Epitaxial Growth of Permalloy Thin Films on MgO Single-Crystal Substrates

Mitsuru Ohtake¹, Takahiro Tanaka¹, Katsuki Matsubara¹, Fumiyoshi Kirino² and Masaaki Futamoto¹

1 Faculty of Science and Engineering, Chuo University, 1-13-27 Kasuga, Bunkyo-ku, Tokyo 112-8551, Japan
2 Graduate School of Fine Arts, Tokyo National University of Fine Arts and Music, 12-8 Ueno-koen, Taito-ku, Tokyo 110-8714, Japan
E-mail: ohtake@futamoto.elect.chuo-u.ac.jp

Abstract. Permalloy (Py: Ni – 20 at. % Fe) thin films were prepared on MgO single-crystal substrates of (100), (110), and (111) orientations by molecular beam epitaxy. Py crystals consisting of fcc(100) and hcp(112₀) orientations epitaxially nucleate on MgO(100) substrates. With increasing the substrate temperature, the volume ratio of fcc(100) to hcp(112₀) crystal increases. The metastable hcp(112₀) structure transforms into more stable fcc(110) structure with increasing the film thickness. Py(110)fcc single-crystal films are obtained on MgO(110) substrates, whereas Py films epitaxially grow on MgO(111) substrates with two types of fcc(111) variants whose orientations are rotated around the film normal by 180° each other. X-ray diffraction analysis indicates that the out-of-plane and the in-plane lattice spacings of these fcc–Py films agree within ±0.4% with the values of bulk fcc–Py crystal, suggesting that the strains in the films are very small. High-resolution transmission electron microscopy shows that periodical misfit dislocations are preferentially introduced in the films around the Py/MgO(100) and the Py/MgO(110) interfaces to reduce the lattice mismatches. The magnetic properties are considered to be reflecting the magnetocrystalline anisotropies of bulk fcc–Py and/or metastable hcp–Py crystals and the shape anisotropy caused by the surface undulations.

1. Introduction
Permalloy (Py: Ni – 20 at. % Fe) is a typical soft magnetic material with fcc structure. Py multilayer films combined with thin MgO layers are investigated for magnetic tunneling junction applications like tunneling magnetoresistance heads and magnetic random access memory devices [1]. For such applications, high-quality epitaxial films are strong candidates since the film uniformity and the magnetic anisotropy are well controlled. Epitaxial films are also useful to investigate the basic structural and magnetic properties [2]. However, the film structure and the magnetic properties vary depending on the substrate orientation and the substrate temperature [3]. In the present study, Py films are prepared on MgO single-crystal substrates with different orientations under similar experimental conditions. The effects of substrate orientation and substrate temperature on the film growth, the structure, and the magnetic properties are investigated.

2. Experimental procedure
Thin films were prepared on polished MgO substrates of (100), (110), and (111) orientations at temperatures between 100 and 500 °C by using a molecular beam epitaxy chamber under base pressures lower than 3×10⁻⁸ Pa. Substrates were heated at 500 °C for 1 h in the chamber to obtain clean surfaces. The surface structure was checked by reflection high energy electron diffraction
(RHEED). The RHEED patterns observed for the MgO substrates exhibited Kikuchi patterns [3]. Py films of 40 nm thickness were deposited by e-beam evaporation of an Ni80Fe20 (at. %) source. The Py film compositions were confirmed to be within Ni = 20 ± 2.5 at. % Fe by energy dispersive X-ray spectroscopy. The deposition rate was kept constant at 0.01 nm/s.

The surface structure during film deposition was studied by in-situ RHEED. The structural property was investigated by out-of-plane and in-plane X-ray diffraction with Cu–Kα radiation (λ = 0.15418 nm). The surface morphology and the cross-sectional microstructure were observed by atomic force microscopy (AFM) and high-resolution transmission electron microscopy (HR-TEM), respectively. The magnetization curves were measured by using a vibrating sample magnetometer.

