Determination of Crystallographic Phase and Estimation of Order Degree for Rare Earth-Transition Metal Alloy Films with Hexagonal Structures

Mitsuru Ohtake and Masaaki Futamoto
Faculty of Science and Engineering, Chuo University, 1-13-27 Kasuga, Bunkyo-ku, Tokyo 112-8551, Japan

A magnetic alloy film consisting of rare earth (R) and transition (T) metals with hexagonal structure may involve crystallographic phases such as R2T7, RT5, and RT3 in addition to RT5 phase. The structures of hexagonal R-T films with the c-axis parallel and perpendicular to the substrate surface are characterized by employing a combination of reflection high-energy electron diffraction (RHEED) and X-ray diffraction (XRD). The crystallographic phase is identified by analyzing a reciprocal lattice map measured by RHEED. The degree of RT5 ordering is estimated by comparing XRD data (intensity, structure factor, Lorentz-polarization factor, and absorption factor) of (0001) superlattice and (0002) fundamental reflections (conventional method). A simplified method for estimating the order degree, where only the intensity ratio of (0001) to (0002) reflection is considered, is proposed for some of the typical RT5 film materials of SmCo5, PrCo5, YCo5, etc. The order degree estimated by employing the simplified method agrees in a small error less than a few percentages with that calculated using the conventional method.

Key words: rare earth-transition metal alloy, magnetic film, hexagonal RT5 phase, order degree

1. Introduction

Magnetic films with high magnetic anisotropy energies have attracted much attention to nanocomposite magnet applications (micromotors, microsensors), magnetic recording applications (energy-assisted perpendicular recording media), etc. Ordered alloys consisting of rare earth (R) and 3d ferromagnetic transition (T) metals with hexagonal DH2 structure such as SmCo5, PrCo5, etc. show high uniaxial magnetocrystalline anisotropy energies (Ku) of $10^6$–$10^8$ erg/cm$^3$ along the c-axis. In order to align the easy magnetization axis parallel or perpendicular to the substrate surface, a well-defined epitaxial film is useful, since the crystallographic orientation can be controlled by the substrate orientation.

SmCo5, PrCo5, NdCo5, YCo5, GdCo5, and SmNi5 epitaxial films with the c-axis lying in the film plane have been prepared on MgO(100) and MgO(110) single-crystal substrates by employing bcc-W, bcc-Cr, bcc-Fe, bcc-(Fe,Co), and hcp-Co underlayers, whereas SmCo5, NdCo5, YCo5, GdCo5, SmFe5, and SmNi5 epitaxial films with the c-axis perpendicular to the substrate surface have been formed on MgO(111) and Al2O3(0001) substrates by using fcc-Cu and hcp-Ru underlayers. However, films deposited at low substrate temperatures tend to include amorphous phase, MgO(111) and Al2O3(0001) substrates by using fcc-Cu and hcp-Ru underlayers. However, films deposited at low substrate temperatures tend to include amorphous phase. There is also a possibility that multiple hexagonal crystallographic phases such as R2T7 ($R \cdot 89 \text{ at.} \% \ T$, $R \cdot 78 \text{ at.} \% \ T$, and $R \cdot 78 \text{ at.} \% \ T$) are involved in addition to RT5 ($R \cdot 83 \text{ at.} \% \ T$) phase. Furthermore, magnetic anisotropy of RT5 material is considered to be enhanced by increasing the long-range order degree (S), similar to the cases of magnetic ordered alloy materials like L11-CoPt29,30, etc. It is thus important to crystallize RT5 phase with a high S value.

Conventional out-of-plane and in-plane X-ray diffractions (XRDs) are not easy to distinguish the hexagonal phases of R2T7, RT5, R2T7, and RT5, since the lattice spacings are very similar each other. Reciprocal lattice map measurement like electron diffraction is effective for the phase identification. Reflection high-energy electron diffraction (RHEED) is useful, since in-situ observation during film formation is often available. On the other hand, an XRD system has better measurement precision of S value when compared with that obtainable with an electron diffraction system. A combination of RHEED and XRD is thus useful for the structural characterization.

