Magnetic uniaxial anisotropy of Fe films grown on vicinal Ag(001)

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Step-induced magnetic anisotropy was investigated in Fe films grown on a vicinal Ag(001) substrate using the surface magneto-optic Kerr effect technique. We found that the step-induced magnetic anisotropy is independent of the Fe film thickness up to 61 ML. Step decoration with Ag or Pd atoms also shows no effect on the step-induced magnetic anisotropy. These results indicate that the step-induced magnetic anisotropy in this system does not localize at the step edges. We suggest that strain inside the Fe film due to the large lattice mismatch between fcc Ag and bcc Fe in the normal direction of the film is the dominant contribution to the step-induced magnetic anisotropy in this system.

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I. INTRODUCTION

Understanding the nature of magnetic anisotropy[1] is crucial to the development of low-dimensional magnetic structures. Magnetic anisotropy is characterized by its strength and functional form. Determining the strength of the magnetic anisotropy requires detailed knowledge of the band structure of the materials,[2] and it remains as a challenge to date for theorists to calculate the strength of the magnetic anisotropy from first principles. The functional form of the magnetic anisotropy, however, depends only on the lattice symmetry. Magnetic thin films offer good experimental systems for studying magnetic anisotropy because translational symmetry is naturally broken in the surface normal direction. After great progress in the study of magnetic surface anisotropy, recent research has been extended to the study of in-plane uniaxial magnetic anisotropy. Based on a symmetry argument, it is easy to show that the in-plane uniaxial magnetic anisotropy cannot exist if the surface normal is an n-fold rotation axis with $n>2$ (e.g., triangle and square lattices).[3]

In an effort to understand the role of lattice symmetry breaking in magnetic anisotropy, experiments have been performed on magnetic thin films grown on vicinal (001) surfaces. The atomic steps on the surface break the fourfold rotational symmetry and thus induce a uniaxial magnetic anisotropy within the film plane. This kind of step-induced magnetic anisotropy was first observed indirectly by ferromagnetic resonance[4] and domain imaging,[5] and then directly by magnetic hysteresis loop measurements.[6,7] With the availability of curved substrates, it is now possible to systematically explore the relationship between step-induced magnetic anisotropy and step density.[8,9] It is now clear that (1) step-induced universal magnetic anisotropy exists in many systems, suggesting that this is a universal property; (2) the easy magnetization axis can be either parallel or perpendicular to the step edges, showing that corrugation-induced shape anisotropy is not the dominant mechanism; (3) changing the step density significantly changes the strength of the magnetic anisotropy; and (4) a simple model based on Néel's pair-bonding model[10] can correctly describe the dependence of the magnetic anisotropy on the step density.

While there has been much activity in applying step-induced magnetic anisotropy to study other important magnetic phenomena such as the spin reorientation transition,[11,12] magnetic interfacial frustration,[13] and magnetic switching,[14,15] the origin of step-induced magnetic anisotropy remains obscure at the microscopic level.[16,17] One of the puzzles is the unusual thickness dependence of the step-induced magnetic anisotropy. If the step-induced anisotropy were localized at the step edges, a $1/d$ thickness dependence of the magnetic anisotropy would be expected, where d is the film thickness. To the best of our knowledge, none of the studied systems shows a simple $1/d$ behavior. In fact, the thickness dependence is either rather weak[18] or even oscillating as a function of film thickness.[7] Obviously, a detailed study of step-induced magnetic anisotropy is needed in order to better understand its microscopic origin. In this paper, we report a detailed study of step-induced magnetic anisotropy in the Fe/Ag(001) system. We found that the step-induced magnetic anisotropy is independent of the Fe film thickness up to 61 ML. In addition, we found little effect of step decoration with Pd or Ag atoms on to the step edges of the Fe films. These results show that the step-induced anisotropy in the normal direction of the film in this system is strongly nonlocal.

