In Situ X-ray Interface Scattering

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- The need for UHV *in situ*
- All nano-science problems cannot be solved in real space: need scattering studies
- Recommendations for in **flexible** situ facility

Collaborators:
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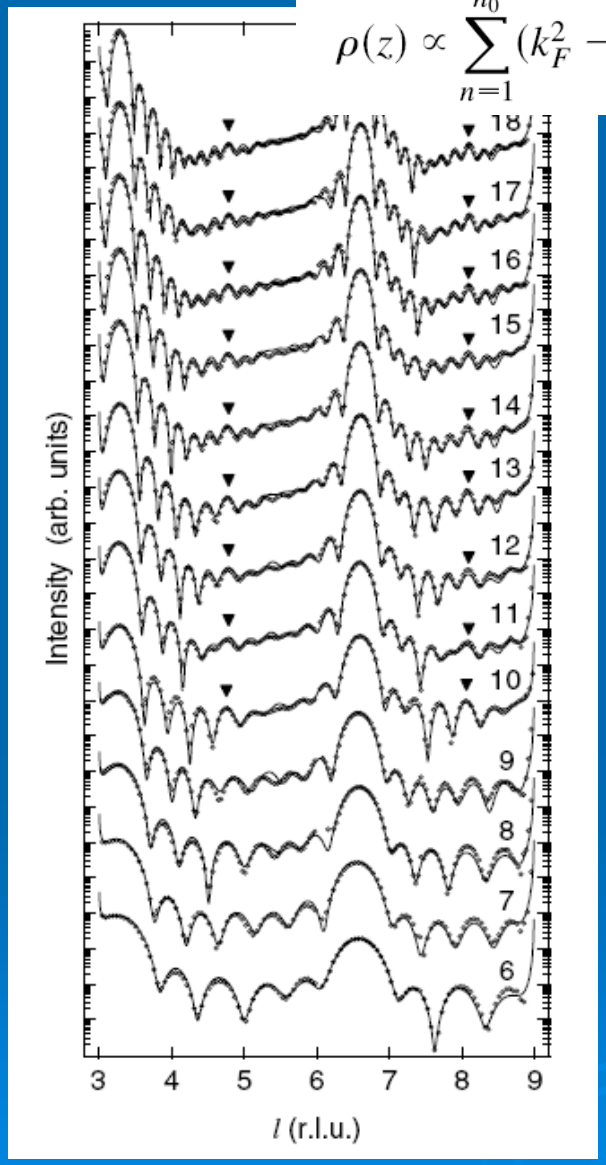
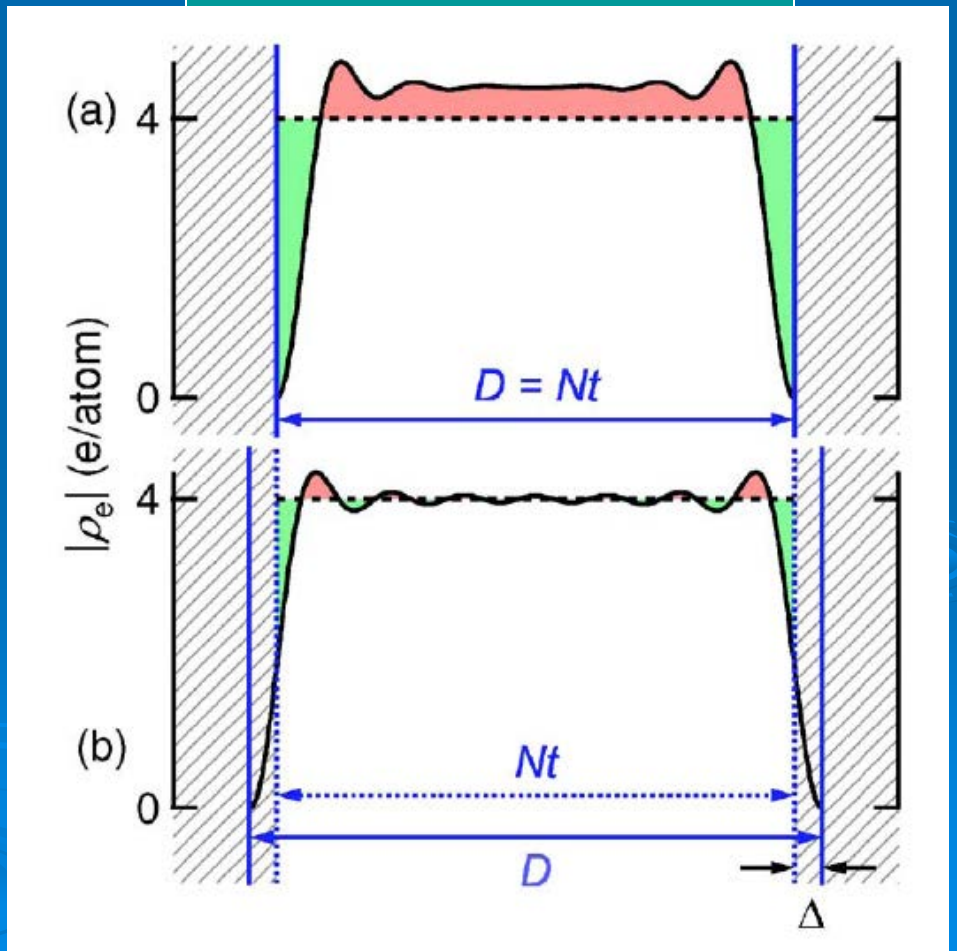
S.T. Hayden, Yiyao Chen, M.Tringides

Electronic Charge Density Oscillations – QM Confinement

$$\rho(z) \propto \sum_{n=1}^{n_0} (k_F^2 - k_z^2)\sin^2(k_z z),$$



In Situ Preparation: Essential



Czoschke, Hong, Basile, Chiang, PRB 72, 072402 (2005); PRL 91, 226801 (2003)

