Magnetostriction Behaviors of Fe$_{100-x}$Co$_x$ Alloy Epitaxial Thin Films under Rotating Magnetic Field

Kana Serizawa$^{1,2}$, Mitsuru Ohtake$^1$, Tetsuroh Kawai$^1$, Masaaki Futamoto$^2$, Fumiyoshi Kirino$^3$, and Nobuyuki Inaba$^4$

$^1$Faculty of Engineering, Yokohama National University, 79-5 Tokiwadai, Hodogaya, Yokohama 240-8501, Japan
$^2$Faculty of Science and Engineering, Chuo University, 1-13-27 Kasuga, Bunkyo-ku, Tokyo 112-8551, Japan
$^3$Graduate School of Fine Arts, Tokyo University of the Arts, 12-8 Ueno-koen, Taito-ku, Tokyo 110-8714, Japan
$^4$Faculty of Engineering, Yamagata University, 4-3-16 Iyonan, Yonezawa, Yamagata 992-8510, Japan

Fe$_{100-x}$Co$_x$ ($x = 0, 30, 50$ 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$_{100-x}$Co$_x$(001) single-crystal and (211) bi-crystal films are respectively obtained on MgO(001) and (110) substrates. Fe$_{100-x}$Co$_x$(110) films are epitaxially grown on MgO(111) substrates with two types of variants, with the crystallographic orientation relationships similar to Nishiya-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 magnetcrysalline anisotropy of Fe$_{100-x}$Co$_x$ 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, $\lambda_{100}$ and $\lambda_{111}$, respectively increase from $+10^{-3}$ to $+10^{-2}$ and from $-10^{-3}$ to $+10^{-5}$ for both Fe$_{100-x}$Co$_x$(001) single-crystal and (211) bi-crystal films. Large $\lambda_{100}$ values are also indicated for the Fe$_{100-x}$Co$_x$(110) epitaxial films ($x = 30, 50$). The present study shows that it is possible to obtain large magnetostriction of $10^{-2}$ 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$^{1-3}$. RE$_2$ (R: Tb, Sm, etc.) alloys show giant magnetostriction coefficients$^9$ 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$^4-10$ 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$^{11-16}$, MgO$^{17-23}$, MgAl$_2$O$_4$$^{22, 23}$, SrTiO$_3$$^{22-24}$, Al$_2$O$_3$$^{25}$, etc. However, the magnetostriction has not been investigated by employing Fe-Co epitaxial films, though there exist reports on the magnetostriction of polycrystalline films$^{8, 25-11}$. In the present study, Fe$_{100-x}$Co$_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$_2$O$_3$(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$_{100-x}$Co$_x$ 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$_{70}$Co$_{30}$, and Fe$_{50}$Co$_{50}$ 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$_{100-x}$Co$_x$ films were formed on MgO(001) and MgO(110) substrates and MgO(111) underlayers hetero-epitaxially grown on Al$_2$O$_3$(0001) substrates. The crystallographic orientation relationship between MgO underlayer and Al$_2$O$_3$ substrate was determined by RHEED as MgO(111)[110] and [111][110] || Al$_2$O$_3$(0001)[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] || Al$_2$O$_3$(0001)[1100] is used below. The thicknesses of MgO(001), MgO(110), and MgO(111)/Al$_2$O$_3$(0001) substrates were respectively 0.30, 0.30, and 0.43 mm, while that of Fe$_{100-x}$Co$_x$ 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/ω-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 isolation table. The details of our measurement system are reported in our previous paper30). The relative length change, Δ/l, was calculated from the following formula,

\[
\frac{\Delta l}{l} = \frac{\Delta S \cdot t^2 \cdot E_e}{3 \cdot L^2} \cdot \epsilon_f \cdot (1 - \nu_f),
\]

