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Can Coherent X-ray Diffraction Imaging Reach Atomic Resolution?

Qun Shen
X-ray Science Division
Argonne National Laboratory

Outline

- *Yes*
- *No*
- *Maybe so …*

- *Signal vs. resolution*
- *Radiation damage*
- *Optimized experiments*

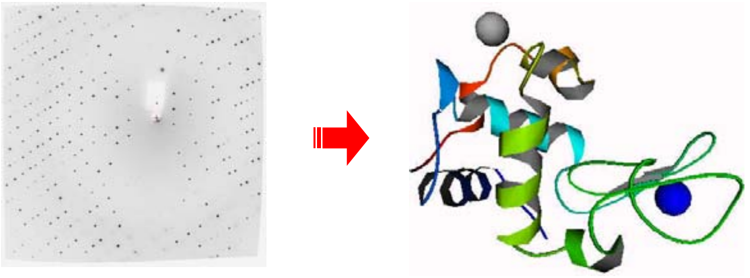
Source like NSLS-II will be critical !

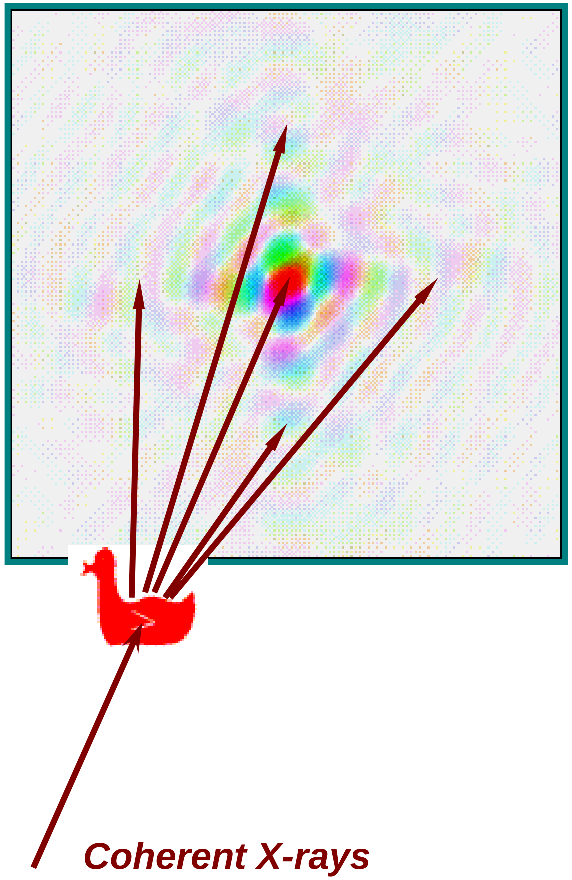
Coherent X-ray Diffraction Workshop, NSLS-II, March 14, 2008

Coherent X-ray Diffraction Imaging (CDI)

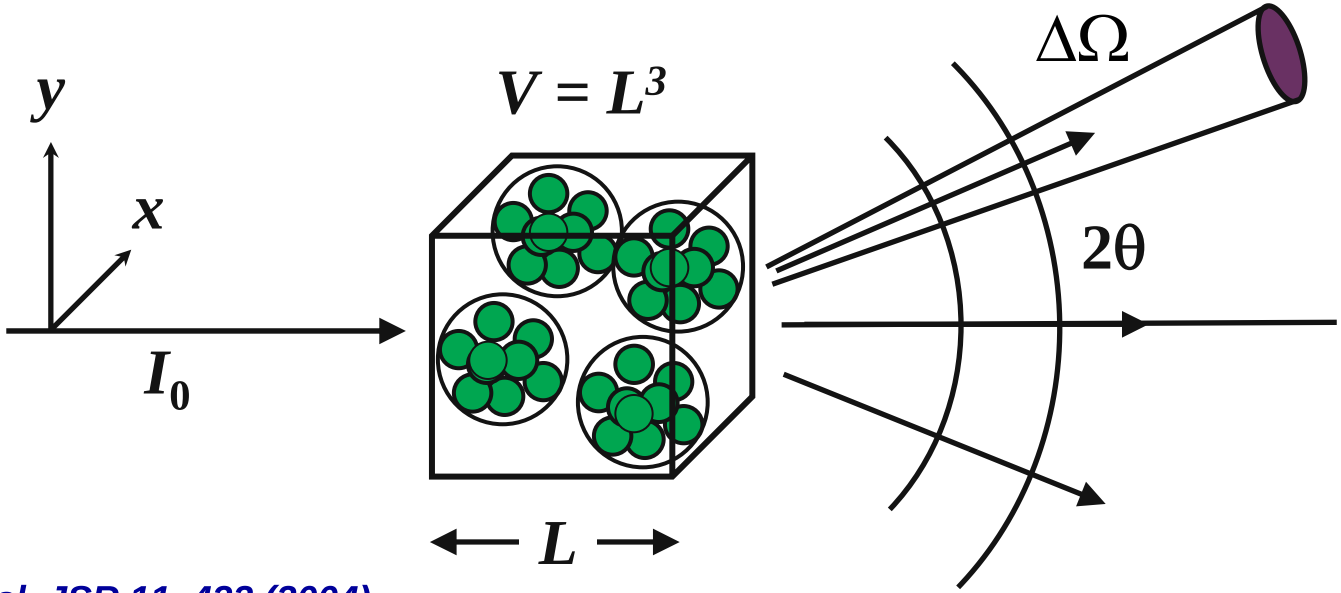
- ➔ Coherent diffraction imaging or microscopy is much like crystallography but applied to noncrystalline materials
- ➔ First proposed by David Sayre in 1980, and first experimental demonstration in 1999 using soft x-rays [Miao, Charalambous, Kirz, Sayre (1999) Nature 400, 342–344]
- ➔ Requires a fully coherent x-ray beam and iterative phase retrieval

Analogous to crystallography





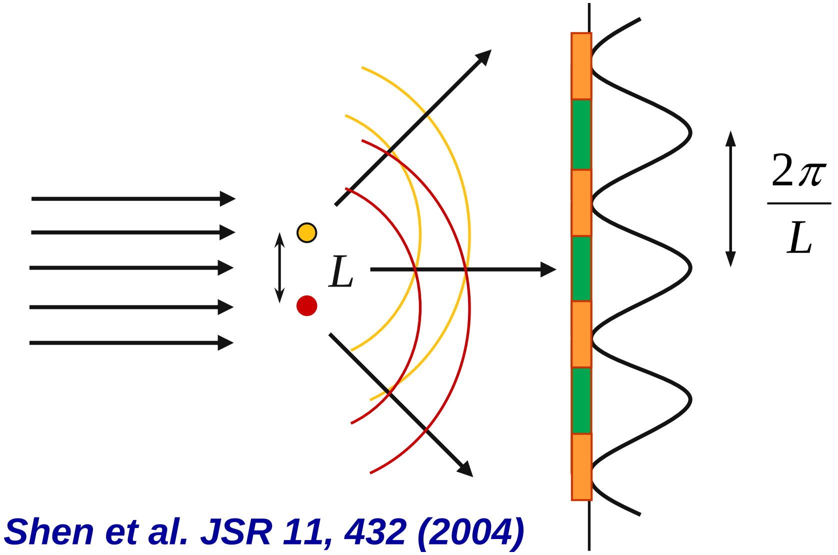
Signal Strength in Coherent Diffraction



Shen et al. JSR 11, 432 (2004)

- Required number of photons (dose) needed incident on sample in order to achieve a given resolution *d* ?
- Does required dose exceed radiation damage limit for that resolution ?
- Is there any way to deal with damage problem ?

Oversampling in Coherent Diffraction Imaging



Shen et al. JSR 11, 432 (2004)

=> *Sampling at frequency $2\pi/L$ in Fourier space is not fine enough to resolve interference fringes!*

=> *Additional measurements in-between $2\pi/L$ are necessary to tell us some interference is going on.*

=> *Minimum oversampling ratio is 2, regardless whether it is 1D, 2D or 3D.*

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

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

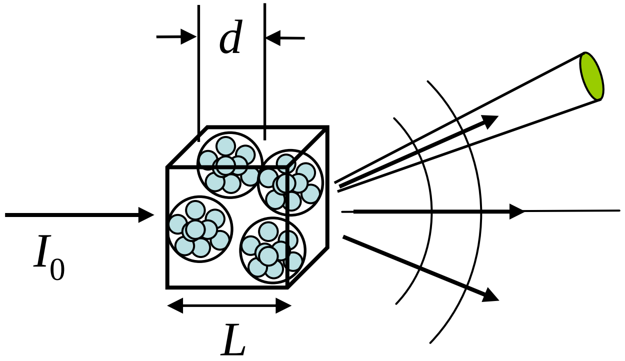
$$\boxed{I = I_0 r_e^2 L \cdot \frac{n_0^2 f^2 d^3 \lambda^2}{4\pi^2}}$$

$$\Delta Q_{\max}^{1D} = \frac{2\pi}{L} \cdot \frac{1}{2} = \frac{\pi}{L}$$

$$\Delta Q_{\max}^{2D} = \frac{2\pi}{L} \cdot \frac{1}{\sqrt{2}} = \frac{\sqrt{2}\pi}{L}$$

$$\Delta Q_{\max}^{3D} = \frac{2\pi}{L} \cdot \frac{1}{\sqrt[3]{2}}$$

An Example: Gold Particles



NSLS-II: doubly focused U20 undulator beam

Coherent flux: $I_0 = 1.4\text{x}10^{13}$ phs/s/μm^2, $\lambda = 1.7$ A

Gold particles: $d = 1$ nm, $f \sim Z = 79$, $L = 1$ μm

Atom density $n_0 = \rho N_0/A$, $\rho = 19.3$ g/cm^3,

$N_0 = 6.022\text{x}10^{23}$/mol, $A = 197$ g/mol

Classical radius of electron: $r_e = 2.82\text{x}10^{-13}$cm

$$\boxed{I = I_0 r_e^2 L \cdot \frac{n_0^2 d^3 f^2 \lambda^2}{4\pi^2}}$$

$$I = 1.4\times10^{13}\times10^{8}\times(2.82\times10^{-13})^2\times10^{-4}\cdot\frac{(19.3\times6.022\times10^{23}/197)^2\times(10^{-7})^3\times79^2(1.7\times10^{-8})^2}{4\pi^2}$$

