Attosecond Science with X-ray FELs

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Linac Coherent Light Source
SLAC National Accelerator Laboratory

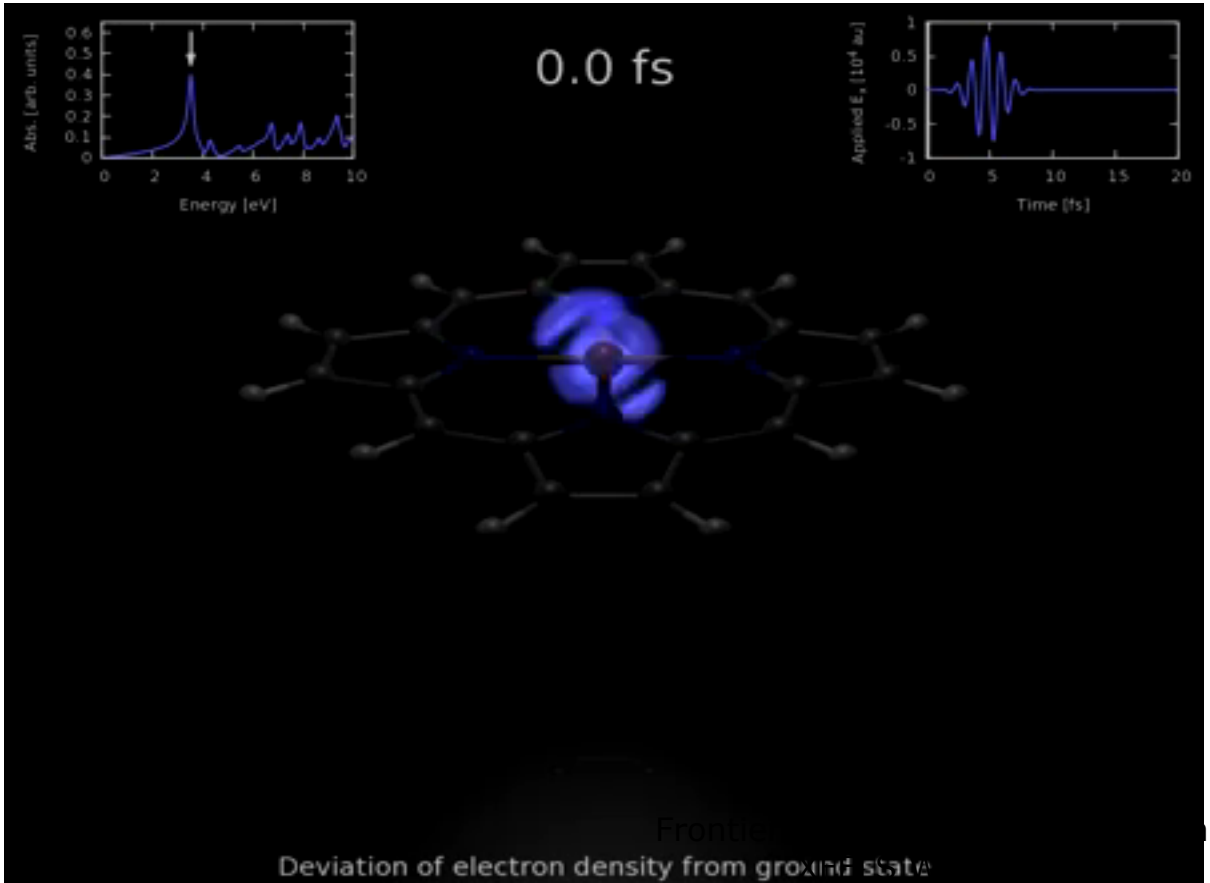
Frontiers of Physical Sciences with XFELS Workshop
November 13, 2019

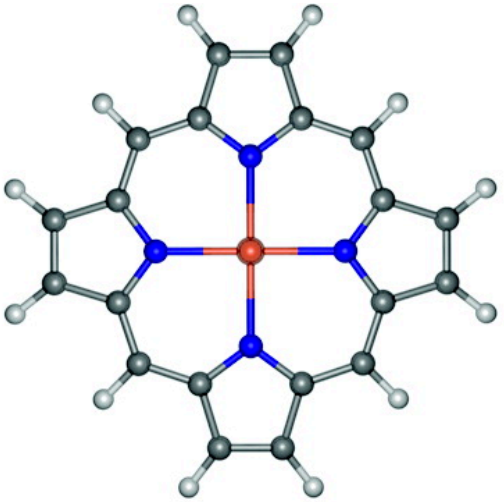
Stanford | PULSE INSTITUTE

LCLS

Coherent Electronic Phenomena

- **Electron motion is responsible for all photochemistry**
- **Our goal is to track the evolution of electrons on their natural time scales**
- **Determine how attosecond scale electronic dynamics (and coherence) effects longer timescale, femtosecond motion.**





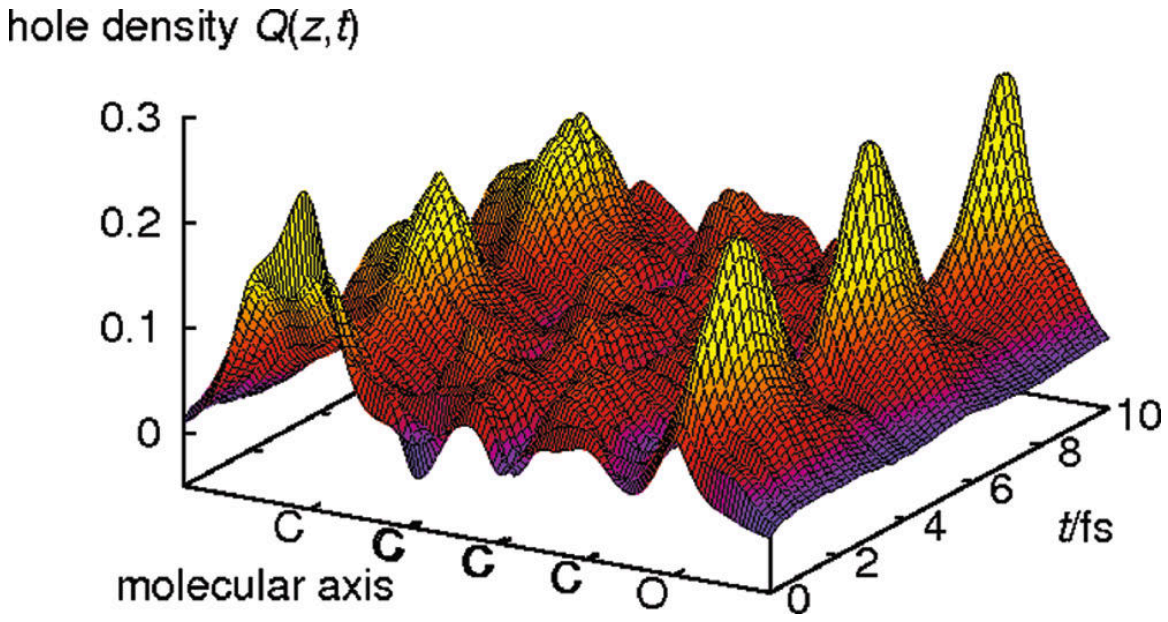
Zinc porphyrin

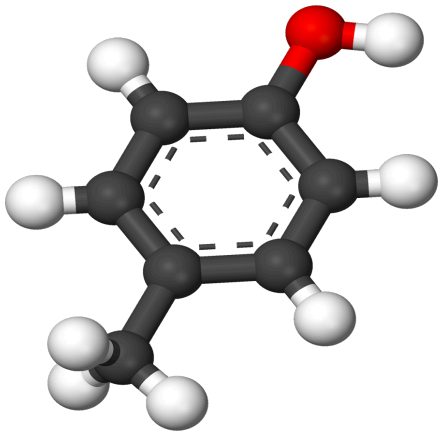
Environmental Molecular Sciences Laboratory (EMSL) @ PNNL: https://www.youtube.com/watch?v=ZYsktRlhMOg

