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

Part II Script 3

Vorlesung im GrK 1355

WS 2013

A. Hemmerich & G. Grübel

Location: SemRm 052, Gebäude 69, Bahrenfeld

Thursdays 12.15 – 13.45

G.Grübel (GR), A.Hemmerich (HE)

Literature

Basic concepts: The quantum theory of light
Rodney Loudon, Oxford University Press (1990)
Quantum Optics
Marlan O. Scully, M. Suhal Zubairy, Cambridge University Press (1997)
Dynamic Light Scattering with Applications
B.J. Berne and R. Pecora, John Wiley&Sons (1976)
Elements of Modern X-Ray Physics
J. A. Nielsen and D. McMorrow, J. Wiley&Sons (2001)

Matter Waves: Bose-Einstein Condensation in Dilute Gases
C. J. Pethick and H. Smith, Cambridge University Press (2002)

Lecture Notes

Part I: ***http://photon.physnet.uni-hamburg.de/fileadmin/user_upload/ILP/Hemmerich/teaching.html/Coherence.pdf***

Part II: **http://photon-science.desy.de/research/studentsteaching/lectures__seminars/ws_13_14/coherence_of_light_grk1355/…..**

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

Part II: G. Grübel

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

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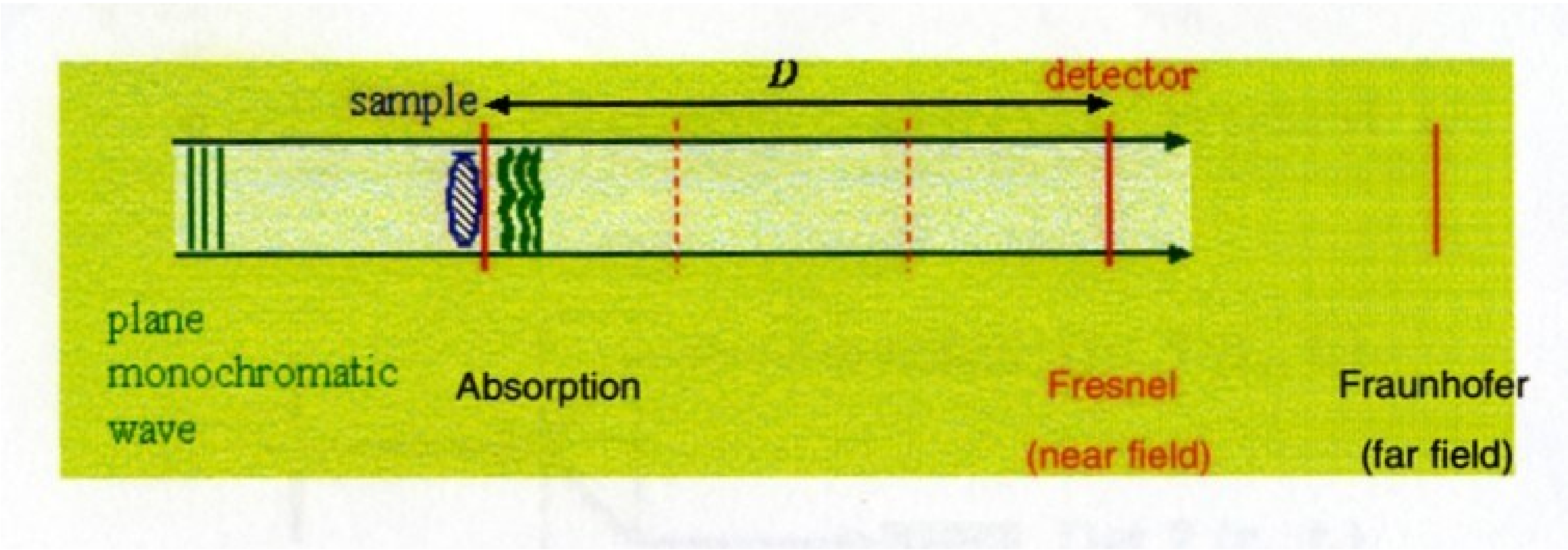
Imaging techniques:

Lensless Imaging, Fourier Transform Holography

Imaging techniques

Parallel beam, without lenses



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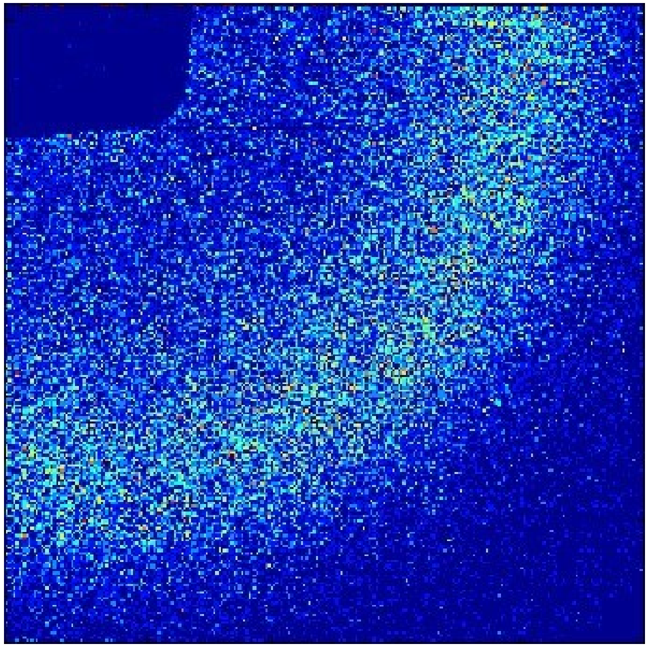
Introduction: Scattering with coherent X-rays

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.

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



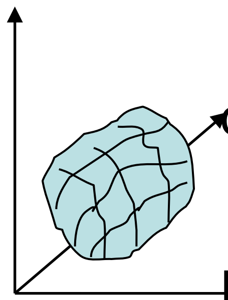
j in coherence volume $c= \xi_t^2 \xi_l$

<u>Incoherent Light:</u>

$S(Q,t) = < S_c(Q,t)>_{V>>c}$

ensemble average

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”

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

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

here (\$) defines a set of N equations 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)

note: *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

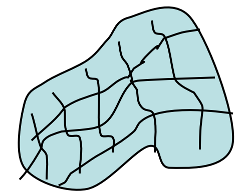
ii) ρ(x) real N variables

Friedel's law: I(**k**) = I(-**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:



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

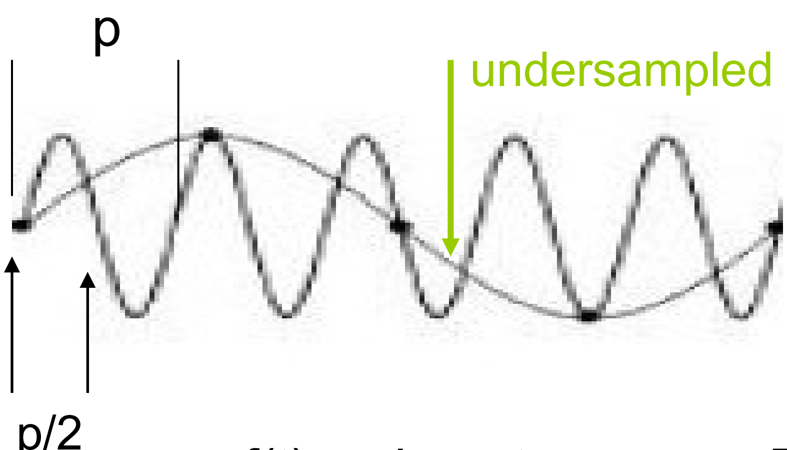
need: $\sigma > 2$ in 1D; $\sigma > 2^{1/2}$ in 2D; $\sigma > 2^{1/3}$ in 3D

