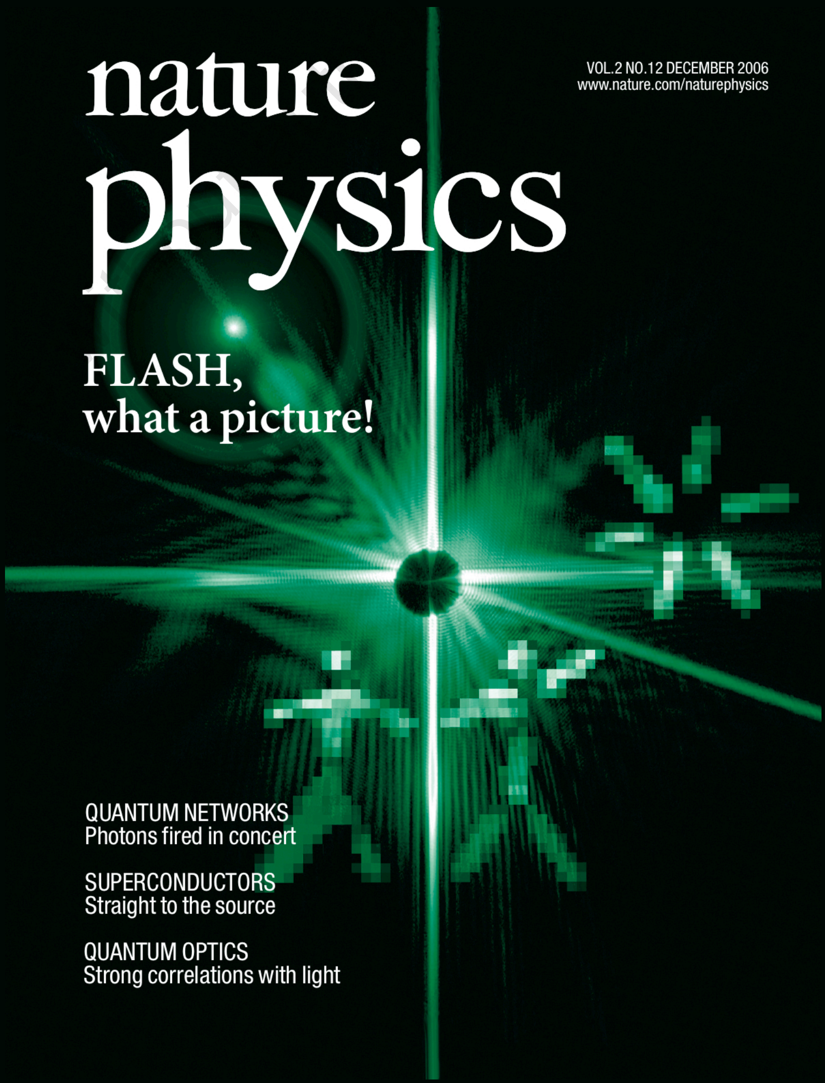
Coherent X-Ray Diffractive Imaging at LCLS, FLASH, and ALS

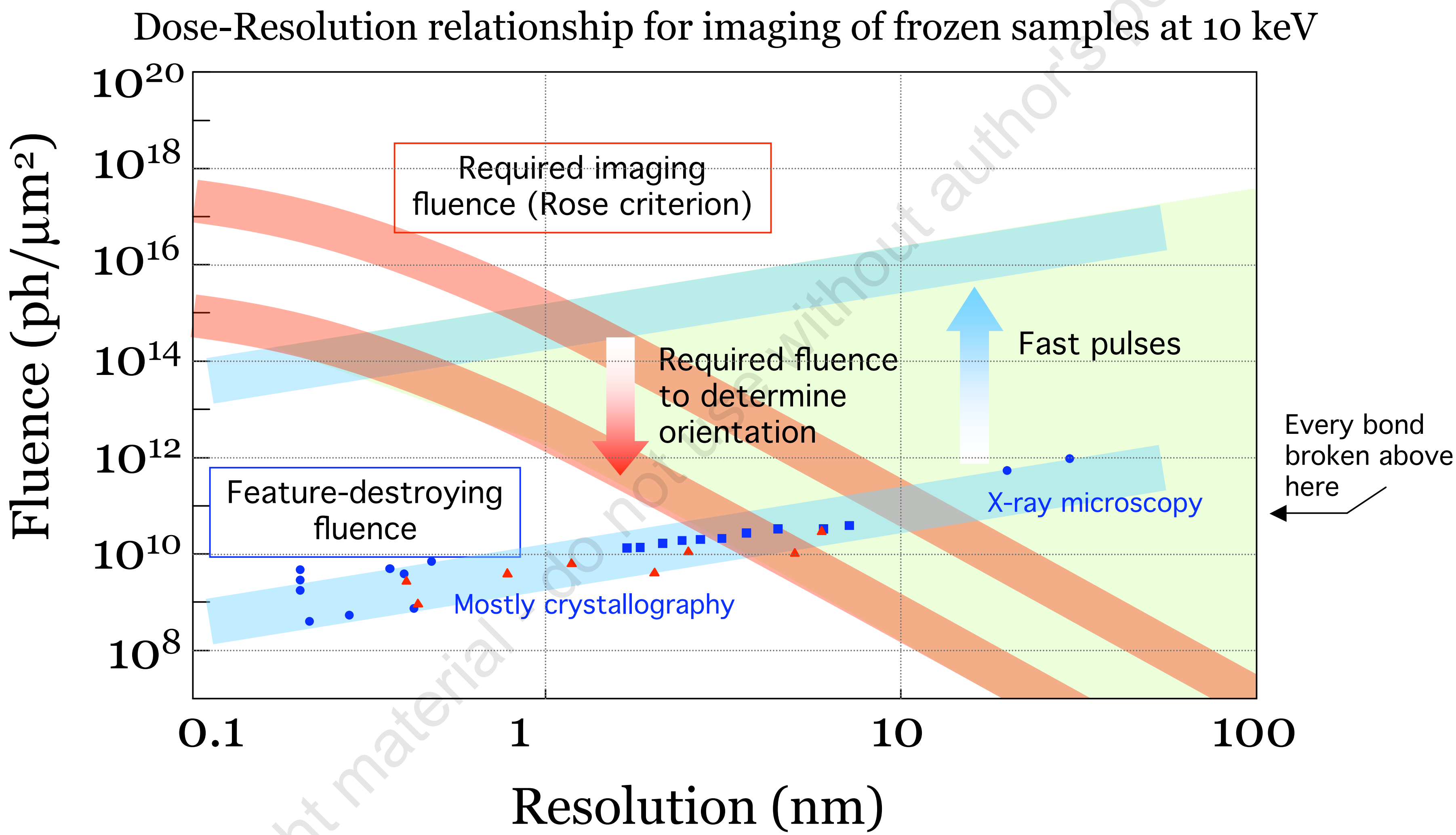
Henry Chapman, LLNL



CENTER for BIOPHOTONICS SCIENCE and TECHNOLOGY

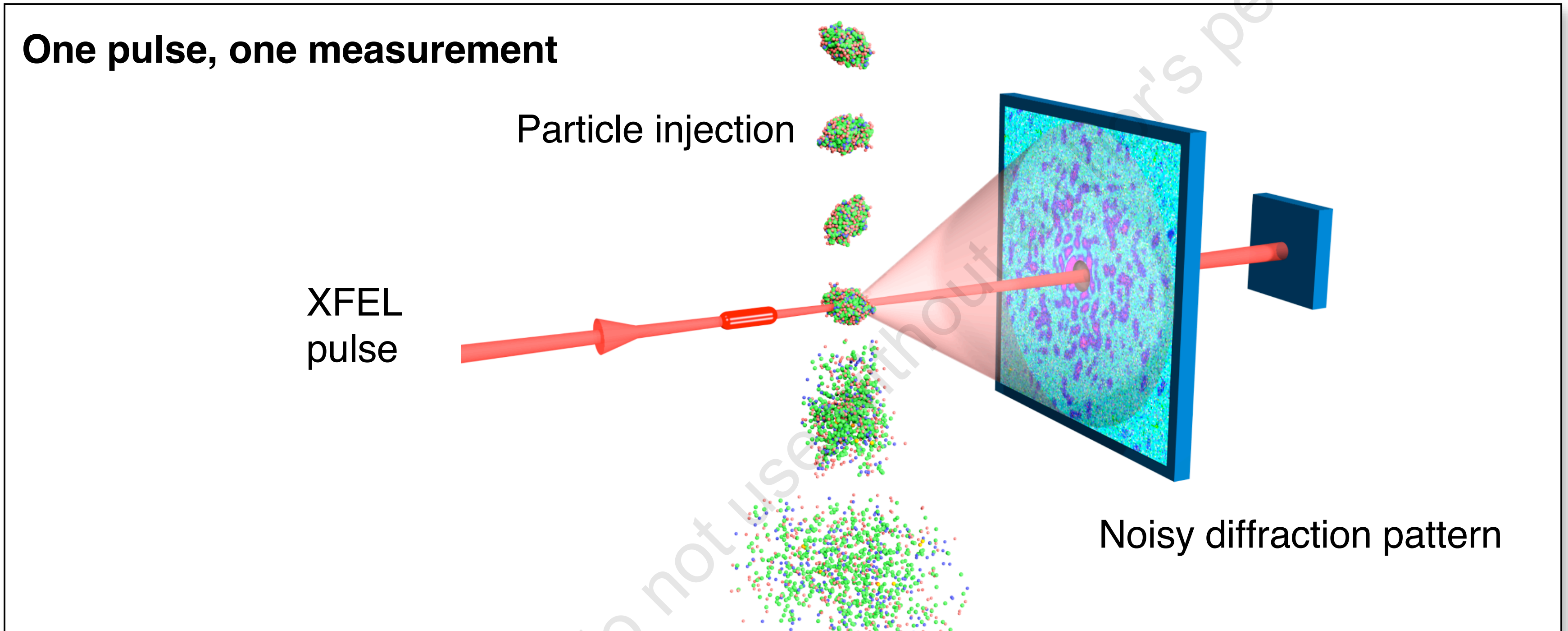
Gulliver workshop, May 2007

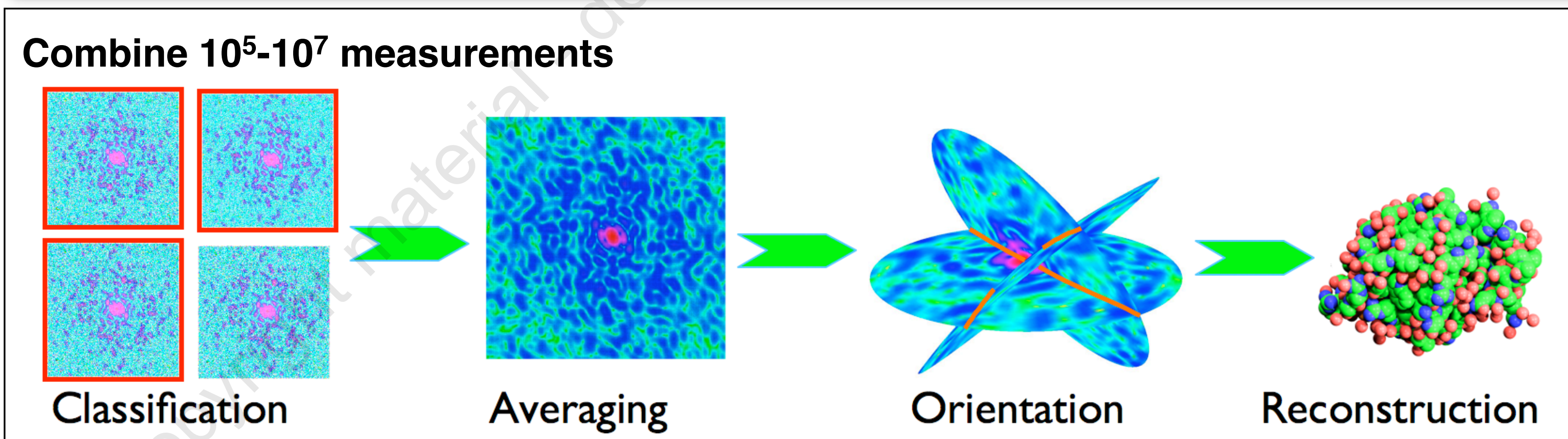
Imaging resolution is limited by radiation damage



Empirical data compiled by Malcolm Howells, LBL

X-ray free-electron lasers may enable atomic-resolution imaging of biological macromolecules





Ultrafast diffractive imaging is a versatile technique

Diffractive imaging recovers an image from diffraction intensities

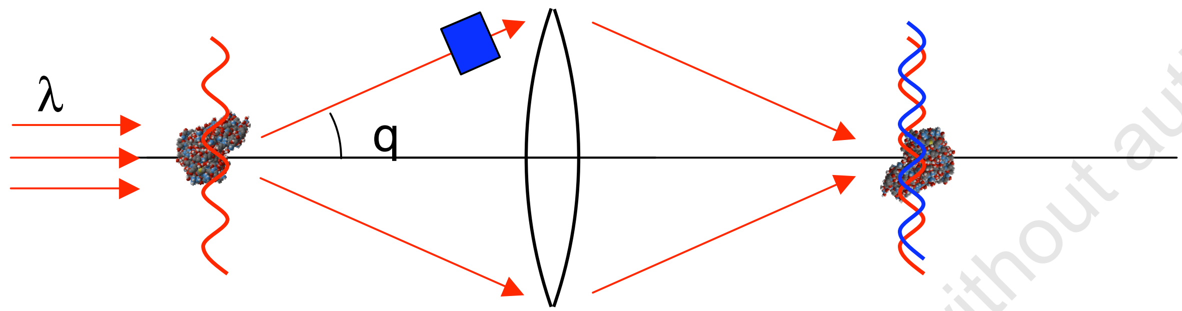
- No lens aberration or NA limitation
- No depth-of-focus limitation for tomography
- Quantitative phase contrast
- Numerical focusing
- Requires isolated objects
- Computationally demanding if no reference wave provided

Ultrafast X-ray pulses allow:

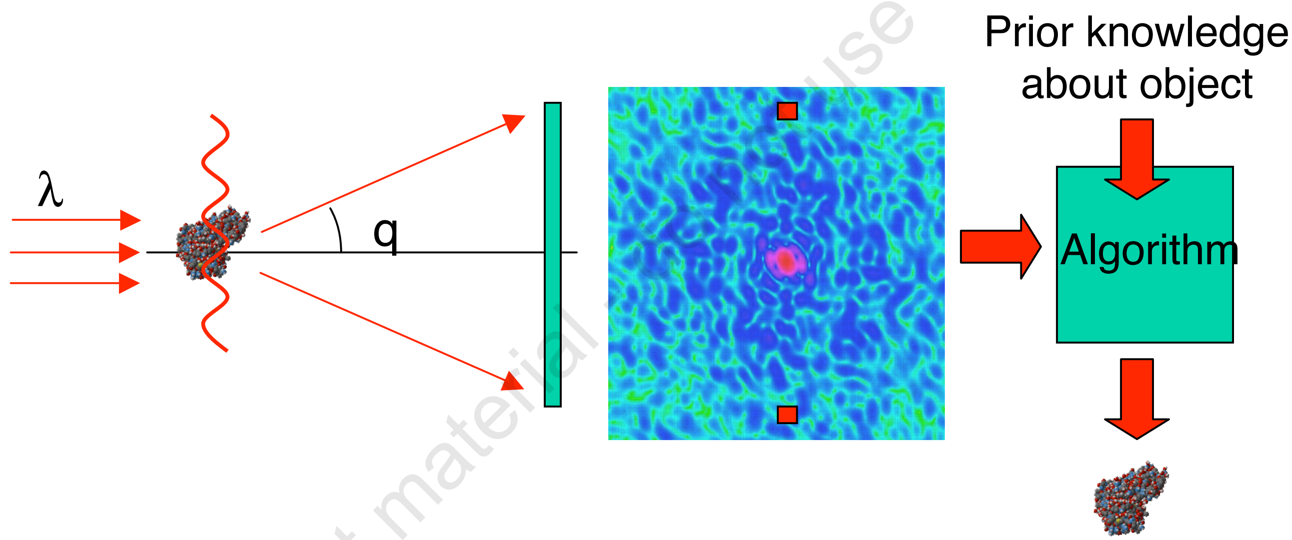
- Imaging beyond static radiation damage limits (to near-atomic resolution)
- Time-resolved imaging
- Imaging of injected wet cells
- Imaging of aligned particles
- Only one exposure per object

Coherent diffractive imaging is lensless

Use a computer to phase the scattered light, rather than a lens



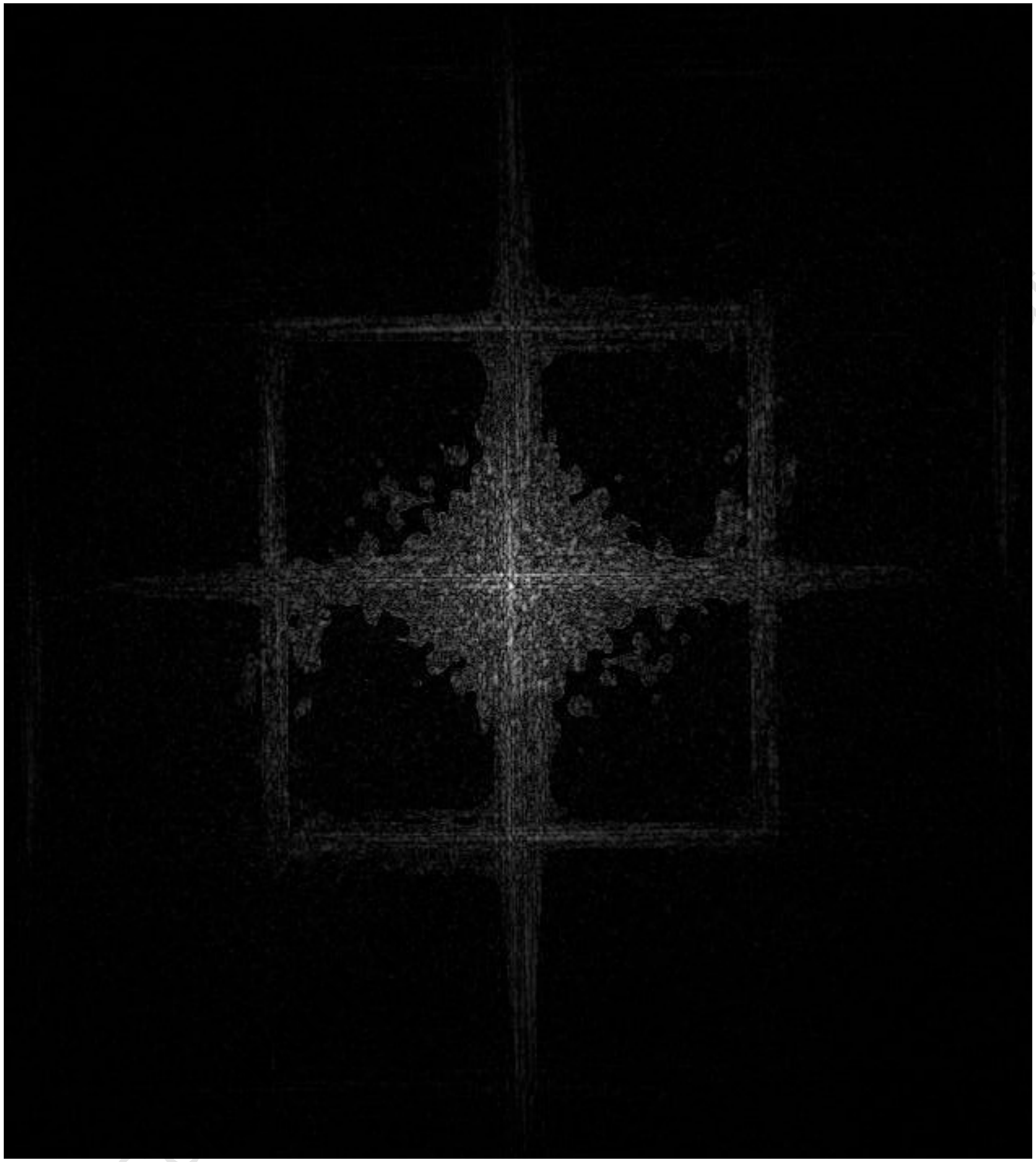
A lens recombines the scattered rays with correct phases to give the image

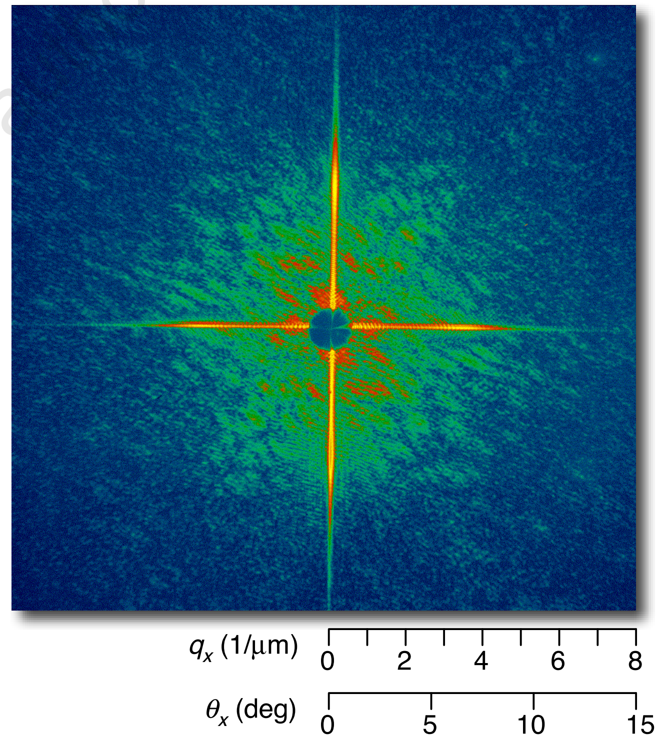


An algorithm finds the phases that are consistent with measurements and prior knowledge

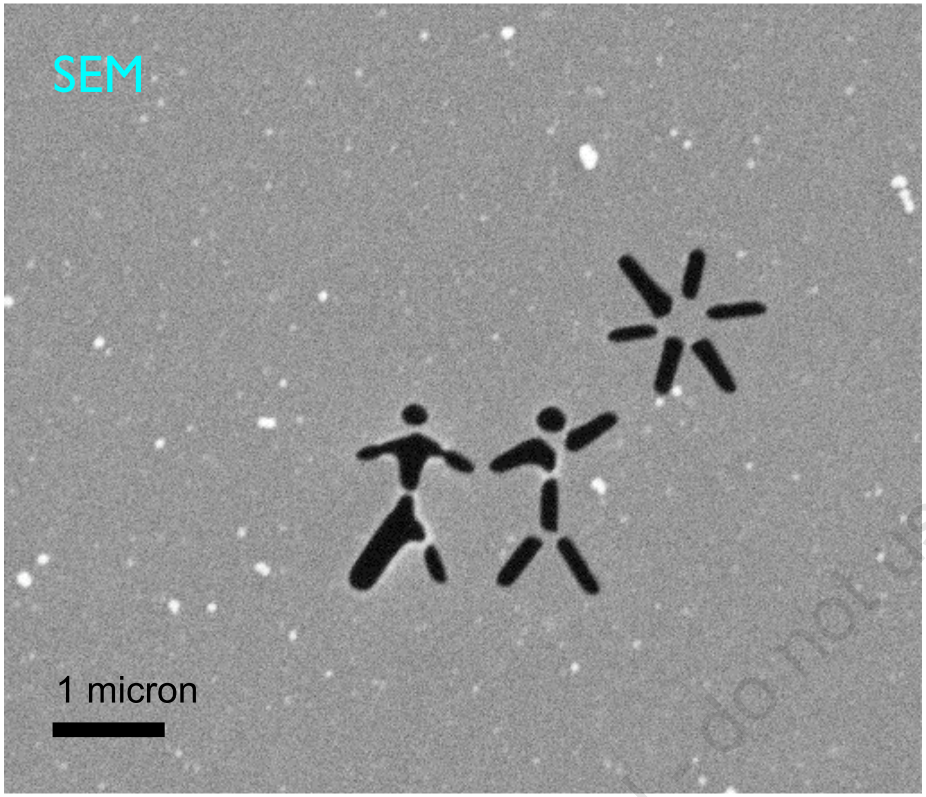
Resolution: $\delta = \lambda / \sin\theta$

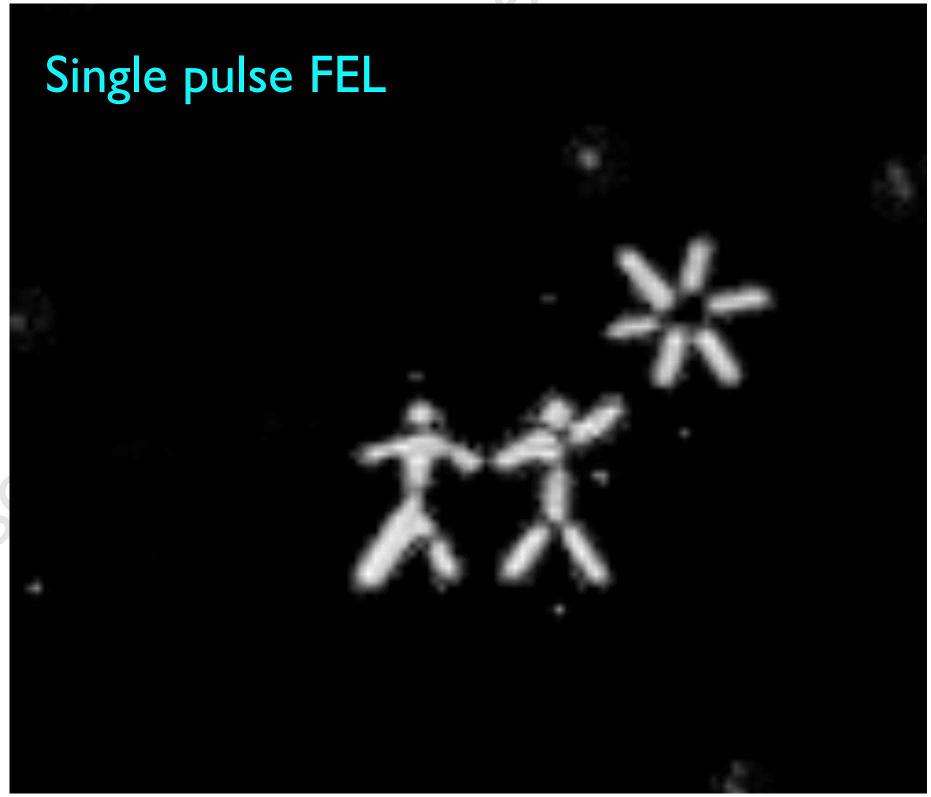
We perform ab initio image reconstruction with our "Shrinkwrap" algorithm



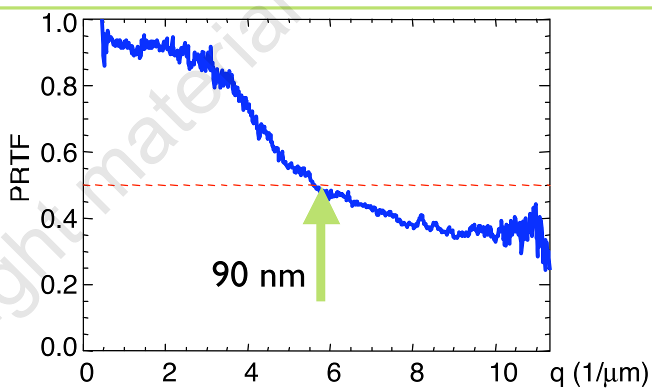


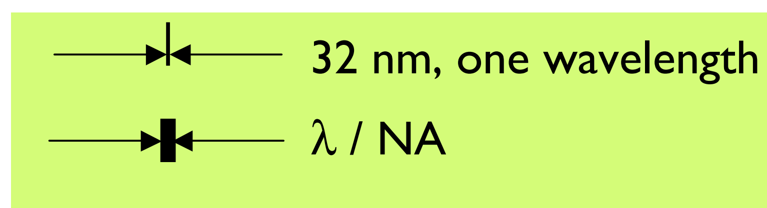
The reconstruction is carried out to the diffraction limit of the 0.26 NA detector



