I. INTRODUCTION

Self-organized surface nanostructures have been widely studied in the last decade because of their regular spatial ordering and specific size control in nano-scale and large area structures. The combination of a self-organized surface nanostructure with various functional materials has been found to possess application potential in many different fields, such as catalysis and spintronics. Moreover, due to the symmetry breaking and finite size effect, the magnetic behavior of ultrathin films is usually dominated by the surface and interface layers. Thus, from the point of view of interface modulation, the combination of magnetic ultrathin films with a nanostructured surface template is expected to be interesting and applicable to technology. Since 1996, T. E. Madey’s group has reported that noble metal covered W(111) undergoes massive reconstruction and exposes {112} oriented facets after 700–1000 K annealing.1–7 Subsequently, study of the faceting phenomena on microscopic tips was performed, because of the technical potential for generating highly coherent and bright electron beams, as well as optimum lateral resolution in scanning probe microscopes (SPM) and focused ion beams.8,9 Self-organized single atom tips also have been demonstrated utilizing faceting phenomena by several groups.10,11

Until now, only limited knowledge of the magnetic tip formation and the magnetic property of the faceted surface covered by a magnetic film was reported.12,13 Our latest study revealed that hydrogen adsorption promotes perpendicular magnetic anisotropy in nanostructured Fe coverage on a Pd/W{112} faceting surface.14 Hydrogen adsorption significantly enhances the perpendicular coercivity by 6–7 times, and shifts the perpendicular-to-in-plane switching critical thickness. Revenant et al. discussed the structural and morphological evolution of Co on a faceted Pt/W(111) surface upon thermal annealing.15 In their study, a CoPt alloy was formed at approximately 800 K.15 However, the magnetic behavior corresponding to the structural and morphological evolution has yet to be explored. Thus, in this experiment, we comparatively investigate Co and Fe magnetic ultrathin films grown on a nanoscale faceted Pd/W{112} surface. The morphology evolution, alloy formation, and corresponding magnetic behavior are systematically studied with the variation of annealing temperature and film thickness.

II. EXPERIMENT

The experiment was carried out in situ in a multifunctional ultrahigh vacuum (UHV) chamber with a base pressure of 2 × 10−10 Torr.3,14 After cycles of oxygen dosing and flashing to ~2000 K, a clean and well-ordered W(111) surface was prepared. The nanoscale surface facets were prepared by annealing the planar Pd/W(111) at 800–1000 K for 10 min. The faceted surface was denoted as Pd/W{112} throughout the rest of this paper, as the exposed facets are of {112} orientation. Fe and Co deposition onto the Pd/W{112} was performed with the sample held at 100 K. Temperature programmed Auger (TPA) spectroscopy was used to monitor the evolution of the element composition in real time during annealing with a heating rate of 4 K/s. The surface morphology was deduced from the low energy electron diffraction (LEED) patterns. A spot-profile analysis LEED (SPA-LEED) was used in this experiment. Therefore, a LEED spot with certain beam energy can be shifted into the screen area by controlling the electrical field in the SPA-LEED. Estimates of strength of bonding between elements and calibration of coverage were examined by temperature programmed desorption
Throughout the study, we use the TPD-calibrated monolayer (TML) as the coverage unit. TML is an operational definition using TPD measurement and might be different for different elements. Our TPD characterization and AES measurement of Co films shows that 1 TML is around 1 pseudomorphic atomic layers, indicating only 1 pseudomorphic atomic layers of Co are strongly boned to a bare W(111) surface. Moreover, the magnetic property was in situ investigated using in-plane and perpendicular magneto optical Kerr effect (MOKE) with the maximum magnetic field of 2000 Gauss at various sample temperatures.

