Impact of high-rank excitations on the accuracy of the unitary coupled cluster downfolding formalism

Nicholas P. Bauman, Bo Peng, and Karol Kowalski

Pacific Northwest National Laboratory, Richland, WA 99354

Since its introduction, the Hermitian form of the downfolding procedure utilizing the double unitary coupled cluster ansatz (DUCC) has been a dynamic tool for advancing quantum chemistry on quantum computers. We have demonstrated that the Hermitian effective Hamiltonians generated through this technique capture a significant portion of a large dimensional calculation in a substantially smaller active space, even with judicious approximations. A comprehensive understanding of the various approximations involved is crucial for the optimal application of the DUCC downfolding procedure.

In a previous paper, we explored the impact of different terms in the commutator expansion on energetics. In this study, we utilized external excitations estimated from standard single-reference coupled cluster methods (SR-CC) to showcase the necessity of higher-rank SR-CC external amplitudes to accurately describe the energies in strongly correlated regimes. Employing occupation-number-representation codes, we constructed the matrix representation of second quantized operators, enabling the exact representation of exponentials of various operators and removing further ambiguity. To systematically investigate correlation effects, we utilized simple model systems, specifically H6 and H8 linear chains, allowing for a continuous variation of correlation effects from weakly to strongly correlated regimes.

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