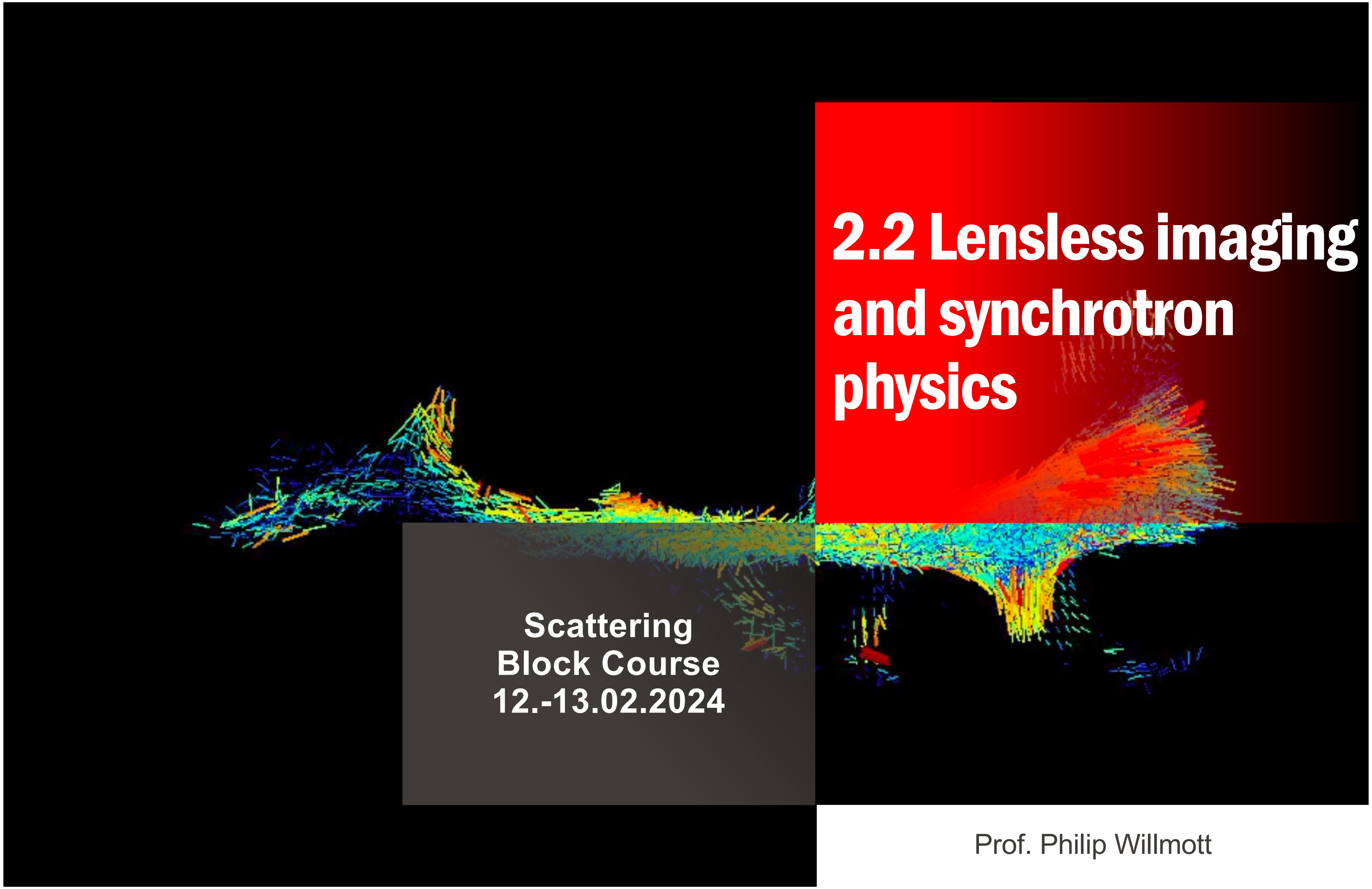
2.2 Lensless imaging and synchrotron physics



Scattering
Block Course
12.-13.02.2024

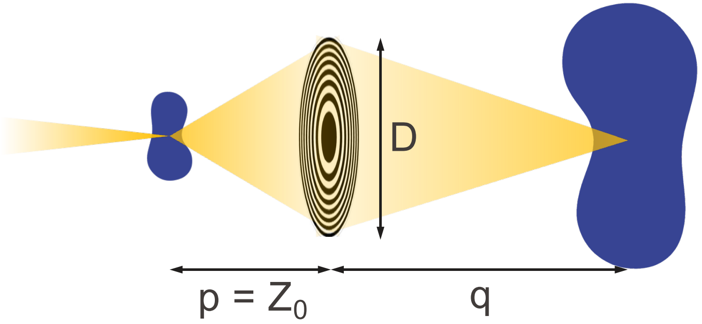
Prof. Philip Willmott

PSI

University of Zurich[UZH]

Lensless imaging – general considerations

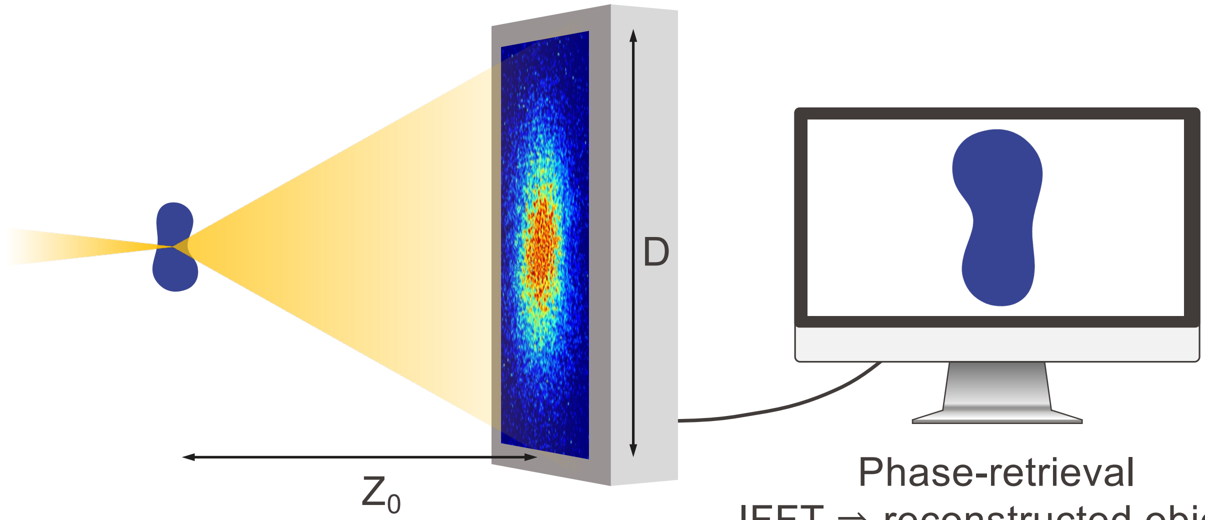
Lens vs lensless



M = q/p

$$\Delta x = \frac{1.22\,\lambda}{2\sin\theta} \approx \frac{1.22\,\lambda\,Z_0}{D}$$

$$Z_0/D \gtrsim 100$$



Phase-retrieval
IFFT ⇒ reconstructed object

Far-field
(Fraunhofer)
diffraction pattern
≡ |FT(object)|^2

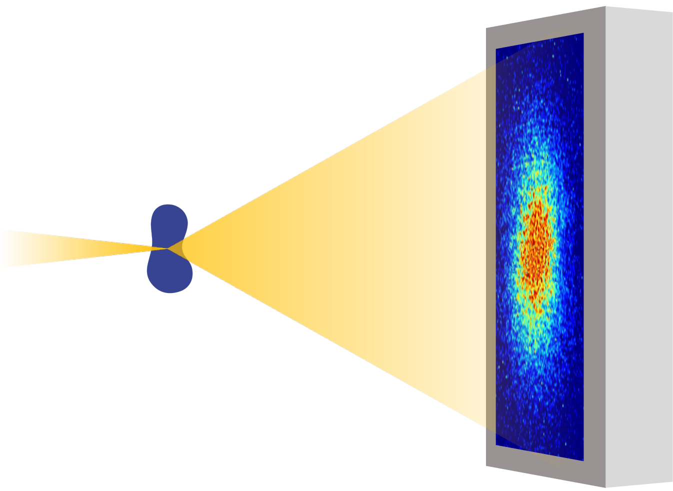
$$\Delta x = \frac{2\,\lambda}{2\sin\theta} \approx \frac{2\,\lambda\,Z_0}{D}$$

👍 No optical aberrations

$$Z_0/D \sim 10$$

👎 $I \propto Q^{-4}$

Lensless imaging and SAXS

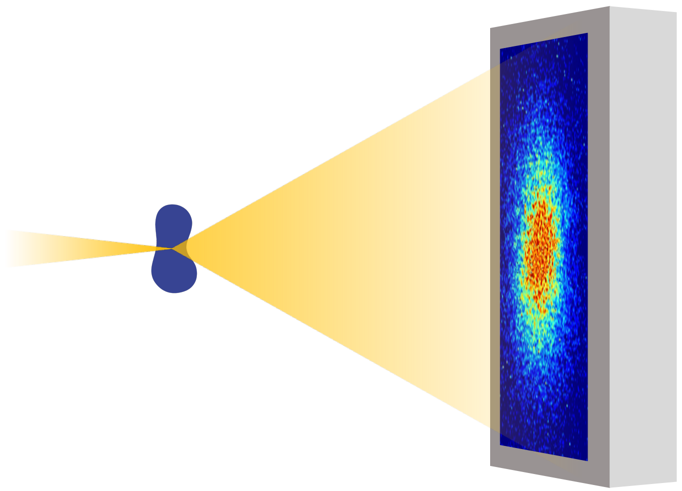


- Also “coherent x-ray diffractive imaging” CXDI
- Coherent illumination of sample
- Transverse (spatial) coherence

$$l_c^{(t)} = \frac{\lambda}{2\Delta\theta} = \frac{\lambda R}{2D}$$

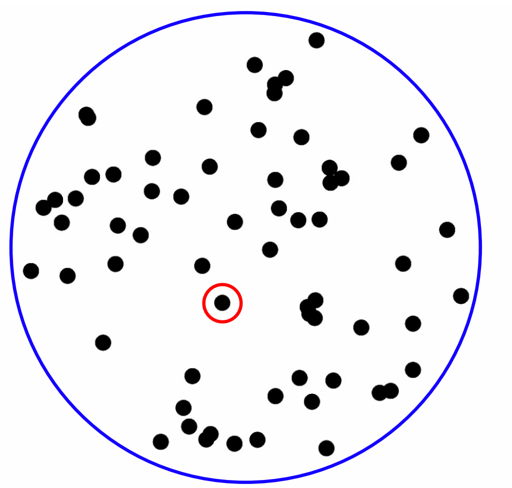
- Lensless-imaging beamlines
  - Long source–sample distance (R)
  - Small source size (D)
  - Highly collimated beam (θ)
  - Transverse coherence length ~ 200 – 1000 μm
  - Minimize optical elements that disrupt wavefront
- DLSRs: increase in coherent flux ~ 10^3 – 10^4!!

Lensless imaging and SAXS

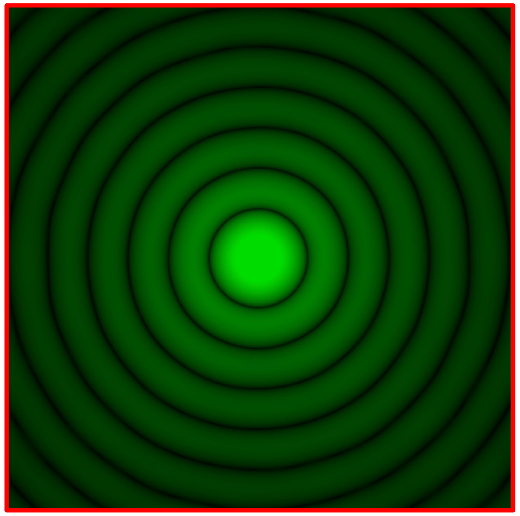


- Diffraction pattern
  - Noncrystalline samples
    - In forward-scattering direction only
  - Crystalline objects
    - Convolution of diffraction pattern due to periodicity and ‘shape function’ defining boundary of object
    - ⇒ regular array of replicas of same pattern
    - Bragg-CXDI

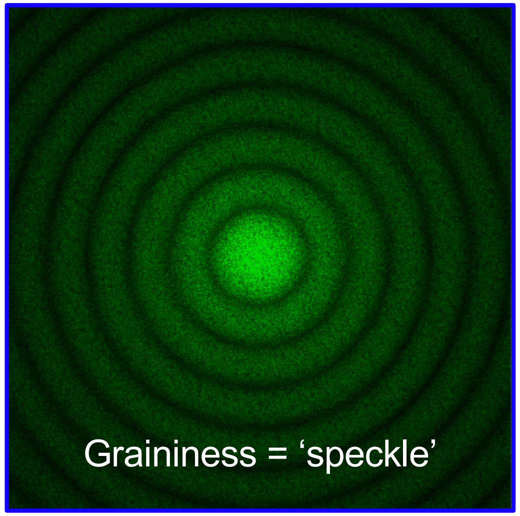
Lensless imaging vs SAXS



Ensemble of
n identical spheres



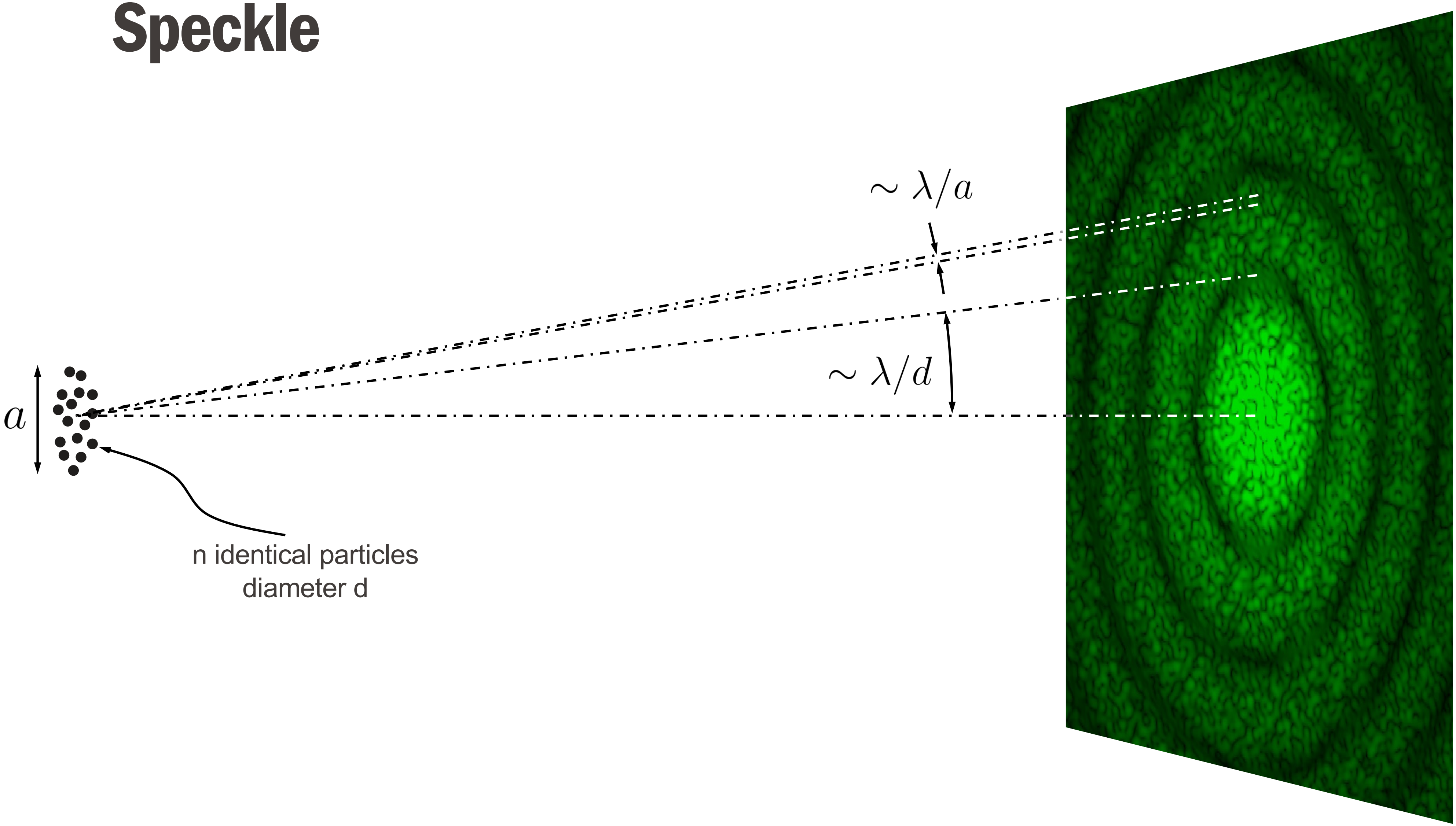
$$I = \sum_{n} |\mathcal{F}(\text{sphere})|^2$$
$$= n\,|\mathcal{F}(\text{sphere})|^2$$



