Preparation and characterization of Co epitaxial thin films on Al₂O₃(0001) single-crystal substrates

Osamu Yabuhara, Mitsuru Ohtake, Yuri Nukaga, and Masaaki Futamoto

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

E-mail: yabuhara@futamoto.elect.chuo-u.ac.jp

Abstract. Co epitaxial thin films were prepared on Al₂O₃(0001) single-crystal substrates in a substrate temperature range between 50 and 500 °C by ultra high vacuum molecular beam epitaxy. Effects of substrate temperature on the structure and the magnetic properties of the films were investigated. The films grown at temperatures lower than 150 °C consist of fcc–Co(111) crystal. With increasing the substrate temperature, hcp–Co(0001) crystal coexists with the fcc crystal and the volume ratio of hcp to fcc crystal increases. The films prepared at temperatures higher than 250 °C consist primarily of hcp crystal. The film growth seems to follow island-growth mode. The films consisting primarily of hcp crystal show perpendicular magnetic anisotropy. The domain structure and the magnetization properties are influenced by the magnetocrystalline anisotropy and the shape anisotropy caused by the film surface roughness.

1. Introduction

Co and Co-based alloy thin films are widely used as magnetic recording media, magnetic heads, etc. The crystal structure of Co film relies on a stacking sequence of close-packed plane, and it varies between hcp \((K_u=5\times10^6 \text{ erg/cm}^3)\) and fcc \((K_1=6\times10^5 \text{ erg/cm}^3)\) phases depending on the material selected for the substrate and the film growth condition. hcp–Co films have been prepared employing underlayers such as Ru, Ag, and Au [1–3], whereas fcc–Co films have been obtained on Cu underlayers [3]. However, stacking faults are generally recognizable in these films. The crystallographic quality gives strong influence on the magnetic properties [4].

Evaluation of stacking sequence has been performed for Co films by transmission electron microscopy [5,6], perturbed angular correlation [7], in-plane X-ray diffraction (XRD) [4], etc. In order to investigate the structural and magnetic properties, it is useful to employ well-defined epitaxial films, since the film uniformity and the magnetic anisotropy are well controlled. In the present study, Co films were prepared on Al₂O₃(0001) single-crystal substrates. The effects of substrate temperature on the structure and the magnetic properties were investigated.

2. Experimental procedure

Co thin films of 40 nm thickness were prepared on polished α-Al₂O₃(0001) single-crystal substrates at temperatures ranging between 50 and 500 °C by using a molecular beam epitaxy chamber under base pressures lower than \(3\times10^{-8} \text{ Pa}\). Substrates were heated at 500 °C for 1 hour in the UHV chamber to obtain clean surfaces. The surface structure was checked by reflection high energy electron diffraction (RHEED). The RHEED pattern observed for the substrate exhibited Kikuchi patterns, indicating that
Co epitaxial thin films with close-packed planes parallel to the substrate surfaces were obtained on 

3. Results and discussion

Co epitaxial thin films with close-packed planes parallel to the substrate surfaces were obtained on
Al₂O₃(0001) substrates for the investigated temperatures. Figure 1 shows the RHEED patterns observed during Co deposition on Al₂O₃(0001) substrates heated at different temperatures. Clear diffraction patterns are observed from the beginning of Co deposition and they remain unchanged during film formation for all samples, as shown, for example, in figures 1(c)–(e). With increasing the substrate temperature, the diffraction spots and streaks become sharper, suggesting that the strain in the film decreases by employing high substrate temperatures. For the films prepared at temperatures ranging between 50 and 200 °C (figures 1(c) and (d)), the RHEED pattern consists of spots corresponding to fcc(111) texture and streaks along the fcc[111] direction, as shown in the RHEED spot map of figure 1(f). The streaks indicate that the film has atomically flat terraces and/or involves stacking faults along the fcc[111] direction. The epitaxial orientation relationship between the fcc–Co crystal and the substrate (figure 2(a)) is determined by the RHEED observation as follows:

Co(111)[110]fcc || Al₂O₃(0001)[11\overline{1}00] (Type A),
Co(111)[1\overline{1}0]fcc || Al₂O₃(0001)[1\overline{1}00] (Type B).

The fcc–Co crystal consists of two variants whose orientations are rotated around the film normal by 180° each other, similar to the cases of fcc–Co film growth on MgO(111) [8] and SrTiO₃(111) [9]. For the films prepared at temperatures higher than 250 °C (for example, figure 1(e)), the RHEED spots are not recognized and only the streaks along the fcc[111] and/or the hcp[0001] direction are observed, as shown in the spot maps of figures 1(f) and (g). By comparing the patterns observed for the film and the substrate, the hcp–Co crystal is considered to be formed in the following crystallographic relationship (figure 2(b)),

Co(0001)[11\overline{2}0]hcp || Al₂O₃(0001)[1\overline{1}00] (Type C).

