ion sputtering to modulate the surface morphology and manipulate the strength and the orientation of the UMA in Fe/MgO(001) [11]. Durand et al [12] found that the extra UMA for Fe on MgO(001) can be removed by rotating the substrate during growth and interpreted the UMA for Fe/MgO(001) in terms of a volume effect rather than in terms of an interface effect because of the absence of a systematic variation of the anisotropy with film thickness. On the other hand, the extra UMA for Fe/GaAs(001) films and Fe films on vicinal surfaces is a linear function of the inverse Fe layer thickness, suggesting that the UMA mainly results from an interface contribution [13]–[16].

In magnetic data storage and spintronic devices, information is stored in the magnetic moment of the film. Therefore, the ability to control the magnetization reversal is a key factor for successful magnetic memory applications. The magnetic switching process in thin films was shown to be sensitive to the specific orientation and strength of the UMA. Depending on the field orientation, hysteresis curves with one, two or three steps were observed in Fe films on various substrates [17]–[20]. Until recently, all of the measured hysteresis loops for the Fe/MgO(001) system only revealed one or two steps [6, 10]. By depositing Fe layers onto MgO(001) substrates at sufficiently large incidence angle of the Fe beam with respect to the surface normal, which induces a pronounced UMA, we succeeded in observing for the first time three-step hysteresis loops for Fe/MgO(001) [21, 22]. Cowburn et al explained the multi-step magnetic switching processes in ultrathin epitaxial Fe films in terms of nucleation and
propagation of 90° and 180° domain walls (DWs). They developed a phenomenological model to account for the angular dependence of the switching fields by considering a nucleation energy $\epsilon_{90^\circ}$ for 90° DWs and a nucleation energy $\epsilon_{180^\circ}$ for 180° DWs \cite{19, 20}. Recently, we found that 180° DW nucleation fails to provide a consistent quantitative explanation for 180° magnetic switching in Fe/MgO(001) films and we introduced a new phenomenological model based on two successive 90° DW nucleations to describe the variation of the switching field for the 180° magnetic transitions \cite{22}.

Magnetic anisotropy can be experimentally measured by Brillouin light scattering (BLS) and ferromagnetic resonance (FMR) \cite{3, 16}. However, since the strength $K_u$ of the extra UMA is very weak for Fe/MgO(001), the UMA values inferred from BLS and FMR have very large error bars \cite{14, 23}. On the other hand, model-based fitting of the angular dependence of the switching fields is able to provide reliable values for the UMA and the DW nucleation energies \cite{19, 24}. Understanding the thickness dependence of the UMA and of the DW nucleation energies for Fe/MgO(001) is important for the design and fabrication of spin-based devices. From a fundamental point of view it is important to identify the origin of the UMA and of the DW pinning. Here, we report on the use of our recently introduced novel mechanism for 180° magnetic switching with either two successive or two separate 90° DW nucleations to evaluate $K_u$ and $\epsilon_{90^\circ}$ for Fe/MgO(001) films grown while rotating the substrates \cite{22}. Both $K_u$ and $\epsilon_{90^\circ}$ turn out be inversely proportional to the thickness of the Fe layers, which suggests that the extra UMA and the DW pinning are pure interface related effects caused by strain relaxation and by steps on the surface. We also compared the magnetic behavior of Fe/MgO(001) films having the same thickness (15 nm), but deposited under different conditions. Relying on the influence of the deposition conditions we are able to discriminate between the ‘volume’ and interface related contributions to the anisotropy.

