

Efficiently emulating fermionic circuits for quantum chemistry

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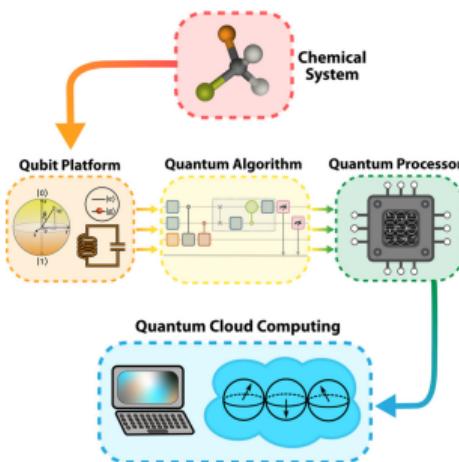
Quantum Chemistry and Quantum Computing

- Chemistry is one of the most highly-anticipated applications of quantum computing.
- Almost every major quantum computing company has a chemistry research division.
- Wavefunctions are inherently quantum objects → may be highly entangled and impossible to represent exactly on a classical computer.

“electronic structure problem” = finding the low-lying energy levels of chemical systems.



Cleaveland Clinic, Ohio



IonQ Forte, Basel

Recent Developments in the Electronic Structure Problem



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Bridging physical intuition and hardware efficiency for correlated electronic states: the local unitary cluster Jastrow ansatz for electronic structure†

Mario Motta, ^a*a Kevin J. Sung, ^bK. Birgitta Whaley, ^{cde} Martin Head-Gordon ^{cf} and James Shee ^{cg}

↪ LUCJ Ansatz, *Chem. Sci.* **14**, 11213 (2023)

Journal of Chemical Theory and Computation > Vol 20/Issue 9 > Article

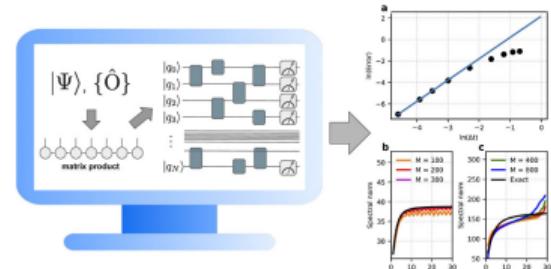
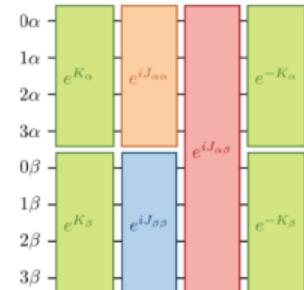


QUANTUM ELECTRONIC STRUCTURE | April 25, 2024

Fast Emulation of Fermionic Circuits with Matrix Product States

Justin Provazza, Klaas Gunst, Huanchen Zhai, Garnet K.-L. Chan, Toru Shiozaki, Nicholas C. Rubin, and Alec F. White*

↪ MPS-FQE Package, *J. Chem. Theory Comput.* **20**, 3719–3728 (2024)



Overview of Open-source Tensor Network Software

	iTensor	TeNPy	quimb
programming language	julia	python	python
supports fermions natively	yes	yes	no*
supports Abelian symmetries	yes	yes	yes
supports non-Abelian symmetries	no*	no*	no*
supports circuits natively	yes	this talk*	yes
supports GPUs	yes	no*	yes
supports GPUs with symmetries	no*	no*	yes

*all of these features are “coming soon”

Motivation:

- ① Develop a best-in-class tensor network emulator for quantum chemistry.
- ② Evaluate wavefunction Ansätze before running on quantum hardware.

Hilbert Space

Andrews, Sung,
Motta, Moreno,
Shee

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Molecular
Hamiltonian

UCC Ansatz

UCJ Ansatz

LUCJ Ansatz

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Orbital Rotation

Coulomb Interaction

LUCJ Layers

Results

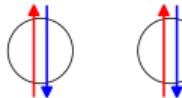
Stretched C₂H₄

N₂ Molecule

Iron-Sulphur Cluster

Conclusion

- Consider (N_α, N_β) electrons occupying N fermion sites.
- The local Hilbert space of each **fermion site** is {empty, up, down, full}.



$$\dim(\mathcal{H}) = 4^N$$

- The local Hilbert space of each **qubit site** is {empty, full}.



$$\dim(\mathcal{H}) = 2^{2N}$$

- Using symmetries, we can greatly reduce the total Hilbert space dimension.
 - **Number symmetry:** $N_e = N_\alpha + N_\beta$.
 - **Spin symmetry:** $2S_z = N_\alpha - N_\beta$.

$$\dim(\mathcal{H}) = \binom{N}{N_\alpha} \binom{N}{N_\beta}.$$

Express the wavefunction in an occupation number basis $|n_0, n_1, \dots, n_{N-1}\rangle$ with $n_i \in \{0, 1\}$.

$$H = \sum_{\sigma, pq} h_{pq} a_{\sigma, p}^\dagger a_{\sigma, q} + \frac{1}{2} \sum_{\sigma\tau, prqs} h_{prqs} a_{\sigma, p}^\dagger a_{\tau, r}^\dagger a_{\tau, s} a_{\sigma, q} + C.$$

one-body tensor two-body tensor constant

- The one-body tensor is an N^2 Hermitian matrix, such that $h_{pq} = h_{qp}^*$.
 - The two-body tensor is an N^4 tensor, such that $h_{pqrs} = h_{rspq}$, $h_{pqrs} = h_{qpsr}^*$.

In a **double-factorized** representation,

$$H = \sum_{\sigma, pq} h'_{pq} a_{\sigma, p}^\dagger a_{\sigma, q} + \sum_{\mu=1}^L \mathcal{U}_\mu \boxed{\mathcal{J}_\mu} \mathcal{U}_\mu^\dagger + C' =$$


diagonal Coulomb operator

Unitary Coupled Cluster (UCC) Ansatz

Gold standard for the electronic structure problem.

