In-situ Studies of Structure and Magnetic Properties of Co Clusters on Au(111)*

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We investigated the surface structure and the local magnetic moment of Co clusters grown on a reconstructed surface of Au(111). We performed an in-situ experiment with scanning tunneling microscopy (STM) and soft X-ray magnetic circular dichroism (XMCD) spectroscopy, to reveal the spin and orbital magnetic moments on the Co atomic site in the well-characterized cluster structure without overlayers. While the spin magnetic moment is comparable to bulk Co, the orbital magnetic moment is considerably enhanced in the clusters whose lateral size is 8.4 nm. [DOI: 10.1380/ejssnt.2014.129]

Keywords: X-ray absorption spectroscopy; Scanning tunneling microscopy; Cobalt; Clusters; Magnetic, structural, and other properties of nanostructures

I. INTRODUCTION

For development of ultra-high-density magnetic recording devices, it is necessary to minimize the magnetic domain size where single-bit information is stored. One of the solutions is to adopt bit-patterned media with predefined single magnetic domains. If a discrete magnetic domain with lateral size of 13×13 nm² stores single-bit information, the recording density of 1 Tbit/in² can be realized in theory [1]. Self-organized magnetic clusters on crystal surfaces have considerable promise as materials for the bit-patterned media, because the size of the individual cluster is smaller than a typical single domain size patterned by lithography. Structure and magnetism of the surface clusters have attracted great interests, since these properties are indispensable information to develop next-generation recording media utilizing the surface magnetic clusters.

A system of Co clusters grown on Au(111) surface is one of promising candidates for the ultra-high-density recording media. Self-assembled Co clusters are aligned regularly on the reconstructed surface of Au(111), whose individual clusters exhibit ultrathin island structure and perpendicular magnetic anisotropy. Voigtländer et al. have reported growth regime and coverage-dependent structure of the Co clusters, which is investigated by scanning tunneling microscopy (STM) [2]. Deposited Co atoms form cluster arrays spontaneously on so-called herringbone structure of the reconstructed Au(111). The nucleation of Co clusters begins at elbow sites of the herringbone structure, leading to Co islands with uniform height of 2 monolayers (ML). The lateral size of the clusters is several nanometers to a few tens of nanometers, which can be controlled by Co coverage.

In addition to the structural studies, magnetic states of the Co clusters on Au(111) have been investigated by T. Koide et al. [3]. They have revealed ferromagnetic order in the Co clusters that are sufficiently large in lateral size, and phase transition to superparamagnetic state with decreasing the cluster size. X-ray magnetic circular dichroism (XMCD) spectroscopy was carried out for Au/Co/Au(111) samples, and the spin and orbital magnetic moments of Co were determined. Enhancement of

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the magnetic moment was revealed for the overlayered Co clusters on Au(111).

So far, experimental researches on structure and magnetism of the Co clusters have been performed independently. Besides, native magnetic property of the Co clusters without overlayers have not been revealed yet. To investigate relationship between the structure and the magnetic properties exactly, in-situ experiments are preferred. In this study, we performed an in-situ experiment with STM and XMCD spectroscopy for overlayer-free Co/Au(111) sample. We estimated spin and orbital magnetic moments of Co in the overlayer-free sample, whose values were compared with the case of the overlayered Au/Co/Au(111).

II. EXPERIMENTAL

All the experiments were performed at soft X-ray beamline HiSOR-BL14 in Hiroshima Synchrotron Radiation Center, Hiroshima University [4]. The endstation of HiSOR-BL14 is equipped with an STM apparatus and a measurement system for X-ray absorption spectroscopy including XMCD experiments [5]. Sample fabrication of ultrathin films is also feasible by means of molecular beam epitaxy in the ultra high vacuum (UHV) environment. A single crystalline Au(111) substrate was cleaned by Ar+ bombardment and annealing. After the cleaning, we examined STM images of the herringbone structure on the reconstructed clean surface, where terrace width was 100-200 nm. To obtain Co clusters, we deposited Co atoms on the substrate at room temperature by e-beam evaporation. The Co clusters were observed by STM with a constant current mode at room temperature. XMCD spectra were measured at the Co L$_{2,3}$ absorption edge by total electron yield method. In the XMCD measurement, external magnetic field of 1.1 T was applied to the sample along surface normal. The direction of magnetic field was switched at each energy step during a photon energy scan of spectral measurement. All the spectra were measured at normal incidence to the surface and the circular polarization of photon was 80%. The measurement temperature was varied from 82 to 298 K.

III. RESULTS AND DISCUSSION

In our integrated experimental station to perform combination experiments of STM and XMCD, electronic and magnetic information can be extracted from the spectroscopic data for the identical sample whose surface structure is determined by atom-scale microscope imaging. Firstly, we fabricated Co clusters on the reconstructed surface of Au(111) by means of molecular beam evaporation in UHV environment. The fabricated sample was sequentially observed with the STM apparatus to examine the cluster shape and size, one of which topographic images is shown in Fig. 1. In the wide-scan image of Fig. 1(a), regularly aligned Co clusters are found in each terrace on the Au(111) surface. The ordered array structure indicates that Co clusters grow from elbow sites of the herringbone structure on the reconstructed surface of Au(111) as previously reported [2]. The close-up STM image in Fig. 1(b) shows that the Co clusters are formed as flat islands with 2 and 3 ML, which are indicated as half-light and bright area in the image, respectively. Each Co cluster is found to be isolated from the neighbors and has polygonal island shape that is elongated elliptically along ⟨110⟩ direction of the Au(111) surface. The average diameter along the minor axis of the Co clusters was estimated at 7.1±0.7 nm from a cross section profile of the island arrays along ⟨112⟩ direction. The average diameter along the major axis was also estimated at 10.0±1.1 nm. The lateral size of the Co clusters is determined to be 8.4 nm, which is defined as a diameter of the equivalent circular cylinder to the elliptical island.

