Influence of substrate roughness on spin reorientation transition of ultrathin Co films on Pd(111)

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We report a drastically different behavior in spin reorientation transition of ultrathin Co films grown on smooth versus rough Pd(111) single crystal substrates. The morphology and magnetic anisotropy of epitaxially grown Co films have been studied with in situ scanning tunneling microscopy (STM) and surface magneto-optical Kerr effects. On an atomically flat substrate, a smooth transition from perpendicular to in-plane magnetization occurs in a thickness interval of over 1.5 ML, beginning at ~4.5 ML. In contrast, rough substrate causes the transition to begin quite earlier at ~3 ML and complete abruptly in less than 1 ML range. Morphology difference of the Co films obtained with STM suggests that on rough substrate, nonuniform coverage of Co leads to locally thicker regions triggering earlier spin reorientation transition. © 2001 American Institute of Physics.

While earliest studies regarding thin ferromagnetic films were based on the assumption of morphologically smooth surface and interface, a number of recent experimental results show that surface and interface roughness in real films greatly affect the magnetic properties such as coercivity, magnetic domain structure, magnetization reversal, magneto-resistance, and spin reorientation transition. For example, in a study of ultrathin Fe films on Ag(001), Schaller et al. observed spin reorientations induced by morphology changes of the top Fe surfaces and explained the results by a change of both the magnetic dipolar and the magnetocrystalline anisotropy due to the roughness. In another observation, Poulopoulos et al. performed a scanning tunneling microscopy (STM) roughness study of Ni/Cu(001) and concluded that while the effect is small, roughness favors the easy magnetization to be out-of-the-film plane. Recent theoretical study by Zhao et al. confirmed a strong dependence of surface magnetization on surface roughness.

So far, most studies on roughness effect have been concerned with a single rough boundary, i.e., the surface, assuming a smooth interface between the substrate and magnetic film. A few studies also have been reported on magnetic films grown on artificially roughened substrates. These results show that substrate roughness can affect the resulting magnetic properties significantly, such as decreased uniaxial magnetic anisotropy and increased coercivity with increasing roughness. Magnetic measurements on these studies were either performed ex situ or only at a number of fixed film thickness. But there are lack of clear observations of the evolution of the magnetic properties on roughened interface with increasing film thickness in the literature. Moreover, there are few direct studies on spin reorientation of ultrathin magnetic films showing the relation of substrate roughness and the spin reorientation even though it is not only a fundamentally interesting issue but also a technologically important issue in developing perpendicular and magneto-optical recording media. A combined in situ study of both structural and magnetic measurement as a function of the film thickness is required to tackle the problem. In this study, we use combined scanning tunneling microscopy (STM) and surface magneto-optical Kerr effects (SMOKE) to directly measure the influence of substrate roughness on the spin reorientation. Our results directly show that the onset of spin reorientation transition is affected by the initial substrate roughness, which subsequently modifies the growth morphology.

The experiment was performed in a custom-designed UHV chamber equipped with an in situ SMOKE measurement as well as a scanning tunneling microscope (STM). The system is best suited for following up the evolution of the magnetic properties of ultrathin magnetic films as a function of the film thickness since the SMOKE measurement is performed at the same position as the deposition without translation of a sample and subsequent realignment of optics. With an externally mounted electromagnet providing an applied magnetic field of up to 2 kOe at the sample position, both the polar as well as the longitudinal SMOKE measurement can be performed by a simple rotation of the sample. This capability is important in determination of the direction of magnetization easy axis for the study of spin reorientation transition. Detailed description of the SMOKE setup has been published elsewhere. A single crystal Pd(111) substrate was sputter cleaned with 1 keV Ar ions and subsequently annealed to obtain a clean, atomically smooth surface. Surface cleanliness was verified with reflection high-energy electron diffraction and STM. Co deposition is carried out from a flux-regulated, well-degassed electron-beam heated rod source at a deposition rate of 0.8 Å/min. The pressure in the chamber remained better than 2 × 10 Torr during deposition. Co deposition rate was calibrated with STM. All growth and measurements were done at room temperature.

After a typical cleaning and annealing procedure, the surface shows large atomically flat terraces as wide as sev-

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eral hundred nanometers as shown in Fig. 1(a). Growth of Co on this smooth surface proceeds initially as a layer-by-layer growth mode gradually turning into three-dimensional (3D)-like structures over 4 ML of coverage as reported earlier. Corresponding evolution of the magnetic properties is summarized in Fig. 2(a), where the polar SMOKE hysteresis loops at each coverage are shown. Ferromagnetic loop appears at Co thickness $>1.5$ ML. Initially the Co film shows perpendicular magnetic anisotropy (PMA) with the magnetization easy axis perpendicular to the film plane. This PMA phase is persistent up to $\sim 4.5$ ML where a spin reorientation transition is observed over a thickness range of 1.5 ML. At 6 ML, the polar SMOKE loop shows a typical hard-axis behavior while the longitudinal SMOKE loop shows a square hysteresis, indicating that the magnetization easy axis now lies parallel to the film plane. STM images obtained at 6 ML coverage show that the surface is characterized with uniformly distributed 3D clusters as demonstrated in Fig. 1(b). Average size of these clusters is $\sim 5$ nm with the rms roughness of 0.26 nm.

