Spin Reorientation Transition of Fe Ultra-Thin Films on Pd(001) Studied by X-Ray Magnetic Circular Dichroism Spectroscopy

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We investigated the magnetic anisotropy of bare and Pd-capped Fe ultra-thin films on the Pd(001) single crystal substrate using soft X-ray magnetic circular dichroism (XMCD) spectroscopy. XMCD spectra indicate that the spin reorientation transition (SRT) occurs in Fe/Pd(001). It is suggested that the SRT in Fe/Pd(001) is related to the film morphology. In Pd/Fe/Pd(001), in-plane magnetic anisotropy was observed. No evidence of perpendicular magnetic anisotropy was obtained, which is in contrast to the L\(_{10}\)-ordered FePd alloy.

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I. INTRODUCTION

Magnetic thin films and multilayers have attracted a lot of interest for several decades because of their low-dimensional magnetic properties. The reduced dimensionality and the broken symmetry at the surface or interface cause many peculiar magnetic properties, such as perpendicular magnetic anisotropy (PMA), enhanced magnetic moment, and critical behavior in the low-dimensional ferromagnetic systems. In particular, magnetic anisotropy in artificial magnetic films and multilayers is one of the important subjects in both fundamental researches and industrial applications. Several multilayers composed of 3d ferromagnetic metals and noble metals are known to possess PMA, which is expected to support a basic technology of high-density magnetic recording media [1–3]. Ferromagnetic ordered alloys with L\(_{10}\)-type crystal structure have been extensively studied as candidate materials for PMA in many years, where natural crystal structure contains an alternate arrangement of monatomic layers of ferromagnetic metals and noble metals [4, 5]. Recently, some types of the alternate monatomic layers have been artificially fabricated on single crystal substrates [6, 7]. Ultra-thin films of Pt/1 ML Fe/Pt(001), that is a single pseudo bilayers of the FePt ordered alloy, has been investigated both experimentally and theoretically [7, 8].

So far, some experimental and theoretical studies on the crystal structure and the magnetic properties of Fe ultra-thin films on Pd(001) have been reported. In the early years, the growth mode of the Fe films on the Pd(001) was very controversial problem. One research group showed the film growth was in two-dimensional (2D) layer-by-layer fashion [9], the other insisted three-dimensional (3D) island growth [10]. Jin et al. observed the surface morphology of Fe/Pd(001) accurately using scanning tunneling microscopy (STM). They revealed that the initial growth of the Fe films is in layer-by-layer mode below 3 ML, and Fe atoms or small clusters are randomly distributed on the surface in the sub-monolayer region [11]. The magnetic properties of Fe/Pd(001) were investigated by several magneto-optical Kerr effect (MOKE) and X-ray magnetic circular dichroism (XMCD) experiments. Liu and Bader reported that Fe films grown on the substrate at room temperature showed in-plane magnetic anisotropy independently of the film thickness, and that Fe films grown at 100 K showed PMA below two monolayers (ML) [9]. Le Cann et al. reported the thickness dependence of the local magnetic moment at the Fe atomic site, and revealed the strong enhancement of the orbital moment in the Fe films below a few ML [12].

Although the formation of the monatomic Fe layer on the Pd(001) substrate have been supported by the STM study by Jin et al., no investigation of magnetic properties of Pd/1 ML Fe/Pd(001) has been conducted. In addition, the magnetism of Fe films in the sub-monolayer region is still in question, because most of previous studies were mainly focused on the magnetic properties of Fe films thicker than a few ML. In order to examine the magnetic properties of Pd/Fe/Pd(001), we need detailed information concerning uncapped Fe films with thickness around 1 ML including the sub-monolayer region. In some cases of magnetic ultra-thin films, it is proved that spin reorientation transition (SRT) is induced by metallic capping layer [13, 14].

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In this study, for the Fe films with the thickness around 1 ML, we investigate the magnetic anisotropy of both bare and Pd-capped Fe/Pd(001) by means of XMCD spectroscopy [15–17]. We have found that the perpendicular magnetization is realized only for the bare Fe films on Pd(001) below the thickness of 1.2 ML, whose magnetization direction is changed into in-plane by the Pd capping. No evidence of PMA has been obtained for Pd/1 ML Fe/Pd(001). The in-plane magnetic anisotropy is quite different from the PMA behavior in ML Fe/Pd(001). The in-plane magnetic anisotropy is realized only for the bare Fe films on Pd(001) below the thickness of 1.2 ML, whose magnetization is simply obtained from the XAS, whose significant signals at 722 and 709 eV, respectively. The broad structure at the higher energy range than 722 eV is originated from a contribution of the Pd substrate. The XMCD spectrum was simply obtained from the XAS, whose significant signals at the $L_{2,3}$ edges indicate an existence of the ferromagnetic order in the Fe film.

FIG. 2: Fe $L_{2,3}$ (a) XAS and (b) XMCD spectra for 1.1 ML Fe/Pd(001). Incident photon angle is normal to the surface. $\mu_+$ and $\mu_-$ represent the absorption spectra for the sample magnetization direction parallel and anti-parallel to the incident photon spin, and the XMCD spectrum is defined as $\mu_- - \mu_+$. 

