Isolating the Step Contribution to the Uniaxial Magnetic Anisotropy in Nanostructured Fe/Ag(001) Films

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(Received 18 October 2005; published 6 February 2006)

We have investigated the possibility of isolating the step-induced in-plane uniaxial magnetic anisotropy in Fe/Ag(001) films on which nanoscale surface ripples were fabricated by the ion sculpting technique. For rippled Fe films deposited on flat Ag(001), the steps created along the ripple sidewalls are shown to be the only source of uniaxial anisotropy. Ion sculpting of ultrathin magnetic films allows one to selectively study the step-induced anisotropy and to investigate the correlation between local atomic environment and magnetic properties.

DOI: 10.1103/PhysRevLett.96.057204 PACS numbers: 75.30.Gw, 75.75.+a, 81.16.Rf

Magnetocrystalline anisotropy is one of the most important properties of magnetic materials and one of the most deeply investigated [1–13]. Because of its local nature, magnetocrystalline anisotropy sensitively depends upon the local atomic environment [14]. Thus, atoms in sites with different coordination (e.g., step, surface, or bulk sites) are each characterized by very different magnetic anisotropies. In low-dimensional magnetic systems such as ultrathin films [1–9], nanowires [10–12], and nanodots [13], atoms at low-coordinated sites are of extraordinary importance as they often give the dominant contribution to the overall anisotropy of the system.

In this respect, a monoatomic step on the surface of a magnetic film is a particularly appealing system, due to both its relevance and simplicity, and much effort has been made to investigate its magnetic anisotropy [2–8]. As a general rule, the selective study of the step-induced anisotropy requires that no other source of magnetic anisotropy having the same symmetry as the atoms at the steps is present. However, the prototype systems for such studies, ultrathin magnetic films grown on vicinal surfaces, suffer from the inevitable presence of sources of uniaxial anisotropy other than surface steps. There, anisotropic strains originated by the step-height difference between substrate and film are present that induce a uniaxial contribution to magnetic anisotropy via bulklike magnetoelastic effects. Such anisotropy unavoidably adds up to any surface contribution, thus preventing its correct evaluation. The understanding of the magnetic properties of atoms at low-coordinated sites would greatly benefit from the availability of a general method that allows one to isolate their contribution.

In this Letter, we report on the possibility of isolating the step-induced uniaxial anisotropy in ultrathin magnetic films, taking advantage of the nanoscale self-assembly processes induced by ion beam irradiation [15–17]. We apply ion sputtering at grazing incidence on ultrathin Fe/Ag(001) films in order to induce the formation of nanometer-sized surface ripples oriented along the projection of the ion beam direction on the surface [11,15,16]. The preferential formation of Fe atomic steps along the ripple sidewalls gives rise to a clearly observable uniaxial anisotropy [11]. For films of various thickness sputtered under identical conditions, the uniaxial anisotropy scales as the inverse film thickness, thus demonstrating that the steps at the surface represent the *only* source of uniaxial anisotropy. By contrast, the uniaxial anisotropy measured for Fe films deposited onto a vicinal-like substrate, fabricated by inducing ripples on a clean Ag(001) surface [16], reveals the presence of bulklike magnetoelastic contributions that prevent the correct evaluation of the step-related properties [5]. The validity of the ion sculpting approach is, therefore, demonstrated for the Fe/Ag(001) system, for which the vicinal approach proves unsuccessful. These findings extend the results already available for the model system Co/Cu(001) [11] and show the general applicability of the method to systems characterized by different crystallographic structure and magnetic properties.

The experiments have been performed in an ultrahigh vacuum system with a base pressure $<1 \times 10^{-10}$ mbars, equipped with facilities for the growth and characterization of ultrathin magnetic films [11,18]. The chamber features the magneto-optical Kerr effect (MOKE), low energy electron diffraction (LEED) with spot profile analysis, and Auger electron spectroscopy (AES). The arrangement of the components within the chamber allows MOKE in either polar or longitudinal configuration to be monitored during film growth. The Ag(001) substrate was prepared by cycles of 1 keV Ar^+ sputtering and annealing at 800 K until a sharp LEED pattern was observed and all contaminants were found below AES detection sensitivity. Iron films have been deposited at normal incidence, in order to prevent any growth-induced magnetic anisotropy [19]. Fe grows on Ag with epitaxial relations $[100]_{Fe} \parallel [110]_{Ag}$ and $[001]_{Fe} \parallel [001]_{Ag}$. The film thickness has been calibrated by the observation in the curves of longitudinal MOKE vs deposition time of the spin reorientation transition (SRT) [20]. The determination of thickness is affected by an uncertainty of the order of $\pm 5\%$.

