Scientific Opportunities at LCLS-II: *the High Repetition Rate Revolution*

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Linac Coherent Light Source, SLAC
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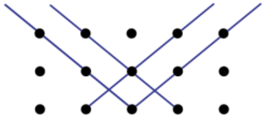
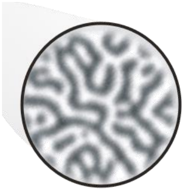
Outline

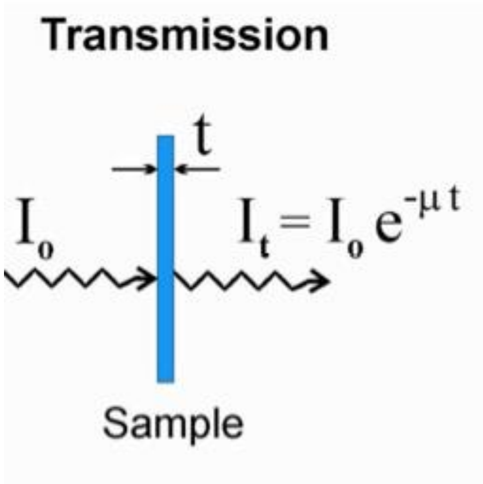
- X-rays: powerful tool for science
- X-ray free electron lasers
- Experiments at LCLS
- The LCLS-II upgrade
- Science opportunities
  - Chemistry
  - Materials physics
- New methods
- Further reading

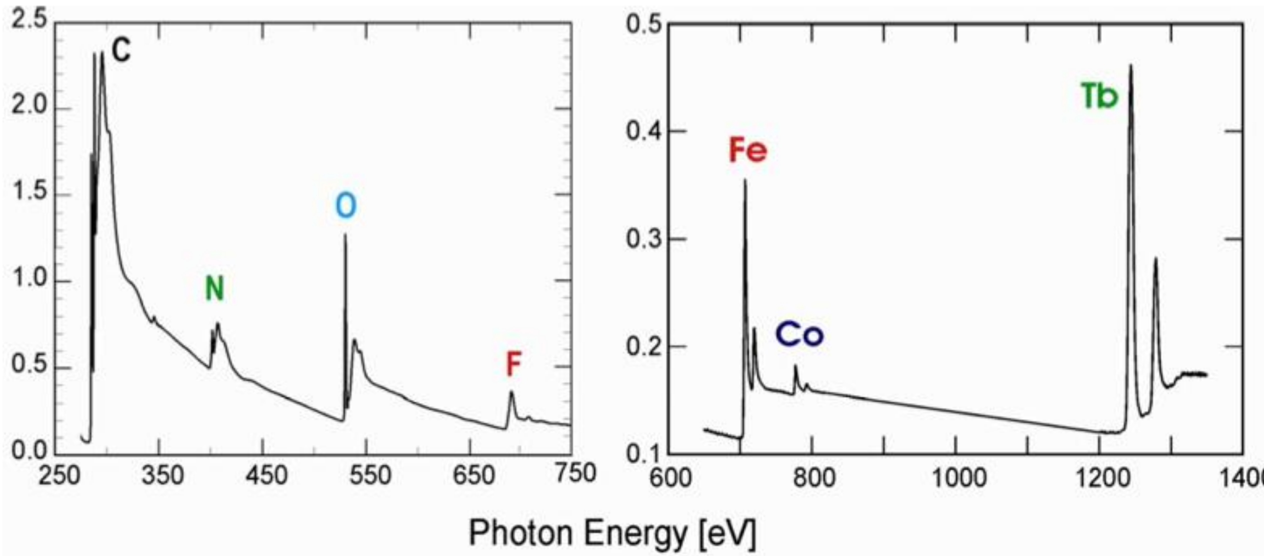
Why do we use x-rays?

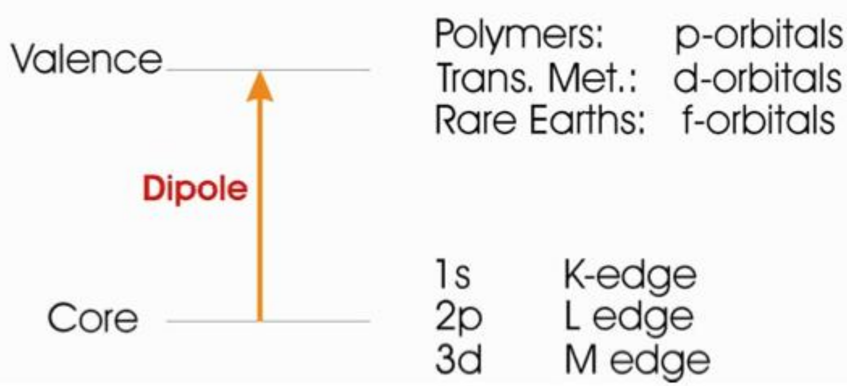
Capability	Technique
Access to atomic length scales	Scattering 
Capture texture exactly and access bulk	Imaging 
Element and chemical sensitivity	Spectroscopy 

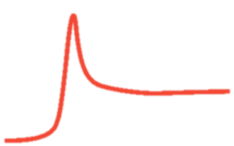
Soft X-ray Absorption Spectroscopy

Element Specific Sensitivity and Contrast







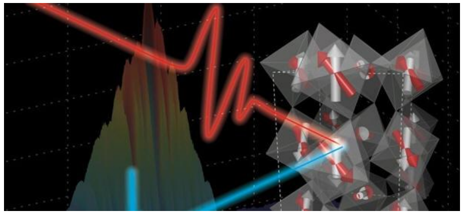


Why do we use X-ray Free Electron Lasers?

X-rays provide element specificity and atomic resolution…

➢ Ultrashort X-ray Pulses

- Study ultrafast (femtosecond) dynamics
- Out-run damage to samples

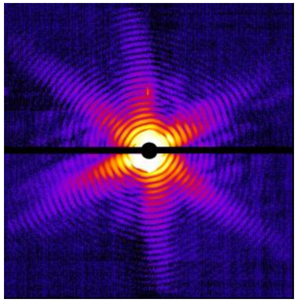


➢ High Energy Per Pulse

- Enables single shot imaging
- Generates unprecedented electric fields

➢ Coherent X-ray Pulses

- Far field scattering methods improved spatial resolution



Operating VUV and X-ray FELs Worldwide

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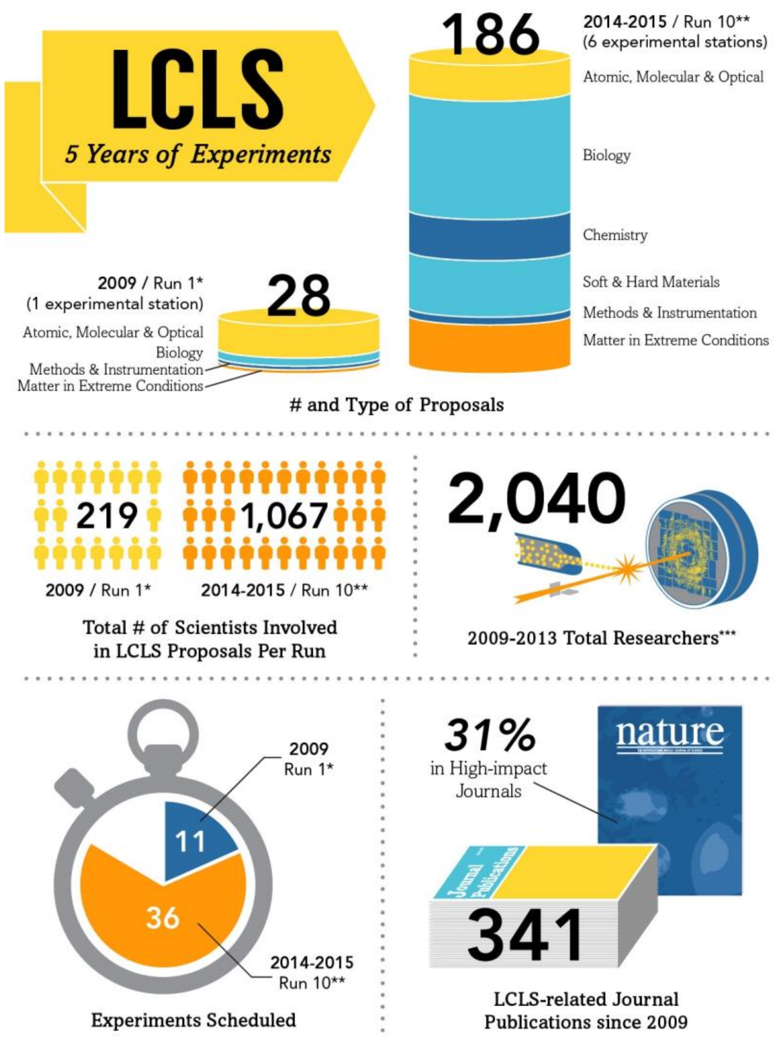
Facility	Location	First light	Energy (keV)	Rep Rate
FLASH	Germany	2006	0.01-0.3	10 Hz (up to 5000Hz)
LCLS	USA	2009	0.25-9.5	120Hz
SACLA	Japan	2011	5-15	60 Hz
FERMI	Italy	2011	0.01-0.4	50 Hz

Many more under development…..

