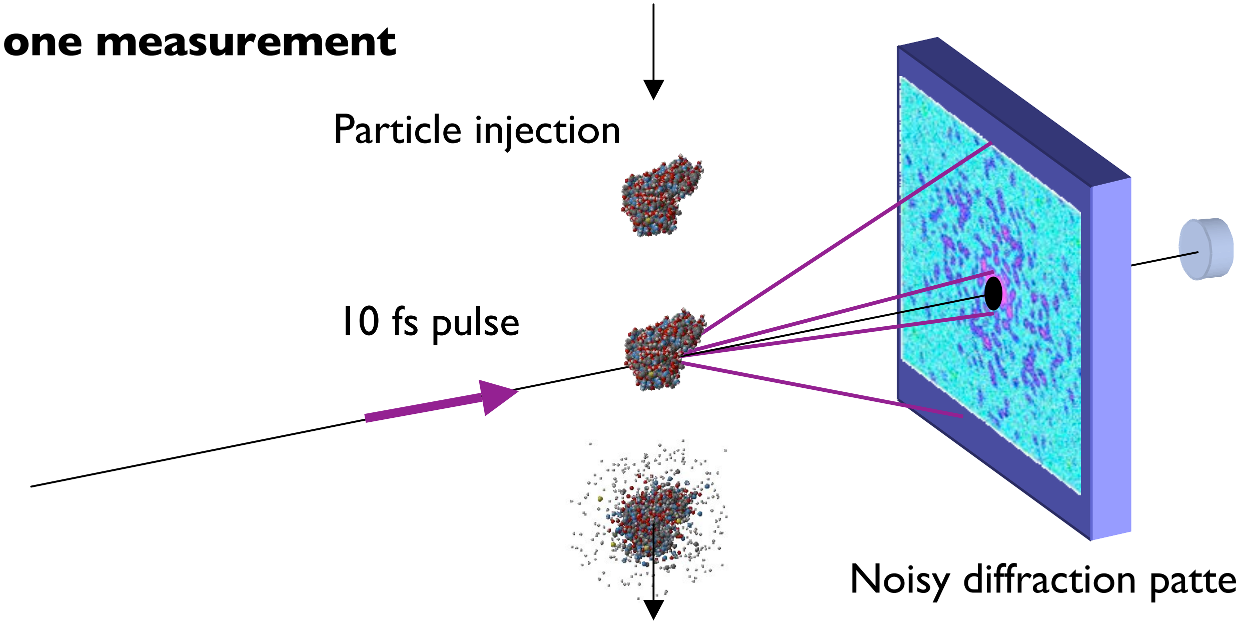
Imaging Update to LCLS SAC, June 2006

Henry Chapman, LLNL

LLNL:	Sasa Bajt, Anton Barty, Henry Benner, Micheal Bogan, Henry Chapman, Matthias Frank, Stefan Hau-Riege, Richard Lee, Richard London, Stefano Marchesini, Tom McCarville, Alex Noy, Urs Rohner, Eberhard Spiller, Abraham Szöke, Bruce Woods
Uppsala:	Janos Hajdu, Gösta Huldt, Carl Caleman, Magnus Bergh, Nicusor Timeneau, David van der Spoel, Florian Burmeister, Marvin Seibert
UC Davis:	David Shapiro
SLAC:	Keith Hodgson, Sebastien Boutet
DESY:	Thomas Tschentscher, Elke Plönjes, Marion Kulhman, Rolf Treusch, Stefan Dusterer, Jochen Schneider
TU Berlin:	Thomas Möller, Christof Bostedt
LBNL:	Malcolm Howells, Congwu Cui
ASU:	John Spence, Uwe Weierstall, Bruce Doak

Single particle diffraction offers many challenges

One pulse, one measurement

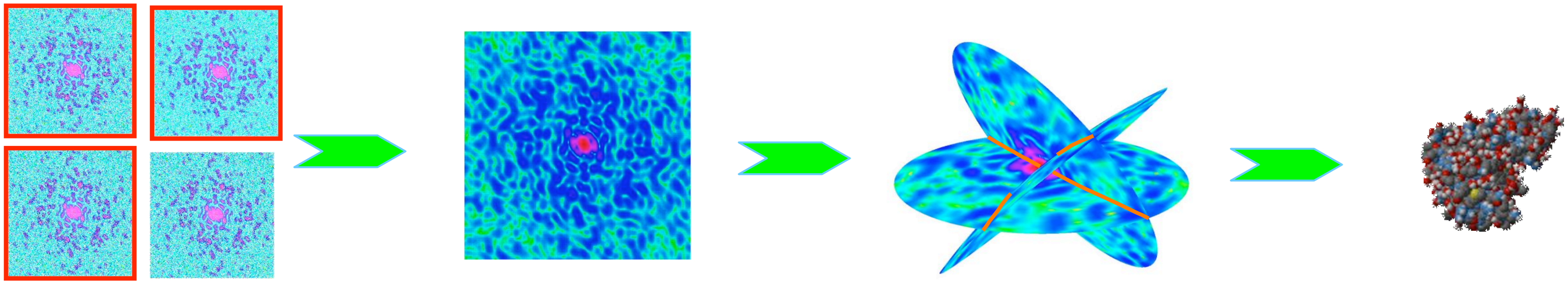


Particle injection

10 fs pulse

Noisy diffraction pattern

Combine 10^5-10^7 measurements



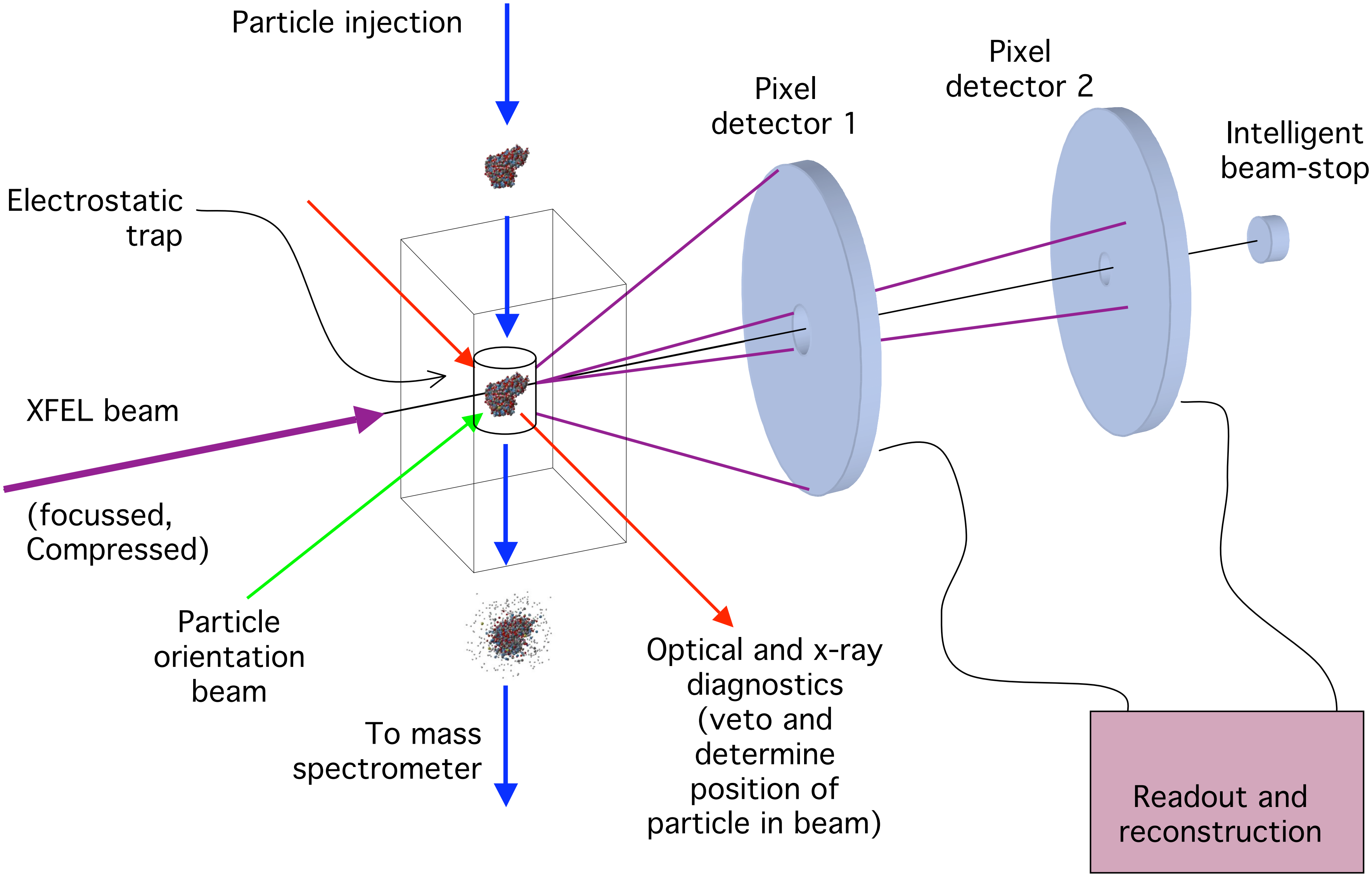
Classification

Averaging

Orientation

Reconstruction

The diffraction imaging interaction chamber and detector arrangement

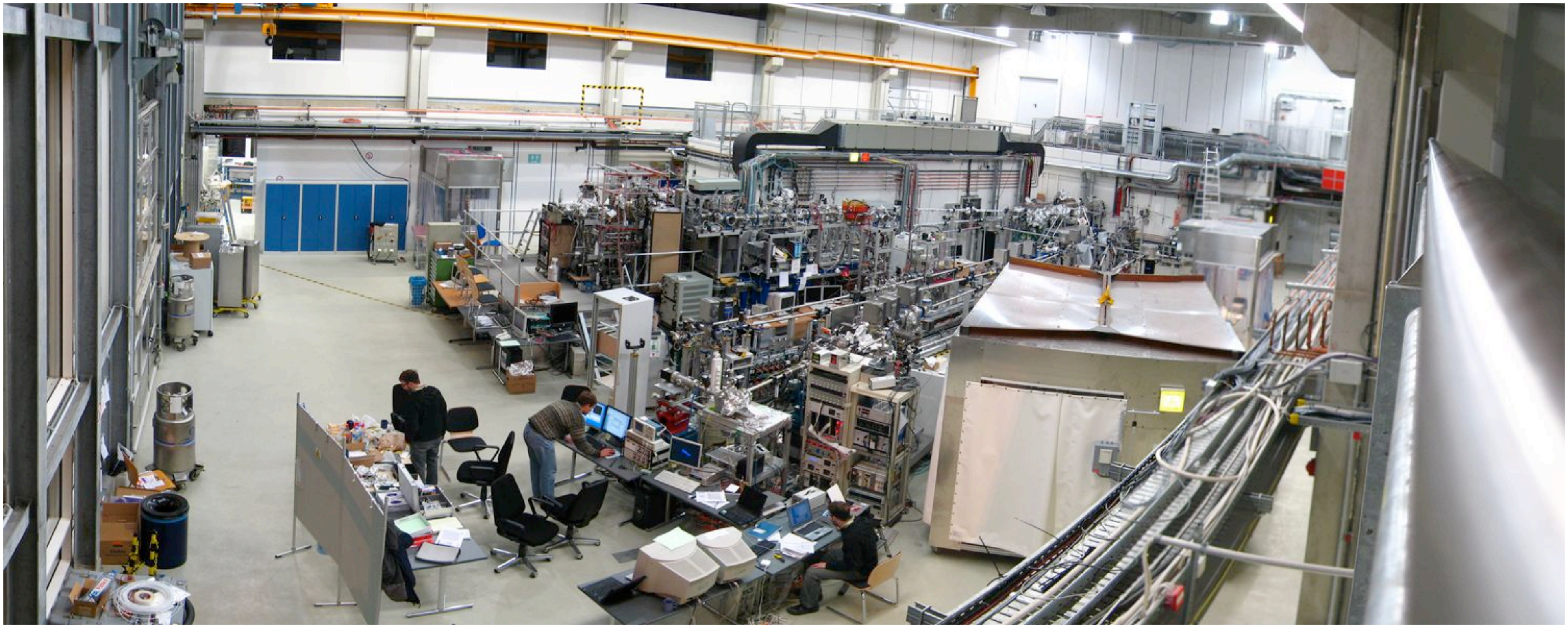


We are developing the first experiments for LCLS at FLASH

The FLASH experiments are essentially a dry-run for LCLS

- Diffraction imaging with a single pulse, to surpass radiation damage limits
- Determine if there is any change in structure of particles during the pulse
- Measure the dynamics of the FEL-particle interaction to validate models
- Develop and demonstrate particle injection and alignment techniques
- Develop optics and instrumentation

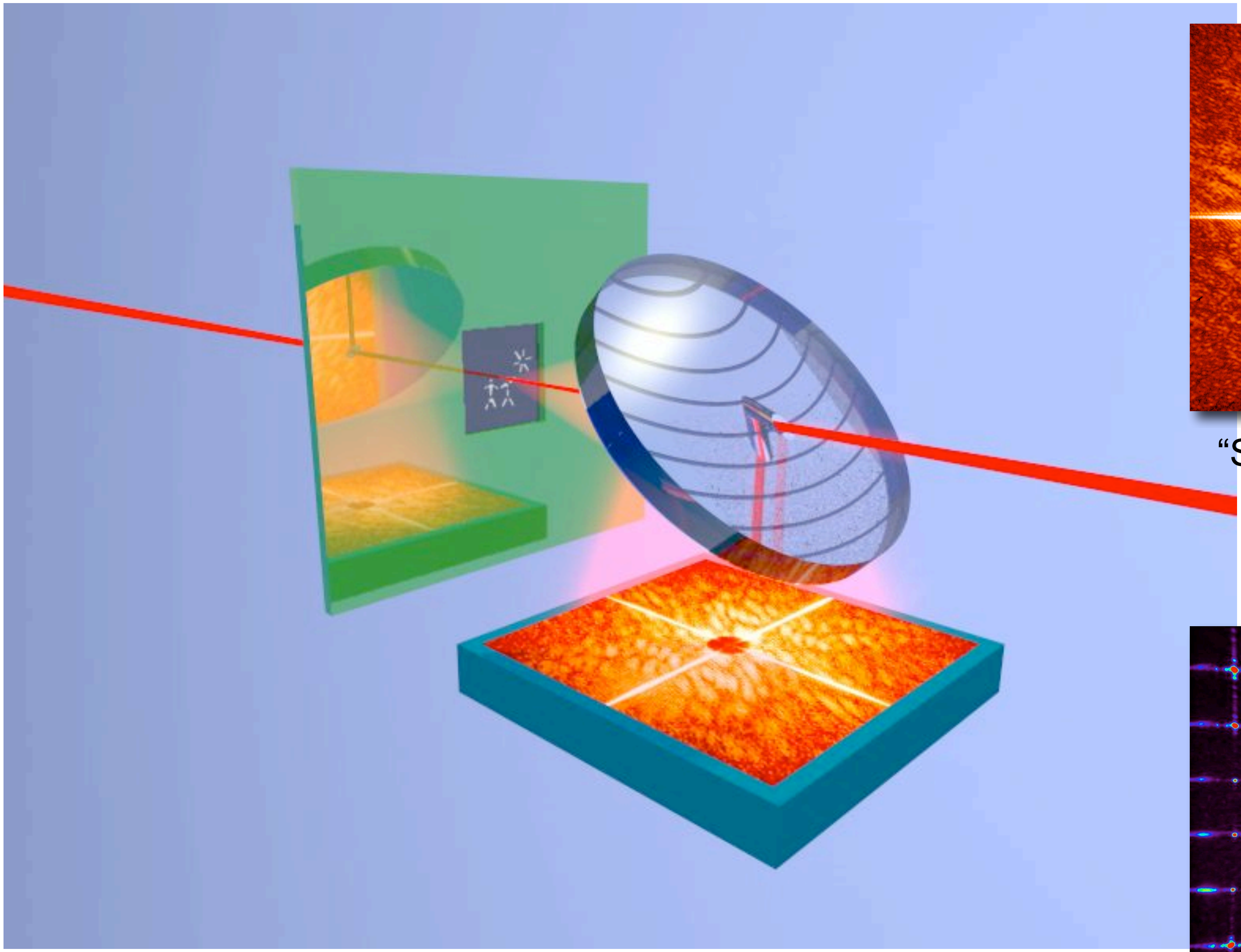
We have carried out experiments at the first soft-X-ray FEL in the world

