High Energy Upgrade: LCLS-II-HE
High Repetition Rate Soft X-rays $\Rightarrow$ Hard X-rays

**Electronic & nuclear coupling**

LCLS-II-HE provides:
- Ultrafast coherent X-rays
- ~1 Ångstrom (~12 keV)
- High repetition rate

**Emergent properties**

**Materials heterogeneity**

**Dynamics**
- excited state
- non-equilibrium
- transient structures

**Fluctuations**
- ground state structure
- spontaneous evolution

**Heterogeneity**
- structural complexity
- ground & excited states

LCLS-II-HE provides new insight to structural dynamics at the atomic scale across a cross-cutting theme of the *Transformative Opportunities* identified by BESAC
LCLS-II-HE: Enabling New Experimental Capabilities

Structural Dynamics at the Atomic Scale

- Expand the photon energy reach of LCLS-II to >12 keV
  - Atomic resolution requires ~1 Å
- ~1,000-fold increase in average spectral brightness re: LCLS
  - Average coherent X-ray power (spatial and temporal) is transformative
- Hard X-ray pulses in a uniform (programmable) time structure at a repetition rate of up to 1 MHz
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![Graph showing photon energy vs. average brightness for LCLS, LCLS-II, LCLS-II-HE, and Eu-XFEL with a y-axis scale from $10^{19}$ to $10^{26}$ and x-axis scale from 2 to 20 keV.]
Dynamics near the FT Limit

• >300x increase in average spectral flux (ph/s/meV) beyond DLSRs
• Spectroscopy & inelastic scattering at high resolution
  • IXS meV resolution up to 20 keV
    sub-meV (dispersive spectrometer, ~10 keV)
  • RIXS ~5 meV (quartz- and sapphire-based analyzers)
• Low-energy modes in quasi-elastic energy region
• Momentum transfer spanning entire Brillouin zone
• Sensitivity (e.g. to electronic vs. lattice modes)
• Excited-state dynamics – near-equilibrium perturbations (5 meV ⇔ 300 fs)
• Excited-state potential mapping with element-specificity
  (e.g. metal-ligand stretch modes)

<table>
<thead>
<tr>
<th>Resolution</th>
<th>Hard X-ray Flux on Sample</th>
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<tbody>
<tr>
<td>LCLS-II-HE seeded (SASE)</td>
<td>~100 meV</td>
</tr>
<tr>
<td></td>
<td>~10^{14} (10^{13}) ph/s</td>
</tr>
<tr>
<td></td>
<td>~10^{13} (10^{12}) ph/s</td>
</tr>
<tr>
<td>ESRF</td>
<td>~10^{13} ph/s</td>
</tr>
<tr>
<td></td>
<td>~10^{11} ph/s (ID28)</td>
</tr>
<tr>
<td></td>
<td>~10^{10} ph/s (ID28)</td>
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<tr>
<td>SPring-8</td>
<td>~10^{12} ph/s</td>
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<td>~10^{11} ph/s</td>
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<tr>
<td></td>
<td>~10^{10} ph/s</td>
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<tr>
<td>APS</td>
<td>~10^{12} ph/s (MERIX)</td>
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<td>~10^{10} ph/s</td>
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<tr>
<td></td>
<td>~10^{9} ph/s (UHRIXS)</td>
</tr>
<tr>
<td>NSLS-II</td>
<td>~10^{10} ph/s</td>
</tr>
</tbody>
</table>
Fluctuations & Heterogeneity
Atomic resolution, Ultrafast time scales, Operating conditions

Photon Correlation Spectroscopy (XPCS)
- “Sequential” real-time mode (fast 2D detectors)
- “Two-pulse” mode (<100 fs) with pulse pairs directly from XFEL
- “Programmable” time structure encoded in X-ray pulse sequence
- High rep rate, lower peak power, sample replacement

Time-domain (and FT) Inelastic X-ray Scattering
- Time-resolved (diffuse) X-ray scattering following impulsive excitation of collective modes
- Perturbative regime – ground-state fluctuations (fluctuation-dissipation theorem)
- Non-equilibrium regime, excited-state dynamics
- High resolution via Fourier-transform of coherent response (1 THz ⇔ 4 meV)
- High-brightness hard X-rays – atomic structure (PDF)

How can we exploit the high rep rate and the potential for $10^8$-$10^{10}$ snapshots/day to:

- Characterize heterogeneous ensembles,
- Capture rare transient events,
- Map spontaneous dynamics *operando*

**Advanced Experimental Approaches**
- Coherent diffractive imaging (and/or serial crystallography) with spectroscopy
- Solution scattering, rapid mixing…
- Fluctuation X-ray scattering

**Advanced Computational Approaches and Data Science**
- Mapping reaction landscapes via diffusion maps, manifold embedding and related Bayesian approaches
- Capturing rare events via automatic pattern recognition and related machine-learning approaches
Workshop Charge

- Identify most important science opportunities (transformational, grand challenge level) that can uniquely be addressed using capabilities of LCLS-II-HE (high rep rate hard X-rays, initially up to 12.8 keV, and in the future beyond 20 keV)
  - Near-term science consistent with initial LCLS-II-HE capabilities and augmented LCLS hard X-ray instrumentation
  - Future science consistent with projected LCLS-II-HE capabilities and advanced instrumentation
  - Succinct statement of why this science is transformational
    - What are important outstanding questions in your field?
    - Why have they not been answered (what is impeding progress, why now, why LCLS-II)?
    - What is the potential broader impact if we can answer these questions (why are they important)?

- Identify relevant experimental approach(es) and key requirements or capabilities – particularly for advanced approaches that are not well developed
  - Instrument(s), computational approaches, optics, endstation(s), detectors, lasers, sample injectors, etc.
    - Photon flux, pulse duration, rep rate, photon energy etc.

- Compare experimental approach to current state-of-the art & assess alternative approaches
  Can the experimental approach leverage existing instrumentation/expertise?
  Where are the gaps, or what R&D is required?
  - Can the science be done with other existing sources?
    (e.g. diffraction-limited synchrotrons, cryo-EM, table-top HHG, etc.)
### Agenda & Sessions

- All are encouraged to present ideas (template provided – see website)
- Breakout summaries at plenary closeout
- Scribes will take notes and collect presented materials for internal use only

#### Chemical dynamics, charge transfer, molecular photocatalysts, natural & artificial photosynthesis

- Kelly Gaffney
- Mike Minitti
- Amy Cordones-Hahn

- Materials Physics: Heterogeneity, nonequilibrium dynamics & spontaneous fluctuations
  - Aaron Lindenberg
  - Mariano Trigo
  - Aymeric Robert

#### Homogeneous & heterogeneous catalysis, interfacial & geo/environmental chemistry

- Simon Bare
- Dennis Nordlund

#### Quantum Materials

- Wei-Sheng Lee
- Diling Zhu
- Hermann Durr

#### Biological Function & Structural Dynamics

- Sebastien Boutet
- T.J. Lane