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BRIGHTER AND FASTER

The promise and challenge of the x-ray free-electron laser

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Starting with the Linac Coherent Light Source in 2009, free-electron lasers are using x rays in new ways. Early results are wowing scientists the world over.

An x-ray free-electron laser pulse is so intense it can knock out a core electron from each of two neighboring atoms in a molecule and momentarily create a molecule with hollow atomic cores. (Courtesy of SLAC.)

New experimental tools and techniques open windows that allow scientists to peek into unexplored scientific realms and to test theoretical predictions. X-ray imaging is unique, both because of the penetrating power of x rays in solid matter—as Wilhelm Röntgen discovered in 1895—and because x-ray wavelengths are short enough to resolve the interatomic spacing in matter via diffraction—Max von Laue's discovery in 1912. Those properties allow scientists to push forward fundamental physical sciences and to find major applications in structural imaging, from new commercial drugs to jet turbine blades.

The early success of the Linac Coherent Light Source (LCLS) has bolstered plans for more accelerator-based x-ray free-electron lasers (XFELs) in Europe and Asia. But the new machines create a challenge: The ultrabright femtosecond pulses generated by XFELs have properties far beyond previous sources. They carry a million times more pulse energy than synchrotron x rays, are 10 000 times shorter, and have coherence that can produce focused x-ray beams with intensities up to 10^{20} W/cm^2, more than a billion times greater than any previously achieved. The XFELs demand new research methods that can take advantage of those characteristics.

Building an x-ray laser

The x-ray source of choice for the past quarter century has been the third-generation synchrotron facility. There, relativistic electrons circulate in a storage ring equipped with magnetic undulators that can produce up to a watt of synchrotron radiation. Today dozens of research synchrotrons around the world serve tens of thousands of scientists.

Synchrotrons are not lasers. Undulator-based synchrotron x rays achieve their brightness through a resonance condition in the photon emission from a relativistic electron traversing many periods of an alternating magnetic field. If the electron's field-induced wiggle motion causes it to slip behind the synchrotron emission by one x-ray wavelength on each oscillation, then radiation from different periods in the undulator will be in phase and will build up coherently. But radiation from different electrons remains incoherent.

Lasing requires that many electrons radiate in phase, and to do so, they must bunch up and wiggle in concert. In 1971 John Madey reasoned that the interaction of the wiggling electrons with the radiation they produced would have the desired effect of creating microbunches separated by the lasing wavelength. Furthermore, Madey showed that the equations governing that process were identical to the laser equations and that such a system was, in fact, a free-electron laser (FEL).[1]

Over the next 20 years several FELs were built and operated in the far-IR, near-IR, and visible wavelength ranges. They typically employed linear accelerators to drive relativistic electron beams through undulator structures collinear with resonant optical cavities to optimize the interaction between the electrons and the light. (See the article by William Colson, Erik Johnson, Michael Kelley, and Alan Schwettman, PHYSICS TODAY, January 2002, page 35.) A real limitation to extending FELs to x rays was the absence of suitable cavities and optics.