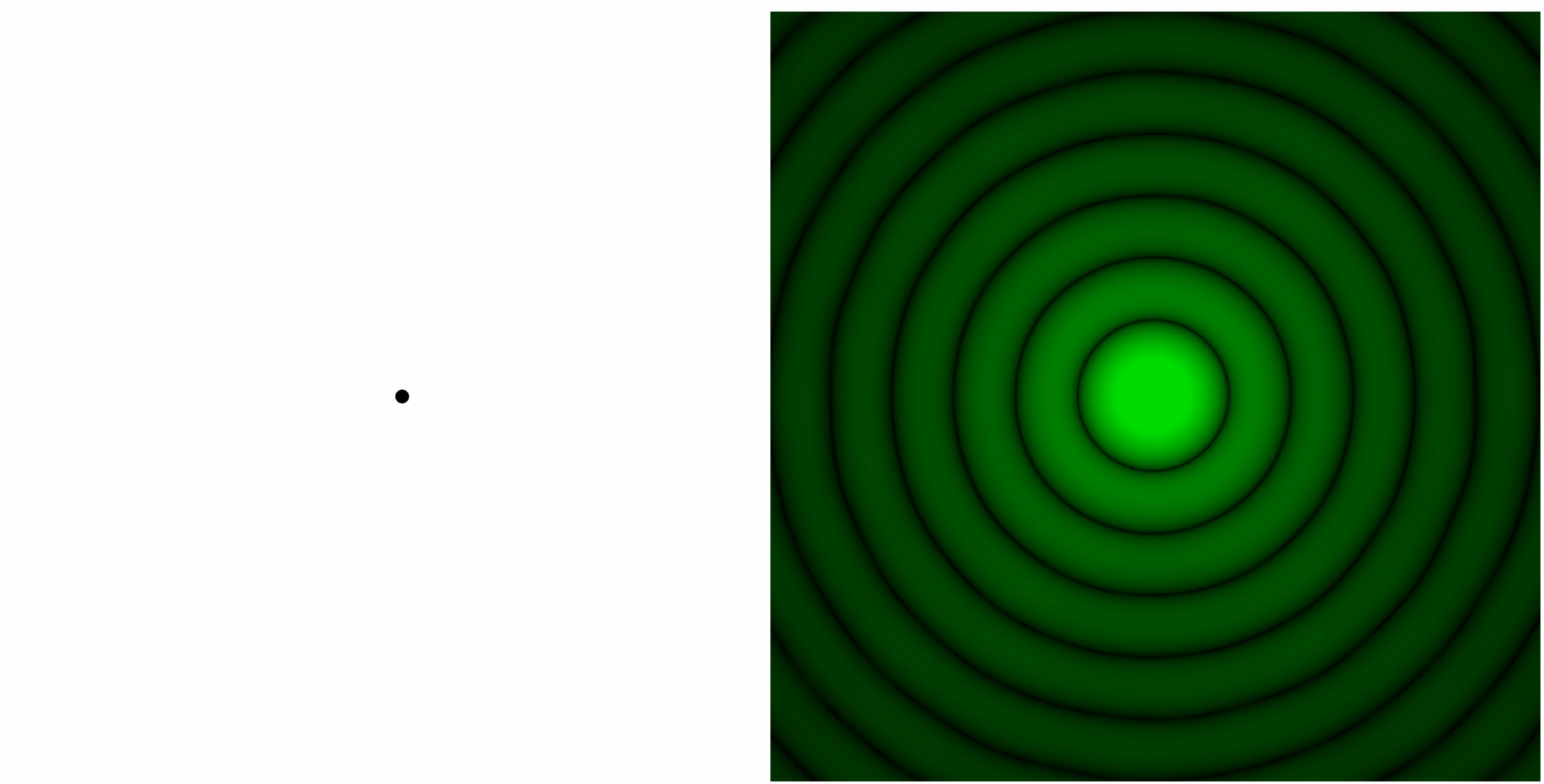
$$I = |\mathcal{F}(\text{ensemble of } n \text{ spheres})|^2$$

Interference between scattering
from individual scatterers

Speckle

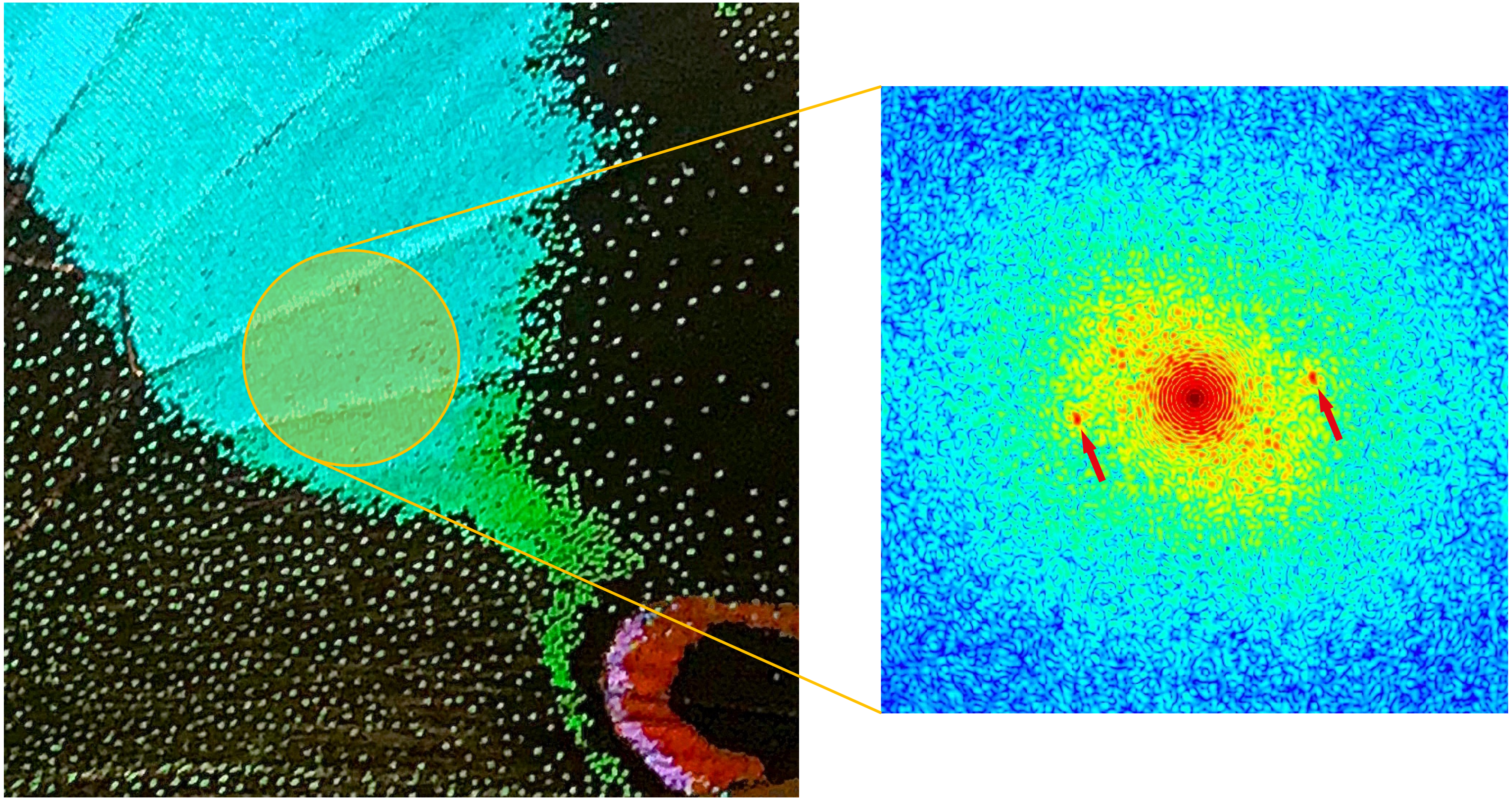


Speckle

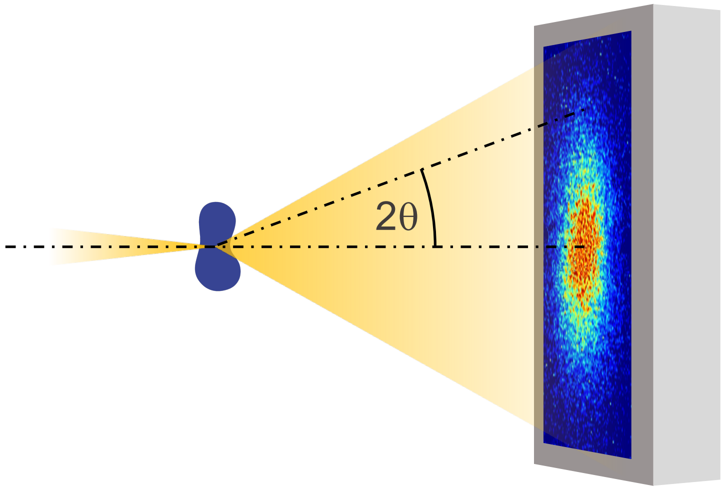


Progression of speckle pattern for increasing ensemble of identical spheres with constant average areal density