Just after the STM observation, we carried out XMCD measurements for the identical sample whose STM images is shown in Fig. 1. The sample was protected from atmospheric exposure or contamination through all the experimental procedure, since the whole of our apparatus is assembled in the same UHV environment. We can access the native features of magnetism and electronic structure in the overlayer-free sample of Co/Au(111) from results of the XMCD measurements. In Fig. 2, we shows X-ray adsorption spectra of Co/Au(111) at Co L$_{2,3}$ edges. We clearly observed an intensity difference at two white lines (L$_3$ and L$_2$) with magnetization direction. To investigate the magnetic moment of Co atomic site in the clusters, we obtained the XMCD spectra from the intensity difference in the absorption spectra. In Fig. 2, the temperature dependence of Co L$_{2,3}$ XMCD spectra is shown, whose intensities are normalized by total absorption. It is found that the amplitude of the XMCD spectra, which corresponds to the magnetization, gradually increases with decreasing temperature. This is because thermal fluctuation of magnetic moment is suppressed in the low temperature region. Full saturation of magnetization is examined below 173 K under the external magnetic field of 1.1 T.

The magneto optical sum rules for XMCD provide the spin and orbital magnetic moments of the magnetic element that corresponds to the absorption edge [6, 7]. Figure 3(a) shows temperature dependence of spin magnetic moment of Co. The plotted values of the spin magnetic moment are defined as effective spin moment including a minor contribution of magnetic dipole moment. Also, it must be noted that these values are not corrected to
FIG. 2: X-ray absorption spectra measured at Co $L_{2,3}$ edges at 173 K, where external magnetic field is applied in parallel ($\mu_+$) and antiparallel ($\mu_-$) directions with respect to the photon helicity vector, and temperature dependence of the XMCD spectra for Co/Au(111) at 82, 173, 203, and 298 K.

the saturated magnetization. It is found again that the increasing value of the spin magnetic moment with decreasing temperature indicates a reduction of the thermal fluctuation of the local spin moment. The saturated value in the low temperature region corresponds to the spin magnetic moment per Co site, whose value is estimated at 1.64±0.10 $\mu_B$ from the average value below 173 K. The value of spin magnetic moment is different from previously reported one (2.03 $\mu_B$) for overlayered Co clusters with the diameter of 8.1 nm [3] but is instead similar to one (1.55 $\mu_B$) for bulk Co [8]. In our case, the 3 ML height Co atoms on each island are not negligible. Analyzing the height distribution obtained from the STM data, we have found the area ratio of 3 ML Co clusters to 2 ML ones is 0.6. The effective cluster size is therefore increased to 9.6 nm, which is proportional to the number of Co atoms per cluster. Taking the effective size into account, our experimental value of the spin magnetic moment is qualitatively consistent with the previous work. The consistency suggests minor effect of the Au overlayer on the spin magnetic moment of the Co clusters in the Au/Co/Au(111) sample.

In Fig. 3(b), ratio of the orbital magnetic moment to the spin magnetic moment is plotted as function of temperature. The orbital magnetic moment mentioned here is its perpendicular component, due to the measurement configuration. The constant ratio can be obtained from the XMCD spectra regardless of whether the magnetization is saturated or not, because the ratio is not dependent on the XMCD amplitude but dependent only on the intensity ratio between the XMCD signals at the $L_3$ and $L_2$ white lines. The ratio value is estimated at 0.19±0.08, confirmed to be constant within a measurement error. The orbital magnetic moment, which is directly deduced from the values of the spin magnetic moment and the ratio, is also estimated at 0.31±0.13 $\mu_B$. The experimental value of orbital magnetic moment of Co is slightly larger than previous one (0.24 $\mu_B$) for the overlayered Co clusters with the diameter of 8.1 nm [3]. It is found that the orbital magnetic moment is enhanced in the Co clusters, whose value is almost two times larger than bulk Co. We attribute the enhancement to the low dimensionality of ultrathin film structures of the individual Co clusters.

IV. CONCLUSIONS

We have measured local magnetic moment on Co atomic site in the Co clusters grown on the herringbone structure of the reconstructed surface of Au(111), by means of XMCD spectroscopy with soft X-ray synchrotron radiation. The sample fabrications, STM observations and XMCD measurements were carried out in situ. We have analyzed XMCD spectra for the overlayer-free sample of Co clusters whose average lateral size is 8.4 nm. It is found that the spin and orbital magnetic moments are 1.64±0.1 $\mu_B$ and 0.31±0.13 $\mu_B$, respectively. The spin magnetic moment is sensitive to the existence of...
third layers on the Co islands, the value of which is reduced toward that in bulk Co. On the other hand, the orbital magnetic moment is enhanced even in Co clusters larger than the previous work. Minor effect of the Au overlayer on the local magnetic moment of Co clusters is found at this Co coverage.

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