Magnetostrictive Behaviors of fcc-Co(001) Single-Crystal Films Under Rotating Magnetic Fields

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Magnetostrictive behaviors of fcc-Co(001) single-crystal films with thicknesses ranging from 40 to 500 nm are investigated under rotating magnetic fields up to 1.2 kOe. The magnetostrictive anisotropy estimated from the magnetization curves is confirmed to be lying with four-fold symmetry, and the anisotropy field is within a narrow range of 900–950 Oe for these films. The easy and the hard axes are lying parallel to <110> and <100>, respectively. The magnetostrictive behavior measured along [110] shows rectangular or bathtub-like waveform and the amplitude of magnetostriction constants (λ₁₀₀, λ₁₁₁) estimated from the amplitudes measured at 1.2 kOe are λ₁₀₀=(85–90)×10⁻⁶ and λ₁₁₁=(40–50)×10⁻⁶, respectively for these films. The λ₁₀₀ values are in agreement with that estimated by the first-principle calculation.

Key words: fcc-Co, magnetocrystalline anisotropy, magnetostriction, single-crystal film

1. Introduction

The crystal structure of Co metal is known to be hexagonal close packed (hcp) at room temperature and it changes to face centered cubic (fcc) over 450°C according to the phase diagram. On the other hand, metastable fcc-Co appears even at room temperature in the films prepared by molecular beam epitaxy or sputtering process. Single-crystal films of fcc-Co(001) are prepared by heteroepitaxial film growth on Cu(001) underlayers, and their magnetic anisotropies are reported to be with the easy and the hard axes along <110> and <100>, respectively. Magnetic properties of fcc-Co are different from those of hcp-Co. However, systematic magnetostriction studies of fcc-Co(001) single-crystal films have not yet been reported.

In the present study, well defined fcc-Co(001) single-crystal films are prepared on MgO(001) single-crystal substrates by employing Pd and Cu underlayers and the magnetostrictive behaviors are investigated under rotating magnetic fields up to 1.2 kOe. The experimental results are analyzed by using a coherent rotation model of magnetization considering the magnetocrystalline anisotropy.

2. Experimental Procedure

Co films with thicknesses of 40–500 nm were prepared on MgO(001) single-crystal substrates of 20×20×0.5 mm³ with Pd (10 nm) and Cu (10 nm) underlayers at 300 °C by using a radio-frequency 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, the MgO substrates were heated at 600 °C for 1 hour in the ultra-high vacuum chamber to obtain clean surface, which was confirmed by RHEED. The underlayers of Pd and Cu were employed to adjust the lattice mismatch (16%) between MgO and fcc-Co and also to promote Co thin film growth with a metastable fcc structure. The film structure was confirmed by RHEED and X-ray diffraction (XRD) with Cu-Kα radiation (λ=0.15418 nm). Magnetization curves were measured by using a vibrating sample magnetometer (VSM). Magnetostriction measurements were carried out by using a cantilever method under rotating magnetic fields up to 1.2 kOe. The rotating speed was 5 rpm. The magnetostriction observation directions were set along [100] and [110] of the MgO(001) substrates. The magnetostriction coefficient λ is calculated from the following formula:

\[
\lambda = \frac{\Delta S \cdot t_f^2}{3L \cdot t_f} \cdot \frac{E \cdot (1 + \nu_l)}{E_f \cdot (1 - \nu_c)}
\]

where ΔS is the measured bending, L is the distance between laser beam points (12.5 mm), t is the thickness, E is Young’s modulus, ν is Poisson’s ratio, and the subscripts of f and s respectively refer to film and substrate. For the single-crystal films and the single-crystal substrates, the values shown in Table 1 are employed because the elastic property of single-crystal sample is anisotropic. In the calculations, the elastic stiffness values of fcc-Ni were employed for the values of fcc-Co because the stiffness values of fcc-Co were unknown.
Table 1  $E$ and $\nu$ values of fcc-Co and MgO materials used in the present study $^{10,11}$.

| Orientation | fcc-Co | MgO |
|-------------|--------|-----|
| $<100>$     | 129    | 185 |
| $<110>$     | 227    | 215 |

3. Results and Discussion

3.1 Crystal structure of Co films grown on Cu/Pd underlayers

RHEED patterns observed for Co films grown on Cu/Pd underlayers are shown in Fig. 1. Similar sharp streaks are observed for the Co films of different thicknesses ranging from 40 to 500 nm, which indicates that the films are single-crystals with smooth surfaces.