3. Results and discussion
Py epitaxial films were obtained on MgO(100) substrates. Figures 1(a)–(c) show the RHEED patterns observed during Py deposition on MgO(100) substrates heated at different temperatures. In early stages of Py film growth at temperatures between 100 and 500 °C, two kinds of RHEED patterns corresponding to fcc(100) and hcp(1120) textures respectively shown in the spot maps of figures 1(d) and (e) are overlapped, as shown in figures 1(a-1)–(c-1). Thehcp RHEED pattern is analyzed to be an overlap of two hcp(1120) reflections, as shown by the spots, B and C, in figure 1(e). Py epitaxial films consisting of fcc(100) and hcp(1120) crystals are formed on the MgO(100) substrates, where the fcc structure is a stable phase, whereas the hcp structure is a metastable phase. The epitaxial orientation relationships are determined by RHEED as follows,

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\begin{align*}
\text{Py}(100)[001]_{\text{fcc}} & \parallel \text{MgO}(100)[001] \quad \text{(Type A),}
\text{Py}(1120)[0001]_{\text{hcp}} & \parallel \text{MgO}(100)[001] \quad \text{(Type B),}
\text{Py}(1120)[1100]_{\text{hcp}} & \parallel \text{MgO}(100)[001] \quad \text{(Type C).}
\end{align*}
\]

The nucleation behavior is similar to the cases of Co [3] and CoNi [4] film growth on MgO(100). The hcp–Py crystal consists of two types of hcp(1120) variants whose orientations are rotated around the film normal by 90° each other. The lattice mismatch between the Py(100)fcc crystal and the substrate is as large as –15.9%. The mismatch between the Py(1120)hcp crystal and the substrate differs depending on the in-plane direction at the Py(1120)hcp/MgO(100) interface, and the misfit values at the interface along the Py[0001]hcp and the Py[1100]hcp directions are –3.1% and +3.0%, respectively. These values

![Figure 1. RHEED patterns observed for Py films grown on MgO(100) substrates at [(a-1),(a-2)] 100 °C, [(b-1),(b-2)] 300 °C, and [(c-1),(c-2)] 500 °C. The film thicknesses are [(a-1),(b-1),(c-1)] 5 nm and [(a-2),(b-2),(c-2)] 40 nm. RHEED spot maps of (d) fcc(100), (e) hcp(1120), and (f) fcc(110). The incident electron beam is parallel to the MgO[001] direction. The symbols, A–C, respectively correspond to the orientation relationships of Type A–C explained in the text.](image1)

![Figure 2. RHEED patterns observed for 5-nm-thick Py films grown on [(a),(b),(c)] MgO(110) and [(e),(f),(g)] MgO(111) substrates at [(a),(e)] 100 °C, [(b),(f)] 300 °C, and [(c),(g)] 500 °C. RHEED spot maps of (d) fcc(110) and (h) fcc(111). The incident electron beam is parallel to the MgO[001] or the MgO[110] direction. The symbols, D–F, respectively correspond to the orientation relationships of Type D–F explained in the text.](image2)
are calculated from the lattice constants of the bulk MgO crystal \(d_{\text{MgO}} = 0.422 \text{ nm}\) [5], the bulk Py crystal \(d_{\text{fcc-Py}} = 3.55 \text{ nm}\) [6] and from those of the hcp–Py film prepared in the present study, which are estimated from the XRD analysis described later. The metastable hcp–Py crystal is considered to be stabilized through hetero-epitaxial growth. With increasing the film thickness, the peak intensities of \(1210\text{a}_\text{h}, 2020\text{b}, 0220\text{b}, 3030\text{b}, 0330\text{b}, 2110\text{c}, \) and \(1210\text{c}_\text{h}\) hcp RHEED spots become stronger, whereas those of \(2130\text{b}\) and \(1230\text{a}\) spots weaken, as shown in figures 1(a-2) and (b-2). The RHEED data suggest that another pattern overlaps with the hcp(1120) pattern. The pattern of increasing spot intensity is analyzed to be an overlap of four fcc(110) reflections, as shown by the spots, \(B'–1, B'–2, C'–1,\) and \(C'–2,\) in the spot map of figure 1(f). The orientation relationships of

