



Influence of layer thickness on the structure and the magnetic properties of Co/Pd epitaxial multilayer films

Kousuke Tobari*, Mitsuru Ohtake, Katsumasa Nagano, Masaaki Futamoto

Faculty of Science and Engineering, Chuo University, Bunkyo-ku, Tokyo 112–8551, Japan

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ABSTRACT

Co/Pd epitaxial multilayer films were prepared on Pd(111)_{fcc} underlayers hetero-epitaxially grown on MgO(111)_{B1} single-crystal substrates at room temperature by ultra-high vacuum RF magnetron sputtering. *In-situ* reflection high energy electron diffraction shows that the in-plane lattice spacing of Co on Pd layer gradually decreases with increasing the Co layer thickness, whereas that of Pd on Co layer remains unchanged during the Pd layer formation. The CoPd alloy phase formation is observed around the Co/Pd interface. The atomic mixing is enhanced for thinner Co and Pd layers in multilayer structure. With decreasing the Co and the Pd layer thicknesses and increasing the repetition number of Co/Pd multilayer film, stronger perpendicular magnetic anisotropy is observed. The relationships between the film structure and the magnetic properties are discussed.

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1. Introduction

Co/Pd multilayer films with perpendicular magnetic anisotropy have been investigated for applications like magnetic recording media [1–3], magnetic random access memory devices [4], etc. The effects of layer thickness [5], underlayer material [6], and Ar gas pressure [7] on the magnetic properties have been investigated by employing Co/Pd multilayer polycrystalline films prepared on glass substrates. The films generally consist of fcc(111) preferred-texture crystals including small volume of crystals with other orientations. The reduction in the Co layer thickness is reported to promote perpendicular magnetic anisotropy [5]. The microstructure and the magnetic properties are influenced by the crystallographic orientation and the layer thickness of Co and Pd layers. Furthermore, the interface structures as well as the strain in the Co and the Pd layers are expected to influence the magnetic properties, where the lattice mismatch between fcc-Co and fcc-Pd is fairly large at about 9%. There is a possibility that the crystal structure of Co may change between fcc and hcp depending on the strain in the multilayer structure.

In order to investigate the detailed structural and the magnetic properties, well-defined epitaxial films are useful, since the film uniformity and the magnetic anisotropy are well controlled. In our previous study [8], Co/Pd epitaxial multilayer films were prepared on Pd underlayers with different orientations by RF magnetron sputtering. The Co/Pd multilayer film formed on Pd(111)_{fcc} underlayer

showed a stronger perpendicular anisotropy than that formed on Pd(011)_{fcc} underlayer. On the contrary, the multilayer formed on Pd(001)_{fcc} underlayer was easily magnetized when the magnetic field was applied along the in-plane direction.

In the present study, Co/Pd multilayer films were prepared on Pd(111)_{fcc} underlayers hetero-epitaxially grown on MgO(111)_{B1} single-crystal substrates by varying the Co and the Pd layer thicknesses from 8.6 and 11.4 nm to 0.09 and 0.11 nm, respectively. Co₅₀Pd₅₀ (at%) alloy films were also prepared on Pd(111)_{fcc} underlayers by employing a CoPd alloy target in order to compare the structural and the magnetic properties with those of Co/Pd multilayer film. The detailed Co and Pd layer structures were studied by *in situ* RHEED during film preparation process and the resulting multilayer film structures were examined by in-plane X-ray diffraction (XRD).

2. Experimental procedure

Thin films were prepared on polished MgO(111)_{B1} substrates by using an RF magnetron sputtering system, where the base pressures were lower than 4×10^{-7} Pa. Substrates were heated at 600 °C for 1 h in the ultra-high vacuum chamber. The distance between the target and the sample was fixed at 150 mm. The Ar gas pressure was 0.67 Pa and the rf powers for Pd, Co, and Co₅₀Pd₅₀ (at%) targets with 3 in. diameter were, respectively, kept constant at 30, 49, and 38 W, where the deposition rate was 0.02 nm/s for all materials.

