The Abdus Salam International Centre for Theoretical Physics

United Nations Educational, Scientific and Cultural Organization

IAEA International Atomic Energy Agency

1936-41

Advanced School on Synchrotron and Free Electron Laser Sources and their Multidisciplinary Applications

7 - 25 April 2008

Coherent Diffractive Imaging with FELs

Henry Chapman

Centre for Free-Electron Laser Science - DESY

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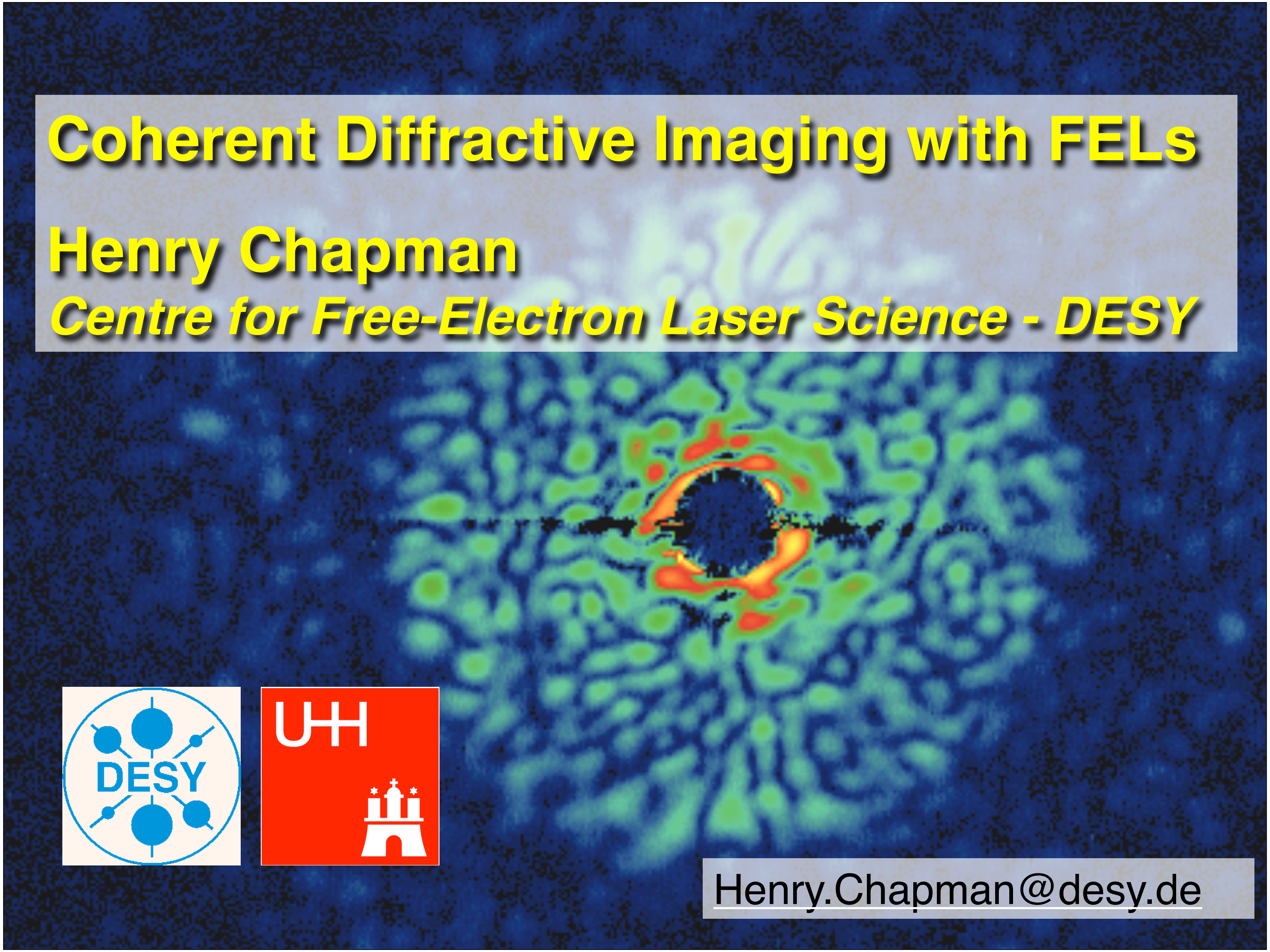
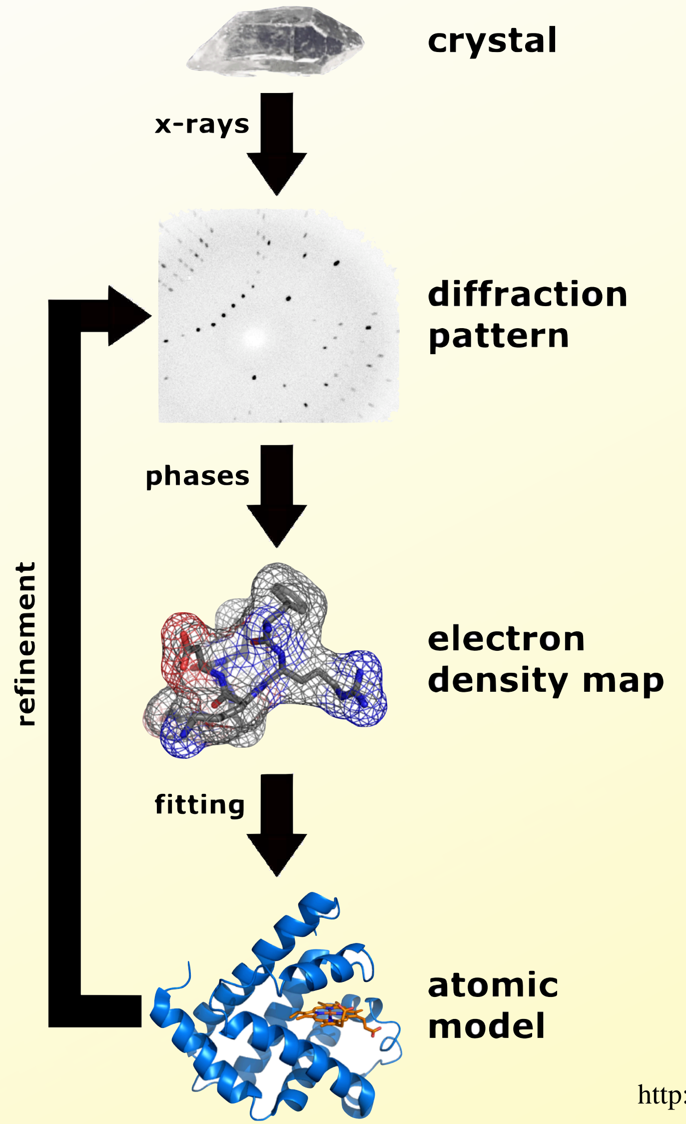


Henry.Chapman@desy.de

Crystallography overcomes the radiation damage and optics limitations, but requires crystals



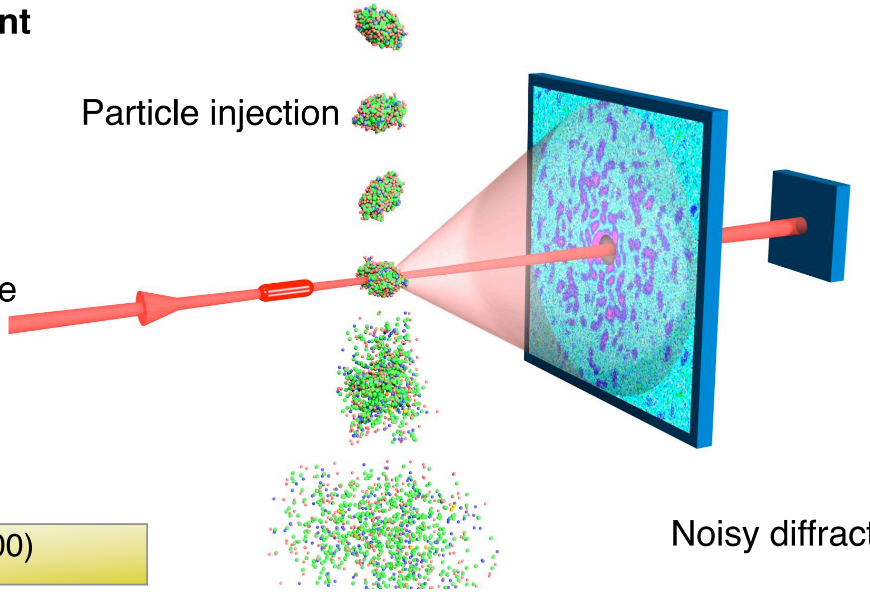
- ★ Radiation damage is spread out over 10^{10} identical unit cells
- ★ Diffraction from unit cells adds up coherently to form strong Bragg peaks
- ★ > 40,000 structures solved (in protein data bank), but ~10,000 distinct structures

- The bottleneck is in growing crystals

http://en.wikipedia.org/wiki/Image:X_ray_diffraction.png

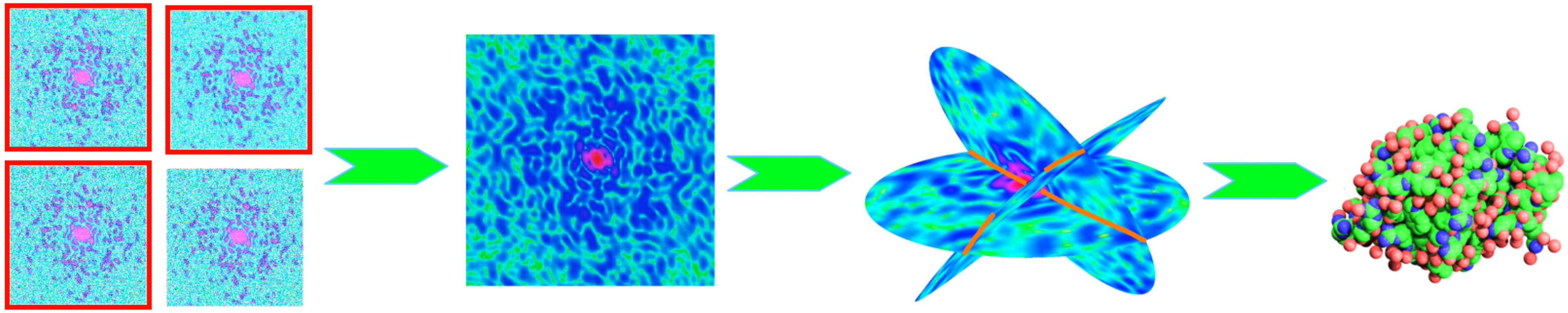
X-ray free-electron lasers may enable atomic-resolution imaging of biological macromolecules

One pulse, one measurement



R. Neutze et al, Nature **406** (2000)

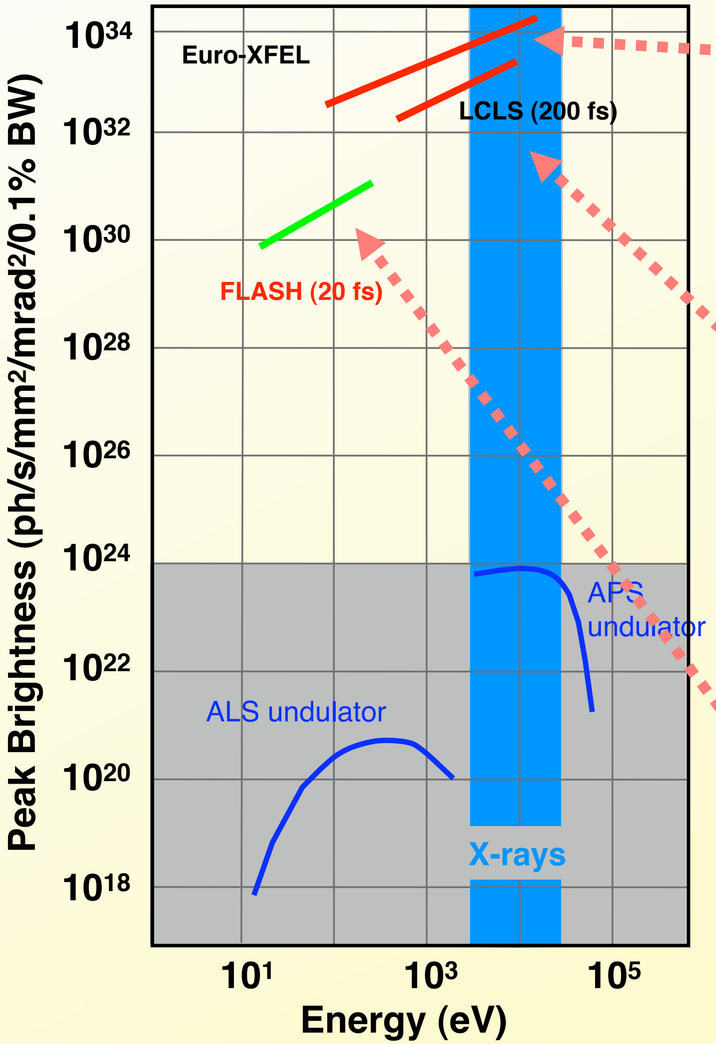
Combine 10^5-10^7 measurements



G. Huldt et al, J. Struct. Biol **144** (2003)

J. Miao, Hodgson, Sayre, PNAS **98** (2001)

X-ray free-electron lasers provide pulses that are *intense, short duration, short wavelength,* and *coherent*

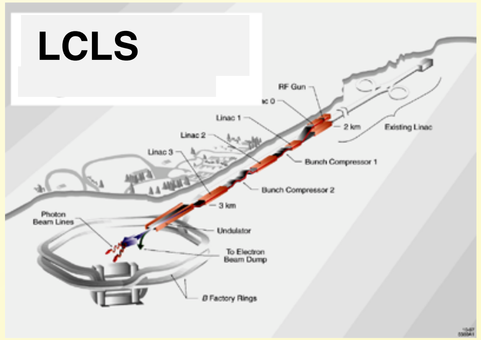


APS=Advanced Photon Source (ANL)
ALS=Advanced Light Source (LBNL)

operational 2013

12 keV, 50 fs, 10^{13} photons

European X-ray FEL,
DESY, Hamburg



operational 2009

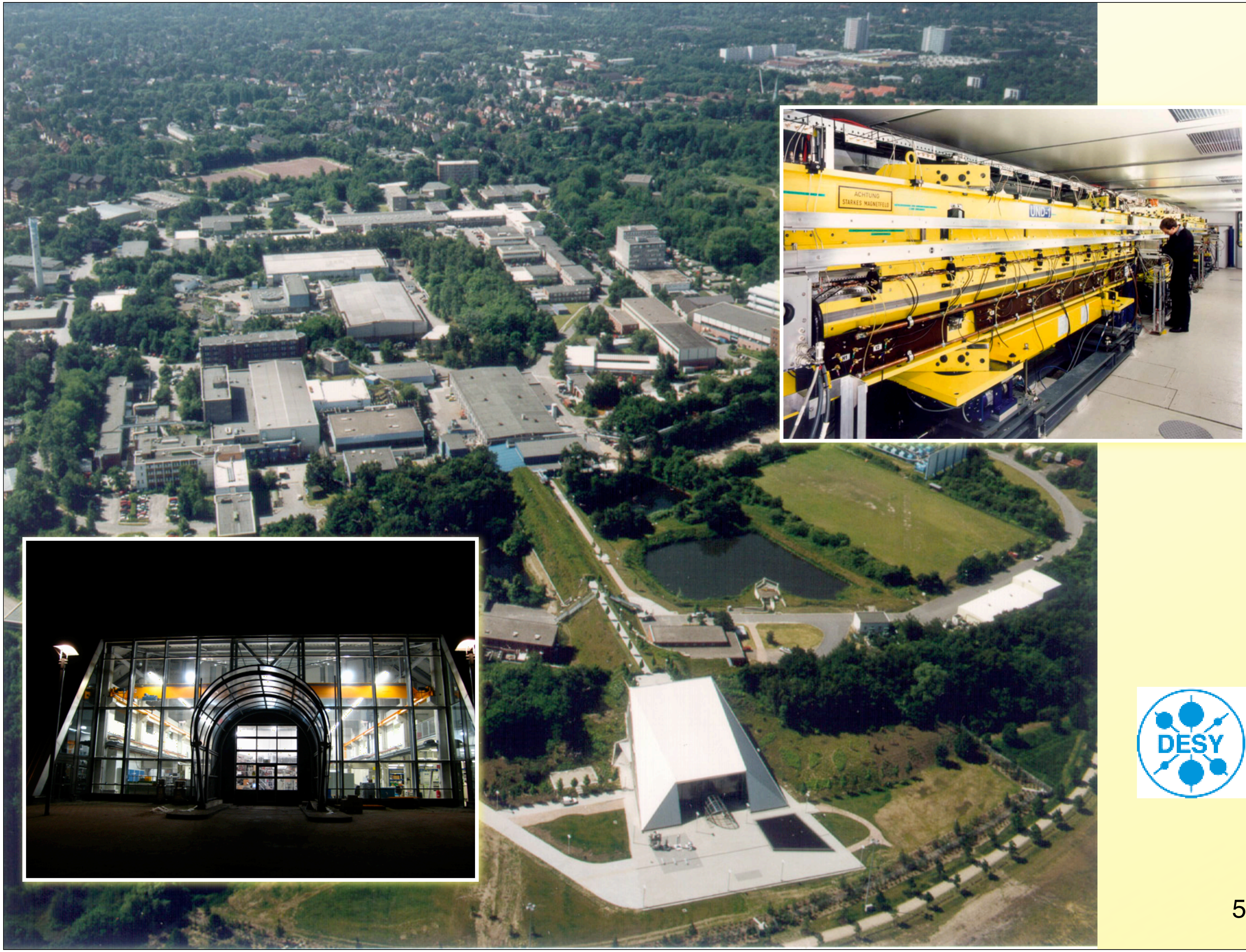
8 keV, 200 fs, 10^{12} photons

Linac Coherent Light Source,
SLAC, Stanford

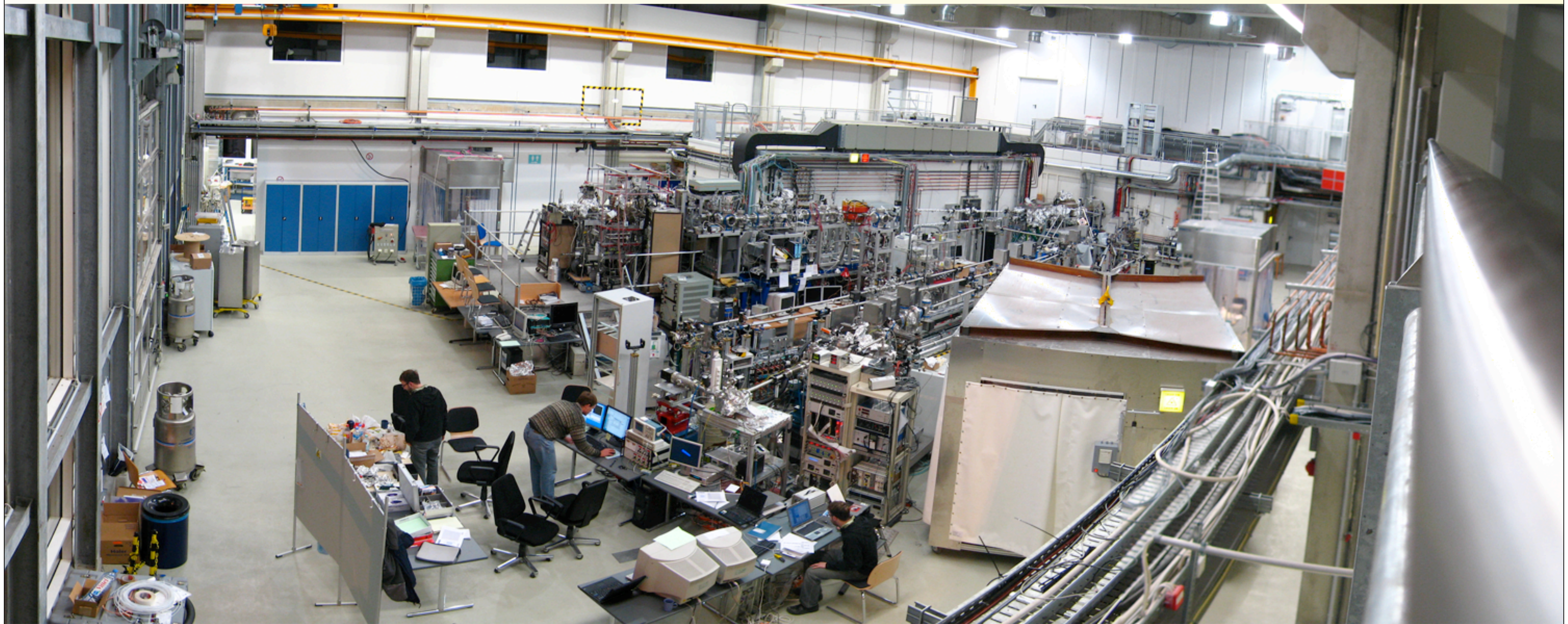
operational now

500 eV, 25 fs, 10^{12} photons

FLASH
DESY, Hamburg


ACHTUNG
STARKES MAGNETFELD
UND-1

We have carried out experiments at FLASH

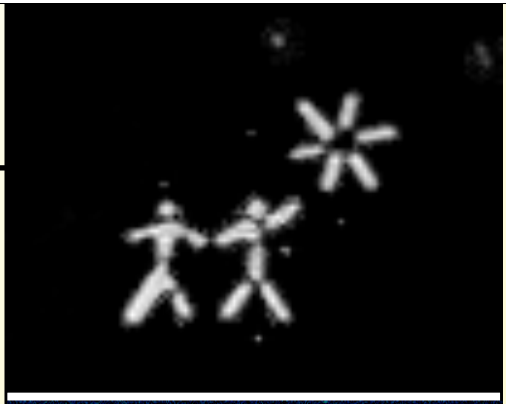


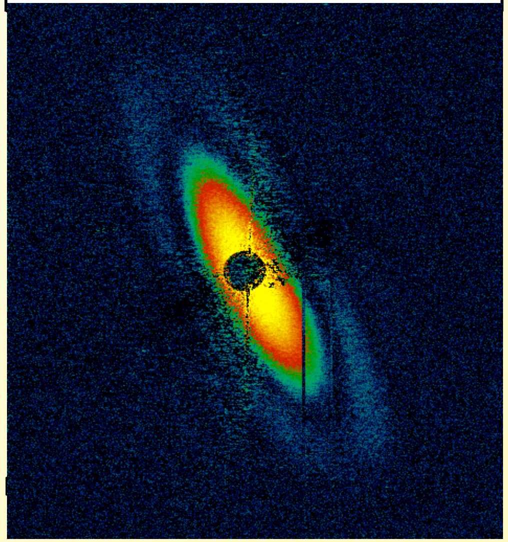
FLASH parameters for the experiments:
32 nm, 13 nm, 7 nm wavelengths
10-30 fs pulses
up to 10^{13} photons/pulse
Single pulses (5 Hz) & pulse trains (140 pulses spaced by 10 µs; 700 Hz)

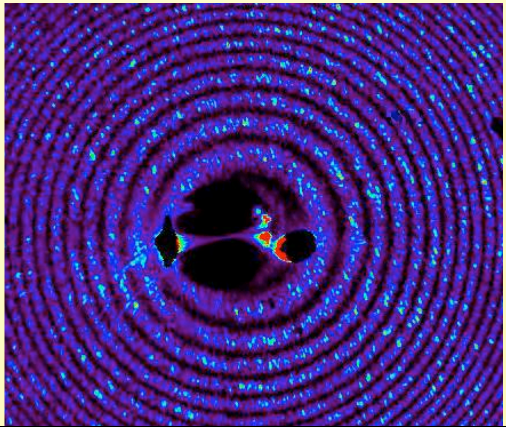
We have overcome many challenges

Using the FLASH free-electron laser we have:

- Demonstrated single-pulse coherent diffractive X-ray imaging;
- Demonstrated single-pulse diffractive imaging of particles injected into vacuum from solution, traveling across the beam at ~100 m/s.
- Examined the dynamics of objects irradiated by FEL and laser pulses. These studies support models that suggest that pulse durations of up to 50 fs could be used to attain 0.3 nm resolution;
- Developed time-resolved imaging of laser ablation

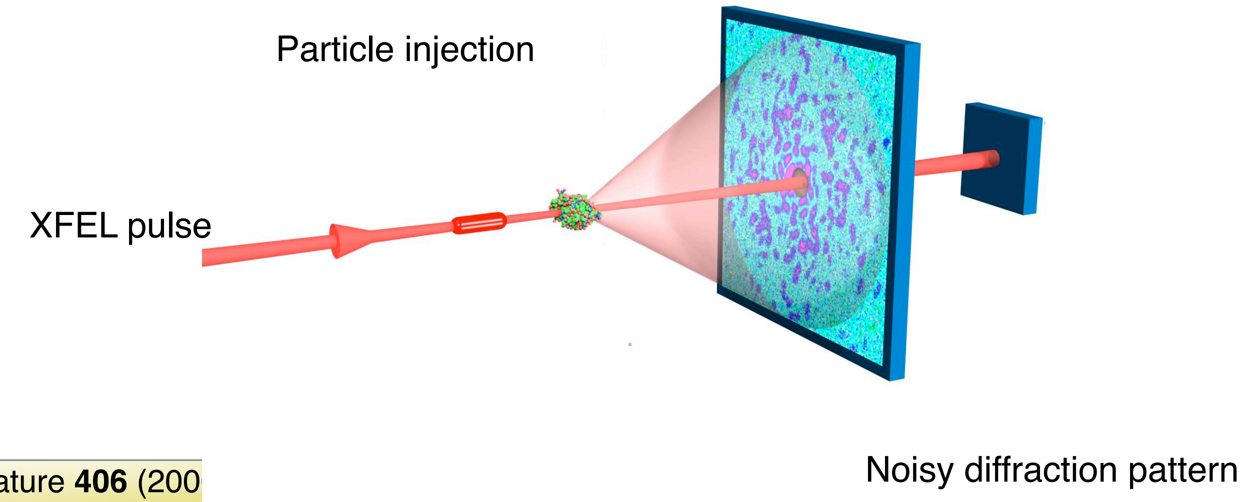






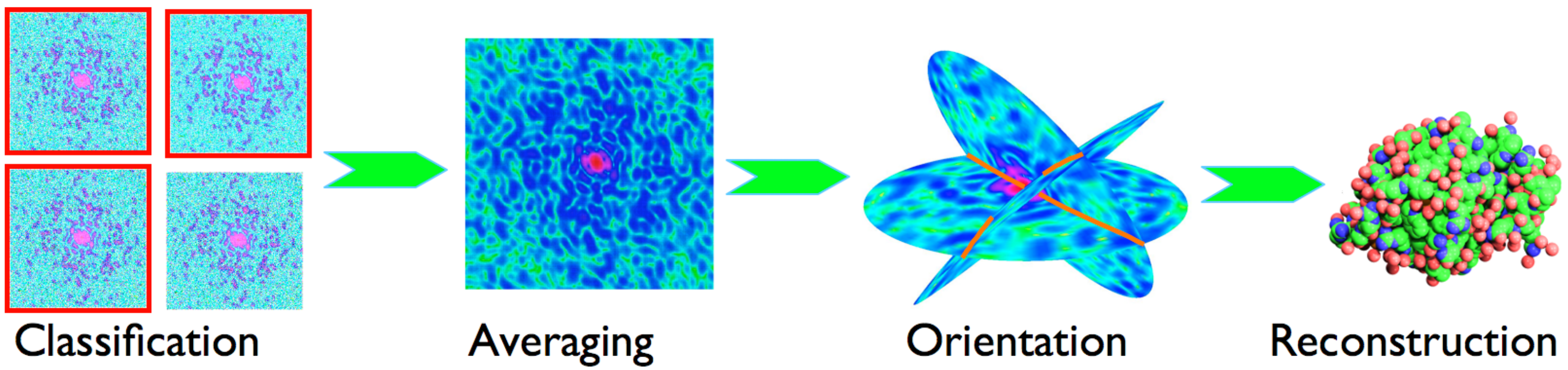
X-ray free-electron lasers may enable atomic-resolution imaging of biological macromolecules

