New Opportunities with Hard X-ray Diffraction Limited Sources

Qun Shen
Photon Sciences Directorate
Brookhaven National Laboratory

Workshop on Science at the Hard X-ray Diffraction Limit
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Outline

- Diffraction limit and coherence
- Coherent imaging and wave propagation
- Coherent diffraction – contrast and scaling law
- Potential high-impact applications
- Summary

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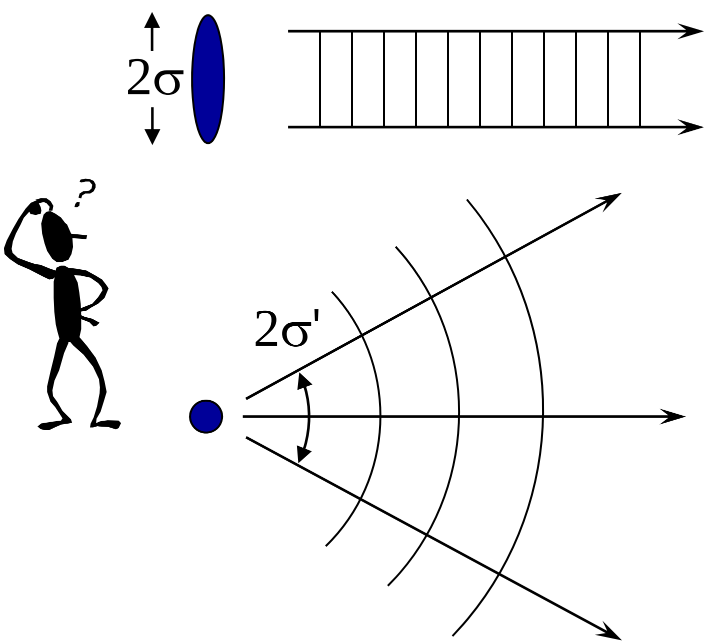
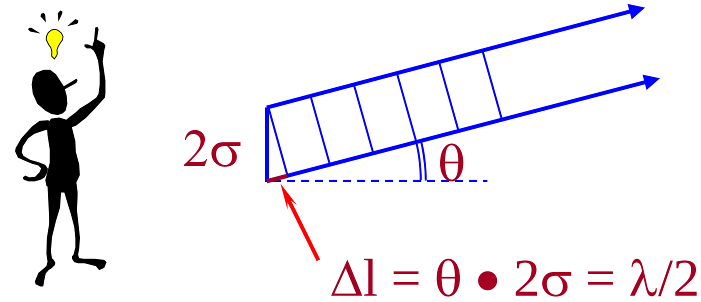
Spatial (Transverse) Coherence





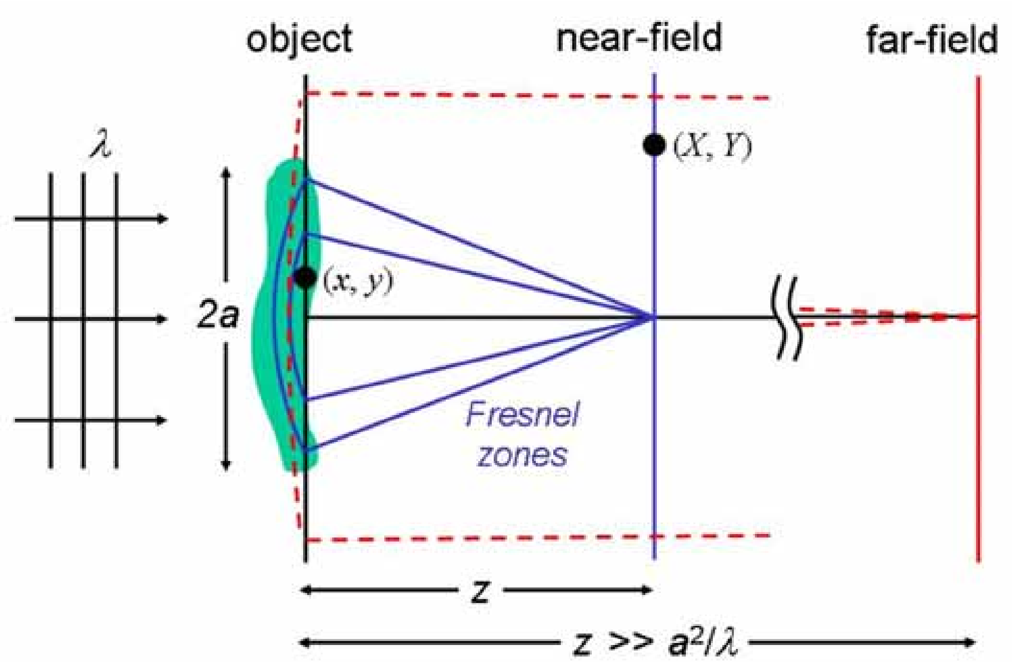
=> $2\theta \bullet 2\sigma \sim \lambda$

$\theta \rightarrow \sigma'$

=> X-ray beam is <u>spatially coherent</u> if phase-space area $2\pi\sigma'\sigma < \lambda/2$

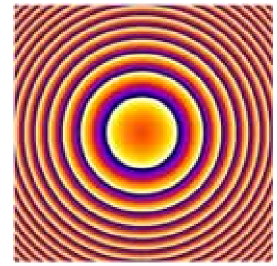
<u>Diffraction limited</u> source: $2\pi\sigma'\sigma = \lambda/2$ or $\varepsilon = \lambda/4\pi$

