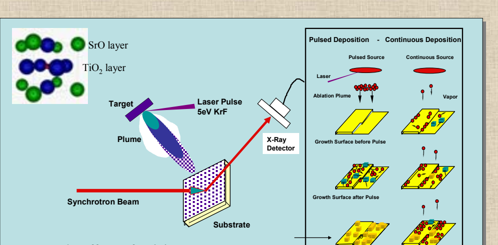
Time-Resolved X-ray Surface Diffraction Study of Surface Transport During Pulsed Laser Deposition of $SrTiO_3$

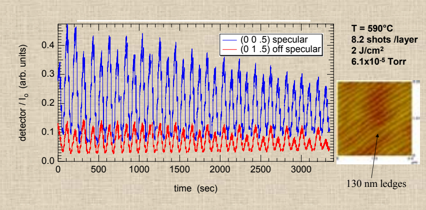
J.Z. Tischler, B.C. Larson, Gyula Eres, C.M. Rouleau, and D.H. Lowndes
(Condensed Matter Sciences Division ORNL)
P. Zschack *(UNICAT, UIUC)*

We describe the use of surface x-ray diffraction (SXRD) for real-time monitoring of oxide thin-film growth during pulsed laser deposition and testing of models that rely on different time scales. The use of SXRD ensures that simple kinematic single scattering analysis can be used to provide direct physical insight into the details of aggregation, crystallization, and surface kinetics involved in the epitaxial growth process.

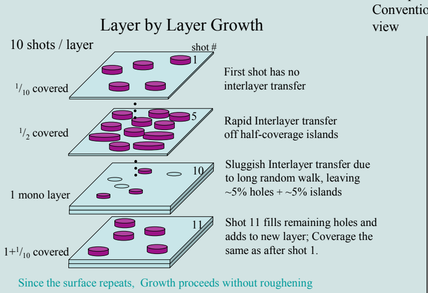
The experiments were performed on the UNICAT undulator beamline using a monochromatic 10 keV x-ray beam. Discrete measurements of the diffracted intensity at the ($00^1/_2$) anti-Bragg position on the specular crystal truncation rod were made at 5ms intervals during the pulsed laser deposition of homoepitaxial $SrTiO_3$. The data suggest that two time scales are present. A simple rate model is unable to explain the abrupt and consistent increase in scattered intensity after half coverage. The early time scale is characterized by a very fast motion which appears prompt in our measurements. We propose the concept of "prompt" motion where some the hot material that lands immediately following the deposition pulse moves down to a lower level, followed by slower more conventional motion thereafter.



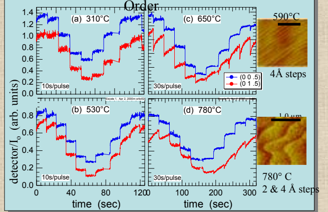
In PLD, the affects of arriving material can be separated in time from evolving material

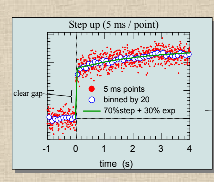


A Simple Conventional view

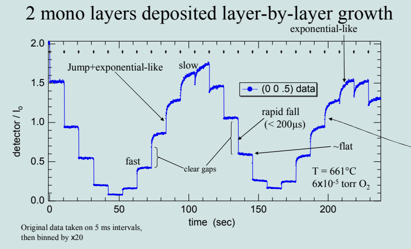


Effect of Temperature on In-Plane Order

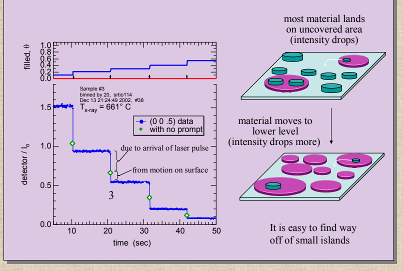




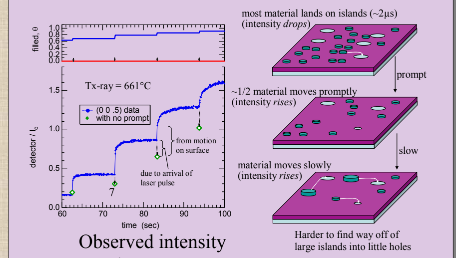
General Features Found in PLD



Intensity *vs.* coverage
Low Coverage



Approaching Full Coverage

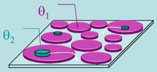


Observed intensity increases

Simple Model Used for Slow Decay

Rate ~ (material on level 2) x (area of holes in bottom)

$$\theta_2' = -\frac{\theta_2(t)\theta_0(t)}{\tau}$$



Where: $b + \theta_2(t) = \theta_0(t)$ is the area of holes

$$b = 1 - [\theta_2(t) + \theta_1(t)] = 1 - \theta_{total}$$

Solution to diff. eq.

$$\theta_2(t) = \frac{be^{bC}}{e^{bt/\tau} - e^{bC}}$$

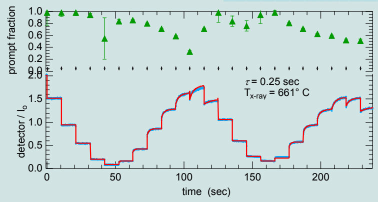
Boundary condition

$$C = \frac{\ln\left(\frac{\theta_2(t=0)}{b + \theta_2(t=0)}\right)}{b}$$

Prompt Fraction and τ

$$\theta_2(t) = \frac{be^{bC}}{e^{bt/\tau} - e^{bC}} \qquad b = 1 - \theta_{total}$$

Prompt fraction ~ 1 for <0.5 Coverage
Prompt fraction <0.5 as coverage ->1
τ =0.25 sec



Summary

- Prompt and slow intensity transients modeled
- Prompt interlayer transfer fraction, f_p
  - ~ 1 for low θ,
  - ~ 1/2 for θ approaching 1
- τ ~0.2-.4 s for 10 sec laser dwell-time
- Slowing of intensity transients results from decreasing hole density
- Random walk precludes complete layer filling

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