

# Ultrathin Co films on Pt(111) and the Co-Pt interface investigated by surface magneto-optical Kerr effect and medium-energy ion scattering spectroscopy

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Magnetic anisotropy evolution of ultrathin Co films grown on a Pt(111) single-crystal surface is investigated by a UHV *in situ* surface magneto-optical Kerr effect (SMOKE) measurement. Perpendicular magnetic anisotropy in ultrathin Co films is found to be persistent up to a thickness of 12 ML where the easy magnetization axis changes its orientation to an in-plane direction. The interface structure of an ultrathin Co overlayer on Pt(111) was investigated with atomic-layer resolution medium-energy ion scattering spectroscopy. For 7-ML Co, interdiffusion begins at 673 K to form a heavily distorted Co-Pt surface alloy layer with little change in SMOKE intensity. However, annealing at 773 K formed a 30-atomic-layers thick Co-Pt substitutional alloy with tensile strain, at which the SMOKE intensity increased by more than 200%. The enhancement of the Kerr intensity is discussed with the interface alloy formation.

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Perpendicular magnetic anisotropy (PMA) in ultrathin magnetic films, multilayers, sandwiches, and alloyed thin films has attracted strong interest due to not only fundamental curiosity but also its applications to perpendicular magnetic and magneto-optical recording media. Co/Pt is one of the most investigated systems exhibiting PMA in addition to a large magneto-optical (MO) Kerr effect required for applications to high-density MO recording media.<sup>1-7</sup> It is known that a Co layer on Pt exhibits PMA at the ultrathin limit. For example, it was predicted theoretically that one Co monolayer (ML) on Pt(111) should show PMA.<sup>8</sup> Indeed, perpendicular domains were observed for a 4-ML-thick Co film on a Pt surface.<sup>9</sup> As the thickness increases, the orientation of magnetization changes from perpendicular to in-plane orientation. This so-called spin reorientation transition (SRT) has also attracted much fundamental interest.<sup>1,10</sup> While a number of experimental investigations have been performed on artificially structured Co-Pt multilayers, sandwiches, and alloys, to our best knowledge, surprisingly few results are available on the magnetic properties of Co ultrathin films on the Pt single-crystal surface. Surface magneto-optical Kerr effect (SMOKE) measurement can provide detailed information on the evolution of magnetic anisotropy such as SRT when measured *in situ* in ultrahigh vacuum without complications arising from the necessary protective overlayers for *ex situ* measurements.<sup>11</sup>

Magnetic anisotropy is also very sensitive to local order and broken symmetry at interfaces. A number of studies have been reported on the effects of intermixing, interface alloy formation, Pt polarization for Co-Pt multilayers, thin films, and novel alloys. However, to our best knowledge, detailed atomic scale characterization of the Co-Pt interface is still lacking, since most conventional surface and interface analysis tools have limited depth resolution. Medium-energy ion scattering spectroscopy (MEIS) has been successfully applied to study various important interfaces with atomic-layer

depth resolution to investigate the composition and structural profiles near the interfaces.<sup>12-14</sup>

In this paper, we report our investigation on the magnetic properties of a Co ultrathin film on a Pt(111) single crystal surface by UHV *in situ* SMOKE measurement combined with structural study by MEIS. The interface structure of a 7-ML Co overlayer on Pt(111) upon annealing at elevated temperatures is studied as well, showing Co interdiffusion into Pt and surface alloy formation along with dramatically enhanced SMOKE response.

The experiment was carried out in a custom-designed ultrahigh-vacuum chamber with a base pressure of  $7 \times 10^{-11}$  Torr. The main chamber is equipped with a three-axis SMOKE measurement setup at the deposition position enabling a detailed *in situ* measurement of the evolution of the magnetic properties without the need for translation of the sample. Details of the SMOKE setup have been published elsewhere.<sup>15</sup> We have measured Kerr ellipticity as well as Kerr rotation simultaneously from the first and second harmonics of a precision photoelastic modulator with frequency of 50 kHz. Kerr effects were measured at polar (longitudinal) configuration where the applied field is normal (parallel) to the film plane.

