Tunable magnetic anisotropy of ultrathin Co layers

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We prepared multilayers, consisting of an epitaxial Au/Co/Au/Cu(111) architecture on Si(111) substrates, to understand the magnetic anisotropy of an ultrathin Co layer in relation to its structure. The room-temperature magnetization of an 8-monolayer (ML)-thick Co layer is predominately either in-plane or out-of-plane depending upon the thickness of the Au underlayer. Specifically, for a Co film grown on a 2-ML-thick Au, the Co film has a *distorted* fcc structure and in-plane magnetic anisotropy. For a Co film grown on a 6-ML-thick Au, the Co film has a *distorted* hcp structure and out-of-plane magnetic anisotropy. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1850592]

Nanometer scale ferromagnetic films exhibit novel magnetic anisotropy behavior that is of fundamental and technological interest.[1] The magnetic anisotropy of an ultrathin film is known to depend upon several external parameters (magnetic layer thickness,[2,3] adjacent nonmagnetic overlayer[2–6] and underlayer,[4,5] temperature,[3] and etc.). For example, Matthes *et al.*[7] studied the influence of alloy composition and thickness on the magnetic properties of ultrathin Ni_xPd_{1-x} alloy films and found that magnetic anisotropy changes from in-plane to out-of-plane with increasing Ni concentration due to the magnetoelastic stress at the interface between film and substrate. A second example involves the interplay between magnetic anisotropy and atomic structure for Fe films grown on Cu(001).[8–10]

Previously, we observed a change of magnetic anisotropy of Co/Au to depend upon the thickness of the Au layer.[4,5] Typically, the magnetic anisotropy of a thin film is in the sample plane (in-plane) in order to minimize demagnetization fields—a consequence of shape anisotropy.[11] However, a competition among shape, magnetocrystalline, and magnetoelastic anisotropies may result in a film having magnetic anisotropy that is perpendicular to the sample plane (out-of-plane).[5] In particular, owing to the large lattice mismatch between Au and Co, epitaxial films of these materials are highly strained; consequently, the magnetic anisotropy of Co might be influenced by strain. Here we investigate the detailed atomic structure of Co films for the different Au underlayer thicknesses. We found that indeed a correlation between the structure of Co and the magnetic anisotropy of the Co/Au system exists. Specifically, we show that the structure of an 8-monolayer (ML)-thick Co film can be controlled by the thickness of the Au underlayer on which the Co film is grown. We further show that the magnetic anisotropy of the Co film depends upon the structure of the film, and therefore, by controlling the structure of the film, we can control the anisotropy of the Co thin film.

For substrates, we used nonmiscut Si(111) wafers, which were immersed in a 2% hydrofluoric acid solution for 2 min prior to film deposition. Once in the molecular beam epitaxy chamber, the substrates were heated to 750 °C for 10 min to eliminate any remaining oxide, hydrogen, and residue, all of which affect the surface quality. After annealing, reflected high-energy electron diffraction (RHEED) and low-energy electron diffraction showed the 7×7 reconstruction of the Si(111) surface, confirming the surface quality.[12] A 4-nm-thick Cu buffer layer was deposited on the Si substrate[13] followed by Au underlayers of various thicknesses, using a computer controlled shutter. The thickness of the Au underlayers varies from 2 to 6 ML (1 ML = 0.236 nm). An 8-ML-thick Co layer (1 ML = 0.204 nm) was then grown on the Au, followed by a thick layer of Au to protect the sample from oxidation. The deposition rate was 0.01 nm/s for all layers with the exception of Cu which was deposited at 0.04 nm/s and all the depositions were performed at ambient temperature.[12]

The bulk magnetizations of the samples were obtained by means of a superconducting quantum interference device susceptometer and by Brillouin light scattering (BLS). *Ex situ* BLS experiments were performed at room temperature using a 532 nm frequency-doubled diode pump Nd-YAG solid state laser light source. *P*-polarized light was focused with a 50 nm focal length camera lens onto the sample at an incident angle of 45°. The diffusely scattered light was collected by the same lens in backscattering geometry, then passed through a cross-polarization analyzer, and analyzed with a 3+3 pass tandem Fabry–Perot interferometer.[14] Atomic structures of the samples were obtained from x-ray diffraction, to determine out-of-plane strain, and high-resolution transmission electron microscope (HRTEM) to identify in-plane structures.

