What Can We Get From Improved Transverse Coherence at a Synchrotron?

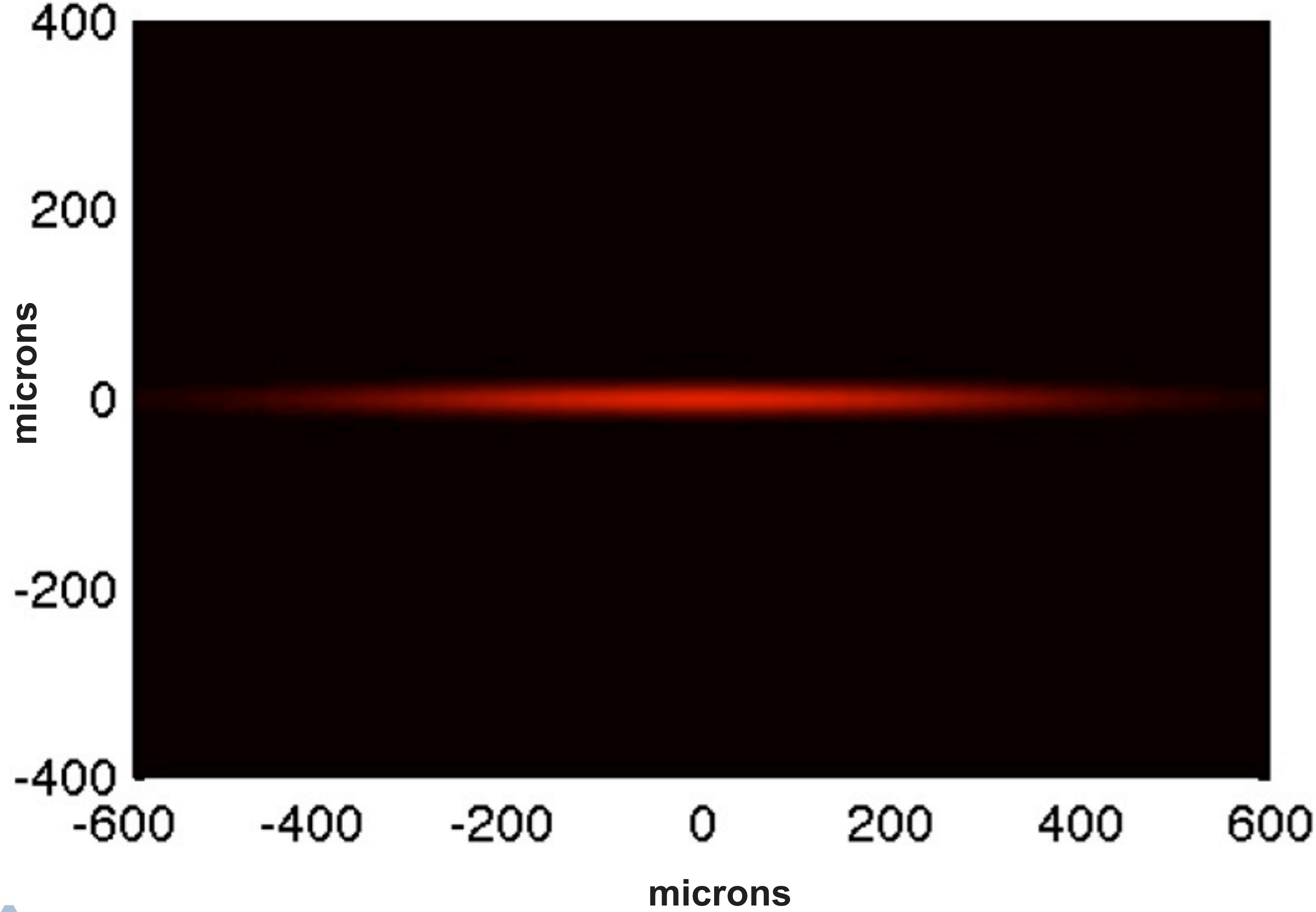
G. Brian Stephenson

SRI Satellite Workshop

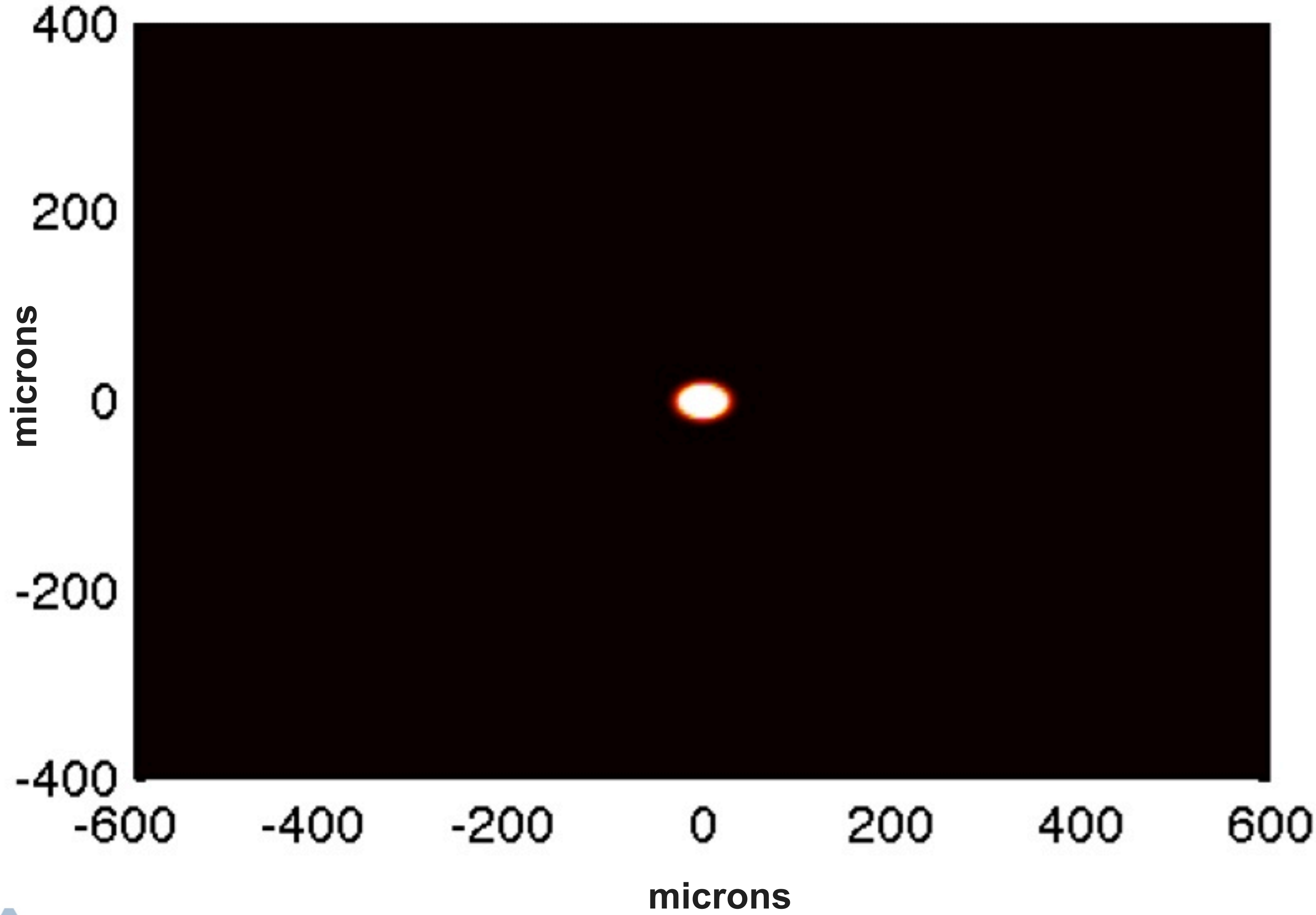
Emerging Opportunities in High Energy X-ray Science: The Diffraction Limited Storage Ring Frontier

