Coherence of light and matter: from basic concepts to modern applications

Part III: G. Grübel

Script 2

Coherence based X-ray techniques

Overview, Introduction to X-ray Scattering, Sources of Coherent X-rays, Speckle pattern and their analysis

Imaging techniques

Phase Retrieval, Sampling Theory, Reconstruction of Oversampled Data, Fourier Transform Holography, Applications

X-ray Photon Correlation Spectroscopy (XPCS)

Introduction, Equilibrium Dynamics (Brownian Motion), Surface Dynamics, Non-Equilibrium Dynamics

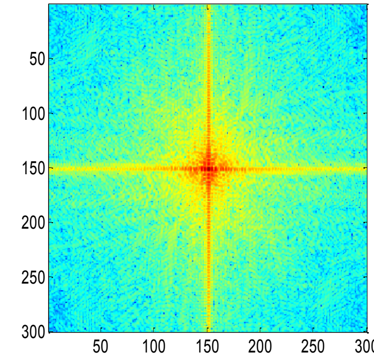
Imaging and XPCS at FEL Sources

Introduction: Experimental Set-Up

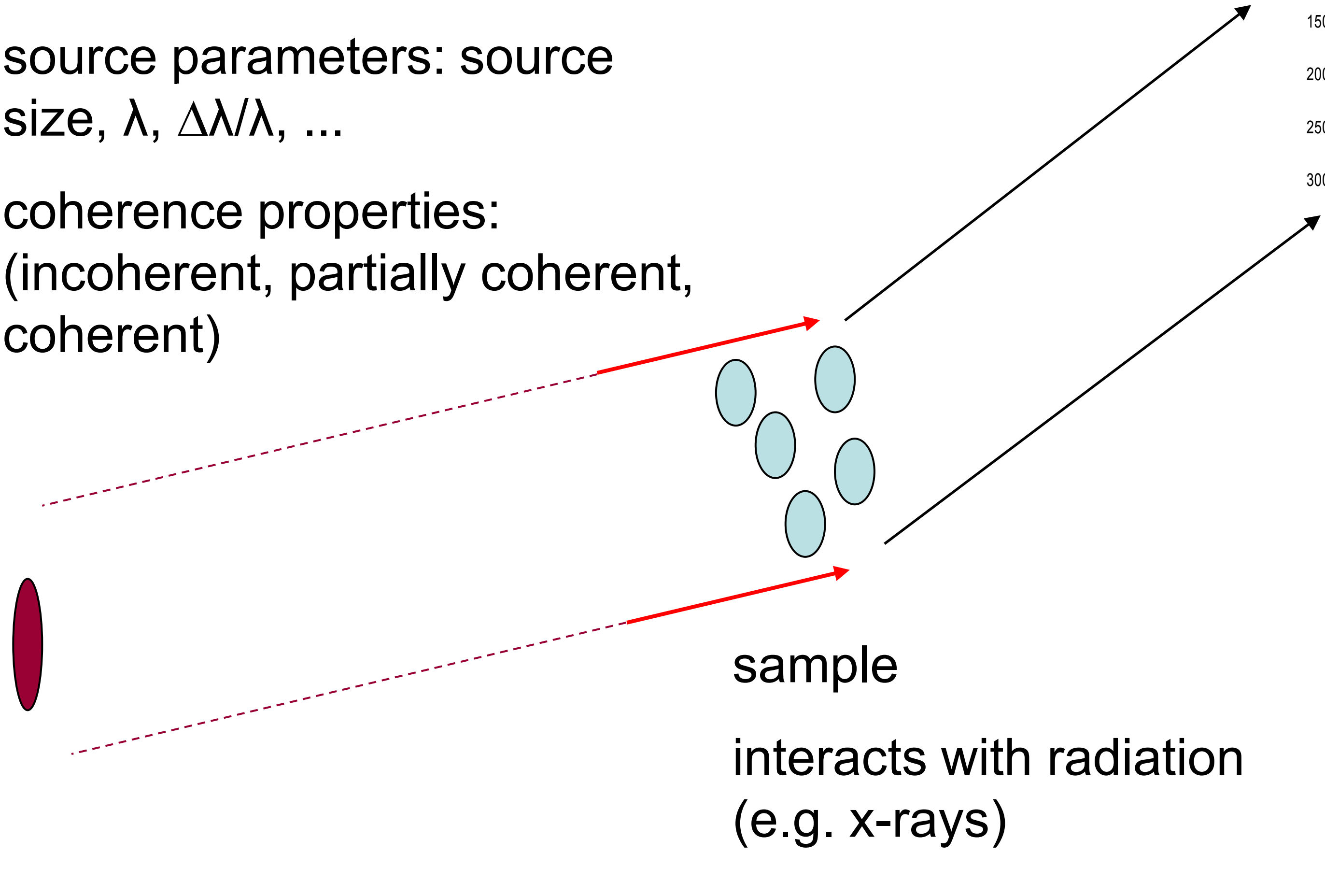
source (visible light, x-rays,..)

source parameters: source size, λ, Δλ/λ, ...

coherence properties: (incoherent, partially coherent, coherent)



detector



sample

interacts with radiation (e.g. x-rays)

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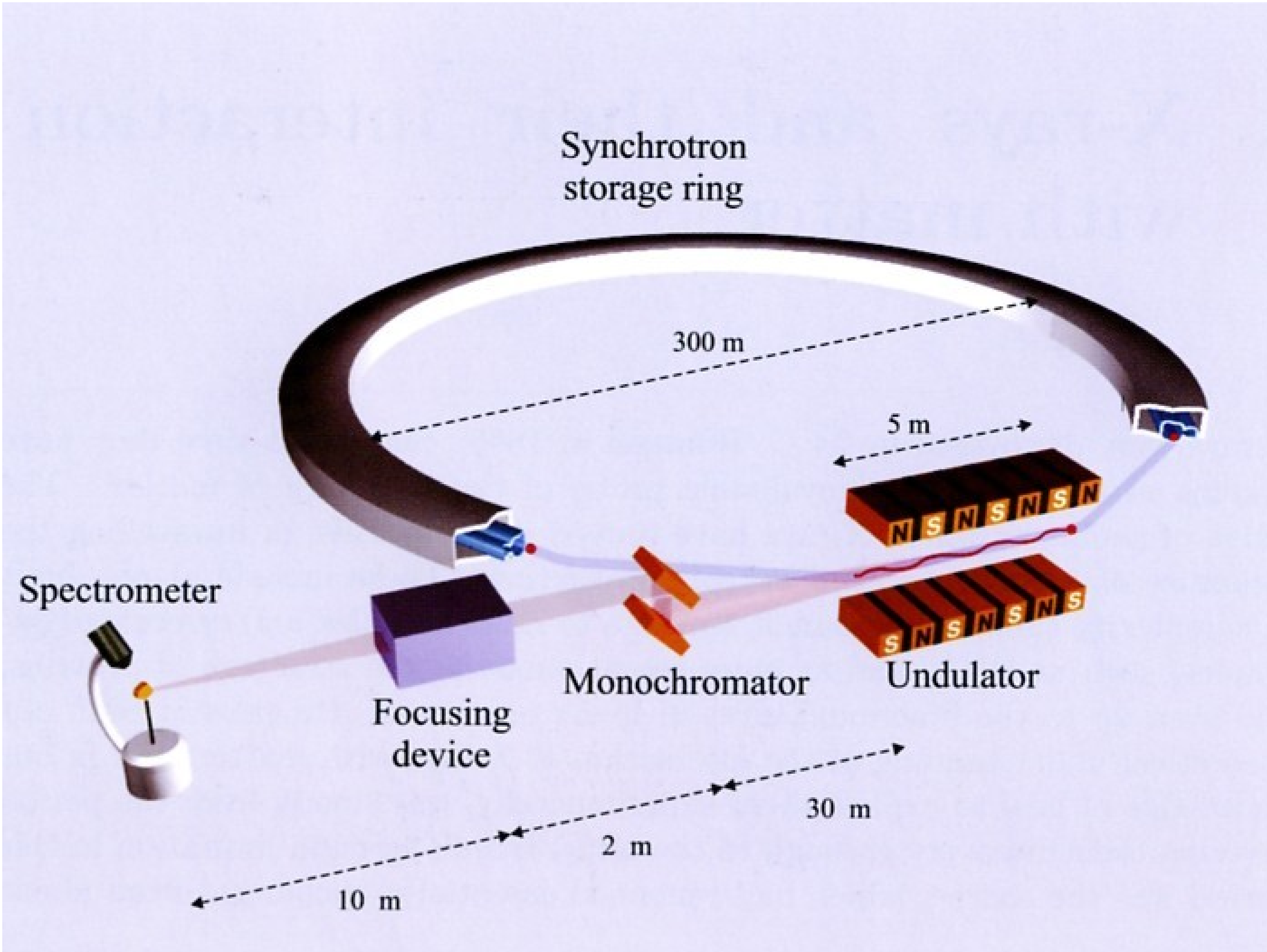
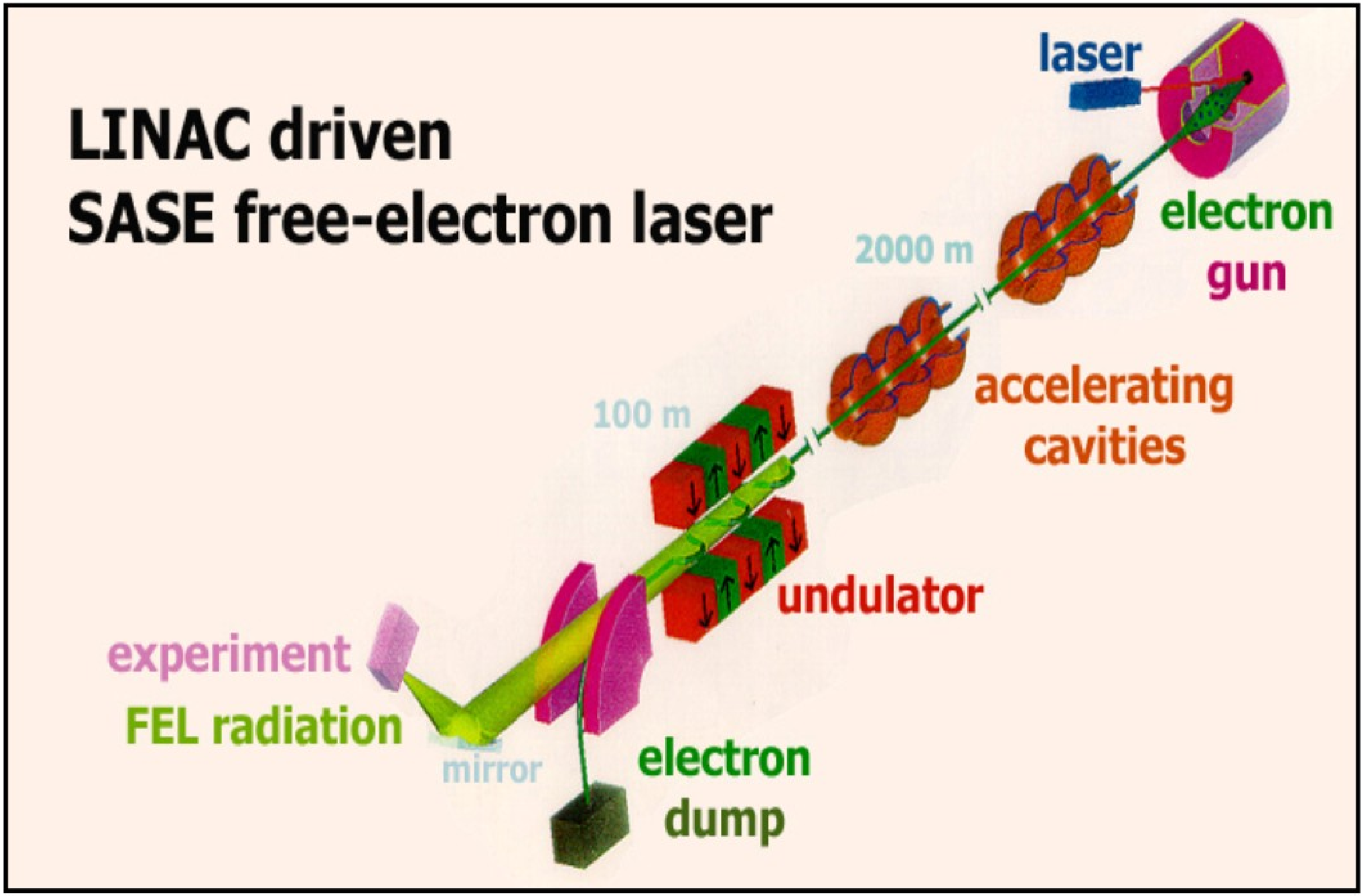
Coherence based X-ray techniques:

Sources of Coherent X-rays

A storage ring facility



A Free Electron Laser (FEL)



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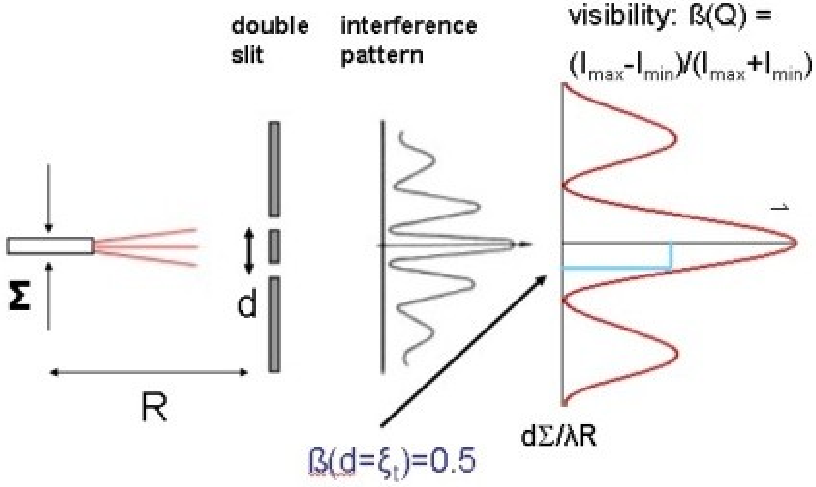
Coherence parameters of an undulator source



Coherent Flux:

$Fc = (\lambda/2)^2 \bullet B$

$= 3.5 \bullet 10^{10}$ ph/s

$B=10^{20}$ ph/s/mm²/mrad²/0.1%bw

$\Delta\lambda/\lambda = 10^{-4}$; $\lambda = 1$Å

Temporal Coherence:

londitudinal coherence length

$\xi_l = \lambda(\lambda/\Delta\lambda) = 1$ µm

$\Delta\lambda/\lambda = 10^{-4}$; $\lambda = 1$ Å

Transverse coherence length:

