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Control of c-axis orientation of $L1_0$-FePd in dual-phase-equilibrium FePd/Fe thin films

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This work establishes a method of controlling the c-axis-oriented structure of α-Fe (soft magnetic)/$L1_0$-FePd (hard magnetic) thin films in the dual-phase compositional region in thermal equilibrium. Two types of thin films were prepared; one is a single-layered thin film, and the other is a multilayered film, [FePd($x$ nm)/Fe($5$ nm)]$_n$ ($x$: thickness, $n$: the number of multilayers), both of which are deposited on silica glass substrates. For single-layered films, the ordering process is retarded by phase separation that requires long-range diffusion. In this case, the (111) oriented domains preferentially grow from the film surface, and the c-axis-oriented structure is not obtained. On the contrary, for multilayered films, each FePd layer can undergo ordering without phase separation subject to the constraint of strong biaxial tensile stress resulting from the difference in the thermal contractions between FePd film and the silica glass substrate. Consequently, the $L1_0$ domains with c-axis orientation in the normal direction of the film surface are preferentially formed in the initial amorphous-like structure, eventually leading to the desired c-axis-oriented structure.


I. INTRODUCTION

Ferromagnetic $L1_0$ ordered alloys FePt, CoPt, and FePd, are under extensive investigation for practical use in perpendicular recording media owing to their significantly large uniaxial magnetocrystalline anisotropy ($K \sim 10^6$ J/m$^3$), where the c-axis is the easy axis of magnetization. The large anisotropy can reduce the size of magnetic nanoparticles down to several nanometers without exhibiting superparamagnetic behavior in terms of the $KV/k_BT$ criterion ($V$ is the volume of magnetic particles, $T$ is the temperature, and $k_B$ is the Boltzmann constant). However, for applications to magnetic recording media, we require well-ordered $L1_0$ particles with their c-axis oriented in the normal direction in the thin films.

Recently, a method to synthesize fully ordered FePt nanoparticles has been reported; in this method, high-melting-temperature SiO$_2$ covers the surface of isolated FePt nanoparticles prepared by Sun’s method, and subsequently the SiO$_2$-covered nanoparticles are annealed at sufficiently high temperature for fully ordering reaction to extract their high coercivity. On the other hand, one can choose an epitaxial-like growth method by utilizing an appropriate substrate as a method of controlling the c-axis orientation of $L1_0$ ordered alloys. It has also been claimed that the difference in the thermal expansion coefficients between FePt or CoPt and the substrate plays a significant role in the c-axis orientation of $L1_0$ alloys. In addition, it has been shown that the c-axis of FePt is oriented in the normal direction of the film surface with the aid of B$_2$O$_3$ interlayers, and some of the present authors have tried to elucidate the mechanism of how the c-axis orientation of FePt or CoPt occurs with the B$_2$O$_3$ interlayers. According this work, the biaxial-strain field due to the difference in the thermal contraction plays a significant role in making c-axis aligned in the normal direction of the thin films; this can be regarded as one of the effects of applying an external field.

A well-known problem raised recently is that the switching magnetic field is still too high for write applications due to the high anisotropy field caused by the large $K$. In order to ameliorate this condition, a composite of soft and hard magnetic materials is effective in reducing the switching field. As a model case, we have recently reported the exchange-coupling magnetic property of the α-Fe/$L1_0$-FePt dual-phase region in the Fe–Pd alloy system, and Schied et al. also reported recently the exchange-coupling behavior of this same system. The Fe–Pd alloy system is attractive in that the soft magnetic α-Fe and hard magnetic $L1_0$-FePd coexist in thermal equilibrium. The intention of this work is to establish the method of controlling the c-axis orientation of $L1_0$ ordered domains in the dual-phase region in Fe–Pd alloy system.

II. EXPERIMENTAL

Thin films were deposited on SiO$_2$ glass substrates by using a three-cathode magnetron sputtering apparatus with 2-in.-diameter Fe and Pd targets; rf (13.56 MHz) sputter depositions were performed at room temperature in an Ar (purity: 99.99%) atmosphere of about 0.3 Pa, after the base pressure reached about 10$^{-5}$ Pa. The distance between the...
cathodes and a rotating substrate was set at 200 mm (as long as possible) to maintain surface flatness and compositional homogeneity.

