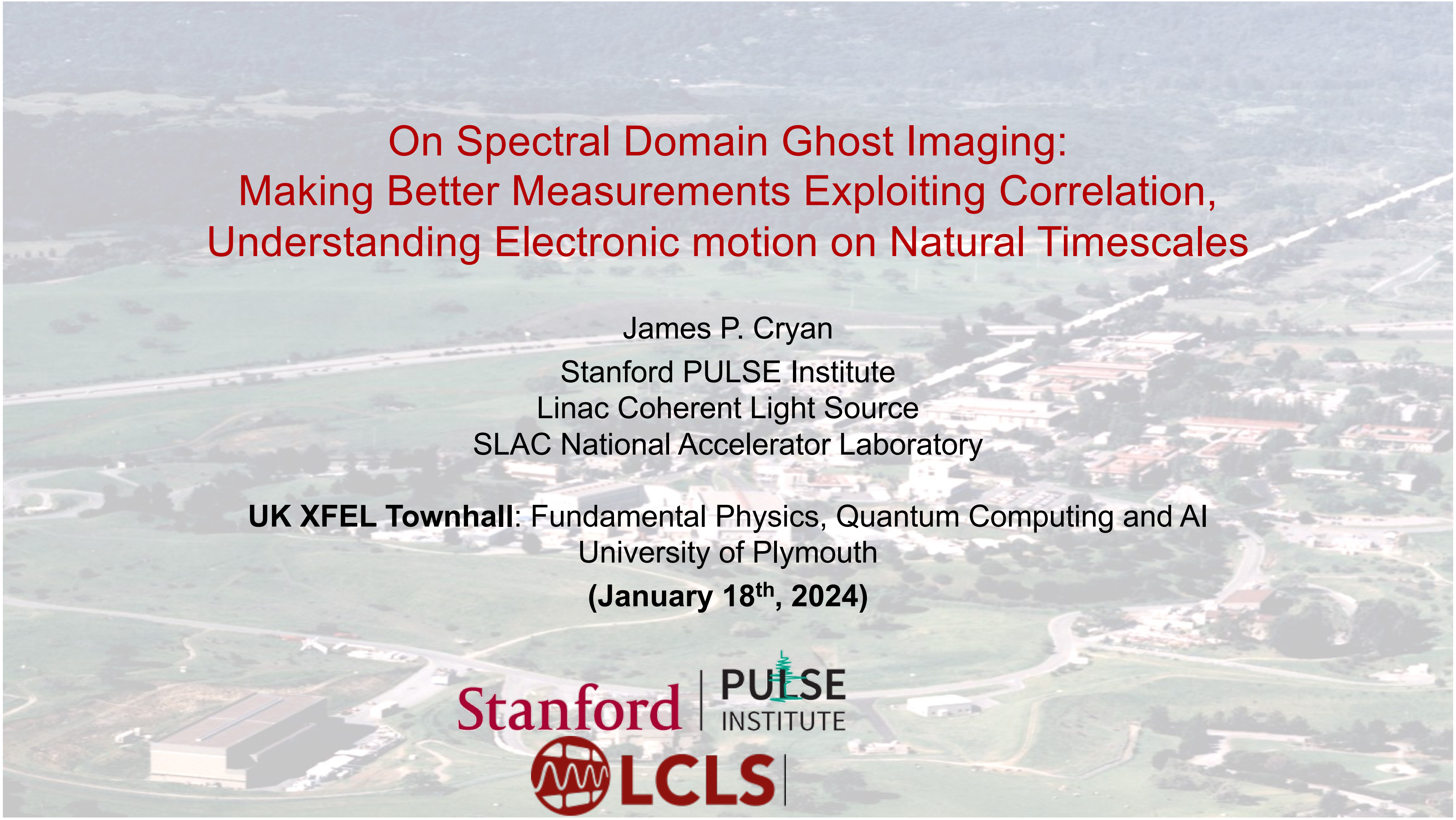


On Spectral Domain Ghost Imaging: Making Better Measurements Exploiting Correlation, Understanding Electronic motion on Natural Timescales

James P. Cryan
Stanford PULSE Institute
Linac Coherent Light Source
SLAC National Accelerator Laboratory

UK XFEL Townhall: Fundamental Physics, Quantum Computing and AI
University of Plymouth
(January 18th, 2024)

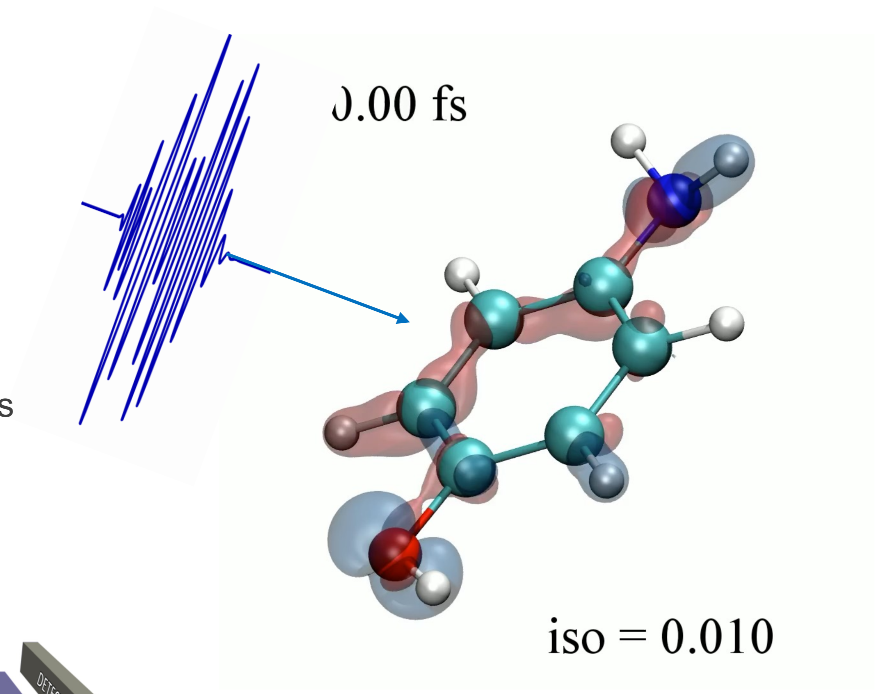
Stanford | PULSE INSTITUTE
LCLS

Coherent Electron Dynamics & the Attosecond Timescale

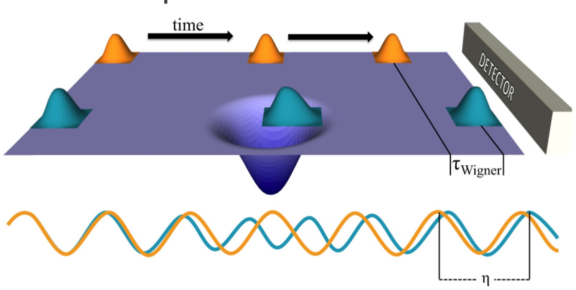
- Electron motion is responsible for photochemical change
  - How does electronic charge (and energy) flow in a molecular system?
  - How is this flow/motion influenced by the mutual interactions between electrons (electron-electron correlation effects)?
  - What role does electronic coherence have on subsequent nuclear motion (chemistry)?

- This is a hard question! We need to start with small model systems
  - $T = 2\pi / \frac{\Delta E}{\hbar} \lesssim 1$ fs when $\Delta E \sim$ few eV
  - We need attosecond measurements

- There is other important interactions that takes place on the attosecond timescale:
  - Photoemission delay
  - Auger-Meitner decay
  - Interatomic Coulombic Decay (ICD)

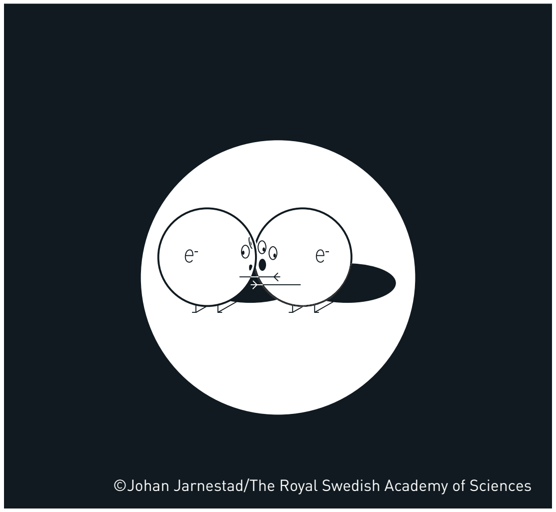


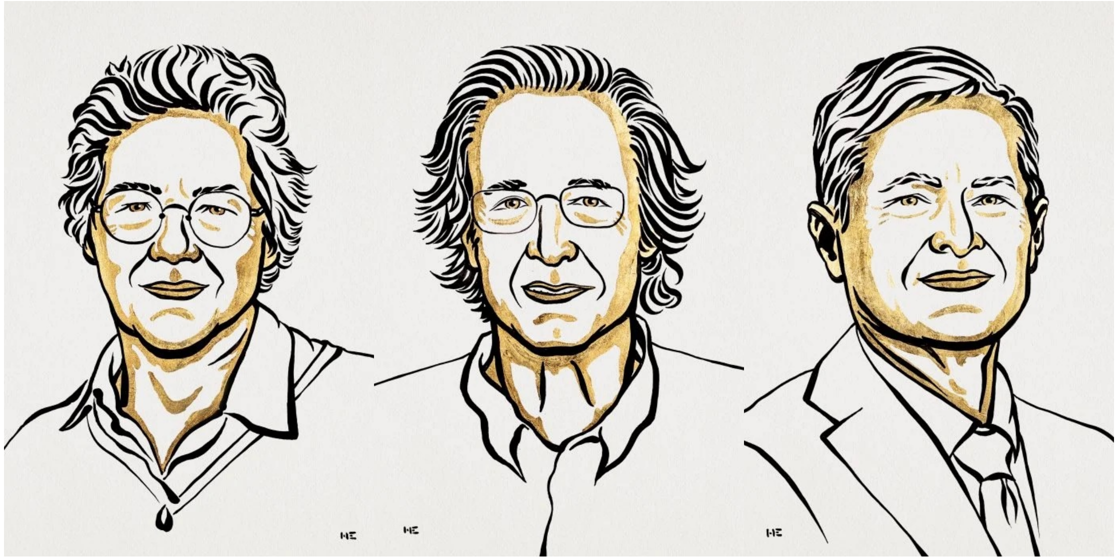
Grell *et al*. Phys Rev. Res. **5** 023092 (2023)