The MEIS experiment was done in a separate UHV chamber with 100-keV  $H^+$  ions incident along the  $[110]$  direction and exiting along the  $[00\bar{1}]$  direction with a scattering angle of  $90^\circ$ . From the precise energy-loss measurement of 100-keV  $H^+$  ions, the energy spectra shows the compositional depth profile with the layer-by-layer depth resolution. The angular distribution of scattered  $H^+$  ions is mainly determined by the position of neighbor atoms. With the incident  $H^+$  ions highly collimated, the angular divergence is better than  $0.01^\circ$ . The minimum in the angular distribution of scattered ion corresponds to the local bond angle. For a blocking dip analysis, the incident  $H^+$  ions are  $4^\circ$  off from the  $[110]$

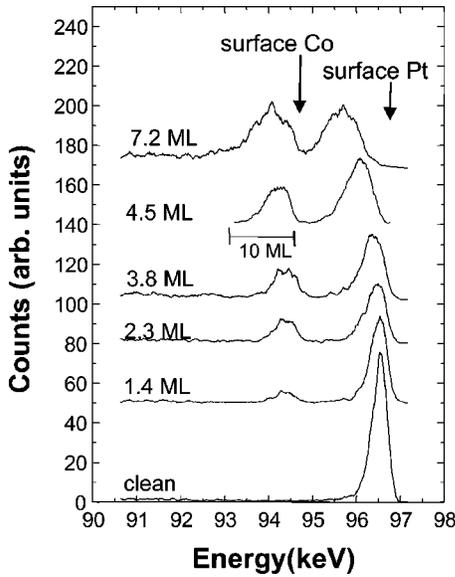


FIG. 1. MEIS energy spectra of a Co overlayer on Pt(111) for various Co coverages.

direction and the angular distribution around the  $[00\bar{1}]$  is measured with a two-dimensional multichannel plate detector. From the shift of the blocking dip, the strain, that is, the local bond angle, can be measured as a function of the depth. Details of the MEIS technique and the system used in this experiment are described elsewhere.<sup>13,14</sup> Special attention was paid to the sample treatment and the Co deposition to ensure identical conditions for the MEIS and SMOKE experiments. A Pt(111) single-crystal substrate was cleaned with cycles of 1-keV  $\text{Ar}^+$  sputtering followed by annealing up to 1000 K. Co was deposited from an electron-beam heated rod source at a rate of 0.4 ML/min. Deposition rates were controlled with the built-in flux monitor. The thickness of the Co overlayer was calibrated with a UHV scanning tunneling microscope and a monolayer in this study corresponds to the number of atoms necessary to completely cover the surface at the density of Pt(111) atoms. The pressure remained at better than  $1.0 \times 10^{-10}$  Torr during the deposition. All deposition and measurements were performed at room temperature.

MEIS energy spectra of Co overlayers for selected Co coverages are shown in Fig. 1. The Co coverage for thick overlayers was estimated based on the electronic stopping power, while the surface Pt peak was used for estimating the coverage in the initial stage around the monolayer since the width of the Co peak is similar to the system resolution. Upon the deposition of about a 1.4-ML Co layer, the surface Pt peak decreased by 50%, which implies that Co atoms do not block the surface Pt atoms completely. It suggests that the initial Co layers do not grow with the lateral spacing of the Pt substrate. The Pt surface peak persists up to 4-ML Co deposition, implying that a significant fraction of the original Pt surface is still exposed. At higher than 4-ML coverages, the Pt surface peak disappears as Co forms a continuous overlayer. Observed broadening of the surface Pt peak could be due to the nonuniform Co overlayer thickness. Our results are consistent with the conclusions from low-energy