$$\Rightarrow \boxed{I = 1.8\times10^{3}\ \ \text{cts/s/pixel}}$$ @ d = 1 nm

Scaling Laws in Coherent Diffraction Imaging

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

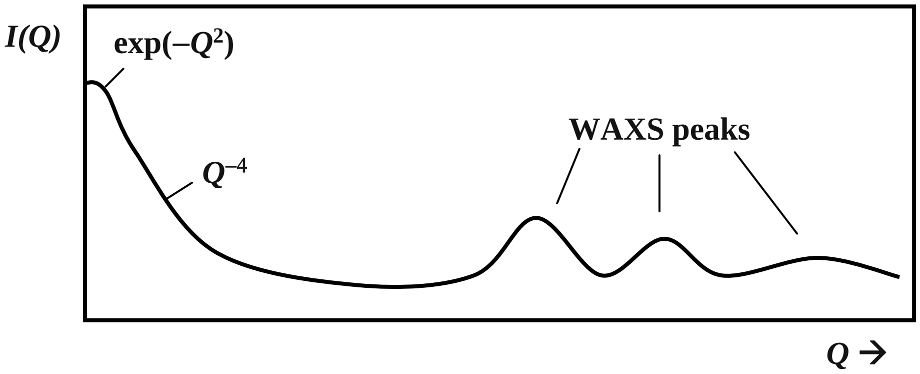
$$I(Q) = I_0 r_e^2 \cdot N\left|S(Q)\right|^2 \cdot \Delta\Omega$$

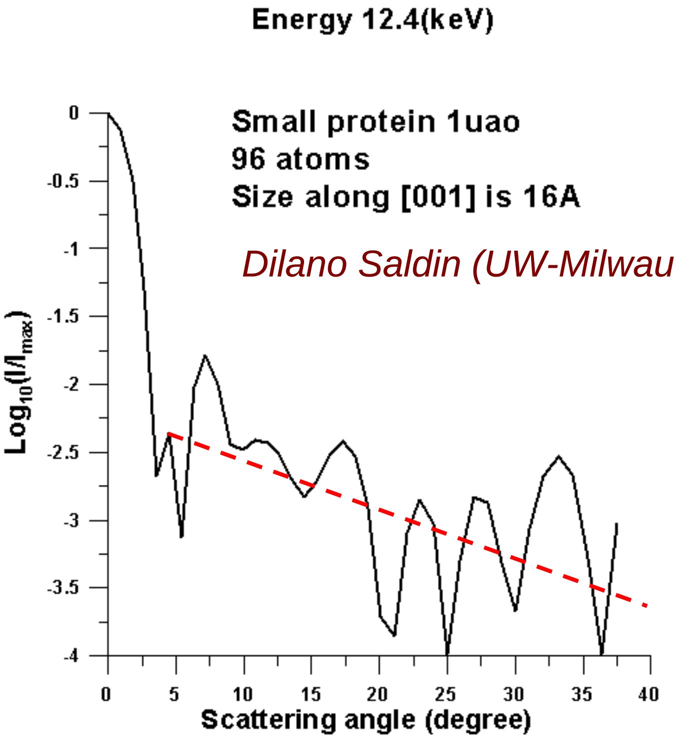
$$\boxed{I \sim I_0 \cdot t\,\lambda^2 d^3}$$

Shen et al. JSR 11, 432 (2004)

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

$$\boxed{I \sim d^4 \lambda^2}$$





Dilano Saldin (UW-Milwaukee)

Required Flux for Time-Integrated Signal $I\Delta t$

Consider fixed 3D volume case:

$$I = I_0 r_e^2 L \cdot \frac{n_0^2 d^3 f^2 \lambda^2}{4\pi^2}$$

➡ Accumulated incident flux:

$$I_0 \Delta t = I\Delta t \cdot \frac{4\pi^2}{r_e^2 L n_0^2 d^3 f^2 \lambda^2}$$

Let 's say $I\Delta t$ = 5 cts/pixel is the minimum time-integrated signal at resolution d, then required incident flux is given by

$$\boxed{I_0 \Delta t = \frac{20\pi^2}{r_e^2 L n_0^2 d^3 f^2 \lambda^2}}$$

<u>Example</u>: carbon (protein) @ d = 1 nm

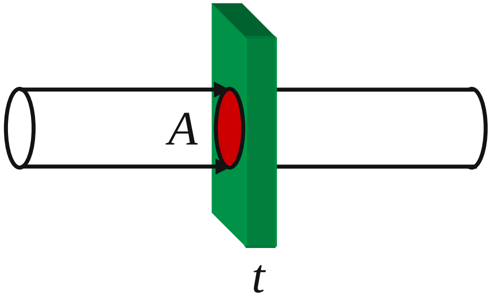
$r_e = 2.82\text{x}10^{-13}$cm, f ~ Z = 6.6, L = 0.1 μm

$n_0 = \rho N_0/A$, ρ =1.35 g/cm^3, A =13 g/mol, N_0=6.022x10^{23}/mol

$$I_0 \Delta t = \frac{20\pi^2}{(2.82\times10^{-13})^2 \times10^{-5} \times(1.35\times6.022\times10^{23}/13)^2 \times(10^{-7})^3 \times6.6^2 (1.5\times10^{-8})^2}$$

$$= 6.48\times10^{21}\ \text{phs/cm}^2 = 6.48\times10^{13}\ \text{phs}/\mu\text{m}^2$$

Converting Flux to Dose



Dose = Energy / Mass (Gy = J/kg)

$$Dose = \frac{(I_0\Delta t)\mu t \cdot E \cdot A}{\rho \cdot At} = \frac{(I_0\Delta t)\mu \cdot E}{\rho}$$

Absorbed photon intensity:

$$I_{abs} = I_0\left(1-e^{-\mu t}\right) \approx I_0\mu t$$

Absorbed power:

$$P_{abs} = I_{abs} \cdot E \cdot A = I_0\mu t \cdot E \cdot A$$

Mass in volume *At*: $M = \rho \cdot At$

Previous Example:

Carbon (protein) @ d = 1 nm

μ/ρ = 4.26 cm^2/g for C, ρ =1.35 g/cm^3,

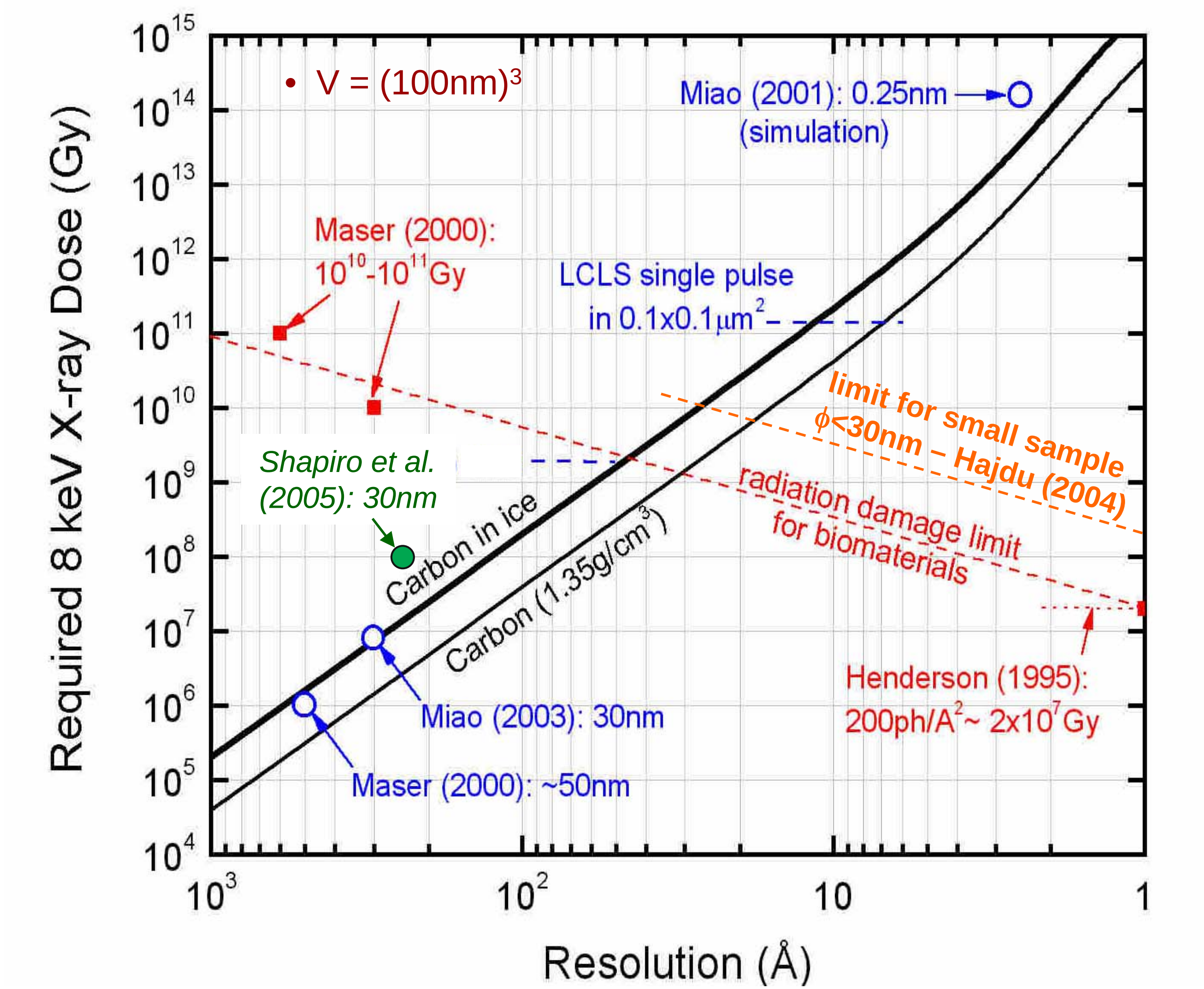
@ E = 8000 eV,

$$I_0\Delta t = 6.48\times10^{21} \text{ phs/cm}^2$$

$$Dose = 6.48\times10^{21}\times4.26\times8000\times1.6\times10^{-19}\times10^3$$

$$= 3.5\times10^{10}\ Gy$$

Radiation Dose vs. Resolution & Damage (biomaterials)



