Atomistic simulations of time-resolved transient absorption spectra by real-time Ehrenfest TDDFTB

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Abstract

Since the advent of ultrafast spectroscopy techniques, pump-probe experiments have been extensively used to study the ultrafast dynamics of molecular and nanoscopic systems after a laser pulse. The interpretation of these experiments to obtain valuable information abut the system poses a major challenge, and several theoretical approaches have helped to elucidate such properties. Although there have been atomistic developments in this direction, there has been a clear need of a computational tool to compute the transient spectra of systems of relevant size during relevant periods of time.

In this seminar I will present our implementation of an electron-nuclear real-time propagation scheme for the calculation of transient absorption spectra. When this technique is applied to the study of ultrafast dynamics of Soret-excited zinc(II)-tetraphenylporphyrin in the sub-picosecond time scale, quantum beats in the transient absorption caused by impulsively excited molecular vibrations are observed. The launching mechanism of such vibrations can be regarded as a displacive excitation of the zinc-pyrrole and pyrrole C-C bonds.