Transient Interlayer Transport in SrTiO₃ Pulsed Laser Deposition Studied by Time-Resolved Surface X-Ray Diffraction

Gyula Eres¹, J.Z. Tischler¹, B.C. Larson¹, C.M. Rouleau¹, and P. Zschack²

¹Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA ²Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, USA

Abstract. Pulsed laser deposition (PLD) is a widely used technique for growing a variety of oxide thin films. Film growth in PLD occurs from an energetic plume of material ejected from a solid target by pulsed laser ablation. The plume consists of a complex mixture of neutral and ionized atoms, molecular fragments, and even small clusters with kinetic energies up to a few hundred eV. The extra kinetic energy provides a transient enhancement of surface mobility that alters the nucleation and growth of thin films. We use time-resolved surface x-ray diffraction (SXRD) to study the formation and evolution of surface structure during PLD of SrTiO₃. The scattered intensity is measured simultaneously at the specular (0 0 ½) and the (0 1 ½) offspecular anti-Bragg positions with microsecond range resolution using avalanche photodiode detectors. The (0 1 ½) transient has an in-plane component and reveals that crystallization into SrTiO₃ unit cells occurs simultaneously with the arrival of the laser plume. The use of x-ray diffraction greatly simplifies growth kinetics studies because in the kinematic limit the x-ray intensity changes correspond directly to coverage changes. Rather than using surface transport models to fit the data, we instead analyze the intensity transients using a model independent approach that allows direct determination of the time-dependent surface coverages from the transient intensities¹. The abrupt change in the intensity corresponds to the fraction of the pulse instantaneously crystallizing in each layer, and the time evolution of the intensity describes interlayer transport from the top of the islands into the growing layer. This analysis reveals that energy-enhanced interlayer transport occurs on a time scale of microseconds or less and it dominates layer filling in PLD growth. A much smaller fraction of material, which is governed by the dwell time between successive laser shots, is transferred by slow, thermally driven interlayer transport processes. In a different set of experiments the time dependence of the entire diffraction profile including the diffuse scattering around the crystal truncation rods is measured using a CCD camera. I will present preliminary results on diffuse scattering to discuss the timedependent spatial distribution of islands during layer filling and their evolution during the dwell time between the laser shots.

REFERENCES

1. J.Z. Tischler, Gyula Eres, B.C. Larson, C.M. Rouleau, P. Zschack, and D.H. Lowndes, *Phys. Rev. Lett.* **96**, 226104 (2006).