Dynamics of Thin-Film Deposition

Joel D. Brock
School of Applied & Engineering Physics
Cornell High Energy Synchrotron Source (CHESS)
Cornell Center for Materials Research (CCMR)
Energy Materials Center at Cornell (EMC^2)
Cornell University

What are the fundamental physical processes involved in thin film deposition?

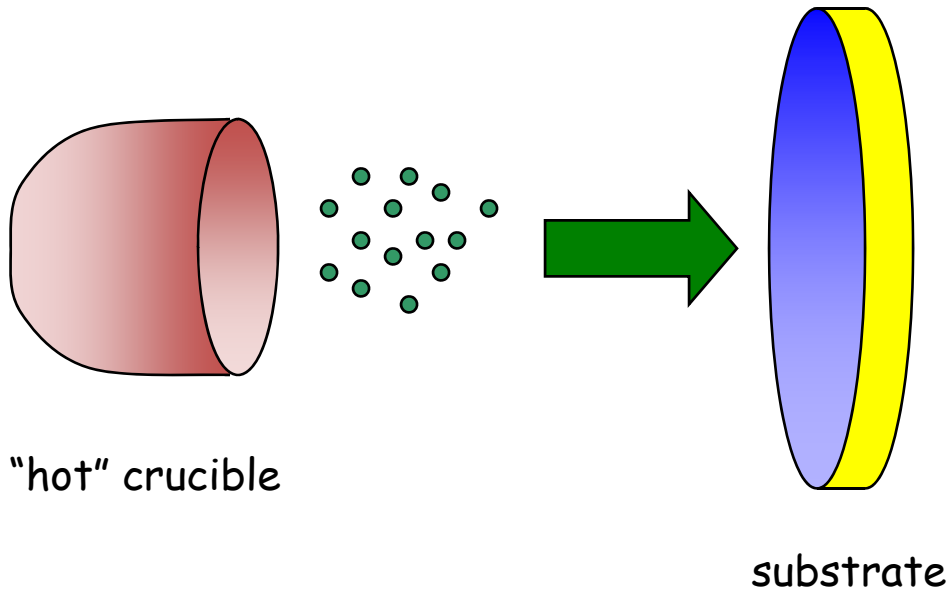
It depends…

Two Limits: Thermal and Energetic

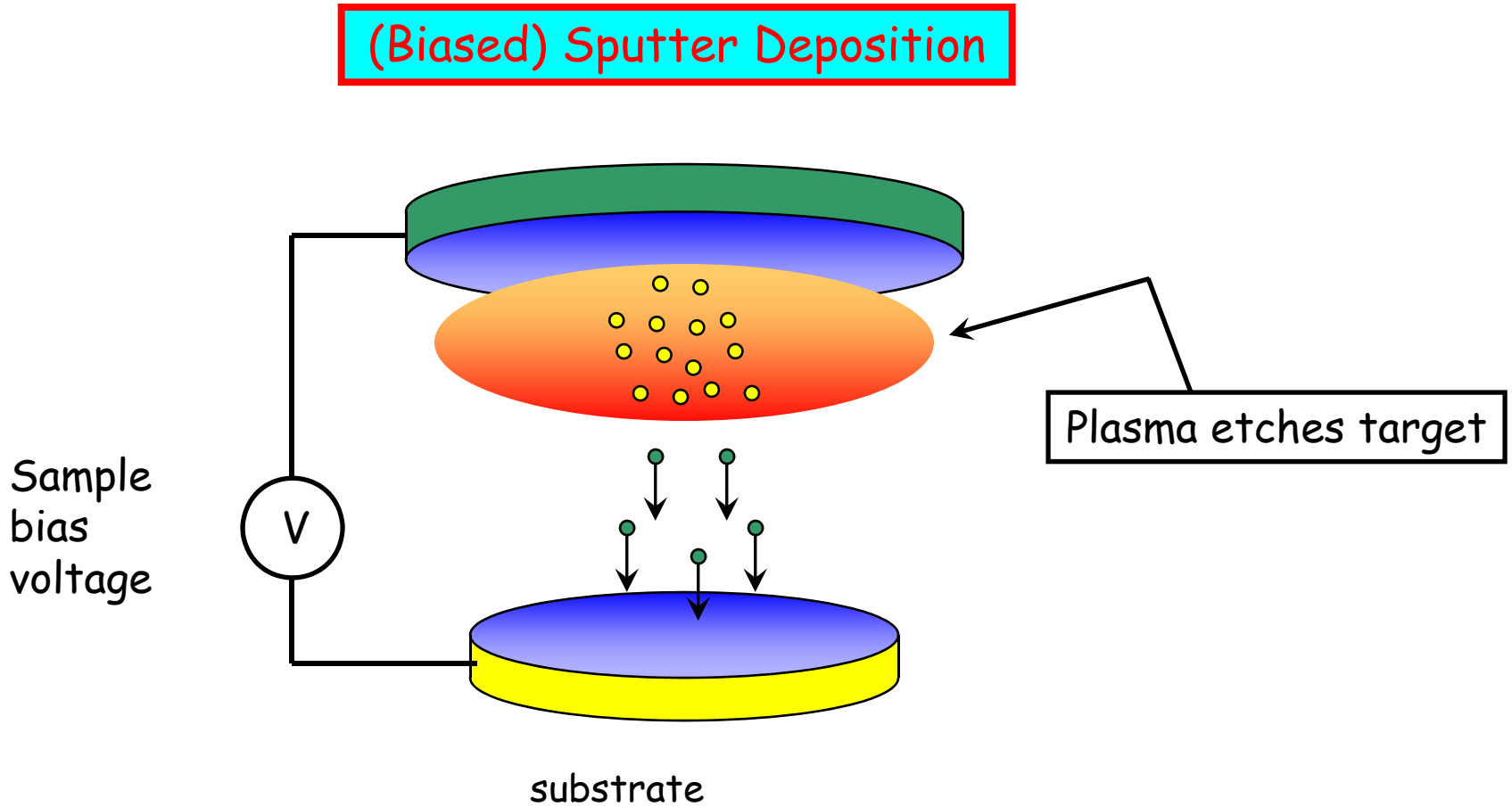
Example: Thermal Deposition

Molecular Beam Epitaxy

