LCLS STRATEGIC FACILITY DEVELOPMENT PLAN

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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 1450 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 638 full-scale user experiments (2009-2019), plus over 270 in-house experiments and 25 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, 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, in SASE or seeded-mode operation, over an energy range from ~250 to ~25,000 eV, at 120 Hz (the high end of this energy range will doubled during the course of 2020 from its prior limit of 13,000 eV). LCLS 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. Recent work has increased the peak power to over 300 GW 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 current suite of 7 instruments have been published, with updates provided online. Their scientific output can be viewed 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 will be 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).

2 Typically 10^{12} ph/s/m^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. 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 project will also double the maximum X-ray energy the existing accelerator can produce, to reach approximately 25 keV (and substantially higher using the third harmonic), providing capabilities unmatched anywhere in the world.

Beyond this, an extension to higher X-ray energy at high repetition rate has long been requested by the LCLS user community. In response, SLAC has developed 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 at least 13 keV and potentially up to 20 keV. The LCLS-II-HE proposal achieved Critical Decision 1 (approval of the selected approach) in September 2018, following endorsement by BESAC and strong community support expressed at a number of workshops. Ongoing work by SLAC is centered on refining the expected “first experiments” for LCLS-II-HE via a series of workshops\(^5\), which are informing the detailed design and instrument requirements.

In parallel, a major upgrade to the MEC instrument is being developed with the DOE Office of Fusion Energy Sciences, to significantly increase the peak power and energy of the pump lasers, with corresponding development of the experimental systems. This is described further in section 4.5.3.

More broadly, a widespread series of developments are underway to prepare for early science on LCLS-II, LCLS-II-HE and MEC, with a view to ensure ongoing scientific impact and development of the field of ultrafast X-ray science. Our plans integrate theory, simulation and experiments; pursue focused R&D in critical technologies; integrate 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 in an organized series of events leading up to first experiments to exploit the new facility capabilities.

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.

It is important to note that the range of developments presented in this document will be selected and prioritized according to the available funding, schedule constraints, and resource availability over the next few years. Not all options will be able to be implemented, hence the need for ongoing strategic prioritization and consultation.

\(^5\) See, for example: https://portal.slac.stanford.edu/sites/conf_public/lclsiihe2018/Pages/default.aspx
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) compared to other XFEL and storage ring facilities (e.g. the use of continuous pulse trains; the use of independently tunable XFEL beams in the soft-, tender, or hard- X-ray regimes; the ability to probe in the ultrafast domain; and the use of X-ray beams of unprecedented average power and spectral brightness).

4. Use the unique in-house knowledge gained from LCLS experiments to evaluate future facility configurations, instrument designs, and experimental aspirations, based on real-world experience.

5. Increase community throughput (via new multiplexing schemes; dedicated end-stations; revised operational models, etc)

6. Ensure any near-term investment in LCLS is consistent with subsequent use in the LCLS-II (-HE) context.

7. Develop systems that are highly robust and configured to run without staff intervention in 'standard modes', in order to enable greater control of experiments by the users, and to provide scalable, sustainable, and scientifically creative roles for LCLS staff.

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 for LCLS-II and LCLS-II-HE;
- **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](mailto:).*
2 SCIENTIFIC DRIVERS

2.1 Example 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

2.1.1 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

2.1.2 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

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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.

**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

### 2.1.3 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

### 2.1.4 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

2.1.5 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

2.2 Example 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 in the latest report from BESAC\(^3\), 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:
i. 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

ii. 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
iii. **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

iv. **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

v. 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 ferroelectricity 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

vi. 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
vii. **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 only 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 is currently 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 take advantage of the new configuration.
- Extends the operating range of the facility from its current limit of ~12.8 keV X-rays to ~25 keV using the existing 120 Hz accelerator.
- Provides for a factor ~2 increase in experimental capacity, addressing a principal limiting factor of LCLS (wherein only ~20-25% of proposals can be scheduled).

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 and a summary of the science opportunities for the next decade.

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 1). The predicted performance of the resultant beam is shown in Figures 2, 3.