For growth on the flat Ag(001), Fe films have been deposited with the substrate held at 300 K. Under these conditions, Fe growth proceeds in 3D mode with the formation of square hillocks [21] and a fraction of an Ag monolayer floating on the surface [21,22]. With regard to magnetic properties, the as-deposited films do not exhibit any in-plane uniaxial anisotropy. The Fe films have been nanostructured by irradiation with 1 keV Ar^+ ions at an incidence angle of 70° from the surface normal and along the Fe [100] direction. The ion sputtering has been performed with the sample held at 350 K. The sputtering temperature has been chosen such that well defined ripples, free of segregated Ag, are formed at high ion fluence, as checked by analysis of LEED spot profiles and AES.

The experiments aimed at the identification of the step contribution to the uniaxial magnetic anisotropy were carried out at low ion fluence [0.2 and 0.4 MLE, 1 monolayer equivalent (MLE) is 1.2×10^{15} ions/cm^2], at which the single-ion impact sites start organizing to build up a ripple structure [16]. At such ion fluences, the amount of material removed due to sputtering is very low. Under these conditions, the thickness of the film after sputtering is well approximated by its initial value, and magnetostatic contributions to the uniaxial anisotropy are negligible [11]. The initial morphology of the films is fairly similar in the thickness range of interest [10–42 monolayers (ML)] [23]: Thus, we assume that, in the low-fluence limit, identical ion doses induce the formation of the same number of steps in Fe films of different initial thickness.

In Fig. 1, we display longitudinal MOKE hysteresis loops measured at 350 K for films of various initial thickness irradiated with a fluence of 0.4 MLE. On the left side, we show loops measured with external field **H** parallel to the bcc [100] direction (parallel to the irradiation direction); on the right side, we show loops measured with **H** parallel to the [010] direction (perpendicular to the irradiation direction). The loops measured with $\mathbf{H} \parallel [100]$ have a square shape with sharp transitions, while those measured with $\mathbf{H} \parallel [010]$ exhibit a characteristic shape composed by two split semiloops with sharp transition fields H_{s1} and H_{s2}. This shape is indicative of the presence of a uniaxial anisotropy with its easy axis aligned parallel to the [100] direction (step edge direction), superimposed to the fourfold anisotropy typical of the Fe/Ag(001) films.

The magnetic anisotropy of this system is described by the cubic (K_c) and uniaxial (K_u) anisotropy constants which enter the expression for the film free-energy density for in-plane magnetization **M**

$$E = \frac{K_c}{4} \sin^2(2\phi) + K_u \sin^2(\phi) - \mathbf{H} \cdot \mathbf{M}, \qquad (1)$$

where ϕ is the angle between **M** and the uniaxial easy axis. Minimization of the free-energy density of Eq. (1) is used to extract K_u and K_c from the hysteresis loops. Since the values of K_c are of marginal interest for our discussion, we will focus on the behavior of K_u. The values of K_u extracted from the experimental data are displayed as a function of the film thickness in Fig. 2 for two different ion fluences. The value of K_u markedly decreases for increasing Fe thickness. Fitting the thickness dependence of K_u according to the simple relation $K_u \propto 1/d$ (inverse-thickness scaling) yields good agreement with the experi-

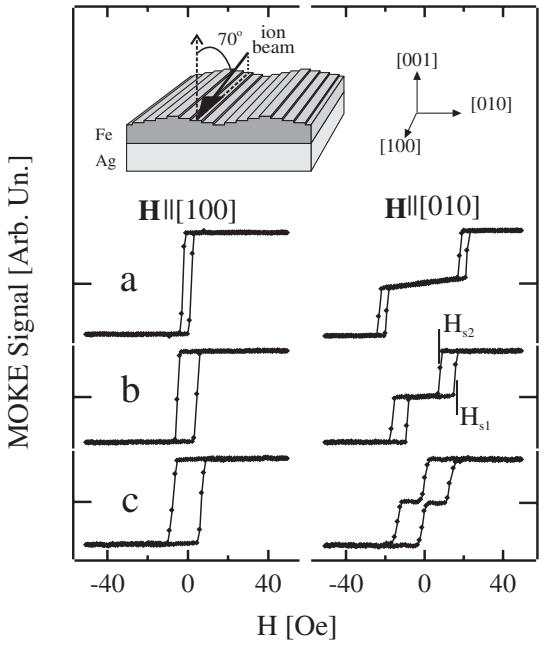

FIG. 1. Longitudinal MOKE hysteresis loops as a function of Fe thickness for films irradiated with 0.4 MLE ion fluence. The external field is parallel to the $[100]_{Fe}$ (left column) and $[010]_{Fe}$ (right column) directions. Initial Fe film thickness was (a) 10, (b) 18, and (c) 32 ML. The loops have been normalized at saturation. The inset depicts the experimental geometry. The sketch of the system is reported for the sake of clarity only.