2. Experimental

Fe films were grown by molecular beam epitaxy (MBE) on MgO(001) substrates supplied by Crystal GmbH. The base pressure of the MBE chamber was below $3 \times 10^{-10}$ mbar. The substrates were first annealed at 700°C during 1 h and held at 150°C during deposition. Fe films were deposited using an electron beam gun at a rate of 0.1 Å s$^{-1}$, as monitored by a calibrated quartz crystal oscillator. The thicknesses determined by this method are accurate to within 10%. The incident Fe beam in the MBE chamber was not perpendicular to the substrate, but at an angle of 30° with respect to the surface normal. In order to avoid growth-induced anisotropy, Fe films with thickness $t_{Fe}$ ranging from 5 to 100 nm were deposited while rotating the substrate. The Fe layers grow according to the well-known Fe(001)[110]∥MgO(001)[100] epitaxial relation with a possible tetragonal distortion caused by the relatively small lattice mismatch \cite{25, 26}. The substrates we used are square with the edges parallel to the MgO ⟨100⟩ direction. Therefore, the epitaxial Fe films have the Fe⟨100⟩ orientation along the in-plane diagonal of the substrates, as illustrated in figure 1(a). In order to further investigate the origin of the extra UMA, we cut three Mg(001) substrates into two halves, and positioned the two halves as illustrated in figure 1(b).

We deposited Fe layers with thickness $t_{Fe} = 15$ nm on the three substrates using three different modes: while rotating the substrate and without rotation, fixing the azimuth of the incoming Fe beam either along the Fe⟨100⟩ or along the Fe⟨110⟩ direction.

The surface topography of the samples was characterized \textit{in situ} by scanning tunneling microscopy (STM). The magnetic properties of Fe/MgO(001) films were measured \textit{ex situ}
Figure 1. The crystalline directions of epitaxial Fe films grown (a) on a square MgO(001) substrate and (b) on two halves of one MgO(001) substrate, which has been cut into two and where the two halves are fixed perpendicularly to each other. For the non-rotation growth modes, the azimuth for the Fe beam is either along the Fe⟨100⟩ or along the Fe⟨110⟩ direction. STM images (200 nm × 200 nm) for Fe layers deposited (c) in rotation mode and with $t_{Fe} = 15$ nm, (d) at an incidence angle of 30° and with azimuth along the Fe⟨110⟩ direction, and with $t_{Fe} = 15$ nm, and (e) in the rotation mode and with $t_{Fe} = 100$ nm. The color contrast (dark to bright) corresponds to 1.4, 3.2 and 3.7 nm for (c), (d) and (e), respectively.

by combined measurements of the longitudinal and transverse magneto-optical Kerr-effect (MOKE) for different field orientations $\phi$, which is defined as the angle between the applied field $H$ and the Fe⟨100⟩ direction (see figure 1(a)). Protective Au layers with a thickness of 4 nm were deposited on top of the Fe films before removing the Fe films from the MBE chamber.

3. Results and discussion

The STM images in figures 1(c) and (d) indicate that a 15 nm thick Fe film grown while rotating the substrate appears as a continuous layer consisting of coalesced small islands about 10 nm in size. On the other hand, the surface of the Fe layer with the same thickness but deposited at an incidence angle of 30° and with azimuth along the Fe⟨110⟩ direction consists of square islands about 15 nm in size with the edges oriented preferentially along the Fe⟨100⟩ direction. The corresponding root-mean-square (rms) roughnesses are 0.125 and 0.486 nm for the surfaces shown in figures 1(c) and (d), respectively. The STM results are consistent with previous studies [11, 27]. The latter studies revealed that the initial growth of Fe on MgO(001) occurs according to the Volmer–Weber mechanism, due to the higher surface free energy of Fe(001) when compared with MgO(001). With increasing Fe film thickness, the three-dimensional Fe
islands coalesce into a continuous layer [28, 29]. Furthermore, our STM results for the samples grown under different conditions suggest that the rotation mode for deposition is able to suppress the three-dimensional Volmer–Weber growth of Fe on MgO(001) and reduce the roughness. On the other hand, the three-dimensional growth is enhanced due to strain relaxation when increasing $t_{Fe}$. The grain size as well as the roughness increase and the grain shape becomes round rather than being square, as illustrated in figure 1(e) for an Fe layer with $t_{Fe} = 100$ nm grown in the rotation mode, where the corresponding grain size and the rms roughness are about 30 and 0.573 nm, respectively.