$\xi_t^2 \bullet \xi_l$ defines coherence volume

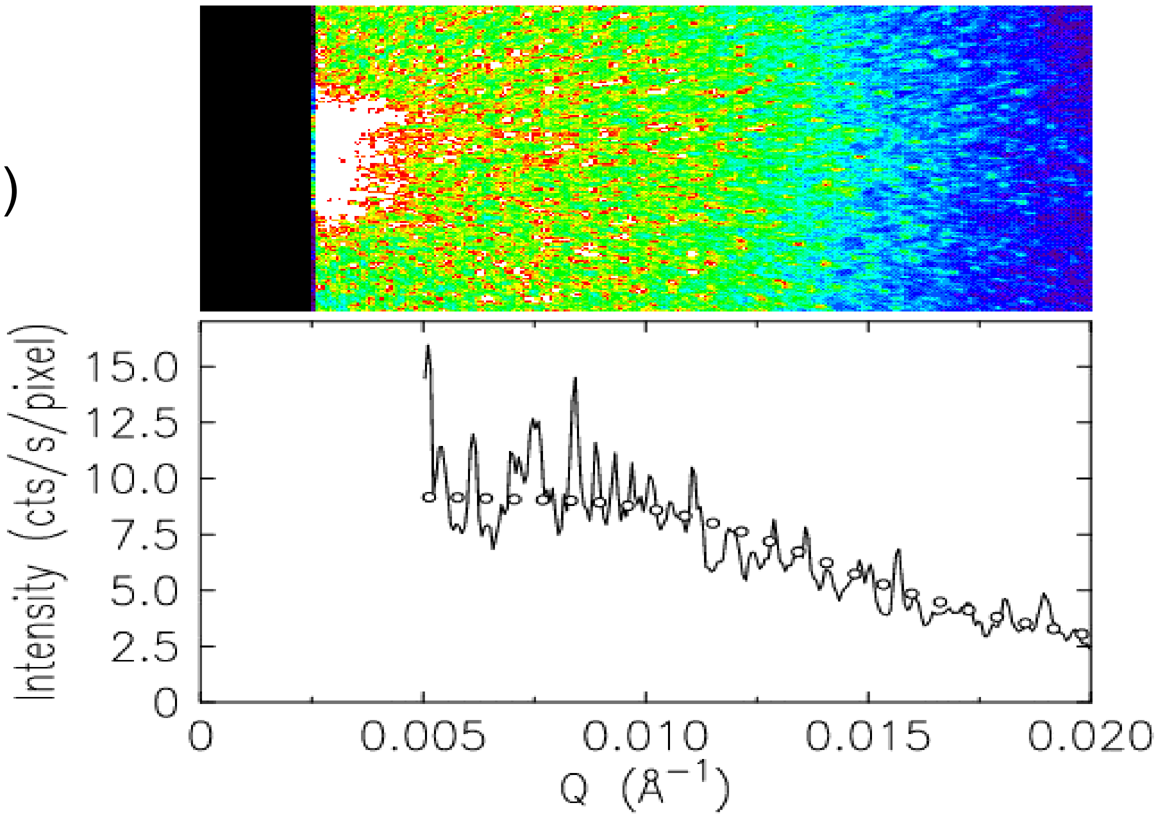
$\xi_t = (\lambda/2)(R/\Sigma)$ = 2.5µm (h), Σ_x=1mm

= 25µm (h), Σ_z=0.1mm

(λ=1Å, R=50m)

Speckle pattern from a porous silica gel

Aerogel
λ=1Å
CCD (22 μm)



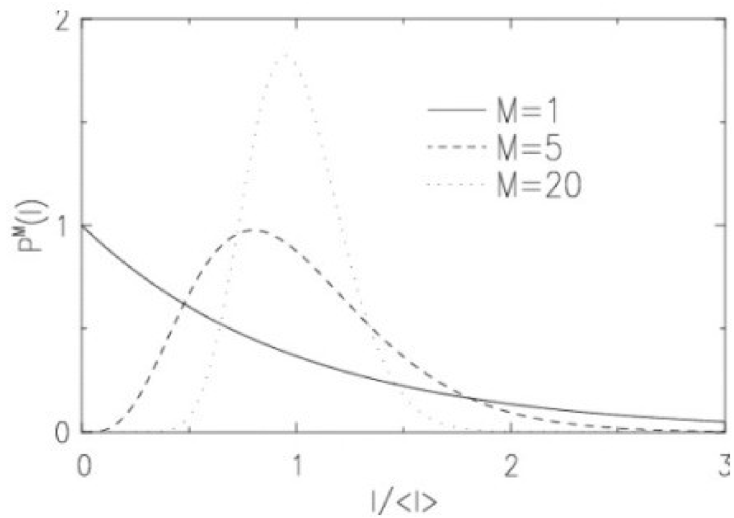
Abernathy, Grübel, et al.J. Synchroton Rad. 5, 37, 1998

Statistical Analysis of Speckle Pattern (1)

If the source is fully coherent and the scattering amplitudes and phases of the scattering are statistically independent and distributed over 2π one finds for the probability amplitude of the intensities:

$P(I) = (1/\langle I\rangle) \exp(- I/\langle I\rangle)$

Mean: $\langle i\rangle$
Std.Dev. σ: $\mathrm{sqrt}(\langle I^2\rangle - \langle I\rangle^2) = \langle I\rangle$
Contrast: $ß = \sigma^2/\langle I\rangle^2 \; 1$



<u>partially coherent illumination:</u>

the speckle pattern is the sum of M

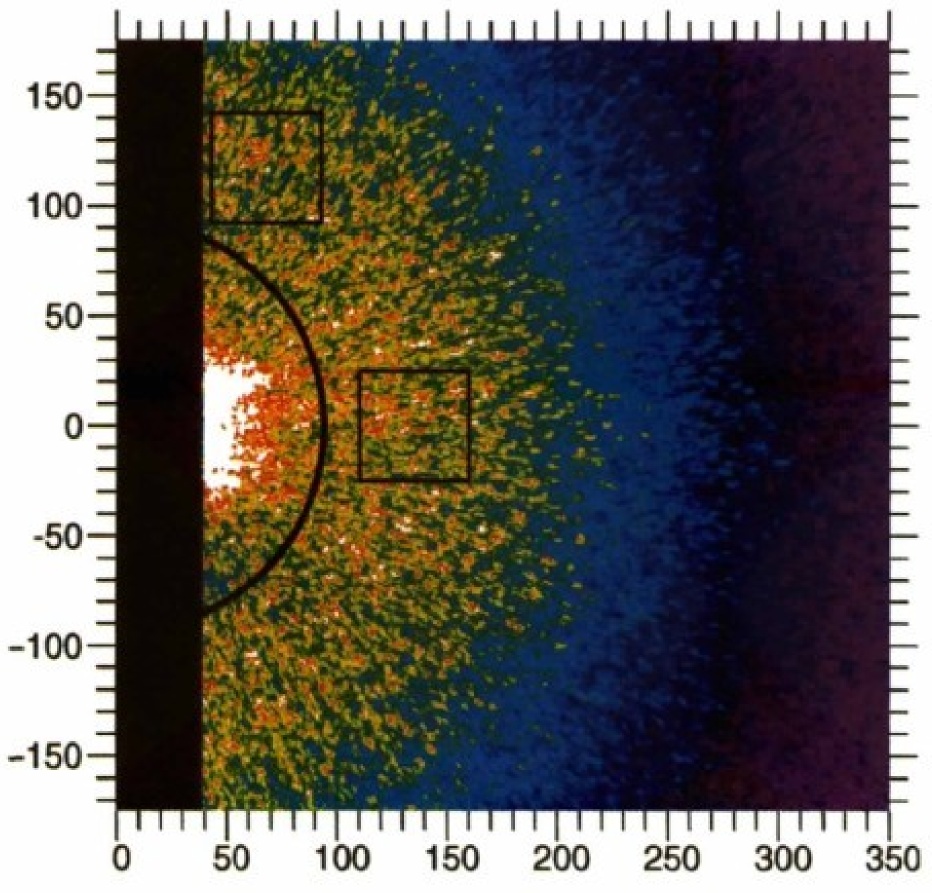
independent speckle pattern

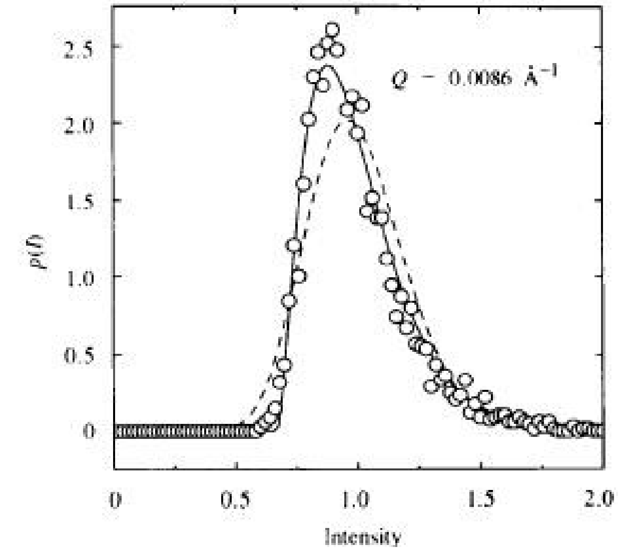
$P_M(I) = M^M \bullet (I/\langle I\rangle)^{M-1}/(\Gamma(M)\langle I\rangle) \bullet \exp(-MI/\langle I\rangle)$

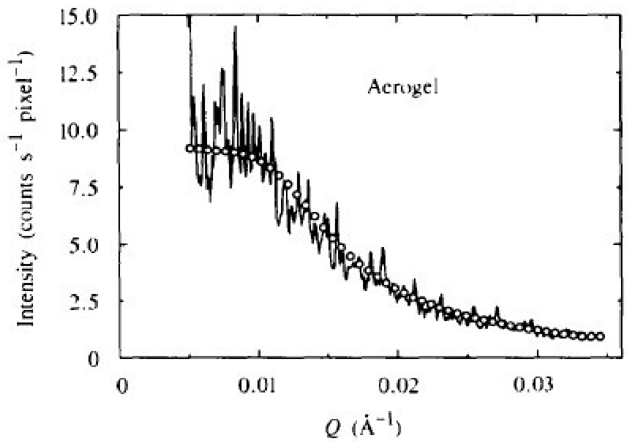
Mean: $\langle I\rangle$
$\sigma = \langle I\rangle/M^{1/2}$
$ß = 1/M$

Statistical Analysis of Speckle pattern (2)

Aerogel, λ=1Å, CCD (22 µm)







normalized two-point correlation function:

$$C(\mathbf{r}_1,\mathbf{r}_2) = [\ \langle I(\mathbf{r}_1)\bullet I(\mathbf{r}_2)\rangle / \langle I(\mathbf{r}_1)\rangle\bullet\langle I(\mathbf{r}_2)\rangle]\ -1$$

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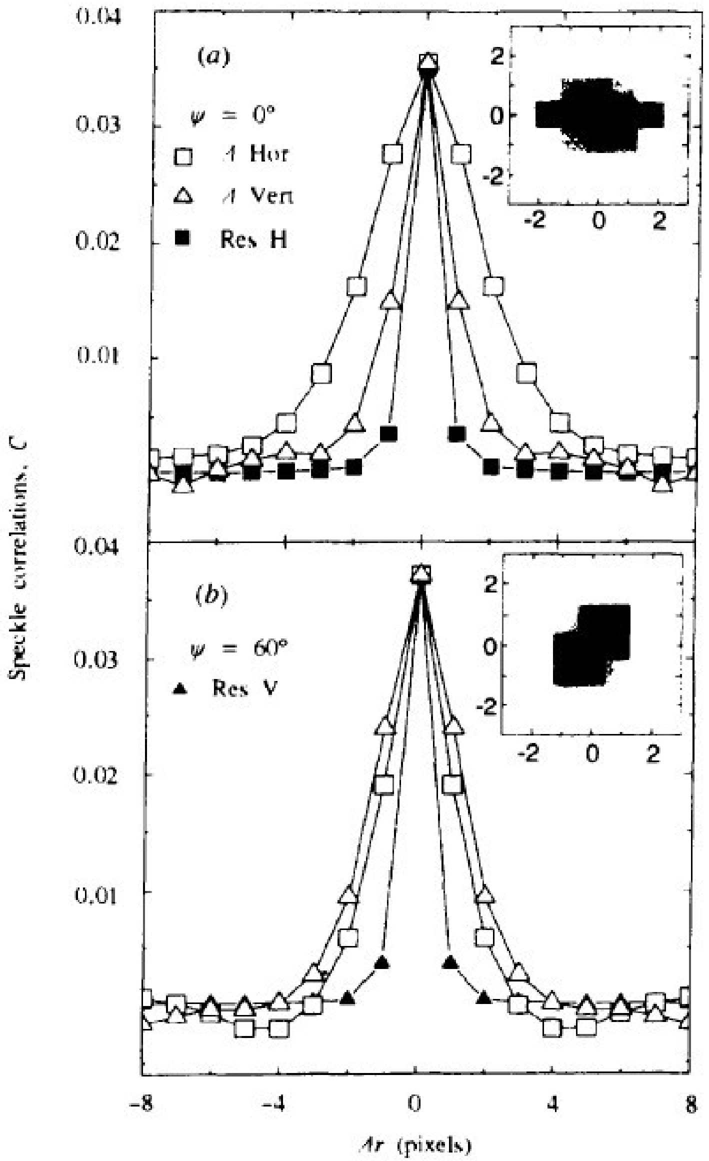
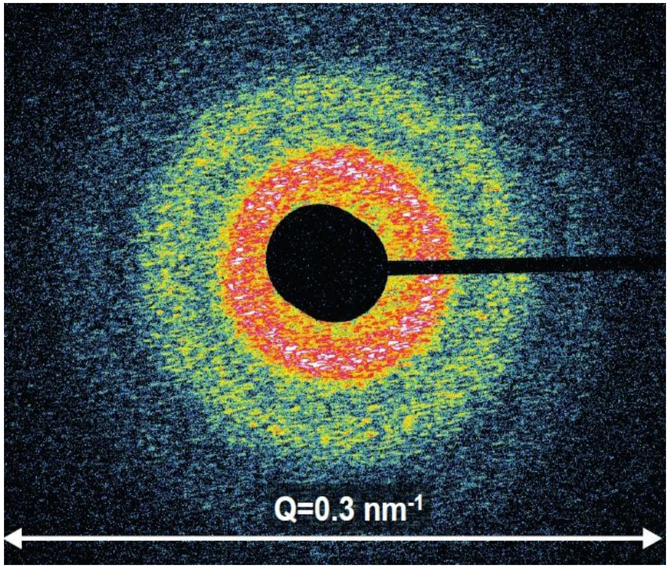
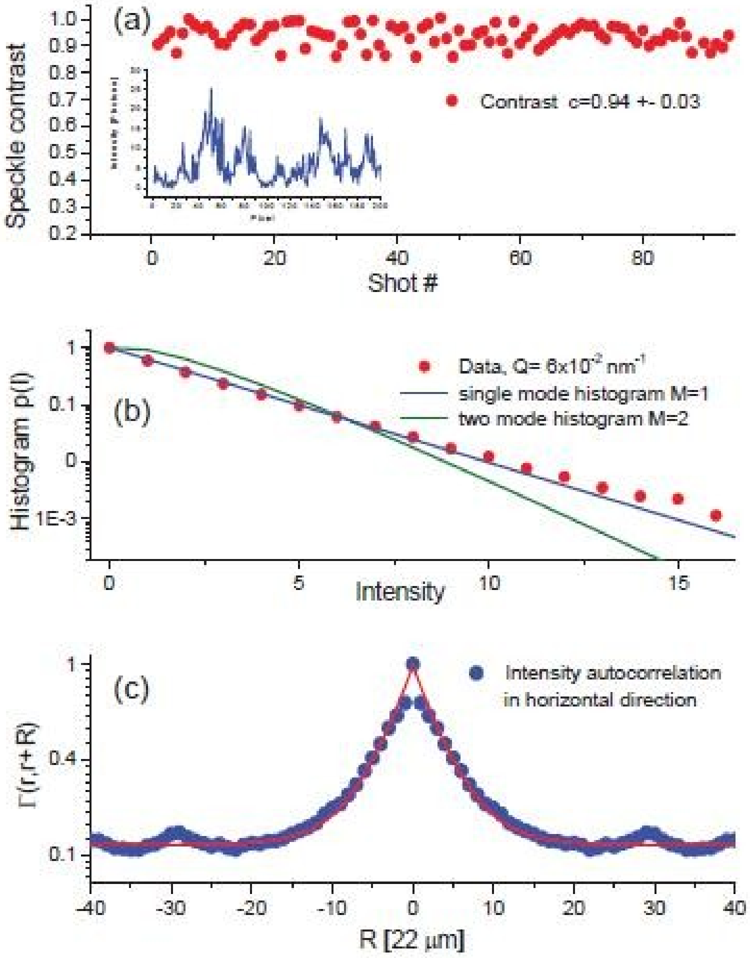


$$C(\mathbf{r}_1,\mathbf{r}_2) = [\, \langle I(\mathbf{r}_1)\bullet I(\mathbf{r}_2)\rangle \,/\, \langle I(\mathbf{r}_1)\rangle\bullet\langle I(\mathbf{r}_2)\rangle] \; -1$$

width: ΔC; contrast: ß = $C(\mathbf{r},\mathbf{r})$

▪ The Linac Coherent Light Source (LCLS)