II. EXPERIMENT

A 10-mm-diameter Ag(001) single-crystal disk was polished mechanically into a curved shape so that the vicinal angle changes continuously across the substrate from 0 to $\sim$20°. The step edges are parallel to the [110] direction of the Ag(001) surface so that the Fe(001) overlayer has step edges parallel to its [110] direction. The substrate was polished mechanically down to a 0.25-μm diamond-paste finish, and then chemical polishing[19] was performed to remove the scratches left from the mechanical polishing. The substrate was further cleaned *in situ* by cycles of Ar^+ sputtering at $\sim$2 keV and annealing at $\sim$600 °C. After this treatment, a clean and well-defined Ag(001) surface is formed as indicated by low-energy electron diffraction (LEED). Fe films were grown epitaxially on the Ag substrate at room temperature with the growth chamber pressure below 4×10^{-10} Torr. The

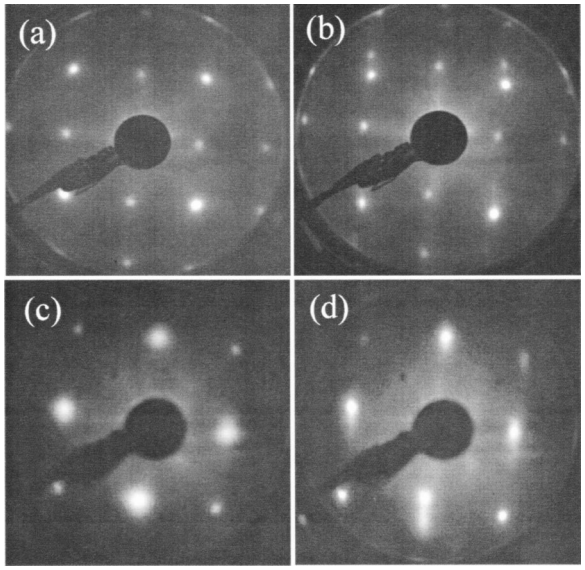

FIG. 1. LEED patterns from (a) flat Ag(001), (b) stepped Ag(001) with ~11° vicinal angle, (c) 25 ML Fe grown on flat Ag(001), and (d) 25 ML Fe grown on stepped Ag(001) with ~11° vicinal angle. The electron energy is ~137 eV in (a) and (b) and ~94 eV in (c) and (d).

deposition rate was ~0.5–1.0 Å/min and was monitored by a quartz thickness balance.

The magnetic properties of the Fe films were measured *in situ* by the surface magneto-optic Kerr effect (SMOKE) using a He-Ne laser (632.8 nm, beam diameter ≈0.2 mm). As the SMOKE laser beam scans across the sample to measure hysteresis loops, its reflection angle simultaneously determines the local vicinal angles at different positions of the substrate. Therefore, the relationship between the step-induced magnetic anisotropy and the step density is obtained systematically from a single curved substrate. Due to the finite beam size, the reflected beam corresponds to a range of vicinal angles. To improve the angular resolution, a slit was placed in the path of the reflection beam to narrow the vicinal angle range down to 0.25°. To make systematic thickness dependent study, Fe wedged samples (10–61 ML) were grown with the slope along the step edges of the vicinal surface. In this way, the film thickness and the vicinal angle can be varied independently.

III. RESULTS

A. LEED results

The LEED patterns of the Ag substrate show very sharp diffraction spots from both the flat [Fig. 1(a)] and the curved [Fig. 1(b)] surfaces. In particular, the well-resolved double LEED spots can be clearly observed on curved substrate for vicinal angles greater than 4°, indicating the formation of periodic atomic steps on the vicinal surface. The separation between the double LEED spots from the vicinal surface increases with the vicinal angle. The ratio of the double LEED spot splitting to the primary LEED spot separation allows us to calculate the local vicinal angle on the curved substrate. As mentioned earlier, the vicinal angle can also be

