X-ray free-electron lasers and ultrafast science at the atomic and molecular scale.

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Albert Einstein in the Bern patent office in 1905.

In one of the four important papers that Einstein published in 1905, he advanced the hypothesis that light can act as if it consists of discrete, independent particles of energy. This proposal was contrary to the accepted theory that light consists of electromagnetic waves, but he showed that his "light quanta" could explain phenomena like the photoelectric effect, the emission of electrons from illuminated metals.

Light is both particles and waves!

Another kind of light: X-rays

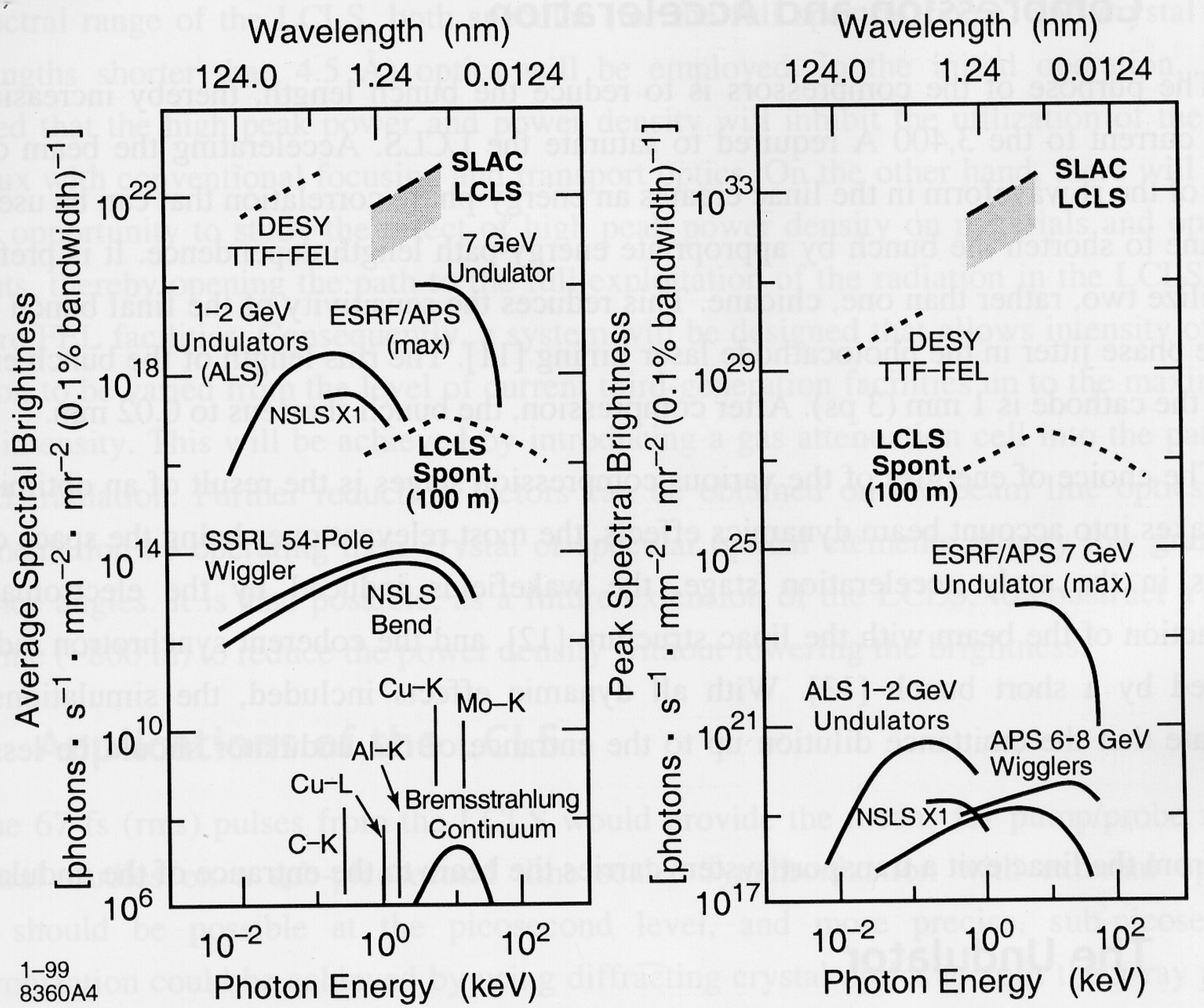
On 8 November 1895, Roentgen was working in his lab, studying the properties of a cardboard-shrouded electrical discharge tube. He was surprised to see that when the tube was operated, an object across the room began to glow. He called the invisible,mysterious and unknown agent doing it X-rays.

12/22/1985
Frau
Roentgen’s
Hand.

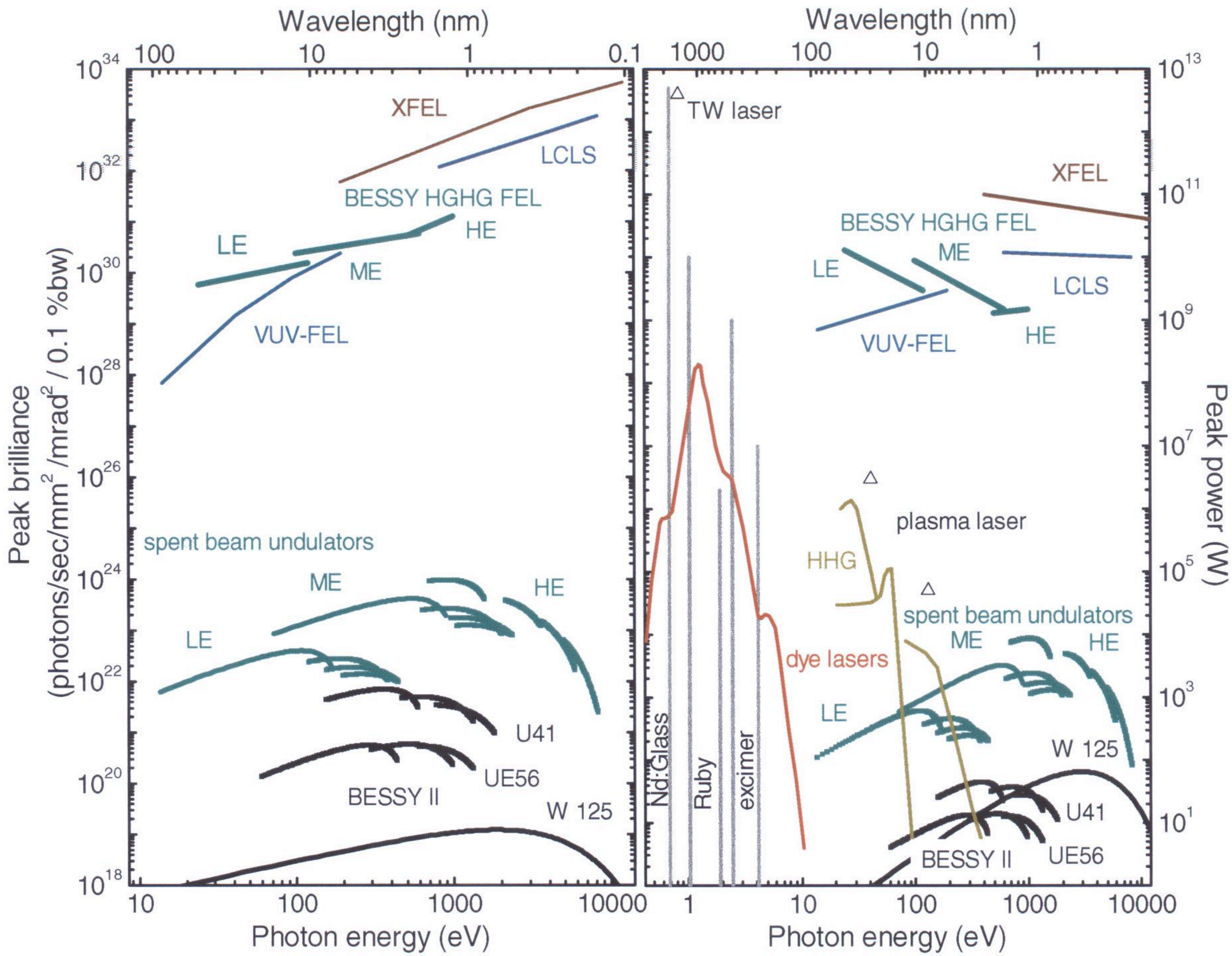
Roentgen won the first Nobel prize in physics in 1901. He did not take any patent on X-rays and their applications.

X-Ray FELs

After many years of research and development an X-ray free-electron laser (X-FEL) operating in the 0.1 nm spectral region, the LCLS, first proposed by C. Pellegrini in 1992, is now being built and will be completed by 2008. Another X-FEL operating at the same wavelength has been proposed at DESY as a European project. Other similar projects are being developed in Japan, China and Korea. Several FELs operating in the few nanometer region are being developed and built in Europe, the US and Asia.



Wavelength (nm)
124.0
1.24
0.0124
Average Spectral Brightness
[photons · s^{-1} · mm^{-2} · mr^{-2} (0.1% bandwidth)$^{-1}$]
10^{22}
10^{18}
10^{14}
10^{10}
10^{6}
SLAC
LCLS
DESY
TTF-FEL
7 GeV
Undulator
1-2 GeV
Undulators
(ALS)
ESRF/APS
(max)
NSLS X1
LCLS
Spont.
(100 m)
SSRL 54-Pole
Wiggler
NSLS
Bend
Cu-K
Mo-K
Al-K
Cu-L
C-K
Bremsstrahlung
Continuum
10^{-2}
10^{0}
10^{2}
Photon Energy (keV)
1-99
8360A4
Wavelength (nm)
124.0
1.24
0.0124
Peak Spectral Brightness
[photons · s^{-1} · mm^{-2} · mr^{-2} (0.1% bandwidth)$^{-1}$]
10^{33}
10^{29}
10^{25}
10^{21}
10^{17}
SLAC
LCLS
DESY
TTF-FEL
LCLS
Spont.
(100 m)
ESRF/APS 7 GeV
Undulator (max)
ALS 1-2 GeV
Undulators
APS 6-8 GeV
Wigglers
NSLS X1
10^{-2}
10^{0}
10^{2}
Photon Energy (keV)



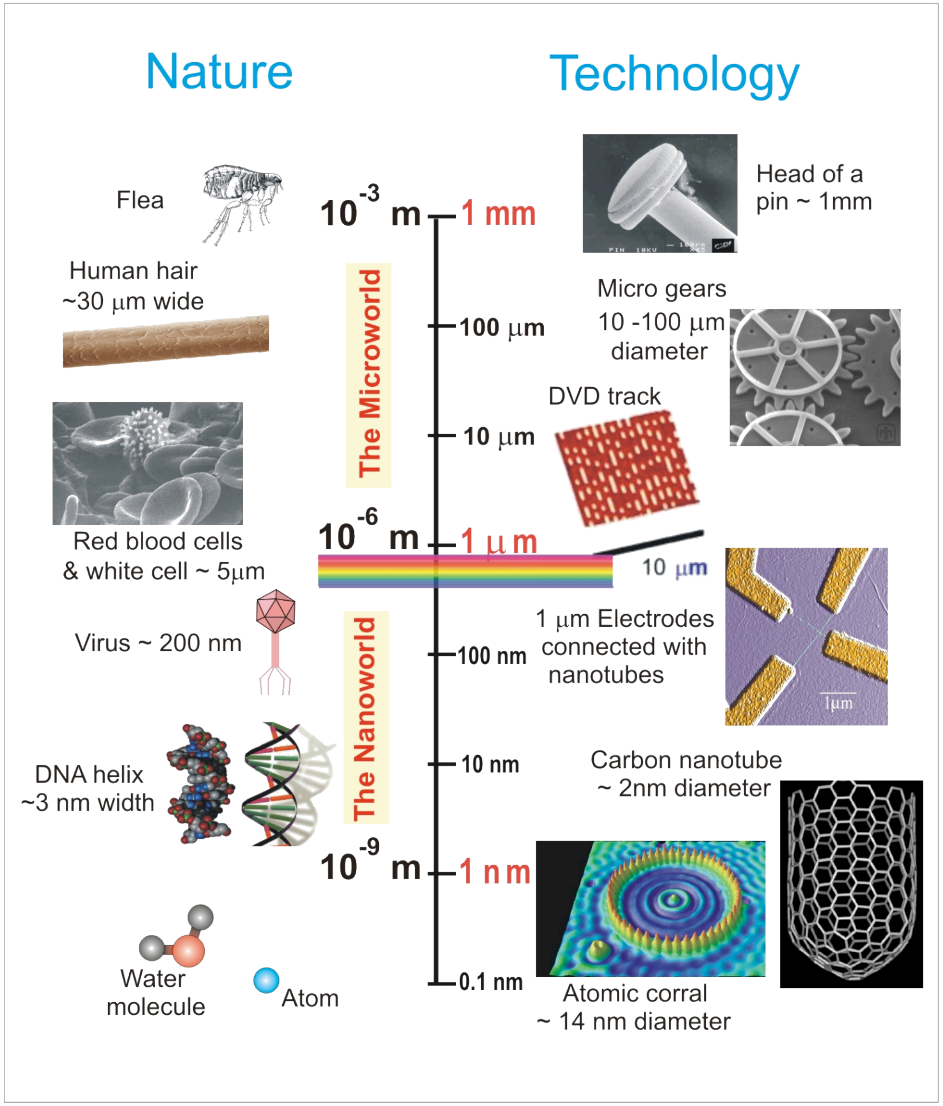
Wavelength (nm)
100
10
1
0.1
XFEL
LCLS
BESSY HGHG FEL
LE
ME
HE
VUV-FEL
spent beam undulators
ME
HE
LE
U41
UE56
BESSY II
W 125
Peak brilliance
(photons/sec/mm^2/mrad^2 / 0.1 %bw)
10^{34}
10^{32}
10^{30}
10^{28}
10^{26}
10^{24}
10^{22}
10^{20}
10^{18}
10
100
1000
10000
Photon energy (eV)
Wavelength (nm)
1000
100
10
1
TW laser
XFEL
BESSY HGHG FEL
ME
LE
LCLS
VUV-FEL
HE
plasma laser
HHG
spent beam undulators
ME
HE
dye lasers
LE
Nd:Glass
Ruby
excimer
W 125
U41
BESSY II
UE56
1
10
100
1000
10000
Photon energy (eV)
Peak power (W)
10^{13}
10^{11}
10^{9}
10^{7}
10^{5}
10^{3}
10^{1}

X-Ray FELs

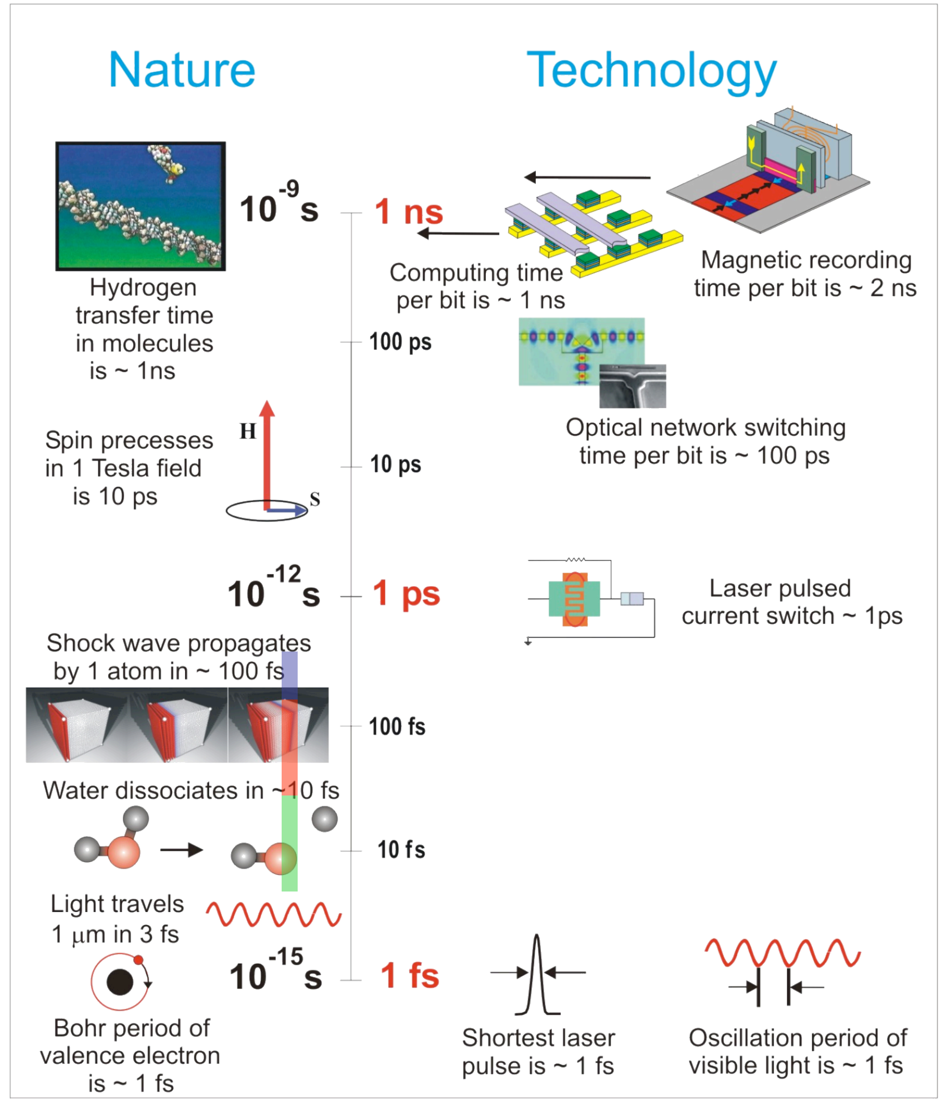
The world-wide interest in X-FELs is motivated by their characteristics of tunability, high peak power, short pulse length, and their promise to open a new field of exploration of matter at the atomic and molecular level with unprecedented time-space resolution.