\[
\begin{align*}
\text{fcc}(110)[1\overline{1}] & \parallel \text{fcc}(110)[1\overline{1}] \parallel \text{hcp}(1120)[0001] \text{ (Type B')} \\
\text{fcc}(110)[1\overline{1}] & \parallel \text{fcc}(110)[1\overline{1}] \parallel \text{hcp}(1120)[1100] \text{ (Type C')} \\
\end{align*}
\]

are determined by RHEED. In these configurations, the closed packed plane of hcp crystal is parallel to that of fcc crystal. The metastable hcp structure is apparently transforming into more stable fcc structure possibly by atomic displacement parallel to the hcp(0001) closed packed plane with increasing the film thickness, similar to the case of hcp–Py(1120) film grown on Au(100) [7]. For the Py films prepared at 100 and 300 °C, diffraction patterns of fcc(100) and hcp(1120)+fcc(110) are observed throughout the course of Py deposition, as shown in figures 1(a-2) and (b-2). When the film is prepared at 500 °C, with increasing the film thickness, the RHEED intensity from fcc(100) crystal increases, whereas that from hcp(1120)+fcc(110) crystal decreases. The result shows that the areal ratio of fcc(100) to hcp(1120)+fcc(110) crystal exposed to the film surface increases with increasing the film thickness. The pattern observed for the 40-nm-thick Py film shows only the fcc(100) reflection, as shown in figure 1(e-2). The hcp–Py phase is considered to be stable on the MgO(100) substrates at lower temperatures.

Py epitaxial films were also obtained on MgO(110) and MgO(111) substrates. Figures 2(a)–(c) and (d)–(f) show the RHEED patterns observed for 5-nm-thick Py films grown on MgO(110) and MgO(111) substrates at different temperatures, respectively. Clear RHEED patterns corresponding to fcc(110) and fcc(111) textures shown in the spot maps of figures 2(d) and (g) are observed from the beginning of Py depositions on MgO(110) and MgO(111) substrates, respectively, and they remain unchanged during film formation for all the films. The fcc(111) pattern is analyzed to be an overlap of two fcc(111) reflections, as shown by the spots, \(E\) and \(F,\) in figure 2(g). Py(110)_{fcc} single-crystal films are formed on MgO(110) substrates, whereas Py(111)_{fcc} bi-crystalline films epitaxially grow on MgO(111) substrates. The epitaxial orientation relationships of

\[
\begin{align*}
\text{Py}(110)[10\overline{1}]_{\text{fcc}} \parallel \text{MgO}(110)[\overline{1}0\overline{1}] \text{ (Type D),} \\
\text{Py}(111)[10\overline{1}]_{\text{fcc}} \parallel \text{MgO}(111)[\overline{1}0\overline{1}] \text{ (Type E),} \\
\text{Py}(111)[10\overline{1}]_{\text{fcc}} \parallel \text{MgO}(111)[\overline{1}0\overline{1}] \text{ (Type F),} \\
\end{align*}
\]

are determined by RHEED. The Py(111)_{fcc} film consists of two types of variants whose orientations are rotated around the film normal by 180° each other.

Figure 3 shows the out-of-plane and the in-plane XRD spectra of Py films grown on MgO substrates with different orientations at 300 °C. For the Py film grown on MgO(100) substrate, Py(200)_{fcc} and Py(1120)_{'hcp}(220)_{fcc} out-of-plane XRD reflections and Py(002)_{fcc} and Py(0002)_{hcp}(111)_{fcc} in-plane reflections are clearly observed, as shown in figure 3(a) and (b). The

\[\text{Figure 3. [(a),(c),(e)] Out-of-plane and [(b),(d),(f)] in-plane XRD spectra of Py films grown on [(a),(b)] MgO(100), [(c),(d)] MgO(110), and [(e),(f)] MgO(111) substrates at 300 °C. The scattering vector in-plane XRD is parallel to (b) the MgO[001] or (d),(f) the MgO[100] direction.}\]
follow the growth mode.

