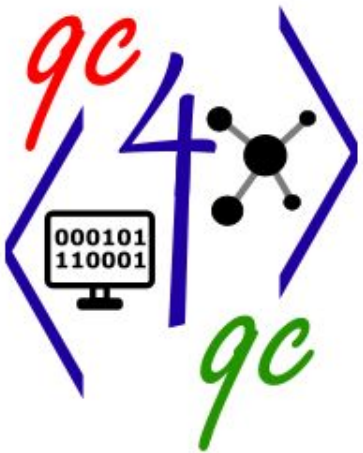
# Refining building blocks in quantum algorithms for electronic structure computations

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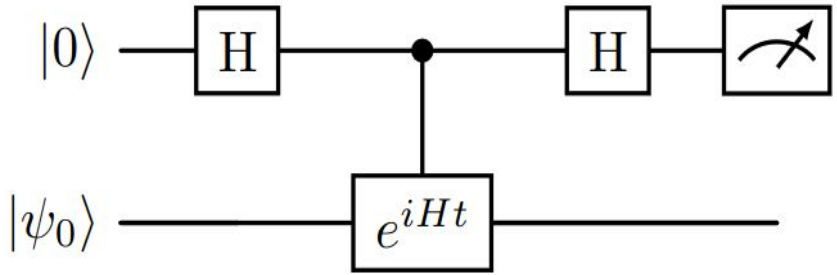
*Advanced Research Department, Qubit Pharmaceuticals*

Qubit PHARMACEUTICALS LCT Laboratoire de Chimie Théorique SORBONNE UNIVERSITÉ

# Introductive quantum circuit




with $H = H^\dagger$ and $H = \sum_{k=0}^{n} E_k \, |\psi_k\rangle \, \langle\psi_k|$

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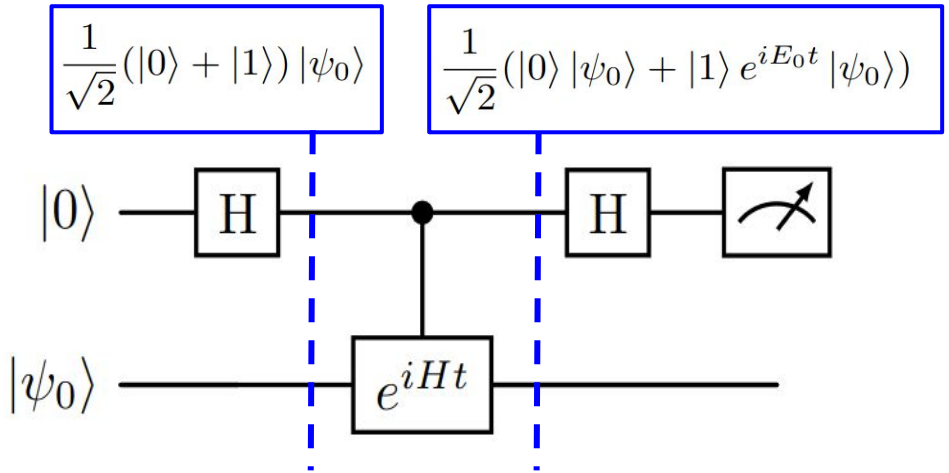


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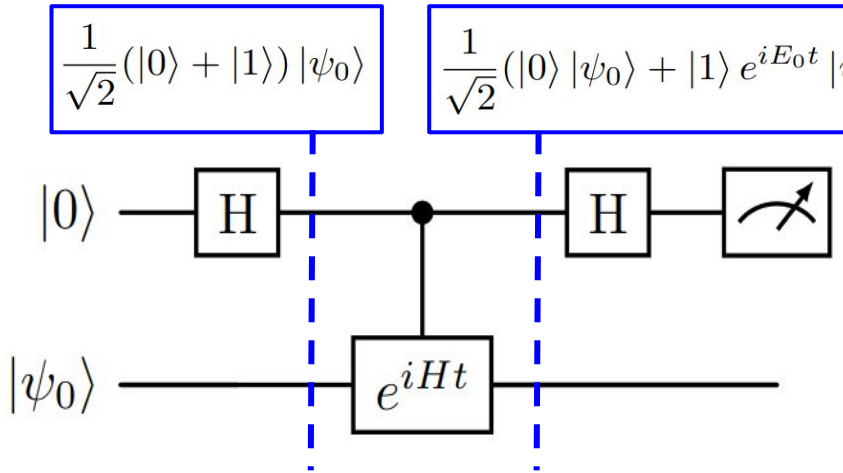


with $H = H^\dagger$ and $H = \sum_{k=0}^{n} E_k |\psi_k\rangle \langle\psi_k|$

The measurement gives an estimation of the phase, here the **ground state energy:**

$$\mathcal{P}_0 = \cos\left(\frac{E_0 t}{2}\right), \quad \mathcal{P}_1 = \sin\left(\frac{E_0 t}{2}\right)$$

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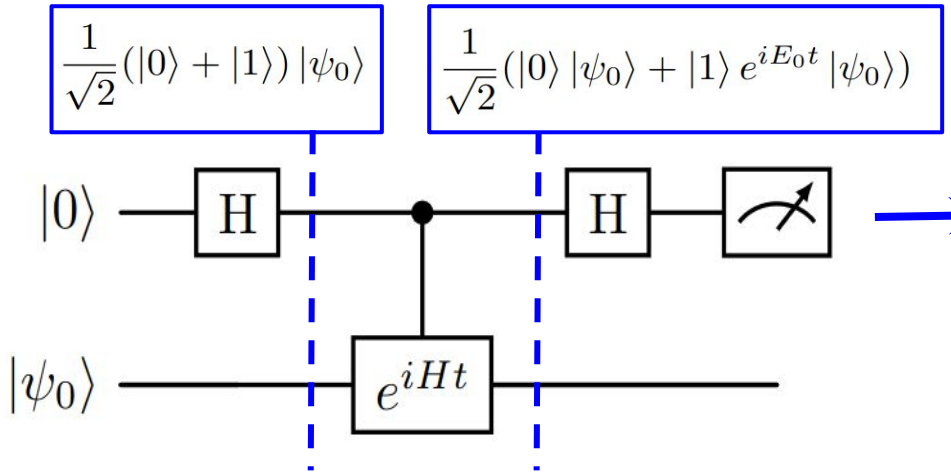


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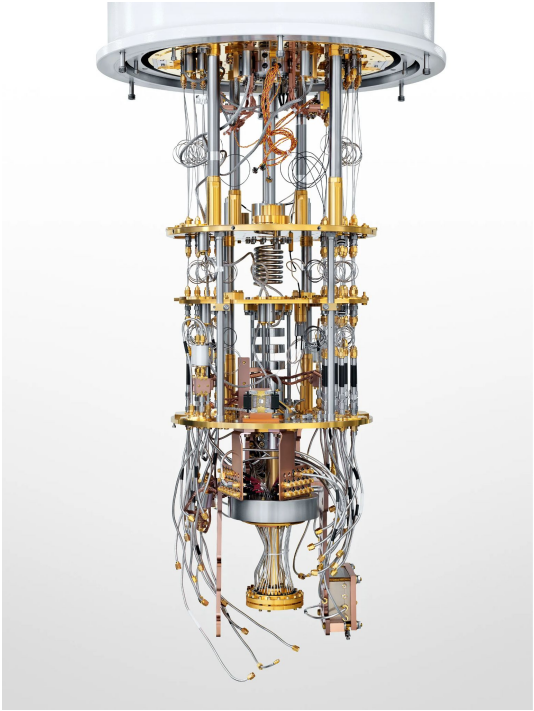
**Two challenges:**

1. Preparing the ground state of *H*
2. Implementing the unitary evolution

# Plan

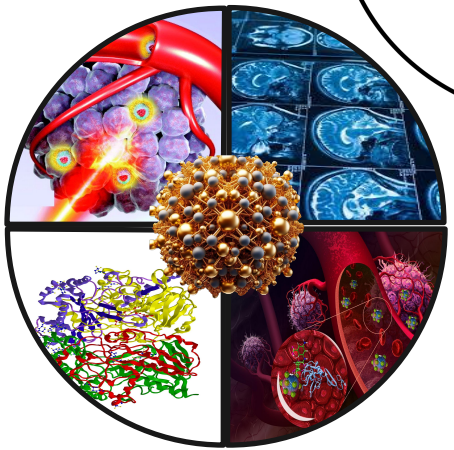


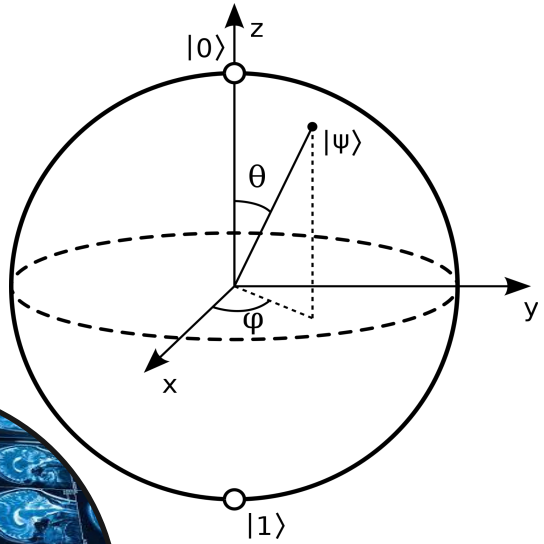
**01** Context - ***Quantum algorithm & chemistry***