J. Chem. Theory Comput. **7**, 1344–1355 (2011)

Coherent Electronic Phenomena

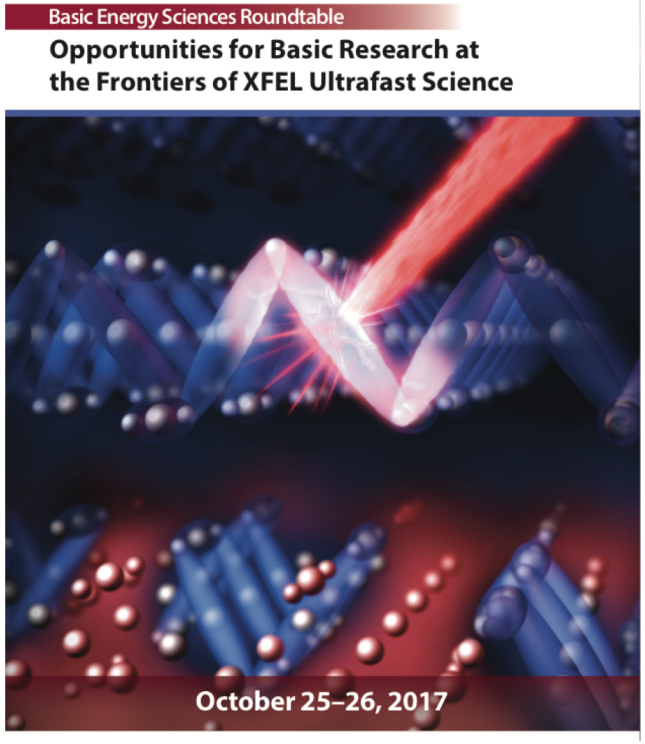
- **Ultrafast X-rays are the ideal tool to create and probe coherent electronic phenomena**
  - **Sub-femtosecond duration**
  - **Atomic site-specificity.**





Outline

- **Tools to probe electron dynamics in molecular systems**
  - **Developing an attosecond XFEL**
- **Recap of some of our work on attosecond timescale dynamics @ LCLS**
- **Opportunities for future attosecond XFELs.**



2. Priority Research Opportunities.....

2.1 PRO 1: Probing and controlling electron motion within a molecule...
Scientific Opportunities and Challenges.....
Current Status, Strategic Research and Implementation.....
Scientific Impact.....

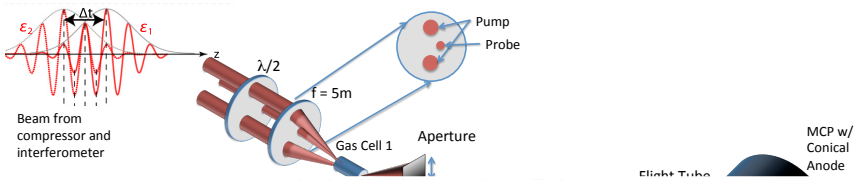
2.2 PRO 2: Discovering novel quantum phases through coherent light-matter coupling.....
Scientific Opportunities and Challenges.....
Current Status, Strategic Research and Implementation.....
Scientific Impact.....

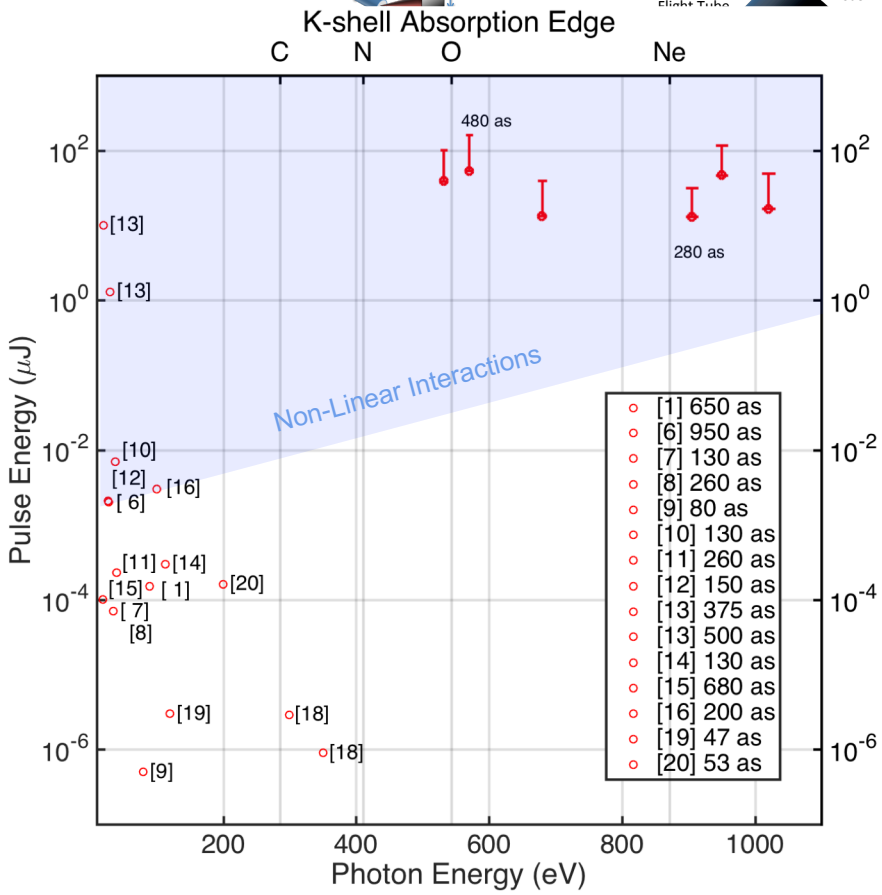
2.3 PRO 3: Capturing rare events and intermediate states in the transformation of matter.....
Scientific Opportunities and Challenges. .....
Current Status, Strategic Research and Implementation.....
Scientific Impact.....