Quantum-Size-Effect Pb Nanocrystals on Si(111)7x7

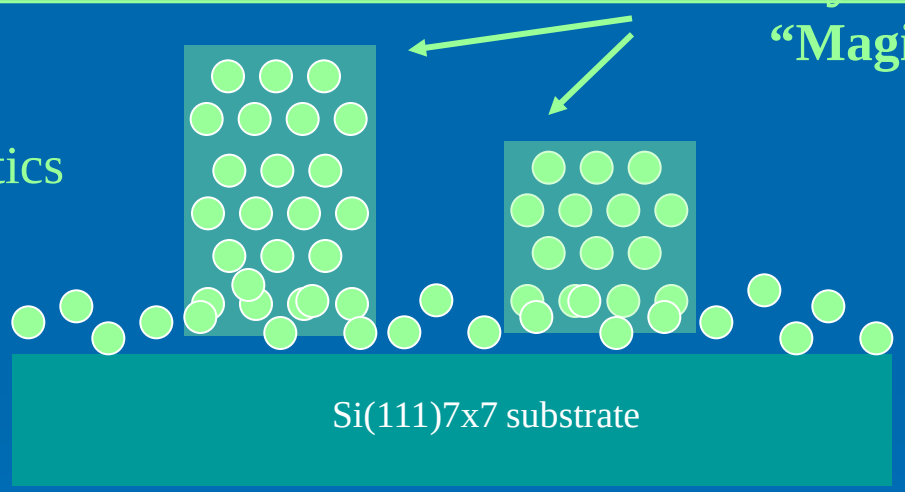
APS 6IDC

Quantum Mechanics Influences Nanocrystal Growth

"Magic Heights"

Discoveries:

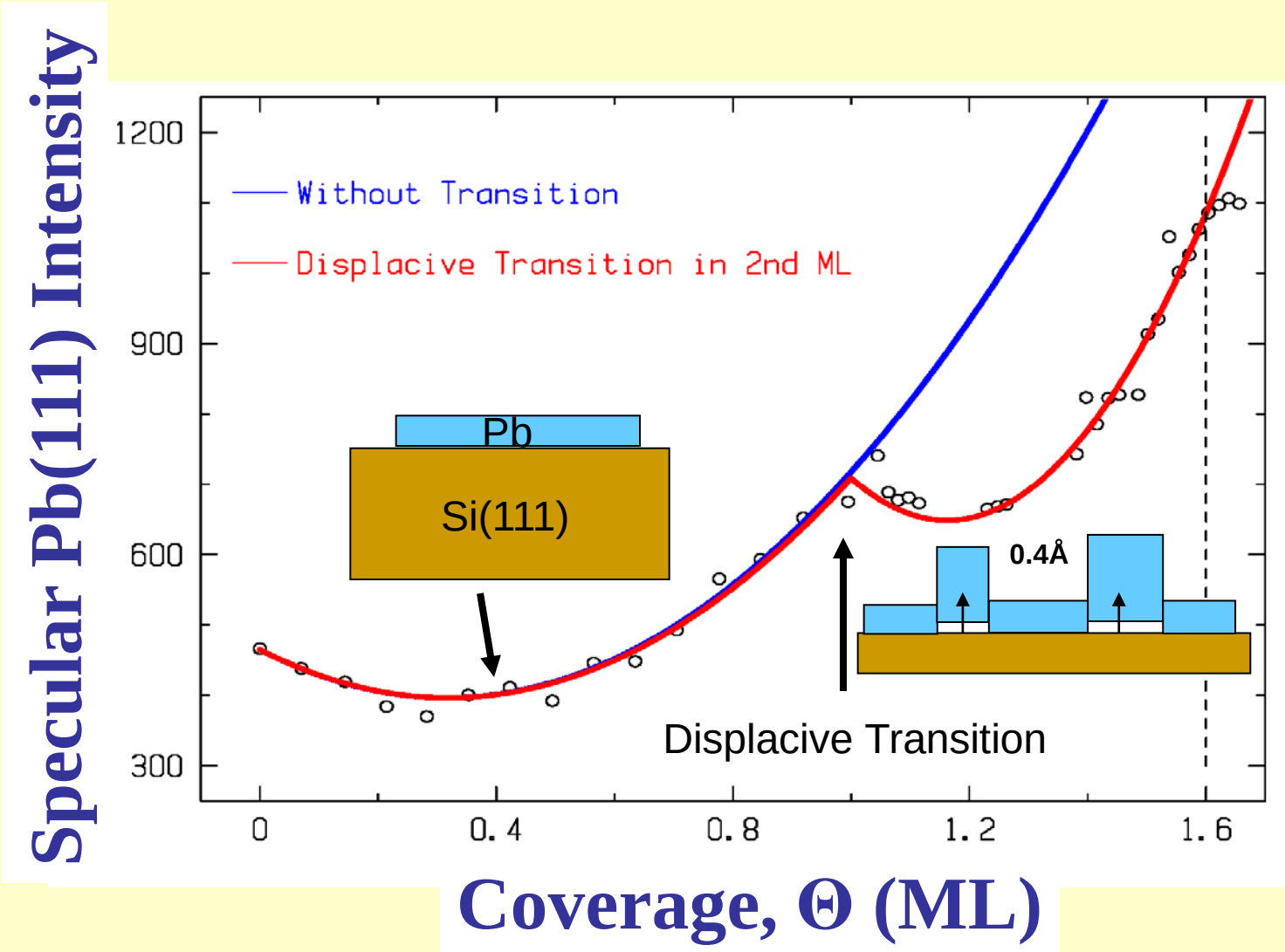
- anomalously (10^3) fast kinetics
- Non-classical coarsening
- Unusual behavior:
  fast growth => most stable structures



