Ag-induced spin-reorientation transition of Co ultrathin films on Pt(111)

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The surface magneto-optical Kerr effect was used to study the magnetic properties of Co ultrathin films deposited on Pt(111). The easy axis of the magnetization changes from the out-of-plane to the in-plane direction after the coverage of Co is larger than 3.5 ML. The spin can reorient to the normal of the surface when the proper thickness of Ag overlayers is deposited on Co/Pt(111) with the in-plane magnetization. The out-of-plane magnetization and its coercivity as a function of Ag coverage were investigated during the spin-reorientation transition. The easy axis of the magnetization can shift back to the in-plane direction after the Ag overlayers are sputtered out. The chemical compositions of the interfaces were measured by Auger electron spectroscopy. The mechanism of the spin-reorientation transition induced by Ag is discussed.

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I. INTRODUCTION

The studies of the properties of ferromagnetic ultrathin-film sandwiches and multilayers have recently attracted much interest experimentally as well as theoretically. One reason for the interest is that the properties of perpendicular magnetic anisotropy and the spin-reorientation transition may apply to magnetic high-density recording, magnetic thin-film sensors, and magnetic thin-film switches.[1,2] A system containing Ag, Co, and Pt is particularly intriguing. Co-Pt ultrathin films and alloys have a perpendicular easy axis of magnetization, high coercivity, low Curie temperature and a large magneto-optical Kerr signal.[3–5] Ag-Co thin films exhibit the phenomena of giant magnetoresistance and superparamagnetism.[6–8] It is interesting to investigate the changes of the magnetic properties after Ag thin films are deposited on top of Co/Pt(111). During the study, the phenomenon of a spin-reorientation transition induced by Ag overlayers was observed. If the coverage of Co on the Pt(111) surface is higher than 3.5 ML, the easy axis of the magnetization changes from the out of plane to the in plane. After Ag overlayers are deposited, the easy axis of the magnetization shifts back to the out of plane. The phenomenon of the spin-reorientation transition occurs usually by changing the thickness of the ferromagnetic thin films[9–13] or tuning the substrate temperature.[2,14–16] Most studies in spin-reorientation transition focused on Fe or Ni thin films. As we know, this study is a report on spin-reorientation transition induced by heterogeneous overlayers in the Co-Pt system.

II. EXPERIMENTAL DETAILS

The experiments were carried out in an ultrahigh-vacuum chamber with the working base pressure about 5×10^{-10} Torr. The single-crystal Pt was polished within 0.5° along the (111) direction. The surface of the sample was cleaned by standard Ar-ion bombardment and annealing cycles in the ultrahigh-vacuum chamber. The bombarding kinetic energy was 2 keV and the annealing temperature reached 1100 K. In order to remove the residual carbon from the Pt surface, oxygen was introduced at a pressure of 1×10^{-7} Torr and the sample was heated to 800 K. The surface was well prepared until a sharp and well-ordered $p(1\times1)$ low-energy electron-diffraction pattern of the Pt(111) surface was observed.

The chamber was equipped with several facilities for the preparation and analysis of thin films and surfaces. The surface structure was obtained by four-grid video low-energy electron diffraction. The chemical compositions of the surface were measured by Auger electron spectroscopy. The purities of Ag and Co were 99.999% and 99.997%, respectively. Ag and Co thin films were deposited by molecular-beam epitaxy. The coverage of Ag and Co was monitored by the relative evolutions of Ag, Co, and Pt Auger signals and was doubly checked by a quartz balance thickness monitor. The deposition rates of Co and Ag were 900 s per monolayer (ML) and 320 s per ML at room temperature, respectively. The magnetic properties were studied *in situ* by the surface magneto-optical Kerr effect. A He-Ne laser with a wavelength of 632.8 nm was used as the light source for the magneto-optical Kerr effect study.