We have prepared following two types of thin films: single-layered thin films and multilayered thin films. In both types of films, the overall thicknesses were about 100 nm. For the single-layered thin films, cosputtering of Fe and Pd were performed simultaneously so as to obtain the objective compositions. For the multilayered thin films, first several nanometers of Fe was sputtered, followed by several nanometers of Fe–50 at. %Pd deposited by co-sputtering, and these procedures were repeated until the total film thickness reached about 100 nm. After the depositions, to attain a fully ordered structure of FePd without oxidation, the thin films were annealed at 550 °C for 6 and 24 h. This is because only ordering processes allowed by short range diffusion occur in the former case, whereas phase separation by long range diffusion is required for the latter case. As a result, the ordering is retarded in the dual-phase compositional region and not in the single-phase compositional region. We also checked that this trend was more prominent for bulk samples of the similar compositions (not presented here). Interestingly, in both cases, the 002 peak position is at a higher angle than that in the powder diffraction file (PDF#02–1440) for L1₀–FePd. From this result, we find that the oriented L1₀ domains have a smaller c-axis than usual, which is due to the stress effect (this is discussed later).

In contrast, the XRD profiles for the multilayered thin films are totally different from the single-layered thin films. As shown in Fig. 2, we cannot see any peaks except the halo peak from the SiO₂ glass in all the as-deposited films, indicating that their structure is an amorphouslike structure or aggregation of tiny small crystals. After annealing at 550 °C for 24 h, however, the diffraction peaks from the L1₀ ordered domains appear. It is here noted that 001 and 002 peaks are very prominent in comparison to the other 110, 111, and 200 peaks, which clearly indicate that the c-axis of L1₀ domains are almost aligned in the normal direction of the film surface in all films. Furthermore, we note that the 200 peak from α-Fe is also strong, while the 110 peak (the strongest line) is almost invisible. That is, it may be suggested that the c-axis

III. RESULTS

Figure 1 shows the XRD profiles obtained for the single-layered thin films of two compositions; (a) Fe–49.4 at. %Pd is in the single L1₀ phase region and (b) Fe–36.3 at. %Pd is in the dual phase (Fe/FePd) region. In the as-deposited films of both compositions, the (111) axis is strongly oriented along the normal direction of the film surface. After annealing at 550 °C for 6 and 24 h, in Fe–49.4 at. %Pd the other peaks (e.g., 200, 002 peaks) appear but in Fe–36.3 at. %Pd the other peaks are invisible after annealing for 6 h but appear after annealing for 24 h. This is because only ordering processes allowed by short range diffusion occur in the former case, whereas phase separation by long range diffusion is required for the latter case. As a result, the ordering is retarded in the dual-phase compositional region and not in the single-phase compositional region. We also checked that this trend was more prominent for bulk samples of the similar compositions (not presented here). Interestingly, in both cases, the 002 peak position is at a higher angle than that in the powder diffraction file (PDF#02–1440) for L1₀–FePd. From this result, we find that the oriented L1₀ domains have a smaller c-axis than usual, which is due to the stress effect (this is discussed later).
of \(L1_0\) domains are cooperatively/simultaneously aligned with the 200 axis of \(\alpha\)-Fe. Figure 3(a) shows the FE-SEM microstructure and composition mappings of Fe and Pd for each multilayered thin film and Fig. 3(b) shows the cross-section TEM micrograph and composition mappings of [FePd(2 nm)/Fe(5 nm)]\textsubscript{14}/SiO\textsubscript{2} film. The denotations, “FePd 5 nm,” “FePd 2 nm,” etc., mean [FePd(5 nm)/Fe(5 nm)]\textsubscript{10}, [FePd(2 nm)/Fe(5 nm)]\textsubscript{14}, etc.

Figure 4 shows the magnetization-field (M-H) curves obtained for single-layered thin films (upper two figures) and multilayered thin films (lower two figures). The insets magnify the curves around the origin. The symbols \(\parallel\) and \(\perp\) mean, respectively, “in-plane” and “out-of-plane” directions of applying the magnetic field during the M-H curve measurement.

