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RHEED intensity oscillations in homoepitaxial growth of $SrTiO_3$ films

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Abstract

The amplitude and periodicity of the reflection high-energy electron diffraction (RHEED) oscillations displayed strong temperature dependence in homoepitaxy of $SrTiO_3$(STO) films. Combining with the AFM observations, the results suggest that the oscillations are not directly related to the layer-by-layer growth.

Keywords: Homoepitaxial $SrTiO_3$ thin film; RHEED; epitaxial growth; atomic force microscopy

Owing to the fundamental and technological importance, there has been extensive revived interest in the epitaxial growth of superconducting and ferroelectric perovskite oxides [1–3]. Among others, reflection high-energy electron diffraction (RHEED) has been used ubiquitously to in-situ monitor the film growth. However, the interpretation of the RHEED intensity oscillations commonly observed has remained as an outstanding debate [1–7]. In this report we present the temperature dependence of RHEED oscillations prevailing on both the as-polished and stepped substrates in STO homoepitaxy.

The details of the pulsed laser deposition system and RHEED setup were described previously [8]. Briefly, a KrF excimer laser with pulse duration of 30 ns was operated at a repetition rate of 1 Hz and an energy density of about 2 J/cm^2. All the films were deposited at an oxygen pressure of 5×10^{-4} Torr. The substrate temperature was controlled to vary from room temperature to 810°C by a resistive heater block. The as-polished STO substrates have atomically flat surface with corrugations of less than 0.5 nm in height. While the buffered NH_4F-HF solution treated stepped substrates are having steps of mostly one unit-cell high and terraces of about 100 nm wide. Both surface structures were confirmed by atomic force microscopy (AFM). The electron beam energy of RHEED was 20 keV with a grazing angle of 0.7° and was directed along [1 0 0] of the STO substrate. With a de Broglie wavelength of about 0.086 nm, the grazing electron beam is slightly off-Bragg conditions, and hence is most sensitive to the edges of surface steps [6].

Figs. 1 and 2 show the oscillations of the RHEED specular reflections as a function of temperature for the homoepitaxial STO films grown on as-polished (Fig. 1) and stepped (Fig. 2) substrates, respectively. In both cases, there are marked changes in the RHEED intensity oscillation with increasing temperature. Previously, the periodic RHEED oscillations have been interpreted as a direct indication of layer-by-layer growth [1–4]. It is noted, however, that both the amplitude and periodicity of the respective oscillations are changing with the deposition temperature. Since, in all cases the repetition rate (1 Hz) and energy density (2 J/cm^2) of the laser were kept the same, the amount of material contained in the plume of every laser pulse should presumably be about the same. As a result, a fixed oscillation periodicity is expected for a strict layer-by-layer growth. Alternatively, the undamped RHEED oscillations can occur as a result of changing step density distribution, while damping results from increasing contribution of third level [6,7]. In this scenario, the lack of oscillation at low temperatures can be interpreted as a consequence of limited diffusivity. It not only hinders the coalescence between

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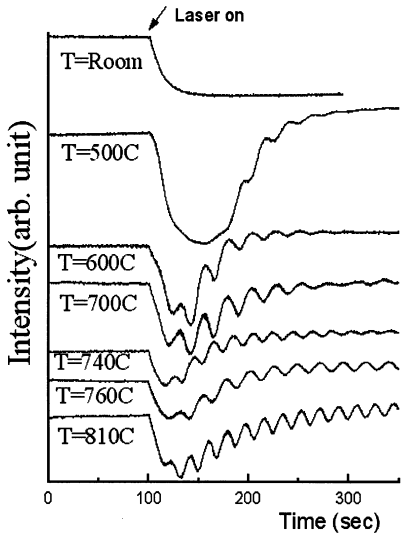

Fig. 1. RHEED oscillation for STO on as-polished substrates.

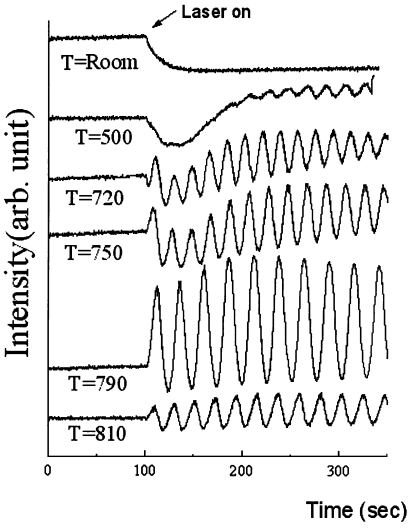

Fig. 2. RHEED oscillation for STO on stepped substrates.

individual nuclei but also distributes steps over multiple layers. At higher temperatures, the enhanced diffusivity helps the redistribution of the surface steps, leading to the oscillatory behavior in RHEED intensity. Indeed, AFM investigations have revealed intimate relations between the film surface structures and the corresponding RHEED intensity variation [8]. This explains, in a consistent way, the strong temperature dependence of the emergence as well as the amplitude and periodicity of the RHEED intensity oscillations observed.

In summary, the in situ RHEED intensity oscillations observed in the homoepitaxial growth of STO (100) films show marked temperature dependence. The results suggest that the origin of the RHEED oscillations may intimately relate to the variations of the surface step distribution rather than serving as a direct indication of layer-by-layer growth.

Acknowledgements

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