Time-dependent Optimized Coupled-cluster Framework for Laser-driven Multielectron Dynamics

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The role of electron correlation [1] is of paramount importance in the theoretical study of laser-driven multielectron dynamics [2]. The multiconfiguration time-dependent Hartree-Fock (MCTDHF) method [3, 4] and time-dependent complete-active-space self-consistent-field (TD-CASSCF) method [5] serve best for the purpose. However, factorial computational cost prohibits large scale applications of these methods. By limiting the configuration interaction (CI) expansion of the wavefunction, a few other methods have been developed [6, 7] compromising the condition of size-extensivity. Therefore, the choice of the exponential coupled-cluster wavefunction as a replacement of the truncated CI wavefunction is a worthy proposal to restore the lost size-extensivity. Recently, we have derived and implemented a gauge invariant time-dependent optimized coupled-cluster (TD-OCC) method considering double and triple excitation amplitudes (TD-OCCDT) with optimized orthonormal orbitals, where both the orbitals and the amplitudes are time-dependent and propagated in time [8].
Additionally, we have implemented methods based on the further approximate coupled-cluster wavefunction in the TD-OCC framework namely time-dependent optimized coupled-electron pair approximation (TD-OCEPA) method and time-dependent optimized second-order many-body perturbation theory (TD-OMP2) to reach out larger chemical systems (See Fig.1, e.g.).

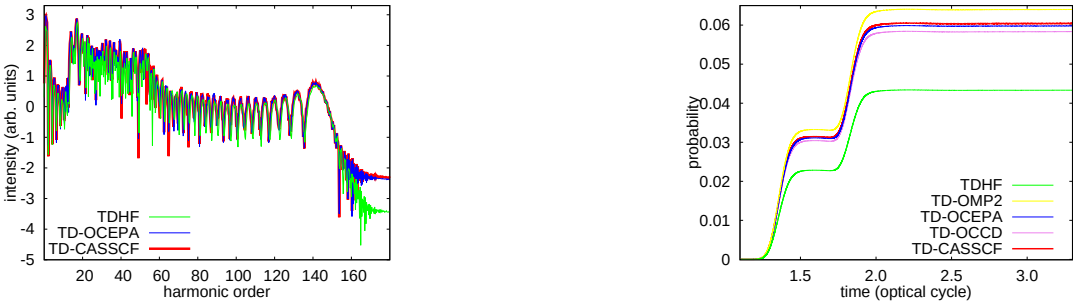

Fig. 1 High-order harmonic generation (HHG) spectra (left) and the time evolution of single ionization probablity (right) of Kr exposed to a laser pulse with a wavelength of 1500 nm and intensity of 1.8×10^{14} W/cm^2. Comparison of the results of TD-CASSCF, TD-OCCD, TD-OCEPA, TD-OMP2 and TDHF methods. All the correlated methods predict similar HHG spectra. Eight valence electrons are correlated among 13 active orbitals for the correlated methods.

References

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