Coherent Wave Propagation



Phase-embedded object:

$$\bar{u}(x,y) \equiv u(x,y) e^{-\frac{i\pi}{\lambda z}\left(x^2+y^2\right)}$$



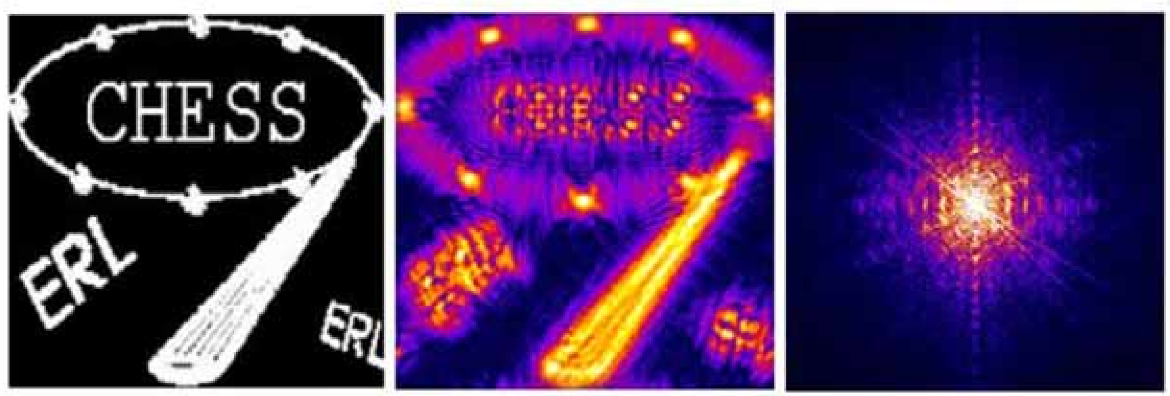
$$F(X,Y) = \frac{i\, e^{-ikR}}{\lambda R} \iint \bar{u}(x,y)\, e^{-\frac{ik}{z}(Xx+Yy)} dxdy$$

- Momentum transfer: $(Q_x, Q_y) = (kX/z, kY/z)$
- Number of Fresnel zones: $N_z = a^2/(\lambda z)$

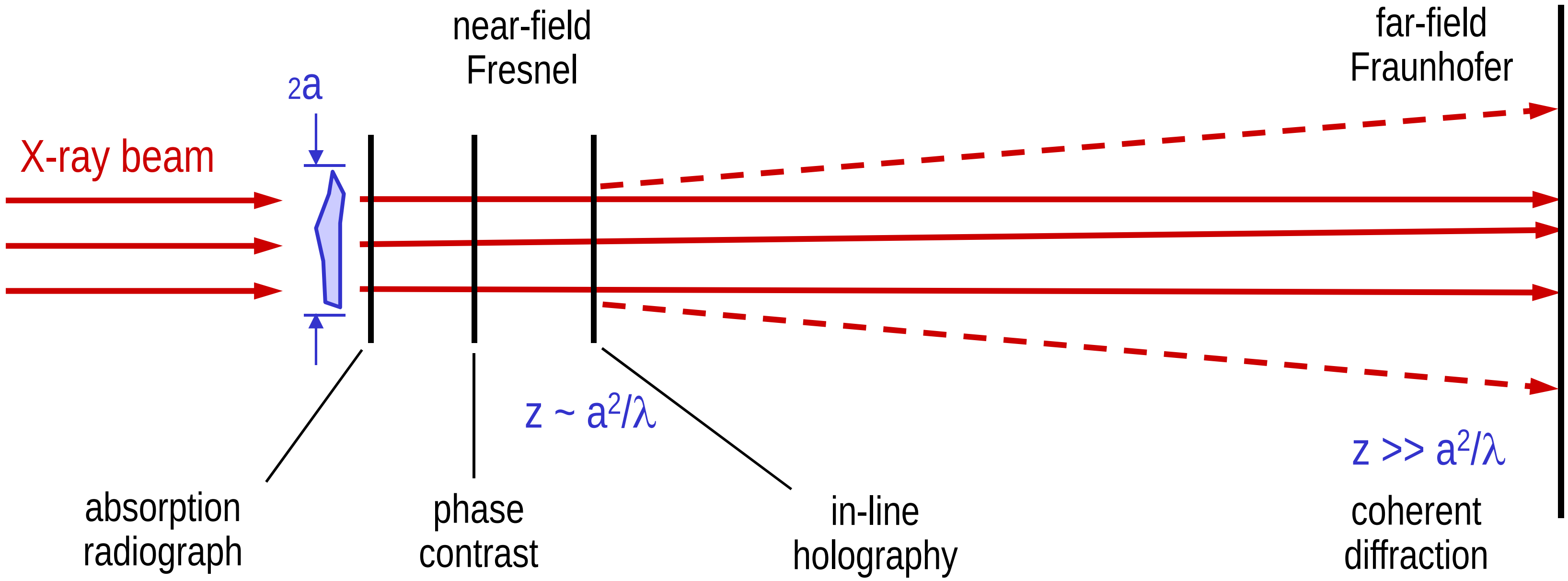
⇒ Unified wave propagation method by Fourier transform

⇒ Unified iterative phasing algorithm development

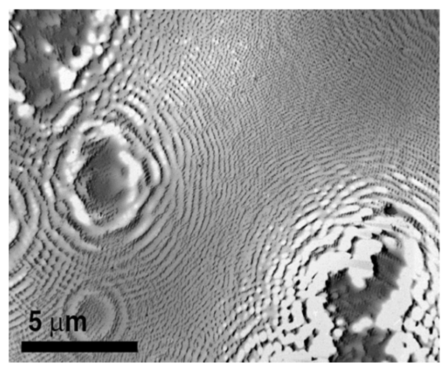
Xiao & Shen, PRB 72, 033103 (2005)



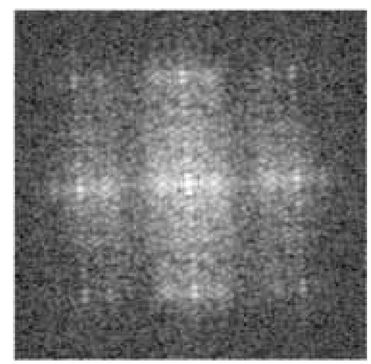
Different Regimes of X-ray Imaging



Kagoshima et al.
JJAP (1999).



Jacobsen (2003).

