MSAE E8235x – X-ray Microscopy

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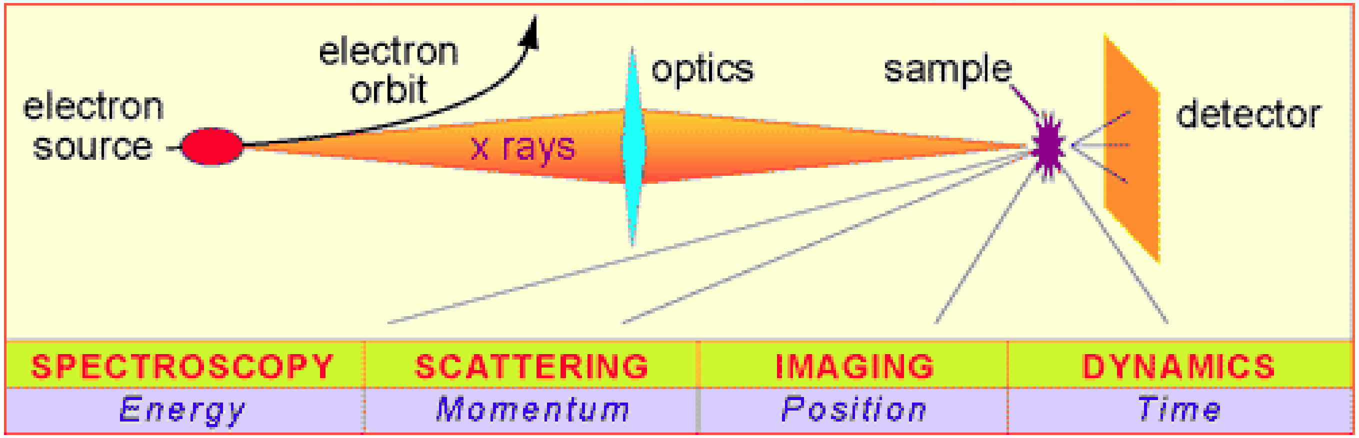
Synchrotron X-ray Techniques for Scientific Research



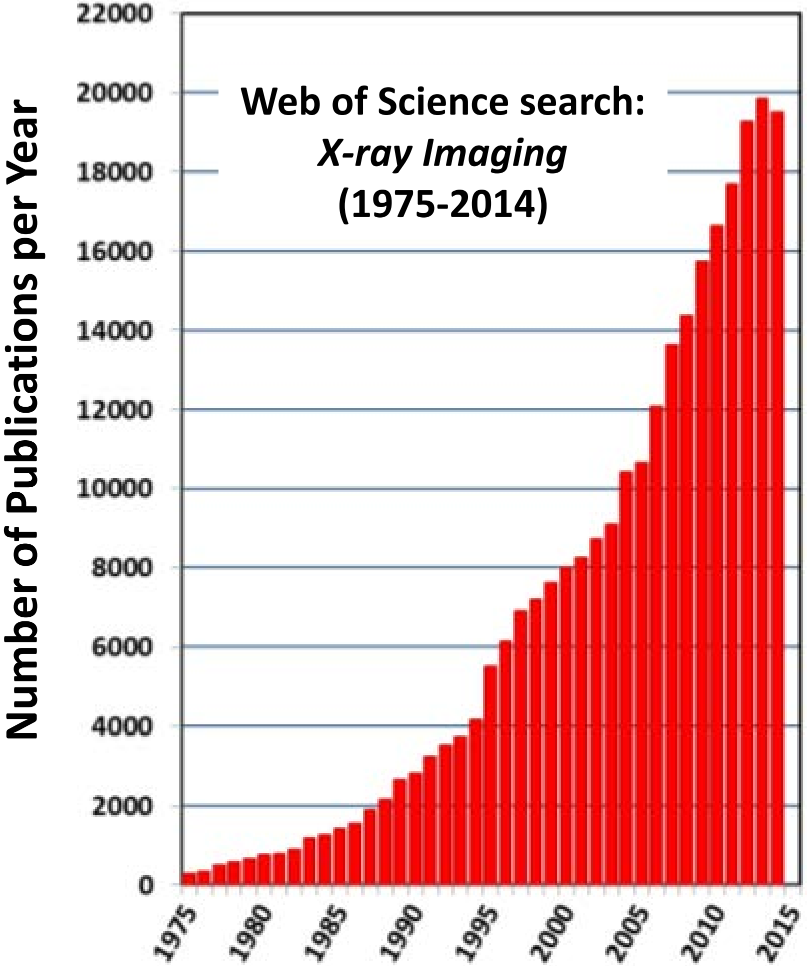
http://www.sc.doe.gov/bes/synchrotron_techniques/

- **SPECTROSCOPY:** used to study the energy & the quantity of emitted particles when incident x-rays are absorbed by the sample ➔ determine the characteristics of chemical bonding and electron energy band structure.
- **SCATTERING**/diffraction: make use of the patterns of scattered x-rays when incident x-rays are deflected by atoms in a sample ➔ determine the structural arrangement of atoms in the sample.
- **IMAGING:** use the intense x-ray beam to obtain structural pictures of the sample with fine spatial resolution and chemical information ➔ used in diverse research areas to image from biological cells to semiconductor chips and fuel cells.
- **Dynamics:** perform spectroscopy, scattering, & imaging measurements vs. time ➔ used to study structural and electronic changes in the material.

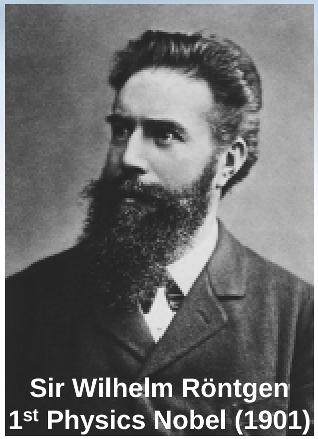
Today's Lecture

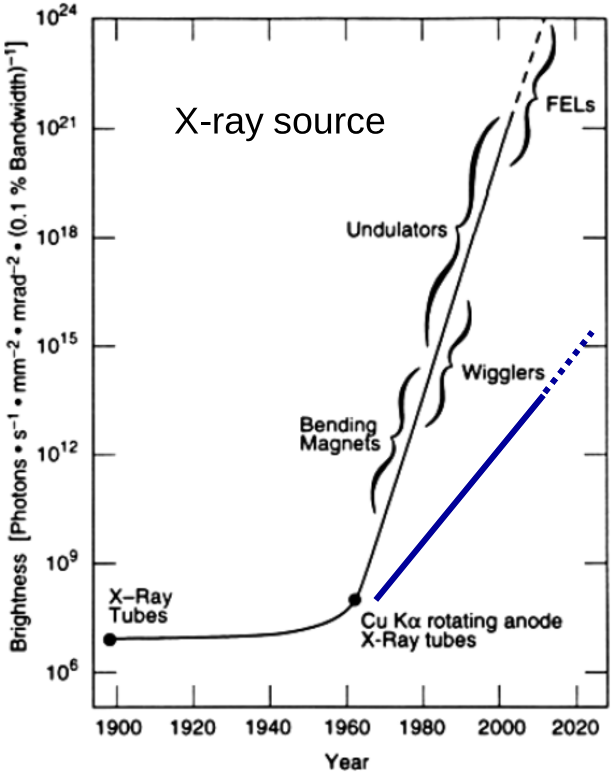
- Introduction to X-ray Imaging/Microscopy
  - absorption & phase contrast
  - Fresnel wave propagation
- Lens-based X-ray Microscopy
  - focusing optics: refractive lens, reflective & diffractive optics
  - Fresnel zone plate: numerical aperture, resolution, magnification
  - full-field transmission x-ray microscope
  - scanning x-ray microscopy
- Lens-less X-ray Microscopy
  - coherent diffraction imaging/microscopy
  - coherent diffraction ptychography
- Application Examples in Each Section

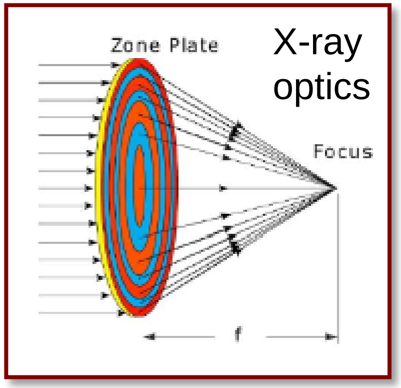
Renewed Interests in X-ray Imaging



Data Source: *Web of Science*

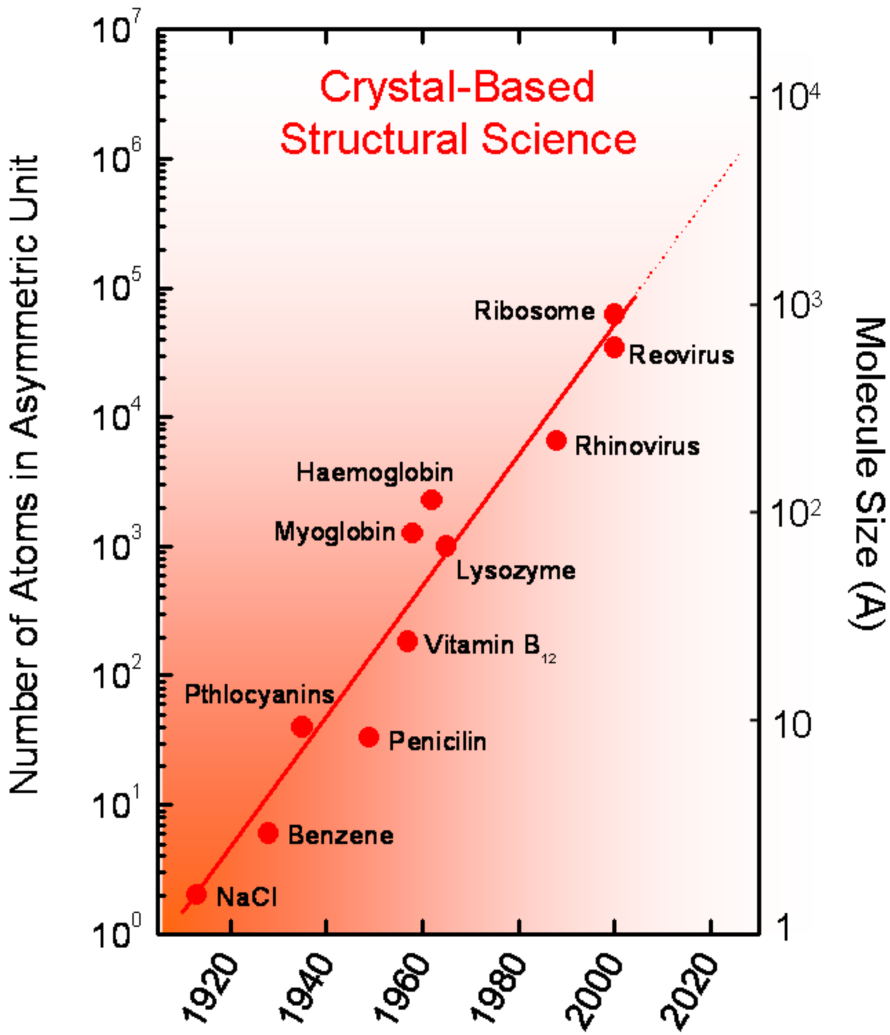




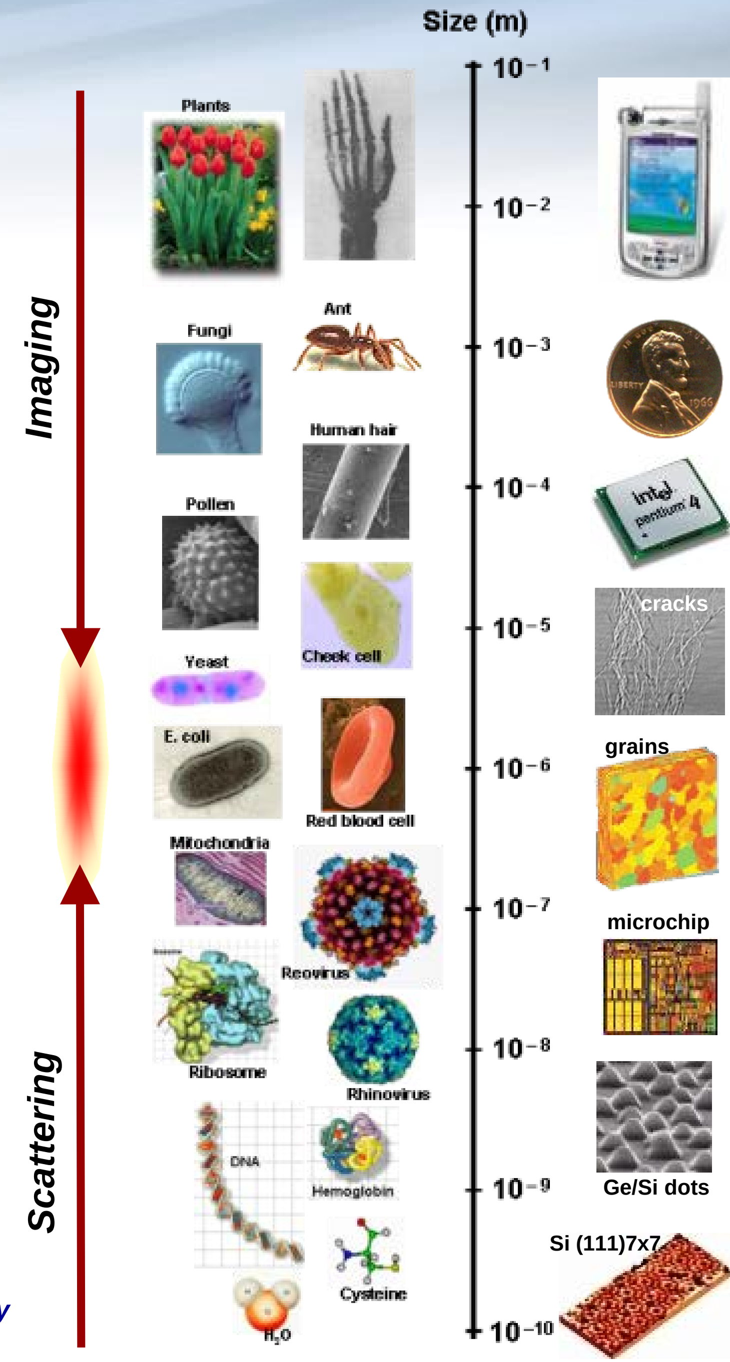


Scientific Interests

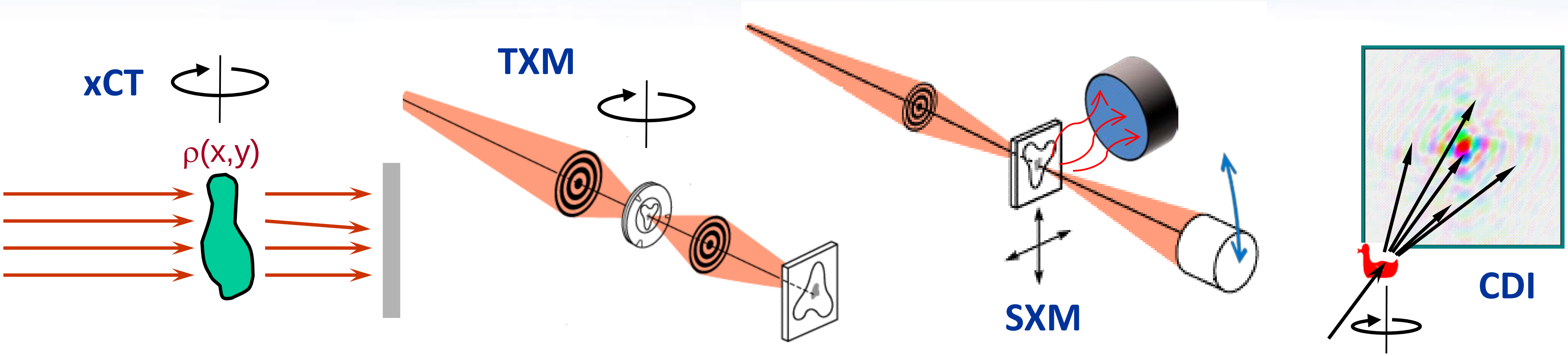
- Structure of non-ideal, non-crystalline materials
- Structure-function relations – heterogeneous



Shen et al.
Physics Today
(March 2006)

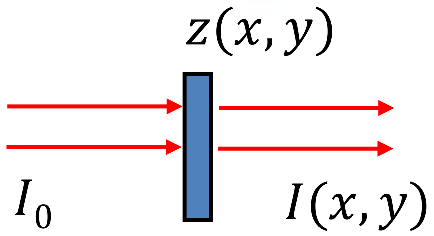


Synchrotron X-ray Imaging & Microscopy Techniques



- **X-ray Tomography (xCT):** images internal structures in 3D ➔ very much like clinical x-ray CT but with both absorption and phase contrasts, and with ~1 μm resolution.
- **Transmission X-ray Microscopy (TXM):** applies an x-ray lens as objective to obtain high resolution image of internal structures in 3D ➔ provides much larger depth penetration than TEM, ideal for in-situ studies of materials in real conditions.
- **Scanning X-ray Microscopy (SXM):** applies an x-ray lens to focused x-ray spot and measures transmitted, fluorescence, and/or diffracted signals as function of raster scan position on specimen ➔ image heterogeneities in sample.
- **Coherent Diffraction Imaging (CDI):** measures x-ray diffraction pattern from non-crystalline specimens ➔ applies phase retrieval to obtain real-space image.

Phase Contrast vs. Absorption Contrast



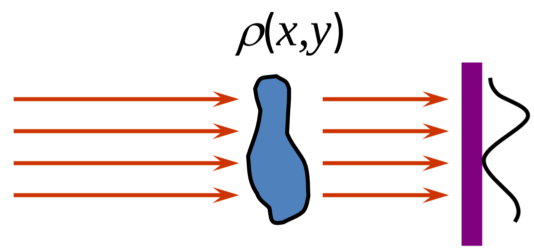
Refraction index: $n = 1 - \delta - i\beta$

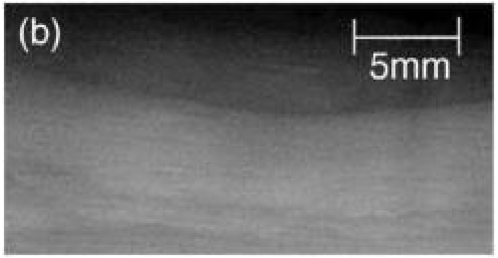
$$E(z) = E_0 e^{-ik_0 nz} = E_0 e^{-ik_0 z(1-\delta-i\beta)}$$
$$= E_0 e^{-k_0 \beta z} \cdot e^{-ik_0 z(1-\delta)}$$

$$I(z) = |E(z)|^2 = |E_0|^2 e^{-2k_0 \beta z} = I_0 e^{-4\pi\beta z/\lambda} = I_0 e^{-\mu z}$$

$\Rightarrow$ Absorption contrast:

$$\mu = \frac{4\pi\beta}{\lambda} \propto \lambda^3$$

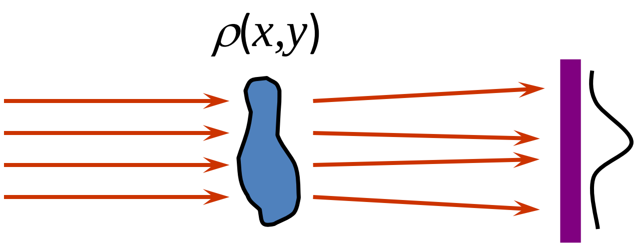


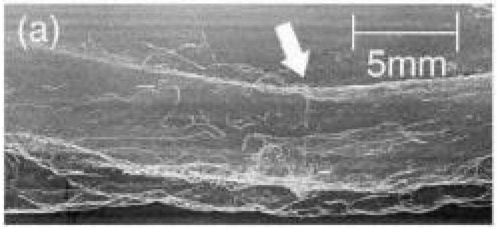


$\Rightarrow$ Phase contrast:

$$\phi = \frac{2\pi\delta z}{\lambda} \propto \lambda$$

$$(\because\ \delta = \frac{r_e N Z \lambda^2}{2\pi})$$





Mori* et al. *(2002): broken rib with surrounding soft tissue

Four Ways to See Phase Contrast

- ***Interferometric imaging***

  ➔ ϕ (phase)

- ***Diffraction enhanced imaging***

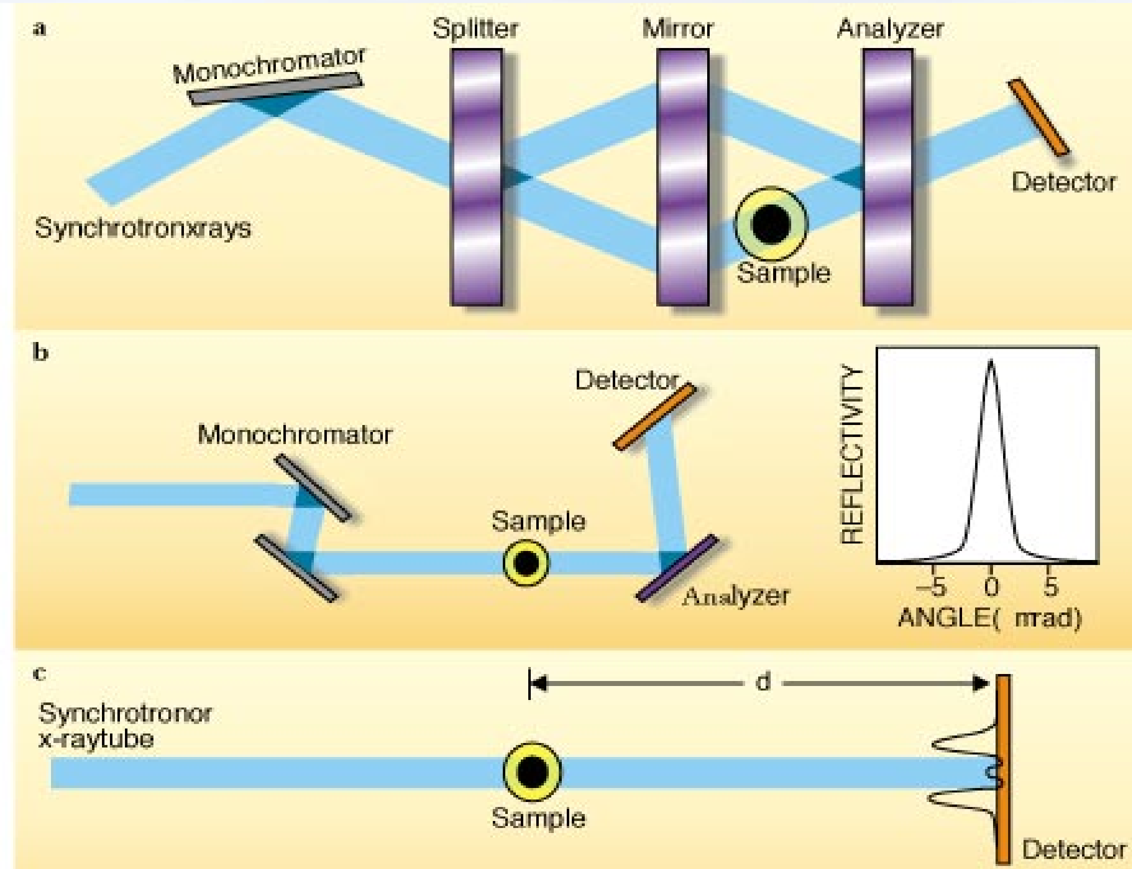
  ➔ $\nabla\phi$ (gradient)

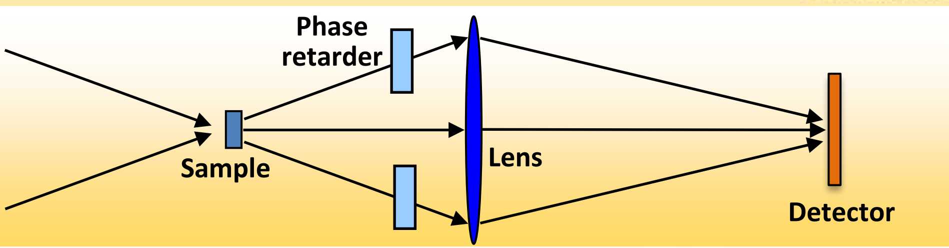
- ***Fresnel wave propagation***

  ➔ $\nabla^2\phi$ (Laplacian)

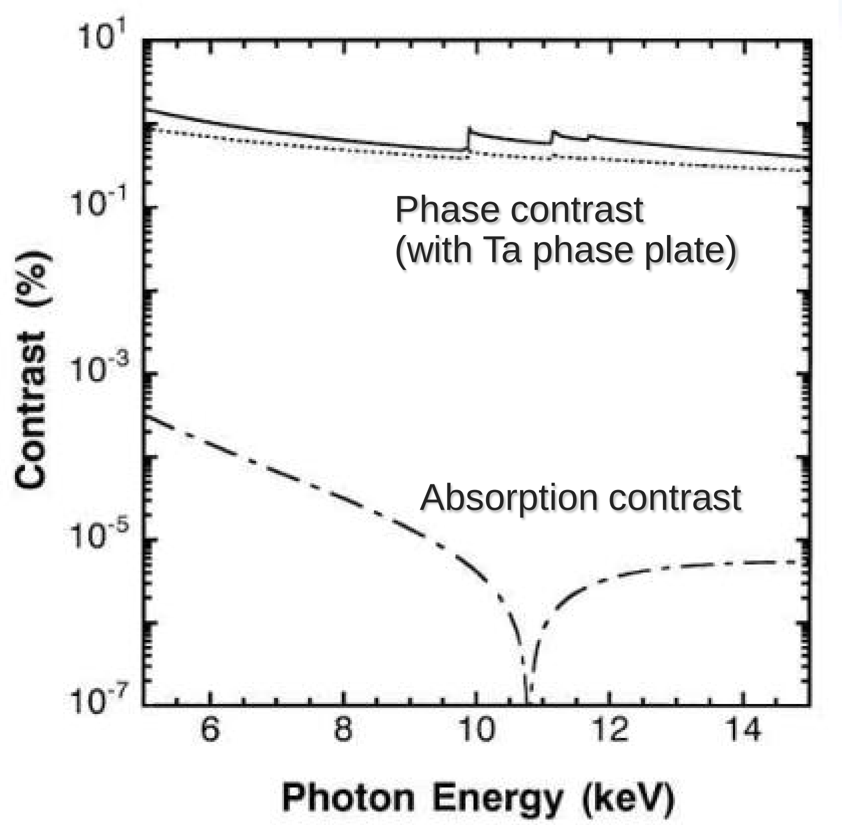
- ***Zernike phase contrast***

  ➔ ϕ (phase)