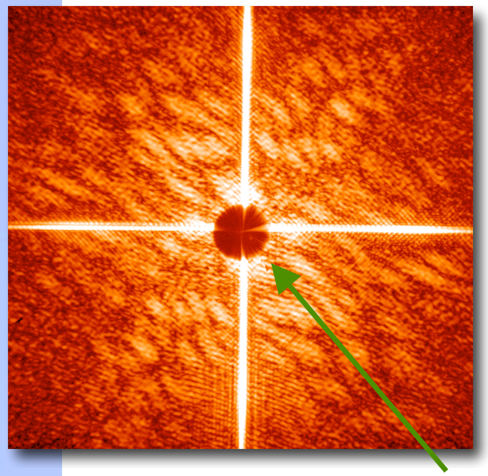


The VUV-FEL at HASYLAB, DESY

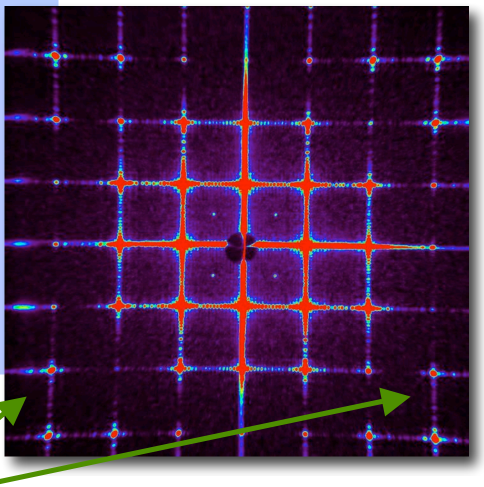
- User facility, FEL radiation to 6 nm wavelength
- Initial FEL Operation August 2005 at 32 nm and <30 fs pulses, 10^{13} photons
- Now lasing at 13 nm

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



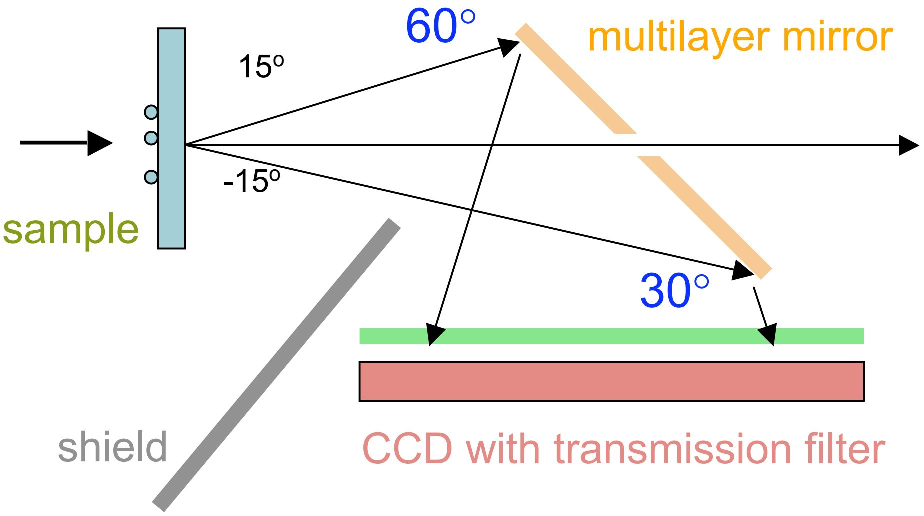


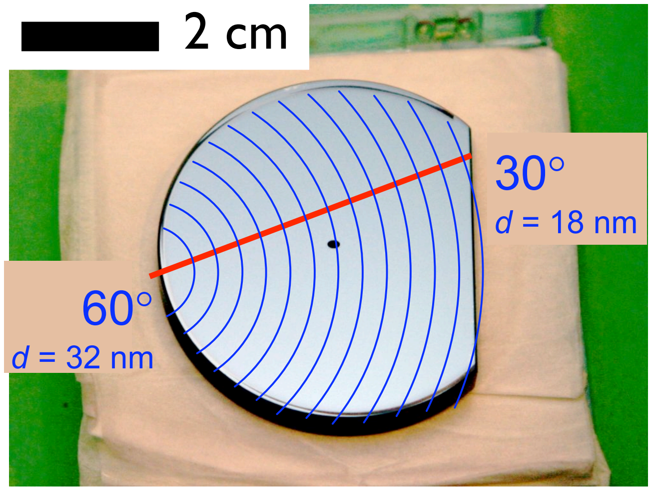
“Soft edge” prevents any scatter from the hole

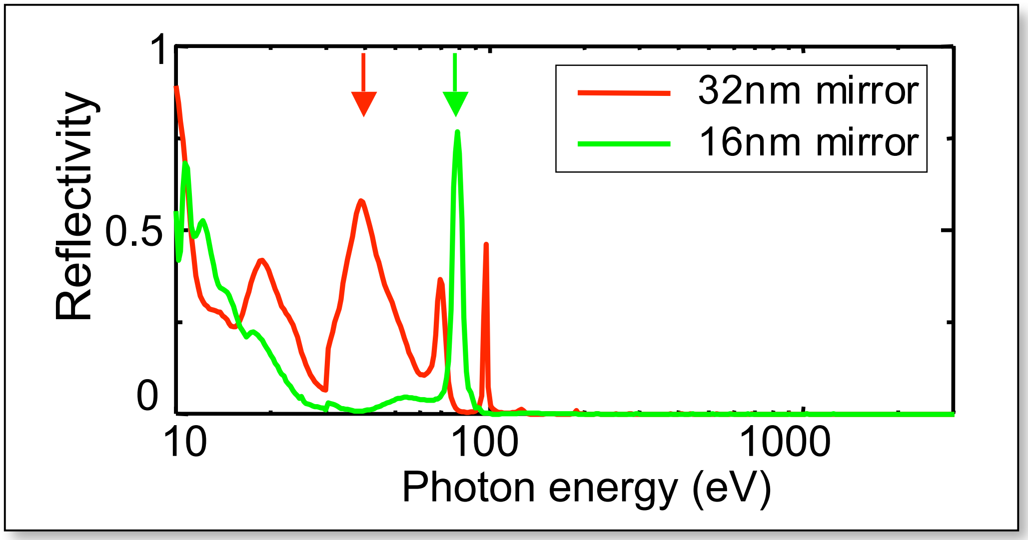


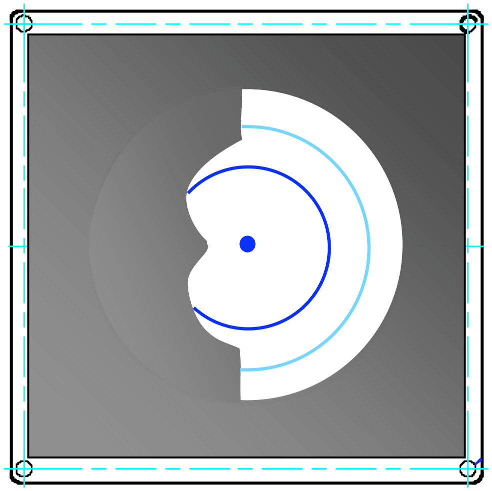
Multilayer reflectivity is uniform across the 30° to 60° gradient

The VUV-FEL diffraction experiment is designed to measure forward scattering with high SNR





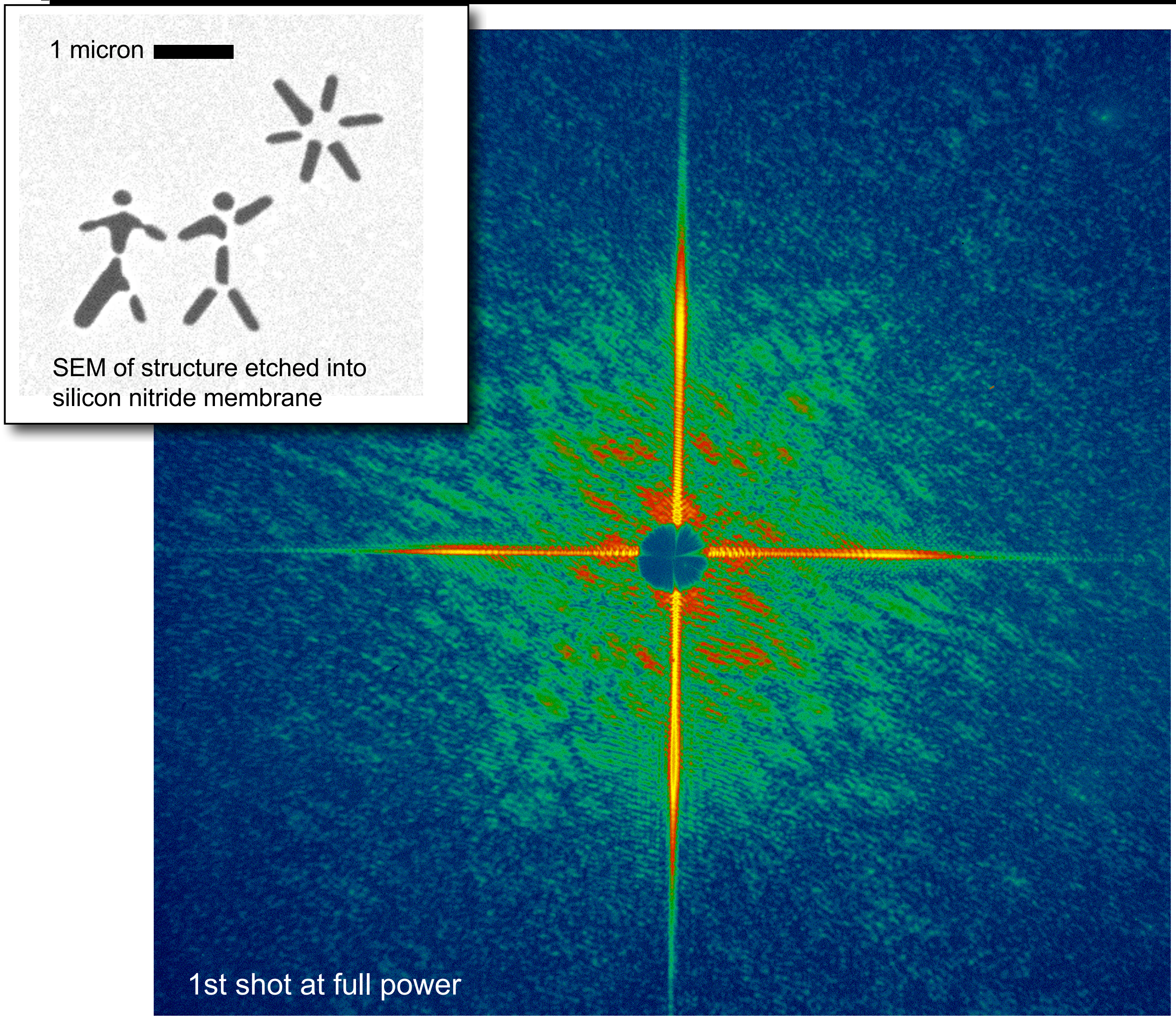




Shadow mask

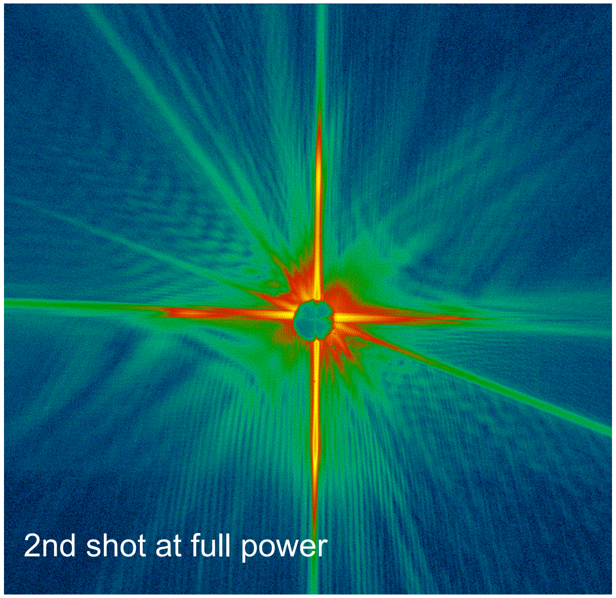
Sasa Bajt, Eberhard Spiller, and Jennifer Alameda

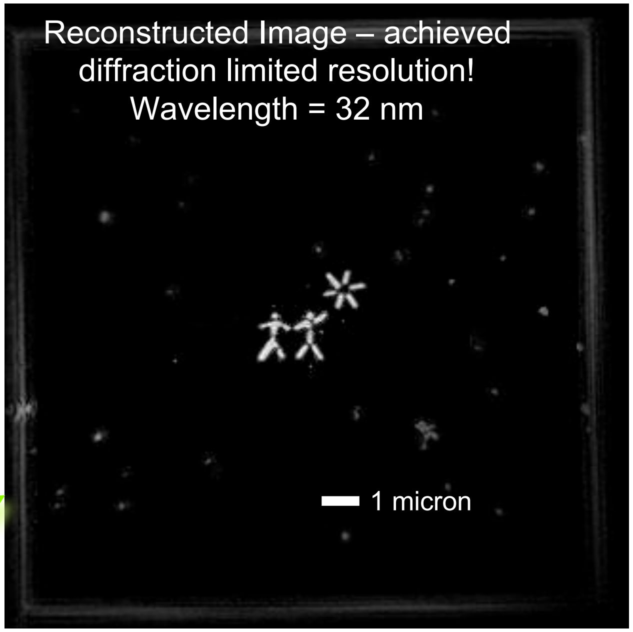
Image reconstructed from an ultrafast FEL diffraction pattern