Single pulse hard X-ray speckle pattern captured from nano-particles in a colloidal liquid (photon wave-length λ= 1.37 Å

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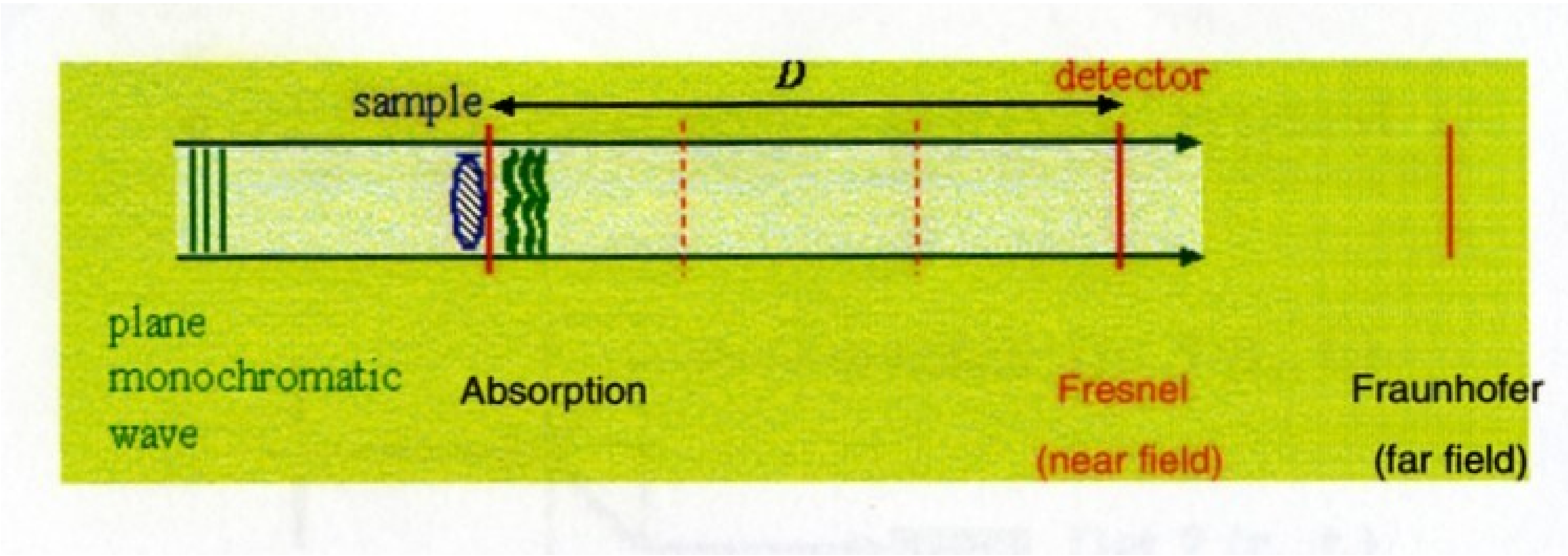
Imaging techniques:

Lensless Imaging, Fourier Transform Holography

Imaging techniques



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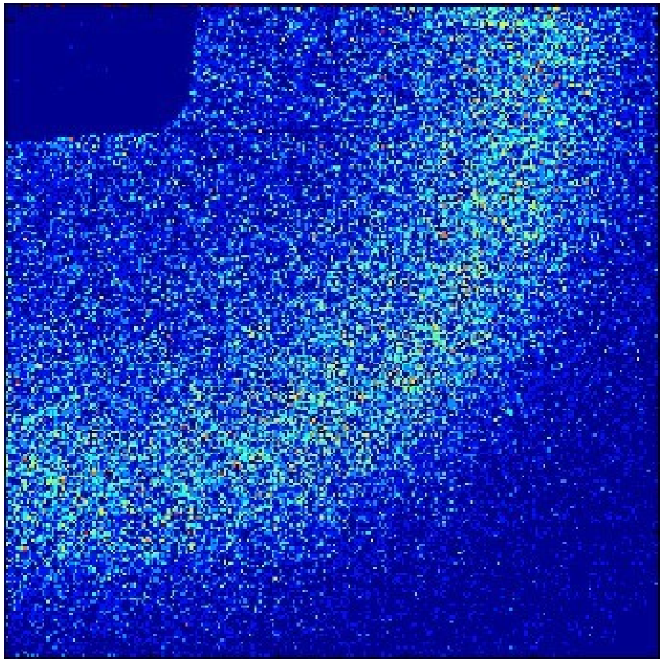
(Lensless) Imaging

If coherent light is scattered from a disordered system it gives rise to a random (grainy) diffraction pattern, known as “speckle”. A speckle pattern is an interference pattern and related to the exact spatial arrangement of the scatterers in the disordered system.

$$I(Q,t) \sim S_c(Q,t) \sim | \sum e^{iQRj(t)} |^2$$

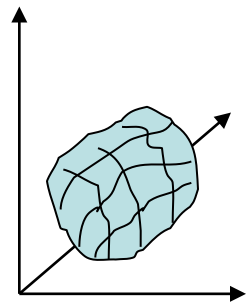


j in coherence volume $c= \xi_t^2 \xi_l$

phase information is lost

 Coherence of light and matter: from basic concepts to modern applications, Vorlesung im GrK 1355, SS 2011 A. Hemmerich & G. Gruebel

The Phase Problem (1)



consider a non-periodic object with mass density $\rho(\mathbf{x})$

$$F(\mathbf{k}) = \int \rho(\mathbf{x}) \exp(2\pi i\, \mathbf{k} \cdot \mathbf{x})\, d\mathbf{x}$$

approximate object ρ(x) and its fourier transform by (N) "elements"

$$F(\mathbf{k}) = \sum_{x=0}^{N-1} \rho(\mathbf{x}) \exp(2\pi i\, \mathbf{k} \cdot \mathbf{x} / N) \qquad (\$)$$

with $I(\mathbf{k}) = | F(\mathbf{k}) |^2$

here ($) defines a set of <u>N equations</u> to be solved for $\rho(\mathbf{x})$ at each pixel

J.Miao et al., J. Opt. Soc. Am.A; Vol.15 (1998)1662

The Phase Problem (2)

<u>note:</u> cannot distinguish between:

$f(x)$

$f(x+x_o)\, e^{i\Phi}$

$f^*(-x+x_o) e^{i\Phi}$

i) ρ(x) complex — 2N variables (real and imaginary part)
N equations

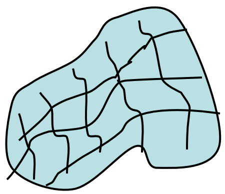
ii) ρ(x) real — N variables

Friedel's law: $I(\mathbf{k}) = I(-\mathbf{k})$| central symmetry:
N/2 equations

problem underdetermined by factor 2

Phase Retrieval and Oversampling

without a priori information eq. ($) cannot be solved uniquely
== > decrease number of unknown variables:



object elements with known scattering density (e.g. zero)

i) use objects with some known scattering density

σ = total number of elements/number of unknown-valued pixels > 2

ii) increase number of known quantities in eq. ($) by “oversampling”

sample the magnitude of a Fourier transform finely enough to get a finite support for the object such that the element values outside the finite support is zero

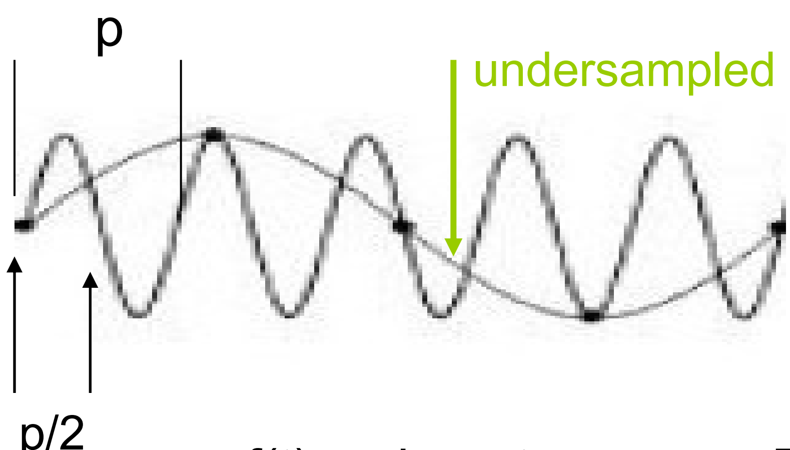
need: $\sigma > 2$ in 1D; $\sigma > 2^{1/2}$ in 2D; $\sigma > 2^{1/3}$ in 3D

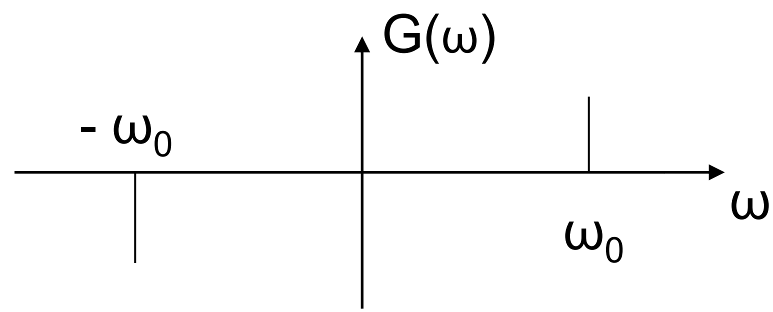
iii) apply iterative algorithms to retrieve phase

iv) resolution determined by maximum momentum transfer Q

Sampling Theory (1)

<u>Sampling Theorem:</u> A signal $f(t)$ that is
i) bandwidth limited
ii) sampled above the Nyquist frequency
is completely determined by its samples.



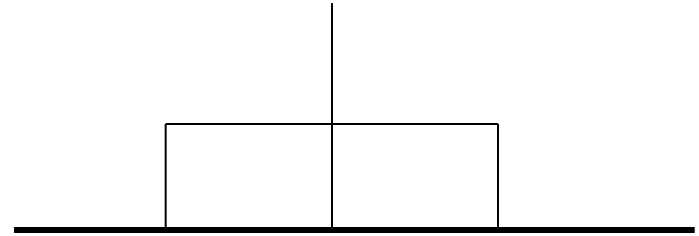


$$f(t) = \sin \omega_0 t \quad ===> \quad \text{FT} \quad <=== \quad G(\omega) = \int f(t)\, e(-i2\pi\omega t)\, dt$$

A signal is called bandwidth limited if it contains no frequencies outside the interval: $[-f_{max}, f_{max}]$. Here f_{max} ($=\omega_0$) is called the bandwidth of the function. The Nyquist frequency is usually given by $f_{NY} = 2 f_{max}$ ($=2\omega_0$).

<u>Note:</u> period $p = 2\pi/\omega_0$ and (Nyquist) sampling period $p_{NY} = 2\pi/2\omega_0 = p/2$.
(undersampling: $p>p_{NY}$; oversampling: $p<p_{NY}$)

Sampling Theory (2)

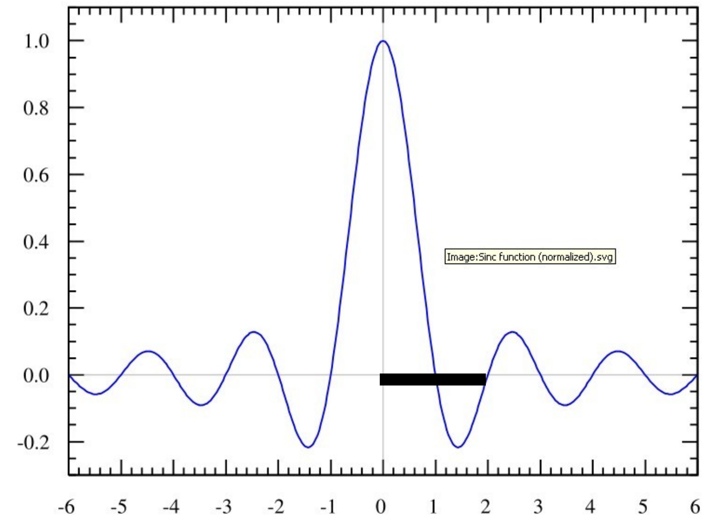


consider $z(t) = 1 \qquad |t| \leq L$

$= 0 \qquad$ elsewhere

its FT is given by

$I(x) = \text{sinc}\,(Lx) = \sin\,(Lx) / Lx$



period $\qquad p = 2\pi/L$

max frequency: $\qquad f_{max} = 2\pi/p = L$

Nyquist frequency: $\qquad f_{Ny} = 2L = 4\pi/p$

sampling period: $\qquad p_{NY} = 2\pi/f_{NY} = p/2$

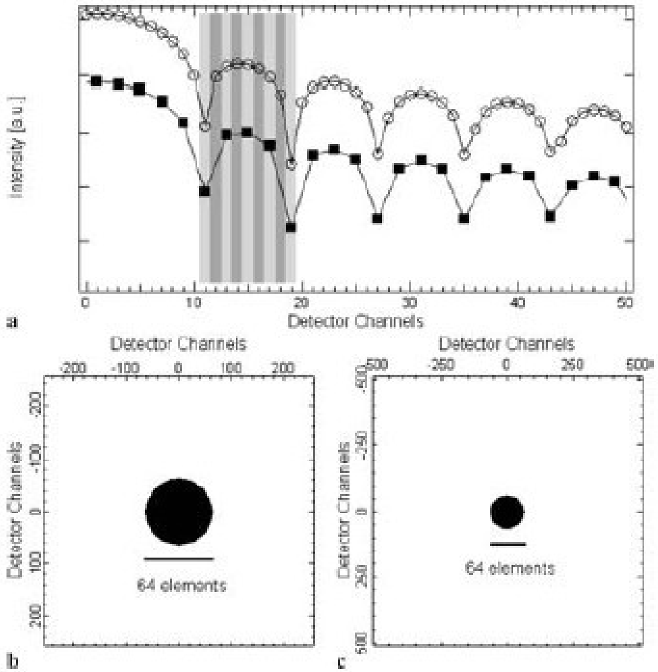
need to sample in fourierspace on a scale $\leq p/2$

“oversampling” corresponds to sampling in “real” space on a scale $\xi > 2L$

Sampling Theory (3)

scattering from a circular slit aperture: $I(q) = I_0 \, |2J_1(qa)/qa|^2$

scattering from a rectangular slit aperture: $I(q) = I_0 \, |\sin(qa)/qa|^2$



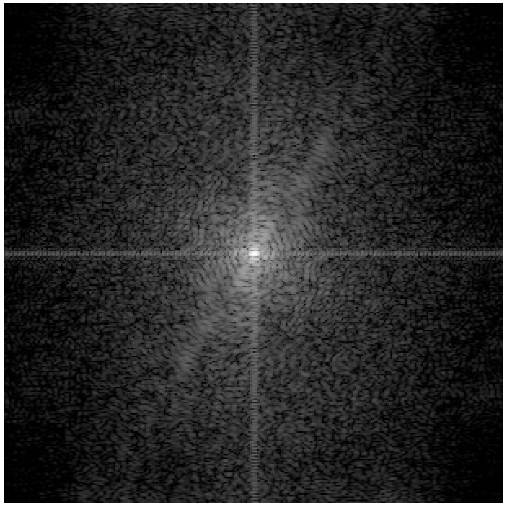
scattering from circular pinhole aperture with a linear oversamp-ling ratio of 4 (8)

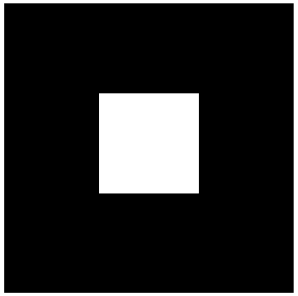
S. Eisebitt et al., Appl. Phys. A80(2005)921

Fourier transform of an oversampled Airy pattern illustrates sampling of an area 4 (8) times bigger than the investigated object.

