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# *Mapping Atomic Structure at Epitaxial Interfaces*

*Roy Clarke, University of Michigan,  
Ann Arbor, MI*



Opportunities for interface science at the ERL

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[royc@umich.edu](mailto:royc@umich.edu)

ERL X-ray Science Workshop:  
Almost Impossible Materials Science, June 16, 2006

# Outline

*At nanoscale, interfaces are everything*

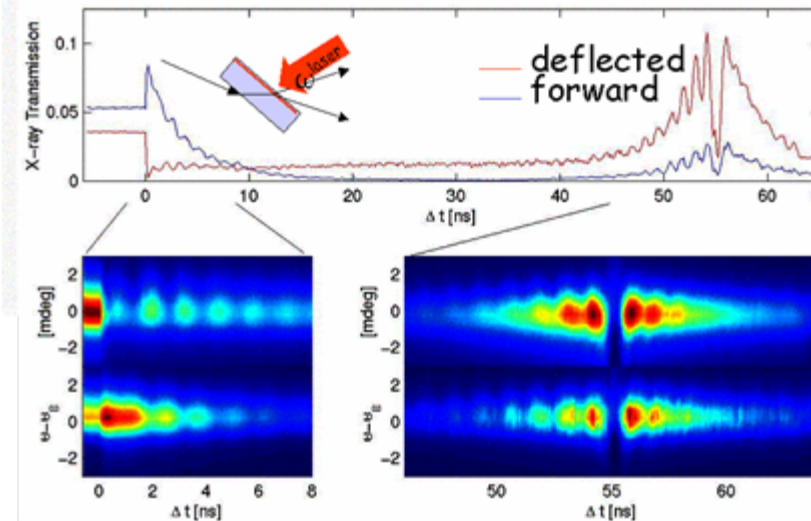
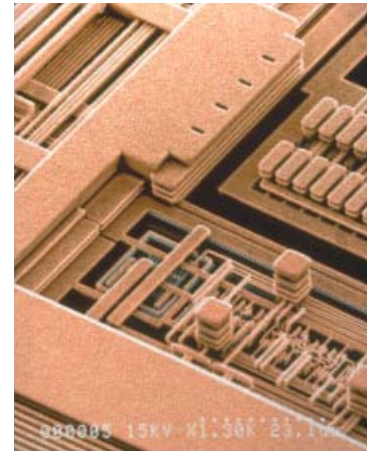
- **Possible now:**

- 3D maps of static thin-film/interface structure with atomic resolution

- Some dynamic information, including strain, phonons, domain walls...

- **Opportunities for ERL:**

- ultrafast imaging at atomic scale
  - tracking growth, deposition processes
  - local probe of nano-structures

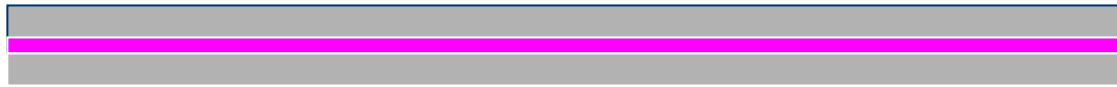


DeCamp, Reis et al. Nature, PRL

# Epitaxial Nanostructures

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Van der Merwe layer-by-layer growth



Quantum wells,  
Superlattices,  
Tunneling devices

Stransky-Krastanov mixed growth



Quantum dots,  
self-assembly

Volmer-Weber island growth



Surface nano-  
patterning

# *Epitaxial Nanostructures*

(example: "6.1Å system")

No-common-atom superlattices

## InAs-on-GaSb:

...Sb-Ga-Sb-Ga-As In-As-In-As...

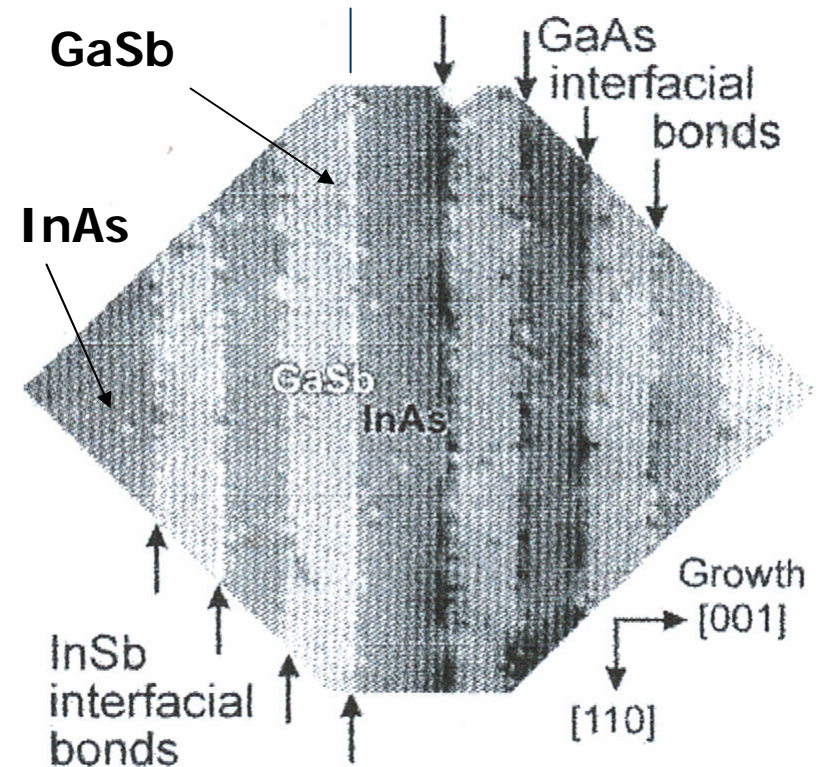
## GaSb-on-InAs:

("inverted interface"):

...As-In-As-In-Sb-Ga-Sb-Ga...

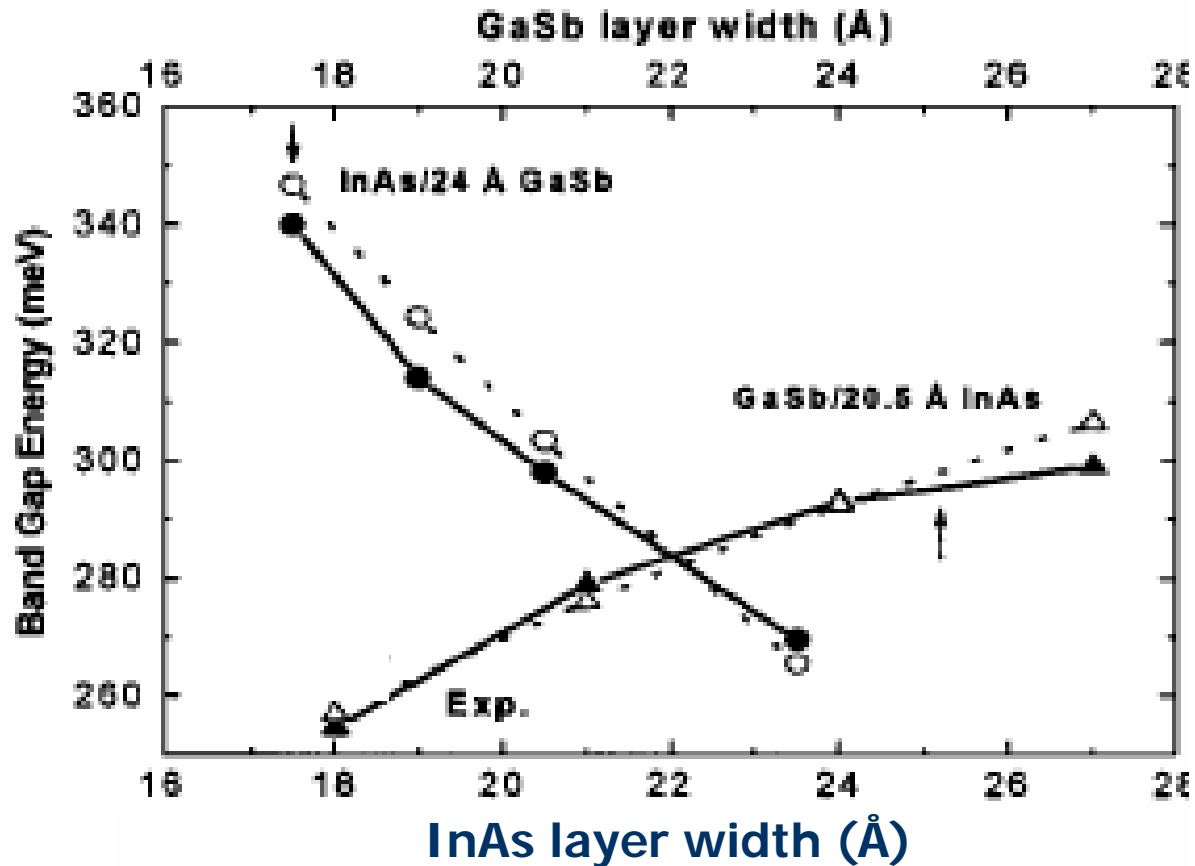
Band-gap tunability 0.2eV to 1.3 eV for IR applications

– but difficult to control growth

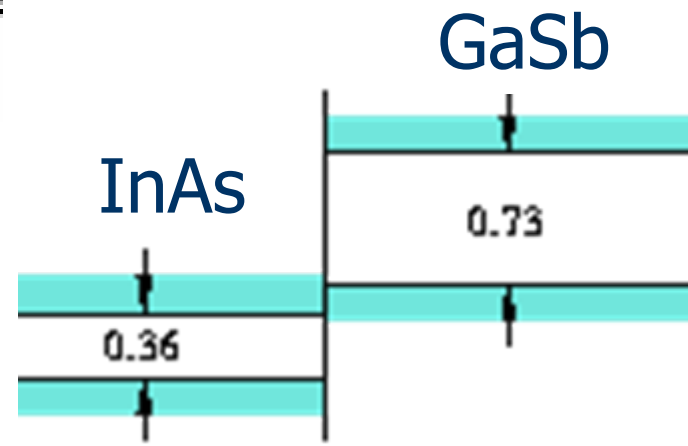


Nosho, Barvosa-Carter, Yang, Bennett, Whitman  
Surf. Sci. 465, 361 (2000).

