Improving VQE accuracy for H4 molecule with shot limits

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QuTech

Ansatz

Default: Hardware-efficient ansatz ***Not good!***

- Barren Plataeu
- Inefficient

Unitary Coupled Cluster(UCC) Ansatz

$$U(\boldsymbol{\theta}) = \exp\left(T(\boldsymbol{\theta}) - T^*(\boldsymbol{\theta})\right)$$

T sums over all excitation: $T = T_1 + T_2 + \cdots$ Practically, $T = T_1 + T_2$ is enough in our case.

Single excitation: $T_1 = \sum \theta_{pr} c_p^* c_r$ p: occupied orbital. r: unoccupied orbital.

Double excitation: $T_2 = \sum \theta_{pqrs} c_p^* c_q^* c_r c_s$ p & q: occupied orbital. r & s: unoccupied orbital.

UCCSD Ansatz

$$T_1 = \sum \theta_{pr} c_p^* c_r \qquad T_2 = \sum \theta_{pqrs} c_p^* c_q^* c_r c_s$$

How to express $U(\boldsymbol{\theta})$ in circuit ansatz form? – **Trotterization**

$$U(\boldsymbol{\theta}) = \exp(T_1(\boldsymbol{\theta_1}) + T_2(\boldsymbol{\theta_2}) - h.c.) \approx \left[\exp(T_1(\boldsymbol{\theta_1}) - h.c.)^{1/n} \exp(T_2(\boldsymbol{\theta_2}) - h.c.)^{1/n}\right]^n$$

Take n=1(A rough but practical choice)

$$U(\boldsymbol{\theta}) \approx \exp\big(T_1(\boldsymbol{\theta_1})\big) \exp(-T_1^*(\boldsymbol{\theta_1})) * \exp\big(T_2(\boldsymbol{\theta_2})\big) \exp(-T_2^*(\boldsymbol{\theta_2}))$$

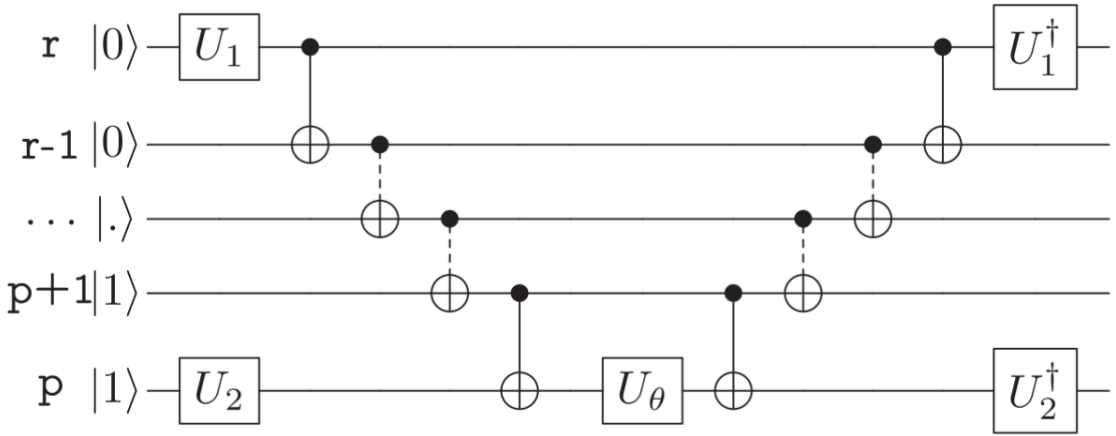
Jordan-Wigner Transform:

$$\exp\big(\theta_{pr} c_p^* c_r\big) = \exp\left(\frac{i\theta_{pr}}{2} \bigotimes_{a=r+1}^{p-1} Z_a Y_r X_p\right)$$

$$\exp\big(\theta_{pqrs} c_p^* c_q^* c_r c_s\big) = \exp\left(\frac{i\theta_{pqrs}}{8} \bigotimes_{b=s+1}^{r-1} Z_b \bigotimes_{a=q+1}^{p-1} Z_a \big(X_s X_r Y_q X_p + Y_s X_r Y_q Y_p + X_s Y_r Y_q Y_p + X_s X_r X_q Y_p - h.c.\big)\right)$$