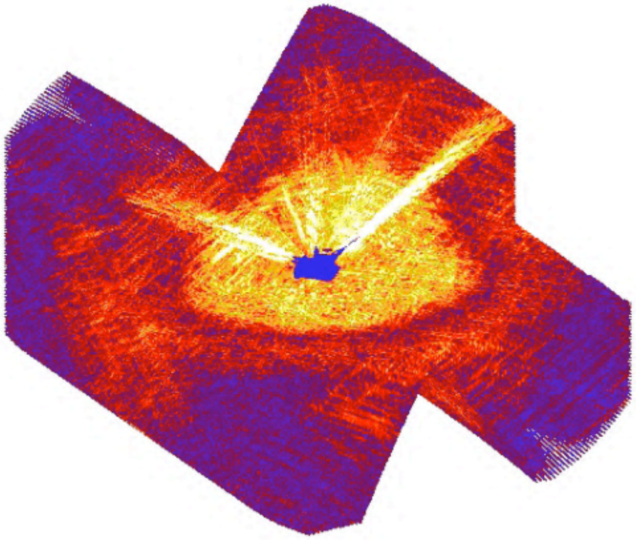


Phase-retrieval transfer function gives an estimate of the resolution of the reconstructed image

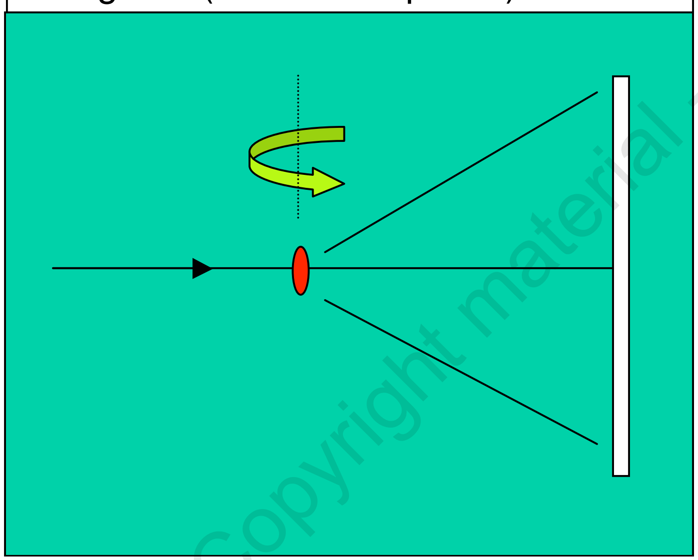




We have reconstructed a 3D X-ray image of a non-crystalline object at 10 nm resolution



Coherent X-ray diffraction data, rotating the sample -70 to +70 degrees (5×10^8 data points)

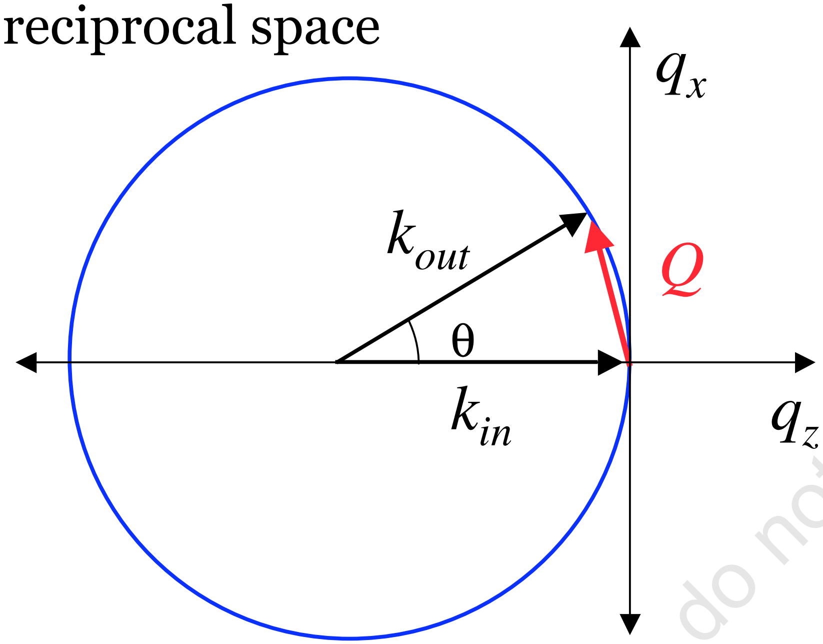


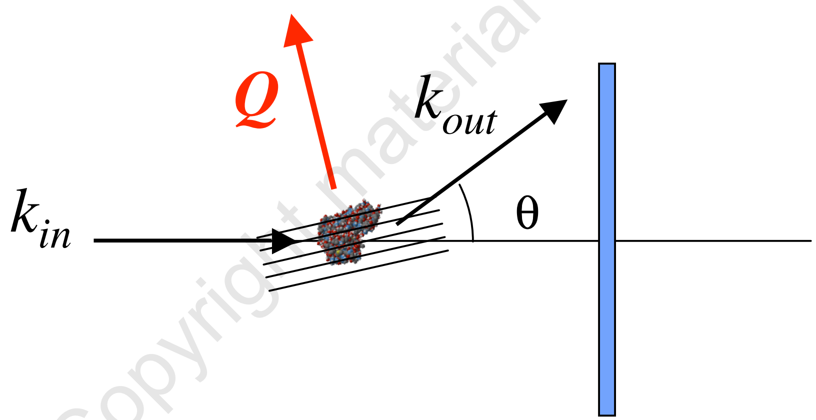
Coherent X-ray diffraction data λ =1.6 nm, from a sample of 50-nm gold spheres arranged on a pyramid

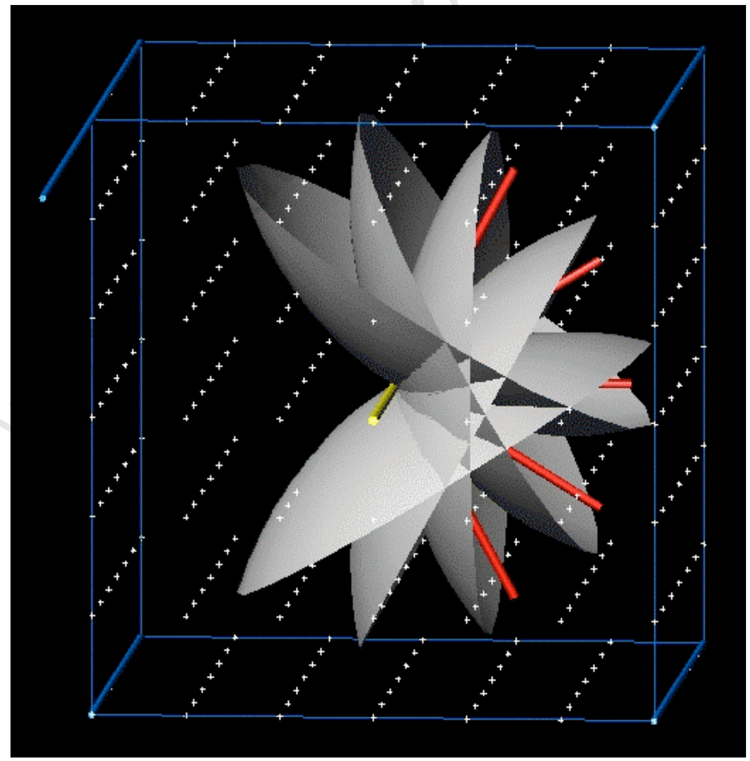
Complete image reconstruction achieved, without any prior knowledge, using our "**shrinkwrap**" algorithm, **parallelized** for 3D on 32-CPU cluster. Resolution = 10 nm

3D reconstruction is achieved by Fourier synthesis

One diffraction pattern gives information on the Ewald sphere in reciprocal space

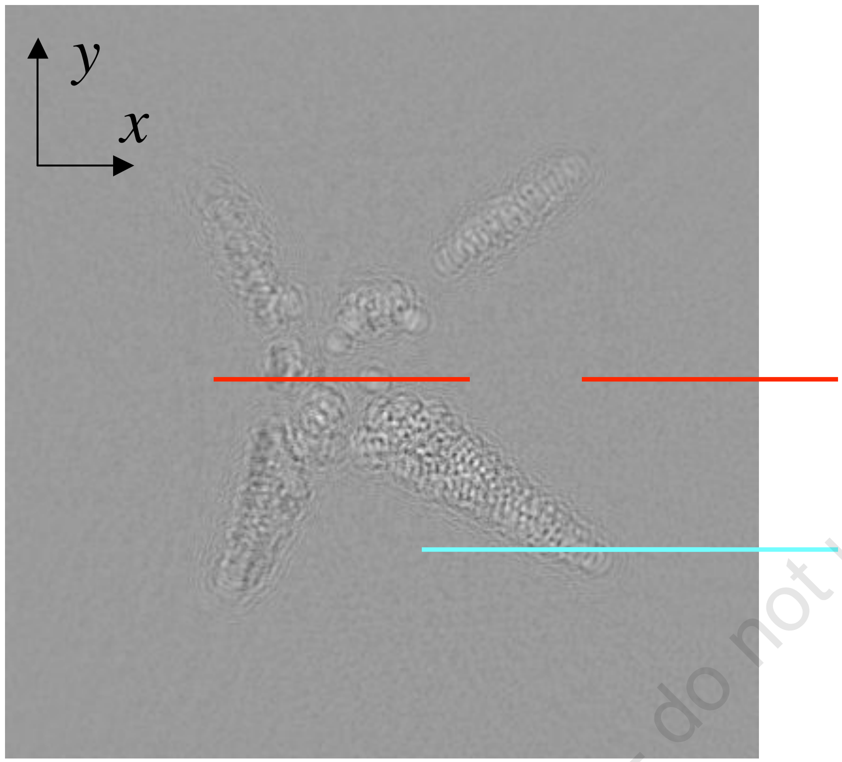


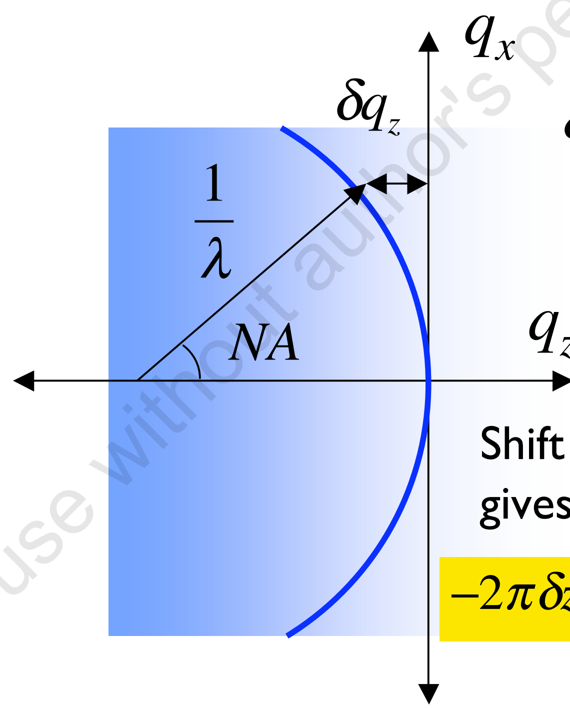




Rotating a sample about one axis only gives imperfect data filling in Fourier space

2D single-view images have depth information





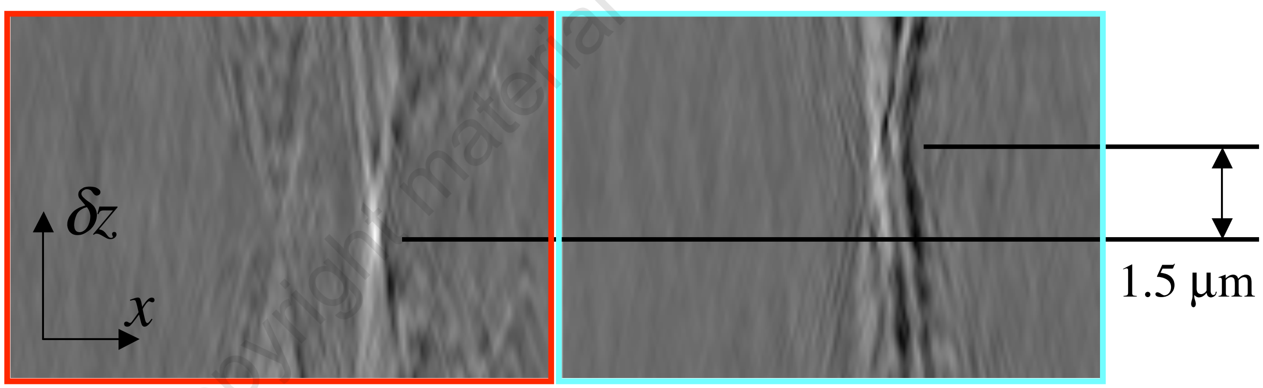
$$\delta q_z = -\frac{\lambda}{2}\left(q_x^2 + q_y^2\right)$$

