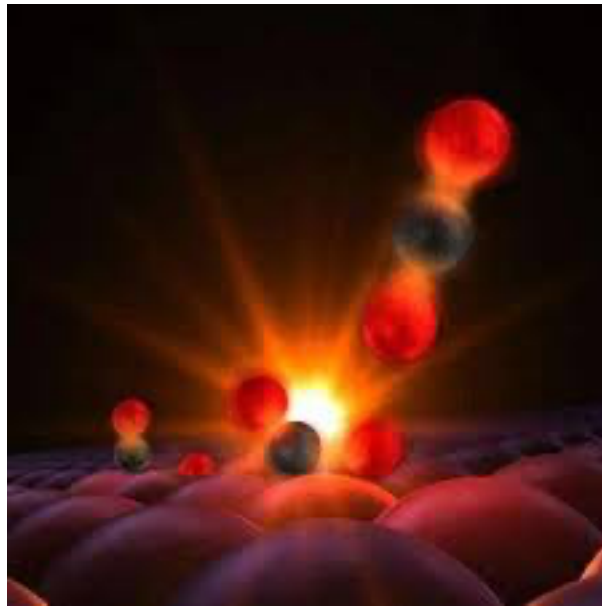


LCLS Science Impact Assessment

February 2023

Submitted to Provost Persis Drell, Interim SLAC Director Stephen Streiffer, and the SLAC Scientific Programs Committee



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Executive Summary

This assessment addresses the scientific impact of the Linac Coherent Light Source (LCLS) from inception in 2009 through 2022, based exclusively on published results, emphasizing the most significant achievements recognizable and appreciated by the science community. We highlight and evaluate the highest impact results from LCLS around specific scientific themes appropriate to the scientific mission of LCLS, SLAC, and the DOE Office of Science.

Given the transformative nature of LCLS, scientific results from LCLS have led to qualitative advances over a broad cross section of scientific fields, ranging from atomic and molecular science, ultrafast chemistry and catalysis, advanced materials, structural biology, high-energy density science, to photon science, as enumerated in this report. While the LCLS is to be credited with many significant scientific discoveries that have natural ties to very specific, applied problems in energy, biology, materials, and chemical sciences, the importance of the fundamental new knowledge that the work at LCLS has created – knowledge that will provide the foundation for transformational advances both seen and unseen at the present time – should not be underestimated.

LCLS has created new areas of science through the development of entirely new experimental methods inspired by its unique capabilities, including atomic scale spatial and temporal resolution, X-ray optical coherence, brilliance, and chemical selectivity. These new methods pioneered by LCLS have been adopted by emerging X-ray free-electron lasers (FELs) worldwide. In addition, well-established X-ray scattering and spectroscopy methods that have been adapted for the first time at LCLS have opened transformative new classes of time-resolved studies possible only with LCLS (or similar X-ray FEL) capabilities. Key experimental methods adapted to LCLS from X-ray science include time-resolved X-ray scattering methods (for example, serial femtosecond crystallography, time-resolved Bragg diffraction, Fourier-transform inelastic X-ray scattering, and time-resolved gas phase scattering), coherent X-ray scattering methods (time-resolved Bragg coherent diffractive imaging, X-ray photon correlation spectroscopy, single particle imaging, and in-flight holographic imaging), and advanced spectroscopy methods (time-resolved multi-modal X-ray scattering and X-ray spectroscopy, attosecond science and methods, and spectral domain correlation spectroscopy). Additionally, LCLS has pioneered advanced methods for investigating matter in extreme conditions, advanced sample delivery methods, nonlinear X-ray science, and advanced operating modes and diagnostics for LCLS.

LCLS has taken deliberate steps to enhance its scientific impact:

- Sustained investment in research and development (R&D) at LCLS, particularly novel source development
- Activities such as multiplexing, protein crystal screening, and standardization of configurations (where possible) that expand access and increase capacity
- Introduction of LCLS Scientific Campaigns, enabling strategic, transformative science with a long-term horizon that complements the investigator-driven proposal process
- Nurturing and sustainment of expert in-house teams for all experiments
- Cultivation of a close and synergistic partnership between LCLS and Stanford University.

These actions are discussed in more detail in the overview section.

Atomic, molecular, and optical science: By inducing energy modulations in the LCLS electron

beam, high-current spikes could be produced in a magnetic chicane, which upon injection into an undulator led to the generation of widely tunable attosecond pulses with an intensity sufficient to drive nonlinear processes. The attosecond pulses generated at LCLS enable attosecond pump-attosecond probe experiments, as well as experiments where core-shell electrons are used to observe electron dynamics from the viewpoint of a particular atom. These attosecond capabilities recently led to a first major scientific result, namely the observation of coherent electron motion during Auger-Meitner decay and should lead to a deeper understanding of the role of electron correlation in quantum materials.

Biological science: LCLS experiments have revealed exquisite detail of ligand binding to several challenging membrane proteins by the use of the serial femtosecond crystallography method, which was developed to capture diffraction of protein crystals from single sample-destroying LCLS pulses. The LCLS has engendered a major advance in the understanding of important enzyme-catalyzed reactions. Using LCLS, short-lived intermediates and transient states of enzyme reactions are simultaneously characterized spectroscopically and “seen” in crystal structures. Further, the femtosecond time scale of LCLS pulses is highly relevant to chemical reactions. Data on the spectroscopic and structural properties of intermediates substantially advance reaction theory and our ability to simulate reaction trajectories in other enzyme systems.

Condensed-phase chemistry and catalysis: Experiments at LCLS have changed the way the scientific community thinks about molecules. Seminal and paradigm-shifting discoveries were enabled by the unique capabilities to probe the electronic and geometric properties of molecules and chemical systems at a more fundamental level than was ever possible before. Completely new observables were revealed, and the field was pushed beyond the boundaries of what is currently known and/or presumed to be true. This created new paradigms for chemical transformations. The fundamental new knowledge and the new approaches provide templates for probing a broad array of reactions spanning chemical, biological, and materials chemistries for new ways of translating the fundamental understanding of chemical systems to solutions of key challenges for a sustainable future.

Gas-phase chemistry: LCLS experiments provide the long sought-after separation of structural and electronic effects, allowing the extraction of molecular dynamics directly from the experimental results. The pioneering experimental capabilities at LCLS include: 1) the ability to record time-resolved X-ray diffraction using the free-electron laser and time-resolved electron diffraction through the Ultrafast Electron Diffraction (UED) instrument; 2) the ability to implement spectroscopic techniques based on site-specific core-level photoabsorption. The UED instrument enabled the study of ring opening of cyclohexadiene with unprecedented sub-ångstrom spatial and femtosecond temporal resolution. The X-ray wavelength of the LCLS allows the development of measurement schemes that are based on diffraction, which are ideally suited for measuring time-dependent structural changes, as well as the development of novel techniques that are uniquely sensitive to time-dependent electronic changes in a molecule, and moreover, from the viewpoint of a particular atom within the molecule.

Materials science and condensed matter physics: LCLS, with complementary advances in theory, simulation, and materials synthesis, has vastly expanded the range of materials phenomena that the materials community can study and ultimately control. Discoveries at LCLS have initiated the coherent control of excitations in a previously inaccessible regime in which it is possible to use light and other experimental tools to interact directly with the relevant physical phenomena. Atomic-scale information derived from X-ray scattering studies at LCLS has

provided an understanding of the transformations of materials between phases, specifically in highly disordered systems for which the dynamics at short time scales have been previously unknown. LCLS has also provided new insights into energetically fragile many-electron dynamics of quantum materials via the separation of coupled dynamics in the time domain, based on its unique combination of selective near-equilibrium perturbation of coupled modes with precision snapshots of the electronic and atomic structure at fundamental length and time scales. Novel techniques and methods pioneered at LCLS will continue to offer new insight into the structure and excited states of solids and nanoscale materials.

Materials in extreme conditions: Experiments at LCLS have provided unique insight into strength and plasticity at the lattice level, enabling links to macroscopic models that bridge micro- to macro-length scales in an area of importance to national security. The Matter in Extreme Conditions (MEC) platform at LCLS has provided completely new insights into pressure induced phase transitions at ultrahigh rates, demonstrating, for example, that the most complex phase a single element can form (an incommensurate host-guest structure) develops in less than a nanosecond. Complex physics that may take place in planetary physics – for example the properties of hydrocarbons in large planets – is being explored with LCLS. Finally, the ability of LCLS to create uniform hot (multi-million Kelvin) plasmas at exactly solid density is providing completely a new understanding of dense quantum plasmas.

Cover: This illustration shows atoms forming a tentative bond, a moment captured for the first time in experiments with LCLS. The reactants are a carbon monoxide molecule, left, made of a carbon atom (black) and an oxygen atom (red), and a single atom of oxygen, just to the right of it. They are attached to the surface of a ruthenium catalyst, which holds them close to each other so they can react more easily. When hit with an optical laser pulse, the reactants vibrate and bump into each other, and the carbon atom forms a transitional bond with the lone oxygen, center. The resulting carbon dioxide molecule detaches and floats away, upper right. See pages 14-15 of this report for more detailed description of this process. (SLAC National Accelerator Laboratory)

Overview

A new scientific frontier opened in 2009 when the world's first X-ray free-electron laser (XFEL), the Linac Coherent Light Source (LCLS) facility, began operations at SLAC National Accelerator Laboratory. XFELs represent a revolution in X-ray science that has the potential to transform the field for the 21st century – providing X-ray beams with a peak brightness that is 9 to 10 orders of magnitude higher, and average brightness 3 orders of magnitude higher, than a synchrotron source. As the world's first XFEL, much of the early research at LCLS has focused on: 1) discovery science, opening and exploring new scientific frontiers; 2) advanced X-ray method development to fully exploit the transformative capabilities of XFELs; 3) pushing the frontier in X-ray capabilities, e.g., from ~100 femtosecond (fs) to ~0.2 fs pulse durations. LCLS is an excellent example of how technological progress creates new methodologies for attacking open scientific problems. From its inception, the strength of LCLS has been the ability to continually improve the performance of the accelerator and the experimental end stations. Significant accelerator achievements include the implementation of self-seeding in both the hard X-ray and soft X-ray ranges, the ability to produce multiple XFEL pulses with independent tunability, and, most recently, the generation of isolated attosecond pulses, as well as pulse trains, tunable across the soft X-ray range. LCLS developed several innovative experimental methods by adapting well-established X-ray scattering and spectroscopy approaches, typically employed at synchrotrons or table-top optical laser laboratories, as well as by creating entirely new experimental methods inspired by the unique capabilities of LCLS.

LCLS is a user facility and thus directly responsive to the needs of the science community. A strong and proactive interaction between the scientific community and the LCLS machine and experimental groups drives pioneering technical developments at LCLS. One notable example is the new capability to generate and manipulate attosecond pulses, unlocking important experimental capabilities such as 4-wave mixing and multi-dimensional spectroscopy. These methods enable the study of critical phenomena ranging from the energy and charge transfer in light harvesting materials to conical intersections in photochemistry.

The substantial scientific accomplishments of LCLS are reflected in both the number of publications (over 1,600 to date) and the number of users (over 3,000 unique users and 13,000 user visits, despite the limitations of only a single beamline). There have been 677 full-scale user experiments and 285 in-house experiments in the 2009-2020 period, reflecting the incredibly high scientific productivity of the facility. Additional indicators of the science impact of LCLS include the steady increase in the annual number of unique users (>1,000 per year, just prior to the COVID pandemic) and the scientific demand as reflected in the average proposal success rate of <25% over the past decade (down to 18% for the latest proposal round).

The early success of LCLS triggered intense development of similar XFEL facilities around the world. Today, there are 5 hard X-ray FELs in operation (LCLS, SACLA in Harima Science Garden City, Japan, PAL-XFEL in Pohang, South Korea, SwissFEL at the Paul Scherrer Institute in Switzerland, and the European XFEL in Hamburg, Germany) and others under construction (LCLS upgrade/LCLS-II High Energy, LCLS-II/HE, at SLAC, and SHINE in China), in addition to 3 extreme ultraviolet (XUV)/soft X-ray facilities (FLASH5 in Hamburg, Germany, FERMI6 in Trieste, Italy, and SXFEL in Shanghai, China). The first generation of XFEL facilities have been based largely on gigaelectronvolt (GeV)-scale electron beams generated by conventional pulsed-RF (radio frequency) accelerator technology, operating at repetition rates in the 10-120 Hertz (Hz) range. It is notable that the LCLS facility inspired not only the design of many similar

copper (Cu)-linac based XFELs, but also the suite of beamlines and experimental instruments at these facilities. Moreover, pioneering experimental techniques developed at LCLS are being used in these newer XFEL facilities, often by scientists who learned the techniques at LCLS. Further, researchers at LCLS have cultivated deep collaborations with XFEL teams and facilities across the world, enhancing growth of the community and capabilities via mutually beneficial sharing of ideas and adopting techniques at LCLS that have been developed elsewhere.