The purpose of present study is to apply the combination to hexagonal R-T films with the c-axis parallel and perpendicular to the substrate surface and to show the usefulness in structural characterization. The crystal structure is identified mainly by RHEED supported by XRD, whereas the order degree is estimated by using XRD data of intensity, structure factor, Lorentz-polarization factor, and absorption factor (conventional method). In the order degree estimation, a simplified method, where only the intensity ratio of (0001) to (0002) reflection is considered, is proposed for some of the typical RT5 materials. The accuracies in order degrees estimated by employing the simplified method are discussed in comparison with those estimated by using the conventional method. In the present paper, the experimental data of SmCo5 (at. %) epitaxial films with the c-axis parallel and perpendicular to the substrate surface which have been reported in our previous papers9,10,17,19,21 are used as examples of RT5 materials.
2. Experimental Procedure

A molecular beam epitaxy system equipped with an RHEED facility was used for film preparation. Sm$_{17}$Co$_{83}$ (at. %) films of 20 nm thickness were deposited on 20 nm thick bcc-Cr(100)$^9$, bcc-Cr(211)$^{10}$, bcc-Fe$_{50}$Co$_{30}$(211)$^{10}$, and fcc-Cu(111)$^{21}$ single underlayers at (a) 300 and (b)–(f) 500 °C. The incident electron beam is parallel to [(a), (b)] Cr[011], (c) Cr[110], (d) bcc-Fe$_{50}$Co$_{30}$(211)$^{10}$, (e) fcc-Co(111)/fcc-Cu(111)$^{19}$, and (f) fcc-Cu(111)$^{21}$ underlayers at (a) 300 and (b)–(f) 500 °C. The details of preparation method are shown in our previous papers.$^{8,10,17,19,21}$

The surface structure during film formation was observed by RHEED. An XRD (ULTIMA IV, Rigaku Corp.) system with the axes of $\lambda$, $\theta$, $\varphi$, and 2$\theta$ was employed. The wavelength of X-ray, $\lambda$, was 0.15418 nm (Cu-K$_\alpha$ radiation). 2$\theta$ out-of-plane measurements were carried out.

3. Determination of Crystallographic Phase

Figure 1(a) shows the RHEED pattern observed for an Sm$_{17}$Co$_{83}$ film deposited on bcc-Cr(100) underlayer at 300 °C$^9$. A diffuse diffraction pattern is observed. The result indicates that an amorphous film is formed. Figure 1(b) shows the RHEED pattern of an Sm$_{17}$Co$_{83}$ film deposited on Cr(100) underlayer at 500 °C$^9$. A clear diffraction pattern is observed. An epitaxial Sm-Co film is obtained on the underlayer. Figures 2(a-1)–(a-4) and (b-1)–(b-4), respectively, show the schematic diagrams of diffraction patterns simulated for (1120) crystals with $R_T$, $R_{T_1}$, $R_{T_2}$, $R_{T_3}$, and $RT_5$ structures formed in the orientation relationships of

\[ R-T(1\overline{1}00)[001] \parallel bcc(100)[110], \]
\[ R-T(1\overline{1}00)[1\overline{1}00] \parallel bcc(100)[110]. \]

The schematic diagrams of Fig. 2(c) are drawn by overlapping Figs. 2(a) and (b). In the cases of $RT$ materials other than Sm-Co, the simulated diffraction patterns are similar to Fig. 2, since the lattice parameters are very close to each other. The observed RHEED pattern is in agreement with the calculated pattern of Fig. 2(c-2). An SmCo$_{5}$(1120) epitaxial film consisting of two variants, whose c-axes are lying in-plane and rotated around the film normal by 90° each other, is formed.

Figure 1(c) shows the RHEED pattern of an Sm$_{17}$Co$_{83}$ film deposited on bcc-Cr(211) underlayer at 500 °C$^{10}$. A clear diffraction pattern involving streaks is observed. Figure 2(d) shows the simulated diffraction patterns of (1100) crystals formed in the orientation relationship of

\[ R-T(1\overline{1}00)[1\overline{1}00] \parallel bcc(211)[1\overline{1}1]. \]

The observed pattern corresponds to the calculated pattern of Fig. 2(d-2). An SmCo$_{5}$(1100) single-crystal film with the c-axis parallel to the substrate surface is obtained on the Cr(211) underlayer. The streaks along the perpendicular direction ($RT(1\overline{1}00)$) in the pattern of Fig. 1(c) indicate that the film possesses an atomically flat surface (or is composed of islands with flat terraces). Figure 1(d) shows the RHEED pattern of an Sm$_{17}$Co$_{83}$ film deposited on bcc-Fe$_{50}$Co$_{30}$(211) underlayer at 500 °C$^{10}$. The diffraction pattern also agrees with the simulated result of Fig. 2(d-2), similar to the case of deposition on Cr(211) underlayer. However, streaks along [0001] are involved in the pattern of Fig. 1(d), which indicates that the film includes a small volume of other hexagonal phases. The reason is possibly due to that Co atoms diffuse from the underlayer into the Sm$_{17}$Co$_{83}$ film and the film composition shifts from the SmCo$_{5}$ stoichiometry to a Co-rich composition.

