Methoden moderner Röntgenphysik I

Coherence based techniques II

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Outline

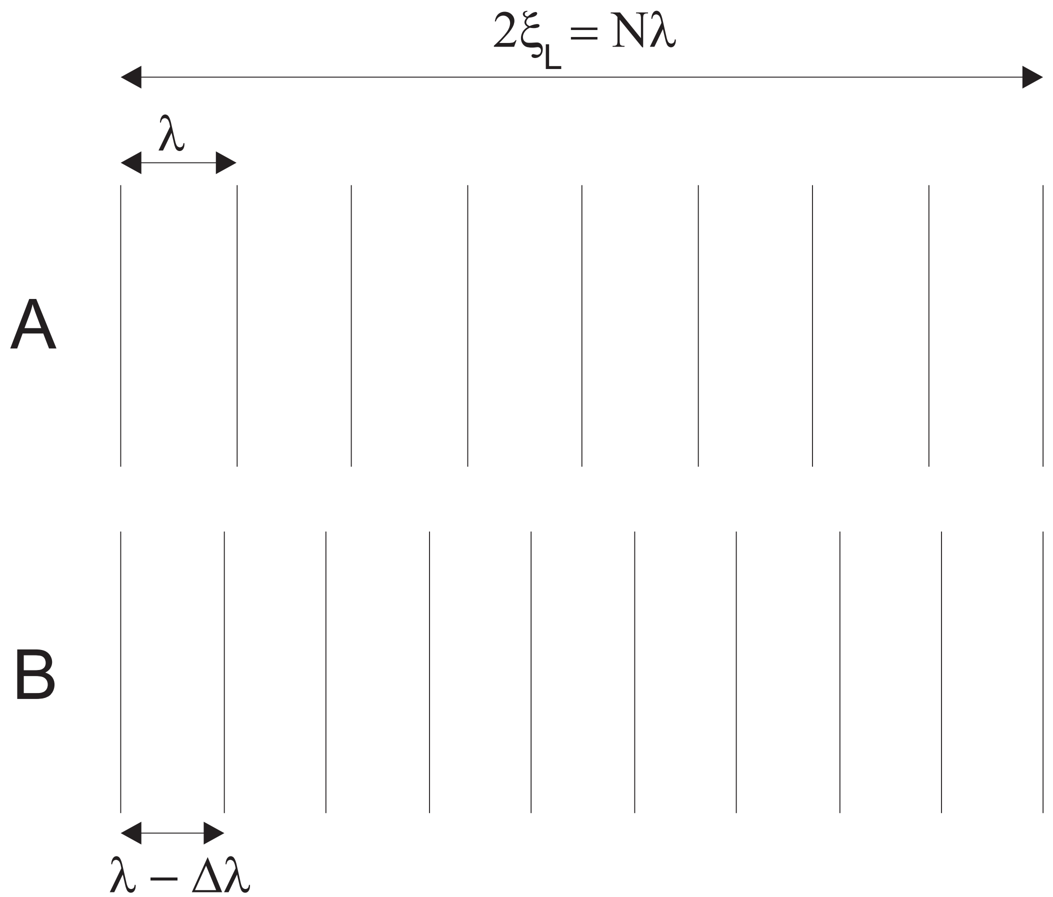
18.12. 2008
Introduction to Coherence

8.01. 2009
- **Structure determination techniques**
- **Oversampling**
- **Coherent Diffractive Imaging**
- **Fourier transform Holography**

15.01.2009
Correlation Spectroscopy

Last lecture

Longitudinal coherence



$$N\lambda = (N+1)(\lambda - \Delta\lambda)$$

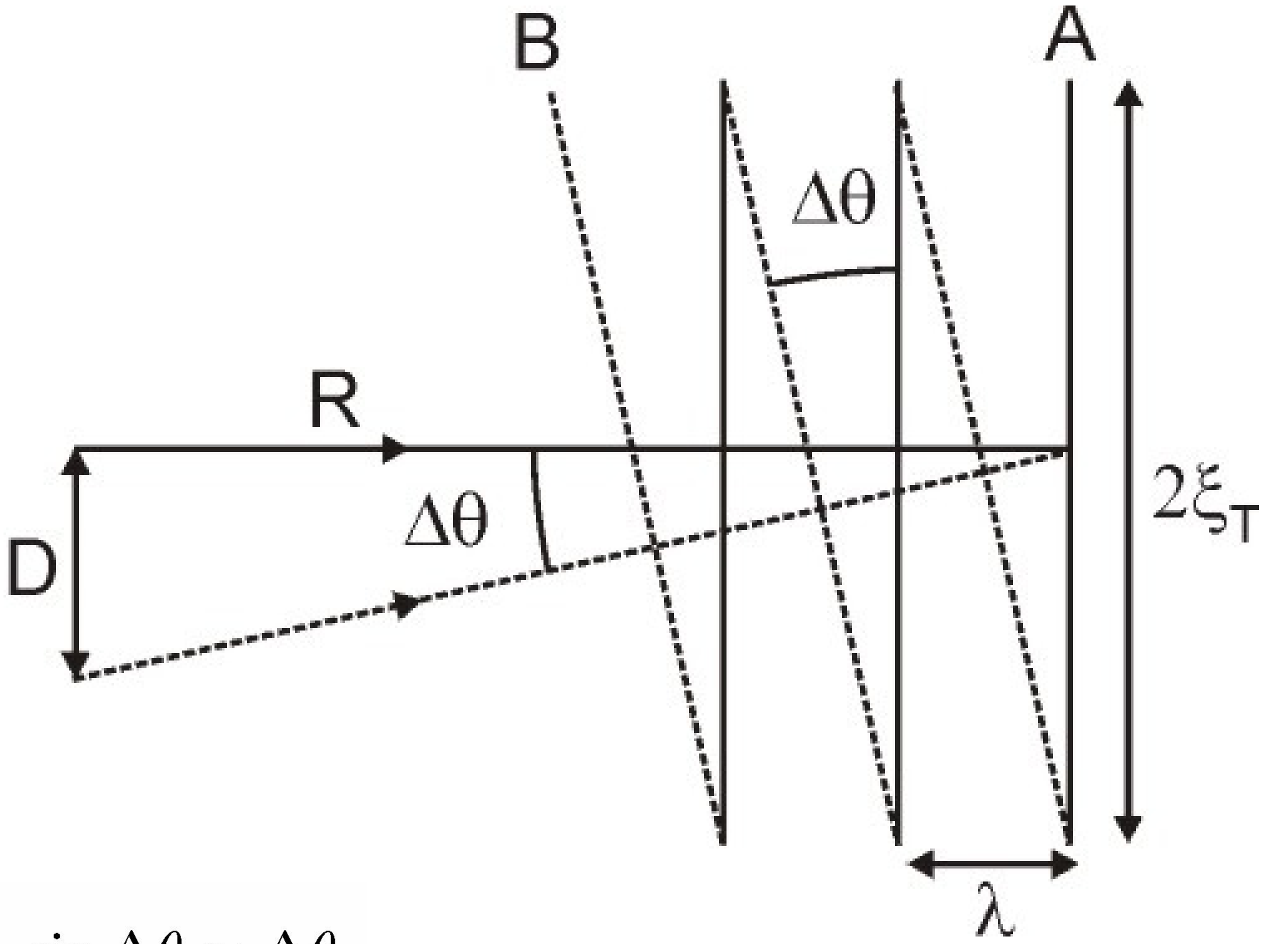
$$(N+1)\Delta\lambda = \lambda$$

$$N \approx \frac{\lambda}{\Delta\lambda}$$

$$\xi_L = \frac{N\lambda}{2} = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

longitudinal coherence depends on bandwidth

Transverse coherence



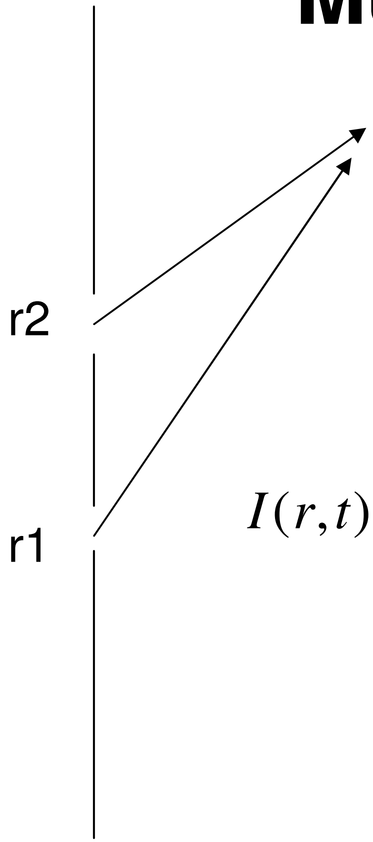
$$\frac{\lambda}{2\xi_T} = \sin \Delta\theta \approx \Delta\theta$$

$$\frac{D}{R} = \tan \Delta\theta \approx \Delta\theta$$

$$\xi_T \approx \frac{\lambda}{2}\frac{R}{D}$$

transverse coherence depends on distance and source size

Mutual Coherence Function



$$V(r,t) = A_1 V(r_1,t) + A_2 V(r_2,t+\tau)$$ Field amplitude

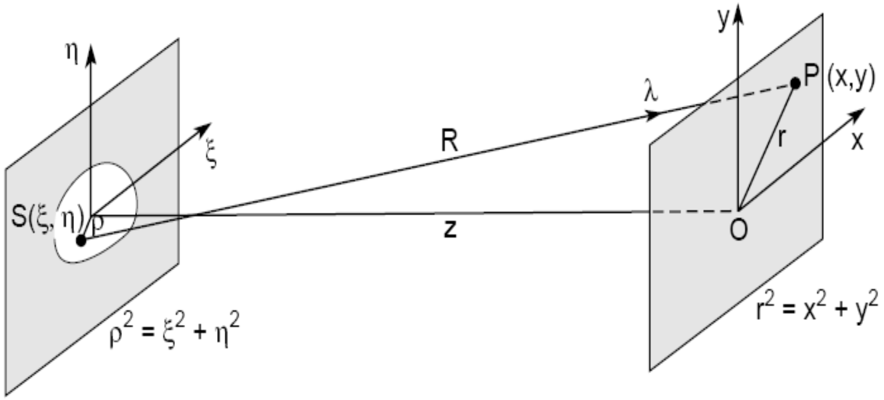
$$I(r,t) = A_1^2 \left|V(r_1,t)\right|^2 + A_2^2 \left|V(r_2,t+\tau)\right|^2 + 2\,\mathrm{Re}(A_1^* A_2 \Gamma(r_1,r_2,\tau))$$ Intensity

$$\Gamma(r_1,r_2,\tau) = < V^*(r_1,t) V(r_2,t+\tau) >$$ Mutual Coherence Function (MCF)

$$\mu(r_1,r_2,\tau) = \frac{< V^*(r_1,t) V(r_2,t+\tau) >}{\sqrt{I(r_1,t) I(r_2,t)}}$$ Complex degree of coherence

$$\left|\mu\right| = \mathrm{Visibility}$$

van Cittert – Zernike Theorem



$$p = \frac{X}{R}, \quad q = \frac{Y}{R}, \quad \psi = \frac{k(X^2 + Y^2)}{R}$$

$$\theta = \frac{r}{z}$$

complex degree of coherence

$$\mu(0,P) = \frac{e^{-i\psi} \iint_S I(\xi,\eta) e^{ik(p\xi + q\eta)} d\xi d\eta}{\iint_S I(\xi,\eta) e^{ik(p\xi + q\eta)} d\xi d\eta}$$

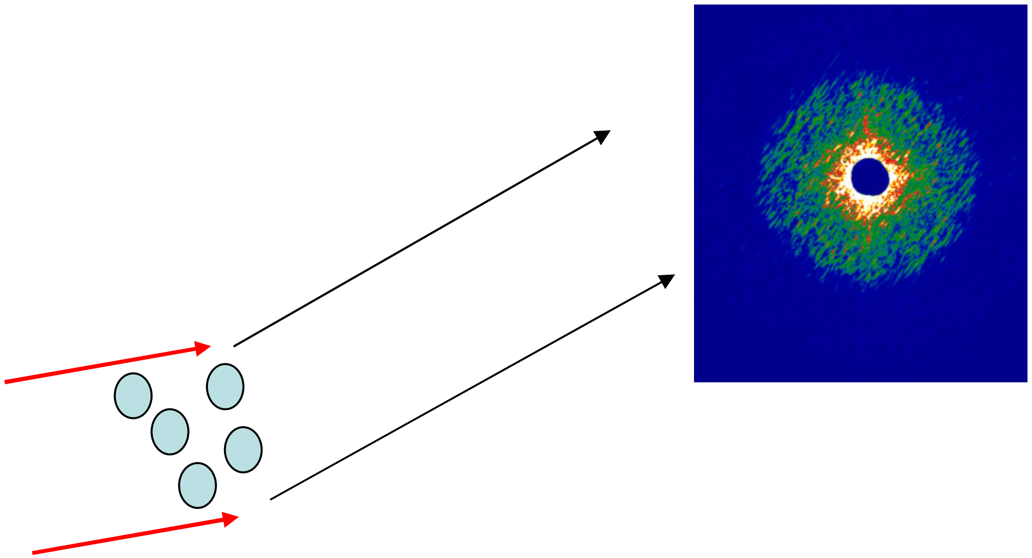
Fourier Transform of the source intensity distribution!

Axial symmetry

$$\mu(0,P) = \frac{e^{-i\psi} \int_0^\infty I(\rho) J_0(k\rho\theta) \rho \, d\rho}{\int_0^\infty I(\rho) \rho \, d\rho}$$

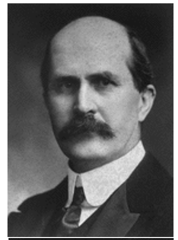
Speckle Pattern

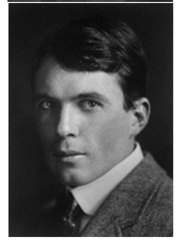
"Everything interferes with everything"

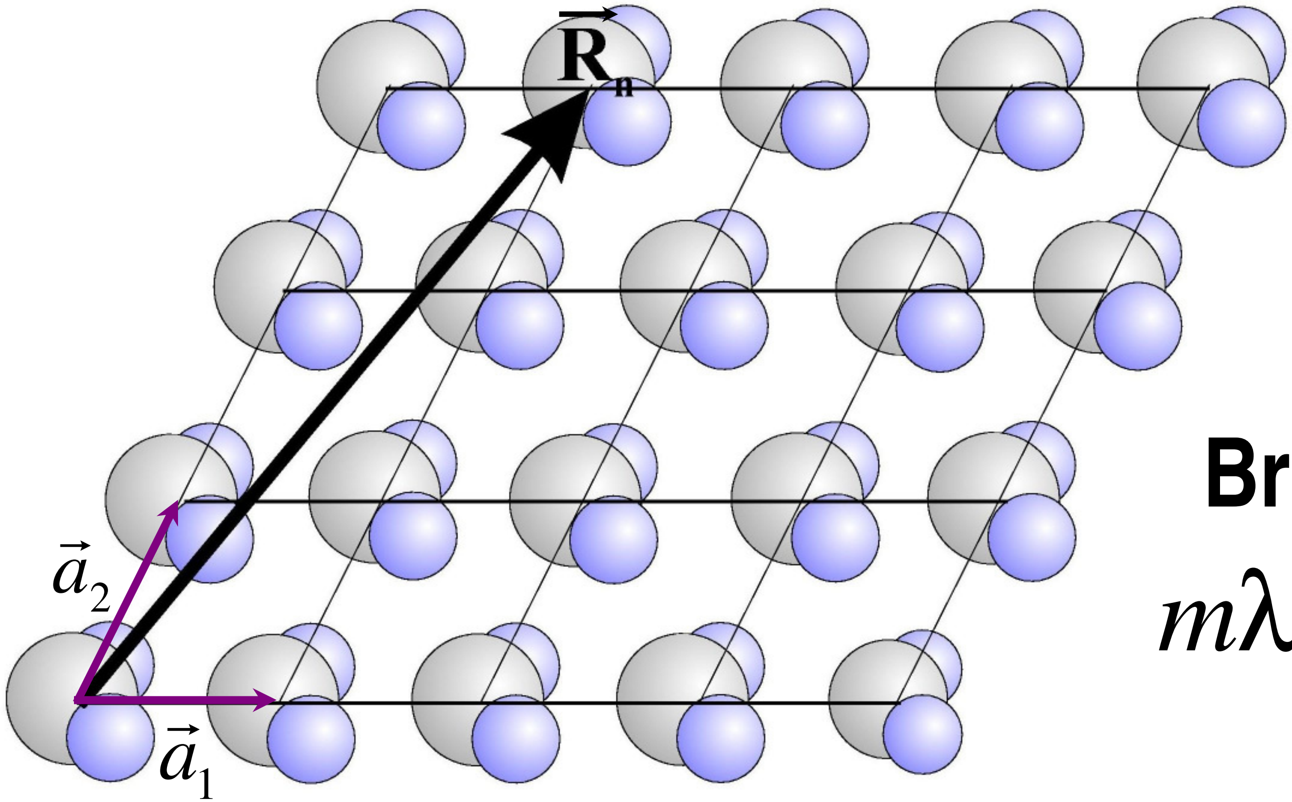


Scattering from a Crystal

William Henry Bragg (1862 - 1942)
William Lawrence Bragg (1890 - 1971)
Nobelpreis 1915







d

Bragg's Law:

$$m\lambda = 2d\sin\theta$$

$$F_{\text{crystal}}(\vec{q}) = \left(\sum_{j=1}^{N} f_j(\vec{q})\, e^{i\,\vec{q}\cdot\vec{r}_j} \right) \cdot \left(\sum_{n=1}^{M} e^{i\,\vec{q}\cdot\vec{R}_n} \right)$$