# Band-gap tuning



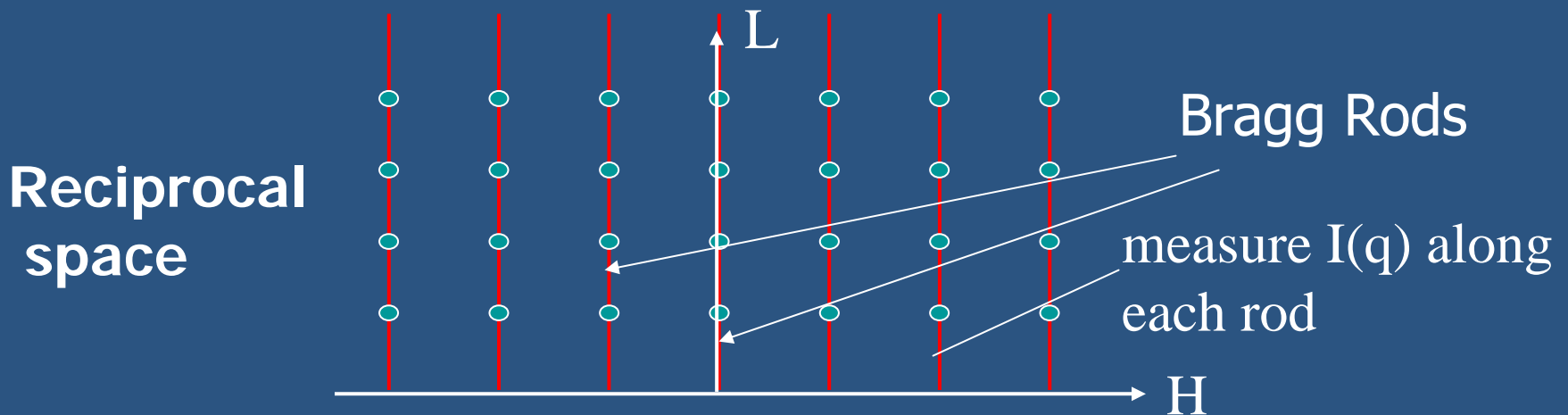
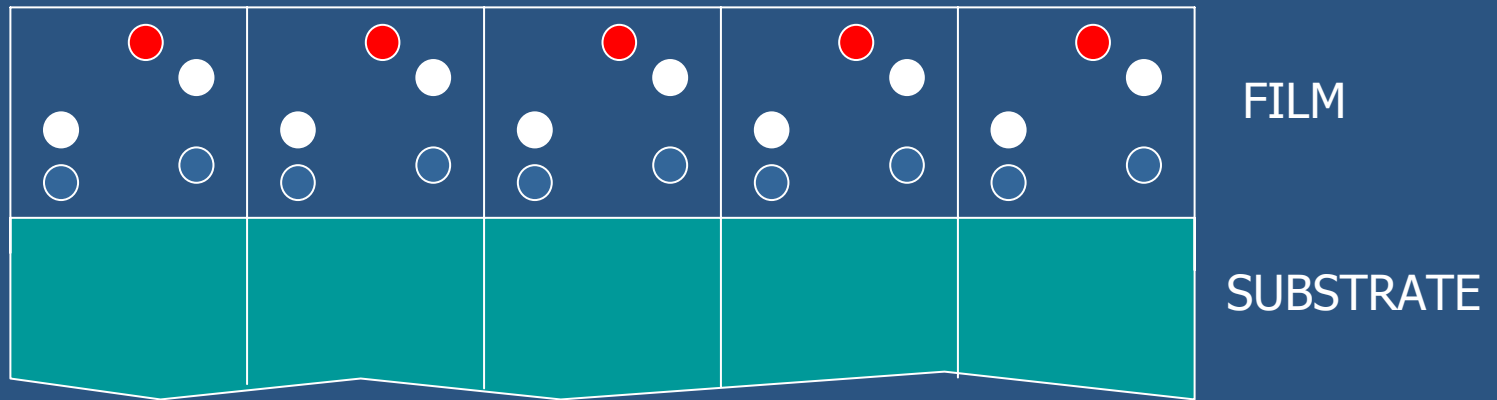
Type II band alignment



Haugen et al. J. Appl. Phys. 96 (2004)

# Epitaxy and 2D periodicity

2D periodic structure coherent with the substrate  
(aka "epitaxial film")

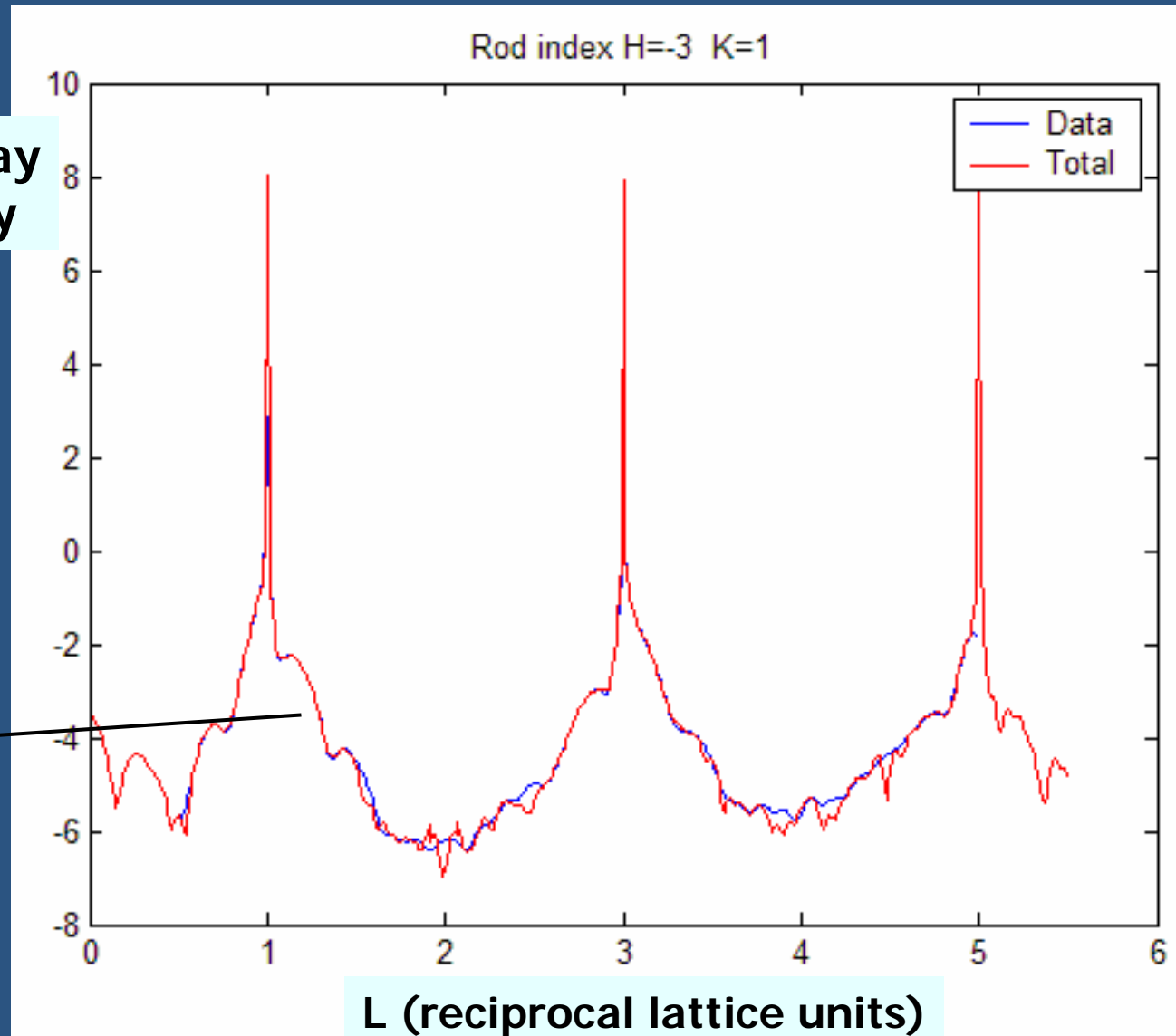


If film and substrate are coherent then get interference fringes along the rods:

**Coherent Bragg Rod Analysis (COBRA)**

# -31L Bragg Rod Profile for sample 811 (InAs on GaSb)

Log x-ray Intensity



(APS, undulator beam line ~ 2hrs/rod need ~ 10 rods)

- large dynamic range: 14 orders!

total

reference  
(substrate)