III. RESULTS AND DISCUSSIONS

Cobalt atoms can grow 3 ML in a layer-by-layer mode on a Pt(111) surface at room temperature.[17] Ag atoms deposited on the 1–3-ML Co/Pt(111) initially grow 2 ML in a layer-by-layer mode and then change to the three-dimensional (3D) island growth mode.[18] The polar and longitudinal Kerr signals versus magnetic field H for different thicknesses of Co on Pt(111) are shown in Fig. 1. Only the polar hysteresis loop was observed when the coverage of Co was less than 3.5 ML. The hysteresis loop shifts gradually to the longitudinal configuration when the coverage of Co is larger than 3.5 ML. It is interesting that the longitudinal Kerr signal starts to appear when the growth of Co thin films turns to the 3D island growth mode. The longitudinal Kerr intensity is stronger than the polar one when the coverage of Co is 4.5 ML. Only the longitudinal hysteresis loop was observed when the coverage of Co was larger than 5.0 ML. The Kerr intensity can be used to represent the magnetization of the measured system since they have a linear relation.[3,5] The result of Fig. 1 indicates that the easy axis of the magnetization of Co ultrathin films is in the out-of-plane direction in the initial growth and then changes gradually to the in plane.

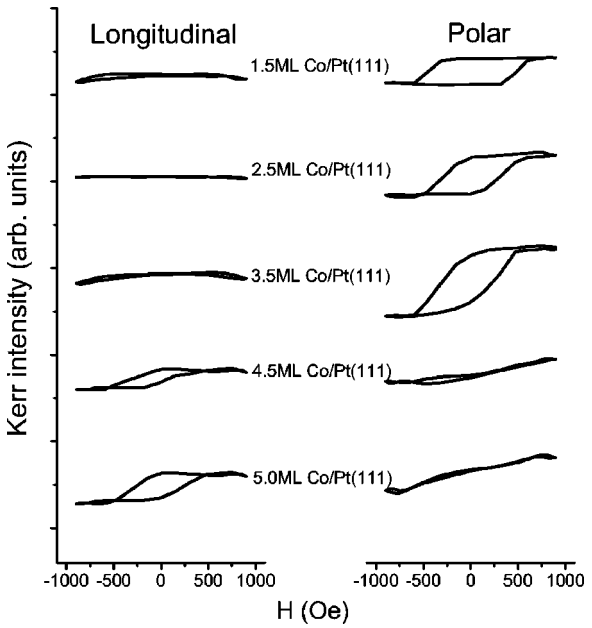

FIG. 1. Kerr hysteresis loops in the polar and the longitudinal configurations versus magnetic field at different coverages of Co deposited on the Pt(111) surface. The easy axis of the magnetization is out of plane when the coverage of Co is less than 3.5 ML and it changes to in plane after the coverage of Co is larger than 5 ML.

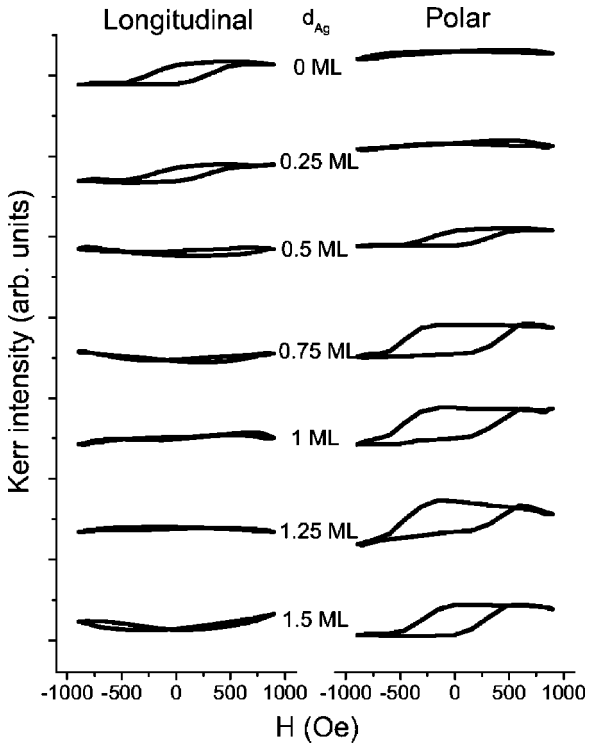

FIG. 2. Kerr hysteresis loops versus magnetic field after Ag overlayers are deposited on 5-ML Co/Pt(111) with increments of 0.25 ML each time. The polar hysteresis loop appears gradually as Ag coverage increases.