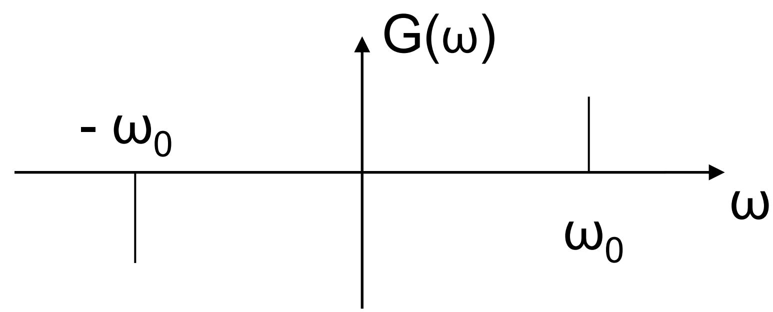
iii) apply iterative algorithms to retrieve phase

iv) resolution determined by maximum momentum transfer Q

Sampling Theory (1)

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



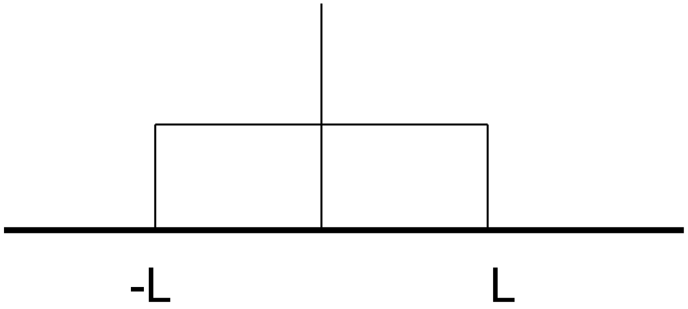


$$f(t) = \sin \omega_0 t \quad ===> \quad \text{FT} \quad <=== \quad G(\omega) = \int f(t)\, e(-i2\pi\omega t)\, dt$$

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 $f_{NY} = 2 f_{max}$ ($=2\omega_0$).

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

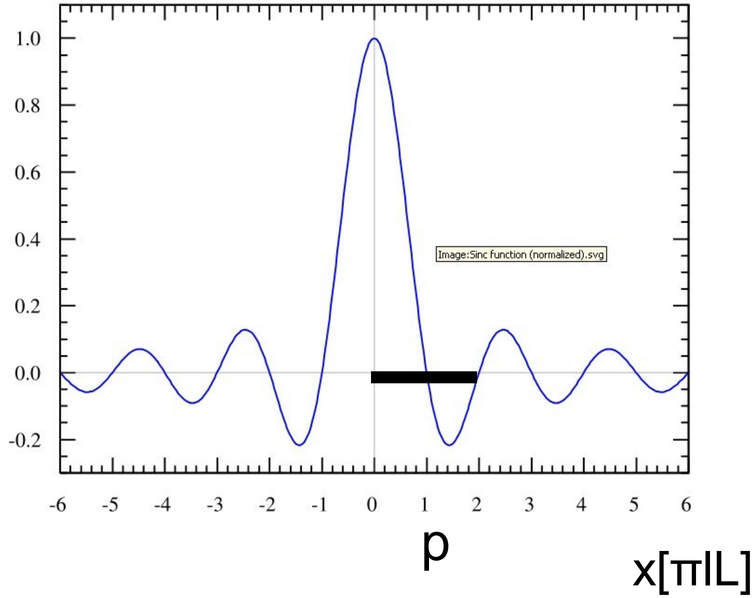
Sampling Theory (2)



consider z(t) = 1 |t| ≤ L
= 0 elsewhere

its FT is given by

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



period: $p = 2\pi/L$
max frequency: $f_{max} = 2\pi/p = L$
Nyquist frequency: $f_{Ny} = 2L = 4\pi/p$
sampling period: $p_{NY} = 2\pi/f_{NY} = p/2$

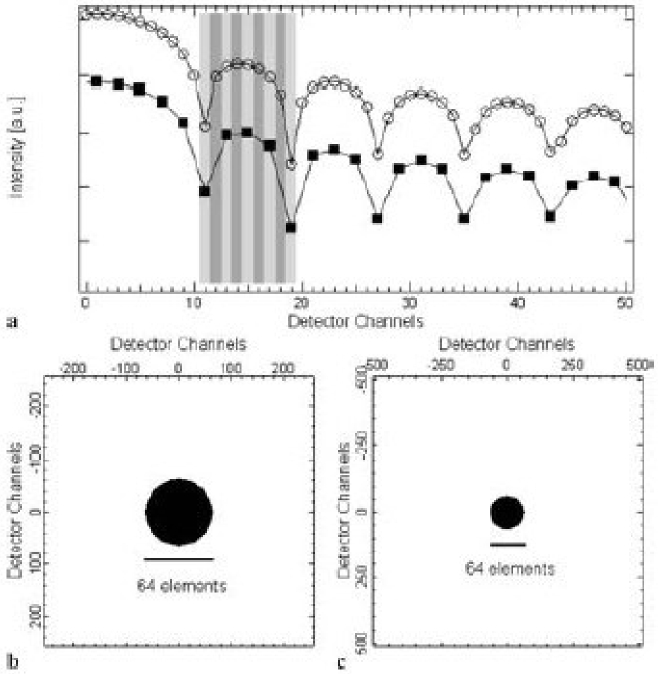
need to sample in fourierspace on a scale ≤ p/2

“oversampling” corresponds to sampling in “real” space on a scale ξ > 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: $I(q) = I_0 |\sin(qa)/qa|^2$



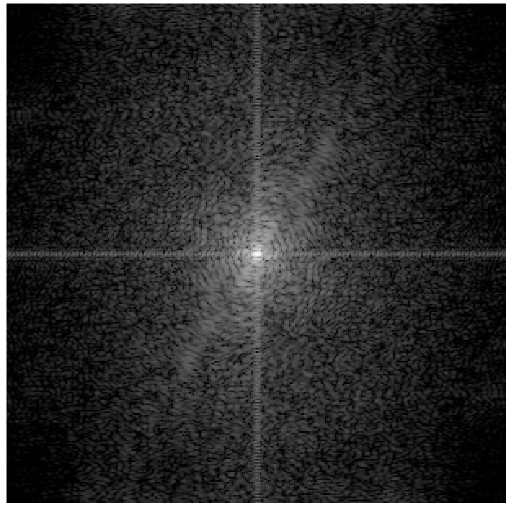
scattering from circular pinhole aperture with a linear oversamp-ling ratio of 4 (8)

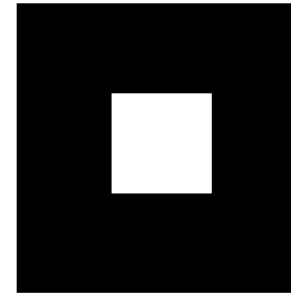
S. Eisebitt et al., Appl. Phys. A80(2005)921

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)

