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Preparation of YCo₅ and GdCo₅ Ordered Alloy Epitaxial Thin Films on Cu(111) Underlayer

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Y₁₇Co₈₃ and Gd₁₇Co₈₃ (at. %) alloy thin films are prepared on Cu(111) underlayers epitaxially grown on MgO(111) substrates at a substrate temperature of 500 °C by molecular beam epitaxy. The growth behavior and the film structure are investigated by *in-situ* reflection high-energy electron diffraction and X-ray diffraction. YCo₅ and GdCo₅ ordered alloy crystals epitaxially grow on the Cu underlayers. The epitaxial films consist of two (0001) variants whose orientations are rotated around the film normal by 30° each other. The epitaxial orientation relationships are (YCo₅ or GdCo₅)(0001)[1 $\bar{1}$ 00] || Cu(111)[11 $\bar{2}$] (type A) and (YCo₅ or GdCo₅)(0001)[11 $\bar{2}$] || Cu(111)[11 $\bar{2}$] (type B). The volume ratios of two variants, $V_{\text{type A}}:V_{\text{type B}}$, in YCo₅ and GdCo₅ films are estimated to be 65:35 and 72:28, respectively. The long-range order degrees of YCo₅ and GdCo₅ films are respectively determined to be 0.63 and 0.65. These ordered alloy films show perpendicular magnetic anisotropies reflecting the magnetocrystalline anisotropies of YCo₅ and GdCo₅ crystals.

Key words: YCo₅, GdCo₅, ordered alloy, epitaxial thin film, perpendicular magnetic anisotropy

1. Introduction

Magnetic thin films with the easy magnetization axis perpendicular to the substrate surface and with the uniaxial magnetocrystalline anisotropy energy (K_u) greater than 10^7 erg/cm³ have been investigated for applications like future recording media with the areal density exceeding 1 Tb/in². A bulk SmCo₅ ordered alloy material with RT_5 -type (R : rare earth metal, T : transition metal) structure (Fig. 1) shows K_u of 1.1×10^8 erg/cm³ along the c -axis.¹⁾ (0001)-oriented SmCo₅ polycrystalline²⁻⁷⁾ and epitaxial⁸⁻¹⁰⁾ films have been prepared on Cu,^{2-5,8,9)} Ru,^{6,7,10)} and Ru-Cr⁷⁾ underlayers.

The Sm and Co sites in SmCo₅ structure can be replaced with other R and T elements, respectively. In our previous studies, SmFe₅¹¹⁻¹³⁾ and SmNi₅^{11,14)} ordered alloy epitaxial films were prepared on Cu(111) underlayers by using a molecular beam epitaxy (MBE) system equipped with a reflection high-energy electron diffraction (RHEED) facility. The crystallographic properties during formations of Sm T_5 alloy films can be investigated by *in-situ* RHEED.

Ferromagnetic ordered alloys consisting of Co and R other than Sm with RT_5 structure such as YCo₅ and GdCo₅ also show K_u values greater than 10^7 erg/cm³. However, there are few reports on the formations of (0001)-oriented RCO_5 epitaxial films. In the present study, Y₁₇Co₈₃ and Gd₁₇Co₈₃ (at. %) materials are deposited on Cu(111) underlayers. The growth behavior and the film structure are investigated.

2. Experimental Procedure

Thin films were deposited on polished MgO(111)

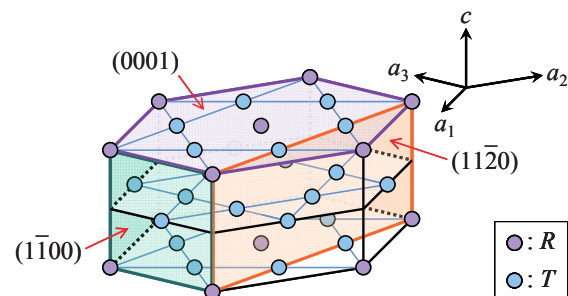


Fig. 1 Schematic diagram of RT_5 structure.

single-crystal substrates by using an MBE system with the base pressure lower than 7×10^{-9} Pa. Pure Y (99.9%) and Gd (99.9%) metals were evaporated by electron beam heating, while pure Co (99.9%) and Cu (99.9999%) materials were evaporated by using Knudsen cells.

The film layer structures were Y₁₇Co₈₃(20 nm)/Cu(20 nm)/MgO(111) and Gd₁₇Co₈₃(20 nm)/Cu(20 nm)/MgO(111). MgO substrates were heated at 500 °C for 1 hour before film formation to obtain clean surfaces. 20-nm-thick Cu underlayers were deposited on the substrates. The epitaxial orientation relationships between Cu underlayer and MgO substrate were Cu(111)[11 $\bar{2}$] || MgO(111)[11 $\bar{2}$] and Cu(111)[$\bar{1}\bar{1}2$] || MgO(111)[11 $\bar{2}$]. Y₁₇Co₈₃ and Gd₁₇Co₈₃ films of 20 nm thickness were formed by co-evaporation of Y and Co or Gd and Co materials. The film composition was confirmed by energy dispersive X-ray spectroscopy to be within 17 ± 2 at. % R ($R = Y$ or Gd), which is nearly the RCO_5 stoichiometry. The substrate temperature during film formation was kept constant at 500 °C.

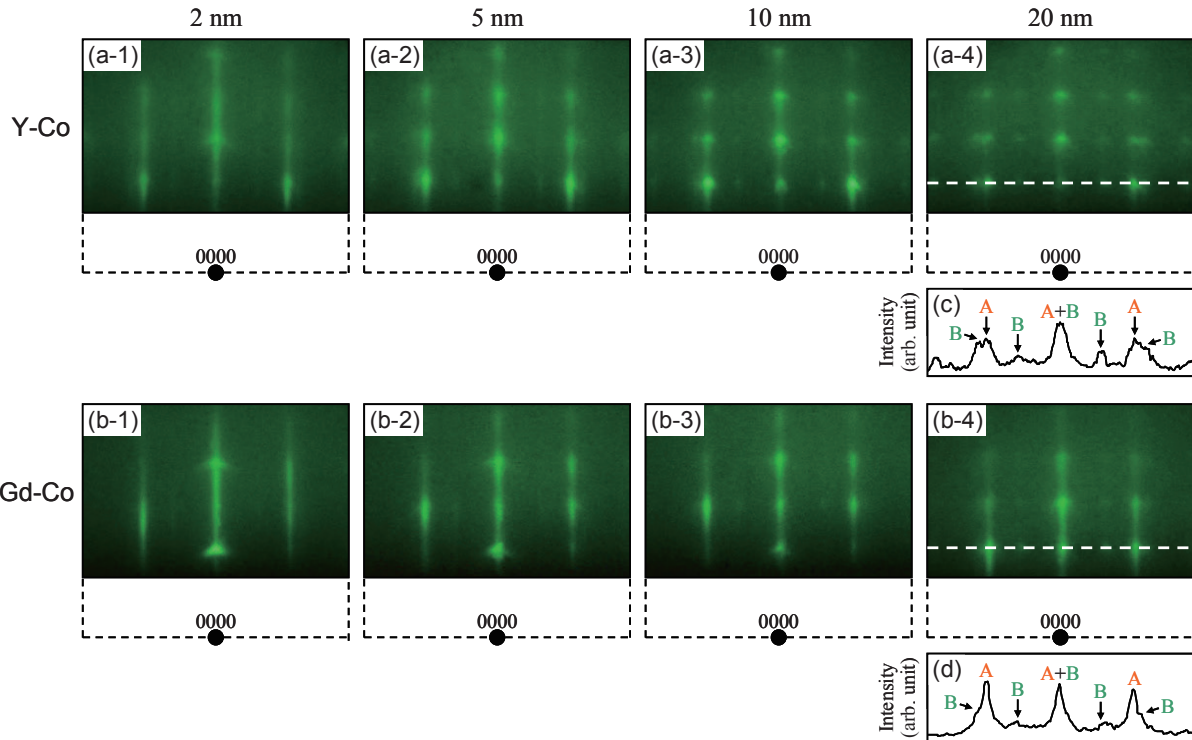


Fig. 2 [(a), (b)] RHEED patterns observed during formations of (a) $Y_{17}Co_{83}$ and (b) $Gd_{17}Co_{83}$ films on Cu(111) underlayers at 500 °C. The film thicknesses are [(a-1), (b-1)] 2, [(a-2), (b-2)] 5, [(a-3), (b-3)] 10, and [(a-4), (b-4)] 20 nm. The incident electron beam is parallel to MgO[112] (\parallel Cu[112], [112]). The intensity profiles of (c) and (d) are measured along the white dotted lines in (a-4) and (b-4), respectively.

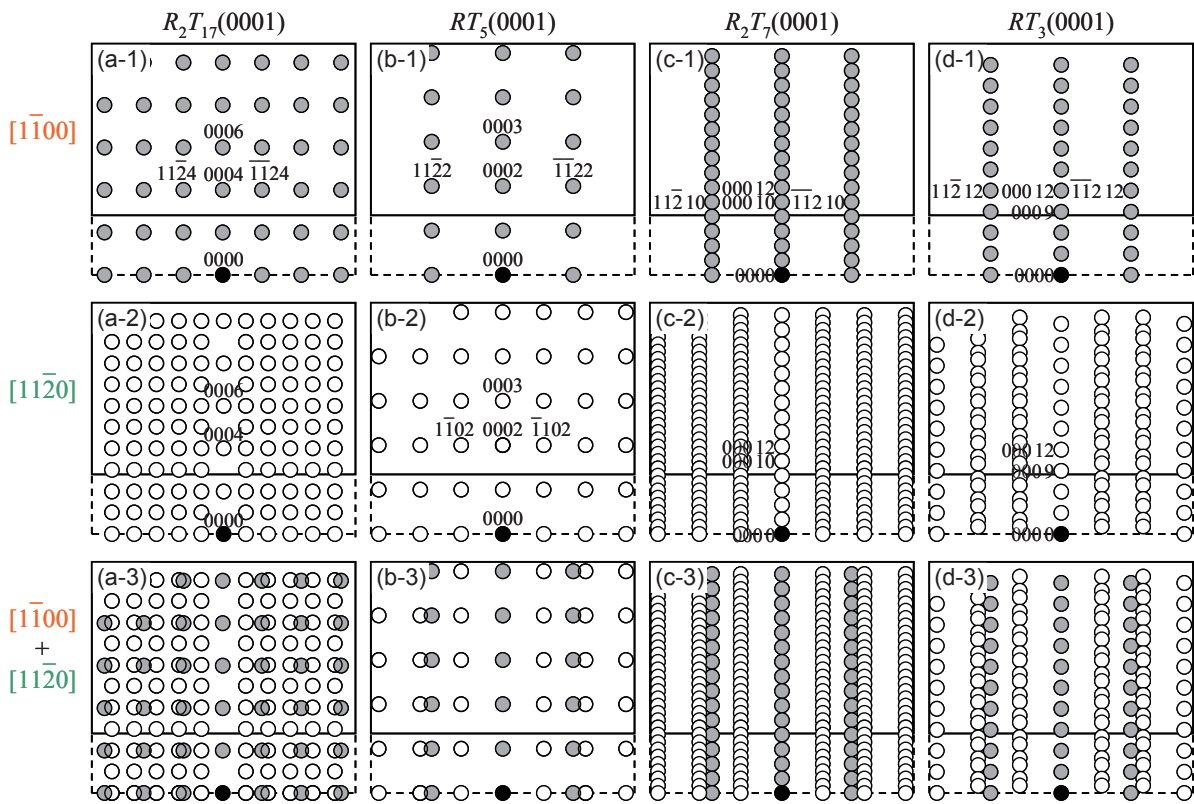


Fig. 3 [(a-1)–(d-1), (a-2)–(d-2)] Schematic diagrams of RHEED patterns simulated for hexagonal (a) R_2T_{17} , (b) RT_5 , (c) R_2T_7 , and (d) RT_3 ordered alloy crystals of (0001) orientation by using the lattice constants of bulk R_2T_{17} ($a/2 = 0.42$ nm, $c/2 = 0.40$ nm), RT_5 ($a = 0.50$ nm, $c = 0.40$ nm), R_2T_7 ($a = 0.50$ nm, $c/9 = 0.40$ nm), and RT_3 ($a = 0.50$ nm, $c/6 = 0.40$ nm) crystals. The incident electron beam is parallel to (a-1)–(d-1) $[1\bar{1}00]$ or (a-2)–(d-2) $[11\bar{2}0]$. Schematic diagrams of (a-3)–(d-3) are drawn by overlapping (a-1)–(d-1) and (a-2)–(d-2), respectively.