Figures 1(e) and (f) show the RHEED patterns observed for Sm$_{17}$Co$_{83}$ films deposited on fcc-Co(111)/fcc-Cu(111)$^{19}$ double and fcc-Cu(111)$^{21}$ single underlayers at 500 °C. Figures 2(e) and (f), respectively, show the schematic diagrams of diffraction patterns simulated for (0001) crystals formed in the orientation relationships of
The incident electron beam is parallel to (a) [0001], (b), (e) [11-20], (a-3)–(g-3) R2T7 (aR2T7 = 0.50 nm, cR2T7/6 = 0.40 nm), and (a-4)–(g-4) RT3 (aRT3 = 0.50 nm, cRT3/6 = 0.40 nm) structures.

The schematic diagrams of (c) and (g) are drawn by overlapping (a) and (c) and by overlapping (e) and (f), respectively.

Fig. 2 Schematic diagrams of RHEED patterns simulated for (a)–(c) (1120), (d) (1100), and (e)–(g) (0001) crystals with (a-1)–(g-1) $R_7T_7$ ($a_{R_7T_7}$ = 0.42 nm, $c_{R_7T_7}$/2 = 0.40 nm), (a-2)–(g-2) $R_T$ ($a_{RT}$ = 0.50 nm, $c_{RT}$ = 0.40 nm), (a-3)–(g-3) $R_2T_7$ ($a_{R_2T_7}$ = 0.50 nm, $c_{R_2T_7}$/6 = 0.40 nm), and (a-4)–(g-4) $RT_3$ ($a_{RT_3}$ = 0.50 nm, $c_{RT_3}$/6 = 0.40 nm) structures. The incident electron beam is parallel to (a) [0001], (b), (e) [1100], or (d), (f) [1120]. Schematic diagrams of (c) and (g) are drawn by overlapping (a) and (c) and by overlapping (e) and (f), respectively.

$R_7T_7(0001)\parallel fcc(111)[11\bar{2}]$, $R_T(0001)\parallel fcc(0001)[11\bar{2}]$.

The schematic diagrams of Fig. 2(g) are drawn by overlapping Figs. 2(e) and (f). The diffraction patterns observed for the films deposited on Co/Cu and Cu underlayers respectively agree with the calculated patterns of Figs. 2(e-2) and (g-2). An SmCo5(0001) single-crystal film is obtained on the Co/Cu double underlayer, whereas an SmCo5(0001) epitaxial film consisting of two variants, whose orientations are rotated around the film normal by 30° each other, is formed on the Cu single underlayer. The streaks along the perpendicular direction indicate formation of flat surface and/or involvement of other phases. Figure 3 shows the cross-sectional high-resolution transmission electron microscope (HR-TEM) image of an Sm1:Co3...
film deposited on Cu underlayer. A layer-by-layer structure consisting of RT5(0001) and RT5(0002) is clearly recognized. Therefore, the streaks are considered to be corresponding to a formation of flat surface, similar to the case of film deposited on Cr(211) underlayer.

4. Conventional Estimation of Order Degree

In order to characterize RT5 crystal by XRD, it is important to understand the superlattice and fundamental reflections. In the RT5 crystal shown in Fig. 4, the position of R atom, \((x_u, y_u, w_u, z_u)\), is \((0, 0, 0, 0)\), whereas those of T atoms, \((x_v, y_v, w_v, z_v)\), are \((2/3, 1/3, –1, 0)\), \((1/3, 2/3, –1, 0)\), \((1/2, 0, –1/2, 1/2)\), \((0, 1/2, –1/2, 1/2)\), and \((1/2, 1/2, –1, 1/2)\). The structure factor \(F_{hkmb}\) of \((hkmb)\) is expressed as

