Changes in the Magnetic Anisotropy of Co Thin Films on Pt(111) Capped by Ag Overlayers

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Two unusual magnetic properties were observed when Co ultrathin films on Pt(111) were capped with Ag overlayers. The perpendicular magnetic anisotropy was significantly enhanced after annealing at high temperatures, and the spin reorientation transition (SRT) could be induced by the Ag overlayers. The perpendicular magnetic anisotropy of 1 ML Ag/1–3.5 monolayers (ML) Co/Pt(111) was enhanced dramatically after Co–Pt alloy formation. The easy axis of the magnetization was reoriented from the in-plane direction to the out-of-plane direction, when Ag overlayers were deposited on 3.5–7 ML Co/Pt(111). The possible mechanisms for the magnetic anisotropy enhancement and the SRT are discussed.

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I. INTRODUCTION

The study of sandwiches and multilayers of ferromagnetic ultrathin films has the prospect of improving high-density magneto-optical devices and magneto resistive sensors and is also fundamental to basic science research [1, 2]. A system containing Ag, Co, and Pt is of particular interest. Co–Pt ultrathin films and alloys have a perpendicular easy axis of magnetization, high coercivity, and large magneto-optical Kerr signal [3, 4]. The adatoms of Ag form a well-ordered state after the formation of the Co–Pt alloy is complete [5]. Ag-Co thin films exhibit the phenomena of giant magnetoresistance and superparamagnetism [6, 7]. The coercivity and the remanence of the Co/Ag multilayers are a function of the annealing temperature, and the coercivity reaches its maximum after being annealed at 400°C [8]. It is interesting to investigate the changes in the magnetic properties after Ag thin films have been capped on Co/Pt(111) and annealed at high temperatures.

In this study, the thickness of the Ag and the Co layers was carefully controlled. The changes in the magnetic anisotropy of the ultrathin films with different coverages were measured at different annealing temperatures. After being capped by ultrathin Ag films, the perpendicular magnetic anisotropy of the Co ultrathin films was significantly enhanced when annealed at a high temperature. Furthermore, the phenomenon of a spin reorientation transition (SRT) induced by the Ag overlayers could be observed. If the Co coverage is thicker than 3.5 monolayers (ML), the easy axis of the magnetization can change from the in-plane direction to the out-of-plane direction after being capped by the Ag overlayer.
II. EXPERIMENT

All the experiments were carried out in situ in an ultra-high vacuum (UHV) chamber. The working background pressure was better than $5 \times 10^{-10}$ Torr. The chamber was equipped with several facilities for the preparation and analysis of thin films and surfaces. The surface structure was observed by low-energy electron diffraction (LEED). A computer-controlled image processor measured the intensity of the diffracted spots in the LEED. The chemical composition of the surface was measured by Auger electron spectroscopy (AES). A hemispherical electron energy analyzer was used in the AES. The surface of the sample was cleaned by the standard Ar ion bombardment and annealing cycles in the UHV chamber. The bombarding kinetic energy was 2 keV and the annealing temperature reached 1100 K. In order to remove residual carbon on the Pt surface, oxygen was introduced at a pressure of $1 \times 10^{-7}$ Torr while the sample was kept at 800 K. The cleaning cycles were repeated until a sharp and well-ordered $p(1 \times 1)$ LEED pattern of the Pt(111) surface was observed.

The Ag and Co purities were 99.999% and 99.997%, respectively. Ag and Co thin films were deposited by means of molecular beam epitaxy (MBE). The coverage of Ag and Co was monitored by the relative evolutions of the Ag, Co, and Pt Auger signals and was double checked by a quartz balance thickness monitor. The Co and Ag deposition rates were 900 s/ML and 320 s/ML at room temperature, respectively. The magnetic properties were studied in situ by the magneto-optic Kerr effect (MOKE). The Kerr signal is sensitive to the magnetization of ferromagnetic thin films in the monolayer range [9, 10]. A He-Ne laser with a wavelength of 632.8 nm was used as the light source for the MOKE study.