SEM of structure etched into silicon nitride membrane

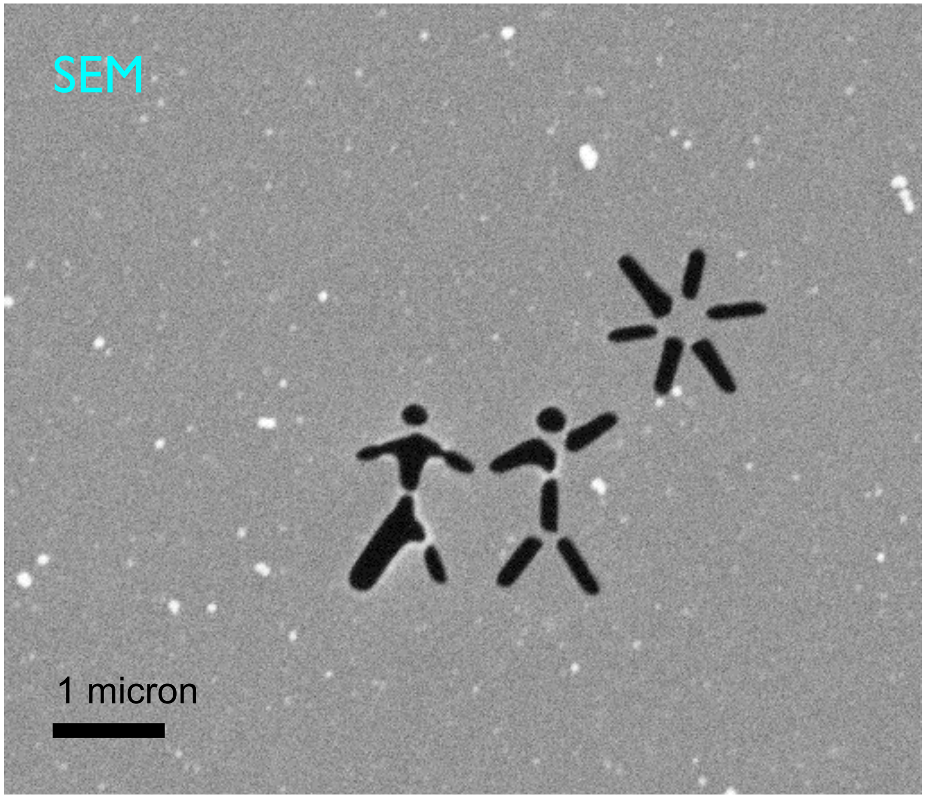
1st shot at full power

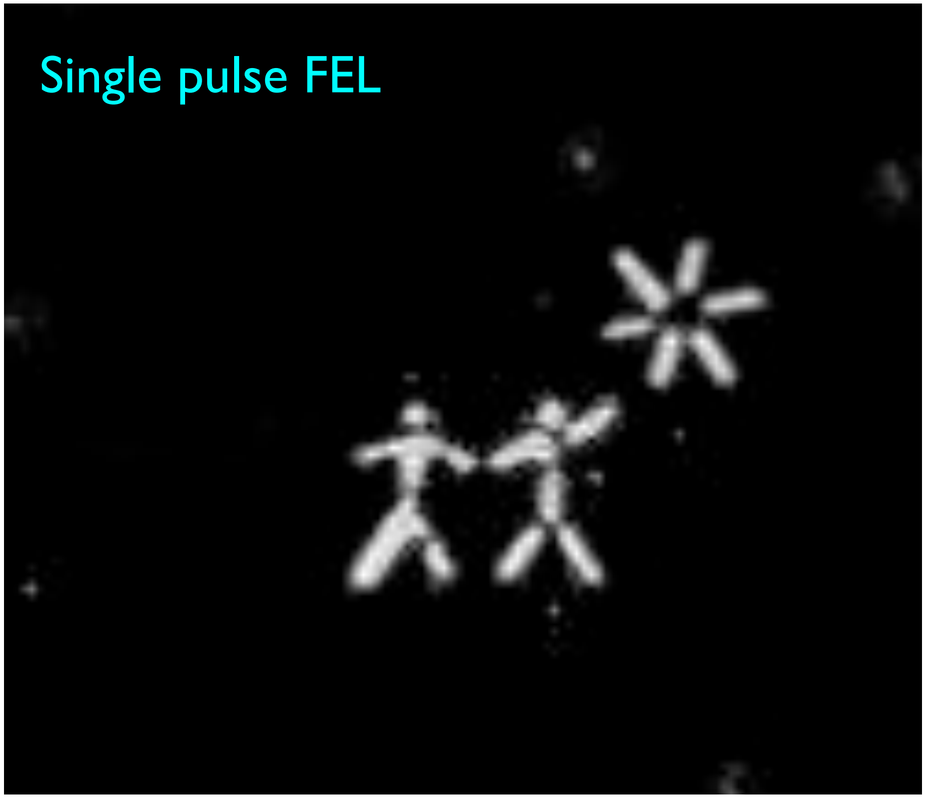

2nd shot at full power


Reconstructed Image – achieved diffraction limited resolution!
Wavelength = 32 nm

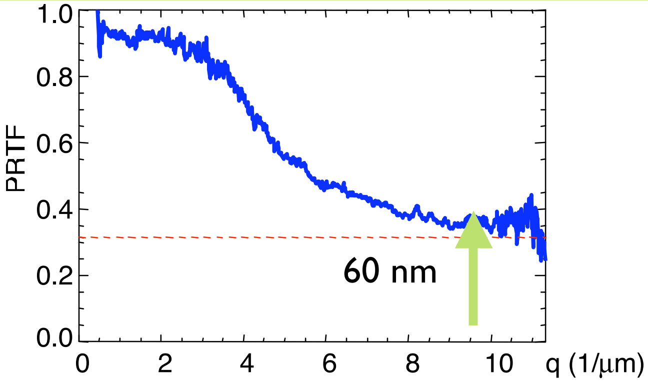
Edge of membrane support also reconstructed

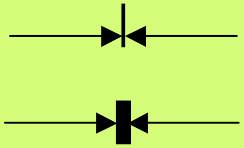
The reconstruction is carried out to the diffraction limit of the 0.26 NA detector





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

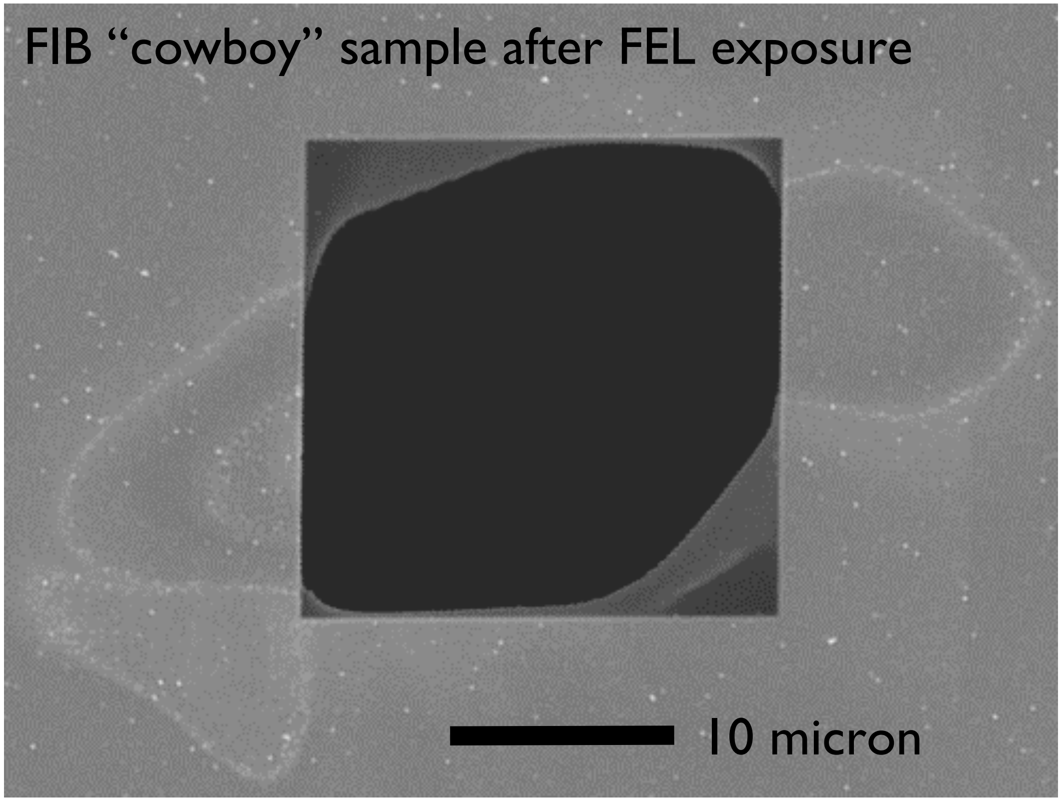




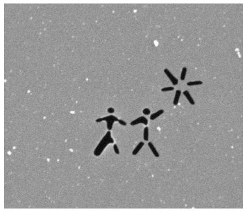
32 nm, one wavelength

λ / NA

The sample is quite damaged by the FEL pulses



before



Pulse energy: 10 μJ or 1 J/cm²

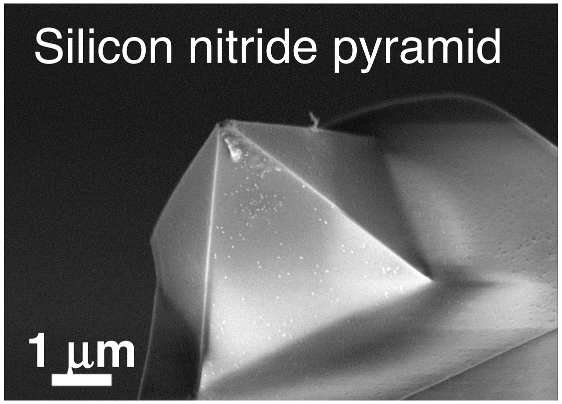
Dose in Si_3N_4: 10^5 J/g or 22 eV/atom

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

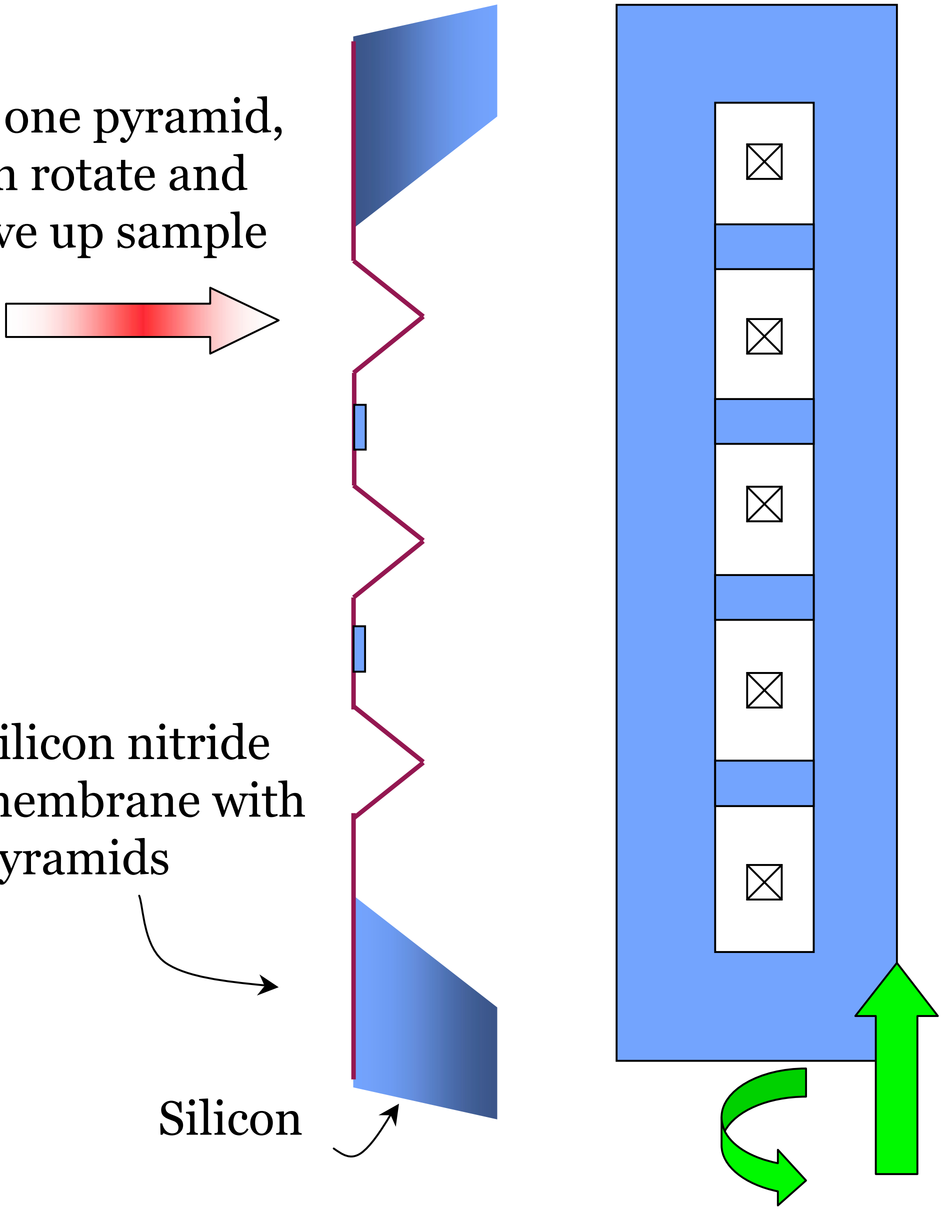
In 25 fs, material will not move more than 3 Å

We will perform 3D imaging of identical objects at FLASH and high-resolution imaging of cells



High-resolution imaging will be carried out at the third harmonic, e.g. a wavelength of 4.5 nm.

Requires 2 µm focus

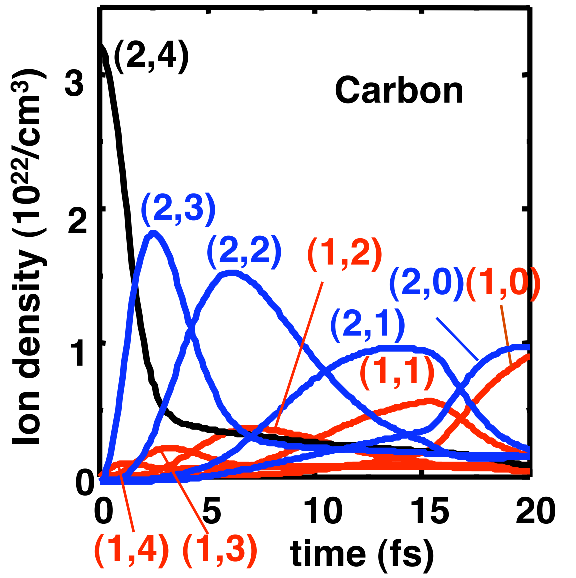


Hit one pyramid, then rotate and move up sample

Silicon nitride membrane with pyramids

Silicon

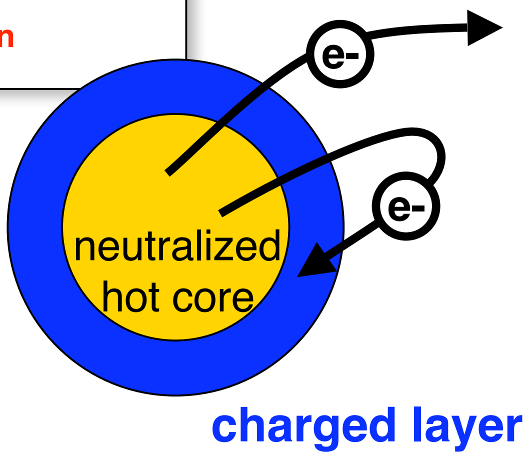
XFEL diffraction of molecules is modified (damaged) by photoionization and motion of atoms



(k,l) =(# K-shell, # L-shell) electrons
black = neutral carbon
blue = valence ionization
red = inner shell ionization

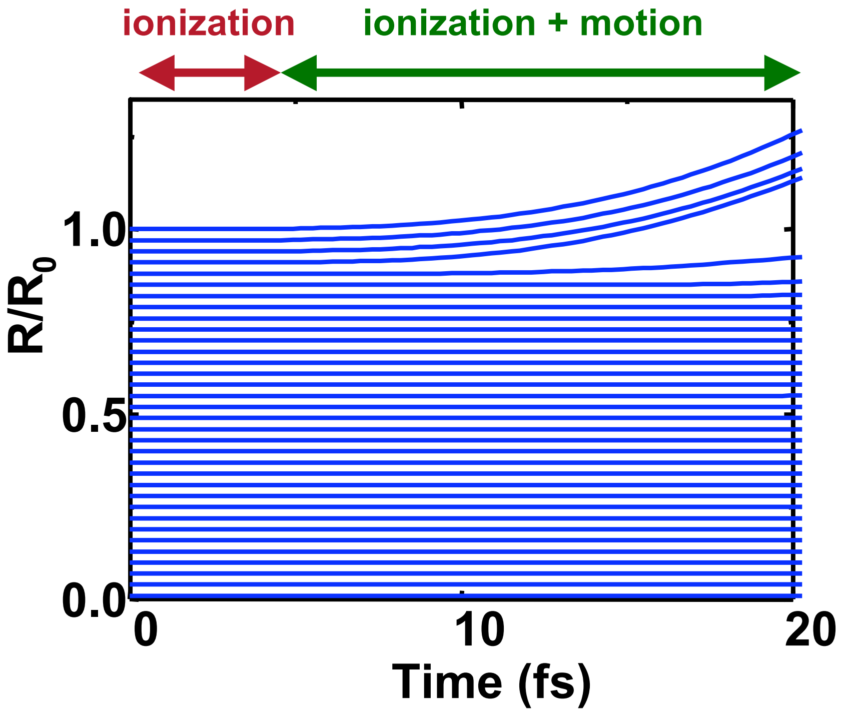
We have developed a *hydrodynamic continuum model* for the atomic motion and the ionization processes that can treat large and small molecules

