

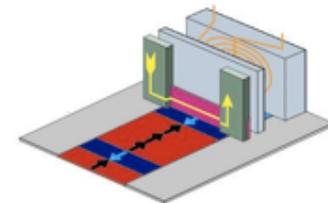
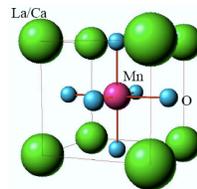
# Resonant Coherent X-ray Imaging

Ian McNulty

*Advanced Photon Source*

Workshop on Diffraction Microscopy, Holography and  
Ptychography using Coherent Beams

Cornell University, 7 June 2011

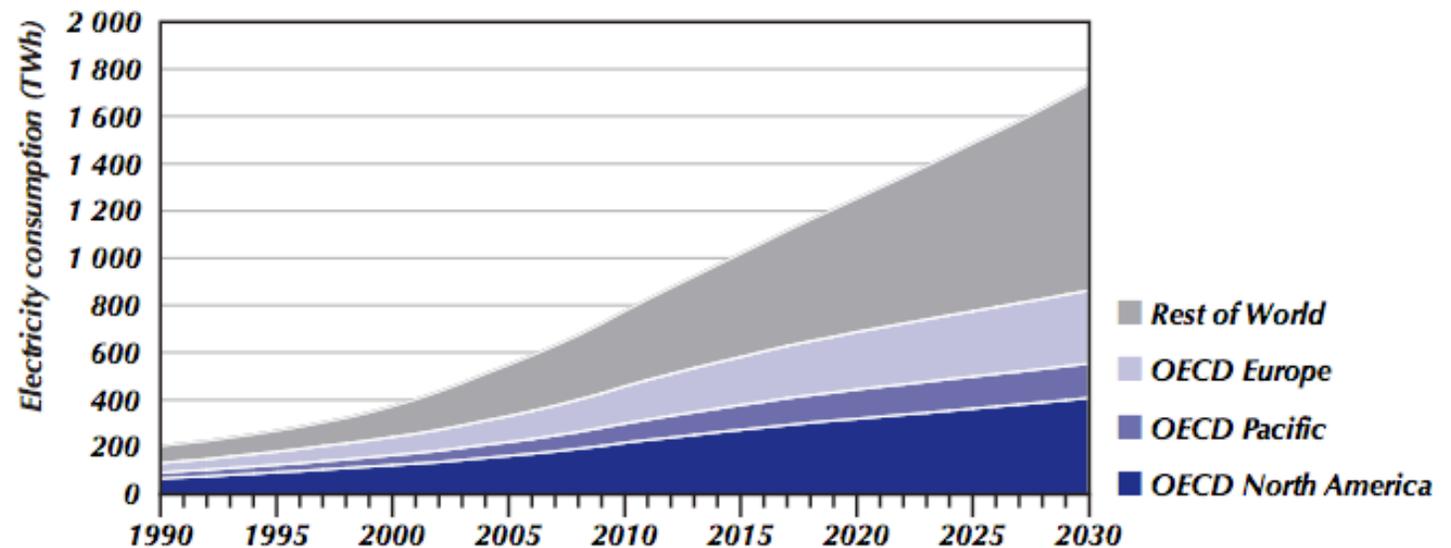


# Gadgets and Gigawatts

By 2010 there will be over 3.5 billion mobile phones subscribers, 2 billion TVs in use around the world and 1 billion personal computers. Electronic devices are a growing part of our lives and many of us can count between 20 and 30 separate items in our homes, from major items like televisions to a host of small gadgets. The communication and entertainment benefits these bring are not only going to people in wealthier nations – in Africa, for example, one in nine people now has a mobile phone. But as these electronic devices gain popularity, they account for a growing portion of household energy consumption.

**Figure 3 • Estimated electricity consumption by ICT and CE equipment in the residential sector, by region, 1990-2030**

Communication technologies (ICT) and consumer electronics (CE) now account for approximately 15% of global residential electricity consumption



Source: IEA estimates.



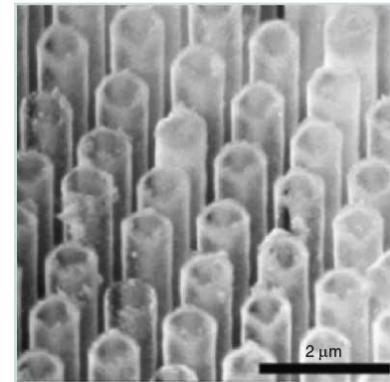
# Imagine ...

- Spin-based non-volatile memory devices and transistors that take almost no power -> "Instant-on" computers



- Ultrafast devices in which a spin-polarized supercurrent propagates over long distances -> sat-phone-in-a-watch

- Room-temperature magnetic field sensors with the sensitivity of a SQUID; efficient, miniature cooling devices and high-power microwave, electron, and x-ray sources



Ferroelectric  $\text{SrBi}_2\text{Ta}_2\text{O}_9$  nanotubes

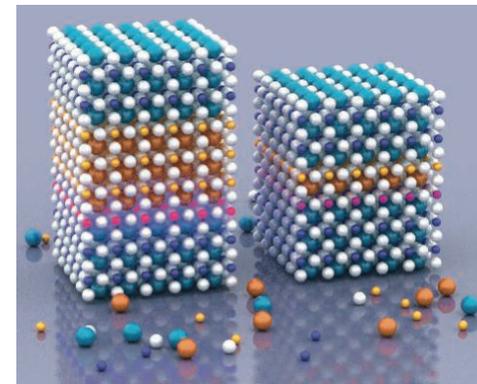
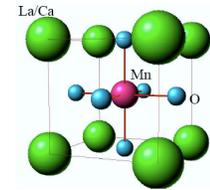
C.A.F. Vaz, Adv. Mater. 20, 1 (2010)

J.F. Scott, Science 315, 954 (2007)



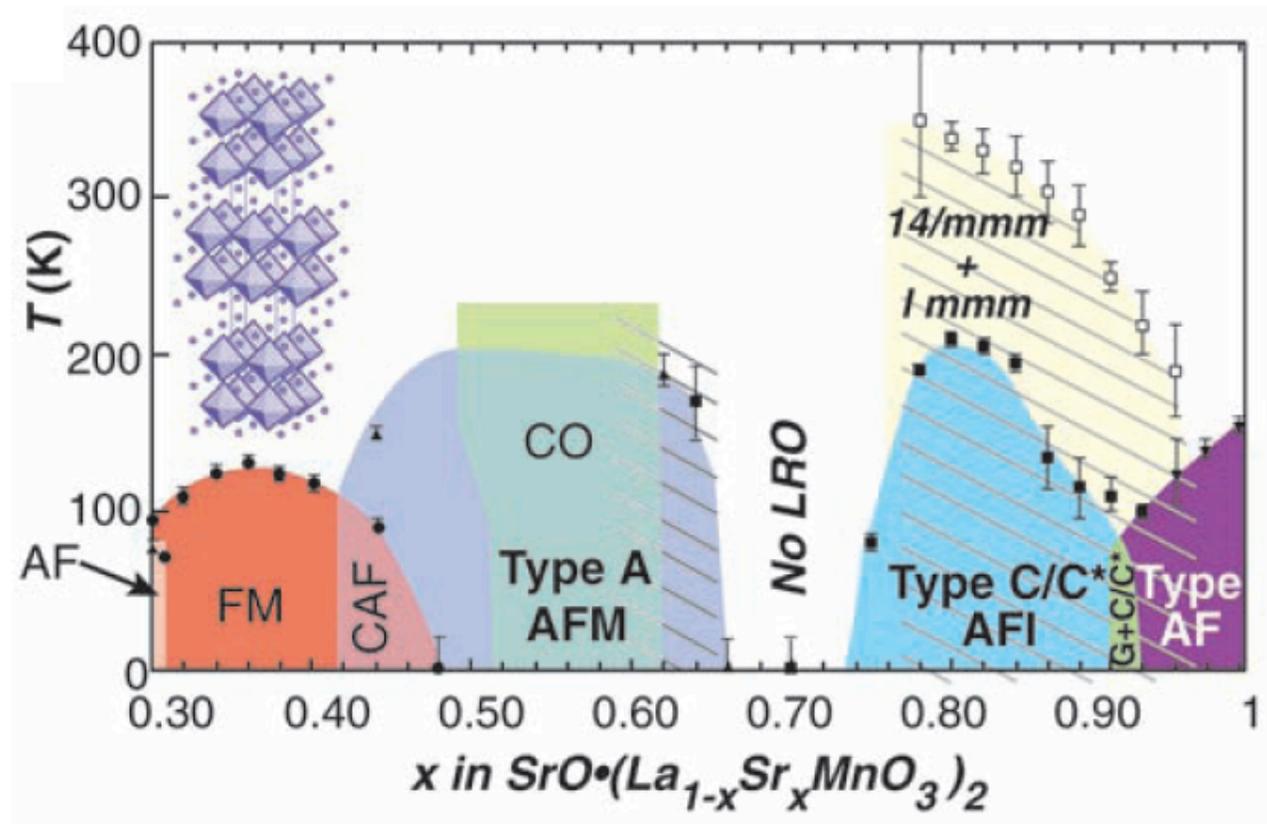
# Challenging science in bulk condensed matter at the sub-10 nm scale

- Understand competing nanoscale phases and ordering with resultant strain in strongly correlated electron materials
- Probe interfaces between different oxides with coupled order parameters, such as in bilayers and heterostructures of magnetoresistive and superconducting materials
- Map domain wall (magnetic, orbital, charge) structure, transport, and fluctuations in magnetic materials and devices with coupled order parameters



# Complex disorder: nanoscale phase separation

Strongly competing ground-state spin, charge, orbital, lattice degrees of freedom give rise to complex phase diagrams → phase separation

