

X-ray Free-Electron Lasers

by CLAUDIO PELLEGRINI & JOACHIM STÖHR

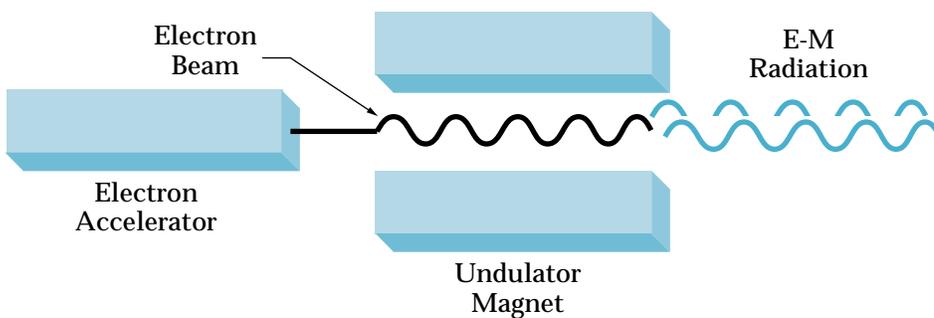
*With advances
in technology,
it is now possible
to realize the
dream of a fully
coherent X-ray
laser.*

CREATING MATTER from the vacuum, taking an atomic scale motion picture of a chemical process in a time of a few femtoseconds ($1 \text{ fs} = 10^{-15} \text{ sec}$) or unraveling the complex molecular structure of a single protein or virus. These are some of the new exciting experiments envisioned with a novel radiation source, the X-ray free-electron laser (FEL).

John Madey and collaborators built the first FEL in the 1970s. It is a powerful and challenging combination of particle accelerator and laser physics and technology. Until recently FELs have been operating at infrared or near ultraviolet wavelengths. A combination of theoretical, experimental, and technological advances has made possible their extension to the X-ray region. X rays have allowed us to see the invisible for almost a century. With their help we have been making great progress in understanding the properties of materials and of living systems. Today the best sources of X rays utilize synchrotron radiation from relativistic electron beams in storage rings. The most advanced, so called third generation facilities, combine storage rings and undulator magnets, and are used by thousands of scientists around the world.

USEFUL AS THEY ARE these facilities have limitations. The minimum X-ray pulse duration is about 50 picoseconds ($1 \text{ ps} = 10^{-12} \text{ sec}$), and the number of photons that one can focus on a small sample is limited. Future experiments will need X-ray pulses with a

- Principles, Properties and Applications



Schematic representation of a SASE free-electron laser.

large number of photons focused on a sample as small as a molecule, squeezed in a time a thousand times shorter, to study the dynamics of atomic and molecular processes. X-ray free-electron lasers can satisfy these requirements, as shown in the table.

AN FEL CONSISTS OF A LINEAR accelerator followed by an undulator magnet, as shown in the figure. The undulator has a sinusoidal magnetic field of period λ_w , usually a few centimeters, and amplitude B_w , typically about 1 tesla. In this field a single electron moves along a sinusoidal, oscillating trajectory, and radiates an electromagnetic wave-train with a number of waves equal to the number of undulator periods, N_w . The wavelength of this spontaneous radiation is equal to the undulator period reduced by a relativistic contraction factor inversely proportional to the square of its energy. This makes it easy to shorten the wavelength by increasing the electron beam energy, and for GeV electron beams one can produce

Undulator Radiation Characteristics

Some typical characteristics of the undulator radiation from third-generation ring-based light sources, and free-electron laser light sources. The emittance is in nm rad; the pulse length in picoseconds; the average and peak brightness are in photons/sec/mm²/mrad²/0.1% bandwidth; the peak power in watts.