- 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



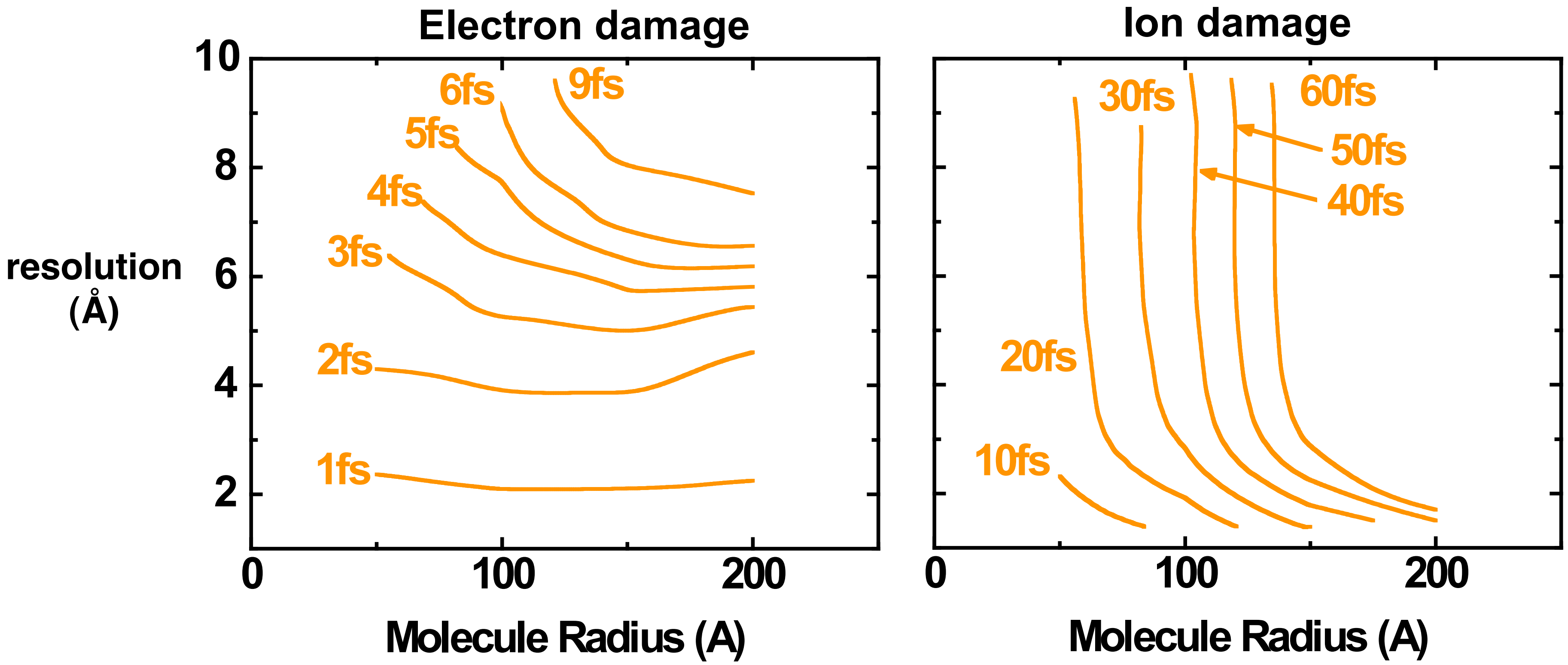
electron escapes if

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



Stefan Hau-Riege, Richard London, Abraham Szoke

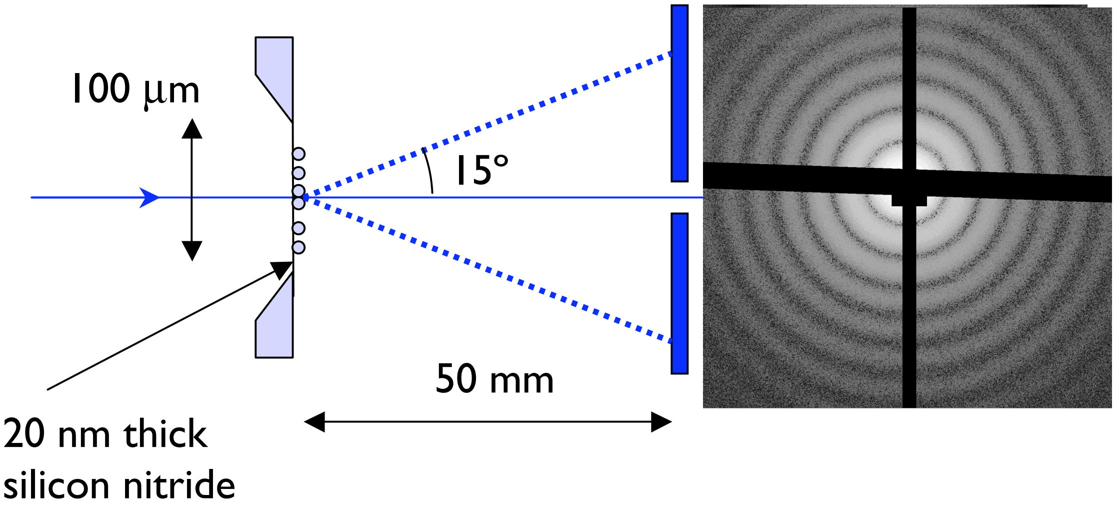
By repairing ionization damage, the imaging is limited by the inertia of the atoms



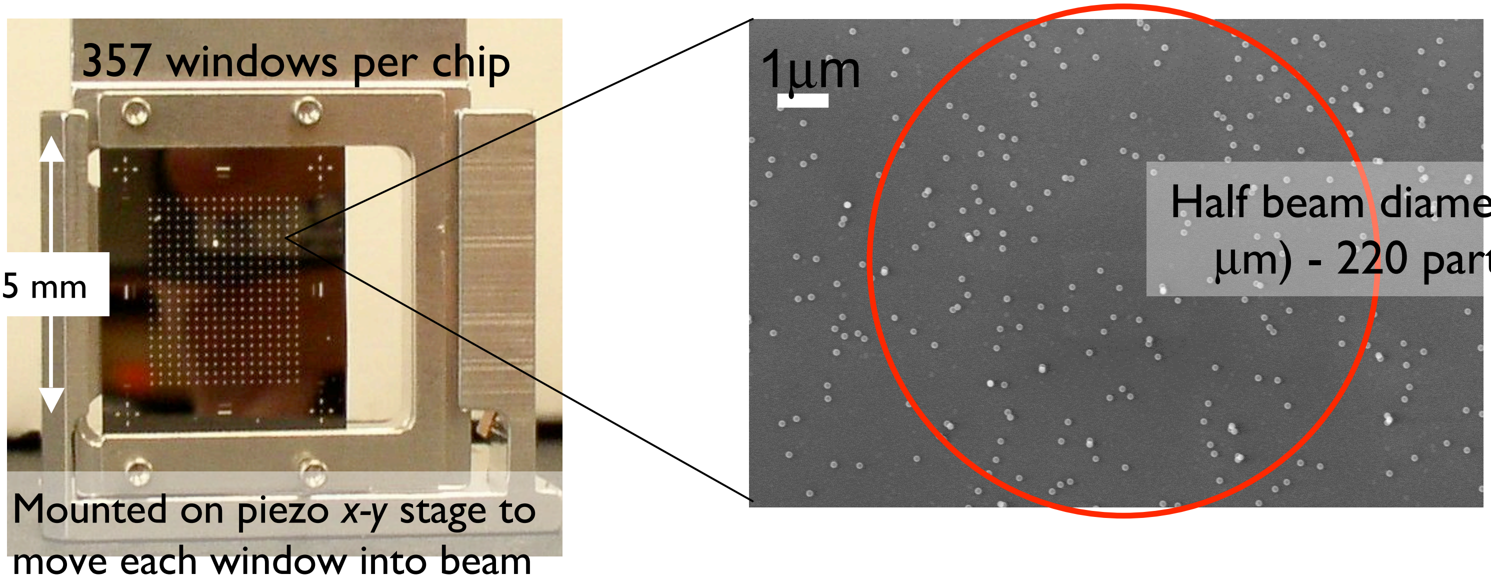
Even longer pulses can be tolerated if a tamper is used, but image classification will be more difficult

(Hau-Riege et al., PRE 2005)

Particle explosion experiments were performed on latex particles on membranes



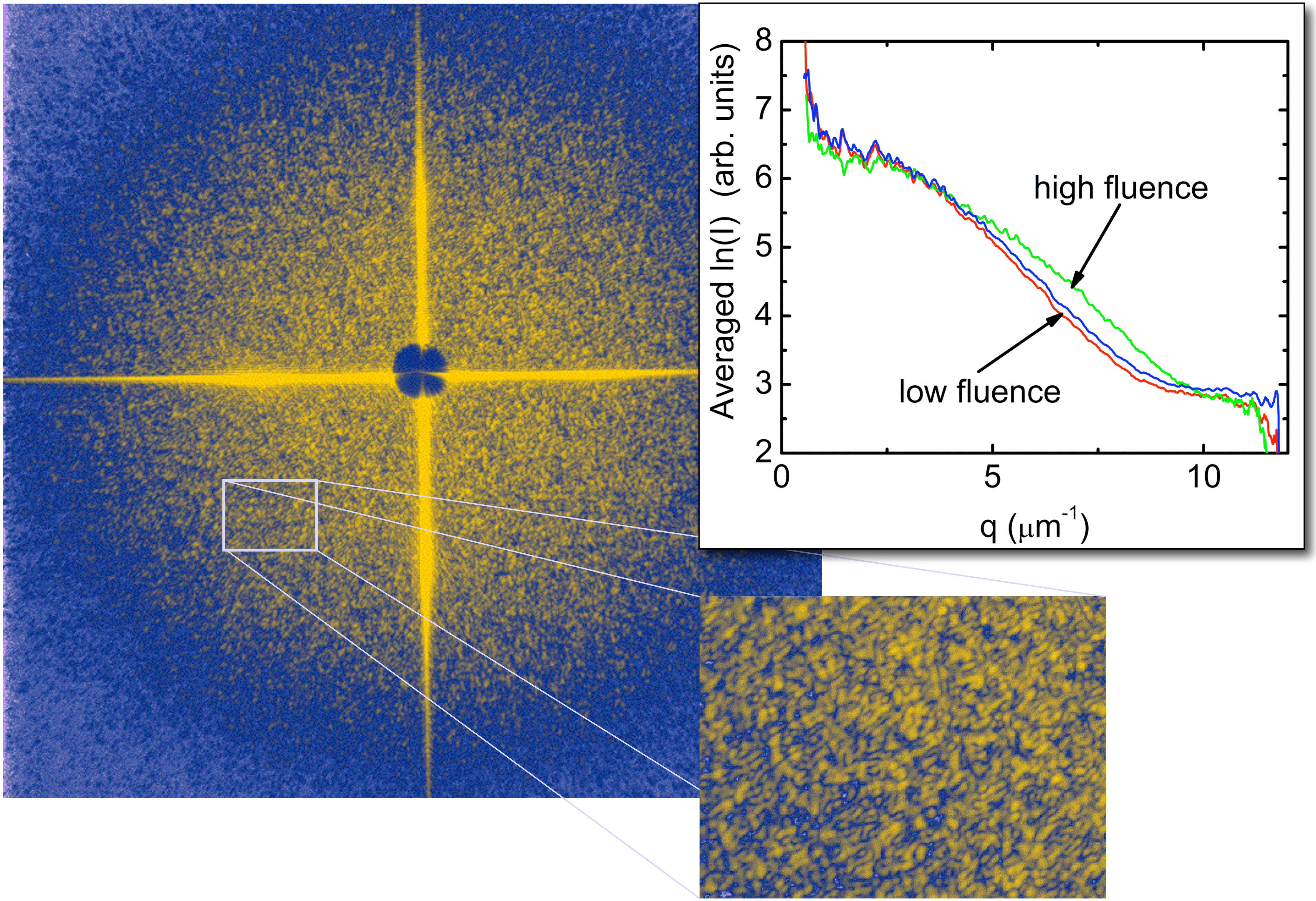
- The particle size is determined by Mie scattering of the VUV-FEL pulse by the particles (FEL pulse is both pump and probe)
- To see a 5% change in radius during pulse, require size distribution of ~1%.



Mounted on piezo *x-y* stage to move each window into beam

Half beam diameter (10 µm) - 220 particles

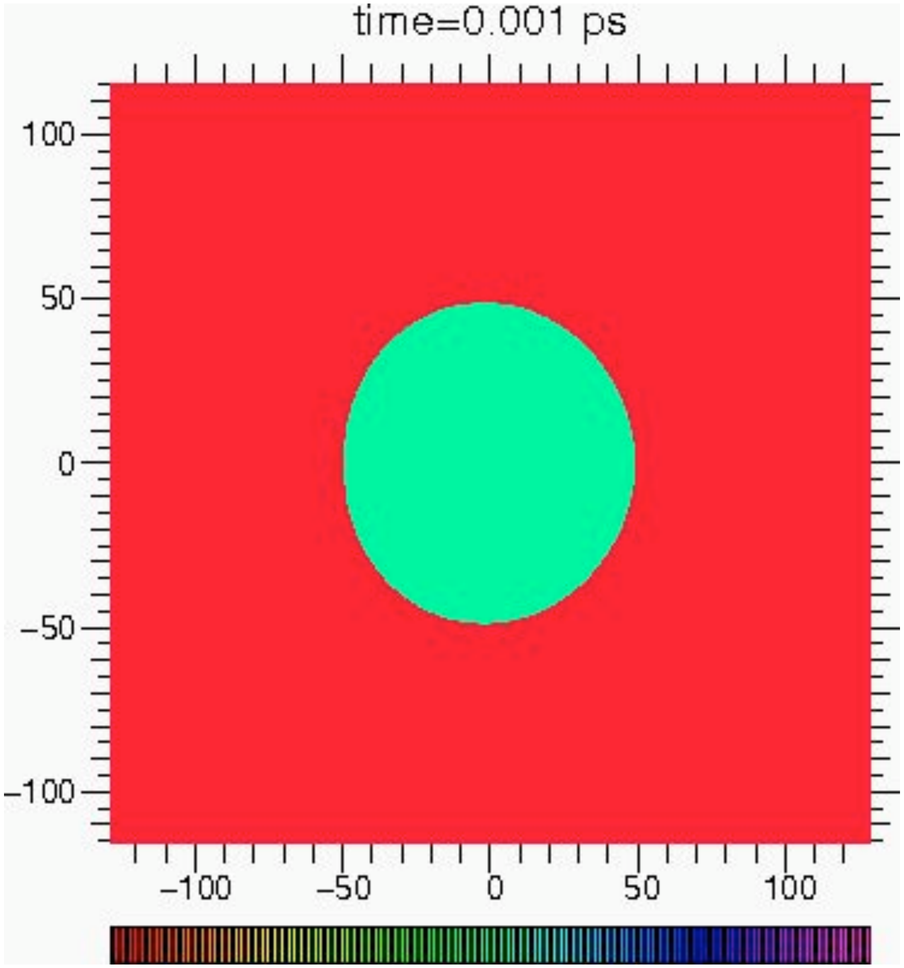
Scattering from balls demonstrates that they retain their shape throughout the duration of the pulse



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

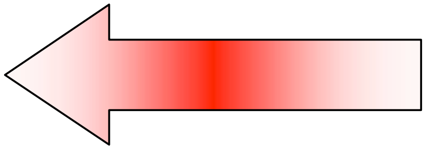
Density

y (nm)



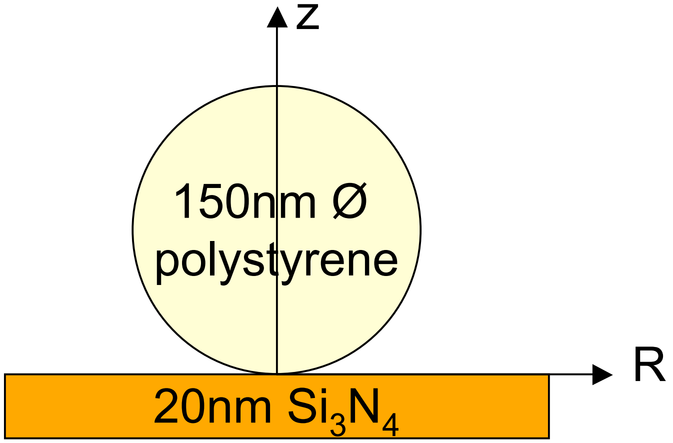
z (nm)

