

Quantum Benchmarking Of Molecular Ground-State Energy Estimation

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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.
- Hence, 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 methylene (**CH₂**) and Benzene (**C₆H₆**) 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.

Electronic Configuration

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

1. Electronic Hamiltonian

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

3. Second Quantization Hamiltonian

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

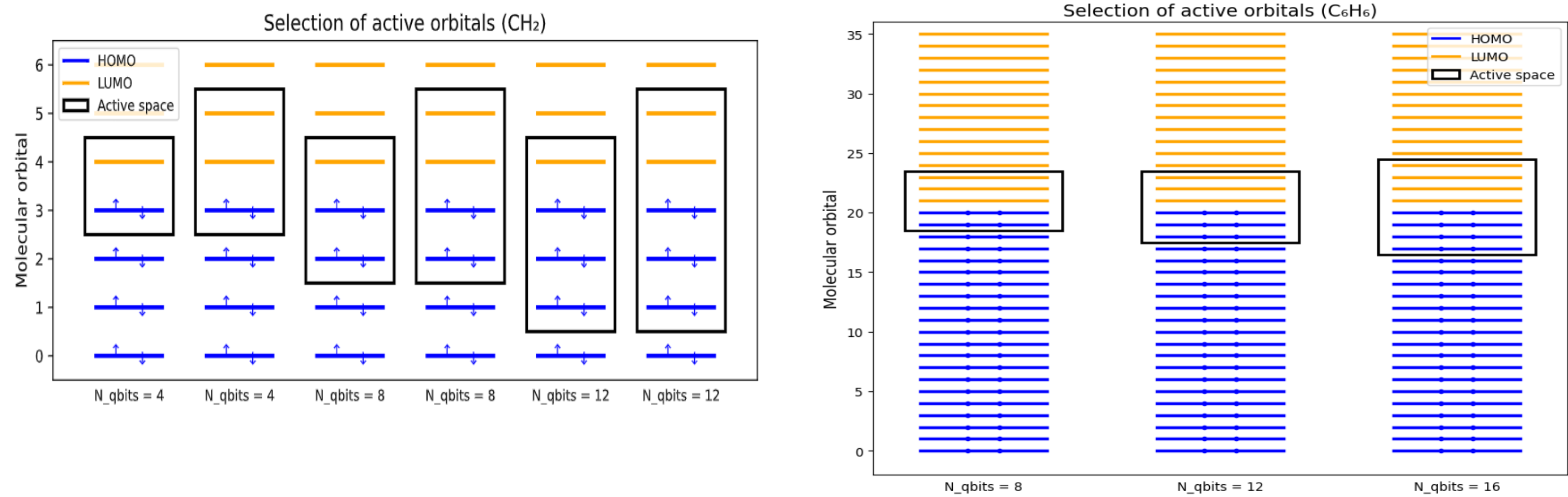
2. Hartree-Fock Approximation

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

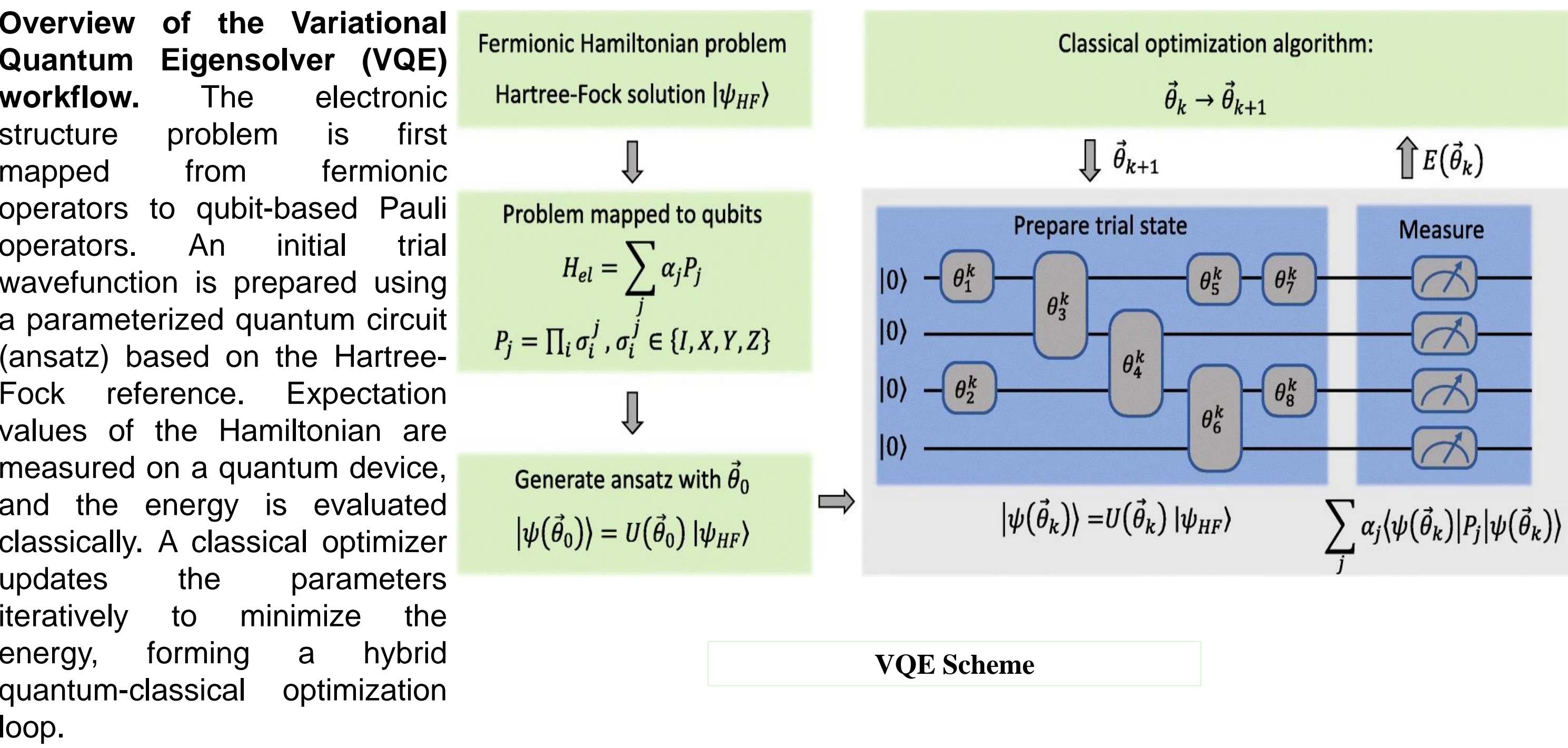
5. Active Space Hamiltonian

Methodology

- We use the CUDA Quantum (CUDA-Q) framework, enabling simulations on both CPU and GPU backends.
- Within the chosen basis set (**sto-3g**) 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.

Conclusion

- 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.

Methylene (CH₂):