Reconstructing the real space structure – the main principle

Gerchberg&Saxton(1972); Fienup (1982)

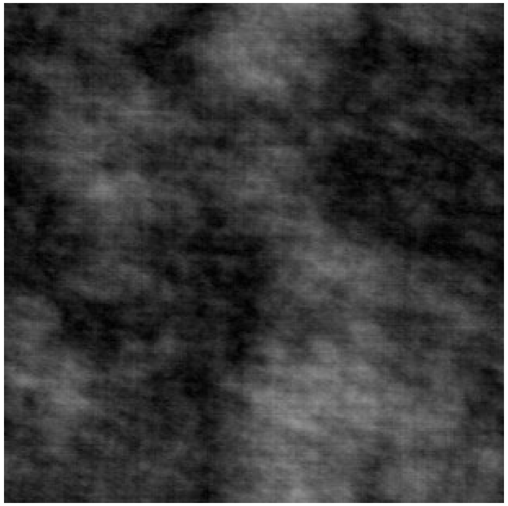




-1. Guess
support in real space

0. Add random phases to measured amplitudes
1. FT into real space
2. Set pixels outside support to zero and use positivity
3. FT into fourier space
4. Substitute amplitudes with measured values

Loop over steps 1-4



courtesy L. Stadler

Reconstructing the real space structure -

Gerchberg & Saxton (1972)

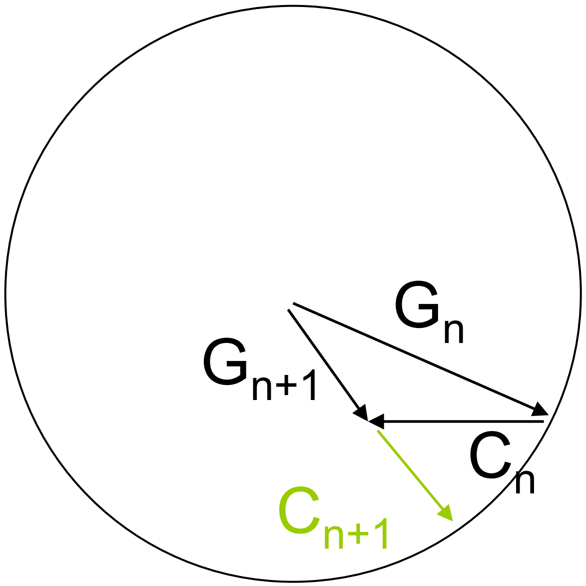
Determination of the phases for given amplitudes in real and Fourier space ($|f(x)|$ and $|F(k)|$, respectively):

0. Adding random phases to the given amplitudes in real space gives an estimate $g_0(x)$.
1. Adjust amplitudes, i.e., $g_1(x)=g_0(x)\,|f(x)|/|g_0(x)|$.
2. FT into Fourier space => $G_1(k)$
3. Adjust amplitudes, i.e., $G_2(k)=G_1(k)\,|F(k)|/|G_1(k)|$.
4. FT back into real space => $g_2(x)$.

By **iterating steps 1 to 4** the estimate $g(x)$ approximates $f(x)$ better and better.

Graphical Illustration

Fourier space

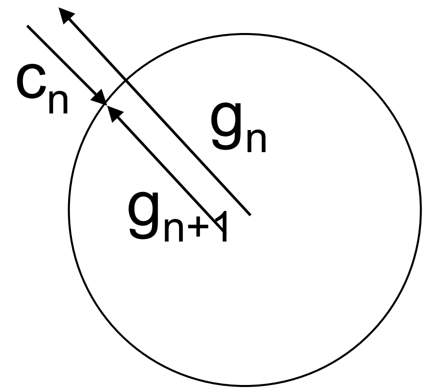


the error

$\Sigma_j\ |c_j|^2$

decreases mono-tonically!

real space



$g_{n+1}=g_n+c_n$

<u>Triangle inequality:</u>

$|G_n| \leq |G_{n+1}| + |C_n|$

$|G_n| - |G_{n+1}|=|C_{n+1}| \leq |C_n|$

Parseval's theorem:

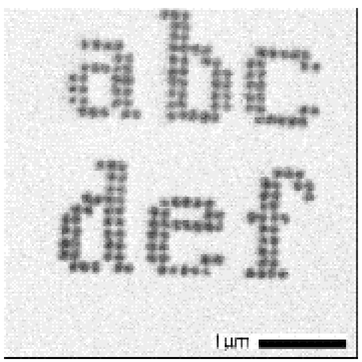
$\Sigma_j|c_{n,j}|^2 = \Sigma_j|C_{n,j}|^2$

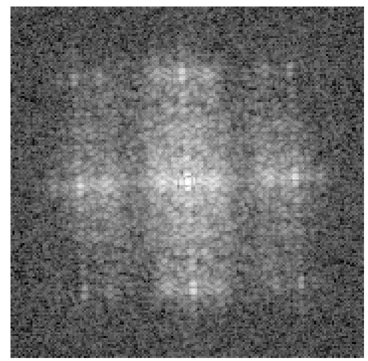
$\Rightarrow \Sigma_j|c_{(n+1),j}|^2 \leq \Sigma_j|c_{(n),j}|2$

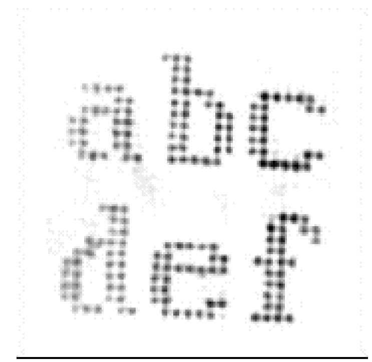
g_n is obtained by FT of G_n, which has the correct amplitude in Fourier space (compare step 3 on the previous slide)

Reconstruction of „oversampled“ data (1)

Reconstruction (phasing) of a speckle pattern: **“oversampling” technique**







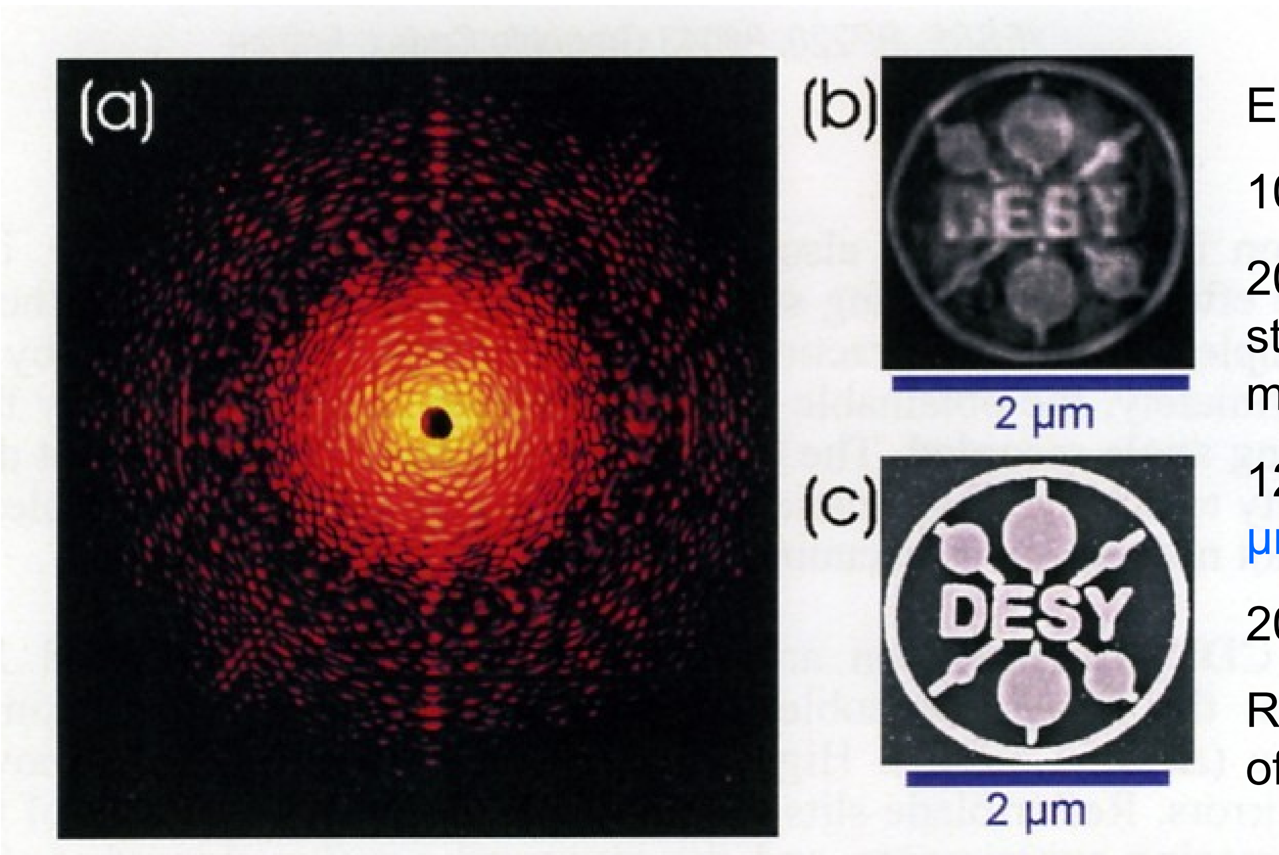
gold dots on SiN membrane (0.1 μm diameter, 80 nm thick)

λ=17Å coherent beam at X1A (NSLS), $1.3 \cdot 10^9$ ph/s 10μm pinhole 24 μm x 24 μm pixel CCD

reconstruction “oversampling” technique

Miao, Charalambous, Kirz, Sayre, Nature, 400, July 1999

Reconstruction of “oversampled” data (2)



E= 8 keV ID10C (ESRF}

10x10 µm^2 beam, L=3.3m

200 nm height gold structures on 50nm Si_3N_4 membrane (c: SEM)

1242x1152 pixel, 22.5 µm^2 pixel CCD in 3.29 m

200 x 3 s exposure (a)

Reconstruction (average of 4 best runs) (b)

oversampling ratio σ = object image / pixelsize = (λ•L/d)/pixel ≈ 10

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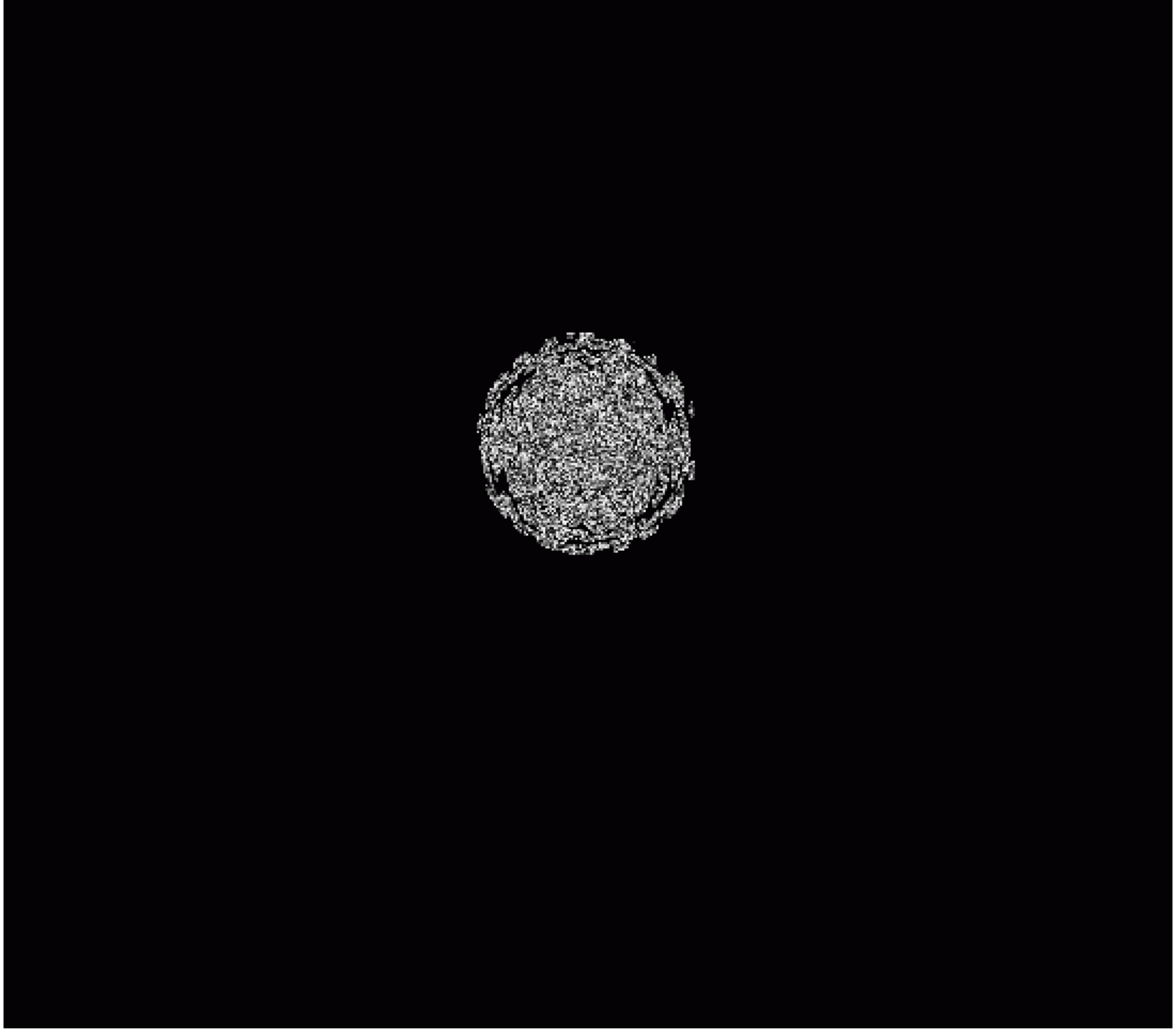
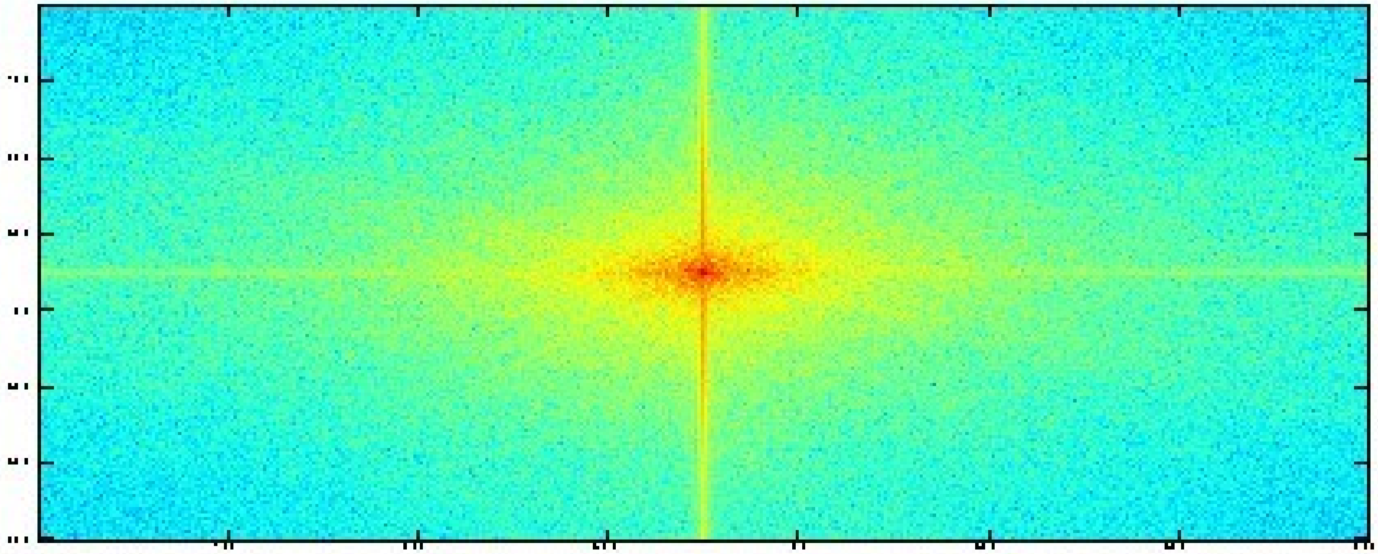
Reconstruction of oversampled data (2)