The surface structure during film deposition was observed by RHEED. The resulting film structure was investigated by $2\theta/\omega$ scan out-of-plane, $2\theta/\chi/\varphi$ scan in-plane, and β -scan pole-figure X-ray diffractions (XRDs) with Cu-K α radiation ($\lambda = 0.15418$ nm). The magnetization curves were measured by superconducting quantum interference device (SQUID) magnetometry.

3. Results and Discussion

Figures 2(a) and (b) show the RHEED patterns of Y₁₇Co₈₃ and Gd₁₇Co₈₃ films deposited on Cu(111) underlayers observed by making the incident electron beam parallel to MgO[11 $\bar{2}$] (\parallel Cu[11 $\bar{2}$], [1 $\bar{1}$ 2]). Figure 3 shows the schematic diagrams of RHEED patterns simulated for hexagonal R_2T_{17} , RT_5 , R_2T_7 , and RT_3 ordered crystals of (0001) orientation. A clear RHEED pattern corresponding to the diffraction pattern simulated for $RT_5(0001)$ surface [Fig. 3(b-3)] starts to be observed from the beginning of deposition and it remains unchanged until the end of film formation for both films. Y₁₇Co₈₃ and Gd₁₇Co₈₃ epitaxial films with RT_5 ordered structure are obtained. The observed RHEED patterns are analyzed to be an overlap of two reflections, as shown by the symbols, A and B, in the RHEED intensity profiles of Figs. 2(c) and (d). The crystallographic orientation relationships are thus determined as follows,

$$(YCo_5, GdCo_5)(0001)[1\bar{1}00] \parallel Cu(111)[11\bar{2}], [\bar{1}\bar{1}2] \\ \parallel MgO(111)[11\bar{2}], \quad (\text{type A})$$

$$(YCo_5, GdCo_5)(0001)[11\bar{2}0] \parallel Cu(111)[11\bar{2}], [\bar{1}\bar{1}2] \\ \parallel MgO(111)[11\bar{2}]. \quad (\text{type B})$$

The epitaxial films consist of two types of (0001) variant whose orientations are rotated around the film normal by 30° each other, which is similar to the growth of SmCo₅ film on Cu(111) underlayer.^{8,9)}

The lattice misfit values of YCo₅ and GdCo₅ crystals with respect to Cu underlayer are respectively -3.4% and -2.9% in the A-type orientation relationship, whereas those are +11.5% and +12.2% in the B-type relationship. Here, the mismatches are calculated by using the lattice constants of bulk YCo₅ ($a_{YCo_5} = 0.4937$ nm),¹⁵⁾ GdCo₅ ($a_{GdCo_5} = 0.4963$ nm),¹⁵⁾ and Cu ($a_{Cu} = 0.3615$ nm)¹⁶⁾ crystals. Although there are fairly large mismatches in the cases of B-type YCo₅ and GdCo₅ variants, epitaxial growth is taking place. The intensity of RHEED spot from A-type variant is stronger than that from B-type variant for both materials [Figs. 2(c), (d)]. The nucleation of A-type variant with smaller lattice misfits seems to be favored.

In order to investigate the volume ratio of two types of variant, β -scan pole-figure XRD was carried out. Figure 4 shows the β -scan XRD patterns of Y₁₇Co₈₃ and Gd₁₇Co₈₃ films measured by fixing the tilt and

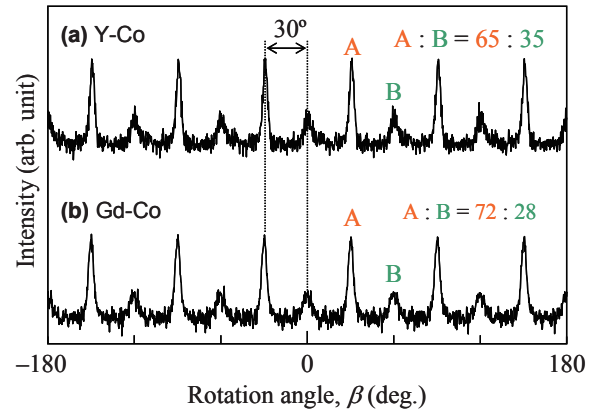


Fig. 4 β -scan pole-figure XRD patterns of (a) Y₁₇Co₈₃ and (b) Gd₁₇Co₈₃ films deposited on Cu(111) underlayers measured by fixing the (α , $2\theta B$) values at (45°, 30.5°). The intensity is shown in linear scale.

diffraction angles of (α , $2\theta B$) at (45°, 30.5°), where YCo₅{1 $\bar{1}$ 01} and GdCo₅{1 $\bar{1}$ 01} reflections are expected to be detectable. Twelve {1 $\bar{1}$ 01} reflections, which originate from the two types of variant, are observed with 30° separation for both films. The volume ratios of A-type to B-type variant in Y₁₇Co₈₃ and Gd₁₇Co₈₃ films are estimated from the integrated intensities of {1 $\bar{1}$ 01} reflections to be 65:35 and 72:28, respectively. It is revealed that the volume ratio of A-type variant is larger than that of B-type variant.

Figures 5(a-1) and (b-1) show the out-of-plane XRD patterns of Y₁₇Co₈₃ and Gd₁₇Co₈₃ films, respectively. $RT_5(0001)$ superlattice and $RT_5(0002)$ fundamental reflections are clearly observed for both films. The out-of-plane XRD confirms the formations of YCo₅ and GdCo₅ ordered phases. Long-range order degree, S , is estimated by comparing the intensities of superlattice and fundamental reflections. The intensity (I) is proportional to structure factor and the complex conjugate (FF^*), Lorentz-polarization factor (L), and absorption factor (A).¹⁷⁾ $F_{(0001)}$ and $F_{(0002)}$ are respectively $S(f_R - f_T)$ and $f_R + 5f_T$,¹⁸⁾ where f is the atomic scattering factor. Therefore, $I_{(0001)}/I_{(0002)}$ is expressed as

$$I_{RT_5(0001)}/I_{RT_5(0002)} = (FF^*LA)_{RT_5(0001)}/(FF^*LA)_{RT_5(0002)} \\ = S^2[(f_R - f_T)^2]_{RT_5(0001)}/[(f_R + 5f_T)^2]_{RT_5(0002)} \\ \times (LA)_{RT_5(0001)}/(LA)_{RT_5(0002)}. \quad (1)$$

By solving this equation, S is given as

$$S = [I_{RT_5(0001)}/I_{RT_5(0002)}]^{1/2} \\ \times (f_R + 5f_T)_{RT_5(0002)}/(f_R - f_T)_{RT_5(0001)} \\ \times [L_{RT_5(0002)}/L_{RT_5(0001)}]^{1/2} \\ \times [A_{RT_5(0002)}/A_{RT_5(0001)}]^{1/2}. \quad (2)$$

The S values of Y₁₇Co₈₃ and Gd₁₇Co₈₃ films are respectively calculated to be 0.63 and 0.65.

Figures 5(a-2) and (b-2) show the in-plane XRD

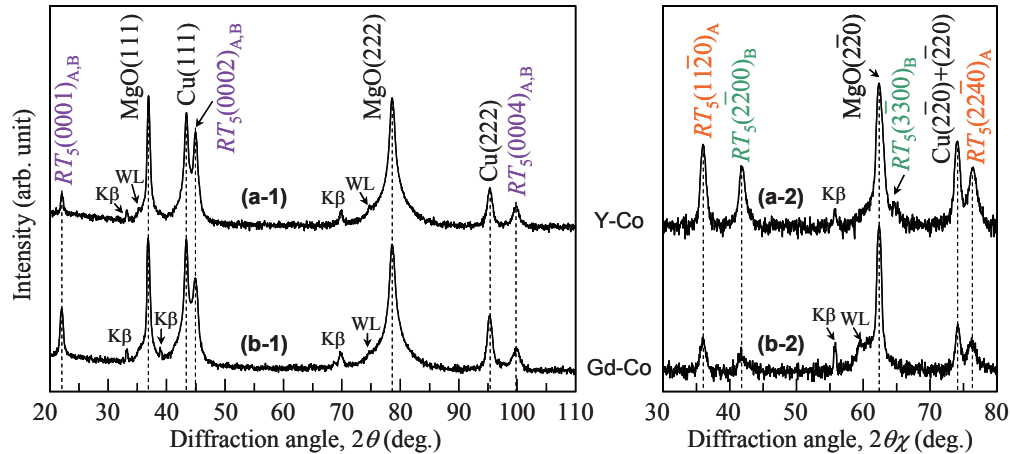


Fig. 5 [(a-1), (b-1)] Out-of-plane and [(a-2), (b-2)] in-plane XRD patterns of (a) $Y_{17}Co_{83}$ and (b) $Gd_{17}Co_{83}$ films deposited on Cu(111) underlayers. The scattering vector of in-plane XRD is parallel to MgO[110]. The small reflections noted as K β and WL are due to Cu-K β and W-La radiations included in the X-ray source, respectively. The intensity is shown in logarithmic scale.

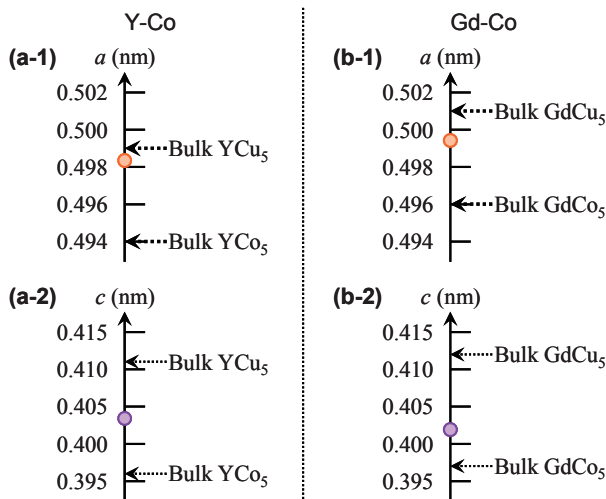


Fig. 6 Lattice constants of [(a-1), (b-1)] a and [(a-2), (b-2)] c of (a) $Y_{17}Co_{83}$ and (b) $Gd_{17}Co_{83}$ films deposited on Cu(111) underlayers.

patterns measured by making the scattering vector parallel to MgO[1 $\bar{1}$ 0]. $RT_5(11\bar{2}0)$ and $RT_5(22\bar{4}0)$ reflections from A-type variant and $RT_5(2\bar{2}00)$ and $RT_5(3\bar{3}00)$ reflections from B-type variant are recognized for both films. The in-plane XRD confirms the epitaxial orientation relationship determined by RHEED.

