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Quantum dynamics of correlated electrons for time-resolved imaging of laser-driven molecules: Formal and computational challenge

The description of the time-resolved electron dynamics of a many-electron atom or molecule subjected to an intense ultrafast laser pulse requires solving the multi-dimensional, many-body Time-Dependent Schroedinger Equations. We show how the many concepts and tools of ab-initio stationary-state Quantum Chemistry, the object of which is to describe stationary molecular electronic structures, can be adapted to this time-dependent context. While that adaptation is straightforward in so far as the dynamics of bound electrons (described by L^2 wavefunctions) is concerned, using for example our non-variational Time-Dependent Multi-Configuration Self-Consistent Field (TDMCSCF) approach [Nguyen-Dang T.T et al, J. Chem. Phys. 127, 174107 (2007).], it presents severe computational problems when large amplitude electron motions, corresponding to ionizations and laser-induced re-scattering processes, are considered. To deal with this problem, a multi-level partition of the many-electron state space is introduced to allow for separate treatments, with separate accuracy criteria with regards to the electron correlation, of the bound and continuum components of the N electron wavefunction. We show how to solve the coupled integral equations resulting from this type of partitioning in such a manner that norm conservation is well enforced, and how concepts derived from the Graphical Unitary Group Approach (GUGA) can be used to organize the computational tasks. Illustrations of the methodology are given on the H_2 (2 electrons) and LiH (4 electrons) molecules.