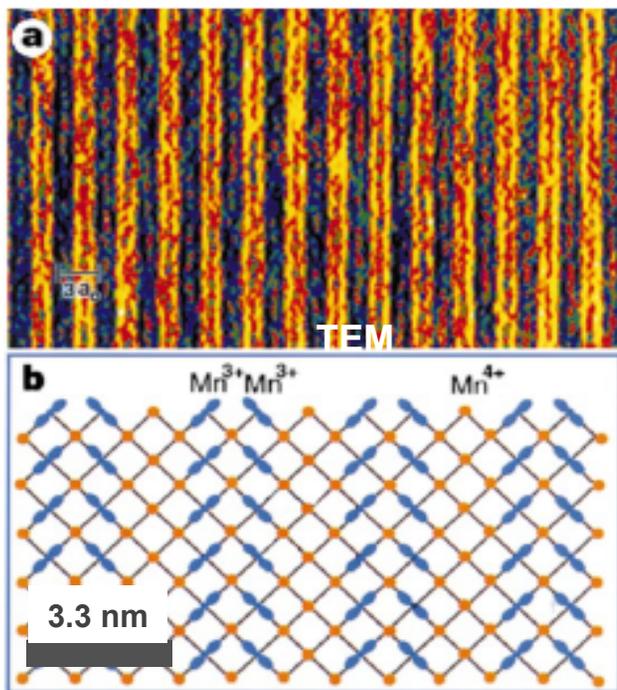


E. Dagotto, Science 309, 262 (2005)



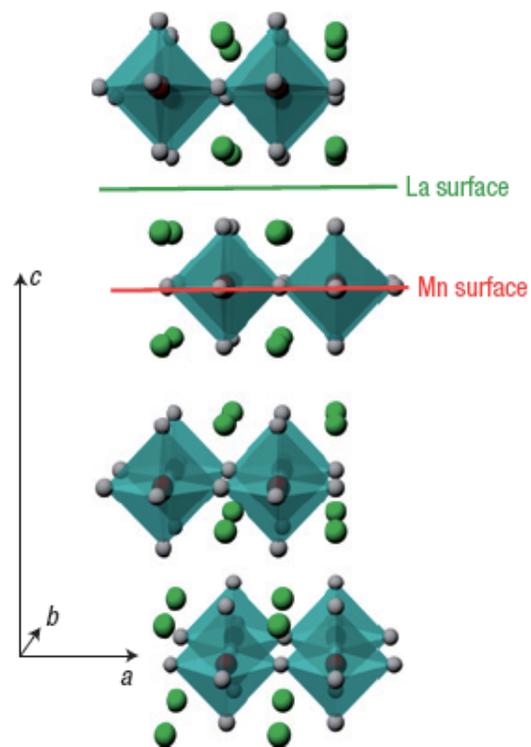
# Charge and orbital ordering have been observed in thin samples and at surfaces

Charge-ordered stripes imaged by TEM. More the exception than the rule that such structures can be seen!



S. Mori, Nature 392, 473 (1998)

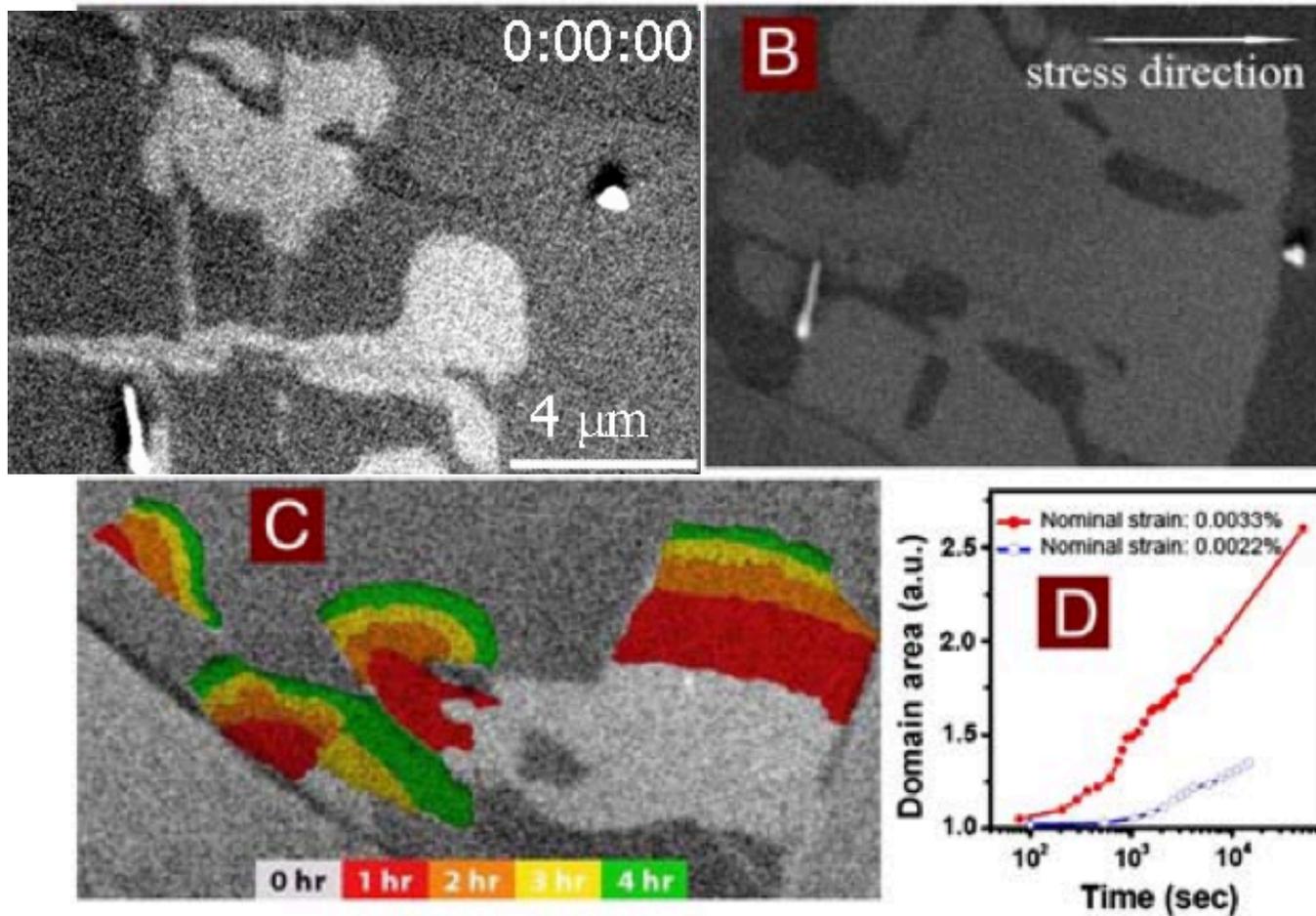
How do surfaces effect orbital ordering compared to the bulk?



Y. Wakabayashi, Nature Materials 6, 972 (2007)



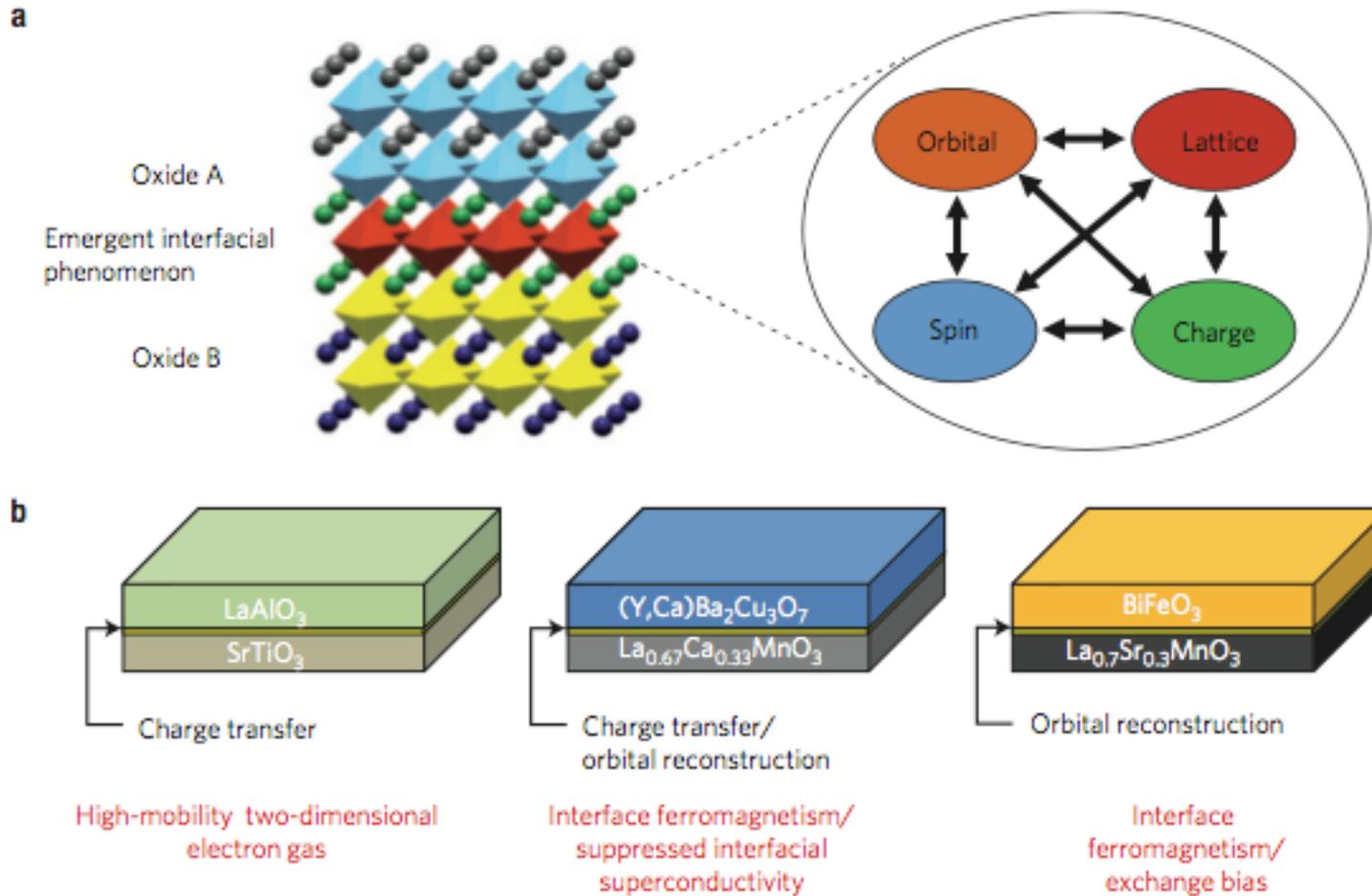
# Surface probes are incredibly powerful ... but many interesting problems are buried in bulk



