Preparation of FePd/MgO/FePd Tri-layer Film on SrTiO₃(001) Single-Crystal Substrate

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An FePd/MgO/FePd tri-layer film is prepared on an SrTiO₃(001) single-crystal substrate by employing a two-step method consisting of sputter deposition at 400 °C followed by annealing at 600 °C. An *L*₁₀-FePd(001) single-crystal lower layer with the *c*-axis perpendicular to the substrate surface is obtained on the SrTiO₃ substrate. An MgO(001) single-crystal layer is formed on the FePd lower layer. An FePd upper layer epitaxially grows on the MgO interlayer. The upper layer consists of *L*₁₀(001) crystal involving a small volume of *L*₁₀(100) crystal with the *c*-axis parallel to the substrate surface. The order degrees of *L*₁₀(001) crystals in the FePd lower and the upper layers are 0.67 and 0.65, respectively. A very flat surface is realized for the tri-layer film. The tri-layer film shows a perpendicular magnetic anisotropy reflecting the magnetocrystalline anisotropy of *L*₁₀(001) crystal. A formation of fully epitaxial *L*₁₀-FePd/MgO/*L*₁₀-FePd tri-layer film with a perpendicular magnetic anisotropy is demonstrated.

Key words: FePd/MgO/FePd tri-layer film, epitaxial growth, *L*₁₀ ordered phase, perpendicular magnetic anisotropy, SrTiO₃(001) substrate

1. Introduction

Perpendicularly magnetized thin films with high uniaxial magnetocrystalline anisotropy (K_u) energies have been investigated for high-density recording media and magnetoresistive random access memory (MRAM) devices. For MRAM device applications, epitaxial (001)-oriented tri-layer films consisting of MgO and *L*₁₀ ordered alloy with high K_u have attracted much attention¹⁻⁷. The crystal structure of FePt-, CoPt-, or FePd-alloy around the equiatomic composition varies between disordered (*A*1) and ordered (*L*₁₀) structures. The *L*₁₀ ordered alloys have K_u values greater than 10⁷ erg/cm³ along the *c*-axis direction⁸. Tri-layer films consisting of MgO and FePt²⁻⁶ or CoPt⁴⁻⁷ have been prepared on (001)-oriented single-crystal substrates. An FePd layer has been employed in combination with a very thin Fe seed layer in the preparation of FePd/MgO/FePt tri-layer on MgO(001) substrate, and a high order degree is reported⁹. The ordering temperature varies depending on the magnetic material and FePd-alloy is reported to show higher order degrees compared with those of FePt- and CoPt-alloy thin films^{10,11}. Thus, FePd-alloy is a desirable magnetic material for both the upper and the lower magnetic layers. However, there are very few reports on the preparation of FePd/MgO/FePd tri-layer structure.

The surface flatness, the crystallographic and the magnetic properties are the important issues in the fabrication of practical magnetic devices in addition to the magnetic film order degree. In our previous studies, *L*₁₀ ordered FePd single-layer films with flat surfaces were prepared on MgO(001) substrates by using a two-step method; low temperature deposition at room temperature¹² or 200 °C¹³ followed by high temperature annealing at 600 °C. In the present study, an FePd/MgO/FePd tri-layer film is formed on an SrTiO₃(001) single-crystal substrate by employing the two-step method. SrTiO₃(001) substrate is employed for accurate evaluation of MgO layer by avoiding the overlap of X-ray diffraction (XRD) reflection. When an MgO substrate is used, it is not easy to extract the crystallographic information of deposited MgO layer. The initial deposition temperature of 400 °C in the two-step method is optimized in a pre-experiment to

promote the epitaxial growth while keeping a very flat surface structure. In the present study, the film growth process and the film structure are investigated in detail for respective layers in a tri-layer structure with very flat surface. The magnetic properties of a tri-layer structure consisting of epitaxial thin films are also investigated.

2. Experimental procedure

Thin films were prepared on SrTiO₃(001) substrates by using a radio-frequency (RF) magnetron sputtering system equipped with a reflection high-energy electron diffraction (RHEED) facility. The base pressures were lower than 4×10^{-7} Pa. Before film formation, SrTiO₃ substrates were heated at 600 °C for 1 h in the ultra-high vacuum chamber to obtain clean surfaces. Figure 1(a) shows the RHEED pattern observed for an SrTiO₃ substrate after heating. A clear diffraction pattern corresponding to a clean SrTiO₃(001) surface is observed, as shown in the schematic diagram of Fig. 1(b). The deposition rates were 0.020 and 0.015 nm/s for Fe₅₀Pd₅₀ (at. %) and MgO targets, respectively.