![RHEED patterns observed for Co films of different thicknesses](image)

Figure 2 shows the out-of-plane and the in-plane XRD patterns observed for these Co films. The fcc(002) and (004) peaks are clearly recognized in addition to the reflections from the MgO substrates and the Cu/Pd underlayers. Therefore, the films are determined to be fcc(001) single-crystal films. The epitaxial orientation relationship to the MgO substrate is determined as

$$\text{fcc-Co}(001)[100] \parallel \text{MgO}(001)[100].$$

The lattice parameters determined from the peak positions of fcc(002) in the out-of-plane and the in-plane XRDs are shown in Fig. 3. The lattice parameter measured parallel to the substrate surface, $a$, is larger than that measured perpendicular, $c$, for the thickness range between 40 and 500 nm, indicating that the fcc-crystal is slightly deformed. This is due to the hetero-epitaxial growth of fcc-Co(001) film on the fcc-Cu(001) under layer, where the lattice parameters of fcc-Cu ($a=0.3614$ nm) is 2.1% larger than that of bulk fcc-Co ($a=0.354$ nm) and the fcc-Co lattice is expanded in lateral direction. The difference of lattice parameters of fcc-Co(001) film decreases with increasing the film thickness as shown in Fig. 3 suggesting that the influence on the lattice strain from the Cu underlayer is decreasing with increasing the Co film thickness. The full width at half-maximum values ($\Delta \theta_{\text{FWHM}}$) is shown in Fig. 4 as a function of film thickness. The $\Delta \theta_{\text{FWHM}}$ values measured from the out-of-plane and the in-plane XRDs are decreasing with increasing the film thickness, which indicate that the crystallographic quality is improved for thicker fcc-Co(001) films. These experimental results confirm that the Co films prepared
in the present study are single-crystal films with fcc(001) orientation involving very small lattice deformation which decreases with increasing the film thickness.

3.2 Magnetization curves of fcc Co(001) films

The magnetization curves of fcc Co(001) films measured with the magnetic field aligned parallel to [100] and [110] are shown in Fig. 5. All the films are showing similar types of M-H curve with the easy and the hard axes parallel to [110] and [100], respectively. The anisotropy field matches to the saturation field for the film thickness between 40 and 500 nm. The anisotropy field is nearly equal to those reported in the references 5-7. The coercivities of these films are ranging between 40 and 50 Oe, which are far smaller than the $H_a$ values (900-950 Oe). Such kind of magnetization curves can be expressed by using the modified coherent rotation model 12). However, the detail of magnetization curve is slightly different depending on the film thickness. The M-H curves along $<110>$ for the (40-200) nm thick films show sharp shoulder, which means that the coherent rotation of magnetization is dominant in the magnetization process. On the other hand, the M-H curve for the 500 nm thick film shows mild shoulder, which means that the coherent rotation mode is slightly disturbed in the film. The small coercivities mean that the magnetic domain wall motions are dominant in the magnetization reverse process under small magnetic fields less than 50 Oe.

3.3 Magnetostrictive behavior of fcc-Co(001) films

Figure 6 shows the magnetostrictive behaviors measured along fcc[110] and [100] for the 40 nm thick film under rotating magnetic fields. The output voltage of 1 V corresponds to 0.2 μm bending of the sample. The rotating angle, $\theta$, corresponds to the magnetic field angle is opposite to $\theta/2$[110] and to [110] for $\theta/[100]$. The output voltages are quite small,

![Fig. 6. Measured magnetostrictive behaviors of 40 nm thick fcc-Co(001) single-crystal film under rotating magnetic fields. The magnetostrictive observation direction is along fcc[110] and [100], respectively.](image-url)
means that the sign of magnetostriction constant is opposite between the two orientations.

