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1st AOFSRR Synchrotron School

Coherent X-ray Imaging

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XFEL and Materials Characterisation Group

Basic Revision of FT

- Recall: any signal may be represented as the sum of sinusoids. Thus all information about the signal is encoded in the values of the spatial frequency, the amplitude and the phase.

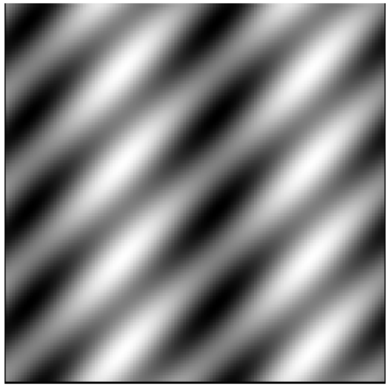
2D FT:

$$F(u_x, u_y) = \iint f(x, y) e^{+i(u_x x + u_y y)} dxdy$$

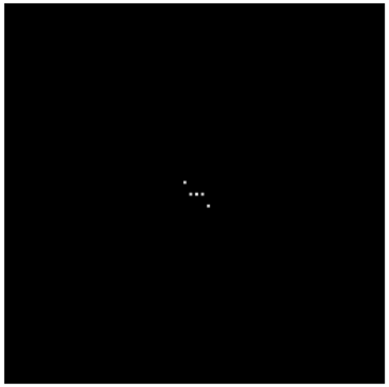
$$f(x, y) = \frac{1}{(2\pi)^2} \iint F(u_x, u_y) e^{-i(u_x x + u_y y)} du_x du_y$$

where: u_x and u_y are the angular spatial frequencies.

Brightness Image

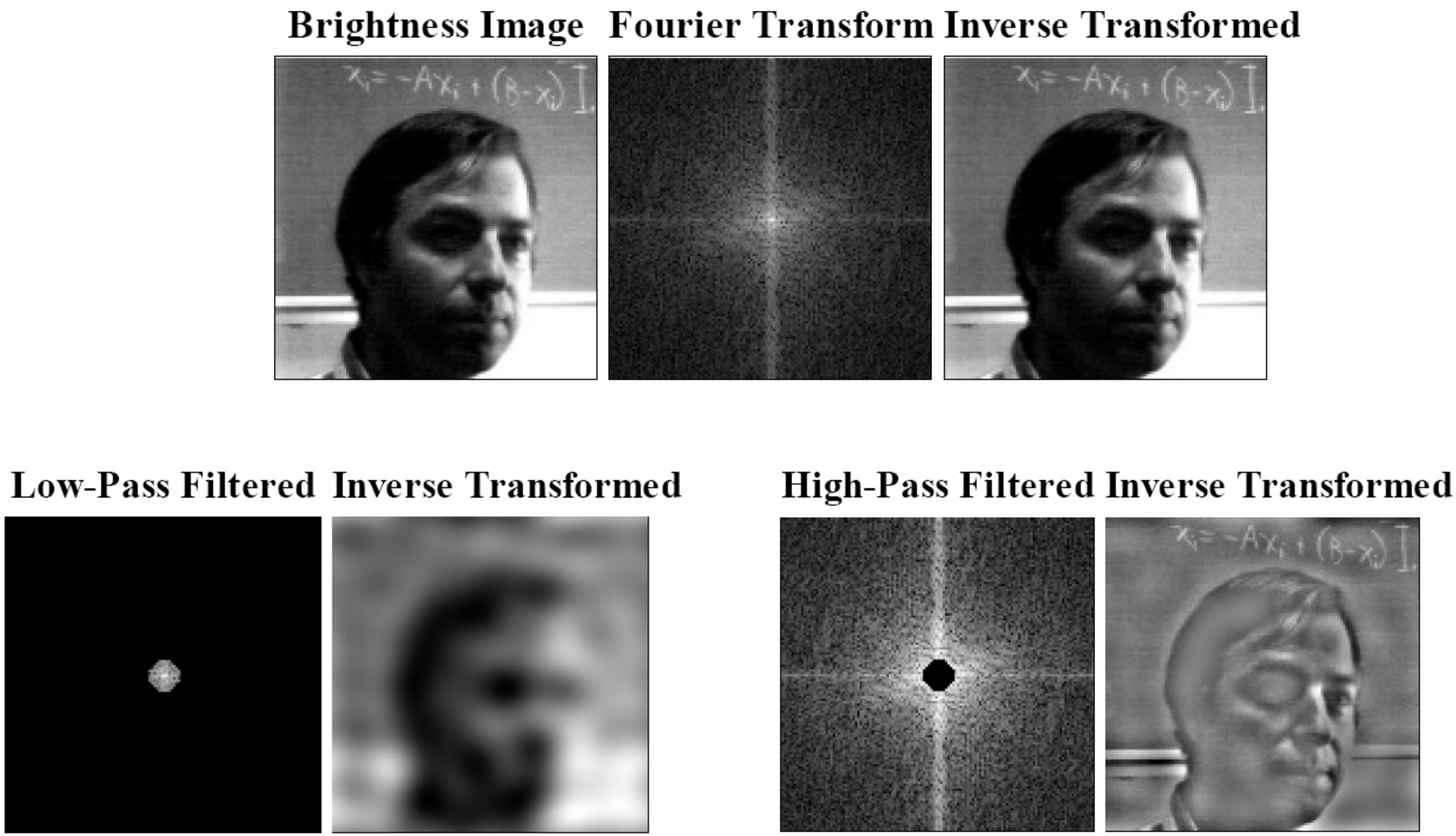


Fourier transform



Lowest spatial frequency represented in the FT (known as the DC term) corresponds to the average brightness across the whole image.
The highest spatial frequency encoded in the FT is known as the Nyquist Frequency

Basic Revision of FT



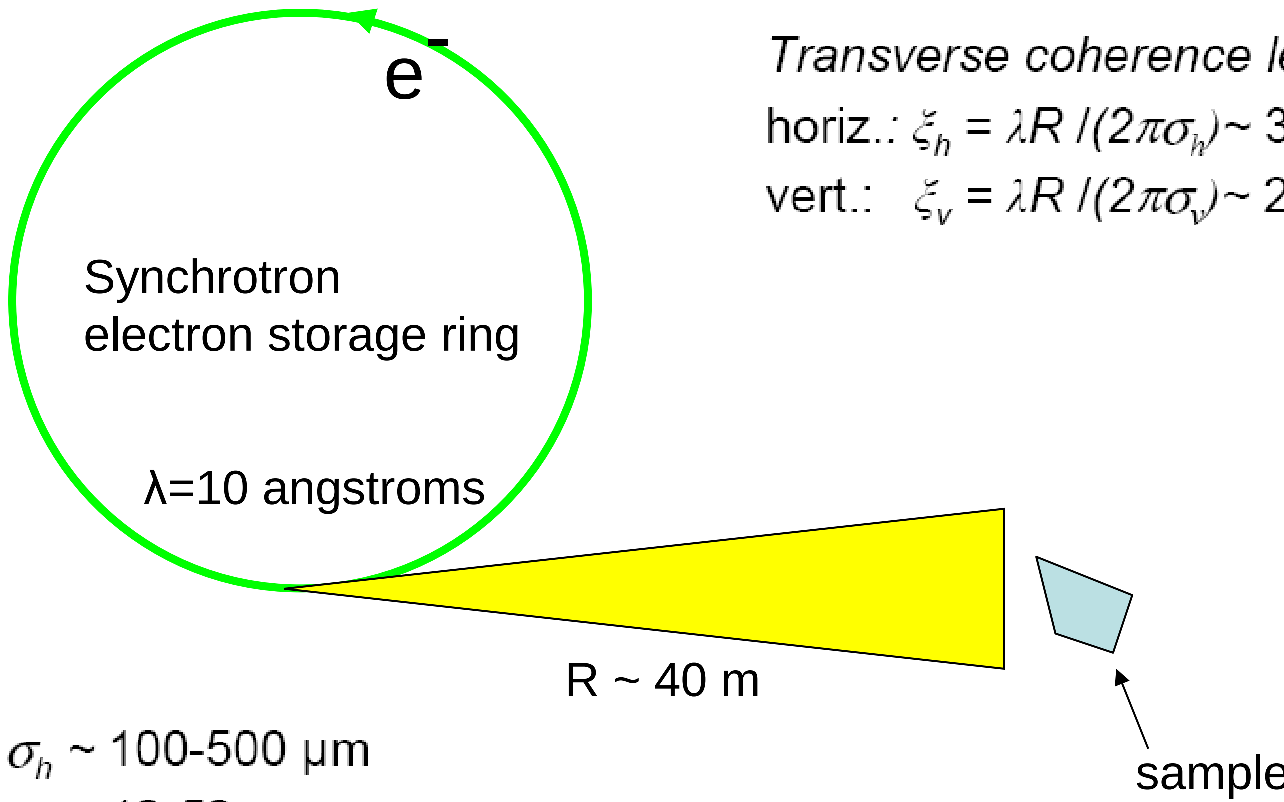
Coherent techniques in X-ray imaging

Gaussian source: $w = 2\pi\sigma$

Transverse coherence length:

horiz.: $\xi_h = \lambda R / (2\pi\sigma_h)$ ~ 3-10 μm

vert.: $\xi_v = \lambda R / (2\pi\sigma_v)$ ~ 25-100 μm



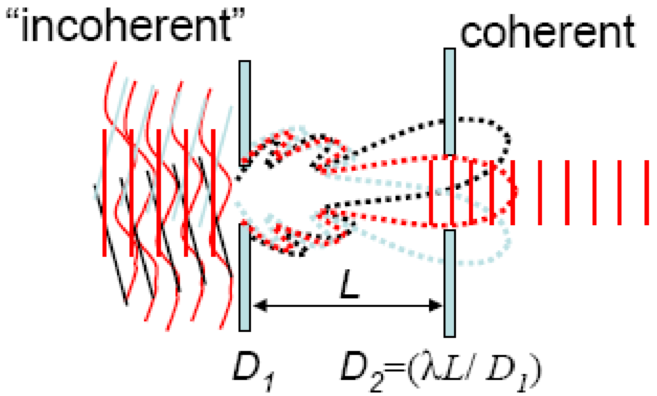
horiz: σ_h ~ 100-500 μm

vert.: σ_v ~ 10-50 μm

The longitudinal coherence at the synchrotron is generally good enough not to destroy the coherence (ΔE/E for a monochromator is typically ~ 10^{-4}).

Coherent techniques in X-ray imaging

- To produce a coherent illumination we can select out a small part of an incoherent beam:



But coherence is not necessary for all X-ray imaging techniques:

Incoherent techniques: Full field X-ray microscopy.

Coherent techniques: Fluctuation microsopy, Holography and diffraction methods.

The Complex Refractive Index

In fact for any material there exist several resonant frequencies, these are the characteristic frequencies at which the atom absorbs and re-emits energy.