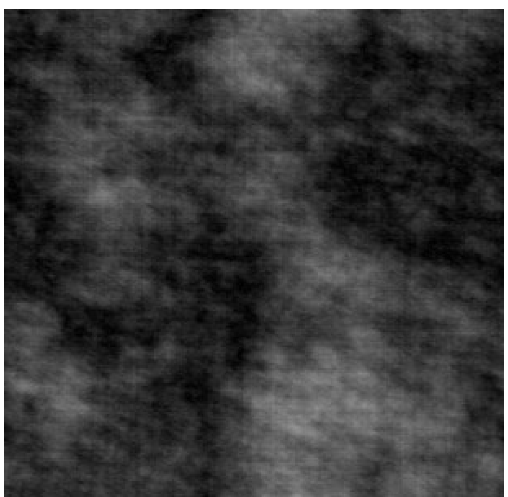




-1. Guess support in real space

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)

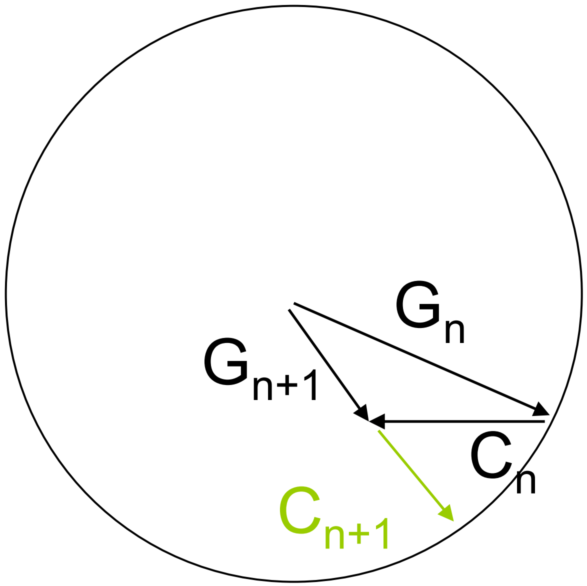
Determination of the phases for given amplitudes in real and Fourier space (|f(x)| and |F(k)|, respectively):

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

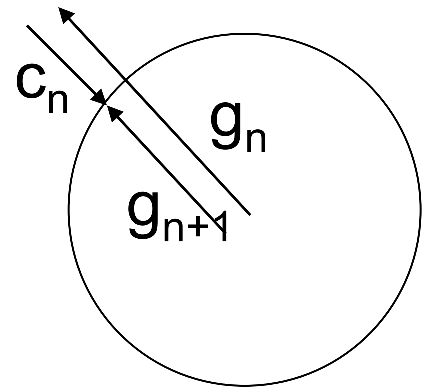


the error

$\Sigma_j |c_j|^2$

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:

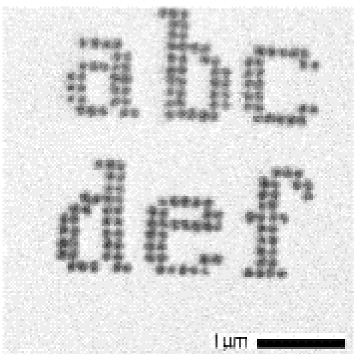
$\Sigma_j|c_{n,j}|^2 = \Sigma_j|C_{n,j}|^2$

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

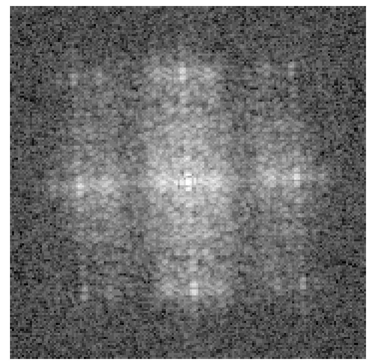
g_n is obtained by FT of G_n, which has the correct amplitude in Fourier space (compare step 3 on the previous slide)

Reconstruction of „oversampled“ data (1)

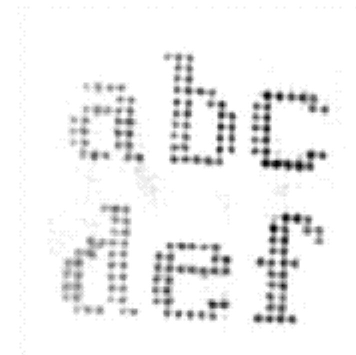
<u>Reconstruction (phasing) of a speckle pattern:</u> **“oversampling” technique**



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



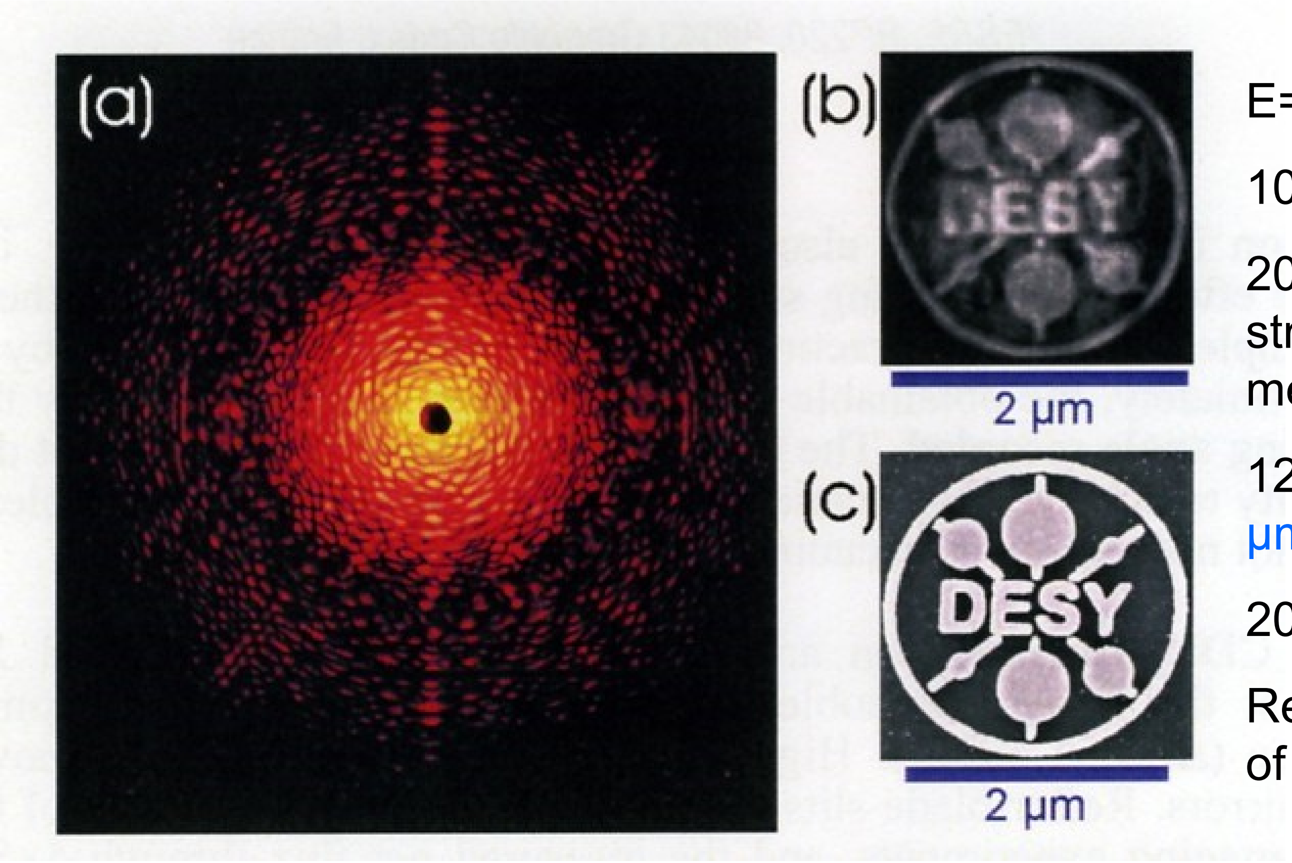
λ=17Å coherent beam at X1A (NSLS), $1.3 \cdot 10^9$ ph/s 10μm pinhole 24 μm x 24 μm pixel CCD



