Towards Practical Quantum Chemistry on Quantum Computers using Localized Active Spaces

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Accurate electronic structure methods for modeling strong electron correlation scale exponentially with system size on classical computers. Richard Feynman's famous quote on quantum simulation of an inherently quantum phenomenon, however, teaches us to use quantum computers for the study of such quantum mechanical problems and provides promise to get rid of the exponential scaling. Yet, the challenges presented by near-term Noisy Intermediate-Scale Quantum (NISQ) devices make this a formidable task. Over the last few years, researchers have employed innovative techniques to harness the power of quantum devices effectively. One promising technique is quantum embedding. Our group has developed a quantum embedding method, localized active space unitary coupled cluster (LAS-UCC) [J. Chem. Theory Comput. 2022, 18, 12, 7205–7217], which introduces a fragment-based multireference wave function into a quantum computer and employs the UCCSD ansatz to perform a Variational Quantum EigenSolver (VQE) calculation to restore inter-fragment correlation using hybrid quantum-classical architecture. However, a case study like the ground state prediction of a Kremer's dimer model demands 774 parameters (cluster amplitudes)—each parameter corresponding to 10-100 gates and over 2000 UCCSD/VQE iterations for convergence, which is currently impractical.

In my presentation, I will discuss a collaboration between the University of Chicago and the Argonne Chemical Lab, where we have developed localized active space unitary selected coupled cluster (LAS-USCC), a multireference amplitude selection scheme that reduces the required parameters in the LASUCC ansatz by at least 90% (to 77 parameters) and reduces UCC/VQE iterations to approximately 170. This scheme enables the ground state prediction within 1 cm^{-1} accuracy, a significant stride towards practical quantum computing applications. Further, I will show other representative examples that demonstrates the method's capability for NISQ devices.