	<i>Third Generation</i>	<i>SASE-Free-Electron Laser</i>	<i>Short pulse SASE-Free-Electron Laser</i>
Wavelength range, nm	1-0.1	1-0.1	1-0.1
Emittance, nm-rad	2	0.03	0.03
Pulse length, ps	15-30	0.06	0.01
Average brightness	10 ²⁰	10 ²²	10 ²¹
Peak brightness	10 ²³	10 ³³	10 ³³
Peak power, W	10 ³	10 ¹⁰	10 ¹⁰

wavelengths of about 1 Å, the size of an atom. The radiation is within a narrow wavelength band, equal to the inverse of the number of undulator periods, which for an X-ray FEL can be a few thousand. The radiation within this energy band is very well collimated, with an opening angle of a few microradians. The number of photons spontaneously emitted from one electron within this energy band and angle is however rather low, about one photon per 100 electrons. In an FEL the electron beam has a large number of electrons, about 10⁹ to 10¹⁰. In this case a phenomenon of self-organization of the electrons, known as the FEL collective instability, can greatly increase the number of photons per electron and the total X-ray intensity. This instability transforms an electron beam with a random electron position distribution into one in which the electrons are clustered in groups regularly

spaced at about 1 Å, producing what could be called a 1-dimensional relativistic electron crystal. The radiation from this crystal has new and exciting properties.

Consider an ensemble of electrons propagating through an undulator. The total radiation field they generate is the sum of the fields generated by all electrons. When the electrons enter the undulator, there is no correlation between their position and the X-ray radiation wavelength. As a result the fields they generate superimpose at random, with a partial cancellation, as shown at the left in the next figure. What the beam produces in this case is called “spontaneous undulator radiation”, and its intensity is proportional to the number of electrons, N_e .

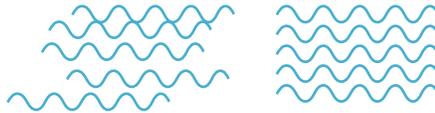
If we could order the electrons so that they were all within a fraction of the radiation wavelength, or

separated by a wavelength, the radiation fields would superimpose in phase, as shown at the right in the figure. The intensity is then proportional to the number of electrons squared, a huge gain. The effect is similar to what you would have during a large party if instead of having all the people talking casually, producing a lot of noise, you could organize them in a well performing choir, all persons singing exactly the same note in perfect tune. The results would probably break all the glasses and windows in the room!

While we do not know how to control the electron position at the Ångstrom level, we can conveniently let nature order the electrons for us, using the FEL collective instability. The way it works is:

- Electrons propagating through the undulator interact with the electromagnetic field generated by other electrons. The interaction changes their energy, and the change is modulated at the X-ray wavelength.
- Within the undulator the trajectory of electrons with larger (smaller) energy is bent less (more). As a result the electrons will bunch together within a wavelength.
- The electromagnetic fields emitted by the bunched electrons superimpose in phase, and the total field amplitude increases. Thus the electron energy change is larger, and the bunching mechanism is stronger.

The end result is that the amplitude of the electromagnetic field grows exponentially. The growth rate is called the gain length and is the most important FEL parameter.



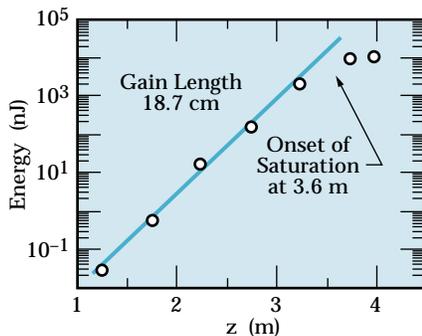
The superposition of the fields generated from many electrons; on the left is the spontaneous radiation case, on the right the free-electron laser case.

The exponential growth saturates when all the electrons are well ordered, and sing perfectly in tune!

The instability can start with the aid of an external electromagnetic field, in which case the system is a high gain FEL amplifier, or it can start from the random synchrotron radiation noise produced by the electron beam at the undulator entrance, a Self Amplified Spontaneous Emission FEL (SASE-FEL). A one-dimensional theory of a SASE-FEL, describing all its properties with a single parameter ρ , was developed in 1984 by Roldolfo Bonifacio, Claudio Pellegrini and Lorenzo Narducci.

