Frontiers of real-time

A. Structure of surfaces, interfaces, domains.
B. Dynamics: relaxation, fluctuations.
C. Resonant and magnetic scattering.
D. Organic films and interfaces.
E. Instrumentation.

ISR Brightness vs X21 and ESRF

- Surface scattering is moderately brightness limited.
- Polarization and XPCS are strongly brightness limited.

Source	Source type	Flux [ph/sec/mr/0.1%BW] *[ph/sec/0.1% BW]	Brightness [ph/s/0.1%BW/mm^2/mr^2]	Ge (001) –(2x1) (3/2,0) [measured cps]
NSLS X21	Wiggler	2×10^{14}	8×10^{16}	5,000
ESRF ID3	Undulator	*3×10^{14}	1×10^{19}	150,000
NSLS-II	IVU20	*1×10^{15}	5×10^{20}	--

Table 1 A comparison of flux and brightness for beamlines used for in-situ thin film growth studies; (1.5 0) fractional order peak on Ge(001)-(2x1). (ESRF measurement from Erik van der Vegt's PhD thesis.)

A. Surface structure

- Surfaces, Interfaces, Domains, Step terraces
  - Reconstructed surfaces, complex interfaces, phase diagrams (e.g. vs. oxygen partial pressure), etc.

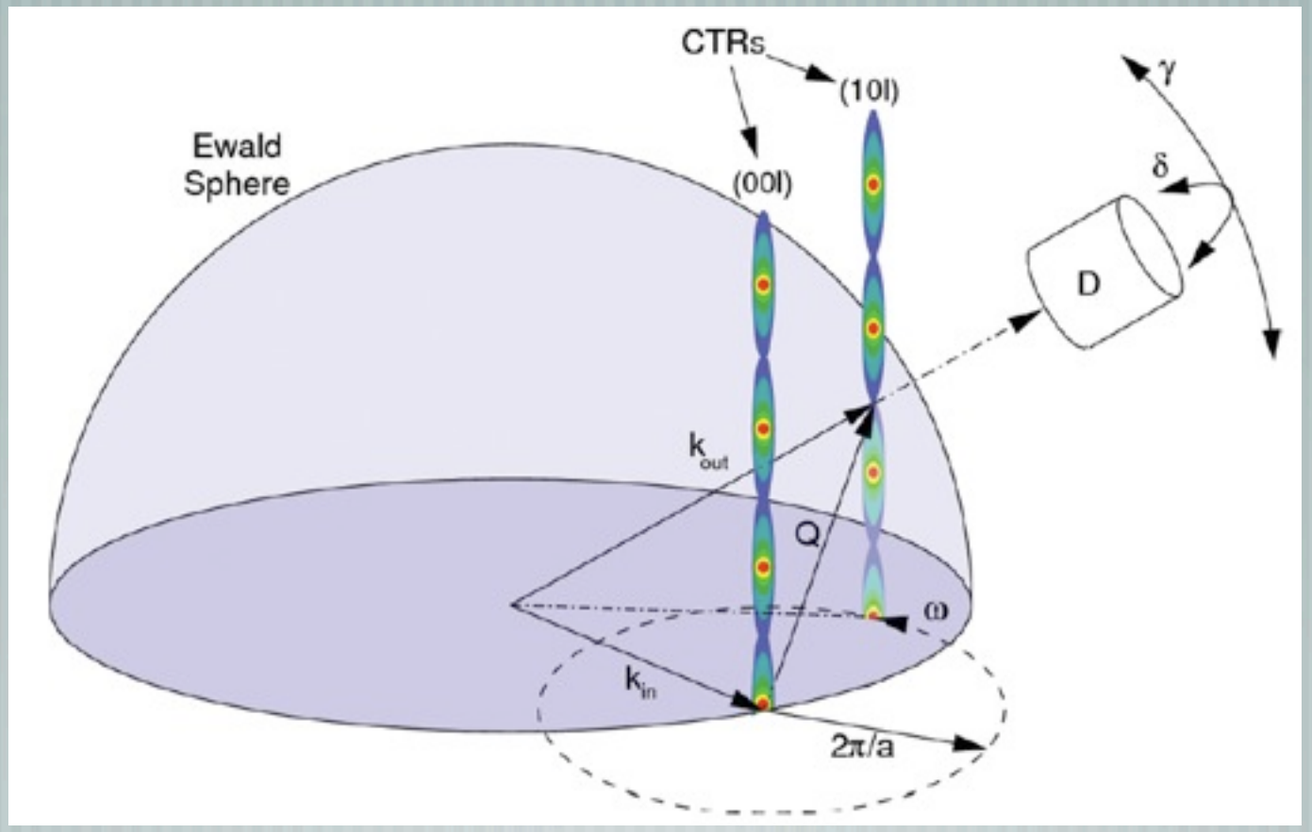
Science Case Perhaps not with the same level of accuracy as the ex-situ measurements because it will be hard to build an in-situ diffractometer with the required level of stability and accuracy. But with in-situ measurements, you can grow a monolayer and then determine the structure, repeat and follow how the structure evolves during film growth. This idea is particularly compelling for growth techniques such as ALD, for systems that exhibit properties that are strongly film thickness dependent (e.g. ferroelectrics, magnetics) , and for mapping surface phase diagrams (e.g. surface reconstructions vs oxygen exposure) Complementary In-situ use of resonance and polarization, etc.

User Demand Want a "house" chamber with most commonly needed capabilities (UHV, good access to reciprocal space, small evaporation cell) Flexibile infrastructure is important (Laser, gas handling, etc.) because each potential user has unique requirements in terms of film deposition and processing.

Performance The ISR beamline will be among the best in the world for this. Much depends on new detectors (high dynamic range, fast readout) and development of scanning and data reduction techniques.

Area detector methods

- Schleputz et al. 2005



- "on-the-fly" scanning [Eng and Rivers].

Real time studies

- Surface domain size vs. time, temperature, adsorption.
- Oscillations, step flow, Coarsening.