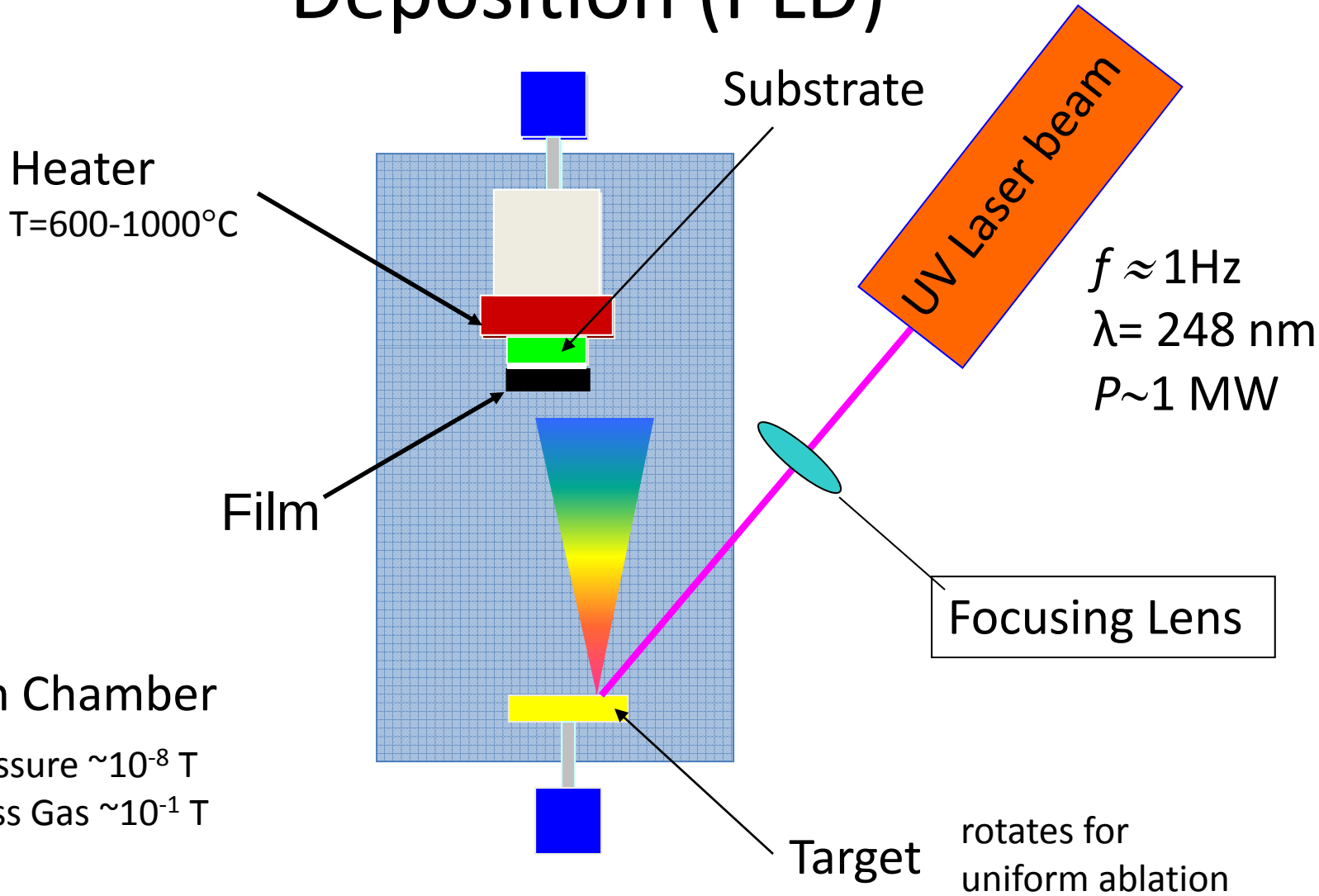
(MBE)

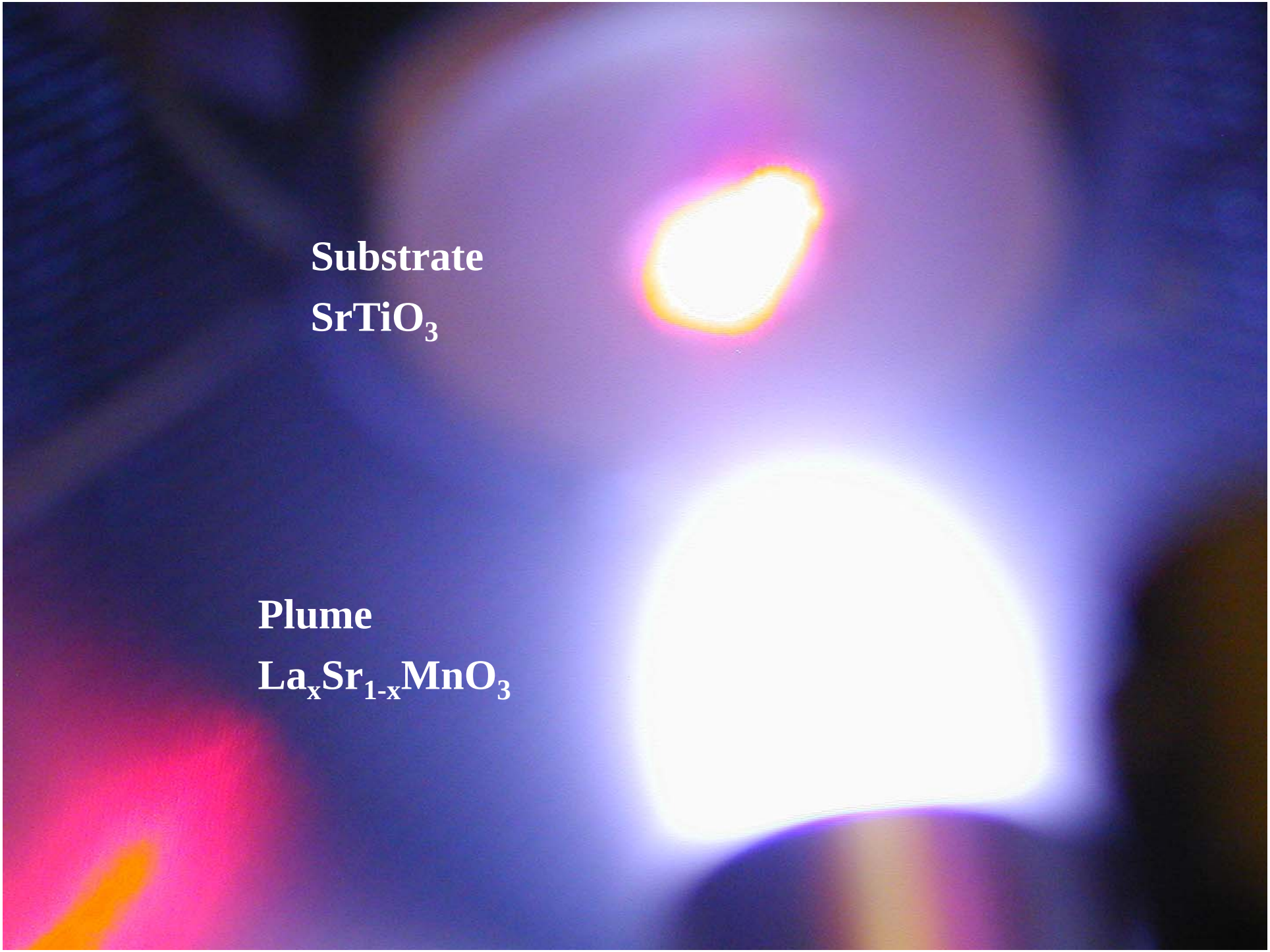


Example: Energetic Deposition



Study both with Pulsed Laser Deposition (PLD)




Substrate
$SrTiO_3$
Plume
$La_xSr_{1-x}MnO_3$

CONDENSED-MATTER PHYSICS

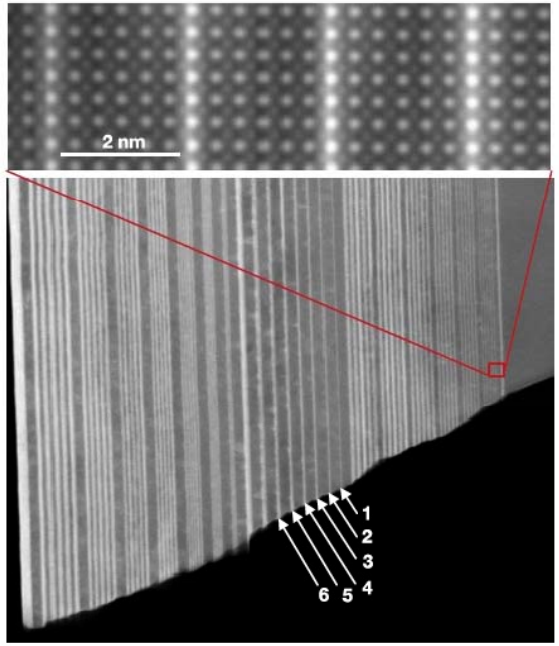
Complex oxides on fire

Oxide materials have long been known as hosts for exotic and useful physical properties. Recent advances in their design and fabrication establish the means to tailor their response for applications.