Shen et al. J. Synch. Rad. 11, 432 (2004)

See also: Marchesini et al. Opt. Express (2003)

To go beyond damage limit:

→ Femtosecond diffraction with XFEL pulses [Hajdu 2000]

→ Self-assembled macromolecule array

→ Continuous stream of molecules oriented by laser [Spence 2004]

→ Small specimen limits [Hajdu 2004]

Radiation Damage in Hard Non-biological Materials

Work by I.C. Noyan group at Columbia University (2008):

Si (008) on SiO_2 peak intensity decreased to 50% upon microbeam irradiation at APS 2-ID-D, with estimated dose of $2x10^{10}$ Gy.

No degradation was observed for SiGe thin-film on Si substrate.

=> Oxygen diffusion plays a crucial role in the degradation process.

=> Radiation damage is a complex problem and is highly sample dependent.

Argonne LDRD for Coherent Diffraction Imaging of Nonperiodic Materials

Subtask 1 – Optimization of coherent diffraction experiments

I. McNulty, Q. Shen, J. Maser, R. Harder

Research and develop optimized arrangements for nanoscale coherent diffraction experiments. The objective is to develop the methods and instrumentation necessary to design and build a future dedicated coherent diffraction user facility at APS.

Subtask 2 – Feasibility of single-particle structural imaging

P. Fuoss, L. Young, J. Maser, E.D. Isaacs

Focus on fundamental experimental and algorithmic issues in single particle imaging by coherent diffraction, including use of nanofocused x-ray beams, sorting of random particle orientations, imaging of laser-aligned nano-objects, radiation effects and resolution limits. The objective is to determine the feasibility of single-particle imaging at the upgraded APS, and to help develop optimized strategies at future coherent sources.

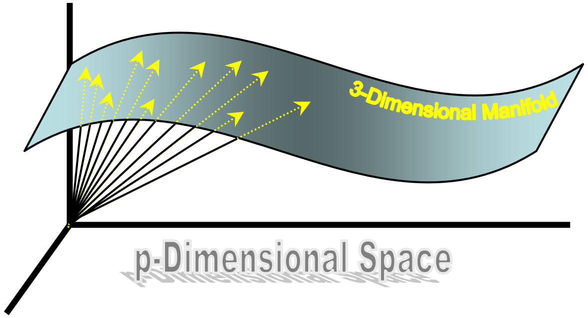
Subtask 3 – Novel applications with coherent diffraction

Q. Shen, I. McNulty, S. Streiffer, E.D. Isaacs

Perform feasibility coherent x-ray diffraction imaging experiments on selected specimens in materials science and biology. The objective is to demonstrate nm-scale resolution and high-impact applications in coherent diffraction and develop a strong user community for the coherent diffraction program at APS.

Key Collaborators/Users:

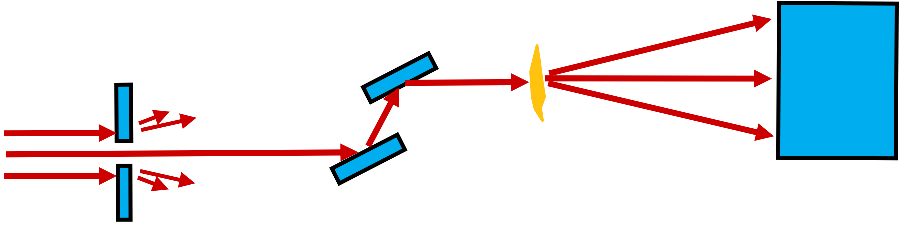
K.A. Nugent (Melbourne)
I.K. Robinson (UCL)
J. Miao (UCLA)
R. Harder (APS)
A. Ourmazd (Milwaukee)
D. Saldin (Milwaukee)
D. Dunand (Northwestern)
S. Vogt (APS)
B. Palmer (Vermont)



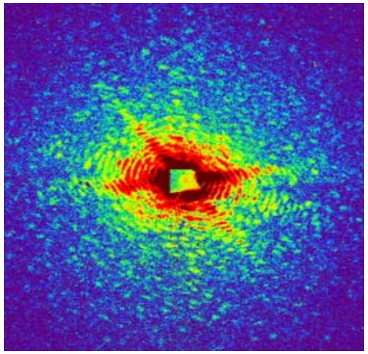
Advanced algorithms (A. Ourmazd)

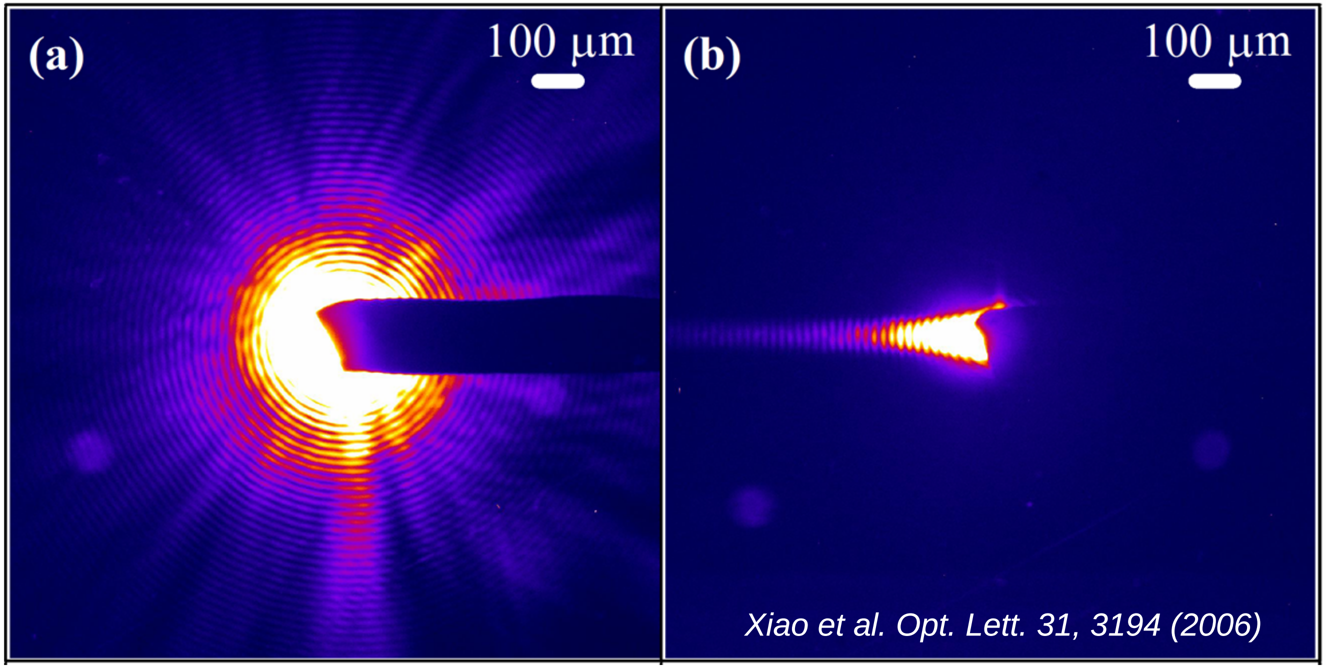
Improving S/N in CDI by Crystal Guard Aperture

- Crystal guard aperture for CDI:



Xiao, de Jonge, Chu, Shen (APS) Opt. Lett. (2006)