For all of the Fe layers either one-step or two-step loops are observed when varying the angle $\phi$, as illustrated by the typical loops in figures 2(a) and (b) that have been obtained by longitudinal and transverse MOKE measurements. The two kinds of hysteresis loops, which were also observed for many other ferromagnetic films with cubic crystal structure, can be understood in terms of a combination of the intrinsic cubic anisotropy $K_1$ and the extra UMA $K_u$ [11, 19]. The orientation and the strength of the UMA can be determined with high precision by fitting the angular dependence of the switching fields. We measured the switching fields for the one-step (we refer to this switching field as $H_{c1}$) and for the two-step (we refer to the lower switching field as $H_{c1}$ and to the higher switching field as $H_{c2}$, respectively) loops as a function of the angle $\phi$ at 5° interval for the Fe layers with different thicknesses, and obtained a behavior similar to the one presented in figure 2(c) for the layer with $t_{Fe} = 25$ nm. The switching fields have a dependence on $\phi$ that is symmetric about the ⟨100⟩ direction, and the angular dependence of $H_{c1}$ reveals a clear, abrupt step when crossing the ⟨110⟩ direction, which confirms the presence of an extra UMA parallel to the ⟨010⟩ direction. Moreover, the cubic easy axis where the one-step loops occur, i.e. the [100] direction corresponding to $\phi = 0^\circ$ in figure 2(c), is also the easy axis for the UMA. In the same way, the cubic easy axis where the two-step loops occur, i.e. the [010] direction ($\phi = 90^\circ$), is the hard axis for the UMA [19].

We ascribe the UMA in Fe/MgO(001), which was always observed along the Fe easy axes in previous works, to the preferential orientation of the Fe grain edges along the ⟨100⟩ direction on the surface. Obviously, the rotation mode for the Fe film growth is not able to remove the additional UMA, which disagrees with the results reported in [12].

In order to properly fit the $\phi$ dependence of the switching fields and evaluate the UMA, the physical mechanisms dominating the magnetization reversal need to be known. Previously, the occurrence of one-step and of two-step loops was interpreted in terms of 180° DW nucleation and in terms of two 90° DW nucleations, respectively [19]. However, such a mechanism is not able to properly describe the variation of the switching fields for the one-step loops (180° magnetic transitions). As illustrated in the inset to figure 2(c), the experimentally observed $H_{c1}$ reveals the presence of a peak when $H$ is applied along the Fe[100] direction ($\phi = 0^\circ$), while the model with 180° DW nucleation predicts a minimum at this angle.

Taking into account the dependence of the magnetic energy on $H$, we are able to understand why 180° DW nucleation cannot account for the 180° magnetic transition in the one-step loops, and are able to identify the appropriate magnetization reversal mechanism. Since the additional UMA in the Fe layers is along the [010] direction, the total energy can be written as $E = (K_1/4)\sin^2 2\theta + K_u\sin^2 \theta - M H \cos (\phi - \theta)$, where $\theta$ is the angle between the [100] direction and the magnetization vector $M$. A detailed quantitative derivation of the relevant switching fields based on minimizing the system energy was presented in [22]. Here, we rely on a more intuitive picture based on the energy landscape resulting from the magnetic anisotropies in our Fe/Mg(001) films. The relevant energy landscapes are presented in figure 3. According
Figure 2. Longitudinal (∥) and transverse (⊥) MOKE loops with (a) one step and (b) two steps, obtained at $\phi = 0^\circ$ and $\phi = 60^\circ$, respectively, for an Fe/MgO(001) film ($t_{Fe} = 25$ nm) grown in the rotation mode. (c) The experimental switching fields $H_{c1}$ (dots) and $H_{c2}$ (squares) as a function of the field orientation $\phi$ and the corresponding theoretical curves for $H_{c1}$ (red), $H_{c2}$ (blue), the ‘virtual’ $H_{c2}$ (dashed gray) and $H_c$ (green).