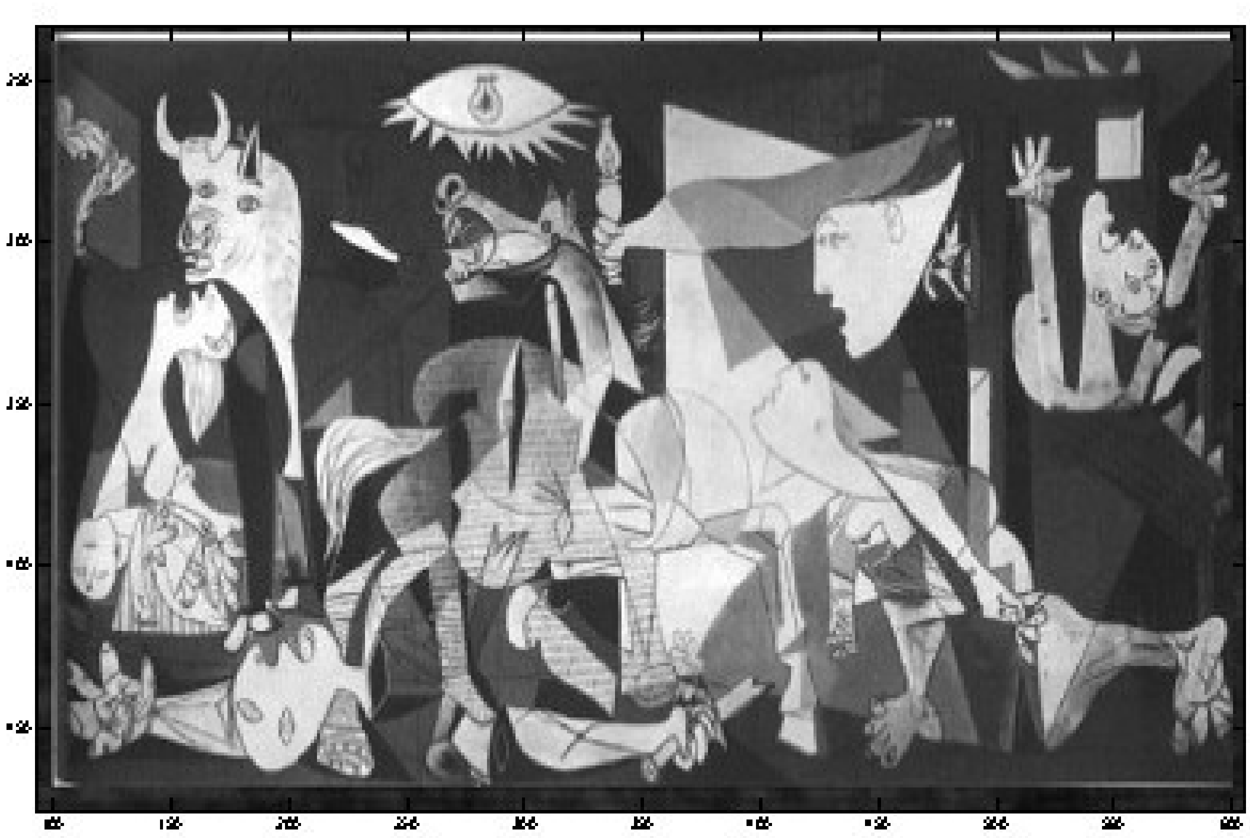
Reconstruction of “oversampled” data (3)

an “unknown” object

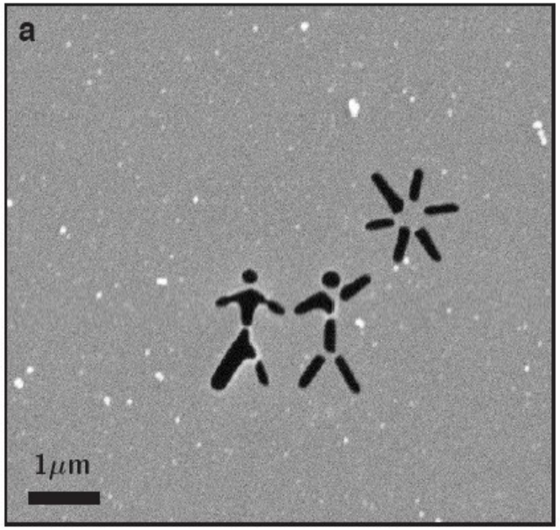


Reconstruction of “oversampled” data (3)

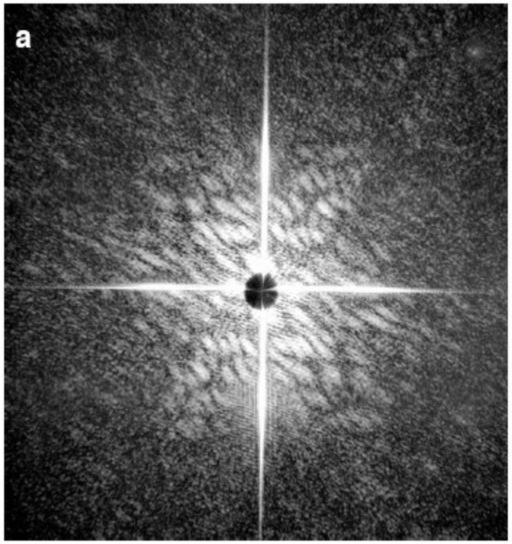
and its reconstruction



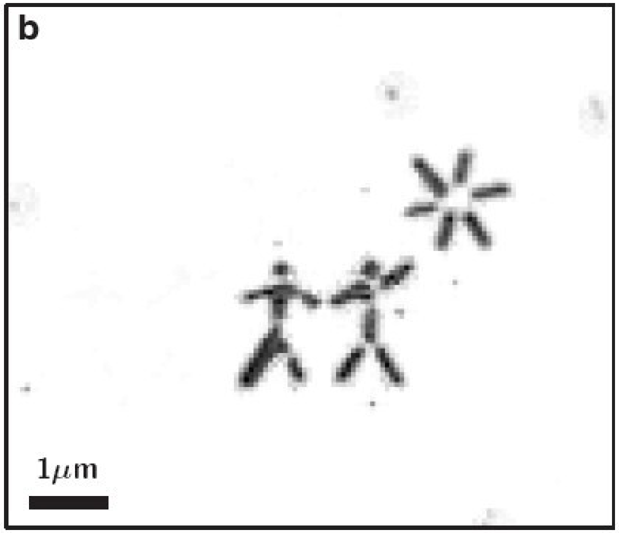
Reconstruction of “oversampled” data (4)



Model structure in 20 nm SiN membrane

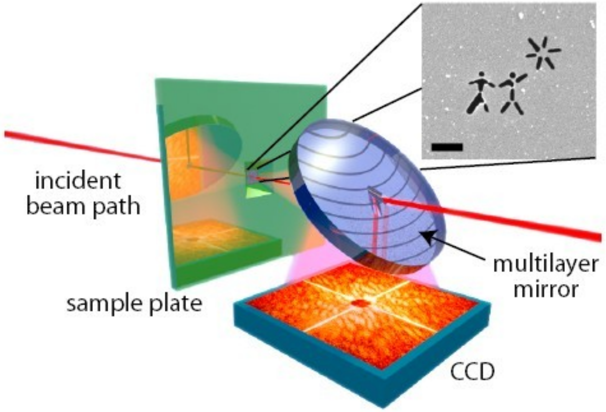


Speckle pattern recorded with a single (25 fs) pulse



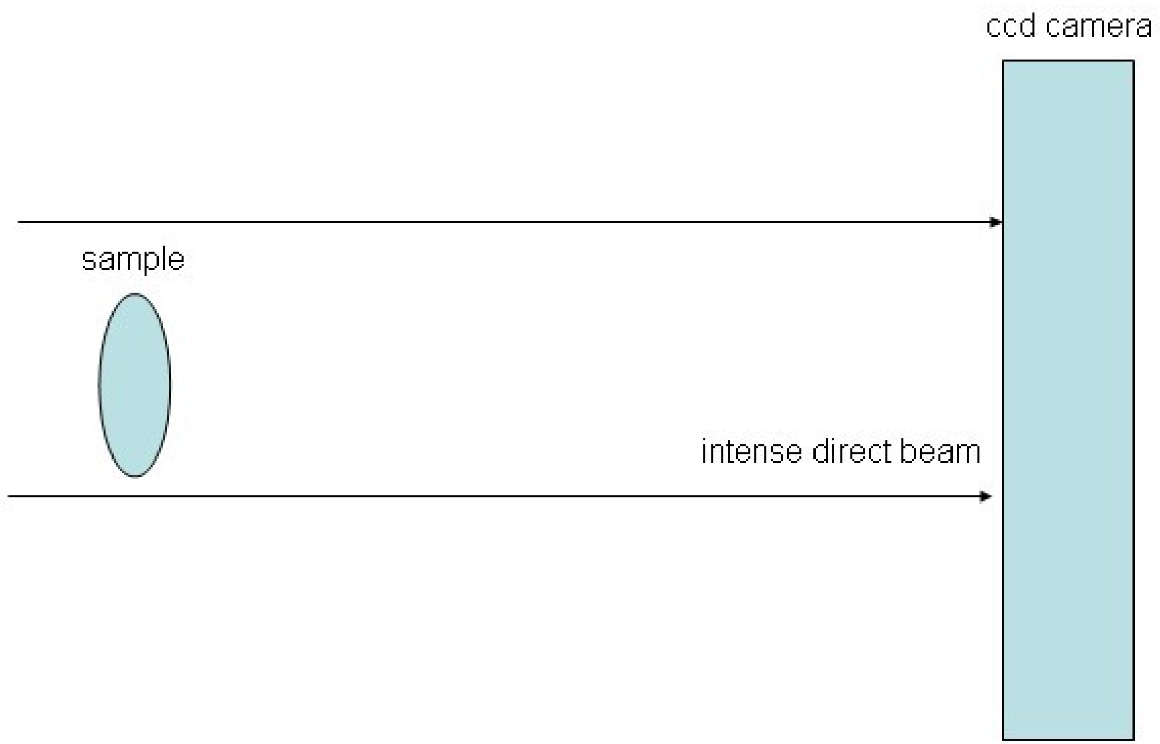
Reconstructed image

Incident FEL pulse:
25 fs, 32 nm,
4 x 10^{14} W cm^{-2} (10^{12} ph/pulse)

