LCLS STRATEGIC FACILITY DEVELOPMENT PLAN

November 2020

LCLS | Linac Coherent Light Source

U.S. DEPARTMENT OF ENERGY | Office of Science

Stanford University

SLAC NATIONAL ACCELERATOR LABORATORY

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1 INTRODUCTION & PURPOSE OF THIS DOCUMENT

A new scientific frontier opened in 2009 when the world's first X-ray free-electron laser (XFEL), the LCLS facility, began operations at SLAC National Accelerator Laboratory, operated by Stanford University on behalf of the US Department of Energy, Office of Basic Energy Sciences (DOE, BES).

The scientific start of LCLS has arguably been one of the most vigorous and successful of any new research facility, impacting a broad cross-section of fields ranging from atomic and molecular science, ultrafast chemistry and catalysis, fluid dynamics, clean energy systems, structural biology, high energy-density science, photon science, and advanced materials

The major scientific accomplishments of LCLS within the first few years of operation are reflected in both the number of publications (over 1600 to date) and the number of users attracted by this novel source (over 3000 unique users and 13,000 user visits, despite the limitations of only a single beamline). There have been 677 full-scale user experiments using the X-ray beam (2009-2020), plus 285 in-house experiments and 30 blocks of protein crystal-screening. As such, the scientific productivity of the facility is incredibly high. The scientific output and impact of LCLS during the first five years are summarized in *Reviews of Modern Physics*[1], and a summary of the scientific highlights of the first 10 years were celebrated earlier this year. An appendix to this document provides a short overview.

The scientific impact of LCLS has been enhanced by a suite of remarkable developments in the facility's capabilities. LCLS can now provide ultrashort pulses (from ~200 attoseconds (as) to >100 femtoseconds (fs)), with unprecedented peak brightness[2], in SASE or seeded-mode operation, over an energy range from ~250 to ~25,000 eV, at 120 Hz. LCLS can provide dual-pulses with relatively arbitrary separation in time (from fs to ~0.5us), with the option of dual color, and variable linear/circular polarization. Recent work has increased the peak power to over 300 GW[3] and has delivered 4 independent pulses in a train that can be separated in increments of 0.35ns to cover a time window of up to ~500ns.

Details of the suite of 7 instruments from the LCLS-I era have been published[4], with updates provided online to cover recent additions for the LCLS-II era. The scientific output can be viewed and searched on the LCLS publications site.

In 2019, an Ultrafast Electron Diffraction (UED) system operating at MeV energy was incorporated into the LCLS User Facility in order to derive maximum scientific benefit from the complementary capabilities offered by this new source. Full details are provided on the MeV-UED website, and access for >50% of its operating time is now offered via open calls for user proposals.

The success of LCLS has been accompanied by the rapid development of a number of new hard-X-ray FEL facilities, including SACLA (Japan), the European XFEL (Germany), PAL-XFEL (Republic of Korea), Swiss-FEL (Switzerland), and construction of SHINE (China), as well as soft X-ray facilities such as FLASH (Germany), Fermi@Elettra (Italy) and SXFEL (China).

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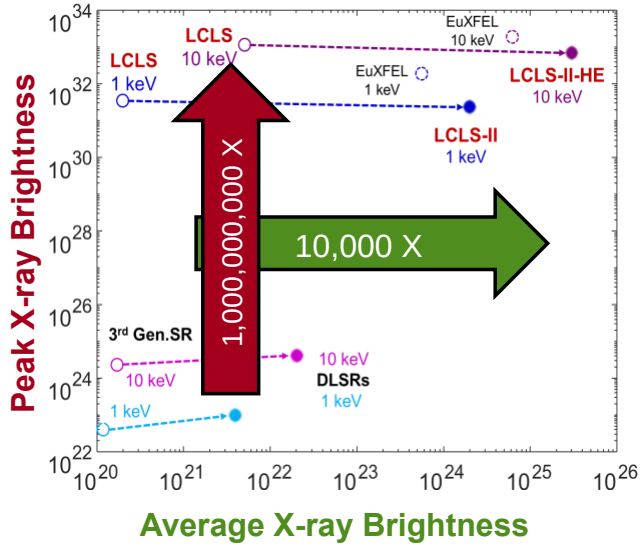
[1] Bostedt, et al., "Linac Coherent Light Source: The first five years", Rev. Mod. Phys, 88, 015007 (2016).
[2] Typically 10^{32} ph/s/mm^2/mrad^2/0.1% BW
[3] See details of operating modes: https://lcls.slac.stanford.edu/machine-faq
[4] See a special edition of the Journal of Synchrotron Radiation (2015) 22, 471 and a publication on MFX

Looking to the future, SLAC and the US Department of Energy Office of Science are pursuing a vigorous and well-coordinated series of developments to keep the LCLS facility in a preeminent state. This is captured as a four-phase plan for the next few years in Figure 1. The scientific priorities that motivate these developments have been derived from the set of Grand Challenges and Transformational Opportunities identified by the DOE-BES leadership and its advisory committees, with specific directions informed by Basic Research Needs reports, focused roundtables, and dedicated LCLS community workshops.

Figure 1 High level development plan for the LCLS facility over the near/medium term



Phase 1: 2020
- **2 LCLS-II variable gap undulators**
- 0.25 to 25 keV at 120 Hz
- XLEAP pulse(s) at 200-400 attoseconds
- 4 pulses at 0.35 ns to >500 ns separation

Phase 2: 2022
- **LCLS-II 4 GeV CW SCRF accelerator**
- 0.25 to 5 keV at 1 MHz
- >5 new endstations

Phase 3: 2027
- **LCLS-II-HE 8 GeV CW SCRF accelerator**
- 0.25 to >15 keV at 1 MHz
- 5 new or upgraded endstations

Phase 4: 2027
- **MEC Upgrade**
- 1 PW at 10 Hz, plus 1 kJ long-pulse
- Dedicated experimental cavern

The LCLS-II Project represents over a billion dollars investment to provide a new, superconducting accelerator in the first kilometer of the SLAC linac tunnel, able to deliver X-rays from 0.2 to 5 keV at up to 1 million pulses per second (compared to the current operation at 120 pulses per second). This leap in repetition rate, and the corresponding average brightness, will transform the scientific breadth and impact of the facility. A suite of new instruments, known as L2S-I, is being deployed to take full advantage of this new source.

Beyond this, an extension to higher X-ray energy at high repetition rate has long been requested by the LCLS user community. In response, DOE launched the LCLS-II-HE ("High Energy") project. This will double the energy of the superconducting linac to 8 GeV, which will extend X-ray energy from a current cutoff of ~5 keV to up to 20 keV. The LCLS-II-HE project achieved Critical Decision 1 (approval of the selected approach) in September 2018 and CD-3a (long-lead procurement) in May 2020. Ongoing work by SLAC is centered on refining the desired experimental plans for LCLS-II-HE via a series of workshops[5], which in turn are informing the instrument and source requirements and their detailed design.

LCLS-II and -HE will be transformative tools for energy science. They will qualitatively change how X-ray scattering, spectroscopy, and imaging are used in ways that were previously impossible, to show how natural and artificial systems function, revealing dynamics on timescales down to the attosecond regime, and mapping spatial response down

[5] See, for example: https://portal.slac.stanford.edu/sites/conf_public/lclsiihe2018/Pages/default.aspx and https://events.bizzabo.com/SLAC-UsersMeeting-2020/agenda/session/332876

to sub-atomic levels. LCLS-II and -HE will enable powerful new methods to capture rare chemical and material events, characterize fluctuating heterogeneous complexes, and reveal underlying quantum phenomena in matter using nonlinear, multi-dimensional, and coherent X-ray techniques only possible with a true X-ray laser.

In parallel, a major upgrade to the **MEC instrument** is being developed with the DOE Office of **Fusion Energy Sciences** (FES). This will significantly increase the peak power (from ~0.1 to >1 Petawatt) and peak energy (from ~60J to >1kJ) of the optical drive lasers, to open up a new era of precision plasma science and research into extreme material dynamics. For this, a new experimental cavern (beyond the Far Experimental Hall) is being designed. Critical Decision 0 (mission need) was approved in January 2019, and CD-1 is planned for the summer of 2021. This project is described further in section 3.4.

The layout of these upgrades is shown in Figure 2 to Figure 4. More broadly, a widespread series of developments are underway to integrate theory, simulation and experiments; pursue R&D in critical technologies; incorporate new approaches to data analysis both onsite and offsite; ensure alignment between the local SLAC science programs and the evolving LCLS facility; and engage the wider user community to exploit the new facility capabilities.

Figure 2 Layout of the accelerator upgrades for LCLS-II and LCLS-II-HE in relation to the existing linac (LCLS-I). The main SLAC site and Stanford campus can be seen in the upper right.

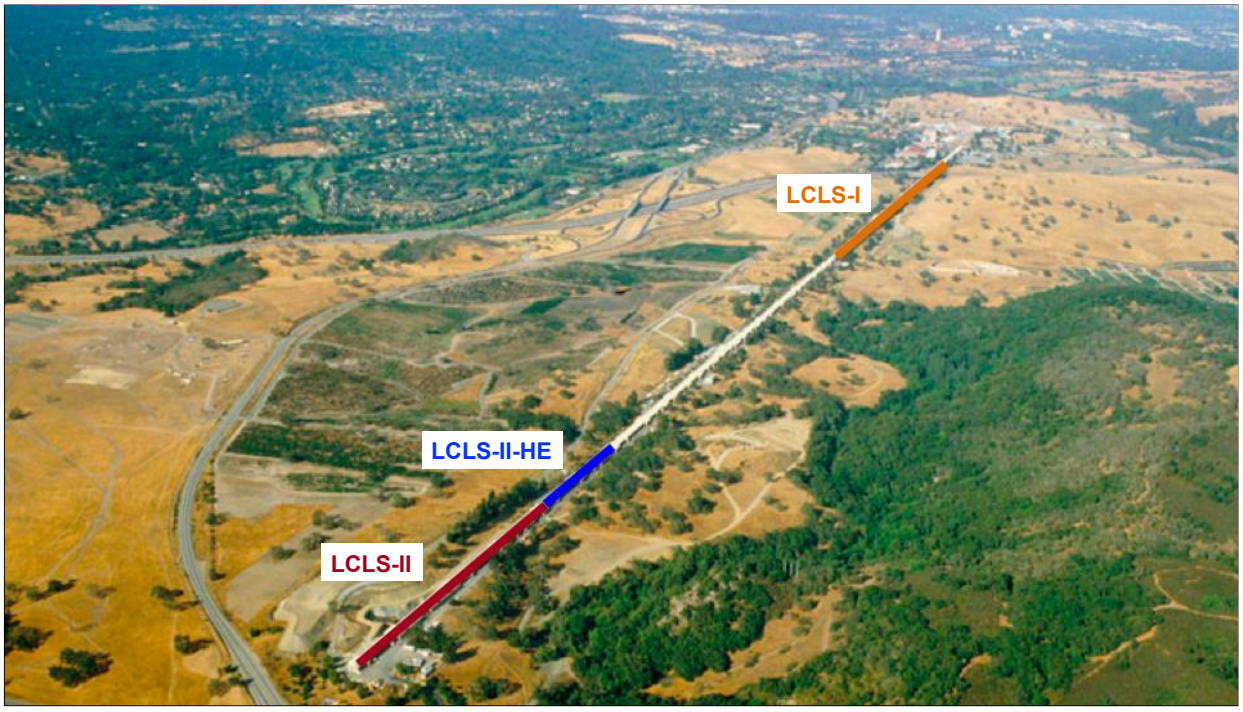
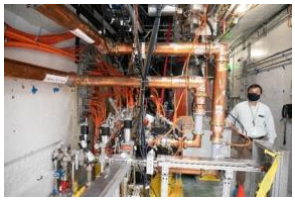
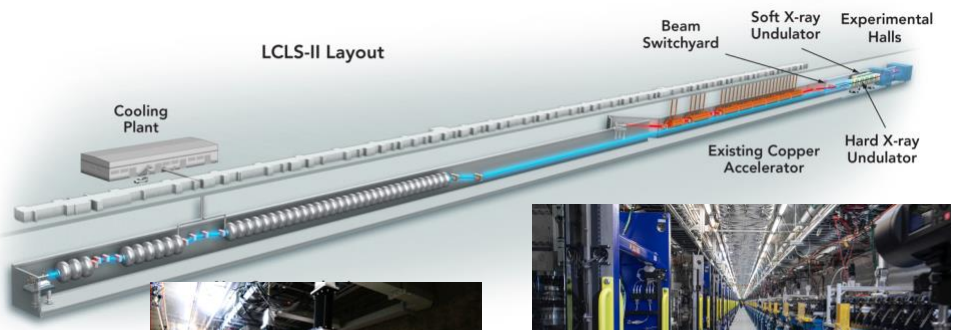


Figure 3 Schematic layouts of the accelerator upgrades

Cryoplant



Injector



Cryomodules

Undulator Hall

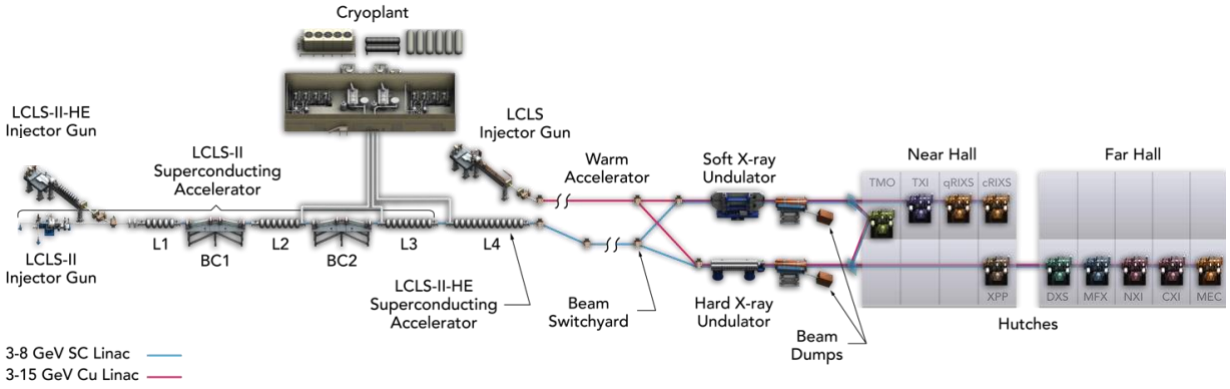
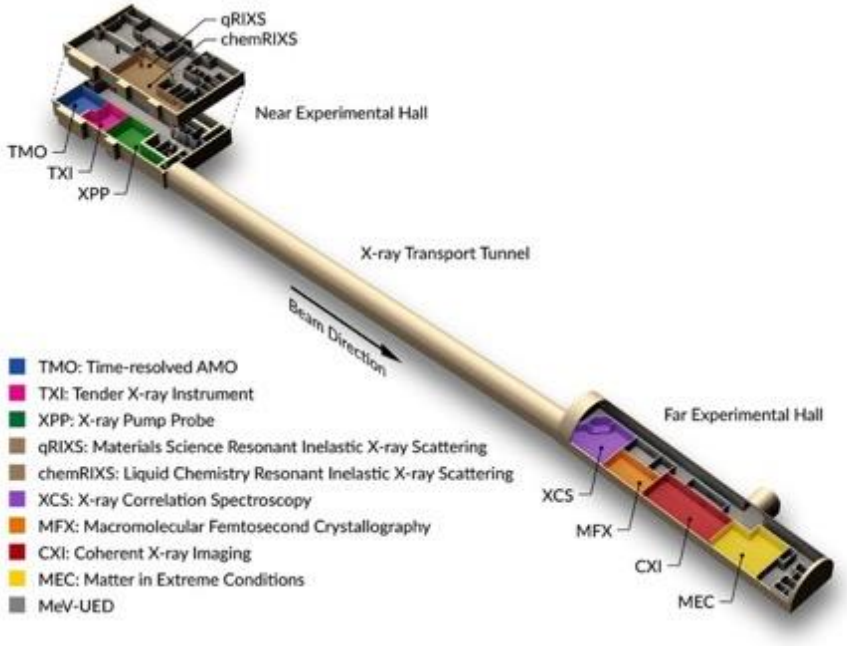
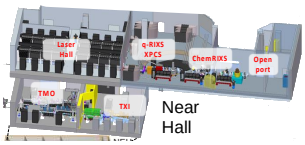


Figure 4 LCLS instrument areas

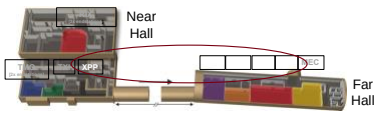


Soft X-ray (LCLS-II)



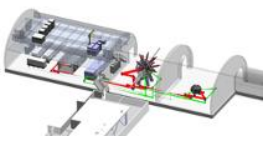
- Atomic physics
- Photo-catalysis
- Surface chemistry
- Quantum materials
- Single Particle Imaging
- Nonlinear X-ray science

Hard X-ray (LCLS-II-HE)



- Complex materials
- Semiconductors
- Biological function & dynamics
- Chemical catalysis
- Gas phase chemistry
- Quantum materials

MEC Upgrade



- Plasma physics
- High field science
- Extreme materials
- Lab astrophysics

The **purpose of this document** is to provide an overview of how the science opportunities defined by the LCLS user community are being translated into facility development activities. This document represents an update to the earlier drafts of this plan published in prior years. **Community feedback is sought**.

The priorities for LCLS development take account of the following **Guiding Principles**:

1. Ensure early LCLS-II experiments are able to adequately exploit the new source characteristics (e.g. via availability of suitable instruments, detectors, lasers, optics, data systems, and robust operation of the accelerator/FEL).
2. Use ongoing LCLS experiments to refine our priorities and assessments of delivery risk. Craft a set of credible pathways to the "ultimate experiments", using a balance of technical studies, high impact intermediary science, and offline developments.
3. Prioritize the exploitation of the unique characteristics of LCLS-II (and subsequently LCLS-II-HE and MEC-U) compared to other XFEL and storage ring facilities (e.g. the use of continuous pulse trains; the use of independently tunable XFEL beams; the ability to probe in the ultrafast domain; and the use of X-ray beams of unprecedented average power and spectral brightness).
4. Broaden the reach of LCLS to new user communities and science areas.
5. Increase community throughput (via new multiplexing schemes; dedicated end-stations; revised operational models, Start-2-End models, offline test-stands, etc)
6. Develop systems that are robust and configured to run with minimal intervention in 'standard modes' in order to enable greater control of experiments by the users, facilitate remote user oversight where possible, and to provide scalable, sustainable, and scientifically creative roles for LCLS staff.
7. Ensure sufficient investment in long-term R&D to explore next-generation concepts.

The top-level facility response to the science opportunities is outlined in the upcoming sections as follows:

- **Section 2** provides a brief recap of key science drivers;
- **Section 3** describes the facility development plans;
- **Section 4** lists Photon Science development priorities;
- **Section 5** lists Accelerator / FEL development priorities;
- **Section 6** discusses next steps;
- **Appendix** provides a brief overview of selected areas of scientific impact to date.

For feedback on the content of this document, please contact the LCLS Director.

2 SCIENTIFIC DRIVERS

2.1 Representative scientific highlights to date

This selection of results from LCLS provides an indication of the types of science that have been addressed to date (with further details and references listed in the appendix), and hopefully will inspire new ideas and directions:

Imaging molecular Motion. The characteristic LCLS ability to directly image the motions of atoms comprising a molecular reaction on fundamental time scales represents a qualitative advance in our ability to link molecular structure with function and reactivity. Hallmark LCLS studies have focused on the evolving structure and bonding of ring-opening reactions, a fundamental building block of organic chemistry. Comparison with theoretical models revealed dominant reaction trajectories and tested long-held assumptions about the electronic structure changes mediating such reactions. Pioneering LCLS experiments pave the way for a wide range of X-ray studies examining gas phase chemistry and structural dynamics associated with ultrafast chemical reactions.

Coupled dynamics of energy flow. A grand challenge in chemistry is to understand how electronic and nuclear configurations couple in molecules and thus mediate reactivity. This knowledge gap profoundly limits our ability to predict charge separation dynamics, impacting the development of robust, cost-effective systems to convert and store solar energy. LCLS has demonstrated the ability to generate and use X-ray pulses that can track the motion of excited-state charge and spin dynamics with element-specific precision, revealing the coupling to subtle coherent motion of the atomic structure, as well as how to trigger specific molecular responses with targeted light pulses.

Understanding natural multi-electron photo-catalysts. Sunlight-driven oxidation of water by photosystem II (PS-II) has generated most of the dioxygen in the atmosphere, central to life on earth. Detailed understanding of this four-photon four-electron process remains a grand science challenge, and an inspiration for inventing artificial photo-catalysts to convert sunlight to fuels. LCLS studies have provided the first damage-free structure determination of all three intermediate PS-II catalysis cycle intermediate states at room temperature, at unprecedented <2Å resolution. These results reveal important new structural details about substrate water binding and the water oxidation mechanism, resolving key discrepancies and driving the refinement of catalysis cycle theoretical descriptions.

Transition states in heterogeneous catalysis. Significant knowledge gaps in catalysis prevent us from fully exploiting and optimizing the transformation of many chemicals on which modern society depends. Short-lived reactive species mediate catalysis but are extremely difficult to characterize. The ultrafast element-specificity of LCLS X-rays have enabled the first direct observation of a transition state in a model surface catalytic reaction: the oxidation of carbon monoxide on a metal bed. This pioneering work opens new opportunities for element-specific studies of catalysis on fundamental time scales for direct input into the theory of heterogeneous catalysis.

Coherent control of complex materials. Coherent light-matter interactions represent a powerful new approach for controlling emergent material properties and creating novel material phases. One important example is coherent control of multiferroic materials based

on manipulating magnetic order with electric fields. The underlying physics, strength, and ultimate speed of magnetoelectric coupling present significant knowledge gaps. Seminal LCLS studies in multiferroic TbMnO3 applied coherent THz excitation of specific magnon modes, while ultrafast resonant X-ray diffraction revealed the coherent spin dynamics. Coherent magnetic switching should be achievable with modest scaling of the THz field. A second important example is coherent excitation of selective lattice modes to manipulate high-case temperature (Tc) superconductivity, such as in yttrium barium copper oxide (YBCO). Comparison of X-ray scattering with density functional theory (DFT) calculations revealed structural changes analogous to directional pressure associated with remarkable enhancement of superconductivity above Tc.

