# Magnetic Anisotropy Multilayers on Al<sub>2</sub>O<sub>3</sub>(0001) with Co Seed Layer

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The influence of Co seed layer on the structure of the Au/Co multilayers was studied by X-ray diffraction and reflection high-energy electron diffraction (RHEED), and compared to its effect on the perpendicular magnetic anisotropy in these films. The Au buffer layer grown on  $Al_2O_3(0001)$  substrates display a large fraction of twin crystalline structure, giving rise to a lack of lateral continuity in the film. The initial deposition of a few monolayers of Co onto  $Al_2O_3(0001)$  substrates prior to deposition of the Au buffer layer yielded (111) epitaxial films without twins. The Co seed layer improves the crystalline quality of Au buffer layer and subsequent multilayer. From the results of X-ray diffraction observations, we confirmed that the interfaces of Au/Co multilayers with Co seed layer are sharper than those with no seed layer. The magnetic anisotropy energy of Au/Co multilayers increased by using the Co seed layer, and the high quality of structure results larger perpendicular magnetic anisotropy. *Key words* : Seed layer, Interface roughness, Perpendicular magnetic anisotropy, Crystal structure, Multilayers

### 1. INTRODUCTION

In recent years the perpendicular magnetic anisotropy of metallic multilayers has been attracting much attention both from a fundamental standpoint and for applications. In several multilayers consisting of Co as a ferromagnetic element, it has been reported that the large uniaxial magnetic anisotropy with the axis perpendicular to the film plane is observed when Co layer thicknesses are smaller than several monolayers. It is known that a Au/Co multilayer system is one of the multilayer systems that exhibit marked perpendicular magnetic anisotropy [1].

Among the many combinations of film and substrate materials, much attention has been paid for the growth of metal on metal since the discoveries of fascinating properties of metallic multilayers, for example, perpendicular magnetic anisotropy and giant magneto-resistance. As has been reported in some metallic multilayers, it is well known that these properties are strongly associated with both the sharpness of interface and the quality of epitaxy. In order to achieve a better understanding of the origins of the properties of metallic multilayers, it is important to control the growth of the thin films.

In order to create multilayers with both well-defined crystal and interfacial structures, we used a seeded epitaxy technique in the growth of Au/Co on single crystal  $Al_2O_3(0001)$  substrates by MBE. Seeded epitaxy has been expected to be a powerful method to improve thin film growth in MBE deposition [2-4] and suputter deposition [5-7]. The key to seeded epitaxy is that one can stabilize multiple crystal structures or orientations of a given element using a single substrate, by changing the initial buffer layer. Seed layers of several nm of Pt, for example, have proven useful for growing (111) oriented epitaxial Co films on  $Al_2O_3(0001)$  substrates [6]. In this paper we show predeposition of a thin Co seed layer on the  $Al_2O_3$  (0001) substrate prior to deposition of a Au buffer layer substrate yielded high structural quality of Au/Co(111) multilayers.

#### 2. EXPERIMANTAL

The Au/Co multilayers were grown by molecular beam epitaxy (MBE) equipped with RHEED. The base pressure was 10<sup>-8</sup> Pa range and the pressure during deposition did not exceed  $5 \times 10^{-7}$  Pa. Film deposition was e-beam evaporation sources for both Co and Au. A Al<sub>2</sub>O<sub>2</sub>(0001) single crystal was used as a substrate. The substrate was cleaned by heating at 850°C for 30 min. Co as a seed laver was deposited on the substrate at the rate of 0.004 nm/s ~0.006 nm/s, and the thickness was varied between 0.2nm ~5nm. After deposition of the Co seed layer, a 50 nm Au buffer layer was deposited at 200°C and was then annealed at 700°C for 60 min to obtain a flat surface. On The buffer layer Au/Co multilayer were fabricated at the deposition rates of 0.004 nm/s ~0.006 nm/s for Co and 0.004 nm/s ~0.006 nm/s for Au, which were controlled by using quartz thickness monitor. The Au/Co multilayers were grown at room temperature.

