XXIX International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC2015) IOP Publishing
Journal of Physics: Conference Series **635** (2015) 112084 doi:10.1088/1742-6596/635/11/112084

The time-dependent two-particle reduced density matrix method: LiH in strong laser fields

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Synopsis We present a time-dependent many-body theory that is based on the propagation of the two-particle reduced density matrix (2-RDM) [1]. We apply our method to simulate the non-linear response of one-dimensional lithium hydride in femtosecond mid-IR laser pulses.

With the availability of laser pulse intensities competing with the internal Coulomb force the electron dynamics in atoms and molecules can be perturbed far beyond the linear regime. Such strong perturbations give rise to fascinating non-linear effects such as above threshold ionization, high harmonics generation and multiple ionization. Along with these advanced experimental techniques the development of sophisticated numerical simulations is required to describe the correlated many-body systems.

We have developed a new approach, the time-dependent two-particle reduced density matrix (TD-2RDM) method, to describe the time-dependent many-body response. As a prototypical molecular many-body system we apply it to the one-dimensional model for lithium hydride (LiH) in strong infrared laser fields. We solve the equations of motion for 2-RDM by approximating the three-particle reduced density matrix (3-RDM) in terms of lower order RDMs via a new reconstruction functional that preserves norm, energy, and spin symmetries during time propagation. We show that approximately enforcing N-representability during time evolution is essential for achieving stable solutions. For the induced dipole moment (Fig. 1) our results are in very good agreement with numerically exact solutions obtained from the multiconfigurational time-dependent Hartree-Fock method. Further we find that the time-dependent 2-RDM method (TD-2RDM) performs consistently better than time-dependent Hartree-Fock (TDHF) and time-dependent density functional theory (TDDFT) within the adiabatic local-density approximation over the entire time interval. This advance opens up the intriguing possibility of solving the many-body problem within the two-particle subspace previously only studied for the ground state of complex atoms and molecules [2, 3].

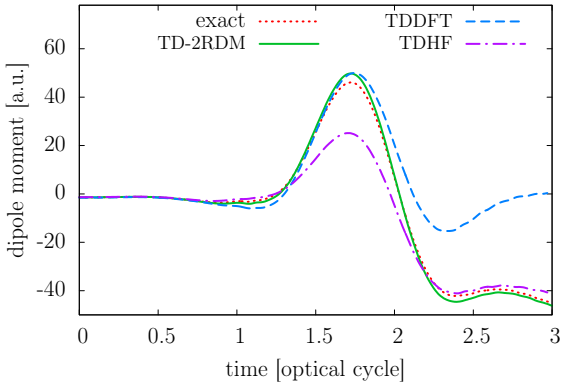

Figure 1. Dipole moment of LiH subject to a laser pulse with $I = 8 \times 10^{14}\mathrm{W/cm^2}$ within the TD-2RDM method compared with the exact (MCTDHF) reference, the TDHF and the TDDFT calculations.

Acknowledgement

This work has been supported by the FWF doctoral school Solids4Fun, FWF SFB-041 ViCoM, and FWF SFB-049 Next Lite, and in part by KAKENHI, the Photon Frontier Network Program of MEXT (Japan), and Center of Innovation Program from JST (Japan).

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