The sustained investment in R&D at LCLS, particularly novel source development, has led to major advances that have driven the science, including spectral seeding, polarization control, progression from 100 fs to 10 fs to few-fs to attosecond-regime pulses, gigahertz (GHz) pulse trains, TW-scale peak powers, and exquisite control of dual pulses including two-color generation. This is further enhanced by LCLS policy to allocate 25% of beam time to in-house development of new techniques and science concepts, aimed at enhancing the community's capabilities.

To increase the scientific impact of LCLS, given the extremely high user demand, LCLS leadership has implemented several activities to expand access and increase capacity. Introduction of "standard configuration" access to instruments has enabled non-expert users to field experiments on a stable platform, thus minimizing the barrier to entry and maximizing productivity. The creation of "protein crystal screening" and other direct-access modes has allowed rapid evaluation of the feasibility of XFEL measurements for a sample or system. Sustained development of real-time, fast feedback data analysis techniques permits a more informed direction of experiments and rapid focus on the time/configuration that is generating interesting and surprising data. The implementation of multiplexing on appropriate LCLS beamlines has been an important avenue for growing capacity; however, it is important to ensure this increased experimental capability is adequately staffed. To address potential staffing issues, LCLS leadership has focused on growing and sustaining expert in-house teams for all experiments, enabling staff to support both experienced and novice users.

The science addressed by LCLS needs strategic direction from LCLS management to complement the single- and few-investigator-driven proposal process. To address this need, LCLS Scientific Campaigns were introduced in August 2020 to provide access for community-wide teams to tackle grand challenge problems that require a series of LCLS experiments over multiple years. The Scientific Campaigns target specific areas where there is great potential for major scientific impact from the unique capabilities of LCLS. The scientific scope, level of ambition, and impact of the proposed research must be well above that of a regular proposal for beam time. Currently, LCLS supports 8 science campaigns, covering atomic, molecular, and optical science, biochemistry, quantum materials, catalysis, and chemical processes and reactivity. Overall, the initial progress from these campaigns has been impressive, enabling transformative science with a long-term horizon.

The close and synergistic partnership between LCLS and Stanford University has been critically important to enhancing the scientific impact of LCLS. Seed funding of new local university-based teams has enabled LCLS to kickstart new directions and build links to a broader community with a particular focus on ultrafast lasers and Earthshot-relevant areas. Stanford's investments in the Arrillaga Science Center, the SLAC/Stanford institutes, and its photon science faculty are all designed to enable cross-fertilization of ideas and to broaden the community of users who can take advantage of XFEL sources. Collaboration between LCLS and Stanford is an area where progress is on a positive trajectory.

In the following sections this assessment addresses the scientific impact of LCLS from inception in 2009 through 2022, based exclusively on published results, emphasizing the most significant achievements recognizable and appreciated by a broader science community. We have deliberately avoided trying to assess science impact based simply on publication citations or similar metrics as these tend to be lagging indicators. Rather, we highlight and evaluate the highest impact results from LCLS around specific scientific themes appropriate to the scientific mission of LCLS, SLAC, and the DOE Office of Science:

- Atomic, molecular, and optical science
- Biological science
- Condensed-phase chemistry and catalysis
- Gas-phase chemistry
- Materials science and condensed matter physics
- Materials in extreme conditions

Atomic, Molecular, and Optical Science

Overall Assessment: Arguably, one of the most exciting developments in ultrafast laser science in the last few years has been the emergence of the novel field of attosecond science. Whereas femtosecond laser technology allows the development of experiments that measure atomic motion, the development of attosecond laser technology permits experiments that elucidate electronic motion in real time. Attosecond science initially developed as a lab-scale experimental science based on the use of moderately intense femtosecond laser systems and the technique of high-harmonic generation (HHG)¹. This approach continues to be quite successful but has at least 2 major limitations. Firstly, it has proven to be extremely difficult to generate attosecond pulses that are intense enough so that they can be used in attosecond pump-attosecond probe experiments. Therefore, the vast majority of attosecond pump-probe experiments are nowadays configured as two-color experiments where attosecond pulses or trains of attosecond pulses are used in combination with a moderately intense replica of the driver laser used in the HHG process, whose optical cycle is employed as a clock with attosecond time resolution. This has permitted certain classes of attosecond experiments, but with major limitations compared to the attosecond pump-attosecond probe experiments that scientists ideally would like to perform. Secondly, the attosecond pulses generated via HHG typically have photon energies below 100 eV, which means that these attosecond pulses predominantly interact with (inner) valence, rather than core-shell, electrons. As such, experiments where core-shell electrons are used to observe electron dynamics from the viewpoint of a particular atom are so far scarce in attosecond science.

In 2020 LCLS scientists achieved a major breakthrough when they reported the generation of attosecond pulses with a pulse energy that is approximately a million times higher than what is typically achieved with a lab-scale HHG source, and moreover, at 10-fold higher photon energies than common lab-scale attosecond light sources². The result was achieved by modifying the properties of the electron beam that underlies the operation of the LCLS free-electron laser (Fig. 1). Coherent infrared radiation emitted in a wiggler by the tail of the electron pulse was used to induce energy modulations in the electron beam, which were subsequently converted into ultrashort high-current spikes in a magnetic chicane. Upon injection into an undulator, widely tunable attosecond pulses were generated with an intensity sufficient to drive nonlinear processes. For example, the first demonstration of these attosecond pulses was soon followed by

an experiment showing that the pulse intensity was high enough to drive a Raman process³.

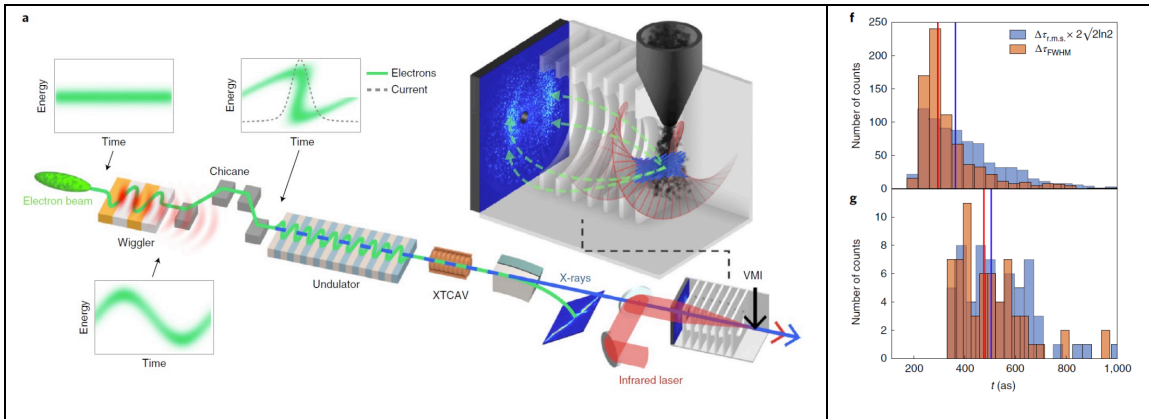


Figure 1. Schematic of the generation of widely-tunable and high-intensity attosecond pulses at LCLS. (left) Isolated attosecond X-ray pulses at the LCLS generated by manipulating the electron bunch prior to injection into an undulator where the lasing occurs. (right) Histograms show the results of single shot attosecond pulse duration measurements for operation of the LCLS at photon energies of (top) 905 eV and (bottom) 570 eV. (Adapted from Reference 2)

Described below is an example of world-leading LCLS impact in the field of attosecond science.

Coherent Electron Motion in Auger-Meitner Decay: Recently, a large international collaboration organized around the theme of attosecond science reported a first major scientific result using the attosecond capabilities of LCLS: experimental results demonstrated the role of coherent electron motion in Auger-Meitner decay⁴. This decay is one of the simplest examples in nature of the effect of electron correlation. It describes a process where an atom or molecule relaxes by means of a mechanism where 2 electrons exchange energy, with the electron that loses energy relaxing into an available lower energy state and the electron that gains energy ionizing and leaving the system. Electron correlation is at the heart of the behaviour of quantum materials that are nowadays intensively investigated due to their highly unusual properties and tremendous application potential.

In the experiment, several core-excited states were simultaneously populated by exciting nitric oxide (NO) molecules with a broadband attosecond X-ray pulse from the LCLS⁴. The dominant relaxation pathway of these core-excited molecules was Auger-Meitner decay, i.e., a refilling of the vacancy in the oxygen core orbital from which the excitation had taken place by 1 electron, accompanied by ionization of another electron. The experiment, as shown in Fig. 2, determined when the Auger-Meitner decay took place by measuring the yield of ionized electrons as a function of the delay with respect to the attosecond X-ray pulse. Remarkably, the decay rate oscillated as a function time due to interference between Auger-Meitner decay processes originating from different excited states. This measurement reveals that motion of the excited electron on the attosecond to few-femtosecond time scale modulates the electron correlation and accordingly, the rate of the Auger-Meitner decay.

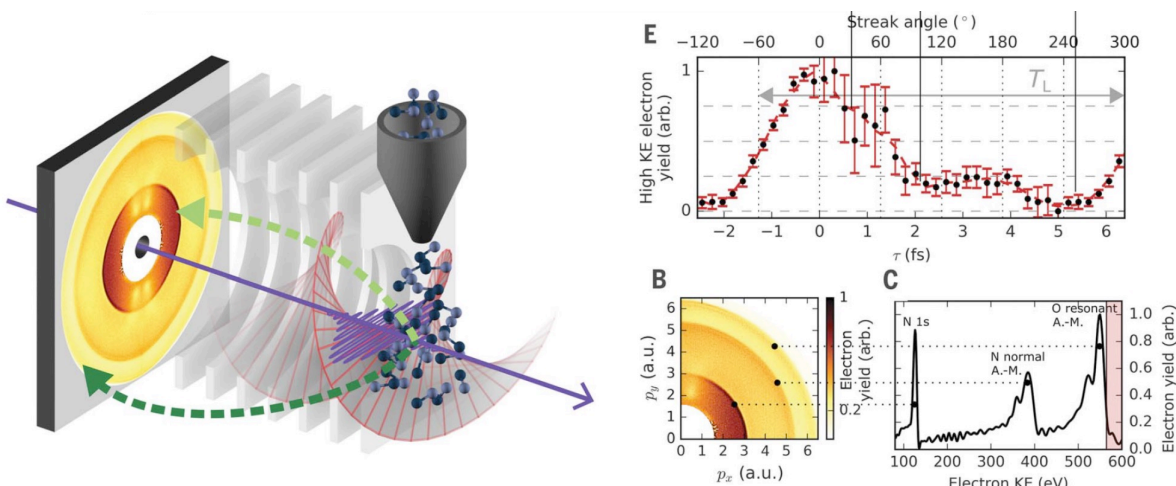


Figure 2. Observation of Auger-Meitner decay in NO. (left) Schematic view of the photoelectron measurement process used to measure the Auger-Meitner decay of core-excited states of NO in real time. By performing the experiment in the presence of a circularly polarized infrared laser field, the time of ionization was mapped onto a position on a position-sensitive detector. (right) Measurement of the time-dependence of the Auger-Meitner yield reveals nonexponential decay due to interference between different core-excited states. (Adapted from Reference 4)

Impact on Theory and Simulation: The pioneering research made possible by LCLS and, in future LCLS-II, creates unprecedented possibilities in the investigation of time-dependent electron dynamics. Accordingly, LCLS experiments are inspiring novel theoretical approaches and the theoretical investigation of innovative experimental measurement schemes. One prominent example is the work by Mukamel and co-workers on multi-dimensional X-ray spectroscopies, which are within reach through unique experimental capabilities of LCLS⁵. The researchers showed how traditional measurement protocols used in the visible/infrared region can be transferred to the X-ray domain. Moreover, LCLS experiments have led to a better theoretical understanding of X-ray-induced radiation damage.