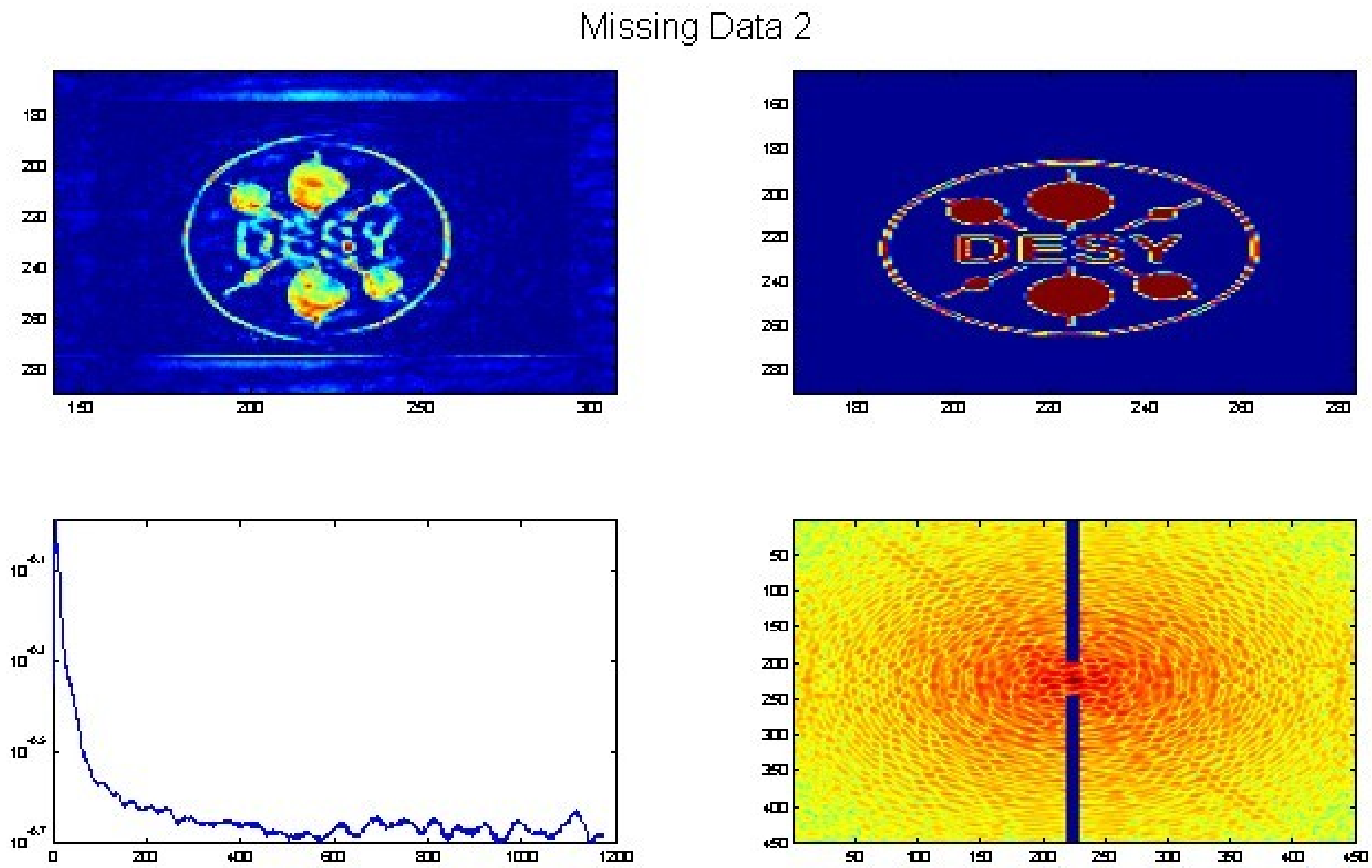


H. Chapman et al., Nature Physics, 2,839 (2006)

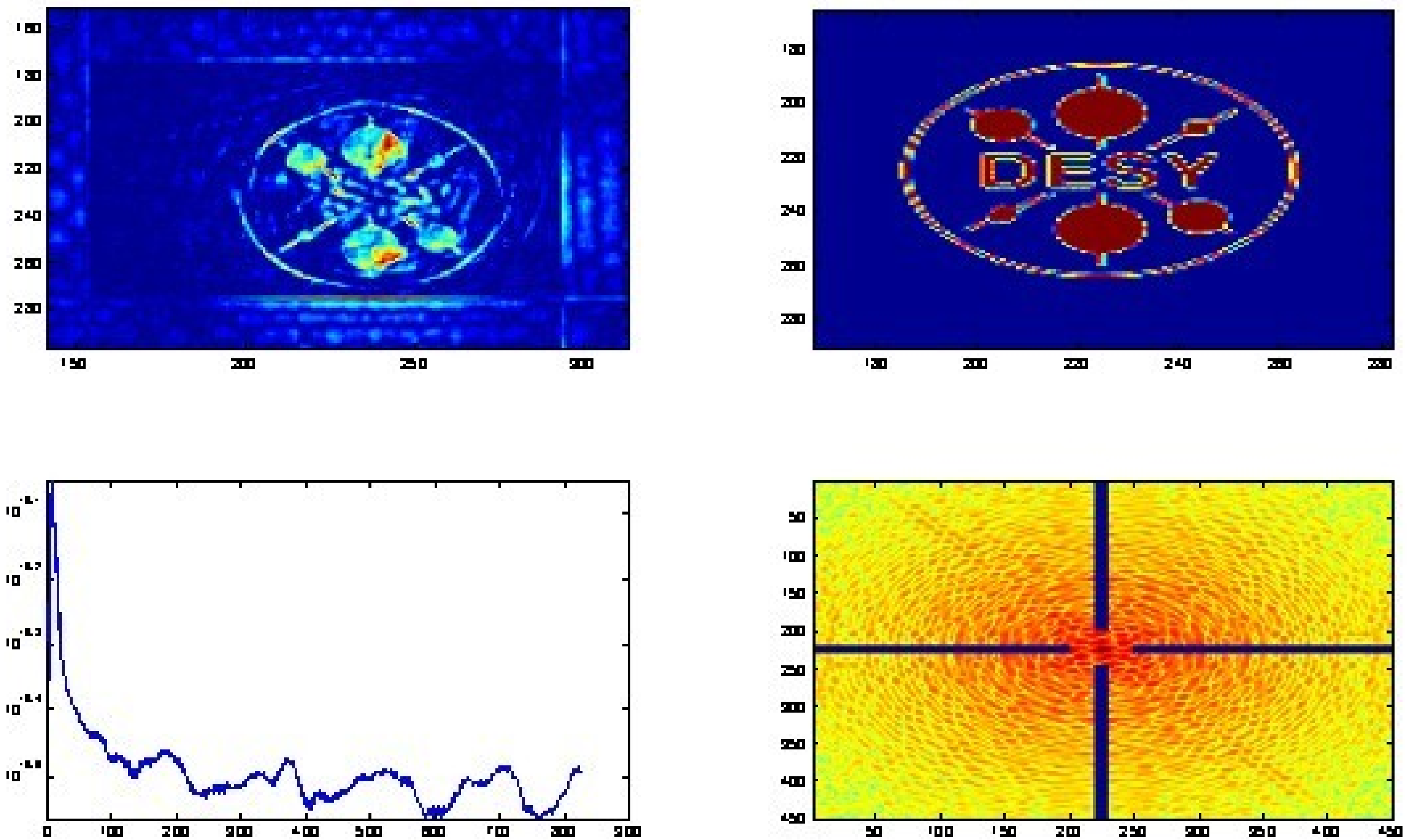
The beamstop problem



Missing Data (1)



- # Missing Data (2)



How to solve the missing data problem

(i) take a low resolution picture of the sample, calculate x-ray intensities and paste the corresponding near forward data into the beamstop area
Miao et al. Nature 400, 342 (1999)

(ii) algorithm approach: calculate F also for the missing pixels and rescale electron densities

Nishino, Miao, Ishikawa, PRB 68,220101 (2003)

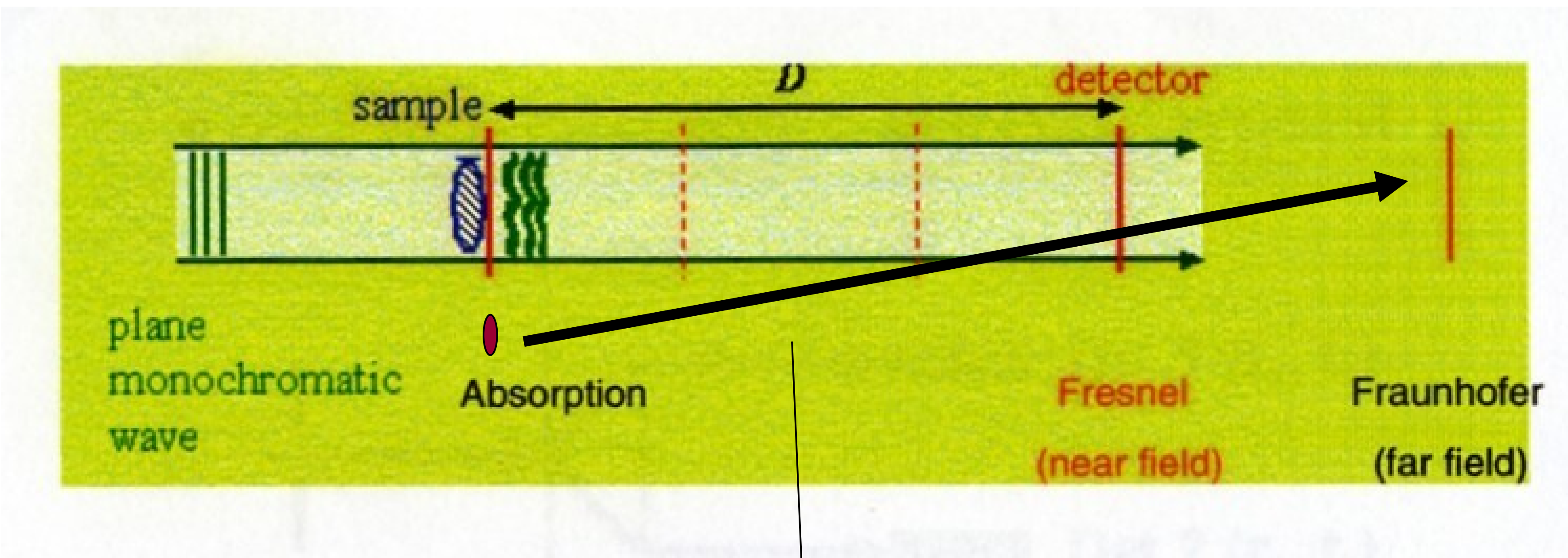
(iii) algorithm approach: use calculated autocorrelation functions as object constraints and update them during the algorithm

S. Marchesini, Chapman et al. PRB 68, 140101

Imaging techniques:

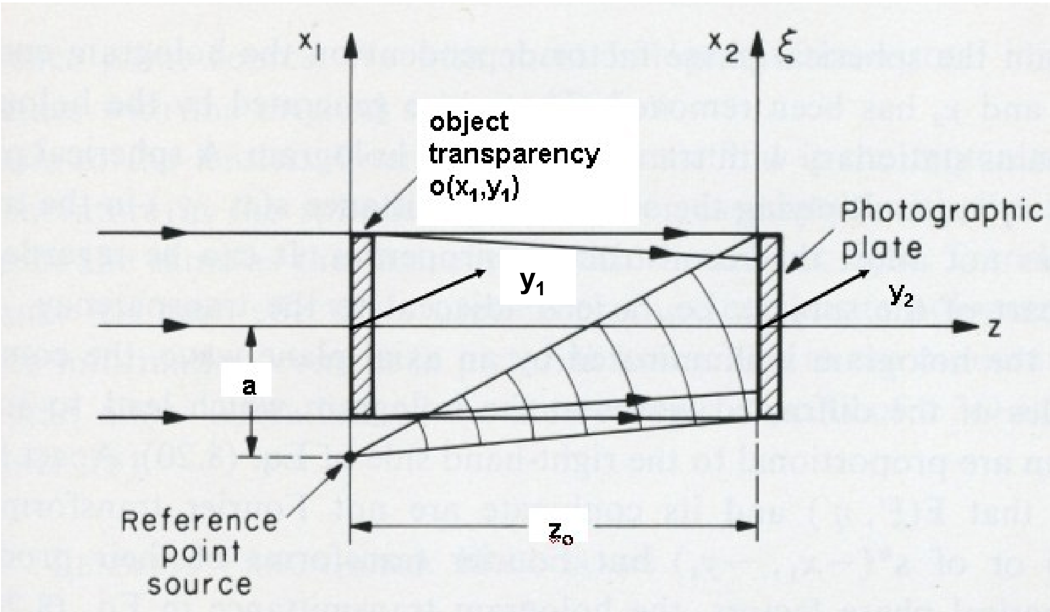
Fourier Transform Holography

- # Imaging techniques:



reference beam: holography

Fourier Transform Holography – FTH (1)



$o(x_1,y_1)$: amplitude of the wave transmitted through object o

$r(x_1,y_1)$: reference wave

R.J. Collier, C.B. Burckhardt, L.H. Lin "Optical Holography", Academic Press (1971)

Fresnel Kirchhoff Theory

$o(x_2,y_2) = (i/\lambda z_o)\exp\{i\pi/\lambda z_o)(x_2^2+y_2^2)\}\ \bar{O}(\xi,\eta)$

$r(x_2,y_2) = (i/\lambda z_o)\exp\{i\pi/\lambda z_o)(x_2^2+y_2^2)\}\ \check{R}(\xi,\eta)\ \exp\{-2i\pi\xi a\}$

with $\bar{O}(\xi,\eta) = \mathrm{FT}\,\{o(x_1,y_1)\}$, $\check{R}(\xi,\eta) = \mathrm{FT}\,\{r(x_1,y_1)\}$, $\xi=x_2/\lambda z_o$, $\eta=y_2/\lambda z_o$

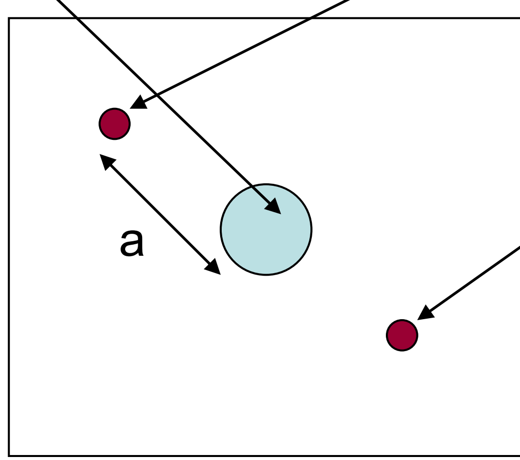
Fourier Transform Holography (2)

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

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

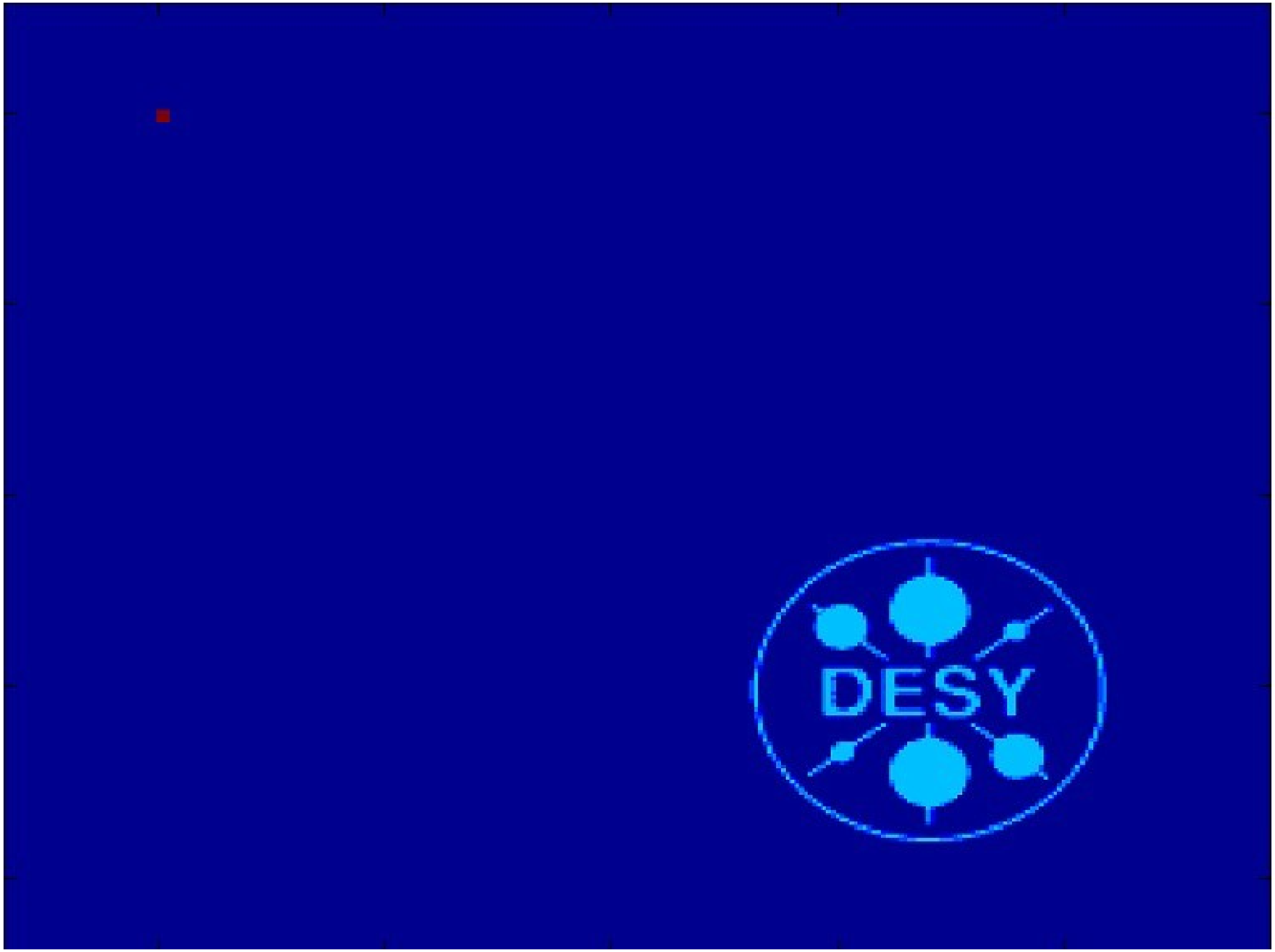
$$I(x_2,y_2) \approx \underbrace{|\check{R}(\xi,\eta)|^2}_{} + |\bar{O}(\xi,\eta)|^2 + \underbrace{\check{R}^*(\xi,\eta)\, \bar{O}(\xi,\eta)\, e^{i\pi a\xi} + \check{R}(\xi,\eta)\, \bar{O}^*(\xi,\eta)\, e^{-i\pi a\xi}}_{}$$