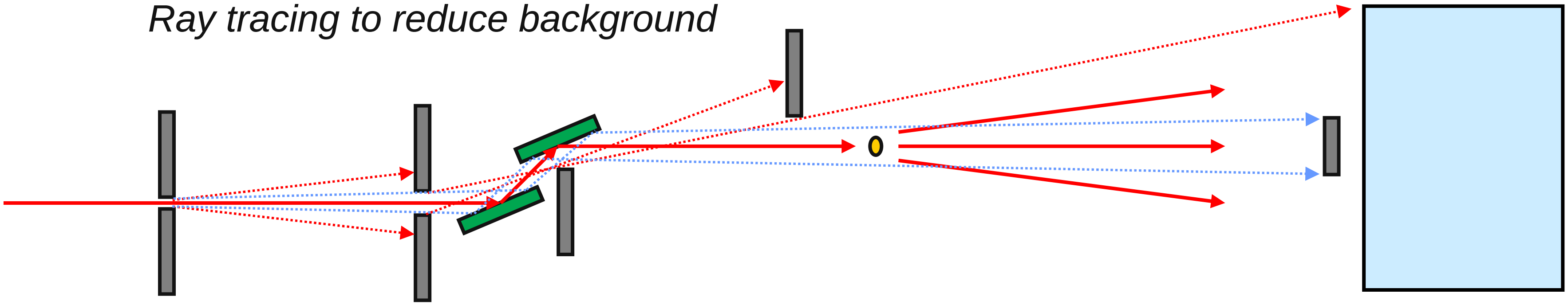




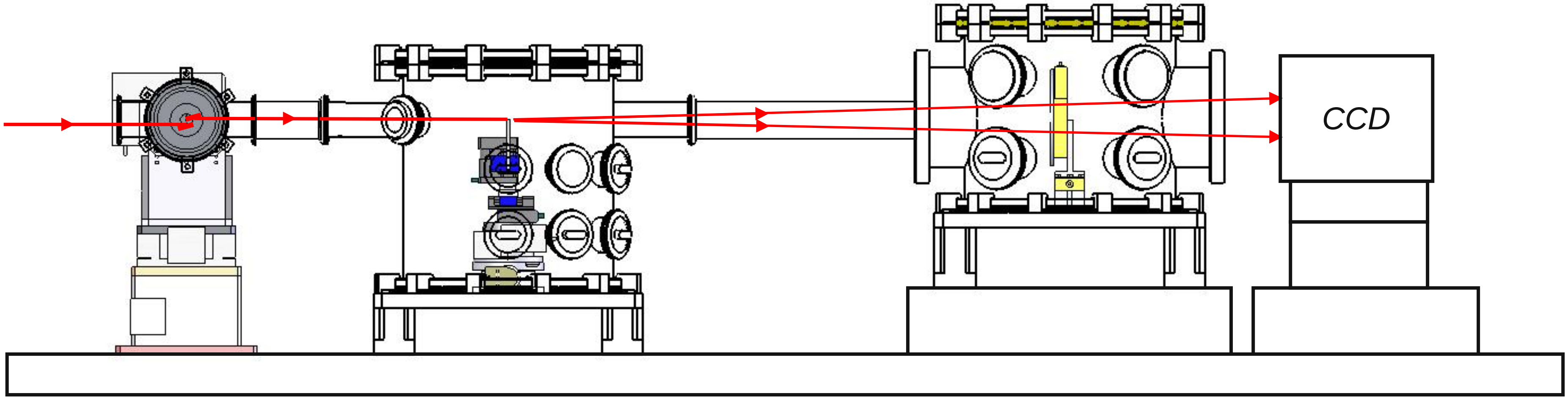
Xiao et al. Opt. Lett. 31, 3194 (2006)

Coherent Diffraction Imaging Experiment Setup

Ray tracing to reduce background

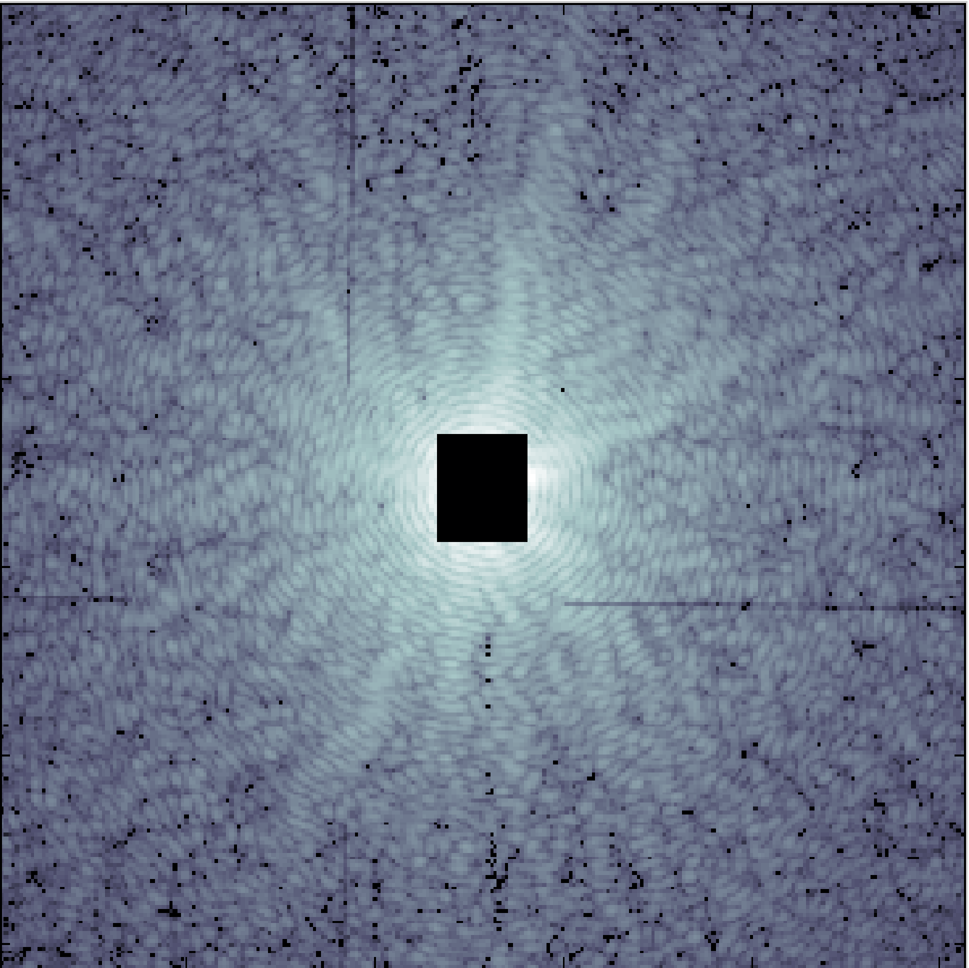


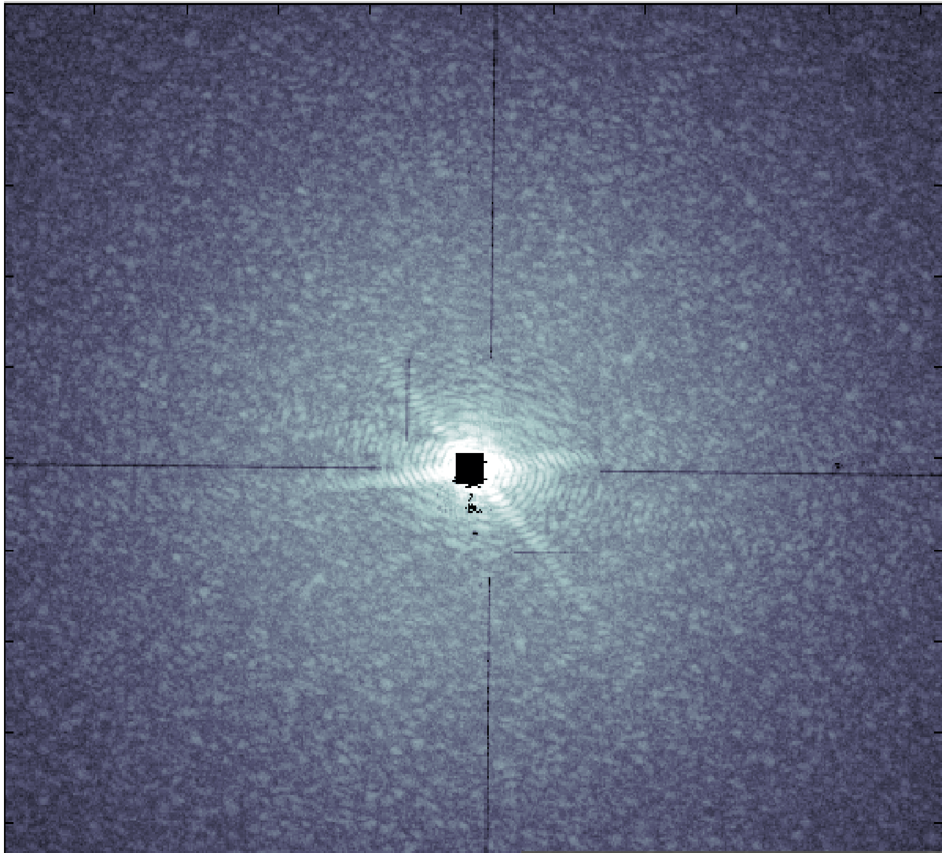
Engineering design by SoonHong Lee (AES-MED)



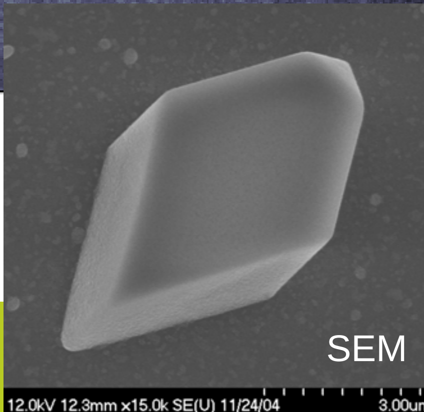
Experiment at 8-ID with Direct Detection CCD

X-ray energy: 7.35 keV
→ Highest angle signal ⇔ ~20nm resolution, limited by counting statistics and size of CCD
→ Parasitic scattering background < 0.1 ph/s



