

Quantum Benchmarking Of Molecular Ground-State Energy Estimation

Khuram Shahzad¹, Rosa Di Felice²⁻³, Guido Goldoni¹⁻²

¹ Department of Physics, Computer Science and Mathematics, University of Modena and Reggio Emilia, 41125 Modena, Italy

² CNR-NANO-Istituto for Nanosciences, 41125 Modena, Italy

³ Department of Physics and Astronomy, University of Southern California, Los Angeles, California, 90089, USA

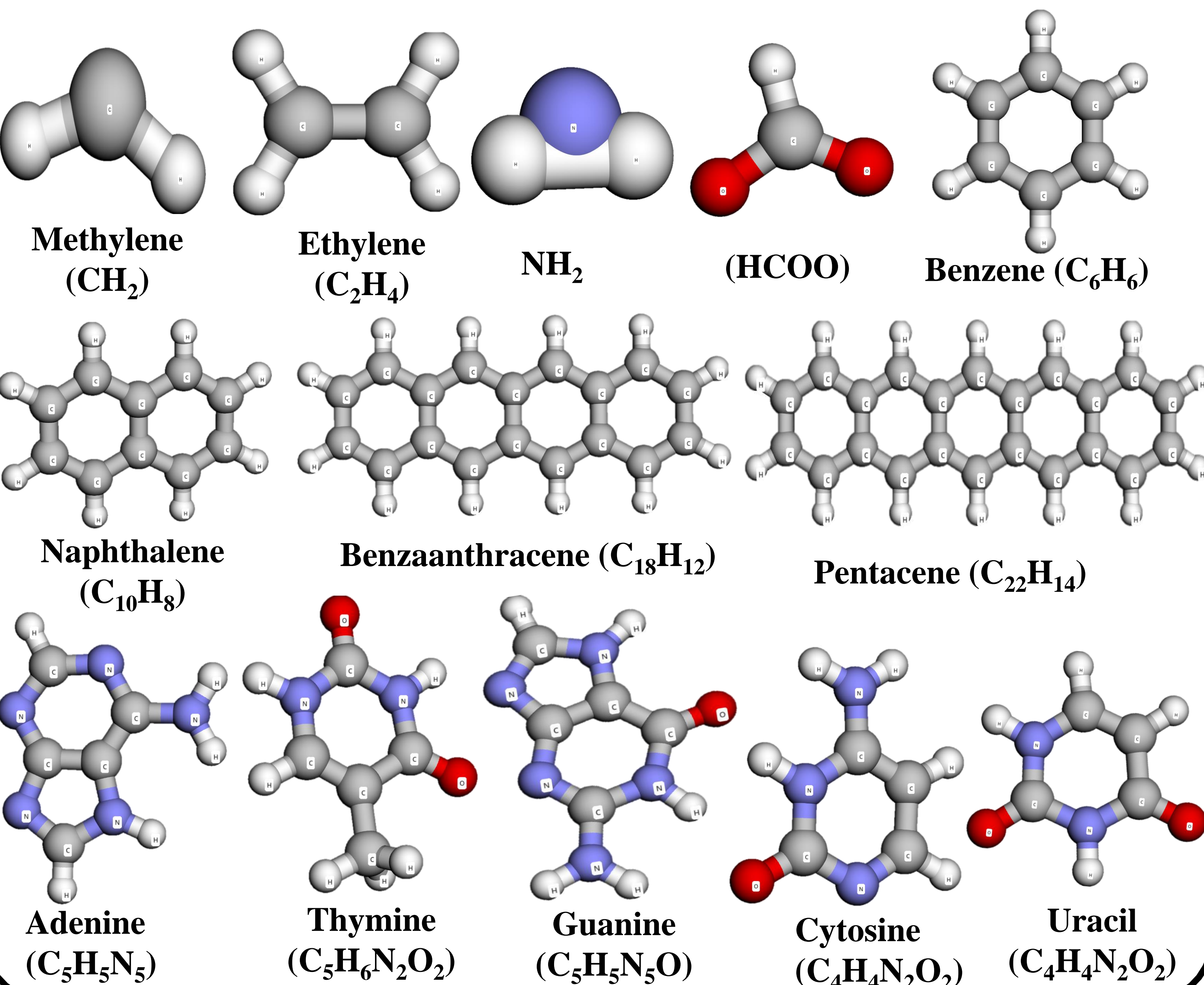
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Overview