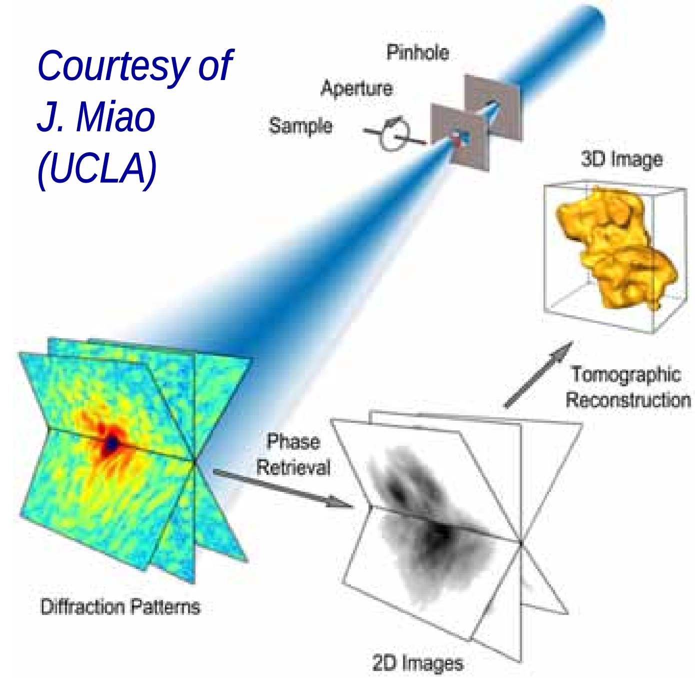


Miao et al.
Nature (1999).

Coherent X-ray Diffraction Imaging / Microscopy

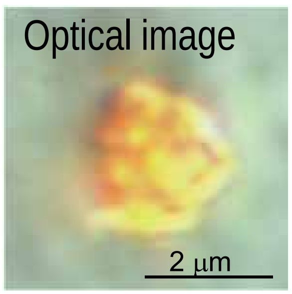
- Coherent diffraction microscopy is much like crystallography but applied to noncrystalline materials
- Requires a highly coherent x-ray beam (available at diffraction-limited source) and iterative phase retrieval

Courtesy of J. Miao (UCLA)

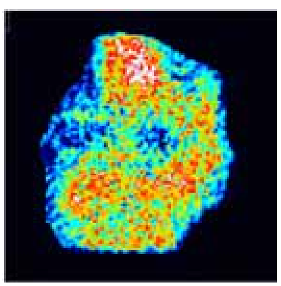


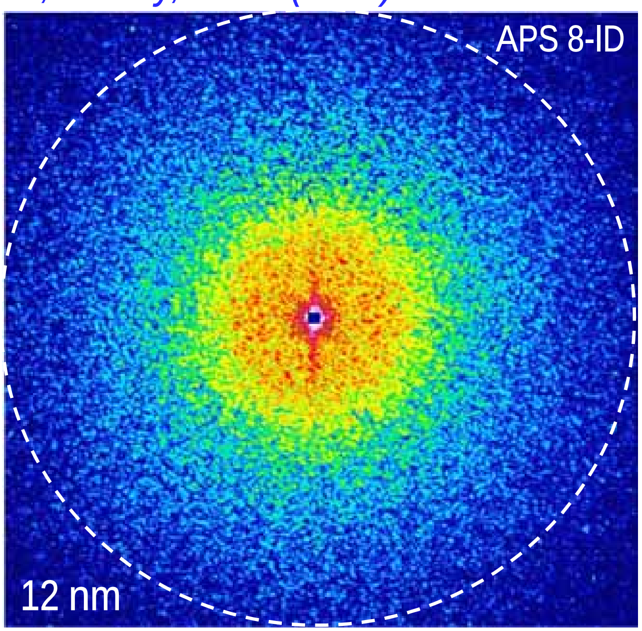
Example: structure of nanoporous gold

Xiao, Shen, Sandy, et al. (APS)

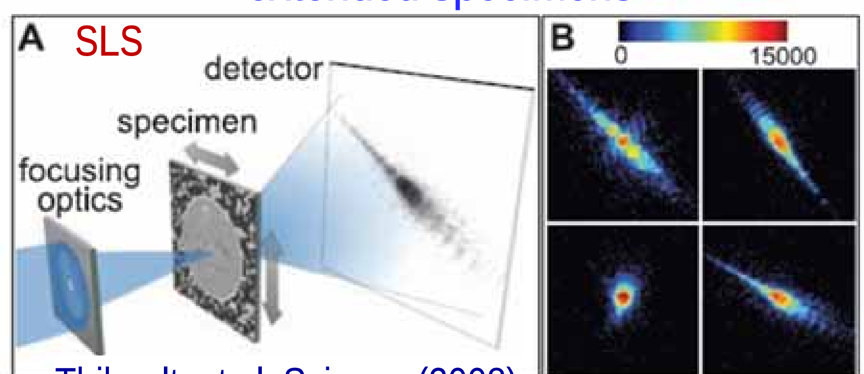


Reconstructed image





CDI combined with STXM: allows study of extended specimens



Thibault, et al, Science (2008)