L. Argenti, *Phys. Rev. A* 95, 043426 (2017)

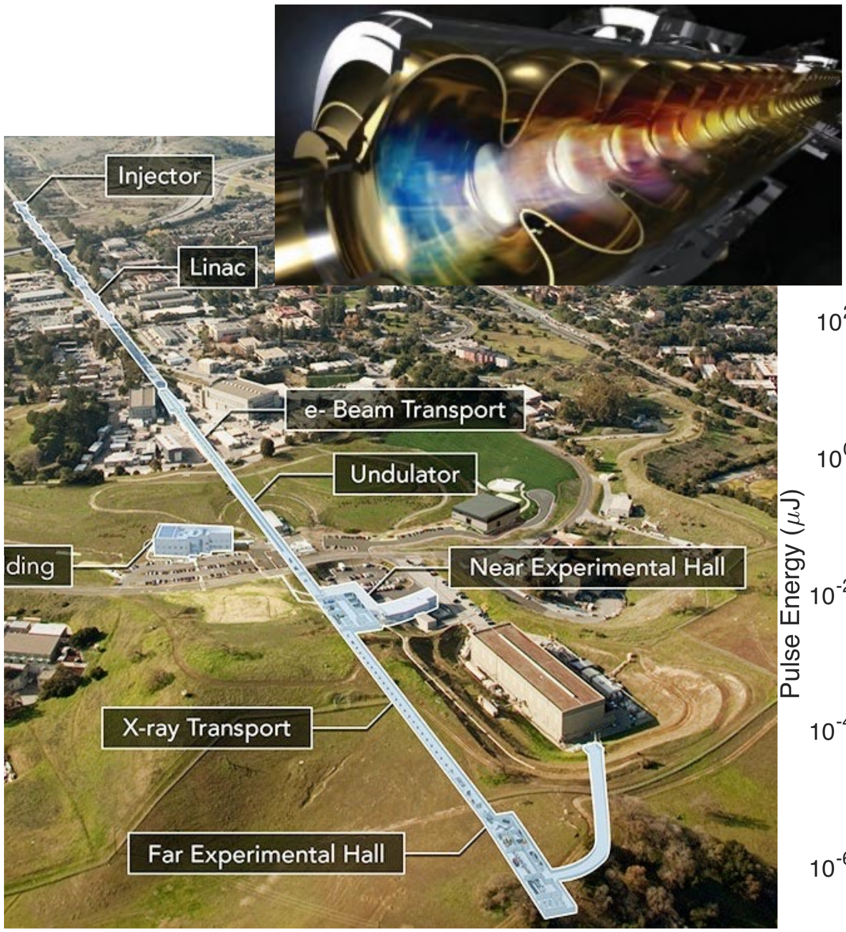
2023 Nobel Prize in Physics

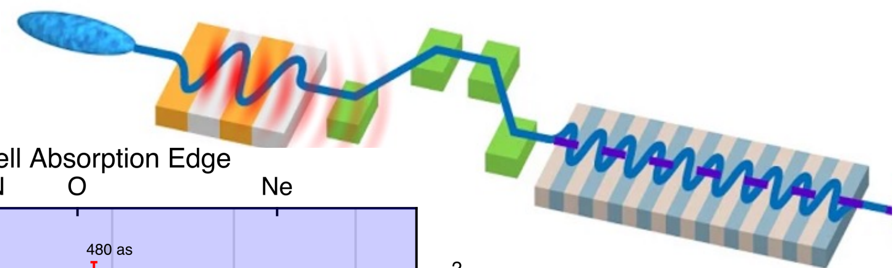


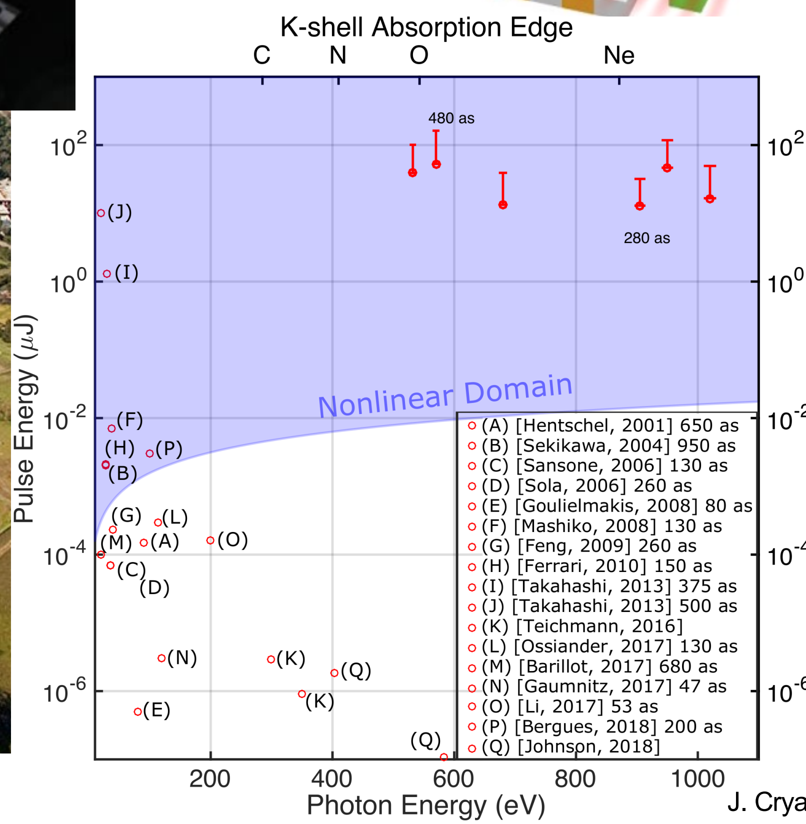


“for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter.”

Attosecond Pulses at LCLS: XLEAP project



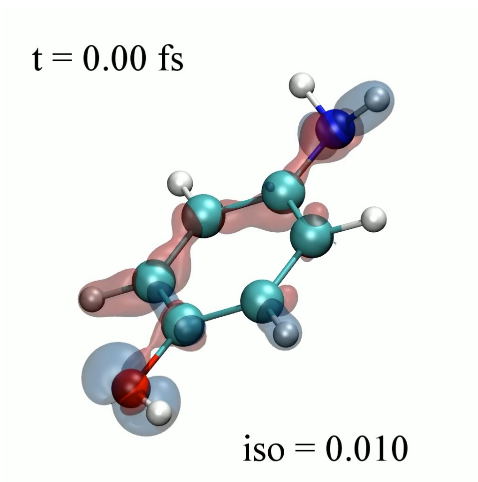




- ESASE for SXR pulses.
- 10's-100's uJ
- High rep. rate coming online

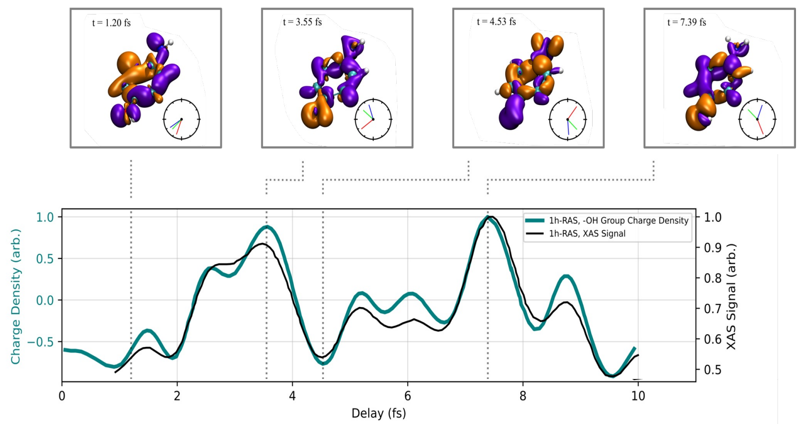
J. Cryan *et al.* AAMOP **71** 1-64 (2022)

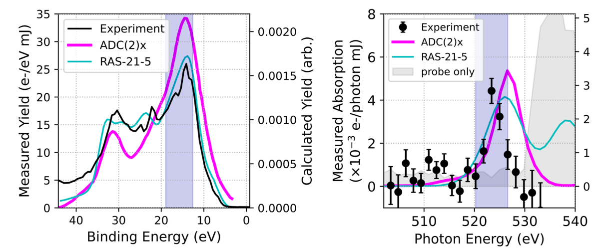
Objectives of the Attosecond Science Campaign at LCLS



Creation and control of nonstationary electronic states

Explore X-ray spectroscopic observables for following ultrafast charge motion with atomic-site specificity

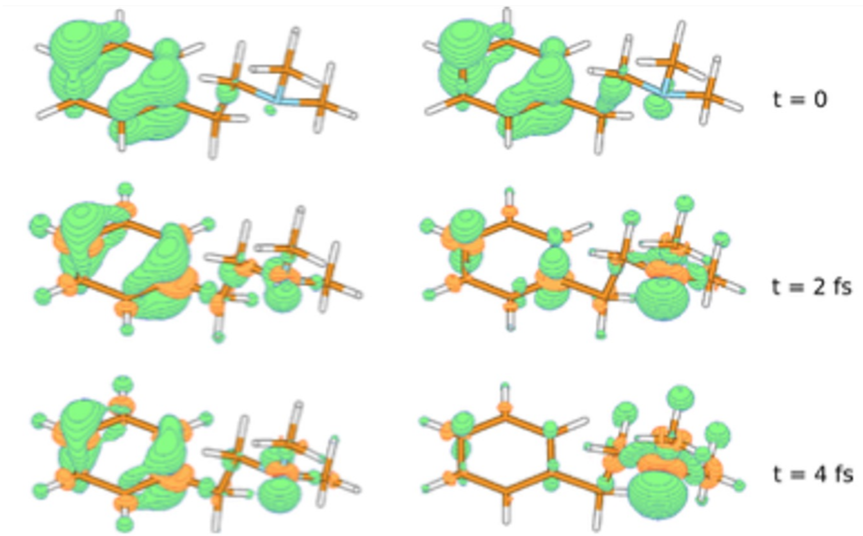




Validation of theoretical predictions through meaningful comparison between experiment and theory

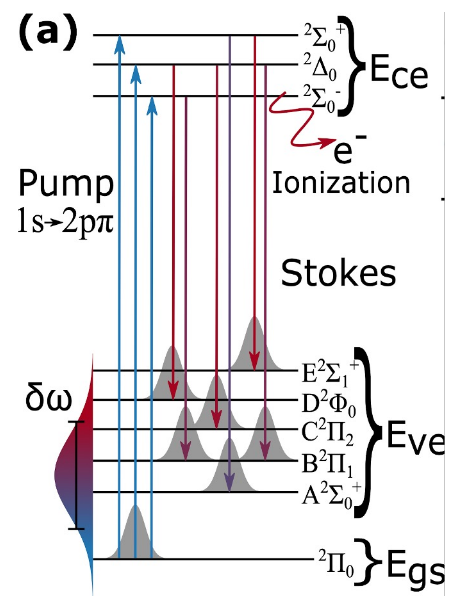
Simulation from Gilbert Grell/Fernando Martin (UAM) and Marco Ruberti (Imperial)

Objectives of the Attosecond Science Campaign at LCLS



Follow charge migration across the molecular backbone and study the coupling of charge motion to nuclear dynamics.

Develop nonlinear X-ray techniques for probing ultrafast dynamics



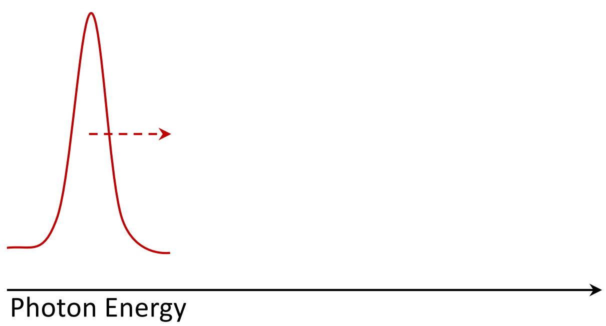
Outline

- Spectral Domain Ghost Imaging: Achieving better measurements thru correlation
  Correlating measurements with high-rate diagnostics to improve measurement fidelity
  - Demonstrations at existing FELs
  - Applying Regression in the temporal domain
- Application to an important problem: Motion of Electron on Attosecond Timescales.
  - Exploiting Spectral Domain Ghost Imaging to follow attosecond electron motion
- Outlook on High Energy Attosecond Pulses:
  - Exploiting X-ray Nonlinearities to study transient chemical processes.