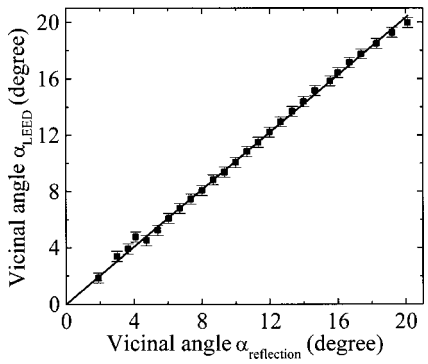

FIG. 2. Relationship between the vicinal angle obtained from Ag double LEED spots and the vicinal angle obtained from a laser beam reflection measurement. The solid line is a linear fit to the data with a slope of 1.02.

obtained by the reflection angle of the SMOKE laser beam. By linearly translating the curved substrate under the illuminating spot of the LEED and SMOKE laser beam, we determined the vicinal angle using both methods. Figure 2 shows the relationship between the vicinal angles determined by the double LEED spots and by laser beam reflection. A linear relation can be clearly seen. In fact, a linear fit to the data yields a slope of 1.02, proving quantitatively that the double LEED spots are from periodic atomic steps of the vicinal surface. Since only fixed angle vicinal surfaces were studied previously, Fig. 2 provides systematic experimental data proving that the double LEED spot splitting is proportional to the vicinal angle.

Figures 1(c) and 1(d) show the LEED patterns of 25 ML Fe grown on flat and vicinal surfaces. After the growth of the Fe film, the single-crystal structure remains, but the LEED spots are no longer as sharp as those from the Ag substrate. The double LEED spots from the vicinal surface evolve into elongated streaks and persist up to 61 ML of Fe (the thickest we studied). The elongated LEED streaks show that the atomic steps after the Fe film growth retain their direction, but the step density exhibits much greater fluctuations than does the step density of the Ag substrate. Nevertheless, the length of the LEED spot elongation increases with the vicinal surface in the same way as the separation between the double LEED spots from the Ag substrate, showing that the average step density of the Fe film follows that of the Ag substrate.

B. Thickness-dependent study

SMOKE measurements were taken on a curved substrate. The Fe magnetization is in the plane of the film in the thickness range studied (10–61 ML), so that only longitudinal SMOKE loops are shown in this paper. Figure 3(a) shows several hysteresis loops at different Fe film thicknesses at a vicinal angle of 10.5°. The magnetic field was applied perpendicularly to the step edges with the misalignment less than 3°. As in our previous results,[8] the hysteresis loops in Fig. 3(a) show that the atomic steps induce an in-plane uniaxial magnetic anisotropy with the easy magnetization axis parallel to the step edges.

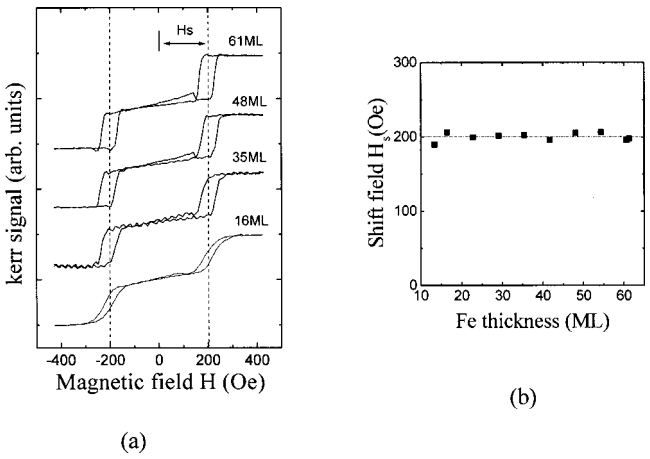

FIG. 3. (a) Hard-axis hysteresis loops of Fe films grown on a curved Ag(001) substrate at a vicinal angle of 10.5°. The shift field H_S (dashed line) measures the strength of the step-induced magnetic anisotropy. (b) The shift field H_S vs the Fe film thickness.

