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## X-ray magnetic circular dichroism at $L_{23}$ edge of Co nanoclusters on Si(111) surface

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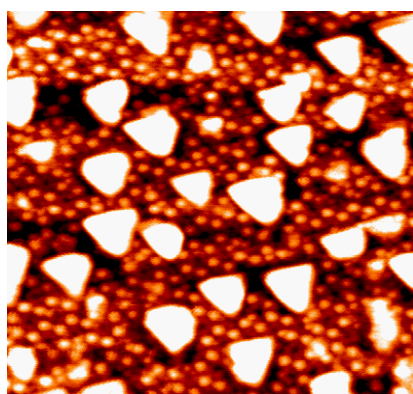
### Abstract

X-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) have been used to study self-assembled Co nanoclusters on Si(111)- $7 \times 7$  substrate. The multiplet fine structures of the XAS spectrum indicate that there is a contribution of  $\text{Co}^{3+}$  atoms in the cluster due to the Co 3d–Al 3p hybridization at the Co–Al interface. The very small and positive  $L_2$  peak compared to the negative peak at the  $L_3$  edge appearing in the XMCD spectrum indicates a localized atomic-like character of the 3d electrons. Estimation based on the sum rules gives a very large value of  $m_L/(m_S + 7m_T) = 0.37$  for the Co nanoclusters.

### 1. Introduction

Small metal clusters with diameters in the size range of 1–10 nm display a lot of novel characteristics [1]. As isolated particles, they can be considered as single giant moments of ferromagnetically coupled atomic spins, i.e. in single-domain (SD) states. There has been a long-standing interest in SD particles driven by their great potential applications in the fields of high-density data storage, sensors and spin electronic devices. Recently, Jia *et al* [2] found that a high-quality ordered Al nanocluster array could be prepared on Si(111)- $7 \times 7$  surface and

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**Figure 1.** The observed STM image (40 nm  $\times$  40 nm). Small white dots are ordered Al nanocluster arrays, on which Co nanoclusters (larger bright spots) in a triangle shape reside. The Co nanoclusters have the same orientation and constant height of 2 ML, and most of the Co nanoclusters have a uniform lateral length of 5.4 nm, which is twice as large as the Si(111)-7  $\times$  7 unit cell.

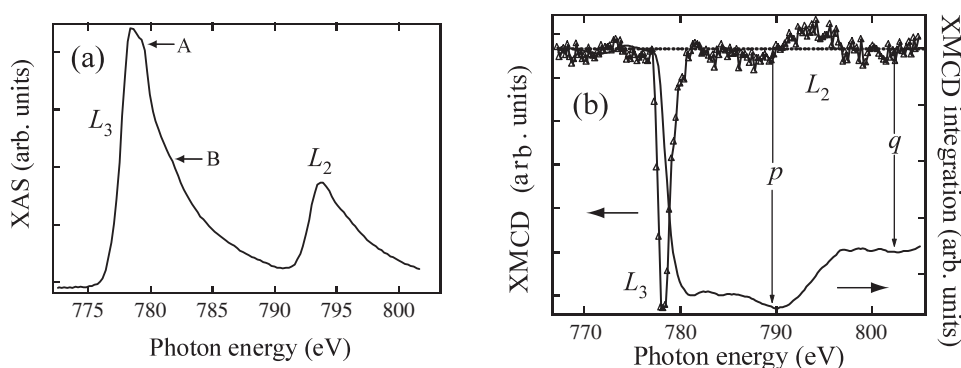
(This figure is in colour only in the electronic version)

exhibit a remarkable thermal stability, which renders an ideal template for growing nano-scale magnetic materials.

In this work, 0.24 ML Al had been evaporated onto a well ordered Si(111)-7  $\times$  7 surface prepared by well established annealing procedures to fabricate the ordered identical Al cluster arrays [2–4]. Subsequently, 1 ML Co was deposited on the template. To avoid the oxidation of the clusters, 2 nm Au film was capped on the sample. As observed by STM, shown in figure 1, the Co nanoclusters in triangle shapes reside on top of the ordered Al cluster arrays and both the clusters have the same orientation. Although their lateral lengths range from 2 to 6 nm, further study shows that about 80% of the Co nanoclusters have a uniform size of 5.4 nm, which is twice as large as the size of the Si(111)-7  $\times$  7 unit cell [5]. The most remarkable feature of these Co nanoclusters is their uniform constant height of 2 monolayers (ML). This is the same as the character of self-assembled Co clusters on Au(111) [6, 7]. The physical origin of the magic height (2 ML constant height) remains unclear. A detailed description of the preparation method and analysis of this kind of Co nanocluster will be given elsewhere [5].