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)

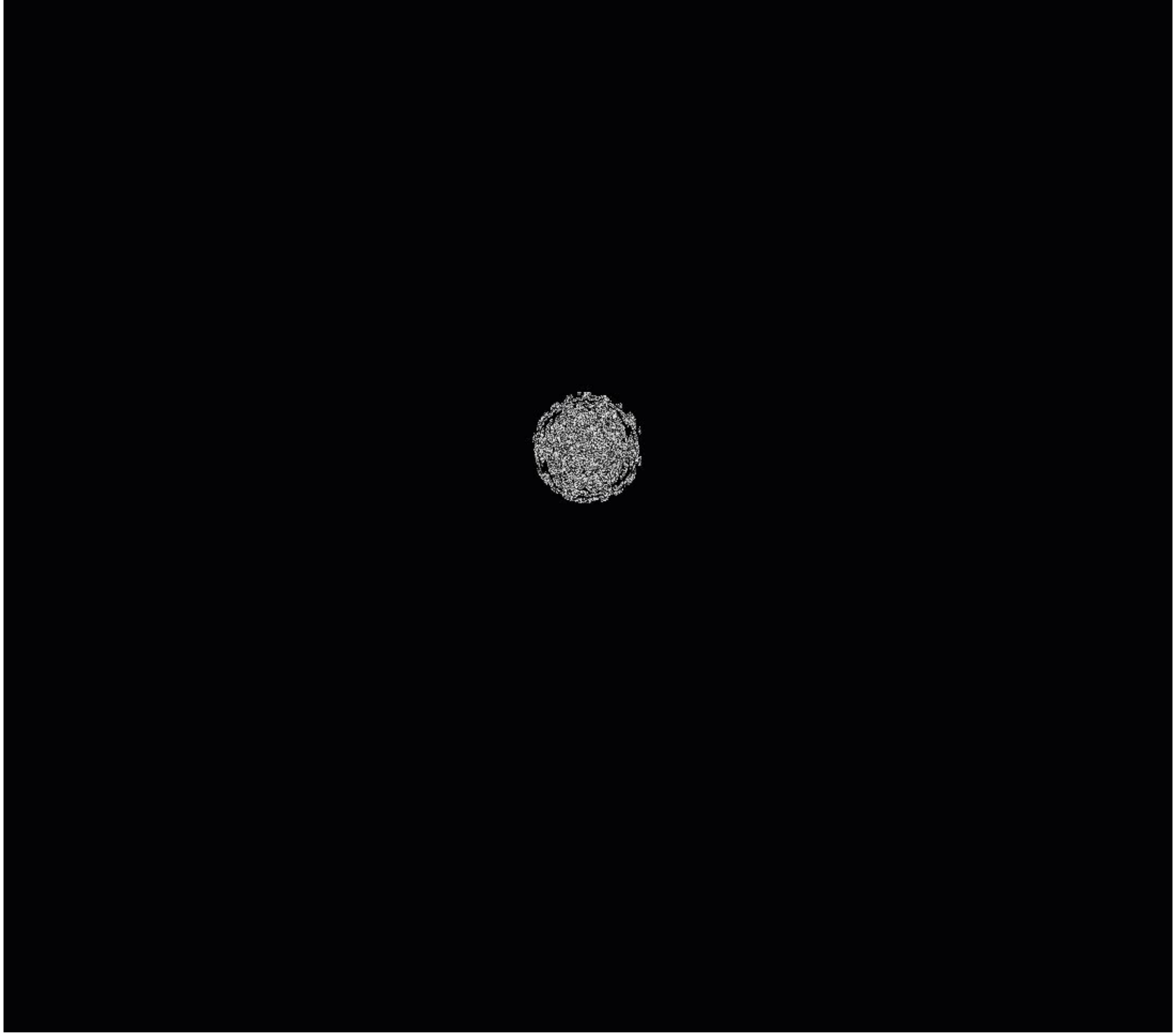
1242x1152 pixel, 22.5 µm^2 pixel CCD in 3.29 m

200 x 3 s exposure (a)

Reconstruction (average of 4 best runs) (b)

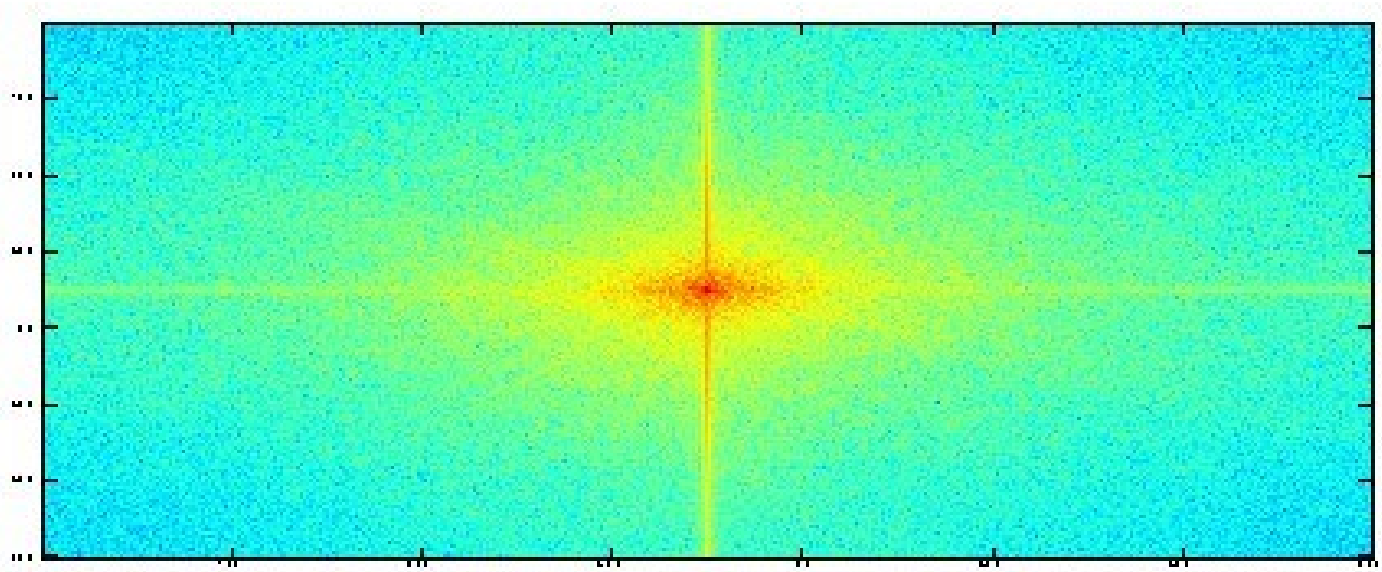
oversampling ratio σ = object image / pixelsize = (λ•L/d)/pixel ≈ 10

