First experiments for LCLS-II

NEH 1.2 - TXI

<u>Overview</u>:

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 two broad areas of science in which the unique capabilities of LCLS-II, offered at the NEH 1.2 experimental beamline, 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)

<u>X-ray Pump/Probe and X-ray Wave-mixing:</u> This new instrumentation will offer a unique capability to combine X-rays from two independent FELs. This will open entirely new fields of nonlinear X-ray science and two-color X-ray pump and X-ray probe methods. For example, Fourier-transform inelastic X-ray scattering (FT-IXS) has been demonstrated at LCLS as a powerful method for probing collective mode dynamics (e.g. phonon coupling) in the excited state. However, present experiments are limited to modes that are accessible by optical excitation (at q=0). First experiments in this area will employ element-specific soft X-ray excitation (e.g. O K-edge, or Cu L-edge) for example to probe charge collective modes of cuprate superconductors.

<u>Coherent X-ray Imaging:</u> Single particle imaging experiments at LCLS have produced single-shot coherent diffraction images of viruses, bacteriophages, organelles, and cyanobacteria to name a few. The latest results yield scattering information (significantly above background) out to 3.5 Å, and push the state-of-the-art for 3D image reconstruction to below 10 nm. The optimum conditions for single shot particle imaging are the subject of much active research, and evidence suggests that the optimum region for single particle imaging is in the tender X-ray range (between 2 keV and 6 keV) which may represent the best compromise between scattering cross-section and resolution. Sample heterogeneity combined with the low number of snapshots (at present low repetition rates) further complicates the assembly of complete data sets. Furthermore, this heterogeneity of objects (e.g.conformational heterogeneity in bio-molecules, or structural heterogeneity in nanocatalysis) is central to understanding how they function.

New instrumentation at LCLS-II will support a wide range of science in the tender X-ray range including: coherent X-ray diffractive imaging, small angle X-ray scattering (SAXS), fluctuation SAXS, and time-resolved SAXS. The high repetition rate will open new opportunities for characterizing heterogeneous ensembles of particles in operating (or near-native) environments.

Typical first experiments in 1.2 - X-ray pump, X-ray probe (XP-XP)

(*= early science candidate)

1. ***Phonon Dynamics**
   - **Importance:** Looking at Phonons in High Tc- Copper Oxide superconductors by pumping the material at the Oxygen K edge (543 eV) and/or the Cu L edge (1,096 eV) while probing the diffuse scattering of the crystal at 5 keV. This method can also take advantage of the unique split undulator techniques allowing for multiple pumping photon energies with fixed delays between them

   - **Measurement:** Fourier Transform Inelastic X-ray Scattering to determine phonon dynamics at specific soft and tender X-ray resonances

   - **Requires:**
     - 10 kHz FEL operation for both SXU and HXU (~4.6 keV) sources
     - Fixed target scanning with cryo cooling
     - 1MP 2D forward scattering detector operating at 10 kHz
     - Robust delay line for temporal overlap <10 fs resolution
     - Streamlined data reduction routines

2. ***Coincidence Catalysis**
   - **Importance:** Combination of soft X-ray energy level spectroscopy and tender X-ray structural imaging to correlate material structure and (re)activity. Demonstration of truly unique capabilities LCLS-II

   - **Measurement:** Soft X-ray emission spectroscopy alongside dynamic tender X-ray structural observations on aerosolized nanoparticles (<10 nm) possibly immersed in reactive carrier gases.

   - **Requires:**
     - 10 kHz FEL operation for both SXU (~500 eV) and HXU (~4.6 keV) sources
     - Aerosol nano to micro particle injection of metali catalysis.
     - >1MP 2D forward scattering detector operating at >2kHz
     - Soft X-ray emission spectrometer operating at >2 kHz
     - Robust delay line for temporal overlap
     - Streamlined data reduction routines
     - Soft X-ray self seeded beam for photoemission measurements

3. ***Femtosecond pump/probe single particle imaging as a probe for non-equilibrium dynamics (electronic[1] and nuclear)**

    - **Importance**: One of the great capabilities of LCLS-II is to advance our understanding of far-from-equilibrium processes. The ability to observe ultrafast structural changes in nanoscopic samples is essential for understanding non-equilibrium phenomena such as FEL-matter interaction, matter under extreme conditions, and ultrafast phase transitions.