The film layer structures are [Pd(t_{Pd} nm)/Co(t_{Co} nm)]_n/Pd(10 nm)/MgO(111)_{B1} and CoPd(20 nm)/Pd(10 nm)/MgO(111)_{B1}. A 10 nm-thick Pd(111)_{fcc} underlayer was prepared by hetero-epitaxial growth

* Corresponding author. Tel.: +81 3 3817 1862; fax: +81 3 3817 1847.

E-mail address: tobari@futamoto.elect.chuo-u.ac.jp (K. Tobari).

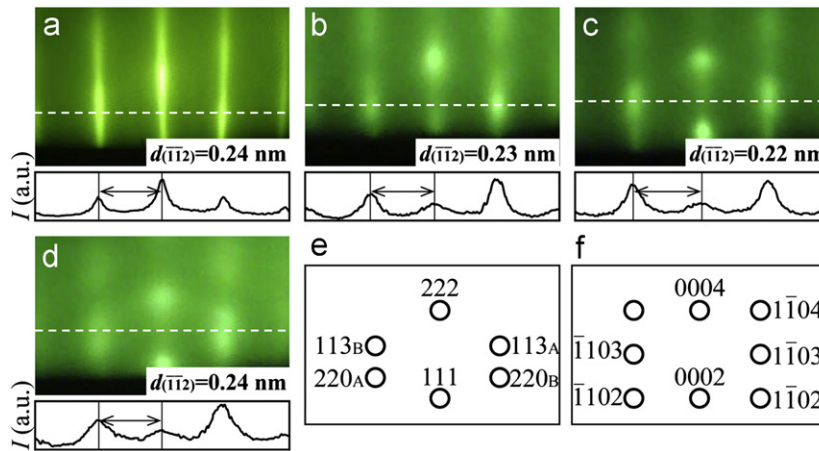
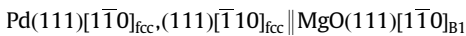


Fig. 1. (a)–(d) RHEED patterns observed for a Pd(11.4 nm)/Co(8.6 nm)/Pd(10 nm)/MgO(111)_{B1} specimen and the RHEED intensity profiles along the fcc[$\bar{1}\bar{1}\bar{2}$] direction. (a) A 10 nm-thick Pd underlayer grown on MgO(111)_{B1} substrate, a Co layer of (b) 1.0 nm and (c) 2.5 nm formed on the Pd underlayer, and an upper Pd layer of (d) 0.2 nm deposited on the Co layer. The incident electron beam is parallel to the MgO[$1\bar{1}0$]_{B1} direction. The in-plane lattice spacings, $d(\bar{1}\bar{1}\bar{2})$, are determined from the distances between RHEED spots shown in the arrows in the intensity profiles of (a)–(d). The spot maps of (e) and (f) correspond to fcc(111) and hcp(0001) textures, respectively. The symbols, A and B, correspond to the orientation relationships of Types A and B explained in the text, respectively.

on an MgO(111)_{B1} substrate at 300 °C. The epitaxial orientation relationship of



was determined by comparing the reflection high energy electron diffraction (RHEED) patterns observed for the Pd underlayer [Fig. 1(a)] and the substrate. And then, a [Pd(t_{Pd} nm)/Co(t_{Co} nm)]_{*n*} multilayer or a 20 nm-thick Co₅₀Pd₅₀ alloy layer was deposited on the underlayer at room temperature. The total thickness and the composition of Co/Pd multilayer were fixed at 20 nm and Pd-50 at% Co, respectively. Here, the thicknesses of Pd and Co layers were, respectively, defined as

$$t_{\text{Pd}} = 20 / \{n[1 + (a_{\text{Co}})^3 / (a_{\text{Pd}})^3]\}, \quad t_{\text{Co}} = [(a_{\text{Co}})^3 / (a_{\text{Pd}})^3] t_{\text{Pd}},$$

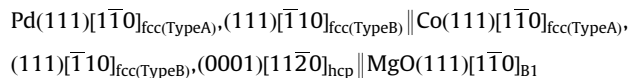
where a_{Pd} and a_{Co} are lattice constants of bulk fcc-Pd crystal ($a_{\text{Pd}} = 0.390$ nm [9]) and bulk fcc-Co crystal ($a_{\text{Co}} = 0.354$ nm [10]). The repetition number, n , was varied from 1 to 100. The compositions of Co/Pd multilayer and Co₅₀Pd₅₀ alloy layer were confirmed by energy dispersive X-ray spectroscopy and the errors were less than 5 at%. The details of film preparation are similar to our previous study [8].