**02** Overlap-ADAPT-VQE - ***Variational state preparation***

**03** Oracle decomposition – ***Multi-control-NOT gate***

**04** Conclusions and Perspectives






# 01

# Context -

*Quantum algorithms for quantum chemistry*

# Mapping chemistry to qubits - *2nd quantisation & Jordan-Wigner*

**Electronic hamiltonian**

$$H = -\sum_i \frac{\nabla_{r_i}^2}{2} - \sum_{i,j} \frac{Z_i}{|R_i - r_j|} + \sum_{i,j>i} \frac{Z_i Z_j}{|R_i - R_j|} + \sum_{i,j>i} \frac{1}{|r_i - r_j|}$$

**Hamiltonian discretisation : 2nd quantisation**

$$H := \sum_{p,q} h_{pq} a_p^\dagger a_q + \sum_{p,q,r,s} h_{pqrs} a_p^\dagger a_r^\dagger a_s a_q$$

**Wavefunctions antisymmetric under particle exchange = fermionic operators satisfy anticommutation relations.**

$$\{a_p, a_q^\dagger\} := a_p a_q^\dagger + a_q^\dagger a_p = \delta_{pq} \quad \text{and} \quad \{a_p, a_q\} := a_p a_q + a_q a_p = 0$$

# Mapping chemistry to qubits - *Occupation basis & Jordan-Wigner*

Mapping *fermions to qubits* & *excitation operators to quantum circuits*

Occupation basis : each qubit stores the *occupation* of a spin orbital

$$\text{Spin-orbital}: \text{ unoccupied} \rightarrow |0\rangle \text{ , occupied} \rightarrow |1\rangle$$

Hartree-Fock state $$|\Psi_{\text{HF}}\rangle = |1\rangle^m \otimes |0\rangle^{n-m}$$

Jordan-Wigner tranform

$$a_p = \left( \bigotimes_{i=0}^{p-1} Z_i \right) \otimes \frac{X_p + iY_p}{2}$$

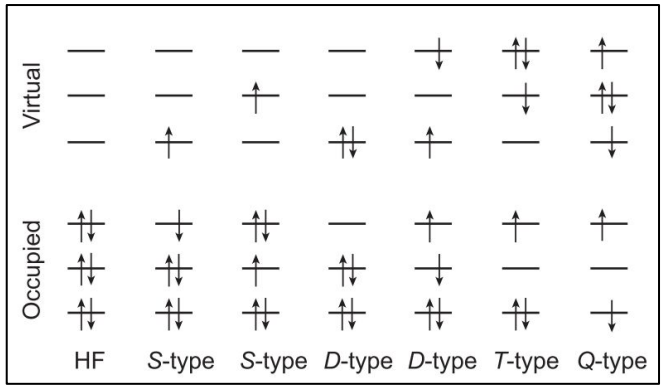
$$a_p |0\rangle_p = 0 \qquad a_p^\dagger |0\rangle_p = |1\rangle_p$$

$$a_p |1\rangle_p = |0\rangle_p \qquad a_p^\dagger |1\rangle_p = 0$$

# Quantum chemistry : electronic wavefunction

## Full-Configuration-Interaction (FCI) wavefunction

$$\Psi_{\mathrm{FCI}} = c_{\mathrm{HF}}\Psi_{\mathrm{HF}} + \sum_{ar} c_a^r \Phi_a^r + \sum_{a<b,r<s} c_{ab}^{rs}\Phi_{ab}^{rs} + \sum_{a<b<c,r<s<t} c_{abc}^{rst}\Phi_{abc}^{rst} + \dots$$




⚠️ Number of Configuration State Functions grow **exponentially** with system size

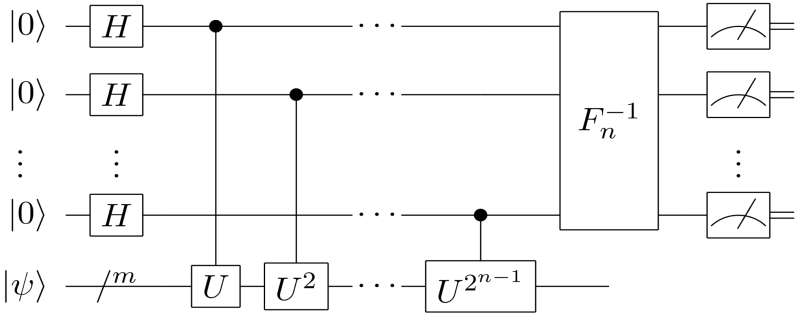
💡 **Quantum computer** stores the **exponentially** increasing wave-function with a **linear number of qubits**

# Quantum algorithms for quantum chemistry

## The Quantum Phase Estimation algorithm

1. Initialisation : $|\psi\rangle = \sum_i a_i |\psi_i\rangle$
2. Projection : ctrl-exp( *i* Ht) and QFT $^+$
3. Final state : $\sum_i \alpha_i |\mathrm{bin}(\varphi_i)\rangle |\psi_i\rangle$

Where $|\mathrm{bin}(\varphi_i)\rangle$ is a binary estimate of the *i*-th eigenvalue




The original QPE algorithm

→ Ground state projection with success probability = $|\alpha_0|^2$

→ Algorithm complexity dominated by ctrl-exp( *i* Ht)

polynomial *assuming efficient initialisation,* i. e. exponential advantage over exact diagonalisation

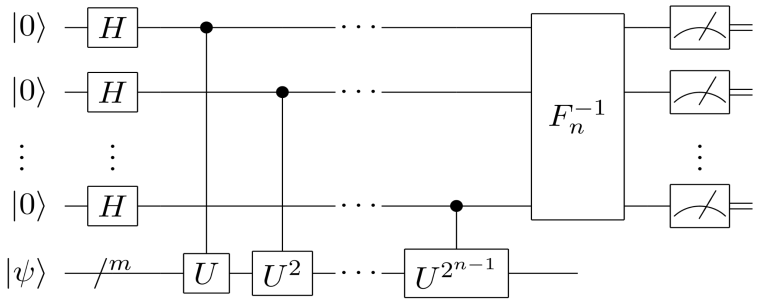
# Quantum algorithms for quantum chemistry

The initial state, spectral decomposition:

$$|\psi\rangle = \sum_{k=0}^{n} \alpha_k |\psi_k\rangle$$

Success probability for ground state:

$$|\langle\psi|\psi_0\rangle|^2 = |a_0|^2$$



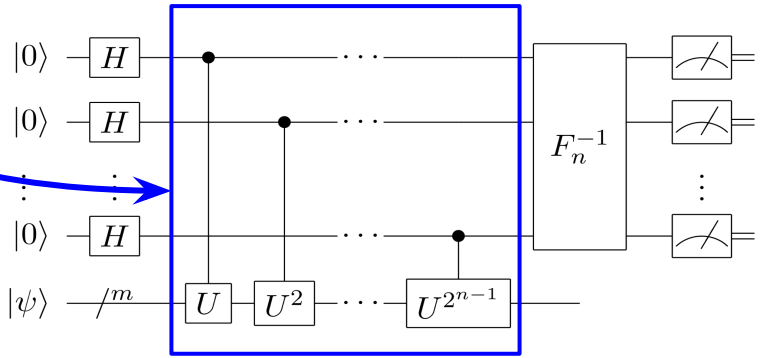

**The initial state should be :**

1. **A good approximation of the Hamiltonian ground state**
2. **Implemented in the quantum device with a compact circuit**

# Quantum algorithms for quantum chemistry

**Multiple Hamiltonian evolution operator**

$\text{ctrl} - e^{iH2^n}$




- The Hamiltonian evolution operator should be efficiently implemented, as it **dominates the overall algorithm complexity**
- Brute force Jordan-Wigner and first order Trotter : the operation can be implemented with a depth of **O(N$^4$)**