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

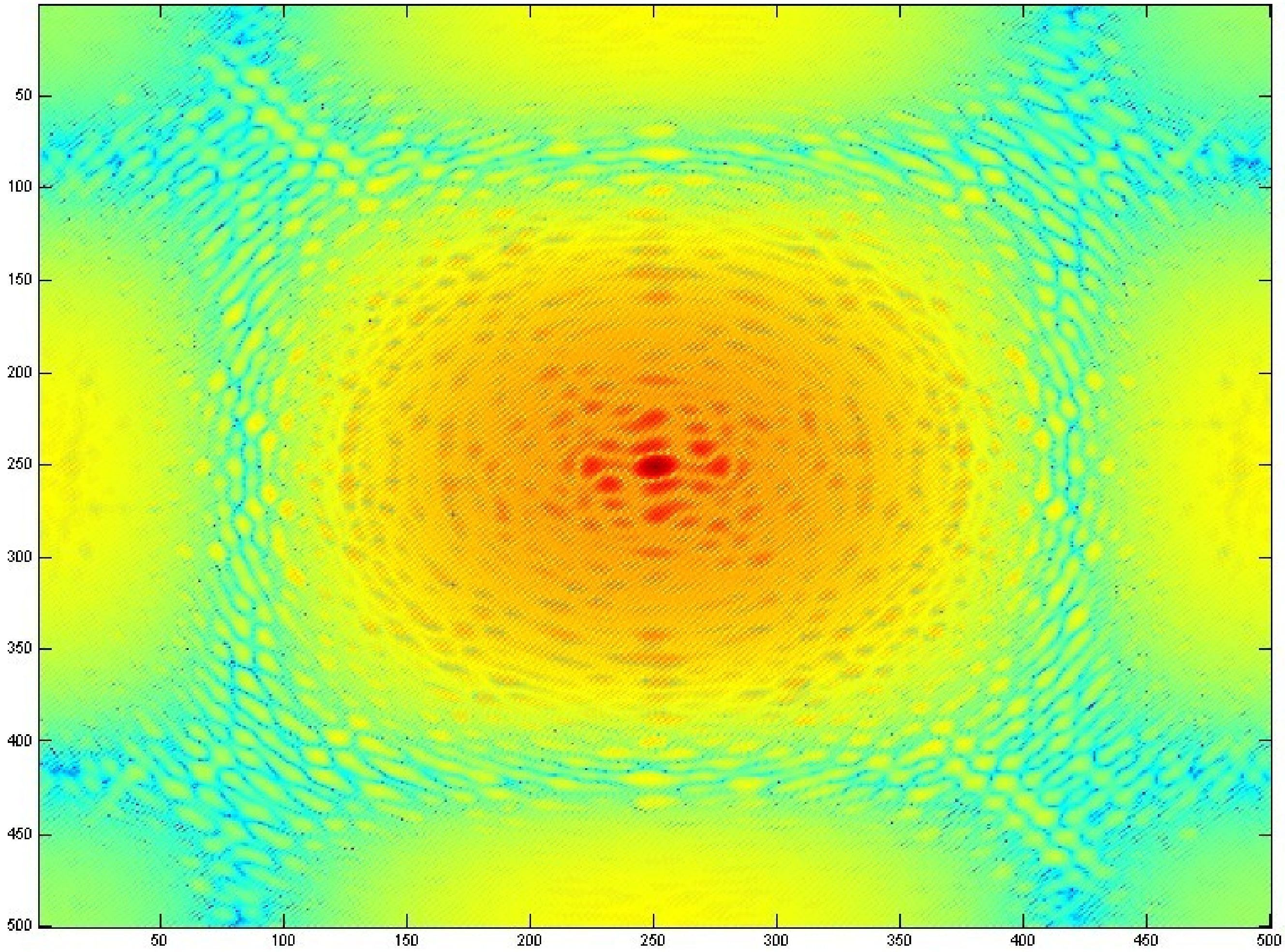


note: resolution determined by size of reference aperture

- # FTH (3): small reference hole



FTH (4)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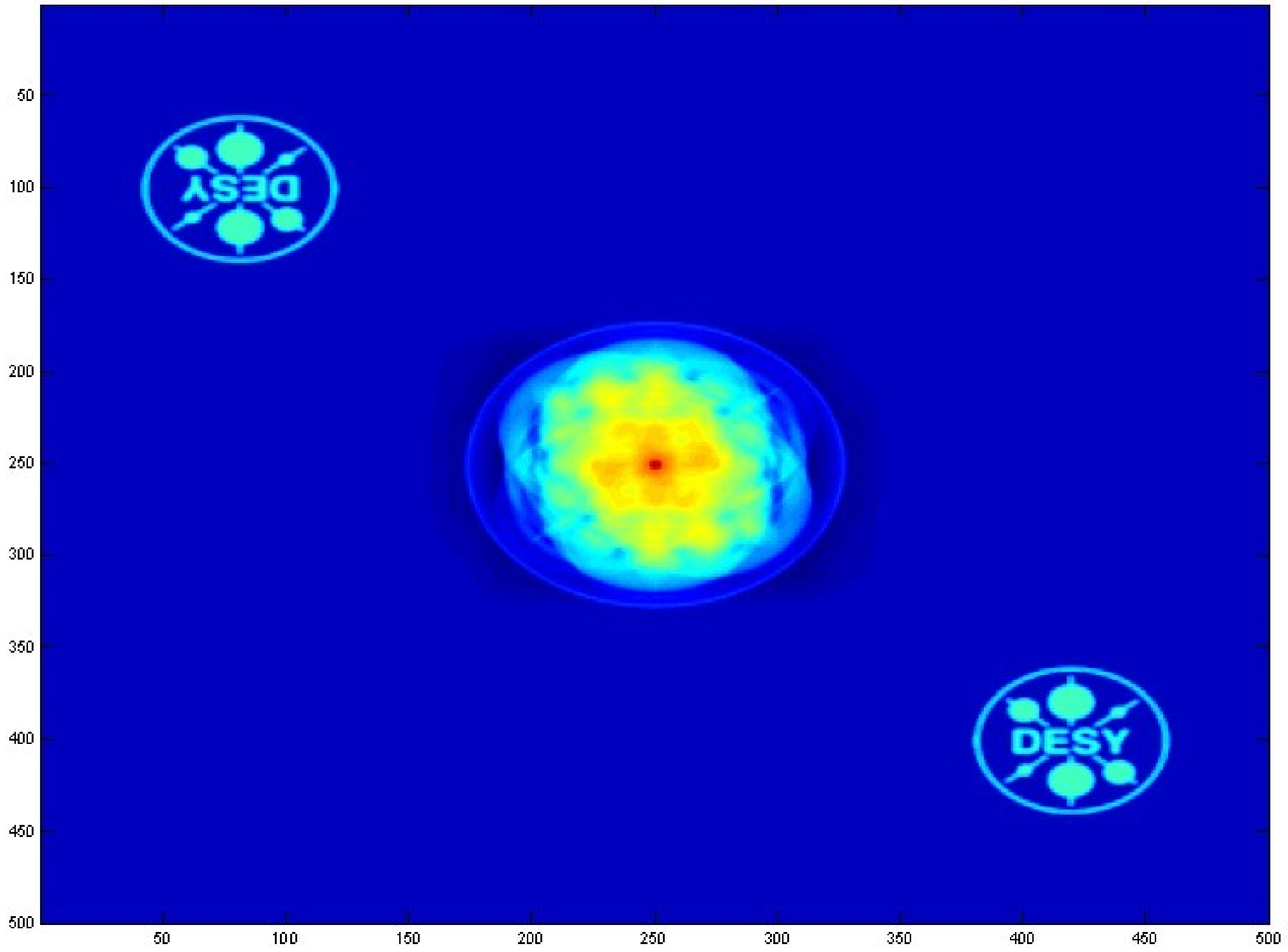
FTH (5)



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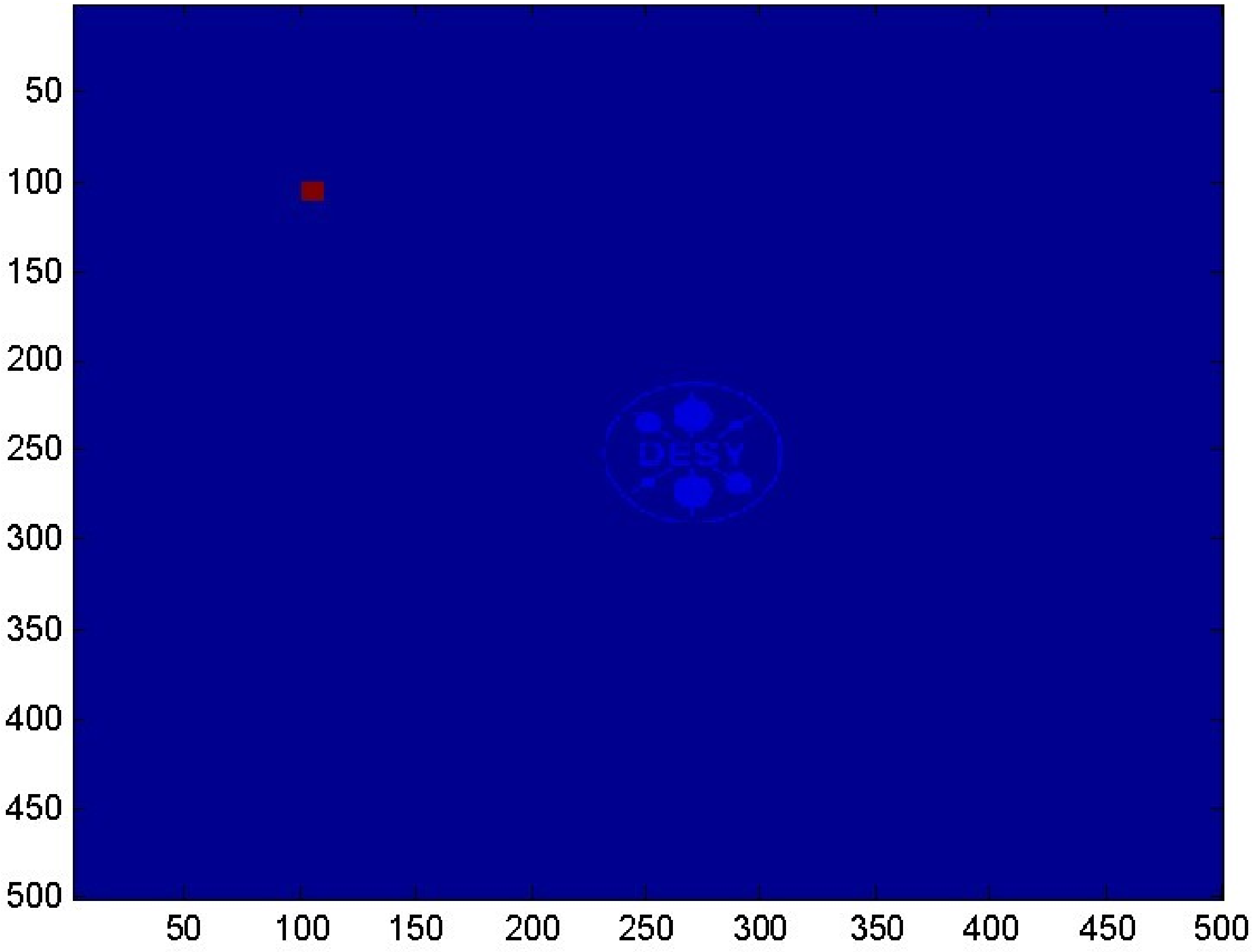
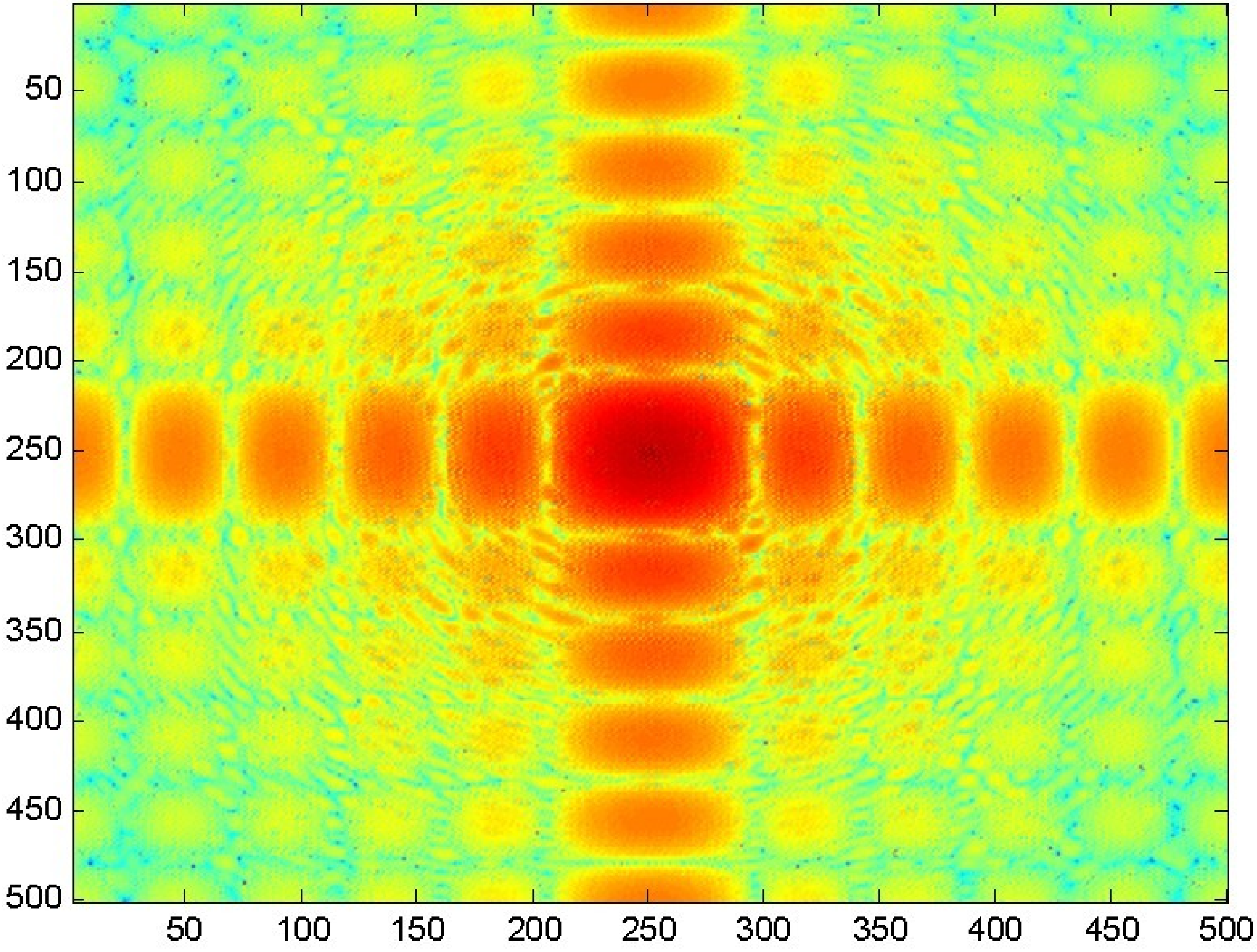
FTH (6): large reference hole