In the X-ray region ρ is about 10^{-3} or less. This means that saturation is reached in about 1000 undulator periods, and that one thousandth of the electron beam kinetic energy is transformed into photons. Consider as an example a 1 Å X-ray SASE-FEL with 15 GeV electron energy, and $\rho \sim 10^{-3}$. In this case the number of photons per electron within the same frequency band and angle is about 1000, a gain of 100,000 over the spontaneous radiation case.

The instability develops if the electron beam has an energy spread smaller than ρ , and a radius and angular spread similar to that of the



Exponential growth and saturation in the VISA experiment at 830 nm, in a 4 m long undulator.

LCLS Parameters

ENERGY SPREAD, PULSE LENGTH, emittance are rms values. Brightness is in the same unit as the earlier table. The energy spread is the local energy spread within 2π cooperation lengths. A correlated energy chirp of 0.1 percent is also present along the bunch.

Electron Beam

Electron energy, GeV	14.3
Emittance, nm rad	0.05
Peak current, kA	3.4
Energy spread, %	0.02
Pulse duration, fs	230

Undulator

Period, cm	3
Field, T	1.32
K	3.7
Gap, mm	6
Total length, m	100

Radiation

Wavelength, nm	0.15
FEL parameter, ρ	5×10^{-4}
Field gain length, m	11.7
Bunches/sec	120
Average Brightness	4×10^{22}
Peak brightness	10^{33}
Peak power, GW	10^{10}
Intensity fluctuations, %	8

X-ray beam. The gain must also be large enough to overcome radiation losses due to diffraction. This means that as the FEL wavelength is reduced the electron beam must satisfy more stringent , and this requires avoiding damaging collective effects in the accelerator. Operating an X-ray SASE-FEL is a balancing act between controlling the unwanted collective effects in the accelerator, and letting the FEL instability develop in the undulator.

IN A WORKSHOP held at SLAC in 1992 to discuss the possibility of developing advanced, fourth generation X-ray sources, Claudio Pellegrini showed that using a novel electron source, the photoinjector, developed by John Fraser, Richard Sheffield, and Edward Gray at Los Alamos, and the new linac technologies developed for the SLAC linear collider, it was possible to build a 1Å X-ray SASE-FEL. A study group coordinated by Herman Winick developed this concept, calling it the Linac Coherent Light Source (LCLS). A design group directed by Max Cornacchia continued the project development until its approval by the Department of Energy. A group at DESY in Germany also became interested in this work and developed its own SASE-FEL project as part of the TESLA linear collider project.

The first experimental observations of the FEL instability were done in 1998 by a UCLA-Kurchatov group, which demonstrated exponential gain over about 4 gain lengths, in a 60 cm long undulator, at a wavelength of 16 mm. A much larger gain, 3×10^5 at 12 μm , was obtained in the

same year, using a 2 meter long undulator by a UCLA-Kurchatov-LANL-SSRL group. In 2000-2001 three SASE-FELs have reached a gain larger than 10^7 and saturation: LEUTL, at Argonne, at 530 and 320 nm in a 20 meter long undulator; VISA, a BNL-SLAC-LLNL-UCLA collaboration, in a 4 meter long undulator, using a beam with characteristics similar to those required for LCLS, as shown in the figure; and TTF, at DESY, at 92 nm, the shortest wavelength for an FEL, with a 15 meter undulator.

These results support the feasibility of LCLS and the TESLA X-ray FEL project. The main difference between these projects is the linear accelerator, an existing room temperature linac for LCLS at SLAC, and a future superconducting linac for TESLA. The LCLS project is scheduled to start operation in 2008. Its characteristics are given in the table.