III. EXPERIMENTAL RESULTS
A. Thermal stability and morphology

In this experiment, the faceted Pd/W{112} surface was prepared by annealing the planar 3.3 pseudomorphic layer of Pd on W(111) at 800-1000 K for 10 min, because 3 pseudomorphic layers is the minimum coverage for inducing the nano-scale facets. Fig. 1 shows the temperature-programmed Auger spectroscopy data of 3, 5, 7, 9, and 11 TML Co films deposited on a faceted Pd/W{112} surface. The Auger signal intensity of Co776 eV, Pd331 eV, and W171 eV are plotted as a function of annealing temperature. The measurement was carried out while the sample was continuously annealed with a heating rate of 4 K/s. As shown in Fig. 1(a), for 3 TML Co, the Co signal only decreased slightly with annealing temperature. 5 TML Co showed a more pronounced reduction when the sample was heated higher than 400 K. 7, 9, and 11 TML Co films were stable below 400 K, and then revealed more significant reduction in Co intensity after 400 K. The stability below 400 K is consistent with the previous studies, in which no alloy was formed at Co/Pd and Co/Fe interface at room temperature. The thicker coverage of Co delayed the transition to a higher critical temperature, which triggered the reduction of Co intensity. After annealing to 600 K, the Co intensity was gradually reduced to the intensity of 3 TML. This indicates that the apparent thickness approached 3 TML for high coverage Co after 700–800 K annealing. The reduction of Co Auger intensity from 400 to 600 K implied that morphology change or inter-diffusion occurred in the Co/Pd/W structure. Since the transition temperature of 400–600 K is much lower than the desorption temperature of Co and the binding energy of Pd/W, the desorption of Co from the Pd/W{112} surface can be excluded and inter-diffusion into the Pd/W{112} surface is not likely to be very serious around room temperature. The possible reason for the Co intensity reduction is the aggregation and 3-dimensional (3D) island formation of Co. The 3D island formation significantly reduced the exposed surface cross-section and thus led to the reduction of Co Auger intensity. Based on this proposed mechanism, after the 3D island formation of Co from 400–600 K, Co coverage on the Pd/W{112} surface becomes thinner and accordingly the Pd and W Auger signals are expected to increase. Actually, in Figs. 1(b) and 1(c), Pd and W Auger signals indeed reveal a significantly increase after 400 K annealing. After 600 K, since the Co morphology was stabilized, the Pd and W Auger signals gradually saturated at a certain value. In comparison with the Co/Pd/W{112} system, Fig. 2 shows the TPA data of 3–11 TML Fe films on the Pd/W{112} surface. As seen in Fig. 2(a), the Fe Auger signal was invariant during 100–300 K annealing. Afterward, the Fe Auger signal significantly decreased. Until ~600 K, the Fe Auger signal gradually approached an apparent thickness of 2–3 TML. Similar to the case of Co/Pd/W{112}, the reduction of Fe Auger signal was accompanied with the enhancement of Pd and W Auger intensity, indicating the 3D island formed after ~300 K annealing.

In order to analyze the temperature dependence of the TAP measurement in more detail, the TPA data of Co and Fe, as shown in Figs. 1(a) and 2(a), were differentiated by annealing temperature. Note that because random noise in TPA curves results in large errors in differentiation, we
performed the differentiation after smoothing the original data by averaging the nearest data points. This smoothing process helps to exclude random noise-induced spikes and meanwhile sustains the original cure evolution. Fig. 3 shows the differentiation of Co and Fe Auger signals as a function of the annealing temperature. For the sake of clarity, the differentiation curves are vertically shifted. The dashed lines are plotted to show the differentiation baseline in each curve. For 3 and 5 TML Co, there are small peaks located between 400–500 K and 500–600 K, respectively. This observation indicates that the Co Auger intensity decreased with a relatively slow rate. For 7 TML Co, the small peaks merged to form a flat-top peak. The differentiation value of the Co signal was invariant from 450 to 550 K, indicating a constant decreasing rate in this temperature region. For the thicker Co films of 9 and 11 TML, the maximum decreasing rate (the peak of differentiation) became larger and appeared at 450–480 K and 480–520 K, respectively. In 3–9 TML Co films, the reduction of Co Auger intensity, i.e., 3D island formation, started at 400 ± 10 K. However, for 11 TML Co film, the Co signal reduction was clearly delayed by 20 K to 420 ± 10 K. In the case of Fe/Pd/W{112}, as shown in Fig. 3(b), similar transition from a slow reduction to a significant single-step reduction was observed with increasing Fe thickness. 3–9 TML Fe films started to show reduced Fe Auger intensity, i.e., 3D island formation, at 320 ± 10 K, while 11 TML Fe film started to show reduced Co Auger intensity at a higher temperature of 340 ± 10 K.
After carrying out element composition analysis by TPA measurement, the temperature-dependent morphology evolution, especially the stability of the faceted surface, was characterized by LEED. Fig. 4 shows the LEED images of 5 TML Co and Fe/Pd/W{112}, measured at 100 K, after annealing at various temperatures. The LEED images were taken using a CCD camera, after shifting the featured spots into the screen area by applying a suitable electric field. The insets in Fig. 4 show the magnified image of the featured spots, indicated by the squares. As shown in Fig. 4(a), after deposition of 5 TML Co, the LEED images taken after 300 K annealing revealed the 3-fold splitting spots, indicating the presence of 3-sided {112}-pyramids in the faceted surface. The 3-fold splitting spots were continually sustained even after 900 K annealing. Thus, the {112} faceted surface was thermodynamically stable in Co/Pd/W{112}. In contrast, no observable LEED spots appeared for 5 TML Fe/Pd/W{112}. After 500–900 K annealing, the 1 × 1 single spots gradually formed. This indicates that the 5 TML Fe coverage did not sustain the Pd/W{112} faceting surface and gradually transferred to the (111) planar surface after high temperature annealing.