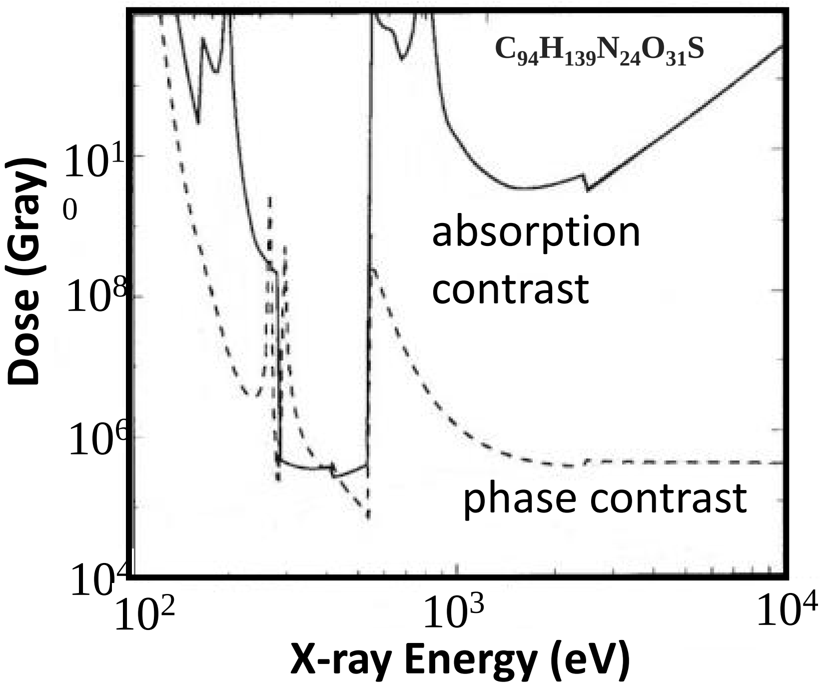
Phase Contrast vs. Absorption Contrast



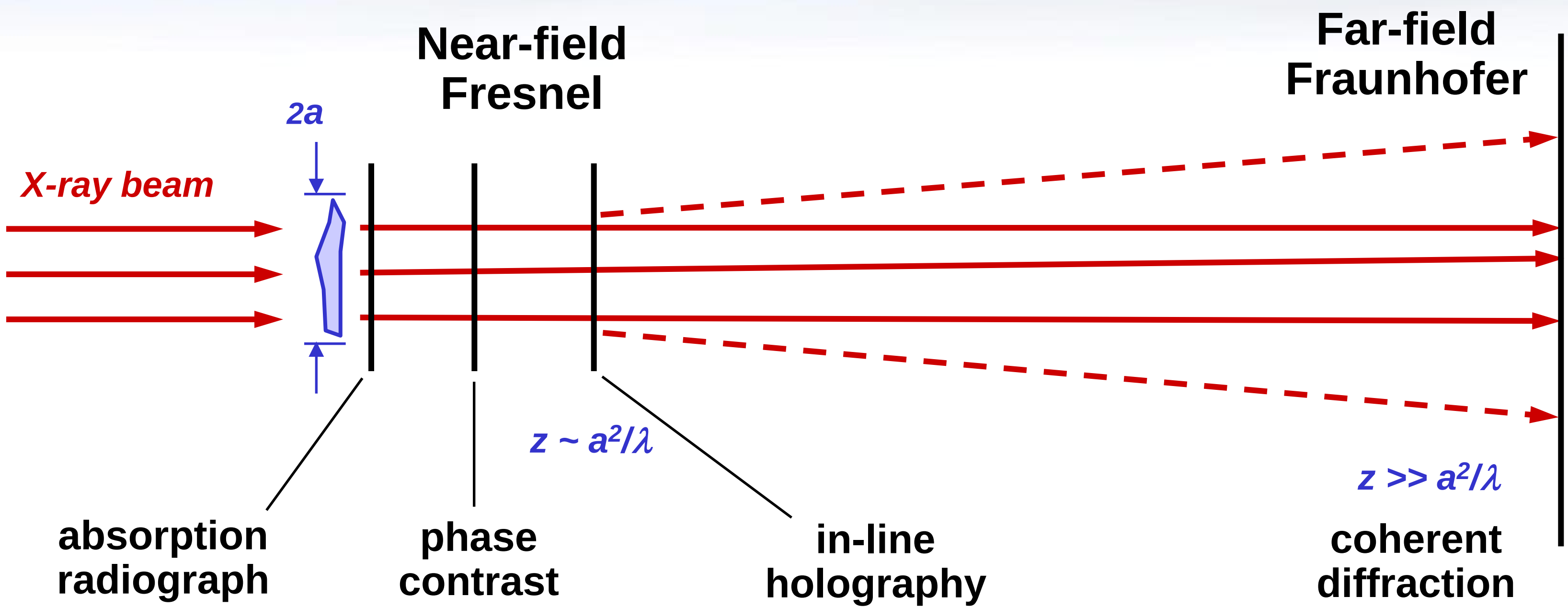
Kagoshima et al. (2001): *protein $C_{94}H_{139}N_{24}O_{31}S$*
ρ=1.35g/cm^3, t=0.1μm in 10μm water

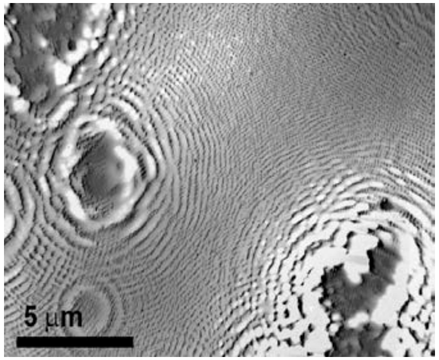
- Phase contrast is x10^4 higher than absorption contrast for protein in water @ 8keV
- Required dose reduced due to phase contrast

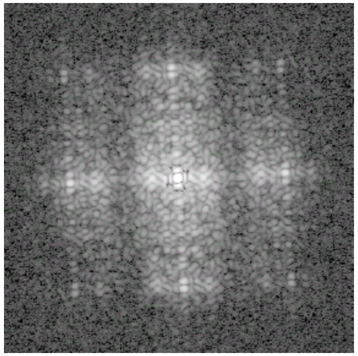
Kirz (1995): *0.05μm protein in 10μm thick ice*



Wave Propogation in X-ray Imaging







Miao et al.
Nature (1999).

Fresnel Wave Field Propagation

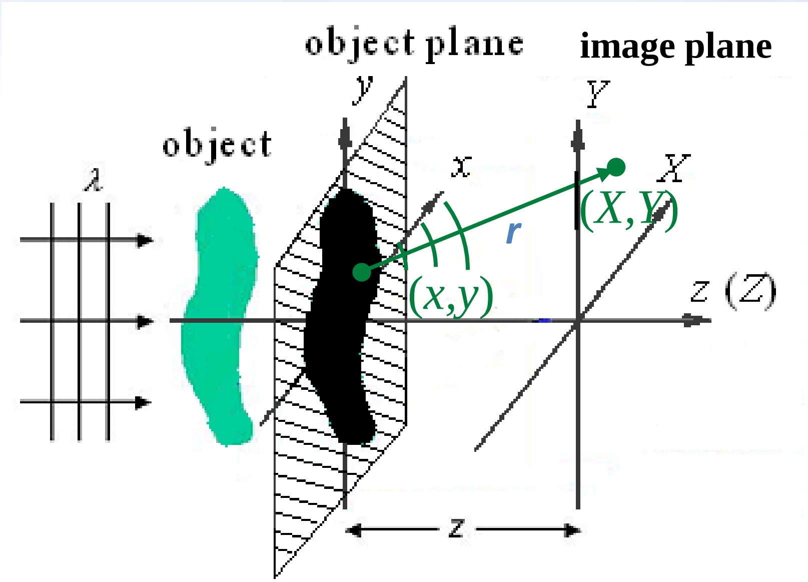


Image plane: plane where image is formed.

Fresnel-Huygens Principle: Wave field on image plane is the coherent sum of all scattered waves from object, weighted by object transmission function.

➔ **Fresnel diffraction for wave propagation**

$$\boxed{F(X,Y) = \iint u(x,y)\frac{e^{-ikr}}{\lambda r}dxdy}$$

$$r = [z^2 + (X-x)^2 + (Y-y)^2]^{1/2}$$

$$\approx \ z + [(X-x)^2 + (Y-y)^2]/2z$$

$$\boxed{F(X,Y) = \frac{e^{-ikR}}{\lambda R}\iint u(x,y) e^{-\frac{i\pi}{\lambda z}(x^2+y^2)} e^{-\frac{i2\pi}{\lambda z}(Xx+Yy)}dxdy}$$

$$R = (X^2 + Y^2 + z^2)^{1/2}$$

Object plane: plane located at z = 0 just downstream of specimen, onto which transmission of incident wave through the specimen is projected.

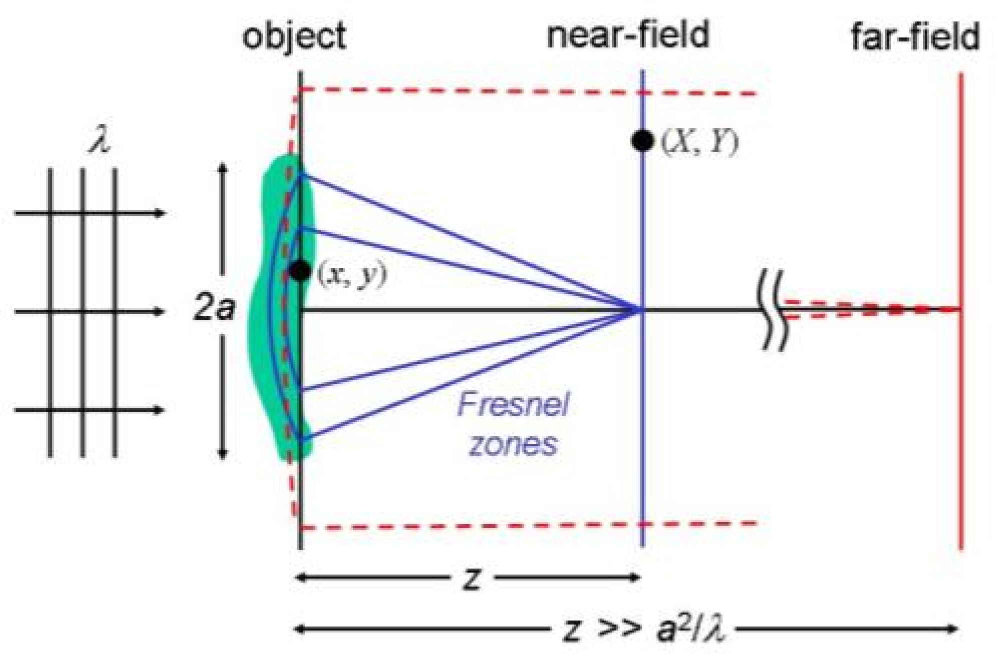
➔ **Wave-field on object plane = transmission function *u(x, y)***

$$u(x,y) = \exp\left\{-ik\cdot\int_{-\infty}^{0}[\delta(x,y,z) + i\beta(x,y,z)]dz\right\}$$

$$\approx \exp(-ik\int_{-\infty}^{0}\delta(x,y,z)dz) \quad \text{(pure phase object)}$$

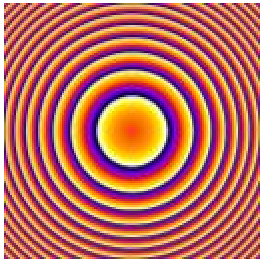
Van der Veen & Pfeiffer, *J. Phys.: Condens. Matter 16, 5003 (2004)*

Distorted Object Approach for Fresnel Propagation



Phase-chirped distorted object:

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



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

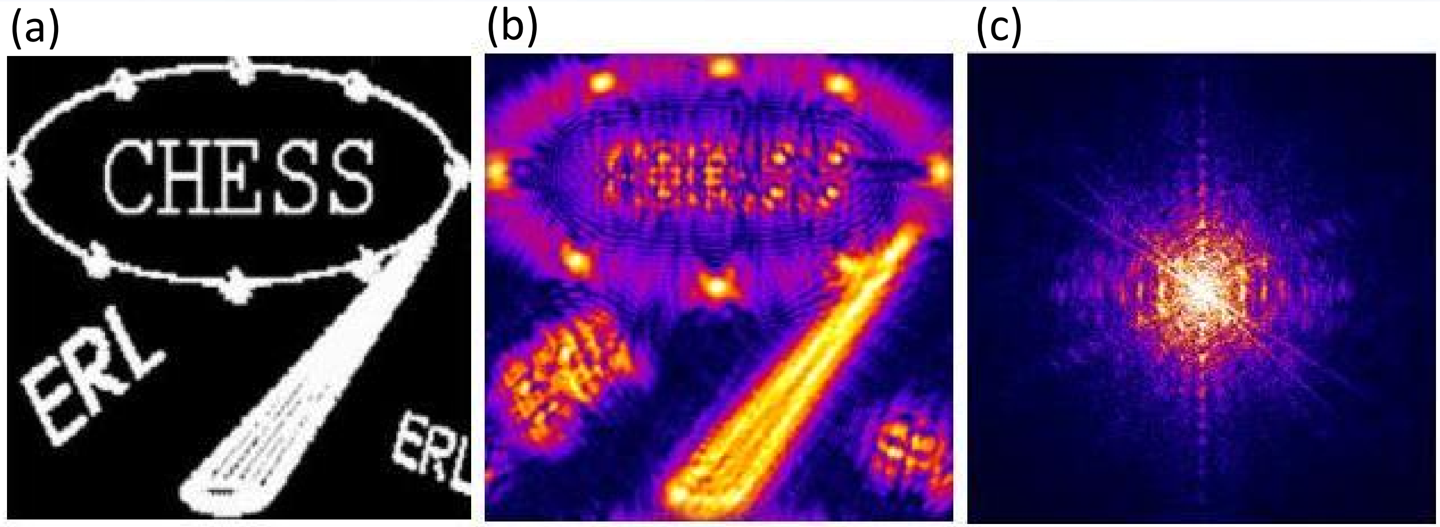
⇒ Unified wave propagation method by Fourier transform

Momentum transfer: $(Q_x, Q_y) = (kX/z, kY/z)$

Number of Fresnel zones: $N_z = a^2/(\lambda z)$

Xiao & Shen, PRB 72, 033103 (2005).

Example of Distorted Object Approach



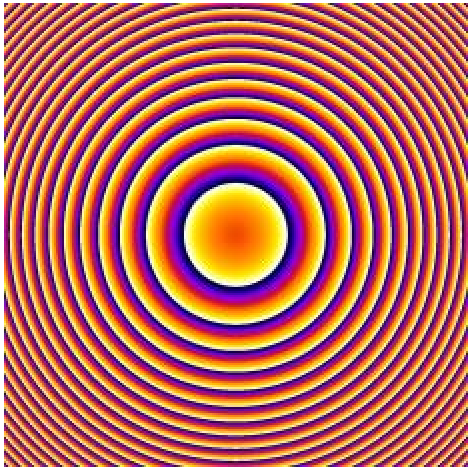
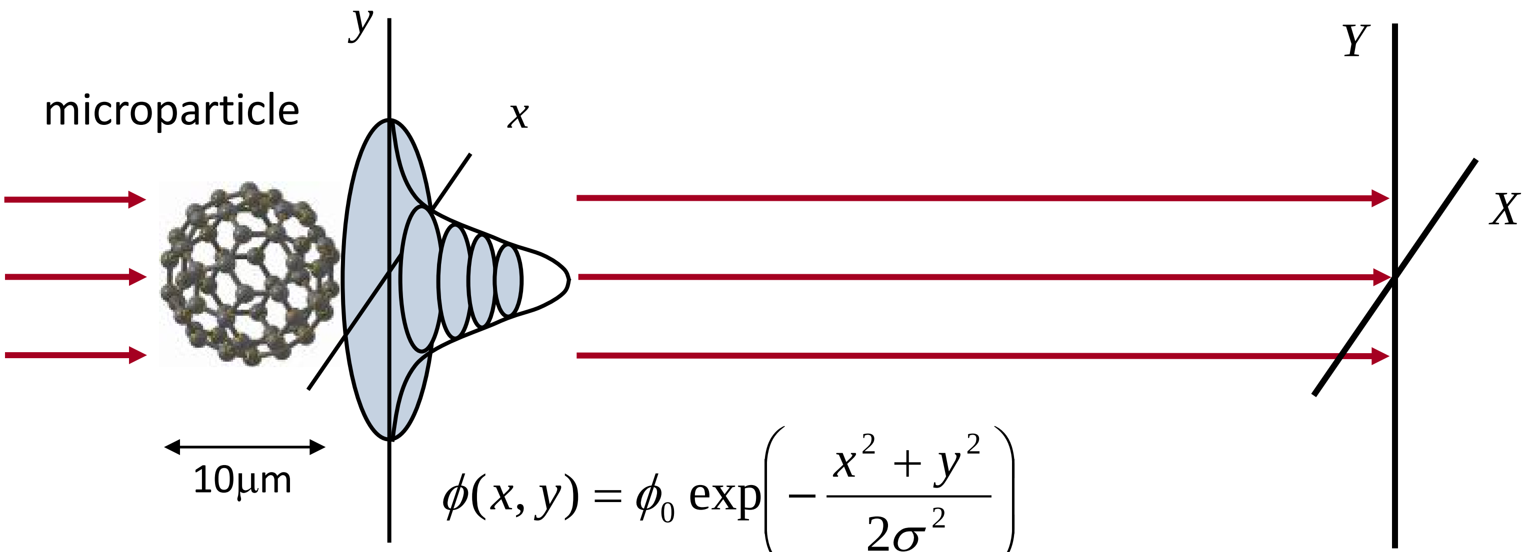


Figure: Simulated diffraction amplitudes $|F(X,Y)|$, of an amplitude object (a) of 10μm x 10μm, with λ = 1 Å x-rays, at image-to-object distance (b) z = 2mm and (c) z = ∞, using the unified distorted object approach with N_z = 500 zones in (b) and N_z = 0 in (c). Note that the diffraction pattern changes from noncentrosymmetric in the near-field (b) to centrosymmetric in the far-field (c).

Xiao & Shen, PRB 72, 033103, July 2005.

Gaussian Phase Object



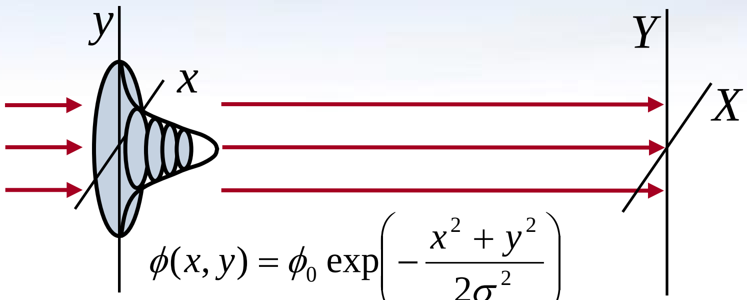
$$\phi(x,y) = \phi_0 \exp\left(-\frac{x^2+y^2}{2\sigma^2}\right)$$

Phase shift by carbon microparticle: size ~ 10μm

$\rho = 2$ g/cm^3, $\lambda = 1$A => $\delta = 1.35\text{x}10^{-6}\lambda^2\rho = 2.7\text{x}10^{-6}$

$\phi = 2\pi\delta\, z / \lambda = 2\pi$ x $2.7\text{x}10^{-6}$ x100,000 / 1 = 0.54π = 1.7 rad

Useful Applications of 2D Gaussian Phase Object

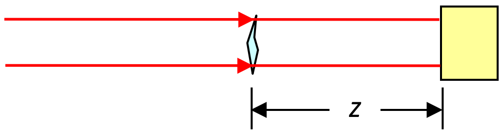


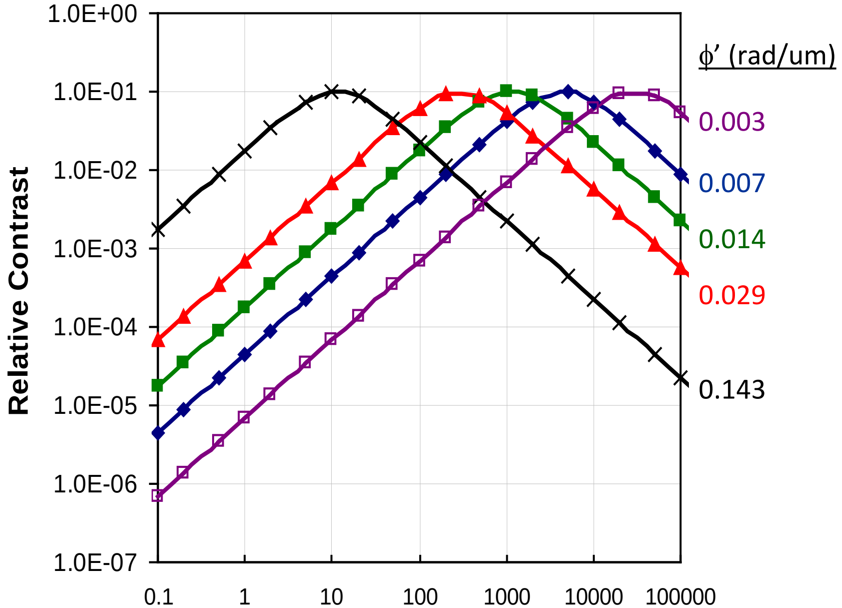
$$\phi(x,y) = \phi_0 \exp\left(-\frac{x^2+y^2}{2\sigma^2}\right)$$

2D Gaussian phase object

➔ Analytical expressions

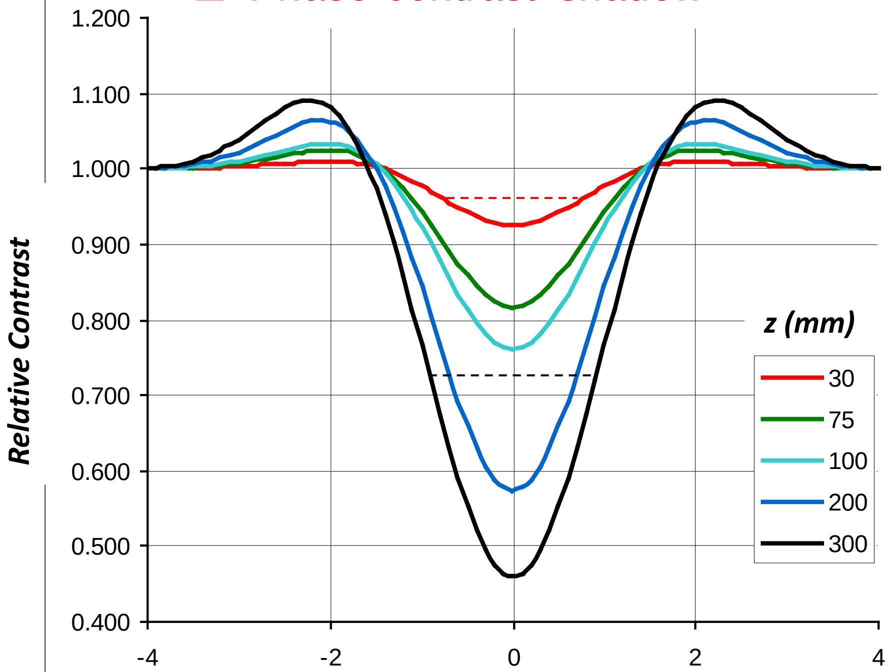
❑ *Phase imaging sensitivity*





Wave Propagation Distance z (mm)

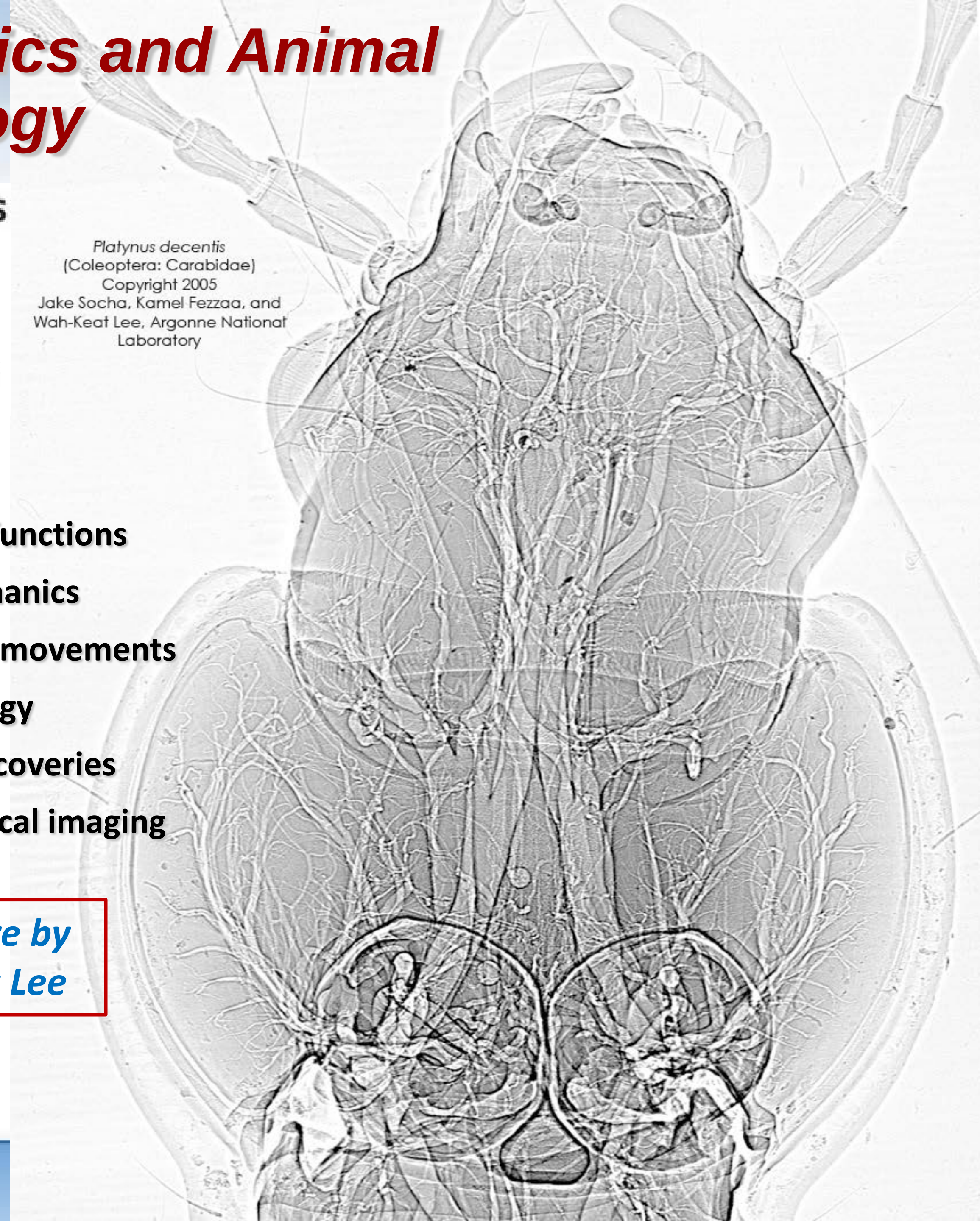
❑ *Phase-contrast 'shadow'*



Imaging Biomechanics and Animal Physiology

Tracheal Respiration in Insects Visualized with Synchrotron X-ray Imaging

Mark W. Westneat,*[1] Oliver Betz,[1,2] Richard W. Blob,[1,3] Kamel Fezzaa,[4] W. James Cooper,[1,5] Wah-Keat Lee[4]
Field museum of Chicago & APS, Argonne National Lab.



