Science opportunities with the upgraded LCLS

Mike Dunne, LCLS Director

BESAC, December 2023

U.S. DEPARTMENT OF ENERGY

Stanford University

LCLS Linac Coherent Light Source

SLAC NATIONAL ACCELERATOR LABORATORY

Today's panel discussion

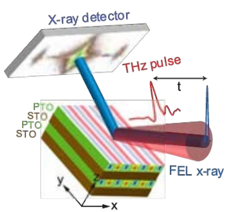
• Mike Dunne (SLAC)	Context: The LCLS facility and the LCLS-II Project	10 mins
• Toni Taylor (LANL)	Scientific Impact of LCLS	10 mins
• Daniel Rolles (KSU)	AMO Physics and Gas-Phase Chemistry	10 mins
• Junko Yano (LBNL)	Condensed Phase Chemistry and Catalysis	10 mins
• Matteo Mitrano (Harvard)	Quantum Materials Science	10 mins
• Q&A / discussion with the panel		40 mins

SLAC

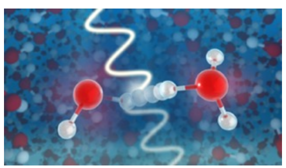
LCLS has been incredibly successful, and is driving a transformation in X-ray science

LCLS performance has shown where coherence, ultrafast pulses, and extreme brightness are most impactful.

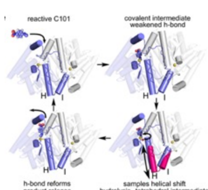
Breakthrough scientific results



Collective dynamics in complex materials

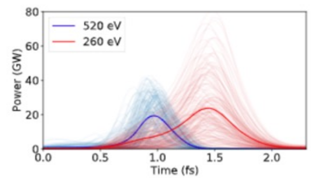


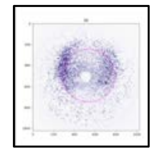
Chemical bond initiation and evolution



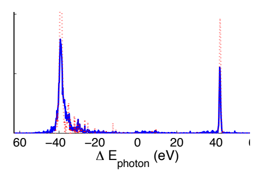
Molecular movies of bio-enzymes

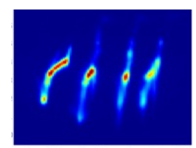
Rapid development of capability



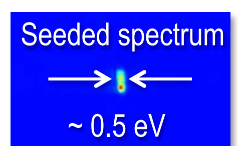


1000x reduction in pulse duration (to 200 as)



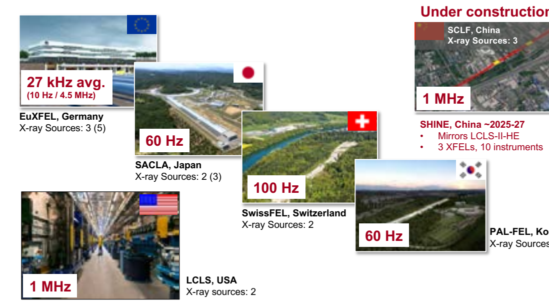


Element-specific dynamics (multi-color, multi-pulse)

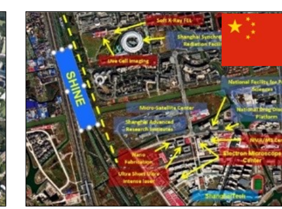


Transform-limited information (temporal coherence)

International response



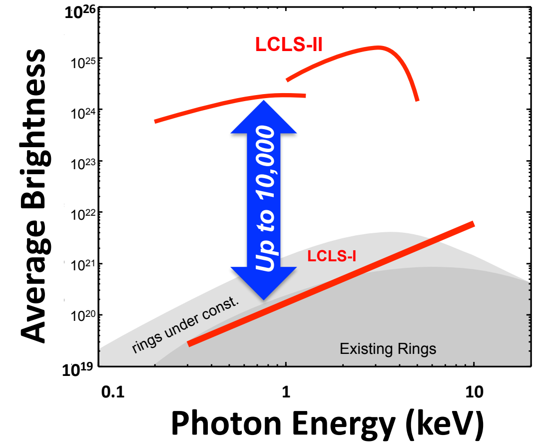
Emergence of XFELs around the world

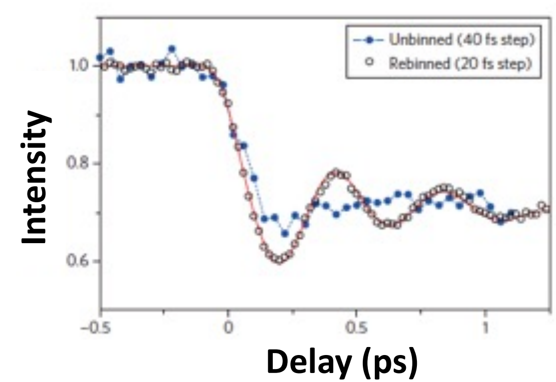


Investments in XFEL science and people

LCLS has driven major advances, but has important limitations

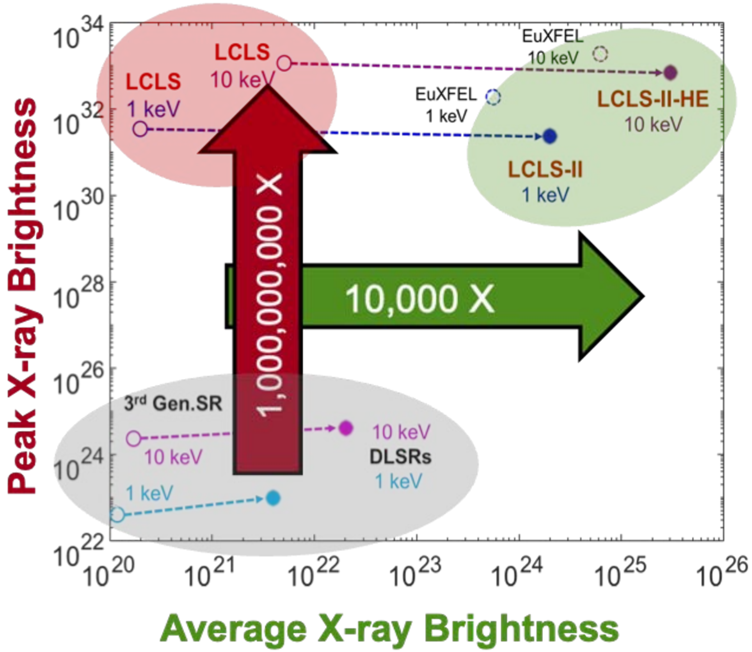
- **Low rep-rate requires high concentration samples, and limited number of time points in 'molecular movie'**
- **Incredible peak brightness, but "conventional" average power**
- **Poor stability of Cu Linac (spatial, Δt, energy)**
- **Inability to meet community demand (single undulator)**

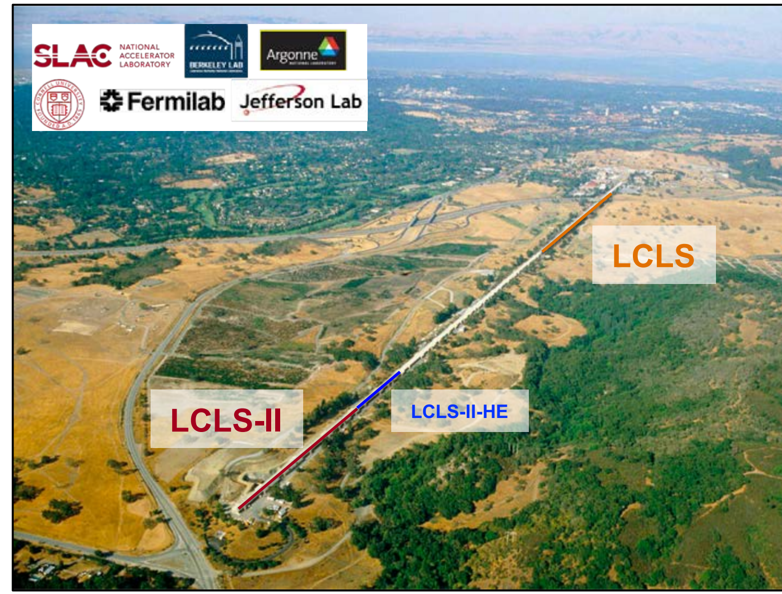




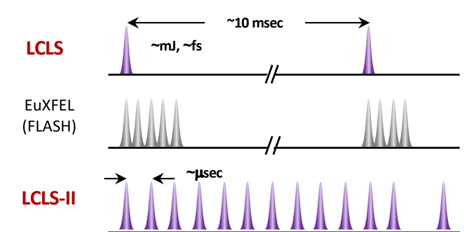
The LCLS facility development plan, thanks to major DOE investments, provides an internationally leading suite of capabilities for our users

LCLS-II delivers a dramatic step in capability, responding to the 2013 BESAC recommendation





From 120 Hz to 1 MHz CW



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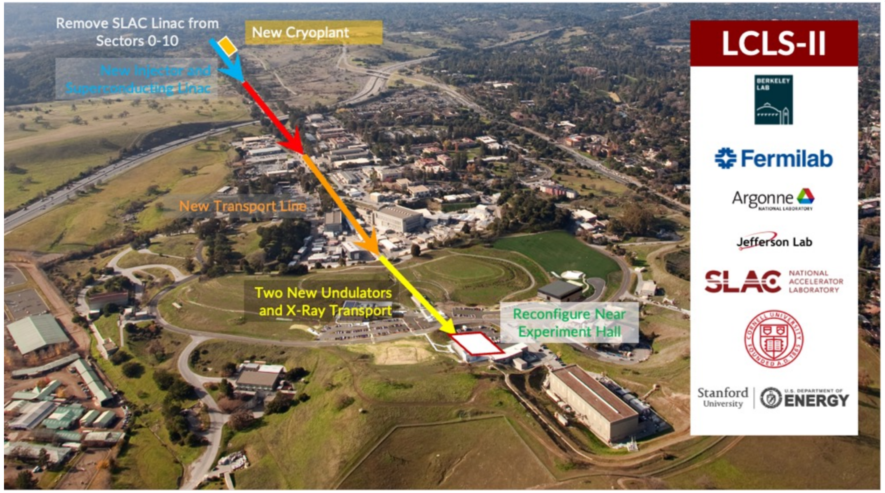
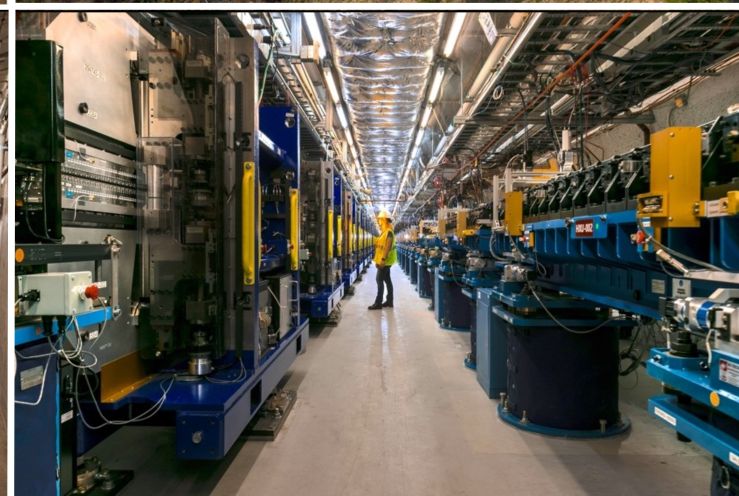
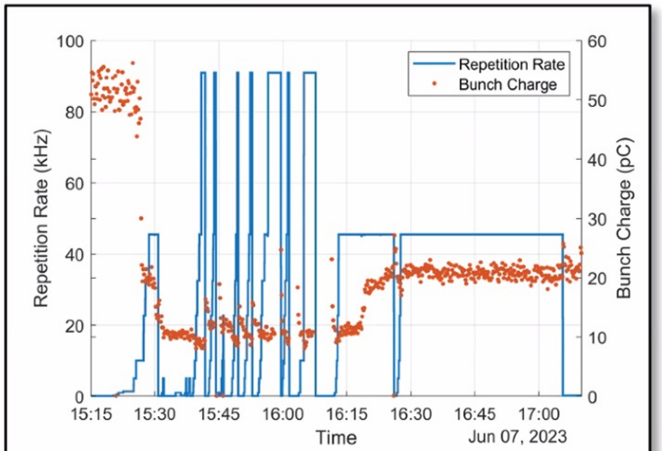
LCLS-II: A remarkable journey over 10 years, >$1 billion