Unit Cell Structure Factor **Lattice Sum**

$F_{uc}(\vec{q})$

Elastic Scattering from a Crystal

Differential Scattering Cross Section

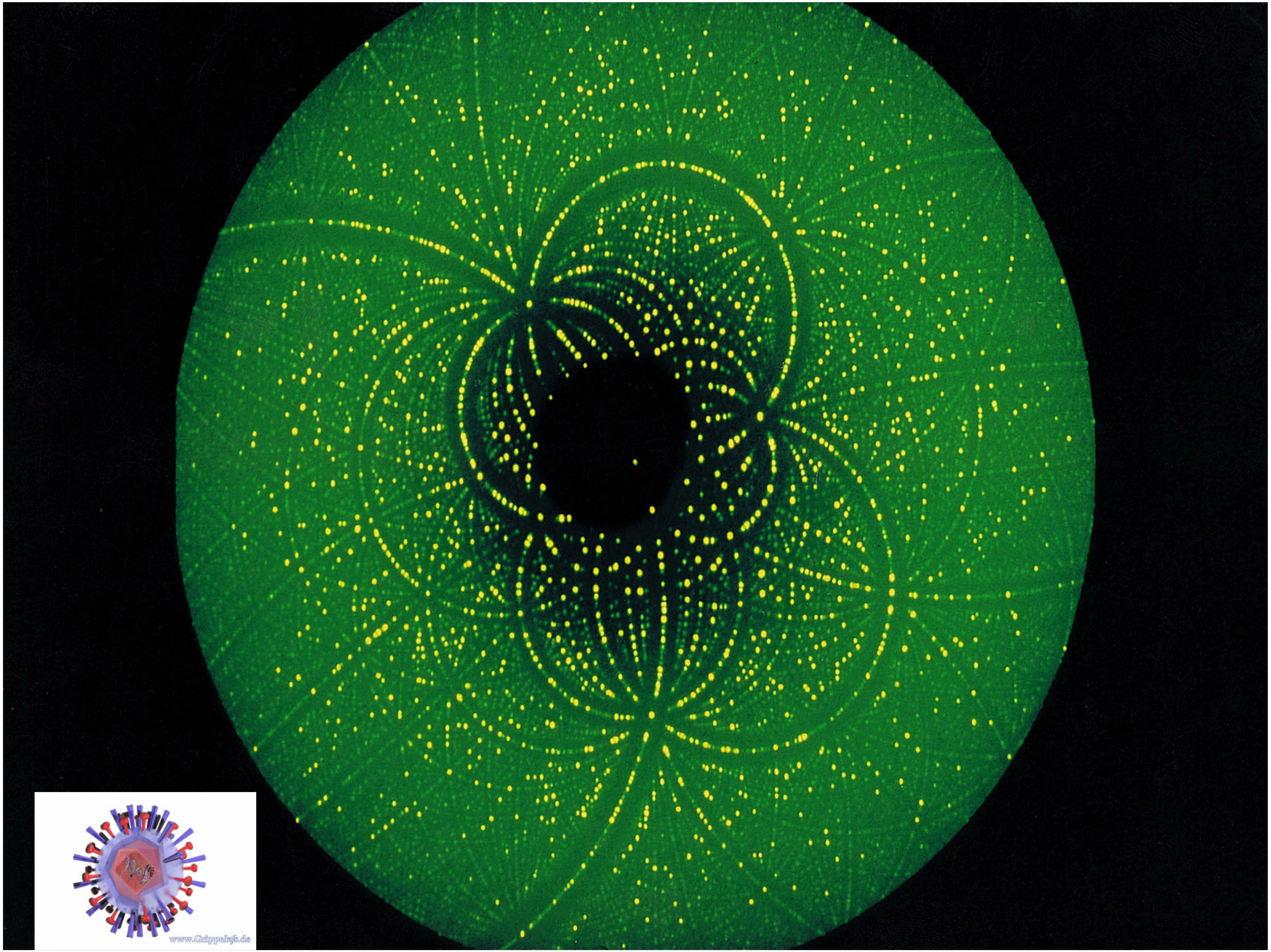
$$\frac{d\sigma}{d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_0 S(\vec{q})$$

Intrinsic Cross Section
Coupling Beam ⇔ Sample

Properties of the Sample without Beam

$$S(\vec{q}) = \left|F_{\text{crystal}}(\vec{q})\right|^2$$

„Phase problem“


www.Grippeinfo.de

Solution to the phase problem for periodic objects
classical crystallography

- direct methods (using the fact that the density is real and positive)
- anomalous X-ray scattering (MAD)
- heavy atoms
- ...

+ atomic resolution
- need for crystals
- x-ray damage

Structure determination of non-periodic objects
a zoo of scanning x-ray techniques

- scanning transmission x-ray microscope
- tomography (medical imaging)
- ...

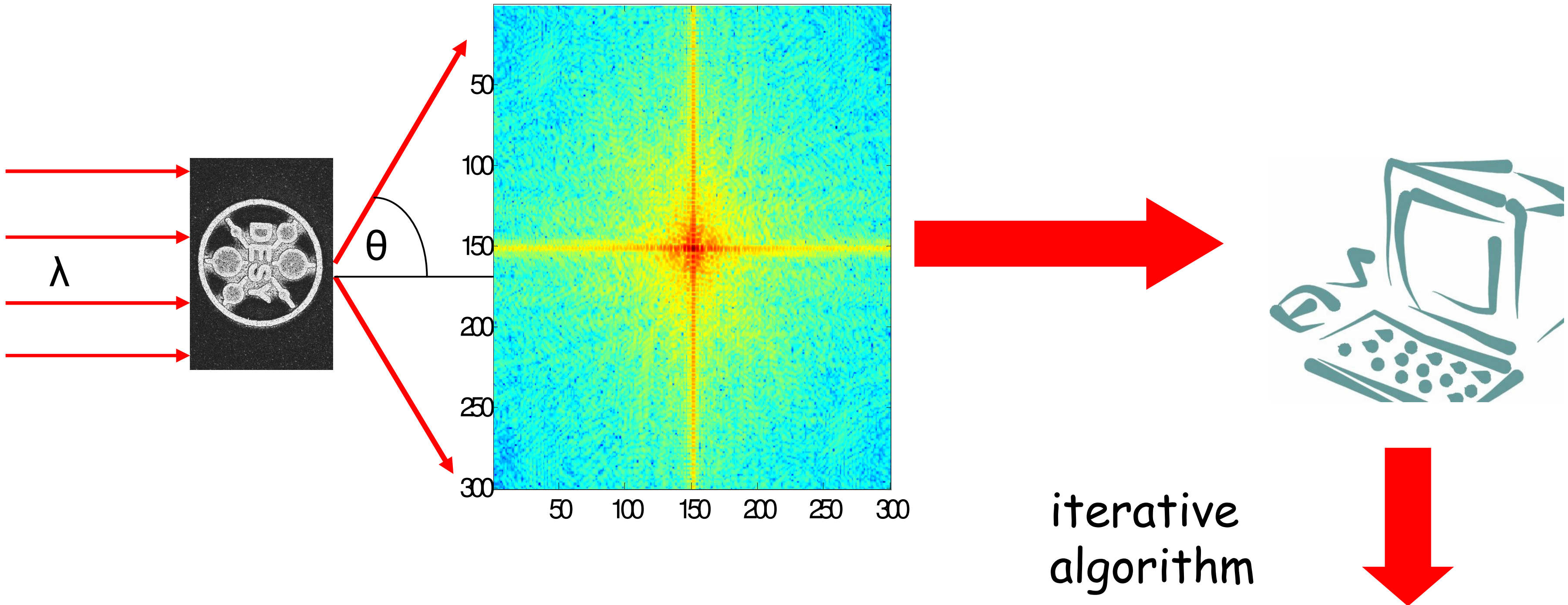
+ no need for crystals
-+ 20-30 nm resolution
- limited dynamics

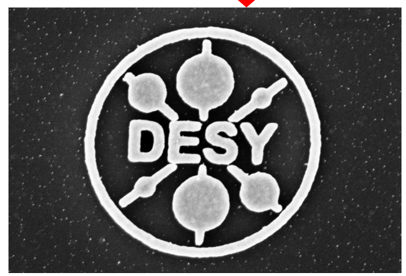
Coherence based techniques for structure determination

Ultrafast (femtoseconds) imaging techniques for non-periodic objects

- **Coherent diffractive imaging**
- **Fourier transform holography**
- **Holographic imaging**
- **Ptychography**
- **and all combinations thereof....**

Structure Determination from Oversampled Speckle Pattern

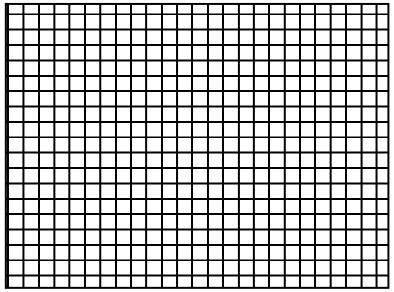




$$\text{max. resolution} \approx \frac{\lambda}{\sin(\theta)}$$

D. Sayre, "Some implications of a theory due to Shannon," Acta Cryst. **5**, 843 (1952).
J. R. Fienup, "Phase retrieval algorithms: a comparison," Appl. Opt. **21**, 2758-2769 (1982).

Phase retrieval and oversampling



M

L

$\rho(x, y, z)$ L x M x N unknown variables

measured quantity |F|^2

sampled at Bragg peak frequency

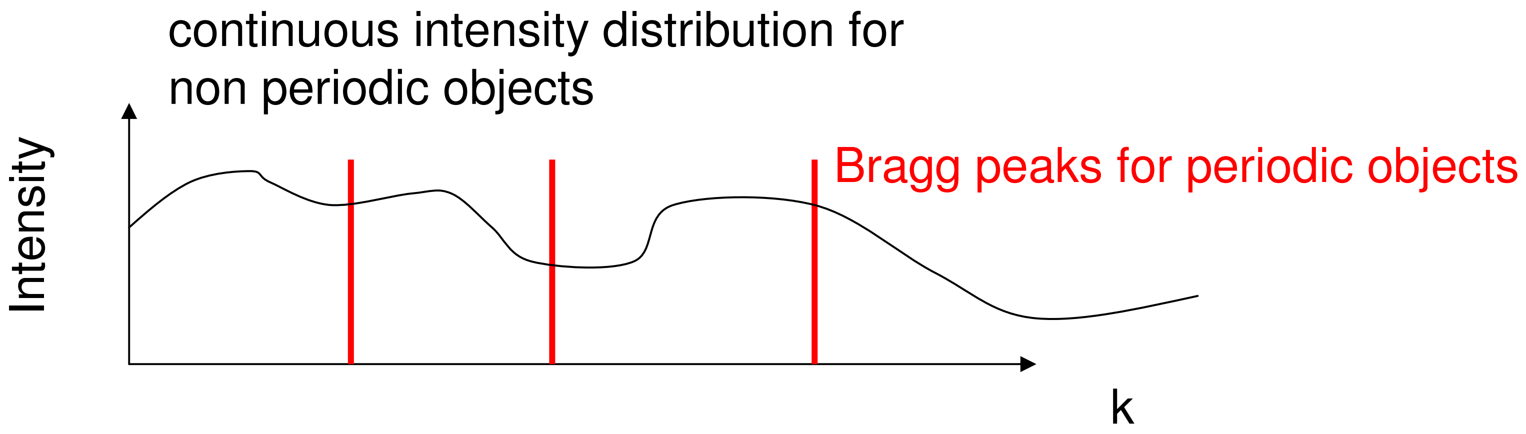
$$|F(k_x, k_y, k_z)| = \left| \sum_{x=0}^{l-1} \sum_{y=0}^{m-1} \sum_{z=0}^{n-1} \rho(x,y,z) e^{2\pi i(k_x x/l + k_y y/m + k_z z/n)} \right|, \quad (1)$$

$$k_x = 0, \ldots, l-1, \quad k_y = 0, \ldots, m-1, \quad k_z = 0, \ldots, n-1$$

Friedel's law

(L x M x N) / 2 independent equations

No inversion possible



Idea: sample k finer than Bragg frequency, e.g. $\sqrt[3]{2}$

$$|F(k_x, k_y, k_z)| = \left| \sum_{x=0}^{l-1} \sum_{y=0}^{m-1} \sum_{z=0}^{n-1} \rho(x,y,z) \times e^{2\pi i [k_x x/(\sqrt[3]{2} l) + k_y y/(\sqrt[3]{2} m) + k_z z/(\sqrt[3]{2} n)]} \right|,$$

$$k_x = 0, \ldots, \sqrt[3]{2} l - 1, \quad k_y = 0, \ldots, \sqrt[3]{2} m - 1,$$

$$k_z = 0, \ldots, \sqrt[3]{2} n - 1.$$

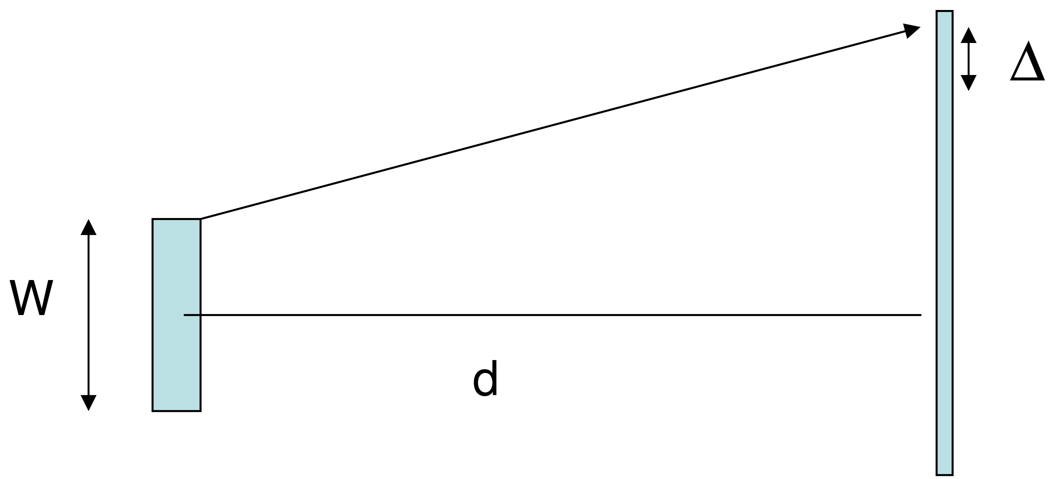
Number of independent equations
= number of unknown variables

$(\sqrt[3]{2})^3$ (L x M x N) / 2 = L x M x N

Shannon's theorem in X-ray scattering

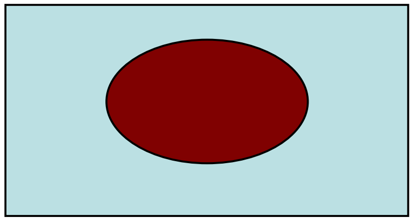
If a diffraction pattern is sampled at spatial frequencies at least twice that corresponding to the size of the sample the phases can be recovered by means of iterative algorithms.

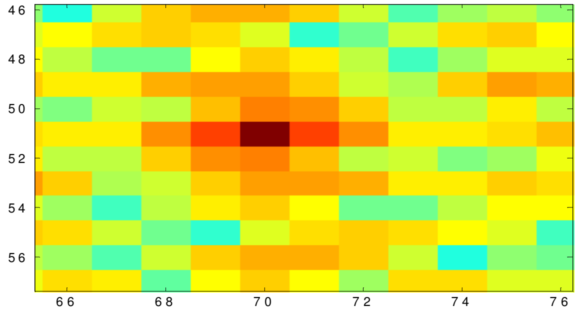
$$\Delta = \frac{1}{2}\frac{\lambda d}{W}$$ sampling in reciprocal space



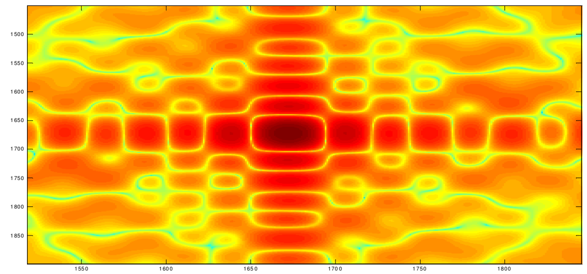
oversampling parameter

$$\sigma = \frac{\text{speckle size}}{\text{pixel size}} = \frac{\lambda d}{WP} \geq 2$$



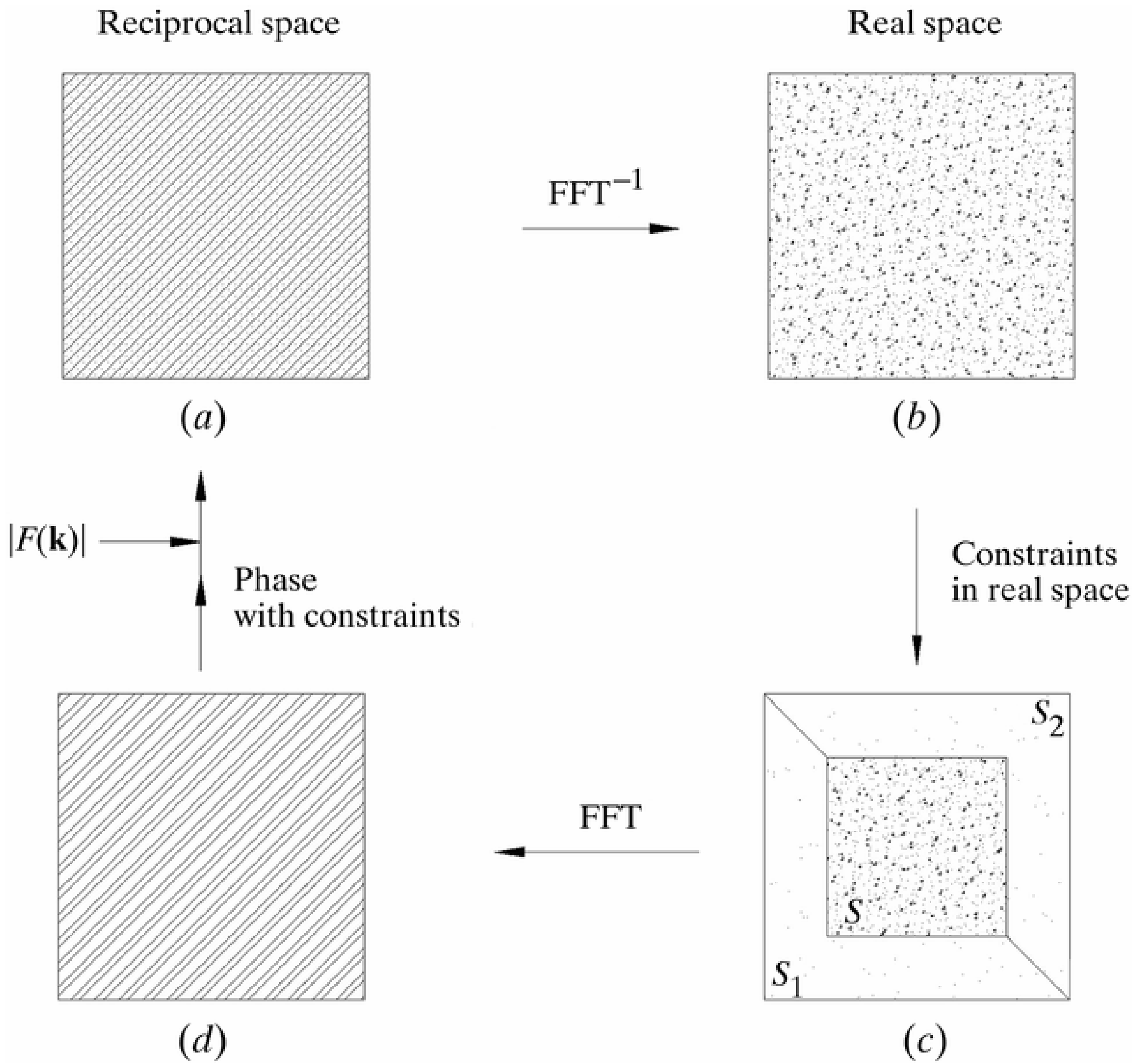


$\sigma = 1$



$\sigma = 770$

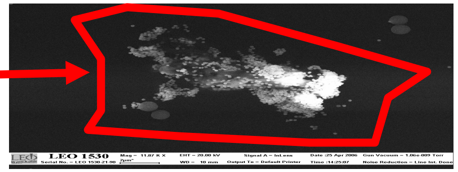
The iterative algorithm due to Gerchberg-Saxton-Fienup



The hybrid-input-output (HIO) algorithm

get some a priori knowledge about the support
i.e. shape of your object

area inside support S



0. Add random phases to the measured amplitudes $|F(\boldsymbol{k})|$ (square root of the measured intensities), which gives $G_1(\boldsymbol{k})$.

