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by

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Abstract

Treating the correlated behaviour of multiple particles is challenging for both theory and experiment. This thesis reports on a variety of experimental investigations aiming to advance the understanding of fundamental processes in atoms and molecules: double ionization, isomerization and dissociation. The emphasis lies on ultra-fast processes, where multiple electrons interact or nuclei move so rapidly that coupling between electronic and nuclear dynamics can not be neglected.

Pulses of light from a synchrotron or laser were used to excite or directly ionize molecules or atoms in the gas phase. The momenta (norms or vectors) of the resulting charged fragments were measured in coincidence using different types of time-of-flight spectrometers. Two papers report on the optimization of momentum imaging spectrometers, one for ions and one adaptable electron–ion hybrid.

Direct double ionization was studied in the time domain for the first time, using a pump–probe method with attosecond pulses and electron interferometry (RABBIT). Theoretical development in combination with a coincident measurement of the two electrons revealed a 500 as group delay of the photoelectron pair from xenon, with respect to propagation in a plain Coulomb potential. Electron correlation was also investigated in single ionization, via the angular distribution of a photoelectron. It was shown that the delay of an electron emitted from C_{60} can be altered by 100 as depending on whether the photon energy lies below or above the resonance frequency of a collective electron oscillation (plasmon).

Soft x-rays were used to core-excite molecules to specific orbitals. The subsequent autoionization and dissociation steps were traced by analysing the momentum vectors of multiple ionic fragments. For carbon dioxide, ultra-fast bending initiated by the Renner–Teller effect could bring the two oxygen nuclei together as O_2^+ before dissociation. For few-femtosecond proton migration in water, the kinetic energy release was found to be correlated with the bond angle. Laser-based pump–probe experiments were made on the femtosecond time scale for singly ionized acetylene. Isomerization and dissociation dynamics with time scales of 50–500 fs were observed and analysed.

Key words
direct double ionization, Born–Oppenheimer approximation, correlation, coincident detection, momentum imaging, photoelectron spectroscopy, high harmonics, synchrotron radiation

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