Tools for Probing Charge Dynamics

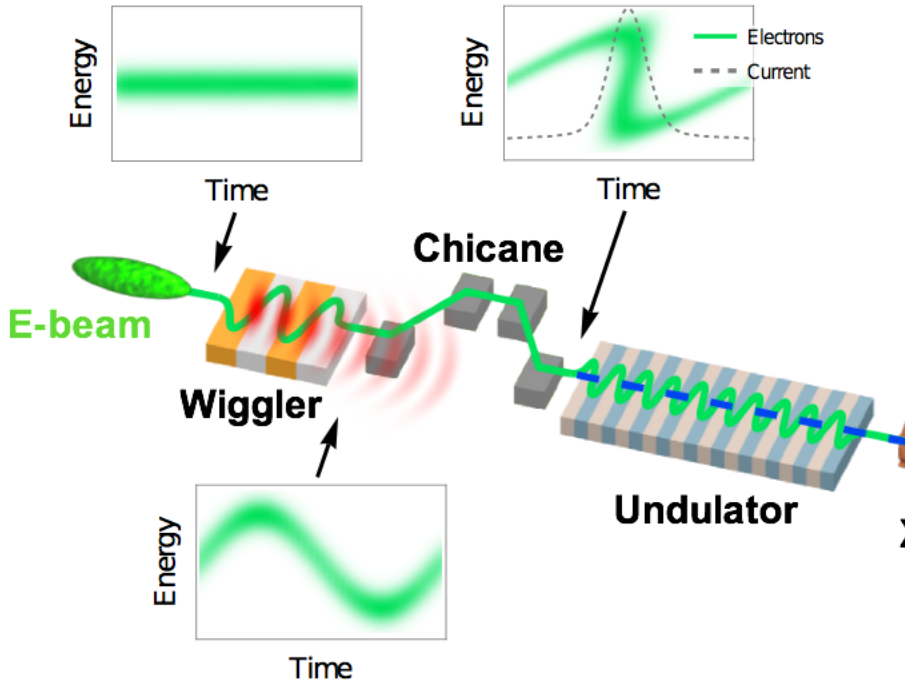
Stanford PULSE Institute LCLS

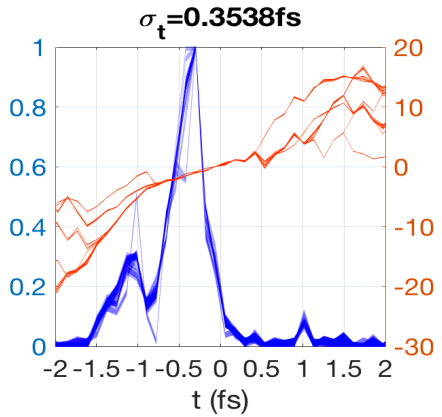
HHG Based Sources

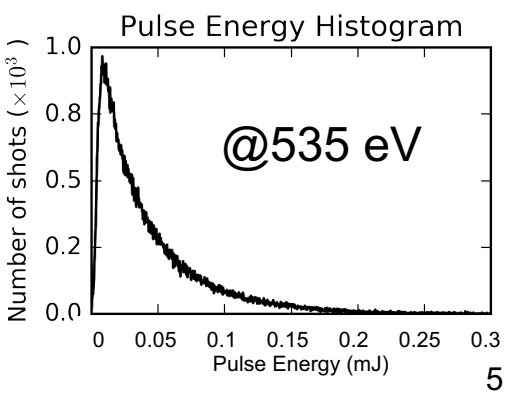




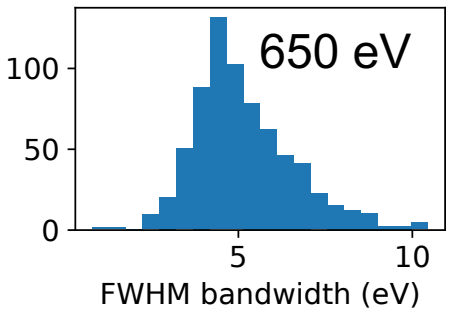
FEL Based Sources

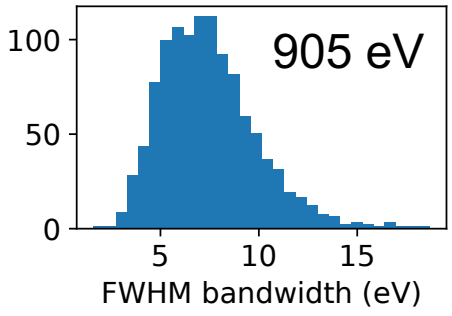


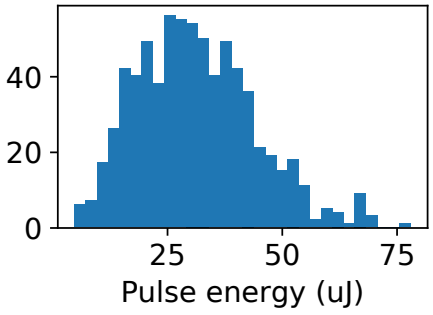


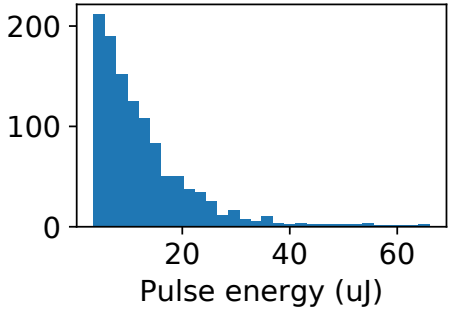


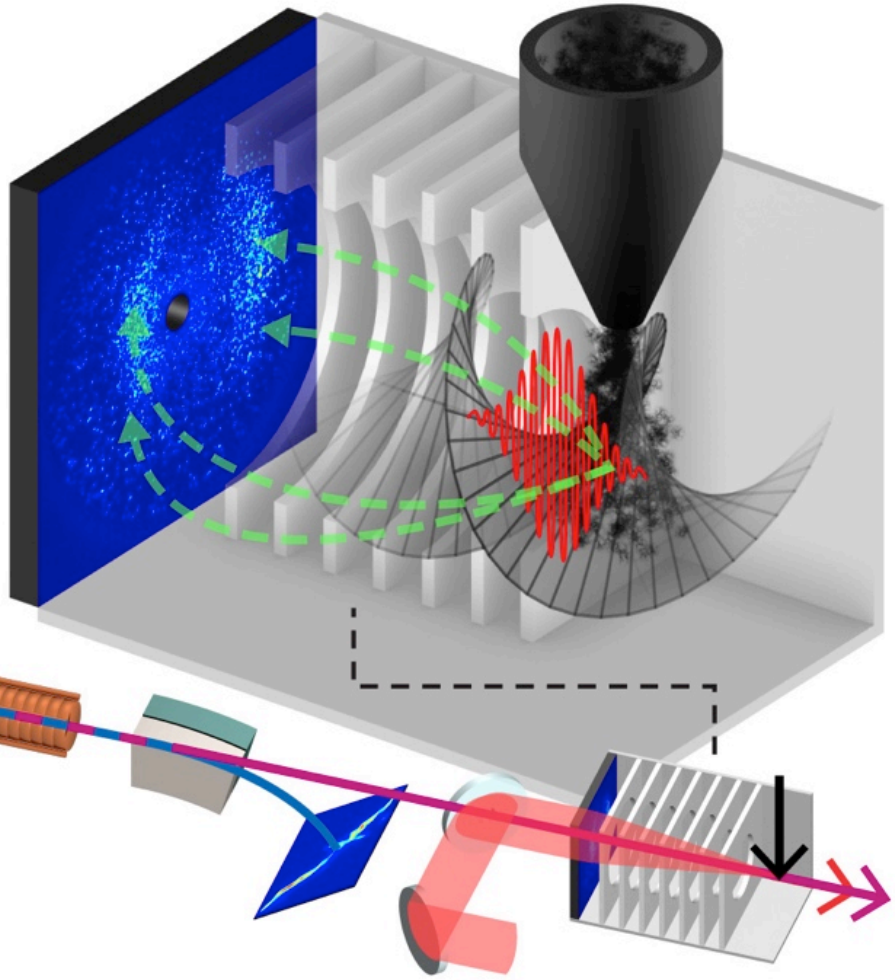
Development of Attosecond XFEL pulses







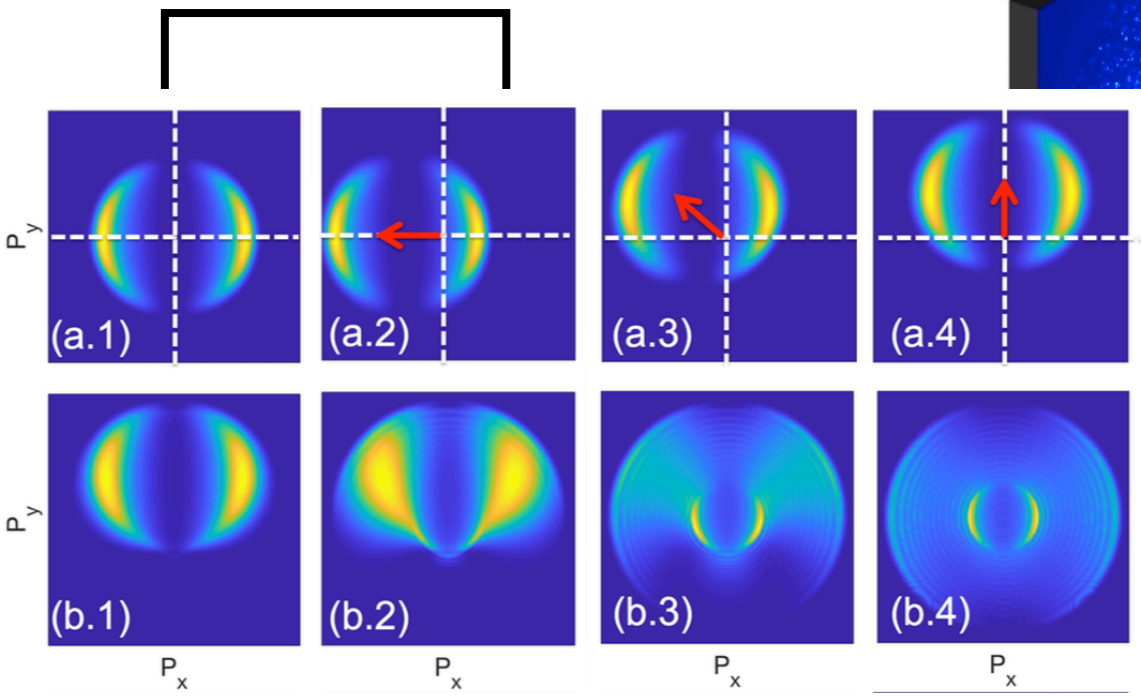


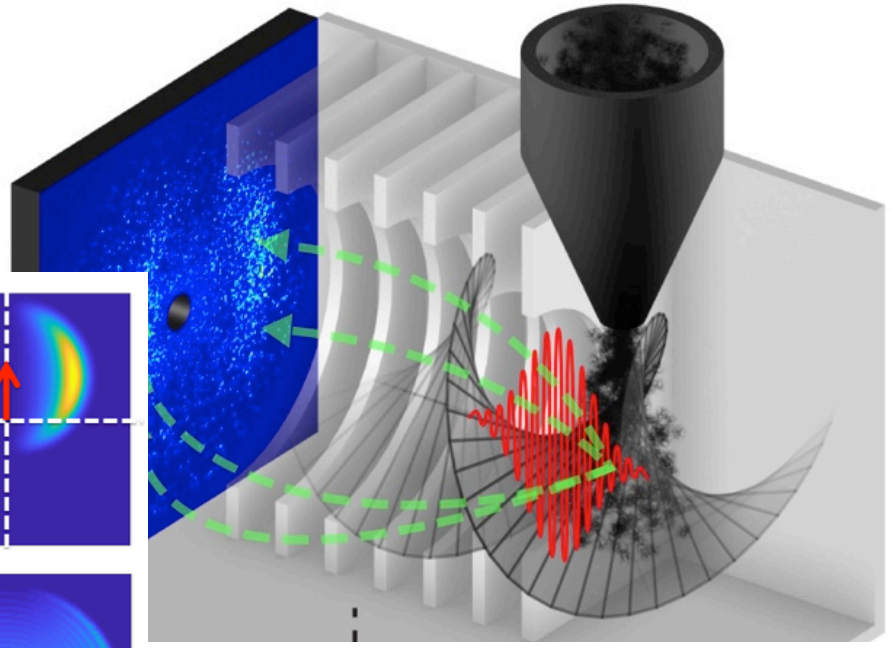


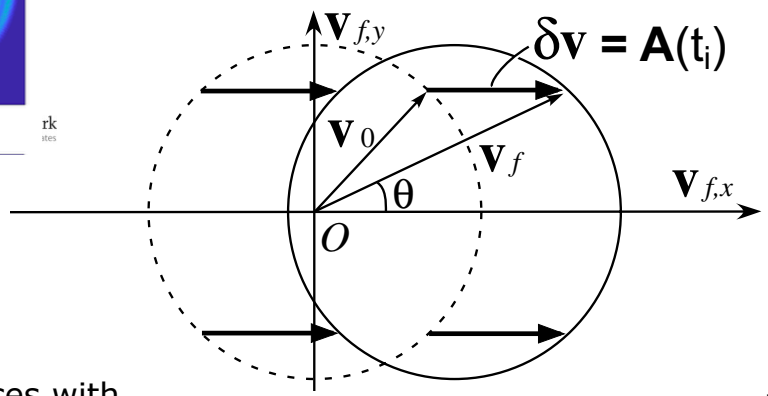
XLEAP: X-ray Laser Enhanced Attosecond pulse Production