## 2. Experimental details

The XAS and XMCD experiments were performed at the BL25-SU of SPring-8 [8, 9]. Circularly polarized light was supplied from a twin-helical undulator, with which almost 100% polarization was obtained at the peak of the first-harmonic radiation. Co  $L_{2,3}$  XAS spectra were measured by means of a total photoelectron yield method by directly detecting the sample current on changing the photon energy ( $h\nu$ ). The measurement was performed with both the incident light and the magnetization perpendicular to the sample surface. The XMCD spectra were taken with a fixed applied magnetic field (1.4 T) and a 1 Hz helicity switching of circularly polarized undulator radiation, which was realized by an on–off sequence of constituent electron orbit bumps [10]. This method is very effective for removing background noise and our experiments show that this is vital for the capped cluster sample. In the present paper, the XMCD spectrum is defined as  $(\mu_+ - \mu_-)$ , where  $\mu_+$  and  $\mu_-$  represent the absorption spectra for the direction of majority spin (which is opposite to the direction of the magnetization) parallel



**Figure 2.** (a) Co  $L_{2,3}$  XAS spectrum (arrows A and B indicate the multiplet structures). (b) Co  $L_{2,3}$  XMCD spectrum and its integrated value (solid curve).  $p$  and  $q$  are the integrals needed in the sum-rule analysis.

and antiparallel to the photon helicity, respectively. The XAS and XMCD measurements were made at a temperature of 43 K.

### 3. Results and discussion

Figure 2(a) shows the Co  $L_{2,3}$  XAS spectrum of the Co nanoclusters, where a background is subtracted from the raw data. We observe  $L_2$  and  $L_3$  core absorption edges at the incident photon energies ( $h\nu$  values) of 778.4 and 793.7 eV, respectively. The superpositions of peaks in the  $L_3$  edge are very clear, as marked by arrows A and B. These multiplet fine structures observed in the Co  $L_{2,3}$  XAS spectrum can be attributed to  $\text{Co}^{3+}$  in an octahedral environment [11]. We assume that this kind of  $\text{Co}^{3+}$  character comes from the Co–Al covalence bonding at the Co–Al interface. The present interfacial Co 3d–Au 5d hybridization would be small and negligible due to the fact that the 5d band of bulk Au is located well below the Fermi level. The possibility of Co 3d–Si 3p hybridization is also excluded because of the significant removal of the Si dangling bonds by the Al nanoclusters [2]. It is well known that the relative integrated intensity ratio between the  $L_3$  and  $L_2$  edges, which is represented as  $R_0 = I(L_3)/I(L_2)$ , can be related to the 3d spin states of the material [12]. Here  $R_0$  is estimated to be 3.1, which is much larger than the statistical value of 2.0. Such a high intensity ratio indicates that the 3d ground state of Co nanoclusters is in a high spin state [12].

The XMCD spectrum for the Co nanoclusters is shown in figure 2(b) (left-hand ordinate axis). One finds a clear XMCD ( $\mu_+ - \mu_-$ ) with negative sign at  $h\nu = 778.4$  eV. Compared to the strong amplitude at the  $L_3$  edge, the peak with positive sign at the  $L_2$  edge is very small. Included in the figure is the integrated value of the XMCD spectrum (right-hand ordinate axis). Here  $q$  denotes the integrated value over the whole  $h\nu$  range ( $L_3 + L_2$ ), and  $p$  represents the integrated XMCD only in the  $L_3$  region. The orbital magnetic moment contribution versus the spin/dipole magnetic moment can be expressed as  $m_L/(m_S + 7m_T) = 2q/(9p - 6q)$  [13, 14]. We find that only  $p/q$ , which is independent of the XAS intensity and the 3d hole number, is needed in the evaluation. The estimated value of the present Co nanocluster is about 0.37, which is much enhanced compared to that of the bulk Co (0.09) [15]. Besides, it is much larger than the values reported in the other Co nanocluster systems [16, 17]. But we find a good correspondence with the results of the Co impurities on alkali metal (Na, K) films when the Co coverage is around 0.07 ML [18]. We attribute the remarkable contribution of the orbital magnetic moment to the very localized 3d electron in the Co cluster.

#### 4. Conclusion

In summary, we have carried out the XAS and XMCD measurements for self-assembled Co nanoclusters on Al-terminated Si(111)- $7 \times 7$  substrate. The multiplet structure of the XAS spectrum indicates nearly the  $\text{Co}^{3+}$  configuration in the Co cluster. The XMCD spectrum has been clearly observed under an applied field of 1.4 T at 43 K. The estimation based on the sum rules gives out a very large contribution of the orbital magnetic moment such as  $m_L/(m_S + 7m_T) = 0.37$ . It is suggested that  $m_L$  is directly linked to the magnetic anisotropy. For the late 3d transition metals, the easy direction of magnetization corresponds to the crystalline axis with the largest value of the orbital magnetic moment  $m_L$  [19]. The present result must be responsible for the magnetic property of the Co cluster.

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