$$q_x = NA/\lambda$$

Shift theorem: shift of δz gives a phase shift of

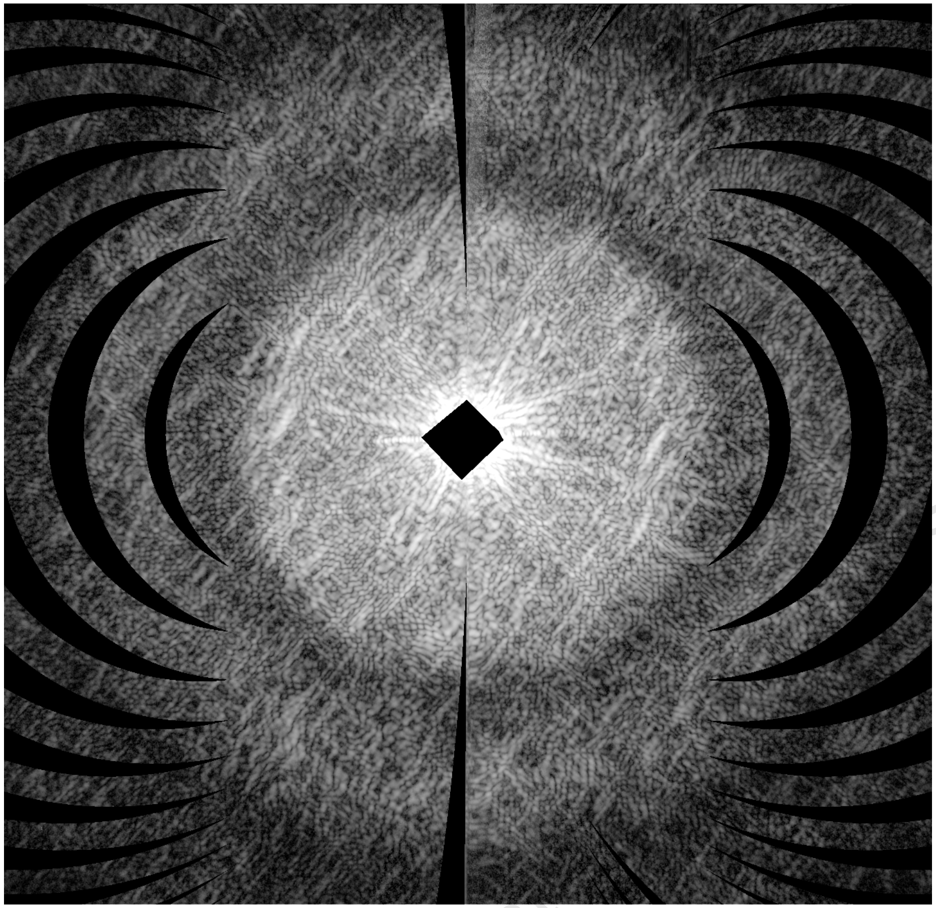
$$-2\pi\delta z q_z = \pi\delta z\lambda\left(q_x^2 + q_y^2\right)$$

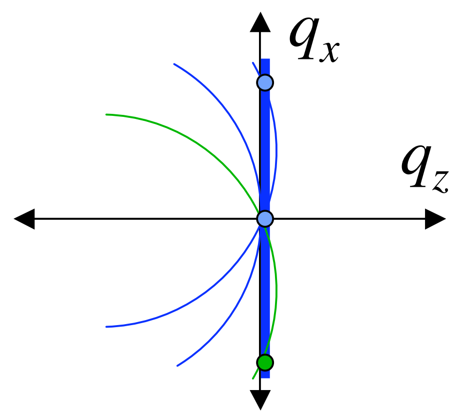
$$D = 1/\delta q_z = -\frac{2\lambda}{NA^2}$$



Images of objects thicker than *D* will differ from the projection image

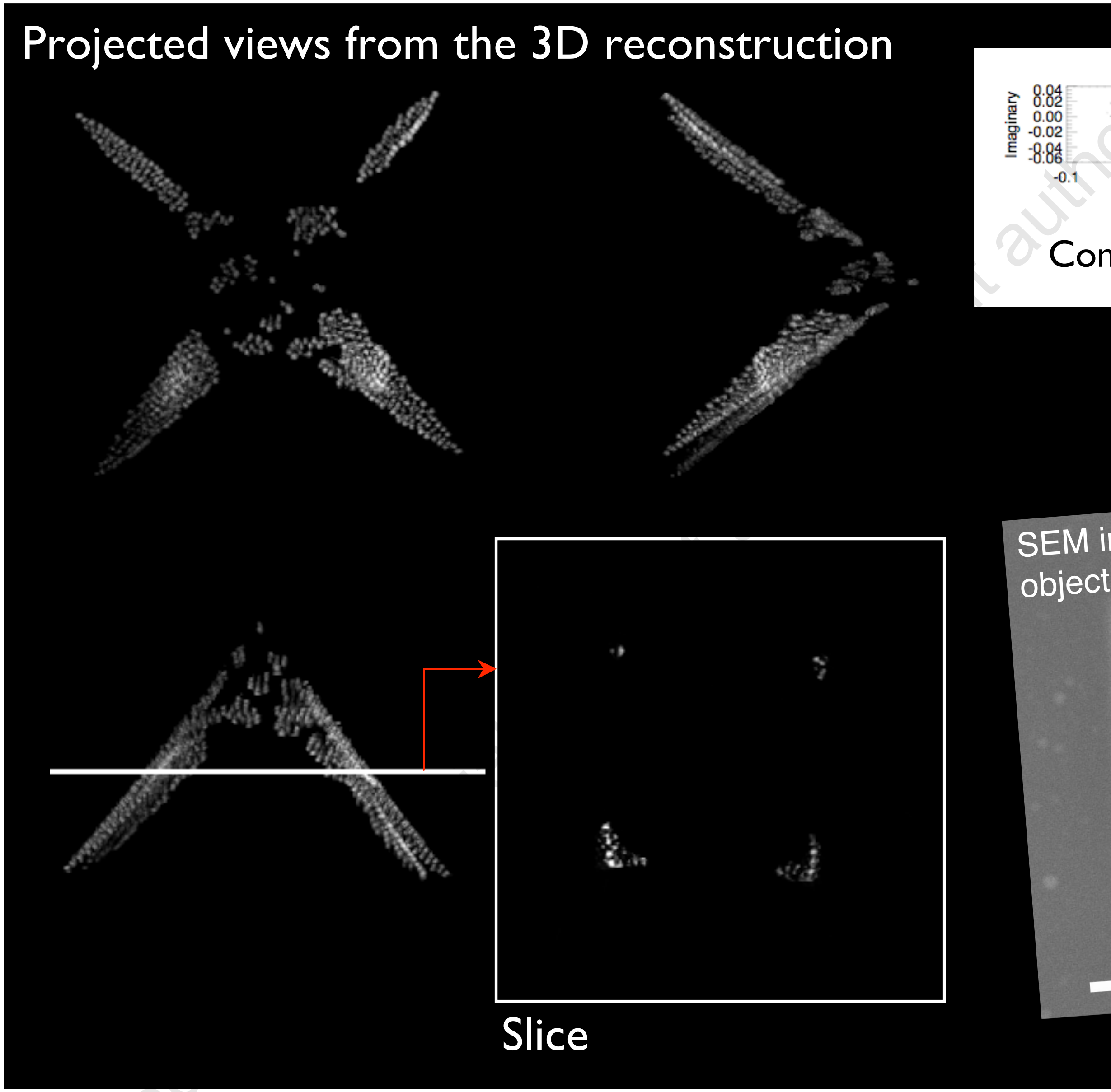
True 2D projection images can be formed from a central section of the 3D diffraction data

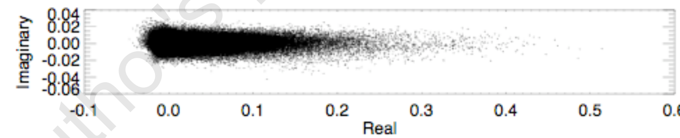




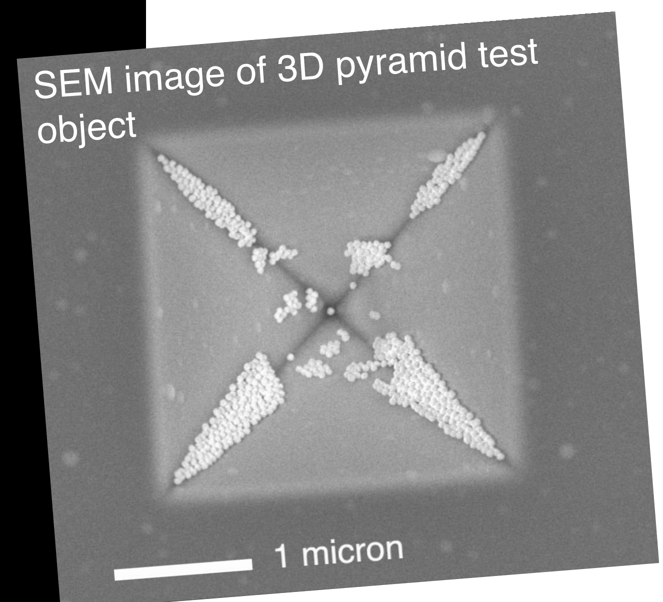
A true projection image is obtained from a **plane central section** of the 3D diffraction data. Data must be collected at many object orientations to achieve this

We have performed full 3D reconstruction with a positivity constraint



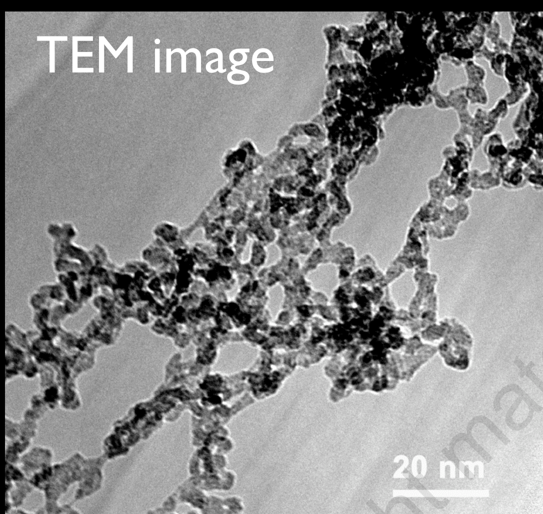


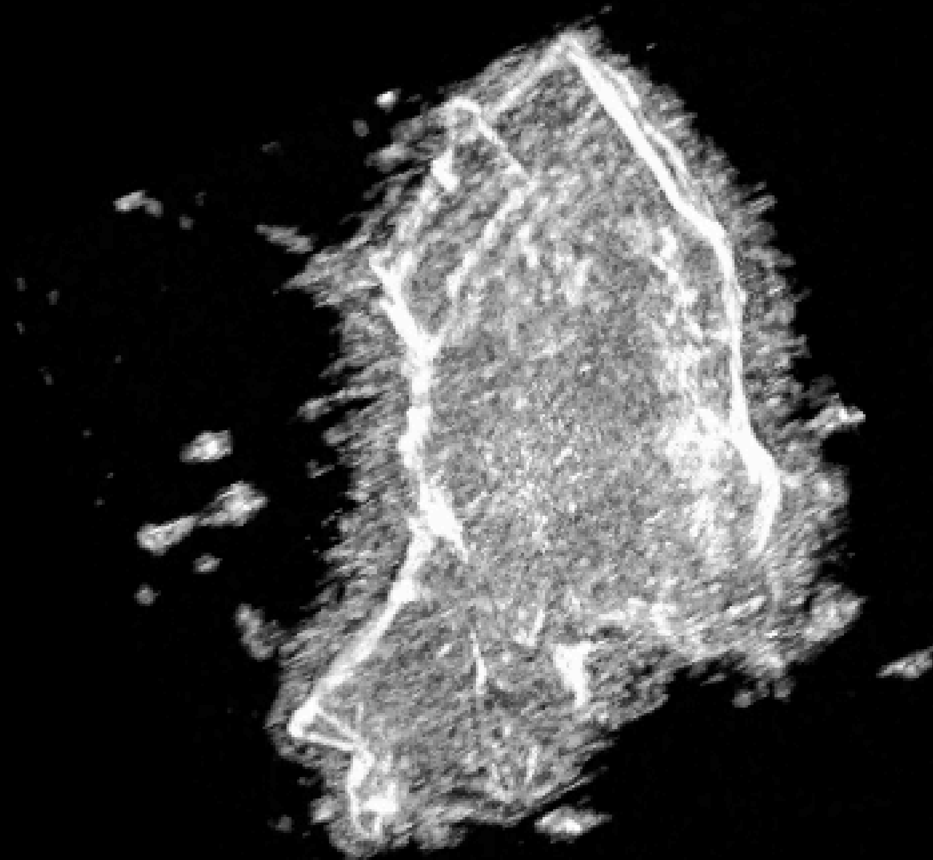
Complex amplitudes of the image



We have performed 3D X-ray imaging of Aerogel foam at 10 nm resolution

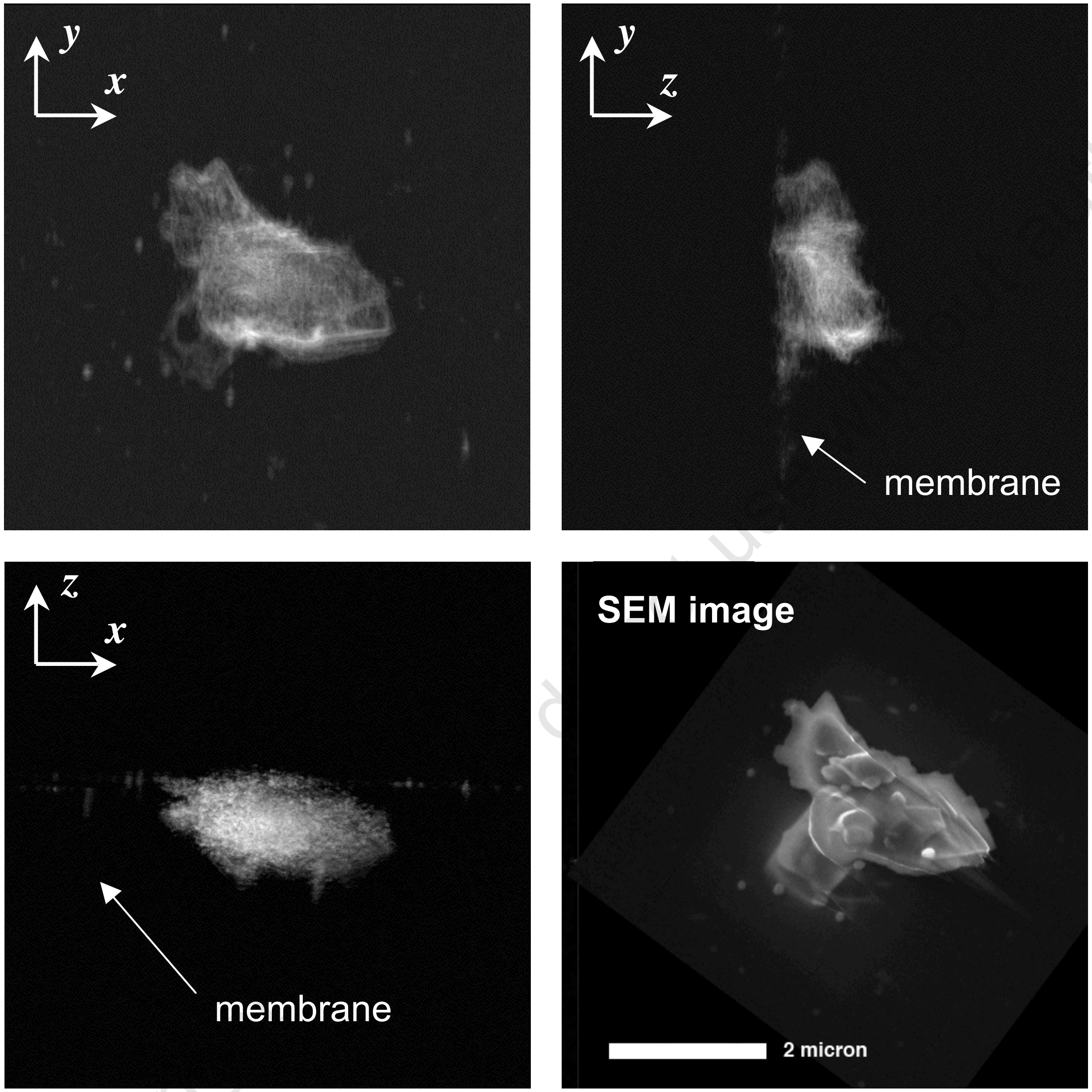
Analysis of the 3D image revealed anisotropy in the structure. Other characterization techniques (TEM, SAXS) could not reveal this