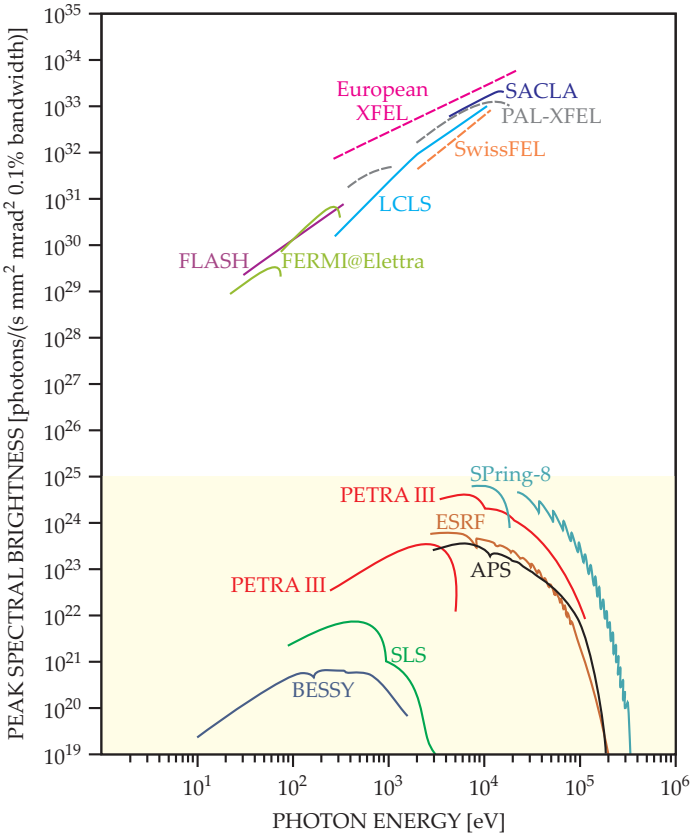

Figure 1. The peak brightnesses of select free-electron lasers (FELs) and synchrotron facilities are shown for a wide photon energy range. The FELs, all in the unshaded region, are up to 10 orders of magnitude brighter than synchrotron sources, in the shaded region. The solid lines are operating FELs and dashed lines are FELs under construction. (Adapted from ref. 16.)

The solution to the optics problem was to realize that a relativistic beam, as a gain medium, has a unique property: It moves at nearly the speed of light and therefore can remain coupled to its own radiation for long distances, even dozens of meters. Thus the spontaneous synchrotron radiation can build up enough to begin the process of microbunching. That method of lasing, called SASE for self-amplified spontaneous emission, has no wavelength limitations.

In SASE, the radiation experiences exponential growth as the electrons in the beam self-organize into microbunches. At FEL saturation, one electron can spontaneously emit about 10^3–10^4 photons. With about 10^9–10^{10} electrons in a bunch, SASE x-ray pulses can have about 10^{12}–10^{14} coherent photons. Figure 1 compares the brightness of FELs with those of some synchrotron radiation facilities.

In 1992 Claudio Pellegrini proposed a hard x-ray SASE FEL using the SLAC linac.[2] Questions regarding the practicality and feasibility of such an x-ray laser, together with the very real complication

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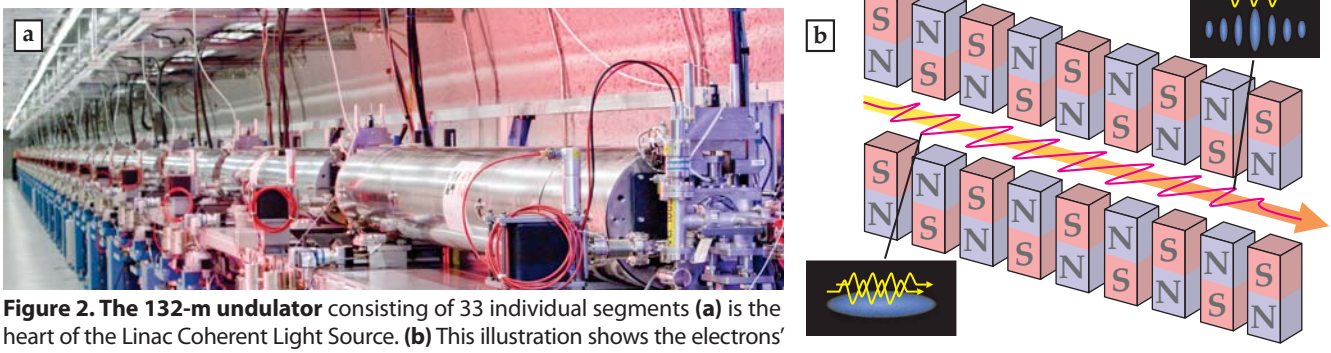

Figure 2. The 132-m undulator consisting of 33 individual segments **(a)** is the heart of the Linac Coherent Light Source. **(b)** This illustration shows the electrons' sinusoidal path (red wavy line) in the magnetic field of the undulator's array of permanent magnets with alternating north and south poles. The yellow arrow indicates the emitted photons. The x rays emitted by different electrons when they first enter the undulator are not coherent (lower left). In self-amplified spontaneous emission, the electrons interact with their own emissions and self-organize into microbunches (upper right) that wiggle synchronously to produce a coherent x-ray laser pulse. (Photo courtesy of SLAC.)

that the SLAC linac was engaged in world-leading high-energy physics at the time, delayed the project.

Meanwhile, a series of experiments at Los Alamos, Brookhaven, and Argonne National Laboratories in the US and the German Electron Synchrotron (DESY) in Europe demonstrated and refined the concept of a SASE FEL. In 2005 FLASH, then called the TESLA Test Facility, a vacuum ultraviolet FEL at DESY, became the first SASE user facility for international science.