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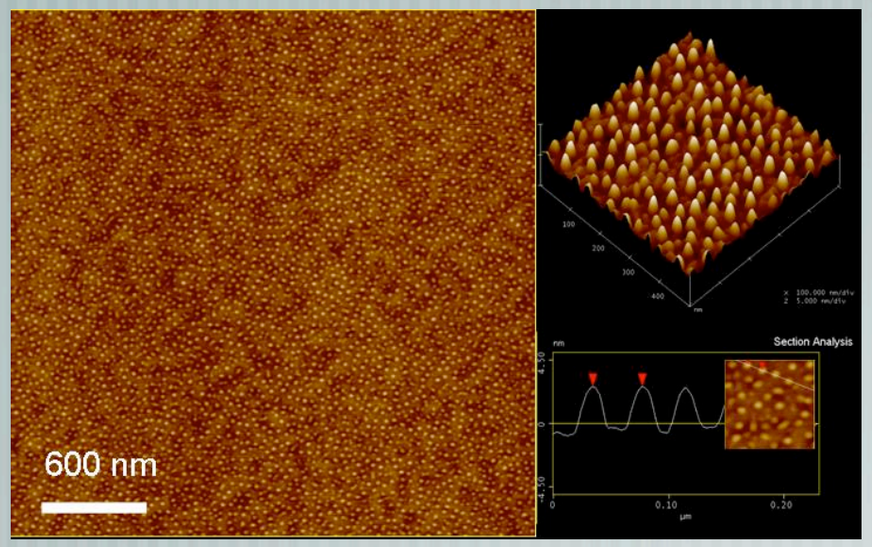
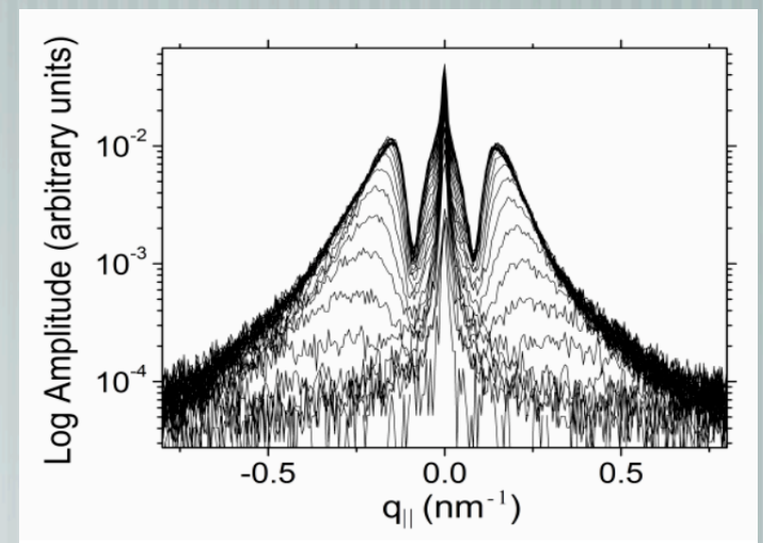
Nanodots on Si, Ozaydin et al, 2007





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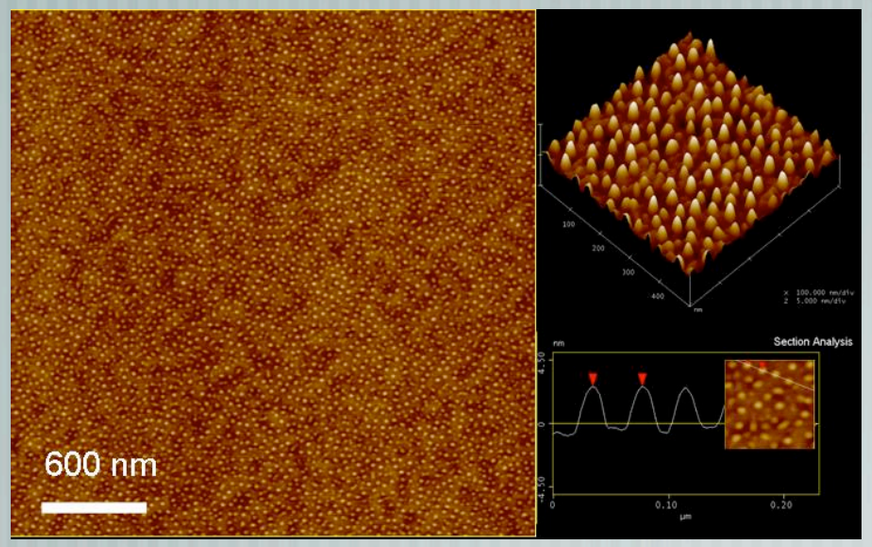

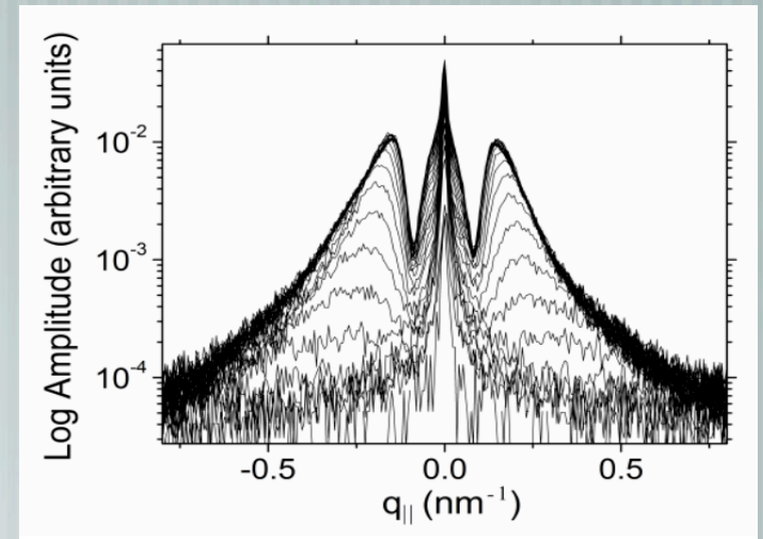

- Combine with anomalous, resonant, magnetic scattering.

B. Dynamics

- Kinetic roughening.
- surface phase transformations. [Pierce , PRL 2009]
- fluctuations near critical point.
- pulsed deposition and relaxation.

X-ray Photon Correlation

- Fluctuations continue after reaching steady-state but are not accessible through average structure factor.
  - Kinetic roughening
  - Surface reconstructions
  - Magnetic or orbital ordering

Speckle from surface

PRL 103, 165501 (2009) PHYSICAL REVIEW LETTERS week ending 16 OCTOBER 2009

Surface X-Ray Speckles: Coherent Surface Diffraction from Au(001)