Future Directions: The landmark experimental result of time-resolving the Auger-Meitner process merely represents the recent starting point of the attosecond research program at LCLS, but this achievement already shows the unique and unprecedented potential that the generation of attosecond laser pulses at LCLS has. In years to come, we may anticipate many breakthrough applications of attosecond pulses at LCLS, including their more widespread use in attosecond pump-attosecond probe experiments. Other examples include observations of ultrafast charge migration and control of chemical reactivity in biomolecules, as well as observation and control of electron correlation in quantum materials. For the foreseeable future, and certainly with the emergence of LCLS-II, we may expect LCLS to be the leading facility worldwide where research of this kind is possible.

Biological Sciences

Overall Assessment: Research to discover the molecular mechanisms of biology at the atomic level has advanced our understanding of biological processes. These discoveries, together with

others from the study of cells, genes, and organisms, continually grow our knowledge of fundamental biology. This knowledge has massively expanded the ability to treat or prevent many diseases, to improve agriculture, and to apply the “green” methods of biology to human endeavors. Advances in the knowledge of biology have been and continue to be dependent on advanced technologies in areas such as imaging, computing, and nanoscale instrumentation. Thanks to the powerful biology-technology combination, we can treat many cancers with biologics, diagnose diseases with molecular precision, develop new antibiotics, thwart pathogens with effective vaccines, develop green catalysts, and raise crops with energy efficiency. X-rays have long been a critical probe in understanding the structure and mechanism of biomolecules. The femtosecond time resolution and intensity of LCLS pulses have provided major advances in 2 aspects of the study of biological macromolecules. The time resolution provides an opportunity to probe the steps of chemical reactions in proteins and to understand the underlying energetics of the reactions. This is especially important for development of biology-based systems that use light energy for manufacturing or other purposes. The intensity of the LCLS pulse allows for detection of scattering or spectroscopy signals that are too weak to detect with less intense X-rays. This expands the number of biological systems that can be examined in atomic detail, especially the proteins that function within the membranes of cells.

The X-ray dose from a single LCLS pulse, however, destroys fragile protein molecules. To circumvent this problem, a plethora of techniques have been developed to flow samples through the pulsed beam and record spectroscopic or diffraction data from a single LCLS pulse. One such technique, the serial femtosecond crystallography (SFX) method, based on the concept of diffract-before-destroy, delivers a stream of micro- or nano-crystals into the XFEL beam, captures a series of diffraction images, each from 1 crystal, and assembles a complete diffraction data set from many thousands of images⁶. SFX has yielded major results in structure-function studies of extremely challenging proteins and in time-resolved studies of chemical or electronic transitions in proteins. It is an excellent example of synergistic advances in biology and technology. The method has since been extended to perform advanced time-resolved experiments beyond structural biology. SFX has been applied in diverse fields including chemistry⁷ and materials science, opening new approaches for materials discovery, synthesis, and nucleation dynamics. The success of SFX at LCLS has triggered a renaissance in serial-crystallography efforts at synchrotron sources. The advanced sample delivery methods pioneered, developed, and implemented at LCLS today are routinely used at XFELs, at synchrotrons around the world, and in fields as diverse as electron cryo-tomography.

Further, in biochemistry, the LCLS enabled experiments to probe chemical and electronic transitions at new and meaningful time scales. How nature guides a chemical or electron-transfer reaction to a specific outcome under the mild conditions of a protein is of considerable fundamental interest and is applicable to the adaptation of natural systems to new functions. Access to the femtosecond regime has enabled a major advance in understanding natural light-driven reactions, most notably how photosynthesis converts light energy to chemical energy. For many systems, short-lived intermediate states in a chemical reaction had been detected spectroscopically but visualizing them was impossible. Thus, many time-resolved experiments include SFX to probe samples for simultaneous spectroscopic characterization and direct visualization of short-lived states. Such experiments using the LCLS pulse as a probe have yielded new insights to the chemical mechanism of photodecarboxylation¹³ and the intermediate states of oxygen evolution by photosystem II^{11,12} in the final water-splitting step of photosynthesis. In other experiments, the LCLS pulse serves as an excitation pump, and a

spectroscopic probe is used to determine features such as the strength of bonds in metal centers of proteins¹⁴ and the excitation and relaxation transitions associated with cofactor reactivity¹⁵. In all cases, computational chemistry is a beneficiary of the data from the femtosecond time scales of these experiments.

Examples of LCLS impact in biological science are ligand binding to a drug target and the trajectory of a light-driven chemical reaction.

Precision Drug Targeting: Receptor proteins embedded in cell membranes signal the cell to respond to environmental changes. Such receptors are ideal drug targets for disease states where changing cellular behavior can be therapeutic. However, the drug must target the intended receptor. This is a tricky problem when the intended receptor is one of the 800 members of the human G-protein-coupled receptor (GPCR) family, which are targets for more than one-third of approved drugs. Structural details at the atomic level are needed to design drugs that bind the intended receptor and not its close relatives. Crystal structures traditionally provide the requisite detail, but it is extremely challenging to purify and crystallize complex GPCRs. SFX has enabled researchers to visualize the atomic details of protein structures when only very tiny crystals can be grown. The emerging story of angiotensin II type 1 and type 2 receptors (AT1R and AT2R, respectively) is an excellent example. AT1R helps maintain cardiac homeostasis, and AT1R blockers such as olmesartan are used to treat heart failure or high blood pressure. AT2R is a target for development of receptor-activating drugs for cardioprotection and receptor-blocking drugs to treat neuropathic pain⁸. Use of SFX at LCLS yielded initial structures of both receptors^{9,10} and ultimately the discovery that the 2 receptors bind chemically similar compounds in strikingly different poses¹⁰ (Fig. 3), information that is crucial for selectively targeting 1 receptor or the other.

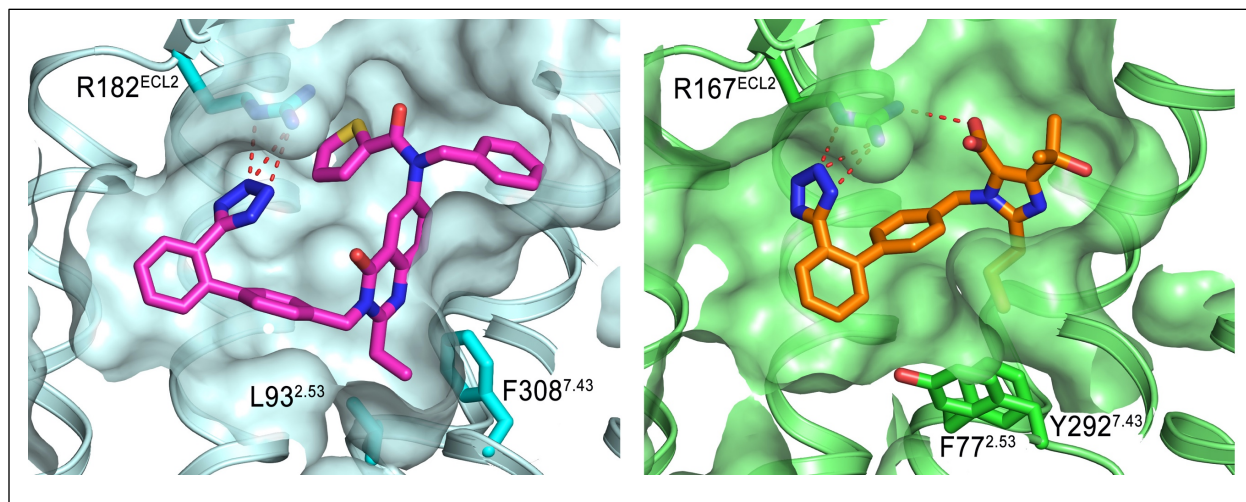


Figure 3. Ligand selectivity in angiotensin II receptors. These equivalent views of AT2R (cyan protein) and AT1R (green protein) show how similar compounds have strikingly different binding poses in the 2 receptors. (left) An AT2R-specific blocker (magenta) is bound to AT2R. (right) The metabolized, active form of the blood pressure drug olmesartan (orange) is bound to AT1R. (Image courtesy of Vadim Cherezov)

How Nature Makes Oxygen from Sunlight: Photosynthesis powers the global oxygen and carbon cycles by capturing light energy from the sun and using it to generate the oxygen in our atmosphere and produce the carbohydrates that feed the world. Proteins operating under mild

conditions perform all steps in the intricate process of transducing light energy to chemical energy. The photosystem II protein (PS II) absorbs 3 photons to catalyze the complex final reactions of splitting 2 water molecules and forming molecular oxygen. Simultaneous SFX and X-ray emission spectroscopy at the LCLS provided high-resolution views of the PS II active site in several previously established metastable states during the catalytic cycle¹¹. Most exciting, 2 previously undetected transient states were discovered, characterized, and visualized^{11,12} (Fig. 4). Understanding how the protein structure controls the sequence of events will inform the design of artificial photosynthetic systems to produce fuels from sunlight, water, and CO₂.

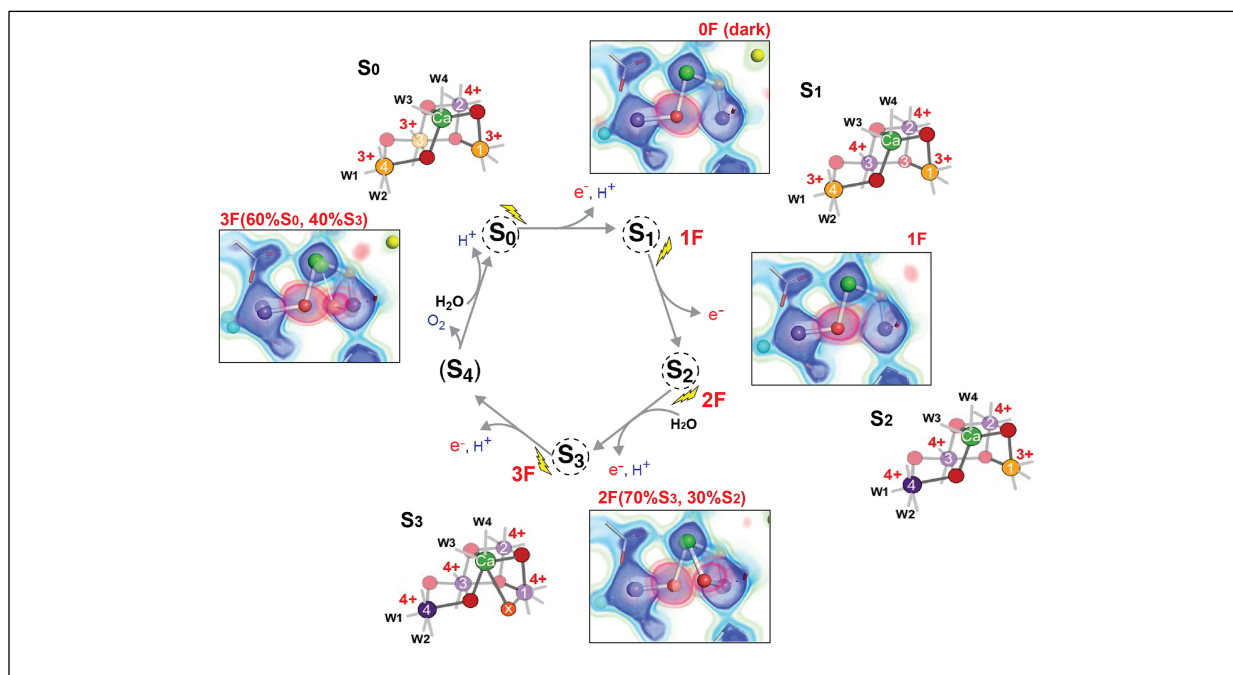


Figure 4. Reaction cycle of water oxidation in PS II. Starting in the dark-stable S₁ state, each light flash advances the oxygen evolving complex, a Mn₄CaO_n cluster, by 1 oxidation state. S₃ is the highest oxidized stable intermediate state. The next photon triggers the formation and release of O₂ via the transient S₄ state and relaxation of the cluster to the most reduced S₀ state, which is returned to the S₁ state by another light flash. The insets show SFX results for each of the S-states with electron density for the metals (green to blue contours) and for the oxygen atoms that form O₂ (orange to red contours). Manganese (Mn³⁺ orange, Mn⁴⁺ magenta), calcium (green), and oxygen (red) are shown as spheres. (Reprinted from Reference 16)

Impact on Theory and Simulation: Simulations are important to the fundamental understanding of the reaction mechanisms of enzymes. Experiments at the LCLS generate spectroscopic and structural data on a new femtosecond time scale that is relevant to chemical and electron-transfer reactions. Thus, the experiments answer the “how” question for enzyme-catalyzed reactions. Simulations ideally answer the “why” question and, by extension, provide a means to predict the behavior of other proteins. The results from experiment are synergistic with computational advances that address complex enzyme systems by combining, for example, molecular mechanics in the protein globally and quantum mechanics locally in reaction centers. The availability of new data on femtosecond reaction time scales together with improvements in theory and advances in the speed and scale of computational methods have the potential to bring

simulations to a new level of reliability and to vastly expand the ability to predict behavior for other systems.