Figure 1 shows the effective magnetization density $[(4\pi M_s)_{\text{eff}}=4\pi M_s-2K_1/M_s]$[15,16] as a function of Au underlayer thickness for a Co 8 ML sample, where M_s is the saturation magnetization of bulk Co (1422 emu/cm^3)[17] and K_1 is the first-order uniaxial anisotropy constant obtained BLS at room temperature. The effective magnetization density is a useful quantity to visualize the magnetization direction of the sample for zero external field. A positive (negative) value of $(4\pi M_s)_{\text{eff}}$ indicates that the magnetic moment of the sample is in-plane (out-of-plane). For the sample with a 2-ML-thick

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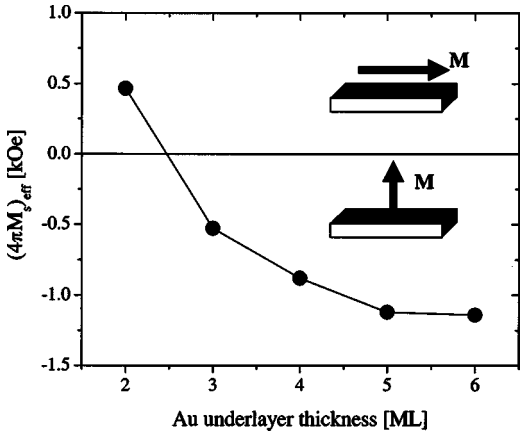

FIG. 1. Room-temperature effective magnetization density as a function of Au underlayer thickness for an 8-ML-thick Co film sandwiched between an *X*-ML-thick Au underlayer and 3.5-nm-thick Au overlayer. The negative (positive) $(4\pi M_s)_{\text{eff}}$ indicates that the magnetic moment of the sample is out-of-plane (in-plane) in zero external field. The solid line is a guide to the eye.

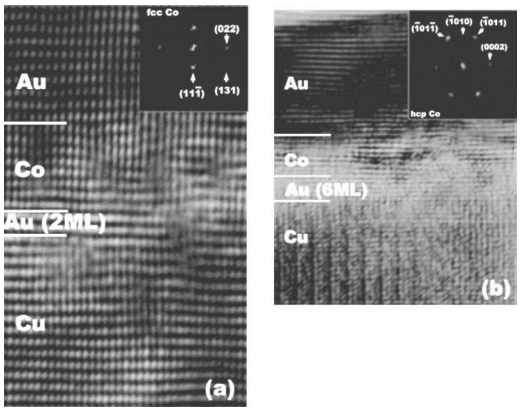

FIG. 3. Cross-sectional HRTEM micrographs of (a) Cu 4 nm/Au 2 ML/Co 8 ML/Au 3.5 nm, and (b) Cu 4 nm/Au 6 ML/Co 8 ML/Au 3.5 nm samples along Si $[\bar{1}10]$ zone axis. The insets show SAD patterns of the 8-ML-thick Co layer. For the 2-ML-thick Au sample, Co exhibits a distorted fcc structure, with a diffraction zone axis of $[2\bar{1}1]$. However, for the 6-ML-thick Au sample, Co has a distorted hcp structure, with a diffraction zone axis of $[1\bar{2}10]$.

Au underlayer, the 8-ML-thick Co exhibits an easy axis that is in the sample plane. However, for the sample with a 3-ML-thick Au underlayer, $(4\pi M_s)_{\text{eff}}$ is negative for the 8-ML-thick Co film indicating the Co film has out-of-plane anisotropy. The effective magnetization density continues to decrease as the Au underlayer thickness increases, reaching a constant value when the Au underlayer is more than 5 ML thick.

Figure 2 shows the room-temperature ferromagnetic hysteresis loop of an 8-ML-thick Co film with (a) 2-ML- and (b) 6-ML-thick Au underlayer, respectively. The external field was applied either along the sample surface (■) or perpendicular to the sample surface (●) for the magnetization measurements. The sample with a 2-ML-thick Au underlayer [i.e., Si (111)/Cu 4 nm/Au 2 ML/Co 8 ML/Au 3.5 nm] exhibits an easy axis in the plane of the sample, consistent with our BLS measurements shown in Fig. 1 [i.e., $(4\pi M_s)_{\text{eff}}$ is positive], as evidenced by the square hysteresis loop when the external field is applied along the sample surface. For the sample with a 6-ML-thick Au underlayer [Si (111)/Cu 4 nm/Au 6 ML/Co 8 ML/Au 3.5 nm], the hysteresis loop shows a square loop (○) when the external field is applied to the perpendicular to the sample surface. Thus, for this sample, the 8-ML-thick Co film has out-of-plane anisotropy at room temperature, consistent with the negative value of $(4\pi M_s)_{\text{eff}}$ in Fig. 1.

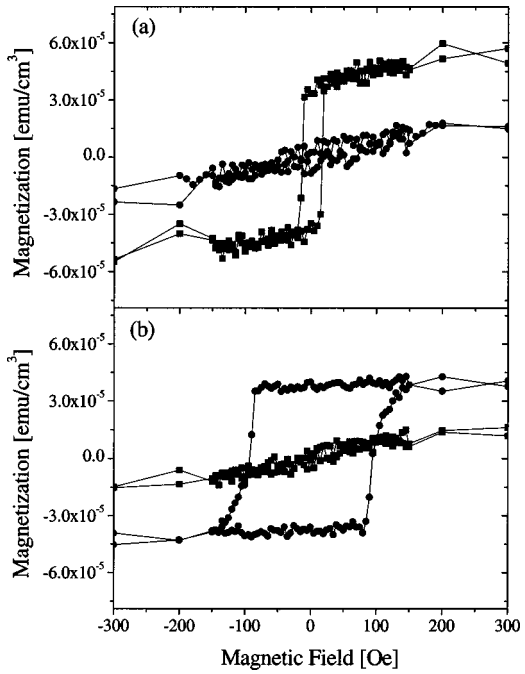

FIG. 2. Magnetization of the 8-ML-thick Co film (a) with 2-ML-thick Au underlayer (b) with 6-ML-thick Au underlayer. The magnetization loop is measured while the external magnetic field is applied either along the sample surface (■) or perpendicular to the sample surface (●).