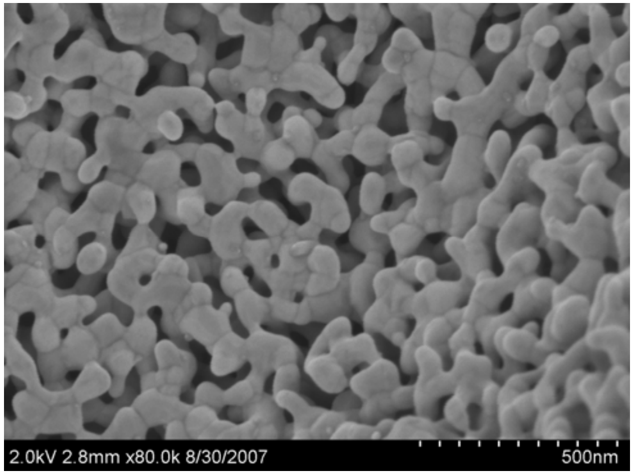


__Problem__: missing data behind beam-stop

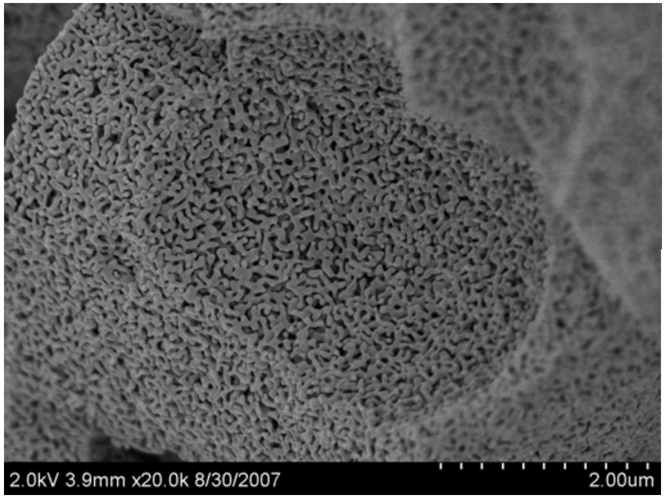


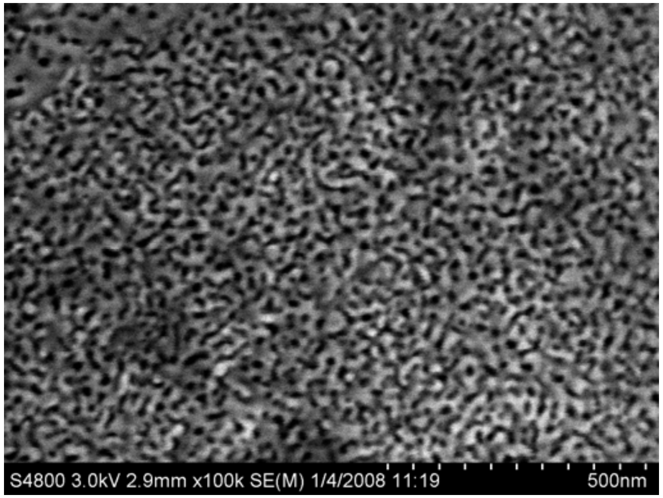
Application Example: Dealloyed Au Nanofoam

Chen, Cox, Dunand (NU)



Ag-Au foil, dealloyed 48 hours

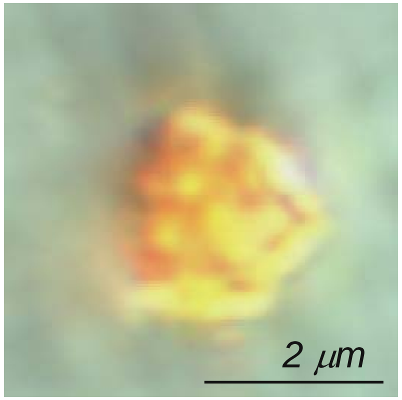


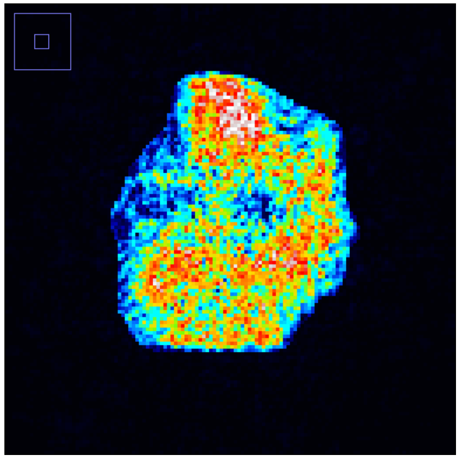


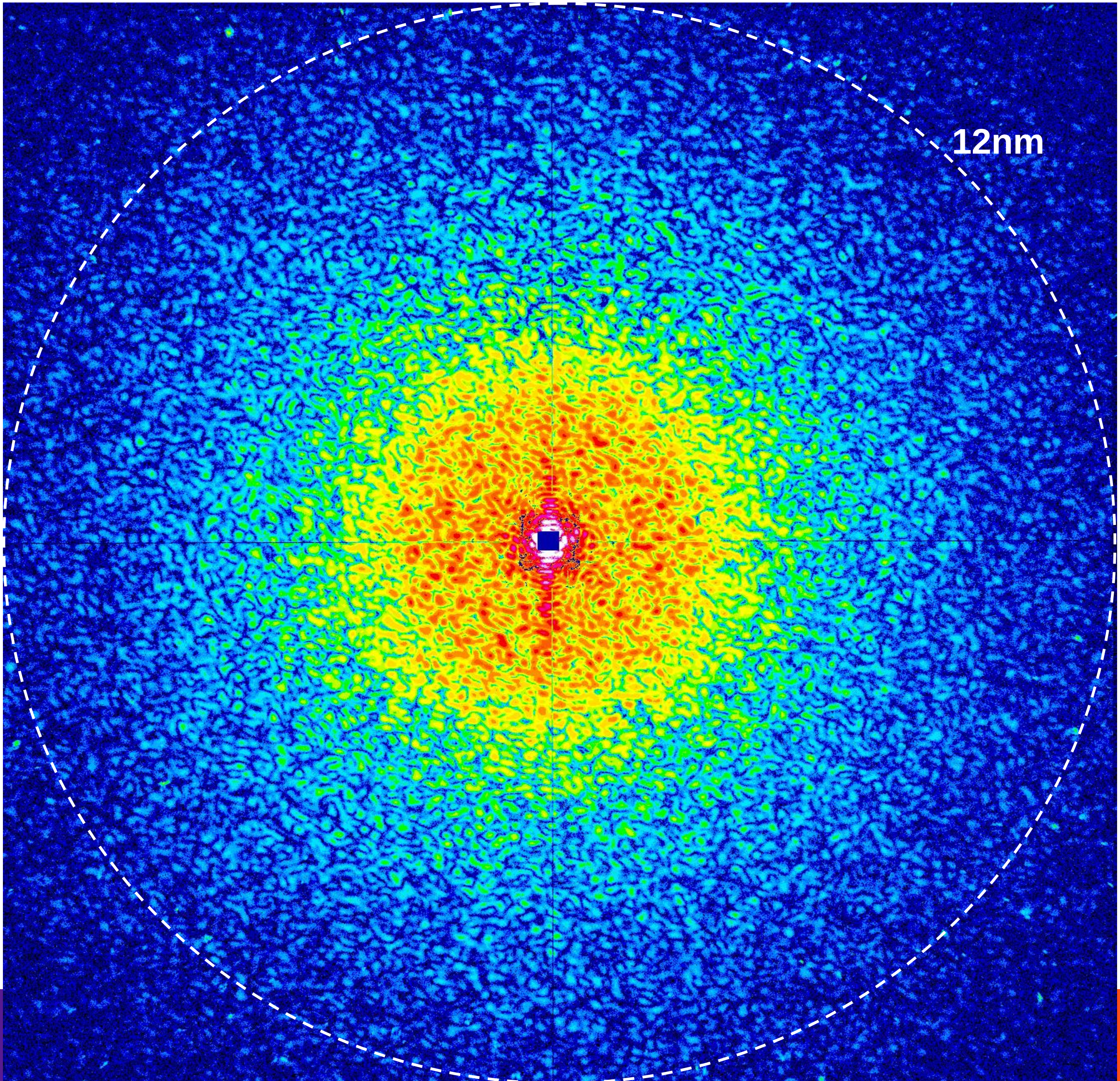
Multiscale porosity, from μm to nm

CDI Experiment at 8-ID on Gold Nanofoam

Xiao, Shen,
Sandy et al.
(APS)
Chen, Dunand
(NU)







Missing-data Problem in CDI

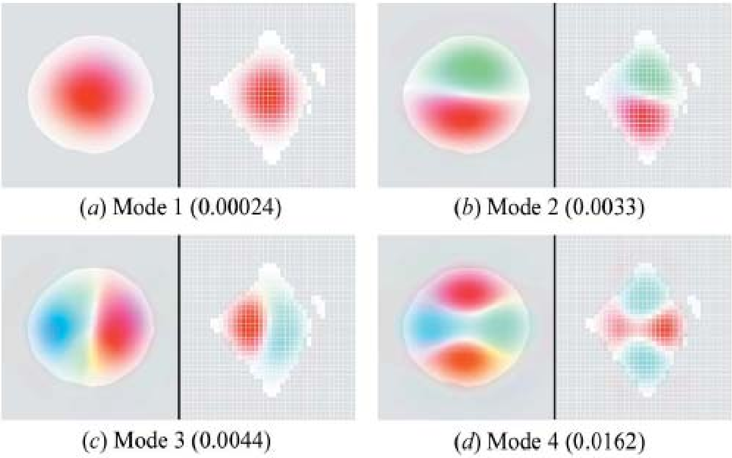
$$\eta_i = \frac{D_i - 1}{2\sigma_i}, \qquad i = x, y, z,$$

FIG. 1. Image reconstruction of oversampled diffraction patterns as a function of the number of missing waves. (a) $\eta_x, \eta_y = 1$. (b) $\eta_x, \eta_y = 2$. (c) $\eta_x, \eta_y = 4$.

in the reconstructed images. The missing structural information cannot be recovered by iterative algorithms no matter whether a tight support is used [7] or the center pixel in the reciprocal-space array is fixed [15]. To reliably solve the missing data problem for general samples, one needs to obtain oversampled diffraction patterns with the missing data confined within the centrospeckle.