One pulse, one measuremer



R. Neutze et al, Nature **406** (200

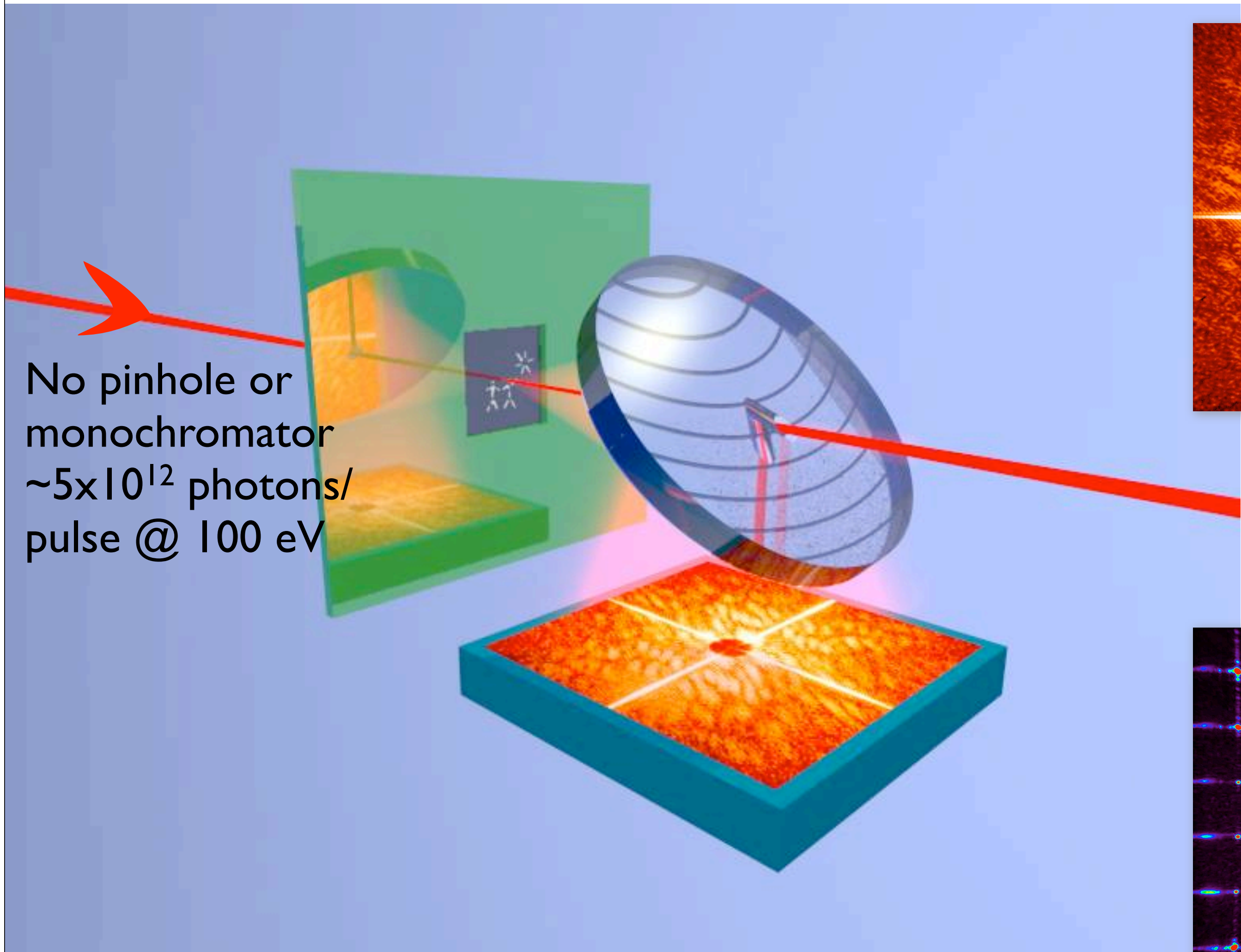
Combine 10^5-10^7 measurements

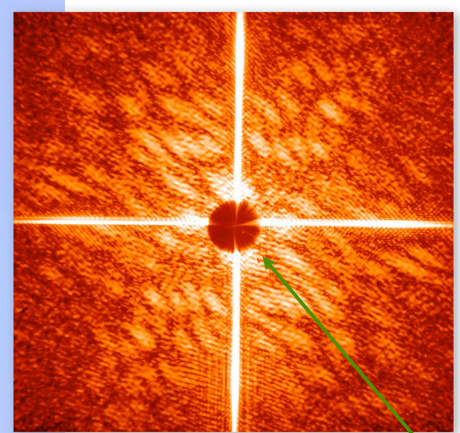


G. Huldt et al, J. Struct. Biol **144** (2003)

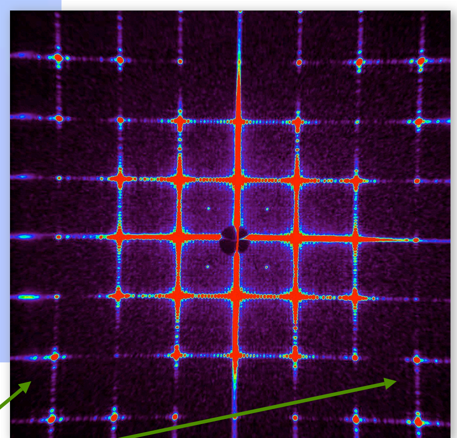
J. Miao, Hodgson, Sayre, PNAS **98** (2001)

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

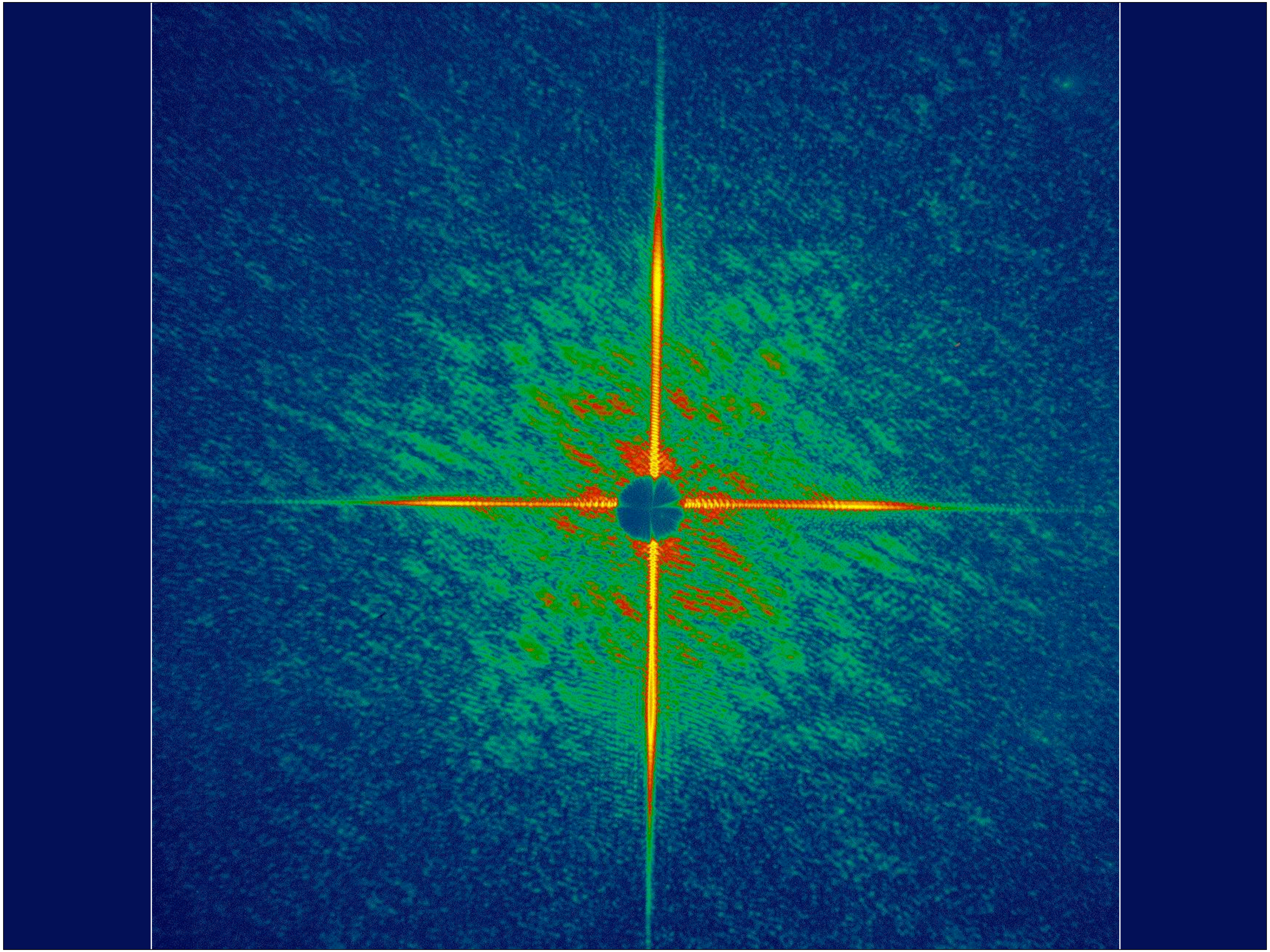


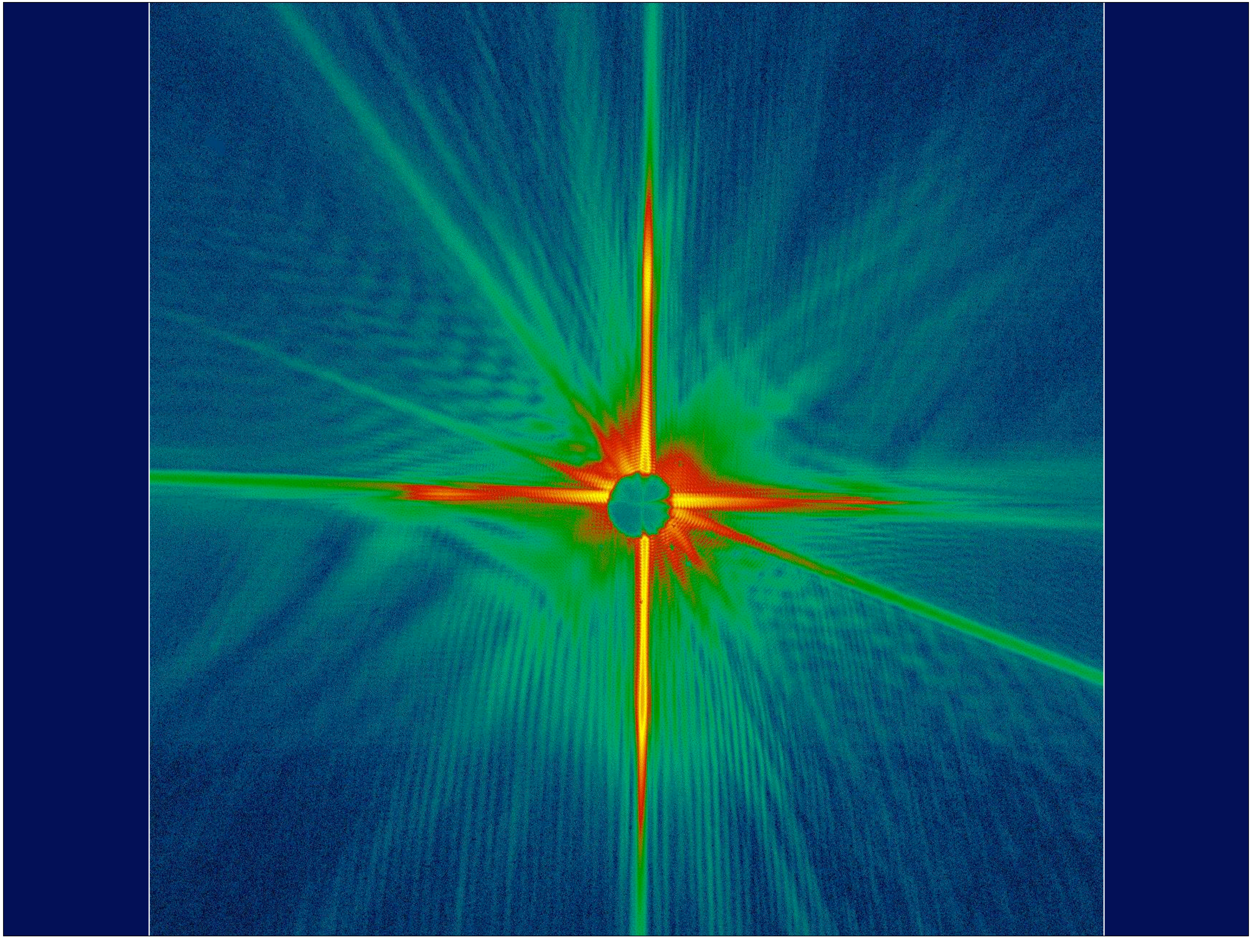


"Soft edge" prevents any scatter from the hole

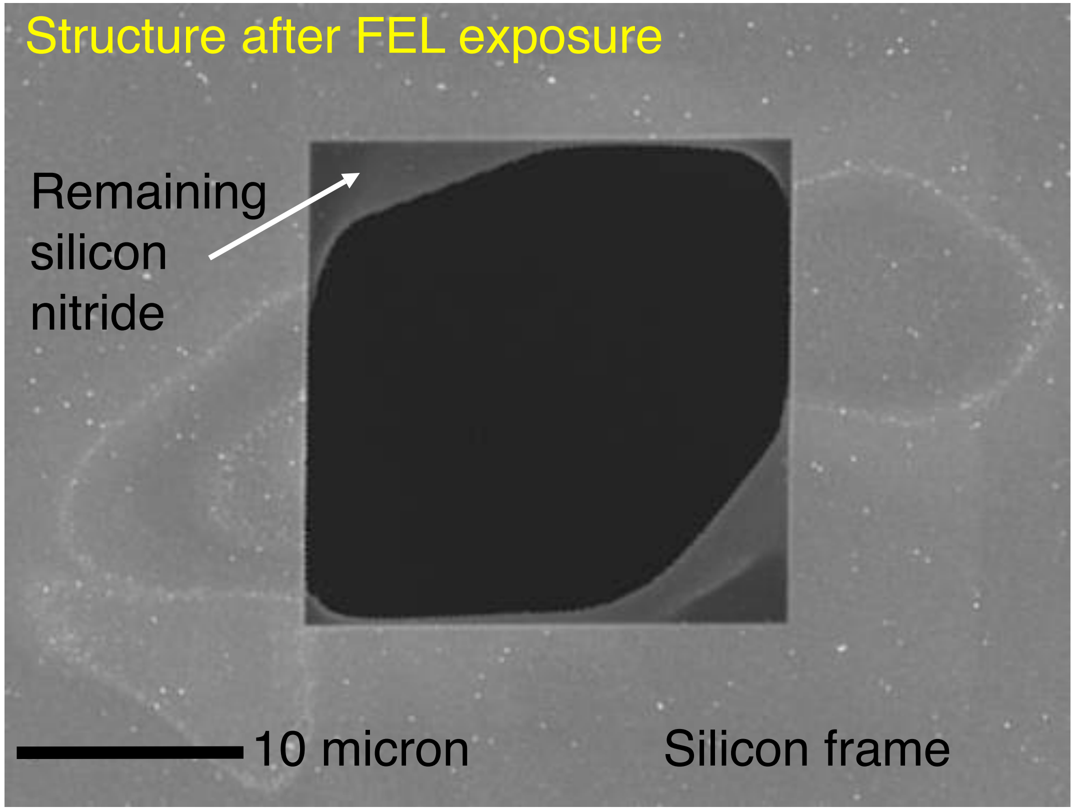


Multilayer reflectivity is uniform across the 30° to 60° gradient

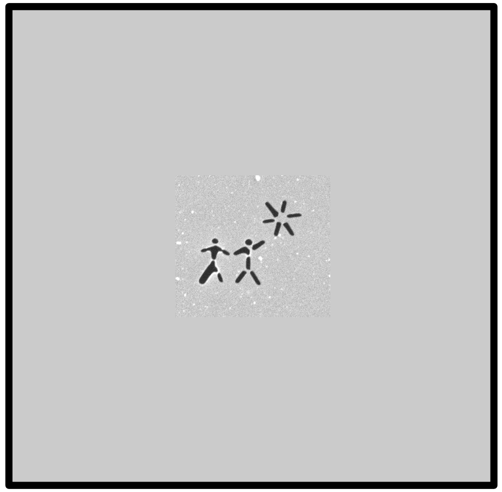




The sample was heated to 60,000 K by the FEL pulse



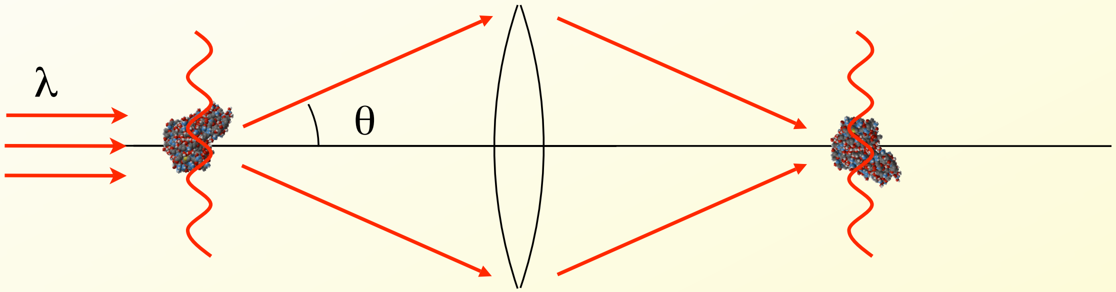
before FEL exposure



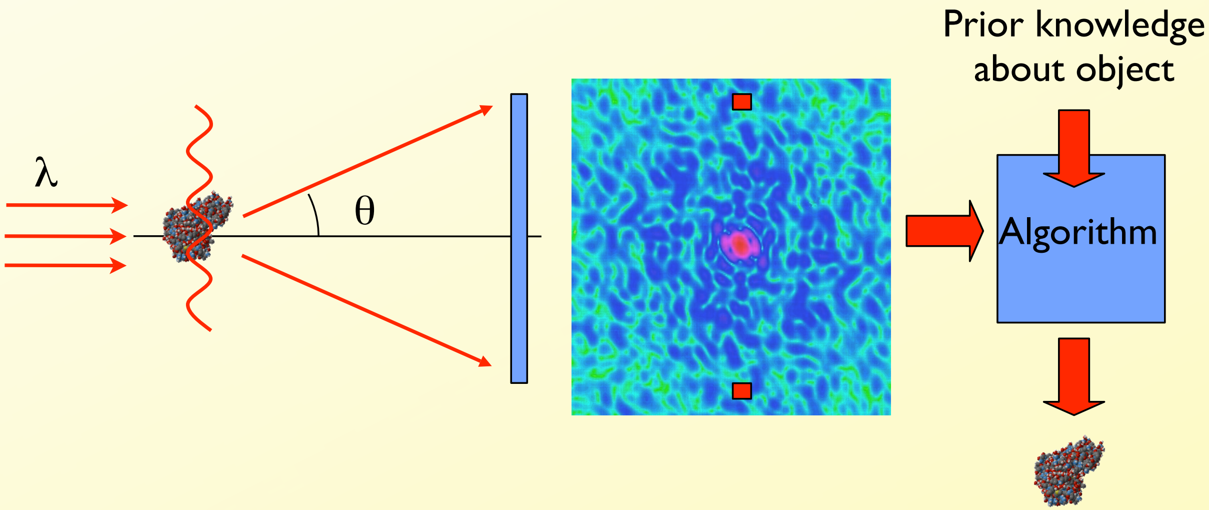
Pulse energy: 10 μJ or 1 J/cm^2
Dose in Si_3N_4: 10^5 J/g or 22 eV/atom
Temperature of 6×10^4 K, or 5.2 eV, ionization/atom of 2.5
Surfaces will expand at sound speed: 1.3×10^6 cm/s
In 25 fs, material will not move more than 3 Å

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



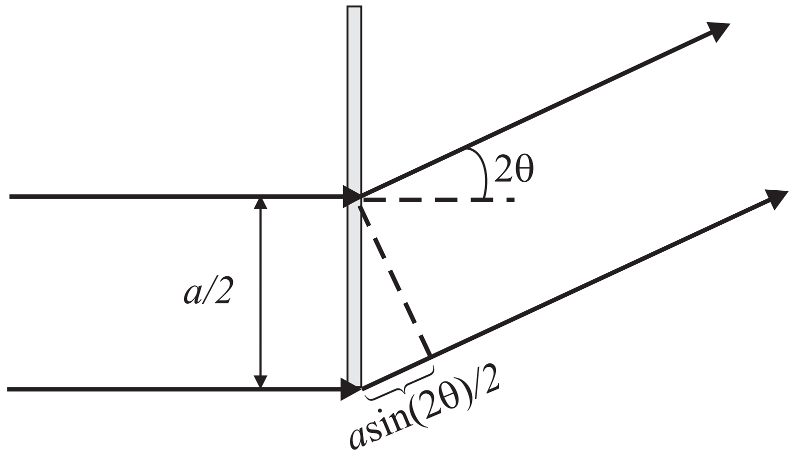
An algorithm finds the phases that are consistent with measurements and prior knowledge

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

J. Fienup, Appl. Opt. **21** 2758 (1987)

J. Miao et al, Nature **400** 342 (1999)

Lensless imaging has strict coherence requirements



$$\frac{\lambda^2}{\Delta\lambda} > a \sin 2\theta .$$

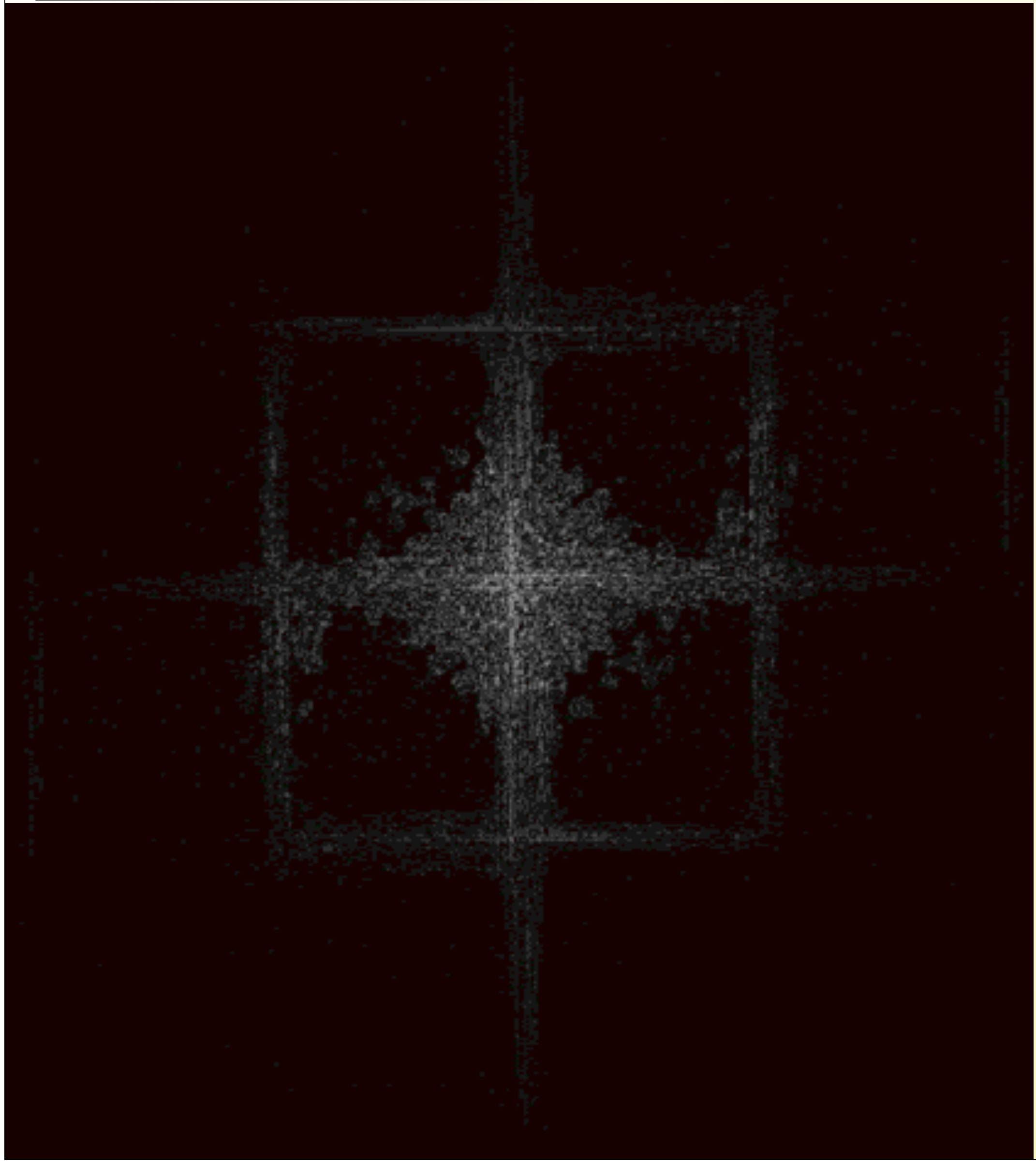
$$\frac{\Delta\lambda}{\lambda} < \frac{s}{a} . \quad = N$$

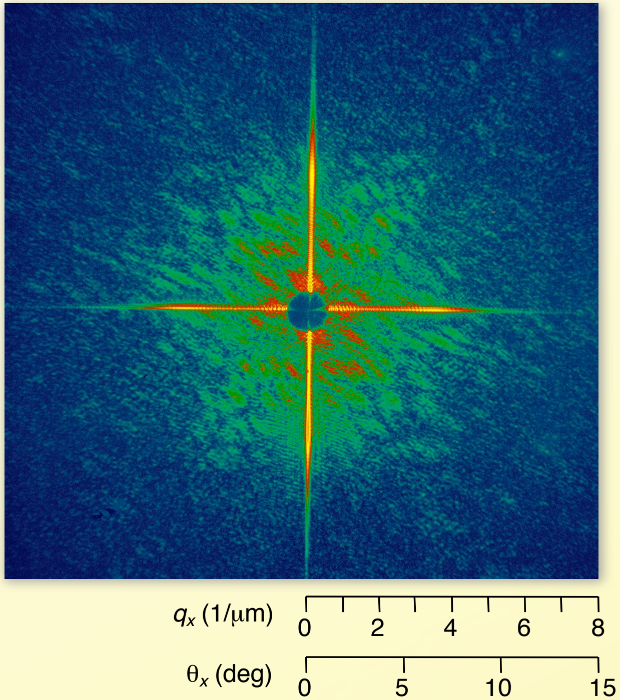
For high spatial resolution, the object size cannot be much larger than the pulse duration