      With the recent advances in X-ray coherent diffractive imaging and X-ray pump/probe capabilities at LCLS and LCLS-II, we will be able to significantly increase the sensitivity and efficiency of time-resolved imaging. Using X-ray diffraction from intense LCLS-II pulses, we will be able to combine femtosecond temporal and high spatial resolutions, and directly capture ultrafast phenomena in nanoscale systems and ion movement.

    - **Measurement:** Time-resolved X-ray pump/probe diffraction from single nanoparticles (atomic clusters, core-shell systems, nanocrystals, etc.). Recording single shot 2D X-ray diffraction from single nanoparticles. Specimen will be injected into the path of the FEL. The first X-ray laser pulse will capture a coherent diffraction pattern of “pristine” sample and initiate a nanoplasma or other X-ray induced dynamical process. The delayed X-ray laser will image the excited nanoparticle in a similar method but at a different photon energy. The difference in photon energy is required as the two X-ray pulses are not coherent with each other (no interference between the pump and probe). Instead appropriate filters placed on different halves of the detector will filter out either the pump or the probe signal (e.g. a 10 μm Teflon filter has a transmission of ~$1.2\text{x}10^{-3}$ at 1.2 keV and a transmission of 0.2 at 2 keV, while a 10 μm Al filter has a transmission of 0.14 at 1.2 keV and $2.0\text{x}10^{-3}$ at 2 keV).

    - **Requirements**:
        - High photon flux, FEL focus preferable smaller than 1 μm.
        - Ability to overlap the 2 foci in both space and time (as well as record time delay)
        - >1MP 2D forward scattering detector operating at >2kHz

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[1] The electronic motion may have to be observed indirectly/non-structurally (via observing effects on K or L edge resonances.) This is due to limits on the maximum photon energy of the LCLS-II upgrade and hence maximum resolution that can be obtained. Of note if a crystal sample is used and if the accelerator outperforms its expected ~5 keV upper photon energy limit then atomic resolution and orbitals will be observed structurally/directly (TXI is planning for this option and has a maximum photon energy of 7 keV on the hard X-ray branch line).

        - High repetition rate from LCLS-II will allow for better statistics will allow for recording a large and precise data set necessary to correct for particle size distribution effects

4. **Femtosecond X-ray reflection from a surfaces far from equilibrium.**

    - **Importance**: This novel approach will enable new insights into core-hole/matter interaction with unprecedented temporal resolution and will pave a way for a new class of ultrafast X-ray optical experiments.

      We propose an X-ray pump/X-ray probe experiment to study transient ionization and recombination mechanisms in X-ray irradiated surfaces with femtosecond resolution. This class of experiments will shed light on little understood ionization avalanche occurring during the first hundred femtoseconds after the beginning of the intense pump pulse and *before* the onset of surface ablation. We will focus two intense FEL pulses on a flat Si surface and directly mirror the Si surface plasma evolution by monitoring transient reflection changes in dependence on the delay between the pump and the probe pulse in 10 femtosecond steps. The ionising pump X-ray laser pulse will release core electrons and initiate complex cascades of subsequent ionisation dynamics such as Auger decays and impact ionization which result in highly non-equilibrium plasma formation. We will tune the X-ray wavelength of the pump pulse to be just below and just above to the Si K-edge Probe measurements will be conducted on either the total external reflection regime of the surface or observe Bragg peaks for intensity changes. We expect two significant effects. First, depletion of K and valence shells will shift the absorption edge to higher energies and suppress absorption. Second, an increasing portion of delocalized (quasifree) electrons will enhance the reflectance efficiency of the probe pulse by up to factor 50. Both effects will be directly detected as changes in reflectivity by a two-dimensional photon detector placed downstream to observe structural changes of the lattice as well as electronic changes.