At SLAC, the LCLS began to lase on the first attempt in April 2009 and exceeded its designed x-ray photon energy and pulse energy within days.[3] Figure 2 shows the facility's 132-m-long undulator where the lasing occurs. When the first research experiments commenced in October 2009, the laser could produce 250-fs pulses with energies of 1 mJ, or, at reduced energies, pulses shorter than 10 fs. That result was a happy surprise to scientists and even experts who only expected 250-fs pulses so early in the game. Over the ensuing six years of operation, the LCLS has improved and widened its parameter range to x-ray energies from 280 eV to more than 10 keV, pulse durations as short as 2–4 fs or as long as 500 fs, and pulse energies that can exceed 3 mJ.

In addition to the LCLS and FLASH, other FELs now operating include the vacuum ultraviolet FEL in Italy called FERMI@Elettra and a hard x-ray FEL called SACLA, part of Japan's RIKEN research institute. A second soft x-ray installation at DESY, FLASH-2, is beginning operations, and a hard and soft x-ray FEL in South Korea (PAL-XFEL) will be completed soon. Other projects under construction include the large European XFEL project at DESY and the SwissFEL at the Paul Scherrer Institute in Switzerland.

A hallmark of FEL research is the presence of many special operating modes. In normal SASE mode, the laser pulse builds up from noise, so it consists of randomly spaced spikes of 1- to 5-fs duration within an energy envelope of about 15 eV. In short-pulse mode, reduced electron-bunch charge creates x-ray pulses with only one or two of those noise spikes.

Scientists can also request two x-ray pulses a few femtoseconds or picoseconds apart, with the same or different wavelengths. There are several ways to produce such a mode, depending on what is most desired—fixed time separation, fixed wavelength separation, tunable separation, or a combination. SASE operation can also be replaced with so-called self-seeding, in which a tunable element, typically a crystal or grating monochromator, is inserted halfway through the undulators to filter a specific wavelength for further amplification; that method provides higher stability and reduces the laser bandwidth.

Science at XFELs

With SASE FELs, researchers can explore x ray–matter interactions in new regimes of time and intensity that synchrotron x rays cannot reach. Synchrotrons were originally developed by the physics community in the 1970s and 1980s, and they quickly became hugely important tools for molecular biology as well as for physics. The same trend may be repeating for XFELs.

This article emphasizes physics research applications, and thus only briefly mentions some of the most exciting new research methods that are aimed at biology, such as serial femtosecond crystallography. (See PHYSICS TODAY, April 2011, page 13.) However, readers are encouraged to refer to some of the recent publications in that area.[4]

Fundamental scientific questions at SASE FELs often involve time and motion. How do atoms move when chemical bonds break? What are the pathways of light-induced atomic motion or radiation damage? Such motion is at the foundation of all chemical reactions. Atomic nuclei in molecules move angstroms in femtoseconds, while valence electrons can move across an interatomic bond in only hundreds of attoseconds. More tightly bound inner-valence and core electrons move even faster.

Laboratory probes based on ultrafast lasers and high harmonics can often match those time scales, particularly for optical wavelengths longer than 20 nm. But XFELs have photon energies sufficient to access core and inner-shell electrons, and their femtosecond pulse durations and high intensities make them a powerful time-resolved probe of molecular structural dynamics. Moreover, x-ray absorption methods can target specific atoms within molecules and thus provide atomic-scale spatial resolution.

Optical absorption or scattering experiments designed to induce femtosecond processes require in-

tense fields for the simple reason that sufficient light must be compressed into an ultrashort pulse. The requirement is exacerbated in the x-ray regime because scattering is far weaker. For example, a typical atomic ionization cross section at 1-keV incident photon energy is 1 Mb, which implies that a 100-fs FEL pulse needs an intensity of 10^{15} W/cm^2 to ionize an atom.

That intensity corresponds to a field amplitude well above the strong-field ionization threshold at visible or IR wavelengths. Not only do XFELs attain such intensity, they reach it with ease. A 1-mJ pulse from the LCLS only needs to be focused to 30 μm to do so. In fact, pulses can be focused to less than 1 μm via grazing-incidence reflections from pairs of bent silicon (Kirkpatrick–Baez) mirrors; so the intensity can be 1000 times greater. Such high intensity provides the opportunity to study strong-field and nonlinear x-ray physics.