This is an advantage if you have many identical copies of a sample in the beam

van der Veen and Pfeiffer, J. Phys **16**, 5003 (2004)

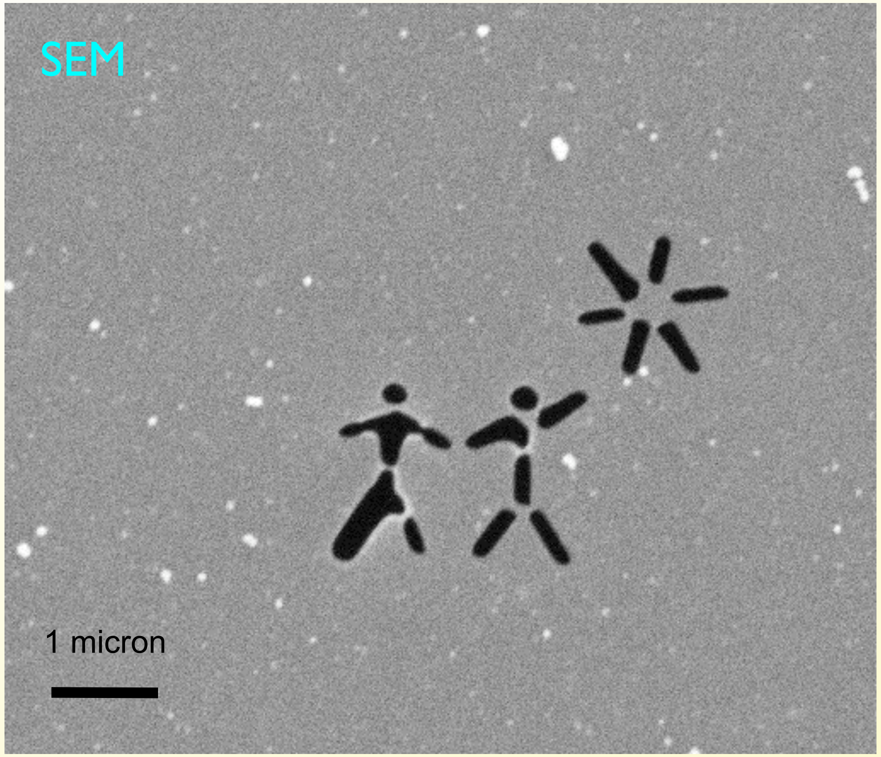
We perform ab initio image reconstruction with our "Shrinkwrap" algorithm

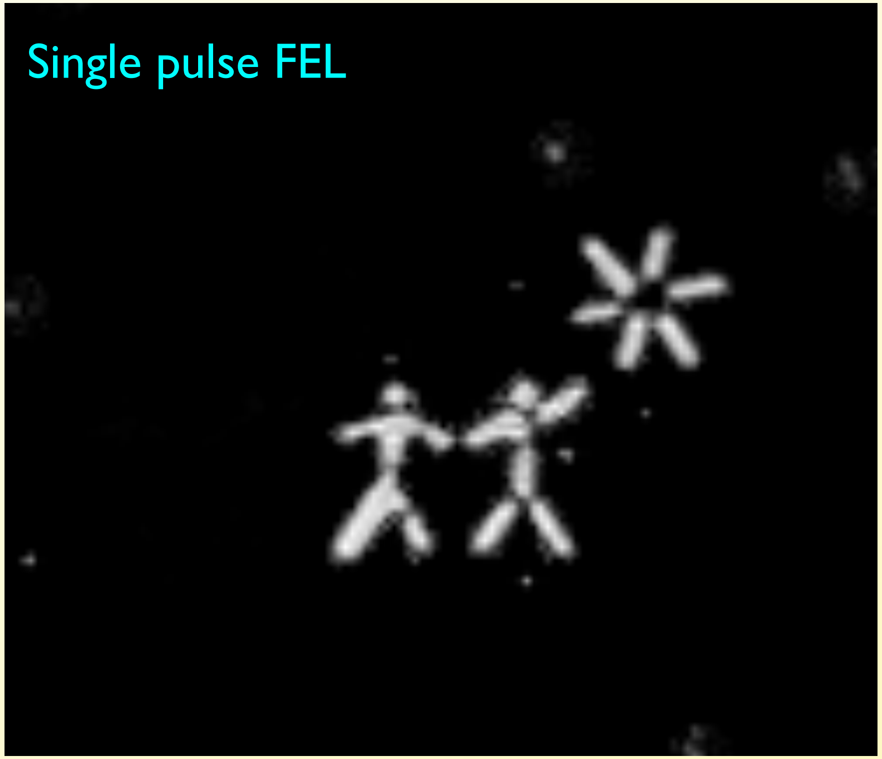




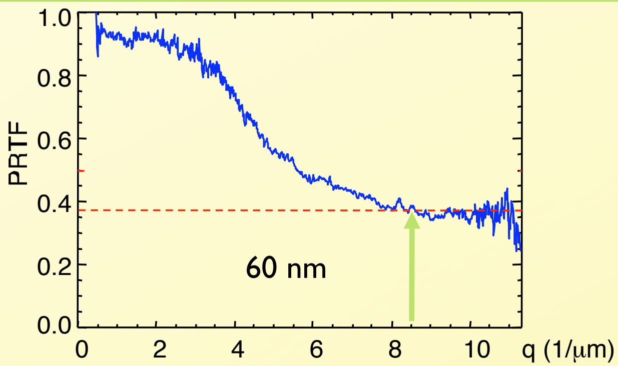
S. Marchesini et al. Phys Rev B **68** 140101 (2003)

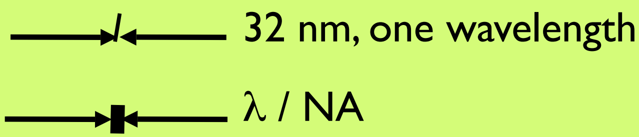
The reconstruction is carried out to the diffraction limit of the 0.26 NA detector





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





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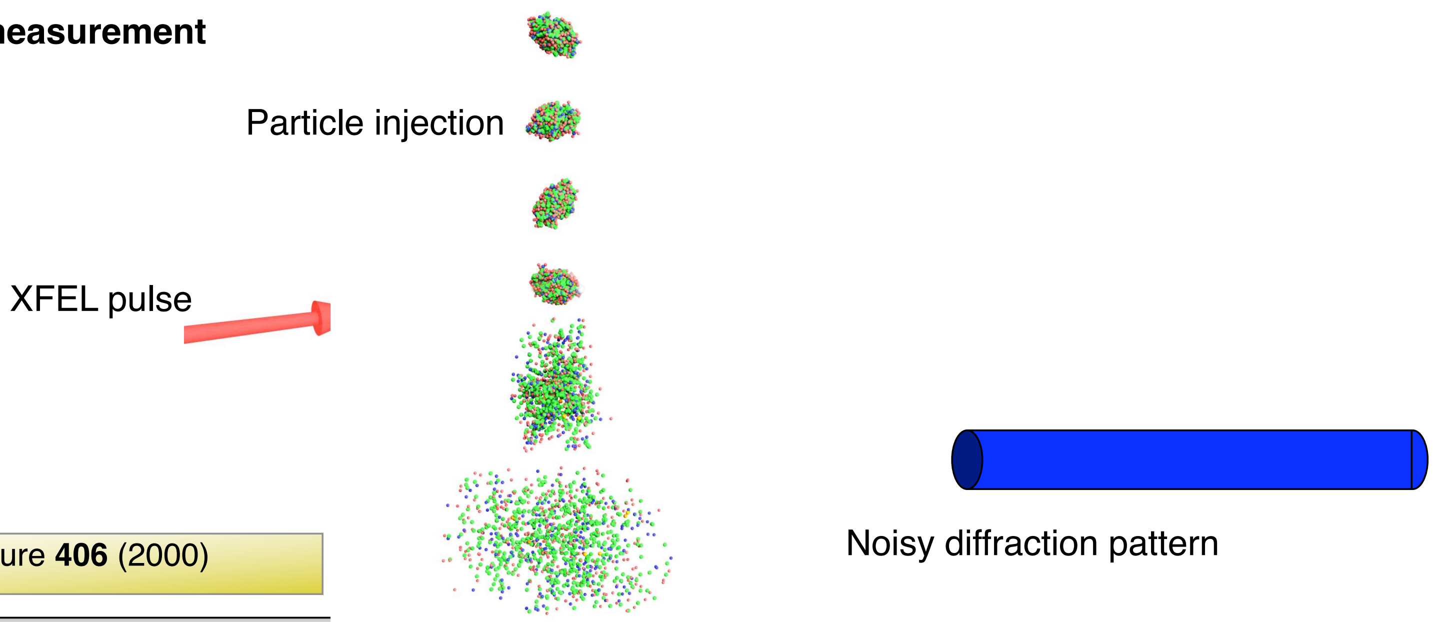

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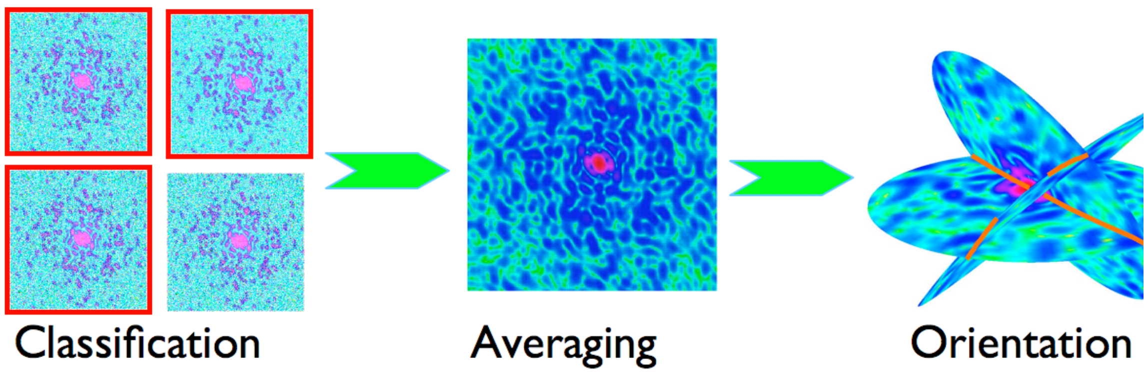


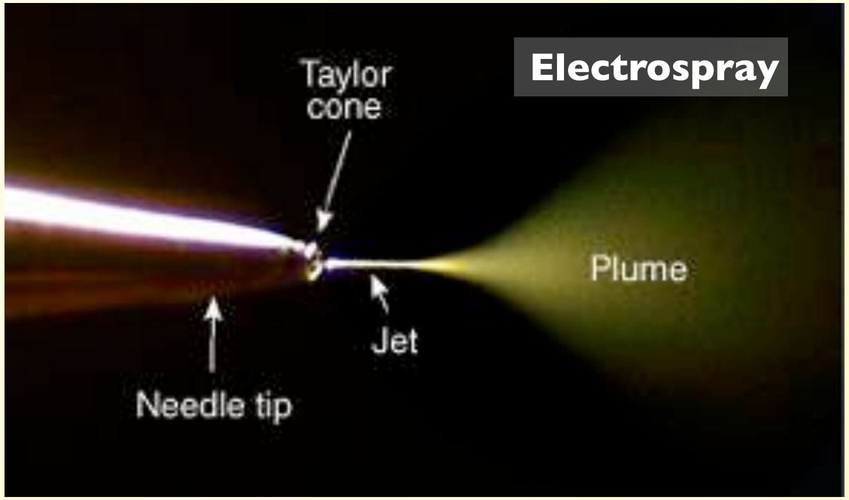
G. Huldt et al, J. Struct. Biol **144** (2003)

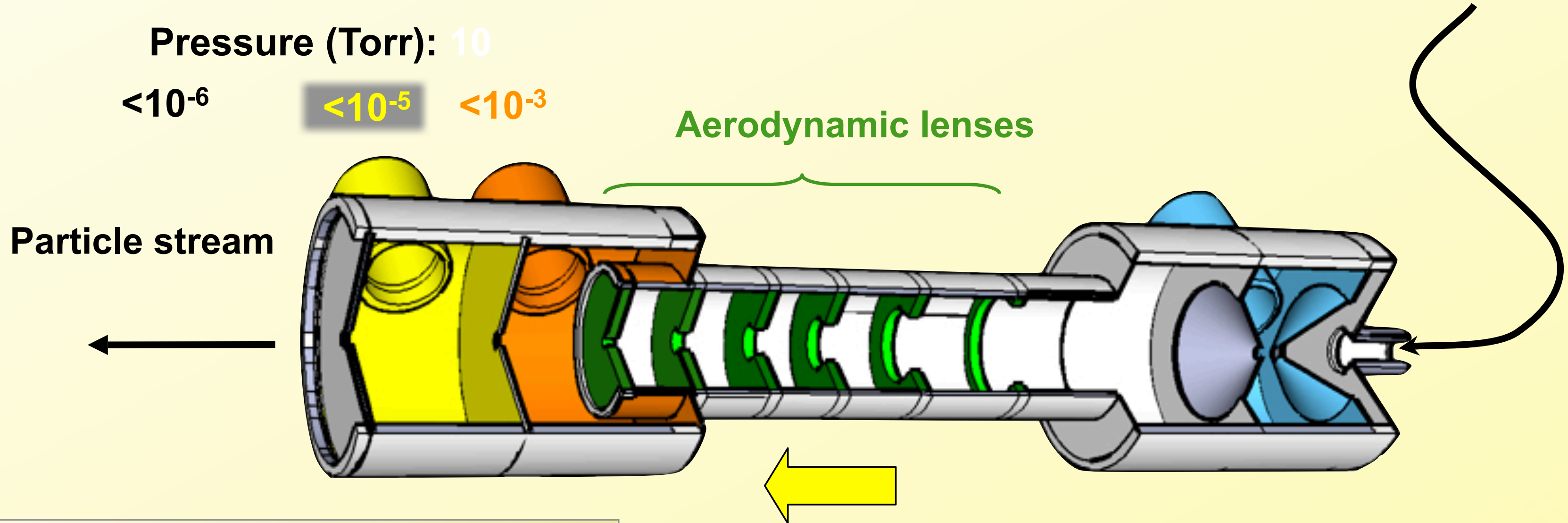
J. Miao, Hodgs

We inject particles into the FEL beam by aerodynamically focusing an aerosol

Challenges:

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





M.J. Bogan et al., Nano Letters **8**, 310 (2008)

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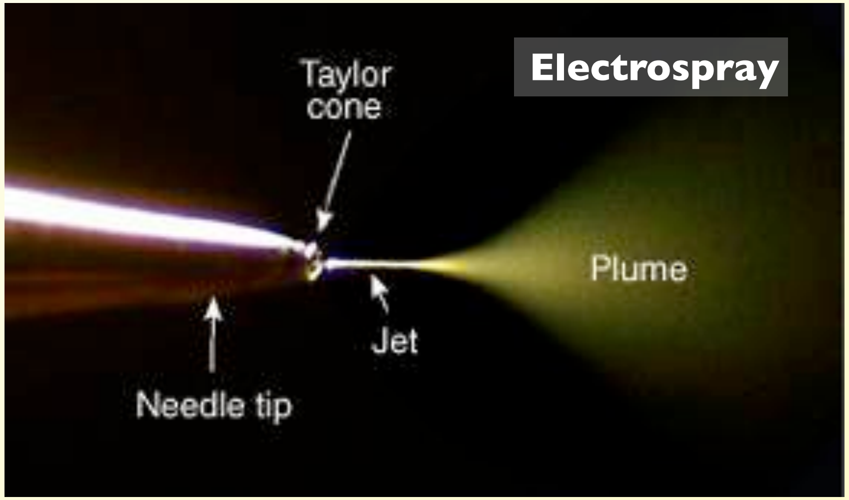
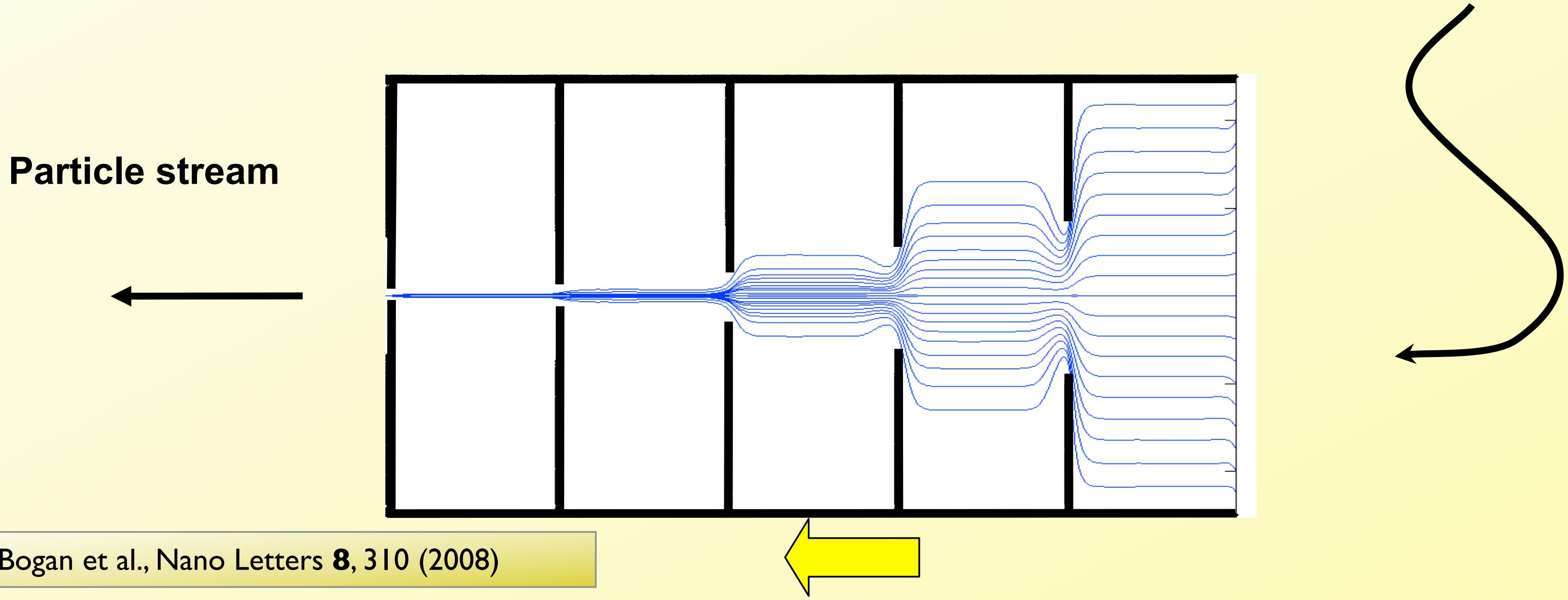
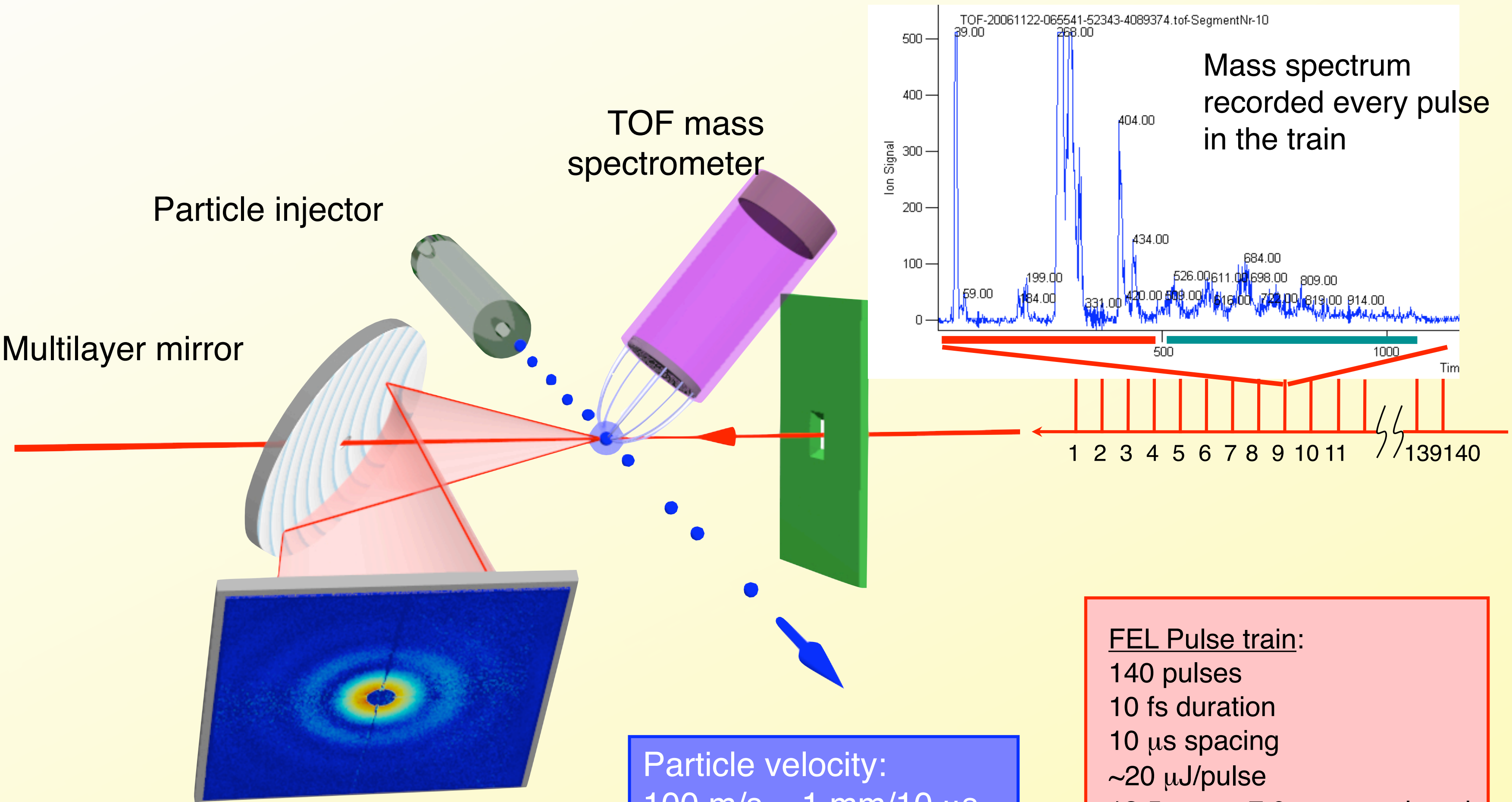




M.J. Bogan et al., Nano Letters **8**, 310 (2008)

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



Single particle diffraction pattern (one hit + 699 no-hits)

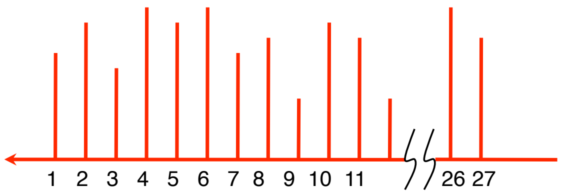
Particle velocity:
100 m/s = 1 mm/10 μs
(c.f. 20 μm beam)

<u>FEL Pulse train</u>:
140 pulses
10 fs duration
10 μs spacing
~20 μJ/pulse
13.5 nm or 7.0 nm wavelength

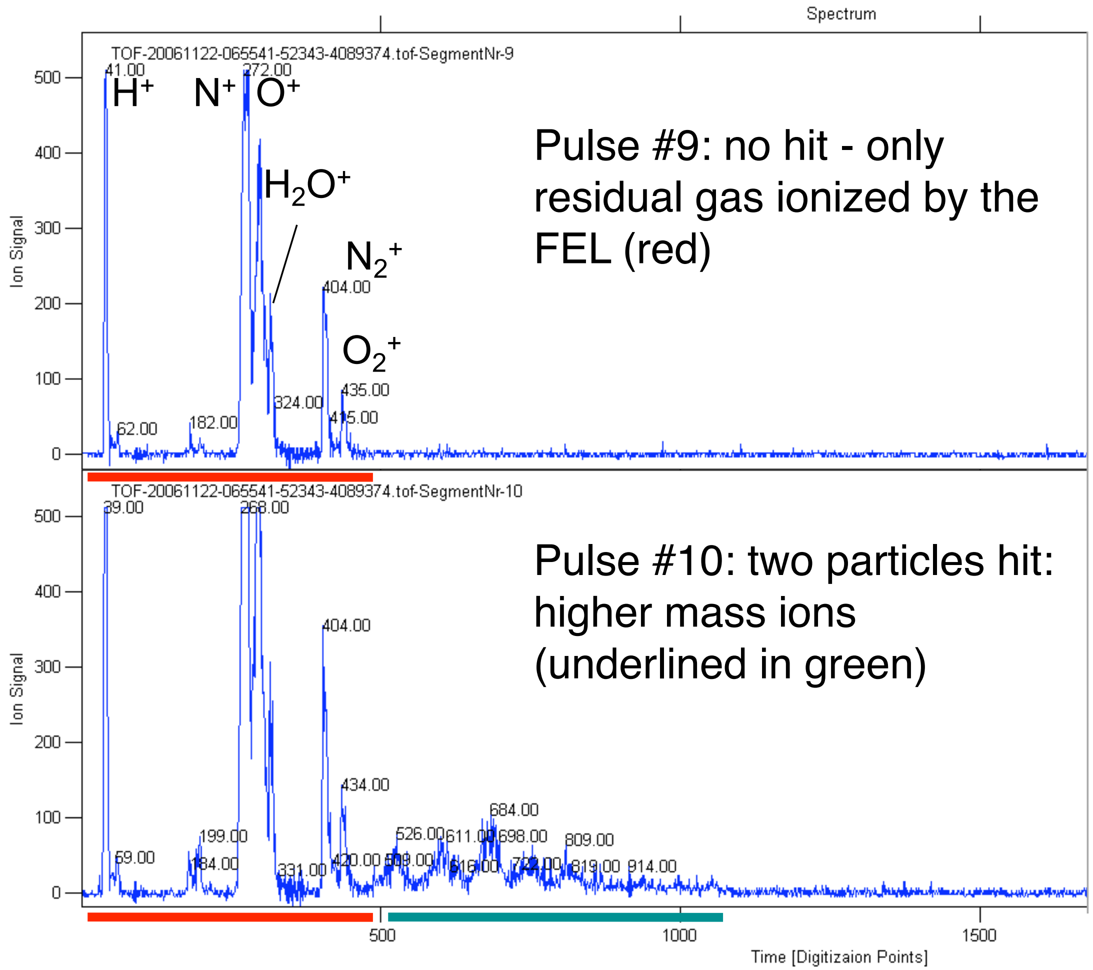
M.J. Bogan et al., Nano Letters **8**, 310 (2008)

A mass spectrum is recorded every FEL pulse

10 fs FEL pulses at 10 μs spacing



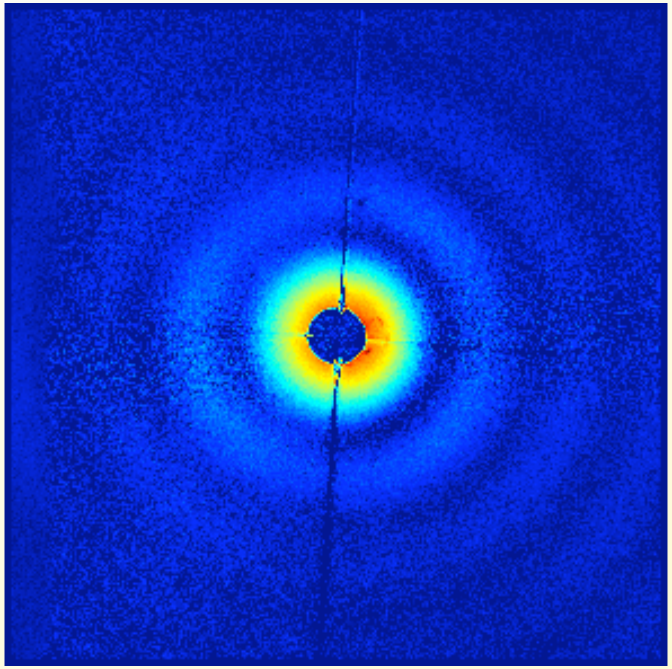
FEL Pulse train:
27 pulses
10 fs duration
10 μs spacing
5 Hz
~7 μJ/pulse
13.5 nm wavelength



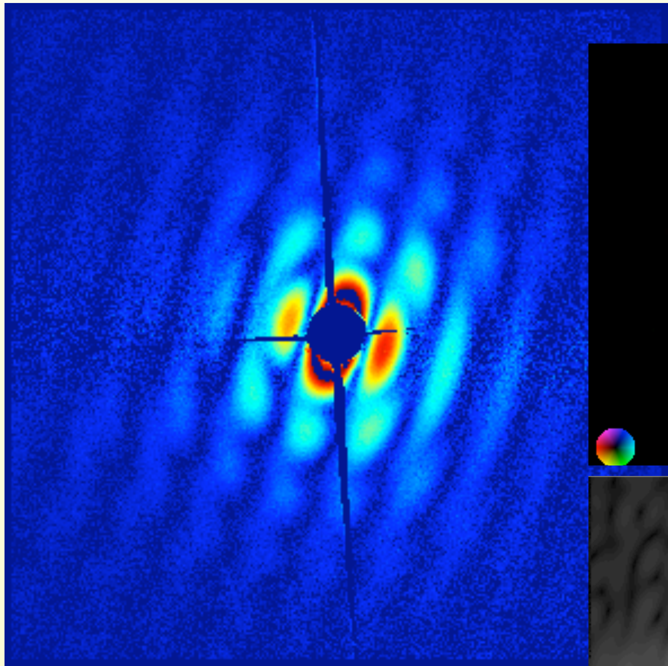
The mass spectra show which pulse in the pulse train had hit and how.