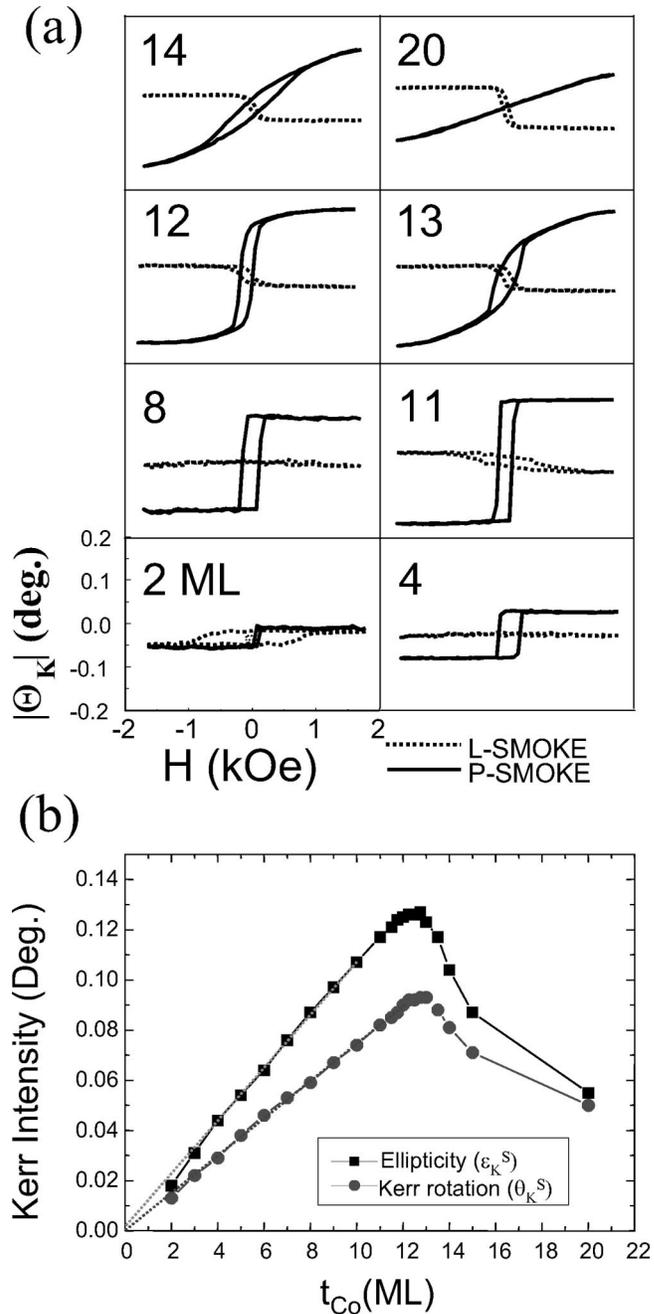


FIG. 2. (a) Polar Kerr hysteresis loops at selected Co thicknesses; (b) saturation Kerr ellipticity ( $\epsilon_K^S$ ) and Kerr rotation ( $\theta_K^S$ ) vs Co thickness.

electron-diffraction studies<sup>5,16</sup> and surface x-ray-diffraction studies<sup>6</sup> where Co is found to grow from the very start with its own lateral spacing and not in pseudomorphism with the substrate, the so-called incoherent epitaxy. Scanning tunneling microscopy studies revealed that Co grows into a multi-leveled surface after  $\sim 4$  ML, in agreement with our interpretation on the broadening of the surface Pt peak.<sup>17,18</sup>

Figure 2(a) shows the Kerr hysteresis loops at selected Co overlayer thicknesses. Here  $|\Theta_K|$  denotes the magnitude of the complex Kerr effect, given by  $|\Theta_K| = (\epsilon_K^2 + \theta_K^2)^{1/2}$ , where  $\theta_K$  and  $\epsilon_K$  are the Kerr rotation and Kerr ellipticity, respec-

tively. A ferromagnetic hysteresis loop appears at 2 ML with PMA, which persists up to 11 ML where a reorientation transition to in-plane anisotropy occurs. Both polar as well as longitudinal hysteresis loops are visible at 2 ML of Co coverage initially, indicating that both perpendicular as well as in-plane magnetization can be energetically stable configurations. The fact that the coercivity for the longitudinal loop is much larger indicates that the energy barrier for domain reversal in the film plane is much higher than that of the other case. This coexistence of perpendicular and in-plane phases has been predicted theoretically<sup>19</sup> and observed experimentally in Co/Pd(111) systems as well.<sup>20</sup> Above 3 ML, only polar hysteresis loops with squareness of 1 occur up to 11 ML; we conjecture that longitudinal hysteresis loops are simply not measurable with the available applied field because of high coercivity for this thickness range.