VUV: below 0.2 keV
Soft X-ray: 0.2-2.0 keV
Hard X-ray: 4-25keV

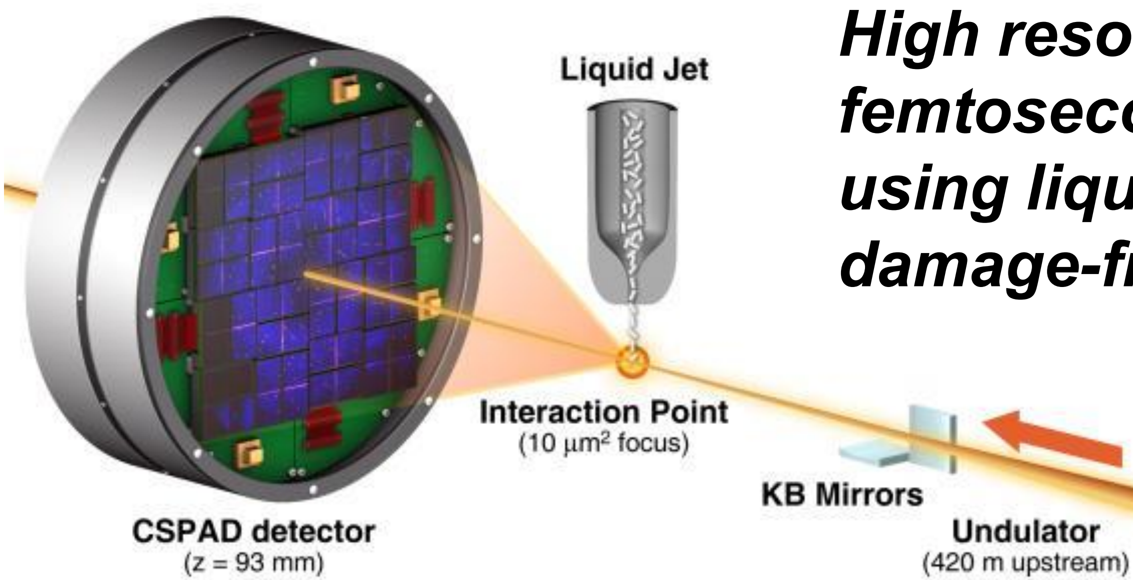
How does LCLS work?

- Users submit proposals twice a year
- Beamtime proposals are evaluated via peer review
- ~20% of proposals are granted beamtime
- Successful proposals are awarded an average 60 hours of LCLS beamtime
- The average user group is ~15 people

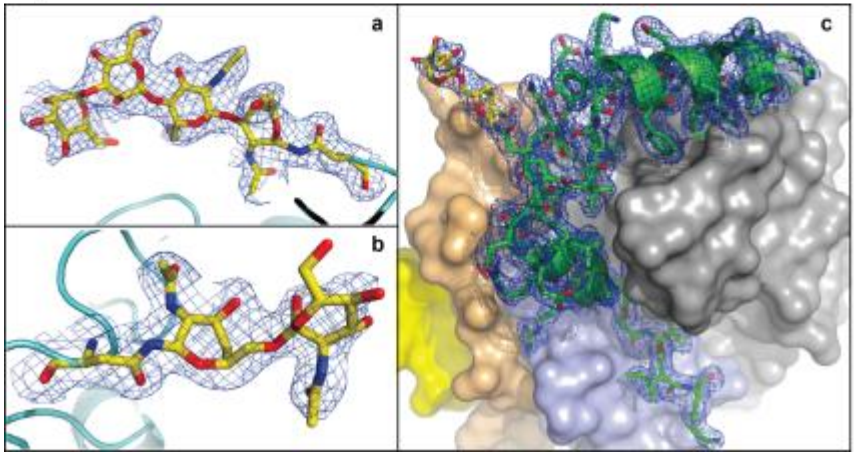


* Run 1, the first operating period at LCLS, was October-December 2009.
** The Run 10 operating period is scheduled October 2014-March 2015.
*** October 2009-October 2013 total number of scientific researchers engaged in approved research at LCLS.

Nano-crystalography at LCLS



High resolution serial femtosecond crystallography using liquid jets can produce damage-free structures



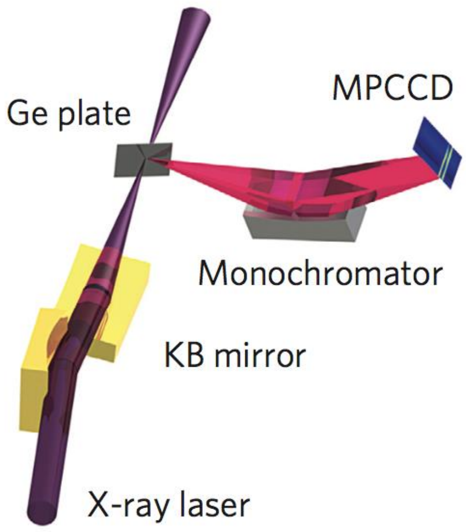
In-vivo grown crystal of a glyco-protein

Redecke *et al*, Science **339**, 6116 (2012)

Boutet *et al*. Science, **337** (6092) 362 (2012)

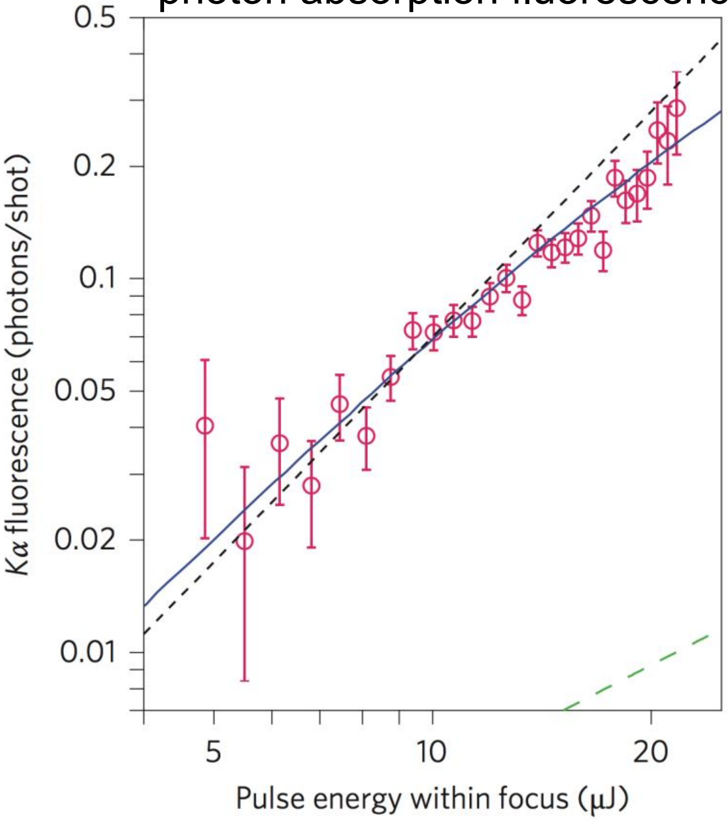
X-ray two-photon absorption at SACLA

First observation of a third order nonlinear process with hard x-rays.



K. Tamasaku, et al., Nature Photonics (2014)

Pulse-energy dependence of germanium two photon absorption fluorescence



Watching surface bonds break in real-time at LCLS:

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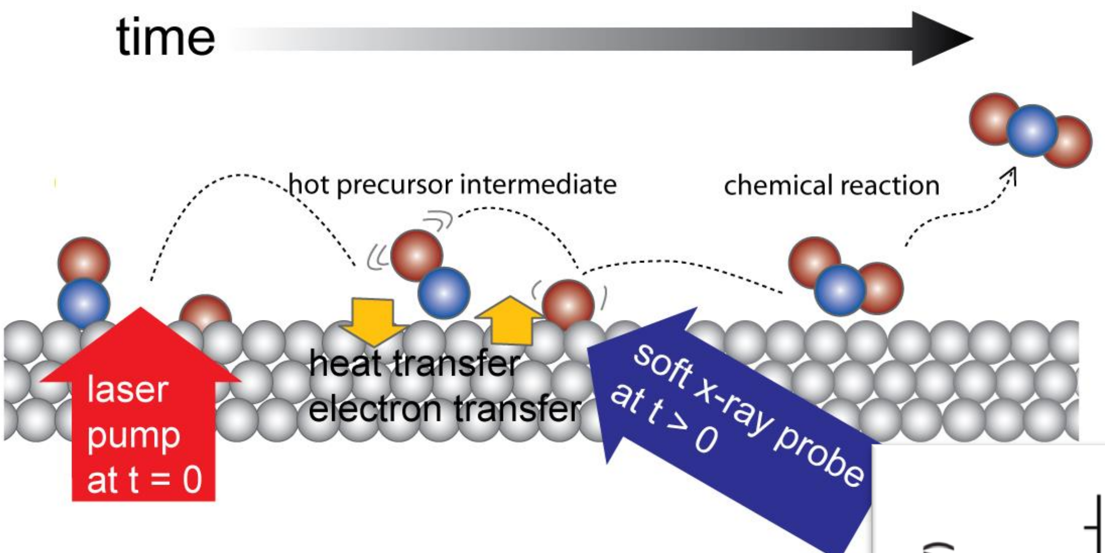
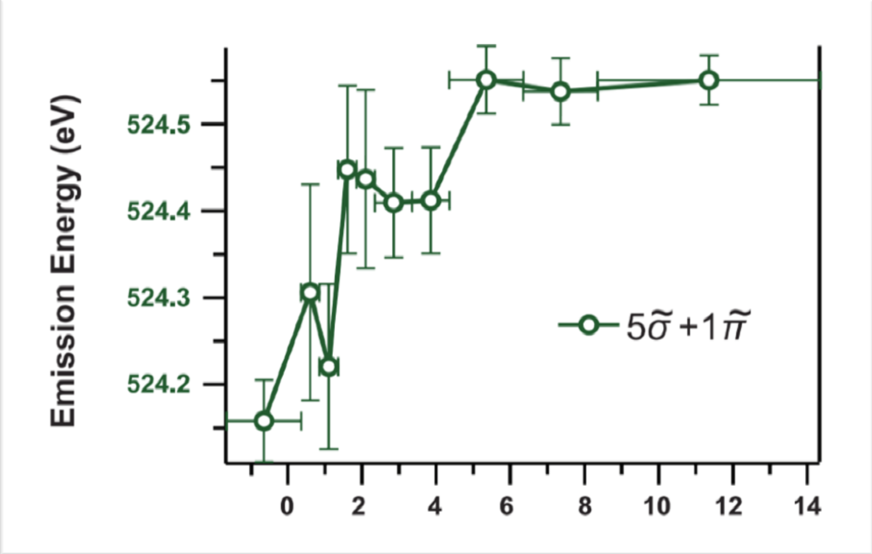
Transient weakly bound state observed in desorption process