An FePd(40 nm)/MgO(40 nm)/FePd(40 nm) tri-layer film was deposited on an SrTiO₃ substrate at 400 °C, and then, the sample was annealed at 600 °C for 1 h. Although a layer thickness around 1 nm is generally used for MRAM devices, a thicker layer of 40 nm was employed in the present study for accurate characterization of epitaxial growth relationship by XRD. In order to characterize the FePd lower and the MgO layers, FePd single-layer and MgO/FePd bi-layer films were also prepared under conditions similar to the case of FePd/MgO/FePd tri-layer film formation.

The film surface structure during sputter deposition process was studied by RHEED. The film structure was investigated by XRD with Cu-K α radiation ($\lambda = 0.15418$ nm). The surface morphology was observed by atomic force microscopy (AFM). The magnetization curves were measured by using a vibrating sample magnetometer.

The notations of crystallographic plane and direction are different between *A*1 and *L*₁₀ structures. In the present study, the *A*1-based lattice notation is applied to the *L*₁₀ structure for simple comparison with the *A*1 structure. The long-range order degree (*S*) value

Fig. 1 (a) A RHEED pattern and (b) the schematic diagram of an SrTiO₃(001) substrate after heating. The incident electron beam is parallel to SrTiO₃[100]. The filled and the open circles respectively correspond to the fundamental and the superlattice reflections.

Fig. 2 RHEED patterns observed for (a) the FePd single-layer, (b) the MgO interlayer, and (c) the FePd upper layer of an FePd/MgO/FePd tri-layer film deposited on an SrTiO₃(001) substrate at 400 °C and observed for (d) the FePd upper layer of tri-layer film after annealing at 600 °C. The incident electron beam is parallel to SrTiO₃[100].

Fig. 3 Schematic diagrams of RHEED patterns simulated for (a) A1(001), (b) L1₀(001), (c) L1₀(100), and (d) B1(001) surfaces. The diagrams are calculated by using the lattice constants of (a) $a_{A1-FePd} = 0.3804 \text{ nm}^{15)}$, (b, c) $a_{L10-FePd} = 0.3850 \text{ nm}$, $c_{L10-FePd} = 0.3715 \text{ nm}^{16)}$, and (d) $a_{B1-MgO} = 0.4212 \text{ nm}^{17)}$. The incident electron beam is parallel to (a) A1[100], (b) L1₀[100], (c) L1₀[010] and L1₀[001], or (d) B1[100]. The filled and the open circles respectively correspond to the fundamental and the superlattice reflections.

is estimated from the XRD data by using the following relation^{13,14)},

$$S = \sqrt{\frac{I_s}{I_f} \times \frac{(f_{Fe} + f_{Pd})^2}{(f_{Fe} - f_{Pd})^2} \times \frac{(L \times D \times A)_f}{(L \times D \times A)_s}}, \quad (1)$$

where I is the integrated intensity, f is the atomic scattering factor of Fe or Pd, L is the Lorentz-polarization factor, A is the absorption factor, D is the temperature factor, and the subscripts of s and f respectively refer to FePd(001) superlattice and FePd(002) fundamental reflections.

3. Results and discussion

Figure 2(a) shows the RHEED pattern observed for

Fig. 4 (a) Out-of-plane and (b) in-plane XRD spectra measured for an FePd single-layer film prepared by deposition on an SrTiO₃(001) substrate at 400 °C followed by annealing at 600 °C. The scattering vector of in-plane XRD is parallel to SrTiO₃[100]. The intensity is shown in a logarithmic scale.

Fig. 5 (a) Out-of-plane and (b) in-plane XRD spectra measured for an FePd/MgO/FePd tri-layer film prepared by deposition on an SrTiO₃(001) substrate at 400 °C followed by annealing at 600 °C. The scattering vector of in-plane XRD is parallel to SrTiO₃[100]. The intensity is shown in a logarithmic scale.

