Magnetostriction Behaviors of Fe\textsubscript{100-x}Co\textsubscript{x} Alloy Epitaxial Thin Films under Rotating Magnetic Field

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Fe\textsubscript{100-x}Co\textsubscript{x} (x = 0, 30, 50 at. %) alloy thin films are prepared on MgO substrates of (001), (110), and (111) orientations by ultra-high vacuum magnetron sputtering. The influences of film orientation and composition on the magnetic anisotropy and the magnetostriction are investigated. Fe\textsubscript{100-x}Co\textsubscript{x}(001) single-crystal and (211) bi-crystal films are respectively obtained on MgO(001) and (110) substrates. Fe\textsubscript{100-x}Co\textsubscript{x}(110) films are epitaxially grown on MgO(111) substrates with two types of variants with the crystallographic orientation relationships similar to Nishiyama-Wasserman and Kurdjumov-Sachs. The (001) single-crystal and the (211) bi-crystal films, respectively, show four- and two-fold symmetric in-plane magnetic anisotropies, which are reflecting the magnetcocrystalline anisotropy of Fe\textsubscript{100-x}Co\textsubscript{x} crystal with the easy magnetization axes parallel to <100> or <111>. On the contrary, isotropic in-plane magnetization properties are observed for the (110) films due to an influence of the variant structure. The magnetostriction is measured under rotating magnetic field by using a cantilever method. As the Co content increases from 0 to 50 at. %, the magnetostriction coefficients, \(\lambda\textsubscript{100}\) and \(\lambda\textsubscript{111}\), respectively increase from +10\(^{-4}\) to +10\(^{-3}\) and from -10\(^{-3}\) to +10\(^{-4}\) for both Fe\textsubscript{100-x}Co\textsubscript{x}(001) single-crystal and (211) bi-crystal films. Large \(\lambda\textsubscript{100}\) values are also indicated for the Fe\textsubscript{100-x}Co\textsubscript{x}(110) epitaxial films (x = 30, 50). The present study shows that it is possible to obtain large magnetostriction of 10\(^{-4}\) by control of the film orientation and composition.

**Key words:** Fe-Co alloy, epitaxial thin film, magnetostriction, rotating magnetic field

1. Introduction

Magnetic thin films with large magnetostriction coefficients have been studied for microelectromechanical-system applications such as actuators, sensors, and vibration energy harvesting devices\textsuperscript{1-3}. \(RF_{2}\) (\(R: \text{Tb, Sm, etc.}\)) alloys show giant magnetostriction coefficients\textsuperscript{4} of 10\(^{-3}\). However, high external magnetic fields are required to show large magnetostriction, since they have high magnetic anisotropies. Furthermore, rare-earth free materials are desirable from the viewpoints of cost and natural resource.

Fe-Co alloys with bcc structure are typical soft magnetic materials and have recently attracted much attention as one of magnetostrictive materials, since they show large magnetostriction coefficients\textsuperscript{5-10} of 10\(^{-4}\). The magnetostriction behavior varies depending on the crystallographic orientation. Therefore in order to investigate the basic magnetostriction properties, it is useful to prepare epitaxial thin films, since the crystallographic orientation can be controlled by the substrate orientation. Fe-Co epitaxial films have been prepared on single-crystal substrates of GaAs\textsuperscript{11-16}, MgO\textsuperscript{17-23}, MgAl\textsubscript{2}O\textsubscript{4}\textsuperscript{24-26}, SrTiO\textsubscript{3}\textsuperscript{27-29}, and Al\textsubscript{2}O\textsubscript{3}\textsuperscript{30}. However, the magnetostriction has not been investigated by employing Fe-Co epitaxial films, though there exist reports on the magnetostriction of polycrystalline films\textsuperscript{31-51}. In the present study, Fe\textsubscript{100-x}Co\textsubscript{x} (x = 0–50 at. %) films are prepared on MgO substrates of (001), (110), and (111) orientations. The influences of film orientation and composition on the magnetization and the magnetostriction properties are systematically investigated.

2. Experimental Procedure

An ultra-high vacuum system consisting of two chambers equipped with radio-frequency (RF) magnetron sputter deposition and reflection high-energy electron diffraction (RHEED) facilities was employed. The base pressure of deposition chamber was lower than 4 \times 10^{-7} Pa. MgO(001), MgO(110), and Al\textsubscript{2}O\textsubscript{3}(0001) single-crystal substrates were used. Before film formation, substrates were heated at 600 °C in the deposition chamber to obtain clean surfaces, which were confirmed by RHEED (not shown here). MgO and Fe\textsubscript{100-x}Co\textsubscript{x} alloy (x = 0, 30, 50 at. %) targets of 3 inch diameter were employed. The distance between target and substrate and the Ar gas pressure were respectively fixed at 150 mm and 0.67 Pa. The RF powers for MgO, Fe, Fe\textsubscript{100-x}Co\textsubscript{x}, and Fe\textsubscript{90}Co\textsubscript{10} targets were respectively adjusted to be 200, 50, 51, and 52 W. Under these conditions, the deposition rate was 0.015 nm/s for MgO, whereas it was 0.020 nm/s for the other materials. The substrate temperature during sputter deposition was kept constant at 300 °C.