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

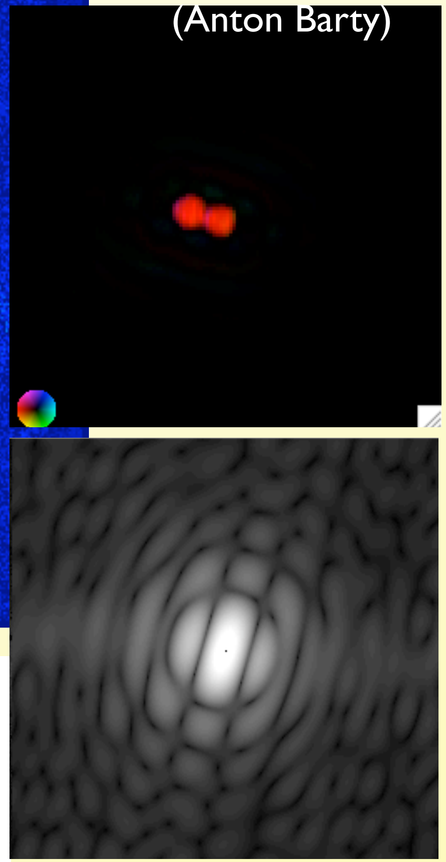
Single ~200 nm particle

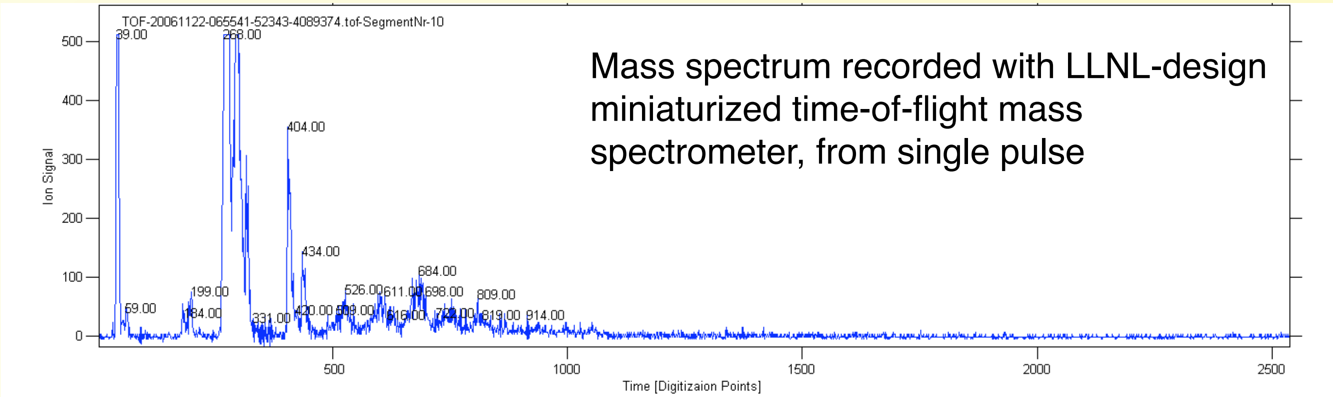


Two particles hit by the one pulse



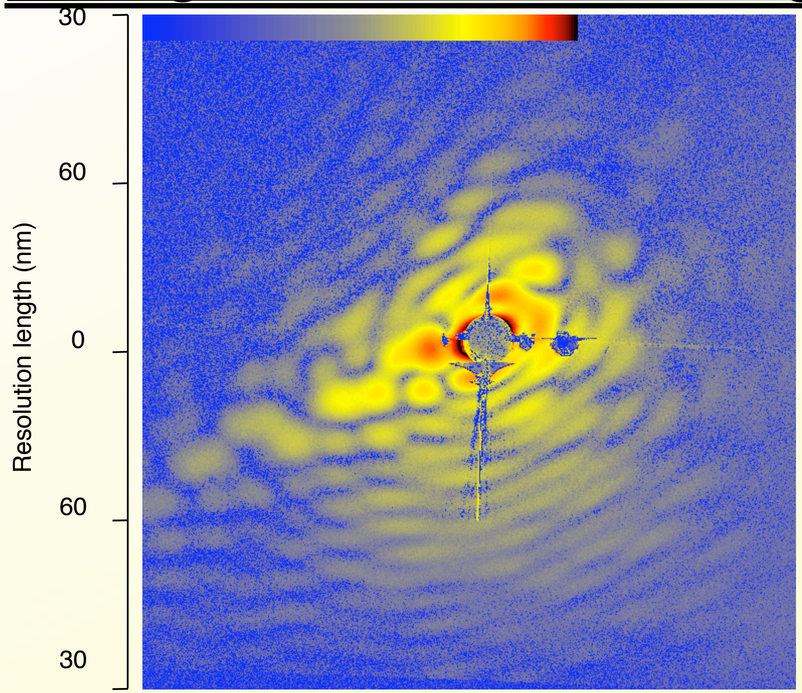
Reconstructed image (Anton Barty)

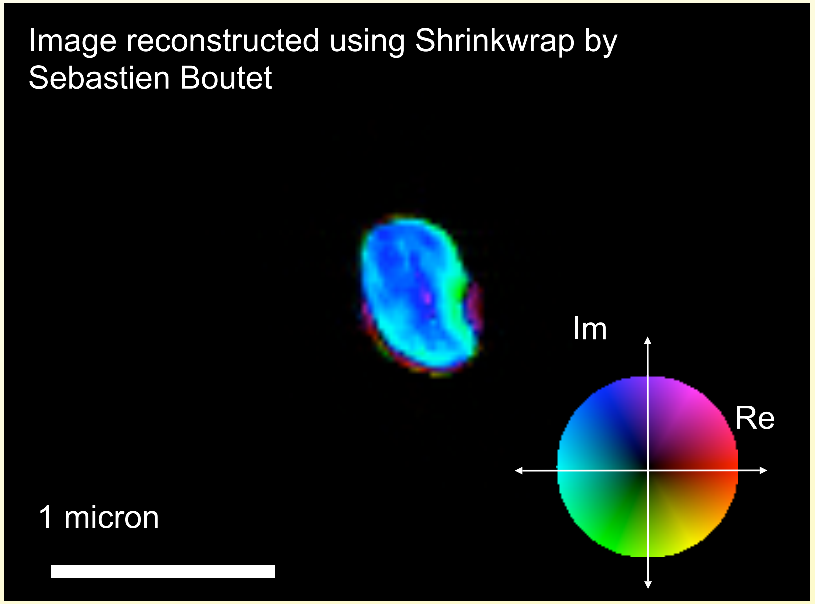




Mass spectrum recorded with LLNL-design miniaturized time-of-flight mass spectrometer, from single pulse

We have performed the first X-ray imaging of free-falling unstained live biological cells





Single shot ~10 fs diffraction pattern recorded at FLASH (DESY) at a wavelength of 13.5 nm of a picoplankton organism.

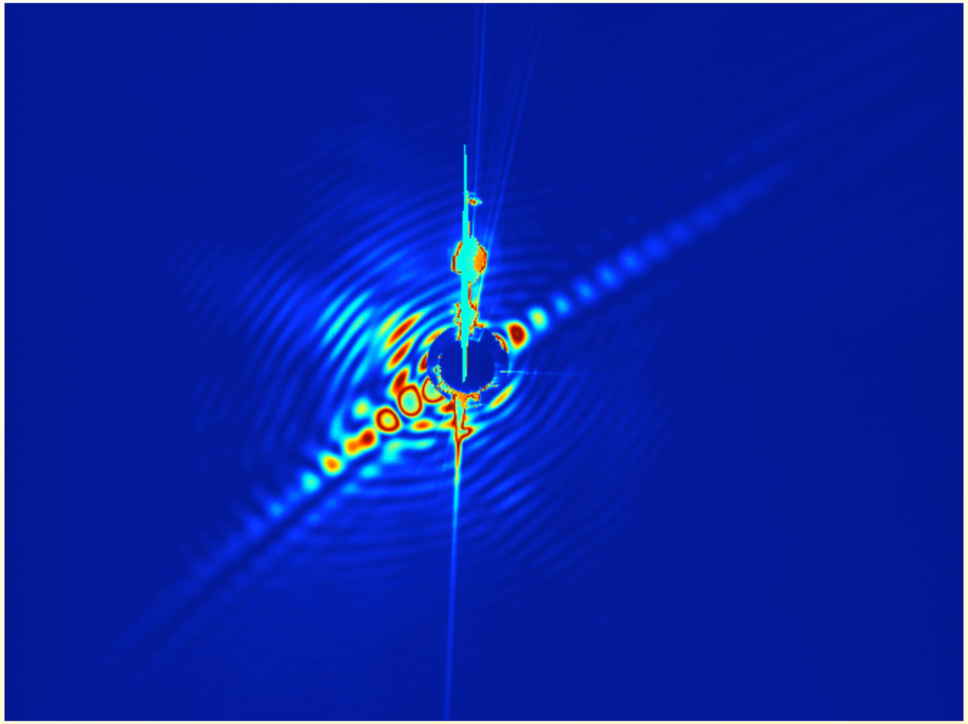
This cell was injected into vacuum from solution, and shot through the beam at 100 m/s

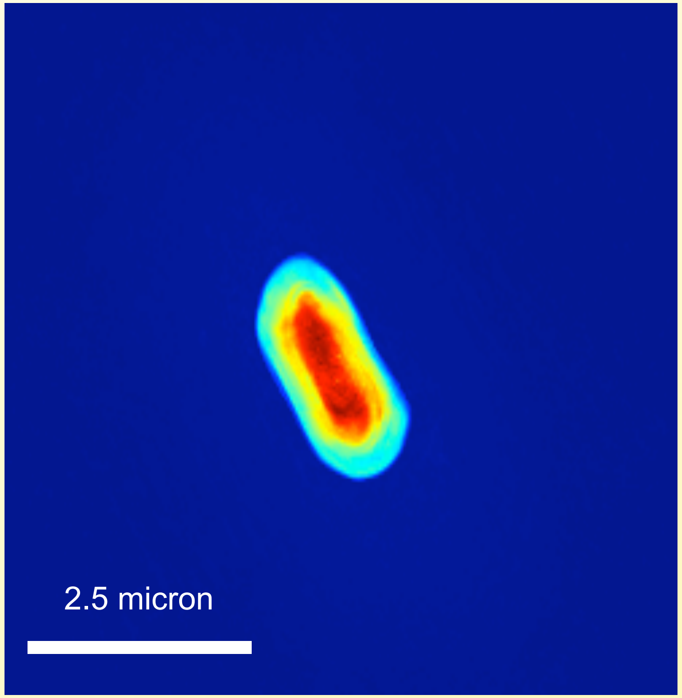
Ostreococcus TEM section
(Wenche Eikrem and Jahn Throndsen, University of Oslo)

J. Hajdu, I. Andersson, M. Svenda, M. Seibert (Uppsala)
S. Boutet (SLAC)
M. Bogan, H. Benner, U. Rohner, H. Chapman (LLNL)

Live picoplankton have been imaged ‘on the fly’ when injected into the FEL beam

March 2007, FLASH, Hamburg
FLASH pulse length: 10 fs
Wavelength: 13.5 nm



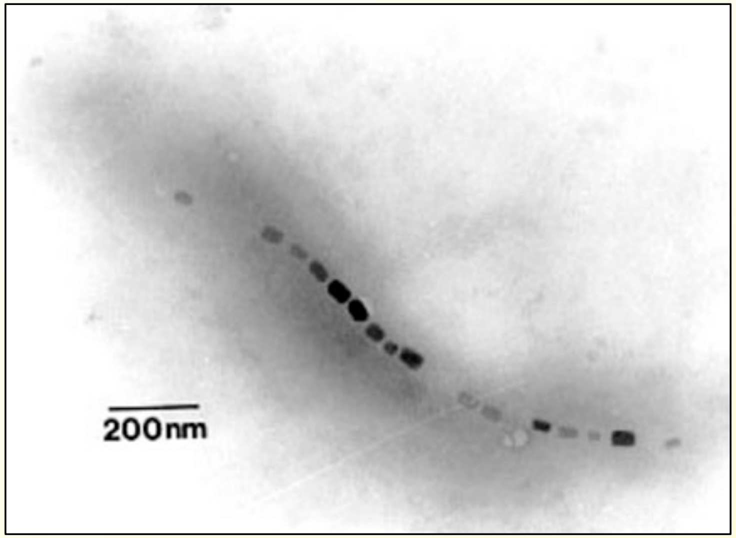


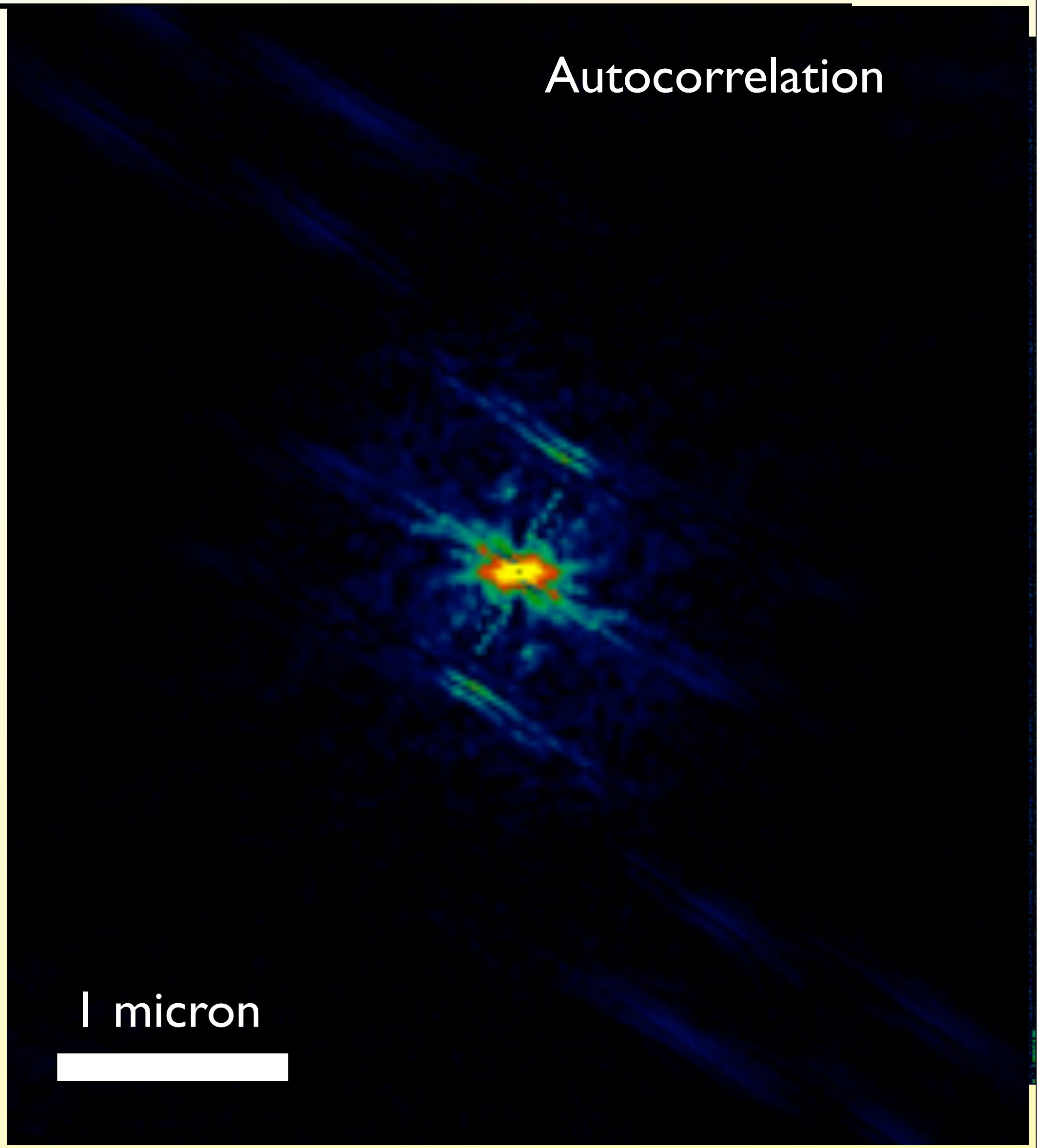
This cell was injected into vacuum from solution, and shot through the beam at 200 m/s

Reconstruction by Filipe Maia, Uppsala

UPPSALA UNIVERSITET

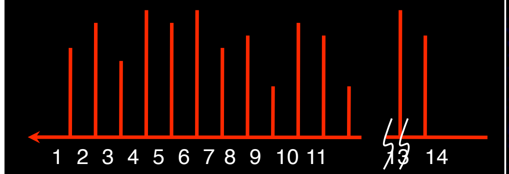
Magnetosomes at 7 nm (on a substrate)