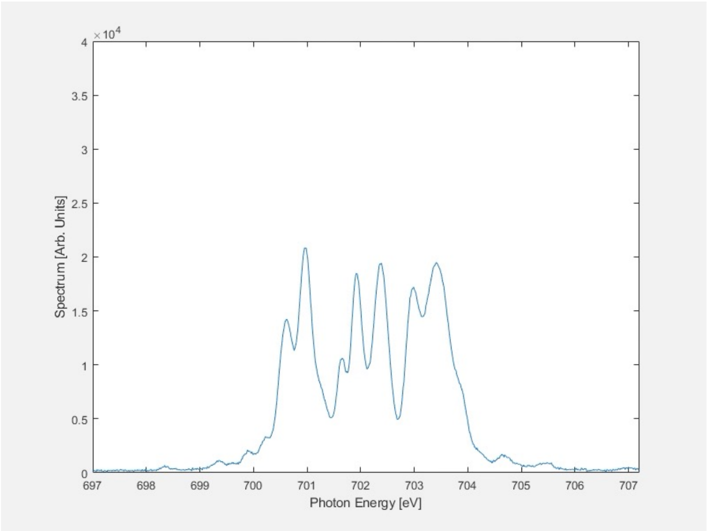
Spectral Variation in SASE FELs

is typically thought of as a disadvantage.

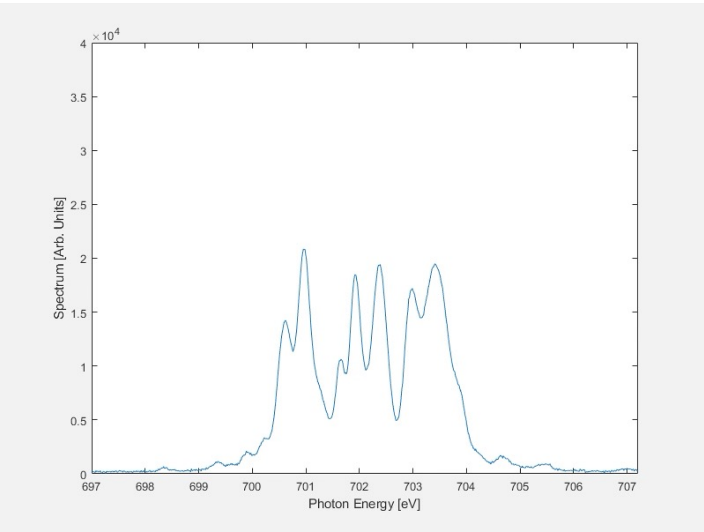
What we *think* we want

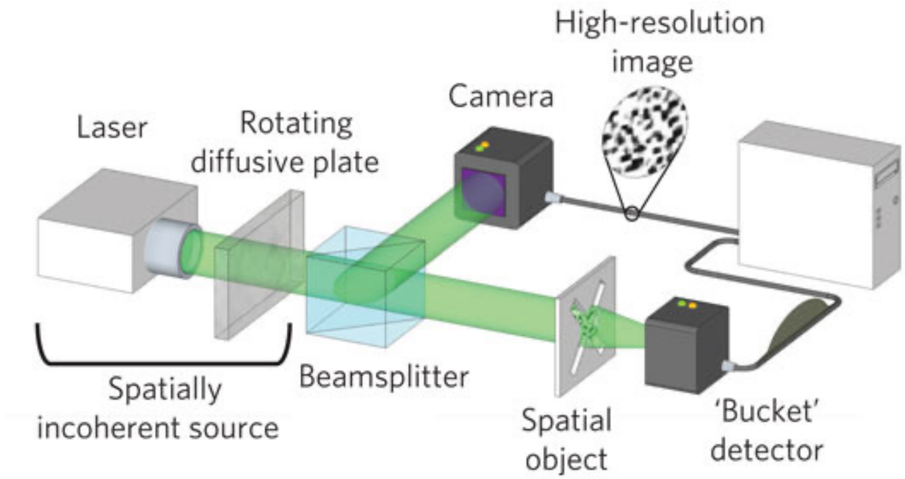


What we have



Making Better Measurements thru Correlation

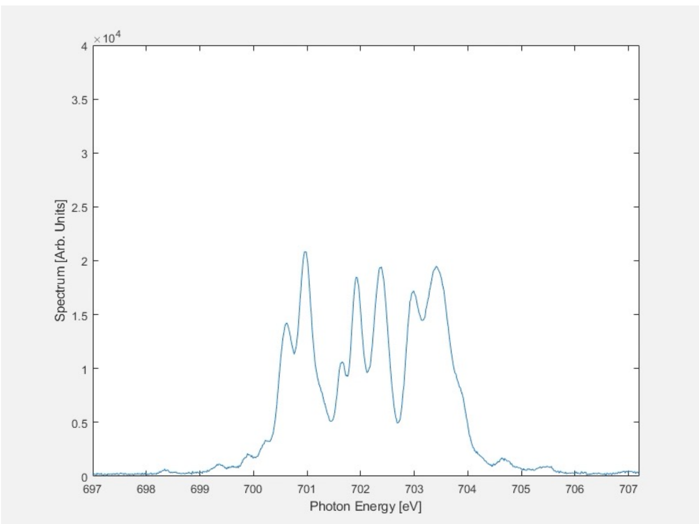


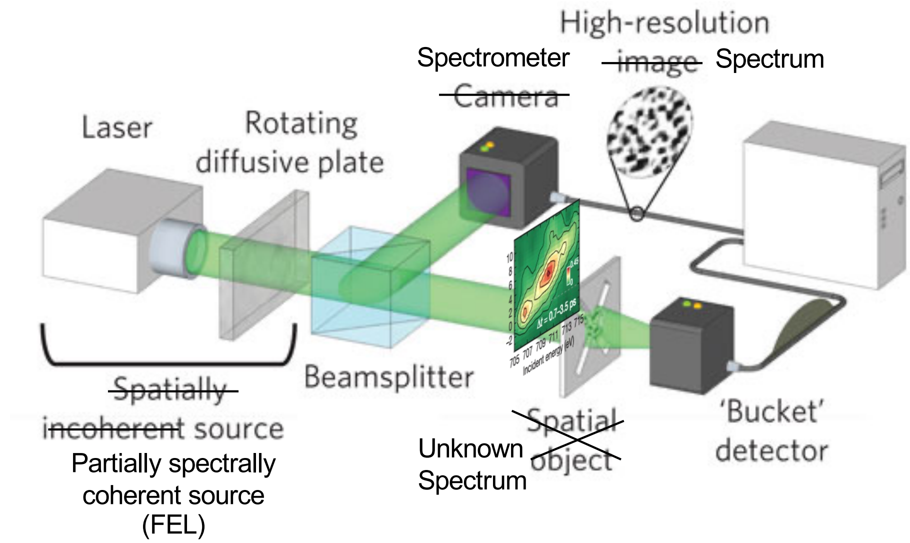


Classical ghost imaging – spatially modulate light source and use correlation between high resolution image and bucket detector to build image of object

P. Janassek, S. Blumenstein and W. Elsäßer, Ghost spectroscopy with classical thermal light emitted by a superluminescent diode, *Phys. Rev. Appl.*, 2018, **9**(2), 021001
C. Amiot, P. Ryczkowski, A. T. Friberg, J. M. Dudley and G. Genty, Supercontinuum spectral-domain ghost imaging, *Opt. Lett.*, 2018, **43**(20), 5025–5028
Kayser, Y., Milne, C., Juranić, P. *et al.* Core-level nonlinear spectroscopy triggered by stochastic X-ray pulses. *Nat Commun* **10,** 4761 (2019)

Making Better Measurements thru Correlation





Research Article Vol. 28, No. 5 / 2 March 2020 / *Optics Express* 5898

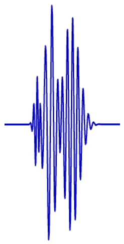
Optics EXPRESS

What are the advantages of ghost imaging? Multiplexing for x-ray and electron imaging

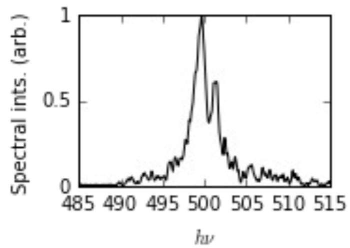
THOMAS J. LANE[1,2] AND DANIEL RATNER[1,3]

P. Janassek, S. Blumenstein and W. Elsäßer, Ghost spectroscopy with classical thermal light emitted by a superluminescent diode, *Phys. Rev. Appl.*, 2018, **9**(2), 021001
C. Amiot, P. Ryczkowski, A. T. Friberg, J. M. Dudley and G. Genty, Supercontinuum spectral-domain ghost imaging, *Opt. Lett.*, 2018, **43**(20), 5025–5028
Kayser, Y., Milne, C., Juranić, P. *et al.* Core-level nonlinear spectroscopy triggered by stochastic X-ray pulses. *Nat Commun* **10,** 4761 (2019)

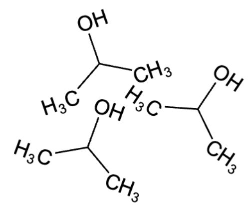
Spectral Domain Ghost Imaging

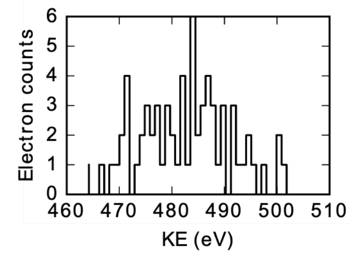


X-ray pulse



Measure incident photon spectrum





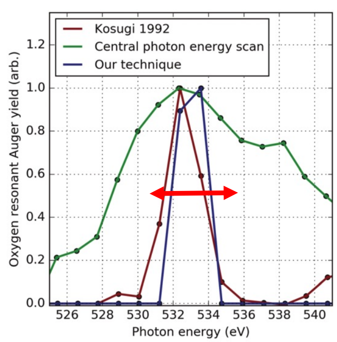
Measure sample response

- e.g. photoelectron KE

$$\boldsymbol{A} \quad \boldsymbol{x} \quad = \quad \boldsymbol{b}$$

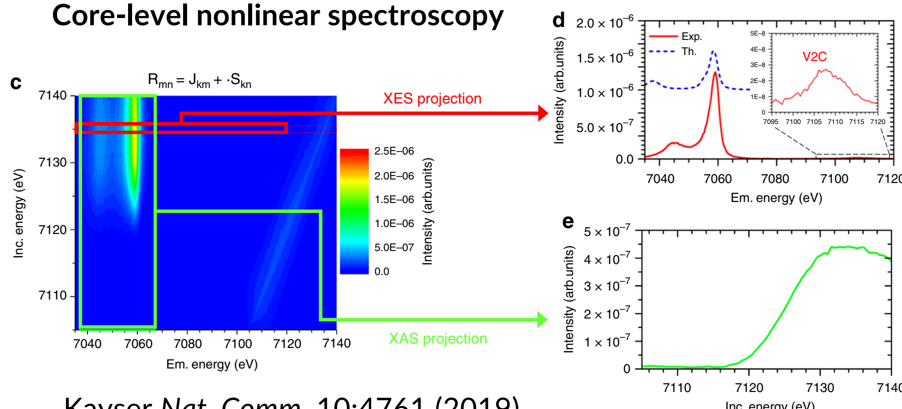
(# shots x # hv bins) (# hv bins x # KE bins) (# shots x # KE bins)