Yoshinori Tokura and Harold Y. Hwang

nature materials | VOL 7 | SEPTEMBER 2008

- Ferroelectricity
- High dielectric Permittivity
- Superconductivity
- Colossal Magnetoresistance
- Ferromagnetism
- Multi-ferroic devices
- 2D electron gas



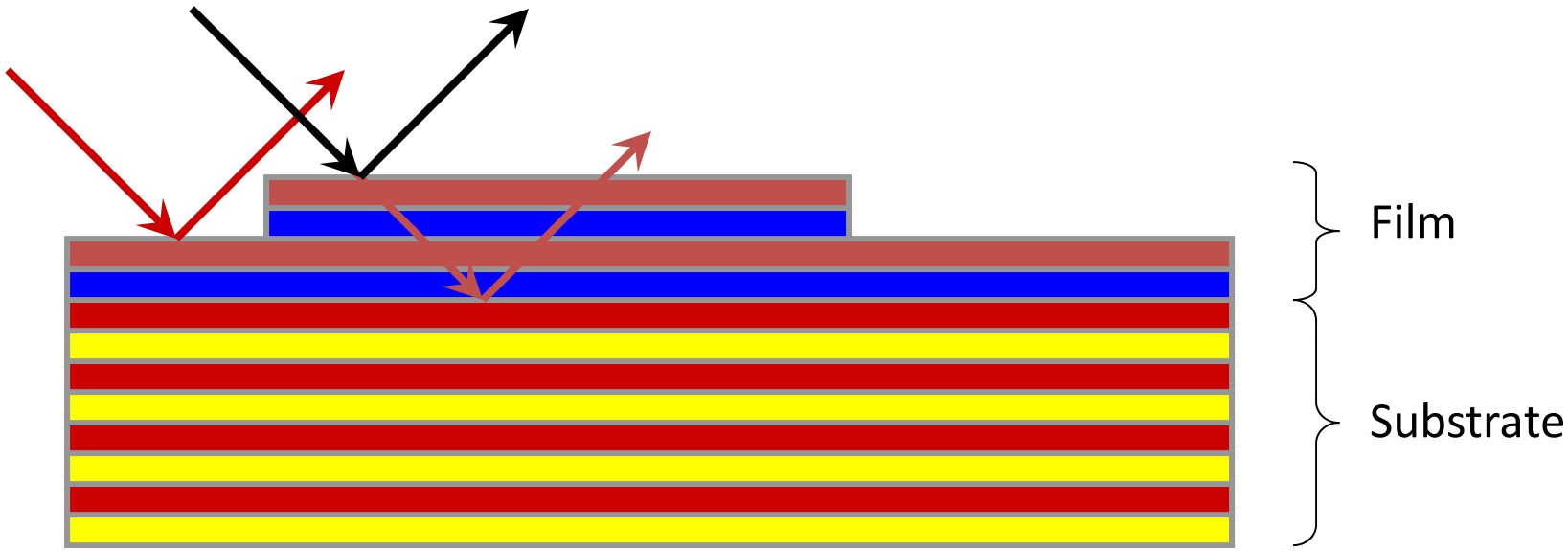
A. Ohtomo, et al, Nature **419, 378 (2002).**

Physical Problem

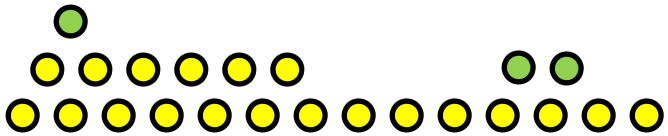
The problem of studying energetic deposition is extremely challenging due to

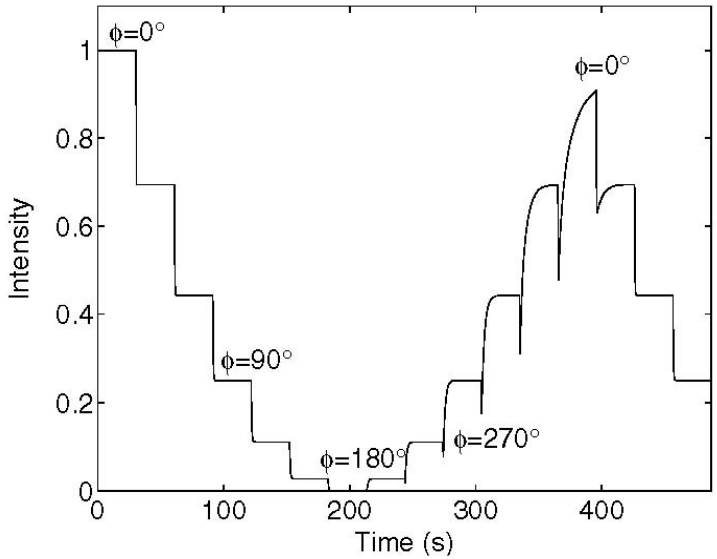
- Length scales vary from atomic to macroscopic (*e.g., from Å to 10's or 100's of μm)*
- Time scales vary from collisions which last for roughly one picosecond to layer completion times on the order of several seconds. (10^{12})

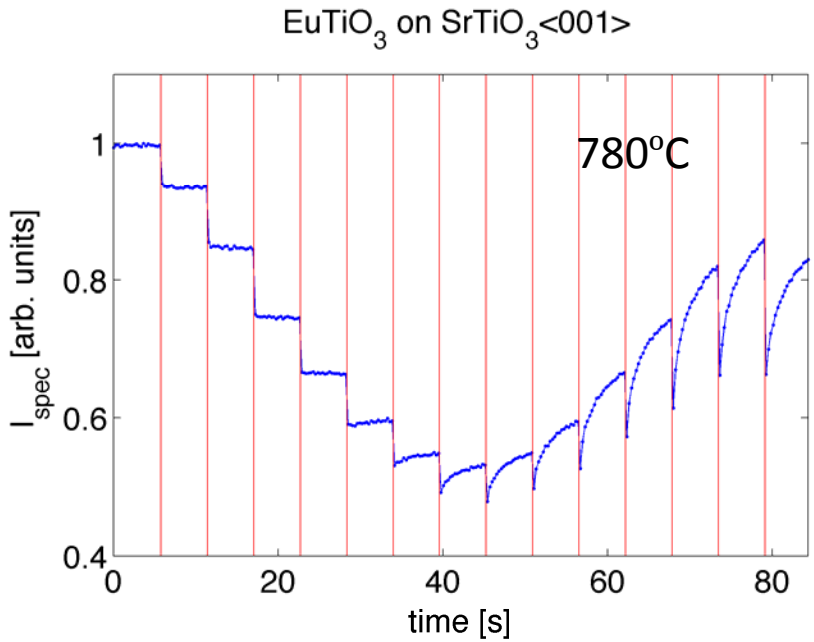
X-Ray Reflectivity (specular)



$$I_{spec} \propto \left| F_{sub}(q) \sum_{n=0}^{\infty} e^{iqna} + F_{film}(q) \sum_{n=1}^{N} \theta_n(t) e^{-iqna} \right|^2$$