an FePd layer deposited on an SrTiO₃(001) substrate at 400 °C. A clear diffraction pattern consisting of only streaks is observed. The pattern corresponds to A1(001) or L1₀(001) single-crystal surface, as shown in the schematic diagrams of Figs. 3(a) or (b). The epitaxial orientation relationships of A1- and L1₀-FePd crystals with respect to the SrTiO₃ substrate are estimated by RHEED as

$$\begin{aligned} \text{FePd(001)[100]}_{A1} &\parallel \text{SrTiO}_3(001)[100], \\ \text{FePd(001)[100]}_{L10} &\parallel \text{SrTiO}_3(001)[100]. \end{aligned}$$

When a streak pattern is observed, the identification of crystal structure is difficult, since the A1(001) and the L1₀(001) patterns are very similar. The crystal structure is therefore determined by XRD, which is described later. Figure 2(b) shows the RHEED pattern observed for an MgO layer deposited on the FePd lower layer at 400 °C. A clear diffraction pattern corresponding to B1(001) single-crystal surface is recognized, as shown in the schematic diagram of Fig. 3(d). The epitaxial

Fig. 6 (a-1)–(d-1) AFM images observed for (a-1) SrTiO₃ substrate and for (b-1) FePd single-layer, (c-1) MgO/FePd bi-layer, and (d-1) FePd/MgO/FePd tri-layer films prepared by deposition on SrTiO₃(001) substrates at 400 °C followed by annealing at 600 °C. (a-2)–(d-2) Cross-sectional profiles measured along the dotted lines in the AFM images of (a-1)–(d-1).

orientation relationship of

$$\text{MgO}(001)[100]_{B1} \parallel \text{FePd}(001)[100]_{A1 \text{ or } L10}$$

is determined by RHEED. Figure 2(c) shows the RHEED pattern observed for an FePd layer deposited on the MgO layer at 400 °C. A diffraction pattern corresponding to $A1(001)$ or $L1_0(001)$ surface is observed, similar to the case of FePd lower layer deposited on an SrTiO₃ substrate. The epitaxial orientation relationship is thus

$$\text{FePd}(001)[100]_{A1 \text{ or } L10} \parallel \text{MgO}(001)[100]_{B1}.$$

Figure 2(d) shows the RHEED pattern observed for the FePd upper layer of the FePd/MgO/FePd tri-layer film after annealing at 600 °C. The diffraction pattern does not change before and after annealing.

The crystal structure of FePd lower layer in the FePd/MgO/FePd tri-layer structure was evaluated by employing a 40-nm-thick FePd single-layer film prepared by deposition on an SrTiO₃ substrate at 400 °C followed by annealing at 600 °C. Figure 4 shows the out-of-plane and the in-plane XRD spectra measured for the single-layer film. FePd(001) superlattice reflection is recognized in addition to FePd(002) fundamental reflection in the out-of-plane spectrum, whereas FePd(001) superlattice reflection is absent in the in-plane spectrum. The result indicates that the FePd lower layer of the FePd/MgO/FePd tri-layer film does not involve $L1_0(100)$ crystal with the c -axis parallel to the substrate surface but consists of $L1_0(001)$ crystal with the c -axis perpendicular to the substrate surface.

Fig. 7 Magnetization curves of (a) FePd single-layer and (b) FePd/MgO/FePd tri-layer films prepared by deposition on an SrTiO₃(001) substrates at 400 °C followed by annealing at 600 °C. SrTiO₃[100] is the direction of applied field for in-plane measurement.

The S value is estimated from the out-of-plane XRD data to be 0.67. A higher S value is expected by further increasing the annealing temperature^{11,18}.

Figure 5 shows the XRD spectra measured for the FePd/MgO/FePd tri-layer film after annealing at 600 °C. In the out-of-plane spectrum, FePd(200) reflection is recognizable in addition to FePd(001) and FePd(002) reflections. The in-plane XRD shows FePd(001) and FePd(002) reflections in addition to FePd(200) reflection. These results show that the FePd upper layer consists of a mixture of $L1_0(001)$ and $L1_0(100)$ crystals. The crystallographic orientation relationship of $L1_0(100)$ crystal with respect to the MgO(001) layer is determined by out-of-plane and in-plane XRD as

$$\text{FePd}(100)[001]_{L10} \parallel \text{MgO}(001)[100]_{B1}.$$

When the atomic arrangements of MgO(001) and $L1_0$ -FePd(100) surfaces which respectively show four-fold and two-fold symmetries are considered, $L1_0$ -FePd(100) crystal with the orientation relationship of