X-ray absorption spectroscopy



Driver *Phys. Chem. Chem. Phys.* **22** (2020)

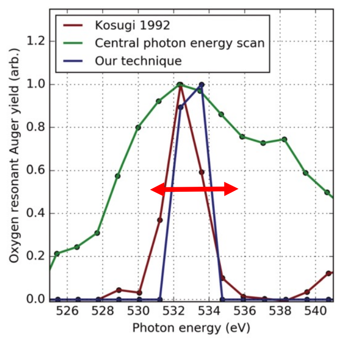
Core-level nonlinear spectroscopy



Kayser *Nat. Comm.* 10:4761 (2019)

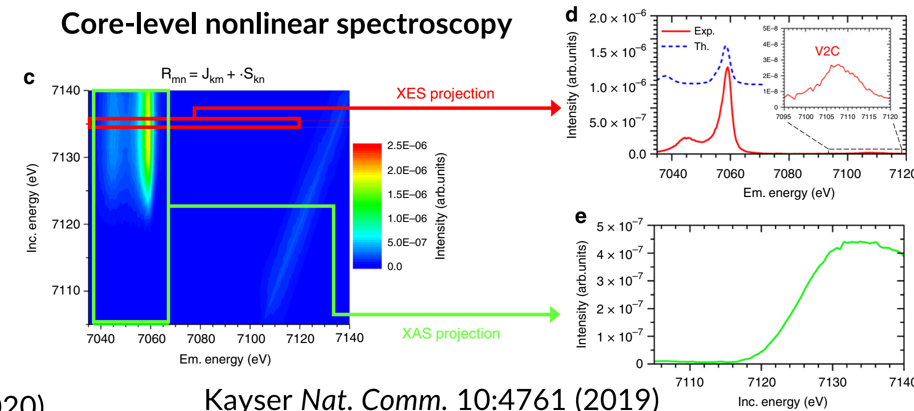
Spectral Domain Ghost Imaging – Sub-bandwidth Resolution

X-ray absorption spectroscopy



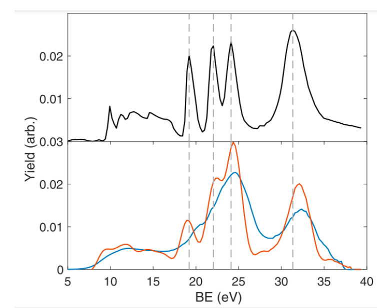
Driver *Phys. Chem. Chem. Phys.* **22** (2020)

Core-level nonlinear spectroscopy



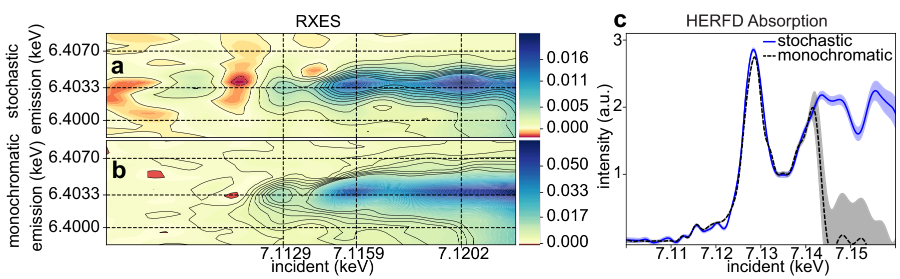
Kayser *Nat. Comm.* 10:4761 (2019)

X-ray photoelectron spectroscopy



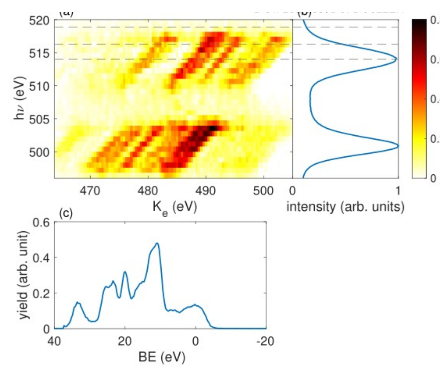
Li *J. Phys. B: At. Mol. Opt. Phys.* **54** 144005 (2021)

Resonant X-ray Emission Spectroscopy



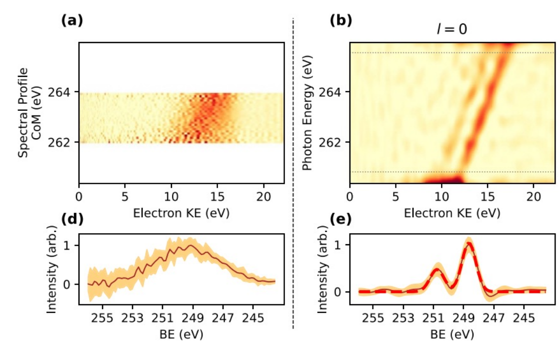
Fuller *Comm. Chem.* **4**:84 (2021)

X-ray pump-probe spectroscopy



Li. *Faraday Discussions* **228** *(2021)*

X-ray velocity map imaging spectroscopy



Wang *New J. Phys.* **25** 033017 (2023)

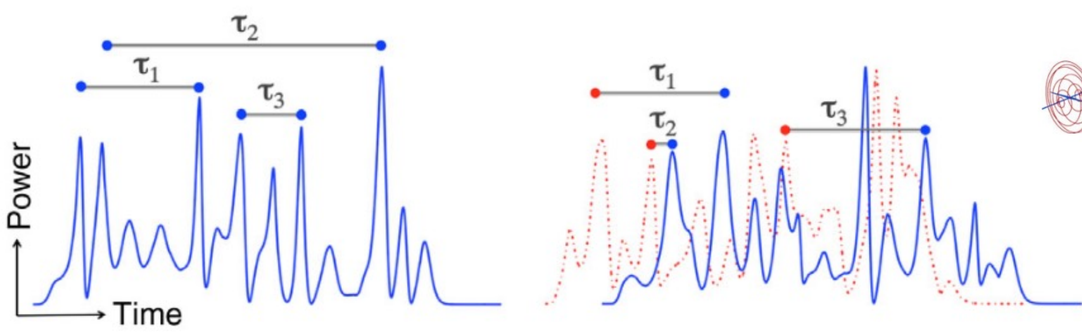
Applying Regression to the Temporal Domain

PHYSICAL REVIEW X **9,** 011045 (2019)

Pump-Probe Ghost Imaging with SASE FELs

D. Ratner,* J. P. Cryan, T. J. Lane, S. Li, and G. Stupakov
SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA

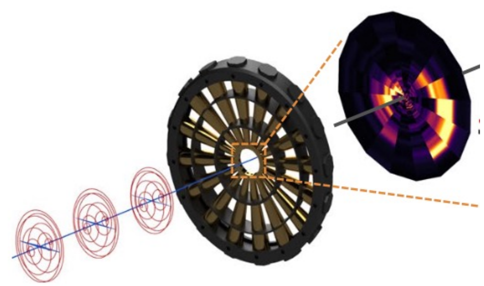
(Received 25 October 2018; published 11 March 2019)

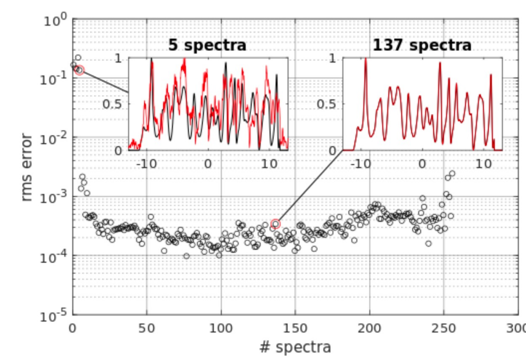


(a) (b)

nature photonics | Hartmann **12** 215–220 (2018) | ARTICLES
https://doi.org/10.1038/s41566-018-0107-6

Attosecond time-energy structure of X-ray free-electron laser pulses





applied sciences

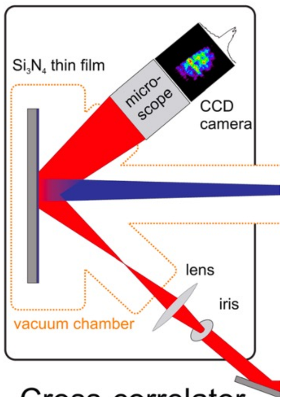
Article

Attoclock Ptychography

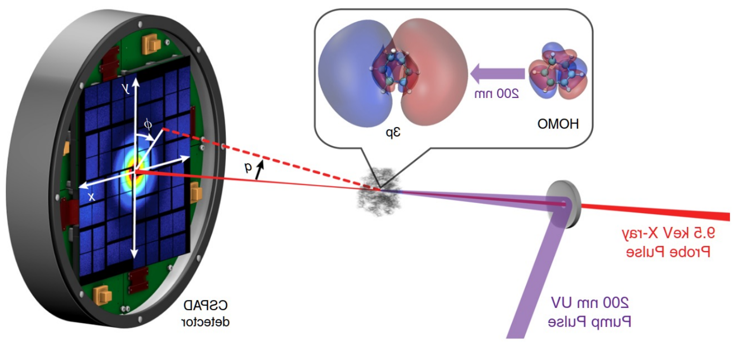
Tobias Schweizer [1,*], Michael H. Brügmann [1], Wolfram Helml [2,3], Nick Hartmann [1,4], Ryan Coffee [4,5] and Thomas Feurer [1]

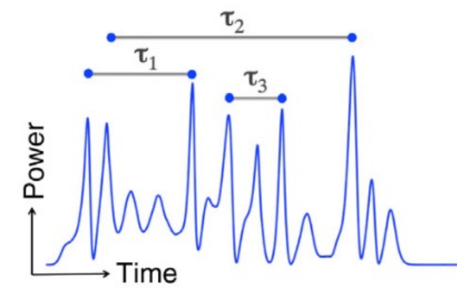
Applying Regression to the Temporal Domain

Making “slow” detectors “ultrafast”



Cross-correlator





(a)

- ”slow” detector which averages over multiple FEL shots
- ”fast” single-shot diagnostic of the relative timing
- Important of high rep. rate FELs