Thickness dependencies of both the saturation Kerr ellipticity ( $\epsilon_K^S$ ) and Kerr rotation ( $\theta_K^S$ ) in the polar hysteresis loops are shown in Fig. 2(b). As is obvious in the graph, both  $\epsilon_K^S$  and  $\theta_K^S$  increase linearly with Co overlayer thickness up to 11 ML. These curves have no appreciable offsets when extrapolated to  $t_{\text{Co}}=0$ . This contrasts with earlier reports by McGee *et al.*<sup>5</sup> on the Co sandwich structure, where significant offsets both in  $\epsilon_K^S$  and  $\theta_K^S$  were observed and attributed to the polarization of nonmagnetic Pt by neighboring Co. The difference can be partially attributed to the fact that there is no additional Co-Pt interface due to the protective Pt overlayer, in our case, compared to the sandwich sample. Even so, our result suggests that Pt polarization is negligible at the interface.

As the Co film thickness increases, the easy axis of magnetization changes from perpendicular to in plane as clearly seen in Fig. 3(a), where the ratio of the remanence Kerr signal ( $\theta_K^R$ ) at  $H=0$  to the saturation Kerr ( $\theta_K^S$ ) is plotted as a function of  $t_{\text{Co}}$ . This spin reorientation transition (SRT) occurs when the shape anisotropy, caused by dipolar interaction, overcomes the surface anisotropy. For a closer look at the transition, we have plotted the angle of the magnetization vector from the surface normal,  $\theta = \cos^{-1}(\theta_K^R/\theta_K^S)$ , assuming a coherent rotation of the easy magnetization direction during SRT, Fig. 3(b). It is immediately clear that the transition shows the characteristics of a smooth second-order transition. With the crossover thickness from perpendicular to in plane to be where  $\theta=45^\circ$ , as in the study of Co on Au(111),<sup>21</sup> our critical thickness is estimated to be  $\sim 12$  ML.

The observed transition thickness of  $\sim 12$  ML for SRT is, to our knowledge, the thickest yet reported for single Co layers grown on a Pt(111) surface. For example, Shern *et al.*<sup>22</sup> reported that the reorientation transition occurred at a coverage of only 4.4 ML in a similar *in situ* SMOKE experiment. From magnetic circular x-ray dichroism of Co on Pt(111), Thiele *et al.*<sup>23</sup> claimed a critical thickness of  $\sim 7$  ML for PMA. Even in the case of *ex situ* measurement on the sandwich structure, McGee *et al.*<sup>5</sup> found PMA up to nine Co layers. Furthermore, in a recent theoretical study, Pustogowa *et al.* predicted a reorientation transition at about 4 ML for Co grown on the Pt(111) surface.<sup>7</sup> Many possibilities exist for this discrepancy among the experiments and