X-Rays have opened the Ultra-Small World
X-FELs open the Ultra-Small and Ultra-Fast Worlds

Ultra-Small



Ultra-Fast



X-Ray FELs: the LCLS

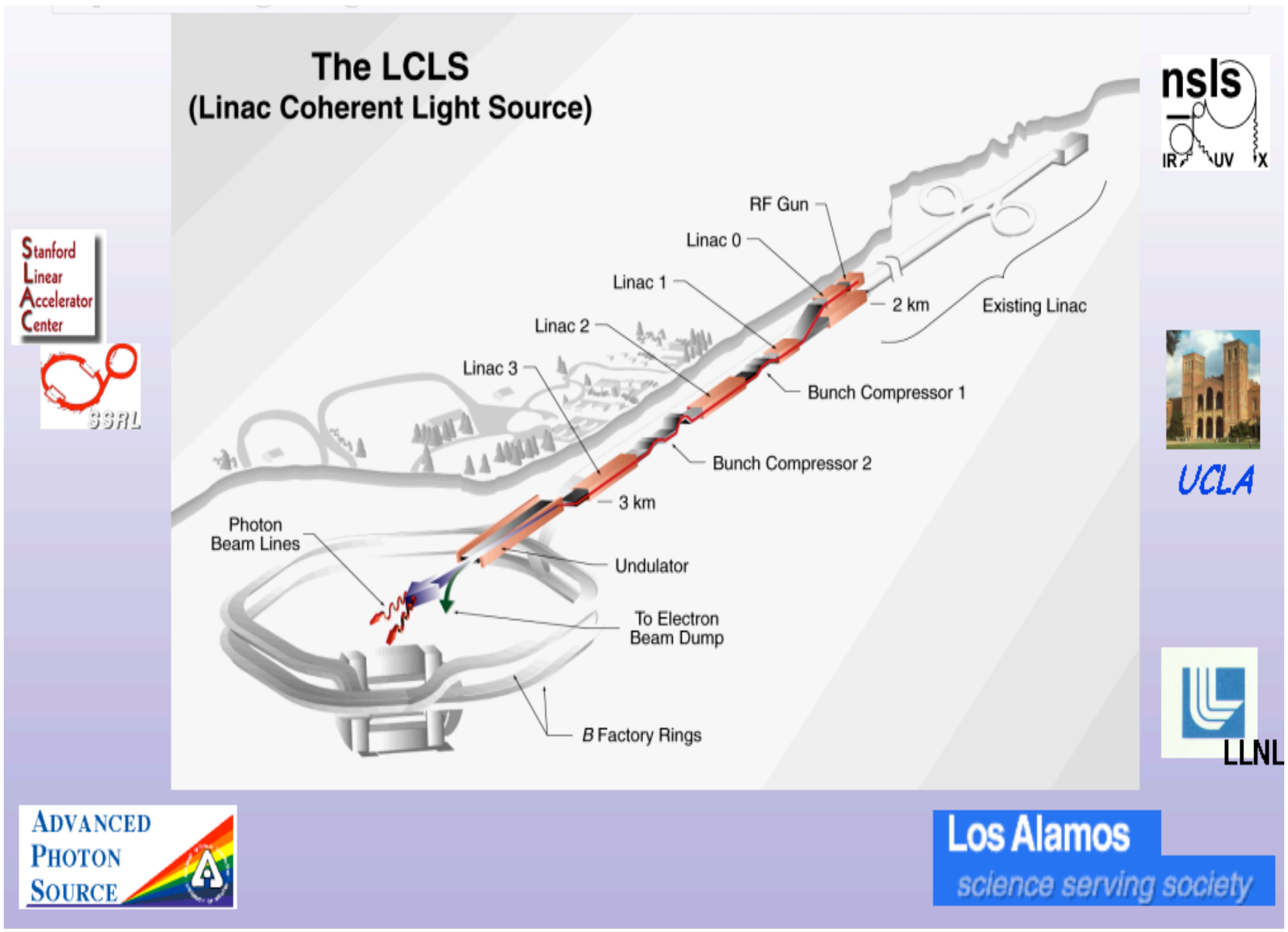
The LCLS, a SLAC-ANL-LLNL-UCLA collaboration, which will begin operating at SLAC in 2009, will offer new ways of studying and constructing nanotechnology devices; will be able to capture the structural rearrangements of atoms in reactions like photosynthesis and catalysis; will create and probe extreme states of plasmas found in the cores of giant planets and proto-stars; and will explore how proteins function as the engines of life.

X-ray SASE-FEL Main Characteristics

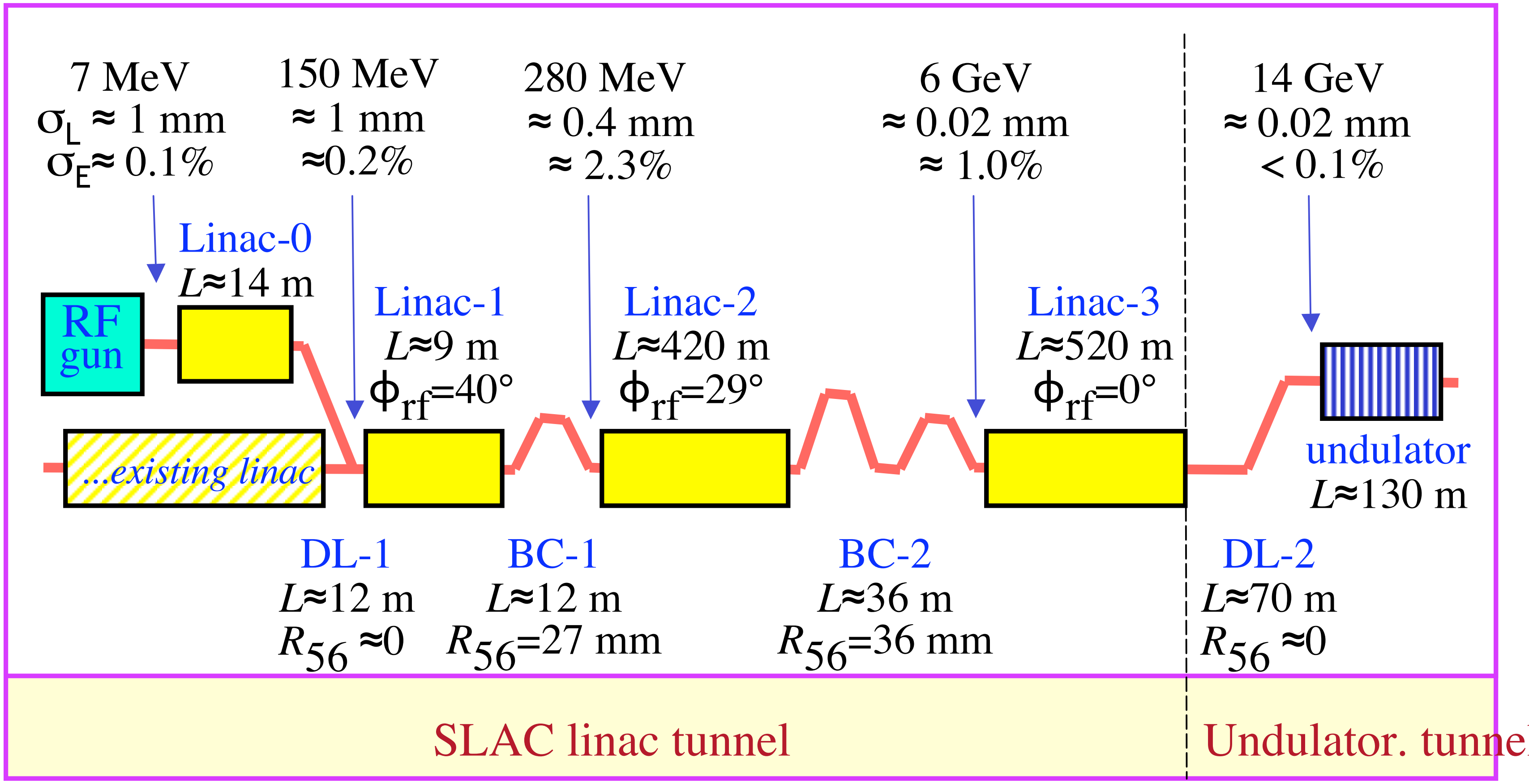
- ~ 10 Gigawatt or more peak power
- ~100 femtosecond pulse length or shorter
- Transversely coherent, diffraction limited
- Line width < 0.001
- Tunable from 15 to 0.5Å

The X-ray FEL is a powerful tool to explore matter and fundamental physics.

LCLS: a SLAC-ANL-LLNL-UCLA collaboration



LCLS schematic



LCLS uses 1 km of the SLAC linac, a new high brightness electron source, and two bunch compressors. The normalized beam emittance is 1 mm mrad. Final peak current 3.4 kA.

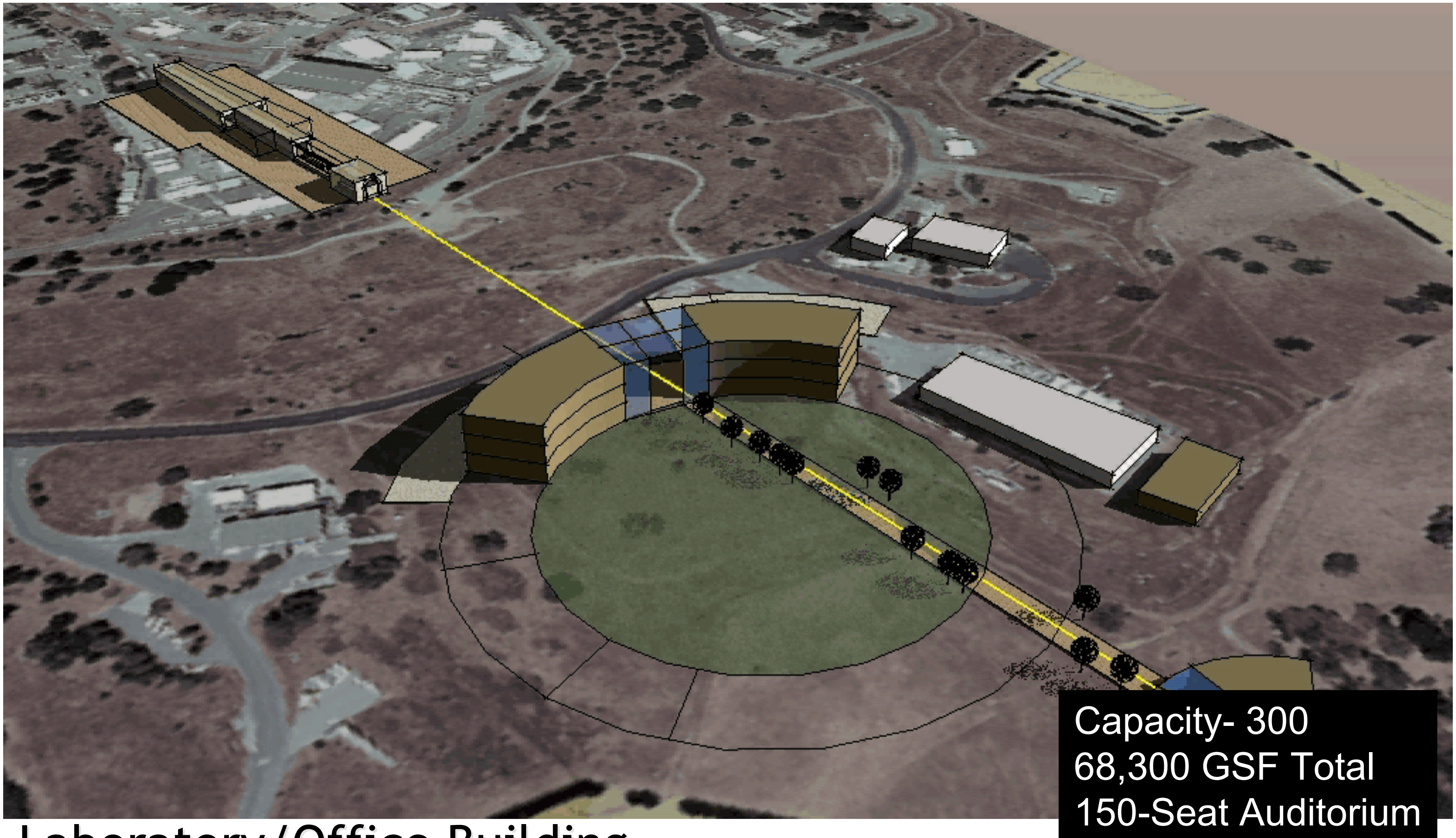
LCLS-Phase I Wavelength	15	1.5	Å
Electron Energy E	5	14.5	GeV
Repetition Rate	120	120	Hz
Saturation Length	32.2	87	m
Peak Power (Average Power)	10.6 (.32)	9 (0.28)	GW (W)
Pulse Duration (rms)	100	100	fs
Energy (Photons)/Pulse	2.6 (20)	2.3 (1.8)	mJ (10^{12})
Peak (Average) Brightness	0.8 (0.39)	12 (2.1)	10^{32} (10^{21})
X-rays Radius (Divergence), rms	37 (3.2)	27 (0.4)	µm (µ rad)

LCLS: a 4th generation light source

- The construction of LCLS is supported by the U.S Department of Energy, Office of Basic Energy Sciences. In a recent review of the US-DOE large projects for the next 20 years the LCLS received one of the highest priority.

- The construction will be completed in 2008, and operation will start at the beginning of 2009.

- The total project cost is about $350M, including new buildings and experimental facilities.



Laboratory/Office Building

Courtesy J. Galayda

X-Ray FELs

The X-ray FEL is based on the high gain regime of FELs, developed mostly in the 80's, including the self amplified spontaneous radiation regime (SASE). Because there are no good optical cavities in the 0.1 nm regime, the X-rays FELs operate in the SASE regime, as a high gain amplifier of spontaneous undulator radiation, using a single pass of a long undulator magnet.

The experimental verification of the SASE-FEL theory was done in the 90's, and had to wait the development, initially done at Los Alamos, of a high brightness electron source, the RF photo-injector, which is now part of all projects.