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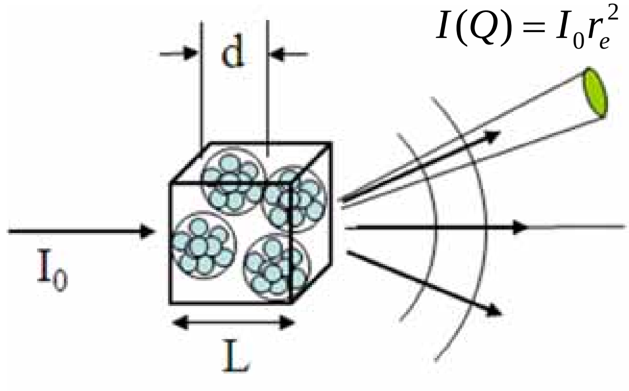
Scaling Laws in Coherent Diffraction Imaging

$$I(Q) = I_0 r_e^2 \cdot N \left| S(Q) \right|^2 \cdot \Delta\Omega$$



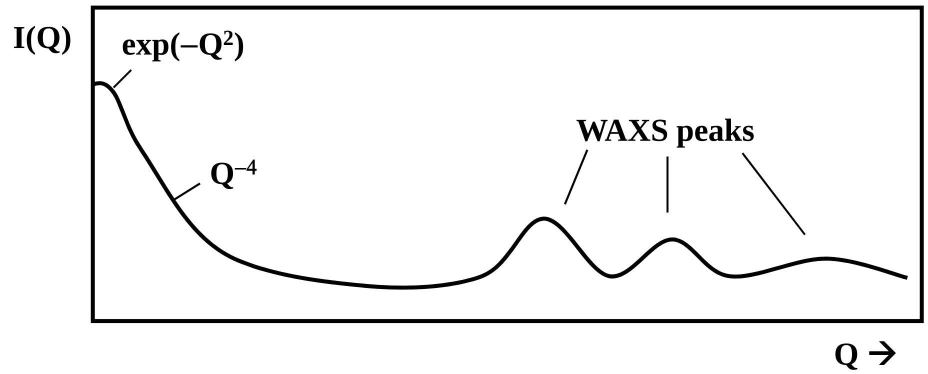
$$\Delta\Omega = \Delta(2\theta)\Delta\phi = \left(\frac{\lambda}{2\pi}\right)^2 \frac{\Delta Q_x \Delta Q_y}{\cos\theta} = \frac{\lambda^2}{2L^2\cos\theta}$$

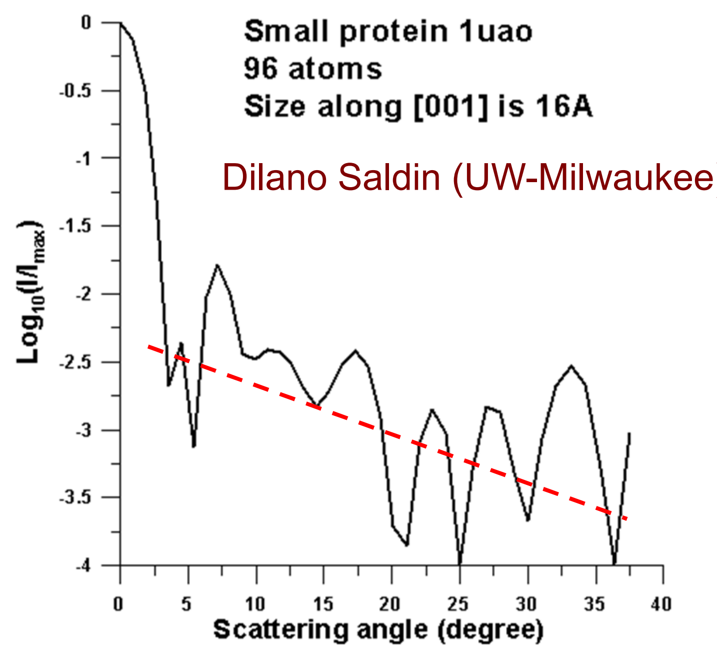
$$I \sim I_0 \cdot t\, \lambda^2 d^3$$

Shen et al. JSR 11, 432 (2004)

Marchesini, Howells, et al.
Optics Express (2003)