As the thickness of the Co thin film increases to 5.0 ML, the magnetization is completely in the in-plane direction.

Ag overlayers were then deposited on the 5-ML Co/Pt(111) surface. The deposition of Ag was stopped after each 0.25 ML in order to perform the Kerr measurement. The hysteresis curves a shown in Fig. 2. The longitudinal Kerr intensity decreases and the polar Kerr intensity increases gradually when the silver thickness d_{Ag} increases. The polar Kerr intensity significantly increases during Ag deposition, and increases significantly when the coverage of Ag reaches 0.5 ML. The shape of the polar hysteresis loop becomes squarer when $d_{\mathrm{Ag}}=0.75$ ML. For subsequent Ag deposition at $0.75\ \mathrm{ML} \leqslant d_{\mathrm{Ag}} \leqslant 1.5\ \mathrm{ML}$, the polar Kerr intensity increases more slowly while the longitudinal Kerr intensity always remains at zero. These results indicate that the easy axis of the magnetization switches from the in-plane to the out-of-plane direction after the Ag overlayers are thick enough, i.e., a spin-reorientation transition can be induced by Ag overlayers.

The Kerr intensity versus the coverage of Ag is plotted in Fig. 3(a). The longitudinal Kerr intensity decreases to zero monotonously as Ag coverage increases, while the polar Kerr intensity starts to grow when Ag coverage is higher than 0.25 ML. The polar Kerr signal arises very quickly when the coverage of Ag is near 0.5 ML. The spin-reorientation-transition switching becomes slow after the coverage of Ag is larger than 0.75 ML. The slow switching can be interpreted as the gradual rotation of the easy axis of the magnetization from the in-plane direction to the out-of-plane one due to the competition between the shape anisotropy and the uniaxial interface anisotropy.[19] Polar Kerr intensity approaches saturation when Ag coverage is 1.0 ML. Since all the Co sites are completely occupied by Ag atoms when the coverage of Ag is 1.0 ML, the increasing thickness of the Ag overlayers larger than 1.0 ML does not further increase the polar Kerr intensity. We define the coverage of Ag as a critical thickness when half of the total spins of Co atoms are reoriented. The critical thickness of 5-ML Co/Pt(111) is 0.5 ML.

The coercivity, H_C, of the out-of-plane magnetization versus Ag coverage is shown in Fig. 3(b). Ag overlayers on the Co/Pt(111) surface can result in the increase of H_C due to the pinning of the spin orientation of Co.[20] Therefore H_C increases when the coverage of Ag increases. H_C reaches the maximum when the coverage of Ag is 1.0 ML. However, it is interesting to discuss why H_C increases when the coverage of Ag is between 0 and 1.0 ML, decreases when the coverage of Ag is higher than 1.0 ML. Engel *et al.*[21] studied the system and in which Co films were epitaxially grown at room temperature on thick, 30–50-nm, Pd(111) buffer layers deposited onto Co-seeded GaAs(110). After a capped Cu overlayer, H_C increases with increasing magnetic anisotropy. In their study, H_C versus the thickness of a Cu overlayer d_{Cu} had the same tendency as that of the anisotropy constant K_1 versus d_{Cu}. From their measurements, H_C also increased to a maximum at around 1 ML and then slightly decreased for $1\ \mathrm{ML} < d_{\mathrm{Cu}} < 5$ ML. The behavior of H_C versus d_{Cu} was the same as ours. Therefore, our observation for the peak in H_C suggests that there is a large increase in the perpendicular interface anisotropy at 1 ML of Ag coverage, and the anisotropy constant slightly decreases for $1.0\ \mathrm{ML} < d_{\mathrm{Ag}} < 1.5$ ML. Weber *et al.*[22] observed that the magnetic anisotropy and co-

