CFEL SCIENCE

SEMINAR

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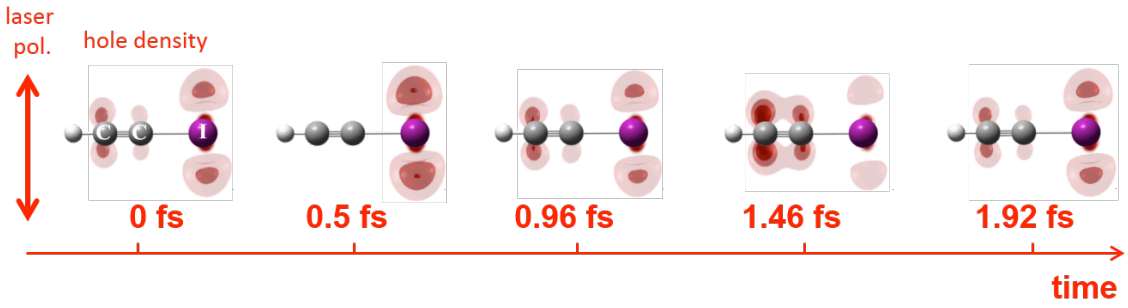
Hans Jakob Wörner

Laboratory of Physical Chemistry, ETH Zurich, 8093 Zurich, Switzerland

Attosecond charge migration and its laser control

Attosecond charge migration has been theoretically identified as one of the key manifestations of electron correlation in the time domain [1]. Experimental implementation of these ideas has remained difficult because of (i) the required temporal resolution, (ii) the selectivity in preparation of the initial hole state and (iii) the ability to trace the time-dependent spatial structure of the hole. Here, we present new developments in high-harmonic spectroscopy that overcome all of these obstacles.

Iodoacetylene (HCCI) serves as the exemplary molecule because HCCI+ displays configurational mixing between its two lowest electronic states (X+ and A+), these two states are strongly dipole coupled by a parallel transition and are well isolated from other electronic states. We impulsively orient the molecules [2,3] and systematically measure the even and odd harmonic intensities and phases of high-harmonic emission from the oriented molecules using multiple wavelengths (0.8, 1.3 and 1.8 mm) and intensities. This complete set of experimental data, combined with a novel theory that includes electron correlation in the description of high-harmonic generation, enables us to experimentally reconstruct both the configuration-interaction parameters and the time-dependent populations and relative phases of the electronic eigenstates as a function of time. Aligning HCCI perpendicular to the probing field, we turn off the effect of the laser field and reconstruct correlation-driven charge migration:



Aligning the molecules parallel to the field, we reconstruct charge migration dominated by the laser field. Varying the wavelength of the laser field [4] enables us to demonstrate extensive control over attosecond charge migration, with population transfers reaching 100%. These results open new perspectives for high-harmonic spectroscopy. Experimental access to configuration-interaction parameters has not been previously demonstrated. The ability to control charge migration through laser fields, provides new opportunities for attosecond time-scale control over molecular reactivity.

[1] L. S. Cederbaum and J. Zobeley, Chem. Phys. Lett. 307, 205 (1999)
[2] P. M. Kraus, A. Rupenyan and H. J. Wörner, Phys. Rev. Lett. 109, 233903 (2012)
[3] P. M. Kraus, D. Baykusheva and H. J. Wörner, Phys. Rev. Lett. 113, 023001 (2014); J. Phys. B 47, 124030 (2014)
[4] A. Rupenyan, P. M. Kraus, J. Schneider, and H. J. Wörner, Phys. Rev. A 87, 031401 (2013); Phys. Rev. A 87, 033409 (2013)

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