In Fig. 2, X-ray absorption spectra (XAS) and XMCD spectrum for 1.1 ML Fe/Pd(001) with out-of-plane magnetization are illustrated as typical spectra in the present experiment. We obtained the XAS for magnetization directions both parallel and anti-parallel to the incident photon spin, denoted by $\mu_+$ and $\mu_-$ in the figure. $L_2$ and $L_3$ white line peaks appear at the photon energies of 722 and 709 eV, respectively. The broad structure at the higher energy range than $L_2$ peak is originated from a contribution of the Pd substrate. The XMCD spectrum was simply obtained from the XAS, whose significant signals at the $L_{2,3}$ edges indicate an existence of the ferromagnetic order in the Fe film.

FIG. 1: Schematics of the XMCD experiment. Sample magnetization and incident photon directions for detecting (a) out-of-plane and (b) in-plane remanent magnetization.

II. EXPERIMENTAL

All experiments were conducted at the BL-14 of HiSOR at Hiroshima Synchrotron Radiation Center (HSRC) [18]. Sample fabrication and XMCD experiments were performed in situ in experimental chambers connected to the beamline in the same ultra high vacuum condition.

Single crystalline Pd(001) surface was prepared by repeated cycles of 1 kV Ar$^+$ bombardment and annealing at 980 K. Negligible contaminations on the surface was observed by Auger electron spectroscopy. Fe films and Pd capping layer were grown by the thermal deposition on the substrate at room temperature. Wedge-shaped Fe films were prepared in situ in order to reveal thickness dependence of signals. The thickness of the Pd capping layer was fixed to 1 ML. Evaporation rate was calibrated by intensity oscillations of reflection high energy electron diffraction (RHEED). We determined the evaporation rate of Fe was about 0.3 ML/min and that of Pd was about 0.2 ML/min. Film thickness was cross-checked by the intensity ratio of Fe $LMM$ (651 eV) to Pd $MNN$ (330 eV) in Auger electron spectra.

XMCD experiments were performed in remanent magnetization to investigate the magnetic anisotropy. During XMCD experiments, the sample was cooled by liquid N$_2$ and the base pressure of the chamber was below 5.0 × 10$^{-8}$ Pa. Schematics of the XMCD experiment is shown in Fig. 1. The sample was magnetized by pulse magnetic field generated by the Helmholtz coil, with magnetic flux density of 0.2 T. Sample magnetization direction was normal (parallel) to the sample surface and incident light angle was set normal (45° off normal) to the sample surface, in order to observe the out-of-plane (in-plane) magnetization. Circularly polarization ($P_c$) of incident light is 0.8 in the energy range of Fe $L_{2,3}$ edges. X-ray absorption spectra were obtained by the total electron yield method.

III. RESULTS AND DISCUSSION

In Fig. 2, X-ray absorption spectra (XAS) and XMCD spectrum for 1.1 ML Fe/Pd(001) with out-of-plane magnetization are illustrated as typical spectra in the present experiment. We obtained the XAS for magnetization directions both parallel and anti-parallel to the incident photon spin, denoted by $\mu_+$ and $\mu_-$ in the figure. $L_2$ and $L_3$ white line peaks appear at the photon energies of 722 and 709 eV, respectively. The broad structure at the higher energy range than $L_2$ peak is originated from a contribution of the Pd substrate. The XMCD spectrum was simply obtained from the XAS, whose significant signals at the $L_{2,3}$ edges indicate an existence of the ferromagnetic order in the Fe film.

Figure 3 (a) shows the normalized Fe $L_{2,3}$ XMCD spectra with several Fe thickness for the bare Fe/Pd(001). Solid and dashed curves represent the XMCD spectra for the out-of-plane and the in-plane remanent magnetization, respectively. The XMCD spectra are normalized by the integrated intensity of the absorption spectra. The spectra measured in the off-normal configuration are additionally multiplied by the geometric correction factor of $\sqrt{2}$. At 1.3 and 1.2 ML, the XMCD signals for in-plane magnetization is clearly observed, whereas those for out-of-plane are hardly detectable. At 1.1 ML, the XMCD signals for both in-plane and out-of-plane are observed. It is explained that the magnetic domains with in-plane and out-of-plane magnetization coexist equally in the film. At 0.9 and 0.7 ML, it is obvious that the in-plane magnetization is clearly observed, whereas those for out-of-plane are hardly detectable.
demagnetization decrease. We can see no XMCD signal at 0.5 ML. This is due to the suppressed Curie temperature ($T_C$) in this coverage. This result is consistent with the $T_C$ vs. Fe thickness curve reported by Liu and Bader, where the $T_C$ is extrapolated to almost 0 K at 0.5 ML [9].