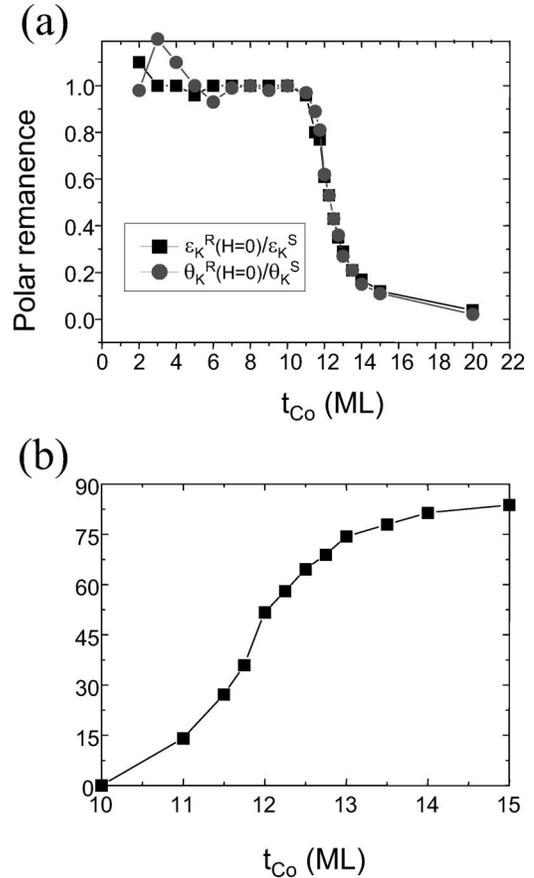


FIG. 3. (a) Plot of polar Kerr remanance at  $H=0$  showing the spin reorientation transition; (b) rotation angle  $\theta = \cos^{-1}(\theta_K^R/\theta_K^S)$  at the SRT thickness.  $\theta=45^\circ$  corresponds to  $\sim 12$  ML.

theoretical predictions, such as substrate conditions, deposition methods and rate, and contamination of the film. However, we believe this discrepancy most likely originates from differences in the initial substrate morphology. Indeed, we have found some variations in the critical thickness in our series of measurements as well, where SRT at as early as 7-ML coverage was observed in a separate measurement even when all other conditions were kept constant. In a related experiment on a Co/Pd(111) system, we have verified by scanning tunneling microscopy combined with SMOKE that substrate roughness affects the critical thickness for the reorientation transition: a rough substrate triggers earlier transition.<sup>24</sup>

For the investigation of the interface structure and its effect on the magnetic properties, we chose a 7-ML Co overlayer on Pt(111) and performed an annealing study. Figure 4 illustrates the changes in the MEIS energy spectra of 7-ML Co on Pt(111) as a function of annealing temperature. The Co and Pt peak shapes changed only slightly after annealing at 573 K for 3 min, which might be due to morphological changes of the film. The peak shapes suggest that the Co overlayer does not interdiffuse at this temperature. Annealing at 673 K for 5 min made the surface Pt peak reappear with a broad width corresponding to  $\sim$  ten atomic layers, while at the same time the Co peak decreased to a smaller peak. At this temperature, no blocking dip was observed, which im-

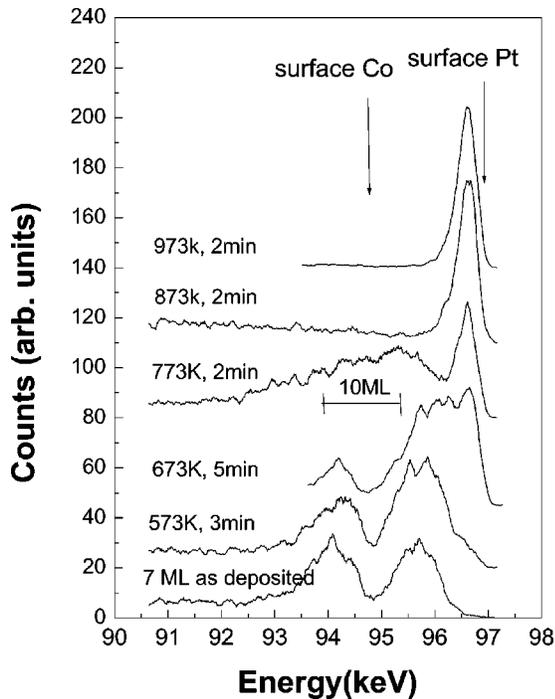


FIG. 4. The change of the energy spectra of 7-ML Co on Pt(111) as a function of annealing temperature.

plies that interdiffusion takes place and forms a heavily distorted or almost amorphous surface alloy.