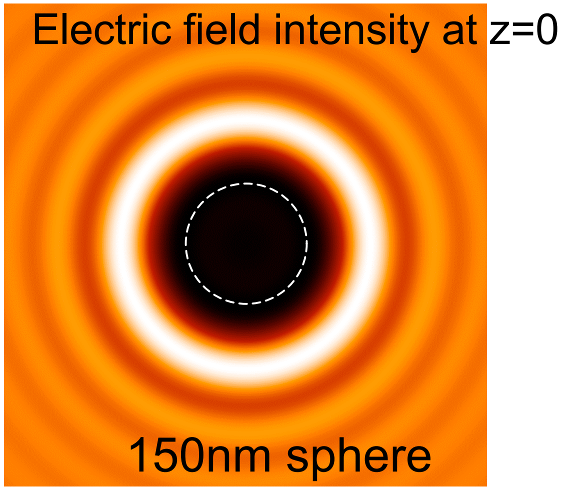
Incident pulse direction

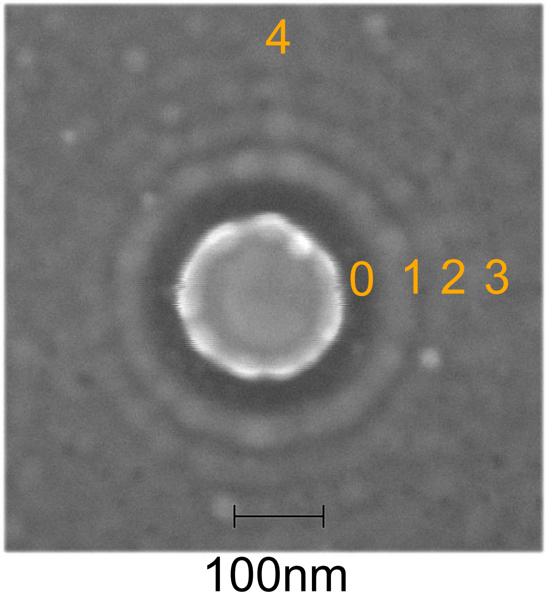


Color scale: 0 g/cm^3 2 g/cm^3

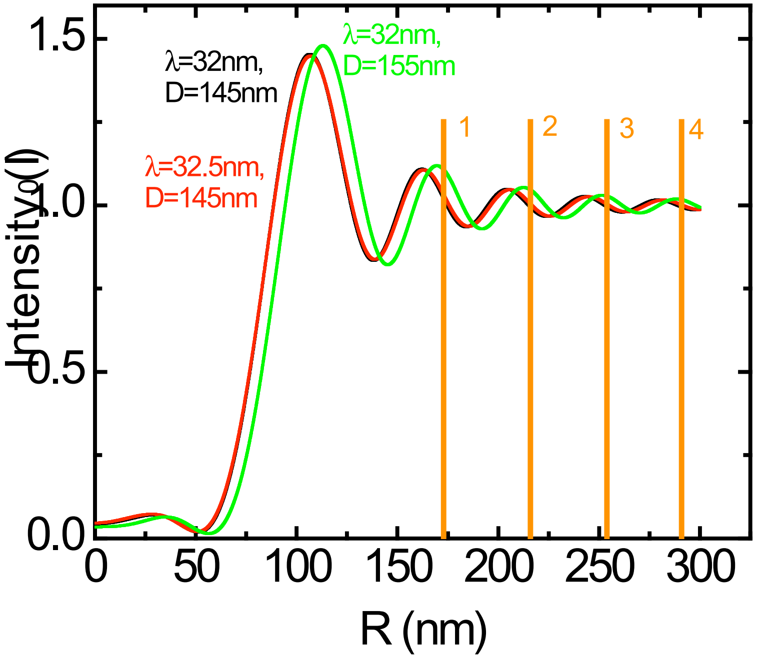
The substrate is a high-resolution detector (at low enough fluence)



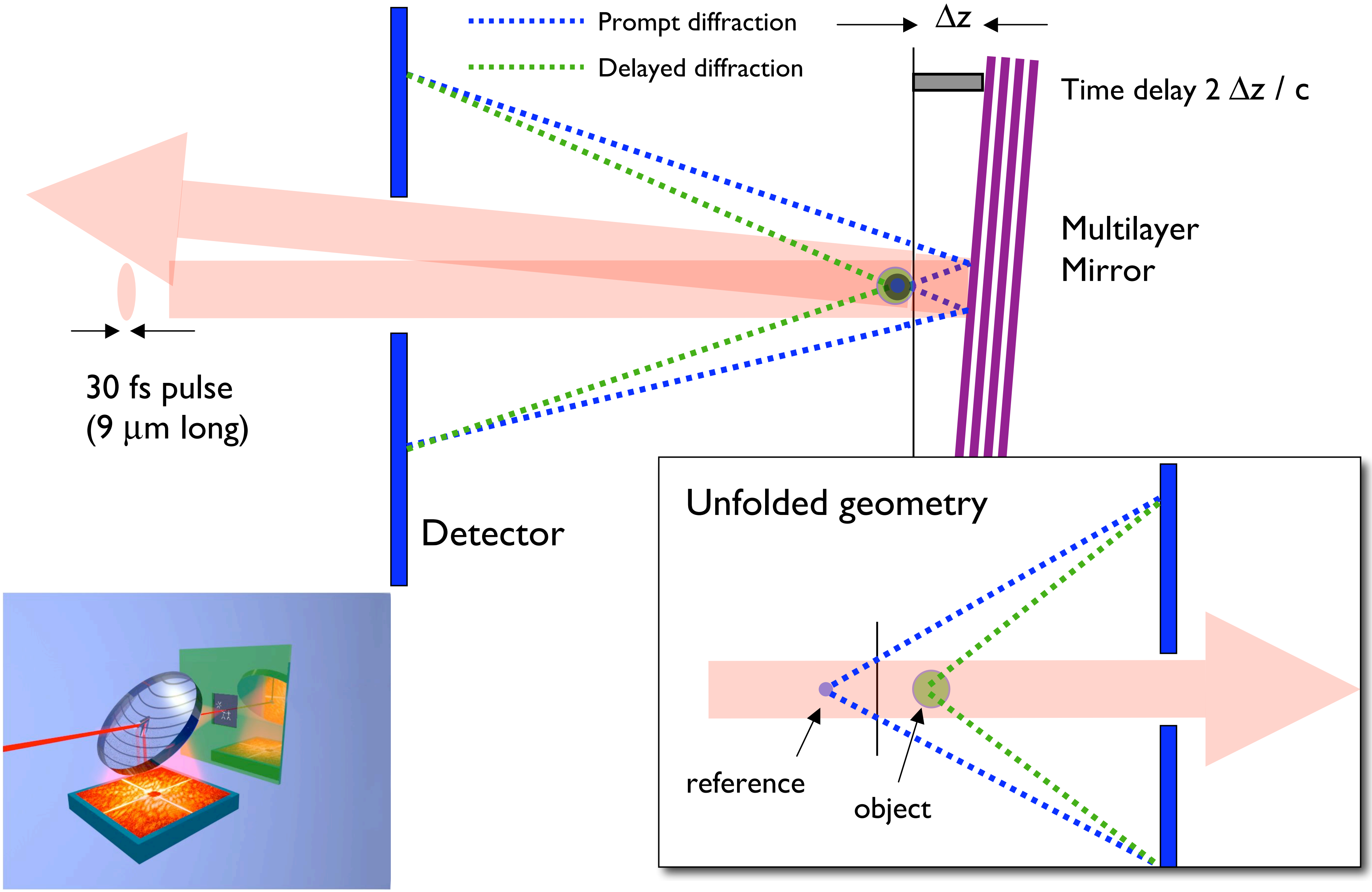




0,1,2,3,4: depression rings



We invented a new method called femtosecond time-delay holography



Prompt diffraction

Delayed diffraction

Δz

Time delay $2\,\Delta z / c$

Multilayer Mirror

30 fs pulse (9 µm long)

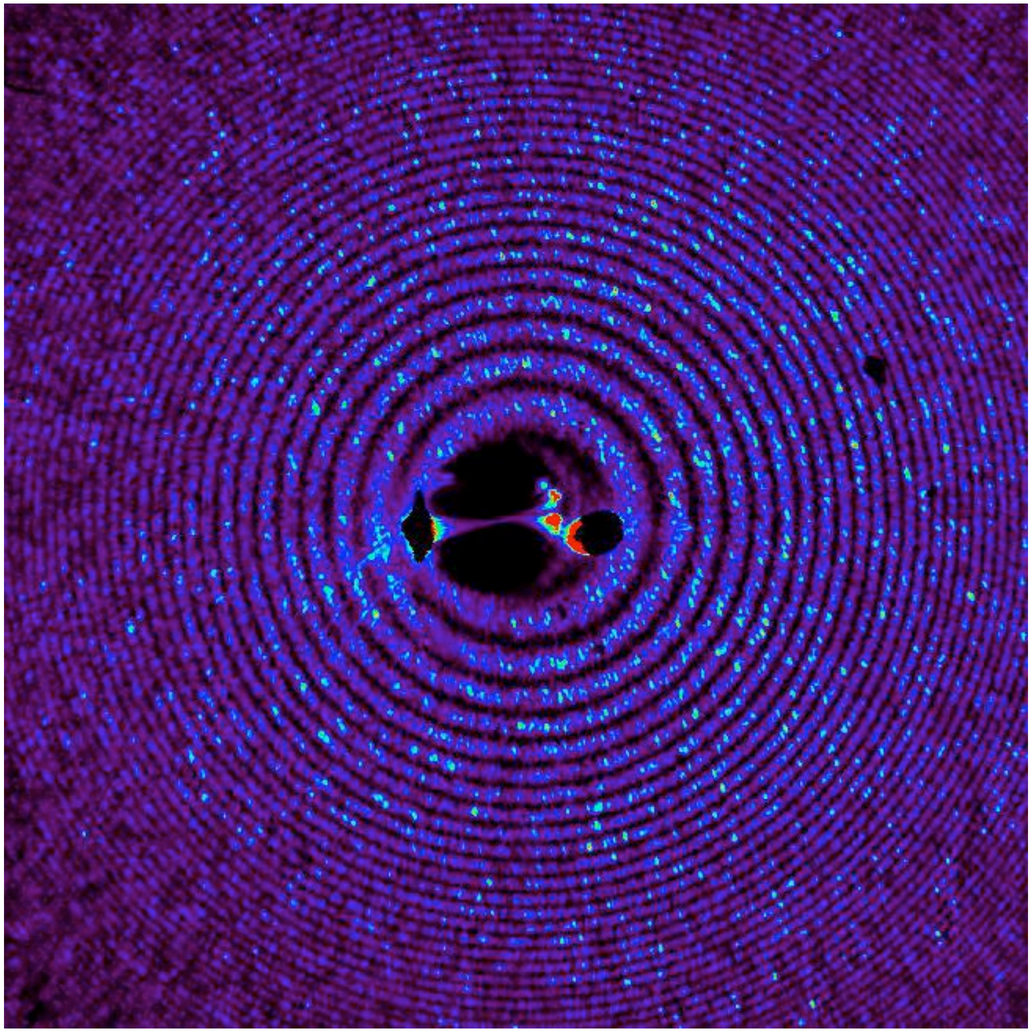
Detector

Unfolded geometry

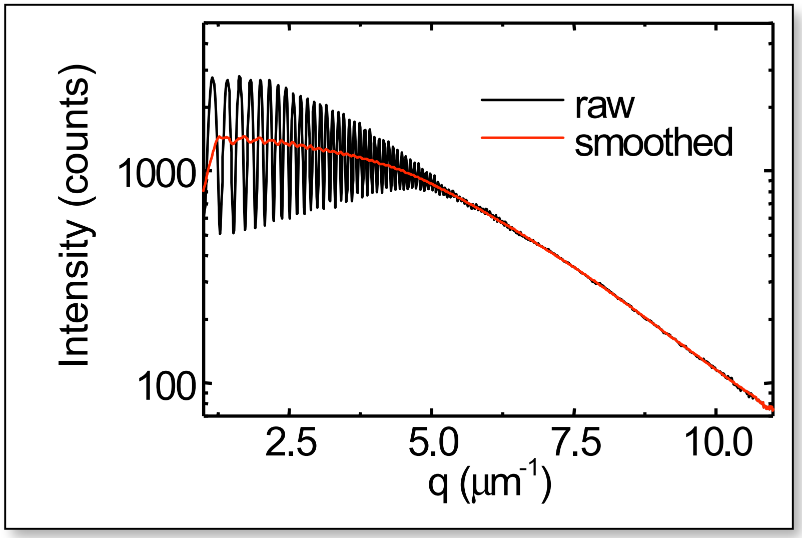
reference

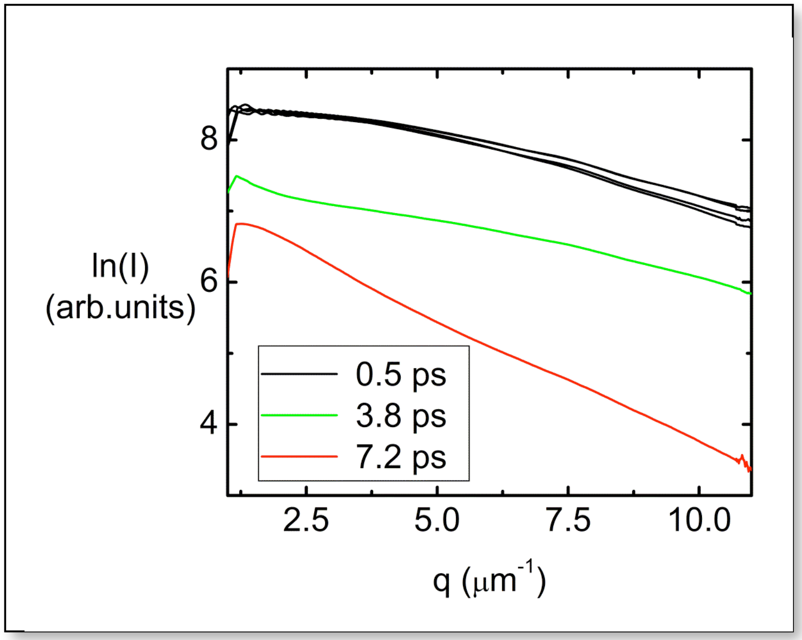
object

First demonstration of time-delay holography with 30 fs time resolution indicates the particle explosion



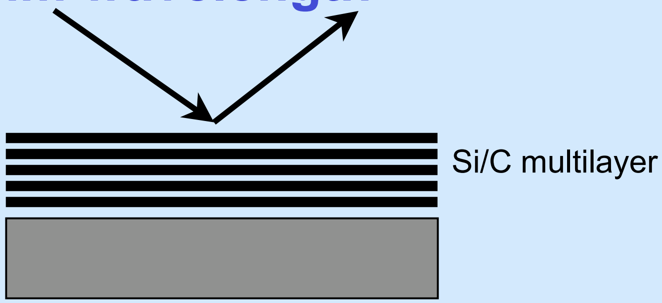
Single shot ultrafast time-delay X-ray hologram, with 300 fs delay



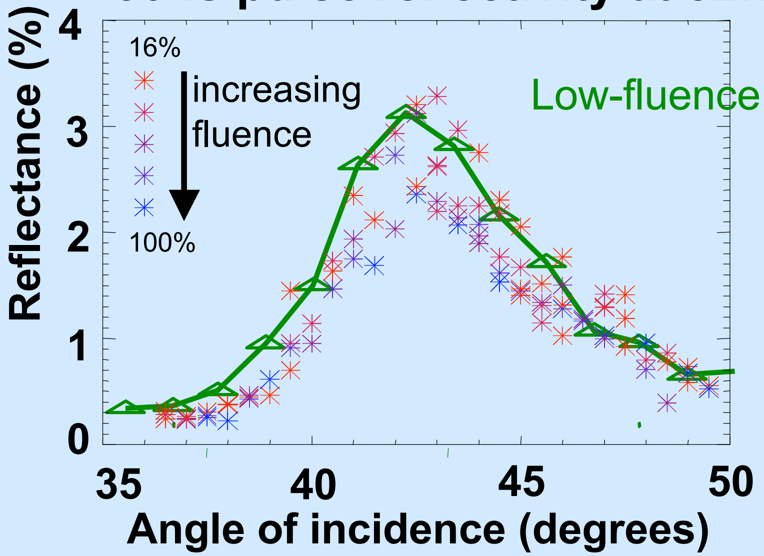


First EUV-FEL experiments show that structural information can be obtained before destruction

