Time-dependent complete-active-space self-consistent field method for multielectron dynamics in intense laser fields

Takeshi Sato\textsuperscript{1} and Kenichi L. Ishikawa\textsuperscript{1}

\textsuperscript{1}Photon Science Center, School of Engineering, University of Tokyo, Tokyo 113-8656, Japan
sato@atto.t.u-tokyo.ac.jp

We propose a flexible time-dependent many-electron theoretical method for multielectron dynamics in intense laser fields, based on the concept of the complete-active-space self-consistent field (CASSCF) \cite{Roos1980}. The method, called TD-CASSCF, allows compact yet accurate representation of ionization dynamics in many-electron systems, thus largely extends the applicability of the rigorous MCTDHF (multiconfigurational time-dependent Hartree-Fock) method \cite{Caillat2005, Kato2004}, while keeping the accuracy.

It is reasonable to expect that in a large molecule interacting with high-intensity, long-wavelength laser, the deeply bound electrons remain non-ionized, while only the higher-lying valence electrons ionize appreciably. For the bound electrons, a closed shell description of the Hartree-Fock type would be acceptable. However, fully correlated treatment is required for ionizing electrons to describe the seamless transition from the closed-shell-dominant ground-state to the symmetry-breaking continuum. The CASSCF provides an ideal ansatz for such problem. It introduces the concept of core and active orbital subspaces, and configuration-interaction expansion is limited to the determinants with core orbitals being doubly occupied. It is possible to further split the core space to frozen-core (timely fixed) and dynamical-core (allowed to vary in time, in response to the field) subspaces. See Fig. 1.

Figure 2 shows the ionization probabilities of one-dimensional LiH-LiH model, induced by a 3-cycle laser pulse of \(\sin^2\) envelope with wavelength 750 nm and peak intensity \(4\times10^{14}\) W/cm\(^2\). As seen in the figure, TD-CASSCF with 4 active electrons closely reproduces the results of MCTDHF with the same number of orbitals. Details of the theory and numerical applications will be presented in this contribution to ECAMP11.

References:
\cite{Roos1980, Caillat2005, Kato2004}.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{Figure1.png}
\caption{TD-CASSCF concept, illustrating a 12 electron system with 4/4/4 electrons in frozen-core/dynamical-core/active orbital subspaces, respectively.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{Figure2.png}
\caption{Ionization probabilities (\(P_n\) for \(n\)-electron ionization) of 1D LiH-LiH as a function of time. Results of TDHF (left), TD-CASSCF with 4 active electron (right), and MCTDHF (black solid lines). Total number of orbitals in both TD-CASSCF and MCTDHF is 8.}
\end{figure}