- 1st monolayer: wetting layer forms
- Higher coverage: islands form

C. A. Jeffrey, Conrad, Feng, Hupalo, Kim, Ryan, Miceli, Tringides. PRL **96**, 106105 (2006)

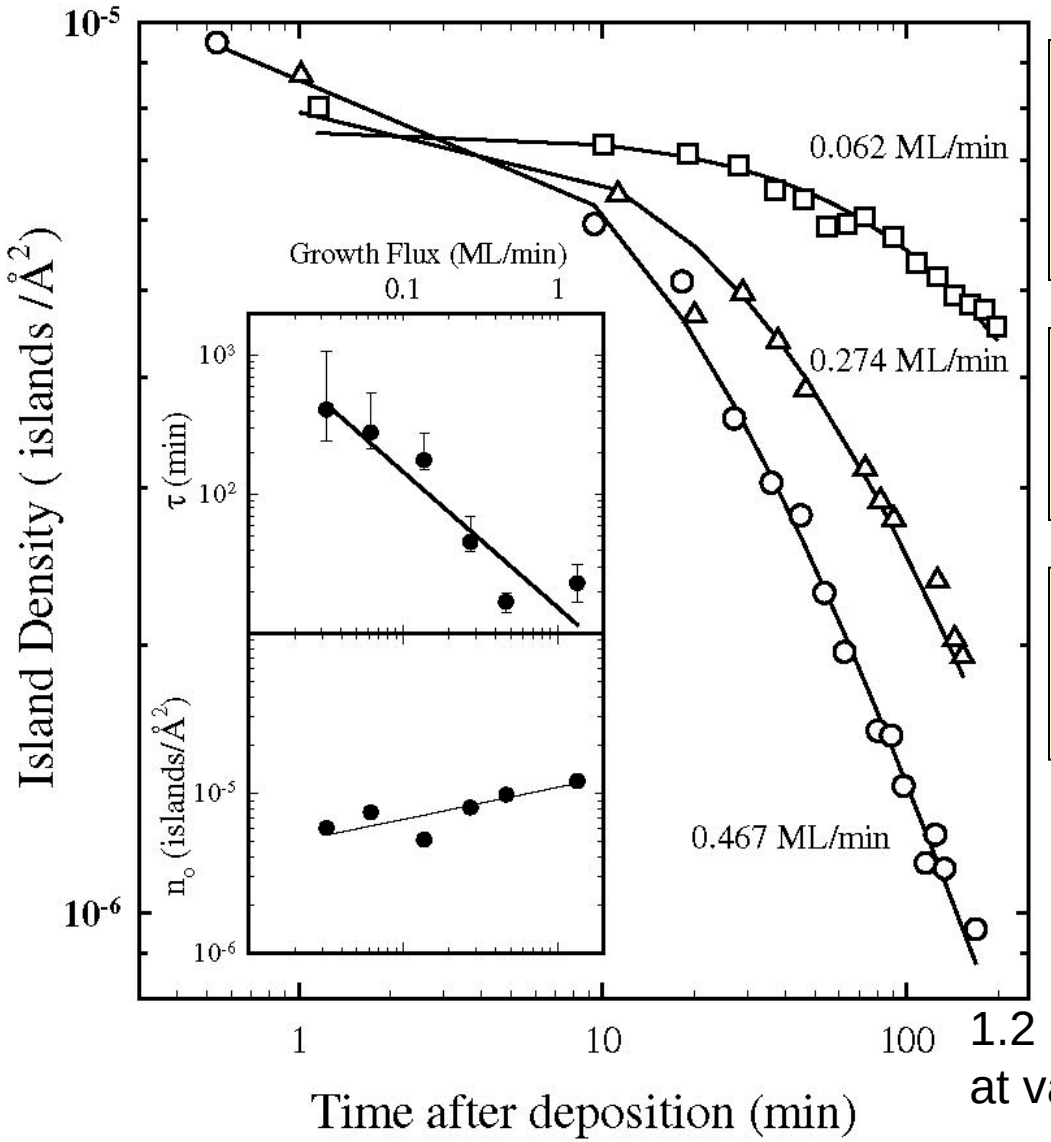
Islands consume the wetting layer & move away from the interface



Pb Nanocrystals: Non-Classical Coarsening

Effect was missed by STM; Seen by In Situ X-ray Scattering

C. A. Jeffrey et al., PRL **96**, 106105 (2006)



- Island densities do not approach each other at long times:
  QSE ⇒ Non-Oswald
  Breakdown of Classical Coarsening

- Time constant τ ~ **1/Flux**
  Strong flux dependence!
  Unexpected!

- Anomalously fast relaxation
  ~1000x faster than expected!
  ⇒allows equilibrium!

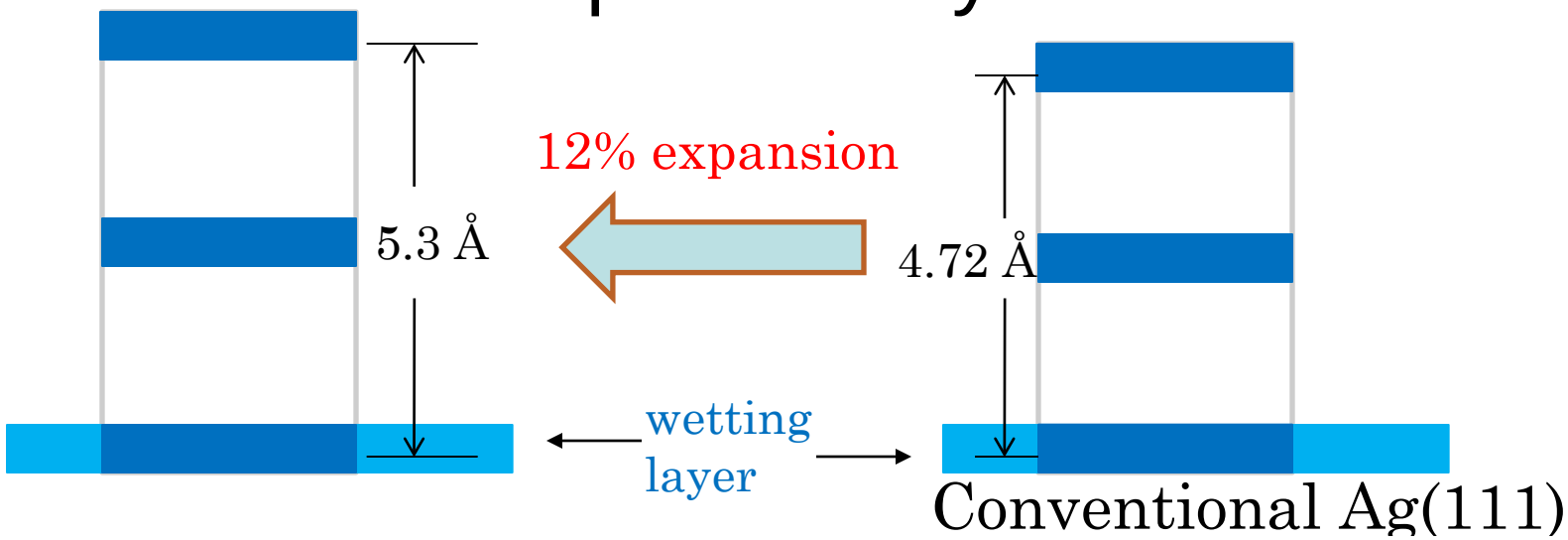
$$n(t) = \frac{n_0}{\left(1 + \frac{t}{\tau}\right)}$$