Correlated phenomena in quantum materials. A hallmark of quantum materials is the dominant influence of quantum-level coupling between charge, spin, orbital, and lattice modes in determining the bulk material properties. LCLS has demonstrated its ability to yield extreme precision measurements using such methods as time-stamping at the femtosecond timescale. A prime example is an LCLS study that provided the first direct measure of electron-phonon coupling strength in the superconductor FeSe purely from experiment. This provided a critical test of theoretical predictions, with results nearly 10-fold larger than the classical model. These results prompted a reassessment of the significance of such coupling in superconductivity in these compounds, and provides a powerful new means of assessing the emergent properties of a broad range of quantum materials.

Extreme material and plasma science. LCLS has demonstrated its ability to create and precisely measure new metastable material phases in extreme environments relevant for high energy density science, planetary science, and fusion research. Examples include the verification of "diamond rain," the calibration of white-dwarf based cosmic clocks, and the laboratory demonstration of Fermi acceleration.

Bioscience. LCLS delivered the first high resolution, room temperature and damage-free atomic structures of biological membrane proteins, used in the targeting of roughly 40 percent of all drugs, along with impact on diseases ranging from Zika to dengue fever, tularemia, African sleeping sickness, hypertension, influenza, and now COVID-19. LCLS has also pioneered the study of biological function by capturing the structural dynamics of biomolecules in action, on timescales ranging from picoseconds to seconds, with key results in systems such as retinal, b-lactamase and RNA riboswitches, resolving long-standing questions for how changes in enzyme structure and dynamics facilitate passage along the reaction coordinate.

2.2 Scientific Drivers for LCLS-II

LCLS-II will be a transformative tool for energy science, qualitatively changing the way that X-ray imaging, scattering and spectroscopy can be used to study how natural and artificial systems function. It will enable new ways to capture rare chemical events, characterize fluctuating heterogeneous complexes, and reveal quantum phenomena in matter, using nonlinear, multidimensional and coherent X-ray techniques that are possible only with X-ray lasers. This facility will provide access to the "tender X-ray" regime (2 to 5 keV) that is largely inaccessible today, and will use seeding technologies to provide fully coherent X-rays in a

uniformly spaced series of pulses with programmable repetition rate and rapidly tunable photon energies.

In the following, we briefly summarize six broad areas of science in which the unique capabilities of LCLS-II will be essential to address critical knowledge gaps at the new scientific frontiers of matter and energy. A complete description of these science opportunities can be found in the report: *New Science Opportunities Enabled by LCLS-II X-ray Lasers* (SLAC-R-1053).[6]

2.2.1 Fundamental Dynamics of Energy & Charge

Charge migration, redistribution and localization, even in simple molecules, are not well understood at the quantum level. These processes are central to complex processes such as photosynthesis, catalysis, and bond formation/dissolution that govern all chemical reactions. Indirect evidence points to the importance of quantum coherences and coupled evolution of electronic and nuclear wavefunctions in many molecular systems. However, we have not been able to directly observe these processes to date, and they are beyond the description of conventional chemistry models. High-repetition-rate soft X-rays from LCLS-II will enable new dynamic molecular reaction microscope techniques that will directly map charge distributions and reaction dynamics in the molecular frame. New nonlinear X-ray spectroscopies offer the potential to map quantum coherences in an element-specific way for the first time.

Experimental Approaches:

- Dynamic molecular reaction microscope
- Time-resolved photoemission spectroscopy
- Time-resolved Hard X-ray scattering
- New nonlinear X-ray spectroscopies: stimulated X-ray Raman, core-hole correlation spectroscopy.

2.2.2 Catalysis & Photo-catalysis

Understanding catalysis and photo-catalysis is essential for directed design of new systems for chemical transformation and solar energy conversion that are efficient, chemically selective, robust, and based on earth-abundant elements. LCLS-II will reveal the critical (and often rare) transient events in these multi-step processes, from light harvesting, to charge separation, to charge migration and subsequent accumulation at catalytically active sites. Time-resolved, high-sensitivity, element-specific spectroscopy enabled by LCLS-II will provide the first direct view of charge dynamics and chemical processes at interfaces, making it possible to pinpoint where charge carriers are lost (within a molecular complex or device) — a crucial bottleneck for efficient solar energy conversion. Such approaches will capture rare chemical events in operating catalytic systems across multiple time and length scales. The unique LCLS-II capability for simultaneous delivery of hard and soft X-ray pulses opens the possibility to follow chemical dynamics (via spectroscopy), concurrent with structural dynamics (substrate scattering) during heterogeneous catalysis.

[6] https://portal.slac.stanford.edu/sites/lcls_public/Documents/LCLS-IIScienceOpportunities_final.pdf

Experimental Approaches:

- Time-resolved X-ray absorption and emission spectroscopy
- Time-resolved resonant inelastic X-ray scattering
- Time-resolved X-ray photoelectron spectroscopy
- Simultaneous soft X-rays (spectroscopy) and hard X-rays (scattering)
- X-ray photon correlation spectroscopy
- New nonlinear X-ray spectroscopies

2.2.3 Emergent Phenomena in Quantum Materials

There is an urgent technology need to understand and ultimately control the exotic properties of new materials – ranging from superconductivity to ferro-electricity to magnetism. These properties emerge from the correlated interactions of the constituent matter components of charge, spin, and phonons, and are not well described by conventional band models that underpin present semiconductor technologies. Fully coherent X-rays from LCLS-II will enable new high-resolution spectroscopy approaches that will map the collective excitations that define these new materials in unprecedented detail. Ultrashort X-ray pulses and optical fields will facilitate new coherent light-matter approaches for manipulating charge, spin, and phonon modes to both advance our fundamental understanding and point the way to new approaches for materials control.

Experimental Approaches:

- Time-resolved and high-resolution resonant inelastic X-ray scattering
- Time-resolved X-ray dichroism
- Coherent X-ray scattering and imaging of domain dynamics
- Time- and spin-resolved hard X-ray photoemission
- X-ray photon correlation spectroscopy

2.2.4 Nanoscale Materials Dynamics, Heterogeneity & Fluctuations

The properties of functional materials are often defined by interfaces, heterogeneity, imperfections, and fluctuations of charge and/or atomic structure. Models of ideal materials often break down when trying to describe the properties that arise from these complex, non-equilibrium conditions. Ultrashort X-ray pulses from LCLS-II will provide element-specific snapshots of materials dynamics to characterize transient non-equilibrium and meta-stable phases. Programmable trains of soft X-ray pulses at high repetition rates will characterize spontaneous fluctuations and heterogeneities at the nanoscale across many decades of time, while coherent hard X-ray scattering will provide unprecedented spatial resolution of material structure, its evolution, and relationship to functionality under operating conditions.

Experimental Approaches:

- X-ray photon correlation spectroscopy
- Time-resolved X-ray scattering

2.2.5 Revealing Biological Function

Biological function is profoundly influenced by dynamic changes in protein conformations and by interactions with molecules and other complexes — processes that span many decades in time. Such dynamics are central to the function of biological enzymes, cellular ion

channels comprised of membrane proteins, and macromolecular machines responsible for transcription, translation and splicing, to name just a few examples. X-ray crystallography at modern synchrotrons has transformed the field of structural biology by routinely resolving simple macromolecules at the atomic scale. LCLS has already demonstrated a major advance in this area by resolving the structures of macromolecules that were previously inaccessible by using the new approaches of serial nano-crystallography and diffract-before-destroy with high-peak-power X-ray pulses. The high repetition rate of LCLS-II portents another major advance by revealing biological function through its unique capability to follow the dynamics of macromolecules and interacting complexes in real time and in native environments. Advanced solution scattering and coherent imaging techniques will characterize, at the sub-nanometer scale, the conformational dynamics of heterogeneous ensembles of macromolecules – both spontaneous fluctuations of isolated complexes and conformational changes that may be initiated by the presence of specific molecules, environmental changes, or by other stimuli. The unique LCLS-II capability for generating two-color hard X-ray pulses will enable entirely new phasing schemes for nano-crystallography, and will resolve atomic-scale structural dynamics of biochemical processes that are often the first step leading to larger-scale protein motions.

Experimental Approaches:

- Time-resolved X-ray scattering
- Time-resolved resonant inelastic X-ray scattering/spectroscopy

2.3 Scientific Drivers for LCLS-II-HE ("High Energy")

LCLS-II-HE will provide the U.S. with a true "discovery science" facility that greatly extends our ability to address the scientific challenges listed above. In particular, LCLS-II-HE will enable precision measurements of structural dynamics on atomic spatial scales and fundamental timescales. Such measurements are needed to underpin many of the transformative opportunities identified by BESAC, by providing detailed insight into the behavior of complex matter in real-world heterogeneous samples on fundamental scales of energy, time, and length.

We highlight seven broad classes of science for which LCLS-II-HE will uniquely address critical knowledge gaps:

2.3.1 Coupled Dynamics of Energy and Charge in Atoms and Molecules

Flows of energy and charge in molecules are the fundamental processes that drive chemical reactions and store or release energy. They are central to energy processes ranging from combustion to natural and man-made molecular systems that convert sunlight into fuels. Understanding and controlling these processes remains a fundamental science challenge, in large part because the movement of charge is closely coupled to subtle structural changes of the molecule, and conventional chemistry models are inadequate to fully describe this. Sharper experimental tools are needed to probe these processes simultaneously at the atomic level and on natural (femtosecond) time scales. LCLS-II-HE will image dynamics at the atomic scale via hard X-ray scattering and coherent diffractive imaging (CDI) to reveal the coupled behavior of electrons and atoms with unprecedented clarity. The combination of hard X-rays with high peak power and high average power will enable new nonlinear

spectroscopies that promise important new insights into reactive chemical flows in complex chemical environments such as combustion.

Grand-challenge science areas addressed:

- Control Matter at the Level of Electrons
- Emergent Properties from Complex Electronic and Atomic Correlations
- Master Energy and Information on the Nanoscale

2.3.2 Catalysis, Photocatalysis, Environmental & Coordination Chemistry

A deeper understanding of the fundamental processes in catalysis, photocatalysis, and interfacial chemistry is essential for directed design of new systems for chemical transformations, energy storage, and solar energy conversion that are efficient, chemically selective, robust, and based on Earth-abundant elements. LCLS-II-HE will reveal the critical (and often rare) transient events in these multistep processes, from light harvesting to charge separation, migration, and accumulation at catalytically active sites. Time-resolved, high-sensitivity, element-specific scattering and spectroscopy enabled by LCLS-II- HE will provide the first direct view of atomic-scale chemical dynamics at interfaces. The penetrating capability of hard X-rays will probe operating catalytic systems across multiple time and length scales. The unique LCLS-II-HE capability for simultaneous delivery of hard and soft X-ray pulses opens the possibility to follow chemical dynamics (via spectroscopy) concurrent with structural dynamics (substrate scattering) during heterogeneous catalysis. Time-resolved hard X-ray spectroscopy with high fidelity, enabled by LCLS-II-HE, will reveal the fine details of functioning biological catalysts (enzymes) and inform the design of artificial catalysts and networks with targeted functionality.

Grand-challenge science areas addressed:

- Beyond Ideal Materials and Systems
- Mastering Hierarchical Architectures in Matter Beyond Equilibrium
- Imaging Matter across Scales
- Data, Algorithms and Computing

2.3.3 Imaging Biological Function and Dynamics

LCLS-II-HE is the ideal, much-desired upgrade to LCLS-II that the structural biology community requires. The combination of high spatial and time resolution with a high repetition rate will make LCLS- II-HE a revolutionary machine for many biological science fields. At high repetition rates, serial femtosecond crystallography (SFX) will advance from successful demonstration experiments to addressing some of the most pressing challenges in structural biology for which only very limited sample volumes are available (e.g. human proteins); or only very small crystal sizes can be achieved (<1 μm); or where current structural information is significantly compromised by damage from conventional X-ray methods (e.g. redox effects in metalloproteins). In all of these cases, high throughput and near- physiological conditions of room temperature crystallography will be qualitative advances. X-ray energies spanning the Se K-edge (12.6 keV) will further enable de novo phasing via molecular replacement and anomalous scattering. Time-resolved SFX and solution SAXS will advance from present few-time snapshots of model systems at high photolysis levels to full time sequences of molecular dynamics that are most relevant for biology. Hard X-rays and high repetition rates will further enable advanced crystallography

methods that exploit diffuse scattering from imperfect crystals, as well as advanced solution scattering and single particle imaging methods to map sample heterogeneity and conformational dynamics in native environments.

Grand-challenge science areas addressed:

- Imaging Matter across Scales
- Characterize & Control Systems away from Equilibrium
- Data, Algorithms and Computing

2.3.4 Materials Heterogeneity, Fluctuations and Dynamics

Heterogeneity and fluctuations of atoms and charge-carriers – spanning the range from the atomic scale to the mesoscale – underlie the performance and energy efficiency of functional materials and hierarchical devices. Conventional models of ideal materials often break down when trying to describe the properties that arise from these complex, non-equilibrium conditions. Yet, there exists untapped potential to enhance materials performance and create new functionality if we can achieve a much deeper insight into these statistical atomic-scale dynamics. Important examples include: structural dynamics associated with ion transport in materials for energy storage devices and fuel cells; nanostructured materials for manipulating nonequilibrium thermal transport; two-dimensional materials and heterostructures with exotic properties that are strongly influenced by electron-phonon coupling, light-matter interactions, and subtle external stimuli; and perovskite photovoltaics where dynamic structural fluctuations influence power conversion efficiency. LCLS-II-HE will open an entirely new regime for time-domain coherent X-ray scattering of both statistical (e.g. XPCS) and triggered (pump-probe) dynamics with high average coherent power and penetrating capability for sensitive real-time, in situ probes of atomic-scale structure. This novel class of measurements will lead to new understanding of materials, and, ultimately, device performance, and will couple directly to both theory efforts and next-generation materials design initiatives.

Grand-challenge science areas addressed:

- Beyond Ideal Materials and Systems
- Mastering Hierarchical Architectures in Matter Beyond Equilibrium
- Imaging Matter across Scales

2.3.5 Quantum Materials and Emergent Properties

There is an urgent technological need to understand and ultimately control the exotic quantum-based properties of new materials – ranging from superconductivity to ferro-electricity to magnetism. These properties emerge from the correlated interactions of the constituent matter components of charge, spin, and phonons, and are not well described by conventional band models that underpin present semiconductor technologies. A comprehensive description of the ground-state collective modes that appear at modest energies, 1-100 meV, where modern X-ray sources and spectrometers lack the required combination of photon flux and energy resolution, is critical to understanding quantum materials. High- resolution hard X-ray scattering and spectroscopy at close to the Fourier limit will provide important new insights into the collective modes in 5d transition metal oxides – where entirely new phenomena are now being discovered, owing to the combination of strong spin-orbit coupling and strong charge correlation. The ability to apply

transient fields and forces (optical, THz, magnetic, pressure) with the time-structure of LCLS-II-HE will be a powerful approach for teasing apart intertwined ordering, and will be a step toward materials control that exploits coherent light-matter interaction. Deeper insight into the coupled electronic and atomic structure in quantum materials will be achieved via simultaneous atomic-resolution scattering and bulk-sensitive photoemission enabled by LCLS-II-HE hard X-rays and high repetition rate.

Grand-challenge science areas addressed:

- Emergent Properties from Complex Electronic and Atomic Correlations
- Harnessing Coherence in Light and Matter

2.3.6 Materials in Extreme Environments

LCLS-II-HE studies of extreme materials will be important for fusion and fission materials applications and could lead to important insights into planetary physics and geoscience. The unique combination of capabilities from LCLS-II-HE will enable high-resolution spectroscopic and structural characterization of matter in extreme states that is far beyond what is achievable today. High peak brightness combined with high repetition rates and high X-ray energies are required to: (i) penetrate dynamically heated dense targets and diamond anvil cells (DAC), (ii) achieve high signal-to-noise data above the self-emission bremsstrahlung background, (iii) probe large momentum transfers on atomic scales to reveal structure and material phases, and (iv) measure inelastic X-ray scattering with sufficient energy resolution and sensitivity to determine the physical properties of materials.

Grand-challenge science areas addressed:

- Characterize & Control Systems away from Equilibrium
- Beyond Ideal Materials and Systems

2.3.7 Nonlinear X-ray Matter Interactions

A few seminal experiments on the first generation of X-ray free-electron lasers, LCLS and SACLA, have demonstrated new fundamental nonlinear hard X-ray-matter interactions, including phase-matched sum frequency generation, second harmonic generation, and two-photon Compton scattering. While nonlinear X-ray optics is still in the discovery-based science phase, advances in our understanding of these fundamental interactions will lead to powerful new tools for atomic and molecular physics, chemistry, materials science, and biology via measurement of valence charge density at atomic resolution and on the attosecond-to-femtosecond timescale of electron motion. The combination of high repetition rate and high peak intensity pulses from LCLS-II-HE will enable high-sensitivity measurements that exploit subtle nonlinear effects. This will transform the nonlinear X-ray optics field from demonstration experiments to real measurements that utilize the nonlinear interactions of "photon-in, photon-out" to simultaneously access transient spectroscopic and structural information from real materials.

Grand-challenge science areas addressed:

- Control Matter at the Level of Electrons
- Emergent Properties from Complex Electronic and Atomic Correlations

3 MAJOR FACILITY DEVELOPMENT

3.1 LCLS-II Project

Working closely with DOE's Office of Science, the LCLS-II Project has been configured to meet a series of requirements laid out by the Basic Energy Sciences Advisory Committee (BESAC), in response to the above scientific drivers.

The LCLS-II design:

- Adds a new, 4 GeV superconducting linac in an existing SLAC tunnel, avoiding the need for excavation.
- Increases the repetition rate from 120 pulses per second to 1 million per second. It will be the world's first X-ray free-electron laser capable of supplying a uniformly-spaced train of pulses with programmable repetition rate.
- Provides a tunable source of X-rays, by replacing the existing undulator (used to generate X-ray laser pulses) with two new ones. This ability to tune the X-ray energy on demand will enable scientists to scan across a wide spectrum – opening up new experimental techniques and making efficient use of the valuable beam time.
- Provides access to an intermediate X-ray energy range (2 – 5 keV) that was largely inaccessible with LCLS, but which is likely critical for studies of new materials, chemical catalysis and biology.
- Supports the latest seeding technologies to provide fully coherent X-rays (at the spatial diffraction limit and near the temporal transform limit).
- Maintains the existing copper-based warm linac and upgrades parts of the existing research infrastructure to extends the operating range of the facility from its prior limit of ~12.8 keV X-rays to ~25 keV (at 120 Hz).
- Provides for a factor ~2 increase in experimental capacity, addressing a principal limiting factor of LCLS.

The detailed scientific potential of the upgraded LCLS facility was explored in a number of consultations with the user community, culminating in a series of user workshops[7] and a summary of the science opportunities for the next decade[8].

The LCLS-II Project is being led by the SLAC National Accelerator Laboratory in collaboration with four other DOE national laboratories: Argonne, Berkeley Lab, Fermilab, Jefferson Lab, and Cornell University (See Figure 5). The predicted performance of the resultant beam is shown in Figure 6.

[7] See: https://lcls.slac.stanford.edu/lcls-ii/meetings-and-reports

[8] See: https://lcls.slac.stanford.edu/lcls-ii/science

Figure 5 Principal partners in the LCLS-II Project

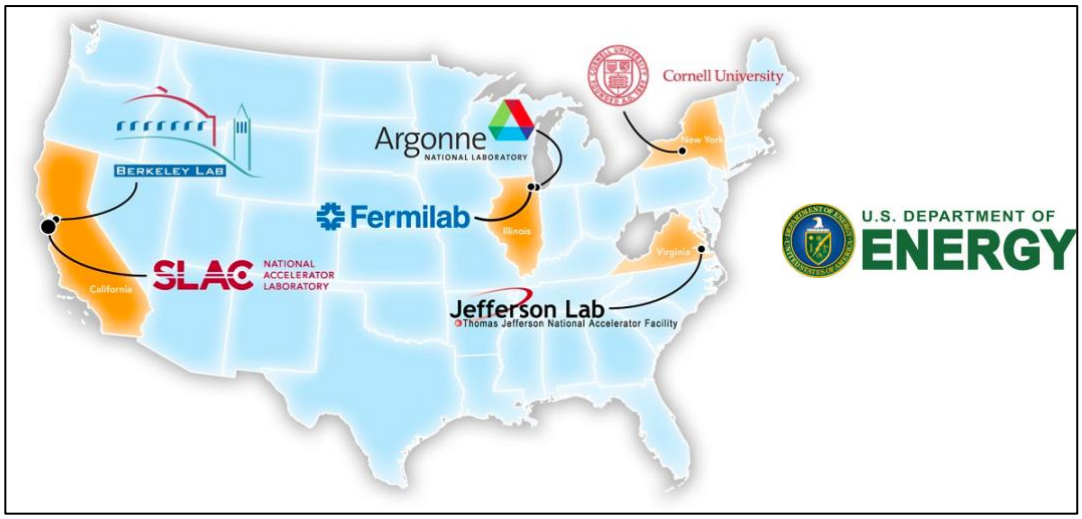
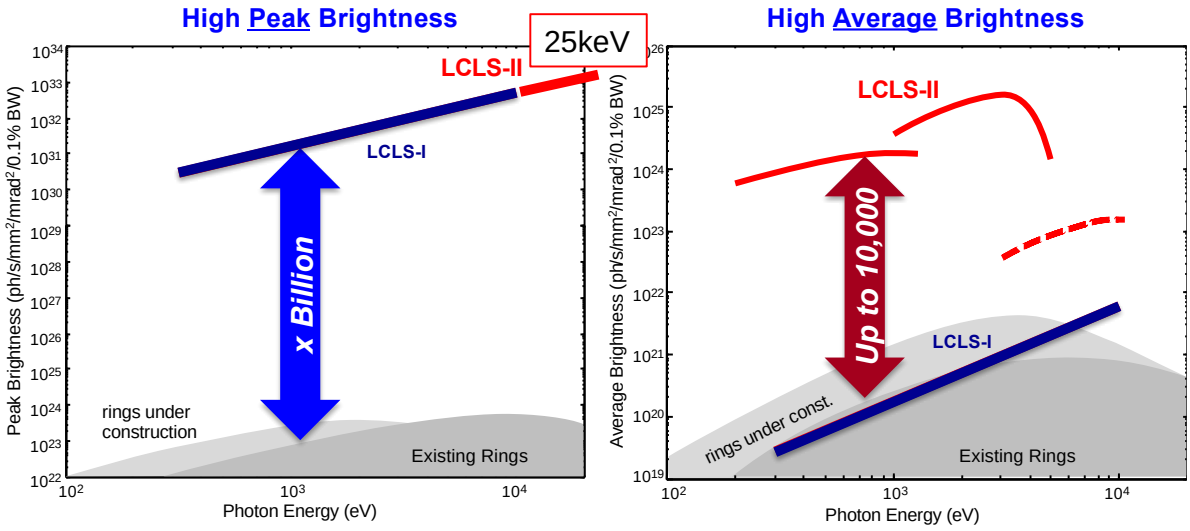
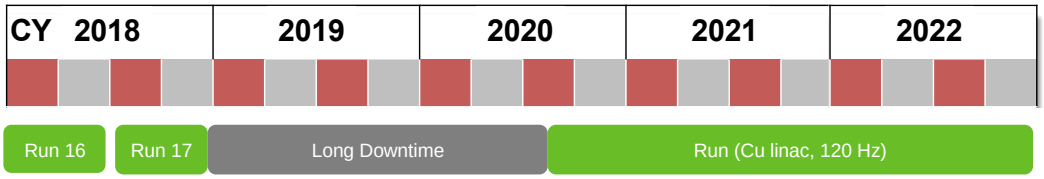


Figure 6 Performance of the LCLS-II upgrade, comparing prior performance (in blue) with the enhanced capabilities (in red; with the 3rd harmonic shown as a dotted line)



The LCLS-II project requires extended periods to modify infrastructure and install new hardware in areas where the LCLS beam is operating. Work in these areas precludes LCLS beams and so LCLS user operations must be suspended. A major downtime was scheduled from mid-December 2018 for the installation of the 2 new variable-gap undulators and a range of new instrument infrastructure. This included a "cross-over beamline" to route the copper linac to the soft X-ray undulator, such that both linacs can feed both undulators. User operation with the 120 Hz copper linac restarted in August 2020, following the installation shutdown and then a COVID-induced extension. Transition to incorporate the MHz LCLS-II beam is now planned for mid-2022 (see Figure 7). LCLS-II-HE and MEC-Upgrade are planned for later in the decade (approximately 2027).

