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A Parallel MCTDHF Code for Few-Electron Systems with Time-Dependent External Fields

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THE MCTDHF METHOD

In this paper we introduce an extension of the time-dependent Hartree-Fock to the Multi-Configuration Time-Dependent Hartree-Fock (MCTDHF) method, which in the limit of infinitely many configurations converges to the exact few-electron time-dependent Schrödinger equation. Denoting the spatial and spin coordinates collectively by $q_i = (\vec{r}_i, s_i)$, the MCTDHF ansatz for the wave function Ψ of f electrons reads

$$\Psi(q_1, \ldots, q_f; t) = \sum_{j_1=1}^{n} \cdots \sum_{j_f=1}^{n} c_{j_1 \cdots j_f}(t) \phi_{j_1}(q_1; t) \cdots \phi_{j_f}(q_f; t)$$

where the coefficients $c_{j_1,\ldots,j_f}$ are taken to be totally antisymmetric with respect to their indices, which leaves only $\begin{pmatrix} n \\ f \end{pmatrix}$ independent c's. Increasing the number of orbitals n allows to systematically include correlation effects. The evolution equations for $c_{j_1 \cdots j_f}(t)$ and $\phi_{j_i}(q_i; t)$ involve the non-linear and non-local mean-field operators and are given in Ref. [1].

For the implementation of the method on a parallel computer the spatial domain of $\phi_{j_1}(q_1; t)$ is distributed over computing nodes. Communication is minimized by a finite-element discretization [2] and by low-rank approximations for the non-local operators. The block diagram of parallelization is given in Fig. 1. We found near linear scaling with the number of nodes. Interaction potentials of arbitrary, numerically defined shape allow application to atoms and molecules as well as quantum dots and nano-structures. Convergence of electron spectra for 1-d model systems is shown in Fig. 2.

CORRELATION AND SPATIAL DIMENSION

Using MCTDHF we solve the time-dependent Schrödinger equation with a strong external laser field in 3 spatial dimensions with 3-d molecular model potentials and Coulomb repulsion between electrons. Laser and molecule parameters are chosen such that a large fraction of the molecules become ionized. Analogous 1-d model systems with the same ionization potentials and properly screened interactions were constructed.

We find pronounced differences between the 3-d system and the 1-d model (Fig. 3): (1) The importance of correlation is greatly overestimated in 1-d and (2) ionization increases with molecule size in 3-d, while the opposite is observed in 1-d.

CONCLUSIONS

The newly developed MCTDHF method can be employed for solving the time-dependent Schrödinger equation for several interacting electrons with general 3-d potentials. We demonstrate favorable scaling in parallel computation and convergence of important observables such as electron spectra. Calculations in 3-d are mandatory, as lower-dimensional models were shown to generate severe artefacts.

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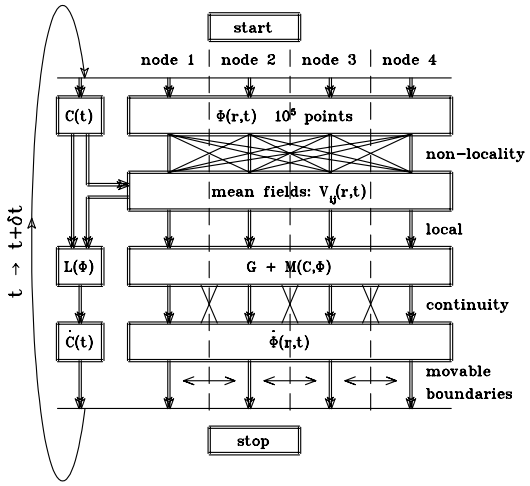

Fig. 1. Flow chart of the parallelization of MCTDHF. Orbitals are distributed over compute nodes. The calculation of mean-field operators requires all-to-all communication. Application of mean field operators is strictly local. Differential operators require minimal nearest-neighbor communication.

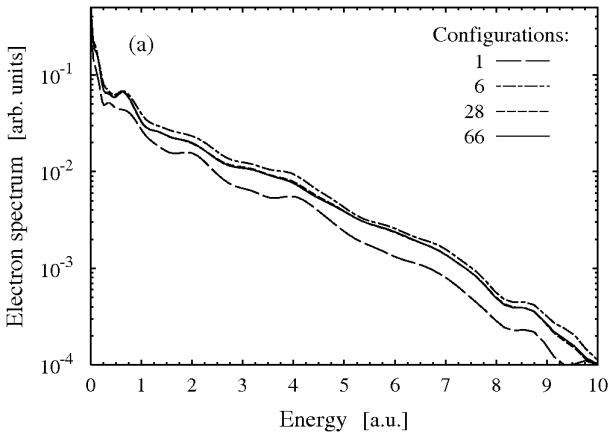

Fig. 2. Convergence of MCTDHF: the photoelectron spectrum from model He atom generated by an 800 nm laser pulse. Calculations with 28 and 66 configurations nearly coincide.

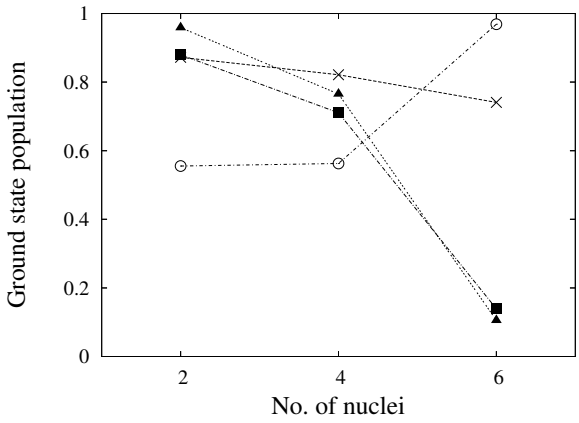

Fig. 3. Ground state depletion for 3-d and 1-d model molecules as a function of molecule size. In 3-d, depletion increases with molecule size for internuclear separations $R = 1.4$ (crosses) and $R = 3$ (full squares), and the Hartree-Fock result is similar to the correlated result (triangles, $R = 3$). In contrast, 1-d models show an decrease of depletion with size (circles).