https://arxiv.org/abs/1906.10649

Angular Streaking at FELs







A co-axial velocity map imaging spectrometer for electrons

S. Li,[1,2,a] E. G. Champenois,[3] R. Coffee,[3,4] Z. Guo,[5] K. Hegazy,[1,3] A. Kamalov,[1,3] A. Natan,[3] J. O'Neal,[1,3] T. Osipov,[4] M. Owens III,[2] D. Ray,[4] D. Rich,[2] P. Walter,[4] A. Marinelli,[2] and J. P. Cryan[3,4,b]

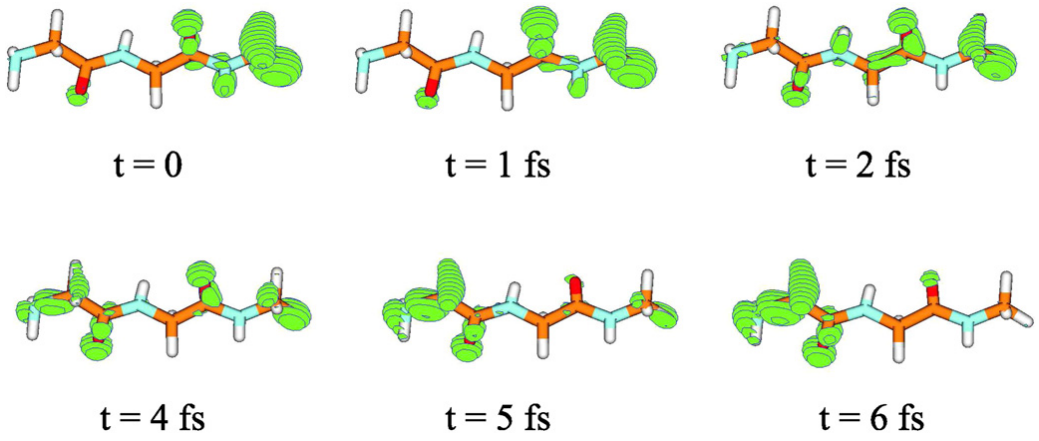
Opportunities for Attosecond Measurements of Electron Dynamics

- **The attosecond scale is the timescale for electronic motion on the atomic length scale.**
- **Coherent charge motion (Charge Migration)**
  - **Superposition of electronic states will evolve on the sub-femtosecond timescale**
  - **Could offer the possibility to control the charge localization in molecules.**
- **Time-resolving electron correlation.**
  - **The exchange–correlation term in the many-body Hamiltonian can have large effects on electronic configuration of a molecule, and thus the chemical bonding.**
  - **Non-sequential ionization**
  - **Auto-ionization, Auger Decay**

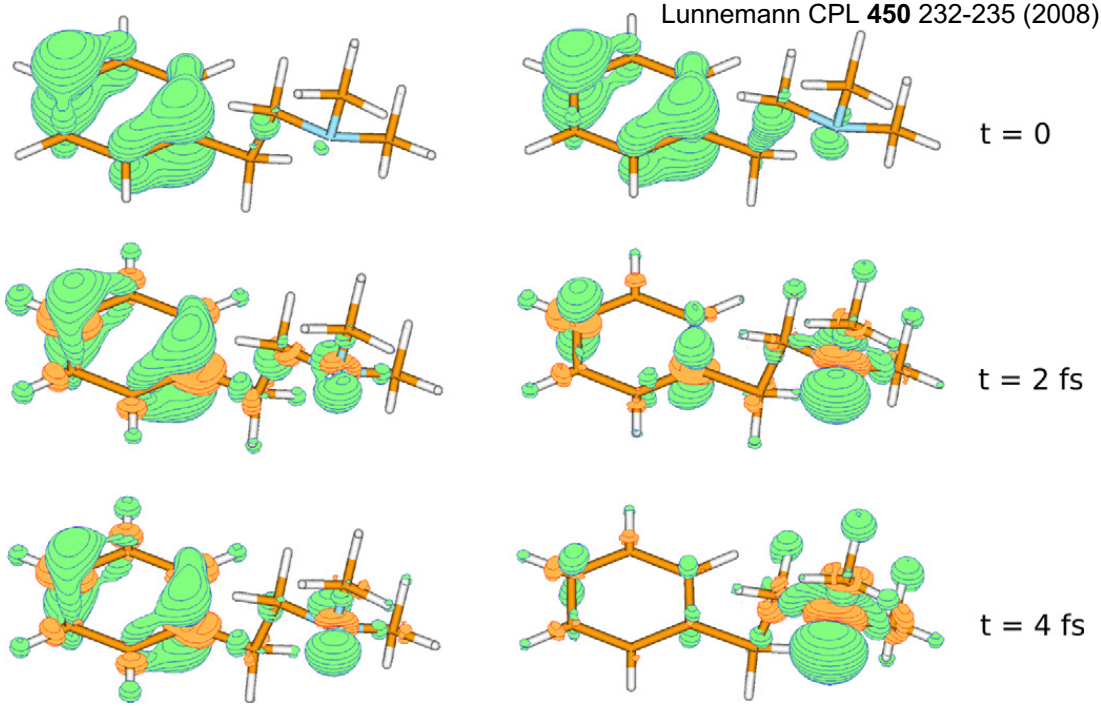
Ultrafast Charge Migration

- **Superpositions of electronic states will evolve on the sub-femtosecond timescale.**
- **Nuclear motion will alter the coherence**
- **We want to determine what role attosecond scale electronic coherence has on longer timescale, femtosecond motion (Chemistry).**
- **Attosecond-pump/attosecond-probe**

Kuleff *Chem. Phys.* **414** 100 2013

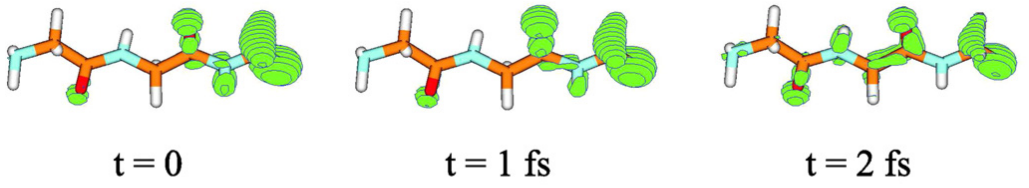


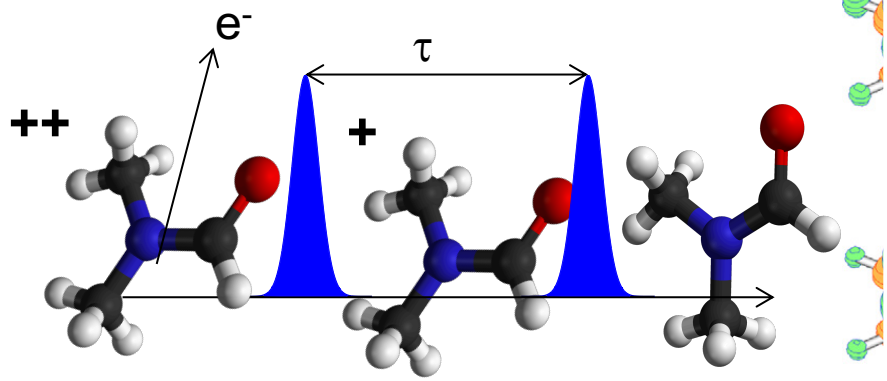
Lunnemann CPL **450** 232-235 (2008)