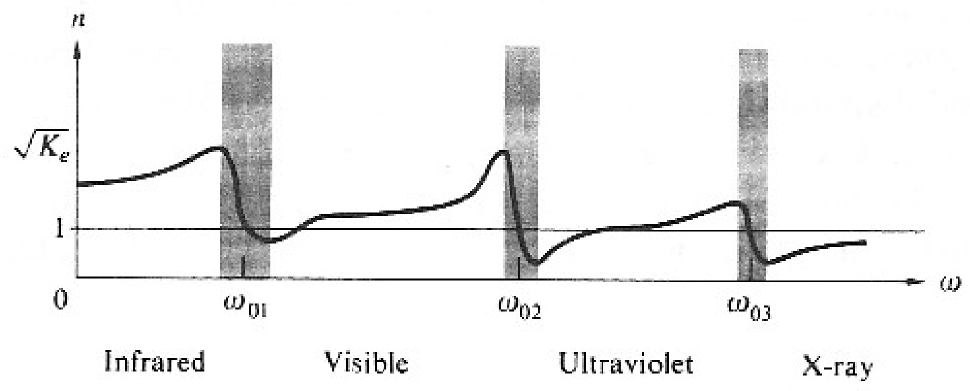


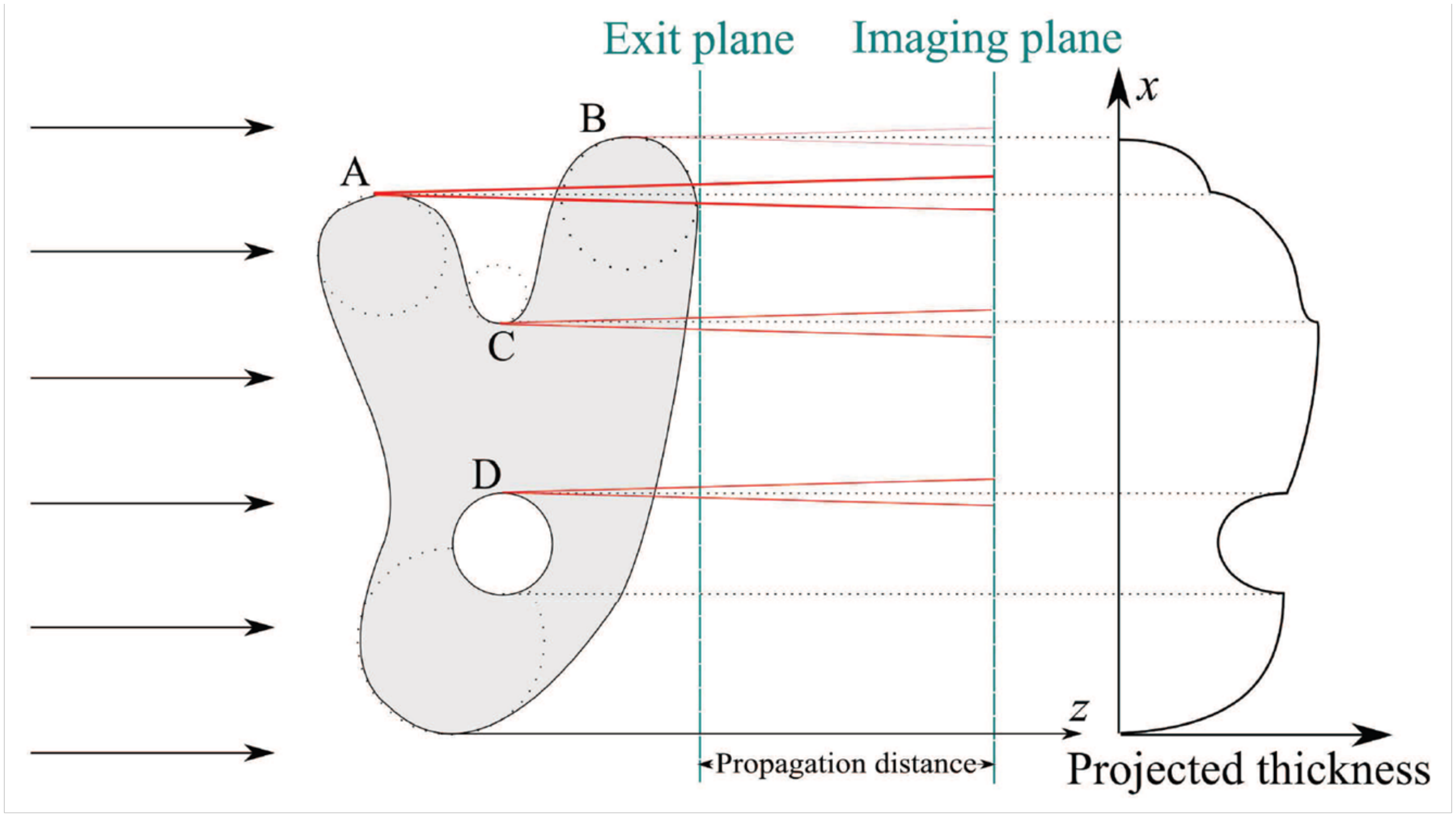
Figure 3.41 Refractive index versus frequency.

The refractive index, for X-ray wavelengths is less than 1. In terms of its real and imaginary components the index of refraction for X-rays is normally given as: $\boldsymbol{n} = 1 - \boldsymbol{\delta} + \boldsymbol{i\beta}$

Where ,$\boldsymbol{\beta}$ the imaginary component, determines the absorption properties of the material and δ the real part of the refractive index modulates the phase.

We can now define a complex transmission function which characterizes the amplitude and phase modulation imparted to an incident wavefield via propagation through a particular material of thickness: $\boldsymbol{T}(\vec{\boldsymbol{r}}) = |\boldsymbol{T}(\vec{\boldsymbol{r}})| = \exp(-\boldsymbol{\beta}(\vec{\boldsymbol{r}})\boldsymbol{kt}(\vec{\boldsymbol{r}}))\exp(\boldsymbol{i\delta}(\vec{\boldsymbol{r}})\boldsymbol{kt}(\vec{\boldsymbol{r}}))$

Imaging and Phase Retrieval

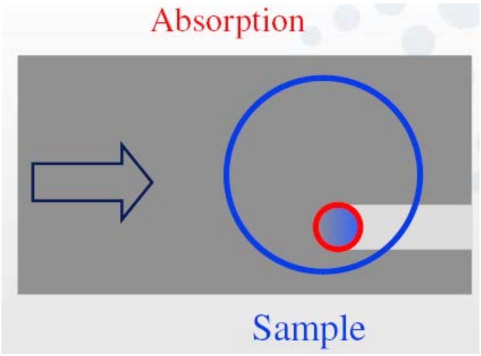


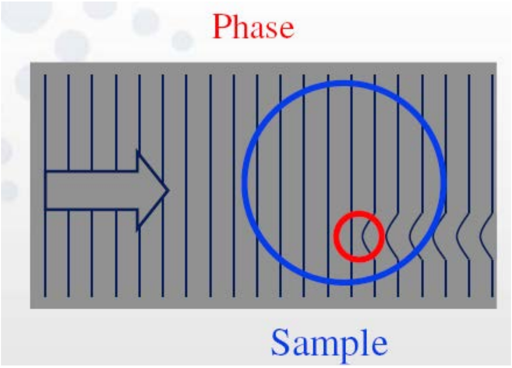
$\psi_0 = E_0 \exp(ikz)$

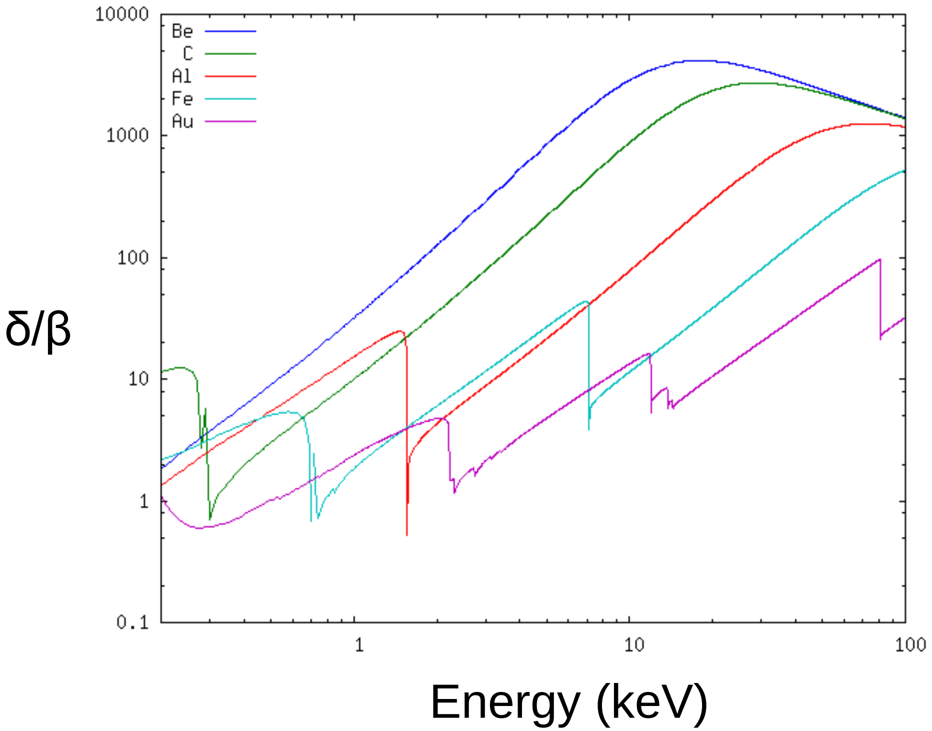
$\psi(z) = \psi_0 \exp(-i\delta kz)\exp(-\beta kz)$

Why Care About Phase Contrast?

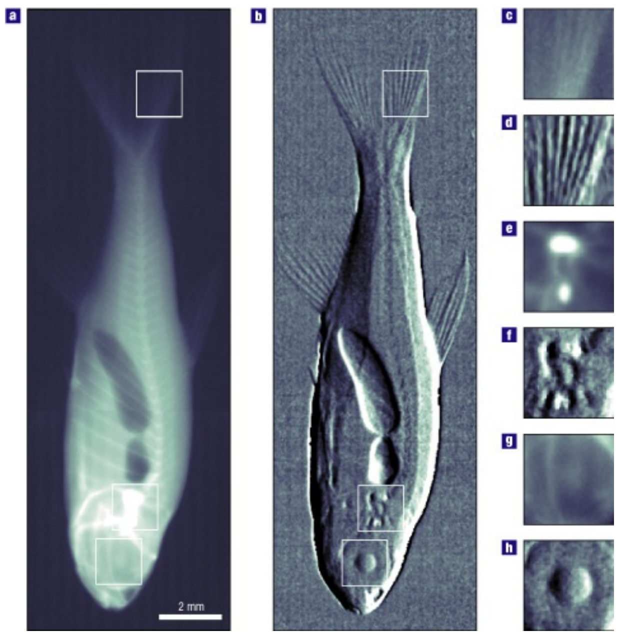
complex refractive index: n=1-δ-iβ.

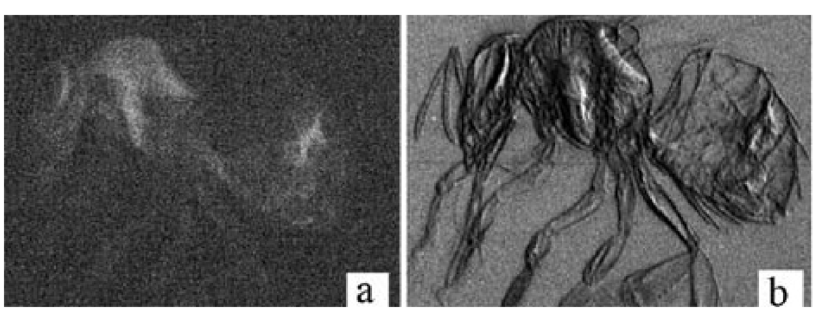


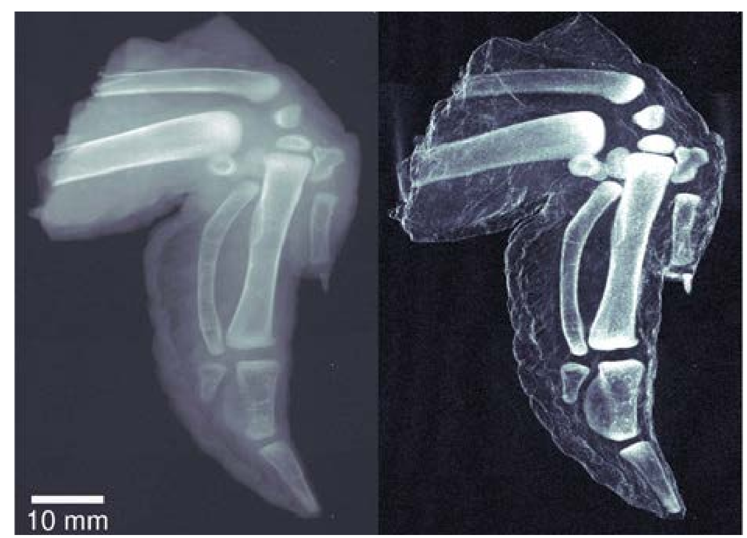




Up to 1000 times or more improvement in contrast that's why!