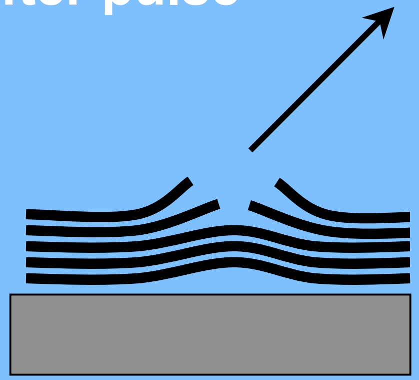
During 30 fs pulse (10^{14} W cm^{-2})
32 nm wavelength



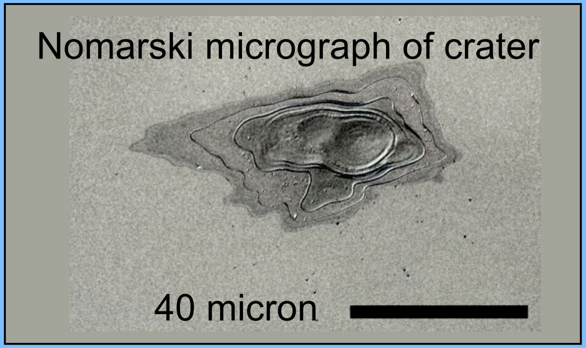
Reflectivity unchanged
Multilayer *d* spacing not changed by more than 0.3 nm

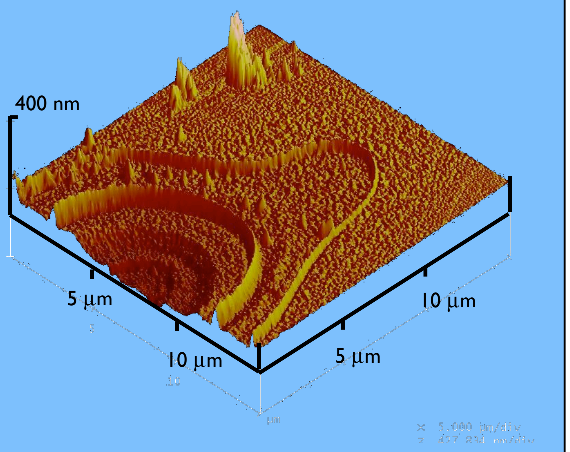

30 fs pulse reflectivity at 32nm

After pulse



Plasma forms, layers ablate

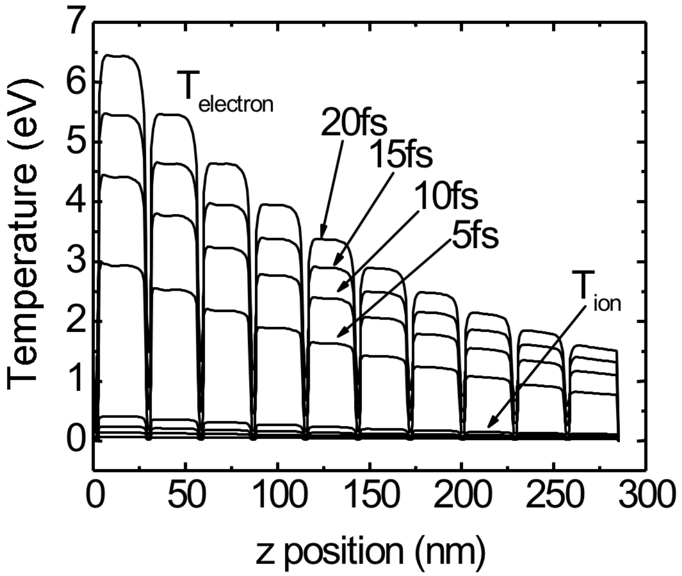


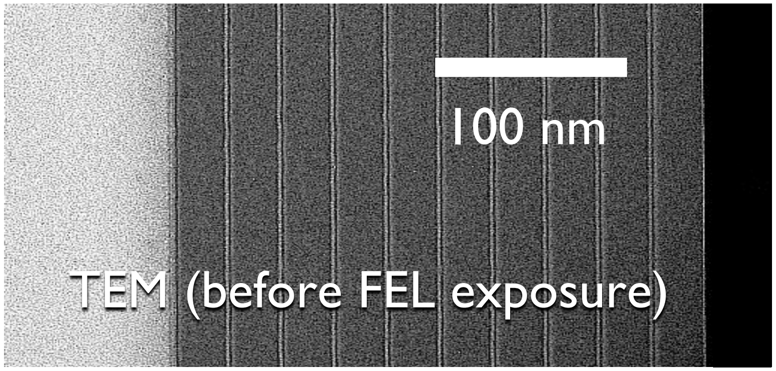


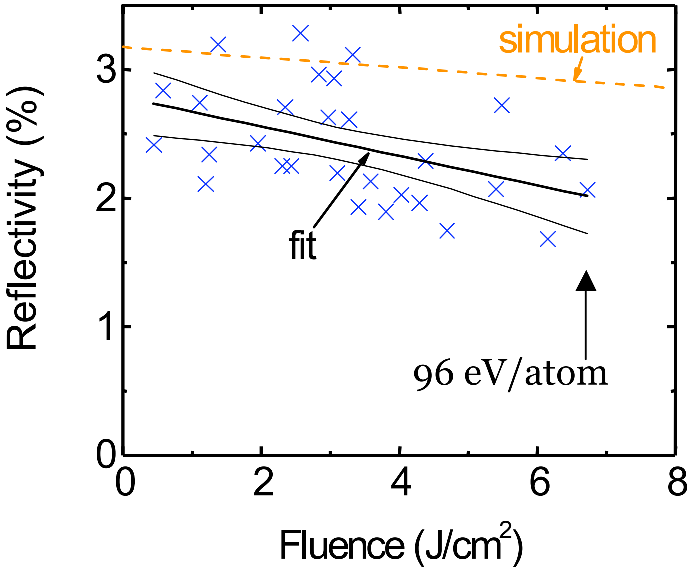
Peak Brightness Collaboration, R. Lee et al
J. Kryzwinski, R. Sobierajski, H. Chapman et al

The multilayer structure can test our hydrodynamic models

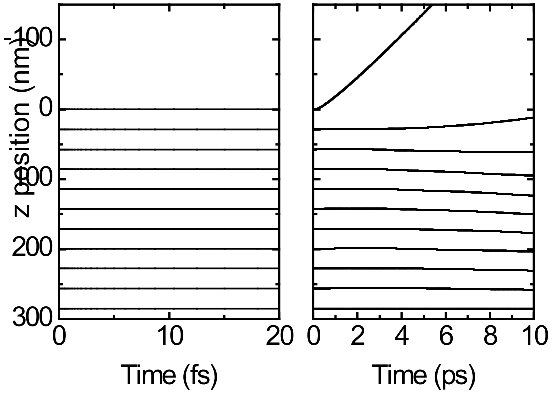
Temperature profile in the multilayer







Motion occurs after the pulse



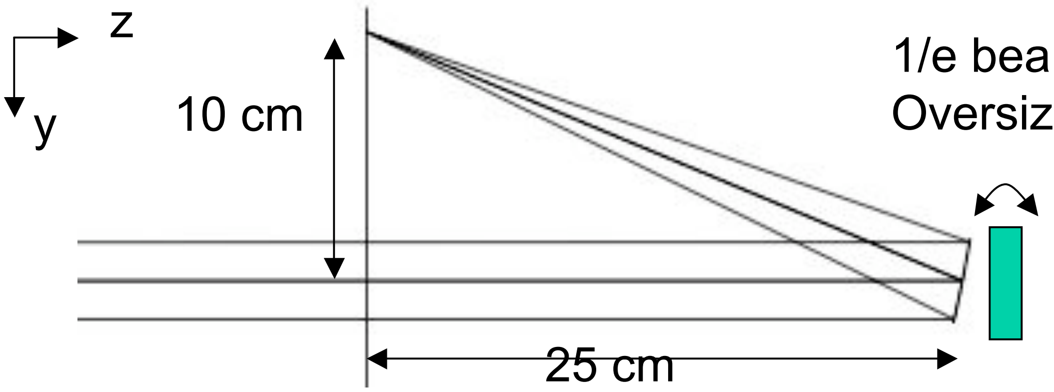
HIGH FIELD EFFECTS

At 32 nm, the high field regime starts at around 10^{17} W/cm^2,
At 6 nm, the high field regime starts at around 2.5 x 10^{18} W/cm^2.

We expect to approach 10^{19} W/cm^2 with submicron focusing this year.

This wiil be needed. It also gives access to a new field of science:
- Studies on non-linearity and multiphoton processes (multiple inner shell ionisations, two-electron ejections)
- Access to hot dense matter regime and to high pressure states
- Magnetic field effects could appear at 10^{18} W/cm^2 at 32 nm
- Relativistic effects above 10^{19} W/cm^2 at 32 nm

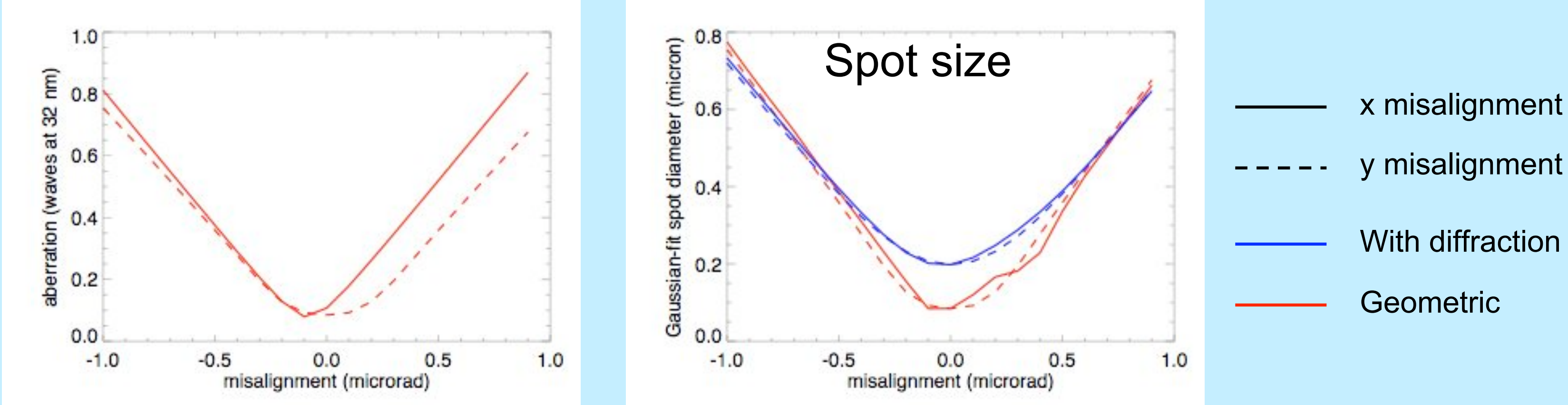
Sub-micron focusing optics could provide ~10^{19} W/cm^2



1/e beam width at optic is 10.4 mm
Oversize optic to 2” diam = 50.8 mm

Mount: must have much better than 50 nm steps (1 microrad over 5 cm)

Optic is not sensitive to x, y displacement (since beam is nearly parallel and optic is a parabola. Optic is sensitive to tilts as shown below. Need better than microradian alignment (use wavefront sensor, but need fine adjustments)

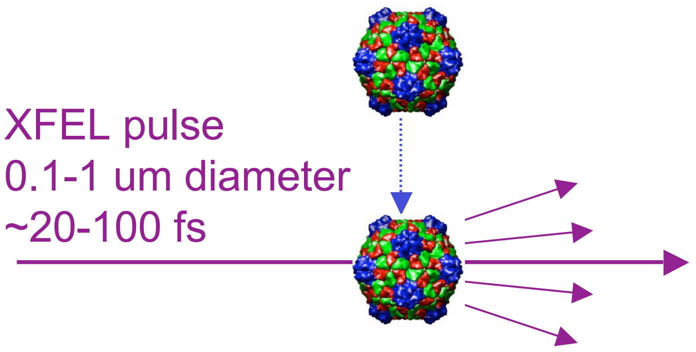


Optic must have <0.5 waves abberration (at 32 nm). That is 16 nm RMS, or 8 nm RMS in surface figure error. i.e. λ/80 (or λ/100 better) surface error at HeNe

SAMPLE INTRODUCTION AND MANIPULATION

The Basic Concept:

1. Take molecules of interest from sample solution
2. Introduce them into the beam

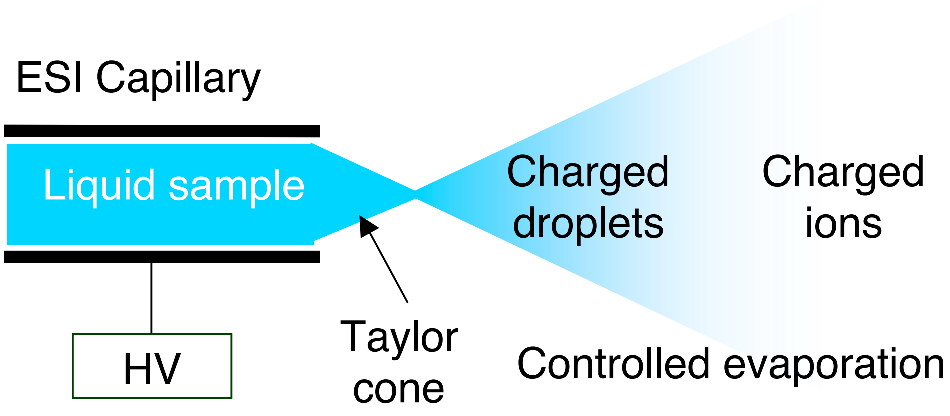


3. Hit them with the XFEL pulse and record diffraction pattern
4. Repeat to get sufficient number of patterns for averaging & image reconstruction.

Challenges:

1. Particle concentration
2. Keeping molecules in “native” conformation
3. Diagnostics: How do we know if a pattern is good?

