

First experiments for LCLS-II

NEH 1.1 - DREAM

Overview:

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 operate in a soft x-ray range (250eV to 1.5 keV), 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 areas of science in which the unique capabilities of LCLS-II, offered at the NEH 1.1 DREAM instrument, 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)

Fundamental dynamics of energy & charge: Charge migration, redistribution and localization, even in simple molecules, are not well understood at the quantum level. These fundamental phenomena are central to complex processes such as photosynthesis, catalysis, and bond formation/dissolution that govern all chemical reactions. Ultrafast soft X-rays at high-repetition-rate from LCLS-II will provide qualitatively new probes of excited-state energy and charge flow and how they work in simple and complex molecular systems. New LCLS-II instrumentation (NEH 1.1) will enable sophisticated coincidence measurement schemes for kinematically complete experiments 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.

First experiments will include: fundamental studies of molecular dissociation trajectories and the role of electronic potential energy surface (PES) gradients in the Franck-Condon region; mapping excited-state isomerization trajectories of acetylene; mapping the ring-opening reaction of cyclohexadiene; capturing non-Born-Oppenheimer relaxation channels in the nucleobase thymine; and capturing the quantum symmetry breaking processes that mediate the emergence of chirality in model systems such as fluoroformaldehyde (HFCO).

Typical first experiments in 1.1 (DREAM) - Fundamental dynamics of energy & charge:

(* = early science candidate)

General time resolved 2 color COLTRIMS (charge migration, energy transfer, bond breaking, vibronic excitation, photoexcitation):

1. *Time-resolved Acetylene isomerization (or similar target with single and/or double hydrogen migration)
 - **Importance:** Detailed and direct study of the time resolved molecular structure and configuration changes, dynamics of the intermediate states in simple systems.
 - **Measurement:** Coincident ion-ion-electron-electron time and momentum resolved detection following IR-pump x-ray probe (or vice-versa, or x-ray/x-ray)
 - **Requires:**
 - COLTRIMS spectrometer
 - 1/20 electron/ion energy resolution (1/30 resolution for long term);
 - 5 deg angular resolution (<3 deg long term)
 - Multi-hit/high rep rate TOF+2D detectors at each end of the spectrometer (up to 5 fragments per detector per laser pulse, with <10ns pulse-pair resolution, <5ns long term)
 - Optical / x-ray timing synchronization (resolution) of <10fs FWHM to map out 3 to 5 steps during 50 fs evolution
 - X-ray focus < 0.3 μm
 - X-ray rep rate 100kHz to 1MHz

2. Optical pump (x-ray) / x-ray probe of HFCO, CHBrClF
 - **Importance:** Imaging “from within” of intrinsic and induced chirality; circular dichroism
 - **Measurement:** Induced by a circularly polarized optical or x-ray pulse chirality is imaged by an outgoing photoelectron originated by the x-ray probe pulse. Photoelectron is detected in coincidence with the fragmenting ions
 - **Requires:**
 - COLTRIMS spectrometer
 - (see above)
 - 800 nm (circular) / x-ray timing resolution of 5fs via ATM
 - Optical / x-ray delay range of 5 - 200fs
 - 5fs x-ray pulses, up to 0.05 mJ
 - <10fs circularly polarized optical laser pulses, with $>10^{13}$ W/cm² peak field

differential molecular-frame photoelectron angular distribution (MFPAD) to complement the dissociation pathways information in this charge transfer study

- **Requires:**
 - COLTRIMS spectrometer (as above)
 - Optical / x-ray timing synchronization of 30fs
 - X-ray laser pulses of <50fs
 - X-ray & optical laser rep rate of >100kHz for high statistics feasibility
 - Optical / x-ray delay range of +/- 1 ps
 - <50fs optical laser pulses, with $>10^{13}$ W/cm² peak field
 - Long term: Future 2 color x-ray pump / probe

Logistical Synopsis: 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.