$$I \sim d^4 \lambda^2$$





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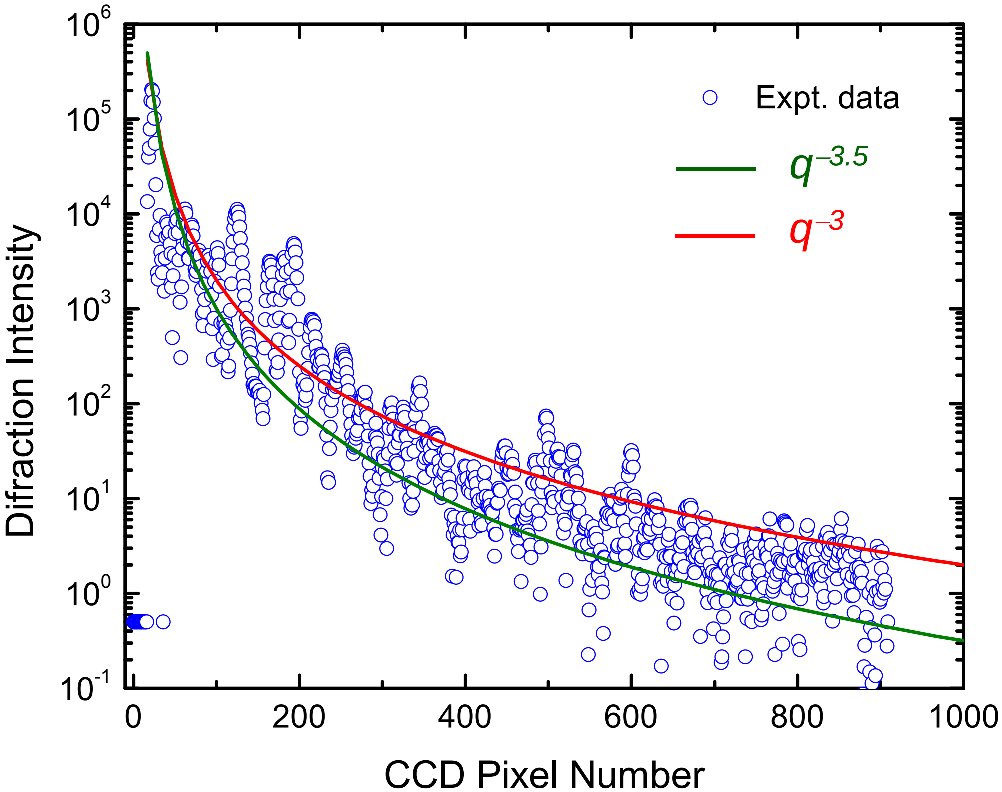
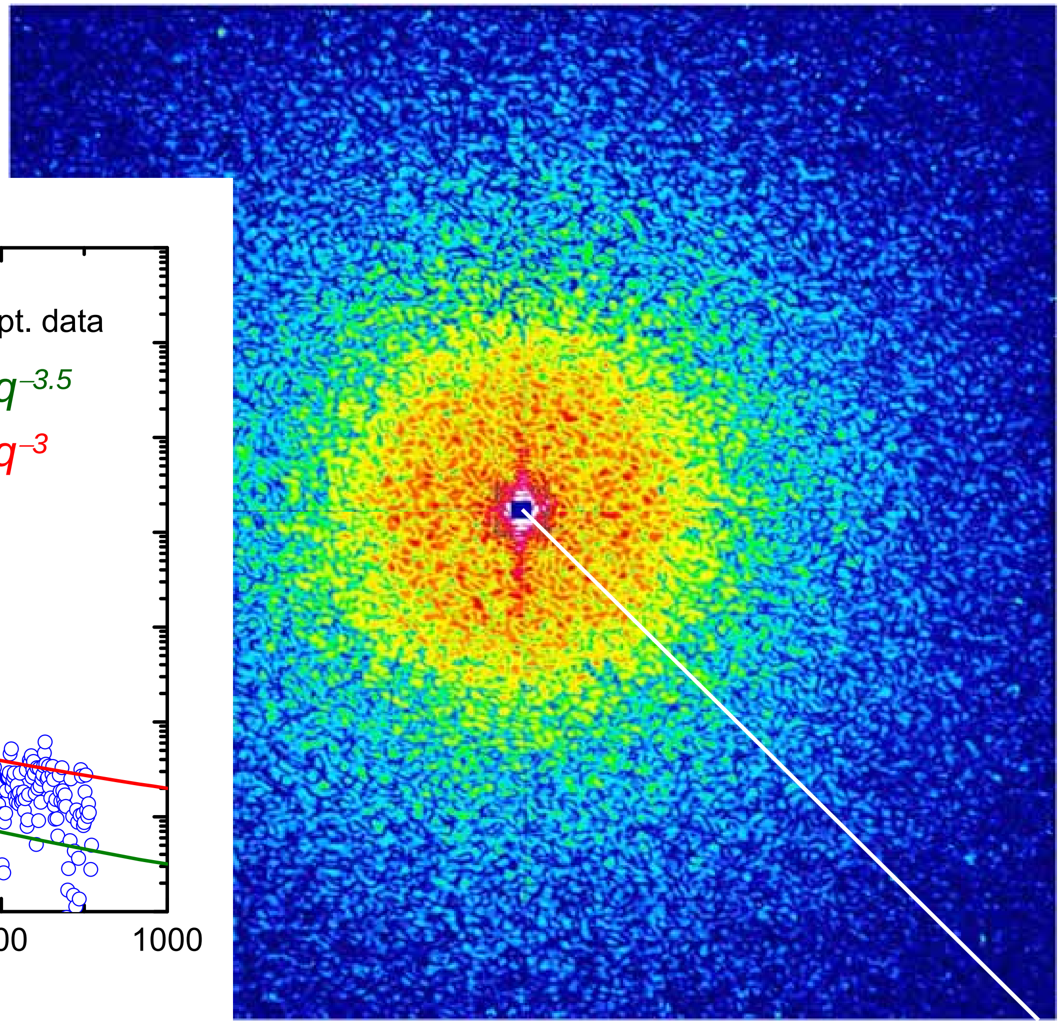
Q-dependence of Coherently Scattered X-Rays

Experiment results:

Gold nanofoam specimen
CDI data to ~ 8 nm





➔ Scaling law depends on specimen heterogeneity length scale !

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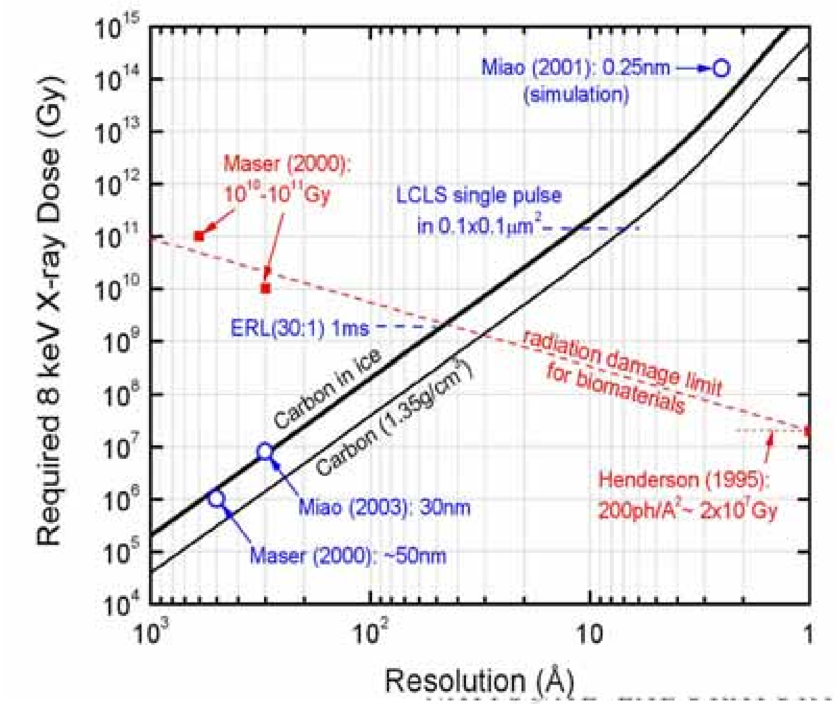
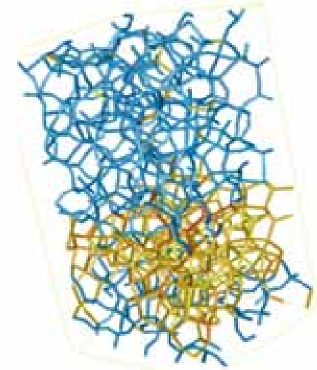
Contrast and Resolution Limit in Coherent Diffraction Microscopy

- Coherent diffraction imaging has the potential to advance structural science to noncrystalline specimens
- In principle, achievable resolution is limited only by x-ray wavelength; However, additional factors apply:
  - Contrast depends on the length scales at which inhomogeneities exist and is momentum-transfer Q dependent, and therefore highest achievable resolution may vary from system to system;
  - No single universal scaling law can be applied to all systems;
  - For radiation sensitive specimens (e.g. biological specimens), radiation damage limits achievable high resolution.
- In all cases, hard x-ray diffraction limited sources will play critical roles in making high-resolution coherent diffraction microscopy feasible and practical.