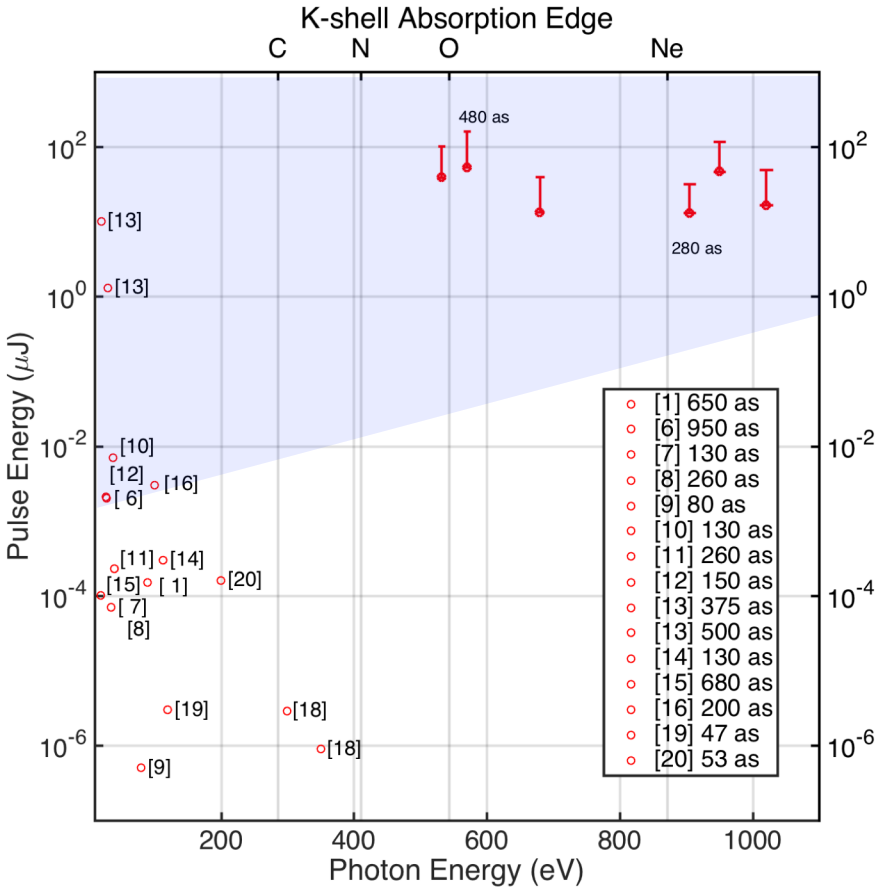


Ultrafast Charge Migration

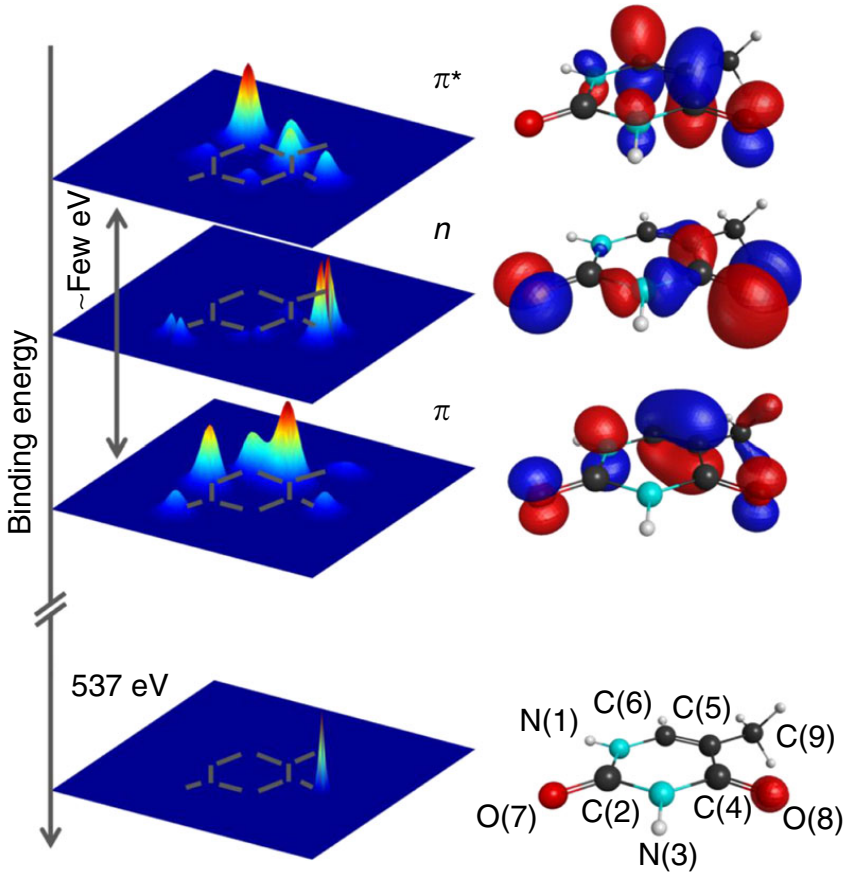
- **Superpositions of electronic states will evolve on the sub-femtosecond timescale.**
- **Nuclear motion will alter the coherence**
- **We want to determine what role attosecond scale electronic coherence has on longer timescale, femtosecond motion (Chemistry).**
- **Attosecond-pump/attosecond-probe**
- **Focus of science campaign @ LCLS**

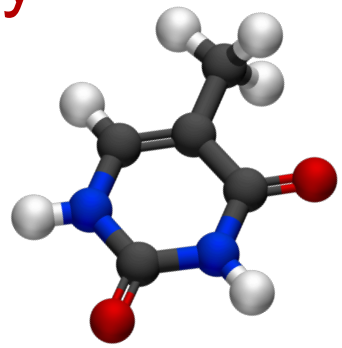




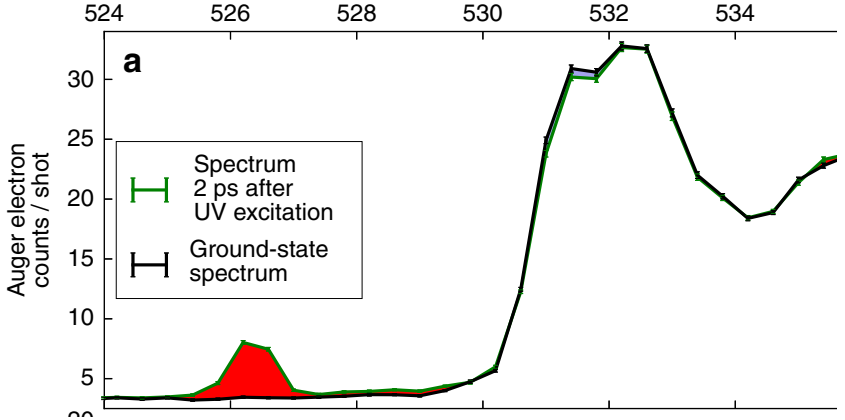


X-Ray Absorption Spectroscopy





- **X-ray absorption probes overlap of valence orbitals with core electrons**
- **Very useful for ultrafast photochemistry**