Fe\textsubscript{100-x}Co\textsubscript{x} films were formed on MgO(001) and MgO(110) substrates and MgO(111) underlayers hetero-epitaxially grown on Al\textsubscript{2}O\textsubscript{3}(0001) substrates. The crystallographic orientation relationship between MgO underlayer and Al\textsubscript{2}O\textsubscript{3} substrate was determined by RHEED as MgO(111)[110] and (111)[110] \parallel Al\textsubscript{2}O\textsubscript{3}(0001)[\text{1100}]. The MgO underlayer consisted of two (111) variants whose orientations were rotated around the film normal by 180° each other. The surface atomic arrangements of the two variants are the same. Therefore, only the crystallographic orientation of MgO(111)[110] \parallel Al\textsubscript{2}O\textsubscript{3}(0001)[\text{1100}] is used below. The thicknesses of MgO(001), MgO(110), and MgO(111)/Al\textsubscript{2}O\textsubscript{3}(0001) substrates were respectively 0.30, 0.30, and 0.43 mm, while that of Fe\textsubscript{100-x}Co\textsubscript{x} film was fixed at 100 nm.

The crystallographic orientation relationship between film and substrate was determined by RHEED. The resulting film
structure was investigated by 2θ/ω-scan out-of-plane and 2θ/φ-scan in-plane X-ray diffractions (XRDs) with Cu-Kα radiation (wave length: 0.15418 nm). The magnetization curves were measured by vibrating sample magnetometry.

The magnetostriction was observed by using a cantilever method under a rotating magnetic field of 1.2 kOe. The bending was measured by using a laser displacement meter fixed on a vibration isolating table. The details of our measurement system are reported in our previous paper38). The relative length change, Δl/l, was calculated from the following formula,

\[
\frac{\Delta l}{l} = \frac{\Delta S \cdot t^2 \cdot E \cdot (1 + \nu)}{3 \cdot L^2 \cdot t \cdot E_0 \cdot (1 - \nu)},
\]

where ΔS was the measured bending, L was the distance between laser beam points (12.5 mm), t was the thickness, E was the Young’s modulus, ν was the Poisson’s ratio, and the subscripts of f and s respectively referred to film and substrate.

The E and the ν values of single crystal vary depending on the crystallographic direction, though E and ν are usually defined in an isotropic elastic body. In the present study, E and ν are respectively defined as \( \sigma_{(001)} \) and \( \epsilon_{(001)} \), where \( \sigma \) is the uniaxial stress applied along \( [g_1, g_2, g_3] \), \( \epsilon \) is the strain occurred along \( [g_1, g_2, g_3] \), and \( \epsilon_f \) is the strain occurred along the direction perpendicular to \( [g_1, g_2, g_3] \) in the film plane (\( [d_1, d_2, d_3] \) \( \perp [g_1, g_2, g_3] \) \( \perp \) out-of-plane direction). Based on the definitions, E and ν of cubic single crystal are respectively expressed39,40 as

\[
1 = \frac{C_{11} + C_{12}}{C_{11} - C_{12}} \left( \frac{C_{11} + 2C_{12}}{C_{11} - 2C_{12}} \right) \left( \chi_1^2 \chi_2^2 + \chi_2^2 \chi_3^2 + \chi_3^2 \chi_1^2 \right),
\]

\[
\nu = -\frac{1}{C_{11} - C_{12}} \left( \frac{C_{11} + 2C_{12}}{C_{11} - 2C_{12}} \right) \left( \chi_1^2 \chi_2^2 + \chi_2^2 \chi_3^2 + \chi_3^2 \chi_1^2 \right),
\]

where \( C_{11}, C_{12}, C_{44} \) were the elastic stiffness values and \( \chi_1, \chi_2, \chi_3 \) are the cosines of angles of \( [g_1, g_2, g_3] \) and \( [d_1, d_2, d_3] \) with respect to the three crystallographic axes \( (a, b, c) \). Tables 1 and 2 summarize the E and the ν values of MgO and Fe single crystals calculated with the reported values of \( C_{11}, C_{12}, C_{44} \) for MgO and Fe single crystals. Figures 1(a) and (b) respectively show the in-plane E and ν distributions of Fe(110) single-crystal film. The E and the ν values of Fe(110) film with multiple variants are respectively regarded as the averages of Fig. 1(a) \( (E = 217 \text{ GPa}) \) and Fig. 1(b) \( (\nu = 0.34) \). In the present study, the E and the ν values of Fe single crystal were used in the calculation of Δl/l for Fe-Co alloy films, since the elastic stiffness values of Fe-Co alloys were unknown. For hexagonal Al2O3 crystal, the reported values41 of E = 407.5±62.5 GPa (345–470 GPa) and ν = 0.285±0.015 (0.27–0.30) were used.

