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Magnetic Properties of Ultrathin Fe Films on Pt(111) and Pd(111): a Surface Magneto-Optic Kerr Effect Study

Wookje Kim and J. H. Choi

School of Physics, Seoul National University, Seoul 151-742

T.-U. Nahm* and S. H. Song

Quantum Photonic Science Research Center and Department of Physics, Hanyang University, Seoul 133-791

S.-J. Oh

School of Physics and Center for Strongly Correlated Material Research, Seoul National University, Seoul 151-742

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We have studied magnetic and structural properties of ultrathin Fe films grown on Pt(111) and Pd(111) substrates by using the *in situ* surface magneto-optic Kerr effect (SMOKE), X-ray photoelectron spectroscopy (XPS), and low-energy electron diffraction (LEED). SMOKE measurements show that perpendicular magnetic anisotropy is present for an as-deposited 4.0 ML (monolayer) Fe film on Pt(111) and partially induced by post-annealing at 450 K for a 2.0 ML Fe film on Pd(111) surface. For thicker films on Pd(111), a strong enhancement in the Kerr signal was observed upon post-annealing at temperatures above 600 K. Underlying reasons, such as morphological change and interdiffusion of Fe atoms into the Pd substrate, are discussed on the basis of the XPS and LEED results.

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I. INTRODUCTION

The observation of perpendicular magnetic anisotropy in Co/Pt multilayer films has initiated a number of investigations on magnetic films because of their potential applications as magnetic recording materials [1,2]. Since Fe is the most familiar ferromagnetic material, there have been many reports on Fe multilayered systems or alloys with nonmagnetic elements such as Au, Ag, Pd, and Pt [3, 4]. Fe/Ag(100) is one of the most studied systems that shows spin reorientation as a function of Fe film thickness [5] and post-annealing temperature [6], and it is expected that many ultrathin magnetic films should exhibit such behavior.

Ferromagnetic thin films on a Pt or Pd substrate enjoy some scientific interest since the magnetic moment of Pt or Pd atoms can be induced by ferromagnetic coupling with adjacent magnetic layers. Also, due to large spin-orbit coupling of Pt, the orbital magnetic moment of Fe can be enhanced by hybridization. There have been many reports on multilayered or alloy films of Fe with Pt or Pd [7–13], and it has been shown that perpendicular magnetic anisotropy (PMA) can be observed in several cases under certain circumstances. In order to understand the microscopic origin of the novel properties of these magnetic systems, it is necessary to study the role of the interface and intermixing between the ad-layer and the substrate.

In this work, we have investigated Fe films grown on Pt(111) and Pd(111) by using the surface magneto-optic Kerr effect (SMOKE), X-ray photoelectron spectroscopy (XPS), and low-energy electron diffraction (LEED). We found that the Fe films on Pt(111) have perpendicular magnetism near 4 ML, and in-plane easy axis above 5 ML. Post-annealing above 600 K induces the intermixing of Fe and Pt, leading to probably Fe_3Pt surface alloy formation, which reduces the magnetization. Perpendicular magnetic anisotropy is only partially induced by post-annealing at 450 K for a 2.0 ML Fe film on Pd(111) surface. For thicker films on Pd(111), an enhancement in the Kerr signal was observed upon post-annealing at temperatures above 600 K.

II. EXPERIMENT

The SMOKE, XPS, and LEED measurements were performed in a home-made ultrahigh vacuum chamber.

*E-mail: tschnahm@hanyang.ac.kr

The base pressure of the chamber was 8×10^{-11} torr. The Pt(111) and Pd(111) substrates were of disk type with diameter 10 mm and thickness 1 mm. The surface was cleaned by several cycles of Ar^+ sputtering at 1 keV and annealing at 870 K in the UHV chamber until the crystal surface showed a well-defined LEED pattern with no contamination detected in the XPS spectrum. The light source for SMOKE measurement was a 10-mW He-Ne laser with $\lambda = 623.8$ nm.