Schematic Representation

Recent (October, 2009) Data

Consider Impact of Plume on Substrate

Assumptions:

1. Impulse approximation
2. Atoms land at random locations on surface

$$\frac{\delta I}{I} = -4\sigma(1-\sigma)$$

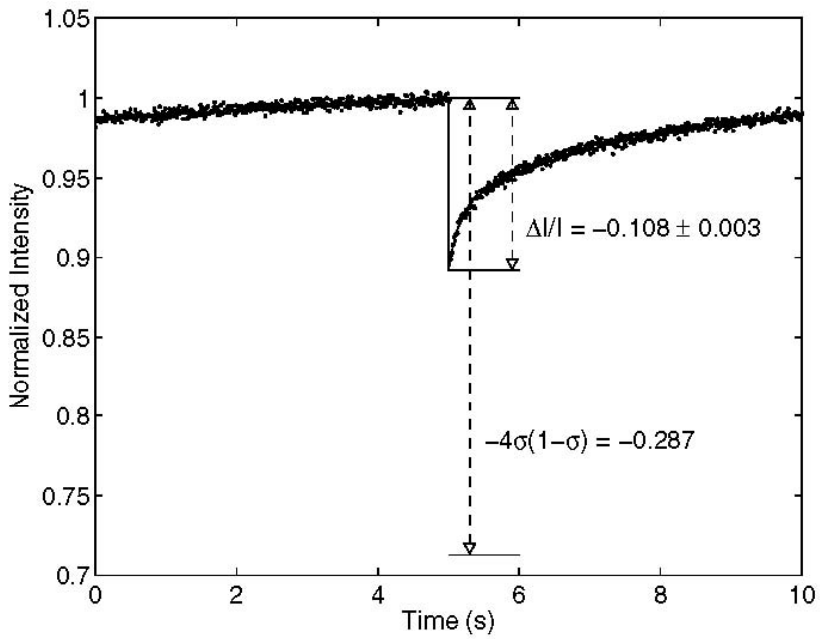
σ = (# of pulses per layer)$^{-1}$

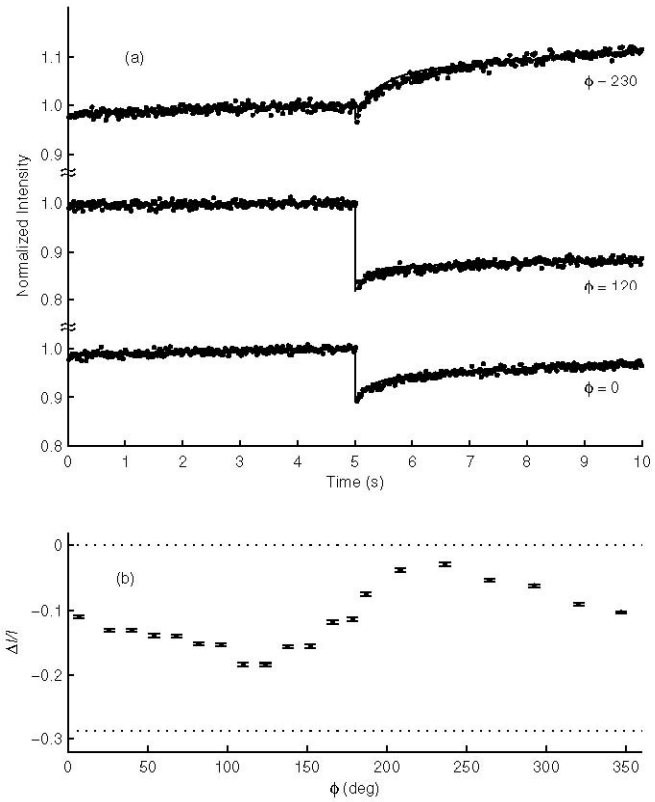
PRL **94,** 036102 (2005) PHYSICAL REVIEW LETTERS week ending 28 JANUARY 2005

Observed Effects of a Changing Step-Edge Density on Thin-Film Growth Dynamics

Aaron Fleet,[1,2] Darren Dale,[3,2] Y. Suzuki,[4] and J. D. Brock[1,2]

[1]*School of Applied and Engineering Physics, Cornell University, Ithaca, New York 14853, USA*
[2]*Cornell Center for Materials Research, Cornell University, Ithaca, New York 14853, USA*
[3]*Department of Materials Science and Engineering, Cornell University, Ithaca, New York 14853, USA*
[4]*Department of Materials Science and Engineering, UC Berkeley, Berkeley, California 94720, USA*
(Received 31 August 2004; published 25 January 2005)