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[1]*Materials Science Division, Argonne National Laboratory, Argonne Illinois 60439, USA*
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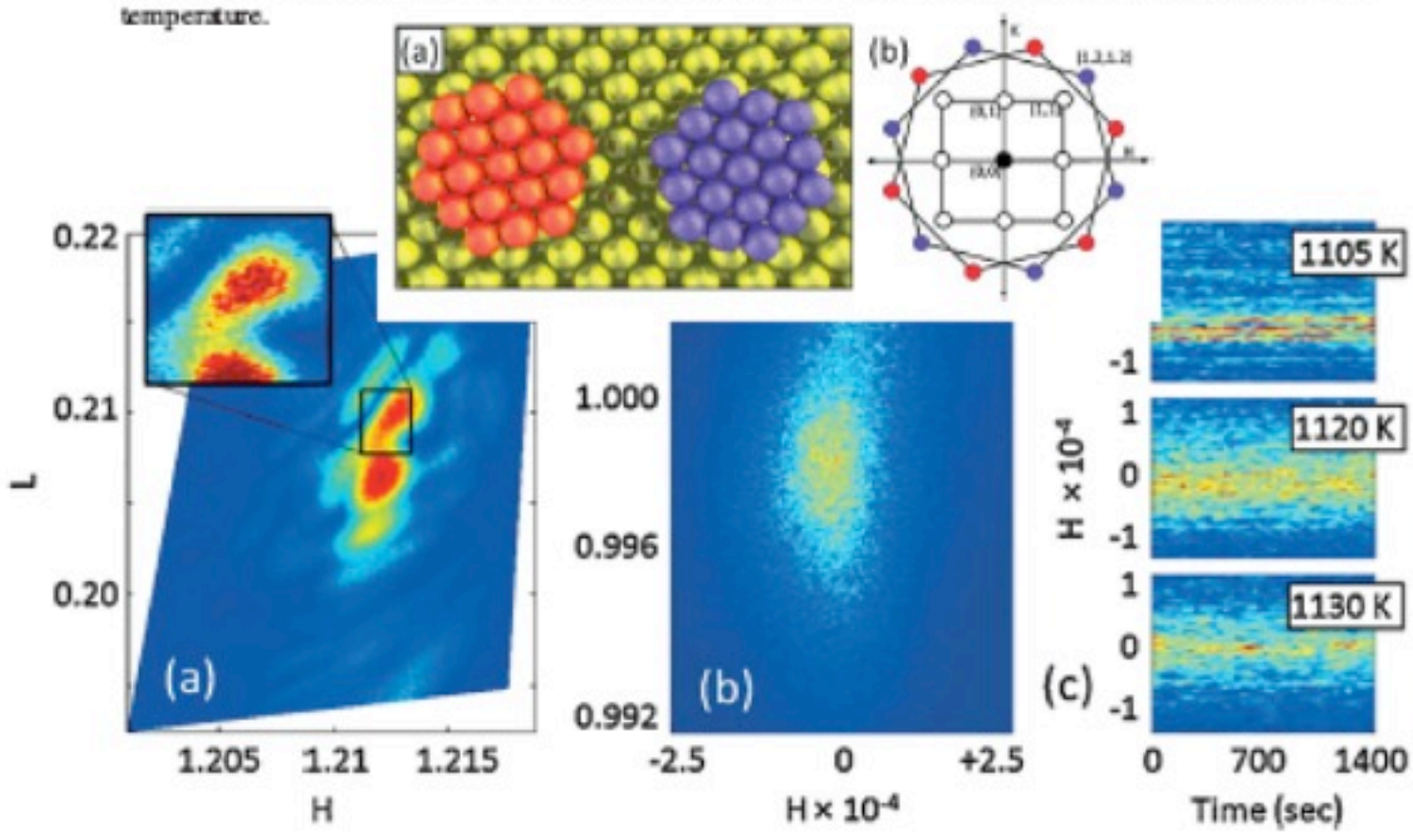
(Received 15 June 2009; published 15 October 2009)

We present coherent speckled x-ray diffraction patterns obtained from a monolayer of surface atoms. We measured both the specular anti-Bragg reflection and the off-specular hexagonal reconstruction peak for the Au(001) surface reconstruction. We observed fluctuations of the speckle patterns even when the integrated intensity appears static. By autocorrelating the speckle patterns, we were able to identify two qualitatively different surface dynamic behaviors of the hex reconstruction depending on the sample temperature.



Relaxation and coarsening

- Coarsening occurs in many thin film systems.
- Kinetic relaxation to steady state occurs in ion bombardment smoothing, kinetic roughening, phase transitions.
  → Get dynamics from XPCS.

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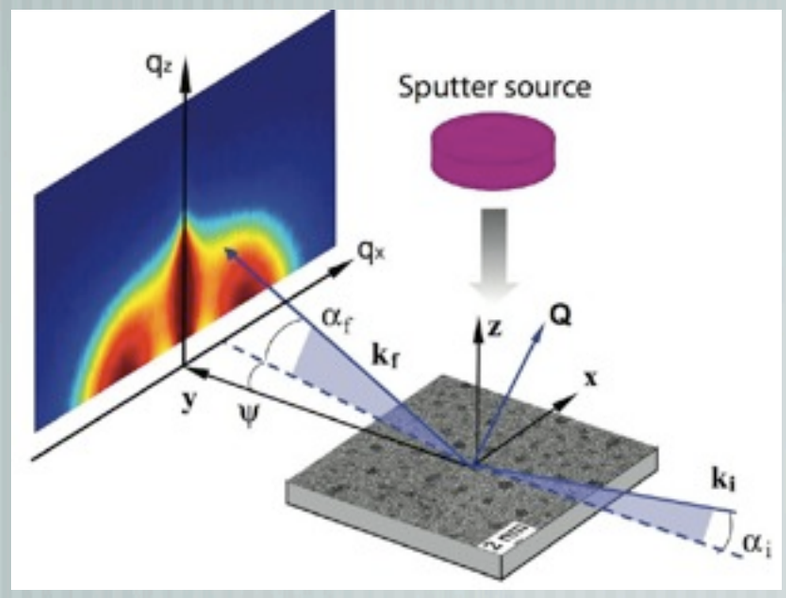
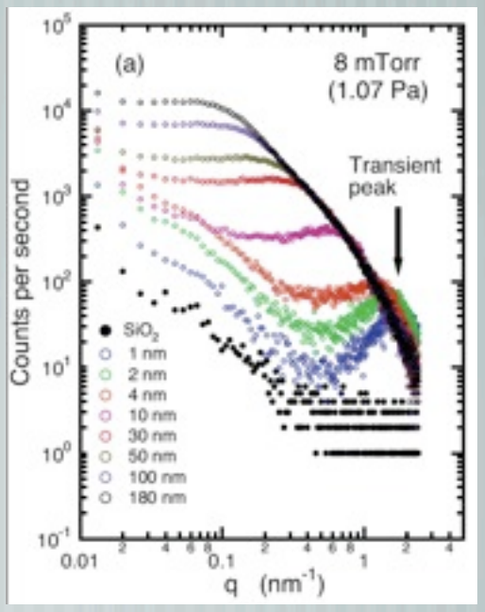
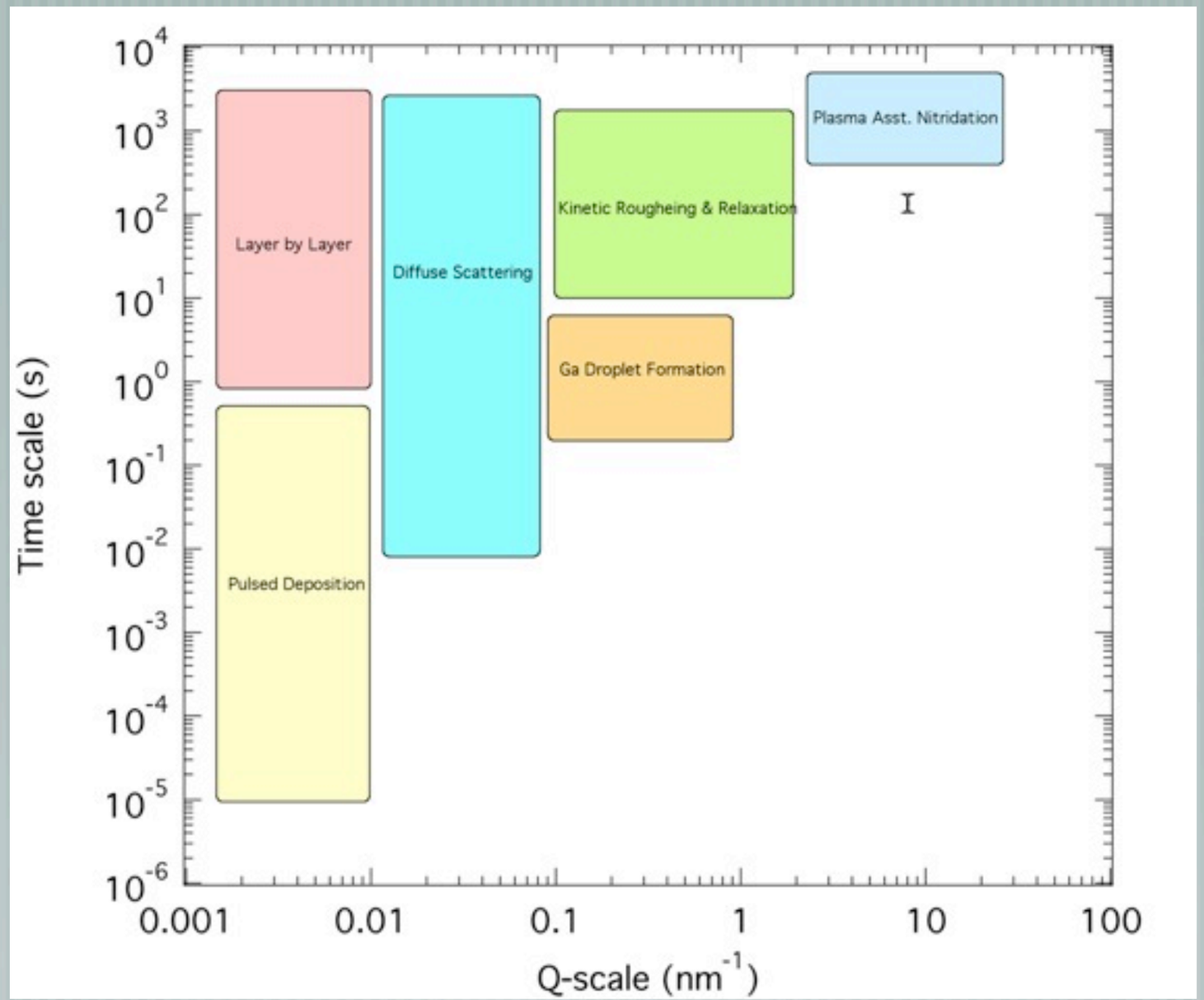
WSi2 Sputter Deposition, Zhou et al, 2010