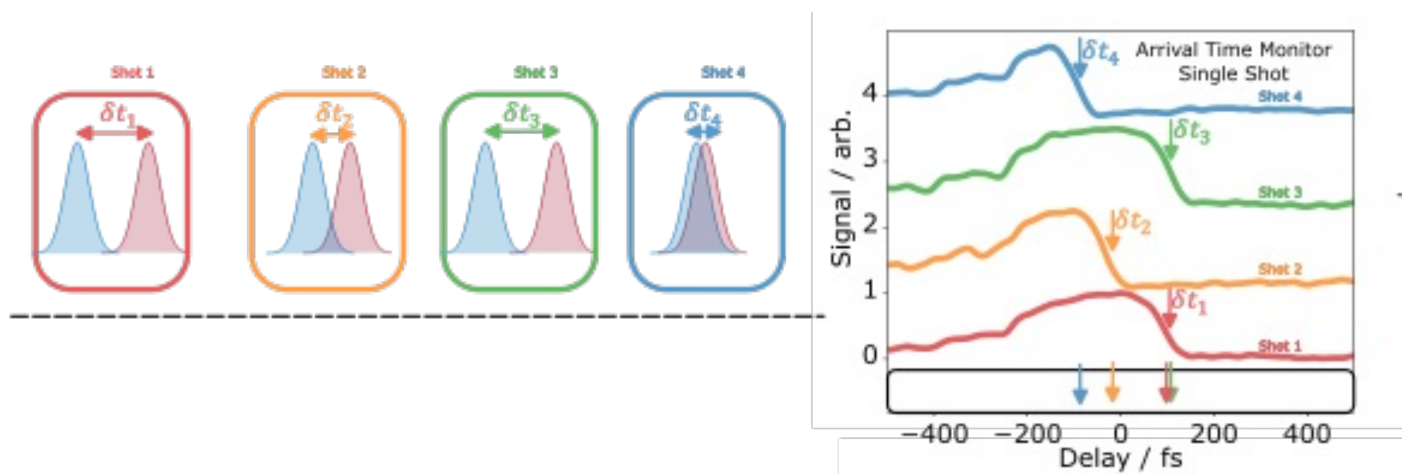
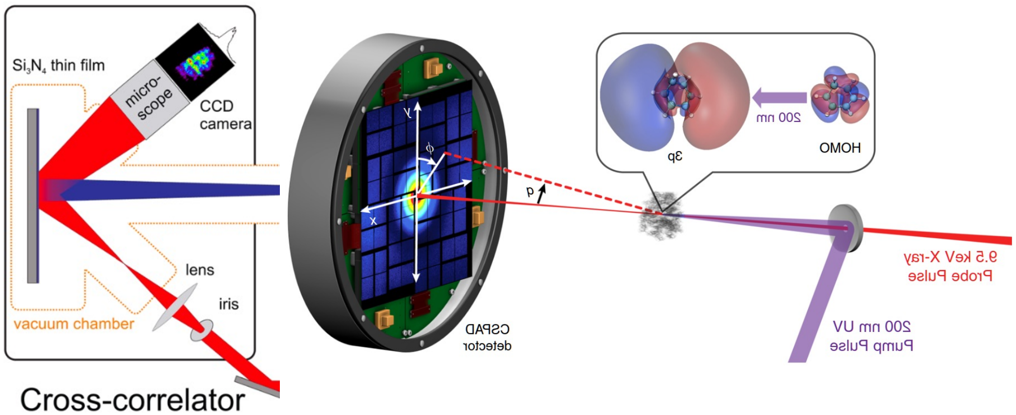


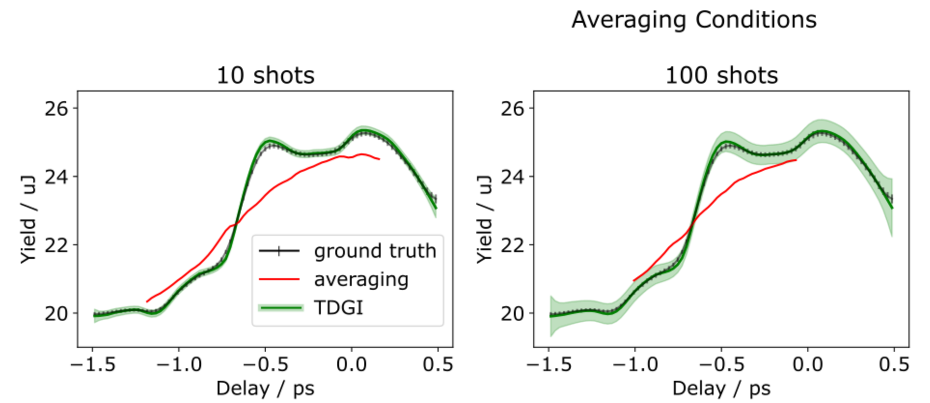
Image Credit: Yong *et al.* Nat. Comm. **11**, 2157 (2020), Schorb *et al.* *App. Phys. Lett.* **100**, 121107 (2012)

Applying Regression to the Temporal Domain

Making “slow” detectors “ultrafast”



Cross-correlator



- ”slow” detector which averages over multiple FEL shots
- ”fast” single-shot diagnostic of the relative timing
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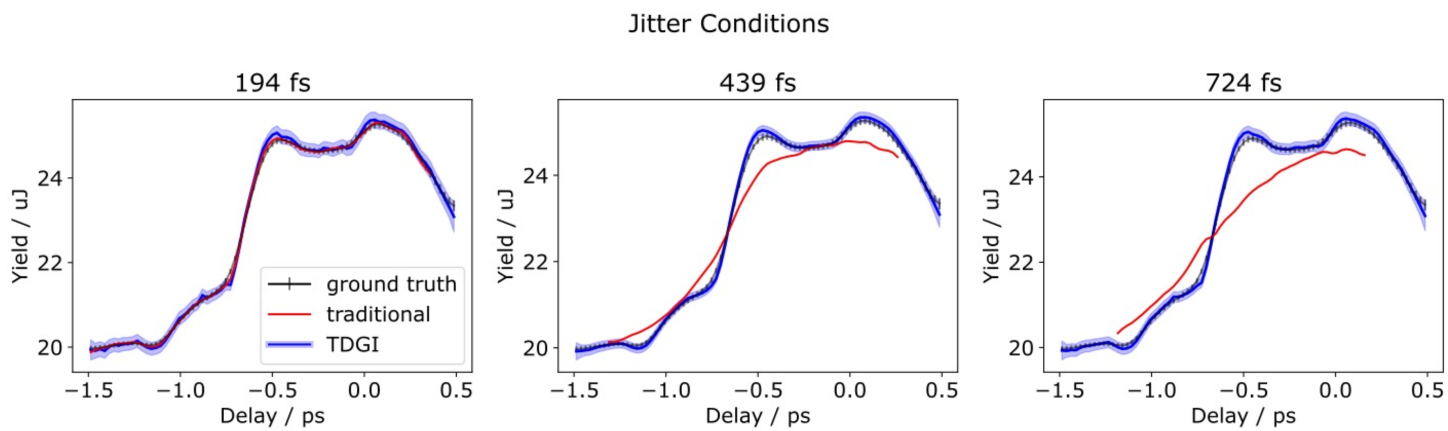
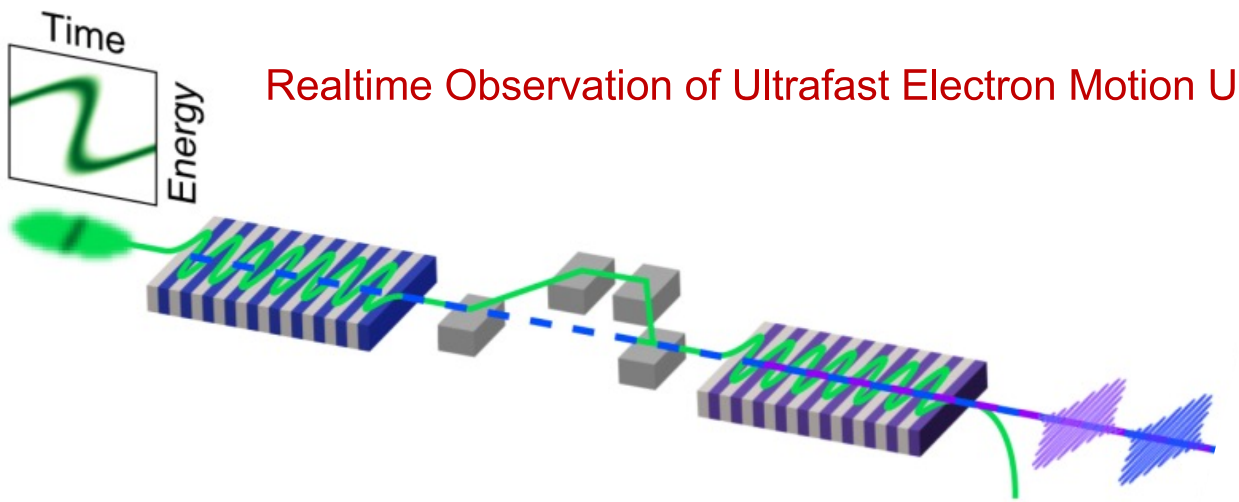


Image Credit: Yong *et al.* Nat. Comm. **11**, 2157 (2020), Schorb *et al.* *App. Phys. Lett.* **100**, 121107 (2012)

Application of Spectral Domain Ghost Imaging

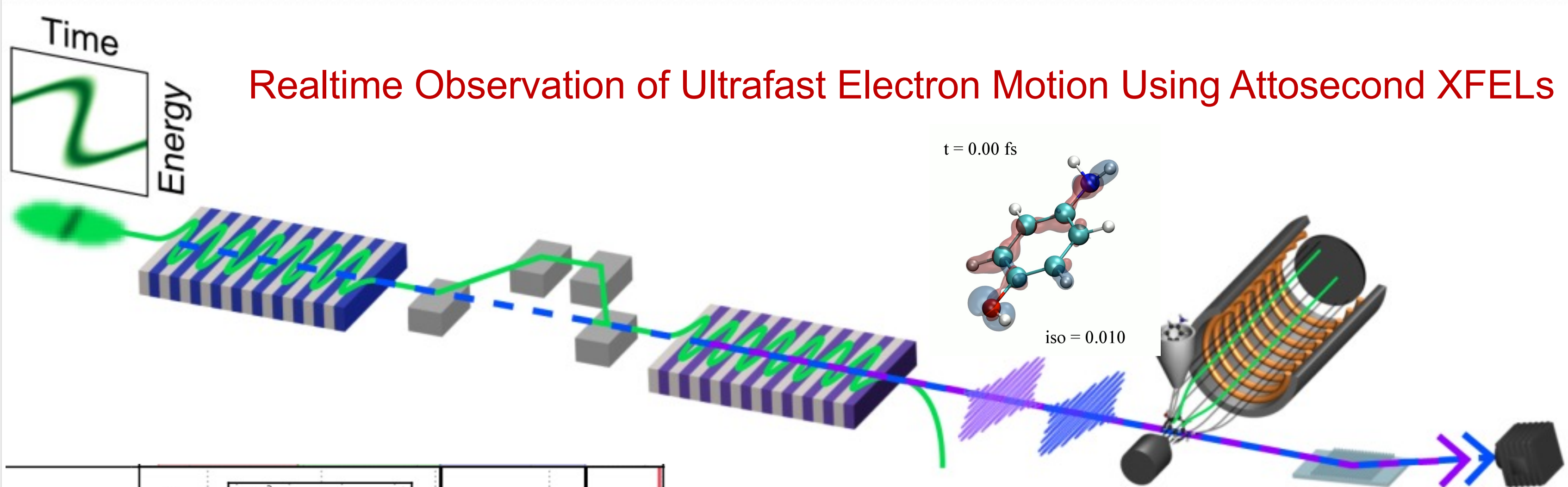
Resolving the Attosecond Motion of Electrons