Speckle

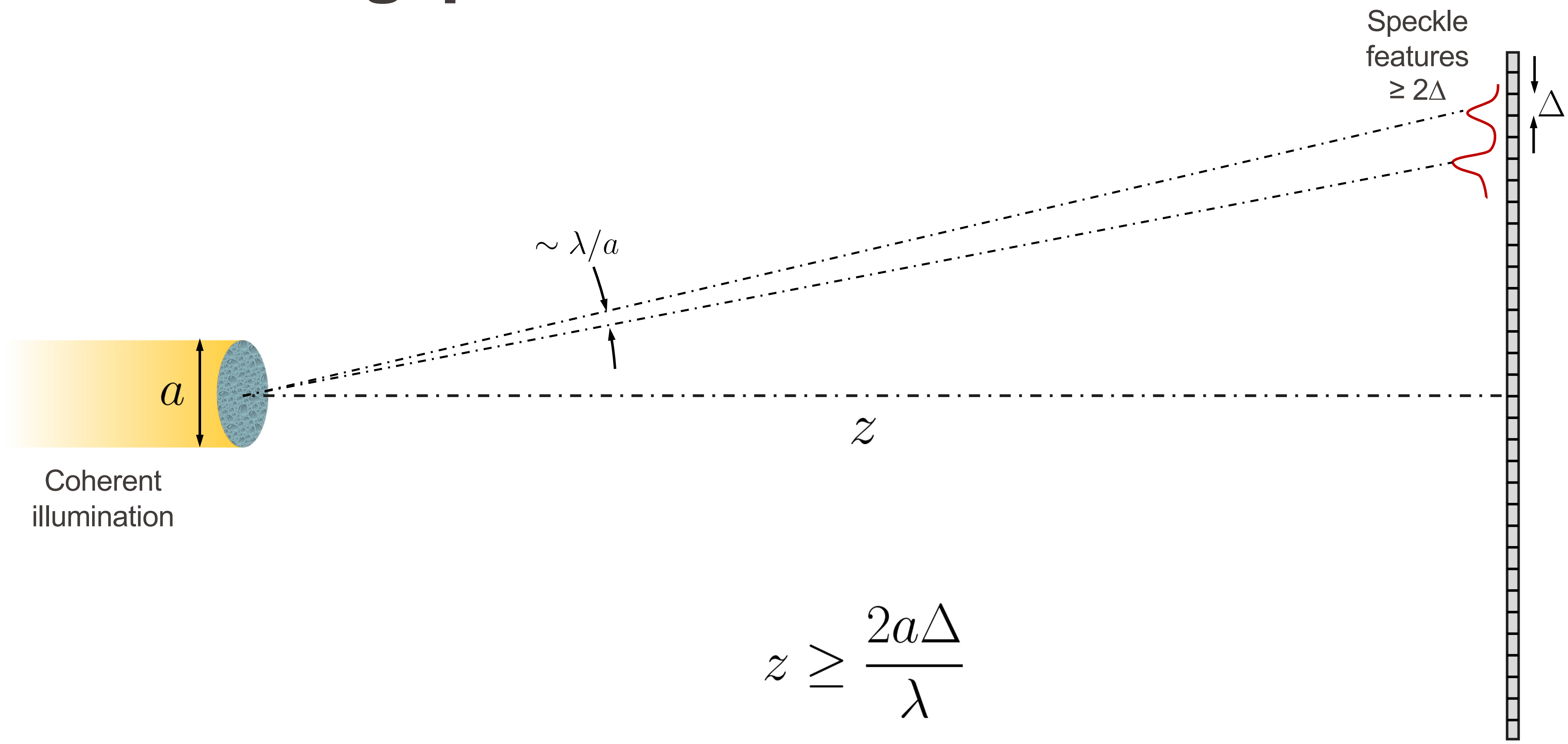


Lensless imaging and DLSRs



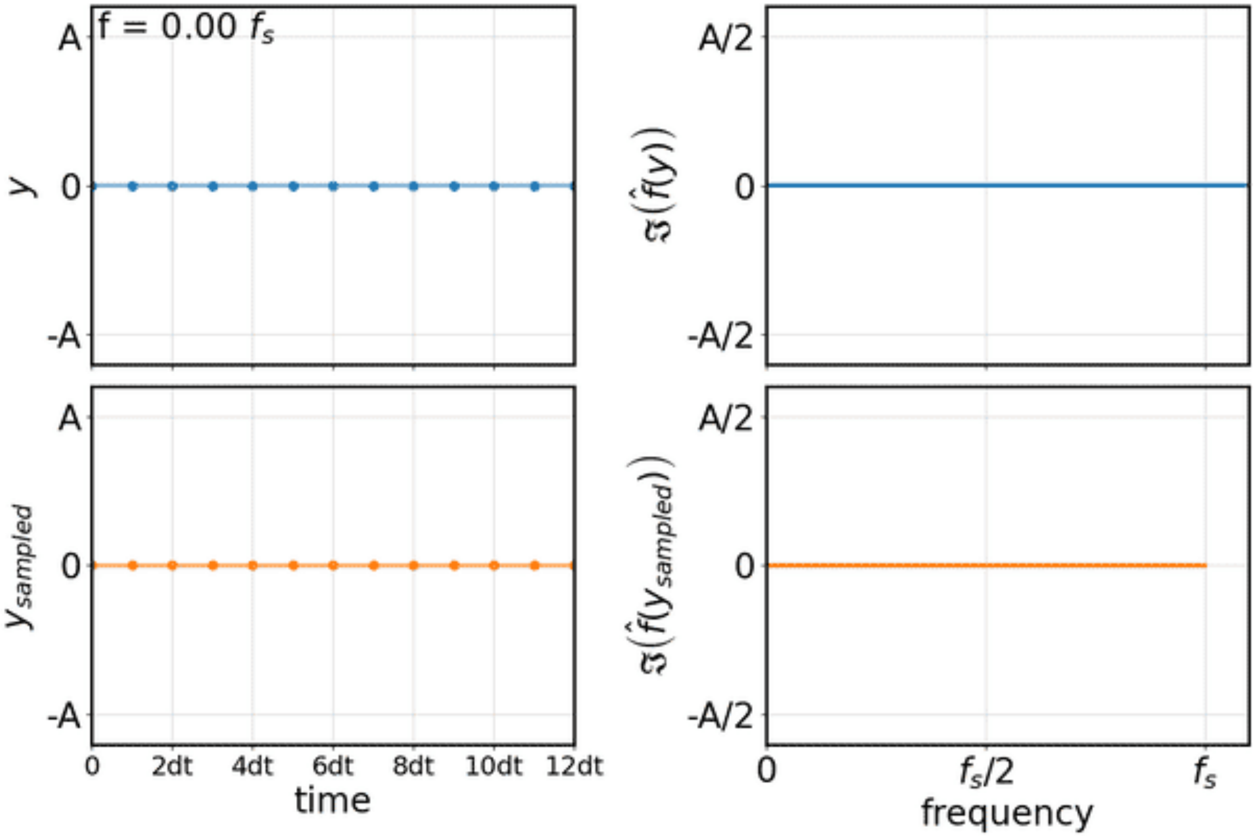
- Signal strength $\propto 1/Q^4 \simeq 1/\theta^4$
- 10^4 increase in coherent flux ⇒ x 10 increase in resolution ~ nm or smaller
- Sample manipulation accuracy becomes impossible
  - Becomes less competitive with e.g., electron imaging
- Exploit higher flux otherwise
  - Faster scanning
  - Higher photon energies
    - Less integrated dose
    - Larger penetration depths

Resolving speckle



$$z \geq \frac{2a\Delta}{\lambda}$$

Nyquist frequency



- Sampling frequency ≥ 2 x highest frequency contained in the signal

$$f_s \geq 2f_c$$

- For a given f_s, the maximum frequency you can accurately represent without aliasing is the Nyquist frequency. The Nyquist frequency equals one-half the sampling frequency

$$f_N = fs/2$$

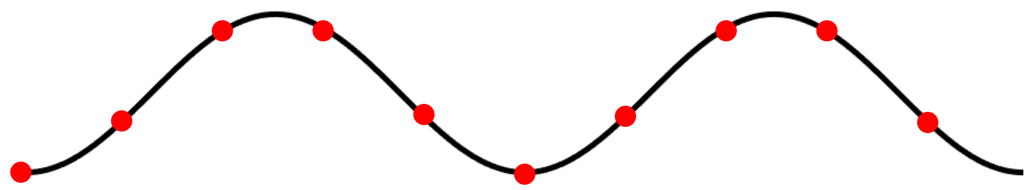
- ”Aliasing”: when

$$f_s < 2f_c$$

Oversampling

- Measuring spatial frequency

$$f_s > f_N$$

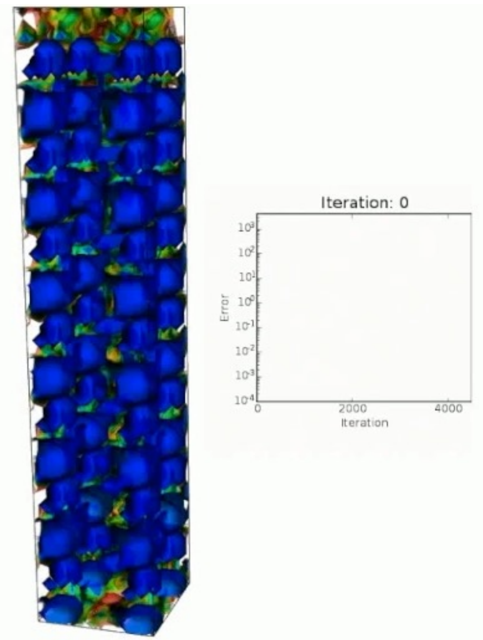


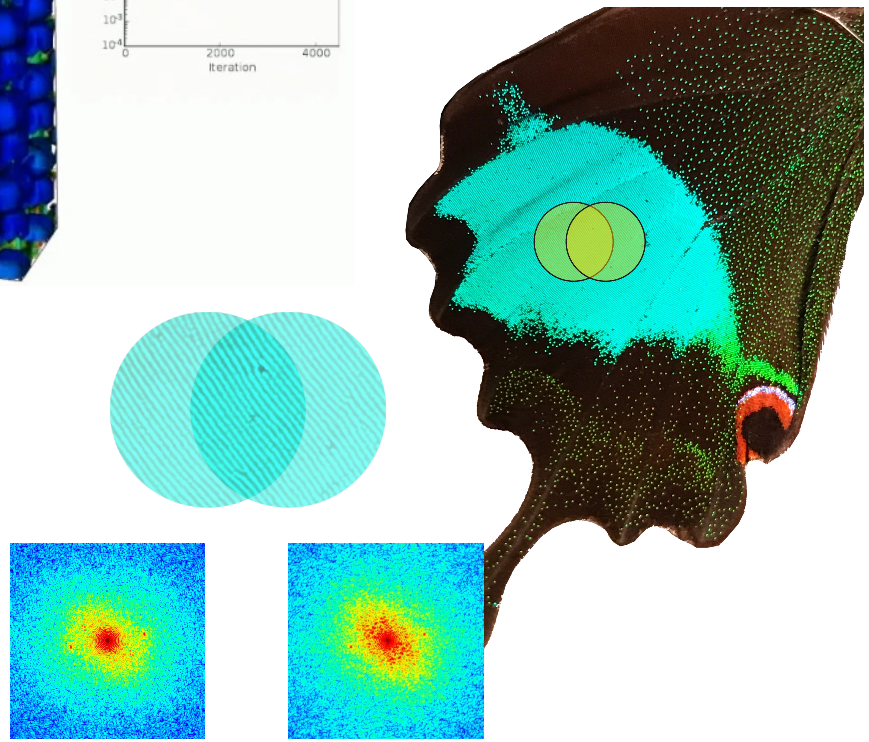
$$f_s = 2.5 f_N$$

$$O = \frac{f_s}{f_N}$$

Redundancy

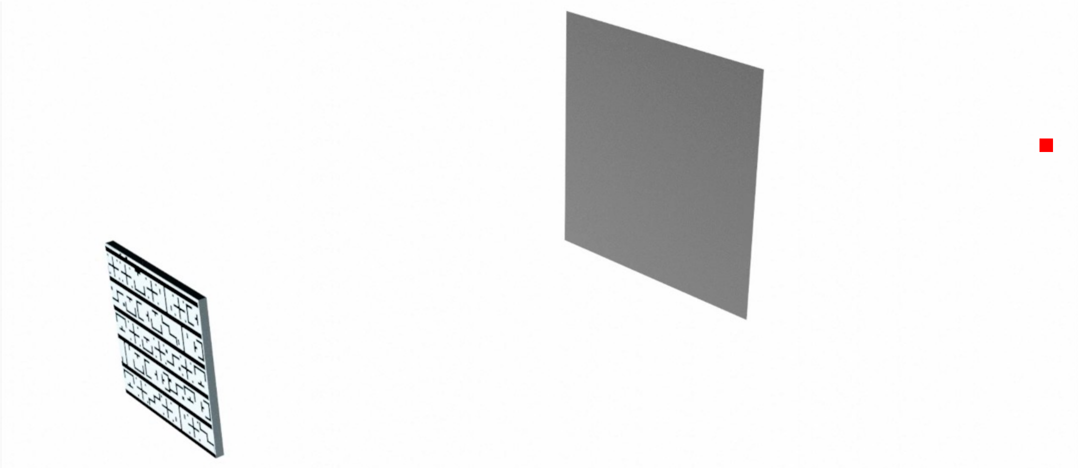
- Additional information/constraints beyond raw scattering data
  - Sparsity of real-space object such as atomicity
  - Physical extent of object
    - “Shrink-wrap”
  - Positivity of scattering (electron) density
  - Symmetry considerations
  - Consistency in overlapping illuminated regions (e.g., in ptychography)
  - …
- Narrows down possible solutions





Coherent x-ray diffractive imaging

Coherent x-ray diffractive imaging

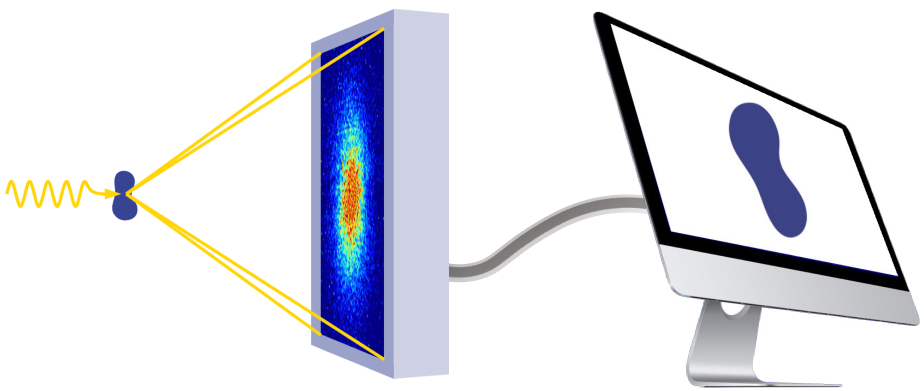


Animations courtesy I. Mochi, Swiss Light Source

- CXDI (or CDI)
- Also ‘lensless imaging’
- Diffraction pattern
  - Noncrystalline sample
    - In forward-scattering direction only
  - Crystalline objects
    - Regular array of replicas of same pattern
    - Bragg-CXDI
  - Oversampling determined by size of coherently illuminated sample
    - Smaller samples ⇒ larger features
- Phase problem resolved typically via phase-retrieval algorithms
  - Gerchberg-Saxton (error reduction)
  - Hybrid input-output (Fienup)
  - Difference-map
  - …

See V. Elser https://opg.optica.org/josaa/abstract.cfm?URI=josaa-20-1-40

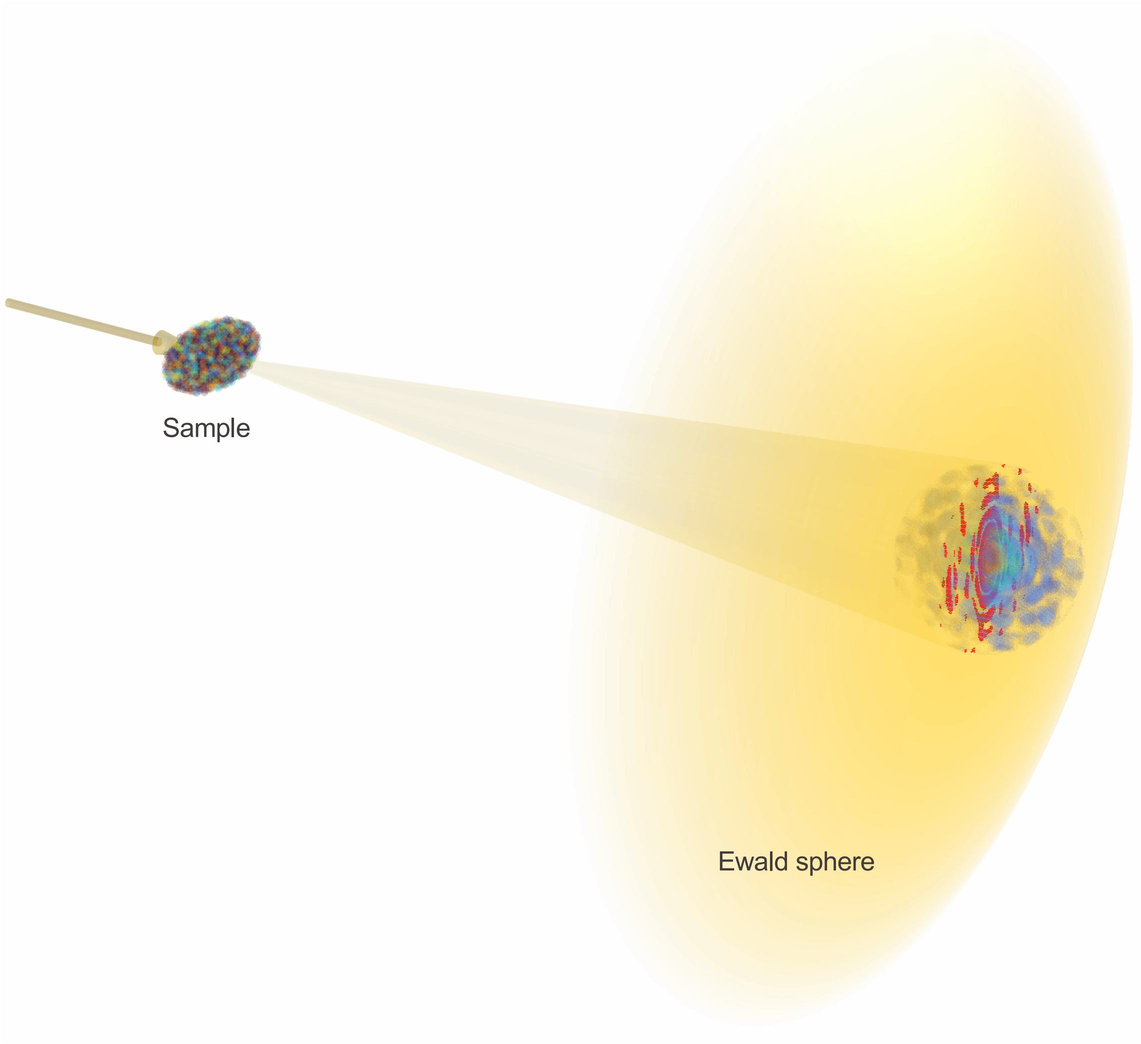
Coherent x-ray diffractive imaging



- XRD
  - Samples have translational symmetry
  - Record far-field scattering (diffraction) pattern
  - Regain real-space structure through IFT
    - Phase problem
  - Unit cells ≲ 200 Å
  - Resolution ≲ Å
- CXDI
  - Same principle as XRD
  - Samples can be crystalline or noncrystalline
    - Scattering pattern: "speckle"
    - Sizes up to ~ μm
    - Requires sample < coherence volume of SR
    - Big improvements with DLSRs!!
    - Resolution down to ~ 10 nm