Reconstruction of oversampled data (2)



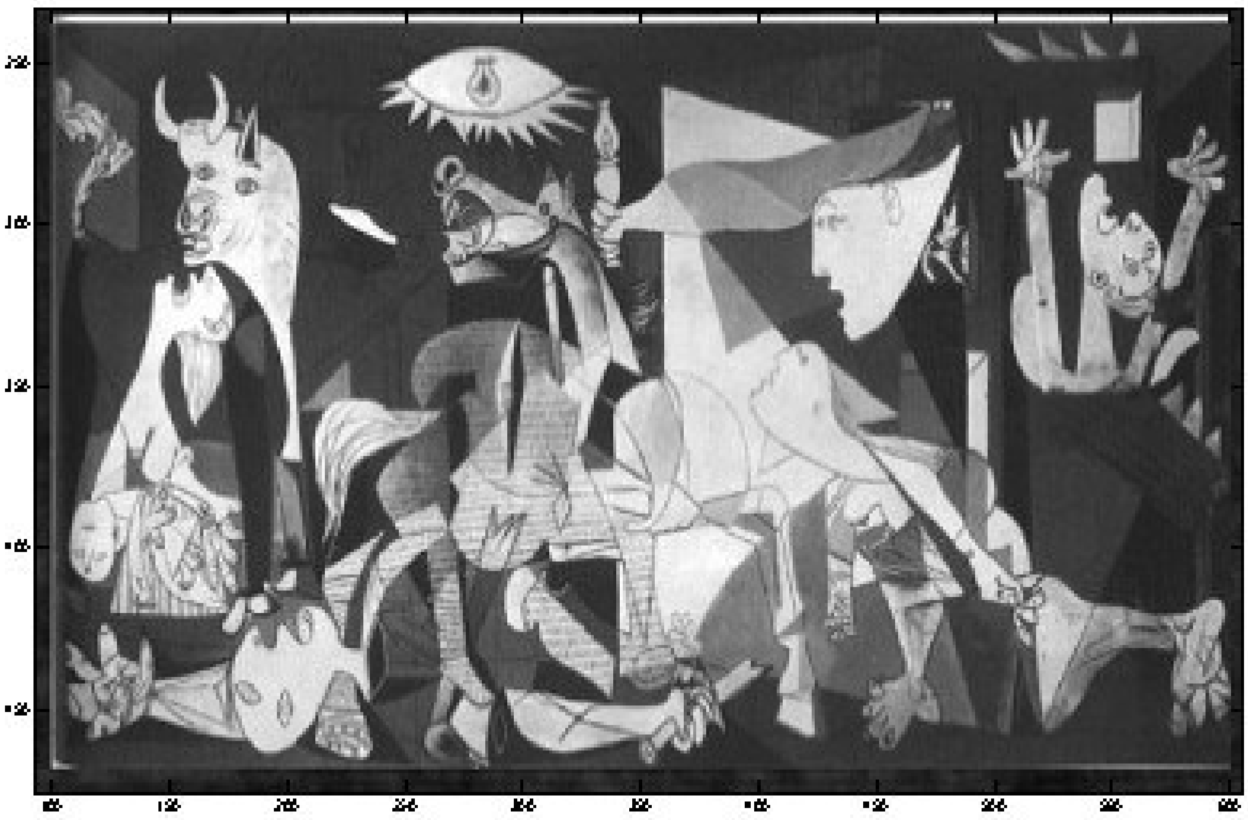
Reconstruction of “oversampled” data (3)

an “unknown” object

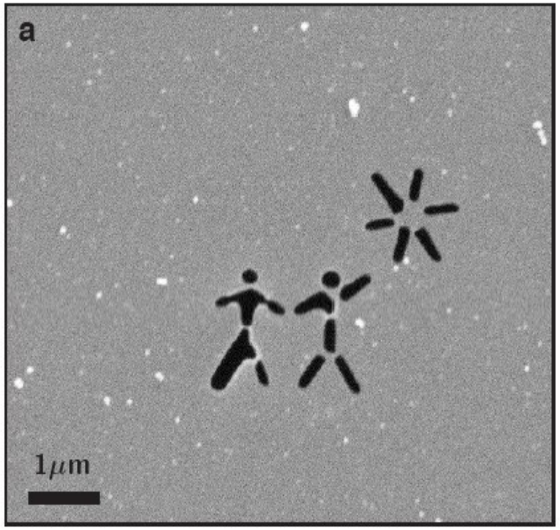


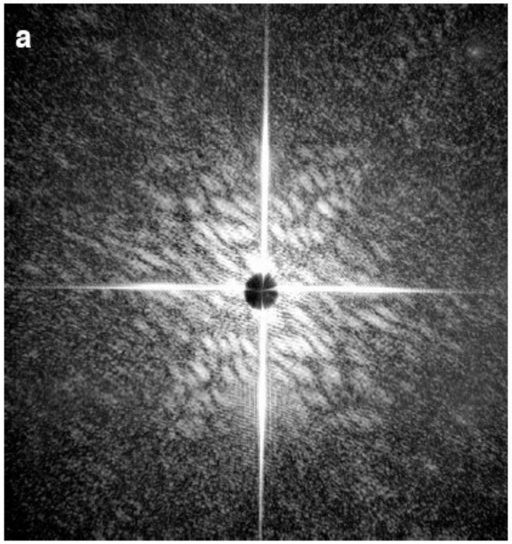
Reconstruction of “oversampled” data (3)

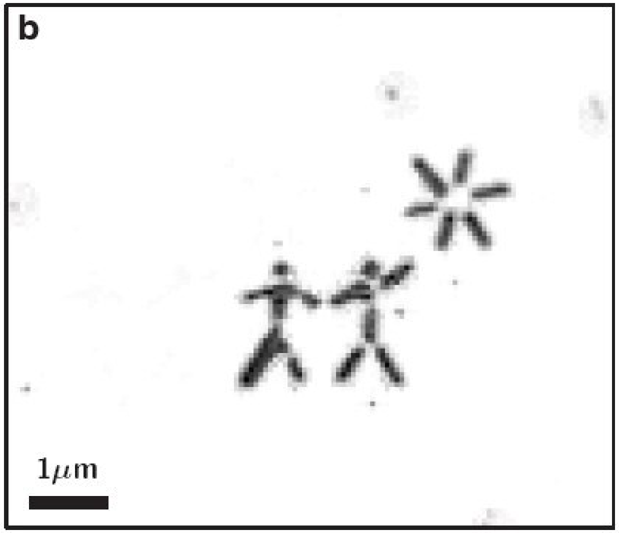
and its reconstruction



Reconstruction of “oversampled” data (4)