Accessible Time and Q



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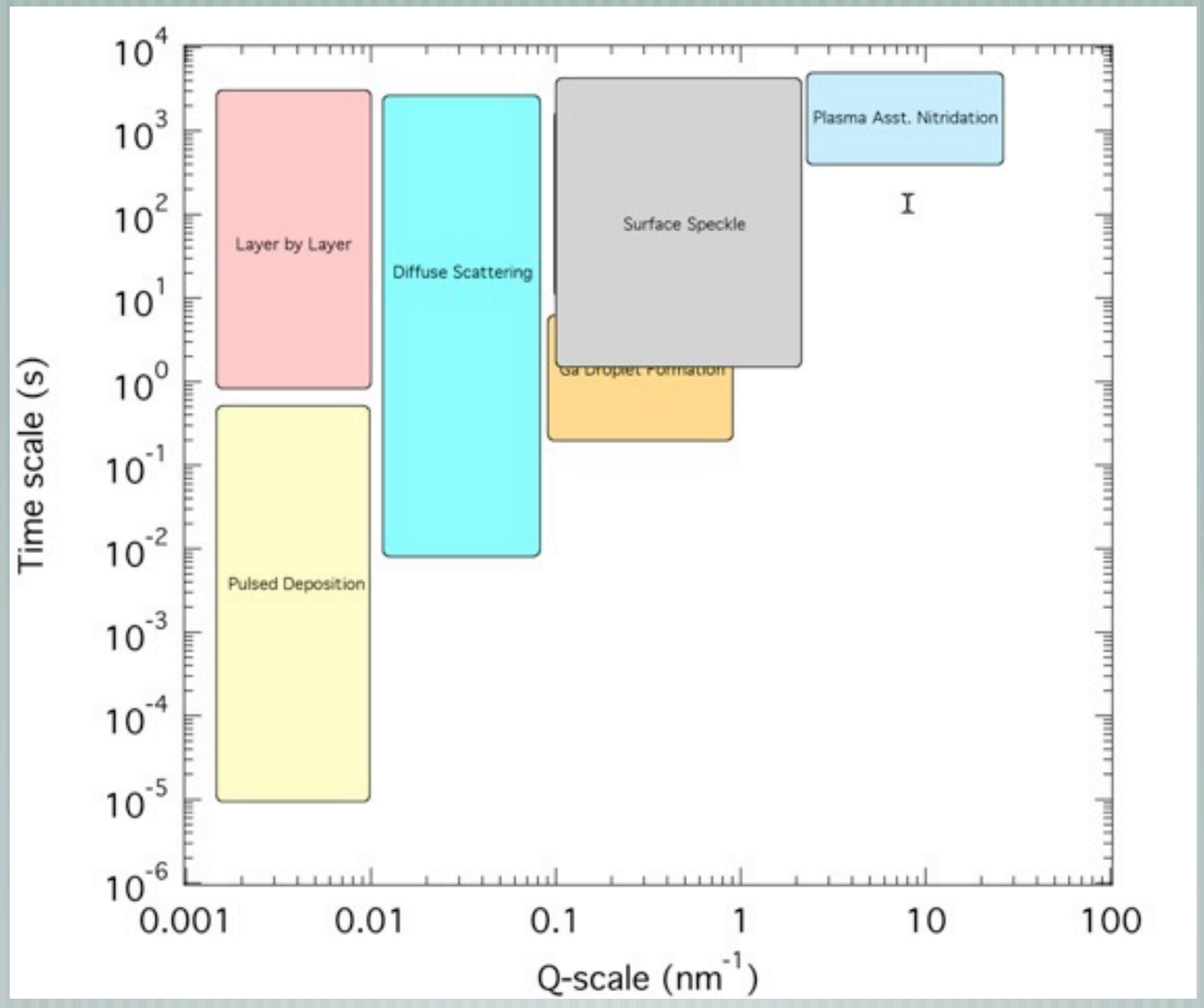