The surface structure during RF-sputter deposition process was studied by RHEED. The film structure was investigated by in-plane X-ray diffraction with Cu-K α radiation ($\lambda = 0.15418$ nm). The magnetization curves were measured using a vibrating sample magnetometer.

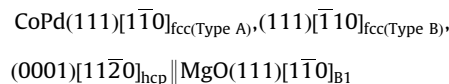
3. Results and discussion

Co/Pd multilayer films epitaxially grew on Pd(111)_{fcc} underlayers for all the samples. Co₅₀Pd₅₀ (at%) alloy films were also obtained on Pd(111)_{fcc} underlayers. Fig. 1 shows the RHEED patterns observed for a Pd(11.4 nm)/Co(8.6 nm) bi-layer film ($n=1$) grown on Pd(111)_{fcc} underlayer. The RHEED patterns observed for the Co layer and the upper Pd layer consist of spots corresponding to fcc(111) texture and streaks along the fcc[111] direction, as shown in Fig. 1(b)–(d). The spots consist of two fcc(111) reflections, as shown by the symbols, A and B, in the RHEED spot map of Fig. 1(e). The streaks observed for the upper Pd layer show that the layer has atomically flat terraces. On the other hand, the streaks observed for the Co layer indicate that the layer has atomically flat terraces and/or involves stacking faults

along the fcc[111] direction. The crystal structure of Co varies between hcp and fcc by introduction of stacking faults parallel to the close-packed plane. The existence of Co(0001)_{hcp} crystal is confirmed by XRD analysis, which is described later. The epitaxial orientation relationships of



are determined by RHEED. The Co layer consists of two-type fcc and one-type hcp variants, whereas the upper and the lower Pd layers consist of two-type fcc variants. The orientations of two-type fcc variants are rotated around the film normal by 180° with respect to each other and the variants with Type A and B relationships have atomic stacking sequences of ABCABC... and ACBACB..., respectively. The in-plane lattice spacings of Pd/Co bi-layer film were measured from the distances between RHEED spots [8]. The in-plane lattice spacing of Co layer gradually decreases and reaches a constant value of 0.22 nm beyond 2.5 nm-thick Co deposition, as shown in Fig. 1(b) and (c). This gradual change in lattice spacing is considered to be partially due to lattice strain caused by the lattice mismatch between Co and Pd layers (−9.0%) and partially due to atomic mixing around the Co/Pd interface. Atomic sites of Co crystal are partly replaced by Pd atoms with larger atomic radius and there is a possibility that a CoPd alloy phase is formed. The formation of CoPd alloy phase is also recognized by XRD. The CoPd alloy phase is formed in the crystallographic orientation relationships of



The in-plane lattice spacing of upper Pd layer is kept constant at 0.24 nm from the beginning of Pd deposition till the end of 11.4 nm-thick Pd layer formation. At the Pd/Co interface, a CoPd alloy phase is also considered to be formed, since the lattice constant of CoPd alloy close to that of Pd crystal [11]. Fig. 2(a) and (b) shows the RHEED patterns observed for a [Pd(0.11 nm)/Co(0.09 nm)]₁₀₀ multilayer film ($n=100$). The variation of in-plane lattice spacing is not recognized for all the Co and the Pd layers, and the lattice spacings of Co and Pd layers are kept constant at 0.24 nm. The atomic mixing is enhanced for thinner Co and Pd layers in Co/Pd multilayer structure. Fig. 2(c) shows the

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