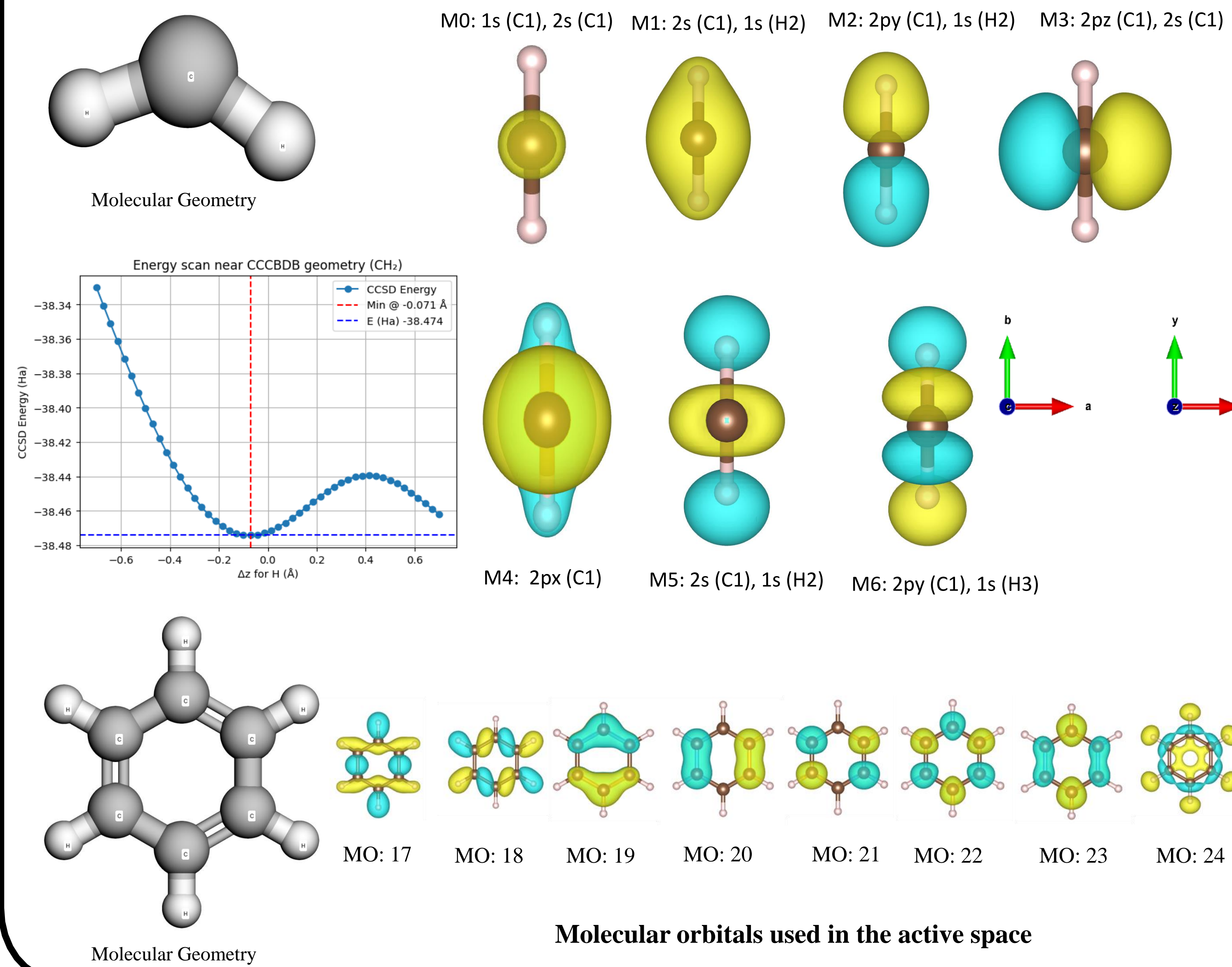
- ✓ For large active space (e.g. nele_cas=6, norb_cas=5), GPU can be ~30× faster than CPU.
- ✓ For small active space, the speedup is marginal or negligible.
- ✓ In one case, CPU was even slightly faster (likely due to overhead/delay in GPU scheduling or under-utilization).

Benzene (C₆H₆):

- ✓ The large active space (e.g. ncore=14, nele_cas=14, norb_cas=8) 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.