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Figure 1 Principal partners in the LCLS-II Project

Figure 2 Performance of the LCLS-II upgrade, comparing current performance (in blue) with the enhanced capabilities (in red; with the 3rd harmonic shown as a dotted line)

Figure 3 Performance table of projected capabilities for LCLS-II
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 includes a “cross-over beamline” to route the copper linac to the soft X-ray undulator, such that both linacs will be able to feed both undulators. Operation will restart with the 120 Hz copper linac in early 2020, and transition to incorporate the LCLS-II beam in late 2021 according to current expectations (Figure 4).

Figure 4 Long range LCLS operations schedule.

A phased increase in capability (for the instruments as well as for the new beams) will take place over the 2021-2023 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.

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.\(^9\)

The top-level design principles being applied to the new instrument area are as follows:

i. Maximize the time spent on scientific data production.
   - Automated alignment of the beam and end-stations where possible.
   - Quantitative goals for the time and effort required for changes in configuration (aiming for changes in “minutes & hours”, not “days and weeks”).

ii. Deliver a design that allows the role of Instrument Scientists and design engineers to focus on the innovative development of their scientific/technical field, so that they can provide LCLS with a greater competitive edge in science delivery.

iii. Use the high rep-rate beam as the primary driver for experimental prioritization and facility design.

iv. Ensure integration of the instrument designs with the required next-generation detectors, pump/probe requirements, in-situ beam diagnostics, etc.

v. An explicit path to move from "early science" to "flagship" experimental goals.

vi. Diagonalize the science objectives at a facility level – not at an instrument level – to allow flexibility in design response.

vii. Assume LCLS-II-HE is implemented to come online roughly mid-decade.

The solution accommodates up to 3 X-ray branch lines from the soft X-ray undulator (SXU), and preserves the current suite of five hard X-ray instruments (XPP, XCS, MFX, CXI, MEC).

The SXU branch lines are created via reflective and grating optics located in the front end enclosure (FEE). They distribute the X-ray beam to 3 hutchs (TMO, RIXS and TXI). As part of this, a new tender X-ray branch line is created that operates on the hard X-ray undulator (HXU) and also delivers beam to TXI to create a ‘dual beam’ instrument area.

This solution involves substantial modifications to the Near Experimental Hall (NEH) to allow the construction of:

- A new X-ray hutch on the NEH first (upper) floor level. This single integrated hutch could potentially be split into two in later years, and accommodates 3 separate endstations (qRIXS, ChemRIXS, and an open port for user-supplied systems).
- Modified X-ray hutchs on the NEH sub-basement level (TMO and TXI, replacing AMO and SXR)
- A major new optical laser laboratory on the upper floor
- Dedicated control rooms for each instrument
- An expanded data/server capability

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\(^9\) See: [https://lcls.slac.stanford.edu/instruments/l2si](https://lcls.slac.stanford.edu/instruments/l2si)
A suite of “First Experiments\textsuperscript{10}” for LCLS-II was developed to drive the design and implementation of these instruments on the basis of the science opportunities described in Section 2. Feedback on these is encouraged.

An overview of the resultant instrument layout is provided in Figure 5, and schematic models of the new instruments are displayed in Figures 6, 7 and 8.

**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 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 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 and high resolution photon spectroscopy. It will host two endstations in tandem, plus an open port for future expansion or user-supplied equipment. The first endstation (qRIXS) is designed for high resolution, momentum resolved resonant inelastic x-ray scattering (RIXS) to study bosonic excitations in solid state samples. The qRIXS endstation 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 designed to accommodate X-ray Photon Correlation Spectroscopy (XPCS) experiments.

The second endstation (ChemRIXS) is designed with an emphasis on soft x-ray spectroscopy experiments on liquid samples, with two x-ray emission spectrometers; a conventional varied line spacing spectrometer and a transition edge sensor (TES) spectrometer.

**Figure 5** Revised instrument layout, as fed from the 2 new undulators from the LCLS-II Project, covering the soft X-ray (SXU) and tender/hard X-ray (HXU) regime.

