Ultrafast coherent electron motion in molecules measured with X-ray free-electron lasers

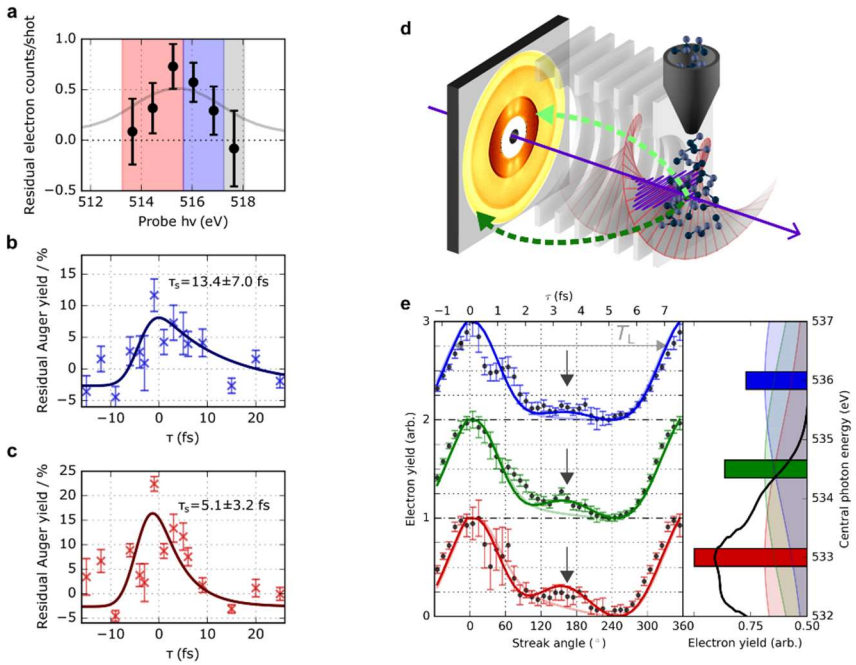
Taran Driver[1,2]

1. *Stanford PULSE Institute, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, U.S.A.*
2. *Linac Coherent Light Source, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, U.S.A.*

Coherent electron motion is initiated and probed on its natural attosecond timescale with few-femtosecond and attosecond soft X-ray pulses from an X-ray free-electron laser (XFEL). We exploit the tunability of the XFEL source to both control the coherent motion, and time-resolve transient valence electron density with atomic site- and state-specificity.

A fundamental question for studies on the attosecond timescale is the dynamical behavior of electronic quantum superposition states. We present recent work performed at the Linac Coherent Light Source (LCLS) on the creation and real-time measurement of coherent electron motion in molecules with an XFEL [1,2]. In an X-ray pump/X-ray probe experiment performed with a few-femtosecond two-color soft X-ray pulse pair, we time-resolve the decay of an inner valence hole to a series of 1-hole-2-particle satellites *via* frustrated Auger-Meitner decay. Inner valence ionization of the isopropanol molecule coherently populates a manifold of highly correlated states in the molecular cation. With a probe pulse resonantly tuned to the oxygen 1s to inner valence transitions, we access the few-femtosecond evolution of the inner valence electron hole with site- and state-specificity.

In a second experiment, we employ attosecond angular streaking with attosecond X-ray pulses [3] tuned to the oxygen 1s→2π resonance in nitric oxide (NO) to produce a coherent superposition of O $1s^{-1}$ $2\pi^2$ core-excited states. The coherent electronic motion of this core-excitation is time-resolved by monitoring the Auger-Meitner electron current using angular streaking. We observe a coherent revival in electron yield corresponding to a rephasing between different angular momentum states. Control of the coherent motion is demonstrated by tuning the central photon energy of the exciting X-ray pulse.



a, b & c, Site- and state-specific measurement of inner valence hole dynamics in photoionized isopropanol. a) pre-edge oxygen absorption of isopropanol at short (<~1 fs) delays following inner valence ionization. b) delay dependence of high-energy (blue) region of absorption feature, corresponding to higher lying inner valence hole state, c) delay-dependence of low-energy region of absorption feature.

d & e, Real-time observation of coherent electron motion in core-excited NO. d) experimental layout for angular streaking of NO molecules in a co-axial velocity map imaging spectrometer [4]. e) control of coherent revival in Auger-Meitner electron current by tuning the central photon energy of the attosecond X-ray pulse.

References

[1] T. Barillot, O. Alexander, B. Cooper, T. Driver *et al.,* under review
[2] S. Li*, T. Driver* *et al.*, under review
[3] J. Duris*, S. Li*, T. Driver *et al.*, *Nature Photonics* **14**, 30–36 (2020)
[4] S. Li *et al.*, *AIP Advances* **8**, 115308 (2018)