Figure 7 LCLS operations schedule.



A phased increase in capability (for the instruments a well as for the new beams) will take place over the 2020-2023 period, as described below. Ramped commissioning is essential since the undulators can be permanently damaged in minutes if the electron losses are not carefully limited at full beam power. Therefore, the beam power allowed in each undulator will be closely controlled and ramped up slowly while undulator magnets are regularly characterized and electron beam losses are carefully monitored and minimized.

3.2 Instrument Development for LCLS-II

Many options exist for the LCLS facility to adapt its current suite of instruments to the opportunities presented by the LCLS-II upgrade. Over the long term this could include the commissioning of new experimental halls, given the ability of the superconducting accelerator to feed up to 8 or 10 undulators. This would greatly relieve the capacity limitations of the current facility, and allow optimized beamlines for each spectral region and/or experimental technique.

In the near term, it is obviously important to ensure that best use is made of the upgraded capabilities, to take full advantage of the high repetition-rate and extended spectral coverage, and to explore the emergent techniques for this new class of X-ray FEL.

A suite of "**First Experiments**[9]" for LCLS-II was developed to drive the design and implementation of the new instruments on the basis of the science opportunities described in Section 2. Feedback on these is encouraged.

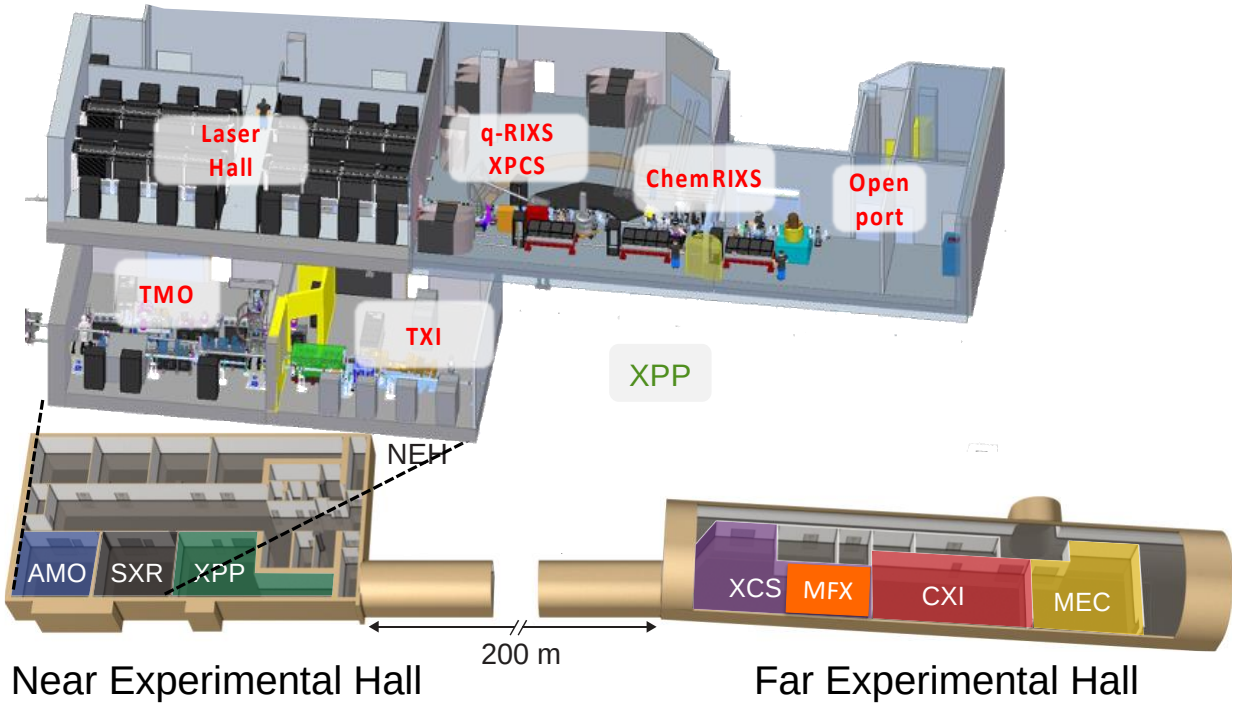
Adaptation of the hard X-ray instruments is straightforward for use of the existing Cu Linac with the new undulator. New optics are required to ensure high quality throughput, extension to 25 keV, and improved beam-splitting options. New detectors are being deployed for higher dynamic range and lower noise, with options for high-sensitivity sensors (for >18 keV) being tested.

Adaptation of the soft X-ray instruments is more involved, as is the extension to tender X-ray coverage (2-5 keV). An approach to optimize the Near Experimental Hall (NEH) has been determined following extensive consultation with the user community. An outline is presented here, and further details can be found on the LCLS website[10].

[9] See: https://lcls.slac.stanford.edu/instruments/neh-1-1/science-drivers, https://lcls.slac.stanford.edu/instruments/neh-1-2/science-drivers, and https://lcls.slac.stanford.edu/instruments/neh-2-2/science-drivers

[10] See: https://lcls.slac.stanford.edu/instruments/l2si

Figure 8 LCLS instrument layout, fed from the 2 new undulators from the LCLS-II Project, highlighting the revised layout of the Near Experimental Hall.



The modified Near Experimental Hall (NEH) and Front End Enclosure (FEE) provides:

- Initially two, and ultimately three new X-ray beamlines from the soft X-ray undulator (SXU) to serve four new instruments (TMO, qRIXS, ChemRIXS and TXI), as well as an open port for user-supplied endstations.
- A hard X-ray beamline to serve the current suite of five hard X-ray instruments (XPP, XCS, MFX, CXI, MEC), as well as to the new TXI instrument to create a 'dual beam' instrument area.
- New hutches (NEH 1.1, 1.2, and 2.2) to accommodate the instruments, and a major new "central laser laboratory" to serve the NEH.
- Dedicated control rooms for each instrument.
- An expanded data/server capability.

An overview of the new instrument layout is provided in Figure 8, and schematic models of the new instruments are displayed in Figures 9, 10 and 11.

NEH-1.1 (TMO) will specialize in atomic and molecular studies in the gas phase, and is configured to utilize both the high per pulse energy from the copper accelerator (120 Hz) as well as high average intensity and high repetition rate (1 MHz) from the superconducting accelerator. NEH 1.1 will support two experimental endstations positioned in tandem. The

first endstation (LAMP/VMI) will employ velocity map and angular resolved spectrometers to measure femtosecond and sub-femtosecond excited state dynamics of atoms and molecules. The second endstation, known as DREAM, will perform coincidence charged particle spectroscopy that yields kinematically complete measurements at each time step of an evolving reaction. This experimental approach, known as a "molecular reaction microscope" will enable the complete spatial reconstruction of the excited-state charge transfer and subsequent dissociation at each time step for a fixed-in-space molecular orientation. This is a powerful new approach for visualizing a broad range of excited-state molecular dynamics.

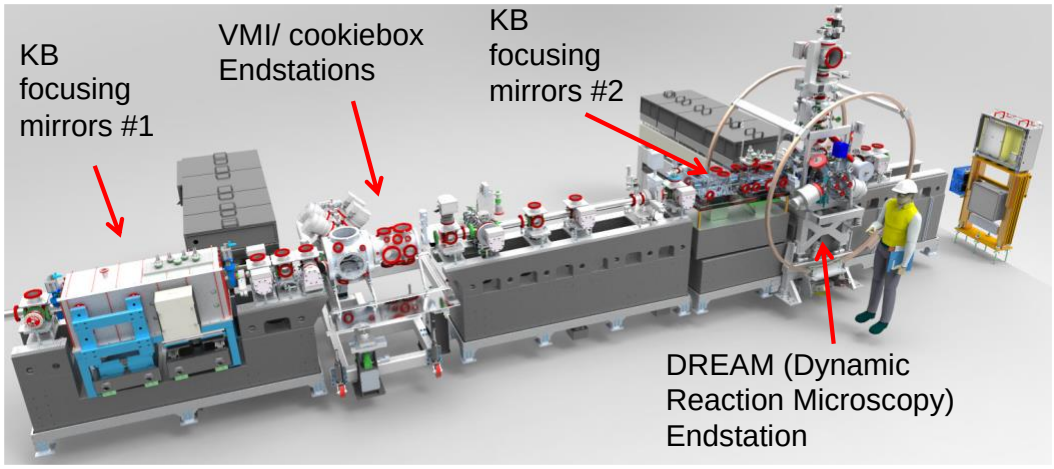
NEH-1.2 (TXI) is a dual-beam instrument, fed by both the SXU and HXU undulators; a feature currently unique among XFEL instruments. The tender x-ray instrument will enable x-ray pump/x-ray probe techniques especially in the emerging field of nonlinear x-ray science, support tender X-ray spectroscopy measurements, and provide a coherent scattering/ forward diffraction instrument for single particle imaging of sub-micron samples. It is designed to accommodate a variety of additional techniques, such as absorption and photoemission spectroscopy, as well as an array of samples from fixed targets to gases, aerosols and liquid jet targets.

NEH-2.2 (RIXS) uses a variable resolution grating monochromator to direct soft X-rays (0.25-1.2 keV) to a series of experimental areas on the upper floor, designed for moderate resolution (~5,000) and high resolution (~30,000) photon spectroscopy. It will host two instruments in tandem, plus an open port for future expansion or user-supplied equipment.

The first instrument in the 2.2 hutch (**qRIXS**) is designed for high resolution, momentum resolved resonant inelastic x-ray scattering (RIXS) to study bosonic excitations in solid state samples. The qRIXS instrument is comprised of a sample chamber and a rotatable spectrometer consisting of grating and detector assemblies (and auxiliary components), continuously covering the range of scattering angles 40-160 degrees in the horizontal plane. The spectrometer is designed to achieve a target resolving power of over 30,000 when integrated with the monochromatic beamline performance, with the option for lower resolving power, ~10,000, and higher throughput through the use of a second grating in the spectrometer. The spectrometer arm is also designed to accommodate X-ray Photon Correlation Spectroscopy (XPCS) and REXS experiments.

The second instrument in the 2.2 hutch (**ChemRIXS**) is designed for soft x-ray spectroscopy of liquid samples, with two x-ray emission spectrometers; a conventional varied line spacing spectrometer and the potential addition of a transition edge sensor (TES). This instrument also provides an open port for user-supplied endstations. Currently under consideration are a momentum microscope (for time-resolved ARPES investigation of quantum materials) and a surface science endstation for ultrsafast chemistry and catalysis research.

Figure 9 Schematic model and photo of NEH1.1 (TMO)



≤ 1 ∝m, 80% T

≤ 0.3 ∝m, 60%T

Endstation	Science	Method	XFEL
IP1 (VMI / cookiebox)	High field physics Photophysics & photochemistry	Ion/Electron Spectroscopy	250 – 2000 keV
IP2 (DREAM)	Fundamental excited state dynamics Charge and energy transfer	Dynamic molecular reaction microscope	250 – 1500 eV >= 100kHz

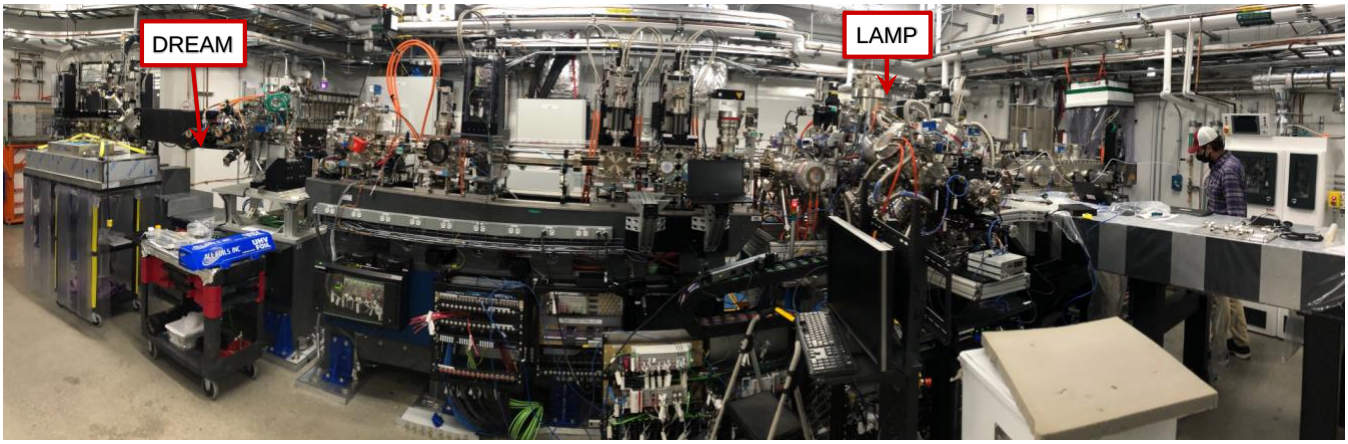
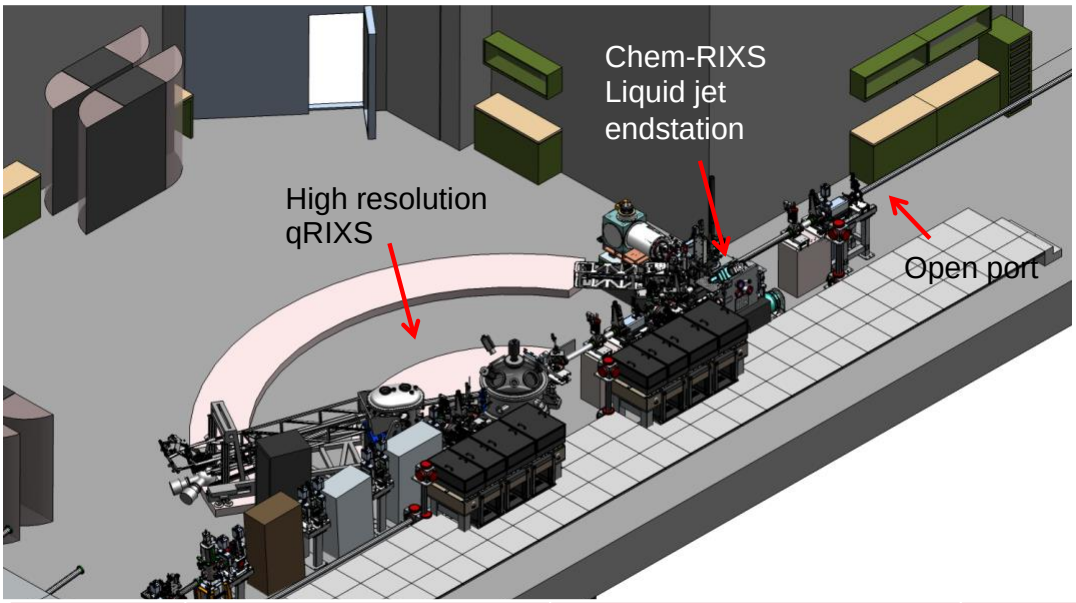


Figure 10 Schematic model and photo of NEH2.2 (RIXS)



Endstation	Science	Methods	XFEL
qRIXS	Emergent phenomena and collective modes in correlated materials	Resonant inelastic X-ray scattering, Resonant diffraction	250 – 1600 eV, >10^{14} ph/s with >30,000 resolving power
ChemRIXS	Heterogeneous catalysis Interfacial chemistry Photo-catalysis	X-ray Absorption & Emission Spectroscopy	250-1600 eV, ≥ 100 kHz, 1000-5000 res. power
qRIXS	Nanoscale material dynamics	XPCS	250 – 1600 eV

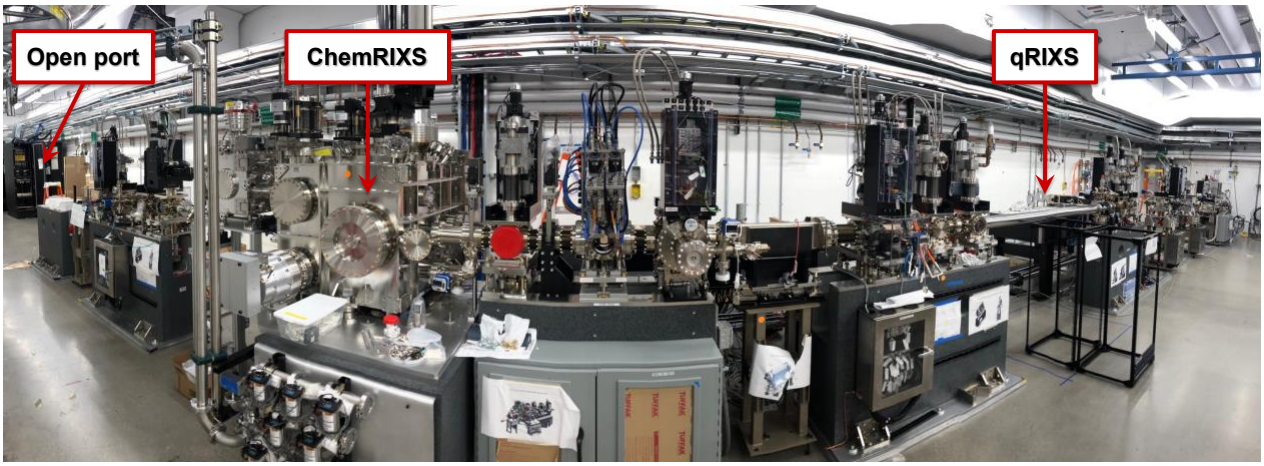
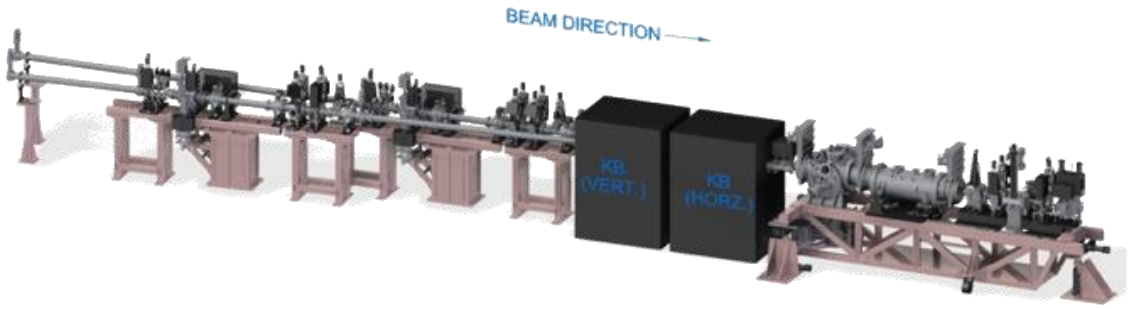


Figure 11 Schematic model of dual-beam NEH1.2 (TXI)



Science	Method	XFEL Requirement
Fundamental dynamics of energy & charge - excited state	X-ray Pump / X-ray Probe	~1 µm focus, 400-5000 eV
Bio-imaging (Revealing biological function)	CDI/SPI	~1 µm focus, 1500-5000 eV
Protein dynamics and SAD phasing (Revealing biological structure)	Protein Crystallography	1-5 µm focus, 3000 to 5000 keV
Revealing biological function and time scales	TR-SAXS/fSAXS	~10 µm focus, 1500-5000 keV
Catalysis Photo-catalysis Bio-spectroscopy	Absorption & emission spectroscopy	~10 µm focus, 250-1500 eV, ≥100 kHz

Figure 12 Nominal timeline for the new instrument suite

FELSource	Inst.	Commissioning
CuRF	XPP, XCS, MFX, CXI, MEC	8/2020
CuRF	TMO (LAMP/cVMI)	10/2020
CuRF	RIX (ChemRIXS)	3/2021
CuRF	RIX (qRIXS – no spect.)	7/2021
CuRF	RIX (qRIXS)	1/2022
CuRF	TMO (MRCOFFEE)	2/2022
CuRF	TMO (DREAM)	3/2022
SCRF	RIX (qRIXS)	8/2022*
SCRF	TMO (DREAM)	8/2022*
SCRF	TMO (MRCOFFEE)	8/2022*
SCRF	RIX (ChemRIXS2.0)	12/2022
SCRF	TXI	1/2023

***Tied to LCLS-II early finish**

The nominal phased delivery schedule for the 4 new instrument areas (dependent on funding and resource availability) is currently as shown in Figure 12.

It is important to note that the detailed design of these instruments is rapidly evolving. Please refer to the **LCLS website** for up-to-date information. This provides details of the scientific fields being served; the experimental approaches being adopted; source characteristics; beamline and endstation designs; and detector/laser system specifications.

