Coherent Diffraction Imaging using X-ray FELs

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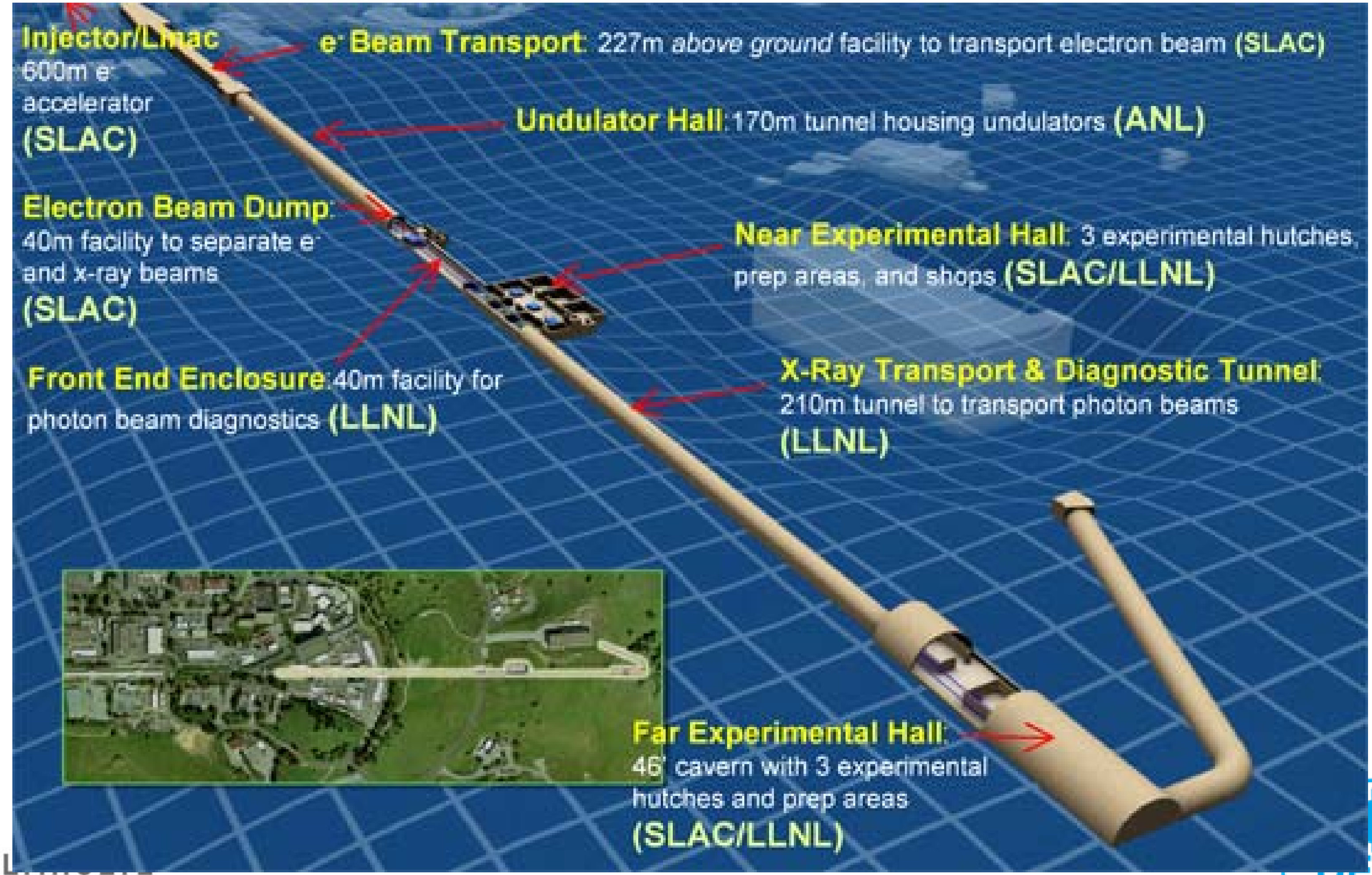
HELMHOLTZ ASSOCIATION

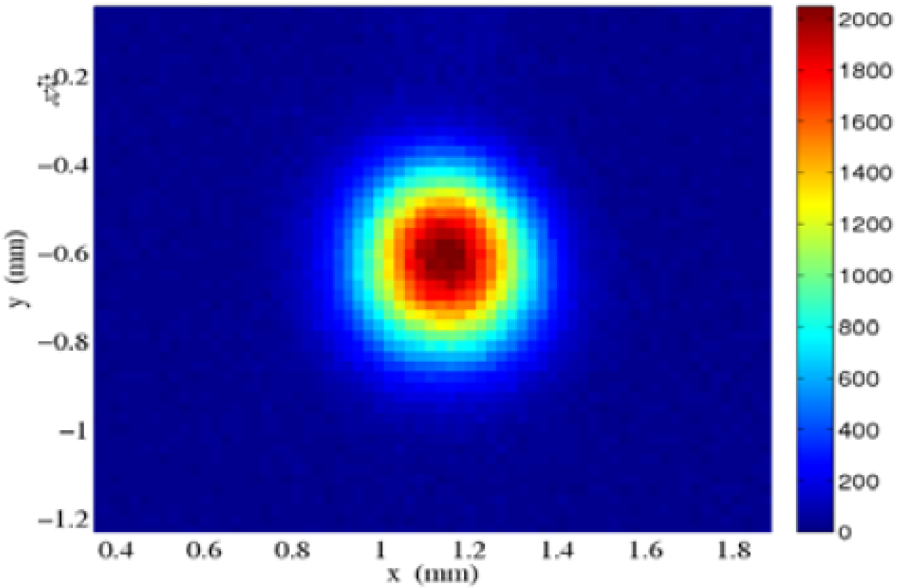
DESY

Revolution in X-ray Physics is just happening

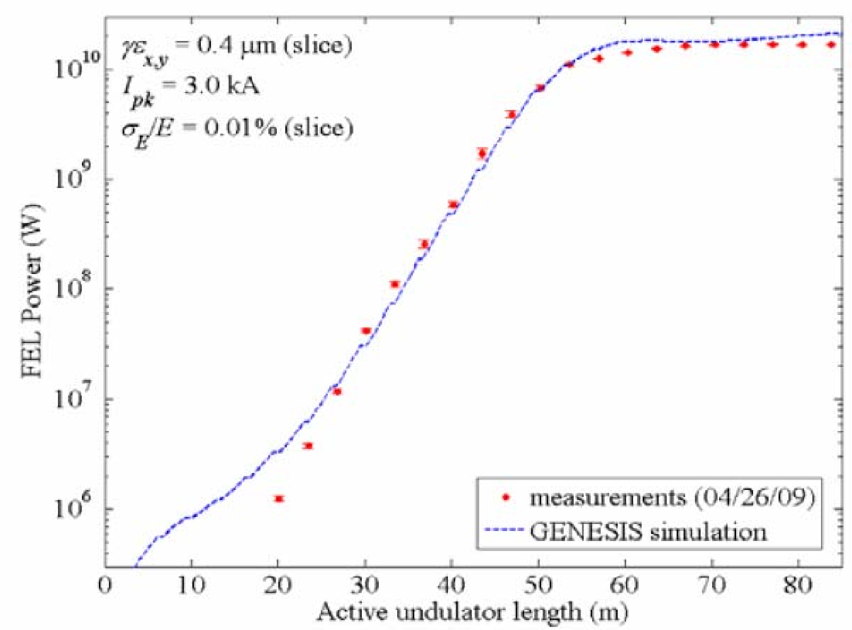


Revolution in X-ray Physics is just happening

April 2009



FEL x-rays at 1.5 Å on a YAG screen 50 m after the last inserted undulator



FEL power gain length measurement (red points) at 1.5 Å made

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P. Emma, *et al.*, http://www-ssrl.slac.stanford.edu/lcls/commissioning/documents/th3pbi01.pdf

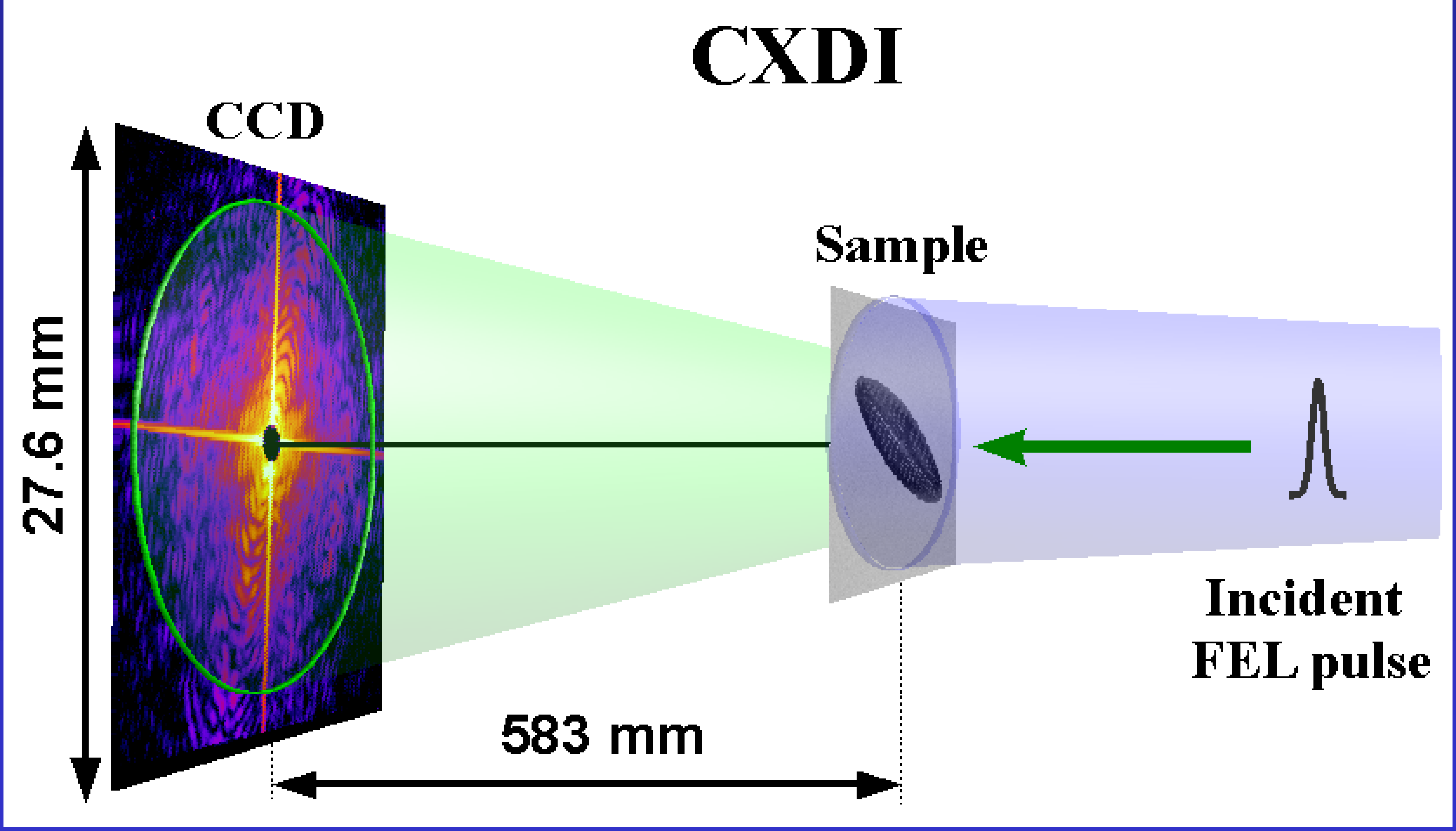
Outline

1. Introduction
2. Coherent Scattering and Phase Retrieval
3. Coherent X-ray scattering and Imaging in Material Science
   - Present status
   - Future applications
4. Requirements for Coherent X-ray scattering beamline at XFEL
5. Summary and Outlook

