

Classical Optimization Strategies for ADAPT-VQE for Chemical Applications

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Introduction



- Quantum simulation of electronic structure is a promising application for demonstrating **quantum advantage** of quantum computers, but many challenges must be overcome to achieve this goal.
- Hybrid variational approaches like VQE are an attractive approach to compute electronic energies using noisy near-term quantum computers. Circuit depth?
- ADAPT-VQE provides a means for determining problem specific compact quantum circuits, thus reducing the required circuit depth to obtain accurate electronic energies.

In this work, we evaluate the possibility of performing ADAPT-VQE calculations for molecules with quality basis sets entirely on classical computers with an eye toward further refinement with quantum hardware.

The **Variational Quantum Eigensolver (VQE)** is a hybrid quantum algorithm for obtaining ground state electronic energies based on the variational principle.

UCCSD Ansatz

$$|\Psi_{\text{UCCSD}}\rangle = \prod_{I=1}^{N_{\text{op}}} \hat{U}_I(\theta_I) |\Psi_0\rangle$$

$$\hat{U}_i^a = \exp[\theta_i^a (\hat{a}_i^a - \hat{a}_a^i)]$$

$$\hat{U}_{ij}^{ab} = \exp[\theta_{ij}^{ab} (\hat{a}_{ij}^{ab} - \hat{a}_{ab}^{ij})]$$