1. Substitute the amplitudes with the measured ones ($\rightarrow G_1'(\boldsymbol{k})$).

2. Fourier transform into real space ($\rightarrow g_1'(\boldsymbol{x})$).

3. Set negative pixels[a] or pixels that lie outside of the support to zero ($\rightarrow g_2(\boldsymbol{x})$).

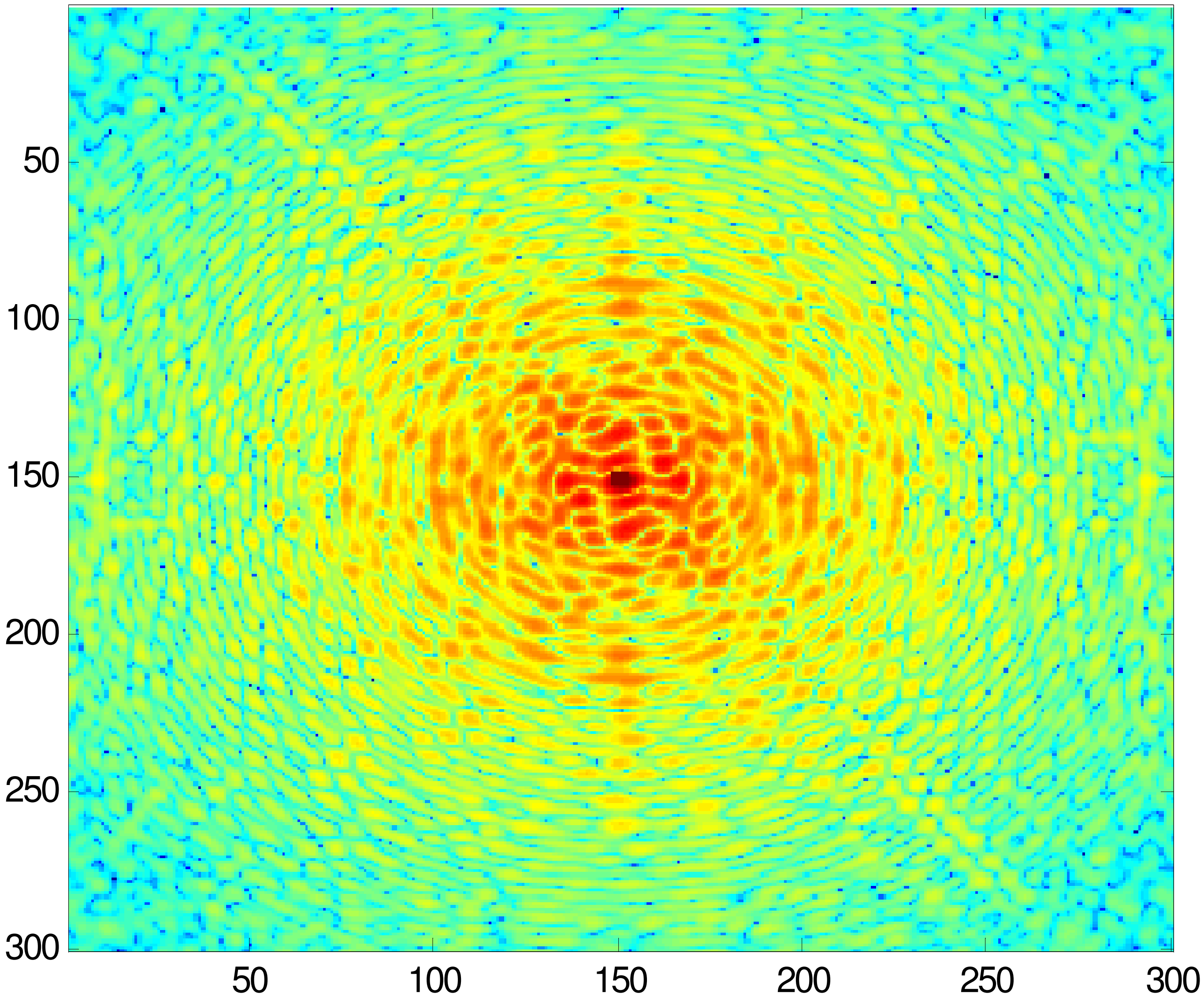
4. Fourier transform back into reciprocal space ($\rightarrow G_2(\boldsymbol{k})$).

$$g_{n+1}(\boldsymbol{x}) = \begin{cases} g_n'(\boldsymbol{x}), & \text{x in support} \\ g_n(\boldsymbol{x}) - \beta_{\mathrm{HIO}} g_n'(\boldsymbol{x}), & \text{x not in support} \end{cases}$$

$$0 < \beta_{HIO} < 1$$

measure of convergence

$$e_{\boldsymbol{k}}^{(n)} = \sqrt{\frac{\sum_{\boldsymbol{k}} \left(|G_n(\boldsymbol{k})| - |F(\boldsymbol{k})|\right)^2}{\sum_{\boldsymbol{k}} |F(\boldsymbol{k})|^2}}.$$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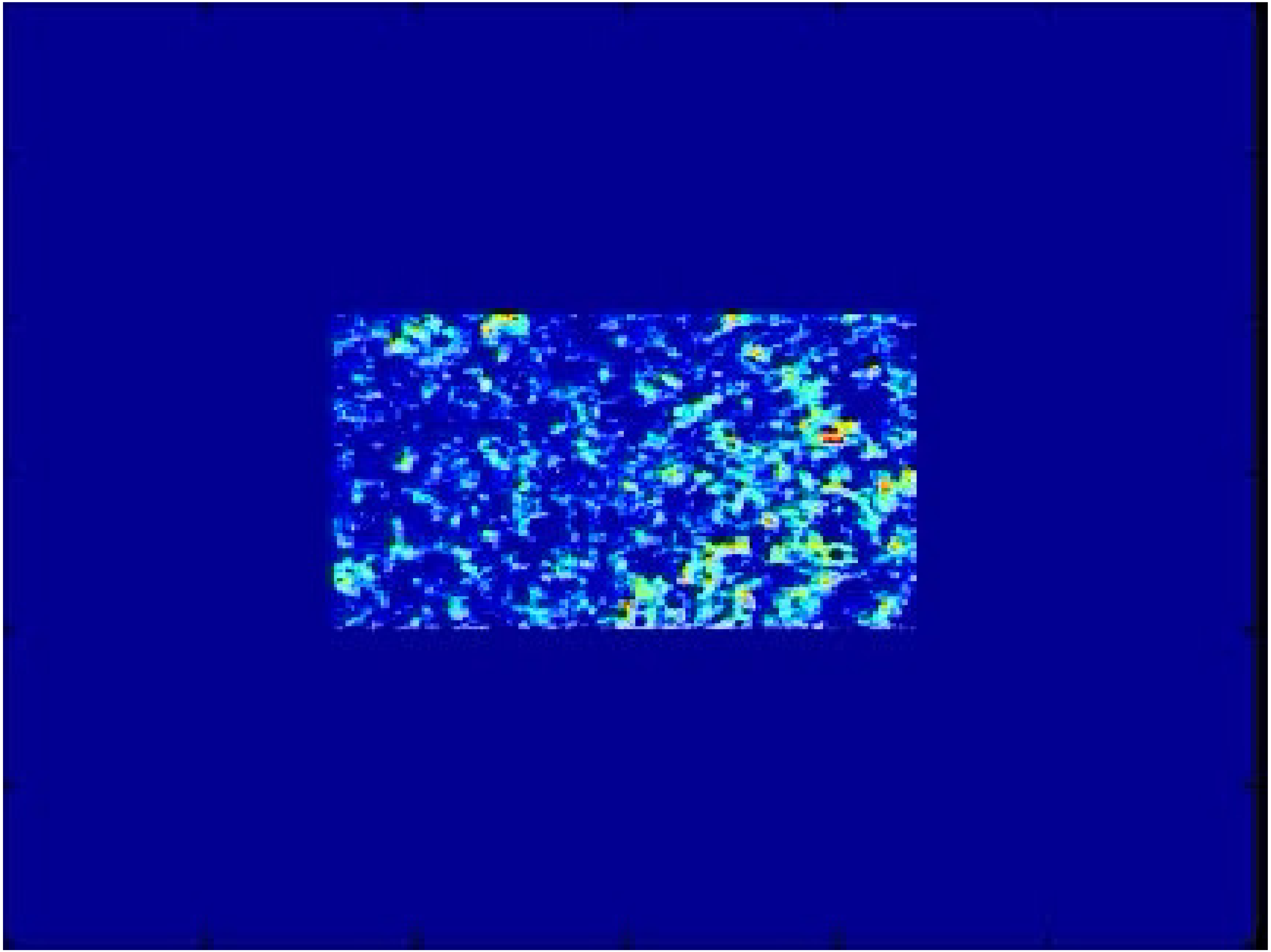
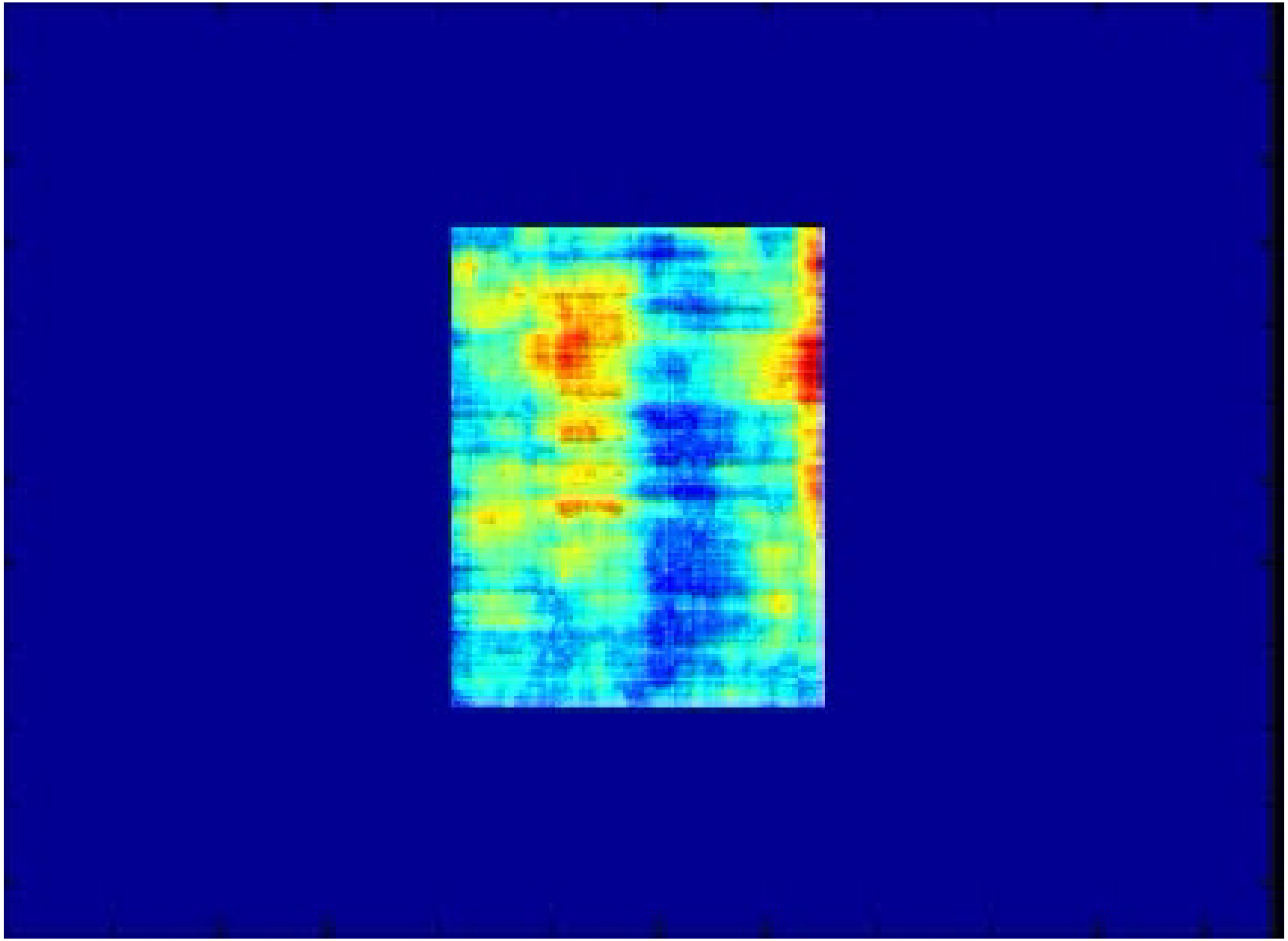
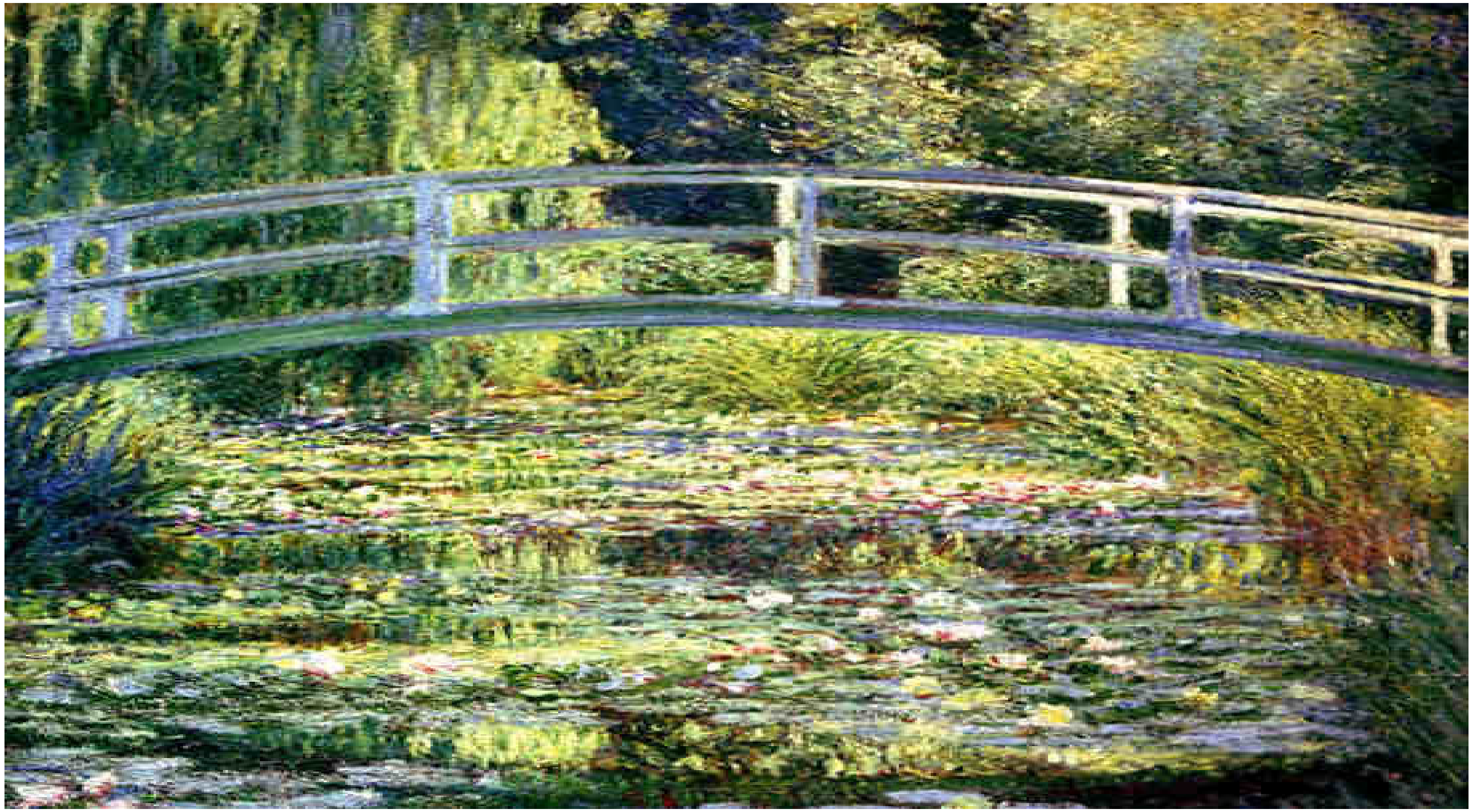


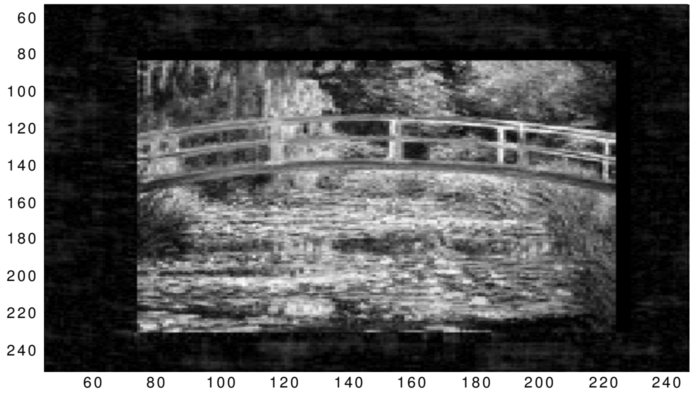


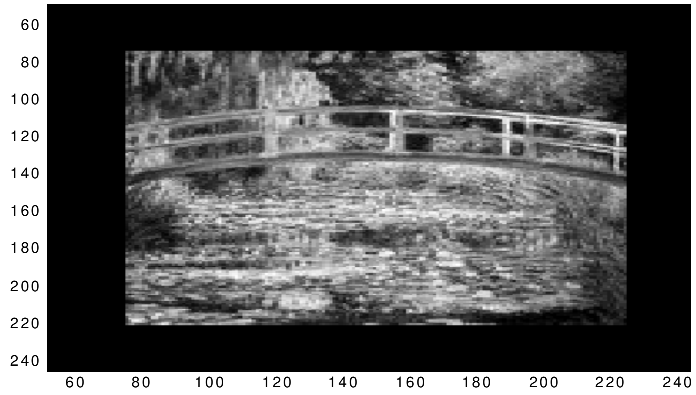
The ‘Object’

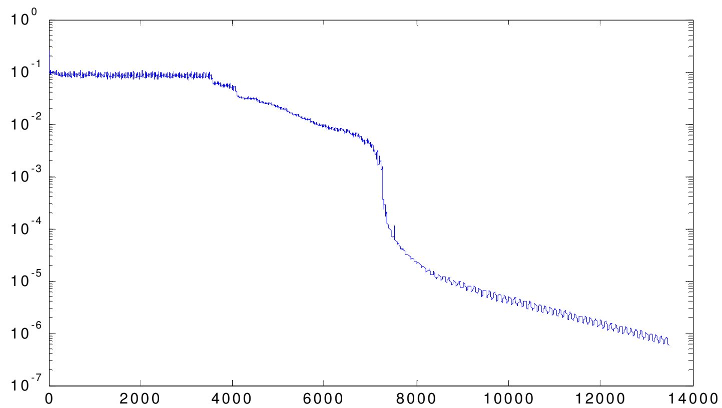


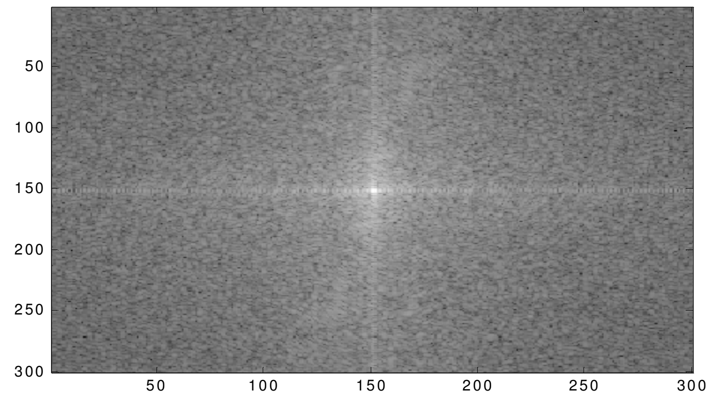
Claude Monet, Seerosenteich II 1899

and its reconstruction...