Geometry

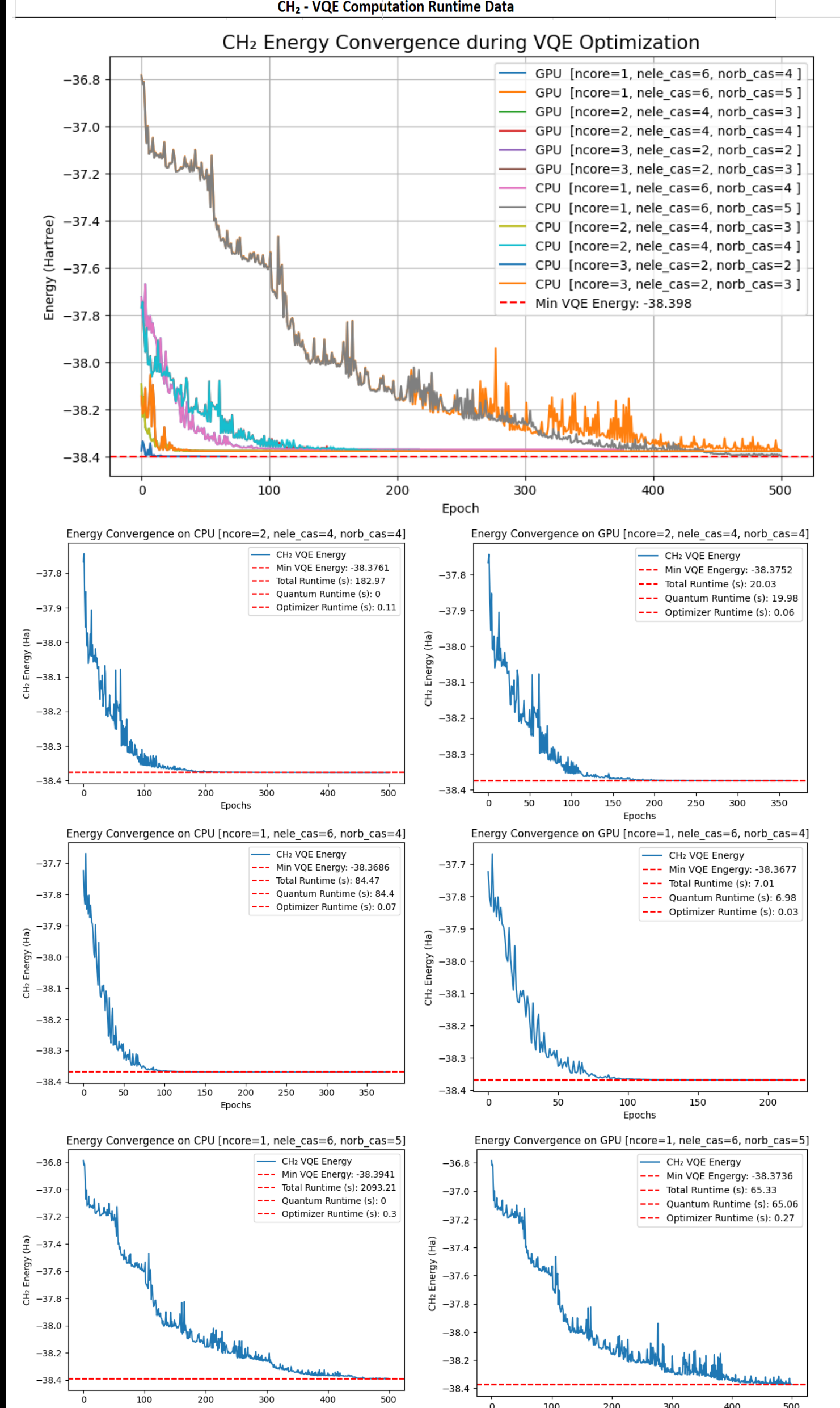
- We take initial geometry coordinates from the Computational Chemistry Comparison and Benchmark Database (CCCBDB).
- Then we scanned more less energy near by it.
- The Isosurface plot of Molecular Orbital using basis set (sto-3g).