T.-H. Kim, PNAS 107, 5272, (2010)



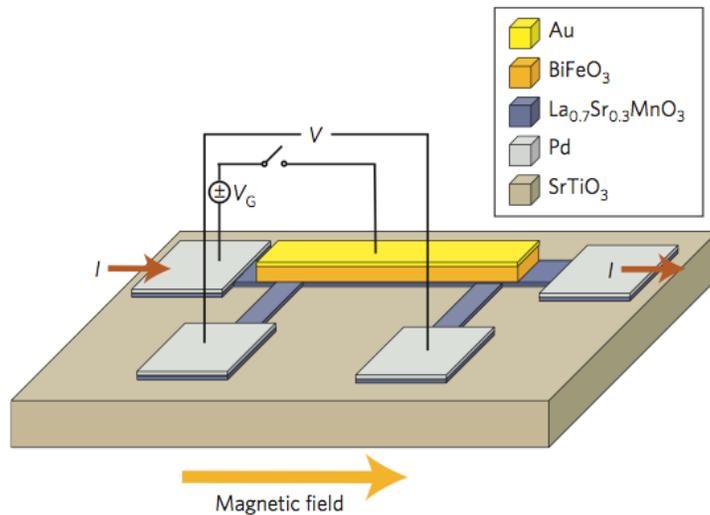
# Combining multiferroics with correlated electron materials enables new, more sensitive devices



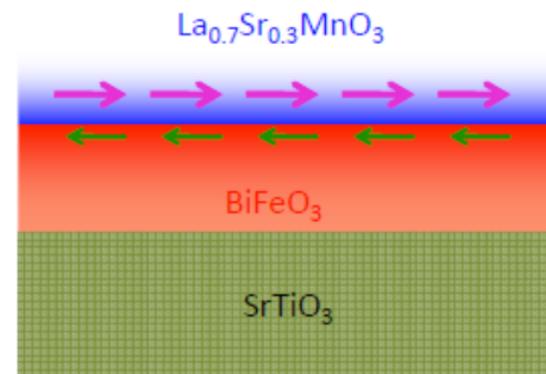
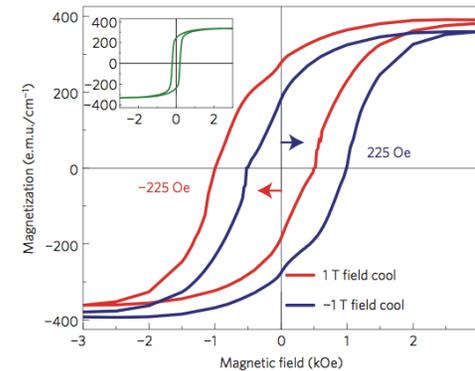
S.M. Wu, Nature Materials 9, 756 (2010)



# How to study spin and strain at domain boundaries, under useful conditions, at the nanoscale?



**Figure 3 | A schematic of the BFO/LSMO field-effect device.** To change the BFO polarization, a voltage pulse  $V_G$  is applied between the gate and the LSMO channel. The magnetic field for MR measurements is applied parallel to the direction of the current.



XMCD spectra at Mn and Fe L-edges strongly suggests that in the first few nm of the BFO film, a new spin structure is present with antiparallel coupling between bulk Mn and interfacial Fe spins. This enhanced magnetism is markedly different from that in the remainder of the BFO film.

S.M. Wu, Nature Materials 9, 756 (2010)



## X-rays offer:

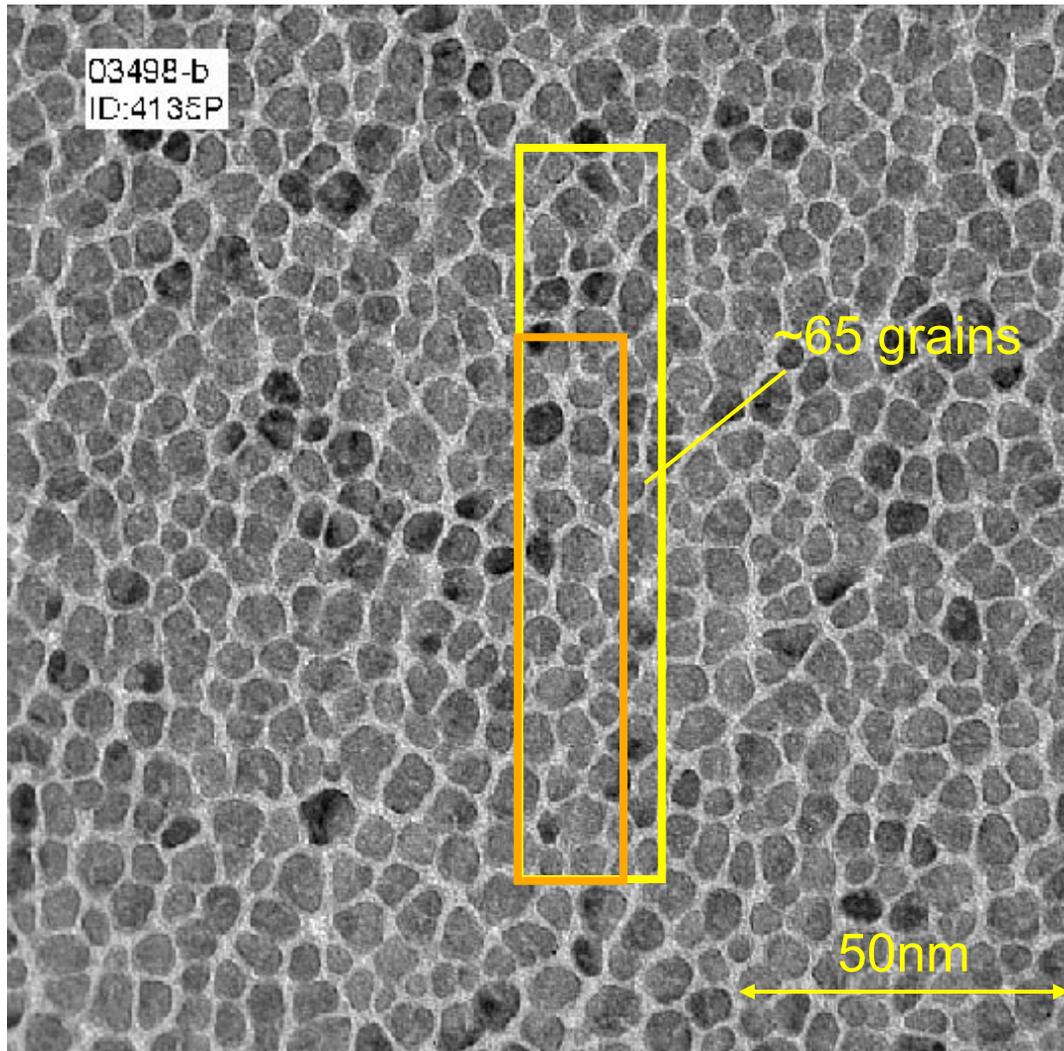
- Short wavelengths, high spatial resolution
- Weakly interacting, high penetration
- Coupling to core-level electrons: “clean” measurement of electronic and chemical states
- Coupling to electronic spin via polarization: probe magnetic and orbital states

## CDI offers:

- Amplitude and phase imaging with strain sensitivity and spatial resolution beyond the limits of x-ray lenses
- Working distance for high and low temperature, pressure, fields



# Beyond today's magnetic storage media



current demonstrations approach 400 Gb/in<sup>2</sup> → 40 grains/bit

## Problem

Why not make one grain per bit (domain limited)?

Too-small grains become thermally unstable

## Opportunity

Denser storage: smaller domains, repeatable switching

- Resolve internal structure during bit reversal, correlate with magnetic behavior

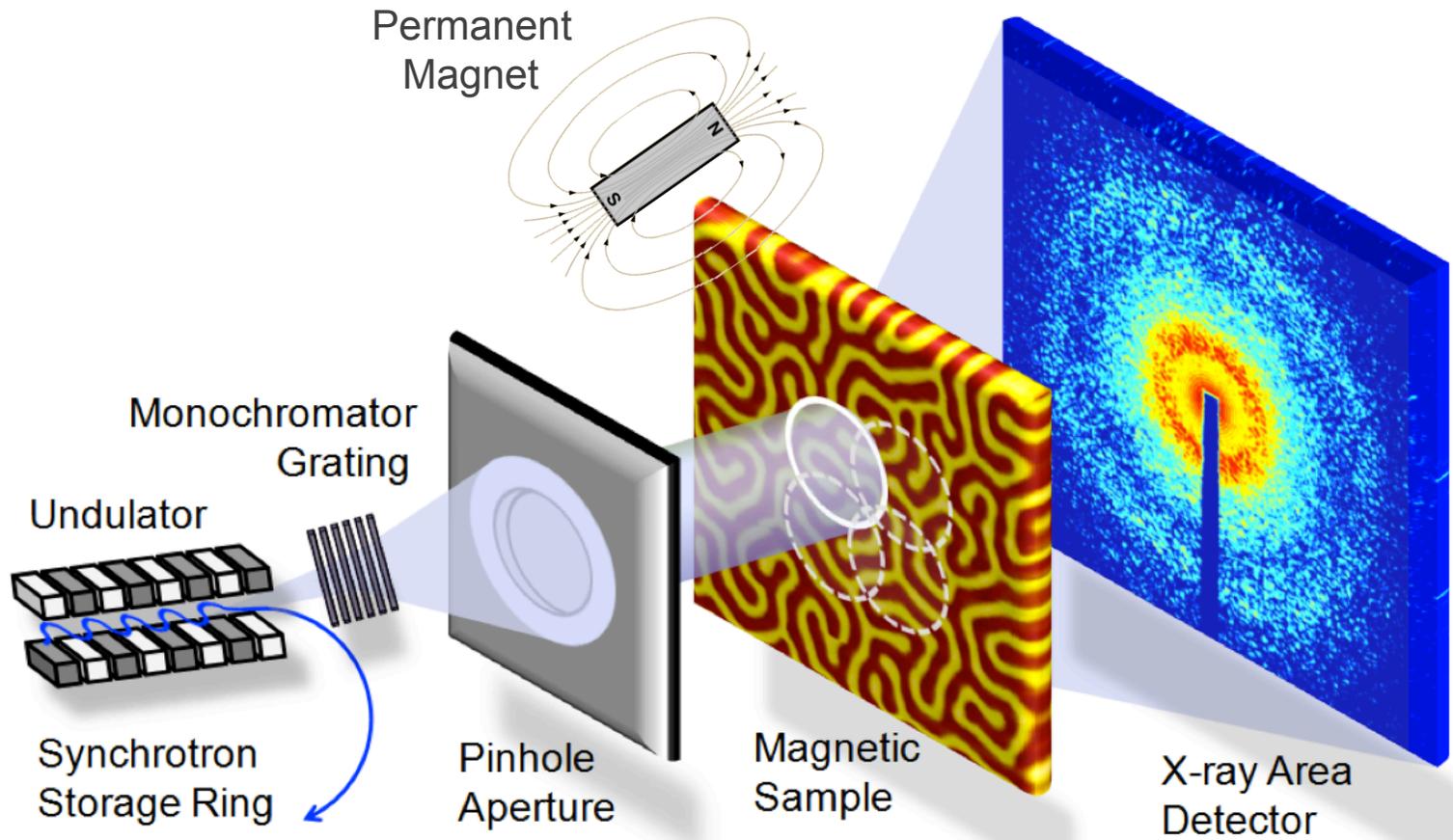
- Reveal functionality of buried layers in complex devices