Single Excitation circuit

- Repeat Twice for each excitation gate

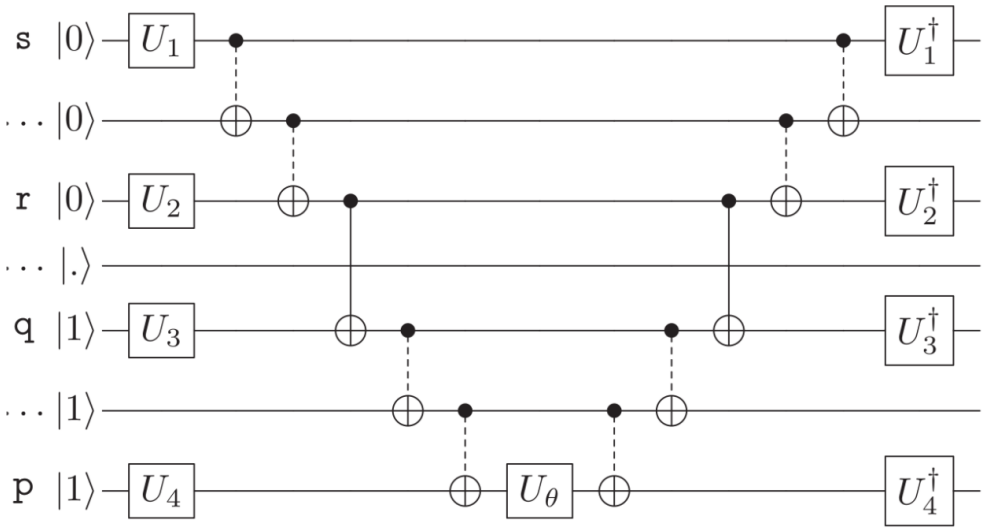


$$[U_1, U_2, U_\theta] = \left\{ \left[R_x(-\pi/2), H, R_z(\theta/2) \right], \left[H, R_x(-\frac{\pi}{2}), R_z(-\theta/2) \right] \right\}$$

Figure: Pennylane

Double Excitation circuit

- Repeat eight times for each excitation gate

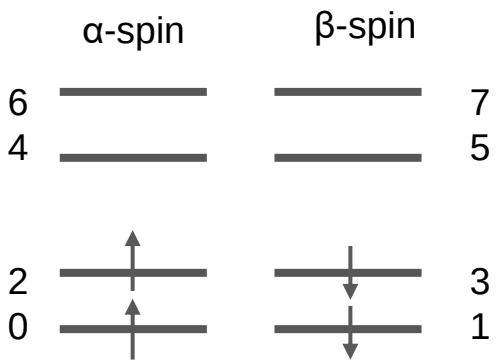


$$[U_1,U_2,U_3,U_4,U_\theta] = \left\{\left[H,H,R_x(-\tfrac{\pi}{2}),H,R_z(\theta/8)\right],\left[R_x(-\tfrac{\pi}{2}),H,R_x(-\tfrac{\pi}{2}),R_x(-\tfrac{\pi}{2}),R_z(\tfrac{\theta}{8})\right],\left[H,R_x(-\tfrac{\pi}{2}),R_x(-\tfrac{\pi}{2}),R_z(\tfrac{\theta}{8})\right],\left[H,H,H,R_x(-\tfrac{\pi}{2}),R_z(\tfrac{\theta}{8})\right],\left[R_x(-\tfrac{\pi}{2}),H,H,H,R_z(-\tfrac{\theta}{8})\right],\left[H,R_x(-\tfrac{\pi}{2}),H,H,R_z(-\tfrac{\theta}{8})\right],\left[R_x(-\tfrac{\pi}{2}),R_x(-\tfrac{\pi}{2}),R_x(-\tfrac{\pi}{2}),H,R_z(-\tfrac{\theta}{8})\right],\left[R_x(-\tfrac{\pi}{2}),R_x(-\tfrac{\pi}{2}),H,R_x(-\tfrac{\pi}{2}),R_z(-\tfrac{\theta}{8})\right]\right\}$$

Figure: Pennylane

Orbitals for H4

- Single Excitation:
  - [0, 4] [0, 6] [2, 4] [2, 6] [1, 5] [1, 7] [3, 5] [3, 7]
  - 8 unique excitation
- Double: - 16 unique excitation
- Can be got from pennylane.qchem.excitations



Other Try

k-Unitary paired-Generalized Coupled Cluster Ansatz (k-UpCCGSD)

- “Generalized”: consider all orbital combinations. Not distinguish occupied or unoccupied orbitals
- “Paired”: only consider paired double excitation: both electrons in the same spatial orbital moved to another spatial orbital.

k=2(36 parameters) is sufficient for H4 to reach chemical accuracy*.