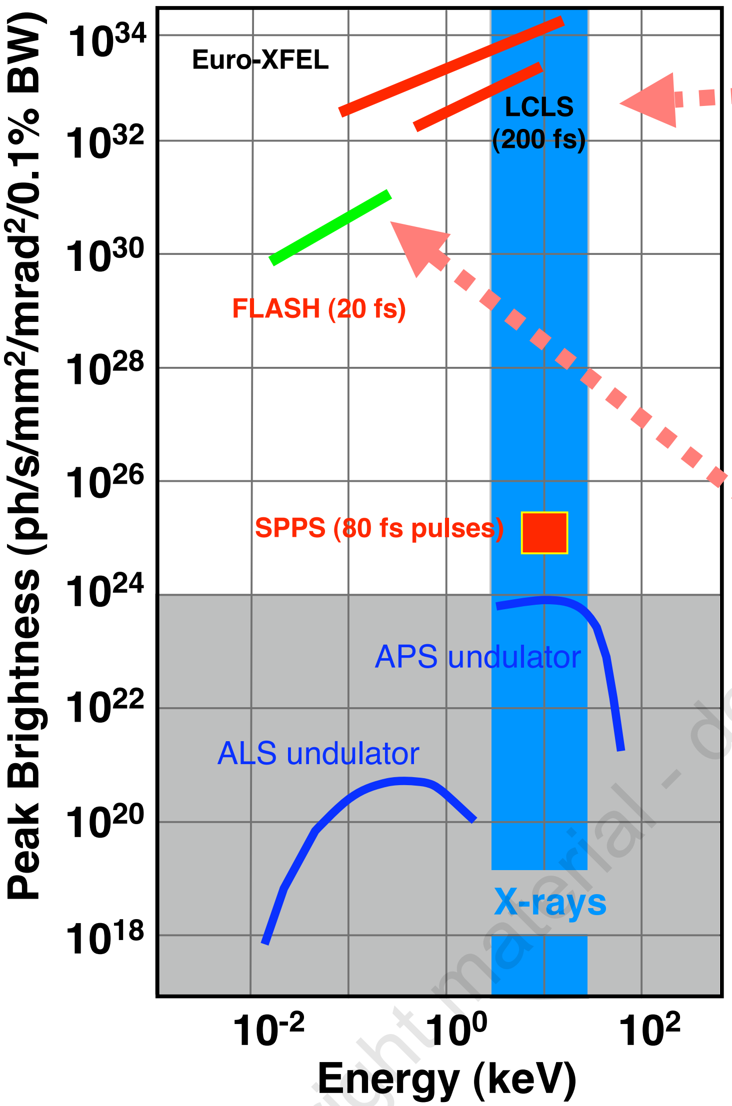
1 micron

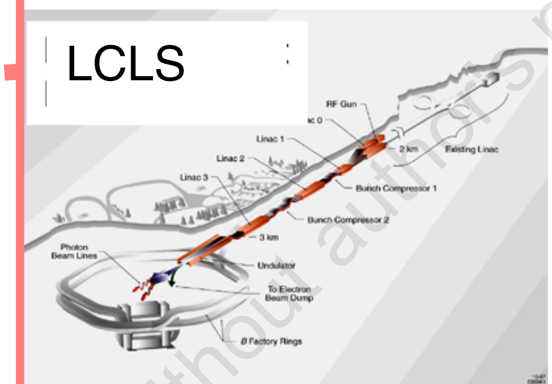
We are using our unique 3D X-ray imaging capabilities to investigate aerogel structure



Ta_2O_5 aerogel (100 mg/cm^3), reconstructed images along orthogonal views.

We are entering a new era in x-ray science





operational 2009

8 keV, 200 fs, 10^{12} photons

Linac Coherent Light Source, SLAC, Stanford

operational now

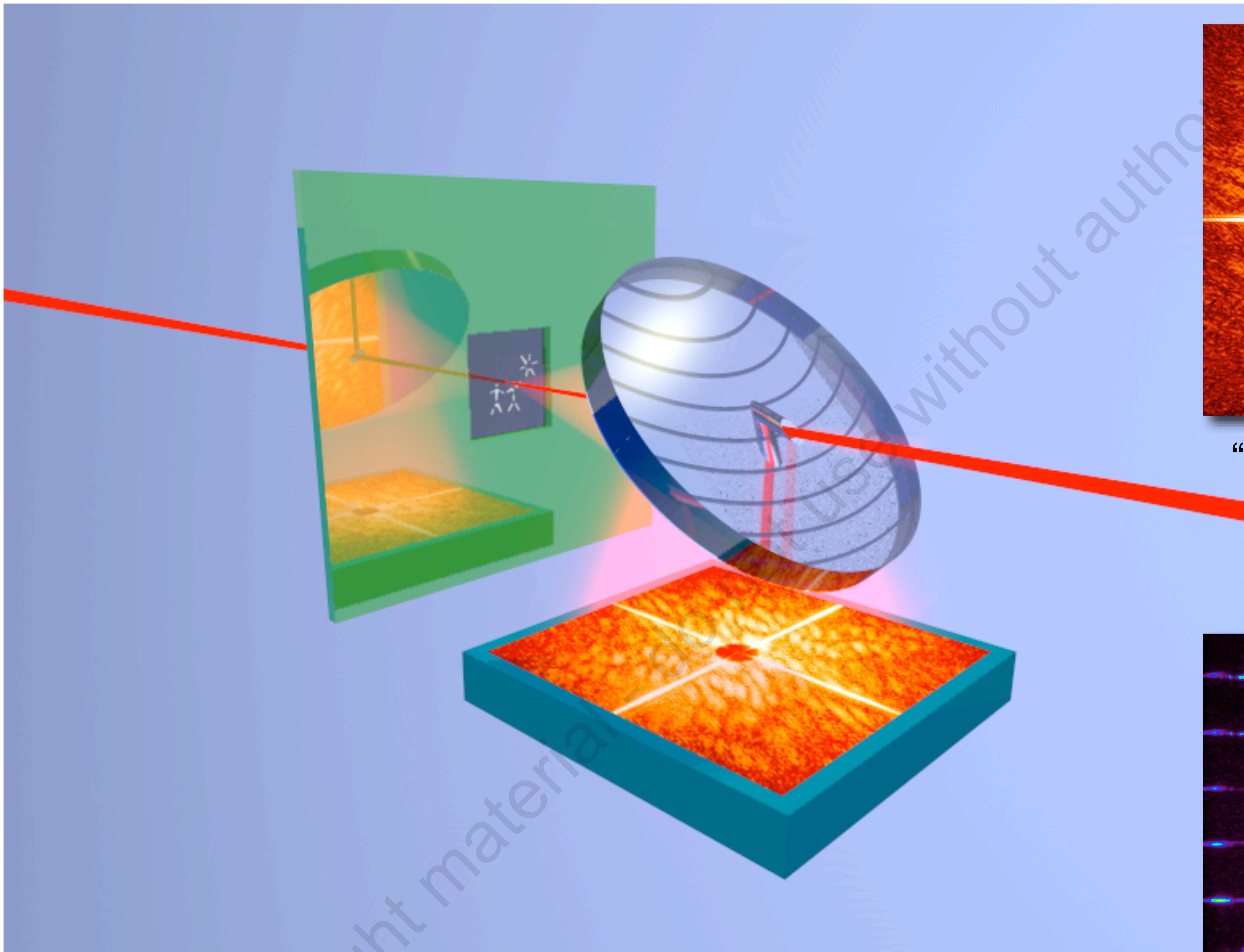
Current: 100 eV, 20 fs

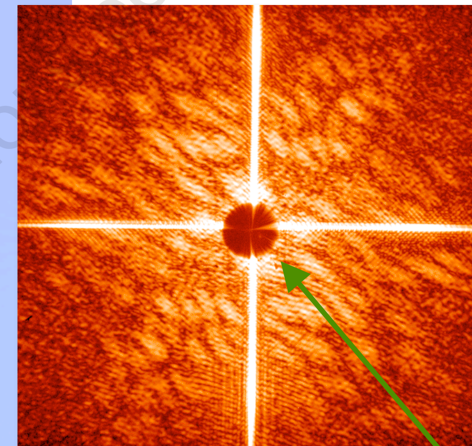
200 eV, 80 fs, 10^{13} photons

Tesla Test Facility DESY, Hamburg

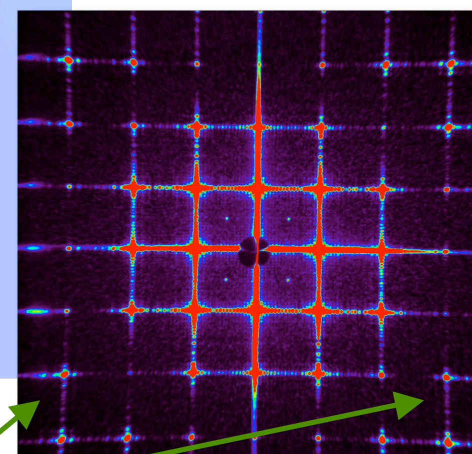
APS=Advanced Photon Source (ANL)
ALS=Advanced Light Source (LBNL)

Our diffraction camera can measure forward scattering close to the direct soft-X-ray FEL beam



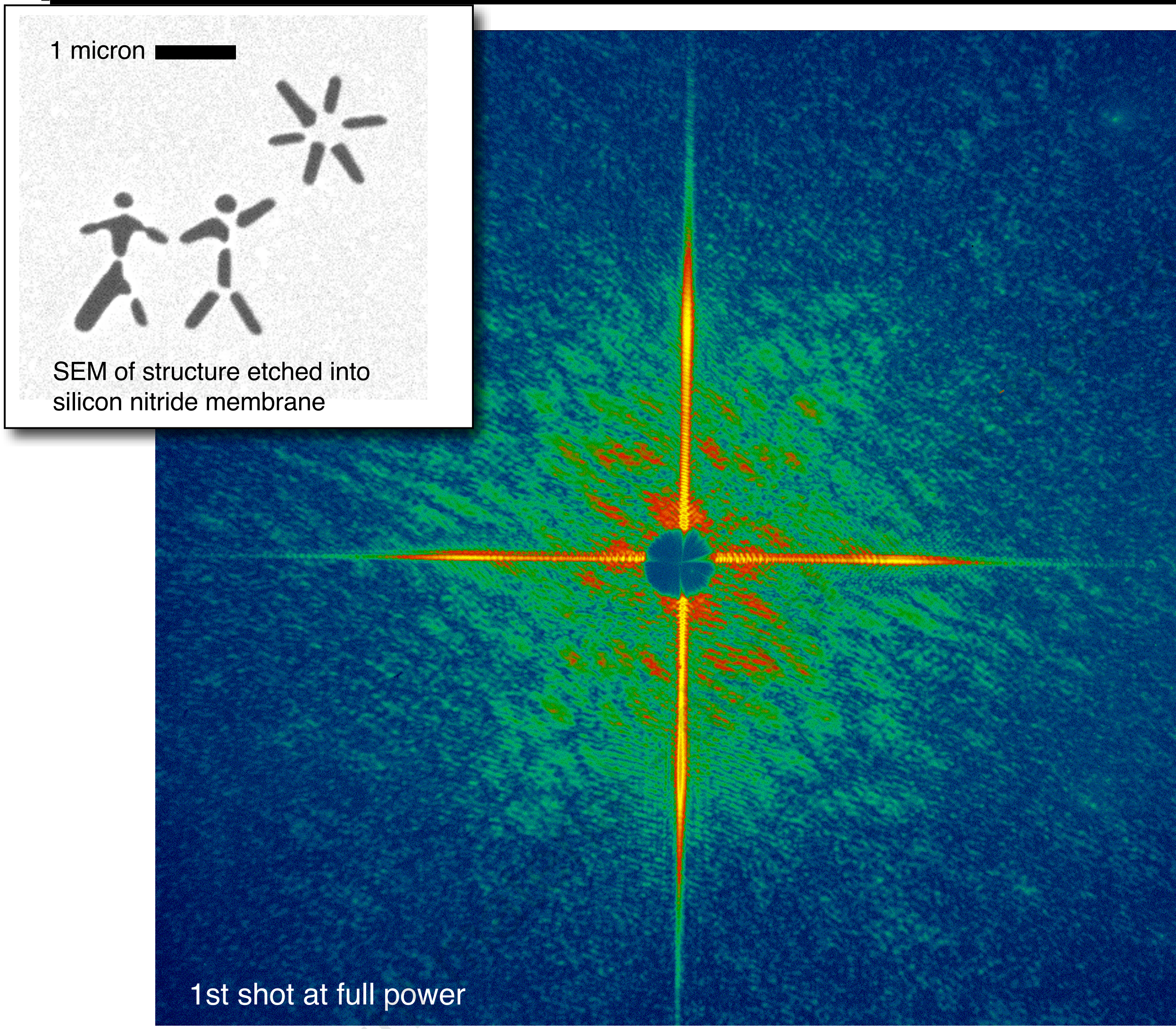


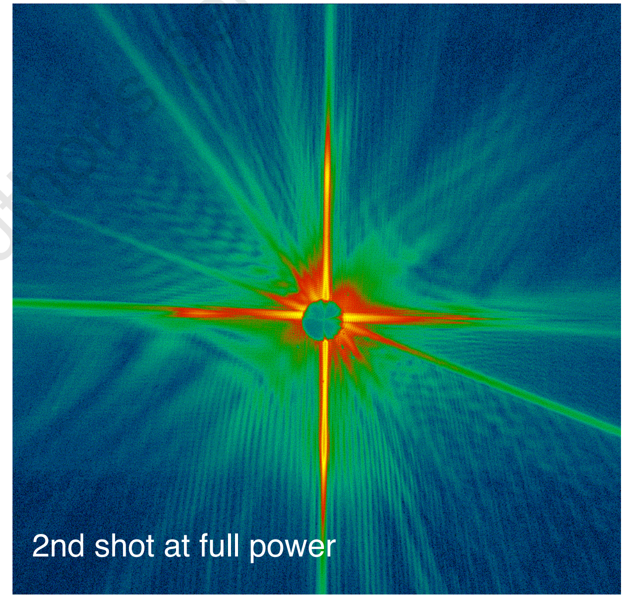
“Soft edge” prevents any scatter from the hole

