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Uniaxial magnetic anisotropy tuned by nanoscale ripple formation: Ion-sculpting of Co/Cu(001) thin films

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We have investigated the growth of surface nanostructures on a Co/Cu(001) film and the growth of Co films on a nanostructured Cu(001) substrate as well as the effect of nanoscale pattern formation on the film magnetic properties. Here we demonstrate by scanning tunneling microscopy measurements and magneto-optic Kerr effect hysteresis curves that low-temperature grazing-incidence ion sputtering can be used to induce the formation of nanoscale ripples which reduce the four-fold symmetry of the Co film to two-fold, thus generating a strong in-plane uniaxial magnetic anisotropy. The nanostructures and the associated uniaxial magnetic anisotropy were found to be stable up to room temperature. © *2004 American Institute of Physics.*
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In the last decade, the influence of nanoscale interface morphology on the magnetic properties of thin films and multilayers has been widely investigated. Apart from fundamental reasons, the interest in this topic was mainly motivated by the fact that many magnetic devices are based on thin film technologies. Coercive field, magnetization reversal processes, as well as giant magneto resistance effect[1,2] are affected by surface roughness which develops during thin film deposition. For the above-mentioned reasons, the possibility of tailoring the surface morphology of thin magnetic films at the nanometer scale has recently attracted a great deal of interest.[3] A particularly attractive topic is the strong in-plane uniaxial magnetic anisotropy induced by the formation of regular nanosized patterns on the film surface. So far, two methods have been proposed to artificially induce uniaxial magnetic anisotropy in thin films by means of nanostructuration: film deposition on vicinal surfaces, where a regular arrangement of parallel steps is induced by the crystal miscut,[4] and grazing-incidence deposition to induce the formation of nano-scale anisotropic mounds.[5–7]

Here we demonstrate that low-temperature grazing-incidence ion sputtering[8–11] can be used to induce the formation of nanostructures which reduce the four-fold symmetry of the Co/Cu(001) film to two-fold, thus provoking a strong in-plane uniaxial magnetic anisotropy. Two routes have been followed: sputtering of a flat Co film grown on Cu(001), and Co deposition on nanostructured Cu substrate. In these two cases, we report on the relationship between the surface morphology and magnetic properties observed by scanning tunneling microscopy (STM) and magneto-optic kerr effect (MOKE) measurements. In addition, we briefly report on the thermal stability of these systems, an important issue from the viewpoint of applications.

The clean Cu(001) substrate was prepared by cycles of ion sputtering with Ar^+ ions at 1 keV and annealing to 800 K. The cleanliness was checked by observation of well-ordered terraces larger than 100 nm by STM and of sharp 1×1 low energy electron diffraction pattern. No contamination was detected by Auger electron spectroscopy. In the first experiment, 12 ML of Co were deposited on the clean Cu(001) surface at normal incidence and at a deposition rate of about 0.25 ML/min. During Co deposition, the substrate temperature was held at 300 K. After the deposition, the substrate was cooled down to 180 K and sputtered with Ar^+ ions at 1 keV. Sputtering was performed along the azimuthal [110] direction at an incidence angle of 70° from the surface normal. During sputtering, the ion flux was about 4 μA/cm^2. The sputtering time was used as a variable parameter. Subsequent STM and MOKE measurements were performed at 140 K in order to inhibit the relaxation of the nanostructures.

Figure 1(a) shows the hysteresis loops of the Co films taken with the magnetic field along the [1-10] direction (per-

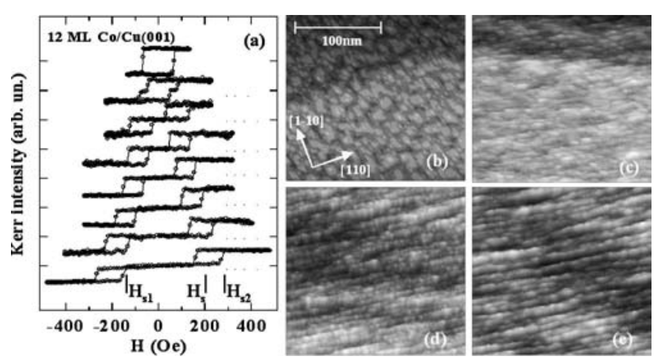

FIG. 1. (a) Hysteresis curves of the flat 12 ML thick Co/Cu(001) film and after ion nanostructuring as a function of the sputtering time. The shift field is defined as $H_s=(H_{s2}-H_{s1})/2$. STM images (200 nm×200 nm) of the flat Co film (b), and after 60 s (c), 660 s (d), and 960 s (e).