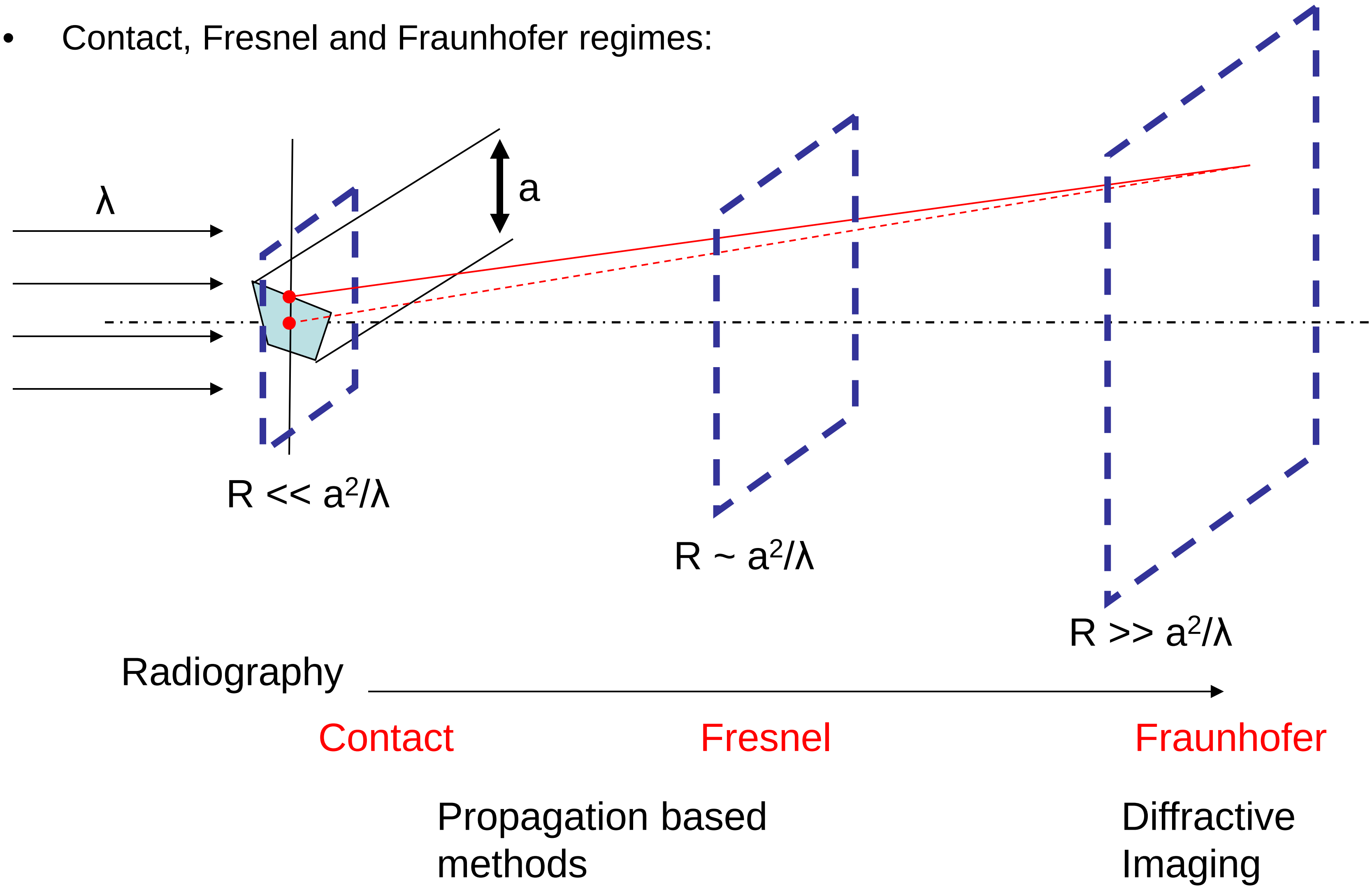




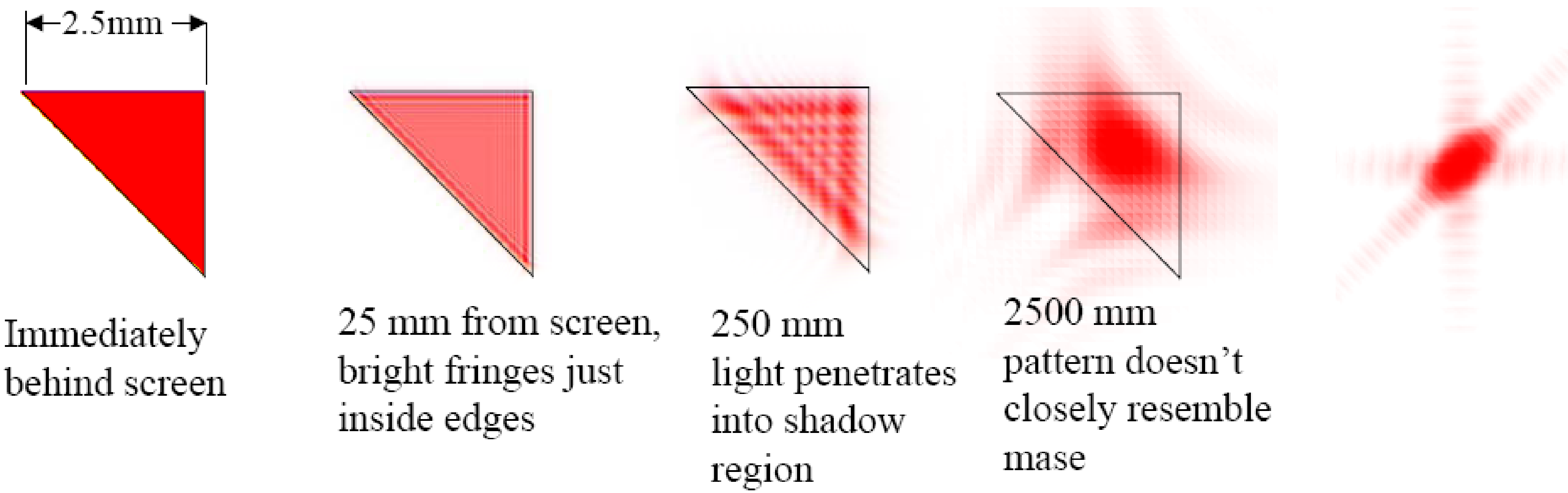


Phase Contrast (I)

- Contact, Fresnel and Fraunhofer regimes:

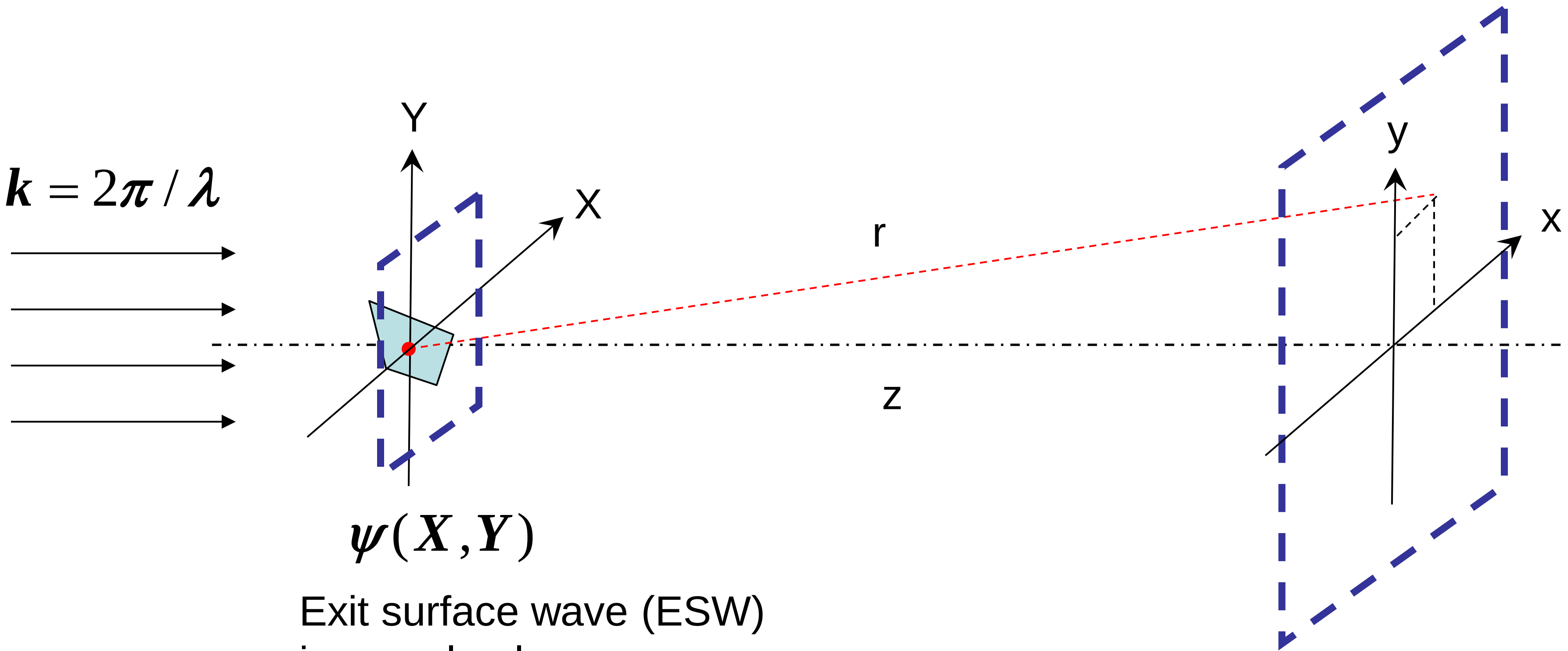


Phase Contrast (I)



In the far-field the diffraction pattern no longer changes just scales with propagation distance.

Phase Contrast (II)



$\psi(X, Y)$

Exit surface wave (ESW)
in sample plane

$\psi(x, y)$

Propagated wave
in detector plane

Phase Contrast (II)

Contact regime: Recall projection approximation for X-rays (weakly interacting). Intensity distribution is simply given by Beer-Lambert for absorption in a homogeneous medium:

$$I = I_o e^{\frac{-4\pi}{\lambda}\int_{t1}^{t2} \beta(t)dt} = I_o e^{-\mu T}$$

(only have access to imaginary part of refractive index)

Fresnel Regime (wavefront curvature cannot be neglected):

$$\psi(x,y) = \frac{\exp(ikz)}{i\lambda z}\exp\left[\frac{ik}{2z}(x^2+y^2)\right]$$

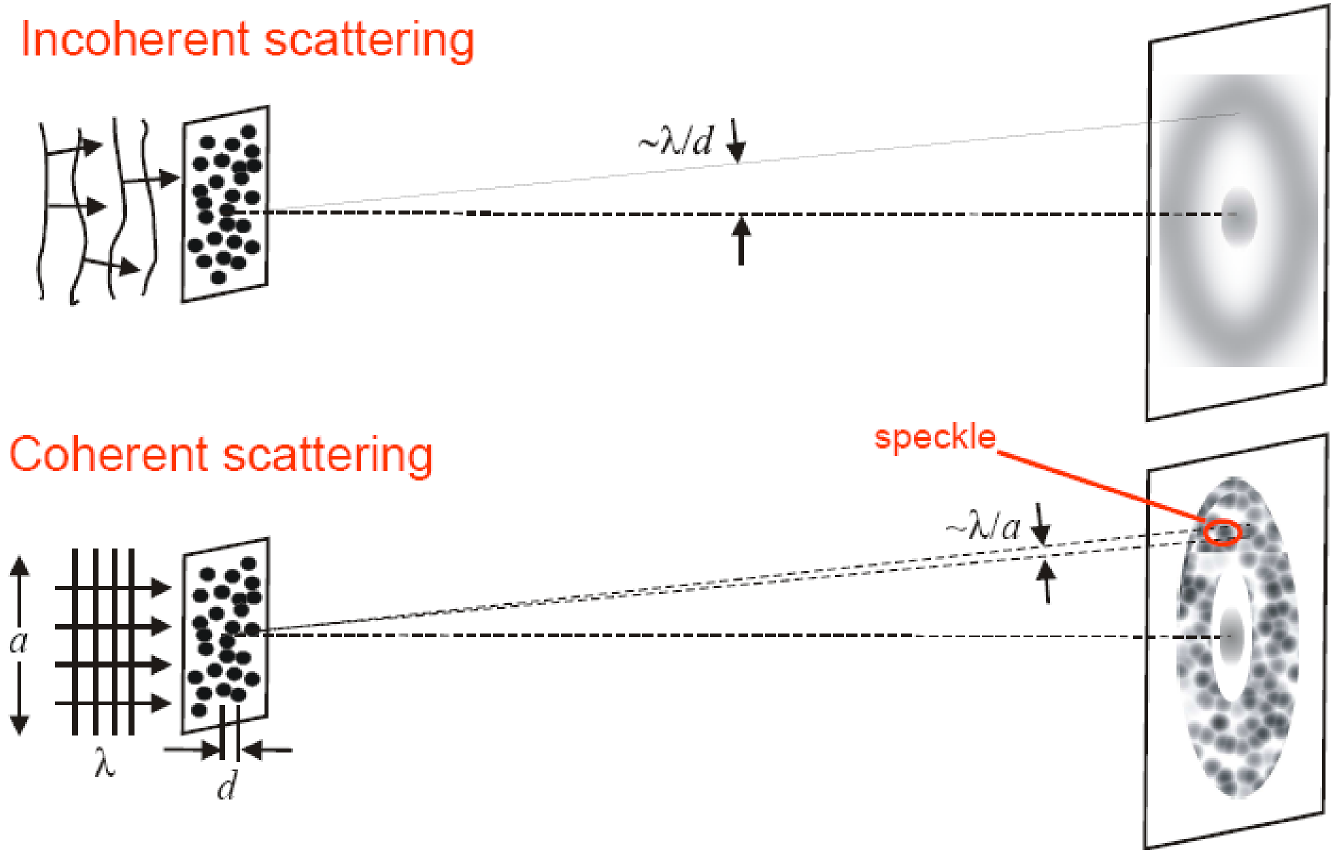
$$\times \int\int \psi(X,Y)\exp\underbrace{\left\{\left[\frac{ik}{2z}(X^2+Y^2)\right]\right\}}_{\text{quadratic terms account for spherical curvature of wavefront}}\exp\left[-\frac{ik}{z}(xX+yY)\right]dXdY$$