- What is happening on the bit side-walls and edges?

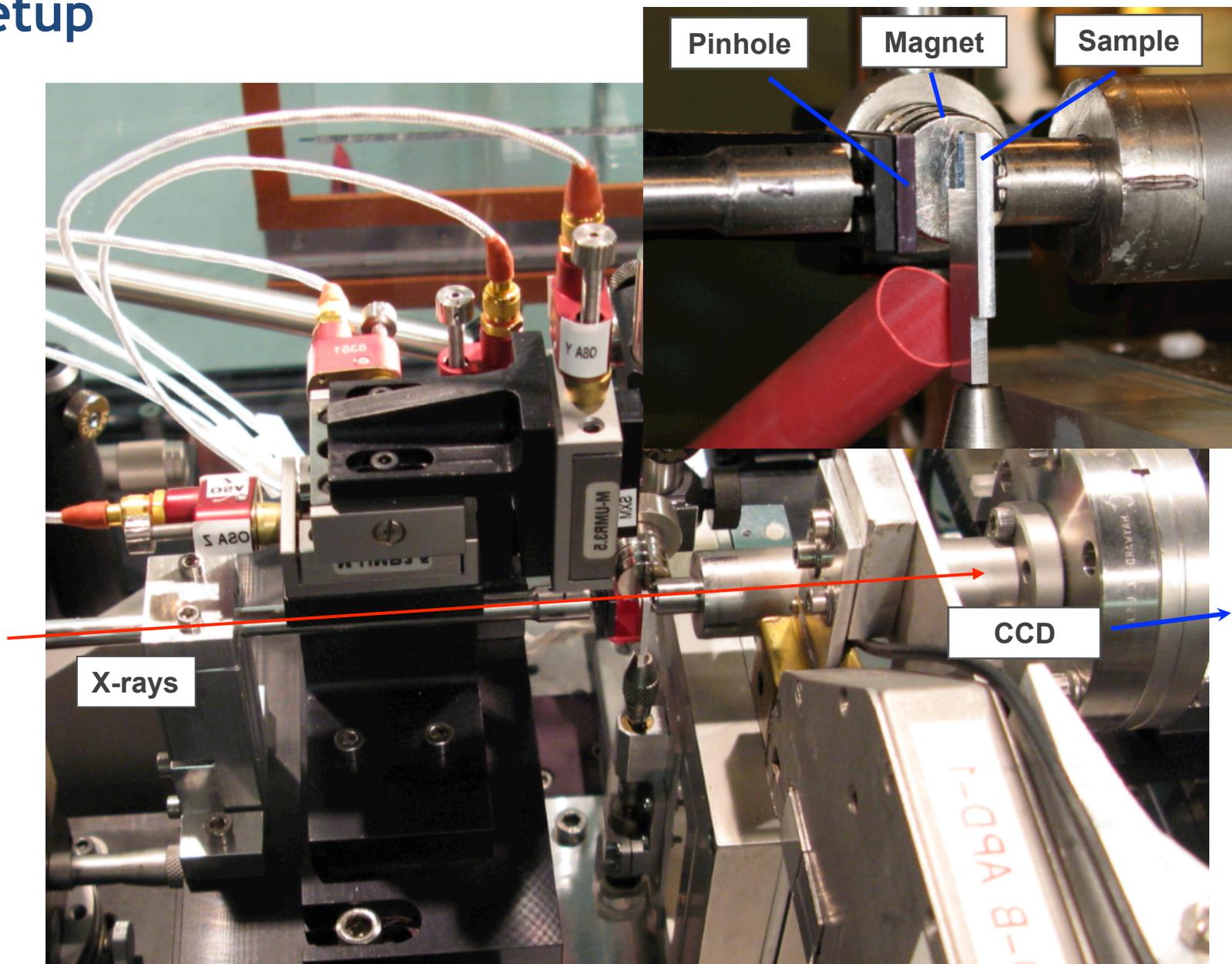
H.J. Richter, MMM (Nov. 2005)



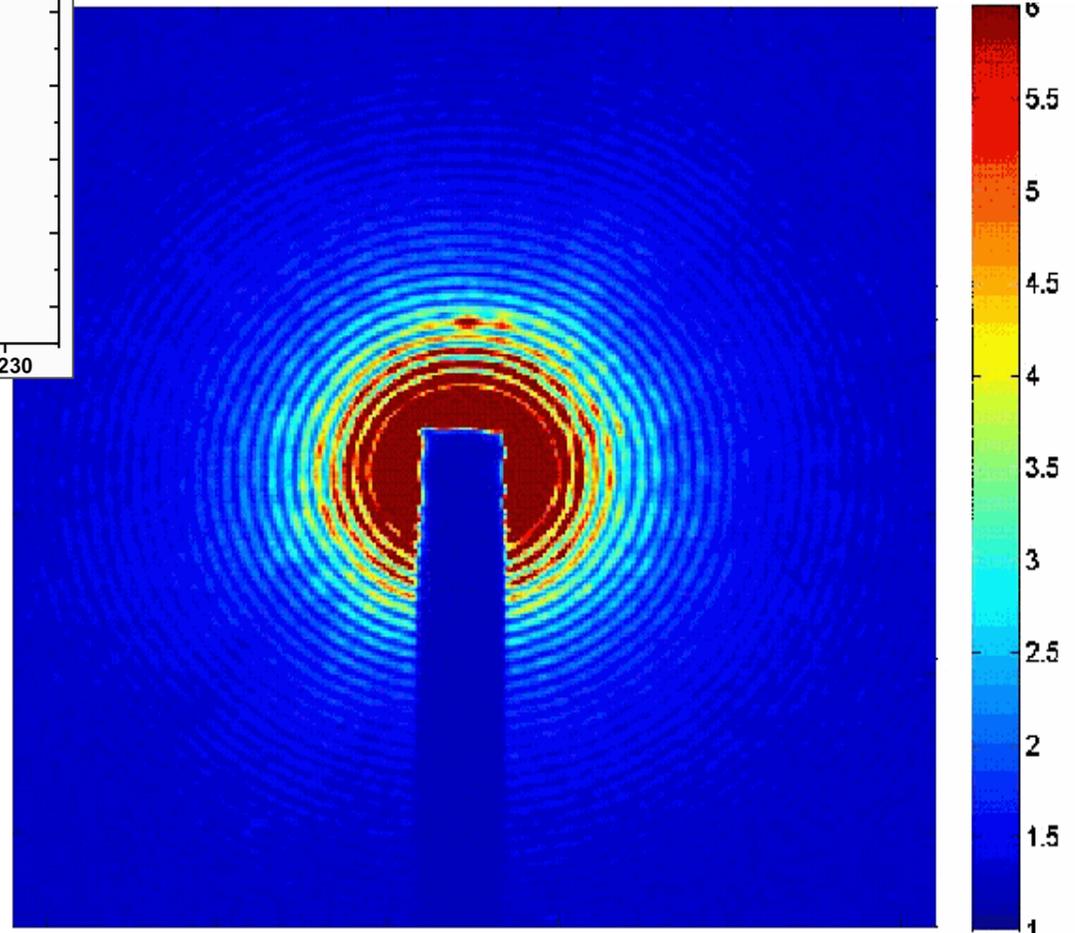
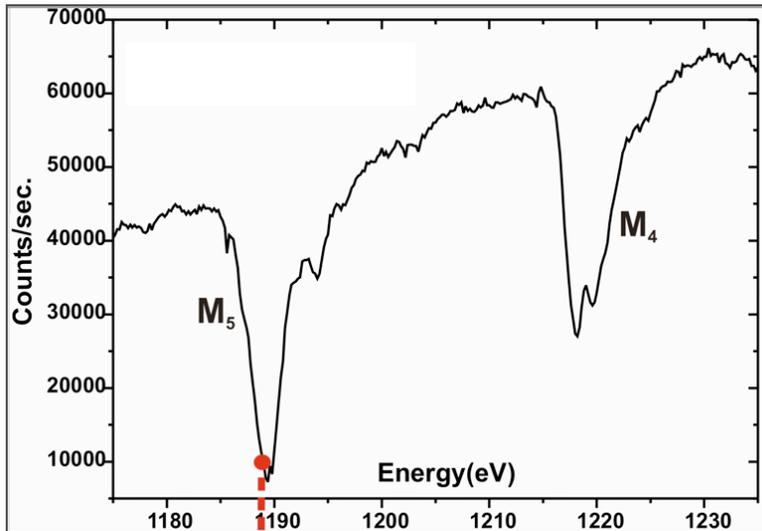
# Lensless approach