LCLS physical characteristics

The LCLS is a SASE-FEL. It is based on the FEL Collective Instability (Bonifacio, Pellegrini, Narducci, Optics Comm. 50, 313 (1984))

- ✓ All key characteristics are given by one universal FEL parameter: $\rho=\{(K/4\gamma)(\Omega_p/\omega_w)\}^{2/3}$

  with $\omega_w=2\pi c/\lambda_w$, Ω_p=beam plasma frequency.
- ✓ Gain Length: $L_G=\lambda_w/4\pi\rho$,
- ✓ Saturation: $P\sim\rho\, I_{beam}\, E$
- ✓ Saturation length: $L_{sat}\sim 10L_G \sim\lambda_w/\rho$
- ✓ Line width: $1/N_w\sim\rho$
- ✓ Cooperation length $L_C=\lambda/4\pi\rho$

FEL Collective Instability Characteristics

- The FEL instability occurs if

$\sigma_E < \rho$ (cold beam)

$\varepsilon \sim \lambda/4\pi$ (Phase-space matching)

$Z_R/L_G > 1$ (Optical guiding)

- Number of photons/electron at saturation

$N_{ph} \sim \rho E/E_{ph}$

For E_{ph}=10keV, E=15 GeV, $\rho=10^{-3}$, $N_{ph} \sim 10^3$

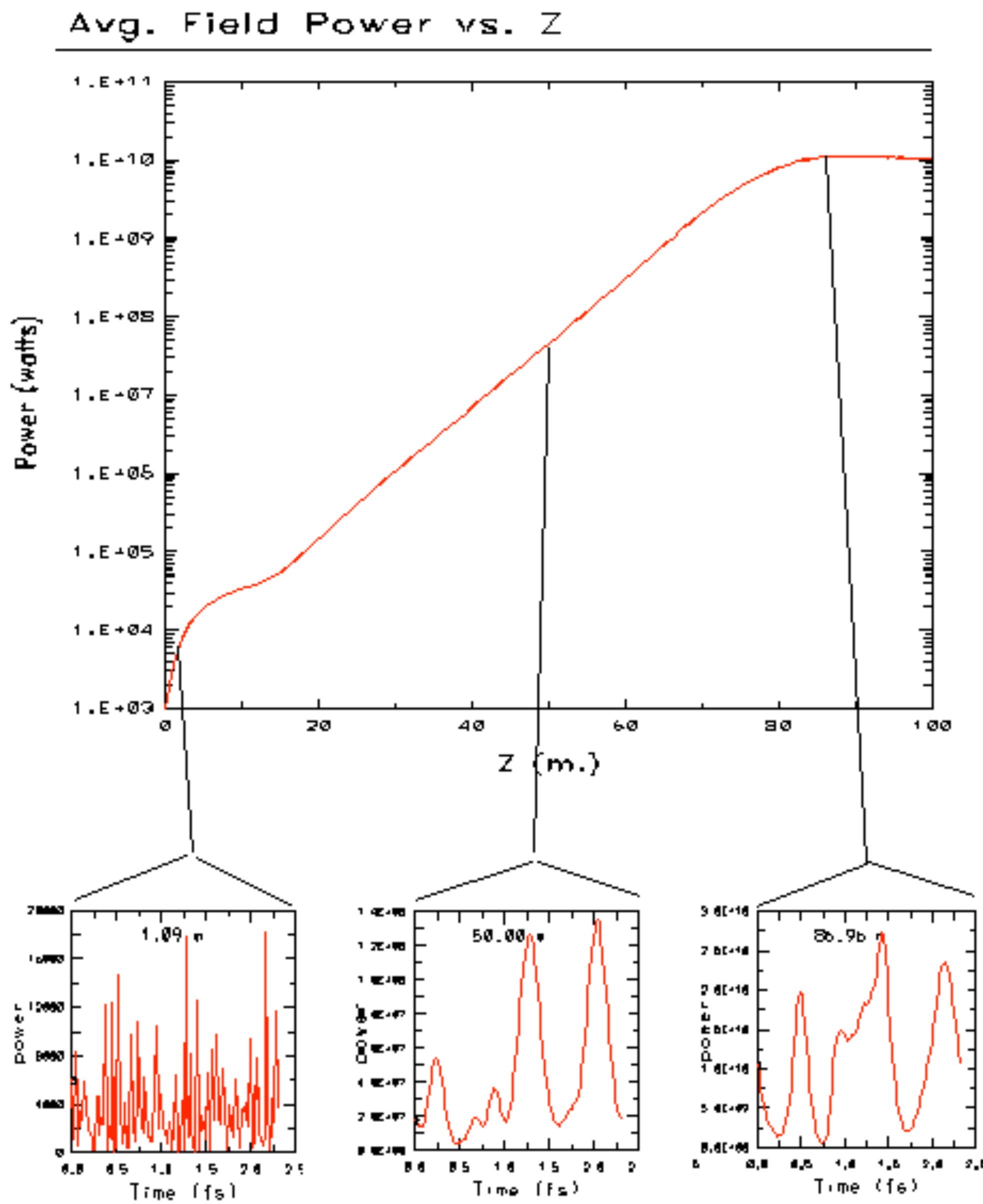
Slippage and Cooperation Length, Time Structure

U C L A

- The radiation propagates faster than the electron (it "slips" by λ per undulator period); thus electrons communicate with the ones in front; total slippage $S=N_w\lambda$.
- Cooperation length (slippage in one gain length) $L_c=\lambda/4\pi\rho$.
- Number of "spikes": bunch length/$2\pi L_c$.

SASE-FEL

Power
and
spikes



U
C
L
A

LCLS Radiation Characteristics

For LCLS L_c=0.34 μm~1fs

$\Delta\lambda/\lambda$=4x10^{-4}

N_S=250, L_P ~100 fs, rms

The Fourier transform line-width is about 100 times smaller than the spike line-width, or $(\Delta\lambda/\lambda)_T$~3x10^{-6}.

Other Properties and Options

Many types of undulators and wigglers can be used:

- helical undulator: no harmonics on axis, circularly polarized;
- planar undulator: rich harmonics content, the third harmonic is amplified;
- X-band radio-frequency undulators, with large gap to period ratio.

X-rays Coherence properties

The LCLS radiation has unprecedented coherence, about 10^9 photons in the coherence volume. The energy of coherent photons can be pooled to create multi-photons excitations and carry out non-linear X-ray experiments. This is a largely unexplored area of science.

3rd gen. beam line

LCLS source

coherence volume 1 x 5 x 50μm

coherence volume 0.1 x 100 x 100μm

contains < 1 photon

contains 10^9 photons

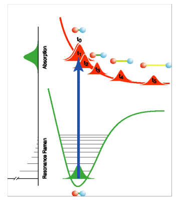
Other LCLS characteristics

LCLS radiation on the third harmonic, at 0.05 nm, will have a peak power of about 100 MW.

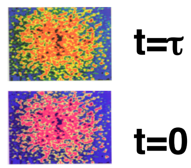
LCLS phase II will reduce the X-ray pulse length from the initial value of about 100 fs to about 10 to 1 fs.

It is also possible to increase the final peak power using longer tapered undulator.

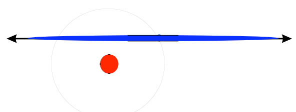
LCLS - The first experiments, starting about 2008



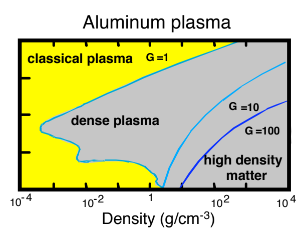
Femtochemistry — Dan Imre, BNL



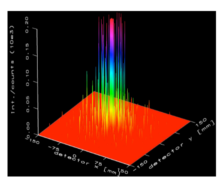
Nanoscale Dynamics in Condensed Matter — Brian Stephenson, APS



Atomic Physics — Phil Bucksbaum, Univ. of Michigan



Plasma and Warm Dense Matter — Richard Lee, LLNL



Structural Studies on Single Particles and Biomolecules — Janos Hajdu, Uppsala Univ.

Program developed by international team of scientists. More than 350 scientists , including many Europeans, participated in a recent meeting to prepare the LCLS experiments.

LCLS Experimental Program: Interaction of high power X-ray beams with matter.

The first LCLS experiment will be aimed at understanding the fundamental interaction process of the high power X-ray beam with atoms, molecules and clusters. It will explore the formation of hollow atoms, where the X-rays can strip electrons from the inside out. It will also study multiphoton processes enabled by the large coherent intensity. Finally, it will investigate the disintegration and explosion of clusters, yielding information on the time scale of the damage caused by the X-ray beam, a fundamental question for other experiments.

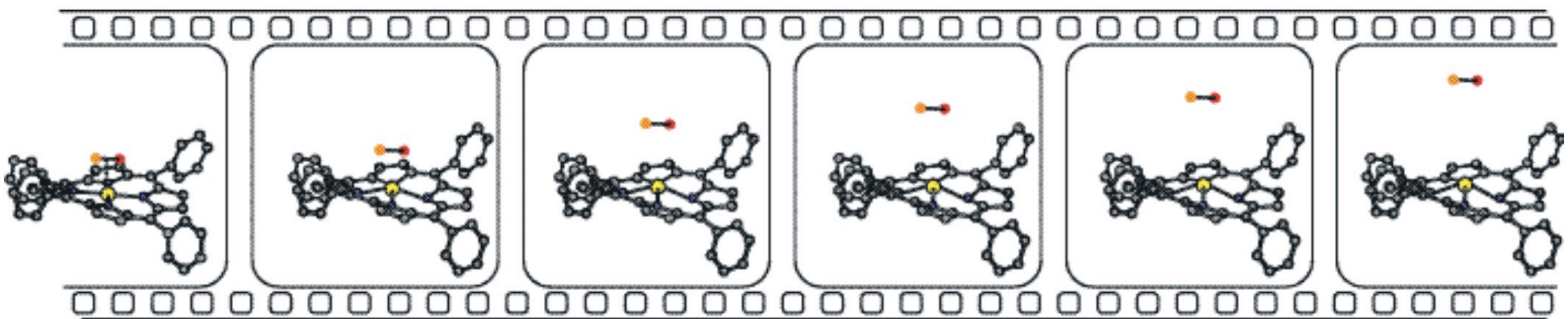
LCLS Experimental Program: Plasmas and warm dense matter.

A second proposed experiment uses LCLS to create and investigate warm (WDM) and hot (HDM) dense states of matter, that exist in astronomical objects and are important for inertial fusion. Conventional lasers have provided limited information on these systems, because they cannot penetrate the high-density matter, and few theories can make any prediction.

LCLS Experimental Program: Femtosecond chemistry

The study of molecular reactions is the heart of chemistry. With ultrafast laser spectroscopy we can glimpse at electron transfer processes in response to an optical excitation pulse. But despite their great success, conventional lasers can only study electronic excitations, and cannot “see” the positions of the atoms during the various transformation stages. This can be done using the LCLS X-rays to take diffraction snapshots.

LCLS Experimental Program: Femtosecond chemistry



Snapshots with atomic resolution and femtosecond time intervals of a molecular dissociation will be made possible by X-ray FELs. The real space images would be reconstructed from ultrashort x-ray diffraction patterns.

These experiments are important because many important discoveries in biology and chemistry can be traced back to the determination of a structure.

Temporal and Spatial Resolution

TIME

The very light systems require a time resolution of a few femtoseconds, while heavier ones can be studied with pulses a few hundred femtosecond long.

BOND LENGTH

The *LCLS* will make it possible to map out the nuclear motions with a resolution of 0.1 Å, which is clearly sufficient.

LCLS Experimental Program: Imaging with coherent photons

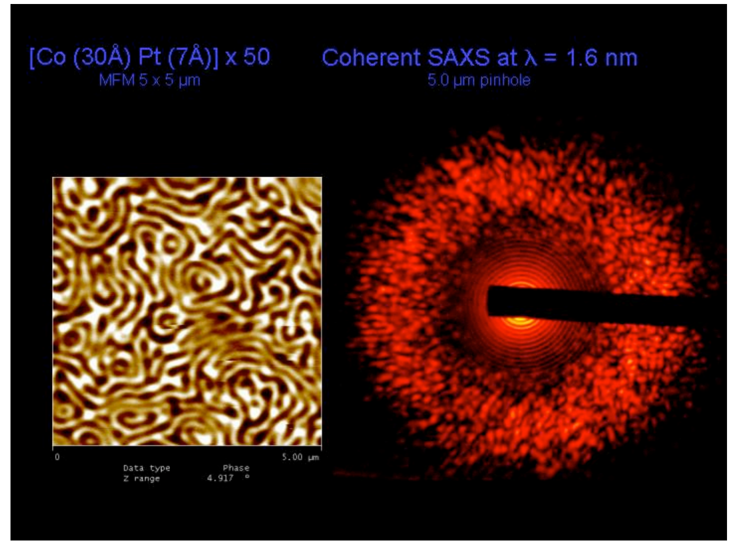
The spontaneous emission of radiation from X-ray tubes or synchrotron radiation sources is incoherent. However if the sample is far from the source, the X-ray phase and amplitude is well defined over a small sample volume, and one can get interference from structures within that volume. Examples are Bragg peaks or small angle scattering peaks, which reflect “structure” within a coherence volume of a few hundred Ångstroms. To extend the structural sensitivity to larger, micrometer, dimensions the x-ray coherence volume must be increased beyond these dimensions. If this is done by moving the sample further from the source, the intensity is reduced.

LCLS Experimental Program: Imaging with coherent photons

With 3rd generation high brightness synchrotron sources these experiments become possible. An example is the coherent diffraction or “speckle” pattern. The “speckle” pattern contains the detailed information on the true structure of the sample in the illuminated area. Because of the small intensity these measurements require integration over long times, seconds or more.

LCLS Experimental Program: Imaging with coherent photons

Magnetic worm domain pattern of a CoPt alloy with perpendicular anisotropy, recorded by transmission x-ray microscopy. Right: Coherent x-ray diffraction or speckle pattern from a 5μm diameter region of the same sample, selected by a 5μm circular aperture. The central rings are the Fraunhofer diffraction pattern from the circular aperture which are well separated from the speckle pattern of the smaller magnetic domains.



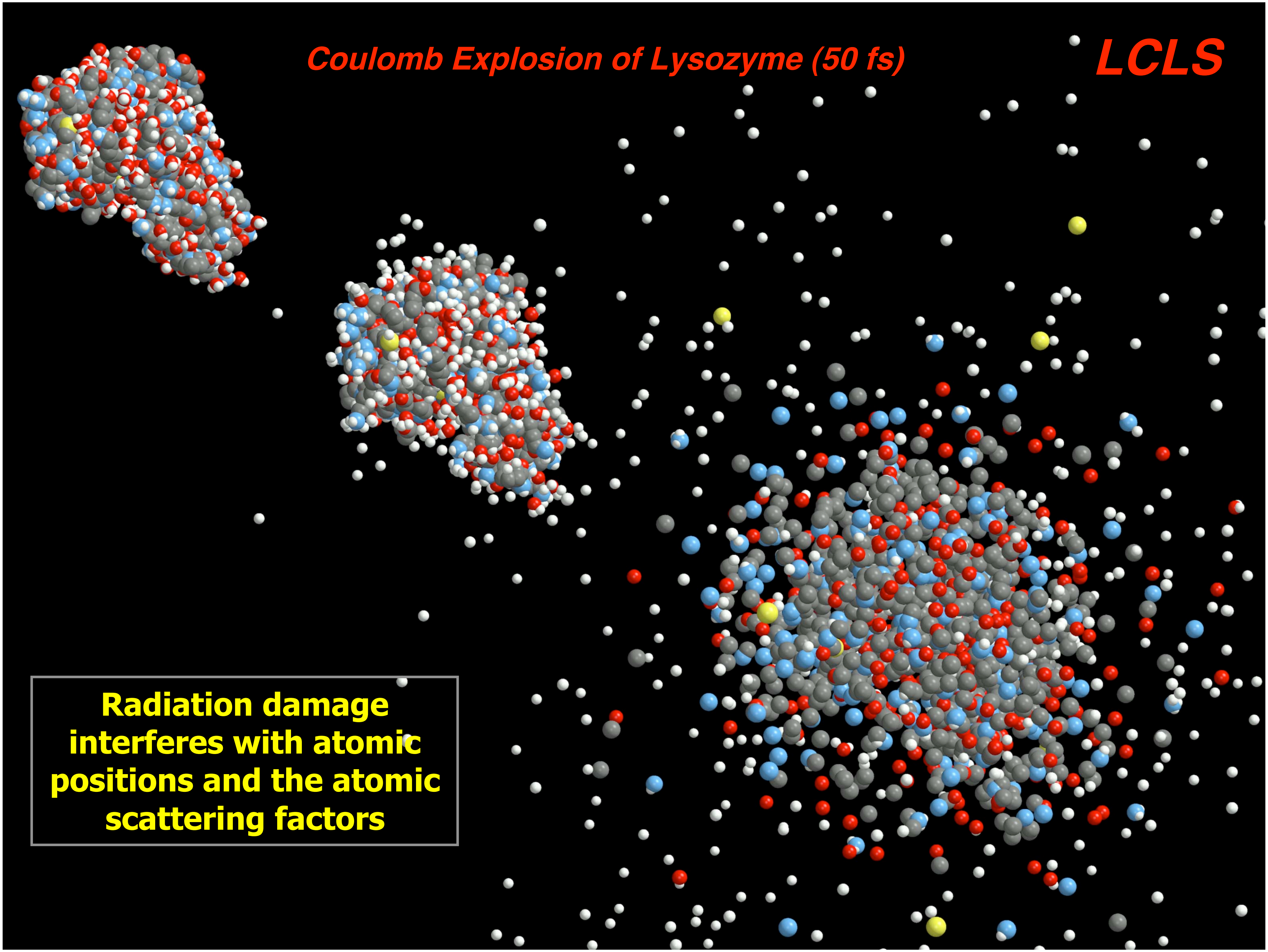
Any change of the sample magnetic structure is reflected by an intensity change in the pattern.

