Slides of the lecture on "X-ray coherence" used during the ESRF/ILL Summer School 2019

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X-ray coherence: introduction and a few applications

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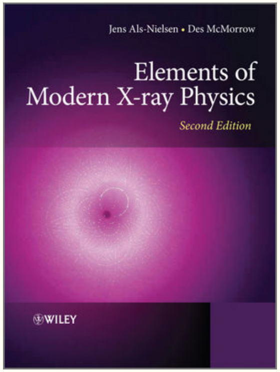
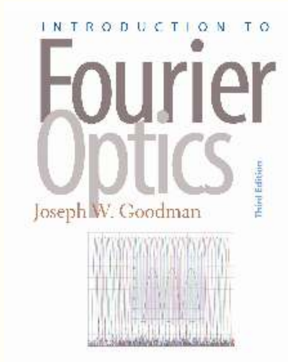
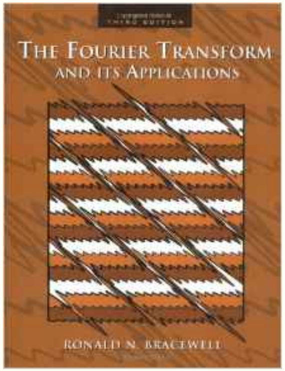
*sites.google.com/view/jcesardasilva

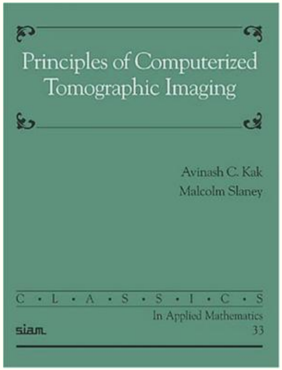
Some books to help

Fundamental ones:

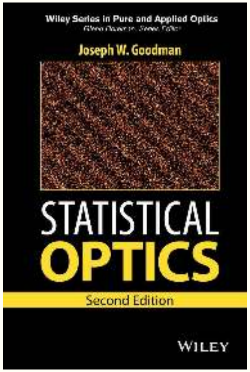


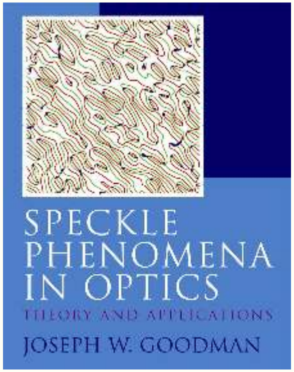


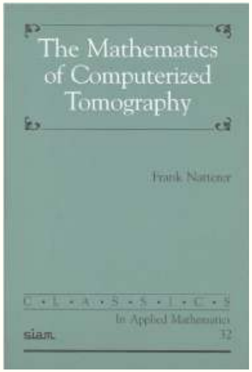


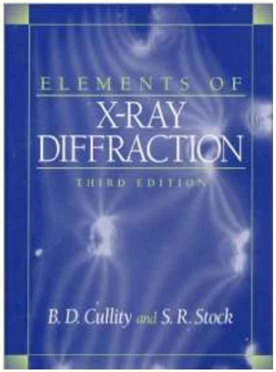


Also important ones:

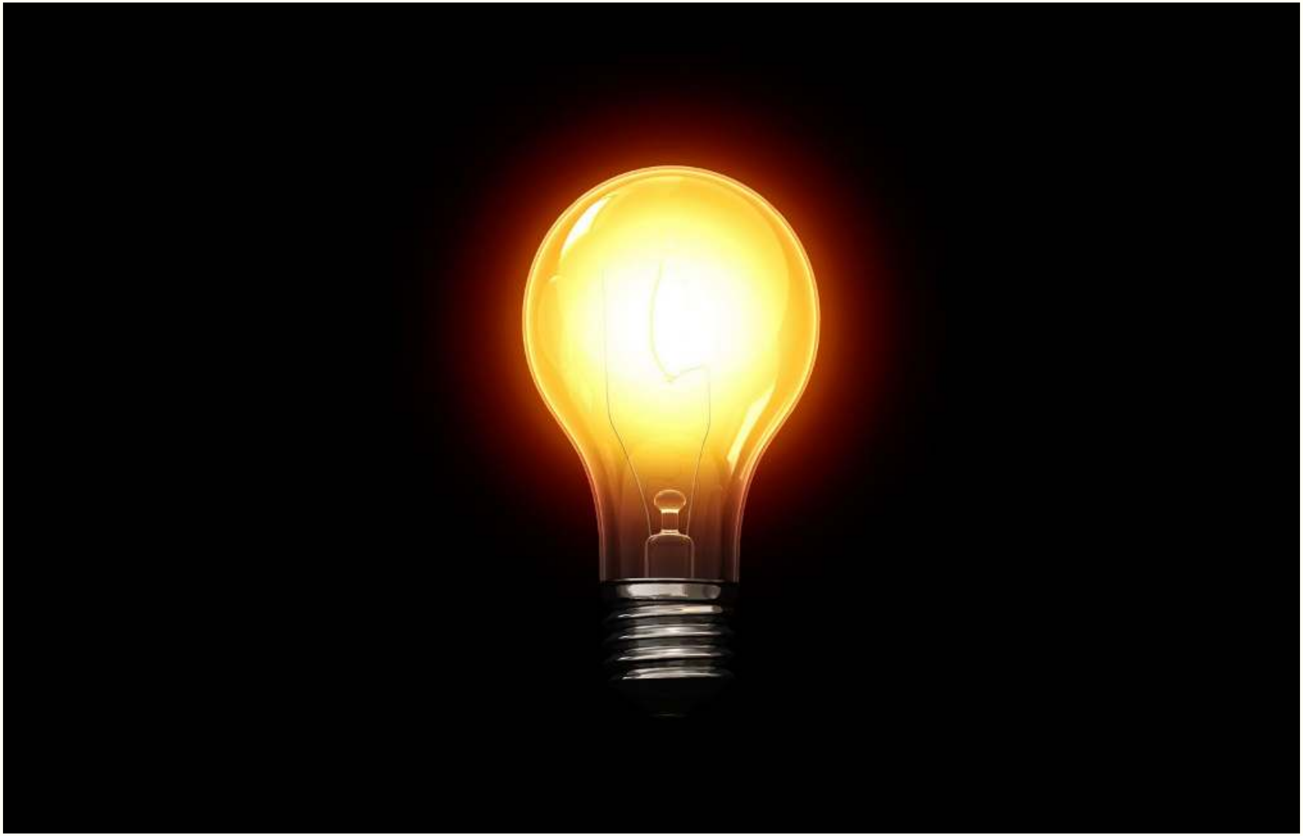




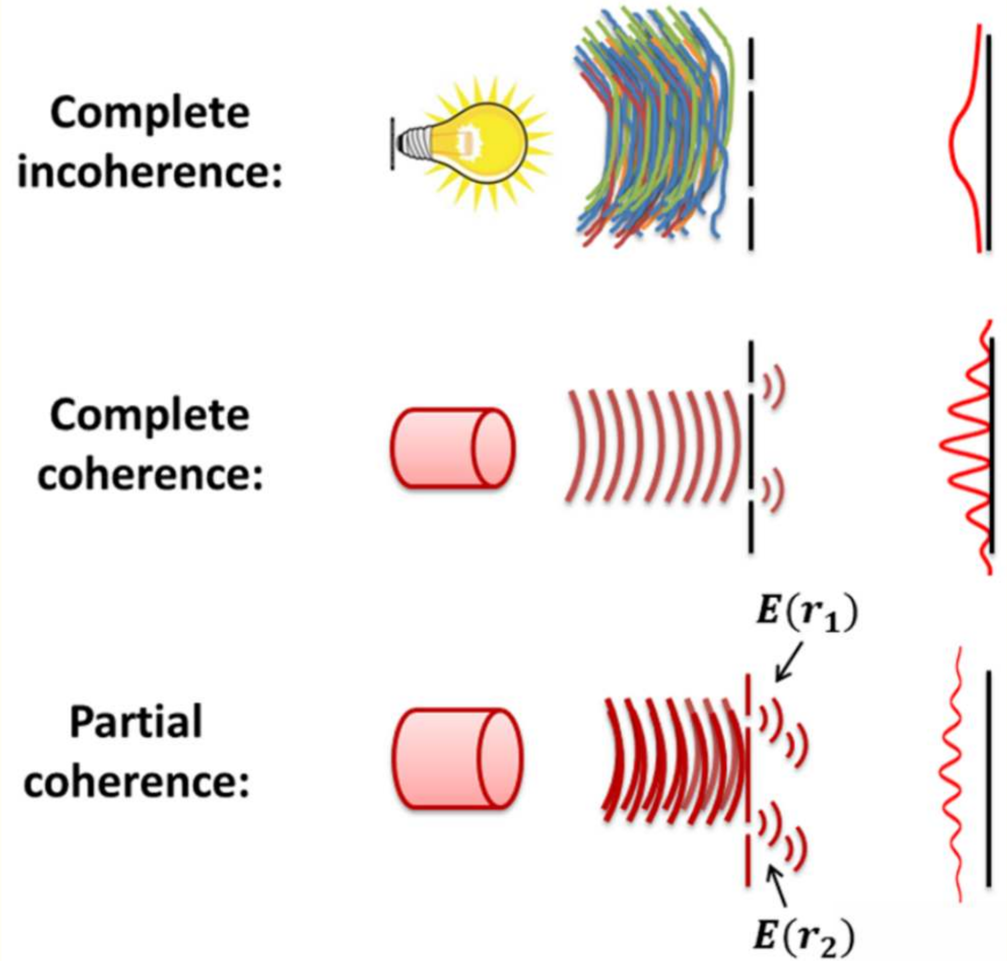




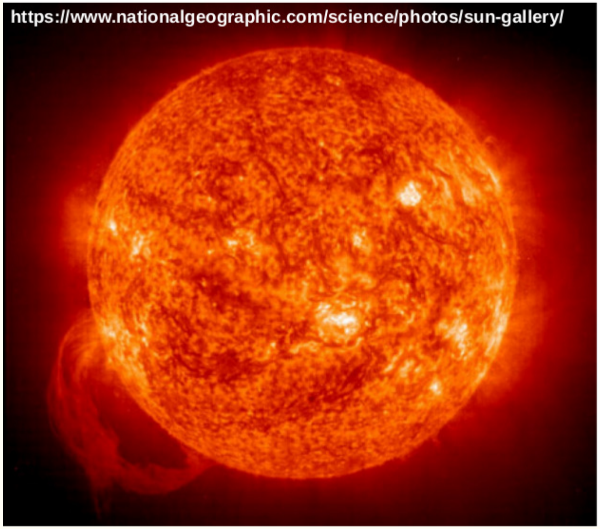
What is light?



Is the light coherent?



Is the sunlight coherent?



Coherent Area (A_c) $\sim 4 \times 10^{-3}$ mm^2

S. Divitt and L. Novotny, Optica 2(2), 95-103 (2015)
https://skullsinthestars.com/2010/06/12/you-could-learn-a-lot-from-a-ducky-the-van-cittert-zernike-theorem/

The light of a star

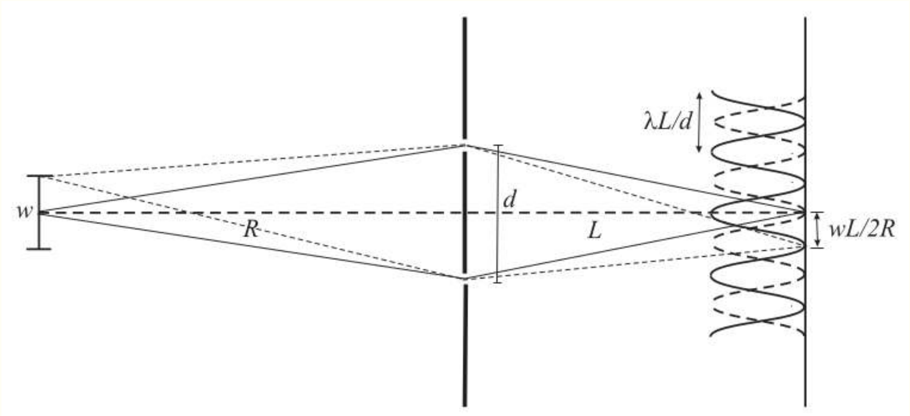
Coherent Area (A_c) $\sim 6\ m^2$

https://skullsinthestars.com/2010/06/12/you-could-learn-a-lot-from-a-ducky-the-van-cittert-zernike-theorem/

The van Cittert-Zernike theorem

W. H. Knox, M. Alonso, E. Wolf, Physics Today 63, 3, 11 (2010)

Transverse Coherence

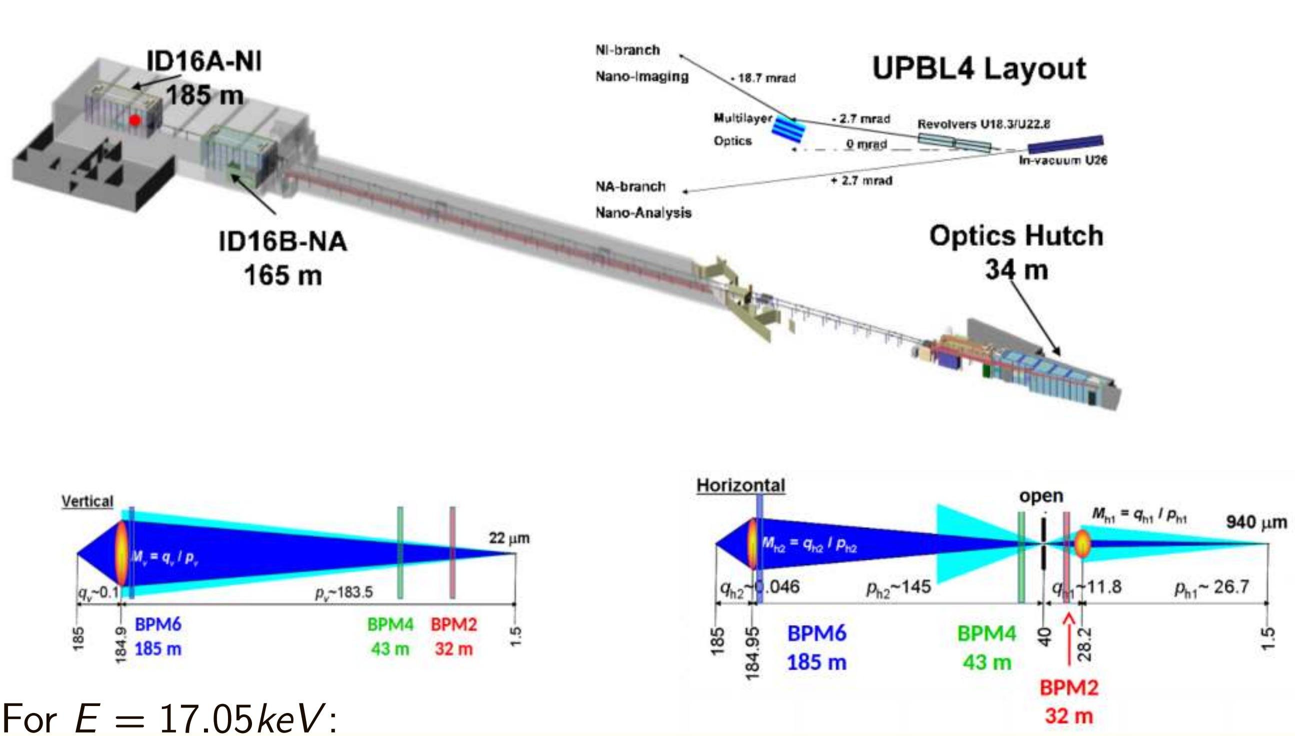


Diffraction patterns from two narrow slits at distance d, originating from the central part of the source (solid curve) and from the edge of the source at height w/2 (dashed curve). The slit distance d is such that the two patterns are in antiphase at $d = \lambda R/w$. This is defined as **transverse coherence length**:

$$\xi_t = \frac{\lambda R}{w}$$

F. van der Veen, F. Pfeiffer, J. Phys.: Condens. Matter 16 (2004) 5003.
J. Goodman, Statistical Optics (book)

ID16A Nano-Imaging beamline - ESRF

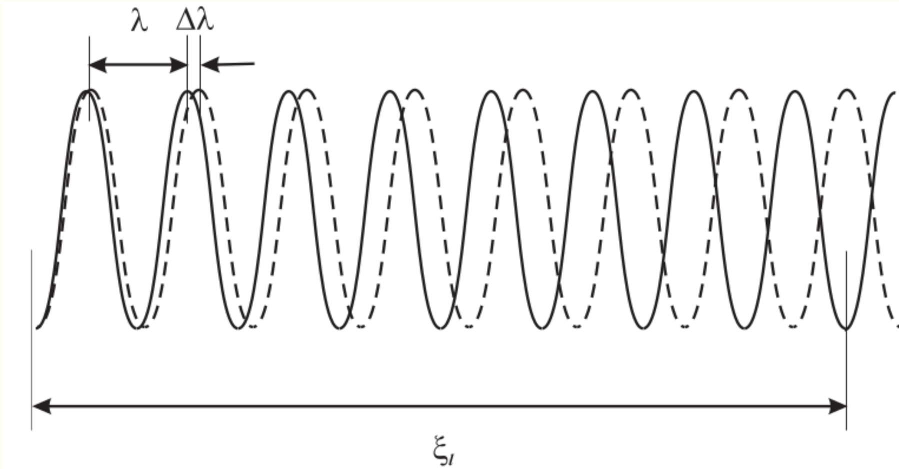


For $E = 17.05 keV$:

$$\xi_{tv} = \frac{\lambda R}{w} \approx 611.6\ \mu m \qquad \xi_{th} = \frac{\lambda R}{w} \approx 210.9\ \mu m$$

J. C. da Silva, et al., J. Synchrot. Radiat. 26, 1751-1762 (2019)

Longitudinal Coherence

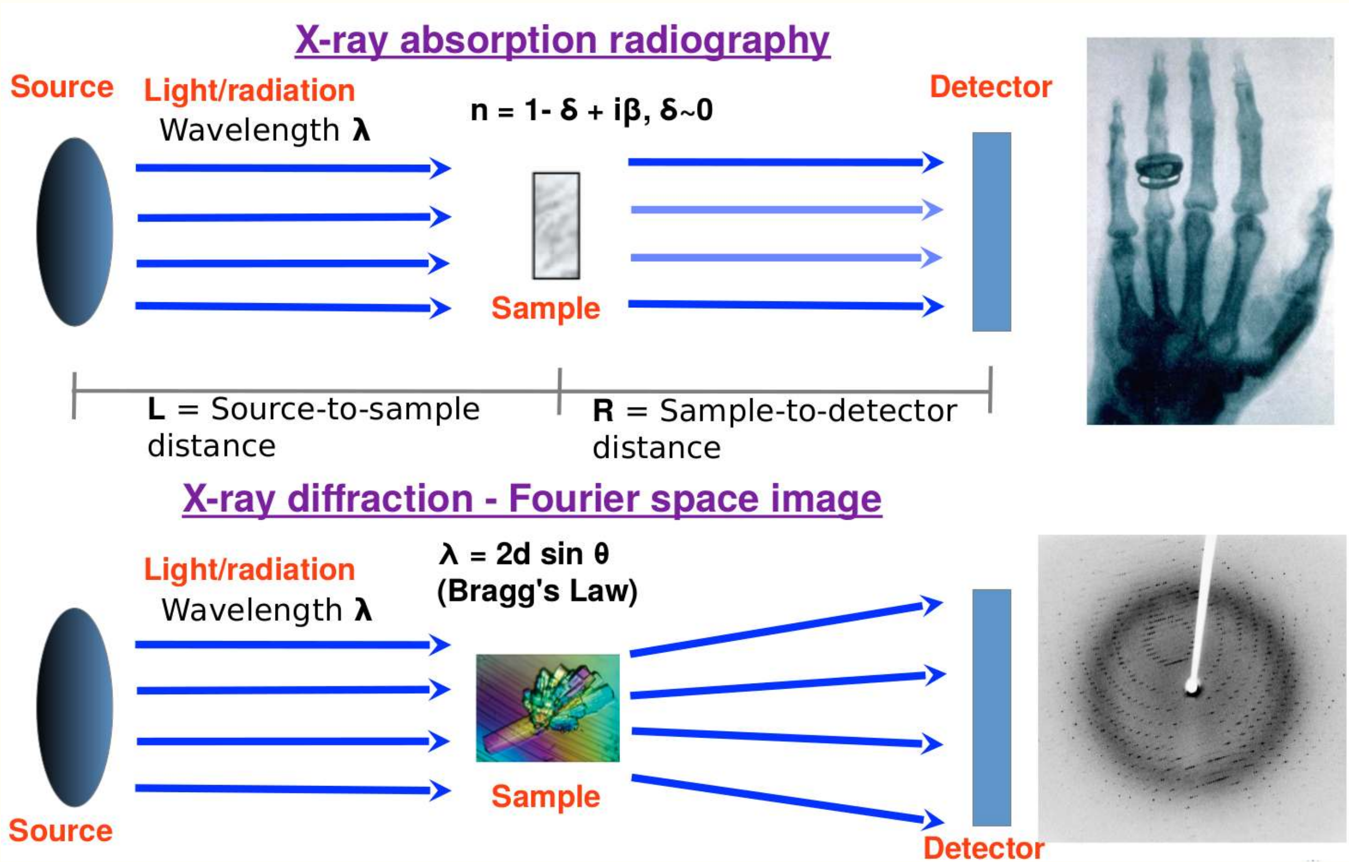


One has $N\lambda = (N - 1/2)(\lambda + \Delta\lambda)$. Solving for N and substituting in $\xi_l = N\lambda$ we find the **longitudinal coherence length**:

$$\xi_l = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

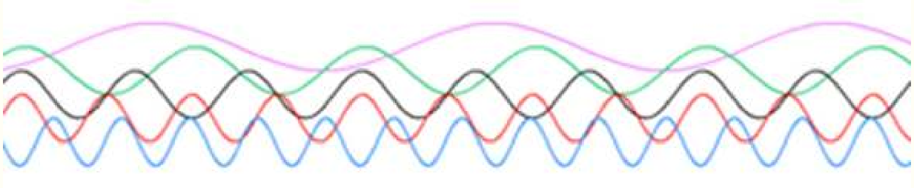
F. van der Veen, F. Pfeiffer, J. Phys.: Condens. Matter 16 (2004) 5003.
J. Goodman, Statistical Optics (book)

Classical X-ray experiments



Beam coherence

Incoherent waves with different frequencies (not monochromatic)



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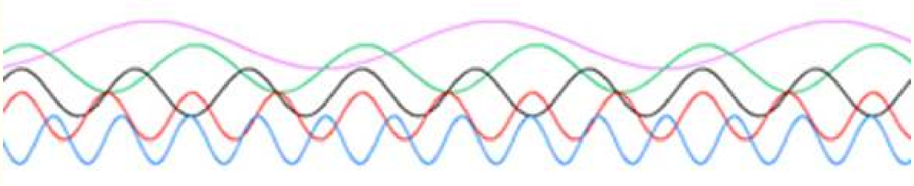

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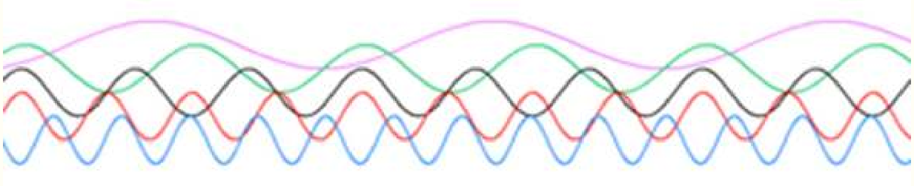
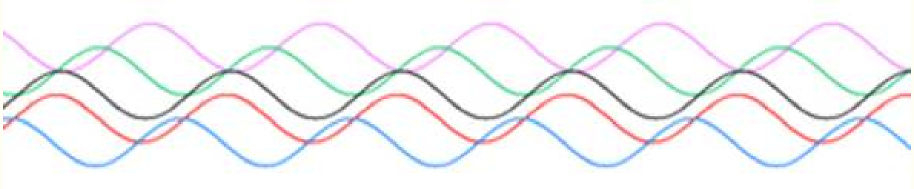
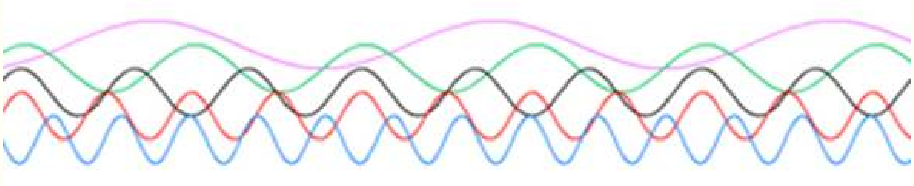


Incoherent waves with same frequency (monochromatic)



Beam coherence

Incoherent waves with different frequencies (not monochromatic)



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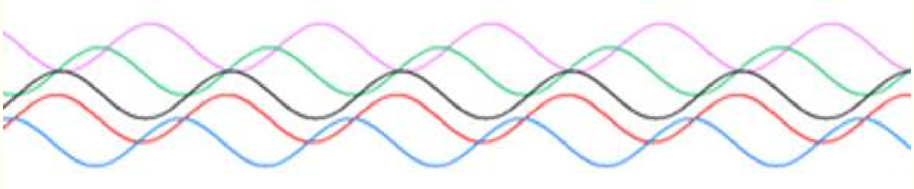
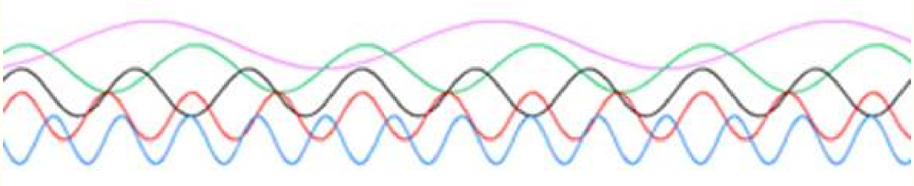
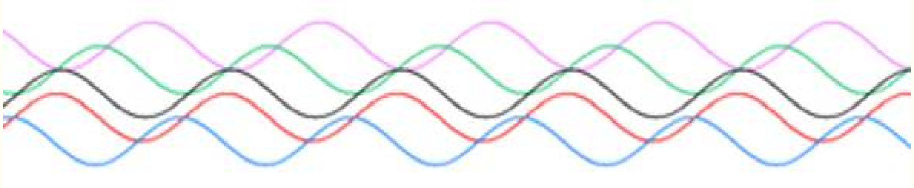


Beam coherence

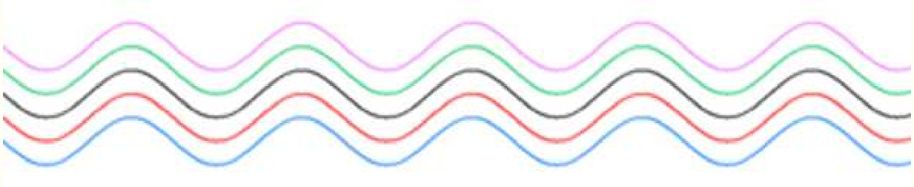
Incoherent waves with different frequencies (not monochromatic)



Incoherent waves with same frequency (monochromatic)

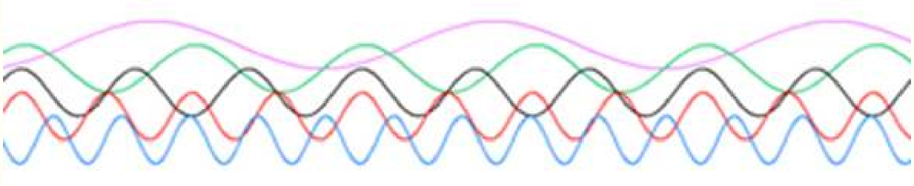


Coherent waves (monochromatic)

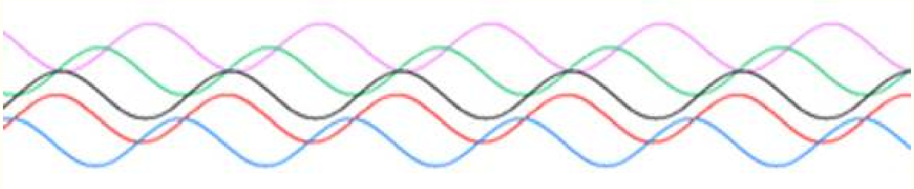


Beam coherence

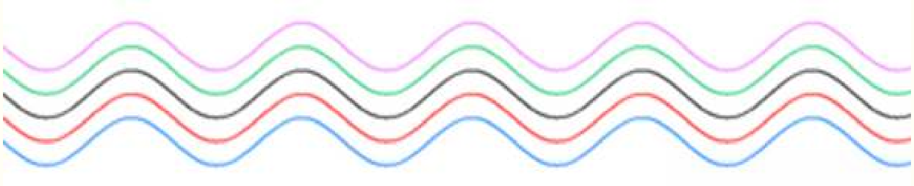
Incoherent waves with different frequencies (not monochromatic)



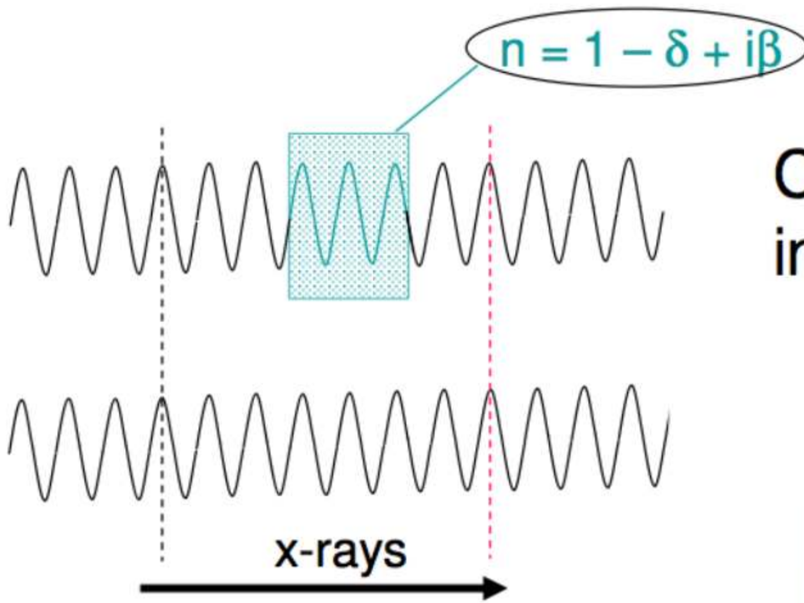
Incoherent waves with same frequency (monochromatic)