Hollow atoms and exploding molecules

Nonlinear x-ray absorption in atoms, molecules, and clusters under intense, short x-ray exposure creates exotic states of matter under extreme conditions. The first experiments performed at the LCLS were attempts to observe strong-field photoionization in light atoms and molecules.[5–7]

Researchers used electron detectors to measure the angular distribution of electrons emitted by inner-shell photoionization and Auger electrons. In the Auger process, an inner-shell vacancy is filled by an electron from a higher energy level. The energy released in that decay is absorbed by another electron, which is then emitted. In addition, ion detectors measured the charge states and kinetic energies of the ions produced by photoabsorption.

Those experiments showed that a focused x-ray laser beam can completely strip all 10 electrons from a neon atom,[5] and that the process is entirely sequential: Each photon is absorbed by preferentially ionizing an inner-shell electron, and the inner vacancy is subsequently filled by an Auger process that emits the next electron. Then the "PA" pair of photoionization (P) and Auger relaxation (A) repeats until fewer than three electrons are left, at which point A is impossible and pure P takes over. In Ne, the most likely sequence to remove all 10 electrons is PAPAPAPAPP for photon energies greater than 1360 eV, the ionization potential of Ne^{9+}.

If the photon energy is less than the ionization potential for that charge state, then the ion is stable against further stripping. Remarkably, even at such high intensities, researchers found no evidence that still higher charge states could be produced by sequential photoexcitation or coherent multiphoton absorption.

In contrast, experiments performed on nitrogen molecules showed the opposite effect—decreased absorption, or x-ray transparency—if the ionization rate greatly exceeds the Auger relaxation rate so that the atom loses both of its core (1*s*) electrons before Auger redistribution can refill them.[6,7] The double ionization, also called double core-hole formation, slows the overall rate for ionization, and for short pulses it suppresses complete stripping of the atoms.

To make such hollow atoms and the accompanying x-ray transparency, the experiments varied the duration of the x-ray pulses without changing the pulse energy or the photon energy. That way the ionization rate changed but the total fluence stayed constant. Pulse-length control, a common feature of ultrafast laser experiments but not standard at synchrotrons, was the first example of a special operating mode for the LCLS. It was accomplished by decreasing the charge of the electron pulse, compressing it, and using gas attenuators to keep the pulse energy constant.

Double core-hole formation in N_2 and the accompanying intensity-induced x-ray transparency were among the first nonlinear effects observed at LCLS. The fundamental atomic and molecular finding is important because it demonstrates that radiation damage, which deteriorates the scattering images of macromolecules such as viruses, can be reduced with the use of ultrashort x-ray pulses even if they are ultraintense—and x rays from the LCLS are both.

The opportunity to study double core vacancies led to Auger spectroscopy and other experiments on the exotic, transient states.[7] In molecules, rapid ionization also leads to so-called two-site double core-hole states in which core vacancies exist simultaneously in neighboring atoms within a molecule. In that case Auger spectroscopy is a powerful tool in chemical analysis because it can differentiate similar molecular environments—for example, carbon monoxide versus carbon dioxide or N_2 versus nitrous oxide.[8]

Such measurements require an electron spectrometer with high resolution, high throughput, and low background. Magnetic bottle time-of-flight electron spectrometers, which determine the energy of ionized electrons by measuring their arrival times at detectors, were introduced in the 1980s to provide good resolution, large solid-angle coverage, and high throughput for low kinetic energy photoemission. They have been reintroduced for FEL-based Auger spectroscopy, for which the electron energies are much higher.

More complex molecules such as fullerenes—60 chemically bonded carbon atoms—provide the opportunity to create and study rapidly evolving nanoscale plasmas during intense x-ray irradiation. They also help benchmark the physical damage that occurs when focused FEL x rays are used in biomolecular structure studies.

During the past decade, researchers have developed several models to study the atomic-scale time evolution of larger molecules irradiated by intense XFEL pulses. X-ray scattering measurements do not give detailed dynamical information for the individual particles. Thus comparison between experiments and models are necessary to validate quantitative predictions.

In the FEL's focus, C_{60} molecules suffer a violent fate: They explode, sometimes into single C atoms, when absorption of 485-eV photons induces ion fragmentation.[9] Figure 3a is an artist's conception of C_{60} exploding in the intense x-ray laser, and figure 3b shows the ionized C yield for all measured ion charge states. Models show that the yield distribution is strongly influenced by secondary ionization processes from the freed electrons, which form a nanoplasma with the highly charged C ions.

