

Implementation of the time-dependent multiconfiguration method for diatomic molecules

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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, we report two real-space implementations of the TD-CASSCF method for diatomic molecules. The first one focuses on the multielectron dynamics within the Born-Oppenheimer approximation. This implementation adopts prolate spheroidal coordinates with spatial grids discretized by a finite-element discrete variable representation (FEDVR), which offers an accurate representation of electron-nucleus interaction and high sparsity of the Hamiltonian operators. In the second implementation, we adopt the fully general TD-MCSCF method^[5,6], in which protons and electrons are treated on an equal footing, to address the joint electron-nuclear dynamics in a strong laser field. Our results show that the present method is well suited to describe not only the laser-induced ionization but also laser-induced dissociation and vibrational excitation.

References

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