A capability of thin-film technology is that unusual phases can be grown in thin-film forms that are not found in bulk. For example, a bcc Co film can be grown on top of GaAs(110) substrates,[18,19] an fcc Co can be grown on top of Cu(111) substrates.[20] However, one complication of thin films is that their structures can be readily influenced by strain, which in turn may affect the magnetic properties of the film. Therefore, it is important to investigate the structure of Co on the thick and thin Au underlayers we have used. We employed cross-sectional HRTEM for a detailed analysis of the in-plane structure of these samples, since (0001) hcp growth cannot be distinguished from (111) fcc growth with RHEED. Further, we should note that reconstruction of the underlying part of the Co film with continued deposition of Co or Au layers can not be observed with RHEED.[21] Cross-sectional HRTEM micrographs shown in Fig. 3 outline the stacking sequence of the samples with 2-ML-thick Au [Fig. 3(a)] and 6-ML-thick Au [Fig. 3(b)] samples, respectively. For both cases, the transmitted electron beam is parallel to the Si $[\bar{1}10]$ zone axis, and the horizontal atomic planes shown in the figure are parallel to the (111)Si surface. For the structure analysis, selected-area diffraction (SAD) patterns of individual layers (Cu, Co, and Au capping layers) and a Fourier analysis of the patterns, using the pattern observed from the Si substrate as a calibration, were used to obtain the interplanar spacings and angles of the each layer. For the 2-ML-thick Au sample [Fig. 3(a)], the SAD pattern of Co is rectangular [see the inset of Fig. 3(a)] with an fcc zone axis

of $[2\bar{1}1]$. Similar rectangular patterns are also observed for the Cu buffer layer and Au overlayer (figures for Cu and Au are not shown here). The crystallographic orientation relation for the thin sample is: Si $[\bar{1}10]\|$Cu $[2\bar{1}1]\|$Co $[2\bar{1}1]\|$Au $[2\bar{1}1]$ and Si $(111)\|$Cu $(11\bar{1})\|$Co $(11\bar{1})\|$Au $(11\bar{1})$. This orientation relationship was determined by electron diffraction from two cross-sectional TEM specimens: one cut normal to Si $\langle 110\rangle$ and other cut normal to Si $\langle 112\rangle$, where Si $\langle 110\rangle$ and Si $\langle 112\rangle$ are two orthogonal directions in the plane of the Si (111) wafer. We performed similar studies on the sample with 6 ML Au, and its HRTEM micrograph and Co SAD pattern are shown in Fig. 3(b). The SAD pattern of Co for the sample with 6 ML thick Au layer [see the inset of Fig. 3(b)] suggests structure of the Co is nearly ideal hcp with $[1\bar{2}10]$ zone axis while the Cu and Au layers remain the $[2\bar{1}1]$ zone axis. Therefore, the crystallographic orientation relations for the thick sample are: Si $[\bar{1}10]\|$Cu $[2\bar{1}1]\|$Co $[1\bar{2}10]\|$Au $[2\bar{1}1]$, and Si $(111)\|$Cu $(11\bar{1})\|$Co $(10\bar{1}0)\|$Au $(11\bar{1})$. Similar observation had been made by Lee *et al.*,[19] in their Co/Au superlattice, which was grown on top of GaAs(110). They observed that after 6-ML-thick Au is deposited on top of the bcc Co, succeeding Co layers showed hcp structure in spite of the large lattice mismatch between Co and Au.

It is noteworthy that 8-ML-thick Co layer exhibits a distorted hcp or distorted fcc structure depending upon that of the Au layer thickness: 6 or 2 ML, respectively. For instance, the SAD pattern for the Co portion of the 2-ML-thick Au sample has $2\% \sim 6\%$ distortion compared to that of bulk fcc Co.[22] We also compared the Co SAD pattern to that of bulk hcp Co. In this case, the amount of distortion would be more than 20%. Thus, we conclude that 8-ML-thick Co is likely fcc rather than hcp for the 2-ML-thick Au sample. For the 6-ML-thick Au sample, the Co film is only minimally distorted from bulk hcp Co. Not shown here are measurements of the out-of-plane strain profiles obtained by x-ray diffraction, which further confirm the conclusions obtained from microscopy.

In summary, we observed that the magnetization of 8-ML-thick Co layer can be tuned to be in-plane to out-of-plane by varying the Au underlayer thickness from 2 to 6 ML. The thickness of the Au underlayer determines whether the Co film on which it is grown has either a *distorted* fcc or *distorted* hcp structure. We attribute the difference in Co structures to strain at the Co/Au interface.

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