July 13-14, 2015

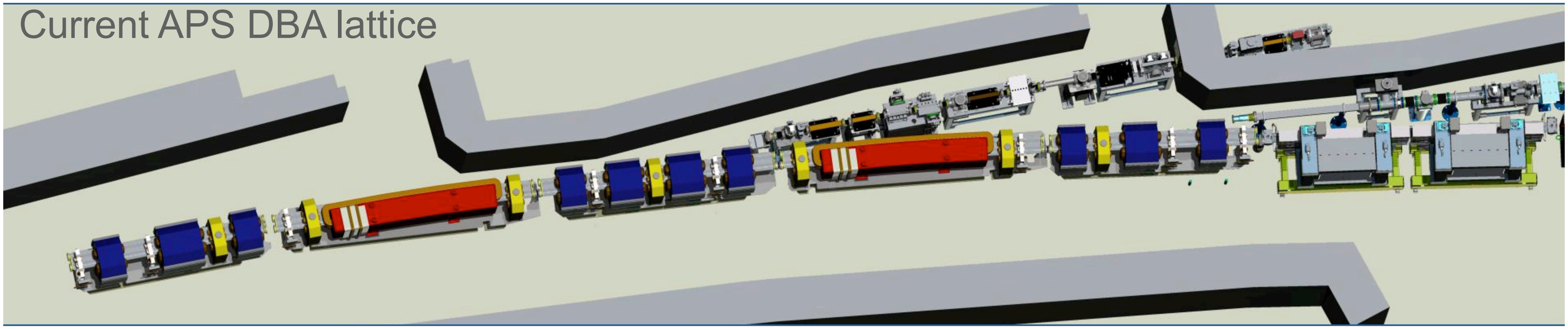
X-ray source size at APS today

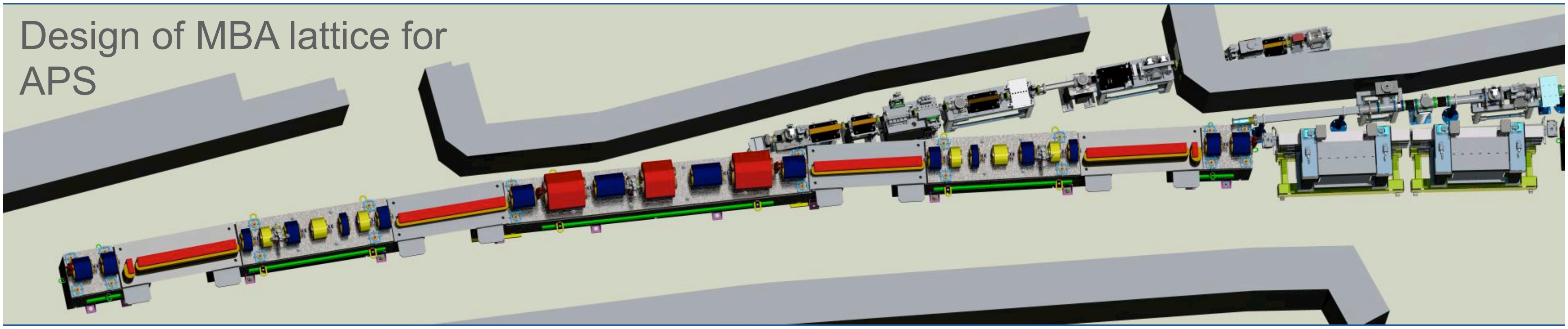


X-ray source size with APS MBA Upgrade



A multi-bend achromat (MBA) lattice for APS

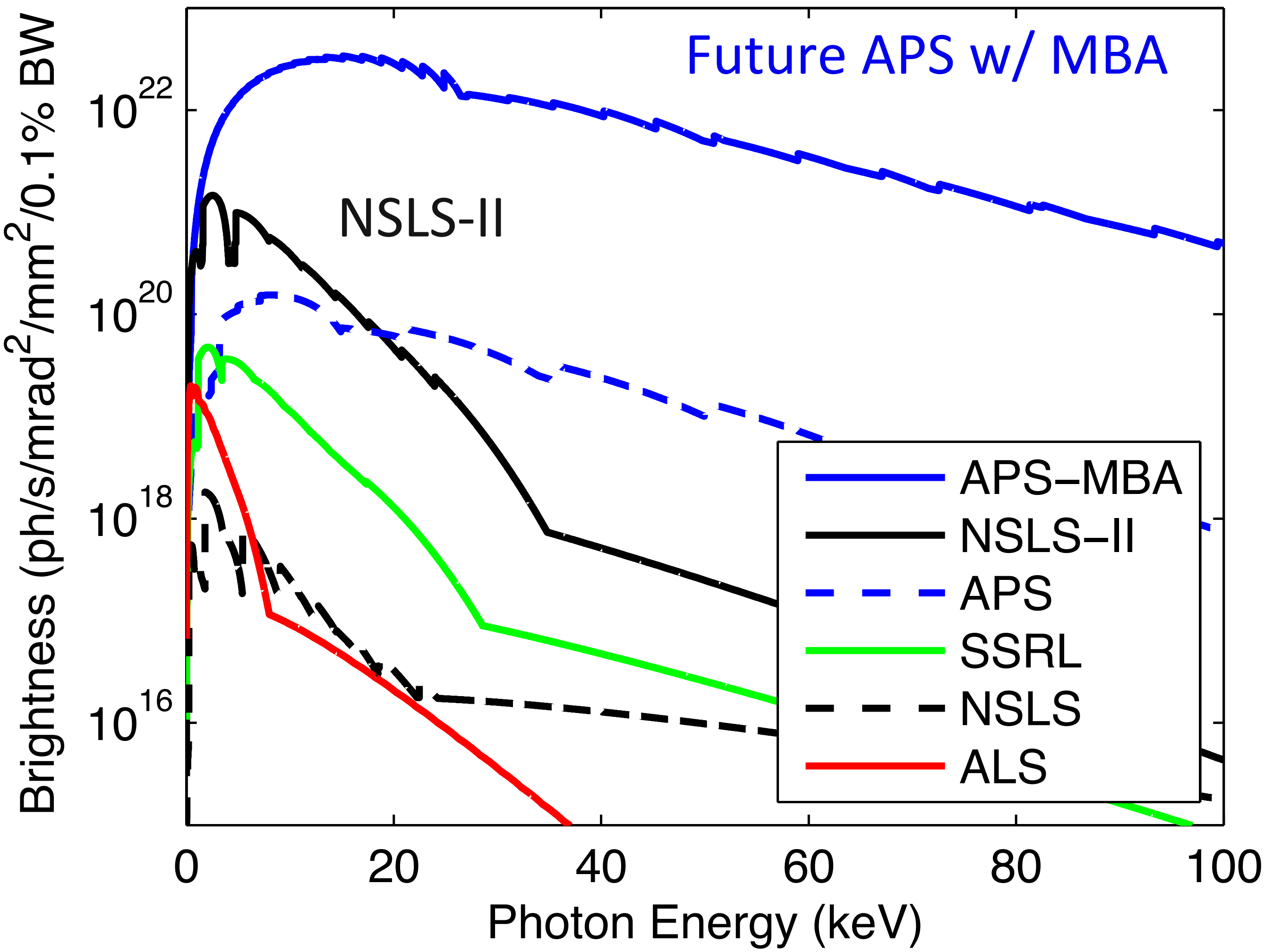




Horizontal emittance scales as N_D^{-3}

Increasing N_D from 2 to 7 improves emittance by a factor of 43

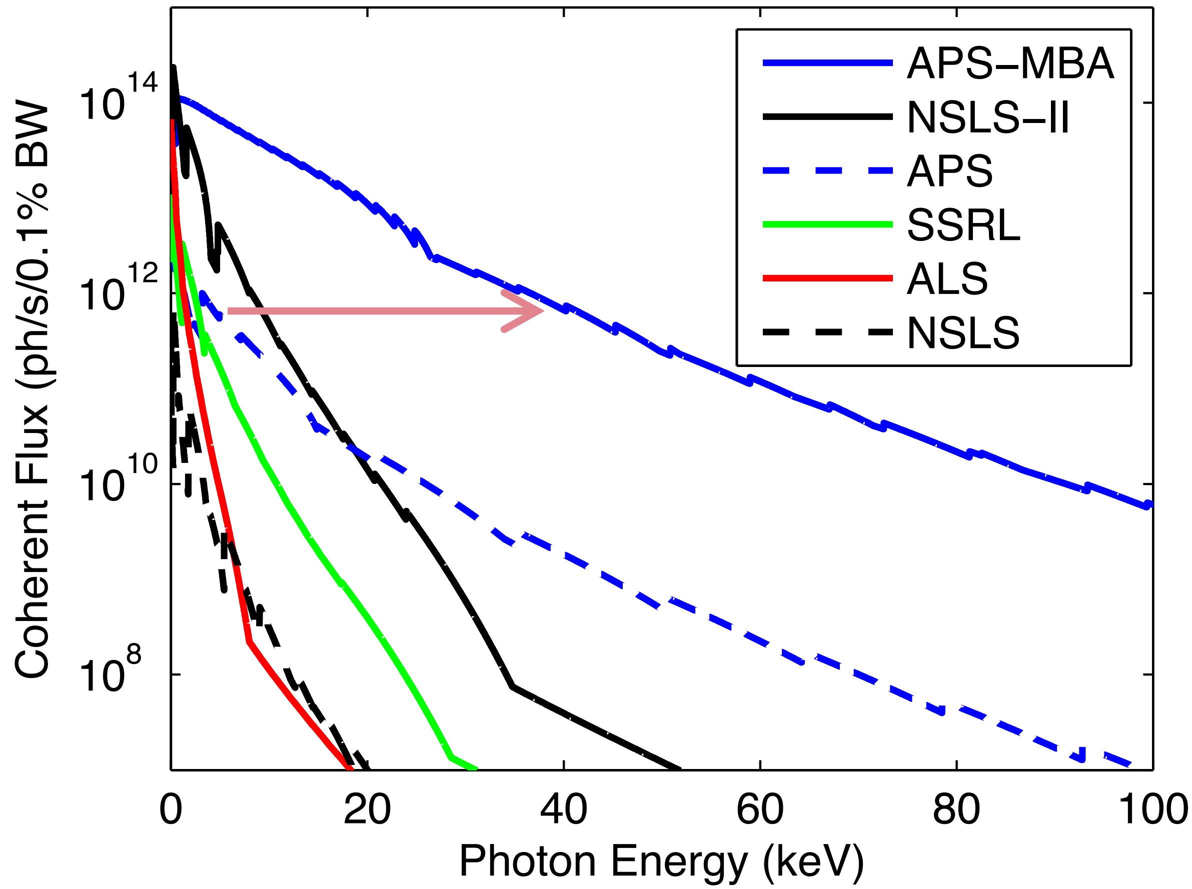
APS-U: Unprecedented brightness and coherence up to high energies



Brightness vs. x-ray energy at top beamlines among BES synchrotron facilities

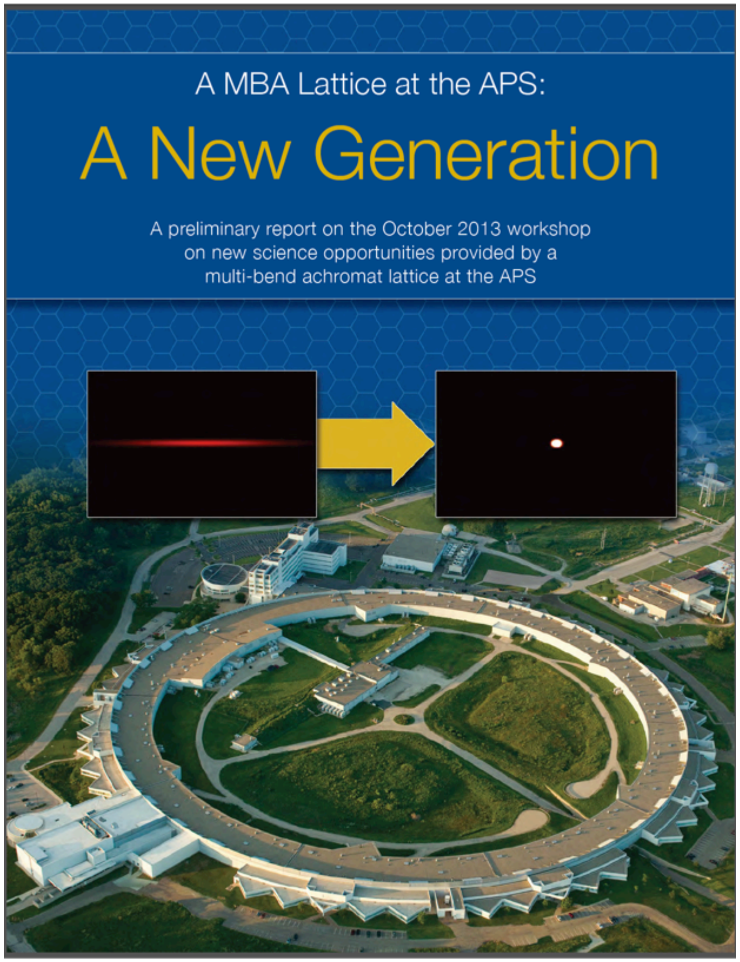
APS-U: Unprecedented brightness and coherence up to high energies

Coherent x-ray experiments possible now at 7 keV will be feasible using penetrating 42 keV light



Coherent flux vs. x-ray energy at top beamlines among BES synchrotron facilities

Current workshop series builds on previous, e.g. Oct 2013 Workshop on New Science Opportunities Provided by a Multi-Bend Achromat Lattice at the APS



- October 21-22, 2013 workshop report posted at www.aps.anl.gov/Upgrade/Workshops/2013/MBA-Technology/