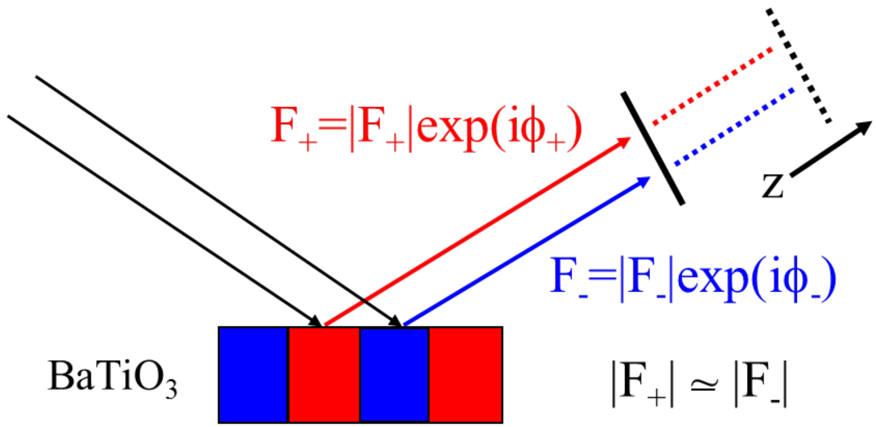
wood beetle

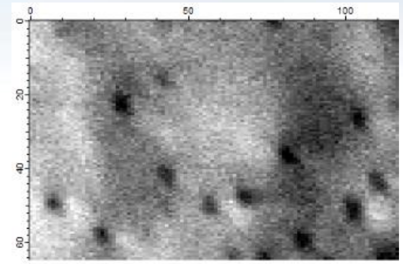
- **Animal functions**
- **Biomechanics**
- **Internal movements**
- **Physiology**
- **New discoveries**
- **Biomedical imaging**

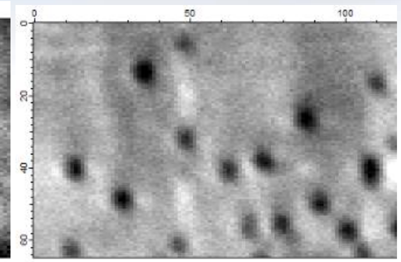
See Lecture by Wah-Keat Lee

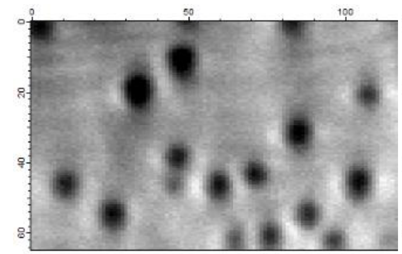
Science (2003) 299, 598-599.

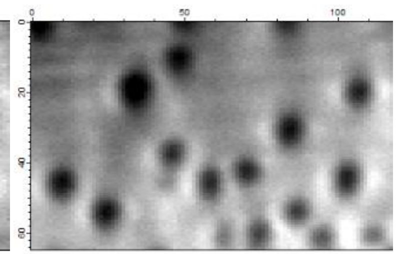
Imaging Ferroelectric Domains by Phase-Contrast X-ray Topography

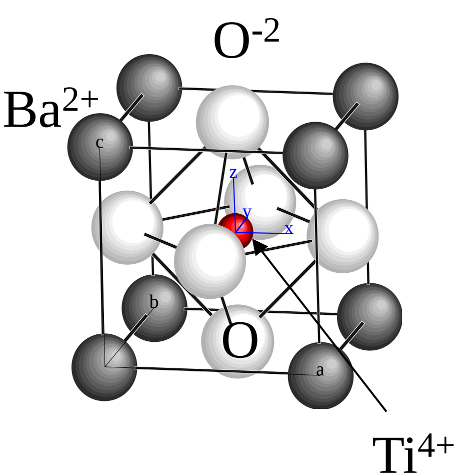


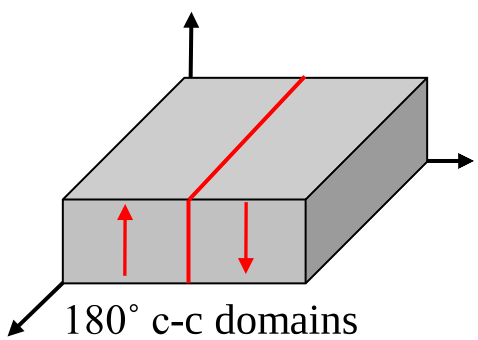

Z=30 mm

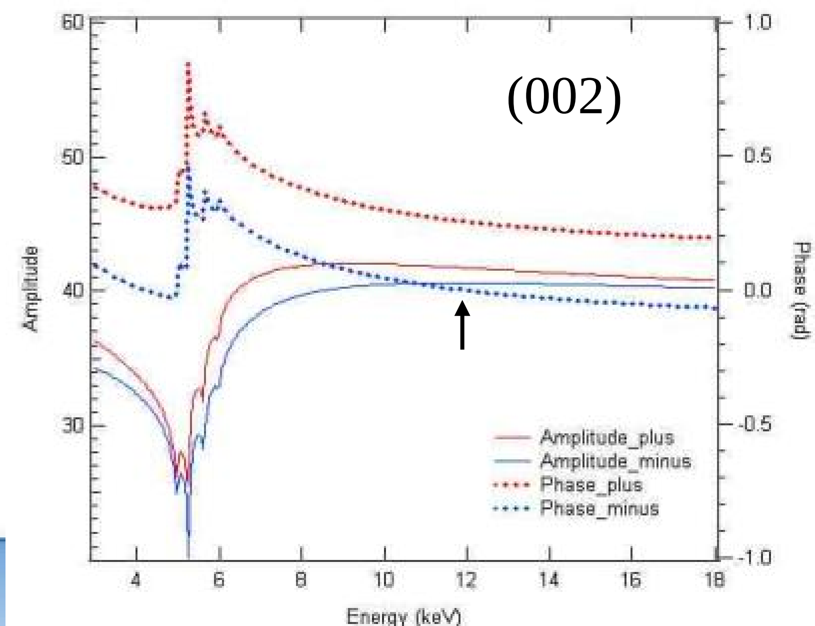

Z=75 mm


Z=150 mm


Z=300 mm







Y. Chu et al. (APS)

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  - coherent diffraction ptychography
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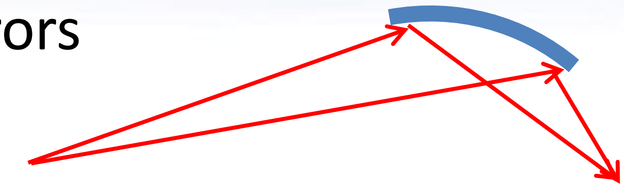
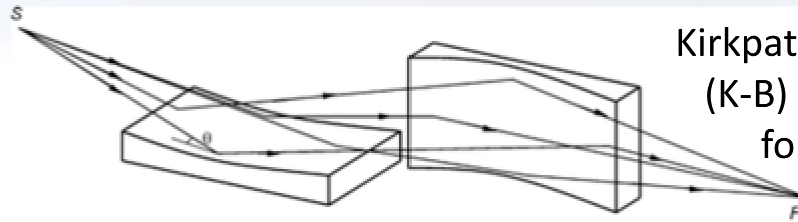
Focusing Optics for X-rays

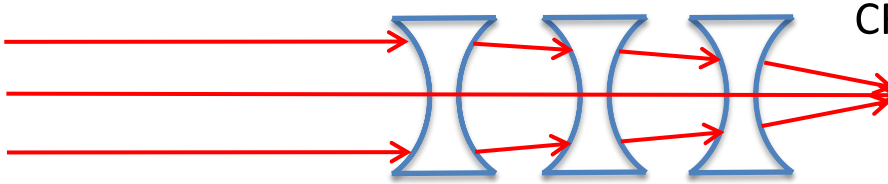
- Mirrors





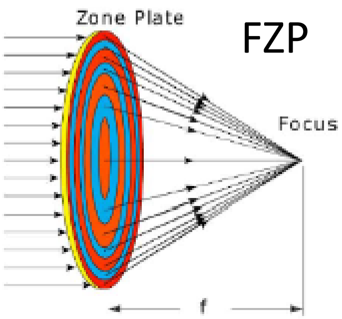
Kirkpatrick-Baez (K-B) mirrors for 2D

- Compound Refractive Lens (CRL)

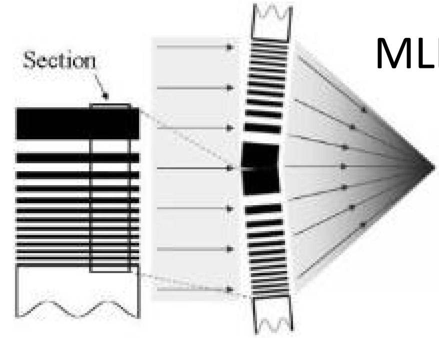


CRL

- Fresnel zone plate (FZP)



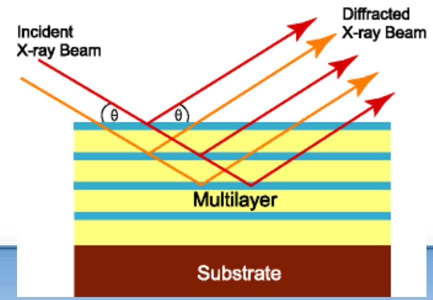
FZP



MLL

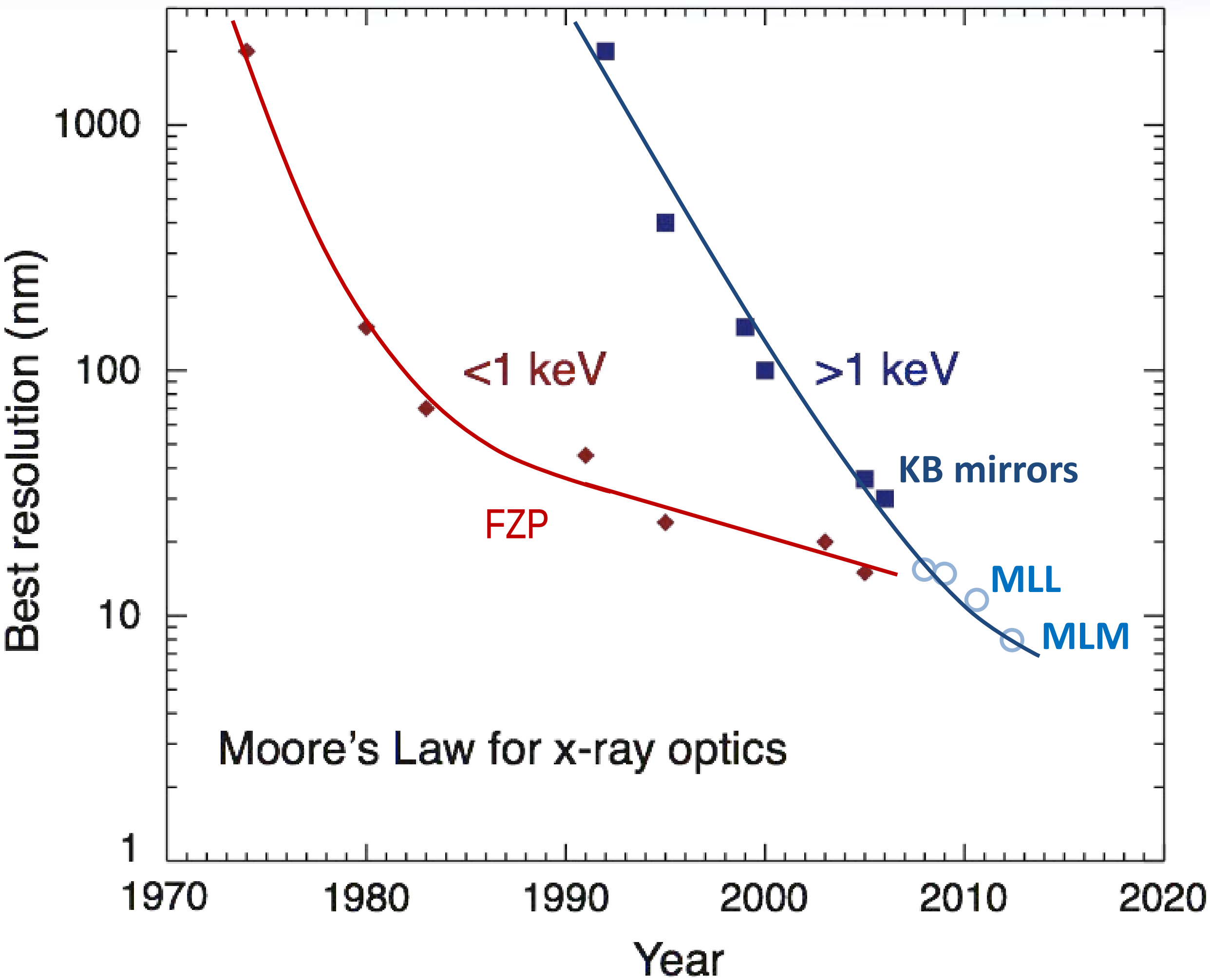
- Multilayer Laue-Lens (MLL)

- Multilayer mirrors (MLM)



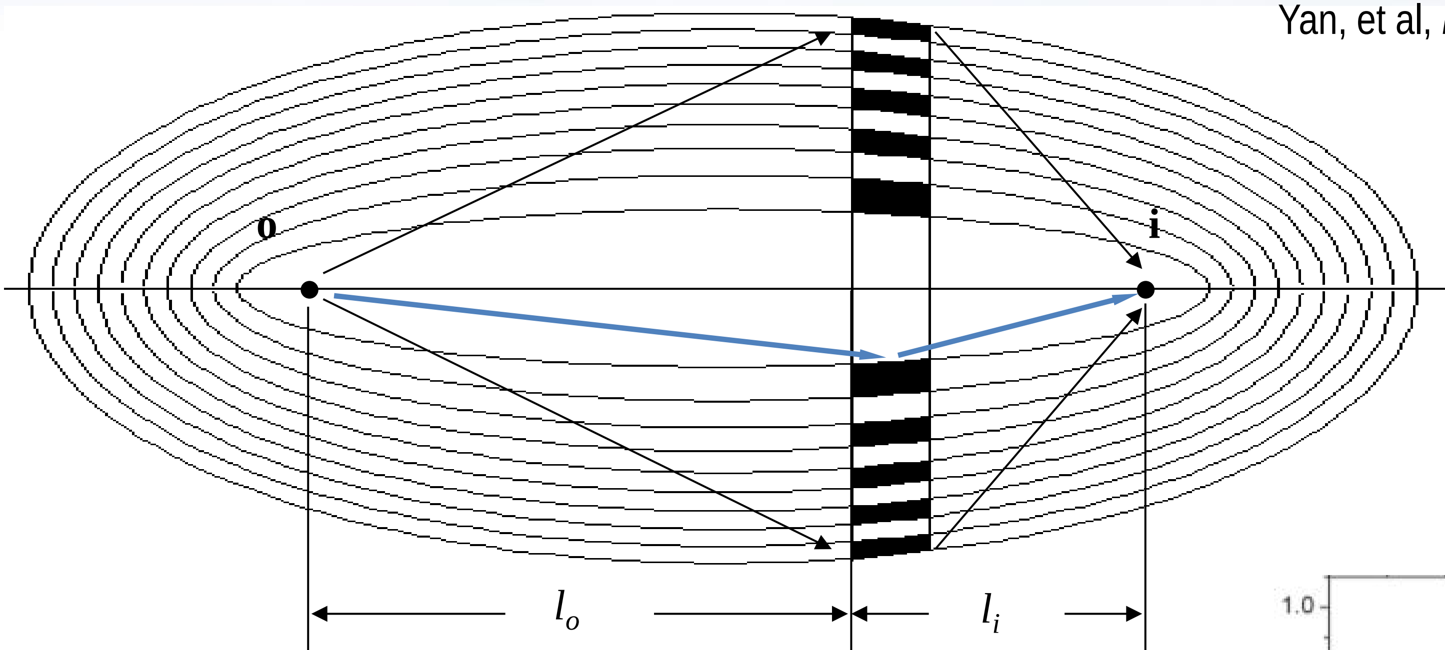
ML Mirror

High-Resolution Nanofocusing X-ray Optics



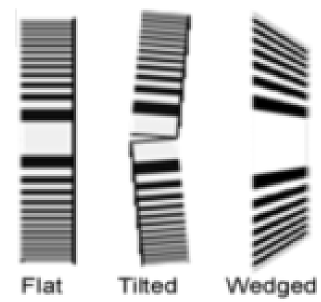
- X-ray focusing optics:
  - Refractive lenses (CRL)
  - Reflective optics (mirrors)
  - Diffractive optics (FZP, MLL, ML mirror)
- Diffractive optics are most promising optics for sub-10 nm focusing
- Fresnel zone plates (FZP) for sub-10 nm focusing of soft x-rays
- Multilayer Laue Lens (MLL) and Multilayer mirrors (MLM) for sub-10nm focusing of hard x-rays

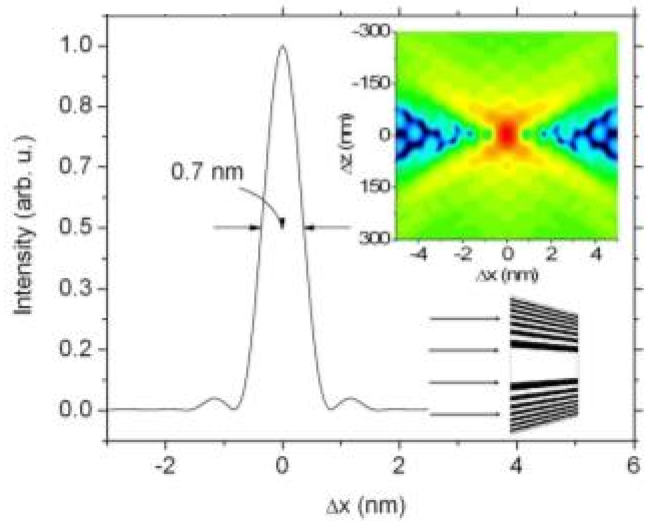
Ideal Focusing – Reflection from Concentric Ellipses



Yan, et al, *Phys Rev B* 76 115438 (2007)

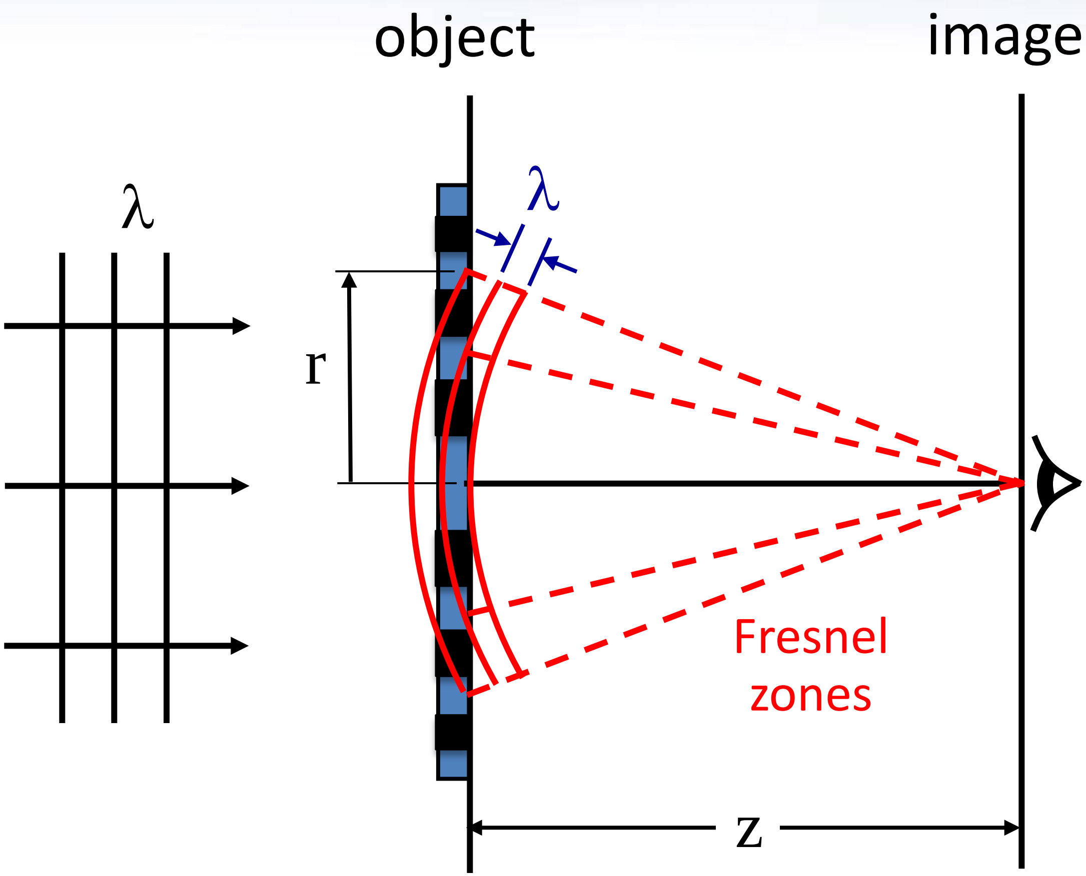
MLL types



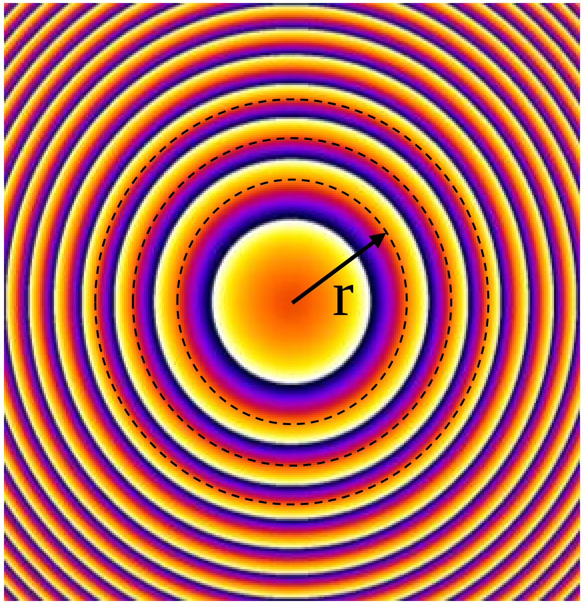


- **<u>MLL</u>**: No hard theoretical limit prevents hard x-rays from being focused to 1-nm by MLL method.
- To achieve 1-nm x-ray focus with high efficiency, **wedged** MLL's are required.
- Ideal MLL is a slice of concentric ellipses involving curves zones

Fresnel Phase Factor → Fresnel Zones

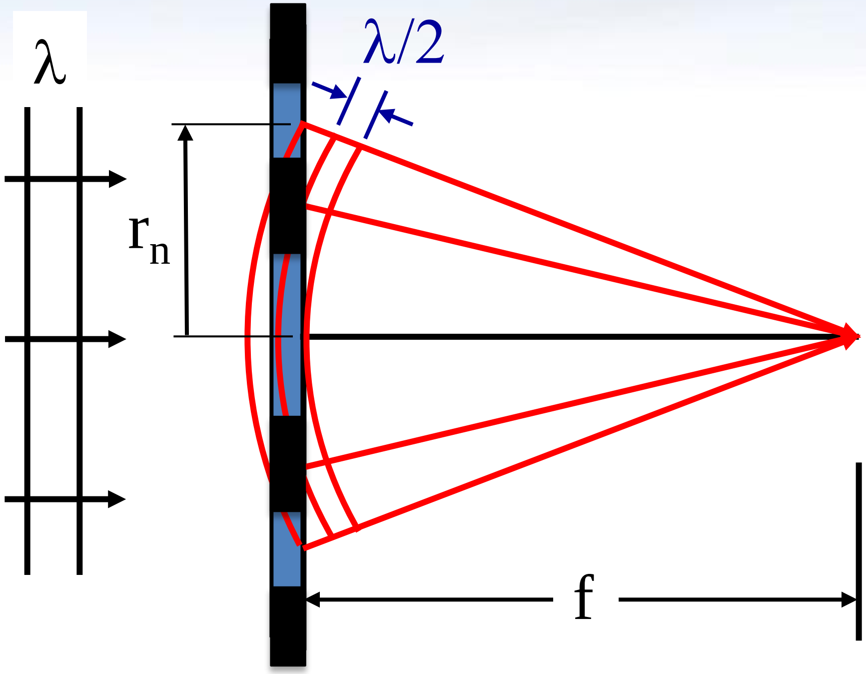


$$e^{-\frac{i\pi}{\lambda z}\left(x^2+y^2\right)} = e^{-\frac{i\pi}{\lambda z}r^2}$$



$$z^2 + r_n^2 = (z + n\lambda)^2 \Rightarrow r_n^2 = 2n\lambda z + (n\lambda)^2 \sim 2n\lambda z \Rightarrow e^{-\frac{i\pi}{\lambda z}r^2} = e^{-i2\pi n}$$

Fresnel Zone Plate (FZP)



Half-period zone radii:

$$r_n^{\,2} = n\lambda f + (n\lambda/2)^2$$

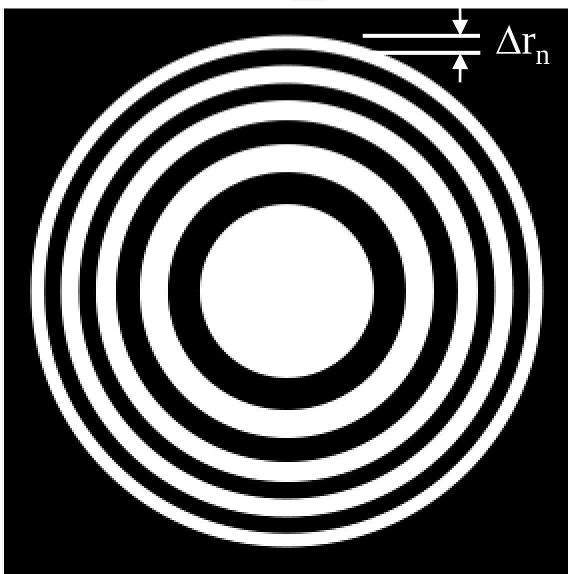
when $f \gg n\lambda$:

$$r_n^{\,2} \simeq n\lambda f$$

outermost zones:

$$(r_n+\Delta r_n)^2 - r_n^{\,2} = \lambda f$$

$$\Rightarrow \boxed{f = 2r_n\Delta r_n/\lambda}$$



outermost zone width

Amplitude FZP: out-of-phase (n=odd) zones are made of absorbing material

Phase FZP: out-of-phase (n=odd) zones are made to provide π phase shift => more efficient

Example of Phase ZP for Hard X-rays

- Using index of refraction for X-rays, $n = 1 - \delta - i\beta$:

$$\delta = 1.35 \times 10^{-6} \lambda[A]^2 \rho[g/cm^3]$$

- For hard X-rays λ = 1.5 A, using Au (ρ = 19.32 g/cm^3) for phase ZP, what is the optimum thickness t ?