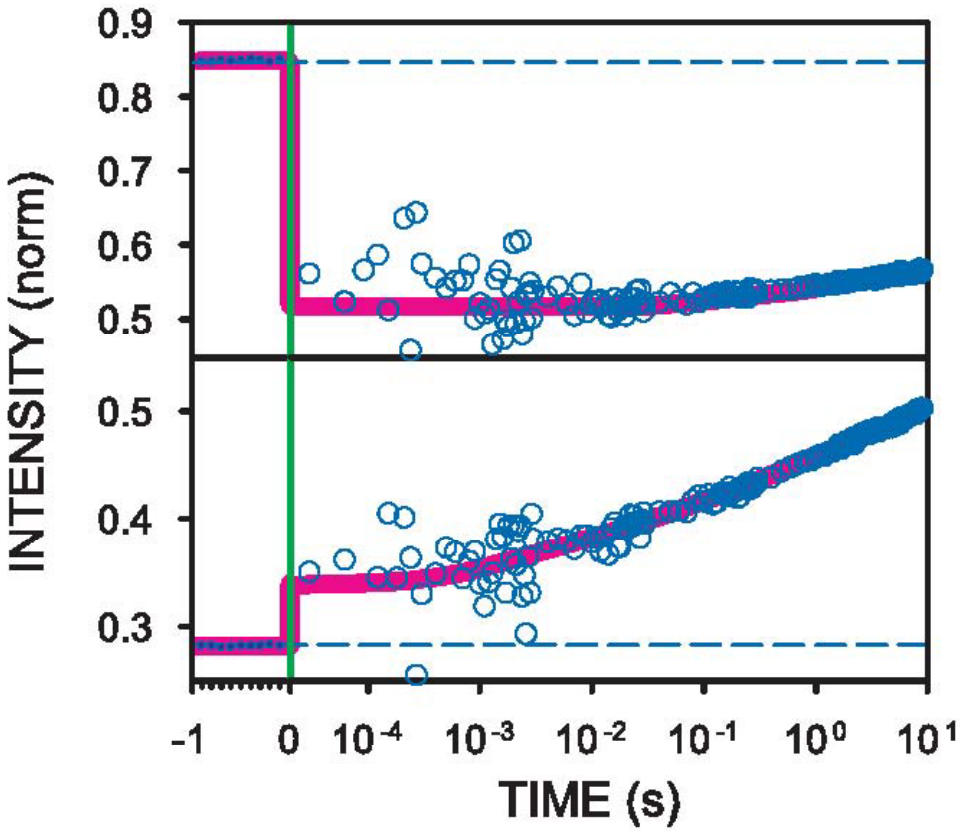




PRL **96,** 226104 (2006) PHYSICAL REVIEW LETTERS week ending 9 JUNE 2006

Nonequilibrium Interlayer Transport in Pulsed Laser Deposition

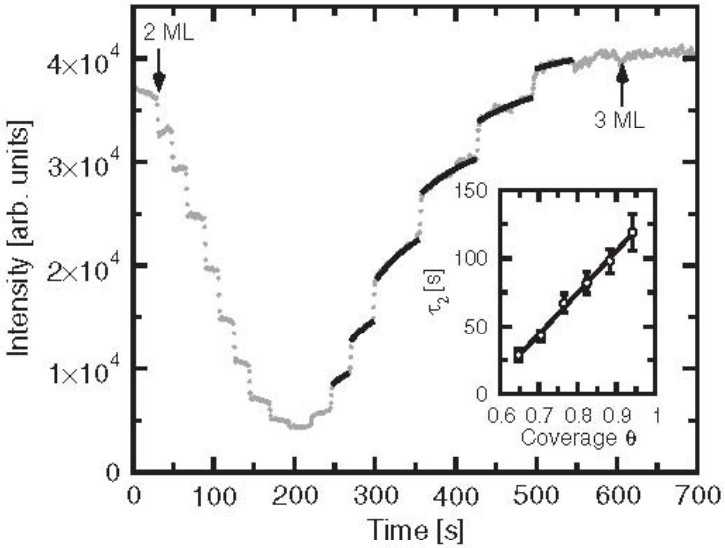
J. Z. Tischler,[1] Gyula Eres,[1,*] B. C. Larson,[1] Christopher M. Rouleau,[1] P. Zschack,[2] and Douglas H. Lowndes[1]
[1]*Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*
[2]*Frederick Seitz Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801, USA*
(Received 28 February 2006; published 6 June 2006)

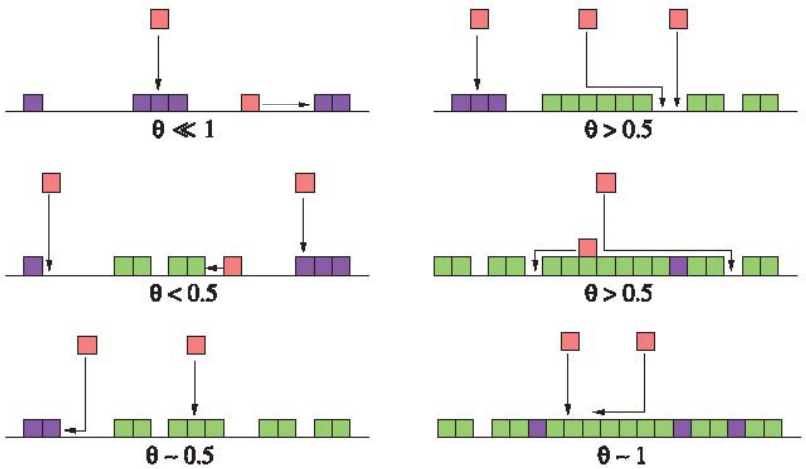


PRL **96,** 176102 (2006) PHYSICAL REVIEW LETTERS week ending 5 MAY 2006

Energetic Surface Smoothing of Complex Metal-Oxide Thin Films

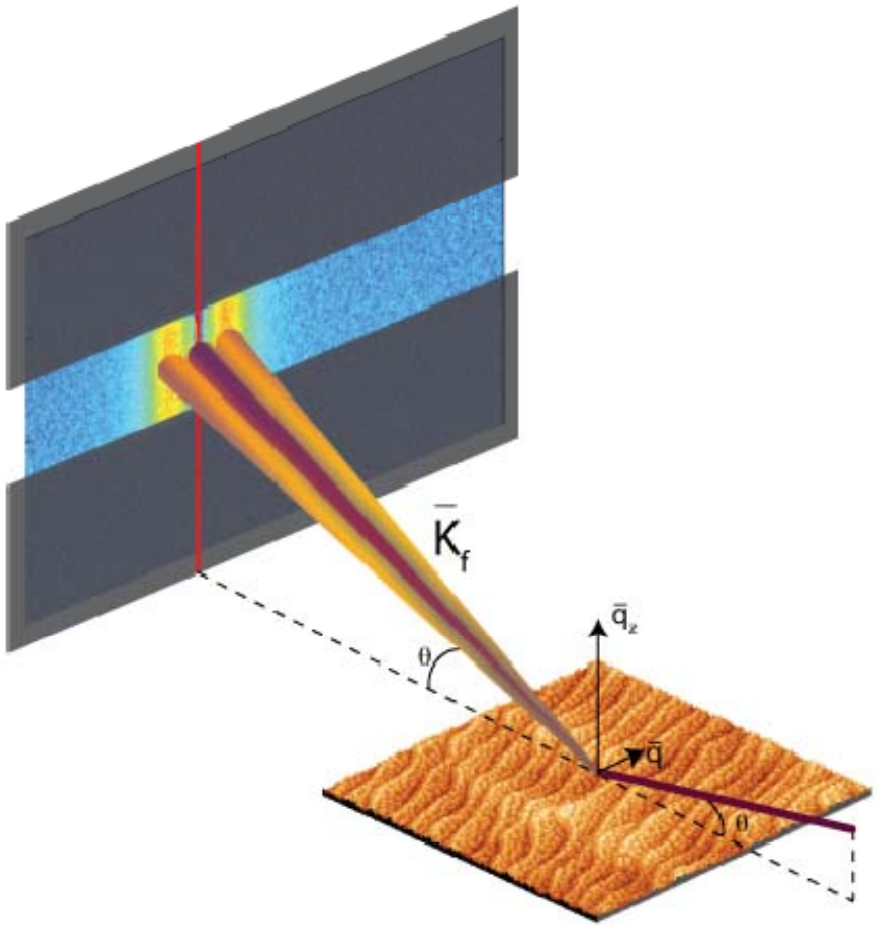
P. R. Willmott,* R. Herger, C. M. Schlepütz, D. Martoccia, and B. D. Patterson
Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen, Switzerland
(Received 19 January 2006; published 2 May 2006)