3.3 LCLS-II-HE Project

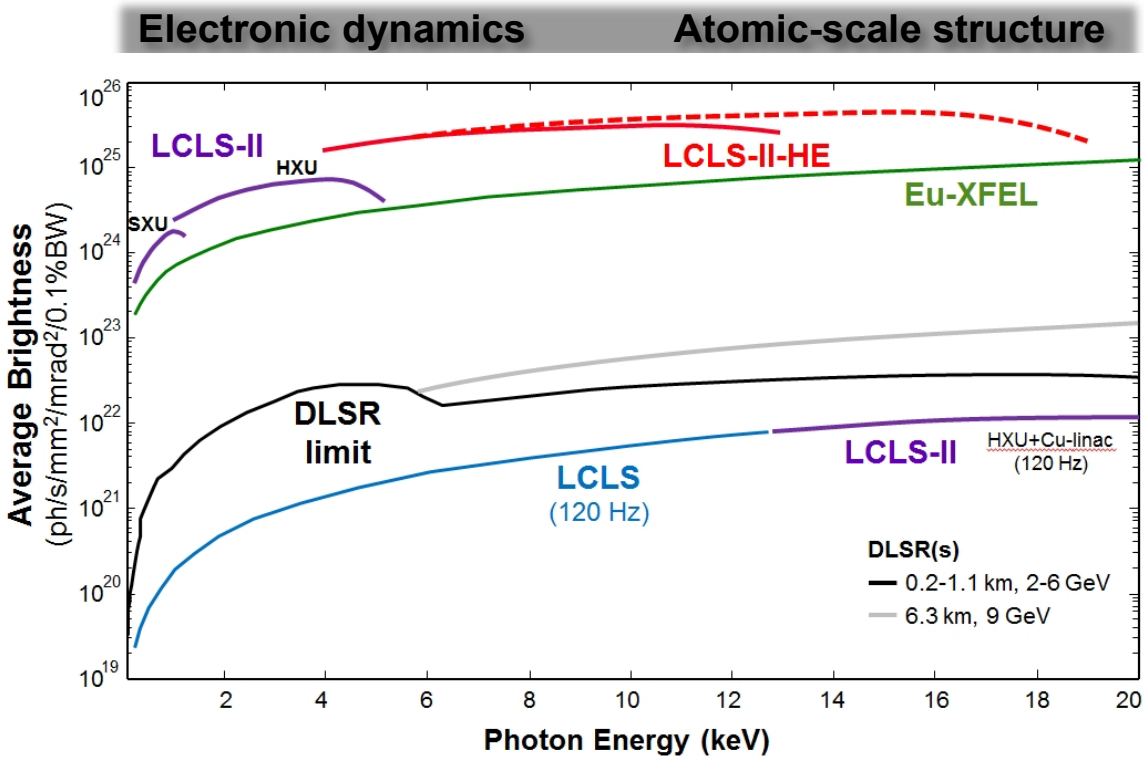
LCLS-II-HE will provide a qualitatively new capability, delivering ultrafast atomic resolution at high average power. The project will extend operation of the high-repetition-rate beam into the critically important "hard X-ray" regime (5-20 keV) that has been used in more than 75% of LCLS experiments to date, providing a major leap in performance to the broadest cross-section of the user community.

The Basic Energy Sciences Advisory Committee (BESAC) assessed this project in 2016, and concluded that *"… the LCLS-II-HE project is considered to be 'absolutely central to contribute to world leading science' and 'ready to initiate construction'".* Subsequent action by DOE led to Mission Need (Critical Decision 0) being approved in December 2016, CD-1 (approval of the selected approach) in Sept 2018, and CD-3a (long-lead procurements) in May 2020.

The energy reach of LCLS-II-HE (stretching from 5 keV up to 20 keV) will enable the study of atomic-scale dynamics with the penetrating power and pulse structure needed for *in situ* and *operando* studies of real-world materials, functioning assemblies, and biological systems.

The performance of LCLS-II-HE in comparison to other X-ray sources is shown in Figure 13.

Figure 13 The performance of LCLS-II-HE will allow access to the 'hard X-ray' regime, providing atomic resolution capability, with an average brightness roughly 300 times the ultimate capability of a diffraction-limited storage ring (DLSR). The predicted performance of LCLS-II-HE is shown in red, with the solid line based on a beam whose emittance is the same as at present; and the dotted line for expected improvements.



The facility will:

- Deliver **two to three orders of magnitude increase in average spectral brightness beyond** any proposed or envisioned diffraction-limited storage ring (DLSR), exceeding the design performance of the European-XFEL.
- Provide **temporal coherence** for high-resolution spectroscopy near the Fourier transform limit with more than **300-fold increase in average spectral flux** (ph/s/meV) for high-resolution studies beyond any DLSR**.**
- Generate ultrafast hard X-ray pulses in a **uniform (or programmable) time structure** at a repetition rate of up to 1 MHz – a qualitative advance beyond the burst-mode nature of the European-XFEL, and a **100,000-fold improvement in temporal resolution** compared to storage ring sources.
- Enable **both accelerators to use both undulators**, representing an unprecedented level of flexibility for the user community**.**

To achieve this, the LCLS-II-HE project will add at least 20 cryomodules with an enhanced performance, doubling the electron beam energy from the superconducting accelerator to 8 GeV and making use of the existing cryogenic cooling capacity and space within the linac tunnel. It will upgrade the soft X-ray undulator to enable operation at 8 GeV alongside the hard X-ray undulator, and will add a new superconducting gun to provide low emittance for the highest energy reach. It will upgrade 5 of the X-ray instruments to take full advantage of the new source from the earliest opportunity (see below).

The facility will take advantage of the myriad beam-sculpting techniques developed on LCLS, including bandwidth control via seeding, multi-pulse operation, and delivery of the 3rd harmonic (opening up new areas of science in the energy range 20 to 50 keV).

LCLS-II-HE will lead to significant scientific impact, enabled by a suite of unmatched technical attributes:

I. **Access to the energy regime above 5 keV:** This is particularly important because it allows analysis of key chemical elements in addition to providing atomic resolution. For example, this regime encompasses Earth-abundant elements that will be needed for large-scale deployment of photocatalysts for electricity and fuel production; it allows study of strong spin-orbit coupling that underpins many aspects of quantum materials; and it reaches the biologically important selenium K-edge, used for protein crystallography.

II. **High repetition rate, ultrafast hard X-rays** from LCLS-II-HE will reveal coupled atomic and electronic dynamics in unprecedented detail. Advanced X-ray techniques will simultaneously measure electronic structure and subtle nuclear displacements at the atomic scale, on fundamental timescales (femtosecond and longer), and in operating environments that require the penetrating capabilities of hard X-rays and the sensitivity provided by high repetition rate.

III. **Temporal resolution:** LCLS-II-HE will deliver coherent X-rays on the fastest timescales, opening up experimental opportunities that were previously unattainable due to low signal-to- noise from LCLS (at 120 Hz) and that are simply

not possible on non-laser sources. The typical limit for synchrotron sources is ~100 ps (100,000 fs), whereas the performance of LCLS has progressed from initial pulse durations of 300 fs down to 5 fs, coupled to the capability for double pulses with independent control of energy, bandwidth, and timing. Ongoing development programs offer the potential for 0.5 fs pulses.

IV. **Temporal coherence**: Control over the XFEL bandwidth will be a major advance for high- resolution inelastic X-ray scattering and spectroscopy in the hard X-ray range (RIXS and IXS). The present scientific impact of RIXS and IXS is substantially limited by the available spectral flux (ph/s/meV) from temporally incoherent synchrotron sources. LCLS-II-HE will provide more than a 300-fold increase in average spectral flux compared to synchrotron sources, opening new areas of science and exploiting high energy resolution and dynamics near the Fourier transform limit.

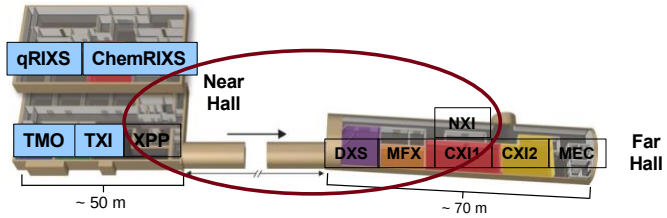
V. **Spatial coherence:** The high average coherent power of LCLS-II-HE in the hard X-ray range, with programmable pulses at high repetition rate, will enable studies of spontaneous ground-state fluctuations and heterogeneity at the atomic scale from μs (or longer) down to fundamental femtosecond timescales using powerful time-domain approaches such as X-ray photon correlation spectroscopy (XPCS). LCLS-II-HE capabilities will further provide a qualitative advance for understanding non-equilibrium dynamics and fluctuations via time-domain inelastic X-ray scattering (FT-IXS) and X-ray Fourier-transform spectroscopy approaches using Bragg crystal interferometers.

VI. **Structural dynamics and complete time sequences:** LCLS achieved early success in the determination of high-resolution structures of biological systems and nanoscale matter before the onset of damage. X-ray scattering with ultrashort pulses represents a step-change in the field of protein crystallography. An important scientific challenge is to understand function as determined by structural dynamics – at the atomic scale (requiring ~1Å resolution) and under operating conditions or in physiologically relevant environments (*e.g.* aqueous, room temperature). The potential of dynamic pump-probe structure studies has been demonstrated in model systems, but the much higher repetition rates of LCLS-II-HE are needed in order to extract complete time sequences from biologically relevant complexes. Here, small differential scattering signals that originate from dilute concentrations of active sites and low photolysis levels are essential in order to provide interpretable results.

VII. **Heterogeneous sample ensembles and rare events:** The high repetition rate and uniform time structure of LCLS-II-HE provide a transformational capability to collect 10^8–10^{10} scattering patterns (or spectra) per day with sample replacement between pulses. By exploiting revolutionary advances in data science (*e.g*. Bayesian analysis, pattern recognition, manifold maps, or machine learning algorithms) it will be possible to characterize heterogeneous ensembles of particles or identify and extract new information about rare transient events from comprehensive data sets.

The instrument development program to meet these scientific goals is under active development, with recent progress and future ideas discussed at community workshops[11]. The plans are to introduce a new inelastic X-ray scattering (DXS) instrument to augment XCS, and to upgrade the exiting instruments for high repetition-rate, including a revised distribution of instruments in the Far Experimental Hall to increase experimental capacity, as shown in Figure 14.

Figure 14 Layout and upgrade plans for the hard X-ray instruments suite with LCLS-II-HE



Instr.	Upgrade Plan	Experimental Techniques	Science Drivers
DXS	High dispersion, tunable mono IXS spectrometer High rate laser, detector, DAQ	• Very high resolution IXS • Coherent scattering (XPCS)	• Quantum information and materials (metastable phases) • Novel functional devices (sensors, computing switching) • Soft/amorphous materials heterogeneity, fluctuations, function • Drivers of material failure (fracture, electrical breakdown)
XPP	Cooled mono & BL diagnostics High rate detector High rate laser pump	• Time-resolved diffraction / scattering • Pump / probe spectroscopy • RIXS	• Versatile platform for molecular structure and energy flow • Femtosecond nanoscale probing of material dynamics • Scanning ptychography (e.g. semiconductor chip design)
CXI	Cooled optics & BL diagnostics High rate detectors, DAQ Split CXI into two endstations.	• Coherent imaging (vacuum) • Spectroscopy	• Enzyme dynamics • Catalysis: chemical reactivity linked to structural dynamics
MFX	Cooled optics, diagnostics. High rate detectors, DAQ	• Coherent imaging (atmospheric)	• Structure-based drug design
NXI	New chamber, detector, DAQ	• Nanoscale imaging and scattering	• Gas phase chemistry • High field, nonlinear X-ray science

XPP
- Monochromatic
- Scattering
- Spectroscopy
- Ambient

DXS
- Monochromatic
- Inelastic Scattering
- XPCS
- XCVS
- X-ray Split & Delay

MFX
- Pink
- ≥ 3 µm focus
- Scattering
- Spectroscopy
- Ambient

CXI
- Pink
- 1 µm focus
- Scattering
- Spectroscopy
- Vacuum

NXI
- Pink
- 0.1 µm focus
- Scattering
- Spectroscopy
- Vacuum

[11] See, for example: https://portal.slac.stanford.edu/sites/conf_public/lclsiihe2018/Pages/default.aspx and https://events.bizzabo.com/SLAC-UsersMeeting-2020/agenda/session/332876

3.4 Matter in Extreme Conditions Upgrade (MEC-U) Project

The field of High Energy Density (HED) plasma science performed at the Matter in Extreme Conditions (MEC) instrument requires high pulse energy and high peak power lasers. The current MEC instrument has two mid-scale laser systems: a temporally-shaped, nanosecond laser system that fires at one shot per few minutes with a pulse energy of 60 J / 10ns; and a Ti:S, high intensity, femtosecond laser system delivering 30-100 TW peak power at 5 Hz.

LCLS engaged in extensive community outreach through workshops and direct interactions to identify the areas of scientific opportunity and the corresponding "flagship experiments" that are desired to be performed over the coming decade and beyond. This has demonstrated a broad-based need for an upgrade that combines three sets of beams: a high-power short pulse laser (1 Petawatt at ~150J / 150fs / 10 Hz); a kilojoule-scale shaped long-pulse laser, and the LCLS X-ray laser. The driving areas of science are listed in the table below.

Category	Key Science	Experimental Configuration
Relativistic Laser-Matter Interactions	Relativistic collisionless shocks; plasma instabilities; ion acceleration mechanisms; relativistic transparency; Gbar plasmas	$>10^{21}$ W/cm^2 target irradiation
	Plasma mixing, ion stopping, atomic physics. Secondary radiation sources to enable fusion material studies (damage cascades / accumulation).	X-rays probe plasma conditions; Secondary sources enable fusion material science on secondary targets
HED Laser Plasmas	Re-shock instabilities; microphysics in turbulence; magnetic interactions; HED plasma flows and turbulence	SPL drives target directly, ion source, or magnetic coil 1 kJ laser transverse to X-rays
Compressed matter	Phase transition kinetics; void collapse; microstructure; planetary interior conditions; ICF ablators	SPL from side 1 kJ laser along X-ray direction;
	Equation of state of WDM; dense plasma atomic physics	High repetition rate 200 J @ 2ω for faint signal accumulation

High-peak-power and high energy laser facilities provide a versatile set of tools for creating plasmas under extreme conditions. When focused to a small spot, ultrashort laser pulses with Petawatt (PW) power deliver light pressure in the gigabar regime and create electric fields high enough to strongly ionize matter and drive electrons to ultra-relativistic speeds. These interactions can also be harnessed to produce bright pulses of particles that can be used to volumetrically heat matter to dense, strongly coupled plasma states, or to produce radiation damage in materials of interest. A tailored high-energy laser can drive shocks in materials to access densities much higher than solid. While creation of such conditions can be achieved today at a handful of laser facilities around the world, the high-energy-density states they generate are difficult to characterize in a quantitative manner using the tools available to them.

To address this, a laser facility with high-power, high-intensity beam parameters that is co-located with hard X-ray laser probing capabilities (i.e. with an X-ray wavelength that allows atomic resolution) will provide the required diagnostic capabilities for fusion discovery science and related fields, with representative areas listed in the table below. This co-location enables novel pump-probe experiments with the potential to dramatically improve our understanding of the ultrafast response of materials in extreme conditions, e.g., found in the environment of fusion plasmas, astrophysical objects, and highly stressed engineering materials.

Recent work at LCLS has also demonstrated exquisite ultrafast measurements of the material structural response to radiation, but higher flux sources of deuterons, neutrons and gamma rays are needed to properly emulate the environment and physics processes that occur in materials next to fusion plasmas. An upgrade to PW- and kJ-scale laser systems holds the potential to validate inter-atomic potentials in molecular dynamics simulations of materials to enable long-term predictions of the material behavior in fusion facilities.

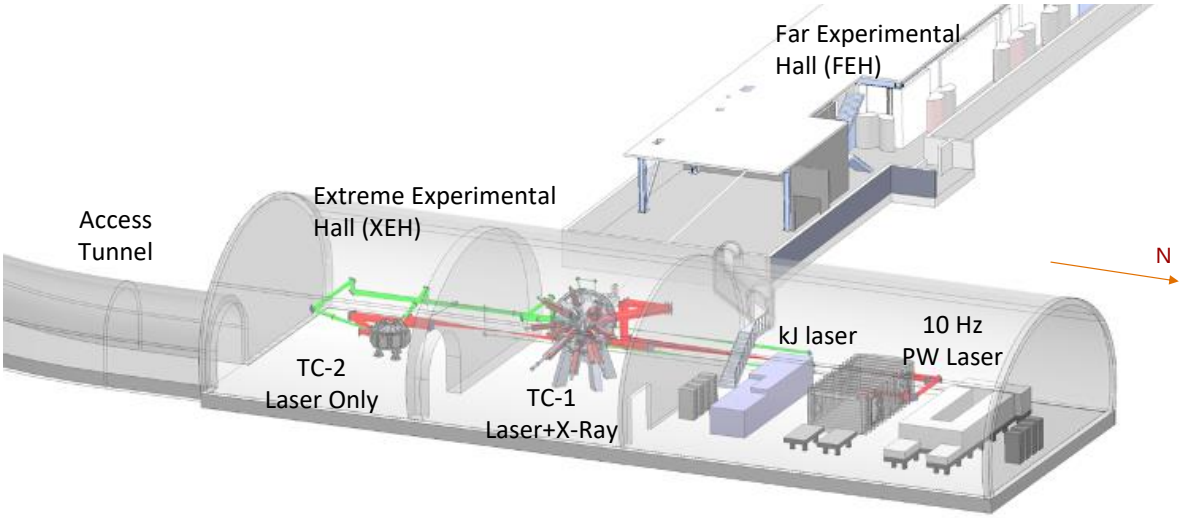
The 'mission need' (Critical Decision 0) for this type of facility was approved by DOE Office of Fusion Energy Sciences (FES) in January 2019.

LCLS is now engaged in a design process in response to this mission need. It is proposed to reconfigure and upgrade the MEC instrument at LCLS by excavating a new experimental hall, as shown in Figure 15 to provide a world-leading open-access user facility that will enable world-leading discovery science in a wide range of high-energy-density physics topics.

This facility is being designed to deliver the following key characteristics, with the point design shown schematically in Figure 15

- **Independent cavern east of the FEH**
- **Laser systems**
  - High rep-rate short pulse: 10 Hz, 150 J, 150 fs, 1 PW
  - High rep-rate long pulse: 10 Hz, 200 J @ 2ω @ 10ns; programmable pulse shape
  - High energy long pulse: > 1 kJ @ 2ω @ 10ns; programmable pulse shape
  - LCLS XFEL (5 to 45 keV)
- **X-ray + laser target chamber**
  - Beam delivery for laser systems and X-rays
  - Optimized to serve a broad plasma physics community (high throughput, rapid reconfiguration, range of illumination geometries and diagnostics)
- **Laser-only target chamber**
  - Beam delivery for laser systems
  - Optimized for flexibility, training, exploration, development

Figure 15 Schematic of the design concept for the upgraded MEC instrument, showing two target chambers in a new experimental cavern to the east of the Far Experimental Hall



4 PHOTON SYSTEM DEVELOPMENT

4.1 Scientific Technique Development

The distinguishing capabilities of the LCLS-II and -HE upgrades (high repetition rate ultrafast X-ray pulses with both longitudinal and transverse coherence, along with two new tunable undulators) will enable entirely new X-ray techniques that will be important for many areas of science, and will significantly advance X-ray techniques that have been pioneered at the LCLS facility to date.

This section outlines the development plans for 5 ambitious new X-ray techniques (focusing on the key instrumentation-related components), and maps them onto the new suite of beamlines. Where appropriate, time will be made available on the existing facility and/or during early operation of LCLS-II to explore these emerging techniques and inform their future potential:

- Fluctuation X-ray scattering – *interacting complexes in natural environments*
- Stimulated X-ray Raman spectroscopy – *ultrafast charge-transfer dynamics*
- Core-hole correlation spectroscopy – *quantum coupling of valence charge states*
- Single-particle imaging – *toward atomic resolution*
- Dynamic multi-view tomography – *chemical mapping of reactive flows*

Fluctuation X-ray scattering - interacting complexes and assemblies in natural environments

Fluctuation X-ray scattering (fSAXS) is a promising route for 3-D imaging of anisotropic ensembles of interacting complexes in natural (solution) environments. fSAXS is an extension of well-developed SAX/WAX techniques, but in a regime in which the X-ray exposure is much shorter than rotational diffusion times. This gives rise to anisotropic scattering patterns (with annular correlations or fluctuations) which contain ~100 times greater information content than typical isotropic SAX/WAX patterns, thus enabling the reconstruction of 3-D objects.

LCLS-II instruments TMO and TXI will enable the full development of fSAXS as a powerful scientific tool. The requirements (and development road map) are quite similar to those for single particle imaging as described above, with the following differences and additional requirements:

- Adjustable X-ray focus from ~1 um to ~50 um (depending on sample concentration etc.)
- Photon energy range: both tender X-rays (2-8 keV[1]) and soft X-rays (0.3-1.2 keV) are needed for different applications, and to exploit larger scattering cross sections, resonant scattering, and transmission in the water-window.
- Moderate initial resolution goal >1 nm
- High repetition rate (limited by detector read out) to exploit the highest average X-ray flux.

[1] Experiments at high repetition rate with energy > 5keV will require LCLS-II-HE

In the fSAXS approach, annular correlations from many scattering patterns (at modest S/N) can be summed. Thus, fSAXS will exploit the high repetition rate of LCLS-II (limited by detector read out) to achieve the highest average X-ray flux. This will be complemented by the development of fSAXS at 120 Hz using the maximum flux/pulse to provide individual scattering patterns with high S/N.

Stimulated X-ray Raman Spectroscopy (SXRS)

Stimulated X-ray Raman spectroscopy represents a broad class of nonlinear X-ray processes that are anticipated to have significant scientific impact, and are uniquely enabled by the capabilities of LCLS-II. Here we development plans and basic requirements for 3 of the most important examples of SXRS: stimulated X-ray emission, coherent X-ray Raman, and X-ray coherent anti-Stokes Raman spectroscopy (XCARS):

Stimulated X-ray emission is a potentially powerful complement to spontaneous X-ray emission (XES or RIXS) processes for probing excited-state valence charge dynamics. Three key advantages are (1) weak emission processes can be enhanced by the stimulating photon, effectively competing with Auger and other relaxation channels, (2) the emitted signal of interest is directional, allowing for efficient 0-D detectors in place of collection optics which cover only a small fraction of the 4π spontaneous emission, and (3) the resolution is determined by the bandwidth of the stimulating pulse, eliminating the need for complex (and inefficient) spectrograph optics. Following are some of the key requirements for developing and exploiting this process:

- Two-color pulses with energy spacing tunable within ~100 eV (XANES region)
- Small energy spacing (~10 eV or ~2%) may exploit the two-bunch FEL approach with ~±35 fs relative time delay
- Larger energy spacing (and larger relative delay range) using the split undulator approach
- <10 fs pulse duration (comparable to the Auger lifetime), ideally close to transform limit
- Photon energy tunable over the soft X-ray range from 0.25-1.2 keV to access lighter elements (C, O, N,…) and the transition-metal L-edges, and in general to exploit larger cross-sections in the soft X-ray range.
- ~1 um focus to optimize X-ray nonlinearity (possible line focus for spatial resolution)

The above parameters can be met at instruments TMO and TXI, and the latter provides greater flexibility to accommodate specialized endstations to detect and develop stimulated X-ray emission for science applications.