Coherent Scattering and

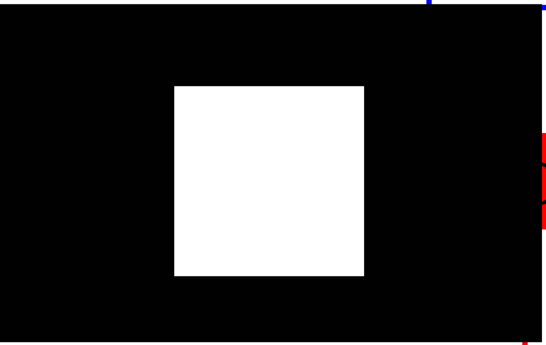
Phase Retrieval

Coherent imaging at FEL sources



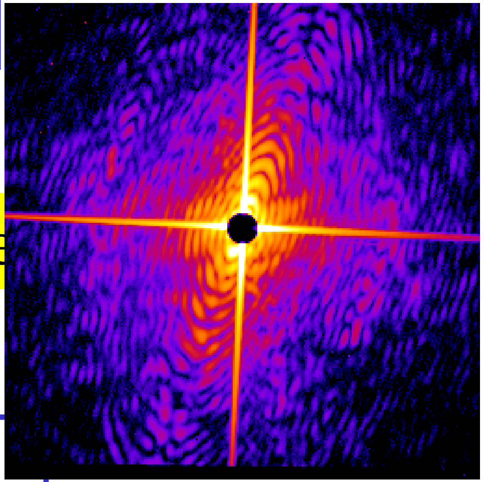
Iterative phase retrieval algorithm

$s_k(\mathbf{x})$ —FFT→ $A_k(\mathbf{q})$



Constraints

Reciprocal Space



$s'_k(\mathbf{x})$ ←FFT^{-1}— $A'_k(\mathbf{q})$

Real space constraints:

use all *a priori* knowledge:

- finite support
- positivity
- …

Reciprocal space constraint:

$$\left|A_k(\mathbf{q})\right| \rightarrow \sqrt{I_{exp}(\mathbf{q})}$$

R.W.Gerchberg & W.O. Saxton, *Optic* (1972) **35**, 237
J.R. Fienup, *Appl Opt.* (1982) **21**, 2758
V. Elser, *J. Opt. Soc. Am. A* (2003) **20**, 40

CXDI in practice is working well but

CXDI is a photon hungry technique

We can benefit from the use of ultrabright pulses of XFEL

CXDI Experiments at XFEL

Coherent X-ray scattering and lensless imaging in materials science

Coherent X-ray scattering and lensless imaging at the European XFEL Facility

I. A. Vartanyants, I. K. Robinson, I. McNulty, C. David, P. Wochner and Th. Tschentscher

J. Synchrotron Rad. (2007). **14**, 453–470

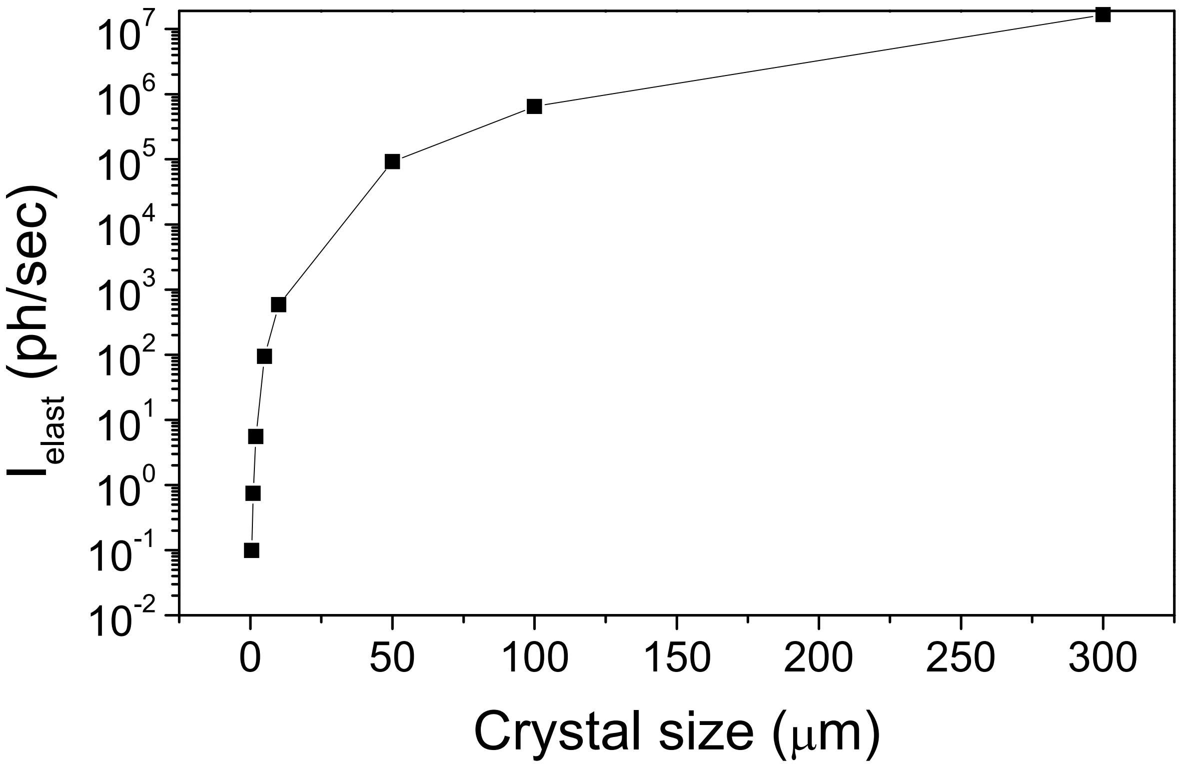
Coherent x-ray scattering and lensless imaging in materials science

Scientific aims

- 3D nanoscale structural analysis using CXDI technique
- main techniques proposed
  - ⇒ **Bragg diffraction for crystalline samples**
  - ⇒ **Forward scattering for non-crystalline objects**
- applications and systems of interest
  - ⇒ **Nanomaterials**
    - **properties of single particles ($100 \times 100 \times 100$ nm^3 in single-shot)**
    - **quantum dots**
    - **anisotropic strain distribution of single islands (on surface or buried)**
  - ⇒ **Mesoscale systems**
    - **obtain information about local properties of bulk structures avoiding averaging**
    - **metals (e.g. dislocations, nucleations) or ceramics (e.g. grain dynamics)**
  - ⇒ **Dynamic processes/fluctuations**
    - **crystal surface morphology as function of time**
    - **spontaneous nucleation of clusters**
    - **crystalline fluctuations in disordered matter**
  - ⇒ **Materials properties during processing**
    - **precipitates/pores growth or shrinkage**
    - **welding**
    - **laser ablation**

J. Synchrotron Rad. (2007). **14**, 453–470

Nanomaterials



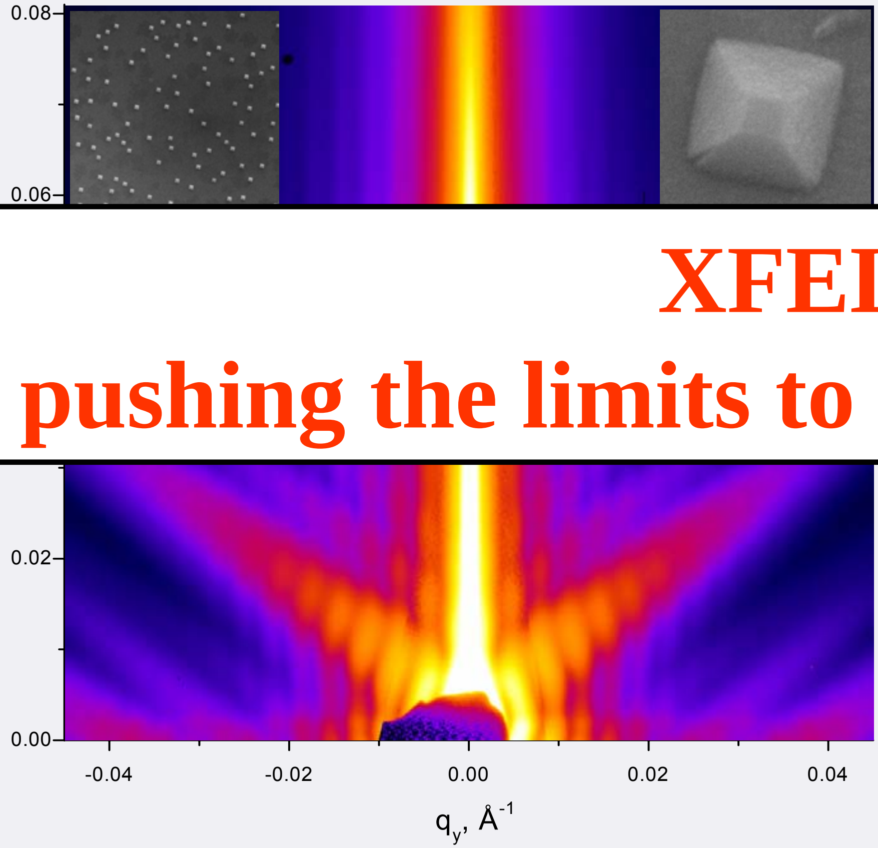
Elastically scattered intensity

incoming flux $I_0=10^{11}$ ph/s·mm^2, E=20 keV

Nanomaterials

(c)