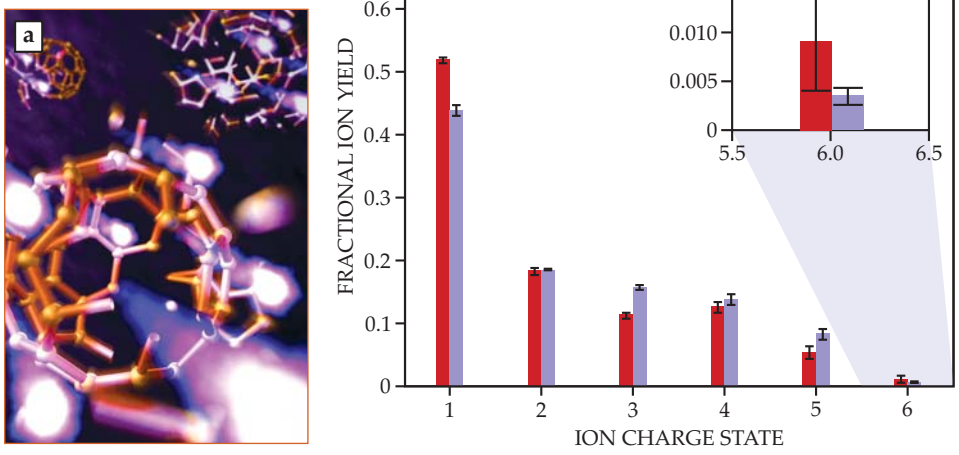

Figure 3. Ion fragmentation of fullerene molecules. **(a)** This illustration helps us visualize the fullerene molecules' violent fate when they encounter the intense x-ray pulse of the Linac Coherent Light Source. Ion fragmentation can lead in some cases to carbon ions stripped of all their electrons. (Courtesy of SLAC.) The individual C-ion yield distribution **(b)** is shown for all charge states subsequent to C_{60} fragmentation. Initial theoretical models had predicted higher yields of C ions, but comparison with the experimental data revealed that a nanoplasma of trapped low-energy electrons recombined with the high-charge C states to suppress the ion yield. A refined model, shown in purple, containing that secondary effect is in excellent agreement with the measured data, shown in red. (Adapted from ref. 9.)

More recent nanoplasma models show that if the target is loaded with hydrogen—for example, a methane cluster—then high charge states in C are suppressed by massive charge transfer from the protons. In biological molecules, the abundance of protons may therefore slow Coulomb explosions. If validated by experiments, that idea may be an important strategy for single-shot x-ray scattering experiments for structure determination.

Pump first, probe later

The motions and locations of nuclei and electrons determine energy flow and charge transfer in molecules. Pump–probe spectroscopy is a stroboscopic method to record those motions. A pump pulse initiates the motions and a probe detects them. The collection of probe signals taken at multiple time delays between the two pulses captures the dynamics.

The x-ray laser can serve as either the pump or probe when it is paired with a short pulse laser of the appropriate wavelength. Pump–probe methods were developed for conventional lasers that produce the pump and probe with a variable optical path difference to set the delay, which can be adjusted easily by small fractions of the pulse duration. The delay is much more challenging when one of the pulses is a local laser and the other is an x-ray laser produced by transporting relativistic electrons more than a kilometer. The delay can drift by picoseconds if the Sun goes behind a cloud and the temperature in the accelerator building changes.

Feedback methods bring the pump and probe within less than a picosecond separation, which is much less than a millionth of the light travel time through the FEL. To improve on that already impressive figure, experimenters have adopted several methods to separately measure the relative arrival of the pump and probe on every laser pulse to be sorted through later. Such sampling methods permit precision approaching 10 fs.

A particularly striking example of a pump–probe method that images how nuclear motion is coupled to charge transfer is a recent experiment[10] that captured x-ray-initiated charge rearrangement during femtosecond laser-induced dissociation of iodomethane (CH_3I). A photon from a short-pulse near-IR laser breaks the I–CH_3 bond. Then while dissociation is under way, a precisely delayed strong x-ray pulse ejects several electrons from the iodine via inner-shell ionization and Auger electron cascade, which suddenly create a strong positive charge that can attract electrons from the receding CH_3. Measurements of the charge and the kinetic energy of the departing fragments have produced an especially rich time- and bond-length-resolved view of the electron transfer process.