to Cowburn et al [19], the energy difference $\Delta E_{180^\circ \rightarrow 0^\circ}$ between Fe magnetized along the $[\bar{1}00]$ and $[100]$ directions is $\epsilon_{180^\circ}$ when the applied field direction approaches the $[100]$ direction within the range of angles $0^\circ < \phi < 45^\circ$. This corresponds to a critical (switching) field $H_c = \epsilon_{180^\circ}/[2M|\cos \phi|]$ (see the green curve in the inset to figure 2(c)). Figure 3(a) schematically illustrates the corresponding energy landscape in the single-domain state as a function of the angle $\theta$. The previously adopted model assumes that the magnetization switches directly from $[100]$ to $[100]$ via $180^\circ$ DW nucleation [19]. Within this model the energy difference between $180^\circ$ and $0^\circ$ is treated in terms of one single barrier without considering the intermediate
Figure 3. Energy landscapes of the Fe layer with the extra UMA along [010] (a) at $H_c$ and (b) at $H_{c1}$ when the field orientation $\phi$ is close to [100] and $0^\circ < \phi < 45^\circ$, and (c) at $H_{c1}$ and (d) at $H_{c2}$ when the field orientation $\phi$ is close to [110] and $0^\circ < \phi < 45^\circ$.

energy minimum at $\theta = 90^\circ$. From figure 3(a) it is, however, clear that in the energy landscape the switching between $\bar{[100]}$ and $[100]$ is governed by two separate energy barriers between $\bar{[100]}$ and $[010]$ and between $[010]$ and $[100]$, respectively. Based on the previous conclusion $\epsilon_{180} = 2\epsilon_{90}$ [19, 20], the relative energy gains are $\Delta E_{180\rightarrow 90} < \epsilon_{90}$ and $\Delta E_{90\rightarrow 0} > \epsilon_{90}$ at $H_c$. Since $\Delta E_{180\rightarrow 90}$ is not sufficiently large to overcome the energy barrier $\epsilon_{90}$ at $H_c$, the magnetization vector can therefore not switch from $\bar{[100]}$ to $[010]$ via $90^\circ$ DW nucleation. When $H = H_{c1}$, where $H_{c1} = (\epsilon_{90} + K_u)/[M(\cos \phi + \sin \phi)]$, the relevant energies are changed to $\Delta E_{180\rightarrow 90} = \epsilon_{90}$ and $\Delta E_{90\rightarrow 0} > \epsilon_{90}$, as illustrated in figure 3(b). Consequently, the domains aligned along the intermediate axis $[010]$ nucleate at $H_{c1}$. Since $\Delta E_{90\rightarrow 0}$ already exceeds $\epsilon_{90}$ at $H_{c1}$, the domains along $[010]$ are unstable and cannot grow. As a result, a second nucleation of domains along the final $[100]$ remanent direction simultaneously occurs at $H_{c1}$. The hysteresis loops therefore only reveal one single step (see figure 2(a)). We refer to such a switching process as two successive $90^\circ$ DW nucleations. In a similar way, the two-step loops can be described by considering the two separate energy barriers. When the applied field direction approaches the $[110]$ direction within the range of angles $0^\circ < \phi < 45^\circ$, $\Delta E_{90\rightarrow 0}$ is still smaller than $\epsilon_{90}$, when $\Delta E_{180\rightarrow 90}$ reaches $\epsilon_{90}$ at $H_{c1}$, as illustrated in figure 3(c). In this case the magnetization jumps from $[\bar{100}]$ to [010] via $90^\circ$ DW nucleation. When further increasing $H$ to $H_{c2}$, where $H_{c2} = (\epsilon_{90} - K_u)/[M(\cos \phi - \sin \phi)]$, $\Delta E_{90\rightarrow 0} = \epsilon_{90}$ and this results in a magnetic switching from [010] to [100] via a second $90^\circ$ DW nucleation, as
illustrated in figure 3(d). This gives rise to hysteresis loops with two distinct jumps at $H_{c1}$ and $H_{c2}$. Moreover, the domain along 90° is stable at $H_{c1} < H < H_{c2}$, and appears as a clear plateau between the two jumps (see figure 2(b)). For the switching mechanism with two successive DW nucleations, which corresponds to one single jump in the hysteresis loop, $H_{c2}$ is no longer relevant for fitting the experimental results, but still indicates that $\Delta E_{90\rightarrow0°} = \epsilon_{90°}$. Whether the magnetization reversal occurs via two successive or via two separate 90° DW nucleations is determined by the relative magnitude of the critical fields $H_{c1}$ and $H_{c2}$. We conclude that for $H_{c1} > H_{c2}$ one-step loops with two successive 90° DW nucleations dominate the magnetic switching. On the other hand, for $H_{c1} < H_{c2}$ two-step loops appear with two separate 90° DW nucleations. Consequently, the critical value for the angle $\phi$, which separates the appearance of the two kinds of loops, can be derived from $H_{c1} = H_{c2}$ and is given by $\tan \phi = K_u/\epsilon_{90°}$ for $0° < \phi < 45°$.