The XPS spectra shown here were obtained with unmonochromatized Mg K_α radiation, with $h\nu = 1253.6$ eV. The overall resolution was 0.9 eV. Since the O Auger spectrum excited by Mg K_α lines partly overlaps with the loss structure of the Fe $2p$ photoemission (PE) lines, we also used Al K_α radiation with $h\nu = 1486.6$ eV to check O contamination on the surface.

The Fe film was deposited on the substrate at room temperature by an e-beam heating method. An iron wire of 99.99 % purity was heated by electron bombardment from a tungsten filament. The film thickness was first calibrated by using a quartz thickness monitor, and was then cross-checked by XPS by using Fe $2p$ and Pt $4f$ or Pd $3d$ core-level PE line intensities at sub-ML coverage, assuming no island growth. The values obtained from these two methods agree with each other within an error of 10 %. In this work, the Fe film thickness was represented in units of one monolayer and 1 ML was set as 1.50 or 1.53 $\times$ 10^{15} Fe atoms cm^{-2} for Pt(111) and Pd(111) surfaces, assuming a pseudomorphic growth of the Fe film on the Pd(111) surface.

III. RESULTS AND DISCUSSION

1. Fe/Pt(111)

The results of SMOKE measurement for Fe films grown on Pt(111) surface are shown in Fig. 1. The 4.1 ML Fe film has perpendicular magnetization, and thicker films have longitudinal easy axis. No ferromagnetic behavior was observed in the 2.0 ML film, probably because the critical temperature was below room temperature. Although the remanence is almost 100 %, the hysteresis curve is not square-like. The post-annealing at 526 K makes the easy axis of the 4.1 ML film change its direction from perpendicular to in-plane. Post-annealing at 800 K leads to the disappearance of the magnetization.

The XPS spectra of Fe $2p$ from 5.1 ML Fe film shown in Fig. 2 can explain these behaviors. Upon post-annealing at 690 K, both Fe $2p$ and Pt $4f$ core levels shift, due to the change in chemical environment, and, for the same reason, the density of states at the Fermi level is reduced. These spectroscopic observations imply that there is some intermixing between ad-layer and substrate. In order to estimate the composition of the Fe concentration of surface alloy Fe_xPt_{1-x}, we use Johansson and Mårtensson's method for treating core-level

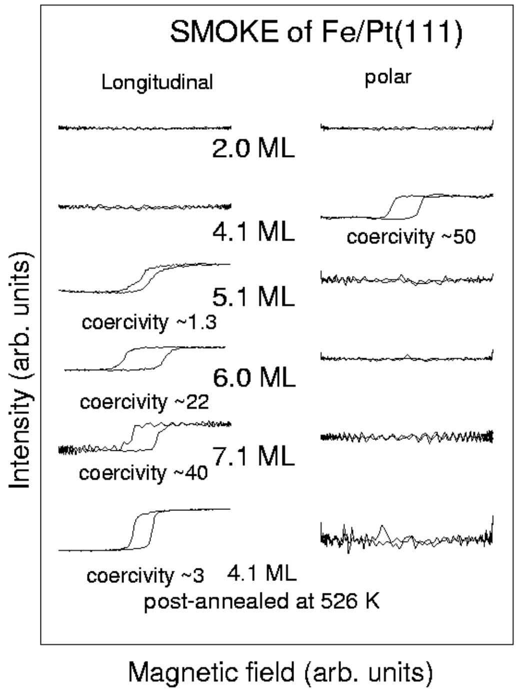

Fig. 1. SMOKE signal of various Fe films on Pt(111). Perpendicular magnetism is only observable for as-deposited 4.1 ML Fe film.