*https://arxiv.org/abs/2103.08505

Measurement optimization: Operator Grouping

- Some observables can be measured at the same sampling circuit
  - Case 1: X_1 and X_2 can be measured at the same time: X_1X_2.(joint operator)
  - Case 2: X_1Y_2 and X_1 can be measured at the same time(Sub operator)

- # of terms to be measured

H4 molecule: 185(before optimization) → 72(after optimized by **meas_optimization**)

Measurement optimization: Weighted Sampling

- Sample noise: Finite statistics
- Weighted Sampling: Allocate more shots to pauli terms with larger coeffcient.
- Hamiltonian: $H = \sum a_i P_i$
- Shots for P_i: $s_i = \frac{|a_i|}{\sum |a_i|} s_{tot}$

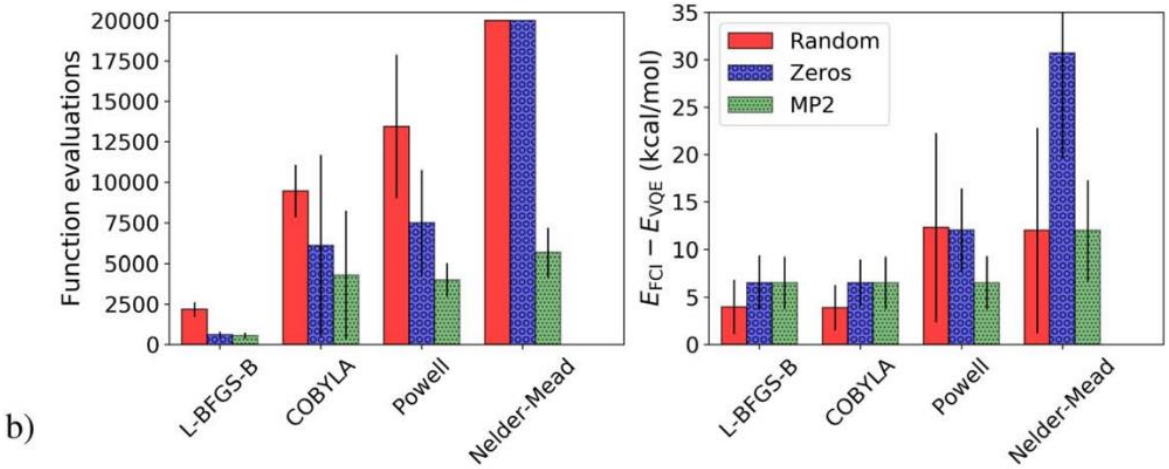
Example: $H = 0.5X_0 + 0.3Y_0 + 0.2Z_0$

The best way to minimize sample variation: 50% shots for X_0, 30% Y_0, 20% for Z_0.

Optimizer - noiseless

L-BFGS-B: Limited-memory Broyden–Fletcher–Goldfarb–Shanno Bound algorithm

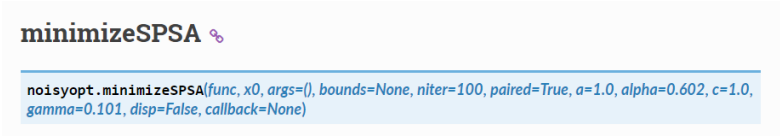
- Overperform COBYLA and Powell.



b)

https://iopscience.iop.org/article/10.1088/2058-9565/aad3e4

Noise-resistant optimizer



If we need to be more frugal:

- Simultaneous perturbation stochastic approximation(SPSA)*

- Random Operator Sampling for Adaptive Learning with Individual Number of shots(Rosalin)**