**Figure 6** Schematic model of NEH1.1 (TMO)

<table>
<thead>
<tr>
<th>Endstation</th>
<th>Science</th>
<th>Method</th>
<th>XFEL</th>
</tr>
</thead>
<tbody>
<tr>
<td>IP1 (VMI/cookiebox)</td>
<td>High field physics, Photophysics &amp; photochemistry</td>
<td>Ion/Electron Spectroscopy</td>
<td>250 – 2000 keV</td>
</tr>
<tr>
<td>IP2 (DREAM)</td>
<td>Fundamental excited state dynamics, Charge and energy transfer</td>
<td>Dynamic molecular reaction microscope</td>
<td>250 – 1500 eV, &gt;= 100kHz</td>
</tr>
</tbody>
</table>
**Figure 7** Schematic model of NEH2.2 (RIXS)

**Figure 8** Schematic model of dual-beam NEH1.2 (TXI)
The nominal phased delivery schedule for the 4 new instrument areas (dependent on funding and resource availability) is currently as shown in Figure 9.

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, unique in the world, delivering ultrafast atomic resolution at high average power. The project is a natural extension to LCLS-II, adding known technology and using existing infrastructure. It will extend operation of the high-repetition-rate beam into the critically important “hard X-ray” regime (>5 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, and Critical Decision 1 in September 2018.

The energy reach of LCLS-II-HE (stretching from 5 keV to at least 13 keV and likely 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 10.

**Figure 10** 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 anticipated 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 proposed or envisioned 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.

- Combine **three independent accelerators** into a single facility, representing an unprecedented level of flexibility for the user community (a new 8 GeV superconducting linac; a separately tunable 3.6 GeV line for the LCLS-II instruments; and the existing 15 GeV Cu-linac). **No other facility in the world will have this capability.**

To achieve this, the LCLS-II-HE project will add 19 cryomodules of the type already being manufactured for LCLS-II, 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.

Therefore, this solution represents a low-risk path with dramatic scientific impact. It will incorporate a linac bypass line to allow simultaneous operation of the soft X-ray and hard X-ray undulator sources with optimum electron beam energies, coupled with 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:

1. **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. This is illustrated in Figure 11.
Figure 11 The spectral region above 5 keV (the design limit of LCLS-II) is a critical area for many types of measurements. Access to Ångström wavelength (~12.4 keV) X-rays is a major enabling step for atomic-scale studies, while the ability to probe Earth-abundant elements and access experimental regimes central to biological structure determination and quantum materials studies will provide a fundamentally new capability for discovery science.

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. The nominal plans are to introduce a new inelastic X-ray scattering (DXS) instrument to augment XCS, and to enable 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 12. However, these plans are the subject of active discussion, and user feedback is sought – both directly and via the various workshops.

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11 See, for example: [https://portal.slac.stanford.edu/sites/conf_public/lclsiihe2018/Pages/default.aspx](https://portal.slac.stanford.edu/sites/conf_public/lclsiihe2018/Pages/default.aspx)
4 PHOTON SYSTEM DEVELOPMENT

4.1 Scientific Technique Development for LCLS-II

The distinguishing capabilities of the LCLS-II upgrade (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 and assemblies in natural environments*
- Stimulated X-ray Raman spectroscopy – *ultrafast charge-transfer dynamics*
- Core-hole correlation spectroscopy – *quantum coupling of valence charge states*

<table>
<thead>
<tr>
<th>Instr.</th>
<th>Upgrade Plan</th>
<th>Science Opportunities</th>
</tr>
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</table>
| XPP    | New detector, upgraded optics | • Understand coupled dynamics of molecular structure and charge & their role in energy flow  
• Characterize materials heterogeneity, fluctuations & link to function |
| DXS    | Dynamic X-ray scattering (High resolution IXS and XPCS capability) | • Map collective excitations & understand their relation to emergent phenomena in complex materials  
• Characterize materials heterogeneity, fluctuations & link to function |
| CXI    | New optics & detector, enhanced DAQ | • Reveal the role of structural dynamics in biological function  
• Catalysis: Reveal the correlation between chemical reactivity & structural dynamics |
| NXI    | New optics & detector, enhanced DAQ | • Reveal the role of structural dynamics in biological function  
• High x-ray peak power capabilities |
| MFX    | New detector, enhanced DAQ | • Reveal the role of structural dynamics in biological function |
• Single-particle imaging – *toward atomic resolution*
• Dynamic multi-view tomography – *chemical mapping of reactive flows*

4.1.1 **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 NEH 1.1 and NEH 1.2 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 µm to ~50 µm (depending on sample concentration etc.)
- Photon energy range: both tender X-rays (2-8 keV\(^{12}\)) 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.