- Ground-state energy estimation (GSEE) and excited-state energy estimation (ESEE) are at the core of predicting and controlling chemical reactions, with huge impact in various technologies.
- Classical algorithms for GSEE scale exponentially with system size and are elusive to even the most powerful supercomputers for useful system sizes.
- Quantum computation (QC) emerges as a powerful alternative with a potentially linear scaling. Results of QC algorithms must be benchmarked on reliable data.
- The Variational Quantum Eigen solver (VQE) is a hybrid quantum-classical algorithm for approximating molecular ground-state energies on near-term quantum devices.
- We demonstrate VQE for **DNA Basis** and **Organic Molecules (NH₂, COOH, Benzene (C₆H₆), Naphthalene (C₁₀H₈), Benz[a]anthracene(C₁₈H₁₂), Pentacene (C₂₂H₁₄)** using quantum software and running on CPU and GPU devices. **While we cannot draw conclusions on energy accuracy, we prove the GPU acceleration of VQE.**

Classical algorithms: Molecular Structure

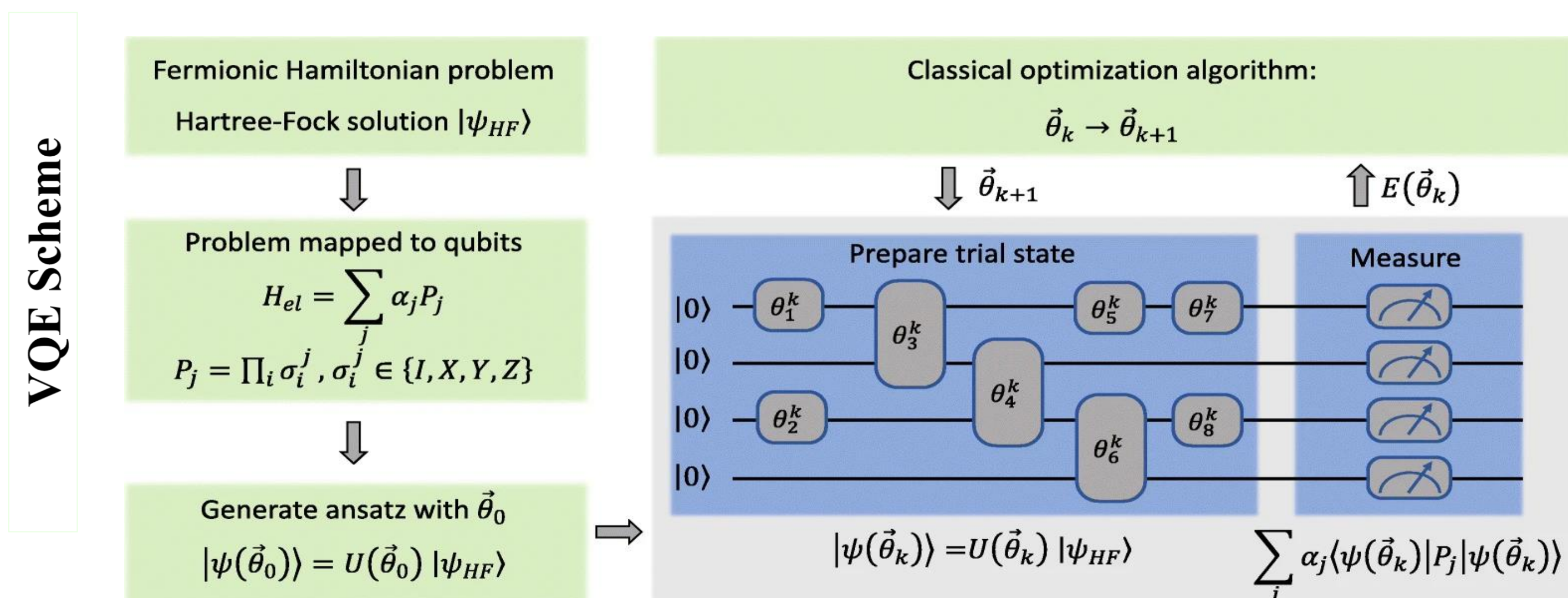
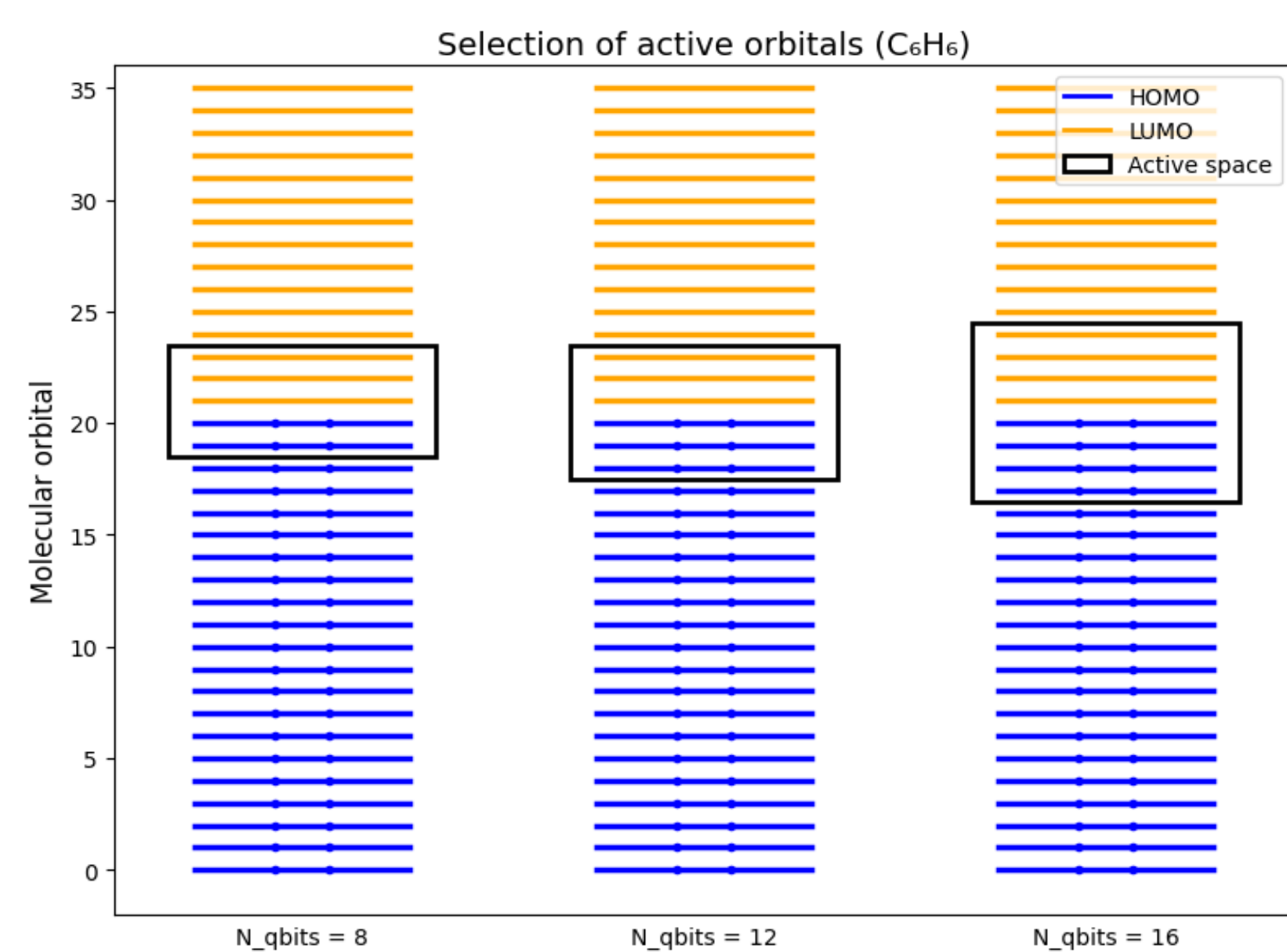
- Geometry obtained from the Computational Chemistry Comparison and Benchmark Database (CCCBDB) [3].
- Experimentally determined and pre-optimized at the CCSD(T)/cc-pVDZ level of theory.