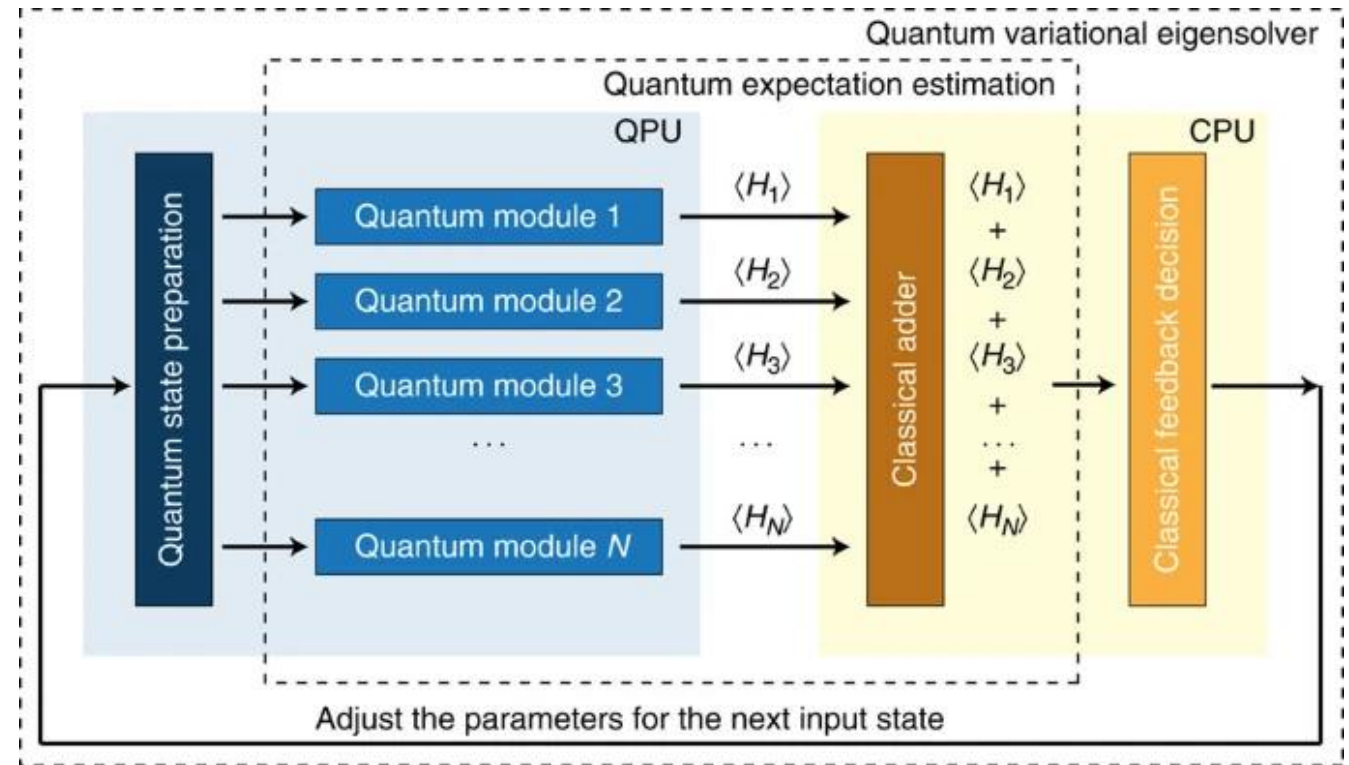


Image from Peruzzo, A.; McClean, J.; Shadbolt, P.; Yun, M.-H.; Zhou, X.-Q.; Love, P. J.; Aspuru-Guzik, A.; O'Brien, J. L. *Nat. Commun.* **2014**, *5*, 4213.

How efficiently can we perform VQE on classical computers?

Sparse Wavefunction Circuit Simulator



The individual UCCSD operators (\hat{U}_I) can be efficiently evaluated on classical computers [*J. Chem. Theory Comput.* **2021**, *17*, 841-847].

We incorporated this result into our **sparse wavefunction circuit simulator** (arXiv:2301.05726) to perform (approximate) VQE calculations using the UCCSD ansatz entirely on classical hardware.