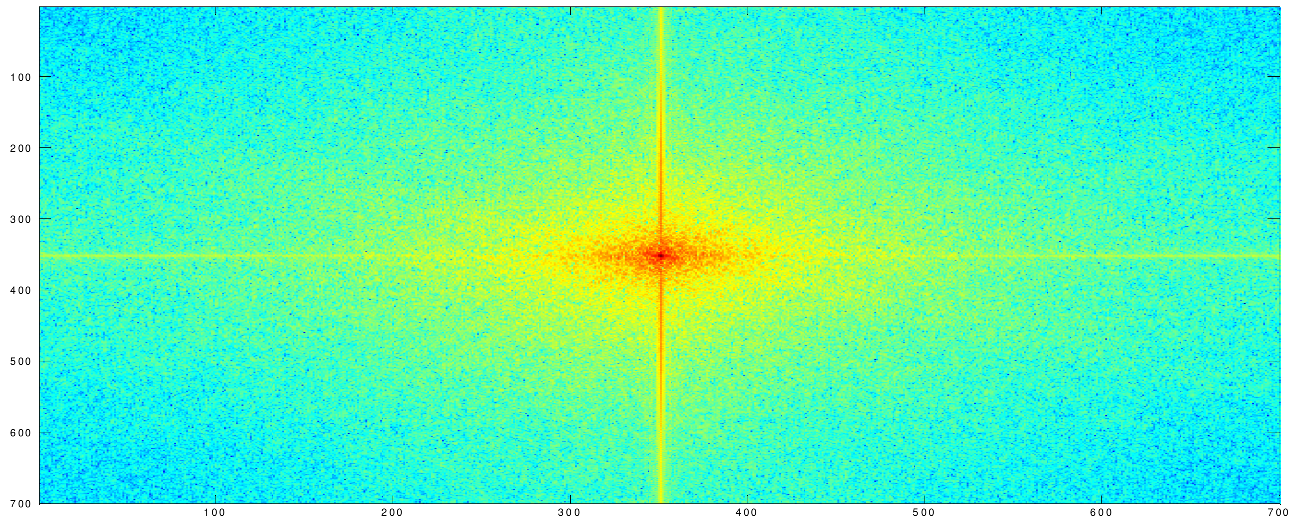


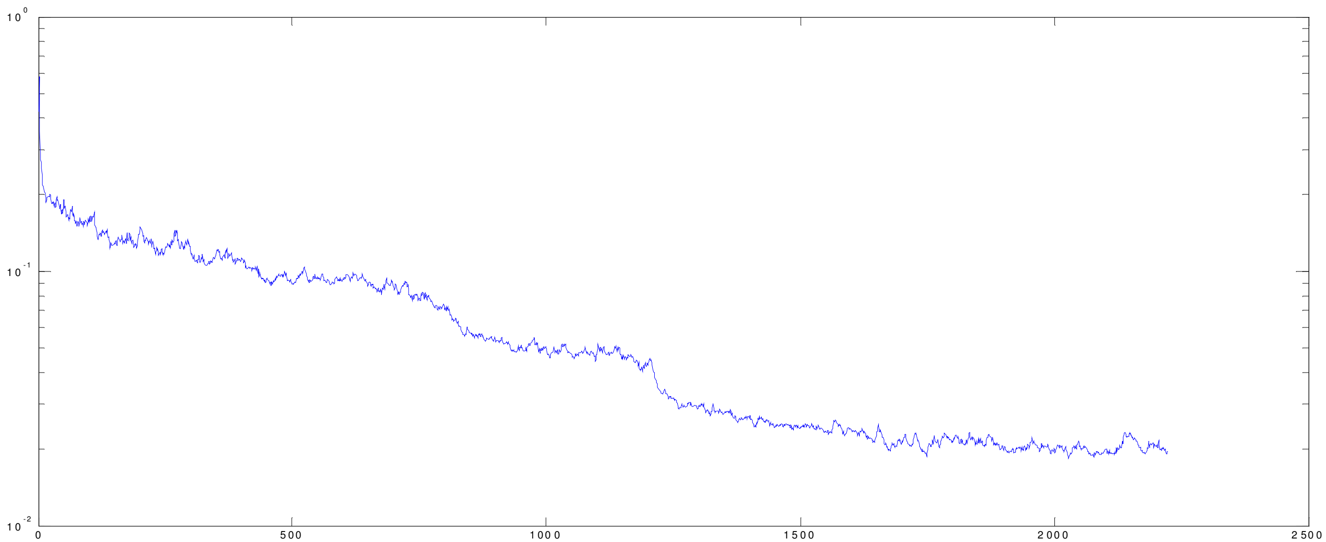






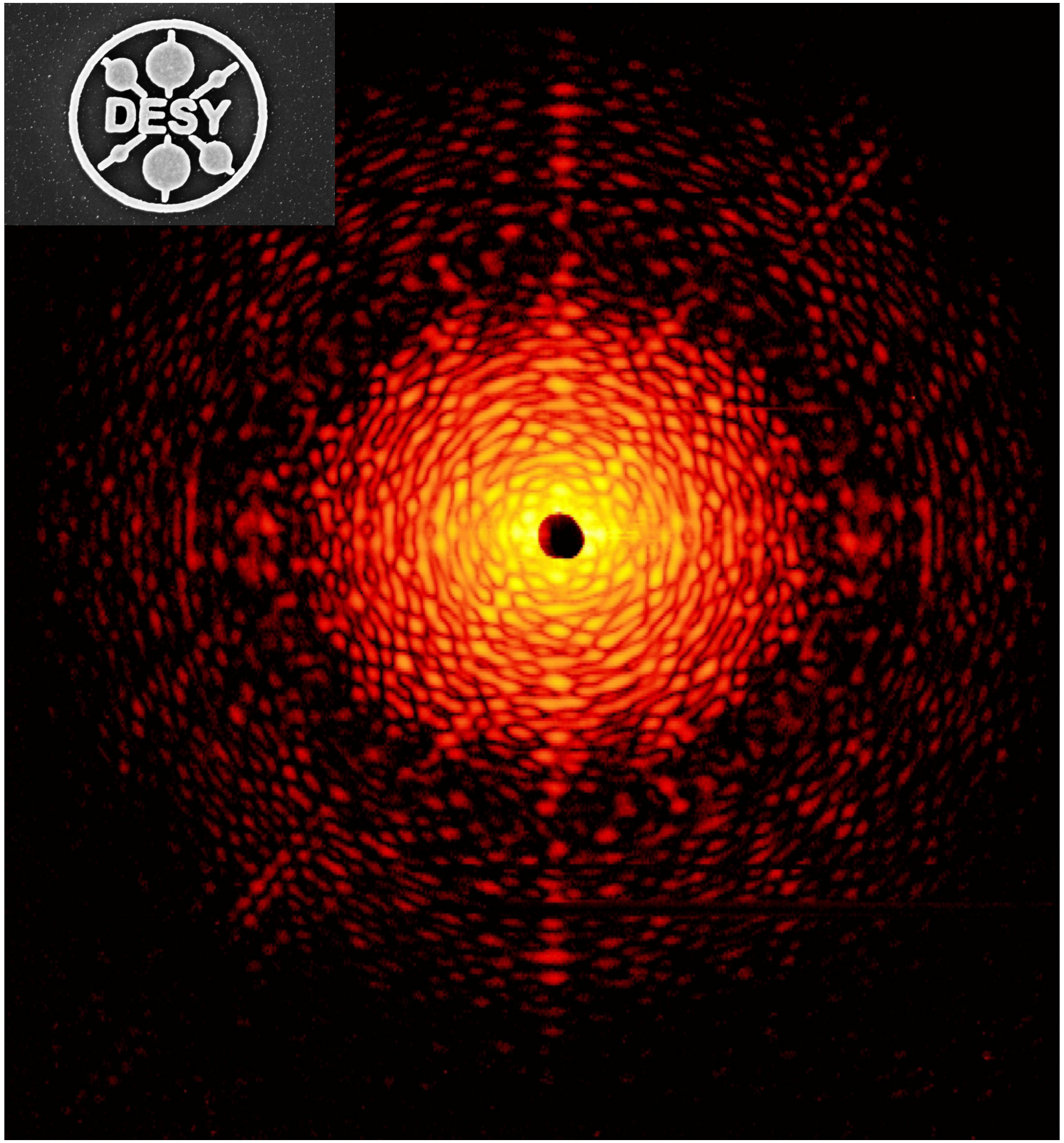
an unknown object





and it’s reconstruction

Experiment using 8 keV Photonen

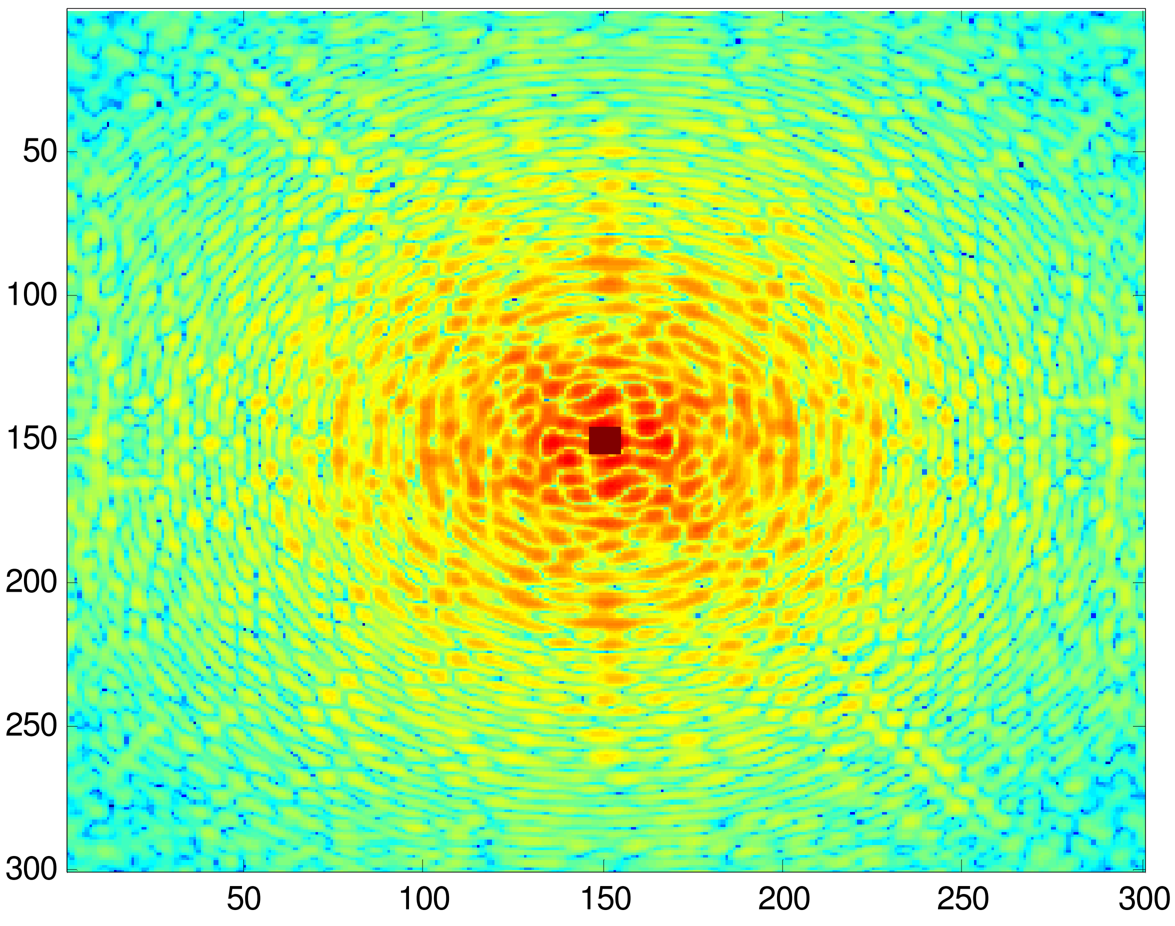


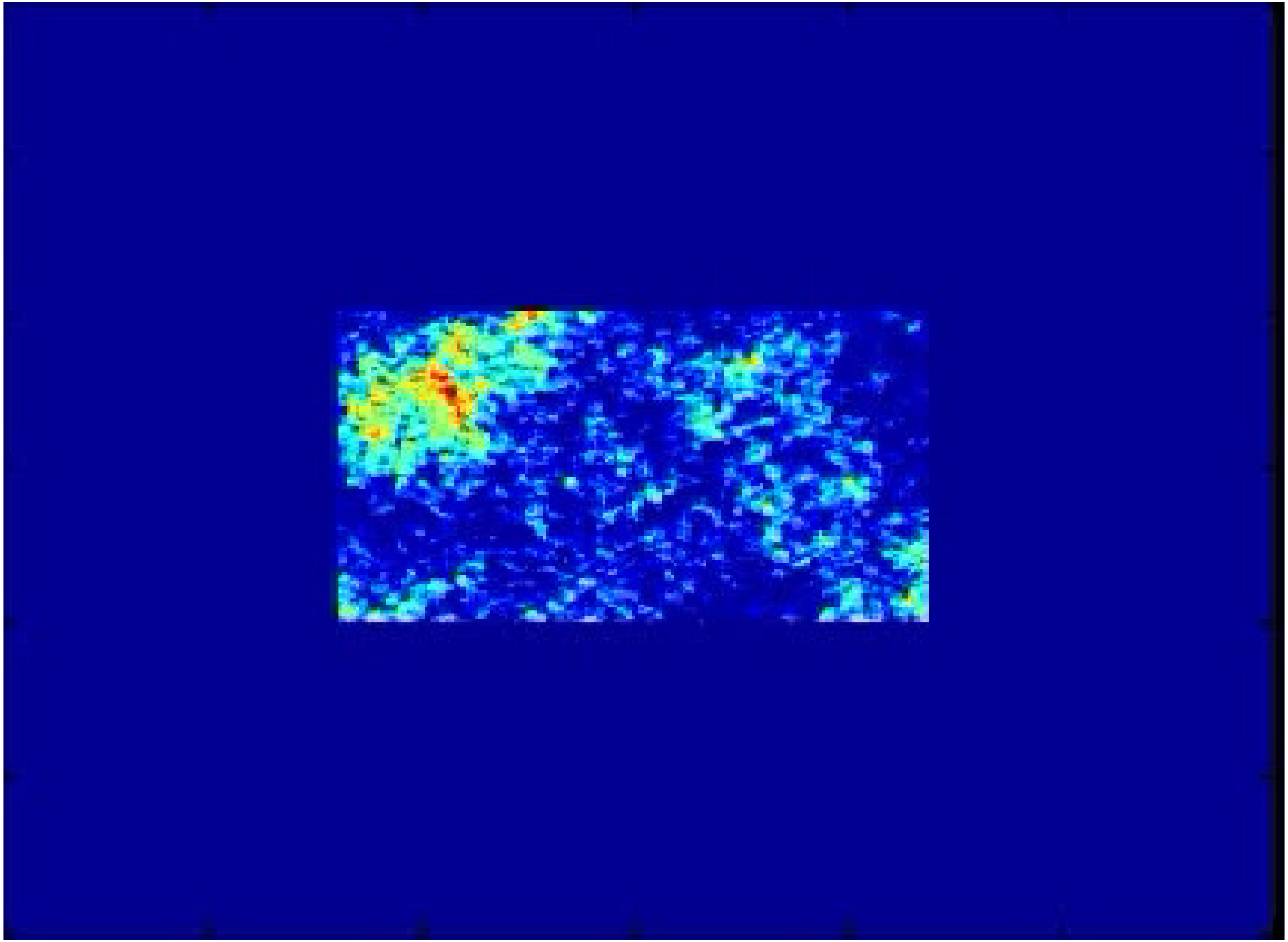
2 microns

Resolution 30 nm

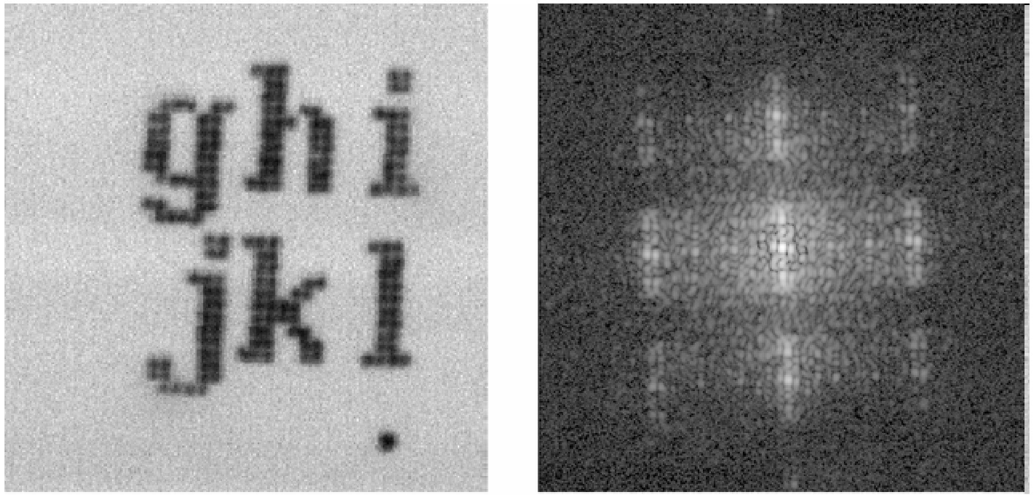
The beamstop problem

Missing Data 1



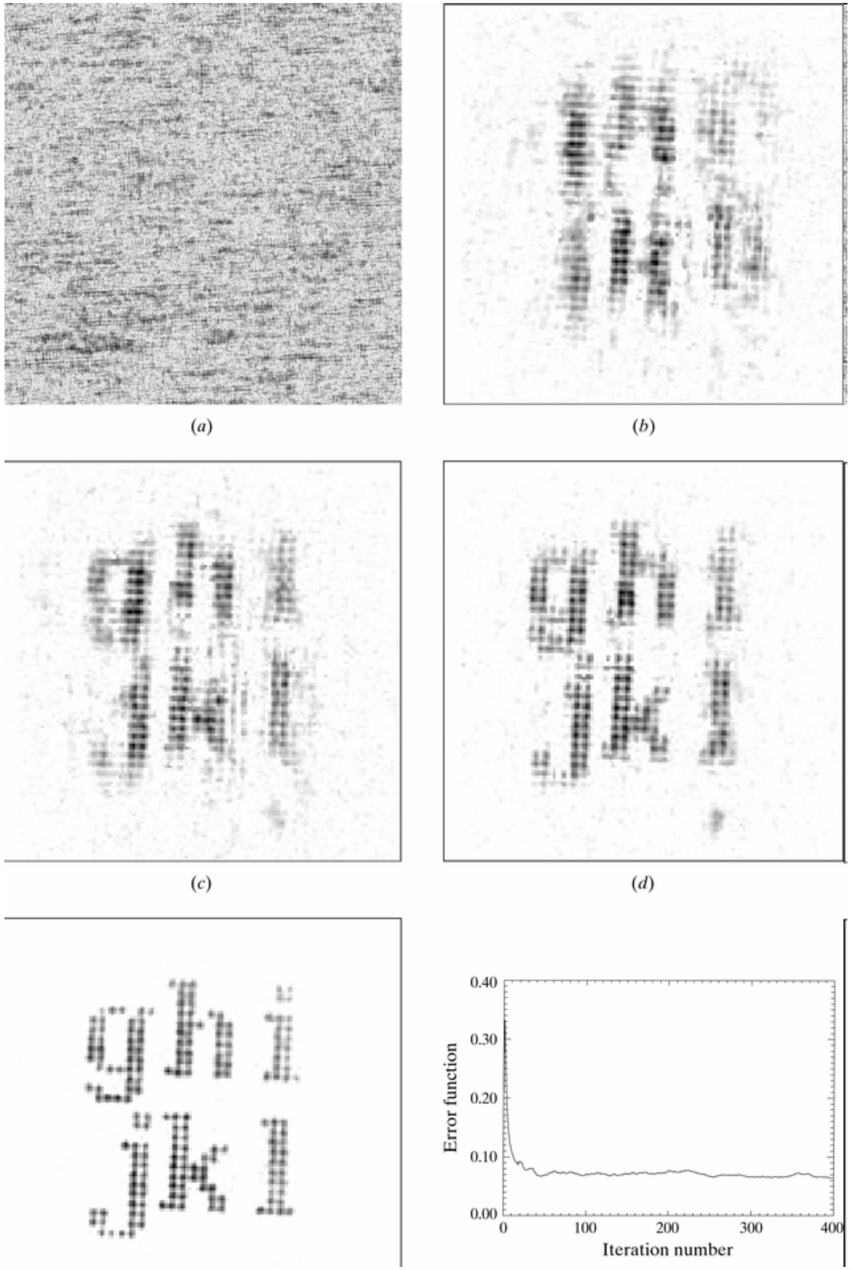


First experimental realization at a synchrotron source



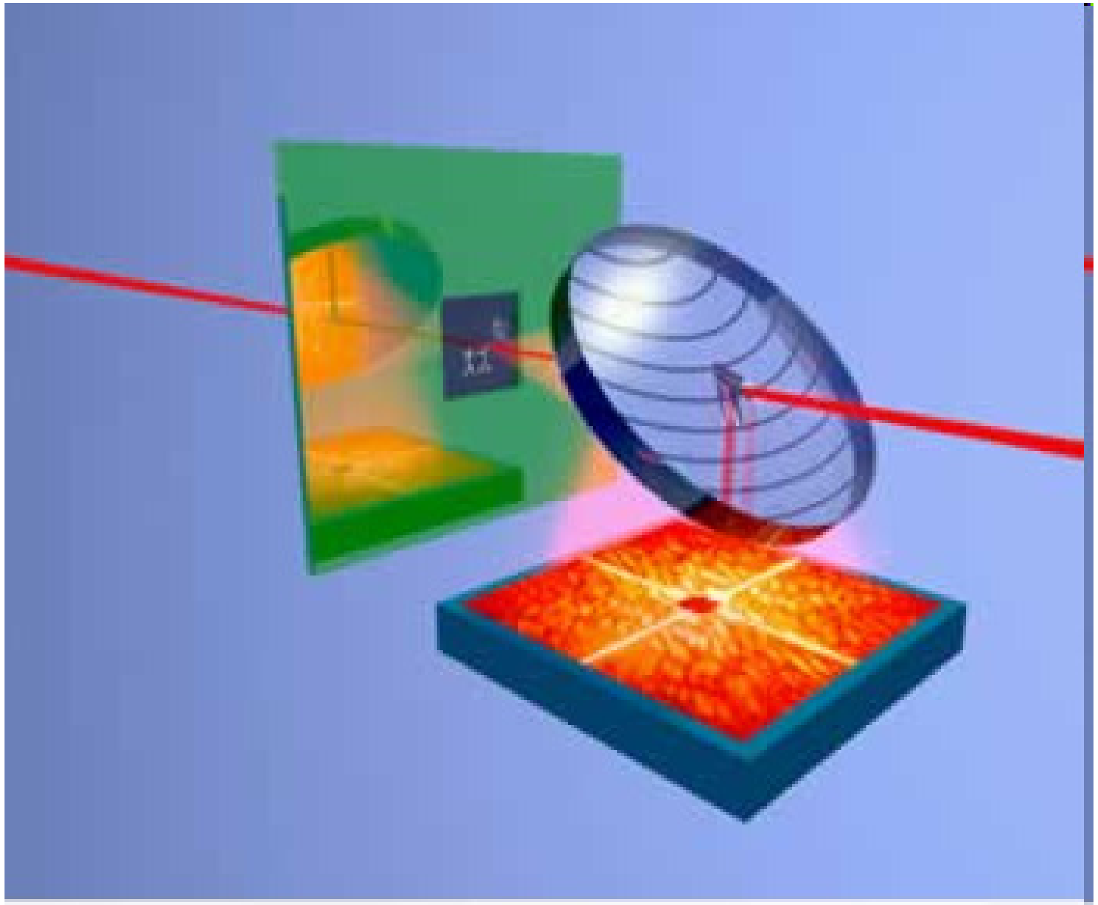
(a) (b)