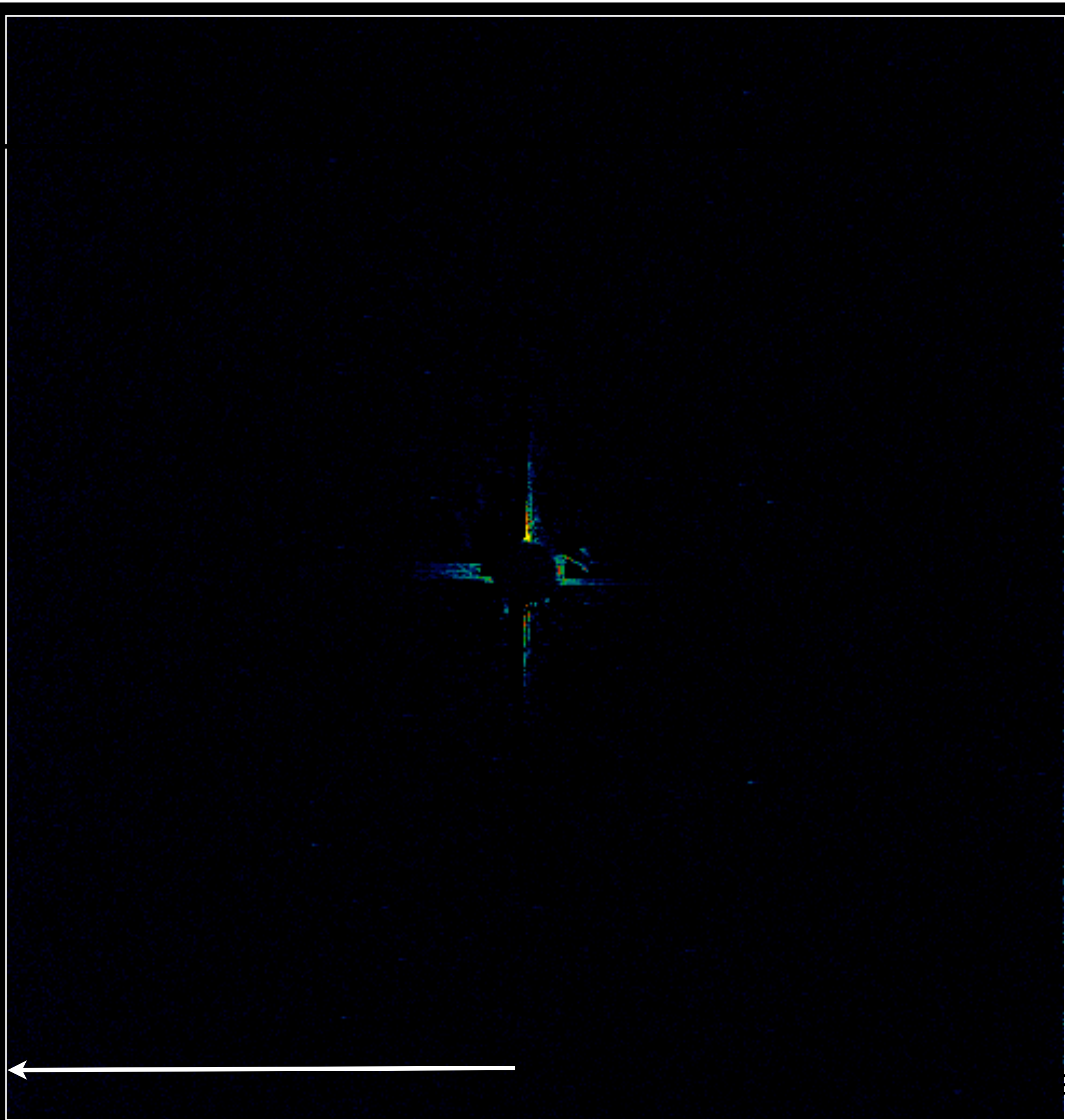


with Ilme Schlichting, Max Planck

latex sphere clusters, measured at 7.0 nm wavelength

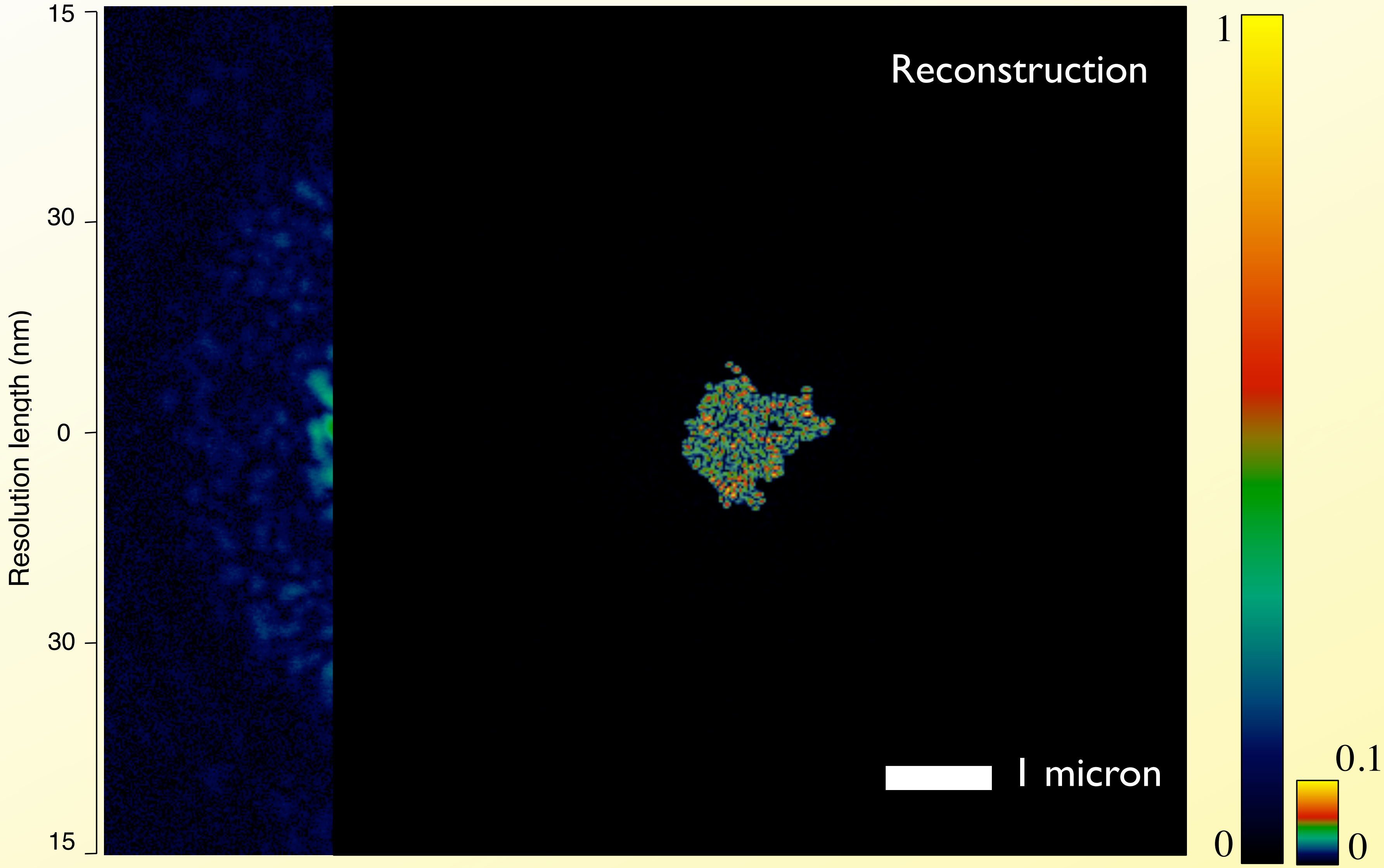


10 fs FEL pulses at 10 μs

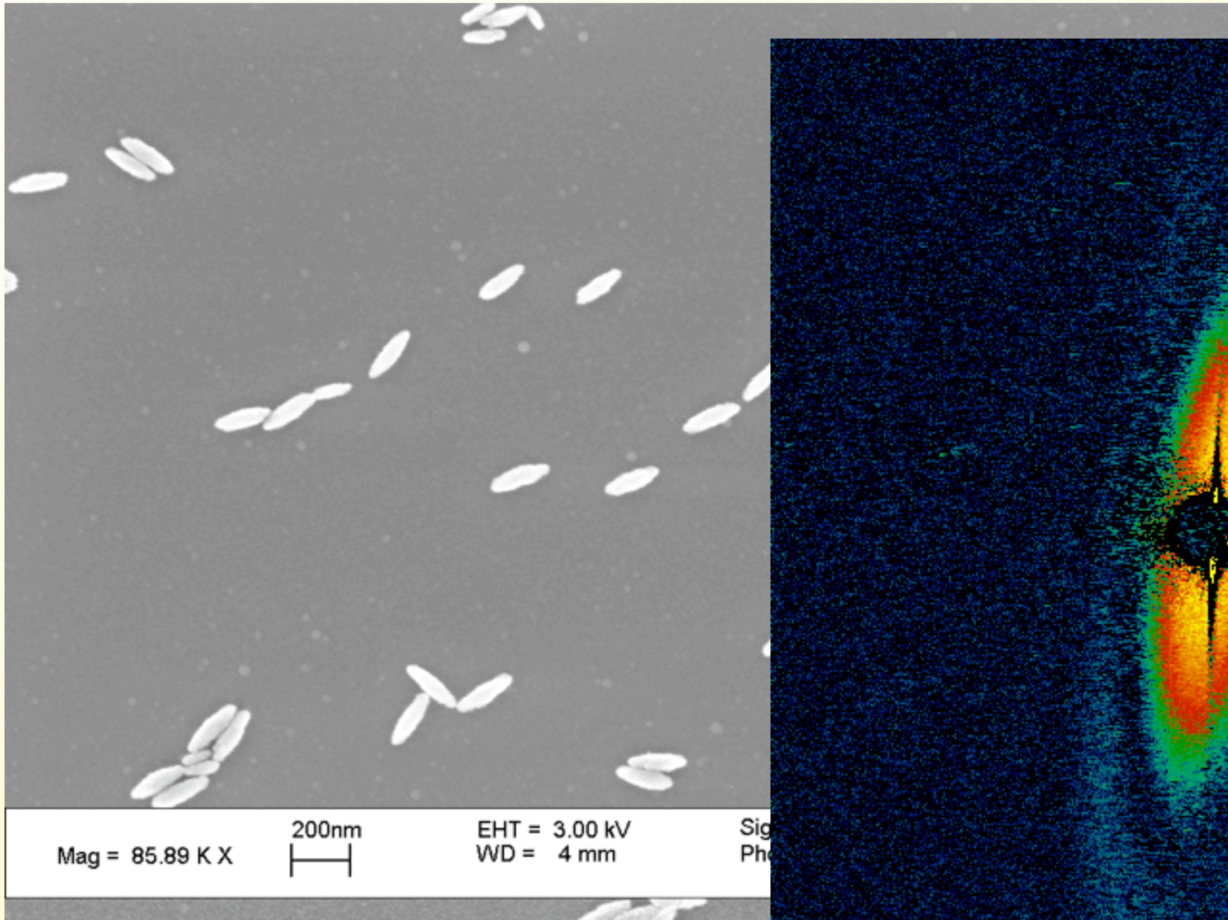


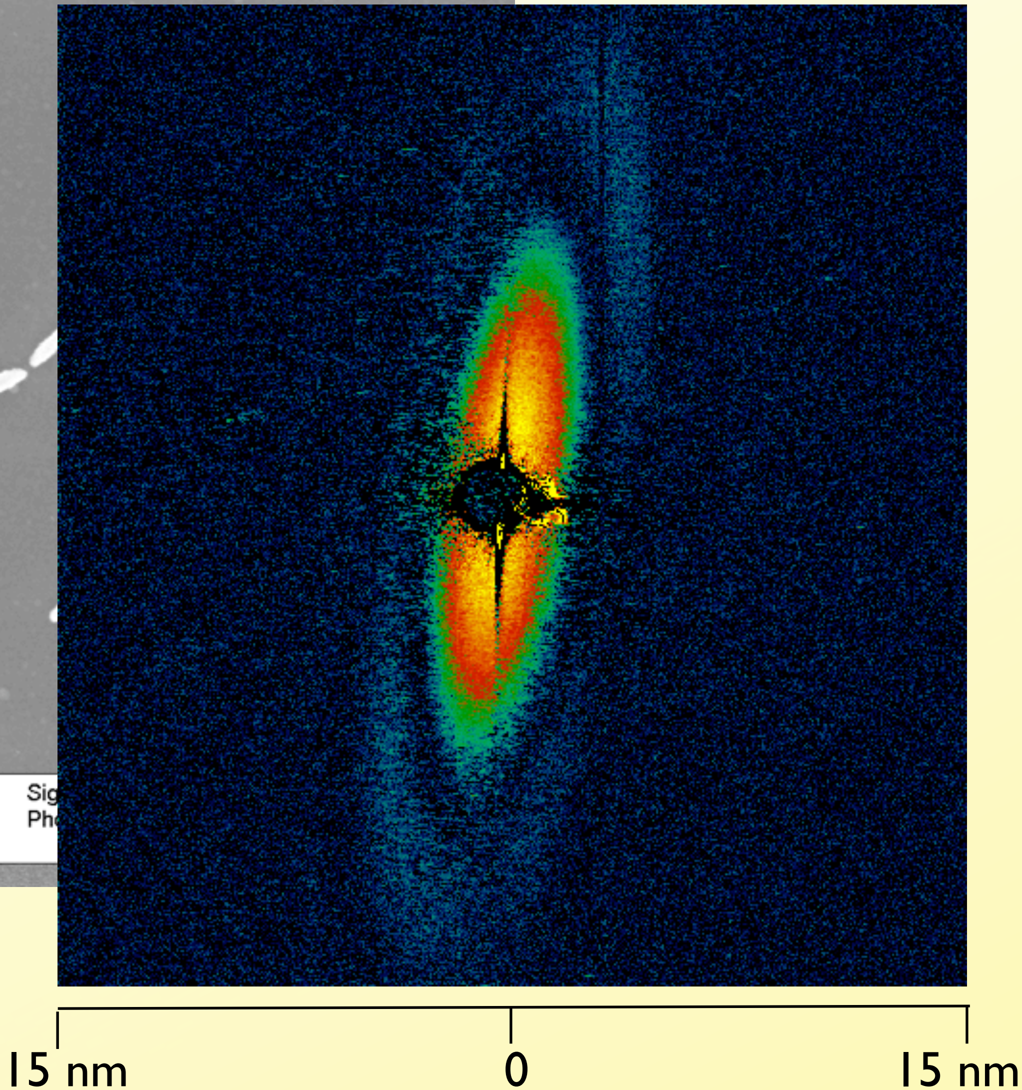
15 nm resolution length

The absence of a substrate gives clean patterns free of aliased scattering sources and plasma radiation



We collected serial diffraction of unoriented iron oxide particles

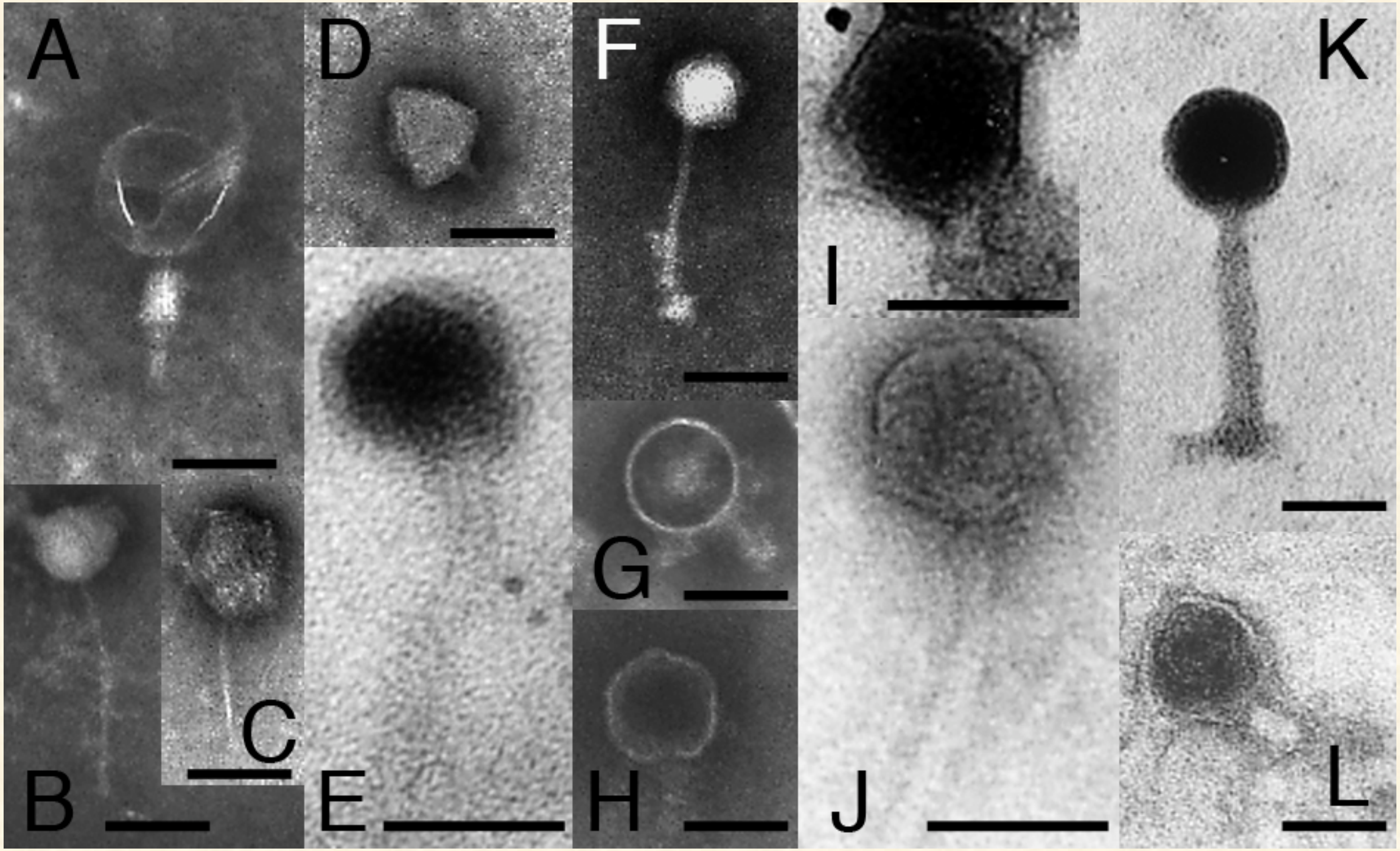




particles: 50 × 200 nm
FEL: 7 nm, 500 pulses/sec

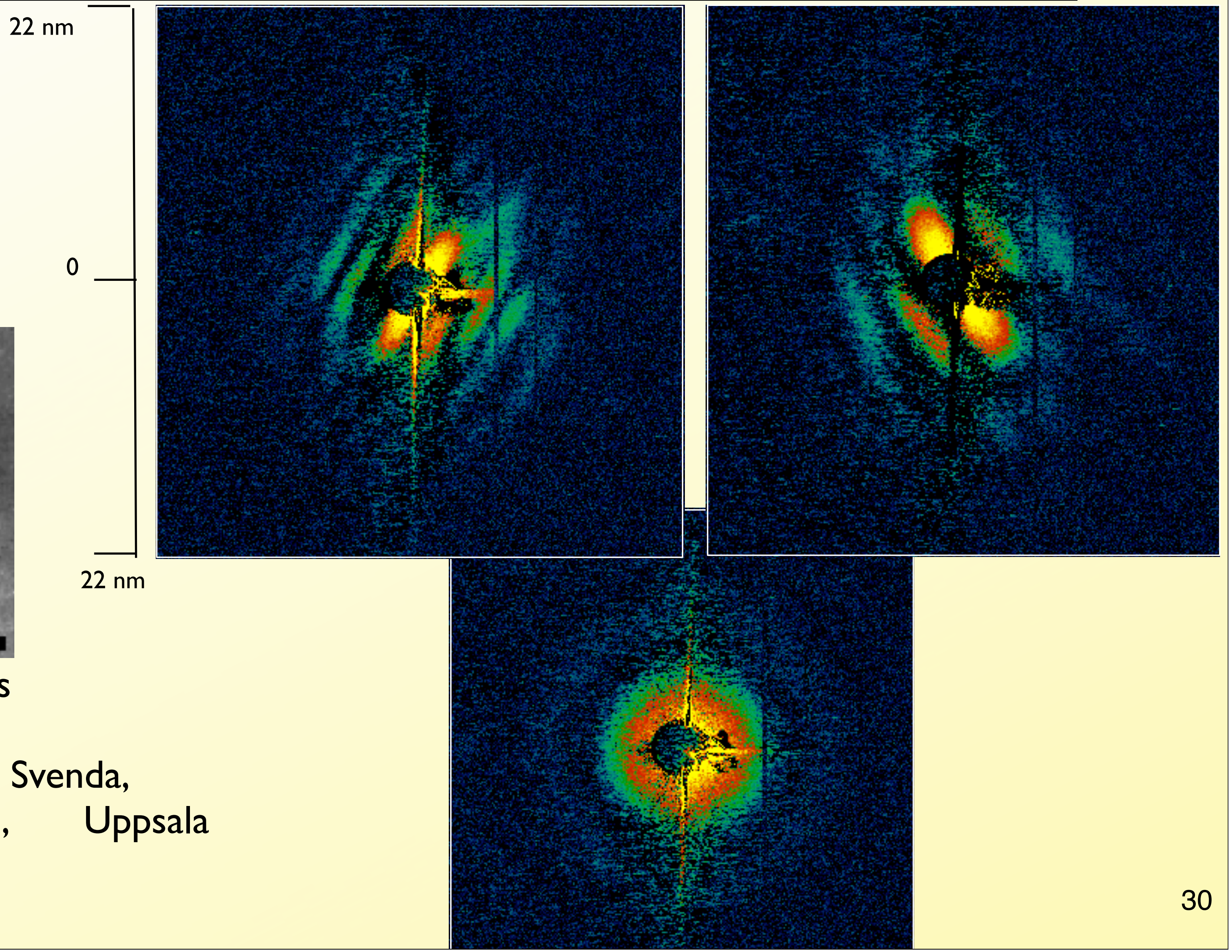
Marine viruses at FLASH

3 different families, different genome sizes, various morphologies



All scale bars = 50 nm

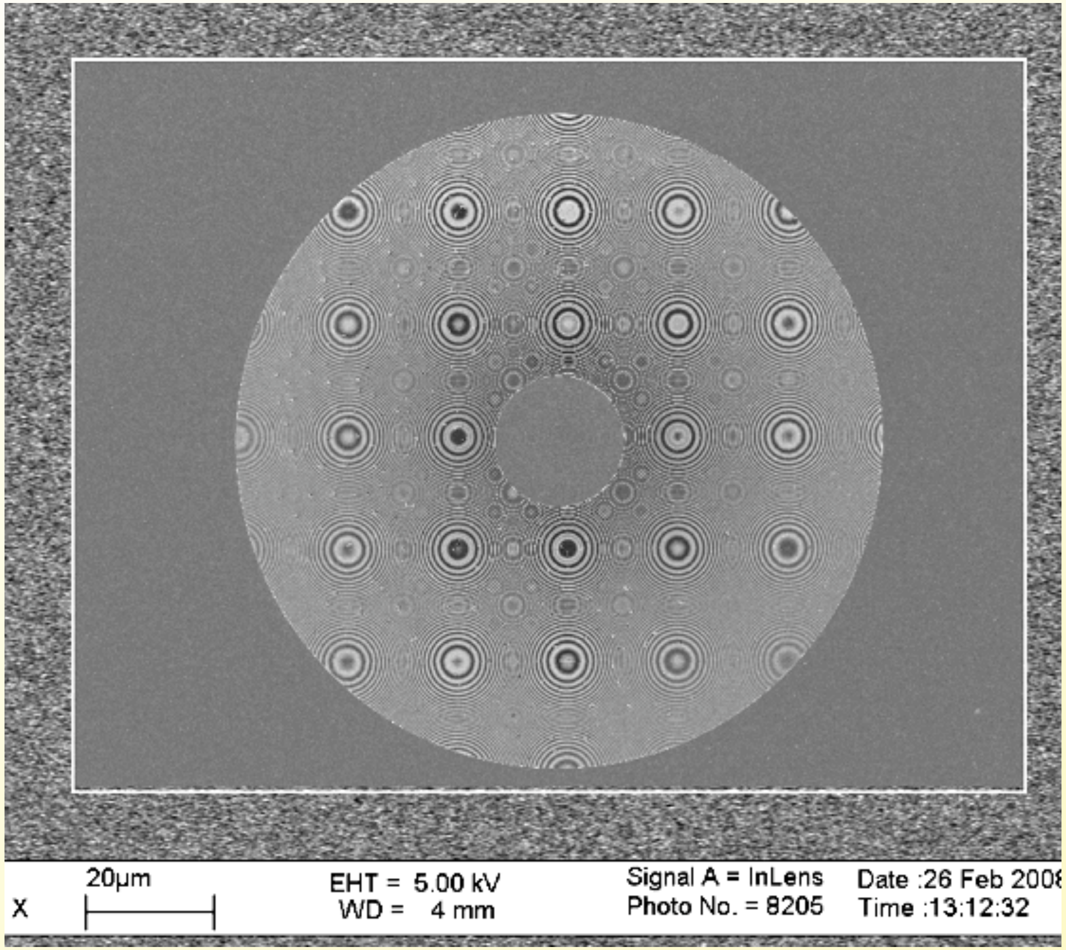
We have recorded many single-pulse patterns of viruses

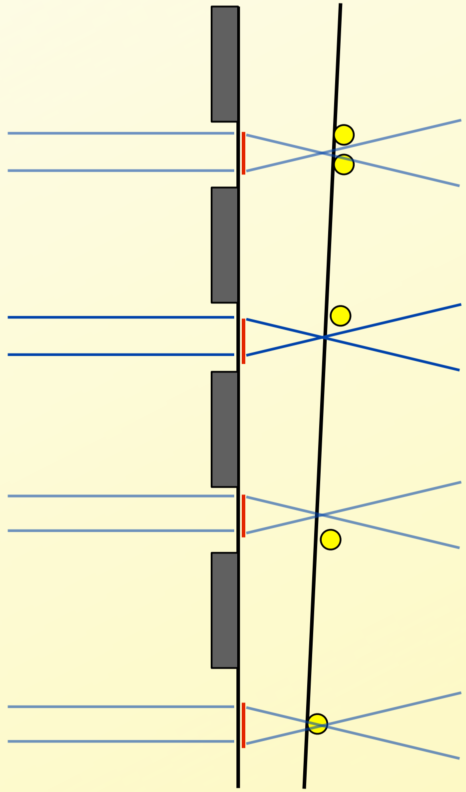


Marine virus

with Martin Svenda,
Janos Hajdu, Uppsala
University

Higher intensity can be achieved using exploding zone plates

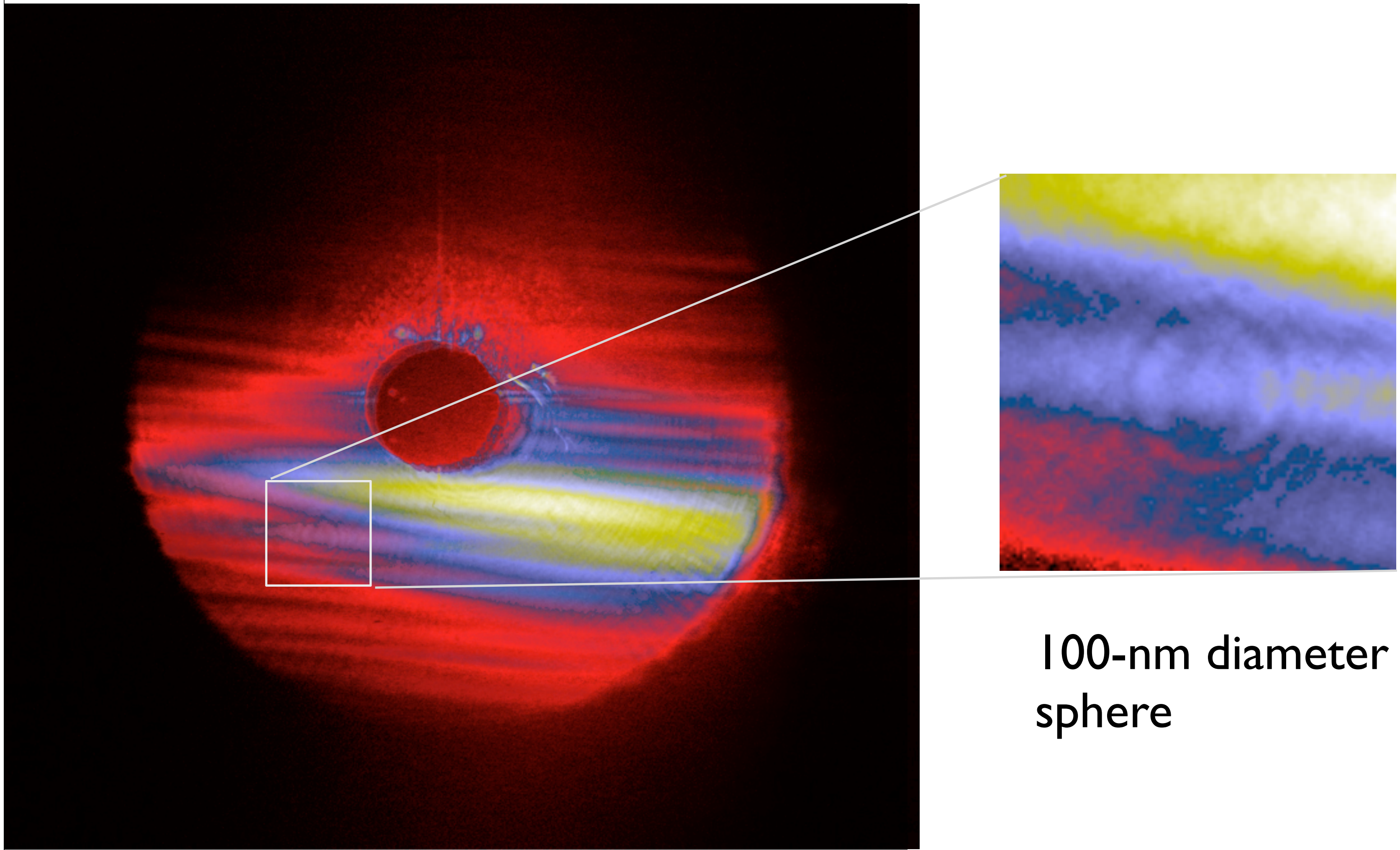




Anne Sakdinawat, CXRO, LBNL

Array of ~200 zone plates
30 nm zone width
100 micron diameter
0.5 mm focus at 7 nm wavelength

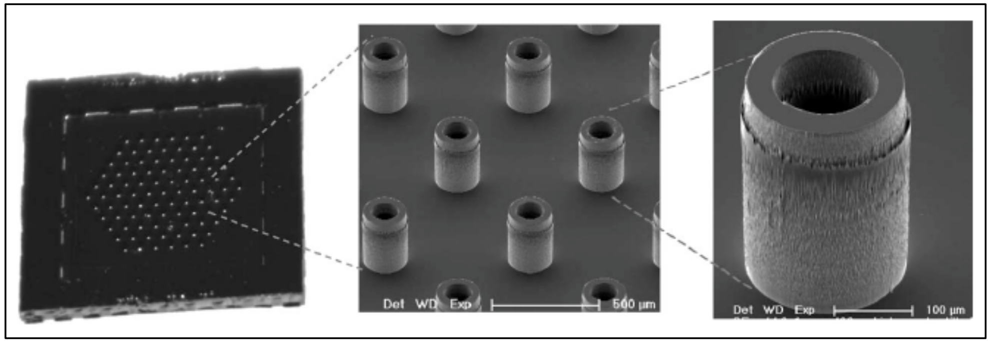
Higher intensity can be achieved using exploding zone plates

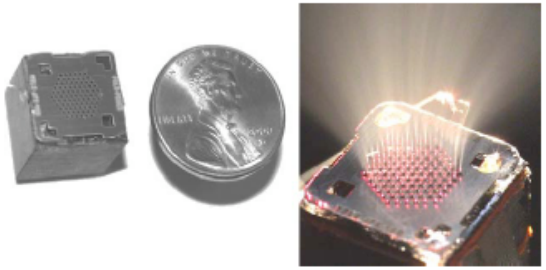


100-nm diameter latex sphere

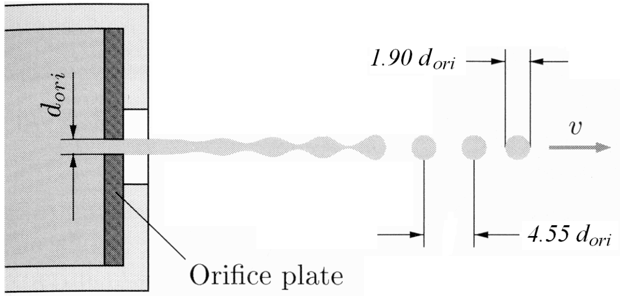
We must develop higher-brightness particle beams