The key idea is to truncate the wavefunction to N_{CUT} determinants after applying each operator (\hat{U}_I) based on the resulting amplitudes without significantly affecting the accuracy.

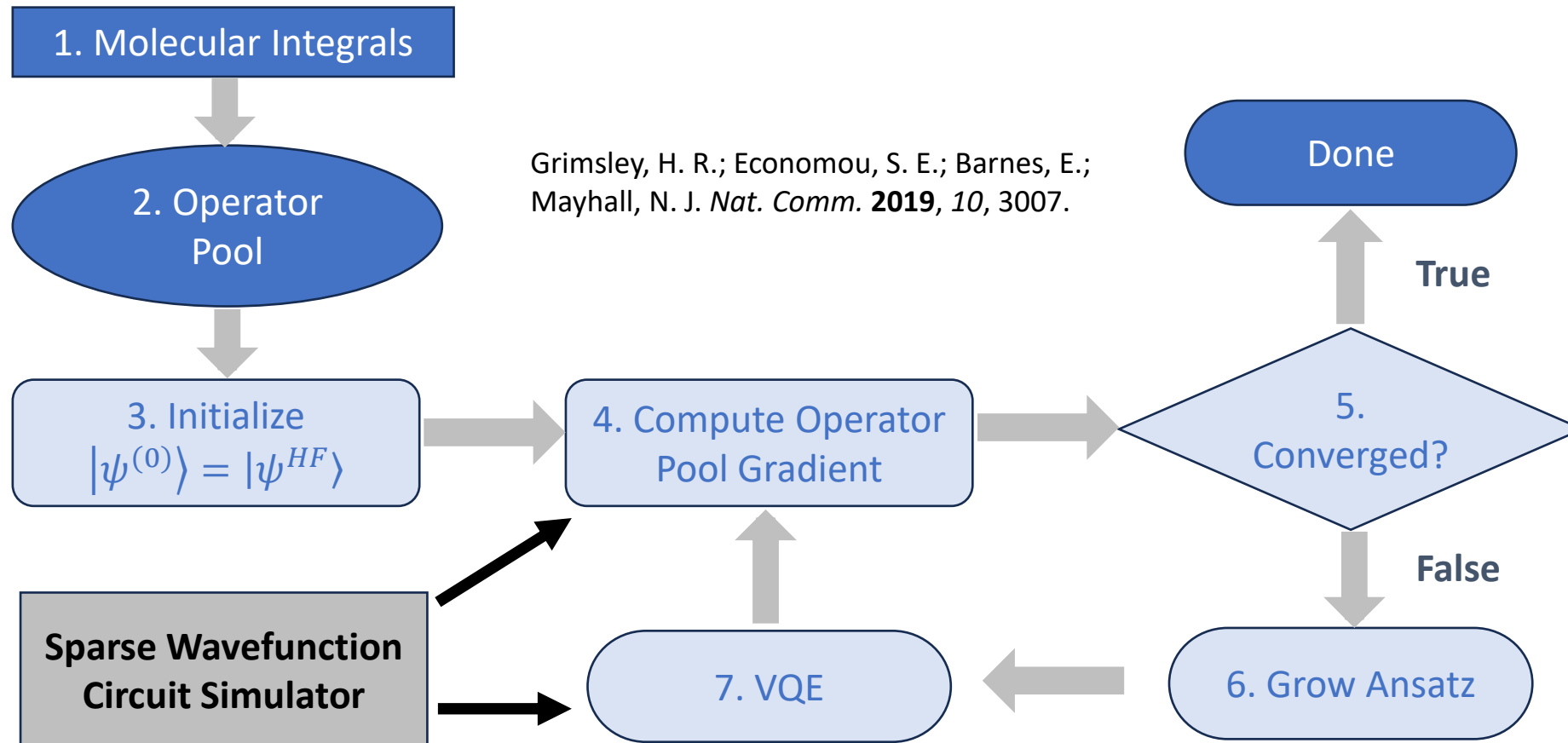
$$|\Psi_{\text{UCCSD}}\rangle = \hat{U}_{N_{\text{op}}}(\theta_{N_{\text{op}}}) \cdots \hat{U}_3(\theta_3) \hat{U}_2(\theta_2) \hat{U}_1(\theta_1) |\Psi_0\rangle$$
A red horizontal line is drawn below the operators in the equation. From this line, five vertical red arrows point upwards to the bottom of each operator: $\hat{U}_{N_{\text{op}}}$, \hat{U}_3 , \hat{U}_2 , and \hat{U}_1 . The arrow under the ellipsis \cdots does not have a vertical line, indicating that the truncation occurs after each of the four operators shown.