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.

4.1.2 **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

\(^{12}\) Experiments at high repetition rate with energy > 5keV will require LCLS-II-HE
cover only a small fraction of the $4\pi$ 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) may exploit 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 µm focus (adjustable) to optimize X-ray nonlinearity (possible line focus for spatial resolution)

The above parameters can be met at instruments NEH 1.1 and NEH 1.2, 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 µm focus (\(~10^{16}\) W/cm$^2$ at 500 eV).
- ~1 µm focus (adjustable) to optimize X-ray nonlinearity

The above parameters can be met at instruments NEH 1.1 and NEH 1.2. 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 $\Delta E \sim 280$ eV (carbon edge)
- Tender X-ray energies 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
- $\sim 1 \mu$m focus (adjustable) to optimize X-ray nonlinearity (possible line focus for spatial resolution)

The above parameters may be met at NEH 1.2 using 3 keV photons from the HXU driven by the SCRF and 2.7 keV from the SXU (e.g. $3\omega \times 900$ eV) similarly driven by a second delayed bunch from the SCRF. Compensation of the delay ($\sim 10$ ns inter-bunch spacing) may be accomplished via Bragg optics. NEH 1.2 provides access for optimized (roll-up) endstations to detect and develop XCARS for a range of science applications.

### 4.1.3 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 $\pm 35$ fs from FEL
- $<10$ fs pulse duration (comparable to the Auger lifetime), ideally close to transform limit
- 0.1-1 $\mu$m focus to enhance X-ray nonlinearity

The above parameters can be met at instruments NEH 1.1 (or NEH 1.2 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).

### 4.1.4 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\(^\text{13}\). 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)

LCLS-II instruments NEH 1.1 and NEH 1.2 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 NEH 1.1 (0.25-3 keV) or NEH 1.2 (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-$q$ 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 NEH 1.1 and NEH 1.2 instruments.

### 4.1.5 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.

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
  • Optical instrumentation for generating and manipulating multiple beams of soft or tender X-rays
  • Fast 2D detectors in the soft and tender X-ray range
  • ~25 µm pixels, 1k×1k, $\sim 10^2$-$10^3$ ph/pixel/pulse
4.2 X-ray Optics Development

4.2.1 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.

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 are 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 13). 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 thermos-mechanical fatigue (with \( \sim 10^{13} \) pulses per operating-year). Evidence from optical irradiation shows that the acceptable absorbed fluence can drop substantially with time.

**Figure 13** Schematic of the REAL optics assembly for a \( \sim 1 \)m long X-ray distribution mirror, combining a cooling system with resistive heating elements to maintain the figure.
4.2.2 Variable resolution beamline for RIXS experiments

A RIXS endstation is 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, namely 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 initial 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 14).

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.

**Figure 14** 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.

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.
**RIXS spectrometer**

The RIXS 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 15).

**Figure 15** Scheme of the proposed 6 m RIXS spectrometer

![Diagram of the 6 m RIXS spectrometer](image)

The expected resolving power is calculated considering a 10µm vertical spot on the sample and 10µm spatial resolution for the detector (Figure 16). 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.

**Figure 16** Expected resolving power for the 1800 l/mm grating. A 10 µm spot size and 10 µm spatial resolution for the detector are considered in the simulation.

![Graph showing resolving power vs photon energy](image)
4.2.3 Split and Delay X-ray optical system

A significant advance for near-equilibrium dynamics is a newly commissioned X-ray optical system for generating two X-ray pulses, introducing a controllable delay with respect to each other (from -10 ps to +500 ps with femtosecond precision), 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.

This system (shown in Figure 17) 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 17 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 18). 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 18 Schematic and photo of a new, compact split and delay system with 4 channel-cuts.
4.3 Advanced Detector Systems

This section provides a very brief overview of the LCLS Detector Strategy. A more comprehensive detector strategy has been detailed separately.