Fraunhofer regime:

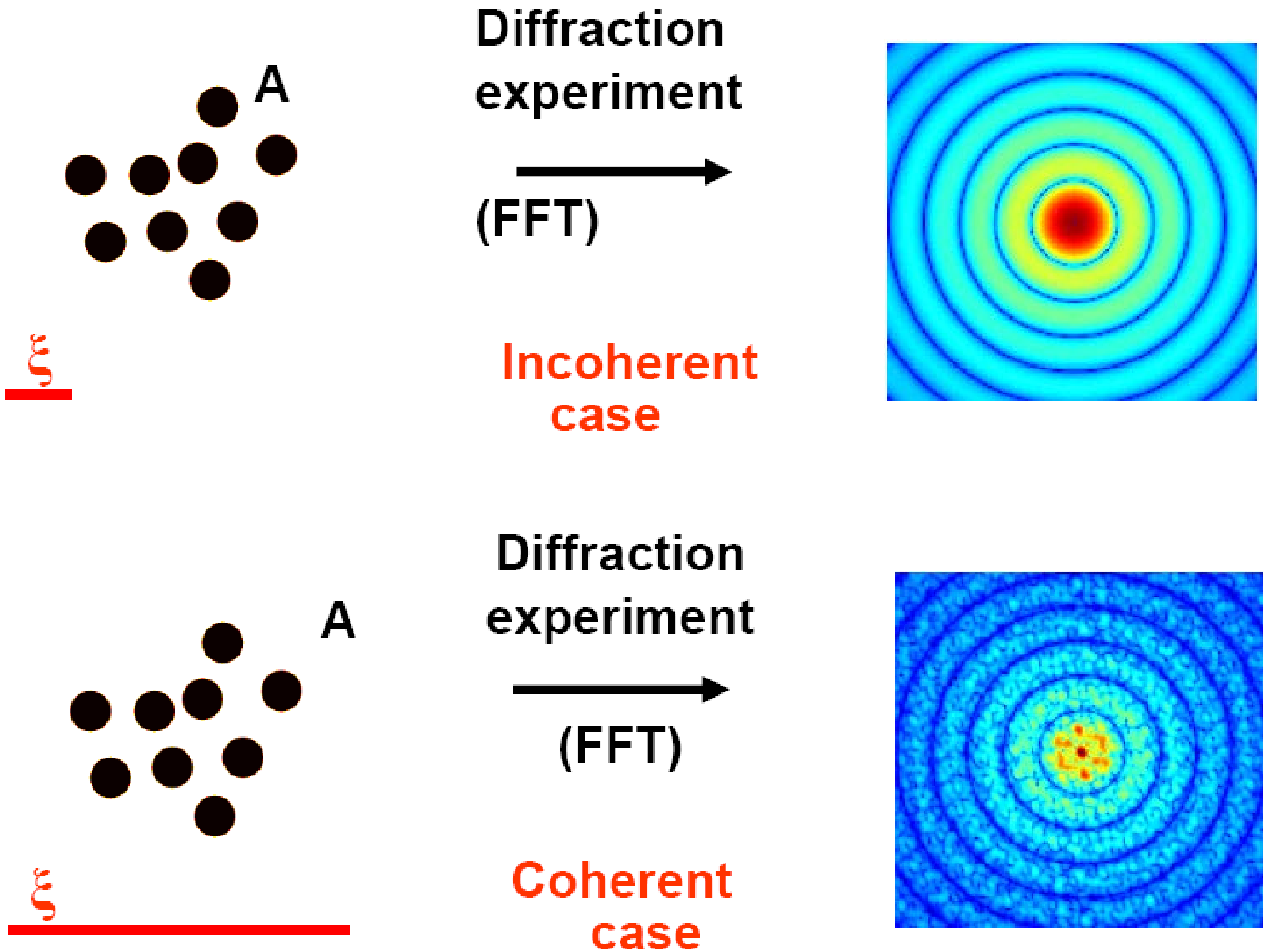
$$\psi(x,y) = \frac{\exp(ikz)}{i\lambda z}\exp\left[\frac{ik}{2z}\left(x^2+y^2\right)\right]\int\int \psi(X,Y)\exp\left\{-\frac{ik}{z}\left[(xX+yY)\right]\right\}dXdY$$

which is just the FT of the sample ESW

Speckle Experiments



Speckle Experiments



The Phase Problem

The phase problem: We have seen that in principle the amplitude and phase of the wave exiting the sample may be recovered from the diffracted wave via an inverse FT.

$$\boldsymbol{f}(\boldsymbol{x}) = \int \boldsymbol{F}(\boldsymbol{u}) \exp(-\boldsymbol{iux})\boldsymbol{du}$$

However in practice we can only directly measure the intensity which we may take to be the square of the magnitude of the FT: $\boldsymbol{I} = \boldsymbol{F}(\boldsymbol{x})^* \boldsymbol{F}(\boldsymbol{x}) = \left|\boldsymbol{F}(\boldsymbol{x})\right|^2$

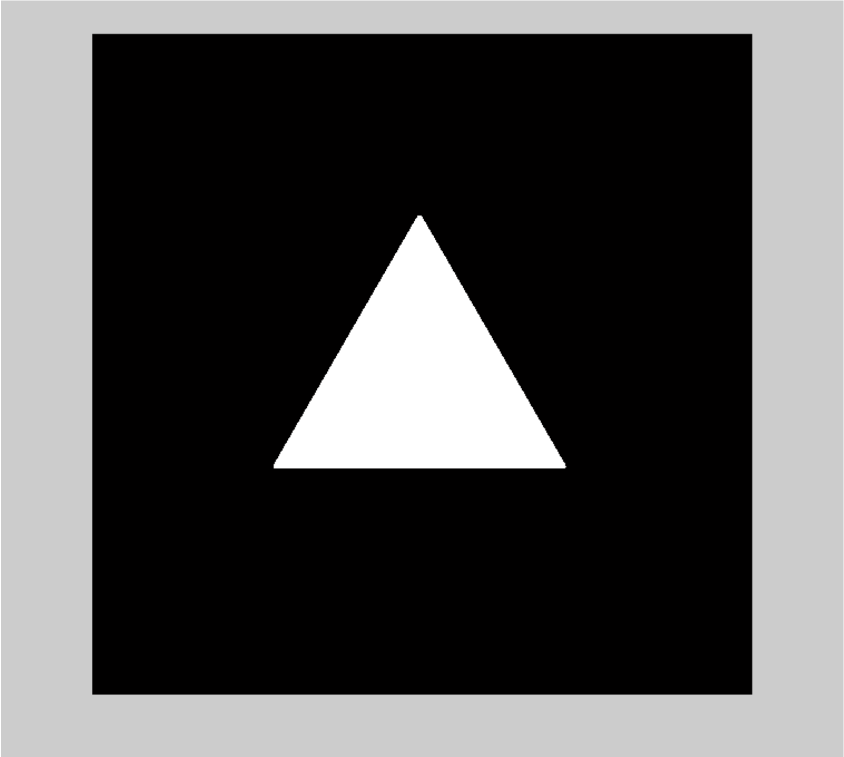
This quantity cannot be used directly to determine the absolute phase of F(x).

We must use indirect means to retrieve the phase of the diffracted wavefield which may then be transformed to give the phase of the wavefield exiting the sample.

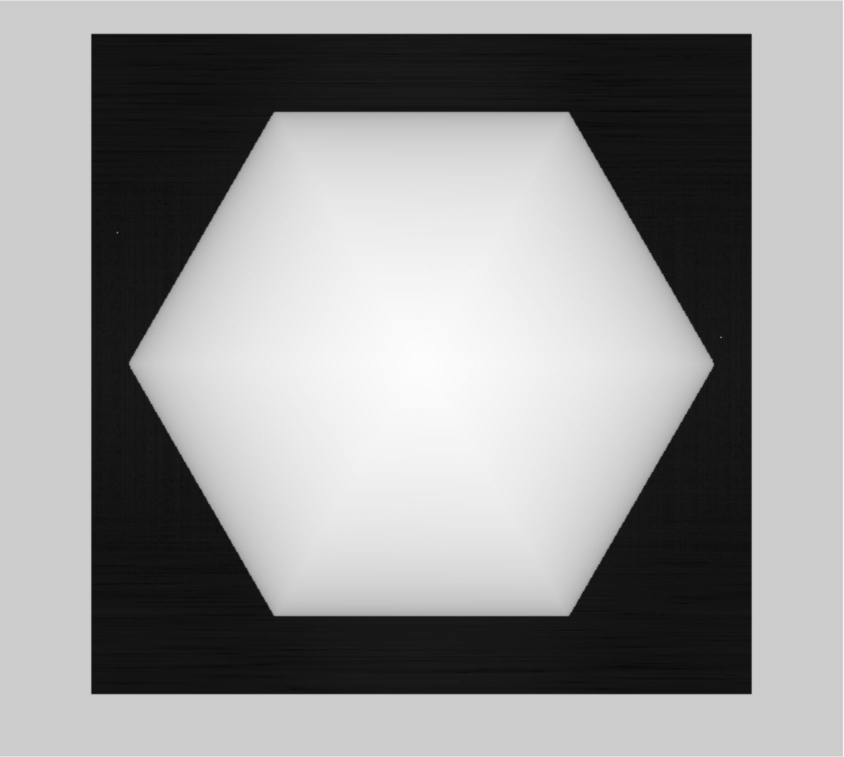
Note that the FT^{-1}: $\int \left|\boldsymbol{F}(\boldsymbol{u})\right|^2 \exp(-\boldsymbol{iux})\boldsymbol{du} = \boldsymbol{f}(\boldsymbol{x}) \otimes \boldsymbol{f}(\boldsymbol{x})$

Where the convolution: $\boldsymbol{f}(\boldsymbol{x}) \otimes \boldsymbol{f}(\boldsymbol{x})$ is known as the autocorrelation function.