Introducing Large Molecules into the Beam Using Electrospray Ionization (ESI)



ESI Capillary

Liquid sample

HV

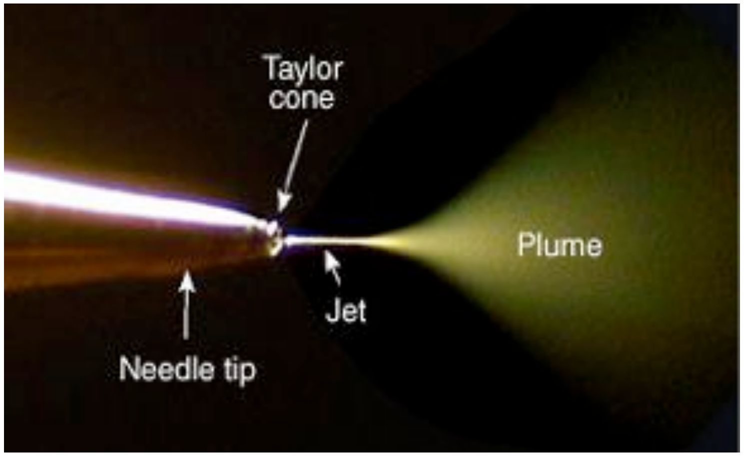
Taylor cone

Charged droplets

Charged ions

Controlled evaporation

www.newobjective.com/electrospray/index.html

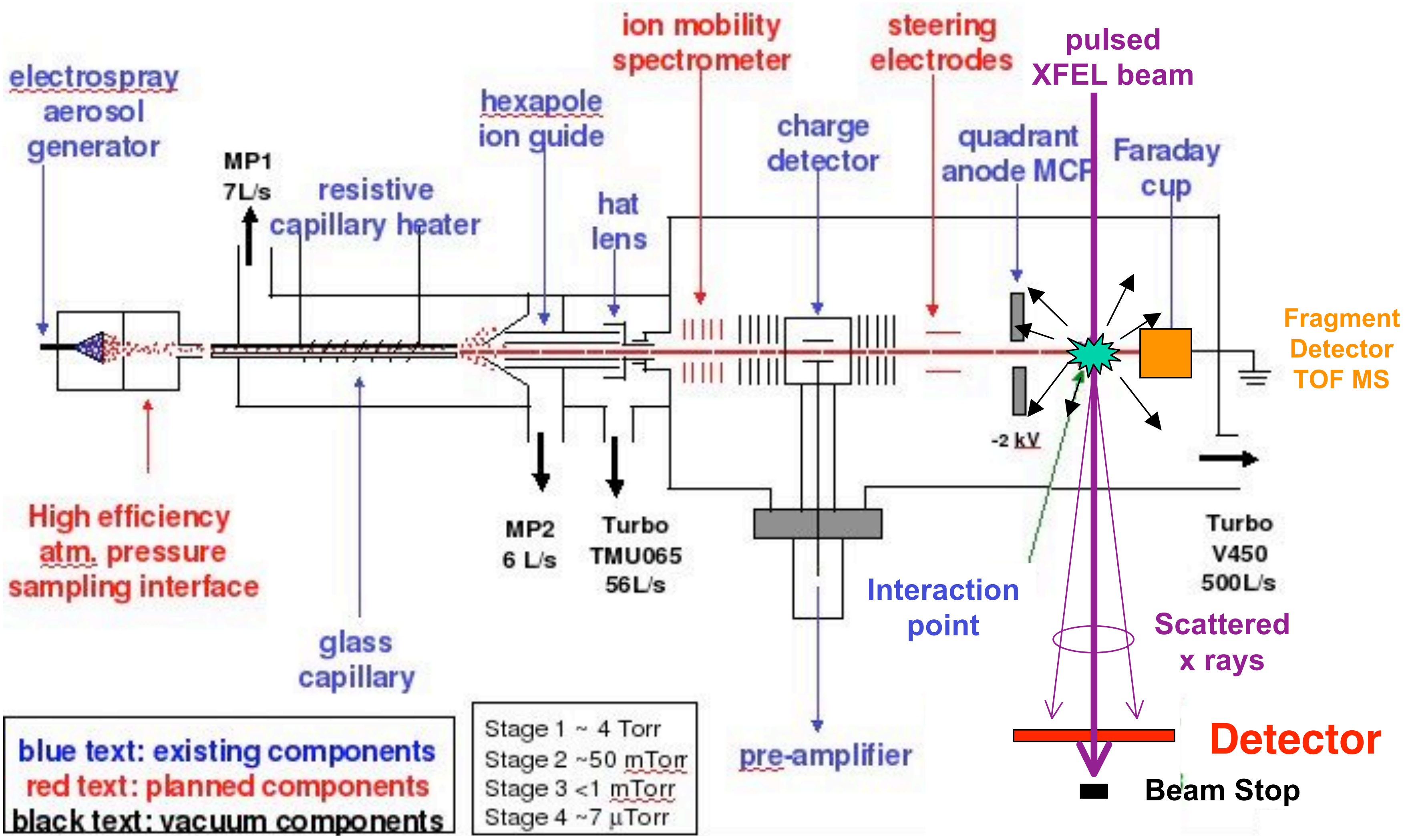


- ESI is widely used to bring macromolecules or viruses into the gas phase, e.g., for mass spectrometry
- ESI can create large number of droplets and molecular ions (~10^{10}/sec at 1 µl/min)
- ESI droplet size adjustable from nm to µm (compared to ~1-50 µm for ink jet droplet generator)
- Both ESI and ink jet can be pulsed as desired
- Droplets / ions can be sucked into vacuum of mass spectrometer or beam line - differential pumping and skimmers

Particle injection system developed at LLNL

(Henry Benner, Matthias Frank, Mike Bogan)

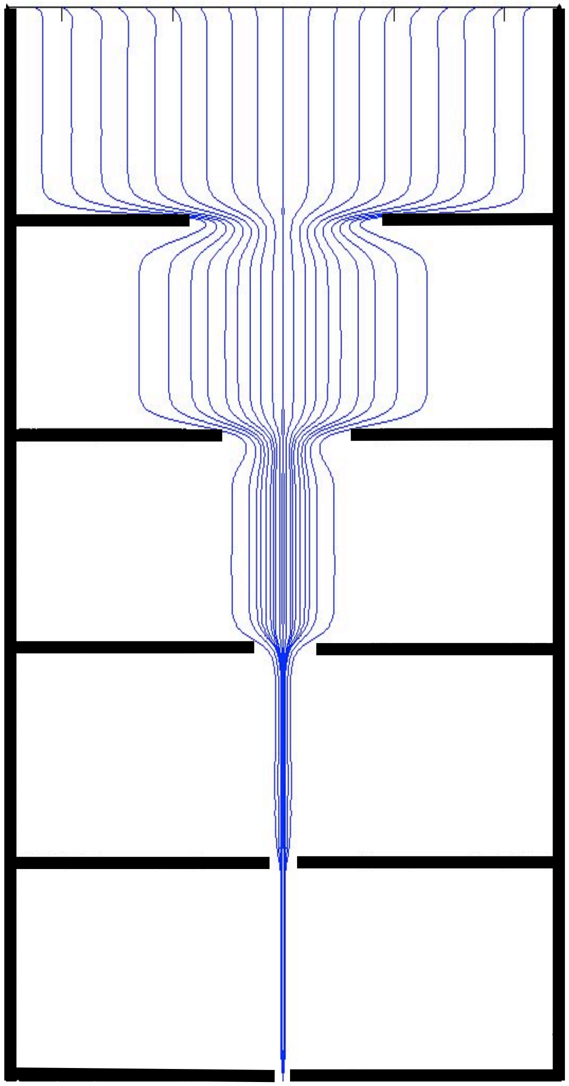
First tests with a pulsed visible laser beam



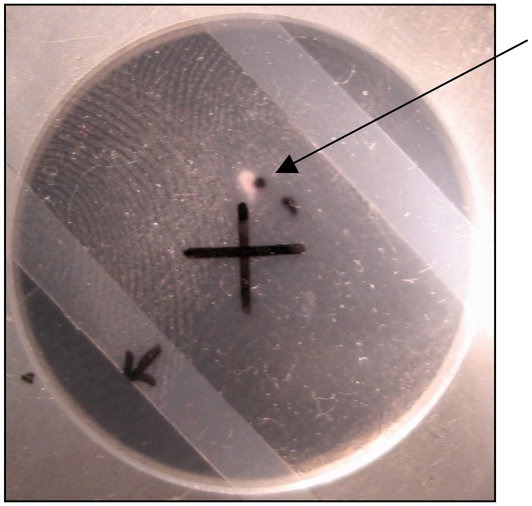
blue text: existing components
red text: planned components
black text: vacuum components

Stage 1 ~ 4 Torr
Stage 2 ~50 mTorr
Stage 3 <1 mTorr
Stage 4 ~7 µTorr

Aerodynamic lens for precision injection of particles into the FEL beam



- Aerodynamic lens: stack of concentric orifices with decreasing openings.
- Can be used to introduce particles from atmosphere pressure into vacuum
- Near 100% transmission
- Creates a tightly focused particle beam. Final focus can be as small as ~10 μm diameter

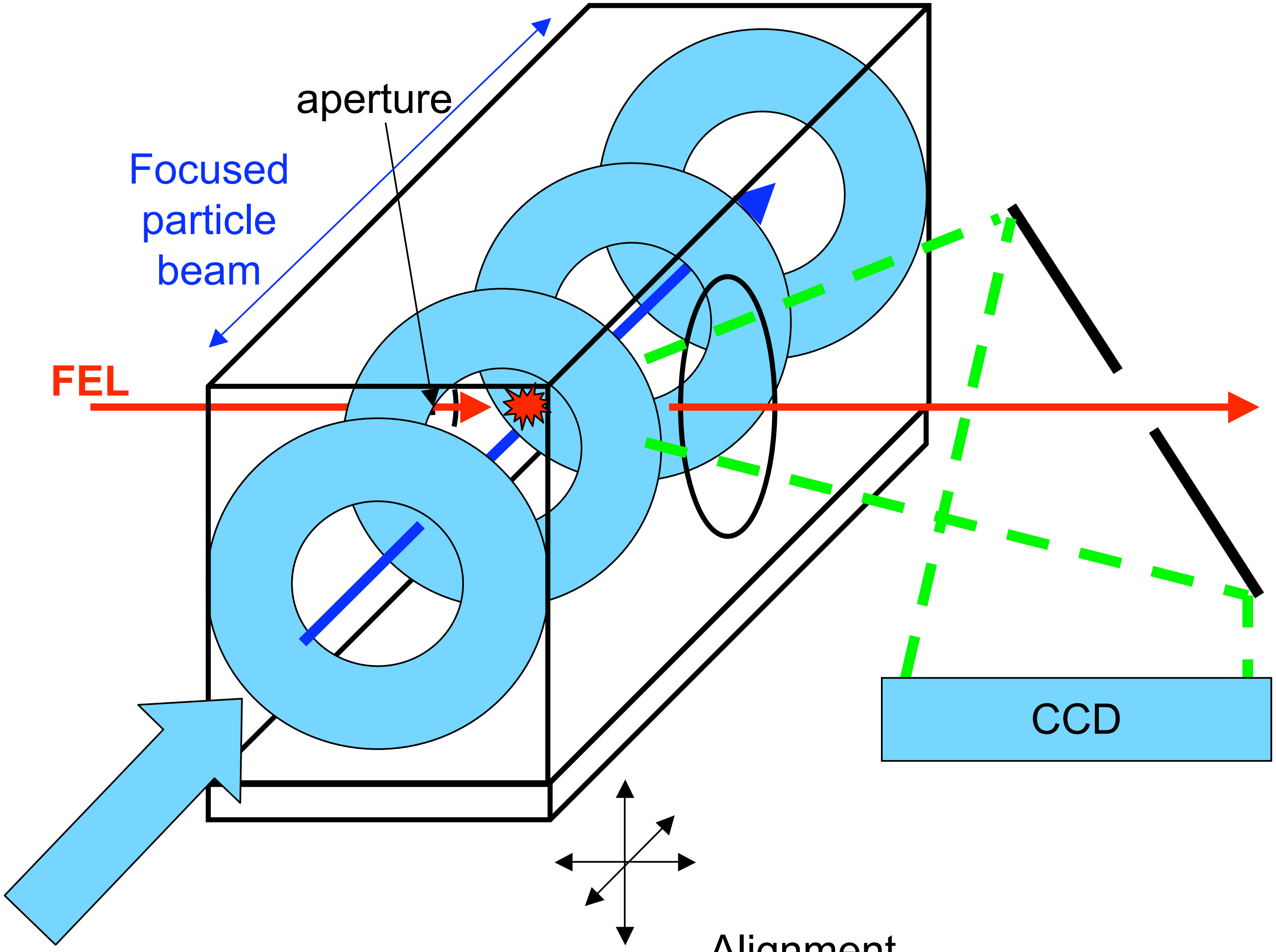


1 μm polystyrene balls sprayed from a distance of *25 cm* for 10 minutes.

Diameter of particle spot deposited on the target: *~500 μm*

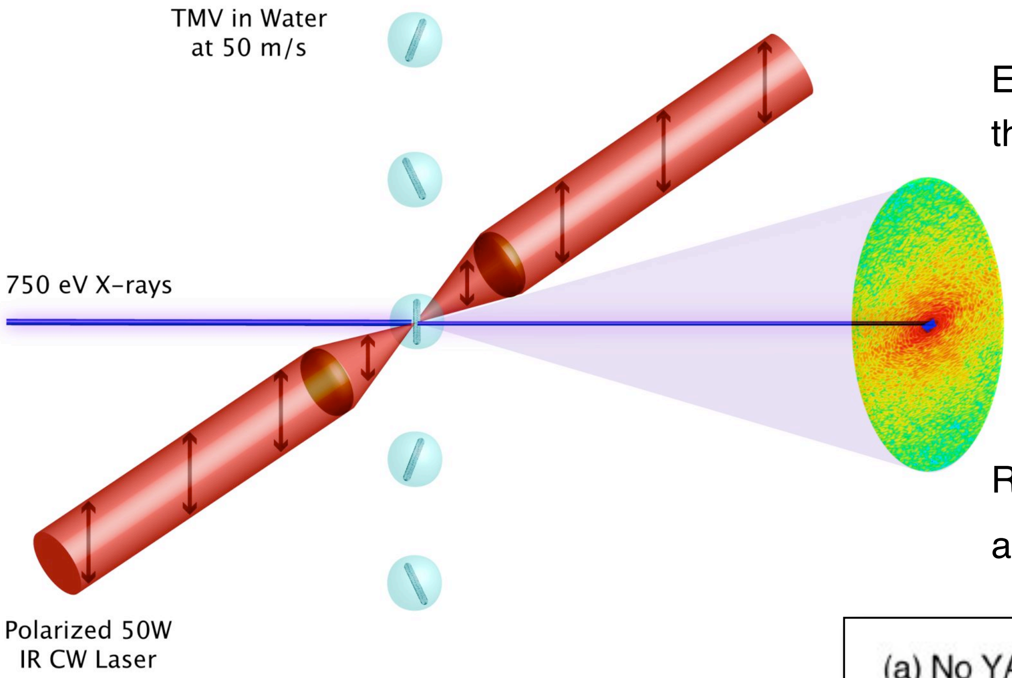
The lens can be aimed just like a gun

We will test such an injector at FLASH later this year



•Unfocused particle beam

Laser alignment will help establish molecular imaging at XFELs and synchrotrons



Equipartition of rotational potential energy with thermal energy gives

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

T - temperature in K

I - laser power in W/cm^2

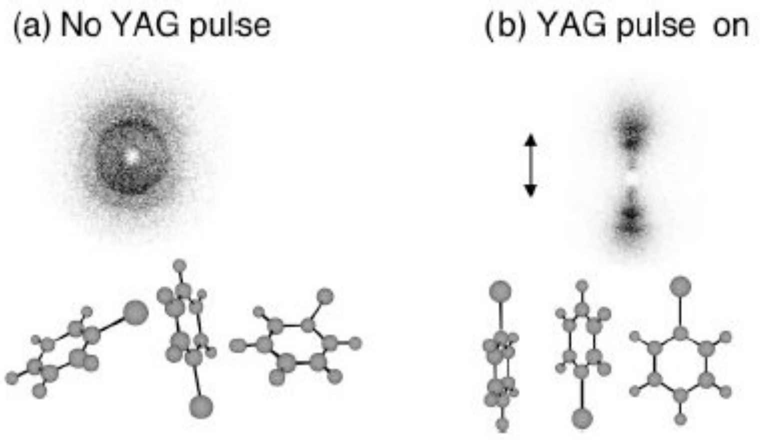
$\Delta\alpha$ - polarizability anisotropy in nm^3