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

A very-fast CCD is being developed in collaboration with LBNL. This MPixel-scale area detector will provide high quantum efficiency in the 250-1200 eV energy range with very low noise and kHz frame rate. To achieve this readout speed, a fully column-parallel CCD together with its corresponding readout and data acquisition system has to be developed. The main difference with conventional CCDs consists in eliminating completely the output transistors (typical source-follower configuration) to directly couple the charge into a low impedance preamplifier, implemented in a readout chip. Each readout channel is then directly wire-bonded to a CCD column. To achieve high quantum efficiency at low photon energy the detector entrance window has to be very thin. A low temperature contact created by molecular beam epitaxy (MBE) will be used to provide good soft X-ray quantum efficiency.

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. This 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 also 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).

Transition Edge Sensor (TES) micro-calorimeters can provide a unique combination of spectral resolution and efficiency. Superconducting detectors are intrinsically capable of very high energy resolution. The challenges with such detectors have been the small number of pixels possible in an array, the low readout rate, a low fill factor (active/total area), and the need for sub-Kelvin cooling. Recent advances in each of these aspects makes TES spectrometers a very interesting alternative to standard grating spectrometers. Ongoing
R&D aims to provide spectrometers for pulsed sources with >10kHz frame rates and ~0.5 eV resolution at 1 keV.

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 ‘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 detector.

The current nominal timeline for detector deployment is as follows:

<table>
<thead>
<tr>
<th>Instrument</th>
<th>Detector Needs</th>
<th>Deployed</th>
<th>Detector</th>
</tr>
</thead>
<tbody>
<tr>
<td>HXR (XPR)</td>
<td>Hard X-ray Area Arrays (120 Hz)</td>
<td>3/2020</td>
<td>ePix100, ePix10k, Jungfrau 4M, Rayonix 3.6M</td>
</tr>
<tr>
<td>NEH 1.1 (LAMP/VMI)</td>
<td>2D ToF Charged Particle (1 MHz)</td>
<td>4/2020</td>
<td>ToF, MCP/hex.anode</td>
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<tr>
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<td>2D High Spatial Resolution (5 μm)</td>
<td>6/2020</td>
<td>STARIXS-CCD</td>
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<tr>
<td>NEH 1.1 (DREAM)</td>
<td>2D ToF Charged Particle (1 MHz)</td>
<td>4/2021</td>
<td>ToF, CookieBox</td>
</tr>
<tr>
<td>NEH 2.2 (GRIKXS)</td>
<td>2D High Spatial Resolution (5 μm)</td>
<td>1/2022</td>
<td>STARIXS-CCD</td>
</tr>
<tr>
<td>NEH 1.2 (TXI)</td>
<td>Tender X-ray Area (≥ 5 kHz, 2.4 MPx)</td>
<td>1/2023</td>
<td>ePixHR</td>
</tr>
</tbody>
</table>

### 4.4 Data Systems Strategy

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 selection of 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.


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 18 months, 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 $10M ExaFEL grant, 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 19.

**Figure 19** A 4-step description of the data flow for LCLS-II and HE.
4.5 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.

4.5.1 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 process.

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 ~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 20, 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.
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 for LCLS-II experiments 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.

### 4.5.2 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\(^\text{14}\).

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.

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For LCLS-II an upgraded version of the current RF timing system (based on RF distribution over stabilized coaxial cable) is planned, employing 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\(^\text{15}\). 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\(^\text{16,17}\). 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.

### 4.5.3 Lasers for High Energy Density Science at the MEC Instrument

The field of High Energy Density (HED) science performed at the Matter in Extreme Conditions (MEC) instrument has laser requirements for high pulse energy and high peak power. The current MEC laser capability is comprised of two laser systems: an Nd:glass, high-energy, temporally-shaped, nanosecond laser system that fires at one shot per few minutes with a pulse energy of 60 J / 10ns; and a Ti:sapphire, high intensity, femtosecond laser system capable of producing 30 TW peak power at 5 Hz. There are multiple, overlapping pathways for expanding capabilities in these laser systems.

