A hybrid quantum/classical computing approach for perturbative energy corrections in unitary coupled cluster theory

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Augmenting low-rank coupled cluster approximations that perturbatively account for higher-rank excitations is a tractable way of accounting for electron correlation. This is perhaps best exemplified by the "gold standard" CCSD(T) method in quantum chemistry, which bolsters the baseline CCSD method with effects of triple excitations via perturbation theory (PT) considerations. Despite this established success, such a synergy between PT and the unitary analog of CC theory (UCC) has not been explored. This is, at least in some part, due to the intractability of performing infinite-order UCC calculations on a classical computer, which arises from a non-terminating Hamiltonian-cluster commutator expansion.

Here, we propose a hybrid computing approach wherein converged amplitudes of low-rank UCCSD and UCCD approximations - constructed on a quantum simulator – are leveraged by a classical computer to construct energy corrections associated with higher-rank cluster operators. Rationale behind the choice in energy correction is shown to be rigorous by studying the properties of finite-order UCC energy functionals. We show that such a scheme is a tractable way of including electron correlation that has been missed as a result of truncating the cluster operator in UCC. We then assess this hierarchy of methods' performance on di and tri-atomic molecules, and further demonstrate the potential impact these methods have for UCC calculations on near and long-term quantum hardware.