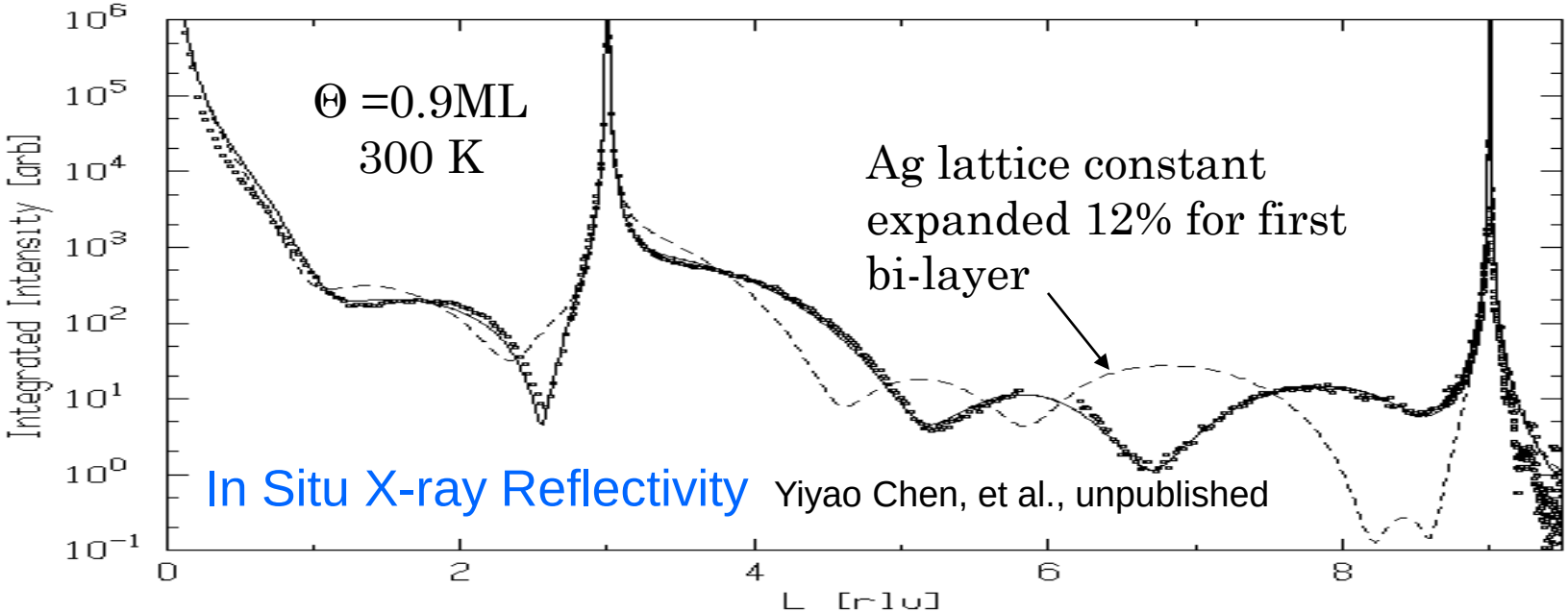
1.2 ML of Pb at 208K
at various flux rates

Ag/Si(111)7x7:

Gavioli et al., PRL **82** 129 (1999);
Goswami et al., Surf Sci. **601** 603(2007);
Unal et al., PRB **81**, 085411(2010)

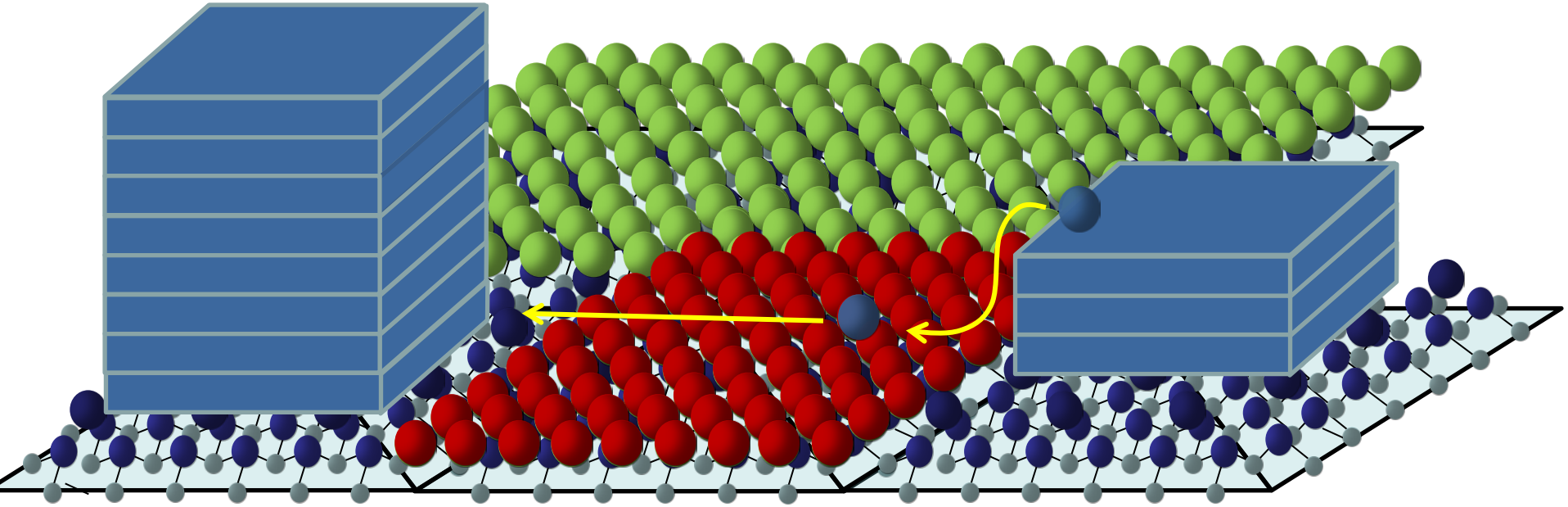
12% anomalous expansion by STM?