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 \(E_{\parallel}\) and \(E_{\perp}\), where E is the uniaxial stress applied along \([g_1; g_2; g_3]\), \(E_{\parallel}\) is the strain occurred along \([g_1; g_2; g_3]\), and \(E_{\perp}\) 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 expressed3,4 as

\[
1 \frac{E}{E_{\parallel}} = \frac{C_{11} + C_{12}}{(C_{11} - C_{12})(C_{11} + 2C_{12})} + \frac{1}{C_{44}} \left( \frac{2}{C_{11}} - \frac{1}{C_{12}} \right) \left( \gamma_1^2 \gamma_2^2 + \gamma_2^2 \gamma_3^2 + \gamma_3^2 \gamma_1^2 \right),
\]

\[
\nu = \left[ \frac{1}{C_{11} - C_{12}}(C_{11} + 2C_{12}) \left( \frac{2}{C_{44}} \right)^{1/2} \gamma_1^2 \gamma_2^2 + \gamma_2^2 \gamma_3^2 + \gamma_3^2 \gamma_1^2 \right]^{1/2},
\]

where \(C_{11}, C_{12}, C_{44}\) are the elastic stiffness values and \((\gamma_1, \gamma_2, \gamma_3)\) and \((\delta_1, \delta_2, \delta_3)\) are respectively 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 \((a, b, c)\).

Figures 2(a)–(c) show the RHEED patterns observed for Fe100–xCo 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 Nishiya-Wasserman (NW)38,39 and Kurdum-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–xCo(001) single-crystal, (211) bi-crystal, and (110) crystal films are respectively epitaxially grown on MgO(001), MgO(110), MgO(111)/Al2O3(0001) substrates for all the compositions of x = 0–50 at. %.

3. Results and Discussion

3.1 Film growth and structure

Figures 2(a)–(c) show the RHEED patterns observed for Fe100–xCo 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 Nishiya-Wasserman (NW)38,39 and Kurdum-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–xCo(001) single-crystal, (211) bi-crystal, and (110) crystal films are respectively epitaxially grown on MgO(001), MgO(110), MgO(111)/Al2O3(0001) substrates for all the compositions of x = 0–50 at. %.

The crystallographic orientation relationships are determined as

\[
\begin{align*}
Fe_{100-x}Co_{x}(001)[110]_{bcc} &|| MgO(001)[100], \\
Fe_{100-x}Co_{x}(211)[011]_{bcc} &|| MgO(110)[001], \quad \text{(type A)} \\
Fe_{100-x}Co_{x}(211)[011]_{bcc} &|| MgO(110)[001], \quad \text{(type B)}
\end{align*}
\]
Fe\textsubscript{100–x}Co\textsubscript{x}(110)[101], (type KS3) Fe\textsubscript{100–x}Co\textsubscript{x}(110)[110], (type KS4) Fe\textsubscript{100–x}Co\textsubscript{x}(110)[111], (type KS5) Fe\textsubscript{100–x}Co\textsubscript{x}(110)[011], (type KS6) which are similar to the case of Fe\textsubscript{100–x}Co film growth by molecular beam epitaxy\textsuperscript{20–23).}

Figures 3(a-1)–(c-1) show the out-of-plane XRD patterns of the Fe\textsubscript{100–x}Co\textsubscript{x} 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)/Al\textsubscript{2}O\textsubscript{3}(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)/Al\textsubscript{2}O\textsubscript{3}(0001) substrates measured by making the scattering vector parallel to MgO[110], MgO[001], and MgO[110] || Al\textsubscript{2}O\textsubscript{3}[1100], respectively. bcc(200) reflection is observed in the patterns of Fe\textsubscript{100–x}Co\textsubscript{x}(001) single-crystal films [Fig. 3(a-2)]. bcc(011) reflection from the A-type variant and bcc(111) reflection from the B-type variant

![Figure 2](image1.png)  
**Fig. 2** (a)–(c) RHEED patterns observed for (a-1)–(c-1) Fe, (a-2)–(c-2) Fe\textsubscript{50}Co\textsubscript{50}, and (a-3)–(c-3) Fe\textsubscript{70}Co\textsubscript{30} films formed on (a) MgO(001), (b) MgO(110), and (c) MgO(111)/Al\textsubscript{2}O\textsubscript{3}(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[100], (b) MgO[001], (c) MgO[110], (d) bcc[110], (e) bcc[011] || [011], or (f) bcc[001] || [111].