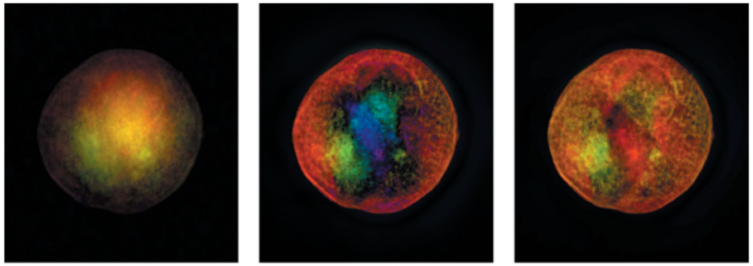
J. Miao et al, PRL, 2005

$$M = \frac{N_C}{\sigma}.$$



(*a*) Mode 1 (0.00024) (*b*) Mode 2 (0.0033)

(*c*) Mode 3 (0.0044) (*d*) Mode 4 (0.0162)

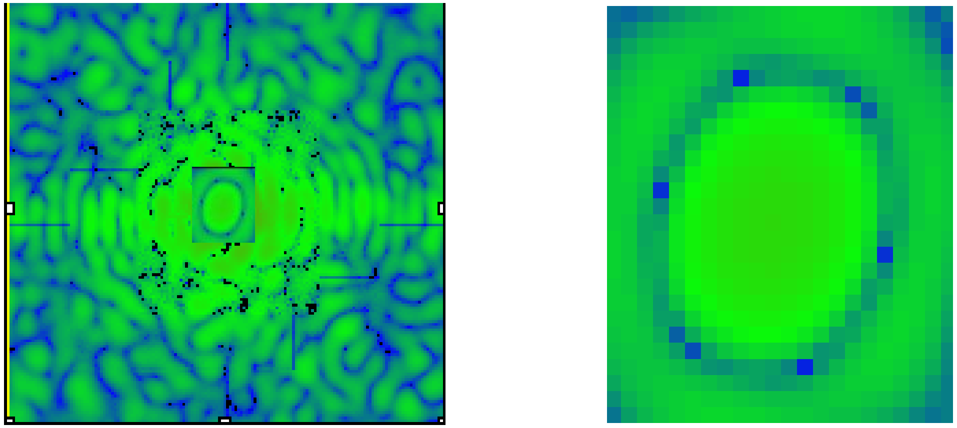


(*a*) (*b*) (*c*)

restoration of the modes using the *ad hoc* variance minimization rule.

V. Elser et al, Acta Cryst., 2006

Solving the CDI Missing-data Problem …



Our data has missing-data region larger than the central speckle. Is the data still usable?

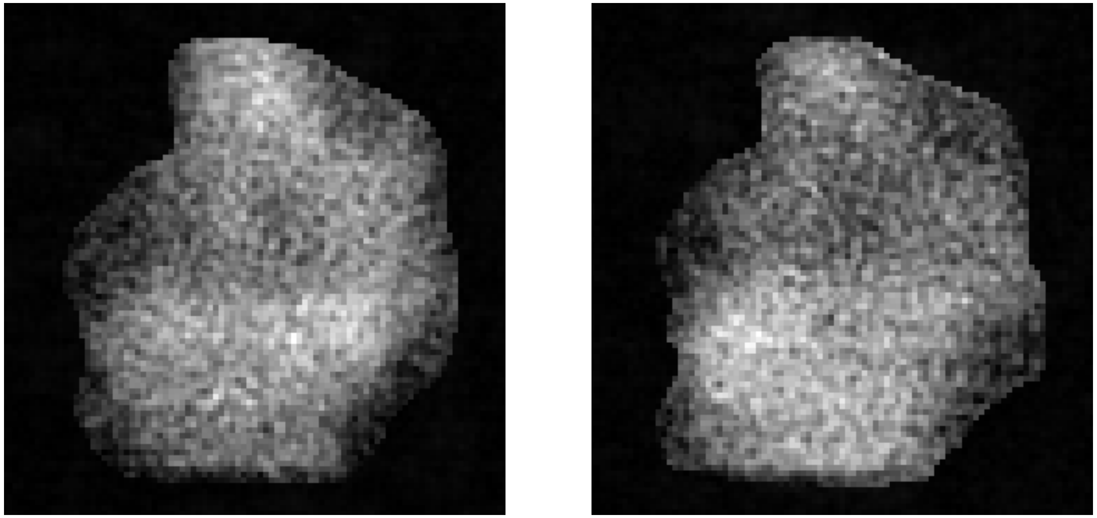
We are developing an algorithm to determine the correct low-Q signal in the missing-data region. The algorithm aims to minimize the following errors:

→ *Global error in real space*
→ *Global error in reciprocal space*
→ *Local error in a vicinity around the missing-data region*

Xiao & Shen, in preparation (2008)

Two Adjacent Projections

DIC optical micrograph

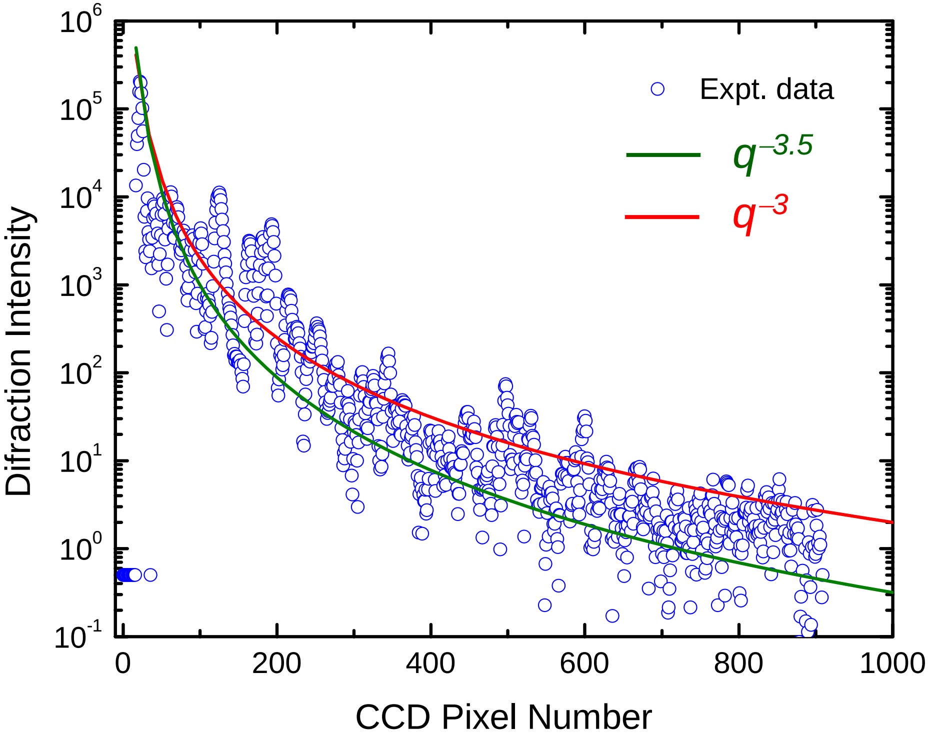


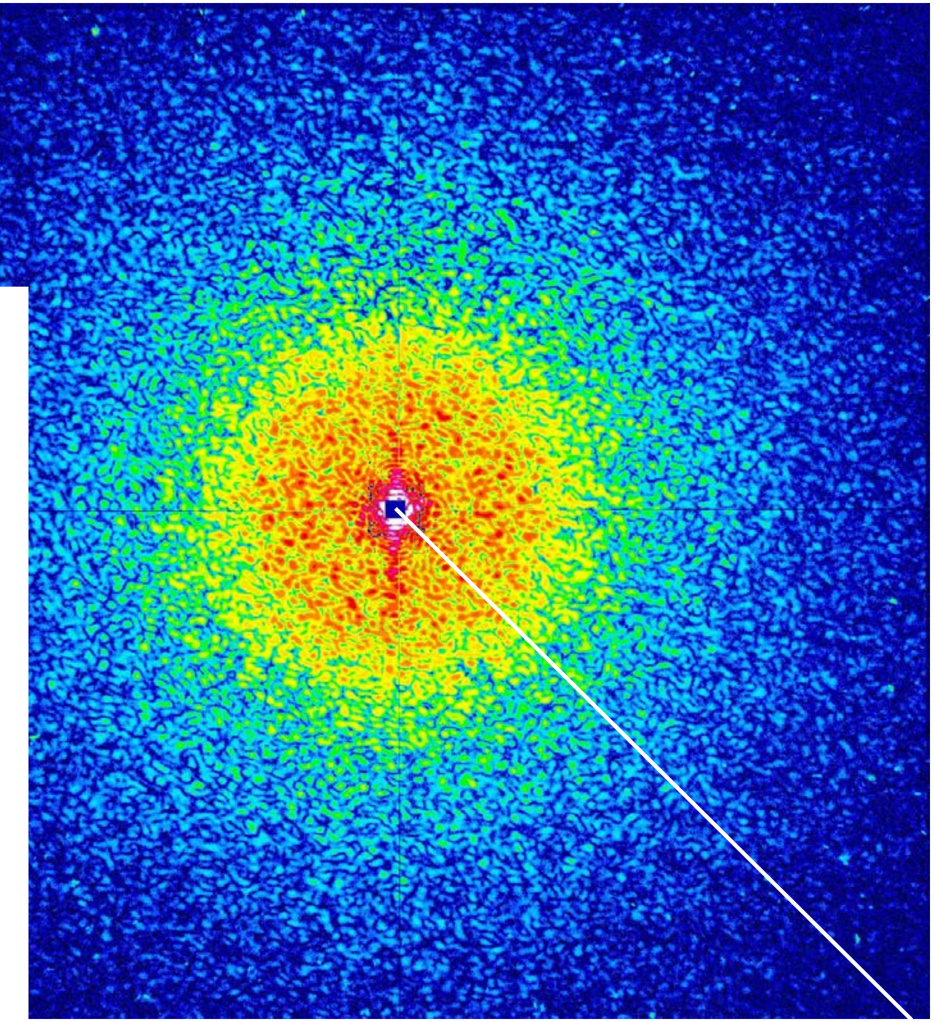
Reconstructed images from projections 5^{o} apart.
Sample size is about 2.1(H)x2.4(V) μm^2.

q-dependence of Coherently Scattered X-Rays

Experiment results:

Gold nanofoam specimen
CDI data to ~ 8 nm





Is Atomic Resolution Possible ?