Coherent X-ray Raman Spectroscopy (CXRS) is a nonlinear (2-photon) X-ray interaction, closely related to the stimulated emission process described above, but configured to create a non-stationary electronic wavepacket in a molecule (or extended solid) that is localized at an atom of interest, based on an X-ray core transition resonance. The wavepacket consists of an exited manifold of valence states, and is a potentially very powerful new means for understanding charge flow in matter (e.g. light harvesting complexes, photo-catalysts, etc.). Following are some of the key requirements for developing and exploiting this process:

- Broad coherent bandwidth (2-5 eV) to couple to a manifold of valence excited states

- <10 fs pulse duration (comparable to the Auger lifetime), ideally close to transform limit
- Photon energy tunable over the soft X-ray range from 0.25-1.2 keV to access lighter elements (C, O, N,…) and the transition-metal L-edges, and in general to exploit larger cross-sections in the soft X-ray range.
- Based on typical transition cross-sections, the "threshold" for nonlinear interaction is estimated to be ~10^{17}-10^{18} photons/cm^2. This corresponds to ~10^9-10^{10} photons/pulse in a 1 um focus (~10^{16} W/cm^2 at 500 eV).
- ~1 um focus (adjustable) to optimize X-ray nonlinearity

The above parameters can be met at instruments TMO and TXI. These also support a range of endstations capabilities (e.g. photo-electron and ion spectroscopy, XAS etc.) for characterizing the electronic wavepacket and associated dynamics. Ultimately we will exploit the two-color capabilities of LCLS-II to apply CXRS at one atomic site as an excitation, and a second CXRS process at a second atomic site as a probe.

X-ray Coherent Anti-Stokes Raman Spectroscopy (XCARS) is a variant of stimulated X-ray emission (as described above), but configured to probe dynamics of low-Z elements within a dense media (e.g. aerosols in ambient gas pressures, or chemistry in a combustion environment) with chemical/element specificity. Following are some of the key requirements for developing and exploiting this process:

- Two-color pulses with ΔE~280 eV (carbon edge)
- Tender X-rays for penetration of dense media (gas at atmospheric pressure, aerosols etc.)
- <10 fs pulse duration (comparable to the Auger lifetime), ideally close to transform limit
- ~1 um focus to optimize X-ray nonlinearity (possible line focus for spatial resolution)

The above parameters may be met at TXI using 3 keV photons from the HXU driven by the SCRF and 2.7 keV from the SXU (e.g. 3ω of 900 eV) similarly driven by a second delayed bunch from the SCRF. Compensation of the delay (~5 ns inter-bunch spacing) may be accomplished via Bragg optics. TXI provides access for optimized (roll-up) endstations to detect and develop XCARS for a range of science applications.

X-ray Core-hole Correlation Spectroscopy

This a two-pulse, two-color multidimensional X-ray technique that is a promising approach to reveal the quantum coupling between excited valence electronic states in molecules (e.g. light harvesting complexes). Following are some of the essential parameters to develop and exploit this process:

- Two-color pulses with >100 eV energy spacing tunable over the soft X-ray range from 0.25-1.2 keV to access different atomic species:
  e.g. C (280 eV), N (410 eV), O (535 eV), Fe L-edge (710 eV), Cu L-edge (910 eV)
- Split undulator approach for large energy spacing
- Relative pulse delay ~±35 fs from FEL
- <10 fs pulse duration (comparable to the Auger lifetime), ideally close to transform limit
- 0.1-1 um focus to enhance X-ray nonlinearity

The above parameters can be met at instruments TMO (or TXI with larger focus) with the caveat that the small focus will likely require the incorporation of a compact KB mirror pair within the endstation chamber (close to the sample).

Single-particle imaging – toward atomic resolution

LCLS and the scientific community have developed a comprehensive road map to advance the development of single particle imaging (coherent diffractive imaging), with a goal of reaching 3 Å resolution of biological objects[2]. Key X-ray source requirement identified in this road map include:

- Target photon energy range: 3-8 keV
- Pulse duration <20 fs with maximum flux/pulse
- Clean X-ray focus of 100-200 nm (minimize beamline scattering)

The TXI instrument will be developed toward meeting as many of these objectives as possible. In addition, an extension of scope to allow the warm Cu linac in combination with the SXU will generate X-rays with >5 mJ/pulse at 120 Hz. Beamline optics will deliver pulses to either TMO (0.25-3 keV) or TXI (0.4-3 keV), with KB mirrors to provide a clean ~500 nm focus.

Detector development for single particle imaging will focus on:

- Optimum quantum efficiency in the tender X-ray range
- Single photon counting at high-$\boldsymbol{q}$ and sub-Poisson noise elsewhere
- ~10^4 dynamics range, <100 μm pixel size
- High read out rates (>1 kHz)

In addition to the development of high-intensity *single-shot* X-ray imaging in the tender X-ray regime, LCLS-II will enable the development of new approaches to biological imaging from *large data sets* of relatively low-contrast scattering patterns collected at high repetition rate (limited by the detector read out). High repetition rate X-rays from the SCRF linac will be available at both TMO and TXI instruments.

Dynamic multi-view tomography – chemical mapping of reactive flows

The high repetition rate of the LCLS-II lends itself to imaging dynamic chemical processes *in-situ* or under operating conditions. The ability to image in 3-D chaotic real time processes such as turbulent flame combustion, liquid jet breakup, fracture propagation in materials, and self-ordering in crystallization provides important new insight and allows for the validation of computational models.

The general theme of 3-D dynamic tomography is the simultaneous view of the sample from multiple directions. Newly developed iterative reconstruction routines allow for only a few views and the additional constraints imposed by motion-tracking optical flow algorithms, gradient approaches, and spatiotemporal analysis constraint the individual images, improving reconstruction and reducing noise. Various optical schemes can be devised to optimize for a particular X-ray energy range of interest.

[2] A. Aquila, A. Barty, C. Bostedt, S. Boutet, G. Carini, et al., "The linac coherent light source single particle imaging road map," *Structural Dynamics*, **2,** 041701 (2015) doi: 10.1063/1.4918726

In the soft X-ray regime, a transmission grating (or convex grating in conical diffraction geometry) may be used to generate multiple diffraction orders. Multilayer mirrors may then be used to provide multiple instantaneous views of the sample from different angles. In the tender X-ray regime, crystals may be used in a wave-front splitting geometry to generate multiple beams, with additional crystals to re-direct the beams onto the sample to provide multiple instantaneous views. Following are some of the essential parameters to develop and exploit this process:

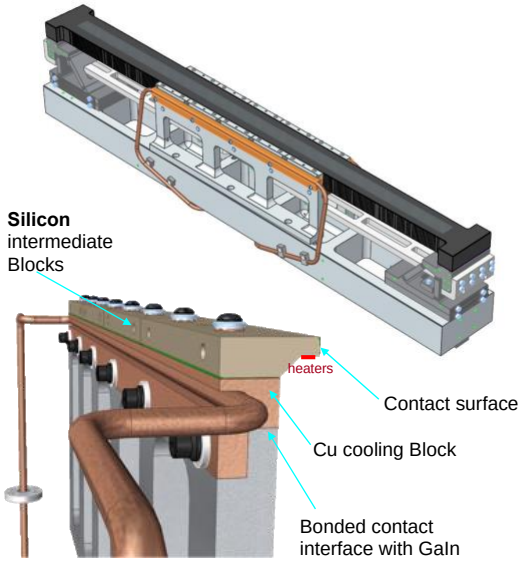
- Maximum average flux in the soft and tender X-ray regimes
- Optics for generating and manipulating multiple beams of soft or tender X-rays
- Fast 2D detectors in the soft and tender X-ray range
- ~25 um pixels, 1k×1k, ~10^2-10^3 ph/pixel/pulse

4.2 X-ray Optics Development

Distribution and focusing mirrors

To preserve the source characteristics, in particular to preserve the wavefront in and out of focus, the demands on mirror shape precision are very high. A shape error of 0.5 nm rms and a radius of curvature larger than several hundred kilometers is needed in most of the energy range. Such precision also needs to be preserved when the mirror is irradiated by the high repetition rate source, with as much as 200 W incident on the mirror. The main difference with respect to synchrotron radiation mirrors is the challenge of implementing liquid nitrogen cooling (due to contamination and vibration issues) and the fact that the footprint on the mirror changes considerably with the photon energy. In fact, a beam footprint smaller than the mirror length (as in the case of LCLS-II for certain energies) induces a significant temperature variation in the tangential direction. The resulting thermal 'bump' around the beam footprint is not spherical and is therefore difficult to correct with a bender.

Figure 1 Schematic of the REAL optics assembly for a ~1m long X-ray distribution mirror, combining a cooling system with resistive heating elements to maintain the figure.



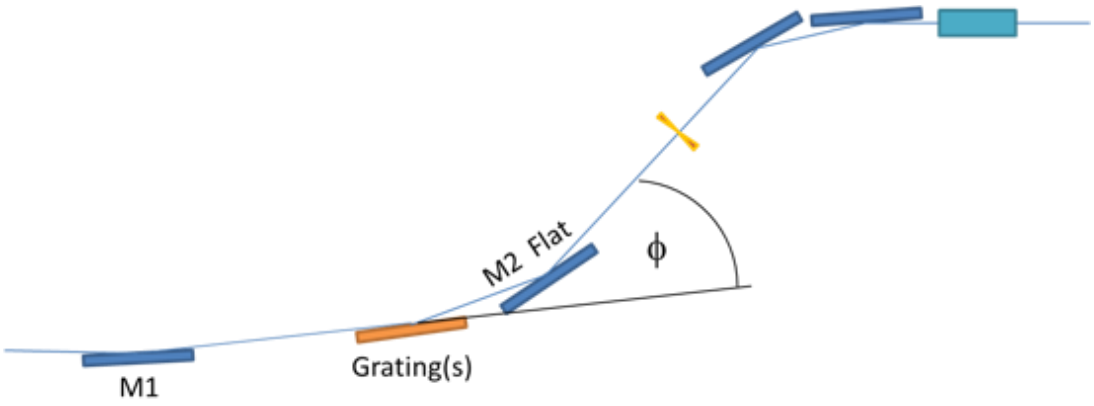
To face such challenges, LCLS is developing a new scheme for active shape control, which combines variable cooling with applied auxiliary heating, and is tailored to the spatial distribution of the incident thermal load. These developments were funded in part directly by DOE-BES via the REAL project, involving a partnership with BNL, ANL and LBNL. The underpinning approach, known as "Thermal Portrait Mirror Figure Control" (TPMFC), may achieve sub-nanometer surface figure error control (see **Figure 1**). An initial finite-element study suggests that TPMFC can reduce surface height errors by more than 10x, compared to traditional, full-length cooling. This should enable the distribution of beams significantly in excess of 100W.

A current focus of the optics research effort is the longevity of the mirrors at these extreme repetition rates due to thermo-mechanical fatigue (with ~10^{13} pulses per operating-year). Evidence from optical irradiation shows that the acceptable absorbed fluence can drop substantially with time.

Variable resolution beamline for RIXS experiments

A pair of RIXS instruments are planned for the upper floor of the NEH, designed to cover the energy range 250 eV to 1500 eV. To deliver the beam, it is necessary to tilt the beam to quite a large angle, 7.1°**,** while ensuring any single angle of incidence is kept below 1.6° in order to reach 1500 eV and to avoid mirror and grating ablation. The concept is to use a first mirror (M1) with fixed incidence angle and use the monochromator system, Grating-M2, to deflect (or diffract) further (Figure 2).

Figure 2 Scheme of the Variable resolution beamline. M1 is a bendable mirror deflecting the beam upward by 2.2° (full angle). The grating-M2 deflects the beam up by further 4.9° and the following two mirrors, one flat and one focusing, bring the beam again parallel to the ground. A sixth mirror is used to focus the beam horizontally.



To obtain variable resolution (from <3,000 to >50,000) it is necessary to both use a few gratings (three minimum) and to change the working configuration of the grating. This is achieved through the use of a bendable first mirror. This can produce a virtual focus behind the grating to reduce the resolving power when needed, preserving the total deflection onto the monochromator. To continue to focus the beam into the fixed exit slit, the angle of incidence on the grating shall be increased when the source becomes negative (e.g. when the beam is focused by mirror M1). This preserves the angle of diffraction by reducing the

number of illuminated lines and the angle of diffraction. Both effects reduce the resolving power. A maximum safe angle of incidence shall be preserved to avoid ablation on the grating. The grating will have a variable groove density. The resolution can be changed by focusing the beam with M1, to change its shape from flat (e.g. no source distance alteration) to a minimum of 3 km of radius of curvature for low resolution.

Considering all the mirrors and the grating efficiency, optimized for flux rather than for high harmonic suppression, the photons per second delivered to the sample can be as high as 10^{15} even for a resolution of 50,000. This large number of photons is mainly due to the high repetition rate of the source. This gives several orders of magnitude of flux enhancement delivered to the sample with respect the most advanced synchrotron beamlines with comparable resolving power.

qRIXS spectrometer

The qRIXS spectrometer for NEH2.2 is based on a spherical VLS grating with fixed angle of incidence and to achieve a net resolving power of 30,000 or higher across the photon energy range. The distance between sample and grating is set to be 1 m and the maximum detector distance from the sample will be below 6 m (Figure 3).

Figure 3 Scheme of the proposed 6 m qRIXS spectrometer

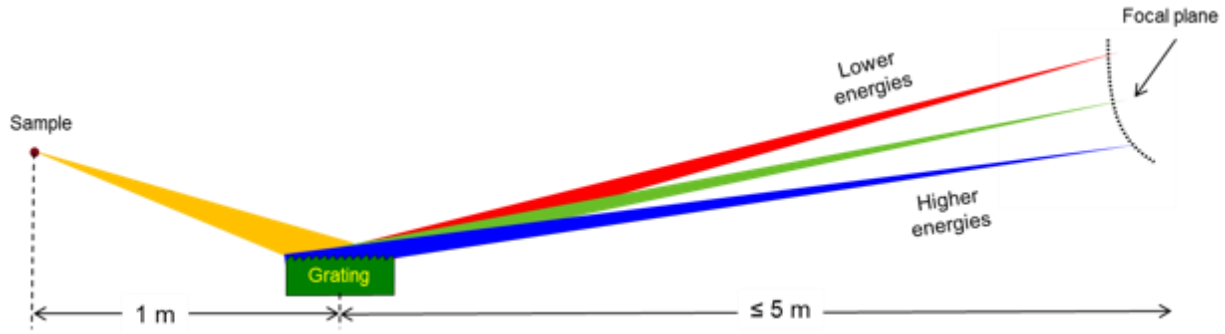
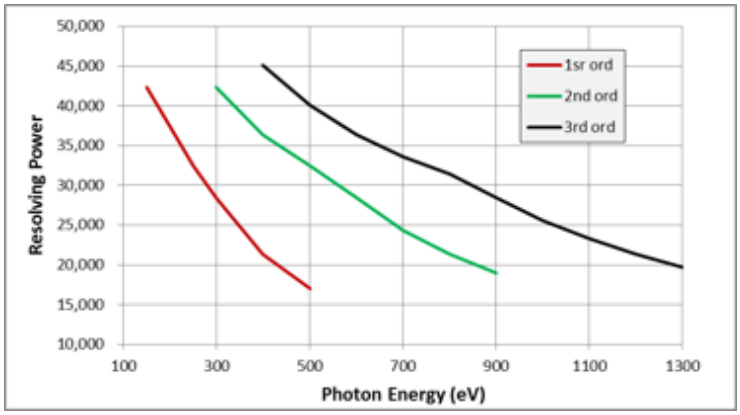


Figure 4 Expected resolving power for the 1800 l/mm grating. A 10 um spot size and 10 um spatial resolution for the detector are considered in the simulation.



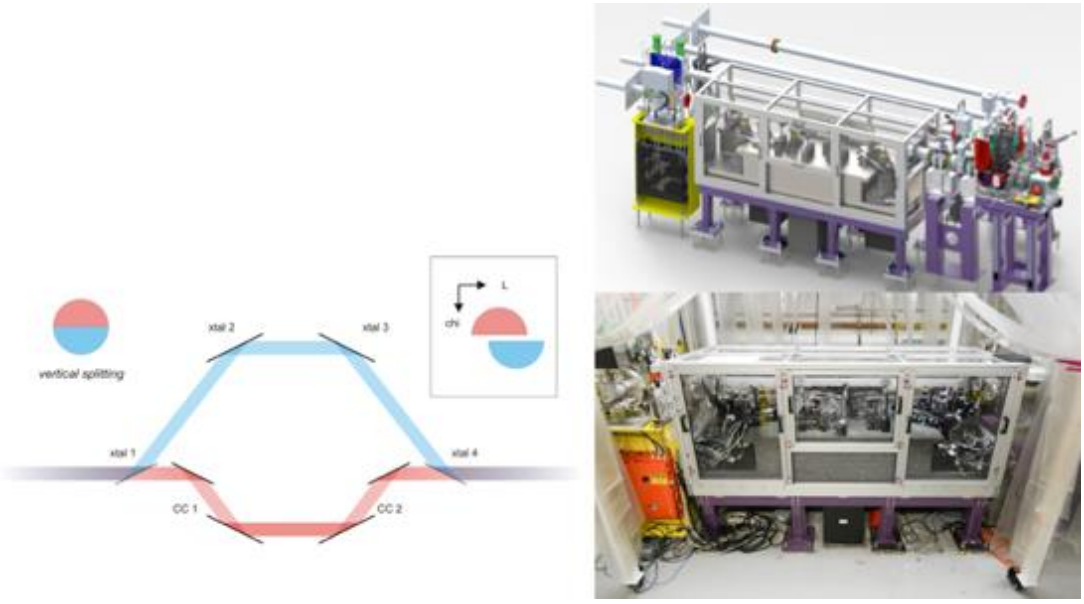
The expected resolving power is calculated considering a 10um vertical spot on the sample and 10um spatial resolution for the detector (Figure 4). A resolving power in excess of 30,000 is expected up to 900 eV, while it is larger than 20,000 over the entire range. At the maximum resolution, a count rate as high as 100,000 counts/sec are expected for standard RIXS spectrum peaks. This is roughly three orders of magnitude higher than the achievable levels on synchrotron sources with lower resolution.

Split and Delay X-ray optical systems

A significant advance for near-equilibrium dynamics is a set of newly commissioned X-ray optical systems for generating two X-ray pulses, introducing a controllable delay with respect to each other, and recombining them with interferometric precision at the interaction point. An initial set of X-ray pump/probe and two-pulse XPCS experiments have been completed that exploit this new "split and delay" capability, for which a robust, easily tunable system has been sought by users since the early days of LCLS operations.

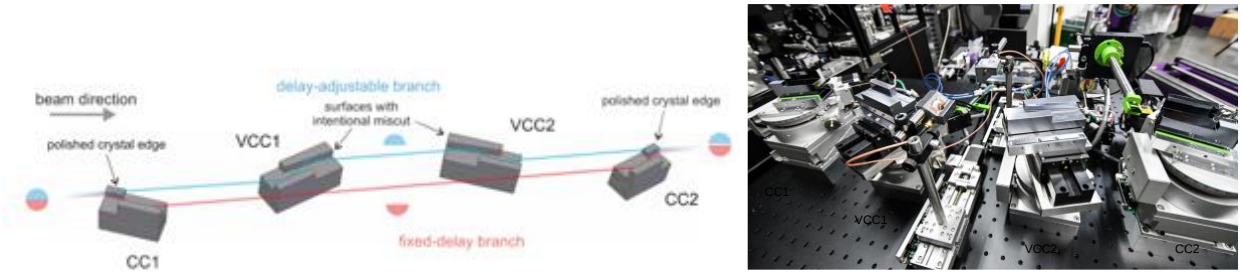
The first system (built for the XCS instrument, shown in Figure 5) operates over an energy range of 7 to 12 keV, with a delay range from -5 to 350 ps at 8 keV, providing continuous coverage by bridging to the multi-bunch operating mode of the accelerator (that has ~350 ps bunch spacing).

Figure 5 Schematic and photo of the XCS split and delay system.



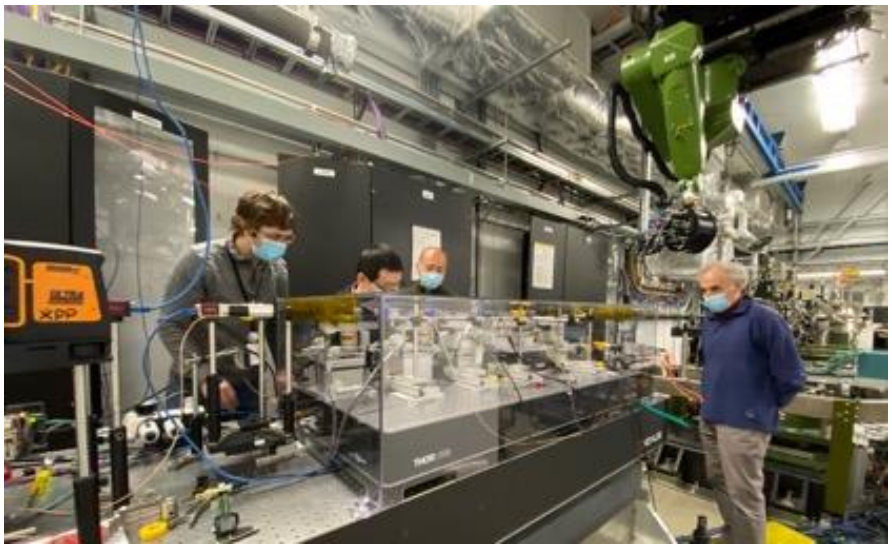
More recently, a compact split and delay system has been installed, with four Si(220) crystals in a horizontal scattering plane (Figure 6). Two of the channel-cuts have non-parallel gaps. The delay is introduced via a linear translation of the crystals that have an intentional mis-cut, so that angular error of the linear translation largely cancels through the two bounces in the opposite direction from a monolithic crystal. This allows for ultra-stable operation over a limited temporal range (0 to 20 ps).