Result

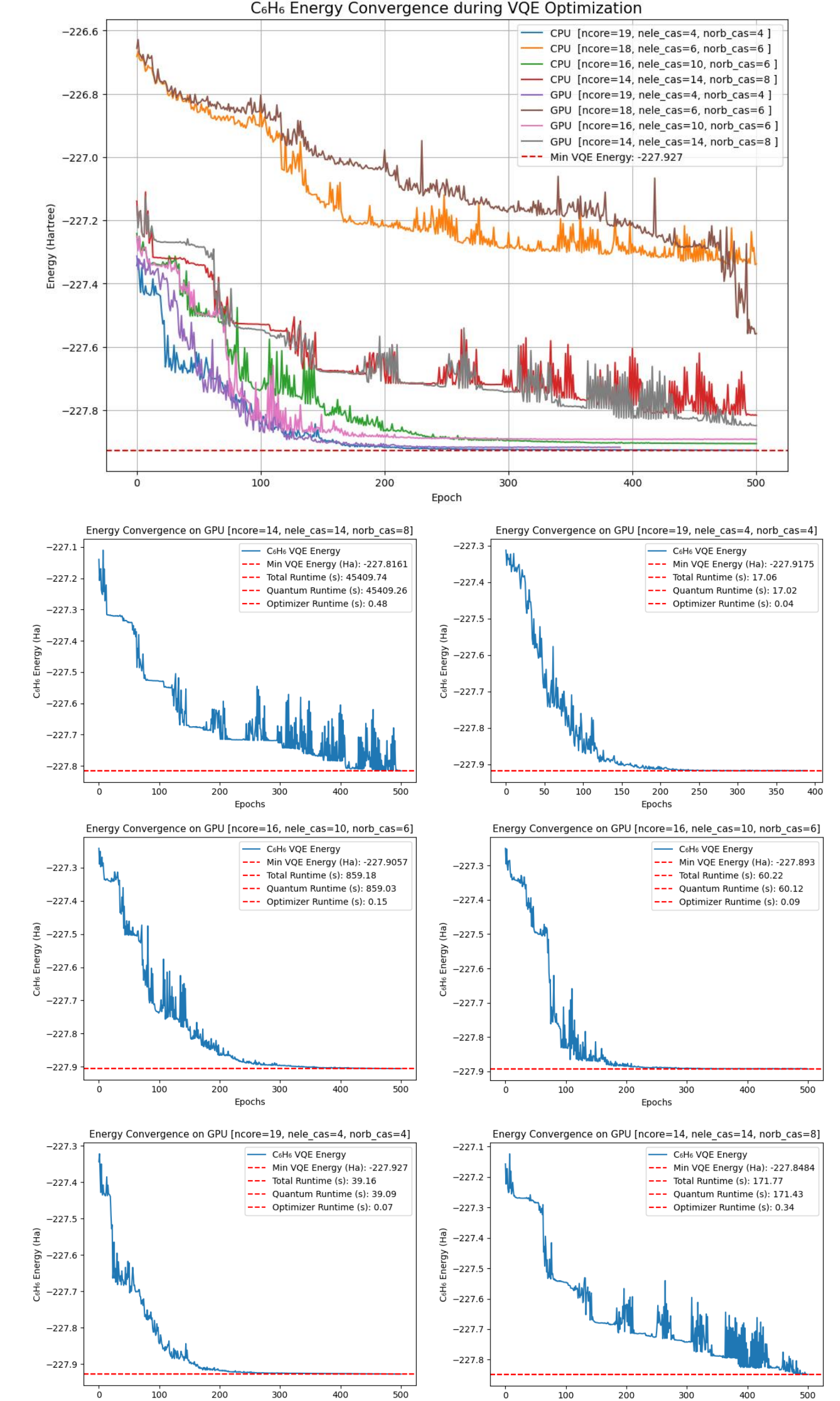
Methylene Results (CH₂)

Active Space	VQE Energy (CPU)	VQE Energy (GPU)	Quantum Simulation Time (s, CPU)	Quantum Simulation Time (s, GPU)	Optimizer Time (s, CPU)	Optimizer Time (s, GPU)	Runtime (s, CPU)	Runtime (s, GPU)	Runtime Speedup (GPU/CPU)
ncore=1, nele_cas=6, norb_cas=4	-38.36862	-38.36769	84.4	0.98	0.07	0.03	84.47	7.01	12.05x
ncore=1, nele_cas=6, norb_cas=5	-38.39409	-38.37362	2092.91	65.06	0.3	0.27	2093.2	65.38	32.04x
ncore=2, nele_cas=4, norb_cas=3	-38.37477	-38.37452	4.84	2.68	0.01	0.01	4.85	2.68	1.68x
ncore=2, nele_cas=4, norb_cas=4	-38.37613	-38.37528	182.87	19.98	0.11	0.06	182.97	20.03	9.13x
ncore=3, nele_cas=2, norb_cas=2	-38.39752	-38.397453	1.96	1.97	0.01	0.01	1.97	1.96	0.99x
ncore=3, nele_cas=2, norb_cas=3	-38.37531	-38.374991	15.22	2.77	0.05	0.01	15.27	2.78	5.49x