LCLS Experimental Program:
Imaging with coherent photons

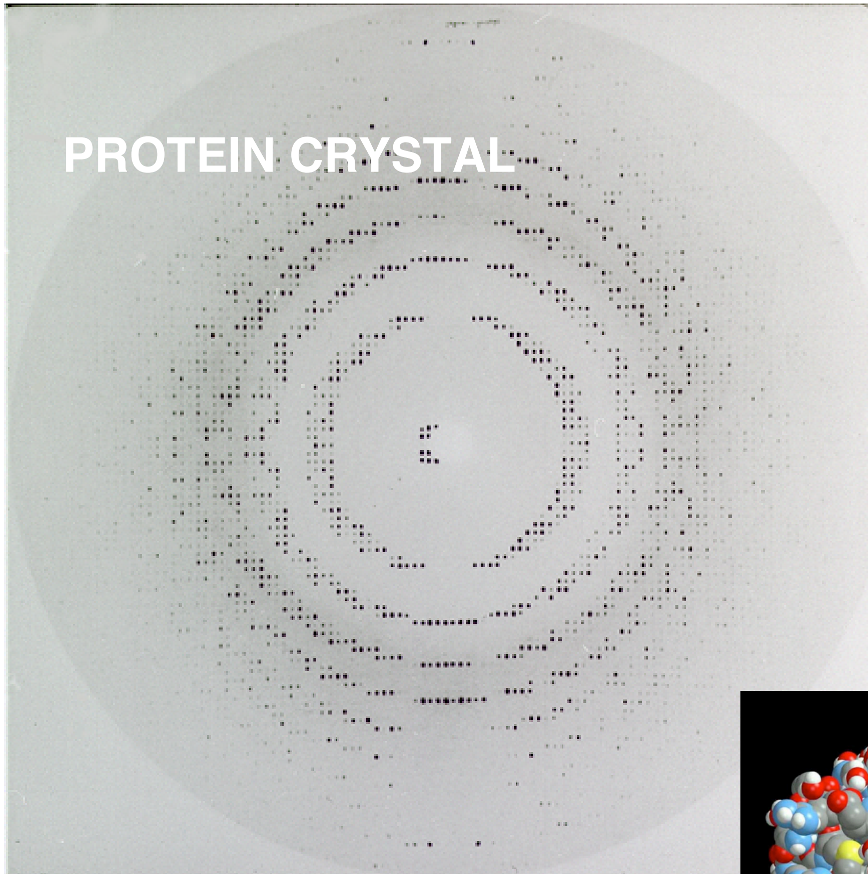
With LCLS a speckle pattern can be recorded in a single shot, opening the door for femtosecond dynamics on the nanoscale. Inversion of the magnetic speckle pattern, using techniques such as oversampling, also promise to give ultrafast real space images of nanostructures. Besides magnetic thin films, systems of interest include various materials undergoing phase transitions, simple and complex fluids or glasses, and correlated materials with complex charge and spin ordering dynamics. These measurements on the nanometer length scale are not only scientifically interesting, but they also constitutes the competitive arena of future technological devices.

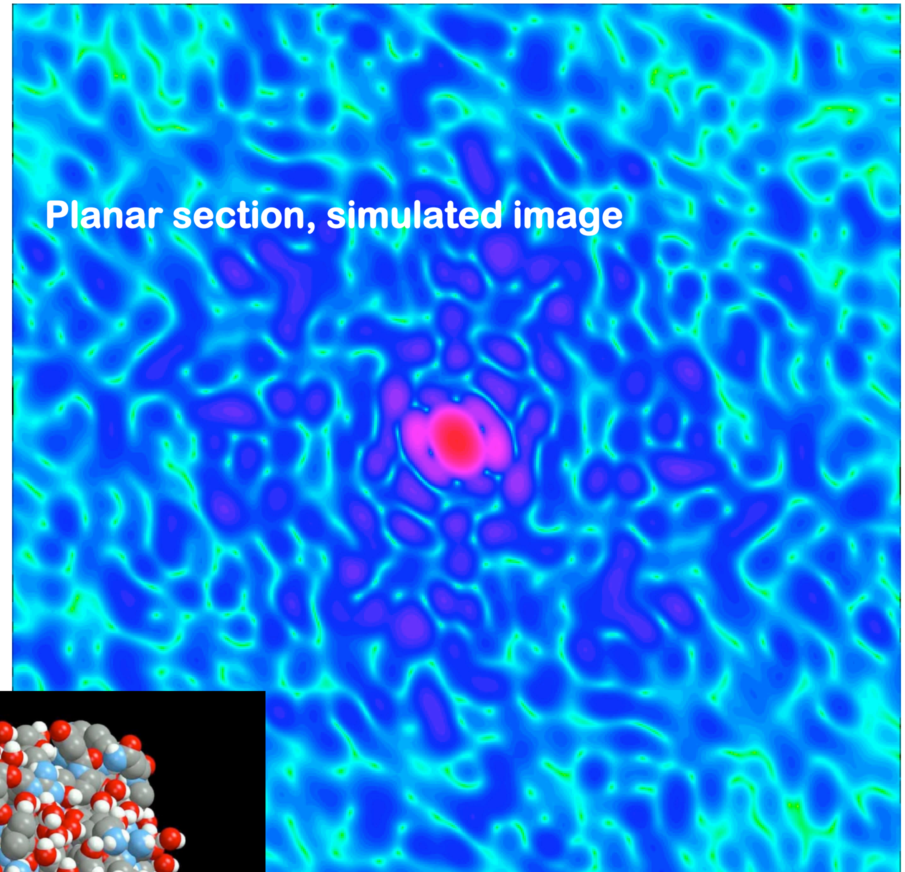
LCLS Experimental Program: Structural biology

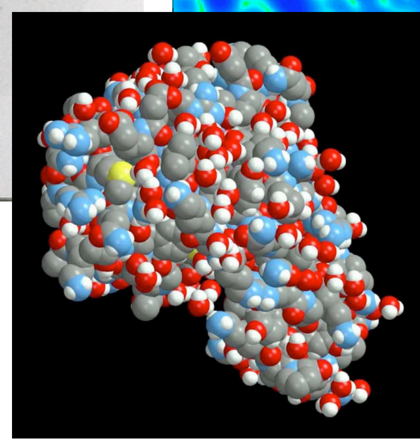
LCLS also holds great promise for structural biology. Today, radiation damage is one of the main obstacles in determining the structure of proteins that cannot be crystallized, like cell membrane proteins, which constitute nearly half of all proteins. With the peak brightness of LCLS the structure of a virus or even a single protein molecule may be determined by recording a three dimensional array of ultrafast diffraction patterns, each recorded in a single shot on a new sample before radiation damage sets in.


Coulomb Explosion of Lysozyme (50 fs)
LCLS
Radiation damage interferes with atomic positions and the atomic scattering factors

Scattering by a Crystal and by a Single Molecule (Janos Hajdu)





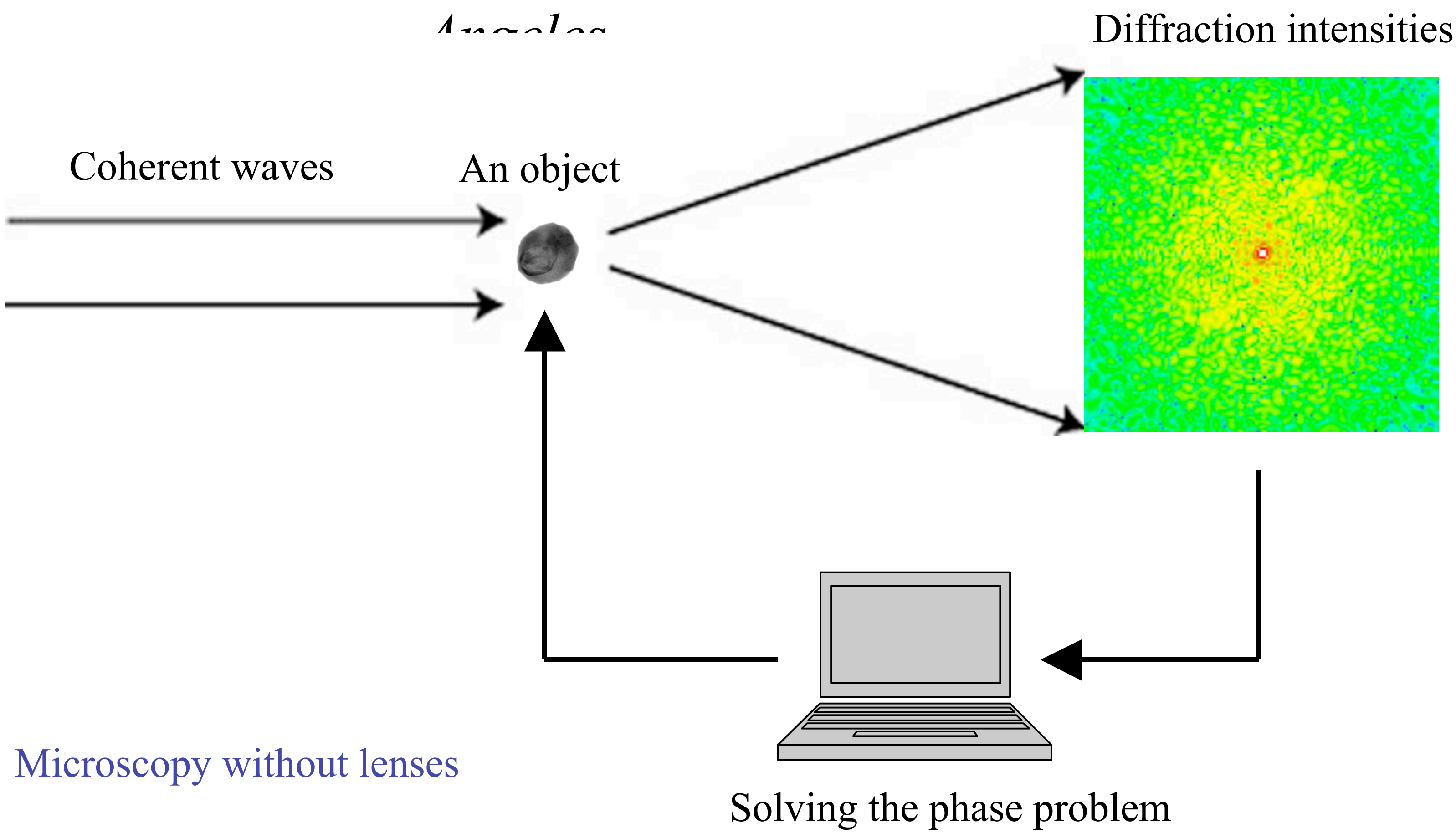


Max. resolution is a function of crystal quality

Max. resolution does not depend on sample quality

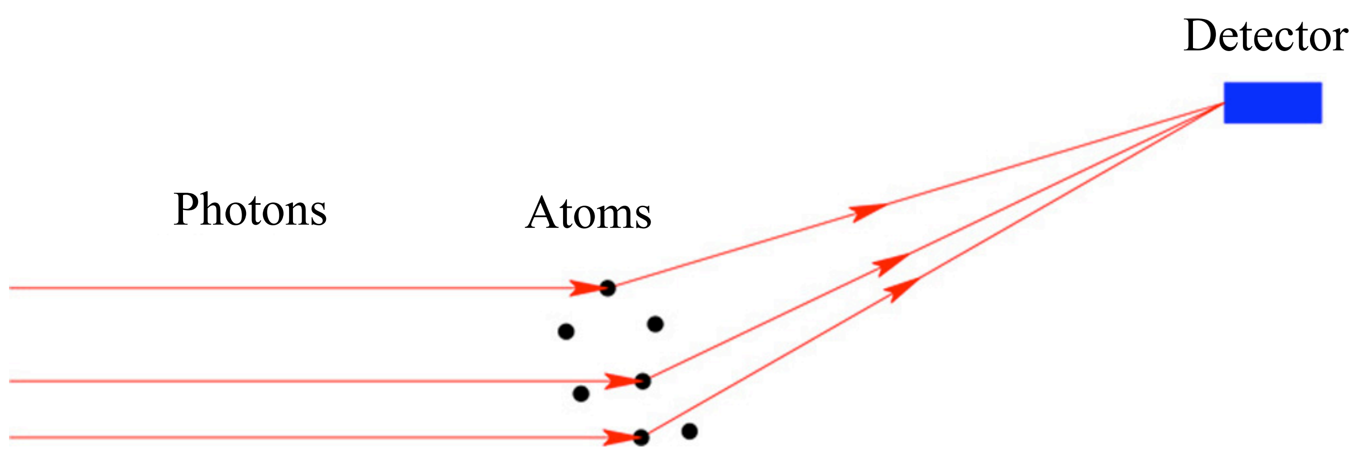
3D Coherent X-ray Diffraction Microscopy

Jianwei (John) Miao *University of California, Los Angeles*



Microscopy without lenses

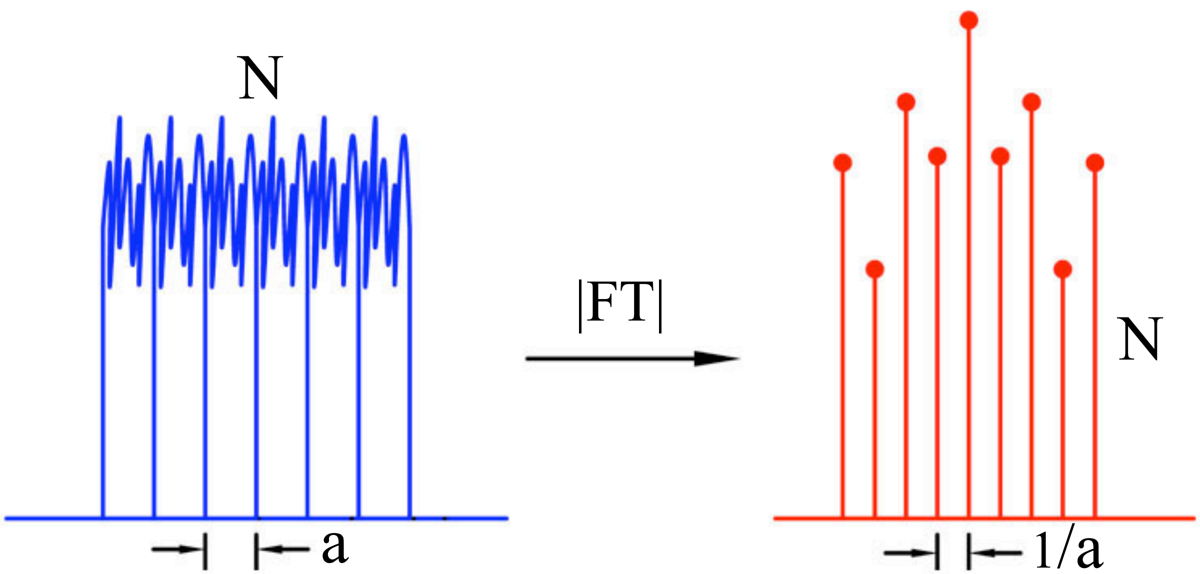
The Phase Problem: A Coherence Effect



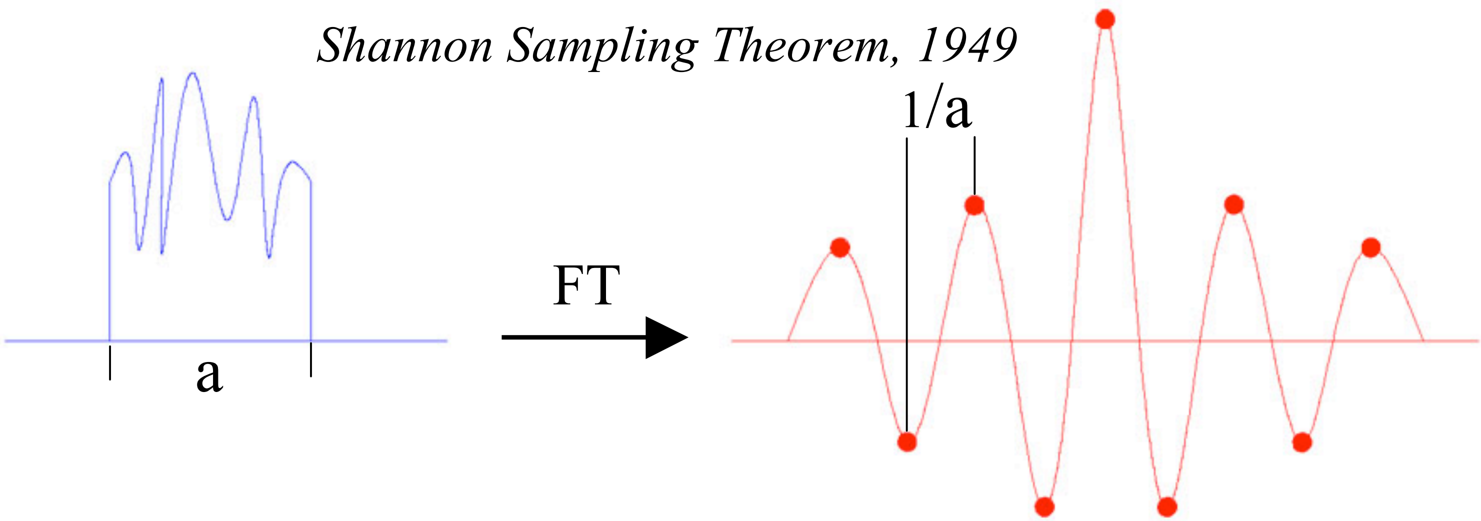
The phase problem is due to the fact that there is no way to distinguish where each photon is scattered from.

