Molecules driven by light: Electron and nuclear dynamics

Peter Saalfrank[a]

[a] *Chemistry Department, University of Potsdam, D-14476 Potsdam, Germany*
peter.saalfrank@uni-potsdam.de

Modern laser technology and ultrafast spectroscopies have pushed the timescales for detecting and manipulating dynamical processes in molecules from the picosecond over femtosecond domains (1 fs = 10^{-15} s), to the so-called attosecond regime (1 as = 10^{-18} s). This way, real-time dynamics of electrons and nuclei after their photoexcitation can be probed and manipulated. On the theory side, powerful quantum dynamical tools have been developed to rationalize experiments on photon-driven molecular species. In the talk, light-induced processes in molecular systems, ranging from "photophysics" and "photochemistry" – will be studied with the help of modern quantum chemistry and molecular quantum dynamics.

In a first, part laser-driven ultrafast electron dynamics will be followed with the help of wavefunction-based ab initio methods such as time-dependent configuration interaction (TD-CI) and the multi-configurational time-dependent Hartree-Fock (MCTDHF) method. Specific examples will be given for state-to-state transitions, creation of electronic wavepackets, and non-linear molecular response (High Harmonic Generation) and its control.

In a second part the vibronic spectroscopy of molecular systems will be considered. For species such as diamondoids the vibronic finestructure in electronic absorption, emission and photoelectron spectra is determined within a time-dependent correlation function approach. This approach has been suggested several decades ago by Heller and coworkers, and is now an efficient method for spectroscopy in complex molecular systems.

Finally, a step to photochemistry is made by considering photoswitching of molecules in an environment. Here, non-adiabatic molecular dynamics with surface hopping allow for detailed insight into the mechanism of photoinduced *trans*-to-*cis* isomerization of azobenzene molecules near surfaces.

References

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