FeCo/MgO systems [10–14]. The Py epitaxial film growth on MgO substrates are also considered to exist between the fcc–Py films and the substrates. The film strain is further decreased by employing a substrate temperature dependence on the lattice spacings of fcc–Py crystals estimated from the XRD misfit dislocations are introduced in the film around the interface is reported for the Au, Ag, Cr, Ni, higher substrate temperature. It is known that misfit dislocations are easily introduced in a film around the Py/MgO interfaces, as shown by the RHEED study. The lattice constants of hcp–Py crystal are estimated from the XRD peaks to be (a, c, c/a) = (2d_{hcp(0002)}, 2d_{hcp(0002)}, c/a) = (0.250 nm, 0.408 nm, 1.63). These values are almost in agreement with those of Py(1120) films grown on Au(100) [8] and Cr(100) [9]. For the Py films grown on MgO(110) and MgO(111) substrates, Py(220)_{hcp}, and Py(111)_{hcp} out-of-plane and Py(220)_{hcp} and Py(220)_{hcp} in-plane XRD reflections are recognized, as shown in figures 3(c),(d) and (e),(f), respectively. Similar XRD patterns are observed for the Py films with the respective orientations prepared at 100 and 500 °C. Figure 4 shows the substrate temperature dependence on the lattice spacings of fcc–Py crystals estimated from the XRD peaks. The lattice spacings are in agreement within ±0.4% with those of bulk fcc–Py crystal. The XRD result shows that the strains in the films are very small, though fairly large mismatches of –15.9% exist between the fcc–Py films and the substrates. The film strain is further decreased by employing a higher substrate temperature. It is known that misfit dislocations are easily introduced in a film around the film/substrate interface to reduce the mismatch between immiscible elements when a weak binding force works between the deposited atoms and the substrate atoms. This type of epitaxial growth where misfit dislocations are introduced in the film around the interface is reported for the Au, Ag, Cr, Ni, FeCo/MgO systems [10–14]. The Py epitaxial film growth on MgO substrates are also considered to follow the growth mode.

Figures 5(a) and (d) show the cross-sectional HR-TEM images of Py(100)_{hcp} and Py(110)_{hcp} crystals grown on MgO(100) and MgO(110) substrates at 300 °C, respectively. Atomically sharp boundaries are recognized between the films and the substrates. Periodical misfit dislocations are preferentially introduced in the Py films around the Py/MgO interfaces, as shown by the \( \perp \) marks in figures 5(a) and (b). At the Py(100)_{hcp}/MgO(100) and the Py(110)_{hcp}/MgO(110) interfaces, the Py(200)_{hcp} and the Py(020)_{hcp} planes, respectively, match with the MgO(200) and the MgO(020) planes in integer ratios like 6:5, 7:6, etc. Figures 5(c) and (e) show the matching ratio distribution. When the matching distribution is considered, the effective lattice mismatches at the Py(100)_{hcp}/MgO(100) and the Py(110)_{hcp}/MgO(110) interfaces decrease from –15.9% to –0.5% and –0.3%, respectively. The small calculated misfit values thus coincide with the previous XRD results. The matching ratio distribution is considered to be delicately varied depending on the substrate temperature, since the film strain is further decreased with increasing the substrate temperature. The small strains in the films are apparently realized by the presence of such periodical misfit dislocations. On the contrary, dislocations are expected to be closed loops at the interface for the Py(111)_{hcp}/MgO(111) system, since fccc(111) is the slide plane. The in-plane lattice strains of Py films grown on MgO(111) substrates are very small, when compared with those of Py films grown on MgO(100) and (110) substrates at the respective substrate temperatures, as shown in figures 4(b), (d), and (f). The result supports that strain relaxation of Py(111)_{hcp}/MgO(111) system is faster.