FIG. 3. (Color online) (a) FE-SEM microstructure and composition mappings of Fe and Pd for each multilayered thin film and (b) the cross-section TEM micrograph and composition mappings of [FePd(2 nm)/Fe(5 nm)]\textsubscript{14}/SiO\textsubscript{2} film. The denotations, “FePd 5 nm,” “FePd 2 nm,” etc., mean [FePd(5 nm)/Fe(5 nm)]\textsubscript{10}, [FePd(2 nm)/Fe(5 nm)]\textsubscript{14}, etc.

FIG. 4. (Color online) The magnetization-field (M-H) curves obtained for single-layered thin films (upper two figures) and multilayered thin films (lower two figures). The insets magnify the curves around the origin. The symbols \(\parallel\) and \(\perp\) mean, respectively, “in-plane” and “out-of-plane” directions of applying the magnetic field during the M-H curve measurement.

IV. DISCUSSION

Figure 5(a) summarizes the degree of the 001 orientation, which was roughly evaluated by the Lotgering method.\textsuperscript{23} According to the method, the degree of orientation \(P\) associated with the [001] direction is defined as

\[
F_{001} = (p - p_0)/(1 - p_0),
\]

where \(p\) is the value of the XRD intensity ratio, given by \(\Sigma l(001)/\Sigma l(hk)\), and \(p_0\) is the value for the perfectly random orientation. This figure also shows the additional results for the other single-layered thin films; Fe–54.2 at. %Pd and Fe–6.0 at. %Pd. In the single-layered thin films, the degrees of 001 orientation, \(F_{001}\), are around 0 (except for Fe–6.0 at. %Pd), whereas \(F_{001}\) in each multilayered thin film is quite high (about 0.8). Thus, the c-axis orientation can

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be markedly observed in the multilayered thin films, although this is not the case for the single-layered thin films.

We now consider why such high degrees of the c-axis orientation are realized in the multilayered thin films. Figure 5(b) show the lattice parameters, $a$ and $c$-axes, and the axial ratio $c/a$ of the $L1_0$ FePd domains embedded in the films. The values of the bulk materials are also shown for comparison. As found from this figure, the $a$-axis lattice parameter is not so different from the value of the bulk sample but the $c$-axis lattice parameter is much lower than the bulk value; resulting, the axial ratio $c/a$ is also very low. These values are evaluated by the 200 peak for $a$-axis and the 002 and 001 peaks for $c$-axis. Since we employed $\theta$-2$\theta$ method in XRD, the crystallographic planes of the crystallites associated with the diffraction are always parallel to the film surface. Therefore, the $c$ parameter evaluated by this method is associated with the crystallites whose $c$-axes are oriented in the normal direction, and the $a$-axis of the $L1_0$ domains oriented in the normal direction is considerably contracted in all the films.

This lowering of the $c$-axis or $c/a$ can be understood by the internal (residual) stress that would be caused by the difference in the thermal contraction between FePd–Fe and a SiO$_2$ glass substrate. During sputter-deposition, the substrate and deposited FePd–Fe are both at high temperature but after deposition they are cooled down to room temperature. In this cooling process, the FePd–Fe alloy is considered to shrink more than the SiO$_2$ glass substrate because the thermal expansion coefficient silica glass is quite low (of the order $10^{-6}$ /K). Thus, the FePd/Fe films are subjected to biaxial tensile stress because they are under a plane-stress condition.

Based on our previous paper, we estimate the magnitude of such a biaxial tensile stress that is applied to the FePd domains embedded in the thin film. Here the experimental axial ratio is expressed as $(c/a)_{\text{exp}}$, and the tetragonal distortion in a stress-free “equilibrium” state (which is measured from the mother fcc-disordered phase) is denoted as $e_{\text{eq}}^c = (1/3)[1-(c/a)_{\text{eq}}]$, where $(c/a)_{\text{eq}}$ is the axial ratio in the stress-free state. Then, the tetragonal distortion is estimated to be $e_{\text{eq}}^c = 0.0123$ by using the axial ratio, $(c/a)_{\text{eq}} = 0.963$, of the bulk sample in Fig. 5(b). Under the plane-stress condition (i.e., $\sigma_3 = 0$), the elastic strain for the in-plane directions $\epsilon_i$ is expressed as