![Figure 3](image2.png)  
**Fig. 3** (a-1)–(c-1) Out-of-plane and (a-2)–(c-2) in-plane XRD patterns of Fe, Fe\textsubscript{50}Co\textsubscript{50}, and Fe\textsubscript{70}Co\textsubscript{30} films formed on (a) MgO(001), (b) MgO(110), and (c) MgO(111)/Al\textsubscript{2}O\textsubscript{3}(0001) substrates. The intensity is shown in logarithmic scale. (d)–(f) Compositional dependences of out-of-plane and in-plane lattice spacings of Fe\textsubscript{100–x}Co\textsubscript{x} films formed on (d) MgO(001), (e) MgO(110), and (f) MgO(111)/Al\textsubscript{2}O\textsubscript{3}(0001) substrates.
seem to be overlapped around $2\theta = 45^\circ$ in the patterns of Fe$_{50}$Co$_{50}$(211) bi-crystal films [Fig. 3(b-2)] by considering the epitaxial orientation relationships determined by RHEED. bcc(002) reflection from the NW1-type variant is recognized, whereas bcc(111) reflection from the KS1-type variant and bcc(111) reflection from the KS4-type variant, which are expected to appear around $2\theta = 55^\circ$, are absent in the patterns of Fe$_{100}$Co$_{100}$(110) epitaxial films [Fig. 3(c-2)], because the bcc(hkl) reflections ($h$, $k$, $l$) are forbidden. The out-of-plane and in-plane XRDs confirm the epitaxial orientation relationships determined by RHEED. Figures 3(d)-(f) summarize the out-of-plane and in-plane lattice spacings of Fe$_{100}$Co$_{100}$ films, which are estimated from the XRD data. The lattice parameters of Fe$_{100}$Co$_{100}$ films agree with small differences within ±0.7% with those of bulk Fe$_{100}$Co$_{100}$ crystal[10], which indicates that the lattice strains of Fe$_{100}$Co$_{100}$ films are small.

### 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$_{50}$Co$_{50}$(001) single-crystal films. The distributions of normalized remnant magnetization, $M_r/M_s$, 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$_{100}$ 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$_{100}$ crystal with the easy magnetization axes parallel to <100>-<001>. Figure 4(c) shows the magnetization property of the Fe$_{50}$Co$_{50}$(001) single-crystal film. Although a four-fold symmetry in in-plane magnetic 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$_{50}$Co$_{50}$ films. It is known that the easy magnetization axes of bulk Fe$_{100}$Co$_{100}$ crystal vary from <100> to <111> when the Co content increases beyond about 40 at. %[8]. Therefore, the in-plane magnetic anisotropy observed for Fe$_{50}$Co$_{50}$ film seems to be reflecting the magnetocrystalline anisotropy of Fe$_{100}$Co$_{100}$ crystal with the easy axes parallel to <111> and the demagnetization field. The magnetic anisotropy of Fe$_{50}$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 $M_r/M_s$ and saturation field ($H_s$) 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]1, [215]1, [251]6, and [215]6 (pink dotted lines in Figs. 5(a-2) and (a-3)), which are respectively obtained by projecting [010]1, [001]1, [010]6, and [001]6 on the (211) surface as shown in Fig. 6. Therefore, the magnetic anisotropy is interpreted to be reflecting the magnetocrystalline anisotropy of Fe70Co30 crystal with the easy axes parallel to <100> and the demagnetization film, similar to the cases of Fe and Fe50Co50(001) single-crystal films. Figure 5(c) shows the magnetic property of the Fe70Co30(211) film. A two-fold symmetry in in-plane magnetic anisotropy is observed. However, the easy magnetization directions are parallel to [011]1, [011]6, and [011]10 (blue solid lines in Figs. 5(c-2) and (c-3)). It is also noted that the Hr values measured along [111]1, γ[111]6, and [111]10 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]1, γ[111]6, and [111]10. On the contrary, the magnetization curves measured along [251]1, [215]1, [251]6, and [215]6, which are respectively obtained by projecting [010]1, [001]1, [010]6, and [001]6 on the film plane, saturate at higher magnetic fields (pink dotted lines in Fig. 5(c-2)). When the Co content increases up to 51%, γ[001]1 and γ[011]6 are coexisting in the (110) epitaxial films and the respective 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 magnetostriction9 is shown as