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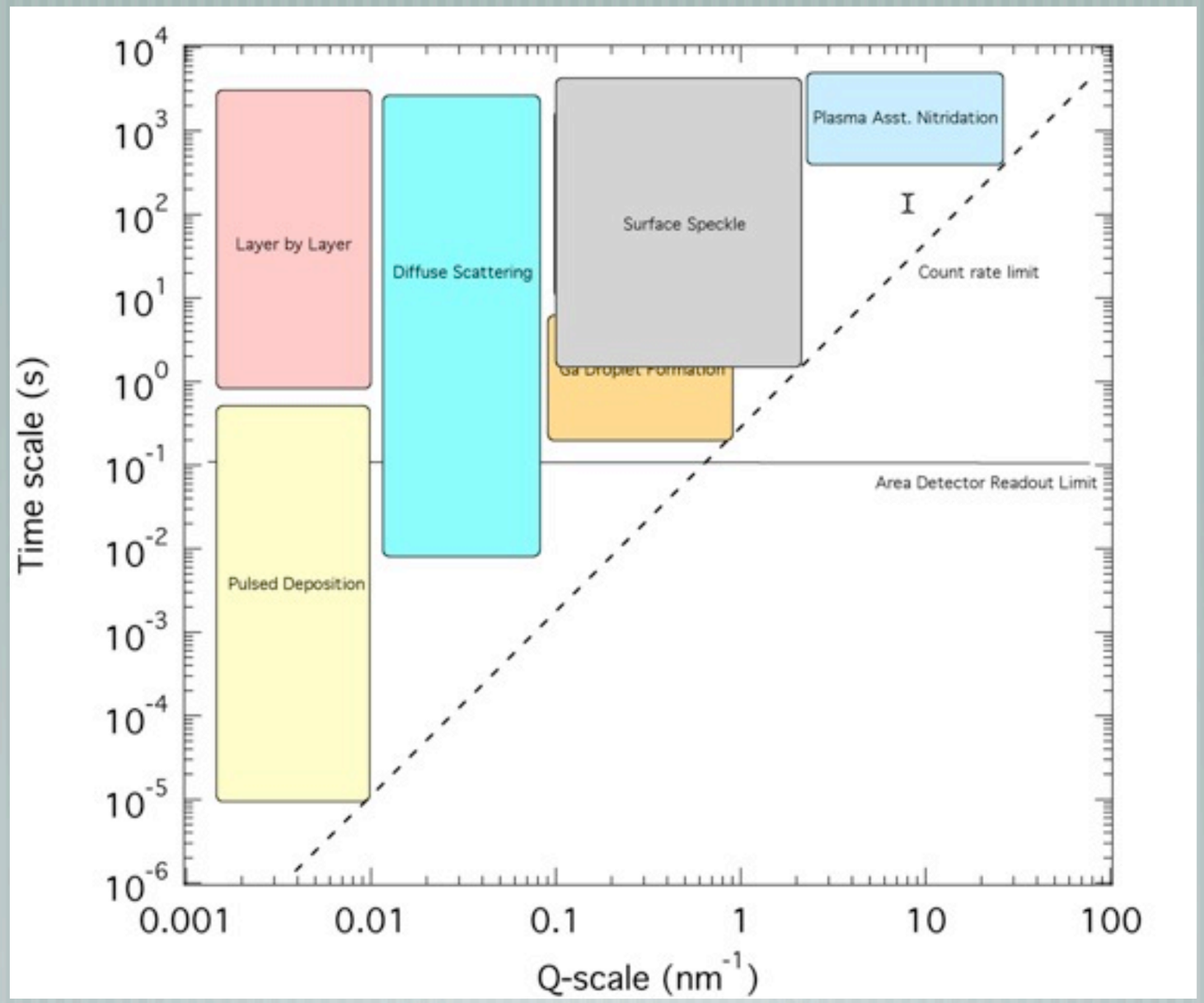