LCLS has engaged in extensive community outreach through workshops and direct interactions to help articulate the case for different types of upgrade, with a preferred path emerging to combine 3 sets of beams: high-energy short pulse beam (~150J in ~150fs, running at up to 10 Hz); kilojoule-scale shaped long-pulse beam, and the LCLS X-ray beam.

High-peak-power and high energy laser facilities provide a versatile set of tools for creating plasmas under extreme conditions. Focused to a small spot, ultrashort laser pulses with 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.

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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. 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 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 Zero) for such a facility was approved by DOE Office of Fusion Energy Sciences (FES) in January 2019.

We propose to reconfigure and upgrade the MEC instrument at LCLS by excavating a new experimental hall, as shown in Figure 21 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.

Figure 21 Schematic of one design concept for the upgraded MEC instrument, showing two target chambers in a new experimental cavern to the east of the Far Experimental Hall.
4.5.4 Upgrades of Pump/Probe Laser Systems

The Ti:sapphire laser systems in the NEH are mature, stable, and nearly identical between AMO, SXR, and XPP. The core NEH laser systems are stable and require 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 on the general use table 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 hutchs. 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.

Further planned 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 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.

Space for R&D efforts on optical sources, such as cryo-cooled THz, <10 fs sources, UV and HHG sources, etc. is now available in two LCLS laboratories, housing systems identical to those used for hutch operations. These labs will also provide laser light to users to test samples, detectors and measurement schemes prior to beamtime. The new Arrillaga Science Center laboratory building at SLAC is scheduled for beneficial occupancy in December 2018, providing space for laser laboratories to further develop sources and experimental capabilities, working closely with researchers from the PULSE Institute and external groups.

Continued enhancement of pump-probe laser capabilities was completed in 2017 in the Far Experimental Hall, with the installation of a new 30fs, 6mJ laser system in XCS. This system provides ultrafast optical pump pulses at wavelengths spanning from the visible to near-infrared, providing similar capabilities to those of XPP in the FEH.

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 will be added to the existing femtosecond Ti:Sapphire laser system, providing ~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. It is expected that similar ultrafast laser systems to those in CXI will also be implemented in the hutch, enabling time-resolved experiments on this instrument.
5 ACCELERATOR SYSTEMS DEVELOPMENT

5.1 Accelerator R&D

The performance of LCLS has been greatly extended since its initial operation. 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 over 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\(^{18}\) 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 22, with further development for LCLS-II now underway.

Figure 22 (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.

• 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\textsuperscript{19} and the slotted foil\textsuperscript{20}, delivering isolated $\sim$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\textsuperscript{21}. 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\textsuperscript{22}.

• A high power mode has been developed using careful compensation of dispersion to reach $\sim$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\textsuperscript{23}. A second high power, short pulse, mode was demonstrated using three-stage amplification for the soft X-ray FEL\textsuperscript{24}. 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\textsuperscript{25}.

• 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.

\textsuperscript{19} Huang et al., Phys. Rev. Lett. 119, 154801 (2017)
\textsuperscript{20} Marinelli et al., Appl. Phys. Lett. 111, 151101 (2017)
\textsuperscript{21} Li et al., Optics Express 26, 4531 (2018)
\textsuperscript{22} Hartmann et al, Nature Photonics 12, 215 (2018)
\textsuperscript{23} Guetg et al., Phys. Rev. Lett. 120, 014801 (2018)
\textsuperscript{24} Lutman et al., Phys. Rev. Lett. 120, 264801 (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.

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<tr>
<th>Project List</th>
<th>Longitudinal Coherence</th>
<th>Advanced Modes</th>
<th>LCLS-II Readiness</th>
<th>Automation</th>
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</thead>
<tbody>
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<td>Attosecond (XLEAP, Nonlinear)</td>
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### 5.1.1 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 be 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 μJ 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.

### 5.1.2 XLEAP-2, enabling attosecond science for LCLS-II

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 23. The energy modulation will be induced in four 10-period wigglers. The wigglers will be built by modifying the existing LCS 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 23** Schematic of the XLEAP-II experimental setup.

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### 5.1.3 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 24 and Figure 25, 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. How much the injector emittance could be improved is not clear, and requires active R&D.
Figure 24 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.