In Fig. 3 (b), we summarize the XMCD intensities at the Fe $L_3$ edge, which are extracted from the normalized XMCD spectra as displayed in the Fig. 3 (a). In the Fe thickness range above 1.2 ML, the intensities for the in-plane magnetization are dominant and those for the out-of-plane are negligible. On the other hand, below 1.2 ML, the out-of-plane magnetization appears and the in-plane magnetization is weakened. Below 0.5 ML, remanent magnetization disappears in both direction due to the reduced $T_C$, as we mentioned above. It is proved that the SRT in Fe films grown on Pd(001) occurs at the thickness of 1.2 ML. As the intriguing feature, the SRT in Fe/Pd(001) is very abrupt. The transition width is very narrow in the thickness (about 0.4 ML) compared to that in other systems. For example, the SRT in Ni/Cu(001) occurs in the range of about 2 ML [19].

To examine the effect of the Pd capping layer on the magnetic anisotropy in the Fe films on Pd(001), we have measured the XMCD spectra for Pd/Fe/Pd(001), whose overlayer thickness is fixed at 1 ML. In Fig. 4, the thick-
ness dependent XMCD spectra and the $L_3$ intensities are summarized for Pd/Fe/Pd(001), in the same manner as presented in the Fig. 3. It is noted that the XMCD signals for the out-of-plane magnetization is completely absent. As a result, only in-plane magnetization has been observed, and no out-of-plane magnetization has been found in the all range of the Fe thickness. The SRT behavior is not found any longer in Fig. 4 (b). The in-plane magnetization is almost constant over the range of 0.7-2.1 ML, and it decreases toward the small thickness from 0.6 ML. Comparing these results with those from Fig. 3 (b), it is clear that the in-plane magnetic anisotropy in Pd/Fe/Pd(001) is present even in the thinner Fe thickness region, where the out-of-plane magnetization is dominant in the bare Fe/Pd(001).

From the present results for the bare Fe films on Pd(001), it is concluded that the in-plane magnetization in thick films is changed into the perpendicular magnetization in the sub-monolayer region by SRT at the thickness of 1.2 ML. As will be discussed below, it is considered that the origin of the PMA below 1.2 ML is related to the film morphology in the sub-monolayer region. The previous STM study indicates that Fe atoms or small clusters are randomly distributed at the sub-monolayer thickness. The density of the Fe clusters on the surface is gradually increased without large island formation as the thickness increases. At the thickness of 1 ML, the initial growth of the Fe film is finished and the 2D film formation is completed as a monatomic layer. It can be pointed out that the SRT nearly coincides with the completion of the 2D film. It is inferred that the cluster feature of the deposited Fe may encourage the PMA below 1 ML, from a viewpoint of shape magnetic anisotropy. Constant remanent magnetization observed even below 1 ML requires some particular magnetic coupling mechanism between the Fe clusters, because spatial continuity of the Fe film should be broken by the cluster formation in the sub-monolayer thickness. We suggest the magnetic coupling between the Fe clusters through the Pd substrate where induced Pd 4$d$ magnetic moment is detected in our Pd $M_{2,3}$ XMCD spectra of Fe/Pd(001) (not shown).

Another point to note is that no evidence of PMA is found in Pd/Fe/Pd(001) around the Fe thickness of 1 ML. Since the capping of Pd on the Fe films below 1.2 ML causes transition from the original perpendicular magnetization into the in-plane one, we consider that the capping contributes to the in-plane magnetic anisotropy rather than the PMA. The evolution of the in-plane magnetic anisotropy is not compatible with bulk and thick films of L1_0-ordered FePd alloy. In contrast to the present case of Pd/Fe/Pd(001), Imada et al. observed PMA in Pt/Fe/Pt(001) at low temperature [7]. The first principle calculation by Tsujikawa et al. showed the reduction of the occupied minority spin electrons of the Fe 3d_{yz} and 3d_{xz} orbital at the Fermi level in Pt/Fe/Pt(001), resulting in the enhancement of the orbital anisotropy that favors the PMA [8]. Because the interfacial electronic structures and the spin-orbit interaction are critical for the magnetic anisotropy in the ultra-thin films, the discrepancy in the magnetic anisotropy between Pd/Fe/Pd(001) and Pt/Fe/Pt(001) may be attributed to the difference between their electronic structures. The substrate crystals of Pd and Pt are so close in lattice constant that we expect minor difference in the film structures significant for the magnetic anisotropy.

IV. CONCLUSION

In summary, we investigated the magnetic anisotropy of bare and Pd-capped Fe ultra-thin films on a Pd(001) substrate by means of XMCD spectroscopy. The thickness dependent XMCD spectra for Fe/Pd(001) indicated that the SRT from out-of-plane to in-plane magnetization occurs around 1.2 ML with increasing thickness. The observed SRT might be related to the film morphology in the sub-monolayer region, where Fe atoms or clusters are randomly distributed on the substrate surface. No evidence of PMA was obtained for Pd/Fe/Pd(001) around the Fe thickness of 1 ML, which is in a strong contrast to the L1_0-ordered FePd alloy. Pd capping layer reorients the perpendicular magnetization to the in-plane one. This in-plane magnetic anisotropy may be attributed to the interface electronic structures.

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