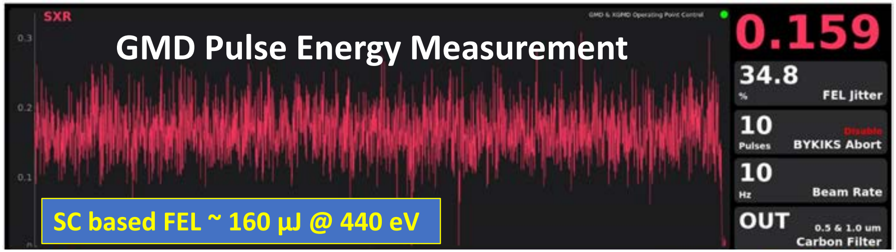


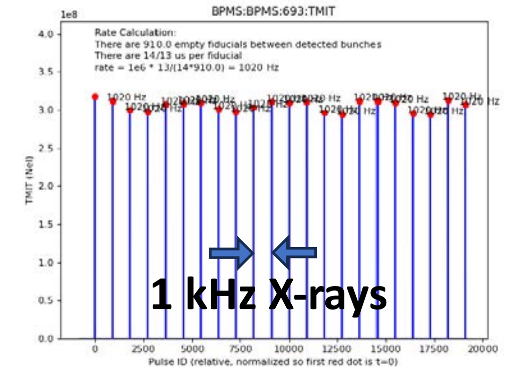
World's first "Superconducting CW X-Rays" 23 Aug 2023

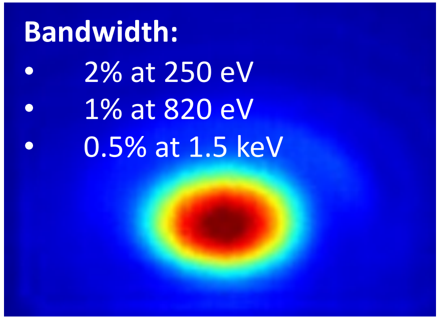
SCRF linac commissioned to 93 kHz



X-ray lasing from 0.25 to 3.8 keV at up to 1 kHz







Key characteristics: High repetition rate, stable beam, tunable undulators

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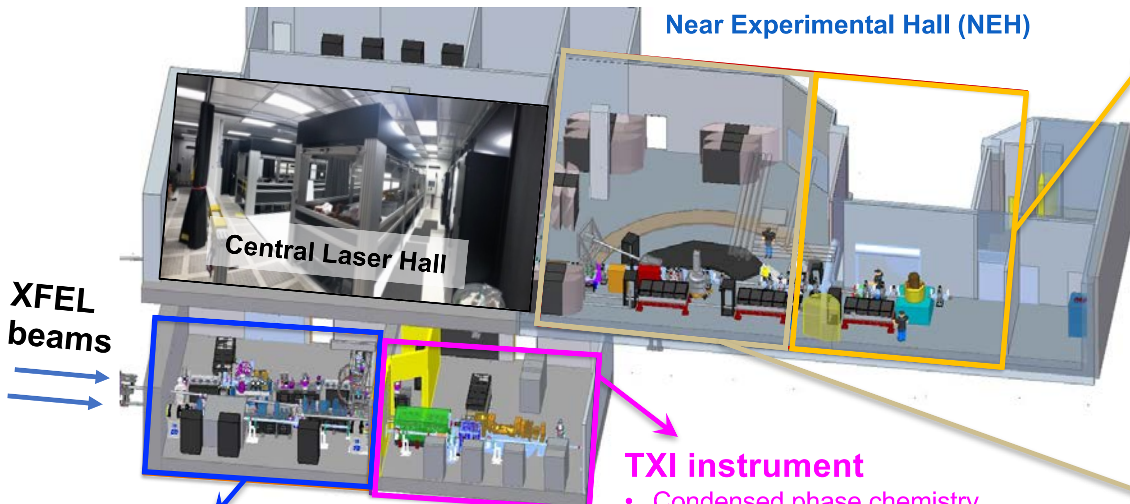
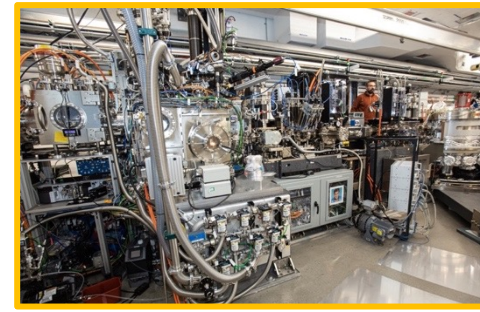
First Light celebration at SLAC, September 2023

SLAC

Extensive community engagement defined a suite of four instruments with 11 endstations to use the new LCLS-II capabilities



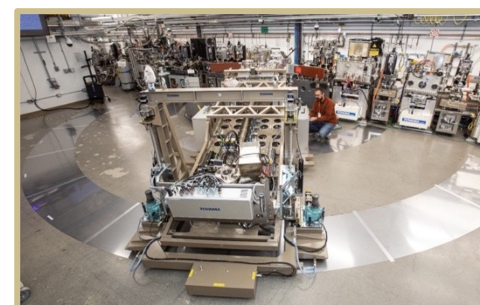
ChemRIXS instrument
- Photo-catalysis
- Surface chemistry



TXI instrument
- Condensed phase chemistry
- Nano-imaging of complex dynamics
- **Dual XFEL beams** for nonlinear science

TMO instrument
- AMO science

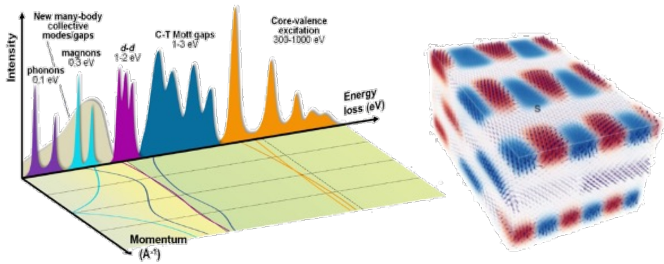
qRIXS instrument
- Quantum materials



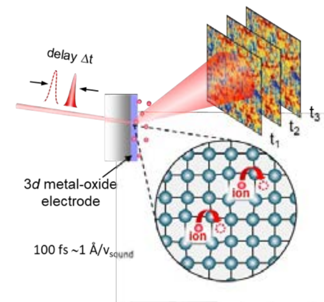
SLAC

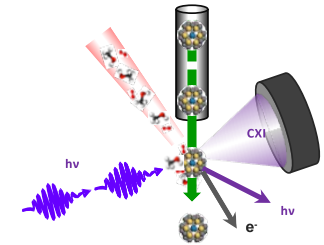
The upgraded flexibility of LCLS serves a broad user community

Quantum materials need **extreme spectral brightness, transform limited pulses, THz**

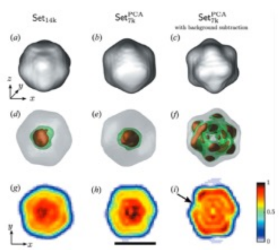


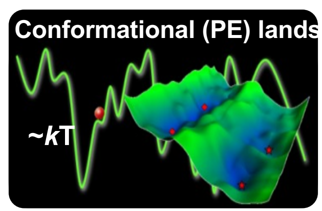
Sustainability and material design need controlled **multi-pulses, MHz rates**



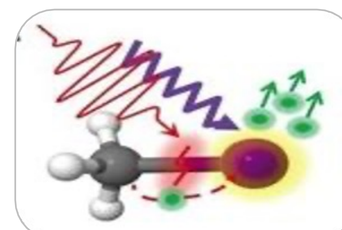


Bio- and Nano- science (e.g., SPI) need **high power (TWs), <10 fs, high rep. rate**

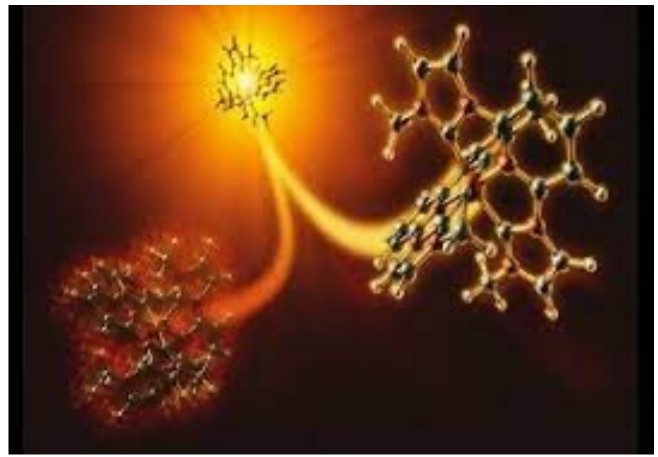




AMO and quantum coherence studies need **attosecond durations, multi-color, rapid tuning**



Scientific Impact of LCLS



Toni Taylor
Los Alamos National Laboratory

Presentation to BESAC, December 12, 2023

Science impact assessment of LCLS (2023)

Background: DOE feedback from the 2022 SLAC Annual Lab Plan

"With the operation of LCLS reaching the decade mark, it is critical for SLAC to evaluate the overall scientific contributions to the various key disciplines supported by LCLS.

This is to ensure that the significant investments in LCLS and its upgrades deliver the maximum scientific and technical impacts. The evaluation will help inform future operations and define an expansion strategy for the next decade."

Panel:

- Antoinette (Toni) Taylor, Los Alamos National Laboratory (Chair)
- Paul Evans, Department of Materials Science and Engineering, University of Wisconsin-Madison
- Claudio Masciovecchio, FERMI FEL, Elettra Sincrotrone Trieste Laboratory
- James McCusker, Department of Chemistry, Michigan State University
- Janet Smith, Department of Biological Chemistry and Life Sciences, University of Michigan
- Marc Vrakking, Max-Born-Institute and the Freie Universität Berlin
- Justin Wark, Department of Physics, University of Oxford
- Philippe Wernet, Department of Physics and Astronomy, Uppsala Universitet

Science impact assessment - Charge

Committee charge

- Identify the key scientific and technical achievements from LCLS research
- Articulate impact in a manner that is accessible to the broader community of scientific, political, policy, and public stakeholders
- Describe how these achievements have been transformative within their field, or beyond
- Briefly outline future opportunities that LCLS can address, including high-level indications of how FEL science must advance in order to address these opportunities

Considerations

- Account for the expected gestation time for science results from novel XFEL technology
- Method development has been a significant focus of the first 10 years of XFELs. Important to identify a small number of the most important achievements in this area.
- In addition to wide-ranging discovery research, the assessment should link to key aspects of the DOE mission, e.g. clean energy, environment, climate change, sustainability, and national security

Source Materials from LCLS

- ❑ Publication record (curated, high-impact)
- ❑ Previous studies of LCLS science impact
  e.g. 5-yr. Rev. Mod. Phys., 10 yr. Anniversary etc.
- ❑ LCLS Science & Instrumentation reviews
- ❑ DOE-BES Triennial Review materials
- ❑ Targeted workshops
  e.g. Solar Energy Photochem., Gas-phase Chemistry
- ❑ Benchmark comparisons
  - Other XFEL facilities
  - Development arc of similar emerging fields
    e.g. synchrotron science, laser science
  - LCLS impact on BES Grand Challenges

LCLS Science Impact Report

December 2022

Table of Contents

LCLS New Methods and links to Science Impact

In the following, we highlight some of the key new methods that underpin the science impact of LCLS. The "new methods" outlined here span a broad range, including:

- Well-established X-ray scattering and spectroscopy methods that have been adapted for the first time at LCLS to open transformative new classes of time-resolved studies that are only possible with LCLS (or similar XFEL) capabilities.
- Entirely new experimental methods inspired by the unique capabilities of LCLS (and similar XFELs).
- Novel XFEL operating modes, diagnostics, and related developments that enable and inspire new experimental approaches with significant projected science impact.