J. Miao, P. Charalambous, J. Kirz, D. Sayre, "Extending the methodology of x-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens," Nature **400**, 342-344 (1999).

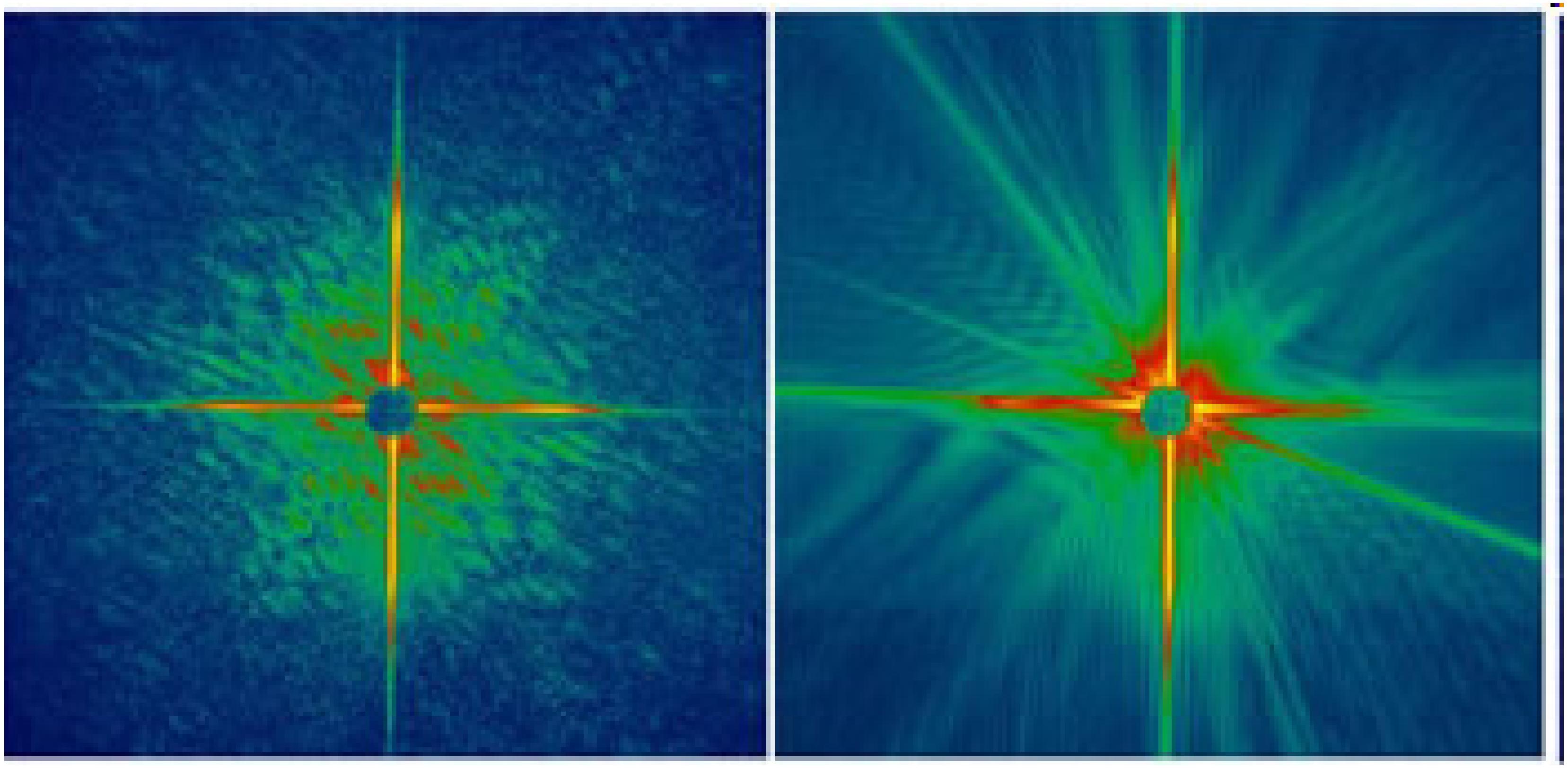


J. Miao, P. Charalambous, J. Kirz, D. Sayre, "Extending the methodology of x-ray crystallography to allow imaging of micrometre-sized non-crystalline specimens," Nature **400**, 342-344 (1999).

First experimental realization at an FEL source



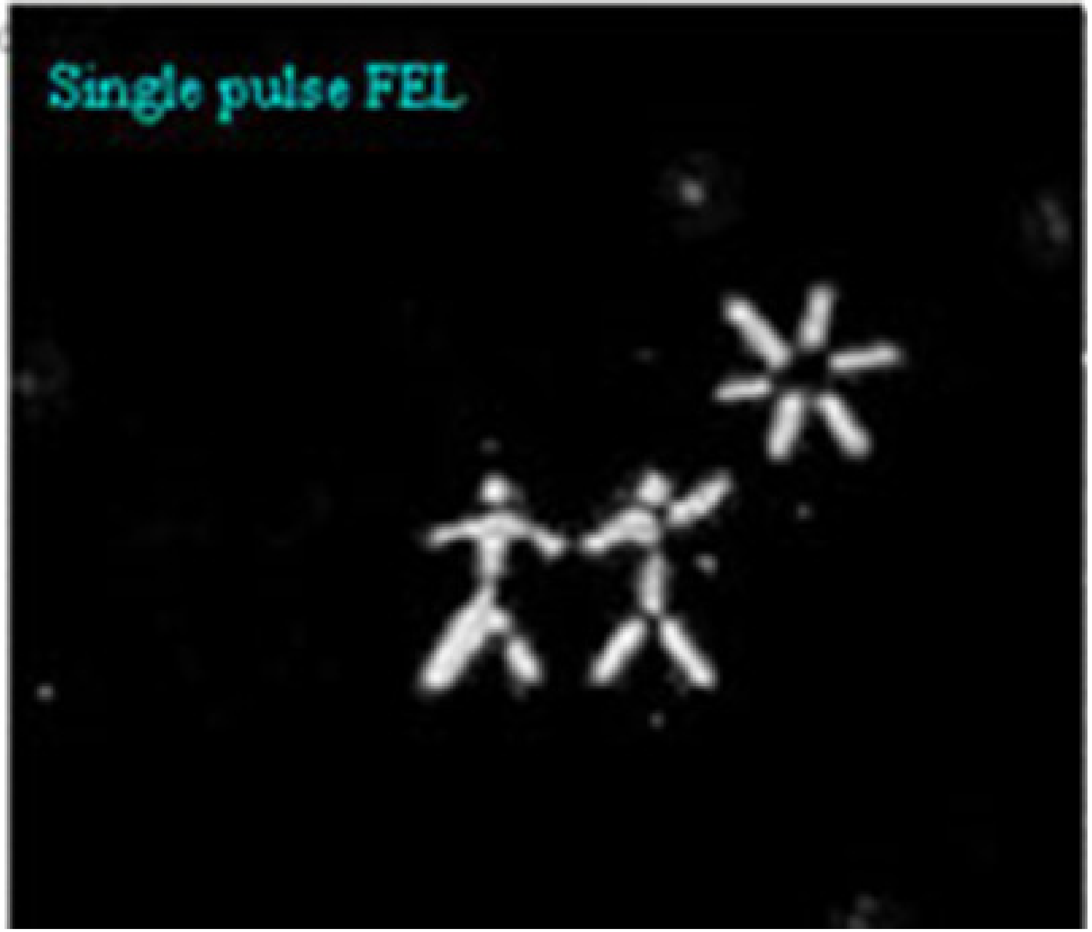
H. Chapman et al. Nature Physics 2, 839 (2006)



pulse #1 pulse #2

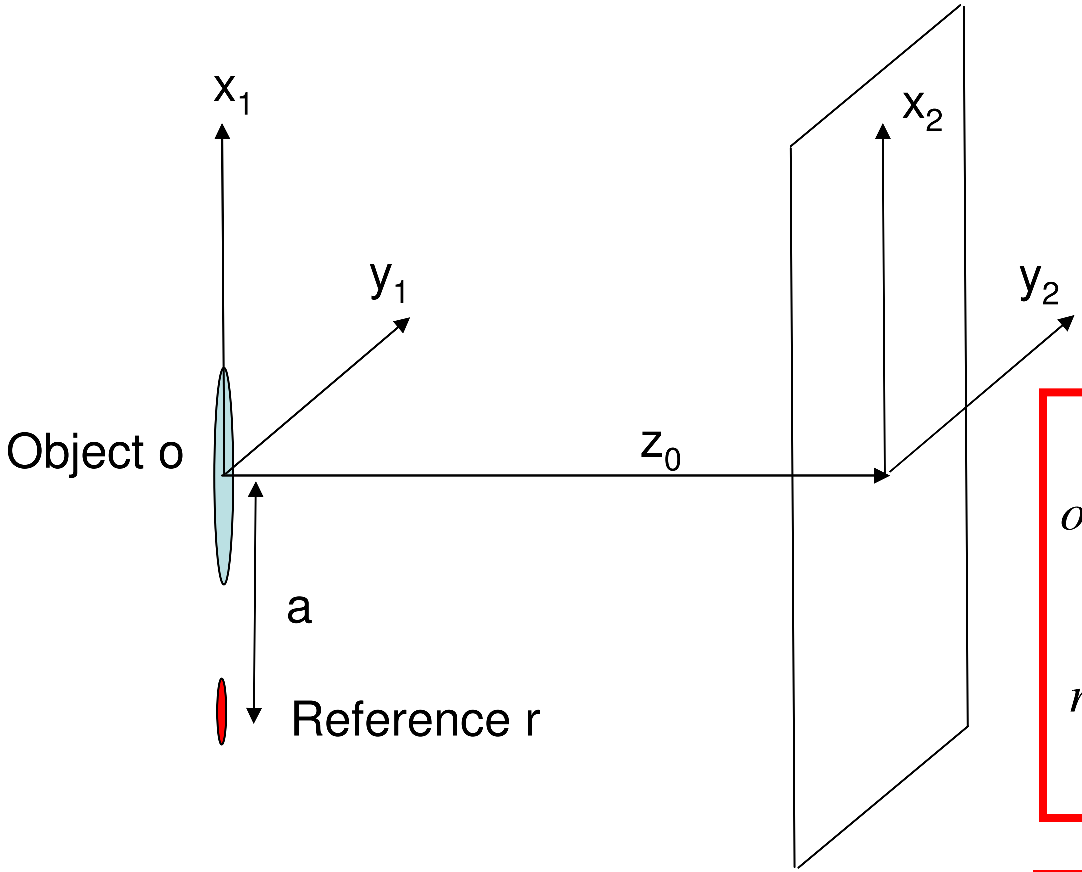
H. Chapman et al. Nature Physics 2, 839 (2006)

