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In-Situ Surface Diffraction: Perovskite Thin Film Growth

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Introduction

The bulk perovskite structures are influenced by ionic radii and by oxygen vacancies. At the surface, the ionic coordination and the oxygen stoichiometry differ from the bulk, causing relaxation and reconstruction. For photoemission spectroscopy and ultra-thin-film applications, the surface region is of great importance. Scanning probes give poor lateral resolution and no depth information, and electron diffraction suffers from strong multiple scattering and charging. Glancing-incidence surface X-ray diffraction overcomes these problems, but requires a large, flat sample and an intense, low-divergence synchrotron radiation beam.

Methods and Materials

At the Materials Science Beamline of the Swiss Light Source [1], a surface diffraction facility has been established with a) an intense wiggler source, b) an in-situ pulsed-laser deposition epitaxial growth chamber [2], and c) a zero-noise, 2D pixel-detector [3]. This facility allows the characterization of the entire near-surface region (2 nm into a perovskite [4]), or alternatively, in-situ dynamical and structural studies during film growth. The present study concerns in-situ growth of $La_{0.66}Sr_{0.34}MnO_3$ (LSMO) on (001) $SrTiO_3$ (STO).

Results

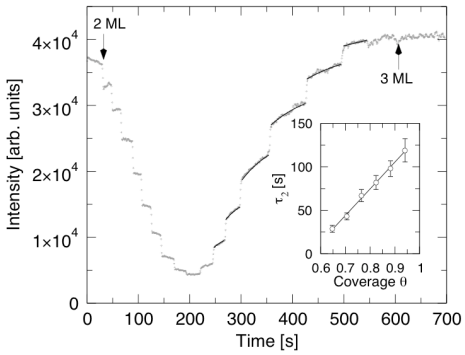

Fig. 1. Reflected X-ray intensity during "burst" growth of the third LSMO monolayer, with inset showing the dependence of the smoothing relaxation time τ_2 on the layer completion θ.

Fig. 1 shows results of a dynamical study of LSMO film growth [5]. A 12.4 keV photon beam was incident onto the 10x10 mm^2 sample, and the reflected intensity was measured at the (0 0 1/2) position along the specular crystal truncation rod (CTR). This signal oscillates during island coalescence growth due to repeated roughening and smoothing and to interference between reflections from the film surface and the film-substrate interface. The Figure follows the reflectivity during deposition of the third monolayer. In order to observe the evolution of the surface roughness, the laser power was delivered in a series of separated bursts. Smoothing of the surface due to diffusion can be followed above 60% layer completion, and a relaxation time constant τ_2 can be extracted as a function of coverage θ (see inset). The linear dependence $\tau_2(\theta)$ with an intercept at θ=0.55 has been interpreted with a model involving the breakup of existing islands by the energetic impinging species [5].

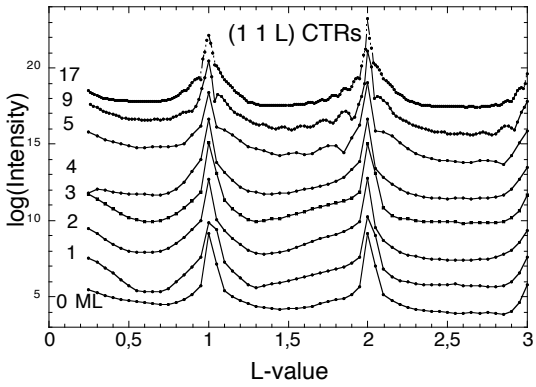

Fig. 2. Monolayer-resolved CTRs of LSMO growth on STO (001).

With the in-situ growth facility, a complete surface diffraction pattern can be taken at any point of the growth process. Fig. 2 shows the (1 1 L) CTRs measured after various numbers of deposited LSMO monolayers. With our pixel detector, such a CTR can be measured in approximately 15 minutes. Note the appearance in the thicker layers of Kiessig interference fringes, demonstrating a very smooth film. We are in the process of analyzing the monolayer-resolved surface structures.

Discussion

The large amount of high-quality data available from the pixel detector allows us to analyze multiply-reconstructed surface structures to large depths [4]. The ability to heavily oversample CTRs should make phase-retrieval analysis methods more feasible for complex oxides.

Acknowledgments

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References

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