

# Atoms and Molecules Changing Shape in Strong Laser Fields — Observed by Attosecond XUV Spectroscopy and Femtosecond X-Ray Imaging

Thomas Pfeifer

*Max-Planck Institute for Nuclear Physics, Heidelberg, 69117, Germany*

email: tpfeifer@mpi-hd.mpg.de

X-ray free-electron lasers (FELs) and attosecond-pulsed high-harmonic generation (HHG) are the products of two parallel — still ongoing — technological revolutions. Providing short bursts of high-frequency light, they are the ideal tools to capture the ultrafast motion of electrons around atoms or molecules. Using these tools along with novel extreme-ultraviolet(XUV) spectroscopy or X-Ray imaging approaches allows insights into the natural dynamics inside these small quantum systems. Moreover, one can observe the effects of strong laser fields pushing and pulling on the electrons, thus modifying the fundamental atomic or molecular spectral and structural properties, starting from most simple atoms and progressing towards increasingly complex molecules.

The first part of this talk covers how to measure, reconstruct, and control two-electron dynamics in doubly-excited states of Helium [1,2]. A two-dimensional XUV spectroscopy scheme employing attosecond pulses characterizes the quantum beats observed for a two-electron wave packet [3]. Transfer of this method to inner-valence transitions in Neon atoms [4] allows the measurement of single-XUV-photon forbidden transitions. This inner-valence/-core site specificity in two-dimensional spectroscopy also paves a route towards direct time-resolved measurements of localized site-to-site electron transfer and charge-migration dynamics in polyatomic molecules.

On the other hand, strong laser fields can change the spatial-structural shape of molecules and break symmetries. In an effort to directly image such processes in C<sub>60</sub> fullerenes in near-infrared (NIR) light, we performed a time-resolved x-ray diffractive imaging experiment at the Linac Coherent Light Source (LCLS). Measurements show different time scales for the deformation and fragmentation of C<sub>60</sub> depending on the NIR laser intensity.

For the future, the combination of time-resolved spectroscopy and imaging promises deep understanding of the fundamental processes at work in laser-driven molecules, and thus may continue to fuel the development of laser-directed “coherent chemistry”.

## References:

- [1] C. Ott, A. Kaldun, L. Argenti, P. Raith, K. Meyer, M. Laux, Y. Zhang, A. Blättermann, S. Hagstotz, T. Ding, R. Heck, J. Madronero, F. Martín, and T. Pfeifer, *Nature* **516**, 374 (2014).
- [2] C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, J. Evers, C. H. Keitel, C. H. Greene, and T. Pfeifer, *Science* **340**, 716 (2013).
- [3] A. Blättermann, C. Ott, A. Kaldun, T. Ding, and T. Pfeifer, Two-dimensional spectral interpretation of time-dependent absorption near laser-coupled resonances, *J. Phys. B* **47**,124008 (2014).
- [4] T. Ding, C. Ott, A. Kaldun, A. Blättermann, K. Meyer, V. Stooß, M. Rebholz, P. Birk, M. Hartmann, A. Brown, H. van der Hart, and T. Pfeifer, *Opt. Lett.* **41**, 709 (2016).