$$\text{FePd}(100)[010]_{L10} \parallel \text{MgO}(001)[100]_{B1}$$

is apparently formed on the MgO(001) layer. Thus, the FePd upper layer consists of one $L1_0(001)$ and two $L1_0(100)$ variants. The c -axes of two $L1_0(100)$ variants are in-plane and rotated around the film normal by 90° each other. The S values of $L1_0(001)$ and $L1_0(100)$ crystals included in the FePd upper layer are respectively calculated from the out-of-plane and the in-plane XRD data to be 0.65 and 0.37. Such difference in the order parameter for $L1_0$ crystals formed in a same film has been reported¹¹. The reason is attributed to the difference of distortion along the different orientations in a film which is caused by a mismatch between the substrate and the film. The S of $L1_0(001)$ crystal is twice larger than that of $L1_0(100)$ crystal. Furthermore, the volume ratio of FePd(100) to FePd(001) crystal is small, because a clear RHEED reflection corresponding to FePd(100) crystal formation is not observed as shown in

Fig. 2(d). The volume ratio of $\text{FePd}(001)_{\text{lower}}$: $\text{FePd}(001)_{\text{upper}}$: $\text{FePd}(100)_{\text{upper}}$ is estimated to be 1 : 0.68 : 0.32. Here, the volume ratio of upper layer is estimated from the out-of-plane XRD data assuming that the XRD intensity is proportional to the crystal volume. The overlap of XRD intensity from the lower layer, where only the $L1_0(001)$ crystal is included, is considered in the estimation. Therefore, the $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer film is considered to show a perpendicular magnetic anisotropy reflecting the magnetocrystalline anisotropy of $L1_0(001)$ crystal.

Figures 6(a-1)–(d-1) show the AFM images observed for SrTiO_3 substrate and for FePd single-layer, MgO/FePd bi-layer, and $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer films prepared by deposition on SrTiO_3 substrates at 400 °C followed by annealing at 600 °C. The delicate differences in the AFM images of Figs. 6(b-1) and (d-1) are possibly reflecting the surface diffusion property of FePd on SrTiO_3 substrate and MgO layer. Figures 6(a-2)–(d-2) show the cross-sectional profiles measured along the dotted lines in the AFM images of Figs. 6(a-1)–(d-1). The surface arithmetical mean roughness (R_a) values of substrate and respective films are 0.1, 0.2, 0.8, and 0.5 nm. Flat surfaces are realized for all the cases, which suggests that flat FePd/MgO interfaces are realized.

Figures 7(a) and (b) show the magnetization curves measured for FePd single-layer and $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer films annealed at 600 °C, respectively. Perpendicular magnetic anisotropies are observed for both films, as expected from the structure analysis by XRD. Similar in-plane magnetization curves are observed when measured along another direction including $\text{SrTiO}_3[110]$, which indicates that the in-plane magnetic anisotropy of epitaxial FePd layer is weak. The perpendicular magnetic anisotropy slightly decreases for the tri-layer film when compared with the case of single-layer film. This is possibly due to that the FePd upper layer film involves a small volume of $L1_0(100)$ crystal. The shape of perpendicular magnetization curve of single-layer film is typical for an $\text{FePd}(001)$ epitaxial film¹⁹⁾. On the contrary, the tri-layer film shows the perpendicular magnetization curve (Fig. 7(b)) different from that of single-layer (Fig. 7(a)). This is possibly due to an increase of coercivity of the FePd upper layer.

In the present study, a formation of fully epitaxial $L1_0\text{-FePd}/\text{MgO}/L1_0\text{-FePd}$ tri-layer film is demonstrated by using an $\text{SrTiO}_3(001)$ single-crystal substrate. The crystallographic orientation, the surface flatness, and the magnetic property are investigated. These are important properties in designing a tri-layer structure for magnetic device applications. It is necessary to reduce the thicknesses of FePd and MgO layers for practical device applications and to further study the influences of layer thickness on the film structure and the magnetic property.

4. Conclusions

An $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer film is prepared on an $\text{SrTiO}_3(001)$ substrate by using a two-step method: deposition at 400 °C followed by annealing at 600 °C. The film growth, the film structure, and the magnetic properties were investigated. The following results are obtained.

- 1) An $L1_0\text{-FePd}(001)$ single-crystal layer with the c -axis perpendicular to the substrate surface

epitaxially grows on an $\text{SrTiO}_3(001)$ substrate. An $\text{MgO}(001)$ single-crystal layer is formed on the FePd layer. An FePd layer deposited on the MgO layer consists of $L1_0(001)$ crystal involving a small volume of $L1_0(100)$ crystal with the c -axis parallel to the substrate surface. A fully epitaxial $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer film is obtained on SrTiO_3 substrate.