Keep only N_{CUT} wavefunction amplitudes with largest magnitude

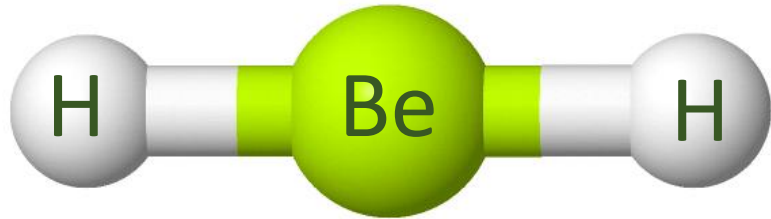
ADAPT-VQE



The objective of ADAPT-VQE is to generate a problem-specific ansatz that is more compact compared to the fixed sized UCCSD ansatz.



Computational Details



Equilibrium: $R_{\text{Be-H}} = 1.3264 \text{ \AA}$

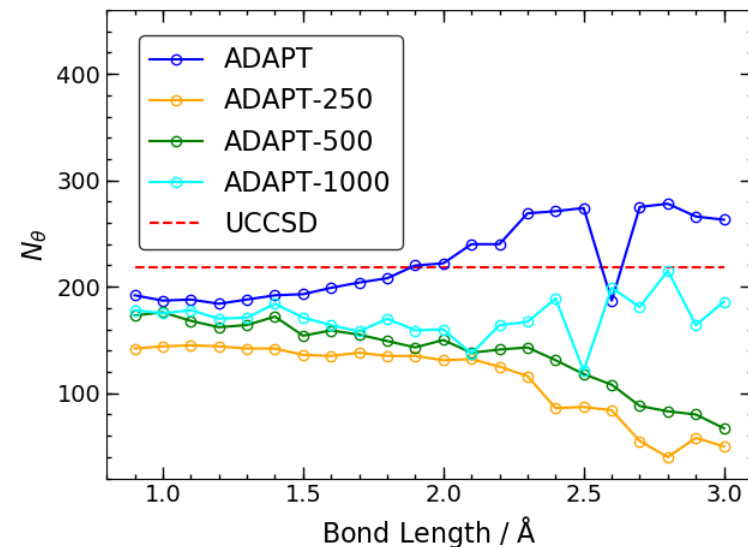
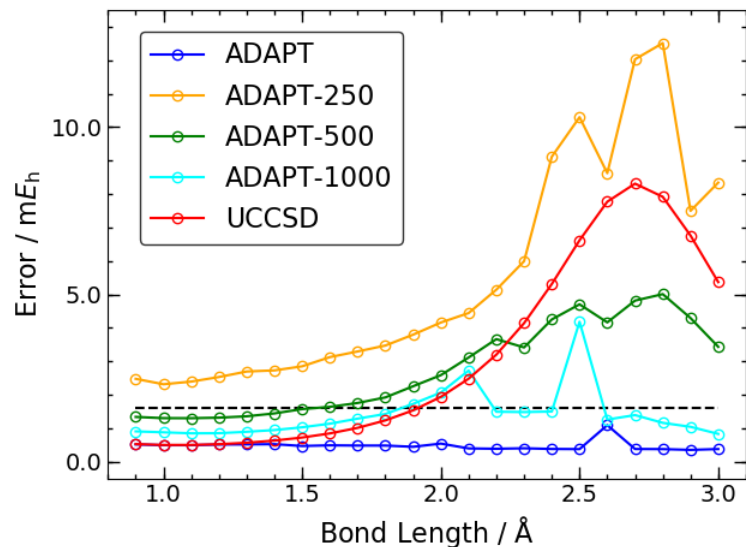
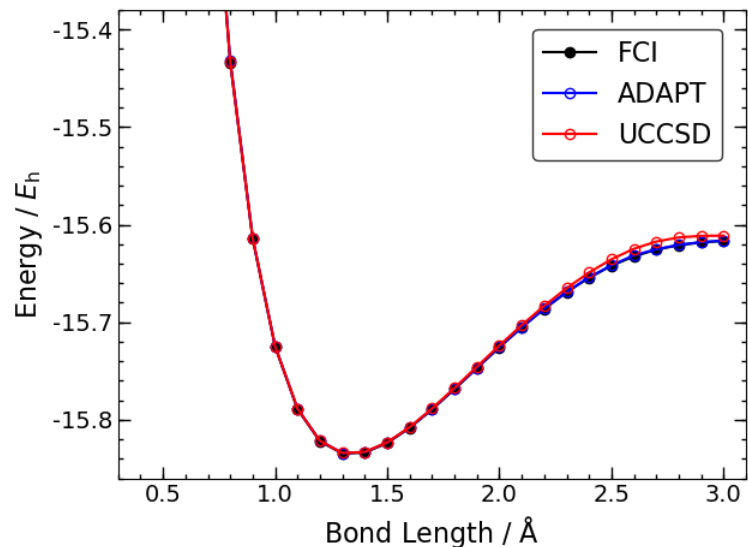
- cc-pVDZ basis set with frozen-core approximation
- PySCF for molecular integrals
- BFGS algorithm from SciPy
- ADAPT-VQE convergence: 10^{-2}
- VQE convergence: 10^{-7}
- Generalized single-double spin-complement operator pool

1. Study convergence behavior and efficiency of ADAPT-VQE.
2. Performance of ADAPT-VQE with sparse wavefunction circuit solver.