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)*

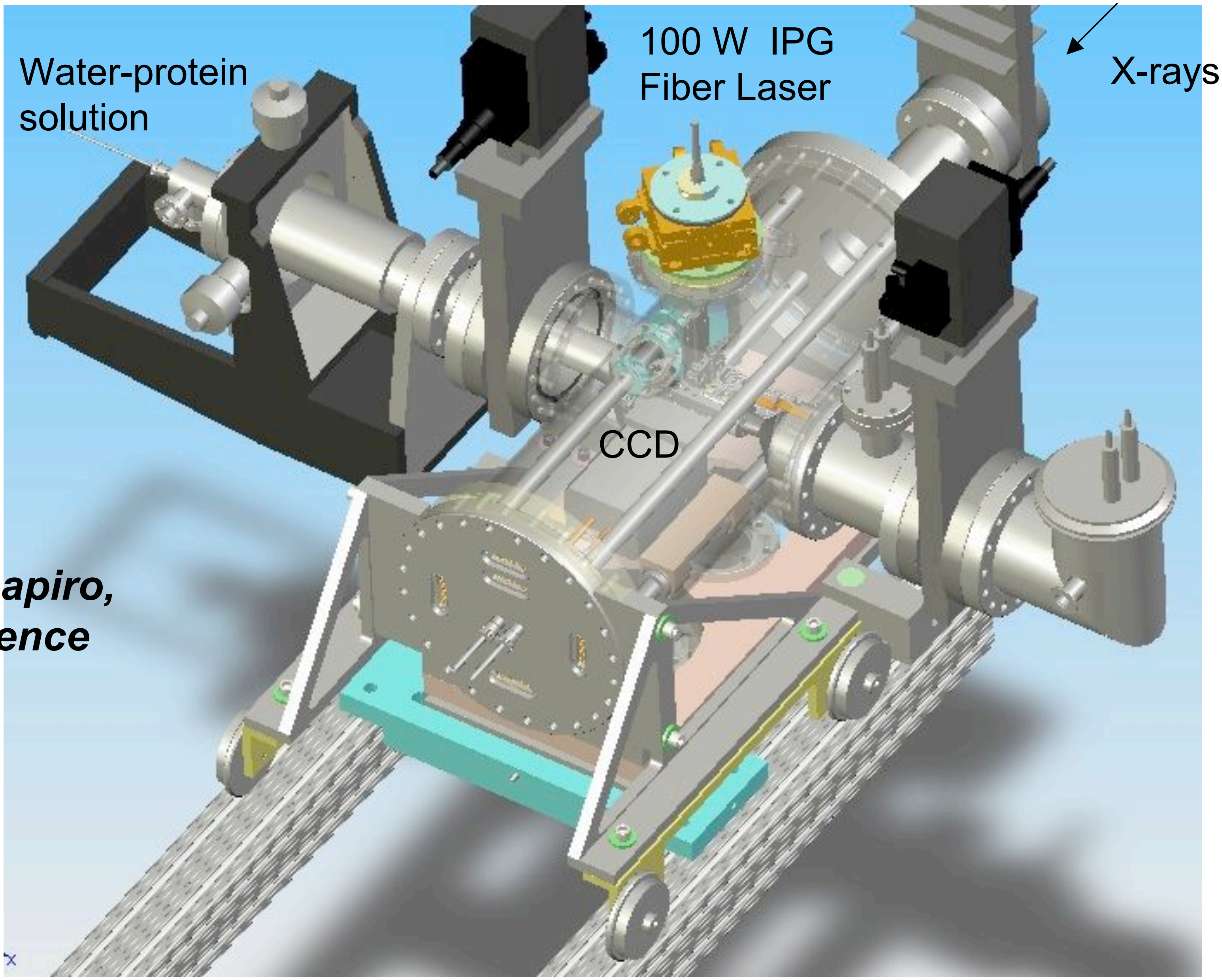
J.C.H. Spence et al., Acta Cryst. A ***61****, 237 (2005)*

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


(a) No YAG pulse (b) YAG pulse on

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

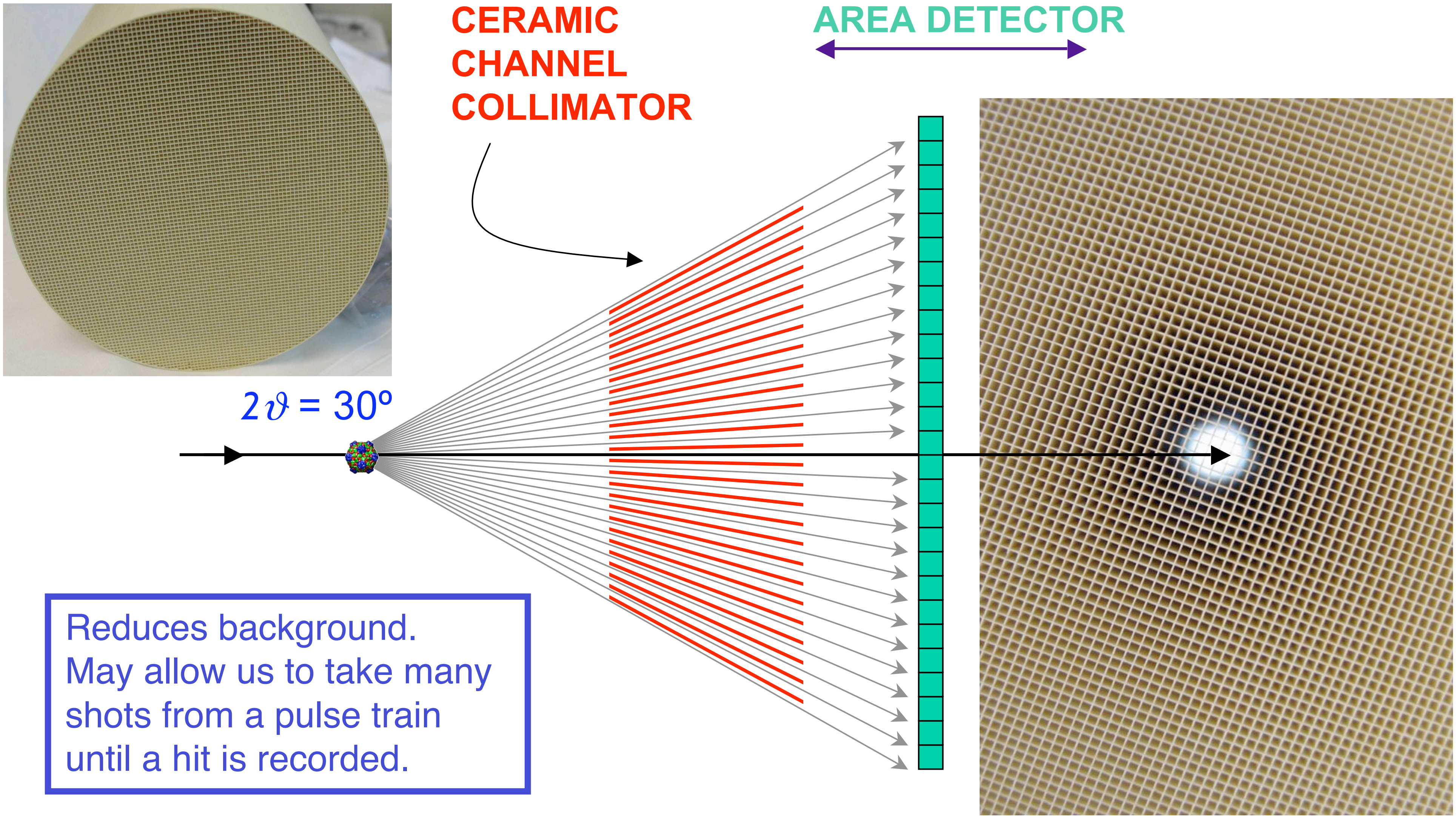
We are testing aligned-particle X-ray diffraction at the ALS



***D. Shapiro,
J. Spence
et al***

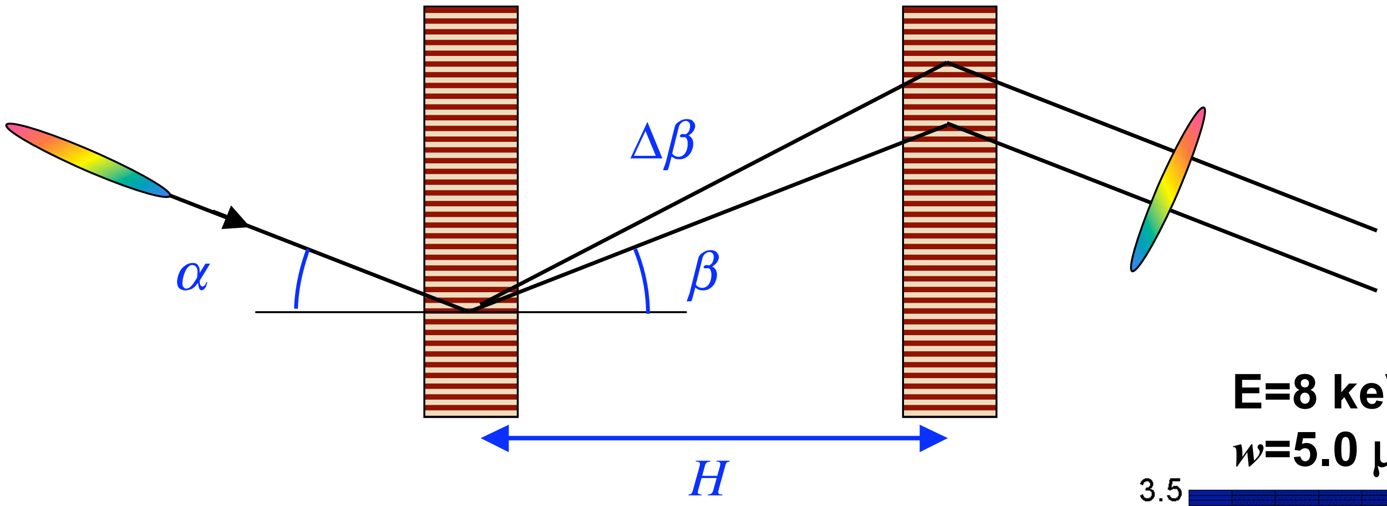
A Ceramic Soller collimator can filter out parastic scattering

Can be manufactured to conical shapes with tapered channels (diameters down to ~10 micrometer)



Reduces background.
May allow us to take many shots from a pulse train until a hit is recorded.

Laue multilayer pulse compressor



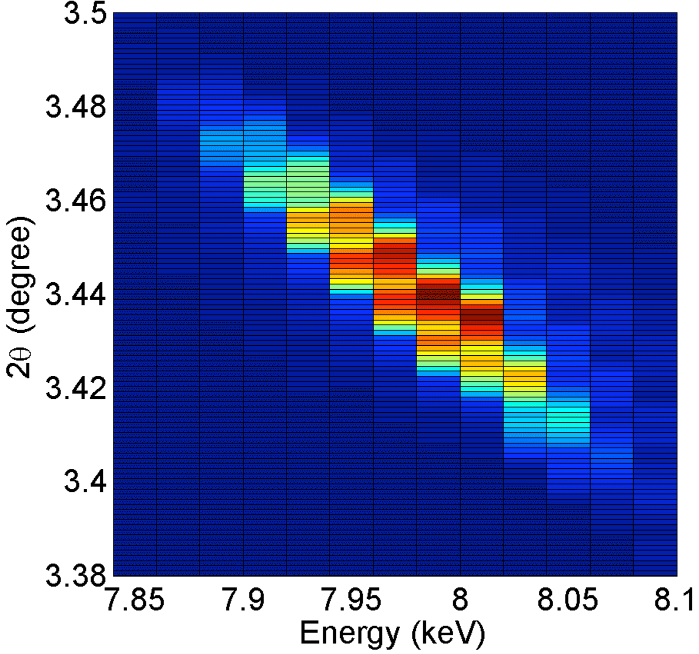
$$\Delta L = \frac{H\,\Delta\lambda}{\lambda}(1+b)^2 \tan^2\theta_B \,\frac{1}{\cos\beta}$$

Experiments at APS:

W/SiC 3280 bilayers, d-spacing = 2.58 nm

Hyon Chol Kang, Brian Stephenson, Saša Bajt

E=8 keV, θ_B=1.7157°, w=5.0 μm, b=1



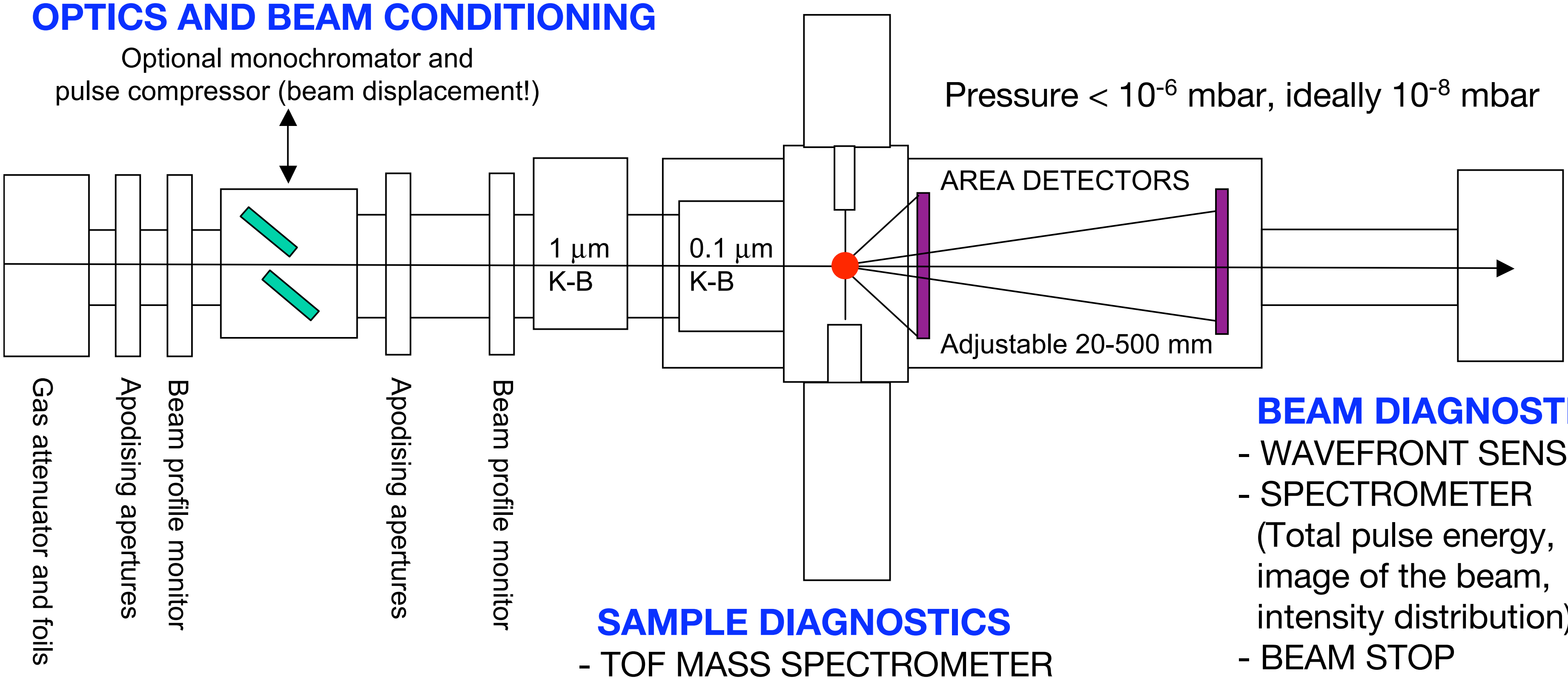
SINGLE PARTICLES, CLUSTER AND BIOMOLECULES: SCHEMATIC LAY OUT OF THE INSTRUMENT

SAMPLE HANDLING

- INJECTION SYSTEM
- LASER FOR PARTICLE ORIENTATION (10^{9}-10^{12} W/cm^{2})
- CRYO EM GONIOSTAT (FLUORESCENCE IMAGE)
- INSPECTION MICROSCOPE

OPTICS AND BEAM CONDITIONING

Optional monochromator and pulse compressor (beam displacement!)



BEAM DIAGNOSTICS

- WAVEFRONT SENSOR
- SPECTROMETER (Total pulse energy, image of the beam, intensity distribution)
- BEAM STOP

SAMPLE DIAGNOSTICS

- TOF MASS SPECTROMETER
- ELECTRON SPECTROMETER
- POSITION-SENSITIVE FLUORESCENCE DETECTOR (to locate/characterise hits)

Initial experiments at LCLS

Follow on from the FLASH experiments

- Diffraction imaging of a cell with a single pulse, to surpass radiation damage limits
- Measure the extent of the Coulomb explosion during the pulse
- Measure the dynamics of the FEL-particle interaction to validate models (pump-probe measurements)
- Diffraction of injected and aligned particles
    - TMV
    - Photosystem 1 nanocrystals and particles
    - Unknown structures
- Single particle diffraction as optics commissioned and injection improved
    - start with Symmetric objects