Bond formation of carbon monoxide on a ruthenium substrate

Electronic structure changes consistent with a weakening of the CO interaction with the substrate but without notable desorption.

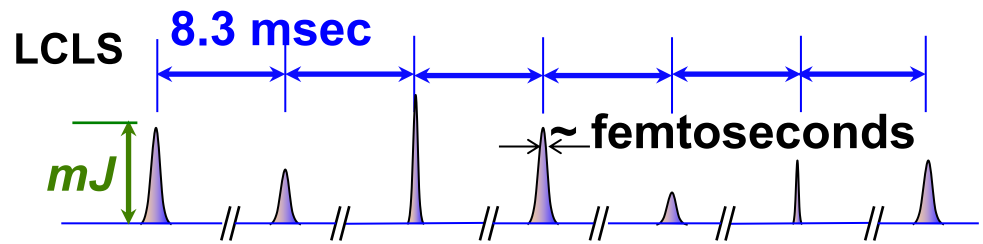


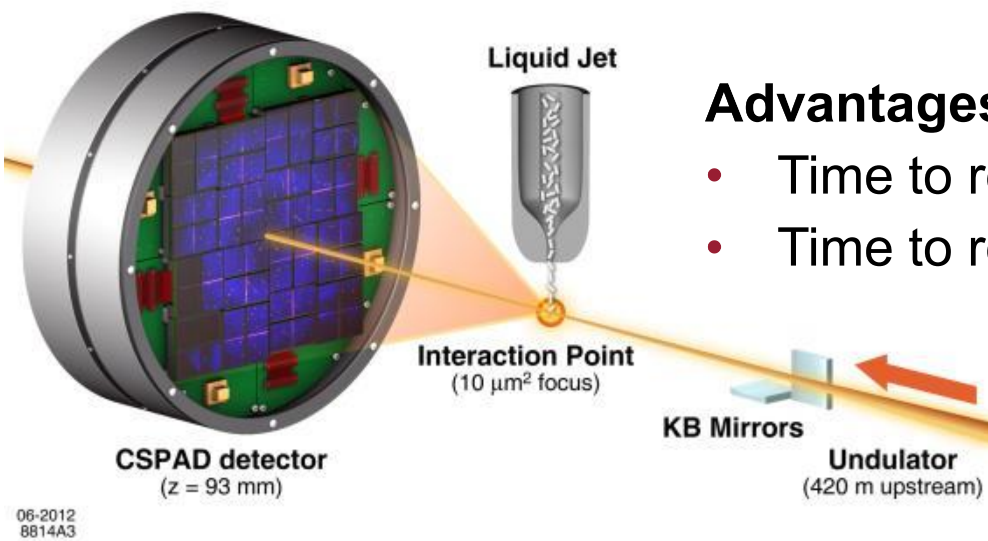
M. Dell'Angela, *et al.*, Science **339** 6125 (2013)
M. Beye, *et al.*, Phys. Rev. Lett. **110** 186101 (2013)

How we detect x-rays at LCLS?

Single shot mode

- All experimental parameter that may change are recorded for each pulse
- Data must be sorted by the *independent variable* after the experiment
- Each x-ray pulse is different



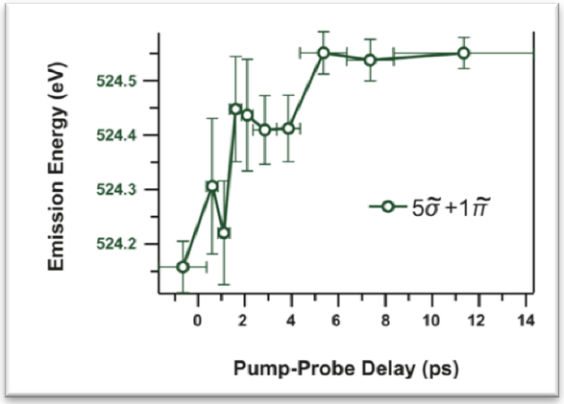


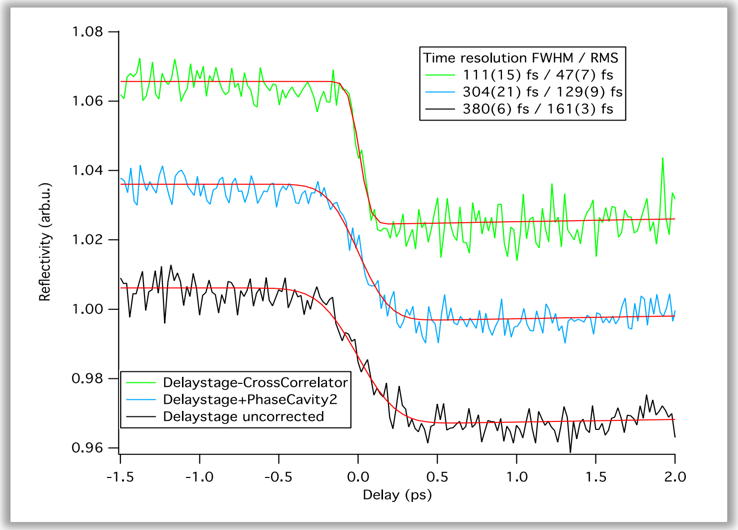
Advantages of single shot mode

- Time to read out detectors
- Time to renew destroyed samples

Limitations to single shot data collection at 120Hz

- Limited data collection volume
  - Low repetition rate (120 Hz)
  - Only 60 hours to collect data
- Source stability
  - Energy
  - Arrival time
  - Duration
  - Wavelength & Bandwidth
- Limited Access
  - Only one x-ray source
  - One size fits all experiments

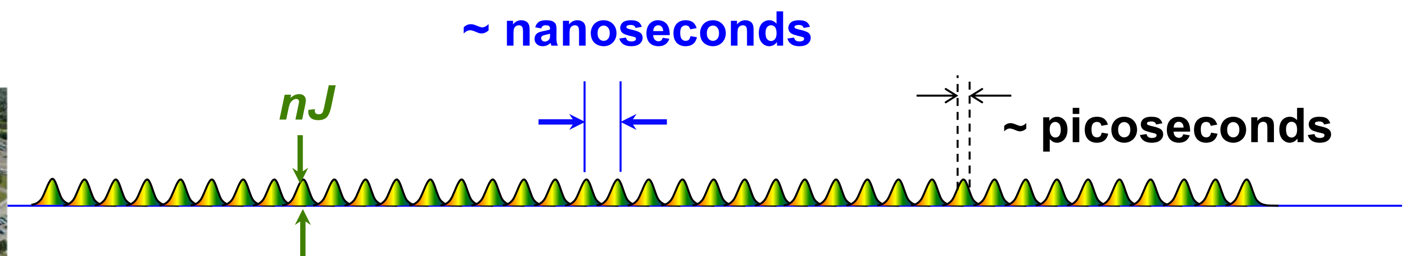




Accumulation Data Collection

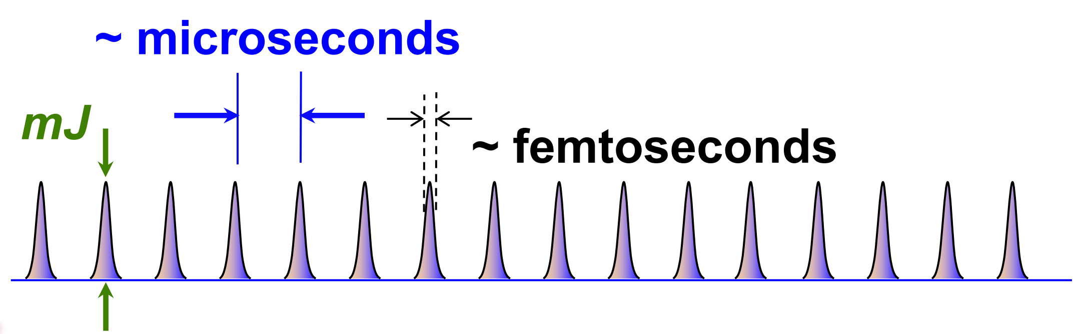
- To the extent each x-ray pulse is the same, we can accumulate the detected photons over many pulses
- But this is exactly how experiments are done at storage ring sources

storage ring X-ray source (NSLS-II)



- Combining the mJ, fs pulses of an FEL with the stability of a storage will revolutionize x-ray experiments.