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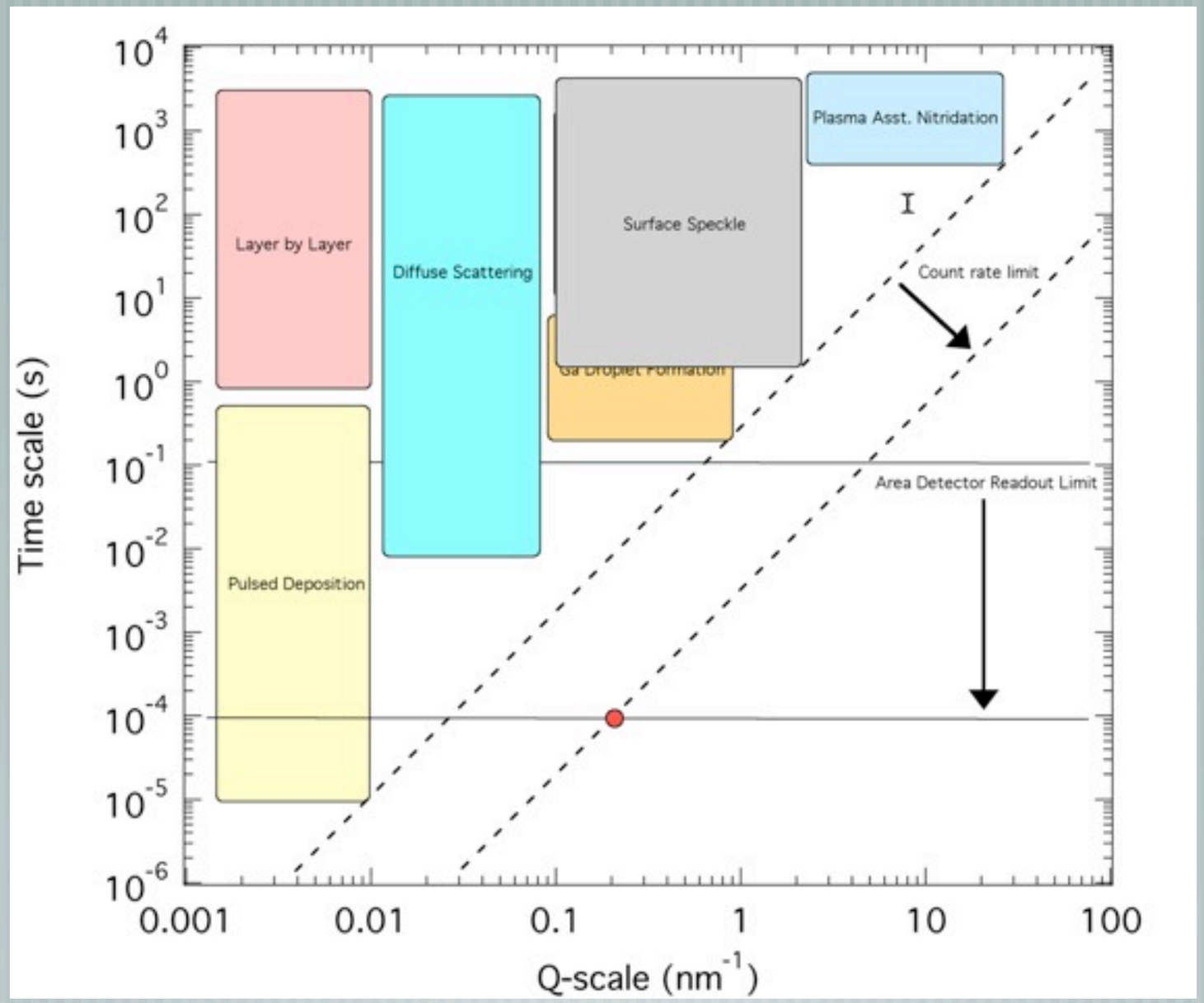
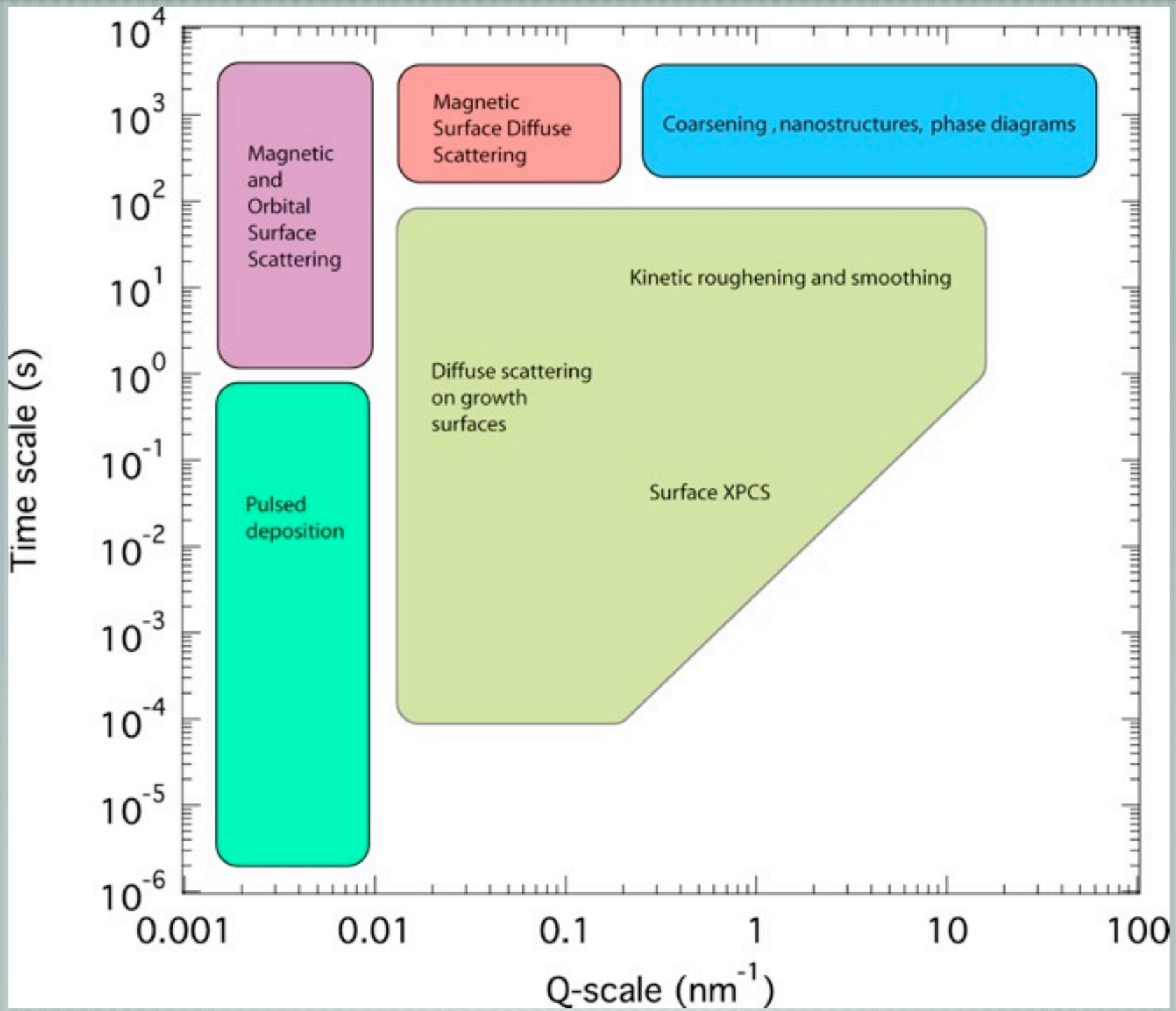
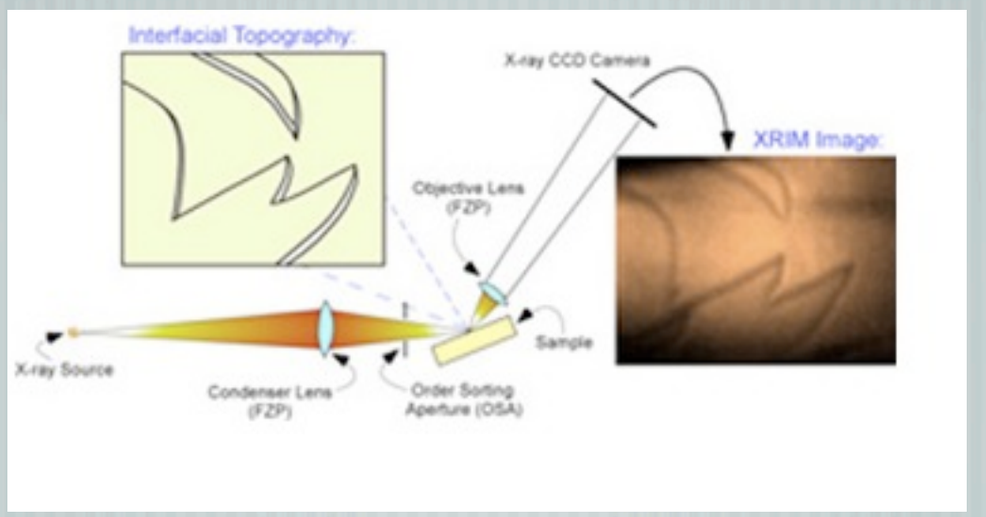


ISR Accessible Time and Q



Spatially resolved studies

- Small beam (1 μm) steps, domains. {ES B beam size??}
  - Domains decorate step structure.
  - Focussed ion beam or lithographically seeded arrays of nanostructures (growth in real time).
- Full field imaging, XRIM [Fenter, 2006]