Figure 6 shows the lattice constants, a and c , of $Y_{17}Co_{83}$ and $Gd_{17}Co_{83}$ films, which are respectively estimated from the peak position angles of $RT_5(22\bar{4}0)$ and $RT_5(0004)$ reflections. Here, the lattice constants of bulk YCo_5 , $GdCo_5$, $Y_{0.8}Cu_{5.4}$, and $GdCu_5$ crystals are cited from Refs. 15, 19, and 20. The a and c values of $Y_{17}Co_{83}$ and $Gd_{17}Co_{83}$ films are between those of bulk YCo_5 and $Y_{0.8}Cu_{5.4}$ crystals and between those of bulk $GdCo_5$ and $GdCu_5$ crystals, respectively. It is reported that Cu atoms of underlayer diffuse into Sm-Co film and

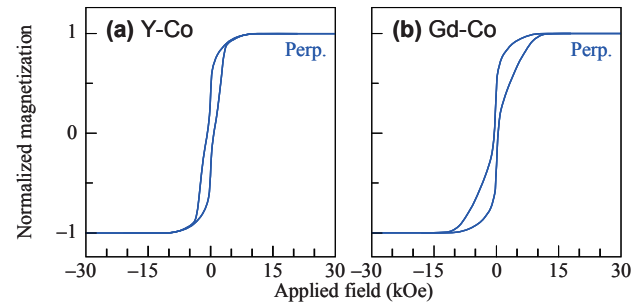


Fig. 7 Magnetization curves of (a) $Y_{17}Co_{83}$ and (b) $Gd_{17}Co_{83}$ films deposited on Cu(111) underlayers.

partially substitute the Co site in $SmCo_5$ structure forming an alloy compound of $Sm(Co,Cu)_5$.^{4,5)} The dissolution of Cu atom into Sm-Co alloy is known to stabilize RT_5 ordered structure.²¹⁻²³⁾ In the present case, Cu atoms are considered to have diffused from the underlayers into the $Y_{17}Co_{83}$ and $Gd_{17}Co_{83}$ films forming alloy compounds of $Y(Co,Cu)_5$ and $Gd(Co,Cu)_5$. It is necessary to confirm the element distribution by using a chemical analysis method.

Figure 7 shows the magnetization curves of $Y_{17}Co_{83}$ and $Gd_{17}Co_{83}$ films measured by applying the magnetic field along the perpendicular direction. These films are easily magnetized, which seems to be reflecting the easy magnetization axis of YCo_5 and $GdCo_5$ ordered alloy crystals.

4. Conclusion

$Y_{17}Co_{83}$ and $Gd_{17}Co_{83}$ thin films are deposited on Cu(111) underlayers at 500 °C. The film growth behavior and the detailed film structure are investigated by RHEED and XRD. YCo_5 and $GdCo_5$ ordered alloy epitaxial films of (0001) orientation are obtained. The films consist of two types of (0001) variant

whose orientations are rotated around the film normal by 30° each other. The S values of YCo₅ and GdCo₅ films are estimated to be 0.63 and 0.65, respectively. Cu atoms are considered to have diffused from the underlayers into the YCo₅ and GdCo₅ films and substitute the Co sites in YCo₅ and GdCo₅ structures forming alloy compounds of Y(Co,Cu)₅ and Gd(Co,Cu)₅. These ordered alloy films show perpendicular magnetic anisotropies reflecting the magnetocrystalline anisotropies of YCo₅ and GdCo₅ crystals.

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Influence of Composition on the Crystal Structure of Fe-Ni Alloy Epitaxial Thin Film Deposited on Cr(211) Underlayer

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Fe_{100-x}Ni_x ($x = 0-100$ at. %) alloy epitaxial films of 10 nm thickness are prepared on Cr(211) underlayers at room temperature by using a radio-frequency magnetron sputtering system. The film growth behavior and the crystallographic properties are investigated by *in-situ* reflection high-energy electron diffraction and pole-figure X-ray diffraction. bcc(211) crystal epitaxially nucleates on the underlayer for the Fe_{100-x}Ni_x films with $x = 0-70$ at. %. The bcc structure is stabilized up to 10 nm thickness for the compositional range of $x = 0-50$ at. %, whereas the Fe₄₀Ni₆₀ and the Fe₃₀Ni₇₀ crystals with bcc structure ($x = 60-70$ at. %) start to transform into fcc structure with increasing the thickness beyond 2 and 5 nm, respectively. The bcc-fcc phase transformation occurs through atomic displacements parallel to bcc(110) and bcc(101) close-packed planes which are 60° canted from the perpendicular direction. The crystallographic orientation relationship is similar to the Kurdjumov-Sachs relationship. When the x value is increased beyond 80 at. %, metastable hcp(1 $\bar{1}$ 00) crystal coexists with bcc(211) crystal. The volume ratio of hcp to bcc crystal increases as the x value increases from 80 to 100 at. %. With increasing the thickness, the hcp crystal also starts to transform into fcc structure through atomic displacement parallel to hcp(0001) close-packed plane, which is similar to the case of bulk phase transformation in the Shoji-Nishiyama relationship.

Key words: Fe-Ni alloy thin film, epitaxial growth, bcc, fcc, hcp, crystal structure, phase transformation

1. Introduction

Thin films of 3d ferromagnetic transition metals or their alloys have been widely studied for applications such as magnetic sensors, magnetic recording media, etc. Recently, magnetic films with metastable structures have attracted much attention, since new possibilities are recognized. For example, magnetic tunnel junction elements prepared by employing Co films with metastable bcc structure have been reported to show high tunnel magnetoresistance ratios¹⁻³. The magnetic and electronic properties are greatly affected by their crystal structures. It is thus important to understand the formation conditions of films with metastable structures.

Fe, Ni, and Fe-Ni alloy materials are typical soft magnetic materials. In the bulk Fe-Ni binary alloy system⁴, there are bcc (A2) and fcc (A1, L1₂) phases. On the contrary, metastable hcp (A3) phase has been recognized for Ni and Fe₂₀Ni₈₀ (at. %) films epitaxially grown on Cr⁵⁻⁹, V⁹, Au^{10,11}, MgO^{12,13}, and Ru¹⁴ materials. Epitaxial thin film growth technique has a possibility in forming films with metastable structures. In our previous studies⁶⁻⁸, Ni and Fe₂₀Ni₈₀ films of 40 nm thickness were sputter-deposited on Cr(211) underlayers at room temperature (RT). The crystallographic property during film formation was investigated by *in-situ* reflection high-energy electron diffraction (RHEED). hcp-Ni and hcp-Fe₂₀Ni₈₀ crystals of (1 $\bar{1}$ 00) orientation nucleated on the underlayers.

However with increasing the thickness, the hcp crystal started to transform into fcc structure and the volume ratio of hcp to transformed fcc crystal increased for both films.

The crystallographic properties are considered to be influenced by the film composition. However, the compositional dependence has not been made clear, yet. In the present study, Fe_{100-x}Ni_x films are prepared on Cr(211) underlayers by varying the composition in the full range, $x = 0-100$ at. %. The growth behavior and the detailed structure property are investigated by *in-situ* RHEED and pole-figure X-ray diffraction (XRD).

2. Experimental Procedure

Thin films were deposited on polished MgO(110) single-crystal substrates by using a radio-frequency (RF) magnetron sputtering system with the base pressure lower than 4×10^{-7} Pa. Fe, Ni, and Cr targets of 3 inch diameter were used and the respective RF powers were fixed at 50, 58, and 40 W. The distance between target and substrate and the Ar gas pressure were kept constant at 150 mm and 0.67 Pa, respectively. Under the conditions, the deposition rate was 0.02 nm/s for all the materials.

MgO substrates were heated at 600 °C for 1 hour before film formation to obtain clean surfaces. 10-nm-thick Cr underlayers were deposited on the substrates at 300 °C. The substrate temperature was used to promote epitaxial growth of Cr underlayer on

e-beam || MgO[1 $\bar{1}$ 0]

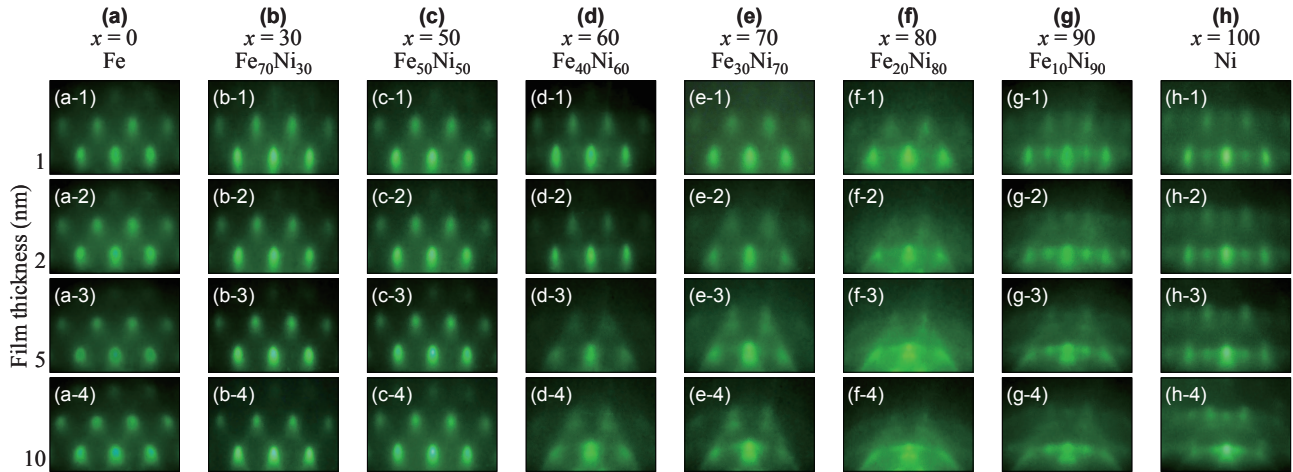


Fig. 1 RHEED patterns observed during formation of (a) Fe, (b) Fe₇₀Ni₃₀, (c) Fe₅₀Ni₅₀, (d) Fe₄₀Ni₆₀, (e) Fe₃₀Ni₇₀, (f) Fe₂₀Ni₈₀, (g) Fe₁₀Ni₉₀, and (h) Ni films on Cr(211) underlayers. The film thicknesses are (a-1)–(h-1) 1 nm, (a-2)–(h-2) 2 nm, (a-3)–(h-3) 5 nm, and (a-4)–(h-4) 10 nm. The incident electron beam is parallel to MgO[1 $\bar{1}$ 0].

e-beam || MgO[001]

