Multiconfigurational Self-Consistent Field Theory with Density Matrix Embedding: The Localized Active Space Self-Consistent Field Method

Scientific Achievement
Development of a new method for computing electronic wave functions which combines the power and flexibility of multiconfigurational methods with the speed and low cost of density matrix embedding theory.

Significance and Impact
Computational simulations are challenging because molecules and materials often have multireference electronic structure, so that computational costs of accurate calculations explode exponentially with size. This is addressed by breaking large multiconfigurational calculations into small coupled subsystem calculations using a modified density matrix embedding theory (DMET) algorithm. This new approach has a formal linear cost scaling with respect to system size, opening the door to accurate wave function calculations on much larger chemical models than previously possible.

Research Details
- DMET method decomposes large system into small, unentangled subsystems
- LAS wave function is designed so that DMET algorithm can be applied, but small-system results can be recombined to iteratively improve a high-quality whole-system electronic wave function
- Existing quantum chemistry codes can be used to solve subsystem problems
- Numerical tests show that LASSCF reproduces results of standard, costly CASSCF method with DMET’s low cost


Fragmentation of the electronic wave function into coupled, unentangled subsystem wave functions

Nanoporous Materials Genome Center
nmgc.umn.edu

Work was performed at University of Minnesota.