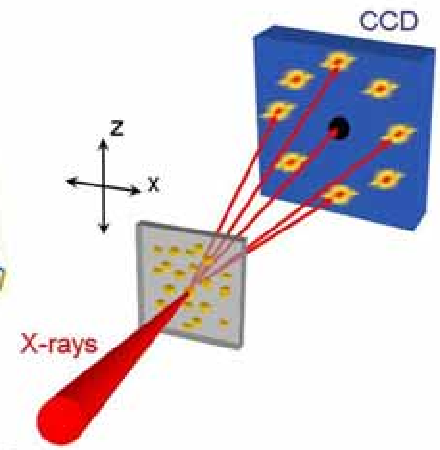


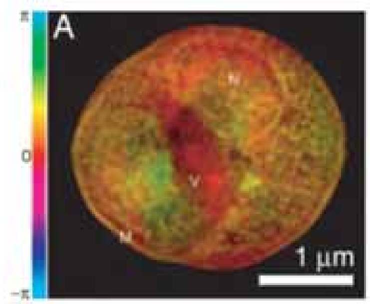
Emerging Potential High Impact Science Areas

- High-resolution Atomic-scale Structures:
  - Atomic-resolution structures of amorphous solids
  - Few-unit-cell nanocrystals of biomolecules
  - 2D crystals of membrane proteins
- High-resolution Nanoscale Structures:
  - Subcellular structures in biological cells
  - Group of biological cells during development
  - Biological bones and tissues
- Model-free Structures of Order/Disorder:
  - Static disorder and dislocations
  - Strain fields
  - Charge/orbital/spin ordering
- Speckle imaging:
  - Information beyond spatial resolution of the optic
  - Spatially resolved speckle statistical averaging

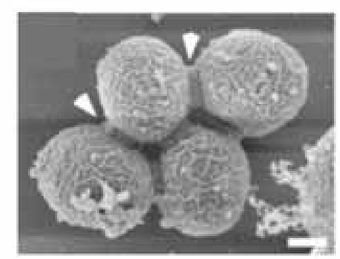


Nature 418, 62 (2002)

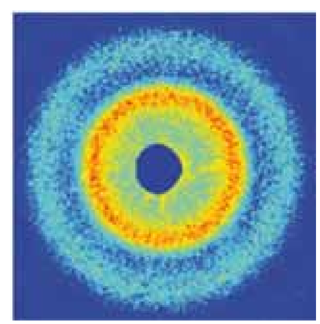




Shapiro et al. PNAS (2005)

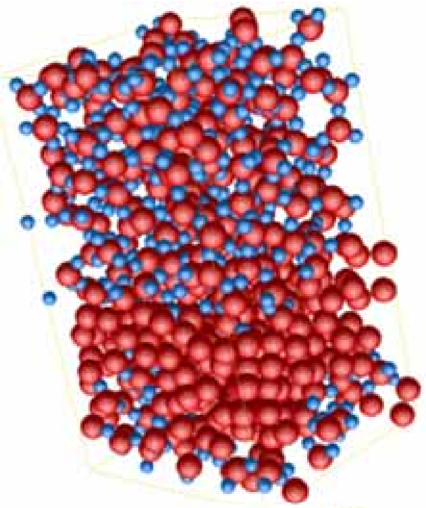


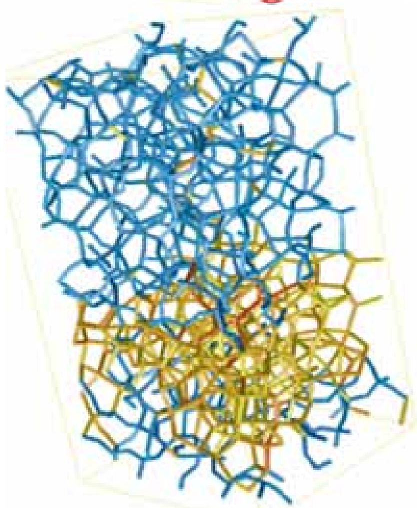
Spores



Wochner et al. PNAS (2009)

