

Attosecond pump-probe spectroscopy of molecular electron dynamics

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Attosecond light pulses allow one to probe the inner workings of atoms, molecules and surfaces on the timescale of the electronic motion. In molecules, sudden ionization by an attosecond pulse is followed by charge redistribution on a time scale from a few-femtoseconds down to hundreds attoseconds, which is usually followed by fragmentation of the remaining molecular cation. Such complex dynamics arises from the coherent superposition of electronic states covered by the broadband attosecond pulse and from rearrangements in the electronic structure of the molecular cation due to electron correlation. To investigate these ultrafast processes, attosecond pump-probe and transient absorption spectroscopies have been shown to be very valuable tools [1-8]. In this talk I will present the results of molecular attosecond pump-probe theoretical simulations in which several molecules, from the simplest H₂ one to the aminoacids phenylalanine and tryptophan, are ionized with a single attosecond pulse (or a train of attosecond pulses) and are subsequently probed by one or several infrared or xuv few-cycle pulses. In all cases, the evolution of the electronic and nuclear densities in the photo-excited molecule or remaining molecular ions can be inferred from the measured (or calculated) ionization or fragmentation yields with attosecond time-resolution, and can be visualized by varying the delay between the pump and probe pulses. The results of this work will serve as a guide of future experimental efforts in more complicated molecules and may open the door to the control of charge transfer in biologically relevant processes [9].

References

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