Results: ADAPT-VQE

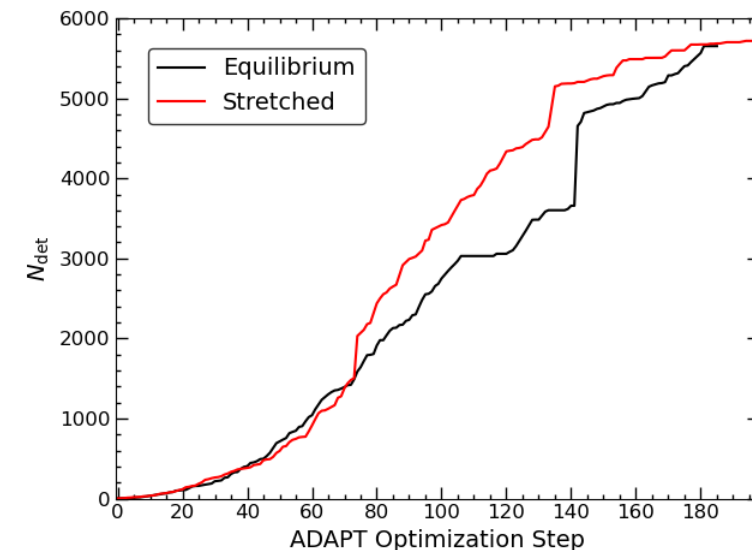
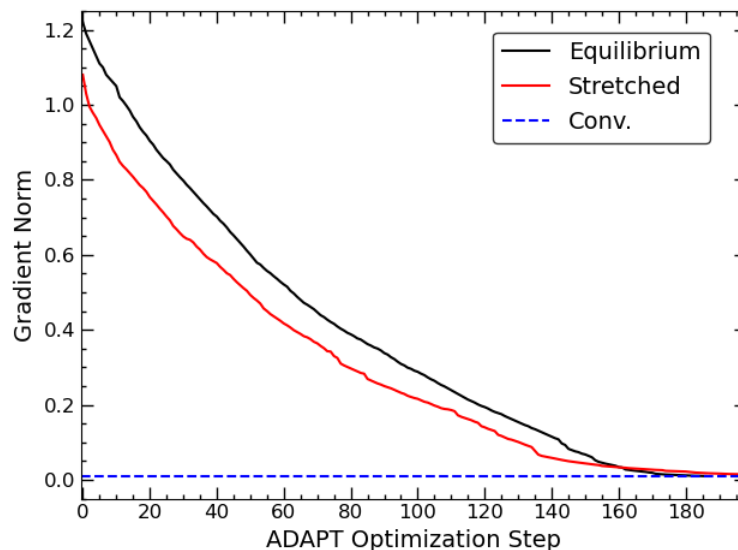
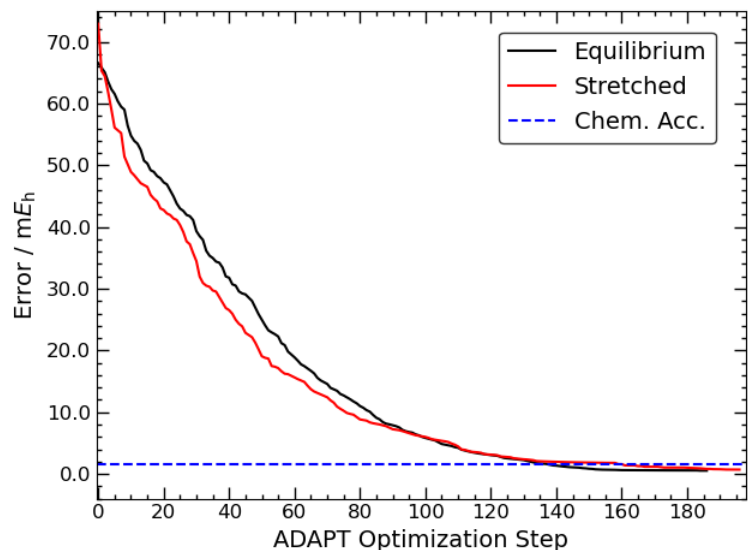


Symmetric dissociation of BeH_2 using the cc-pVDZ basis set and the frozen-core approximation.