Review: top level summary

“Given the transformative nature of LCLS, scientific results from LCLS have led to ***qualitative advances over a broad cross section*** *of scientific fields …*

LCLS is to be credited with ***many significant scientific discoveries*** *that have natural ties to very specific, applied problems in energy, biology, materials, and chemical sciences …*

… the importance of the fundamental new knowledge *that the work at LCLS has created – knowledge that will provide the foundation for transformational advances both seen and unseen at the present time –* ***should not be underestimated*** *…*

LCLS has ***created new areas of science*** *through the development of entirely new experimental methods inspired by its unique capabilities.”*

Highlighted the enabling steps taken by LCLS :
- Sustained investment in R&D (targeted, strategic)
- Scientific Campaigns to address grand challenge questions
- Enhanced capacity (multiplexing, standard configurations, PCS)
- Nurturing and sustaining expert staff
- Close partnerships with Stanford


LCLS Science Impact Assessment
February 2023

Submitted to Provost Persis Drell, Interim SLAC Director Stephen Streiffer, and the SLAC Scientific Programs Committee

Submitted by the LCLS Science Impact Assessment Committee members:
Antoinette (Toni) Taylor, Los Alamos National Laboratory (Chair)
Paul Evans, Department of Materials Science and Engineering, University of Wisconsin-Madison
Claudio Masciovecchio, FERMI FEL, Elettra Sincrotrone Trieste Laboratory
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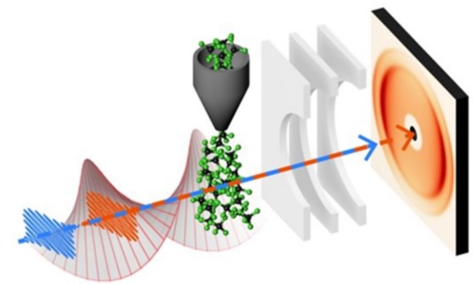
Justin Wark, Department of Physics, University of Oxford
Philippe Wernet, Department of Physics and Astronomy, Uppsala Universitet

Review – detailed findings (1/2)

AMO science:

Major breakthrough in attosecond science. Pioneering research on charge dynamics with elemental specificity.

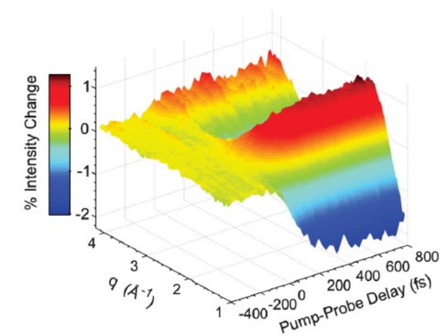
- *Examples*: X-ray induced transparency of Aluminum, creation of hollow atoms, and observation of non-sequential ionization (nonlinear X-ray science). Direct observations of coherent electron motion during Auger-Meitner decay.
- ***Future***: *"For the foreseeable future… expect LCLS to be the leading facility worldwide"*



Gas-phase chemistry:

Direct observables separating structural and electronic effects, with sub-Angstrom and femtosecond resolution, revolutionizing our understanding of ultrafast chemical transformations

- *Example*: Ring opening reaction dynamics of organic molecules, modeling photobiology relevant to vitamin-D, proving that UV excitation leads to an opening of the aromatic ring – via conical intersection on ~100fs timescale
- *Future:* Molecular-frame (CEI) imaging to reveal full 3D molecular structure evolution

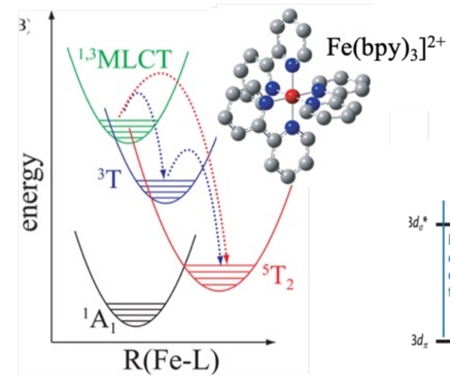


Review – detailed findings (2/2)

Condensed-phase chemistry and catalysis:

"LCLS has made a number of seminal and paradigm-shifting discoveries … provides the foundation to translate fundamental understanding of chemical systems to address key challenges for a sustainable future"

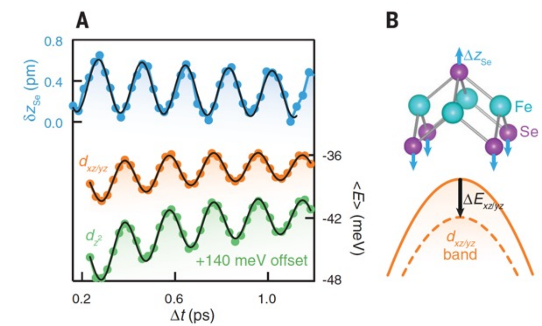
- *Example*: Revealed the coupling of structure, spin states, and reactivity via evolution of frontier orbitals in transition metal complexes – driving studies in photoredox catalysis and solar cell technology
- ***Future***: Complete studies of functional complexes in operating environments



Materials science and condensed matter physics:

Understanding and control of complex materials with coupled order parameters, and properties mediated by heterogeneity, fluctuations and disorder at the nanoscale

- *Example*: Understanding the interactions that lead to the emergence of superconductivity: the enhancement of electron –phonon coupling strength via correlation effect in Fe-based superconductors.
- ***Future***: Advanced scattering methods (diffuse, coherent, FT-IXS) → complex/quantum & applied materials

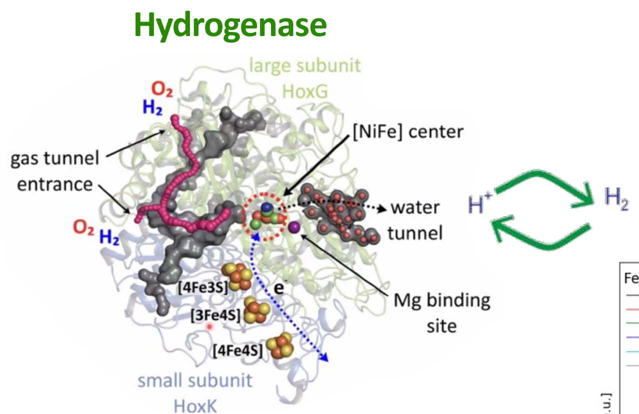


Enzyme Catalysis for Clean Energy

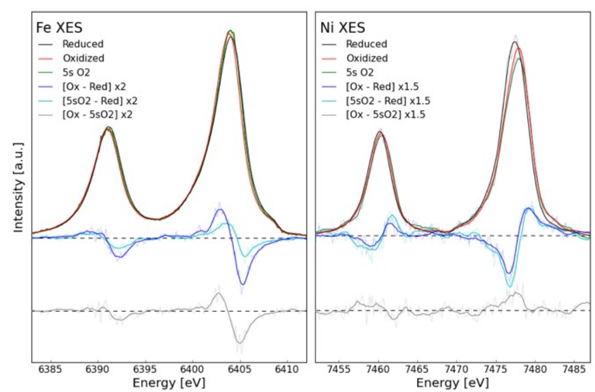
LCLS Scientific Campaign
J. Kern et al.

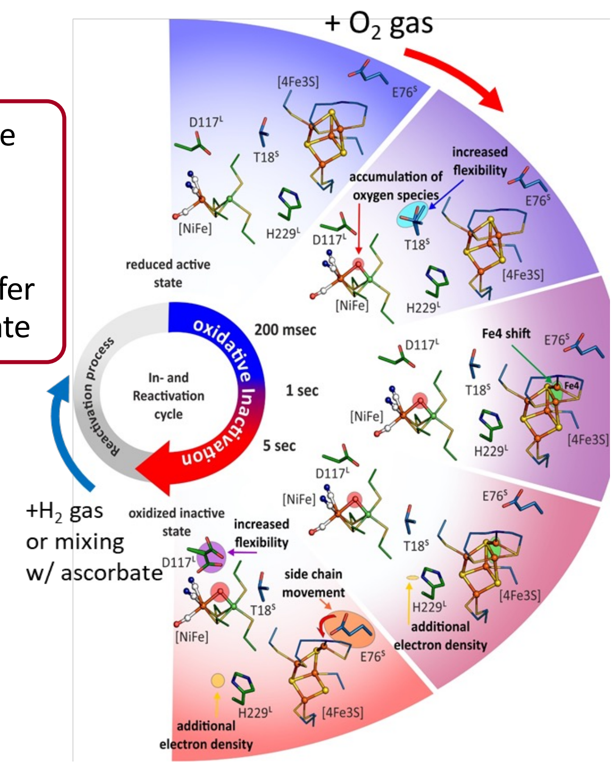
Link atomic structural dynamics with chemical/electronic dynamics of metalloenzymes that catalyze reactions relevant for the energy economy

Multimodal (scattering + spectroscopy), *Operando*



- Clear stepwise change at Ni/Fe site first followed by proximal cluster
- XES dynamics of Ni and Fe differ, matching structural observations
- Thr18 may influence proton transfer during inactivation to form Ni_B state

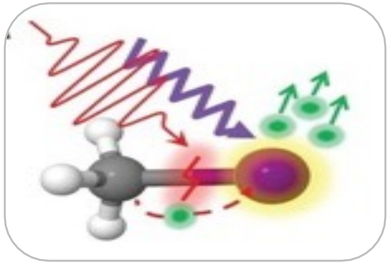




LCLS-II will transform our understanding of dynamics in real-world materials and chemical science systems

Charge dynamics on fundamental timescales

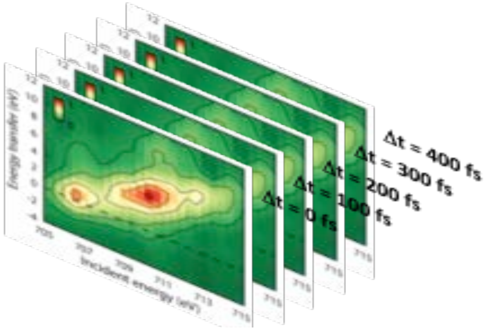
- Reveal coupled electronic and nuclear motion in molecules
- Capture the initiating events of charge transfer chemistry with sub-fs resolution



Ultrafast

Molecular dynamics with exquisite resolution

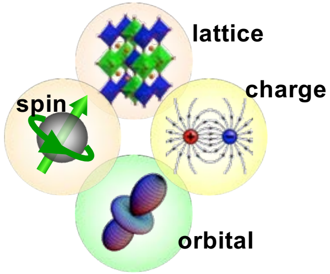
- Measure element-specific, local chemical structure and bonding
- Study efficient, robust, selective photo-catalysts



High repetition rate

Emergent phenomena in quantum materials

- Connect spontaneous fluctuations, dynamics and heterogeneities on multiple length- and time- scales to bulk material properties
- Study interacting degrees of freedom (e.g. unconventional superconductors)



Extreme brightness

LCLS-II represents a substantial leap in performance for ultrafast and precision science

Atomic, Molecular, and Optical Science & Gas-Phase Chemistry

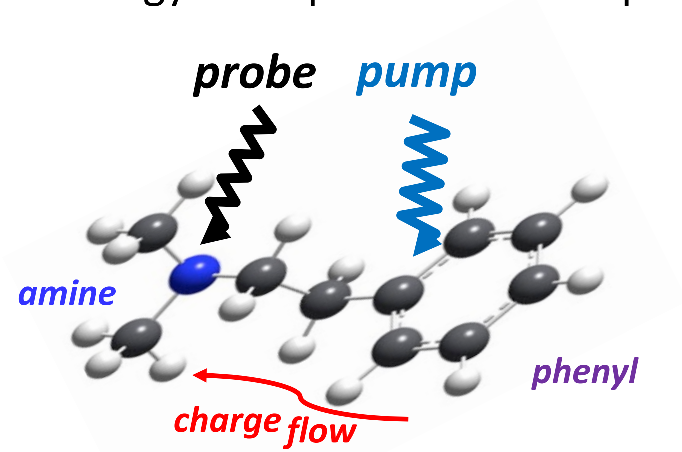
Daniel Rolles

J. R. Macdonald Laboratory
Kansas State University

Challenges and Opportunities in AMOS & Gas-Phase Chemistry

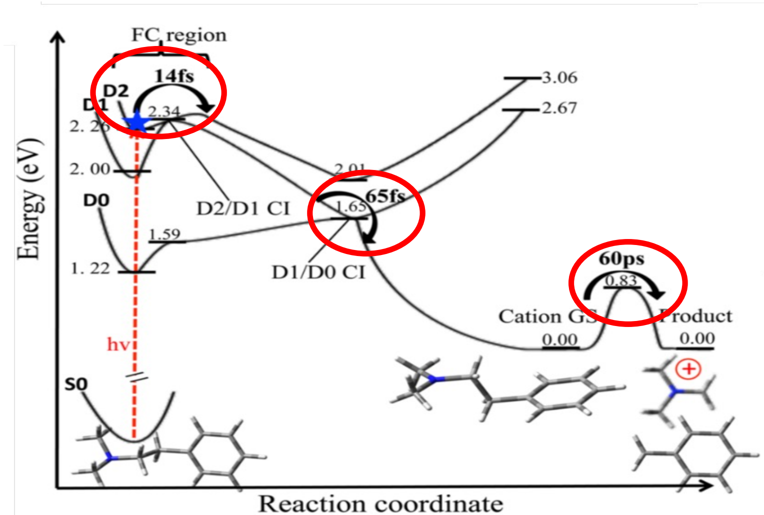
Probing charge and energy flow at the molecular level

Example: energy absorption at chromophore leads to charge & energy flow across the molecule



2-phenylethyl-N,N-dimethylamine (PENNA)

prototype system for charge transfer in peptides



Grand Challenges*

- How do we control matter at the level of electrons & atoms?
- How do we characterize and control matter away from equilibrium?
- How do we design and synthesize matter with tailored properties?