\[
F_{RT5(hkmb)} = [qf_R + (1- q)f_T] \times \sum u_1 \sum u_2 e^{2\pi i(hx_u + ky_u + kz_u)}
+ [rf_T + (1- r)f_R] \times \sum v_5 \sum v_5 e^{2\pi i(hx_v + ky_v + lz_v)},
\]

where \(q\) and \(r\) are respectively the ratios of R and T atom positions occupied with R and T atoms to the total R and T atom positions \((1/6 \leq q \leq 1, 5/6 \leq r \leq 1)\) and \(f\) is atomic scattering factor. \(q\) and \(r\) are respectively shown by using \(S\) as

\[
q = (1/6)(1 – S) + S = (5/6)S + 1/6, \quad (2)
\]

\[
r = (5/6)(1 – S) + S = (5/6)S + 5/6. \quad (3)
\]

By substituting Eqs. (2) and (3) into (1), \(F_{hkmb}\) is given as

\[
F_{RT5(hkmb)} = [(5S + 1)f_R/6 + (5 – 5S)f_T/6] \times \sum u_1 \sum u_2 e^{2\pi i(hx_u + ky_u + kz_u)}
+ [(S + 5)f_T/6 + (1 – S)f_R/6] \times \sum v_5 \sum v_5 e^{2\pi i(hx_v + ky_v + lz_v)}
= [(5S + 1)f_R/6 + (5 – 5S)f_T/6]
\]

\[
\times e^{(2/3)\pi i(2h + k)} + e^{(2/3)\pi i(h + 2k)}
\]

\[
+ e^{\pi i(h + l)} + e^{\pi i(k + l)} + e^{\pi i(h + k + l)}, \quad (4)
\]

Therefore,

\[
F_{RT5(0001)} = S(f_R – f_T), \quad (5)
\]

\[
F_{RT5(0002)} = f_R + 5f_T. \quad (6)
\]

The reflection from RT5(0001) is superlattice, whereas that from RT5(0002) is fundamental. Thus, the \(S\) value can be calculated by comparing XRD data of RT5(0001) and RT5(0002) reflections.

Reflection intensity \(I\) is proportional to \(F\) and the complex conjugate \((F^*)\), Lorentz-polarization factor \((L)\), and absorption factor \((A)\)

\[
L(\text{in-plane XRD}) = \frac{(1 + \cos^22\theta)\sin^2\theta}{\sin^2\theta}, \quad (7)
\]

\[
L(\text{out-of-plane XRD}) = \frac{(1 + \cos^22\theta)\sin^2\theta}{\sin^2\theta}. \quad (8)
\]

\([A(\text{in-plane XRD}) = \frac{1 - e^{-\mu(2\Lambda/\sin \theta)}\cos \theta}{2\mu}, (9)\]

\([A(\text{out-of-plane XRD}) = \frac{1 - e^{-\mu(2\Lambda/\sin \theta)}\cos \theta}{2\mu}, (10)\]

where \(\mu\) is the linear absorption coefficient of RT5 alloy and \(2\Lambda/\sin \theta\) and \(2\Lambda/\sin \theta\) are the X-ray path lengths. Here, \(A\) corresponds to \(\sigma\) in the case of in-plane XRD, whereas that corresponds to film thickness \((d)\) in the case of out-of-plane XRD. Therefore,
A_{\text{in-plane XRD}} \approx 1/2\mu, \quad (11)
A_{\text{out-of-plane XRD}} = \frac{1 - e^{-\mu(2t/sin\theta)}}{2\mu}. \quad (12)

The intensity ratio of \( RT5(0001) \) to \( RT5(0002) \) reflection is expressed as
\[
\frac{I_{RT5(0001)}}{I_{RT5(0002)}} = S\left[ \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)}} \right]^{1/2} \times \frac{[L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \times [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2}. \quad (13)
\]

Therefore, \( S \) is shown as
\[
S = \left[ \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \right]^{1/2} \times \frac{[L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \times [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2}. \quad (14)
\]

In order to estimate the order degree by using the conventional method of Eq. (14), it is necessary to consider the ratios of \( I_{RT5(0001)}/I_{RT5(0002)} \), \( (f_R + 5f_T)_{RT5(0002)}/(f_R - f_T)_{RT5(0001)} \), \( [L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \), and \( [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2} \).