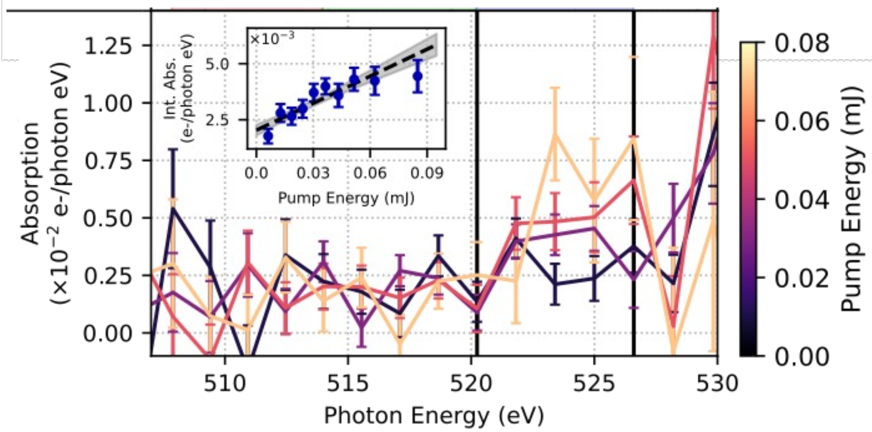
Realtime Observation of Ultrafast Electron Motion Using Attosecond XFELs

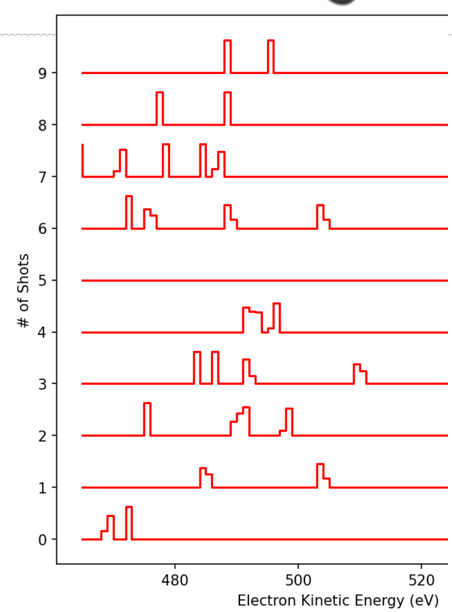


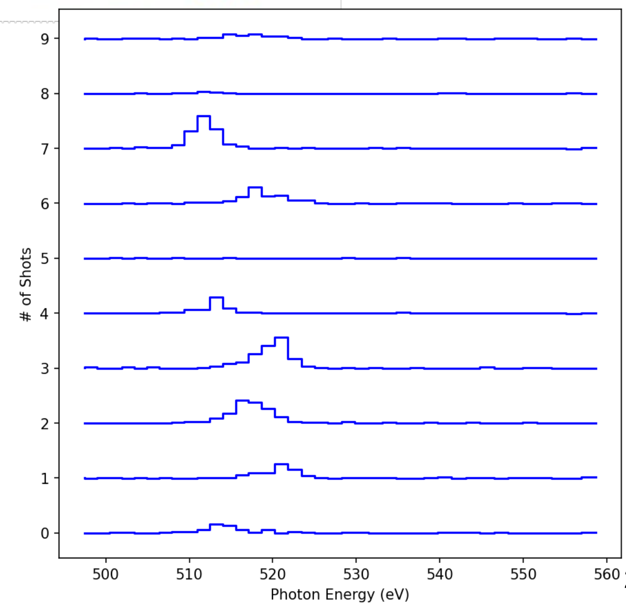
Realtime Observation of Ultrafast Electron Motion Using Attosecond XFELs

Stanford PULSE Institute
LCLS

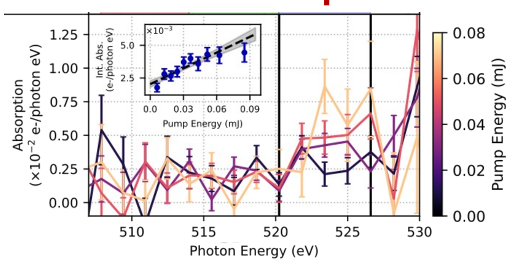


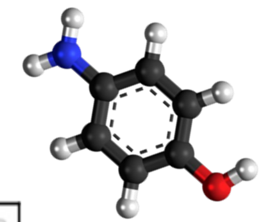


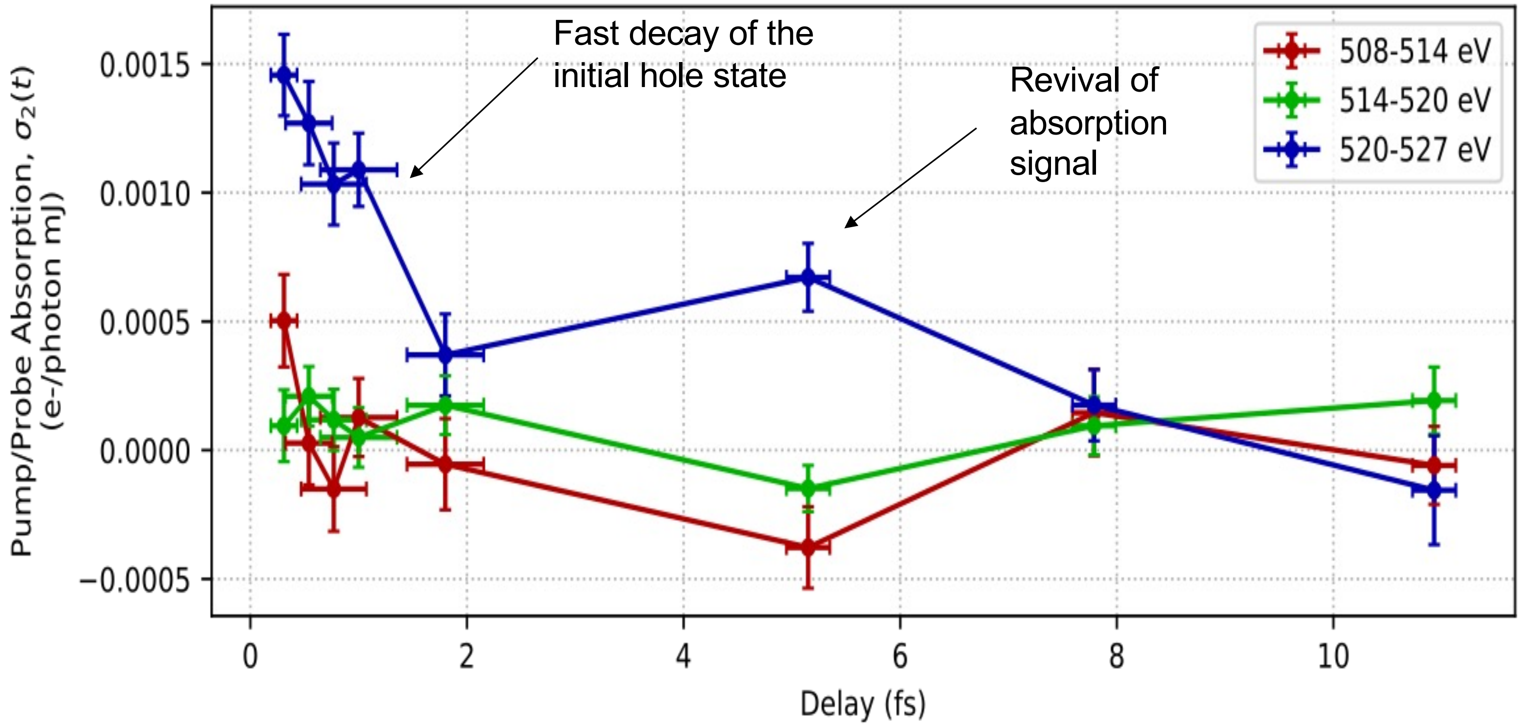




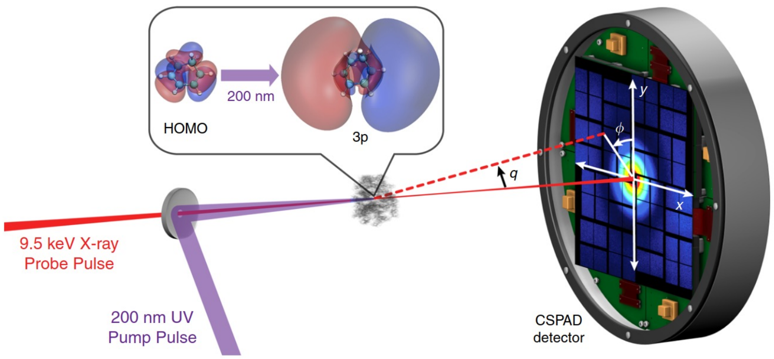
Time Dependence of Absorption Feature

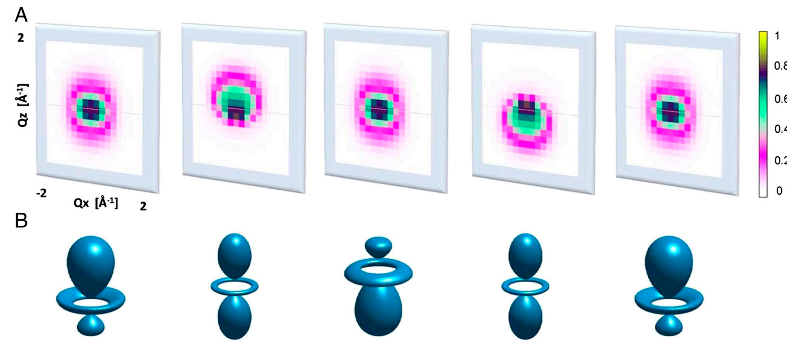






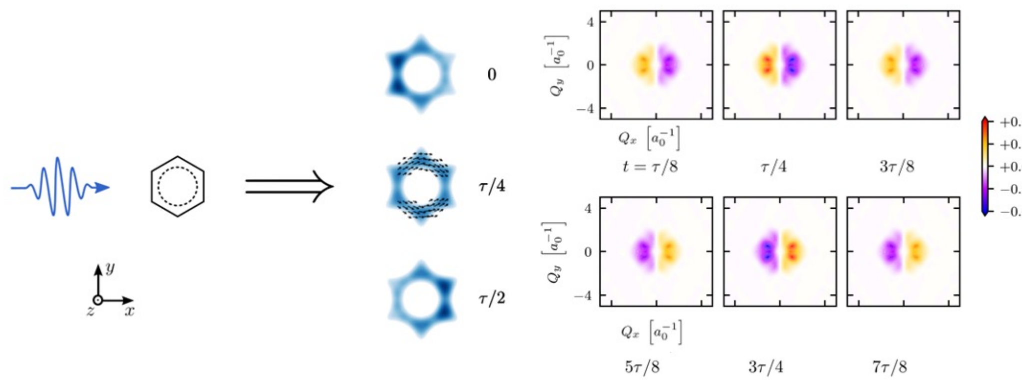
Opportunity for fully coherent HXR FEL:
HXR Diffractive Imaging of Ultrafast Charge Motion





Dixit PNAS **109** 1163 (2012)
Charge Motion in atomic hydrogen

- Time-resolved diffractive imaging has become an invaluable tool for probing molecular dynamics
- We can extend this to image electronic motion
- Requires good stabilization to external laser source. (sub-optical-cycle tagging?)