The surface Pt peak became clean and sharp after annealing at 773 K for 2 min. The broad peak in the energy spectra reflects the distribution of Co atoms diffused into the Pt substrate, which extends over 30 atomic layers with a peak around nine atomic layers below the surface. The disappearance of the Co peak with diffusion suggests that the diffused Co atoms occupy substitutional sites in the Pt lattice. Our analysis of the blocking dips at this annealing temperature showed that the Pt atoms between 8 and 11 layers below the surface are under tensile strain.<sup>25</sup> A clear dip shift of  $1.0^\circ$  to the lower scattering angle was observed, reflecting a 3.7% vertical contraction of the (111) interlayer distance due to the diffused substitutional Co atoms. Assuming a uniform distribution of 7-ML Co over 30 Pt atomic layers, the Co concentration could be estimated to be approximately 25%. Considering the 10% smaller covalent radius of Co compared with Pt, a maximum of 3.7% vertical contraction of the Pt lattice with 25% Co surface alloying would be reasonable.

Further annealing at 873 K enhanced the Pt surface peak and the broad peak became a background that increases with depth. The increased background suggests that the Co atom distribution and associated lattice distortion are quite extended, estimated to be tens of atomic layers. Annealing even further up to 973 K for 2 min regenerates the clean Pt MEIS energy spectra as shown in Fig. 1.

The SMOKE measurements were performed for the 7-ML Co overlayer under the annealing conditions identical to the case of MEIS measurements. The results are summarized in Fig. 5. Initially the as-grown 7-ML Co exhibits a slightly canted orientation of magnetization as indicated by the presence of a longitudinal hysteresis loop with a less than perfect

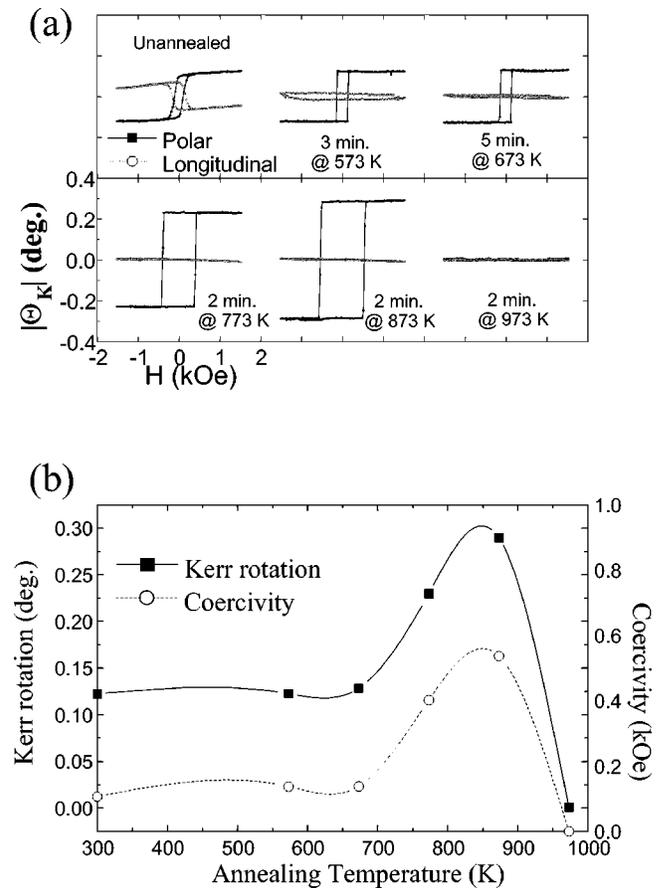


FIG. 5. (a) Kerr hysteresis loops of 7-ML Co on Pt(111), before and after annealing at different temperatures; (b) Kerr intensity and coercivity of 7-ML Co on Pt(111) as a function of annealing temperature.

polar loop. However, the polar hysteresis loop became square upon annealing at 573 K for 3 min, indicating that morphology change in the Co film promotes the PMA. The longitudinal hysteresis loop shows a minor loop indicating that the coercivity is larger than our available field. Further annealing at 673 K for 5 min enhanced the polar Kerr intensity slightly, with no trace of the longitudinal hysteresis loop. After 2-min annealing at 773 K, a dramatic increase in both the saturated Kerr rotation and the coercivity is observed. The Kerr rotation and the coercivity continued to increase after 2-min annealing at 873 K. The maximum enhancement in the Kerr signal is  $\sim 230\%$  at 873 K. The ferromagnetic signal disappeared completely after annealing at 973 K for 2 min. These observations indicate that heavily distorted surface alloying with high Co concentration, resulting from annealing up to 673 K, does not contribute significantly to the enhancement of the SMOKE signal. However, the Kerr signal increases dramatically as soon as Co atoms form a surface alloy with Pt crystalline lattice atoms by occupying the substitutional sites. The Kerr signal continues to increase as Co atoms form a dilute alloy with a concentration as low as probably a few percent, as estimated by the MEIS data.