3. Results and Discussion

3.1 Film growth and structure

Figures 2(a)–(c) show the RHEED patterns observed for Fe100-Co films with different compositions formed on MgO(001), MgO(110), and MgO(111)/Al2O3(0001) substrates, respectively. Figures 2(d)–(f) illustrate the diffraction patterns simulated for bcc(001) single-crystal, bcc(211) bi-crystal, and bcc(110) crystal with Nishiyama-Wasserman (NW)38,39 and Kurdumov-Sachs (KS)40 variants, respectively. The details of the simulations have been shown in our previous papers20–23). The observed patterns of Figs. 2(a)–(c) are respectively in agreement with the simulated patterns of Figs. 2(d)–(f). Therefore, Fe100-Co(001) single-crystal, (211) bi-crystal, and (110) crystal films are respectively epitaxially grown on MgO(001), MgO(110), and MgO(111)/Al2O3(0001) substrates for all the compositions of x = 0–50 at. %. The crystallographic orientation relationships are determined as

\[
\begin{align*}
\text{Fe}_{100} \cdot \text{Co}_{(001)[110]} & \parallel \text{MgO(001)[100]}, \\
\text{Fe}_{100} \cdot \text{Co}_{(211)[011]} & \parallel \text{MgO(110)[001]}, \quad \text{(type A)} \\
\text{Fe}_{100} \cdot \text{Co}_{(211)[011]} & \parallel \text{MgO(110)[001]}, \quad \text{(type B)}
\end{align*}
\]
MgO[001], or MgO[110], and MgO[111]/Al₂O₃[0001] substrates. Schematic diagrams of RHEED patterns simulated for Fe₇₀Co₃₀ single-crystal, Fe₅₀Co₅₀ bi-crystal, and Fe-Co₆₀ multi-crystal with NW and KS variants. The incident electron beam is parallel to MgO[001], MgO[001], and MgO[111]/Al₂O₃[0001] substrates, respectively. Figures 3(a-2)–(c-2) show the in-plane XRD patterns of the epitaxial films formed on MgO[001], MgO[110], and MgO[111]/Al₂O₃[0001] substrates measured by making the scattering vector parallel to MgO[110], MgO[001], and MgO[111]/Al₂O₃[0001], respectively. bcc(200) reflection is observed in the patterns of Fe₇₀Co₃₀[001] single-crystal films [Fig. 3(a-2)], bcc(011) reflection from the A-type variant and bcc(011) reflection from the B-type variant.

Figures 3(a-1)–(c-1) show the out-of-plane XRD patterns of the Fe₇₀Co₃₀ epitaxial films with different orientations. bcc(002), bcc(211), and bcc(110) reflections are observed for the films formed on MgO[001], MgO[110], and MgO[111]/Al₂O₃[0001] substrates, respectively. Figures 3(a-2)–(c-2) show the in-plane XRD patterns of the epitaxial films formed on MgO[001], Fe-Co₆₀, and MgO[111]/Al₂O₃[0001] substrates measured by making the scattering vector parallel to MgO[110], MgO[001], and MgO[111]/Al₂O₃[0001], respectively.

Fig. 2 (a)–(c) RHEED patterns observed for (a-1)–(c-1) Fe, (a-2)–(c-2) Fe₇₀Co₃₀, and (a-3)–(c-3) Fe₅₀Co₅₀ films formed on (a) MgO[001], (b) MgO[110], and (c) MgO[111]/Al₂O₃[0001] substrates. (d)–(f) Schematic diagrams of RHEED patterns simulated for (d) bcc(001) single-crystal, (e) bcc(211) bi-crystal, and (f) bcc(110) multi-crystal with NW and KS variants. The incident electron beam is parallel to (a) MgO[010], (b) MgO[001], (c) MgO[110], (d) bcc[110], (e) bcc[011]∥[011], or (f) bcc[001]∥[111].