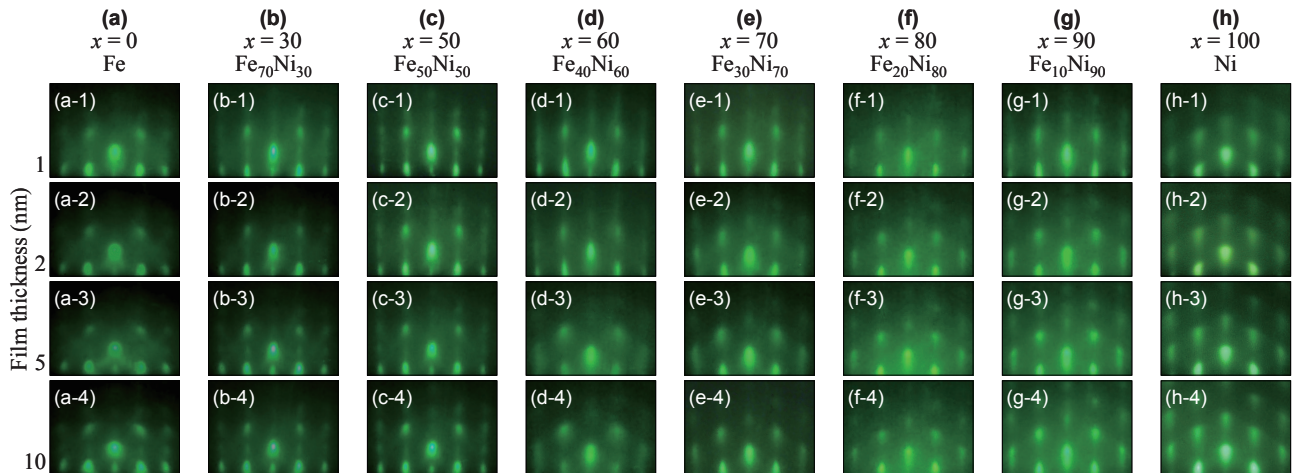


Fig. 2 RHEED patterns observed during formation of (a) Fe, (b) Fe₇₀Ni₃₀, (c) Fe₅₀Ni₅₀, (d) Fe₄₀Ni₆₀, (e) Fe₃₀Ni₇₀, (f) Fe₂₀Ni₈₀, (g) Fe₁₀Ni₉₀, and (h) Ni films on Cr(211) underlayers. The film thicknesses are (a-1)–(h-1) 1 nm, (a-2)–(h-2) 2 nm, (a-3)–(h-3) 5 nm, and (a-4)–(h-4) 10 nm. The incident electron beam is parallel to MgO[001].

MgO(110) substrate. The crystallographic orientation relationship between Cr underlayer and MgO substrate was Cr(211)[$\bar{1}\bar{1}\bar{1}$], [$\bar{1}\bar{1}\bar{1}$] || MgO(110)[$\bar{1}\bar{1}\bar{0}$].⁶⁾ After cooling the samples down to RT, Fe_{100-x}Ni_x ($x = 0, 30, 50, 60, 70, 80, 90, 100$ at. %) alloy films of 10 nm thickness were formed on the Cr underlayers by alternative deposition of Fe and Ni layers. The layer structure was [Fe(1 - δ nm)/Ni(δ nm)]₁₀/Cr(10 nm)/MgO. The alloy formation and the film uniformity were checked by X-ray reflection. The Ni-Fe alloy composition was controlled by changing the δ value ($0 \leq \delta \leq 1$). The film composition was confirmed by energy dispersive X-ray spectroscopy and the errors were less than 3 at. % from the x values.

The film surface structure during growth process was observed by RHEED. The resulting film structure

was investigated by $2\theta/\omega$ -scan out-of-plane and pole-figure XRDs with Cu-K α radiation ($\lambda = 0.15418$ nm). The surface morphology was observed by atomic force microscopy (AFM). The magnetization curves were measured by vibrating sample magnetometry.

3. Results and Discussion

3.1 Structure of Fe_{100-x}Ni_x films ($0 \leq x \leq 50$ at. %)

Figures 1(a)–(c) and 2(a)–(c) show the RHEED patterns observed during formation of Fe_{100-x}Ni_x films with $x = 0$ –50 at. % on the Cr(211) underlayers. Here, two kinds of incident electron beam direction are used to identify the variant structure. The electron beam is parallel to MgO[$\bar{1}\bar{1}\bar{0}$] in Figs. 1(a)–(c), whereas that is parallel to MgO[001] in Figs. 2(a)–(c). MgO[001] is the

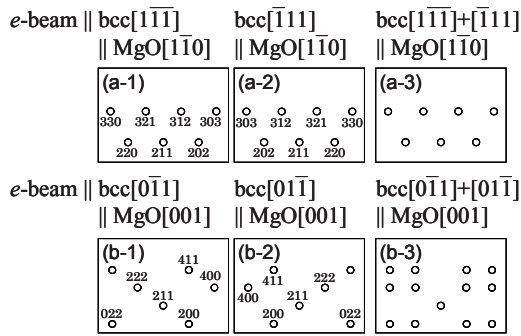


Fig. 3 Schematic diagrams of RHEED patterns simulated for bcc(211) crystal. The incident electron beam is parallel to (a-1) bcc[111], (a-2) bcc[111], (b-1) bcc[011], or (b-2) bcc[011]. Schematic diagrams of (a-3) and (b-3) are drawn by overlapping (a-1) and (a-2) and by overlapping (b-1) and (b-2), respectively.

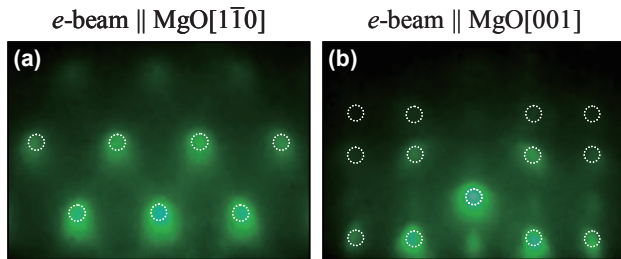


Fig. 4 RHEED patterns observed for a 10-nm-thick Fe film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate [Figs. 1(a-4), 2(a-4)] overlapped with the reflection spots simulated for bcc(211) bi-crystal [Figs. 3(a-3), (b-3)]. The incident electron beam is parallel to (a) bcc[111]+bcc[111] || MgO[110] or (b) bcc[011]+bcc[011] || MgO[001].

direction rotated around the film normal by 90° with respect to MgO[110]. Clear RHEED patterns are observed from the beginnings till the ends of film formation. The films are epitaxially growing on the underlayers for the compositional range.

Possible diffraction patterns were calculated for bcc (A2) crystal. Figures 3(a-1)–(b-2) show the schematic diagrams of diffraction patterns of bcc(211) crystal simulated by making the incident electron beam parallel to bcc[111], bcc[111], bcc[011], or bcc[011]. bcc[111] or bcc[111] is the direction rotated around the film normal by 90° with respect to bcc[011] or bcc[011]. The experimental RHEED data shown in Figs. 1(a)–(c) (e-beam || MgO[110]) are in agreement with both simulation results shown in Fig. 3(a-1) (e-beam || bcc[111]) and Fig. 3(a-2) (e-beam || bcc[111]). However, the observed RHEED patterns shown in Figs. 2(a)–(c) (e-beam || MgO[001]) are partially matching with the calculated patterns of Fig. 3(b-1) (e-beam || bcc[011]) and Fig. 3(b-2) (e-beam || bcc[011]). Therefore, there is a possibility that bcc(211) bi-crystal is formed on the underlayer. Figures 3(a-3) and (b-3) are the schematic diagrams obtained by overlapping Figs. 3(a-1) and (a-2) and by overlapping Figs. 3(b-1) and (b-2), respectively.

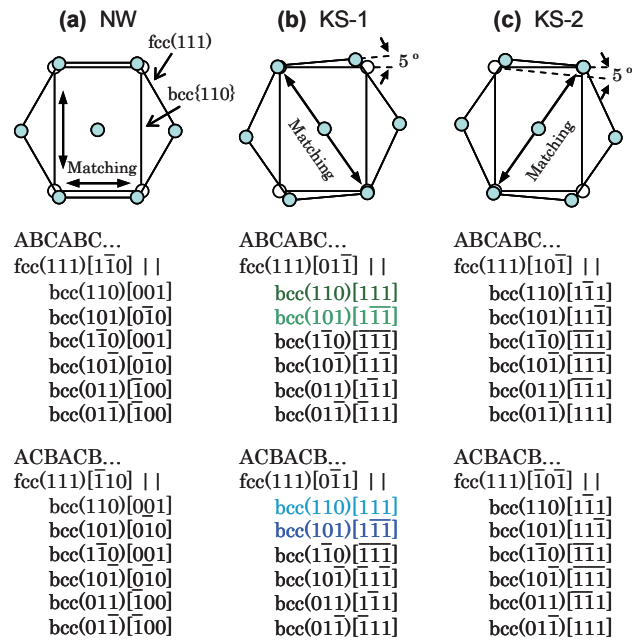


Fig. 5 Schematic diagrams and crystallographic orientation relationships of (a) NW, (b) KS-1, and (c) KS-2.

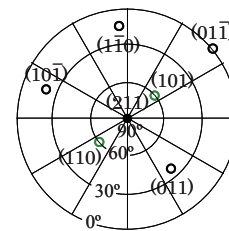


Fig. 6 Pole-figure map showing the configurations of bcc{110} planes around bcc(211) pole.

The observed RHEED patterns agree very well with the overlapped patterns of Figs. 3(a-3) and (b-3), as shown in Fig. 4. The crystallographic orientation relationships between bcc(211) crystal and MgO(110) substrate are thus determined as follows,

$$\text{bcc-Fe}_{100-x}\text{Ni}_x(211)[1\bar{1}\bar{1}] \parallel \text{Cr}(211)[1\bar{1}\bar{1}], [\bar{1}11] \parallel \text{MgO}(110)[1\bar{1}0], \quad (\text{Type I})$$

$$\text{bcc-Fe}_{100-x}\text{Ni}_x(211)[\bar{1}11] \parallel \text{Cr}(211)[1\bar{1}\bar{1}], [\bar{1}11] \parallel \text{MgO}(110)[1\bar{1}0]. \quad (\text{Type II})$$

The films consist of two bcc(211) variants whose orientations are rotated around the film normal by 180° each other.

3.2 Structure of Fe_{100-x}Ni_x films (60 ≤ x ≤ 70 at. %)

Figures 1[(d), (e)] and 2[(d), (e)] show the RHEED patterns observed for Fe₄₀Ni₆₀ and Fe₃₀Ni₇₀ films. RHEED patterns corresponding to the simulated diffraction patterns from bcc(211) bi-crystal [Figs. 3(a-3), (b-3)] are observed for the Fe₄₀Ni₆₀ film thinner than 2

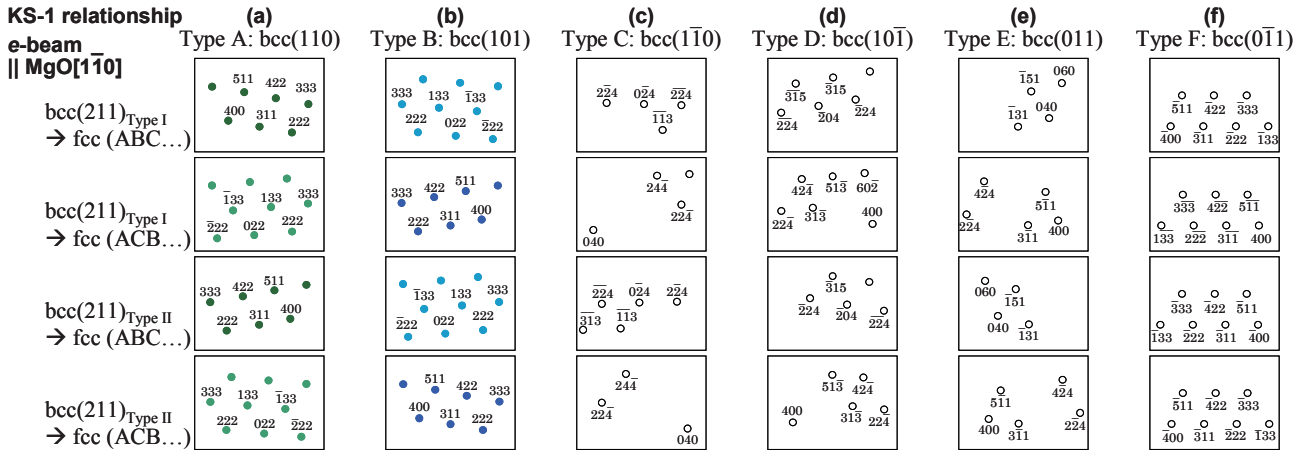


Fig. 7 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along fcc[111] transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(10 $\bar{1}$), (e) bcc(1 $\bar{1}$ 0), and (f) bcc(0 $\bar{1}$ 1) planes in the KS-1 relationship. The incident electron beam is parallel to MgO[1 $\bar{1}$ 0].

