Coherent high-harmonic spectroscopy of electron dynamics in molecules

Hans Jakob Wörner

Laboratorium für Physikalische Chemie

ETH Zürich, Switzerland

Electrons determine most properties of matter, yet observing their motion is a major challenge. In the past years, the emerging tools of attosecond science have enabled progress towards this goal. In this presentation we will discuss recent advances in the development of high-harmonic spectroscopy as a method for probing electronic structure and dynamics in molecules. Exploiting the coherence properties of high-harmonic generation in a transient grating geometry enables one to measure both the amplitude and phase of the emitted extreme-ultraviolet radiation in a self-referenced (homodyne) detection. Aligned molecules can be measured relative to a randomly aligned sample, giving access to the complex photorecombination dipole moments. The dynamics of photoexcited molecules can be temporally resolved against the unexcited molecules in the sample, resulting in a high sensitivity. Our recent experiments on the photodissociation of Br_2 show that high-harmonic spectroscopy probes the evolution of the electronic structure of the molecule along the dissociation coordinate. In NO_2, the method reveals the change in electronic symmetry that occurs when the photoexcited wave packet crosses a conical intersection and the subsequent statistical photodissociation dynamics.