- # FTH (7)



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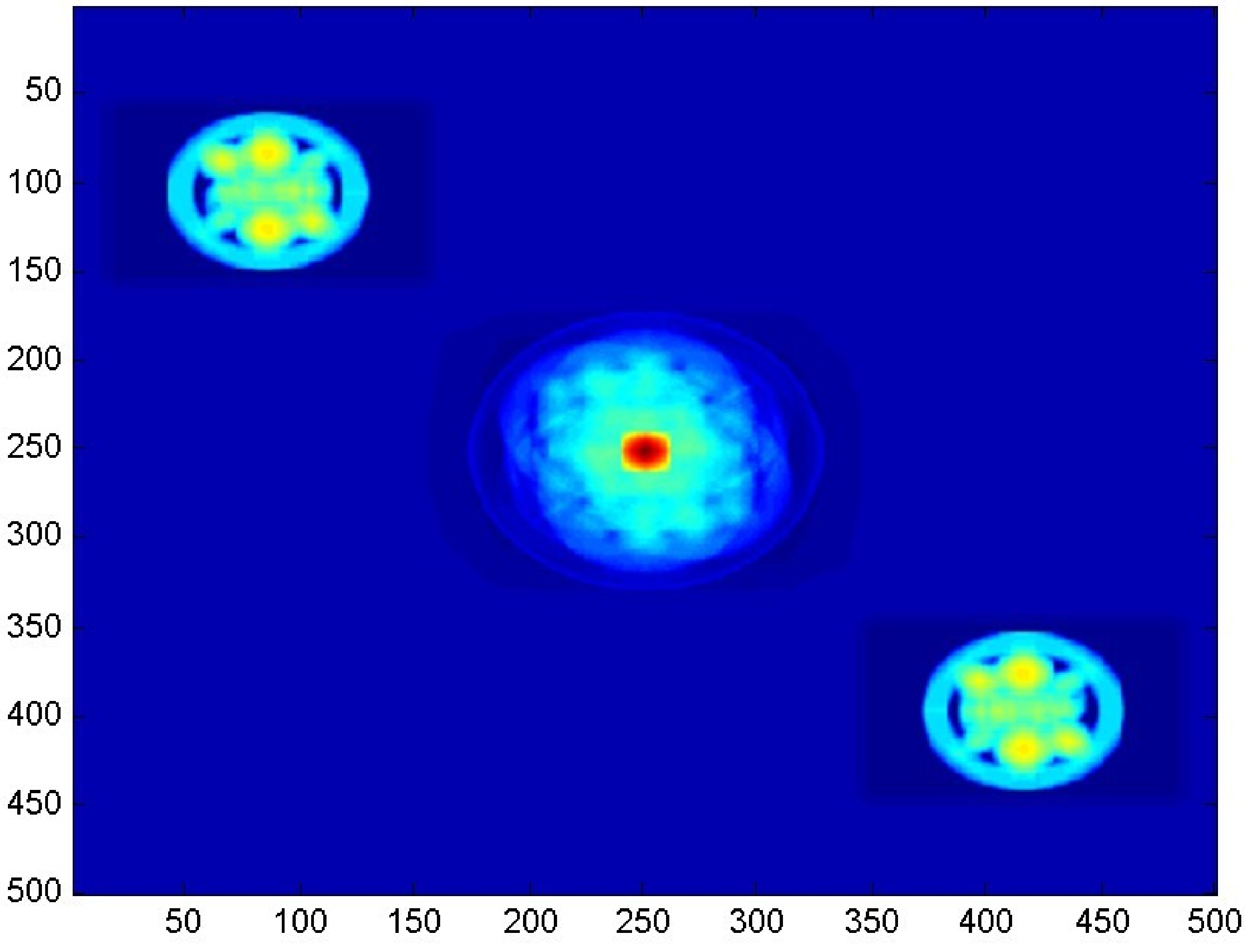
- FTH (8)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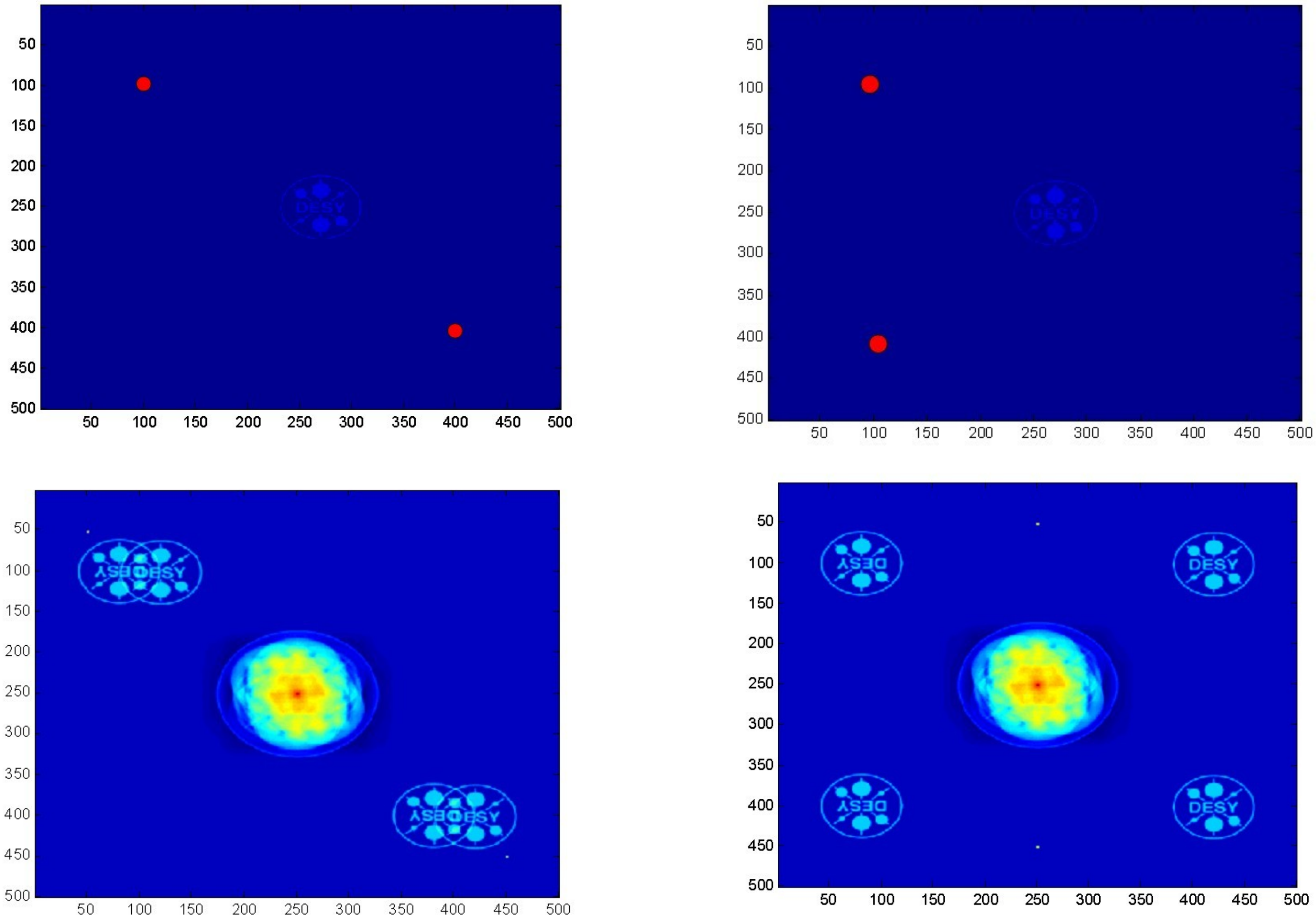
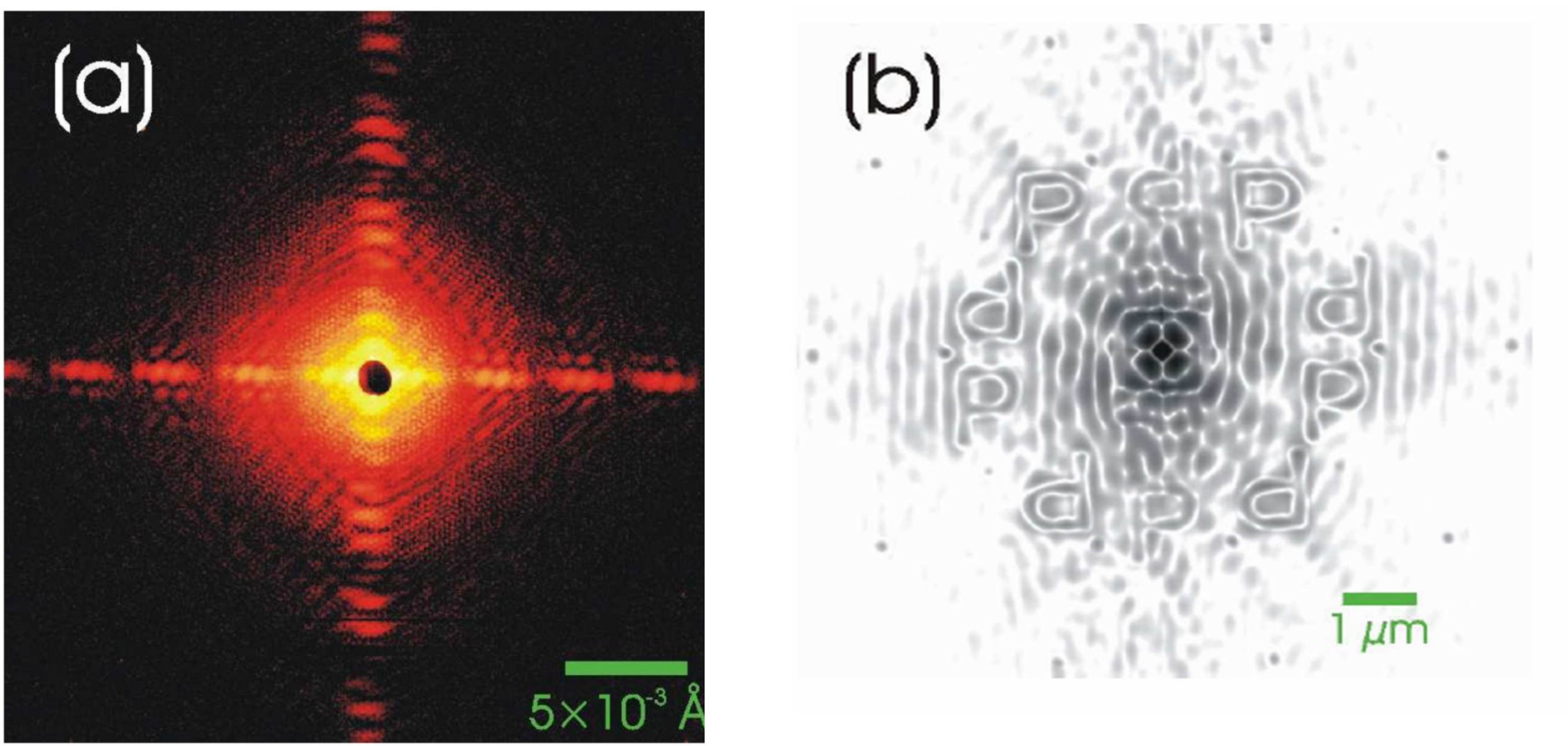
FTH (9): more than one reference hole

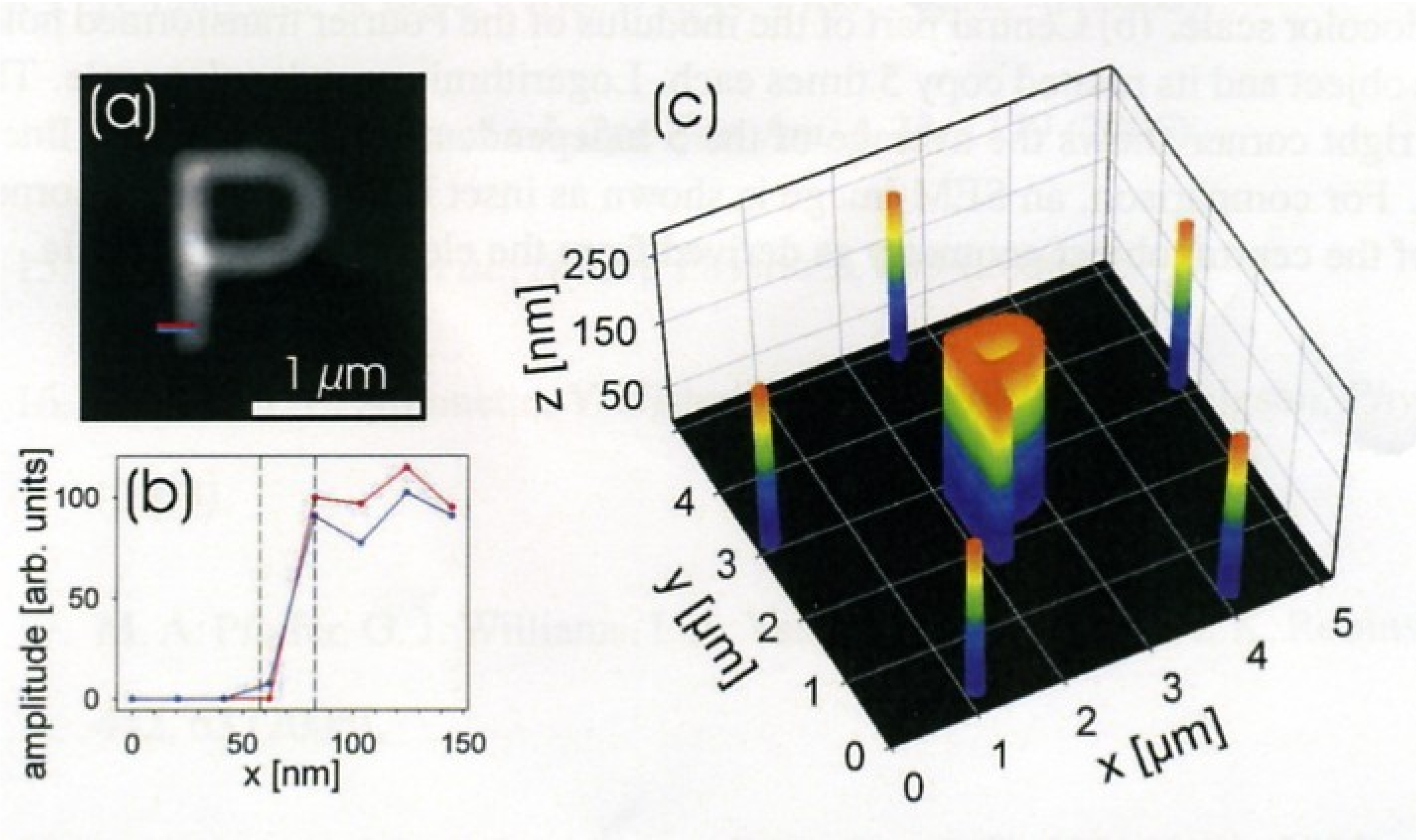


FTH (10)

Au reference structure (letter P): letter elements 200 nm in width and 220 nm heigh on 50 nm Si_3N_4-membrane. 5 Au reference dots of 175 nm diameter and 220 nm height on a circle of 2.5 μm around the sample. 200x 3s exposures (≈$1.4x10^8$ ph/s through $10x10μm^2$ at 8 keV.



FTH (10):



(a) average of 100 phase retrieval runs
(b) slices yielding a resolution of ≈ 25 nm
(c) visualization of the object as determined from the electron density profile