2019 LCLS-II



LCLS-II: A Revolution in X-ray Science

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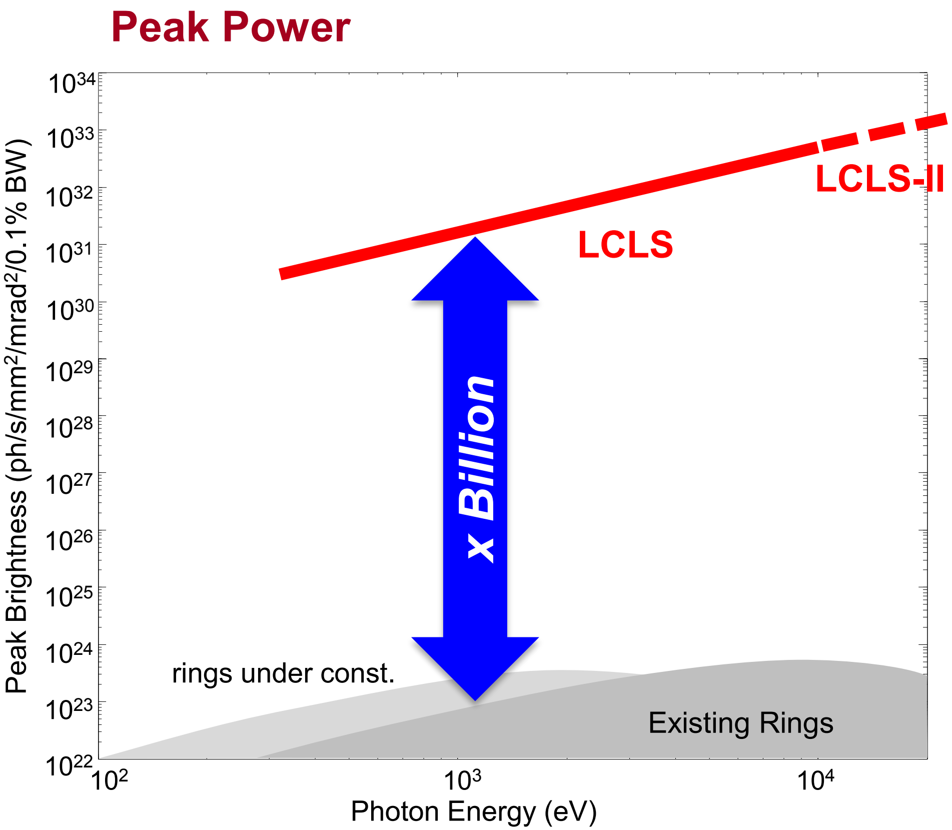
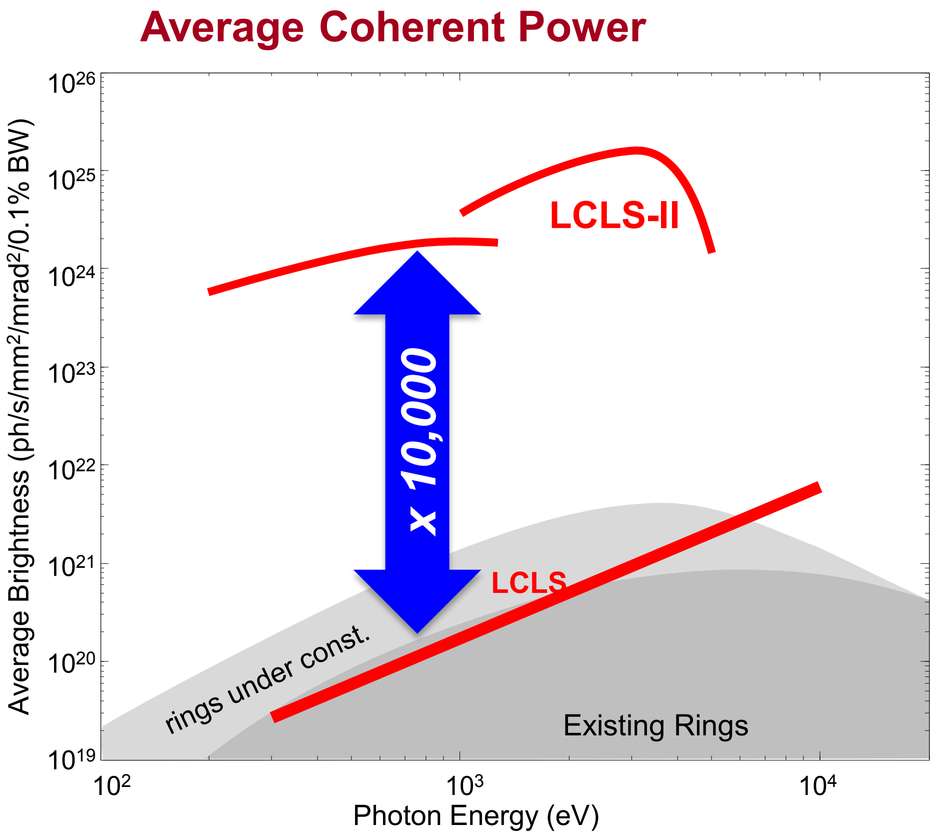
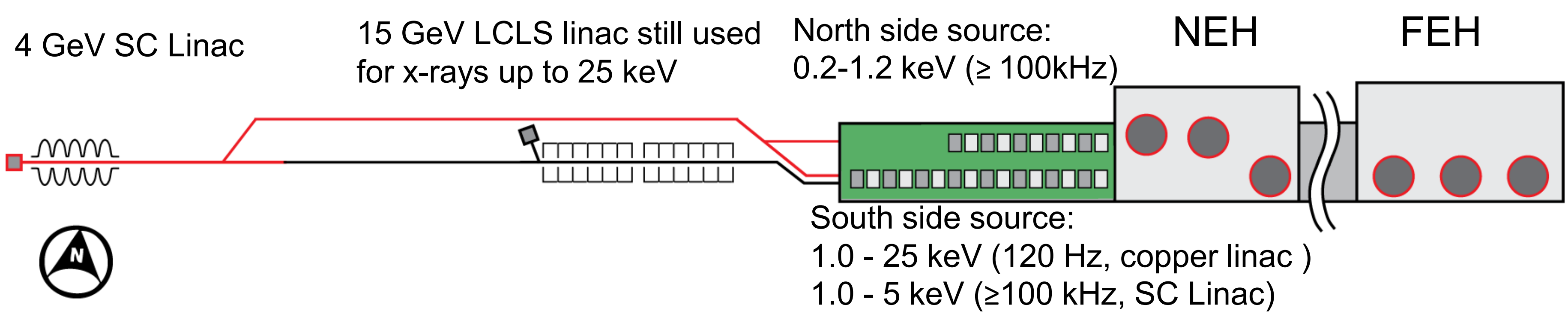




LCLS-II:
- Repetition rate
- Stability
- Coherence (seeding)
- Photon energy reach

LCLS-II Upgrade Project Scope

Accelerator	<u>**Superconducting linac**</u>: 4 GeV
Undulators in existing LCLS-I Tunnel	New variable gap (north) New variable gap (south), replaces existing fixed-gap und.
Instruments	Re-purpose existing instruments (instrument and detector upgrades needed to fully exploit)



Development of Science Drivers
LCLS-II Science Opportunities Workshops

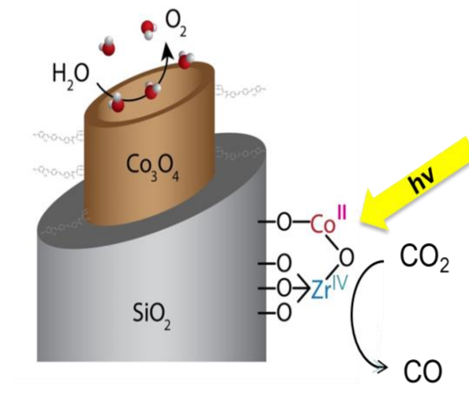
Science opportunities workshops held at SLAC in February, 2015

Workshop Registrants	
Chemistry	165
Materials Physics	264
Life Sciences	149
MEC Breakouts	116
Unique Registrants	**410**

Predictive understanding of catalysis

LCLS-II Science Opportunity

- Understand the fundamental processes that occur on metal surfaces during catalytic reaction conditions in order to design new, efficient, and selective catalysts
- Provide a robust structure-function relationships for materials in electronic excited states
- Understanding and predicting photon driven phenomena

