Ultrafast Coherent Diffraction Imaging with a Soft X-Ray Free-Electron Laser

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Just before XFEL pulse

During the pulse

After pulse

Diffraction pattern

FEL 2006, 1st September

This work was performed under the auspices of the U. S. DOE by LLNL under Contract No. W-7405-ENG-48.
Acknowledgements


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 Kuhlman, Rolf Treusch, Stefan Dusterer, Jochen Schneider

TU Berlin: Thomas Möller, Christof Bostedt, Matthias Hoener
We are entering a new era in x-ray science

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

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**Euro-XFEL**
- Peak Brightness: $10^{34}$
- Energy: 10 keV

**LCLS (200 fs)**
- Peak Brightness: $10^{32}$
- Energy: 100 keV

**FLASH (30 fs)**
- Peak Brightness: $10^{30}$
- Energy: 1000 keV

**SPPS (80 fs pulses)**
- Peak Brightness: $10^{28}$
- Energy: 10 keV

**APS undulator**
- Peak Brightness: $10^{26}$
- Energy: 100 keV

**ALS undulator**
- Peak Brightness: $10^{24}$
- Energy: 10 keV

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**LCLS operational 2009**
- 8 keV, 200 fs, $10^{12}$ photons
- Linac Coherent Light Source, SLAC, Stanford

**FLASH operational now**
- Current: 38 eV, 30 fs
- 500 eV, 80 fs, $10^{13}$ photons
- Tesla Test Facility
- DESY, Hamburg
Radiation damage affects atomic scattering factors and atomic positions.

50 fs
4x10$^{14}$ photons/μm²
12 keV

Radiation damage affects atomic positions and atomic scattering factors

Coulomb explosion of Lysozyme

20 fs
$4 \times 10^{14}$ photons/μm$^2$
12 keV

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

One pulse, one measurement

Particle injection

10 fs pulse

Noisy diffraction pattern

Combine $10^5$-$10^7$ measurements

Classification

Averaging

Orientation

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

FLASH 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
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.
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

Prior knowledge about object

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

Resolution: \[ \delta = \frac{\lambda}{\sin \theta} \]

First demonstration with X-rays: John Miao, P. Charalambous, J. Kirz and D. Sayre, Nature 400 (1999)
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.

90 nm

32 nm, one wavelength

$\lambda / NA$
The sample is quite damaged by the FEL pulses

FIB “cowboy” sample after FEL exposure

Melted silicon nitride

Silicon frame

10 micron
We have performed full 3D X-ray imaging of non-crystalline material at 10 nm resolution

Coherent X-ray diffraction data $\lambda = 1.6$ 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 16-node cluster. Resolution = 10 nm
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.

357 windows per chip

20 nm thick silicon nitride

100 μm

50 mm

15°

Half beam diameter (10 μm) - 220 particles

1 mm
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 explosion takes longer than expected from our hydrodynamic model

- Experiments and simulations show a similar trend of the particle exploding
- The onset of explosion occurs later than predicted
- Measurements will be improved with better pulse diagnostics and shorter wavelength
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

With J. Kryzwinski, R. Sobierajski, L. Juha et al
There is no motion at 3Å during the pulse, but the change in optical constants is larger than expected.
Our model predicts atomic resolution imaging is feasible

S. Hau-Riege, R. London, A. Szoke, G. Huldt
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