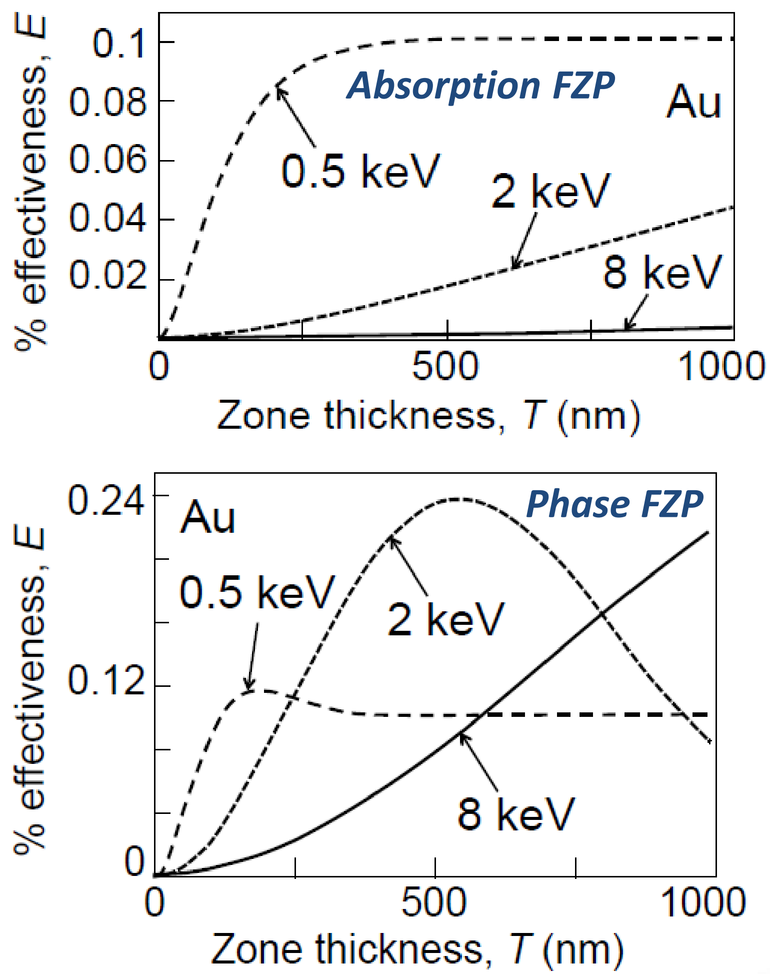
$$\Delta\phi = \frac{2\pi\delta t}{\lambda} = \pi$$

$$t = \frac{\lambda}{2\delta}$$

$$= \frac{1}{2 \times 1.35 \times 10^{-6} \times 1.5 \times 19.32}$$

$$t = 12780\ \text{Å} = 1.278\ \mu m$$

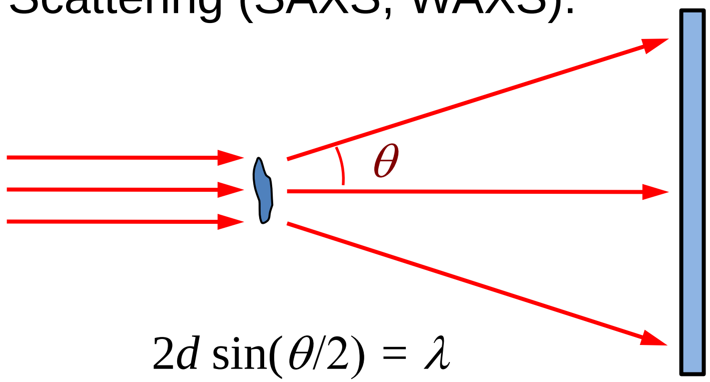
Materials **2012**, *5*, 1752-1773; doi:10.3390/ma5101752



Resolution of Focusing Optics

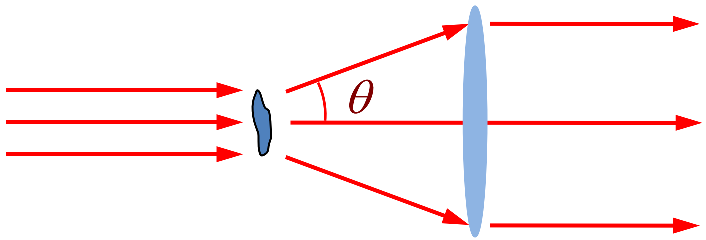
- X-ray lens (FZP) collects scattered or emitted photons and can focus the collected X-rays to a small spot
- smallest achievable focus or *optical resolution* δ (*half d-spacing*) is determined by the largest collection angle θ
- Sine of the largest collection angle by a lens is called *Numerical Aperture* (NA)

Scattering (SAXS, WAXS):

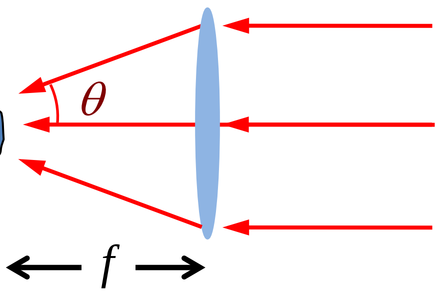


$$2d \sin(\theta/2) = \lambda$$

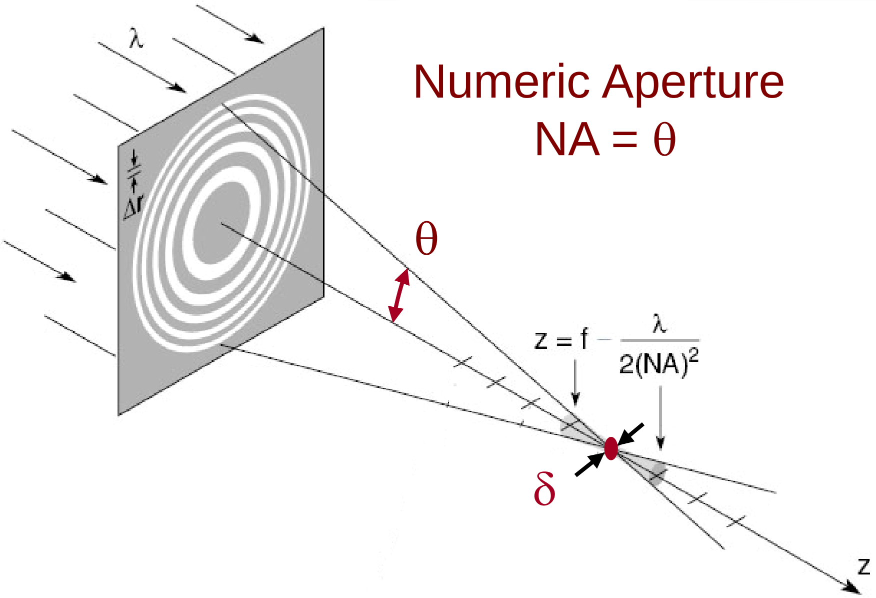
Collecting Scattered X-rays with X-ray lens:



Optical Reciprocity Theorem:



Rayleigh Resolution and Numerical Aperture



Diffraction Limit: $\delta * \theta \sim \lambda/2$

$\Rightarrow$ resolution $\delta \sim 0.5\lambda$ / NA

$\boxed{\text{Rayleigh resolution: } \delta = 0.61\lambda / \text{NA}}$

Bragg's law for outermost 'period':

$2d \sin(\theta/2) \sim \lambda \sim d\,\theta\,; \quad d = 2\Delta r_n$

$\Rightarrow \quad \delta \sim \Delta r_n$

$\boxed{\delta = 1.22\Delta r_n}$

Example: X-ray $\lambda = 1$ A, resolution $\delta = 10$ nm

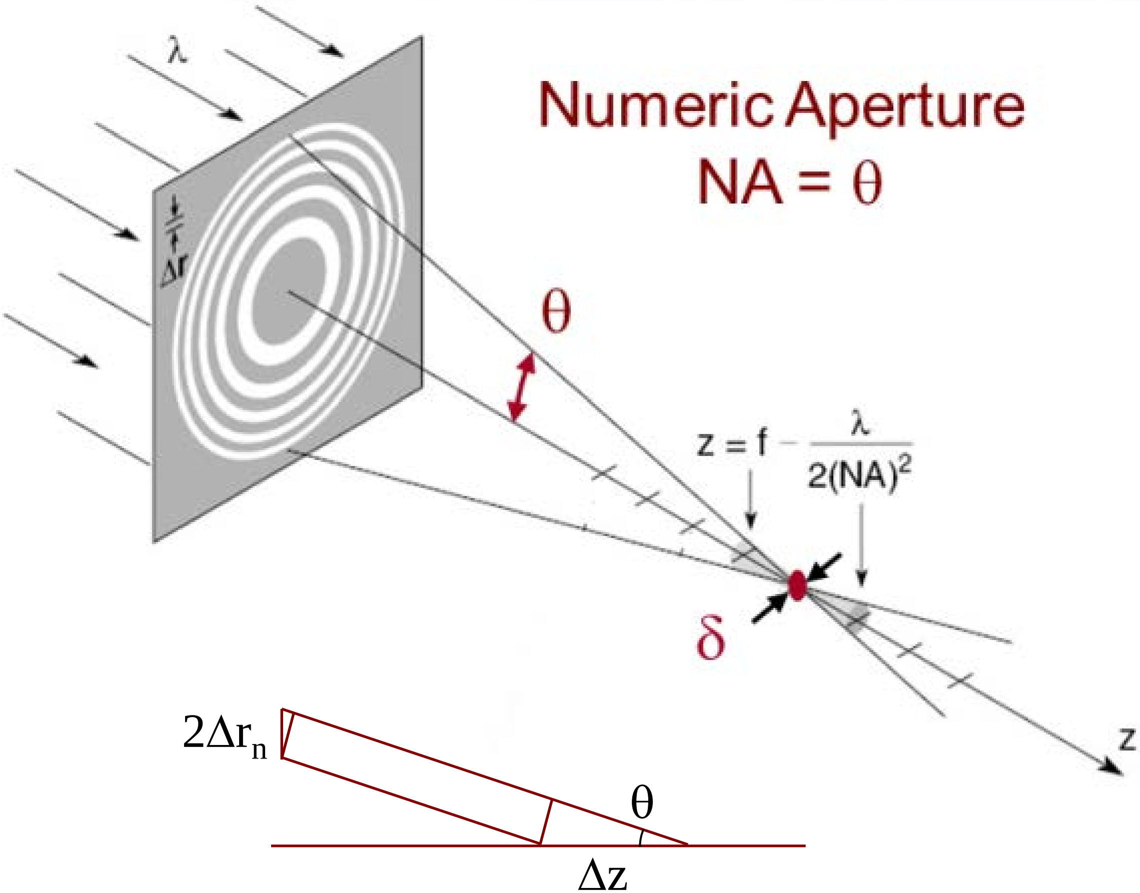
Numerical aperture:

$\text{NA} = 0.61 \times 0.1 / 10 = 0.006$ radians

$= 6$ mrad

Numerical aperture is small for x-rays, even for nm-scale focusing optics

Depth of Focus



Depth of focus Δz is the projection along the θ direction of the outermost period onto optical axis:

$$\Delta z = 2\Delta r_n \cos\theta \,/\, \sin\theta$$

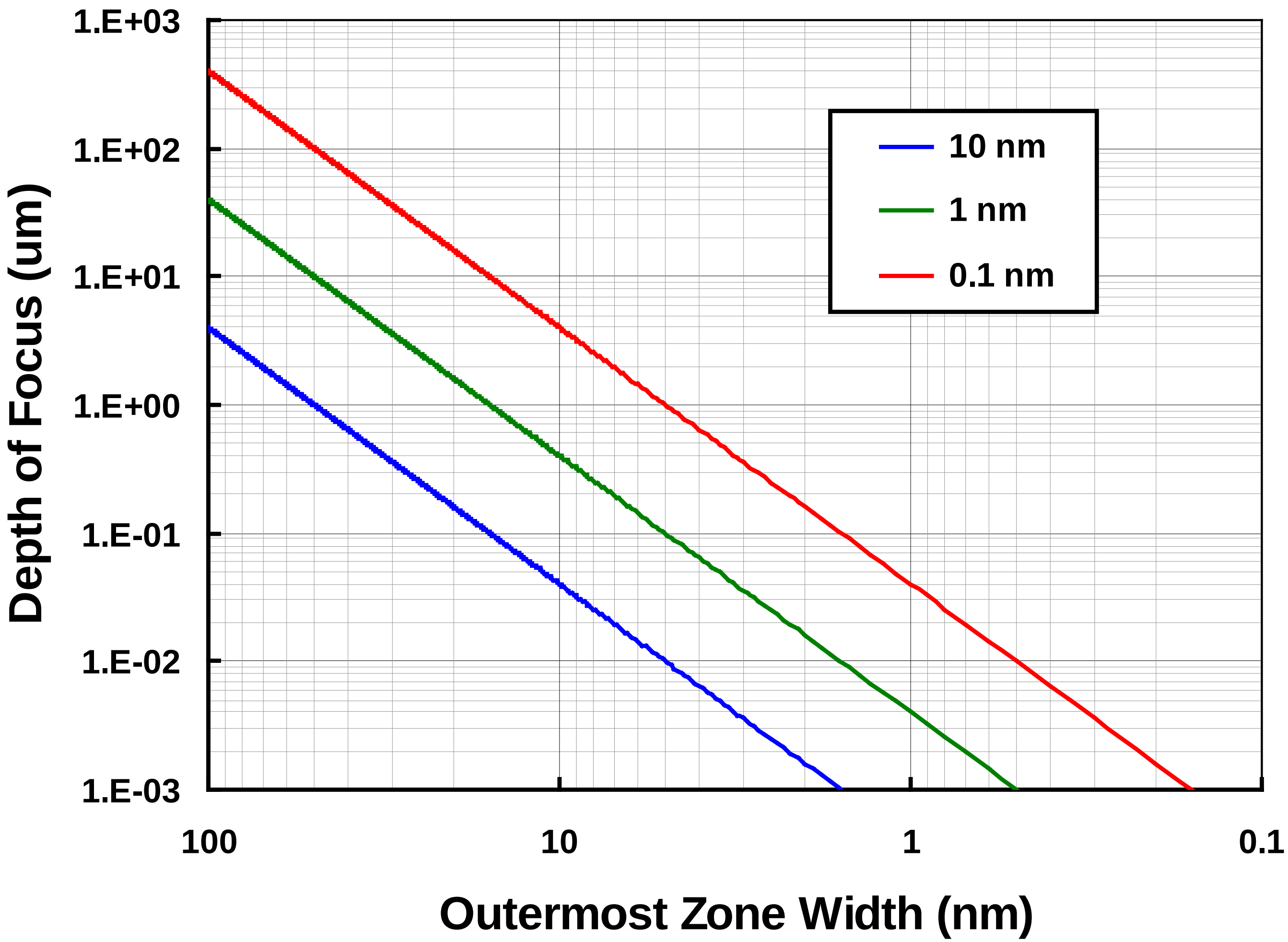
$$\Rightarrow \boxed{\begin{aligned}\Delta z &= 2\Delta r_n \,/\, \theta \\ &= 4(\Delta r_n)^2 / \lambda\end{aligned}}$$ for FZP

Using $\Delta r_n = \dfrac{\lambda}{2\,NA}$

$$\Rightarrow \boxed{\Delta z = \frac{\lambda}{(NA)^2}}$$ for focusing optic in general

➔ Note the quadratic dependence on resolution for a given X-ray energy

Typical Depth of Focus for X-rays



Example: Fresnel Zone Plate (FZP)

FZP design: X-ray $\lambda = 1$ A $= 0.1$ nm,

Resolution $\delta = 1.22\Delta r_n = 20$ nm

$f = 20$ mm

$\Rightarrow$ Outermost zone width:

$\Delta r_n = 20 \text{ nm} / 1.22 = 16 \text{ nm}$

$\Rightarrow$ ZP radius:

$r_n = f\,\lambda / (2\Delta r_n) = 20 \times 0.1 / (2 \times 16) = 0.0625 \text{ mm}$

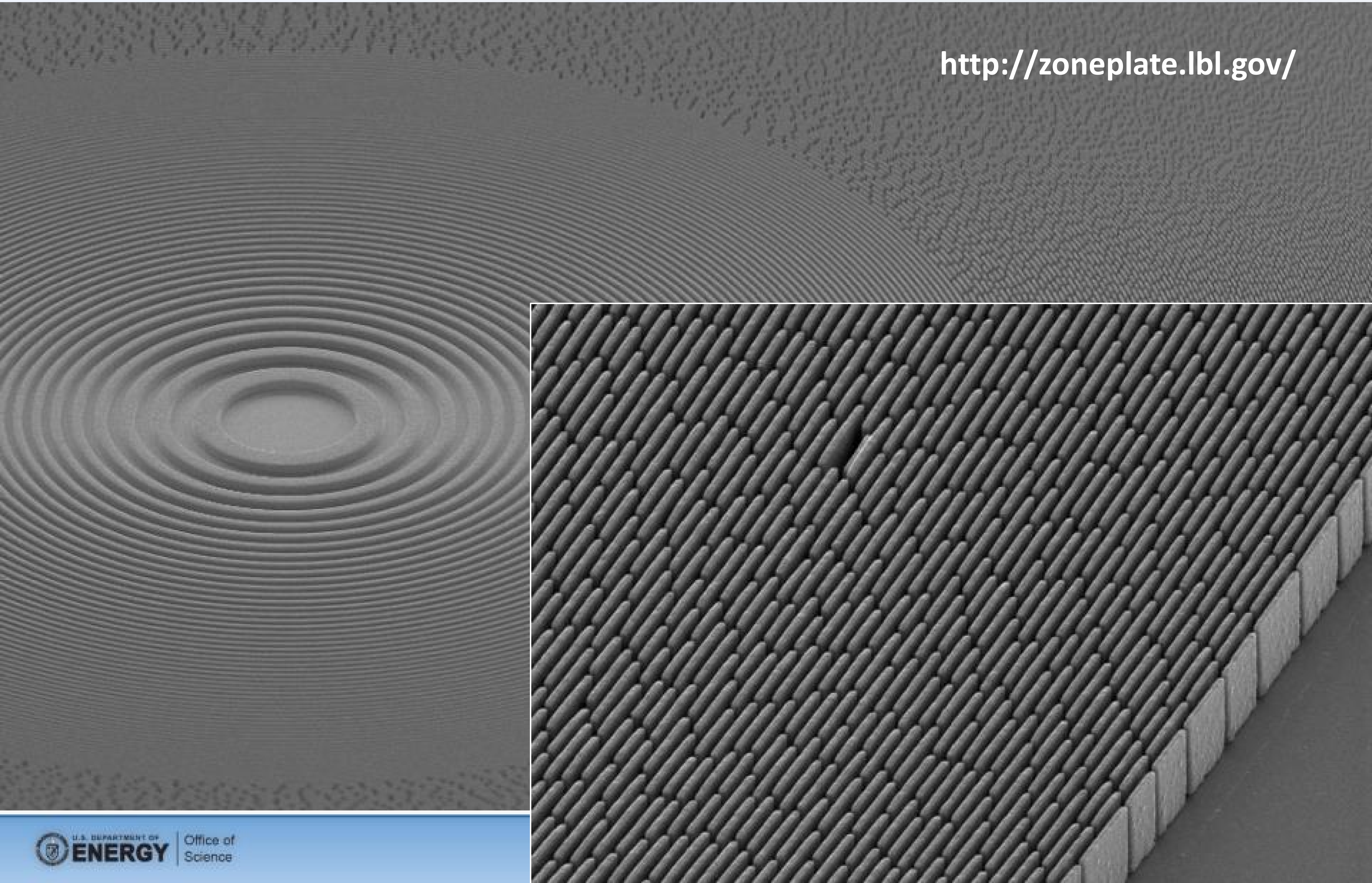
$\Rightarrow$ Number of zones:

$n = r_n^2 / (\lambda f) = 0.0625^2 / (0.1\text{x}10^{-6} \times 20) = 1953$

$\Rightarrow$ Numeric aperture:

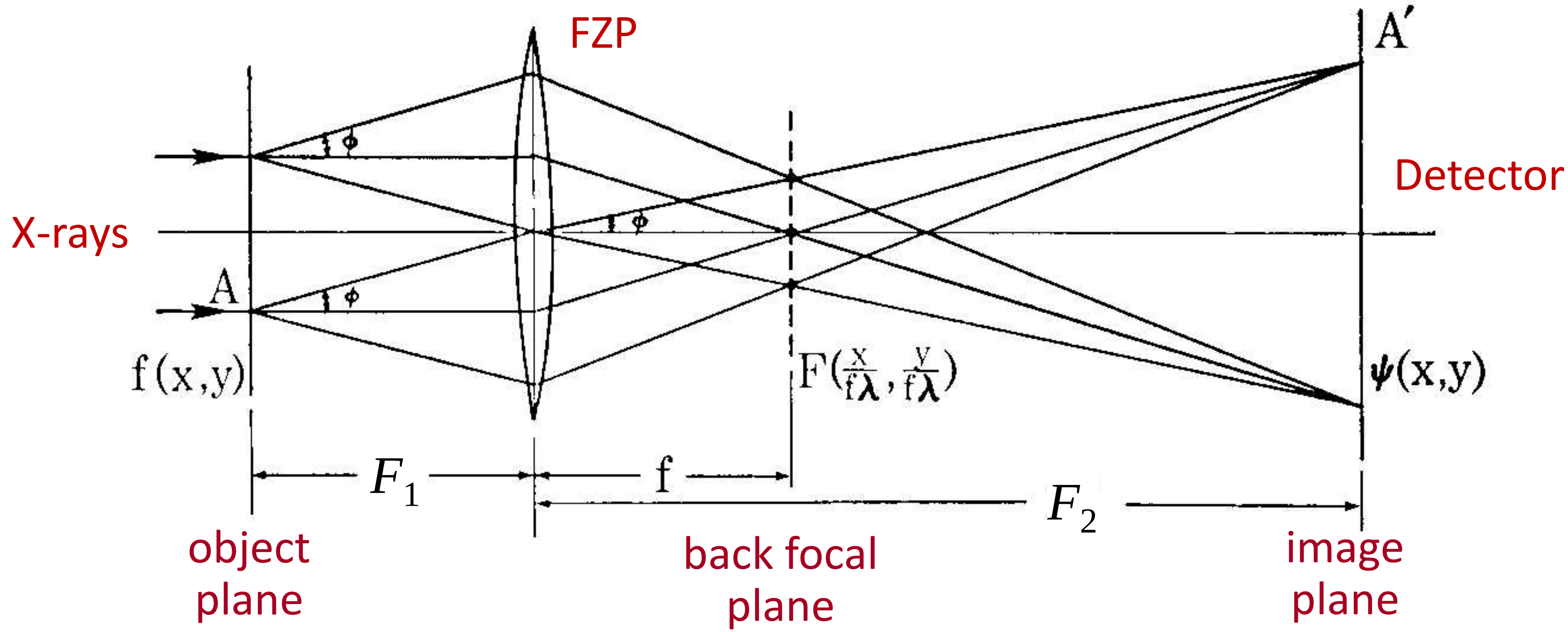
$NA = r_n / f = 0.0625 / 20 = 0.003125 \text{ rad} = 3.125 \text{ mrad}$

Scanning Electron Micrograph of a Zone Plate



http://zoneplate.lbl.gov/

Lens-Based Full-field Transmission X-ray Microscope (TXM)