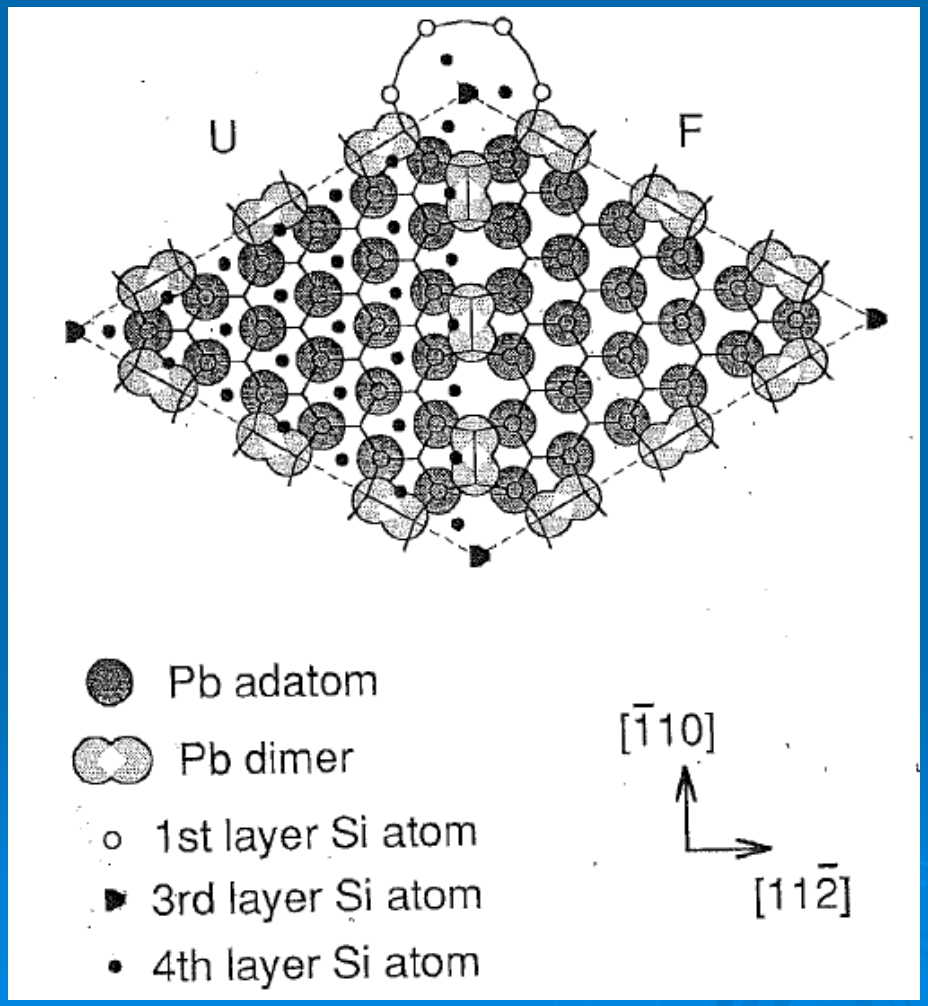
Coarsening of Pb QSE Islands is Facilitated by the Wetting Layer

What is the structure of the Pb Wetting Layer on Si(111)7x7?

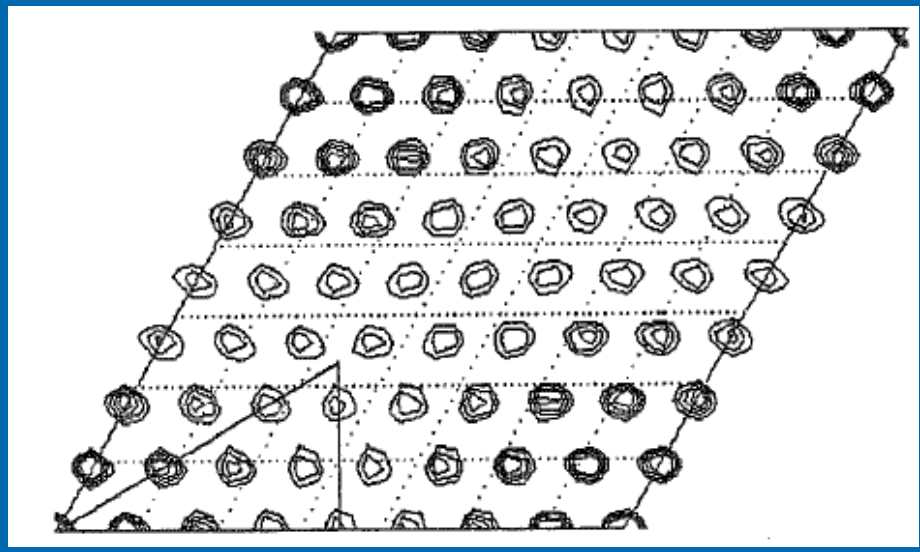


Pb Wetting Layer Structure on Si(111)7x7

STM Structure: Pb7x7 (in situ)



X-ray Structure: Pb8x8 (ex situ)