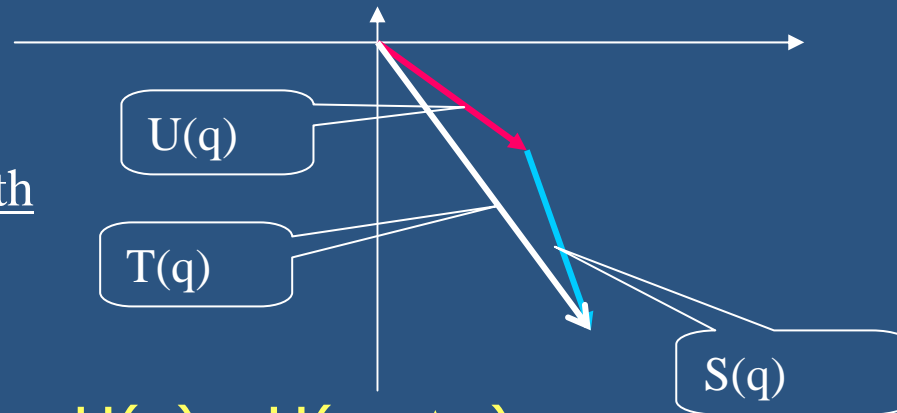
Unknown (film)

$$\text{Scattering factor } T(\mathbf{q}) = S(\mathbf{q}) + U(\mathbf{q})$$

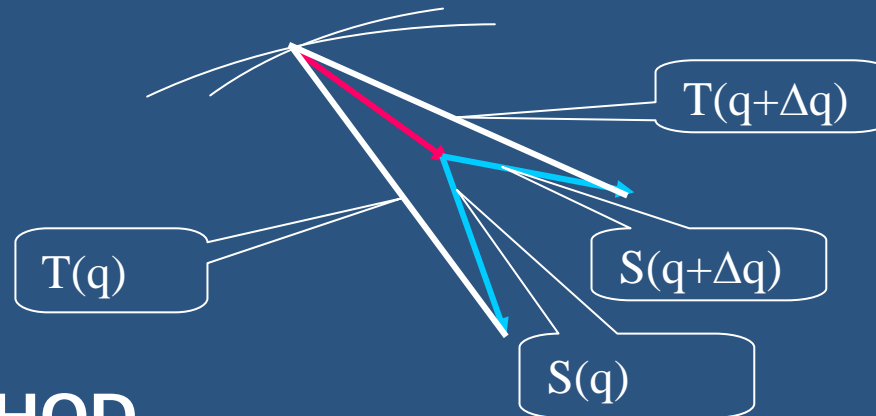
Fourier transform of  $T(\mathbf{q})$   
 $\Rightarrow$  electron density

In the complex plane

know only length  
of  $T(\mathbf{q})$   
from intensity



Approximation:  $U(\mathbf{q}) = U(\mathbf{q} + \Delta\mathbf{q})$

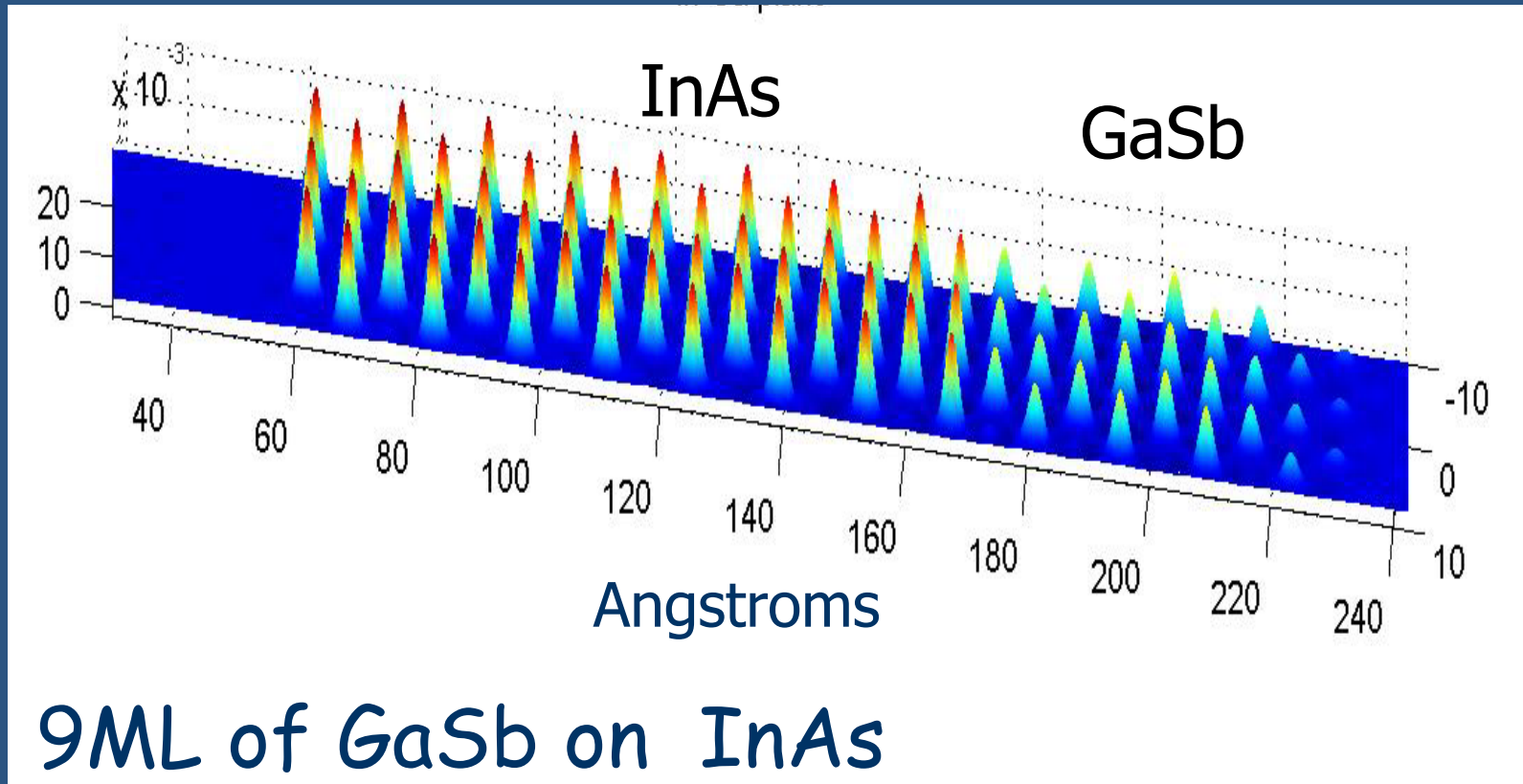


## COBRA METHOD

Sowwan et al. *Phys. Rev. B* 66, 205311 (2002).