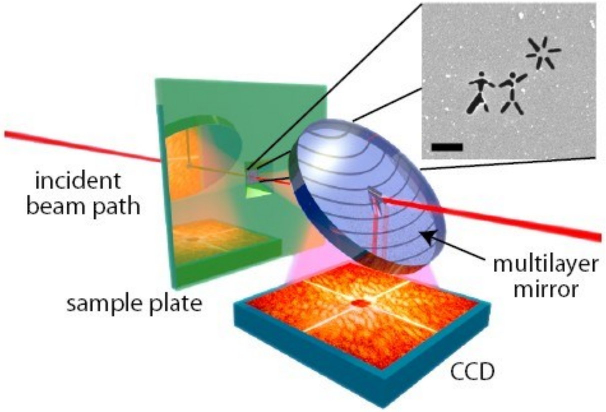


Model structure in 20 nm SiN membrane

Speckle pattern recorded with a single (25 fs) pulse

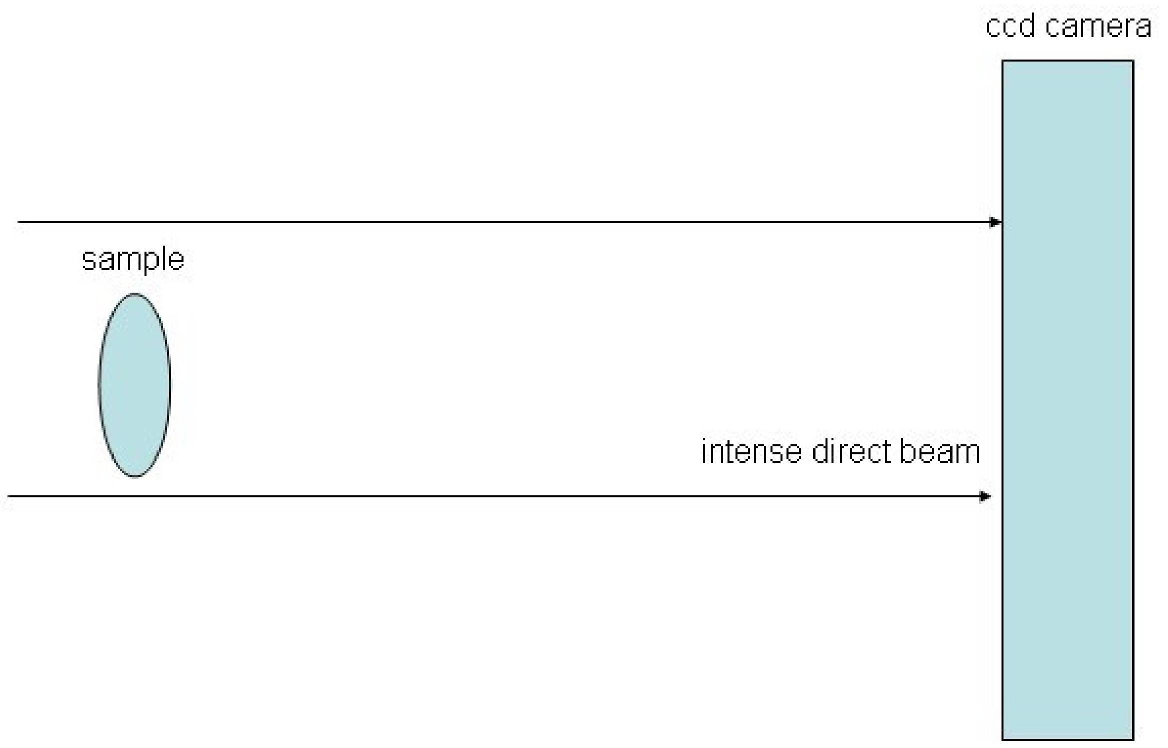
Reconstructed image

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

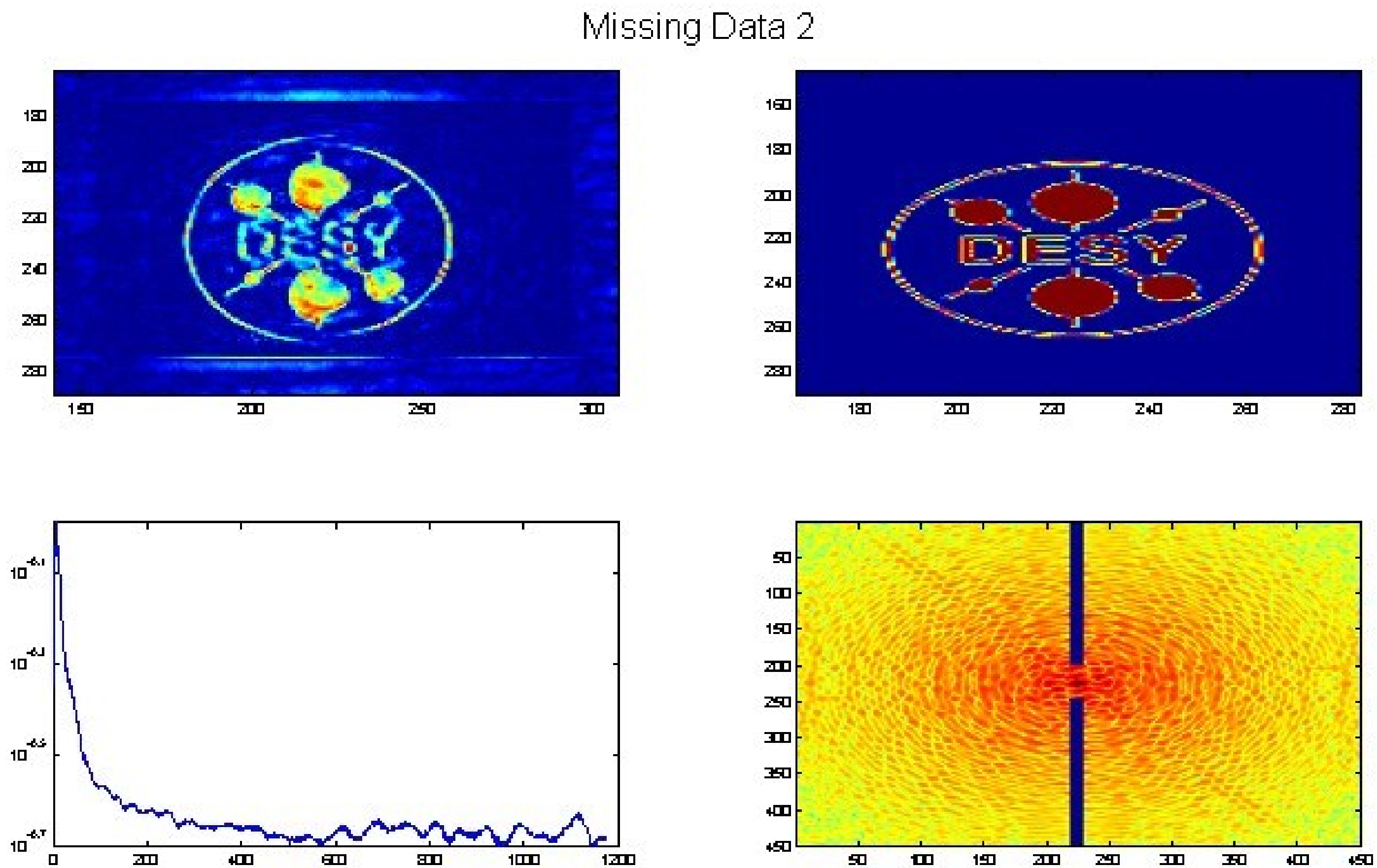


H. Chapman et al., Nature Physics, 2,839 (2006)

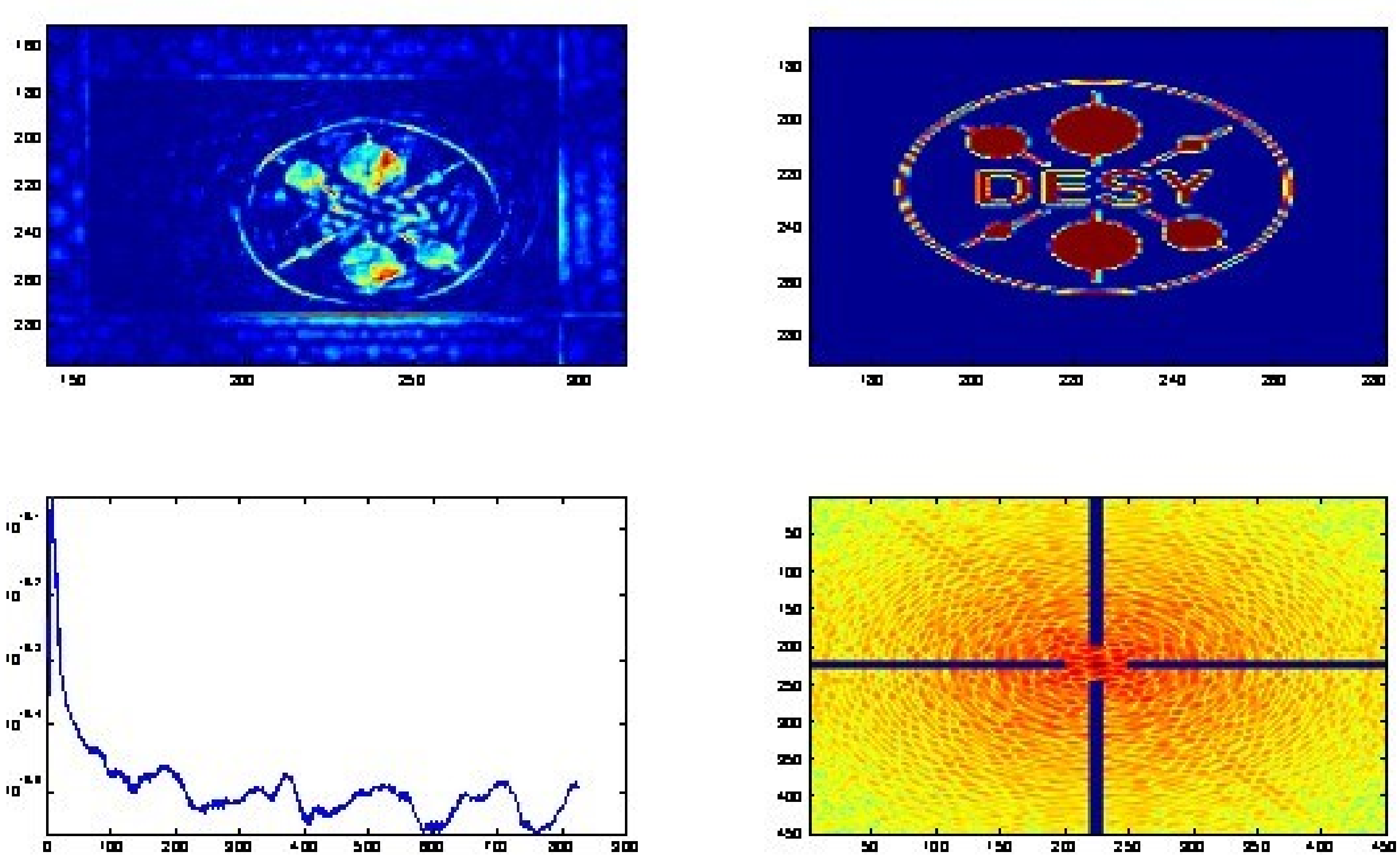
The beamstop problem



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)

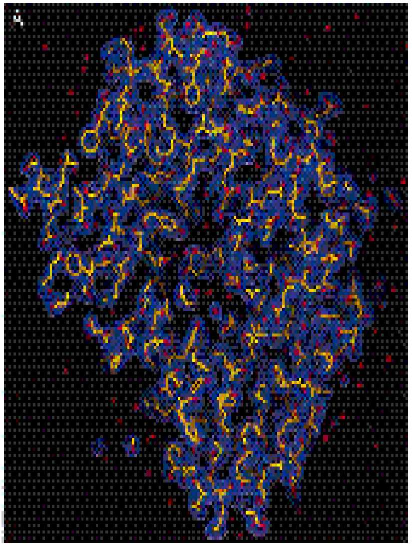
(ii) algorithm approach: calculate F also for the missing pixels and rescale electron densities

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