Fe₇₀Co₃₀ films formed on (a) MgO[001], (b) MgO[110], and (c) MgO[111]/Al₂O₃[0001] substrates. The scattering vector of in-plane XRD is parallel to (a-2) MgO[110], (b-2) MgO[110], or (c-2) MgO[110]∥Al₂O₃[0001]. The intensity is shown in logarithmic scale. (d)–(f) Compositional dependences of out-of-plane and in-plane lattice spacings of Fe₇₀Co₃₀ films formed on (d) MgO[001], (e) MgO[110], and (f) MgO[111]/Al₂O₃[0001] substrates.

Fig. 3 (a-1)–(c-1) Out-of-plane and (a-2)–(c-2) in-plane XRD patterns of Fe, Fe₇₀Co₃₀, and Fe₅₀Co₅₀ films formed on (a) MgO[001], (b) MgO[110], and (c) MgO[111]/Al₂O₃[0001] substrates. The scattering vector of in-plane XRD is parallel to (a-2) MgO[110], (b-2) MgO[110], or (c-2) MgO[110]∥Al₂O₃[0001]. The intensity is shown in logarithmic scale. (d)–(f) Compositional dependences of out-of-plane and in-plane lattice spacings of Fe₇₀Co₃₀ films formed on (d) MgO[001], (e) MgO[110], and (f) MgO[111]/Al₂O₃[0001] substrates.
Fig. 4 (a-1)–(c-1) In-plane magnetization curves and (a-2)–(c-2) $M_s/M_i$ distributions measured for (a) Fe, (b) Fe$_{30}$Co$_{70}$, and (c) Fe$_{20}$Co$_{80}$(001) single-crystal films grown on MgO(001) substrates. The applied magnetic field directions are shown by using the crystallographic directions of Fe$_{100}$-Co$_{50}$ film.

Fig. 5 (a-1)–(c-1) In-plane magnetization curves and (a-2)–(c-2) $M_s/M_i$ and (a-3)–(c-3) $H_i$ distributions measured for (a) Fe, (b) Fe$_{30}$Co$_{70}$, and (c) Fe$_{20}$Co$_{80}$(211) bi-crystal films grown on MgO(110) substrates. The applied magnetic field directions are shown by using the crystallographic directions of two Fe$_{100}$-Co$_{50}$(211) variants. The films show four-fold symmetric in-plane anisotropies. The easy magnetization directions are observed along [100], [010], [100], and [010] (blue solid lines in Figs. 4(a-2) and (b-2)), which is reflecting the magnetocrystalline anisotropy of Fe$_{100}$-Co crystal with the easy magnetization axes parallel to <100>. Figure 4(c) shows the magnetization property of the Fe$_{20}$Co$_{80}$(001) single-crystal film. Although a four-fold symmetry in in-plane anisotropy is recognized, the easy magnetization directions are parallel to [110], [110], [110], and [110] (orange dotted lines in Fig. 4(c-2)), which are different from those observed for Fe and Fe$_{30}$Co$_{70}$ films. It is known that the easy magnetization axes of bulk Fe$_{100}$-Co$_{50}$ crystal vary from <100> to <111> when the Co content increases beyond about 40 at. %$^6$. Therefore, the in-plane magnetic anisotropy observed for Fe$_{20}$Co$_{80}$ film seems to be reflecting the magnetocrystalline anisotropy of Fe$_{100}$-Co$_{50}$ crystal with the easy axes parallel to <111> and the demagnetization field. The magnetic anisotropy of Fe$_{100}$-Co$_{50}$(001) film varies depending on the composition, similar to the case of bulk crystal.

3.2 Magnetic anisotropy

Figures 4(a-1) and (b-1) show two typical examples of in-plane magnetization curves measured for the Fe and the Fe$_{30}$Co$_{70}$(001) single-crystal films. The distributions of normalized remnant magnetization, $M_s/M_i$, are summarized in Figs. 4(a-2) and (b-2). The applied magnetic field directions are shown by using the crystallographic directions of Fe$_{100}$-Co$_{50}$ film. The films show four-fold symmetric in-plane magnetic anisotropies. The easy magnetization directions are observed along [100], [010], [100], and [010] (blue solid lines in Figs. 4(a-2) and (b-2)), which is reflecting the magnetocrystalline anisotropy of Fe$_{100}$-Co crystal with the easy magnetization axes parallel to <100>. Figure 4(c) shows the magnetization property of the Fe$_{20}$Co$_{80}$(001) single-crystal film. Although a four-fold symmetry in in-plane anisotropy is recognized, the easy magnetization directions are parallel to [110], [110], [110], and [110] (orange dotted lines in Fig. 4(c-2)), which are different from those observed for Fe and Fe$_{30}$Co$_{70}$ films. It is known that the easy magnetization axes of bulk Fe$_{100}$-Co$_{50}$ crystal vary from <100> to <111> when the Co content increases beyond about 40 at. %$^6$. Therefore, the in-plane magnetic anisotropy observed for Fe$_{20}$Co$_{80}$ film seems to be reflecting the magnetocrystalline anisotropy of Fe$_{100}$-Co$_{50}$ crystal with the easy axes parallel to <111> and the demagnetization field. The magnetic anisotropy of Fe$_{100}$-Co$_{50}$(001) film varies depending on the composition, similar to the case of bulk crystal.