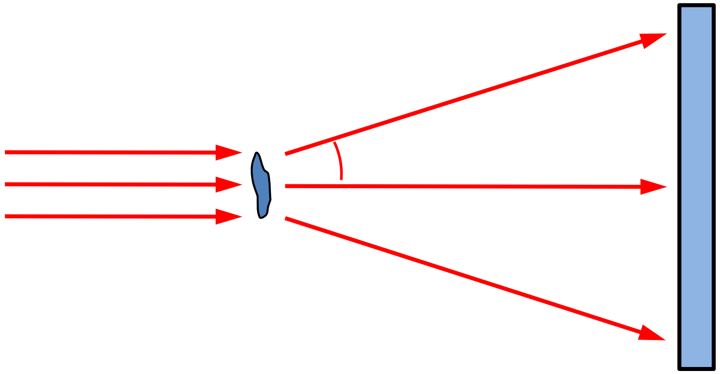
⇒ TXM works just like any transmission microscope by use of lens equation

$$\frac{1}{F_1} + \frac{1}{F_2} = \frac{1}{f}$$

⇒ Magnification $M = F_2 / F_1$

Analogy – Scattering vs. Imaging

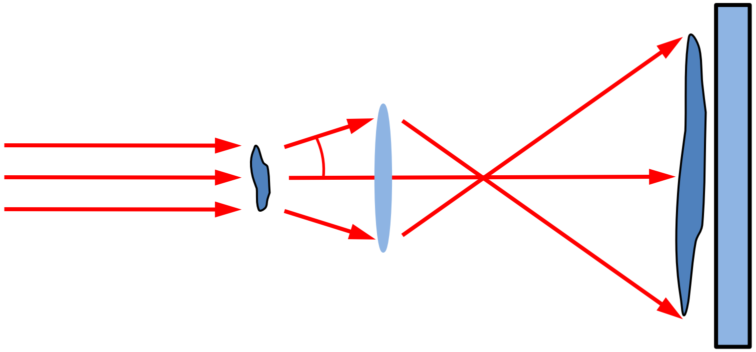
Scattering (SAXS, WAXS, CDI):



$$\frac{1}{F_1}+\frac{1}{F_2}=\frac{1}{f}$$

→ X-ray lens (FZP) collects scattered photons and projects to an enlarged real-space image on image plane as determined by lens equation

Imaging with X-ray lens (TXM):



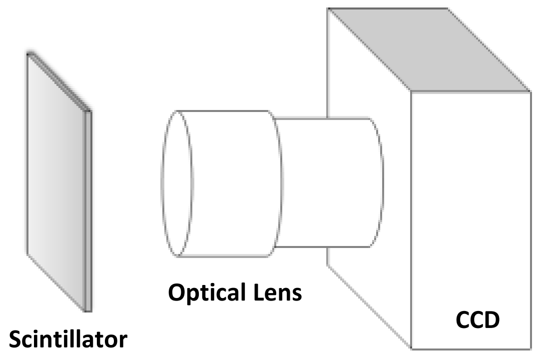
→ Achievable resolution in a TXM is determined by the resolution of the ZP objective lens, or the largest scattering angle collected by the ZP objective

→ Currently Fresnel zone plate resolution is limited by nano-fabrication technology to 20-30 nm for hard x-rays

2D Area Detector for X-rays

- Pixel Array Detector – pixelated sensors (1Kx1K or 2Kx2K) for direct X-ray detection with on-pixel & readout electronics
  - typical pixel size 10-100 um
  - sensitive to single x-ray photon
  - good for scattering experiments

- Lens-coupled CCD – using a scintillator to convert X-rays to visible light, which is then captured by a visible light microscope with an objective lens + CCD
  - typical CCD pixel ~10 um, 1Kx1K or 2Kx2K
  - scintillator x-ray to visible efficiency is low (<1%)
  - overall, good for x-ray microscopic imaging
  - visible light objective 5x to 20x → effective pixel size 2 to 0.5 um on scintillator screen
  - effective pixel size on sample plane is 20 to 5 nm if TXM magnification is M = 100



Example of Transmission X-ray Microscope

<u>ZP Objective</u>: f = 20 mm, Diameter = 0.16 mm, X-ray λ = 0.1 nm

=> NA = (0.16/2)/20 = 40 mrad

=> $\Delta r_n = f\lambda / (2r_n)$ = 20 x 0.1 / 0.16 = 12.5 nm

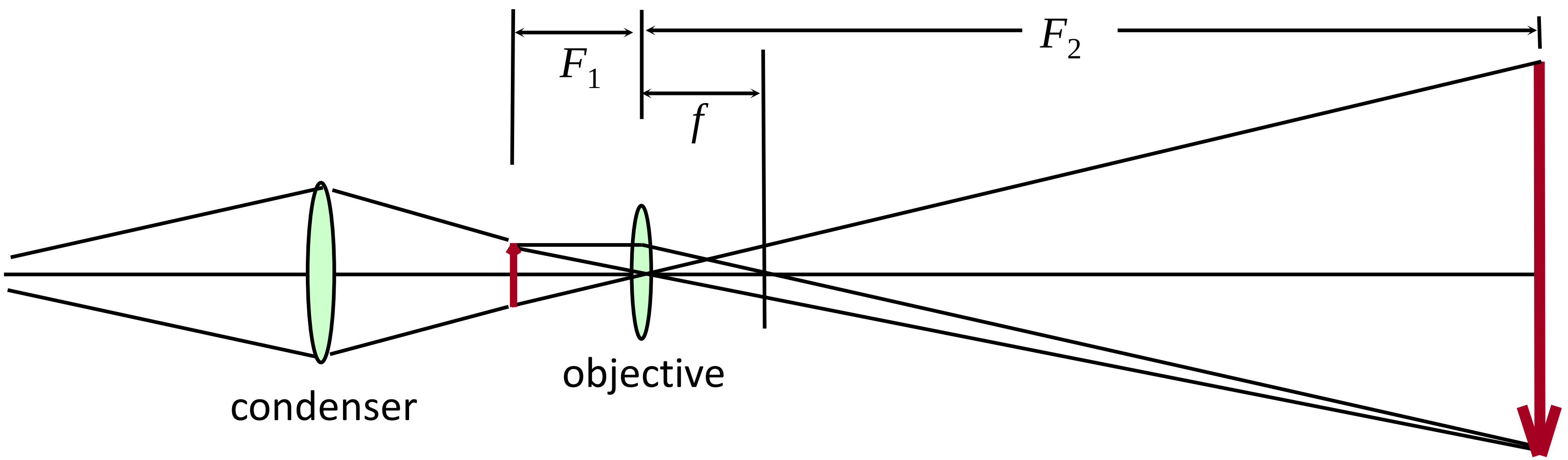
ZP resolution $\delta = 1.22\Delta r_n$ = 1.22 x 12.5 = 15 nm

<u>TXM Settings</u>: Object distance F_1 = 20.2 mm

=> Image distance F_2 = 2020 mm

=> Magnification $M = F_2/F_1$ = 100

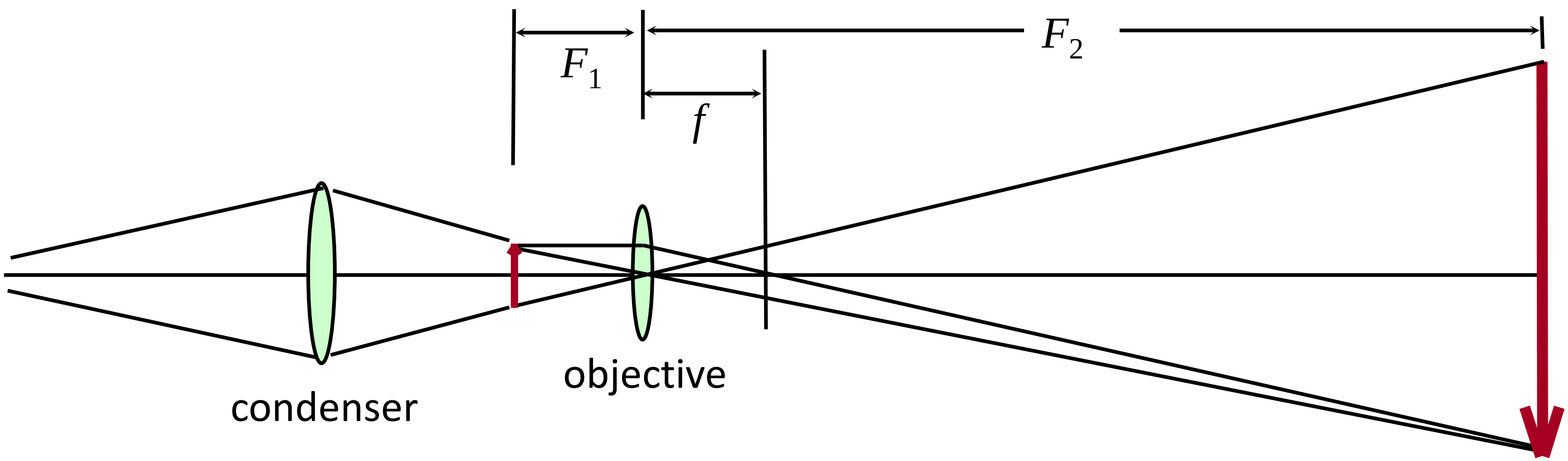
$$\boxed{\frac{1}{F_1}+\frac{1}{F_2}=\frac{1}{f}}$$



Oversampling

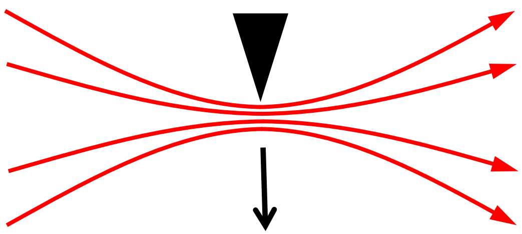
Oversampling Ratio: defined as the number of physical pixels used in imaging measurement within the optical resolution of the imaging system; Oversampling provides better measurements statistics in low-contrast cases

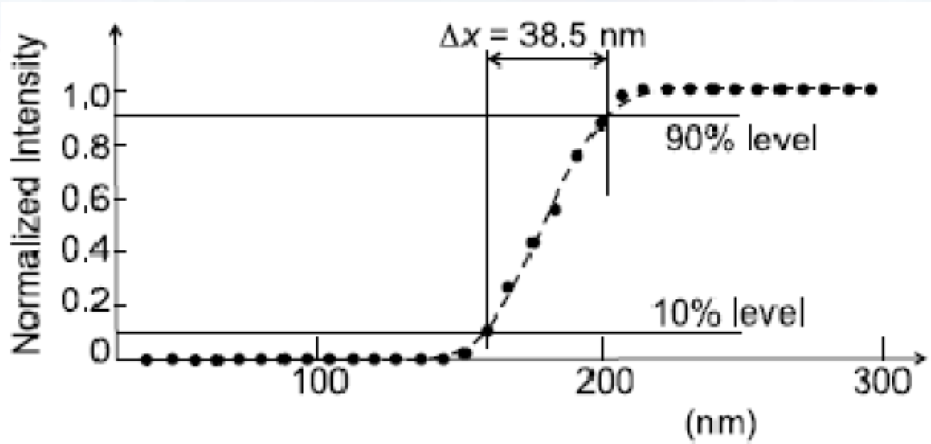
Back to our example: assuming detector 2k x 2k CCD with pixel size 10 um
objective lens 20x
=> Effective pixel size on scintillator = 10/20 = 0.5 um
=> Effective pixel size on sample = 0.5 um / 100 = 5 nm
Since optical resolution = 15 nm, the oversampling ratio in this case is 3x



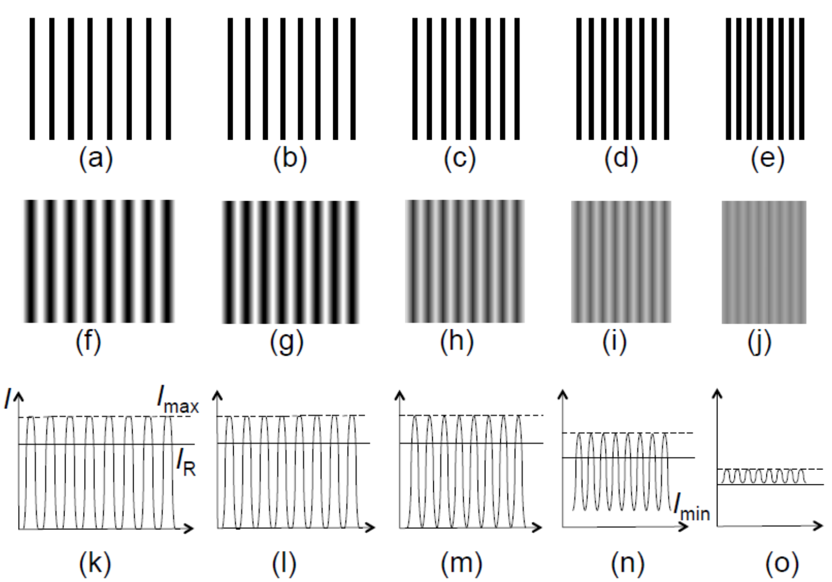
Experimental Determination of Resolution

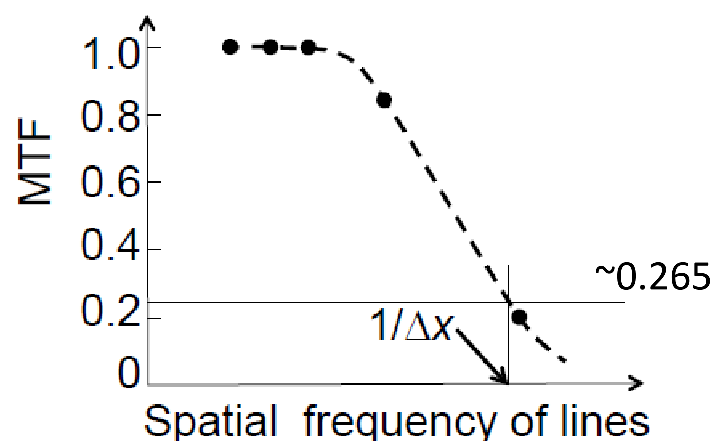
Knife-edge scan:
→ scan a sharp edge across the focus





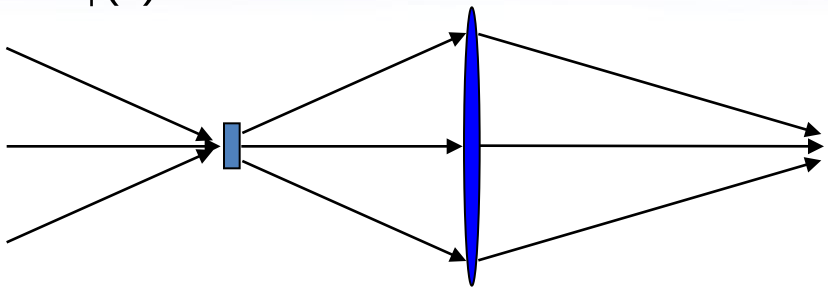
Modulation Transfer Function (MTF): a special case of *Power Spectrum Analysis*
→ plot observed contrast as a function of spatial frequency





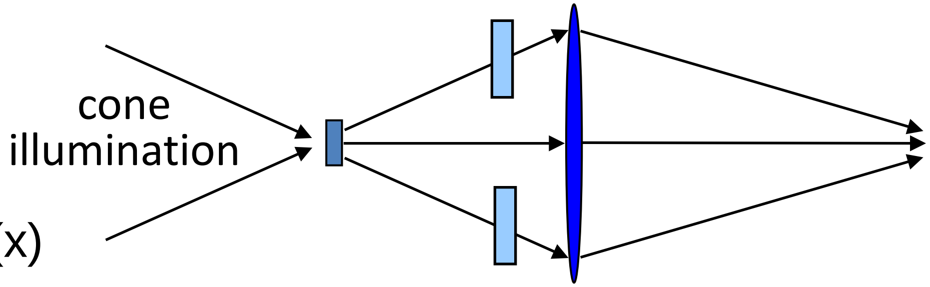
Zernike Phase Contrast: Principle

For pure phase object: $F(x) = e^{i\phi(x)} \sim 1 + i\phi(x)$



=> No absorption contrast: $|F|^2 \sim 1$

Zernike method in visible optics:



=> $F'(x) \sim \pm i + i\phi(x)$
=> Phase contrast: $|F'|^2 \sim 1 \pm 2\phi(x)$

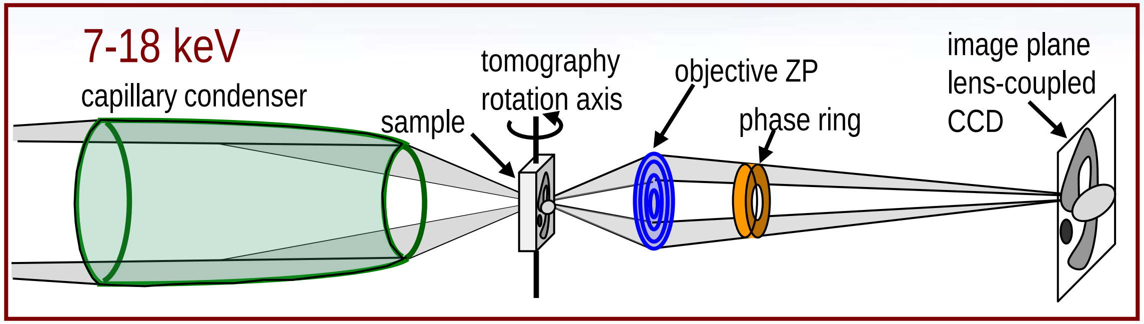
phase plate: $\Delta\phi = \pm\pi/2$
in zeroth-order (un-scattered photons)

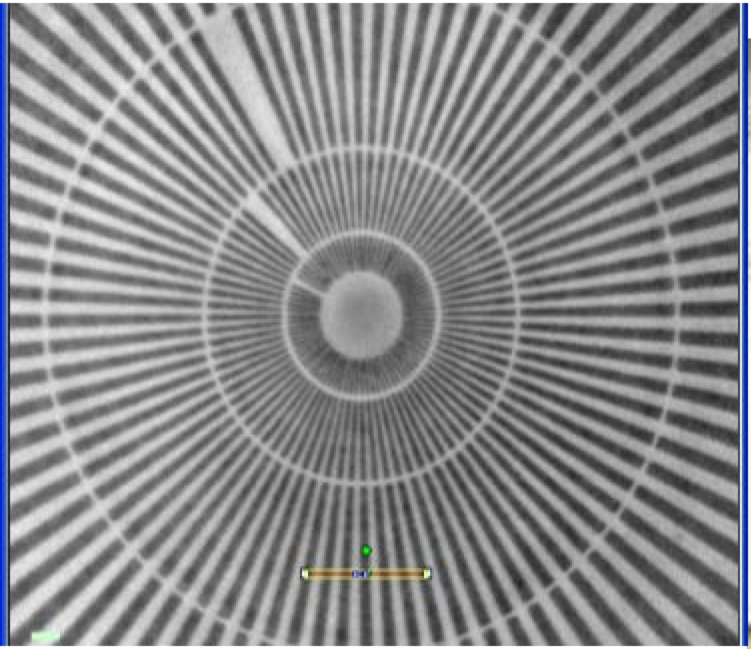
X-ray Zernike Phase Contrast:

Soft X-rays: Schmahl et al. (1995). BESSY, 0.5 keV.
Hard X-rays: Kagoshima et al. (2001). SPring8, 10keV.

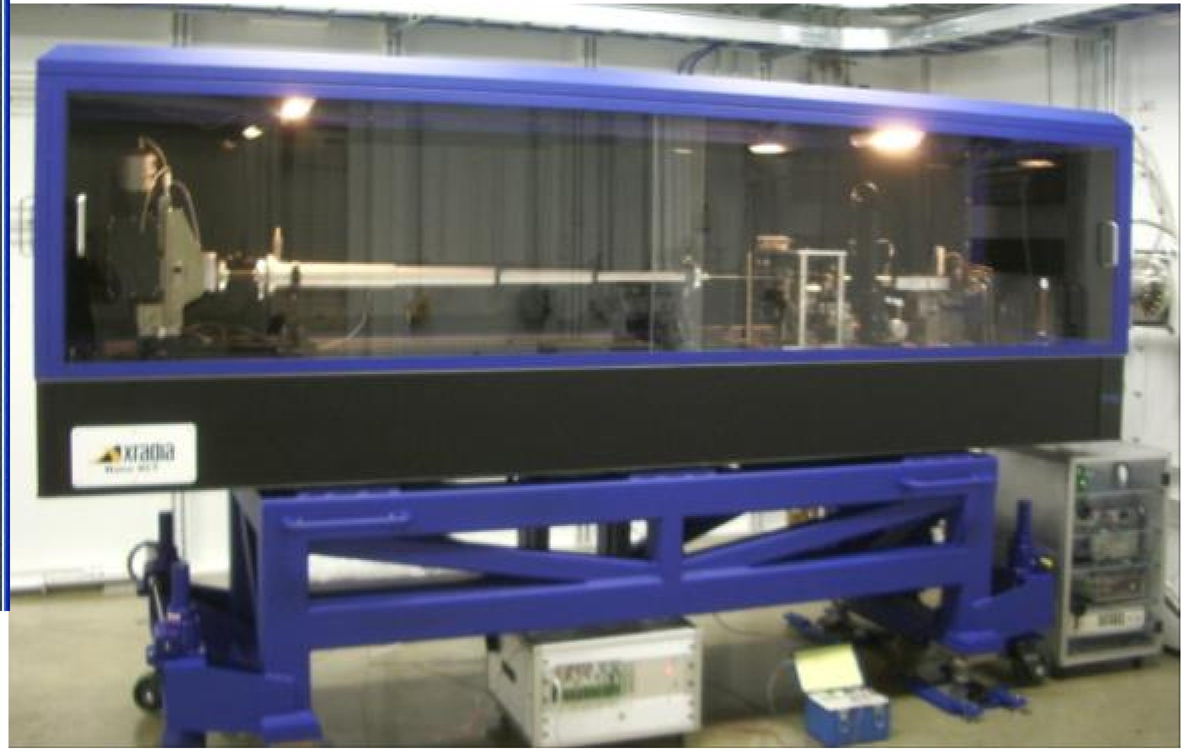
Transmission X-ray Microscopy (TXM) by Xradia (now Zeiss)

- Absorption & Zernike phase-contrast
- 30 nm FZPs, 3D tomography
- Exposure time: 50ms - seconds

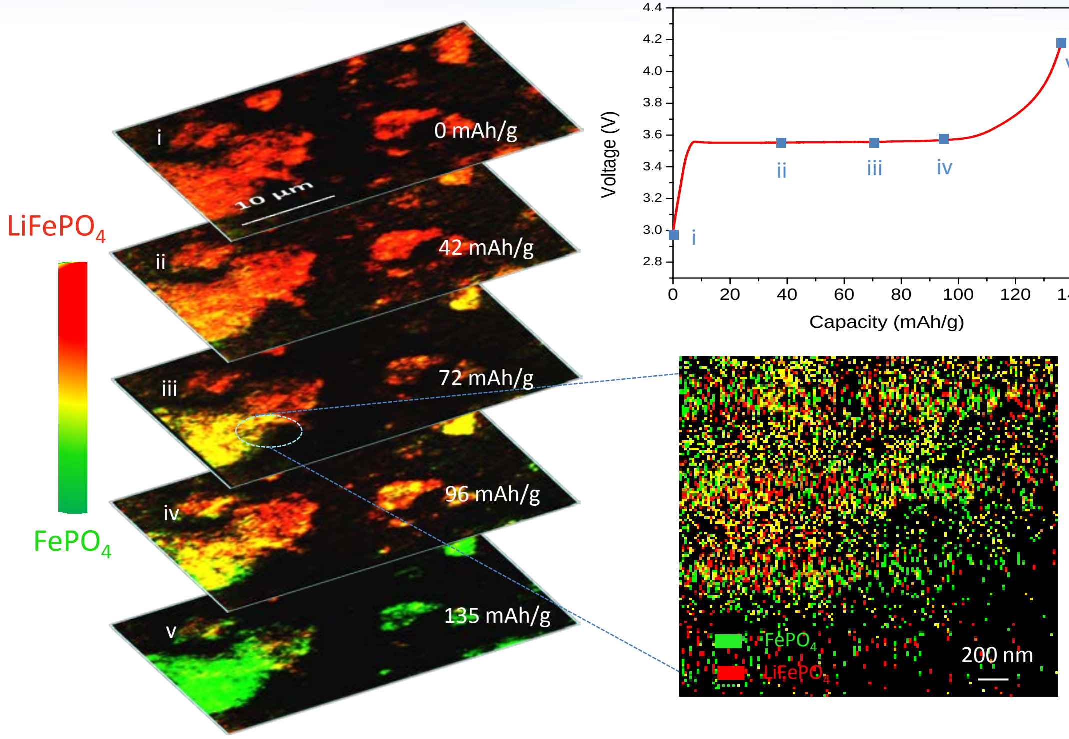


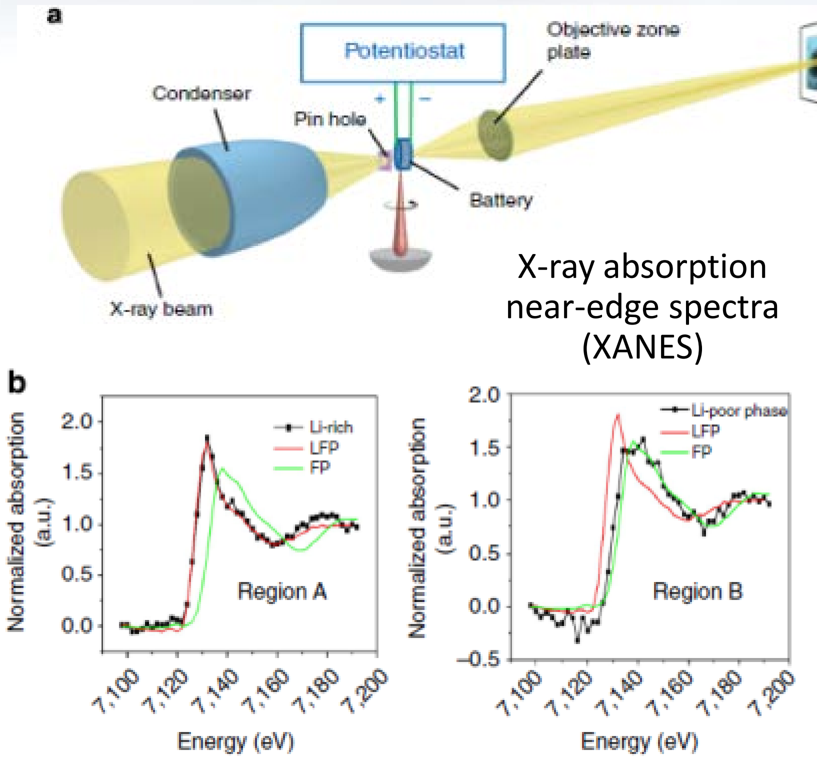


10 keV Zernike Phase Contrast



In-Operando 2D Chemical Mapping of Multi-particle Li-Battery Cathode



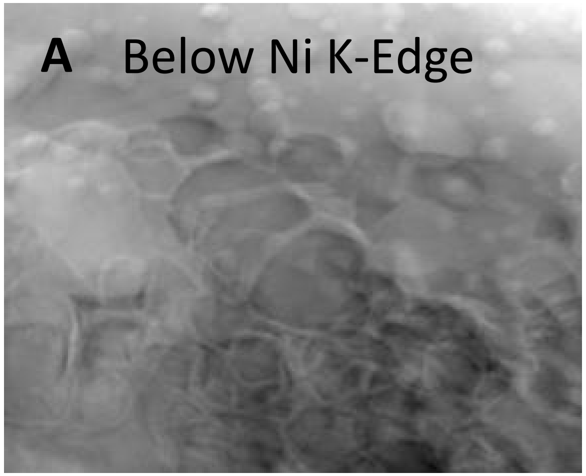


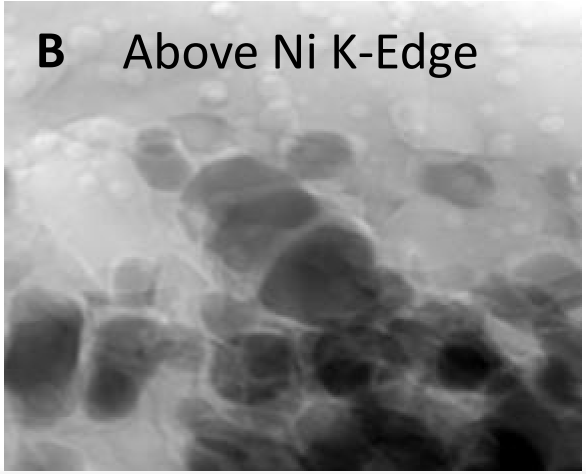
In operando 2D chemical mapping of multi particle $LiFePO_4$ cathode during fast charging. The closeup frame shows that as the sample charges, some regions become completely delithiathed ($FePO_4$, green) while others remain completely lithiated (red). This inhomogeneity results in a lower overall battery capacity than can be attained with slower charging, where delithiation occurs more evenly throughout the electrode

