Attosecond time-resolved molecular dynamics

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The intrinsic timescale of dynamical processes that occur at molecular length-scales is ultrafast. It ranges from femtosecond (1 fs = 10^{-15} s) to picosecond (1 ps = 10^{-12} s) timescales when considering structural changes in small molecules, and can involve attosecond (1 as = 10^{-18} s) timescales in the case of electronic processes. Correspondingly, the emergence of attosecond science in the last decade has had a major impact on our understanding of light-induced processes. Attosecond science aims at probing electronic motion on the atomic length-scale, and – more generally - is concerned with the ultrafast motion of charges (including electrons, holes and – in some cases - protons), as well as the interactions between them. Electrons play a crucial role in nature, as they link atoms together in the process of forming molecular bonds and define the optical properties of macroscopic materials.

In the last few years our group has taken first steps towards the implementation of attosecond pump-probe techniques in molecular systems. In a first step, we have investigated dissociative ionization of H_2 under the influence of an isolated attosecond pulse that was followed – at a variable time delay - by a few-cycle infrared laser pulse. Selective localization of the single remaining bound electron on either of the two protons was measured as a function of XUV-IR time delay and revealed the importance of both coupling of the electronic degrees of freedom and electron entanglement on the attosecond to few-femtosecond timescale.

Probing of attosecond dynamics using an attosecond pulse is relevant to attempts to observe charge migration processes on attosecond to few-femtosecond timescales, which have been predicted in the literature. In recent experiments, we have shown that variations in molecular charge densities associated with polarization of the molecules in an electric field can be reflected in photoionization yield measurements. Together with recent results showing that ionic hole wavepackets can be formed by strong-field ionization this suggests a possible way to study charge migration.