Figure 5(a-1) shows the hysteresis curves of the Fe(211) bi-crystal film. The distributions of $M_s/M_i$ and saturation field ($H_i$) are respectively summarized in Figs. 5(a-2) and (a-3). The applied field directions are shown by using the crystallographic directions of two (211) variants. The Fe film shows a two-fold
symmetric in-plane magnetic anisotropy. The easy magnetization directions are observed along [251]A, [215]A, [251]B, and [215]B (pink dotted lines in Figs. 5(a-2) and (a-3)), which are respectively obtained by projecting [010]A, [001]A, [010]B, and [001]B on the (211) surface as shown in Fig. 6. Therefore, the magnetic anisotropy is interpreted to be reflecting the magnetocrystalline anisotropy of Fe100–Co30 crystal with the easy axes parallel to <111> and the demagnetization film, similar to the cases of Fe and Fe2Co0(001) single-crystal films. Figure 5(c) shows the magnetic property of the Fe90Co10(211) film. A two-fold symmetry in in-plane magnetic anisotropy is observed. However, the easy magnetization directions are parallel to [011]A+[011]B and [011]A+[011]B (blue solid lines in Figs. 5(c-2) and (c-3)). It is also noted that the M/s values measured along [111]A+[111]B and [111]A+[111]B are not so high (orange dotted lines in Fig. 5(c-3)), though the M/Ms values are low (orange dotted lines in Fig. 5(c-2)). Therefore, the film is moderately easily magnetized along [111]A+[111]B and [111]A+[111]B. On the contrary, the magnetization curves measured along [251]A, [215]A, [251]B, and [215]B, which are respectively obtained by projecting [010]A, [001]A, [010]B, and [001]B on the film plane, saturate at higher magnetic fields (pink dotted lines in Fig. 5(c-3)). When the Co content increases up to 50 at. %, the easy and the hard magnetization axes are respectively considered to be parallel to <111> and <100>6).

Figure 7 shows the in-plane magnetic properties of the Fe100–Co30(110) epitaxial films with NW and KS variants formed on MgO(111)/Al2O3(0001) substrates. The films show almost isotropic magnetization properties. Nine (110) variants are coexisting in the (110) epitaxial films and the respective magnetic anisotropies are overlapped. Therefore, isotropic magnetization properties are considered to be observed.

3.3 Magnetostriction of (001) single-crystal films

The relative length change, Δl/l, of a cubic crystal caused by magnetostriction6) is shown as

\[
\frac{\Delta l}{l} = \frac{3}{2} \lambda_{100} (a_1^2 \beta_1^2 + a_2^2 \beta_2^2 + a_3^2 \beta_3^2 - \frac{1}{3}) + 3 \lambda_{111} (a_1 a_2 \beta_2 \beta_3 + a_3 a_1 \beta_3 \beta_1 + a_2 a_3 \beta_1 \beta_2),
\]

where \(\lambda_{100}\) and \(\lambda_{111}\) are the magnetostriction coefficients, \((a_1, a_2, a_3)\) and \((\beta_1, \beta_2, \beta_3)\) are respectively the cosines of the angles of magnetization and observation directions with respect to the three crystallographic axes \((a, b, c)\).

When the magnetization rotates in a (001) plane under in-plane rotating magnetic field as shown in Fig. 8(a), the crystallographic direction of magnetization is shown as \((\cos \phi, \sin \phi, 0)\), where \(\phi\) is the angle of magnetization direction with respect to [100]. The \((a_1, a_2, a_3)\) values are thus expressed as

\[
(a_1, a_2, a_3)_{[001]} = (\cos \phi, \sin \phi, 0).
\]

When the observation directions are parallel to [100] and [110], the \((\beta_1, \beta_2, \beta_3)_{[100]}\) and \((\beta_1, \beta_2, \beta_3)_{[110]}\) values are respectively expressed as

\[
(\beta_1, \beta_2, \beta_3)_{[100]} = (1, 0, 0),
\]

\[
(\beta_1, \beta_2, \beta_3)_{[110]} = (0, \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}).
\]

