Time-Dependent Two-Component Electronic Structure Theory

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We have formulated an ab initio time-dependent two-component spinor method to study the non-collinear spin dynamics of many-electron systems. We employed a direct integration of the time-dependent non-relativistic two-component Hartree-Fock/Kohn-Sham equation using atom-centered basis functions and a unitary propagation approach with a modified midpoint algorithm. An analysis tool based on the Hirshfeld partitioning scheme has been developed to analyze the time-dependent spin magnetization. For the simple one-electron system of the hydrogen atom and collinear multi-electron system of the lithium atom, the real-time two-component Hartree-Fock simulations yield the same results as the analytical Larmor precessions. As an important application of our methodology, we have simulated spin dynamics of a non-collinear Li3 trimer in response to an external magnetic field. The switching of the magnetization at each lattice node was observed during the dynamical simulation. This method has been extended to the Ehrenfest dynamics framework that can be used to model spin-flip processes in molecular dissociation dynamics.