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Preparation of $L1_0$ ordered FePd, FePt, and CoPt thin films with flat surfaces on MgO(001) single-crystal substrates

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Abstract. FePd, FePt, and CoPt alloy thin films are prepared on MgO(001) single-crystal substrates by employing a two-step method consisting of sputter deposition at low-temperature (room temperature) followed by high-temperature annealing at 600 °C. FePd, FePt, and CoPt single-crystal films with disordered structure ($A1$) grow epitaxially on MgO substrates at room temperature. By annealing these disordered films at 600 °C, a transformation from $A1$ to $L1_0$-ordered phase occurs. The order degrees of annealed FePd, FePt, and CoPt films are 0.63, 0.38, and 0.16, respectively. The films before annealing have very flat surfaces and the flat surfaces are kept after annealing. The arithmetical mean surface roughness values of annealed FePd, FePt, and CoPt films are 0.3, 0.1, and 0.2 nm, respectively. The two-step method is considered to be useful for preparation of FePd, FePt, and CoPt alloy thin films with flat surfaces, which are necessary for practical applications.

1 Introduction

$L1_0$ ordered alloy thin films with high uniaxial magnetocrystalline anisotropy energies (K_u) such as FePd, FePt, and CoPt have been investigated for applications like perpendicular magnetic recording media, perpendicularly magnetized magnetoresistance random access memory devices, etc. For such practical applications, realization of very flat surface is required in addition to achievement of high order degree and control of the c-axis direction. However, high process temperature is generally necessary to obtain $L1_0$ ordered structure [1–6]. FePd, FePt, and CoPt thin films deposited at high substrate temperatures around 600 °C had rough surfaces, though $L1_0$ ordered phase was formed in these films [7]. In our previous study, $L1_0$ ordered FePd alloy epitaxial films with very flat surfaces were realized on MgO single-crystal substrates of (001), (110), and (111) orientations by using a two-step method, low-temperature deposition at 200 °C followed by high-temperature annealing at 600 °C [8]. In the present study, the two-step method is applied to FePt and CoPt films in addition to FePd film. The detailed structure and the magnetic properties of these films are compared.

2 Experimental procedure

FePd, FePt, and CoPt alloy films were prepared on polished MgO(001) single-crystal 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, substrates were heated at 600 °C for 1 h in the chamber to obtain clean surfaces. The surface structure was checked by RHEED. The RHEED patterns observed for substrates exhibited *Kikuchi* patterns (not shown here), indicating that the surfaces were clean and smooth. The arithmetical mean surface roughness (R_a) values measured by atomic force microscopy (AFM) were less than 0.1 nm. $Fe_{50}Pd_{50}$, $Fe_{50}Pt_{50}$, and $Co_{50}Pt_{50}$ (at. %) alloy targets of 3 in. diameter were employed. The distance between target and substrate was 150 mm. The RF powers for FePd, FePt, and CoPt targets were respectively fixed at 35, 43, and 45 W and the Ar gas pressure was kept constant at 0.67 Pa, where the deposition rate was 0.02 nm/s for all materials. FePd, FePt, and CoPt films of 40 nm thickness were deposited on MgO(001) substrates at room temperature (RT). The thickness was employed for accurate structural characterization of the films through X-ray diffraction (XRD) analysis. Then, the films were annealed at 600 °C for 1 h. The film compositions were confirmed by energy dispersive X-ray spectroscopy and the errors were less than 4 at. % from the target compositions. The structural properties were investigated by RHEED and XRD with Cu-Kα radiation ($\lambda = 0.15418$ nm). The surface morphology was observed by AFM. The R_a values were estimated from AFM data taken from of 2 μm ×2 μm area. The magnetization curves were measured by using a vibrating sample magnetometer.