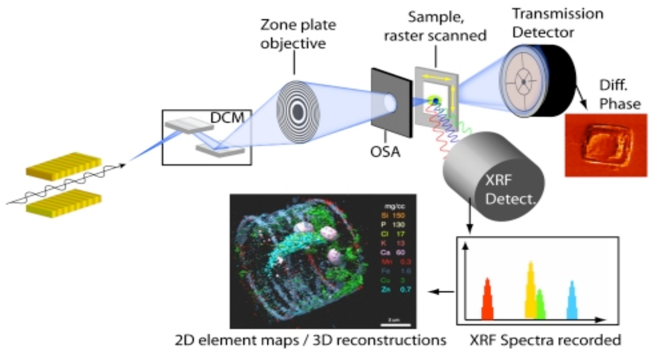
Outline: Main points

- High energy x-rays
  - Penetrate to enable in situ
  - Allow high q for atomic-scale resolution
- High transverse coherence enables:
  - Nanobeam focusing
  - Coherent diffractive imaging
  - Correlation spectroscopy

A new regime of scattering and spectroscopy with nanobeams: *nanoXRF*, *nanoXRD*, *nanoXAS*, *nanoRIXS*

X-ray fluorescence nano-tomography

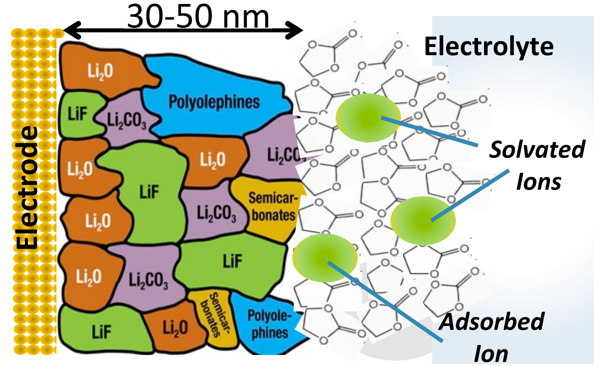
3D elemental mapping of functional mesostructures



de Jonge, et. al., PNAS 107, 15676 (2010).

nanoXRD

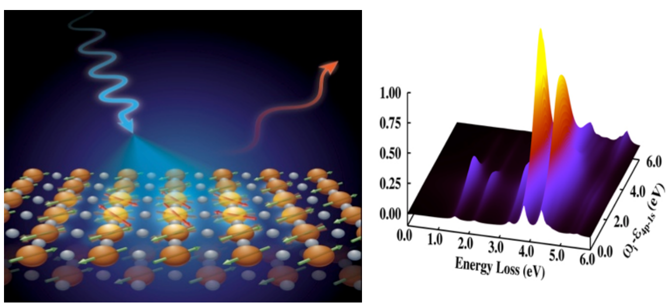
Formation, structure, and function of the solid-electrolyte interphase in batteries



JCESR

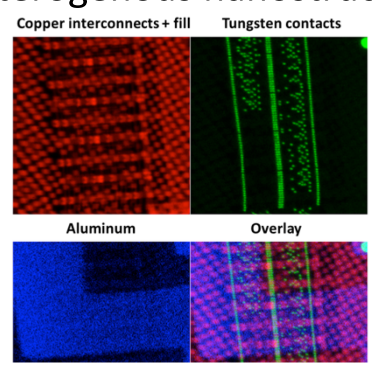
nanoRIXS

understanding coupled excitations in heterogeneous materials and nanostructures



nanoXRF

Understanding elemental composition in heterogenous nanostructures

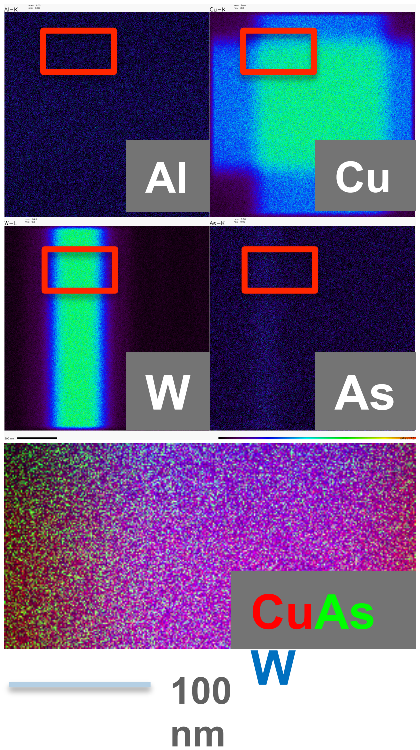


CMOS semiconductor

MBA will vastly expand the capability and capacity of scanned x-ray probes: high flux at resolution approaching 1 nm.

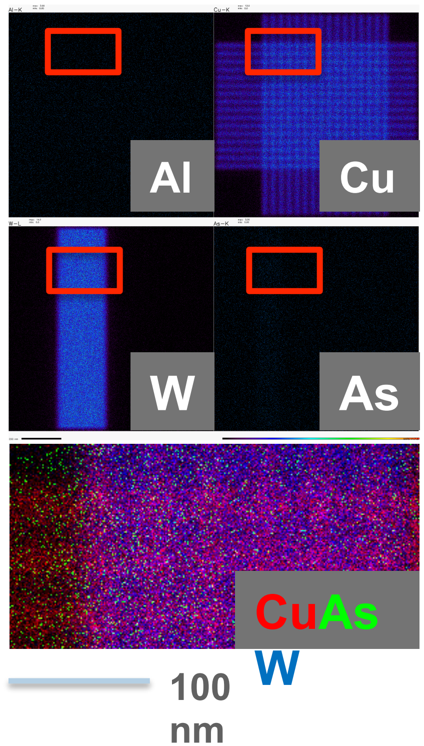
APS MBA upgrade, 5 nm spatial resolution: revolutionary

2-ID-D today, 120 nm spatial resolution: "work horse"



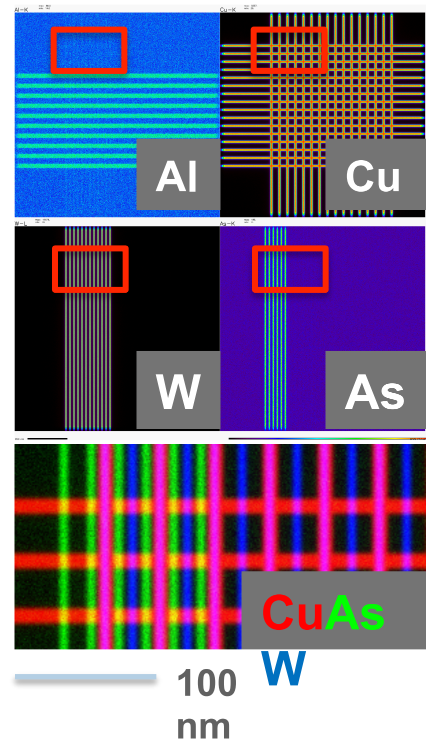
- Cannot resolve W, Cu structures, As doping
- Cannot detect Al

26-ID nanoprobe today, 35 nm spatial resolution: "cutting edge"



- Cannot resolve W structures, As doping
- Cannot detect Al
- Resolve Cu structures

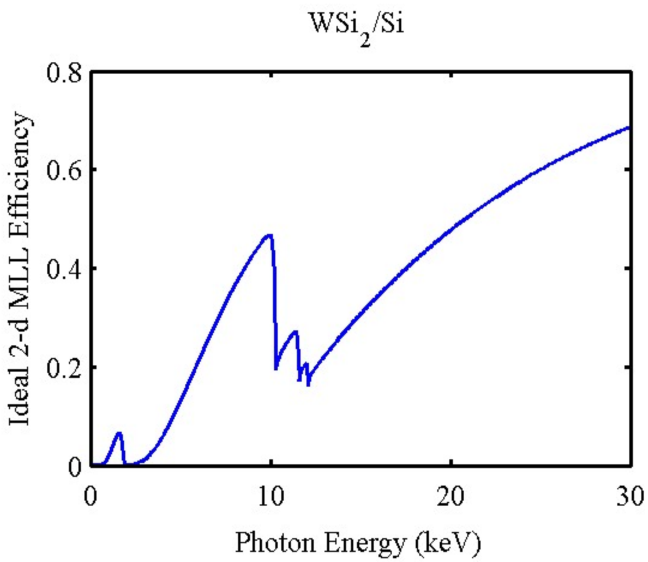
Simulation of MBA upgrade, 5 nm spatial resolution: "revolution"

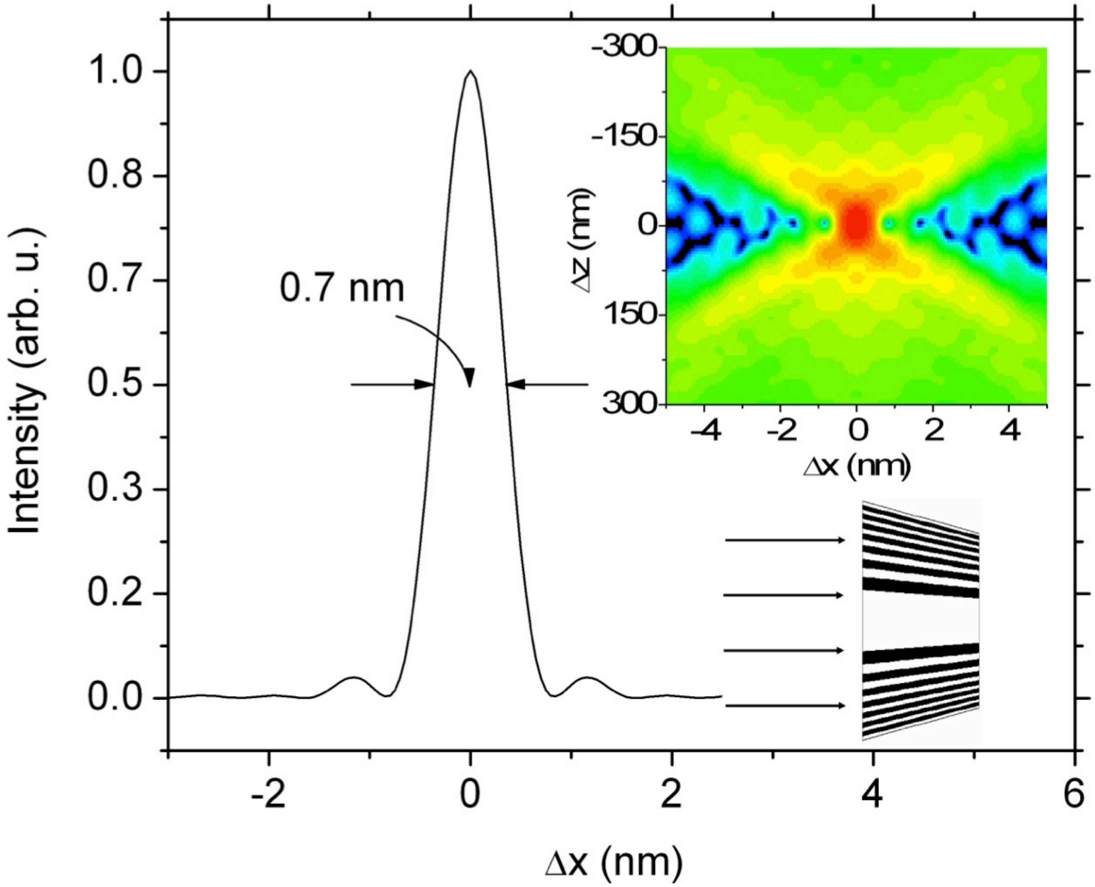


- Resolve Cu, W structures, and As doping
- Detect and resolve Al

Near-Atomic-Scale Focusing Possible with "Wedged" Multilayer Laue Lens

- Calculations show that focusing to below 1 nm is possible using "wedged" layers
- Efficiency increases at higher photon energy