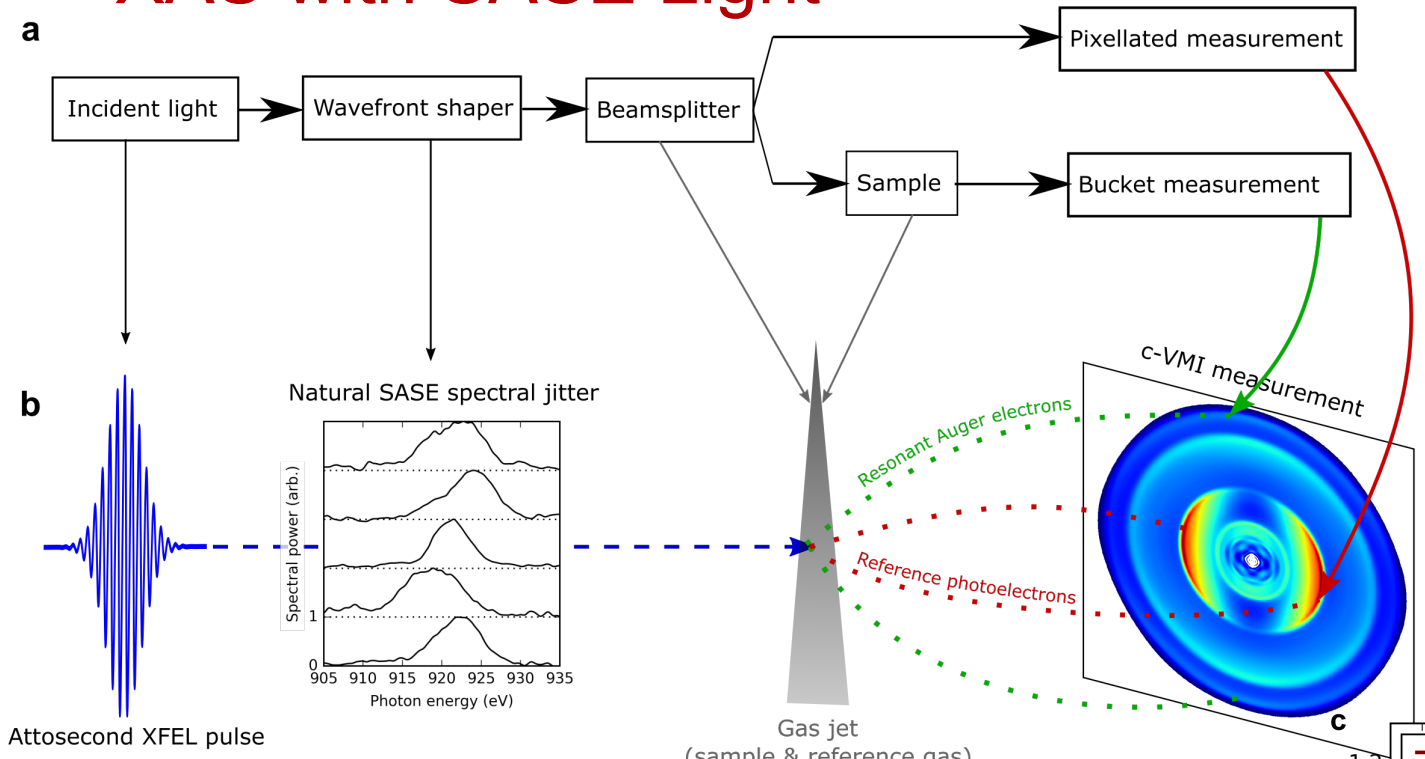
ARTICLE

DOI: 10.1038/s41467-017-00069-7 OPEN

Probing ultrafast $\pi\pi^*/n\pi^*$ internal conversion in organic chromophores via K-edge resonant absorption

T.J.A. Wolf[1], R.H. Myhre[1,2], J.P. Cryan[1], S. Coriani[3,4], R.J. Squibb[5], A. Battistoni[1], N. Berrah[6], C. Bostedt[7,8,9], P. Bucksbaum[1,10], G. Coslovich[7], R. Feifel[5], K.J. Gaffney[1,11], J. Grilj[12], T.J. Martinez[1,13], S. Miyabe[1,13,14], S.P. Moeller[7], M. Mucke[15], A. Natan[1], R. Obaid[6], T. Osipov[7], O. Plekan[16], S. Wang[1], H. Koch[1,2] & M. Gühr[1,17]

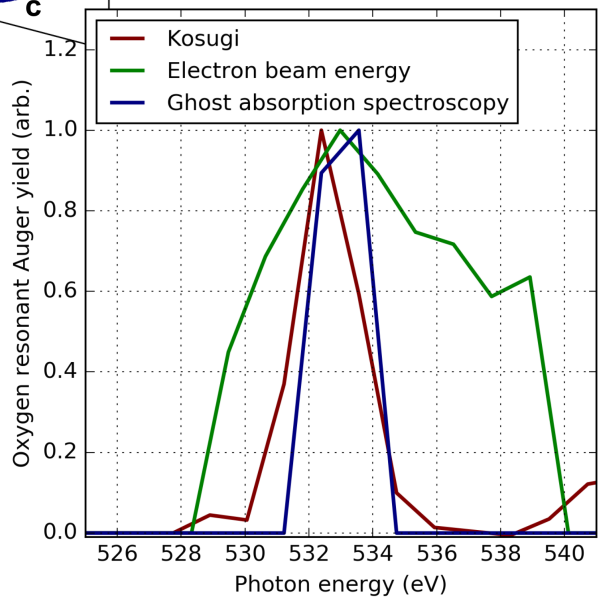
XAS with SASE Light



Short pulses -> Large bandwidth

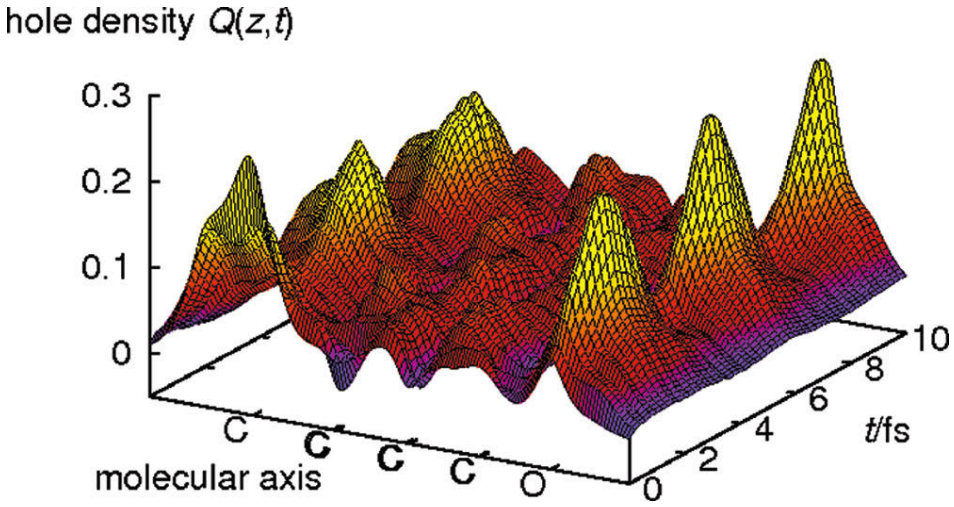
Hard to do precision Spectroscopy

Variation is key to recovering resolution

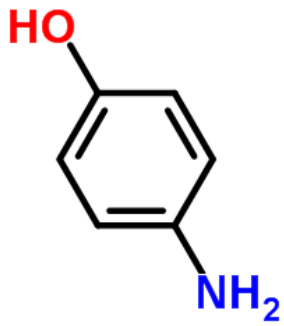


Attosecond Transient Absorption Spooktroscopy:
a ghost imaging approach to ultrafast absorption spectroscopy
Driver and Li *et al.* PCCP accepted (https://arxiv.org/abs/1909.07441)

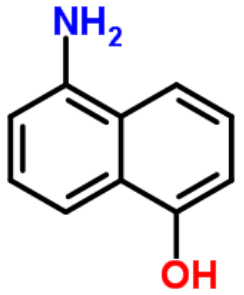
Molecular Effects on Electronic Coherence



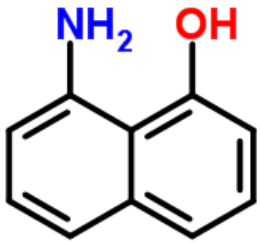
A



B



C



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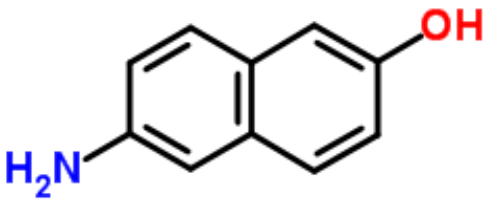

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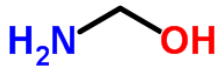
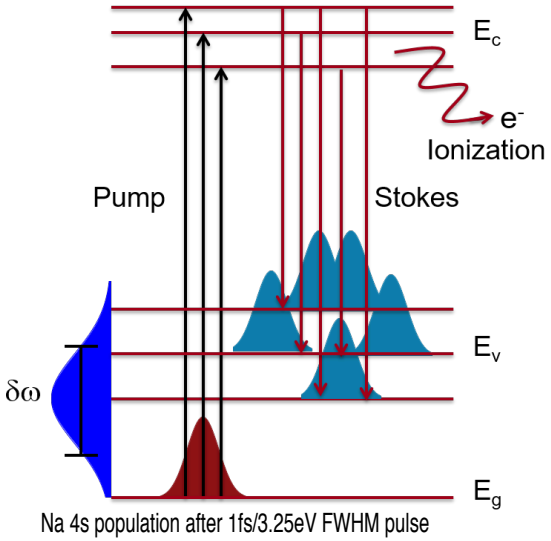


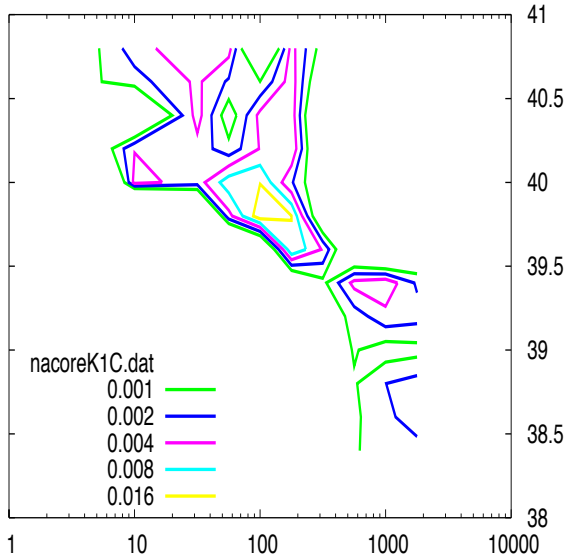
F

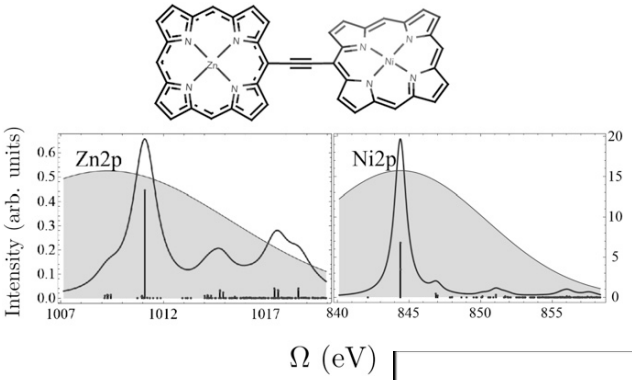
- **Can coherent electronic excitations provide a route to by-pass IVR in controlling molecular reactions?**
  - **Chem. Phys. Lett. 285, 25-33 (1998)**
  - **Nat. Photonics 8 195-204 (2014)**
- **Measure coherence lifetime**
- **Effect of structure on charge migration.**
  - **Amino-phenol derivatives.**