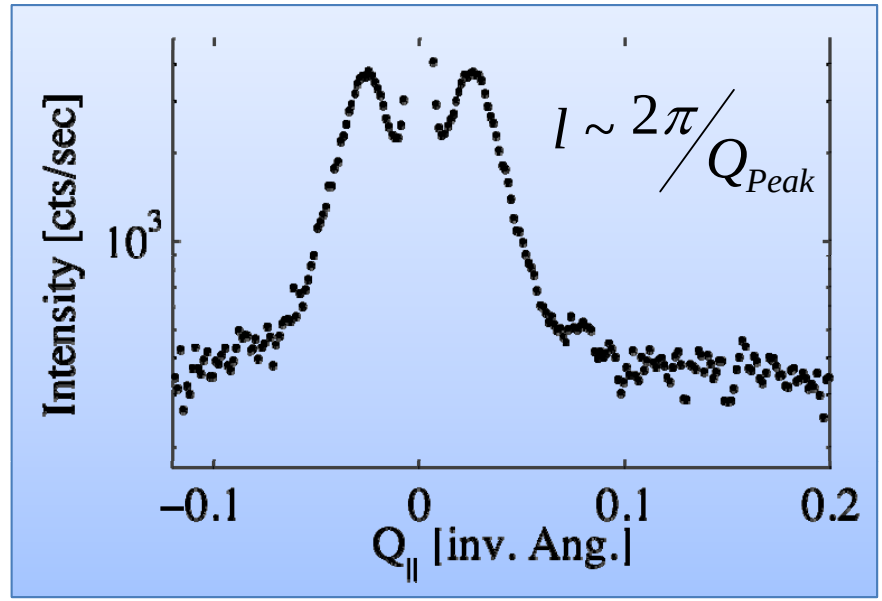




Argues that "island break up" is responsible for smooth growth during PLD.

Scattering Geometry





200 ms Time Resolution

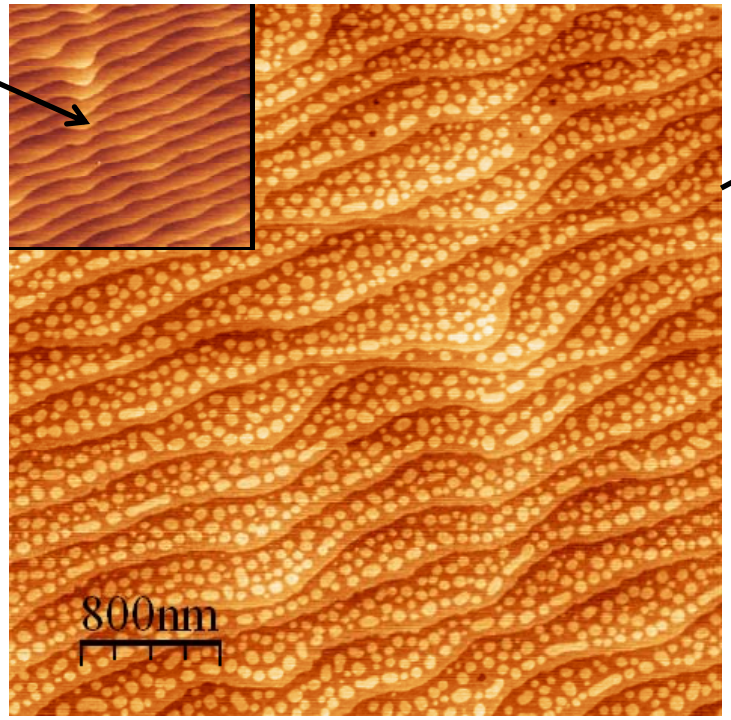
- Optimum between diffuse signal and thermal background