By substituting Eqs. (5)–(7) into (4), the relative length changes measured along [100] and [110] under in-plane rotating...
magnetic field, $\Delta l/\lambda_{100}(\phi)$ and $\Delta l/\lambda_{110}(\phi)$, are given as

$$\frac{\Delta l}{\lambda_{100}}(\phi) = \frac{3}{4} \lambda_{100} \cos 2\phi + \frac{1}{4} \lambda_{111},$$

$$\frac{\Delta l}{\lambda_{110}}(\phi) = \frac{3}{4} \lambda_{111} \sin 2\phi + \frac{1}{4} \lambda_{111},$$

which are respectively cosine and sine waves. It is noted that the phases of $\Delta l/\lambda_{100}(\phi)$ and $\Delta l/\lambda_{110}(\phi)$, respectively, reverse depending on the signs of $\lambda_{100}$ and $\lambda_{111}$, as shown in Figs. 8(b) and (c). Furthermore, the $\lambda_{100}$ and the $\lambda_{111}$ values can be estimated by using the following relations,

$$\lambda_{100} = \frac{4}{3} \left[ \frac{\Delta l}{l_{100}}(\phi = 0) - \frac{\Delta l}{l_{100}}(\phi = 45^\circ) \right],$$

$$\lambda_{111} = \frac{4}{3} \left[ \frac{\Delta l}{l_{110}}(\phi = 45^\circ) - \frac{\Delta l}{l_{110}}(\phi = 90^\circ) \right].$$

Figure 8(d-1) shows the $\Delta l/\lambda_{100}(\phi)$ measured for the Fe$_{100}$Co$_{30}$(001) single-crystal films with different compositions. The phases of observed waves are in agreement with that of calculated wave of Fig. 8(b-1). The $\lambda_{100}$ value is thus positive for all the Fe$_{100}$Co$_{30}$(001) films. Figure 8(d-2) shows the $\Delta l/\lambda_{110}(\phi)$ measured for the Fe$_{100}$Co$_{30}$(001) films. The phases of waves observed for Fe and Fe$_{50}$Co$_{50}$ films agree with that of calculated wave of Refs. 7–8, whereas the phase of wave observed for Fe$_{70}$Co$_{30}$ film is in agreement with that of wave of Fe$_{50}$Co$_{50}$ films, while that is positive for the Fe$_{50}$Co$_{50}$ film.

Figure 8(e) shows the $\lambda_{100}$ and the $\lambda_{111}$ values plotted as a function of Co content. The $\lambda_{100}$ and the $\lambda_{111}$ values increase with increasing the Co content. The Fe$_{50}$Co$_{50}$ film shows a large $\lambda_{100}$ value of $+234 \times 10^{-6}$ and a small $\lambda_{111}$ value of $-5 \times 10^{-6}$. On the contrary, a large $\lambda_{100}$ value of $+274 \times 10^{-6}$ and a moderately large $\lambda_{111}$ value of $+78 \times 10^{-6}$ are observed for the Fe$_{50}$Co$_{50}$ film.

### 3.4 Magnetostriiction of (211) bi-crystal films

When the magnetization rotates in a (211)$_\lambda$ plane as shown in Fig. 9(a), the crystallographic direction of magnetization is shown as $[-\sin\chi/\beta, \cos\chi/\beta, \sin\chi/\beta, -\cos\chi/\sqrt{2}, \sin\chi/\sqrt{2}, -\cos\chi/\sqrt{2}]$, where $\chi$ is the angle of magnetization direction with respect to [011]$_\lambda$ (|| MgO(001)). The $(\alpha_1, \alpha_2, \alpha_3)_{211A}$ values are thus expressed as

$$(\alpha_1, \alpha_2, \alpha_3)_{211A} = (\sin\chi/\sqrt{2}, \cos\chi/\sqrt{2} \sin\chi/\beta, \cos\chi/\sqrt{2} \sin\chi/\beta, \cos\chi/\sqrt{2} \sin\chi/\beta).$$

When the observation directions are parallel to [011]$_\lambda$ and [111]$_\lambda$, the $(\beta_1, \beta_2, \beta_3)_{011A}$ and the $(\beta_1, \beta_2, \beta_3)_{111A}$ values are respectively expressed as...
(β, θ, β1)011A = (0, 1/2, 1/2).

(β, θ, β1)111A = (1/2, 1/2, 1/2).

(13)

By substituting Eqs. (12)–(14) into (4), the Δl/l_{111}A(x) and the Δl/l_{111}B(x) are respectively given as

\[
\frac{\Delta l}{l_{111}A}(x) = \frac{1}{4} \Delta \lambda_{111} \cos 2x + \frac{1}{8} \Delta \lambda_{100} + \frac{1}{4} \Delta \lambda_{111}.
\]

(14)

Since the Fe_{100–x}Co_{x}(211) films consist of two types of variants, A and B, it is necessary to take into account the Δl/l_{111}A(x) and the Δl/l_{111}B(x), which are respectively shown as follows,

\[
\frac{\Delta l}{l_{111}A}(x) = \frac{1}{4} \Delta \lambda_{111} \cos 2x + \frac{1}{8} \Delta \lambda_{100} + \frac{1}{4} \Delta \lambda_{111}.
\]