For a (001) stepped ferromagnetic film with in-plane magnetization, the magnetic energy for a magnetic field H applied along the hard axis is

$$E = K_u \cos^2\phi - MH\cos\phi - K\cos^2\phi\sin^2\phi. \quad (1)$$

Here K_u is the uniaxial magnetic anisotropy, M is the magnetization of the film, and ϕ is the angle between the $\vec{M}$ and $\vec{H}$.

For an antiferromagnetically coupled (001) sandwich with in-plane magnetization, the magnetic energy within a magnetic field H applied along the (100) direction is

$$E = JM_1M_2\cos(\phi_1-\phi_2) - M_1H\cos\phi_1 - M_2H\cos\phi_2 - K_1M_1^2\cos^2\phi_1\sin^2\phi_1 - K_2M_2^2\cos^2\phi_2\sin^2\phi_2, \quad (2)$$

where J is the antiferromagnetic coupling strength across the spacer layer, and K_i, M_i, and ϕ_i $(i=1,2)$ are the magnetic anisotropy, the magnetization, and the angle between the magnetization and the magnetic field of the ith magnetic layer. For two identical ferromagnetic layers $(M_1=M_2=M, K_1=K_2=K)$, the magnetization vectors of $\vec{M}_1$ and $\vec{M}_2$ should be symmetric relative to the direction of the magnetic field, i.e., $\phi_1=-\phi_2=\phi$. Under this condition, Eq. (2) becomes

$$E = 2JM^2\cos^2\phi - 2MH\cos\phi - 2K\cos^2\phi\sin^2\phi - 2JM^2. \quad (3)$$

Equations (1) and (3) are identical except for a factor of 2 and a constant. That is why the hard-axis hysteresis loop of a stepped magnetic thin film has the same shape as that of an antiferromagnetically coupled sandwich. It was shown that Eq. (3) has an analytical solution for the hysteresis loop[20] that produces most of the characteristic properties observed in experiment, especially the properties of zero remanence and two side loops under appropriate conditions. The saturation field calculated from Eq. (3), however, is not proportional to J. The reason is that Eq. (3) is based on single-domain magnetization, but in reality magnetic domains always appear in the region of the magnetization switching. Nevertheless, the middle point of the side loop, referred to as the shift field (H_S), is usually taken as a measure of the antiferromagnetic coupling strength. We then follow this convention in this paper to take the shift field H_S as a measure of the step-induced magnetic uniaxial anisotropy.

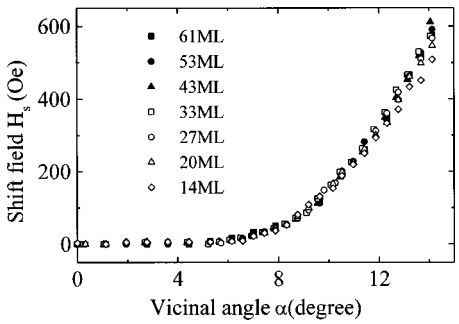

FIG. 4. Shift field H_S vs the vicinal angle α with several different Fe film thicknesses.

Despite some detailed differences in the shape of side loops which depend on the domain propagation during magnetization switching, Fig. 3(a) shows that the step-induced magnetic anisotropy depends very little on the Fe film thickness. Detailed measurements along the Fe wedge [Fig. 3(b)] indeed confirm that the step-induced magnetic anisotropy is independent of the Fe film thickness in the thickness range studied. To make sure that this statement is also true at other vicinal angles, we performed SMOKE measurements as a function of the vicinal angle (α) at different Fe thicknesses (Fig. 4). First, we observed that the H_S increases with α, consistent with our earlier results.[8] Second, all data collapse onto a single curve, showing that the step-induced magnetic anisotropy is independent of the Fe film thickness at all vicinal angles. We note that the shift field for 14 ML Fe is slightly smaller than the others for $\alpha > 13°$. This is probably due to nonuniformity of the substrate.