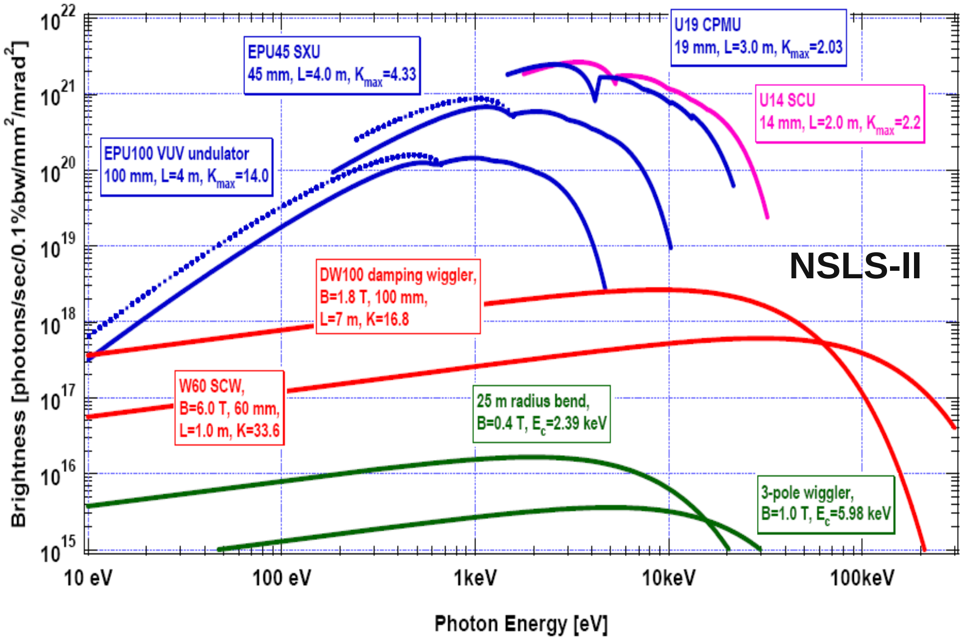
Results at current APS:

Specimen: Au nanofoam
CDI data: to ~ 7 nm, in total of 2 hrs.
CCD detector: PI direct detection
Optics: , Si (111) mono
10 μm pinhole
crystal guard aperture
X-ray wavelength: 1.7 A

APS: B = 5×10^{19} ph/s/0.1%/mm^2/mr^2
$\sigma_x = 270\ \mu m$, $\sigma_y = 12\ \mu m$
Coh. area @ 65m: 17μm x 390μm

If full coherence area is focused:
gain of 1.7x39 = 66x
=> 2 hrs → 1.8 min.

Scaling to 3.5A = 0.35nm resolution:
need x20^3 = x8000 more photons
→ x8000 longer collection time
→ 1.8min x8000 = 14400 min
= 240 hrs !



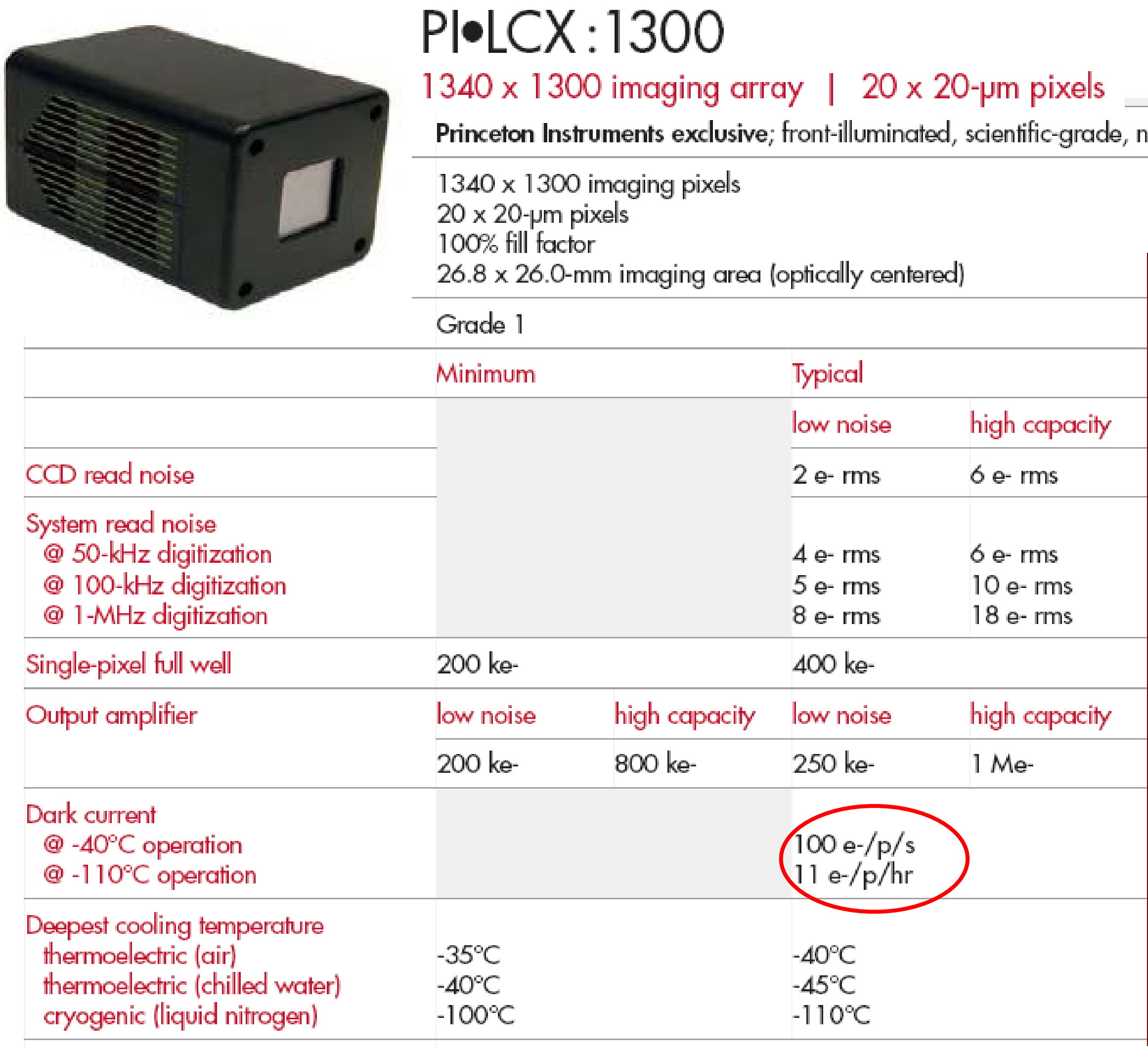
NSLS-II: B = 2×10^{21} ph/s/0.1%/mm^2/mr^2
@ 7 keV with U20 undulator

=> gain of 40x in coherent flux → 6 hrs

=> dual U20 in long straight → 3 hrs.
=> Ge (111) mono. → 1.5 hrs.

➔ A bright source like NSLS-II is essential to atomic resolution CDI

Detector Improvements



PI•LCX:1300

1340 x 1300 imaging array | 20 x 20-µm pixels

Princeton Instruments exclusive; front-illuminated, scientific-grade, non-MPP, deep-depletion device

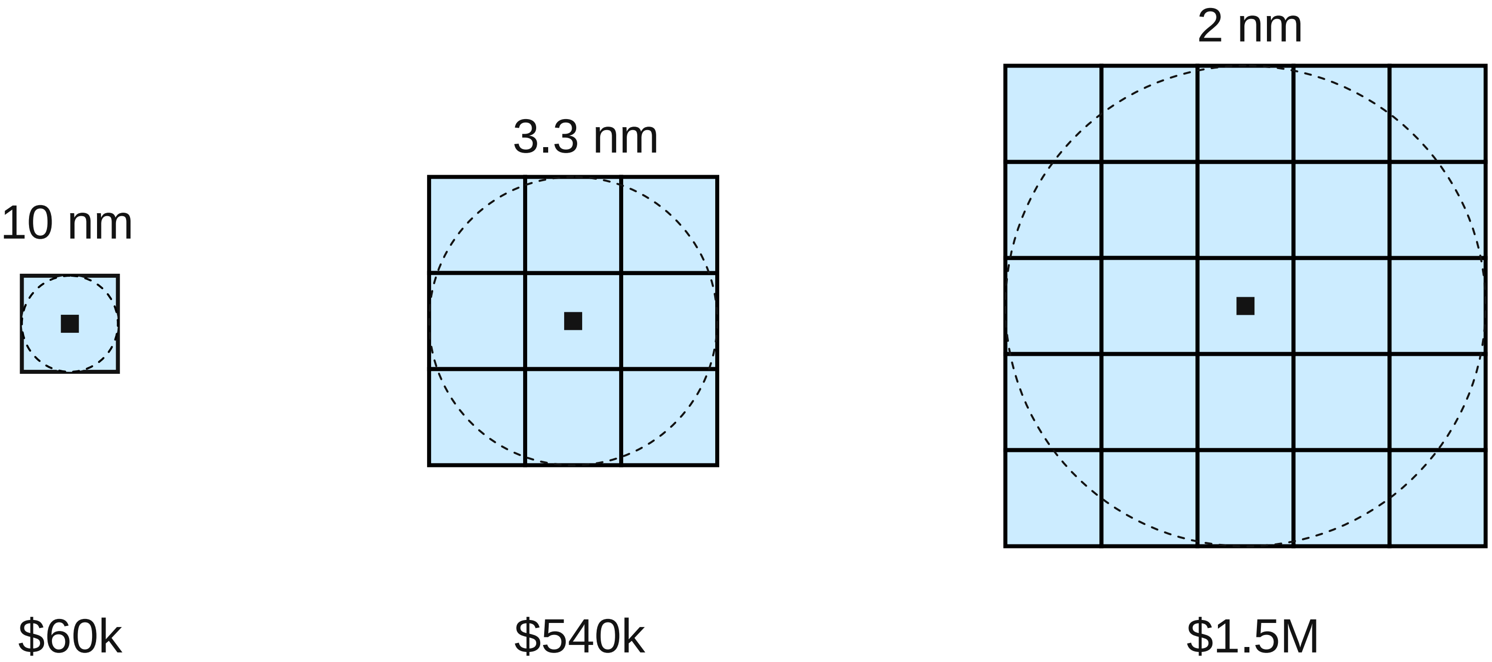
1340 x 1300 imaging pixels
20 x 20-µm pixels
100% fill factor
26.8 x 26.0-mm imaging area (optically centered)

Grade 1

	Minimum		Typical	
			low noise	high capacity
CCD read noise			2 e- rms	6 e- rms
System read noise @ 50-kHz digitization @ 100-kHz digitization @ 1-MHz digitization			4 e- rms 5 e- rms 8 e- rms	6 e- rms 10 e- rms 18 e- rms
Single-pixel full well	200 ke-		400 ke-	
Output amplifier	low noise	high capacity	low noise	high capacity
	200 ke-	800 ke-	250 ke-	1 Me-
Dark current @ -40°C operation @ -110°C operation			100 e-/p/s 11 e-/p/hr	
Deepest cooling temperature thermoelectric (air) thermoelectric (chilled water) cryogenic (liquid nitrogen)	-35°C -40°C -100°C		-40°C -45°C -110°C	

Pilatus 6M detector
(from Dectris)

More Detectors !!