    - **Measurement:** Small & large angle X-ray reflectance from X-ray laser-superheated surface. The first X-ray probe will ionize the surface of the substrate and delayed, second pulse will reflect from the excited surface near grazing incidence or on a Bragg peak. The measurement will consist of the X-ray reflectance efficiency and shape

    - **Requirements**:

        - High photon flux, focus preferable smaller than 1 μm

        - >1MP 2D forward scattering detector operating at >2kHz
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        - High repetition rate from LCLS-II will allow for better statistics and highly systematic studies.
        - X-ray emission spectrometer to observe Si K-alpha and K-beta emission (1.74 and 1.84 keV respectively, with a resolution of 0.25 eV)

5. **X-ray four-wave mixing (Stimulated X-ray Raman Spectroscopy)**

    - **Importance**: The new capabilities offered by TXI will allow for a non-collinear geometry and for significant difference in photon energies used in X-ray Pump/X-ray Probe methods. One such method that can take advantage of this is the nonlinear method of X-ray stimulated Raman scattering. Non-stimulated Raman scattering or spectroscopy is an inelastic scattering technique that is used as both a laser spectroscopy technique and as an X-ray spectroscopy technique. The technique looks at differences in the photon energy of an incident photon with that of a scattered photon. The energy difference, as well as the system's geometry, provides information on phonons and other vibrational modes. In the X-ray regime it has also been used to study electronic transitions as well as core holes of low Z materials in bulk. Stimulated Raman spectroscopy, also called Coherent anti-Stokes Raman spectroscopy (CARS) or Coherent Stokes Raman spectroscopy, is similar to the technique of Raman spectroscopy, except a dressing pumped field is used to stimulate the transitions. The dressing/pump field causes a stimulated excitation, and makes it into a coherent process, dramatically increasing the signal output over the non-simulated case. X-ray stimulated Raman spectroscopy could potentially become a powerful method to study electronic transitions in materials.

    - **Measurement:** Two X-ray pulses that are separated by the transition photon energy of interest, overlap and interact with the sample. This produces produces a Raman signal that is spatially separate (unique k-vector axis) to that of either the stimulating pump field or the probing stokes pulse. A 2D downstream detector observes the intensity of the Raman signal.

    - **Requirements**:

        - High photon flux, focus preferable smaller than 1 μm

        - >0.25MP 2D forward scattering detector operating at >2 kHz operating >1 m from interaction.
        - Ability to observe, measure, and synchronize the two LCLS pulses to <10 fs (duration of the pump and stokes pulses)
        - High repetition rate from LCLS-II will allow for better statistics and highly systematic studies.

Typical first experiments in 1.2 - Forward Scattering

(*= early science candidate)

6. ***Single Particle Imaging**
    - **Importance:** First ever in-situ difference measurement between bacterial and human ribosomes maps
    - **Measurement:** Collect ~10^6 diffraction patterns of aerosolized single proteins, orient these diffraction patterns and place them in a 3D volume of average intensity. Possibly observe dynamics with a pump laser triggering a chemical reaction
    - **Requires:**
        - Aerosol injection system (for higher hit rates, sample consumption reduction, reduce clustering & removal of liquid around the molecule)
        - Aerosol viewing
        - Hit/Miss Veto data system
        - 2 detector pa**nels that are at least 400 x 200 pixels, Rep rate > 2 kHz (10 kHz is better)**
7. ***Fluctuation X-ray Scattering**
    - **Importance:** Real time movie of antibiotic binding to ribosomes
    - **Measurement:** Collect ~10^5 fluctuation diffraction patterns of ribosomes in liquid with antibiotics with varying mixing time. It is possible to enhance the signal and observe the RNA by taking two data sets: one above and one below the phosphorus edge.
    - **Requirements:**
        - Liquid mixing jet system
        - On axis viewing
        - 2 detector panels that are at least 1000 x 1000 pixels Rep rate > 2 kHz (10 kHz is better)

Logistical Synopsis for executing experiments: In conjunction with the TTO plan laid out by the LCLS-II Project, early science experiments occurring in the abovementioned timeframe will be led by LCLS staff, with significant contribution from relevant users in the community. Experiments will serve as a bridge between technical commissioning and the start of general user access.

Initially, equal access will be split between photon operations (LCLS) and the Accelerator Directorate for machine development (MD). Typical photon experiments will range from 24-36 hours, with equal time given back to the MD program. Technical changes between early science experiments will be severely limited and strictly enforced. Only minimal changes to the instrument's baseline scope will be considered.