The above experiment suggests that the step-induced anisotropy is not localized at the step edges; otherwise, a $1/d_{\mathrm{Fe}}$ dependence would have been expected. To further clarify this issue, we performed step decoration experiments.

C. Step decoration with Pd and Ag

Step decoration is a powerful method to single out the effect of step edges. By growing a small fraction of foreign atoms on the ferromagnetic surface, it was shown that the step-induced magnetic anisotropy can be greatly influenced if the foreign atoms migrate to the step edges. This effect has been used as a proof of the local nature of the step-induced magnetic anisotropy in the Co/Cu(001) system.[7,9] We performed the same experiment in the Fe/Ag(001) system with Ag and Pd as the foreign atoms. To obtain an equal amount of atoms per atomic step at all vicinal angles and to promote the movement of deposited atoms to move to the step edges, we used the side growth method whereby the evaporator faced to the side of the curved substrate [Fig. 5(a)].

Two atomic rows of Ag were grown on a 15-ML Fe surface, and SMOKE measurements were made before and after

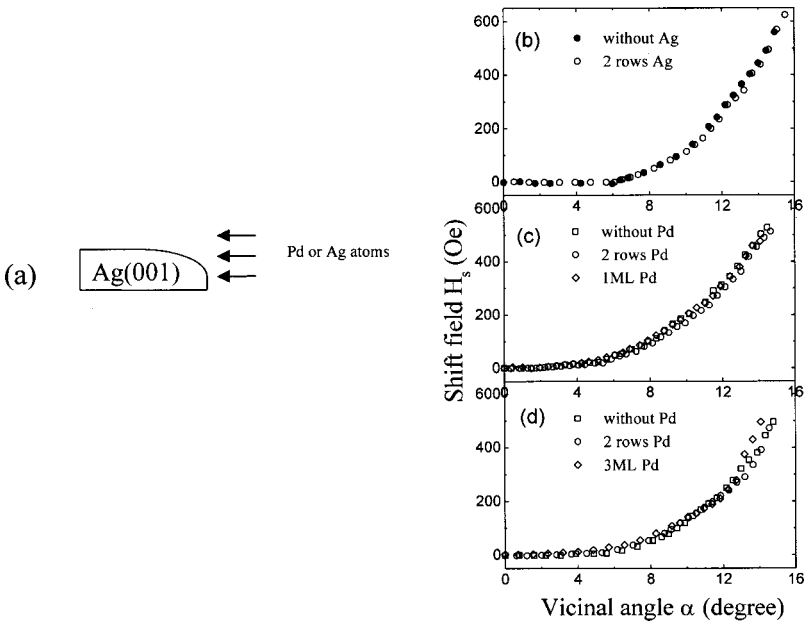

FIG. 5. (a) Schematic drawing of the side-growth geometry. The shift field H_S vs the vicinal angle α of (b) 15 ML Fe prior to and after Ag step decoration on the Fe film, (c) 30 ML Fe prior to and after Pd decoration on the Fe film, and (d) 27 ML Fe prior to and after Pd decoration on the Ag substrate.

the step decoration. As shown in Fig. 5(b), we observed no effect of the Ag decoration on the step-induced magnetic anisotropy over the entire vicinal angle range. Growing 10 ML Ag at normal incidence on top of the Fe film also had no effect on the step-induced magnetic anisotropy. This result again suggests that the step-induced magnetic anisotropy in the Fe/Ag(001) system is not localized at the step edges.