Methodology

- We use the CUDA Quantum (CUDA-Q) framework, enabling simulations on both CPU and GPU backends.
- Within the 6-31G and cc-pVDZ basis set for the molecular Hamiltonian, we downsize the active space by freezing “core” orbitals and restricting the simulation to chemically relevant “reactive” orbitals, thus balancing computational cost with chemical accuracy.

- The molecular Hamiltonian is constructed via PySCF and OpenFermion libraries, and the UCCSD ansatz is implemented as the variational circuit within CUDA-Q.
- We systematically evaluate energy convergence, qubit and parameter counts, and computational runtimes across various active space configurations.



Overview of the Variational Quantum Eigensolver (VQE) workflow. The electronic structure problem is first mapped from fermionic operators to qubit-based Pauli operators. An initial trial wavefunction is prepared using a parameterized quantum circuit (ansatz) based on the Hartree-Fock reference. Expectation values of the Hamiltonian are measured on a quantum device, and the energy is evaluated classically. A classical optimizer updates the parameters iteratively to minimize the energy, forming a hybrid quantum-classical optimization loop.

Molecular Hamiltonian

$$\hat{H} = \sum_{i=1}^N \left(-\frac{1}{2} \nabla_i^2 - \sum_A \frac{Z_A}{r_{iA}} \right) + \sum_{i < j} \frac{1}{r_{ij}}$$

1. Electronic Hamiltonian

$$\Psi_{HF} = \frac{1}{\sqrt{N!}} \det[\psi_1(\mathbf{x}_1) \psi_2(\mathbf{x}_2) \cdots \psi_N(\mathbf{x}_N)]$$