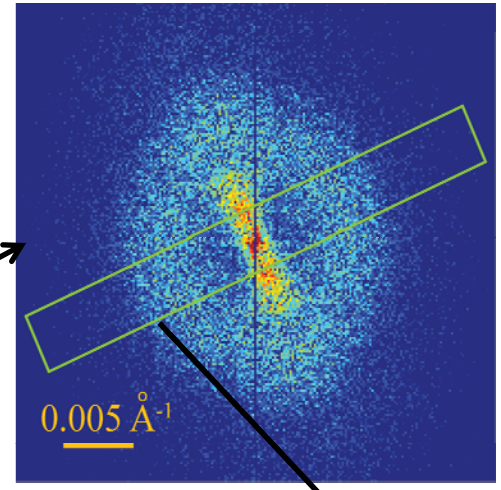
$$q_z = 0.275 \text{ r.l.u.}$$

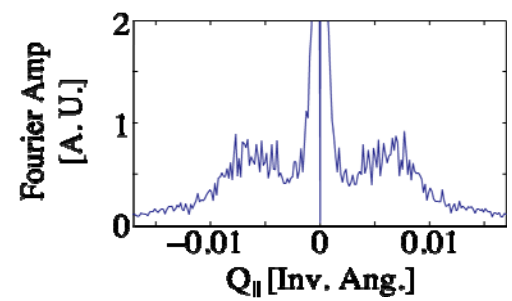
Atomic Force Microscopy

11.3 Monolayers of $SrTiO_3$

Starting Surface

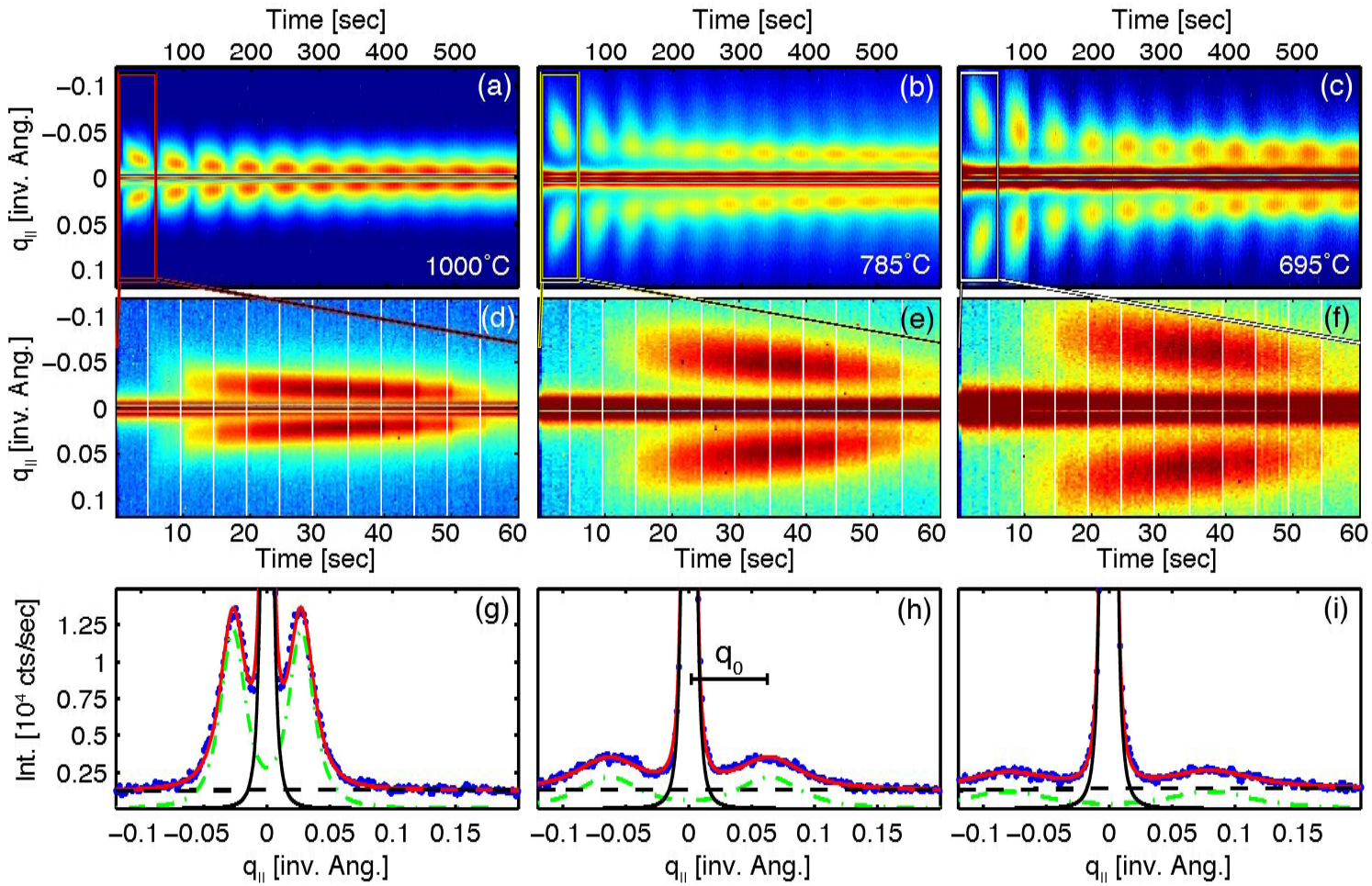






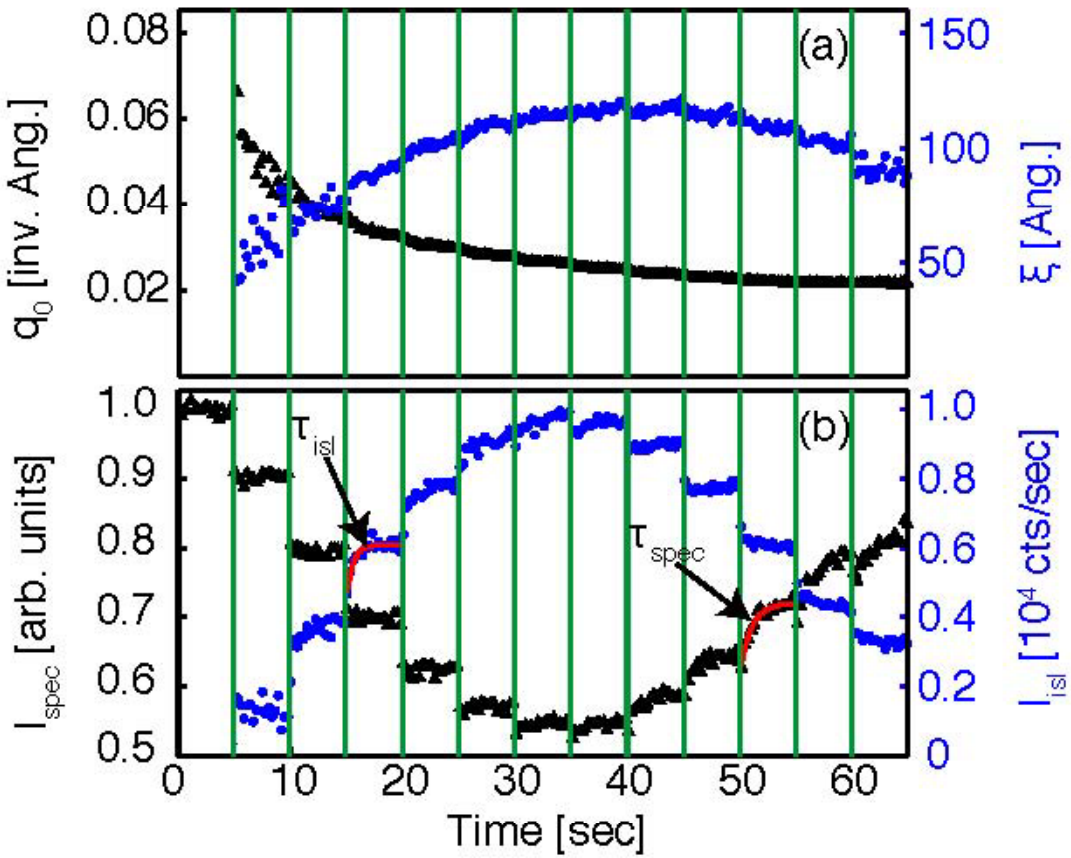
Excellent real space images of small number of stationary islands.

Time-resolved Diffuse Scattering



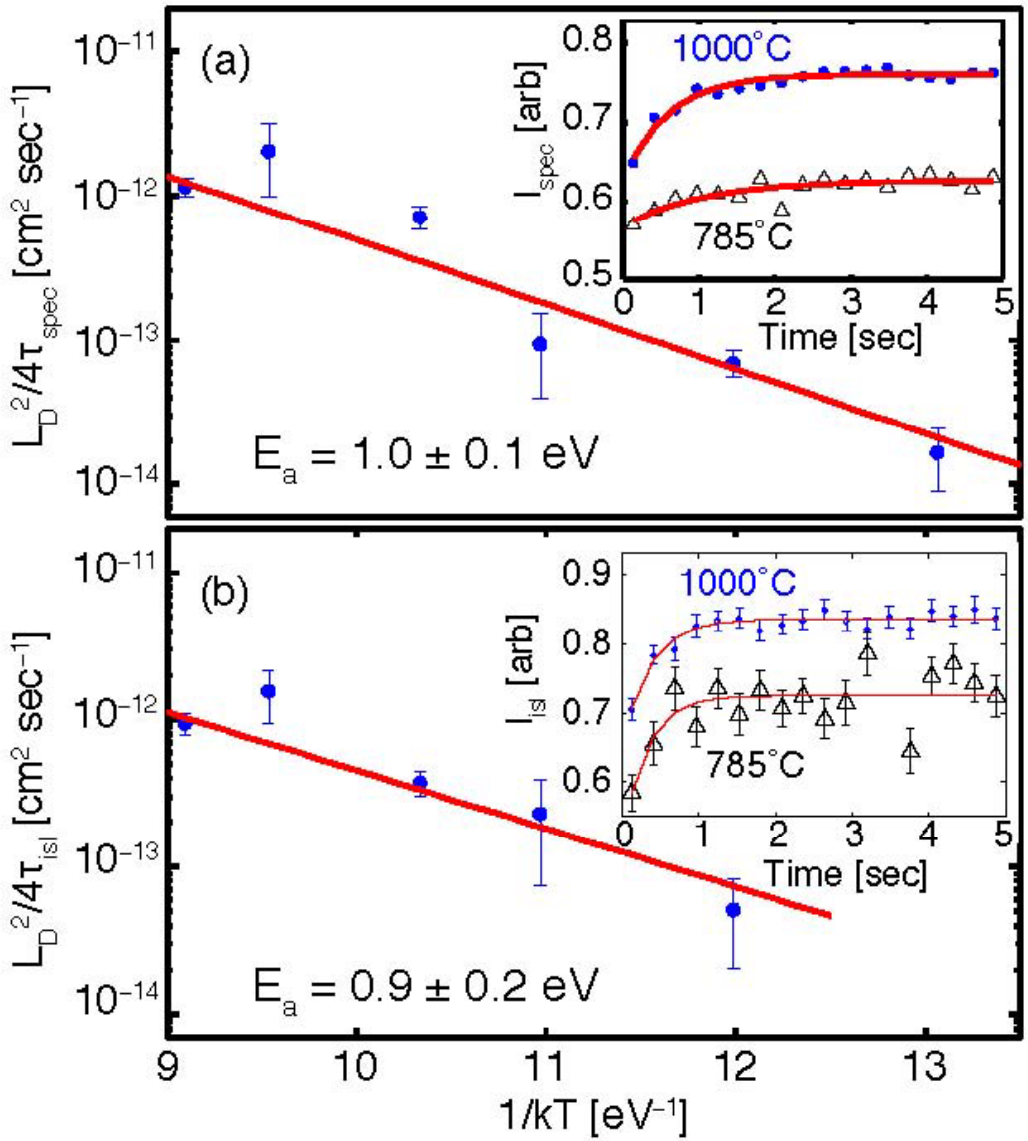
$$I_{diff}\left(\vec{Q}\right)=\frac{I_0}{\left(1+\xi^2\left(Q-Q_0\right)^2\right)^{3/2}}$$

Best Fit Parameters



No jump in spacing between islands as laser fires. Therefore, island break up isn't significant in this particular case.