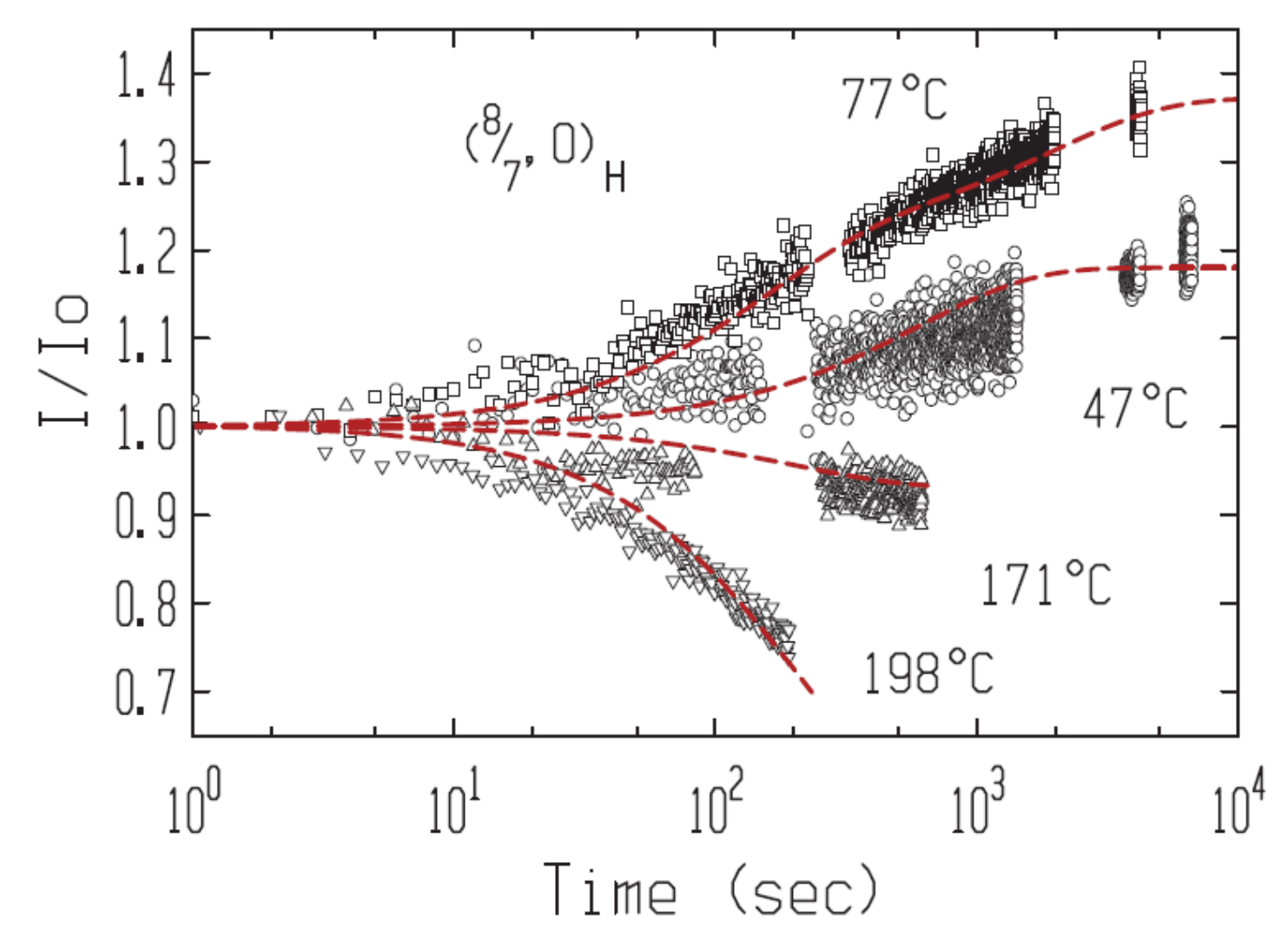


R. Feidenhans'l, F. Grey, M. Nielsen, R. L. Johnson, *Kinetics of Ordering and Growth at Surfaces*, ed. M.G. Lagally, (Plenum 1990)

I.S. Hwang, R.E. Martinez, C. Liu, G. Golovchenko, Surf. Sci, **323**, 241 (1995)

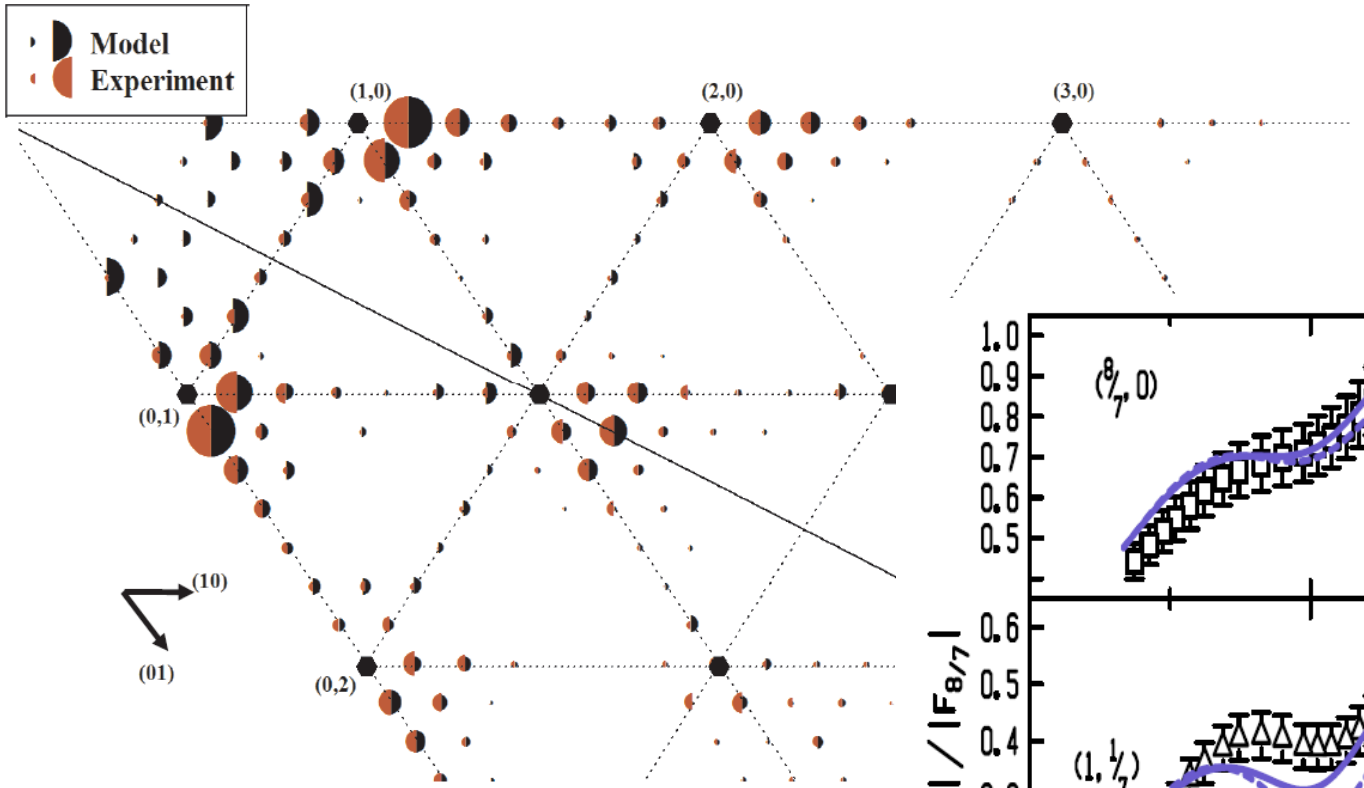
In Situ **X-ray Diffraction Studies**: Non-equilbrium Wetting Layer