In more complex molecules, the charge transfer dynamics induced by laser excitation may involve several atoms and many bonds, so that the final-state ion-fragment charges and energies are not simply related to the initial charge transfer. However, the spectra of electrons from x-ray absorption and prompt Auger relaxation are snapshots of the molecule at the time of x-ray absorption. Time-resolved transient Auger spectroscopy measurements enable such snapshots.

Auger electron emission is a nonradiative electron-cooling mechanism to remove the energy deposited in an x-ray-induced core vacancy—a valence electron fills the core hole and a second valence electron carries away the excess energy. Auger emission bands identify the atomic species. For example, in oxygen-containing molecules, the O *KLL* Auger emission process is the decay of an electron from the n = 2 energy level to the n = 1 level, together with the emission of another n = 2 electron at about 500-eV kinetic energy.

The fine structure within the emission band reveals the density and energy of valence electrons in the vicinity of the O atom. Transient shifts in the Auger spectrum as a function of the x-ray probe's delay following photoabsorption can therefore provide a clear signature of the redistribution of energy in a molecule.

Transient Auger probe methods were recently employed to study the time it takes for thymine ($C_5H_6N_2O_2$), one of the four nucleobases of DNA, to rid itself of the potentially damaging effects of UV light absorbed from the Sun.[11] Researchers tuned 50-fs LCLS laser pulses above the O 1*s* absorption edge, and observed changes in the oxygen Auger spectrum with 75-fs time resolution. The results revealed that the thymine molecule internally and

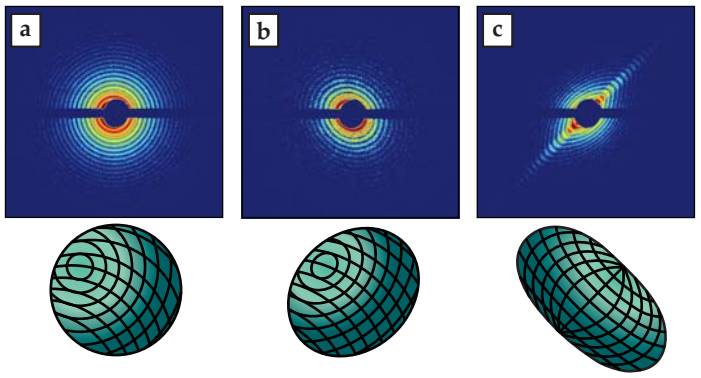

Figure 4. Diffraction images of pure helium droplets made with femtosecond x-ray pulses. **(a–c)** Images represent circular, elliptical, and streaked patterns, respectively, with corresponding three-dimensional representations of the droplet shapes placed below each diffraction pattern. Diffracted x-ray intensities are displayed in a logarithmic color scale. **(d)** A droplet outline is reconstructed from the diffraction pattern in panel c using an inverse Fourier transform. (Adapted from ref. 14.)

harmlessly redistributes substantial excitation energy within 200 fs of absorbing UV light. The experiment helped to answer an important question about photoprotection, and it demonstrated that transient Auger spectroscopy can map changes in a molecule.

Femtosecond x-ray pulses can act as both pump and probe. Several methods for splitting the x-ray beams have been devised. The simplest is wavefront division, in which a split mirror intercepts half of the x-ray beam on each half of the mirror, and then the two mirror halves are adjusted to produce a delay. A significant advantage of the method is that the geometry of the setup precisely determines the delay, so no synchronization of separate sources is required.

The high temporal resolving power of split-and-delay methods was demonstrated at FLASH, where researchers used a split mirror combined with imaging spectrometers to measure interatomic coulombic decay in Ne_2. The process is similar to Auger relaxation except that the two electrons that participate in the relaxation come from different atoms. In the FLASH experiment,[12] the neon dimer's 150-fs interatomic coulombic decay lifetime was measured with the remarkable precision of ±5 fs.

Techniques that use x rays as both pump and probe can also map out more complex processes following x-ray absorption. A recent example from the LCLS is the measurement of ultrafast isomerization initiated by x-ray core ionization in deuterated acetylene (DCCD). The isomerization consisted of a rapid migration of a deuteron from one C site to the other to yield vinylidene (CCDD). The experiment used deuterium rather than hydrogen to reduce background and because the heavier deuteron moves more slowly.