Future Directions: The SFX method will mature, enabling the study of more challenging systems of environmental or biomedical importance. Improved sample-delivery methods will reduce the quantities needed for a successful experiment and open the method to samples that are extremely challenging to produce.

Theorists and computational chemists will assimilate new data from femtosecond time-resolved kinetic and structural experiments and improve the ability to understand and hence predict the behavior of enzyme systems. The improved predictions will be applied to the development of greener, more efficient biocatalysts via genome mining and protein engineering.

Condensed-Phase Chemistry and Catalysis

Overall Assessment: The scientific impact of the LCLS in condensed-phase chemistry and catalysis is based on its ability to address questions in the context of chemical systems that lie at the heart of efforts to create design-specific platforms for complex chemical transformations. The impacted areas range from artificial photosynthesis to advanced materials chemistry to the development of new approaches for the synthesis of pharmaceuticals and commodity chemicals. Breakthroughs in all of these areas require the development of new approaches to study the most fundamental aspects of the electronic and geometric properties of molecules and materials and of their reaction profiles, pushing beyond the boundaries of what is currently known and/or presumed to be true to realize new paradigms for chemical transformations. The use of photons to effect or probe such transformations has a long history, and its implementation in the ultraviolet, visible, and infrared regions of the spectrum is well established. The LCLS was conceived to extend these efforts into a fundamentally new regime – the X-ray region. Many of the most fundamental questions underpinning all of the concepts aimed at finding new ways for effective and efficient chemical transformations can only be answered in this regime. While the LCLS is to be credited with many significant scientific discoveries that have natural ties to very specific, applied problems in the chemical sciences, the importance of the fundamental new knowledge in condensed-phase chemistry through work at LCLS should not be underestimated as it provides the foundation for new approaches to translating fundamental understanding of chemical systems to solutions of key challenges for a sustainable future.

This report highlights 2 specific areas where work at the LCLS has had a significant impact: 1) revealing the structure, spin states, and evolution of frontier orbitals following photoexcitation and excited-state evolution of metal complexes; 2) capturing short-lived reaction intermediates in catalytic cycles (both natural and artificial). In both areas, the LCLS has made a number of seminal and paradigm-shifting discoveries, all made possible by its unique capabilities to probe atoms, molecules, and materials at a more fundamental level than was previously possible.

Structure, Spin, and Frontier-Orbital Evolution in Metal Complexes: In this section, we shall highlight examples that will illustrate how the fundamental science arising out of the LCLS has not simply offered up new discoveries. It has changed the way people think about molecules.

One of the most ubiquitous types of atoms found in both synthetic and natural chemical systems are transition metals. They form the basis of the vast majority of catalysts used in industries ranging from pharmaceuticals to fine chemicals to materials produced on the multi-ton scale. In

biology, the presence of transition metal ions defines the active sites of catalysts that allow life to exist. Additionally, transition metals represent a frontier in the development of new materials that will aid in the realization of the next quantum revolution in information science. One of the unique characteristics about metal ions is that they often possess unpaired electrons. While this is rarely seen in organic systems save for certain, specific instances, it is more common than not to encounter transition metal ions as paramagnetic components of a chemical system (both synthetic and biological). It is precisely this paramagnetic nature that is often responsible for much of the unique reactivity observed for such systems (the Mn ions in the active site of the water oxidation center of photosystem II as one such example). Most, if not all, transformations involving transition metals (e.g., photoredox catalysis or photocatalytic bond activation) involve a change in the spin properties of the metal ion or specific changes in frontier-orbital interactions as key mechanistic components of those transformations. Conventional thinking would hold that the spin and geometric properties and, by extension, the reactivity of a molecule are independent of each other. There has long been ample evidence in the literature to suggest that this assumption was not only incorrect, but was also driving research in the development of new approaches to chemical transformations relying on spin-state changes down the wrong path. Frontier-orbital interactions, in turn, being an elusive and complex fundamental quantum-mechanical or quantum-chemical property of transition metal complexes, are hard to probe experimentally.

This is precisely where work at the LCLS changed the rules of the game. Using prototypical transition metal complexes – $[\text{Fe}(\text{bpy})_3]^{2+}$ (where bpy is 2,2'-bipyridine) and $\text{Fe}(\text{CO})_5$ – and by following how these react to the absorption of visible light, researchers at the LCLS devised new ways to simultaneously probe the evolution of the spin, structure, and bonding properties of transition metal complexes (Fig. 5, left panel). Pioneering experiments on $[\text{Fe}(\text{bpy})_3]^{2+}$ revealed a disconnect between the evolution of the system along the structural and spin trajectories and provided, for the first time ever, a glimpse into how these 2 critical parameters that define the physical and photophysical properties of a metal complex play off each other (Fig. 5, middle panel).¹⁷

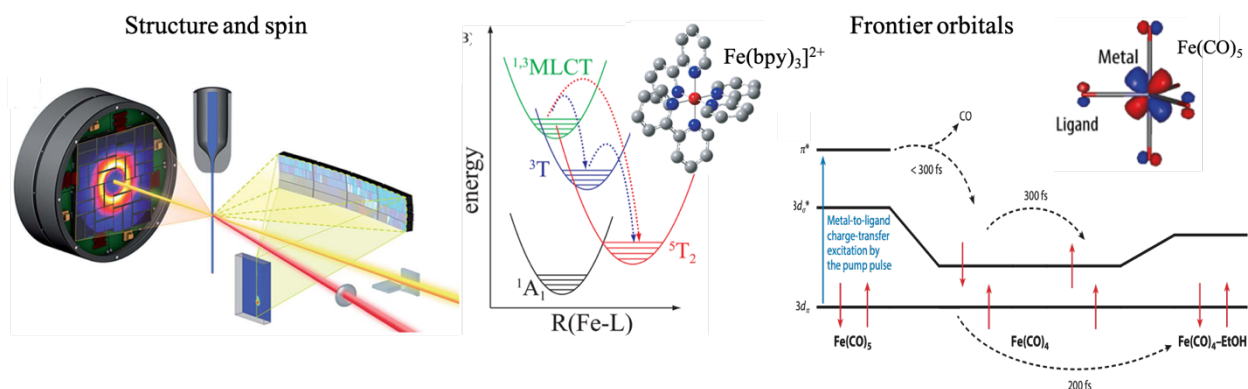


Figure 5. Structure, Spin, and Frontier-Orbital Evolution in Metal Complexes (left) Schematic of the experimental setup¹⁷. (Adapted from Reference 18) (middle) Potential energy surface diagram depicting excited-state evolution in $[\text{Fe}(\text{bpy})_3]^{2+}$ in solution following charge-transfer excitation. The involvement of the ^3T state, which had never been detected before despite decades of work on this system, was only identified due to the technology developed at the LCLS. (Adapted from Reference 19) (right) Reaction coordinate diagram for a photosubstitution reaction involving the

replacement of a bound CO molecule with a solvent (in this case, ethanol). Insight into the evolution of orbital energetics that define the interplay between the thermodynamics and kinetics governing this reaction was made possible by techniques developed at the LCLS²⁰. (Adapted from Reference 21)

In the years since this result was published, the use of ultrafast X-ray methods to simultaneously examine the reactivity of metal complexes along both of these coordinates has become the gold standard at facilities around the world and is being applied to problems ranging from solar cell technology to photoredox catalysis. This work has truly redefined the field, and its implications are continuing to reverberate across the scientific community.

In contrast to the photophysical processes illuminated by the aforementioned work, studies on Fe(CO)₅ established a new approach for probing chemical reactions, specifically those leading to bond breaking and bond formation (Fig. 5, right panel).²⁰ Specifically, the methodology pioneered at the LCLS allows for directly probing valence electrons in a transition metal complex. The valence electrons are involved in the making and breaking of chemical bonds and are therefore the key for monitoring the progress of any chemical transformation. In the case of Fe(CO)₅, photoexcitation results in the loss of one of the CO ligands (i.e., the cleavage of a Fe-carbon bond), resulting in the formation of a coordinatively unsaturated, reactive Fe(CO)₄ transient species. This species can access different reaction trajectories at this point, chief among them structural reorganization of its now 4-coordinate structure and/or sequestration of a new ligand (in this case ethanol) in a bond-forming reaction. The data acquired at the LCLS allowed the direct monitoring of this reaction sequence, therefore providing information concerning the energetics and kinetics of the system. This information would have been difficult, if not impossible, to obtain through any other means. This study has thus provided a template that can be used to probe a broad array of reactions spanning chemical, biological, and materials chemistries.

Capturing Reaction Intermediates in Catalysis: The temporal resolution and atomic specificity available through the X-rays at LCLS have proven invaluable for probing details of catalytic transformations (both natural and synthetic) that were simply indeterminable prior to the LCLS coming online. This is best demonstrated with the results from the LCLS on CO oxidation on a metal surface²² and water splitting by the photosystem II protein (PS II) complex^{11, 23}.

Elucidating the underlying reasons for selectivity in heterogeneous catalysis requires knowledge of how reaction intermediates determine reaction pathways and kinetics at an atomistic level: this is exactly what the LCLS has made accessible (Fig. 6). With time-resolved and element specific X-ray spectroscopy, researchers were able to capture the short-lived and intermediate configurations of CO molecules and O atoms on a ruthenium surface²². They could show how these two entities move and meet on the surface to form an elongated and transient form of CO-O that then transforms into the stable configuration of CO₂ before the molecule is released from the surface. Capturing short-lived reaction intermediates yields new and essential insight into how and on which time scale the binding site, local geometry, and charge state of adsorbed species enable surface reactions in heterogeneous catalysis.

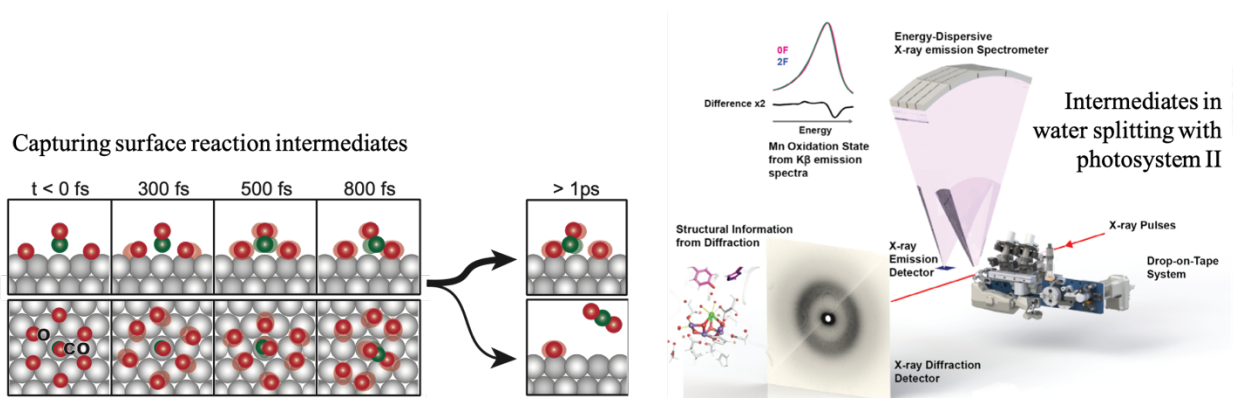


Figure 6. Capturing Reaction Intermediates in Catalysis (left) Depiction of the sequence of reaction steps for CO oxidation on a ruthenium surface: (upper row) side and (bottom row) top views of the surface²². Femtosecond X-ray spectroscopy developed at the LCLS showed for the first time how oxygen atoms are activated and collide with CO on the surface, how and when the decisive transition state is formed, and how the system either (last panel, top) is trapped in an intermediate configuration or (last panel, bottom) eventually proceeds to releasing the final CO₂ product. (Adapted from Reference 22) (b) Schematic of the approach developed at the LCLS that uniquely enabled probing atomic positions during water splitting with the photosystem II protein complex through X-ray diffraction and simultaneously mapping the local electronic structure in the water oxidation center with X-ray spectroscopy^{11, 23}. The ability to probe the system as it functions at room temperature and before it changes due to X-ray illumination was possible due to the femtosecond X-ray pulses generated at the LCLS. (Adapted from Reference 16)