While it is known that interface roughness and interdiffusion tend to decrease the strength of the PMA,<sup>26</sup> our results suggest otherwise. In a recent SMOKE experiment on Co/

Pt(111), Shern *et al.* found that the Kerr signal increases up to 200% for 1 ML of a Co overlayer after annealing at 710 K.<sup>22</sup> Similar enhancements were reported in Kerr spectroscopy measurements on a Co/Pt trilayer.<sup>27</sup> However, the origin of the enhanced Kerr signal is still not clearly understood. Ferrer *et al.*<sup>6</sup> determined that the transformation from an fcc to hcp structure upon annealing results in  $\sim 25\%$  enhancement of the magnetization at the interface in a resonant magnetic surface x-ray-diffraction experiment of Co on Pt(111). But the magnetic-moment increase alone cannot account for the large enhancement in the polar Kerr rotation by as much as 200%. A formation of an ordered Co-Pt alloy has been suggested by Train *et al.*<sup>27</sup> to explain the enhancement. Since a substantial electronic band-structure change is involved with chemical ordering in the Co-Pt alloy,<sup>4</sup> it may be responsible for the dramatic increase of the Kerr rotation. To verify this point, we performed magneto-optical Kerr spectroscopy (MOKE) on the annealed sample and compared it with the spectrum of the unannealed sample. For the *ex situ* measurement, the unannealed sample was covered with 1 nm of Pd to protect from oxidation. Indeed, as demonstrated in Fig. 6, our MOKE studies in the photon energy of 1.5–3 eV reveal very contrasting spectra between the unannealed sample and the annealed one at 873 K, and the Kerr rotation of the annealed sample was distinctly enhanced compared to the unannealed one. The spectra clearly show that there is a substantial difference in the electronic structure between the two samples. The spectrum of the annealed sample also exhibits a strong similarity to the spectrum of the ordered Co-Pt alloy, confirming this assertion.

In summary, we have investigated the magnetic and structural properties of Co films on a Pt(111) surface and Co-Pt

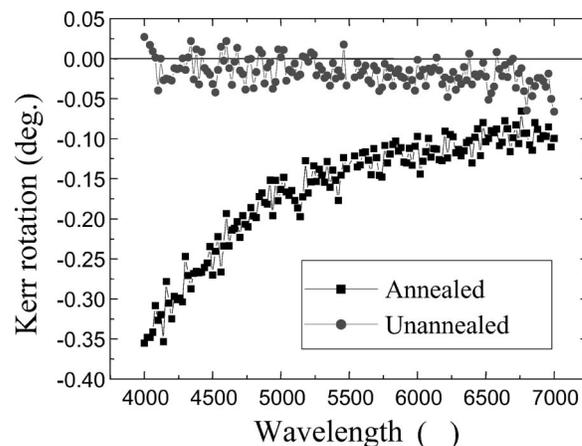


FIG. 6. Kerr spectra obtained on unannealed 7-ML Co and annealed 7-ML Co at 873 K for 2 min.

interface using SMOKE and MEIS. Ultrathin films of Co exhibit PMA up to 12 ML, contrary to other studies, before going through a smooth second-order spin reorientation transition to in-plane anisotropy. We also found that, after annealing 7-ML Co on Pt(111), an interface alloy under tensile strain was formed. This alloy formation was accompanied by a dramatic enhancement in the polar SMOKE signal. Our study shows that interface control is very important for the applications of Co-Pt systems to magnetic and magneto-optical recording media.

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