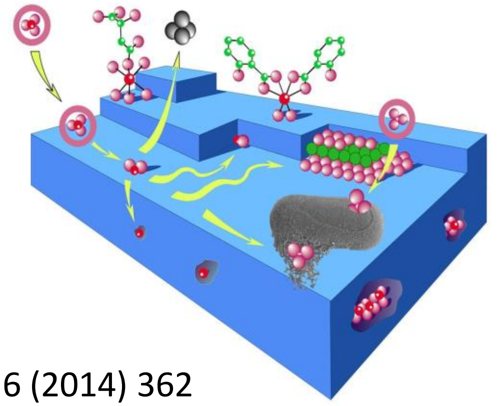


Significance and Impact

- Light harvesting & charge separation are fundamental to understanding natural & artificial photo-catalytic systems
- Interfacial chemistry and charge-transfer in real time & under reactive conditions

Strengths of SRF source

- High average power at high rep rate (moderate peak power)

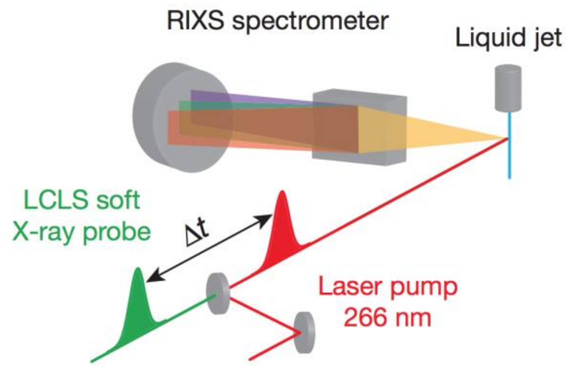


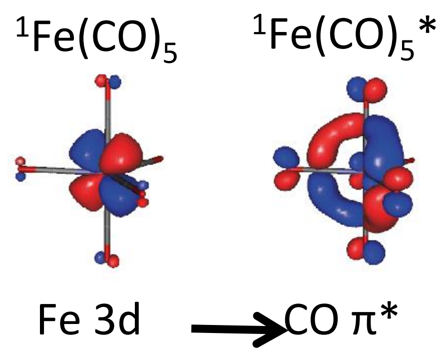
Heinz Frei et al. Nature Chem. 6 (2014) 362

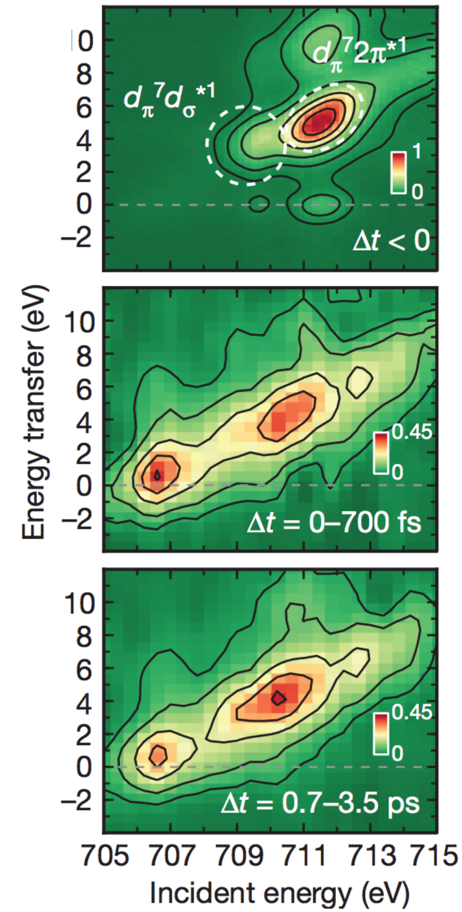
Example: X-ray Raman Studies of Molecular Dynamics

- Soft X-ray RIXS maps molecular orbitals & their evolution
- Element-specific: transition-metals & ligands
- Local chemical structure & bonding
- Current limitations:
  - Sensitivity - observe only large molecular changes, in model complexes, at high concentrations
  - Limited time information - average X-ray flux (rep rate)

Ultrafast X-ray Raman Spectroscopy (resonant inelastic X-ray scattering – RIXS)







P. Wernet et al., *Nature*, 520, **78** (2015)

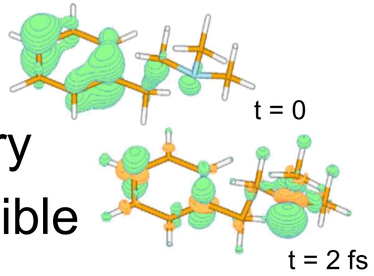
Chemistry: Charge migration and redistribution

Follow molecular transformations & bond formation

LCLS-II Science Opportunity

- Map electron dynamics on sub-angstrom and sub-femtosecond scales and reveal coupled electronic and nuclear motion in molecules

Significance and Impact



- Charge migration initiates all charge transfer chemistry
- Dynamics on fundamental time scale have been invisible before this

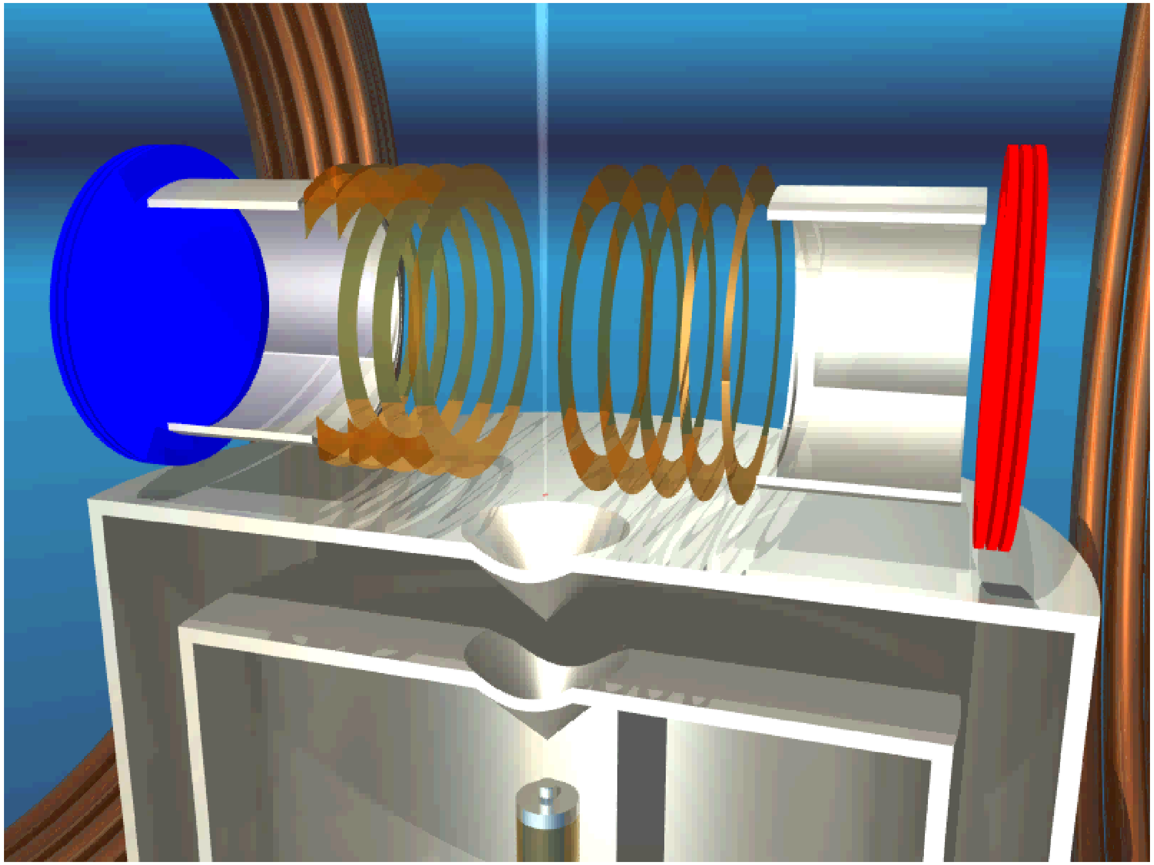
Strengths of SRF source

- Coherent bandwidth and pulse intensity are essential for transient impulsive electronics
- 2-color (element selectivity)
- High rep rate for rare events and coincidences

Chemistry: Reaction Microscope (COLTRIMS)

COLTRIMS: Cold Target Recoil-Ion Momentum Spectroscopy

- Entanglement & correlation dynamics in many electron/nuclei systems
- Time-resolved energy & angular correlations between electrons & ions
- 2-color X-ray pump/probe is chemical/element specific
- Rare coincidence events ($\sim 10^{-5}$) ⇒ high repetition rate



