Massively-parallel simulation of reduced-scaling ground and excited state coupled-cluster methods

Marjory C. Clement[a*] and Edward F. Valeev[b]

[a]Department of Chemistry, Virginia Tech
Blacksburg, VA United States

[b]Department of Chemistry, Virginia Tech
Blacksburg, VA United States

Abstract: To greatly increase the reach of conventional many-body techniques like coupled-cluster (CC) we must employ numerical approximations that reduce the prohibitive high-order computational complexity as well as deploy our reduced-scaling CC algorithms on the largest machines available. With these complementary ideas in mind we implemented a simulation framework for reduced-scaling CC methods based on the massively-parallel implementation of ground- and excited-state CC methods in the re-engineered Massively Parallel Quantum Chemistry (MPQC) package [1]. Although the current implementation retains the high complexity of the conventional CC methods, this allows rapid exploration of reduced-scaling ansatz and provides a production-quality solver component for reduced-scaling massively parallel coupled-cluster currently under development.

Both the ground and excited state approaches can utilize arbitrary compressions of the cluster amplitudes, including those based on pair-natural orbitals (PNOs) [2,3]. The idea behind PNOs is quite simple: for each pair of electrons in a system, only a few orbital products contribute substantially to the pair-correlation wave function and pair energy, with the number of PNOs per pair essentially independent of the system size. Furthermore, the PNOs determined using an approximate first-order guess provide a useful subspace for solving the coupled-cluster equations. Using the PNOs as the template, we devise similar compressions to the right- and left-hand vectors in EOM-CCSD. The robust convergence of the simulated reduced-scaling CCSD(T) and EOM-CCSD energies will be demonstrated with respect to the single rank-truncation parameter for systems of unprecedented size.

References

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