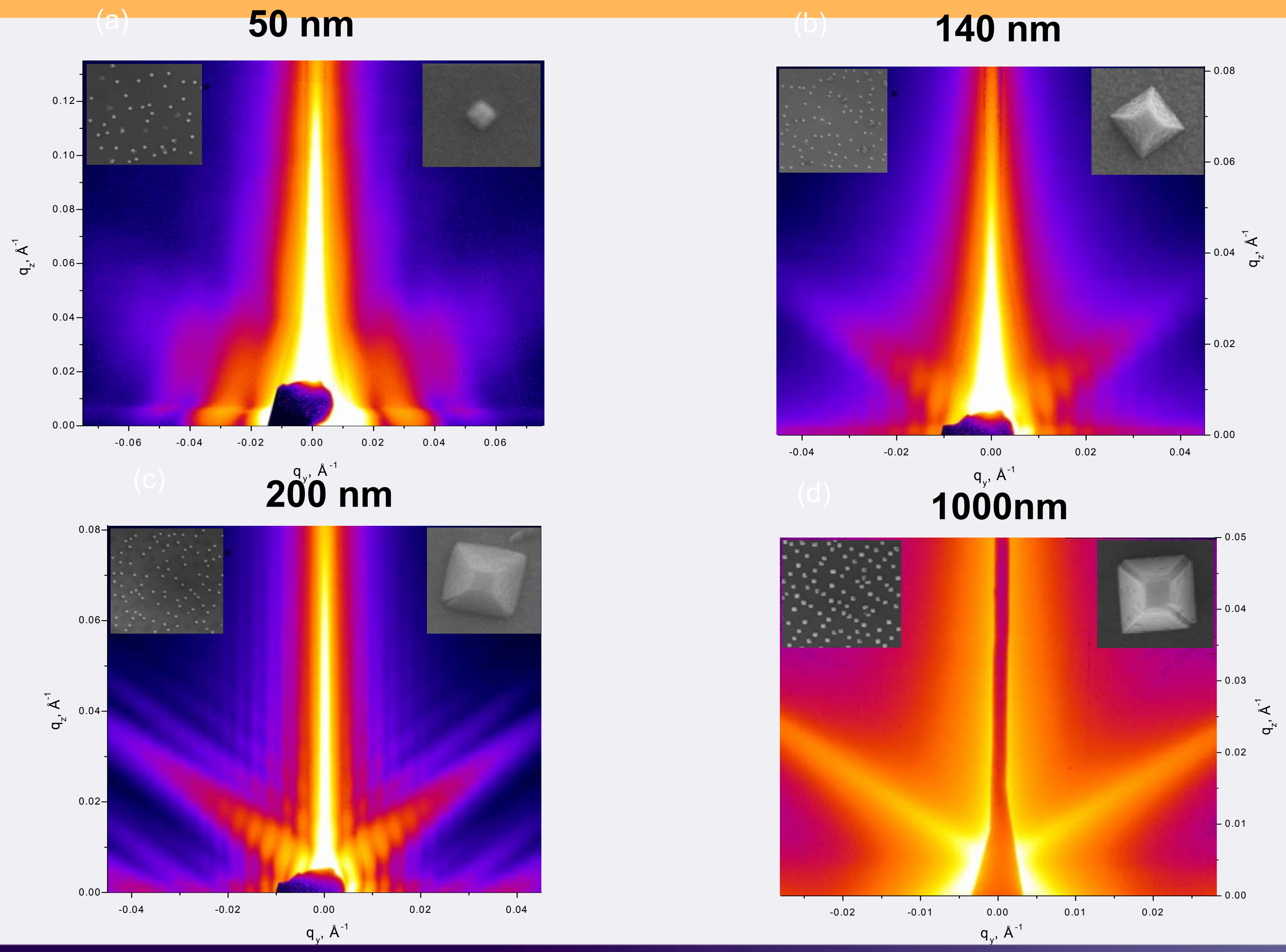
200 nm



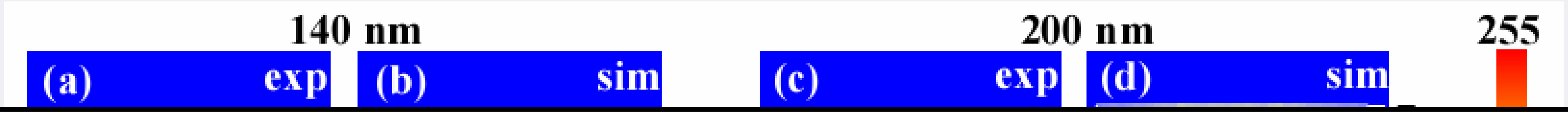
Coherent GISAXS scattering

XFEL
pushing the limits to 10 nm and below

Coherent GISAXS Measurements



Results of reconstruction



Extension to XFEL is straightforward

Time- dependent studies (reactions, oxidation, *etc.*)

An estimate of resolution: **10-15 nm !**

Not accessible by any other X-ray methods

A. Zozulya, O. Yefanov, I. Vartanyants *et al.*, Phys. Rev. B **78**, 121304R (2008)

Extension to XFEL

Scattered intensity for a narrow size distribution:

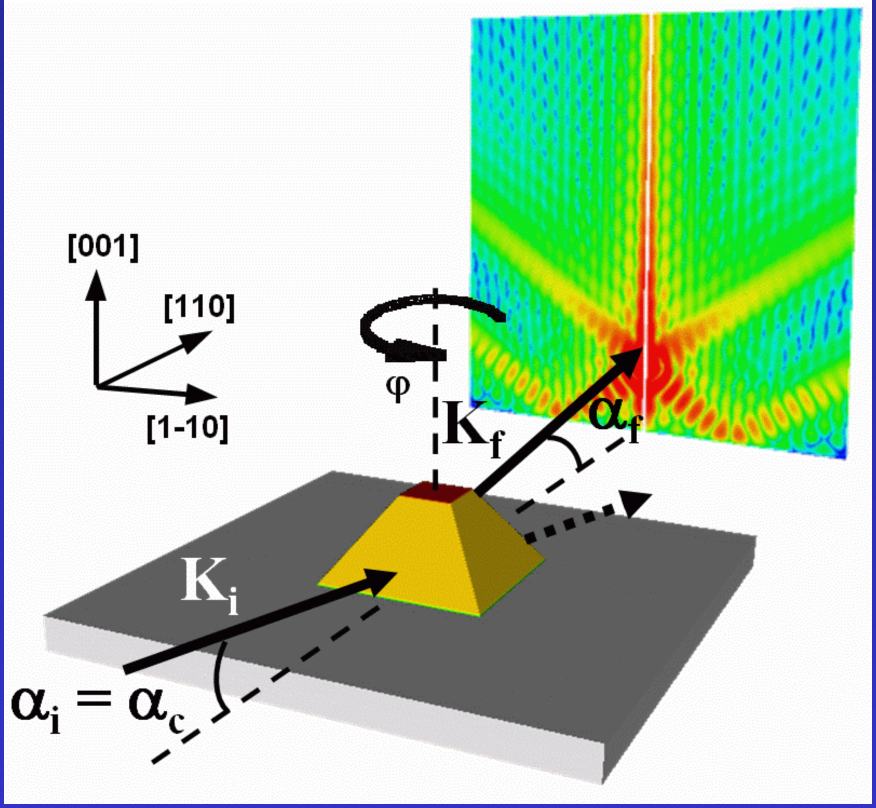
$$I(\mathbf{q}) = N I_{av}^{coh}(\mathbf{q}) \cdot \left[1 + \frac{1}{N} \sum_{i \neq j} \exp\left[i\mathbf{q}(\mathbf{R}_i - \mathbf{R}_j) \right] \right]$$

Experimental parameters for each sample during measurements

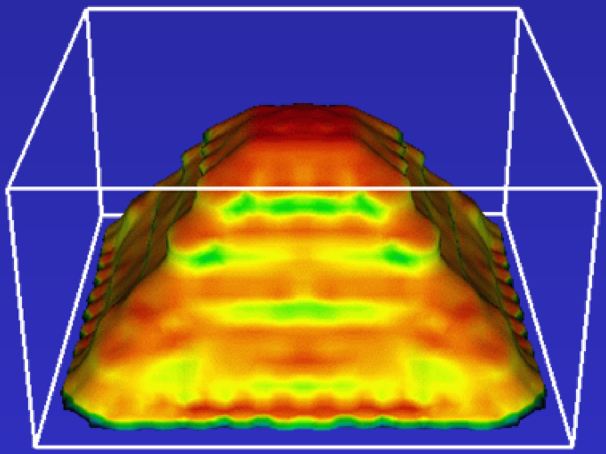
Sample	Average island size, nm	No. of illuminated islands, $N_{islands}$	Average distance, nm	Incoming flux, ph/s	Total incoming flux, ph	Incoming flux per averaged island, ph/s	Total incoming flux per averaged island, ph	Incoming flux per averaged island×$N_{islands}$, ph/s	Total incoming flux per averaged island×$N_{islands}$, ph
1	50	$3.5\cdot10^{7}$	200	$7.5\cdot10^{11}$	$5.4\cdot10^{15}$	$2.5\cdot10^{2}$	$1.8\cdot10^{6}$	$8.7\cdot10^{9}$	$6.3\cdot10^{13}$
2	140	$4.7\cdot10^{6}$	650	$3\cdot10^{12}$	$7.8\cdot10^{15}$	$2\cdot10^{3}$	$5.2\cdot10^{6}$	$9.4\cdot10^{9}$	$2.4\cdot10^{13}$
3	200	$2.5\cdot10^{6}$	900	$3\cdot10^{12}$	$1.2\cdot10^{16}$	$4\cdot10^{3}$	$1.6\cdot10^{7}$	$1\cdot10^{10}$	$4\cdot10^{13}$
4	1000	$7.2\cdot10^{5}$	2400	$1\cdot10^{12}$	$3.6\cdot10^{14}$	$1\cdot10^{5}$	$3.6\cdot10^{7}$	$7.2\cdot10^{10}$	$2.6\cdot10^{13}$

Extension to 3D imaging?

Coherent X-ray Diffraction Tomography



Incident angle: $\alpha_i = \alpha_c = 0.22^\circ$
Saxs tube: CCD at 2 meters
Azimuthal scan ω : 1° steps



Reconstructed shape of an island

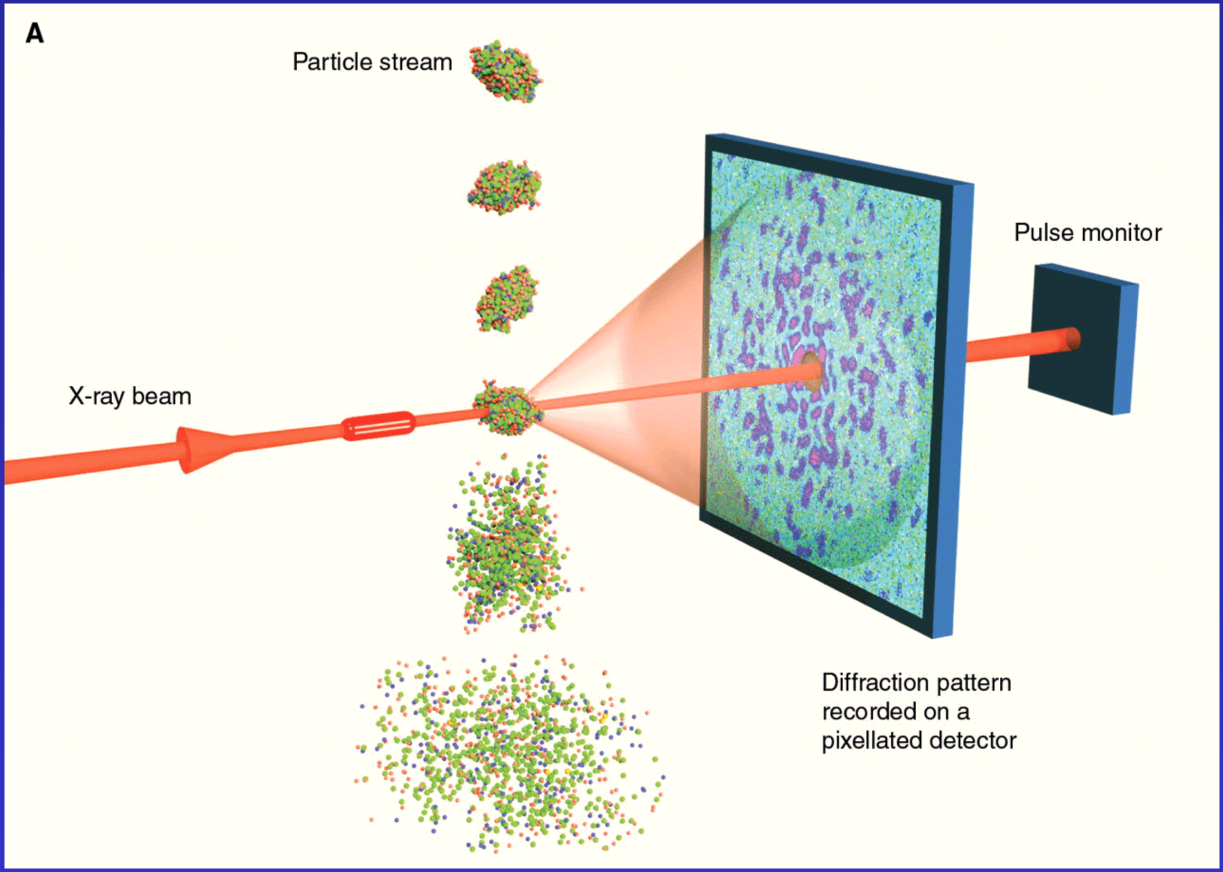
O. Yefanov *et al.*, APL **94** 123104 (2009)