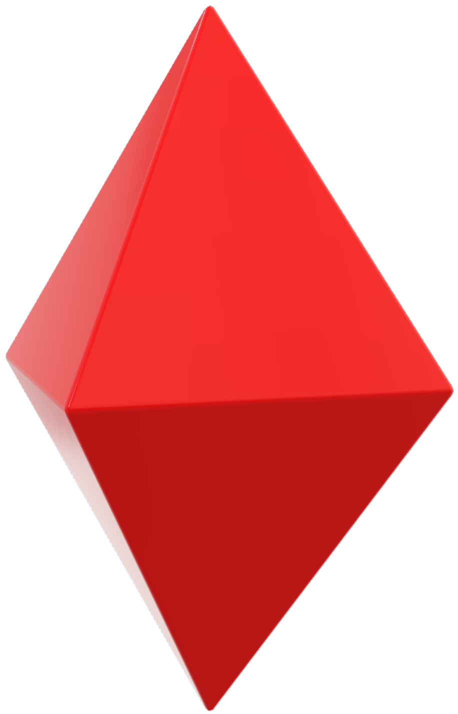
See also H.N. Chapman and K.A. Nugent
https://www.nature.com/articles/nphoton.2010.240

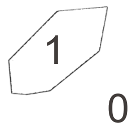
CXDI



- CXDI in forward-scattering direction
  - Used for noncrystalline samples
  - Sample bathed in coherent x-rays
    - Limits sample size
  - Requires rotation of sample at least by 180°, or even 360° if close to an absorption edge (scattering pattern loses its centrosymmetry – this doubling of information contributes to redundancy)

Bragg CXDI – a perfectly regular starry firmament



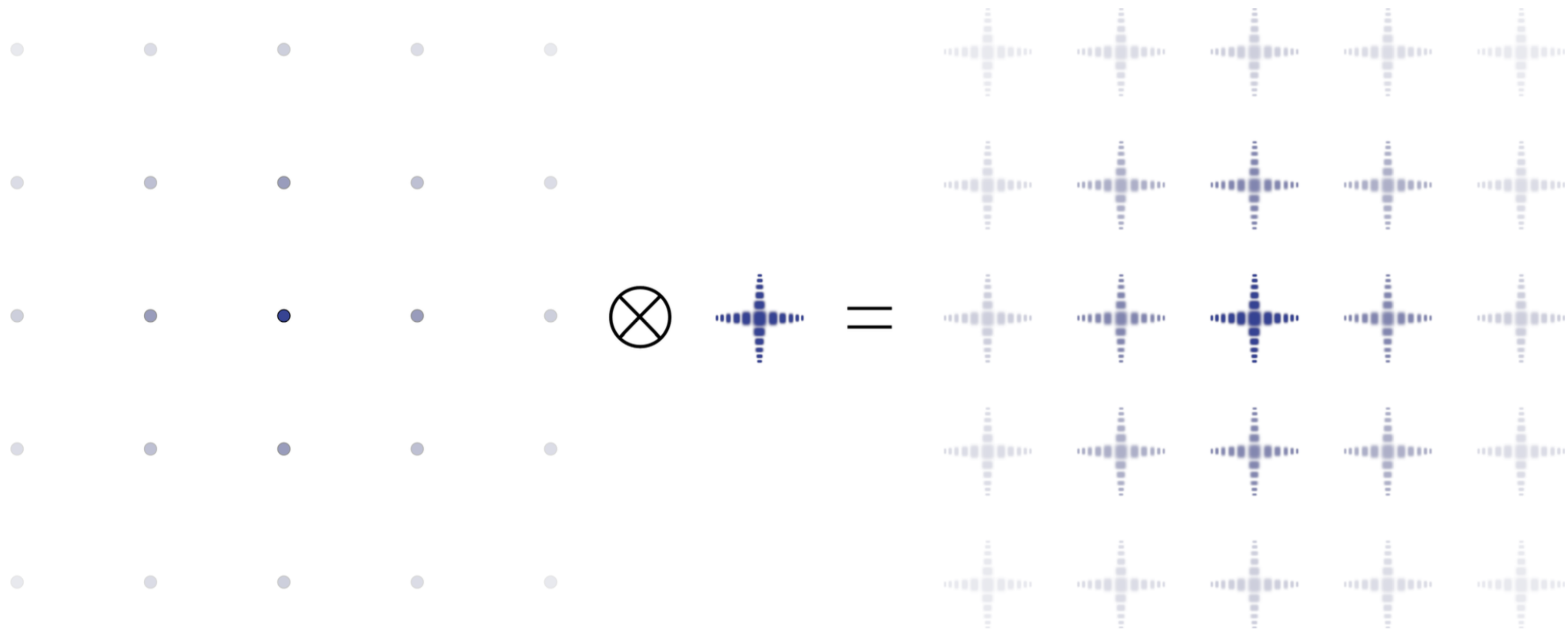


Perfect large crystal x Nanosized volume = Nanocrystal

Bragg CXDI – a perfectly regular starry firmament

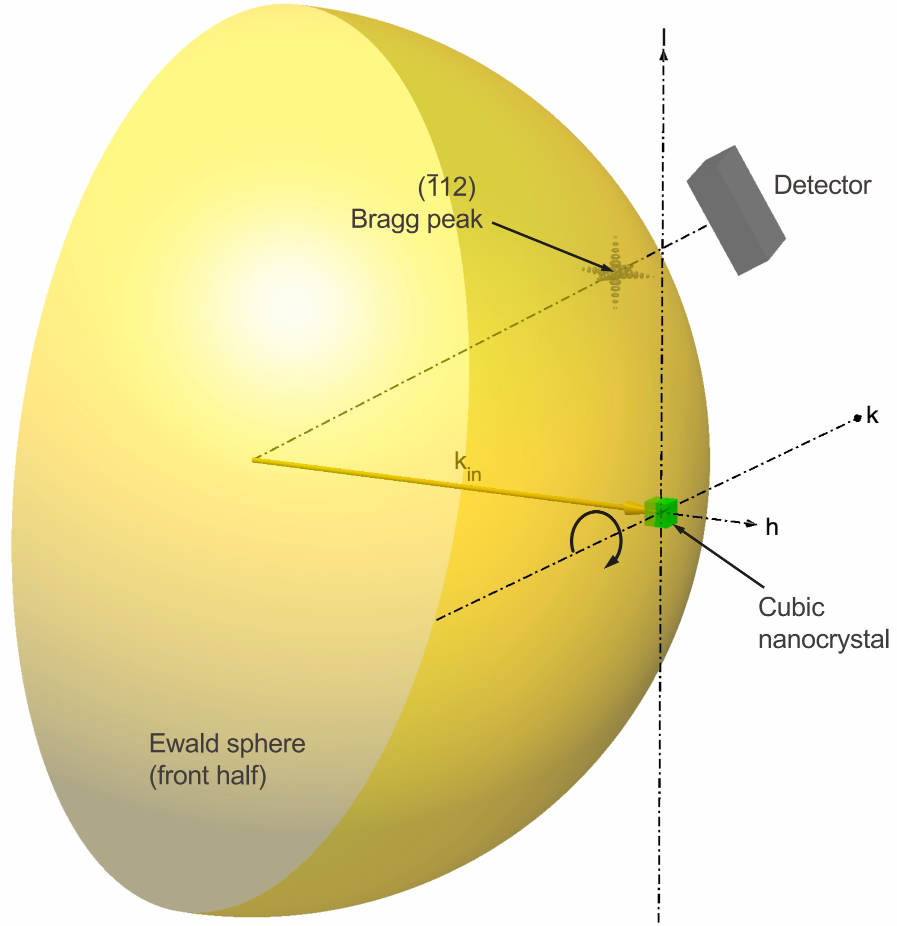
$$\mathcal{F}(A \times B) = \mathcal{F}(A) \otimes \mathcal{F}(B)$$

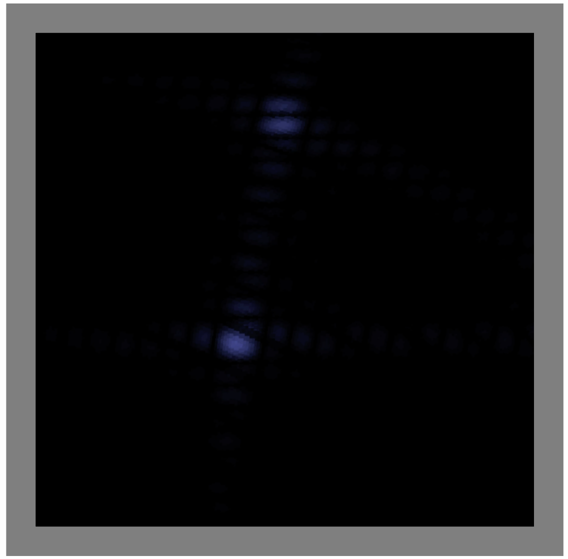
Bragg CXDI – a perfectly regular starry firmament



Diffraction pattern of large crystal ⊗ Shape function Diffraction pattern of nanocrystal

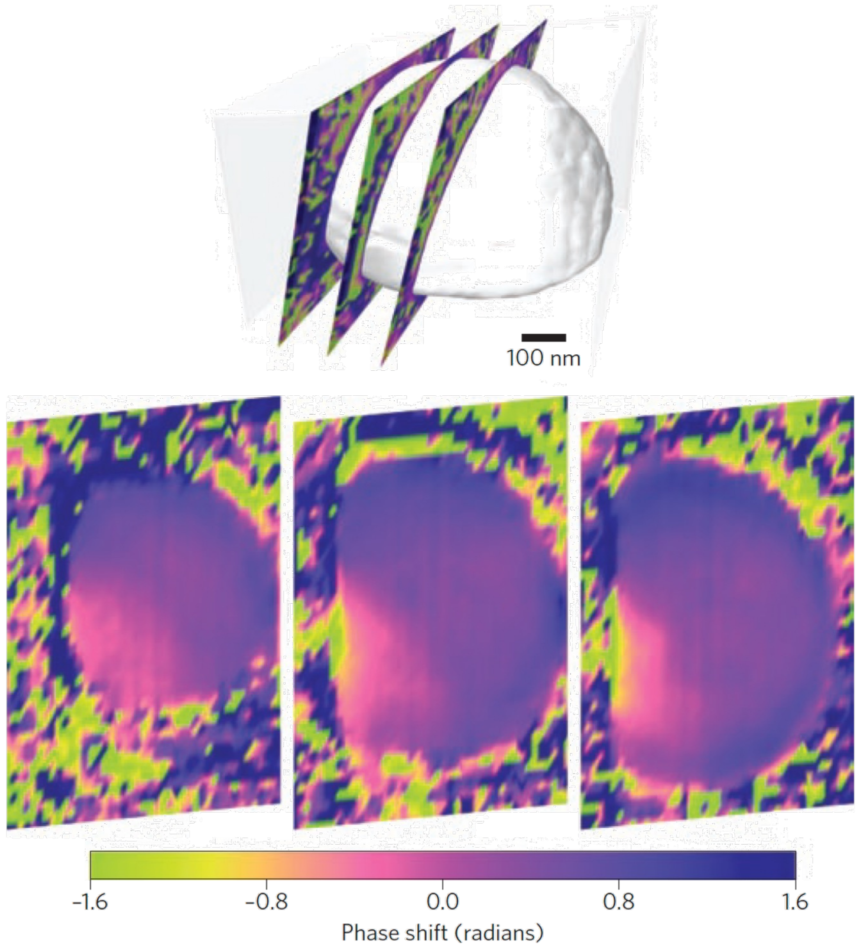
Bragg CXDI





Rotation angles ~ few degrees

Applications of Bragg CXDI

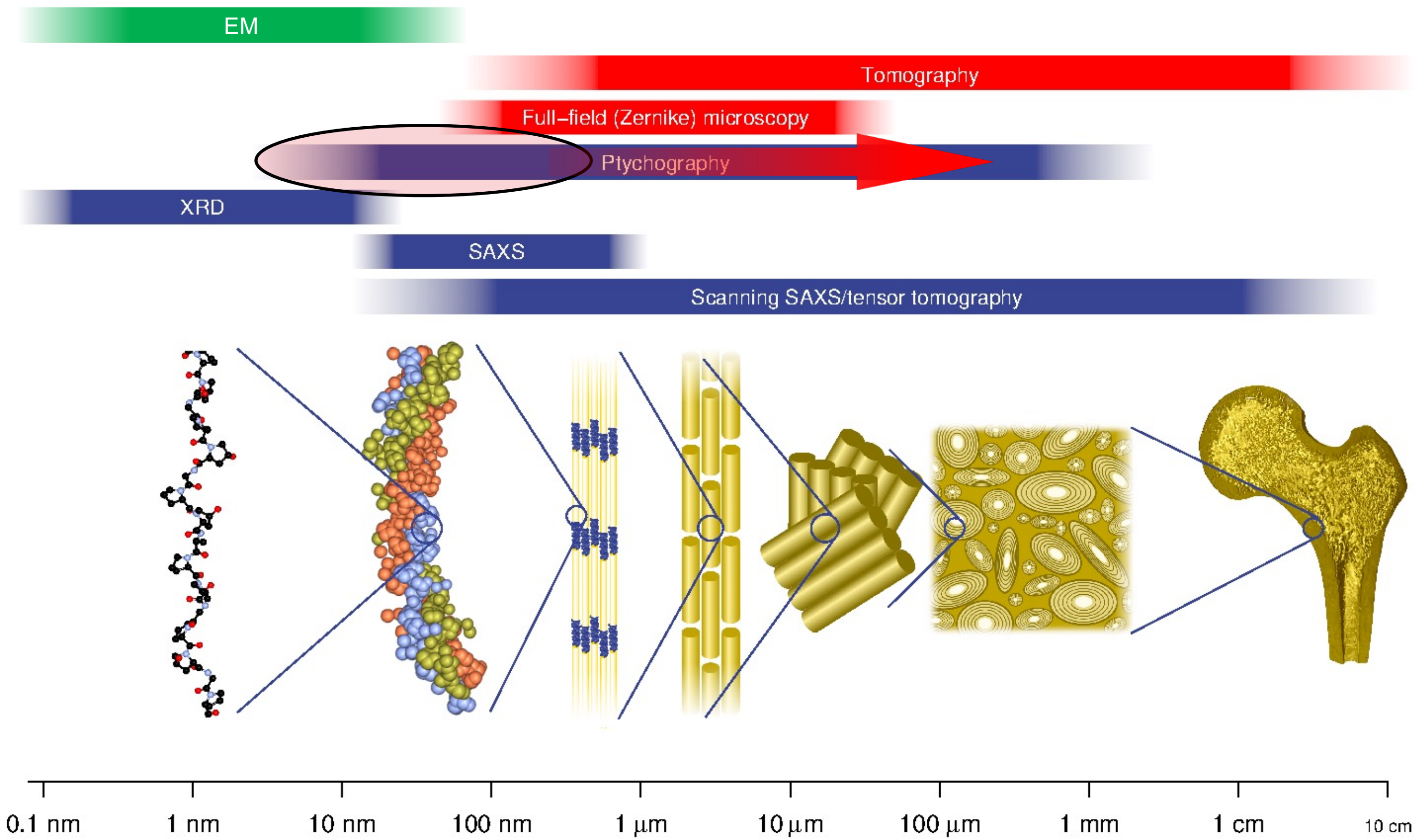


- Phase information arises from strains within the crystal
  - ⇒ Bragg CDI yields high resolution 3-D images of strain from within a nanocrystal in direction of Q
- Bragg diffraction away from (000) direction
  - ⇒ scattering object does not need to be physically isolated
  - Nonperiodic substrates or those with different lattice constants will be invisible to the diffraction process
  - ⇒ use Bragg CXDI to study the impact of an interface with the nanocrystal
- Several different Bragg spots (different Qs)
  - ⇒ 3-D strain tensor within nanocrystal