Coherent waves (monochromatic)

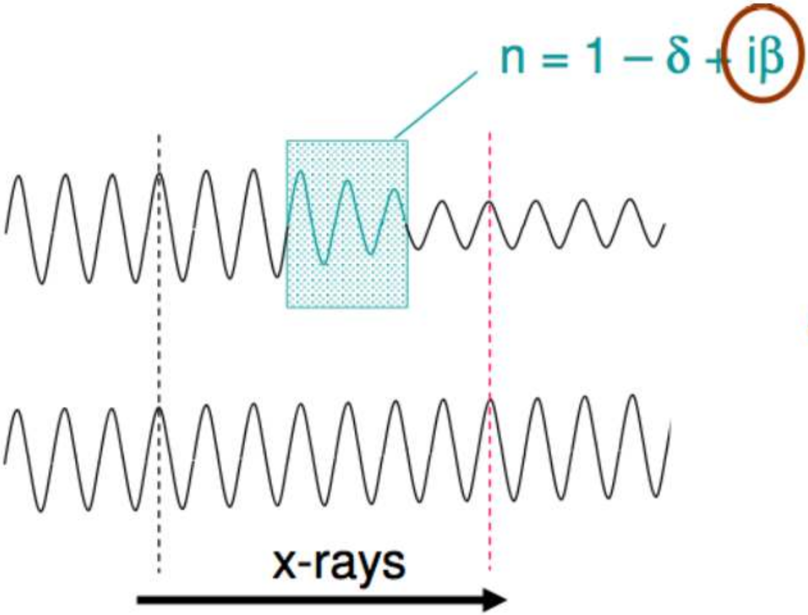


Absorption vs. Phase contrast



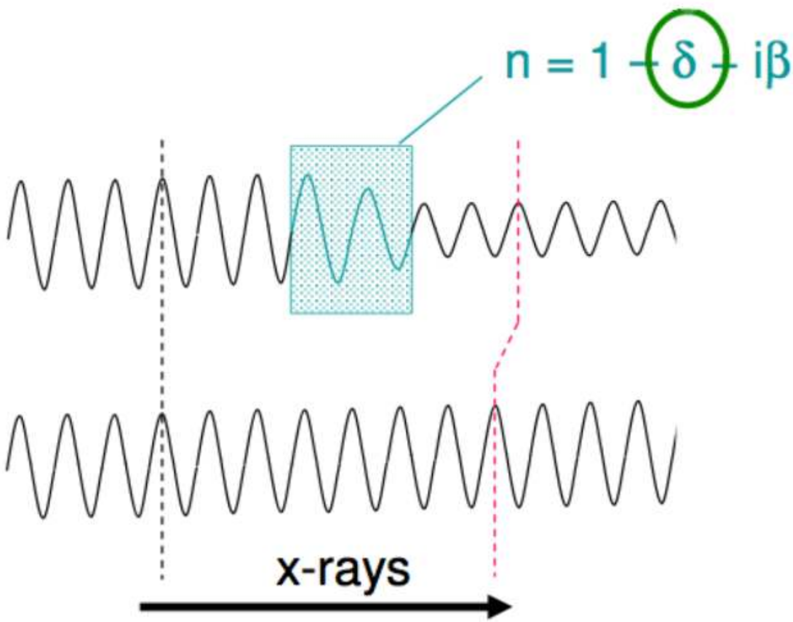
Complex-valued index of refraction

Absorption vs. Phase contrast



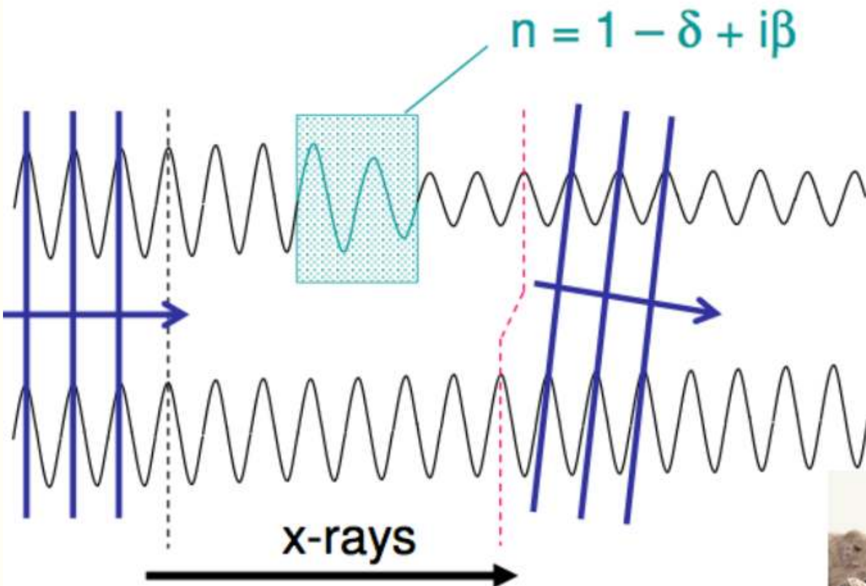
Absorption

Absorption vs. Phase contrast



Phase shift

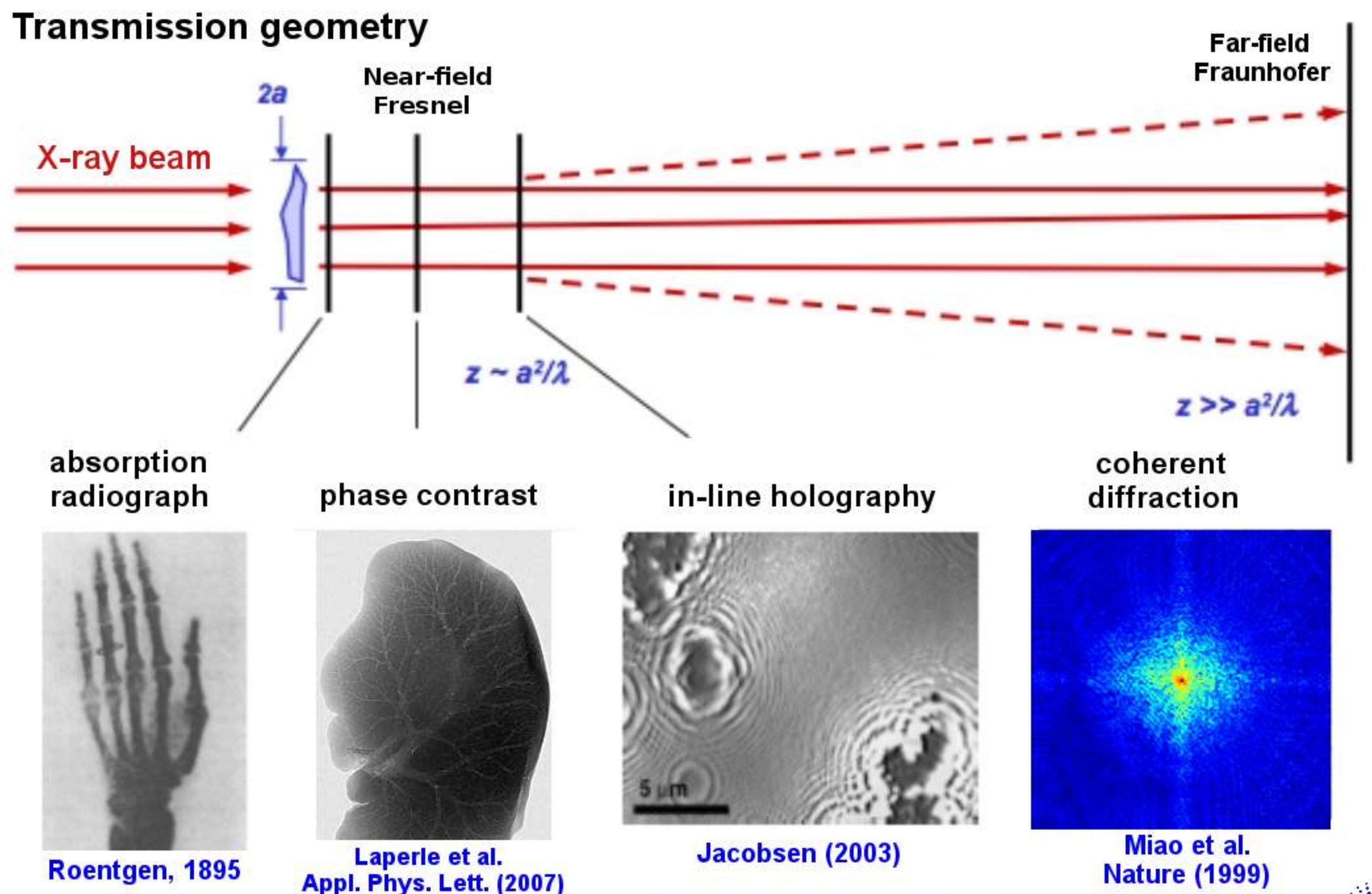
Absorption vs. Phase contrast



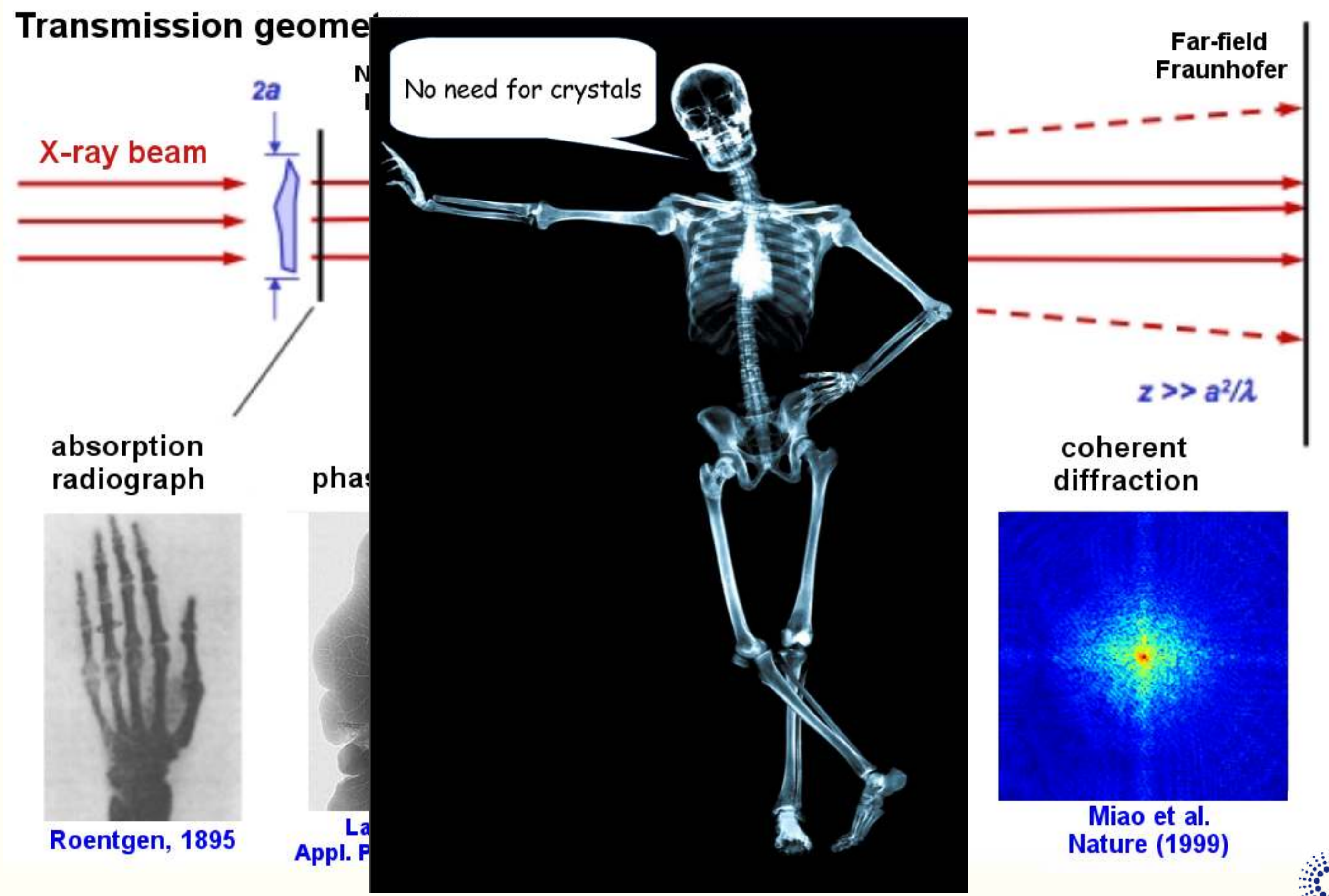
Phase shift

→ Refraction

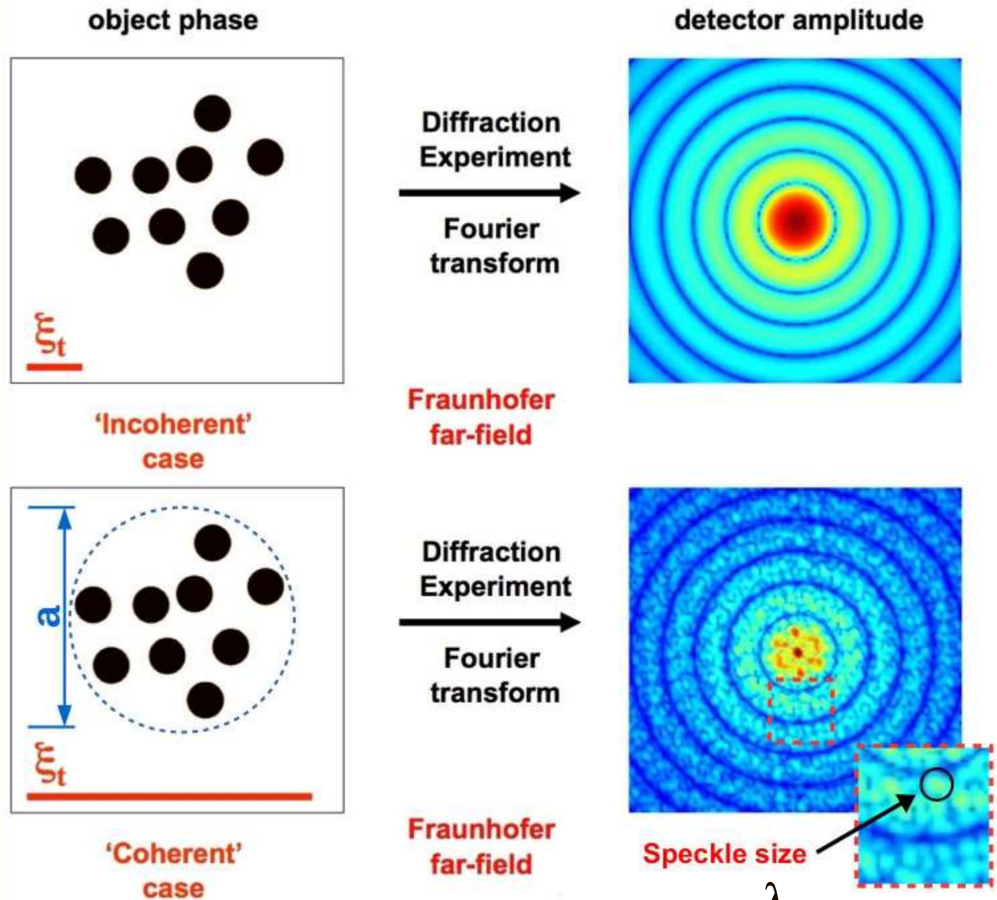
Different X-ray imaging regimes



Different X-ray imaging regimes



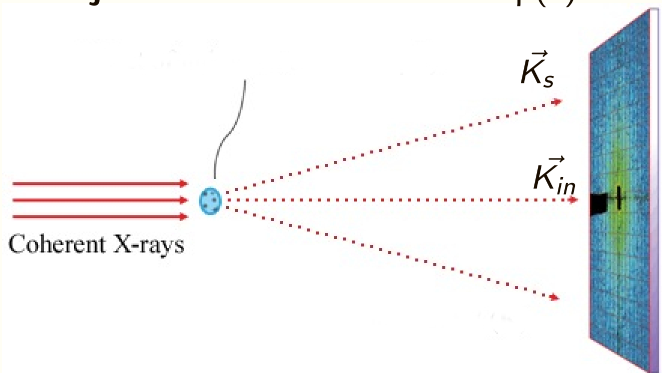
Speckles



$$\Delta\theta \approx \frac{\lambda}{a}$$

Coherent X-ray diffraction in the Fraunhofer regime

Object of unknown structure $\rho(\vec{r})$



Intensity, $I(\vec{q})$

Scattering vector

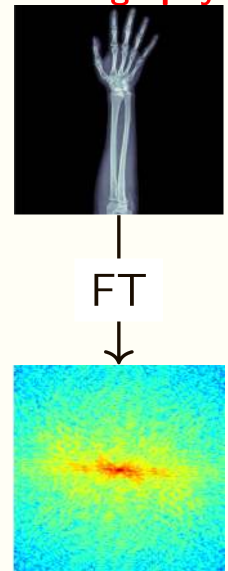
$$\vec{q} = \vec{k}_{in} - \vec{k}_s$$

Phase are lost

$$I(\vec{q}) = |E(\vec{q})|^2$$

$$E(\vec{q}) = |E(\vec{q})|e^{i\phi(\vec{q})}$$

Radiography



Intensity

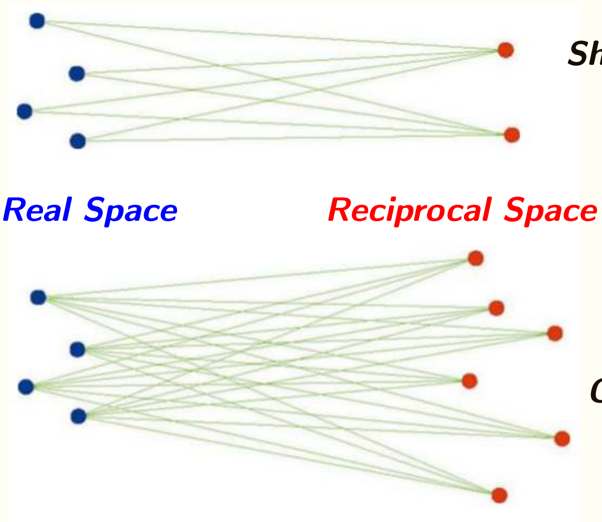
Images, yes... but, Fourier transformed ones

$$E(\vec{q}) = \underbrace{\int \rho(\vec{r})e^{-2\pi i\vec{r}\cdot\vec{q}}d\vec{r}}_{\text{Fourier Transform}} = \mathcal{F}\{\rho(\vec{r})\}$$

Patterson Function: $\mathcal{F}\{|E(\vec{q})|^2\} = (\rho \star \rho)(\vec{r}) = \int \rho(\vec{r}\prime + \vec{r})\rho(\vec{r}\prime)d\vec{r}\prime$

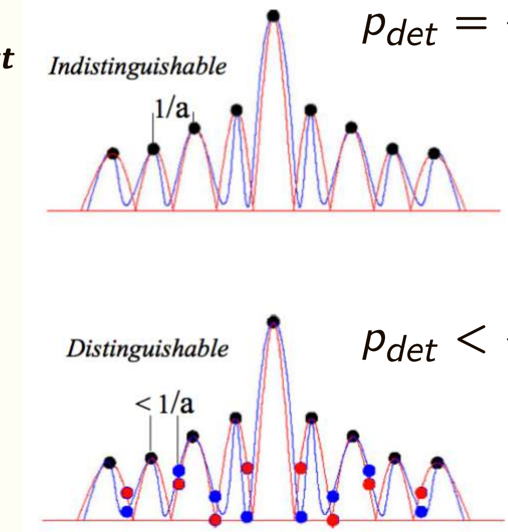
Phases are lost, but can be retrieved...

Oversampling of the intensities



Shannon-Nyquist sampling

Oversampling



$$p_{det} = \frac{\lambda z}{a}$$

$$p_{det} < \frac{\lambda z}{2a}$$

Sampling in the real space:

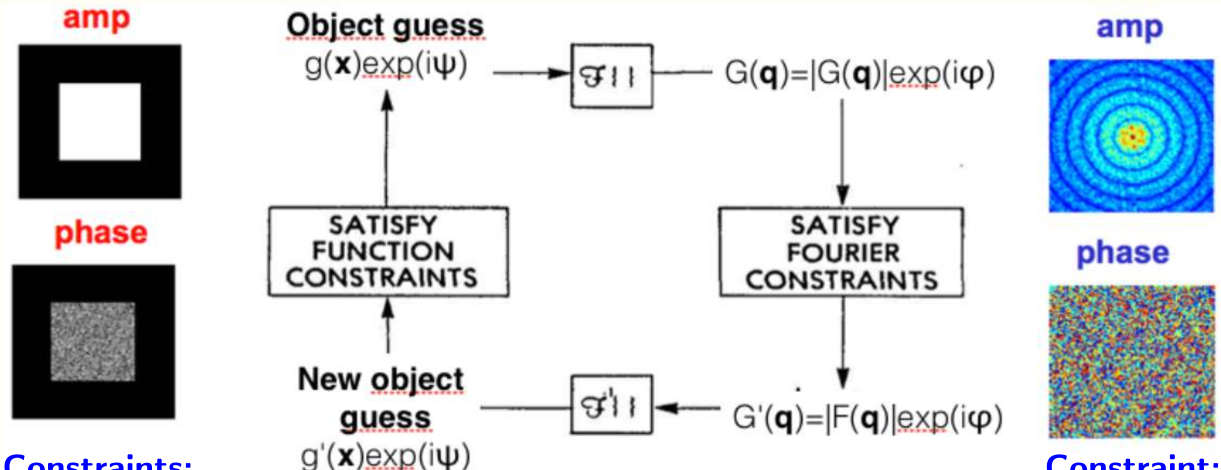
$$x \rightarrow \left[-\frac{L_x}{2} : \Delta x : \frac{L_x}{2} - \Delta x\right]$$

Sampling in the Fourier Space:

$$f_x \rightarrow \left[-\frac{1}{2\Delta x} : \frac{1}{L_x} : \frac{1}{2\Delta x} - \frac{1}{L_x}\right]$$

$$f_{Nyquist} = \frac{1}{2\Delta x}$$

J. Goodman, Fourier Optics (book)
D. Sayre, Acta Cryst. 5, 843 (1952)
J. Miao, D. Sayre, H. Chapman, JOSA A 15, 1662 (1998)



Constraints:
- Finite Support
- Positivity
- *a priori* knowledge

J.R. Fienup, J. Opt. Soc. Am. 3 27(1978).
J. R. Fienup, Appl. Opt. 21, 2758 (1982).
R. W. Gerchberg and W.O. Saxton, Optik 35, 237 (1972).

Constraint:

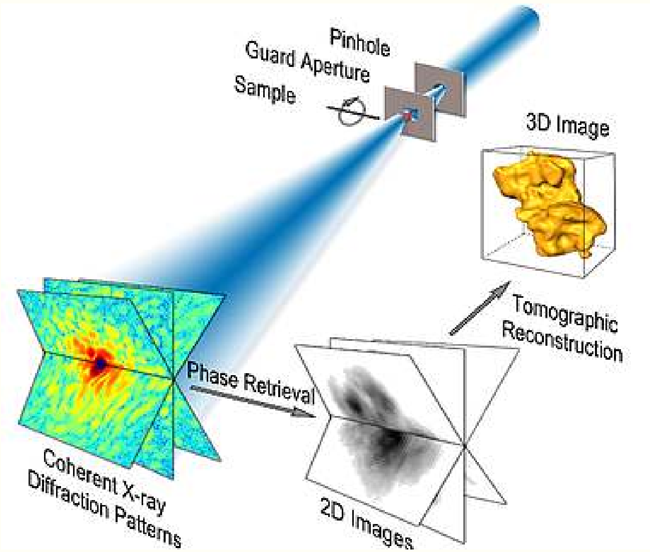
$$|G(\vec{q})|$$

$$|F(\vec{q})| = \sqrt{I_{exp}(\vec{q})}$$

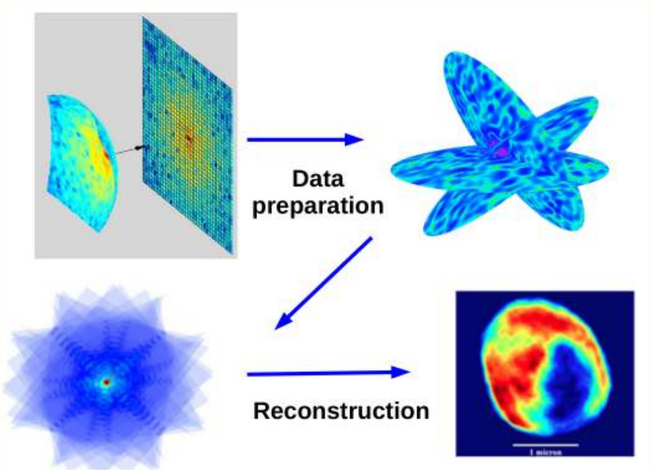
We cycle iteratively between Real and Reciprocal space through Fourier Transformations while imposing the constraints of each space until convergence is reached.