The notations of crystallographic plane and direction are different between disordered ($A1$) and ordered ($L1_0$)

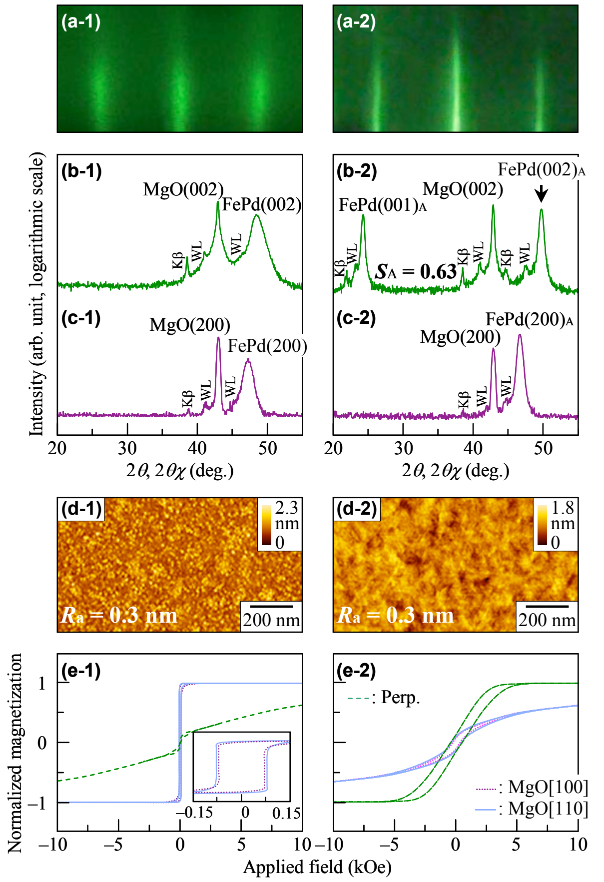

Fig. 1. (a) RHEED patterns, (b) out-of-plane and (c) in-plane XRD spectra, (d) AFM images, and (e) magnetization curves (a-1, b-1, c-1, d-1, e-1) of an FePd film deposition at RT and (a-2, b-2, c-2, d-2, e-2) of the film annealed at 600 °C after deposition at RT. The incident electron beam of RHEED and the scattering vector of in-plane XRD are parallel to MgO[100].

structures. In the present study, $A1$-based lattice notation is applied to the $L1_0$ structure for simple comparison with the $A1$ structure. The long-range order degree (S) is estimated from the XRD data by using the equation [8, 9],

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

where I is the integrated intensity, f is the atomic scattering factor, 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 (001) superlattice and (002) fundamental reflections.

3 Results and discussion

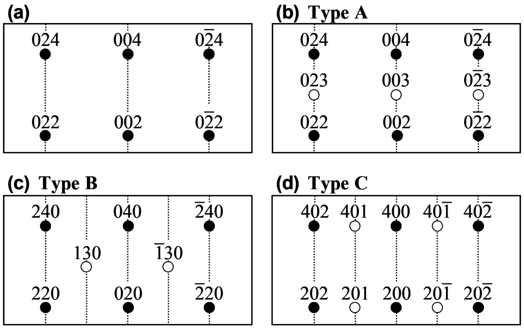

Fig. 2. RHEED spot maps corresponding to (a) $A1(001)$, (b) $L1_0(001)$, (c) $L1_0(010)$, and (d) $L1_0(100)$ surfaces. The incident electron beam is parallel to [(a), (b)] [100], (c) [001], or (d) [010]. The filled and the open circles respectively correspond to the fundamental and the superlattice reflection.

Figures 1(a-1) and (a-2) show the RHEED patterns observed for an FePd film deposited at RT and the film annealed at 600 °C after deposition at RT, respectively. Clear diffraction patterns consisting of only streaks are observed for both films. The patterns correspond to (001) texture of $A1$ or $L1_0$ structure, as shown in the RHEED spot maps of figure 2. When streak patterns are observed, it is difficult to distinguish the diffraction patterns between $A1(001)$ and $L1_0(001)$ textures, because these patterns seem very similar. In order to determine the crystal structure of these films, out-of-plane and in-plane XRD analyses were carried out. Figures 1(b-1), (c-1), (b-2), and (c-2) show the XRD spectra measured for FePd films before and after annealing. For the film before annealing as shown in figures 1(b-1) and (c-1), only the fundamental reflections of FePd(002) and FePd(200) are respectively observed in the out-of-plane and the in-plane XRDs, indicating that the film is with $A1$ structure. For the annealed film, FePd(001) superlattice reflection is recognized in addition to FePd(002) fundamental reflection in the out-of-plane spectrum as shown in figure 1(b-2), whereas FePd(001) superlattice reflection is absent in the in-plane spectrum as shown in figure 1(c-2). The result shows that the annealed film does not involve $L1_0(010)$ and $L1_0(100)$ crystals whose c-axes are in-plane but consists of $L1_0(001)$ crystal with the c-axis perpendicular to the substrate surface. The crystallographic orientation relationships of FePd films before and after annealing with respect to MgO substrate are determined by RHEED and XRD as