10 nm

3.3 nm

2 nm

$60k

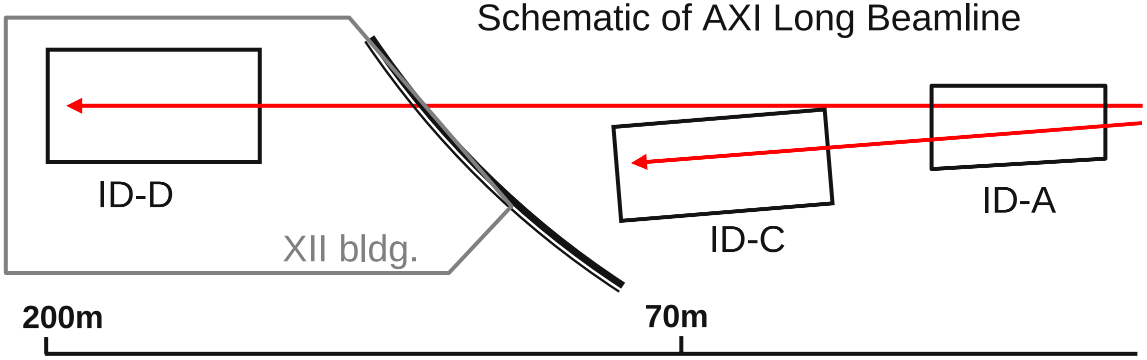
$540k

$1.5M

Proposed Advanced X-ray Imaging (AXI) Long Beamline

→ X-ray Imaging Institute (XII) proposed by APS; Building may be funded by State of Illinois

→ Could enclose a 150-200m long imaging beamline, and provide office & lab (optics, detectors, etc.) spaces to staff and visitors


Schematic of AXI Long Beamline

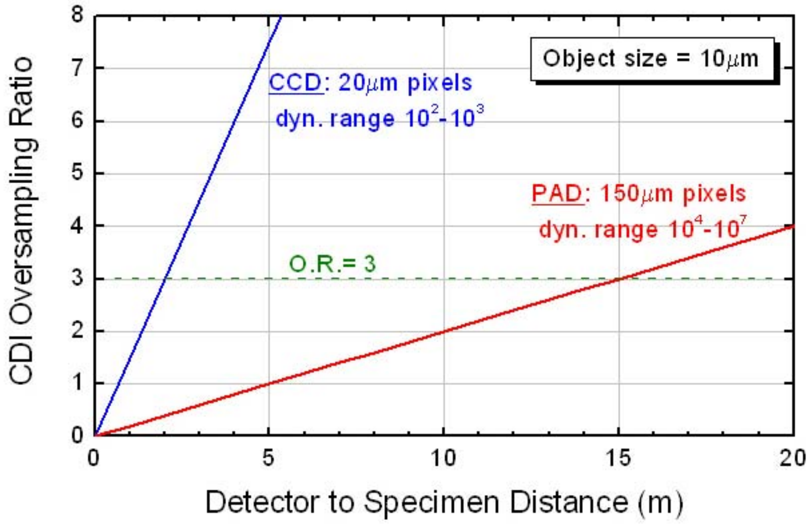
New Imaging Capabilities at Long BL

- High phase sensitivity:
  → internal defect & crack propagations in low-Z materials e.g. polymer foams & nanocomposites
- 100x field of view:
  → opens up comparative biology to larger animals – essential for understanding of evolutionary transitions
  → biomedical imaging and physiological studies on small animals such as mouse, critical for medical applications
- 20m-long CDI hutch:
  → unique at APS, allows the use of high-dynamic-range area detectors such as Pilatus pixel arrays for CDI → crucial in reaching <10nm resolution

Advanced X-ray Imaging (AXI-CDT)

→ CDT Co-Directors: Jon Harrison (ASU)
John Miao (UCLA)
Ian McNulty (APS)

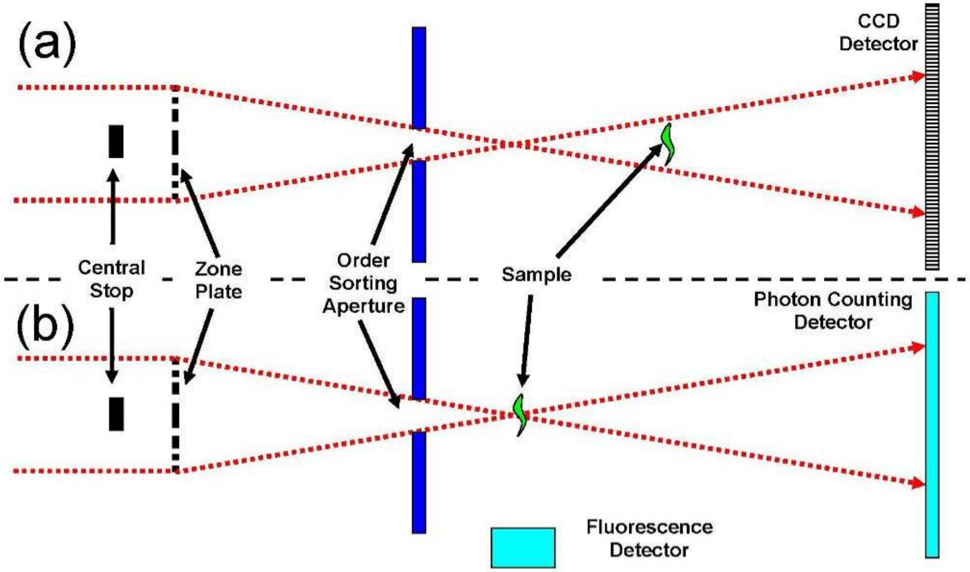
→ Letter of Intent submitted to APS, Nov. 2007

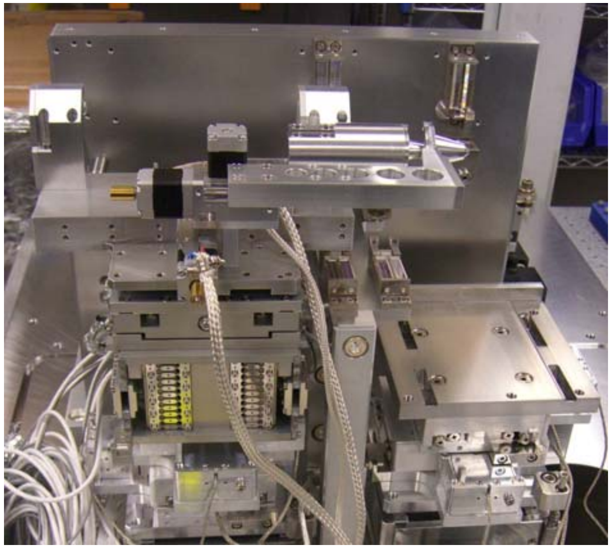


Hybrid Coherent Diffraction Imaging Microscopes

- Combining Fresnel CDI with STXM:

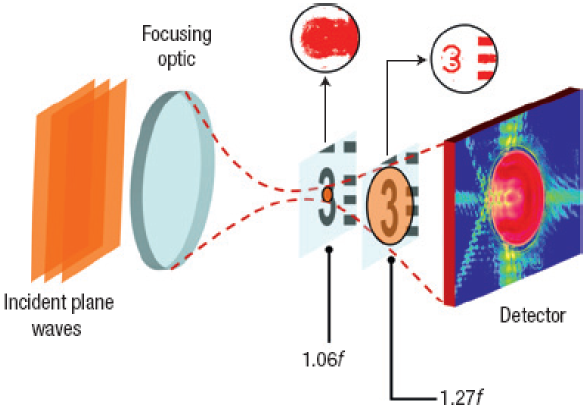
Andrew Peele & Keith Nugent





- Combining Ptychrography CDI with STXM:

X-ray ptychography using a <10nm nanoprobe may present the best opportunity to reach atomic resolution.



Summary

- ***X-ray coherent diffraction imaging*** ***applications are presently limited to ~ 7nm in hard materials and ~ 30nm in biological specimens.***

- ***Radiation damage*** ***is the primary concern to reaching atomic resolution imaging of biological specimens. The ultimate resolution may very well be limited to a few nm.***

- ***For hard radiation resistant materials*** ***atomic resolution may be achievable, but it requires optimized experiment conditions, including sources, optics, detectors, algorithms, and combining CDI with SXM.***

- ***Advanced data analysis algorithms*** ***may allow optimized data treatment in low S/N experiments and thus substantially improve the achievable resolution by CDI technique.***

- ***NSLS-II should play a leading role in developing the atomic resolution CDI potential since it will offer brightest x-rays and highest coherence in the next 5-10 years.***

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