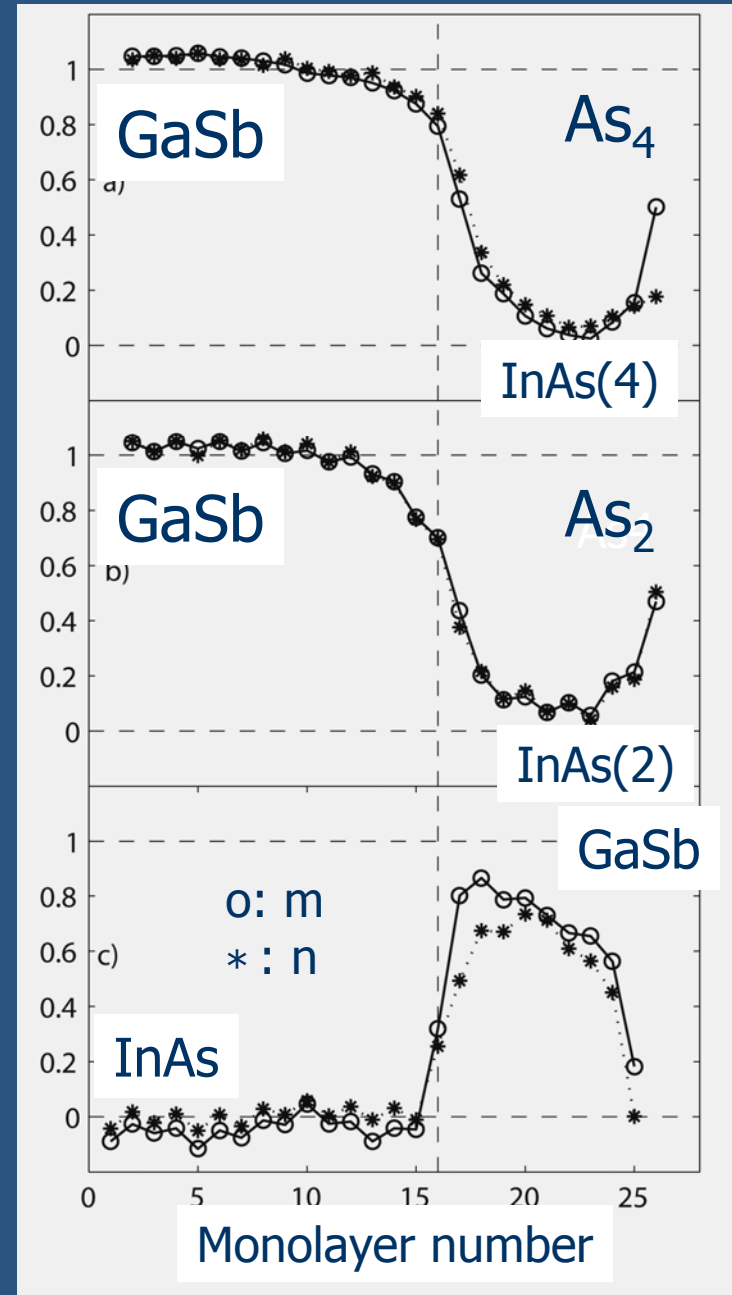
# COBRA MAP of Group III (In,Ga) plane



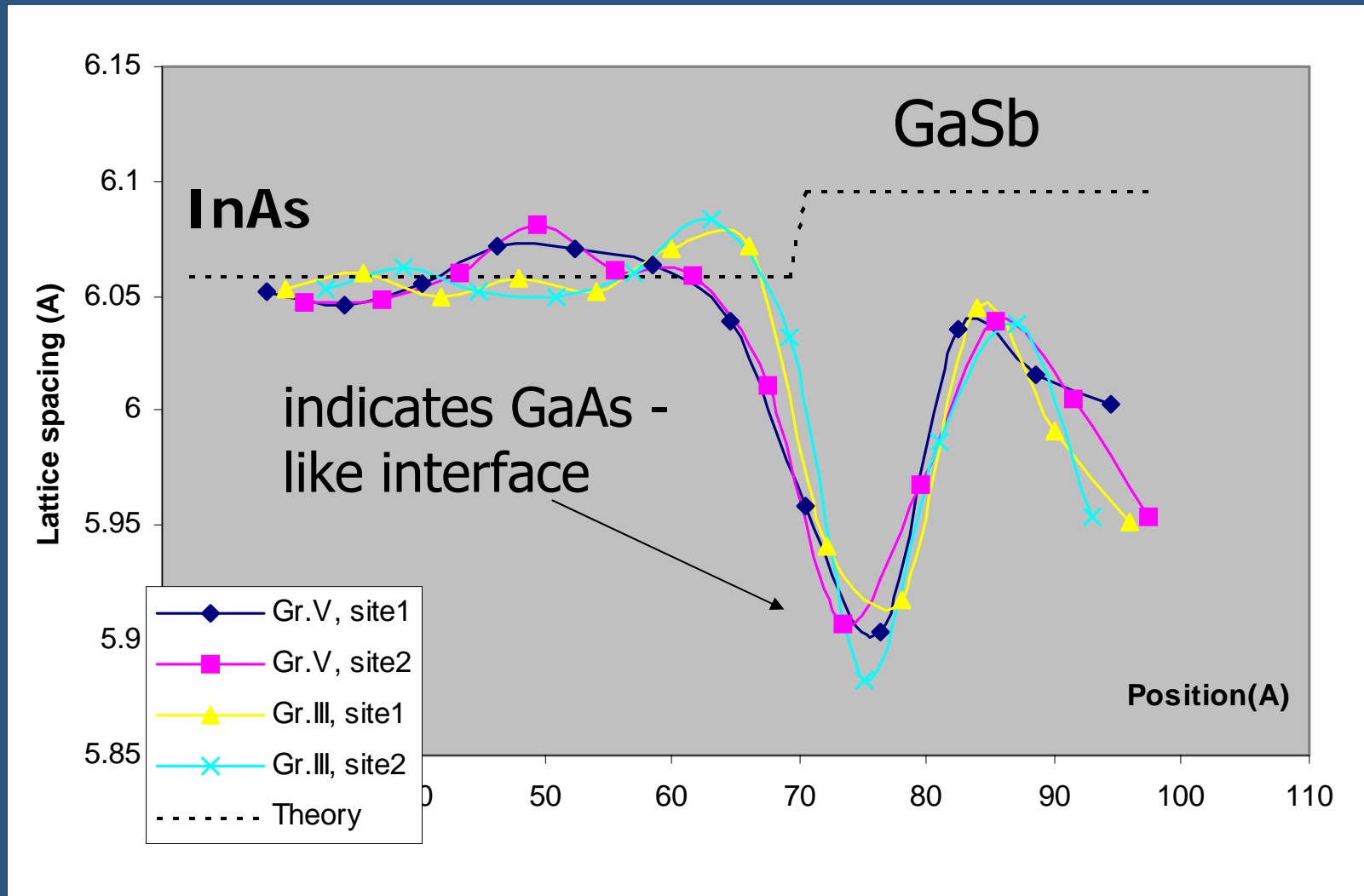
- very coherent interface
- surface roughness ~ 3ML

# Composition profiles determined from COBRA electron density maps

Assumes quaternary system:



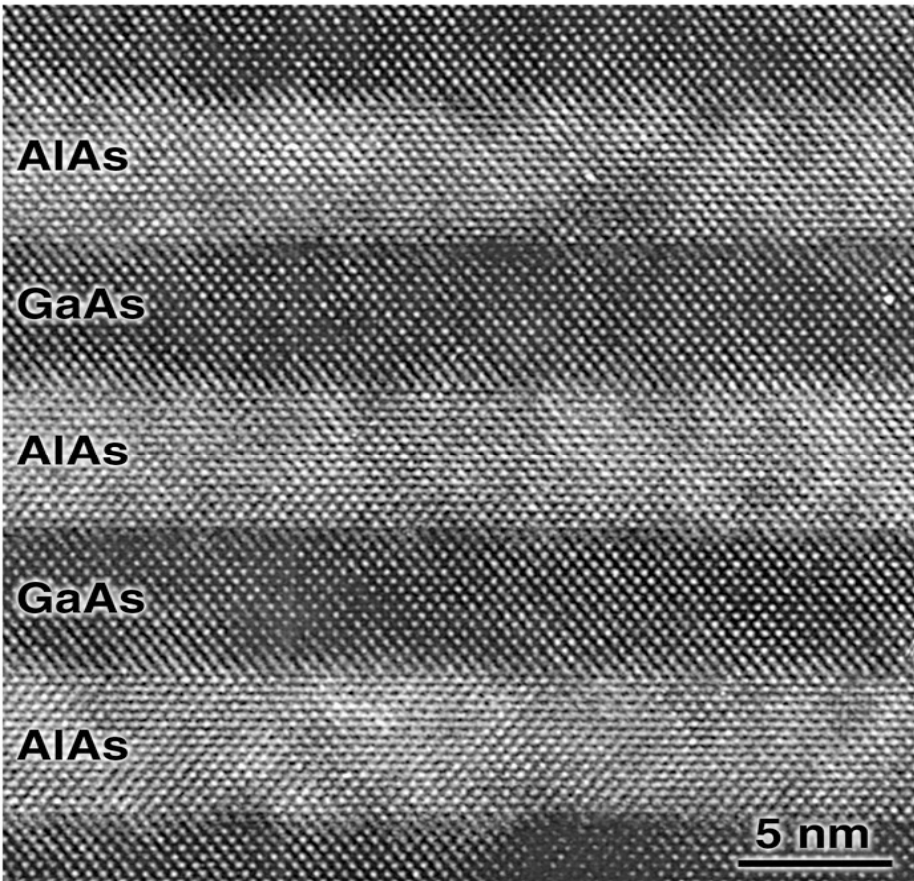
# GaSb on InAs lattice spacing



(Determined from Gaussian fit to electron density peaks)

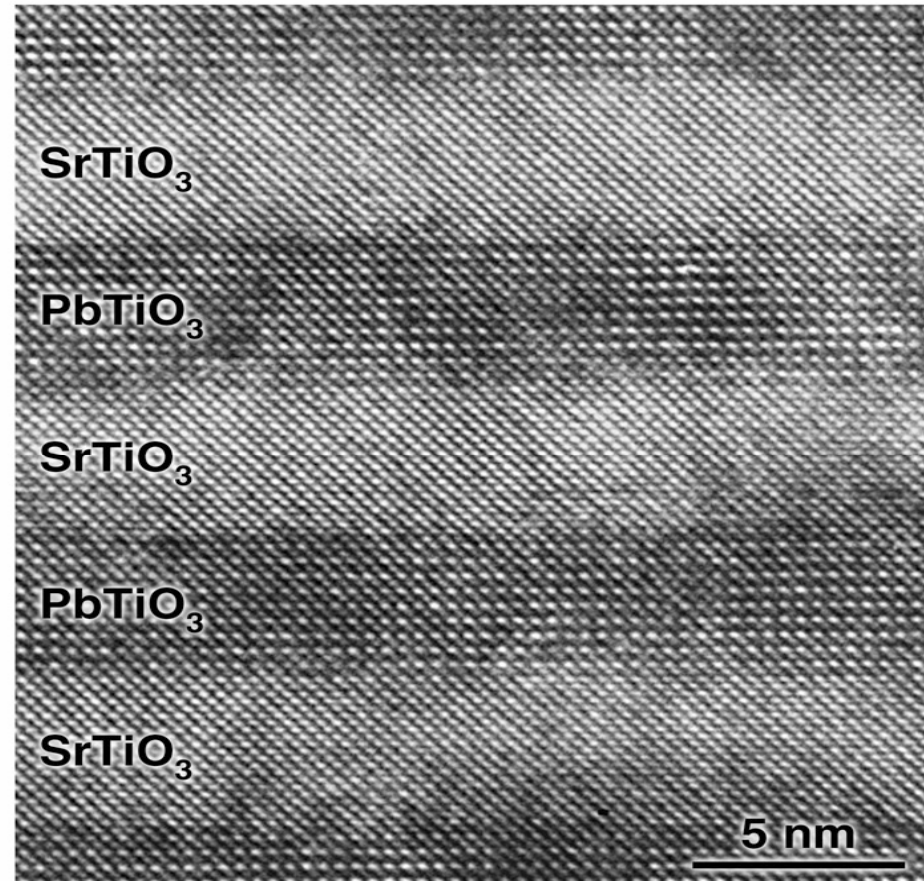
# Ferroelectric nanostructure

## AlAs / GaAs Superlattice



A. K. Gutakovskii *et al.*,  
Phys. Stat. Sol. (a) **150** (1995) 127.

## PbTiO<sub>3</sub> / SrTiO<sub>3</sub> Superlattice

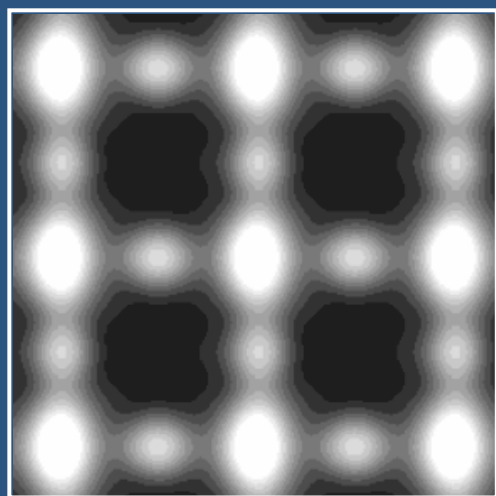


Film – Schlom Group (Penn State)  
HRTEM – Pan Group (Univ. Michigan)