When the crystal structure of Al₂O₃ is considered, there are two possibilities of Co crystal nucleation on Al₂O₃(0001). The crystal structure of Al₂O₃ is α–corundum and the (000 l_odd) and the (000 l_even), respectively, consist of Al and O atoms, as shown in figure 2. The atomic arrangements of Al and O layers are rotated around the film normal by 30° each other. One possibility is that Co atoms of fcc(111) or hcp(0001) fit with the Al atoms of Al₂O₃(000 l_odd), where the lattice mismatch is calculated to be 5.6%. Another possibility is that Co atoms fit with the O atoms of Al₂O₃(000 l_even), where the mismatch increase up to 8.9%. The experimental result shows that the arrangement of Co atoms is similar to that of O atoms, as shown in figure 2. Theoretical simulation has shown that 3d ferromagnetic transition metal film growth on Al₂O₃ substrate is strongly influenced by the O atoms located on the substrate surface [10]. The Co film growth on Al₂O₃(0001) is also considered to be influenced by the atomic arrangement of O atoms of Al₂O₃.
apparently increases with increasing the substrate temperature. Al2O3(0001) substrates, respectively. The Co(111) fcc and/or Co(0002) hcp out-of-plane and Co(220) fcc and/or Co(1120) hcp in-plane XRD reflections are clearly observed for all samples. The rocking curves were measured for the out-of-plane and the in-plane XRD peaks, as shown, for example, in the insets of figures 4(c) and (d). Figures 4(c) and (d) show the substrate temperature dependence of the values of full width at half maximum of rocking curves observed for the out-of-plane and the in-plane peaks, \( \Delta \theta_{50} \) and \( \Delta \theta_{50} \). With increasing the substrate temperature, \( \Delta \theta_{50} \) and \( \Delta \theta_{50} \) decrease, indicating that the film strain decreases by employing higher substrate temperatures. The respective in-plane lattice strains are slightly larger than the out-of-plane lattice strains, possibly due to the accommodation of lattice mismatch between the film and the substrate.

Figures 5(a)–(e) show the AFM images. The film growth seems to follow island-growth mode. fcc(111) and/or hcp(0001) islands with hexagonal shape are recognized for the Co epitaxial films prepared in a temperature range between 300 and 500 °C, as shown in figures 5(c)–(e).
shows the substrate temperature dependence on the surface roughness ($R_a$). The $R_a$ does not increase much up to the substrate temperature of 200 °C. With increasing the substrate temperature beyond 250 °C, the $R_a$ starts to increase. Figures 5(g)–(k) show the MFM images. Stripe magnetic domain structure typical for films with perpendicular anisotropy is observed for the films prepared in a temperature range between 250 and 500 °C, as shown in figures 5(j)–(k). Such magnetic domain structure is considered to be reflecting the magnetocrystalline anisotropy of hcp–Co crystal with $c$–axis normal to the substrate surface. Figure 5(l) shows the substrate temperature dependence on magnetic domain width. The domain width slightly increases with increasing the substrate temperature. Figures 5(m)–(q) show the magnetization curves. The films are easily magnetized when the magnetic field is applied parallel to the in-plane direction, and isotropic in-plane magnetization curves are observed. The perpendicular direction is the hard axis due to demagnetizing field, although the crystallographic easy axis of hcp–Co is perpendicular to the substrate surface. Figure 5(r) shows the substrate temperature dependence on the remanent magnetization ($M_r$) and the coercivity ($H_c$) of the films. With increasing the substrate temperature, the out-of-plane $M_r\perp$ increases, whereas the in-plane $M_r//$ decreases. This result is considered to be partially due to the magnetocrystalline anisotropy of hcp–Co crystal and partially due to the shape anisotropy of large islands which form the Co films prepared at higher substrate temperatures. Furthermore, with increasing the substrate temperature beyond 250 °C, the in-plane $H_c//$ starts to increase, possibly due to suppression of domain wall motion by the crevasses in the film existing between Co islands, as shown in figures 5(c)–(e). The crystal structure and the surface roughness influence the magnetic properties of Co epitaxial thin films.

4. Conclusion
Co epitaxial thin films were prepared on Al$_2$O$_3$(0001) single-crystal substrates. The effects of substrate temperature on the structure and the magnetic properties were investigated. The films grown at temperatures lower than 150 °C consist of Co(111)$_{fcc}$ crystal. With increasing the substrate temperature, Co(0001)$_{hcp}$ crystal coexists with fcc crystal and the volume ratio of hcp to fcc crystal increases. The film growth seems to follow island-growth mode. The film strain decreases by employing higher substrate temperatures. Perpendicular magnetic anisotropy is observed for the films consisting primarily of hcp phase. The domain structure and the magnetization properties are influenced by the crystal structure and the surface roughness of the films.

Acknowledgment
A part of this work was supported by NEDO–Japan, MEXT–Japan and STFJ (Science and Technology Foundation of Japan). M. Ohtake is a Research Fellow of the Japan Society for the Promotion of Science.

References
[1] Krishnan K M, Takeuchi T, Hirayama Y and Futamoto M 1994 IEEE Trans. Magn. 30 5115
[2] Gong H, Rao M, Laughlin D E and Lambeth D N 1999 J. Appl. Phys. 85 4699
[3] Ohtake M, Futamoto M, Kirino F, Fujita N and Inaba N 2008 J. Appl. Phys. 103 07B522
[4] Hinata S, Yanagisawa R, Saito S and Takahashi M 2009 J. Appl. Phys. 105 07B718
[5] Wong B Y, Shen Y and Laughlin D E 1993 J. Appl. Phys. 73 07C303
[6] Nukaga Y, Ohtake M, Kirino F and Futamoto M 2010 J. Phys.: Conf. Ser. 200 072071
[7] Barradas N P, Wolters H, Melo A A, Soares J C, da Silva M F, Leal J L, Melo L V and Freitas 1994 J. Appl. Phys. 76 6537
[8] Nukaga Y, Ohtake M, Yabuhara O, Kirino F and Futamoto M 2010 J. Magn. Soc. Jpn. 34
[9] Yabuhara O, Nukaga Y, Ohtake M, Kirino F and Futamoto M 2010 J. Magn. Soc. Jpn. 34 78
[10] Zhang W, Smith J R and Evans A G 2002 Acta Mater. 50 3803
[11] Kachkachi H and Dimian M 2002 Phys. Rev. B 66 174419
[12] Bozorth R M 1993 Ferromagnetism (New York: Wiley–IEEE press) p 264