III. RESULTS AND DISCUSSION

Cobalt atoms can grow 3.5 ML in a layer-by-layer mode on a Pt(111) surface at room temperature [11]. Ag atoms deposited on the 1–3 ML Co/Pt(111) will initially grow 2 ML in a layer-by-layer mode and then change to the three-dimensional (3-D) island growth mode [12]. The easy axis of the magnetization is in the out-of-plane direction when the growth of Co thin films on Pt(111) is in the layer-by-layer growth mode, and turns to in-plane gradually when it is in the 3-D island growth mode [9, 10, 13]. The polar Kerr intensity ($I_K$) versus magnetic field ($H$) of 1 ML Co/Pt(111) is shown in Fig. 1 (a). After being capped by 1 ML of Ag on 1 ML Co/Pt(111), the $I_K-H$ curves are shown in Fig. 1 (b). Comparing these two polar hysteresis curves, shows that the coercivity ($H_C$) increases from 280 Oe to 745 Oe after a 1 ML Ag overlayer is deposited. The polar hysteresis curves for 2 ML Co/Pt(111) before and after depositing a 1 ML Ag overlayer are shown in Fig. 1 (c) and Fig. 1 (d), respectively. $H_C$ increases from 475 Oe to 585 Oe. No longitudinal hysteresis loop was observed for 1 and 2 ML Co/Pt(111), whether the Ag overlayer was deposited or not. The shapes of all the hysteresis curves shown in Fig. 1 are close to a square. This indicates that the easy axis of the magnetization is in the out-of-plane direction. The saturated polar intensity does not significantly change when the Ag overlayers are capped, but $H_C$ increases significantly when a 1 ML Ag overlayer is capped on the Co thin film. It
FIG. 1: The hysteresis curves measured at room temperature with a polar MOKE configuration. (a) 1 ML Co/Pt(111), (b) 1 ML Ag/1 ML Co/Pt(111), (c) 2 ML Co/Pt(111), (d) 1 ML Ag/2 ML Co/Pt(111).

It is interesting that the increase in $H_C$ after depositing the 1 ML Ag overlayer becomes small when the Co coverage increases.

We annealed the 1 ML Ag/2 ML Co/Pt(111) thin film at different temperatures for 5 minutes each, then measured the corresponding Kerr signal at the annealing temperature. The polar Kerr intensity versus the temperature during the temperature raising process is shown in curve 1 of Fig. 2. The Kerr signal has an anomalous enhancement from $T = 600$ K, and reaches a maximum at 700 K. Then $I_K$ decreases rapidly, reaching zero at 780 K. The Curie temperature of this system is about 780 K. After carefully studying the AES signals of Co, Ag, and Pt in the annealing process, we found that the Pt signal increased and the Co signal decreased significantly, while the Ag signal remained constant for temperatures between 600 K and 700 K. These results are similar to our previous report on a 1 ML Ag/1 ML Co/Pt(111) system [12]. We conclude that the enhancement of the polar Kerr intensity occurs due to the Co–Pt alloying formation.

An unusual phenomenon occurs during the cooling process as shown in curve 2 of Fig. 2. $I_K$ increases when the sample temperature decreases, reaching a maximum at 450 K, but $I_K$ decreases with temperature when $T < 450$ K. $I_K$ approaches zero at room temperature; i.e. the magnetization disappears near room temperature. It is especially interesting that the magnetization versus temperature described in curve 2 is reversible. No in-plane magnetic anisotropy is observed during the process. The magnetic property
FIG. 2: The polar Kerr intensity versus the sample temperature for 1 ML Ag/2 ML Co/Pt(111). Curve 1: the Polar Kerr intensity has an anomalous enhancement after T>600 K and reaches a maximum at 700 K during the first heating process. Curve 2: In the cooling process, the Polar Kerr intensity reaches a maximum at 450 K, and drops close to zero at room temperature. It is a reversible process.

The magnetization is almost in the in-plane direction when the coverage of the Co
FIG. 3: The hysteresis curves measured at room temperature. (a) 4 ML Co/Pt(111), (b) 1 ML Ag/4 ML Co/Pt(111), (c) 5 ML Co/Pt(111), (d) 1 ML Ag/5 ML Co/Pt(111). The polar hysteresis loop appears after depositing the 1 ML Ag overlayer.