The ability to capture reaction intermediates with X-rays from the LCLS also led to a quantum leap in understanding light-triggered water splitting and oxygen evolution in plants^{11,23}. The X-ray flashes were bright and short enough in duration to illuminate the system as it functioned in a room-temperature solution and in a stroboscopic fashion to render sequences of 3-dimensional maps of atomic positions and sequences of charge states of the active center of the protein¹¹. Importantly, scientists were able to see this information before the protein was damaged by the very same X-ray illumination because the X-ray flashes were shorter than the typical time scale for X-ray illumination induced damage²³. With the LCLS capability to follow the coupled structural and chemical sequences of events with simultaneous X-ray diffraction and spectroscopy at well-defined time intervals between the visible light triggers and the X-ray flashes, researchers were able to elucidate how structural parameters control the reaction kinetics in water splitting and how the protein environment and hydrogen-bonding network in the system accommodate the dynamically evolving energy landscape. Understanding what facilitates the complex proton-coupled electron transfer chemistry of the water oxidation center of PS II is critical for determining how nature solved the problem of free-energy leveling (i.e., accessing multiple oxidation states of a system without building up a Coulombic barrier to the desired multi-electron chemistry). The lessons learned can be adapted for artificial constructs to carry out other multi-electron, multi-proton transformations (e.g., the reduction of N₂ to NH₃, a 6-electron, 6-proton reaction that is the basis of the Haber-Bosch process) that employ cheap, earth-abundant elements.

Impact on Theory and Simulation: Excited-state pathways in ultrafast dynamical processes and reaction barriers in kinetically driven processes of chemical, biomolecular, and catalytic systems

always result from a complex interplay of atomic structural and electronic structural motions. Theory is essential in making predictions for the outcomes of such processes and for retrospective and detailed interpretations of experimental data. The atomic and orbital resolution capabilities of ultrafast X-ray experiments at the LCLS are invaluable for testing such predictions. With these capabilities, completely new observables, such as the real-time observation of atomic distance or molecular orbital evolution during chemical transformations are also accessible. These results can then be directly compared to simulations of the transformations, whereas before the advent of the LCLS, researchers made inferences based on the initial and final configurations. Building on this simulation-observations comparison, theoretical simulations can next be used to find ways to modulate the outcome of the process by modulating the probability of a certain dynamical pathway or modulating the height of a reaction barrier. This interactive, synergistic approach based on new X-ray experimental observables and directed theoretical descriptions is already transforming the way scientists are thinking about understanding existing chemical processes and, more importantly, designing new ones that have yet to be imagined.

Future Directions: The proven ability of the LCLS to probe dynamically evolving electronic and geometric properties of molecules with atomic resolution combined with its more than 1,000-fold increase in repetition rate and photon flux at the future LCLS-II/HE will yet again transform the fields of condensed-phase chemistry and catalysis. Leaving behind studies of model systems or demonstration cases, we will be able to probe, at functional conditions and in real time during chemical transformations, virtually any specifically designed (photo)catalyst or any metalloenzyme, irrespective of current limitations imposed by, e.g., concentration of the studied element or quantum yield of the targeted reaction path. Complete mapping of all degrees of freedom as a function of time, from the catalytically active site to the whole catalyst in solution and including information on the concerted motions of surrounding solution and substrate molecules, whether for a homogeneous photocatalyst, for a protein, at a surface, or at an interface, promises to revolutionize the way we conceive and design future systems for chemical transformations. Completely new frontiers will be accessed by hitherto unexplored combinations of new capabilities at the LCLS-II/HE, and these may include: 1) revelations of how conical intersections direct molecular relaxations in non-Born-Oppenheimer systems in solution excited by intense sub-femtosecond X-ray pulses and pairs of pulses, 2) tunable surface and interface sensitivity in heterogeneous catalytic reactions with high-repetition-rate femtosecond pulses for photoemission studies, and 3) probing the valence d-electron interactions locally at the active sites of 3d transition-metal enzymes at functional conditions with the unprecedented average femtosecond flux and sample delivery methods enabling the study of ultra-dilute systems.

Gas-Phase Chemistry

Overall Assessment: The advent of ultrafast laser techniques has revolutionized our understanding of the way in which molecules undergo chemical transformation. Using ultrafast lasers, pump-probe experiments can be configured, where a “pump” laser initiates a molecular process of interest and where a “probe” laser subsequently interrogates the time evolution of the system at a variable delay. In 1999, Ahmed Zewail, one of the pioneers of this novel field of femtochemistry, was awarded the Nobel Prize in chemistry²⁴.

Femtochemistry experiments require that the interaction of the probe laser with the evolving

molecule leads to the measurement of a suitable observable. This is where “traditional” femtochemistry experiments have run into limitations. These experiments typically use visible or ultraviolet probe lasers and therefore observe the molecular dynamics by interacting with the valence electrons, for example, measuring photoabsorption or photoionization processes. This gives the experiment a sensitivity to both structural changes and changes in the electronic configuration of the molecule, with the two often being difficult to separate. As a result, the interpretation of femtochemistry experiments often requires a quite advanced understanding of the molecular dynamics prior to the experiment. For this reason, the femtochemistry research community has been searching for measurable observables that better separate structural and electronic effects and permit extraction of the molecular dynamics directly from the experimental results. These capabilities now exist for the first time at LCLS, due to several novel and pioneering experimental capabilities. These include: 1) the ability to record time-resolved X-ray diffraction using the LCLS free-electron laser²⁵ and time-resolved electron diffraction using the LCLS UED instrument²⁶; 2) the ability to implement spectroscopic techniques based on site-specific core-level photoabsorption²⁷. Two examples below serve to highlight the impact that LCLS has had on gas-phase chemistry.

Ring Opening in 1,3-cyclohexadiene: Diffraction arises due to an interference between scattering processes that occur at multiple scattering sites and provides direct information on the distance between these scattering centers. Therefore, scattering of X-rays from the LCLS free-electron laser or of relativistic electrons provides researchers direct insight into structural transformations in molecules without needing to rely on accompanying theoretical work. These capabilities are highlighted with a study on ring opening in 1,3-cyclohexadiene, a textbook example of an electrocyclic reaction in organic chemistry and a model for photobiological reactions in vitamin D synthesis²⁶. Electron diffraction from ground state molecules reveals the presence of 2 dominant C-C bond distances in the molecule, corresponding to the bond distance between 2 adjacent carbon atoms and the bond distance between 2 non-adjacent carbon atoms located at different positions on the aromatic ring (Fig. 7, left panel). UV laser excitation (the “pump”) dramatically changes the electron diffraction pattern, with a decrease of intensity at the 2 aforementioned bond distances, and the emergence of a novel C-C bond distance that exceeds the size of the unexcited molecule (Fig. 7, left panel). As such, the experiment proves that UV excitation leads to an opening of the aromatic ring and moreover, researchers were able to determine that this process occurs on an ultrafast time scale of little more than 100 fs, due to the involvement of a conical intersection that rapidly funnels the photoexcited molecule toward reaction products. The combination of sub-ångstrom spatial and femtosecond time resolutions demonstrated in the experiment is unique and unprecedented.

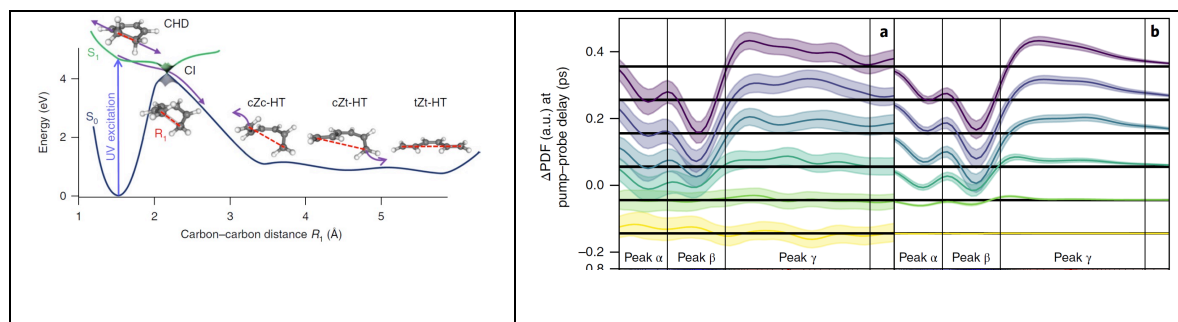


Figure 7. UV-induced ring opening of 1,3-cyclohexadiene, as probed in a UED experiment: (left) schematic of the chemical reaction, showing the reaction path via a conical intersection;

(right) (a) experimental and (b) theoretical UV-induced transient changes in the measured pair distribution functions (PDFs), showing a time-dependent decrease of short C-C distances (α and β) and increase of longer C-C distances (γ), implying that a ring opening process has occurred. (Adapted from Reference 26)

Relaxation Dynamics of Glycine: The X-ray wavelength of the LCLS permits the development of not only measurement schemes that are based on diffraction, which are ideally suited for measuring time-dependent structural changes, but also novel spectroscopic techniques that specifically address core electrons. These techniques are uniquely sensitive to time-dependent electronic changes in a molecule and moreover from the viewpoint of a particular atom within the molecule. A pioneering example at LCLS involved a study of photoinduced dynamics of glycine (Fig. 8), the simplest stable amino acid²⁷. Upon excitation of the molecule with UV light, a bonding electron was promoted to an anti-bonding orbital, leaving behind a hole in the valence shell. However, this hole was not static and rapidly localized at an atomic site. In terms of the potential energy curves of the molecule, this process corresponds to a $\pi\pi^* \rightarrow n\pi^*$ internal conversion via a conical intersection. Using time-resolved near edge X-ray fine structure absorption (tr-NEXAFS) around the oxygen K-edge, this process could be directly observed, exploiting the significantly different X-ray absorption cross sections for the $\pi\pi^*$ and $n\pi^*$ electronic configurations. From the time delay between the UV laser-induced depletion of ground state contributions in the NEXAFS spectrum and the emergence of new features related to the excited state population, researchers could determine that this purely electronic rearrangement of the molecule took about 60 fs.

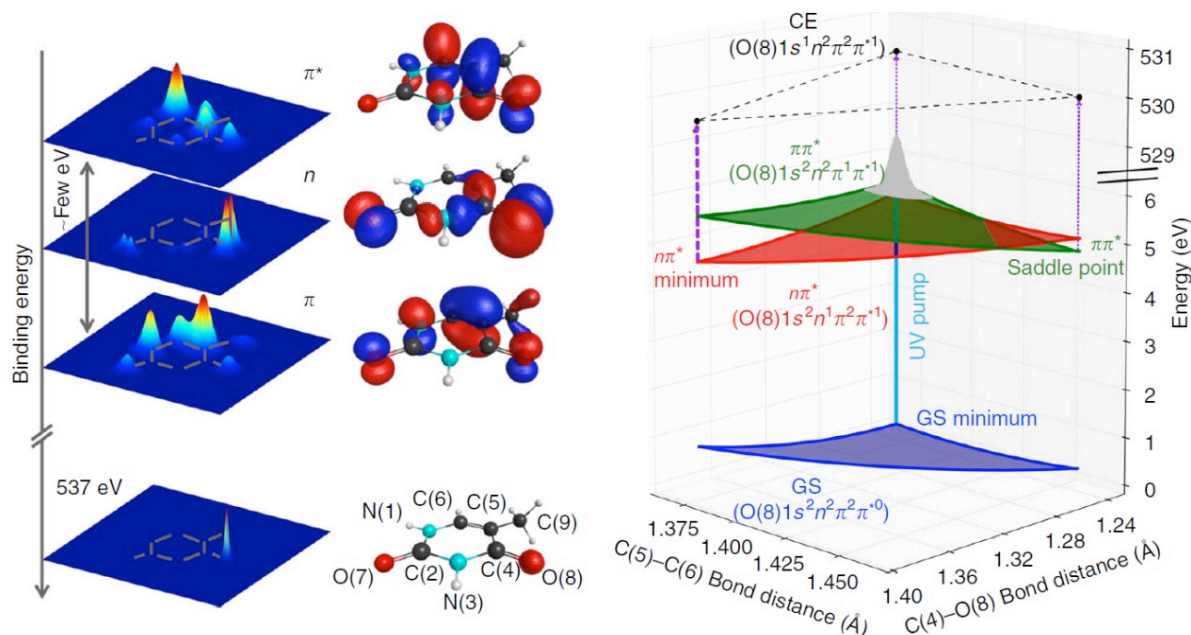


Figure 8. Photoinduced dynamics of glycine: The absorption of UV light promotes a bonding electron to an anti-bonding orbital and is rapidly followed by a relaxation process via a conical intersection. At LCLS this process was observed in real time using tr-NEXAFS around the oxygen K-edge. (Adapted from Reference 27)

Impact on Theory and Simulation: The availability at LCLS of intense, tunable X-ray pulses that can be used in transient absorption experiments has stimulated the development of novel

computational approaches where the time-resolved photoinduced molecular dynamics is calculated along with its “on the fly” observation via X-ray, electron diffraction, or NEXAFS²⁸. Moreover, the X-ray and electron diffraction experiments at LCLS have led to deeper theoretical understanding of the role of structural and electronic contributions to elastic and inelastic scattering processes.