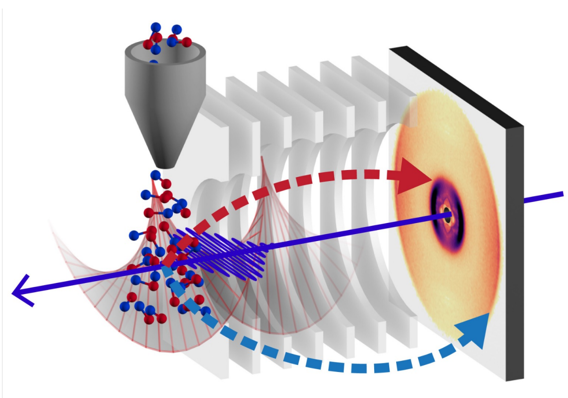


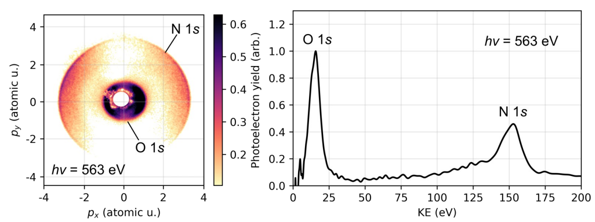
Hermann *Phys Rev Lett* 2020, **124**(1)**:** 013002
Probing Electronic Flux Density

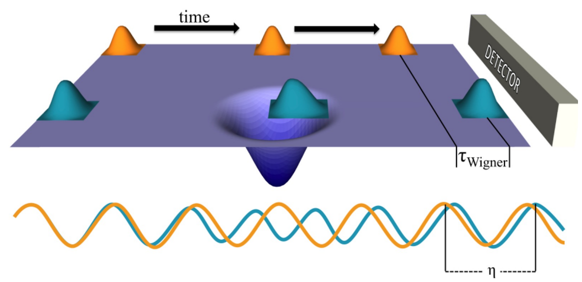
Yong Nat. Comm. **11**, 2157 (2020)
Stankus Nat. Chem. **11**, 716 (2019)
Ruddock Ang. Chem. (131) 6437 (2019)
Minitti Phys. Rev. Lett. **114**, 255501(2015)

Additional Results from LCLS with Attosecond X-ray Pulses

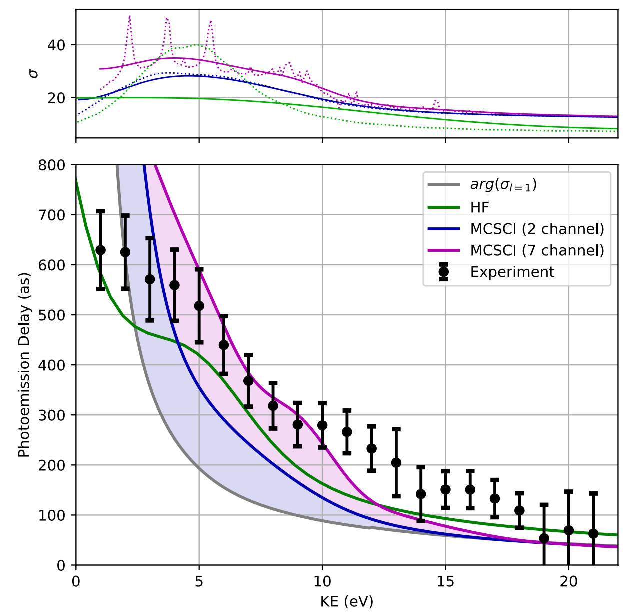
- Resolving photoemission time delays at x-ray wavelengths
- X-ray Imaging with sub-femtosecond pulses
  – Tais Gorkhover





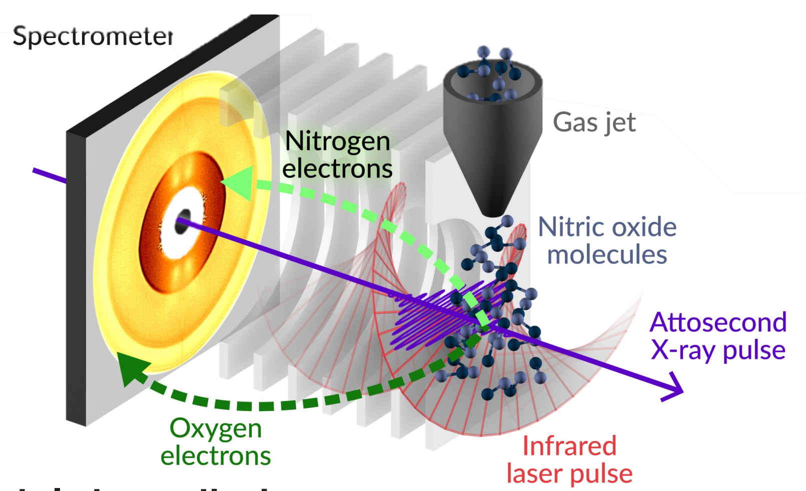


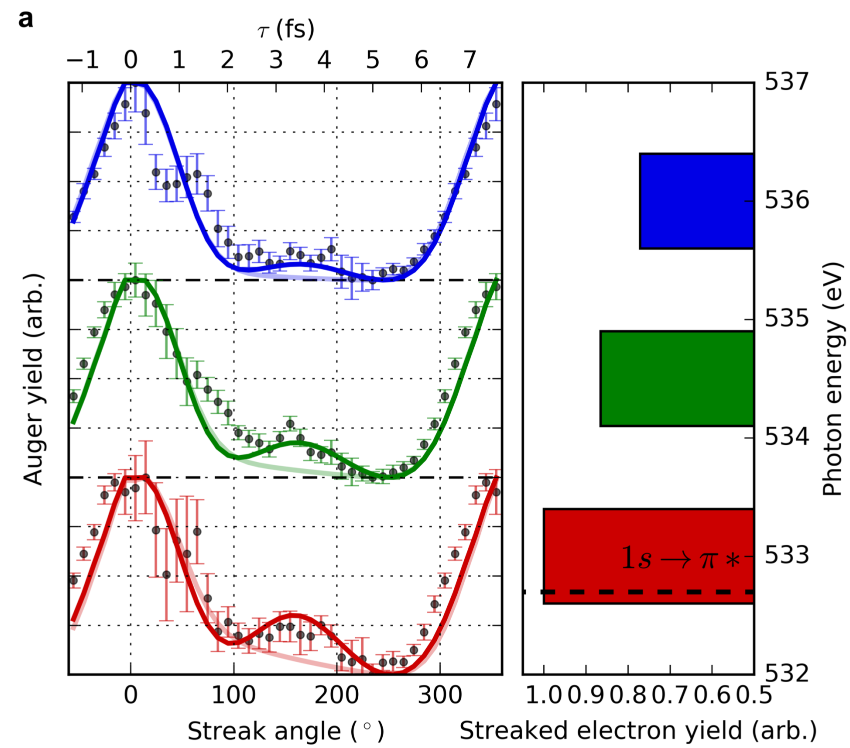
L. Argenti, *Phys. Rev. A* 95, 043426 (2017)



Additional Results from LCLS with Attosecond X-ray Pulses

- Resolving photoemission time delays at x-ray wavelengths
- X-ray Imaging with sub-femtosecond pulses
- Probing electronic coherence in core-excited states



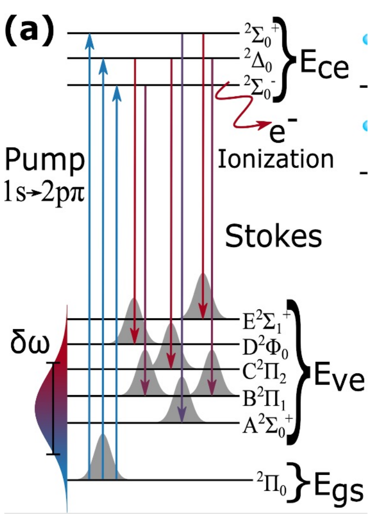


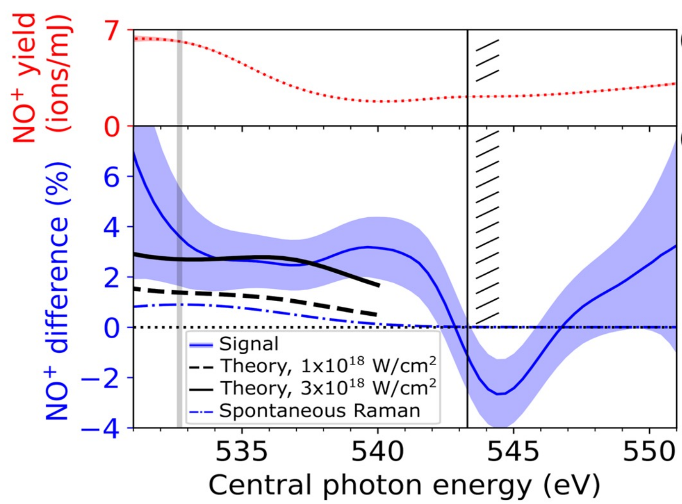
Attosecond coherent electron motion in Auger-Meitner decay

Science **375**, 285–290 (2022)

Additional Results from LCLS with Attosecond X-ray Pulses

- Resolving photoemission time delays at x-ray wavelengths
- Probing electronic coherence in core-excited states
- Probing electronic coherence in core-excited states
- Nonlinear X-ray methods for creating and probing electronic coherence





PHYSICAL REVIEW LETTERS **125,** 073203 (2020)

Editors' Suggestion | Featured in Physics

Electronic Population Transfer via Impulsive Stimulated X-Ray Raman Scattering with Attosecond Soft-X-Ray Pulses

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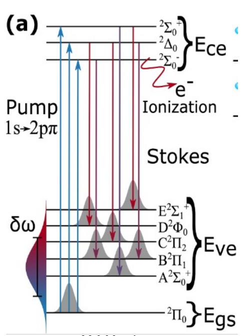
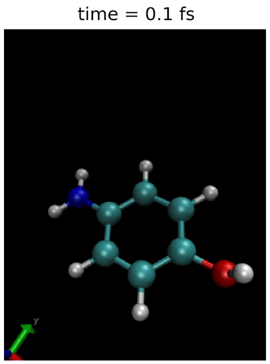
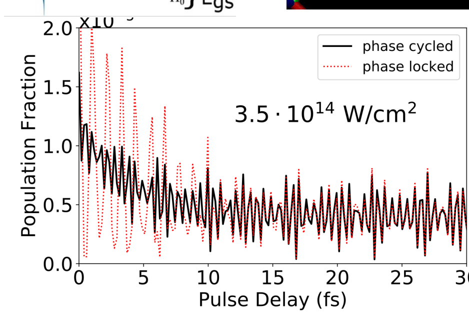
Nonlinear Spectroscopy at X-ray Wavelengths

Impulsive Stimulate Raman Scattering:
A fast kick for creating and probing charge motion







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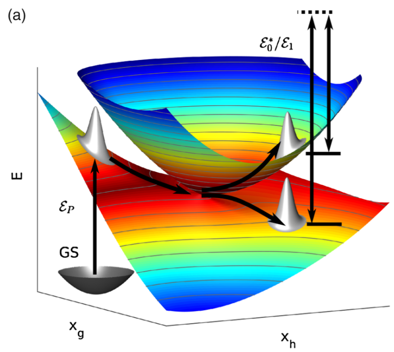
Calculation courtesy Antonio Picon

TRUECARS:
Electron dynamics (and coherence) near conical intersections



Nam JPC Lett. 12 12300 (2021)
Cavelleto PRX 11 011029 (2021)
Keefer PNAS 117 24069 (2020)
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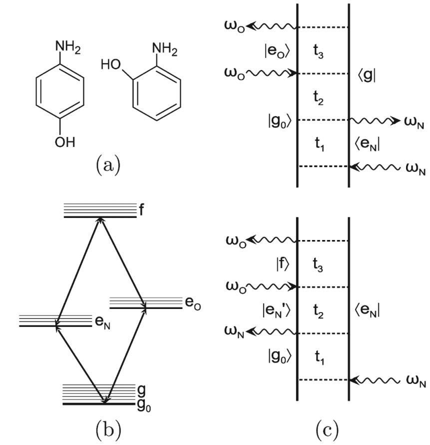
Core-hole Correlation Spectroscopy:
2D-spectroscopy to extract electronic coupling and correlation.