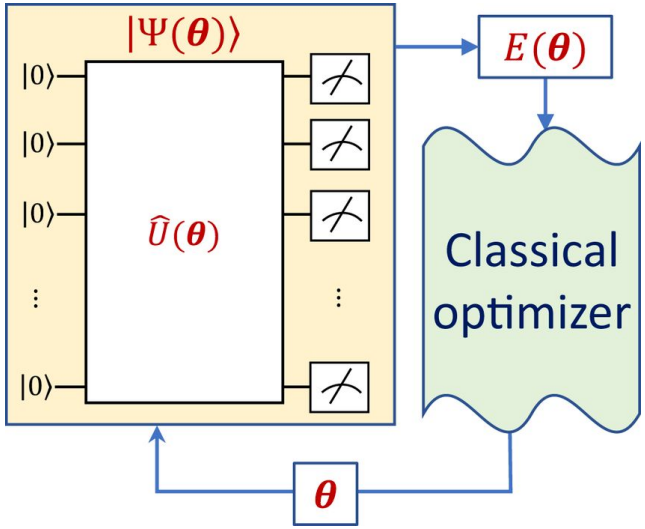
# Quantum algorithms for quantum chemistry - VQE

VQE relies on the variat $\langle\psi(\theta)|H|\psi(\theta)\rangle \geq E_0$

$|\psi(\theta)\rangle$ is a parameterised quantum state (ansatz)
$E_0$ is the lowest eigenvalue: the ground-state energy.

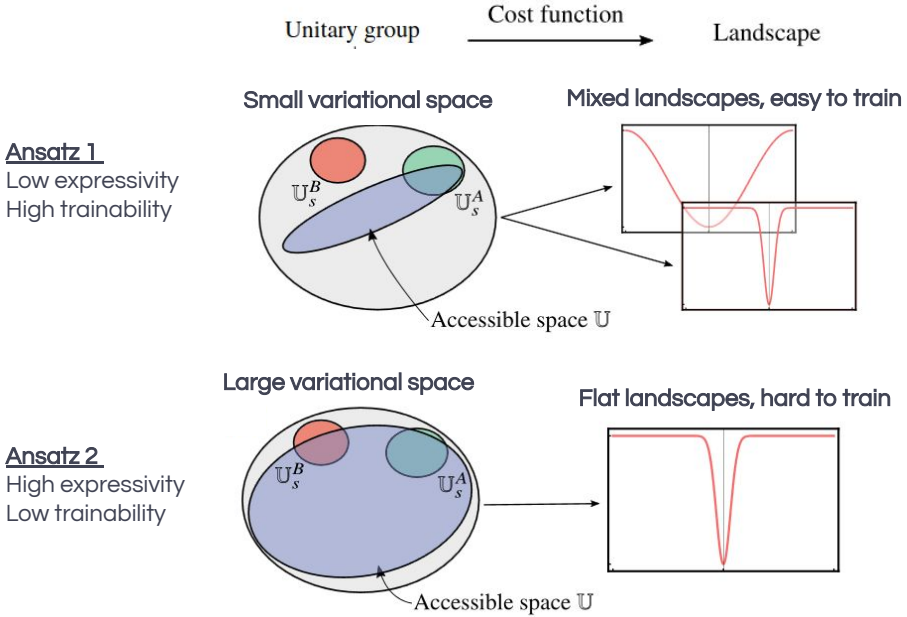
This implies we can approximate the ground-state wavefunction and its energy by computing:

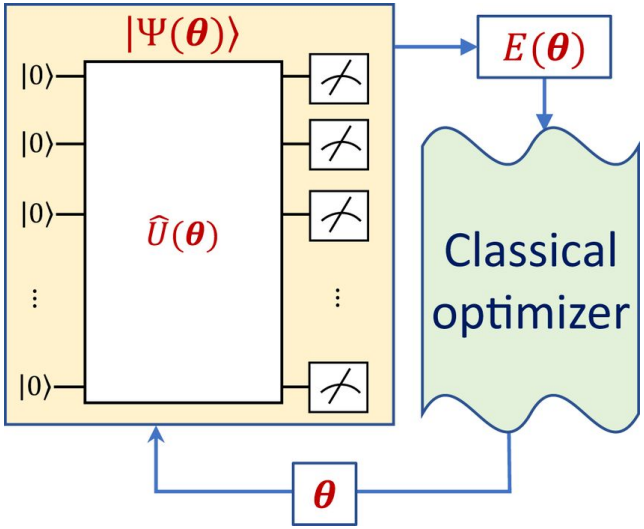
$$\min_{\theta} \langle\psi(\theta)|H|\psi(\theta)\rangle$$



# Quantum algorithms for quantum chemistry - VQE

**Challenge: Ansatz trainability, expressivity and circuit depth**



# Quantum algorithms for quantum chemistry - VQE

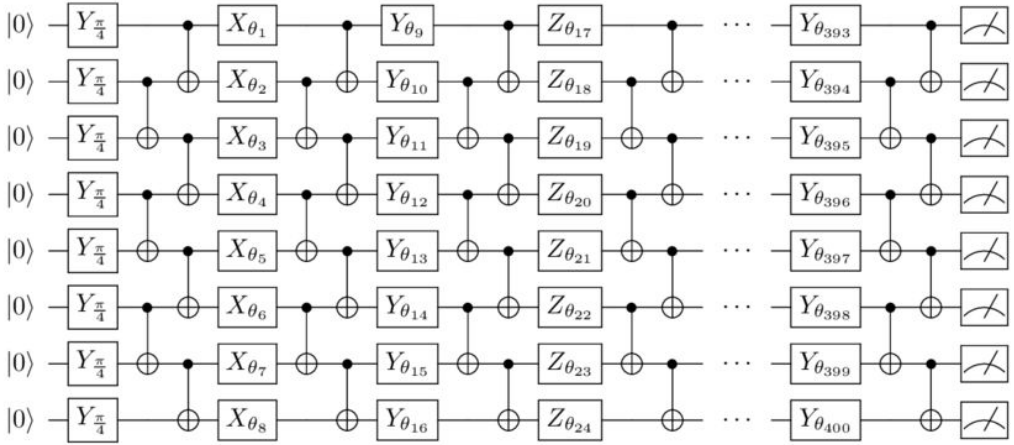
**Limitations** :

- Noisy optimisation of a multi-dimensional non-convex cost function, with limited energy evaluations.
- Convergence to very accurate energy (chemical accuracy) is uncertain, it needs an ansatz of **(i) high trainability, (ii) high expressivity, and (iii) a compact quantum circuit.**
- Quantum resource estimates are pessimistic about having an advantage with VQE in chemistry.

**But**:

- Quantum state preparation for QPE is less critical, as it only needs to prepare a state having a **non-zero overlap** with the ground state.
- VQE could provide an accurate parameterised quantum state as initial state for QPE