The Autocorrelation function

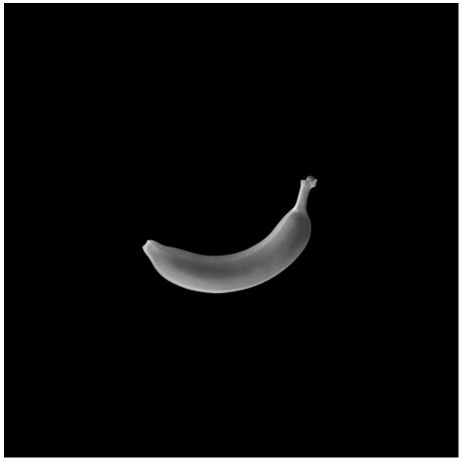


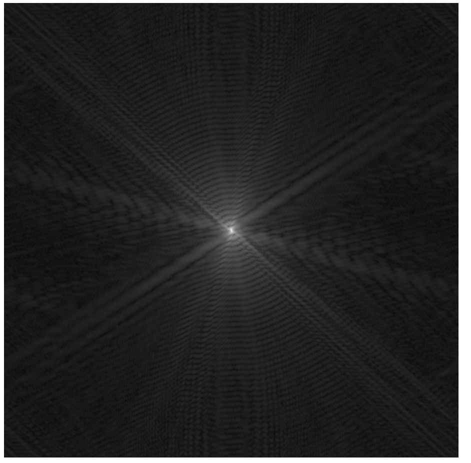
triangle



log of autocorrelation of triangle

Autocorrelation and Sampling





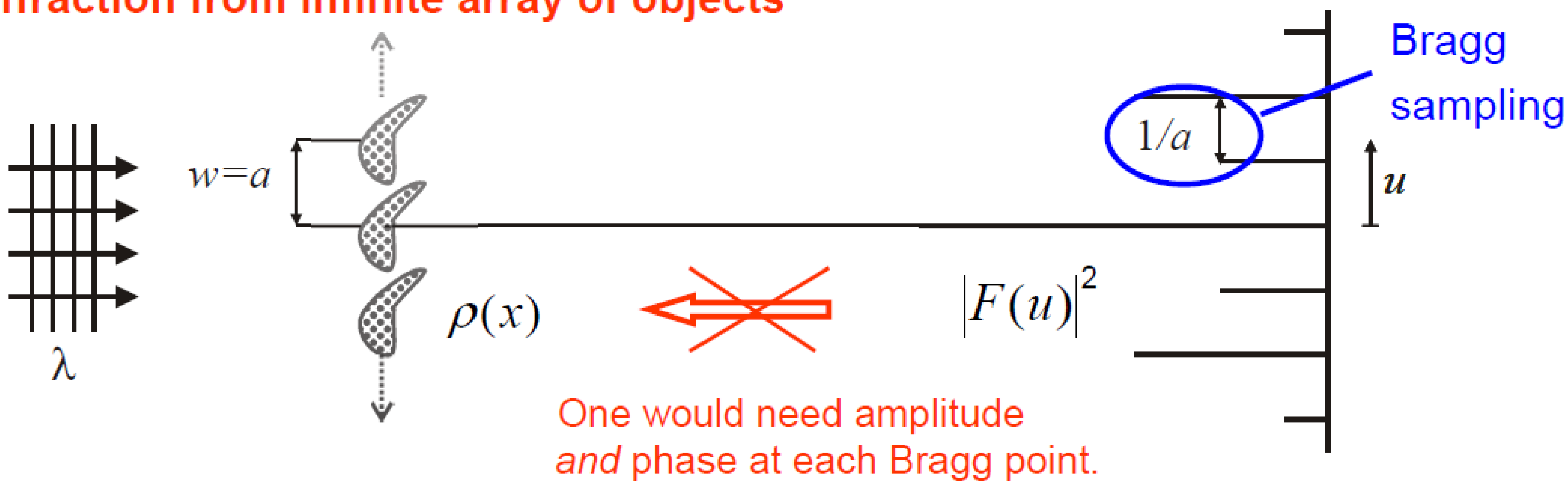
autocorrelation + Banana

autocorrelation of Banana

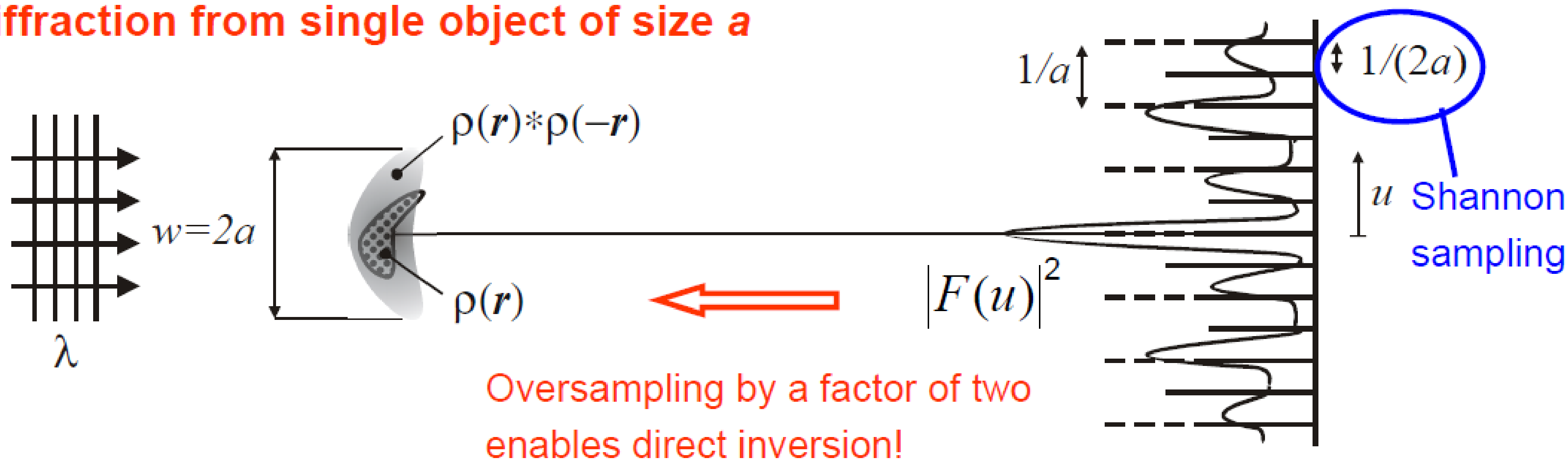
Coherent Diffractive Imaging

- Bragg diffraction Vs. continuous diffraction

Diffraction from infinite array of objects



Diffraction from single object of size *a*



Phase Retrieval

There are numerous approaches to obtaining the phase of the sample ESW, the main approaches are:

Propagation based phase imaging: Transport of Intensity (TIE), Transfer function approach.

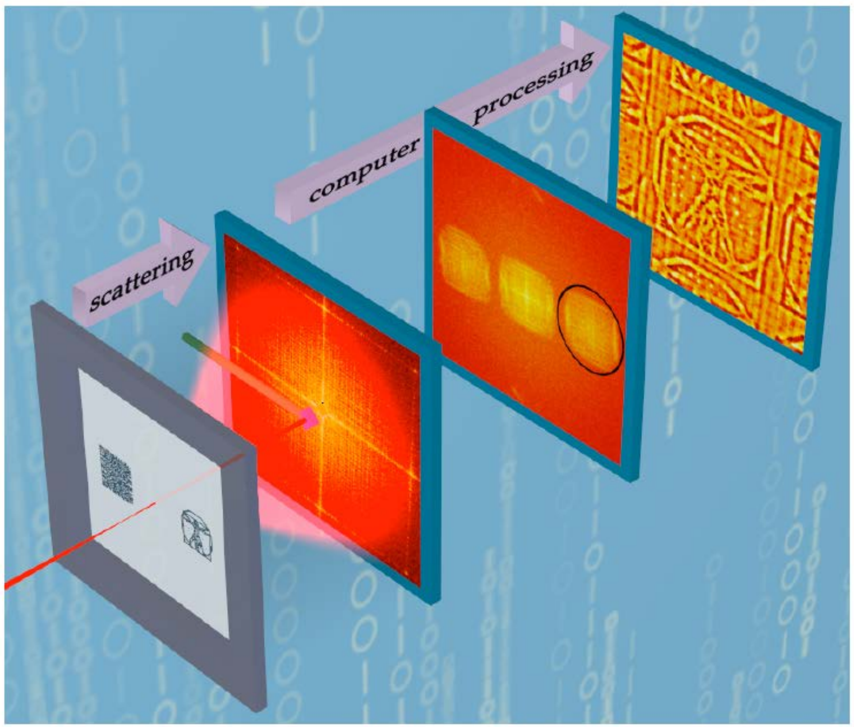
Interference methods: In-line Holography, interferometry.

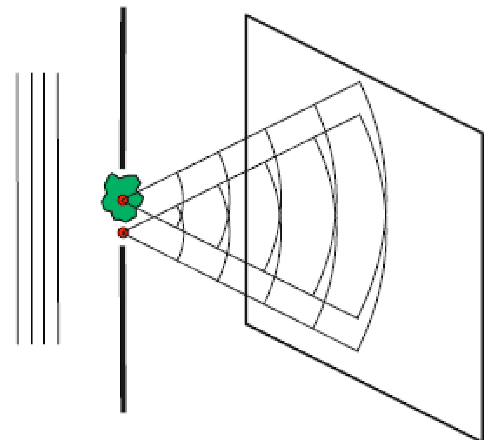
Diffraction techniques: Fourier Transform Holography, Coherent diffractive imaging.

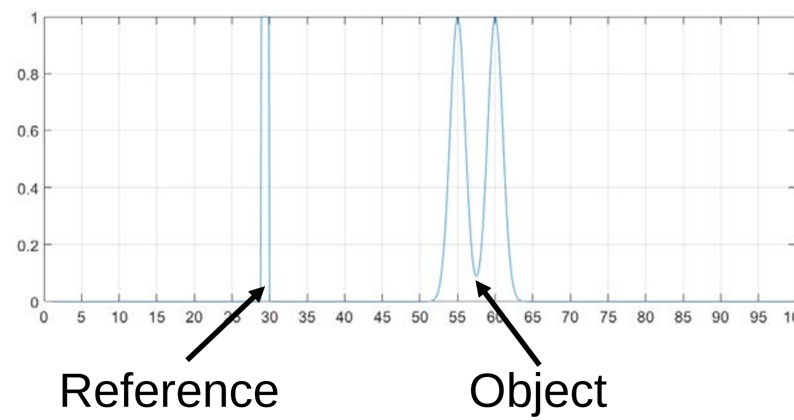
Here we will look at two diffraction techniques (far-field).

Fourier Transform Holography

- Holography: interference between sample and reference waves.







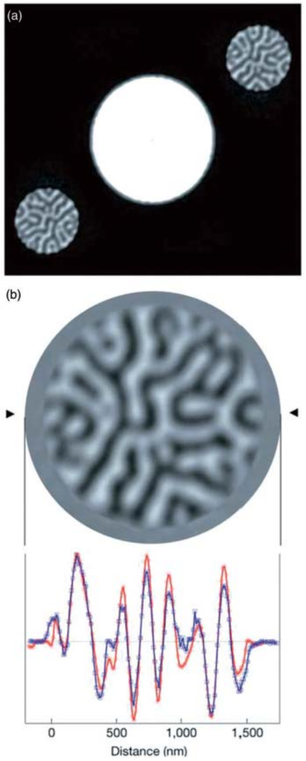
Principles behind FTH

The autocorrelation function in FTH which is obtained from applying the FT^{-1} to the far-field intensity distribution (assuming no phase information) is the sum of four terms:

$$G_T(\vec{r}) = |A|^2 \delta(\vec{r}) + G_f(\vec{r}) + Af^*(\vec{r} - \vec{d}) + A^* f(\vec{r} + \vec{d})$$

Terms:

1. Autocorrelation of aperture
2. Autocorrelation of object
3. Conjugate image of object
4. Primary image of object

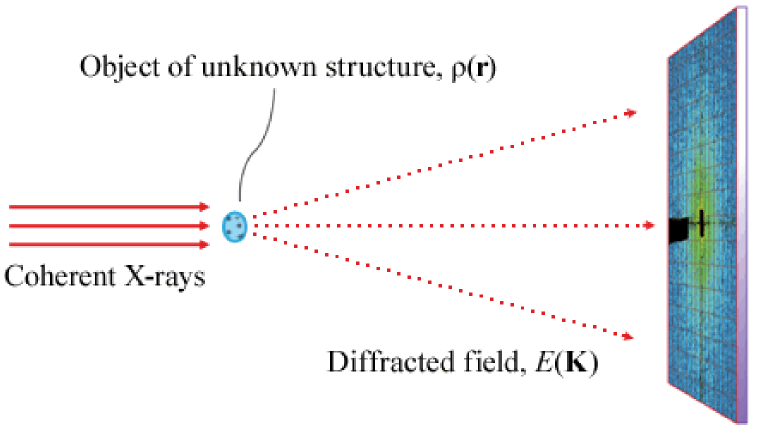


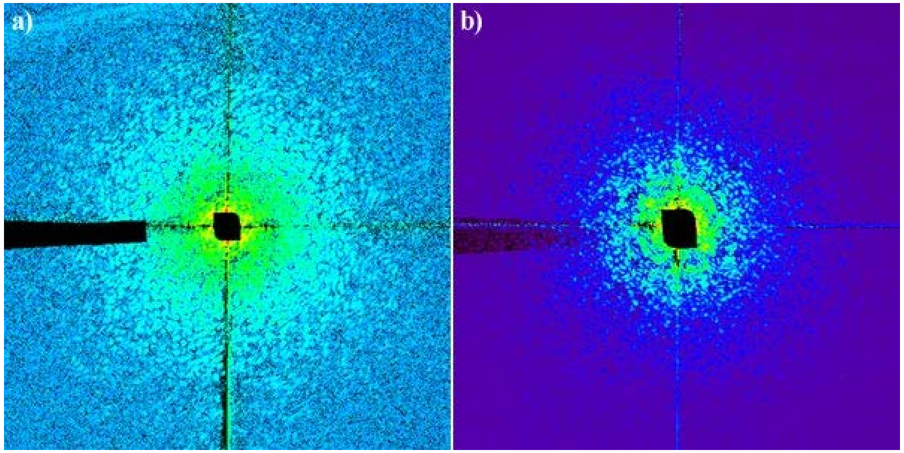
Magnetic domain structure

In practice the finite width of the reference aperture means that the δ function should be replaced by a known function . The primary and conjugate images are then convolved with the autocorrelation and self convolution respectively of . Provided is still reasonably sharp (i.e. close to being a δ function) the blurring this convolution introduces does not reduce the resolution of the reconstruction significantly.

