

Anomalous Growth-mode Transition During the Initial Growth Stages of SrRuO₃ Thin Films on Atomically Flat Single-terminated SrTiO₃ (111) Surface

Jaewan Chang* and Sang-Koog Kim

Research Center for Spin Dynamics and Spin-Wave Devices, Seoul National University and Nanospinics Laboratory, Department of Materials Science and Engineering, Seoul National University, Seoul 151-744, Republic of Korea

1. Introduction

SrRuO₃ (SRO), a conducting ferromagnetic oxide, has attracted much attention due to its favorable integration with other functional oxides into heteroepitaxial-multilayered electronic devices[1], which would require atomically sharp interfaces. Although oxide films grown on SrTiO₃ (STO) (001) substrates have been extensively studied[2-4], the growth of SRO films on STO (111) is lacking, due the difficulty of achieving an atomically well-defined, single-terminated STO (111) surface. However, the recent successful fabrication of such a STO (111) surface by Chang *et al.* [5] has opened the door to layer-by-layer growth of SRO thin films on STO (111). In this presentation, we report on observations of the layer-by-layer growth and growth-mode transitions of SrRuO₃ thin films on atomically flat Ti⁴⁺ single-terminated SrTiO₃(111) substrates.

2. Experiments

SRO thin films were grown on Ti⁴⁺ single-terminated STO (111) substrates by pulsed layer deposition (PLD). The specifics of the preparation of single-terminated STO (111) substrates were described in our previous paper[5]. For stoichiometric ablation, a KrF excimer laser (wavelength $\lambda=248$ nm) was focused on a sintered SRO target with a fluence of 2.5 J/cm². The oxygen pressure was maintained at 100 mTorr. During the SRO film deposition, the growth modes were monitored by in situ high-pressure reflection high-energy electron diffraction (RHEED). A 20 keV electron beam with an incidence angle of around 1° was directed along[1-10] azimuth. The RHEED diffraction patterns and intensity profiles in specular reflection geometry were recorded using a charge-coupled device camera with an acquisition software. The surface morphologies of the grown SRO films were probed by atomic force microscopy (AFM).

3. Results and Discussion

Over the first ~9 unit-cell layers, the dominant growth mode changes from island to layer-by-layer for the growth rate of 0.074 unit cells/s and the growth temperature of 700°C. The adoption of a lower growth rate of 0.0185unit-cell/s or a higher substrate temperature of 800°C was discovered and allowed the layer-by-layer growth mode to dominate over the former island growth regime, but ended up replacing the former layer-by-layer by a new growth mode regime. This growth mode was characterized by damped oscillations of RHEED intensity and decrease in RHEED intensity after deposition stop. The observation by RHEED could not be explained by conventional multi-level two-dimensional growth due to limited-mobility-induced kinetic roughening or step-flow growth. AFM observations revealed that the growth mode is related to a surface feature characteristic of the (111)-oriented STO

surface.

4. Conclusions

In the course of growing SrRuO₃ films, the governing growth mode of interest can be manipulated by changing the growth temperature and the growth rate and this change allows the selection of the desired layer-by-layer mode. The present study thus paves the way for integrations of SrRuO₃ thin layers into (111)-orientated oxide heterostructures, and hence multi-functional devices, requiring control of the sharp atomic-level interfaces and the layer-by-layer growth mode.

5. References

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