Reconstruction



H. Chapman et al. Nature Physics 2, 839 (2006)

Fourier Transform Holography



Fresnel-Kirchhoff Theory

$$o(x_2, y_2) = \frac{i}{\lambda z_o} e^{i\frac{\pi}{\lambda z_0}(x_2{}^2 + y_2{}^2)} \hat{O}(\xi, \eta)$$

$$r(x_2, y_2) = \frac{i}{\lambda z_0} e^{i\frac{\pi}{\lambda z_0}(x_2{}^2 + y_2{}^2)} \hat{R}(\xi, \eta) e^{-2i\pi\xi a}$$

$$\xi = \frac{x_2}{\lambda z_0}$$

$$\eta = \frac{y_2}{\lambda z_0}$$

$$\hat{O}(\xi, \eta) = FT[o(x_1, y_1)]$$

$$\hat{R}(\xi, \eta) = FT[r(x_1, y_1)]$$

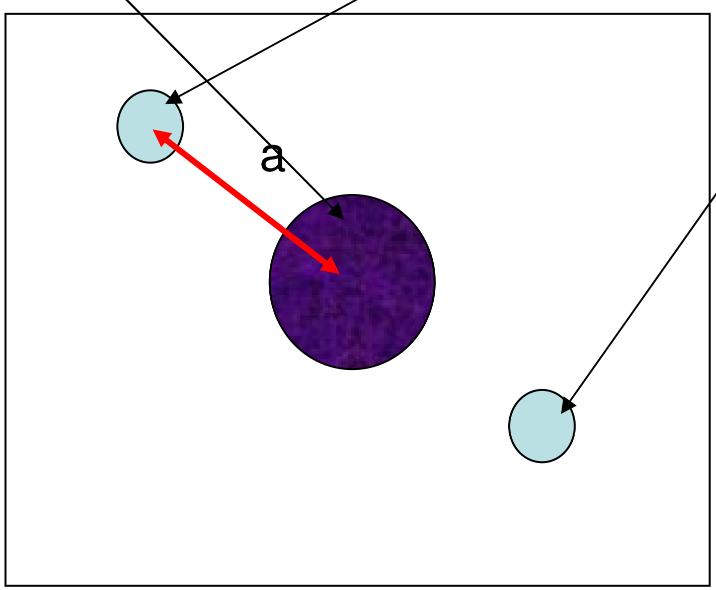
$$\hat{O}^*(\xi,\eta) = \hat{O}(-\xi,-\eta)$$

$$I(x_2, y_2) = | r(x_2, y_2) + o(x_2, y_2) |^2$$

$$I(x_2, y_2) = | r(x_2, y_2) |^2 + | o(x_2, y_2) |^2 + r^*(x_2, y_2) o(x_2, y_2) + r(x_2, y_2) o^*(x_2, y_2)$$

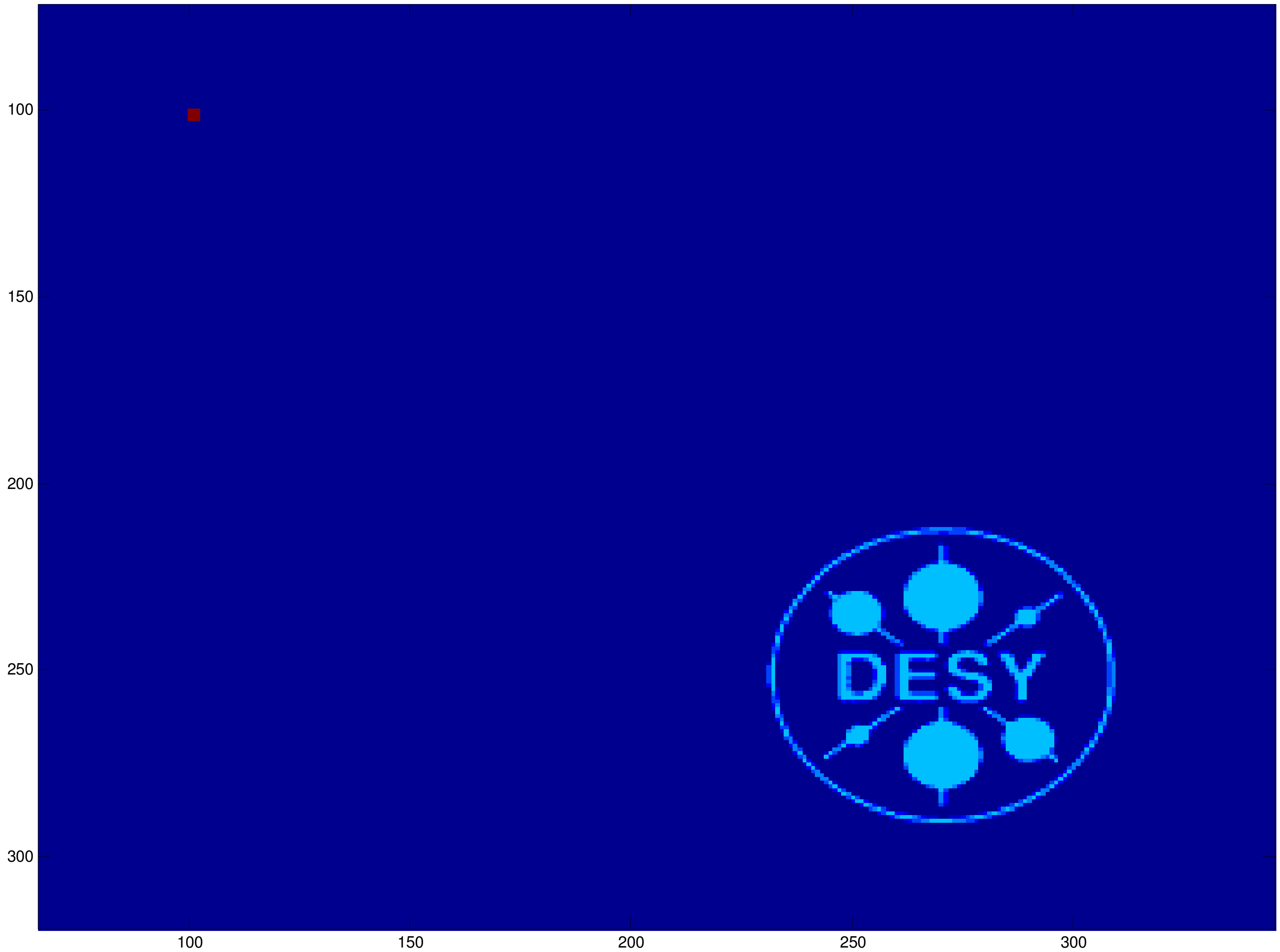
$$I(x_2, y_2) \propto | \hat{R}(\xi,\eta) |^2 + | \hat{O}(\xi,\eta) |^2 + \hat{R}^*(\xi,\eta)\hat{O}(\xi,\eta) e^{i\pi a\xi} + \hat{R}(\xi,\eta)\hat{O}^*(\xi,\eta) e^{-i\pi a\xi}$$

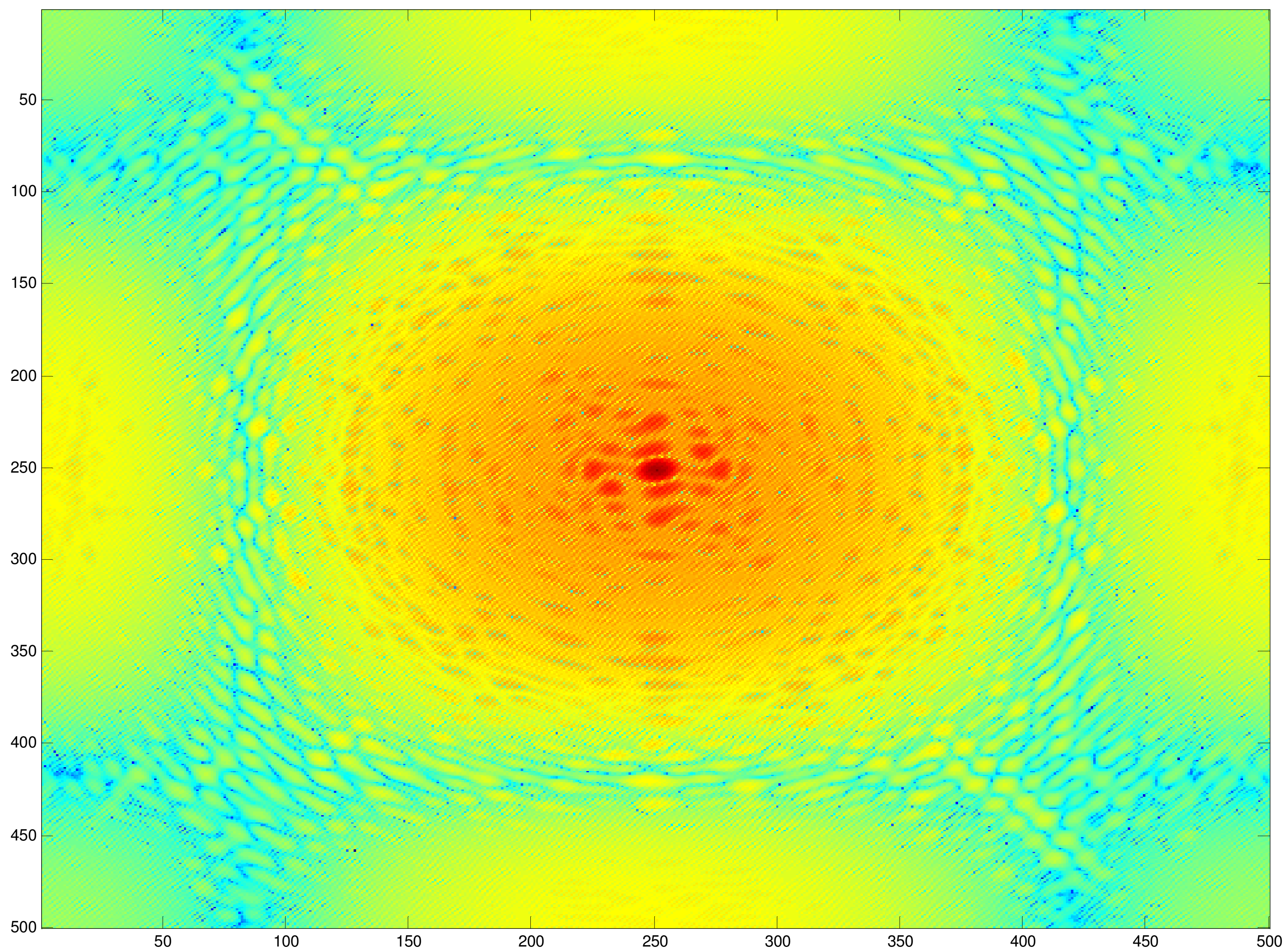
$$FT[r^*(-x,-y) \otimes o(x+a, y)] + FT[r(x, y) \otimes o(-x-a,-y)]$$



Convolution with the reference objects limits the resolution

Small reference hole





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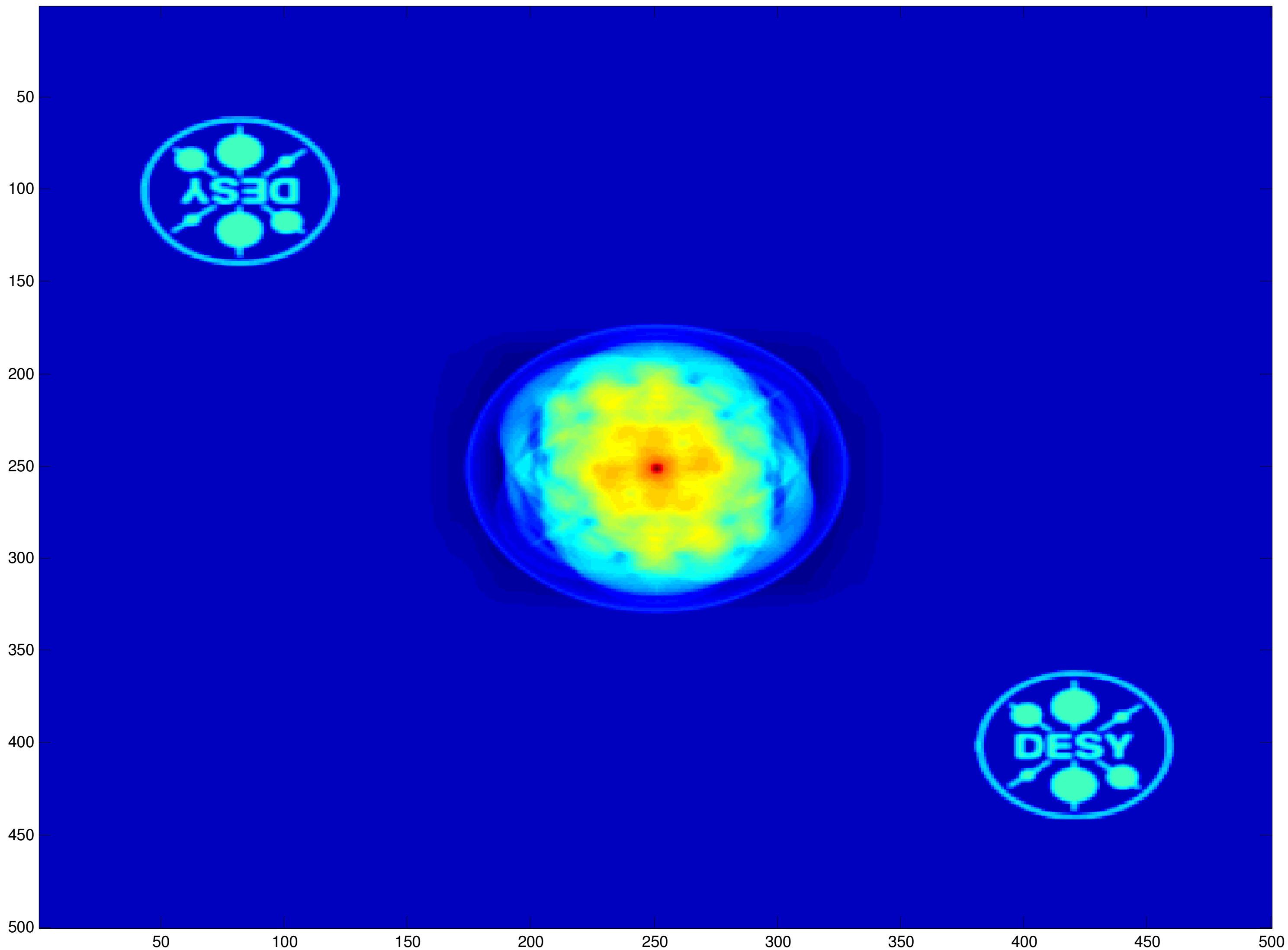
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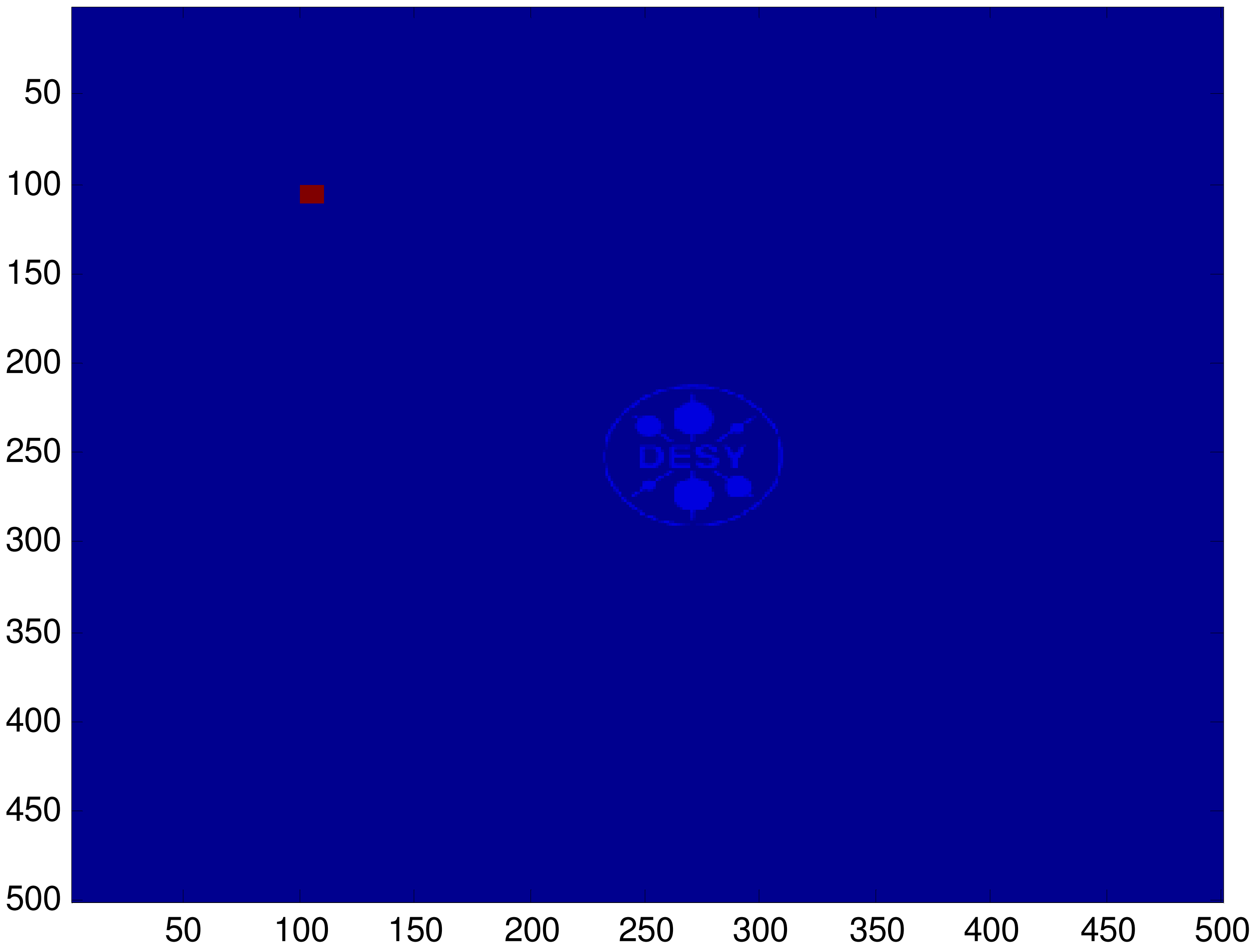
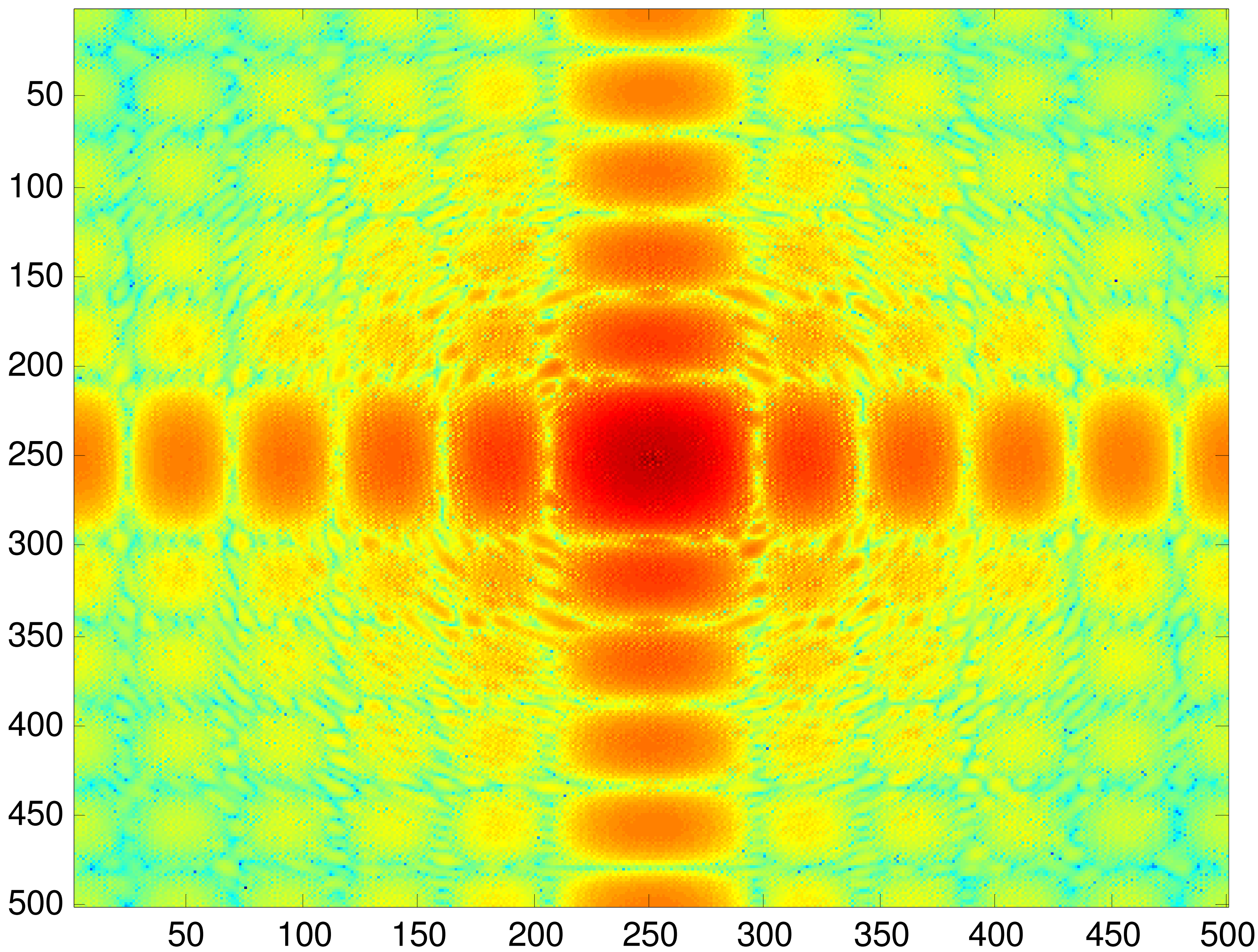
Large reference hole