(15)

(0.7)

Therefore, when the magnetostriction is measured along MgO[001] ( || Fe_{100–x}Co_{x}[01 1]A + [0 1 1]A) and MgO[1 0 0] ( || Fe_{100–x}Co_{x}[1 1 1]A + [1 1 1]A), the averages of relative length changes of types A and B variants, Δl/l_{MgO[001]}(x) and Δl/l_{MgO[1 0 0]}(x), are respectively given as

\[
\Delta l/l_{MgO[001]}(x) = \frac{1}{2} \left[ \frac{\Delta l}{l_{111}A}(x) + \frac{\Delta l}{l_{111}B}(x) \right] = \frac{1}{8} \Delta \lambda_{100} + \frac{5}{8} \Delta \lambda_{111} \cos 2x + \frac{1}{8} \Delta \lambda_{100} + \frac{1}{8} \Delta \lambda_{111},
\]

(16)

\[
\Delta l/l_{MgO[1 0 0]}(x) = \frac{1}{2} \left[ \frac{\Delta l}{l_{111}A}(x) + \frac{\Delta l}{l_{111}B}(x) \right] = -\frac{3}{4} \Delta \lambda_{111} \cos 2x + \frac{1}{4} \Delta \lambda_{111}.
\]

(17)

(18)

which are shown in Figs. 9(b) and (c). Furthermore, the λ_{100} and the λ_{111} values can be estimated by using the following equations,

\[
\lambda_{100} = 8 \left[ \frac{\Delta l}{l_{MgO[001]}}(x = 0º) - \frac{\Delta l}{l_{MgO[001]}}(x = 45º) \right],
\]

(19)

\[
\lambda_{111} = -\frac{4}{3} \left[ \frac{\Delta l}{l_{MgO[1 0 0]}}(x = 90º) - \frac{\Delta l}{l_{MgO[1 0 0]}}(x = 135º) \right].
\]

(20)

(21)
Figure 9(d) shows the $\Delta l_{MgO(001)}(\chi)$ and the $\Delta l_{MgO(110)}(\chi)$ measured for the Fe$_{100-x}$Co$_x$(111) films. Figure 9(e) summarizes the $\lambda_{100}$ and the $\lambda_{111}$ values as a function of Co content in Fe$_{100-x}$Co$_x$(001) single-crystal films. The Fe$_{50}$Co$_{50}$(211) film shows a large $\lambda_{100}$ value of +236±10$^4$ and a moderately large $\lambda_{111}$ value of +97±10$^4$.

3.5 Magnetostriiction of Fe$_{100-x}$Co$_x$(110) epitaxial films with NW and KS variants

When the magnetization rotates in a (110) plane as shown in Fig. 10(a), the crystallographic direction of magnetization is shown as $[\sin \psi/\sqrt{2} \sin \phi/\sqrt{2} \cos \phi]$. Here, $\psi$ is the angle of magnetization direction with respect to (001). The $(\alpha_1, \alpha_2, \alpha_3)$ values are thus expressed as

$(\alpha_1, \alpha_2, \alpha_3) = [\sin \psi/\sqrt{2}, -\sin \phi/\sqrt{2}, \cos \phi]$. (23)

When the angle of in-plane observation direction with respect to [001] is shown as $\phi$, the $(\beta_1, \beta_2, \beta_3)$ values are expressed as

$(\beta_1, \beta_2, \beta_3) = [\sin \phi/\sqrt{2} - \sin \phi/\sqrt{2}, \cos \phi]$. (24)

The $\Delta l_{\sin \phi/\sqrt{2} - \sin \phi/\sqrt{2}, \cos \phi}^i$ is thus given by substituting Eqs. (23) and (24) into (4) as follows,

$$\Delta l = \frac{2}{3} \lambda_{100} \left[ \sin^2 \psi \sin^2 \omega - \cos^2 \psi \cos^2 \omega - \frac{1}{3} \right]$$

$$+ 3 \lambda_{111} \left[ \sin^2 \psi \sin^2 \omega + \cos^2 \psi \cos^2 \omega \right]$$ (25)