B. Magnetism

Fig. 5(a) shows Kerr magnetic hysteresis loops of n TML Co films on Pd/W{112}, measured right after deposition at 100 K. Stable square hysteresis loops were observed in the in plane direction for 3–11 TML Co. No observable magnetic hysteresis loops were found in the perpendicular direction. Both the magnetic coercivity $H_c$ and remanence monotonically increased with Co coverage. The summarized Kerr remanence is plotted as a function of film thickness in Fig. 6. The $H_c$ of Fe films decreased monotonically from 22 Oe to 10 Oe, when the thickness was increased from 3 to 11 TML. In contrast, the $H_c$ of Co film significantly increased from 50 Oe to 118 Oe, when the thickness was increased from 3 to 11 TML. The different thickness-dependent evolutions of $H_c$ in Co and Fe films might be due to the strain relaxation in the crystalline structure, or the domination of volume-contributed magnetic anisotropy in thicker films.

In our previous study, the magnetic hysteresis loop of 1.3 TML Fe/Pd/W{112} was observable at 100 K.\textsuperscript{14} The Fe magnetization switched from a perpendicular to in-plane direction at 2 TML. Hydrogen adsorption significantly enhanced the perpendicular coercivity by 6–7 times, and shifted the switching critical thickness to 2.8 TML.\textsuperscript{14} Above 3 TML, the Fe films revealed stable in-plane anisotropy. Fig. 5(c) shows the in-plane hysteresis loops of 3–11 TML Fe/Pd/W{112} films. In contrast to the $H_c$ increase in Co films, the $H_c$ of Fe gradually decreased with increasing thickness. The Kerr remanence of Fe films, as plotted in Fig. 5(d), also revealed a linear relation with the film thickness. The magnetic dead layer, estimated by extrapolating the linear fitting line, is 0.5 ± 0.5 TML.

The $H_c$ of the as-deposited Co and Fe films are plotted as a function of film thickness in Fig. 6. The $H_c$ of Fe film decreased monotonically from 22 Oe to 10 Oe, when the thickness was increased from 3 to 11 TML. In contrast, the $H_c$ of Co film significantly increased from 50 Oe to 118 Oe, when the thickness was increased from 3 to 11 TML. The different thickness-dependent evolutions of $H_c$ in Co and Fe films might be due to the strain relaxation in the crystalline structure, or the domination of volume-contributed magnetic anisotropy in thicker films.