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pendicular to the ripple axis) after ion sputtering with different ion doses. For sputtering time as low as 60 s, the hysteresis loops along the [1-10] splits into two semiloops while the one measured along the [110] direction stays unvaried. The origin of the modification of the hysteresis loops is the change in the magnetization reversal process induced by uniaxial magnetic anisotropy along the [1-10] direction.[12] In particular, the intensity of uniaxial magnetic anisotropy is proportional to the splitting H_s of the two semiloops. After 60 s, the observed anisotropy is still weak and the magnetic remanence is not zero. After 120 s of sputtering, the hysteresis loop is completely split and the magnetic remanence becomes now zero. Subsequent ion dosing further increases the magnetic anisotropy.

The behavior of the magnetic anisotropy can be qualitatively correlated to the evolution of the surface morphology. Figures 1(b)–1(e) show the STM images of the Co films after sputtering for 0, 60, 660, and 960 s, respectively. On the initial 12 ML thick Co film [Fig. 1(b)], we can see square islands on the terraces with the step edges aligned along the [110] equivalent directions.[13,14] After 60 s of sputtering [Fig. 1(c)], we can already find an incipient ripple-like periodic modulation. After sputtering for 660 and 960 s [Figs. 1(d) and 1(e)], we can clearly observe a well developed ripple structure along the ion incidence direction. As in the case of single-crystal metal surfaces,[9] the development of the ripple amplitude observed by STM well obeys a power law trend, while the wavelength is almost constant. This trend favorably increases the density of [110] steps and such unbalancing is known to induce uniaxial anisotropy.[4] Moreover, the ripple corrugation is expected to induce magnetic shape anisotropy. Both contributions of magnetic anisotropy are expected to increase in agreement with the MOKE results [Fig. 1(a)]. We note here that the observed changes in the magnetic anisotropy cannot be attributed to the role of gas adsorption: test experiments exposing the flat Co film to comparable Ar pressures in absence of the ion beam show no detectable change in the magnetic properties.

An important issue from the viewpoint of possible applications, is the thermal stability of the artificial nanostructures and of the related uniaxial magnetic anisotropy. Figure 2(a) shows the temperature dependence of the shift field H_s of a Co film sputtered for 480 s in the above-mentioned conditions. The sample was annealed at each temperature for 600 s, while the MOKE measurements were performed at low temperature. Starting from a shift field of about 175 Oe, as the substrate temperature is increased up to 250 K, H_s stays unchanged. At temperatures around 300 K, H_s starts slightly diminishing, and at 350 K definitively disappears. In independent experimental runs, the stability of the magnetic anisotropy was further observed after 12 h at room temperature. The observed behavior of H_s is caused by the relaxation of the ripple structures due to the Co atom migration on the Co/Cu(001) surface. Figures 2(b) and 2(c), respectively, show the asymmetry of the nanostructure shape[15] and the surface roughness (standard deviation of surface height) extracted from the STM images. From these results, we conclude that the smoothening of Co ripples begins near 300 K while, at higher temperature, both their shape asymmetry and the unbalance in step density rapidly decrease, in agreement with the results of the MOKE measurements and with the one found for Co films deposited at grazing-incidence on Cu(001).[16]

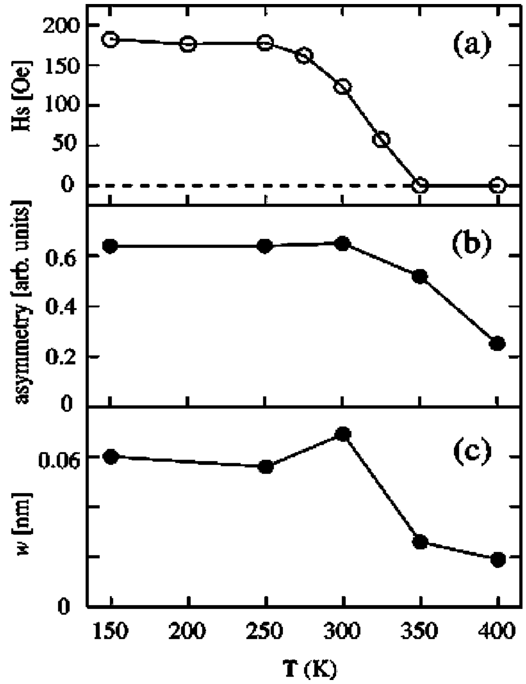

FIG. 2. Shift field (a), nanostructure shape asymmetry (b), and roughness (c) of a 12 ML thick Co/Cu(001) film sputtered at 180 K for 480 s, as a function of the annealing temperature.

