48th Midwest Theoretical Chemistry Conference
University of Pittsburgh
June 9–11, 2016

Black-Box, Real-Time Simulations of Transient Absorption Spectroscopy

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We introduce an atomistic, all-electron, black-box electronic structure code to simulate transient absorption (TA) spectra and apply it to simulate pyrazole and a GFP-chromophore derivative. The method is an application of OSCF2, our dissipative extension of time-dependent density functional theory. We compare our simulated spectra directly with recent ultrafast spectroscopic experiments. We identify features in the TA spectra to Pauli-blocking, which may be missed without a first-principles model. An important ingredient in this method is the stationary-TDDFT correction scheme recently put forward by Fischer, Govind, and Cramer that allows us to overcome a limitation of adiabatic TDDFT. We demonstrate that OSCF2 is able to reproduce the energies of bleaches and induced absorptions as well as the decay of the transient spectrum with only the molecular structure as input.