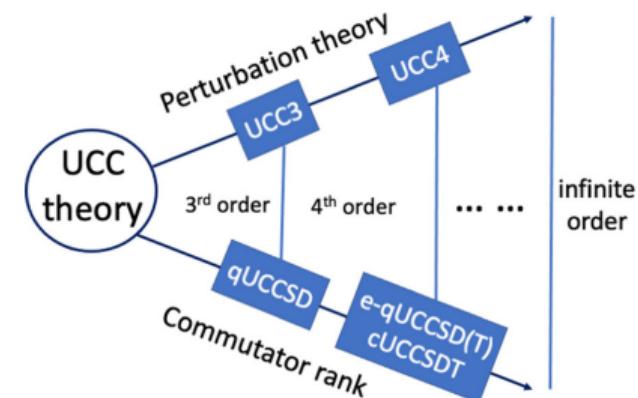
anti-Hermitian cluster operator

$$|\Psi_{\text{UCC}}\rangle = \exp(\sigma) |\Phi_0\rangle, \quad \text{where } \sigma = T - T^\dagger.$$

Hartree-Fock wavefunction

We define

$$T = \underbrace{\sum_{ia} t_i^a a_a^\dagger a_i}_{\text{singles (S)}} + \underbrace{\sum_{ijab} t_{ij}^{ab} a_a^\dagger a_b^\dagger a_j a_i}_{\text{doubles (D)}} + \dots$$

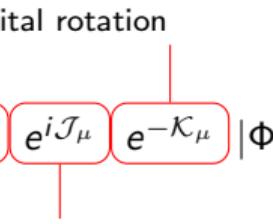


Transformed $e^{-\sigma} H e^\sigma$ is now Hermitian but commutator expansion is nonterminating. We typically take the lowest-order reasonable truncation, which is quadratic (q) \rightarrow qUCCSD.

Unitary Cluster Jastrow (UCJ) Ansatz

An alternative Ansatz focuses on capturing short-range two-body correlations.

$$|\Psi_{\text{UCJ}}\rangle = \prod_{\mu=1}^L e^{i\mathcal{K}_\mu} e^{i\mathcal{J}_\mu} e^{-i\mathcal{K}_\mu} |\Phi_0\rangle, \quad \text{where} \quad \begin{cases} \mathcal{K}_\mu = \sum_{\sigma,pq} K_{pq}^\mu a_{\sigma,p}^\dagger a_{\sigma,q} \\ \mathcal{J}_\mu = \sum_{\sigma\tau,pq} J_{\sigma\tau,pq}^\mu n_{\sigma,p} n_{\tau,q} \end{cases}$$

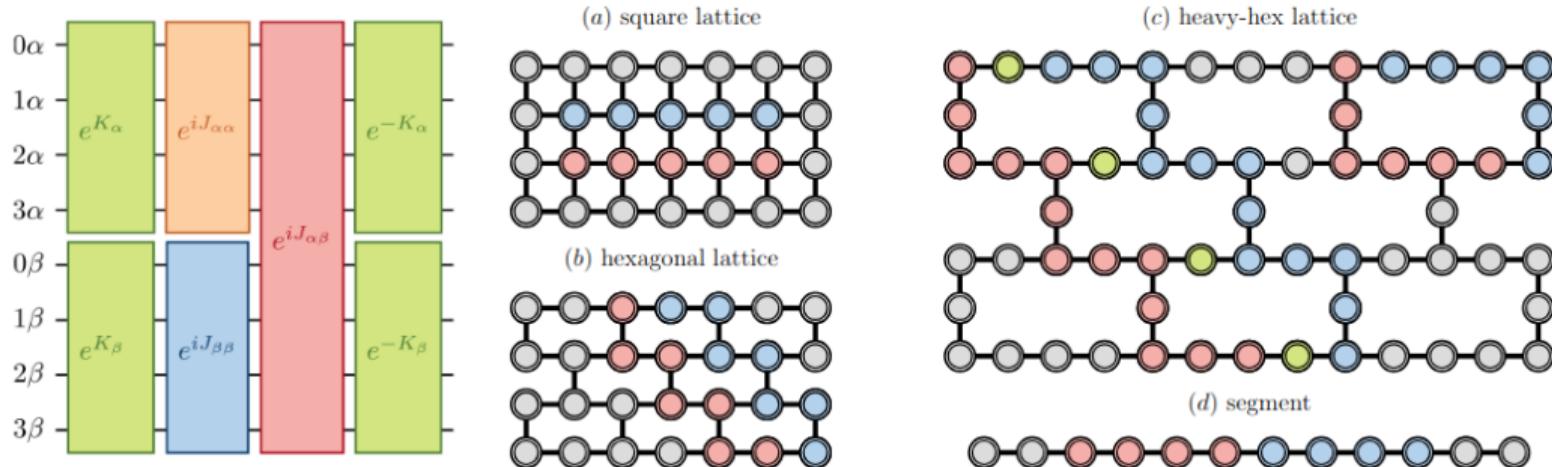
orbital rotation

diagonal Coulomb interaction

- ✓ can be derived as a double-factorized low-rank decomposition of the qUCCD Ansatz.
- ✓ full configuration interaction (FCI) wavefunction is recovered by stacking layers, L .
- ✓ promising accuracy with reduced circuit depths compared to qUCCSD.
- ✗ diagonal Coulomb interaction requires all-to-all connectivity or fermionic swap network.

Local Unitary Cluster Jastrow (LUCJ) Ansatz

Inspired by the Hubbard model, we impose a **sparsity constraint** on the diagonal Coulomb matrix that enforces **locality** on a given qubit topology.

$$|\Psi_{\text{LUCJ}}\rangle = \prod_{\mu=1}^L \mathcal{U}_\mu e^{i\mathcal{J}_\mu} \mathcal{U}_\mu^\dagger |\Phi_0\rangle, \quad \text{with} \quad \mathcal{J}_\mu = \frac{1}{2} \sum_{\sigma\tau,pq} J_{\sigma\tau,pq}^\mu n_{\sigma,p} n_{\tau,q}.$$