M.A. Pfeifer *et al.*, Nature **442**, 63–66 (2006) https://www.nature.com/articles/nature04867

Ptychography

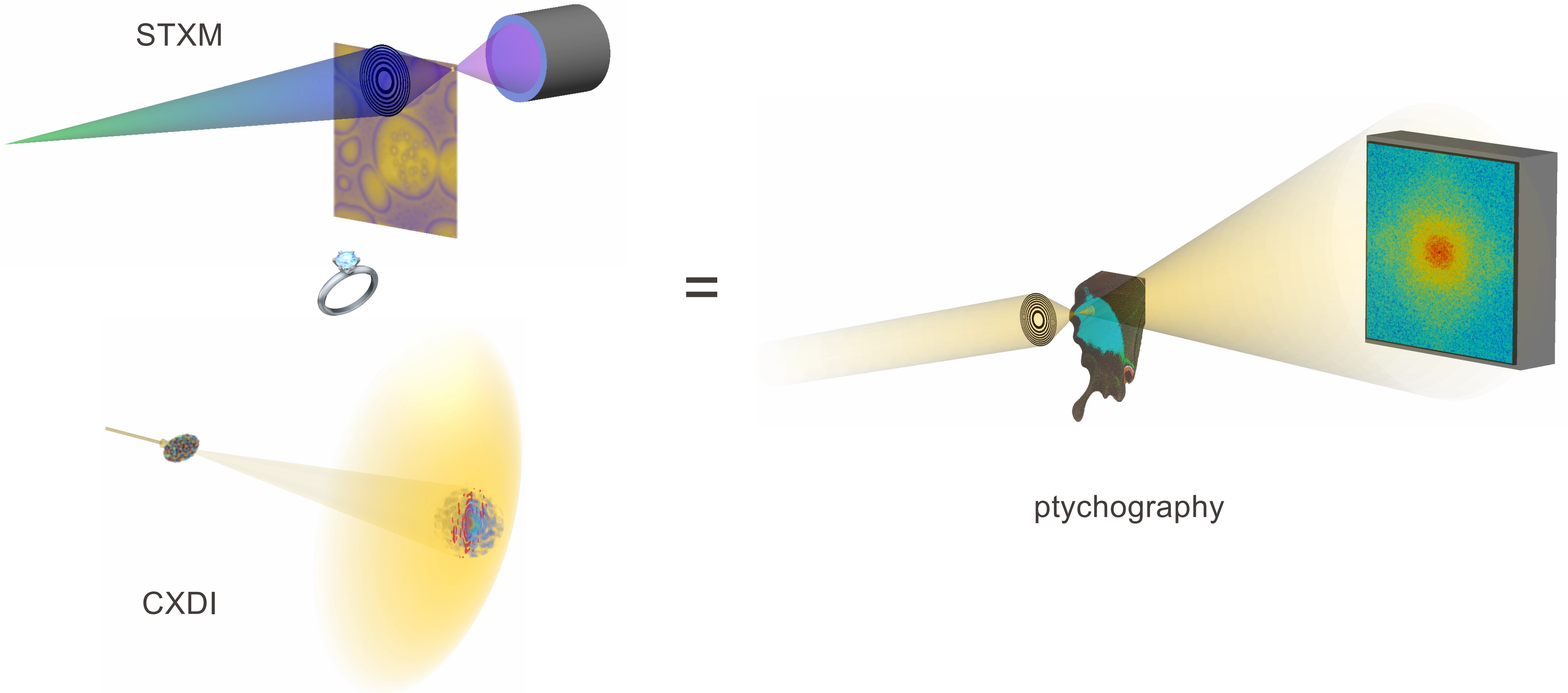
Role of ptychography



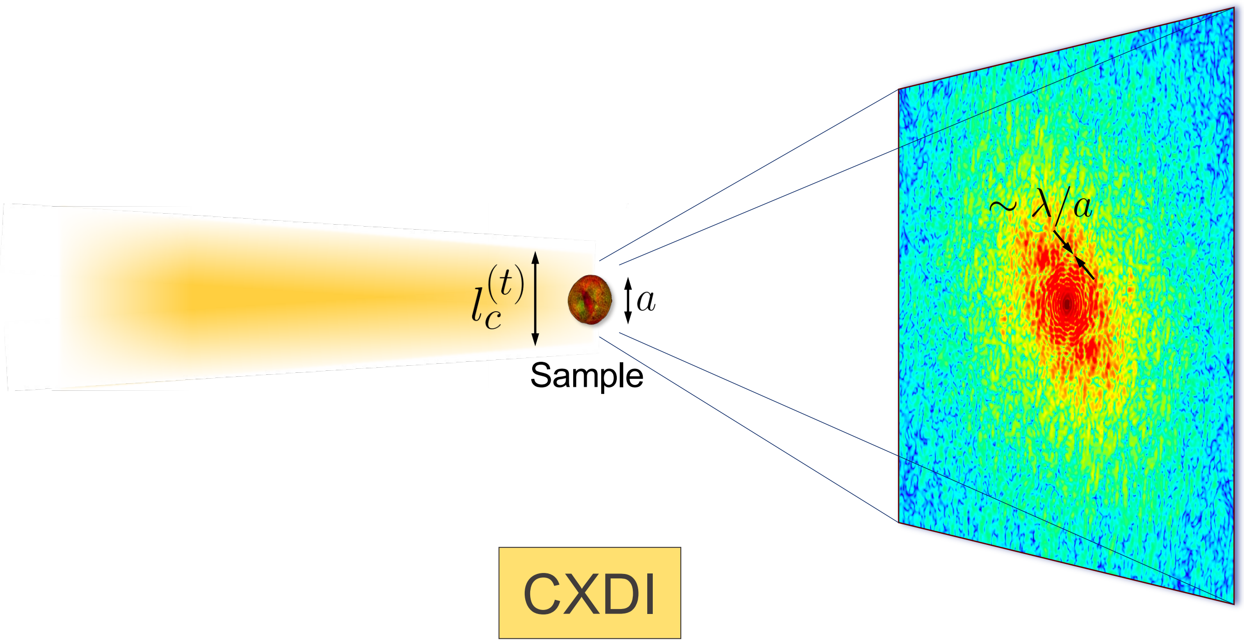
Architectural hierarchy of bone

- Bridge resolution gap between full-field tomographies and XRD/electron microscopy
- Scanning aspect allows high resolution down to few nm on extended samples with macroscopic dimensions limited only by IT considerations (and absorption lengths, not normally a problem for HXR)
- Spatial resolution determined by
  - Largest scattering angle
  - Stability of sample movements
  - NOT by size of illumination or step size

The perfect marriage



CXDI vs ptychography

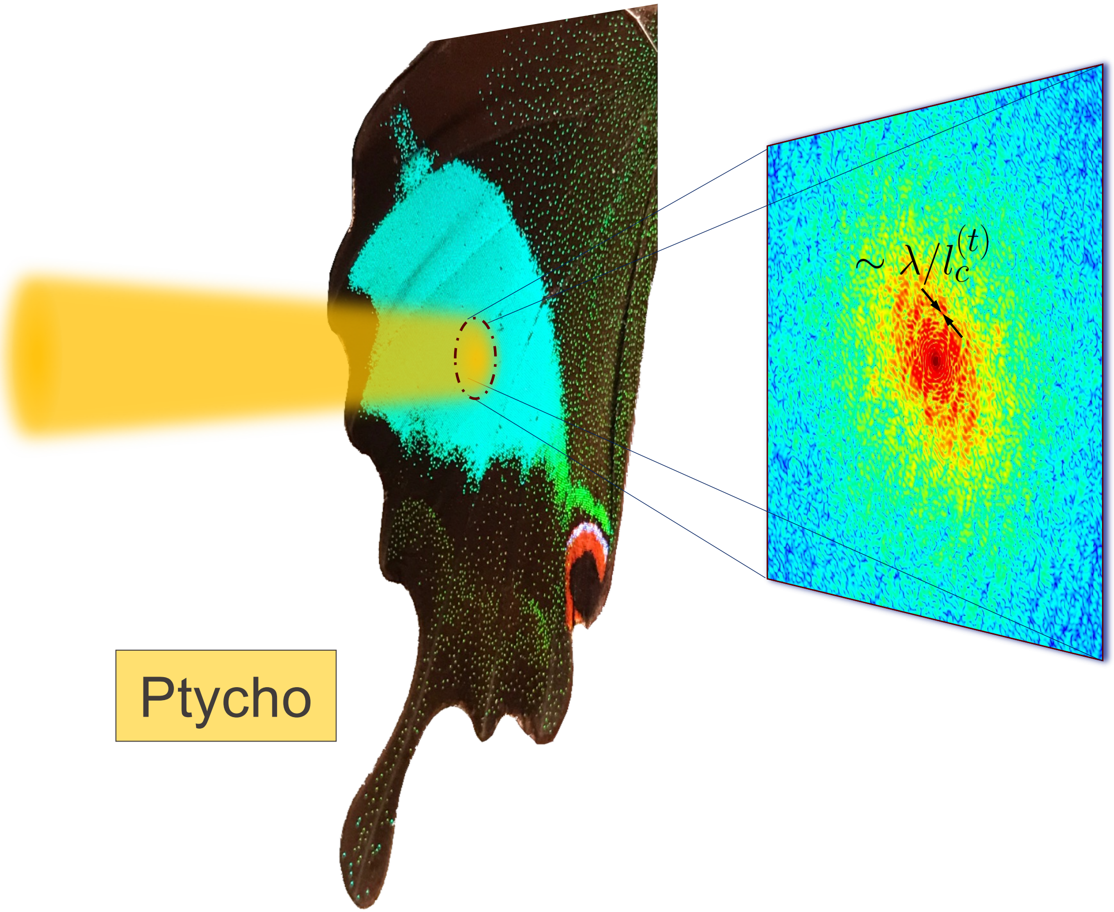


- Sample flooded with coherent radiation:

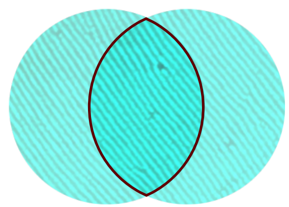
$$l_c^{(t)} > a$$

- Speckle and oversampling determined by sample size a
- Redundancy provided by
  - Positive electron density
  - Approximate maximum/minimum electron densities
  - Overall sample size (if known)

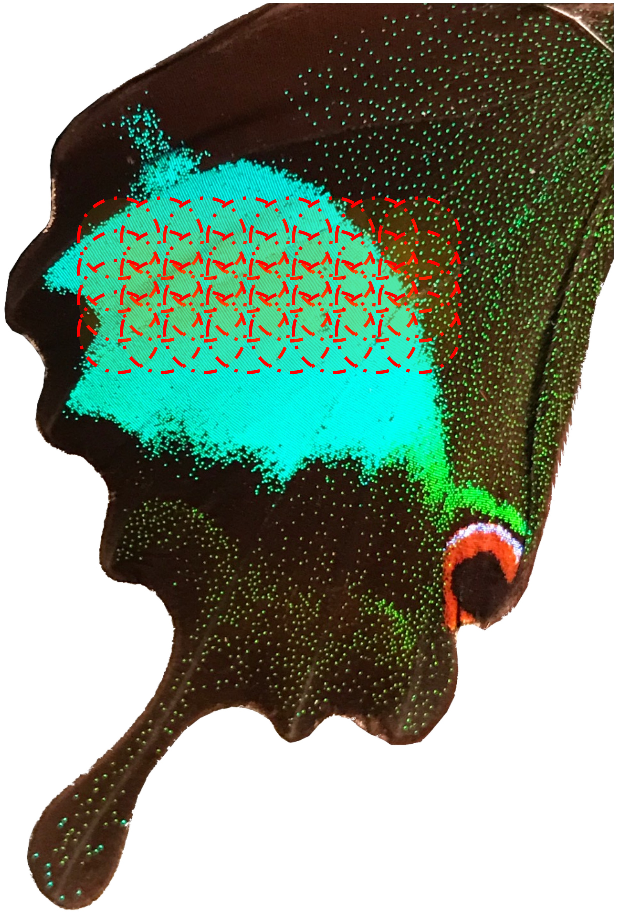
CXDI vs ptychography



- Extended sample
  - Larger than $l_c^{(t)}$
  - Part of sample illuminated with coherent radiation
- Speckle and oversampling determined by illumination size $l_c^{(t)}$
- Raster sample with step sizes < $l_c^{(t)}$
  - Marriage of CXDI and STXM
- Redundancy (real-space constraint) provided by
  - Overlap between adjacent recordings – solutions must be the same in real-space



Experimental considerations in ptychography



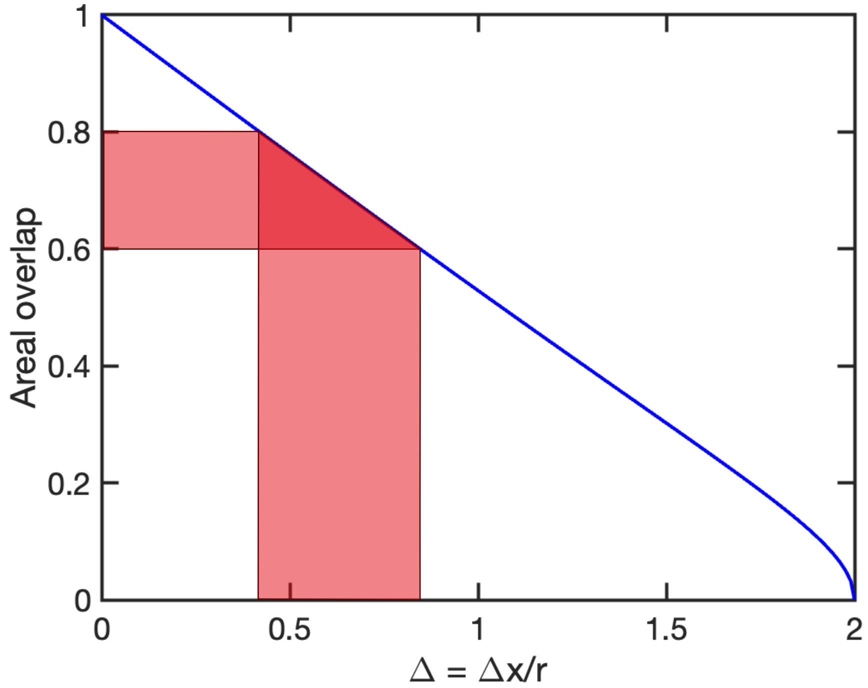
- HXR: ~ 0.5 – 2 Å
- SXR: near absorption edges e.g., magnetic materials
  - L-edges 600 – 900 eV
- Size of illumination ~ μm
- Depth of field* $T \sim 5(\Delta x)^2/\lambda$
  - Δx = desired resolution
  - ⇒ T ~ 1 – 10 μm for HXR and 10-nm resn.
- Optimal areal overlap† between adjacent illuminations ~ 60 – 80%
- Nested iteration to determine (imperfect) incident wavefront‡
- DLSRs: increase in coherent flux ~ 10^3!!