# Setup



# Resonant scattering vs. photon energy



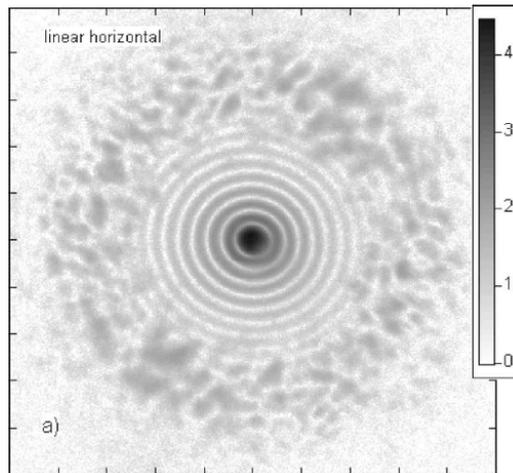
# Coherent diffraction with linearly polarized light

Each helicity:  $|FT\{\text{probe} + \text{object}\}|^2 = |\hat{P}|^2 + |\hat{O}|^2 + (\text{cross-terms})$

RCP:  $I_R = [\hat{P} + \hat{O}_R]^2$

LCP:  $I_L = [\hat{P} + \hat{O}_L]^2$

For LP = RCP + LCP, we measure:  $I_R + I_L = 2(\hat{P}^2 + \hat{O}^2)$

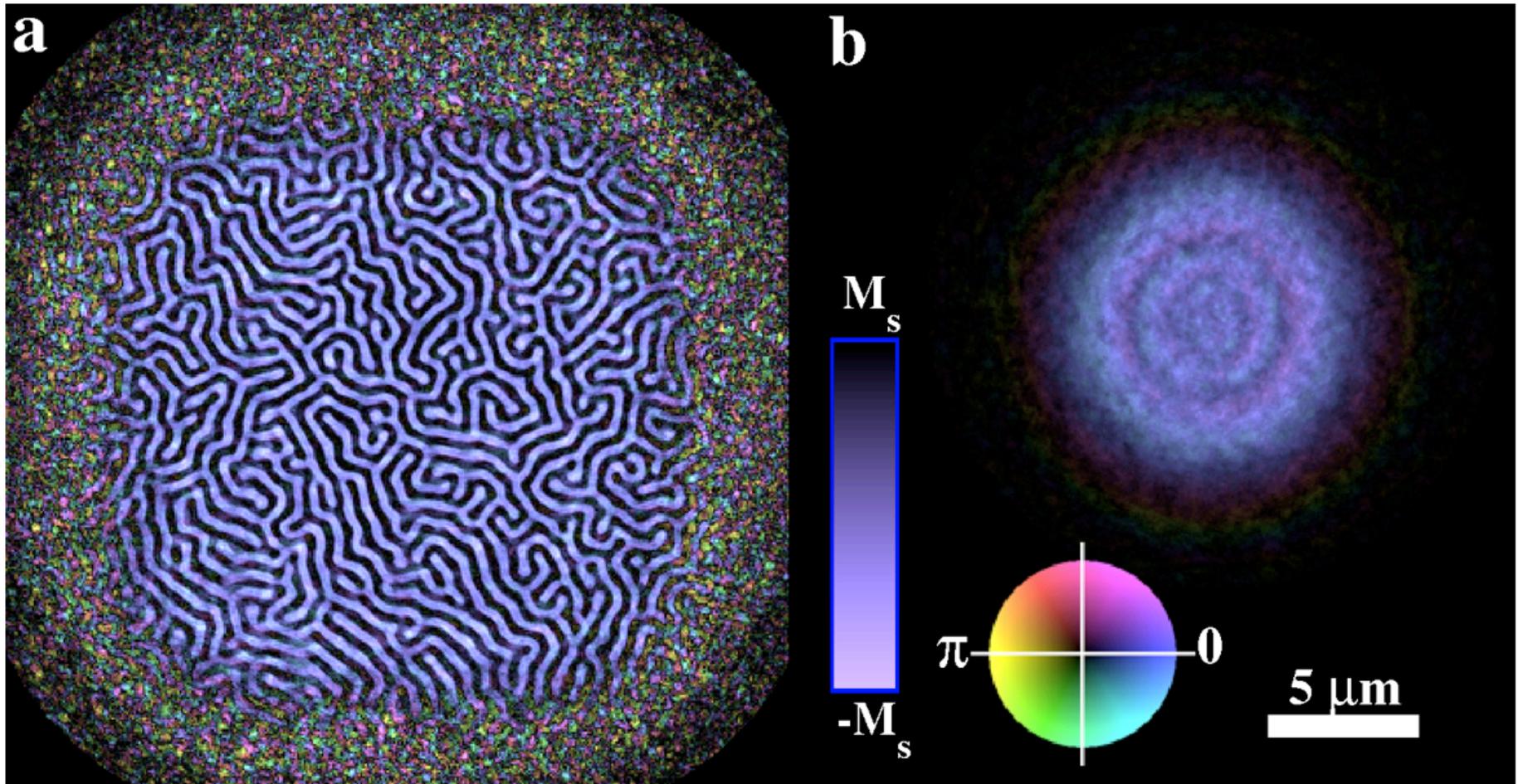


- Resonant scattering does not interfere with probe function
- Can just subtract probe (sample-out measurement) !

S. Eisebitt, PRB 68, 104419 (2003)



# Reconstructed domain structure and probe function

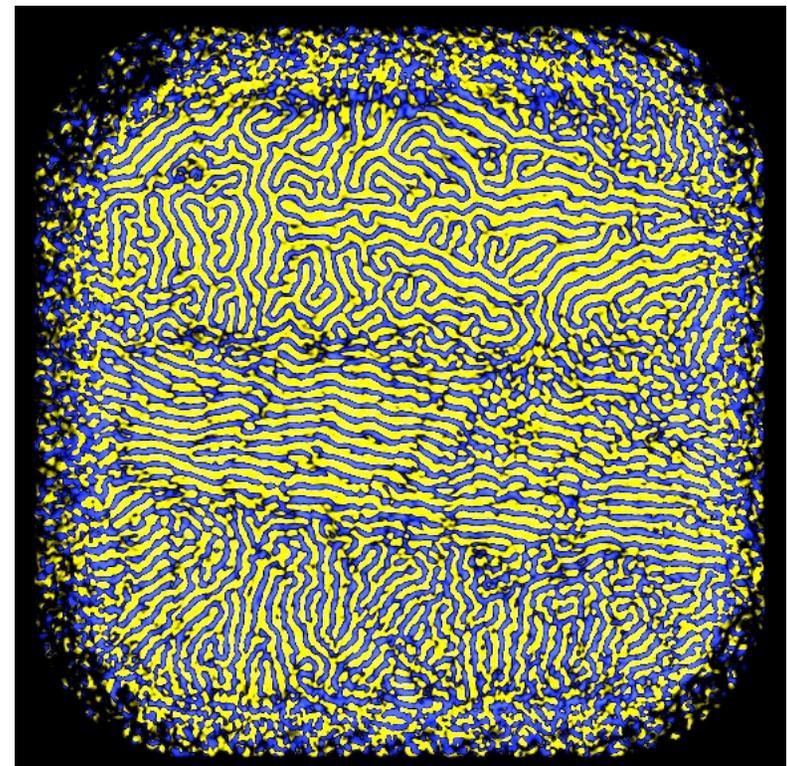
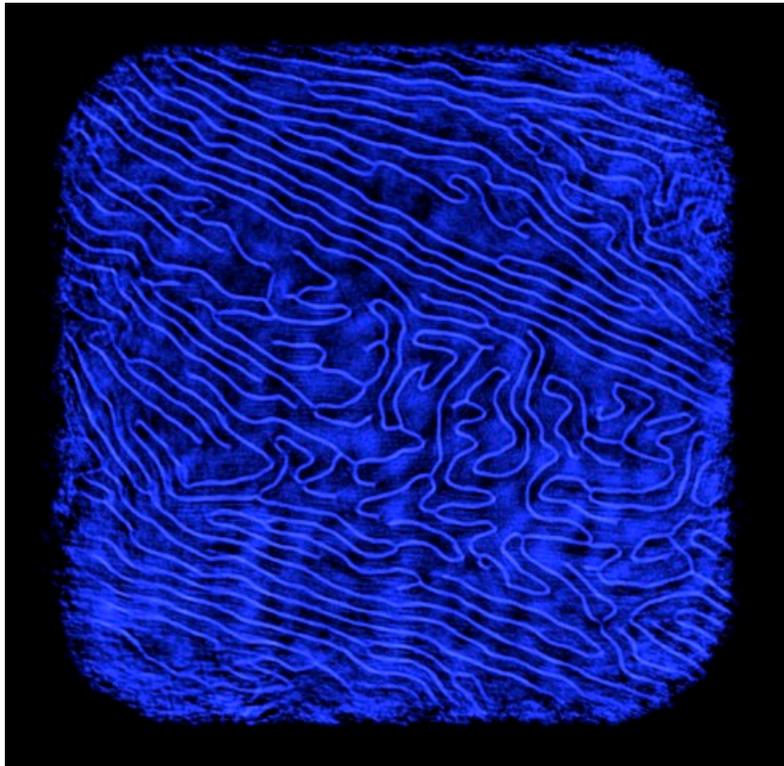


Sample scan:  $(14 \times 2 \mu\text{m})^2 \times (40 \times 1 \text{ s})$  exposures