X-ray Diffraction by Crystals

Laue, W. Bragg and L. Bragg, 1912

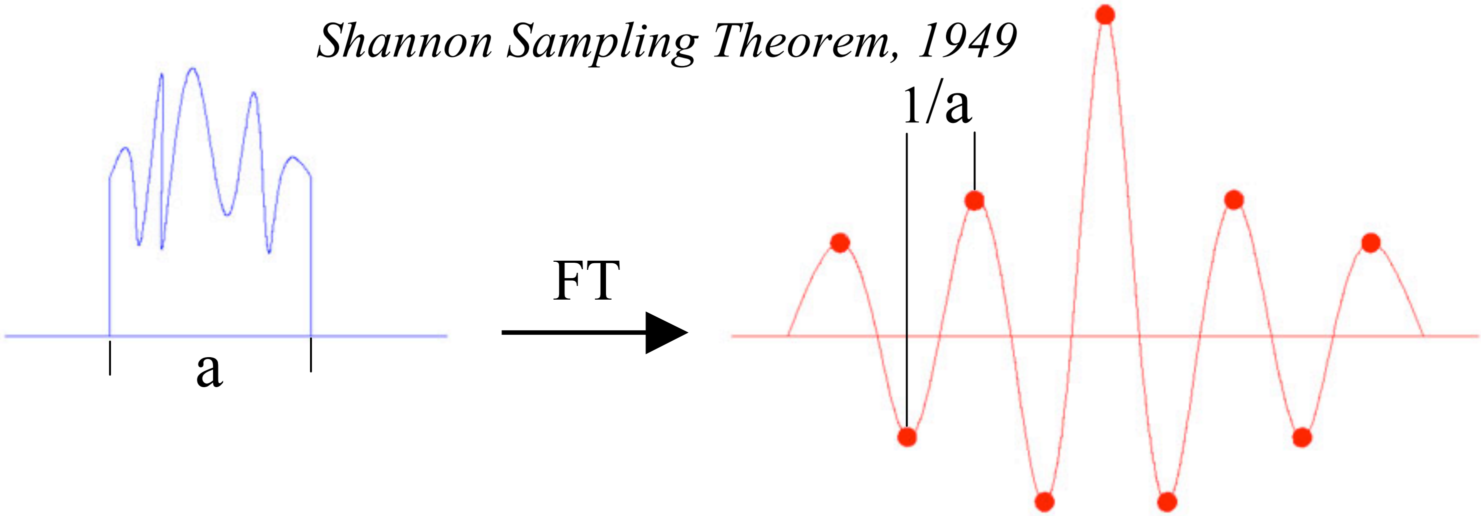


Shannon Sampling vs. Bragg-peak Sampling

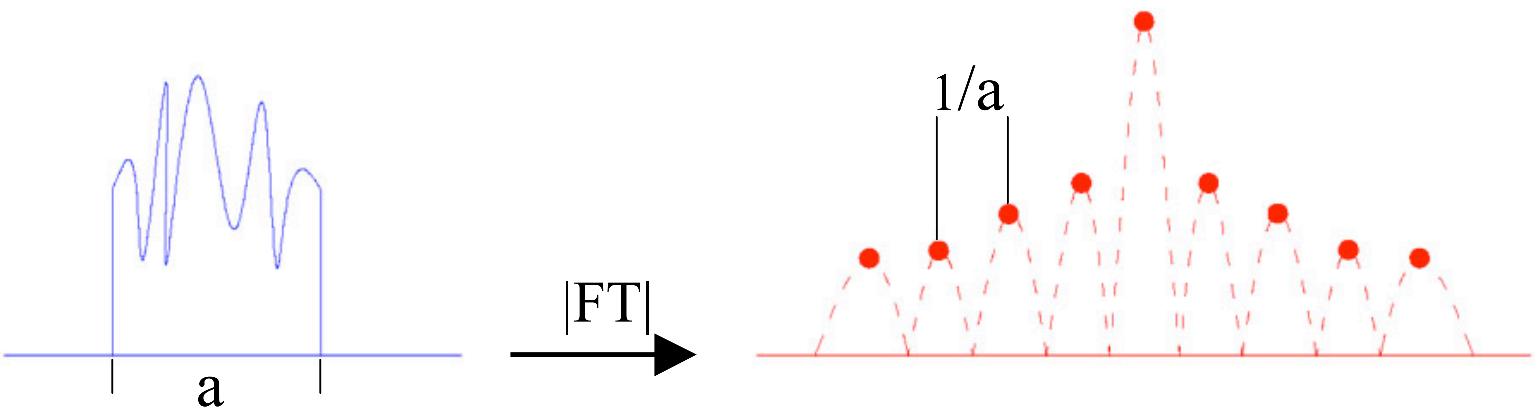


Shannon Sampling vs. Bragg-peak Sampling

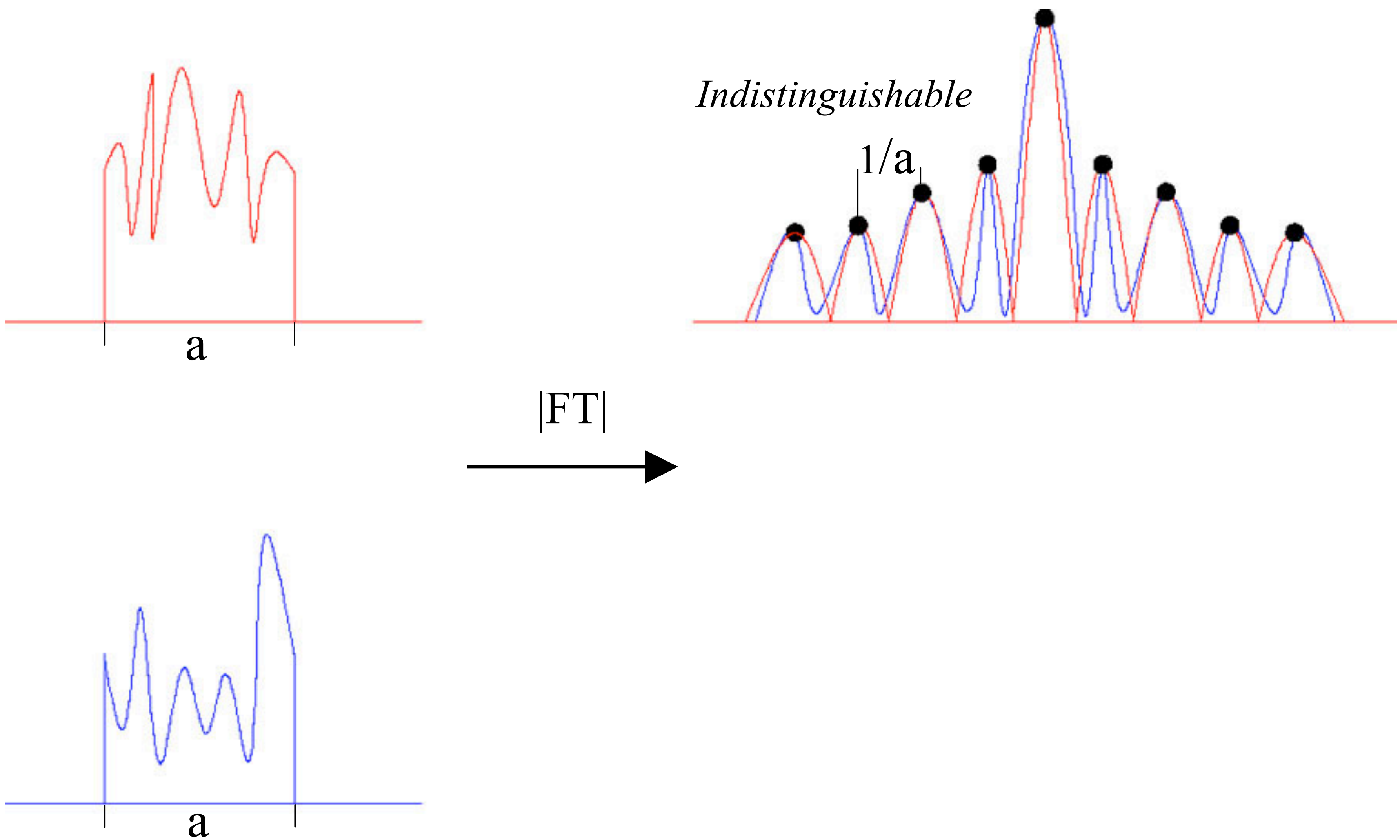
Shannon Sampling Theorem, 1949



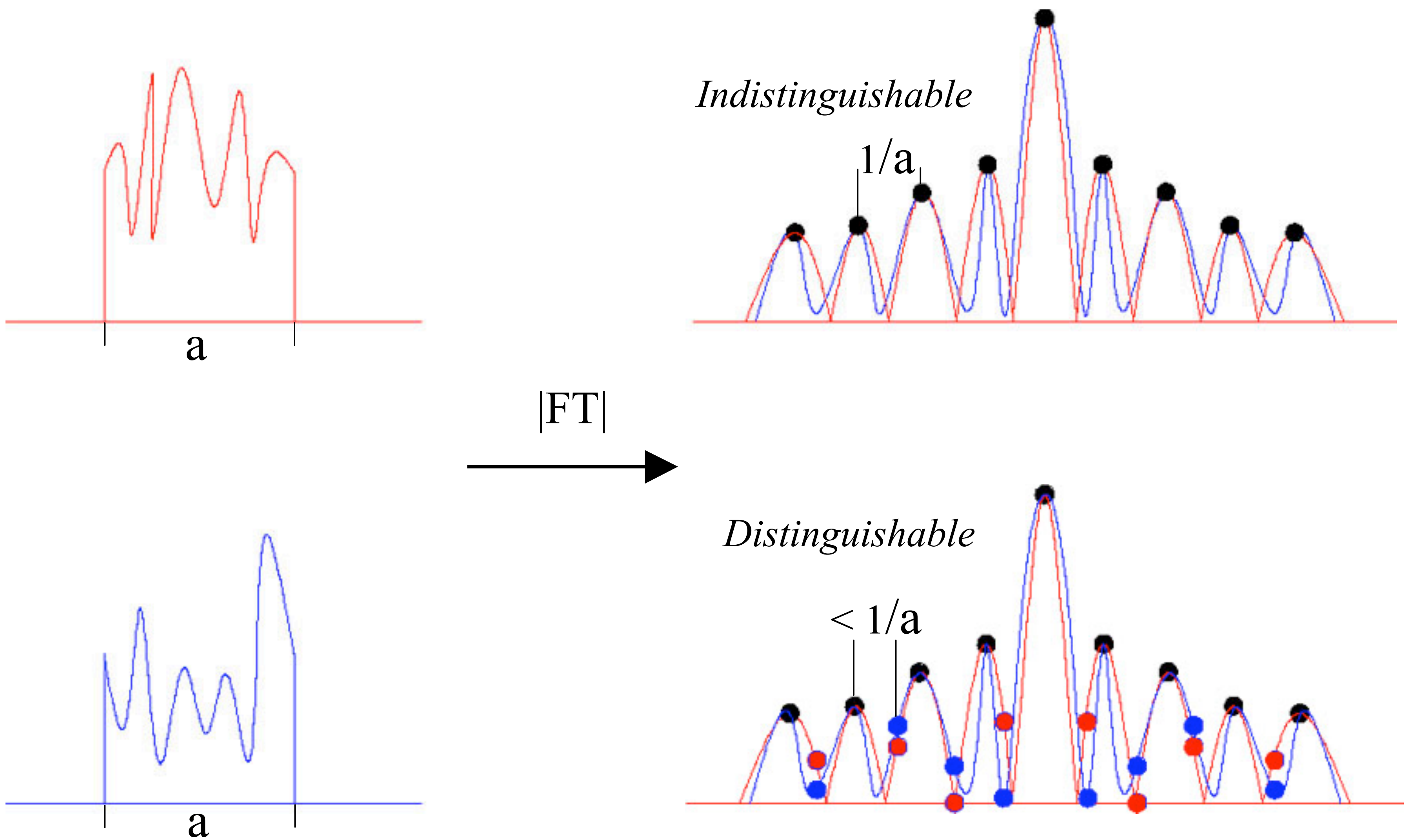
Bragg-peak sampling, 1912



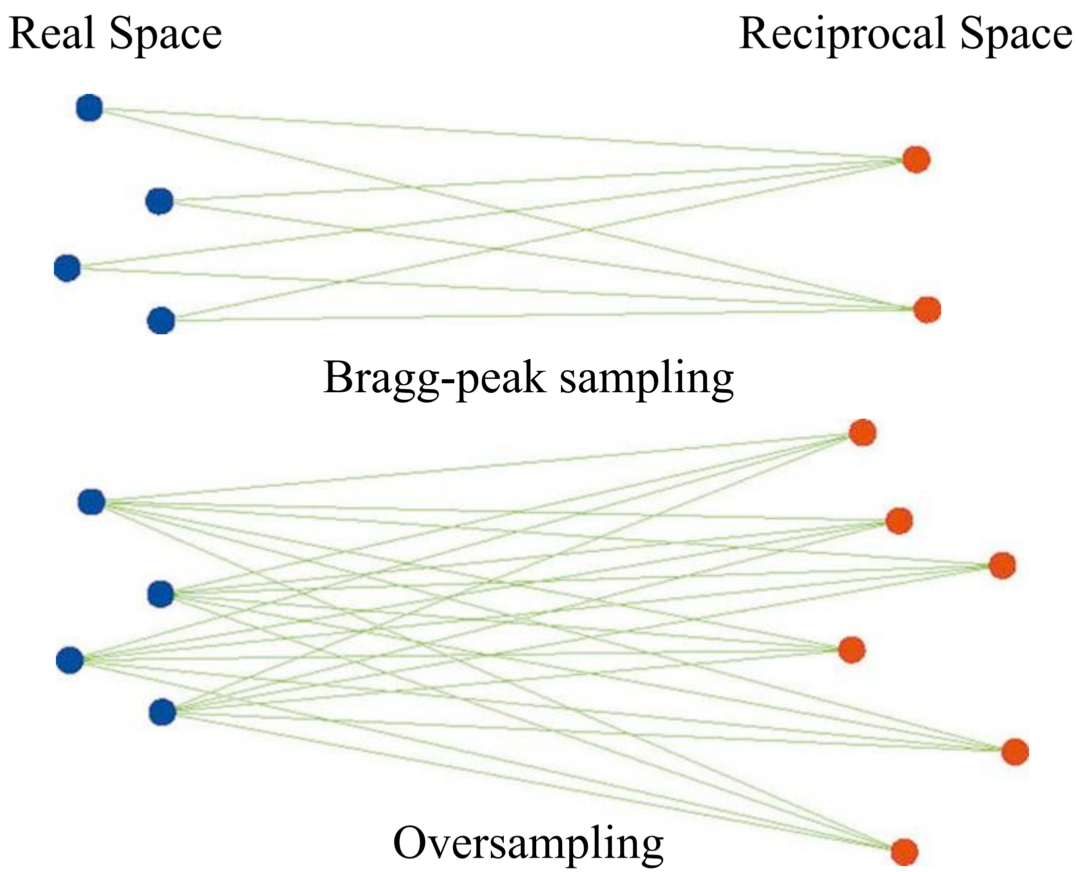
Bragg-peak Sampling vs. Oversampling



Bragg-peak Sampling vs. Oversampling



The Physical Explanation to the Oversampling Method



Better coherence ⇒ More correlated intensity points ⇒ Phase information

Miao, Sayre & Chapman, *J. Opt. Soc. Am.* A **15**, 1662 (1998).