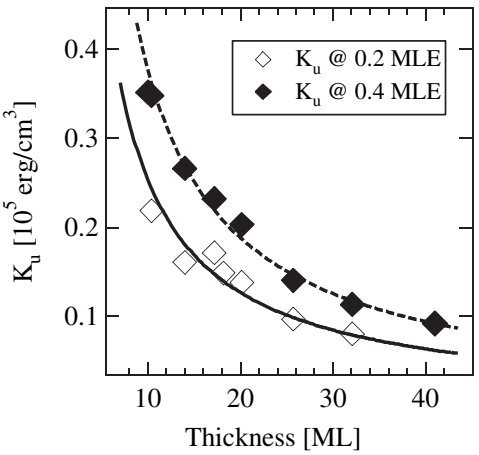

FIG. 2. Uniaxial magnetic anisotropy constant K_u as a function of initial film thickness for two different ion fluences. The lines are fits to the experimental data according to the equation $K_u \propto 1/d$, where d is the film thickness. The experimental uncertainty is comparable to the marker size.

mental data. The observation of the inverse-thickness scaling of K_u directly proves that, under these experimental conditions, the uniaxial anisotropy originates *only* at the film surface. Therefore, we *selectively* measure the step-induced uniaxial anisotropy.

To the best of our knowledge, experiments performed on vicinal substrates always failed to identify the step-induced contribution to uniaxial anisotropy, due to the ubiquitous presence of strain-induced uniaxial anisotropy, and different behaviors were observed whenever different film or vicinal-substrate combinations were employed [3,5,7]. In the case of Fe films grown on vicinal Ag(11n), the strain originates from the large step-height difference between film and substrate due to the bcc/fcc character of the system. These films do exhibit a uniaxial magnetic anisotropy [4], which was found to be constant in the 14–61 ML thickness range and, therefore, ascribed to bulklike contributions [5].

In order to show that the possibility of isolating the step-induced anisotropy is affected by the presence of sources of uniaxial anisotropy other than the steps, we have studied the thickness and the adsorbate dependence of the uniaxial anisotropy for Fe films deposited on a Ag(001) surface previously rippled by ion sculpting [16].

In this experiment, the clean Ag(001) substrate was first irradiated with 30 MLE of 1 keV Ar^+ ions incident at an angle of 70° from the surface normal and in the plane defined by the fcc [001] and [110] directions, with the sample held at 180 K. After this treatment, LEED spot profile analysis showed that ripples oriented along the [110] fcc direction had formed. The sidewalls of the ripples consisted of facets at an angle of 11° with respect to the surface plane. Under these conditions, a large unbalance in the number of steps oriented along the ripple direction and normal to it had developed. Fe films of various thickness up to 20 ML were then deposited at 170 K, and their anisotropy measured at the same temperature. The low temperature growth inhibits Ag segregation [22] and avoids thermal smoothening of both the substrate and the film, preserving the uniaxial symmetry of the system.

The hysteresis loops measured with **H** perpendicular to the ripples (**H** ∥ bcc[010]), reported in Fig. 3 as a function of thickness, are split in two semiloops. The hysteresis loops measured with **H** parallel to the ripples (not reported) are square. The loop shape, therefore, suggests that also in this case a uniaxial anisotropy superimposed to a cubic anisotropy term is present, with the uniaxial easy axis lying along the ripple direction [4,5].

Figure 4 shows the thickness dependence of the anisotropy constant K_u extracted from the loops of Fig. 3. Clearly, the thickness dependence of K_u bears no resemblance of the $1/d$ behavior, as it increases from a value below our detection sensitivity at 7 ML (immediately following the SRT), up to $K_u = (0.8 \pm 0.1) \times 10^5$ erg/cm^3 at 13 ML, and thereafter saturates. The saturation of