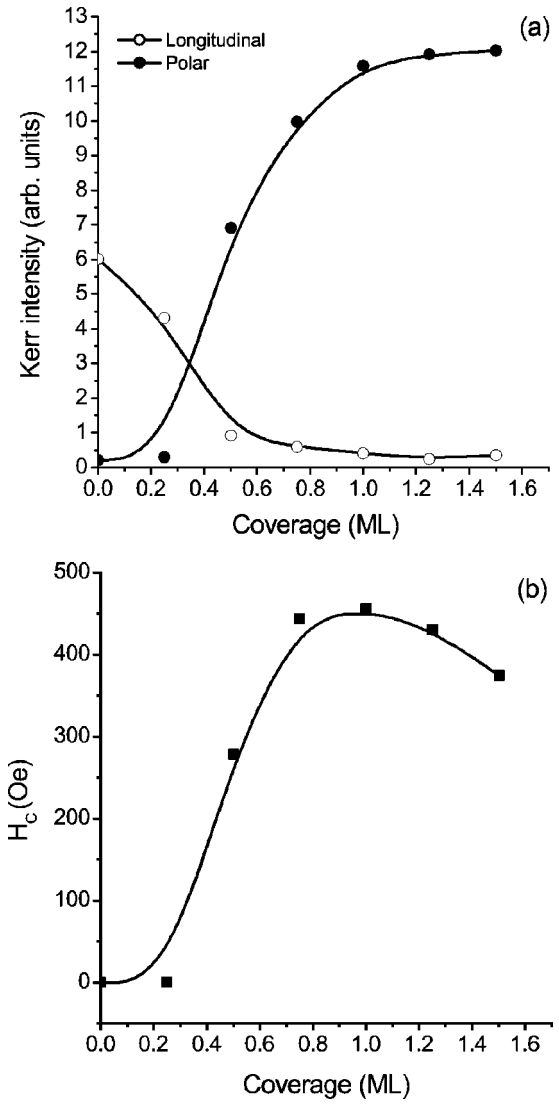

FIG. 3. (a) Longitudinal and polar Kerr intensities as a function of Ag coverage on 5-ML Co/Pt(111). The polar Kerr intensity increases quickly when $d_{\mathrm{Ag}}=0.5$ ML, and saturates when $d_{\mathrm{Ag}}=1$ ML. (b) Coercivity as a function of Ag coverage on 5-ML /Pt(111). The maximum of H_C is located at 1 ML of Ag coverage. The continuous lines along data points are just a guide to the eye.

ercivity in Co films epitaxially grown on Cu(100) osillate as a function of the Co thickness with a period of 1 ML. They proposed that these oscillations contribute to the periodic variation of the film morphology alternating between filled and incompletely filled atomic layers. The same behavior of H_C oscillation in a Fe/Pd(100) system was observed by Childress *et al.*[23] They also proposed a possible roughness modulation during growth of successive Pd layers. In the growth of Ag overlayers, as soon as the layer is completed in 1 ML, islands form in the next layer. The Ag film morphology changes from flat to rough when the coverage of Ag is larger than 1 ML. The film roughness of Ag causes a slight decrease in H_C as shown in Fig. 3(b). Another possibility is that it could arise from the hybridization of electronic states at the Ag/Co interface. The magnetic interface anisotropy is very sensitive to the details of the electronic band structure.[24]