5. Simplified Estimation of Order Degree

The parameters of \( f_R, f_T, L, \) and \( A \) vary depending on the diffraction angle and the material properties. The approximate diffraction angles of superlattice and fundamental reflections can be respectively evaluated from the \( c \) parameters of bulk \( RT5 \) crystals, \( c_{bulk} \), as
\[
(2\theta_{\chi} \text{ or } 2\theta)_{RT5(0001)} = 2\arcsin(\lambda/2c_{bulk}), \quad (15)
(2\theta_{\chi} \text{ or } 2\theta)_{RT5(0002)} = 2\arcsin(\lambda/c_{bulk}). \quad (16)
\]

When a new parameter of \( C \) is introduced as
\[
C = \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)} \times [L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \times [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2}, \quad (17)
\]
Eq. (14) can be simply expressed as
\[
S = C \times \left[ \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \right]^{1/2}. \quad (18)
\]

Table 1 summarizes the \( c \) parameters of typical magnetic \( RT5 \) materials\(^{32–35} \), the calculated diffraction angles of \( RT5(0001) \) and \( RT5(0002) \) reflections, the \( f \) values of \( R \) and \( T \) atoms at the diffraction angles\(^{36} \), and the \( \mu \) values of \( RT5 \) materials at 8.048 keV (Cu-K\( \alpha \) radiation)\(^{37} \). Table 2 shows the \( C \) values of \( RT5 \) films with the \( c \)-axis lying in the film plane. Table 3 and Fig. 7 show the thickness dependences of \( C \) values of \( RT5 \) films with the \( c \)-axis perpendicular to the substrate surface. Here, it is noted that the \( S \) value is slightly

\[ A_{\text{in-plane XRD}} \approx 1/2\mu, \]
\[ A_{\text{out-of-plane XRD}} = \frac{1 - e^{-\mu(2t/sin\theta)}}{2\mu}. \]

The intensity ratio of \( RT5(0001) \) to \( RT5(0002) \) reflection is expressed as
\[
\frac{I_{RT5(0001)}}{I_{RT5(0002)}} = S\left[ \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)}} \right]^{1/2} \times \frac{[L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \times [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2}. \]

Therefore, \( S \) is shown as
\[
S = \left[ \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \right]^{1/2} \times \frac{[L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \times [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2}. \]

In order to estimate the order degree by using the conventional method of Eq. (14), it is necessary to consider the ratios of \( I_{RT5(0001)}/I_{RT5(0002)} \), \( (f_R + 5f_T)_{RT5(0002)}/(f_R - f_T)_{RT5(0001)} \), \( [L_{RT5(0002)}]/[L_{RT5(0001)}])^{1/2} \), and \( [A_{RT5(0002)}]/[A_{RT5(0001)}])^{1/2} \).

### Table 1

| Material    | \( c \) Parameters (\( \text{Å} \)) | \( \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \) | \( \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)}} \) | \( \frac{[L_{RT5(0002)}]}{[L_{RT5(0001)}]} \) | \( \frac{[A_{RT5(0002)}]}{[A_{RT5(0001)}]} \) |
|-------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|
| SmCo5(0001) | \( c \) (bulk)                  | 8.0 × 10^{-2}                     | 1.94 × 10^{-1}                   | 0.660                             | 0.659586                         |

### Table 2

| Film        | \( C \) Values | \( \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \) | \( \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)}} \) | \( \frac{[L_{RT5(0002)}]}{[L_{RT5(0001)}]} \) | \( \frac{[A_{RT5(0002)}]}{[A_{RT5(0001)}]} \) |
|-------------|----------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|
| SmCo5(0001) | 0.59371        | 0.659586                          | 0.660                             | 0.659586                         |                                  |

### Table 3

| Thickness (\( t \)) | \( C \) Values | \( \frac{I_{RT5(0001)}}{I_{RT5(0002)}} \) | \( \frac{(f_R + 5f_T)_{RT5(0002)}}{(f_R - f_T)_{RT5(0001)}} \) | \( \frac{[L_{RT5(0002)}]}{[L_{RT5(0001)}]} \) | \( \frac{[A_{RT5(0002)}]}{[A_{RT5(0001)}]} \) |
|---------------------|----------------|-----------------------------------|-----------------------------------|-----------------------------------|-----------------------------------|
| 50 Å                | 0.59371        | 0.659586                          | 0.660                             | 0.659586                         |                                  |
Table 1  Crystallographic properties of typical magnetic RT₅ film materials. The c parameters are cited from Refs. 32–35). fR, fT, and μ are calculated by using the data reported in Refs. 36) and 37).