3D CDI using tomography or 3D Fourier transform

Tomo CDI

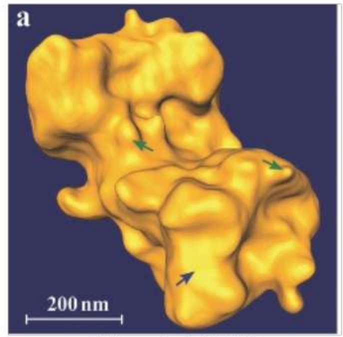


3D CDI

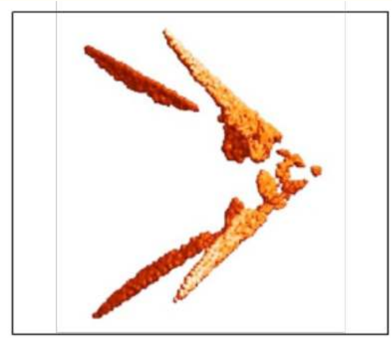


Some examples in the literature

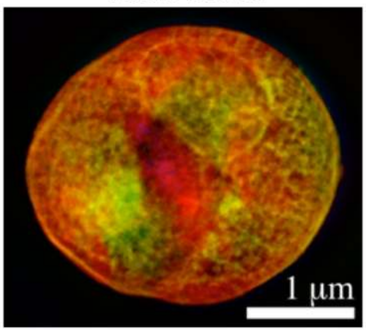
Miao *et al.* (1999)
Nature **400** 342.



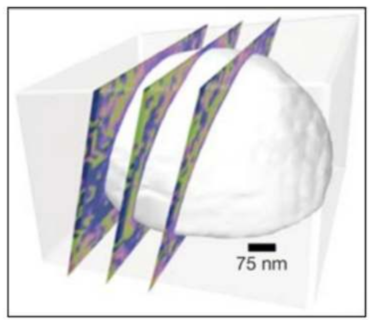
Miao *et al.* (2006)
Phys Rev Lett **97** 215503.



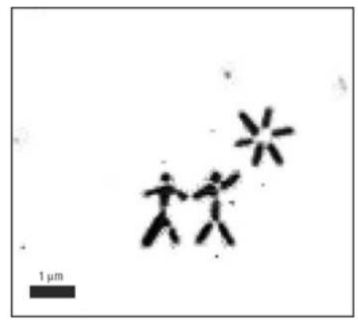
Chapman *et al.* (2006)
J. Opt. Soc. Am. A **23** 1179.



Shapiro *et al.* (2005)
Proc. Natl. Acad. Sci. **102** 15343.



Pfeifer *et al.* (2006)
Nature **442** 63.



Chapman *et al.* (2006)
Nature Physics **2** 839.

There are some difficulties

CDI poses some challenges for detector technology

- very high dynamic range
- small pixel size and very large active detection area
- noise-free detectors

Isolated particles

- specimens need to be isolated
- limited to small specimens
- tremendous oversampling of the intensities

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Algorithm stagnation problems

- slow convergence rate
- twin-images
- low-frequency information is hard to recover, but essential

Ptychography

Pty·chog·ra·phy *noun*

German: Falte

from Greek: $\pi\tau\upsilon\xi$ *= fold (crease)*

and: $\gamma\varrho\alpha\varphi\acute{\eta}$ *= writing, drawing*

Dynamische Theorie der Kristallstrukturanalyse durch Elektronenbeugung im inhomogenen Primärstrahlwellenfeld

Von R. Hegerl und W. Hoppe

Some time ago a new principle was proposed for the registration of the complete information (amplitudes and phases) in a diffraction diagram, which does not – as does Holography – require the interference of the scattered waves with a single reference wave. The basis of the principle lies in the interference of neighbouring scattered waves which result when the object function $\varrho(x, y)$ is multiplied by a generalized primary wave function $p(x, y)$ in Fourier space (diffraction diagram) this is a convolution of the Fourier transforms of these functions. The above mentioned interferences necessary for the phase determination can be obtained by suitable choice of the shape of $p(x, y)$. To distinguish it from holography this procedure is designated "ptychography" ($\pi\tau\upsilon\xi$ = fold). The procedure is applicable to periodic and aperiodic structures. The relationships are simplest for plane lattices. In this paper the theory is extended to space lattices both with and without consideration of the dynamic theory. The resulting effects are demonstrated using a practical example.