Multiplexed electrospray nozzles increase aerosol density

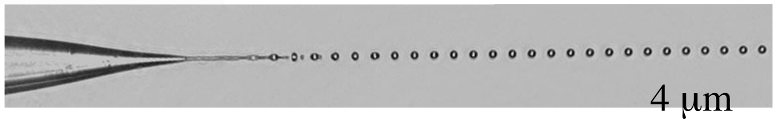




Droplet beams are collimated and generated at MHz rates

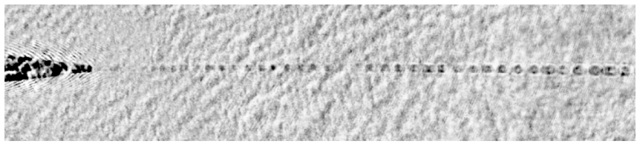


Rayleigh in vacuum

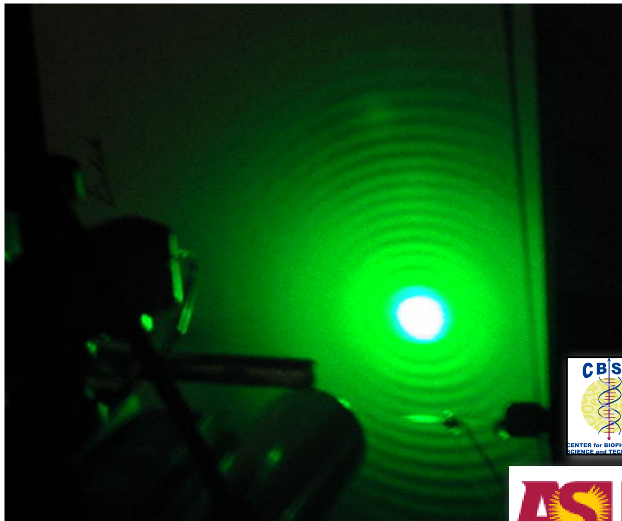


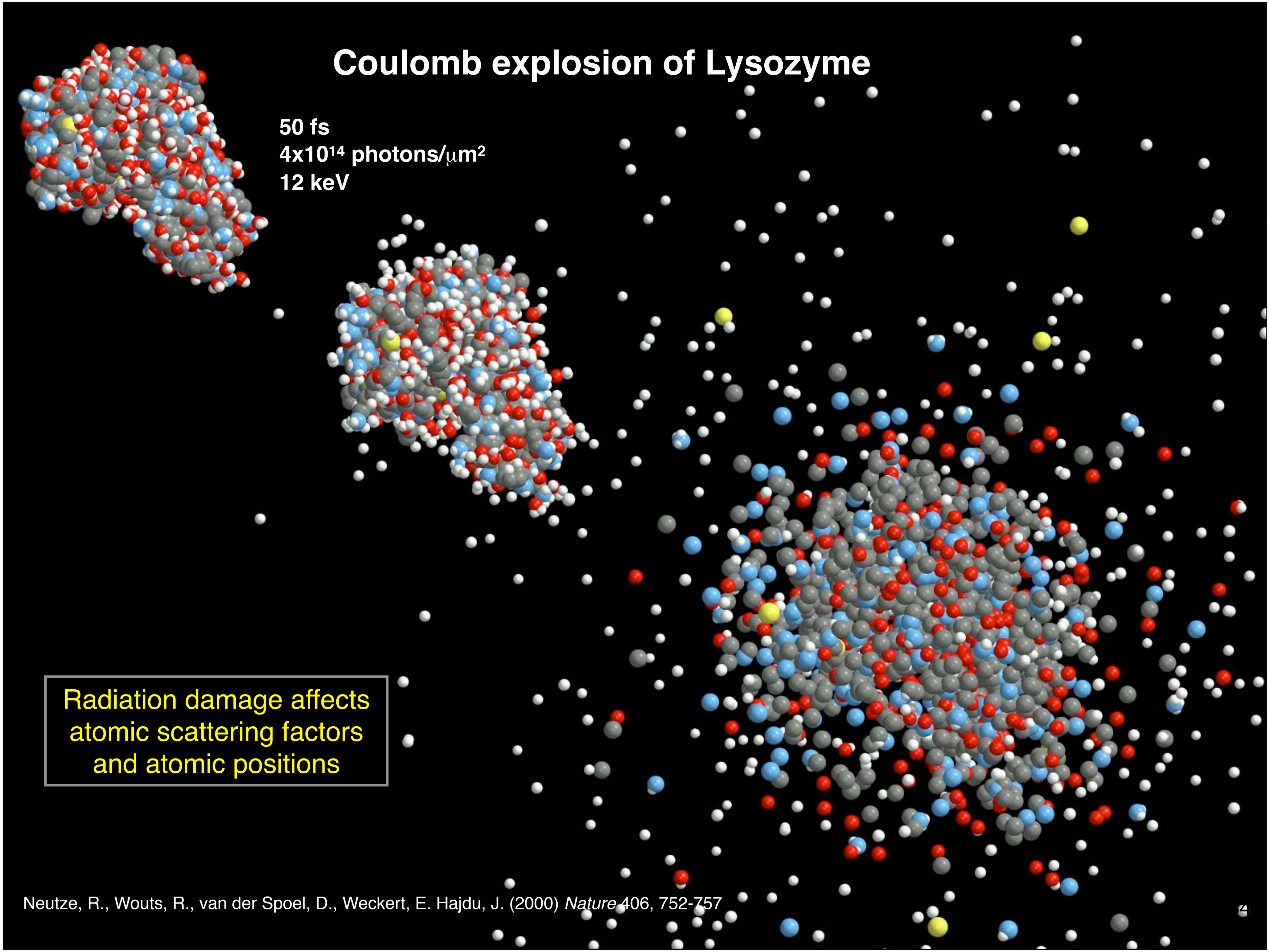
4 µm

Electrospray in vacuum

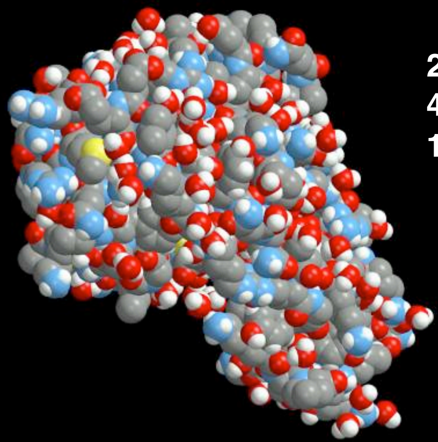


0.3 µm

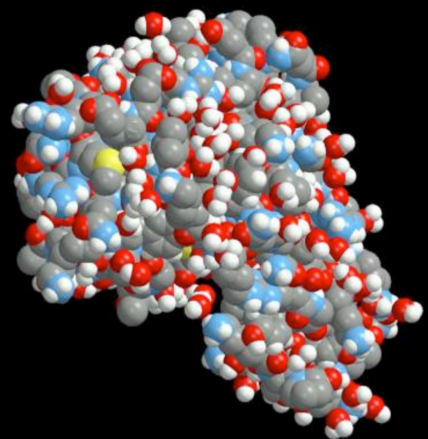


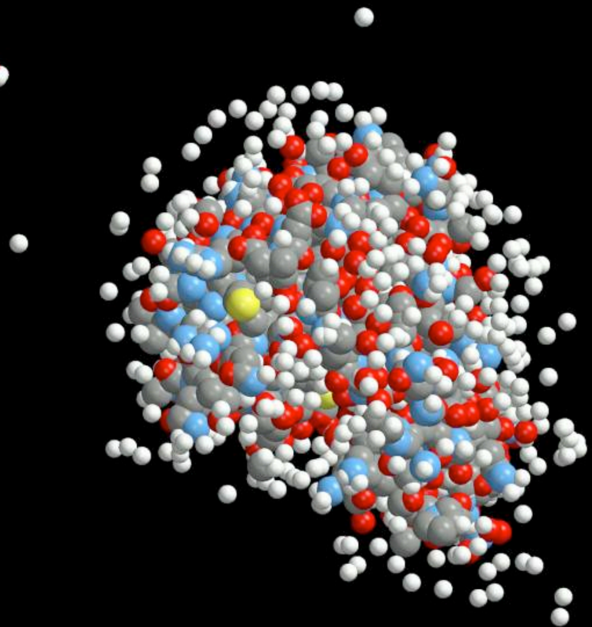

Coulomb explosion of Lysozyme
50 fs
4x10^14 photons/μm^2
12 keV
Radiation damage affects atomic scattering factors and atomic positions
Neutze, R., Wouts, R., van der Spoel, D., Weckert, E. Hajdu, J. (2000) Nature 406, 752-757

Coulomb explosion of Lysozyme



20 fs
4×10^{14} photons/μm^2
12 keV



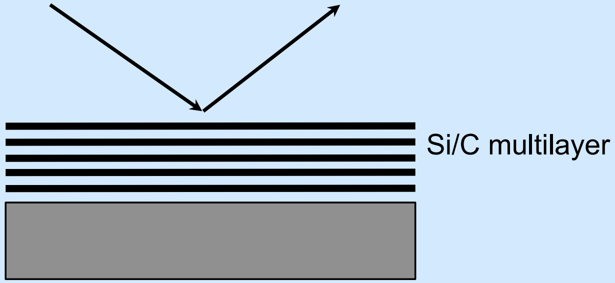


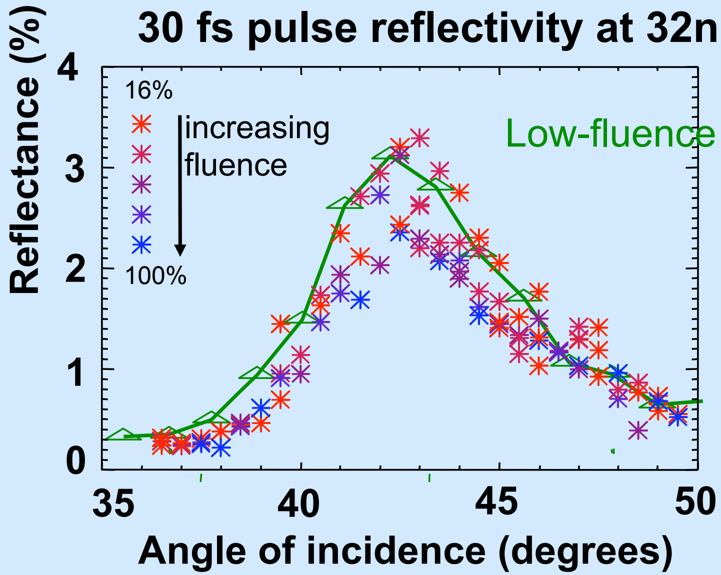
Radiation damage affects atomic positions and atomic scattering factors

Neutze, R., Wouts, R., van der Spoel, D., Weckert, E. Hajdu, J. (2000) *Nature* 406, 752-757

First EUV-FEL experiments show that structural information can be obtained before destruction

During 30 fs pulse (10^{14} W cm^{-2})
32 nm wavelength



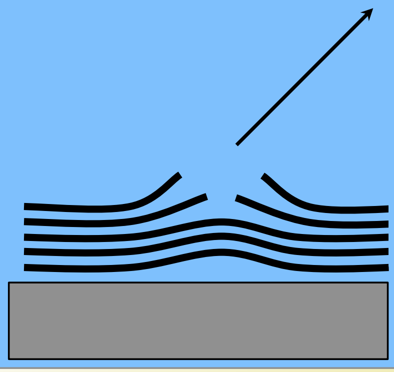


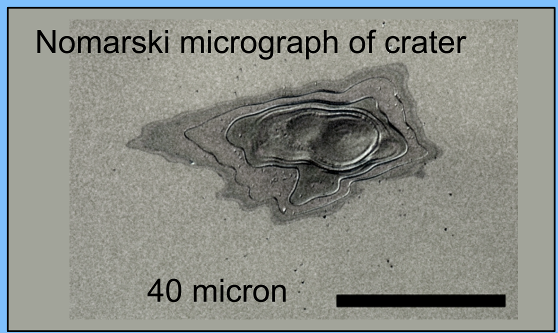
Reflectivity unchanged

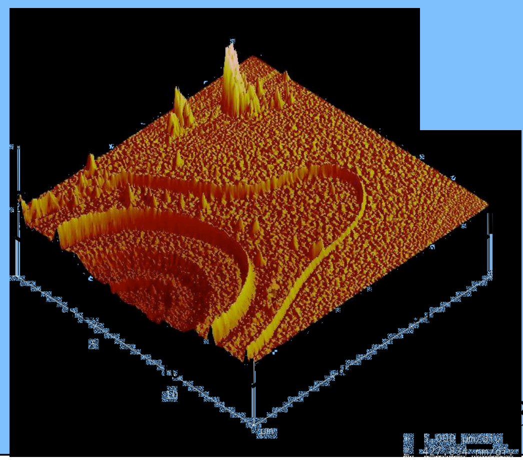
Multilayer *d* spacing not changed by more than 0.3 nm

After pulse

Plasma forms, layers ablate

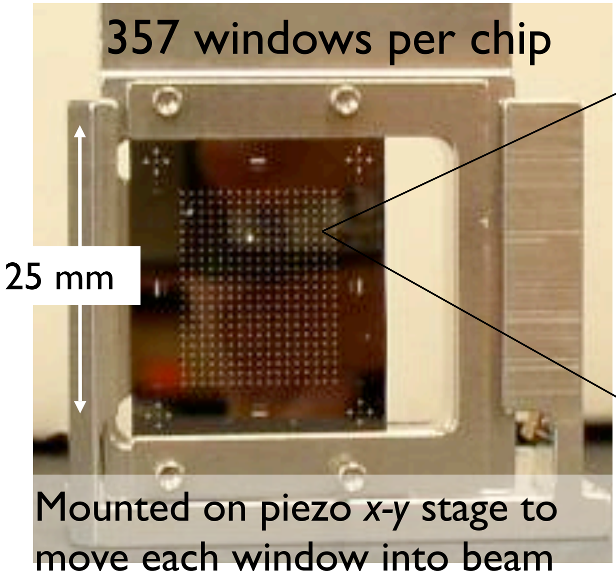


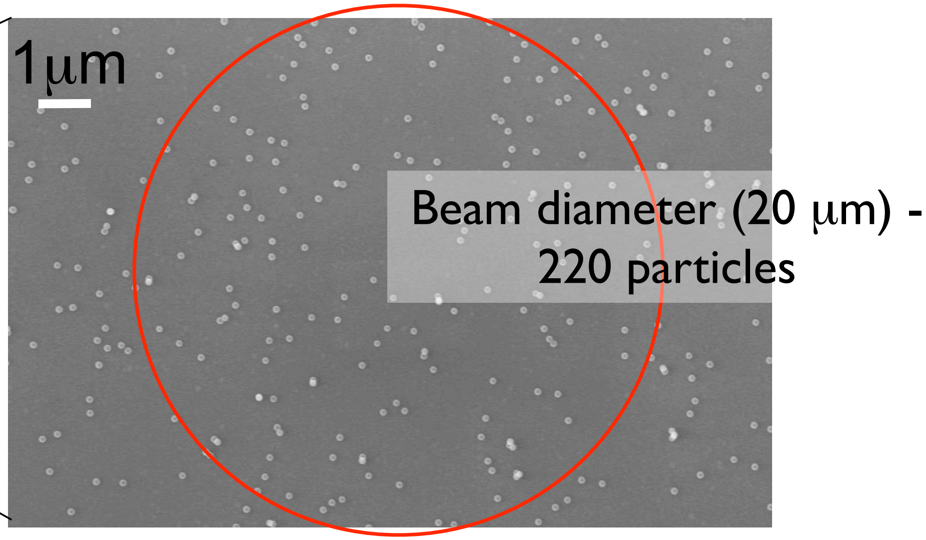




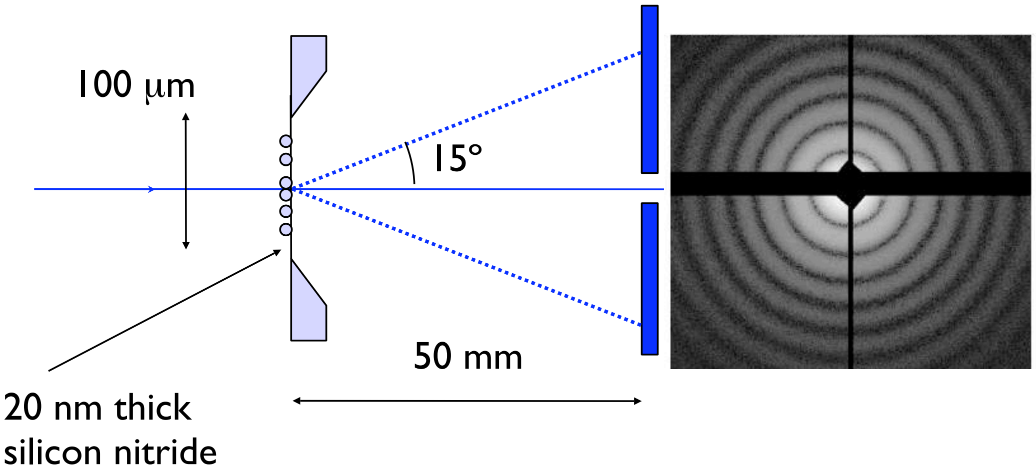
S.P. Hau-Riege et al., Phys Rev Lett **98**, 145502 (2007)

Particle explosion experiments were performed at FLASH on latex particles on membranes



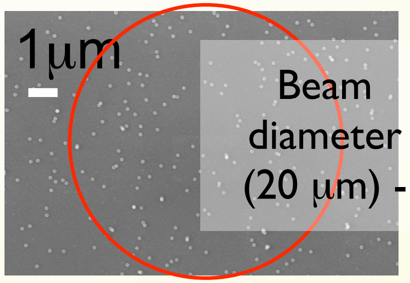


Latex particles on membrane

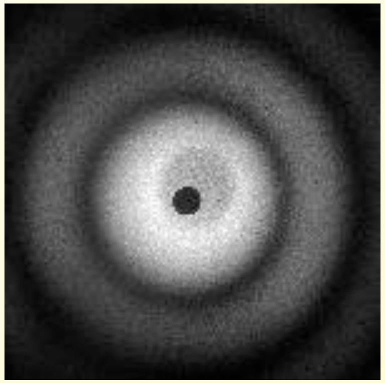


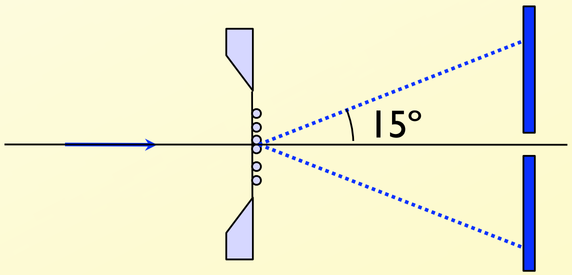
The particle size is determined by Mie scattering of the FEL pulse by the particles (FEL pulse is both pump and probe)

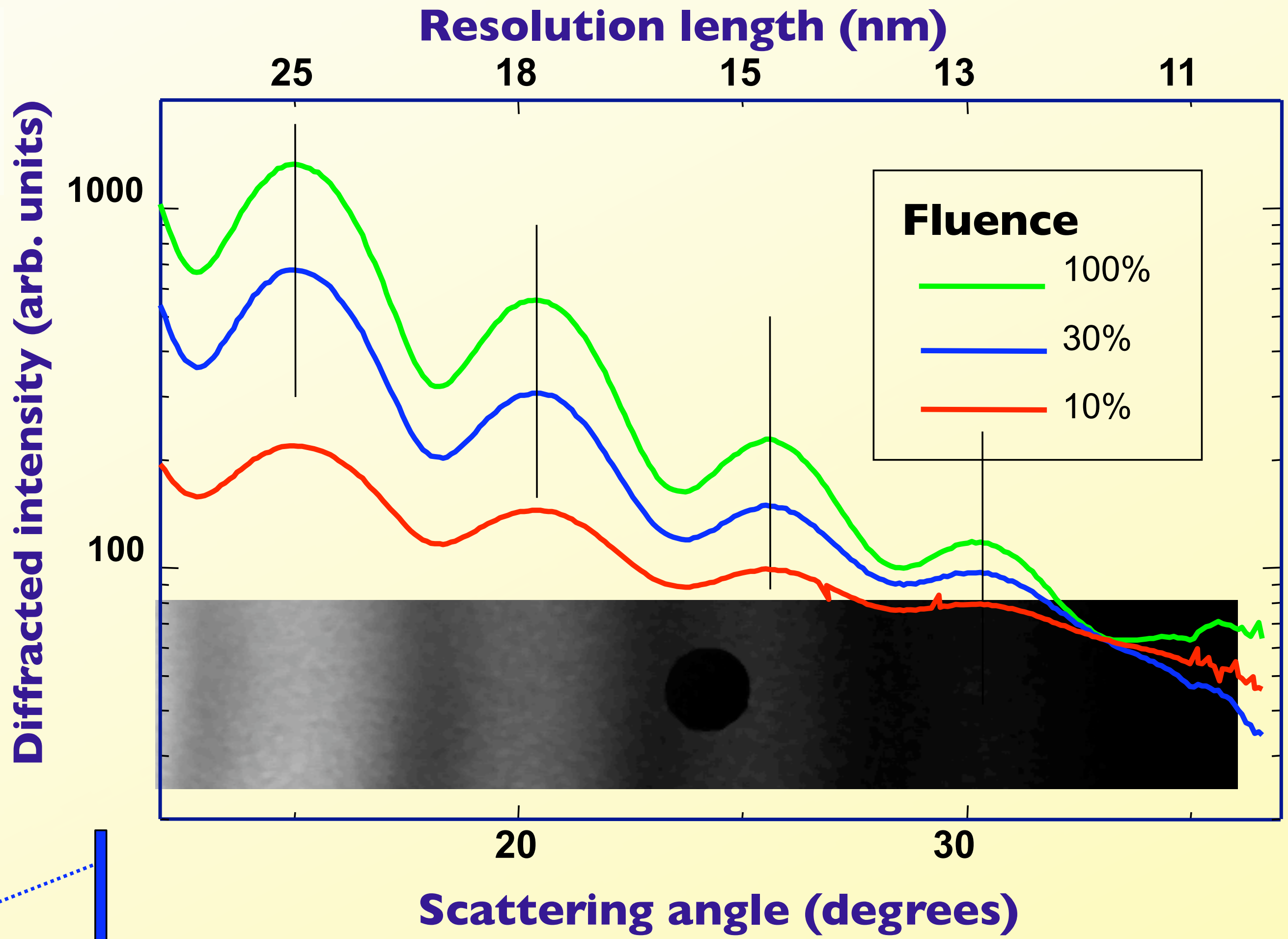
High-angle diffraction shows no change in structure of particles greater than 12 nm



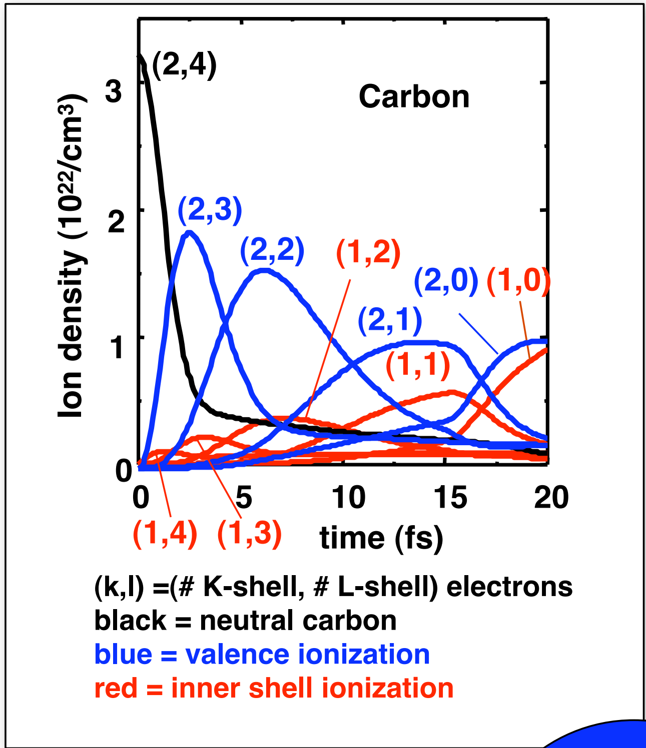
Latex particles on membrane







XFEL diffraction of molecules and clusters is modified (damaged) by photoionization and motion of atoms

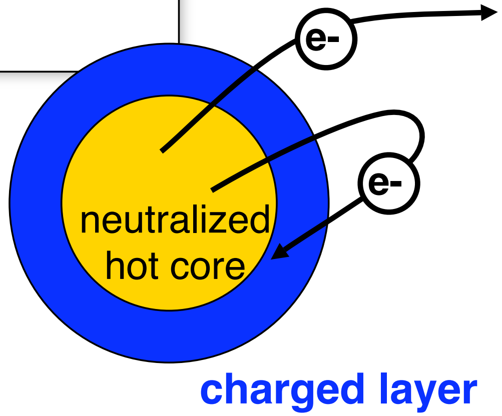


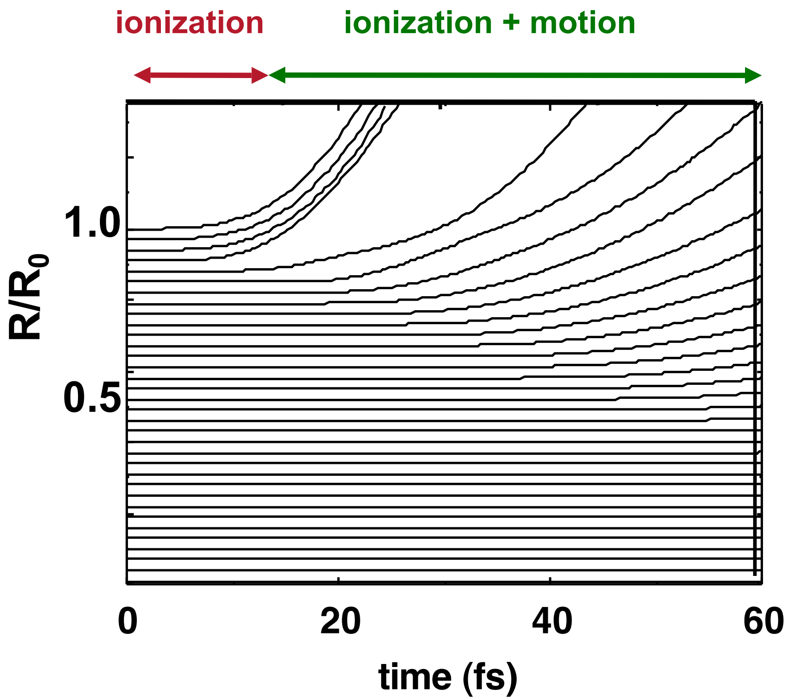
We have developed a *hydrodynamic continuum model* for the atomic motion and the ionization processes that can treat large and small molecules