# Quantum algorithms for quantum chemistry - VQE



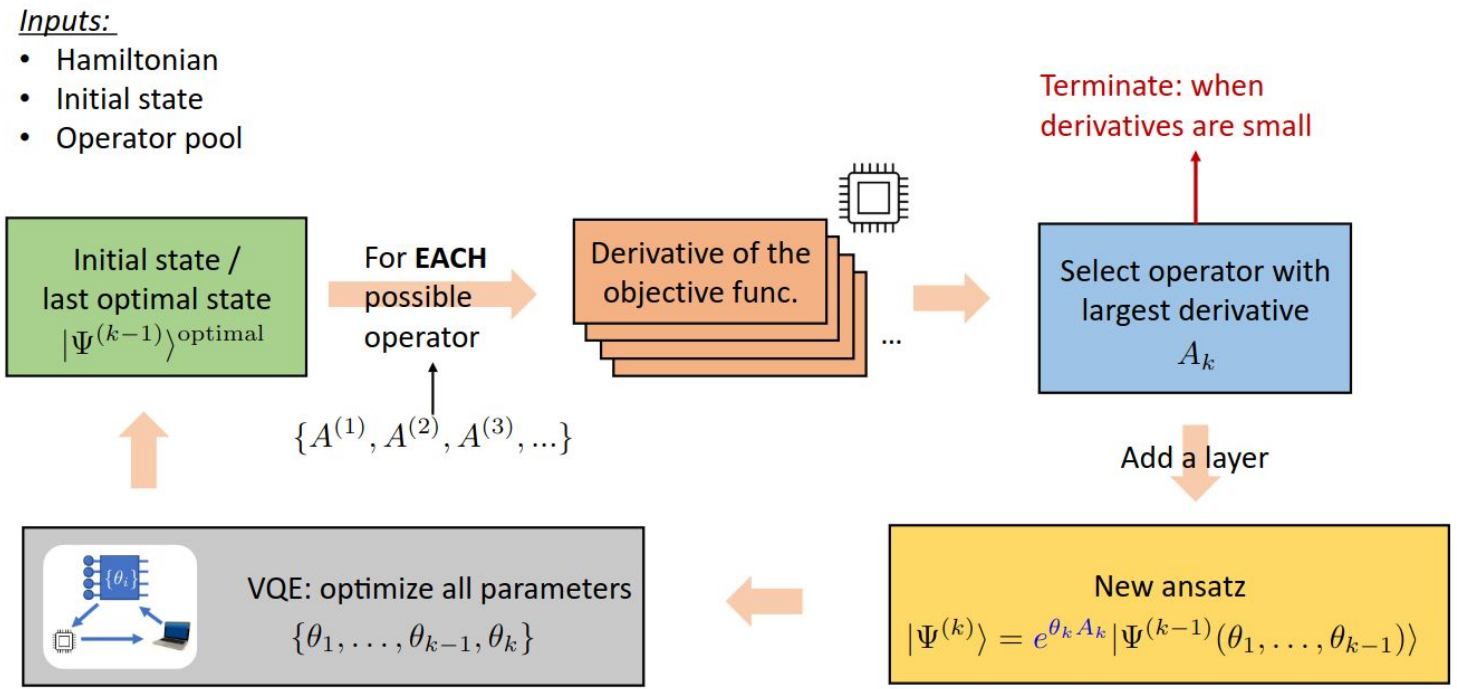

Typical Hardware-efficient ansatz : trainability issues

# Quantum algorithms for quantum chemistry

## ADAPT-VQE : compact and accurate ansatze

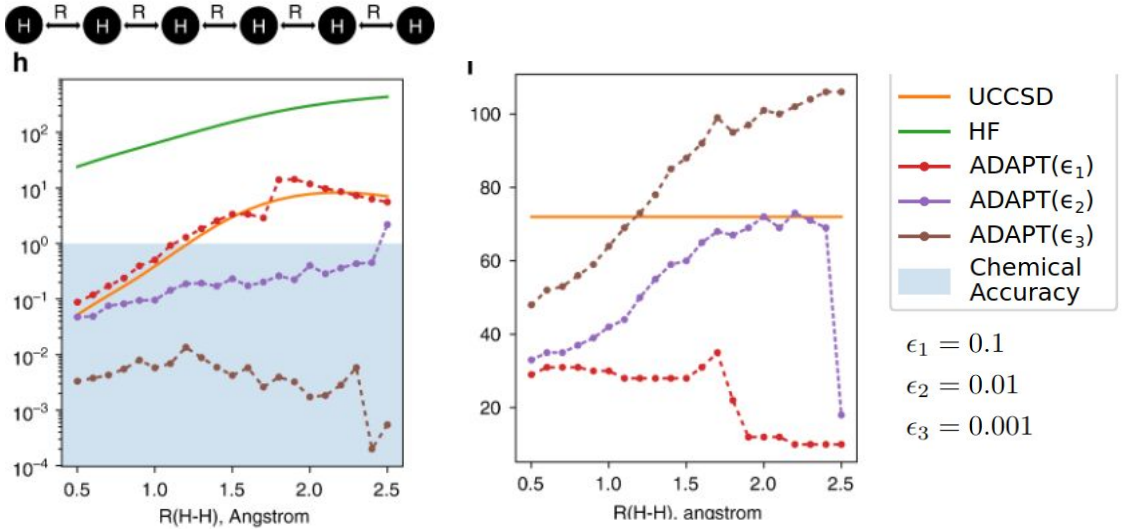
Grimsley, Economou, Barnes, Mayhall, Nature Communications 10, 3007 (2019)

Slide taken from talk : UCLA, Sophia Economou, Adaptive quantum simulation algorithms



# Quantum algorithms for quantum chemistry

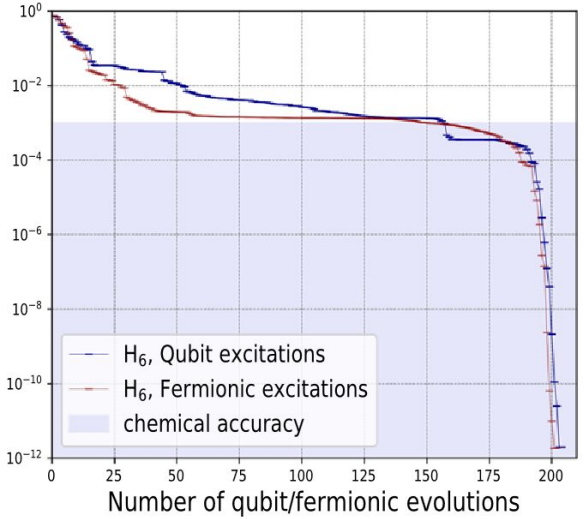
## ADAPT-VQE : compact and accurate ansatze



# Quantum algorithms for quantum chemistry

**But sometimes face energy plateaus issues**

Local minima & barren plateaus



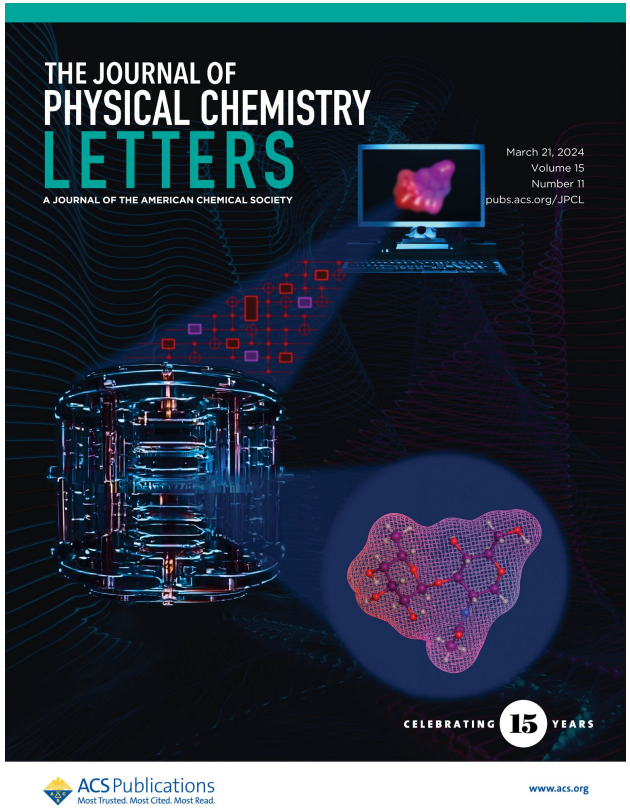

Unwise addition of operators is a problem:

- **Overparameterised** ansatz
- Increase the **circuit depth**
- No energy improvements

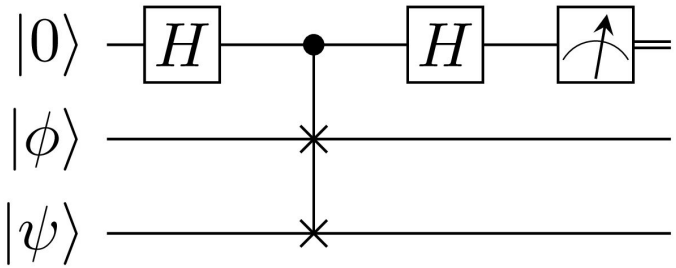
# 02

# Overlap-ADAPT-VQE

*Variational state preparation*

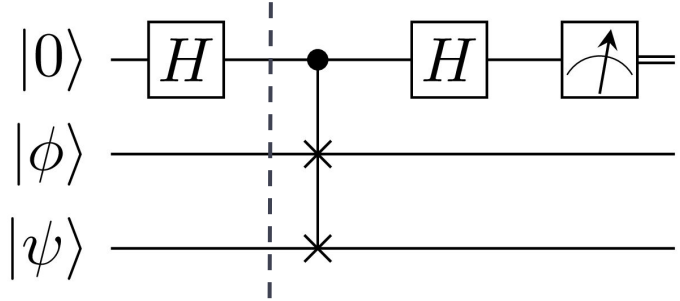


# Introductive quantum circuit



# Introductive quantum circuit

$$\frac{1}{\sqrt{2}}(|0,\phi,\psi\rangle + |1,\phi,\psi\rangle)$$



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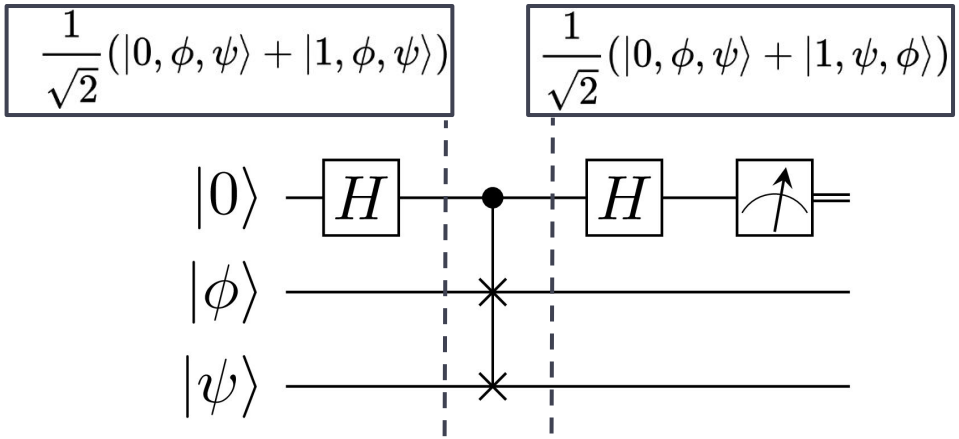