Future Directions: The investigations described above could already be successfully carried out using the 120 Hz repetition rate of the LCLS. The 1 MHz repetition rate of LCLS-II will open completely new possibilities. An exciting example is the use of Coulomb explosion imaging (CEI) for monitoring time-dependent structural dynamics. The high intensity of the focused X-ray pulses at LCLS makes it possible to ionize single molecules dozens of times within a single X-ray laser pulse. Such an ionization process makes the molecule highly unstable and leads to an explosion into many charged fragments. With current detection systems, it is possible to measure the three-dimensional velocity of every one of these charged fragments simultaneously. As shown in recent experiments at the European XFEL (the European counterpart of the LCLS, which can produce as many as a few thousand X-ray pulses per second), a precise measurement of all these velocities allows a reconstruction of the structure of the intact molecule before the Coulomb explosion occurs²⁹. The MHz repetition rate of LCLS-II will make it possible to implement such CEI measurements as part of a pump-probe sequence and will thus create the possibility to measure snapshots of evolving molecular structures right after a pump laser has initiated a structural rearrangement. Experiments like this, and many more that the unique properties of LCLS and LCLS-II inspire, provide a unique view of the structural and electronic response of molecules to incident light, and as such, lay the foundation for a better understanding and, consequently, more widespread exploitation of photoinduced molecular processes.

Materials Science and Condensed Matter Physics

Overall Assessment: Future advances in renewable energy production and storage, quantum information, computing technologies, and sustainable manufacturing rely on achieving precise understanding and control of phenomena in materials science and condensed matter physics. The most challenging problems involve many atoms or electrons, exhibit coupling across multiple length scales, amplify small perturbations, and/or involve fluctuations and nanoscale heterogeneity that enable the emergence of novel phases. Such phenomena span a range of length- and timescales, including those that are uniquely accessible by LCLS (and other XFELs) but not accessible via other techniques including optics, synchrotron radiation, and electron microscopy. LCLS, with complementary advances in theory, simulation, and materials synthesis, has vastly expanded the range of materials phenomena that the community can study and ultimately control. Key strengths of LCLS in this area include time resolution, X-ray optical coherence, brilliance, and chemical selectivity of the experimental capabilities of LCLS. Two areas that serve to highlight the impact that LCLS has had in condensed matter physics and materials science are described below.

Control of Magnetic Order in Multiferroic Materials: An important frontier in condensed matter science is to develop the physical principles that provide the means to control the interaction of usually separated phenomena. These principles are challenging because they can involve interactions that are not typically considered in the predictions of the steady-state configurations of the materials. Emerging physical concepts to meet this challenge include coherence in the

excitation of new phenomena, which is the potential to use specific modes of excitation, for example a specific terahertz (THz) frequency, to induce changes in specific modes of responses such as the magnetization or other subtle forms of order. Experimental progress in this area prior to LCLS had been difficult because the time scales of the phenomena of most interest lie in the femtosecond to picosecond (ps) regimes where specific THz optical modes offer important resonances with the responses of materials. Ultimately, the opportunity to create and control new phases of matter with new functional properties is enabled by harnessing coherent light-matter interactions.

Multiferroic materials are compounds that exhibit both magnetic order and order in their electronic systems. The existence of these multiple simultaneous interacting forms of order suggests that their dynamics might be related and that an optical signal may provide the means to control both forms of order. The magnetic order in multiferroic materials is of particular interest due to potential applications in devices employing the dynamics of magnetism at the nanometer scale, termed spintronics. The frontier of spintronics involves developing the means to change the magnetic order using electric fields at very short time scales. However, the underlying physics and ultimate speed of magnetoelectric coupling needed for such precise selection of the changes remains largely unexplored.

Experiments at LCLS using ultrafast resonant X-ray diffraction revealed that the means for the control of spin dynamics exists in a prototypical multiferroic material with the composition TbMnO_3 . The predicted coupling of magnetism and electronic order in this system is termed an electromagnon mode – a simultaneous oscillation of the electronic and magnetic order. The LCLS study discovered that exposure to a brief intense few-cycle THz light pulse, consisting of only a few electric field cycles, coherently excited specific modes of electromagnons, as shown in Fig. 9. By tuning the excitation into resonance with the electromagnon mode, the experiments demonstrated atomic-scale magnetic structures can be directly manipulated with an electric field of light on a sub-picosecond time scale³⁰.

Further experiments at LCLS have revealed that the principles of control can be extended to a broad range of condensed matter excitations. These areas include the organization and dynamics of novel forms of chiral electronic order, termed ferroelectric vortices. Experiments at LCLS discovered that these new forms of order lead to new modes of excitation and that such excitations are coherently excited by THz optical pulses³¹.

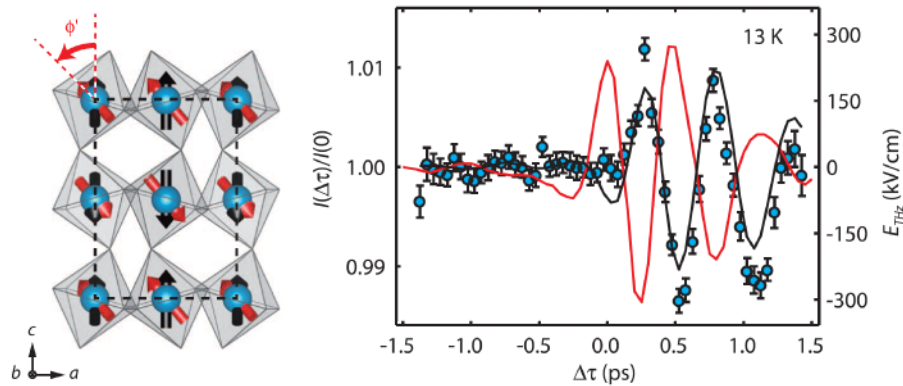


Figure 9. Toward coherent control of solid-phase excitations and coherent oscillation of the magnetic spins within the multiferroic material TbMnO_3 . (left) Pattern of the oscillation of the spins: The excitation results in a specific pattern of tilting between the excited and steady-state

spin pattern, as indicated by the red and black arrows, respectively. (right) Time dependence of the electromagnetic field, shown as the red lines: The X-ray scattering signature of the spin oscillation, shown as the filled circles, coherently follows the THz excitation. (Adapted from Reference 30)

Temporal Dynamics of Highly Disordered Materials: Dynamic heterogeneity, fluctuations, and disorder at the nanoscale are ubiquitous in functional materials and in processes ranging from electronic materials for information storage to advanced manufacturing. These areas each exploit transient nonequilibrium processes to create new material phases with novel microstructures and properties. Despite their broad impact across multiple classes of materials and time scales, dynamics in heterogeneous systems have remained extremely challenging to understand because the times over which materials reconfigure are extremely short and the length scales involved can be as small as the distances between atoms. The scientific impact of LCLS in this area arises from the transformative capabilities to capture the atomic structure and possibly stochastic dynamics of complex materials without long-range order at the fundamental atomic scale and under the transient nonequilibrium conditions where new metastable phases nucleate and emerge.

A particularly compelling example of transformative LCLS science in this area is in the observation of the structural evolution of phase change materials important for computer memory applications. Here, studies using time-resolved diffuse X-ray scattering revealed that a liquid-to-liquid phase transition underpins the phase-change memory (PCM) process^{32,33}. In PCMs, the volume of material storing 1 bit of information is cycled between glassy and crystalline states, as shown in Fig. 10, providing nonvolatile information storage. The dynamics of the glassy-crystalline transition are crucial and had not been resolved on an atomic scale. More fundamentally, this is a region in which LCLS helped to address a significant gap in experiments, theory, and computation. Previous uncertainty regarding the atomic structural dynamics, in particular the temperature dependence of the atomic mobility, had precluded a directed optimization and design of PCM materials.

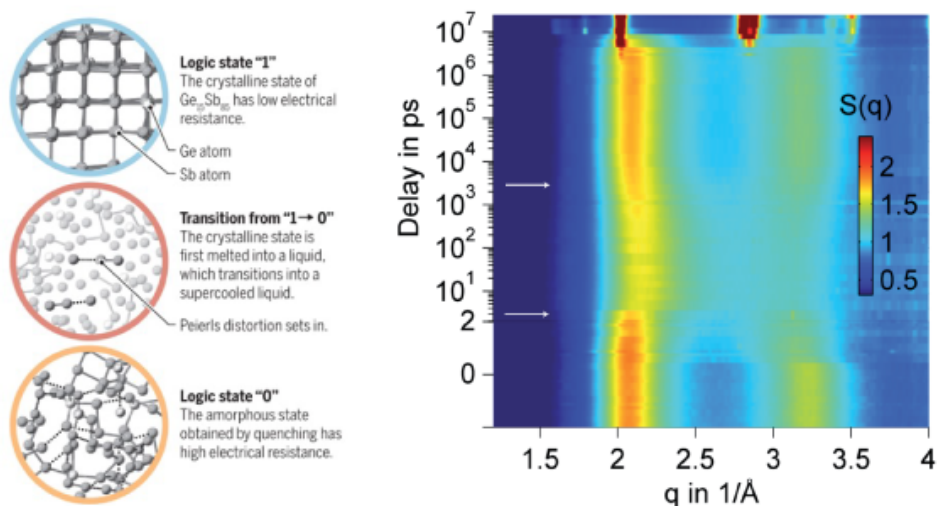


Figure 10. Dynamics of highly disordered phase-change materials. (left) Atomic-scale configurations of the bits “1” and “0” stored in the phase-change memory, and the mechanism of the change from crystalline to amorphous phase that underpins these devices. (right) X-ray signatures of the phase transformation, visualized as the time evolution (measured in ps) after

optical excitation of the X-ray scattering function $S(q)$ that indicates the degree of order at spatial frequency q in the PCM material. (Adapted from References 32 and 33)

Time-resolved diffuse X-ray scattering studies at LCLS provided the first atomic-scale view of the melt-quenching and crystallization process (Fig. 10) and represent a significant qualitative advance in our understanding in this area. These studies revealed that 2 distinct liquid phases are at the heart of PCM functionality, namely a strong liquid with reduced mobility and a fragile liquid with higher mobility, yielding a new design principle: to control the threshold for the liquid-liquid phase transition, compared to the threshold for melting, to maximize the temperature window for operation.

Impact on Theory and Simulation: A common feature among the areas of impact of LCLS in condensed matter physics and materials science has been that experiments at LCLS have stimulated new directions of theory and simulation. In the area of coherent manipulation of solids, new theoretical and computational efforts have been stimulated to describe and predict new phenomena in light-matter interaction, to predict the dynamics of the excited electrons and other excitation modes, and to discover novel modes of collective excitations and mechanisms for ultrafast control that are now within reach of experiments³⁴. Theoretical efforts have described and elucidated effects at length scales ranging from atomic to mesoscopic and have been based on density functional theory, molecular dynamics, and continuum models. Examples include the development of methods that fundamentally enable the interpretation of spectroscopic and scattering experiments to extract previously unknown parameters of electronic models³⁵. More generally, LCLS experiments have stimulated the development of advanced combined computational/experimental methods that include artificial-intelligence-guided data analysis and strategies for comparing theory with vast arrays of experimental data.