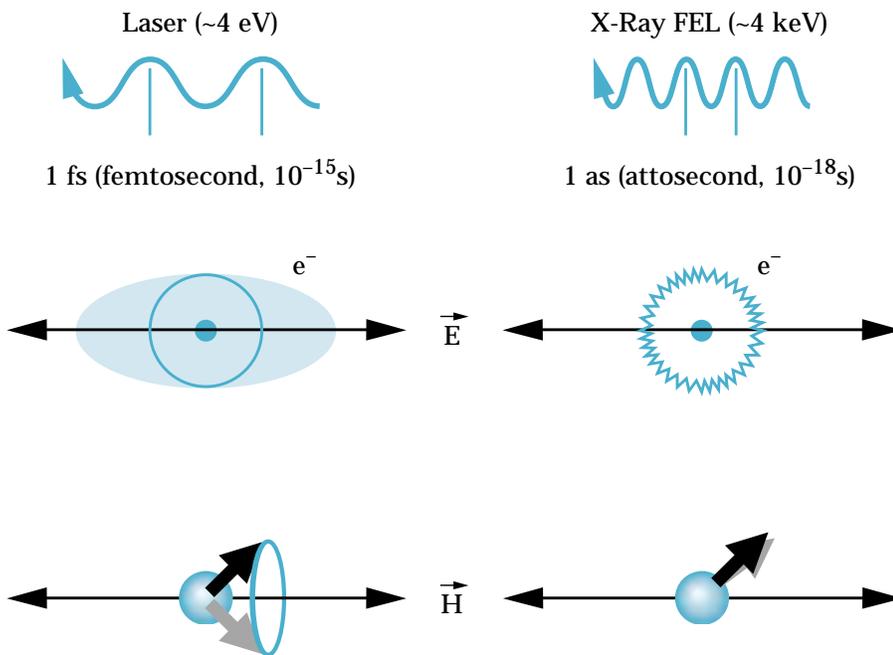
WHEN THE LCLS was first discussed, an overriding concern was the destructive power of its X-ray beam, an extraordinary beam with a peak power of about 10 GW. Wouldn't such a beam simply destroy everything put in its path, from X-ray optics to the sample itself? Traditionally X rays have been used as a gentle probe of matter, avoiding radiation damage caused by the beam itself. While some of the excitement surrounding LCLS is based on utilization of the high power to create new forms of matter, there are fortunately also ways to use its X rays as a gentle probe.

X rays, like visible light, are electromagnetic waves. In the case of light the wave oscillation period is

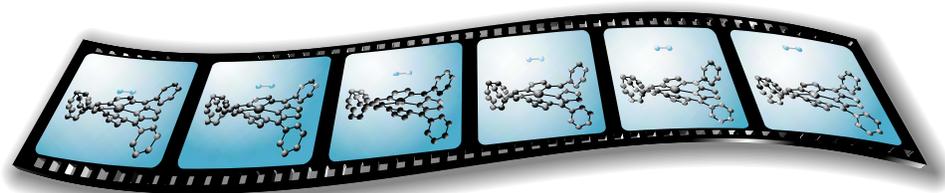
about 1 fs, while for X rays it is 1000 times faster. The LCLS beam has a peak power of 10 GW in a $100 \times 100 \mu\text{m}^2$ area near the undulator exit, corresponding to peak electric and magnetic fields of 2 volts/Ångstrom and 60 tesla, respectively. Focusing the beam by state-of-the-art methods to $1 \times 1 \mu\text{m}^2$ the field strength grows to 200 volts/Ångstrom and 6,000 tesla, strong enough to pull electrons out of atomic shells. For the focused LCLS beam the magnetic fields rival the exchange fields in ferromagnets, and would affect all magnetic interactions in matter. These numbers appear to confirm our worst fears—that nothing would survive the LCLS beam.

Fortunately, the very rapid variation of the fields with time saves the day. Just as humans cannot hear very high frequencies because our eardrums cannot follow the rapid movement of the sound waves, matter becomes insensitive to high frequency fields because the electron charge or spin cannot follow them. While the most intense visible lasers can ionize atoms by the sheer strength of the electric field, converting a material into a plasma, the thousand times faster oscillating fields of X rays only cause the electrons and spins to quiver, as illustrated in the figure.

In addition, one can minimize the interaction of the X rays with the



Distortion of the electron orbit and spin direction in an atom by strong oscillating electric and magnetic fields for typical laser and X-ray frequencies. For lasers, the field frequency is comparable to the Bohr orbit frequency of valence electrons, and the spins can precess back and forth by a large angle around the strong magnetic field. For X rays the electrons and spins cannot follow the fast fields and just quiver.