Timur Osipov, SLAC

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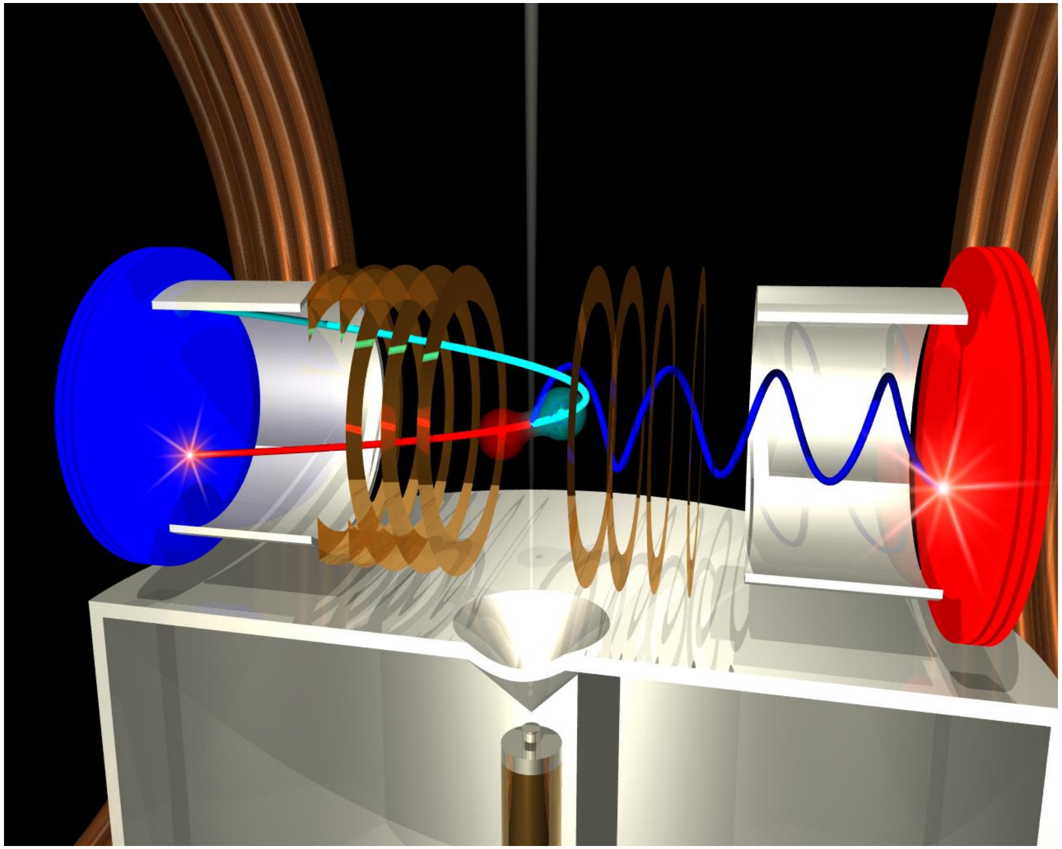


Timur Osipov, SLAC

Materials Physics: Revealing interactions among degrees of freedom in high temperature SC cuprates

LCLS-II Science Opportunity

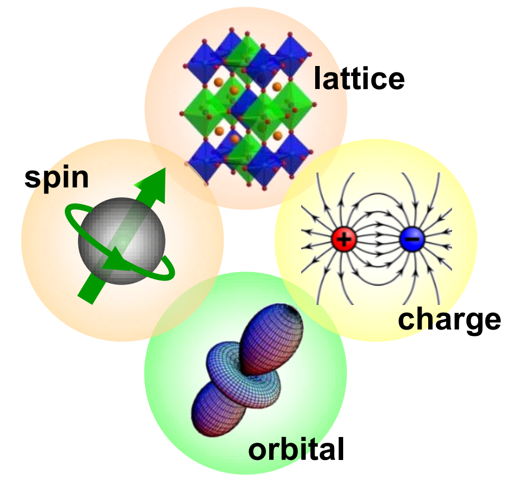
- Magnetic, lattice, and charge degrees of freedom are strongly intertwined makes it difficult to understand the mechanism of HTSC.

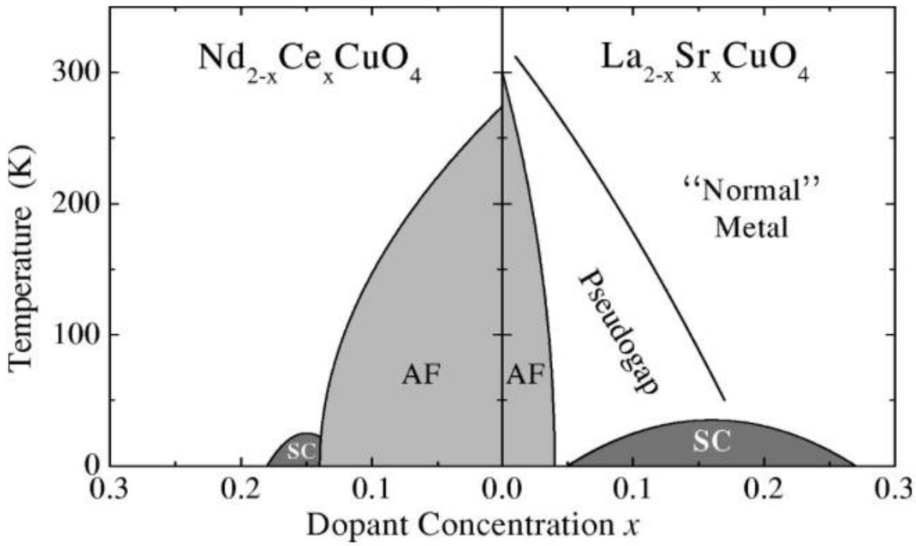
Significance & Impact

- Clarify interactions among different degrees of freedom in high T_c cuprates, that may provide important clues to reveal its mechanism.
- Pathway to manipulate novel phase and perhaps lead to SC with even higher T_c.

Strengths of SRF source

- Time-resolved RIXS with Fourier-transform limited time and energy resolution.





SRF2015, Bill Schlotter wschlott@slac.stanford.edu

Wei-Sheng Lee, SLAC

Materials Physics: Collective excitations using RIXS

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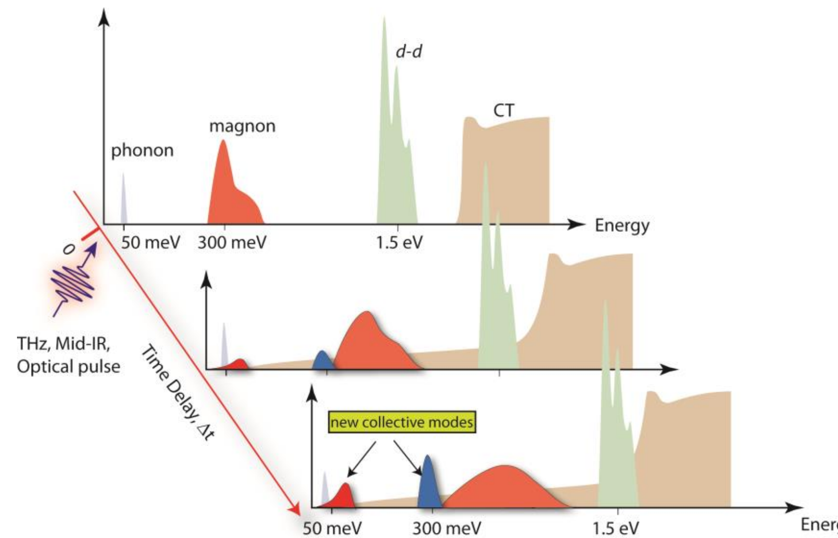
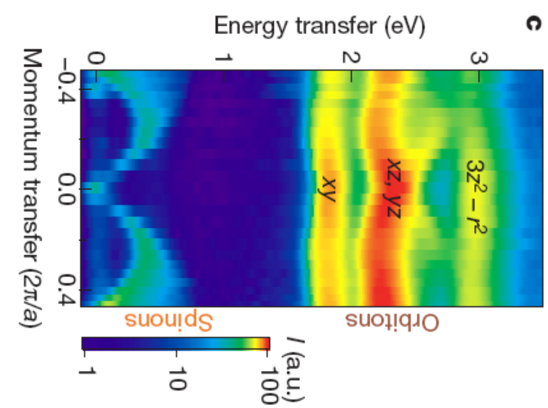
- ***High-resolution*** RIXS probes critical collective charge modes (element specific)
- ***Dynamic*** RIXS reveals response of collective modes to control fields and tailored excitations (60 fs ⇔ 30 meV)
  - ***- light-induced superconductivity***
  - ***- vibrational-driven insulator to metal transitions***
- Hierarchy of collective excitations in cuprates can be seen in energy-momentum domain.



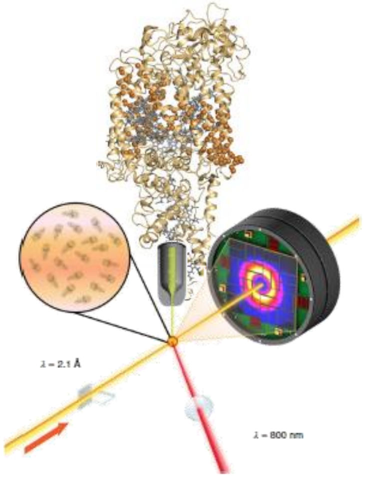


Nature 485, 82 (2012).