| Material  | c (nm)  | 2θχ or 2θ (deg.) | fR (0001) | fT (0002) | μ (m⁻¹)  |
|-----------|---------|------------------|-----------|-----------|----------|
|           |         | (0001)           | (0002)    |           |          |
| YCo₅     | 0.3969  | 22.40            | 45.72     | 34.04     | 23.79    |
| LaCo₅    | 0.3976  | 22.36            | 45.63     | 50.16     | 23.79    |
| CeCo₅    | 0.4016  | 22.13            | 45.15     | 51.31     | 23.84    |
| PrCo₅    | 0.3988  | 22.29            | 45.49     | 52.57     | 23.81    |
| NdCo₅    | 0.3977  | 22.35            | 45.62     | 53.61     | 23.80    |
| SmCo₅    | 0.3971  | 22.39            | 45.69     | 55.73     | 23.79    |
| GdCo₅    | 0.3967  | 22.41            | 45.74     | 57.62     | 23.78    |
| TbCo₅    | 0.3982  | 22.33            | 45.56     | 58.95     | 23.80    |
| DyCo₅    | 0.3988  | 22.29            | 45.49     | 60.03     | 23.81    |
| HoCo₅    | 0.3996  | 22.25            | 45.39     | 60.88     | 23.82    |
| ErCo₅    | 0.3981  | 22.33            | 45.57     | 62.14     | 23.80    |
| SmFe₅    | 0.414   | 21.5             | 43.7      | 56.1      | 22.9     |
| SmNi₅    | 0.3974  | 22.37            | 45.66     | 55.74     | 24.87    |
| SmCu₅    | 0.4099  | 21.68            | 44.19     | 55.99     | 26.36    |

Table 2  C values of RT₅ film materials with the c-axis lying in the film plane.

| Material  | Thickness (nm) | Thickness, t (nm) |
|-----------|----------------|-------------------|
|           | (0 < t < ∞)    | 5  | 10  | 20  | 50  | 100 | 500 | 1000 | 5000 | 10000 |
| YCo₅      | 5.63           | 3.99 | 3.99 | 4.00 | 4.03 | 4.08 | 4.46 | 4.84 | 5.62 | 5.63  |
| LaCo₅     | 2.43           | 1.72 | 1.72 | 1.73 | 1.75 | 1.77 | 1.97 | 2.16 | 2.43 | 2.43  |
| CrCo₅     | 2.36           | 1.67 | 1.68 | 1.68 | 1.70 | 1.73 | 1.94 | 2.13 | 2.36 | 2.36  |
| PrCo₅     | 2.27           | 1.61 | 1.61 | 1.61 | 1.63 | 1.66 | 1.86 | 2.04 | 2.27 | 2.27  |
| NdCo₅     | 2.20           | 1.56 | 1.56 | 1.57 | 1.58 | 1.61 | 1.81 | 1.98 | 2.20 | 2.20  |
| SmCo₅     | 2.08           | 1.47 | 1.48 | 1.48 | 1.50 | 1.53 | 1.72 | 1.89 | 2.08 | 2.08  |
| GdCo₅     | 1.99           | 1.41 | 1.41 | 1.42 | 1.43 | 1.46 | 1.65 | 1.81 | 1.99 | 1.99  |
| TbCo₅     | 1.94           | 1.37 | 1.37 | 1.38 | 1.39 | 1.42 | 1.59 | 1.75 | 1.93 | 1.94  |
| DyCo₅     | 1.89           | 1.34 | 1.34 | 1.35 | 1.36 | 1.39 | 1.55 | 1.70 | 1.89 | 1.89  |
| HoCo₅     | 1.86           | 1.32 | 1.32 | 1.32 | 1.34 | 1.35 | 1.49 | 1.63 | 1.86 | 1.86  |
| ErCo₅     | 1.81           | 1.28 | 1.29 | 1.29 | 1.30 | 1.32 | 1.45 | 1.59 | 1.81 | 1.81  |
| SmFe₅     | 1.98           | 1.40 | 1.40 | 1.41 | 1.42 | 1.45 | 1.63 | 1.79 | 1.98 | 1.98  |
| SmNi₅     | 2.23           | 1.58 | 1.58 | 1.58 | 1.59 | 1.61 | 1.73 | 1.86 | 2.21 | 2.23  |
| SmCu₅     | 2.46           | 1.74 | 1.75 | 1.75 | 1.76 | 1.78 | 1.90 | 2.03 | 2.44 | 2.46  |

Table 3  C values of RT₅ film materials with the c-axis perpendicular to the substrate surface.

