Implementation of the time-dependent Hartree-Fock method for diatomic molecules on prolate spheroidal coordinates

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Multielectron dynamics plays an important role when atoms and molecules are subject to ultrashort intense laser pulses. Numerical investigations of multielectron dynamics and correlation effects require highly nonperturbative treatments, which are very challenging. In order to tackle this challenge, the time-dependent multiconfiguration self-consistent-field (TD-MCSCF) methods have been developed[1,2]. As one example of TD-MCSCF methods, we have developed and numerically implemented the time-dependent complete-active-space self-consistent-field (TD-CASSCF) method for atoms[3,4], with which stable and highly accurate simulations of strong-field phenomena are achieved within reasonable computational cost.

Due to extra degrees of freedom, the response of molecules to strong fields is considerably more complicated than that of atoms. Diatomic molecules, as one kind of simplest molecules, are of fundamental importance in this sense. In this contribution, as a starting point of TD-MCSCF methods, we report 3-dimensional real-space implementation of the time-dependent Hartree-Fock (TDHF) method for diatomic molecules. Our implementation adopts prolate spheroidal coordinates with spatial grids discretized by a finite-element discrete variable representation (FEDVR), which offers accurate representation of electron-nucleus interaction and high sparsity of the Hamiltonian operators. As an example, we apply this method to the simulation of high harmonic generation (HHG) from CO. The ground state energy obtained from our program is -112.791 a.u., which matches well with the value -112.789 a.u. obtained by the PSI4 program with Aug-cc-pVQZ basis set. The resulting high-harmonic spectrum is shown in Fig. 1.

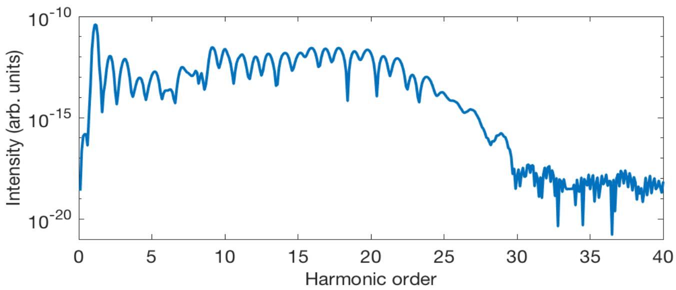

Figure 1: HHG spectrum of CO irradiated by a laser pulse with a wavelength of $\lambda = 800\,nm$, a peak intensity of $I = 6 \times 10^{13}$ W/cm^2 and the foot-to-foot pulse duration of $\tau = 6T$ with T being the optical period of the laser field.

References

[1] T. Sato and K. L. Ishikawa, Phys. Rev. A **88**, 023402 (2013).

[2] Y. Li, T. Sato, and K. L. Ishikawa, Phys. Rev. A **99**, 043401 (2019).

[3] T. Sato, K. L. Ishikawa, I. Brezinová, F. Lackner, S. Nagele, and J. Burgdörfer, Phys. Rev. A **94**, 023405 (2016).

[4] Y. Orimo, T. Sato, A. Scrinzi, and K. L. Ishikawa, Phys. Rev. A **97**, 023423 (2018).