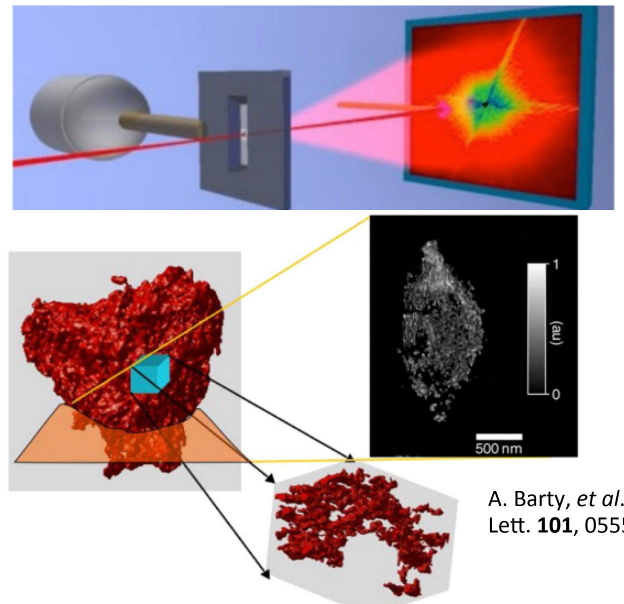


H. Yan et al., *Physical Review B* **76**, 115438 (2007)

Coherence provides transformation in X-ray imaging

Coherent Diffraction Imaging

- Resolution limited by wavelength and sample stability – not optics.
- Recover real and imaginary parts of refractive index: magnetization, composition, bonding configuration.
- Challenge: reach atomic scale.

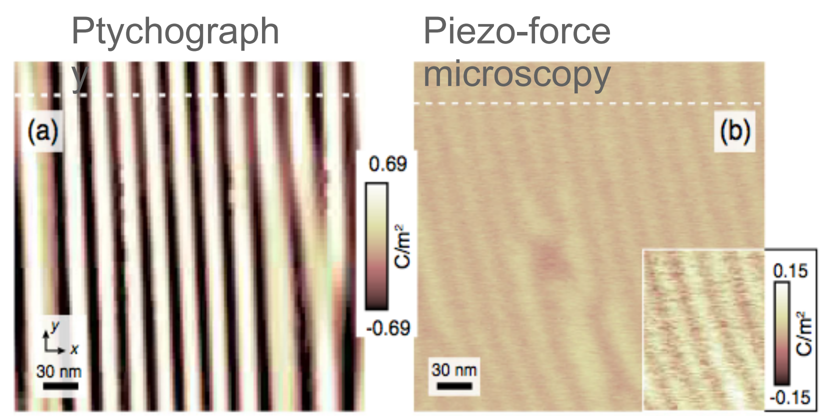


Nanofoam Diffraction Pattern/ Reconstruction

A. Barty, *et al.*, Phys. Rev. Lett. **101**, 055501 (2008)

Wavelength-Resolution Ptychography

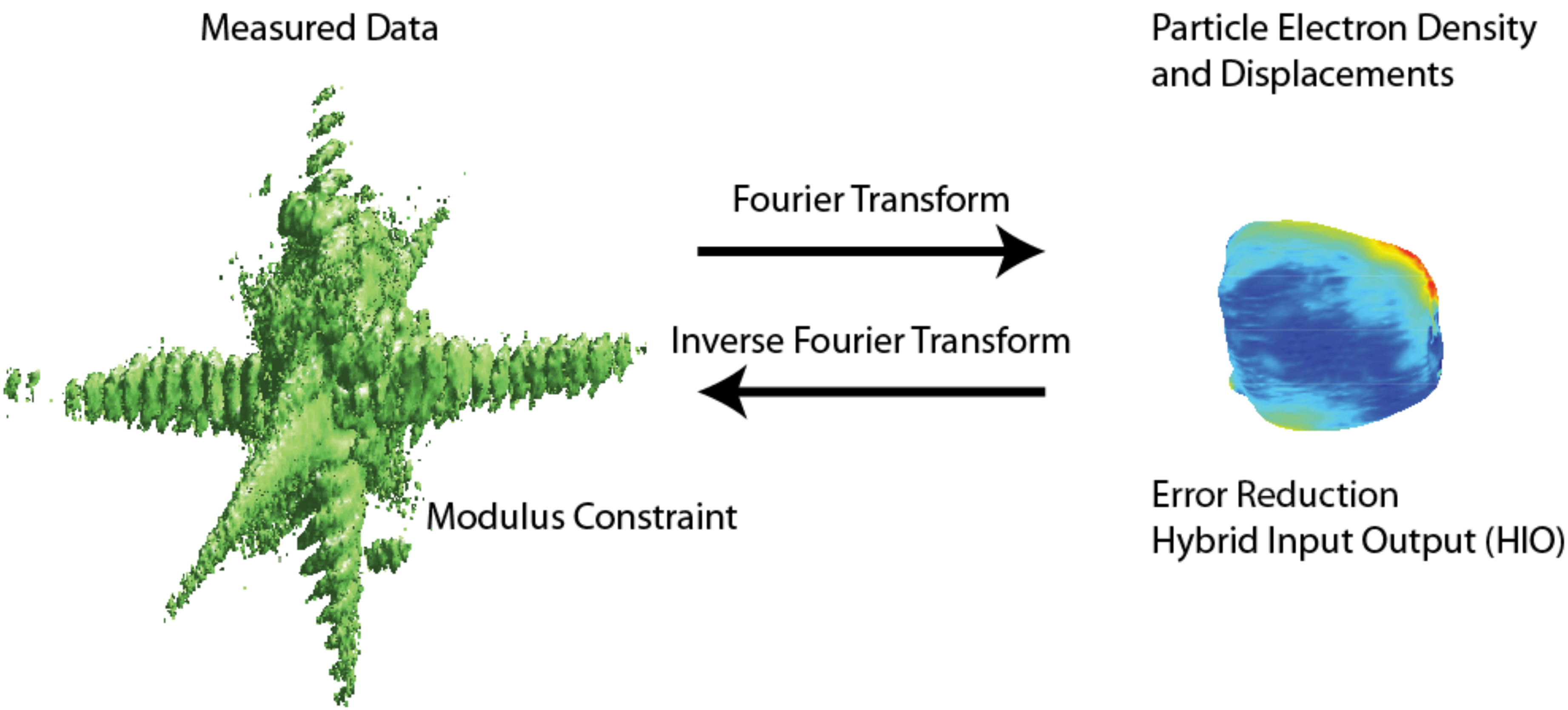
- CDI adapted to continuous samples with scanned-beam **ptychography**, resolution far better than focused X-ray spot size.
- Coherent imaging techniques to approach wavelength resolution from improved coherent flux



Polarization Domains in $PbTiO_3$
Hruszkewycz *et al.*, PRL (2013)

MBA will enable *in operando*, multimodal imaging approaching atomic resolution.