As for the effect of thermal annealing on magnetism, Fig. 7 shows an example of a 7 TML Co film. The MOKE hysteresis loops of 7 TML Co/Pd/W{112} were measured at 100 K, after sequential annealing at different temperatures for 30 min. Fig. 7(b) summarizes the $H_c$ and Kerr remanence evolution as a function of annealing temperature. The $H_c$ was significantly enhanced by more than 2 times, from 87 Oe to 185 Oe, when the annealing temperature reached 400 K. Meanwhile, the Kerr remanance was invariant until 350 K. Afterward, 400 K annealing resulted in ~15% reduction of Kerr remanence. Fig. 8(a) summarizes the $H_c$ evolutions of the various Co films measured at 100 K, after annealing at different temperatures. The curves reveal the same tendency of monotonic increasing of $H_c$. The $H_c$ first increased slightly after 200–300 K and then was significantly enhanced by about 2–3 times when the sample was annealed at 400–450 K. 300 K annealing was enough to induce the disappearance of magnetic hysteresis loops for 3 TML Co. In 5–11 TML Co films, further annealing at 450–500 K also led to the disappearance of hysteresis loops.

In contrast to the annealing-induced $H_c$ enhancement in Co films, the $H_c$ of Fe films was always invariant after annealing to 300 K, as shown in Fig. 9(a). Although 300 K annealing did not change the $H_c$, the Kerr remanance was reduced by 10%–30%, depending on the film thickness. Further annealing at 350 K always resulted in the disappearance of the hysteresis loops. Due to the disappearance of Kerr remanance after 450–500 K annealing for Co films or 350 K annealing for Fe films, we also tried to check if the magnetization switched to the perpendicular direction, i.e., the surface normal direction, using polar-MOKE. There were no observable MOKE signals in the perpendicular direction for the Co and Fe films after annealing. This result excluded the possibility of annealing-induced spin-reorientation transition. Since the MOKE signals (Kerr remanence) of Co and Fe disappeared at
the temperatures much lower than the expected Curie temperature, there should be some other reasons for this phenomenon. Based on the TPA measurement, other possible mechanisms for the annealing-induced disappearance of MOKE signals should be the 3D-islands formation and the possible Pd-diffusion, which are discussed later.

IV. DISCUSSION

From the TPA measurement of Co films, as shown in Figs. 1 and 3(a), the thin films are stable up to at least \( \sim 400 \) K. For a thicker Co film of 11 TML, the change of TPA-signal is delayed to a higher temperature of \( \sim 420 \) K. This indicates that thicker films reveal a higher thermal stability. The Auger signals drastically change from 450 to 550 K, because of the formation of Co 3D-islands. Comparatively, as shown in Figs. 2 and 3(b), for Fe films, the as-deposited element composition and morphology are sustained until \( \sim 320 \) K. A higher thickness of 11 TML delays the critical temperature to \( \sim 340 \) K. Massive composition and morphology reorganizations occur from 380 to 500 K. 3D-islands of Fe form after this transition temperature region. The surface free

FIG. 6. Magnetic coercivity \( H_c \) of 3–11 TML Co and Fe on Pd/W\{112\} plotted as a function of film coverage. The \( H_c \) of Co increases monotonically and the \( H_c \) of Fe decreases monotonically with the increasing coverage.

FIG. 5. (a) and (c) Kerr magnetic hysteresis loops of \( n \) TML Co and Fe deposited on Pd/W\{112\}, as measured at 100K. (b) and (d) The Kerr remanence deduced from (a) and (c), plotted as a function of Co coverage. The lines show the linear fitting of the experimental data.
energy of Pd(111) is $\gamma_{Pd} = 1.6\text{--}1.9\ \text{J/m}^2$, which is smaller than that of Co $\gamma_{Co} = 2.0\text{--}2.7\ \text{J/m}^2$ and Fe $\gamma_{Fe} = 2.2\text{--}2.7\ \text{J/m}^2$.\textsuperscript{21,22} Furthermore, the lattice mismatch of fcc-Co on Pd is $-9.1\%$ and that of fcc-Fe on Pd is $-7.7\%$.\textsuperscript{21,23} The small surface free energy of Pd and the large lattice mismatch between Pd and Co (Fe) prevent the formation of smooth epitaxial Co (Fe) film on the Pd surface. Thus, the annealing-driven aggregation and 3D-island formation is straightforward. Apparently, in comparison, the Co films are more stable than are Fe films, and can be sustained to a higher annealing temperature, i.e., $\sim80\ K$ higher than the transition temperature of Fe films. This is not surprising since bulk Fe is of bcc structure. Although the lattice mismatch of fcc-Fe/Pd is smaller than that of fcc-Co, the Fe films may undergo a structural transformation from fcc to the bulk-like bcc structure, in which the lattice mismatch between Fe and Pd is even larger and the bonding between Fe and Pd becomes weaker. Accordingly, the Fe films are relatively unstable in comparison with the Co films.