The Iterative Algorithm

Real Space   Reciprocal Space

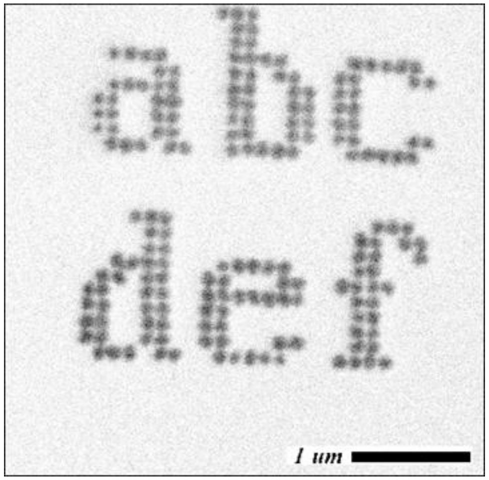
$$\begin{tikzcd}
\rho_j^{'}(\mathbf{r}) \arrow[d, "\mathbf{B}"] & & F_j^{'}(\mathbf{k}) \arrow[ll, "FFT^{-1}"'] \\
\rho_j(\mathbf{r}) \arrow[rr, "FFT"] & & F_j(\mathbf{k}) \arrow[u, "\mathbf{A}"']
\end{tikzcd}$$

A: $$F_j^{'}(\mathbf{k}) = \left| F_{\exp}(\mathbf{k}) \right| \times e^{\varphi_{j-1}(\mathbf{k})}$$

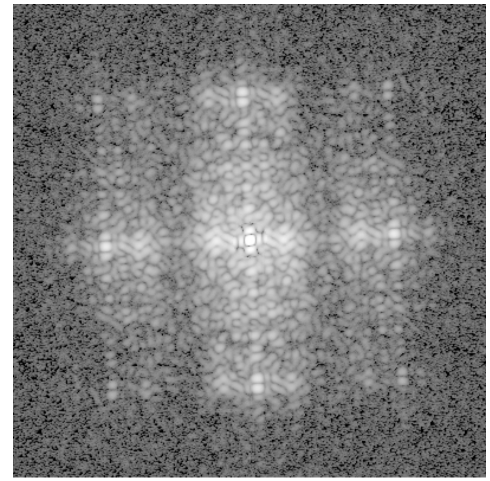
B: $$\rho_j(\mathbf{r}) = \begin{cases} \rho_j^{'}(\mathbf{r}) & if\ \mathbf{r} \in S \cap \rho_j^{'}(\mathbf{r}) \geq 0 \\ \rho_{j-1}(\mathbf{r}) - \beta \times \rho_j^{'}(\mathbf{r}) & if\ \mathbf{r} \notin S \cup \rho_j^{'}(\mathbf{r}) < 0 \end{cases}$$

Fienup, *Opt. Lett.* 3, **27** (1978).

The First Experimental Demonstration

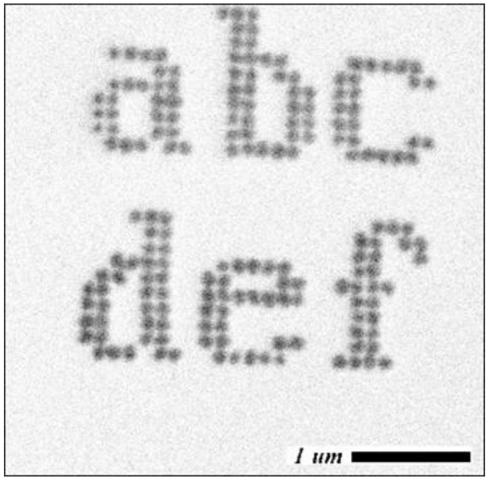


(a) A SEM image



(b) An oversampled diffraction pattern (in a logarithmic scale) from (a).

The First Experimental Demonstration



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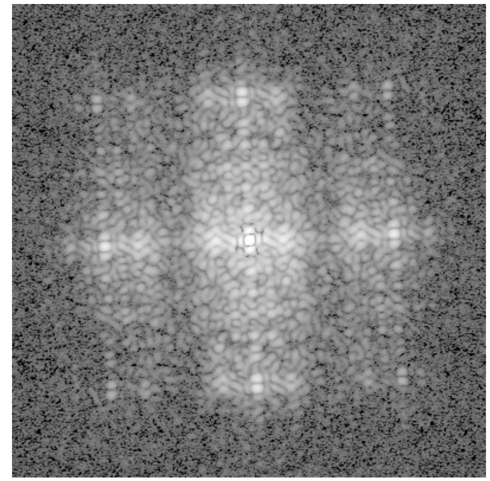
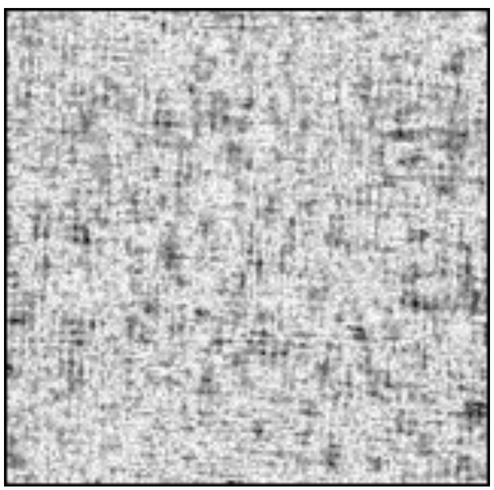
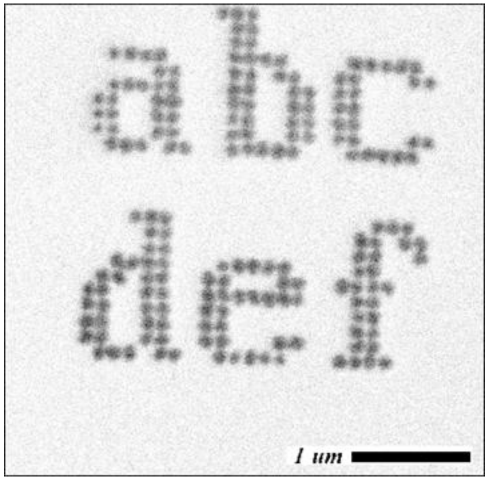


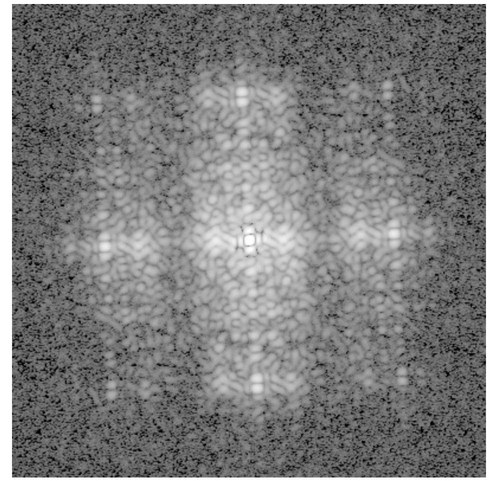
(b) An oversampled diffraction pattern
(in a logarithmic scale) from (a).



The First Experimental Demonstration



(a) A SEM image

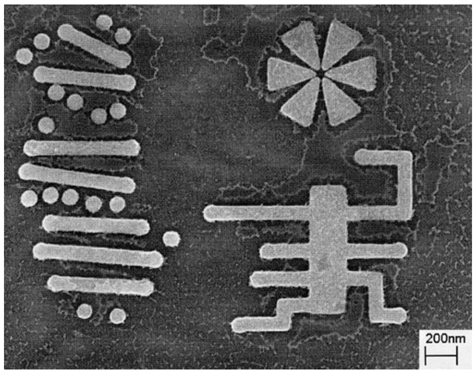


(b) An oversampled diffraction pattern (in a logarithmic scale) from (a).

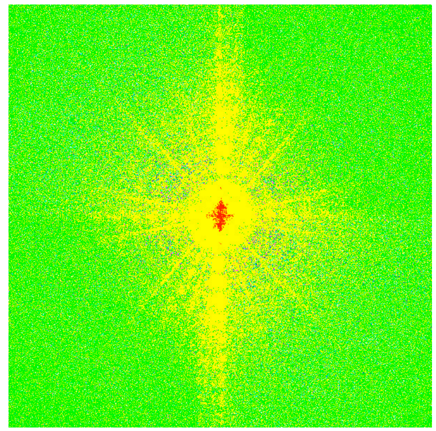
(c) An image reconstructed from (b).

Miao, Charalambous, Kirz & Sayre, *Nature* **400**, 342 (1999).

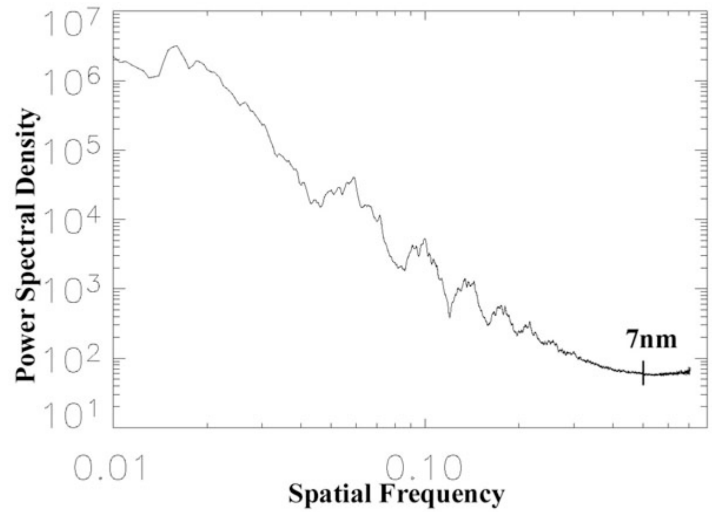
Imaging Nanostructures at 7 nm Resolution



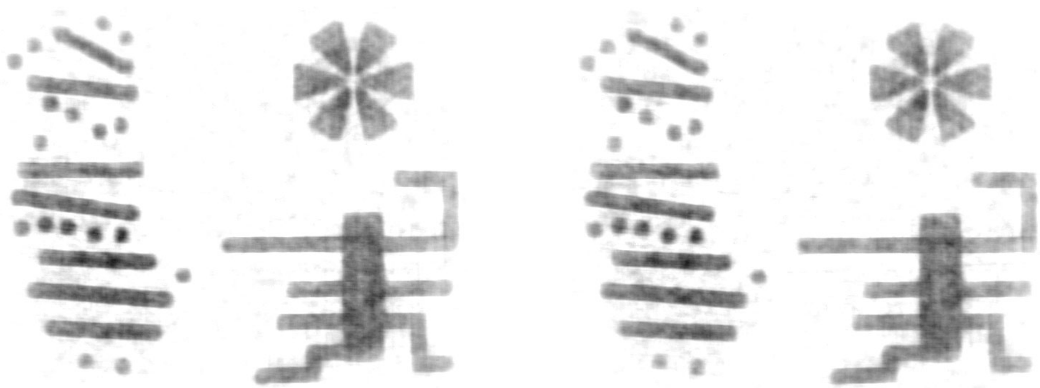
(a) A SEM image of a patterned sample made of Au



(b) A coherent diffraction pattern from (a)

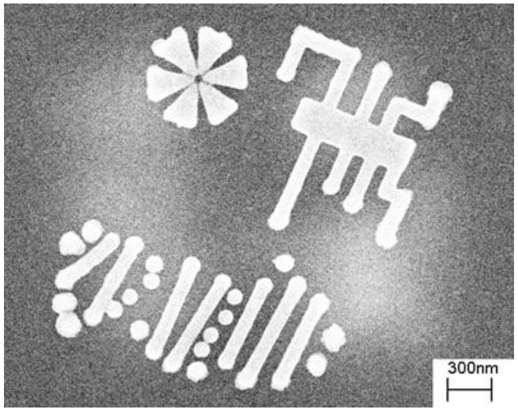


(c) Power spectral density of the diffraction pattern

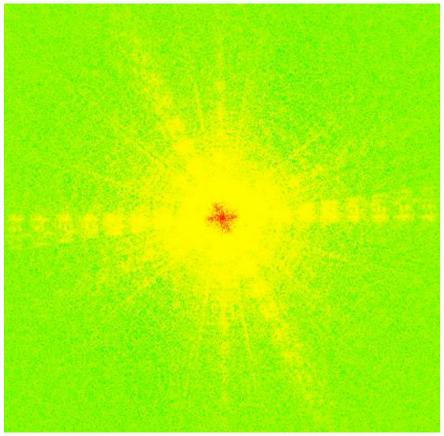


(d) Images reconstructed from (b) with two different initial seeds.

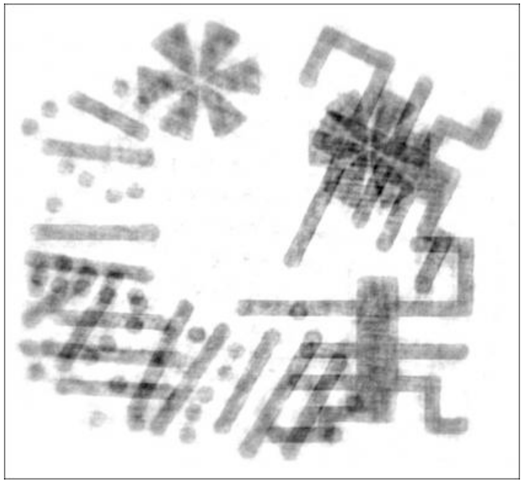
3D Imaging of a Nanoscale Material



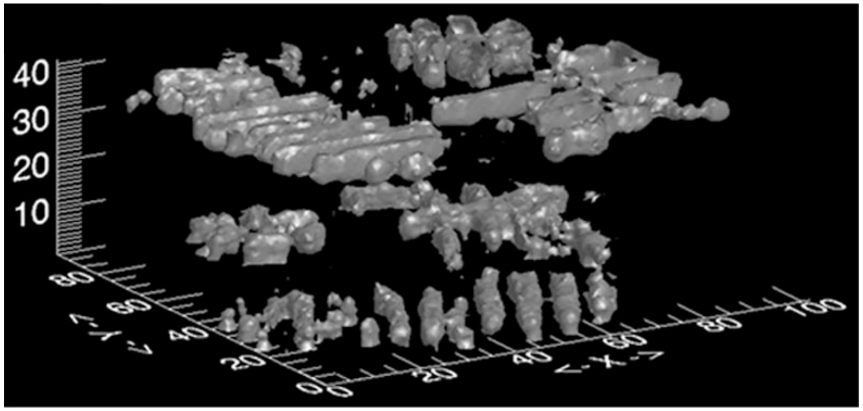
(a) A SEM image of a double-layered sample made of Ni (~2.7 x 2.5 x 1 μm^3)



(b) A coherent diffraction pattern from (a)



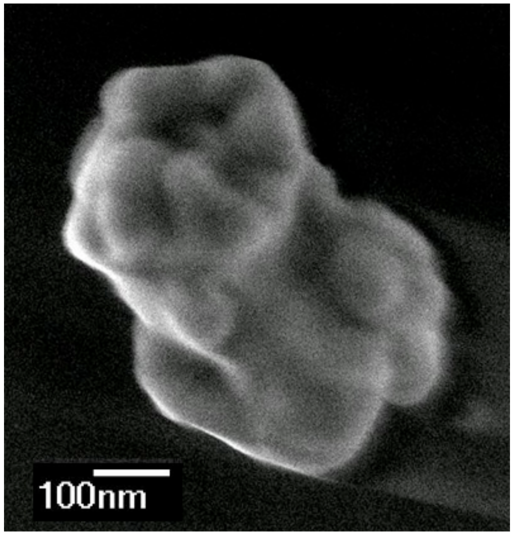
(c) An image reconstructed from (b)

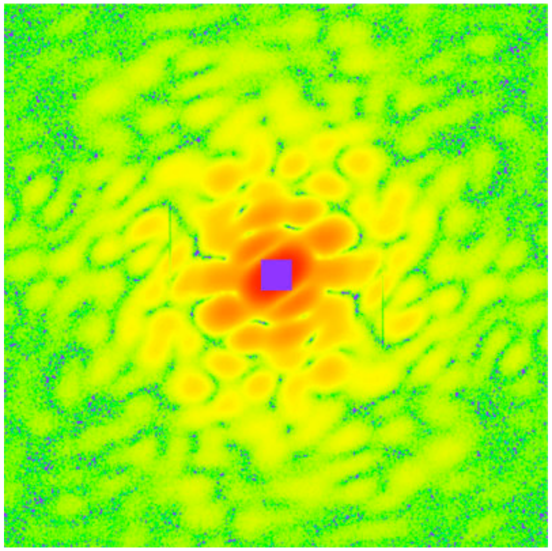


(d) An iso-surface rendering of the reconstructed 3D structure

Miao *et al*., *Phys. Rev. Lett.* **89**, 088303 (2002).

