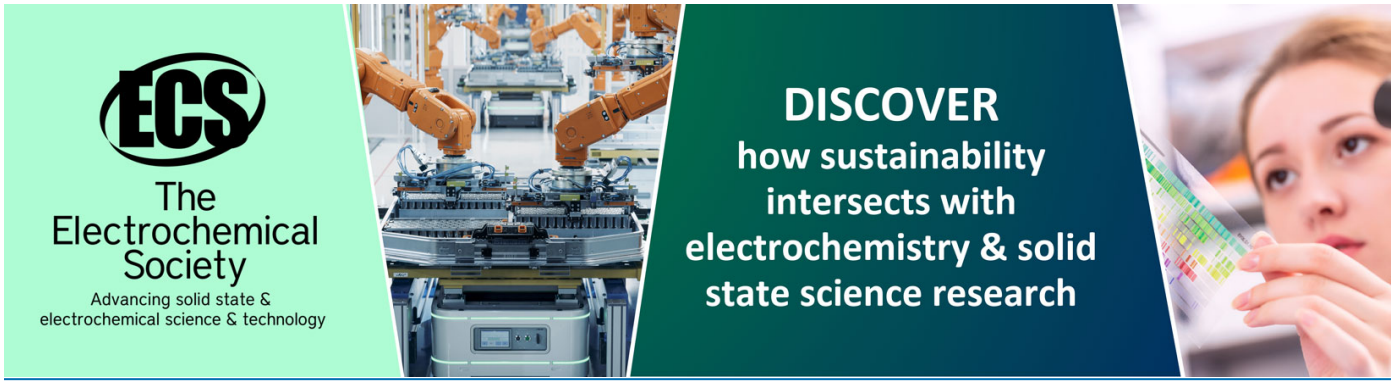
Attosecond charge migration and its laser control



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Attosecond charge migration and its laser control

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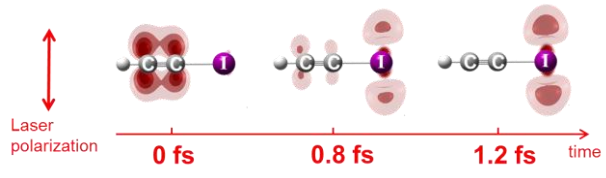
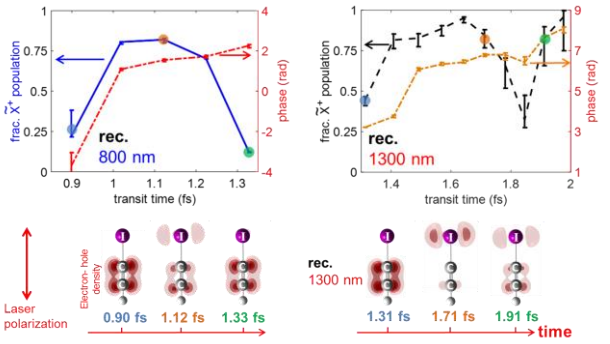
Synopsis: We advance high-harmonic spectroscopy to resolve molecular charge migration in time and space and simultaneously demonstrate extensive control over the process. A multidimensional approach enables us to reconstruct both quantum amplitudes and phases with a resolution of better than 100 attoseconds and to separately reconstruct field-free and laser-driven charge migration. Our techniques make charge migration in molecules measurable on the attosecond time scale and open new avenues for laser control of electronic primary processes.

The spatial migration of an electron hole following excitation or ionization is a fundamental process in nature and engineering. Charge migration controls the regioselectivity of chemical reactions, energy transport in biological systems and underlies molecular electronics. Moreover, charge migration following ultrafast ionization has been predicted to provide direct information on electron correlation in molecules [1]. The extreme spatio-temporal scales of charge migration pose major challenges to the latest techniques of time-domain spectroscopy.

Here, we experimentally study charge migration on the attosecond time scale and demonstrate laser control over the dynamics. We develop high-harmonic spectroscopy (HHS) further to perform a direct reconstruction of time-dependent populations and phases of electronic eigenstates from measured amplitudes and phases of the harmonic radiation with a resolution of about 100 as. Photorecombination dipole moments and the angular variation of strong-field ionization rates of the relevant eigenstates of the cation are the necessary input in the reconstruction. This procedure allows us to characterize field-free charge migration in spatially-oriented iodoacetylene (ICCH) molecules as depicted in the picture below.



We further demonstrate extensive laser control over the charge migration by controlling the orientation of the molecules [2,3], which enables us to separately reconstruct field-free and laser-driven charge migration. The laser-driven charge migration fundamentally differs from the field-free process. We find significant population transfer between the two relevant electronic eigenstates for both investigated wavelengths (800 nm and 1300 nm) as depicted below. This is mediated by a strong dipole coupling between the two lowest-lying eigenstates of the cation.



Our results enable the direct reconstruction of electronic state populations. They thus constitute a unique experimental access to the rich variety of field-free charge migration in molecules. Moreover, laser-control over charge migration in molecular systems opens new perspectives for controlling chemical reactions on the electronic time scale [4].

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