Imaging of Biological samples with XFEL

Low scattered signal

Radiation damage

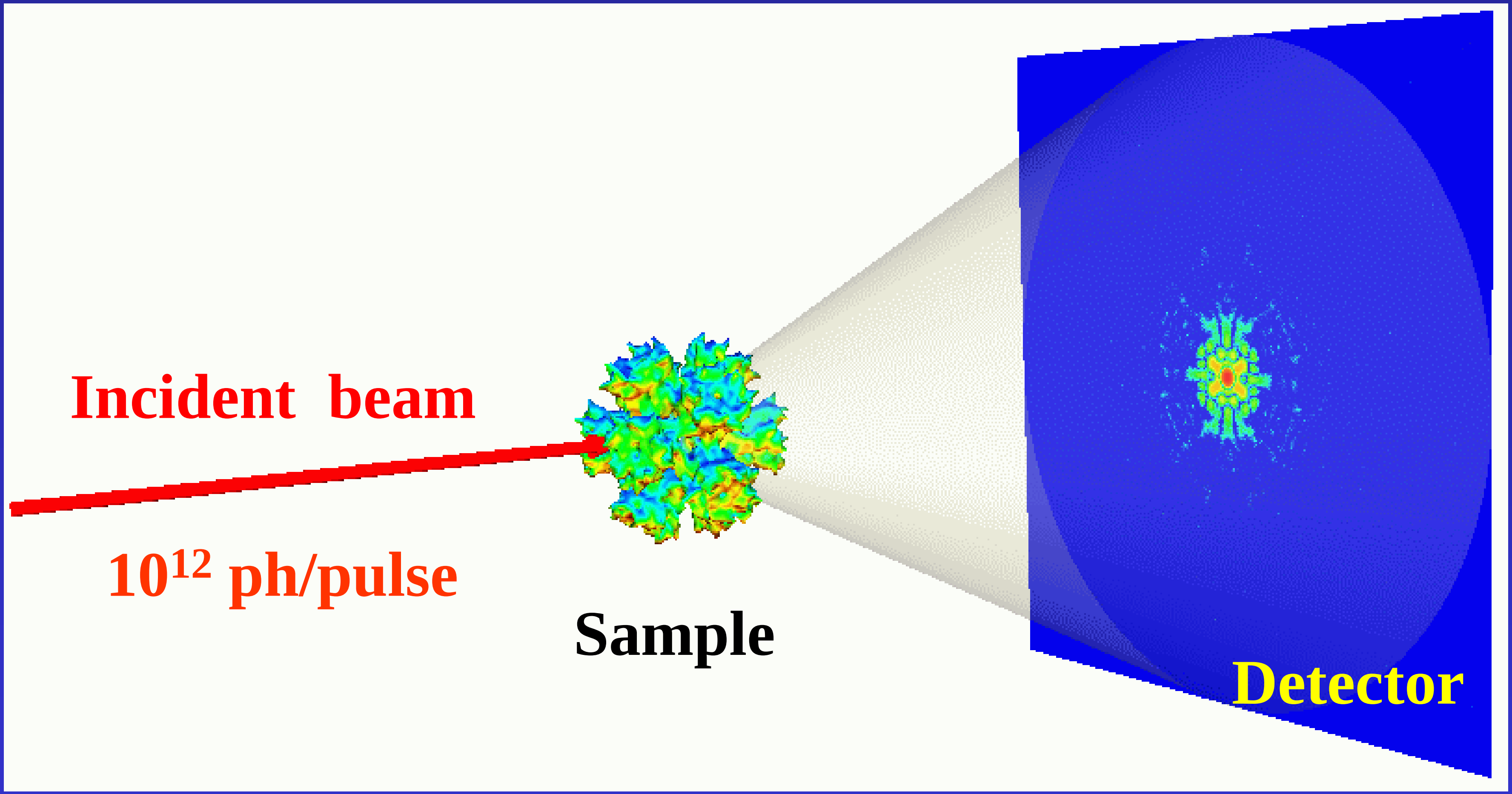
Imaging of biomolecules with femtosecond X-ray pulses



An elegant idea is based on measuring a sufficiently sampled diffraction pattern from a *series of injected* biological samples (viruses, molecules)

K. J. Gaffney and H. N. Chapman, Science (2007)

Imaging of biomolecules with femtosecond X-ray pulses



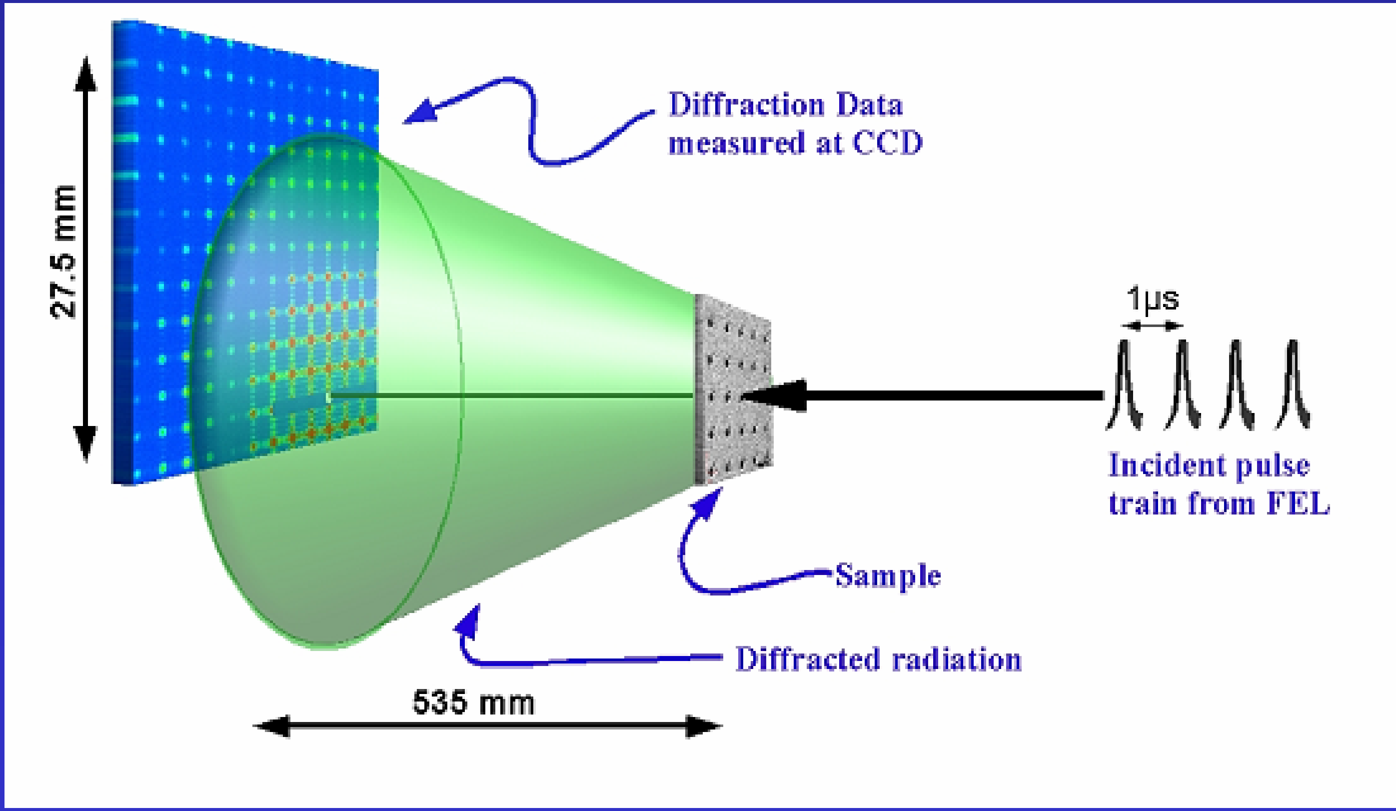
Single shot will produce a low resolution noisy image

FLASH Facility at DESY

• Jan 2005	first lasing at 32 nm
• Aug 2005	start of user experiments
• Energy range	~0.3 - 1GeV
→	~6.5 - 60 nm (~20-200 eV)
• Peak power	1-10 GW
• Pulse duration	<100 fs
• Bandwidth $\Delta\lambda/\lambda$	~0.5 %

Coherent-pulse 2D Crystallography at FEL

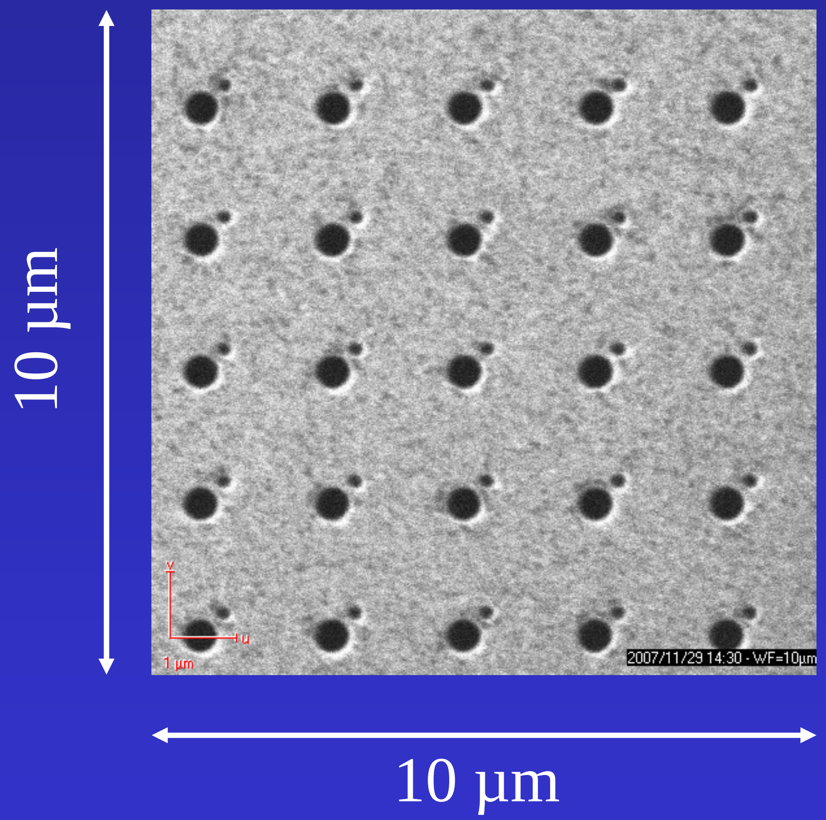
measurements at fundamental 7.97 nm



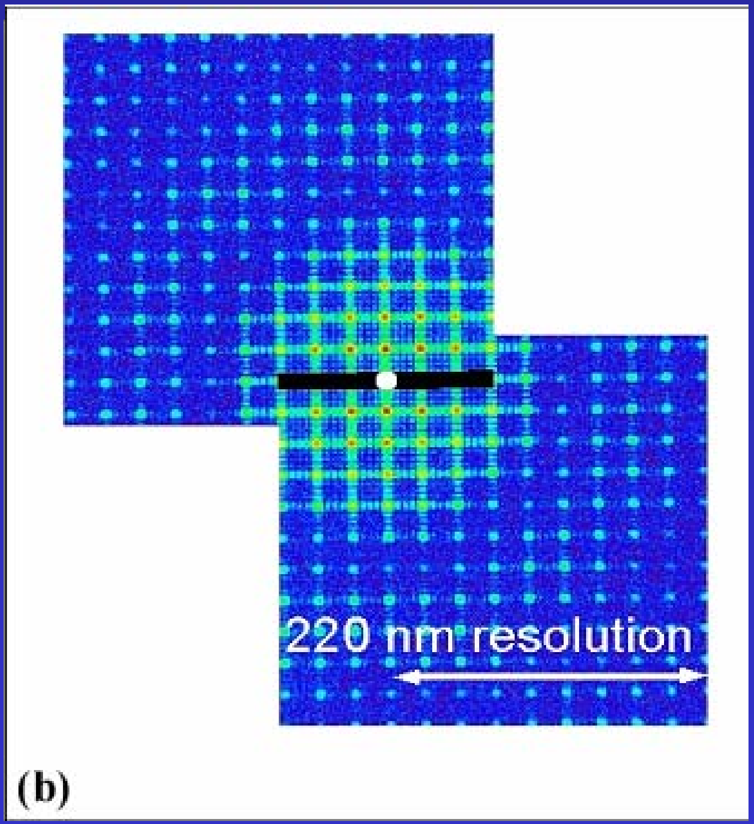
Scheme of experiment

Mancuso *et. al.* PRL **102,** 035502 (2009)