Life Sciences & Matter in Extreme Conditions at LCLS-II

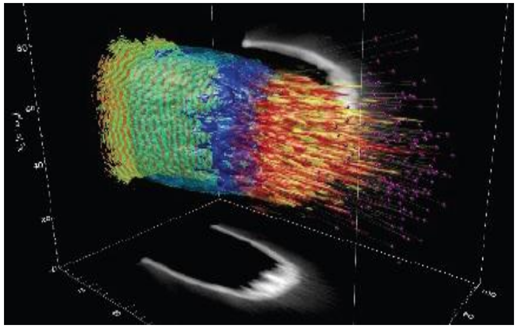
Life Science

- Small-scale structural dynamics at Å resolution
  - Serial nano-crystallography
- Large scale conformational dynamics
  - Molecular movies – single particle imaging (2-6 keV)
  - Solution scattering – fluctuation SAX



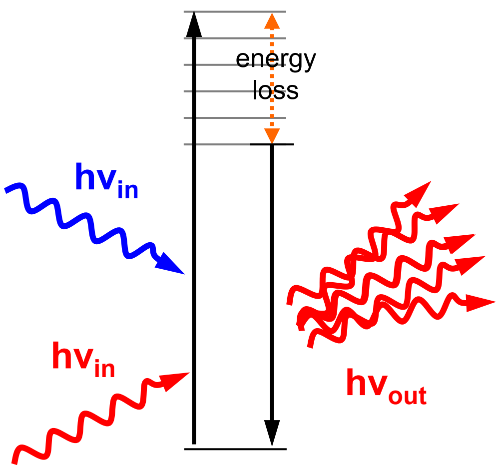
Matter in Extreme Conditions

- Warm & hot dense matter – lab. astrophysics
- Rapid compression, shock & impact physics
- Material weakening and hydrodynamic "flow" on ultrafast time scales



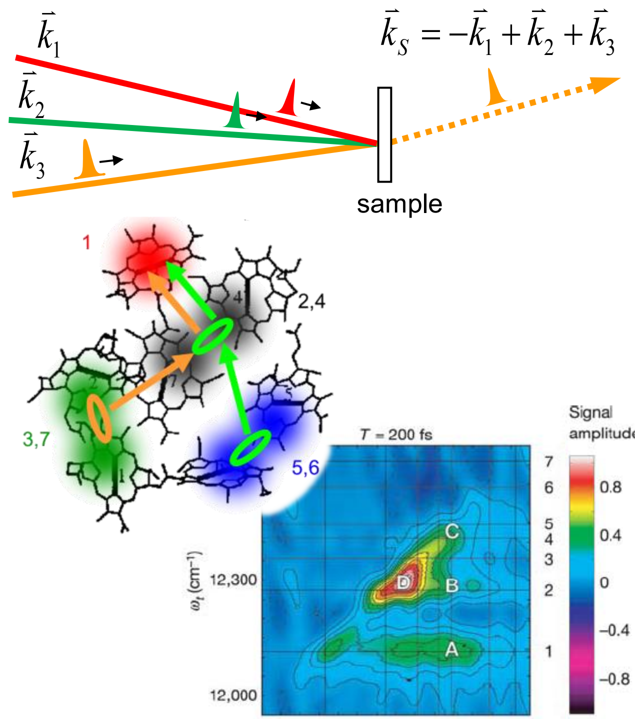
LCLS-II will enable completely new x-ray methods

Time-resolved X-ray Raman, stimulated emission



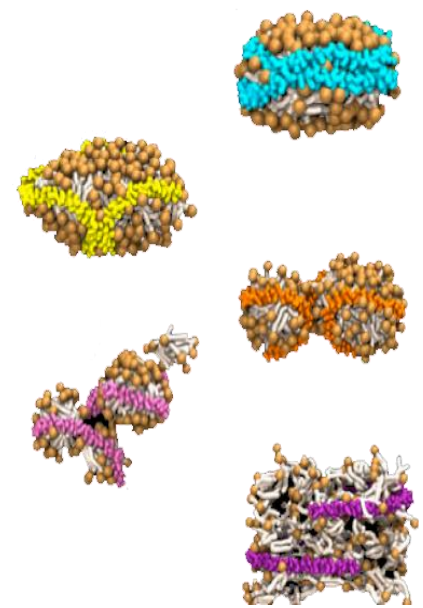
X-ray emission spectrum

Multi-dimensional nonlinear spectroscopy



Pump-probe

Macromolecular assembly & dynamics



Structure of single molecules

This was just a taste of what LCLS-II will enable

Chemistry

- ❑ Fundamental charge migration & redistribution

Dynamic reaction microscope	High rep rate
Stimulated X-ray Emission Spec.	Coherence (few fs), 2-color
Photoemission	Soft, tender X-rays
X-ray scattering	Hard X-rays

- ❑ Predictive understanding of photo-catalysis

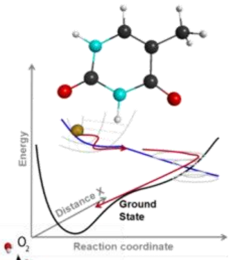
Time-resolved X-ray Raman	High rep rate
(X-ray absorption/emission)	Soft, tender X-rays
Time-resolved photoemission	Coherence (FT limit), 2-color

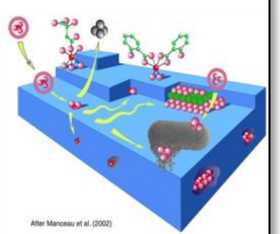
- ❑ Heterogeneous catalysis - in real time & *operando*

Time-resolved photoemission	High rep rate
(ambient pressure)	Soft, tender X-rays
Res. coherent X-ray scattering	Hard X-rays + soft X-rays

- ❑ Combustion & aerosol chemistry

Flash tomography	High rep rate
Stimulated X-ray Emission Spec.	Soft, tender X-rays
Coherent X-ray scattering	2-color





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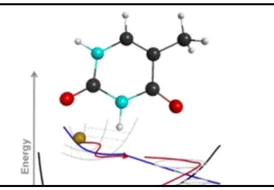
Chemistry



- ❑ Fundamental charge migration & redistribution

Dynamic reaction microscope Stimulated X-ray Emission Spec. Photoemission X-ray scattering	High rep rate Coherence (few fs), 2-color Soft, tender X-rays

- ❑ Predictive understar

Time-resolved X-ray Ram
(X-ray absorption/emissio
Time-resolved photoemis

- ❑ Heterogeneous cata

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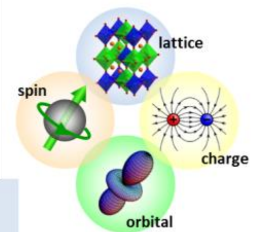
Res. coherent X-ray scatt

- ❑ Combustion & aeros

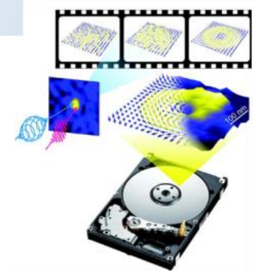
Flash tomography
Stimulated X-ray Emission
Coherent X-ray scattering

Materials Physics



- ❑ Understand & control emergent phenomena in quantum systems with interacting degrees of freedom

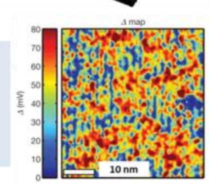
Time & momentum resolved X-ray Raman Time-resolved hard X-ray photoemission Time-, spin-, imaging- photoemission	High rep rate Soft, tender, hard (3ω) X-rays Coherence (FT limit)



- ❑ Understand & control nonequilibrium spin states at fundamental length & time scales

Time-resolved X-ray dichroism Coherent, resonant scattering Hard X-ray photoemission (time/spin)	High rep rate Soft, hard (3ω) X-rays Polarization

- ❑ Nanoscale heterogeneity, fluctuations, & Dynamics



X-ray photon correlation (XPCS) X-ray scattering THz pump/X-ray probe	High rep rate Soft, tender, hard (ω, 3ω) X-rays Programmable pulse sequences

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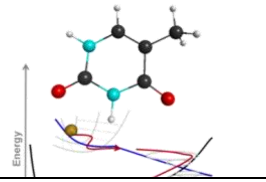
SLAC

Chemistry



- ❑ Fundamental charge migration & redistribution

Dynamic reaction microscope	High rep rate
Stimulated X-ray Emission Spec.	Coherence (few fs), 2-color
Photoemission	Soft, tender X-rays
X-ray scattering	

- ❑ Predictive understar

Time-resolved X-ray Rama
(X-ray absorption/emissio
Time-resolved photoemis

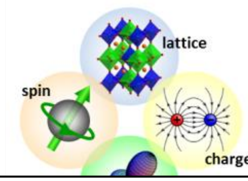
- ❑ Heterogeneous cata

Time-resolved photoemis
(ambient pressure)
Res. coherent X-ray scatt

- ❑ Combustion & aeros

Flash tomography
Stimulated X-ray Emission
Coherent X-ray scattering

Materials Physics



- ❑ Understand & control emergent phenomena in quantum systems with interacting degrees of freedom

Time & momentum resolved X-ray R
Time-resolved hard X-ray photoemis
Time-, spin-, imaging- photoemission

- ❑ Understand & control nonequ at fundamental length & time

Time-resolved X-ray dichroism
Coherent, resonant scattering
Hard X-ray photoemission (time/spi

- ❑ Nanoscale heterogeneity, flu

X-ray photon correlation (XPCS)
X-ray scattering
THz pump/X-ray probe

Life Sciences

- ❑ Understanding the dynamics of biological complexes & molecular machines
  - In physiological environments & on natural time scales
    structure alone provides limited insight to biological function
- ❑ Small-scale structural dynamics at Å resolution