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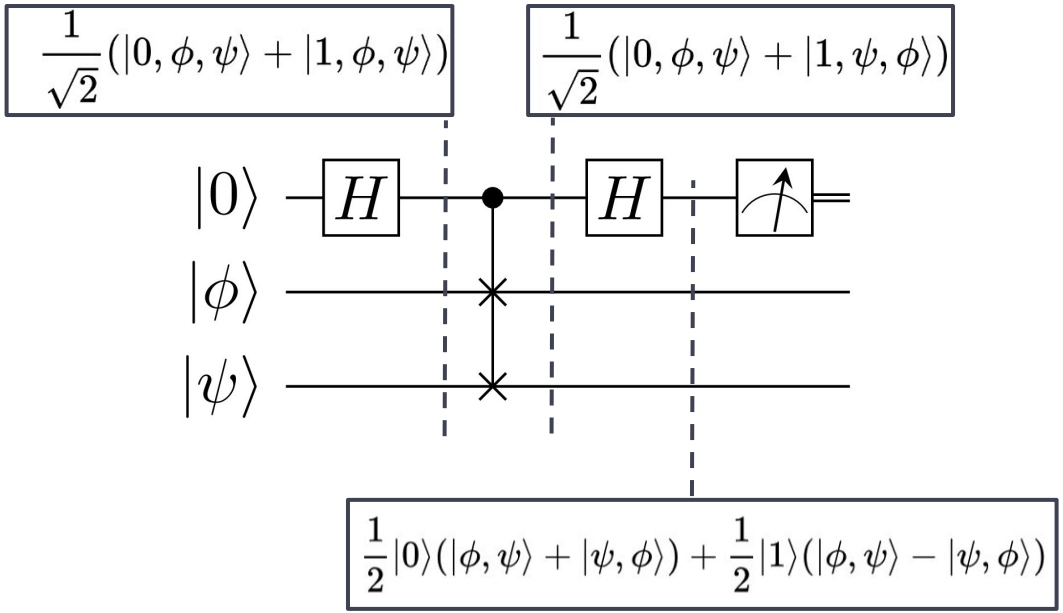

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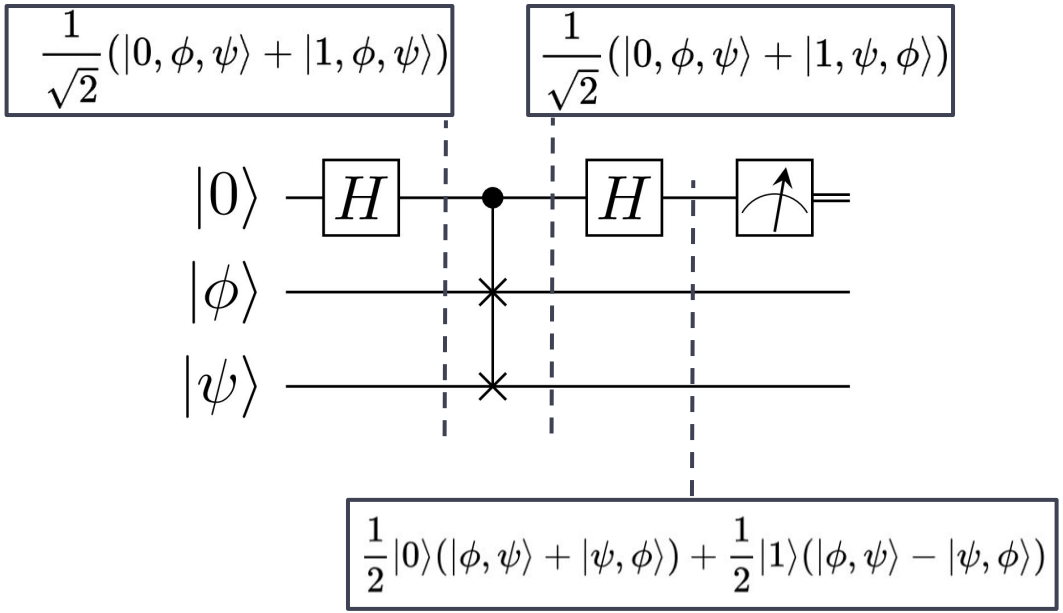



$$P(\text{First qubit} = 0) = \frac{1}{2}\Big(\langle\phi|\langle\psi| + \langle\psi|\langle\phi|\Big)\frac{1}{2}\Big(|\phi\rangle|\psi\rangle + |\psi\rangle|\phi\rangle\Big) = \frac{1}{2} + \frac{1}{2}|\langle\psi|\phi\rangle|^2$$

# Overlap-ADAPT-VQE

Feniou, Hassan, Giner, Maday, Piquemal, Communications Physics 6, 192 (2023)

Grows the ansatz by maximizing its overlap with any intermediate target wave-function that captures some electronic correlation.

Inputs :

1. Target/reference wave-function
2. Pool of operators
3. Initial state

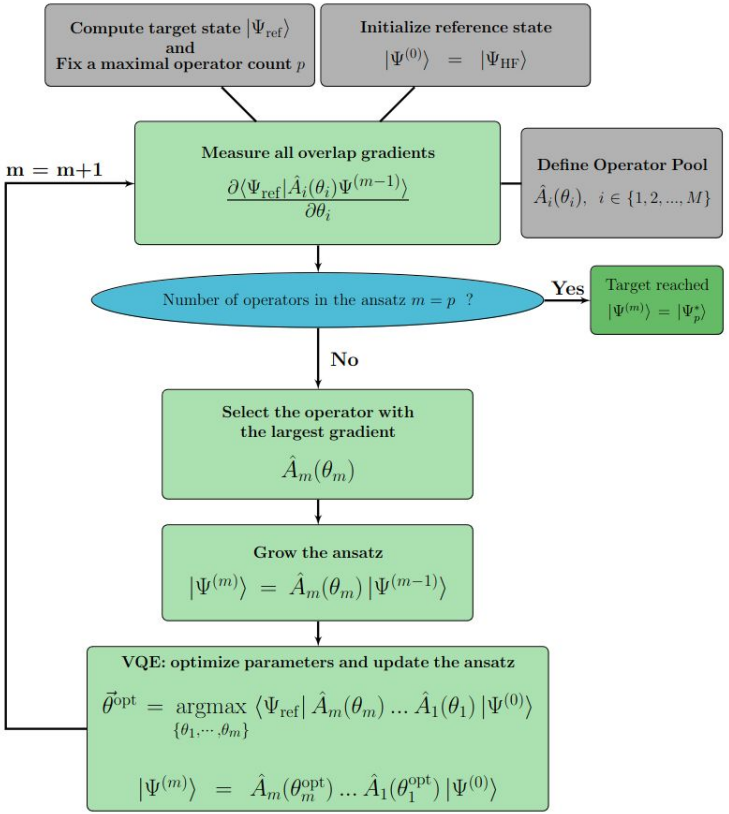
Procedure :