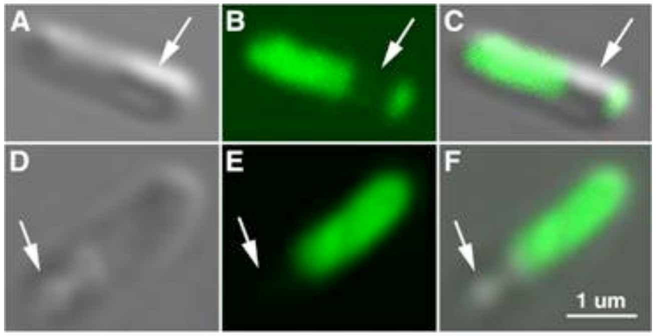
3D Imaging of a Single GaN Quantum Dot Nanoparticle



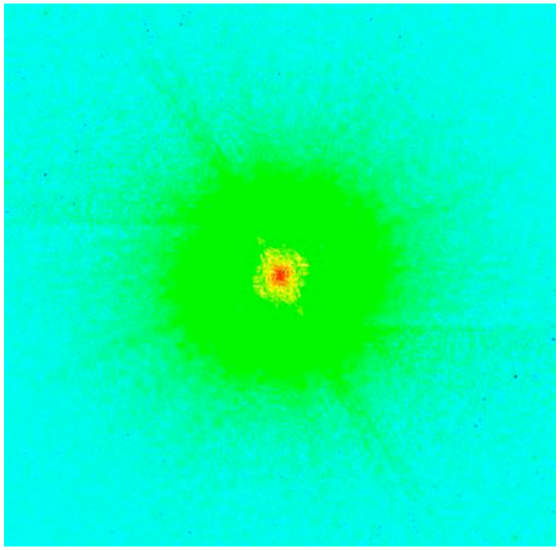


Miao *et al*., *Phys. Rev. Lett.* **95**, 085503 (2005) .

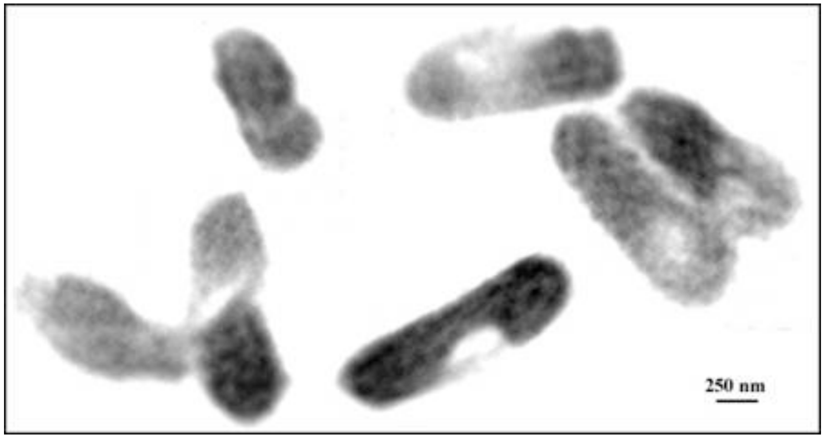
Imaging *E. Coli* Bacteria



(a) Light and fluorescence microscopy images of *E. Coli* labeled with manganese oxide



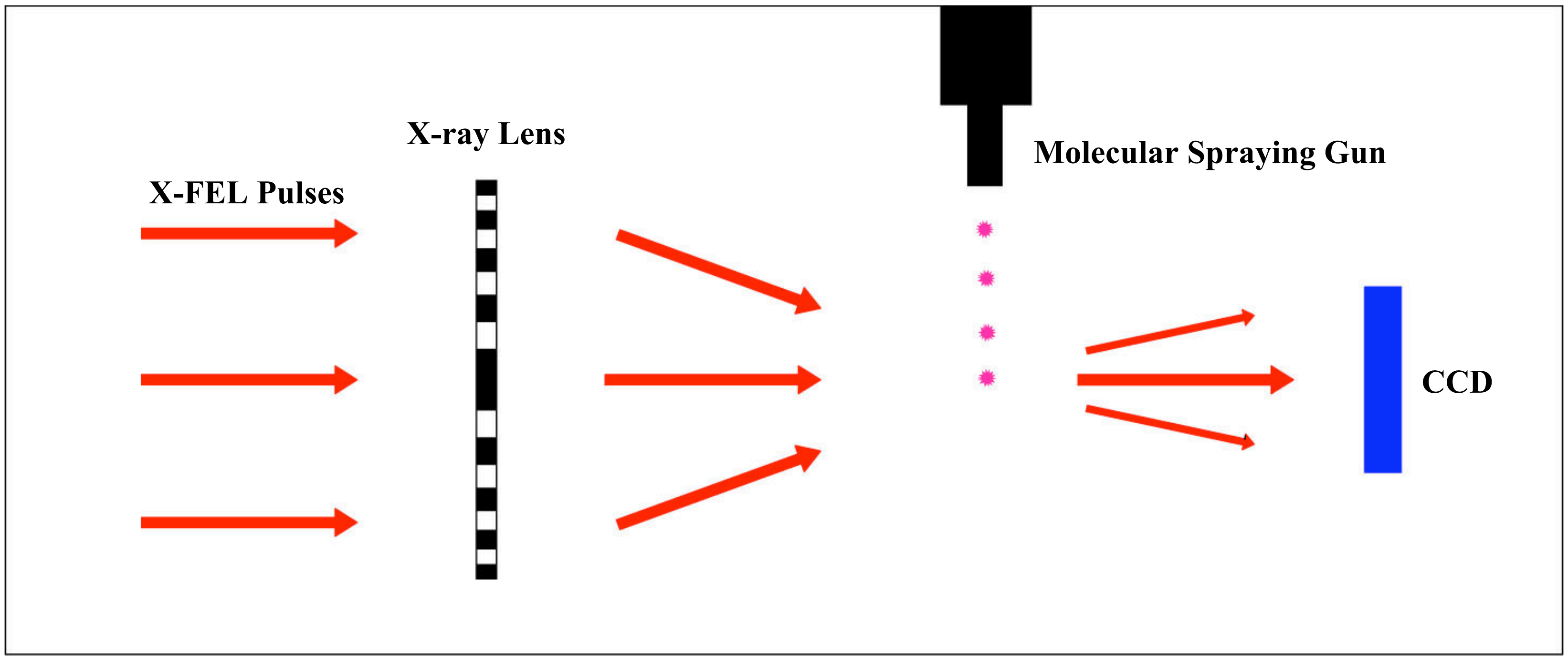
(b) A coherent X-ray diffraction pattern from *E. Coli*



(c) An image reconstructed from (b).

Miao *et al*., *Proc. Natl. Acad. Sci. USA* **100**, 110 (2003).

A Potential Set-up for Imaging Single Biomolecules Using X-FELs

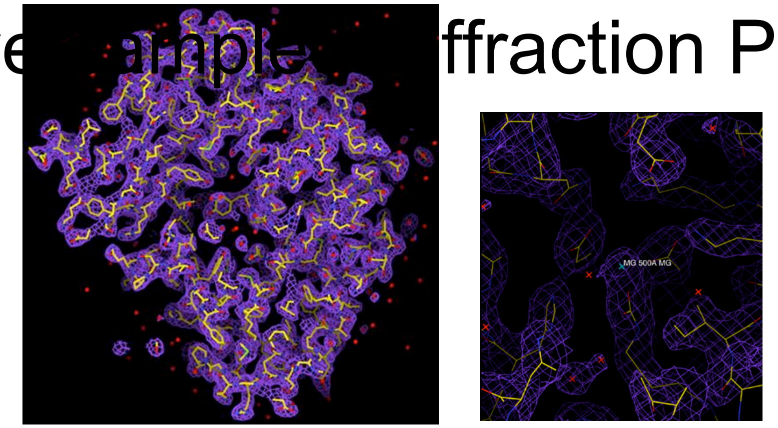


Radiation Damage

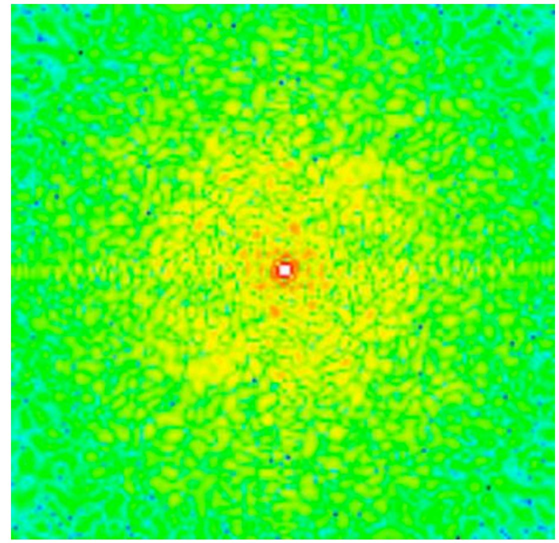
Solemn & Baldwin, *Science* 218, 229 (1982). Neutze *et al*., *Nature* 400, 752 (2000).

When an X-ray pulse is short enough (< 50 fs), a 2D diffraction pattern may be recorded from a molecule before it is destroyed.

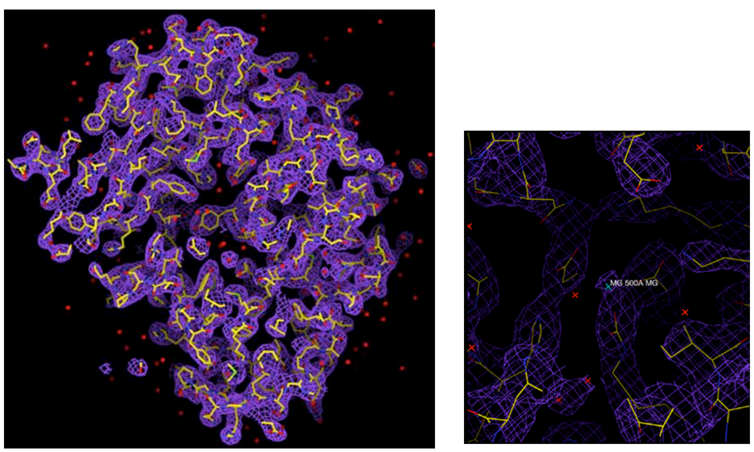
Electron Density Reconstruction of Rubisco Molecules from Simulated Oversampled Diffraction Patterns



(a) The 3D electron density map of a rubisco molecule and its active site (from PDB)



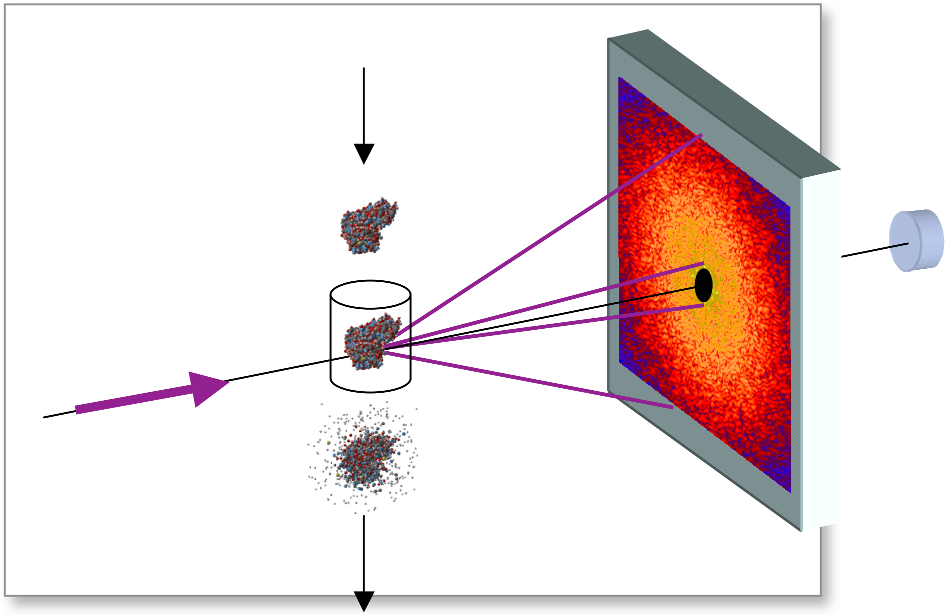
(b) A section of the oversampled 3D diffraction pattern with Poisson noise, assembled from 3 x 10^5 simulated 2D diffraction patterns.

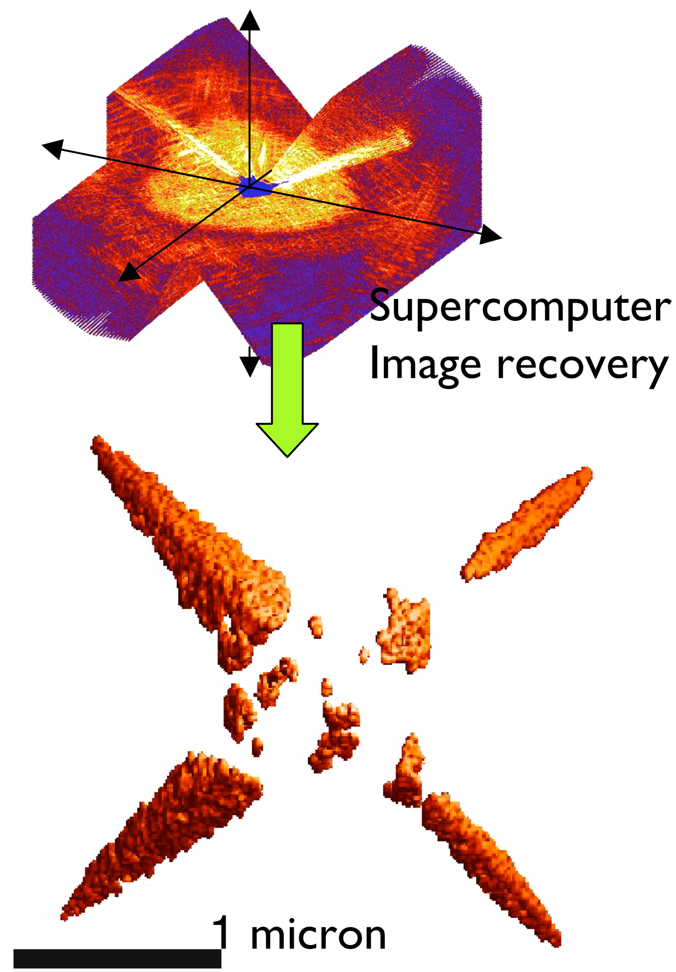


(c) The reconstructed 3D electron density map

Miao, Hodgson, Sayre, *Proc. Natl. Acad. Sci. USA* 98, 6641 (2001).