The Kerr intensity versus the coverage of Ag for 4 ML Co/Pt(111) is shown in Fig. 4(a). The longitudinal Kerr intensity decreases monotonically to zero as the Ag coverage increases. It almost disappears after the Ag coverage is greater than 0.5 ML. The polar Kerr intensity rises very fast when the coverage of Ag is between 0.25 ML and 0.75 ML. The SRT switching becomes slow after the Ag coverage becomes greater than 0.75 ML. This slow switching can be interpreted as the gradual rotation of the easy axis of thin film is larger than 3.5 ML. The 4 ML Co/Pt(111) hysteresis curves are shown in Fig. 3(a). Only a longitudinal Kerr signal was detected, no polar Kerr signal was found. After a 1 ML Ag overlayer was deposited on the 4 ML Co/Pt(111) surface, the longitudinal Kerr intensity disappeared and the polar hysteresis loop appeared as shown in Fig. 3(b). A similar phenomenon was observed for 5 ML Co/Pt(111). The hysteresis curves before the deposition of Ag are shown in Fig. 3(c). Only the longitudinal Kerr effect is obvious. The polar hysteresis in a square shape appeared after a 1 ML Ag overlayer was deposited on the 5 ML Co/Pt(111), as shown in Fig. 3(d). These results indicate that the easy axis of the magnetization has switched from the in-plane direction to the out-of-plane direction after the 1 ML Ag overlayer is capped. A spin reorientation transition for Co thin films on a Pt(111) surface can be induced by Ag overlayers. Since the property of the perpendicular magnetization anisotropy can be used in high-density magneto-optic recording, this observation is especially interesting.

The Kerr intensity versus the coverage of Ag for 4 ML Co/Pt(111) is shown in Fig. 4(a). The longitudinal Kerr intensity decreases monotonically to zero as the Ag coverage increases. It almost disappears after the Ag coverage is greater than 0.5 ML. The polar Kerr intensity rises very fast when the coverage of Ag is between 0.25 ML and 0.75 ML. The SRT switching becomes slow after the Ag coverage becomes greater than 0.75 ML. This slow switching can be interpreted as the gradual rotation of the easy axis of
FIG. 4: (a) The longitudinal and polar Kerr intensity as a function of Ag coverage on 4 ML Co/Pt(111). The polar Kerr intensity increases quickly when \(d_{Ag} = 0.5\) ML, and saturates when \(d_{Ag} = 1\) ML. (b) The Coercivity as a function of Ag coverage on 5 ML Co/Pt(111). The \(H_C\) maximum is located at the coverage of 1 ML Ag.

the magnetization from the in-plane direction to the out-of-plane one, due to competition between the shape anisotropy and the uniaxial interface anisotropy [14]. The polar Kerr intensity approaches saturation when the Ag coverage is 1.0 ML. Since all the Co sites are completely occupied by Ag atoms when the Ag coverage is 1.0 ML, the increasing thickness of Ag overlayers larger than 1.0 ML does not further increase the polar Kerr intensity. \(H_C\) of the out-of-plane magnetization versus the Ag coverage is shown in Fig. 4(b). \(H_C\) increases when the Ag coverage increases, reaching a maximum when the Ag coverage is 1.0 ML. It is interesting that both the polar Kerr intensity and \(H_C\) reach their peak value when the Ag coverage is 1.0 ML. The behavior of \(H_C\) versus \(d_{Ag}\) is interesting. Engel et al. [15] proved that \(H_C\) has the same tendency as that of the anisotropy constant \(K_1\) in a Cu/Co/Pd(111) system. Since the behavior of \(H_C\) in our system is similar to that of the Cu/Co/Pd(111) system, we believe that in our system \(K_1\) increases monotonically when the Ag coverage is less than 1 ML, and decreases slightly when the Ag coverage is larger than 1 ML.

Fig. 5 shows the dependence of \(H_C\) as a function of the sample’s temperature. \(H_C\) is about 375 Oe for 1 ML Ag/4 ML Co/Pt(111). We measured \(H_C\) at each temperature after the system was in thermal equilibrium. Usually \(H_C\) decreases when the temperature of a ferromagnetic thin film increases, because the thermal energy can help to reverse the magnetization of the magnetic domains. \(H_C\) in this system is interesting. During the heating process, \(H_C\) decreases drastically between 300 K and 350 K. The trend of the decrease slows when T is higher than 350 K. The curve for \(H_C\) versus T is totally different for the cooling down process. \(H_C\) increases significantly after the annealing, from 375 Oe to 430 Oe at room temperature. This shows that the annealing at a high temperature has caused an increase of \(H_C\). In our AES study, the formation of Co–Pt alloy causes the
increase of $H_C$.