Coherent Diffractive Imaging

- Coherent diffractive imaging

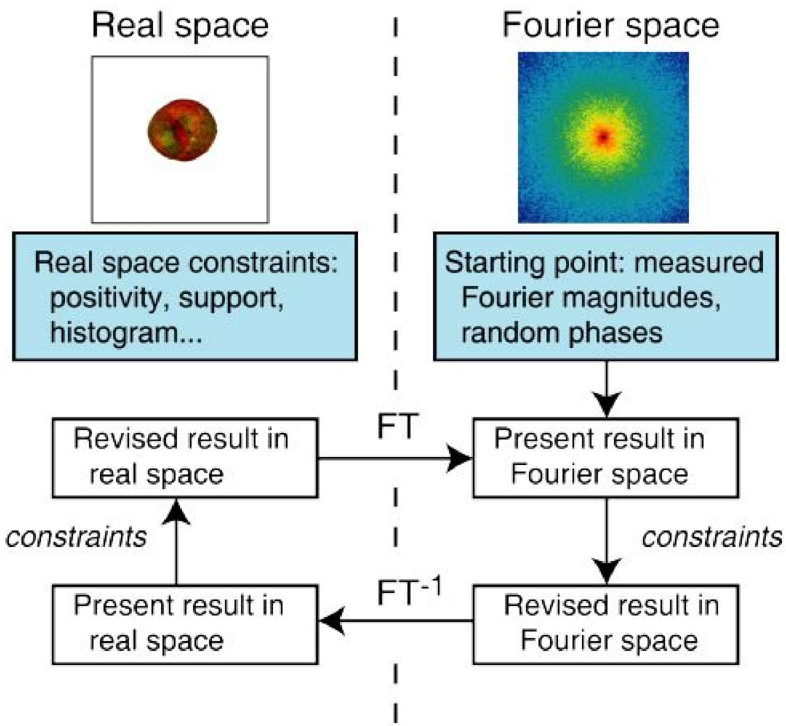




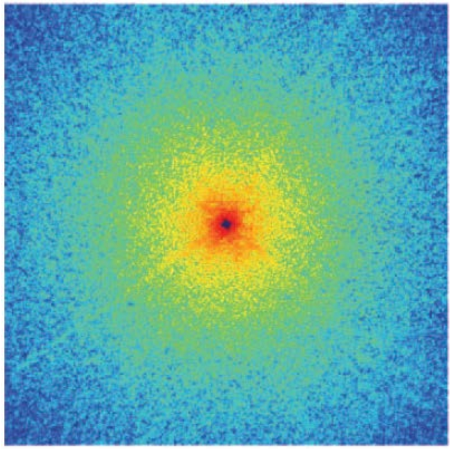
Diffraction from unstained bacteria

Algorithms: error reduction

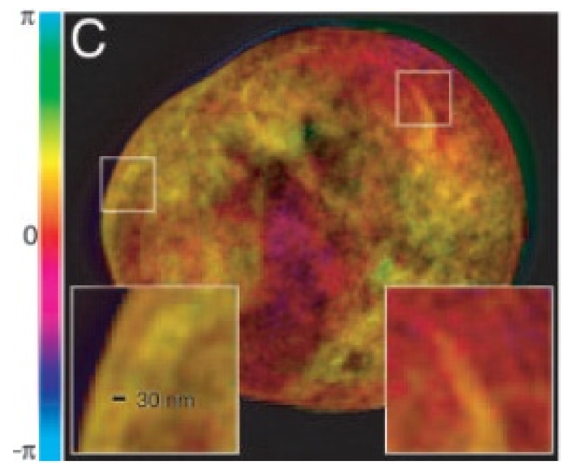
- G-S/Error reduction



Start from this:

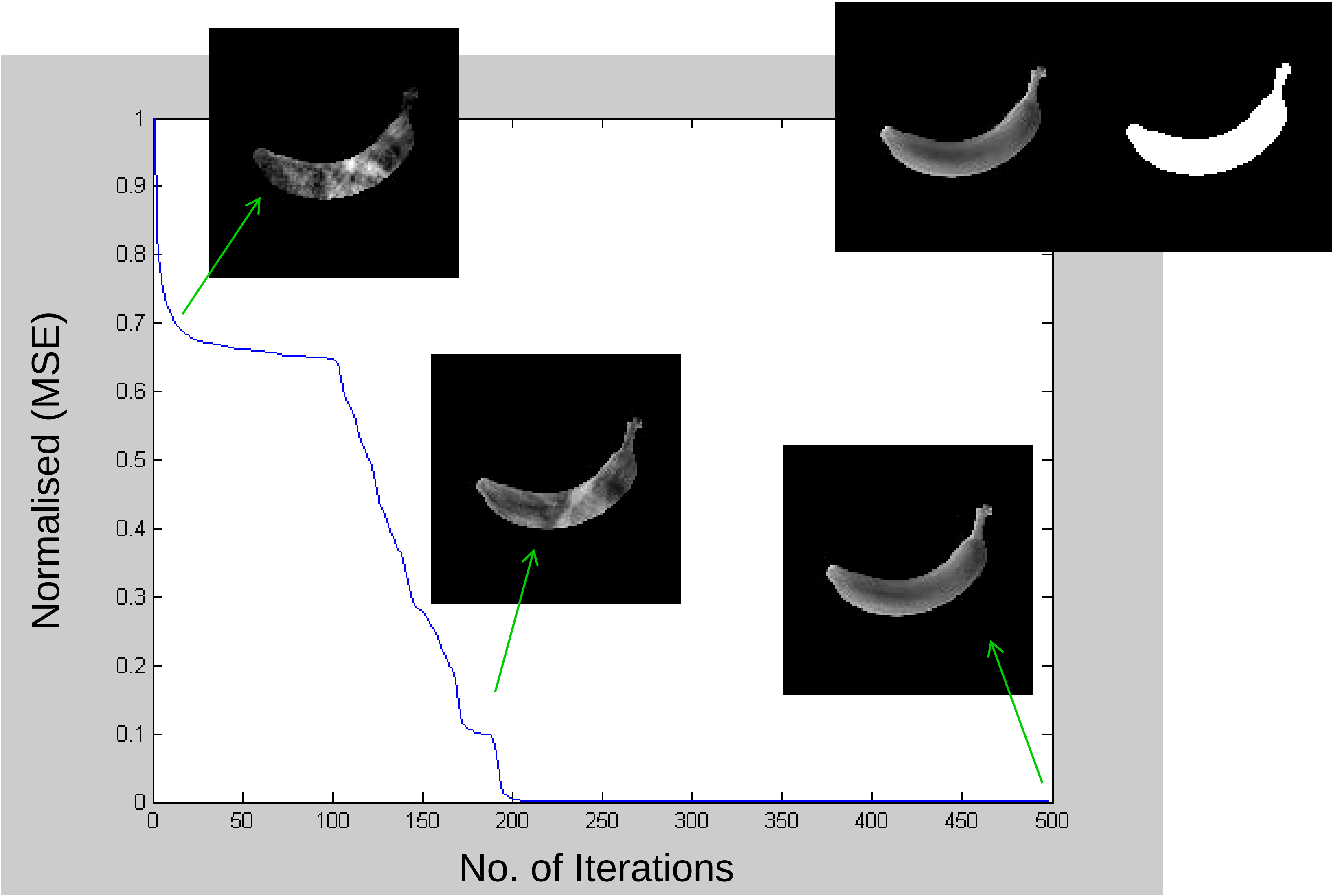


End with this:



