Implementation of the first-principles simulation based on time-dependent multiconfiguration self-consistent field methods for polyatomic molecules under intense laser pulses

Yuki Orimo[1], **Takeshi Sato**[1,2,3], **Kenichi L. Ishikawa**[1,2,3]

1. Department of Nuclear Engineering and Management, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
2. Photon Science Center, Graduate School of Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
3. Research Institute for Photon Science and Laser Technology, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

Abstract: We present an implementation of time-dependent multiconfiguration self-consistent field (TD-MCSCF) methods based on the adaptive finite element method for first-principles simulations of general molecules under intense laser pulses.

Electronic dynamics induced by the interaction of strong laser fields and ultrafast light pulses with matter has attracted great research interest in the past decade. In our previous works, to theoretically investigate multielectron dynamics under laser pulses we have proposed first-principles approaches based on the time-dependent multiconfiguration self-consistent field (TD-MCSCF) method [1], which are referred to as the time-dependent complete-active-space self-consistent field (TD-CASSCF) method and the time-dependent occupation-restricted multiple-active-space (TD-ORMAS) method [2-5]. These methods enable accurate simulations of multielectron dynamics considering electronic correlation with quite reduced computational costs compared to the full-CI approach. However, their computational costs are still expensive to simulate large systems such as polyatomic molecules.

To achieve larger scale first-principles simulations, we have developed an implementation of the TD-MCSCF methods based on the adaptive finite element method [6], which provides flexible discretization and successfully treats polyatomic structures in numerical simulations. Figure 1 shows a part of the 3D mesh used for a hydrogen molecule. In the red-colored area around nuclear positions, where wave functions can drastically vary due to the singularities of the Coulomb potential, the fine mesh discretization is applied. On the other hand, in the blue-colored area distant from nuclei, the coarse mesh is used. This adaptivity and flexibility enable the use of a large simulation box with less computational costs while keeping the accuracy of the discretization. Since the adaptive finite element method may generate a quite fine mesh, which leads to numerical instability of time-propagation, we have introduced a stable propagator based on the iterative Lanczos method. Furthermore, the present implementation is highly parallelized for distributed memory computers and able to simulate multielectron dynamics much faster than our previous implementation.

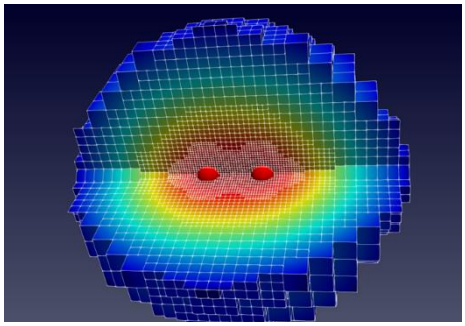

Fig. 1 A part of the 3D mesh used in the adaptive finite element method. This mesh is generated for a hydrogen molecule. The two red spheres indicate nuclear positions.

In this talk, I will give the detail of our implementation including spatial discretization, stable time-propagation and parallelization, and some examples of simulations such as high harmonic generation from molecules.

References

[1] K. L. Ishikawa and T. Sato, IEEE J. Sel. Topics Quantum Electron. 21 8700916 (2015)
[2] T. Sato and K. L. Ishikawa, Phys. Rev. A 88, 023402 (2013).
[3] T. Sato and K. L. Ishikawa, Phys. Rev. A 91, 023417 (2015).
[4] R. Sawada, T. Sato and K. L. Ishikawa, Phys. Rev. A 93, 023434 (2016).
[5] T. Sato, K. L. Ishikawa, *et al.*, Phys. Rev. A 94, 023405 (2016).
[6] D. Arndt, *et al.*, J. Numer. Math., 28, 131-146 (2020)