The above observations about the thermal stability of Co and Fe films are strongly correlated to the temperature-dependent evolutions of their magnetism. For Co films, as shown in Fig. 8(b), 450--500 K annealing is enough to induce the disappearance of magnetic hysteresis loops. For Fe films, 350 K annealing leads to the disappearance of magnetic hysteresis loops. The critical annealing temperatures for the disappearance of MOKE hysteresis loops, i.e., 450 K for 5--9 TML Co and 350 K for 3--11 TML Fe, are close to the starting temperature of TPA variation, not the temperature of the maximum change rate. This is because of the annealing time effect. The TPA measurements shown in Figs. 1 and 2 were carried out with a heating rate of 4 K/s, while the thermal annealing in the MOKE measurements shown in Figs. 8 and 9 was carried out for 30 min in each step. Although the
change rates of composition and morphology at 450 K for Co and 350 K for Fe are not fast, a long annealing time of 30 min is still enough for the complete transformation of the morphology form a uniform film to 3D islands plus a thin wetting layer. When the sample morphology transferred from a uniform film of few nm thickness to 3D islands of a large size, e.g., hundreds of nm in height, the MOKE signal is significantly reduced to a hundredth of the original signal, because of the reduction of the apparent cross-section and the limited probing depth of MOKE (<50 nm). Therefore, after the annealing-induced morphology transition to 3D islands with a thin wetting layer, the 3D islands did not contribute an observable MOKE signal in our measurement. Two possible reasons are proposed for this observation. One is that the Pd atoms seriously mixed with Co or Fe atoms in the thin wetting layer and the Pd-Co or Pd-Fe alloys can be paramagnetic while the Pd content is too high. Or the intermixing is not so serious and the Pd-alloys are still ferromagnetic, but the alloying effect significantly enhanced the magnetic coercivity over the maximum magnetic field (1200 Oe) of our MOKE setup. Further experiments which are beyond scope of the current report are still needed to clarify the detailed mechanism.

In the TPA measurement shown in Figs. 1 and 2, for 5–11 TML Co (Fe) films, the thermal annealing reduced the Co (Fe) Auger signal to an apparent thickness of ~3 TML, but, meanwhile, the Pd Auger signal increased to a value higher than the initial Pd signal of 3 TML. This means there was still a Co (Fe) layer with an apparent thickness of ~3 TML covering the surface, but the Pd underlayer, which was initially an intermediate layer under Co (Fe), became more visible under Auger electron spectroscopy after annealing. This implies the possible diffusion of Pd into the Co (Fe) overlayer. This deduction about the possibility of Pd-diffusion is also consistent with the proposed reasons for the disappearance of the Kerr signal after 450 K (350 K) annealing for the Co (Fe) films. Moreover, the de-faceting phenomenon observed by LEED also supports the proposed Pd-diffusion explanation. In this experiment, upon annealing, an ultrathin overlayer of Pd can induce the W(111) surface to undergo massive reconstruction to expose [112] oriented facets. The faceting is driven by minimization of the surface energy but limited by kinetics. Previous studies of Pd/W(111) show that a critical coverage (about 3 pseudomorphic layers) of Pd is necessary to induce faceting and the surplus Pd tends to form 3D islands, as revealed by Auger electron spectroscopy (AES) and scanning tunneling microscopy (STM). The Pd/W{112} faceted surface might be destroyed by Pd-interdiffusion or modification of surface energy after depositing an overlayer and subsequent heating.