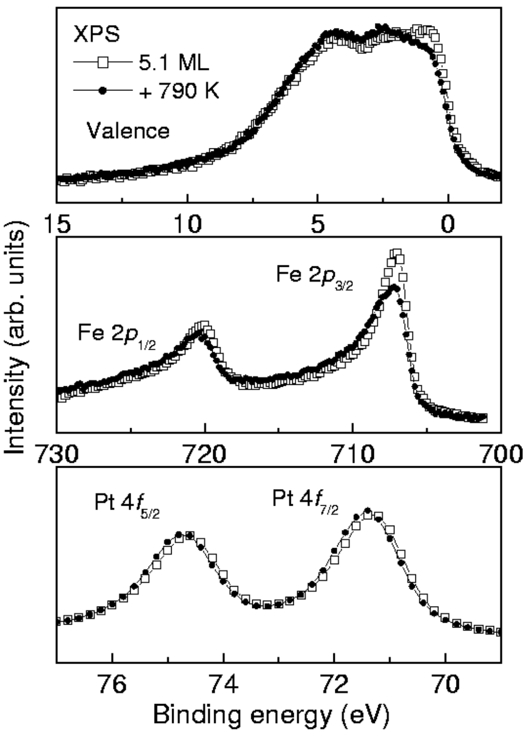

Fig. 2. XPS spectra of 5.1 ML Fe film on Pt(111): due to intermixing between Fe and Pt, the electronic structure is changed and both Fe $2p$ and Pt $4f$ levels are shifted.

binding-energy shifts in alloys [14–16] as in Ref. 17. This method is based on a Born-Haber cycle involving the heat of formation usually calculated after Miedema *et al.* [18]. The Fe 2p binding-energy shift calculated in a simplified scheme [14] is $\Delta E = 0.25\ (1 - x)$ eV. Since the measured value of the shift is +0.20 eV, we can estimate that the surface alloy composition, on average, is Fe 20 at.%. This change in the Fe 2p binding energy is also accompanied by a shift in Pd 3d peak position by +0.20 eV. By applying the same method to the Pt 4f binding energy shift, we found that $\Delta E = 0.82\ x$ eV. From this equation, the composition is Fe 24 at.%, in agreement with the estimation using the Fe 2p binding-energy shift. Since this value is very close to the composition ratio of $FePt_3$, we checked the possibility of surface ordered-alloy formation by low energy electron diffraction (LEED). We indeed observed extra spots upon annealing the films at 750 K, and this confirmed the existence of $FePt_3$ surface alloy which was paramagnetic at room temperature.

2. Fe/Pd(111)

Figure 3 shows variation of the SMOKE signals from a 2.0 ML Fe film in two different post-annealing situations. After room-temperature (RT) deposition, no hysteresis loop can be observed for either longitudinal or polar directions, and thus an Fe film on Pd(111) is paramagnetic at RT up to this thickness. Upon 450 K annealing, hysteresis curves are observed for both directions, which indicates that the magnetization vector is canted. The coexistence of in-plane and out-of-plane hysteresis curves is commonly observed in systems which show spin reorientation [6,19]. Upon 600 K post-annealing, the hysteresis loop disappears again for both directions.

For thicker films, different behavior of Kerr signals is observed after post-annealing. Ferromagnetic Kerr signals in the longitudinal direction were observed after RT deposition, but a polar signal could not be induced by any annealing process. Upon 600 K (660 K) post-annealing of a 2.5 ML (5.5 ML) film, there is an increase in coercivity as well as in Kerr intensity. Further annealing at 750 K leads to a reduction of coercivity for the 2.5 ML film. The enhancements of the Kerr signals for the 2.5 and 5.5 ML films were observed to be more than 200 % and 300 %, respectively [20]. There might have been some change in reflectivity upon post-annealing, but it is not probable that this reflectivity change alone caused such strong enhancements. For a Co-Pt(111) surface alloy, it had been estimated that reflectivity changes could explain only about one-third of the observed 300 % enhancement of the Kerr signal [21].

Microscopically, the coupling between the electric field of light and the electron spin within a magnetic medium occurs through the spin-orbit interaction, and the Kerr effect manifests itself because of the unbalanced population of electron spins for ferromagnetic materials. Therefore, a change in the Kerr signal can result from changes in spin-orbit interaction in ferromagnetic materials. In previous calculations [22,23], spin-orbit coupling enhancement was expected in the cases of Fe or Co multilayers or intermetallics with Pd or Pt, which in turn affects the polar Kerr rotations. The interdiffusion will cause some of the Fe atoms to be in contact with Pd atoms; this is different from the low temperature case, where only Fe atoms at the interface can be influenced by adjacent Pd atoms.