A difference less than a few percentages is considered to exist between the cbulk value and the c value of a prepared film (cfilm). Here, the error of c parameter is defined as

\[ \gamma = \frac{|c_{\text{film}} - c_{\text{bulk}}|}{c_{\text{bulk}}} \times 100. \]
Figure 7 Thickness dependences of C values of RT₅ films with the c-axis perpendicular to the substrate surface. The thickness is shown in logarithmic scale.

Figure 8 Influences of γ on the errors of S values of RT₅ films with the c-axis lying in the film plane estimated by using the C values. Figure 8 shows the influences of γ on the errors of S values of RT₅ films estimated by the simplified method,

\[ \varepsilon = \left( \frac{C_\gamma - C}{C} \right) \times 100 \]  

(20)

The ε values originating from γ less than ±5.00% are estimated to be within ±2.33%. Therefore, the simplified method is useful for estimation of order degree with a high accuracy.

6. Summary

The structures of magnetic RT₅ films with c-axis parallel and perpendicular to the substrate surface are characterized by using a combination of RHEED and XRD. The crystal structure is determined by analyzing a reciprocal lattice map measured by RHEED. The degree of RT₅ ordering is estimated by comparing XRD data of (0001) superlattice and (0002) fundamental reflections. Furthermore, a simplified method of estimating the order degree is proposed for typical magnetic RT₅ materials. The order degree estimated by employing the simplified method agrees in a small error less than a few percentages with the value calculated by using the conventional method. The simplified method is useful for estimation of order degree with a high accuracy.

Acknowledgments A part of this work was supported by JSPS KAKENHI Grant Numbers 25420294 and 26820117 and Chuo University Grant for Special Research.