Impulsive Electronic Raman Scattering: A Swift Kick for Initiating Charge Motion

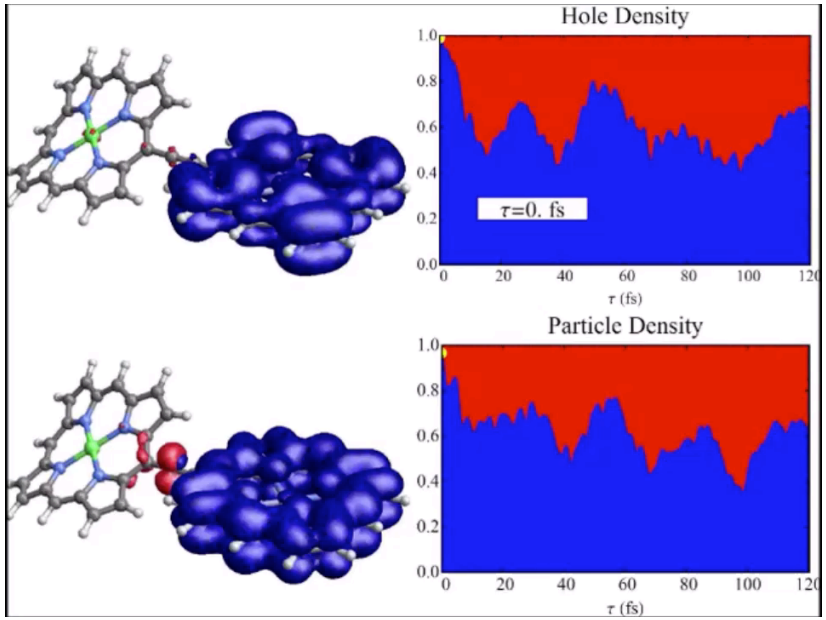


Na 4s population after 1fs/3.25eV FWHM pulse





Biggs PNAS **110** 15597 (2013)



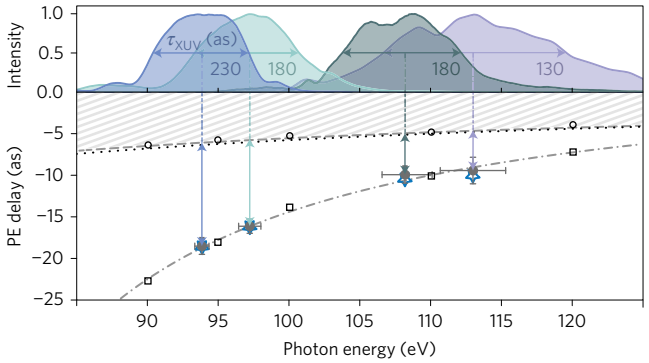
MCTDHF Calculation
(Haxton) arXiv:1609.04175

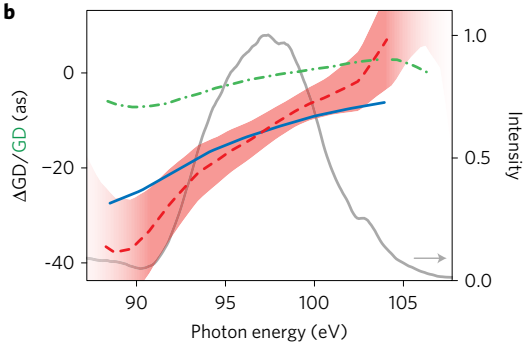
Electron Correlations in Photoionization

nature physics
ARTICLES
PUBLISHED ONLINE: 7 NOVEMBER 2016 | DOI: 10.1038/NPHYS3941

Attosecond correlation dynamics

M. Ossiander[1,2*], F. Siegrist[1,2], V. Shirvanyan[1,2], R. Pazourek[3], A. Sommer[1], T. Latka[1,2], A. Guggenmos[1,4], S. Nagele[3], J. Feist[5], J. Burgdörfer[3], R. Kienberger[1,2] and M. Schultze[1,4*]

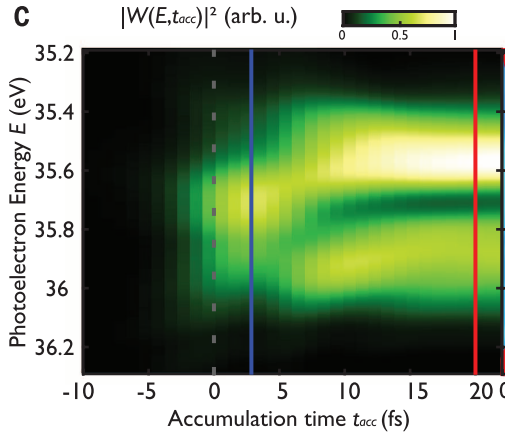




CHEMICAL PHYSICS

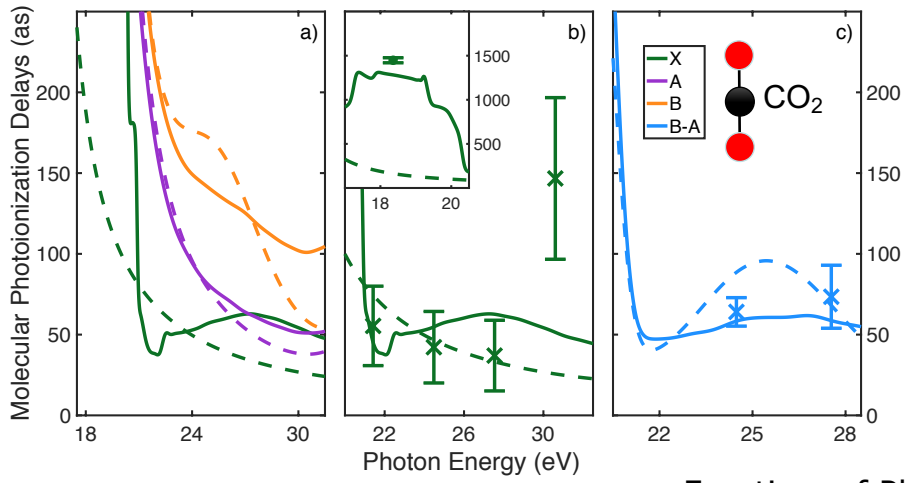
Attosecond dynamics through a Fano resonance: Monitoring the birth of a photoelectron

V. Gruson,[1*] L. Barreau,[1*] Á. Jiménez-Galan,[2] F. Risoud,[3] J. Caillat,[3] A. Maquet,[3] B. Carré,[1] F. Lepetit,[1] J.-F. Hergott,[1] T. Ruchon,[1] L. Argenti,[2†] R. Taïeb,[3] F. Martín,[2,4,5‡] P. Salières[1‡]

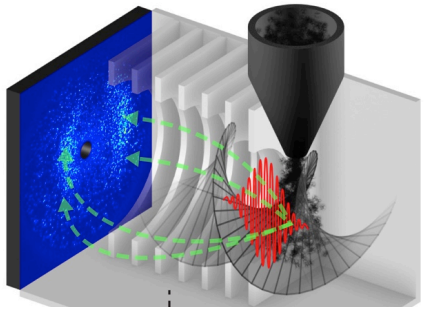


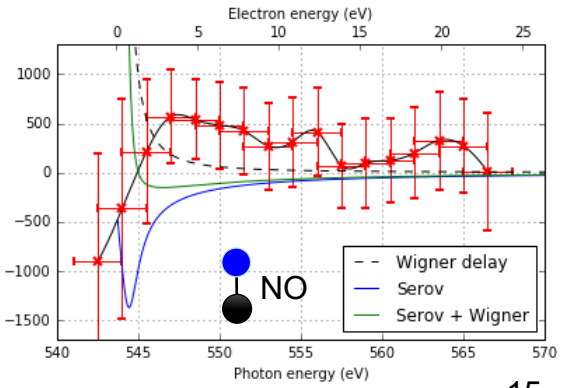
Inter-Channel Coupling in Molecular Systems in the Time Domain