The morphology of the surface after deposition was confirmed by RHEED. The periodic compositional modulations and crystallographic structures of these multilayers were analyzed by X-ray diffractions (XRD) using CuKa radiation. The magnetic properties of the multilayers were investigated by means of vibrating sample magnetometer (VSM). All measurements were carried out at room temperature. The strength of the magnetic field applied perpendicular and in-plane to the thin film. The values for the magnetic anisotropy were obtained from the area between the in-plane and perpendicular magnetization curves as measured in VSM.

#### 3. RESULTS AND DISCUSSION

Fig.1 shows RHEED patterns of the surface of Au buffer layers and Al<sub>2</sub>O<sub>3</sub>(0001) substrates. The electron beam with the energy of 20keV impinged on the film surface at a glancing angle of about 1.8°. The smoothness of the film surface is confirmed by the elongation of the RHEED streaks. Fig. 1(a), (b) shows the RHEED diffraction patterns on a Al<sub>2</sub>O<sub>2</sub>(0001) substrate after annealing at 850° for 30min. The incident beam is parallel to the [1100](Fig. 1(a)) and  $[2\overline{1}\ 10]$  (Fig. 1(b)) direction. These patterns exhibit strong Kikuchi features and characteristic of smooth surface. Fig. 1(c), (d) shows the RHEED patterns of 50 nm Au buffer layer deposits on the Al<sub>2</sub>O<sub>3</sub>(0001) substrate annealed for1h at 700°C. The diffraction patterns consist of double streaks, indicating that the growth of Au(111) on Al<sub>2</sub>O<sub>2</sub>(0001) is two-dimensional with twins related two equivalent epitaxial orientations. The RHEED pattern of Fig. 1(d) is almost the same as the pattern diffracted from the incident azimuth rotated by 30° (Fig. 1(c)). Identical RHEED images could be repeated after every change of the angle by 30°. Without using the Co seed layer, the twins formation could not be avoided by varying the growth conditions. The RHEED patterns of the Au buffer layer with predeposition of 0.2 nm of Co as a seed layer were also same patterns without the Co seed layer (not shown).

With predepositions of more than 0.4 nm of Co on  $Al_2O_3(0001)$  prior to the deposition of Au, the crystal structures of the Au buffer layers were drastically changed. Fig. 1(e),(f) shows the RHEED patterns of  $Al_2O_3(0001)/Co3nm/$ Au50nm after annealing at 700°C for 1h. The rod spacing and crystal orientation of Fig. 1(e),(f) indicates that the RHEED patterns are of the fcc-Au(111) surface. The incident beam is parallel to the  $[0\bar{1}1]$  (Fig. 1 (e)) and  $[\bar{1}\bar{1}2]$  (Fig. 1(f)) direction it. The sharp single streaks and the Kikuchi lines in Fig. 1(e),(f) indicates that the surface is smooth and ordered. They also show that the Au buffer layer surface had the same crystallographic symmetry as the  $Al_2O_3(0001)$  substrate. The epitaxial relationship between Au(111) and  $Al_2O_3(0001)$  is determined from the RHEED patterns as follows:

Au[111]  $\parallel$  Al<sub>2</sub>O<sub>3</sub>[0001] and Au(110)  $\parallel$  Al<sub>2</sub>O<sub>3</sub>(1010).

The growth of thin films of Pt on  $Al_2O_3(0001)$  substrates, by MBE, is reported by R. F. C. Farrow *et al.* [6]. They find that the seed layers of several nm of Pt films on  $Al_2O_3(0001)$  substrates have a high structural perfection. The second-neighbor Pt atoms in Pt(111) plane from a hexagon which is only 0.9% larger than the basal-plane unit of  $Al_2O_3(0001)$ . On the other hand, the second-neighbor Au atoms in Au(111) plane is 4.8% larger than the basal-plane unit of  $Al_2O_3(0001)$ . Scince Au has a lager lattice misfit to the substrate, Au buffer layers are not epitaxially grown on  $Al_2O_3(0001)$ . Co seed layers were also grown on  $Al_2O_3(0001)$  substrates with twins. But Au buffer layers deposited on the Co seed layer have monocrystalline structure with no twins. We have never achieve a bet-



Fig. 1 RHEED patterns of the surface of  $Al_2O_3(0001)$  after annealing at 850° for 30min (a, b), 50 nm Au buffer layer deposits on  $Al_2O_3(0001)$  substrate annealed for 1h at 700°C (c, d), and 500Å Au buffer layer deposits on  $Al_2O_3(0001)/Co3nm$  annealed for 1h at 700°C (e, f). The incident electron beam was parallel to  $Al_2O_3[1\overline{1}00]$  (a, c, e) and  $Al_2O_3[2\overline{1}\overline{1}0]$  (b, d, f).