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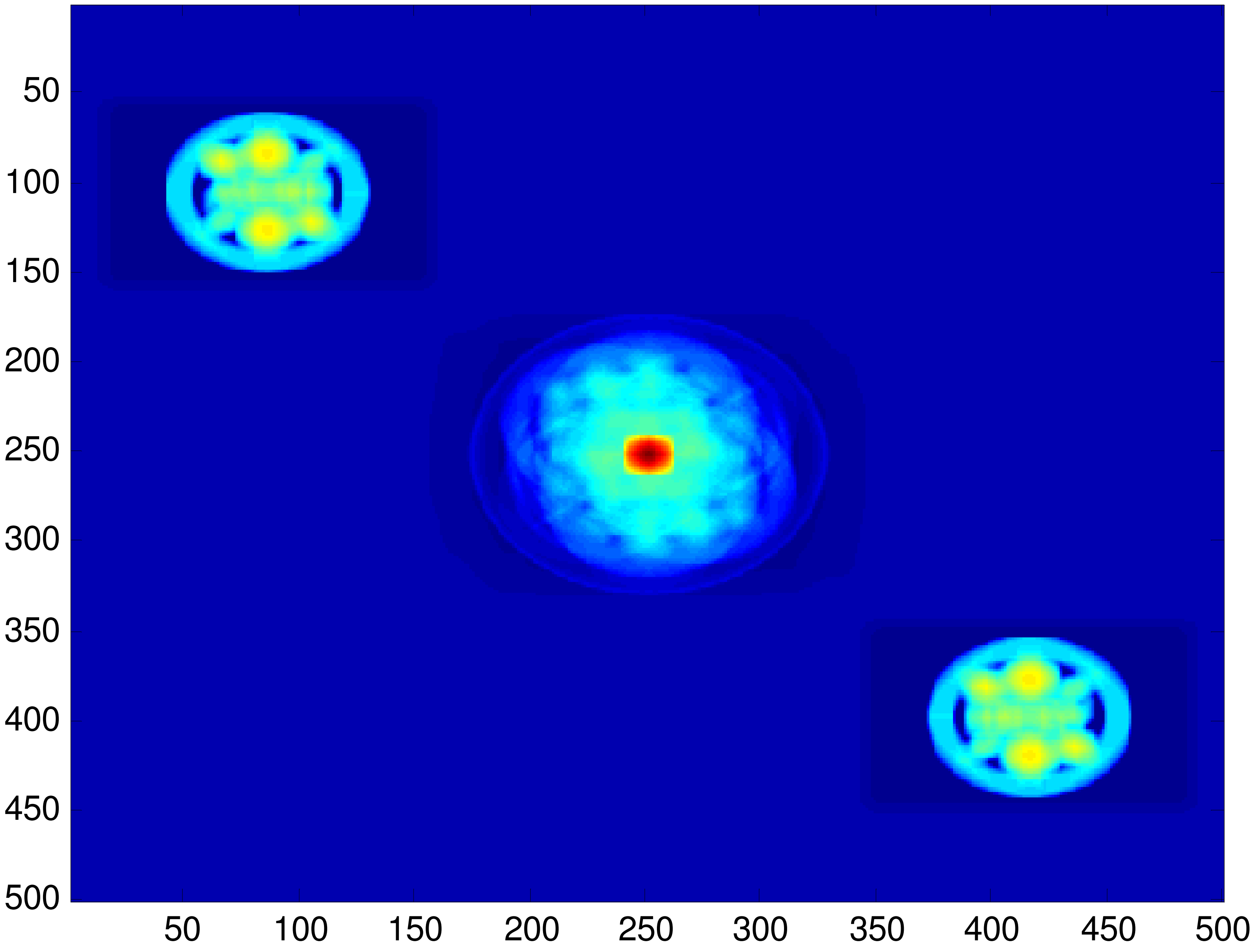
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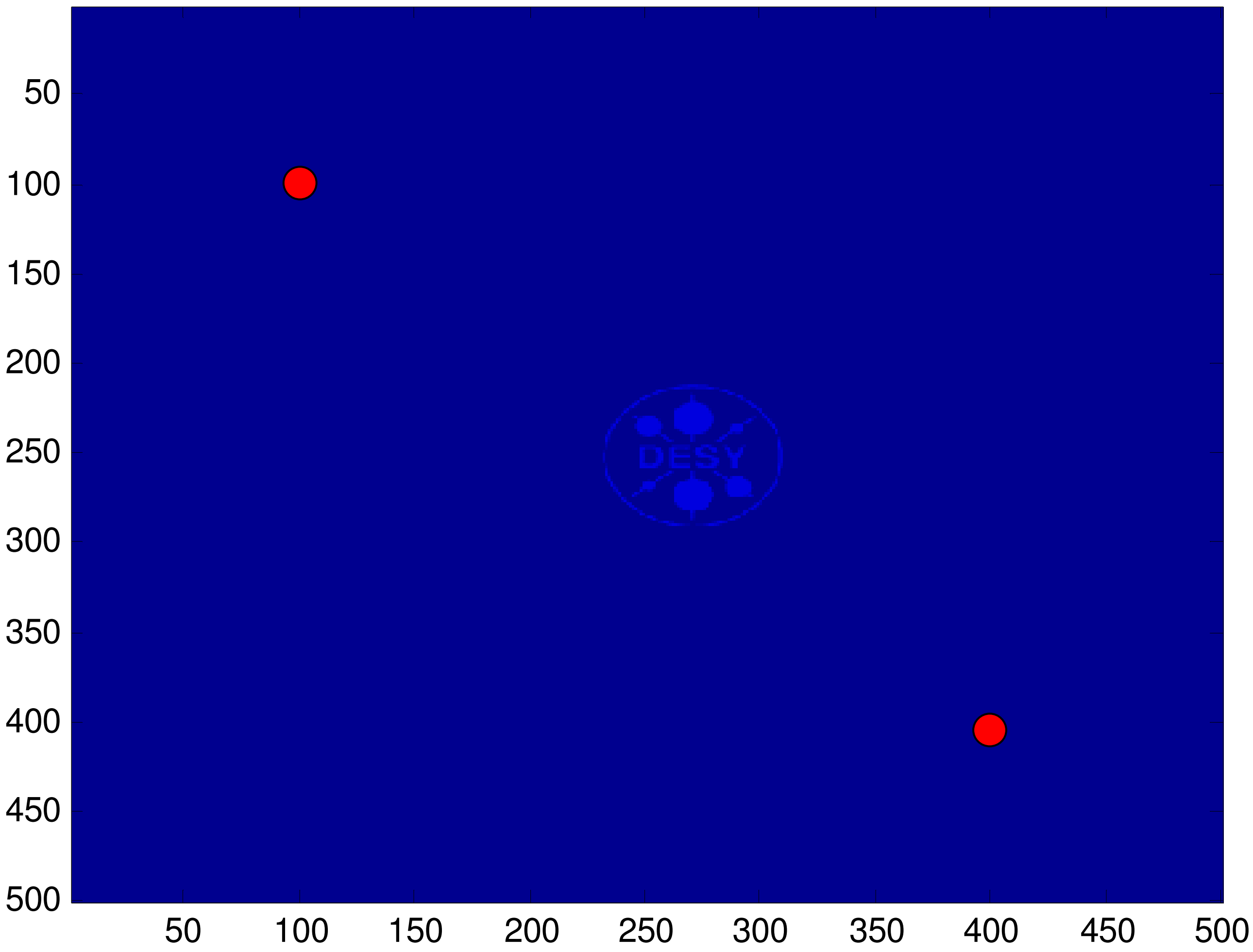
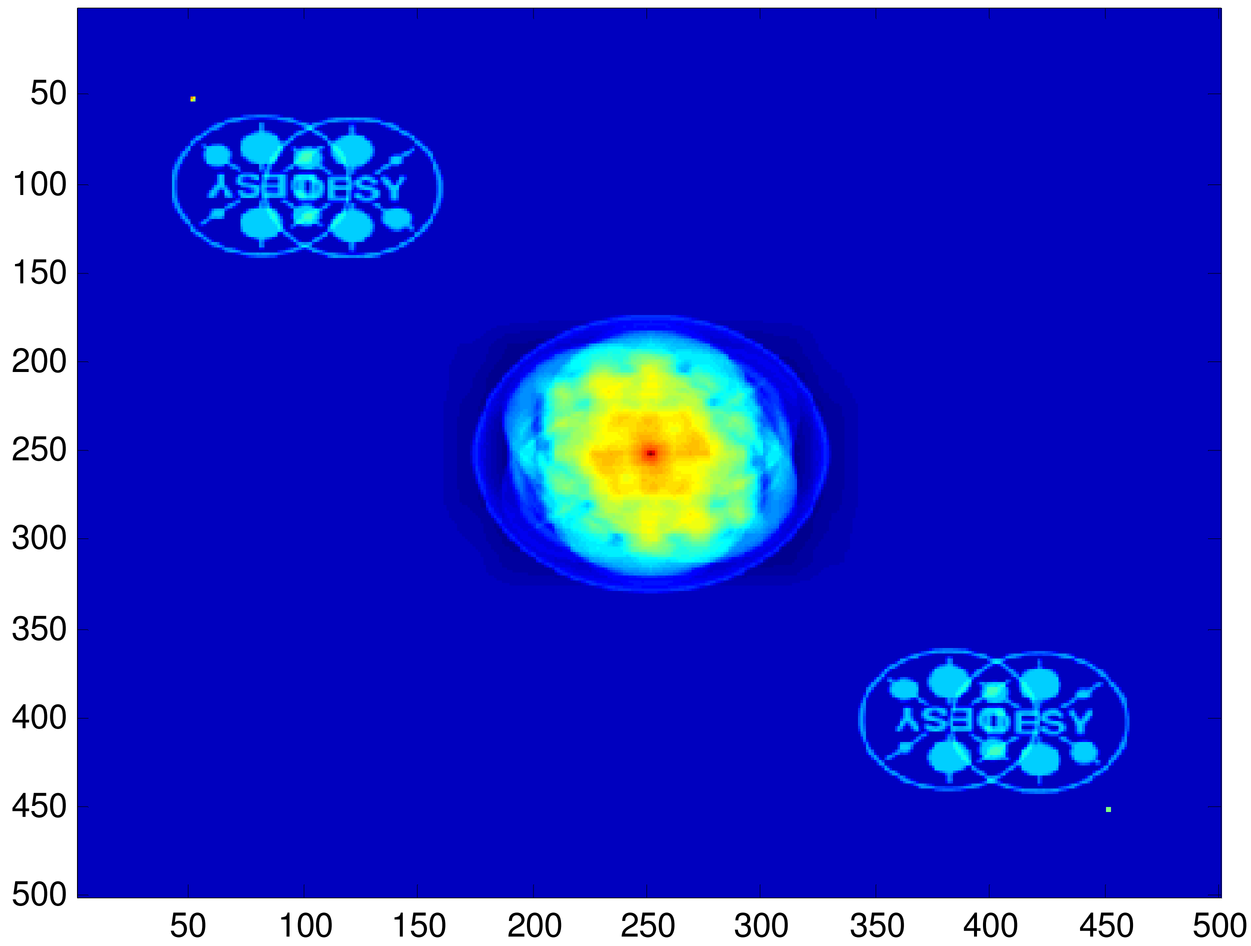
More than one reference hole

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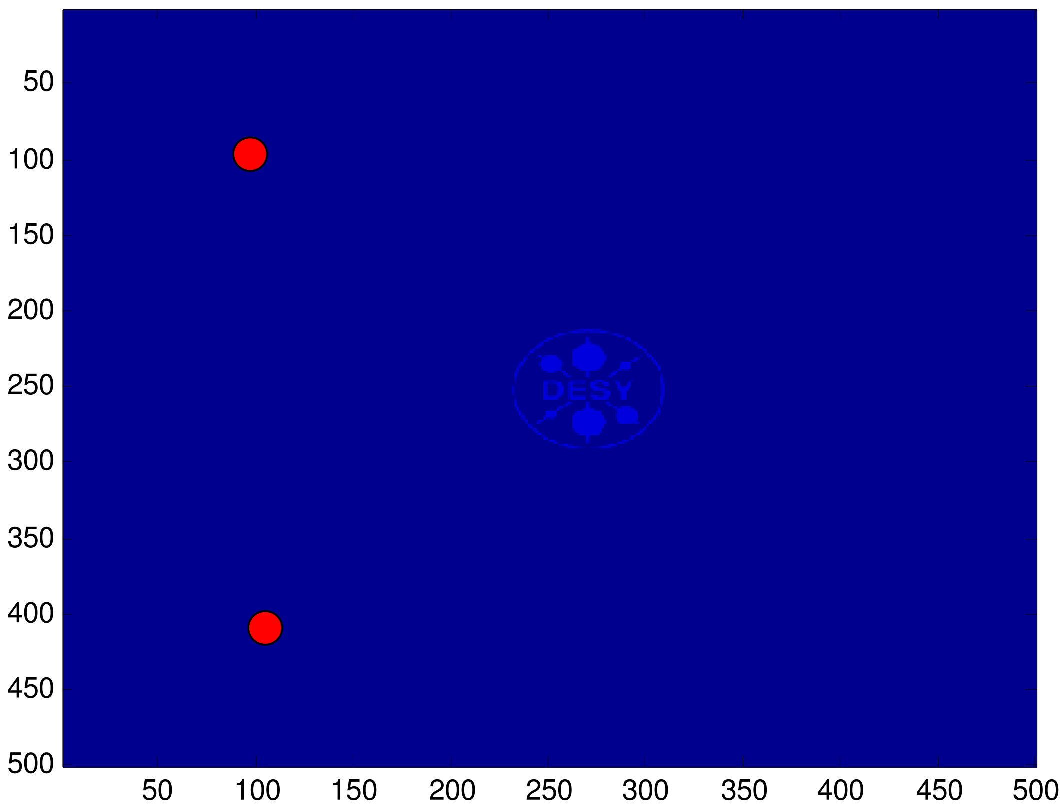

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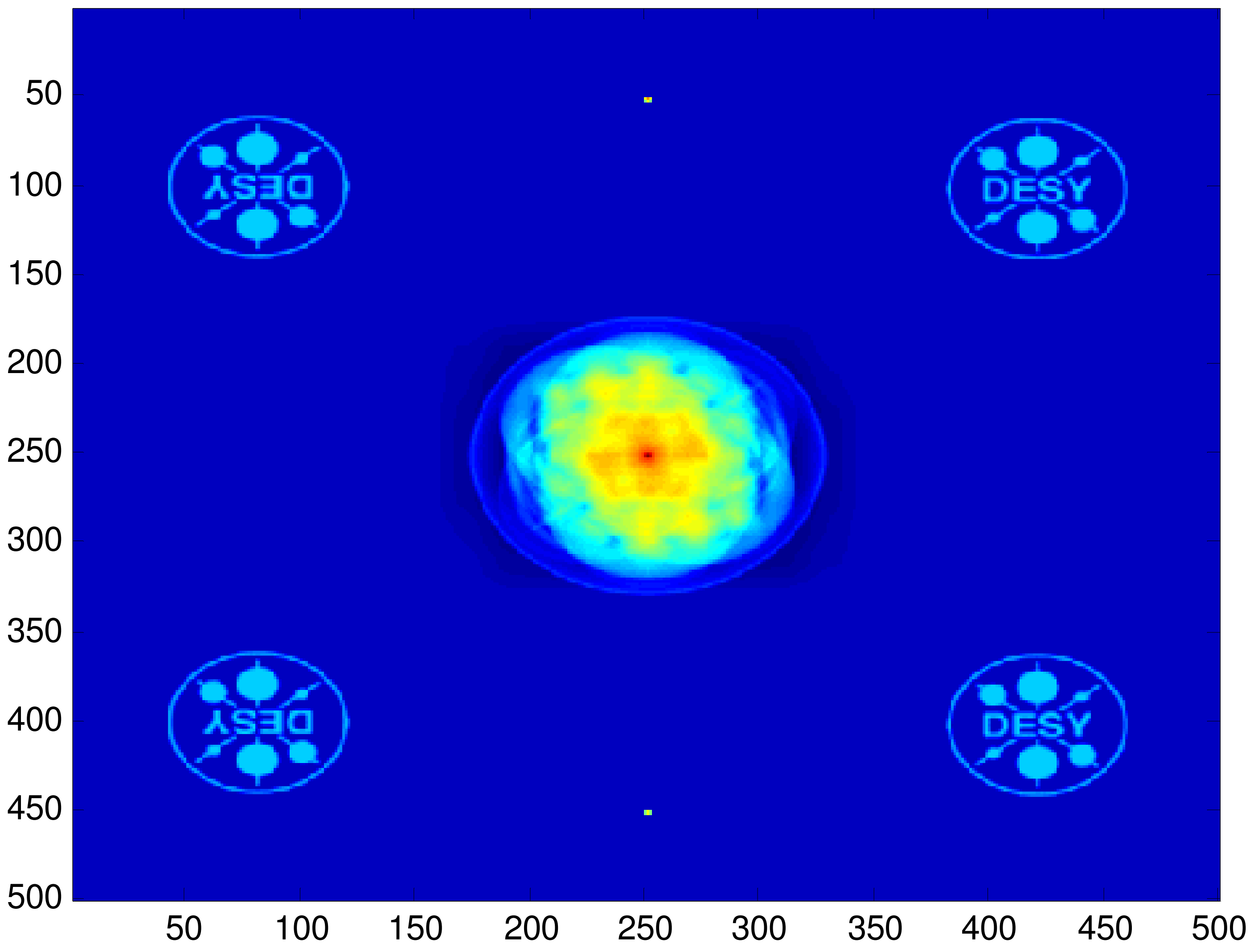
DESY