- 2) The S values of $L1_0(001)$ crystal in FePd lower and upper layers are 0.67 and 0.65, respectively. The S of $L1_0(100)$ crystal in the FePd upper layer is 0.37. The volume ratio of $\text{FePd}(001)_{\text{lower}}$: $\text{FePd}(001)_{\text{upper}}$: $\text{FePd}(100)_{\text{upper}}$ is estimated to be 1 : 0.68 : 0.32.
- 3) The $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer film prepared by two-step method has a flat surface with the R_a value around 0.5 nm.
- 4) The $\text{FePd}/\text{MgO}/\text{FePd}$ tri-layer film shows a perpendicular magnetic anisotropy reflecting the magnetocrystalline anisotropy of $L1_0(001)$ crystal.

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References

- 1) Y. Taniguchi, Y. Miura, K. Abe, and M. Shirai: *IEEE Trans. Magn.*, **44**, 2585 (2008).
- 2) M. Hagiuda, S. Mitani, T. Seki, K. Yakushiji, T. Shima, and K. Takanashi: *J. Magn. Magn. Mater.*, **310**, 1905 (2007).
- 3) P. de Person, P. Warin, M. Jamet, C. Beigne, and Y. Samson: *Phys. Rev. B*, **76**, 184402 (2007).
- 4) N. Inami, H. Naganuma, T. Hiratsuka, G. Kim, T. Miyazaki, K. Sato, T. J. Konno, M. Oogane, and Y. Ando: *J. Magn. Soc. Jpn.*, **34**, 293 (2010).
- 5) N. Inami, G. Kim, T. Hiratsuka, H. Naganuma, M. Oogane, and Y. Ando: *J. Phys.: Conf. Ser.*, **200**, 052008 (2010).
- 6) T. Hiratsuka, G. Kim, Y. Sakuraba, T. Kubota, N. Inami, H. Naganuma, M. Oogane, T. Nakamura, K. Takanashi, and Y. Ando: *J. Appl. Phys.*, **107**, 09C714 (2010).
- 7) G. Kim, Y. Sakuraba, M. Oogane, Y. Ando, and T. Miyazaki: *Appl. Phys. Lett.*, **92**, 172502 (2008).
- 8) T. Klemmer, D. Hoydick, H. Okumura, B. Zhang, and W. A. Soffa: *Scr. Metall. Mater.*, **33**, 1793 (1995).
- 9) S. Yoshimura, S. Omiya, G. Egawa, and H. Saito: *IEEE Trans. Magn.*, **47**, 4417 (2011).
- 10) S. Yoshimura, S. Omiya, G. Egawa, H. Saito, and J. Bai: *J. Phys.: Conf. Ser.*, **266**, 012114 (2011).
- 11) M. Ohtake, S. Ouchi, F. Kirino, and M. Futamoto: *J. Appl. Phys.*, **111**, 07A708 (2012).
- 12) A. Itabashi, M. Ohtake, S. Ouchi, F. Kirino, and M. Futamoto: to be published in *Eur. Phys. J. B*, (2012).
- 13) A. Itabashi, M. Ohtake, S. Ouchi, F. Kirino, and M. Futamoto: *IEEE Trans. Magn.*, **48**, 3203 (2012).
- 14) B. D. Cullity, in *Elements of X-Ray Diffraction* (Addison-Wesley, Massachusetts, 1956) pp. 104–137.
- 15) L. Yiping, G. C. Hadjipanayis, C. M. Sorensen, and K. J. Klabunde: *J. Appl. Phys.*, **75**, 5885 (1994).
- 16) Y. V. Pal'guyev, A. A. Kuranov, P. N. Syutkin, and F. A. Didorenko: *Phys. Met. Metallogr.*, **42**, 46 (1976).
- 17) Z. Feng, V. S. Babu, J. Zhao, and M. S. Seehra: *J. Appl. Phys.*, **70**, 6161 (1991).
- 18) P. Caro, A. Cebollada, F. Briones, and M. F. Toney: *J. Cryst. Growth*, **187**, 426 (1998).
- 19) D. Ravelosona, A. Cebollada, F. Briones, C. D. Paniagua, M. A. Hidalgo, and F. Batallan: *Phys. Rev. B*, **59**, 4322 (1999).

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