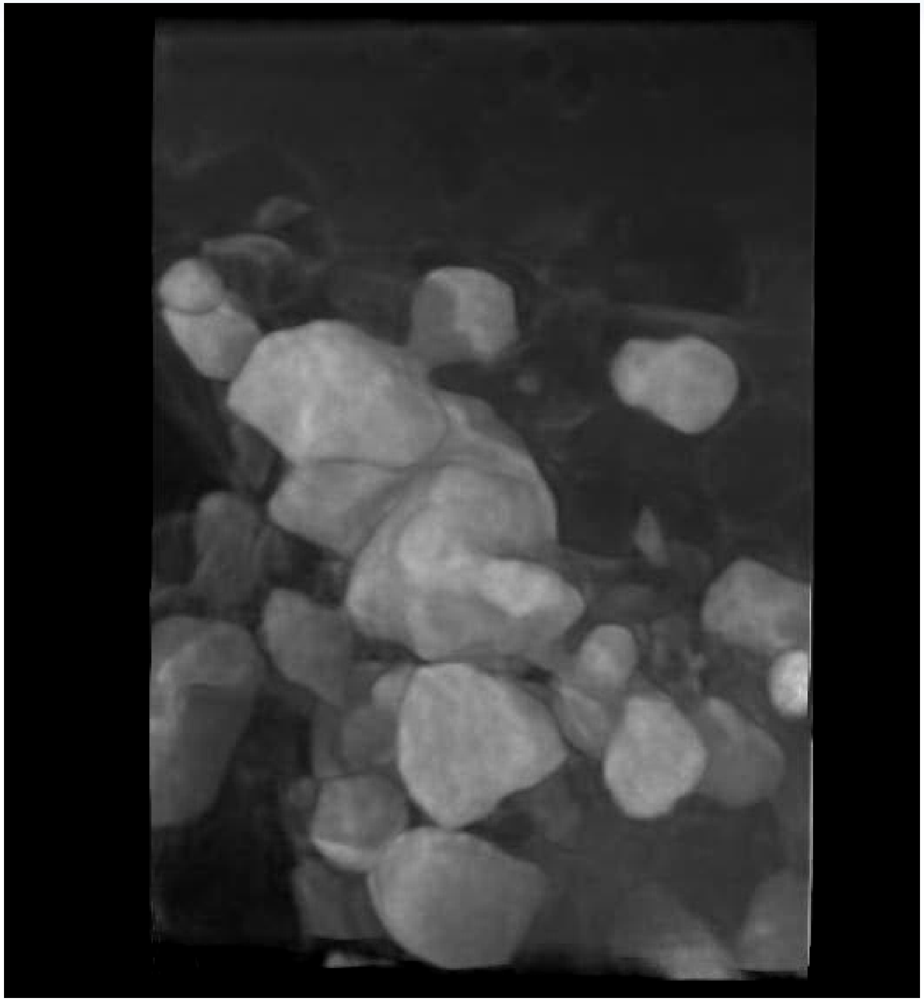
"In operando tracking phase transformation evolution of lithium ion phosphate with hard X-ray microscopy", Jiajun Wang, Yu-chen Karen Chen-Wiegart, Jun Wang, ***Nature Comm.***, **DOI:10.1038/ncomms5570, 2014**

3D Element-Specific Nano-Tomography

Solid Oxide Fuel Cell (SOFC) Pore Structures: Yittria Stablized Zirconia YSZ (electrolyte) / NiO (anode)







W. Chiu (U. Conn.), Y.S. Chu (NSLS-II, BNL)

3D TXM Observation of Nanoscale Changes in Rechargeable Battery Electrode *In-Operando*

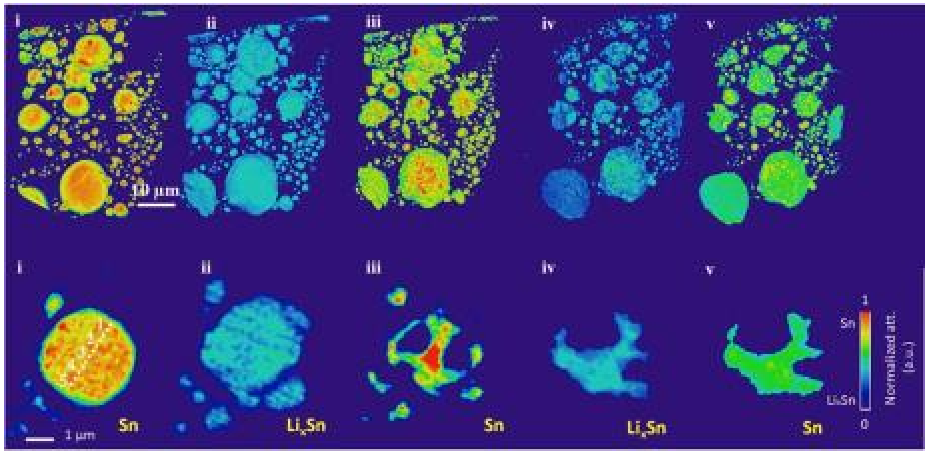
Jiajun Wang, Yu-chen Karen Chen-Wiegart, Jun Wang. *Angewandte Chemie 53, 4460* (2014)

Scientific Achievement

Made the first 3D nano-imaging observations of microstructural evolution of a lithium-ion battery anode during electrochemical reactions in a real battery cell as it discharges and recharges

Significance and Impact

Understanding the mechanism leading to electrode degradation points to new ways to engineer battery materials to increase the capacity & lifetime of rechargeable batteries

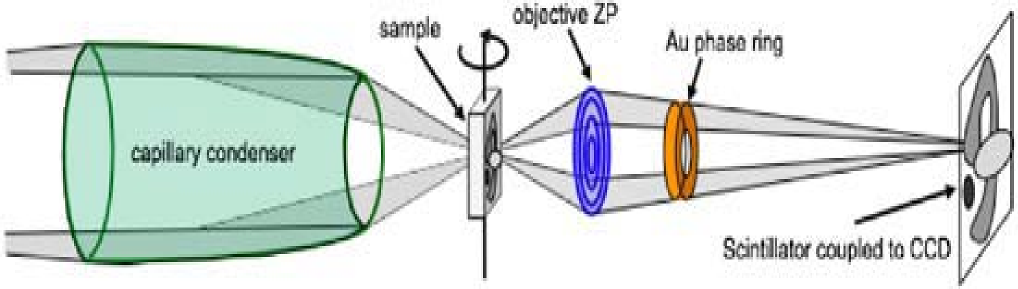


Battery materials during charge and discharge: a) 3D morphological evolution of Sn particles during the first two lithiation–delithiation cycles; b) Pseudo cross-sectional images of a single Sn particle during the first two cycles. The particle shows severe fracture and pulverization at the initial stage of cycling, but stays mechanically stable afterwards while the electrochemical reaction still proceeds reversibly. The color scale corresponds to the normalized linear attenuation coefficient, representing direct visualization of the chemical composition.

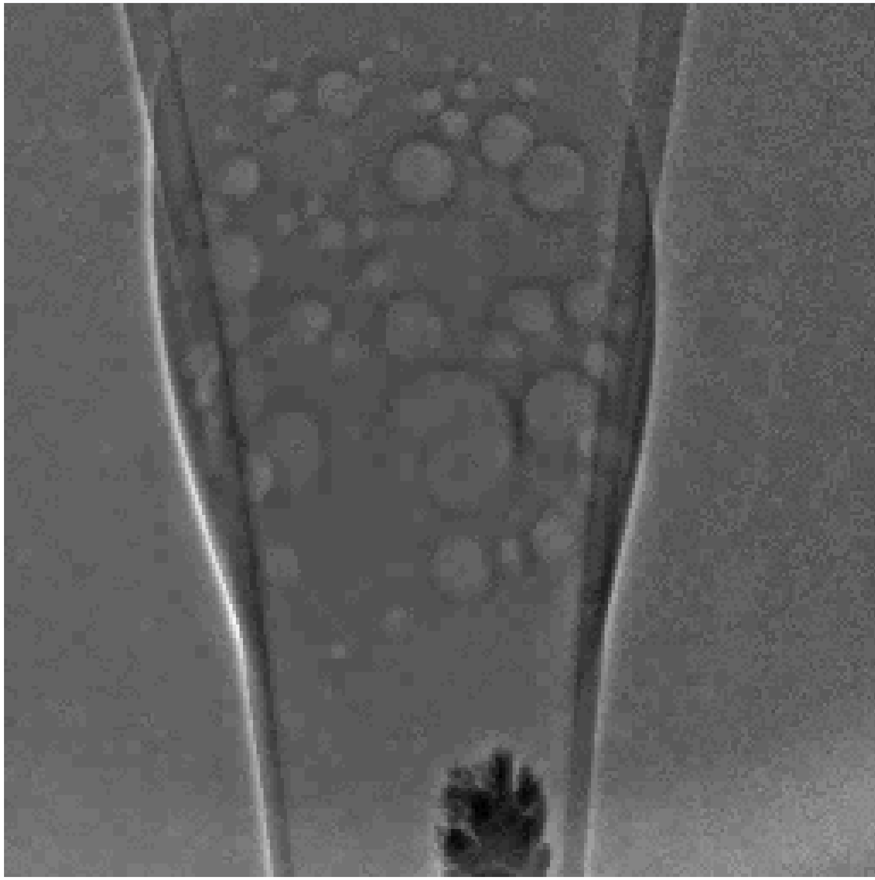
Research Details

- Built a fully functioning battery cell with all three battery components -- the electrode being studied, a liquid electrolyte, and the counter electrode -- supported by relatively transparent materials to allow transmission of the x-rays, and contained within a quartz capillary measuring one millimeter in diameter.
- Produced more than 1400 2D x-ray images of the anode material with a resolution of approximately 30 nanometers, which were later reconstructed into 3D images.

Real Time Nano-Imaging of Electrochemical Growth



- Modern x-ray lenses (Fresnel zone plates) and bright x-ray sources enable x-ray microscopic imaging at nano-scale spatial resolutions
- Example: In-situ studies of dendritic growth of Cu in $CuSO_4$ solution by electrical potential (APS 32-ID, 8 keV)
  - Frame rate: 100 msec/frame
  - TXM FOV: 22 μm



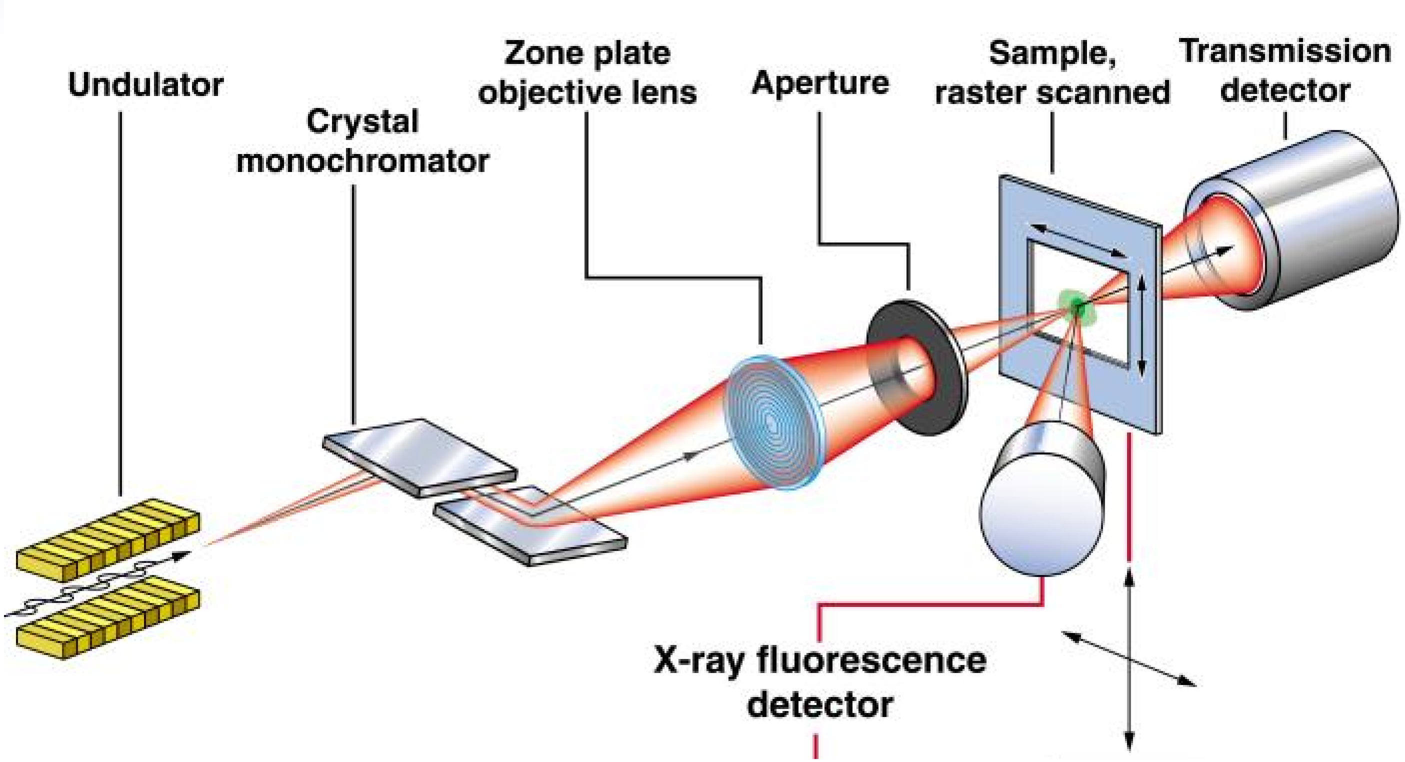
J. Yi, S. Wang (ANL), Y-K. Hwu (Acad. Sinica, Taiwan), J. H. Je (POSTECH, Korea), Y. S. Chu (NSLS-II, BNL)

*APPLIED PHYSICS LETTERS **97**, 033101 (2010)*

Today's Lecture

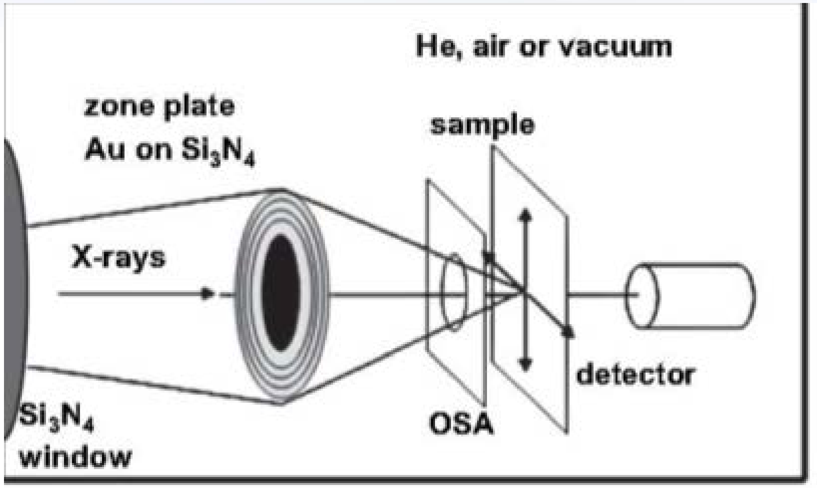
- Introduction to X-ray Imaging/Microscopy
  - absorption & phase contrast
  - Fresnel wave propagation
- Lens-based X-ray Microscopy
  - focusing optics: refractive lens, reflective & diffractive optics
  - Fresnel zone plate: numerical aperture, resolution, magnification
  - full-field transmission x-ray microscope
  - scanning x-ray microscopy
- Lens-less X-ray Microscopy
  - coherent diffraction imaging/microscopy
  - coherent diffraction ptychography
- Application Examples in Each Section

Scanning X-ray Microscope



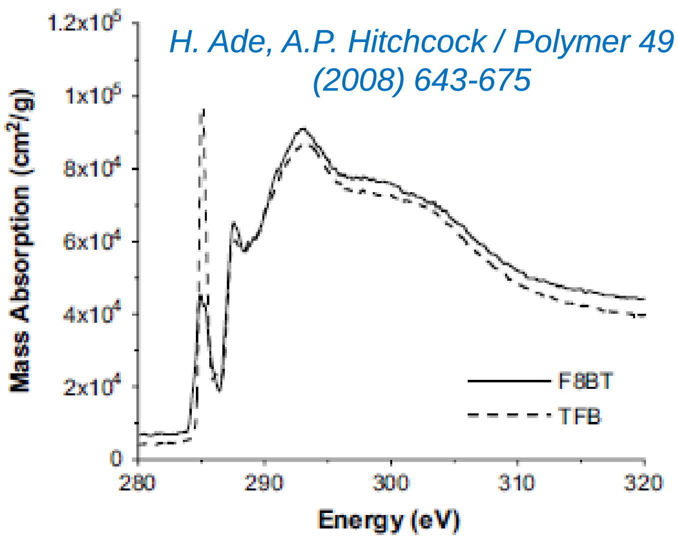
- In scanning X-ray microscopy or micro/nanoprobe, contrast is obtained as a function of scanning position of the sample
- Contrast may be almost anything that can be measured, *e.g.* x-ray transmission (STXM), x-ray fluorescence (SFXM), diffraction (micro-diffraction), scattering signal, total electron yield, ...

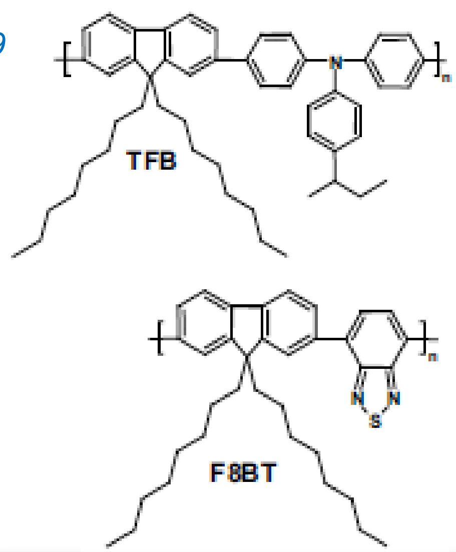
STXM Study of Fuel Cell & Organic Materials

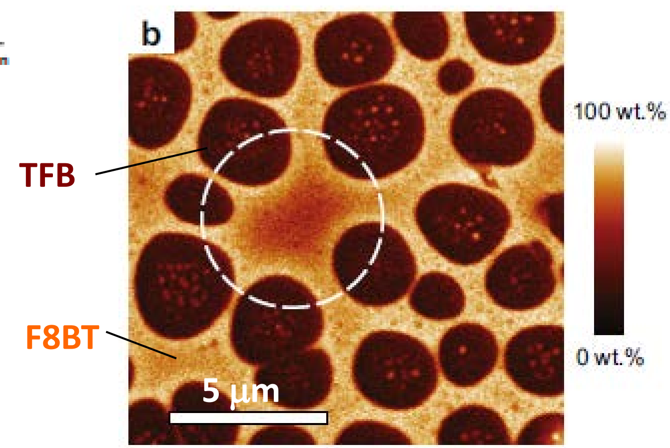


Scanning transmission x-ray microscope (STXM)

- Organic photovoltaic (OPV) devices fabricated from conjugated polymers are potentially economic alternatives to present device technologies.
- Enriched and sharp TFB domain interface with F8BT leads to efficient charge transport and promotes charge capture and recombination → Efficient performance of TFB:F8BT-based LEDs.





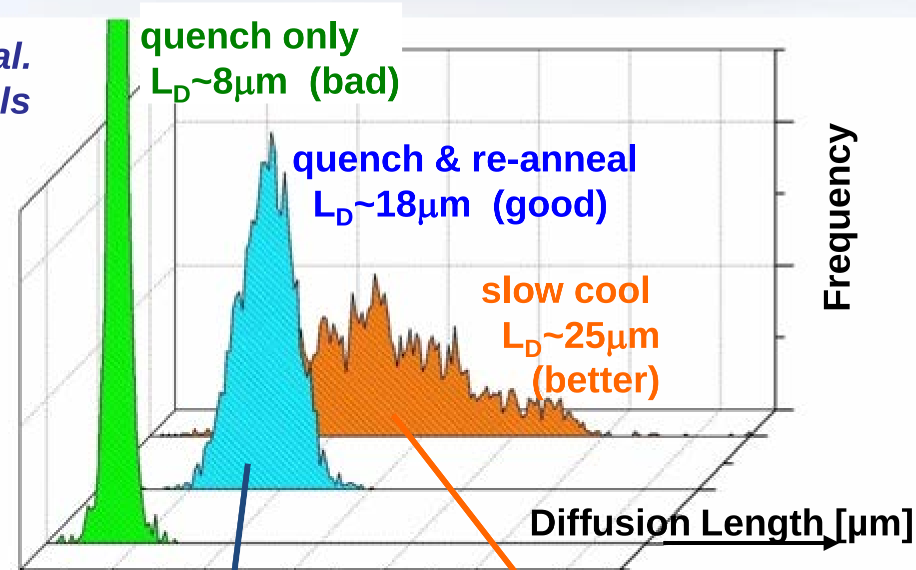


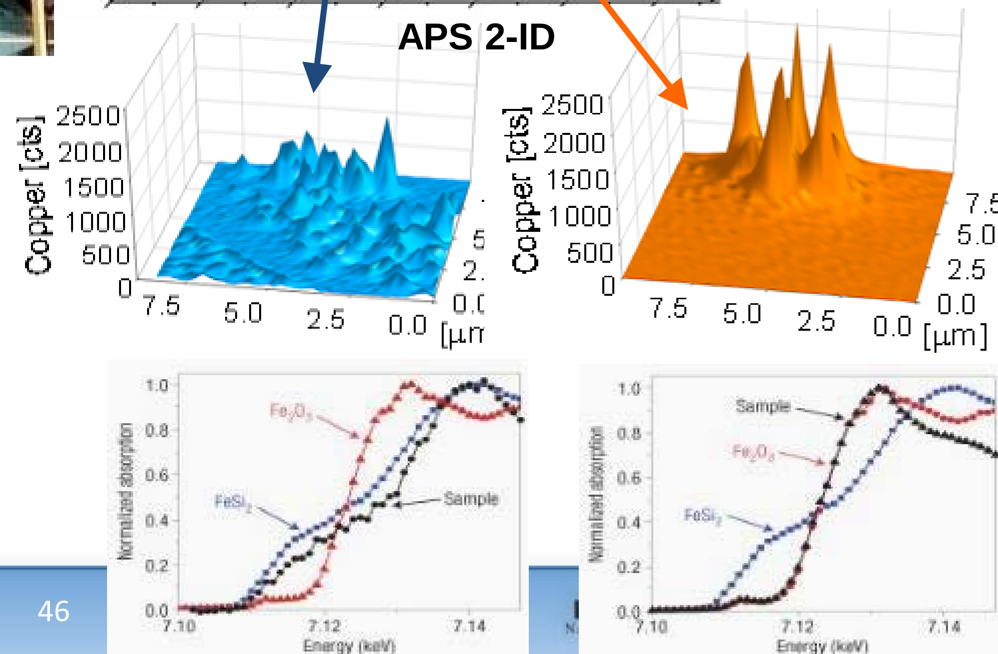
F8BT composition map

Defect Engineering for Less-Costly Solar Cells

Multicrystalline (mc-Si)

