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Correlation-driven charge migration following double ionization and attosecond transient absorption spectroscopy

The dynamics of electrons govern elementary processes such as chemical reactions and charge transport in molecular systems. With the advent of attosecond pulses, it is becoming possible to steer and to probe these dynamics on their natural timescale [1]. In this context, correlation-driven charge migration, i.e., electron dynamics in molecular ions following prompt ionization and which originate from electron correlation and orbital relaxation [2], is of current interest. Theoretical investigations concerning singly ionized molecules have revealed that correlation-driven charge migration is diverse, associated with many facets and it is anticipated to represent a first step of a charge transfer [3]. However, although correlation-driven charge migration has already been predicted in 1999, its experimental demonstration still remains outstanding.

In this talk, an extension of the concept of correlation-driven charge migration to doubly ionized molecules will be presented addressing the charge rearrangement following double ionization induced by light pulses as well as the charge redistribution in the course of electronic decay processes associated with secondary electron emission. Moreover, it will be discussed how correlation-driven electron dynamics are reflected in the time-resolved absorption cross section. Thereby, a promising route for tracing them in real time is identified.