*M. Holler *et al*., Sci. Rep. **4** 3857 (2014)

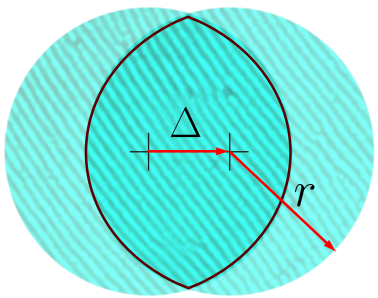
†O. Bunk *et al*., Ultramicroscopy **108** 481 (2008)

‡P. Thibault *et al*., Ultramicroscopy **109** 338 (2009)

Experimental considerations in ptychography



$$\text{Overlap} = \frac{2}{\pi}\arccos\left(\frac{\Delta}{2}\right) - \frac{\Delta}{2\pi}\sqrt{1-(\Delta/2)^2}$$



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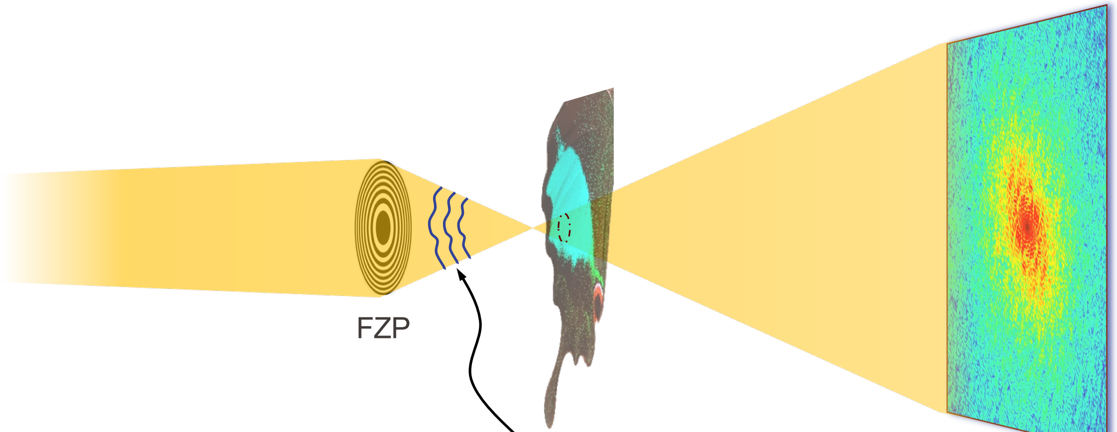

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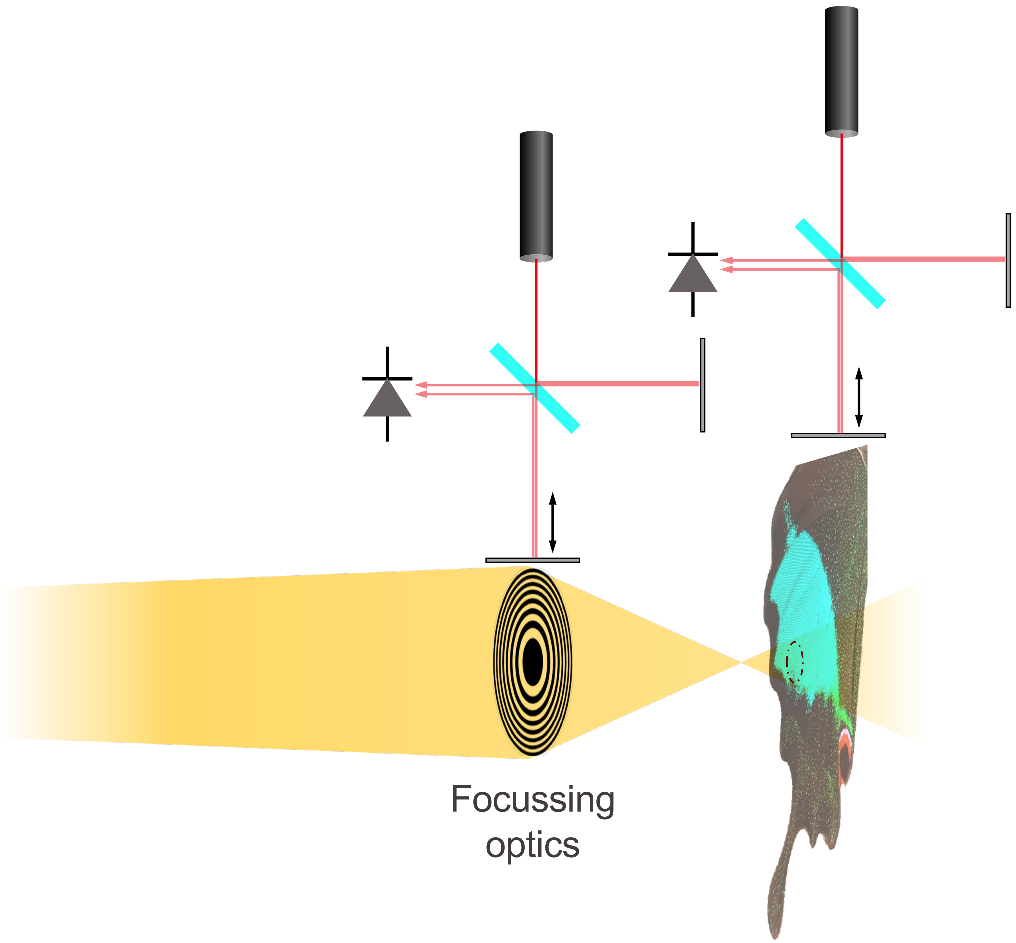
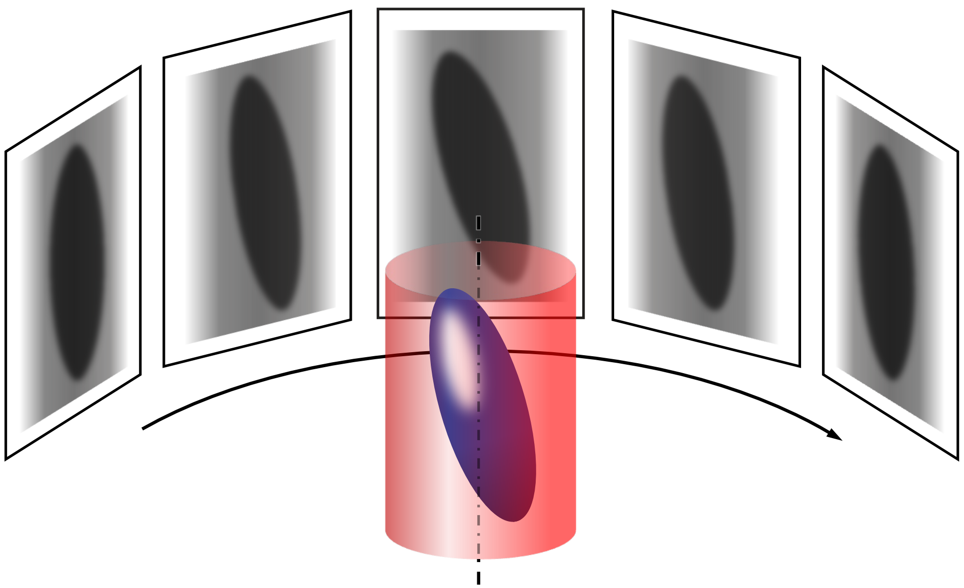
Sample manipulation



- Sample illumination accuracy better than desired spatial resolution
  - Optics (FZP, OSA, pinhole, etc) – fixed
  - Sample – controlled movements
  - Both x- and y-directions
- Interferometric control using lasers
- Avoid long-term drift
  - Especially problematic in cryogenically cooled samples
- In case of ptychographic tomography (PXCT, see next video) rotation control also required
  - Axis wobble

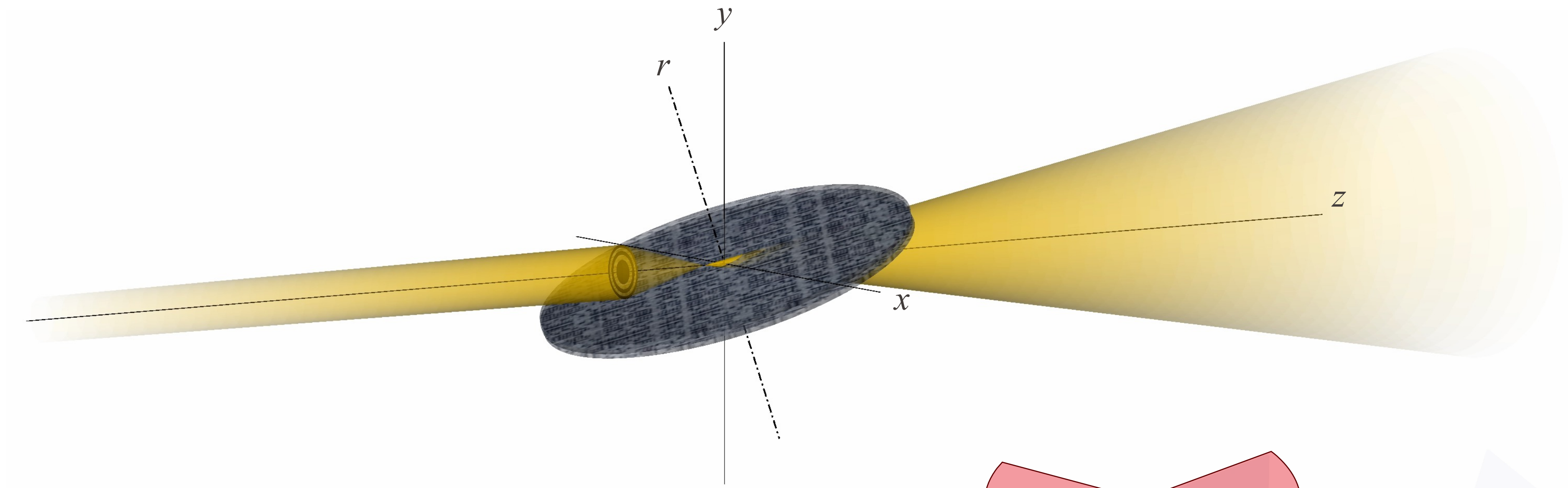
*M. Holler *et al.*, Sci. Rep. **4** 3857 (2014)

Ptychographic tomography

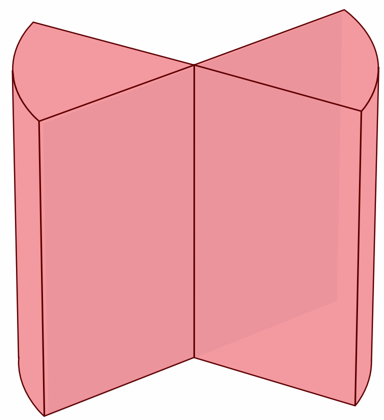


- PXCT combines
  - Ptychography
  - Tomography
- 3D reconstruction with ~ few 10s nm resolution
  - Ptychographic 2D reconstructions at different projection angles
  - Tomographic 3D reconstruction from ptychographic reconstructions
- Density variations < 1% possible

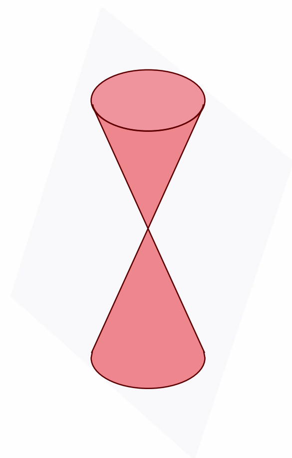
Ptychographic laminography



- Ptychographic x-ray laminography “PyXL”
- Used for extended samples in two dimensions (flat objects)
  - Sample rotation axis r tilted relative to x-y plane perpendicular to incident radiation (tomography)
  - Also scan sample laterally (ptychography)
  - Offset angle means some of reciprocal space cannot be accessed