Shapiro et al. PNAS, 2005

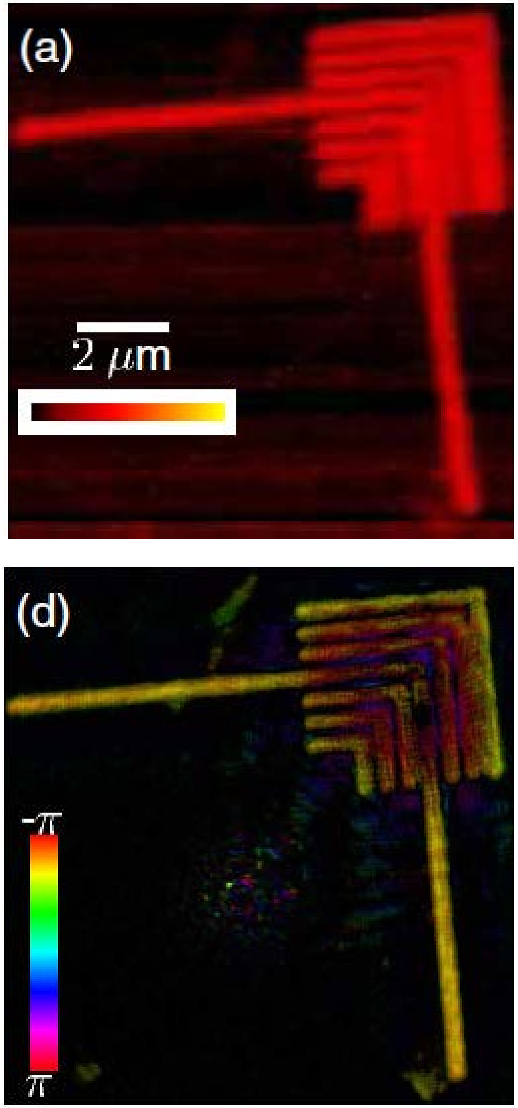
The plane wave Banana



Fresnel Coherent Diffractive Imaging

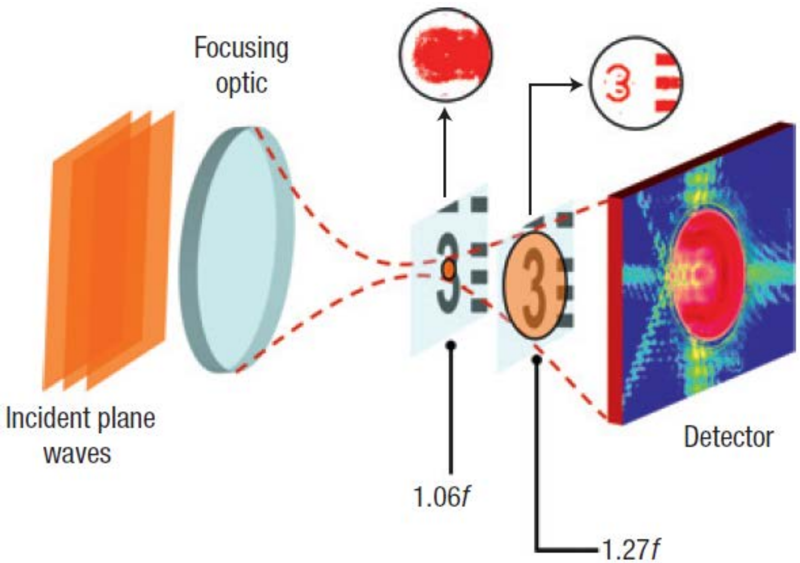
CDI with curved incident illumination

Williams et al., PRL, 2007



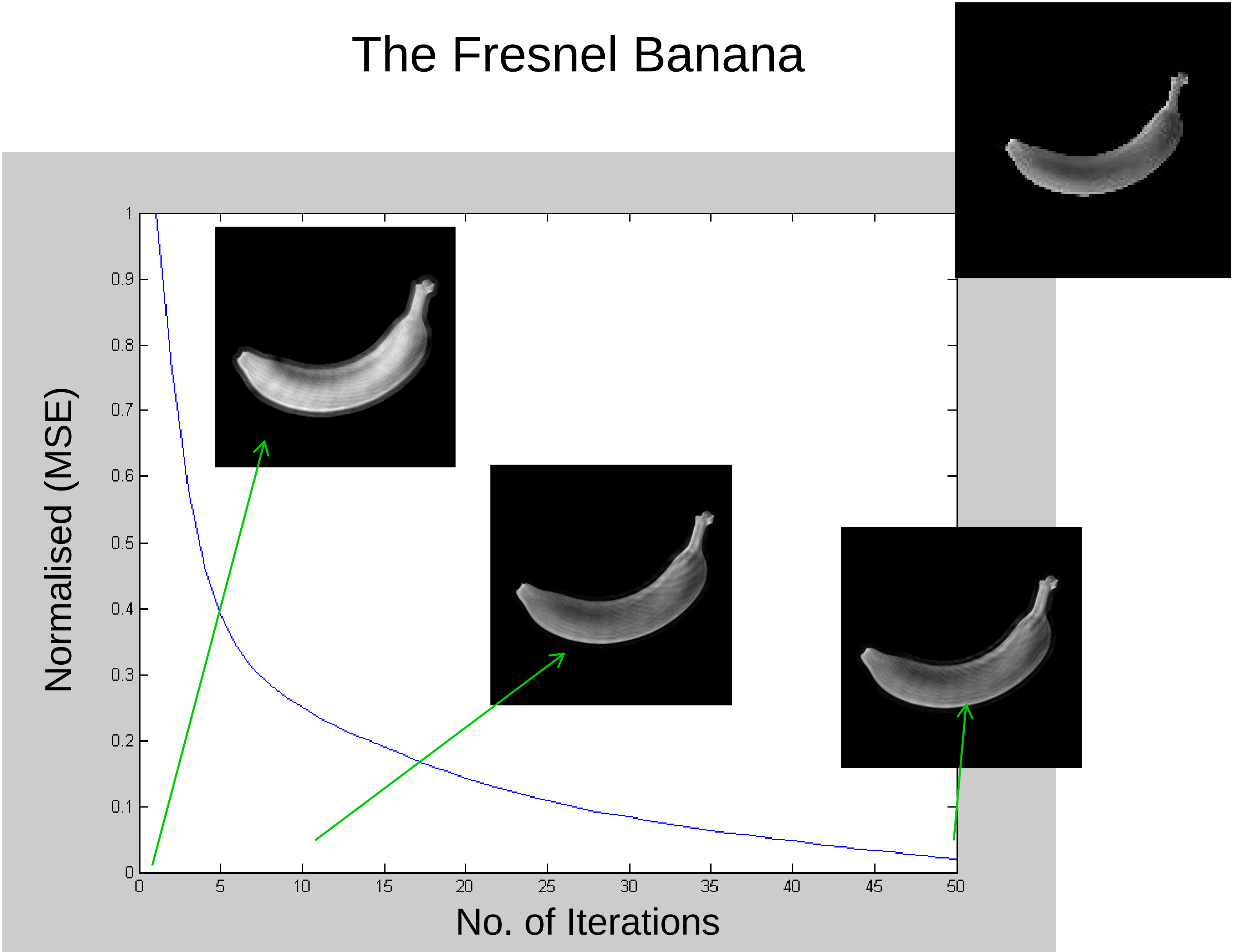
Benefits:

Favourable convergence characteristics
More robust with respect to partial coherence
Can image ‘extended objects’

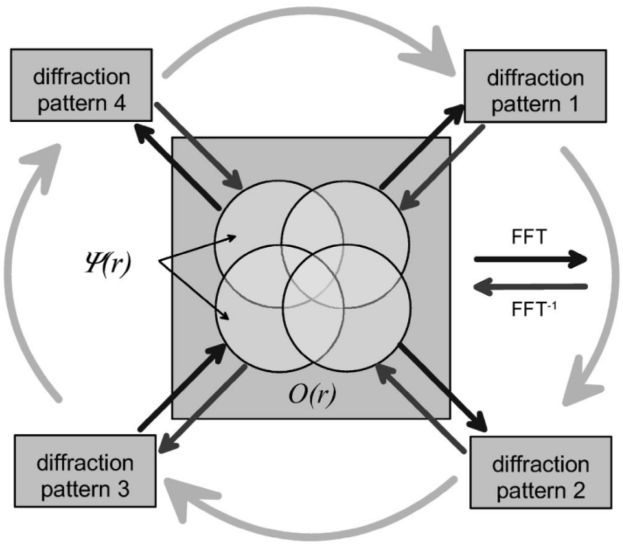


Abbey et al., Nature Phys., 2008

The Fresnel Banana

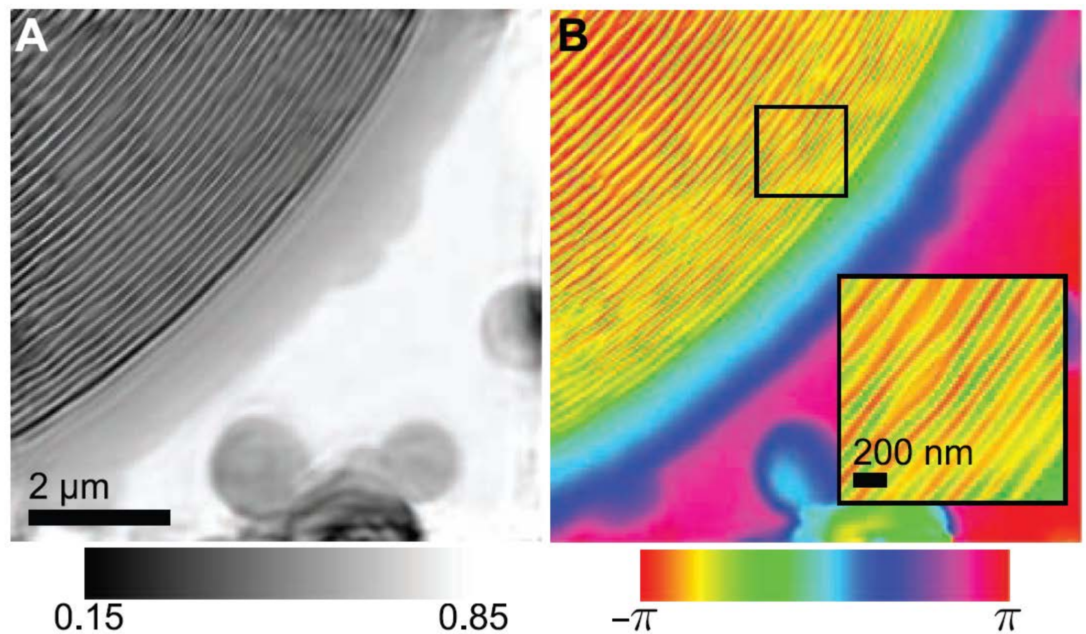


Ptychographic Diffractive Imaging



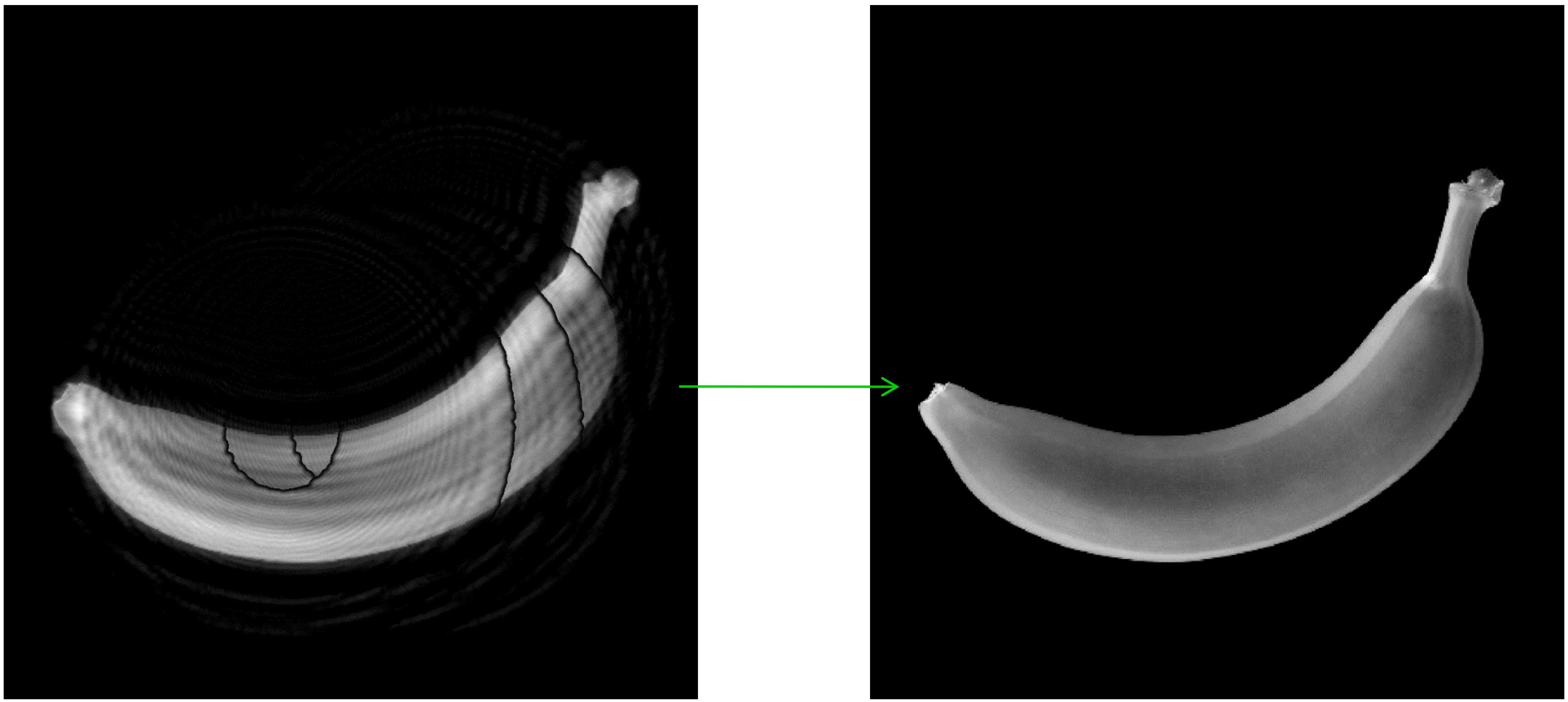
The Ptychographic Iterative Engine (PIE) is a means of reconstructing extended objects with plane wave illumination using the redundant information contained in the overlap of multiple exposures.

The method has been shown to converge rapidly and provides very high quality images of extended objects.