Fig.2 RHEED patterns of completed Au/Co multilayers with(a) and without(b) a 3 nm of a Co seed layer. The incident electron beam was parallel to  $Al_2O_3[1\overline{100}]$ .

ter understanding of the mecanism of Au growth on the Co seed layer.

Fig.2 shows RHEED patterns of completed Au/Co multilayers with (Fig. 2(b)) and without (Fig.2(c)) 3 nm of the Co seed layer. The thickness of Co and Au layer was fixed at 1 nm and 2 nm respectively. The total number of bilayers was set to 20. Both of them maintained structure in the early stages up to a final layer of multilayer. In other words, the multilayers with no seed layer display a large fraction of twin crystalline structure, giving rise to a lack of lateral continuity in the film, and the multilayers



Fig.3 Low-angle X-ray diffraction patterns of Au/Co multilayers with a no Co seed layer. (a) [Co0.2nm/Au2nm]<sub>50</sub>, (b) [Co0.6nm/Au2nm]<sub>25</sub>, (c) [Co1nm/Au2nm]<sub>20</sub>, (d)[Co2nm/Au2nm]<sub>10</sub>.

with the Co seed layer display single crystalline structure.

We examined the effect of the Co seed layer on the structures of interface using X-ray diffraction. From highangle X-ray diffraction patterns, we confirmed that all the samples have a (111) orientation along the growth direction (not shown). A set of low-angle X-ray diffraction patterns of Au/Co multilayers with and without a 3 nm Co seed layer are shown in Fig. 3 and Fig. 4, respectively. The finite-size peaks resulted from interface of X-ray refractions from the film surfaces and the film-substrate interface appear for Co seeded epitaxial multilayers. The Bragg peak intensity of Co seeded epitaxial multilayers is stronger than that of normal ones. The increase of Bragg peak intensity and the appearance of finite-size peaks suggest the decrease of cumulative random variations in layer thickness whose magnitude gives indirect information of degree of layer roughness [8,9]. Therefore, we consider the Co seed layer also decreased interfacial roughness and made interfaces more flat. The use of this seeded epitaxy results in a highly improved structural quality of Au/Co multilayers.

Fig.5 shows the effective magnetic anisotropy energy  $K_u$  per unit volume of magnetic layer as a function of magnetic layer thickness  $t_{co}$ . If the total magnetic anisotropy



Fig.4 Low-angle X-ray diffraction patterns of Au/Co multilayers with a 3nm Co seed layer. (a)  $[Co0.2nm/Au2nm]_{50}$ , (b)  $[Co0.6nm/Au2nm]_{25}$ , (c)  $[Co1nm/Au2nm]_{20}$ , (d) $[Co2nm/Au2nm]_{10}$ .

originates from two-dimensional magnetic anisotropy at interfaces  $K_s$  per unit area and volume-dependent anisotropy  $K_v$  per unit volume,  $K_u$  can be phenomenologically expressed as,

$$K_{u} \cdot t_{c_{0}} = K_{v} \cdot t_{c_{0}} + 2K_{s}$$
(1)

The  $K_v$  contains contributions from shape, magnetocrystalline and magneto-elastic anisotropy. The anisotropy data show linear thickness dependence, except for  $t_{co} < 0.8$ nm, where the values are below the straight line. Since, in the Au/Co multilayers, the Co layers are strained in the regions near the interfaces and misfit dislocations are distributed widely in those regions [10]. If the strains in Co layers decrease linearly with increasing distance from the interfaces to Au,  $K_u \cdot t_{co}$  has a falloff at a small  $t_{co}$  values as shown in Fig. 5.