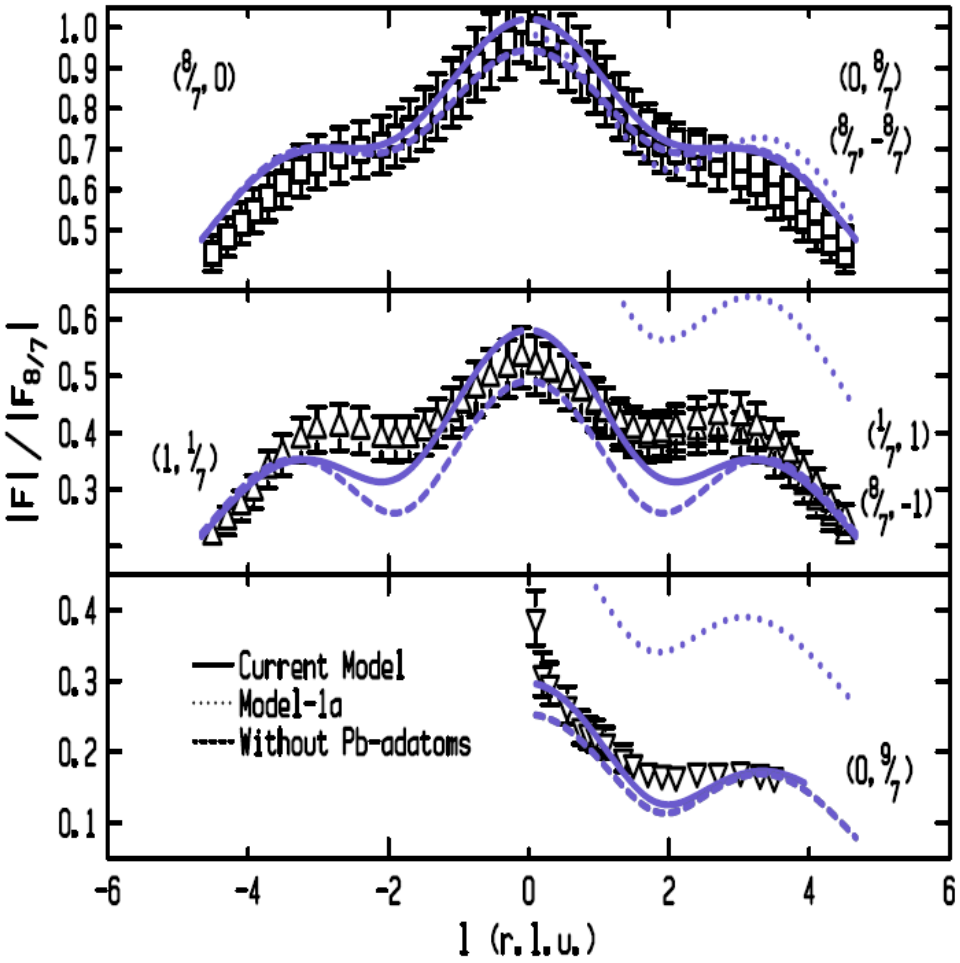
- Structure changes over span of 200 C
- Need in situ diffraction to see non-equilibrium behavior



