Effect of surface roughness on magnetism of ultrathin Co films

M Sakamaki1, K Amemiya
Institute of Materials Structure Science, High Energy Accelerator Research Organization, Tsukuba, 305-0801 Ibaraki, Japan
E-mail: masako.sakamaki@kek.jp

Abstract. Interface roughness effect on magnetic anisotropy of Co/Au are studied by using a Au substrate with “sharp” and “rough” surfaces by means of conventional and depth-resolved XMCD techniques. From the Co L-edge XMCD and sum rule analysis, the critical Co thickness of SRT from in-plane to perpendicular magnetization are found to be shifted from ~0.8 to ~1.1 nm by Au capping. We also found the sample with “rough” interface favors in-plane magnetization more than the sample with “sharp” interface. Compared to “sharp” sample, “rough” sample has smaller $m/m_s$ values, and no significant probing-depth dependence is observed. These differences in the magnetic state at the interface between “sharp” and “rough” might be caused by structural changes. Moreover, $m/m_s$ is lager for “sharp” sample especially at the interface. This can be interpreted as a large perpendicular anisotropy at the “sharp” interface, i.e. large $K_s$, while the magnetic anisotropy of the “rough” sample is determined by $K_v$ more than by $K_s$.

1. Introduction
It is known that surface/interface roughness of magnetic thin films affects magnetic properties, such as coercivity, domain structure, and magnetic anisotropy [1-4]. These magnetic properties greatly affect the application to e.g. magnetic recording, and data storage devices. We here focus on roughness-induced anisotropy for ultrathin Co films grown on a Au(111) substrate. Several mechanisms have been invoked to explain its origin: (i) edge atom, whose electronic structure is different from atoms inside the island, would feel anisotropic potential and change the anisotropy of whole film via exchange coupling [5], (ii) strain at the interface as a magneto-elastic coupling [6], and (iii) dipolar interaction as a magnetostatic contribution [7]. However, there are few experimental/theoretical

1 To whom any correspondence should be addressed.
discussions that include all of these contributions. The structural properties strongly depend on the preparation conditions, quality of the substrate and so on. It is therefore important to perform magnetic and structural investigations on the same samples. In this respect, we used wedge-shaped Co thin layers in combination with in-situ local probes such as x-ray magnetic circular dichroism (XMCD) and low-energy electron diffraction (LEED) allowing a thickness dependence study of magnetic and structural properties. In addition, we applied depth-resolved XMCD technique [8] to the sample with different degree of roughness, which was controlled by Ar sputtering on the substrate, and evaluated the surface/interface and inner-layer magnetic moments as well as magnetic anisotropy of Co.

2. Experimental

2.1. Sample preparation
A Au(111) single crystal was cleaned by repeated cycles of Ar sputtering (2 keV) and subsequent annealing at ~950 K. We then separately fabricated rough and sharp surfaces on single substrate by sputtering only the half area of the sample. The roughness of the surface by Ar sputtering was confirmed by LEED. Wedge-shaped Co films were deposited on Au(111) at the substrate temperature of 300 K with the electron bombardment evaporation of a Co rod. The deposition rate of the Co films was determined by Auger electron spectroscopy (AES) prior to the sample preparation. The thickness of the Co layer varied from 0 to 2.0 nm. The Au-covered Co films were also prepared by subsequently evaporating Au at the substrate temperature of 300 K with the electron bombardment evaporation of a Au wire supported on a W rod. The Au deposition rate was also calibrated by AES prior to the sample preparation.

2.2. X-ray magnetic circular dichroism measurement
In addition to the total electron yield XMCD, which provides averaged information over the whole sample, the depth-resolved XMCD technique [8] was adopted. The depth-resolved XMCD provides depth-resolved and element specific magnetic information. In this technique, the probing depth of the spectrum is determined by the effective escape depth of the electrons, which depends on the electron emission angle \( \theta_d \). The emitted electrons were collected separately at different detection angles by using an imaging type detector consisting of a microchannel plate (MCP), a phosphor screen and a CCD camera. All the spectra with different probing depths were recorded at once. Partial electron

Figure 1. Co L-edge XMCD spectra for (a) Co/Au(111) and (b) Au/Co/Au(111) with “sharp” interface at the Co thickness of 0.9 nm. (c) XMCD spectra for Au/Co/Au(111) with “sharp” and “rough” interfaces at the thickness of 1.0 nm. All the spectra were taken at NI.
yield mode with a retarding voltage of 500 V was adopted, so that the Co LMM Auger electrons were mainly collected. The effective electron escape depth was experimentally determined at each $\theta_d$ from the thickness dependence of the edge-jump intensity of the films. XMCD measurements were performed on the samples remanently magnetized by a pulse magnetic field (~700 Oe) before each measurement. The spectra were recorded at normal (NI) and grazing (GI) x-ray incidences (0° and 60° from normal) to study magnetic anisotropy. All the measurements were performed at room temperature in a vacuum chamber connected to beamline BL-16A at the Photon Factory.

3. Results and discussion

It was reported that Co thin films grown on a Au(111) substrate exhibit the spin reorientation transition (SRT) from in-plane to perpendicular magnetization by capping with Au [9,10]. In the present study, the magnetic structure at the surface and interface has been investigated in order to clarify the origin of the SRT. Figure 1 shows Co L-edge XMCD spectra of Co/Au(111) with and without Au capping at the Co thickness of 0.9 nm. Here we show the result of the sample without Ar⁺ sputtering (we call this sample as “sharp” interface sample). The spectra were taken at NI, in which only the surface normal component of sample magnetization is detected, in the total electron yield mode. The XMCD intensity drastically increases upon Au capping, indicating that Au capping surely enhances perpendicular magnetic anisotropy. Co showed perpendicular magnetic anisotropy at the thickness of ~0.7-1.1 nm for “sharp” sample after Au capping. In the case of “rough” sample, perpendicular anisotropy was seen at ~0.6-1.0 nm. Figure 1(c) shows Co L-edge XMCD spectra of Au/Co/Au(111) with “sharp” and “rough” interfaces at the Co thickness of 1.0 nm. The sample with “rough” interface shows smaller XMCD intensity, which means more in-plane preference compared to the sample with “sharp” interface at the Co thickness of 1.0 nm.