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

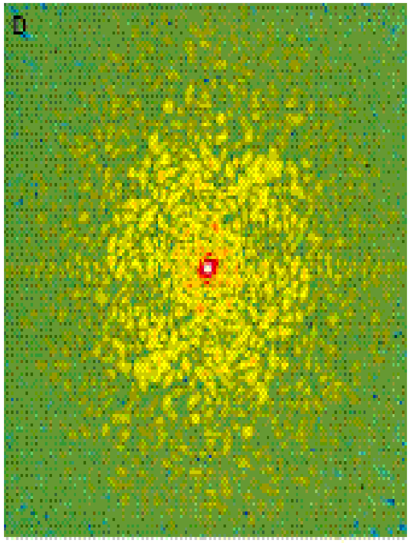
S. Marchesini, Chapman et al. PRB 68, 140101

An approach to three-dimensional structures of biomolecules by using single-molecule diffraction images: A simulation

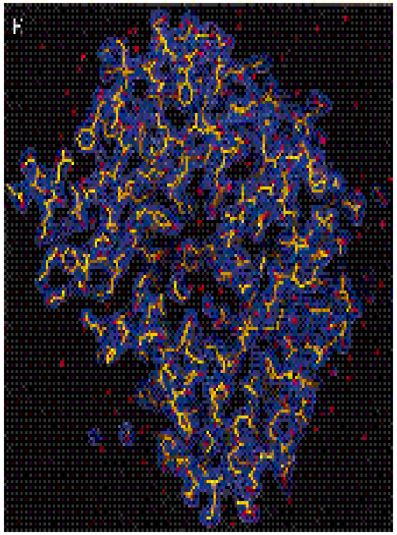


3-D structure (2.5 Å resolution) of rubisco molecule.

(106 kDa)



Top view of a section (kz=0) of 3-D scattering pattern from 10^6 single molecules (of known relative orientation) each "exposed" by a single 10 fs XFEL pulse (λ=1.5Å, 0.1μm beamsize) containing $2 \cdot 10^{12}$ photons.



Reconstructed 3-D pattern (from 250 2-D projections). Phasing by "oversampling" technique.