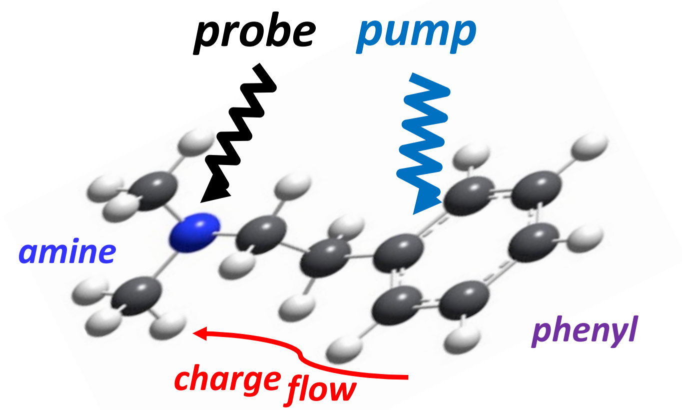
Only LCLS-II can interrogate these processes on natural time and length scales

*BES Science Grand Challenges and Transformational Opportunities Reports

Challenges and Opportunities in AMOS & Gas-Phase Chemistry

Probing charge and energy flow at the molecular level

Example: energy absorption at chromophore leads to charge & energy flow across the molecule



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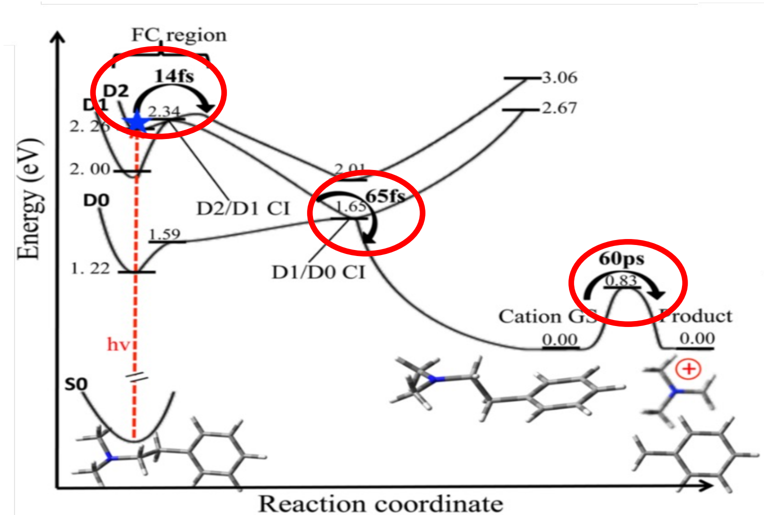


Technical Opportunities of LCLS-II:

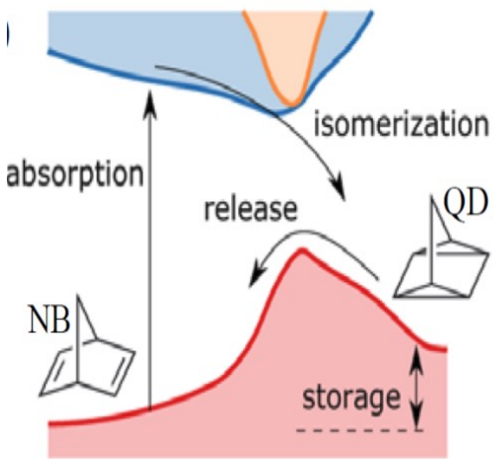
- Ability to generate **few-femtosecond to attosecond, multicolor X-ray pulse pairs**
- Visualize **charge flow** in molecules on the **attosecond timescale** (2023 Nobel Prize)
- Time-resolved X-ray photoelectron spectroscopy: **element and site specificity**; sensitivity to both electronic and atomic/nuclear structure and dynamics in the direct vicinity of specific molecular sites
- **High repetition rate** enables measurements with weak contrast and allows more differential measurements (angle resolved & molecular-frame measurements *via* coincidences with ions)

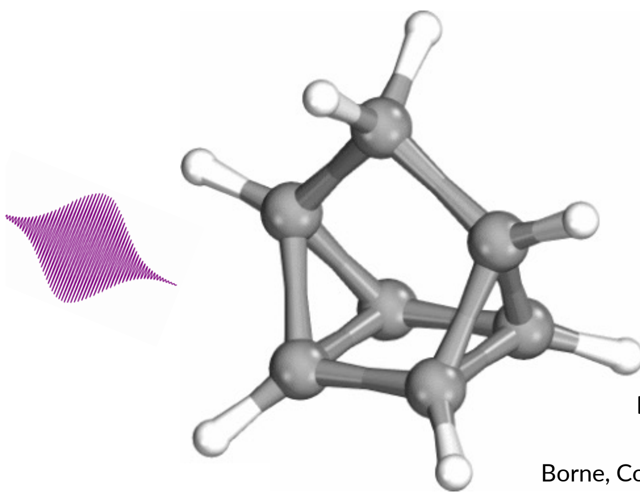
Challenges and Opportunities in AMOS & Gas-Phase Chemistry

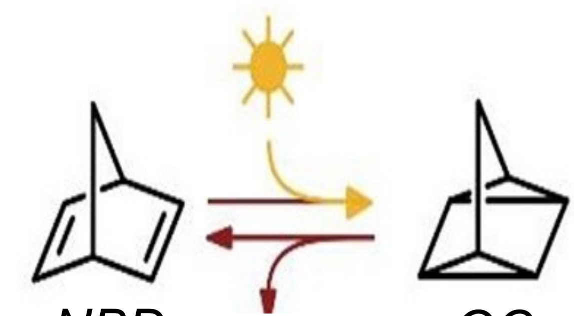
Imaging molecular rearrangement during chemical reactions (bond breaking and bond formation; motion of electrons & atoms)

Example: Interconversion reaction of QC and NBD – proposed, e.g., for solar energy storage

- Highly strained 3-member rings in QC hold a lot of energy
- Could store usable energy that would be released with the reconversion to norbornadiene
- Was also considered as possible rocket fuel by US Airforce







NBD
norbornadiene

QC
quadricyclane

Borne, Cooper, DR, et al. Nat. Chem. (in press)

Only LCLS-II can interrogate these processes on natural time and length scales

Information on **structural dynamics** *via* several **complementary imaging techniques**:
ultrafast X-ray & electron diffraction; highly correlated 3-D information via **Coulomb explosion imaging**

Understanding Complex Photochemical Reaction Landscapes

Scientific Opportunity

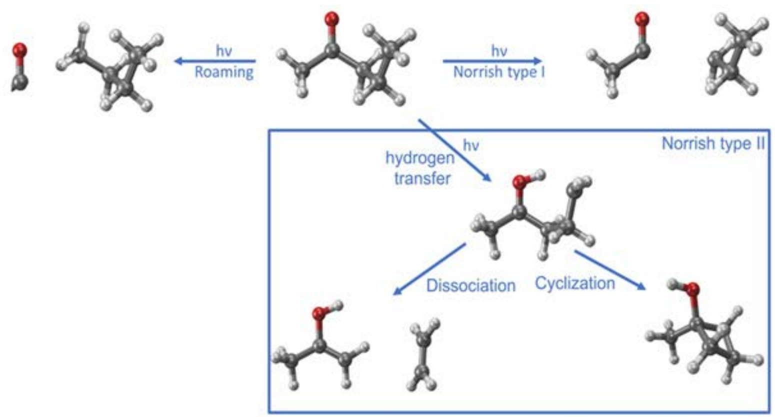
- Detailed mapping of complex photochemical reaction networks involving multiple parallel and rare reaction pathways

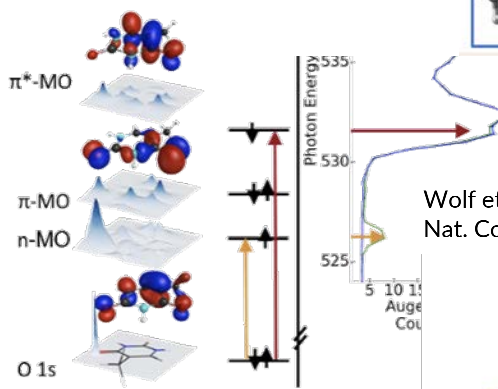
Significance and Impact

- Benchmark high-level quantum chemical simulations
- Establish photochemical design principles, a current major knowledge gap

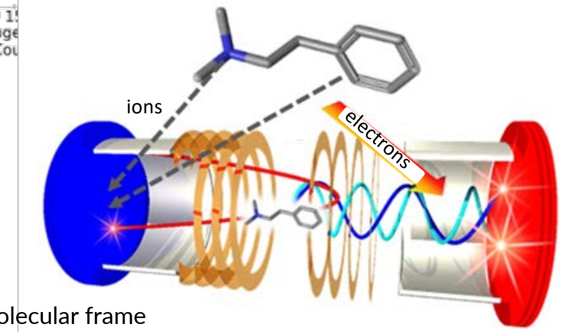
LCLS-II Approach

- Coincidence spectroscopy in molecular reaction microscope disentangles reaction pathways
- Exploit high repetition rate (capture rare events), and high intensity of LCLS-II soft X-rays, with element and site specificity





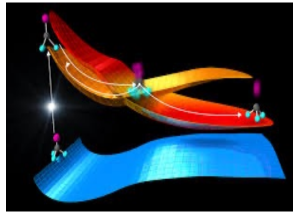
Wolf et al.
Nat. Commun. 8, 29 (2017)



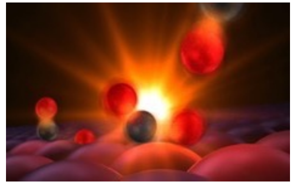
Molecular Reaction Microscope
– reconstruct reactions in the molecular frame

LCLS-II high repetition rate captures rare events via coincidence spectroscopy

Summary: Challenges and Opportunities in AMOS & Gas-Phase Chemistry

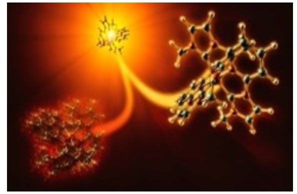


- ✓ **Probing charge and energy flow at the molecular level**
- ✓ **Imaging molecular rearrangement during chemical reactions**
- ✓ **Disentangling complex photochemical reaction landscapes**



BES Grand Challenges:

- How do we control matter at the level of electrons & atoms?
- How do we characterize and control matter away from equilibrium?
- How do we design and synthesize matter with tailored properties?