Having proved that grazing-incidence ion sculpting can promote the formation of nanoscale ripples on Co films and to modify the magnetic anisotropy, one is led to the question whether the deposition of Co films on a prepatterned Cu(001) substrate allows the control of the magnetic properties in a similar way. Figure 3(a) shows the four hysteresis loops taken along the [1-10] direction (perpendicular to the ripple axis) after the deposition of 1, 2, 4, and 6 ML of Co on the presputtered Cu(001) substrate with the following conditions: 1 keV Ar^+ ion, $\theta_1 = 70°$, 4 $\mu A/cm^2$ for 660 s along [110]. For the 1 ML thick Co film, the magneto-optic signal as a function of the applied magnetic field is characterized by

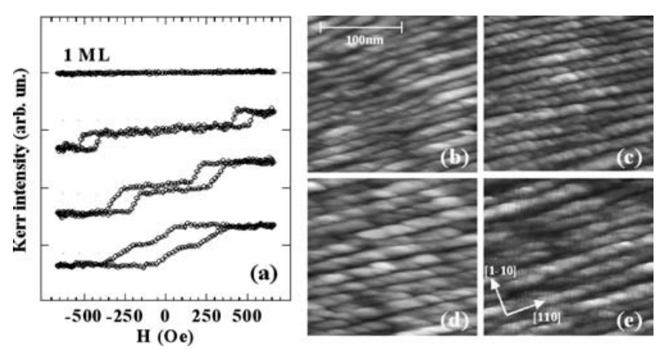

FIG. 3. (a) Hysteresis curves for Co films deposited on the nanostructured Cu(001) surface as a function of the Co coverage. STM image (200 nm ×200 nm) of the clean ripple structure made on Cu(001) surface at 180 K (b), after the deposition of 1 ML of Co on it and annealing at 300 K (c), after sample annealing up to 350 K (d), and after the deposition of further 2 ML of Co at 300 K.

a large even component which is probably induced by second-order effects related to the strong in-plane anisotropy.[17] Such an even component strongly reduces for higher Co coverages and definitively disappears for the 6 ML thick Co film. In Fig. 3(a), the odd part of the Kerr signal is shown. The flat Kerr signal for the 1 ML thick Co film indicates that the maximum field generated in our experimental apparatus (650 Oe) is not able to induce the switching of the magnetization along the [1-10] direction. After 2 ML Co deposition, we can observe a very large separation of two semiloops corresponding to a shift field of about 500 Oe. Increasing Co coverage, the two semiloops get closer, and finally they merge after the deposition of 6 ML of Co. On the other hand, the hysteresis loop taken along [110] (parallel to the ripple axis) on the 4 ML film had a typical easy-axis square shape.

We may simply interpret this behavior assuming that the growth of Co on the Cu ripple structure is conformal thus leaving the step density on the Co film unchanged during the growth. Since a surface-type behavior is expected for the magnetic anisotropy originating by atoms at step site, the anisotropy intensity is reduced by the increase of the total volume of Co film. In addition, the hysteresis loop for the 6 ML thick Co film appears to be considerably tilted with respect to the ones shown in Fig. 1(a). The different shape of the hysteresis loops may be caused by the existence of the dual interface and by a larger magnetostatic contribution due to this film geometry. However, this point needs further investigations.

We made related STM measurements to investigate the morphology and thermal stability of the Co film grown on the Cu ripple structure. Figure 3(b) shows the STM image of a well-ordered ripple structure obtained on the Cu(001) surface by Ne-ion sputtering: 1 keV, 3 μA, 900 s, $\theta_1 = 70°$ along [110] direction at 180 K. We deposited 1 ML of Co on this ripple structure at 110 K and observed this Co film by STM at room temperature [see Fig. 3(c)]. Contrary to our expectation, the ripple structure covered by 1 ML of Co was stable also at room temperature whereas the clean Cu ripple structure is completely smeared out at room temperature because of the strong atom migration.[11] We can infer that the extra interface bonding at the Cu–Co interface well inhibits atom migration and the relaxation of the ripple structures. The morphological stability of this film was confirmed also after annealing at 350 K for 600 s [Fig. 3(d)]. We then added 2 ML of Co to the 1 ML thick Co film at room temperature [Fig. 3(e)] and did not find any distinct change of the surface morphology, thus concluding that Co film growth on the ripple structures can be considered conformal to a good approximation.

In conclusion, we investigated the formation of nanoscale periodic patterns on Co films grown on flat and prepatterned Cu(001) substrates. In both cases, we found that ripple structures, produced by means of grazing-incidence ion sculpting, can deeply modify the magnetic properties of the Co film and in particular induce a strong in-plane uniaxial anisotropy which persists at room temperature. The ion sculpting technique, thus allows us to tailor *in situ* in a fast and effective way fine morphological details of the nanostructures as well as the magnetic properties of the film over extended macroscopic areas of the substrate. Its simplicity and the generality of the results are expected to be easily extended to other systems of technological interest.

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