For the comparison with the smooth substrate, a rough surface was prepared by sputtering the smooth surface with 1 keV Ar$^+$ ion for 10 min at a near normal incidence. As-sputtered surface was imaged with STM before the Co deposition as seen in Fig. 3(a). The surface still retains overall topographical features such as large terraces and steps but the terrace is no longer atomically flat. They are left with uniform mounds $\sim 7$ nm in size and $\sim 1$ nm in height with the rms roughness of 0.53 nm. These images show close similarities with recent ion sputtering experiments. Frost, Schindler, and Bigl reported the formation of similar mounds on ion sputtered InP surface, which even turned into a highly regular pattern of close-packed mounds for certain sputter conditions.

SMOKE evolution with Co coverage on the roughened surface is illustrated in Fig. 2(b). Onset of ferromagnetic signal is at $\sim 1.5$ ML, and at low coverage the film also exhibits PMA similar to the smooth case. Comparing with the smooth case in Fig. 2(a), the most noticeable difference is the critical thickness at which the spin reorientation transition (SRT) takes place. The SRT begins at smaller thickness of $<3$ ML, and almost ends at 3.5 ML. These comparisons are summarized in Fig. 4. The plot of the ratio of remanence magnetization to the saturated magnetization, $M_R/M_S$, shows clearly the difference in the critical thickness for the SRT. There is also a notable difference in the range of transition.
the transition is gradual over 1.5 ML in the smooth surface, while it is abrupt over a small thickness range of ~0.5 ML in the roughened surface. One might call the SRTs in the rough and smooth cases the first- and second-order phase transition, respectively.\textsuperscript{15} For comparison, Purcell et al.\textsuperscript{16} reported reorientation transition at ~8 ML for the Pd/Co/Pd(111) system while Boukari et al.\textsuperscript{17} observed the transition as early as 2 ML of Co coverage. One possible reason for this variance in the reported transition thickness could be the substrate roughness as demonstrated in this study.

A STM image of 4 ML Co covered rough surface is shown in Fig. 3(b). The surface looks similar to the substrate surface of Fig. 3(a), i.e., uniform distribution of mounds. On closer inspection, the average size of the mounds is now ~10 nm, slightly larger, and average height of them is less than 1 nm. Co deposition actually had a smoothing effect, as can be seen in the line profiles, Figs. 3(c) and 3(d), where the height fluctuations in (c) have almost twice the frequency of that in (d). While the vertical rms roughness of 0.54 nm of this surface is comparable to that of beginning surface, the average size of the mounds is larger. Therefore we consider that Co preferentially fills the gaps of the initial substrate mounds instead of covering mounds and valleys in uniform fashion. This would imply that there are regions of locally thicker Co, mainly in the valley, which will be responsible for the early onset of the SRT in the rough case. Assuming that Co completely fills the valley, local thickness is estimated to be as thick as ~2 nm from the line scans [Figs. 3(c) and 3(d)]. In the case that Co is uniformly deposited according to the rough substrate, one might expect a later onset of the SRT since the magnetic dipolar anisotropy favoring in-plane magnetization is reduced.\textsuperscript{18}

The coercivities of the polar hysteresis loops are compared in Fig. 4(b). Clearly, $H_C$ of the roughened substrate before SRT is much smaller than $H_C$ of smooth substrate. Observed decrease of the coercivity on the rough substrate is in contrast to the general trend that the coercivity increases with surface/interface roughness.\textsuperscript{8-10} Recent theoretical calculations have attributed this general increase of the coercivity with roughness to the roughness-induced demagnetizing effect.\textsuperscript{19} However, these predictions are only valid in the limit where the film thickness is much greater than the interface width. This assumption certainly is not valid for ultrathin films of a few monolayers on artificially roughened substrate with roughness of many layers. In fact, our STM images suggest that the film actually became smoother with the deposition of a few monolayers of Co due to the fact that Co preferentially fills the gaps of the mounds. We believe that this difference in coercivity is related to the different growth morphology and the resulting difference in the magnetization reversal behavior. Co films grown on smooth substrate form a continuous film, where the reversal should proceed via domain nucleation and displacement of domain walls. However, on the roughened surface, Co grows as isolated volumes of clusters filling the gaps, where the reversal should be through the rotation of magnetization of each cluster. STM images reveal that these clusters are at most 7 nm in size, with less than 1 nm in thickness for the measured thickness range, resulting in an estimated volume of ~50 nm\textsuperscript{3}. This is close to the superparamagnetism limit.\textsuperscript{20,21} Thus the observed smaller coercivity can be explained with thermally assisted rotation of these clusters.

In summary, we have compared the evolution of the surface morphology and magnetic properties of ultrathin Co films grown on smooth and artificially roughened Pd(111) substrates. It is shown that the substrate roughness affects the onset of the spin reorientation as well as the coercivity of the ultrathin Co film. STM observation of the substrate and final surface shows that the resulting morphology of the film is responsible for these differences. Our results suggest that the control of substrate roughness is essential for manipulating the thickness range where the magnetization orientation is normal to the film plane, which is desirable for technological applications to perpendicular magnetic recording and magneto-optical recording media.

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