Figure 6 Schematic and photo of a new, compact split and delay system with 4 channel-cuts.



In 2020, a new amplitude splitting technique is being tested that incorporates the double channel-cut system combined with a pair of tranmission gratings. This has been designed[3] to cover a delay range of 15 ps with a sub-20 fs time resolution at 10 keV, sub-nanoradian stability, and preservation of the wavefront for high quality coherent beam overlap (see Figure 7). Initial indications are that this system provides exquistite performance.

Figure 7 Schematic of the amplitude split/delay system, and its installation in the XPP instrument.



[3] Haoyuan Li et al, Optics Letters 45(7), 2086-2089 (2020). https://doi.org/10.1364/OL.389977

4.3 Sample Delivery Systems

Sample delivery capabilities for liquid systems is for the most part common between LCLS instruments. The mechanical systems to position nozzles to deliver samples to the X-ray interaction region differ but the nozzle technology as well as the liquid and gas pressure systems to deliver and monitor flow parameters are common. LCLS maintains, deploys and operates high-performance liquid chromatography (HPLC) pumps, gas pressure regulators, gas mass flow controllers, as well as flowmeters and sample cooling, shaking and switching systems. The SED group supports the operation of Gas Dynamic Virtual Nozzle (GDVN), Double Flow Focusing Nozzles (DFFN), Lipidic Cubic Phase (LCP) and other high viscosity nozzles as well as the Microfluidic Electrokinetic Sample Holder (MESH) and its rapid mixing variants. Several sample environment and delivery themes emerge as priorities for development:

- Versatility/Optimization for specific needs of various classes of bio complexes (buffers, other conditions…..), as well as different methods such as solution scattering or SPI.
- Operational efficiency (robust, reliable, automated, user friendly, low- maintenance).
- Reduced sample volume demands (to enable access to precious proteins and the most important bioscience problems) consistent with LCLS science advantages for:
    - Mapping dynamics in near-physiological conditions
    - Exploiting diffract-before-destroy to mitigate radiation effects
    - Higher repetition rates
- Push the state-of-the-art for mix/inject to access faster dynamics of interest across a broader range of bioscience problems beyond pump/probe

4.4 X-ray Detector Systems

The present suite of detectors operating at 120 Hz has evolved through many iterations of calibration and optimization cycles to reach the current operational status. Data acquisition and analysis platforms and routines tailored to specific experiments have been developed, refined, and made available for non-expert users. They take into consideration both the event level sorting/filtering as well as performance optimization for processing very large data sets. Figure 8 provides a list of detectors commonly used at the hard X-ray instruments.

Figure 8 Pixel array imaging detectors currently available at LCLS.

	Pixel size	**Num. of Pixels**	**Dynamic range** (8 keV photons)	**Status**
CSPAD	110 μm	0.14M, 2.3M	0.3-1,000	To be retired (120 Hz)
ePix100	50 μm	0.5M	0.05-1,000	Many available (120 Hz) for low noise studies
ePix10k	100 μm	0.14M, 2.1M	0.05-11,000	Available for experiments in 2020 (currently 120 Hz, with development plans up to 25 kHz)
Jungfrau	75 μm	0.5M, 1M	0.05-10,000	Available for experiments (up to 2 kHz)
Rayonix MX-340-XFEL	44 μm	3.6M (2x2 binning)	5-50,000	Nominally 10 Hz (up to 120 Hz with higher binning ratio)
Andor Zyla	6.5 μm	4M	N/A	Low noise optical sensor, used with scintillator based imaging system.

Future development requirements can be summarized as follows:

- **Imaging non-identical objects**: A 2D detector is required with large dynamic range, high QE for tender/hard X-ray energies, capable of operating at rates of up to 100 kHz. Frames contain information in the majority of pixels, thus requiring the readout of the entire frame.
- **Stochastic dynamics**: XPCS techniques will benefit from a 2D integrating detector with fine spatial resolution operating at the full MHz pulse rate. Information in these experiments is "sparse" and confined in a limited number of pixels per frame, with each pixel containing a limited number of photons (small dynamic range). Extracting information using a sparsified readout would allow efficient implementation at MHz rates with contained data volumes.
- **Rare events**: This would benefit from a detector operating at the full 1 MHz rate. Information in rare events is sparse in time, and so an event-driven triggered detector capable of extracting frames centered on a rare transient event would produce acceptable data rates. Coarse MHz frames could be potentially used to generate feature-based triggers locally, via automatic pattern recognition and related machine-learning approaches.
- **Molecular dynamics** (e.g. COLTRIMS): To take full advantage of the timing options of FELs (multi-bunch, multi-polarization and sub fs-pulses), a highly segmented TOF particle detector accommodating hit rates of 6khits/s/cm2 , and a time resolution of 100ps is needed.
- **Extreme states**: Fast framing imaging detectors are needed to take full advantage of the new laser drivers and diagnostics capabilities from LCLS, such as an X-ray pulse train that allows measurements of material dynamics with ultrafast temporal resolution at multi-GHz rates.

To achieve these goals, multiple pathways are being developed in parallel, to reduce the risk to delivery in time for LCLS-II.

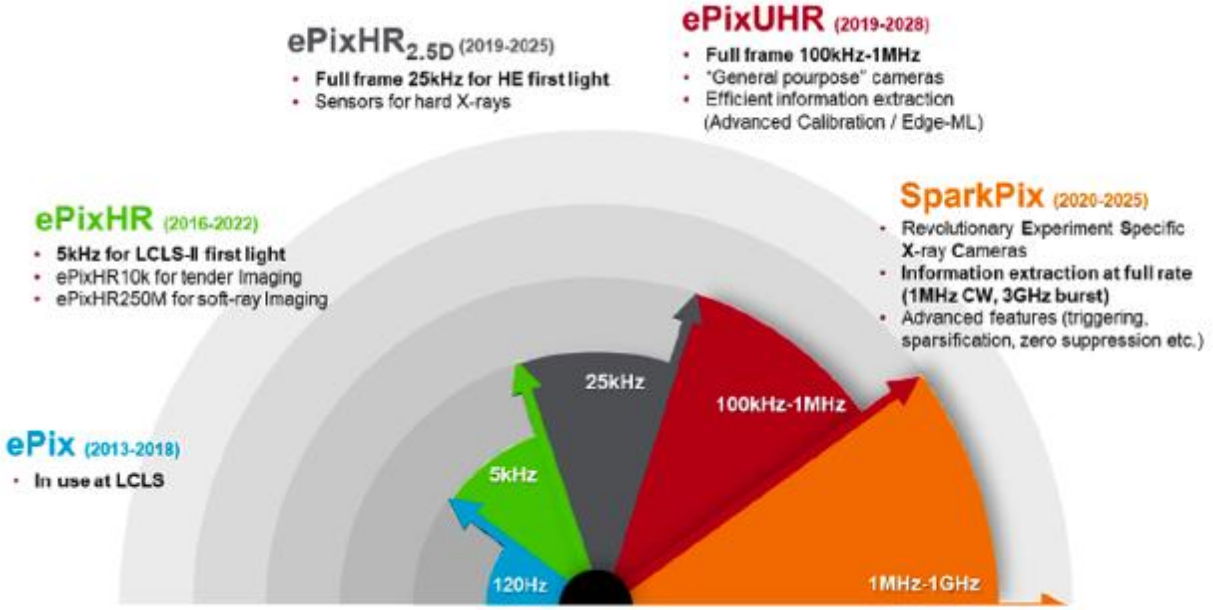
The most taxing requirements are to provide area detectors for soft and tender X-rays at a readout speed of few kHz for the initial period of LCLS-II operations, scaling to tens of kHz thereafter for full-frame, full-depth readout, and MHz rates for sampled/sparsified readout. Similar performance will be required in the hard X-ray regime for LCLS-II-HE. Alongside this, SLAC is pursuing the development of Transition Edge Sensor (TES) technology to enable spectroscopy at very high repetition rates with sub-eV resolution. More conventional spectroscopy and detection systems employing 1D readout are already available, able to function at the full (1MHz) repetition rate.

To cover the entire soft-, to tender-, to hard- X-ray regime, hybrid pixel array detectors based on the ePix family are being developed at SLAC, as shown in Figure 9. Two parallel development paths have been identified. The ePixHR/UHR path: a platform-based approach extension of the present generation of ePix detector family toward full frame readouts up to 100kHz, and a new class of experiment specific detectors named SparkPix dedicated to a class of experiment where information extraction in real-time can push operation to 1MHz with optimized performance and with manageable data throughput.

The ePix class of detectors is currently able to provide a readout speed matching the LCLS repetition rate (120 Hz) and support Region of Interest (ROI) mode for faster readout of a subset of pixels. In the next generation, these cameras will be able to provide multi-kHz readout speed with trigger and veto capabilities, while maintaining low noise and high maximum signal (10,000 8 keV photons equivalent). Extension to 10's kHz operation is

under development, for deployment in the mid-2020s. Thanks to the intrinsic scalability of hybrid pixel array detectors, large areas can be covered and maintained in a sustainable way. Specifically, two major development projects are underway: ePix-M (based on CMOS technology for the soft X-ray regime), and ePix-HR (as an extension to the current ePix-100 and ePix-10k systems for the tender/hard-ray regime). The current versions of these ePix detectors can be modified to use different sensor materials, such as Ge or GaAs, to increase the quantum efficiency at higher photon energy (>18 keV).

Figure 9 Development path at SLAC of 2D X-ray detectors for LCLS.



To complement this development for soft X-ray applications, a "very-fast CCD" (vfCCD) is being developed in collaboration with LBNL. This MPixel area detector will provide high quantum efficiency in the 250-1200 eV energy range with very low noise and kHz frame rate.

To address high field physics applications, a detector is needed that is capable of measuring the 3d momentum for hundreds of particles per laser shot in coincidence at very high pulse repetition rates. A prototype SparkPix 'Tixel' detector is under development at SLAC to meet this need, as well as exploration of the potential use of Timepix systems being developed at CERN and LBL. Such systems may also be adaptable for 'sparsified' readout for applications such as X-ray photon correlation spectroscopy, with the potential of increasing frame rate to up to 100 kHz. If successful, this type of detector could impact multiple other experimental techniques, with the potential to replace the delay-line detectors in COLTRIMS and coincidence spectrometers (while removing the number-of-hits-per-shot limitation), phosphor-based velocity-map-imaging detectors (removing the rep-rate limitation), and supersede most other position-sensitive particle detectors.

4.5 Data Systems

With LCLS-II and HE, the repetition rate will increase from 120 Hz to 1 MHz, requiring significant growth of the present data environment (computing platforms, network, storage and data processing software, and the manner in which users access their data). These experiments present some challenging characteristics for the computing and software systems:

- Fast feedback on the quality and experimental implications of the streaming data, on timescales of seconds-to-minutes.
- 24/7 system availability; high reliability; and accessibility by a broad user community.
- Bursts of short jobs, requiring very short startup time on large scale compute platforms.
- Storage represents a significant fraction of the overall system (in cost and complexity).
- Throughput between the front-end electronics and the storage layers and between storage and processing is a critical element of the system.
- Speed and flexibility of the development cycle (due to the wide range of experiments, the rapid turnaround required, and the need to modify data analysis during experiments.
- Significant increase in the data throughput compared to LCLS-I, from GB/s to TB/s.

Together, these attributes call for an extensive investment in the enabling software for data extraction, processing, and analysis.

Another critical feature is the deployment of a system to veto the readout process for uninteresting events, and/or reduce the data throughput through other means, such as regions of interest, or on-chip processing to extract the required information. Development of such a "**Data Reduction Pipeline**" (DRP) requires in-depth consultation for each class of experiments, to arrive at a practicable balance between data throughput and subsequent data mining capability. This is currently underway, working with the LCLS Users Executive Committee (UEC) and individual user groups to determine the most appropriate solutions.

At the beginning of 2016 we published a document describing the strategy for LCLS data analysis:https://portal.slac.stanford.edu/sites/lcls_public/Documents/LCLSDataAnalysisStrategy.pdf.

This strategy is motivated by the above characteristics, combined with a drive to decrease the *time to science*, and to ensure that *no user is left behind*. The latter point is driven by the fact that most of the advanced algorithms for analysis of LCLS science data have been developed by external groups with enough resources to dedicate to a leading-edge computing effort. Smaller groups with good ideas may be hindered in their ability to conduct science by not having access to these advanced algorithms or forced into collaborations with larger groups. LCLS support for externally developed algorithms and the development of in-house algorithms for some specific science domains, would alleviate this problem.

Update on data systems development

Extensive effort has been devoted to this topic over the past 3 years, integrating the input from a wide-ranging series of community workshops, and coordinating the effort across the 5 BES light sources and from other national laboratories in the US. Quantitative, time-phased

requirements have been derived from a detailed analysis of over 20 anticipated experimental workflows, scaled according to expected scheduling demands in the LCLS-II and LCLS-II-HE era. The approach, and provisional conclusions, were the subject of a wide-ranging review held at SLAC in October 2017. Substantial funding has been allocated to the definition and development of the required computing platforms, scalable software architectures, and efficient algorithms for real-time data reduction, assessment, and deep analysis. This work has coordinated funding and direction from LCLS Operations (tied to the L2S-I instrument program), BES/ASCR (e.g., the ExaFEL and LLAna grants, and funding for machine learning), and multi-laboratory efforts in algorithm and simulation development.

The scientific objectives of the suite of "First Experiments" for LCLS-II and LCLS-II-HE were assessed, and a set of over 20 scientific workflows defined for each of the representative experiments, mapping each to the envisioned instruments and detectors, and their evolution over time.

For each experiment, 4 phases of data processing were assessed: (i) data extraction and reduction, (ii) real-time fast feedback to inform the experimental execution, (iii) off-shift data analysis, (iv) long-term data interpretation.

The suite of notional experiments was distributed according to a detailed mock schedule covering the next few years (including a nominal mid-decade transition to LCLS-II-HE). A large-scale spreadsheet was constructed that analyzed:

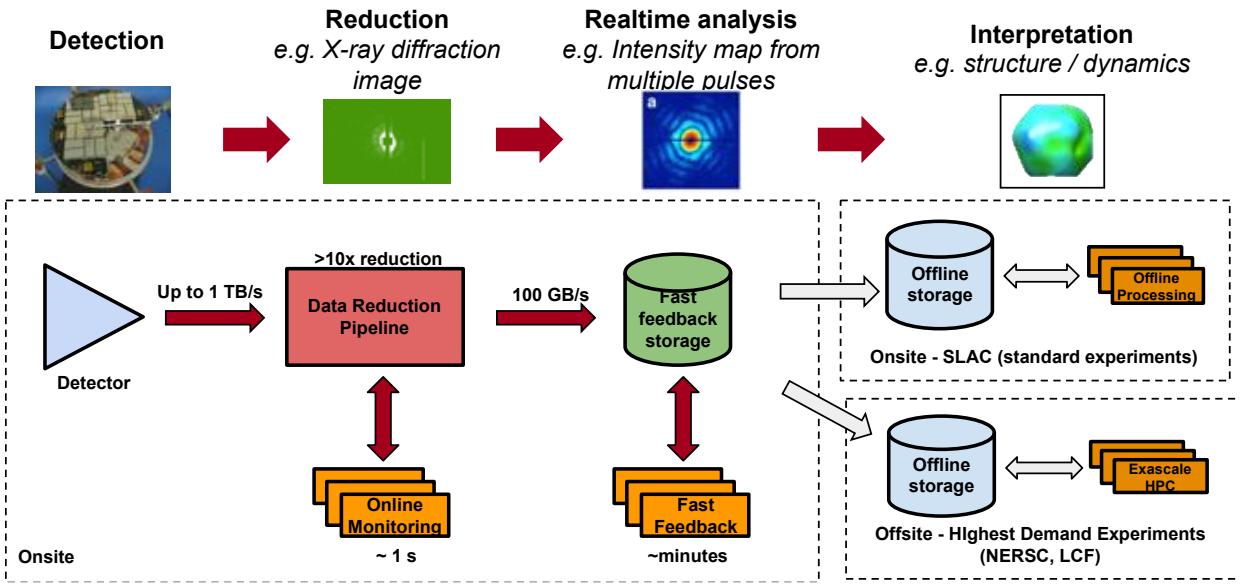
- Distribution of experiments: # shifts allocated per instrument and technique;
- Typical uptimes per experiment, based on extrapolated LCLS experience-to-date;
- Detector rates for each LCLS-II instrument (and their increase over time);
- Data reduction capabilities based on the experimental techniques (this could vary from factors of 2 to over 10^6 rate reduction, dependent on the data type);
- Algorithm processing times for each experimental technique, for real-time assessment and for long-term analysis.

From this, a set of quantitative, time-phased requirements were defined for the hardware, networking, storage, and associated data processing systems.

Where possible, LCLS will pursue partnerships with other facilities and make every effort to leverage the work and expertise of the community to solve common problems. LCLS personnel will encourage the collaboration between software engineers, instrument scientists and users to ensure that code and algorithms are shared. Open source tools will be used where feasible, and shared repositories will be used to store and distribute code to users. LCLS personnel will work closely with the users and scientific staff to provide resources to install, understand, and use the analysis software. By participating in the development of these algorithms and actively communicating with users, LCLS staff can ensure that these algorithms are portable and reusable around the world.

A schematic of the new workflow for LCLS-II era experiments is shown in Figure 10.

Figure 10 A 4-step description of the data flow for LCLS-II and HE.



In collaboration with the other DOE-BES light sources, a strategic approach for development of this area has been proposed. Known as DISCUS ("Distributed Infrastructure for Scientific Computing for User Science"), this recognizes that coupling the BES Light Sources to advanced computing capabilities will enable new opportunities over the next decade:

- Leverage the exabyte (EB) of data per year across the Light Sources to increase the scientific knowledge base
- Utilize 10's PFLOPS to 1 EFLOPS of on-demand peak computing to reduce time to science
- Unlock exceptionally challenging experiments by utilizing billions of core hours per year
- Fully leverage the AI/ML capabilities now being developed across the facilities

Unified solutions are required in order to leverage efficiencies of scale, and to provide facility users with the ability to easily and transparently manipulate data across the complex; this is needed now more than ever to ensure the productivity of remote users due to the COVID-19 landscape.

Six areas of development are being incorporated into our joint planning:

i. **Algorithms & AI/ML –** New methods to enable advanced production, processing, analysis and interpretation of data;

ii. **Scalable Software Library –** Scalable common software tools for reduction, processing, analysis, interpretation, and visualization that operate across a multi-tiered computing landscape;

iii. **Workflow & Orchestration Tools –** Couple beamline instruments at facilities with edge, local, Laboratory, and ASCR computing resources to run data reduction, processing, analysis, interpretation, and visualization tasks;

iv. **Seamless Real-time On-demand Computing –** Enable fast feedback and experiment steering across the range of computing resources within the complex;

v. **Networking Improvements –** Transparently connect instruments, facilities and Laboratories across the complex;

vi. **Discoverable Data Repositories –** Provide sufficient and sustainable storage, cataloging, searching, and publication services for knowledge dissemination and future scientific discovery.

4.6 Laser Systems

Approximately 75 percent of all experiments at the LCLS require optical laser excitation to initiate a reaction or state within the sample under investigation. The vast majority of the physical phenomena under investigation by pump/probe techniques occur on a timescale of picoseconds to femtoseconds, requiring both optical and X-ray sources to have femtosecond pulse duration and a well-defined and controllable temporal separation. LCLS-II will present challenges for optical lasers primarily in two broad areas: (1) increasing the repetition rate of the lasers to match that of the LCLS-II X-ray beam (up to ~1 MHz) with suitable pulse energy, (2) maintaining low temporal jitter and drift relative to the X-ray beam. Related challenges will include managing high average power beams with robust personnel and machine safety systems, producing sufficient pulse energy at wavelengths of interest where the conversion efficiency is low (e.g., THz, deep-UV), and producing and maintaining increasingly shorter pulse durations to take full advantage of the high time fidelity of the X-rays and optical beams for experiments on extremely short time scales. Alongside this, new opportunities in high energy density (HED) science will drive demand for increased optical laser intensity and very high per-pulse energy, as described in Section **Error! Reference source not found.**.

Lasers for LCLS-II

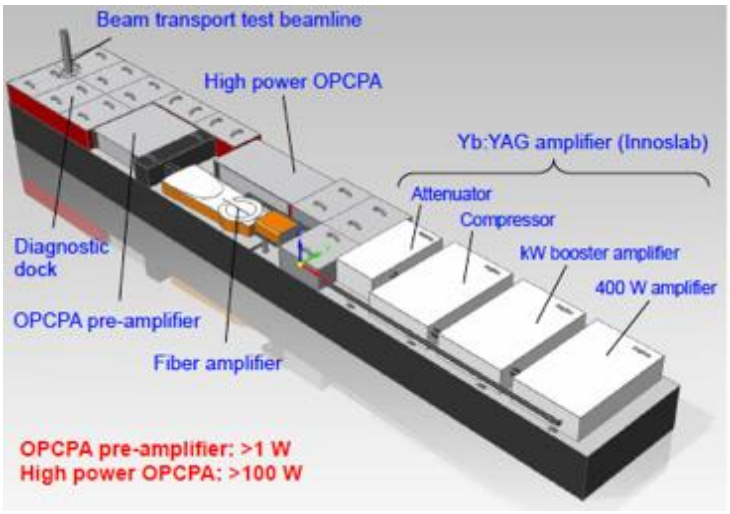
In the baseline design for the LCLS-II superconducting accelerator, the maximum repetition rate for electron bunches (and therefore X-ray pulses) is ~1 MHz. While specific experiments may be limited by the rep-rate of detectors, sample refresh rates, or related considerations, the majority of science opportunities will require lasers for pump-probe measurements at THz to EUV frequencies and high average power. Pump-probe systems of this scale have a major impact on the overall facility layout, requiring extensive tradeoff analyses and close integration throughout the design.

LCLS has initiated an R&D program to develop a 100 W average power (1 mJ at 100 kHz or 0.1 mJ at 1 MHz) laser system to assess the feasibility of applying this technology to LCLS-II. Based on Optical Parametric Chirped Pulse Amplification (OPCPA), a target specification of ≥1 mJ was selected to provide sufficient peak power to drive low-efficiency wavelength conversion processes such as THz generation by optical rectification, which has a $\sim 10^{-5}$ conversion efficiency. Other targeted baseline performance parameters include 15 fs FWHM pulse duration (scalable to <10fs), broad spectral tunability, and timing synchronization jitter ≤20 fs RMS from 0.01 to 1 MHz.