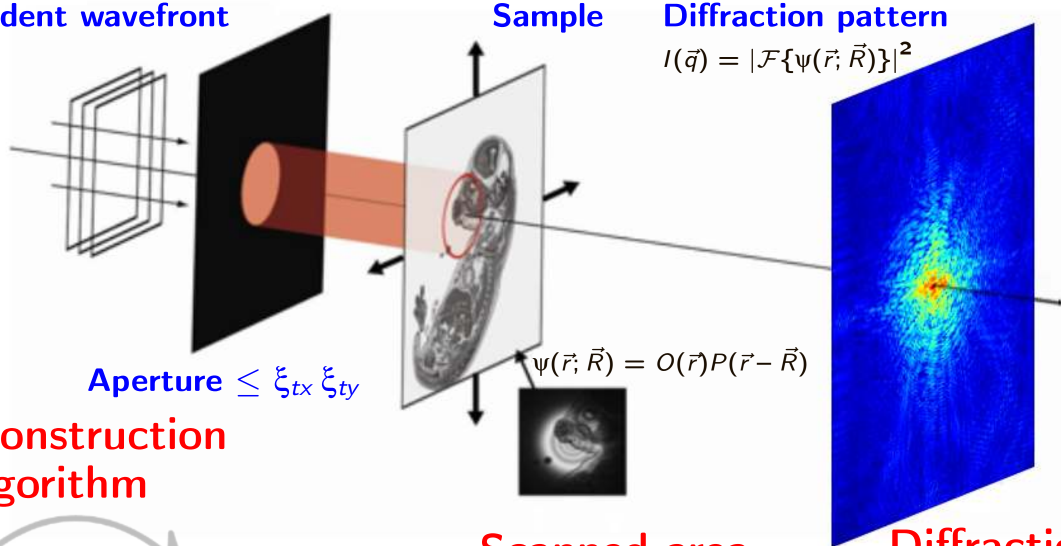
Hoppe, Acta Cryst A 25 (1969) 508; Hegerl and Hoppe, Ber Physik Chemie 74 (1970) 1148

Transverse diversity - Far-field X-ray Ptychography

exploiting the transverse diversity

Incident wavefront **Sample** **Diffraction pattern**

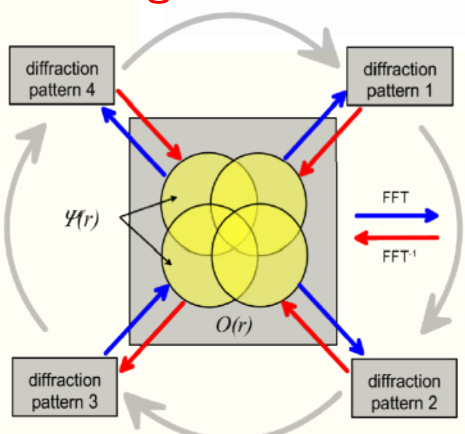
$I(\vec{q}) = |\mathcal{F}\{\psi(\vec{r};\vec{R})\}|^2$



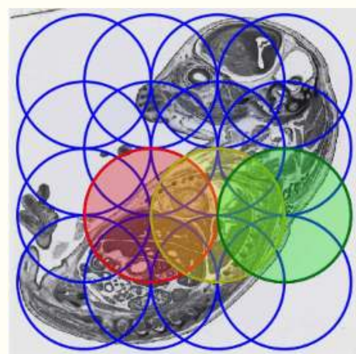
$\psi(\vec{r};\vec{R}) = O(\vec{r})P(\vec{r}-\vec{R})$

Aperture $\leq \xi_{tx}\,\xi_{ty}$

Reconstruction algorithm



Scanned area

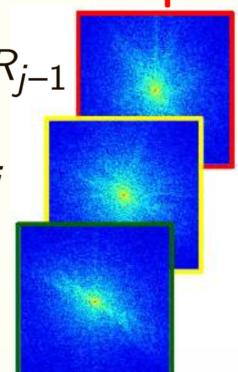


Diffraction patterns

R_{j-1}

R_j

R_{j+1}



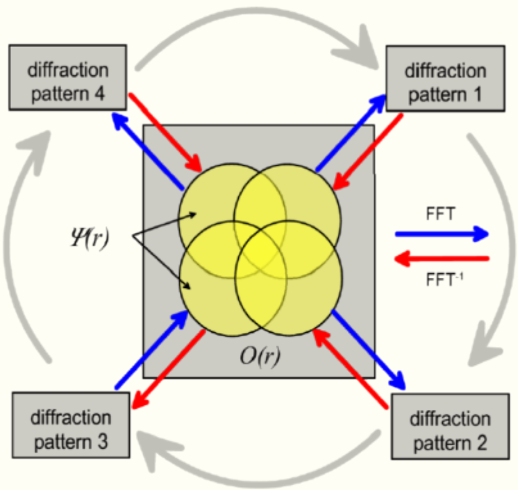
J. C. da Silva, A. Menzel, Opt. Express 23 (2015) 33812. / J.M. Rodenburg et al., Phys. Rev. Lett. 98 (2007) 034801

X-ray ptychography - the reconstruction problem

Find the object $O(r)$ and the complex-valued incident illumination $P(r)$ (the probe) consistent with the measured intensities:

$$\mathrm{I}(\vec{q};\vec{R}) = \left| \int_{-\infty}^{\infty} \mathrm{O}(\vec{r})\mathrm{P}(\vec{r}-\vec{R})e^{-i\vec{q}\cdot\vec{r}}d\vec{r} \right|^2$$

Iterative phase retrieval



Fourier constraints

Each "view" satisfies its own Fourier Constraint.

Overlap constraints

Overlapping regions agree and the incident wave field is unique.

Redundancy

It allows to simultaneously reconstruct the probe and the object.

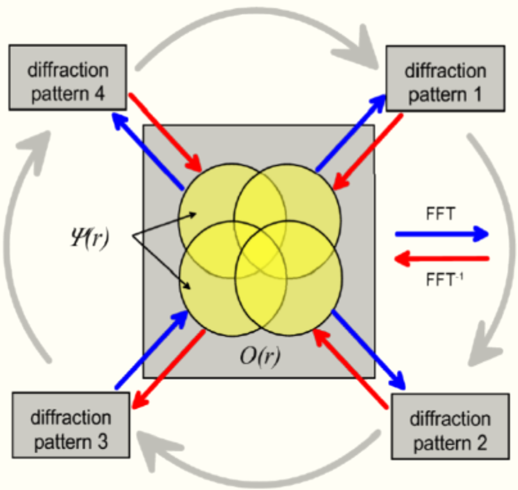
M. Dierolf et al., Europhysics New 39 (2008) 22.
P. Thibault et al., Ultramicroscopy 4 (2009), 338.
H.M.L. Faulkner, J.M. Rodenburg, Phys. Rev. Lett. 93 (2004), 023903
J. C. da Silva and A. Menzel, Opt. Express 23 (2015) 33812.

X-ray ptychography - the reconstruction problem

Find the object $O(r)$ and the complex-valued incident illumination $P(r)$ (the probe) consistent with the measured intensities:

$$\mathrm{I}(\vec{q};\vec{R}) = \left| \int_{-\infty}^{\infty} \mathrm{O}(\vec{r})\mathrm{P}(\vec{r}-\vec{R})e^{-i\vec{q}\cdot\vec{r}}d\vec{r} \right|^2$$

Iterative phase retrieval



Fourier constraints

Each "view" satisfies its own Fourier Constraint.

Overlap constraints

Overlapping regions agree and the incident wave field is unique.

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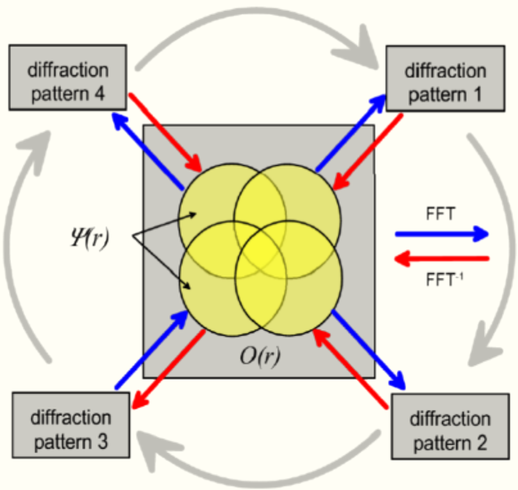

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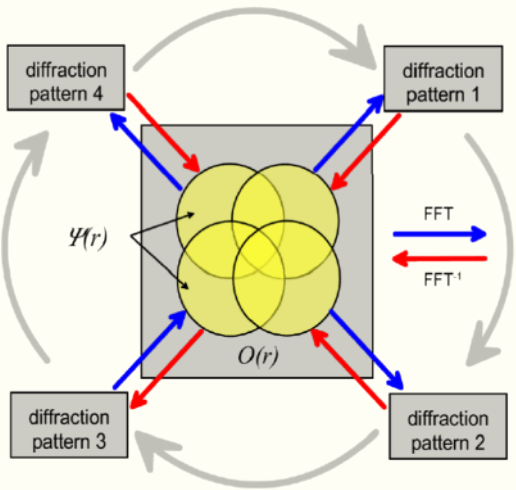

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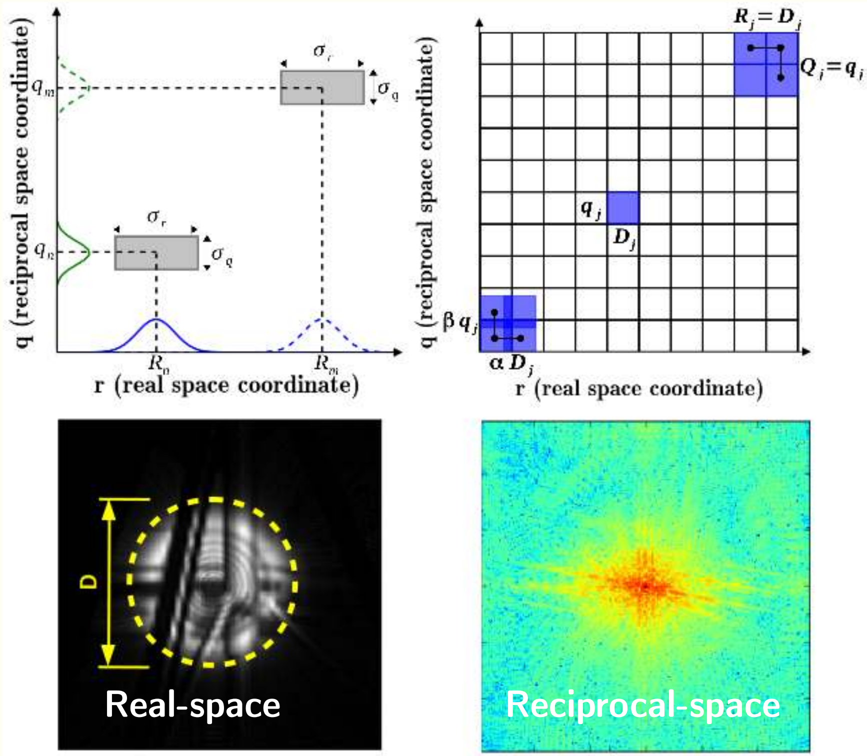
H.M.L. Faulkner, J.M. Rodenburg, Phys. Rev. Lett. 93 (2004), 023903
J. C. da Silva and A. Menzel, Opt. Express 23 (2015) 33812.

Sampling requirements for Ptychography



$\psi(\vec{r};\vec{R}) = O(\vec{r})P(\vec{r}-\vec{R})$ $\quad$ $I(\vec{q}) = |\mathcal{F}\{\psi(\vec{r};\vec{R})\}|^2$

J. C. da Silva, A. Menzel, Opt. Express 23 (2015) 33812.

Let us define:

$$R_j = \alpha D_j$$

$$Q_j = \beta q_j$$

where: $\beta = \dfrac{1}{\mathcal{O}}$

Density of boxes:

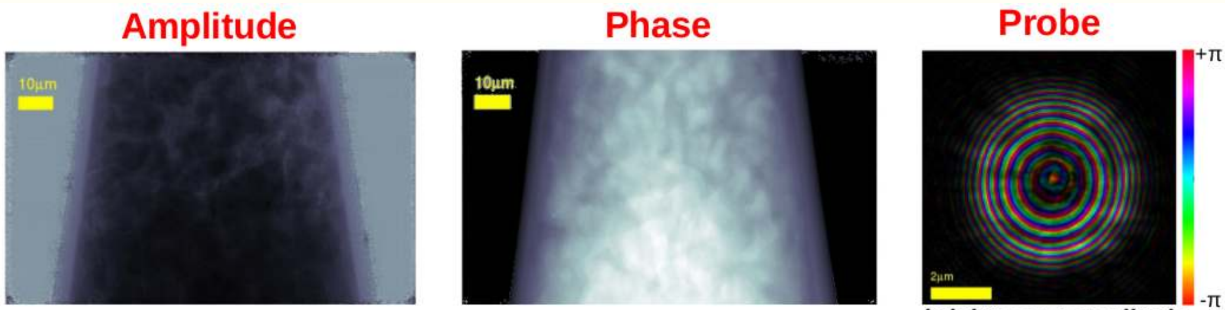
$$\frac{2\pi}{(\alpha D_j)(\beta q_j)} \geq 1$$

Since $D_j q_j = 2\pi$:

$$\boxed{\frac{1}{\alpha\beta} > 1}$$

Oversampling condition

Welcome to the third dimension



brightness : amplitude
color : phase

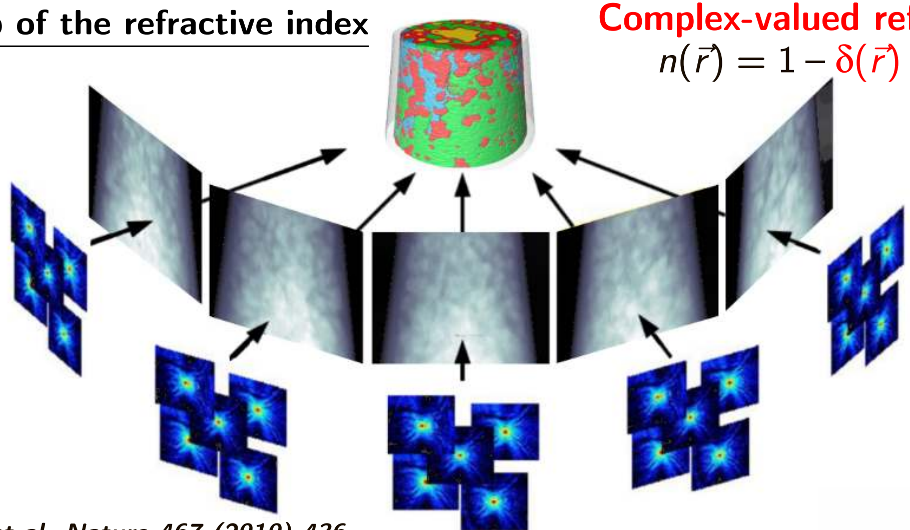
J. C. da Silva et al. Langmuir 31 (2015) 3779

Ptychographic X-ray Computed Tomography (PXCT)

3D map of the refractive index

Complex-valued refractive index

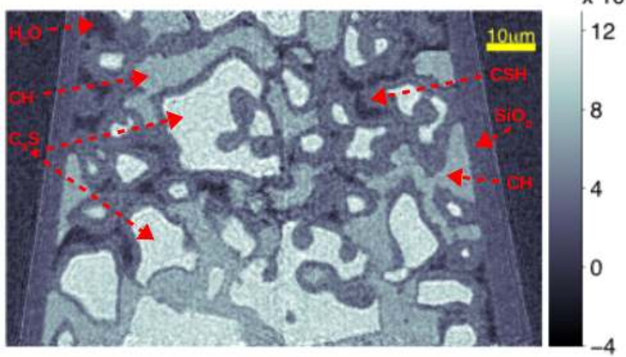
$$n(\vec{r}) = 1 - \delta(\vec{r}) + i\beta(\vec{r})$$