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Results: Convergence



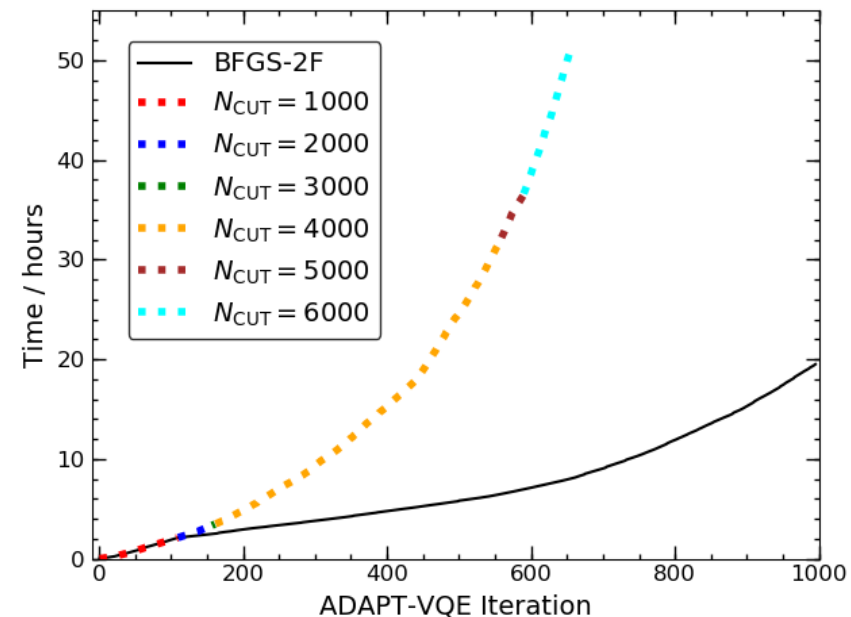
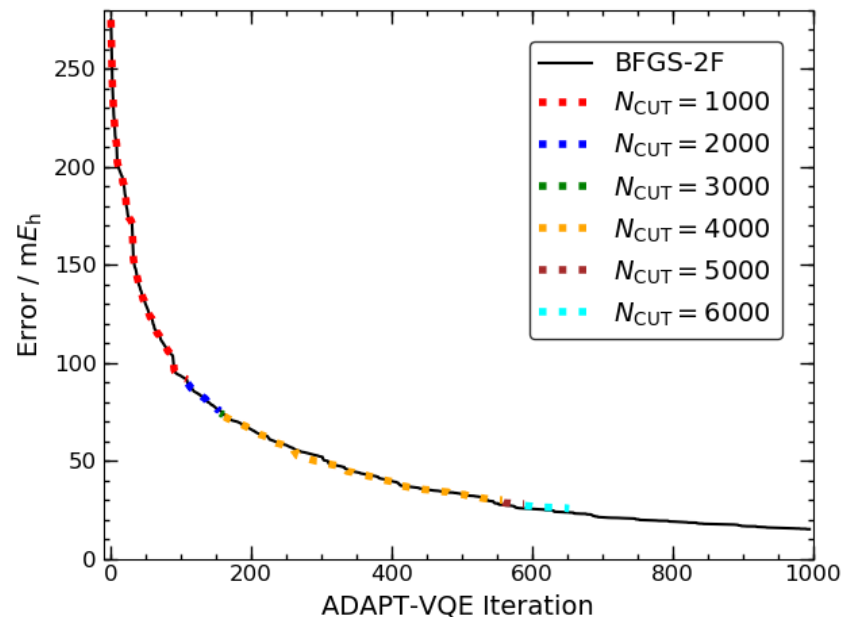
- Smooth convergence in terms of energy and the ADAPT-gradient but slows as approach to chemical accuracy.
- Even though we reach chemical accuracy near step 140, the ADAPT calculation continues for nearly 50 more steps.
- The number of determinants with non-zero ($>10^{-16}$) amplitudes increases with each ADAPT step.

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Results: C_2 and BFGS-2F



For large problems, full optimization becomes challenging, so we developed a method, **BFGS-2F**, extending ADAPT-VQE using the N_{CUT} parameter of our SWCS. For C_2 at the equilibrium geometry, we arrived at an error of **12.7 mE_h** vs an error of **26.9 mE_h** for the UCCSD ansatz.



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Conclusions



- With the SWCS, we can extend the scope of electronic structure problems studied with VQE and ADAPT-VQE.
- We demonstrated the performance SWCS for BeH_2 (46 qubits) and C_2 (52 qubits).
- Extended to variants like TETRIS-ADAPT-VQE and CEO-ADAPT-VQE.
- SWCS can incorporate downfolded Hamiltonians.
- Benchmark results expected on future quantum computers.
- Pre-optimization strategy for refinement on quantum hardware.

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