A. Kamalov and J. P. Cryan in prep



Shape Resonance in X-ray Absorption in Time-Domain





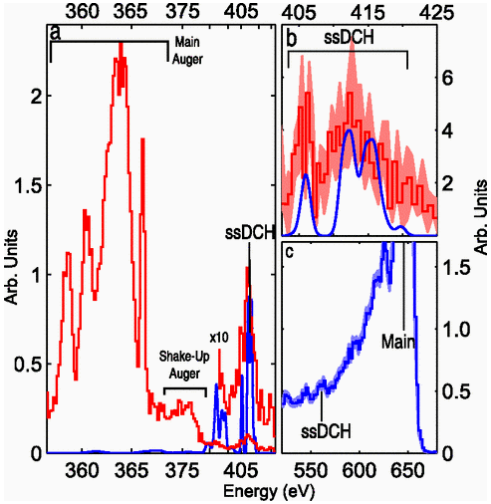
Non-sequential Ionization

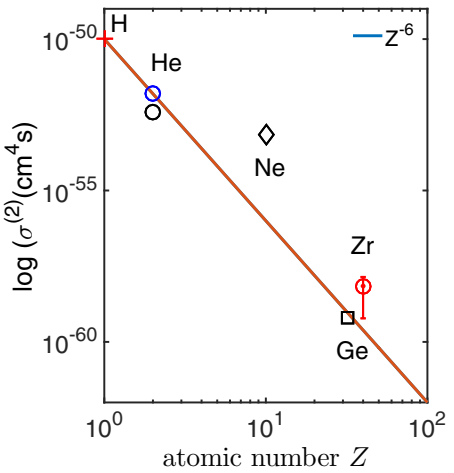
- **Early LCLS experiments observed sequential ionization**

PRL 105, 083004 (2010) PHYSICAL REVIEW LETTERS week ending 20 AUGUST 2010

Auger Electron Angular Distribution of Double Core-Hole States in the Molecular Reference Frame

James P. Cryan,[1,2,*] J. M. Glownia,[1,3] J. Andreasson,[4] A. Belkacem,[5] N. Berrah,[6] C. I. Blaga,[7] C. Bostedt,[8] J. Bozek,[8] C. Buth,[1,9] L. F. DiMauro,[7] L. Fang,[6] O. Gessner,[5] M. Guehr,[1] J. Hajdu,[4] M. P. Hertlein,[10] M. Hoener,[6,10] O. Kornilov,[5] J. P. Marangos,[11] A. M. March,[12] B. K. McFarland,[1,3] H. Merdji,[1,13] V. S. Petrović,[2] C. Raman,[14] D. Ray,[12,15] D. Reis,[1,3] F. Tarantelli,[16] M. Trigo,[1] J. L. White,[3] W. White,[8] L. Young,[12] P. H. Bucksbaum,[1,2,3] and R. N. Coffee[1,8]





PHYSICAL REVIEW A 94, 043418 (2016)

Nonsequential two-photon absorption from the *K* shell in solid zirconium

Shambhu Ghimire,[1,*] Matthias Fuchs,[2] Jerry Hastings,[3] Sven C. Herrmann,[4] Yuichi Inubushi,[5] Jack Pines,[4] Sharon Shwartz,[6] Makina Yabashi,[5] and David A. Reis[1,7,†]

- **Nonsequential Double Ionization**

$$N_{SI}^{(S)} = \int dt\, N_{GS}(t)\sigma_1^{(1)} I(t) \propto \tau$$

$$N_{DI}^{(S)} = \int dt\, N_{SI}(t)\sigma_2^{(1)} I(t)$$

$$\propto \sigma_2^{(1)} N_{SI}\, \tau \propto \sigma_1^{(1)}\sigma_2^{(1)}\tau^2$$

$$N_{DI}^{(NS)} = \int dt\, N_{GS}(t)\sigma^{(2)} I^2(t) \propto \sigma^{(2)}\tau$$

$$\sigma_1^{(1)}\sigma_2^{(1)} >> \sigma^{(2)}$$

$$\frac{N_{DI}^{(S)}}{N_{DI}^{(NS)}} = \frac{\sigma_1^{(1)}\sigma_2^{(1)}}{\sigma^{(2)}}\tau$$

Short pulse suppress sequential process

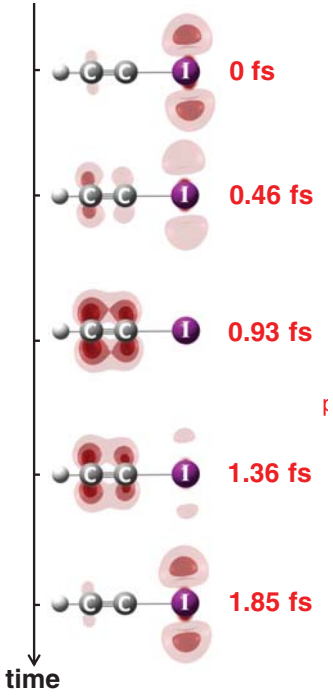
- **Push far below core-hole lifetime**
- **Time-resolve electron emission**

Molecules in Strong-Laser Fields

ATTOSECOND DYNAMICS

Measurement and laser control of attosecond charge migration in ionized iodoacetylene

P. M. Kraus,[1] B. Mignolet,[2,3] D. Baykusheva,[1] A. Rupenyan,[1] L. Horný,[1] E. F. Penka,[4] G. Grassi,[1] O. I. Tolstikhin,[5] J. Schneider,[1] F. Jensen,[6] L. B. Madsen,[7] A. D. Bandrauk,[4] F. Remacle,[2] H. J. Wörner[1]*



- **Another route to studying ultrafast charge motion**

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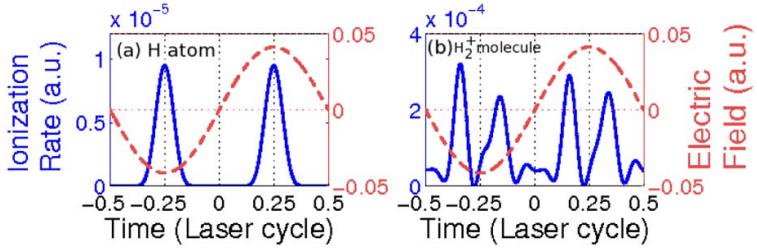
Laser-driven nonadiabatic electron dynamics in molecules

M. R. Miller, Y. Xia, A. Becker,* and A. Jaroń-Becker



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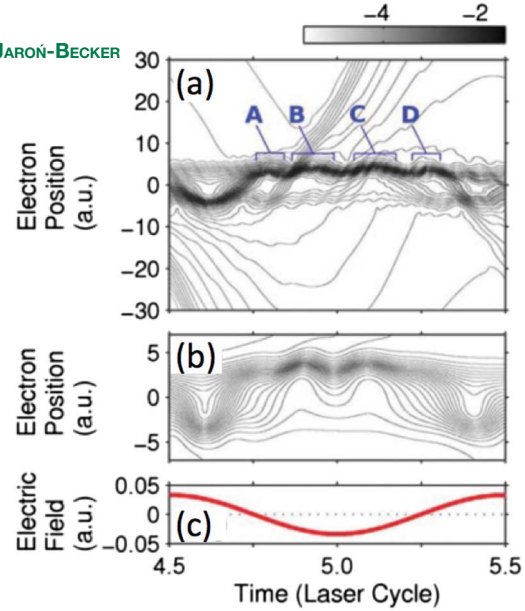
Enhanced ionisation of polyatomic molecules in intense laser pulses is due to energy upshift and field coupling of multiple orbitals

Sonia Erattupuzha[1], Cody L Covington[2], Arthur Russakoff[2], Erik Lötstedt[3], Seyedreza Larimian[1], Václav Hanus[1], Sergiy Bubin[4], Markus Koch[5], Stefanie Gräfe[6], Andrius Baltuška[1], Xinhua Xie (谢新华)[1], Kaoru Yamanouchi[3], Kálmán Varga[2] and Markus Kitzler[1,7]



- **Probing on electron motion in strong-fields**
- **Non-linear probing (Raman)**

Summary

- Sub-femtosecond X-ray pulses provide a unique opportunity to study the motion of electrons on the atomic length scale.
- This is critical for understanding the role of electronic coherence in photochemical reactions
- Such technology would also allow for time-resolving electron-electron interactions (or correlations)
- Requirements:
  - 10-200 μJ pulse energies
  - Sub-femtosecond pulse duration
    - Within factor 3 of transform limit would be most useful.
  - Soft X-ray (< 1keV)
  - Two-color pulses,
    - 100's eV separations between colors.
    - Tunable delay (nice to scan through zero)
  - 1 μm^2 focus
  - Synchronized NIR laser field
- Measurement can be better (cheaper, easier) than control.