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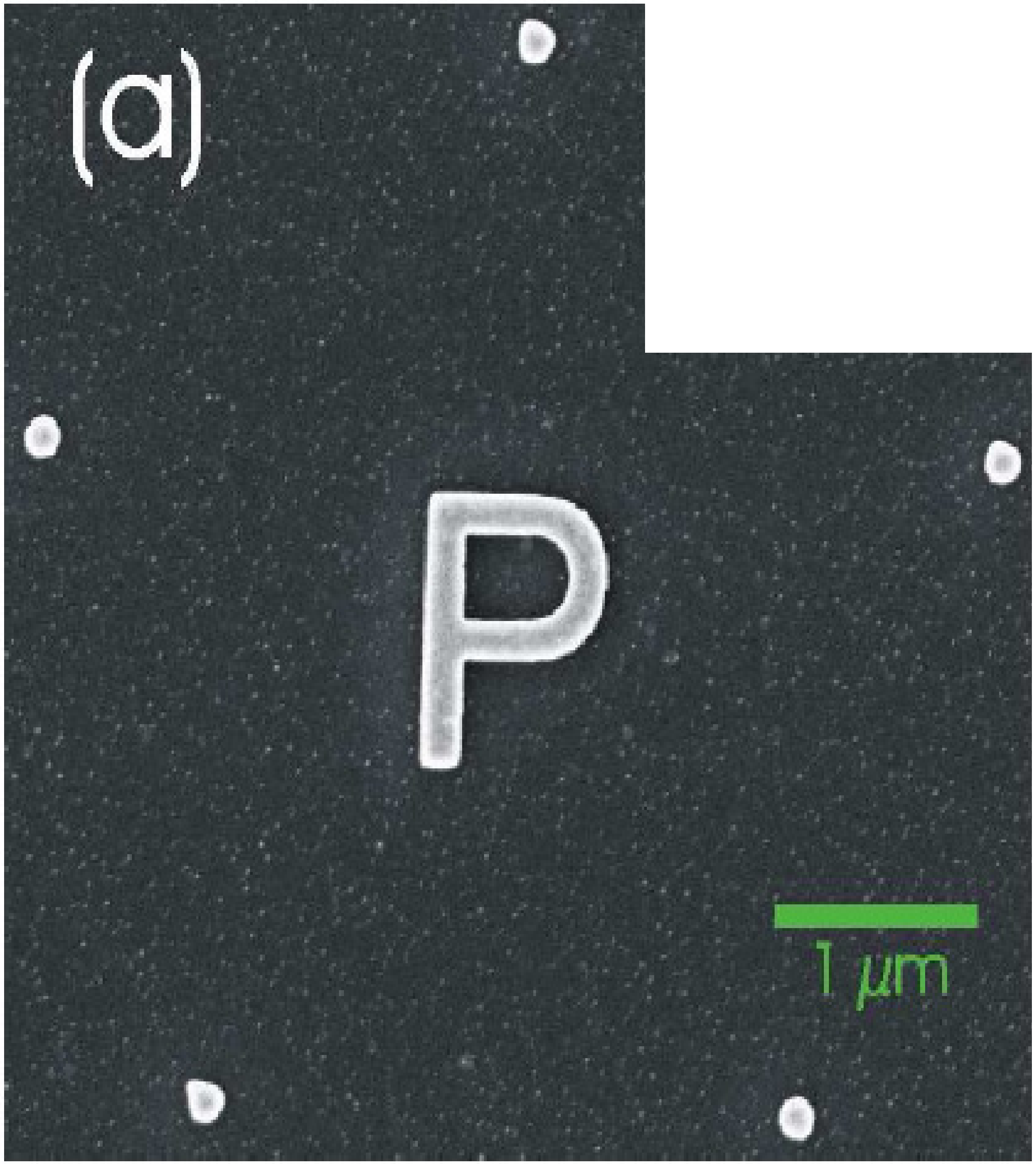
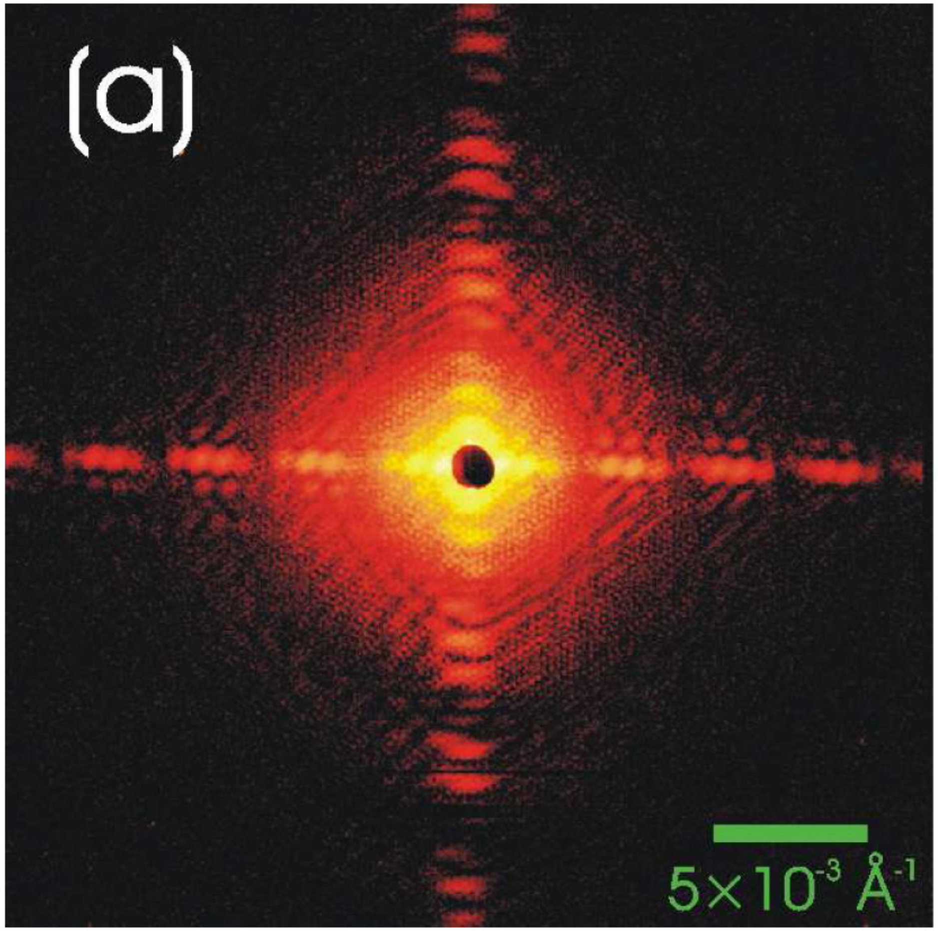
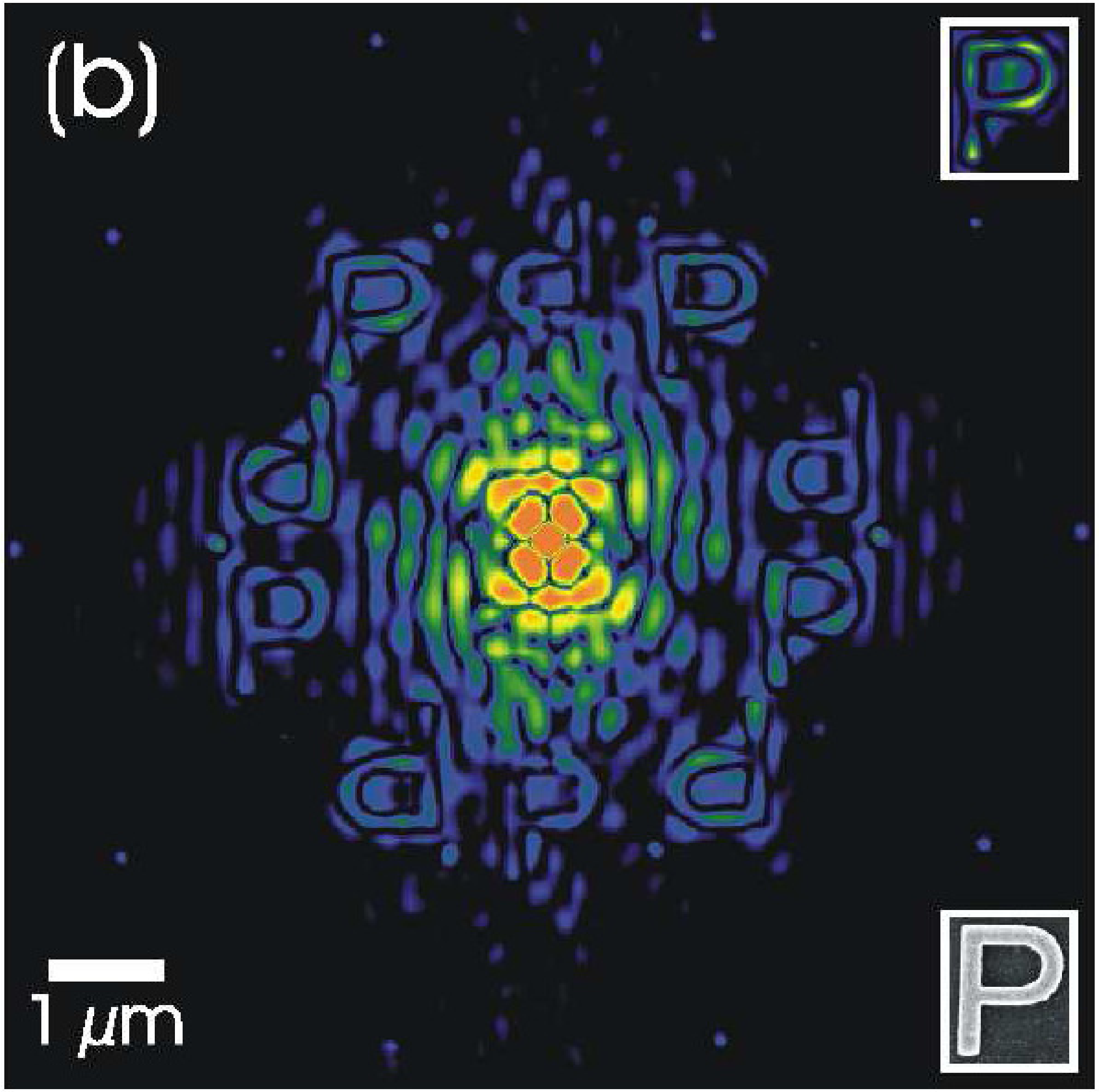
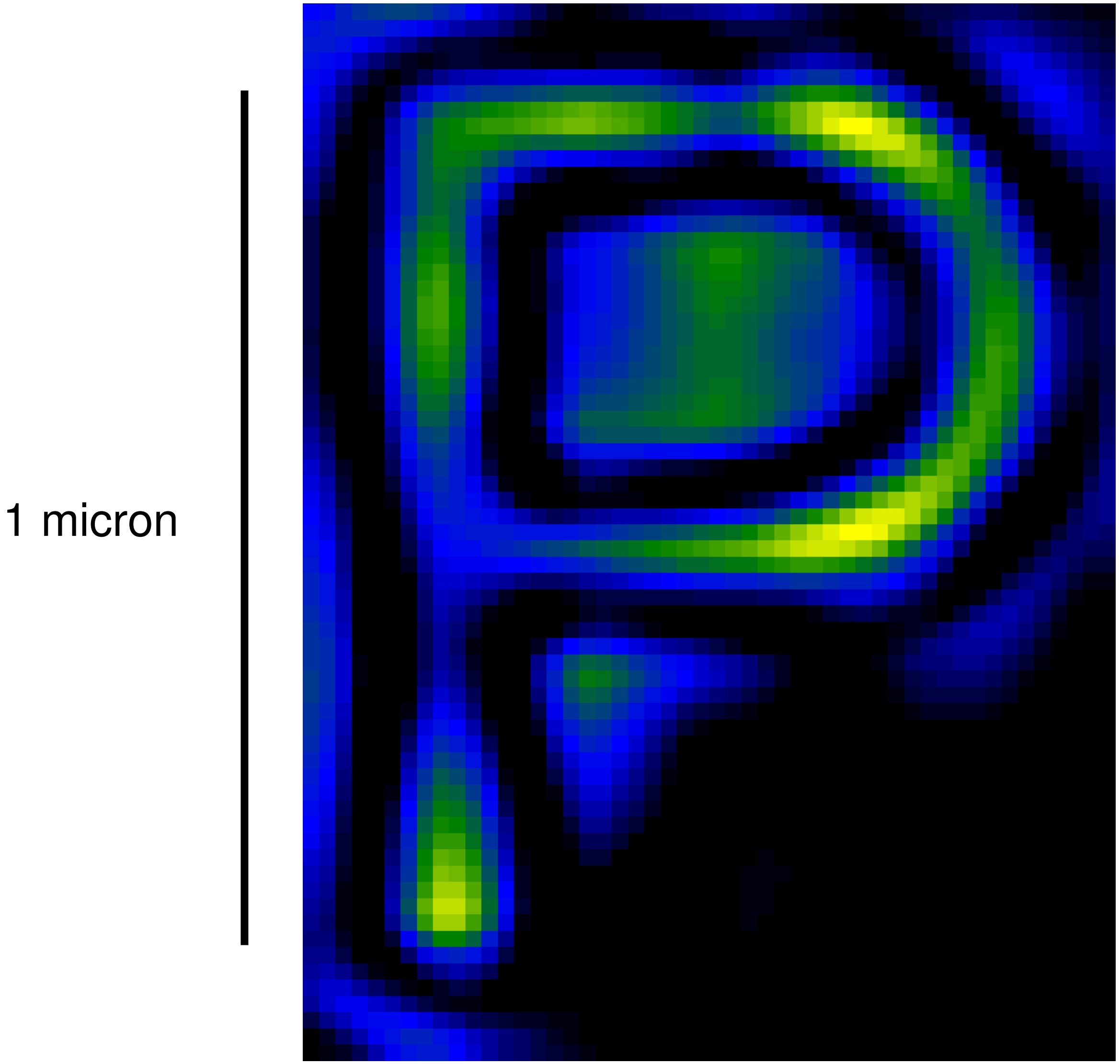

(a)
P
1 μm

First experimental FTH realization using hard X-rays



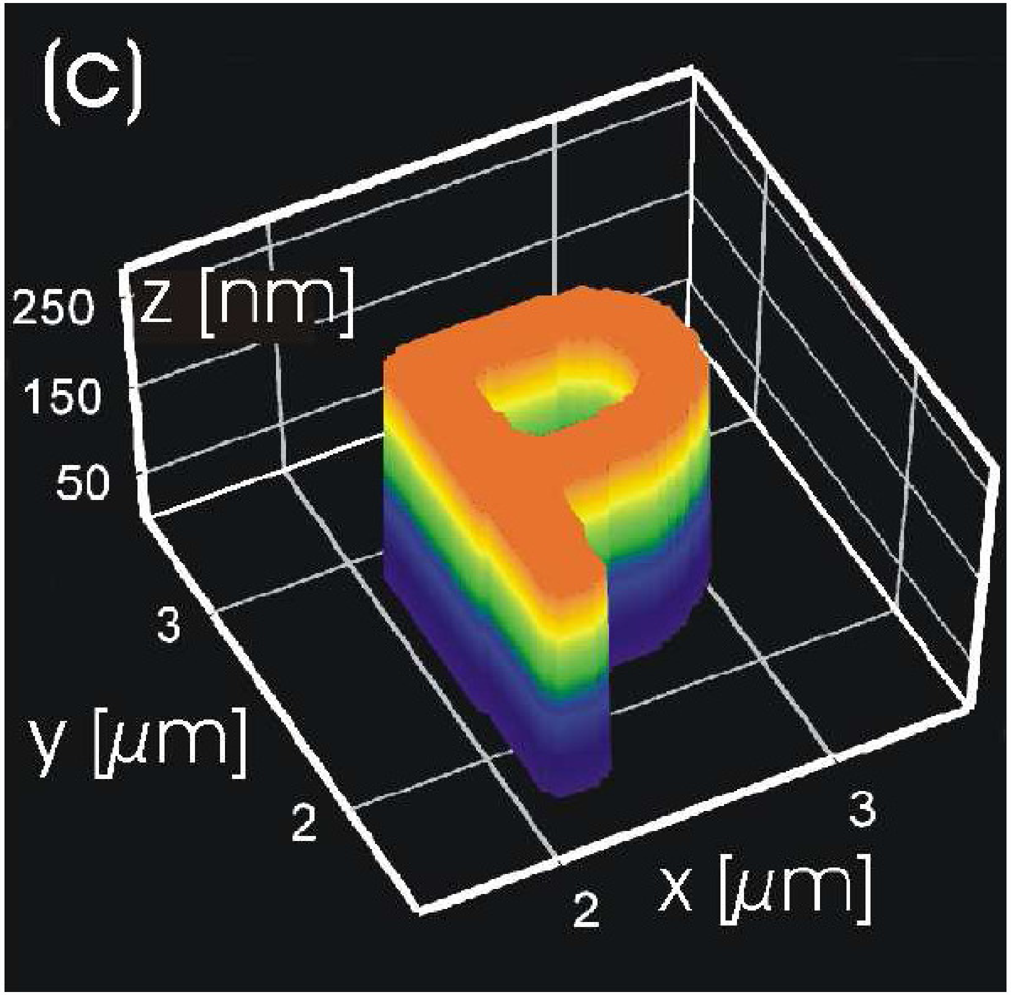
Experiment with 0.15 nm Photonen




1 micron

Combination of Holography and Phase Retrieval

Resolution
20 nm



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