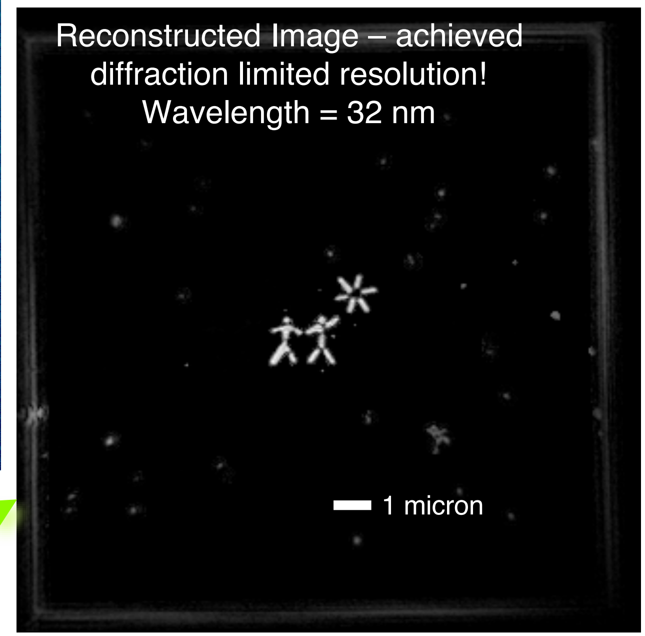


Multilayer reflectivity is uniform across the 30° to 60° gradient

Image reconstructed from an ultrafast FEL diffraction pattern



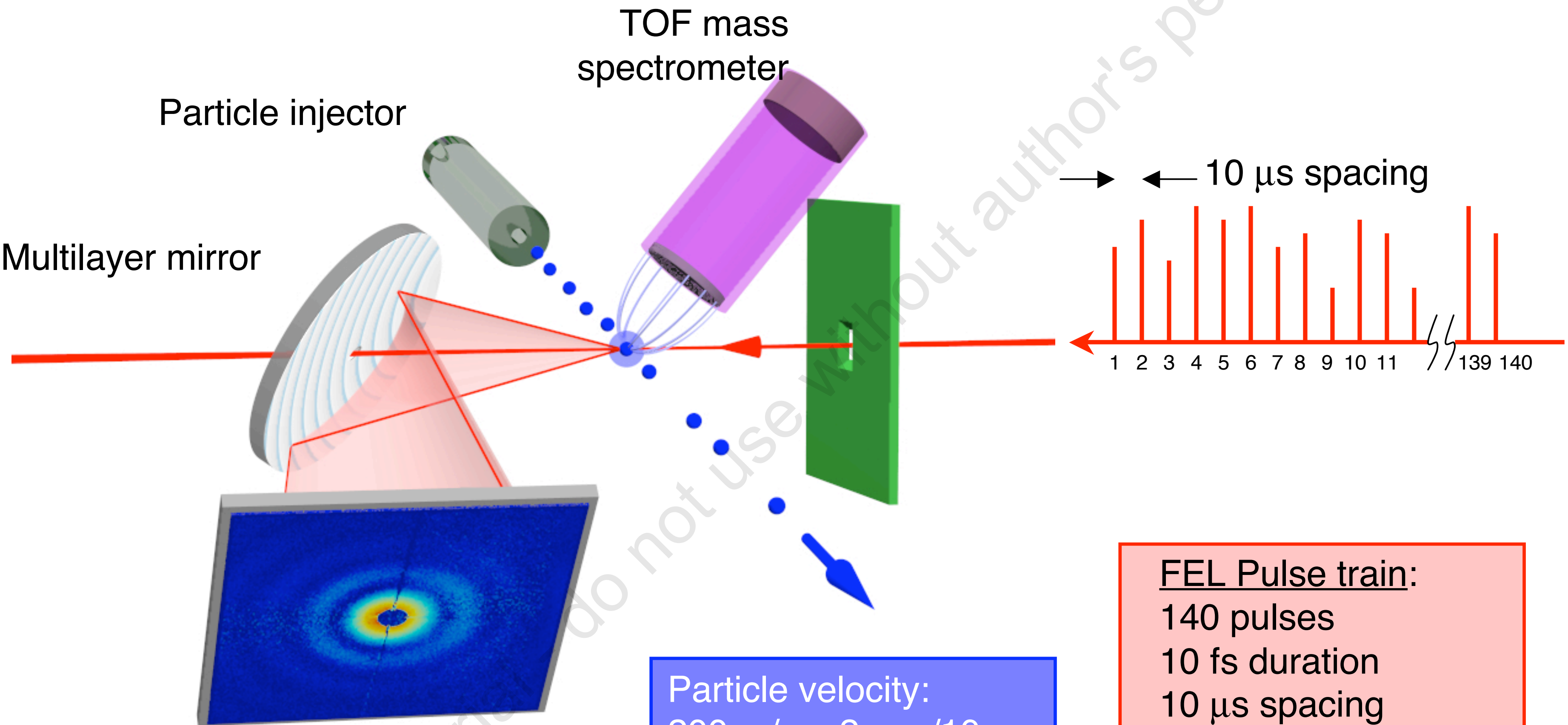




Edge of membrane support also reconstructed

Chapman et al. Nature Physics **2** 839 (2006)

Single-particle FEL diffraction of “on-the-fly” particles has been demonstrated for the first time



Particle velocity:
200 m/s = 2 mm/10 μs
(c.f. 20 μm beam)

FEL Pulse train:
140 pulses
10 fs duration
10 μs spacing
~20 μJ/pulse
13.5 nm wavelength

Single particle diffraction pattern (one hit + 269 no-hits)

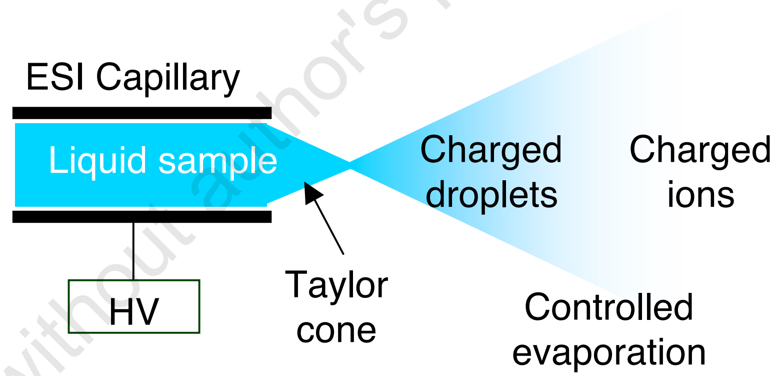
We generate particle streams by electrospray aerosol generation and aerodynamic focusing

Challenges:

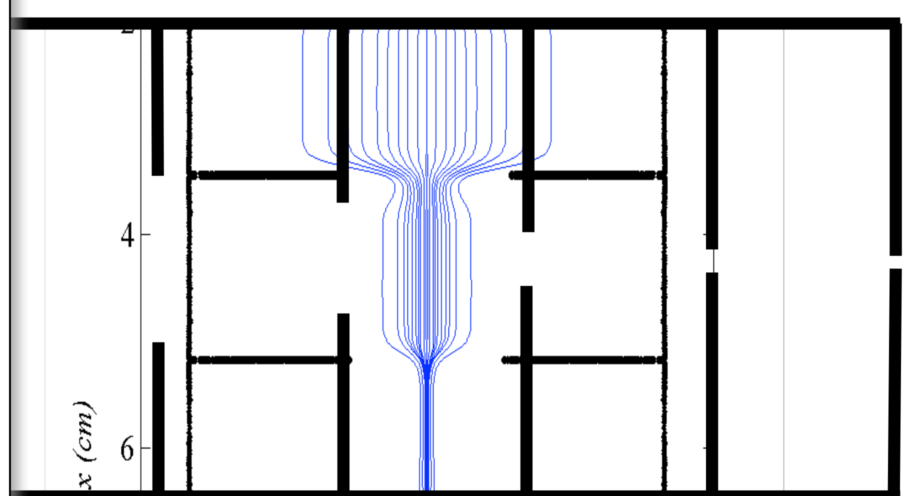
- High enough particle density in the beam
- Having a pure sample
- Keeping molecules in "native" conformation
- Diagnostics and control of particle trajectories

Electrospray Approach:

- Charged-reduced electrospray and aerodynamic and electrostatic trajectory control
- Purification through size selection by mobility
- Extremely sensitive charge detection and mass detection to give status of FEL-particle interaction

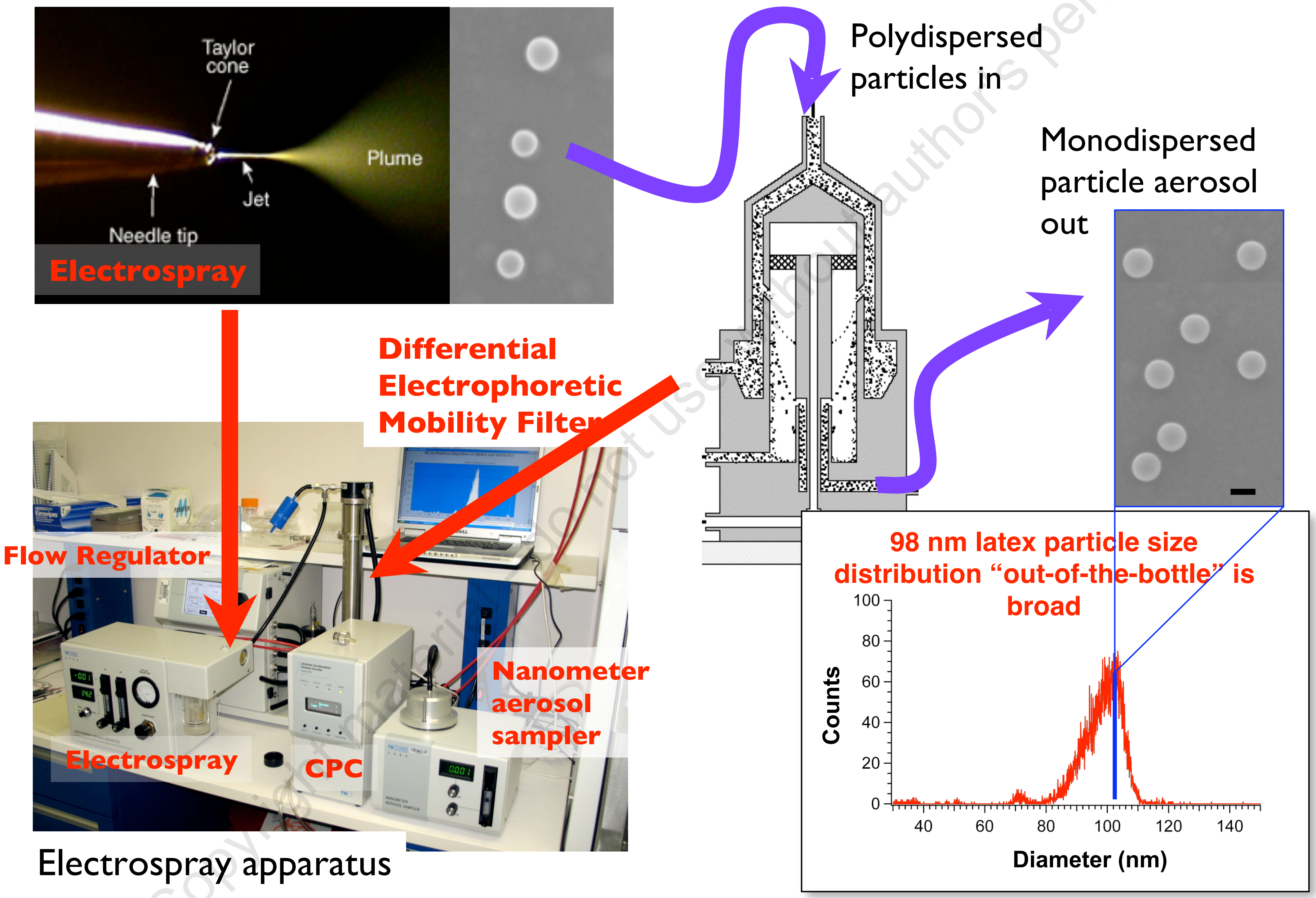


Electrospray



Aerodynamic lens

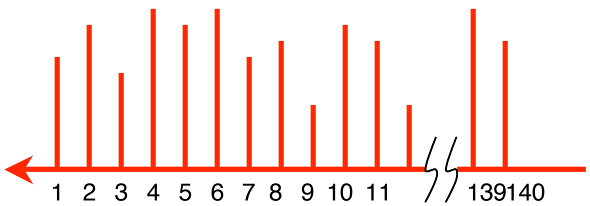
The electrospray system is extremely versatile and we can select particle size and charge