A. Tripathi et al., PNAS (2011)

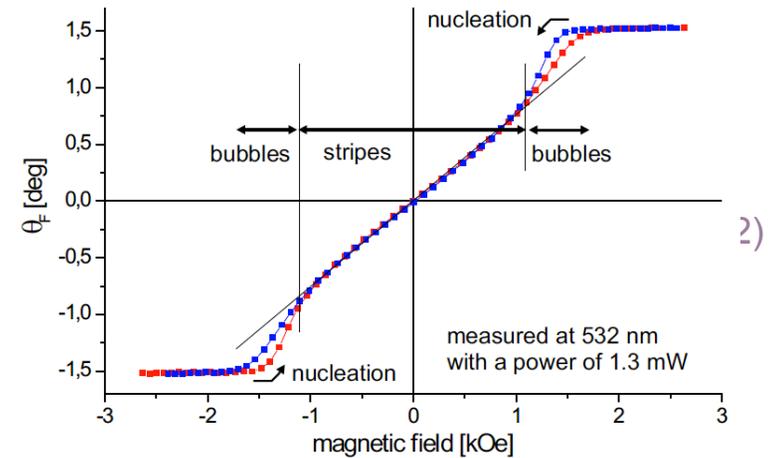


Many domain configurations are possible,  
depending on the magnetic history of the system

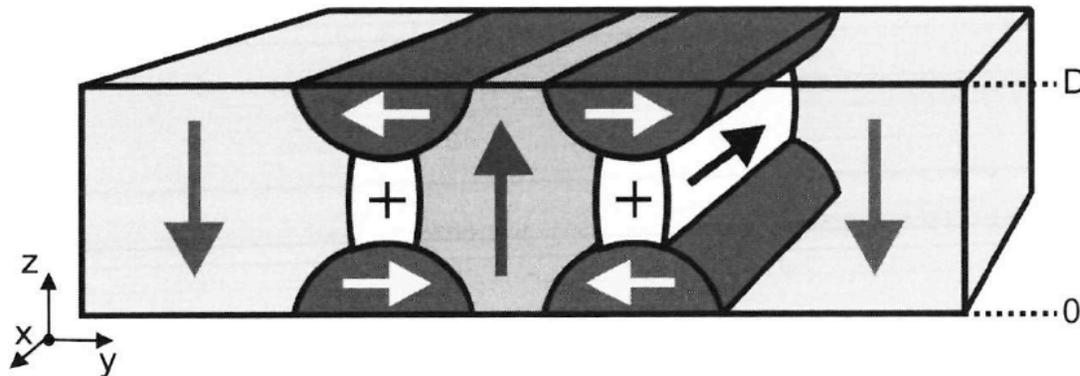


# Open questions

- Take GdFe system through entire hysteresis curve.
  - Does structure reproduce?



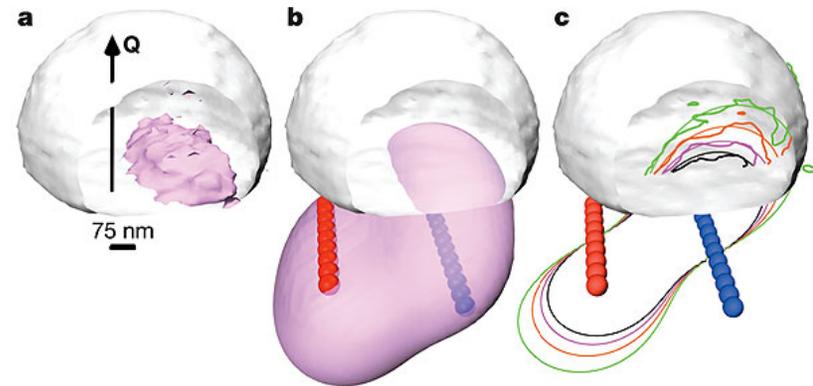
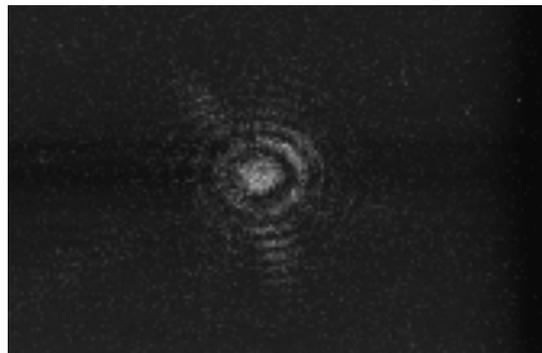
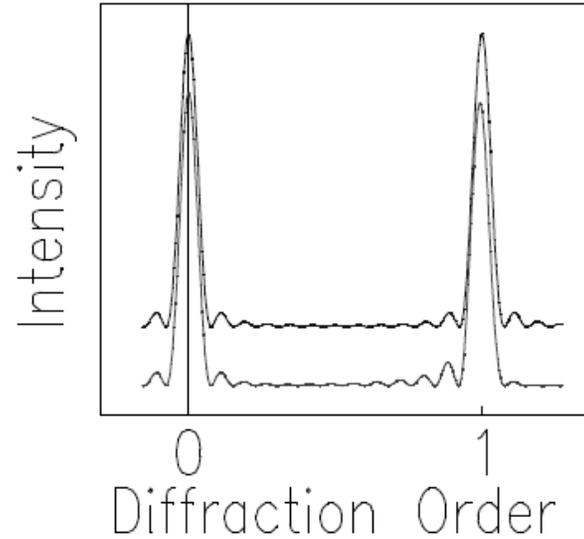
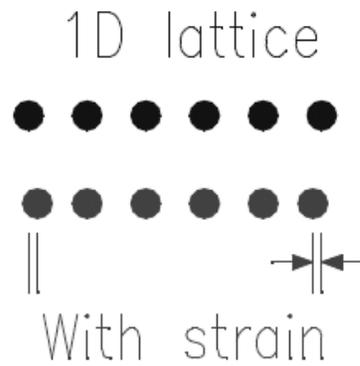
- With enough flux, can we resolve the domain walls themselves?



J.F. Peters, PhD Thesis (2003)



# Asymmetries in the diffraction yield lattice strain

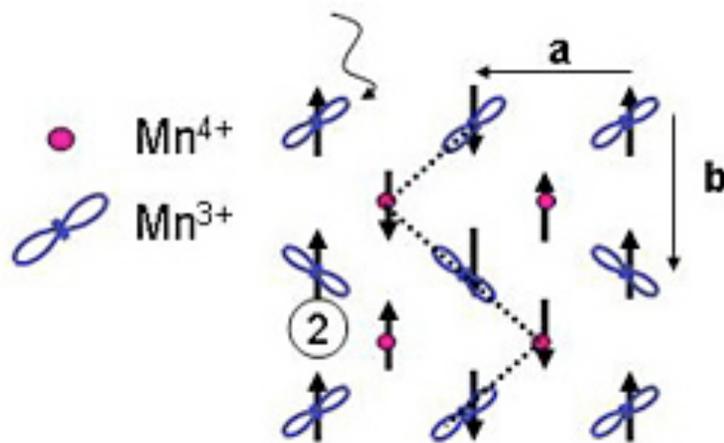


M. Pfeifer, *Nature* 442, 63 (2006)

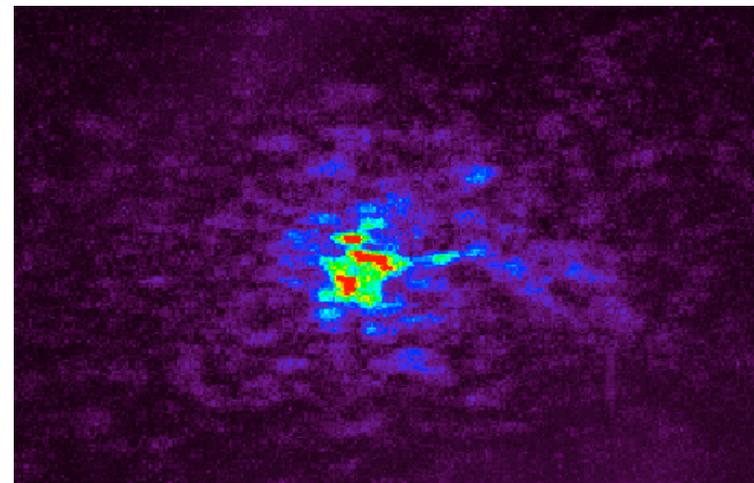


# Probing order parameters in the manganites

- **Colossal magnetoresistance** is exquisitely sensitive to competing orbital, charge, spin, and lattice degrees of freedom. Harnessing it may lead to denser magnetic storage and new spintronic and multiferroic devices.
- What are the order parameters and dynamics in doped manganites (e.g. in  $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ )? What do they tell us about nanoscale phase separation and CMR?



Some Mn atoms have an extra valence electron, causing orbitals to have a site-specific orientation, forming superlattice.