In order to verify the validity of the proposed magnetization reversal mechanism, we fit the switching fields for one-step loops by the theoretical equation for $H_{c1}$ and for two-step loops by the equations for $H_{c1}$ and $H_{c2}$ [22]. The switching fields can be consistently predicted for the full range of angles $\phi$, with only one set of $\epsilon_{90°}$ and $K_u$ values, as illustrated in figure 2(c). In particular, the maximum in the switching field around $\phi = 0°$ can be nicely fitted. For the range of angles $\phi$ where the one-step loops occur, we have added to figure 2(c) the ‘virtual’ field $H_{c2}$ by relying on the same set of fitting parameters. Obviously, the one-step and the two-step loops occur according to our model for $H_{c1} > H_{c2}$ and for $H_{c1} < H_{c2}$, respectively.

It should be noted that the direct transition via 180° DW nucleation (green curve in figure 2(c)) predicts a lower switching field than the switching field corresponding to our reversal mechanism with two successive 90° DW nucleations (red curve in figure 2(c)). In principle, the magnetization should reverse by the mechanism corresponding to the lower coercivity. However, as indicated by the quality of the fitting in the inset of figure 2(c), our experimental results reveal that the magnetization reversal mechanism is clearly not the 180° DW nucleation (green curve), although this mechanism predicts a lower coercivity. This can be linked to the fact that 180° DW nucleation treats the energy difference between 180° and 0° in terms of one single barrier without considering the energy minimum at 90°. Taking into account the energy minimum at 90°, our approach with two successive 90° DW nucleations is able to provide a much better fit of the experimental data. This is not because the corresponding switching of the magnetization is energetically more favorable, but because it appropriately takes into account the extra energy minimum at 90°.

The fitting parameters $K_u$ and $\epsilon_{90°}$ for Fe/MgO(001) films with $t_{Fe}$ varying between 5 and 100 nm have been collected in figure 4. $K_u$ can be decomposed into a volume contribution $K_u^V$ and an interface contribution $K_u^I$ according to $K_u = K_u^V + K_u^I/t_{Fe}$ [30]. Since a DW can be nucleated at and pinned by impurities and defects located within the volume and/or at the interface, $\epsilon_{90°}$ can in a similar way also be expressed as $\epsilon_{90°} = \epsilon_{90°}^V + \epsilon_{90°}^I/t_{Fe}$. A fitting based on such a decomposition reveals that $\epsilon_{90°}$ and $K_u$ are inversely proportional to $t_{Fe}$ with the surface related parameters $K_u^I/M = 2.21$ mT and $\epsilon_{90°}^I/M = 4.42$ mT. Within the experimental error the volume contributions are zero, which indicates that the UMA and the DW nucleation/propagation for our Fe layers are pure interface related effects. $K_u^V$ and $\epsilon_{90°}^V$ consist of contributions from the interface between the Fe films and the substrate and from the interface between the Fe films and the Au capping layers.