ADAPT-VQE with a projector on the target state as hamiltonian:

$$H = |\Psi\rangle_{\text{ref}} \langle\Psi|_{\text{ref}}$$

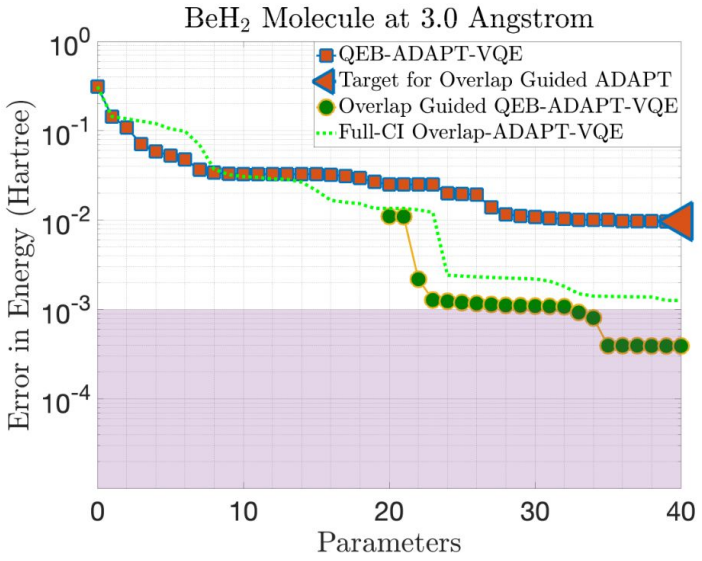
# Overlap-ADAPT-VQE

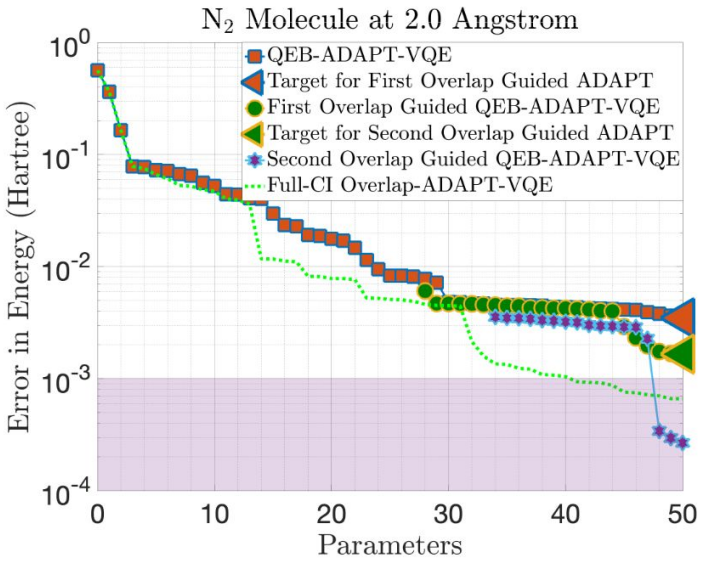
Feniou, Hassan, Giner, Maday, Piquemal, Communications Physics 6, 192 (2023)



# Overlap-ADAPT-VQE

Example target wavefunction : ADAPT-VQE ansatz stuck in an energy plateau

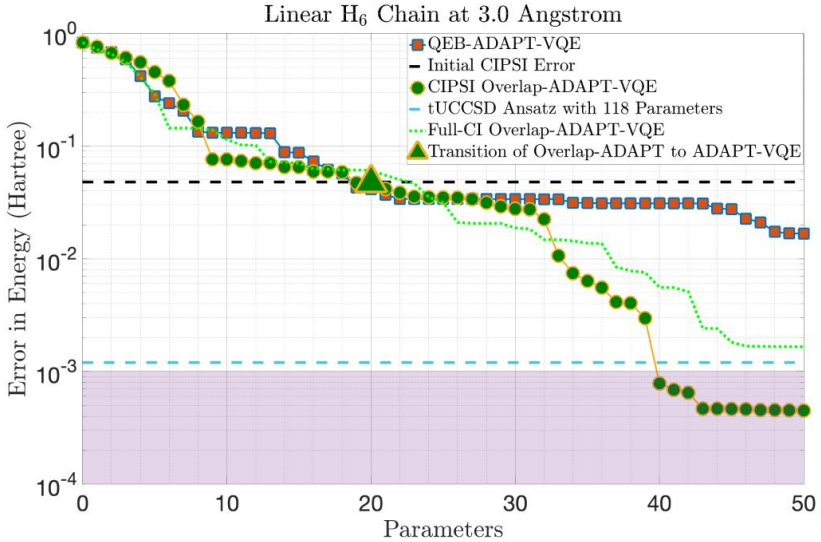


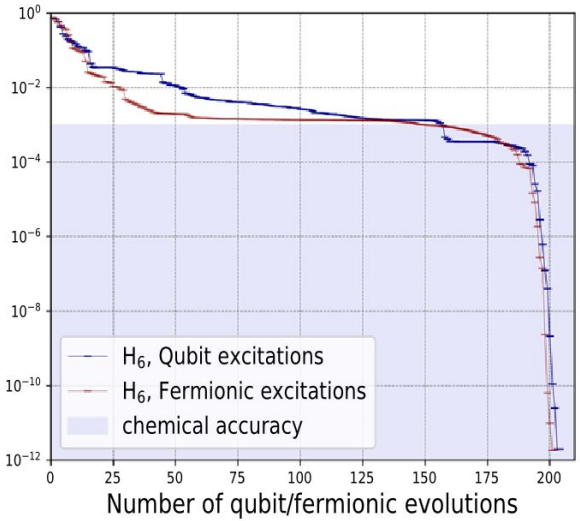




The Overlap-ADAPT-VQE ansatz achieves **higher accuracy** and/or **shorter circuits**

# Overlap-ADAPT-VQE

Example target wavefunction : Classical CIPSI wave-function




Initialising the ansatz with a CIPSI-Overlap-ADAPT-VQE procedure drives the ADAPT-VQE to chemical accuracy with **40 parameters** instead of **over 150** .

# Overlap-ADAPT-VQE : QPE initialisation

Feniou, C., Adjoua, O., Claudon, B., Zylberman, J., Giner, E., & Piquemal, J. P. (2024). Sparse quantum state preparation for strongly correlated systems. *The Journal of Physical Chemistry Letters*, *15*(11), 3197-3205.

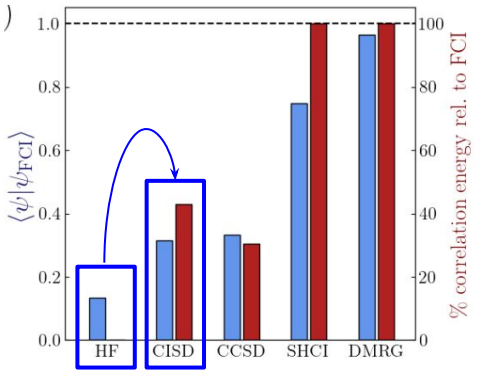
📝 The **overlap** between **initial state** and **ground state** drives the **success** probability in QPE.

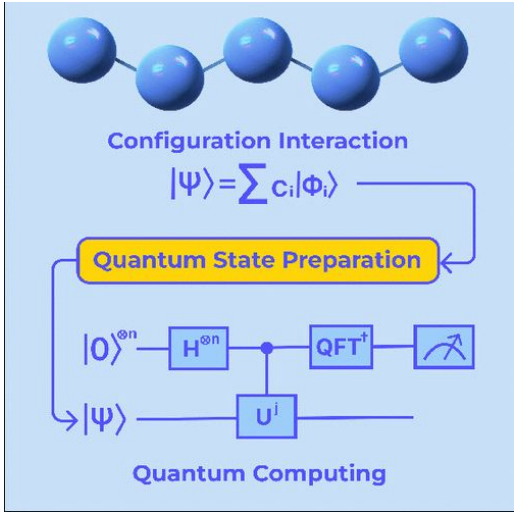
⚠️ For **strongly correlated systems** , the overlap between **Hartree-Fock** state and ground state **vanishes** (orthogonality catastrophe)

💡 Taking **classically-derived** correlated wavefunction as **initial states** would enhance the ground state support

Strategy: Quantum state preparation of CI wave-function

Fomichev, Stepan, et al. *arXiv:2310.18410* (2023).



