Spin-reorientation transitions in ultrathin Co films on Pt(111) and Pd(111) single-crystal substrates

Jeong-Won Lee,* Jong-Ryul Jeong, and Sung-Chul Shin
Department of Physics and Center for Nanospinics of Spintronic Materials, Korea Advanced Institute of Science and Technology, Daejeon 305-701, Korea

Jonggeol Kim
LG Chem Research Park Ltd., Daejeon 305-380, Korea

Sang-Koog Kim
Nanospintronics Laboratory, School of Materials Science and Engineering, College of Engineering, Seoul National University, Seoul 151-744, Korea

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Spin-reorientation transition (SRT) behaviors in ultrathin Co films grown on both Pt(111) and Pd(111) single-crystal substrates are investigated with *in situ* magneto-optical Kerr effects. At the region of SRT from the perpendicular to in-plane magnetization with the increasing of Co thickness, canted out-of-plane orientations are observed, from which we can determine not only the effective second- (K_2) but also fourth- (K_4) order anisotropy constants. Both the constants, which can be separated into bulk $K_{2b(4b)}$ and interface anisotropy terms $K_{2s(4s)}$ are essential to understand the stable canted out-of-plane magnetization. We found that the value of K_{2s} for Co/Pt(111) is larger than that for Co/Pd (111), resulting in the later onset thickness and wider range of the observed SRT, while the negative small values of K_{4s} in the both systems yield the stable canted phases. In addition, we observed different polarization effects of Pd and Pt near the interfaces which might also contribute the contrasting SRT behaviors.

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Ultrathin magnetic films have the property that the magnetic anisotropy prefers out-of-plane or in-plane magnetization depending on the film thickness or temperature. Perpendicular magnetic anisotropy (PMA) in ultrathin magnetic films has been one of the most attractive subjects due to its application to ultrahigh-density information storage. It is well known that Co films on Pt(111) and Pd(111) substrates exhibit PMA at the monolayers (ML) regime, where the magnetocrystalline anisotropy caused by a broken symmetry at the interfaces is sufficient to overcome the demagnetizing energy originated from the shape anisotropy.[1–4] As the film thickness increases, the magnetic easy axis changes from perpendicular toward in-plane orientation, which is a so called thickness-driven spin-reorientation transition (SRT). In the present work, we compare SRT behaviors in Co films grown on between Pt(111) and Pd(111) single-crystal substrates, especially under the context of their second- (K_2) and fourth- (K_4) order anisotropy constant flows.[8] Observed difference in the transition thickness between the two substrates is explained by differences in their interface anisotropy terms and likely by the polarization effects of Pt and Pd at the interfaces as well.

The Co films were grown on Pt(111) and Pd(111) single-crystal substrates at a rate of 0.4 ML/min by *e*-beam evaporation. The substrates were cleaned by a few cycles of 1 keV Ar^+ ion sputtering and annealing up to 1000 K. Well-defined terrace structures of the Pd(111) and Pt(111) surfaces were confirmed by a reflection high-energy electron diffraction (RHEED) and a scanning tunneling microscope (STM). The magneto-optical Kerr effect (MOKE) measurements as well as the Co growth were performed in an ultrahigh vacuum (UHV) chamber maintained under a base pressure of 1 $\times 10^{-10}$ Torr. The *in situ* MOKE measurements were carried out at the same position in the growth chamber without any sample transportation, which made the experiment convenient in the sample growth and measurement cycle with a step of 0.5-ML Co thickness. HeNe laser with a wavelength of 632.8 nm and silicon detectors were used at an incident angle of 45°. Using a lock-in technique with a precise photoelastic modulator of 50 kHz and crystal polarizers with extinction ratio of better than 10^{-5}, we simultaneously measured the Kerr rotation as well as ellipticity with an accuracy down to $\sim 0.001°$. Details of the measurement system were described elsewhere.[5]

We have *in situ* measured the polar and longitudinal MOKE signals after every Co deposition of 0.5 ML coverage. Figure 1(a) and 1(b) show the evolution of the polar Kerr hysteresis loops of Co films grown on Pt(111) and Pd(111) substrates. For both substrates, the square loops start