$$A1\text{-FePd}(001)[100] \parallel \text{MgO}(001)[100],$$

$$L1_0\text{-FePd}(001)[100]_{\text{Type A}} \parallel \text{MgO}(001)[100].$$

Although $A1$-FePd(001) crystal is expected to transform into $L1_0$-FePd crystals with three kinds of orientations, (001), (010), and (100), by taking into account the crystallographic symmetries of $A1$ and $L1_0$ structures, the transformation is taking place mainly along the $A1$-FePd[001]. The reason seems to be related with the lattice strain in $A1$-FePd(001) film. The in-plane and the out-of-plane lattice constants, a and c, of FePd film before annealing are estimated by XRD to be 0.3848 and 0.3764 nm, respectively. The c value is 2.2% smaller than the a value. In the ordered $L1_0$ crystal, the lattice parameter of c is 4.4% smaller than that of a. Therefore, the c-axis of $L1_0$ structure is considered to have favored to be perpendicular to the substrate surface during the transformation from $A1$ disordered structure to the $L1_0$ ordered phase. The transformation seems to be influenced by the strain within the film of $A1$ disordered structure. The S value of annealed film is estimated from the out-of-plane XRD data to be 0.63. Figures 1(d-1) and (d-2) show

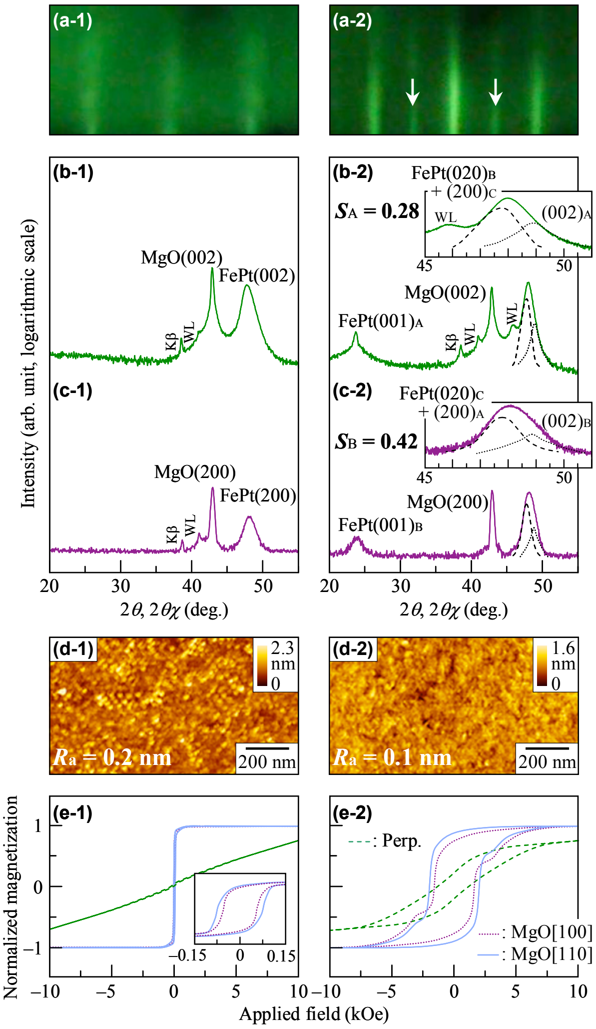

Fig. 3. (a) RHEED patterns, (b) out-of-plane and (c) in-plane XRD spectra, (d) AFM images, and (e) magnetization curves (a-1, b-1, c-1, d-1, e-1) of an FePt film deposition at RT and (a-2, b-2, c-2, d-2, e-2) of the film annealed at 600 °C after deposition at RT. The arrows in (a-2) show the superlattice reflections from $L1_0(010)$ and $L1_0(100)$ crystals. The incident electron beam of RHEED and the scattering vector of in-plane XRD are parallel to MgO[100].

the AFM images. The R_a value is 0.3 nm for both films before and after annealing. It is clearly shown that the film before annealing has very flat surface and the flat surface is kept after annealing. An $L1_0$ ordered FePd film with very flat surface is realized by using the two-step method. Figures 1(e-1) and (e-2) show the magnetization curves. The film before annealing shows an in-plane magnetic anisotropy and the in-plane easy magnetization direction is observed along the MgO[011] (|| $A1$-FePd[011]). The magnetization property is considered to be reflecting the magnetocrystalline anisotropy of $A1$-FePd crystal and the demagnetization field, since $A1$-FePd crystal has the easy magnetization axes along <111>. A perpendicular magnetic anisotropy is obtained for the annealed film which includes the $L1_0(001)$ ordered phase.