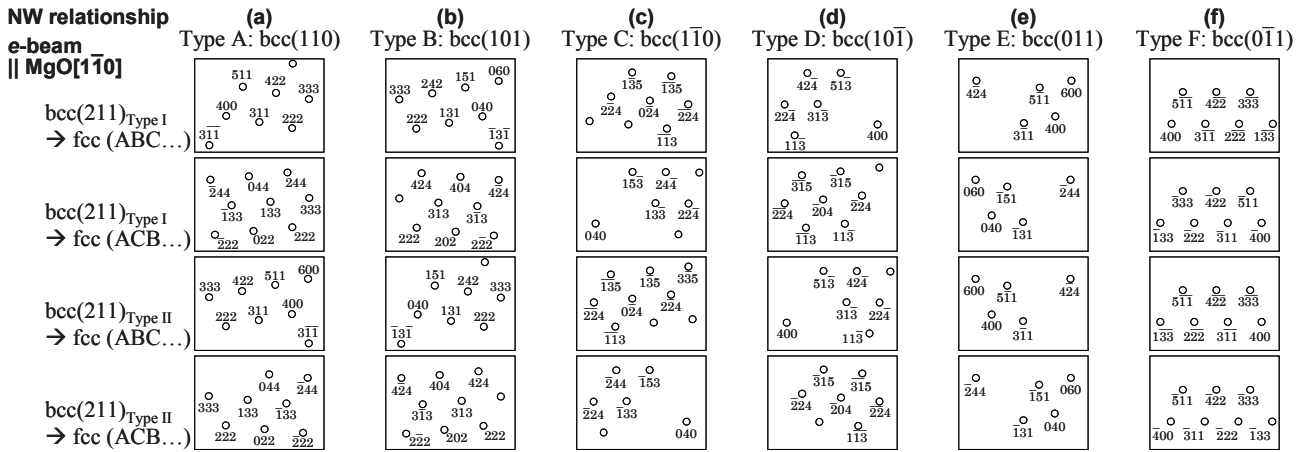


Fig. 8 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along fcc[111] transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(10 $\bar{1}$), (e) bcc(1 $\bar{1}$ 0), and (f) bcc(0 $\bar{1}$ 1) planes in the NW relationship. The incident electron beam is parallel to MgO[110].

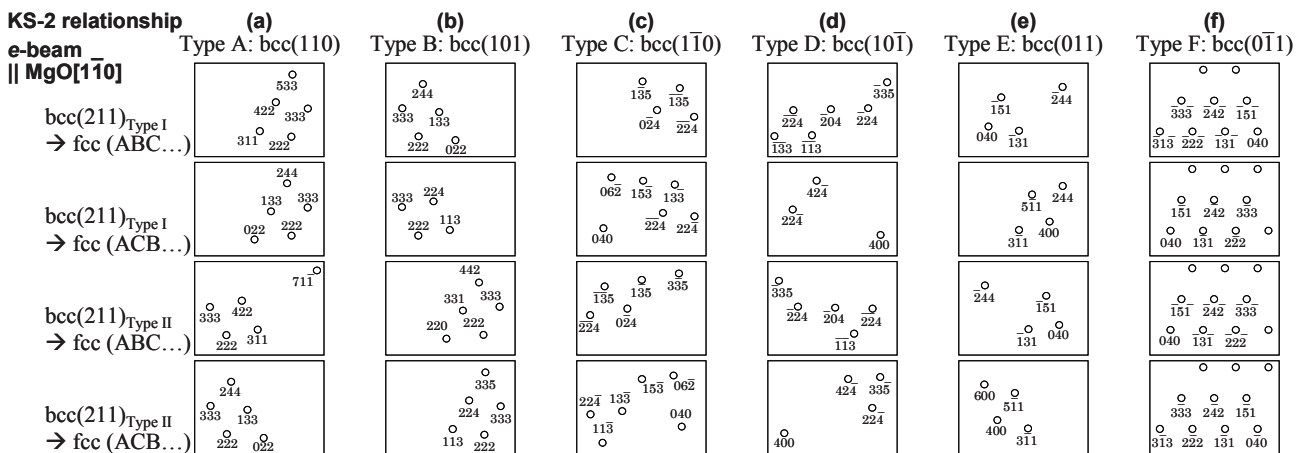


Fig. 9 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along fcc[111] transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(10 $\bar{1}$), (e) bcc(1 $\bar{1}$ 0), and (f) bcc(0 $\bar{1}$ 1) planes in the KS-2 relationship. The incident electron beam is parallel to MgO[110].

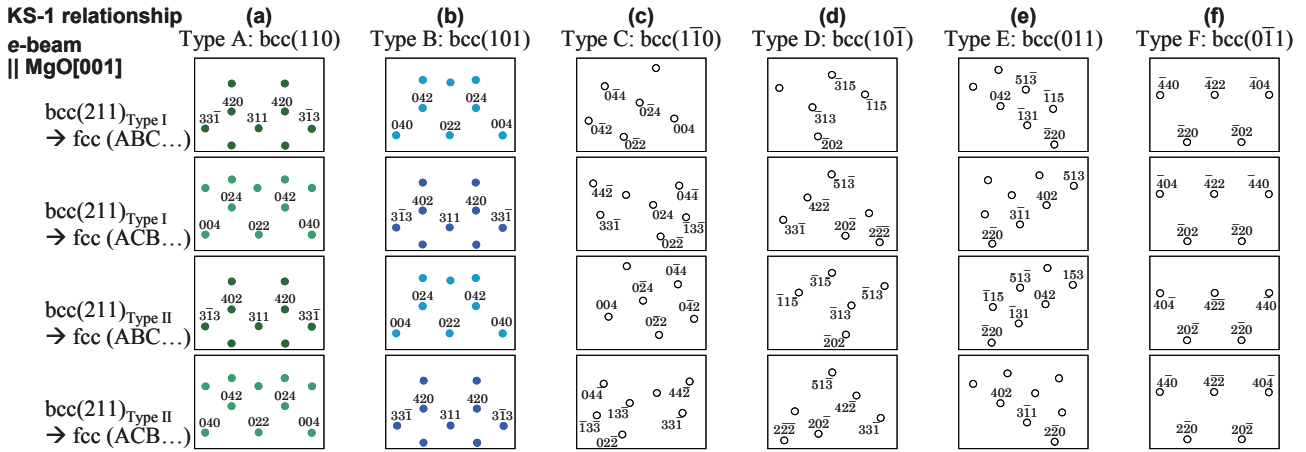


Fig. 10 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along $fcc[111]$ transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(101), (e) bcc(110), and (f) bcc(011) planes in the KS-1 relationship. The incident electron beam is parallel to MgO[001].

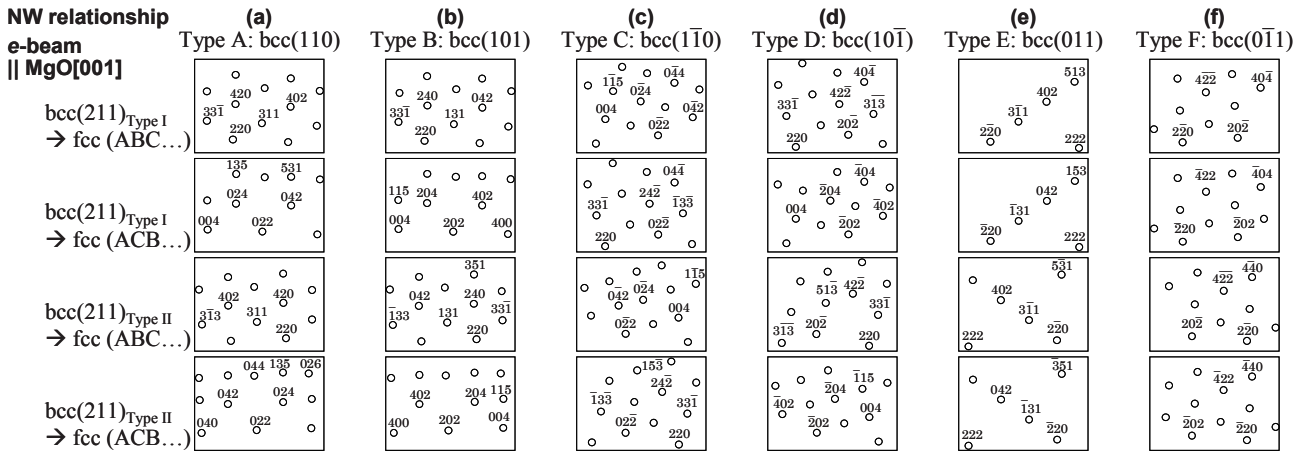


Fig. 11 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along $fcc[111]$ transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(101), (e) bcc(110), and (f) bcc(011) planes in the NW relationship. The incident electron beam is parallel to MgO[001].

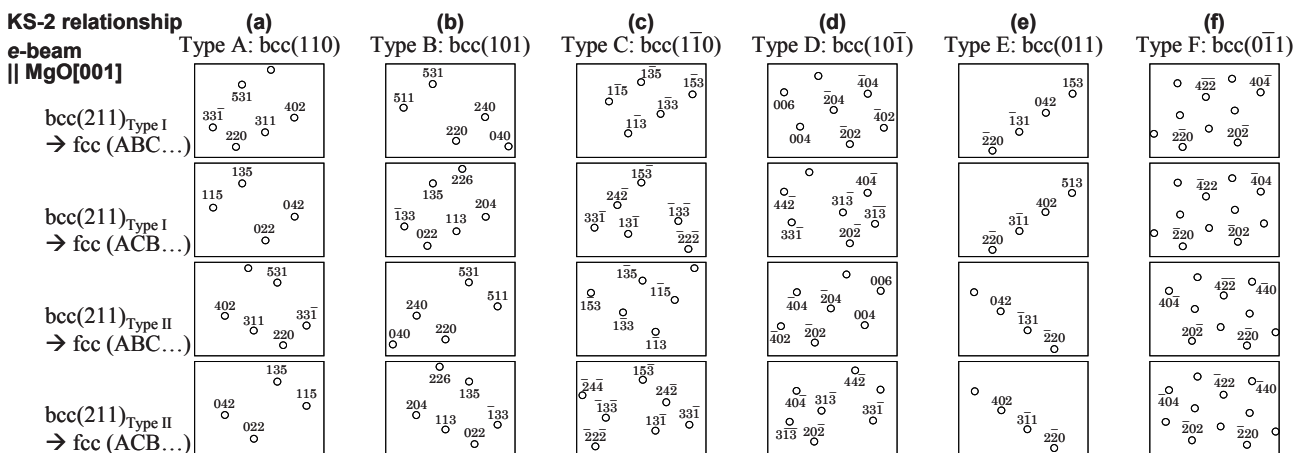
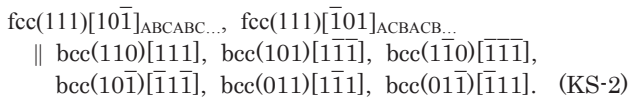
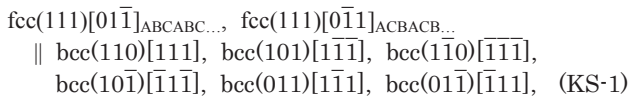
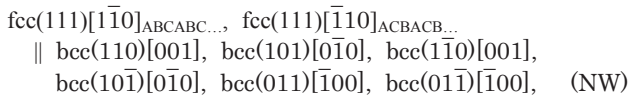


Fig. 12 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of ABCABC... and ACBACB... along $fcc[111]$ transformed from bcc(211) crystals with I- and II-type epitaxial orientation relationships through atomic displacements parallel to (a) bcc(110), (b) bcc(101), (c) bcc(011), (d) bcc(101), (e) bcc(110), and (f) bcc(011) planes in the KS-2 relationship. The incident electron beam is parallel to MgO[001].

nm [Figs. 1(d-1)–(d-2), 2(d-1)–(d-2)] and the Fe₃₀Ni₇₀ film thinner than 1 nm [Figs. 1(e-1), 2(e-1)]. With further increasing the thickness [Figs. 1(d-3)–(d-4), 1(e-2)–(e-4), 2(d-3)–(d-4), 2(e-2)–(e-4)], RHEED spots become broader and diffraction spots other than bcc crystal appear for both films, indicating that phase transformation is taking place. The thickness stability of bcc crystal formation is decreased as the *x* value increases from 50 to 70 at. %.