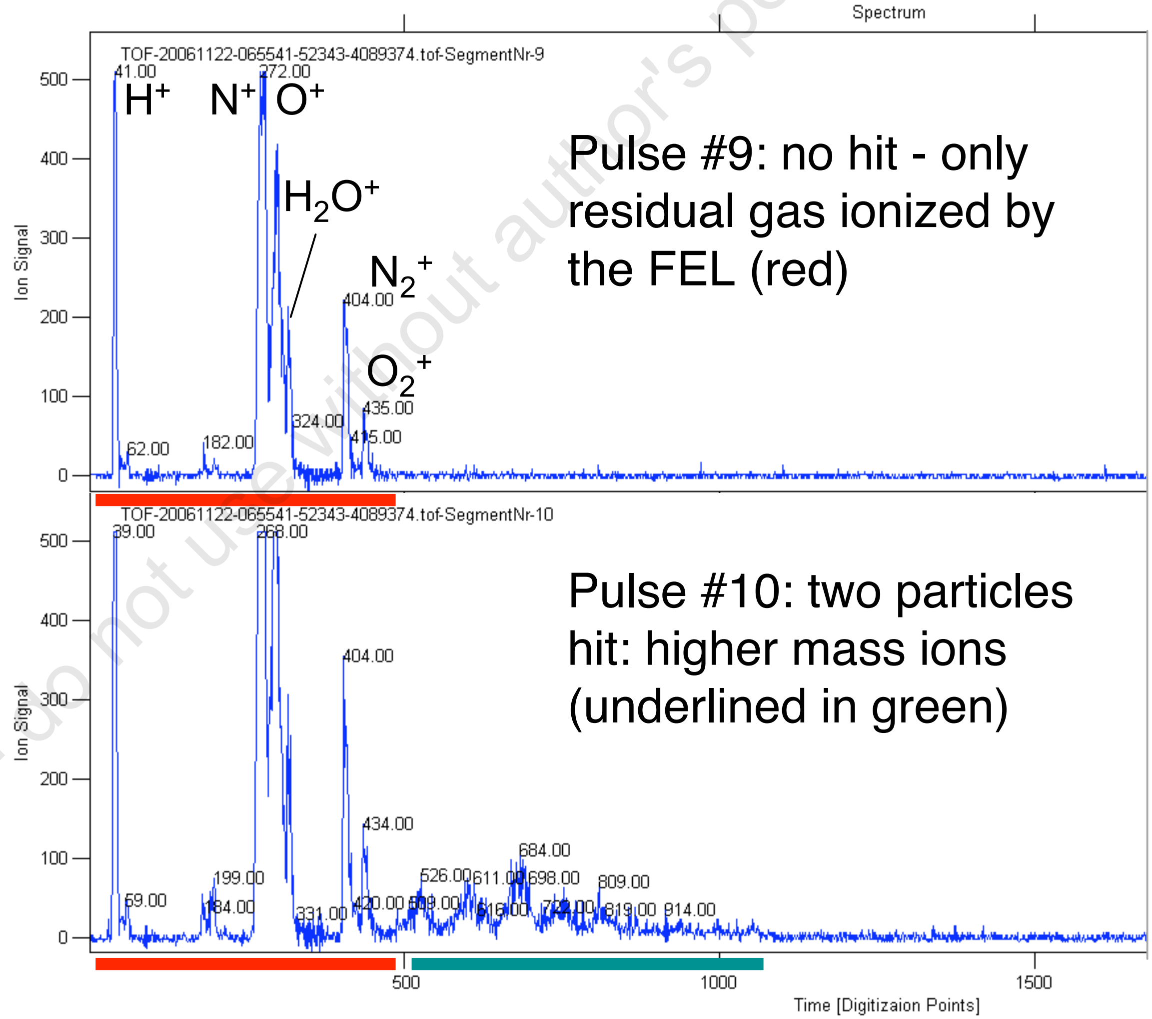
Electrospray apparatus

A mass spectrum is recorded every FEL pulse

10 fs FEL pulses at 10 µs spacing



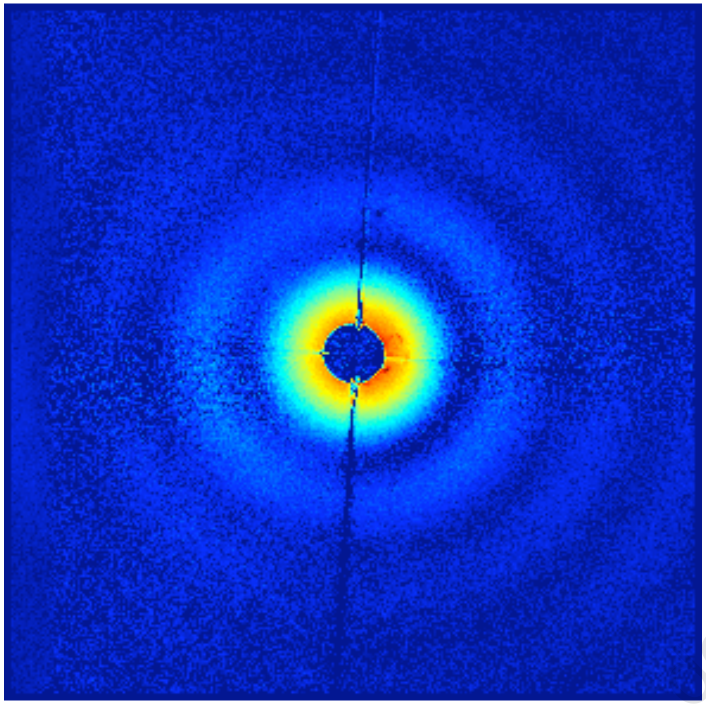
FEL Pulse train:
140 pulses
10 fs duration
10 µs spacing
5 Hz
~20 µJ/pulse
13.5 nm wavelength



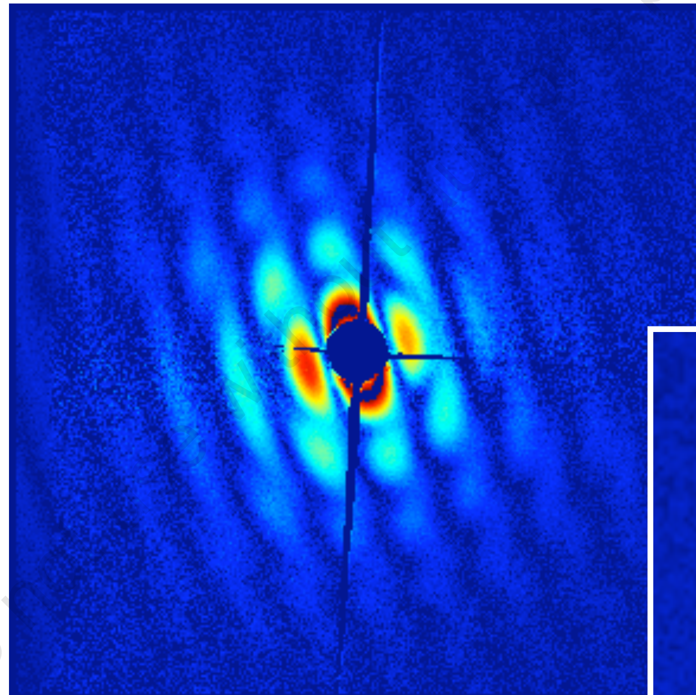
The mass spectra show which pulse in the pulse train had hit and how.

Single-particle FEL diffraction of “on-the-fly” particles has been demonstrated for the first time

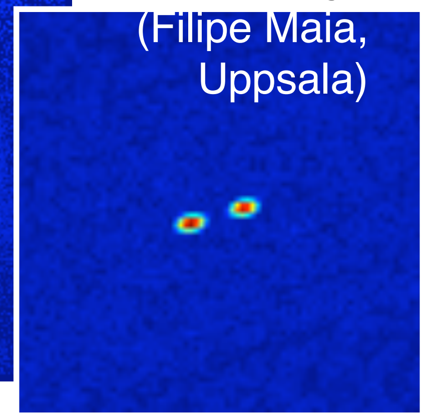
Single ~200 nm particle

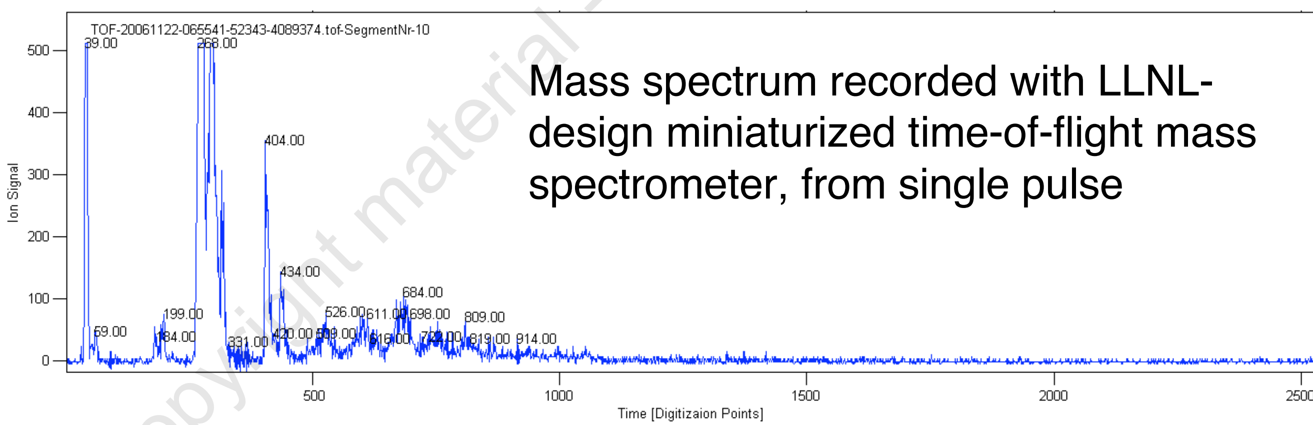


Two particles hit by the one pulse



Reconstructed image (Filipe Maia, Uppsala)

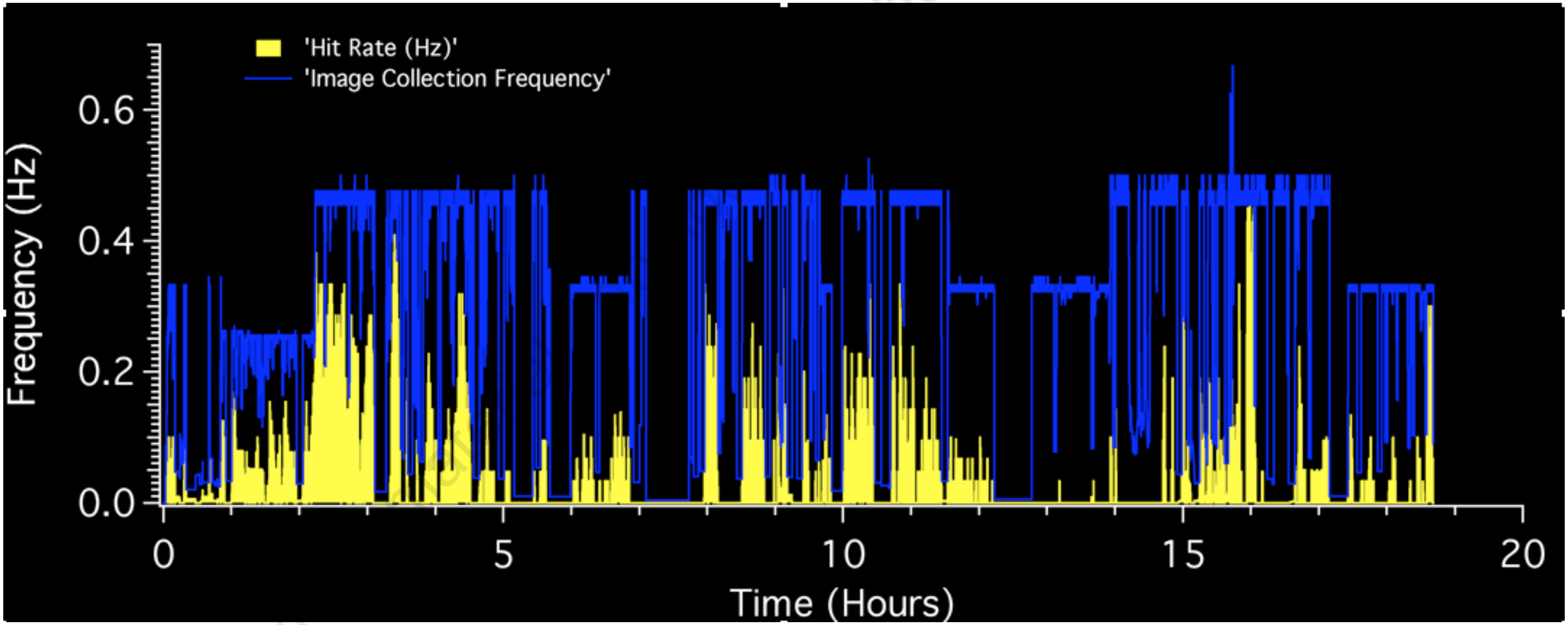




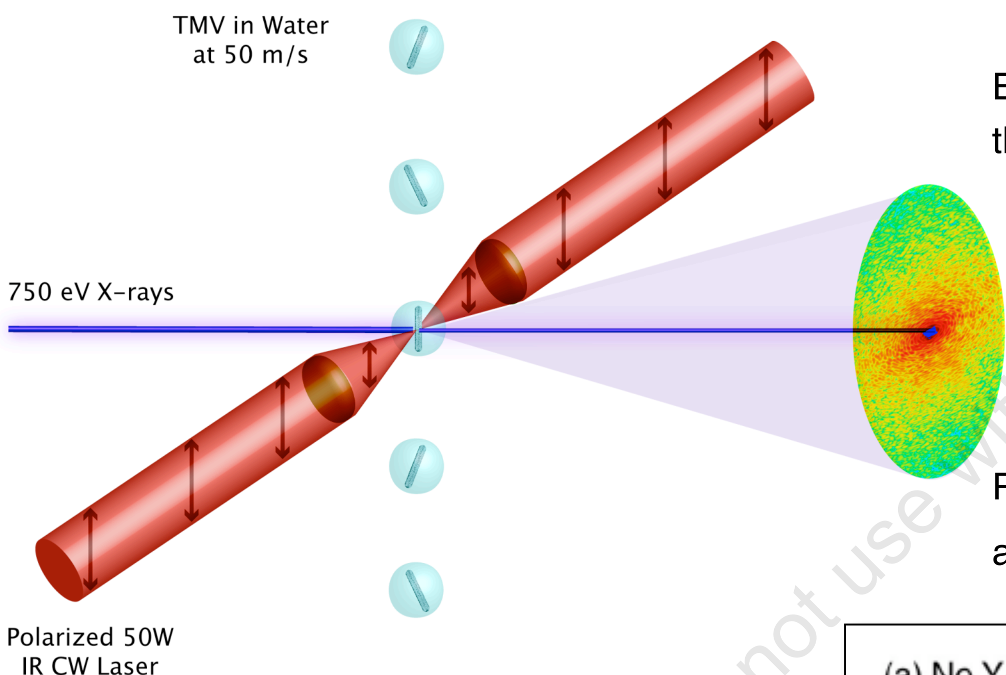
Mass spectrum recorded with LLNL-design miniaturized time-of-flight mass spectrometer, from single pulse