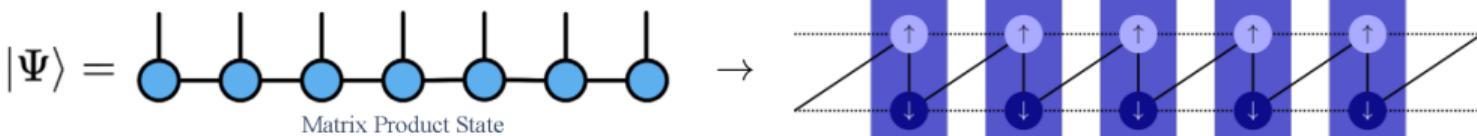


Fermion↔Qubit Mapping

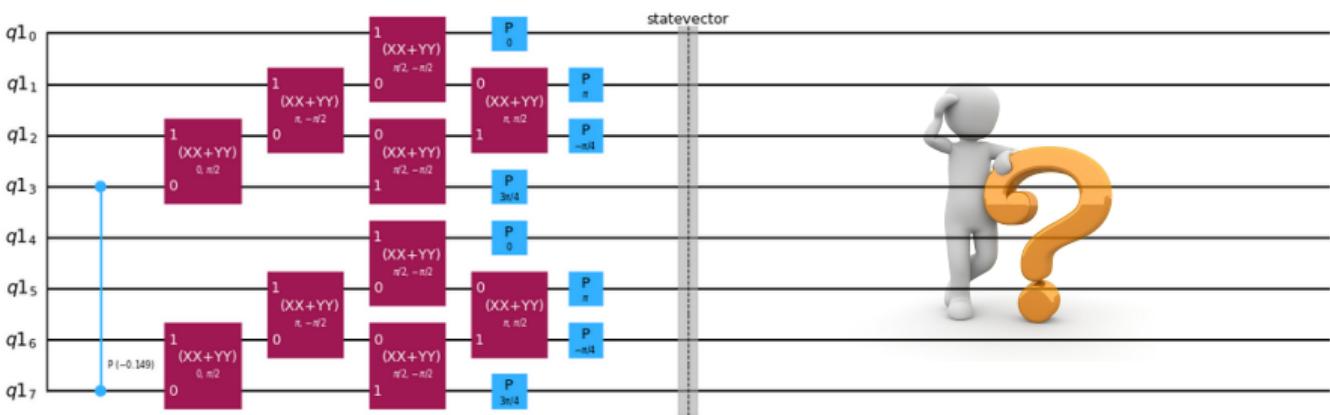
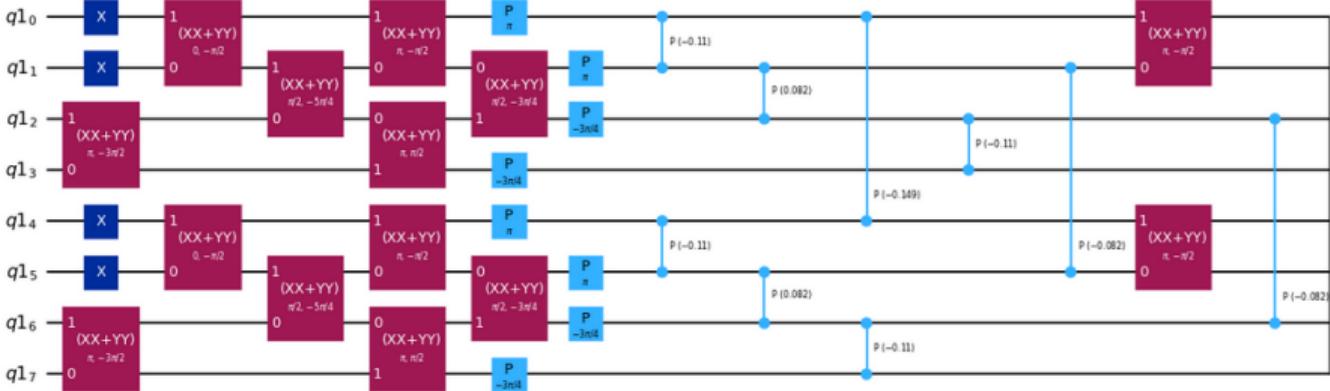
There are many ways to map fermions↔qubits, which find a balance between the locality of the operators versus the locality of the state: Jordan-Wigner, Bravyi-Kitaev, parity mapping, Verstraete-Cirac, Derby-Klaasen, etc.

We choose the **Jordan-Wigner mapping**, due to the 1D locality constraints on $|\Psi\rangle$.

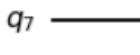
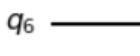
$$\begin{cases} n_i = (Z_i + 1)/2 \\ a_i = (-1)^{\sum_{l < j} n_l} (X_i - iY_i)/2 \\ a_i^\dagger = (-1)^{\sum_{l < j} n_l} (X_i + iY_i)/2 \end{cases} \leftrightarrow \begin{cases} X_i = (a_i^\dagger + a_i)Z_{i-1} \dots Z_0 \\ Y_i = i(a_i^\dagger - a_i)Z_{i-1} \dots Z_0 \\ Z_i = 2n_i - 1 \end{cases}$$



Anatomy of an LUCJ Circuit ($|\Psi_{\text{LUCJ}}\rangle = \prod_{\mu=1}^L \mathcal{U}_\mu e^{i\mathcal{J}_\mu} \mathcal{U}_\mu^\dagger |\Phi_0\rangle$)



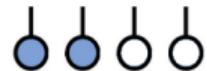
Step 1: Prepare Hartree-Fock State



We start with an “optimal” Slater determinant initial state.

- Fermion language:

- Prepare a product state by initializing N sites: $\{\uparrow\downarrow, \uparrow\downarrow, \emptyset, \emptyset\}$.
- HF state has electrons occupying the lowest numbered sites.

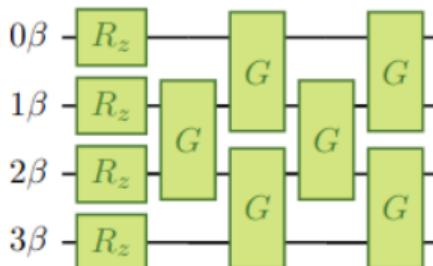
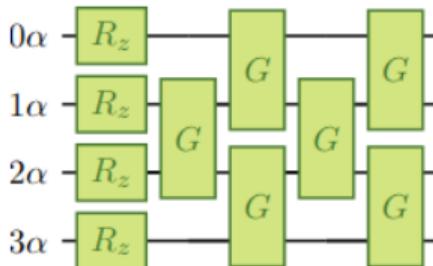


- Qubit language:

- Prepare a product state of $2N$ qubits by applying X gates.
- HF state has electrons occupying lowest numbered orbitals.

Step 2: Apply Orbital Rotation

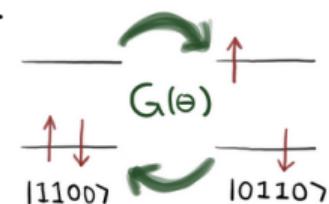
Fermion language



$$\mathcal{U}a_{\sigma,i}^\dagger \mathcal{U}^\dagger = \sum_j U_{ji} a_{\sigma,j}^\dagger.$$

Any unitary matrix can be decomposed into **Givens rotations**:

$$U = DG_{N-1}^* G_{N-2}^* \dots G_0^*.$$



- ① Apply a layer of number interactions.
- ② Apply a sequence of Givens rotations.