Atomic steps due to the miscut of the substrate and the strain resulting from the lattice mismatch between the Fe film and the substrate are possible mechanisms, which can give rise to

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Figure 4. Dependence of $K_u$ (dots) and $\epsilon_{90^\circ}$ (squares) on the Fe film thickness $t_{Fe}$ for the Fe/MgO(001) films grown in the rotation mode, as obtained from our analysis of the $\phi$ dependence of the switching fields. The full curves correspond to a $1/t_{Fe}$ dependence.

a magnetic anisotropy and energy that are inversely proportional to the Fe film thickness [16]. Since the extra UMA is very small, a miscut, which is even less than 10′ for our MgO(001) crystal substrates, has to be taken into account. On the other hand, the crystal lattice distortion results in strain in epitaxial films, giving rise to magnetoelastic anisotropy. The strain is relaxed via the formation of dislocations with increasing film thickness, and these dislocations can then give rise to pinning of DWs.

We estimate the contributions related to atomic steps and to strain by comparing the three pairs of samples with $t_{Fe} = 15$ nm grown in different modes. Each pair of samples has a collinear UMA along an Fe easy axis, i.e. [100] for part I and [010] for part II of the MgO substrate (see figure 1(b)), with an UMA strength that is listed in table 1. We ascribe the clear difference in UMA to a competition between the contributions related to atomic steps and to strain. We expect that for our Fe layers the strain is sensitive to the substrate heating and other deposition conditions in the MBE growth chamber. The deposition conditions should be similar for the two parts of the MgO substrates, implying the strain-related UMAs occur along perpendicular crystallographic directions for the two parts. On the other hand, the miscut for a single crystal is oriented along a well-defined crystallographic direction, implying the step-induced UMA for parts I and II will be along the same crystallographic direction. Comparing the UMA for the three pairs of samples, we come to the following consistent interpretation of the experimental results: (i) the strain-induced UMA is along the [100] direction and along the [010] direction for parts I and II, respectively; (ii) the strain-related contribution to the UMA is slightly larger than the miscut-related contribution; (iii) when the two contributions are perpendicular, the contributions partially cancel and the UMA strength is reduced, which is the case for the part I; (iv) when the two contributions are collinear, the UMA strength is enhanced, which
is the case for the part II. Our results for the samples grown without rotating the substrate reveal that oblique deposition at an angle of 30° and with azimuth along the ⟨100⟩ direction is able to induce an UMA of about 0.10 mT. On the other hand, oblique deposition with azimuth along the ⟨110⟩ direction has almost no influence on the UMA. This dependence on the oblique deposition conditions was the subject of a previous systematic study\cite{21}. The growth-induced UMA is easy to control, which can be used to compensate the contribution related to the strain or to the miscut. This implies that we can obtain Fe/MgO(001) films with pure cubic anisotropy such as the sample grown at 30° and with azimuth along Fe[100] (i.e. part I). The UMA related to the strain and to the miscut may either be enhanced or canceled for different relative orientations of the two contributions. On the other hand, the dislocations resulting from the strain relaxation and the steps caused by the miscut, which both act as pinning centers for DW nucleation and propagation, do not depend on the orientation. In order to obtain the DW nucleation energy, one can simply add the nucleation energies for the two contributions. Consequently, the parts I and II that are positioned perpendicularly during deposition, have the same DW nucleation energy $\epsilon_{90°}$, which we determine to be about 0.35 mT for all six samples listed in table 1. Based on our experimental results, it is clear that the DW nucleation energy $\epsilon_{90°}$ is mainly determined by the film thickness $t_{Fe}$, regardless of the strength $K_u$ of the UMA. This conclusion is also confirmed by our recent related work. As indicated in\cite{21}, for films with the same nominal thickness but deposited at different oblique incidence angles and with azimuth along Fe[010], $K_u$ rapidly increases with increasing deposition angle, while $\epsilon_{90°}$ remains almost constant.