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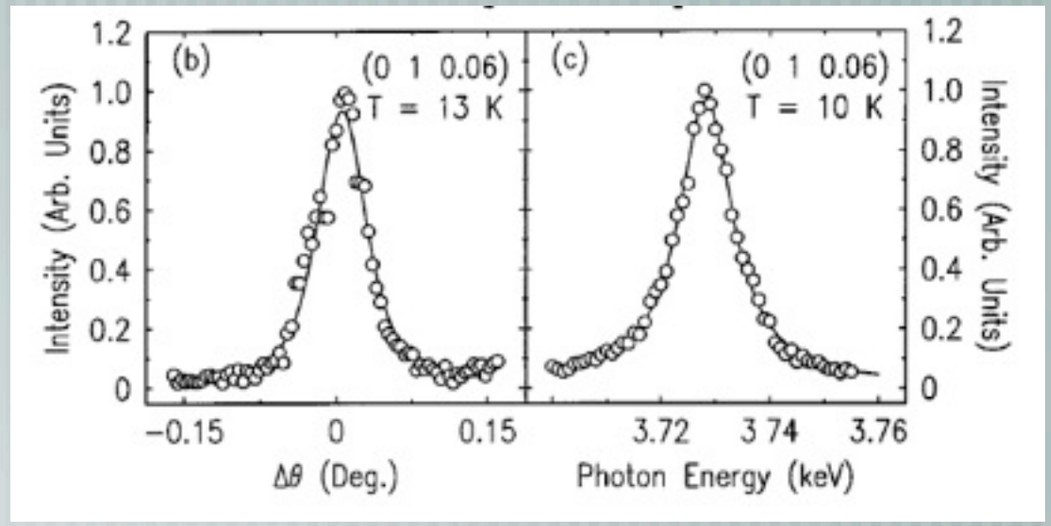

UO_2 surface magnetic scattering, Watson, 1996

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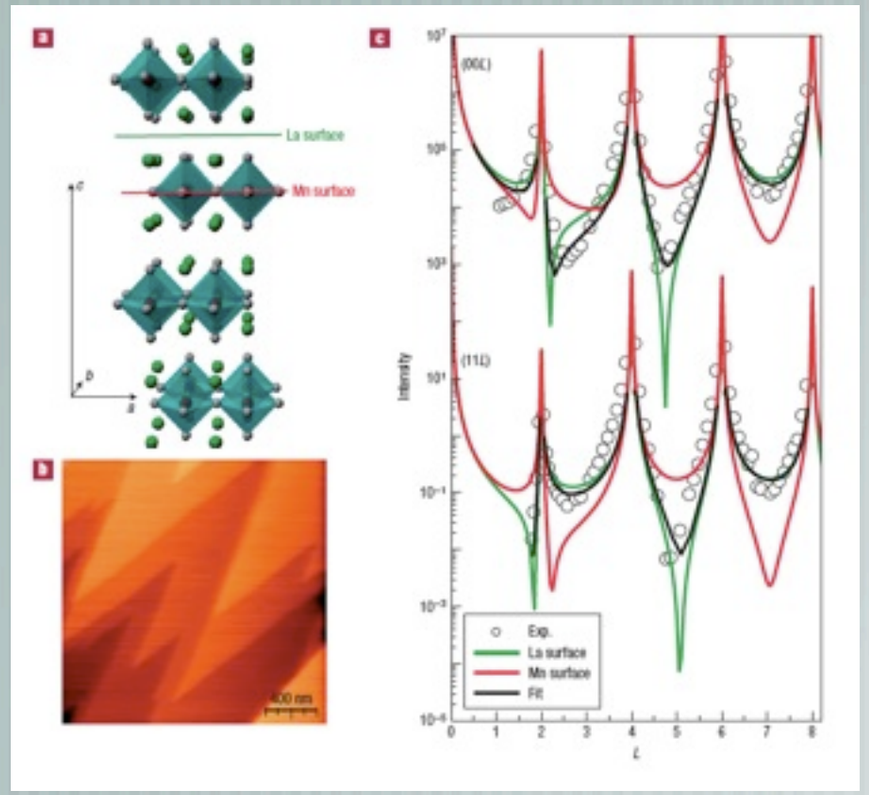
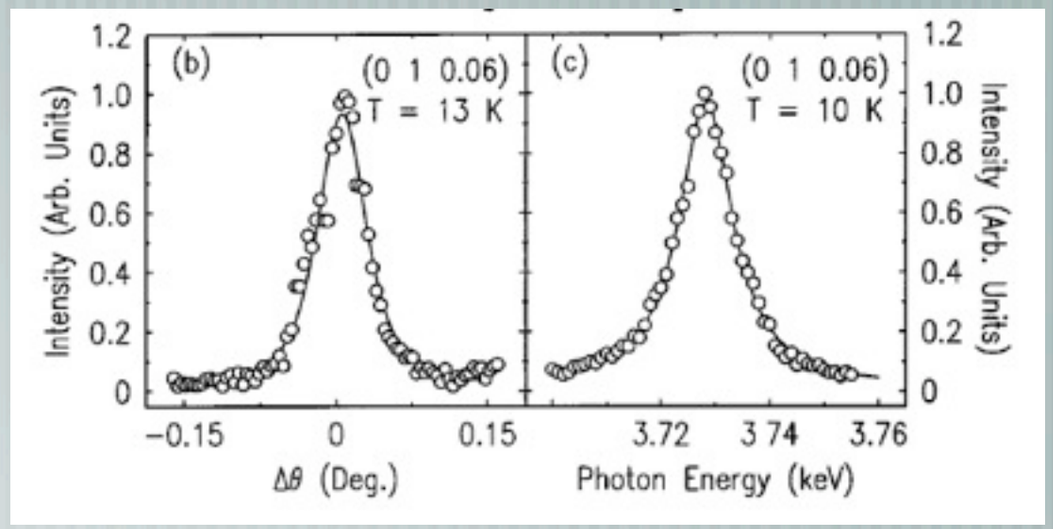


LSMO orbital surface ordering, Wakabayashi et al. 2007



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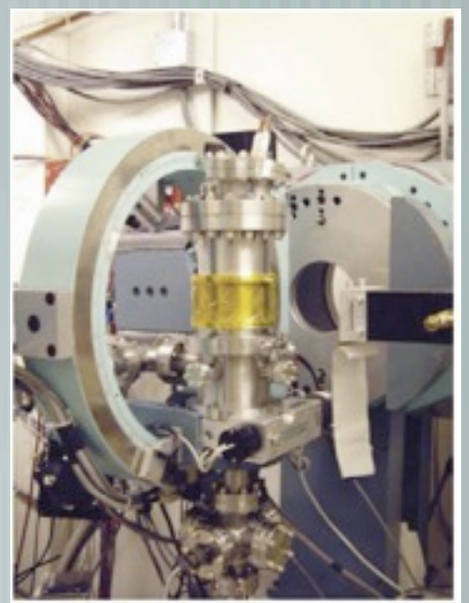