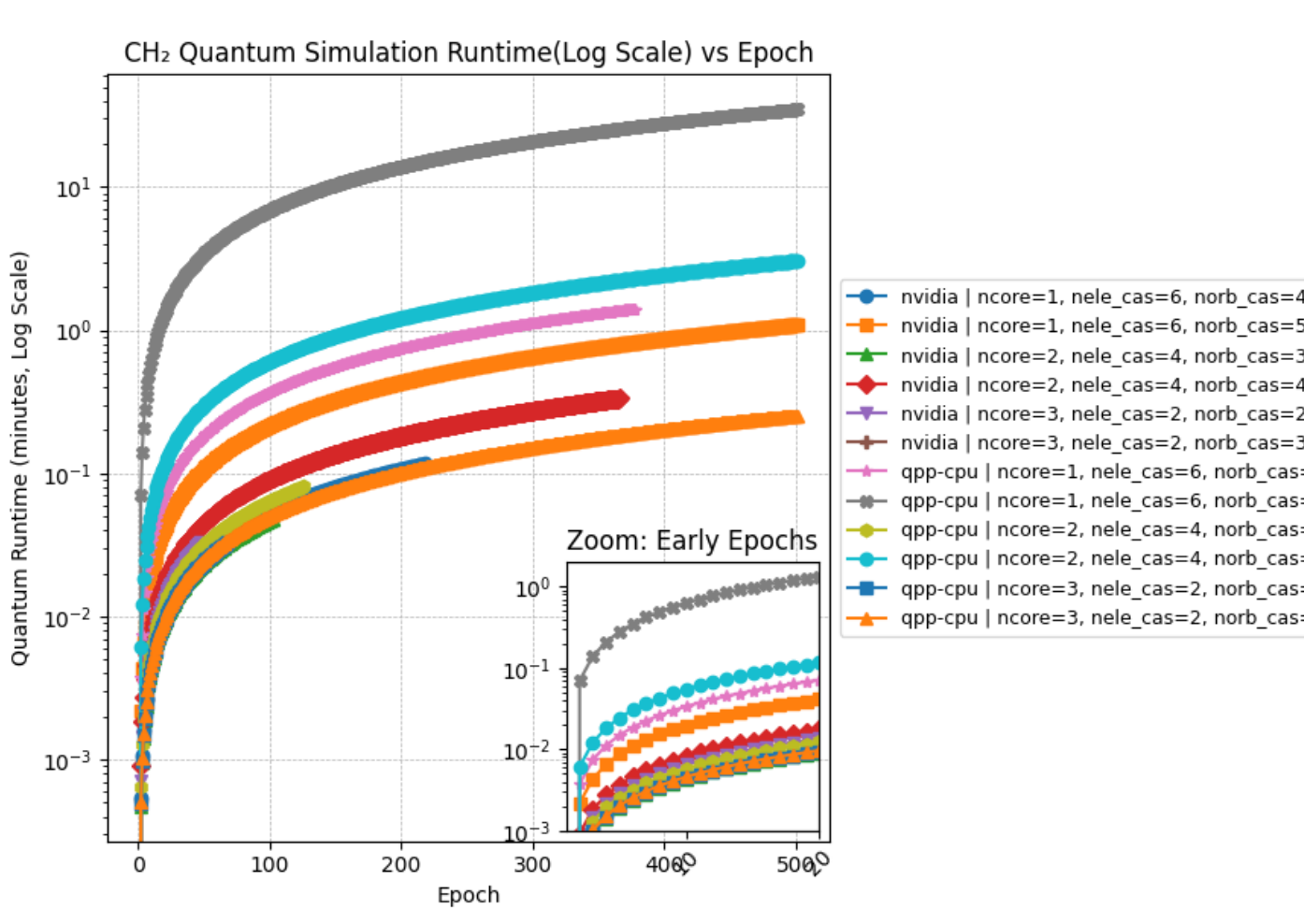


Benzene Results (C₆H₆)