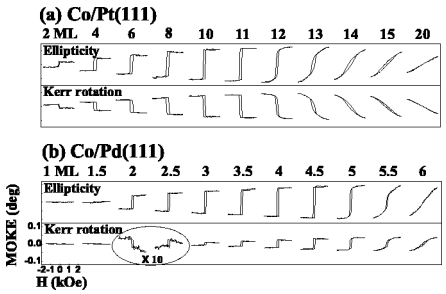

FIG. 1. Evolution of the polar Kerr hysteresis loops with Co thickness on (a) Pt(111) and (b) Pd(111) substrates. For 2 and 2.5 ML Co films on Pd(111), the Kerr rotation loops are magnified to clearly see the polarity change.

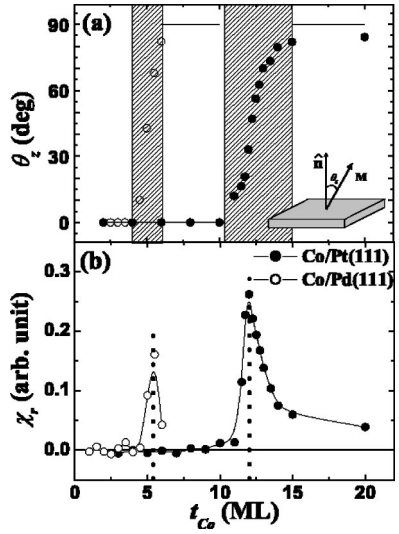

FIG. 2. (a) The canted angle θ_z of the magnetization vector from the film normal as a function of Co film thickness t_{Co}. The spin reorientation transition occurs in the shaded region. (b) Polar Kerr susceptibility at the remanent state (χ_r) as a function of t_{Co}. The dotted line indicates the critical thickness t_{Co}^c.

to appear at 2 ML coverage. Then, the loops become slanted from 10 ML for Pt(111) and 4.5 ML for Pd (111). The evolution of the hysteresis loops vs the Co thickness t_{Co} clearly exhibits that perpendicular magnetization switches to in-plane orientation as t_{Co} increases through the transition region of spin reorientation. Overall trends in the SRT behavior vs t_{Co} are similar for both substrates, but the onset thicknesses from perpendicular to in-plane orientation and the thickness ranges of transition are different.

In order to compare different SRT behaviors in both substrates, we first plot the canted angles θ_z from the film normal at the remanent state, i.e., at zero field as a function of t_{Co}, as shown in Fig. 2(a). The canted angles are determined from $\cos^{-1}[m_z/(m_x^2+m_y^2+m_z^2)^{1/2}]$, where m_x, m_y, and m_z are the magnetization components along each axis. The vectorial determination of all the magnetization components from MOKE signals was reported elsewhere.[6] In the plots of θ_z vs t_{Co} for Co/Pd (111) and Co/Pt (111), different transition onsets and ranges are evident, and also supported by the polar Kerr susceptibility, χ_r, defined by $dm_z/dH|_{H=0}$ as seen in Fig. 2(b).[13] The critical thickness t_{Co}^c, where χ_r has a maximum, are determined to be $\sim$ 5.5 ML Co on Pd(111) and $\sim$12 ML Co on Pt(111). Before starting SRT, χ_r remains almost zero due to the square shapes of the hysteresis loops, while the rapid increases up to t_{Co}^c and then decreases in the transition region represent the distinct SRT behaviors.

In order to understand the underlying physics of the difference in SRTs on Pd(111) and Pt(111) single-crystal substrates, we determine K_2 and K_4, both of which are essential for the canted orientation existing in the SRT region. To determine K_2 and K_4 values as a function of t_{Co}, we use their correlation with θ_z. For a uniaxial anisotropy system in the second-order approximation the free energy density in the absence of an applied magnetic field is given by