When a bulk bcc material transforms into fcc structure, there are two possible crystallographic orientation relationships of Nishiyama-Wasserman^{15,16} (NW) and Kurdjumov-Sachs¹⁷ (KS),



The phase transformation occurs through atomic displacements from bcc(110), bcc(101), bcc(011), bcc(10 $\bar{1}$), bcc(1 $\bar{1}$ 0), and bcc(0 $\bar{1}$ 1) close-packed planes to fcc(111) plane, as shown in Fig. 5. The configurations of bcc{110} planes in a bcc crystal with the (211) plane parallel to the substrate surface are shown in the pole-figure map of Fig. 6.

Figures 7–12 show the schematic diagrams of RHEED patterns simulated for fcc (*A1*) crystals with the atomic stacking sequences of ABCABC... and ACBACB... along fcc[111] transformed from bcc(211) crystals with I- and II-type orientation relationships through atomic displacements parallel to bcc{110} planes in the KS-1 [Figs. 7, 10], the NW [Figs. 8, 11], and the KS-2 [Figs. 9, 12] relationships. Figures 13(a)–(f) are drawn by overlapping all the schematic diagrams shown in Figs. 7–12, respectively. Here, the electron beam is parallel to MgO[1 $\bar{1}$ 0] in Figs. 13(a)–(c), whereas that is parallel to MgO[001] in Figs. 13(d)–(f). The RHEED patterns observed for the Fe₄₀Ni₆₀ film thicker than 5 nm [Figs. 1(d-3)–(d-4), 2(d-3)–(d-4)] and the Fe₃₀Ni₇₀ film thicker than 2 nm [Figs. 1(e-2)–(e-4), 2(e-2)–(e-4)] are different from any patterns shown in Fig. 13. The bcc{110} slide planes where the phase transformation occurs are considered to be influenced by the strain caused by accommodation of the lattice misfit between film and underlayer¹⁸.

Figures 14(a)–(f) are obtained by overlapping the schematic diagrams of patterns simulated for fcc crystals transformed through atomic displacements parallel to bcc(110) and bcc(101) planes [Figs. 7[(a), (b)], 8[(a), (b)], 9[(a), (b)], 10[(a), (b)], 11[(a), (b)], 12[(a), (b)]].

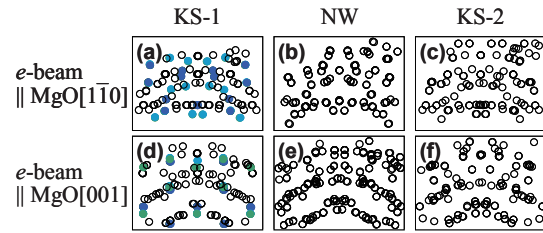


Fig. 13 Schematic diagrams of RHEED patterns simulated for fcc crystals transformed through atomic displacements from bcc(110), bcc(101), bcc(011), bcc(10 $\bar{1}$), bcc(1 $\bar{1}$ 0), and bcc(0 $\bar{1}$ 1) planes to fcc(111) plane in [(a), (d)] the KS-1, [(b), (e)] the NW, and [(c), (f)] the KS-2 relationships. The incident electron beam is parallel to (a)–(c) MgO[1 $\bar{1}$ 0] or (d)–(f) MgO[001]. The schematic diagrams of (a)–(f) are drawn by overlapping all the schematic diagrams shown in Figs. 7–12, respectively.

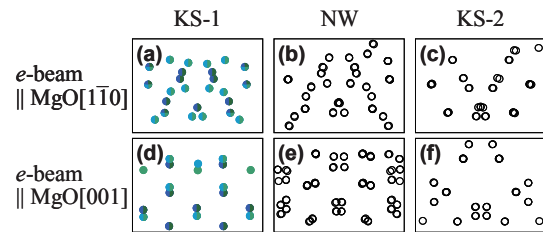


Fig. 14 Schematic diagrams of RHEED patterns simulated for fcc crystals transformed through atomic displacements from bcc(110) and bcc(101) planes to fcc(111) plane in [(a), (d)] the KS-1, [(b), (e)] the NW, and [(c), (f)] the KS-2 relationships. The incident electron beam is parallel to (a)–(c) MgO[110] or (d)–(f) MgO[001]. The schematic diagrams of (a)–(f) are drawn by overlapping all the schematic diagrams shown in Figs. 7[(a), (b)]–12[(a), (b)], respectively.

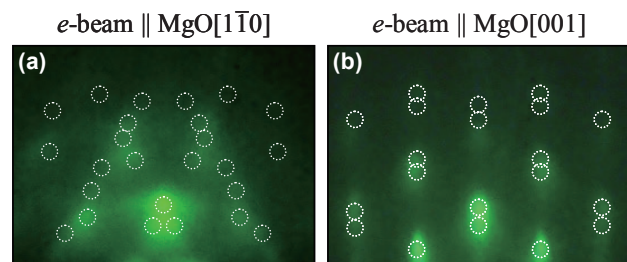


Fig. 15 RHEED patterns observed for a 10-nm-thick Fe₃₀Ni₇₀ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate [Figs. 1(e-4), 2(e-4)] overlapped with the reflection spots simulated for fcc crystals transformed through atomic displacements from bcc(110) and bcc(101) planes to fcc(111) plane in the KS-1 relationship [Figs. 14(a), (d)]. The incident electron beam is parallel to (a) MgO[110] or (b) MgO[001].

The RHEED patterns observed for the Fe₄₀Ni₆₀ film thicker than 5 nm [Figs. 1(d-3)–(d-4), 2(d-3)–(d-4)] and the Fe₃₀Ni₇₀ film thicker than 2 nm [Figs. 1(e-2)–(e-4), 2(e-2)–(e-4)] are in agreement with the simulated patterns of Figs. 14(a) and (d), as shown in Fig. 15. The

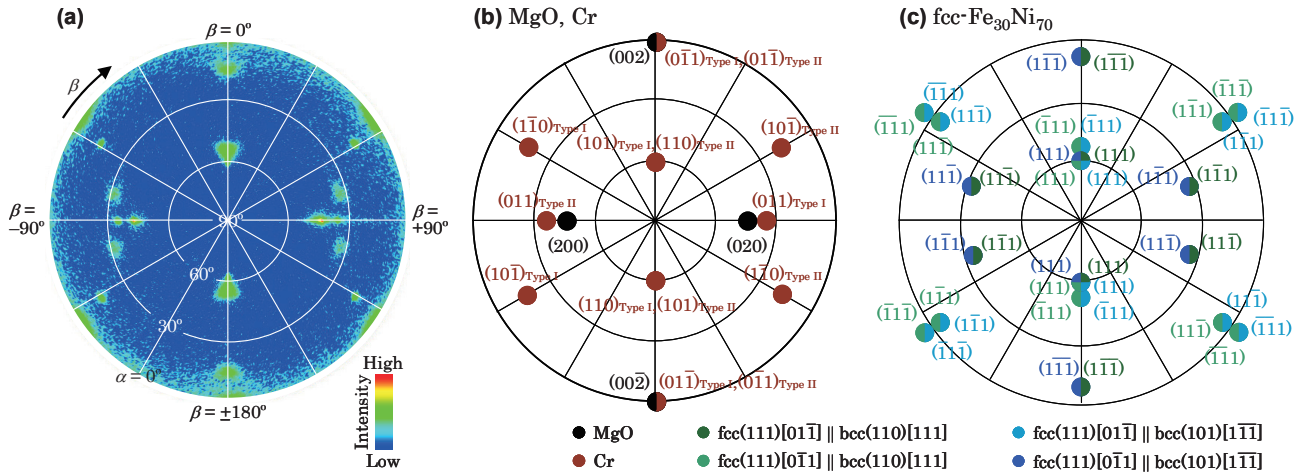


Fig. 16 (a) Pole-figure XRD pattern measured for a 40-nm-thick Fe₃₀Ni₇₀ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate. The diffraction angle of $2\theta B$ is fixed at 44° . The intensity is shown in logarithmic scale. [(b), (c)] Schematic diagrams obtained (b) by overlapping the diffraction patterns simulated for MgO(110) substrate and Cr(211) underlayer and (c) by overlapping the patterns calculated for the fcc crystals transformed from bcc structure in the crystallographic orientation relationships determined by RHEED.

result shows that the phase transformation is not taking place through atomic displacements from six bcc{110} planes but from bcc(110) and bcc(101) planes, which are 60° inclined from the perpendicular direction, in the KS-1 relationship.

When the bcc(211) epitaxial bicrystalline films transform into fcc structure through atomic displacements parallel to bcc(110) and bcc(101) planes in the KS relationship, the films consist of four fcc crystals. fcc(311) is 0.5° tilted from the substrate surface for the fcc crystals with the stacking sequences of ABCABC... and ACBACB... along fcc[111] transformed from bcc(211) crystals with I- and II-type relationships through atomic displacements parallel to bcc(110) and bcc(101) planes, respectively. On the other hand, fcc(011) plane is 5° canted from the substrate surface for the fcc crystals with the stacking sequences of ACBACB... and ABCABC... along fcc[111] transformed from bcc(211) crystals with I- and II-type relationships through atomic displacements parallel to bcc(110) and bcc(101) planes, respectively. These low-index planes of transformed fcc crystals tend to be more inclined from the substrate surface in the cases of NW and KS-2 relationships. Therefore, the phase transformation in the KS-1 relationship seems to be favored.

In order to confirm the phase transformation orientation relationships, pole-figure XRD was carried out. Figure 16(a) shows the pole-figure XRD pattern of a 40-nm-thick Fe₃₀Ni₇₀ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate. Here, the film thickness of 40 nm is employed so that the reflections from transformed fcc crystals are detected more strongly. The diffraction angle of $2\theta B$ is fixed at 44° , where MgO{200}, Cr{110}, and fcc-Fe₃₀Ni₇₀{111} reflections are expected to be detectable. Figures 16(b) and (c), respectively, show the schematic

diagrams obtained by overlapping the diffraction patterns simulated for MgO(110) substrate and Cr(211) underlayer and by overlapping the patterns calculated for the fcc crystals transformed from bcc structure in the crystallographic orientation relationships determined by RHEED. The measured pole-figure XRD pattern of Fig. 16(a) apparently agrees with an overlap of the simulated patterns of Figs. 16(b) and (c). The pole-figure XRD confirms the orientation relationships determined by RHEED.