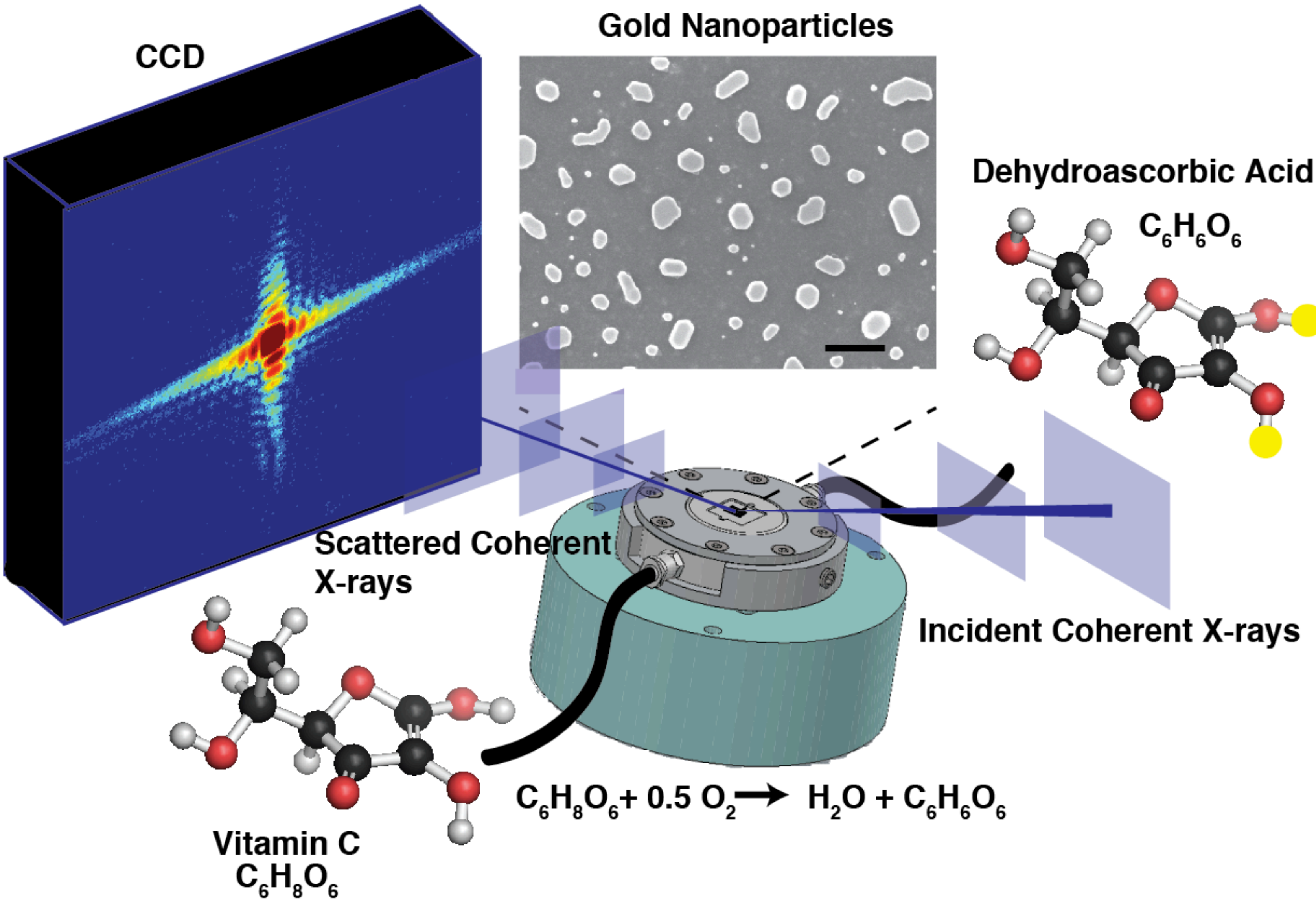
Bragg Coherent Diffraction Imaging

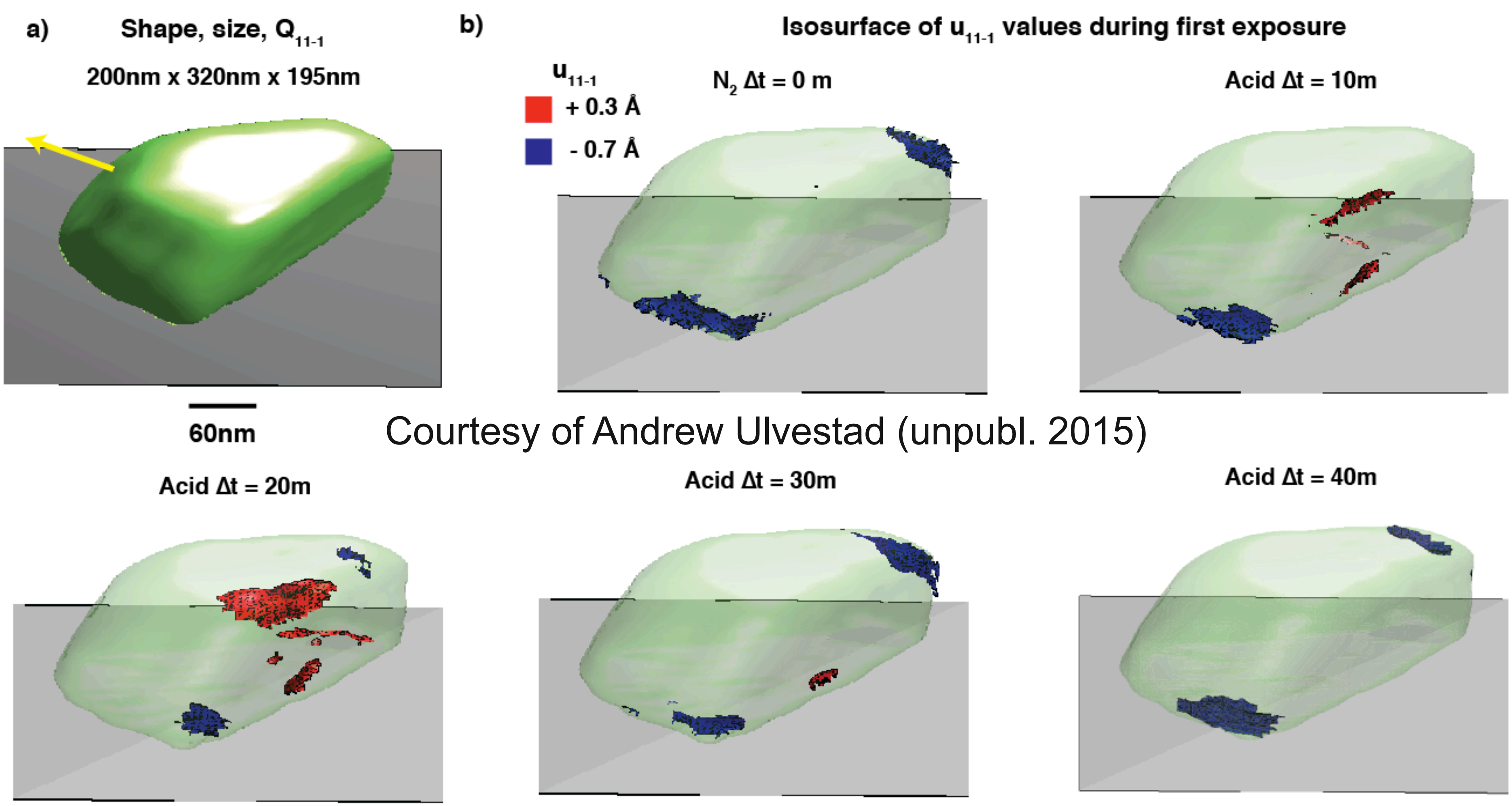


- Basic idea: record coherent diffraction, use computer methods to obtain image of strain fields
- Powerful, new type of imaging enabled by high coherent x-ray flux, still being explored

Catalysis by Gold Nanoparticles

Courtesy of Andrew Ulvestad (unpubl. 2015)

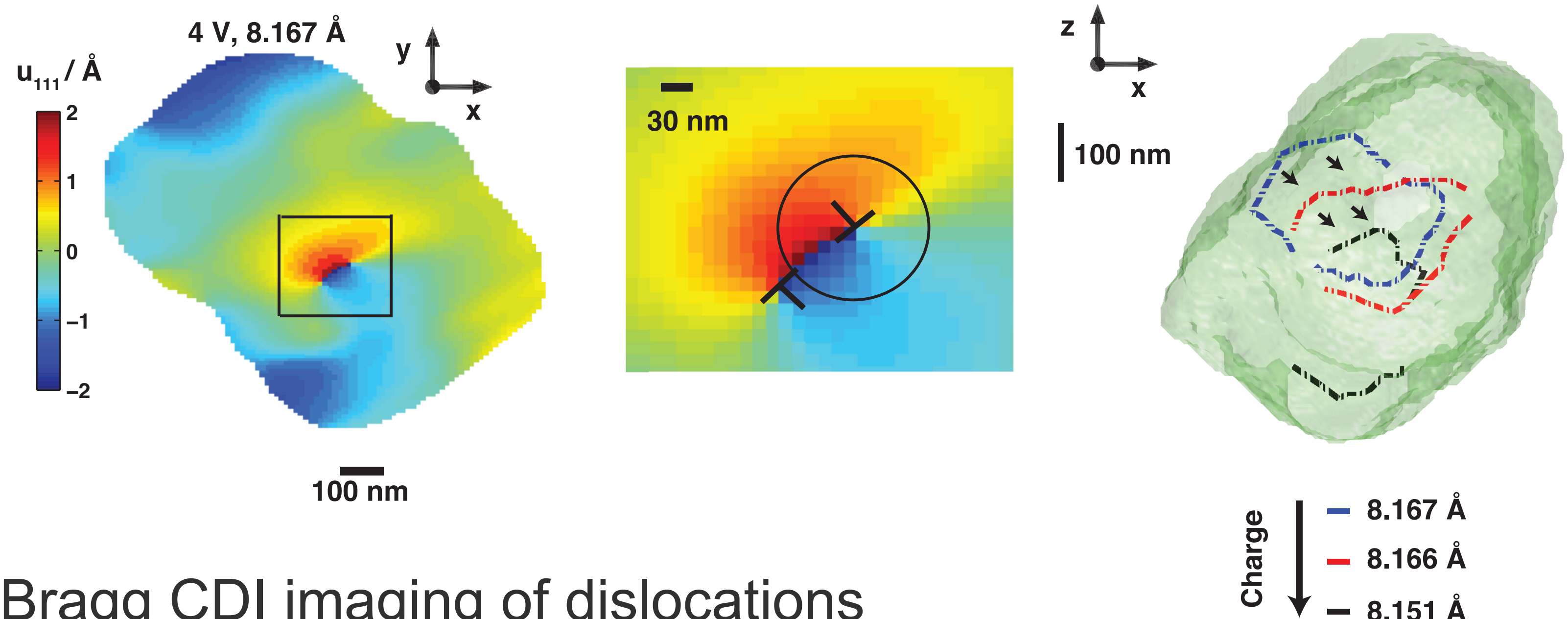




Courtesy of Andrew Ulvestad (unpubl. 2015)

- Observing distribution of catalytic activity on a single particle is a major breakthrough
- Direct observation of "hot spots" suggested in other work
- Provides new understanding of atomic-scale surface structure giving best performance, and how to optimize catalysts

In Situ Imaging of Edge Dislocations in "Aged" Single Cathode Nanoparticles



- Bragg CDI imaging of dislocations
- Can see dynamics during cycling
- Quantitative displacement field, gives local material properties

A. Ulvestad, O. Shpyrko et al., Science 348, 1344 (2015)

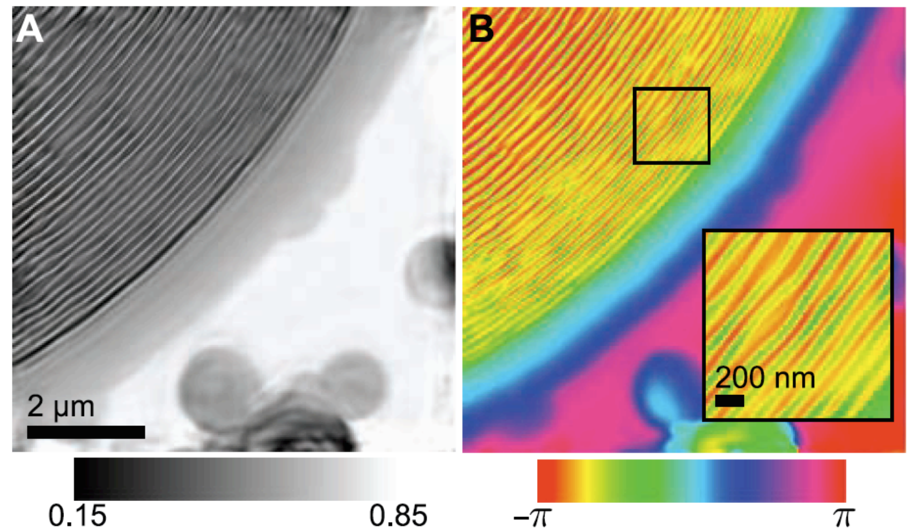
Ptychography: coherent diffraction imaging of region of extended objects

High-Resolution Scanning X-ray Diffraction Microscopy

Pierre Thibault,[1]* Martin Dierolf,[1] Andreas Menzel,[1] Oliver Bunk,[1] Christian David,[1] Franz Pfeiffer[1,2]

Coherent diffractive imaging (CDI) and scanning transmission x-ray microscopy (STXM) are two popular microscopy techniques that have evolved quite independently. CDI promises to reach resolutions below 10 nanometers, but the reconstruction procedures put stringent requirements on data quality and sample preparation. In contrast, STXM features straightforward data analysis, but its resolution is limited by the spot size on the specimen. We demonstrate a ptychographic imaging method that bridges the gap between CDI and STXM by measuring complete diffraction patterns at each point of a STXM scan. The high penetration power of x-rays in combination with the high spatial resolution will allow investigation of a wide range of complex mesoscopic life and material science specimens, such as embedded semiconductor devices or cellular networks.