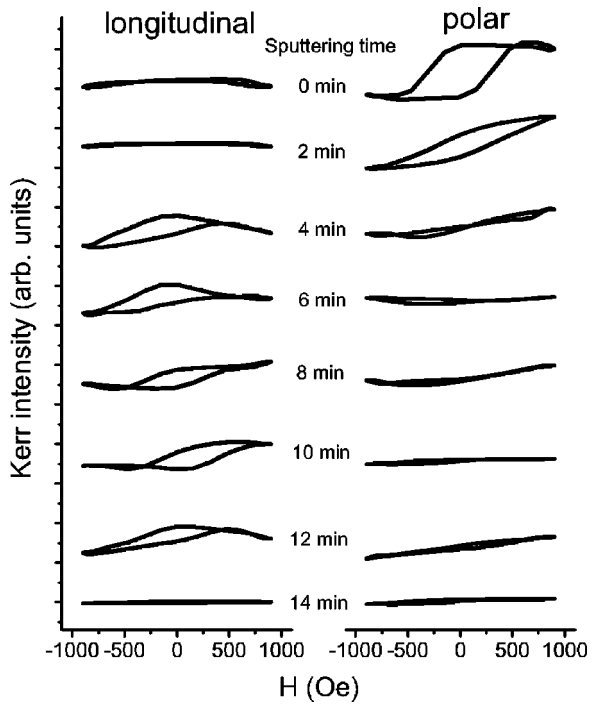

FIG. 4. The evolution of the longitudinal and polar hysteresis loops for 1-ML Ag/5-ML Co/Pt(111) during the Ar-ion sputtering. The longitudinal hysteresis loop appears gradually during the sputtering. The polar hysteresis loop disappears as Ag atoms are removed.

The spin-reorientation transition induced by Ag can occur for the Co/Pt(111) system which exists in both polar and longitudinal hysteresis loops initially [e.g., 4.5-ML Co/Pt(111) in Fig. 1]. The critical thickness of Ag overlayers reduces as the coverage of ferromagnetic Co thin films decreases. The spin-reorientation transition induced by Ag also exists for Co coverages of more than 5 ML. However only parts of Co spins can be reoriented if the coverage of Co is higher than 7 ML. In our observation, both the in-plane and the out-of-plane magnetization appear regardless of the thickness of Ag deposition when the coverage of Co is larger than 7 ML. We will interpret this phenomenon later.

Argon ions with a kinetic energy of 1.6 keV and a pressure of 1.0×10^{-5} Torr were used in the depth profile for 1-ML Ag/5-ML Co/Pt(111) at room temperature. The hysteresis loops were measured at every 2-min sputtering. The results are shown in Fig. 4. The longitudinal Kerr signal recovers the original value and the polar Kerr signal disappears gradually after the Ag overlayers are removed gradually. The in-plane magnetic hysteresis loop was observed after 4 min of sputtering. After further sputtering, parts of Co thin films are removed and the surface becomes rough. Therefore the shape of the longitudinal hysteresis loop is not as square as that of the original one. Auger electron spectroscopy was used to monitor the evolution of the compositions in the interfaces during the sputtering. The results of the depth profile are shown in Fig. 5. The Ag 351-eV Auger signal decreases very quickly because Ag overlayers are on the topmost layer. The Co 775-eV Auger signal slightly increases initially and then decreases slowly because a 5-ML Co thin

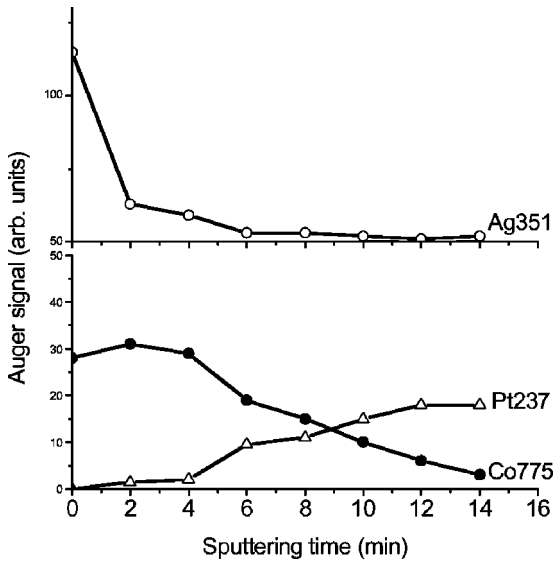

FIG. 5. Depth profiling for the study of the composition at the interfaces of 1-ML Ag/ 5-ML Co/Pt(111) during the sputtering in Fig. 4. The evolutions of Ag, Co, and Pt Auger signals indicate that the spin-reorientation transition is induced by Ag overlayers. The continuous lines along data points are just a guide to the eye.