Figure 7 shows the magnetostrictive behaviors measured for the 100 nm thick film. The output amplitude is increasing in proportional to the film thickness. The waveforms measured along [110] and [100] resemble to those for the 40 nm thick film, namely the output waveform changes from rectangular to bath-tub like with increasing the magnetic field and the output intensity is kept almost at a similar amplitude along [110], whereas the output waveform measured along [100] is triangular and the amplitude increases with increasing the magnetic field.

\[ \text{Output (V)} \]
\[ \lambda / \text{fcc[110]} \]
\[ 0.2 \text{kOe} \]
\[ 0.3 \text{kOe} \]
\[ 0.5 \text{kOe} \]
\[ 0.7 \text{kOe} \]
\[ 1.0 \text{kOe} \]
\[ 1.2 \text{kOe} \]
\[ \lambda / \text{fcc[100]} \]
\[ 0.2 \text{kOe} \]
\[ 0.3 \text{kOe} \]
\[ 0.5 \text{kOe} \]
\[ 0.7 \text{kOe} \]
\[ 1.0 \text{kOe} \]
\[ 1.2 \text{kOe} \]

\[ \text{Rotation angle (deg.)} \]
\[ 0 \]
\[ 90 \]
\[ 180 \]
\[ 270 \]
\[ 360 \]

**Fig. 7** Measured magnetostrictive behaviors of 100 nm thick fcc-Co(001) single-crystal film under rotating magnetic fields. The magnetostriiction observation direction is along fcc[110] and fcc[100], respectively.

The output phases are reversed similar to the case of 40 nm thick film (Fig. 6). The waveforms for the 200 nm thick film shown in Fig. 8 are also similar to those for the 100 nm thick film. However, the waveforms for the 500 nm thick film are slightly different from those for 40 or 100 nm thick films. The waveform along [110] is not rectangular but bath-tub like even under a small magnetic field of 0.2 kOe. In the case that the magnetization process is based on a coherent rotation mode, the waveform should be rectangular under a sufficiently small magnetic field compared with its anisotropy field as reported in the previous report. In the case that the magnetization process is based on a coherent rotation mode, the waveform should be rectangular under a sufficiently small magnetic field compared with its anisotropy field as reported in the previous report. It is notable that the films with sharp rectangular M-H curves (Fig. 5), which are of the 40 and 100 nm thick films, show rectangular waveforms and the film with mild square M-H curve measured for the 500 nm thick film (Fig. 5) shows a bath-tub like waveform even under a small magnetic field. Therefore, the behavior of 500 nm thick film suggests that the magnetization process is deviating slightly from the coherent mode.

**Fig. 8** Measured magnetostrictive behaviors of 200 nm thick fcc-Co(001) single-crystal film under rotating magnetic fields. The magnetostriiction observation direction is along fcc[110] and fcc[100], respectively.

These unique magnetostrictive behaviors can be explained by considering coherent rotation of magnetization under a rotating magnetic field with taking into account the magnetocrystalline anisotropy. In the case that the intensity of rotating magnetic field is comparable to the magnetocrystalline anisotropy field, the magnetization tends to keep its direction parallel to the easy magnetization axis and tends to leave its direction from the hard magnetization axis. Only in the case where the intensity of rotating magnetic field is strong enough to overcome the anisotropy field, the magnetization will rotate under an influence of the applied rotating magnetic field and the magnetostrictive waveform is expected to be sinusoidal.
These behaviors are similar to those for bcc(001) single-crystal films with four-fold magnetocrystalline anisotropy \(^{12, 13}\).

A phenomenological model is described as follows. The change in length by magnetostriction is given by the following equation for a cubic crystal \(^{14}\).

\[
\frac{\delta l}{l} = \frac{3}{2} \lambda_{100} \left( a_1^2 \beta_1^2 + a_2^2 \beta_2^2 + a_3^2 \beta_3^2 - \frac{1}{3} \right) + 3 \lambda_{111} \left( a_1 \cdot a_2 \cdot \beta_3 + a_2 \cdot a_3 \cdot \beta_1 + a_3 \cdot a_1 \cdot \beta_2 \right).
\]  

(2)

Here \(\lambda_{100}\) and \(\lambda_{111}\) are the longitudinal magnetostriction for [100] and [111] directions, \(\alpha\)'s are the direction cosines of the magnetization with respect to the crystal axes, and \(\beta\)'s the direction cosines of the measured length change. Since the change in length is a function of direction cosine of magnetization in this equation, it is based on a coherent rotation model. In a coherent rotation model, the free energy is given by the following equation considering magnetocrystalline anisotropy energy \((K_i)\) and Zeeman energy \((M_s H_\alpha)\).