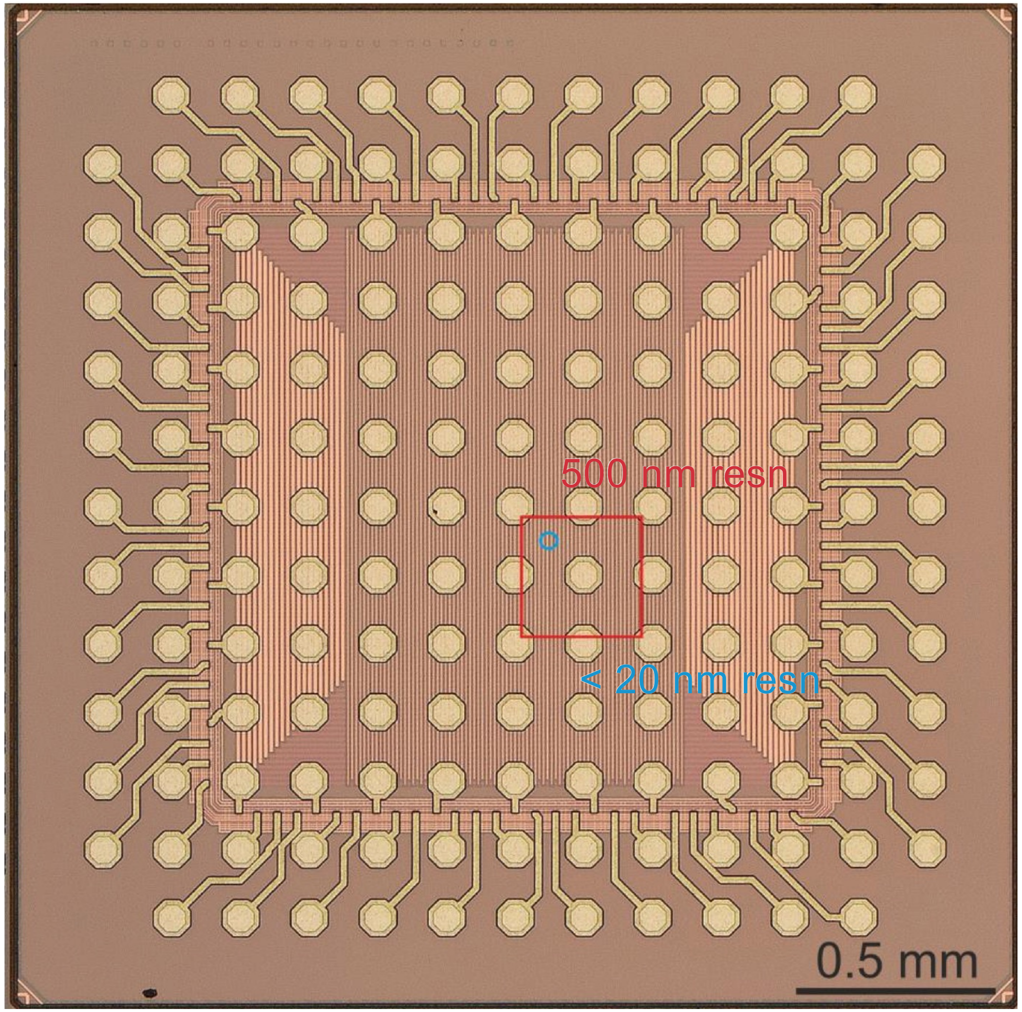


“Normal” tomography of flat objects



PyXL

Example – nondestructive study of chip architecture

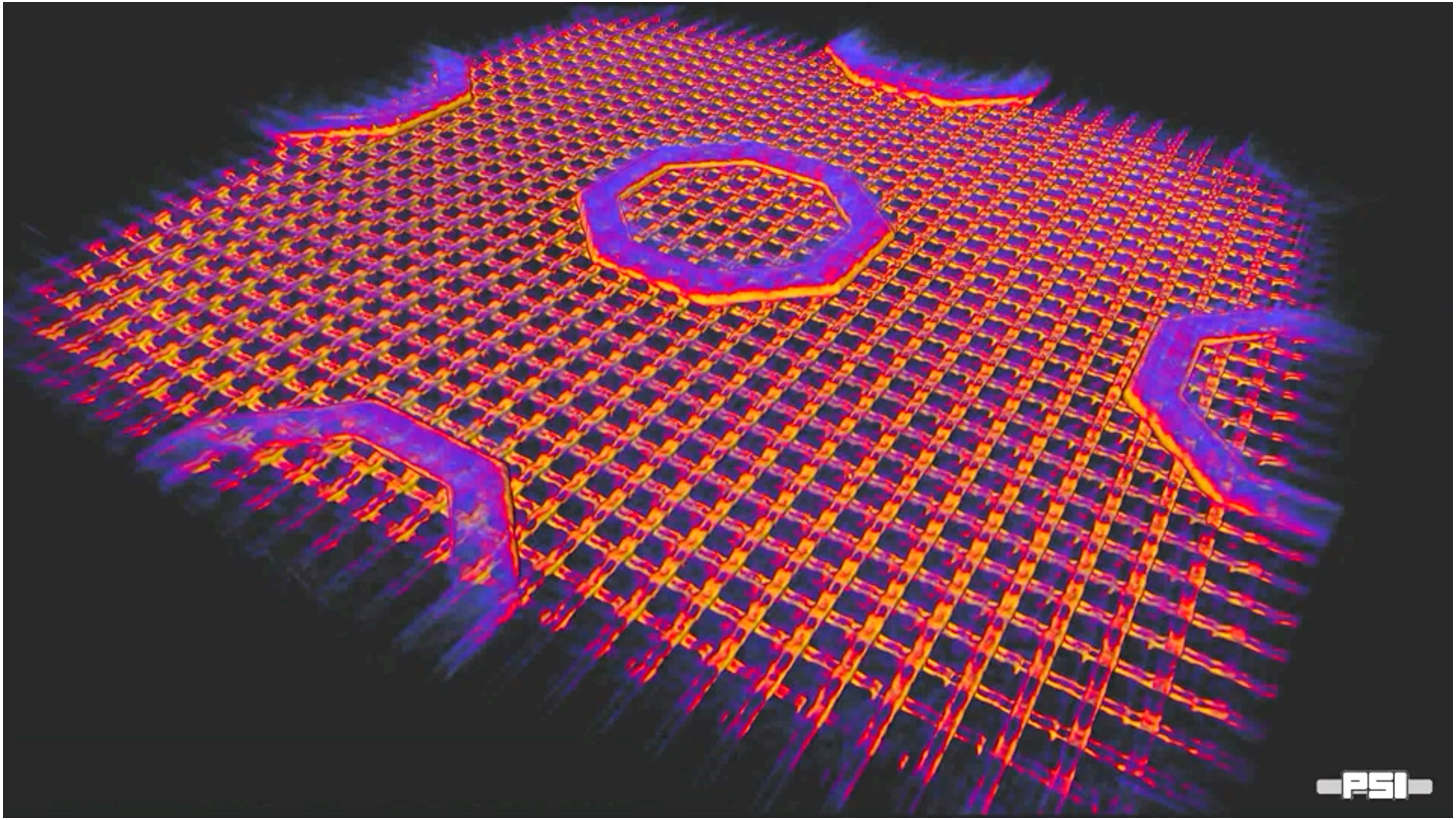


- PyXL @ 6.2 keV
- Tilt angle off vertical θ = 61°
- Integrated circuit chip with 16-nm fin field-effect transistor technology
- Size of radiation on sample T = 4 μm
- Number N of angular projections between 0 and 360° = 2872
- Theoretical resolution

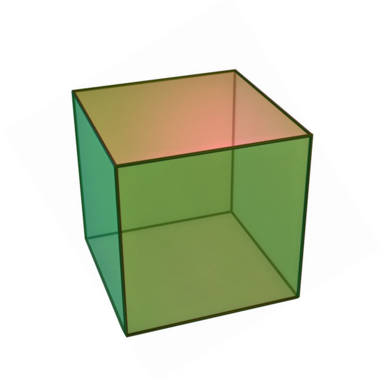
$$\Delta r = \pi \frac{T}{N} \tan\theta = 7.6 \text{ nm}$$

- Actual resolution 19 nm

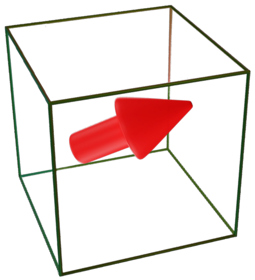
M. Holler *et al*. Nature Electronics
https://doi.org/10.1038/s41928-019-0309-z



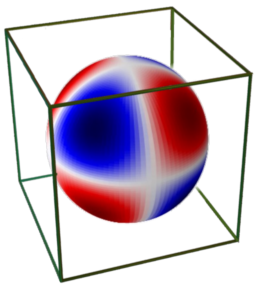
From scalar to vector to tensor



- “Standard” CXDI, ptychography, laminography yield scalar properties for each voxel
  - e.g., electron density

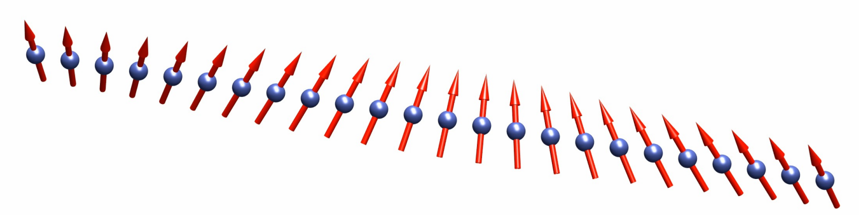


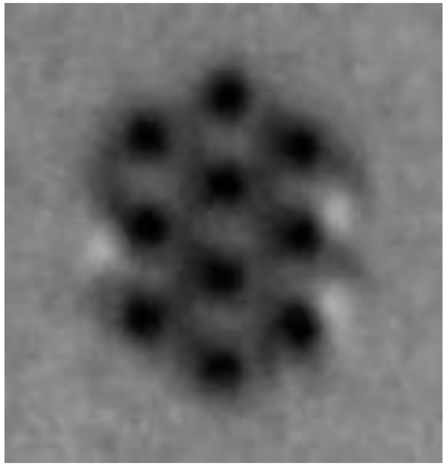
- Progress to directional properties in each voxel
  - Magnetic direction
  - Piezoelectricity
  - …



- Tensor properties within a voxel also possible
  - Full 3D spatial distribution of given property within voxel

Vector PXCT – nanomagnetism

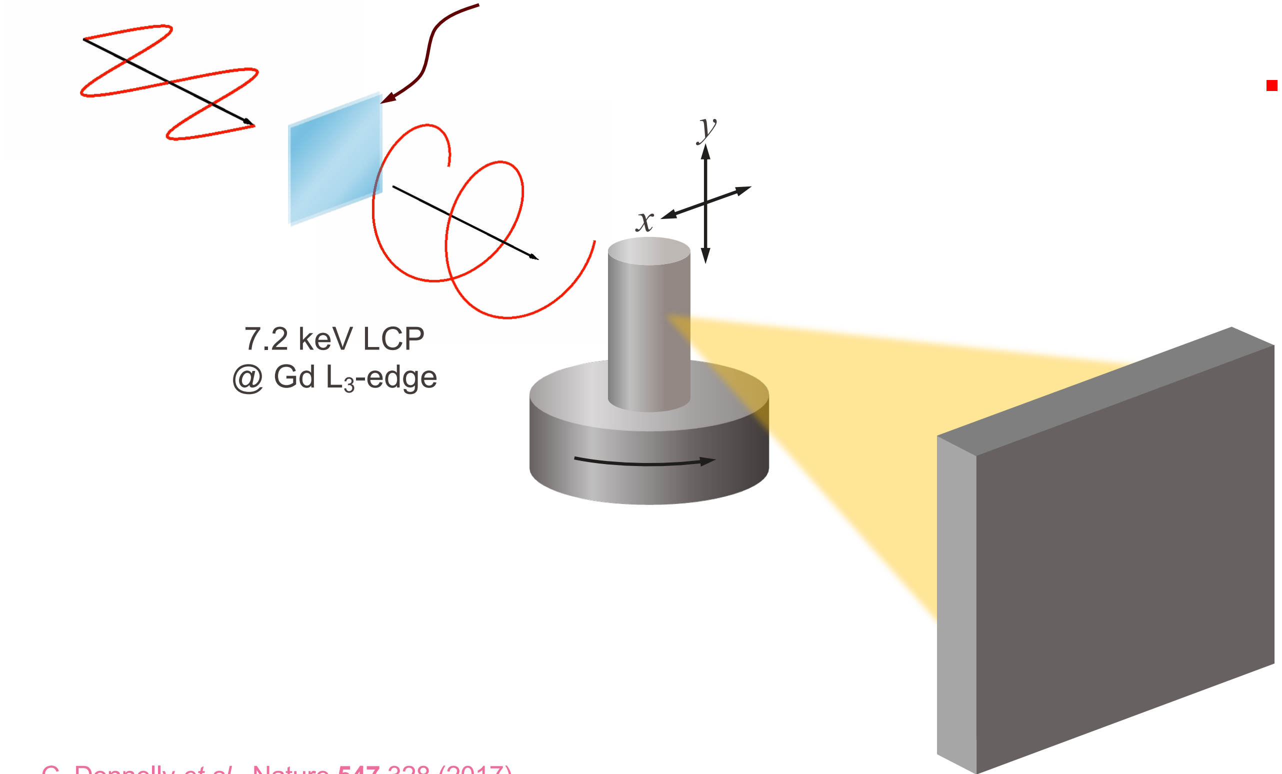


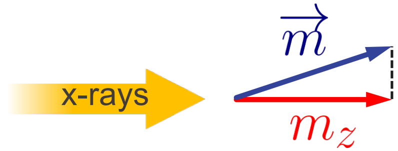


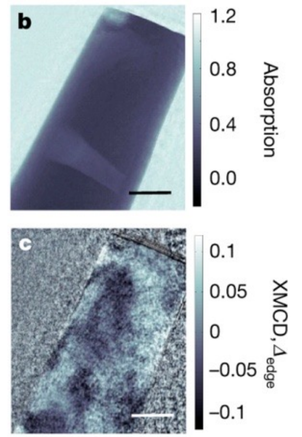
- 3D study of nanoscale magnetic materials
  - Basic research
  - Spintronics
  - Storage
  - Energy-harvesting industry
- Transmission electron microscopy
  - Limited to depths < 10 nm
- SXR microscopy
  - Limited to depths < 20 nm
- HXR transmission to many microns
  - $GdCo_2$
  - L-edge of Gd @ ca. 7.2 keV

Vector PXCT – magnetism

- $GdCo_2$ pillar, 5 μm diameter
  - Rotate and scan x- and y-directions
- Circular polarized x-rays sensitive to component of magnetization in the propagation direction
- Rotating the sample therefore changes the absorption coefficient
  - ⇒ domain contrast
  - In contrast to standard absorption in lensless imaging