3.3 Structure of Fe_{100-x}Ni_x films ($80 \leq x \leq 100$ at. %)

Figures 1[(g), (h)] and 2[(g), (h)] show the RHEED patterns observed for Fe₁₀Ni₉₀ and Ni films. Diffraction patterns from hcp(1100) surface shown in the schematic diagrams of Fig. 17 are overlapped with those from bcc(211) surface [Figs. 3(a-3), (b-3)] for the Fe₁₀Ni₉₀ film thinner than 1 nm [Figs. 1(g-1), 2(g-1)] and the Ni film thinner than 5 nm [Figs. 1(h-1)-(h-3), 2(h-1)-(h-3)], as shown in Fig. 18. Therefore, these films are composed of a mixture of hcp(1100) and bcc(211) crystals. Fe₁₀Ni₉₀ and Ni crystals with metastable hcp structure are stabilized through hetero-epitaxial growth on Cr(211) underlayer, similar to the cases of previous studies⁵⁻⁸. The crystallographic orientation relationship between hcp-Fe_{100-x}Ni_x(1100) crystal and Cr underlayer is

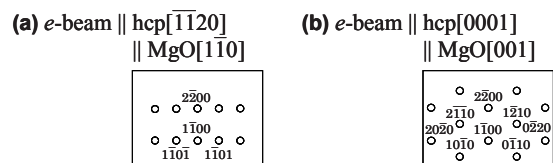


Fig. 17 Schematic diagrams of RHEED patterns simulated for hcp(1100) crystal. The incident electron beam is parallel to (a) hcp[1120] or (b) hcp[0001].

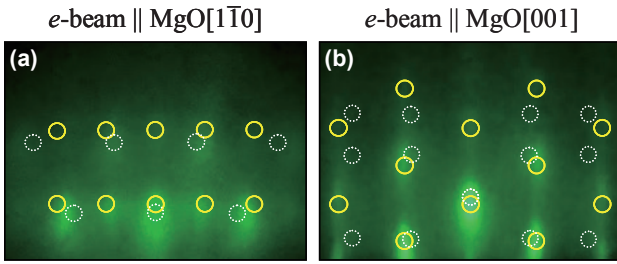


Fig. 18 RHEED patterns observed for a 1-nm-thick $\text{Fe}_{10}\text{Ni}_{90}$ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate [Figs. 1(g-1), 2(g-1)] overlapped with the reflection spots simulated for hcp(1100) [Fig. 17] and bcc(211) [Figs. 3(a-3), (b-3)] crystals. The incident electron beam is parallel to (a) MgO[110] or (b) MgO[001]. The diffraction patterns consisting of yellow solid and white dotted circles correspond to Fig. 17 and Figs. 3[(a-3), (b-3)], respectively.

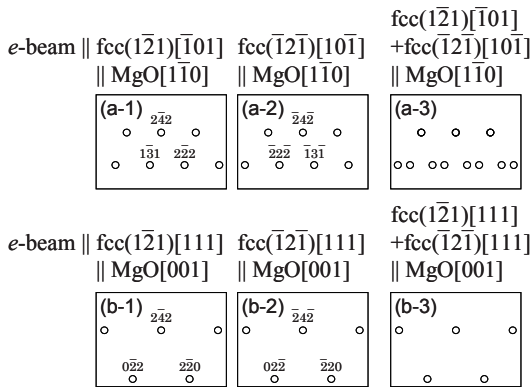


Fig. 19 Schematic diagrams of RHEED patterns simulated for fcc crystals with the atomic stacking sequences of [(a-1), (b-1)] ABCABC... and [(a-2), (b-2)] ACBACB... along fcc[111] transformed from hcp(1100) crystal with III-type orientation relationship through atomic displacement parallel to hcp(0001) plane in the SN relationship. The incident electron beam is parallel to (a) MgO[110] or (b) MgO[001]. Schematic diagrams of (a-3) and (b-3) are drawn by overlapping (a-1) and (a-2) and by overlapping (b-1) and (b-2), respectively.

determined by RHEED as

$$\text{hcp-Fe}_{100-x}\text{Ni}_x(\bar{1}\bar{1}00)[\bar{1}\bar{1}20] \parallel \text{Cr}(211)[1\bar{1}\bar{1}], [\bar{1}11] \parallel \text{MgO}(110)[\bar{1}\bar{1}0]. \quad (\text{Type III})$$

As the thickness further increases [Figs. 1(g-2)–(g-4), 1(h-4), 2(g-2)–(g-4), 2(h-4)], RHEED spots become broader, suggesting that the phase transformations from hcp and bcc crystals to fcc structure are taking place. The details of hcp-fcc phase transformation in hcp(1100) film are shown in our previous paper⁶. The hcp-fcc transformation occurred through atomic displacement from hcp(0001) to fcc(111) plane. The crystallographic orientation relationship between hcp and fcc crystals was

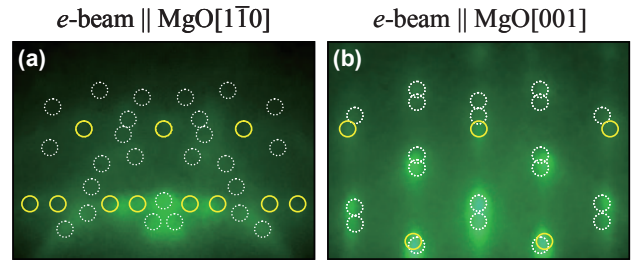


Fig. 20 RHEED patterns observed for a 10-nm-thick $\text{Fe}_{10}\text{Ni}_{90}$ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate [Figs. 1(g-4), 2(g-4)] overlapped with the reflection spots calculated for fcc crystals transformed through atomic displacements from hcp(0001) to fcc(111) plane in the SN relationship [Figs. 19(a-3), (b-3)] and the reflection spots simulated for fcc crystals transformed through atomic displacements from bcc(110) and bcc(101) planes to fcc(111) plane in the KS-1 relationship [Figs. 14(a), (d)]. The incident electron beam is parallel to (a) MgO[110] or (b) MgO[001]. The diffraction patterns consisting of yellow solid and white dotted circles correspond to Fig. 19[(a-3), (b-3)] and Figs. 14[(a-3), (b-3)], respectively.

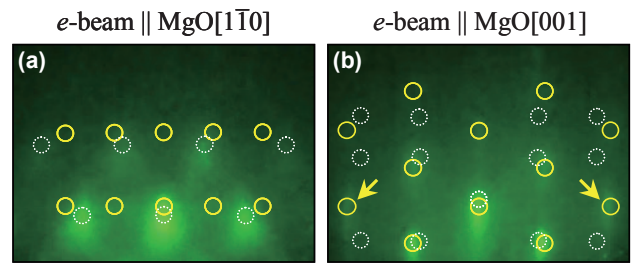


Fig. 21 RHEED patterns observed for a 1-nm-thick $\text{Fe}_{20}\text{Ni}_{80}$ film deposited on Cr(211) underlayer hetero-epitaxially grown on MgO(110) substrate [Figs. 1(f-1), 2(f-1)] overlapped with the reflection spots simulated for hcp(1100) [Fig. 17] and bcc(211) [Figs. 3(a-3), (b-3)] crystals. The incident electron beam is parallel to (a) MgO[110] or (b) MgO[001]. The diffraction patterns consisting of yellow solid and white dotted circles correspond to Fig. 17 and Figs. 3[(a-3), (b-3)], respectively.

$$\text{fcc}(\bar{1}21)[\bar{1}01], \text{fcc}(\bar{1}21)[10\bar{1}] \parallel \text{hcp}(\bar{1}100)[\bar{1}\bar{1}20] \parallel \text{Cr}(211)[1\bar{1}\bar{1}], [\bar{1}11] \parallel \text{MgO}(110)[\bar{1}\bar{1}0], \quad (\text{SN})$$

which was similar to the Shoji-Nishiyama (SN) relationship^{19,20}.

Figure 19 shows the schematic diagrams of RHEED patterns calculated for fcc crystals transformed in the SN relationship. The RHEED patterns observed for the $\text{Fe}_{10}\text{Ni}_{90}$ film thicker than 2 nm [Figs. 1(g-2)–(g-4), 2(g-2)–(g-4)] and the Ni film of 10 nm thickness [Figs. 1(h-4), 2(h-4)] are considered to be consisting of the diffraction patterns of Figs. 14[(a), (d)] and 19[(a-3), (b-3)], as shown in Fig. 20.

Figures 1(f) and 2(f) show the RHEED patterns observed during $\text{Fe}_{20}\text{Ni}_{80}$ film formation. The RHEED

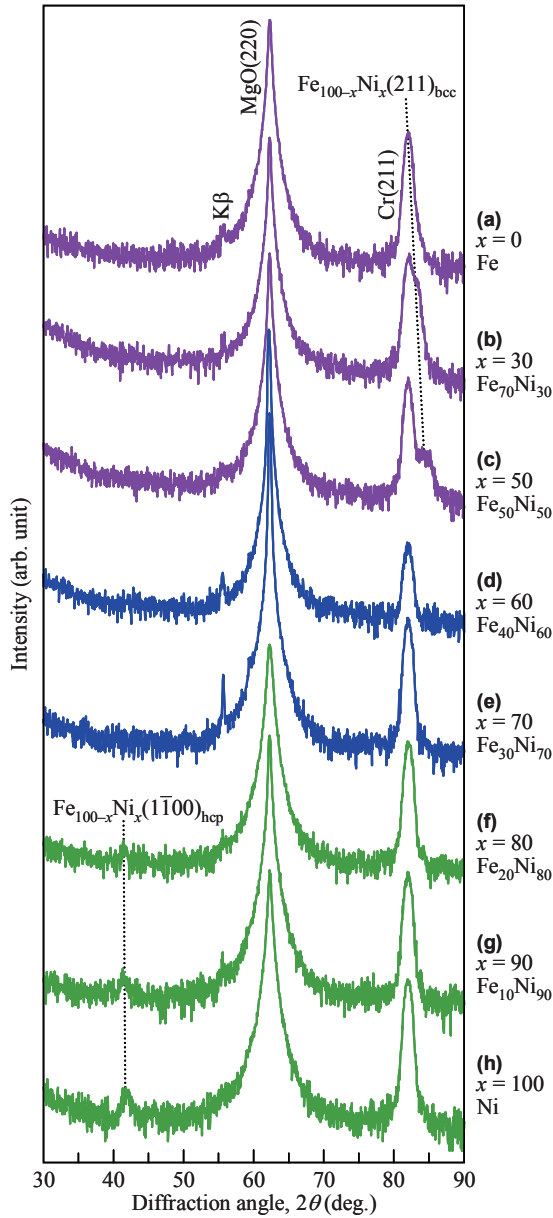


Fig. 22 Out-of-plane XRD patterns of (a) Fe, (b) Fe₇₀Ni₃₀, (c) Fe₅₀Ni₅₀, (d) Fe₄₀Ni₆₀, (e) Fe₃₀Ni₇₀, (f) Fe₂₀Ni₈₀, (g) Fe₁₀Ni₉₀, and (h) Ni films deposited on Cr(211) underlayers hetero-epitaxially grown on MgO(110) substrates. The intensity is shown in logarithmic scale.