\[
\frac{\Delta l}{l} = \frac{3}{2} \lambda_{110} (a_1^2 \beta_1^2 + a_2^2 \beta_2^2 + a_3^2 \beta_3^2 - \frac{1}{3}) + 3\lambda_{111} (a_1a_2 \beta_1 \beta_2 + a_1a_3 \beta_1 \beta_3 + a_2a_3 \beta_2 \beta_3),
\]

where λ110 and λ111 are the magnetostriction coefficients, (a1, a2, a3) and (β1, β2, β3) 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 \(\cos \theta, \sin \varphi, 0\), where \(\varphi\) is the angle of magnetization direction with respect to [100]. The (a1, a2, a3) values are thus expressed as

\[
(a_1, a_2, a_3)_{\text{parallel}} = (\cos \theta, \sin \varphi, 0).
\]

When the observation directions are parallel to [100] and [110], the (β1, β2, β3)_{100} and the (β1, β2, β3)_{110} values are respectively expressed as

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

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

By substituting Eqs. (5)–(7) into (4), the relative length changes measured along [100] and [110] under in-plane rotating
and (c). Furthermore, the phases of which are respectively cosine and sine waves. It is noted that the magnetic field, measured for the Fe 100–directions. (b) Fe100–(001) single-crystal films with different compositions. (c) Compositional dependences of λ100 and λ111. The bulk values in (c) are cited from Refs. 5–8 and 42.

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

(8)

\[ \frac{\Delta l}{l_{111}}(\phi) = \frac{3}{4} \lambda_{111} \sin 2\phi + \frac{1}{4} \lambda_{100}, \]

(9)

which are respectively cosine and sine waves. It is noted that the phases of \( \Delta l/l_{100}(\phi) \) and \( \Delta l/l_{110}(\phi) \), 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,

\[ \lambda_{100} = \frac{4}{3} \left[ \frac{\Delta l}{l_{100}}(\phi = 0°) - \frac{\Delta l}{l_{100}}(\phi = 45°) \right]. \]

(10)

\[ \lambda_{111} = \frac{4}{3} \left[ \frac{\Delta l}{l_{111}}(\phi = 0°) - \frac{\Delta l}{l_{111}}(\phi = 90°) \right]. \]

(11)

Figure 8(d-1) shows the \( \Delta l/l_{100}(\phi) \) measured for the FeCoCo[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 FeCoCo[001] films. Figure 8(d-2) shows the \( \Delta l/l_{100}(\phi) \) measured for the FeCoCo[001] films. The phases of waves observed for Fe and FeCoCo films agree with that of calculated wave of Fig. 8(c-2), whereas the phase of wave observed for FeCoCo film is in agreement with that of wave of Fig. 8(c-1). Therefore, the \( \lambda_{111} \) value is negative for the Fe and the FeCoCo films, while that is positive for the FeCoCo 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 FeCoCo film shows a large \( \lambda_{100} \) value of +234×10–6 and a small \( \lambda_{111} \) value of –5×10–6. On the contrary, a large \( \lambda_{111} \) value of +274×10–6 and a moderately large \( \lambda_{111} \) value of +78×10–6 are observed for the FeCoCo film.

3.4 Magnetostriction of (211) bi-crystal films

When the magnetization rotates in a (211), plane as shown in Fig. 9(a), the crystallographic direction of magnetization is shown as \([ -\sin\chi/\sqrt{3}, \cos\chi/\sqrt{3}, \sin\chi/\sqrt{3} - \cos\chi/\sqrt{3} + \sin\chi/\sqrt{3}] \), where \( \chi \) is the angle of magnetization direction with respect to [011] \( \Delta \) (MgO[001]). The \( (a_1, a_2, a_3)_{(211)} \) values are thus expressed as

\[ (a_1, a_2, a_3)_{(211)} = (\frac{\sin\chi}{\sqrt{3}}, \frac{\cos\chi}{\sqrt{3}}, \frac{\sin\chi}{\sqrt{3}} + \frac{\cos\chi}{\sqrt{3}} + \frac{\sin\chi}{\sqrt{3}}). \]

(12)