Figures 3(a-1) and (a-2) show the RHEED patterns observed for an FePt film deposited at RT and the film annealed at 600 °C after deposition at RT, respectively. Clear diffraction patterns are observed for both films. The RHEED pattern observed for FePt film before annealing corresponds to (001) texture of $A1$ or $L1_0$ structure, similar to the case of FePd film. On the contrary, for the annealed FePt film, superlattice reflections from $L1_0(010)$ and $L1_0(100)$ crystals are recognized, as shown by the arrows in figure 3(a-2). The annealed film involves $L1_0(010)$ and $L1_0(100)$ crystals. Figures 3(b) and (c) show the XRD spectra. For the film before annealing, only fundamental reflections of FePt(002) and FePt(200) are observed, suggesting that the film has $A1$ structure. For the annealed film, FePt(001) superlattice reflections are observed in both the out-of-plane and the in-plane spectra. The annealed film thus includes three variants, $L1_0(001)$, $L1_0(010)$, and $L1_0(100)$. The crystallographic orientation relationships of FePt films before and after annealing with respect to MgO substrate are determined as

$$A1\text{-FePt}(001)[100] \parallel \text{MgO}(001)[100],$$

$$L1_0\text{-FePt}(001)[100]_{\text{Type A}}, (010)[001]_{\text{Type B}}, (100)[010]_{\text{Type C}} \parallel \text{MgO}(001)[100].$$

The $A1$-FePt single-crystal is apparently transforming into three $L1_0$ variants. The lattice constants, a and c, of FePt film before annealing are estimated by XRD to be 0.3810 and 0.3785 nm, respectively. The difference between the c and the a values is 0.7% and it is far smaller than the case of FePd film. Therefore, the transformation is considered to occur along both the perpendicular and the in-plane directions. The S_A value of $L1_0(001)$ crystal is estimated from the out-of-plane XRD reflections to be 0.28, whereas the S_B value of $L1_0(010)$ crystal is calculated from the in-plane reflections to be 0.42. The volume ratio of $L1_0(001)$: $L1_0(010)$: $L1_0(100)$ are estimated from the FePt(002) and the FePt(020) + (200) reflections to be 32: 34: 34. Thus the total S value of FePt is $0.32 \times S_A + 0.24 \times S_B + 0.34 \times S_C = 0.32 \times S_A + 2(0.24 \times S_B) = 0.38$. Figures 3(d-1) and (d-2) show the AFM images. The R_a values of FePt films before and after annealing are 0.2 and 0.1 nm, respectively. An $L1_0$ ordered FePt film with flat surface is obtained by the two-step method, similar to the case of FePd film. Figures 3(e-1) and (e-2) show the magnetization curves. The film before annealing shows an in-plane magnetic anisotropy. A perpendicular anisotropy is enhanced for the annealed FePt film due to that the film includes the $L1_0(001)$ phase. Higher in-plane and out-of-plane coercivities are observed for the annealed film, when compared with the case of film before annealing. This is possibly due to suppression of domain wall motion by the boundaries of $L1_0$ variants.

Figures 4(a-1) and (a-2) show the RHEED patterns observed for CoPt films before and after annealing. Streak patterns corresponding to (001) texture of $A1$ or $L1_0$ structure are observed for both films. Figures 4(b) and (c) show the XRD spectra. For the film before annealing, only fundamental reflections are observed, indicating that the film is a disordered film. For the annealed film, very weak out-of-plane CoPt(001) superlattice reflection is recognized. The result shows that the order degree is very low. The S value is estimated to be 0.16. In the in-plane spectrum, no superlattice