SCIENCE VOL 321 18 JULY 2008 379



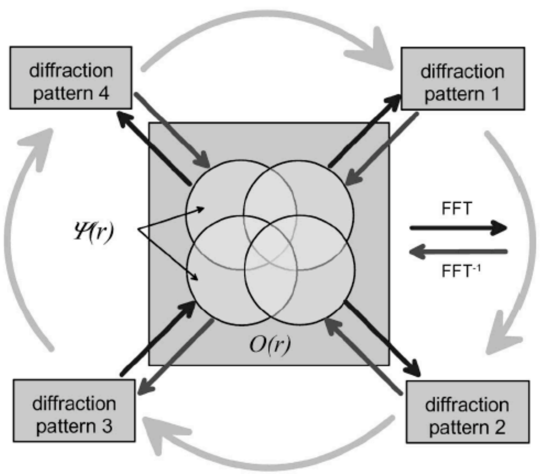
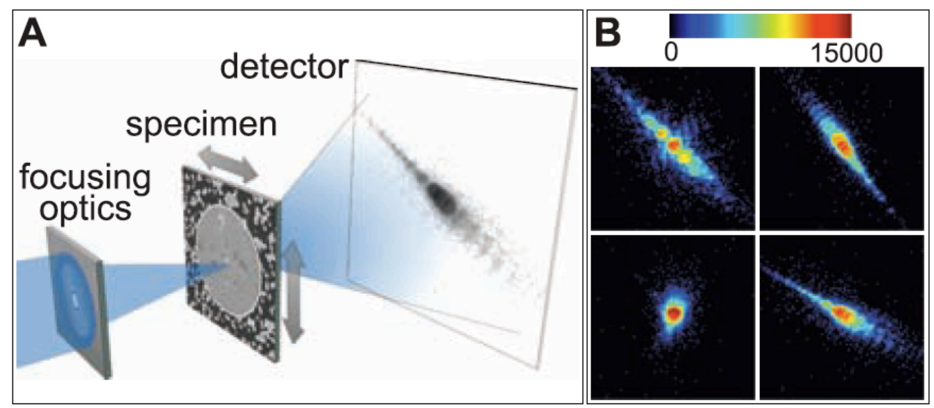


FIG. 2. Diagram of the phase-retrieval algorithm. The outer circular arrows indicate the position stepping within one iteration. The arrows within indicate (inverse) Fourier transforms and the desired input-output information.



First x-ray demonstration: Rodenburg *et al.*, *Phys. Rev. Lett.* **98**, 034801 (2007). Pilatus at Swiss Light source c-SAXS beamline

Observing individual point defects inside functioning devices with single-atom sensitivity

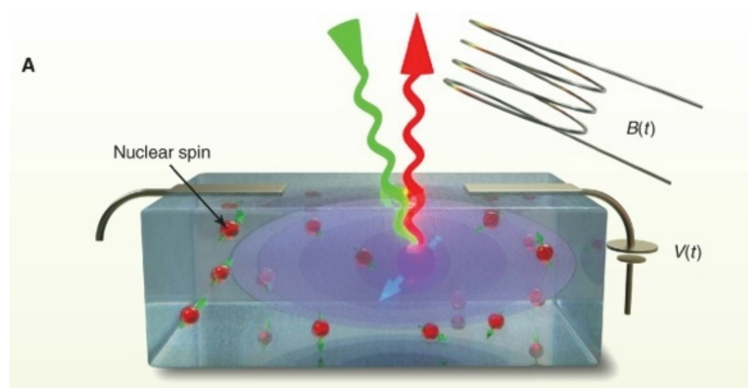
Opportunity

- Quantum spintronics: cryptography, sensing, and quantum computers
- Manipulate interacting arrays of "designer atoms"

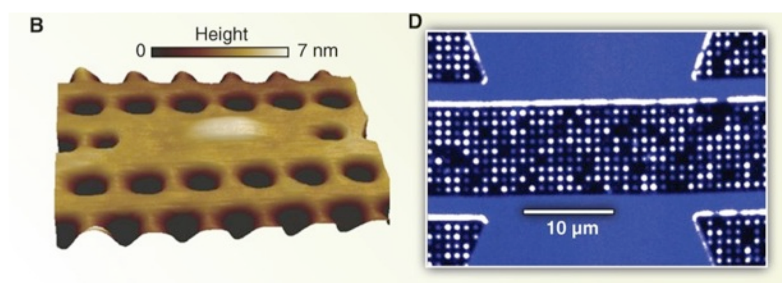
Gains From APS MBA Lattice

- Single-atom sensitivity for fluorescence
- Point defect strain fields at nm resolution; Bragg CDI measurements

Entangled spin states of single point defects in wide-bandgap semiconductors



Why do properties of each defect differ?



D. D. Awschalom et al., *Science*, 2013

Now: structure and composition of point defects are uncharacterized
APS MBA upgrade: gives sensitivity to strain and composition of single atoms through **factor of 100 to 1,000 improvement** in brightness

Ptychographic imaging of a single point defect

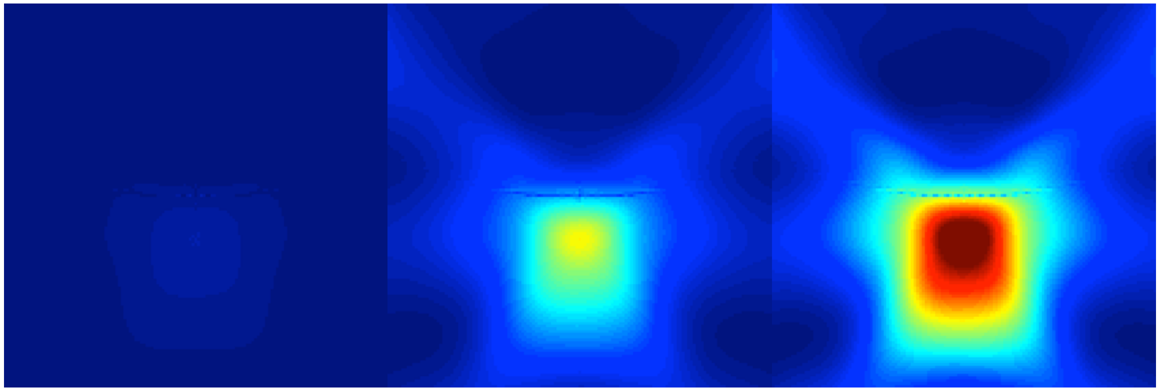
Characterize the nature and strain field of a single point defect

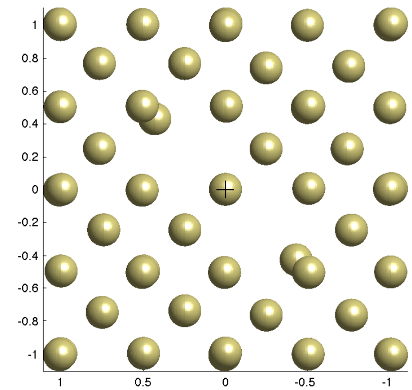
Scattering difference from defect, for various offsets

10 nm 5 nm 0 nm

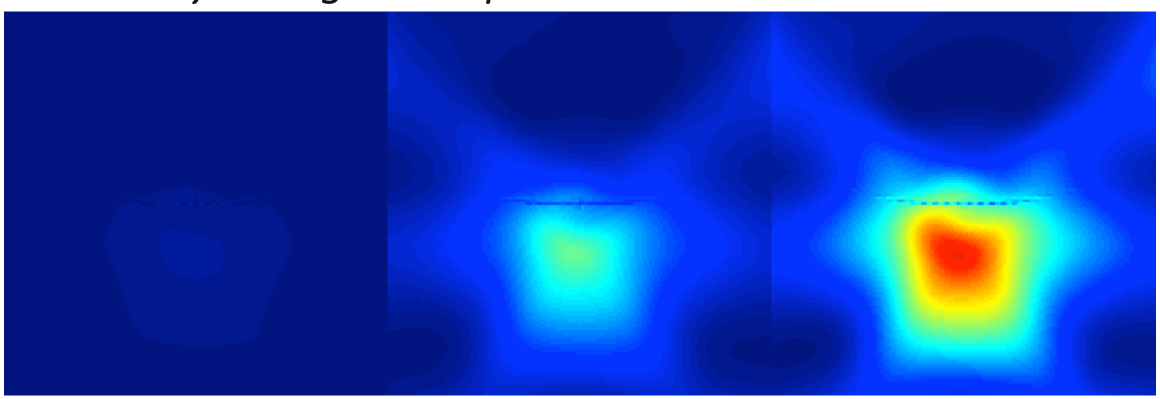
Simulated structure from MD

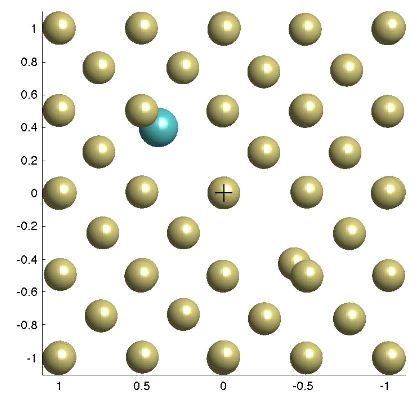
Vacancy in diamond





Vacancy-nitrogen complex in diamond





Unpublished calculations by S. Hruszkewycz and P. Zapol (2014)

Fast fluctuations with XPCS

X-ray photon correlation spectroscopy

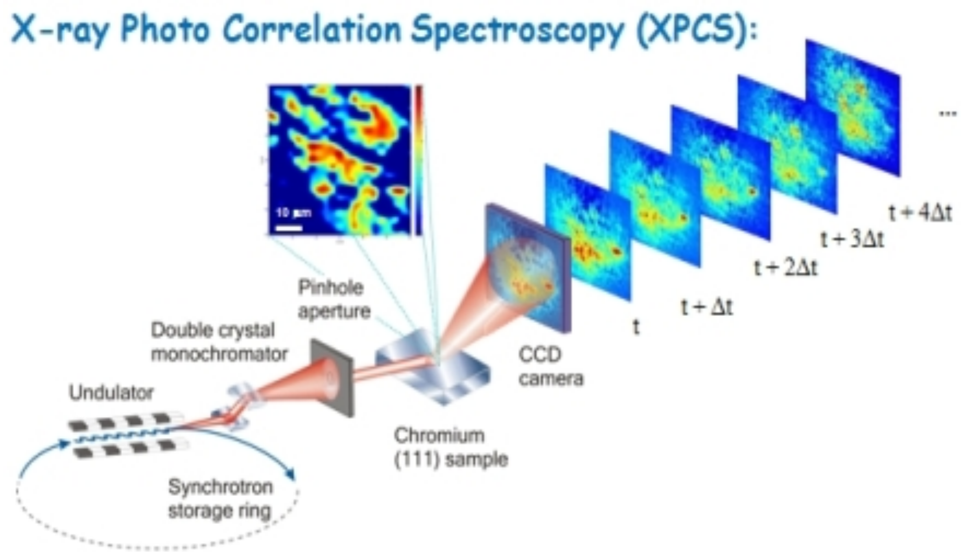
- Chemical, magnetic, and structural fluctuations

Accessible time scale proportional to (coherent flux)²

- 100 to 1000-fold increased brightness improves time resolution by 10^4 to 10^6

MBA enables ns-resolution studies of nm-scale fluctuations in

- Reaction-diffusion
- Self-assembly
- Domain wall motion
- Complex order parameters