As shown in Figs. 5 and 6, the $H_c$ of as-deposited Co films increased with thickness, while the $H_c$ of as-deposited Fe films decreased with thickness. The $H_c$ evolution with film thickness in Co and Fe films shows the opposite trends. In previous studies on magnetic ultrathin films, the increase of film thickness may have changed the crystalline structures, e.g., strain relaxation or massive structural transition, which can correspondingly change the magnetic behavior. Furthermore, in low coverage region, the magnetic anisotropy is dominated by the surface and interface anisotropy, while in a thick film, the surface effect gradually decays and the volume magnetic anisotropy becomes dominant. In the Fe/Pd/W{112} films, a spin reorientation transition (from perpendicular to in-plane) occurred at ~2.0 TML Fe. In the SRT transition region, the $H_c$ of perpendicular and in-plane components was enhanced. After the SRT, the Fe revealed stable in-plane anisotropy and the $H_c$ gradually decreased from 22 Oe to 10 Oe when the thickness was increased from 3 TML to 11 TML. The decreasing trend and the small $H_c$ for the Fe films is reasonable, because the small crystalline magnetic anisotropy of bulk Fe, $K = 4.4 \times 10^4 \text{J/m}^3$, gradually dominated the magnetic behavior. As compared with Fe, bulk Co reveals a much larger crystalline magnetic anisotropy $K = 45 \times 10^4 \text{J/m}^3$.  

![FIG. 9. Summarized magnetic coercivity $H_c$ (a) and Kerr remanence (b) of 3–11 TML Fe/Pd/W{112} plotted as a function of annealing temperature. The measurement temperature is 100 K.](image-url)
For example, in the case of fcc-Co films on Cu(100), the bulk-contribution of magnetic anisotropy $K_b \approx 100 \times 10^3 \text{J/m}^3$ is larger than the surface-contribution of magnetic anisotropy. Thus, in thicker Co films, the larger bulk-crystalline magnetic anisotropy gradually dominates the magnetic behavior and thus leads to a larger $H_c$. Moreover, the possible strain relaxation of fct-Co films and the possible fct-to-bct structural transition of Fe films on Pd might also affect the thickness-dependent evolution of the magnetic behavior.

As shown in Figs. 8 and 9, thermal annealing increased $H_c$ enhancement in Co films, but $H_c$ was invariable in Fe films. Thin films of ordered L10 alloys such as FePd, FePt, CoPd, and CoPt have attracted much attention in recent years because of their large uniaxial magnetocrystalline anisotropy. The large coercivity ($H_c$) of FePd and CoPd films has been reported. Both the alloying and ordered L10 structure are crucial for the magnetic behavior. Usually Co films, as reported in several articles, can sustain a stable fcc or fct structure on a fcc-substrate, like Pd. The Pd diffusion into Co films forms a crystalline structure similar to L10, or fct structure on a fcc-substrate, like Pd. The Pd diffusion and magnetic properties were systematically characterized for investigation. The morphology, element composition, and thermal annealing in Fig. 9 is not enough to improve the ordering, it is reasonable to expect a minor effect on the magnetic properties, e.g., the coercivity $H_c$.

V. SUMMARY

3–11 TML of Co and Fe thin films were deposited on a nanostructured substrate, the Pd/W[112] faceting surface, for investigation. The morphology, element composition, and magnetic properties were systematically characterized with different annealing temperature. The Co thin films aggregated and formed 3D islands at 400–450 K. Co covered {112} facets remained invariant even after further annealing to 900 K. The Fe thin films aggregated and formed 3D islands at lower temperatures, 300–350 K. There was no observable faceted structure after annealing from 300 to 900 K. The magnetic coercivity ($H_c$) of as-grown Co and Fe films increased and decreased, respectively, with increasing thickness from 3 to 11 TML, due to the dominance of their bulk magnetocrystalline anisotropy. The $H_c$ of Co films was significantly enhanced by 2–4 times after annealing at 400–450 K, but the $H_c$ of Fe films sustained invariable upon thermal annealing. The reason is likely the very different crystalline structures in the fct-Co and bcc-Fe films, which lead to various Pd-diffusion effects on magnetism. These comparative studies on 3–11 TML Co and Fe films on the faceted Pd/W[112] surface clearly reveal the significant differences between the two systems and provide detailed information about the thermal stability based on various aspects.

These findings of this study are valuable for future applications.

ACKNOWLEDGMENTS

This work was financially sponsored by National Science Council of Taiwan under Grant Nos. NSC 99-2112-M-001-035, NSC 99-2112-M-003-009-MY3, NSC 102-2923-M-003-002-MY3, and NSC 102-2112-M-003-003-MY3.