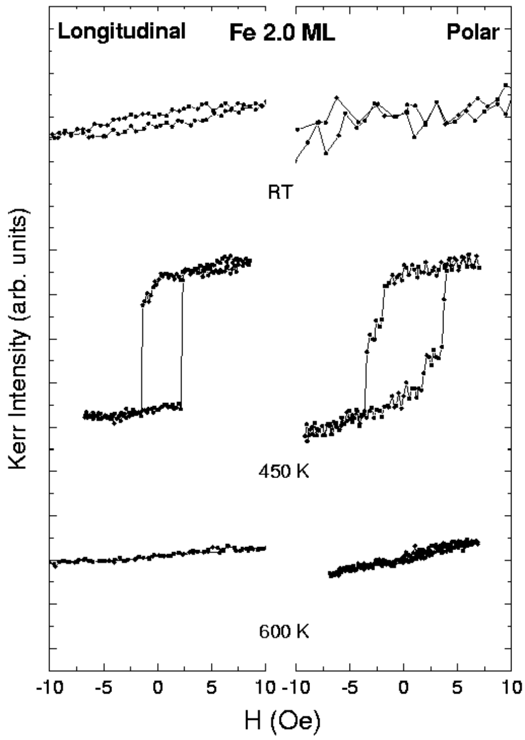

Fig. 3. SMOKE signals from a 2.0 ML Fe film on Pt(111) after room temperature deposition, with 450 and 600 K post-annealing (from Ref. 20).

Similar analysis on the XPS spectra of Fe/Pd(111), as in the Fe/Pt(111) case, has also been reported [20]. The result shows that the Fe concentration is 9 at.% when the films were post-annealed at 750 K, which was actually in agreement with 15 Fe at.% determined from the comparison of the photoelectron intensity ratio in Ref. 20.

Another piece of information we can get from a change in peak intensities is that there is some dewetting of the Fe layers upon 450 K annealing. While the Fe 2p peak intensity is reduced, the Pd 3d peak intensity is enhanced upon 450 K annealing. The apparent enhancement of both peak intensities upon 600 K annealing seems to be due to the onset of interdiffusion. We can infer that a 2.0 ML Fe film post-annealed at 450 K is relatively flat without interdiffusion at the interface. For a 2.0 ML film, since we have observed hysteresis loops both in longitudinal and polar directions, we can conclude that magnetic anisotropy is induced by a suitable change in morphology. Similar results have been reported in other systems such as Fe/Cu(100) [24] and Fe/Ag(100) [6]. The induced PMA in all these cases can be interpreted as a result of an increase in surface anisotropy after post-annealing, the magnitude of which is more than enough to compensate the decrease in roughness-induced dipolar surface anisotropy [6].

IV. SUMMARY

In summary, the magnetic and morphological properties of ultrathin Fe films grown on a Pt(111) and on a Pd(111) surfaces were investigated by using SMOKE, XPS, and LEED. By combining the experimental results, the change in morphology by a suitable annealing process was shown to induce ferromagnetism in the Fe/Pd(111) system around a critical Fe film thickness of about two monolayers. A flat surface with a sharp interface could be achieved by 450 K post-annealing, and magnetizations both in longitudinal and in polar direction were concurrently induced. Further annealing at 600 K completely demagnetized the film, because of dewetting and the onset of Fe-diluted surface alloy formation. For the Fe/Pt(111) system, intermixing was so great that post-annealing led to the Fe_3Pt ordered surface-alloy. Since the bulk Fe_3Pt is paramagnetic at room temperature, the disappearance of the ferromagnetic behavior upon annealing at 750 K can be explained.

For thicker films on Pd(111), a different type of change of the Kerr signal was observed. An enhancement of the Kerr signal was observed upon annealing steps at around 600 K and 660 K, accompanied by a reduction in the relative XPS intensity ratio and an enhancement of the Fe $2p$ PE satellite peaks. These observations indicate the formation of ferromagnetic surface alloy as a result of Fe interdiffusion. These results show that the interdiffusion as well as the morphological changes can affect the magnetic and magneto-optical properties of the ultrathin magnetic films.

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