IMPRS Seminar 12 April 2023
4:45 pm at MPI-PKS

Multiphoton processes in molecules with time-dependent configuration interaction

Speaker: Jiri Suchan, UCT Prague

Abstract: Time-dependent electronic structure methods are gaining popularity for modeling ultrafast processes in molecules and materials. I will focus on non-linear optical properties and present a new method for calculating two-photon absorption intensities using time-dependent configuration interaction (TD-CI). The approach is based on tracking the system response to the applied electric field and subsequent reconstruction of the transition tensor. Thanks to the extended Floquet formalism, it is possible to numerically follow individual multiphoton channels with attosecond resolution and relate them to the fundamental equations for light absorption. This non-perturbative solution can be easily extended to capture higher-order effects.

As a test system, I use a model chromophore of green fluorescent protein, which two-photon absorption spectrum is profoundly blue-shifted. Analysis of non-Condon effects along several normal modes and various attempts to model complete molecular spectra will be presented.