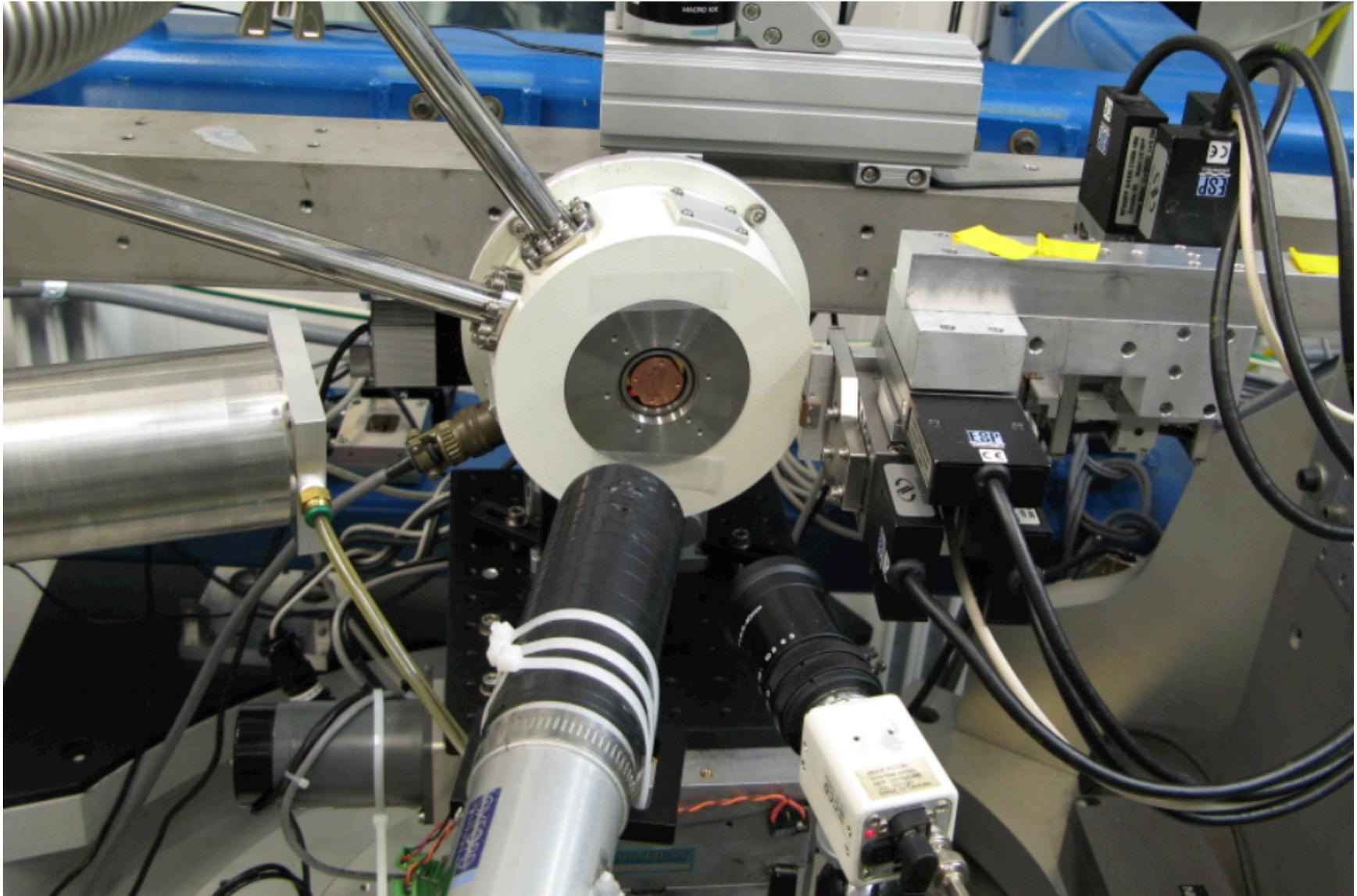


Resonant (010) "orbital speckle" from LaMnO<sub>3</sub> at 6.555 keV and 130 K (below T<sub>Néel</sub>) using a 1 μm beam. 300-800 nm domains consistent with Nelson (2002).

**Speckle contrast is good, but signal is weak: Exposures took ~100 s at APS. 3D and fast dynamics are inaccessible!**

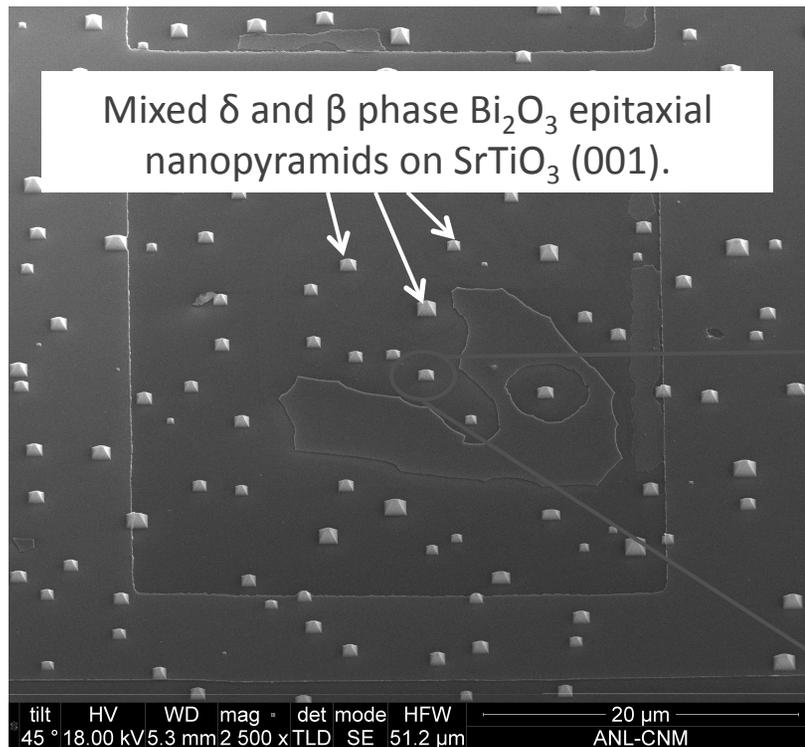


# Bragg coherent diffraction at cryo temperatures

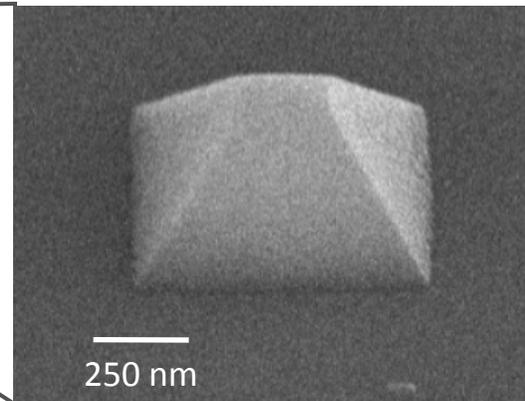
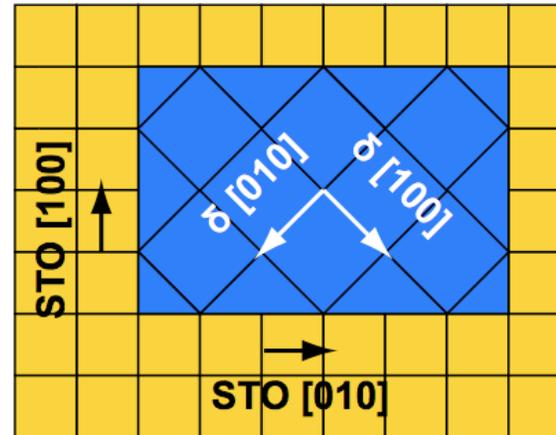


# Strain in $\text{Bi}_2\text{O}_3$ nanocrystals

- The high temperature  $\delta$ -phase  $\text{Bi}_2\text{O}_3$  is an exceptional oxygen conductor.
  - Epitaxially stabilized at room temp.



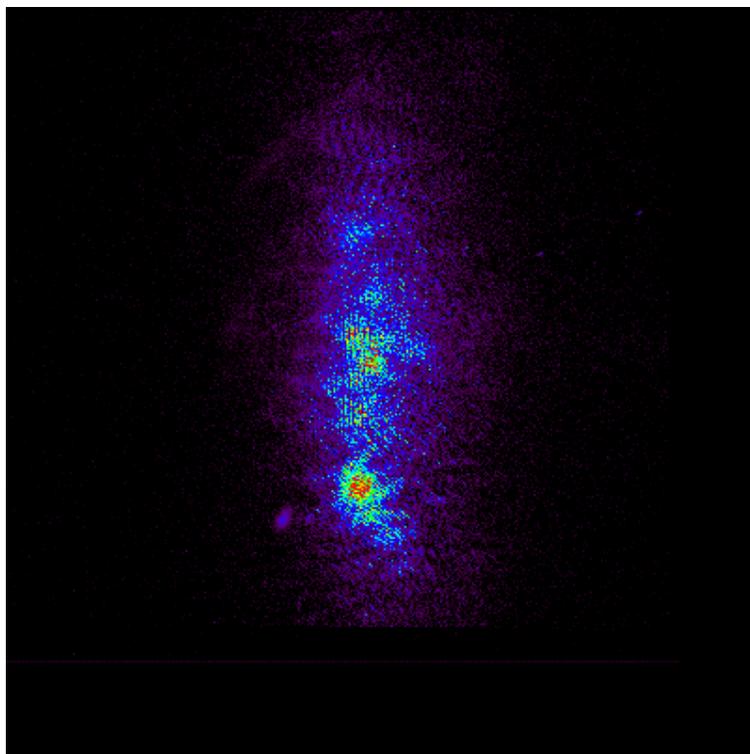
In-plane lattice orientations



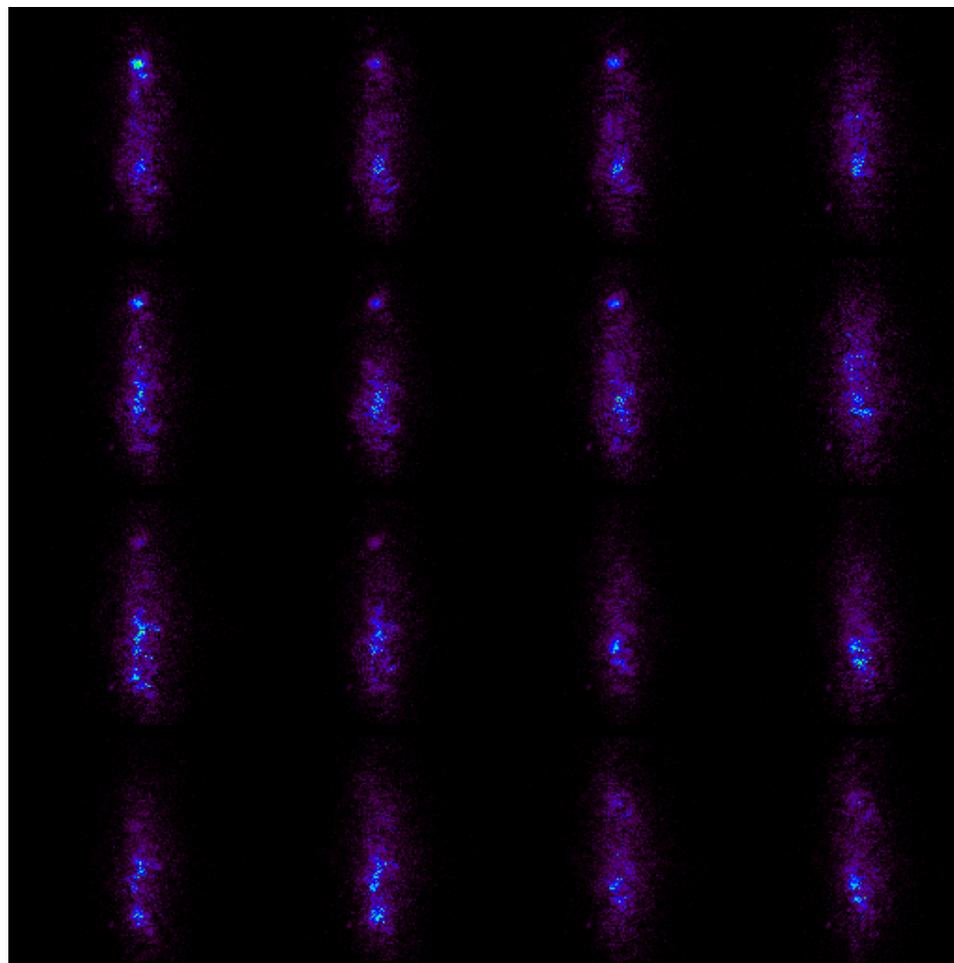
What is the role of elastic interfacial strain in phase stabilization and differentiation in  $\delta$  and  $\beta$   $\text{Bi}_2\text{O}_3$  nanostructures?



