Efficient Implementation of a Large-Scale v2RDM-Driven CASSCF Method

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Abstract: Complete active space self-consistent field (CASSCF) methods are successful in tackling static correlation problems in computational chemistry, but the conventional configuration interaction CASSCF approach limits the size of the active space. An alternative strategy is to use the two-electron reduced density matrix (2RDM) as the variational variable instead of the wavefunction. Our previous work demonstrates that a variational 2RDM (v2RDM)-driven CASSCF approach allows for much larger active spaces (e.g., 50 electrons in 50 orbitals with the simultaneous optimization of 1892 orbitals).[1] In this work, we describe recent improvements to our v2RDM-driven CASSCF implementation which includes efficient tensor manipulations through the use of the libtensor library,[2] new strategies for improved convergence, and incorporation of effective core potentials. These improvements increase the number of potential applications, which include our interest in actinide chemistry.

References

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