$$G(\theta, \phi) = \prod_{\sigma} e^{i\phi a_{\sigma,p}^\dagger a_{\sigma,p}} e^{\theta(a_{\sigma,p}^\dagger a_{\sigma,q} - a_{\sigma,q}^\dagger a_{\sigma,p})} e^{-i\phi a_{\sigma,p}^\dagger a_{\sigma,p}}.$$

$$N(\theta) = \prod_{\sigma} e^{i\theta a_{\sigma,p}^\dagger a_{\sigma,p}}$$

Step 2: Apply Orbital Rotation

Qubit language

Andrews, Sung,
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LUCJ Layers

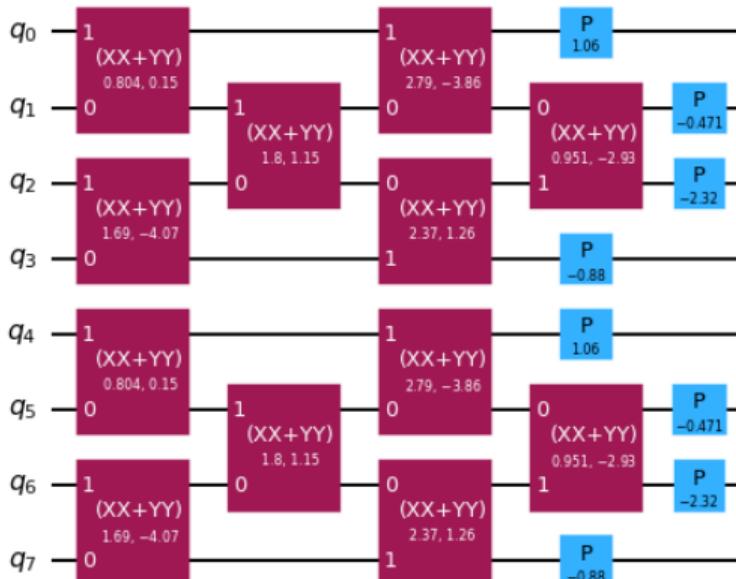
Results

Stretched C₂H₄

N₂ Molecule

Iron-Sulphur Cluster

Conclusion



- ① Apply a sequence of XX+YY gates.
- ② Apply a layer of phase gates.

$$XY(\theta, \beta) = RZ_0(-\beta)e^{-i\frac{\theta}{4}(XX+YY)}RZ_0(\beta)$$

$$P(\theta) = e^{i\theta/2}RZ(\theta)$$

NB: XX+YY and XY-YX rotations are equivalent up to $S = \sqrt{Z}$ gates.

Step 2: Apply Orbital Rotation

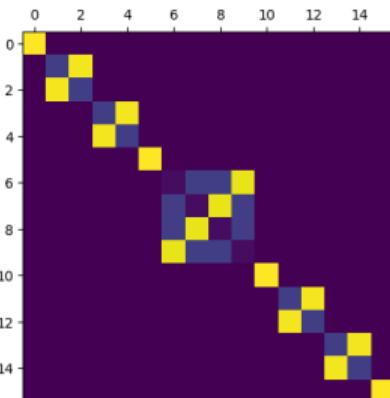
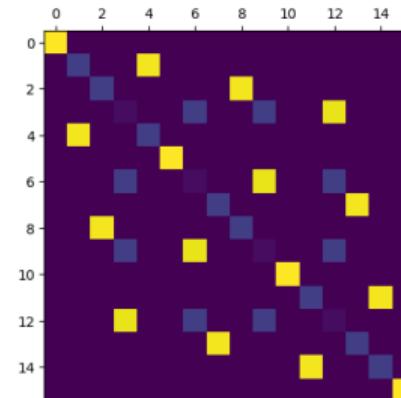
Interlude: symmetries

For fast fermionic emulation, it is crucial to exploit number and spin symmetries.

Consider the tensor product space of fermionic 2-qudit gates: $\{\emptyset, \uparrow, \downarrow, \uparrow\downarrow\} = \{0, 1, 2, 3\}$.

$$\begin{pmatrix} 0 \\ 1 \\ 2 \\ 3 \end{pmatrix} \otimes \begin{pmatrix} 0 \\ 1 \\ 2 \\ 3 \end{pmatrix} = \begin{pmatrix} 00 \\ 01 \\ 02 \\ 03 \\ 10 \\ 11 \\ 12 \\ 13 \\ 20 \\ 21 \\ 22 \\ 23 \\ 30 \\ 31 \\ 32 \\ 33 \end{pmatrix} \rightarrow \begin{pmatrix} (N_e, 2S_z) \\ 22 \\ 20 \\ 02 \\ 23 \\ 32 \\ 00 \\ 12 \\ 13 \\ 21 \\ 03 \\ 30 \\ 12 \\ 33 \\ 01 \\ 10 \\ 31 \\ 13 \\ 11 \end{pmatrix}$$

$$G \rightarrow PGP^T$$



Step 3: Apply Coulomb Interaction

Fermion language

Andrews, Sung,
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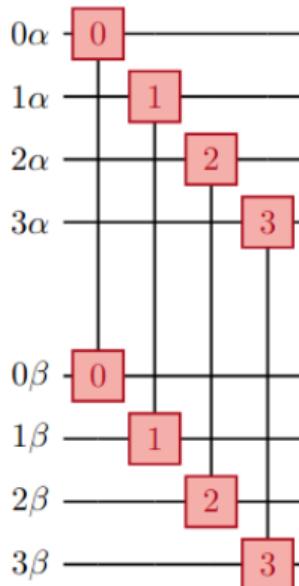
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Conclusion



$$e^{i\mathcal{J}} = \exp \left(\frac{i}{2} \sum_{\sigma\tau,pq} J_{\sigma\tau,pq} n_{\sigma,p} n_{\tau,q} \right)$$

- ① Apply a sequence of number-number interactions.
- ② Apply a layer of on-site interactions.

$$NN(\theta) = \prod_{\sigma} e^{i\theta a_{\sigma,p}^{\dagger} a_{\sigma,p} a_{\sigma,q}^{\dagger} a_{\sigma,q}}$$

$$OS(\theta) = e^{i\theta a_{\alpha,p}^{\dagger} a_{\alpha,p} a_{\beta,p}^{\dagger} a_{\beta,p}}$$