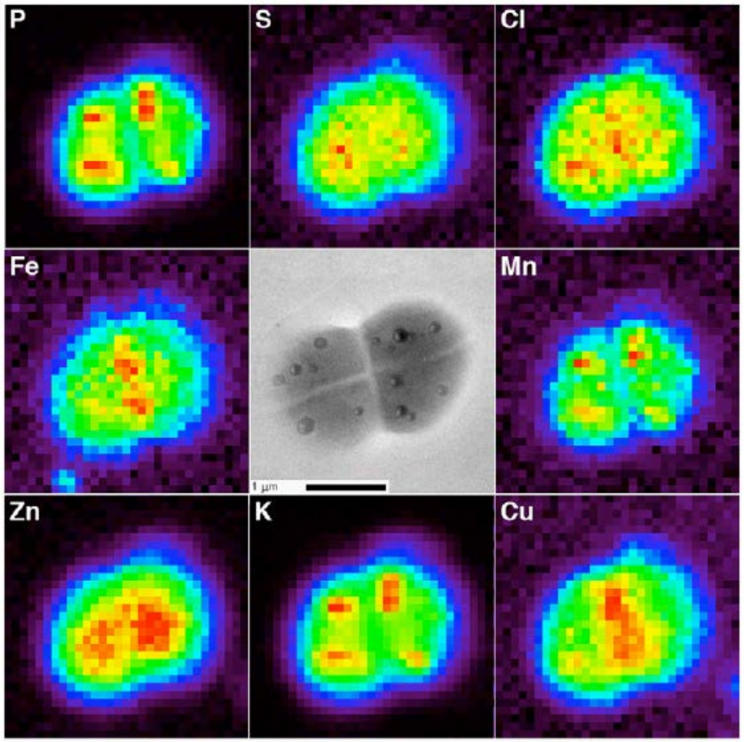
Buonassisi et al. Nature Materials (2005)





- Multicrystalline Si is an inexpensive alternative to costly monolithic Si for solar cells
- X-ray fluorescence microscopy is used to identify defect regions in mc-Si and to develop processes to improve device performance

SFXM → Elemental Map → Critical Biological Information



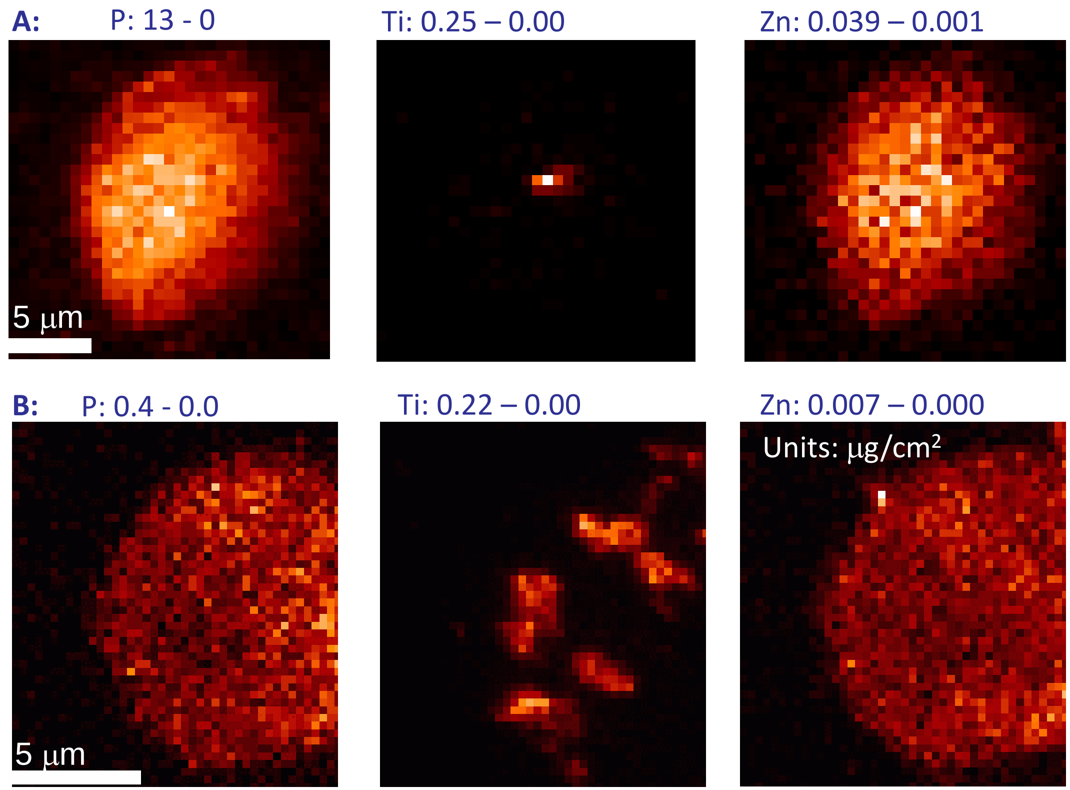
Protein Oxidation Implicated as the Primary Determinant of Bacterial Radioresistance

M. J. Daly, E. K. Gaidamakova, V. Y. Matrosova, A. Vasilenko, M. Zhai, R. D. Leapman, B. Lai, B. Ravel, S-M. W. Li, K. M. Kemner, J. K. Fredrickson

PLoS Biology | www.plosbiology.org 0001 April 2007 | Volume 5 | Issue 4 | e92

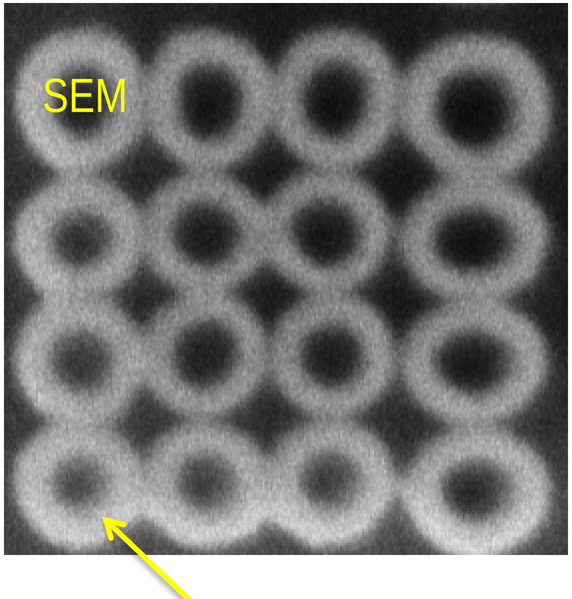
Woloschack et al. Nature Materials 2, 343 (2003)

- Map Ti distribution using X-ray fluorescence, to quantify success rate of TiO_2-DNA transfection, & visualize target
- A: nanocomposites targeted to nucleolus
- B: nanocomposites targeted to mitochondria



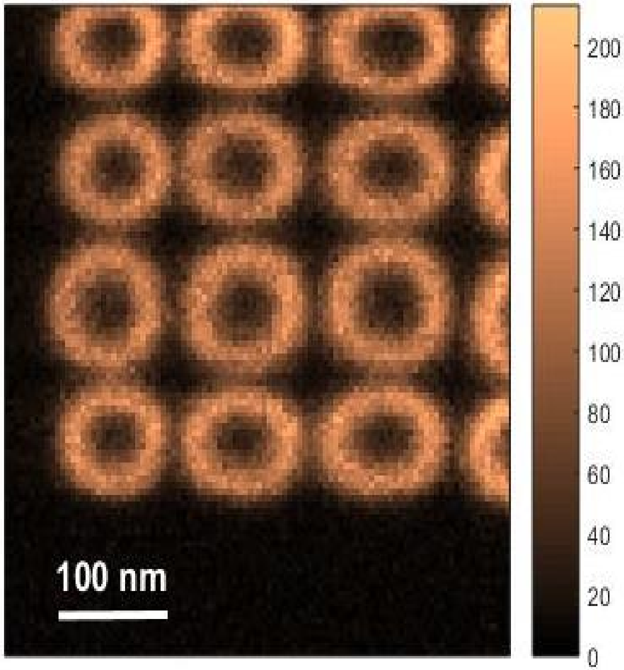
NSLS-II HXN: Fly-scan Nano-Imaging at 15 nm Resolution

Pt test pattern (200 nm thick)
(made by M. Liu, CFN)



20 nm wide

HXN: 12 keV @ 50 mA current



Pt XRF fly-scan: 5nm/pixel

Dwell time: 0.5s/pixel

Spatial Resolution: ~15 x 15 nm (FWHM),
~11 x 13 nm (PDS, based on cutoff frequency)

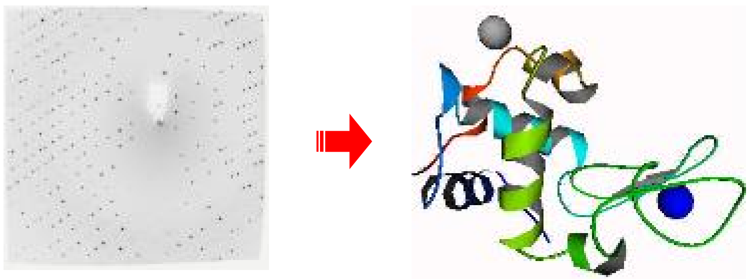
Today's Lecture

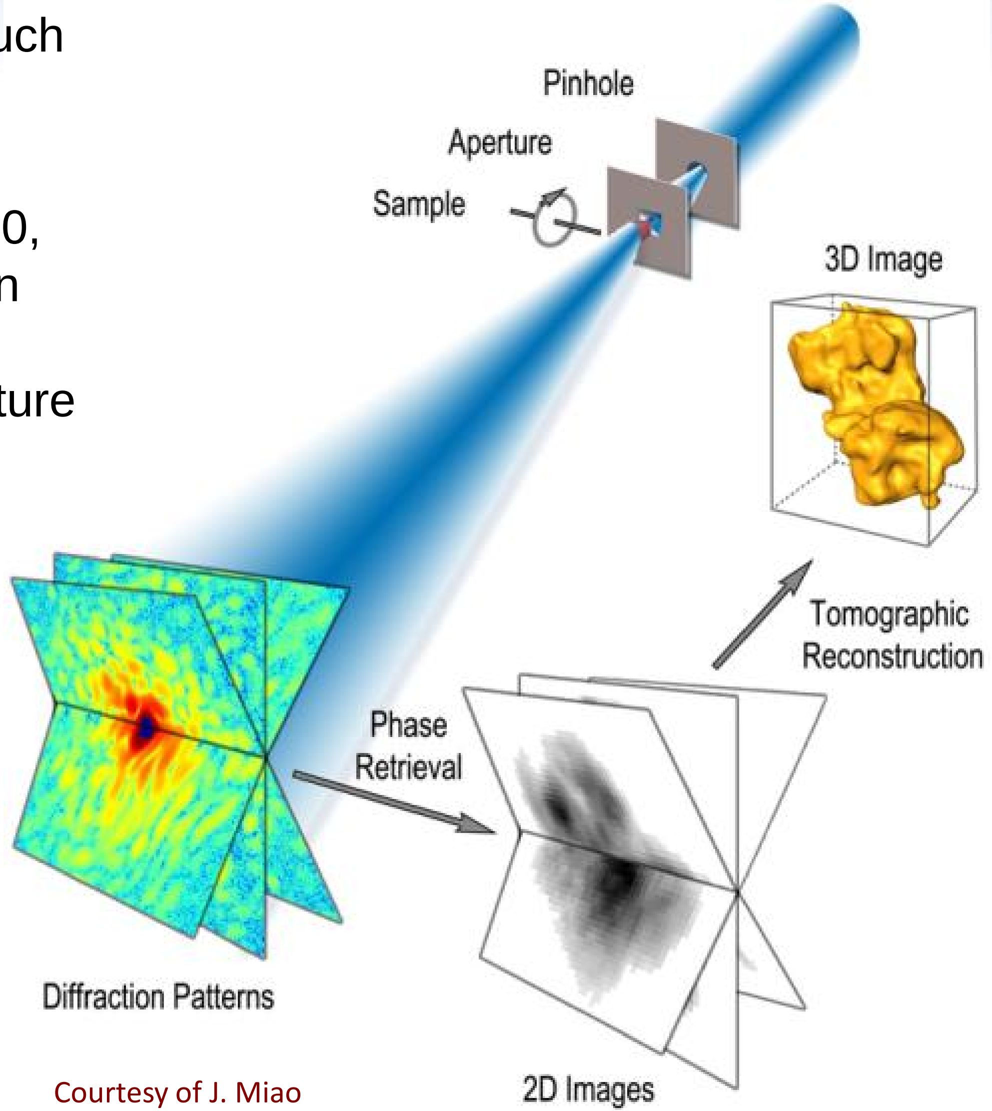
- Introduction to X-ray Imaging/Microscopy
  - absorption & phase contrast
  - Fresnel wave propagation
- Lens-based X-ray Microscopy
  - focusing optics: refractive lens, reflective & diffractive optics
  - Fresnel zone plate: numerical aperture, resolution, magnification
  - full-field transmission x-ray microscope
  - scanning x-ray microscopy
- Lens-less X-ray Microscopy
  - coherent diffraction imaging/microscopy
  - coherent diffraction ptychography
- Application Examples in Each Section

Coherent X-ray Diffraction Imaging (CDI)

- ➔ Coherent diffraction 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

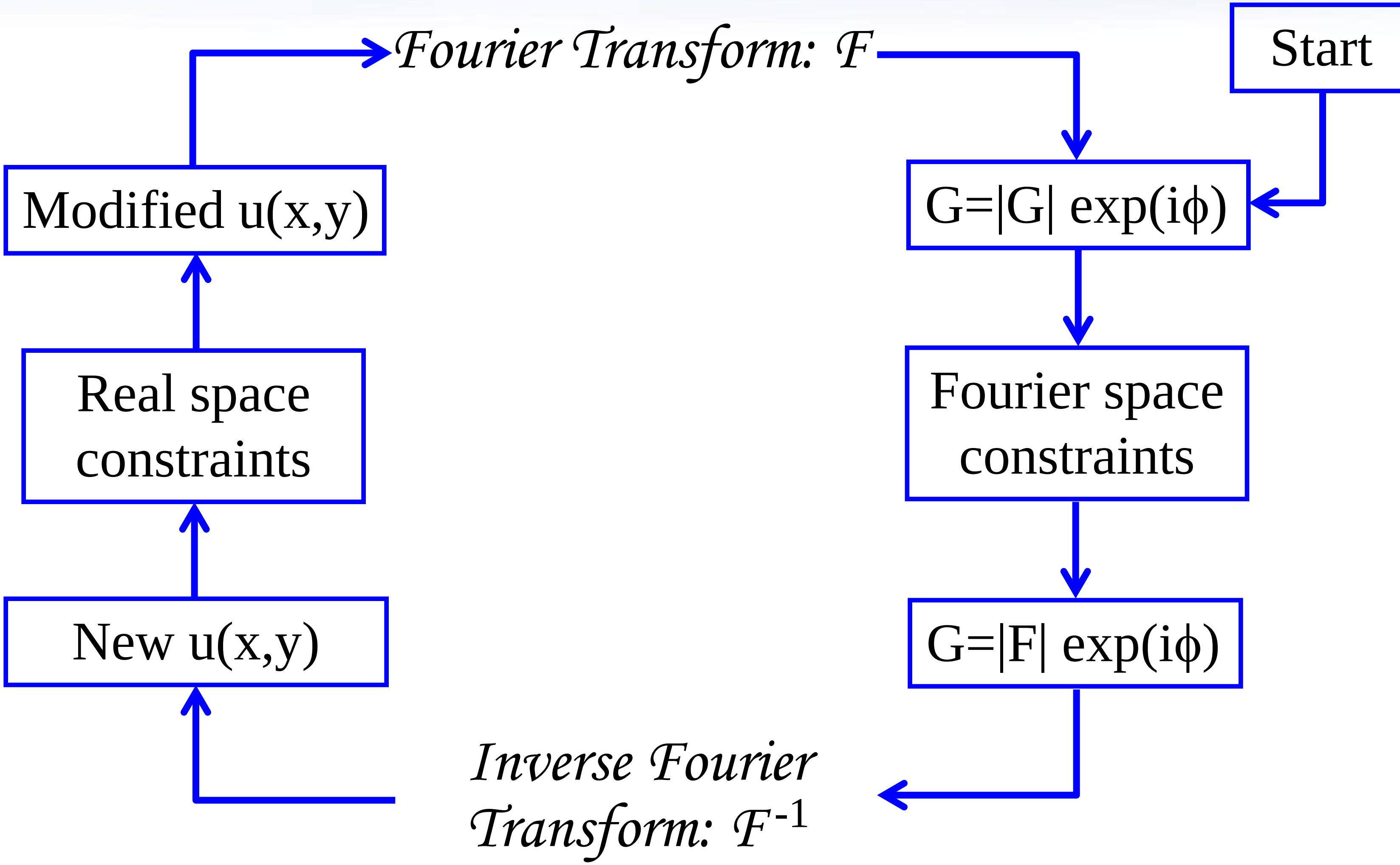
Analogous to crystallography



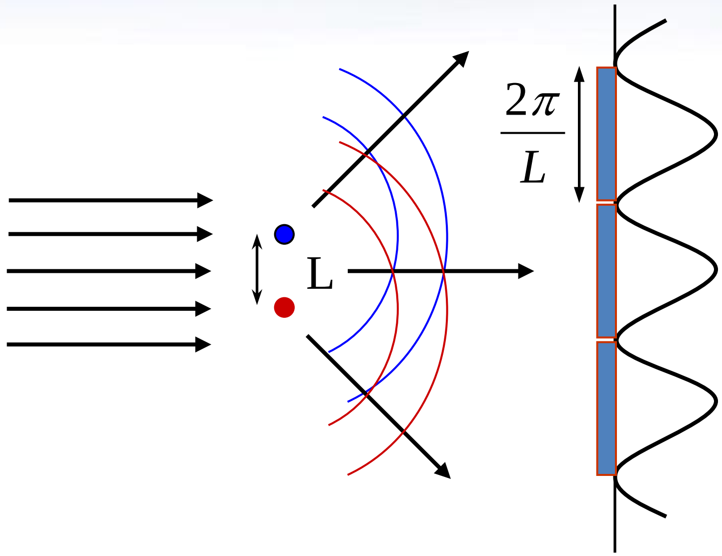


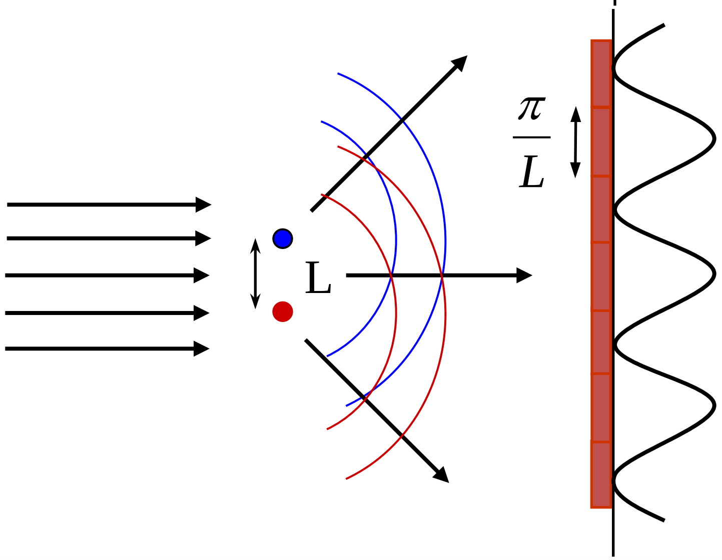
Courtesy of J. Miao

Iterative Phase Retrieval



Oversampling Requirement in CDI: Angular Resolution





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

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

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

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

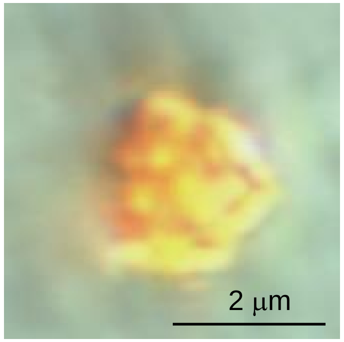
$$\Delta Q_{\max}^{2D} = \frac{2\pi}{L} \cdot \frac{1}{\sqrt{2}} = \frac{\sqrt{2}\pi}{L}$$

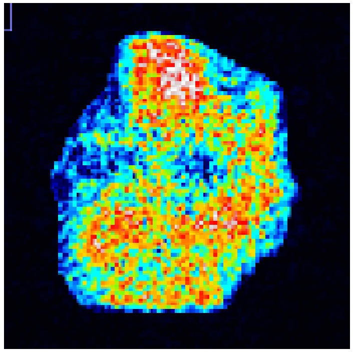
$$\Delta Q_{\max}^{3D} = \frac{2\pi}{L} \cdot \frac{1}{\sqrt[3]{2}}$$

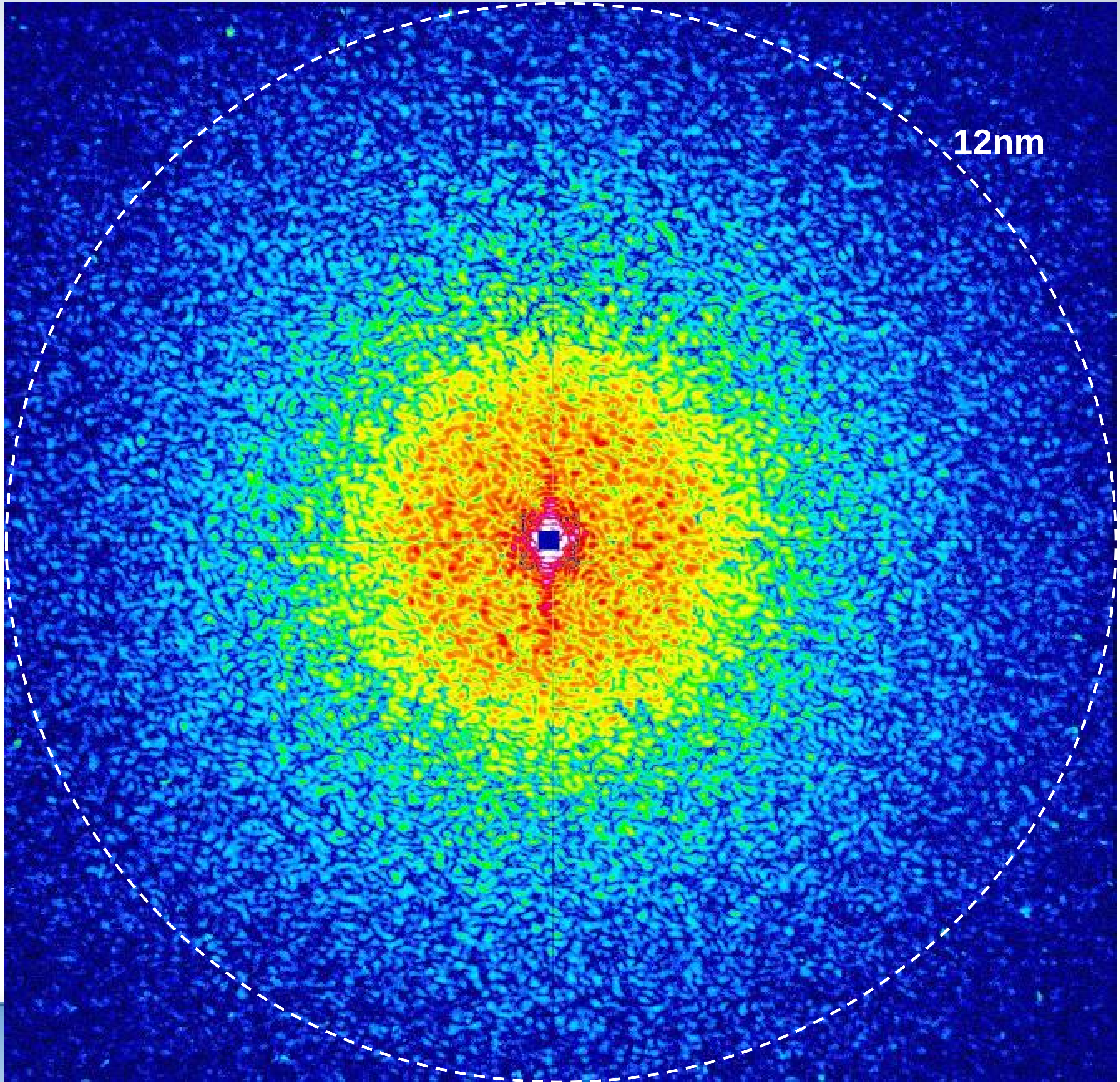
CDI Experiment on Nanoporous Gold

Xiao, Shen, Sandy (APS)
Chen, Dunand (NU)