- Allows for trapping and secondary effects (such as inverse Bremsstralung, 3-body recombination)
- Damage is dominated by ionization at short times
- A tamper reduces motion

electron escapes if

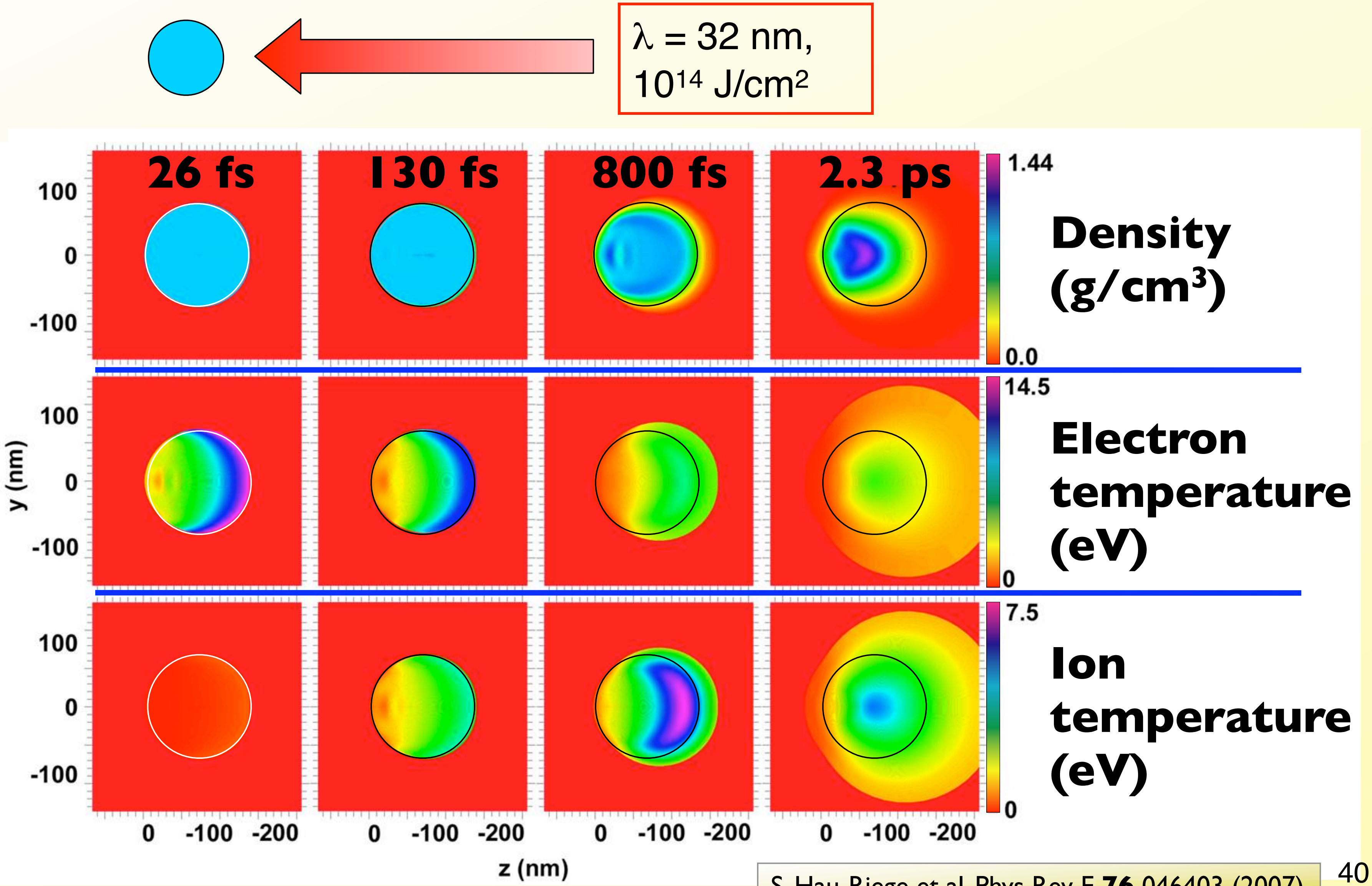
$$E > \frac{3eQ}{2R}$$





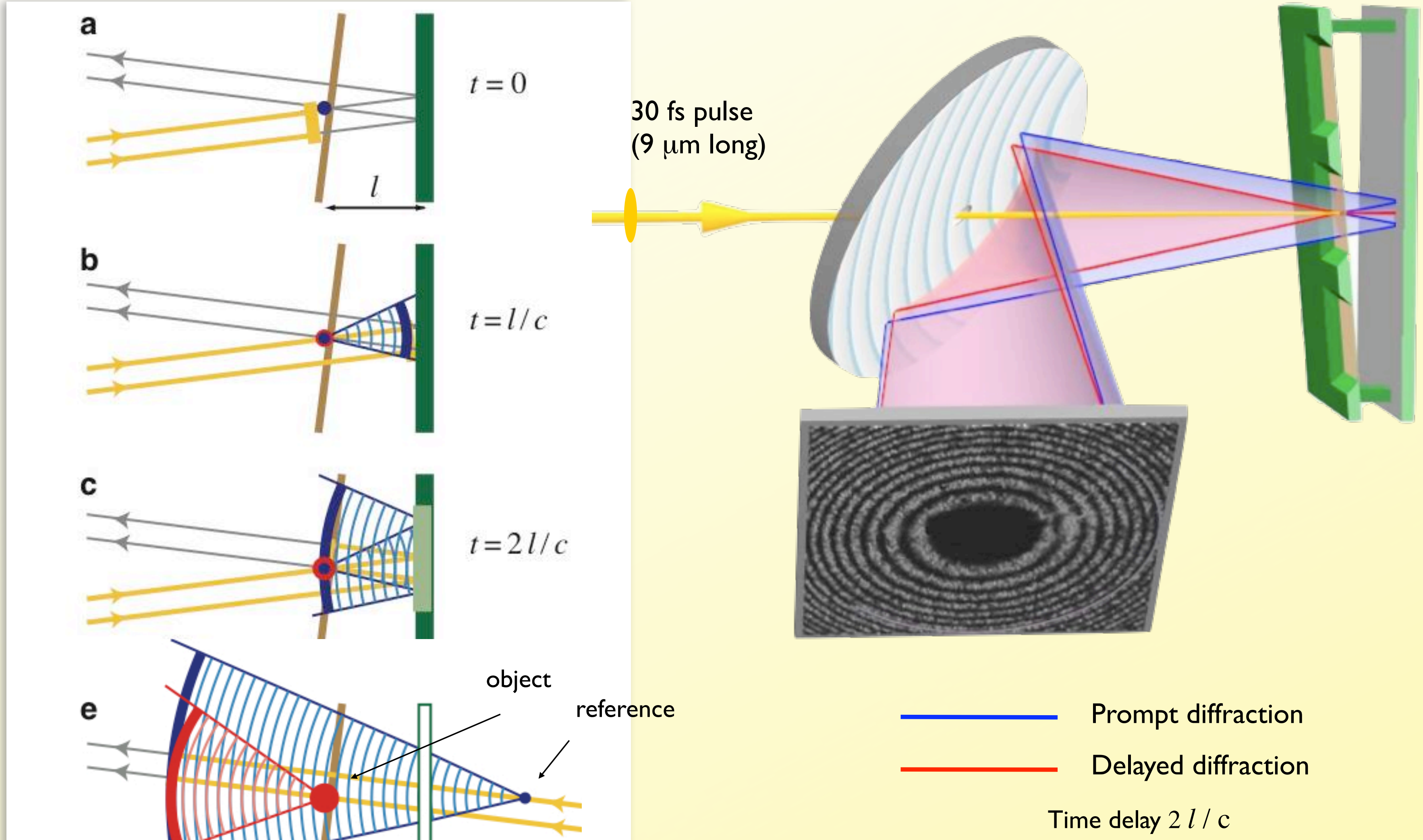
S. Hau-Riege et al, Phys Rev E **69**, 051906 (2004)

Our VUV hydrodynamic code shows that latex spheres start exploding in ~ 2 ps



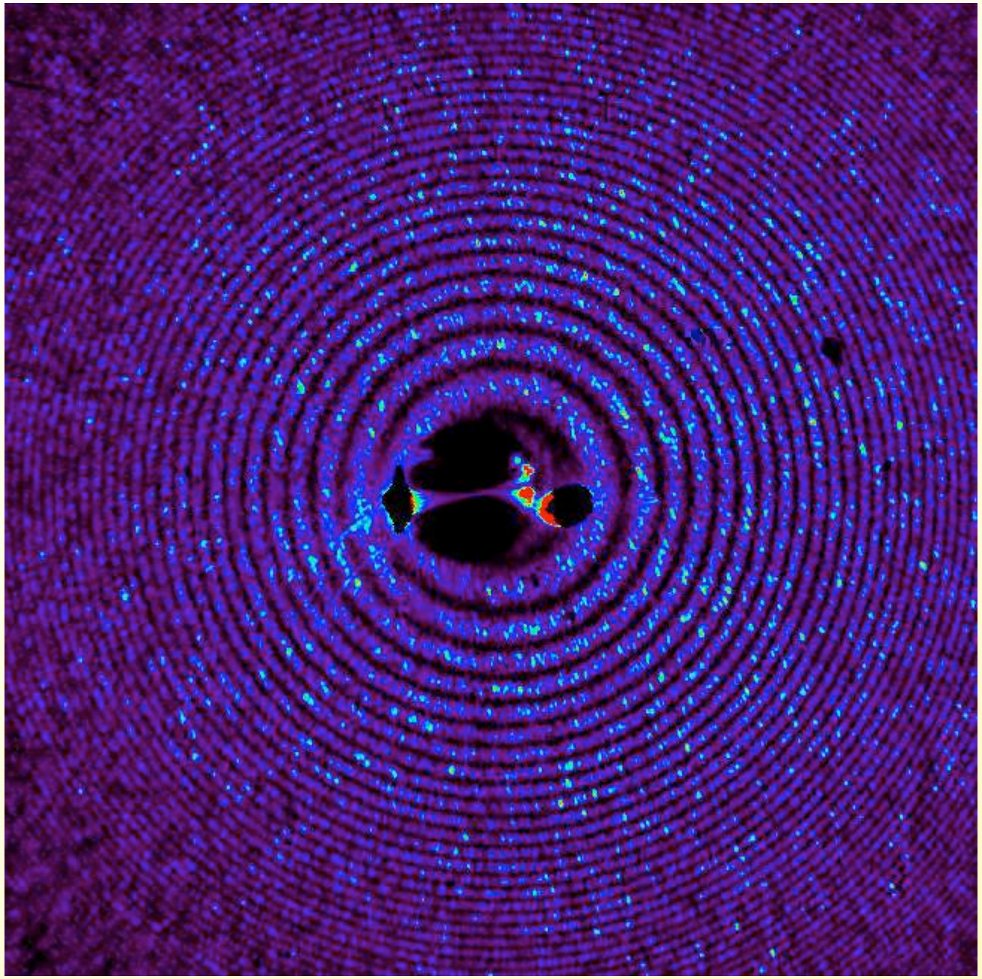
S. Hau-Riege et al, Phys Rev E **76** 046403 (2007)

We invented a new method called femtosecond time-delay holography

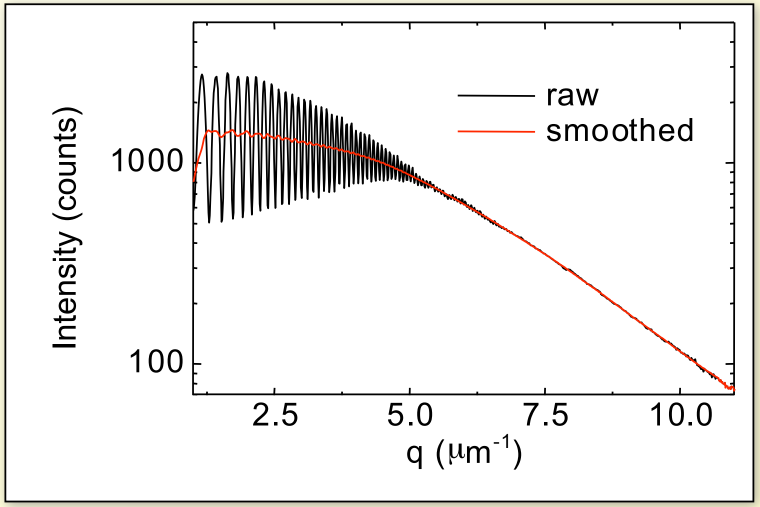


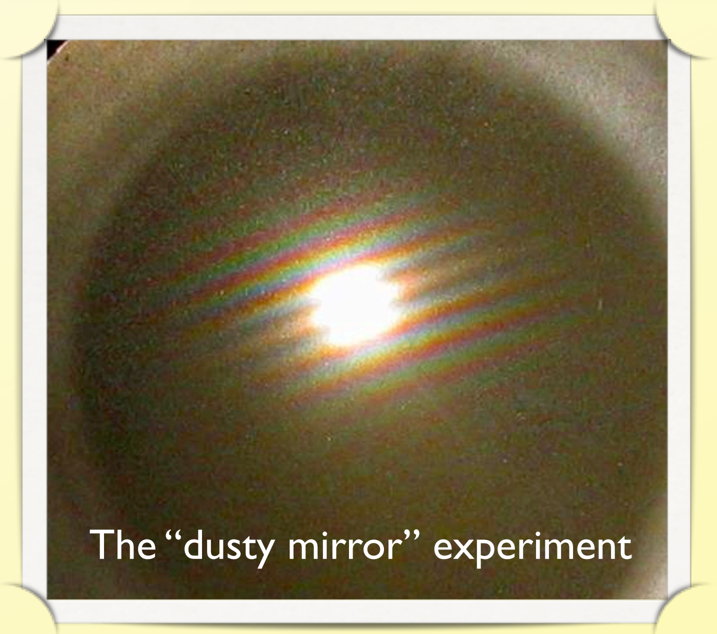
H. Chapman et al., Nature **448** 676 (2007)

First demonstration of time-delay holography with 3 fs time resolution indicates the particle explosion

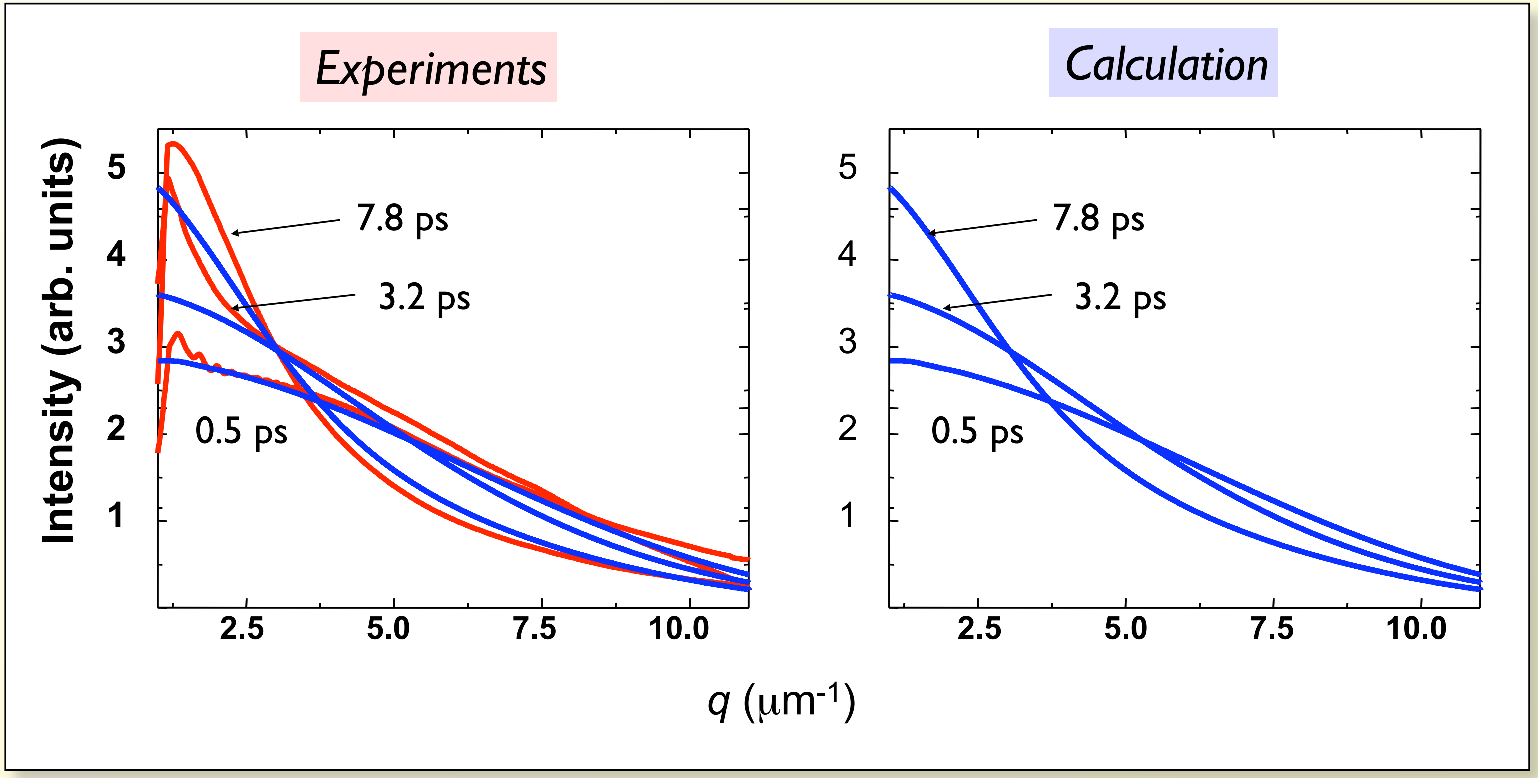


Single shot ultrafast time-delay X-ray hologram, with 300 fs delay





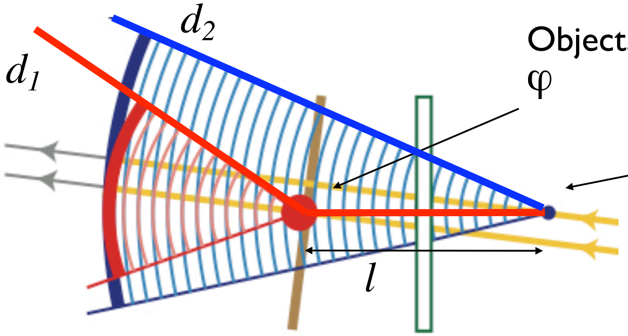
The explosion is in good agreement with our hydrodynamic model



- The structure factor narrows, showing the particle exploding
- The lower resolution shape of the explosion is different than expected
- This is the first high-resolution observation of particle explosions

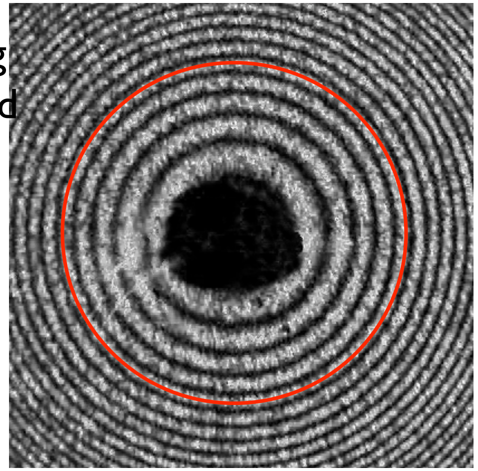
H. Chapman et al., Nature **448** 676 (2007)

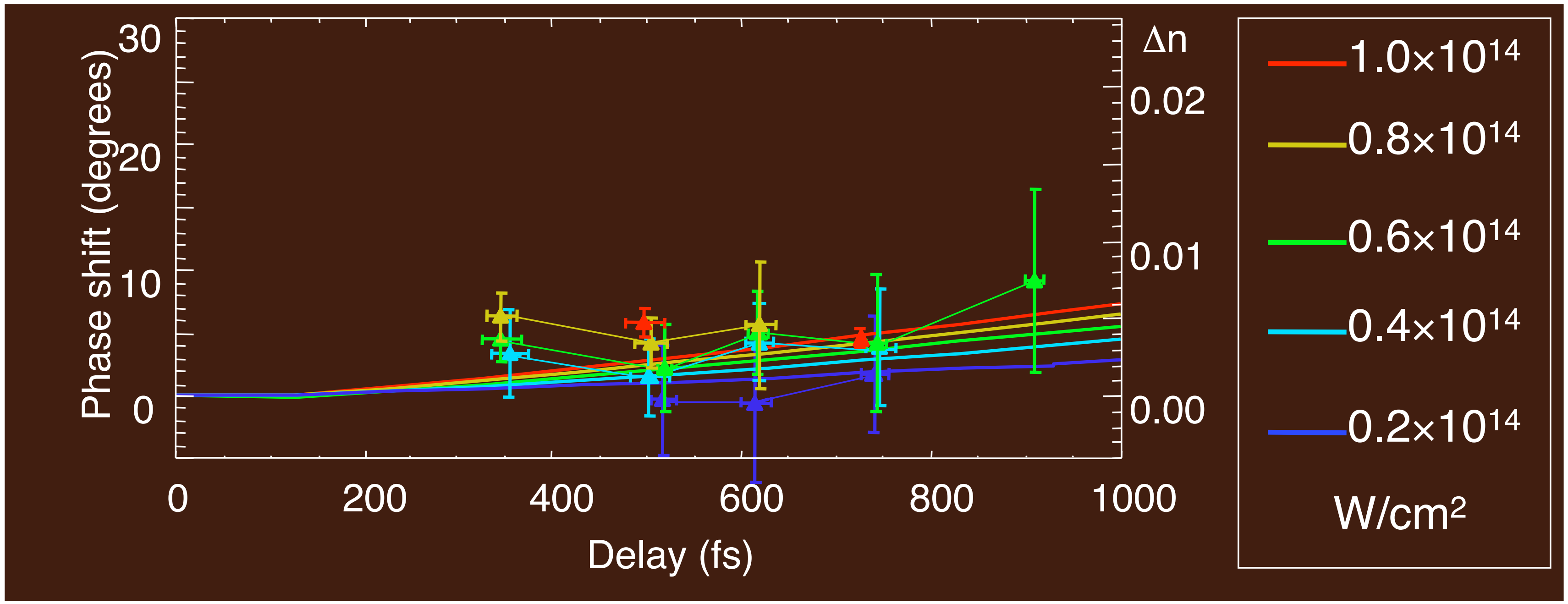
We interferometrically measure the change in optical density of the particle at short delays



Rings occur when $l + \varphi + d_1 = d_2 + N\lambda$

Fitting the ring radii gives l and φ





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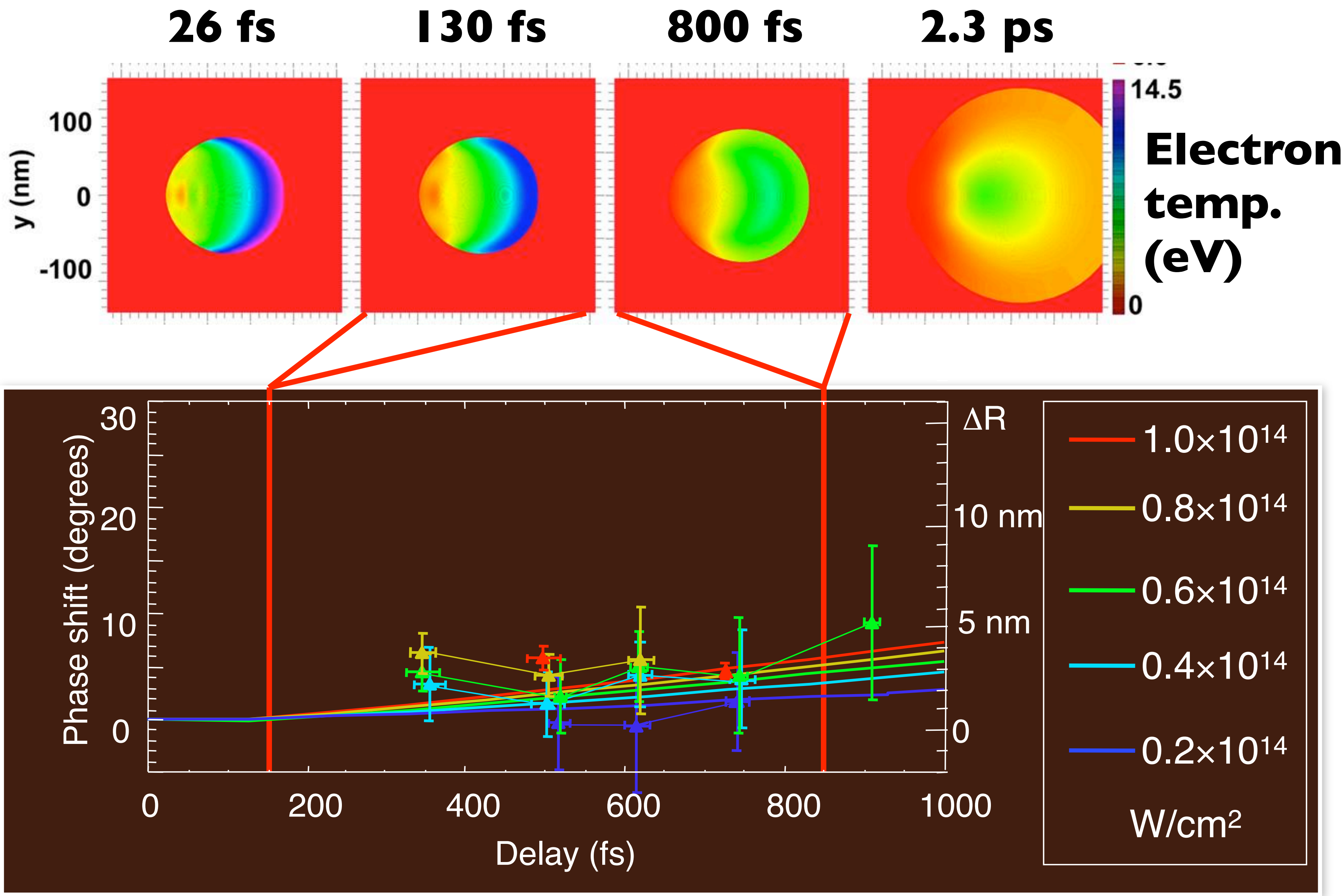
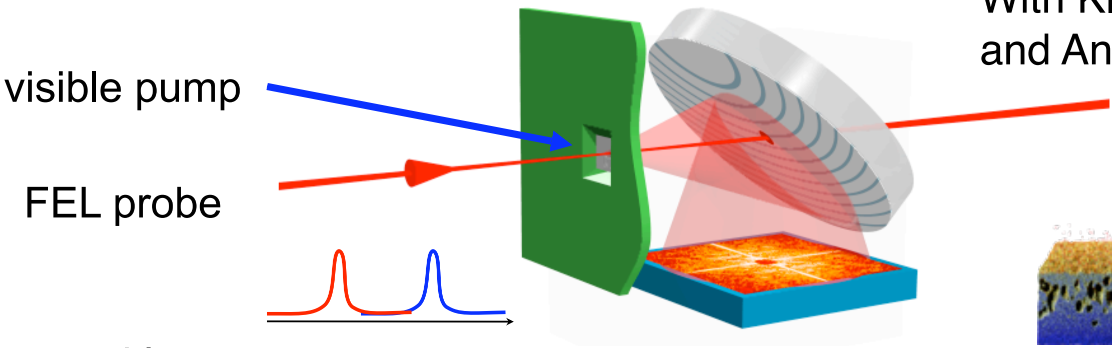
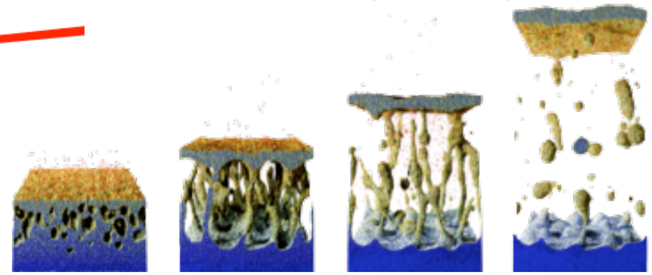


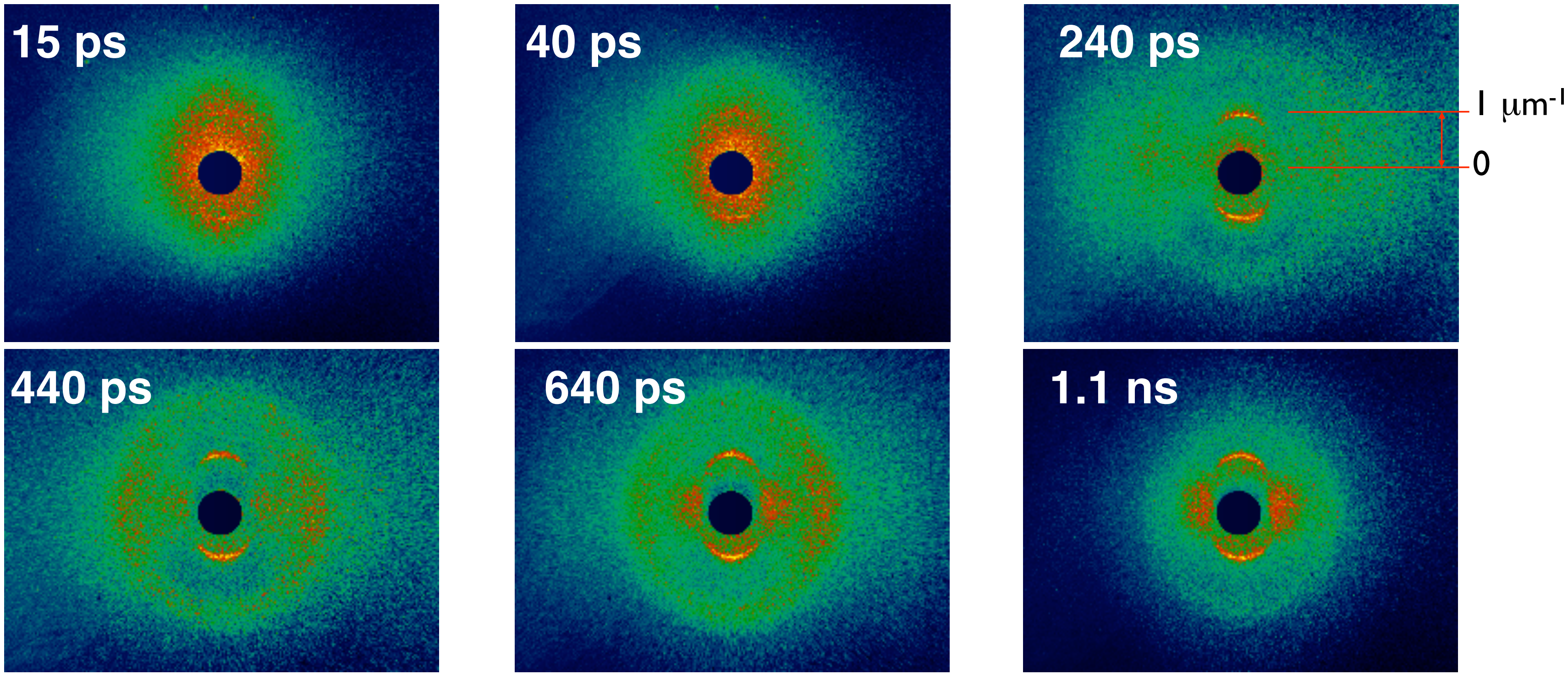
We performed ultrafast coherent X-ray diffraction to study ablation of materials