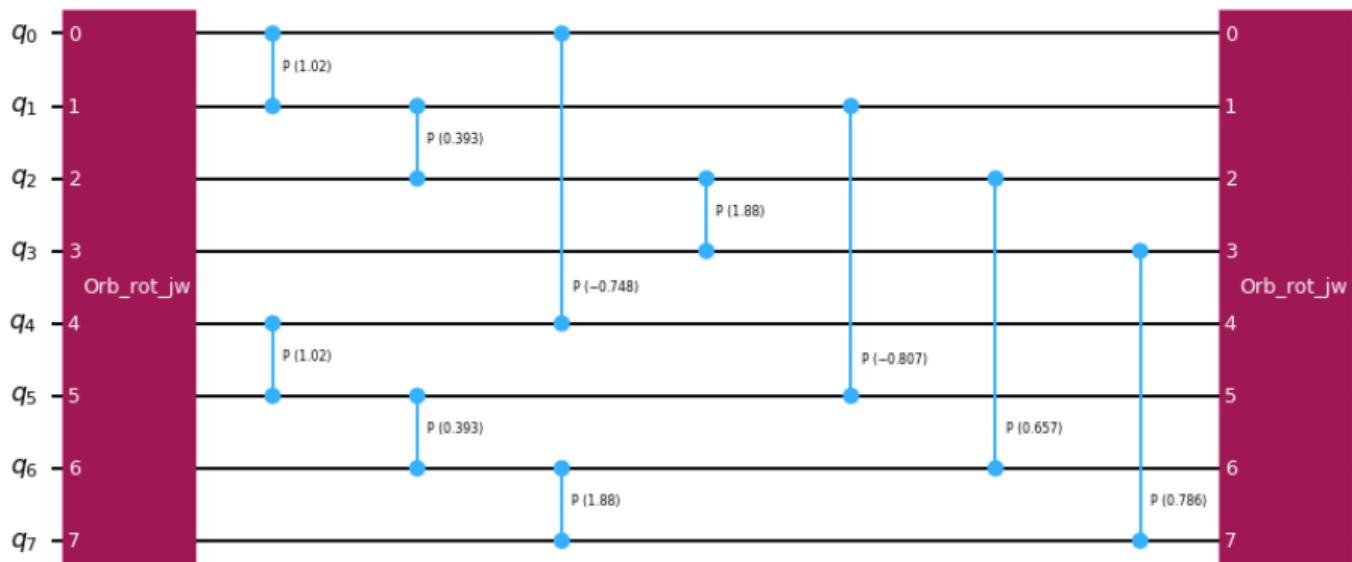


Step 3: Apply Coulomb Interaction

Qubit language

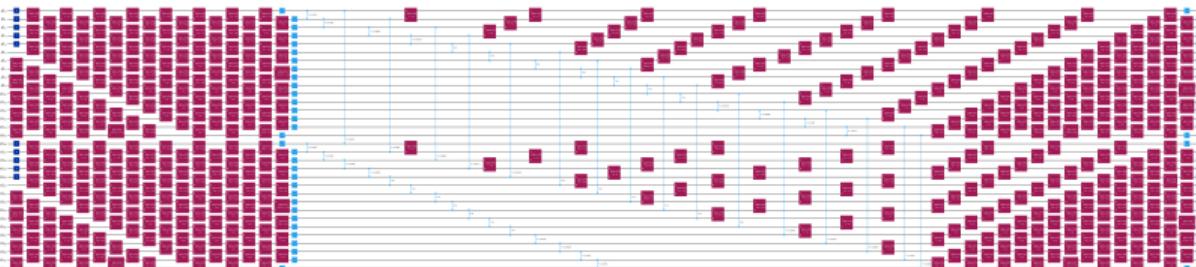
- 1 Apply a sequence of controlled phase gates.

$$CP(\theta) = I \otimes |0\rangle\langle 0| + P(\theta) \otimes |1\rangle\langle 1|.$$

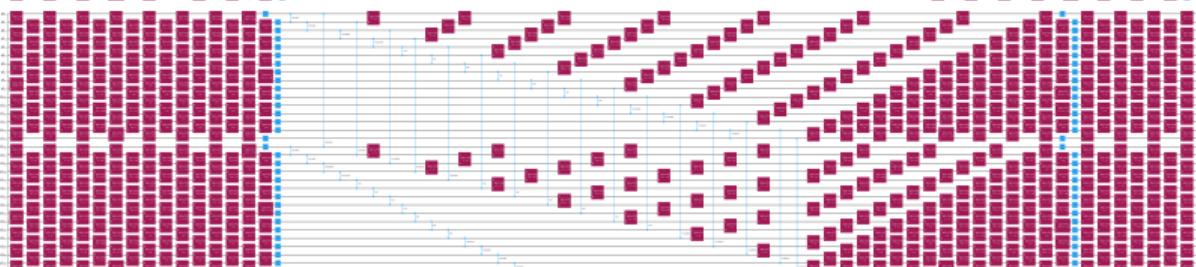


Step 4: Repeat Layers ($|\Psi_{\text{LUCJ}}\rangle = \prod_{\mu=1}^L \mathcal{U}_\mu e^{i\mathcal{J}_\mu} \mathcal{U}_\mu^\dagger |\Phi_0\rangle$)

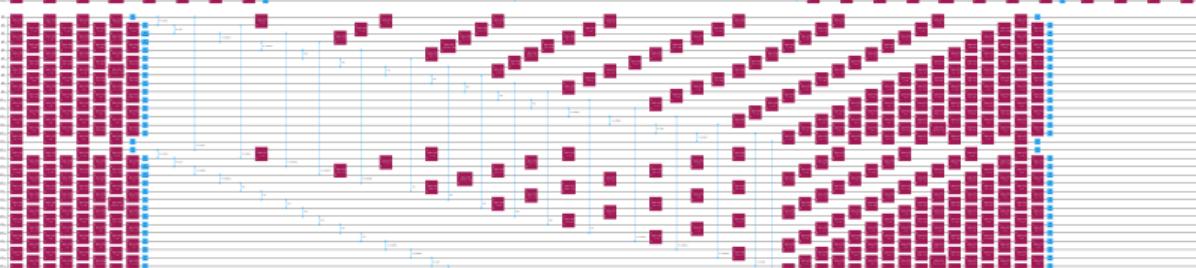
L=1



L=2



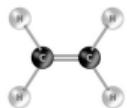
L=3



etc.

