

Time-dependent restricted-active-space self-consistent field theory for laser-driven electron dynamics

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Inspired by a recent work on orbital adaptive time-dependent coupled-cluster theory [1], we have developed a new theoretical framework named time-dependent restricted-active-space self-consistent field (TD-RASSCF) theory for time-dependent many-electron dynamics. Optimising the orbitals at each time step with incorporating the RAS ansatz, i.e., decomposing the orbital space into several subspaces, among which electron transitions are conditionally allowed, one can expand the wave function accurately using relatively small numbers of orbitals and configurations. This computationally economical approach will make it possible to analyse laser-induced many-electron dynamics of large atoms and molecules beyond the capability of the multiconfigurational time-dependent Hartree-Fock (MCTDHF) method [2].

To demonstrate the computational efficiency of the TD-RASSCF theory, we investigated the one-dimensional (1D) beryllium atom. Figure 1 shows the laser induced dipole moment obtained from three different methods: the time-dependent Hartree-Fock (TDHF), the MCTDHF, and the MCTDHF-doubles (-D) methods, where the latter is a special case of the TD-RASSCF theory. The MCTDHF and -D results are in good agreement despite of large reduction of the number of configurations in the MCTDHF-D calculation.

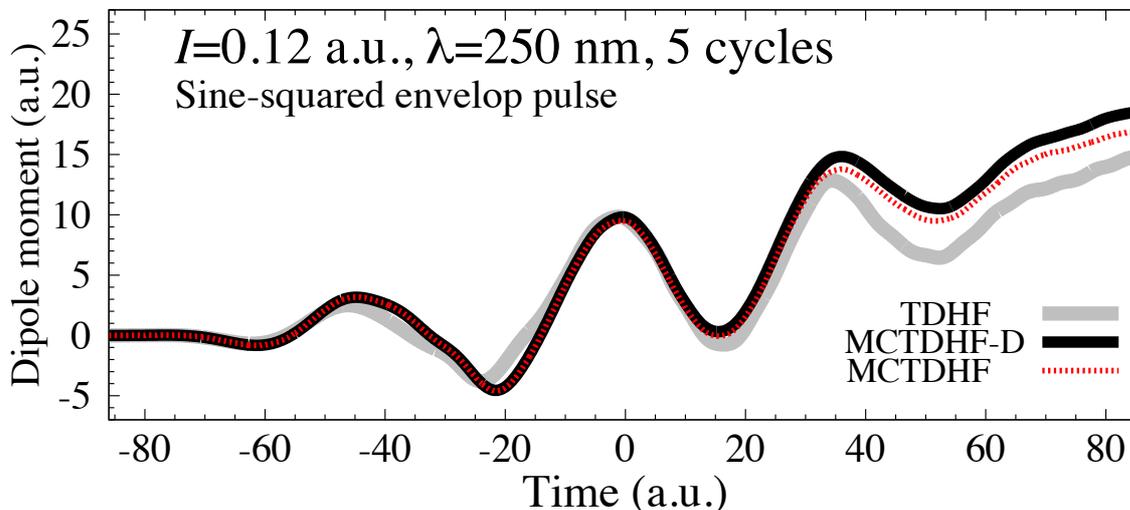


FIG 1. Dipole moment of the 1D beryllium atom induced by an intense laser pulse. Solid gray line: TDHF, solid black line: MCTDHF-D, and dotted (red) line: MCTDHF. In the MCTDHF and -D calculations, 12 spatial orbitals were employed, and consequently the wave functions were expanded by 4356 and 491 configuration state functions, respectively. The figure includes a specification of laser parameters, from which the value of the Keldysh-parameter is estimated to be $\gamma=1.2$.

References:

- [1] S. Kvaal, J. Chem. Phys. **136**, 194109 (2012).
- [2] J. Caillaud *et al.*, Phys. Rev. A **71**, 012712 (2005).