In situ time-resolved x-ray scattering study of pulsed-laser deposition thin-film growth

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Introduction

The high brilliance of third-generation synchrotron light sources provides sufficient scattering intensities that millisecond-range time-resolved measurements can be performed using the weak surface scattering crystal truncation rods (CTRs). As a result, it is now possible to study the time dependence associated with surface crystallization and the dynamics of surface evolution using x-ray diffraction. Pulsed-laser deposition (PLD), utilizing short, ~25 ns excimer laser pulses, ablates material nearly stoichiometrically from a target to a heated substrate [1]. Typically, the ablated plume of target material arrives at the substrate in times substantially less than a ms, and often as short as a few tens of μs. Thus, PLD provides an opportunity to investigate the kinetics of surface aggregation different from that of molecular beam epitaxy (MBE) by making possible a separation of the deposition and aggregation phases. In this report, we describe essential features of a newly developed time-resolved PLD surface diffraction system and we present initial time-resolved measurements on homoepitaxial growth of $SrTiO_3$.

Methods and Materials

The diffraction system, shown schematically in Figure 1, uses the 2 + 2 geometry [2], where there are four axes (two on the sample and two on the detector). This geometry keeps the ablation target fixed relative to the sample. Also shown is an independently rotating mirror on the χ axis to maintain the excimer laser ablation pulses at a fixed position on the target for all values of χ. This arrangement makes it possible to do PLD at all orientations. The chamber can be operated at temperatures up to 800^{O} C with a background gas of up to 1 torr O_2 and/or other buffer gases.

Time-slice measurements of CTRs are performed using three channels of multiscaler data; two detectors and an incident beam monitor. Measurements typically cover a range of 1 second before and 10 to 20 seconds following individual laser pulses using time slices of 5 ms. By firing the laser one second after initiating time-slice measurements, an accurate measure of diffraction intensity before as well as immediately after the deposition of the ablated growth pulse is obtained. A fast NaI detector (Oxford, Cyberstar X1000) counts reliably at our maximum rate of ~10^5 Hz.

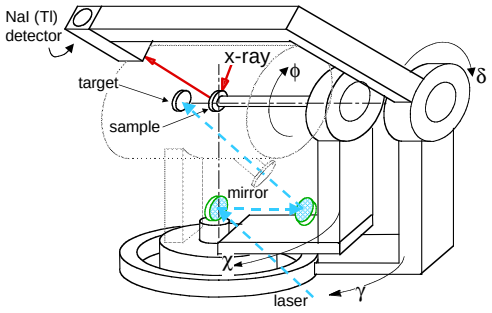

Figure 1: Diffractometer geometry. The sample rotates on two axes, χ and ϕ, and the detector rotates on γ and δ. Specular scans are taken by scattering horizontally and in-plane by scattering vertically.

Results

Initial results using this system to study homoepitaxial growth of $SrTiO_3$ are shown in Figure 2. Scattered intensity at the (001/2) specular anti-Bragg position is plotted as a function of time during the growth of five $SrTiO_3$ layers; similar oscillations were measured simultaneously at the off-specular (0 1 1/2) anti-Bragg position. The damping of the contrast of the oscillations about the midpoint intensity indicates departure from layer-by-layer growth and the beating in the maximum intensity of the oscillations is an indicator of the density/stoichiometry of the deposited layer. Figure 2b shows intensity transients associated with increases in coverage and surface evolution associated with individual laser pulses.

Discussion

We are presently modeling the observed time dependence for single laser shots in terms of the abrupt arrival of Sr, Ti, and O and subsequent crystallization/aggregation. The prompt arrival and crystallization of these constituents into bilayer $SrTiO_3$ produces the abrupt jump at each laser pulse, while the evolution of the surface step geometry of the $SrTiO_3$ bilayers leads to the upward recovery, which continues until the following laser shot. RHEED

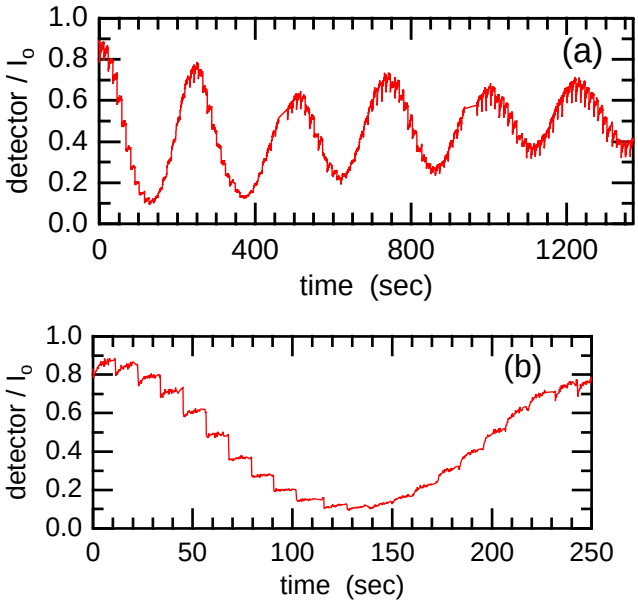

Figure 2: Time-resolved diffraction for $SrTiO_3$ homoepitaxy at (0 0 1/2) anti-Bragg position. (a) Oscillations from layer-by-layer growth, and (b) expanded view of individual pulse reflectivity transients for the first oscillation period.

measurements [3] on such oxide systems have revealed similar effects; however, detailed quantitative analyses of the full oscillation time structure have not been performed. The addition of x-ray measurements to nonequilibrium thin-film and multilayer growth investigations will provide complementary information on evolution at buried interfaces in heterostructure growth as well as provide for quantitative modeling of surface structure and evolution.

Acknowledgments

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