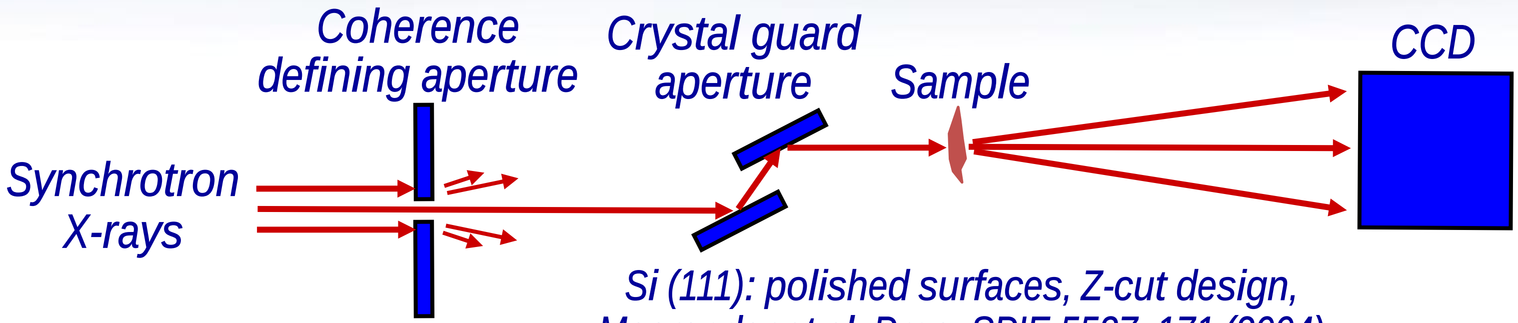
APS 8-ID



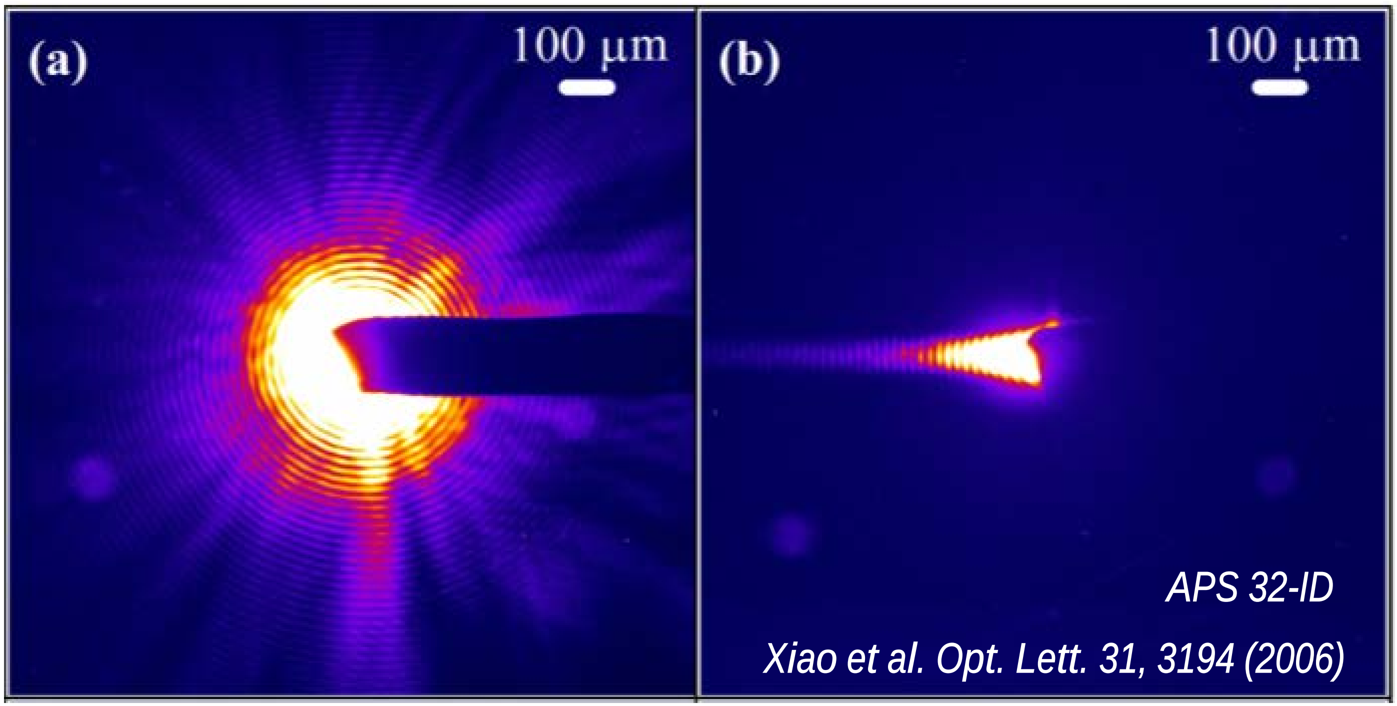




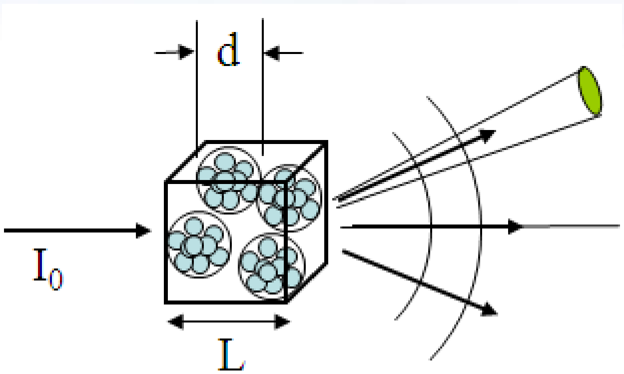
Improving S/N in CDI with Crystal Guard Aperture

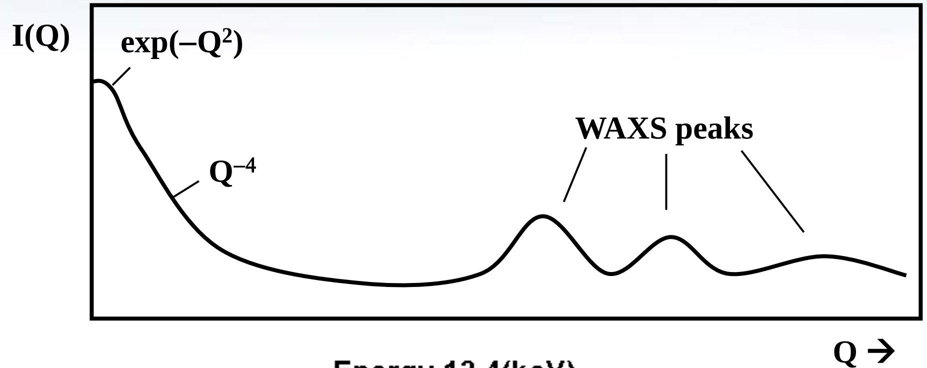


Si (111): polished surfaces, Z-cut design,
Macrander et al. Proc. SPIE 5537, 171 (2004)



Scaling Laws in Coherent Diffraction Imaging





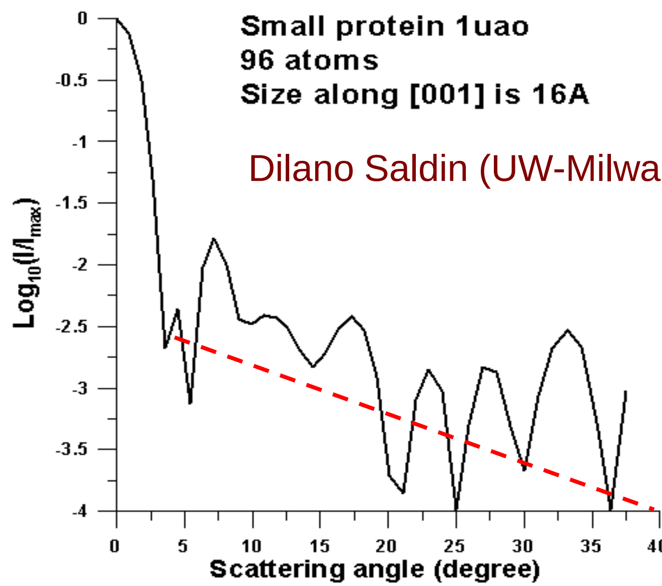
Energy 12.4(keV)

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

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

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

Shen et al. JSR 11, 432 (2004)



Dilano Saldin (UW-Milwaukee)

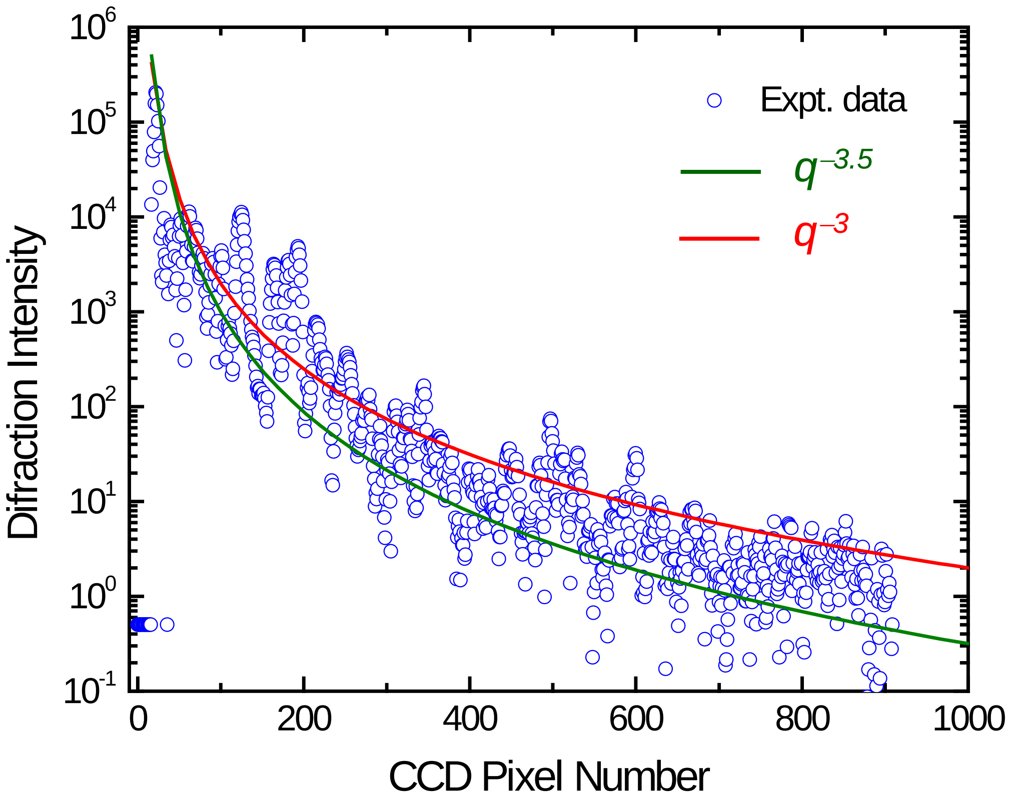
Marchesini, Howells, et al.
Optics Express (2003)

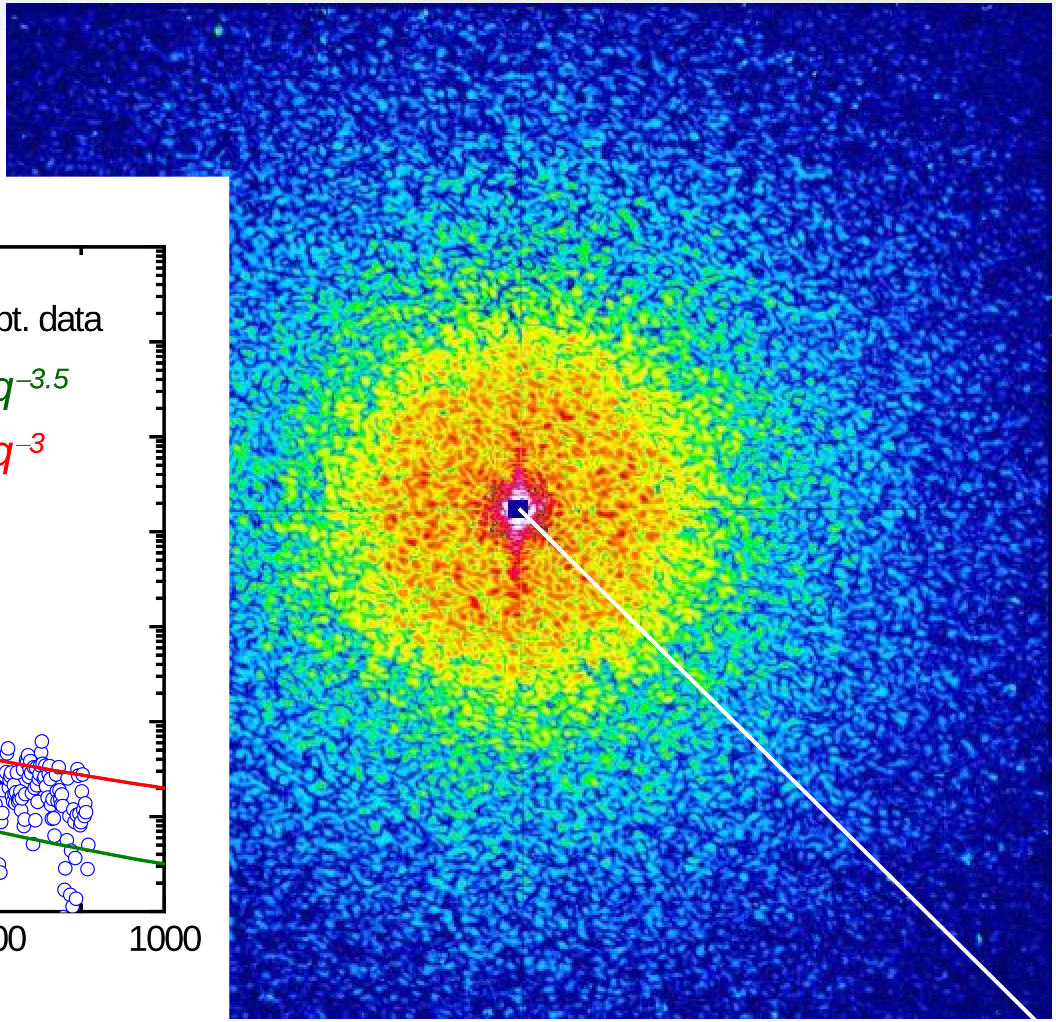
$$\boxed{I \sim d^4\lambda^2}$$

Q-dependence of Coherent Scattering Signal

Experiment results:

Gold nanofoam specimen
CDI data to ~ 8 nm

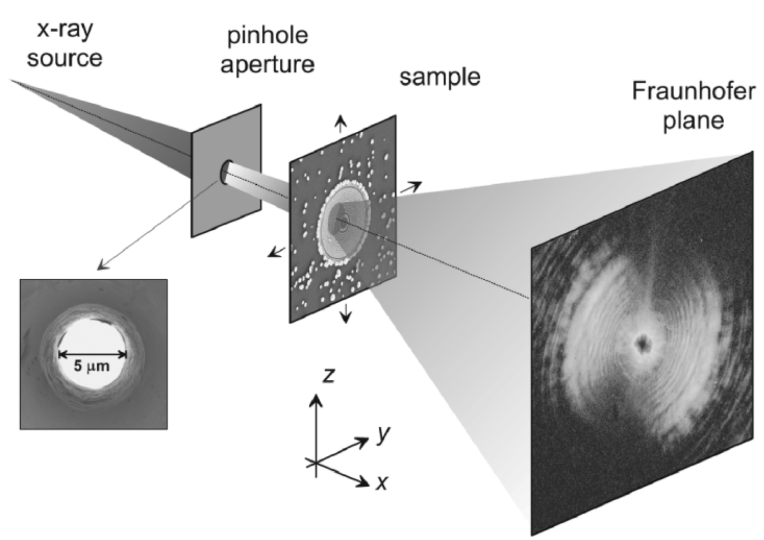




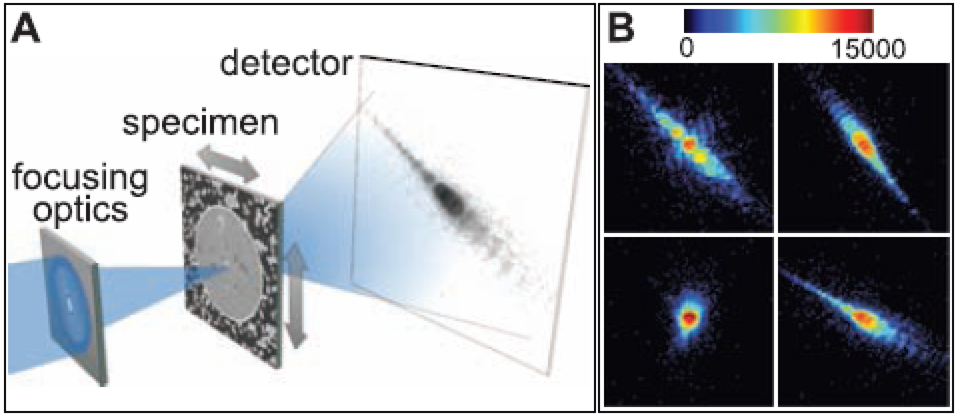
➔ Scaling law depends on specimen heterogeneity length scales !

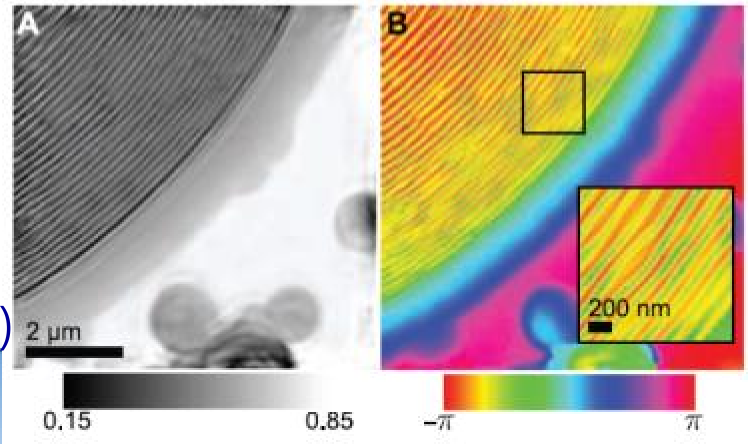
Extended Object: Ptychography = CDI w/ STXM

- Ptychography: moving-aperture lensless imaging [Rodenburg, et al, PRL (2007)]: overlapped sampling regions provide additional constraints in real space.
- Combining CDI with STXM: performing CDI measurements in a scanning x-ray microprobe.



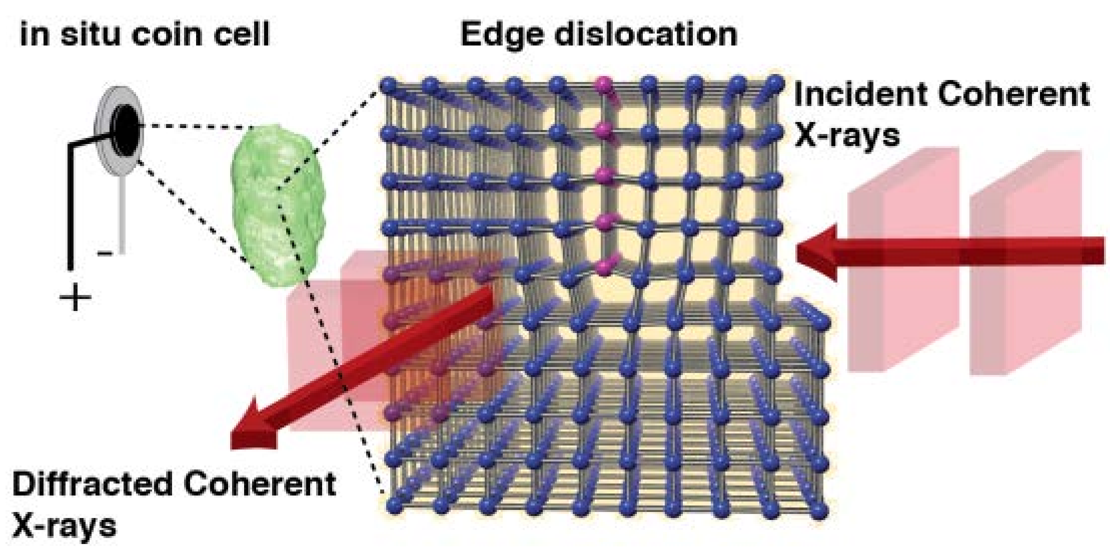
Rodenburg, et al, PRL (2007)

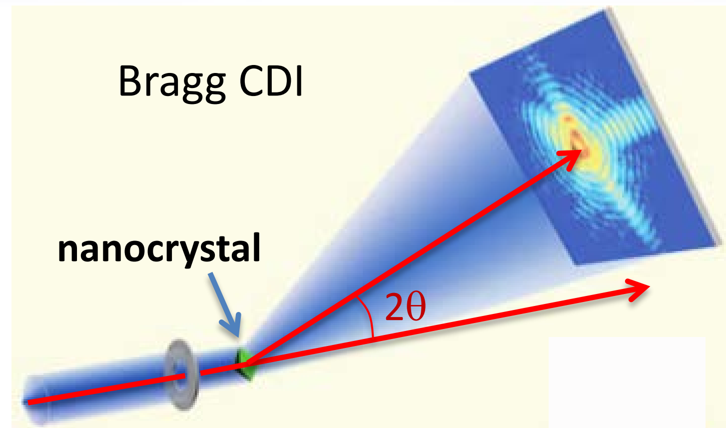




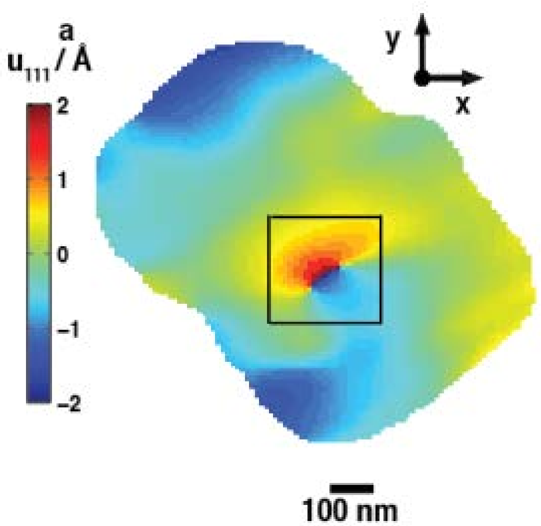
Thibault, et al, Science (2008)

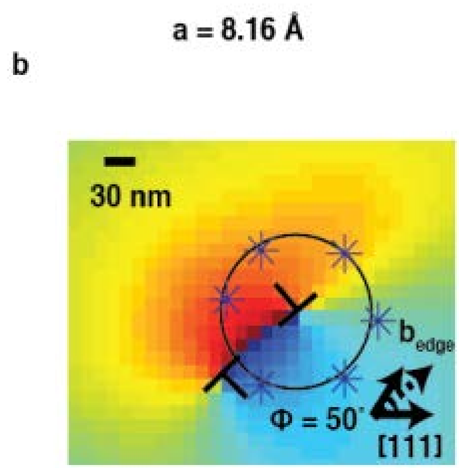
CDI in Bragg Geometry – Imaging Strain Fields in Nanocrystals



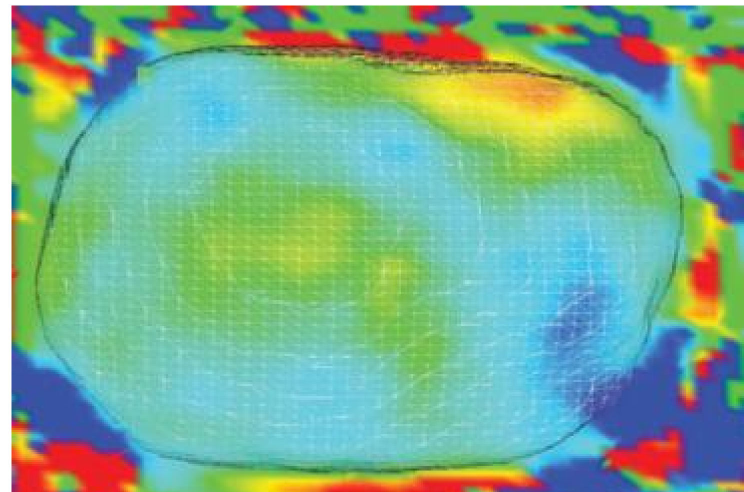


I. K. Robinson, et al, Phys. Rev. Lett. 87, 195505 (2001).





Ulvestad, et al., Science 348, 1344 (2015)



Robinson & Harder, Nature Mat. (2009) doi: 10.1038/nmat2400.

Sensitivity to Strain Field in Nanocrystal

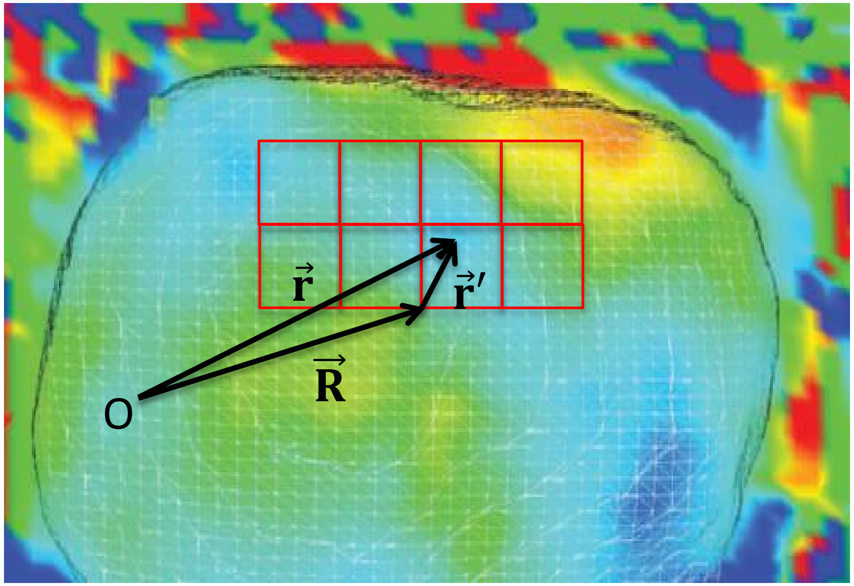
$$S(\mathbf{Q}) = \int \rho(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}}\, d\mathbf{r}$$

$Strain\ field\colon\quad \mathbf{r} \rightarrow \mathbf{r} + \mathbf{u}(\mathbf{r})$

$\mathbf{u}(\mathbf{r}) \sim atomic\ displacement$

$$S(\mathbf{Q}) = \int \rho(\mathbf{r}) e^{i\mathbf{Q}\cdot[\mathbf{r}+\mathbf{u}(\mathbf{r})]}\, d\mathbf{r}$$

$$= \int \rho(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{r}} e^{i\mathbf{Q}\cdot\mathbf{u}(\mathbf{r})}\, d\mathbf{r}$$



$Note\ that\ \mathbf{u}(\mathbf{r})\ is\ slow\ varying\ compared\ to\ atomic\ positions$:

$$S(\mathbf{Q}) \approx \int \{e^{i\mathbf{Q}\cdot\mathbf{u}(\mathbf{R})} \rho(\mathbf{r}')\}\, e^{i\mathbf{Q}\cdot\mathbf{r}'}\, d\mathbf{r}' = \int \rho_{\mathbf{R}}(\mathbf{r}')\, e^{i\mathbf{Q}\cdot\mathbf{r}'}\, d\mathbf{r}'$$

$where\ \rho_{\mathbf{R}}(\mathbf{r}) = \rho(\mathbf{r}) e^{i\mathbf{Q}\cdot\mathbf{u}(\mathbf{R})} = \rho(\mathbf{r}) e^{i\phi}$ ➔ Strain field as additional phase in complex density function

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Today's Homework

MSAE E8235x Homework #8 November 4, 2015

A transmission X-ray microscope at an X-ray wavelength of 1 Å makes use of a Fresnel zone plate objective lens with a diameter of 80 μm and a focal distance of 20 mm.

(a) What is the achievable resolution of this X-ray microscope?

(b) A lens-couple CCD is used for this microscope. The 2k x 2k CCD has a pixel size of 15 μm. If a 20x objective is used for visible light, what is the effective pixel size at its scintillator screen?

(c) Suppose an oversampling ratio of 3x is required, what should the magnification be set at? Determine the sample distance F_1 and the image distance F_2 for this magnification.