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.

mirrors and other optical elements, due mostly to the photoelectric effect, by choosing materials with a small atomic number to build them.

Three general classes of experiments have been proposed for LCLS. In the first the X-ray beam is used to probe the sample without modifying it, as is done in most experiments today. In the second class, X rays are used to induce non-linear photo-processes or produce states of matter with extremely large temperature and pressure. In the third class the ultra-fast nature of the LCLS pulse is used to obtain a snapshot of the structure of the sample before the effects of radiation damage set in.

The LCLS can be used for all three types of experiments. The unfocused X-ray beam size at the exit of the undulator is about $100 \times 100 \mu\text{m}^2$, the diameter of a fine needle. At a typical experimental station located about 200 meters from the undulator exit, the size expands to only $200 \times 200 \mu\text{m}^2$. This beam can be focused by state-of-the-art techniques to about $0.1 \times 0.1 \mu\text{m}^2$ with maybe a factor of 10 loss in power. This beam size is about the size of

a virus or the smallest structures that can be lithographically fabricated today on an electronic chip. Through variable focusing the flux density of the X-ray beam can therefore be tuned by a staggering factor of about one million.

THE X-RAY PHOTON flux typically delivered to a sample in today's synchrotron radiation experiments is about 10^{12} - 10^{13} photons/sec. It typically takes one second to record a diffraction pattern of a sample. From the diffraction pattern we can then construct a real space image of the atomic positions in the sample. Because of the time duration of the measurement only the average position of the vibrating atoms can be observed. LCLS can give the same number of X rays obtained now in 1 second in an ultra-fast pulse of about 100 femtoseconds.

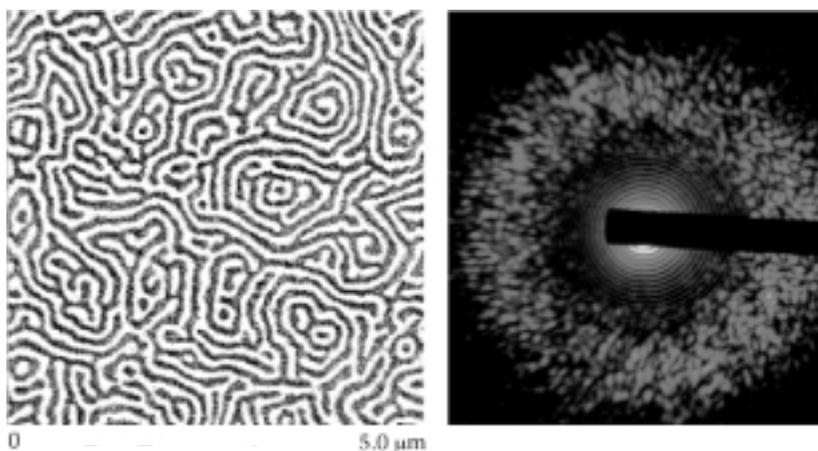
This time is hard to imagine. It is so short that sound would travel only the distance of one atom, and the fastest "thing" we know, light, would travel only about $50 \mu\text{m}$ or the width of human hair. It is also the time with which atoms, the building

blocks of materials, vibrate. Hence LCLS will allow us not only to “see” the invisible, down to the size of an atom, but also to take snapshots of the motion of atoms and of ultra-small, so called nanoscale, objects like molecules and clusters. This is illustrated in the figure, showing schematically how a small molecule separates from a bigger molecule, a chemical process called dissociation. Today we can only imagine such a movie, but with LCLS we will be able to record it and then play it back to look at the detailed process.