To make sure that the above results were not associated only with Ag, Pd step decoration was also performed on a 30-ML Fe film. It is well known that a ferromagnetic layer can induce a magnetic moment in Pd at the Pd/ferromagnetic interface. Because of the strong spin-orbit interaction in Pd, the induced moment also has effect to enhance the magnetic anisotropy. As for the stepped film, it was shown that Fe on stepped Pd(001) has a much stronger step-induced magnetic anisotropy than Fe on stepped Ag(001). Moreover, the Pd also has a strong effect on the α dependence of the step-induced magnetic anisotropy and the Curie temperature of the Fe film.[21] Thus a significant change of the stepped induced magnetic anisotropy would be expected for Pd step decoration on the Fe/Ag(001) system if the anisotropy were localized at the step edges. As shown in Fig. 5(c), the addition of two atomic rows of Pd on the 30 ML Fe/stepped Ag(001) has no effect on the step-induced magnetic anisotropy. To be sure that all the step edges were decorated with the Pd atoms, 1 ML Pd was grown on top of the 30 ML Fe at normal incidence, and we found that the step-induced magnetic anisotropy remains unchanged. To further distinguish the steps at the Fe/vacuum interface from the steps at the Fe/Ag interface, we performed the same Pd step decoration experiment on the Ag substrate prior to the growth of a 27-ML Fe film. Again, we observed no effect of the Pd on the step-induced magnetic anisotropy even with a seed layer of up to 3 ML Pd between the Fe and Ag [Fig. 5(d)]. The Ag and Pd step decoration experiments show that the step-induced magnetic anisotropy in the Fe/stepped Ag(001) system is not localized at the step edges.

IV. DISCUSSION

The origin of the step-induced magnetic anisotropy is not well understood at present. The popular explanation of the step-induced magnetic anisotropy is based on Néel's pair-bonding model[10] in which the magnetic anisotropy is generated by the spin-orbit interaction through the nearest-neighbor electronic hybridization. The absence or modification of the nearest neighbors for atoms at the step edges will generate magnetic anisotropy in thin films grown on vicinal surface. A phenomenological anisotropy function can be derived using Néel's pair-bonding model,[22] which can successfully explain the α dependence of the step-induced magnetic anisotropy. However, there are two simplifications in this model. First, $3d$ electrons in transition metals are not localized, so that it is questionable how good the approximation of using only the nearest neighbors is. Second, the strain in the interior of the film is neglected in the model, so that even if the nearest-neighbor approximation is valid, the localized nature of the model should result in a $1/d$ dependence of the step-induced magnetic anisotropy, contradicting experimental observations.

The present experiment shows that the step-induced magnetic anisotropy in the Fe/stepped Ag(001) system is almost independent of the Fe film thickness. This is a strong indication that the step-induced magnetic anisotropy is non local, i.e., the symmetry breaking in this system should occur not only at the step edges, but also in the interior of the Fe film. In fact, such an effect has been observed in magnetic surface anisotropy, where lattice distortion breaks the cubic symmetry throughout the ferromagnetic film.[23] For stepped thin films, the deviation from the $1/d$ dependence in the step-induced magnetic anisotropy suggests that lattice distortion due to the atomic steps also occurs throughout the ferromagnetic film. Lattice distortion is usually caused by lattice mismatch between the substrate and overlayer film and will persist until dislocations relax the strain. The thickness

independence of the step-induced magnetic anisotropy in the Fe/stepped Ag(001) system indicates that the step-induced magnetic anisotropy in the system is dominated by the lattice distortions of the Fe film. The lattice constants of fcc Ag, and bcc Fe are 4.08 and 2.87 Å, respectively. Thus bcc Fe(001) fits very well to fcc Ag(001) (0.8% misfit) by aligning the Fe [100] to Ag [110]. That is why bcc Fe can grow epitaxially on Ag(001). However, there is a $\sqrt{2}$ difference in the lattice constant in the film normal direction between bcc Fe(001) and fcc Ag(001). Such a large lattice mismatch is usually ignored in considering Fe film growth on flat Ag(001), but obviously cannot be ignored for Fe films grown on stepped Ag(001). The lattice distortion at the step edges should induce a significant strain in the ferromagnetic Fe film inducing magnetic anisotropy. If the strain extends into the interior of the ferromagnetic film, a deviation from the $1/d$ dependence in the step-induced magnetic anisotropy predicted by Neel's pair-banding model will then be expected. Our results from the Fe stepped Ag(001) system suggest that the lattice distortion is probably the dominant contribution to the step-induced magnetic anisotropy in this system.

