



am Donnerstag, den 11. Juni 2015
spricht um 16 Uhr c. t. im Großen Hörsaal

Prof. Dr. Hans-Jakob Wörner
(ETH Zürich)

zu dem Thema

Controlled attosecond dynamics in molecules

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.

In this talk, I will discuss the experimental investigation of charge migration in a molecular cation on the attosecond time scale. High-harmonic spectroscopy (HHS) has been developed further to perform a direct reconstruction of time-dependent populations and phases of electronic eigenstates with a resolution of 70-130 as. The reconstruction employs measured amplitudes and phases of the high-harmonic emission from oriented molecules at multiple wavelengths of the driving field.

We demonstrate this technique using iodoacetylene (ICCH) as an example. We use control over the spatial orientation of the molecule [1-3] to separate field-free from laser-controlled charge migration. For molecules aligned perpendicular to the laser polarization, charge migration takes place under conditions similar to the absence of an external field. In particular, we determine the shape of the hole created by strong-field ionization and its subsequent time evolution. We further demonstrate extensive laser control over charge migration by orienting the molecules parallel or anti-parallel to the laser polarization. The laser-controlled charge migration fundamentally differs from the field-free process. We find significant population transfer between the two lowest field-free electronic eigenstates for both investigated wavelengths (800 nm and 1300 nm). The population transfer furthermore strongly depends on the head-to-tail orientation of the molecules.

References

- [1] P. M. Kraus, A. Rupenyan, H. J. Wörner, Phys. Rev. Lett. 109, 233903 (2012)
- [2] P. M. Kraus, D. Baykusheva, H. J. Wörner, Phys. Rev. Lett. 113, 023001 (2014)
- [3] P. M. Kraus et al., Nat. Commun. 6, 7039 (2015)