Ionization, dissociation, and Coulomb explosion of H_2 in intense laser fields: A theoretical study based on moving-grid approach in phase space

Zhongyuan Zhou[1] **and Shih-I Chu**[2]

Department of Chemistry, University of Kansas, Lawrence, KS, 66045, USA

Synopsis: A moving-grid approach based on coupled coherent states and classical trajectories is developed for the study of dynamics of polyatomic molecules in intense laser fields. In this approach, the initial state and trajectories are determined by solving the imaginary time-dependent Schrödinger equation using diffusion Monte Carlo method. The wave function is then calculated by propagating the initial state on the moving grids guided by the classical trajectories. This approach has been applied to study ionization, dissociation, and Coulomb explosion of H_2 in intense laser fields as a demonstration.

Ab initio calculation based on fully quantum mechanical approach is still a challenge for the study of dynamics of polyatomic molecules in intense laser fields due basically to the huge numbers of configurations involved. This difficulty may be conquered by using a moving-grid approach based on couple coherent states in phase space. This approach has been applied to the study of double ionization of He in strong laser field [1]. In this paper we extend the moving-grid approach to the polyatomic molecules interacting with intense laser fields. As a demonstration, we apply the approach to explore ionization, dissociation, and Coulomb explosion of H_2 in intense laser fields.

The moving-grid approach characterizes a quantum system in phase space with coherent-state representation (CSR) wave functions [2] that depend on classical trajectories represented by coordinates and momenta. The basic equation of the moving-grid approach is an integro-differential equation which is derived from the time-dependent Schrödinger equation by expanding the wave function in coherent states and deliberating the evolution of trajectories (moving grids). The trajectories are determined by Hamiltonian canonical equation with quantum corrections.

The CSR wave functions are calculated by solving the basic integro-differential equation together with the Hamiltonian canonical equation for the moving grids. The initial CSR wave function and initial values of trajectories are estimated and optimized simultaneously by the solution of imaginary time-dependent Schrödinger equation with diffusion Monte Carlo method [3,4]. The spin symmetry of the initial CSR wave function is taken into account by using symmetrized and antisymmetrized coherent states [4].

To investigate the full dynamics of H_2 in intense laser fields, we consider both electron and nucleus motions. The electron motion is described by the CSR wave functions in the moving-grid approach while the nucleus motion is depicted by the classical dynamics. In CSR, the singularities of Coulombic potentials of electrons and nuclei are removed completely [1,4]. The CSR wave functions of electrons are computed under the consideration of the nuclear motion. The probabilities of ionization, dissociation, and Coulomb explosion of H_2 are then calculated for different laser intensities and frequencies with different numbers of trajectories. The results change with the number of trajectories. The scaling rules of Monte Carlo method are used to estimate the final results. Our results are in well qualitative agreement with available experimental and other theoretical results.

References

[1] D.V. Shalashilin, M.S. Child, and A. Kirrander, *Chem. Phys.* **347,** 257 (2008).
[2] D.V. Shalashilin and M.S. Child, *Chem. Phys.* **304,** 103 (2004).
[3] I. Kosztin, B. Faber, and K. Schulten, *Am. J. Phys.* **64,** 633 (1996).
[4] D.V. Shalashilin and M.S. Child, *J. Chem. Phys.* **122,** 224108 (2005); **122,** 224109 (2005).

[1] E-mail: zvzhou@ku.edu
[2] E-mail: sichu@ku.edu