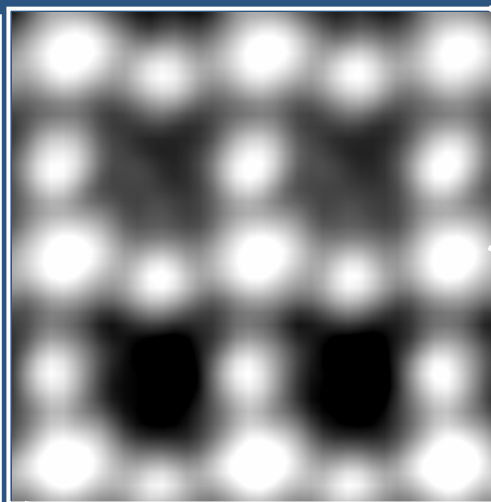
Jiang *et al.*, APL 74, 2851 (1999).



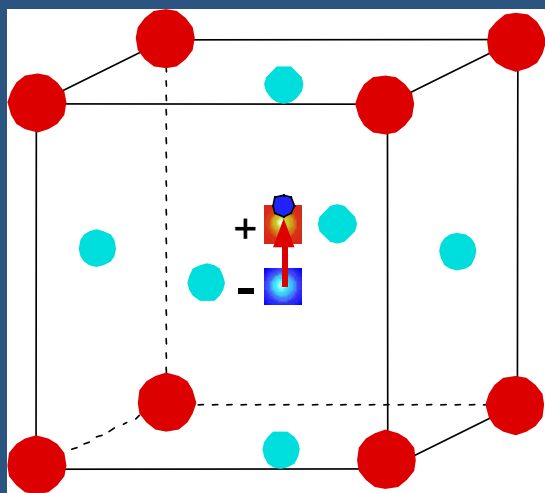
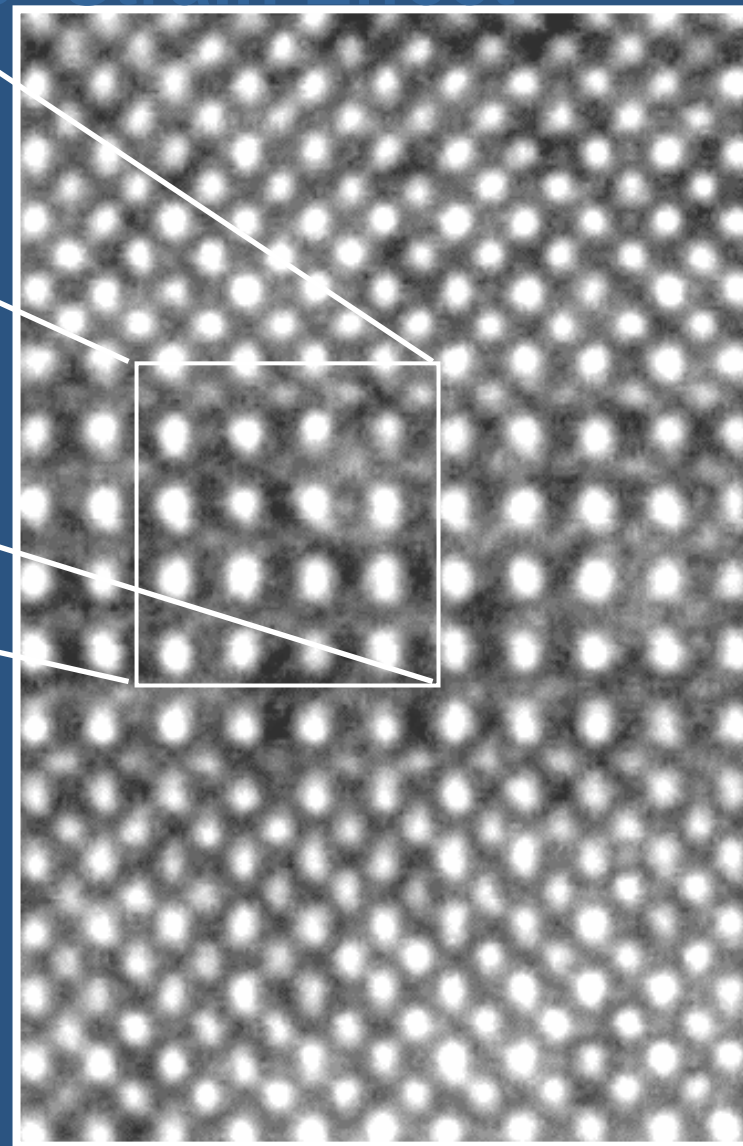
# Ferroelectrics: Superlattice- Strain Effect