In addition to the spin-orbit interaction, dipolar interaction is an another source of the magnetic anisotropy. For a flat film, dipolar interaction generates $2\pi M^2$ shape anisotropy which favors the magnetization in the plane of the film. For a stepped film, Bruno pointed out[24] that magnetic poles distributed along the step edges could generate a magnetic shape anisotropy that favors an easy axis parallel to the step edges. More recently, Arias and Mills obtained a parameter-free analytical form of roughness-induced magnetic anisotropy.[25] Because of the dipolar origin, the anisotropy has a rather weak thickness dependence. Wolfe *et al.* performed an experiment on Fe/MgO(001) with off-axis growth and found that roughness with a preferred direction indeed induces uniaxial magnetic anisotropy with the magnitude in agreement with the theory. However, in order to generate a comparable magnetic anisotropy the amount of film roughness is much greater than that of the epitaxially grown stepped thin films.[26] In addition, some stepped films show the easy magnetization axis perpendicular to the step edges,[6,18] in contrast to the roughness-induced magnetic anisotropy. Therefore it is unlikely that the film roughness is the dominant contribution to the step-induced magnetic anisotropy.

It should be mentioned that our result is different from the result obtained from a similar system of Fe/Ag/GaAs(001) in which a Ag seed layer was grown on stepped GaAs(001) with a vicinal angle of 2°. In this system, Leeb *et al.*[27] found that the easy magnetization axis is perpendicular to the step edges and that the step-induced magnetic anisotropy is inversely proportional to the Fe film thickness. This different result is probably due to the use of a different substrate or the effect of segregation of Ga or As. Nevertheless, the observed $1/d$ dependence in their system may suggest that the strain in their system is well relaxed by dislocations. Another interesting comparison is Fe/stepped Cr(001) in which the strain is presumably much smaller than in Fe/stepped Ag(001) because of the ideal lattice match between bcc Fe and bcc Cr. Escorcia-Aparicio *et al.* performed a step decoration experiment by growing a row of Au atoms on the stepped Fe surface and observed an obvious change of the step-induced magnetic anisotropy.[28] This result suggests that once the strain is absent or very small, the step-induced magnetic anisotropy is localized to the step edges.

The last issue in the Fe/stepped Ag(001) system concerns the nature of the strain inside the Fe film. If the strain is dominated by the lattice mismatch in the direction normal to the film, the strain inside the Fe film must increase with the step density. Then if the strain is the dominant contribution to the step-induced magnetic anisotropy, the step-induced magnetic anisotropy should increase with the step density in the Fe/stepped Ag(001) system, consistent with the α-dependence experimental result. On the other hand, the thickness independence of the step-induced magnetic anisotropy suggests that the strain per unit volume at a given vicinal angle should be a constant for Fe film in the 10–61 ML range. At present, we do not have a microscopic theory of the step-induced magnetic anisotropy (under this picture) because such a theory would require the knowledge of the step-induced strains. It will be a very interesting future experiment to investigate the growth of Fe on stepped Ag(001) to understand the strain inside the Fe film.

V. SUMMARY

In summary, we have shown that the step-induced magnetic anisotropy in the Fe/stepped Ag(001) system does not depend on the Fe film thickness in the 10–61 ML thickness range. Step decorations with Ag and Pd atoms also have no effect on the step-induced magnetic anisotropy. All these results show that the step-induced magnetic anisotropy in the Fe/stepped Ag(001) system is not localized at the step edges. We propose that the strain caused by the large lattice mismatch in the direction normal to the film is the dominant contribution to the step-induced magnetic anisotropy in this system.

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