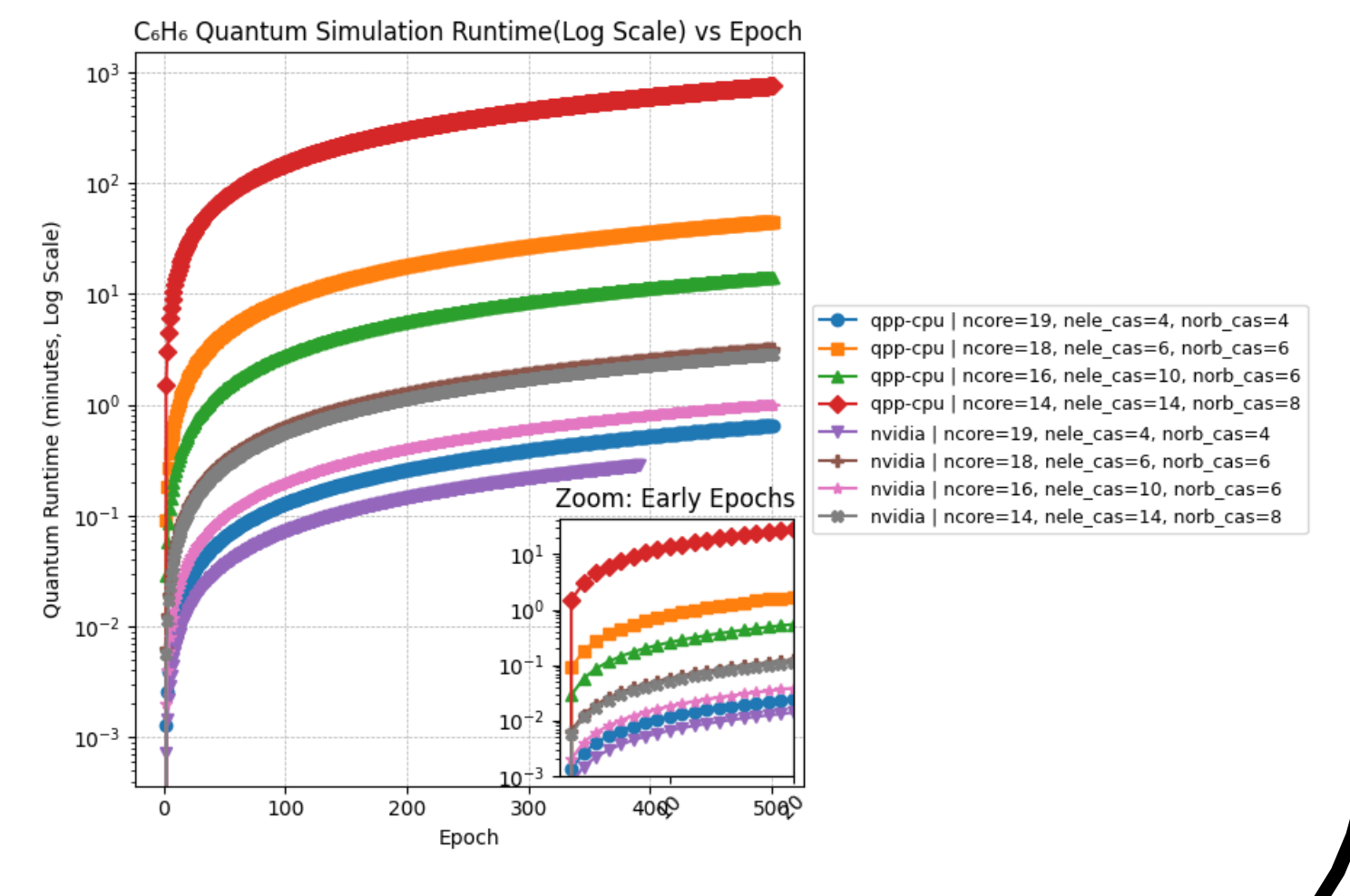
Active Space	VQE Energy (CPU)	VQE Energy (GPU)	Quantum Simulation Time (s, CPU)	Quantum Simulation Time (s, GPU)	Optimizer Time (s, CPU)	Optimizer Time (s, GPU)	Runtime (s, CPU)	Runtime (s, GPU)	Runtime Speedup (GPU/CPU)
ncore=19, nele_cas=4, norb_cas=4	-227.91746	-227.92704	39.09	7.02	0.07	0.04	39.16	7.06	2.30x
ncore=18, nele_cas=6, norb_cas=6	-227.33727	-227.55781	2703.1	192.31	0.72	0.66	2703.81	192.97	14.01x
ncore=16, nele_cas=10, norb_cas=8	-227.89304	-227.90568	859.03	60.12	0.15	0.09	859.18	60.22	14.27x
ncore=14, nele_cas=14, norb_cas=8	-227.84837	-227.81613	45409.3	171.43	0.48	0.34	45409.7	171.77	264.41x



Comparative Analysis of VQE w.r.t Epoch



Comparative Analysis of VQE w.r.t Epoch



References

1 Ieva Liepuoniute, Kirstin D Doney, Javier Robledo-Moreno, Joshua A Job, Will S Friend, and Gavin O Jones. Quantum-centric study of methylene singlet and triplet states. arXiv preprint arXiv:2411.04827, 2024.

2 Konstantin M Makushin and Aleksey K Fedorov. Simulating methylamine using a symmetry-adapted, qubit excitation-based variational quantum eigensolver. Quantum Reports, 7(2):21, 2025.