Coherent-pulse 2D Crystallography at FEL



Sample: prepared by FIB



Symmetrized Diffraction Pattern

Coherent-pulse 2D Crystallography at FEL

By using periodic array: we increase signal to noise ratio

$$S / N \sim \sqrt{N_{uc}}$$

Conditions of experiment

- beam size on the sample position: 150×150 μm^2
- flux: 4.5×10^{10} ph/pulse
- coherent flux on the sample area (0.2 sec):
  1.5×10^{10} ph/pulse train

This is at least one order of magnitude higher than at the 3rd generation synchrotron sources

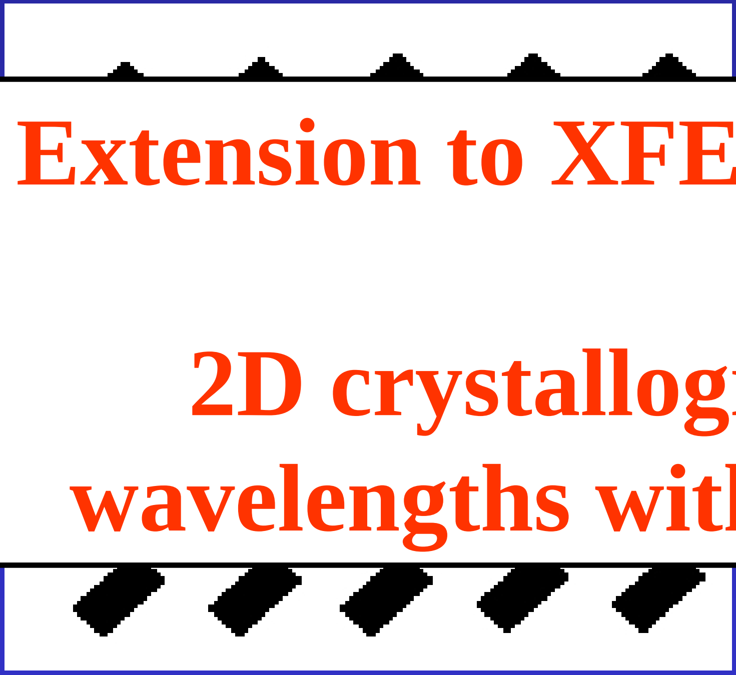
How to get an ultimate resolution?

Recipe:

- **To increase signal to noise ratio**
- **To break the symmetry**

Result of reconstruction

Increased signal to noise ratio due to 5 ×5 times binning



Extension to XFEL is straightforward

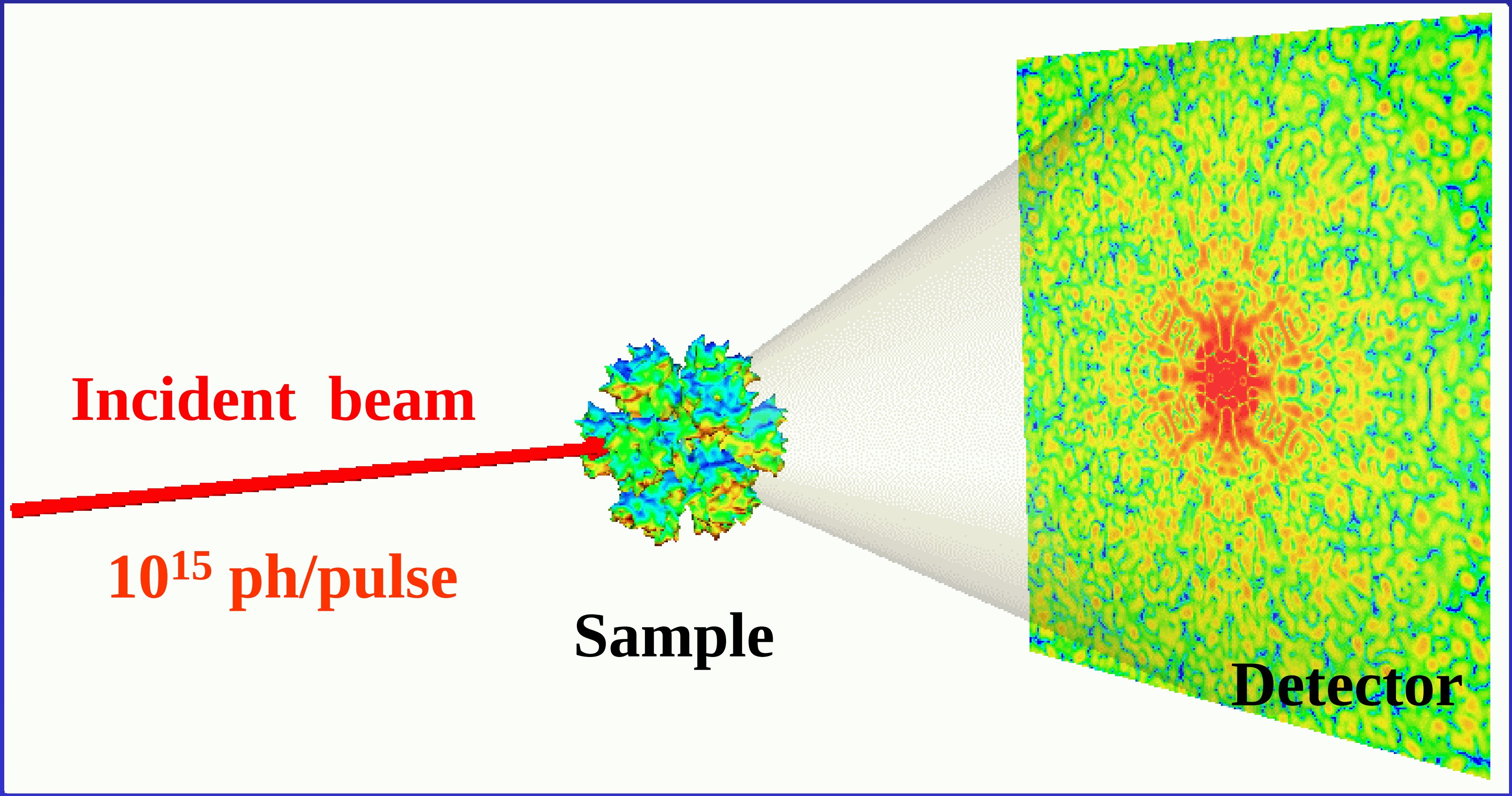
2D crystallography at shorter wavelengths with single FEL pulses

Support used in reconstruction

Reconstructed Image

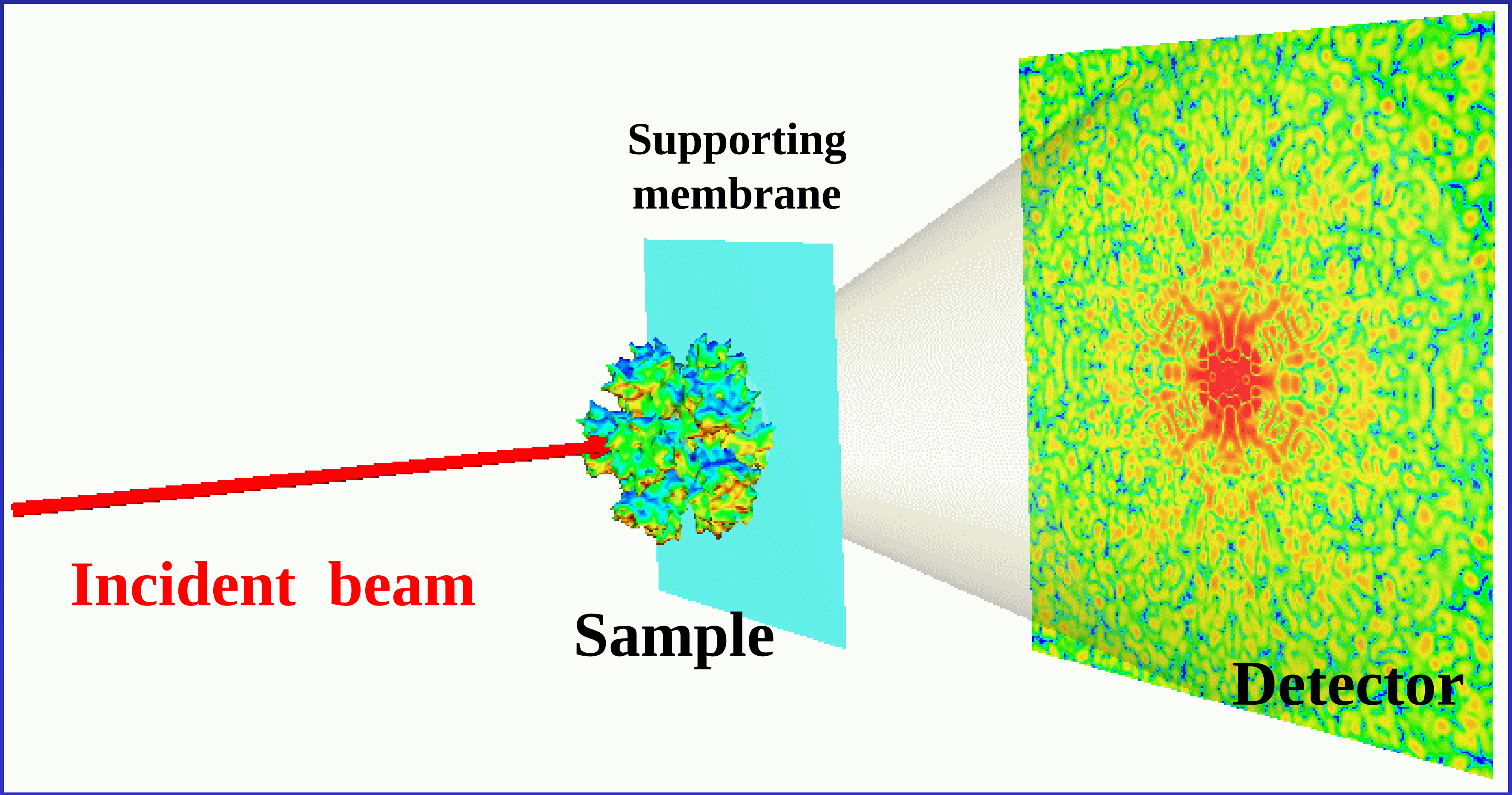
Resolution estimate: 220 nm

Imaging of biomolecules with femtosecond X-ray pulses



To obtain a 3D image of biological samples with sub-nanometer resolution many *reproducible* copies will need to be measured

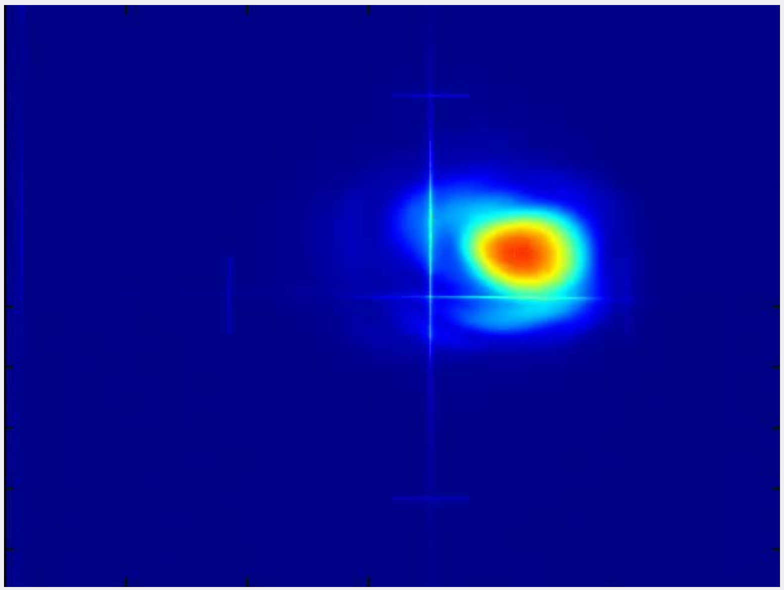
Imaging of biomolecules with femtosecond X-ray pulses