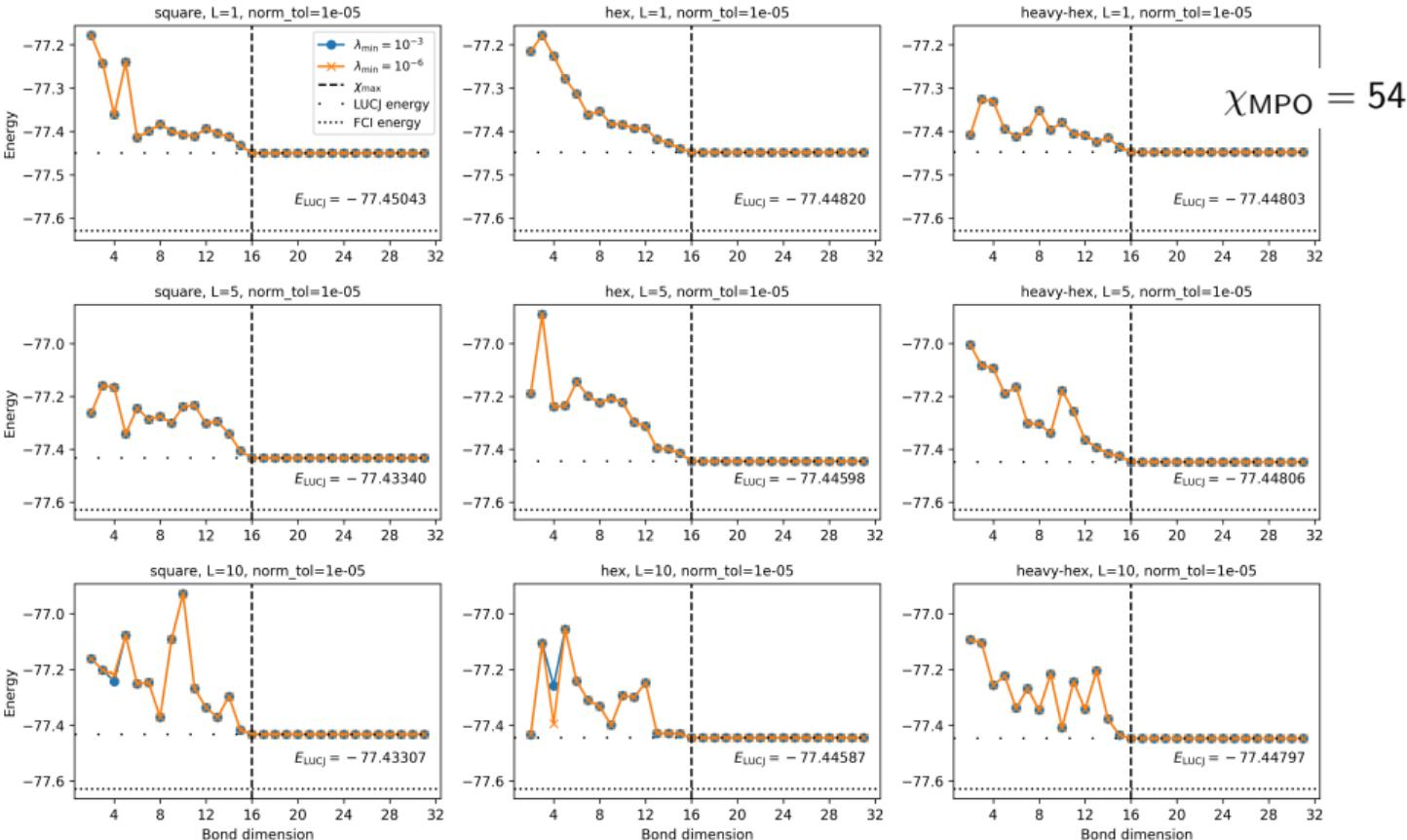
Efficiently
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for quantum
chemistry

Stretched C₂H₄



c2h4_sto-6g_4e4o_d-2.68

(16 electrons)

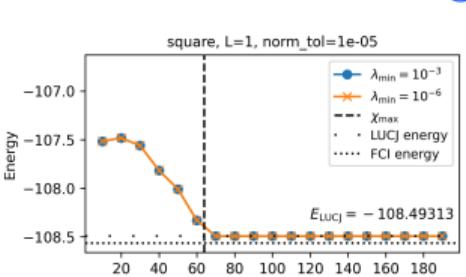


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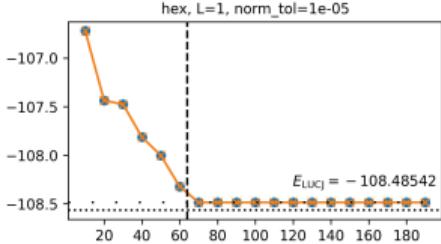
N₂ Molecule



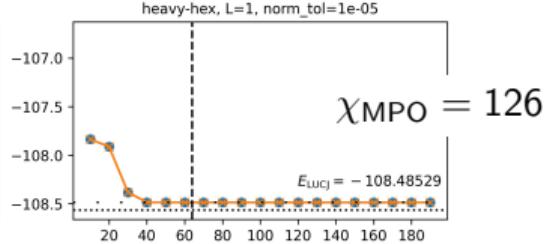
(14 electrons)



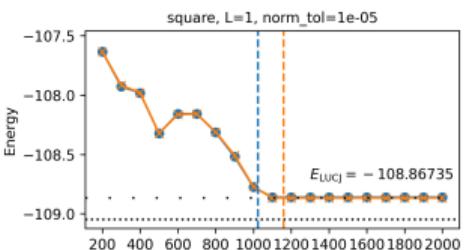
(1) n2_sto-6g_6e6o_d-1.00



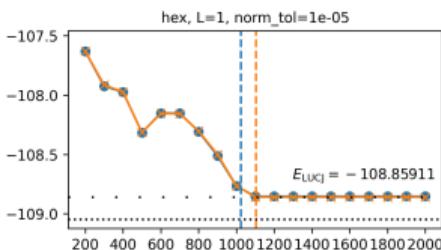
hex, L=1, norm_tol=1e-05



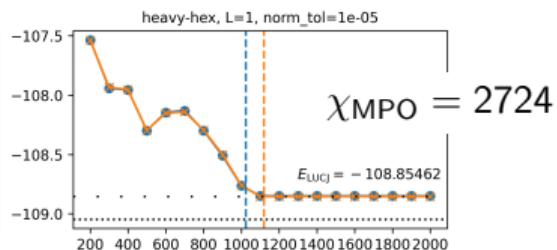
$\chi_{\text{MPO}} = 126$



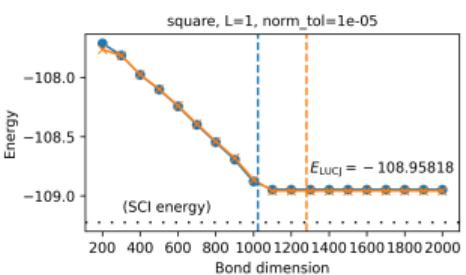
(2) n2_6-3lg_10e16o_d-1.00



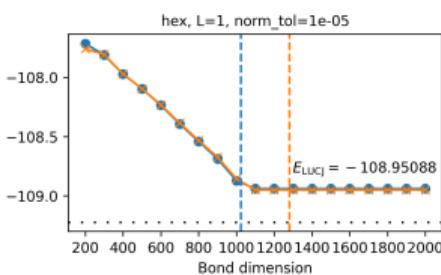
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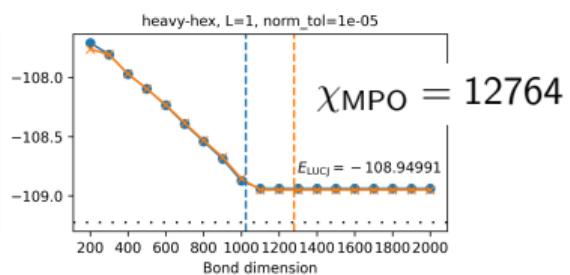
$\chi_{\text{MPO}} = 2724$