film is located in the middle. The Pt 237-eV Auger signal increases monotonously. The sputtering in the initial 4 min had removed most of the Ag overlayers. The Co thin film still had some residuals from $t=6$ min to $t=12$ minutes in Fig. 5. This explains why we can observe the in-plane hysteresis loops during the sputtering as shown in Fig. 4. Co thin films were almost sputtered out at $t=14$ min. Therefore the longitudinal Kerr signal also disappeared in Fig. 4. The results of Auger electron spectroscopy during the sputtering are consistent with the evolution of the in-plane magnetic hysteresis loop appearing and the out-of-plane hysteresis loop disappearing as shown in Fig. 4.

It is interesting to know what the mechanism is for the spin-reorientation transition induced by Ag in this system. Recently Robach *et al.*[25] reported that adsorption of CO gas on the Co/Pt(111) surface causes a flipping of the easy direction of magnetization from parallel to perpendicular for Co films of about four atomic layers in thickness. Their interpretation of this unusual phenomenon is that CO reduces the magnetic moment of the surface Co atoms and leaving the inner atoms unaffected, and the remaining magnetic Co film has a thickness smaller than the critical thickness, and therefore it exhibits a perpendicular easy axis of magnetization. Since a spin-reorientation transition can occur at any coverage of Co lower than 7 ML, the mechanism in our system may be different. Beauvillain *et al.*[26] studied the enhancement of perpendicular magnetic anisotropy of Co/Au(111) after having capped Au, Cu, or Pd using the measurement of the surface magneto-optical Kerr effect, and showed that only the interface contribution to the anisotropy changes with the overlayer thickness. Following this conclusion, the change of the interface anisotropy must be involved for our calculation.

The effective anisotropy of the uncovered Ag film in our system can be written as[27]

$$K_{eff}=K_V+\frac{K_S^{\mathrm{Pt}}}{d_{\mathrm{Co}}}+\frac{K_S^{UHV}}{d_{\mathrm{Co}}}, \tag{1}$$

where K_V is the volume anisotropy, K_S^{Pt} is the interface magnetic anisotropy between Co and Pt, and K_S^{UHV} is the interface magnetic anisotropy between Co and the vacuum. We can assume that the volume anisotropy and the interface anisotropy between Co and the substrate Pt do not change after Ag is deposited on the Co/Pt(111) surface. When the fraction of Ag surface coverage is α, the effective anisotropy can be written as

$$K_{eff}=K_V+\frac{K_S^{\mathrm{Pt}}}{d_{\mathrm{Co}}}+\frac{K_S^{UHV}}{d_{\mathrm{Co}}}(1-\alpha)+\frac{K_S^{\mathrm{Ag}}}{d_{\mathrm{Co}}}\alpha, \tag{2}$$

