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Layer-by-layer Growth and Growth-mode Transition of $SrRuO_3$ Thin Films on Atomically Flat Single-terminated $SrTiO_3$ (111) Surfaces

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1. Introduction

Not only as a ferromagnetic functional component in (111)-oriented functional heteroepitaxial devices, but an electrode in (111)-oriented ferroelectric capacitors, free from fatigue and imprinting behaviors, $SrRuO_3$ (SRO) thin films would be one of promising candidates to be integrated into such (111)-oriented heterostructures, as have been favorably integrated into (001)-oriented heterostructures[1,2]. The key to realizing (111)-oriented oxide heterosturctures is the control of sharp heterointerfaces on the atomic level, and thus the control of a favorable growth mode is prerequisite. Despite such a strong need for (111)-oriented oxide heterostructures, realizing the layer-by-layer growth of SRO films on $SrTiO_3$ (STO) (111) has yet to be reported because an atomically well-defined, single-terminated STO (111) surface has proved difficult to achieve. A recent accomplishment of successful fabrications of such a STO (111) surface by Chang et al. [3], however, has opened the door to the layer-by-layer growth of SRO thin films, enabling SRO thin films to be integrated into (111)-oriented oxide heterostructures. In this presentation, we report on anomalous growth-mode transitions at the initial growth of SRO thin films on single-terminated STO (111) substrates.

2. Experimental Method

SRO thin films were grown on Ti^{4+} single-terminated STO (111) substrates by pulsed layer deposition (PLD). The details of the preparation of single-terminated STO (111) substrates were described in our previous report[3]. For stoichometric ablation, a KrF excimer laser (wavelength λ=248 nm) was focused on a sintered SRO target with a fluenceof 2.5 J/cm^2. 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). The RHEED diffraction patterns and intensity profiles in specular reflection geometrywere 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.

3. Results and Discussion

Over the first ～9 unit cells, the dominant growth mode changed from island to layer-by-layer for the growth rate of 0.074 unit cells/s and the growth temperature of 700ºC [4]. The observed island growth mode between the third and the ninth unit-cell layers may not be understood in terms of kinetics alone. The abrupt growth-mode transitions and the reentrance of the layer-by-layer mode above the ninth unit-cell layers could be explained by the electrostatic polarity effect[5] and its compensation, associated with the polar nature of the constituent planes of the films. A-few-monolayer-thick SRO film can be an insulator due to its low dimensionality, and when the SRO film becomes thicker than ~1.98 nm, the film shows a metallic behavior, as found in Ref. 6. Accordingly, in the thickness regime showing insulating properties, the SRO film exhibited a rough surface, while in the thickness regime showing metallic behaviors, free charge carriers compensated the electric field inherent to the SRO films, and the relevant electrostatic polarity effect did not affect the roughening of the SRO film morphology any more. The adoption of a lower growth rate of 0.0185 unit cells/s or a higher growth temperature of 800ºC was found to allow the layer-by-layer growth mode to dominate over the former island growth regime, but ended up replacing the former layer-by-layer by an unidentified growth mode regime [4]. Moreover, in the course of growing SRO films, the governing growth mode of interest could be manipulated by changing the growth temperature and the growth rate, which change allowed for the selection of the desired layer-by-layer mode.

4. Conclusion

We grew SRO thin films on atomically flat single-terminated STO (111) substrates. We found 3D island mode, layer-by-layer mode, and a certain unidentified mode in different thickness ranges, as well as growth mode transitions between them, which can be controlled by varying the growth rate and temperature parameters. These growth parameters can affect kinetic parameters such as adatom mobility and flux, which can, in turn, determine growth modes. Controllable growth parameters allow for the layer-by-layer growth of SRO films over a wide thickness range as well as for control of atomically sharp heterointerfaces.

This work was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Education, Science, and Technology (Grant No. 20100000706).

5. References

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