Absorption of the first x ray is followed by rapid Auger decay to initiate isomerization from the acetylene dication $DCCD^{2+}$ to the vinylidene dication $CCDD^{2+}$. The motion can be captured by absorption of a second ultrafast x-ray pulse, which leads to a Coulomb explosion into four ions C^+, C^+, D^+, and D^+. The methodology produces a molecular movie that shows the evolution in time of the ion momenta revealing that nonadiabatic interactions can couple nuclear motion to the electronic relaxation in the first 12 fs following core ionization.[13]

Dynamics of correlated systems

Superconductivity and superfluidity have been studied using most techniques available to condensed-matter physicists. Now XFELs can view the superconducting or superfluid transition with femtosecond resolution and thereby capture dynamics.

Micron-scale individual droplets of liquid helium-4, injected into a vacuum, quickly cool by evaporation through the superfluid transition temperature, so the droplets are ideal laboratories for studying the onset of superfluidity. (See the article by Peter Toennies, Andrej Vilesov, and Birgitta Whaley, PHYSICS TODAY, February 2001, page 31.) Pulsed femtosecond XFELs can image those droplets one at a time by coherent x-ray diffraction. A recent experiment at the LCLS employed single-shot femtosecond x-ray coherent diffractive imaging, illustrated in figure 4, in which a micron-focused x-ray beam scatters from a micron-scale individual xenon-doped helium droplet, and the scattered x rays are collected on a high sensitivity CCD detector.[14] (See PHYSICS TODAY, November 2014, page 16.) Droplets could be detected with as few as 10^8 atoms—less than 100 nm.

As a droplet cools below the superfluid transition temperature, its angular momentum becomes compacted into quantized vortices, which attract the Xe atoms. Coherent x-ray scattering then reveals both the shape of the overall droplet and the arrangement of vortices into a regular lattice.

Two unanticipated findings emerged from the images. First, the shapes of the droplets indicate that their rotation can exceed the classical stability limit for rotating liquid He. Second, the density of vortices is about five orders of magnitude higher than ever measured in larger superfluid systems. Both results point to very high values of angular momentum for the quantum system. They show that studies of superfluid He droplets can explore new excitation regimes in quantum fluids, and lead to a deeper understanding of quantum collective phenomena.

Superconductivity also has transient features on nanometer scales, such as the recent discovery that IR-frequency pulses can drive resonant structural deformations in high-temperature superconducting yttrium barium copper oxide crystals. Those deformations lead to coherent interlayer transport, a signature of superconductivity. The effect has been dubbed transient room-temperature superconductivity, but its true nature is still controversial.

A collaboration used 6.7-keV x rays from the LCLS to measure the changes in YBCO following excitation with 300-fs mid-IR pulses—a challenging wavelength for ultrafast pulses. The atomic displacements last for a few picoseconds, which is long enough to clearly resolve both the structural change and the relaxation back to the normal state.[15]

The experiment required high statistical precision in diffraction intensity, a high degree of synchronization between the pump laser and the x-ray probe, and a state-of-the-art IR source. Transient x-ray diffraction produced the kind of detailed information necessary to test models of structural properties that help drive superconductivity. The work also introduced the use of photonic driving to control—either to enhance or to reduce—the superconducting coupling in those materials.

A bright future

The launch of operations at the LCLS, FLASH, SACLA, and FERMI@Elettra has been a resounding success. Research applications will continue to expand as laser improvements lead to new capabilities such as intense attosecond x rays. Other future developments, either under discussion or in planning, include x-ray laser beams at photon energies above 50 keV and repetition rates as high as 1 MHz. A second laser at SLAC, LCLS II, is expected to provide 100-kHz x-ray beams within the next few years.

Still more advanced designs based on plasma wake-field or dielectric laser acceleration might also eventually shrink the size and cost of XFELs to make them available for hospitals, university research labs, or even commercial applications. Such developments would parallel the history of improvements and new applications in conventional lasers. Research work in that direction has already begun.

In the meantime, scientific research with XFELs will add to our understanding of the interaction of matter with photons and reveal the intricacies of phenomena still not understood in physics, chemistry, and biology. The combination of short wavelength and femtosecond time structure provides a unique tool for investigating nanoscale structure and dynamics.

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