Figure 6 shows the AFM images of Py films grown on MgO substrates of (100), (110), and (111) orientations. The film growth on all the substrates seems to follow the Volmer-Weber (island-growth mode). With increasing the substrate temperature, the \( R_s \) value increases. Py(100)_{hcp} islands with \{111\} and \{110\} facets, Py(110)_{hcp} islands with \{111\} and \{100\} facets, and Py(111)_{hcp} islands with \{001\} and \{110\} facets are respectively recognized for the Py films formed on MgO substrates of (100), (110), and (111) orientations at 500 °C. The orientations of facets were estimated from the cross-
Figure 5. Cross-sectional HR-TEM images of (a) Py(100)fcc and (c) Py(110)fcc crystals grown on MgO(100) and MgO(110) substrates at 300 °C observed along (a) the MgO[001] and (d) the MgO[110] directions, respectively. The \( \perp \) marks show the positions of misfit dislocations. Distributions of lattice matching ratio, (b) Py(200):MgO(200) and (d) Py(020):MgO(020).

Figure 6. AFM images of Py films grown on [(a-1),(a-2),(a-3)] MgO(100), [(b-1),(b-2),(b-3)] MgO(110), and [(c-1),(c-2),(c-3)] MgO(111) substrates at [(a-1),(b-1),(c-1)] 100 °C, [(a-2),(b-2),(c-2)] 300 °C, and [(a-3),(b-3),(c-3)] 500 °C.

sectional profiles of AFM images (not shown here). Figure 7 shows the magnetization curves of Py films grown on MgO substrates of (100), (110), and (111) orientations. The in-plane magnetic anisotropy varied depending on the film orientations. For the Py/MgO(100) system, magnetic properties of Py(100)$_{\text{fcc}}$ and Py(1120)$_{\text{hcp}}$+$\text{(110)$_{\text{fcc}}$}}$ are considered to be overlapped. Py films grown on MgO(100) substrates are easily magnetized when the magnetic field is applied along the MgO[011] direction, whereas the magnetization curves measured along the MgO[001] direction saturate at higher applied fields, as shown in figures 7(a-1)–(a-3). The magnetization behavior is similar to the cases of Co(100)$_{\text{fcc}}$+$\text{(1120)$_{\text{hcp}}$}}$ [3] and CoNi(100)$_{\text{fcc}}$+$\text{(1120)$_{\text{hcp}}$}}$ [4] films epitaxially grown on MgO(100) substrates. The Py films grown on MgO(110) substrates show an uniaxial anisotropy. The easy magnetization direction is observed along the Py[110]$_{\text{fcc}}$ direction. The in-plane magnetization curves are almost isotropic in in-plane measurements for the Py films grown on MgO(111) substrates. The in-plane magnetization property is considered to be reflecting the magnetocrystalline anisotropies of bulk fcc–Py and/or metastable hcp–Py crystals. The film coercivity increases greatly for the films prepared
Figure 7. Magnetization curves of Py films grown on [(a-1),(a-2),(a-3)] MgO(100), [(b-1),(b-2),(b-3)] MgO(110), and [(c-1),(c-2),(c-3)] MgO(110) substrates at [(a-1),(b-1),(c-1)] 100 °C, [(a-2),(b-2),(c-2)] 300 °C, and [(a-3),(b-3),(c-3)] 500 °C.

at 500 °C, possibly due to suppression of domain wall motion by the crevasses in the films existing between large Py islands, as shown in figures 6(a-3), (b-3), and (c-3). The surface morphology influences the coercivities of Py films.

4. Conclusion

The epitaxial growth, the film structure, and the in-plane magnetization properties of Py films on MgO single-crystal substrates of (100), (110), and (111) orientations were investigated. Py epitaxial films consisting of fcc(100) and hcp(1120)+fcc(110) crystals are formed on MgO(100) substrates. Py (110)fcc single-crystal and Py(111)fcc bi-crystalline films epitaxially grow on MgO(110) and MgO(111) substrates, respectively. The lattice spacings of these Py films are in agreement within ±0.4% with the values of bulk Py crystal. The strains in the Py films formed on MgO substrates are very small, which is associated with periodical introduction of misfit dislocations. The in-plane magnetic properties are influenced by the magneto crystalline anisotropies of bulk fcc–Py and/or metastable hcp–Py crystals and the shape anisotropy caused by the surface undulations.

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