Imaging Update to LCLS SAC, June 2006
Henry Chapman, LLNL


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

Particle injection

Electrostatic trap

XFEL beam
(focussed, Compressed)

Particle orientation beam

To mass spectrometer

Pixel detector 1

Pixel detector 2

Optical and x-ray diagnostics (veto and determine position of particle in beam)

Readout and reconstruction

Intelligent beam-stop
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. Multilayer reflectivity is uniform across the 30° to 60° gradient. “Soft edge” prevents any scatter from the hole.
The VUV-FEL diffraction experiment is designed to measure forward scattering with high SNR.

- Multilayer mirror
- 60°
- 30°
- Sample
- Shadow mask
- Reflectivity
- 32 nm mirror, 16 nm mirror
- Photon energy (eV)
- 10, 100, 1000
- 0, 0.5, 1
- Sasa Bajt, Eberhard Spiller, and Jennifer Alameda
Image reconstructed from an ultrafast FEL diffraction pattern

1st shot at full power
2nd shot at full power

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

SEM of structure etched into silicon nitride membrane

Edge of membrane support also reconstructed
The reconstruction is carried out to the diffraction limit of the 0.26 NA detector.
The sample is quite damaged by the FEL pulses

FIB “cowboy” sample after FEL exposure

Pulse energy: 10 µJ or 1 J/cm²
Dose in Si₃N₄: 10⁵ J/g or 22 eV/atom
Temperature of 6×10⁴ K, or 5.2 eV, ionization/atom of 2.5
Surfaces will expand at sound speed: 1.3×10⁶ 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.

Hit one pyramid, then rotate and move up sample.

High-resolution imaging will be carried out at the third harmonic, e.g. a wavelength of 4.5 nm. Requires 2 \( \mu \text{m} \) focus.

Silicon nitride pyramid

Silicon nitride membrane with pyramids

Silicon
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

\[ E > \frac{3eQ}{2R} \]
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%.

- Half beam diameter (10 μm) - 220 particles
- Mounted on piezo x-y stage to move each window into beam
- 357 windows per chip
- 20 nm thick silicon nitride

![Image of particle explosion setup with measurements and labels](image-url)
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.
The substrate is a high-resolution detector (at low enough fluence)

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

Electric field intensity at z=0

Intensity vs. R (nm)

150nm sphere

$\lambda = 32\, \text{nm}, D = 155\, \text{nm}$

$\lambda = 32.5\, \text{nm}, D = 145\, \text{nm}$
We invented a new method called femtosecond time-delay holography.

Time delay: $2 \Delta z / c$

Multilayer Mirror

30 fs pulse (9 µm long)

Detector

Unfolded geometry

Reference object

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} \text{ W cm}^{-2}) with a 32 nm wavelength. Si/C multilayer reflectivity is unchanged, and multilayer spacing is not changed by more than 0.3 nm.

After pulse, plasma forms, and layers ablate. The peak brightness collaboration includes R. Lee et al., J. Kryzwinski, R. Sobierajski, H. Chapman et al.
The multilayer structure can test our hydrodynamic models.
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 $2.5 \times 10^{18}$ W/cm$^2$.

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

This will 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 $\sim 10^{19} \text{ W/cm}^2$

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)

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

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

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

[Image of ESI process]

[Link to ESI website: www.newobjective.com/electrospray/index.html]
Particle injection system developed at LLNL
(Henry Benner, Matthias Frank, Mike Bogan)

First tests with a pulsed visible laser beam
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.
Laser alignment will help establish molecular imaging at XFELs and synchrotrons

\[
\langle \Delta \theta^2 \rangle = \frac{T}{3 \times 10^{-8} I \Delta \alpha}
\]

- \( T \) - 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:

\[
d = \frac{L}{2} \Delta \theta
\]

---


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, \( \theta_B = 1.7157^\circ \), \( w = 5.0 \, \mu m \), \( b = 1 \)
SINGLE PARTICLES, CLUSTER AND BIOMOLECULES: SCHEMATIC LAY OUT OF THE INSTRUMENT

OPTICS AND BEAM CONDITIONING
Optional monochromator and pulse compressor (beam displacement!)

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

Pressure < 10^{-6} mbar, ideally 10^{-8} mbar

AREA DETECTORS
Adjustable 20-500 mm

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

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