Coherent Diffraction Imaging of Amorphous Structures

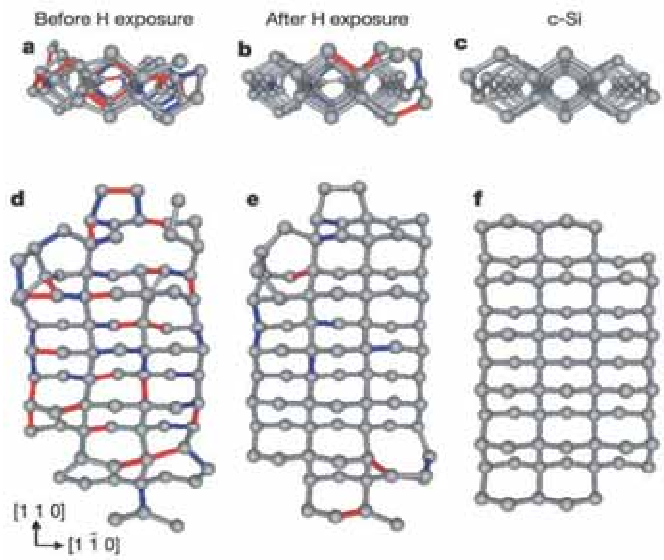




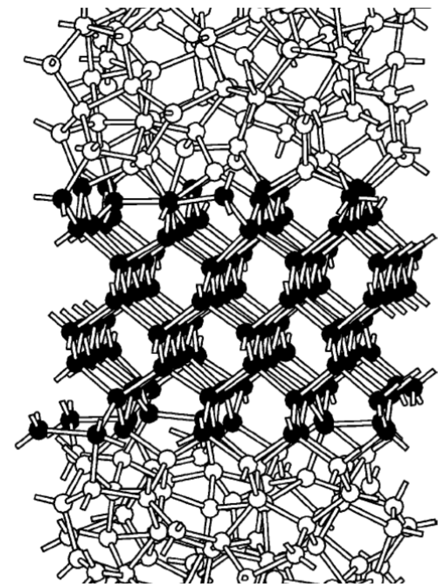
Nature 418, 62 (2002)

Amorphous Silicon (α-Si): *Veit Elser (Cornell)*

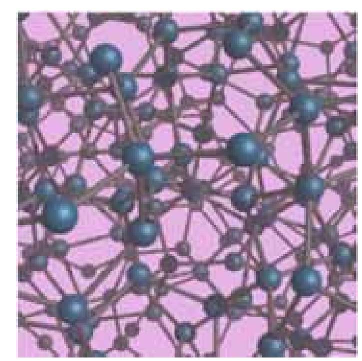
- atomic resolution structure data still lacking
- one of grand challenges in solid state physics



- High-resolution Amorphous Structures: Coherent diffraction microscopy could become a unique in-situ experimental technique to provide direct atomic-resolution structures for amorphous solids which are generally radiation resistant



PRB 58, 4579 (1998)



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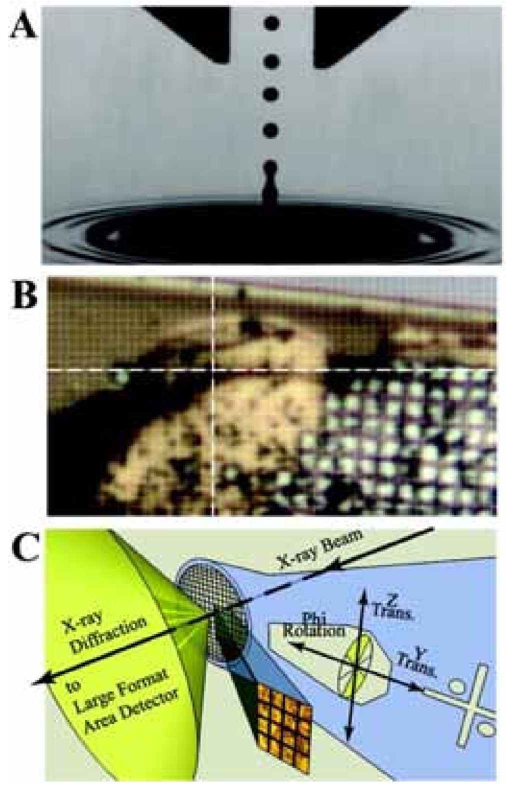
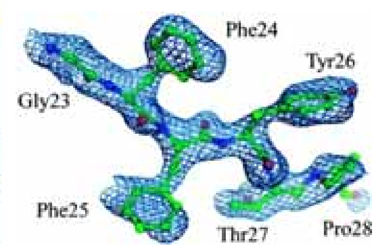
Structural Biology w/ Few Unit-Cell Nanocrystals

- Ability to obtain suitable crystals is a bottle neck in structural biology today
- With new experimental capabilities developed to handle small nanocrystals, perhaps only a few unit-cell in size, coherent diffraction microscopy could become a viable method to dramatically increase our structural knowledge of biological macromolecules
- Recently developed Acoustic Drop Ejection technique by Soares et al. (BNL) enables automatic mounting of microcrystallites onto micromesh by acoustic ejection of nL-sized droplets onto micromesh
- Successful structure determinations have been performed under cryo-protection
- One may expect that this type of technology may be extended to nanocrystals and to coherent diffraction microscopy from these nanocrystals

A.S. Soares, et al, "Acoustically Mounted Microcrystals Yield High-Resolution X-ray Structures," Biochemistry, ***50*** *(21), 4399 (2011).*



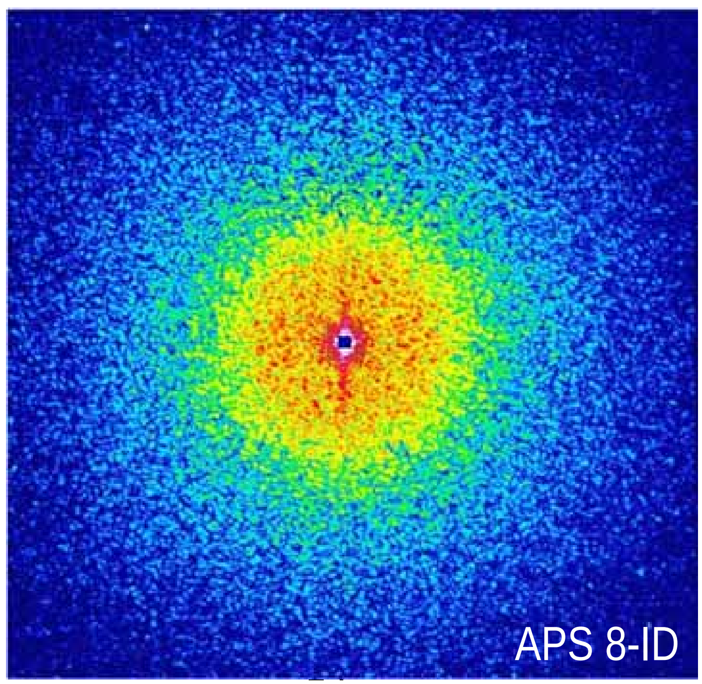
Left: (A) Stroboscopic photomicrograph of a single 2.5 nL water droplet launched via ADE from the liquid surface. (B) Image of the insulin microcrystal slurry supported on a micromesh used to determine the structure. The 20 μm × 20 μm x-ray beam is centered at the intersection of the white cross hairs. (C) Illustration of the concepts for the raster-scanning x-ray diffraction strategy with a microdiffractometer and a several-micrometer wide x-ray beam. Below: High-quality electron density maps obtained from ADE-mounted, serial microcrystallography.