M. W. Gramlich et al., Phys. Rev. B **84**, 075433 (2011).

In Situ Preparation: correctly anneal into equilibrium structure



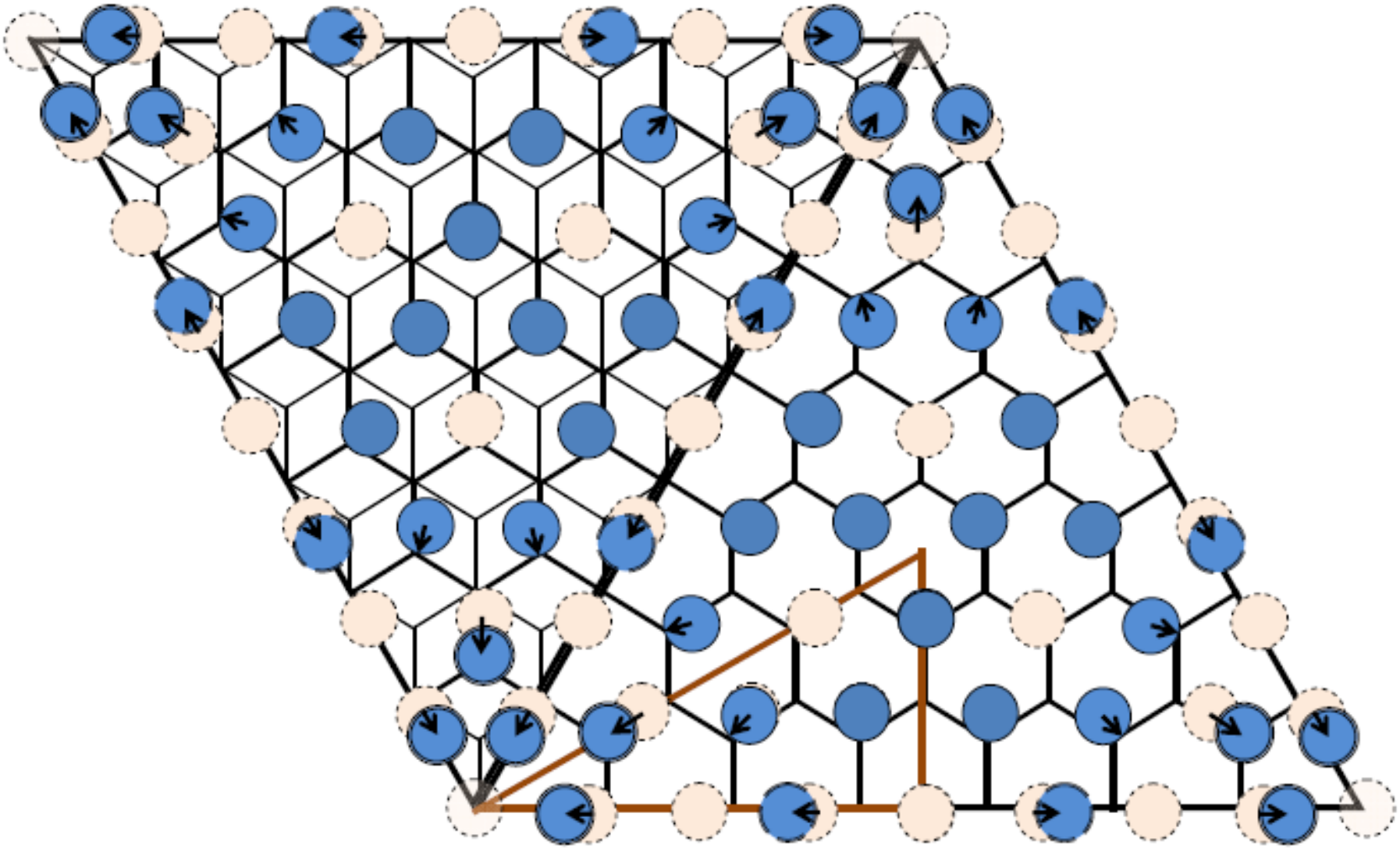


- 2D Diffraction
- Crystal Truncation Rods for fractional and integer rods

M. Gramlich et al., unpublished

Equilibrium Structure from X-ray Scattering

M. Gramlich et al., unpublished



Need For UHV In Situ Studies

- *Fabrication*: Most nano-materials must be fabricated in situ
  - large surface area
  - sensitive to environment

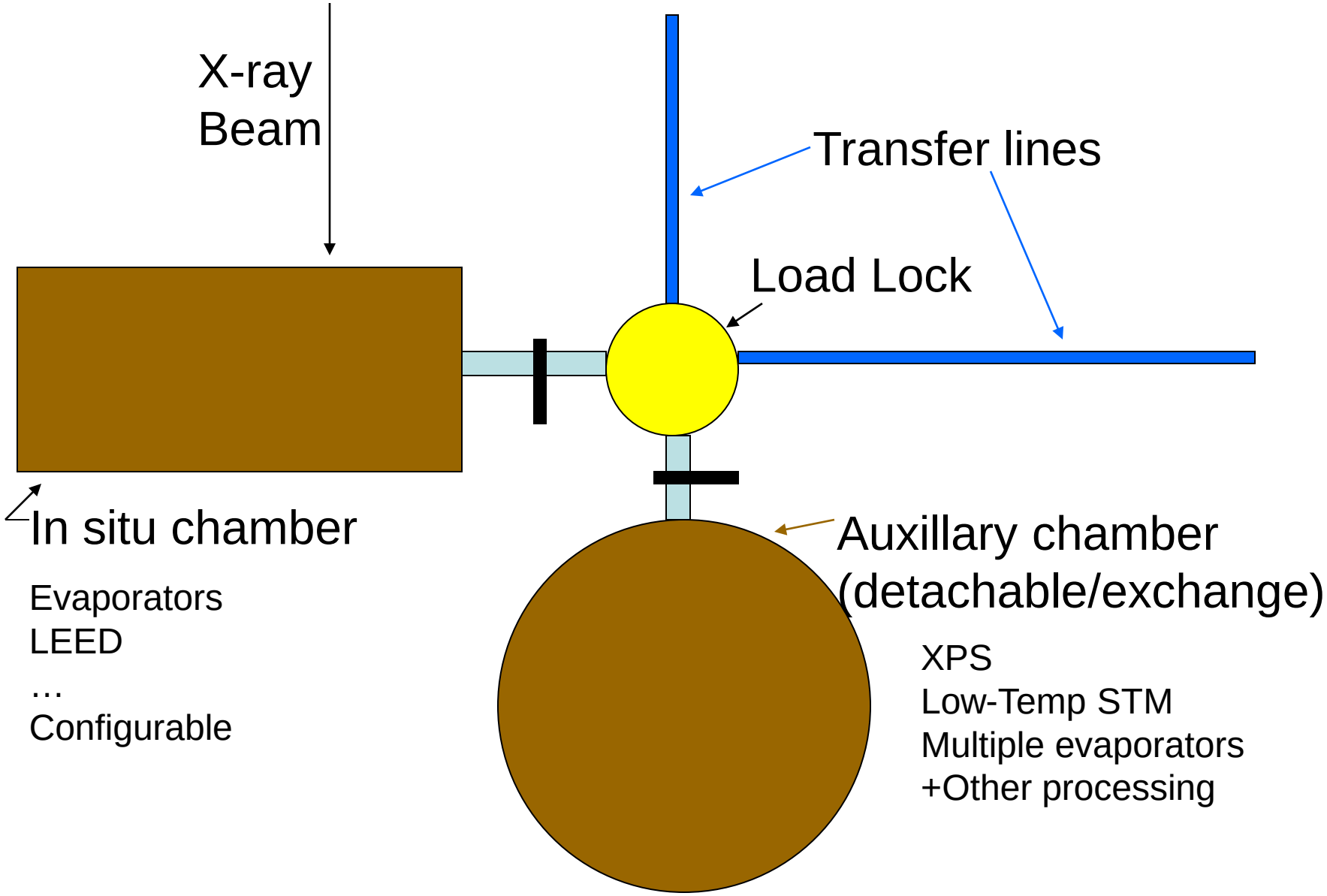
- *Modification*: science through physical evolution
  - temperature
  - time
  - coverage, growth, flux rate
  - gas exposure

- Nano-scale science involves interfaces
  - with environment
  - with supporting surface

Future Capabilities

- Flexibility is critical
  future is unknown; facilitate new ideas
- Science is about mechanisms: simple systems!
  Deposit (evap) small quantities and study
  Metals & Si on anything – always important
- Flexible Design:
  10 K < T < 1500 C
  - Multiple evaporation sources; gas handling
  - Ancillary tools: LEED, Auger, XPS, sputtering…
  - Low Temp STM: correlate with other studies
  - Broad reciprocal space range (160° x 90°)
  -
- Technical Support is critically essential

Schematic of a flexible instrument lay out



Technical Support/Expertise

- Look to Neutron Sources as example:
  - 1 to 2 “Instrument-Responsible” scientists plus assistants for each instrument!
- Know & develop the instrument
- Reliability
- Cultivate the user base
- Bring in NEW users who would not otherwise come