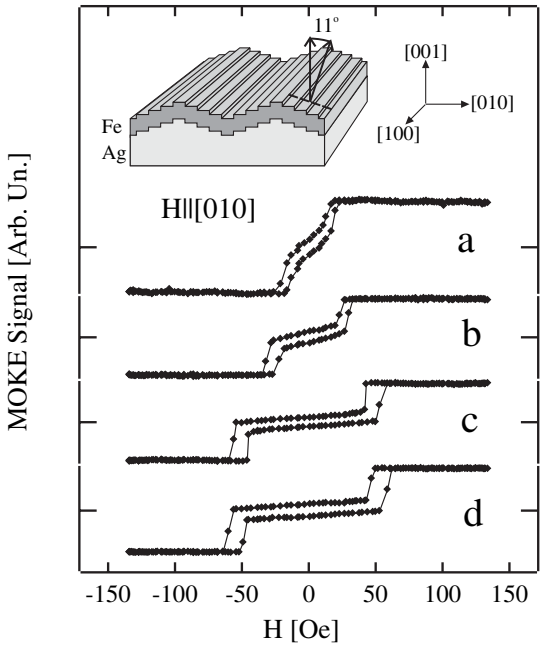

FIG. 3. Longitudinal MOKE hysteresis loops for Fe deposited on nanostructured Ag(001) as a function of thickness. The external field is parallel to the bcc [010] direction. The Fe thickness was (a) 7.5, (b) 8.5, (c) 15.5, and (d) 20 ML. The loops have been normalized at saturation. The inset depicts the experimental geometry. The sketch of the system is reported for the sake of clarity only.

K_u above 13 ML agrees with previous studies of Fe grown on vicinal Ag [5] and is a clear indication that a bulklike uniaxial anisotropy is present. We attempt to determine the presence of surface contributions to the measured uniaxial magnetic anisotropy by exposing the surface to adsorbates [5,24] that are known to affect the anisotropy by modifying the step electronic structure. In our case, oxygen was dosed on a 20 ML thick Fe film grown on a rippled Ag(001) surface. The exposure was performed with the sample held

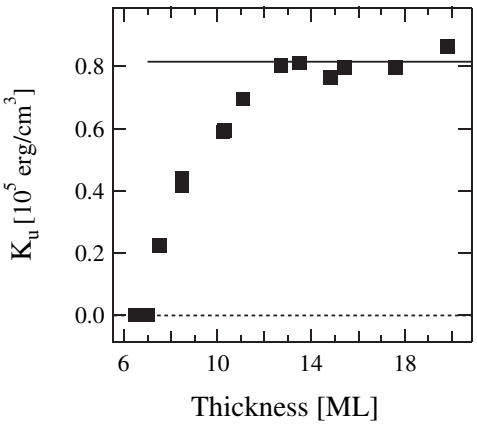

FIG. 4. In-plane uniaxial magnetic anisotropy constant as a function of film thickness for Fe films grown on a nanostructured Ag(001) surface. The continuous line represents the saturation value for K_u.

at 170 K in order to effectively avoid any change of morphology, due to interaction with oxygen [25,26], and to inhibit Ag segregation. We observed that the uniaxial anisotropy had increased by a factor of 2 after exposure to 10 L of O_2 (1 L = 10^{-6} Torr s). A similar effect has been observed for exposure to CO. From the large variation of K_u, taking place at exposures for which we expect that bulk sites are very weakly affected, we conclude that a surface-type uniaxial anisotropy is clearly present, too.

At variance with the previous case of ion-sculpted Fe films on flat Ag(001), we observe that, for Fe films deposited on an ion-sculpted Ag(001) substrate, there is simultaneous presence of bulklike and step-related contributions to the magnetic anisotropy with the same uniaxial symmetry. Under these conditions, there is not, to the best of our knowledge, a method that allows the isolation of the surface step properties.

In summary, we have shown that the ion sculpting technique permits one to selectively address the step-induced magnetic anisotropy in ultrathin magnetic films. In order to show the effectiveness of this method, we applied it to the Fe/Ag(001) system, for which other approaches have not succeeded to measure the step contributions alone. The present results extend and generalize the findings for the model system Co/Cu(001) [11], thus showing the wide applicability of this approach for system endowed with different anisotropy and crystal structure. The availability of such a general and effective method will allow one to experimentally test theoretical predictions as well as to investigate the influence of other variables, such as surface orientation, film, or substrate combination and the role of adsorbates on the magnetic anisotropy of atoms at low-coordinated sites. The information thus obtained can extend the knowledge of the correlation between the symmetry of the atomic environment and the magnetic anisotropy, allowing the fine tailoring of the anisotropy of nanosized aggregates in the quest for novel magnetic properties.

Financial support from the MIUR programs FIRB No. RBNE017XSW, FIRB No. RBNE01YLKN, and PRIN No. 2004029329 is acknowledged.

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