The too much difference cannot be seen in the curves between the Co seeded epitaxial multilayers and normal ones. The value obtained for  $K_s$  of the Co seeded epitaxial multilayers from Fig. 4, 0.446  $\pm$  0.010  $\times$  10<sup>-7</sup> J/cm<sup>2</sup>, is the almost same value for  $K_s$  of normal ones from Fig. 4, 0.452  $\pm$  0.014  $\times$  10<sup>-7</sup> J/cm<sup>2</sup>. However, the  $K_s$  value of the Co seeded epitaxial multilayers deduced from Fig. 4 is -6.67  $\pm$  0.16  $\times$  10<sup>-1</sup> J/cm<sup>3</sup>, which is lager than that of the normal ones by 10% (-7.33  $\pm$  0.22  $\times$  10<sup>-1</sup> J/cm<sup>3</sup>). The Co seeded



Fig. 5 Dependence of  $K_{\mu} \cdot t_{c_0}$  on  $t_{c_0}$  for Au/Co multilayers.

Table I Anisotropy energy of  $K_v$ ,  $K_{d'}$  and  $K_{MC} + K_{ME}$  of Au/Co multilayers with and without 3 nm of Co seed layer.

Sample	K , (× 10 <sup>-1</sup> J/cm <sup>3</sup> )	K d (× 10 <sup>-1</sup> J/cm <sup>3</sup> )	$K_{MC} + K_{ME}$ (× 10 <sup>-1</sup> J/cm <sup>3</sup> )
Co seed layer	6.67±0.16	-11.7±0.7	5.03 ± 0.72
No seed layer	-7.33 ± 0.22	-11.9±1.1	4.58±1.08

epitaxial multilayers exhibit a magnetic easy axis perpendicular to film plane up to a larger magnetic layer thickness than multilayers without Co seed layer.

Volume anisotropy energy  $K_{v}$  is generally written as,

 $K_{v} = K_{d} + K_{MC} + K_{ME}$ (2)where the first term  $K_d$  is the demagnetization energy which is shape contribution,  $K_d = -2\pi M_s^2$  where  $M_s$  is the saturation magnetization per unit volume, and  $K_{MC}$  is magnetocrystalline energy of the bulk Co, and the last magnetoelastic energy  $K_{MF}$  is the contribution due to stress and the resulting strain. Table I summarizes the  $K_{v}$ ,  $K_{d}$ , and  $K_{MC}$  +  $K_{\mu\nu}$  value of Au/Co multilayers. The value obtained for  $K_{\mu}$ of the Co seeded epitaxials multilayer is almost same as normal ones. The  $K_{MC} + K_{ME}$  value of the Co seeded epitaxial multilayers is lager than that of the normal ones by ~10%. The increase of  $K_{v}$  with addition of the Co seed layer may be explained to be due to structural change and/ or strain contribution. It shows that the magnetic anisotropy energy increased by using the seed layer, and high quality of structure results larger perpendicular magnetic

## anisotropy.

#### 4. CONCLUSION

We have investigated the effect of a thin Co seed layer on the growth of a Au buffer layer and Au/Co multilayers on  $Al_2O_3(0001)$  single crystal substrates by MBE. The main results obtained are summarized as follows;

(1) The Au buffer layer grown on  $Al_2O_3(0001)$  substrates display a large fraction of (111) twin crystalline structure. On the contrary, the Au buffer layer with predeposition of more than 0.4 nm of a Co seed layer onto  $Al_2O_3(0001)$  substrates prior to deposition of the Au buffer layer yielded (111) epitaxial films with no twins.

(2) The Co seed layer improves the crystalline quality of subsequent Au/Co multilayers. From the results of Xray diffraction observations, the interfaces of Au/Co multilayers with Co seed layer are sharper than those with no seed layer. The use of the Co seed layer improved the periodicity and structural quality in this system.

(3) The magnetic anisotropy energy of Au/Co multilayers increased by using the Co seed layer, and the high quality of structure results larger perpendicular magnetic anisotropy.

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