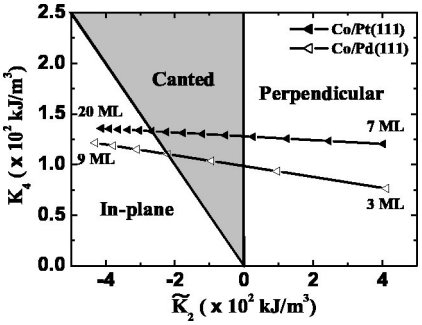

FIG. 3. Anisotropy constant flows on the $\tilde{K}_2$-K_4 plane with varying Co thickness. The perpendicular, canted, and in-plane phases are separated as noted in the figure.

$$E = K_2 \sin^2\theta + K_4 \sin^4\theta + \frac{1}{2}\mu_0 M^2 \cos^2\theta, \qquad (1)$$

where the first, second, and third terms are the second- and fourth-order anisotropy energies, and the demagnetization energy, respectively. By minimizing E with respect to θ, one can obtain the three solutions for the stable phases of magnetization orientations, i.e., $\theta = 0°$, 90°, and $\sin^2\theta = (\frac{1}{2}\mu_0 M^2 - K_2)/2K_4$. Those angles correspond to perpendicular, in-plane, and canted out-of-plane phases, respectively.[7–9] Since the interfaces between Co and Pd(111) or Pt(111) substrates are present in the whole system, K_2 and K_4 could be separated into the bulk and interface terms $K_2 = K_{2b} + K_{2s}/t_{\mathrm{Co}}$ and $K_4 = K_{4b} + K_{4s}/t_{\mathrm{Co}}$. From the Fig. 2(a), the onset (t_{Co}^o) and end (t_{Co}^e) thicknesses of transition are determined, which correspond to the cross points from perpendicular to canted phase and from canted to in-plane phase on the $\tilde{K}_2$-K_4 plane, respectively, where $\tilde{K}_2 = \frac{1}{2}\mu_0 M^2 - K_2$. One can determine the values of K_{2s} and K_{4s} from the two conditions with taking Co bulk values[10] for K_{2b} and K_{4b}: (i) $\tilde{K}_2 = 0$ at $t_{\mathrm{Co}} = t_{\mathrm{Co}}^o$ and (ii) $K_4 = -\frac{1}{2}\tilde{K}_2$ at $t_{\mathrm{Co}} = t_{\mathrm{Co}}^e$.[8] The numerical calculation results in $K_{2s} = 0.78$ mJ/m^2; $K_{4s} = -0.041$ mJ/m^2 for Co/Pd(111), and $K_{2s} = 1.8$ mJ/m^2; $K_{4s} = -0.034$ mJ/m^2 for Co/Pt(111). The large difference in the determined values of K_{2s} is evidently responsible for the contrasting onset thickness as well as transition range between both substrates.

The observed SRT behaviors in the Co/Pd(111) and Co/Pt(111) systems could be well explained under the context of the anisotropy flow. By adopting the calculated values, in Fig. 3 we plot the anisotropy flows on the $\tilde{K}_2 - K_4$ plane for the Co/Pd(111) and Co/Pt(111) systems with varying $t_{\mathrm{Co}} = 3$–9 ML and 7–20 ML, respectively. It is clear that the larger value of K_{2s} in the Co/Pt(111) than that in the Co/Pd (111) gives rise to the later onset and wider range of the SRT, while the negative small values of K_{4s} in the both systems yield a stable canted phase during the SRT's. As seen in Fig. 3, the SRT's in both the substrates proceed via a stable canted phase, i.e., a typical second-order (smooth) SRT as t_{Co} increases. However, there is a large difference in the onset thicknesses of the transition between the two substrates. The SRT from perpendicular to in-plane orientation starts at 4.5 ML Co for Pd(111), and proceeds through the thickness

range of 1.5 ML. On the other hand, the transition on Pt(111) substrate occurs in the range of $t_{Co}=10$-15 ML's. Our results vividly witness that the contrasting behavior is essentially originated from the different interface anisotropies sensitively dependent on the substrate material. For instance, in the Co/Au(111) system with $K_{2s}=0.66$ mJ/m^2 and $K_{4s}=-0.12$ mJ/m^2, Oepen *et al.*[11] reported an onset thickness of 3.7 ML and the transition via a coexistence phase within a 0.4 ML thickness.