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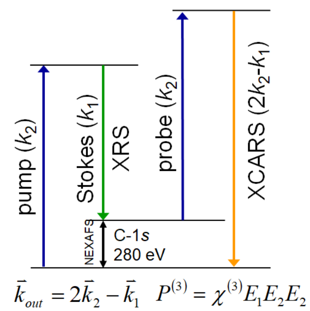
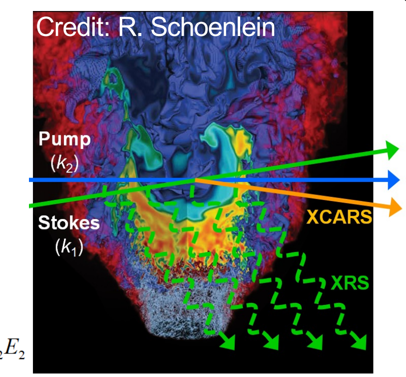
XCARS (and re-DFG):
wave-mixing technique to probe SXR XAS with HXR



$\bar{k}_{out} = 2\bar{k}_2 - \bar{k}_1 \quad P^{(3)} = \chi^{(3)} E_1 E_2 E_2$



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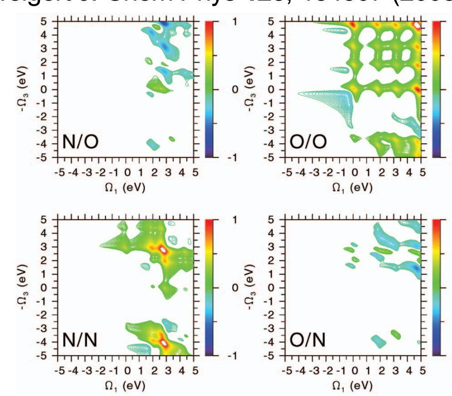
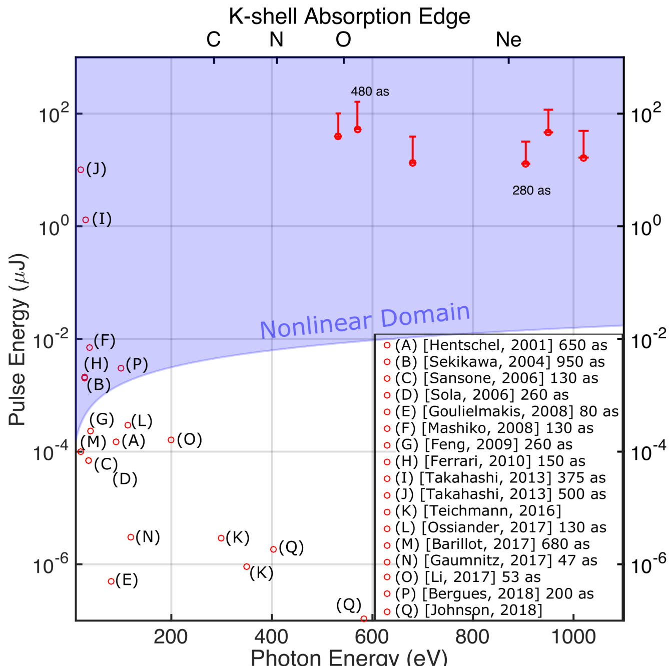
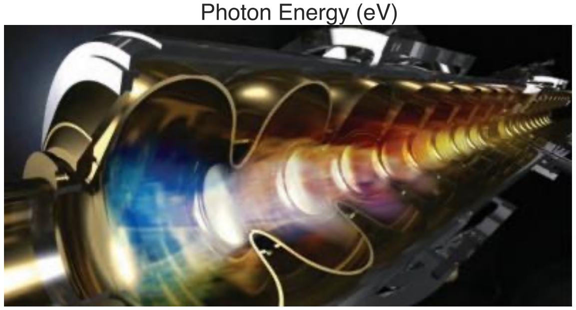


Attosecond Science for Problems in Chemical Sciences





- **We can study the ultrafast motion of electrons in chemical systems**
  - This is a crucial part of any photochemical process
  - **Can electronic coherence be important to the transfer of charges in molecules?**
    - Bottom-up approach, studying small molecules (which means fast evolution)
- **Technique development:**
  - Non-linear x-ray spectroscopies which are sensitive to electronic motion in chemical systems
  - Multi-dimensional spectroscopies to probe correlations in electronic structure with atomic-site specificity.
    - Important for linking to more complex systems.
  - **Developing machine learning approaches to data analysis of large datasets**
    - Employ machine learning to make better measurements with XFELs.
    - Better spectroscopy, better time resolution

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Attosecond Science Group @ PULSE

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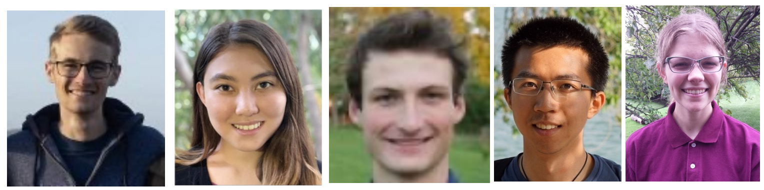

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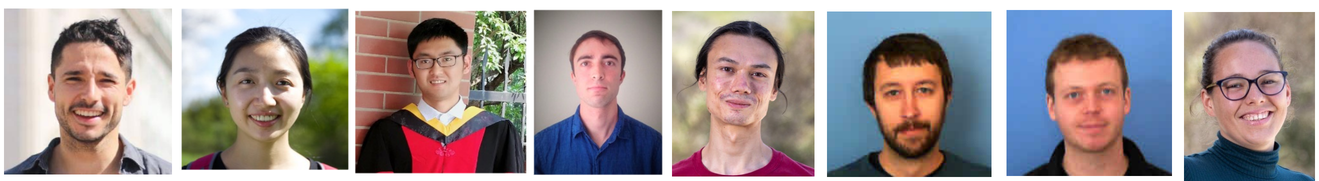

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Kansas State University

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TMO Instrument Team @ LCLS

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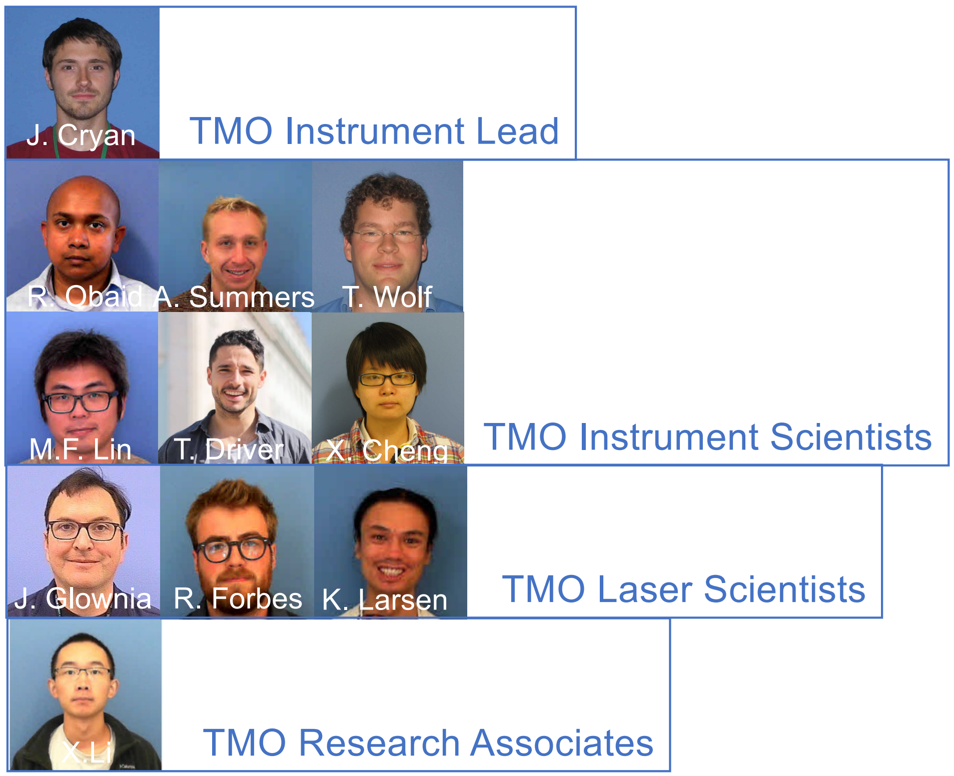

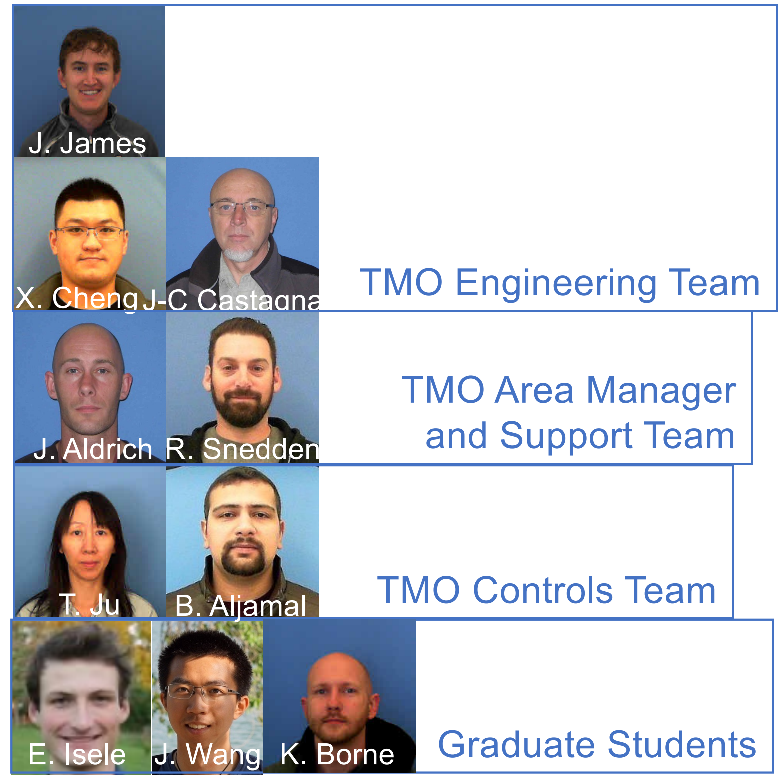

Attosecond Campaign @ LCLS – Impulsive Ionization

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