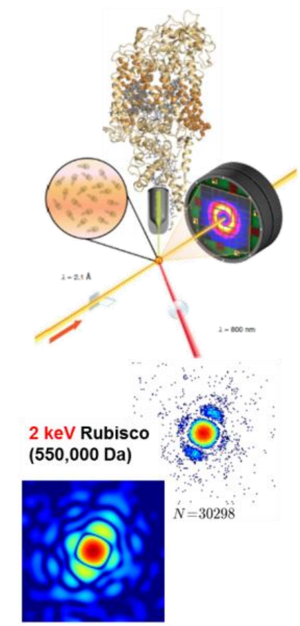
Anomalous phasing (Se – 12.5 keV)	Hard X-rays
Native phasing (S – 2.5 keV)	2-color
Resonant scattering (Na, Mg, P, Cl)	Tender X-rays

- ❑ Large scale conformational dynamics

Single-particle imaging	Tender X-rays
• Single-shot	High intensity (>5 mJ/pulse)
• Multi-shot	High rep rate
Fluctuation SAXS	High rep rate, tender X-rays

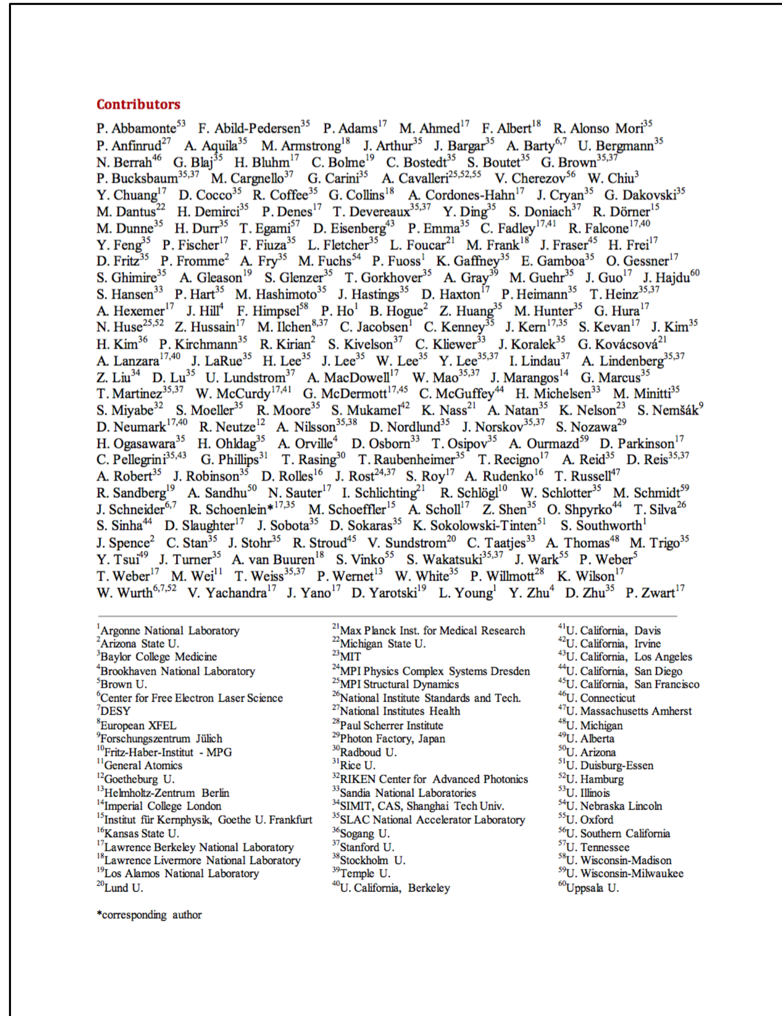
- ❑ Electronic structure and biological function

Time-resolved RIXS	High rep rate
Time-resolved XES, XAS	Soft, tender, hard X-rays
X-ray scattering	Coherence (FT limit), 2-color



LCLS-II Science Opportunities Document

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



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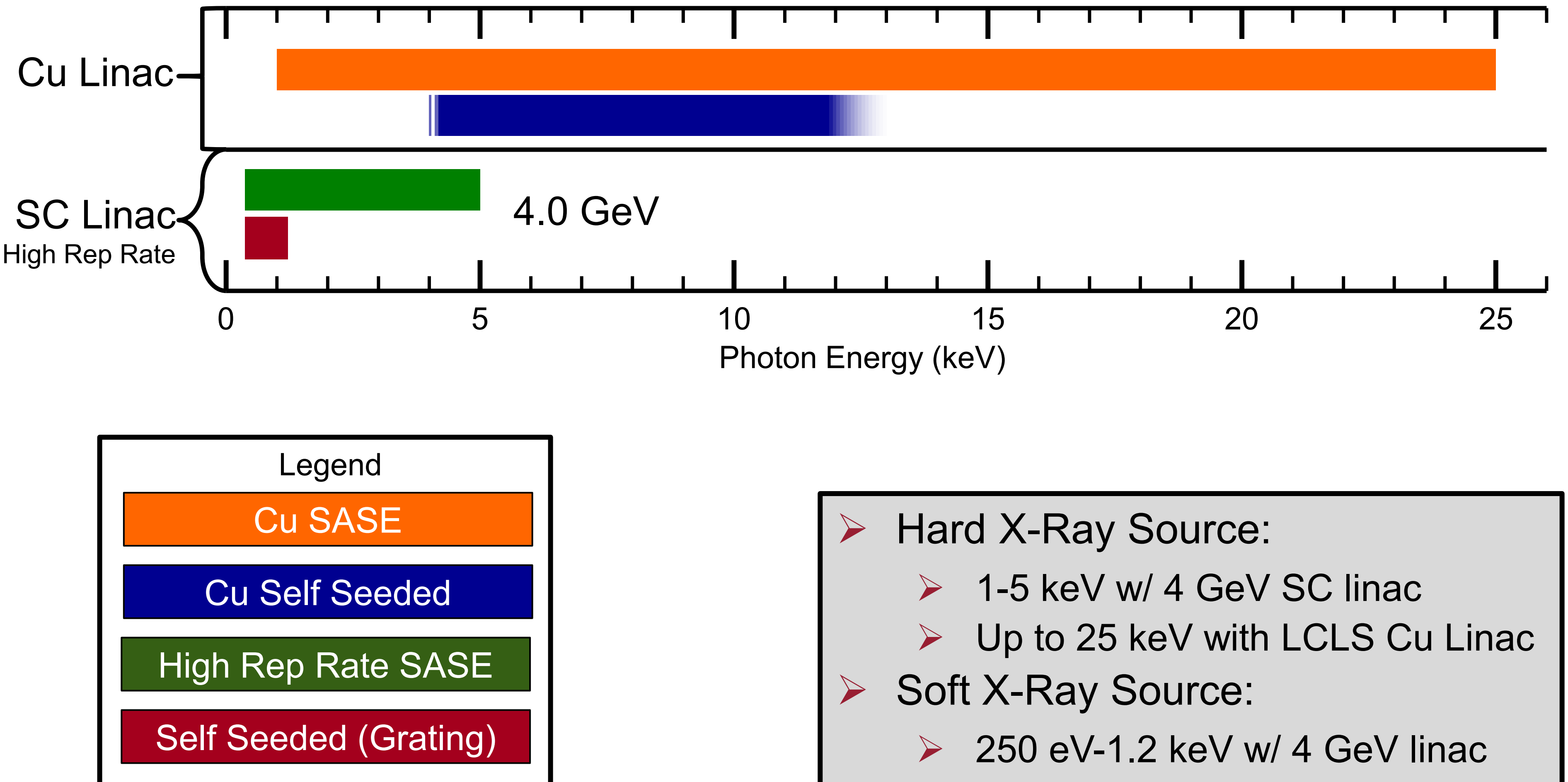
High Repetition Rate Revolution

- LCLS-II upgrade will deliver
  - High repetition rate → 10^4 fold increase in data collection
  - High stability → high throughput measurements
  - Second source capable of multiplexing → doubles access
- New Scientific Opportunities at LCLS-II
  - Photo and heterogeneous catalysis
  - Follow molecular transformations & bond formation
  - Revealing interacting degrees of freedom in correlated electron systems

The Linac Coherent Light Source (LCLS), SLAC National Accelerator Laboratory, is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515.

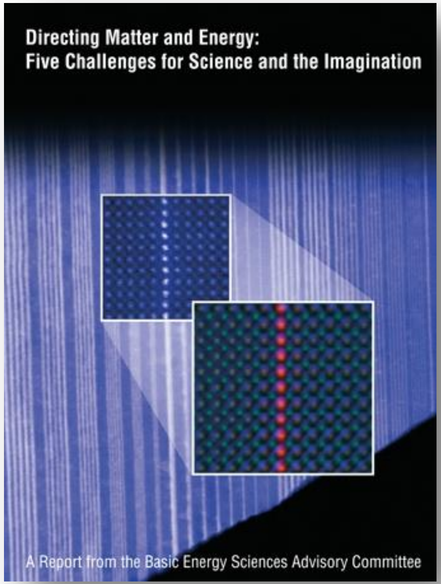
LCLS-II Operating Energy Range



- Hard X-Ray Source:
  - 1-5 keV w/ 4 GeV SC linac
  - Up to 25 keV with LCLS Cu Linac
- Soft X-Ray Source:
  - 250 eV-1.2 keV w/ 4 GeV linac
    - 200 eV requires <4 GeV

Guiding Scientific Themes

Five Grand Challenges for Science and the Imagination (2007)



Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science (2015)