3.1. XMCD sum rule analysis

Figure 2 shows XMCD sum rule [11] analysis for Co/Au(111) with and without Au capping. 3d hole number $n_{3d}$, effective spin magnetic moment $m_s$, and orbital to spin moment ratio $m_l/m_s$ are plotted as a function of Co thickness. The critical thickness of SRT, which is estimated from the thickness dependence of $m_s$, is shifted from ~0.8 to ~1.1 nm by Au capping. The sample with “rough” interface favors in-plane magnetization more than the sample with “sharp” interface at thicker region for both Co/Au(111) and Au/Co/Au(111). On the other hand, the reduction in $m_s$ at thinner region (<~0.6 nm) is not attributed to the SRT but to the phase transition to the super-paramagnetic region.

![Figure 2. Sum rule analysis of (a) Co/Au(111) and (b) Au/Co/Au(111). 3d hole number $n_{3d}$, effective spin magnetic moment $m_s$, and orbital to spin moment ratio $m_l/m_s$ are plotted.](image-url)
We thus conclude magnetostatic contribution, which stabilizes perpendicular magnetization under the presence of surface roughness [7], is negligible in the investigated samples. For \( m/m_s \), the sample with “sharp” interface shows slight larger values. The orbital moment corresponds to magnetic anisotropy constant \( K \) [12], and it means the “sharp” interface has larger \( K \). This is consistent with the result of \( m_s \). In the case of Co/Au(111), \( m/m_s \) shows slight decrease as the Co thickness increased (>~0.6 nm), and the slope of “sharp” sample is gentler than that of “rough”. The change in the slope cannot be due to the difference in \( K_s \), surface component of \( K \), but the difference in \( K_v \), volume part of \( K \). Generally the structural change, e.g. strain, structural phase transition, affects the magnitude/sign of \( K_v \). From these results, the structural difference between “sharp” and “rough” is expected. In order to separately discuss \( K_s \) and \( K_v \), structural/magnetic depth profiling is required.

3.2. Depth-resolved XMCD analysis

In the depth-resolved technique, a set of spectra at different probing depths is obtained. The XMCD spectrum recorded at smaller probing depth contains more contribution from the top Co layer, and is more surface/interface sensitive. According to XMCD sum rule, we obtained effective spin magnetic moment \( m_s \), and orbital to spin moment ratio \( m_l/m_s \) at each probing depth. Figure 3 shows probing depth dependence of \( m_s \) and \( m_l/m_s \) for Au/Co/Au(111) at the Co thickness of 0.9 nm. Both “sharp” and “rough” samples show decrease in \( m_s \) at smaller probing depth, indicating that the Co atoms at the interface have smaller effective spin moment compared to the inner layers. On the other hand, \( m_l/m_s \) increases at smaller probing depth for “sharp” sample, suggesting that the orbital magnetic moment is enhanced at the interface. These results agree with the theoretical calculation [13], and they can be explained as e.g. structural changes at the interface, lack of magnetic atoms above the film, and hybridization with the non-magnetic atoms. So far, the enhancement of the orbital moment at the interface had not been experimentally observed directly. Compared to “sharp” sample, “rough” sample shows smaller \( m_l/m_s \) values, and no significant probing-depth dependence is observed. It indicates the interface has similar orbital moment to that in the inner layers for “rough” sample. We suppose these differences in the magnetic state at the interface between “sharp” and “rough” might be caused by structural changes. Especially the uniform magnetic state observed on “rough” sample might be due to the structural phase transition or structural relaxation.

![Figure 3. Probing depth dependence of spin magnetic moment \( m_s \) and orbital to spin moment ratio \( m_l/m_s \) for Au/Co/Au(111) at the Co thickness of 0.9 nm.](image)
Moreover, the orbital moment is larger for “sharp” sample especially at the interface. This can be interpreted as a large perpendicular anisotropy at the “sharp” interface, i.e. large $K_s$. On the other hand, the magnetic anisotropy of the “rough” sample might be determined by $K_v$ more than by $K_s$.

4. Summary
We applied conventional and depth-resolved XMCD techniques to study the interface roughness effect on magnetic anisotropy of Co ultrathin films using a Au substrate with “sharp” and “rough” surfaces. From the Co $L$-edge XMCD and sum rule analysis, we found the critical Co thickness of SRT from in-plane to perpendicular magnetization is shifted from $\sim 0.8$ to $\sim 1.1$ nm by Au capping. We also found the sample with “rough” interface favors in-plane magnetization more than the sample with “sharp” interface. No significant probing-depth dependence of orbital moment was observed for “rough” sample, indicating that the orbital moment at the interface is similar to that in the inner layers. On the other hand, the orbital moment is larger for “sharp” sample especially at the interface. This might be due to a large perpendicular anisotropy at the “sharp” interface, i.e. large $K_s$. We suppose these differences in the magnetic state at the interface between “sharp” and “rough” might be caused by structural changes. Especially the uniform magnetic state observed on “rough” sample might be due to the structural phase transition or structural relaxation. A depth-structural study is underway.

5. References
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