$$\epsilon_i = 1 - 2e_{\text{eq}}^c - (1 + e_{\text{eq}}^c)(c/a)_{\text{exp}}$$

and the “in-plane” stresses $\sigma_1$ and $\sigma_2$ are given by

$$\sigma_1 = \sigma_2 = \frac{c_{11}^2 + c_{11}c_{12} - 2c_{12}^2}{c_{11}}\epsilon_i,$$

where the subscripts 1, 2, and 3 are assigned for the in-plane directions [Eqs. (1) and (2)] and out-of-plane direction [Eq. (3)] of a thin film, and $c_{ij}$ denote the elastic constants of FePd. Tentatively, we assume that the elastic constants of ordered and disordered FePd are equal to the single-crystal elastic constants of an fcc disordered FePd. Since $(c/a)_{\text{exp}}$ is approximately 0.951 as shown in Fig. 5(b), the in-plane stresses are estimated to be $\sigma_1 = \sigma_2 \approx 700$ [MPa]. Thus, the experimental low $c/a$ ratio indicates that considerably large tensile stresses are being applied to the FePd domains in the films.

Based on the biaxial tensile stress discussed above, we address the $c$-axis orientation mechanism in the FePd/Fe films. Figure 6 illustrates the ordering and phase-separation process under the biaxial tensile stress applied to the film for...
both single and multilayered thin films. In the multilayered
thin film case, long range diffusion is not required because
each FePd layer satisfies the compositional requirement (for
two phases are originally separated in the as-deposited stage
for the multilayered films) so that it can easily undergo only
ordering. In contrast, single-layered thin films need long
range diffusion for phase separation to occur. For multilay-
ered thin films under the biaxial tensile stress, c-axis-oriented
domains are preferentially formed in each layer, except in the
vicinity of the surface (because the (111)-oriented domains
would be favorable there in terms of reducing the surface
energy and releasing the elastic stress). Thus, c-axis-oriented
FePd domains are likely to be formed or chosen from the
tiny small crystals owing to the biaxial tensile stress in the
multilayered thin film. On the contrary, this situation is thoro-
guously different for single-layered thin films. As mentioned
above, this system (i.e., of Fe-rich composition in the Fe–Pd
alloy system) requires phase separation with long range
diffusion, in addition to the ordering process. In terms of dif-
fusion, long range diffusion, and therefore ordering and phase
separation, can take place relatively easily in the vicinity of
the surface, whereas their kinetics would be retarded inside
the film. Near the surface the (111)-oriented ordered do-

V. CONCLUSIONS

In conclusion, we have devoted this work to the investiga-
tion of the c-axis-oriented structure formation of FePd/Fe
thin film in the dual phase region in thermal equilibrium, and
have reached several conclusions. First, in the dual phase
region, phase separation with long range diffusion is required
in addition to the ordering kinetics to attain thermal equilib-
rium during annealing. Second, in the multilayered thin
films, the ordering in each layer easily occurs without long
range diffusion under the constraint of a strong biaxial ten-
sile stress. Consequently, the c-axis-oriented FePd domains
are preferentially formed or chosen from many tiny crystals,
and they eventually become dominant. On the contrary, in
the single-layered thin films, the ordering process itself is
retarded by the indispensable phase separation accompanied
by long-range diffusion. In this case, the (111)-oriented
domains tend to grow preferentially near the surface in terms
of reducing the surface energy and releasing the elastic energy.
Consequently, the c-axis-oriented structure cannot be at-
tained for the single-layered thin films. Finally, to attain con-
trol of the c-axis orientation in L10 ordered alloys, the re-
sidual biaxial tensile stress must be effectively exploited, the
(111)-oriented crystals should be reduced as low as possible
in the as-deposited film, and the ordering process must be
prohibited from occurring at the surface of the film.

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FIG. 7. (Color online) XRD profiles obtained after annealing the
two films, FePd(10 nm)/Fe(5 nm)/SiO2 and
Fe(20 nm)/FePd(10 nm)/Fe(5 nm)/SiO2, at 550 °C for 24 h.