where K_S^{Ag} is the interface magnetic anisotropy between Co and Ag. The fraction $\alpha=d_{\mathrm{Ag}}/d_{\mathrm{Ag(1\ ML)}}$ with $0\leqslant\alpha\leqslant1$. We take $\alpha=1$ when the coverage of Ag is larger than 1 ML. If we neglect high-order terms, the anisotropy energy density can be written as $E=K_{eff}\sin^2\theta$, where θ is the angle between the magnetization and the surface normal. The direction of the easy axis of the magnetization can be determined from a minimum requirement of E. The volume anisotropy, which mainly includes the crystalline anisotropy and the shape anisotropy, favors the in-plane magnetization for the Co thin film on Pt. The interface magnetic anisotropy between Co and Pt, and between Co and Ag, favor the out-of-plane magnetization. From the reported values, K_V is negative (-6.8×10^6 erg/cm^3),[28] K_S^{Pt} is positive ($K_S^{\mathrm{Pt}}=0.57$ erg/cm^2),[29] K_S^{UHV} is negative ($K_S^{UHV}=-0.17$ erg/cm^2),[26,27] and K_S^{Ag} is positive ($K_S^{\mathrm{Ag}}=0.35$ erg/cm^2).[30] Before Ag deposition, $K_{eff}>0$ in the initial growth of Co because the $K_S^{\mathrm{Pt}}/d_{\mathrm{Co}}$ term is dominant in Eq. (1) and d_{Co} is very small. Therefore the easy axis of the magnetization is perpendicular to the surface in the Co initial growth. $K_{eff}<0$ after further deposition of Co because d_{Co} becomes larger. Therefore the easy axis of the magnetization is in the plane when the coverage of Co is thick enough.

After Ag deposition on 5-ML Co/Pt(111), K_{eff} changes sign from negative to positive because $K_S^{\mathrm{Pt}}/d_{\mathrm{Co}}$ and $K_S^{\mathrm{Ag}}/d_{\mathrm{Co}}$ in Eq. (2) are positive. Therefore the easy axis changes from the in-plane direction to the out-of-plane one. One can obtain the same conclusion from comparing Eq. (2) and Eq. (1). All terms are identical except that K_S^{Ag} is used instead of K_S^{UHV} after Ag deposition. Since K_S^{Ag} is positive and K_S^{UHV} is negative, K_{eff} in Eq. (2) becomes positive after the proper thickness of Ag deposition. Therefore the easy axis of the magnetization rotates 90° from the in-plane to the out-of-plane direction.

Further calculation in Eq. (2) reveals that K_{eff} becomes negative when d_{Co} is larger than 6.6 ML. This indicates that the magnetization favors the in-plane direction when the thickness of Co is larger than 6.6 ML. This explains why only parts of the Co spins reorient in our observation regardless of the thickness of Ag being deposited when the coverage of Co is larger than 7 ML.

Although the results of the numerical calculation of the change in the interface anisotropy are consistent with our observations, the physical reasons need further discussion. The lattice mismatch between Ag and Co is very large. The strain causes the interface anisotropy to change sign from negative in K_S^{UHV} to positive in K_S^{Ag}. Another possible physical origin is that the hybridization of electronic states at the Ag/Co interface could cause significant alternations of the magnetic interface anisotropy.[24,26] Indeed, the electron structures of Cu ultrathin films on Co(0001) have some important changes that can be observed by photoemission.[31] The hybridization of the Ag and Co wave functions changes the magnetic anisotropy. Both the strain due to the large lattice mismatch and the hybridization of electron states at the Ag/Co interface are the possible mechanisms of spin-reorientation transition in our system.

IV. CONCLUSION

The easy axis of the magnetization is the in-plane direction when the coverage of the Co thin film on the Pt(111) surface is larger than 5 ML. After 1 ML of Ag overlayers is deposited on 5 ML Co/Pt(111) surface, the easy axis of the magnetization reorients from the in-plane to the out-of-plane direction. The increasing thickness of the Ag overlayers does not further induce the spin-reorientation transition after all the Co sites are completely occupied by Ag atoms. The easy axis of the magnetization transits back to the in plane if Ag overlayers are sputtered out. The critical thickness of the spin-reorientation transition for Ag overlayers reduces when the coverage of the Co thin film decreases. The thickness between Ag and Co is the main contribution for the spin-reorientation transition. Both the strain due to large lattice mismatch and the hybridization of electron states at the Ag/Co interface causing the change in interface magnetic anisotropy are the possible mechanisms of the spin-reorientation transition induced by Ag.

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