bulk BaTiO<sub>3</sub>

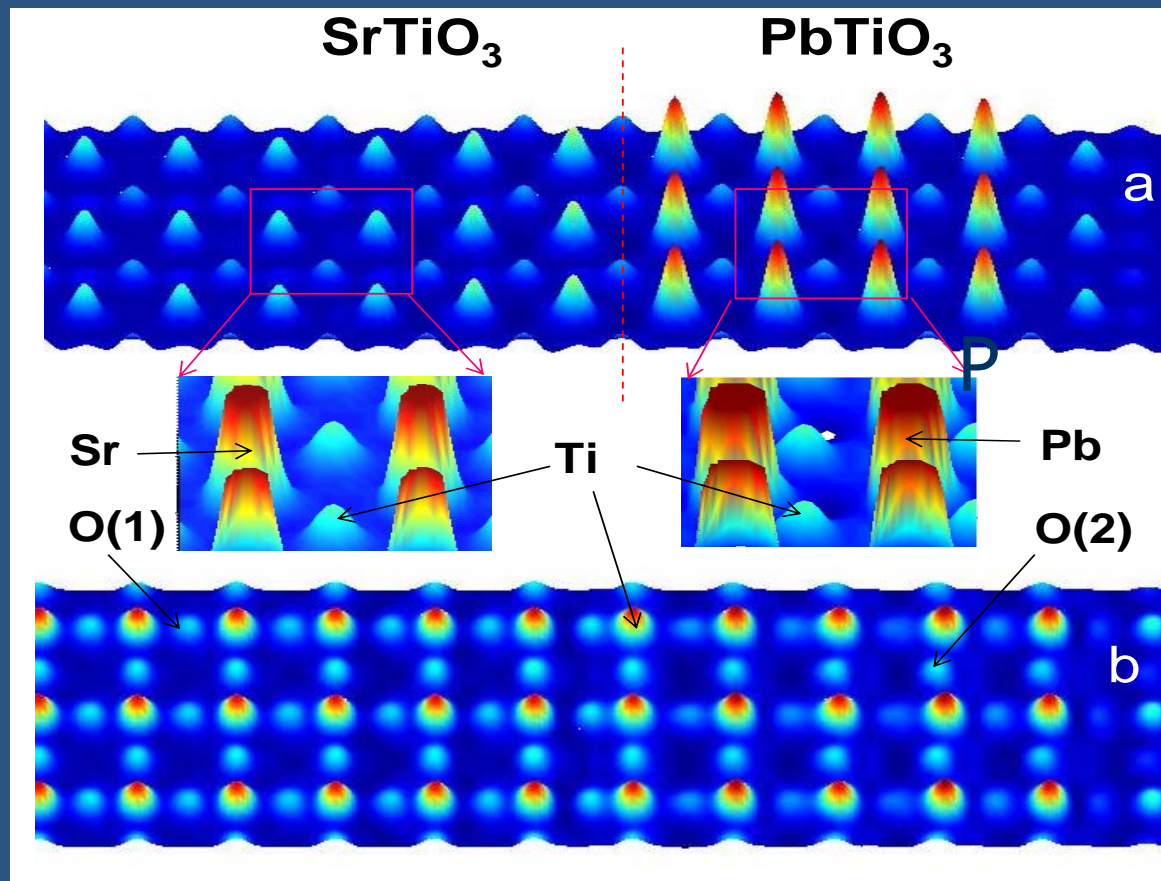


strained BaTiO<sub>3</sub>

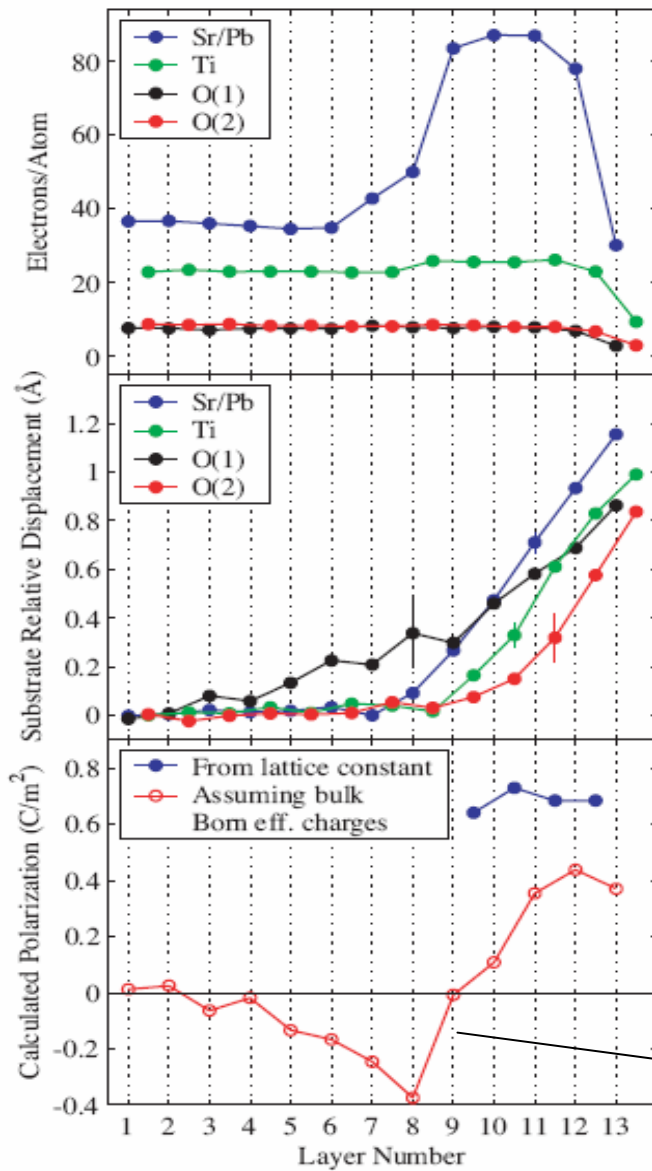


HRTEM image  
[(BaTiO<sub>3</sub>)<sub>6</sub>/(SrTiO<sub>3</sub>)<sub>5</sub>]<sub>20</sub>

# COBRA MAP OF FERROELECTRIC INTERFACE



Sample made by MOCVD, Stephenson group –in-situ facility, APS



# COBRA results on ultrathin film of $\text{PbTiO}_3$

Fong, Cionca, Yacoby, Stephenson, Eastman, Fuoss, Streiffer, Thompson, Clarke, Pindak, Stern, PRB 2005

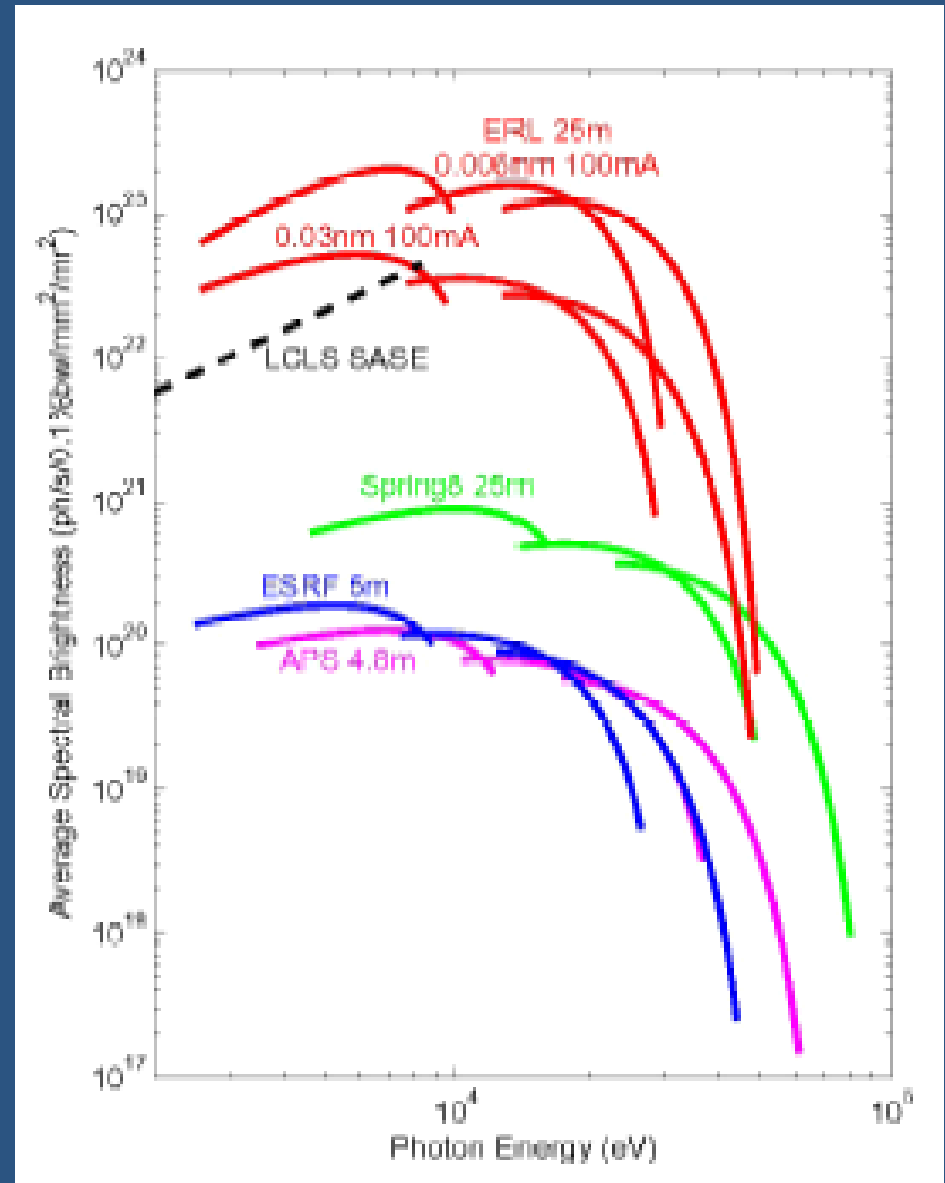
Interface polarization reversal ?

Towards the EFL...

Smaller and faster

→ nm : fs

High rep-rate  
and tunability are  
great features!!





# ERL characteristics for in-situ materials dynamics studies

Modes:	Short-Term Goals			Long-Term Goals		Unit
	(A) Flux	(B) High-Coherence	(C) Short-Pulse	(D) Ultra High-Coherence	(E) Ultra Short-Pulse	
Energy	5	5	5	5	5	GeV
Macropulse current	100	25	1	100	1	mA
Bunch charge	77	19	1000	77	10000	pC
Repetition rate	1300	1300	1	1300	0.1	MHz
Transverse emittance (norm. rms)	0.3	0.08	5.0	0.06	5.0	mm.mrad
Transverse emittance (geometric at 5GeV)	31	8.2	511	6.1	511	pm
Bunch length (rms)	2000	2000	50	2000	20	fsec
intra-bunch Energy spread (fractional; rms)	2E-4	2E-4	3E-3	2E-4	3E-3	
Beam power	500	125	5	500	5	MW
Beam loss	< 1	< 1	< 1	< 1	< 1	μA

kW average power (fiber) lasers on the horizon (mJ/pulse at 1MHz)

USING EVERY X-RAY PHOTON

# Ultrafast Laser Deposition

## Plasma Plume Imaging & Spectroscopy

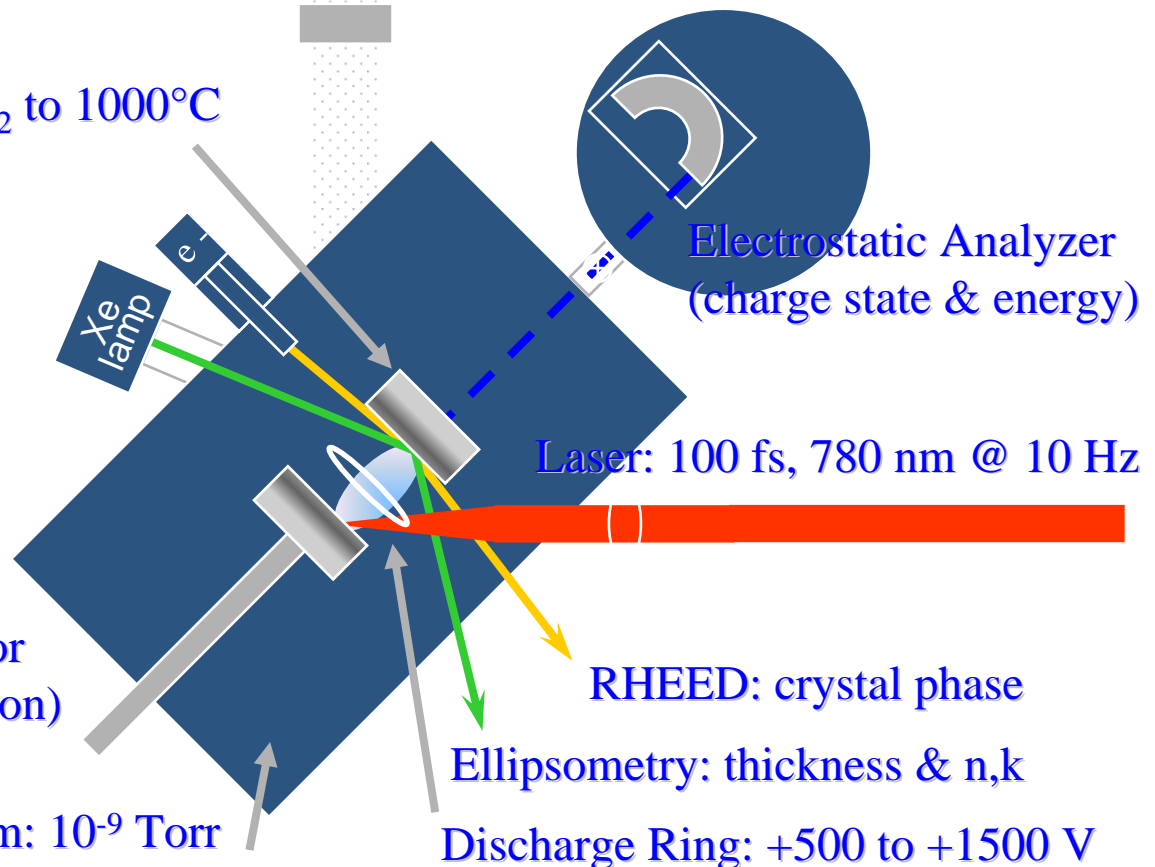
2" Substrate Holder:

- temperature: liquid N<sub>2</sub> to 1000°C
- rotation & translation

Load-Lock for  
Substrate Transfer

Multi-Target Selector  
(4 targets with rotation)

Vacuum: 10<sup>-9</sup> Torr  
N<sub>2</sub> backfill: 10<sup>-4</sup> Torr



Electrostatic Analyzer  
(charge state & energy)

Laser: 100 fs, 780 nm @ 10 Hz

RHEED: crystal phase

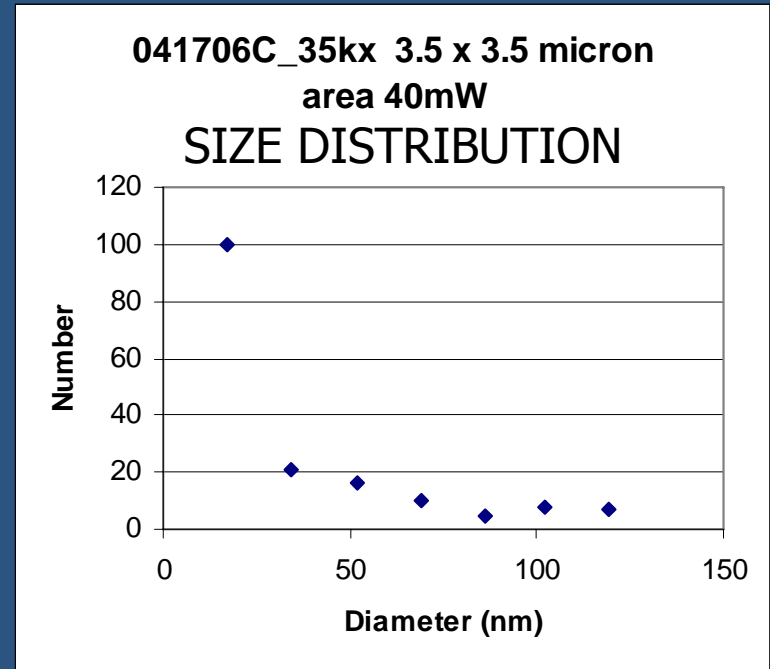
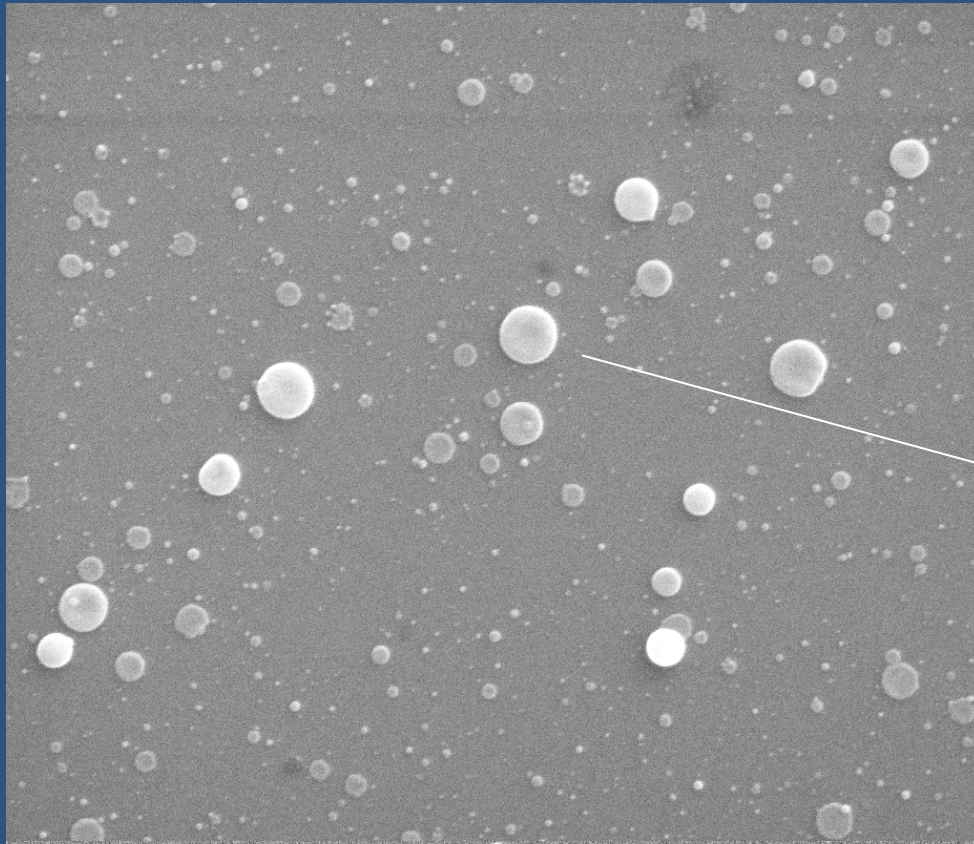
Ellipsometry: thickness & n,k

Discharge Ring: +500 to +1500 V

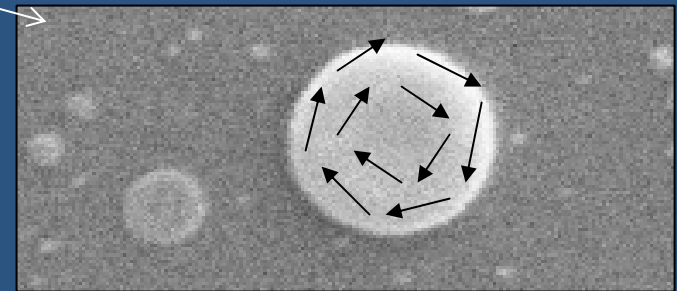
# Co nano-disks on Si

ultrafast laser deposition

810nm, 50 fs, 1mJ/pulse 1 kHz



200nm

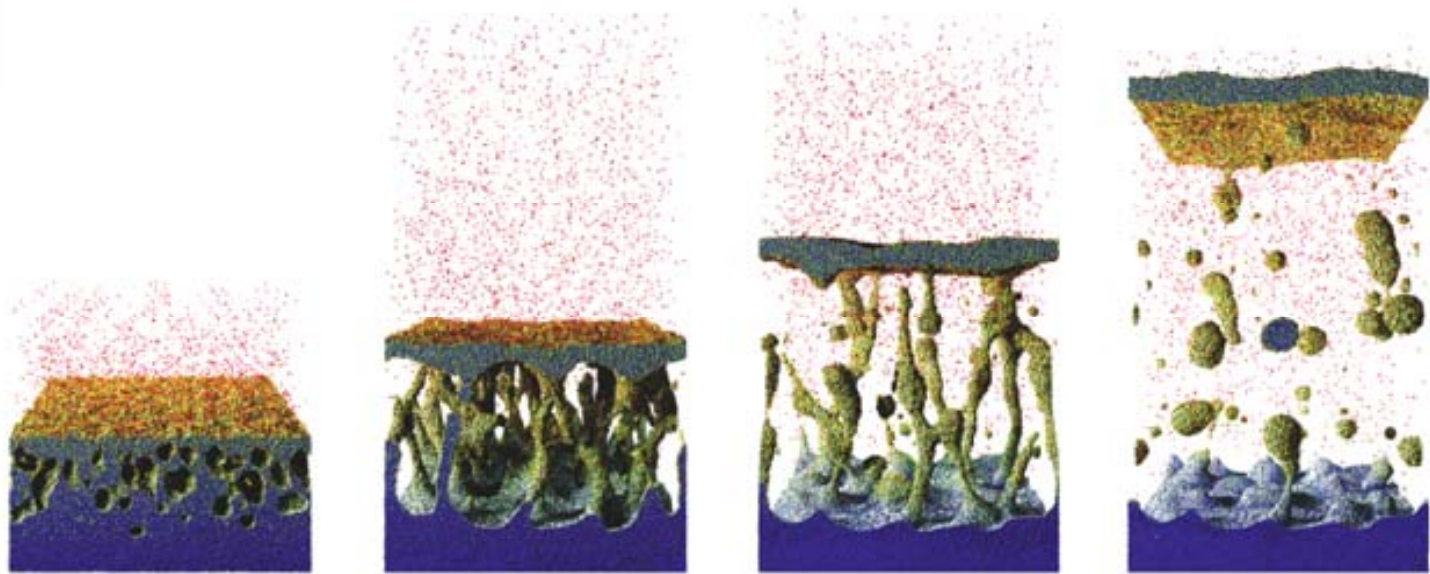


spin vortex state

35kx

	HV	mag	tilt	WD	curr	dwell	4/25/2006	← 500 nm →
	20.00 kV	35000 x	-0 °	4.3 mm	1.7 nA	30 μs	12:14:54 PM	

