Homoepitaxial growth of α -Al₂O₃ thin films on atomically stepped sapphire substrates by pulsed laser deposition at room-temperature

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Influence of the atomic-scale surface morphology of sapphire substrates on the epitaxial growth and crystallinity of Al₂O₃ thin films was investigated by pulsed laser deposition (PLD) toward sapphire substrate engineering. Homoepitaxial growth of α -Al₂O₃ thin films on R-plane sapphire (01 $\bar{1}2$) substrates with 0.35-nm-high atomic steps and approximately 20-nm-wide terraces was achieved at room-temperature (approximately 20°C) by PLD. Epitaxial growth of the α -Al₂O₃ thin films was confirmed by in situ reflection high-energy electron diffraction (RHEED). The obtained epitaxial film surface was atomically stepped similar to the sapphire substrate before deposition.

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1. Introduction

Single crystalline α -aluminum oxide (sapphire) substrates have been widely used as insulating substrates for epitaxial growth of semiconductors such as blue-light-emitting GaN or Si (silicon-on-sapphire).^{1)–4)} In addition, atomically stepped sapphire substrates⁵⁾ have been applied for constructing nanostructures of inorganic materials^{6),7)} and controlling molecular orientations of organic materials.⁸⁾ For high-performance mechanical, optical, and microelectronic applications, patterning the sapphire surface with features sized from the micrometer to nanometer scale is strongly desired.

So far, nanometer-scale sapphire surface engineering using top-down methods such as focused-ion beam etching, reactive dry etching, femtosecond laser ablation^{9)–12)} has been extensively studied.^{13)–16)} In the development of atomically regulated electronic devices, low-temperature sapphire substrate surface processing should be effective for obtaining ultra-smooth surfaces and sharp interfaces. Previously, we reported low-temperature homoepitaxial growth of α -Al₂O₃ films on sapphire substrates using an electron-beam-assisted pulsed laser deposition (PLD) process for fabricating nanopatterns on the sapphire substrates.¹⁷⁾ The α -Al₂O₃ thin films were found to grow homoepitaxially on the substrate only in the electron-beam-irradiated region during film deposition, whereas the α -Al₂O₃ thin film was amorphous in the non-irradiated region.¹⁸⁾ In this study, we report the PLD homoepitaxial growth of α -Al₂O₃ thin films at room-temperature (RT) without electron-beam irradiation by atomically controlling components of substrate surface morphology such as atomic step density.

2. Experimental procedure

The two vicinal substrates used in this study were mirror-polished R-plane sapphire (01 $\bar{1}2$) wafers with apparent miscut angles of 0.15 and 1.0°; these substrates were annealed at 1000°C for 3 h in air in order to develop atomically stepped surfaces with approximately 100-nm and 20-nm wide terrace widths, respectively, as previously reported.⁵⁾ Prior to installing a substrate into the ultra-high vacuum PLD chamber, the substrate was cleaned with acetone and ethanol, and then dried in a stream of dry nitrogen.¹⁹⁾ The Al₂O₃ thin films were grown on the atomically stepped sapphire (01 $\bar{1}2$) substrates without heated intentionally in O₂ gas of 1.0×10^{-6} Torr at RT (approximately 20°C) by PLD using an α -Al₂O₃ single-crystal target and a focused KrF excimer laser (wavelength: 248 nm, pulse duration: 20 ns, laser fluence: 2.3 J/cm², and frequency: 3 Hz). The surface morphologies of the films were examined in air using an atomic force microscope (AFM, Hitachi High-Tech Science Corporation, Nanocute) in contact mode. The crystal structure and epitaxial growth of the films were investigated by in situ reflection high-energy electron diffraction (RHEED) and ex situ X-ray diffraction (XRD). The film thickness was evaluated using a stylus surface profiler (KLA Tencor, AlphaStep D-100).

3. Results and discussion

Figure 1 shows AFM surface images of the two different stepped sapphire (01 $\bar{1}2$) substrates. As seen in Fig. 1(a), the ultra-smooth surfaces comprises of approximately 100-nm-wide atomically flat terraces, separated by 0.35-nm-high straight atomic steps. From the observed values of terrace width and step height, the crystallographic off-angle of the vicinal surface shown in Fig. 1(a) was estimated to be approximately 0.2°, which was close to the miscut apparent angle of 0.15°. In

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Fig. 1. AFM topographic images (1000 nm $\times$ 1000 nm) of the atomically stepped sapphire (01 $\bar{1}2$) substrates surface with a terrace width of (a) approximately 100 nm and (b) approximately 20 nm.