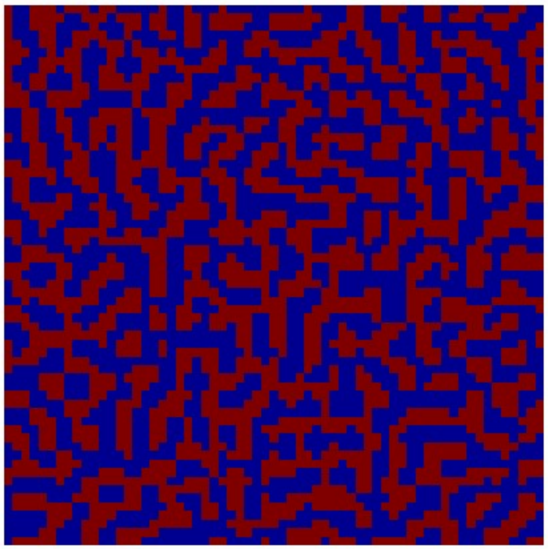
***Time to probe 1 ns fluctuations*:**

Today:	50,000 hours
MBA + modern BL:	5 hr to 3 min

MBA will revolutionize our ability to probe fluctuations at molecular length scale and nanosecond time scale

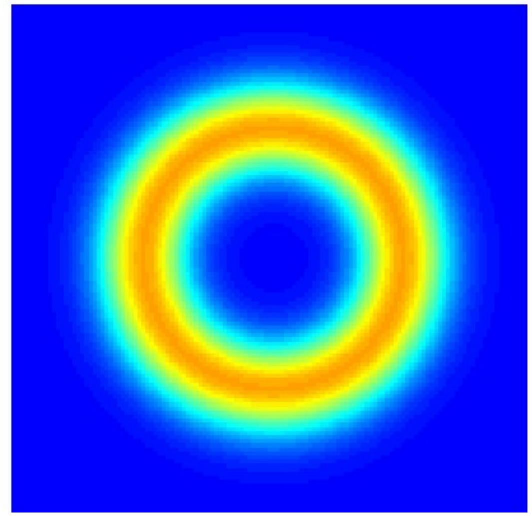
Scattering from Disorder: Speckle

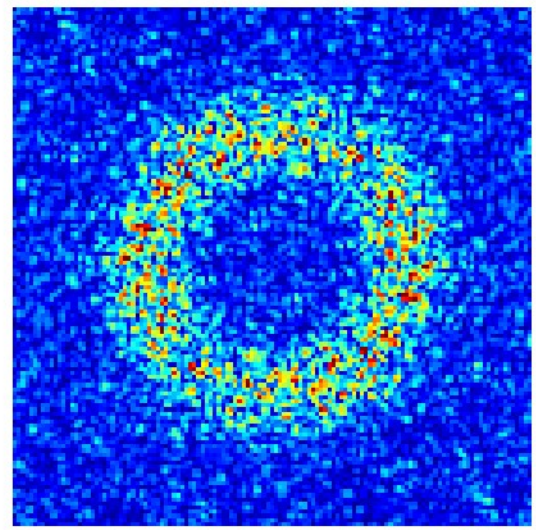
sample with disorder
(e.g. domains)



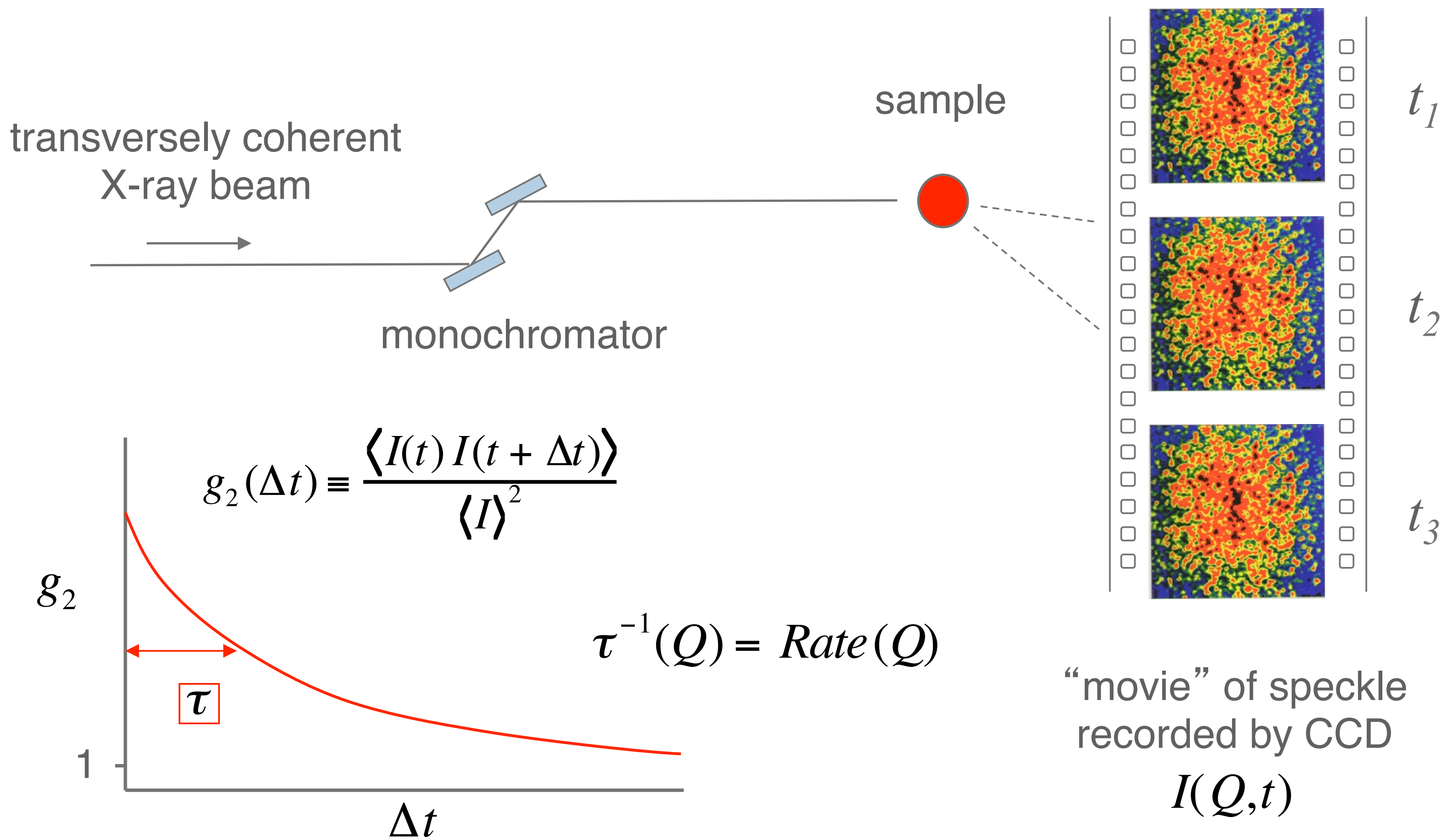
- *Incoherent Beam: Diffuse Scattering*
  - Measures averages, e.g. size, correlations
- *Coherent Beam: Speckle*
  - Speckle depends on exact arrangement

scattering





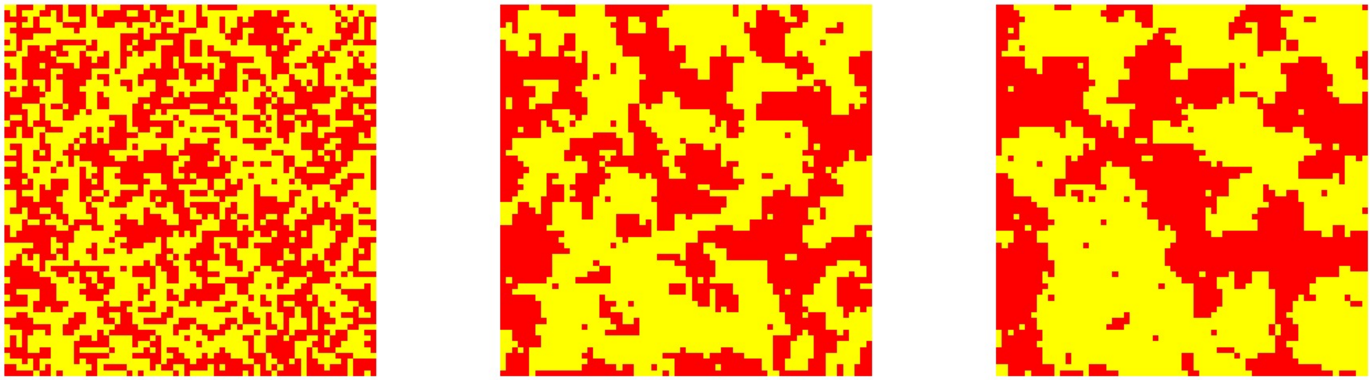
X-ray Photon Correlation Spectroscopy



Speckle Reveals Equilibrium Dynamics

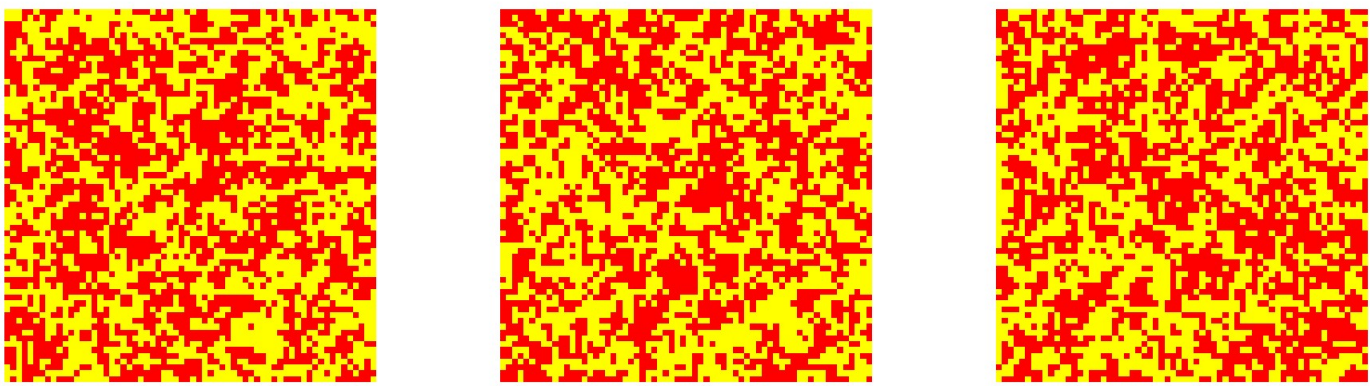
A. Non-equilibrium dynamics: average structure changes