XFELs will enable 3D atomic-resolution imaging





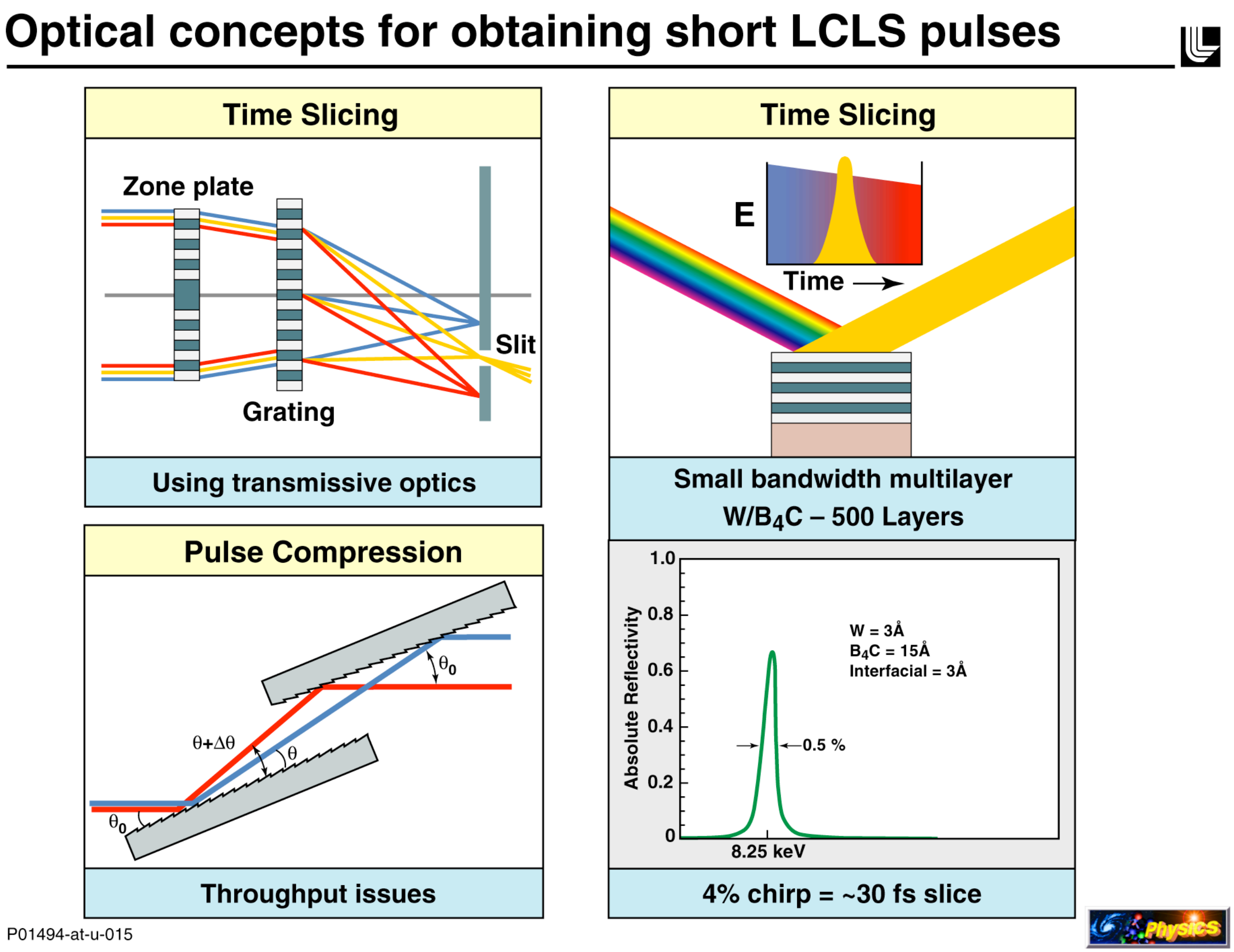
- Ultra-fast X-ray pulses will allow us to measure diffraction from single molecules. The fluence required will destroy the molecule, but we freeze this motion with a short pulse
- There are no atomic-resolution lenses for X-rays. We generalize crystallography to non-periodic structures. We have demonstrated this with the world's highest resolution 3D images of non-crystalline material

Courtesy Henry Chapman, LLNL

Short bunches: Very Important!

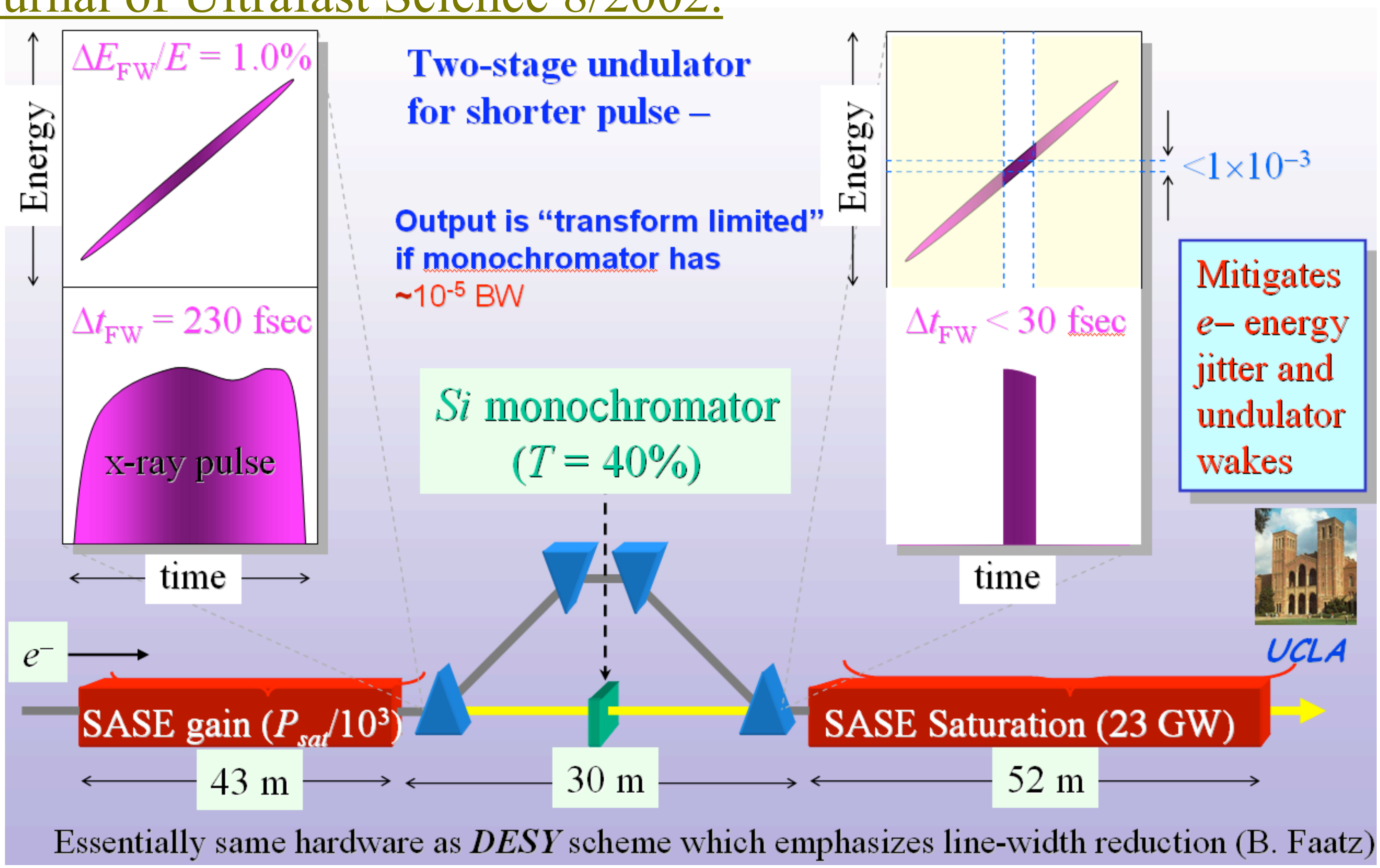
In its initial configuration the full width LCLS pulse duration is about 200 femtoseconds. Even though this is hundred times shorter than in storage rings, it can be further reduced to about 1/10 of this value or less. Several schemes to achieve ultrashort pulses have been proposed. Some use an energy-longitudinal position correlation in the electron bunch, and, as a result, in the X-ray pulse. A short X-ray pulse is then obtained by slicing out part of the bunch with a monochromator.
Other methods change the current or emittance distribution along the electron bunch to obtain lasing only in a short part.

Some optical concepts to obtain short pulses from energy chirped electron beam.



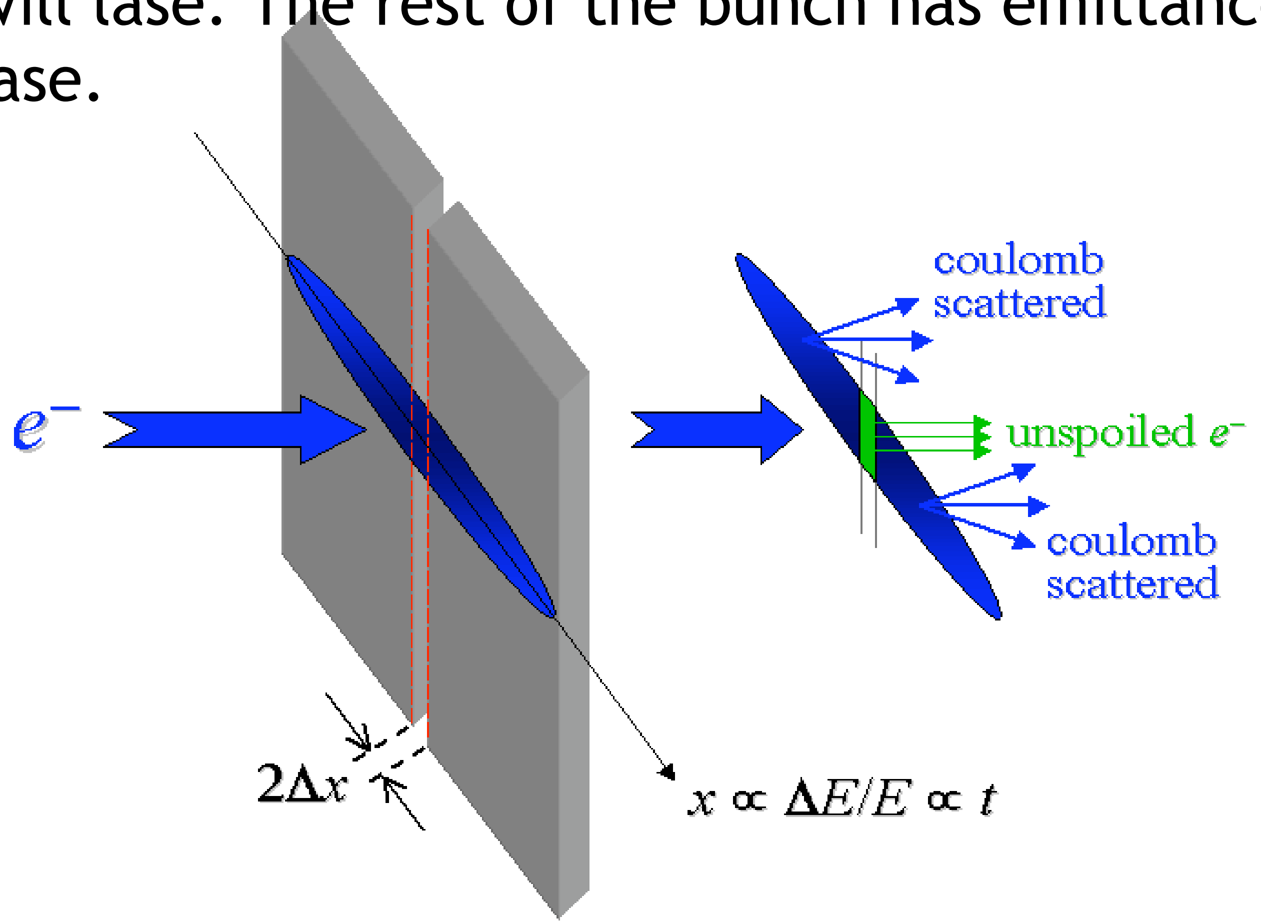
Two-Stage Chirped-Pulse Seeding in LCLS

C. Schroeder, J. Arthur, P. Emma, S. Reiche, and C. Pellegrini, JOSA B 19,, 1782-1789, (2002). Virtual Journal of Ultrafast Science 8/2002.

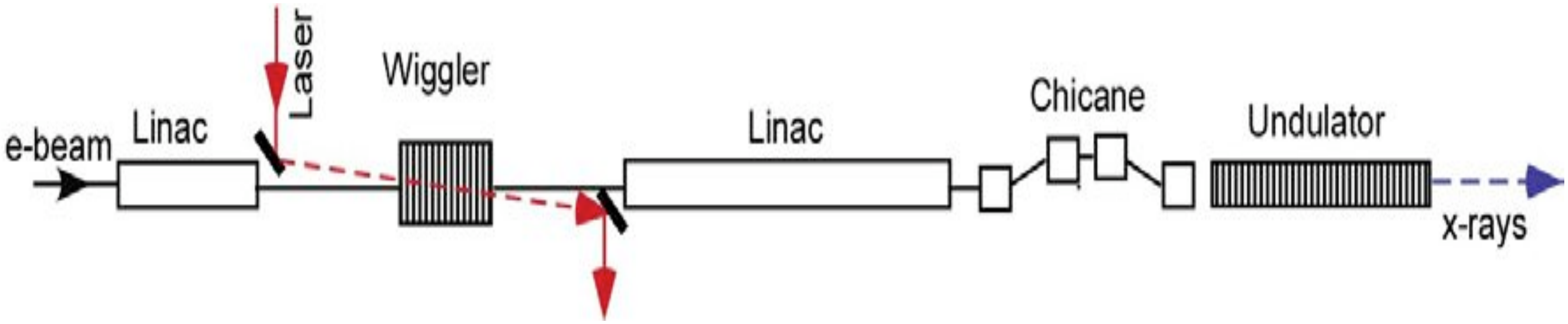


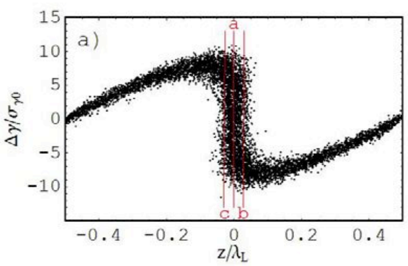
Slotted spoiler method to produce femtosecond pulses. P. Emma, Z. Huang, et al., Ph. Rev. Lett. 2004

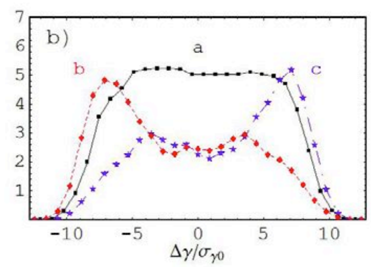
Slotted spoiler at the center of a chicane leaves a narrow, un-spoiled beam center, which has small emittance and will lase. The rest of the bunch has emittance too large to lase.

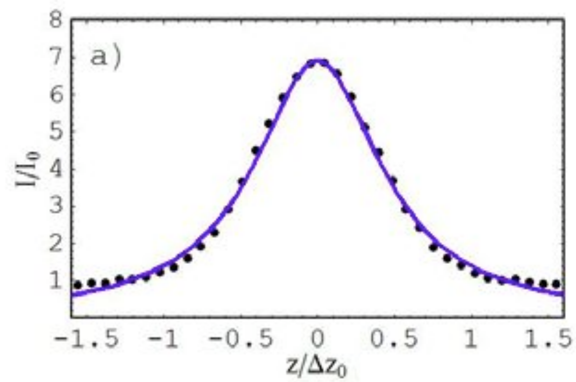


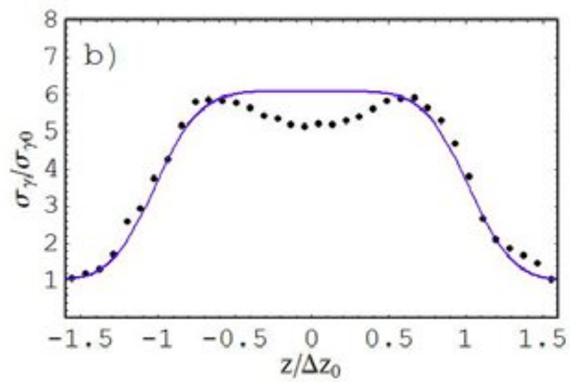
Enhanced SASE. A. Zholents, LBNL55938 and PRL



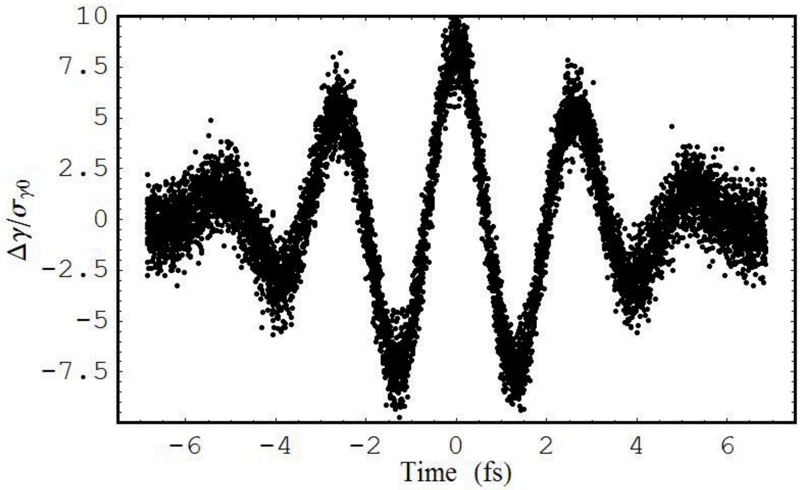








ESASE at LCLS (Zholents)



Modulating Laser at wavelength 2.2 mm, power 6 GW

Modulator magnet with ten periods, 16 cm long, field 2T (K=29),at E=2GeV..

FEL parameter, of modulated beam 8x10^{-4},twice as large as the non modulated beam.

FEL radiation at 0.15 nm, with Peak power of 230 GW and pulse length of 0.2 fs.

Conclusions

- The progress in the physics and technology of particle beams, and the exploitation of the FEL collective instability, has made possible to design and build a powerful X-ray FELs in the 1Å spectral region.
- The unique characteristics of the X-ray pulse will open new areas of research in physics, chemistry and biology.
- R&D work needs to be done in areas like X-ray optics, synchronization of the X-ray probe pulse with a pump pulse, and detectors to fully exploit the potential of the system.