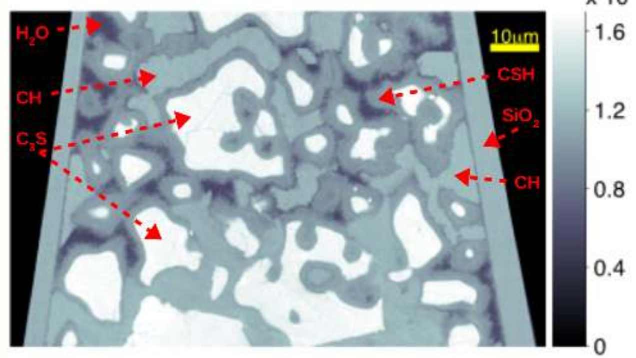
M. Dierolf et al. Nature 467 (2010) 436

$$\underbrace{Ca_3SiO_5}_{\text{Alite } C_3S} + \underbrace{(1.3+\textcolor{red}{y})H_2O}_{\text{water}} \rightarrow \underbrace{1.3\,Ca(OH)_2}_{\text{Calcium hydroxide } CH} + \underbrace{(CaO)_{1.7}(SiO_2)(H_2O)_{\textcolor{red}{y}}}_{\text{Calcium-Silicate-Hydrated } CSH}$$

Absorption index β



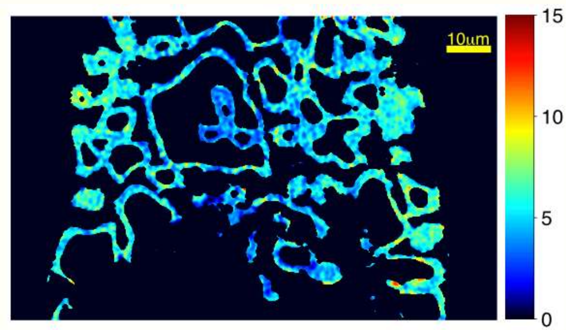
Refractive index decrement δ



$$\left(\frac{\mu}{\rho}\right)_{CSH} = \omega_{CaO}\left(\frac{\mu}{\rho}\right)_{CaO} + \omega_{SiO_2}\left(\frac{\mu}{\rho}\right)_{SiO_2} + \omega_{H_2O}\left(\frac{\mu}{\rho}\right)_{H_2O}$$

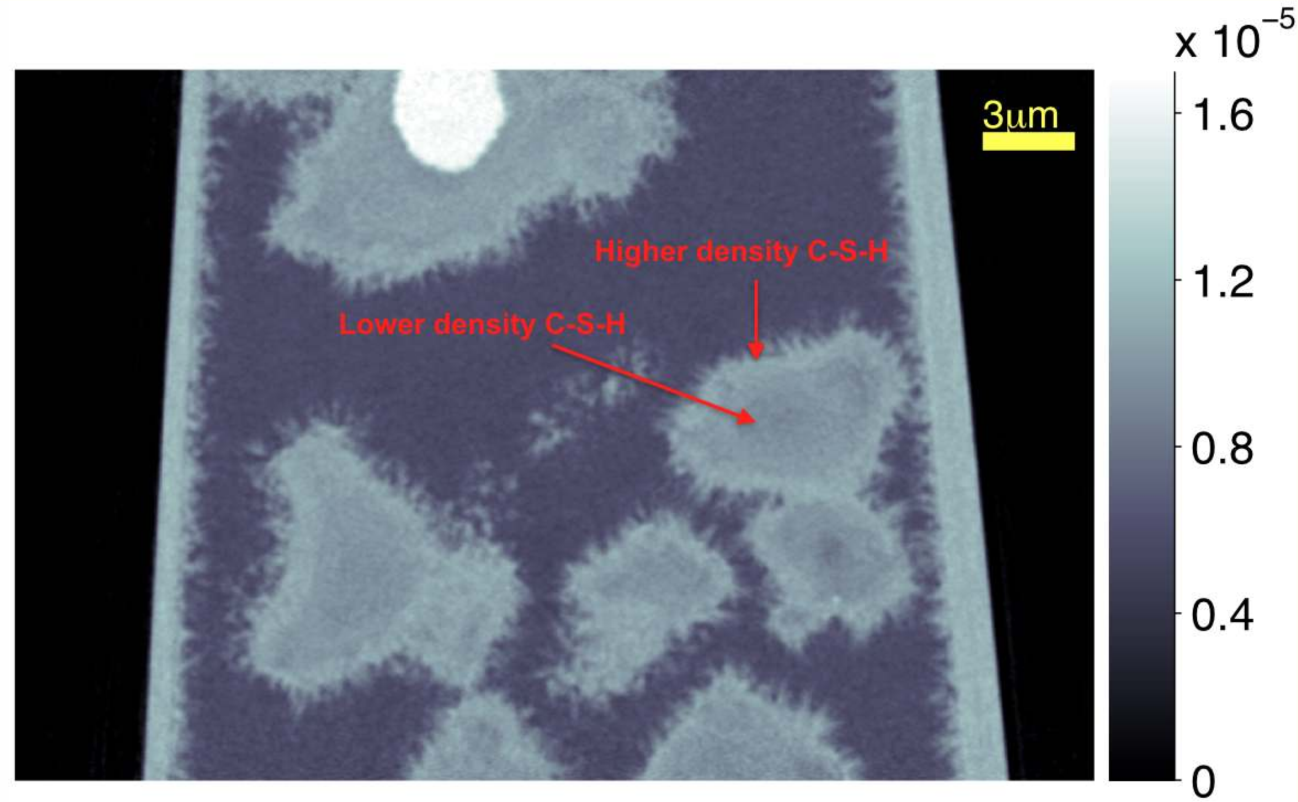
Materials	ρ $(g.cm^{-3})$	$\rho_{ex.}$ $(g.cm^{-3})$
Water	0.99 ± 0.01	1.00
CH	2.18 ± 0.01	2.211
C_3S	3.10 ± 0.01	3.064
Capillary	2.19 ± 0.01	2.203
$CSH_{(\textcolor{red}{y=5.2\pm0.4})}$	$\mathbf{1.83 \pm 0.01}$	$\mathbf{1.83\ (y=5)}$

Water content of CSH:



J. C. da Silva et al., Langmuir 31 (2015) 3779.

Unexpected high density C-S-H at the border



J. C. da Silva et al., Langmuir 31, (2015), 3779.

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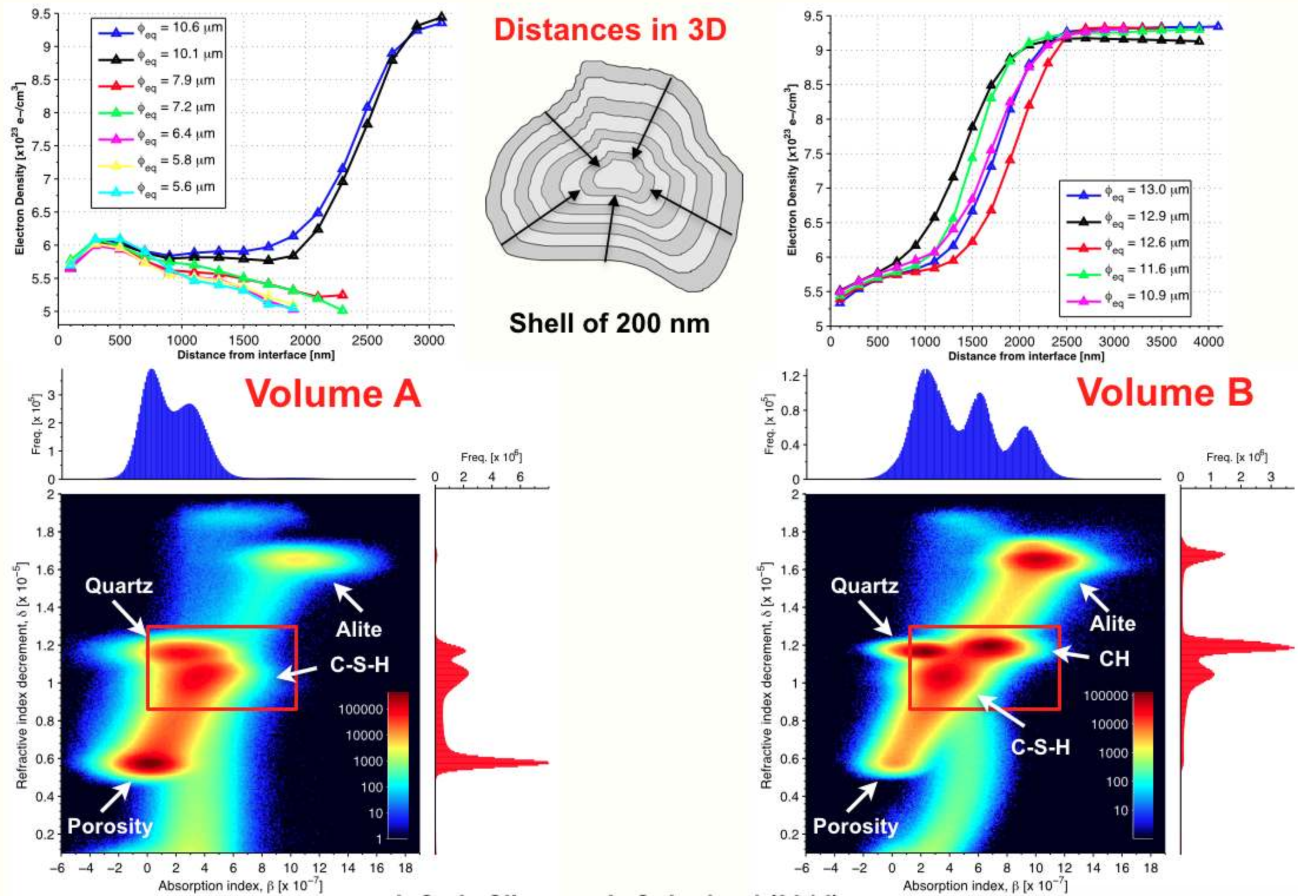
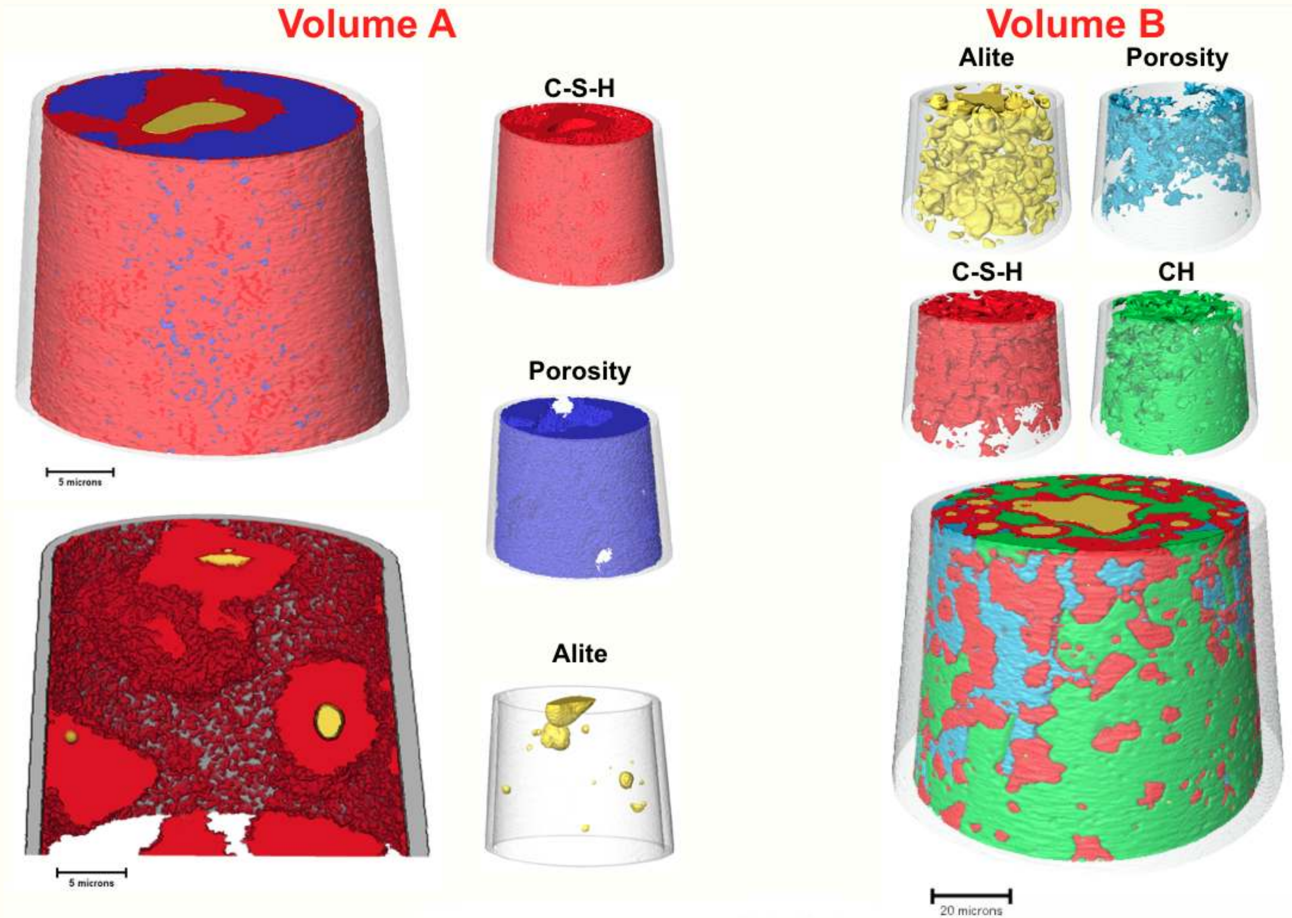


J. C. da Silva et al., Langmuir 31, (2015), 3779.

How the phases are seated to each other



J. C. da Silva et al., Langmuir 31, (2015), 3779.

Eco-friendly cement: the hydration of ye'elemite

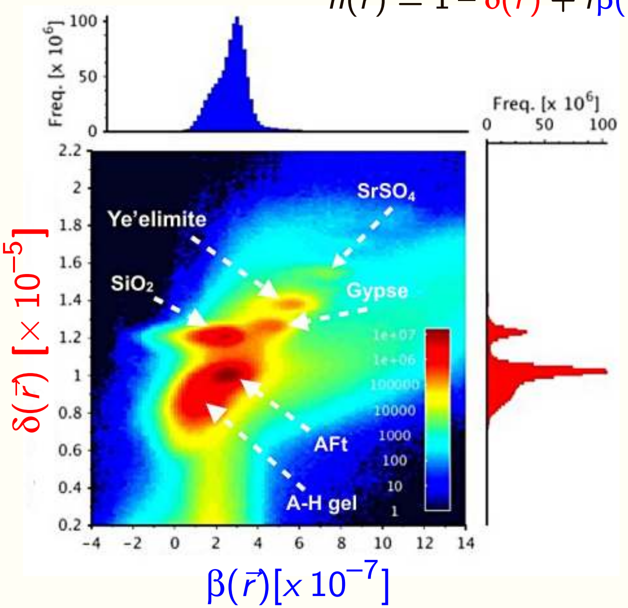
$$\underbrace{Ca_4Al_6SO_{16}}_{\text{ye'elemite}} + \underbrace{2CaSO_4.nH_2O}_{\text{gypsum}} + \underbrace{(38-2n)H_2O}_{\text{water}} \rightarrow \underbrace{Ca_6Al_2(OH)_{12}(SO_4)_3.26H_2O}_{\text{ettringite (AFt)}} + \underbrace{4(CaO)_{0,04}Al(OH)_3.nH_2O}_{\text{Al-hydroxyde (A-H gel)}}$$