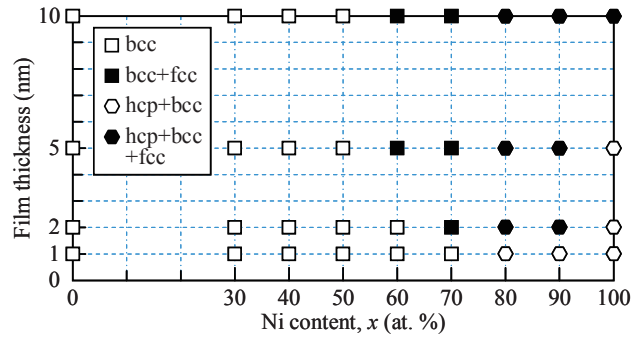


Fig. 23 Phase transformation map of Fe_{100-x}Ni_x films.

patterns observed for the 1-nm-thick film [Figs. 1(f-1), 2(f-1)] seems to involve the reflection spots from hcp(1 $\bar{1}$ 00) crystal as shown in Fig. 21, though the reflection intensity of hcp(1100) crystal is fairly weaker than that of bcc(211) crystal. With increasing the thickness beyond 2 nm [Figs. 1(f-2)–(f-4), 2(f-2)–(f-4)], the RHEED pattern changes to an overlap of diffraction patterns from fcc crystals transformed from bcc and hcp structures [Figs. 14[(a), (d)], 19[(a-3), (b-3)]].

Figure 22 shows the out-of-plane XRD patterns measured for the Fe_{100-x}Ni_x films of 10 nm thickness with different compositions. bcc-Fe_{100-x}Ni_x(211) reflections are observed for the Fe, the Fe₇₀Ni₃₀, and the Fe₅₀Ni₅₀ films ($x = 0-50$ at. %), while those are absent for the Fe₄₀Ni₆₀ and the Fe₃₀Ni₇₀ films ($x = 60-70$ at. %) where the bcc-fcc phase transformation is taking place. hcp-Fe_{100-x}Ni_x(1 $\bar{1}$ 00) reflections are recognized not only for the Fe₁₀Ni₉₀ and the Ni films ($x = 90-100$ at. %) but also for the Fe₂₀Ni₈₀ film ($x = 80$ at. %). As the x value increases from 80 to 100 at. %, the intensity of hcp(1 $\bar{1}$ 00) reflection increases, indicating that the volume of hcp crystal nucleated on Cr(211) underlayer is increased with increasing the Ni content. By considering the RHEED and the XRD data, the phase transformation map of Fe_{100-x}Ni_x films is determined as shown in Fig. 23.

3.4 Surface morphology and magnetic property

Figure 24 shows the AFM images observed for the Fe_{100-x}Ni_x films deposited on Cr(211) underlayers. These

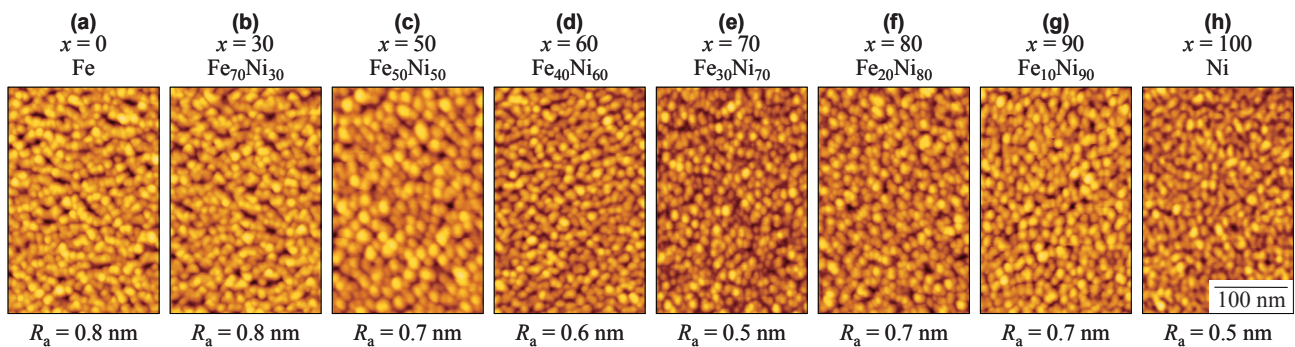


Fig. 24 AFM images observed for (a) Fe, (b) Fe₇₀Ni₃₀, (c) Fe₅₀Ni₅₀, (d) Fe₄₀Ni₆₀, (e) Fe₃₀Ni₇₀, (f) Fe₂₀Ni₈₀, (g) Fe₁₀Ni₉₀, and (h) Ni films deposited on Cr(211) underlayers hetero-epitaxially grown on MgO(110) substrates.

films have flat surfaces with the arithmetical mean roughness (R_a) values lower than 1 nm.

Figure 25 shows the magnetization curves measured for the $\text{Fe}_{100-x}\text{Ni}_x$ films. The magnetic field is applied along $\text{MgO}[1\bar{1}0]$, $\text{MgO}[001]$, or the direction rotated around the film normal by 51° with respect to $\text{MgO}[1\bar{1}0]$, which is the direction obtained by inclining $\text{bcc}[010]$ of $\text{bcc}(211)$ crystals with I- and II-type

orientation relationships to the film plane. The effective easy magnetization direction is observed along $\text{MgO}[001]$ for the films with $x = 0-70$ at. %, whereas that is recognized along $\text{MgO}[1\bar{1}0]$ for the films with $x = 80-100$ at. %. The magnetic properties seem to be delicately influenced by the crystal structure, the film composition, the configuration of transformed fcc crystals, etc.

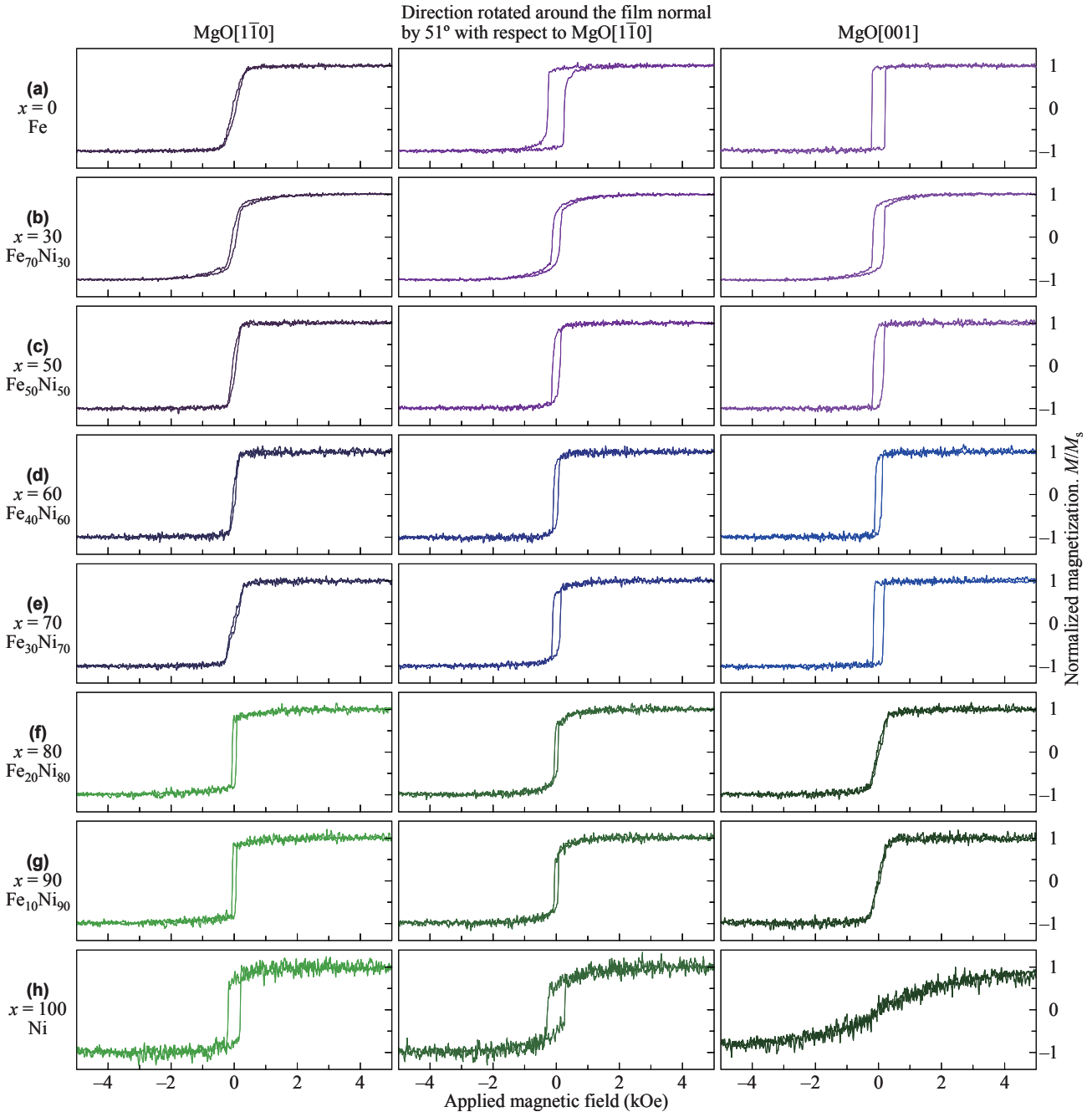


Fig. 25 Magnetization curves measured for (a) Fe, (b) $\text{Fe}_{70}\text{Ni}_{30}$, (c) $\text{Fe}_{50}\text{Ni}_{50}$, (d) $\text{Fe}_{40}\text{Ni}_{60}$, (e) $\text{Fe}_{30}\text{Ni}_{70}$, (f) $\text{Fe}_{20}\text{Ni}_{80}$, (g) $\text{Fe}_{10}\text{Ni}_{90}$, and (h) Ni films deposited on $\text{Cr}(211)$ underlayers hetero-epitaxially grown on $\text{MgO}(110)$ substrates. The magnetic field is applied along $\text{MgO}[1\bar{1}0]$, $\text{MgO}[001]$, or the direction rotated around the film normal by 51° with respect to $\text{MgO}[1\bar{1}0]$, which is the direction obtained by inclining $\text{bcc}[010]$ of $\text{bcc}(211)$ crystals with I- and II-type orientation relationships to the film plane.

4. Conclusion

Fe_{100-x}Ni_x alloy epitaxial films of 10 nm thickness are prepared on Cr(211) underlayers by varying the composition in the full range of $x = 0\text{--}100$ at. %. The film growth behavior and the crystallographic properties are investigated by RHEED and XRD. bcc(211) crystal nucleates on the underlayer for the Fe_{100-x}Ni_x films with $x = 0\text{--}70$ at. %, whereas hcp(1 $\bar{1}$ 00) crystal coexists with bcc(211) crystal in the cases of $x = 80\text{--}100$ at. %. With increasing the thickness, the bcc(211) and/or the hcp(1 $\bar{1}$ 00) crystals start to transform into fcc structure for the films with $x \geq 60$ at. %. As the x value increases, the critical thickness up to which bcc crystal stability is maintained decreases, whereas that for hcp crystal increases. The bcc-fcc and the hcp-fcc phase transformations occur through atomic displacements from bcc(110) and bcc(101) close-packed planes and from hcp(0001) close-packed plane to fcc(111) close-packed plane, respectively. The crystallographic orientation relationships are similar to the Kurdjumov-Sachs and the Shoji-Nishiyama relationships.

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