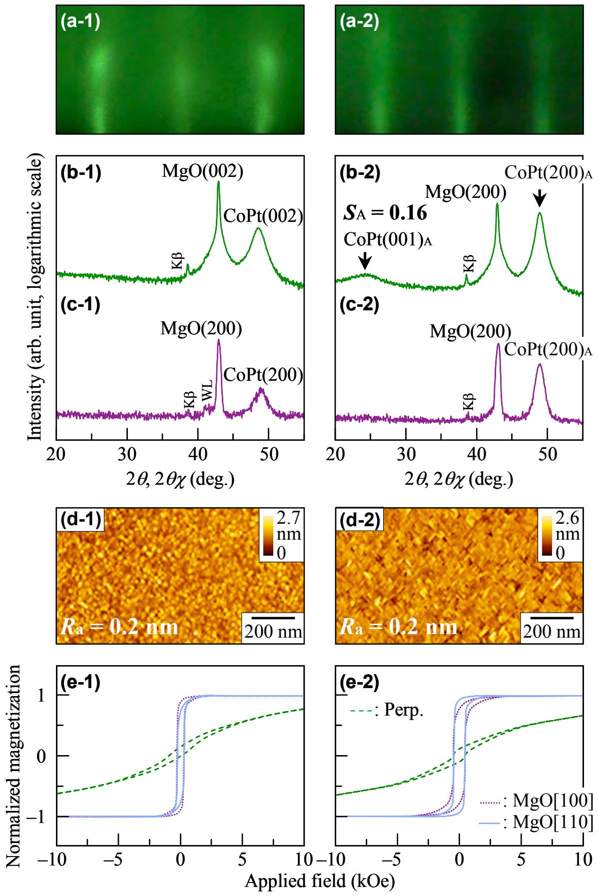

Fig. 4. (a) RHEED patterns, (b) out-of-plane and (c) in-plane XRD spectra, (d) AFM images, and (e) magnetization curves (a-1, b-1, c-1, d-1, e-1) of a CoPt film deposition at RT and (a-2, b-2, c-2, d-2, e-2) of the film annealed at 600 °C after deposition at RT. The incident electron beam of RHEED and the scattering vector of in-plane XRD are parallel to MgO[100].

reflections are recognized. However, there is a possibility of existence of $L1_0$(010) and $L1_0$(100) crystals, because the XRD detection sensitivity may not be strong enough to detect very small volume of $L1_0$(010) and $L1_0$(100) crystals. The crystallographic orientation relationships of CoPt films before and after annealing with respect to MgO substrate are determined as

$$A1\text{-CoPt}(001)[100] \parallel \text{MgO}(001)[100],$$

$$L1_0\text{-CoPt}(001)[100]_{\text{Type A}} \parallel \text{MgO}(001)[100].$$

The lattice constants, a and c, of CoPt film before annealing are estimated by XRD to be 0.3726 and 0.3753 nm, respectively. The c value is in agreement with the a value within a small difference of 0.7%. In this case, the transformation from $A1$ to $L1_0$ structure is expected to take place in almost similar probabilities along the three directions. Figures 4(d-1) and (d-2) show the AFM images. The R_a value is 0.2 nm for both films before and after annealing. Figures 4(e-1) and (e-2) show the magnetization curves. These films show in-plane anisotropies, since the order degree of annealed film is not so high. The in-plane easy magnetization direction is different between the two films. The film before annealing is easily magnetized when the magnetic field is applied along the MgO[001] (|| CoPt[001]). The magnetic anisotropy is considered to be affected by the film strain, similar to the case of $A1$-Co(001) film grown on Cu(001) underlayer [10]. On the other hand, the easy magnetization direction is observed along the MgO[011] (|| CoPt[011]) for the annealed film, similar to the cases of FePd and FePt films.

In the present paper, $L1_0$ ordered FePd, FePt, and CoPt epitaxial films with very flat surfaces are realized by the two-step method. Achievement of higher order degrees, which seems to be possible by employing a higher annealing temperature, is required for practical device applications. It seems also necessary to investigate the effect of film thickness on the surface roughness, since thermal diffusion of atoms are affected by film thickness.

4 Conclusion

$L1_0$ ordered films of three kinds of materials, FePd, FePt, and CoPt, with very flat surfaces are obtained on MgO(001) single-crystal substrates by employing a two-step method; low-temperature deposition at RT followed by high-temperature annealing at 600 °C. The S values of annealed FePd, FePt, and CoPt films are 0.63, 0.38, and 0.16, respectively. The R_a values are less than 0.3 nm for all the films. The magnetic properties are influenced by the order degree and the direction of $L1_0$ ordered phase. The two-step method is considered to be useful for realization of very flat film surface, which is necessary for practical device applications.

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