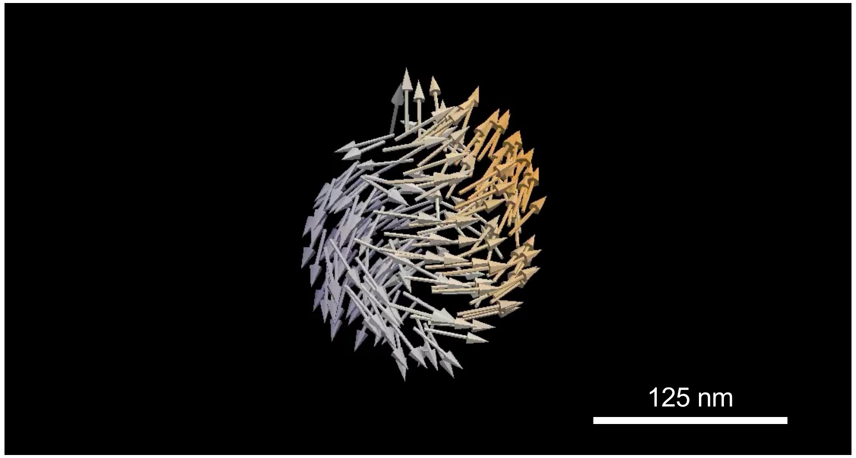


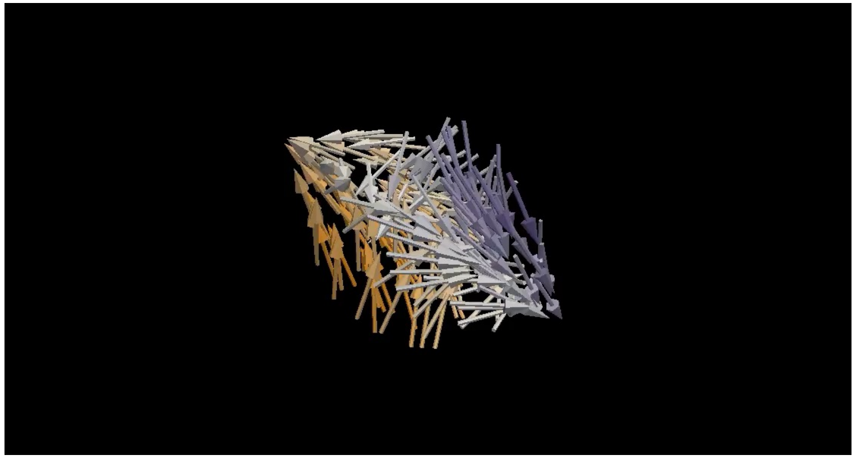




C. Donnelly *et al.*, Nature **547** 328 (2017)

Vector PXCT – magnetism

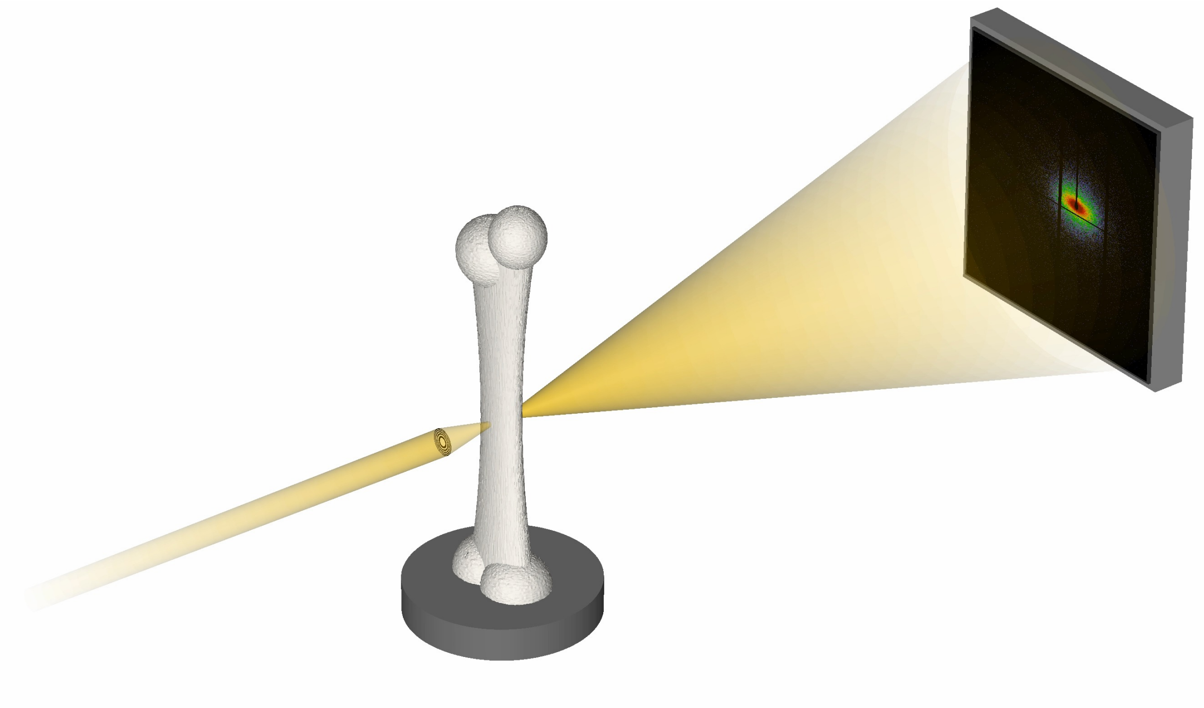




C. Donnelly *et al.*, Nature **547** 328 (2017)

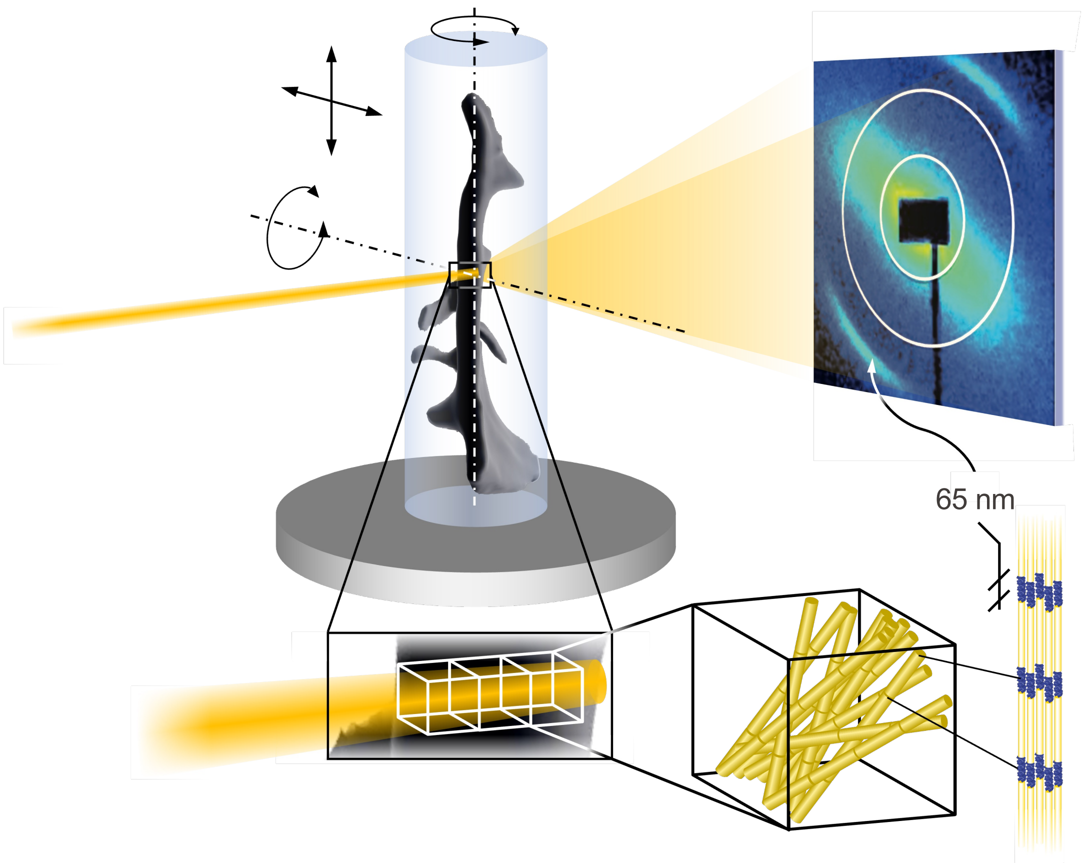
- "Magnetic vortices" form when electron spins swirl in a circle within a plane
  - At the centre of the circle, swirl becomes tighter and tighter – "Bloch point"
  - Eventually magnetization at the core tilts out of the plane
  - Prior to this work, magnetic vortices widely studied in 2D systems, but remained only a theoretical prediction in 3D
- Vortices shown to be surprisingly stable to both temperature and externally applied magnetic fields
  - Stability thought to be provided by pinning to domain walls

Scanning SAXS tensor tomography



- SSTT
- In each voxel (x, y, z) study a tensor property
  - Physical size
  - Intensity distribution
- Requires six degrees of freedom

SAXS tensor tomography



- $I(\theta,\phi,x_s,y_s,x_d,y_d)$
  - Tensor property
- Within a given voxel @ (x,y,z)
  - $I(Q,\theta,\phi)$
- Detector sees integrated signal of all voxel contributions along direction of propagation
  - Separate using tomographic methods

SAXS tensor tomography

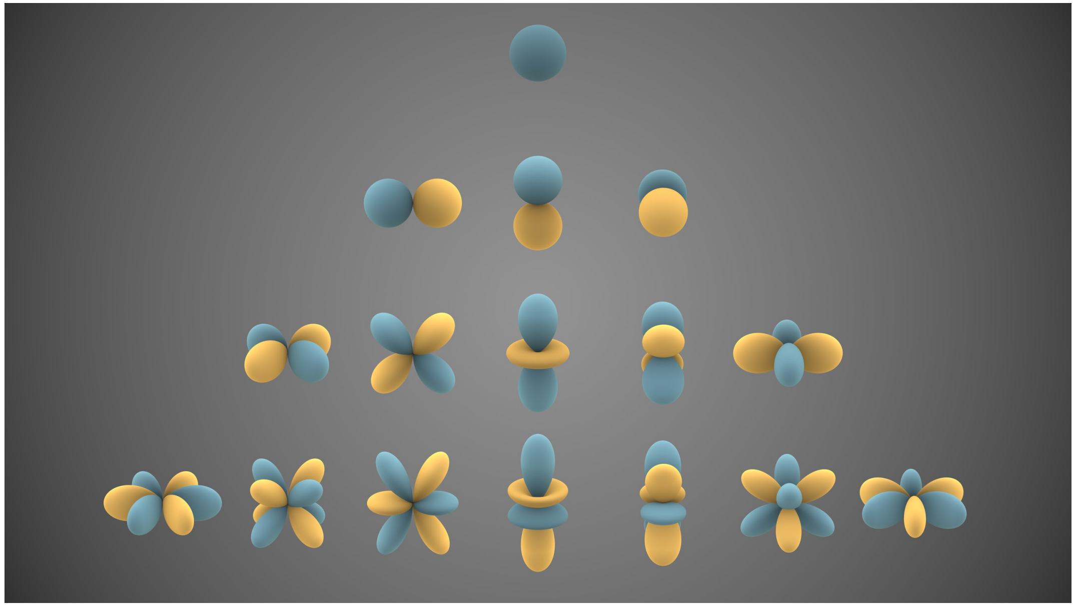
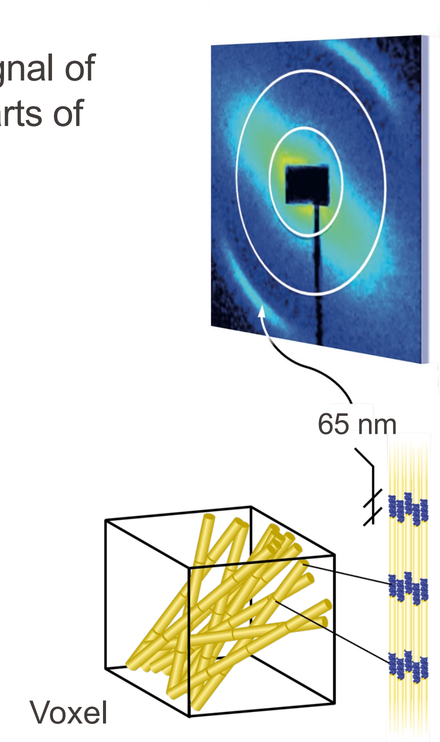
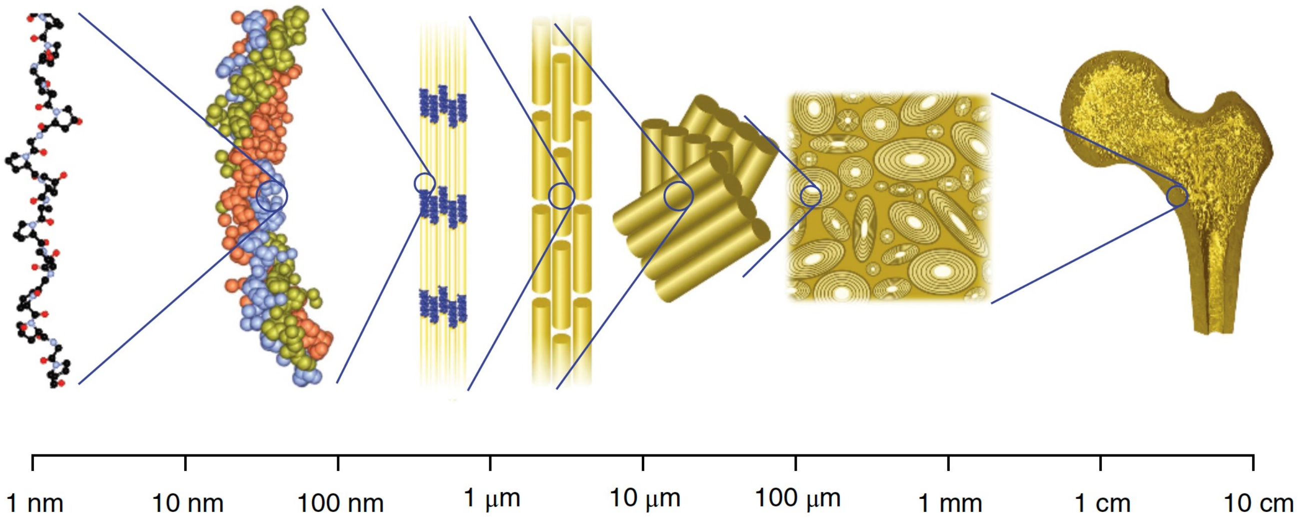


Image: Inigo.quilez Creative Commons

- Any distribution $I(\theta, \phi)$ can be described as a linear combination of spherical harmonics Y_l^m
- Angular distribution of intensity of given Q-value(s) associated with certain SAXS features
  - e.g., 65-nm signal of mineralized parts of collagen fibrils



SAXS tensor tomography – example: down to the bone

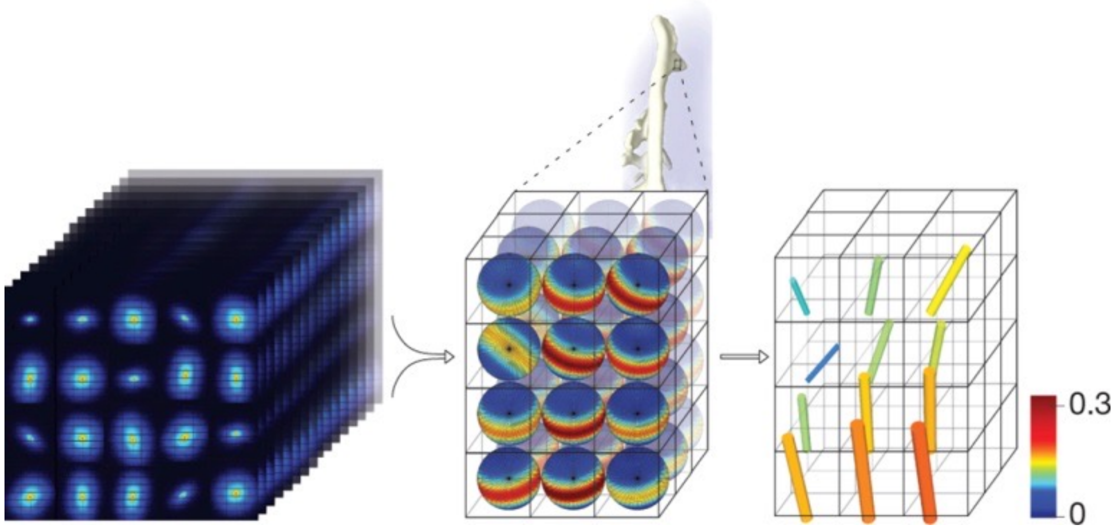


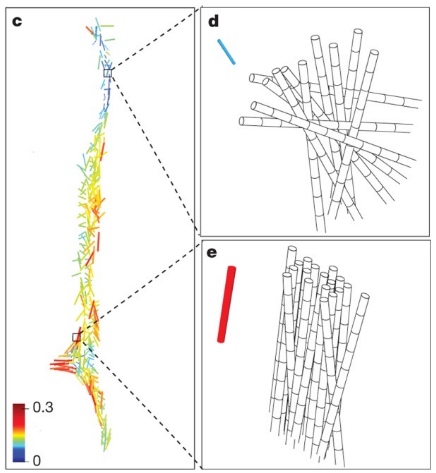
- Biology
  - Principle of hierarchical ordering
  - Maximize functionality
    - Strength
    - Robustness
  - Minimize weight and energy cost

- Bone
  - Multiple length scales
    - Collagen molecules (nm)
    - Microfibrils
    - Fibrils
    - Lamellae
    - Osteons (mm)

See M. Liebi *et al.*, https://doi.org/10.1038/nature16056

SAXS tensor tomography – example





- Human trabecular (spongy) bone
- Pencil beam ϕ = 25 μm
- Concentrate on 65-nm feature of mineralized collagen microfibrils
- > 10^6 SAXS patterns
  - Reconstruct 3D reciprocal-space map for each voxel
  - Model using spherical harmonics
    - Provides representation of nanoscale structure distribution
  - From reconstruction
    - Main ultrastructure orientation depicted by orientation of the cylinder
    - Degree of orientation the orientation of the cylinder illustrated by
      - Colour: indicating the ratio of anisotropic scattering to total scattering
      - Length of the cylinders: total scattering intensity

SAXS tensor tomography – example