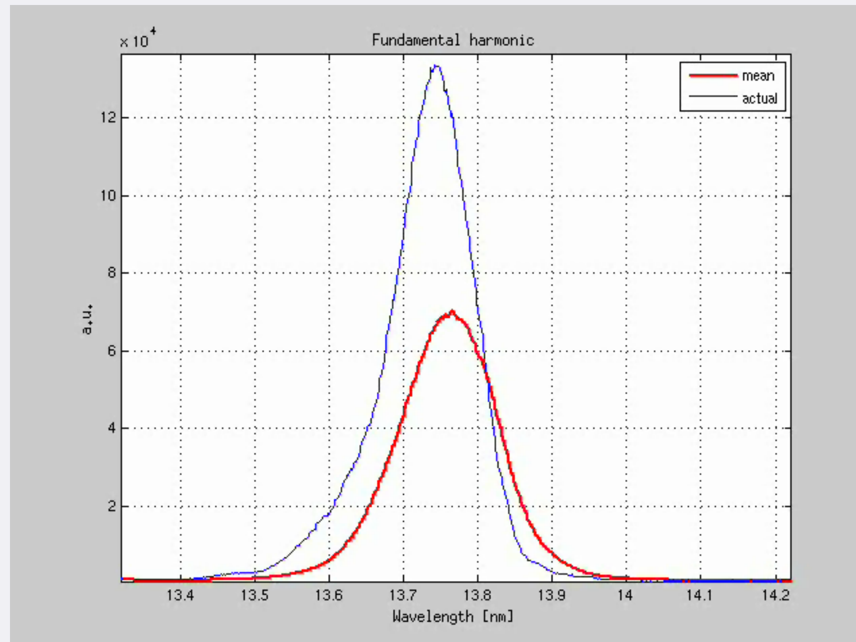


For *non-reproducible* biological samples (e.g. cells) supported on the membrane same 2D projection of electron density will be measured as for injected particles

SASE: Shot to Shot Fluctuations

Spectral distribution at 13,7 nm

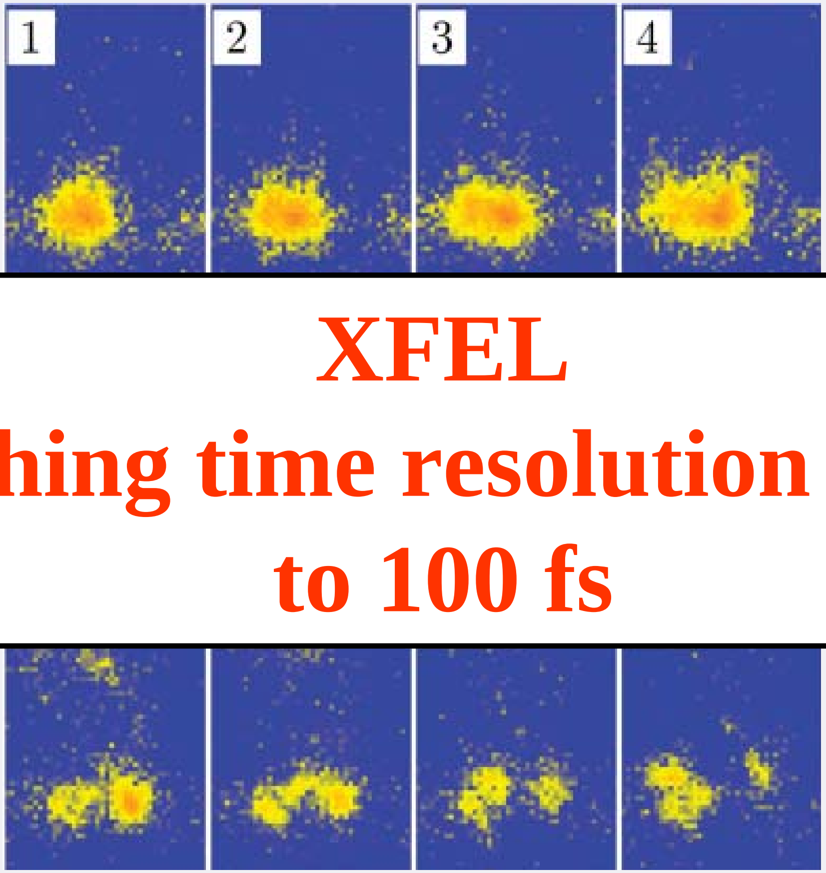




W. Ackermann et al.
Operation of a free-electron laser from the extreme ultraviolet to the water window
Nature Photonics 1, 336 (2007)

Study of the Early Stages of Protein Crystallization

Ferritine crystal measured at room temperature



XFEL
pushing time resolution limits to 100 fs

Evolution of a Bragg peak over time.
Each image is taken **95 s** apart.

S. Boutet&I. Robinson , J. Synchr.Rad. (2006)

Requirements for Coherent X-ray Scattering beamline at European XFEL

Coherent x-ray imaging – Instrument requirements

Requirements

- solid samples
- tunable soft x-ray FEL radiation
- hard x-ray FEL radiation
- high energy spontaneous radiation
- large area detectors

X-ray beam requirements

- photon energy
  - ⇒ **0.5 – 1.0 keV (transition metal L)**
  - ⇒ **(3)8-12 keV (surfaces & bulk)**
  - ⇒ **60-90 keV (hard materials)**
  - ⇒ **bw 10^{-4}-10^{-3} (long. coherence)**
- beam size
  - ⇒ **100-1000 nm**
- time domain
  - ⇒ **10 Hz rep. rate**
  - ⇒ **Pulse trains for 200 ns timescale**
  - ⇒ **10 fs synchronisation**

Instrumentation

- photon diagnostics
  - ⇒**photon flux, coherence/wavefront, spectral distribution**
- sample manipulation & environment
  - ⇒**UHV conditions**
  - ⇒**solid samples**
  - ⇒**fast exchange and sample alignment (~1 μm)**
  - ⇒**surface preparation techniques**
- detector requirements
  - ⇒**x-ray area detectors covering a large portion of reciprocal space**

I. Vartaniants

J. Synchrotron Rad. (2007). **14**, 453–470

Coherence at European XFEL

Is it a fully coherent source?

I. Vartaniants, A. Singer, DESY Preprint, DESY 09-114 (2009)

General Theoretical Approach
based on Statistical Optics

Mutual coherence function:

$$\Gamma(\mathbf{r}_1,\mathbf{r}_2,\tau)=\langle E(\mathbf{r}_1,t)E(\mathbf{r}_2,t+\tau)\rangle$$

Cross-spectral density function:

$$W(\mathbf{r}_1,\mathbf{r}_2,\omega)=\int_{-\infty}^{\infty}\Gamma_{12}(\tau)e^{-i\omega\tau}d\tau$$

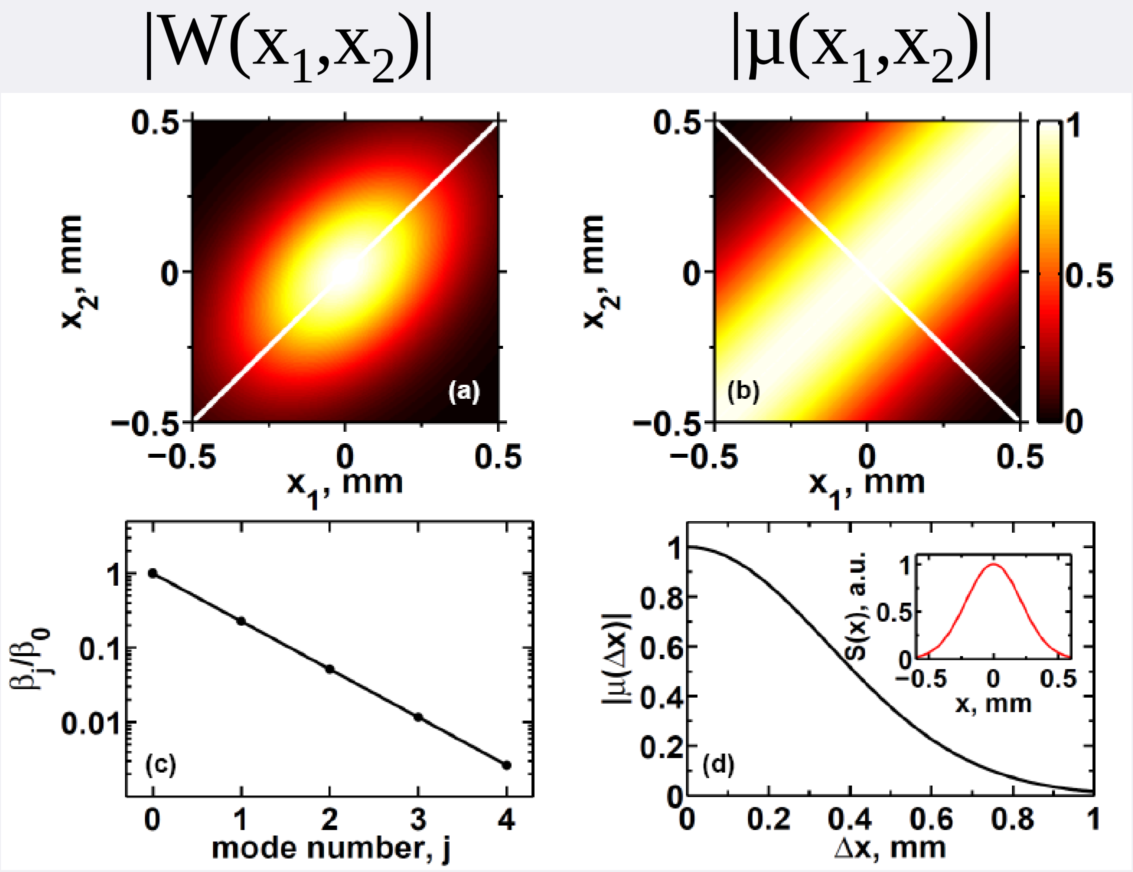
CSD of a partially coherent field can be represented as a sum of independent coherent modes

$$W(\mathbf{r}_1,\mathbf{r}_2)=\sum_{j=0}\beta_j\psi_j^*(\mathbf{r}_1)\psi_j(\mathbf{r}_2)$$

β_j and $\psi_j(\mathbf{r})$ are the eigenvalues and eigenfunctions that satisfy Fredholm integral equation

$$\int W(\mathbf{r}_1,\mathbf{r}_2)\psi_j(\mathbf{r}_1)d\mathbf{r}_1=\beta_j\psi_j(\mathbf{r}_2)$$

Calculations for European XFEL



Parameters of European XFEL for SASE I undulator:

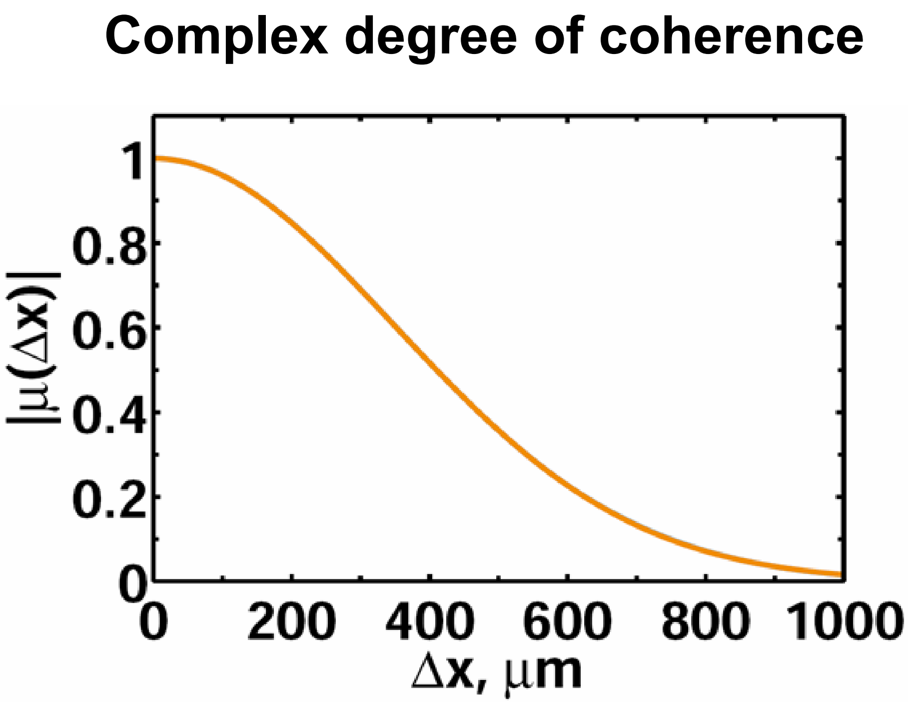
(E=12 keV, z=500m)

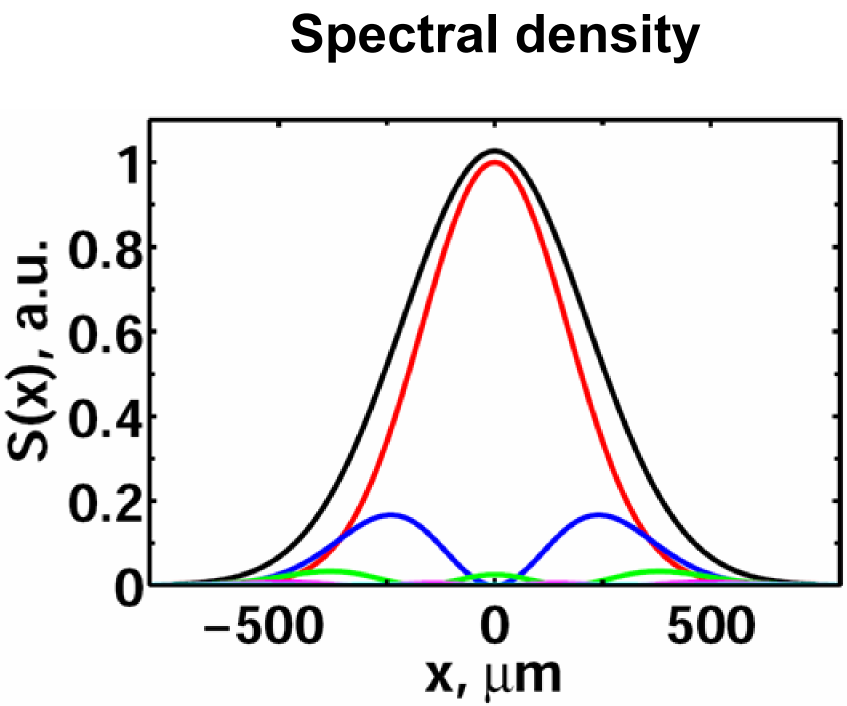
σ=30 μm
σ'=0.42 μrad

Degree of coherence: 63%

I. Vartaniants

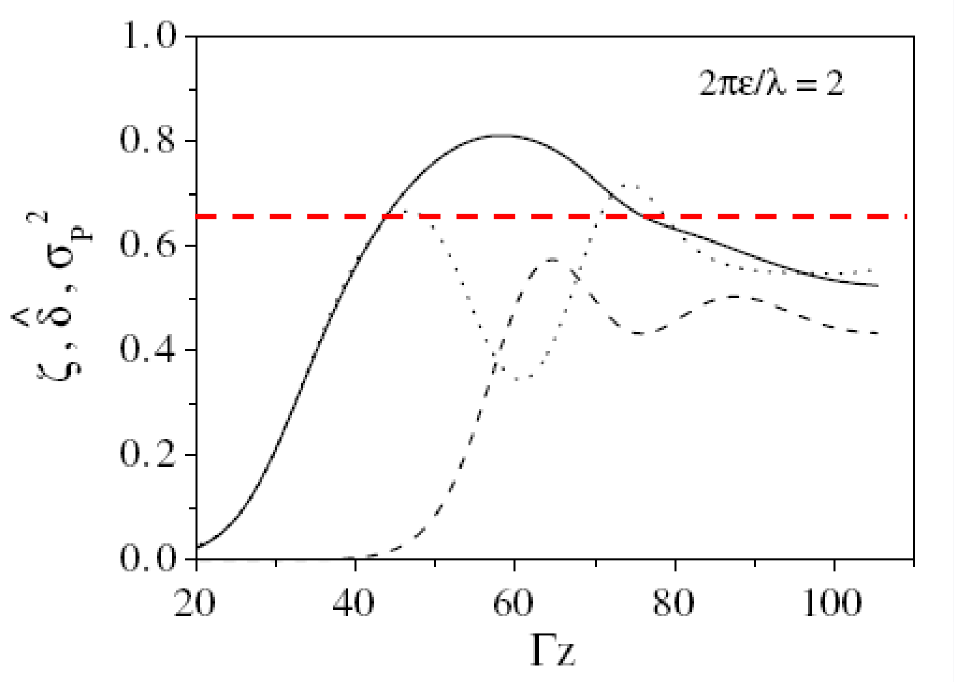
Contribution of each mode





Five modes

Transverse Coherence of XFEL



$$\gamma(\mathbf{r},\mathbf{r}') = \frac{\langle E(\mathbf{r},t)E(\mathbf{r}',t)\rangle}{\sqrt{I(\mathbf{r},t)}\sqrt{I(\mathbf{r}',t)}}$$

$$\varsigma = \frac{\int |\gamma(\mathbf{r},0)|^2 \; I(\mathbf{r})d\mathbf{r}}{\int I(\mathbf{r})d\mathbf{r}}$$

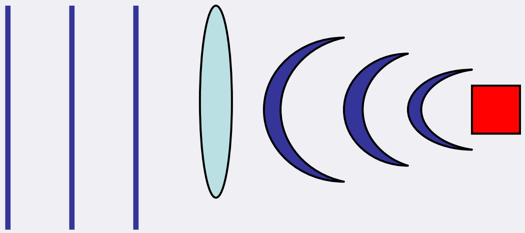
Degree of coherence: 65%

Degree of transverse coherence in SASE1 is mainly limited by the contribution of higher radiation modes

E.L. Saldin, *et al.*, Optics Communications **281** (2008), 1179

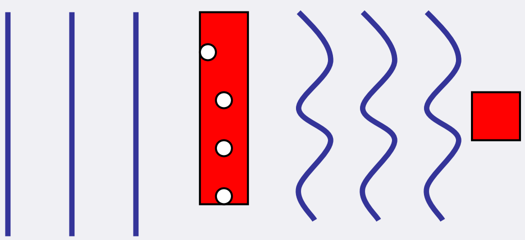
Optics

Optics in Coherent X-ray beam



Controlled change of the wavefront
(known additional phase)

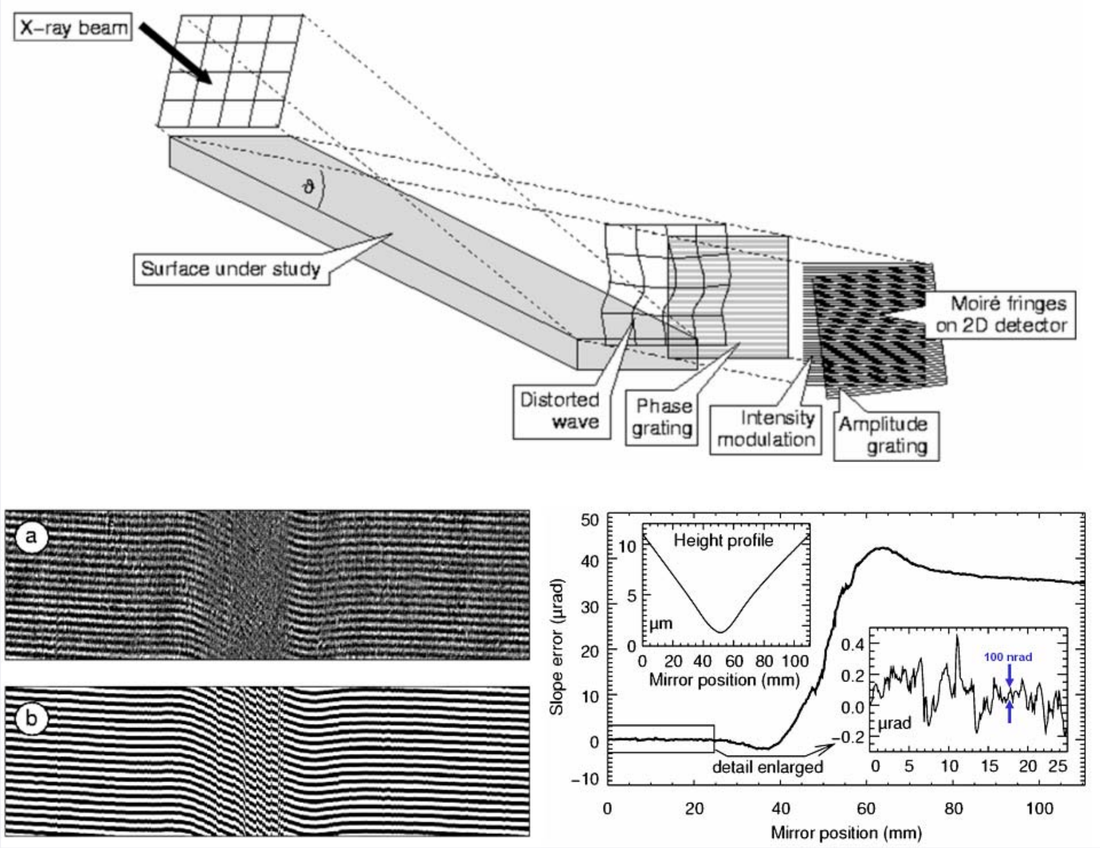
'Perfect' or Diffraction Limited Optics



Uncontrolled change of the wavefront
(unknown phase)
Artifacts in reconstruction

Non Perfect Optics

Coherence or Wave Front Preserving Optics



Setup for measurement of hard x-ray wave front distortions using a shearing interferometer (PSI)