# Overlap-ADAPT-VQE : QPE initialisation

**Selected-CI wave function, is mapped as a sparse quantum state in the computational basis (particle-, spin-symmetries).**

$$|\Psi_{\mathrm{SCI}}\rangle = \sum_{i \in \mathcal{V}} c_i \, |\phi_i\rangle = c_0 \, |11110000\rangle + c_1 \, |11100100\rangle + \ldots + c_n \, |00001111\rangle$$

# Overlap-ADAPT-VQE : QPE initialisation

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**Quantum State Preparation can be achieved by direct loading of expansion coefficients at the corresponding determinants/computational basis vectors, or with a variational algorithm.**

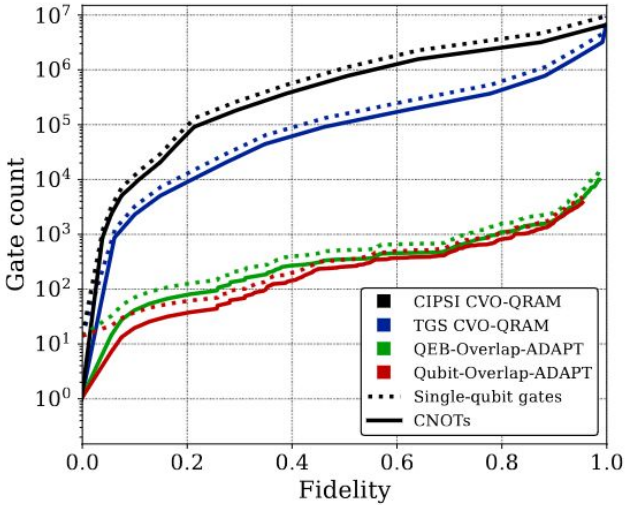
**Direct loading** : loading *M* determinants in a *n*-qubit space has a circuit depth complexity in with CVO-QRAM*

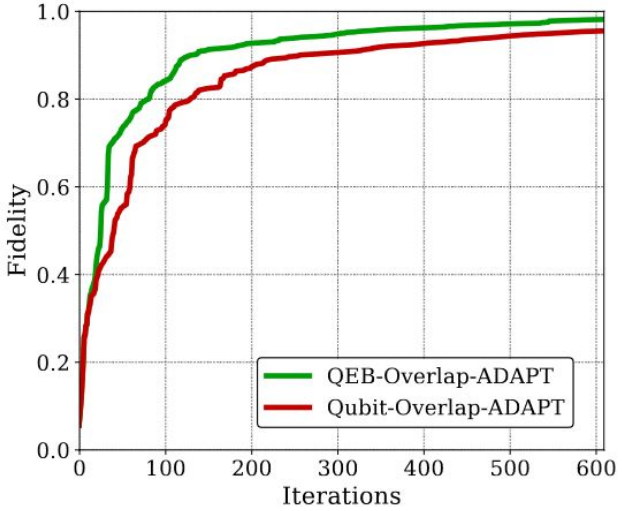
**Variational algorithm** : approaching the target state with parameterised unitary rotations, with Overlap-ADAPT-VQE, undeterministic success

*de Veras, T. M. L.; da Silva, L. D.; da Silva, A. J. Double sparse quantum state preparation. Quantum Information Processing 2022, 21, 204

# Overlap-ADAPT-VQE : QPE initialisation

Feniou, C., Adjoua, O., Claudon, B., Zylberman, J., Giner, E., & Piquemal, J. P. (2024). Sparse quantum state preparation for strongly correlated systems. *The Journal of Physical Chemistry Letters*, *15*(11), 3197-3205.




- 28-qubit simulation on GPU-accelerated emulator
- Overlap-ADAPT-VQE yields much smaller circuits than CVO-QRAM for any target fidelity
- With ~100 iterations, it reaches the fidelity of a 10k determinants CI wavefunction

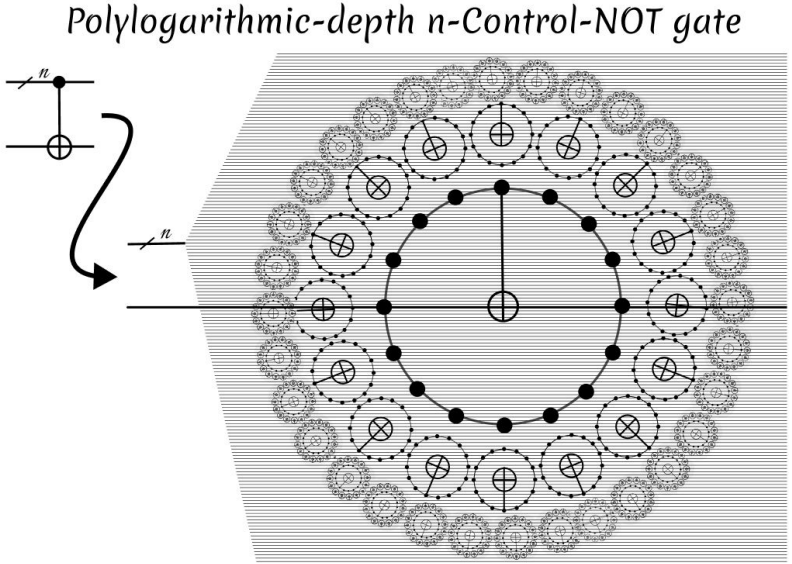
# Overlap-ADAPT-VQE : QPE initialisation

What can we conclude from this?

- It is likely **large CI wave-functions could be encoded as compact Ansatze** in the quantum device, the **Overlap-ADAPT-VQE algorithm can achieve** such encoding
- However, it here needs to classically simulate the whole quantum circuit: **NOT SCALABLE**

How to scale this process to the **100-qubit regime and beyond** ?

- **Approximate** quantum circuit simulation with MPS
- **Loading CI wavefunction** with CVO-QRAM, then **compacting with Overlap-ADAPT-VQE**




# 03

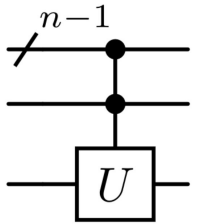
# Oracle decomposition

***Multi-control NOT gate***

# Refining oracle decomposition as quantum circuit

- All FTQC quantum algorithms rely on oracles (Black-box unitary operations)
- The algorithm complexity is expressed as ‘number of calls to the oracle’
- Can these black-box oracles be *efficiently* decomposed into usual quantum gates?


Typical QPE oracles for Hamiltonian evolution operator



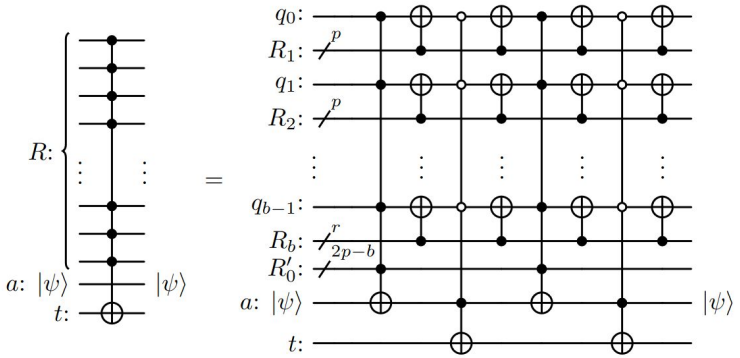

Multi-control-*U gate* used in CVO-QRAM

# Refining oracle decomposition as quantum circuit

## Polylog-depth decomposition of multi-control NOT gates

Claudon, B., Zylberman, J., Feniou, C. *et al.* Polylogarithmic-depth controlled-NOT gates without ancilla qubits. *Nat Commun* 15, 5886 (2024).

- Multi-control operations are building blocks of **countless quantum algorithms**
- State-of-the-art decompositions scale **linearly with the number of controls**
- We proposed a **polylogarithmic decomposition**




| Method | Circuit depth | Borrowed |
|---|---|---|
| Barenco 1 [15] | $48n - 148$ | 1 |
| Barenco $n-2$ [15] | $24n - 43$ | $n-2$ |
| Proposition 1 | $43\log(n)^3 - 1287$ | 1 |

TABLE II: Circuit depth of borrowed ancillae methods. The depths are numerically fitted in the range from $10^2$ to $10^7$ control qubits.

# Refining oracle decomposition as quantum circuit

Parallelised Cp(X) gates => Depth $O(\sqrt{n})$

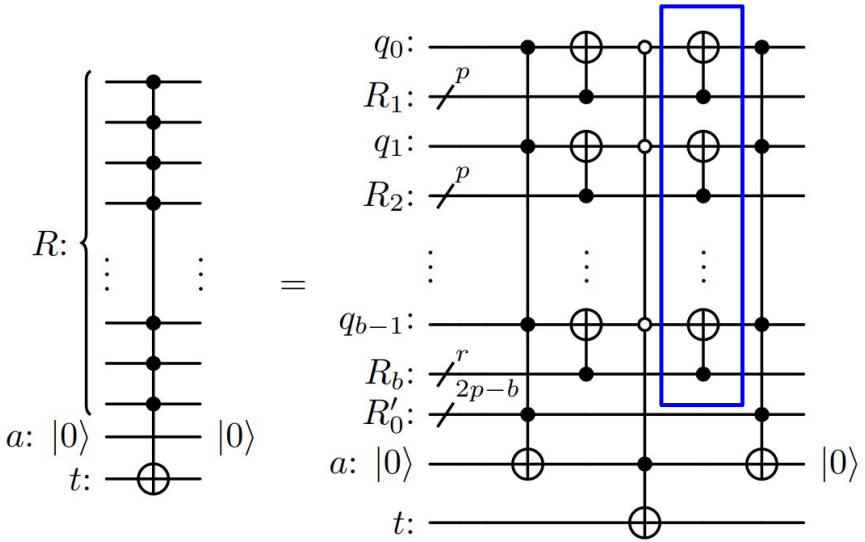



FIG. 1: $n$ controlled $\mathcal{C}_R^t$ using the zeroed ancilla $a$, where $p = \lfloor \sqrt{n} \rfloor$ and $R_i$ is a register of at most $p$ qubits