2. Hartree-Fock Approximation

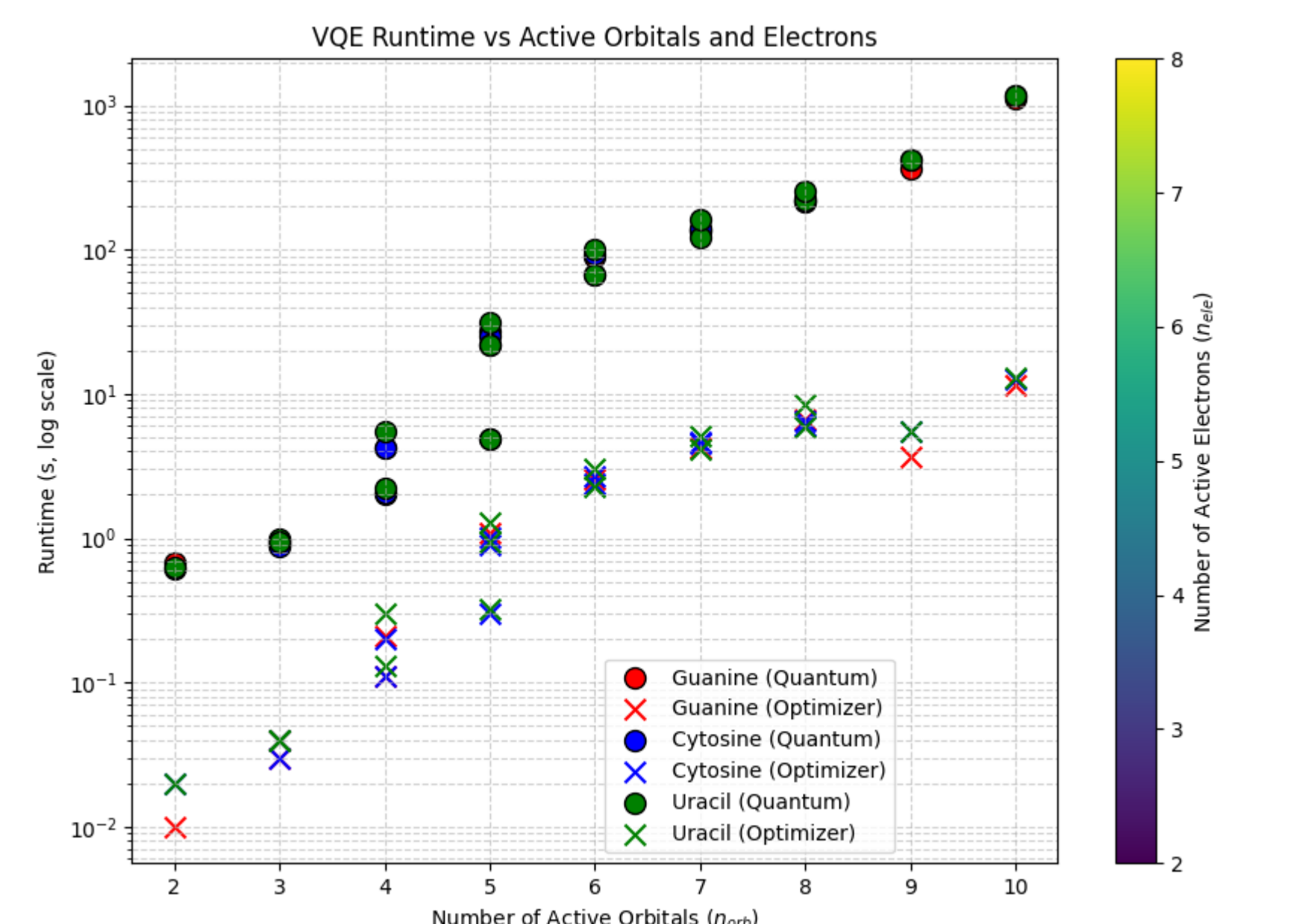
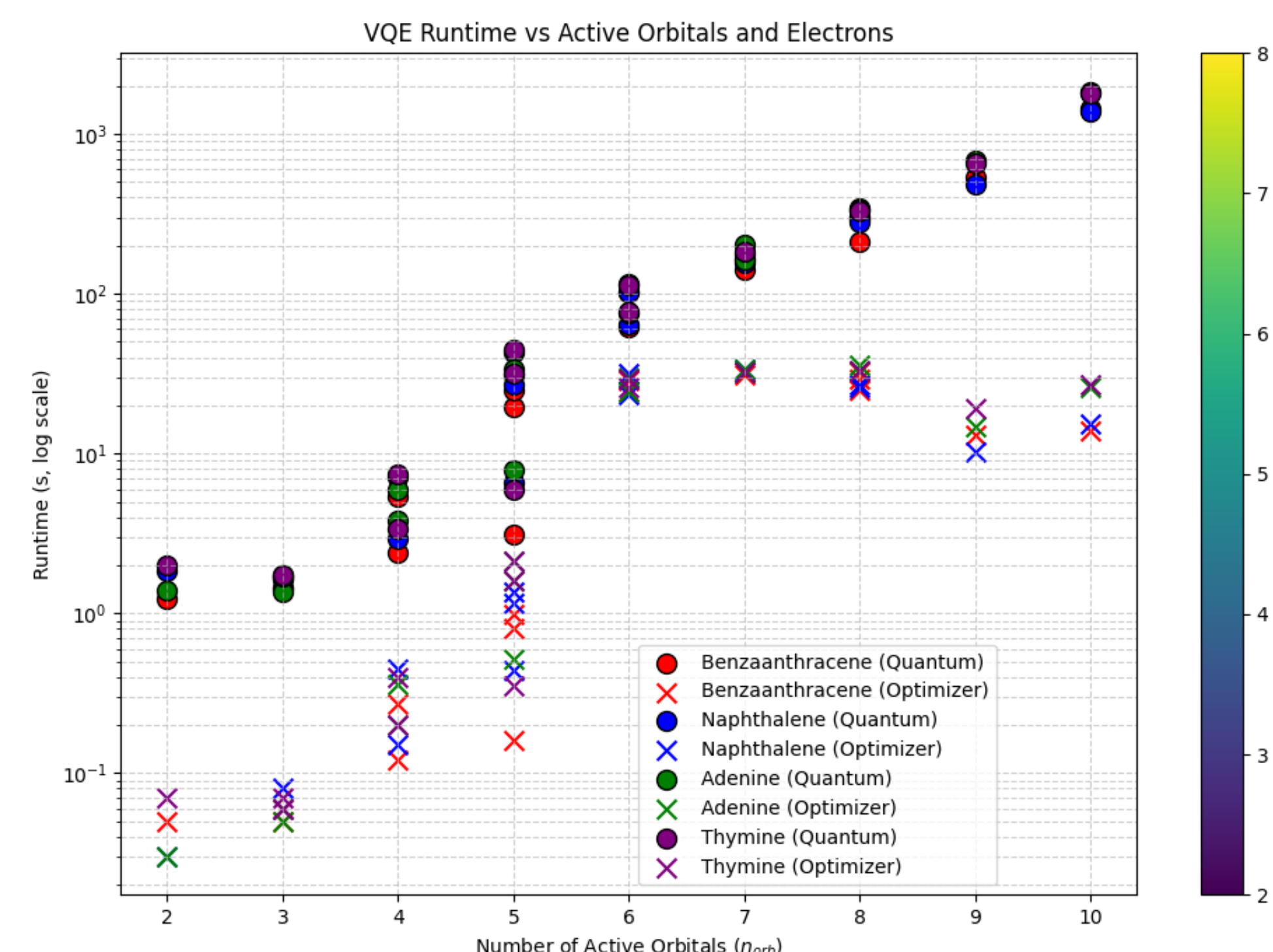
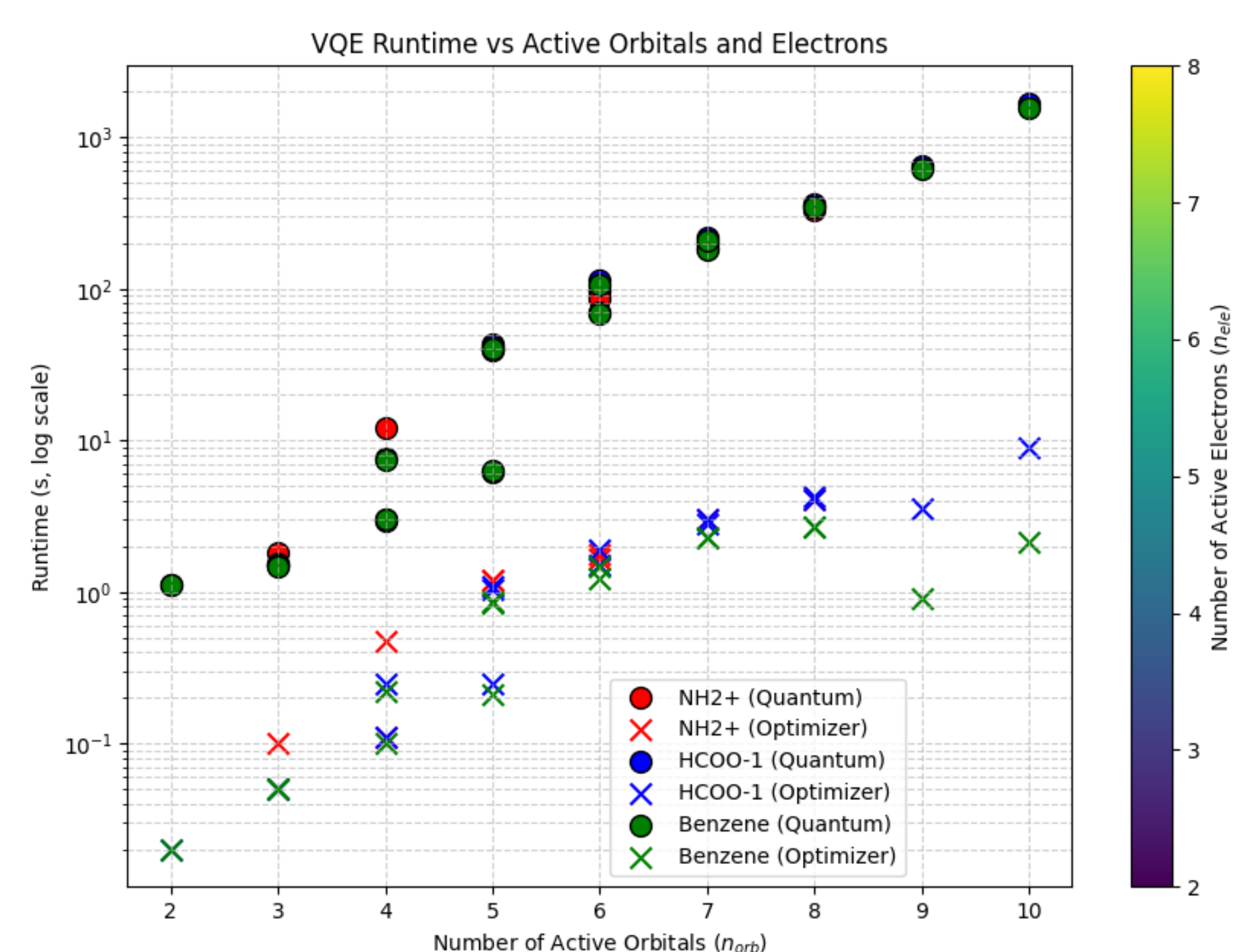
$$\hat{H} = \sum_{pq} h_{pq} a_p^\dagger a_q + \frac{1}{2} \sum_{pqrs} h_{pqrs} a_p^\dagger a_q^\dagger a_r a_s$$

3. Second Quantization Hamiltonian

$$H^{AS} = \sum_{pq}^{\text{active}} \tilde{h}_{pq} \hat{a}_p^\dagger \hat{a}_q + \frac{1}{2} \sum_{pqrs}^{\text{active}} g_{pqrs} \hat{a}_p^\dagger \hat{a}_q^\dagger \hat{a}_r \hat{a}_s + E_{\text{core}}$$

5. Active Space Hamiltonian

Results



Summary

- Our results show a significant acceleration in runtime for GPU based simulations compared to CPU implementations, while maintaining comparable energy accuracy within mHa differences.
- The gain increases monotonically with the size of the active space.
- The approach is generalizable to other molecular species, including open-shell molecules.

- ✓ The large active space shows over 260× speedup, highlighting GPU superiority for large-scale quantum simulations.
- ✓ For the smallest case, GPU is still about 2.3× faster than CPU.

References

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