Fig. 2. RHEED patterns taken along [2 $\bar{1}\bar{1}0$] of the substrate for (a) the sapphire (01 $\bar{1}2$) substrate with approximately 100-nm wide terrace and (b) the Al₂O₃ thin film grown on the substrate of (a).

contrast, a densely stepped surface appeared for the high-miscut-angle substrate shown in Fig. 1(b). The terrace width of the surface shown in Fig. 1(b) was approximately 20 nm. The crystallographic angle of this narrow-stepped substrate was calculated from the step height and terrace width to be approximately 0.9°, which was close to the miscut apparent angle of 1.0°. It is noted that the density of atomic steps of the vicinal surface shown in Fig. 1(b) was approximately five times larger than that of the substrate surface shown in Fig. 1(a).

Figure 2 reveals the RHEED patterns of (a) the stepped sapphire substrate with an approximately 100-nm-wide terrace recorded before the film growth and (b) the Al₂O₃ film (approximately 80-nm thick) surface grown on the wide terraced substrate shown in Fig. 2(a). The halo RHEED pattern was observed for the film shown in Fig. 2(b). Thus, the amorphous Al₂O₃ film grew on the stepped sapphire (01 $\bar{1}2$) substrate with approximately 100-nm-wide terraces at RT, as shown in Fig. 1(a).

In contrast, as shown in **Fig. 3**, the streaky RHEED patterns along two different directions were observed for the film (approximately 70-nm-thick) grown on the sapphire (01 $\bar{1}2$) substrate with approximately 20-nm-wide terraces at RT, as shown in Fig. 1(b). The RHEED pattern of the Al₂O₃ thin film shown in Fig. 3(a) was identical to that of the sapphire substrate shown in Fig. 2(a). The streaky RHEED pattern of the grown film in Fig. 3 were periodically changed with the same RHEED azimuth angle as that of the sapphire (α -Al₂O₃) substrate, indicating the epitaxial growth of the α -Al₂O₃ thin film. **Figure 4** shows XRD 2θ - θ profiles of the Al₂O₃ film deposited at RT on the sapphire (01 $\bar{1}2$) substrate with approximately 20-nm-wide terraces. There were observed no other peaks except for the diffraction peaks from α -Al₂O₃ (01 $\bar{1}2$) planes of the film and the substrate. These results on RHEED and XRD indicates that the single-crystal α -Al₂O₃ thin film was grown homoepitaxially

Fig. 3. RHEED patterns taken along (a) [2 $\bar{1}\bar{1}0$] and (b) [2 $\bar{2}01$] azimuth of the sapphire substrate for the Al₂O₃ thin film grown at RT on the substrate as shown in Fig. 1(b).

Fig. 4. XRD 2θ - θ profiles of the Al₂O₃ film deposited on the sapphire (01 $\bar{1}2$) substrate with approximately 20-nm-wide terraces at RT.

Fig. 5. AFM topographic image (1000 nm $\times$ 1000 nm) of the Al₂O₃ thin film grown at RT on the narrow-stepped sapphire substrates as shown in Fig. 1(b).

on the narrow-stepped sapphire (01 $\bar{1}2$) substrate. The AFM surface image of the RT-grown α -Al₂O₃ epitaxial thin film on the narrow-stepped sapphire (01 $\bar{1}2$) substrate of Fig. 1(b) is shown in **Fig. 5**. It is seen that the obtained homoepitaxial thin film presented an atomically stepped surface similar to the substrate surface shown in Fig. 1(b).

These results suggest that the terrace width and step density of the atomically stepped sapphire (01 $\bar{1}2$) substrate affect the surface migration of film precursors and nucleation behavior near the step edges of the atomically flat terraces. It is thought that more film precursors would reach the step edges on the narrower terraces, resulting in enhanced nucleation and epitaxial growth near the step edges. Further investigations are necessary to elucidate the detailed growth mechanism of RT-deposited homoepitaxial α -Al₂O₃ thin films on the atomically stepped sapphire substrates.

4. Summary

The influence of the atomic-scale surface morphology of sapphire substrates on PLD homoepitaxial growth and crystallinity of Al_2O_3 thin films was investigated in order to develop a new approach for sapphire substrate engineering. Homoepitaxial $\alpha\text{-Al}_2\text{O}_3$ thin films were obtained at RT on densely stepped sapphire (01 $\bar{1}$ 2) substrates with 0.35-nm-high atomic steps and approximately 20-nm wide terraces. The obtained film surface was atomically stepped similar to the sapphire substrate before deposition. In contrast, amorphous Al_2O_3 thin films were obtained on the stepped sapphire (01 $\bar{1}$ 2) substrates with a wider terrace width of approximately 100 nm. These results suggest a correlation between the RT homoepitaxial growth behavior of Al_2O_3 thin films and atomic step density of the sapphire substrates.

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