Our present results confirm that all 180° magnetic transitions in the Fe/MgO(001) system can be consistently described in terms of two successive 90° DW nucleations. We demonstrated before that a 180° magnetic transition also occurs for the second step of the three-step hysteresis loops in Fe/MgO(001) films that are deposited at oblique incidence at a sufficiently large angle with respect to the MgO substrate normal\cite{21}. Moreover, we expect that the validity of our model will not be restricted to specific ferromagnetic materials and to specific substrates. The magnetization reversal mechanism with two successive 90° DW nucleations should be applicable as well for similar systems, including both ferromagnetic films and magnetic semiconducting films on various cubic substrates. We note that in previous works on films with cubic symmetry the range of angles for which the one-step loops or the three-step loops occur was quite small, and the magnitude of the switching fields for 180° magnetic transitions relatively low. Consequently, the maximum that occurs for the dependence on the field orientation of the switching field around the cubic easy axes (see inset in figure 2(c)) may be hard to observe experimentally and may easily be overlooked. Nevertheless, we note that, e.g., the experimental results presented in\cite{31} for Ni/MgO(001) and in\cite{32} for Fe/GaAs(001) confirm the occurrence of a maximum that is consistent with our model for magnetization reversal with two successive 90° DW nucleations. Finally, we note that ultra-fast time-resolved

Table 1. $K_u$ for the three pairs of Fe/MgO(001) films ($t_{Fe} = 15$ nm) grown in three different modes (see text).

| Sample part | $K_u$ (rotation) | $K_u$ (along Fe[010]) | $K_u$ (along Fe[010]) |
|-------------|-----------------|----------------------|----------------------|
| I           | 0.04 ± 0.01 mT  | 0.00 ± 0.01 mT       | 0.03 ± 0.01 mT       |
| II          | 0.10 ± 0.01 mT  | 0.21 ± 0.02 mT       | 0.13 ± 0.01 mT       |
MOKE measurements may be able to directly detect the intermediate domain formation that is involved in the process of the two successive DW nucleations. Indeed, the DW nucleation and propagation are perturbed by defects and roughness [33], implying magnetic switching is not as sharp as predicted by theory.

4. Conclusions

In summary, we have investigated Fe films with thicknesses varying between 5 and 100 nm and grown on flat MgO(001) substrates while rotating the substrates. For all samples hysteresis loops with one step and two steps were observed at different field orientations, which indicates that an additional UMA is superimposed on the intrinsic fourfold in-plane magnetic anisotropy and this extra UMA cannot be removed by rotating the substrate during the Fe film growth. The magnetization reversal mechanism for Fe/MgO(001) is explained in terms of a model with either two successive or two separate 90° DW nucleations. We rely on an intuitive description that is based on the energy landscape of a system with combined cubic anisotropy and an extra UMA in order to understand why the magnetization reverses by two successive 90° DW nucleations and why the previously introduced mechanism with 180° DW nucleation fails to account for the experimentally observed 180° magnetic transition. Both the UMA and the DW nucleation energy, which are evaluated using our phenomenological model, turn out to be inversely proportional to the thickness of the Fe layers, which suggests that the extra UMA and the DW nucleation/propagation for the Fe layers are pure interface related effects. By comparing six 15 nm thick Fe/MgO(001) films deposited under different conditions, we have moreover found that these interface related effects originate from the presence of atomic steps due to the substrate miscut and from strain relaxation resulting from lattice mismatch.

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