References

1) K. J. Stmat: Handbook of Ferromagnetic Materials (Elsevier Science B. V., New York, 1988) pp. 131–209.
2) E. E. Fullerton, C. H. Sowers, J. P. Pearson, S. D. Bader, X. Z. Wu, and D. Lederman: Appl. Phys. Lett., 69, 2438 (1996).
3) E. E. Fullerton, J. S. Jiang, C. Rehm, C. H. Sowers, S. D. Bader, J. B. Patel, and X. Z. Wu: Appl. Phys. Lett., 71, 1579 (1997).
4) M. Benaissa, K. M. Krishnan, E. E. Fullerton, and J. S. Jiang: IEEE Trans. Magn., 34, 1204 (1998).
5) M. J. Pechan, N. Teng, J. D. Stewart, J. Z. Hilt, E. E. Fullerton, J. S. Jiang, C. H. Sowers, and S. D. Bader: J. Appl. Phys., 87, 6686
6) A. Singh, R. Tanm, V. Neu, S. Fahler, C. -G. Oertel, W. Skrotzki, L. Schultz, and B. Holzapfel: *J. Appl. Phys.*, 97, 093902 (2005).
7) A. Singh, V. Neu, S. Fahler, L. Schultz, and B. Holzapfel: *J. Magn. Magn. Mater.*, 290–291, 1259 (2005).
8) V. Neu, K. Häfner, and L. Schultz: *J. Magn. Magn. Mater.*, 322, 1613 (2010).
9) T. Yanagawa, Y. Hotta, M. Yamada, M. Ohtake, F. Kirino, and M. Futamoto: *J. Appl. Phys.*, 115, 17A759 (2014).
10) A. Suzuki, T. Yanagawa, Y. Hotta, M. Yamada, M. Ohtake, F. Kirino, and M. Futamoto: Proc. 23rd International Workshop on Rare Earth and Future Permanent Magnets and Their Applications (2014) pp. 114–115.
11) A. K. Patra, V. Neu, S. Fähler, H. Wendrock, and L. Schultz: *J. Appl. Phys.*, 100, 043905 (2006).
12) A. K. Patra, V. Neu, S. Fähler, R. Groetzschel, S. Bedanta, W. Kleemann, and L. Schultz: *Phys. Rev. B*, 75, 184417 (2007).
13) M. Seifert, L. Schultz, and V. Neu: *J. Appl. Phys.*, 106, 073915 (2009).
14) M. Seifert, L. Schultz, R. Schäfer, V. Neu, S. Hankemeier, S. Rößler, R. Frömter, and H. P. Oepen: *New J. Phys.*, 15, 013019 (2013).
15) Y. Hotta, M. Yamada, A. Suzuki, M. Ohtake, M. Futamoto, F. Kirino, and N. Inaba: Digests of the 38th Annual Conference on Magnetics in Japan 2014 (2014) p. 17.
16) Y. Hotta, M. Yamada, T. Yanagawa, M. Ohtake, F. Kirino, N. Inaba, and M. Futamoto: *EPJ Web Conf.*, 75, 04009 (2014).
17) M. Ohtake, Y. Nukaga, F. Kirino, and M. Futamoto: *J. Cryst. Growth*, 311, 2251 (2009).
18) Y. Nukaga, M. Ohtake, F. Kirino, and M. Futamoto: *IEEE Trans. Magn.*, 44, 2891 (2008).
19) M. Ohtake, Y. Nukaga, F. Kirino, and M. Futamoto: *J. Appl. Phys.*, 105, 07C315 (2009).
20) M. Ohtake, Y. Nukaga, F. Kirino, and M. Ohtake: *J. Appl. Phys.*, 107, 09A706 (2010).
21) M. Yamada, T. Yanagawa, Y. Hotta, M. Ohtake, F. Kirino, and M. Futamoto: *EPJ Web Conf.*, 75, 04010 (2014).
22) M. Seifert, L. Schultz, and V. Neu: *J. Appl. Phys.*, 107, 09A711 (2010).
23) M. Yamada, Y. Hotta, A. Suzuki, M. Ohtake, M. Futamoto, F. Kirino, and N. Inaba: Digests of the 38th Annual Conference on Magnetics in Japan 2014 (2014) p. 18.
24) M. Ohtake, O. Yabuhara, Y. Nukaga, F. Kirino, and M. Futamoto: *J. Appl. Phys.*, 107, 09A708 (2010).
25) O. Yabuhara, M. Ohtake, Y. Nukaga, F. Kirino, and M. Futamoto: *J. Magn.: Conf. Ser.*, 200, 082026 (2010).
26) T. Yanagawa, M. Ohtake, F. Kirino, and M. Futamoto: *EPJ Web Conf.*, 40, 06007 (2013).
27) M. Yamada, Y. Hotta, T. Yanagawa, M. Ohtake, F. Kirino, and M. Futamoto: *IEEE Trans. Magn.*, 50, 2101604 (2014).
28) S. Okamoto, N. Kikuchi, O. Kitakami, T. Miyazaki, Y. Shimada, and K. Fukamichi: *Phys. Rev. B*, 66, 024413 (2002).
29) H. Sato, T. Shimatsu, Y. Okazaki, H. Muraoka, H. Aoi, and S. Okamoto: *J. Magn.: Phys.*, 103, 07E114 (2008).
30) M. Ohtake, T. Suzuki, M. Futamoto, F. Kirino, and N. Inaba: *IEEE Trans. Magn.*, 50, 2104004 (2014).
31) B. D. Cullity: *Elements of X-Ray Diffraction* (Addison-Wesley: Reading, Massachusetts, 1956) pp. 104–137.
32) A. E. Dwight: *Trans. Am. Soc. Met.*, 53, 479 (1961).
33) S. E. Haszko: *Trans. Metall. Soc. AIME*, 218, 763 (1960).
34) J. H. Wernick and S. Geller: *Acta Crystallogr.*, 12, 662 (1959).
35) C. Lin, Y. Sun, Z. H. Lou: *J. Appl. Phys.*, 63, 3592 (1988).
36) http://pmsl.planet.sci.kobe-u.ac.jp/~seto/ (As of Oct. 24, 2014).
37) http://physics.nist.gov/PhysRefData/FFast/html/form.html (As of Oct. 24, 2014).

Received Oct. 26, 2014: Revised May 25, 2015: Accepted Jul. 9, 2015