In order to characterize the magnetostriiction of an epitaxial film with multi-variant structure, it is necessary to take into account the volume ratio of each variant and the respective relative length changes. However, there are as many as 9 variants in the Fe$_{100-x}$Co$_x$(110) epitaxial films prepared in the present study. Therefore, the in-plane orientation can be regarded as being random and the average of $\Delta l(\psi)$ of each variant is expressed as

$$\Delta l(\psi) = \frac{1}{2\pi} \int_0^{2\pi} \frac{\Delta \psi}{[\sin \phi/\sqrt{2} - \sin \phi/\sqrt{2}, \cos \phi]} d\omega$$

$$= \frac{3}{16} \left[ \lambda_{100} - \lambda_{111} \right] \cos 2\psi + \frac{1}{16} \left[ \lambda_{100} + 3\lambda_{111} \right]$$, (26)

which is shown in Fig. 10(b). Although the relationship of $\lambda_{100} > \lambda_{111}$, $\lambda_{100} = \lambda_{111}$, or $\lambda_{100} < \lambda_{111}$ can be determined by considering the phase of observed wave, the values of $\lambda_{100}$ and $\lambda_{111}$ cannot be estimated in the case of the Fe$_{100-x}$Co$_x$(110) epitaxial film. The $(\lambda_{100} - \lambda_{111})$ value is shown as

$$(\lambda_{100} - \lambda_{111}) = \frac{16}{3} \left[ \Delta l(\psi = 0^\circ) - \Delta l(\psi = 45^\circ) \right]$$, (27)

Figure 10(c) shows the $\Delta l(\psi)$ measured for the Fe$_{100-x}$Co$_x$(110) films. The phase of wave observed for Fe film is in agreement with that of calculated wave of Fig. 10(b-2), whereas the phases of waves measured for Fe$_{70}$Co$_{30}$ and Fe$_{50}$Co$_{50}$ films agree with that of wave of Fig. 10(b-1). The result shows that the $\lambda_{100}$ value is smaller than the $\lambda_{111}$ value for the Fe film, while the $\lambda_{100}$ value is larger than the $\lambda_{111}$ value for the Fe$_{70}$Co$_{30}$ and the Fe$_{50}$Co$_{50}$ films. Figure 10(d) shows the $(\lambda_{100} - \lambda_{111})$ values plotted as a function of Co content. As the Co content increases, the $(\lambda_{100} - \lambda_{111})$ value increases. The Fe$_{70}$Co$_{30}$ and the Fe$_{50}$Co$_{50}$ films show large $\lambda_{100} - \lambda_{111}$ values, indicating that large $\lambda_{100}$ values are obtained.

Large $\lambda_{100}$ values are obtained, even if Fe-Co films are prepared on MgO substrates with different orientations. Therefore, well-defined epitaxial Fe-Co films have potentials to achieve large magnetostriiction.

![Fig. 10](image-url)

Fig. 10 (a) Schematic diagram showing the magnetization and the observation directions with respect to the typical crystallographic directions. (b) $\Delta l(\psi)$ calculated for (110) epitaxial multi-crystal films with (b-1) $\lambda_{100} > \lambda_{111}$ and (b-2) $\lambda_{100} < \lambda_{111}$. (c) $\Delta l(\psi)$ measured for Fe$_{100-x}$Co$_x$(110) epitaxial multi-crystal films formed on MgO(111)/Al$_2$O$_3$(0001) substrates. (d) Compositional dependences of $(\lambda_{100} - \lambda_{111})$. The bulk values in (d) are calculated by using $\lambda_{100}$ and $\lambda_{111}$ values in Refs. 5–8 and 42.
4. Conclusion

Fe\textsubscript{100-x}Co\textsubscript{x} (x = 0–50 at. %) alloy epitaxial films are prepared on MgO substrates with different orientations. The magnetization and the magnetostriction properties are characterized. Fe\textsubscript{100-x}Co\textsubscript{x}(001) single-crystal and (211) bi-crystal films are respectively obtained on MgO(001) and (110) substrates, whereas Fe\textsubscript{100-x}Co\textsubscript{x}(110) films with nine variants are epitaxially grown on MgO(111) substrates. The (001) single-crystal and the (211) bi-crystal films, respectively, show four- and two-fold symmetric in-plane magnetic anisotropies, which are reflecting the magnetocrystalline anisotropy of Fe\textsubscript{100-x}Co\textsubscript{x} crystal with the easy magnetization axes parallel to <100> or <111>. The easy magnetization directions vary depending on the film composition and orientation. On the contrary, isotropic in-plane magnetization properties are observed for the (110) epitaxial films due to an influence of the variant structure. The magnetostriction behavior under rotating magnetic field is studied. As the Co content increases, the λ\textsubscript{100} values are also indicated for both Fe\textsubscript{100-x}Co\textsubscript{x}(001) single-crystal and (211) bi-crystal films. Large λ\textsubscript{111} values are also indicated for the Fe\textsubscript{50}Co\textsubscript{50} and the Fe\textsubscript{60}Co\textsubscript{40} epitaxial films. The present study shows that it is possible to obtain large magnetostrictive of 10\textsuperscript{-4} by control of the film orientation and composition.

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