TDDFT calculation for electron-ion dynamics in molecules under intense laser pulse

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Recent development of laser technology has enabled us to study various phenomena that occur under high-intensity (10^{13}~10^{15}[W/cm^2]) and ultrashort (10^{-15}~10^{-18}[sec]) laser pulse. The pulse duration of such laser pulse is comparable with the period of the electronic excitations in molecules and is much shorter than the period of molecular vibrations. The strength of the electric field of the laser pulse is comparable to the Coulomb field from ions inside molecules. Therefore, the electron motion in the ultrashort laser pulse is characterized by nonlinear and nonperturbative dynamics. To describe them, the time-dependent density-functional theory (TDDFT) is expected to be useful.

We have been developing a three-dimensional description of electron-ion dynamics in molecules employing the TDDFT. In our computational procedure, the time-dependent Kohn-Sham equation for electrons is solved in real-time and real-space introducing uniform grid points in the three-dimensional Cartesian coordinate.[1)] The forces acting on ions are evaluated every time step, and the Newton equation for ions is solved simultaneously. [2)]

As our first analysis for coupled electron-ion dynamics, we calculate the Coulomb explosion of molecules under intense laser pulse. The Coulomb explosion proceeds following the formation of highly charged molecule by intense laser pulse. By detecting the kinetic energy of the ions after the Coulomb explosion, one may investigate the ion distance when the Coulomb explosion occurred. Although the Coulomb explosion has been studied extensively, unified picture for the process is still under development. To obtain intuitive picture, the three-dimensional first-principles simulation for the Coulomb explosion will be useful. Therefore, we here apply the TDDFT taking the Coulomb explosion of N_2 molecule as an example.

Our typical calculation is shown in Fig. 1 and 2. In this example, we use 800[nm], 30[fs] laser with maximum intensity of 4.1×10^{15} [W/cm^2]. The polarization direction of the laser is set parallel to the molecular axis. Fig. 1 shows that the bond of N_2 molecule is broken around 20~25fs. In Fig. 2, we show the force acting on ions as a function of time. This figure clearly shows that the force acting on ion is much weaker than the force of two point particles with the same charge, when the two ions are not yet separated. After about 25 fs, the electron is localized around each ions, as seen in Fig. 1, and consequently the force is practically equal to Coulomb force as is seen in Fig. 2.

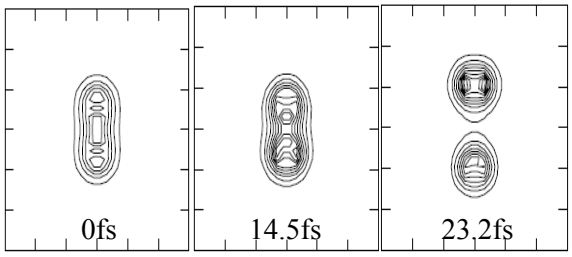

Fig. 1: Snapshot of the electron density of N_2 molecule in the laser pulse.

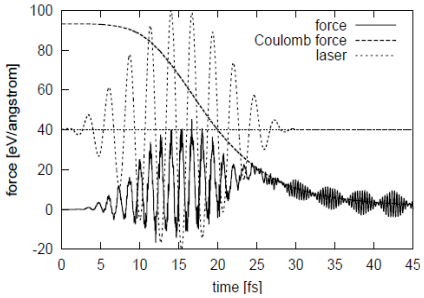

Fig. 2: The force acting on ion is shown by solid curve. Coulomb force between point-like N^{+2} ions as a function of the distance is shown by dashed curve.

1) K. Yabana and G. F. Bertsch, Phys. Rev. B54, 4484(1996)
2) O. Sugino and Y. Miyamoto, Phys. Rev. B59, 2579(1999)