\[
E = \frac{1}{8} K_i (1 - \cos 4\theta) + M_s H_\alpha \cdot \cos (\phi - \theta).
\]  

(3)

Here \(\theta\) and \(\phi\) are the direction of \(M_s\) and \(H_\alpha\) with respect to the [100] axis in the (001) plane, respectively. For an observation direction of the change in length along [110] in the (001) plane, the equation (2) is rewritten as the following equation.

\[
\frac{\delta l}{l} = \frac{1}{4} \lambda_{100} + \frac{3}{4} \lambda_{111} \sin 2\theta.
\]  

(4)

This equation shows that the direction depending part depends only on \(\lambda_{111}\), namely \(\lambda_{111}\) can be estimated from the measured amplitude along [110]. The magnetostrictive behavior along [100] can also be calculated. In this case, the equation (2) is rewritten as the following equation.

\[
\frac{\delta l}{l} = \frac{1}{4} \lambda_{100} \left(1 + 3 \cos 2\theta\right).
\]  

(5)

In the case, the direction depending part of the change in length depends only on \(\lambda_{100}\), namely \(\lambda_{100}\) can be estimated from the measured amplitude along [100].

In order to calculate the change in length, we need to know the angle \(\theta\) under an applied rotating magnetic field. The angle \(\theta\) is determined so as to minimize the free energy, \(E\) in the equation (3). The calculated results are shown in Fig. 10 for \(h=1\) and \(2\), where \(h\) is defined as \(h=H_\alpha/H_\alpha\). Therefore, the calculated results for \(h=1\) are corresponding to the experimental results under \(H_\alpha=1.0\) kOe and can explain the characteristic waveforms such as bath-tub like or triangular. The magnetostriction constants, \(\lambda_{100}\) and \(\lambda_{111}\) can be estimated from the amplitudes of output. Fig. 11 shows the output amplitude as a function of applied magnetic field up to 1.2 kOe. The amplitude along [110] keeps almost a same level and is well saturated in the

![Fig. 10](image)

Calculated magnetostrictive behaviors along (a) [110] and (b) [100], \(\lambda_{100}\) and \(\lambda_{111}\) are assumed to be \(10 \times 10^{-6}\). \(K_i\) is assumed to be negative. \(h\) is defined as \(h=H_\alpha/H_\alpha\), where \(H_\alpha\) is anisotropy field.

![Fig. 11](image)

Magnetic field dependence of output amplitude measured along (a) [110] and [100] of fcc-Co(001) single-crystal film sample with the thickness of 50, 100, 200, and 500 nm, respectively.

![Fig. 12](image)

Film thickness dependences of magnetostrictiction constants \(\lambda_{100}\) and \(\lambda_{111}\) estimated for fcc-Co(001) single-crystal films.
magnetic field range. However, the amplitude along [100] increases with increasing the magnetic field and the amplitudes for 40 nm, 100 nm, and 200 nm thick films are almost saturated at 1 kOe but that for 500 nm thick film is still approaching to saturation at 1.2 kOe. The estimated magnetostriction constants are shown in Fig. 12. The $\lambda_{100}$ shows large positive value ranging from 85 to $90 \times 10^{-6}$ and shows small film thickness dependence. The $\lambda_{111}$ shows negative value ranging from $-40$ to $-45 \times 10^{-6}$ and also shows small film thickness dependence. The large positive value is consistent to the value calculated by a first principle method 15).

4. Conclusion

The magnetostrictive behaviors of fcc-Co(001) single-crystal films are investigated under rotating magnetic fields. The magnetostrictive behaviors along fcc[110] show rectangular or bath-tub like waveform and the amplitude is kept at almost a similar level, while the behaviors along fcc[100] show triangular and the amplitude increases with increasing the magnetic field. The behaviors can be explained by the coherent rotation model of magnetization assuming a four-fold magnetocrystalline anisotropy in (001) plain. The magnetostriction constant $\lambda_{100}$ is determined to be large positive ranging from 85 to $90 \times 10^{-6}$, which agrees with that obtained by theoretical calculation. $\lambda_{111}$ is negative ranging from $-40$ to $-45 \times 10^{-6}$. The thickness dependences of $\lambda_{100}$ and $\lambda_{111}$ are confirmed to be very small for the fcc-Co(001) films.

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