AMO Science Contributions & Objectives:

- *Gas-phase / single-molecule studies allow for quantitative comparison to theory (benchmarking quantum chemistry calculations)*
- *Fundamental measurements in model quantum systems drive advances in predictive theory*
- *Detailed understanding of the coupling between electronic and nuclear degrees of freedom*
- *Long-term goal: influence & control charge and energy flow and chemical transformations at the level of atoms and electrons.*
- *Synthesis of molecules and materials with required functionality*

Condensed Phase Chemistry and Catalysis

Junko Yano

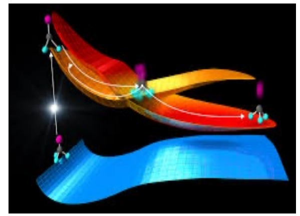
Lawrence Berkeley National Laboratory

Directed chemical reactivity for sustainability:

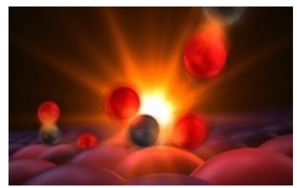
Energy efficient ♦ Chemically Selective ♦ Durable ♦ Earth-abundant

Grand Challenges*

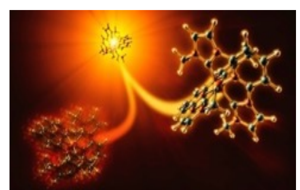
- ❑ How do we design and synthesize matter with *tailored properties*?



- ❑ How do we characterize and control matter *away from equilibrium*?



- ❑ How do we control matter at *the level of electrons & atoms*?



Chemical Science Objectives

- ➢ *To achieve a deeper fundamental understanding of chemical reactivity*
- ➢ *To exploit nonequilibrium dynamics to direct chemical outcomes*
- ➢ *To develop design principles for synthesis of molecules and materials with required functionality*

Only LCLS-II can interrogate these processes on natural time and length scales

*BES Science Grand Challenges and Transformational Opportunities Reports

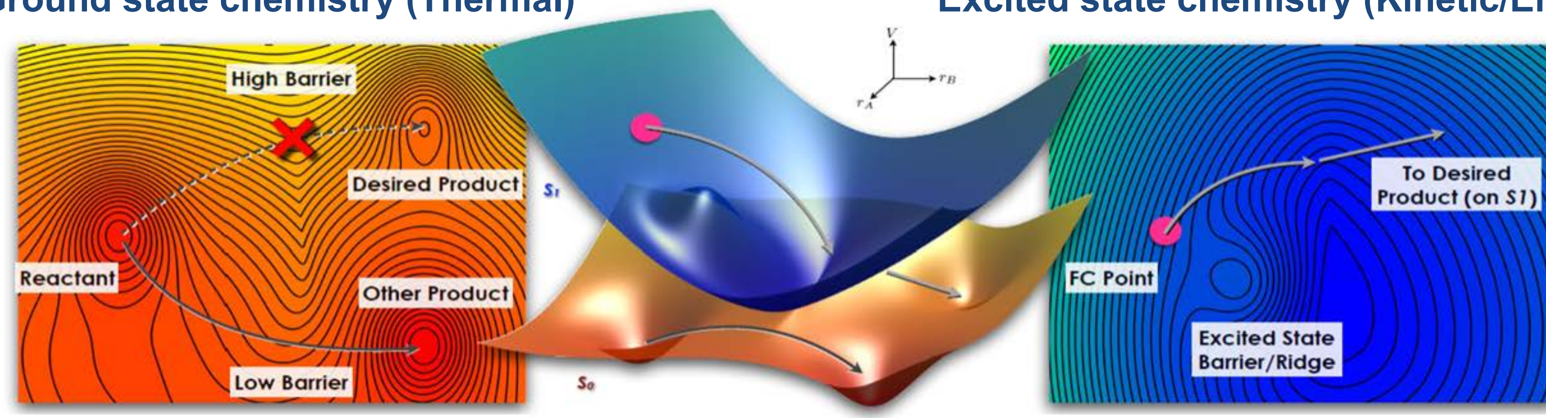
Harnessing Driven Dynamics for Chemical Transformations

Using non-equilibrium states to direct chemical outcomes

- Systems driven by photons and electrons enable rare and functional configurations to be accessed with higher probability
- Understanding, and ideally controlling, non-equilibrium phenomena has foundational significance of potential importance to energy technology

Ground state chemistry (Thermal)

Excited state chemistry (Kinetic/Entropic)



Co-design novel modes of excitation with new molecular & material systems to efficiently and selectively drive chemical transformations

Capturing Catalytic Processes for Controlling Photocatalytic Reactivity

Scientific Opportunity

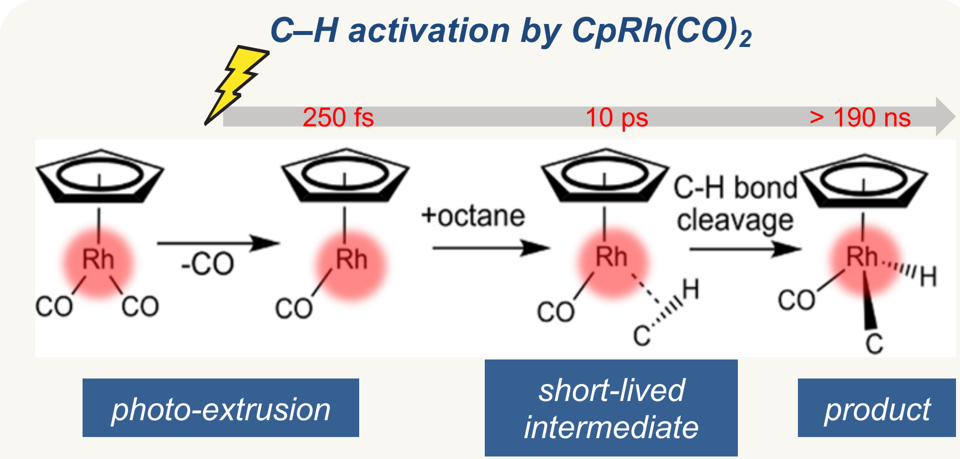
- Mapping the evolution of frontier orbitals in transition metal photocatalysts with atom specificity

Significance and Impact

- Understanding of catalytic processes, such as C-H activation, towards the orbital-level control of catalytic reactivity
- Validate design rules for efficient & robust catalysts from abundant elements (3*d* transition metals)
- Strong link to high-level theory

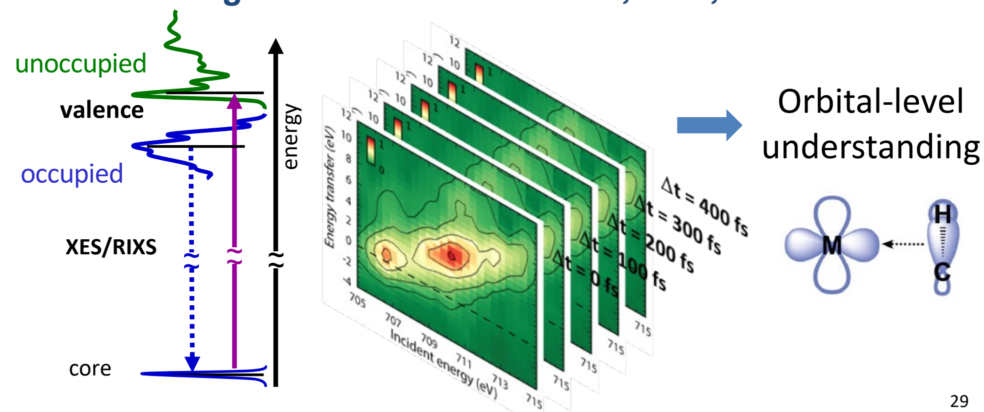
LCLS-II Approach

- High photon flux enables photon-hungry observables such as time-resolved resonant inelastic X-ray scattering (RIXS)
- Dedicated instrumentation for advanced soft and tender X-ray spectroscopy



Jay et al., Science 380, 955–960 (2023)

Tracking C–H activation w/ XAS, XES, and RIXS



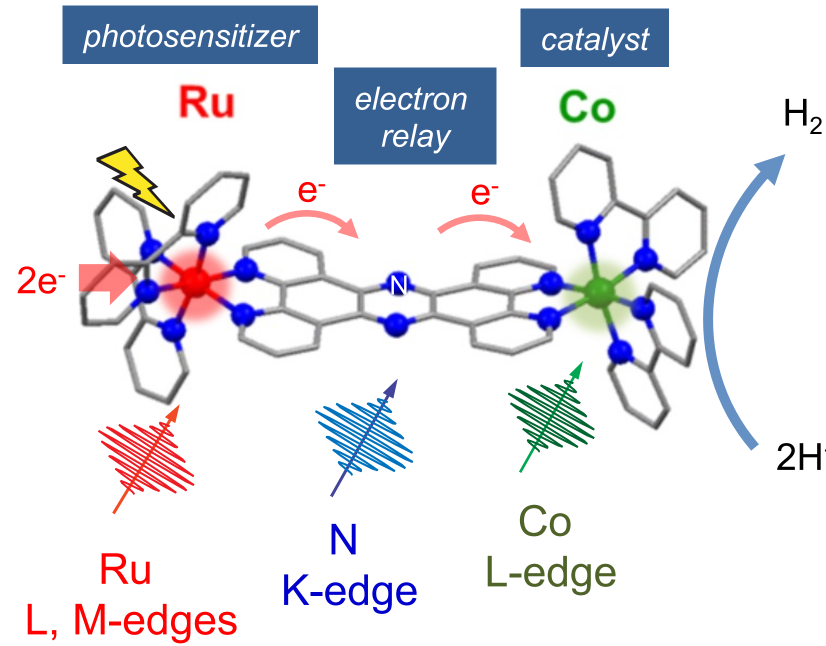
Revealing the full sequence of electronic dynamics in multi-electron catalysts with element specificity

Scientific Opportunity
- Reveal coupling of valence charge structure and subtle changes in molecular structure
- Map entire catalysis cycle, capturing rare transient events (transition states)
- Operando characterization of multi-electron photocatalysts

Significance Impact
- Design of efficient multi-electron catalysts with sequential charge transfer
- Link to theory & simulation

LCLS-II Approach
- Time-resolved spectroscopy maps excited states: valence structure (frontier orbitals)
- High throughput *operando* RIXS at ~100 meV resolution

Following charge transfer cascade



with element specificity

Adapted from
Canton et al., J. Phys. Chem. Lett. 2013, 4, 1972

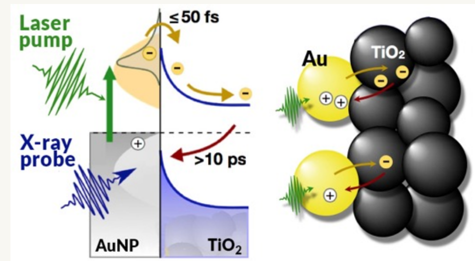
Understanding Surface Reactivity in Heterogeneous Systems

Scientific Opportunity
- Following chemical transformations of adsorbates on catalytic surfaces

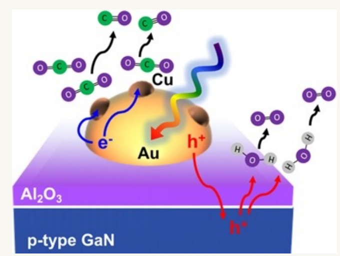
Significance and Impact
- Approaching transition states and identifying intermediates:
  - Understanding of reaction mechanisms
  - Optimization of reaction conditions

LCLS-II Approach
- High average brightness in combination with surface sensitivity of soft X-ray spectroscopy: beyond proof-of-principle experiments to *operando* investigations
- Tailored approaches to trigger reaction :
  - Non-equilibrium electronic excitation
  - Resonant vibrational excitation

Charge transfer at interfaces

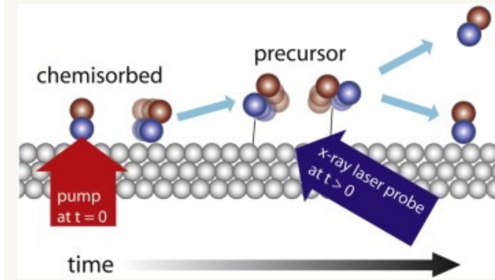


Borgwardt *et al., J. Phys. Chem. Lett.* 2020, 11, 14, 5476.



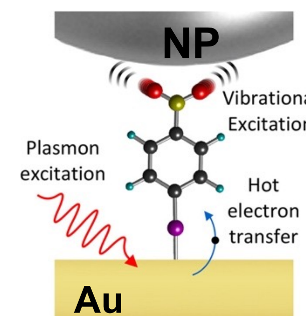
Li et al. ACS Energy Lett. 2021, **6**, 5, 1849.