Arrhenius Behavior



What don't we know?

- What happens during plume collision?
  - Preliminary tr-fluorescence data (not shown) indicates different species arrive at distinct times.
  - tr-current data (not shown) indicates shock waves in plume.
  - What are effects on nucleation, aggregation, coarsening at early times?
  - How fast is fast?
  - Would like to perform XPCS on fast time scales
- How to understand diffuse scattering data obtained at coalescence?
  - Transition from islands to circular pits or to long thin valleys?
  - Tr-Coherent diffraction?

What would we like?

- Pulse Length

  Time scale for rearrangement of atoms is 1 picosecond. ($k=1eV/a^2$, $a = 3Å$, $m=10^{-25}kg$)

$$T_{vib} = 2\pi(k/m)^{1/2} \approx 1ps$$

- Pulse Intensity

  For short pulses, energy deposition is adiabatic.

$$N_{\max} = \frac{3k_B L^2}{E\sigma_{abs}} \Delta T_{\max},$$

$L=100\mu m$, $\Delta T=1 \rightarrow N_{max} \leq 10^8$

What would we like?

- Repetition Rate

  Want to keep flux of modern 3rd generation sources (radiation damage makes higher fluxes less useful).

  $10^{13\text{-}14}$/second → Repetition rate ≈ 1-100 MHz

- Wave length

  For atomic resolution structural studies need

  $\lambda \leq 1.5$Å

- Coherence (focusing)

  Diffraction Limited

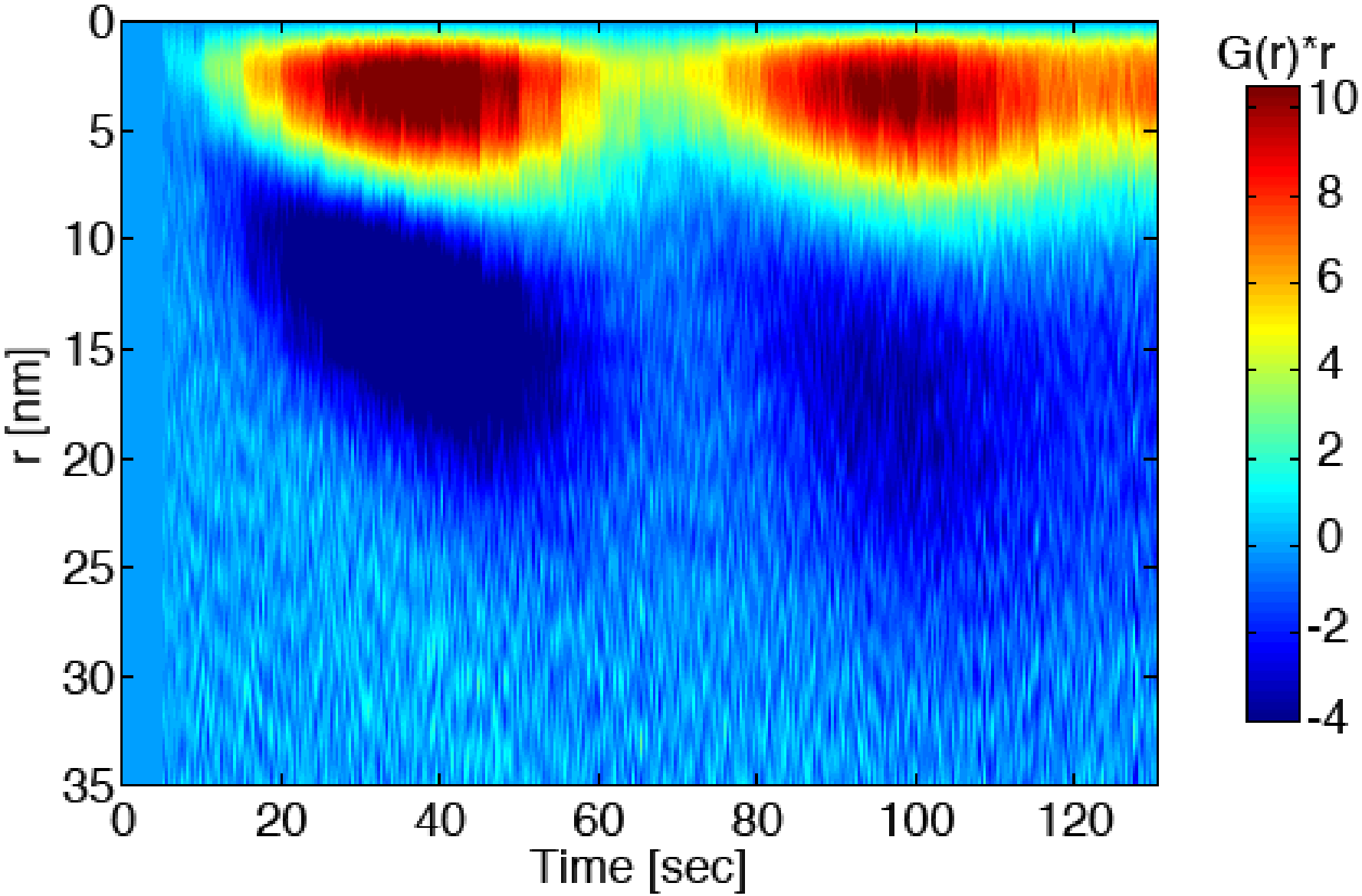
Next Steps: Going beyond line-shape analysis for diffuse scattering

$$I_{diff}(q_{\parallel}, t) = 2\pi |f|^2 \int_0^{\infty} \rho d\rho \, J_0(q_{\parallel}\rho) \, \langle \delta n(0,t) \delta n(\rho,t) \rangle$$

$$G(r,t) \equiv \langle \delta n(0,t) \, \delta n(r,t) \rangle$$

G(r,t) is maximum amount of information available in a scattering experiment

G(r,t)*r


0
5
10
15
20
25
30
35
r [nm]
0
20
40
60
80
100
120
Time [sec]
G(r)*r
10
8
6
4
2
0
-2
-4

Summary

- Need to push both time- and length-scales of our measurements to make progress.
- Moving away from periodic systems.
- Non-destructive, structural studies of biologically, chemically, and technologically relevant materials put tight constraints on properties of photon beams.
- For sources to have wide and deep impact, need to heed these constraints.

Acknowledgements

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