(3) n2_cc-pvdz_10e26o_d-1.00



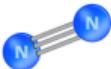
hex, L=1, norm_tol=1e-05



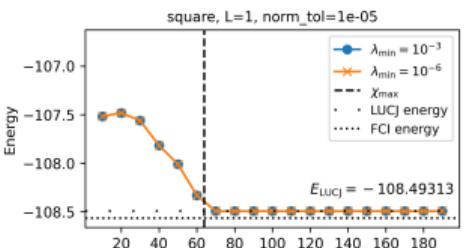
$\chi_{\text{MPO}} = 12764$

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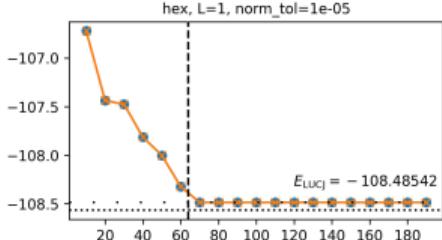
N₂ Molecule



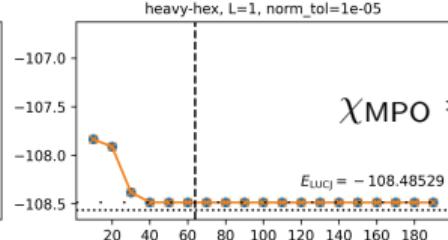
(14 electrons)



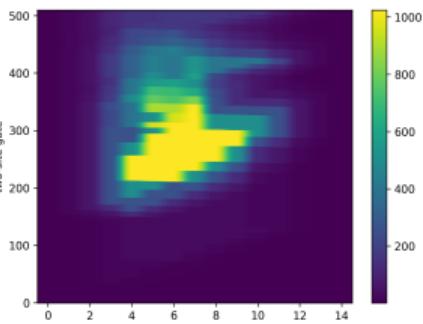
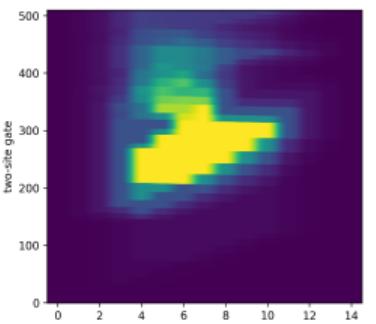
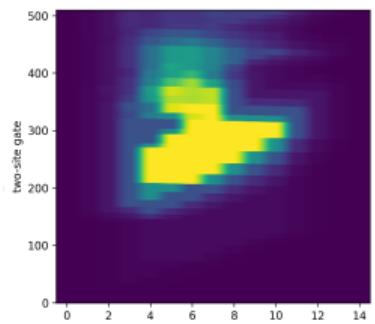
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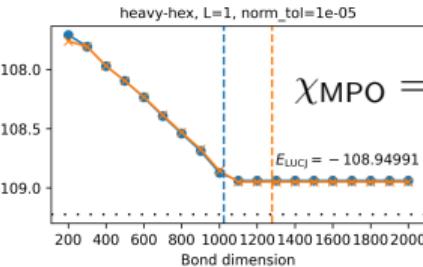
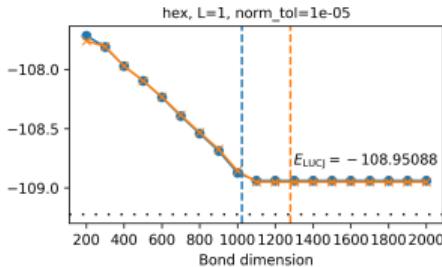
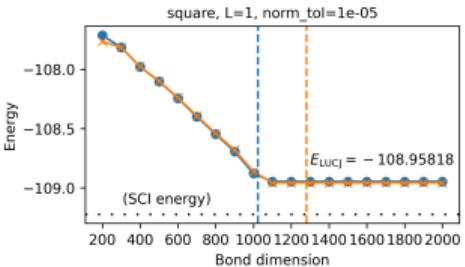
hex, L=1, norm_tol=1e-05



$\chi_{\text{MPO}} = 126$



2724



$\chi_{\text{MPO}} = 12764$

Andrews, Sung,
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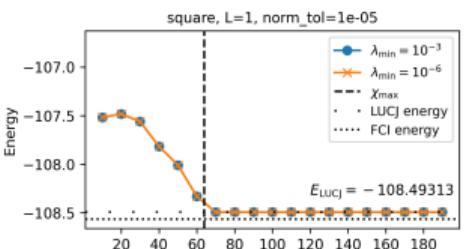
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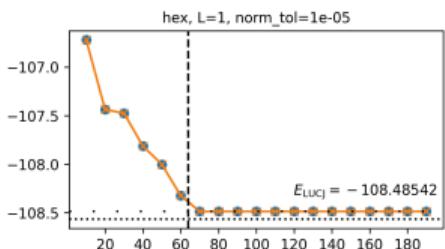
N₂ Molecule



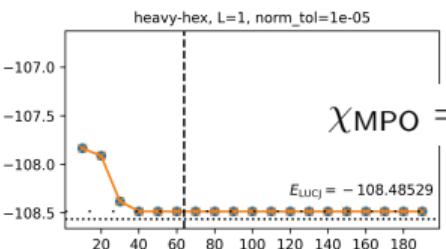
(14 electrons)



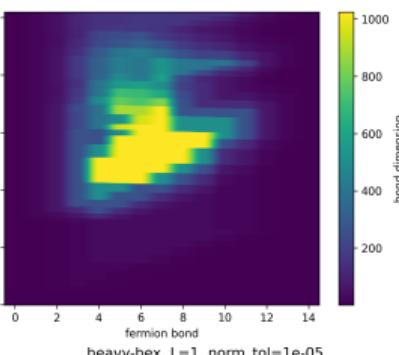
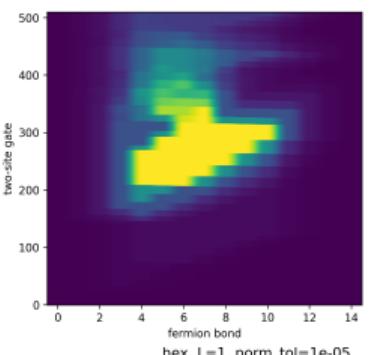
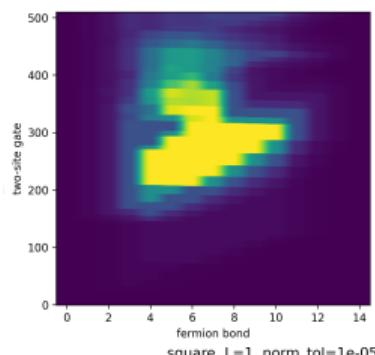
(1) n2_sto-6g_6e6o_d-1.00