Surface bond breaking/forming



Nilsson *et al., Chem. Phys. Lett.* 2017, 675, 145.

Tailored approaches to trigger reaction



Shin et al., J. Am. Chem. Soc. 2023, 145, 22, 12264

Understanding Enzyme Design Principles

(example: Light-driven water oxidation in Photosystem II)

Scientific Opportunity

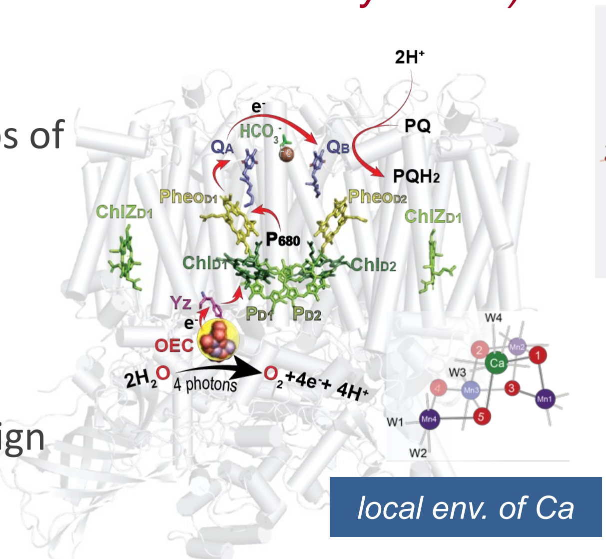
- Understanding the structure-function relationships of reaction centers in biological systems, such as photosystem II, at electronic structural level

Significance and Impact

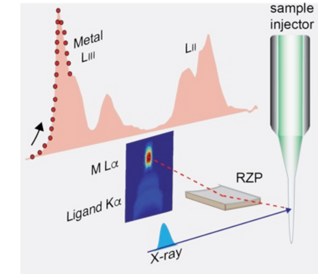
- Understanding of nature's approaches to multielectron/proton catalysis will inform the design principles of artificial systems tailored for specific reactions

LCLS-II Approach

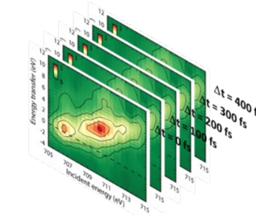
- Brightness of LCLS-II with element & site specificity of soft X-ray spectroscopy:
  - Access to highly dilute biological reaction centers
  - Spectroscopic observables with rich information content (2p3d RIXS)
- New *operando* approaches to triggering biochemical reaction dynamics, e.g. fast mixing, temperature, etc.



local env. of Ca

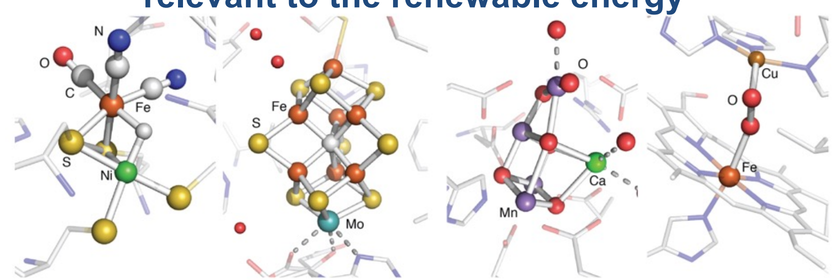


Kubin et al., Struct. Dynam. 2017, 4, 054307



charge/spin on Mn

Examples of metalloenzyme reactions relevant to the renewable energy



NiFe H_2ase: $2H^+ \rightarrow H_2$ | FeMoS Nitrogenase: $N_2 \rightarrow NH_3$ | Mn_4Ca Water oxidase: $2H_2O \rightarrow O_2 + 4H^+$ | FeCu Cyt c oxidase: $O_2 + 4H^+ \rightarrow 2H_2O$

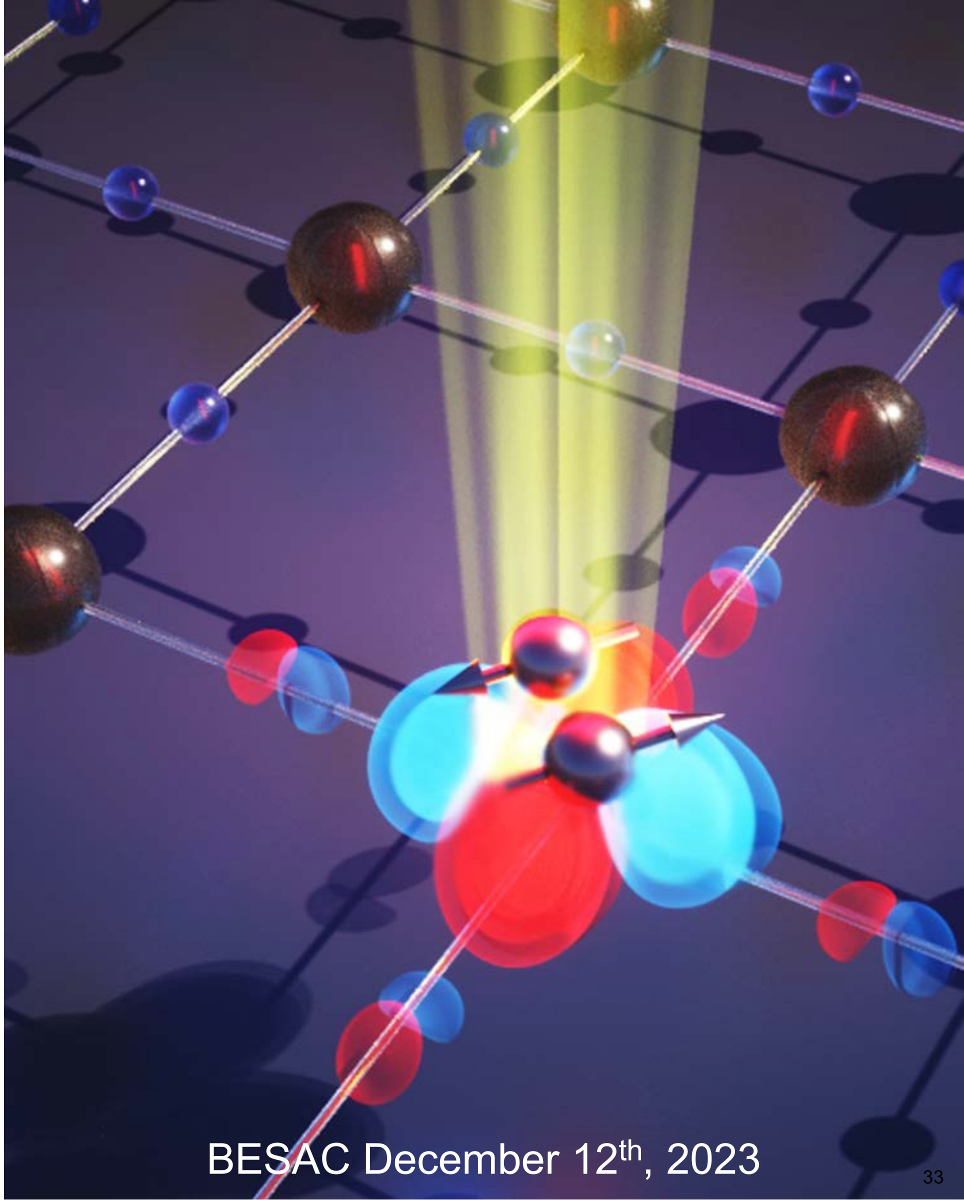
Quantum Materials Science at the LCLS-II

Matteo Mitrano

Assistant Professor

Department of Physics
Harvard University
https://mitrano.physics.harvard.edu

U.S. DEPARTMENT OF ENERGY | Office of Science

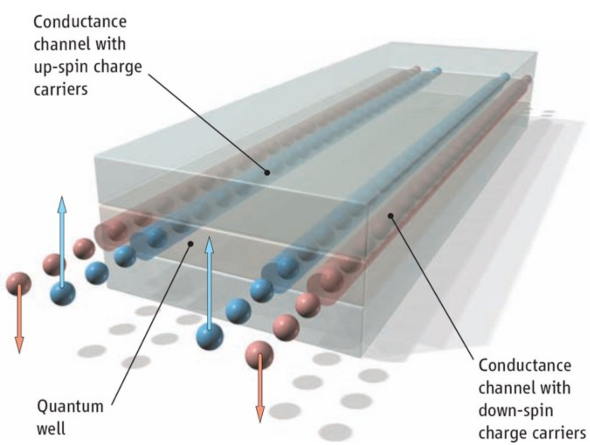


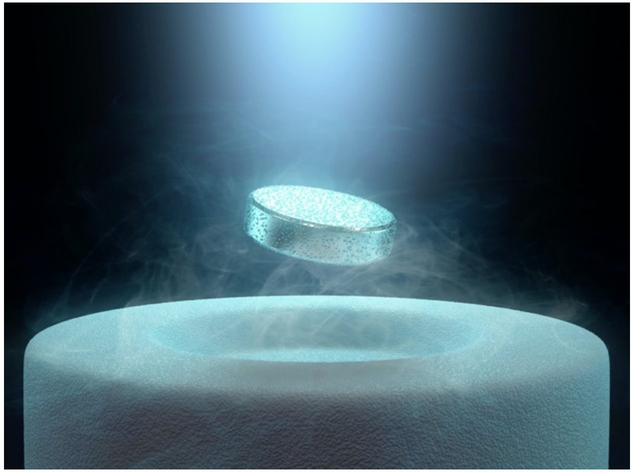
BESAC December 12th, 2023

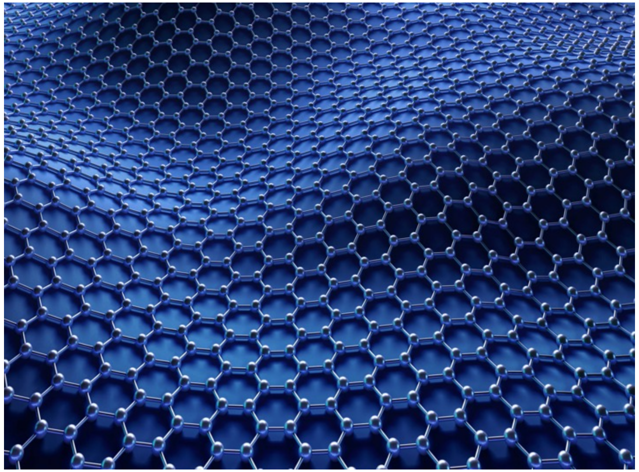
Relevance of quantum materials

"[…] Material systems where quantum effects remain manifest over a wider range of energy and length scales"

*B. Keimer & J. E. Moore, Nature Phys **13,** 1045–1055 (2017)*







- Energy applications
- Quantum information
- Material science

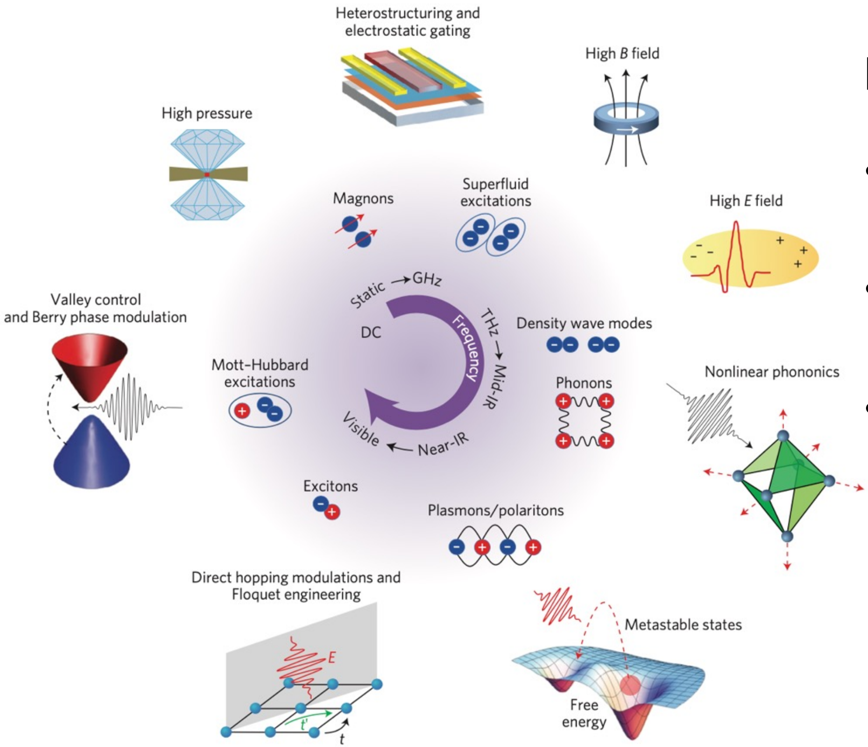
Key to Energy Earthshots Initiative
e.g., Long Duration Storage Shot™

Grand challenges in quantum materials' science

- ❑ How to control complex correlations that give rise to remarkable properties of materials?
- ❑ How can we master energy and information flow on the nanoscale to create new technologies?
- ❑ Beyond ideal materials: How fluctuations, heterogeneity, interfaces, and disorder impact functioning of real materials
- ❑ How do we characterize and control matter away – especially very far away – from equilibrium?

