Real-time electron dynamics with correlated wavefunction methods

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In recent years, due to progress made in generating and controlling intense laser fields, the timescale of dynamical processes in atomic and molecular systems has been pushed into the attosecond domain (1 as=10^{-18} s). In parallel with experiments, theoretical methods are being developed to treat explicitly time-dependent electronic motion after photoexcitation. This talk describes correlated, explicitly time-dependent, wavefunction based N-electron methods as alternatives to real-time density functional theory, and their application to selected molecular problems.

The focus of the talk is on many-electron methods in which the time-dependent N-electron wavefunction is expanded as a sum of Slater determinants. The first approach of this type to be described is time-dependent configuration interaction (TD-CI), where only the coefficients of the determinants are time-dependent. The second approach is the time-dependent complete active space SCF method (TD-CASSCF), for which both the coefficients and Slater determinants are time-dependent. Extensions of the methods to include ionization, dissipation, and optimal control strategies for excited electron dynamics, are also presented.

The methods will be applied (i) for laser-pulse excitation and switching of real molecules without [1] and with [2] dissipation and / or ionization, (ii) for the calculation of response properties of small molecules [3], (iii) for long-range intermolecular charge transfer [4], and (iv) for controlled electron dynamics in molecules [5].

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