When the observation directions are parallel to [011] \( \Delta \) and [111] \( \Delta \), the \( (\beta_1, \beta_2, \beta_3)_{(011)} \) and the \( (\beta_1, \beta_2, \beta_3)_{(111)} \) values are respectively expressed as
Fig. 9 (a) Schematic diagram showing the magnetization and the observation directions with respect to the typical crystallographic directions. (b) $\Delta l_{/\text{MgO[001]}(\phi)}$ and (c) $\Delta l_{/\text{MgO[110]}(\phi)}$ calculated for (211) bi-crystal films with (b-1) $\lambda_{100}^{\phi}+5\lambda_{111}^{\phi} > 0$, (b-2) $\lambda_{100}^{\phi}+5\lambda_{111}^{\phi} < 0$, (c-1) $\lambda_{111}^{\phi} < 0$, and (c-2) $\lambda_{111}^{\phi} > 0$. (d-1) $\Delta l_{/\text{MgO[001]}(\phi)}$ and (d-2) $\Delta l_{/\text{MgO[110]}(\phi)}$ measured for Fe$_{50}$Co$_{50}$ (211) bi-crystal films formed on MgO[110] substrates. (e) Compositional dependences of $\lambda_{100}$ and $\lambda_{111}$. The bulk values in (e) are cited from Refs. 5–8 and 42.

\begin{align}
(\beta_1, \beta_2, \beta_3)_{011A} &= (0, \frac{1}{2\sqrt{2}} \phi, -\frac{1}{2\sqrt{2}} \phi), \\
(\beta_1, \beta_2, \beta_3)_{111A} &= (-\frac{1}{2\sqrt{2}} \phi, \frac{1}{2\sqrt{2}} \phi, \frac{1}{2\sqrt{2}} \phi). 
\end{align}

By substituting Eqs. (12)–(14) into (4), the $\Delta l_{/011A}(\phi)$ and the $\Delta l_{/111A}(\phi)$ are respectively given as

\begin{align}
\Delta l_{/011A}(\phi) &= -\frac{3}{4} \lambda_{111} \cos 2\phi + \frac{1}{4} \lambda_{111}, \\
\Delta l_{/111A}(\phi) &= \frac{\lambda_{011}}{2} \cos 2\phi + \frac{3}{8} \lambda_{111} \cos 2\phi + \frac{1}{8} \lambda_{111}.
\end{align}

Since the Fe$_{50}$Co$_{50}$ (211) films consist of two types of variants, A and B, it is necessary to take into account the $\Delta l_{/011A}(\phi)$ and the $\Delta l_{/111A}(\phi)$, which are respectively shown as follows,

\begin{align}
\Delta l_{/011A}(\phi) &= \lambda_{011} \cos 2\phi + \frac{3}{8} \lambda_{111} \cos 2\phi + \frac{1}{8} \lambda_{111}, \\
\Delta l_{/111A}(\phi) &= -\frac{3}{4} \lambda_{111} \cos 2\phi + \frac{1}{4} \lambda_{111}.
\end{align}

Therefore, when the magnetostriiction is measured along MgO[001] (|| Fe$_{50}$Co$_{50}$[01 T]$_{\phi}^\perp$[0 T 1]$_{\phi}^\parallel$ and MgO[1 0 0] (|| Fe$_{50}$Co$_{50}$[1 1 1]$_{\phi}^\perp$[1 1 1]$_{\phi}^\parallel$), the averages of relative length changes of types A and B variants, $\Delta l_{/\text{MgO[001]}(\phi)}$ and $\Delta l_{/\text{MgO[110]}(\phi)}$, are respectively given as

\begin{align}
\Delta l_{/\text{MgO[001]}(\phi)} &= \frac{1}{2} \left[ \lambda_{100}^{\phi} + \frac{5}{8} \lambda_{111}^{\phi} \right] \cos 2\phi + \frac{1}{8} \lambda_{100}^{\phi} + \frac{1}{8} \lambda_{111}^{\phi}, \\
\Delta l_{/\text{MgO[110]}(\phi)} &= \frac{1}{2} \left[ \lambda_{100}^{\phi} + \frac{5}{8} \lambda_{111}^{\phi} \right] \cos 2\phi + \frac{1}{8} \lambda_{100}^{\phi} + \frac{1}{8} \lambda_{111}^{\phi},
\end{align}