hex, L=1, norm_tol=1e-05



$\chi_{\text{MPO}} = 126$



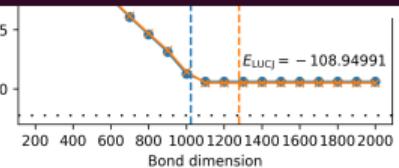
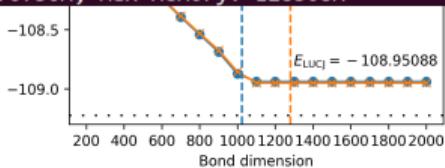
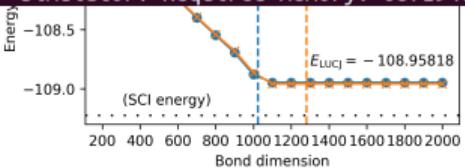
2724

square, L=1, norm_tol=1e-05

hex, L=1, norm_tol=1e-05

heavy-hex, L=1, norm_tol=1e-05

```
qiskit.exceptions.QiskitError: 'ERROR: [Experiment 0] Insufficient memory to run circuit circuit-166 using the statevector simulator. Required memory: 68719476736M, max memory: 128308M , ERROR: Insufficient memory to run circuit circuit-166 using the statevector simulator. Required memory: 68719476736M, max memory: 128308M'
```



Andrews, Sung,
Motta, Moreno,
Shee

Introduction

Theory

Hilbert Space

Molecular
Hamiltonian

UCC Ansatz

UCJ Ansatz

LUCJ Ansatz

Fermion Mapping

Method

Circuit Anatomy

Hartree-Fock State

Orbital Rotation

Coulomb Interaction

LUCJ Layers

Results

Stretched C₂H₄

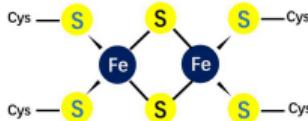
N₂ Molecule

Iron-Sulphur Cluster

Conclusion

Efficiently
emulating
fermionic circuits
for quantum
chemistry

Iron-Sulphur Cluster



$\text{Fe}_2\text{S}_2 = (84 \text{ electrons})$

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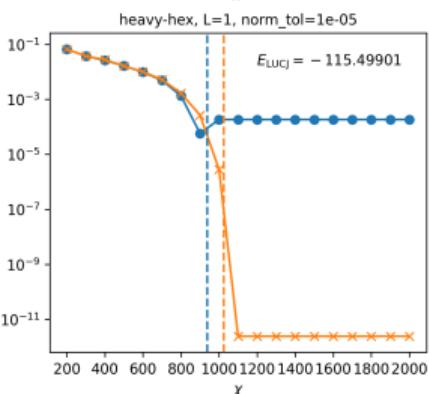
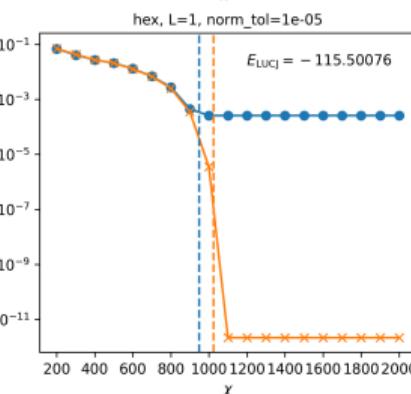
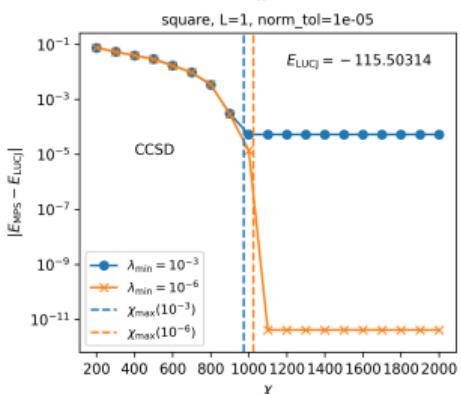
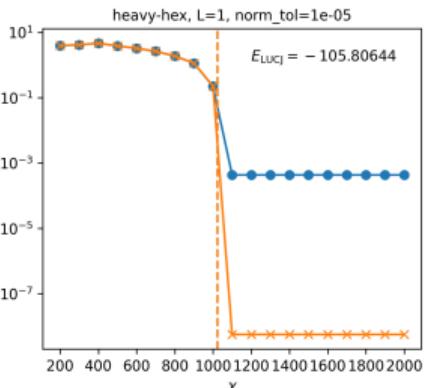
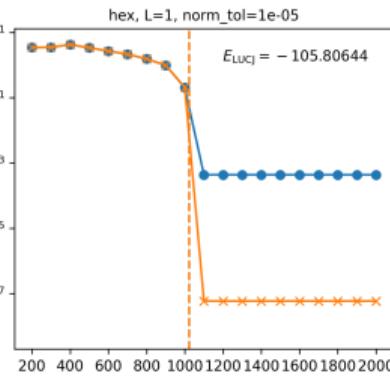
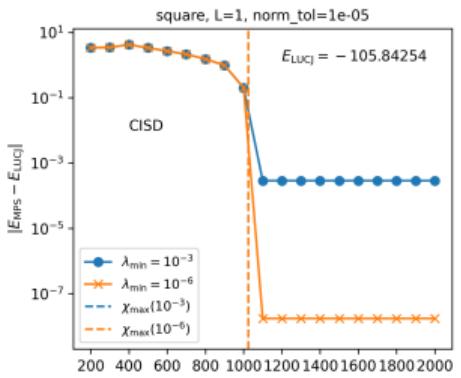
Results

Stretched C_2H_4

N_2 Molecule

Iron-Sulphur Cluster

Conclusion

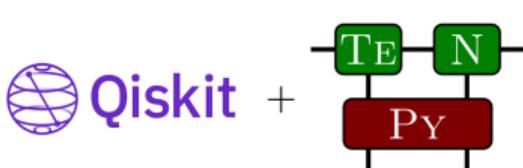


Conclusion

- We have constructed an open-source MPS emulator of fermionic quantum circuits, which takes advantage of number and spin symmetry.
- We have interfaced Qiskit and TeNPy, such that fermionic circuits can be constructed either directly, or by transpiling a qubit circuit.
- We have used this to efficiently emulate the LUCJ Ansatz at system sizes that are costly or inaccessible on current quantum hardware.

Outlook:

- Variational MPS optimization methods and realistic noise models.
- Larger strongly-correlated molecules targeting industrial applications.
- Improved wavefunction Ansätze and regimes of quantum advantage.



Contributions always welcome!