Identify:

Bivariate histogram: Phase vs. Absorption

$\delta(\vec{r})$ $\beta(\vec{r})$

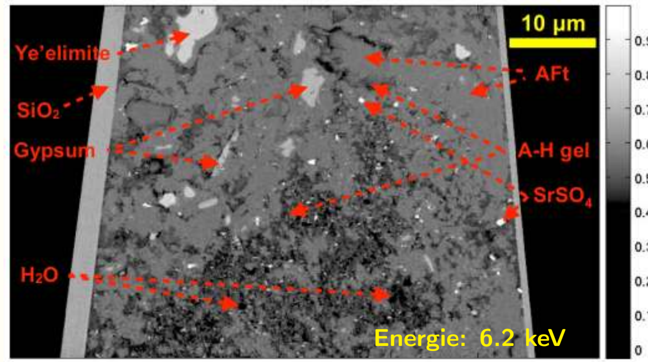
$$n(\vec{r}) = 1 - \delta(\vec{r}) + i\beta(\vec{r})$$



A. Cuesta et al., J. Phys. Chem. C 121, 3044 (2017)

Characterize:

3D map of the electron density ($e^-/Å^3$)



Quantify:

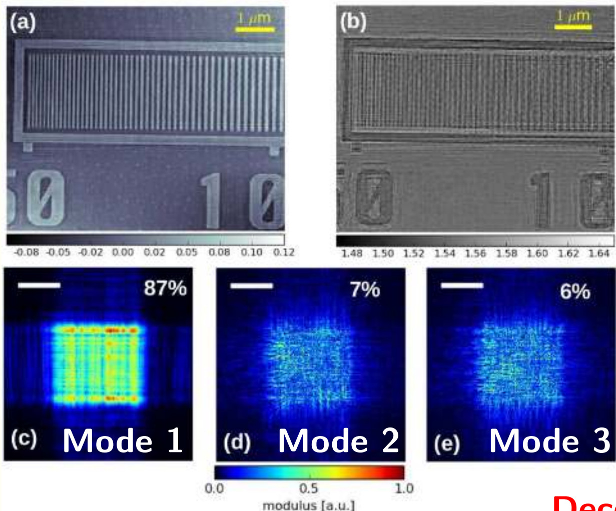
Mass density and stoichiometry

Material	ρ $(g.cm^{-3})$	$\rho_{ex}(g.cm^{-3})$
ye'elemite	2.58	2.60
gypsum	2.28	2.30
ettringite (AFt)	1.77	1.78
A-H gel (**n=2.3**)	**1.48**	**N.A.**

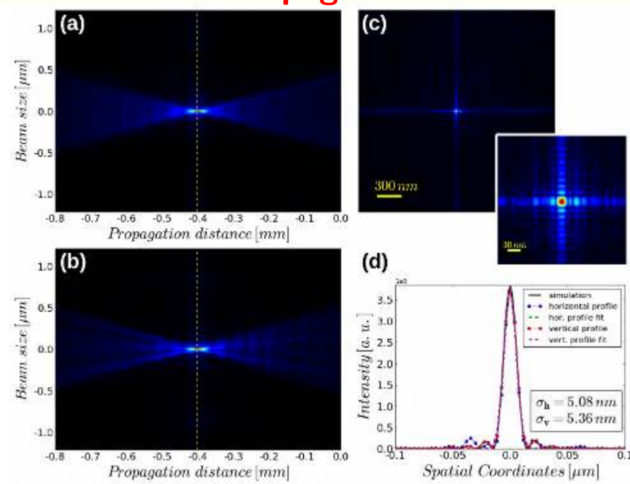
Jupyter Notebook

Smallest ever high energy (33.6 keV) focal spot

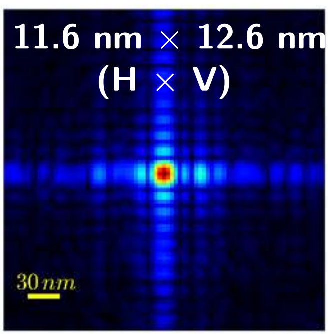
Phase **Amplitude**



Wave Propagation

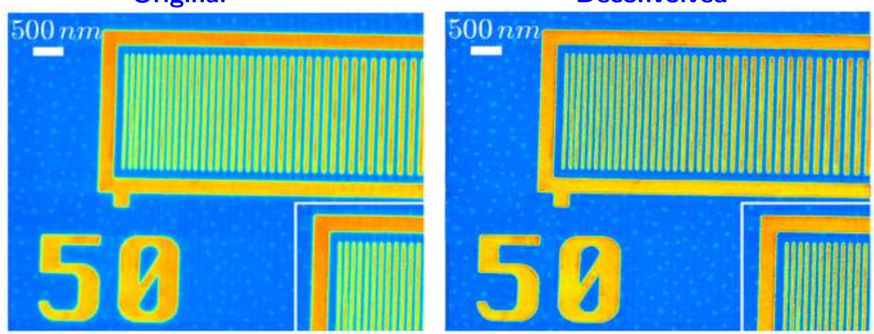


Focal spot



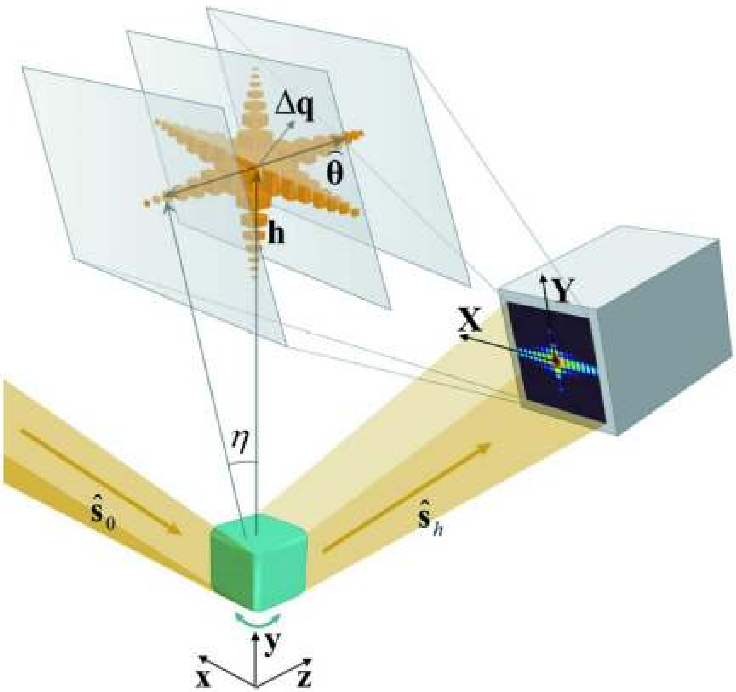
Deconvolution of XRF data

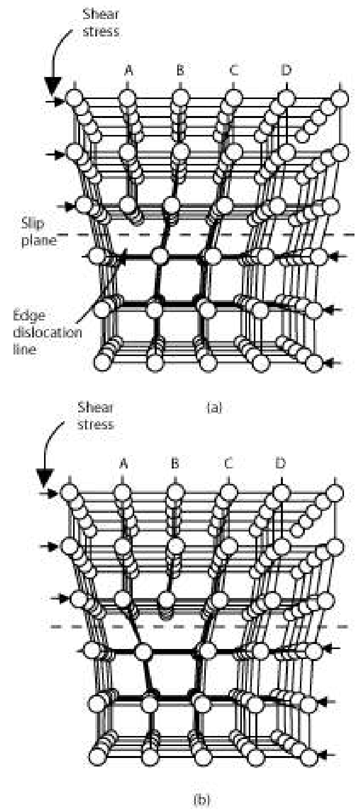
Original Deconvolved



J. C. da Silva et al., Optica 4(5), 492-495 (2017).

Bragg coherent diffraction imaging (Bragg CDI)



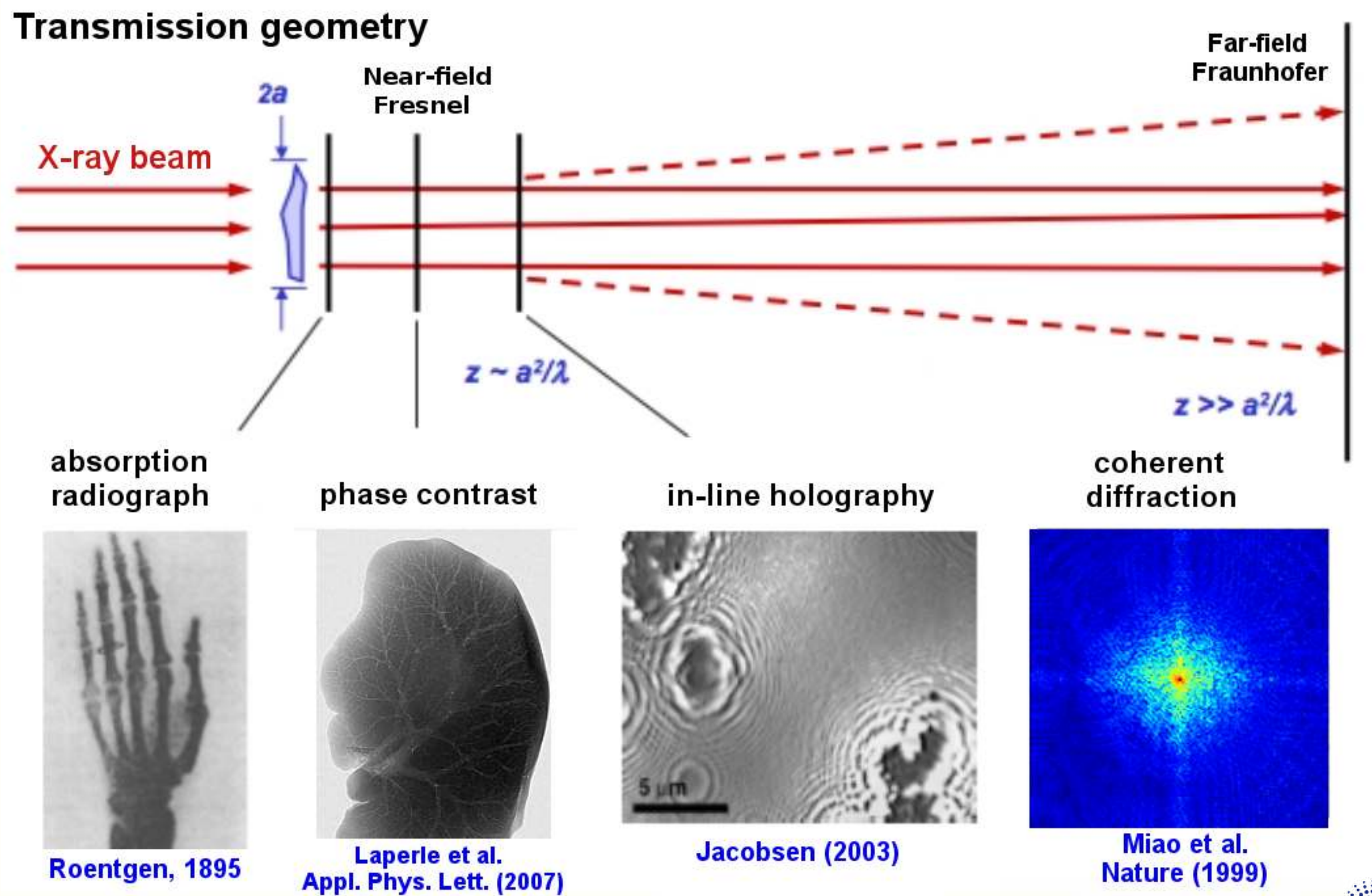


M. A. Pfeifer, et al., Nature 442, 63 (2006)

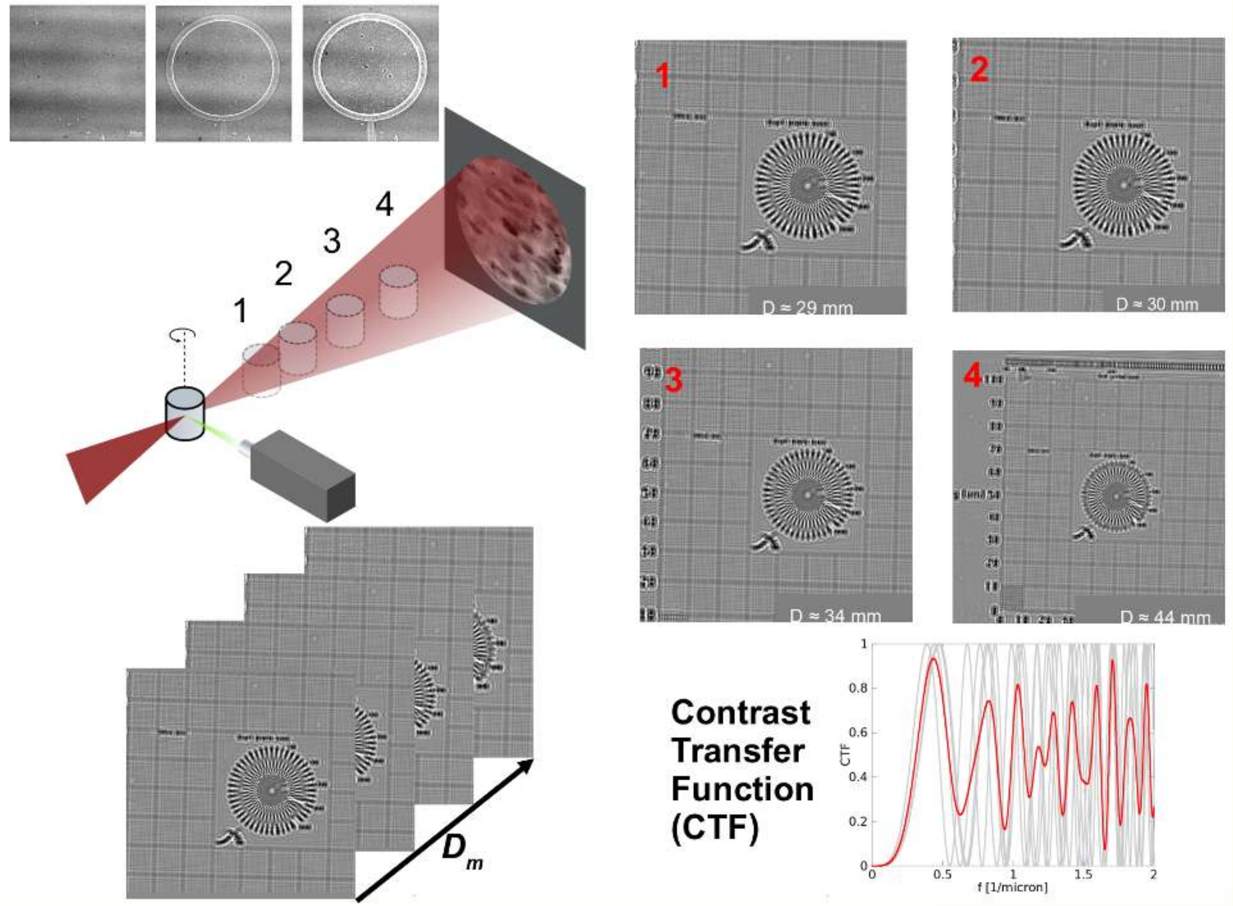
P. Godard, et al., Nature Communications 2, 568 (2011)