In the thin film regime, the Kerr signal depends linearly on the thickness of the magnetic layer by the additivity law.[12] Our results also show the linear dependence of the Kerr signal on the Co thickness up to t_{Co}^c. In addition to this linear dependence, however, the Kerr signals for Co/Pd and Co/Pt show a contrasting behavior. Very interestingly, in Fig. 1(b) the negative polarity of the Kerr rotation loop at 2 ML Co on Pd(111) changes into the positive one at 2.5 ML Co, which might be caused by the polarization effect of Pd by Co at the interface. To our knowledge, we report, for the first time, an experimental evidence for the polarity change of the loops from the MOKE measurements. To convincingly verify the polarization effect, we plot the thickness dependence of the saturation values of the polar Kerr ellipticity ϵ^{sat} and rotation θ^{sat} as shown in Fig. 4. Both ϵ^{sat} and θ^{sat} increase linearly with t_{Co} up to t_{Co}^c as shown in Fig. 2(b). The ϵ^{sat} curves for both Pd(111) and Pt(111) substrates have no significant offset when extrapolated to $t_{Co}=0$ with the linear fits to the data prior to t_{Co}^c. The θ^{sat} curve for the case of Pd(111), however, has a large negative offset. Our results are different from the earlier reports, where significant offsets in both ϵ^{sat} and θ^{sat} were observed in the sandwiched Co films on Pt(111) (Ref. 14) and Pd(111) (Ref. 15) substrates. The discrepancy seems to be attributed to the fact that there is no additional Co-Pt (or Pd) interface in our *in situ* measurements compared to their wedged sandwich samples. Our results suggest that the polarization effect is more dominant in Pd than Pt, which is in accordance with theoretical predictions[16,17] and the experimental observation of charge transfer from Co to Pd at the interface.[18] Note that the polarity change of the Kerr rotation and the negative offset in Co/Pd(111) might imply a probable opposite direction of the polarized Pd moments with respect to Co moments. As evidenced by distinctively different onset thicknesses of SRT's in Co/Pd(111) and Co/Pt(111), the difference in the polarization effect between those systems may influence the interface anisotropy terms being essential for the SRT's in ultrathin Co films on both Pt(111) and Pd(111) substrates.

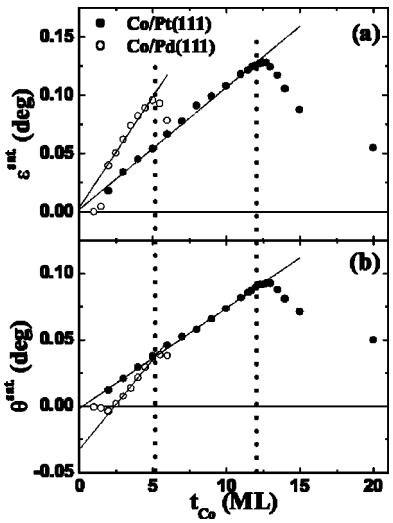

FIG. 4. (a) Polar Kerr ellipticity ϵ^{sat} and (b) rotation θ^{sat} values at the saturation field vs t_{Co}. The vertical dashed lines stand for the critical thickness t_{Co}^c. The solid lines are linear fits to the data.

In conclusion, we compared the spin-reorientation transition behaviors in ultrathin Co films grown on both Pt(111) and Pd(111) single-crystal substrates using *in situ* magneto-optical Kerr effects. With increasing Co thickness in the monolayer regime, the canted phases of magnetization were observed during the transition from perpendicular to in-plane magnetization on both substrates. We could determine the interface anisotropy terms $K_{2s(4s)}$ which are essential to understand the stable canted phase of magnetization. The larger value of K_{2s} for Co/Pt(111) in comparison with that of Co/Pd(111) results in the later onset thickness and wider range of the observed SRT from perpendicular to in-plane orientation, while the negative small values of K_{4s} in the both systems yield the stable canted phases. In addition, we observed different polarization effects of Pd and Pt near the interfaces which might also contribute the contrasting SRT behaviors.

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*Electronic address: phleejw@mail.kaist.ac.kr

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