The particle injection system operates at high efficiency

24hr shift, 18.68 hours of data collection
26 sample changes, 14 different samples
16639 patterns collected, 1873 patterns contained particle scattering
11.6% of patterns contained particles
0.05 Hz average hit rate, maximum >0.5 Hz (camera limited)



Laser alignment will help establish molecular imaging at XFELs



Equipartition of rotational potential energy with thermal energy gives

$$\left\langle \Delta\theta^2 \right\rangle = \frac{T}{3\times10^{-8}\, I\Delta\alpha}$$

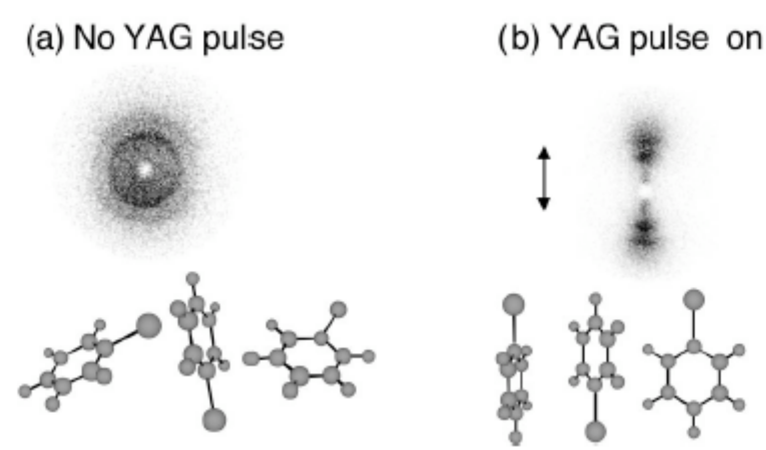
T - temperature in K
I - laser power in W/cm^2
$\Delta\alpha$ - polarizability anisotropy in nm^3

Resolution is limited by the degree of alignment: $d = (L/2)\,\Delta\theta$

J.C.H. Spence and R.B. Doak, Phys. Rev. Lett. ***92****, 198102 (2004)*

J.C.H. Spence et al., Acta Cryst. A ***61****, 237 (2005)*

D. Starodub et al. J. Chem Phys ***123****, 244304 (2005)*



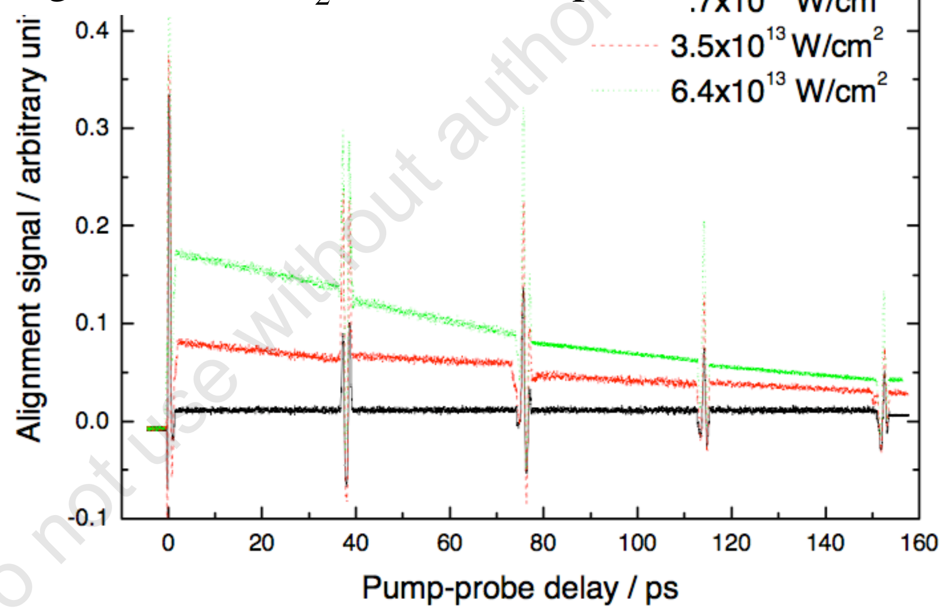
Larsen, J. Chem Phys ***111****, 7774 (1999).*

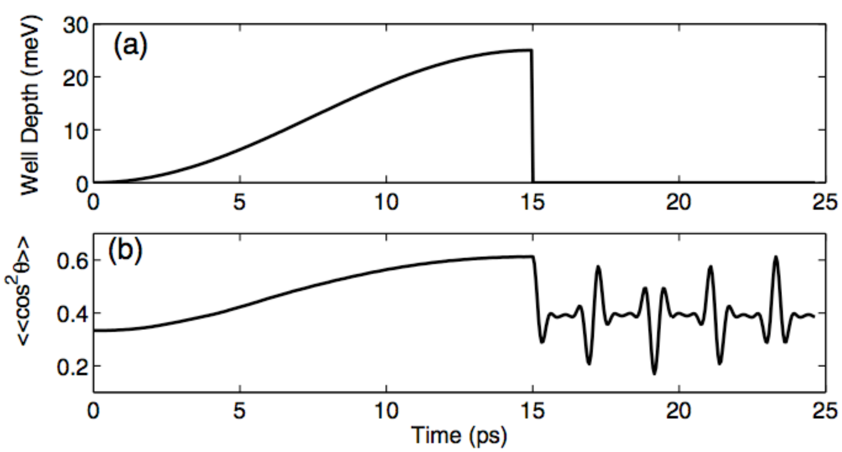
FEL pulses can probe laser alignment interactions

Laser fields can align particles but they may also distort them.

Methods exist for impulsive and adiabatic field-free alignment. An ultrafast FEL pulse can probe alignment

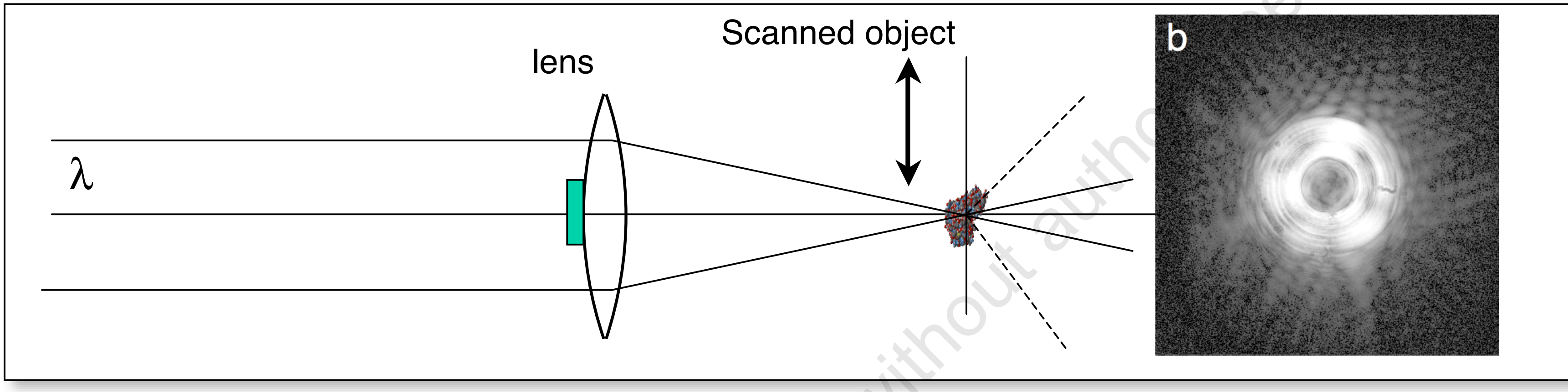
Xu, Optics Express 2006
Alignment of CS_2 with 100 fs pulse

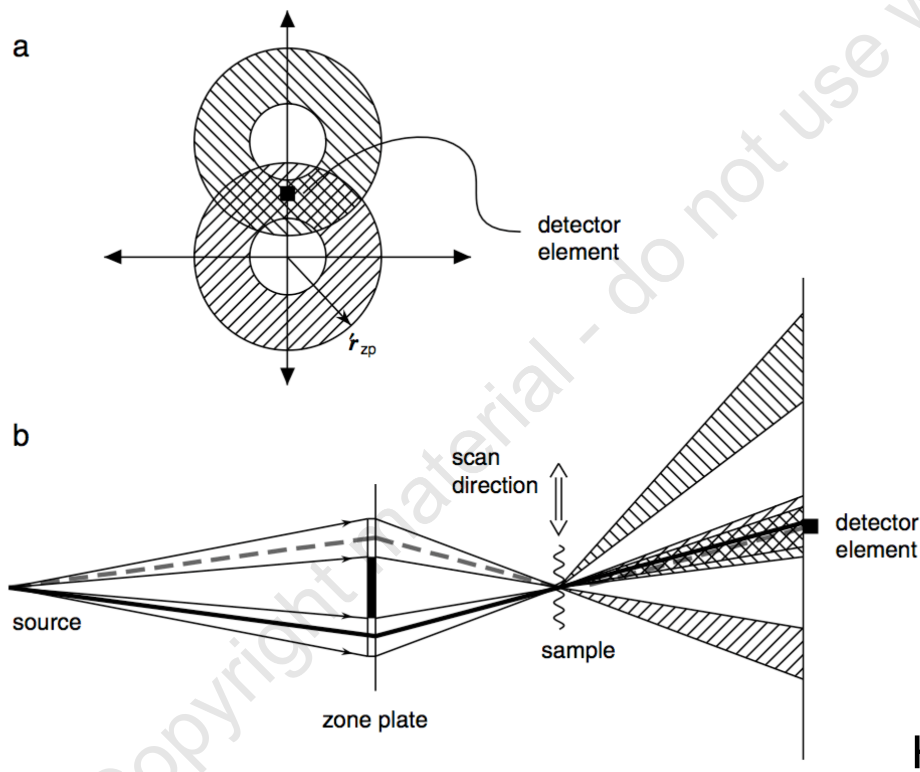


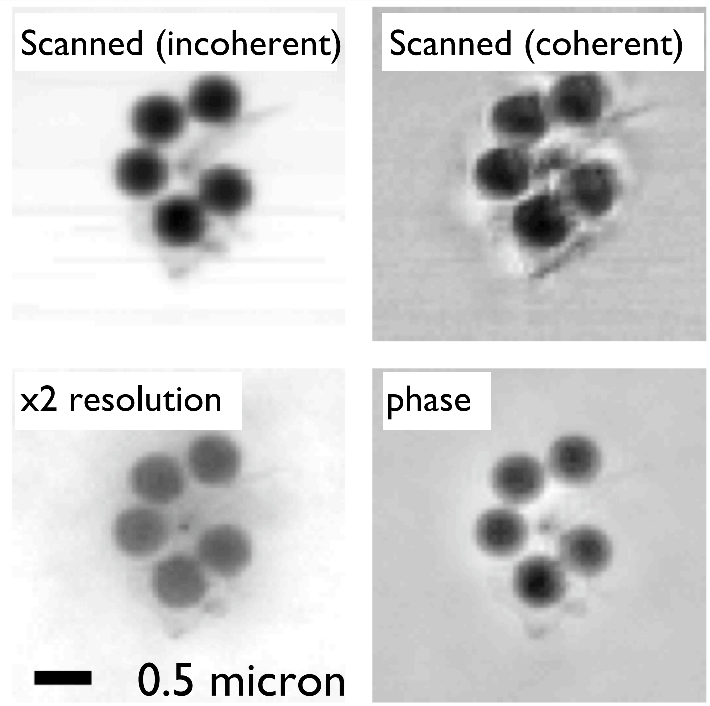


Underwood, PRL 2003
Alignment with 15 ps, fast switch-off

Coherent diffractive imaging can be combined with lenses to increase robustness







H. Chapman, Ultramicros. **66** 153 (1996)

Acknowledgements

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SLAC: Keith Hodgson, Sebastien Boutet

DESY: Thomas Tschentscher, Elke Plönjes, Marion Kulhman, Rolf Treusch, Stefan Dusterer

TU Berlin: Thomas Möller, Christof Bostedt

LBNL: Malcolm Howells, Congwu Cui

ASU: John Spence, Uwe Weierstall, Bruce Doak