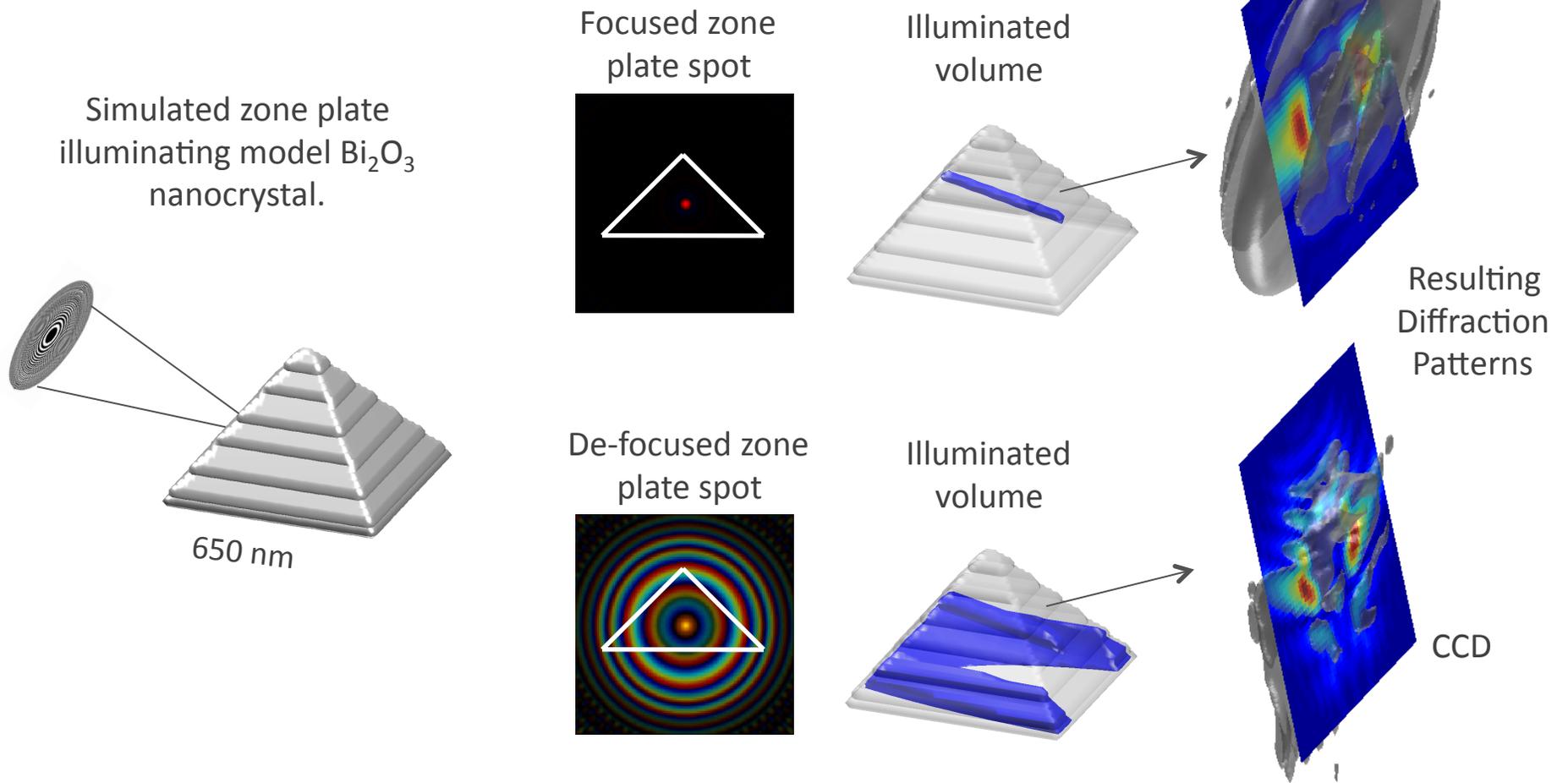
# Scanning Bragg data: $\text{Bi}_2\text{O}_3$ nanocrystals



9 keV,  $\sim 1.5 \mu\text{m}$  beam,  $0.5 \times 3 \mu\text{m}$  steps



# Convergent beam Bragg diffraction



Diffraction complexity changes with focus

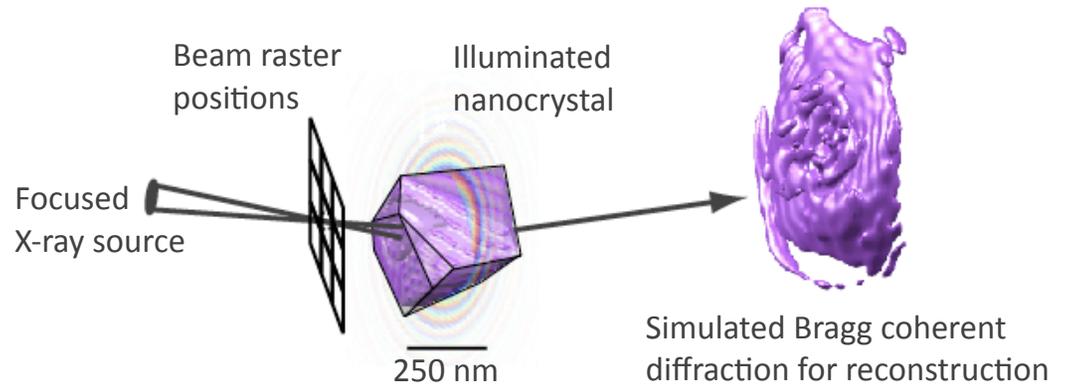


# Simulations are working!

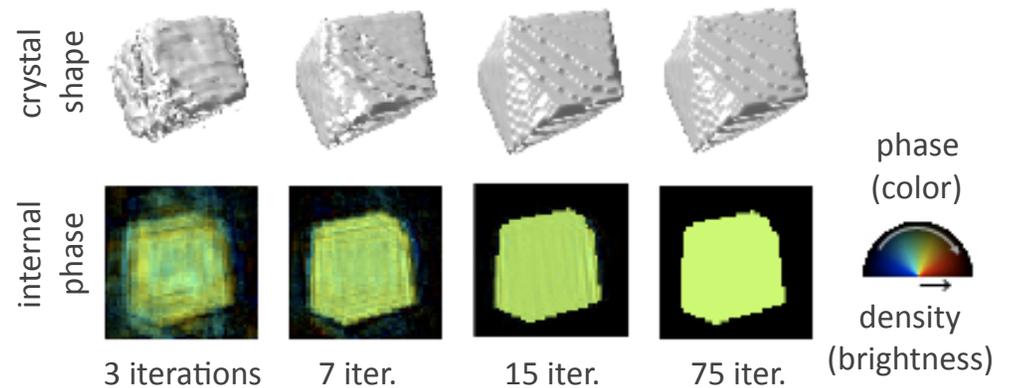
- **Achievement**
  - Successful simulation of 3D Bragg ptychography with focused beam
  - Relaxes the sample constraints of plane wave Bragg coherent imaging
  
- **Significance**
  - Enables the study of densely arranged, self-similar nanocrystals with focused coherent beams
  
  - Provides a roadmap to internal lattice strain imaging in extended crystal systems

S. O. Hruszkewycz,  
Opt. Lett. 36, 2227 (2011)

## Simulated nanocrystal diffraction



## Ptychographic reconstruction



## What do we need?

- Controllable polarization
- Stable beam, instrumentation
- Cryo to  $\sim 10$  K, magnetic field to  $\sim 2$  T
- Faster area detectors!

## What are the challenges?

- Getting to 3D: how to both scan and rock while studying same volume in sample (sphere of confusion problem)
- Maintaining stability across wide temperature range -> *more flux helps!*



# Conclusions

- We can probe magnetic ordering by resonant coherent diffractive imaging.
- We are making progress on scanning Bragg CDI, with both plane and convergent beams. However, significant challenges remain to handle sphere of confusion.
- We are currently at the ~20 nm scale on a 3rd generation SR with high contrast samples. Given 3rd-4th power scaling, can expect few-nm resolution with the ERL (worse for weakly scattering systems). Combined with resonant methods This opens up new territory, e.g. to study buried domain walls, that *cannot be addressed by other methods.*
- **The ERL offers unique truly unique opportunities for coherent x-ray imaging of both order and disorder in condensed matter exhibiting emergent properties.**

Preliminary layout view of an ERL upgrade to CHESS in the present CESR tunnel. A new superconducting cavity at (I) and accelerated to 2.5 GeV in the first half of the main linac, then to 5 GeV in the second half. The green lines show 18 possible beamline locations. The electron source is at the ESR (not shown). Spent electrons are sent to the dump (D). Their energy is extracted and the spent electrons are then sent to the dump (D).

Two superconducting linacs in one tunnel accelerate the electrons to 5 GeV. Person shown for scale.



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*and Thank you !*