Future Directions: Two novel techniques to probe materials properties have been developed at LCLS, and we expect these techniques to be applied to important problems in materials in the future. Fourier-transform inelastic X-ray scattering enables the measurement of low-energy collective modes in condensed matter to map the dynamic structure factor in the nonequilibrium regime. This technique measures low-lying excitations with very high resolving power, orders of magnitude better than previous inelastic scattering measurements based on dispersive techniques³⁶. Time-resolved Bragg coherent diffractive imaging³⁷ exploits spatially coherent X-rays via the information content in the speckle pattern in the vicinity of Bragg peaks to reconstruct the morphology and internal lattice deviations of crystals with nanoscale spatial resolution and picometer deformation. Both techniques rely on the unique properties of LCLS and will benefit greatly from the enhanced performance expected with LCLS-II.

The impact of LCLS in the coherent control of materials phenomena is exciting because it provides new principles for the design of materials exhibiting enhanced interactions between functional properties of materials, including magnetism, electronic order, and superconductivity, with the optical and electrical signals that can be used to control them. The studies of multiferroics highlighted here have opened the broader field of the coherent evaluation and control of the dynamics of solids. Future work at the LCLS (and its future upgrades) can reveal the interactions among the fundamental excitations of solids such as phonons, magnons, and topological excitations with far more precision than has previously been possible. Understanding these excitations and developing the means to control them via materials synthesis and to excite them specifically via electromagnetic fields has tremendous potential for future scientific and technological impact. In the area of materials research, the liquid-structure and phase

transformation insight, only available from LCLS studies, points to the future science impact of LCLS science in related areas of advanced manufacturing, such as flash annealing, laser ablation, and additive manufacturing, which seek to exploit nonequilibrium conditions to create new materials and structures with novel properties and with greater energy efficiency. These and other areas of future impact will employ the spatial and temporal coherence of the X-ray radiation at LCLS in new ways, building on areas of technical advances made in the first years of LCLS operation.

Materials in Extreme Conditions

Overall Assessment: There can be little doubt that LCLS has had a transformative effect on the study and understanding of materials under extreme conditions. Before the advent of LCLS, ultrafast diffraction from dynamically compressed matter was already a burgeoning field, but, based on relatively crude laser-plasma generated X-ray sources, diffraction data was of poor quality and sparse, owing to low repetition rates. The exquisite diffraction patterns that can be produced on femtosecond time scales at LCLS rival those obtained at synchrotrons in quality, and data throughput is high. LCLS has made major contributions to the understanding of shock induced plasticity and polymorphic phase transformations, and a large and growing international community now pursues this field. Furthermore, as the focused output of LCLS can attain intensities, to direct onto targets, that were previously confined to ultrafast lasers in the optical domain, new vistas in the study of dense plasmas have been opened, significantly impacting the understanding of such systems, requiring rigorous quantum approaches to model warm dense matter, and providing new insights into plasma properties. Three areas that serve to highlight the impact that LCLS has had in materials in extreme conditions are described below.

Material Deformation and Plasticity: Several aspects of the science of materials that underpin important parts of the DOE/NNSA (National Nuclear Security Administration) stockpile stewardship program require knowledge of a material's strength (i.e., its resistance to deformation) at ultrahigh strain rates. Being able to predict how metals, in particular, deform under impact is clearly key for many aspects of national security. What has long been sought after is a coherent thread of understanding from the microscale of atomic dimensions, via the mesoscale of dislocations and grains, up to the macroscale phenomena simulated in large-scale hydrocodes relevant to the final situations of interest. As reported in the 2019 review, femtosecond diffraction experiments at LCLS looking at shocked tantalum showed, for the first time, how lattice rotation and twinning varied as a function of shock strength³⁸. The experiments used polycrystalline targets with a specific texture (i.e., distribution of the orientation of the grains) such that the azimuthal position of intense diffraction regions within Debye-Scherrer rings encoded the angle that the lattice planes made with the sample normal. As the material was shocked, the lattice rotation increased with pressure. The observed lattice rotation compared relatively well (but not perfectly) with classical molecular dynamics simulations, but computational limits restrict such atomic-level simulations from encompassing the full phenomena (such as, for example, grain-grain interactions). The data showed a "knee" (see Fig. 11, right panel) that at the time could not be explained.

However, further analysis of this groundbreaking data, using crystal-plasticity simulations that are further up the length scale³⁹, have led to a much deeper understanding of the dislocation dynamics and multiplication mechanisms that now give an almost perfect match between

experiment and theory, and resolve the initially perplexing “knee” in the measured lattice rotation as a function of shock pressure.

Importantly, these simulations specifically further inform the best analytic model we have for tantalum, the so-called Livermore Multi-Scale model that can be used in hydro simulations. For the first time, thanks to LCLS, the micro-meso-macro link has been successfully made. Admittedly, this result is for one material with a specific texture (i.e., orientation of grains and grain sizes, etc.), but this should not detract from the highly significant fact that well-designed femtosecond diffraction experiments are informing fundamental questions about materials science that are directly related to stock-pile stewardship in a way that has never been done before.

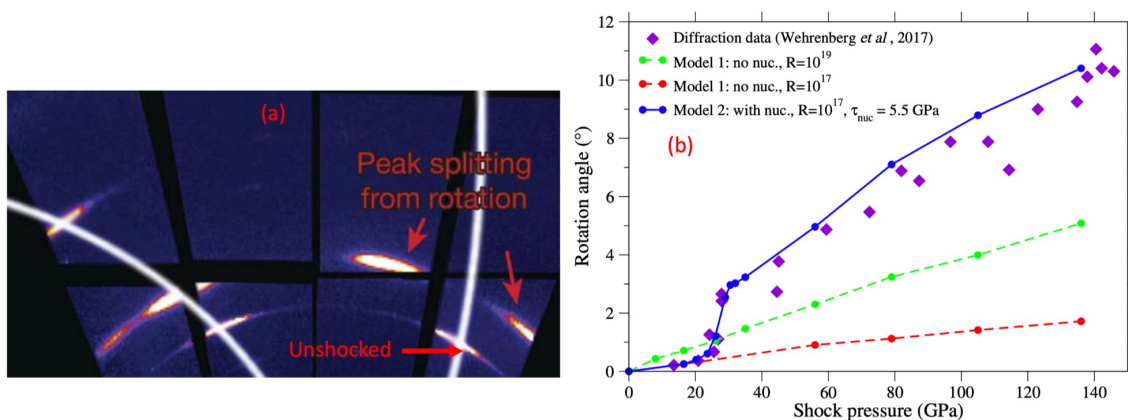


Figure 11. Femtosecond diffraction experiments on shocked tantalum. (left) Diffraction from tantalum shocked to 1.6 megabar (Mbar). The Debye-Scherrer rings come from the {110} planes. When shocked, the ring expands, and the region of high diffraction intensity splits in two and moves azimuthally, from which the angle of the lattice with respect to the target normal can be measured. (right) The diamonds show the measured angle as a function of shock pressure. The “knee” at 25 GPa can now be explained by modifications to the Livermore Multi-Scale model, thus linking micro physics to a macro model. (Adapted from References 38 and 39)

Dynamic Compression and Planetary Science: Diffraction experiments at LCLS have pioneered our understanding of polymorphic pressure (shock) induced phase transitions at ultrahigh rates. Highlights have included the observation, within a few hundred picoseconds, of the generation of the most complex crystalline phase that a single element can exhibit – the so-called incommensurate host-guest structure (e.g., see Reference 40). These experiments, where the compression is applied for a few nanoseconds and the diffraction pattern captured in under 100 femtoseconds, produced exquisitely detailed data owing to the monochromatic nature of the FEL beam and the lack of background radiation (in contrast to laser plasma X-ray sources, which are inherently noisy).

Furthermore, novel materials synthesis and chemistry have been investigated via atomic-scale structural dynamics of hydrocarbons under conditions comparable to 10,000 km below the surface of Neptune (~150 GPa, ~5,000 K). These conditions were transiently created via a 2-step laser shock front⁴¹.

The impact of these experiments is that they are, in principle, enabling the study of a whole new region of the phase diagram of matter. This is because dynamic compression can (at least with slightly more energetic optical lasers than those currently sited at MEC) produce pressures that are not achievable by static techniques. This achievement could in the future take us to pressures well above a terapascal (TPa, about 10 Mbar), although that ultimate goal is likely to require upgrades to the optical lasers that sit alongside the FEL, as we discuss in the section on future developments.

Thus, LCLS has pioneered, and now has the opportunity to further explore, states of matter that have hitherto not been produced on earth. However, experiments at LCLS have already demonstrated that there can be significant differences between the phases observed, and/or phase boundaries observed, under dynamic compression, as compared with static compression. This raises the question: Are these differences due to shear induced by uniaxial compression, due to kinetic effects, or both? Legitimate claims of relevance to planetary science, in many instances, will require a better understanding of the effects of the up to 25 orders of magnitude difference in time scale between LCLS experiments and planetary formation.

Physics of Dense Plasmas: From the very start of operations, LCLS has led the way in high intensity X-ray-solid matter interactions to produce highly ionized plasmas at exactly solid density (e.g., see Reference 42). A series of experiments have brought into question the electronic structure of very dense plasmas. This has had a huge impact on the field and has promoted a significant amount of theoretical work geared toward treating such systems from a fundamentally quantum-mechanical point of view – i.e., approaching the physics of such systems as much from a condensed matter perspective as from a plasma perspective. Innovative high energy density (HED) experiments and scattering techniques are starting to provide direct information on kinetic processes (such as collision rates) within such systems.

Part of the power of LCLS in this field results from its ability to, for the first time, uniformly heat solid matter, via penetrating X-rays, to multi-million-degree temperatures on a time scale short enough for the system to remain at exactly solid density. Most previous approaches to create hot dense plasmas almost inevitably produced large gradients in temperature and density.

As a result of this work, there is now a far better realization of what is needed to understand these so-called “quantum plasmas.” In simple terms, this means breaking free of the traditional demarcation in plasma physics of designating electrons as either bound or free and taking an attitude more aligned with a quantum solid-state approach, which considers a spectrum of “boundness” and “localization” to better understand the plasma properties.

Impact on Theory and Simulation: The experimental work at LCLS has had a significant impact on the way researchers approach modelling and data visualization. For example, in the case of dynamic compression experiments, where the use of X-ray diffraction is ubiquitous, modelers now routinely use molecular dynamics or crystal plasticity codes to directly output simulated diffraction profiles, not only to compare with experimental data after the event, but also to inform the design of experiments before they are performed. Furthermore, and as alluded to in the section above, experimental data has informed the plasticity models, spanning the length scales.

There has also been a huge impact on atomic kinetics calculations that are widely used within the optical laser-plasma community. In particular, the intense X-ray excitation that LCLS affords means that many standard codes, which had not included the relevant atomic configurations (or

super-configurations) within them, do not accurately predict the observed spectra, and that has led to considerable efforts to create codes specifically designed for XFEL-matter interactions. Furthermore, and relevant to the above discussion, the observation of ionization thresholds at energies differing from standard theory⁴³ has led to many groups now employing density functional theory to attempt to understand matter under these conditions.

Future Directions: We envisage several new opportunities that will emerge over the coming years, although many of these will require developments and investment in the optical lasers that sit alongside the FEL, as much as (if not more than) in the FEL technology itself. In addition, resolving challenges in several areas will require increased theoretical efforts.

We note the design concept, from the MEC Upgrade Conceptual Design Report⁴⁴, envisions a kilojoule (kJ)-class optical laser sited alongside the FEL facility, although the repetition rate envisaged will still be relatively low compared with, for example, the 0.1 kJ system at the European XFEL. This certainly should enable access to the TPa regime during nanosecond planar compression experiments. The high intensity petawatt system will also enable new science, and it is pertinent to note that most of the work in HED science to date has come from the nanosecond systems, with exploitation of the high intensity optical regime not yet garnering as much attention.

It is also important to observe that almost all the experiments performed on shocked matter at MEC have been performed in planar geometry. Presumably, this is partly because there are only 2 beams and partly because in shock (or ramp compression) experiments, it is often desirable to measure pressure, which is done via VISAR (a technique that effectively uses the rear surface of the target as an interferometer to measure surface motion). However, as is well known in the Inertial Confinement Fusion (ICF) community, much higher pressures can be achieved in converging geometries. This could well be worth exploring, even at modest laser energies, whether cylindrically or spherically converging experiments could be conducted, potentially opening a whole new set of experimental conditions.