SLAC-R-1053 report "New Science Opportunities Enabled by LCLS-II X-ray Lasers

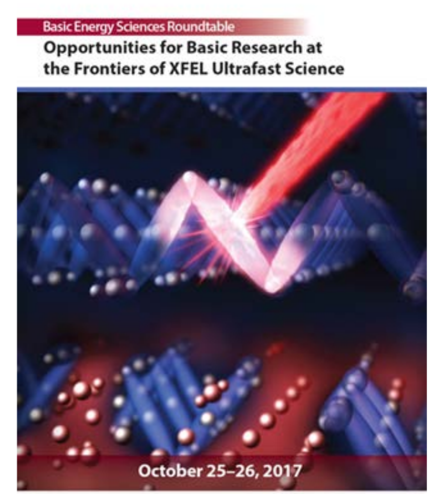
Role of ultrafast light-matter interaction



Basov, Averitt, & Hsieh, Nat. Mater. **16**, 1077 (2017)

Key questions for ultrafast science

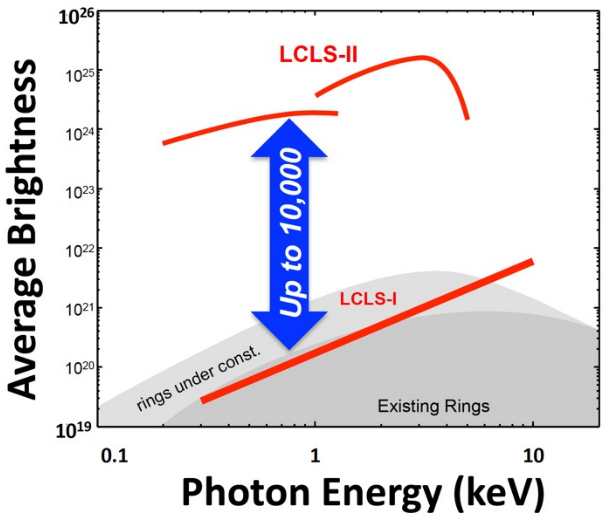
- Can light be used to create novel quantum phases of matter without equilibrium analogues?
- Can ultrafast lasers introduce quantum coherence or create new topological states?
- Can we stabilize novel transient quantum phases for practical use?

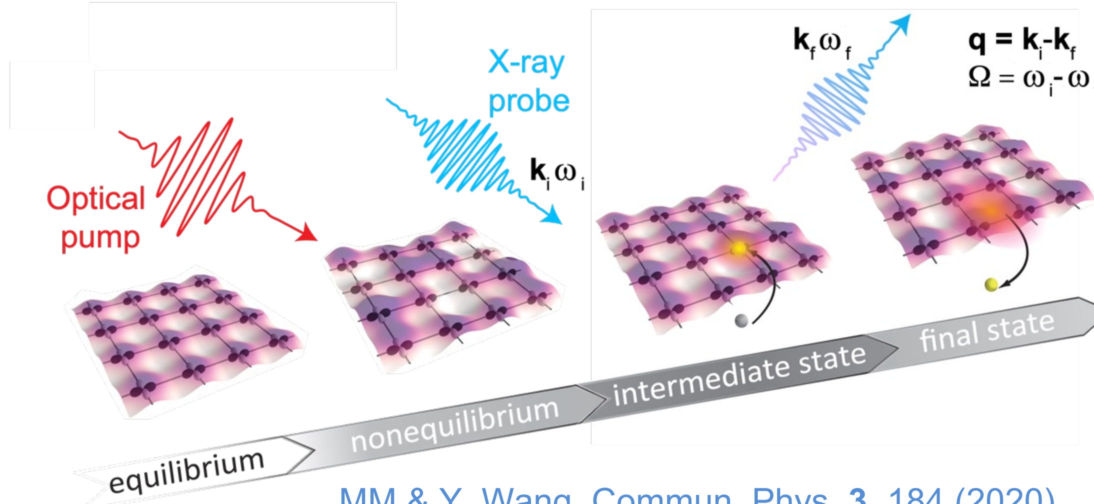


Goal: **dynamically inducing new phenomena and functionalities**

Opportunities offered by LCLS-II

M. Dunne's talk





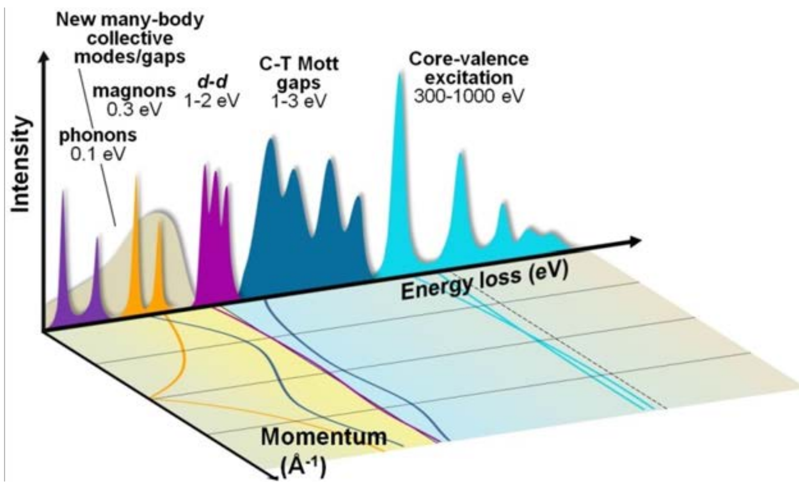
MM & Y. Wang, Commun. Phys. **3**, 184 (2020)

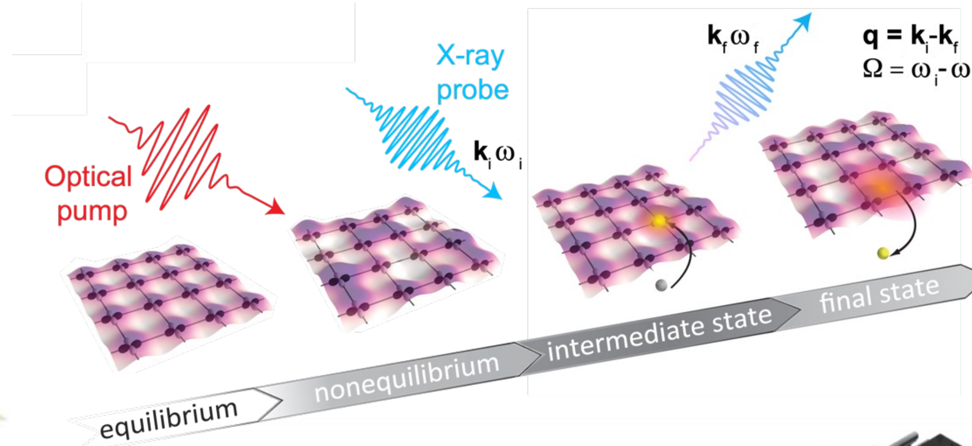
Brightness enhancement prompts **qualitative change** in scientific opportunities moving from x-ray diffraction towards spectroscopic techniques (trRIXS, XPCS, trARPES)

Primary impact

- Revealing emergent nonequilibrium quantum phases
- Dynamically capturing nanoscale fluctuations and heterogeneity
- High-precision measurement of collective modes of quantum materials in and out of equilibrium

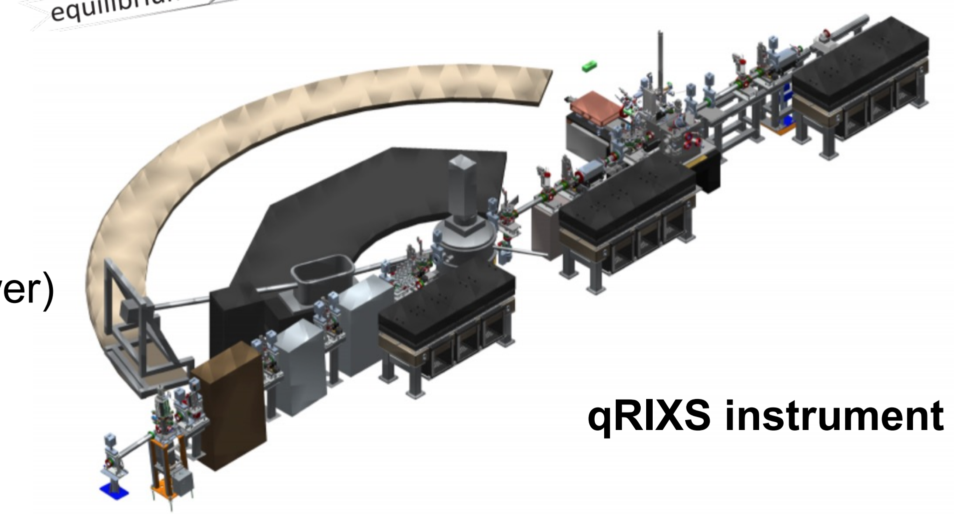
Time-resolved resonant inelastic x-ray scattering





LCLS-II Approach

- Collective modes at ~30-100 meV scale
  - High average power (moderate peak power)
  - High Spectral brightness
- Coherence (energy resolution) near the transform limit
- 2D (q and ω) maps of collective modes

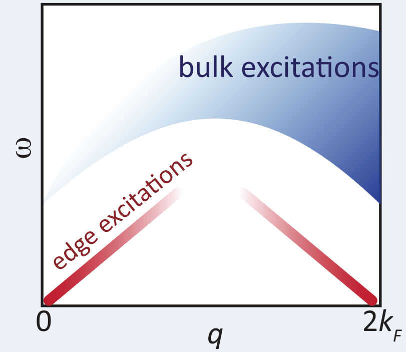


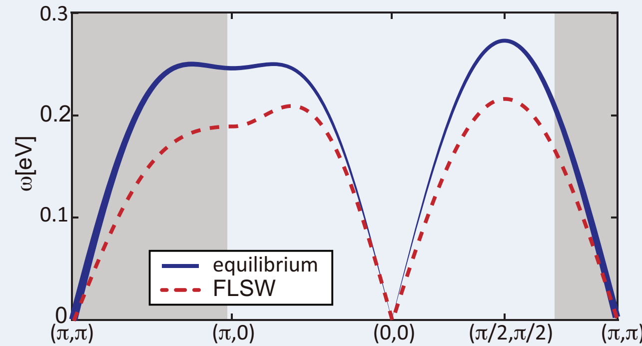
qRIXS instrument

High spectral brightness necessary to provide energy resolution at the relevant timescale