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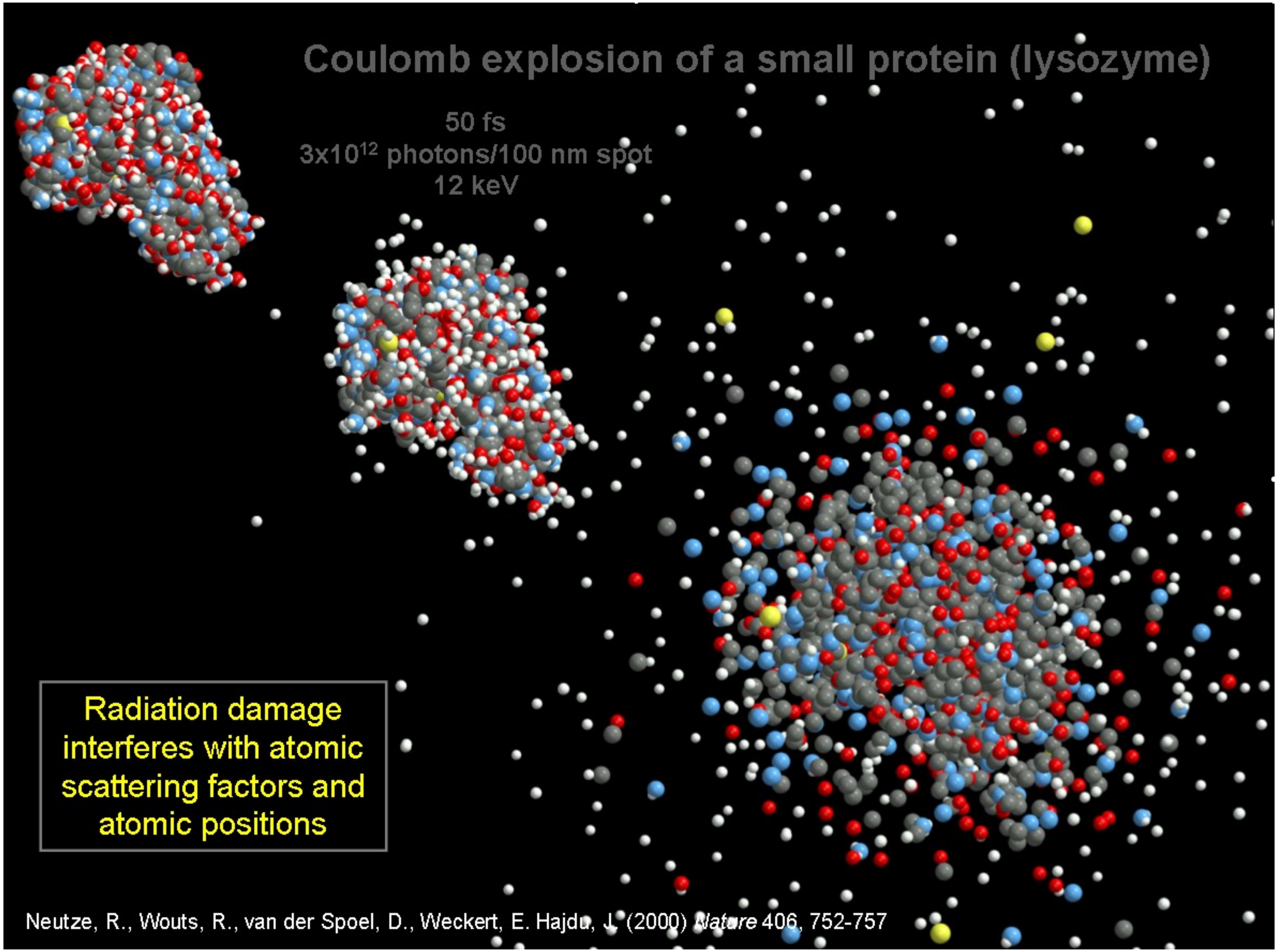
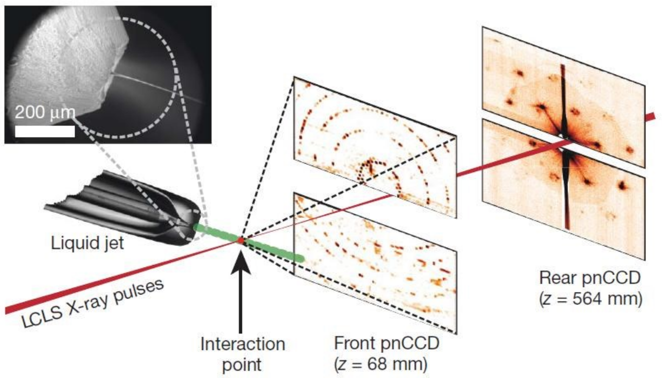
<u>NOTE:</u> Radiation Damage

J. Miao, K.O. Hodgson and D. Sayre, PNAS, 98, 6641 (2001)

Beam – Sample Interaction



Femtosecond X-ray protein nanocrystallography

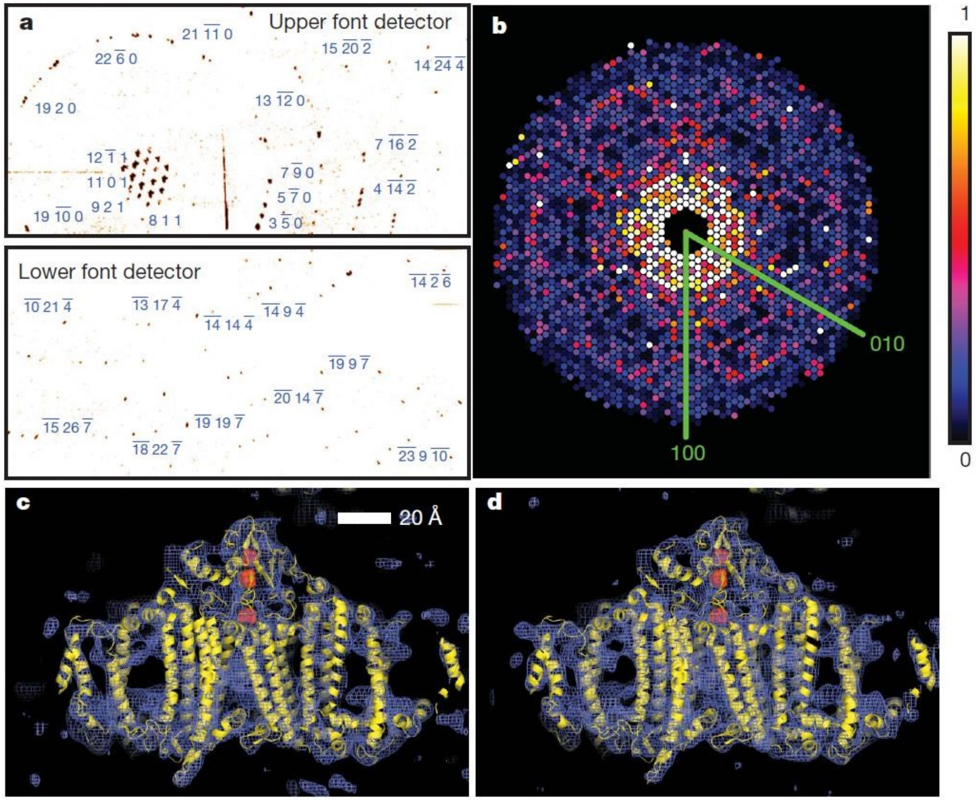


Nanocrystals (200 nm to 4 microns) of photosystem I flow in their buffer solution in a gas-focused, 4-mm-diameter jet at a velocity of 10ms/s perpendicular to the pulsed 1.8 keV X-ray FEL beam that is focused on the jet.

(a) Diffraction pattern recorded on the front pnCCDs with a single 70-fs pulse after background subtraction and correction of saturated pixels. The resolution in the lower detector corner is 8.5A˚.

(b) Precession-style pattern of the [001] zone for photosystem I, obtained from merging femto-second nanocrystal data from over 15,000 nanocrystal patterns.

(c, d) Region of the electron density map calculated from the 70-fs data (c) and from conventional synchrotron data truncated at a resolution of 8.5A˚ and collected at a tempera-ture of 100K (d)



Henry N Chapman et al., Nature 470. 73 (2011)