The SRT induced by Ag can occur when the Co coverage is less than 7 ML. However, only parts of the Co spins can be reoriented if the coverage of Co is higher than 7 ML. According to our observations, both the in-plane and the out-of-plane magnetization appear regardless of the thickness of the Ag deposition, when the coverage of Co is larger than 7 ML. It is interesting to know what the mechanism is, for the SRT induced by Ag, in this system. If we neglect the high order terms, the anisotropy energy density can be written as $E = K_{eff} \sin^2 \theta$, where $\theta$ is the angle between the magnetization and the surface normal. The direction of the easy axis of the magnetization can be determined from the minimum requirement of $E$. The effective anisotropy [16] of the uncovered Ag film can be written as

$$K_{eff} = K_V + \frac{K_{S}^{Co-Pt}}{d_{Co}} + \frac{K_{S}^{UHV-Co}}{d_{Co}},$$

(1)

where $K_V$ is the volume anisotropy, $K_{S}^{Co-Pt}$ is the interface magnetic anisotropy between Co and Pt, and $K_{S}^{UHV-Co}$ is the interface magnetic anisotropy between the vacuum and Co. We can assume that the volume anisotropy and the interface anisotropy between Co and the Pt substrate do not change after Ag is deposited on the Co/Pt(111) surface. Then the effective anisotropy when the coverage of Ag is 1 ML can be written as

$$K_{eff} = K_V + \frac{K_{S}^{Co-Pt}}{d_{Co}} + \frac{K_{S}^{Ag-Co}}{d_{Co}}.$$

(2)

$K_{S}^{Ag-Co}$ is the interface magnetic anisotropy between Ag and Co. Comparing equations (1) and (2), the $K_{S}^{UHV-Co}$ term is just replaced by the $K_{S}^{Ag-Co}$ term after the Ag overlayer is
deposited. Since $K_{S}^{UHV-Co}$ is negative [16] and $K_{S}^{Ag-Co}$ is positive [17], $K_{eff}$ changes the signal from negative to positive when a 1 ML Ag overlay exists. Therefore, the in-plane magnetization changes to out-of-plane magnetization. If Co is over 7 ML, $K_V$ is dominant and $K_{eff}$ is always negative. The direction of the magnetization is in-plane, no matter how thick the Ag is capped.

From the above discussion, the increase of the interface anisotropy after Ag is capped on Co thin films is the main physical reason for the SRT. It is interesting to discuss the mechanism for the changes in the interface anisotropy further. The lattice mismatch between Ag and Co is large (the nearest-neighbor distances are $a_{Co} = 0.251$ nm, $a_{Ag} = 0.289$ nm). The strain causes the interface anisotropy to change its sign from negative for $K_{S}^{UHV-Co}$ to positive for $K_{S}^{Ag-Co}$. Another possible physical origin is that the hybridization of the electronic states at the Ag/Co interface could cause significant alternations of the magnetic interface anisotropy [18, 19]. Indeed, for the electron structures of Ag ultrathin films on Co/Pt(111), some important changes have been observed by ultraviolet photoemission spectroscopy [12]. The peak height of the Fermi edge decreases and the binding energy of the $d$-band changes from 4.0 eV to 4.5 eV and 5.3 eV double peaks after Ag ultrathin films are deposited. The hybridization of the Ag and Co wave functions changes the magnetic anisotropy. Weber et al. reported that a strong change of the magnetic anisotropy, induced by hybridization of the Cu and Co wave functions, switches the in-plane magnetization direction of 90° for the Cu/Co/Cu(100) system [20]. Both the strains due to the large lattice mismatch and the hybridization of electron states at the Ag/Co interface are possible mechanisms for the SRT in our system.

IV. CONCLUSIONS

The perpendicular magnetic anisotropy shows a significant enhancement during high temperature annealing, after Ag was capped on to a Co/Pt(111) surface. The hybridization of the electronic states of Co and Pt due to interface alloy formation, and the uniaxial interface anisotropy between Ag and Co, are possible mechanisms for the enhancement. After a 1 ML Ag overlay was deposited on a 3.5–7 ML Co/Pt(111) surface, the easy axis of the magnetization was reoriented from the in-plane to the out-of-plane direction. Both strains due to the large lattice mismatch and the hybridization of the electron states at the Ag/Co interface are possible mechanisms for the SRT.

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