Time-resolved resonant inelastic x-ray scattering

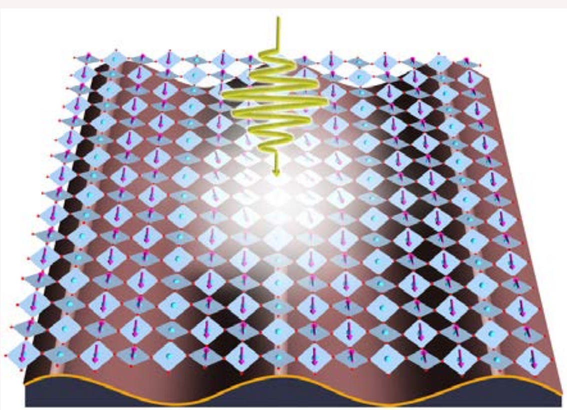
Scientific Opportunities





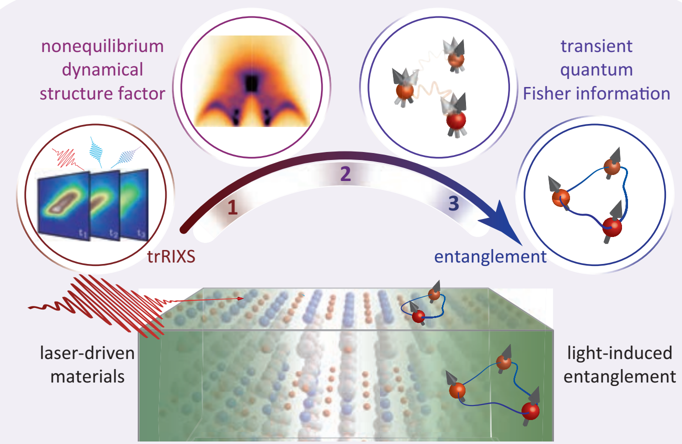
Probe collective modes in and out of equilibrium

MM & Y. Wang, Commun. Phys. **3**, 184 (2020)
Y. Wang et al. Commun. Phys. 4, 121 (2021)



D. Fausti et al. Science **331**, 189 (2011)
W. Hu et al. Nat. Mater. **13**, 705(2014)
D. Nicoletti et al. PRB **90**, 100503 (2014)
MM et al. Sci. Adv. **5**, eeax3346 (2019)

Reveal emergent light-driven phases



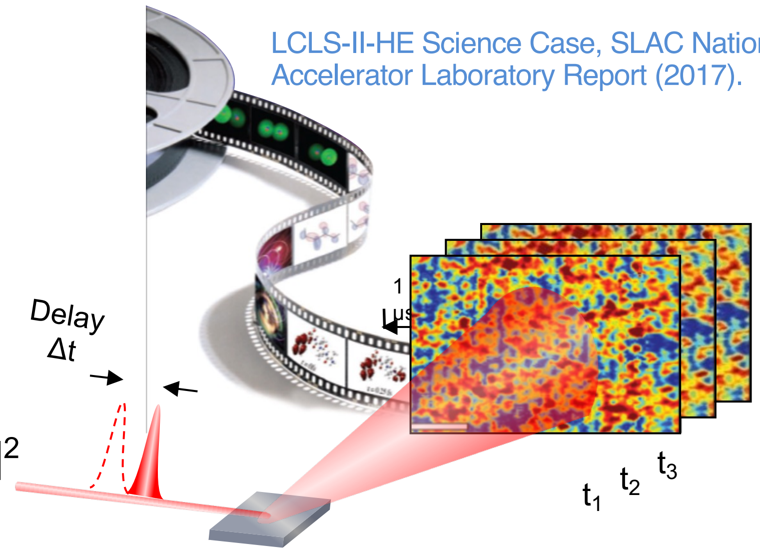
Hales et al., *Nat. Commun* **14**, 3512 (2023)

Quantify many-body entanglement

Dynamic coherent X-ray scattering (XPCS)

LCLS-II Approach

- Dynamic coherent X-ray scattering (XPCS)
  - Maps fluctuating material heterogeneity
  - Nanoscale resolution, element specific (resonant)
- Sequential XPCS, 2-pulse XPCS (programmable LCLS-II pulse structure)
- Accessible time resolution: S/N ~[Ave. Brightness]^2



LCLS-II-HE Science Case, SLAC National Accelerator Laboratory Report (2017).

Scientific Opportunity

- Map spontaneous fluctuations and heterogeneity on multiple time- and length-scales
- Nanoscale fluctuations and control transport of electrons, spins, phonons, and ions.
- Role of defects and heterogeneity in phase transitions and domain motion

XPCS at LCLS-II

Sequential:
- Limited by camera frame rate

2-pulse XPCS:
(programmable pulses)
- >1 μs ➜ 5 ns (RF buckets)
- 1 ps ➜ 10 fs (two-pulse mode)

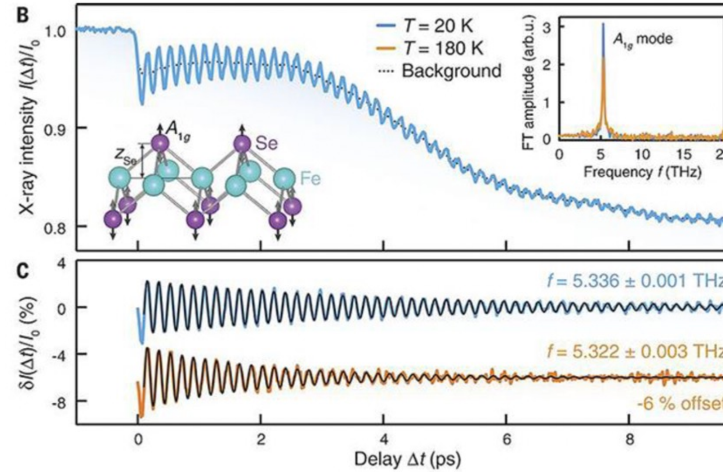
Time-resolved electron spectroscopy

LCLS-II Approach

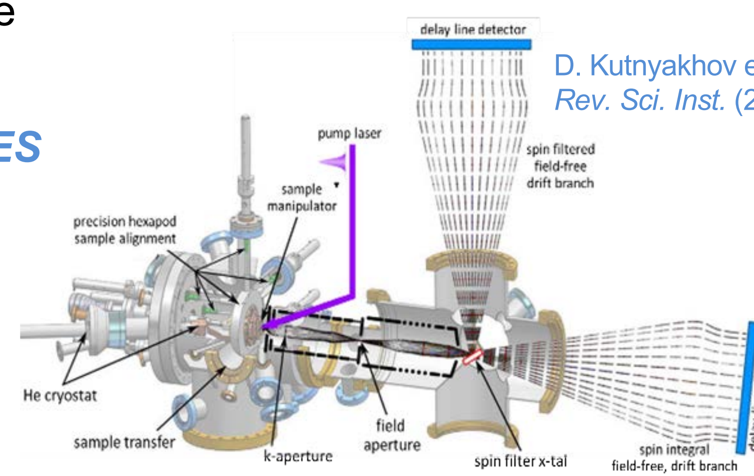
- Advanced momentum microscope (new endstation):
  - High repetition rate and high spectral brightness for high-resolution 3D momentum maps of electronic structure (ARPES)
  - Time/energy resolution at the FT-limit
  - Element-specific (resonant) photoelectron diffraction (XPD)
- Capture metastable phases and dynamic response to pump excitation

Momentum (k) microscope: multi-modal tr-ARPES & element-specific photo-electron diffraction

High spectral brightness, repetition rate, and ultrafast pulse duration of LCLS-II are essential for tr-ARPES & tr-XPD



S. Gerber et al. Science **357**, 71 (2017)



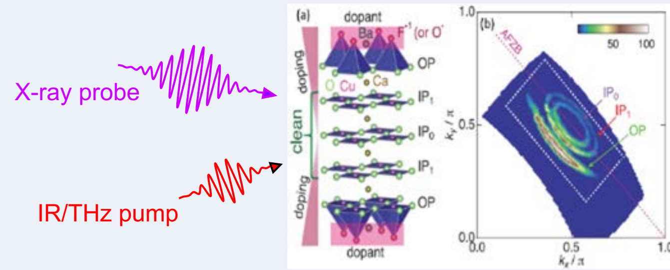
D. Kutnyakhov et al., *Rev. Sci. Inst.* (2020).

Time-resolved electron spectroscopy

Scientific Opportunities

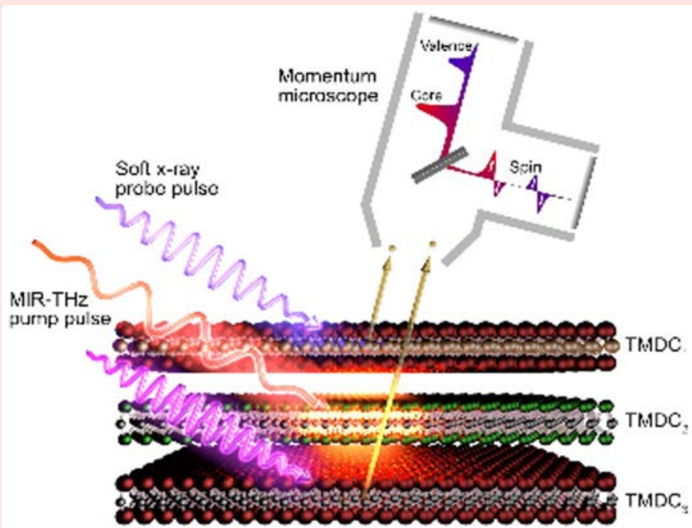
- Probe couplings in quantum materials through multimodal measurements
- Reveal photoinduced states of matter

Dynamics of complex oxides and interfaces

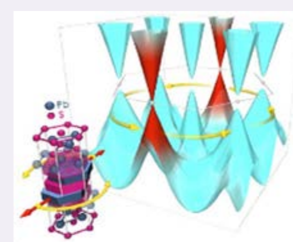


Charge
Spin
Orbital
Lattice

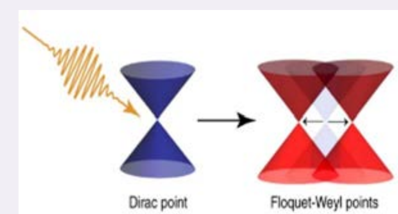
Dynamics across stacked layered materials



Ultrafast engineering of topological matter



Lattice Control

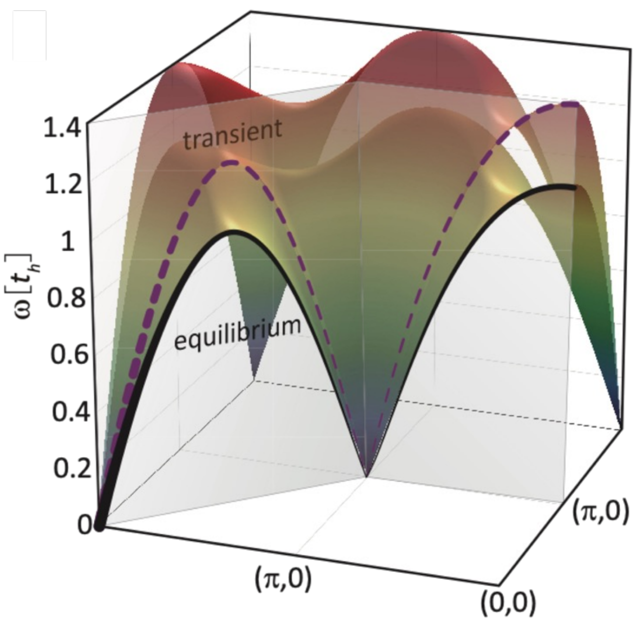


Floquet Control

Z.X. Shen, K. Rossnagel, G. Schoenhense et al.

Conclusion

LCLS-II will support new spectroscopic probes of quantum materials in and out of equilibrium (trRIXS, XPCS, trARPES)



*MM & Y. Wang, Commun Phys **3**, 184 (2020)*

- trRIXS will provide new access to collective modes, driven phases, and many-body entanglement.
- XPCS will inform about heterogeneity and domain dynamics at higher frequencies than current experiments
- trARPES will enable studies of driven topological phases, oxide interfaces, and artificial quantum materials
- Key technical advances: high brightness, 2-pulse XPCS, k-microscope, high-resolution RIXS

Today's panel discussion

• Mike Dunne (SLAC)	Context: The LCLS facility and the LCLS-II Project	10 mins
• Toni Taylor (LANL)	Scientific Impact of LCLS	10 mins
• Daniel Rolles (KSU)	AMO Physics and Gas-Phase Chemistry	10 mins
• Junko Yano (LBNL)	Condensed Phase Chemistry and Catalysis	10 mins
• Matteo Mitrano (Harvard)	Quantum Materials Science	10 mins
• **Q&A / discussion with the panel**		**40 mins**

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