time = 0 time = 1 time = 4



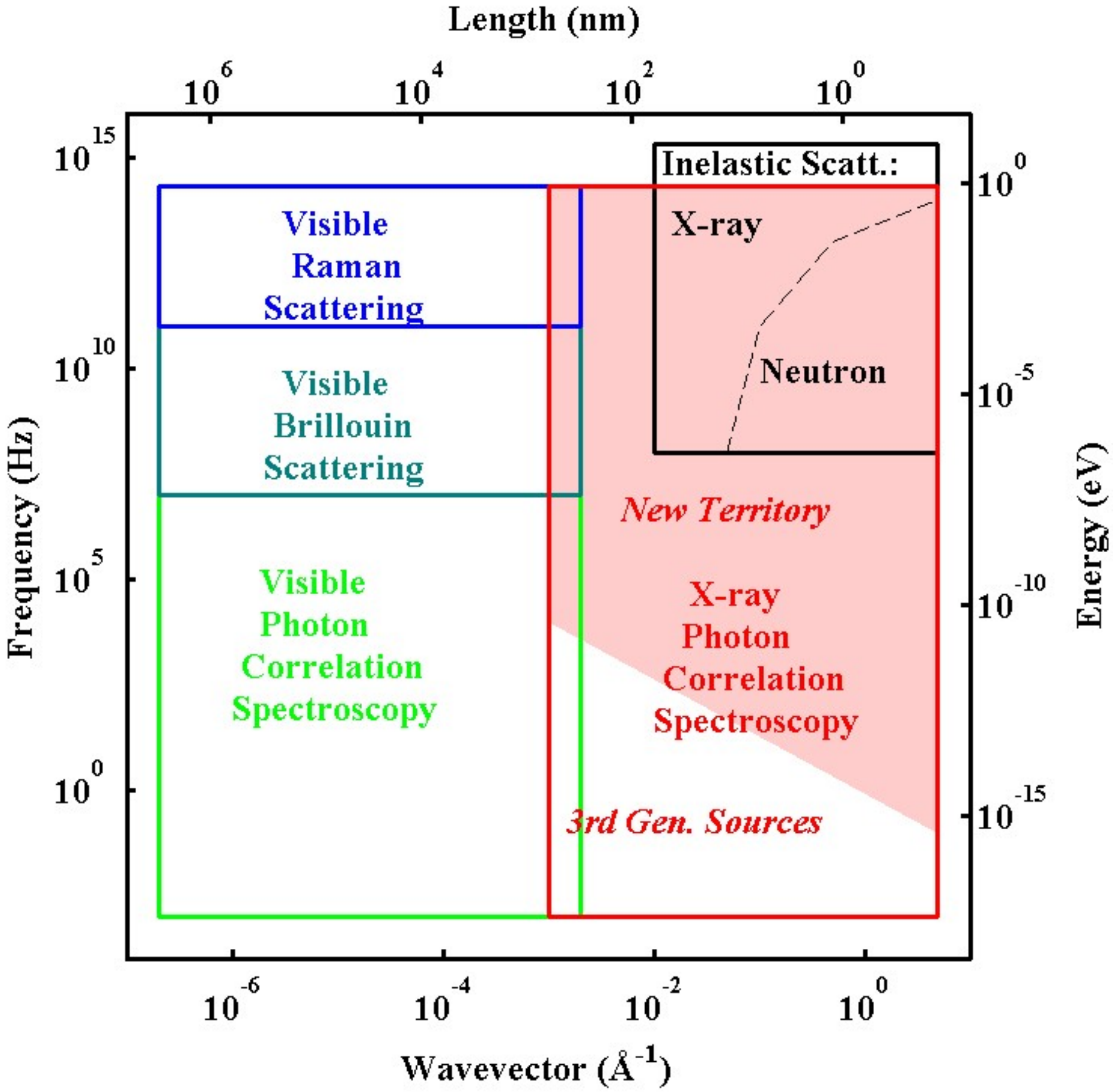
B. Equilibrium dynamics: average structure is static

time = 0 time = 1 time = 4



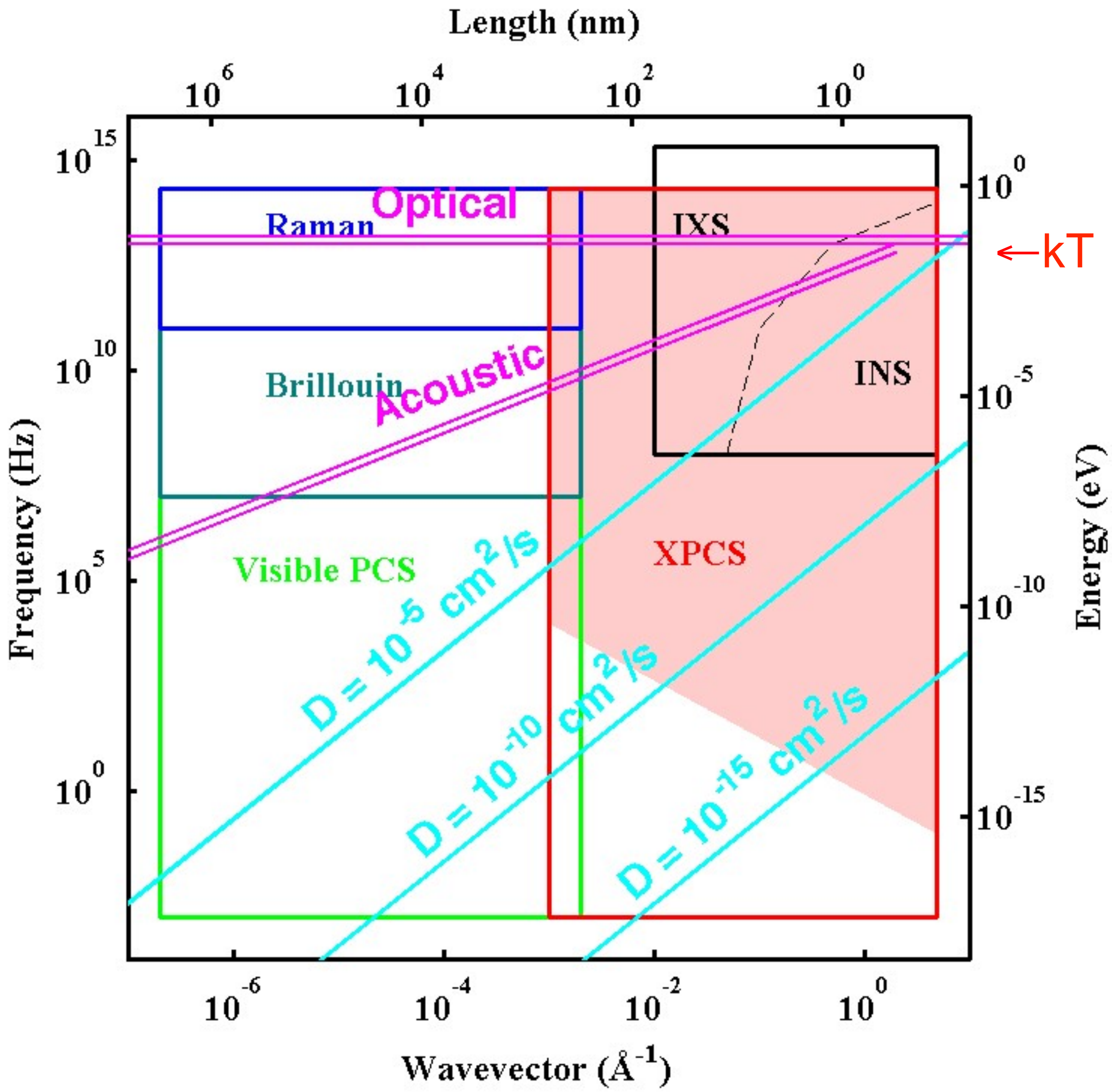
G.B. Stephenson, A. Robert, G. Grübel, Nature Mater. ***8****, 702 (2009)*

New Territory for XPCS: Large Q, Small τ



- To date, most XPCS experiments have been small-angle scattering in order to obtain sufficient signal
- Higher coherent flux will allow large-angle scattering studies of atomic scale dynamics, and studies at faster time scales

Large Q: Small Length Scales => Fast Time Scales



- Typical time scales of processes (e.g. mass diffusion) are faster at smaller length scales
- Mass diffusion time scales at molecular length scales are typically inaccessible by XPCS at third generation sources

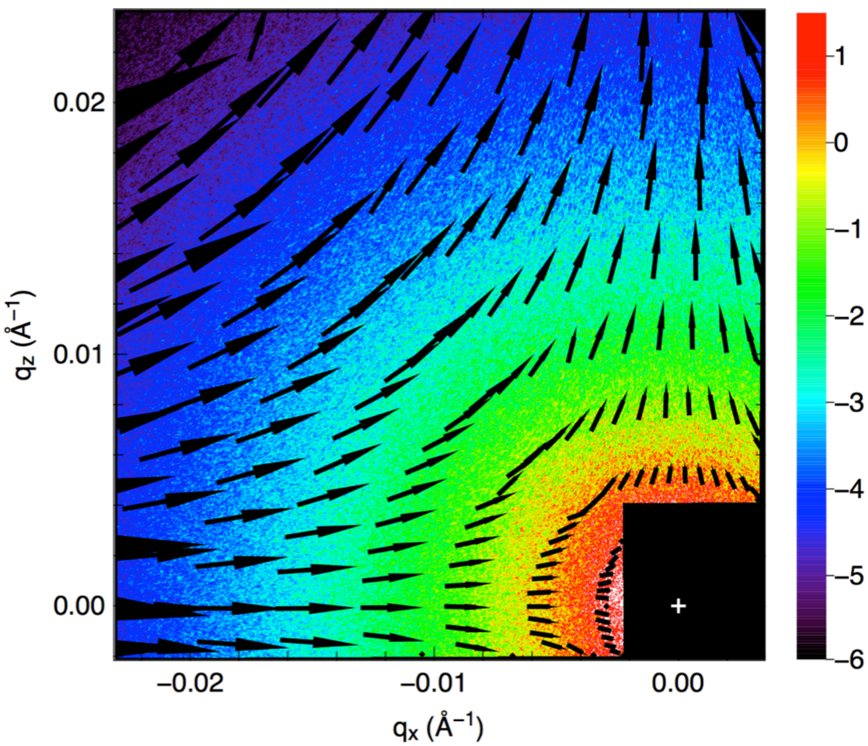
Going beyond the pair correlation function

Can we understand the time evolution of deformation and flow in condensed matter?

The APS with MBA lattice upgrade will allow:

- Molecular-scale motion in fluids and glasses mapped on sub-micron scale (e.g., transition to turbulent flow)
- 3-D strain tensor mapping in real-time of materials evolving under loading conditions
  - $\sim I(t,q)I(t+\tau, q+\delta q)$
- 10^2x-10^3x brightness yields 10^4 to 10^6 increase in dynamic range
- Nanosecond time scale, nanometer spatial resolution

Model system: spatiotemporal correlations in colloidal-decorated elastomer



Speckle shifts superimposed on scattering from a 20 micron region of a rubber sample undergoing flow in a stress-strain cell. Shifts are scaled by 200.

M. Sutton and J. L'Hermitte, unpub. (2014)

Summary

- High energy x-rays
  - Penetrate to enable in situ
  - Allow high q for atomic-scale resolution
- High transverse coherence enables:
  - Nanobeam focusing: high speed, could approach atomic scale resolution
  - Coherent diffractive imaging: sensitivity to surface chemistry, to single point defects
  - Correlation spectroscopy: nanosecond / nanometer resolution, higher order correlations