Thibault et al., Science, 2008

The Ptychographic Banana



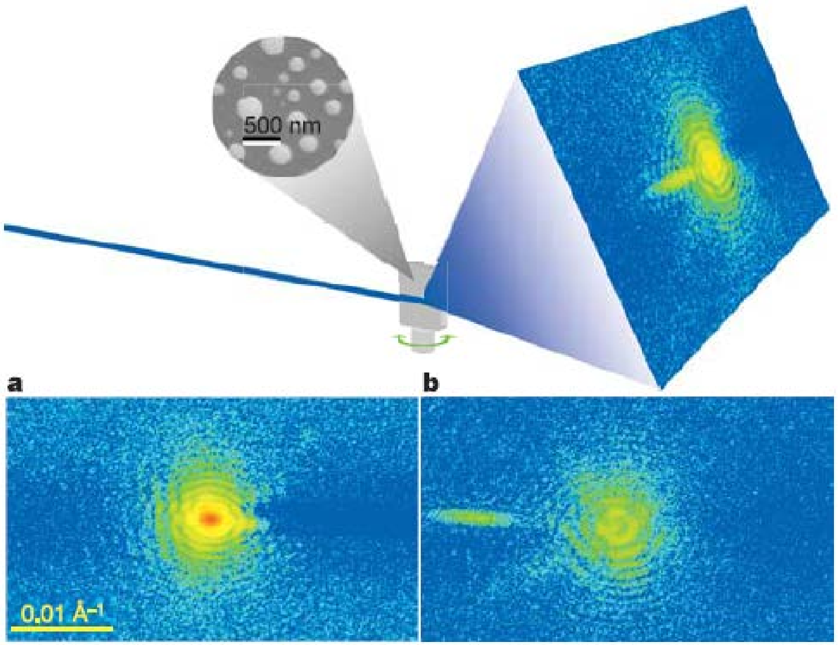
Initial ‘Guess’ Final Result

Bragg diffraction and CDI

Bragg diffraction from a finite crystal:

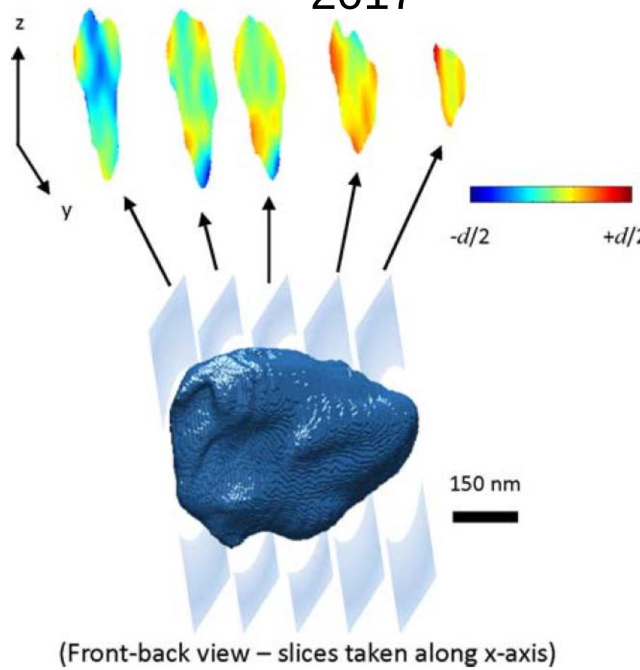
Each Bragg spot is itself a continuous diffraction pattern containing information about the overall shape of the crystal.

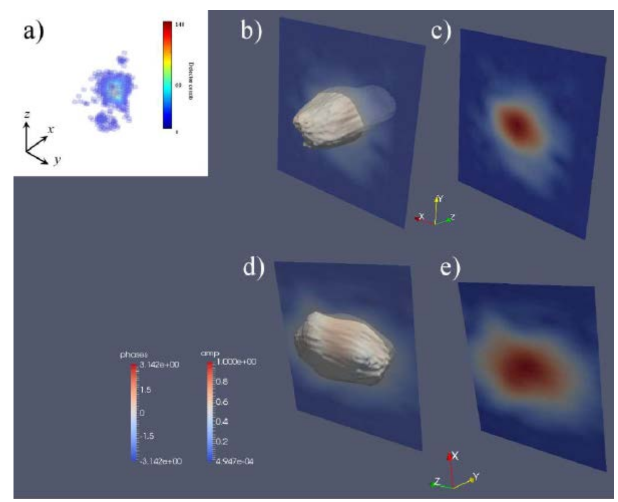
Departure from perfect spatial symmetry is indicative of strain within the nanocrystal



Robinson et al., Nature, 2007

Maqbool et al., IJNT, 2017

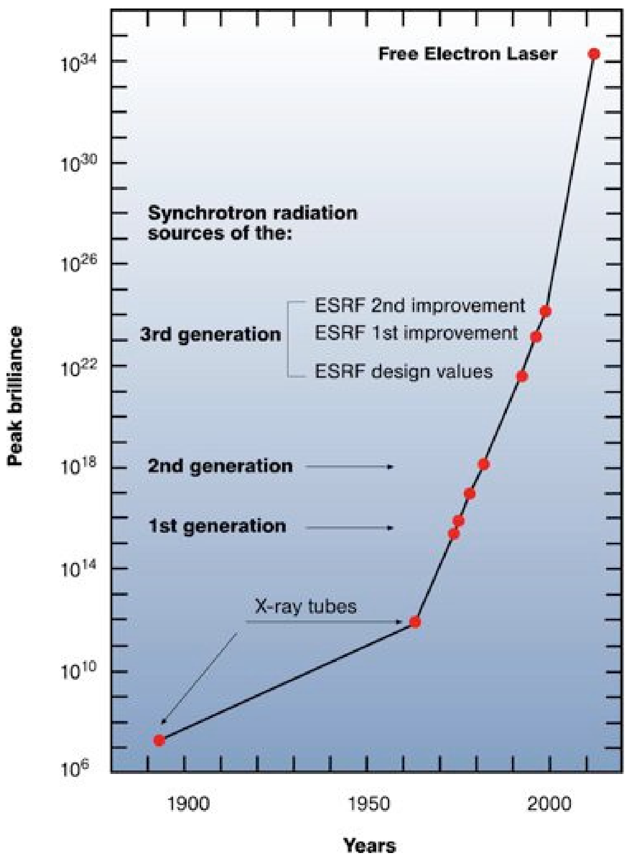


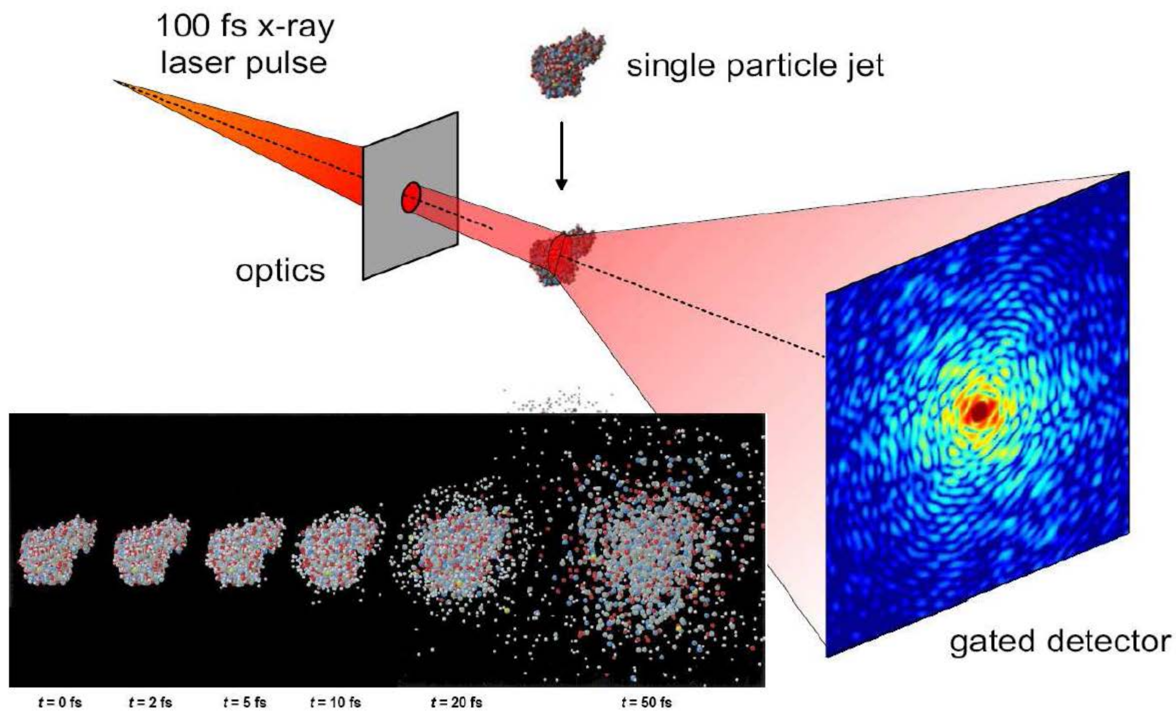


Coughlan et al., Journal of Optics, 2016

Future Directions: XFELS

Peak brilliance many times brighter than conventional 3rd generation sources.

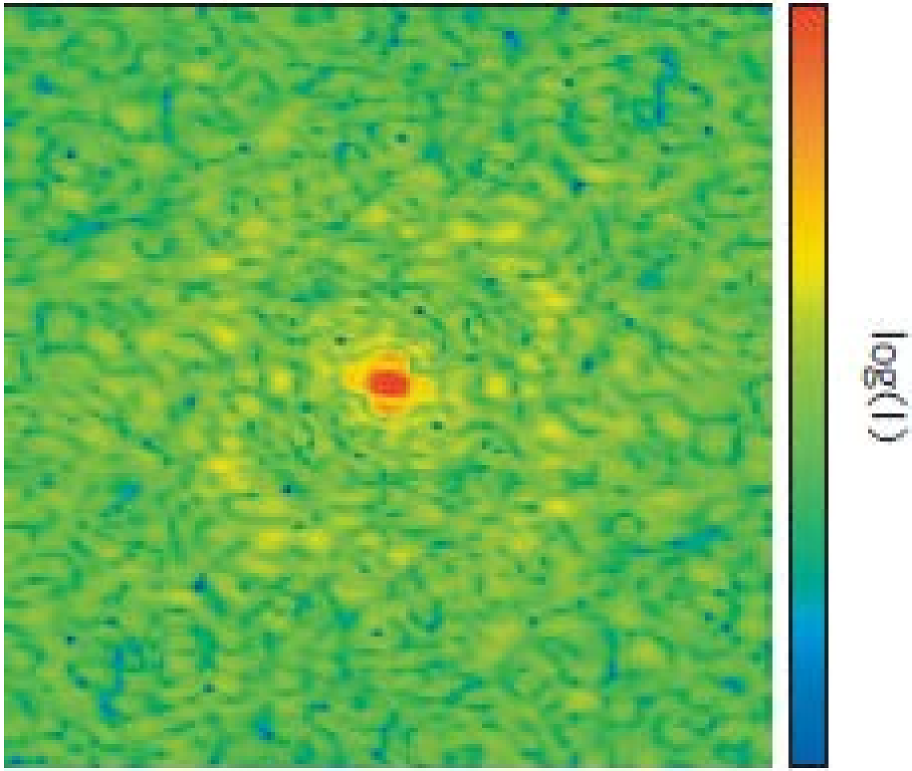


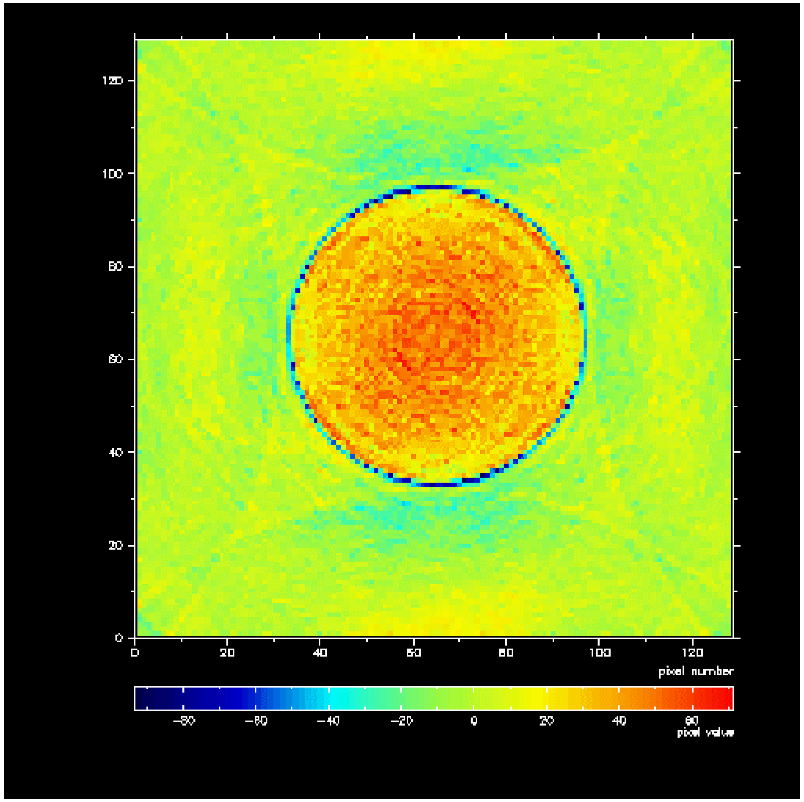


At very short pulse durations < 5 fs there is the potential for imaging single molecules using CDI.

Neutze et al., Nature, 2000

Reconstruction of Bacteriorhodopsin





Quiney & Nugent et al., Nature Physics, 2011

Opportunities in Australia

Look into crystal ball

Melbourne scientists in molecule breakthrough

MARK DUNN

MELBOURNE researchers have accidentally discovered how to transform molecules into a new type of crystal — a

and molecules, the machines of life," Prof Abbey said.

"Being able to see these structures in new ways will

nut with a sledgehammer and instead of destroying it and shattering it into a million pieces, we instead created a differ-