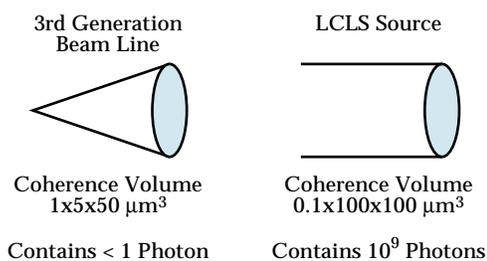
The spontaneous emission of radiation from X-ray tubes or synchrotron radiation sources is chaotic or incoherent by nature. At first sight it is surprising that it permits us to observe X-ray diffraction phenomena, originating from interference. Under certain conditions, incoherent sources can be a source

of coherent X rays. It all depends on the experimental geometry and on the dimensions of interest in the sample! If the sample is far from the source, the X-ray phase and amplitude is well defined over a certain 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, say 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 cost is a reduced intensity.

Only with the advent of high brightness third generation synchrotron sources have such experiments become practical. An example is the



Left: Magnetic worm domain pattern of CoPt alloy with perpendicular anisotropy, recorded by transmission X-ray microscopy. Right: Coherent X-ray diffraction or speckle pattern from a 5 μm diameter region of the same sample, selected by a 5 μm 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.



Comparison of the coherence properties of a state-of-the-art third generation beam line with that of an XFEL.

coherent diffraction or “speckle” pattern shown on the previous page. The pattern arises from the magnetic worm domains in a CoPt alloy, shown on the left. The speckle image on the right was recorded in transmission through a $5 \mu\text{m}$ diameter aperture, which defines the lateral X-ray coherence. The speckle pattern contains the detailed information on the true structure of the sample in the illuminated area. Any change of the magnetic structure of the sample would be reflected by an intensity change in the speckle pattern. 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 promises 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.

On a typical undulator beam line on a third generation X-ray facility, equipped with a monochromator of 0.01 percent bandpass, the X-ray beam is coherent within a micrometer size volume, as shown in the next figure. The number of photons in the coherence volume, the degeneracy parameter, scales linearly with the source brightness and with the third power of the wavelength and is on average less than one. Therefore all present day diffraction experiments are based on single photon interference effects.

In contrast, the LCLS radiation has unprecedented coherence,

about 10^9 photons in the coherence volume. The energy of coherent photons can be pooled to create multiphoton excitations and carry out nonlinear X-ray experiments. This is a largely unexplored area of science.

In its initial configuration the LCLS pulse duration is about 200 femtoseconds. Even though this is hundred times shorter than in storage rings, it can probably be further reduced to about 1/10 of this value. Several schemes to achieve ultrashort pulses have been proposed. They all use an energy-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. A promising scheme using this approach and two undulators has been studied, and could be implemented in LCLS.

THE EXPERIMENTS with LCLS and TESLA will cover a wide range of fields, from high-energy physics, to atomic physics, plasma physics, chemistry, materials science, condensed matter physics, and biology. Here we can only mention some of the present ideas and explain why they are expected to break truly new ground.

The very 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 clus-

ters, yielding information on the time scale of the damage caused by the X-ray beam, a fundamental question for other experiments.

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.

The study of molecular reactions is the very heart of chemistry. With ultrafast laser spectroscopy we can catch a glimpse of 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 is, of course, the domain of X rays, and LCLS will permit us to take diffraction snapshots as shown in the earlier figure. The importance of these experiments is underscored by the fact that many important discoveries in biology and chemistry can be traced back to the determination of a structure.

Another experiment, similar to that shown in the “speckle” figure, pushes the envelope in probing ordering phenomena in hard and soft condensed matter on the important nanometer length scale, which cannot be seen by optical photons, over a broad range of time scales. This area is not only scientifically interesting, but it constitutes

the competitive arena of future technological devices.

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.

Finally, just as the application of conventional lasers has been accompanied and driven by the R&D on lasers themselves, LCLS will also have an R&D program, exploring new ways to improve its characteristics.

THE HISTORY of and experience with three generations of synchrotron radiation sources has taught us that the above experiments are at best the tip of the iceberg of scientific opportunities. It is safe to predict that we have not yet thought of the most important experiments that eventually will be done with this new class of radiation sources—X-ray free electron lasers! Quoting Antoine de Saint-Exupery (The Wisdom of the Sands), “The most important task before us may be . . . *not to foresee it, but to enable it.*”