We also note that experiments to date have largely involved either compressing solid state matter from ambient conditions to high pressures, or isochorically heating ambient matter to produce solid density high temperature plasmas. No work has yet been performed combining these 2 approaches, which would lead to even further as-yet-unexplored regions of the phase diagram of matter.

Summary and Future Outlook

LCLS has transformed the ability to study chemical, biological, and materials processes and dynamics at the atomic scale, with elemental specificity and on unprecedented time scales. In a decade, the LCLS has transitioned from creating and optimizing a photon beam of extraordinary brightness to developing the tools required for making and interpreting unique measurements to applying these to important, long-standing science problems. The speed and impact of this transition rivals those in other scientific fields. Moreover, the community is strongly engaged in developing the new precision techniques needed to pursue the transformational science enabled by LCLS, as measured by the large number of unique users and user visits, the demand for beam time, and the high scientific productivity of the facility. Finally, LCLS has changed how we perform light source science from relatively small teams to larger groups, similar to the particle

physics community. This transition, while increasing the overall impact of LCLS in the long term, may have impacted near-term productivity.

Given the transformative nature of LCLS, the first decade of operations has been characterized by discovery science, pioneering method development, and important demonstration experiments that highlight the scientific potential of this new X-ray source. In the second decade, transformative expansion of LCLS capabilities continues, with a pending ~10,000-fold enhancement in coherent power in the soft and tender X-ray regimes (LCLS-II, 2023), extending to >13 keV in the hard X-ray regime (LCLS-II-HE, 2028). At the same time, LCLS research is expanding beyond discovery science to programs with deeper impact in key application areas, including complex “real-world” materials, functional chemical systems and assemblies, and biological complexes operating in near-physiological environments. We note that this increase in capability and scope of LCLS science will require additional operational support to fully realize the scientific potential of LCLS.

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Appendix I: Charge to the LCLS Scientific Impact Assessment Committee

Dr. Stephen Streiffer, Vice President for SLAC, Stanford University, provided the following charge to the committee:

The goal of this assessment is as follows:

- 1) Identify and articulate the key scientific and technical achievements resulting from research that has been performed at LCLS, in a manner that is accessible to the broader community of scientific, political, policy, and public stakeholders.
- 2) Describe how these achievements have been transformative within their field or discipline, along with broader impact as appropriate.
- 3) Very briefly outline the committee's sense of future opportunities that LCLS can address, including very high-level indications of how free-electron laser science must advance in order to address these opportunities.

Appendix II: Bios for the LCLS Science Impact Assessment Committee

Antoinette (Toni) Taylor (Chair) is a Laboratory Fellow at Los Alamos National Laboratory (LANL). Formerly, she was the Associate Laboratory Director for Physical Sciences at LANL, overseeing R&D of ~1,000 researchers in physics, materials science, manufacturing science, and accelerator technologies. She received her bachelor of science, master of science, and PhD degrees in physics from Stanford University where she was a Hertz Foundation pre-doctoral and doctoral fellow. At LANL, she has served as the director of the Center for Integrated Nanotechnologies, a joint Sandia/LANL nanoscience center funded through DOE Basic Energy Sciences, the leader of the Materials Physics and Applications Division, and the Deputy Associate Director for Chemistry, Life and Earth Sciences. Her research interests include ultrafast dynamical processes in quantum and nanoscale materials, electromagnetic metamaterials, and the development of spatially and temporally local probes. Taylor is the author or co-author of over 370 peer-reviewed articles resulting from this research. She has served in professional societies and chaired international conferences in these areas. She is a fellow of the American Physical Society, Optica, and American Association for the Advancement of Science. Taylor is a member of the SLAC Scientific Program Committee and chairs the LCLS Proposal Review Committee for Hard Condensed Matter.

Paul Evans is a Professor of Materials Science and Engineering at the University of Wisconsin-Madison. His research interests include the development and application of X-ray scattering methods addressing scientific challenges in electronic materials, as well as the synthesis and characterization of complex oxides. Evans is a user of U.S. and international free-electron laser and synchrotron light sources, including the LCLS. He serves on multiple advisory committees and standing review panels in the light-source community. He is the author or co-author of more than 150 papers and is an inventor on 11 issued or filed patents.

Claudio Masciovecchio, Director of the FERMI FEL, Elettra Sincrotrone Trieste Laboratory is an experimental physicist with a track record that includes more than 200 articles in peer-reviewed journals and over 100 invited talks and plenary lectures at international congresses. His expertise ranges from the study of liquids to samples in extreme thermodynamic conditions and from nanomaterials to biophysics. Masciovecchio has well-established experience in the development, construction, and exploitation of complex instrumentation for research in physics, chemistry, and biology, as demonstrated by the international awards he received: Kai Siegbahn Prize (Uppsala, Sweden, 2012), Innovation Award on Synchrotron Radiation (Berlin, Germany, 2015), and Outstanding Scientist Award (Bari, Italy, 2016). He is principal investigator of a number of projects as, for instance, an European Research Council grant devoted to the development of ultrafast nonlinear optics with chemical selectivity. He is currently the general director of the FERMI FEL, a user facility that attracts scientists worldwide to carry out research at the cutting edge in the fields of chemistry, physics, and biology.

Jim McCusker was born in New Haven, Connecticut, in 1965. A graduate of Bucknell University (where he majored in chemistry with minors in physics and music), McCusker enrolled in the doctoral program at the University of Illinois at Urbana-Champaign in 1987 and carried out research in physical-inorganic chemistry under the guidance of Professor David N. Hendrickson. He was awarded a 2-year post-doctoral fellowship from the National Institutes of Health in 1992 to work with Professor Thomas J. Meyer at the University of North Carolina,

then began his independent academic career at the University of California at Berkeley as an assistant professor of chemistry in the fall of 1994. The initial paper out of his group was the first application of femtosecond spectroscopy to understand the photophysics of a coordination charge-transfer complex, specifically, a delineation of the ultrafast nature of excited-state evolution in $[\text{Ru}(\text{bpy})_3]^{2+}$. While at UC Berkeley, McCusker was awarded the Department of Chemistry Teaching Award in 1999 in addition to being named an Alfred P. Sloan Fellow (1998-2000) and a Hellman Faculty Fellow of the University of California (1997-1998). McCusker moved his research group to Michigan State University (MSU) in 2001 where he is currently MSU Foundation Professor of Chemistry. The central themes of his research group continue to focus on a synergy between synthesis and ultrafast spectroscopy to probe the light-induced properties of transition metal-containing molecules – as they impact the development of solar energy conversion strategies and photoredox catalysis – as well as the interplay of spin, magnetism, and the excited-state dynamics of molecular systems. An associate editor for physical-inorganic chemistry at Chemical Science since 2015, he has been recognized at Michigan State with the 2014 Junior Faculty Mentoring Award, the 2016 College of Natural Science Outstanding Faculty Award, and the 2018 William J. Beal Outstanding Faculty Award by the President of Michigan State University. He was named a fellow of the Royal Society of Chemistry in 2016, a recipient of their Chemical Dynamics Award in 2020, and most recently a recipient of the 2023 Inter-American Photochemical Society Award in Photochemistry. He has published nearly 100 articles in journals including Science, Nature, Chemical Science, and the Journal of the American Chemical Society, was elected chair of the 2012 Electron Donor-Acceptor Interactions and 2023 Photochemistry Gordon Research Conferences, and has given in excess of 300 invited seminars at universities and conferences in more than 20 countries around the world.

Janet Smith is the Martha L. Ludwig Distinguished University Professor of Biological Chemistry, Rita Willis Professor of the Life Sciences, Professor of Biophysics, and Associate Director of the Life Sciences Institute at the University of Michigan. She is an internationally recognized leader in structural biology who has made path-breaking contributions in the use of X-rays to determine crystal structures of proteins. Her research takes a structure-based approach to understand the function of biosynthetic enzymes, viral toxins, and antiviral proteins. She has degrees in chemistry (bachelor of science, Indiana University of Pennsylvania) and biochemistry (PhD, University of Wisconsin). She was a founder and is currently scientific director of the General Medical Sciences and Cancer Institute Structural Biology Facility (GM/CA) facility for macromolecular crystallography at the Advanced Photon Source. The GM/CA team designed and implemented a world-leading microcrystallography capability that opened the way for many advances in structural biology. Smith has published more than 200 papers. She has chaired or served on numerous advisory panels and boards for the National Institutes of Health (NIH) and many other national and international scientific committees and research facilities. Smith is a fellow of the American Association for the Advancement of Science (2008) and the American Crystallographic Association (2018), a member of the National Academy of Sciences (2020), and a recipient of the Dorothy Crowfoot Hodgkin Award from the Protein Society (2021) and the Mildred Cohn Award in Biological Chemistry from the American Society for Biochemistry and Molecular Biology (2022). She received an NIH MERIT Award (1998) for her productive and impactful research and service to the scientific community. Smith is a member of the SLAC Scientific Program Committee.

Marc Vrakking, Director at the Max-Born-Institute and professor for Ultrashort Physics at the Freie Universität Berlin. Vrakking completed his PhD at the University of California at Berkeley in 1992. After postdoctoral positions at the National Research Council (Ottawa) and the Vrije Universiteit Amsterdam, he led a scientific group at the FOM Institute for Atomic and Molecular Physics (AMOLF) in Amsterdam from 1997 to 2011. While at AMOLF, he led a research program focusing on the use of ultrashort (femtosecond and attosecond) XUV and X-ray laser pulses in studies of time-resolved atomic and molecular dynamics. In March 2010 he was appointed as director at the Max-Born Institute (MBI) in Berlin and as a professor of physics at the Freie Universität Berlin. At MBI, Vrakking is the head of Division A (“attosecond science”) and leads a team of researchers that are both further developing and applying techniques to study electron dynamics on attosecond time scales as well as nuclear dynamics on femtosecond time scales.

Justin Wark, Professor of Physics University of Oxford, received his degree in physics from the University of Oxford in 1982, and his PhD in plasma physics from Imperial College in 1985. He was awarded a Royal Society University Research Fellowship at the University of Oxford, where he set up a research group working in high power laser-matter interactions. His research includes high harmonic generation, XUV lasers, X-ray spectroscopy, and the development and use of novel X-ray sources in studying shock and isentropic compression of solid-state matter via X-ray diffraction. His research group has recently been highly active in exploiting so-called 4th generation light sources – XUV and X-ray free-electron lasers, which have a spectral brightness over a billion times greater than those of any synchrotron. He has used such sources to both create and diagnose matter under extremes of density, temperature, and pressure. He is currently a fellow, tutor, and professor of physics at Trinity College, director of the Oxford Centre for High Energy Density Science, U.K. representative on the Helmholtz International Beamline for Extreme Fields at the European XFEL, fellow of the American Physical Society, and winner of the 2015 John Dawson Award for Excellence in Plasma Physics Research.

Philippe Wernet is a Professor in Photon Science at Uppsala University (Sweden). He is an experimental physicist by training, conducting fundamental research on chemical and biochemical systems. He aims at watching chemical reactions as they take place with X-ray lasers, synchrotrons, and lab sources. He uses these as high-speed X-ray cameras to select which atomic site in a molecule to look at and which orbital to follow in real time during the reaction. With his group and collaborators, he develops new X-ray based approaches to the electronic structure of small molecules, metal complexes, and metalloproteins with the aim to discover basic principles that drive their transformations and determine how they work. Wernet received his PhD in physics from Universität Hamburg (Germany) based on X-ray research at Deutsches Elektronen-Synchrotron DESY. After a postdoc at the Stanford Synchrotron Radiation Laboratory (SSRL, Stanford, USA) he became a staff scientist at Helmholtz-Zentrum Berlin (BESSY II, Berlin, Germany). Wernet has been teaching atomic and molecular physics, chemical dynamics, and optics (habilitation in physics at Technische Universität Berlin, Germany). He contributed to the science cases and instrument designs at a number of large-scale X-ray facilities. He is responsible for the interdisciplinary research program on chemical and bio-molecular physics at the Faculty of Science and Technology and for the strategic direction of ultrafast processes at the Department of Physics and Astronomy, both at Uppsala University. Wernet is serving in a number of scientific advisory committees at large-scale X-ray facilities.