This system, shown in Figure 11, is based on a commercial oscillator for seeding the amplifier and the pump laser for passive synchronization, a commercial Yb-fiber pre-amplifier, and several stages of OPCPA pumped by a commercial Yb:YAG amplifier producing >1 kW average power with a few picosecond pulse duration.

The architecture of the system is designed to enable enhanced performance through upgrades or alternative modes of operation. Substantial development is required in terms of characterizing thermal properties of nonlinear optical crystals for OPCPA and modeling the amplification process prior to design and operation of an amplifier.

Figure 11 Schematic and key performance parameters for the high repetition rate OPCPA laser system for LCLS-II pump/probe operation



OPCPA	
pulse energy	1 mJ, 100 W (signal)
wavelength range	0.7-1 μm
pulse duration	15 fs
	<5 fs, 60 W with spectral broadening
pulse energy	1.5 mJ, 150 W (signal)
wavelength range	1.4-1.7 μm
pulse duration	50 fs
	<10 fs with spectral broadening
pulse energy	1 mJ, 100 W (idler)
wavelength range	2.6-3.8 μm
pulse duration	70 fs
	<15 fs with spectral broadening
pump laser	
pulse energy	15 mJ, 1.5 kW
wavelength range	1.03 μm
pulse duration	1.5 ps
	<200 fs with spectral broadening
other secondary sources	
Harmonics (OPCPA and pump)	SHG, THG and FHG
tunable OPA	(pumped with 1 mJ, 50 fs, 800 nm)
pulse energy	μJ-tens of μJ
wavelength range	200 nm-20 μm
pulse duration	> 40 fs
THz source	DFG or Optical rectification
pulse energy	to be investigated
wavelength range	0.1-100 THz

More broadly, the development of high average power, ultrashort laser systems has developed rapidly over the past few years, both in results from research institutions, and in commercial laser products (e.g. with regard to Ti:Sapphire systems, and fiber lasers that offer breakthrough potential with emerging research on coherent combining techniques and non-linear spectral broadening technology). The choice of laser technologies that will be implemented will be informed by continued clarification of the scientific requirements for LCLS-II science, continued advancement in the capabilities of commercial lasers, and the knowledge gained as part of the of the above R&D project.

Laser Synchronization and Timing

Taking full advantage of the temporal resolution of femtosecond X-ray pulses and femtosecond optical lasers in pump-probe experiments requires timing measurement and control of the variable delay between the pulses on a timescale from a few tens of fs to sub-fs. At present, LCLS has a pulse-to-pulse timing jitter relative to the accelerator radio-frequency (RF) distribution of approximately 60 fs RMS, integrated over a bandwidth of 0.1 to 100 kHz. Optical lasers must be locked to the accelerator RF distribution with similar or better timing jitter, and drifts in the laser beam path and RF distribution must be controlled

to the ~1 ps level. A detailed description of the timing distribution and oscillator locking electronics developed at SLAC has been published[4].

The CW RF drive and high Q of the superconducting accelerator cavities of LCLS-II promises to significantly reduce the timing jitter, possibly to as low as a few fs. This improvement will drive a corresponding demand for improvements in the jitter of the pump-probe lasers and timing distribution systems, particularly in experiments that probe ultrafast dynamics of transient systems.

For LCLS-II an upgraded version of the current RF timing system (which is based on RF distribution over stabilized coaxial cable) will employ RF-over-fiber.

An R&D project is currently underway to evaluate a second timing distribution system technology based on pulsed femtosecond laser signals over optical fiber utilizing optical balanced cross-correlators for timing detection[5]. In the cited reference, remote two-color optical-to-optical synchronization was demonstrated with 3.3 fs RMS slow drift over 24 hours between a Ti:sapphire oscillator and an Er-doped fiber laser. Similar systems are already in use (DESY/FLASH) or planned (European XFEL and SwissFEL) for precision synchronization of optical laser with FEL X-ray beams.

To complement the stabilized timing system, cross-correlation techniques that provide a pulse-by-pulse measurement of the X-ray/optical synchronization have proven to be highly useful at LCLS to date[6,7]. Investigations are now underway to study methods of scaling cross-correlation techniques up to the 0.1-1 MHz repetition rate of LCLS-II, and to improve the temporal resolution of this technique, ideally down to the <10 fs level.

Upgrades of Pump/Probe Laser Systems

The Ti:sapphire laser systems for 120 Hz operation in the NEH are mature and stable, requiring only routine maintenance and incremental upgrades such as replacing older hardware, e.g. to improve timing jitter performance, and adding new or improved diagnostics. The mid-IR OPA setup has been very successful in reducing set-up time and providing consistent output and diagnostics of the mid-IR source. Similar mobile, well-defined configurations will be developed for the optical THz source and for the <10 fs hollow fiber source. Further improvements in THz conversion efficiency by cryogenic cooling of LiNbO3 crystals is being developed by researchers at LCLS, and this will be engineered for deployment in the LCLS hutches. Other R&D activities on THz sources include the development of collinearly-pumped THz using organic crystals, CEP-stabilized mid-IR pulses, and narrow-band, multi-cycle THz sources.

Laser system improvements include: (1) upgrades to accommodate <25 fs laser pulses for higher temporal resolution in pump/probe experiments; (2) dedicated harmonics and OPA

[4] Gumerlock, K, et al. FEL2014, Basel, Switzerland, 2014.
[5] Li, H, et al., Remote two-color optical-to-optical synchronization between two passively mode-locked lasers. *Opt. Lett.* 2014**,** *39* (18), 5325
[6] Schorb, S, et al., X-ray–optical cross-correlator for gas-phase experiments at the Linac Coherent Light Source free-electron laser. *Appl. Phys. Lett.* 2012**,** *100* (12), 121107
[7] Bionta, M. R., et al., Spectral encoding of X-ray/optical relative delay. *Opt. Express* 2011**,** *19* (22), 21855

configurations for each instrument to reduce time spent moving and aligning shared systems; (3) additional diagnostics for on-line monitoring during experiments; (4) development of extended capabilities in wavelength (XUV, UV, THz) and pulse duration (<10 fs); (5) further development of existing mid-IR and THz techniques and diagnostics.

R&D efforts on optical sources, such as cryo-cooled THz, <10 fs sources, UV and HHG sources, etc. are underway in two LCLS laboratories, housing systems identical to those used for hutch operations. These labs also provide laser light to users to test samples, detectors and measurement schemes prior to beamtime. The new Arrillaga Science Center (ASC) laboratory building at SLAC now provides space for laser laboratories to further develop sources and experimental capabilities, working closely with researchers from the PULSE Institute and external groups.

A 30fs, 6mJ laser system in XCS provides ultrafast optical pump pulses at wavelengths spanning from the visible to near-infrared, providing similar capabilities to those of XPP.

In CXI, an increasing number of experiments require higher pulse energy and changes in wavelength using OPAs, with both at nanosecond and femtosecond lasers. Typically, each of these configurations must be set up from scratch and disassembled after the experiment. To increase operational efficiency, we are developing stable, interchangeable configurations of standard optical setups such as an OPA stage or harmonic conversion. To enhance the laser capabilities and meet user demand, a compact multi-pass amplifier provides ~20mJ pulse energies for driving shocks in materials and extending wavelength conversion capabilities to the mid-IR and THz.

In the MFX instrument a tunable nanosecond laser system has been installed to generate pump wavelengths spanning 410-2200 nm. Similar ultrafast laser systems to those in CXI will enable time-resolved experiments on this instrument.

5 ACCELERATOR SYSTEMS DEVELOPMENT

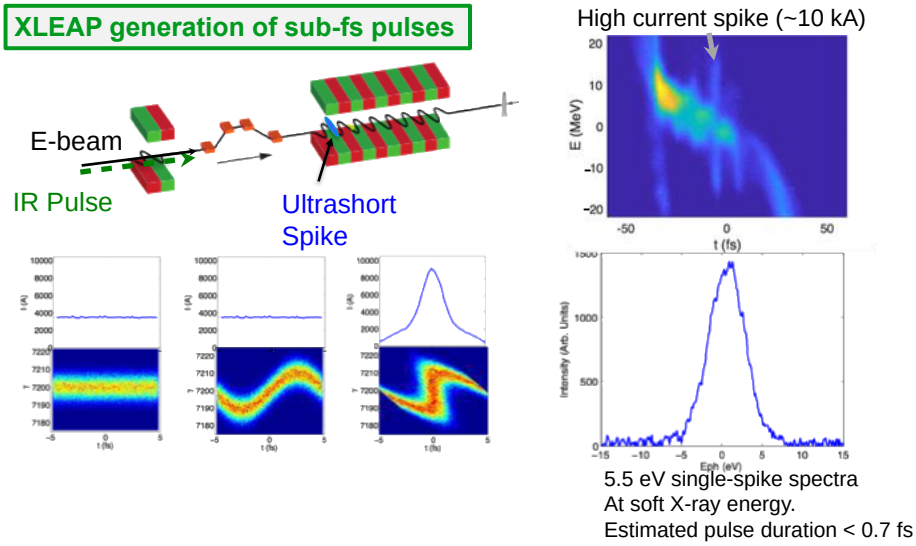
The performance of LCLS has been greatly extended since its initial operation, with an overview provided in a FAQ and performance table. LCLS can now provide ultrashort pulses (from ~200 attoseconds (as) to >100 femtoseconds (fs)), with unprecedented peak brightness, in SASE or seeded-mode operation, over an energy range from ~250 to ~25,000 eV, at 120 Hz. It regularly provides dual-pulses with relatively arbitrary separation in time (from fs to ~0.5us), with the option of dual color, and variable linear/circular polarization (for soft X-rays). Recent work has provided 4 independent pulses in a train, and increased the peak power to >300 GW.

Most recently:

- Progress has been made in verifying the microbunching instability origins of the pedestal in soft X-ray self-seeding through experimental measurements, and a new project has been launched to improve the laser heater and suppress the pedestal.

- Substantial focus has been applied to the generation of sub-femtosecond (attosecond) pulses, in response to the priority research opportunities identified in this domain. The XLEAP sub-femtosecond soft X-ray pulse generation program[8] has been commissioned and has already deployed new operating modes to LCLS users using self-modulation that scale well to high-repetition rate machines. This capability is needed to probe atom-specific core-to-valence transitions in molecules, and offers unprecedented coherent bandwidth to excite the entire valence spectrum, enabling control of the initial state of the system being studied. Current status is shown in Figure 12, with further development for LCLS-II now underway.

Figure 12 (anticlockwise from top left): schematic of the XLEAP concept; plots of the electron phase space; generation of high bandwidth single spike, consistent with <0.7fs.



[8] http://accelconf.web.cern.ch/AccelConf/ipac2017/papers/wepab118.pdf

- To complement this, a set of advanced beam configurations for attosecond pulses in the hard X-ray energy range have been demonstrated at LCLS using nonlinear compression[9] and the slotted foil[10], delivering isolated ~180 attosecond pulses (14.4 eV coherent bandwidth). Further work to increase the peak power of these pulses is underway, which would have a substantial impact on our ability to measure complex molecular structures prior to any meaningful atomic motion (or "damage").

- To improve our ability to measure such pulses, an algorithm to reconstruct the time structure from angular streaking has been developed[11]. A multinational team led by LCLS scientists has developed an "X-ray attoclock," capable of measuring the time and energy structure of XFEL pulses that are less than a femtosecond long. This tool will open up new opportunities for capturing unprecedented details of charge dynamics and molecular motion during the initiating events that drive chemical reactions[12].

- A high power mode has been developed using careful compensation of dispersion to reach ~300 GW peak power in a 10 fs hard X-ray pulse and a low photon energy bandwidth of 0.18% at 6.6 keV from a 6 kA peak current electron beam. This represents a factor-3 increase in peak power for LCLS[13]. A second high power, short pulse, mode was demonstrated using three-stage amplification for the soft X-ray FEL[14]. This keeps LCLS able to provide the highest power XFEL pulses available.

- The recently developed fresh-slice scheme has been highly successful in improving a wide range of operating modes, including short bunch, two-pulse, two-color, multi-stage (high power), among others. A new concept to make a match-based (rather than orbit-based) slice control reduces beam loss and will help extend fresh-slice modes to LCLS-II, and has now been demonstrated experimentally[15].

- The range of available multi-bunch, multi energy beams has been extended to > 100 ns bunch separation. A technique based on drive laser pulse splitting has been developed to deliver four bunches per pulse with several hundreds of picosecond separation. An extension of up to 16 bunches per pulse is planned.

[9] Huang et al., Phys. Rev. Lett. 119, 154801 (2017)
[10] Marinelli et al., Appl. Phys. Lett. 111, 151101 (2017)
[11] Li et al., Optics Express 26, 4531 (2018)
[12] Hartmann et al, Nature Photonics 12, 215 (2018)
[13] Guetg et al., Phys. Rev. Lett. 120, 014801 (2018)
[14] Lutman et al., Phys. Rev. Lett. 120, 264801 (2018)
[15] Chao et al., Phys. Rev. Lett 121, 064802 (2018)

Continuation of a wide-ranging R&D program remains a priority for LCLS operations, with the current focus being on readiness of new capabilities for LCLS-II. A list of R&D projects underway is shown in the following table. Examples of some major development activities are provided below.

Project List	Longitudinal Coherence	Advanced Modes	LCLS-II Readiness	Automation
Attosecond (XLEAP, Nonlinear)	X	X	X	
DELTA-II polarization control		X	X	
Seeding options	X		X	
Multi-bunch (4 to 16 pulses)		X		
Ultrafast kicker (for TXI)		X	X	
Laser heater shaping (for MBI)	X	X	X	
Dechirper phase space control	X		X	
Machine learning XTCAV analysis		X	X	X
THz development assessment		X	X	

5.1 DELTA Polarization Control

Many X-ray techniques, especially in the soft X-ray spectral range, require circularly polarized light. In the hard X-ray regime, quarter wave plates using 'off Bragg' diffraction have been very effective and this is the approach that will be taken for LCLS-II. Test of this approach to ensure its viability is important, and can made use of the existing LCLS facility to develop a robust optical configuration.

In the soft X-ray regime, one polarizing "Delta" undulator is currently installed at the end of the LCLS-I undulator line, providing ~200 uJ of circularly polarized X-rays at a very high degree of polarization (>99%). To reach saturation, it is currently estimated that three DELTA undulators will be required for the LCLS-II SXR line. The additional length of the DELTA will not only enhance the intensity of the circularly polarized X-rays but also strongly suppress harmonics on axis. These will be real advantages for many spectroscopies enabled by LCLS-II in the soft X-ray range. Adoption of this "afterburner" configuration will be a high priority for early operation of the upgraded facility.

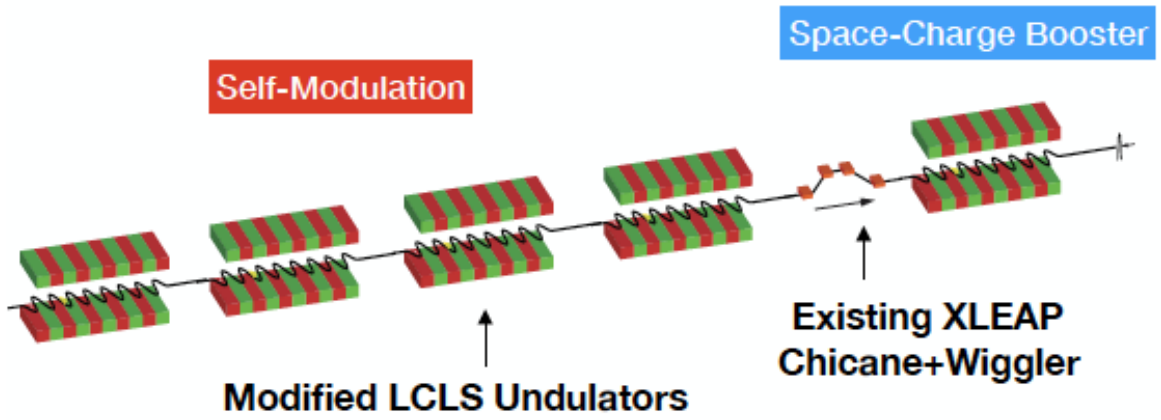
5.2 XLEAP-2 Attosecond Pulses

The XLEAP-II project will be based on the self-modulation variant of this scheme, due to its robustness (this uses the coherent undulator radiation emitted by the tail of the electron bunch to modulate the core of the bunch). This is to be installed in the soft X-ray line of LCLS-II. A schematic representation of the experimental setup is shown in Figure 13. The energy modulation will be induced in four 10-period wigglers. The wigglers will be built by modifying the existing LCLS undulators. The existing variable-gap XLEAP wiggler will be used as a space-charge booster after a magnetic chicane. Since this scheme is based on a passive modulator, it naturally scales to the high-repetition rates envisioned for the LCLS-II superconducting linac. The new setup will allow for an energy modulation up to 50 MeV

(peak to peak), which can be used for experiments that go well beyond the current attosecond FEL capabilities.

The large energy modulation generated by the XLEAP-II modulator can be used to generate two-color sub-fs pulses with a double-taper scheme. By dividing the undulator in two parts with two different tapers one can achieve the chirp/taper matching condition on two independent slices in the electron bunch, generating two sub-fs pulses with different photon energies. A small chicane in the middle of the undulator can be used to introduce a variable delay between the two pulses and use them for X-ray pump/X-ray probe experiments. The delay between the two pulses is locked to the resonant wavelength in the wiggler, allowing for sub-fs arrival time jitter between the two pulses.

Figure 13 Schematic of the XLEAP-II experimental setup.



5.3 Emittance improvements for LCLS-II-HE

At these high photon energies desired for LCLS-II-HE (>10 keV), the FEL performance is limited by the beam emittance in the undulator. Given the expected performance of the LCLS-II injector and the LCLS-II HXR undulator, a minimum beam energy of 8 GeV is required to generate 13 keV FEL photons. Improvements to the beam emittance would increase the attainable photon energy. This is illustrated in Figure 14 and Figure 15, which shows the sensitivity of the FEL to the electron beam energy for various values of the emittance. As an example, 13 keV photons could be generated with a 6.7 GeV beam if the beam emittance at the undulator could be reduced to 50% of the nominal LCLS-II emittance. Conversely, photon energies in the ~18 keV range could be produced by ~factor 3 improvements in emittance for an 8 GeV beam. Note that IMPACT simulations suggest that a factor of 2 reduction at the undulator would require at least a factor of 4 reduction at the injector due to space charge driven emittance dilution. A superconducting gun is being designed for the LCLS-II-HE

Project to deliver the performance required to extend the X-ray energy reach into the 15-20 keV range.

Figure 14 Analytical estimates of X-ray pulse generation with post- saturation taper are shown benchmarked against Genesis simulations of the LCLS-II 20 pC beam at various beam energies and with emittances scaled from the nominal bunch emittance.

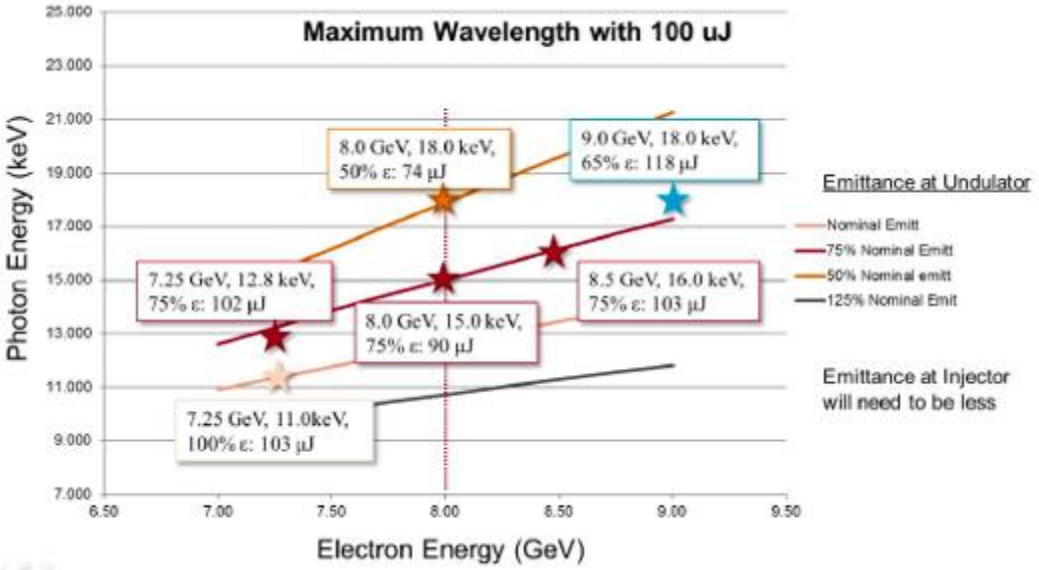
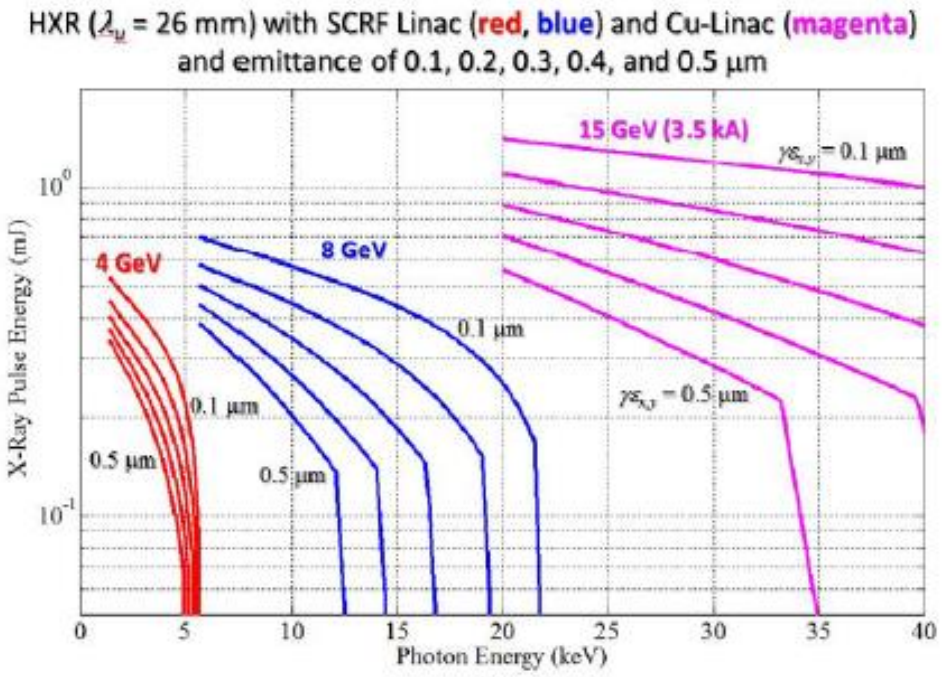


Figure 15 Estimates of spectral range based on ideal beams and the Ming-Xie approximations for the LCLS-II-HE SRF and the CuRF linacs.