With Klaus Sokolowski-Tinten (Essen) and Andrea Cavalleri (Oxford)

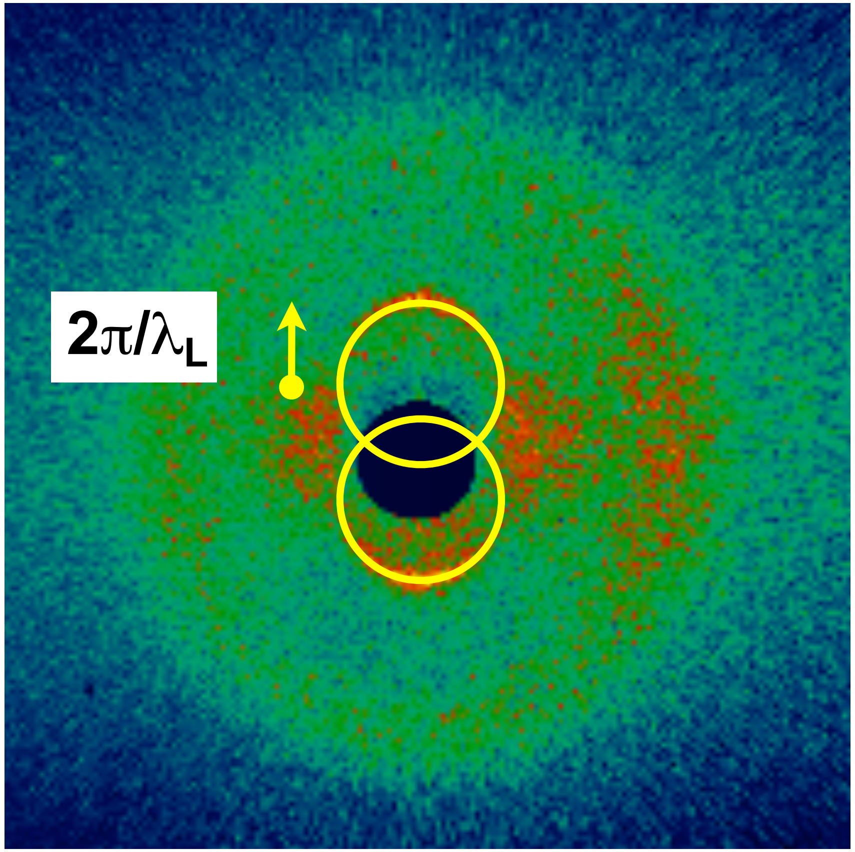


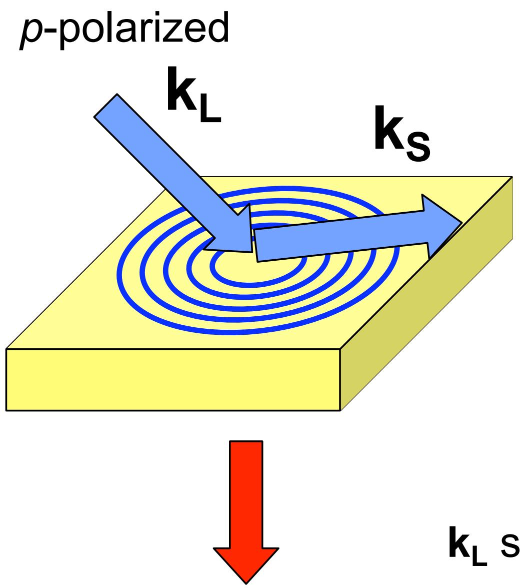


10 ps visible pump

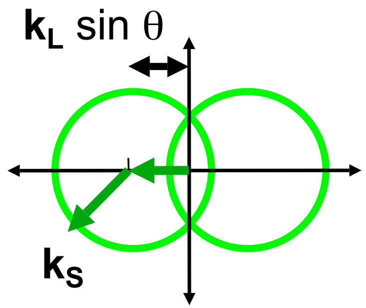


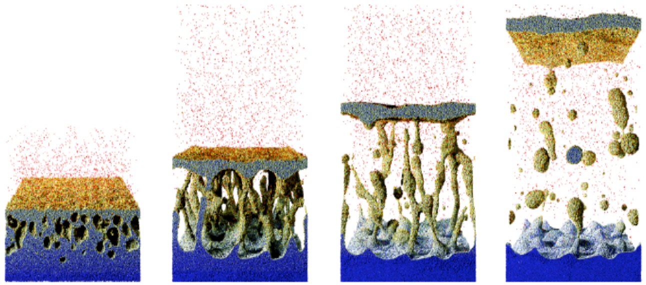
Scattering from Light-Induced Periodic Surface Structures is observed





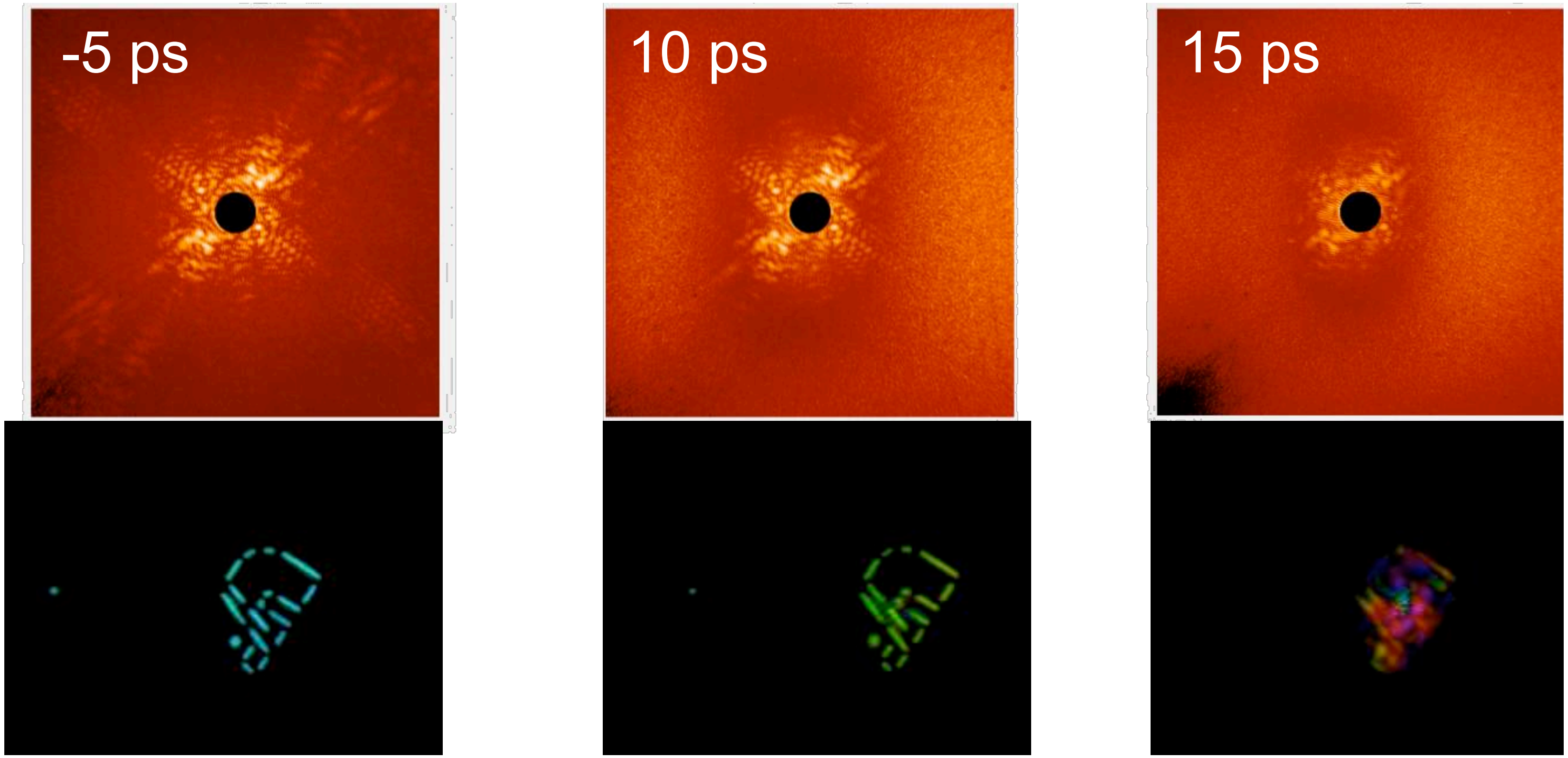
FEL scatters from structure $|\mathbf{k}_L - \mathbf{k}_S|$

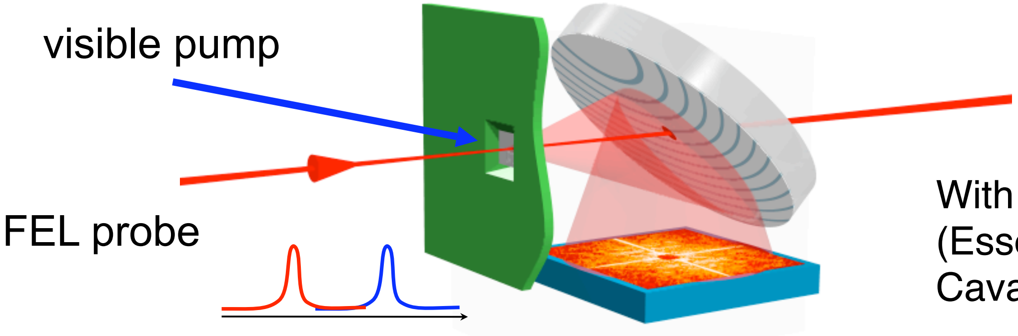


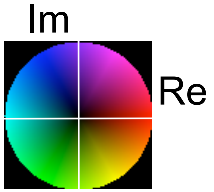


Speckle may be due to diffraction from phase-separated plasma. MD model: Gilmer, LLNL

Laser-induced dynamics can be imaged

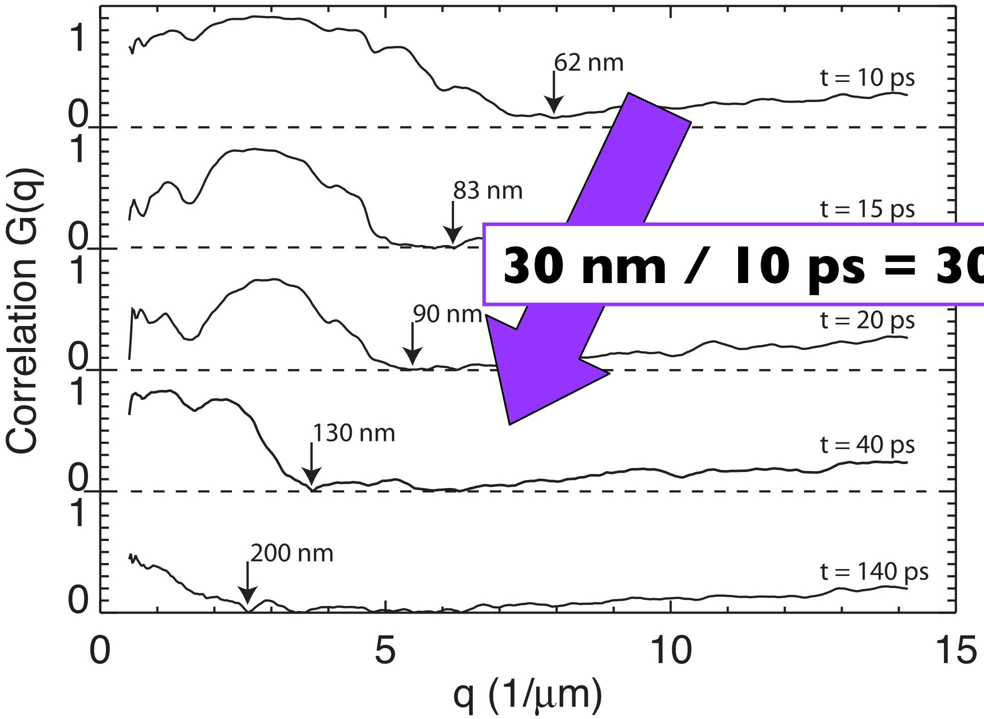




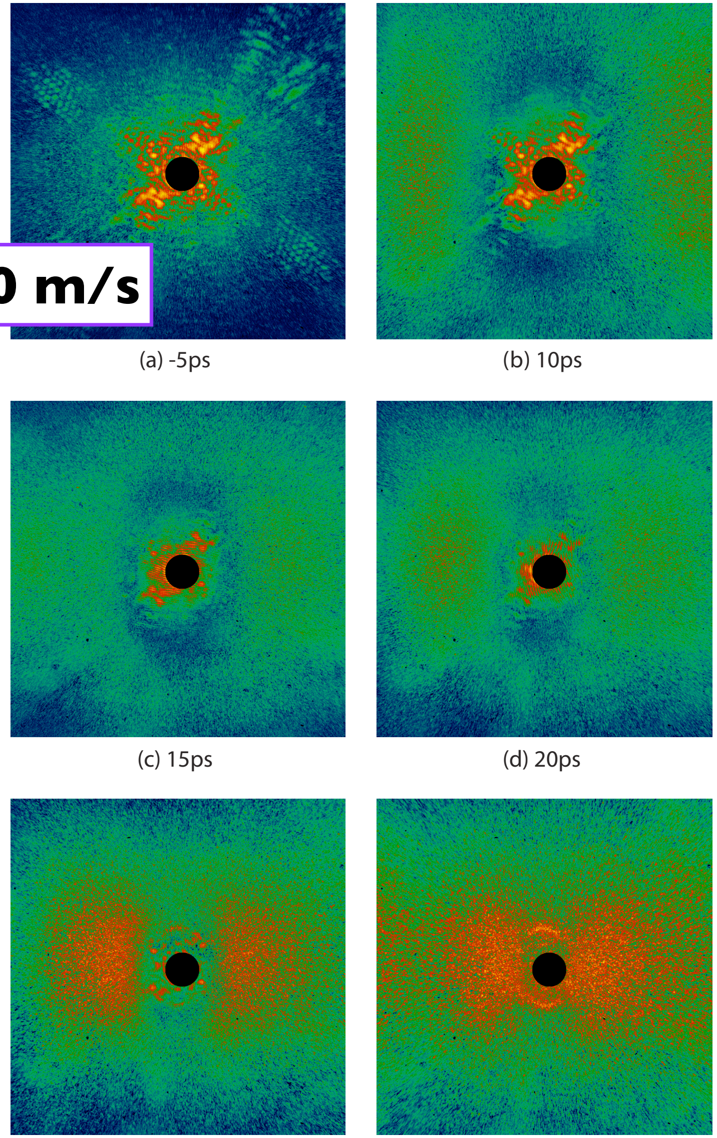


With Klaus Sokolowski-Tinten (Essen) and Andrea Cavalleri (Oxford)

Patterns can be cross correlated to reveal the danamics of the structure



30 nm / 10 ps = 3000 m/s

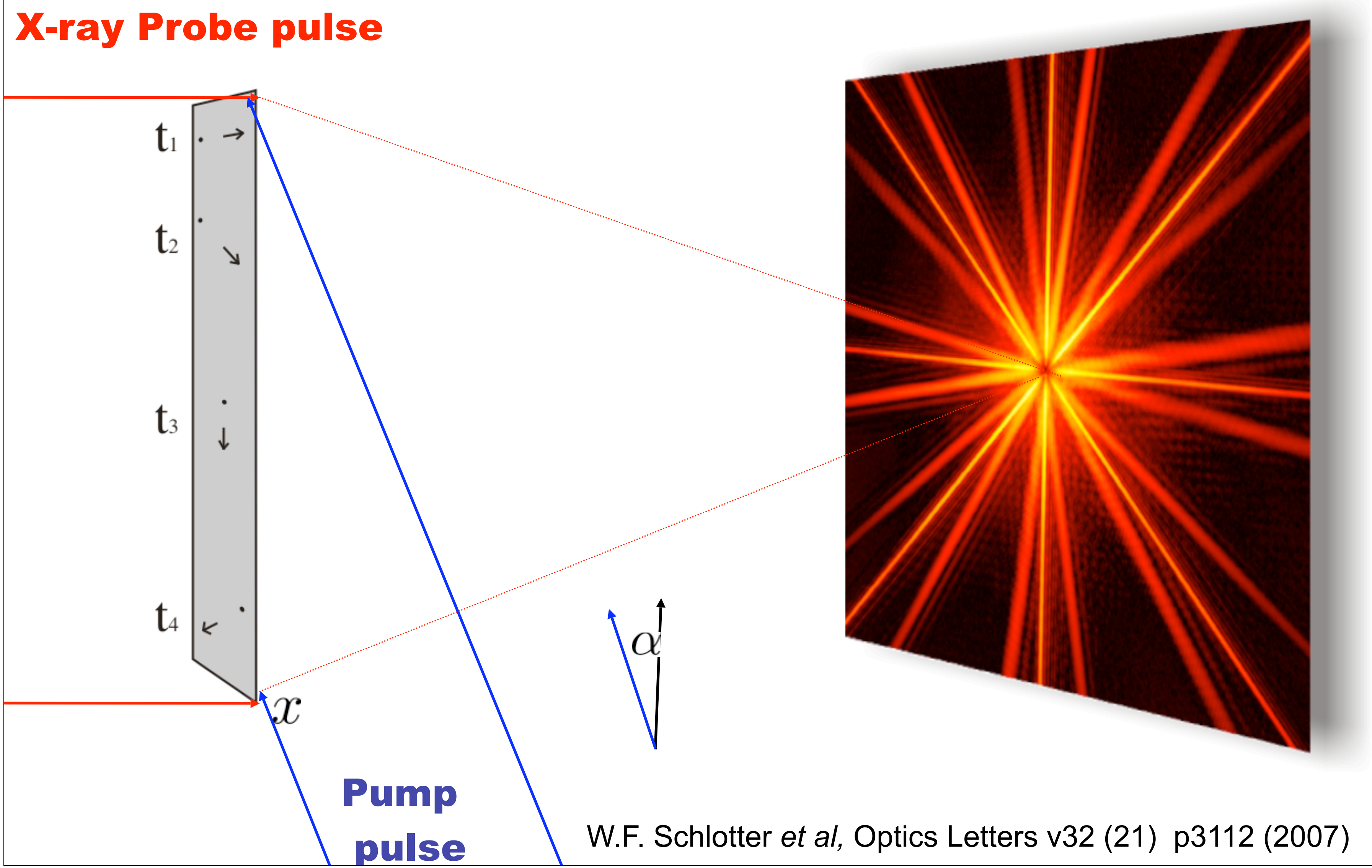


(a) -5ps (b) 10ps (c) 15ps (d) 20ps (e) 40ps (f) 140ps

$$G(q) = \frac{I_1'(q) \bullet I_2(q)}{\|I_1'(q)\| \|I_2'(q)\|}$$

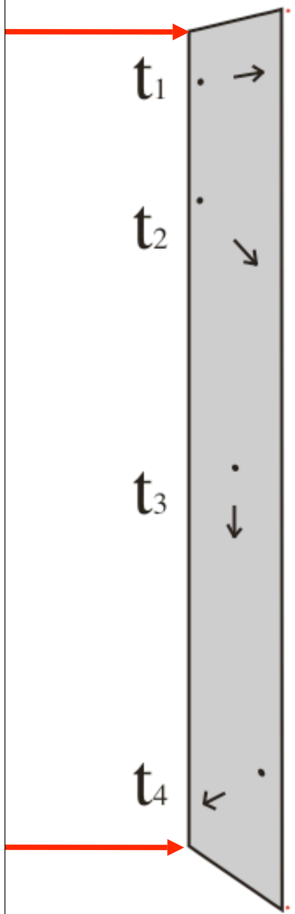
$$I_n' = I_n(q) - < I_n(q) >$$

Ultrafast Stopwatch

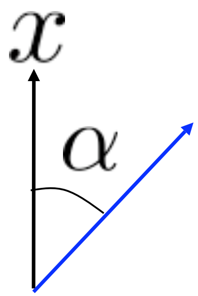


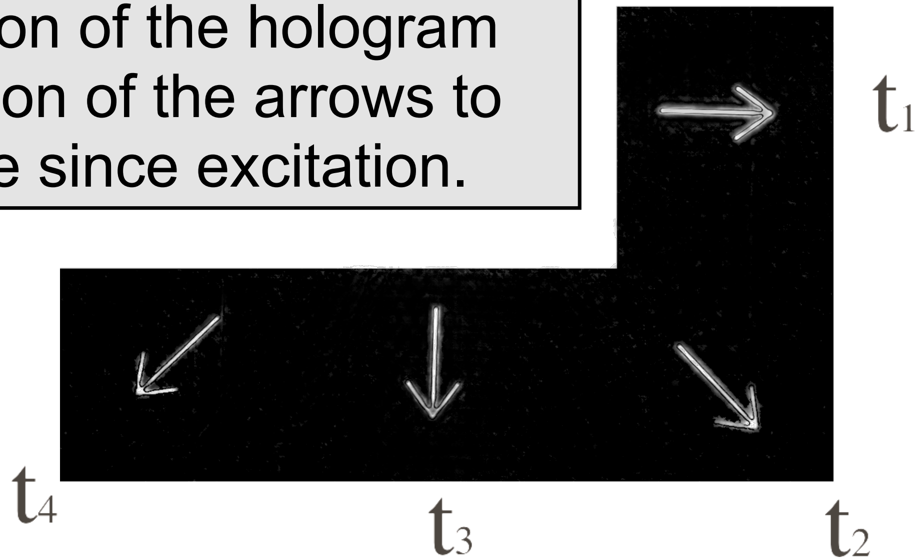
W.F. Schlotter *et al,* Optics Letters v32 (21) p3112 (2007)

Single Shot Stopwatch



The reconstruction of the hologram relates the position of the arrows to their relative time since excitation.





$$t(x) = x\frac{\cos(\alpha)}{c}$$

$$\boxed{c = 0.3\ \mu\text{m/fs}}$$

x	α	$t(x)$
180 μm	33°	500 fs
50 nm	33°	140 as

W.F. Schlotter *et al,* Optics Letters v32 (21) p3112 (2007)

Acknowledgments

LLNL:	Saša Bajt, Anton Barty, Henry Benner, Michael Bogan, Matthias Frank, Stefan Hau-Riege, Richard Lee, Richard London, Urs Rohner, Eberhard Spiller, Abraham Szöke, Bruce Woods
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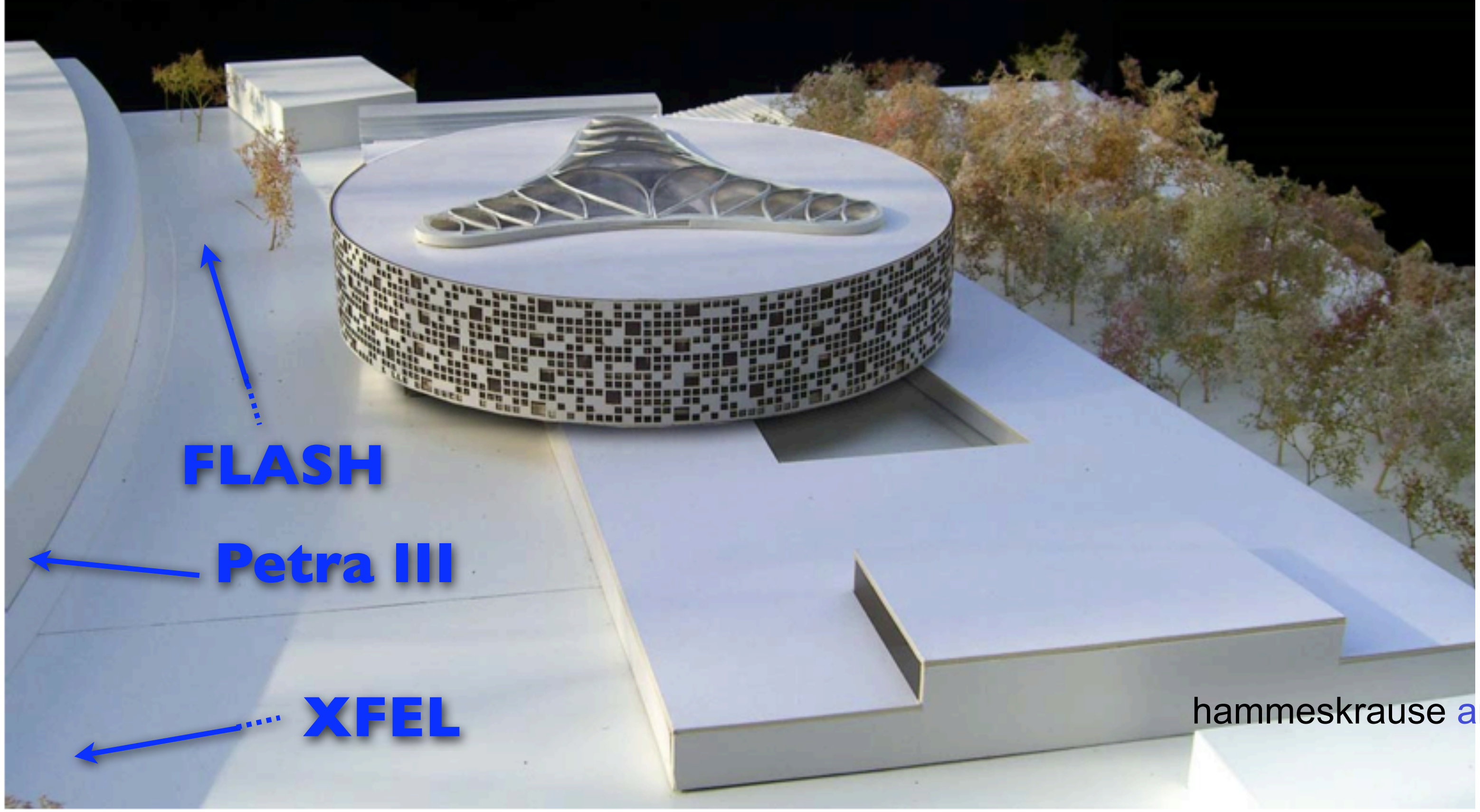
SLAC:	Sebastien Boutet
LBNL:	Stefano Marchesini, David Shapiro
DESY:	Saša Bajt, Joachim Schulz, Thomas Tschentscher, Elke Plönjes, Marion Kulhman, Rolf Treusch, Stefan Dusterer, Jochen Schneider
TU Berlin:	Thomas Möller, Christof Bostedt
Essen:	Klaus Sokolowski-Tinten
Oxford:	Andrea Cavalleri
ASU:	John Spence, Uwe Weierstall, Bruce Doak

Center for Free-Electron Laser Science (CFEL)

Coherent Imaging & Scattering	*Henry Chapman*
Structural Dynamics	*Andrea Cavalleri*
3 more groups to come	
Max Planck Advanced Study Group	*Joachim Ullrich*
U Hamburg Advanced Study Group	*Wilfried Wurth*



hammeskrause architekten