![Graph showing X-ray pulse generation](image)

Figure 25 Estimates of spectral range based on ideal beams and the Ming-Xie approximations for the LCLS-II-HE SRF and the CuRF linacs.

![Graph showing spectral range](image)

5.1.4 X-ray pump Two-Pulse (variable delay)

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. A Summary of 2-pulse, 2-color modes of operation is shown in the table below. See FAQ for latest information.

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 currently developing a high-speed “kicker” magnet to send the LCLS-II beam to both undulators with a ~5ns delay. This would be compensated for by X-ray optics, to provide coincident (or arbitrarily delayed) 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 ‘combine’ the third harmonic of the SXR undulator with a similar energy HXR undulator pulse at repetition rates in the 100kHz scale.

5.1.5 High resolution 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.

5.1.6 High Peak Power and Extended Energy for LCLS-II

Driving the LCLS-II soft X-ray undulator with the high-energy electron beam from the warm copper SLAC linac would generate pulses with very high energy (>5 mJ) in the tender (2-5 keV) X-ray regime at 120 Hz. The peak power levels from this source are calculated to exceed the TW level, opening up new capabilities for single particle imaging.

Such a modification to the beam switchyard (allowing either accelerator to feed either undulator) is now underway, via a “copper to soft” cross-over line. This provides obvious operational advantages, allowing both accelerators to feed both undulators.
6  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 Project and the LCLS-II-HE extension, based on the assessments of science opportunities conducted over the past three years or so.

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.

Overall, the following drivers are guiding our selection of investment priorities:

- Enabling successful completion of the LCLS-II project.
- Driving high impact science for early operations (following commissioning). This will initially use the 120 Hz beam, and subsequently the high rep-rate SCRF beam.
- Charting a clear trajectory to “flagship” experiments on each instrument.
- Instrument priorities will incorporate the following:
  - Instrument development matched to the phased performance of LCLS-II and the scientific needs of the user community:
    - (i) NEH1.1 attosecond science endstation (VMI/cookiebox), followed by the DREAM dynamic reaction microscope;
    - (ii) NEH 2.2 ChemRIXS moderate-resolution SXR monochromatic endstation, followed by the high resolution monochromatic qRIXS / XPCS endstation;
    - (iii) NEH1.2 Tender X-ray instrument;
    - (iv) Adaptation of the other hard-X-ray instruments (XPP, CXI, XCS, MFX, MEC), pending LCLS-II-HE.
  - Detector, Laser, Controls and Data Systems consistent with LCLS-II science priorities and efficient and effective user delivery.
- Increased user throughput and operational efficiency.
- The budget for long-term R&D (accelerator and photon systems) will be kept approximately at its current level to preserve long-term capability development.
- All other (intermediate-scale) development projects will be treated as the principal area of flexibility, subject to community requirements and LCLS ability to deliver in a timely manner. These will be prioritized in an integrated manner for the facility for each year and the 5/10-year period.
- Upgrade to PW-scale peak-power and high pulse energy (>100 J) laser systems for MEC will be treated as a separate activity, dependent on its own funding stream.

Feedback should be provided directly to the LCLS Director.
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 FeII polypyridyls have revealed important new insight to the excited-state charge and spin dynamics and their coupling to subtle atomic structural dynamics. 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 3MC and 5MC 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 3d 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. 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 Mn4CaO5 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 Biomolecules 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, 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 magnetostrictive coupling present a significant knowledge gap. LCLS experiments using ultrafast resonant X-ray diffraction revealed the spin dynamics in multiferroic TbMnO₃ driven by an intense few-cycle terahertz (THz) light pulse tuned to resonance with an electromagnon mode. 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 YBa₂Cu₃O₆+\(x\) (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-O₂ distances within each bilayer, a contraction of these atomic distances between bilayers, and subtle buckling of the O-Cu-O bonds. 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 \(\text{YBa}_2\text{Cu}_3\text{O}_{6.67}\).\(^{27}\) 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 \(\text{FeSe}\) – 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.
under transient high-pressures. Importantly, quantitative studies enabled by LCLS ultrafast X-rays enable direct comparison to theoretical models and simulations.