*https://noisyopt.readthedocs.io/en/latest/api.html#minimizespsa

**https://arxiv.org/abs/2004.06252

Parameter initialization

- MP2 theory

SingleExcitation DoubleExcitation

$$t_i^a = 0; \quad t_{ij}^{ab} = \frac{h_{ijba} - h_{ijab}}{\epsilon_i + \epsilon_j - \epsilon_a - \epsilon_b},$$

h_{ijba} is the two-electron integral for orbital i,j,b,a
ϵ_i is the HF energy for orbital i.

https://iopscience.iop.org/article/10.1088/2058-9565/aad3e4

Other try

- Identity Block

Choose parameter $\boldsymbol{\theta}$, such that the whole starting ansatz is identity operation.

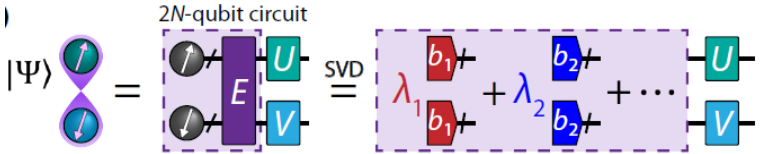
$$U(\boldsymbol{\theta}^{init}) = \prod_{m=M}^{1} U_m(\boldsymbol{\theta}_m) = \prod_{m=M}^{1} \left(\prod_{l=L}^{1} U_l(\theta_{l,1}^m) \prod_{l=1}^{L} U_l(\theta_{l,1}^m)^\dagger \right) = \prod_{m=M}^{1} I_m = I.$$

https://quantum-journal.org/papers/q-2019-12-09-214/pdf/

Using Symmetry

- Qubit tapering(https://arxiv.org/abs/1701.08213)
- Bipartite system(PRX QUANTUM 3, 010309 (2022))

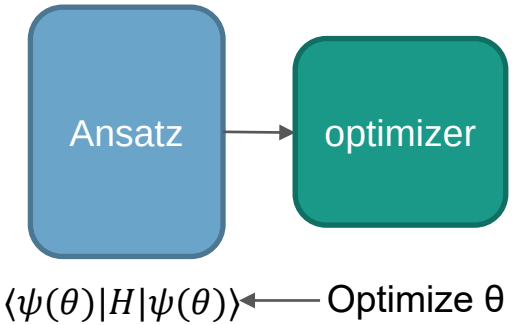
Core idea: smaller system size with more terms to evaluate.

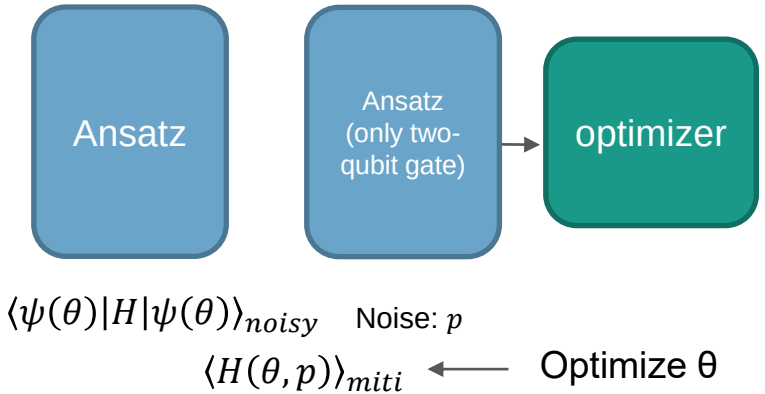


Error mitigation proposal

Mitigation with noise-estimation circuit(PRL 127, 270502 (2021))

- Quantum devices suffer from depolarizing noise
- Two-qubit gates are the main source of depolarizing error
- The original paper is designed for quantum simulation, but the idea should also work for VQE.





Result

- We don’t implement all the ideas mentioned here. However, by UCCSD ansatz, weighted-sampling and measurement optimization, we reached quasi-chemical accuracy(~4mH).
- If shot limit is 3~4 times larger, chemical accuracy can be reached(already tested).

- Error mitigation approachs have not been implemented in code yet. But they can be easily packaged given the access to real noisy device.

I learned a lot during this Challenge.
Many thanks for your attention!