5.4 Two-Pulse Two-Color

X-ray pump X-ray probe experiments open a very broad array of new scientific possibilities. Extensive machine development effort has recently been devoted to developing 2-pulse, 2-color techniques, with notable success.

An important advance beyond two pulses with the same energy and variable delay has been two pulses with different colors and variable delay. Accelerator-based techniques have addressed this within a finite parameter space: two colors with separations on order of 1 % have been demonstrated with delays ranging from 50 fs in soft X-rays to ~150 fs in hard X-rays. Also, two pulses separated by one or more RF buckets (up to >100ns separation) can be provided with different colors. For delays between the 100 fs scale and the few hundred ps to ns scale, X-ray "split and delay" optics solutions are used. See FAQ for latest information.

In addition, LCLS-II offers a unique potential: to bring the SXR and HXR undulator beams to one experiment with variable delay. The Cu linac and SCRF linac will have relative time jitter of ~100 fs, and with timing diagnostics this can provide high peak power pulses to the experiment again with very different photon energies. For high repetition rate operation, SLAC is developing a high-speed "kicker" magnet to send the LCLS-II beam to both undulators with a ~5ns delay at a rate of at least 10 kHz. This will provide coincident (or arbitrarily delayed) dual beams in the new TXI instrument. There are many variants of this scheme, for example using the SCRF linac for both the SXR and the HXR undulators and 'combining' the third harmonic of the SXR undualtor with a similar energy HXR pulse.

5.5 Soft X-ray Self-seeding

The system envisioned for self-seeding at high resolution is an upgraded version of the existing soft X-ray self-seeding scheme. The SXR self-seeding scheme is described extensively elsewhere (Proc. SPIE, vol. 8849, p. 88490A). It is a monochromator inserted into the undulator chain of LCLS that collects the SASE radiation produced by the first 8 undulators, selects a narrower bandwidth, and uses the resulting monochromatic beam to seed the electrons beam into the following undulator section.

The resolving power of the existing system is of the order of 5,000. Since the resolving power is dominated mainly by the diffraction limit contribution of the grating, e.g. the number of illuminated lines, one can increase the resolving power by moving the source point further upstream, e.g. by opening the gap of the last few undulators just before the monochromator.

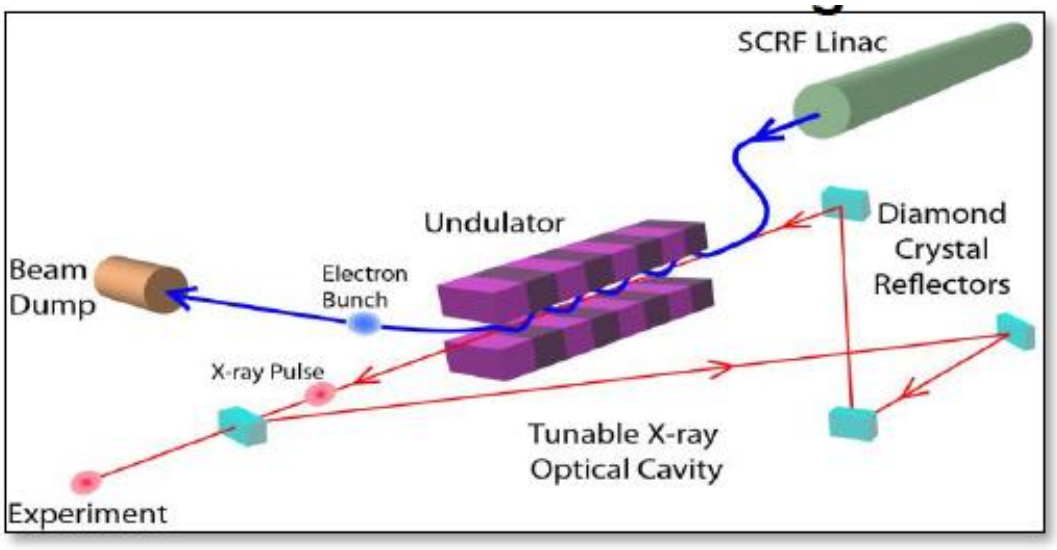
By opening the gap of the last undulators before the grating, one can increase the resolving power almost arbitrarily. However, one must also increase the power delivered to the grating, to preserve the power required to seed the electron beam. Within the projected damage limit of the grating, a resolving power of the order of 30,000 or slightly higher may be achievable.

5.6 RAFEL – Regenerative Amplifier FEL

Funded by DOE-BES, and in partnership with Argonne National Lab and RIKEN (Japan), SLAC is developing the design of a high gain X-ray cavity that would direct the X-ray beam around

a multi-pass round trip of the X-ray undulator, using a suitably high rep-rate beam[16]. This is predicted to result in an enhancement of the XFEL brightness by two to three orders of magnitude and high longitudinal coherence near the Fourier Transform limit, via the periodic FEL amplification and Bragg-monochromatization of the beam. See Figure 16. Output from the cavity could either be CW or pulsed (via Q-switching or microbunch rotation and off-axis lasing). RAFEL is similar to the X-ray FEL oscillator (XFELO) concept, but with reduced sensitivity to optics quality and alignment. Recent tests on a 21 meter cold-cavity at LCLS show suitable optics now exist, capable of sustaining the required efficiency.

Figure 16 Schematic of the RAFEL cavity.



5.7 Superconducting Undulators

Superconducting Undulators (SCU) provide the ability to reach higher X-ray energies, and provide much greater flexibility for advanced FEL modes (such as seeding) due to the shorter gain length. Development of SCUs is being undertaken with Argonne, with a proposal to build a 3-cryostat SCU prototype, and to test the performance at LCLS. Further investment is needed to develop advanced SCU performance, including planar and helical undulators, multiple undulator packages in the same cryostat, and new materials to enable higher fields and shorter periods.

[16] Z. Huang and R.D. Ruth, PRL 96, 144801 (2006); G. Marcus et al, doi:10.18429/JACoW-FEL2019-TUP032

6 SUMMARY AND NEXT STEPS

The purpose of this document is to lay out a suite of facility development priorities for the next 5+ years of LCLS operation in the context of the LCLS-II, LCLS-II-HE and MEC-U Projects, based on the assessments of science opportunities developed with the user community.

It is intended that this document (and subsequent updates) be used as the basis of a broad, ongoing consultation across the community, to help prioritize the suite of developments according to available funding and resource limitations, and to drive partnerships for delivery of capabilities that extend beyond the internal LCLS portfolio.

XFEL-based science requires the coupled development of the source, beamline diagnostics, experimental systems, data analysis, and a highly responsive operations environment. SLAC provides LCLS with a strong, co-located science program and world-leading technology capabilities. Stanford University adds a truly differentiating ability to identify and execute game-changing research through its faculty, joint institutes, student programs, and joint grants. Developments of the LCLS facility are thus able to take full advantage of the highly integrated nature of SLAC/Stanford, coupled to the extensive expertise across the DOE complex, and the breadth of the international user community.

A defining characteristic of LCLS (building from a long-standing approach at SSRL) has been the cultivation of a highly knowledgeable and service-oriented scientific and technical facility staff that have the skills, resources and capabilities to translate user science objectives into successful experiments. This requires deep engagement from the initiation of the proposal, through detailed planning and execution, coupled to investment in data workflows, use of start-2-end models to optimize performance, and offline lab capabilities to prepare samples, pump/probe systems, and novel diagnostics and detectors. The user community is strongly encouraged to make full use of the assembled expertise and knowledge within the facility staff to explore new opportunities and to help optimize their scientific plans.

Feedback to the LCLS Director on this document is welcomed.

7 Appendix – Scientific impact of LCLS

Here we provide a summary of some examples of the LCLS science impact, highlighting areas of science where bodies of LCLS research are judged to have a lasting impact by significantly advancing our understanding of important science questions, or in changing the direction and thinking in 10 selected scientific fields.

(i) High Intensity X-ray Matter Interaction

The advent of ultrafast X-ray lasers, with peak power densities of 10^{18}-10^{20} W/cm^2, has opened an entirely new regime in the science of light-matter interaction. Compared to the optical regime, intense X-ray fields are not well described by a quasi-static assumption (i.e. that the oscillating field is slow compared to the bound electron motion). Furthermore, the dominant X-ray interaction is with core and inner-shell electrons. Thus, intense X-ray pulses ionize atoms from the inside out, with the ultrafast relaxation dynamics that fill inner-shell vacancies (on time scales comparable to the X-ray pulse duration) playing a critical role. Seminal LCLS studies in low-Z elements (Ne) revealed that sequential single-photon ionization dominates in the 10^{18} W/cm^2 regime, producing a fully-stripped atom. The rapid photo-ejection of inner-shell electrons produces "hollow" atoms, double core-hole states, and X-ray-induced transparency.[1] In contrast, LCLS studies in high-Z elements revealed charge states far beyond the single-photon ionization limit, and first revealed the importance of transient resonance effects in enhancing X-ray multi-photon ionization.[2, 3] Most recently, LCLS studies of intense X-ray interaction with polyatomic molecules containing a high-Z atom show that the ionization physics is qualitatively different from that of isolated atoms or small molecules.[4] These results reveal a considerable enhancement in ionization driven by ultrafast charge transfer within the molecule. This charge transfer rapidly refills the core-hole states that are initially created in the heavy atom, providing further targets for inner-shell ionization and resulting in the emission of more than 50 electrons during a single X-ray pulse. LCLS experimental studies of high-intensity X-ray matter interactions have driven the development of advanced theoretical models with predictive capability for guiding future XFEL experiments.

(ii) Imaging Molecular Motion: Femtosecond X-ray Scattering of an Electrocyclic Chemical Reaction

Directly imaging the motions of atoms comprising a molecular reaction, on fundamental time scales, represents a qualitative advance in our ability to link molecular structure with function and reactivity. Ultrafast X-ray pulses from LCLS are indispensable for this purpose, and the ability to record "molecular movies" has opened an important new scientific frontier. Hallmark LCLS studies have focus on the evolving ring structure of 1,3-cyclohexadiene (CHD) as it opened up to form linear 1,3,5-hexatriene.[5] CHD is a prototypical example of an electrocyclic reaction; it plays an important role in the understanding of a large class of organic reactions; and features prominently in synthetic processes, photochemical switches, and natural product synthesis. Ring-shaped molecules are also abundant in biochemistry and form the basis for many drug compounds. Scattered X-rays from LCLS provided structural snapshots roughly every 25 fs during the 200 fs lifetime of the reaction. Comparison with theoretical models reveals the atomic structural details of four dominant reaction trajectories that are a direct consequence of the electronic excitation of the molecular π bonds. This time-resolved observation of an

evolving chemical reaction triggered by light paves the way for a wide range of X-ray studies examining gas phase chemistry and the structural dynamics associated with ultrafast chemical reactions.

(iii) Coupled Dynamics of Energy Flow: Molecular Structure and Charge

Understanding ultrafast nonadiabatic chemical dynamics during molecular photo-transformations is a grand challenge in chemistry because the close coupling of electronic and nuclear configurations mediates reactivity in ways that we are unable to accurately predict. For example, this knowledge gap limits our ability to predict and control the most fundamental properties of electronic excited states, including lifetime and charge-separation dynamics, and thus impedes our ability to develop robust, cost-effective complexes to convert and store solar energy. Polypyridyl iron complexes are model systems for investigating such phenomena, and LCLS studies of Fe^{II} polypyridyls have revealed important new insight to the excited-state charge and spin dynamics and their coupling to subtle atomic structural dynamics.[6-8] From these detailed multi-modal ultrafast X-ray studies (combining X-ray scattering and spectroscopy) it was possible to determine for the first time the excited state lifetimes, the role of coherent structural distortions, the locations of intersections between potential energy surfaces, and the transition probabilities between surfaces. Most significantly, coherent ultrafast electronic transitions are observed between the ^{3}MC and ^{5}MC states that directly coincide with coherent modulation of the ligand (Fe-N) bond distance. This work further demonstrates the power of ultrafast multi-modal XFEL methods for mapping in unprecedented detail the fundamental electronic excited state dynamics that underpin many useful light-triggered molecular phenomena involving earth-abundant 3*d* transition metal complexes.

(iv) Probing the Transition State Region of a Catalytic Reaction

Heterogeneous catalysis (e.g. hydrogenation and oxidation reactions) is central to the transformation of many chemicals on which modern society depends. However, significant knowledge gaps prevent us from fully exploiting and optimizing these processes based on prediction and directed design. Catalytically active species on surfaces are extremely difficult to characterize because of their high intrinsic reactivity and short lifetimes. Thus, despite theoretical predictions of various active intermediates, they have never been experimentally characterized. Transition states have been directly observed for the first time via element-specific time-resolved. X-ray absorption spectroscopy (XAS) using ultrafast pulses from LCLS.[9] Ultrafast optical pulses excite the motions of CO and O species on a Ruthenium surface, allowing the reactants to collide, and facilitating the formation of new electronic states associated with bond formation between CO and O with a distribution of bond lengths characteristic of the transition state. This first experimental characterization of the transition state in a surface catalytic reaction provides direct input into the theory of heterogeneous catalysis.

(v) Understanding the Function of a Natural Multi-electron Photo-catalyst

Sunlight-driven oxidation of water by photosystem II (PS-II) in plants, algae and cyanobacteria has generated most of the dioxygen in the atmosphere and is central to life on earth. The four-electron redox chemistry of water oxidation is accomplished by the Mn_4CaO_5 cluster in the oxygen-evolving complex (OEC) within PS-II. A detailed

understanding of the process of O-O bond formation and O_2 evolution in the cluster remains a grand science challenge, and will provide a foundation for the development of modified complexes and bio-inspired artificial photo-catalysts for converting sunlight to fuels. LCLS studies have provided the first damage-free structure determination of all the intermediate states (S_1, S_2, S_3) of the multi-electron catalysis cycle – at room temperature, and at unprecedented (<2 Å) resolution. [10-12] These results reveal important new structural details about substrate water binding and water oxidation mechanism, resolving key discrepancies between prior experiments, and ruling out suggested hypotheses for the catalysis cycle.

(vi) Damage-free Atomic Structures of Biological Membrane Proteins in Near-native Environments

Membrane proteins represent a dominant fraction of all proteins, they play a major role in biology, and are the target of more than 50% of all drugs. Nevertheless, understanding the structure and function of membrane proteins remains a major challenge because conventional X-ray crystallography and cryo-EM methods are often unable to resolve the structure of membrane proteins at suitable resolution. LCLS structural studies of membrane proteins are having a significant impact on the field of structural biology.[13-16] G-protein-coupled-receptors (GPCRs) are an important example: they represent the largest class of membrane proteins, and they play an essential role in mediating signaling in the cells of organisms (through coupling to G-proteins or arrestins). A hallmark study at LCLS resolved the structure of the membrane-protein rhodopsin (responsible for vision) bound to arrestin.[16] The unprecedented resolution of these measurements (~3.5 Å) revealed new details of the rhodopsin/arrestin complex that suggest a common model for the interaction between GPCRs and arrestins. Structural details of such a binary protein complex are essential to facilitate the development of drug compounds that might selectively modulate GPCR signaling pathways for the treatment of diseases. More broadly, the successful development at LCLS of powerful new methods of serial femtosecond crystallography (SFX) and "diffract-before-destroy" have opened new areas of structural biology by enabling damage-free structure determination of membrane proteins, metallo-enzymes, and small bio-crystals,[17, 18] in near-native environments, and have triggered a renaissance in conventional protein crystallography methods.[19, 20]

(vii) Understanding Biological Function: Capturing Structural Dynamics of Bio-molecules in Action

For decades, biological function has been inferred from static structures obtained primarily from crystallographic measurements. However, functioning biological systems are not static, and probes of structural dynamics are essential to reach a deeper understanding the function of macromolecular complexes whose structures change over a range of time and length scales, often in response to changes in local environment (temperature, pH) or interaction with small molecules or other external stimuli. LCLS studies of structural dynamics of biological macromolecules in near-native environments represent a qualitative advance in our understanding of biological function. [21-23] LCLS studies of light-sensitive complexes such as bacteriorhodopsin and photoactive yellow protein, have created molecular movies of the fundamental structural dynamics with unprecedented temporal and spatial resolution. Beyond the limited class of naturally photo-active

proteins, LCLS has pioneered structural dynamics studies of functioning macromolecular machines such as riboswitches, which are structural elements of messenger RNA (mRNA) that are central in genetic regulation. The function of ribosomes, and signaling of the downstream expression platform, is mediated by conformational changes in response to the binding of a ligand to a specific domain. LCLS studies captured the dynamic structural response of riboswitches to the binding of a ligand for the first time, [24] by using ultra-small riboswitch crystals, timing the diffusion of a ligand to initiate the reaction, and exploiting SFX to capture the structural intermediates. These experiments determined four transient structures that support a reaction mechanism model with at least four states and illustrate the structural basis for signal transmission. These results further demonstrate the potential of "mix-and-inject" time-resolved serial crystallography to study biochemically important interactions between biomacromolecules and ligands, including those involving large conformational changes.

(viii) Coherent Control of Complex Materials

Coherent light-matter interactions represent a powerful new approach for controlling emergent material properties and for creating novel metastable (nonequilibrium) material phases. Especially important is the ability to drive a material directly on the low energy scales at which fundamental excitations are found, e.g. phonons, plasmons, magnons or other collective modes. Seminal LCLS studies in this area combined coherent THz excitation for tailored manipulation of complex materials with ultrafast X-rays for quantitative characterization of new metastable material phases. One important example is the coherent control of multiferroic materials. Interest in this area stems from potential applications for controlling magnetic order by electric fields. However, the underlying physics, strength, and ultimate speed of magnetoelectric coupling present a significant knowledge gap. LCLS experiments using ultrafast resonant X-ray diffraction revealed the spin dynamics in multiferroic $TbMnO_3$ driven by an intense few-cycle terahertz (THz) light pulse tuned to resonance with an electromagnon mode. [25] The results show that atomic-scale magnetic structures can be directly manipulated with a coherent THz electric field on a sub-picosecond time scale. Projections indicate that complete coherent magnetic switching should be achievable with modest scaling of the THz field. A second important example is the coherent excitation of selective lattice modes for manipulating high-T_c superconductivity. LCLS experiments resonantly excited Cu-O stretch modes in the cuprate superconductor $YBa_2Cu_3O_{6+x}$ (sample above the transition temperature T_c). Careful comparison of X-ray scattering with DFT calculations showed evidence of phonon rectification (ionic Raman scattering) manifest as a dilation of $Cu\text{-}O_2$ distances within each bilayer, a contraction of these atomic distances between bilayers, and subtle buckling of the O-Cu-O bonds.[26] These structural changes are associated with trastic changes in the electronic structure, including a remarkable enhancement of superconductivity above T_c.

(ix) New Insight into Correlated Phenomena in Quantum Materials

A hallmark of quantum materials is the dominant influence of quantum-level coupling between charge, spin, orbital, and lattice modes in determining the macroscopic material properties. LCLS studies using ultrafast X-rays have significantly advanced our understanding of quantum materials by enabling the observation and separation of coupled interactions in the time domain. One important example is in understanding the

universal existence of charge density wave (CDW) correlations in high-T_c cuprates and the role of CDW in establishing superconductivity. LCLS studies revealed for the first time the structure of the CDW phase existing in the same phase-space with superconductivity in the high-T_c cuprate $YBa_2Cu_3O_{6.67}$.[27] Time-resolved X-ray scattering measurements in combination with a transient high magnetic field (28 Tesla) to suppress superconductivity, revealed an unexpected three-dimensionally ordered CDW phase. This discovery of the field-induced CDW provides long-sought information to bridge the gap in cuprate phenomenology, which is critical to uncover the mechanism of high-T_c superconductivity. A second important example is in understanding the role of electron-phonon (e-ph) coupling in high-T_c superconductors. Multi-modal LCLS studies that correlated time-resolved ARPES measurements of electronic structure, with time-resolve X-ray scattering measurements of atomic displacements, provided the first direct measure of e-ph coupling strength in the pnictide superconductor FeSe – purely from experiment.[28] Such direct measurements provide a critical test of theoretical predictions, and in the case of FeSe, the e-ph coupling is found to be nearly 10-fold larger than predictions. This has prompted a reassessment of the significance of e-ph coupling in facilitating superconductivity in pnictide compounds. More broadly, this approach provides a powerful new means to directly determine the role of e-ph coupling in creating the emergent properties of a broad range of quantum materials.

(x) Understanding the Creation of New Metastable Material Phases in Extreme Environments

LCLS provides important new insight, at the atomic scale, of non-equilibrium states associated with material phase transitions in extreme environments. The most important open scientific questions relate to the physical and structural properties of extreme states of matter that can be produced only in dynamic transient experiments that are uniquely accessibly using ultrafast X-ray methods. Such information is of central importance to fundamental research in numerous fields, including high energy density science, geoscience, planetary science, laboratory astrophysics, relativistic laser plasmas, and fusion research. One important example is in understanding the novel chemistry that occurs at extreme pressures and temperatures that are characteristic of the interior of icy giant planets in our solar system. Recent LCLS experiments investigated the atomic-scale structural dynamics of hydrocarbons under conditions comparable to 10,000 km below the surface of Neptune (~150 GPa, ~5,000 K), transiently created via a two-step laser shock front. In-situ ultrafast X-ray scattering measurements, revealed a phase separation of carbon and hydrogen, from the covalent bonds of the polymer chain molecules, and the subsequent formation of nano-diamonds.[29] These results are the first unambiguous observation of high-pressure diamond formation from hydrocarbons. They further demonstrate the necessity of high-pressure (in addition to temperature) to initiate the carbon-hydrogen separation, and imply that diamond precipitation requires ~10-fold higher pressure than previously indicated by static compression experiments. These results highlight the importance of novel chemistry for an accurate understanding of mass-radius relationships and material constituent constraints that underpin the classification of exoplanets. Additional examples of LCLS research that has significantly advanced our understanding of materials in extreme environments include atomic-scale studies of the evolution of microstructure associated with elastic and plastic deformation in Cu[30] and Ta

[31] under transient high-pressures. Importantly, quantitative studies enabled by LCLS ultrafast X-rays enable direct comparison to theoretical models and simulations.

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