# Probing the initial stages of ultrafast ablation



**Snapshots of MDCASK simulation  
of the ablation of Cu (From Gilmer et al.,  
unpublished research)**

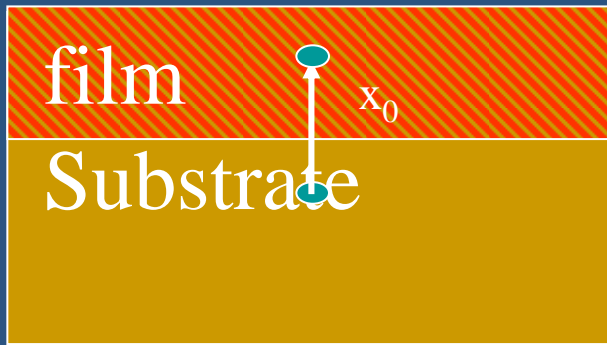
Timescale of ERL matches simulation  
Laser pump – x-ray (diffraction/XAFS) at high rep rates

# Detectors !

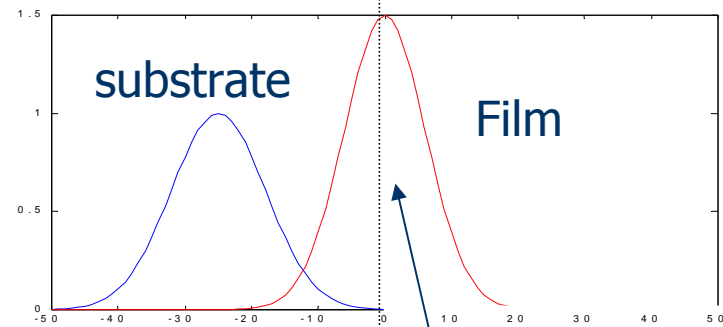
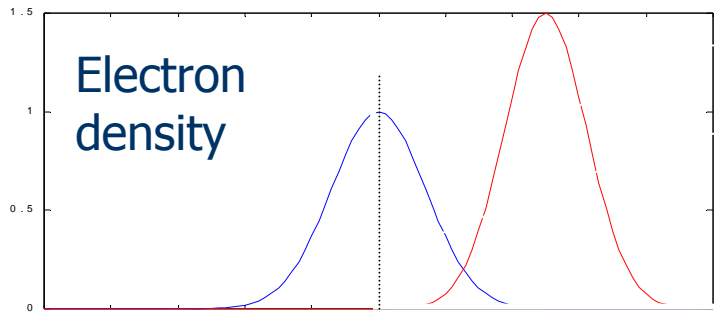
- Investment lags source development
- Need fast, gated area detectors –badly!
  - efficient direct detection PADs
  - x2 from cross-scan, more from // channels
  - $\leq n$  sec gating
  - $10^5$  dynamic range
  - multiple read architecture
  - modular
  - .....

# Summary

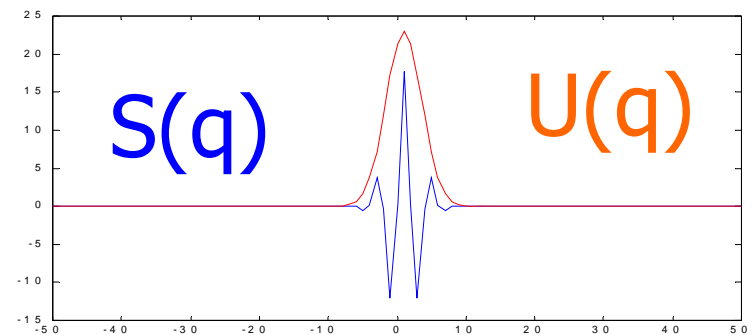
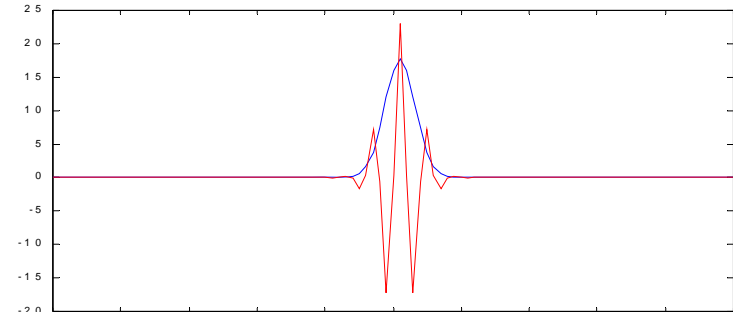
- ERL source could enable dynamic imaging of atomic/spin motions associated with collective phenomena  
(e.g. fast switching with laser impulse)
- deposition processes also, esp. for nanostructures
- combination of fast rep-rate and short pulse matches well to laser pump development  
(high average power fiber lasers)



## Real space



## Reciprocal space

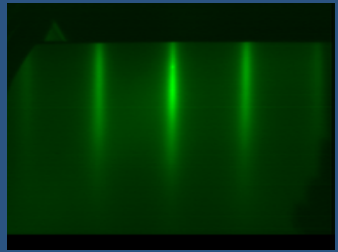


This choice of origin satisfies the COBRA approximation: slowly varying  $U(q)$

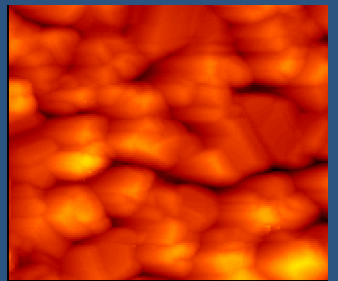


# MBE Growth and In-situ Analysis

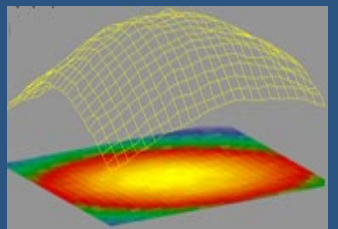
Integrated  
MBE/RHEED/STM/MOS  
capabilities:



RHEED: Real-time surface structure characterization (annealing)



STM: surface morphology, roughness, coarsening



MOS: curvature mapping gives mismatch strain information.





# Typical Growth Conditions

GaSb on InAs  $T_{\text{sub}} = 450\text{C}$

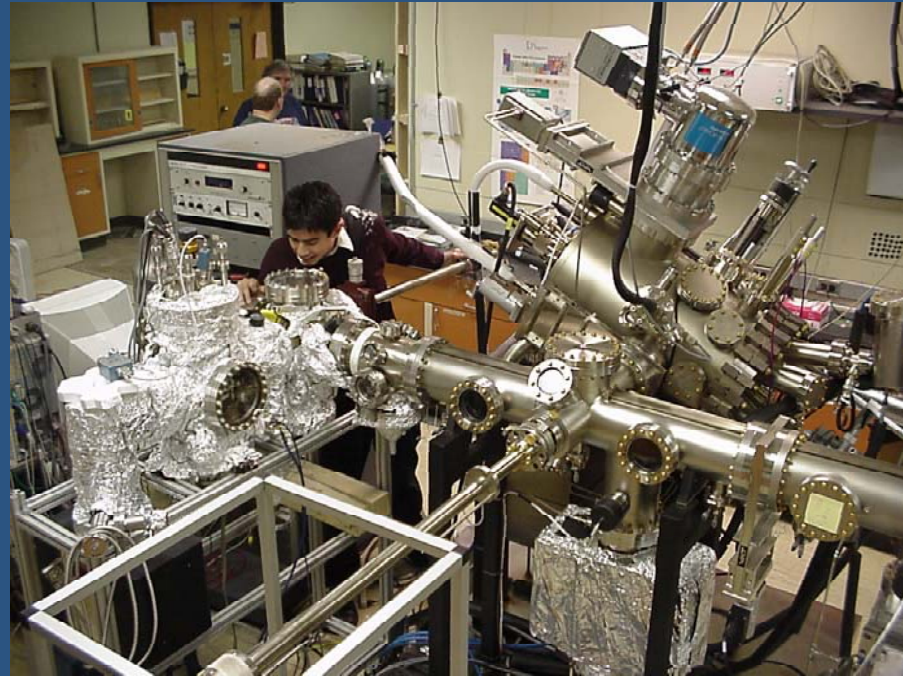
GaSb 9ML MBE (30sec)

Ga/Sb 1ML/1ML MEE (3s/2s)

Sb 1ML MEE (2s)

In 1ML MEE (2s)

InAs 0.2 micron



*Combined MBE growth chamber and STM chamber. Mirecki Millunchick Lab, University of Michigan*

In-situ: k-Space Associates KSA 400 and MOSS

# Sample 814 GaSb on InAs

In = 49  
Ga = 31

Exp ratio  $\sim 1.6$   
Ideal = 1.6

Accumulation of  
As at interface

As = 33  
Sb = 51

Exp ratio  $\sim 1$   
Ideal = 0.65