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

\begin{align}
\lambda_{100} &= \frac{8 \Delta l_{/\text{MgO[001]}(\phi)}}{\Delta l_{/\text{MgO[110]}(\phi)}} \left( \lambda_{100}^{\phi} + \frac{5}{8} \lambda_{111}^{\phi} \right) \cos 2\phi + \frac{1}{8} \lambda_{100}^{\phi} + \frac{1}{8} \lambda_{111}^{\phi}, \\
\lambda_{111} &= -\frac{4}{3} \left[ \lambda_{100}^{\phi} + \frac{5}{8} \lambda_{111}^{\phi} \right] \cos 2\phi + \frac{1}{8} \lambda_{100}^{\phi} + \frac{1}{8} \lambda_{111}^{\phi}.
\end{align}

Reference
3.5 Magnetostriction 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 $\sin\psi/\sqrt{2}$ $-$ $\sin\psi/\sqrt{2}$ $\cos\psi$. Here, $\psi$ is the angle of magnetization direction with respect to [001]. The $(\alpha_1, \alpha_2, \alpha_3)$ values are thus expressed as 

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

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

$$(\beta_1, \beta_2, \beta_3) = (\sin\omega/\sqrt{2}, -\sin\omega/\sqrt{2}, \cos\omega).$$

The $\Delta l_{\text{Fe}70\text{Co}30}$ $\sin\psi/\sqrt{2}$ $\cos\psi$ is thus given by substituting Eqs. (23) and (24) into (4) as follows:

$$\Delta l/I = \frac{2}{3} \lambda_{100} \left( \frac{\sin^2\psi\sin^2\omega}{2} + \cos^2\psi\cos^2\omega - \frac{1}{3} \right) + 3 \lambda_{111} \left( \frac{\sin^2\psi\sin^2\omega}{4} + \cos^2\psi\cos^2\omega \cos\omega \right).$$

In order to characterize the magnetostriction 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 $\Delta l/I(\psi)$ of each variant is expressed as 

$$\frac{\Delta l}{I}(\psi) = \frac{1}{2\pi} \int_0^{2\pi} \frac{\Delta l}{I}(\psi, \omega) \, d\omega$$

$$= \frac{3}{16} \sum \lambda_{100} - \lambda_{111} \cos^2\omega + \frac{1}{16} \left( \lambda_{100} + 3\lambda_{111} \right),$$

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

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

Figure 10(c) shows the $\Delta l/I(\psi)$ 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$_{50}$Co$_{50}$ and Fe$_{70}$Co$_{30}$ 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$_{50}$Co$_{50}$ and the Fe$_{70}$Co$_{30}$ 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$_{50}$Co$_{50}$ and the Fe$_{70}$Co$_{30}$ 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 magnetostriction.

Fig. 10 (a) Schematic diagram showing the magnetization and the observation directions with respect to the typical crystallographic directions. (b) $\Delta l/I(\psi)$ calculated for (110) epitaxial multi-crystal films with (b-1) $\lambda_{100} > \lambda_{111}$ and (b-2) $\lambda_{100} < \lambda_{111}$. (c) $\Delta l/I(\psi)$ 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 magneto crystalline 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 magnetic anisotropy is 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 $\lambda_{100}$ and the $\lambda_{111}$ values, respectively, increase from +10\textsuperscript{8} to +10\textsuperscript{4} and from $-10^{5}$ to $+10^{5}$ for both Fe\textsubscript{100-x}Co\textsubscript{x}(001) single-crystal and (211) bi-crystal films. Large $\lambda_{100}$ values are also indicated for the Fe\textsubscript{50}Co\textsubscript{50} and the Fe\textsubscript{50}Co\textsubscript{50}(110) epitaxial films. The present study shows that it is possible to obtain large magnetostriiction of $10^{-4}$ by control of the film orientation and composition.

References

38) G. Wasserman: Arch. Eisenhüttenwes, 16, 647 (1933).

Received Nov. 4, 2018; Revised Feb. 15, 2019; Accepted Feb. 27, 2019