S. O. Hruszkewycz, et al., Nature Materials 16, 244-251 (2017)

Different X-ray imaging regimes

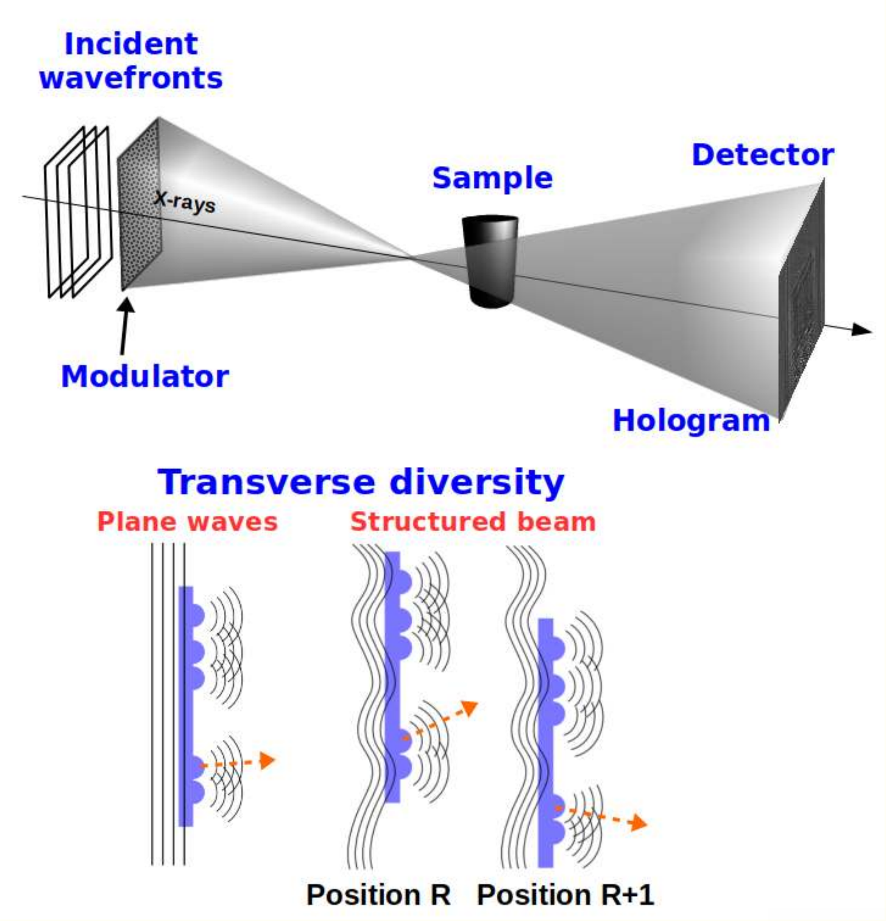


In-line Holography



P. Cloetens et al., Appl. Phys. Lett. 75, (1999), 2912.

Near-field ptychography

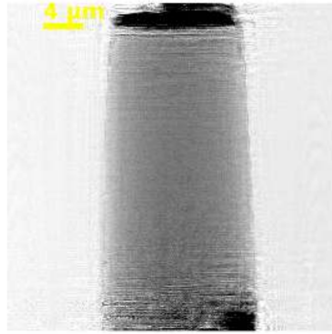


M. Stockmar et al., Sci. Rep. 3, 1927 (2013)

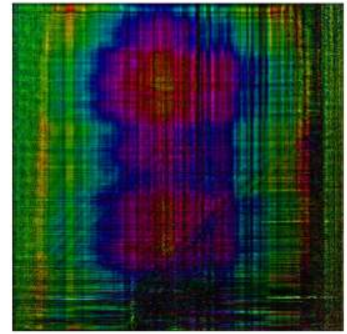
Welcome to the third dimension

Phase

Amplitude



Probe



Ptychographic X-ray Computed Tomography (PXCT)

Complex-valued refractive index

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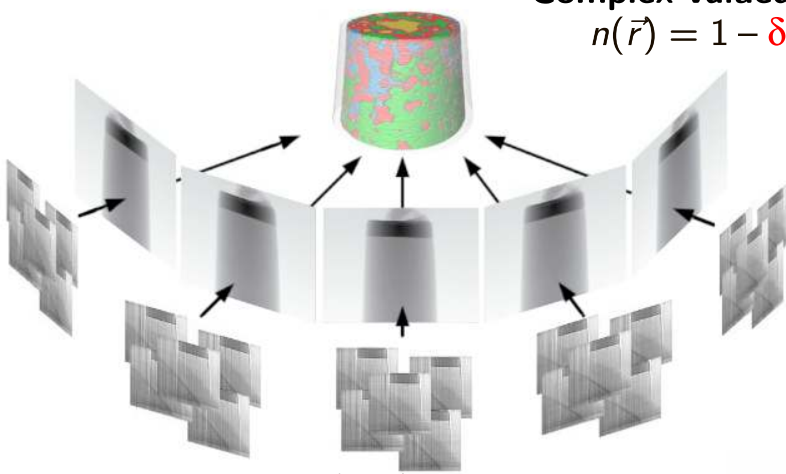
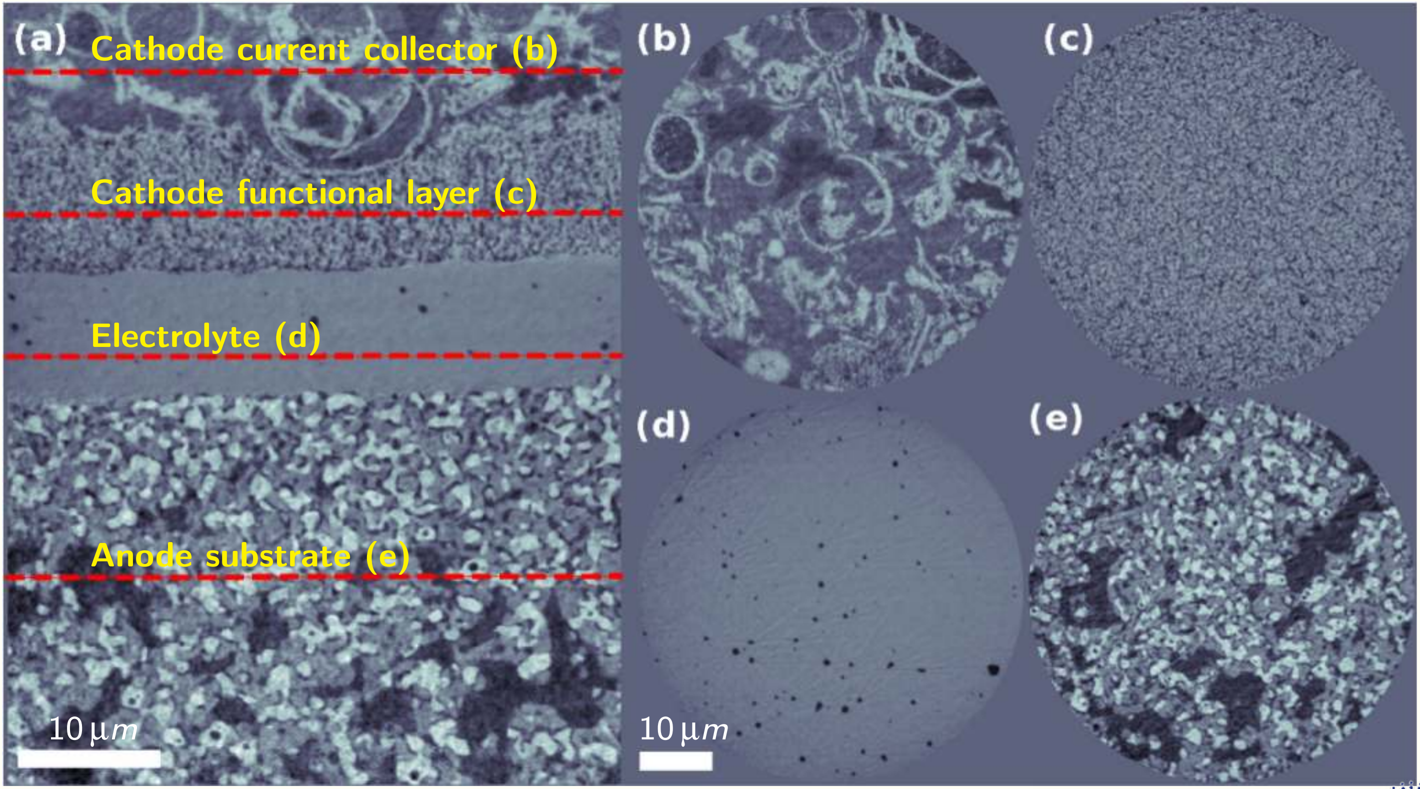


M. Stockmar et al. Opt. Express 23, 12720 (2015)

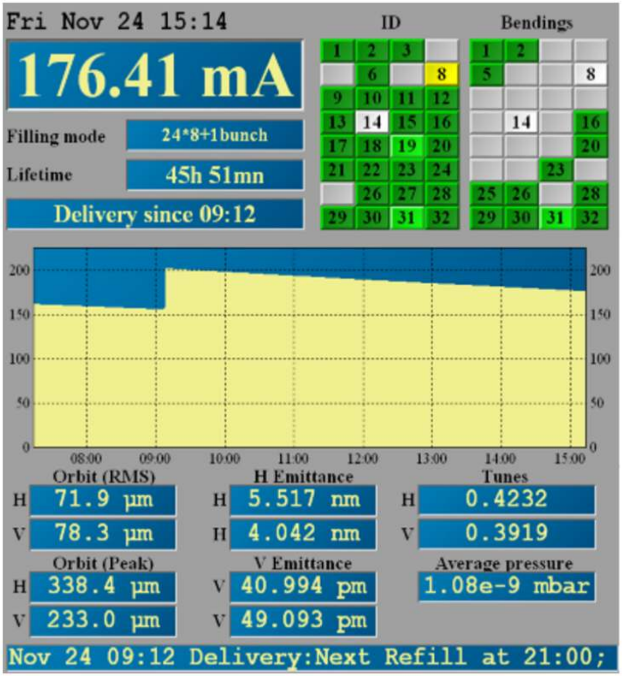
The different structures of a SOFC

SOFC: Solid Oxide Fuel Cell

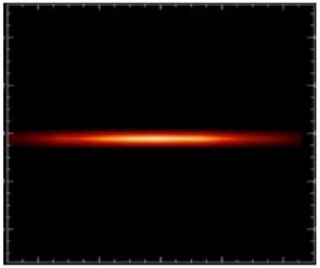


M. Stockmar et al., Opt. Express 23(10), 12720 (2015)

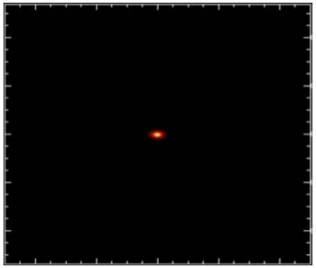
ESRF - Extremely Brilliant Source (EBS)

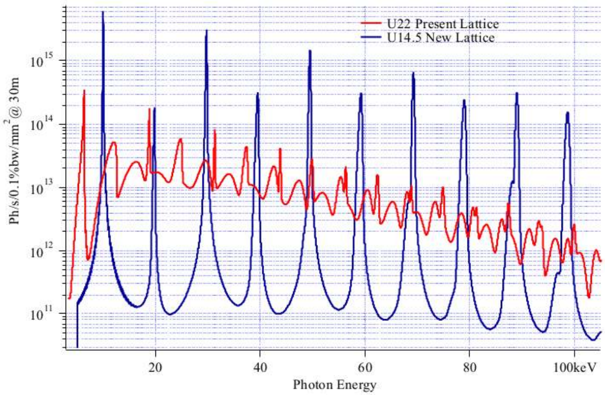


Current source

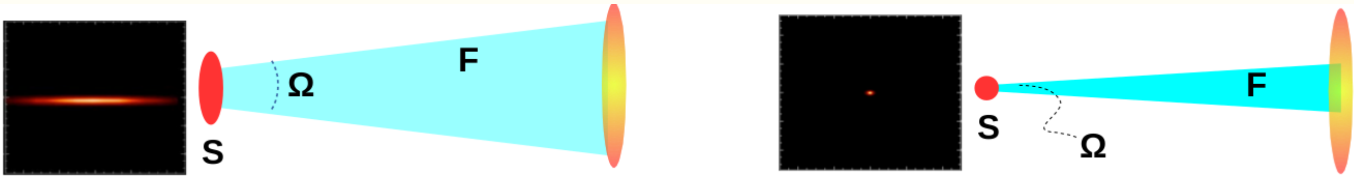


New source (Expected)





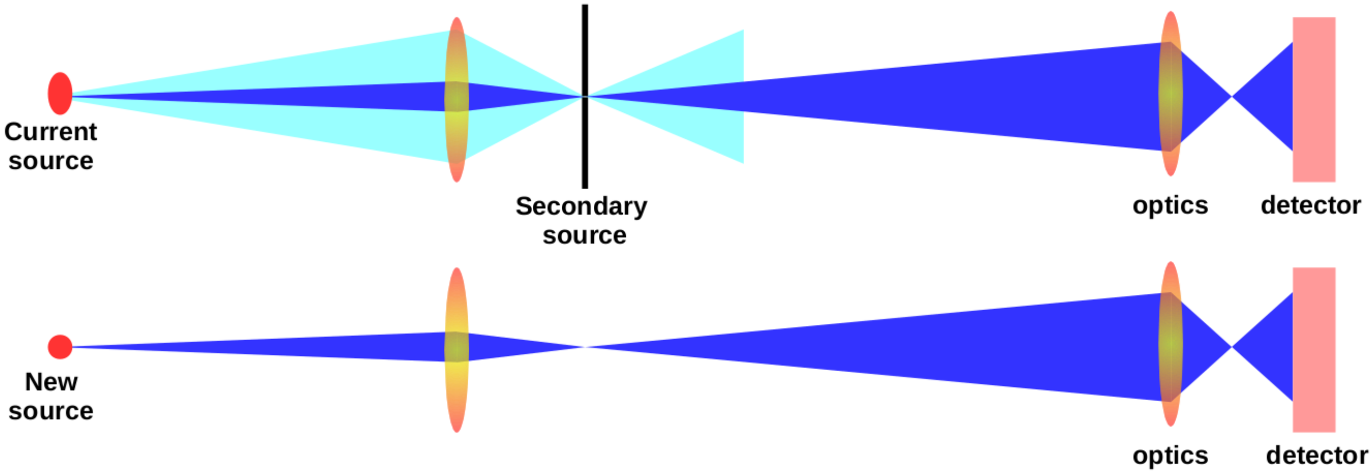
More brilliance and transverse coherence



Ω = angular divergence
S = source area
F = Flux

$$\text{Brilliance} = (\text{const.}) \times \frac{F}{S\Omega}$$

$$\text{Emittance} = S\Omega$$



Conclusions and remarks

Coherent X-ray imaging (CDI)

➔ **High resolution**, small samples, isolated specimens, works with perfect plane waves.

Far-field ptychography (FFP)

➔ **High resolution**, high penetration, combination with spectroscopy, tomography, works with plane waves or structured wavefield.

In-line holography

➔ **Large field-of-view**, high resolution, high penetration, tomography, faster than NFP, works with perfect plane waves.

Near-field ptychography (NFP)

➔ **Large field-of-view**, relatively high resolution, high penetration, tomography, faster than FFP, works with structured wavefields.

Thank you for your attention!

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