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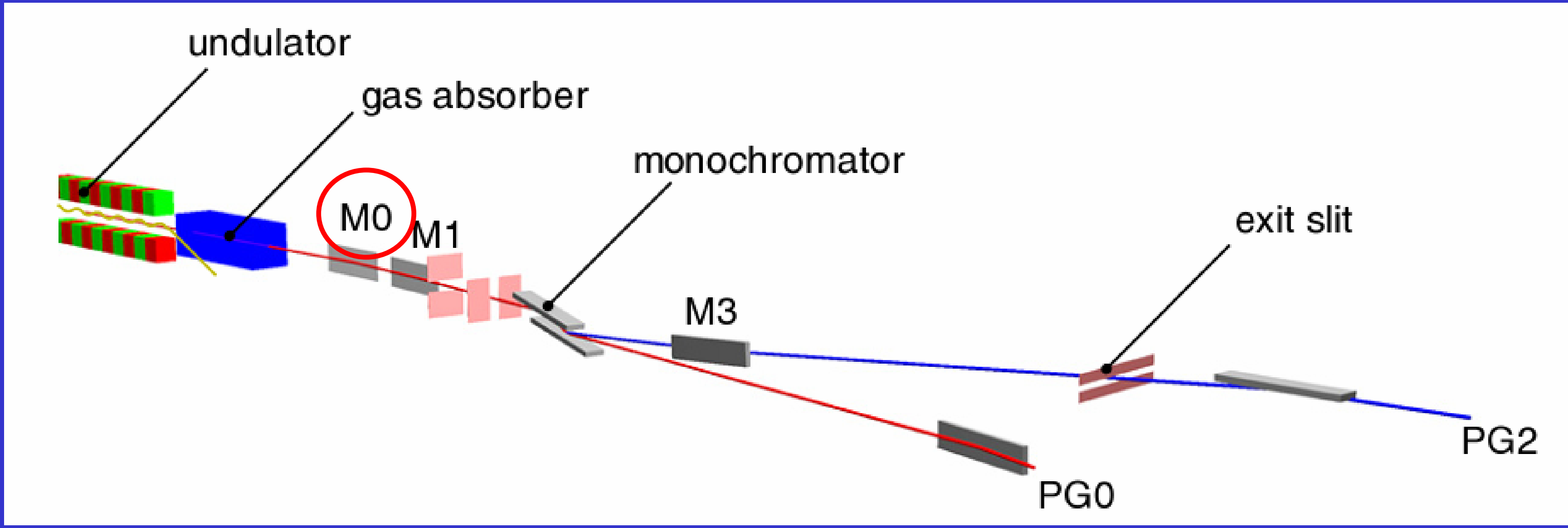
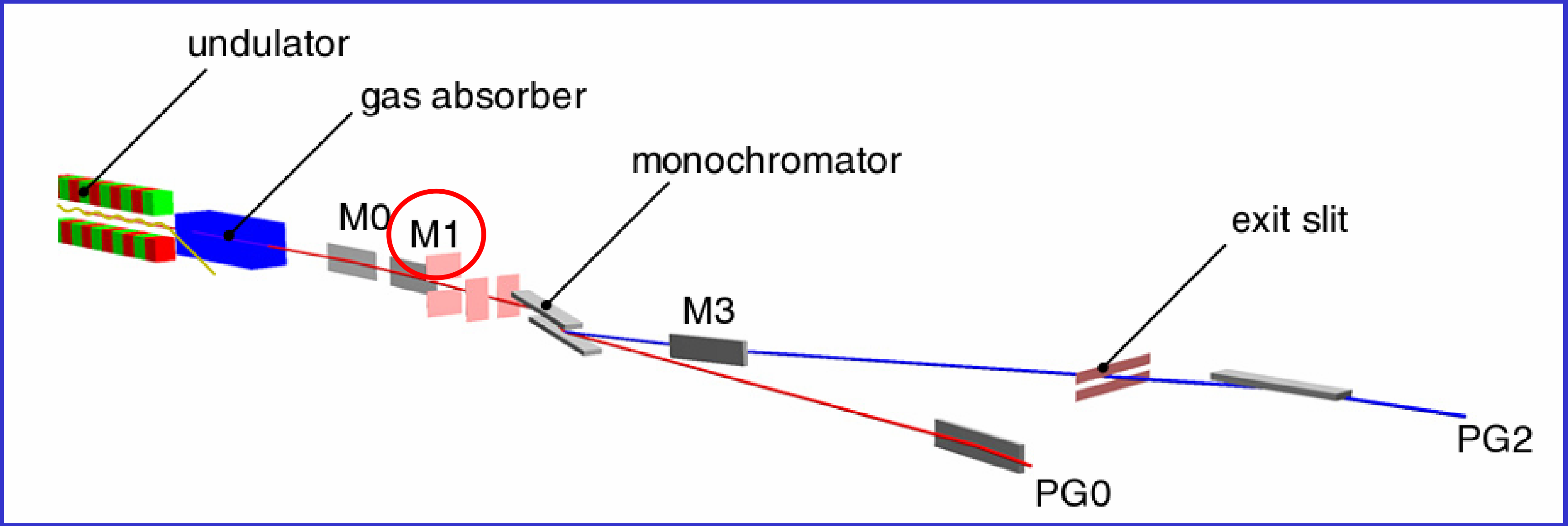
Beamline PG2 (PG0) @ FLASH



- Plane grating monochromator
- Two gratings: 200 lines/mm & 1200 lines /mm
- Optics are coated with diamond like carbon (DLC)

M. Wellhöfer *et al., J. Opt. A: Pure Appl. Opt.* **9** (2007) 749

Beamline PG2 (PG0) @ FLASH



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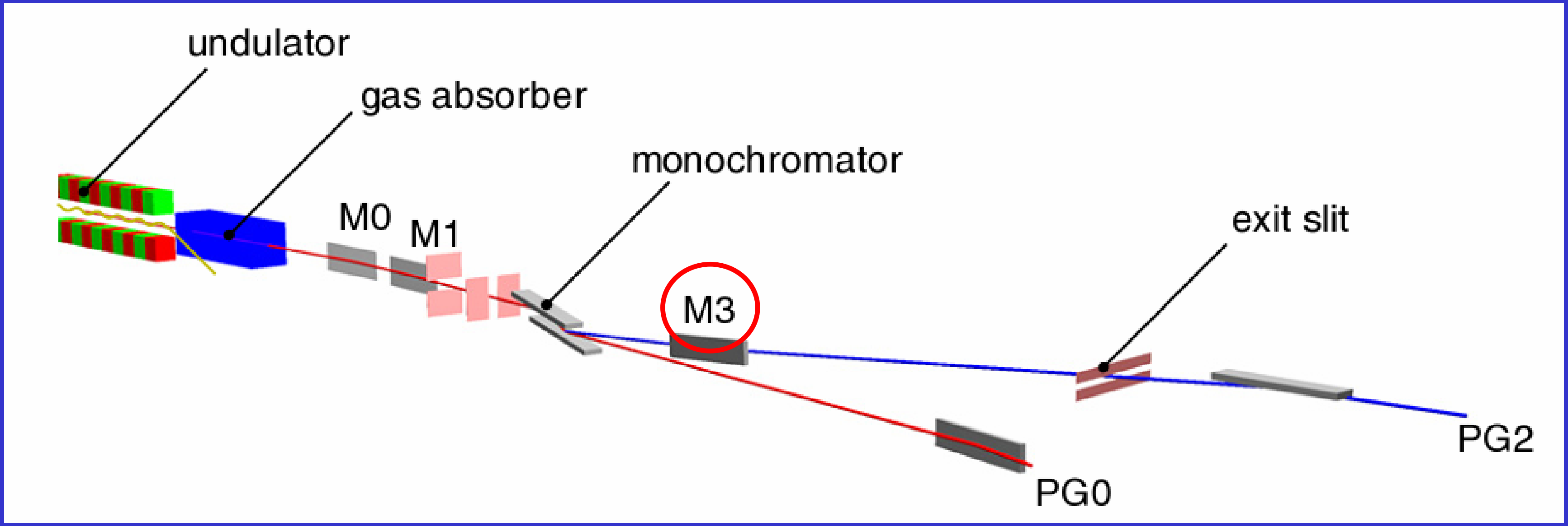

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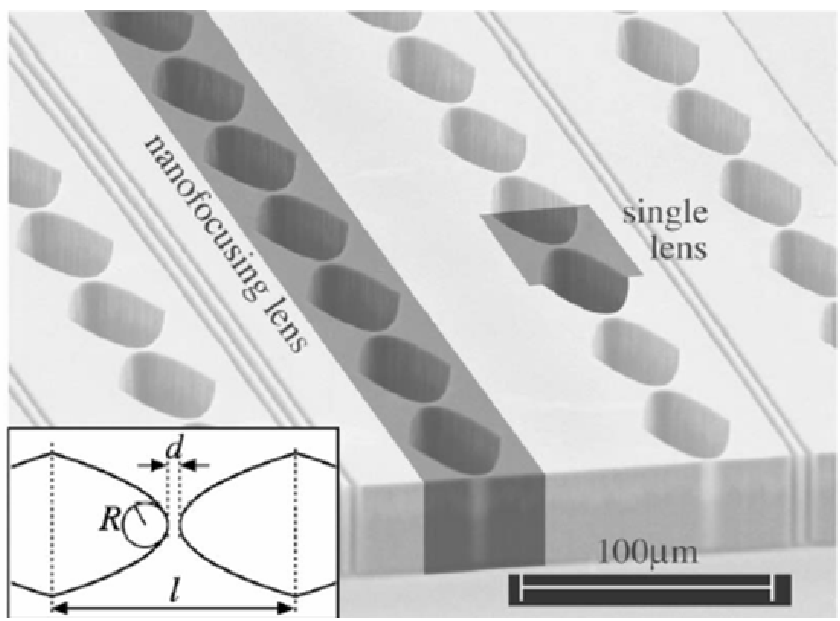
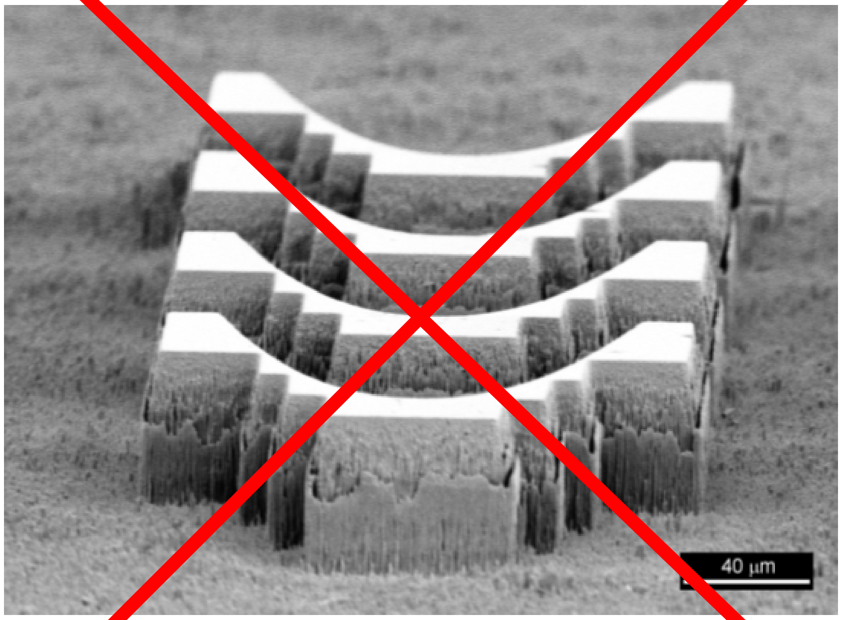
M. Wellhöfer *et al., J. Opt. A: Pure Appl. Opt.* **9** (2007) 749

Focusing Optics





Nano-focusing Si lenses made by electron beam litography
C. Schroer (2005)

Diamond planar refractive lenses fabricated using similar technique

Detector requirements

- 2D detectors
- Dynamic range (10^5)
- 4K×4K, 10K ×10K plus
- Tiling or robot arm design
- Fast readout speed

Data acquisition: Computer requirements

Size	Memory needed	
	Single Precision	Double Precision
256^3	336 MB	592 MB
512^3	2.6 GB	4.6 GB
1024^3	21 GB	37 GB
2048^3	168 GB	296 GB

Size	32-CPU G5 cluster speed	
	3D FFT time	Time for 3D reconstruction
256^3	73 msec	10 min
512^3	850 msec	1.5 hrs
1024^3	7.9 sec	14 hrs
2048^3	-	

*2000 iterations, 2 FFT's per iterationplus other floating point operations
code: C with mvapich, dist_fft (Altivec-optimized MPI FFT)
see http//images.apple.com/acg/pdf/20040827_GigaFFT.pdf

H. Chapman, et al. J. Opt. Soc. Am. (2006)

Summary

We want to use the full power of XFEL:

Experimental realization of CXDI at XFEL is straightforward and community is ready to use XFEL for coherent imaging
TODAY

On femtosecond time scales

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- O. Yefanov *(HASYLAB)*
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- R. Kurta *(HASYLAB)*

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- A. Zozulya (now at *EMBL, Hamburg)*

- **E. Weckert (*HASYLAB*)**
- *and many others …*

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[1] Deutsches Elektronen-Synchrotron (DESY), Notkestraße 85, 22607 Hamburg

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Thank you for your attention