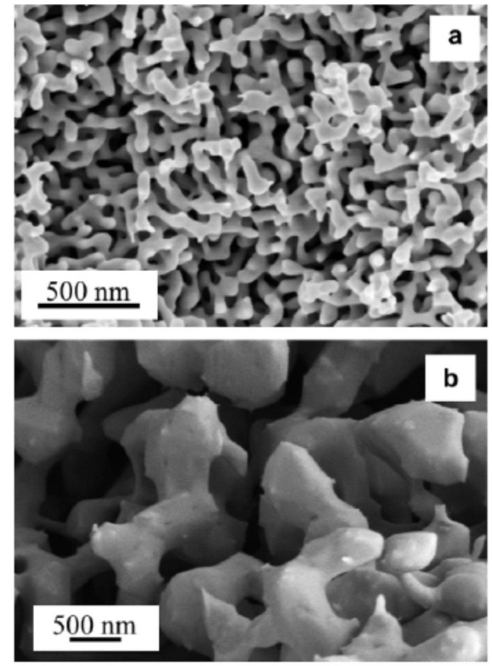


Work performed on NSLS beamline X25 and APS 23ID-D

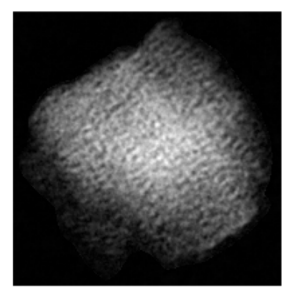
Nanoporous Materials

- Considerable interests in catalytic and sensor applications due to large surface-to-volume ratios
- Mechanical behavior of nanofoam materials have been a subject of intensive studies and discussion
- Metal nanofoams are potentially useful as voltage-tunable microelectronic devices, such as actuators, magnets, & resistors, because of their electronically tunable physical properties
- Coherent diffraction imaging at nm-scale resolutions on nanoporous materials will provide the necessary information to correlate structure to function of these materials
- Diffraction-limited x-ray source is essential for achieving ~nm scale





Xiao, Shen, et al. unpublished



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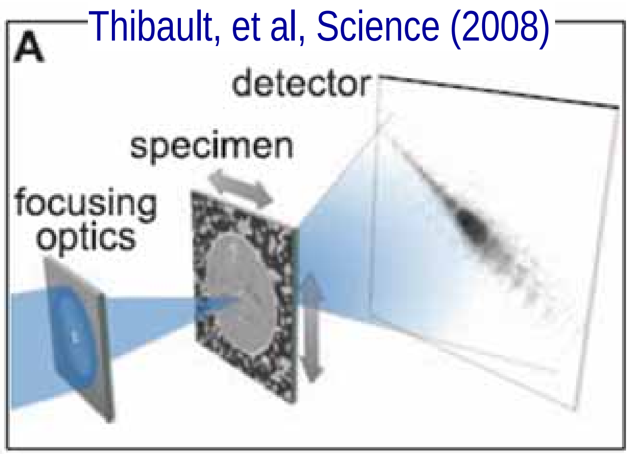
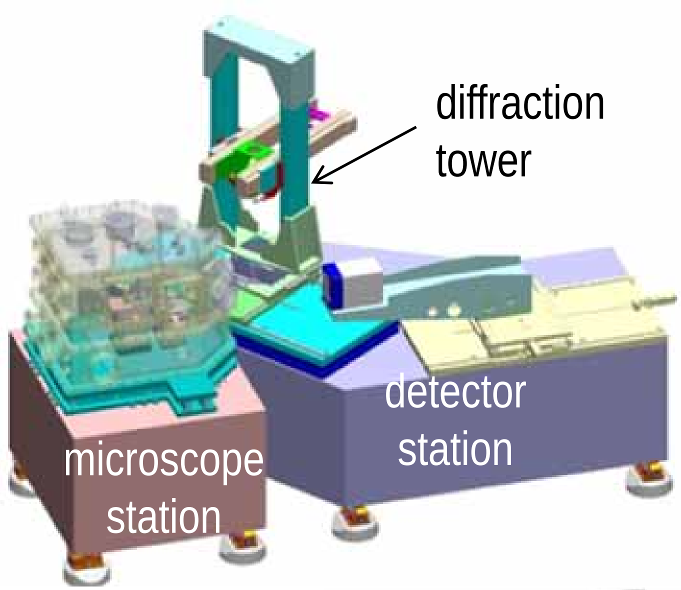
Integrated CDI-Ptychography-SXM

- Diffraction limited source would essentially transform any scanning x-ray microscope into a potential coherent ptychography instrument
- Such integrated coherent SXM would allow
  - Structural information beyond the spatial resolution set by focusing optics
  - Integrated ptychography and multi-modal SXM capabilities, including statistical speckle metrology contrast mechanism
- Such system would be ideal to study all types of heterogeneous materials
- NSLS-II: both HXN and SRX beamlines have been designed to accommodate these potential capabilities





NSLS-II HXN
Y. Chu et al. (BNL)

Summary

- Coherent diffraction imaging has the potential to advance structural science to noncrystalline specimens
- High-impact applications can be categorized according to density inhomogeneity length scales:
  - High-resolution atomic-scale structures
  - High-resolution nanoscale structures
  - Model-free structures of order/disorder
  - Speckle imaging
- Future diffraction-limited x-ray sources such as ERL or USR will play critical role in enabling high-resolution coherent diffraction microscopy in high-impact applications