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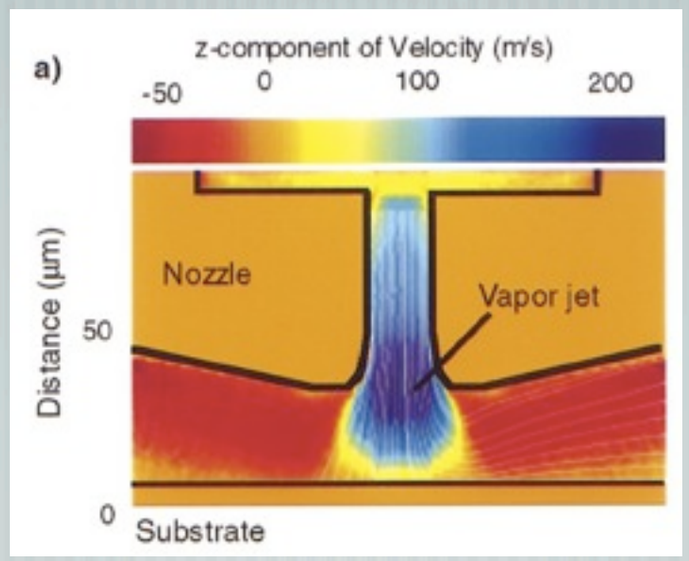
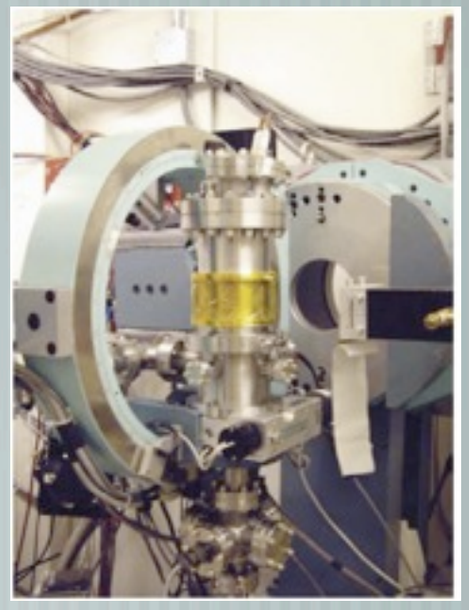


Shtein et al., 2004



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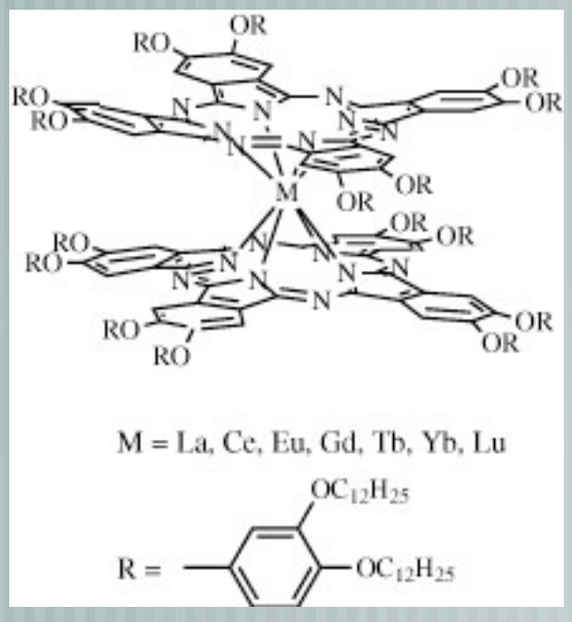

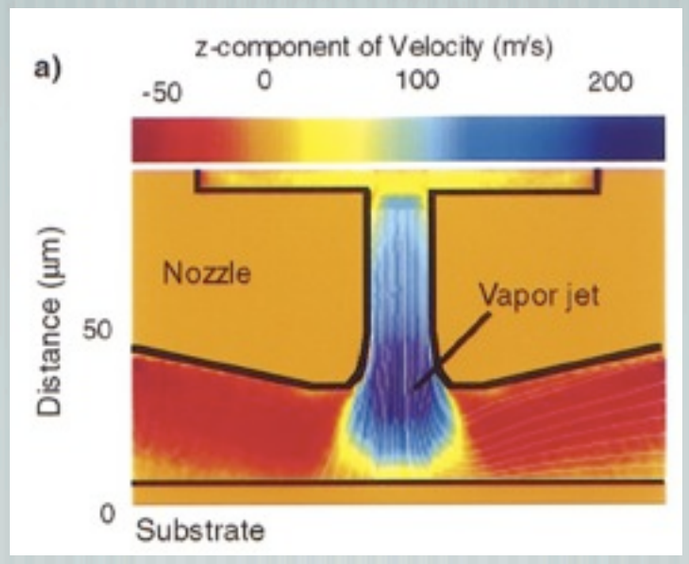

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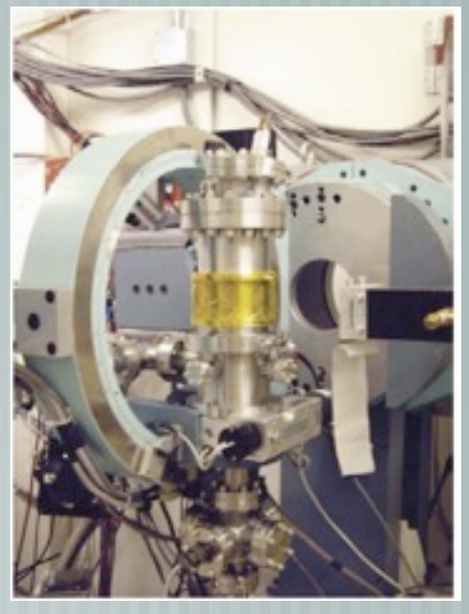
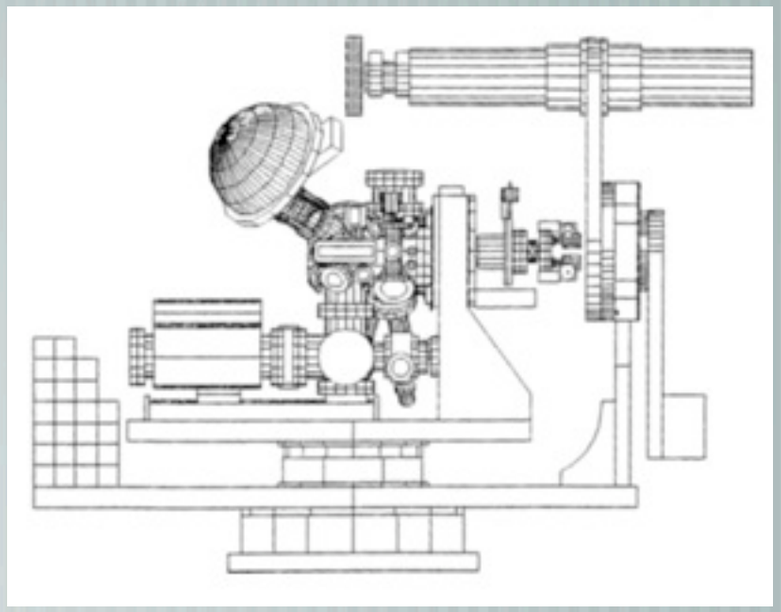


Headrick et al., 2004

E. Technical requirements

- Energy tunable, focussed beam 1 -100 μm, high coherence.
- Area detectors with small pixel size (10 μm), improved dynamic range, and fast readout.
- Diffractometer for medium to large chambers in "roll-on" mode.
- "House" growth chamber.⟶
- Nearby setup space.



Lyman et al., 1997