# Refining oracle decomposition as quantum circuit

Parallelised Cp(X) gates => Depth $O(\sqrt{n})$

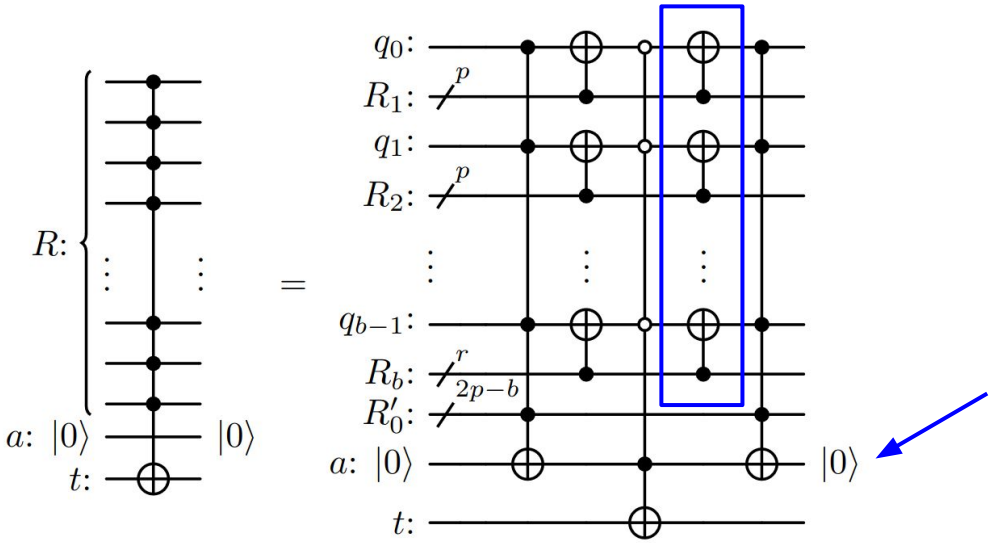



FIG. 1: $n$ controlled $\mathcal{C}_R^t$ using the zeroed ancilla $a$, where $p = \lfloor \sqrt{n} \rfloor$ and $R_i$ is a register of at most $p$ qubits

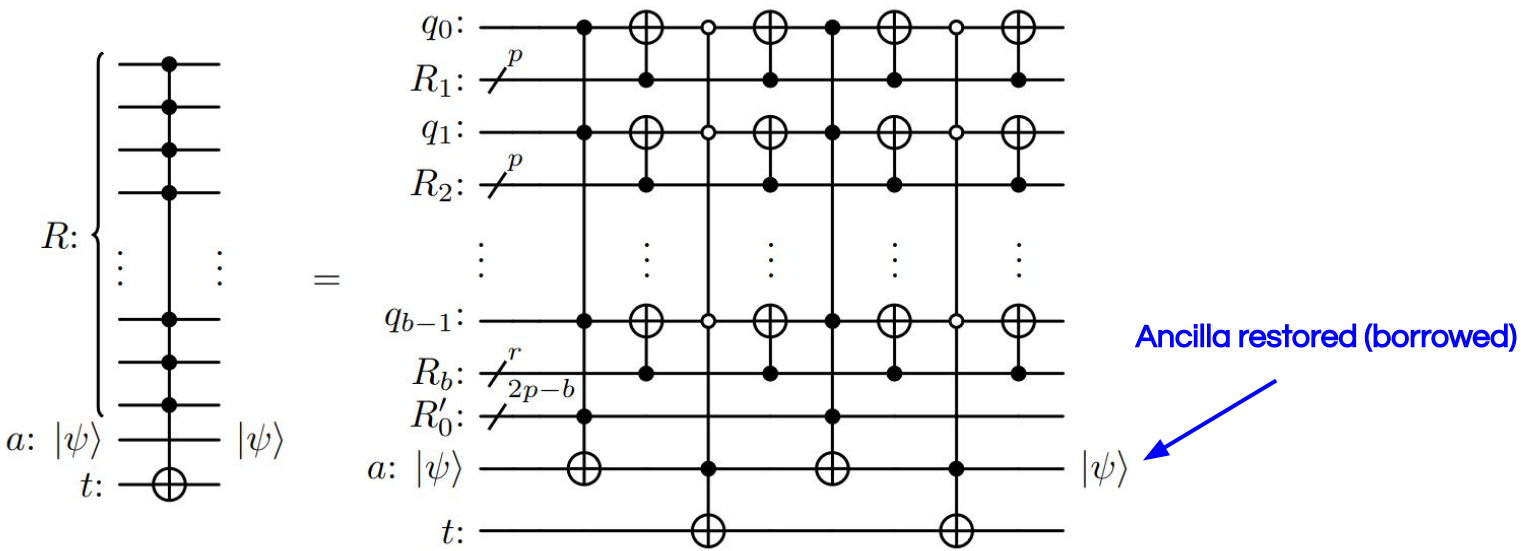
**5 layers of depth** $O(\sqrt{n})$ involving $C_{\sqrt{n}}(X)$ gates (already a quadratic advantage)

Each layer **could be cut down**

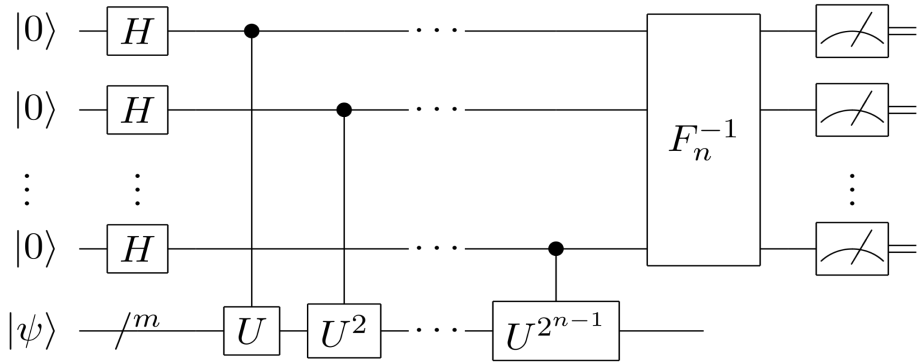
But here need zeroed ancilla qubits...

# Refining oracle decomposition as quantum circuit

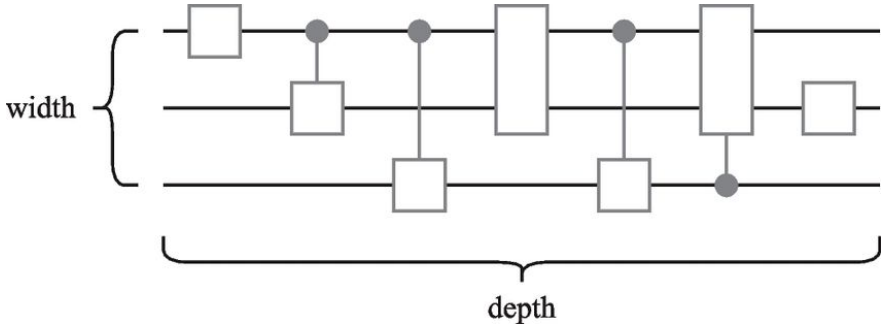
Add a few layers to transform the *zeroed*-ancilla to a *borrowed*, then divide-and-conquer as there are enough *borrowable* ancilla qubits on each Cp(X) layer => circuit depth $\Theta\left(\log(n)^3\right)$



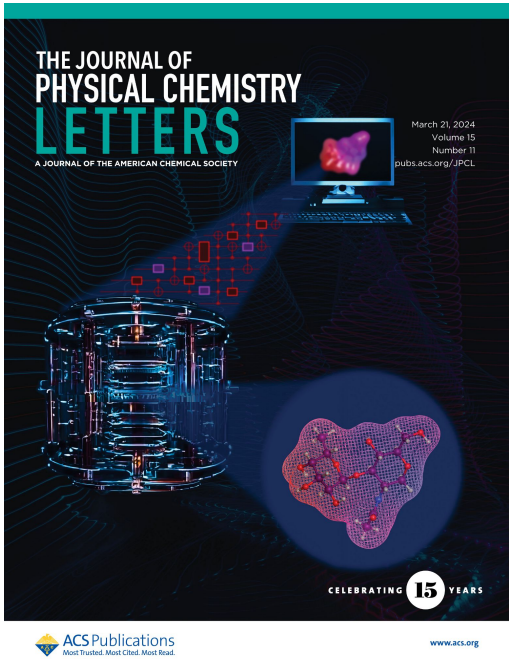
# Conclusion and perspectives

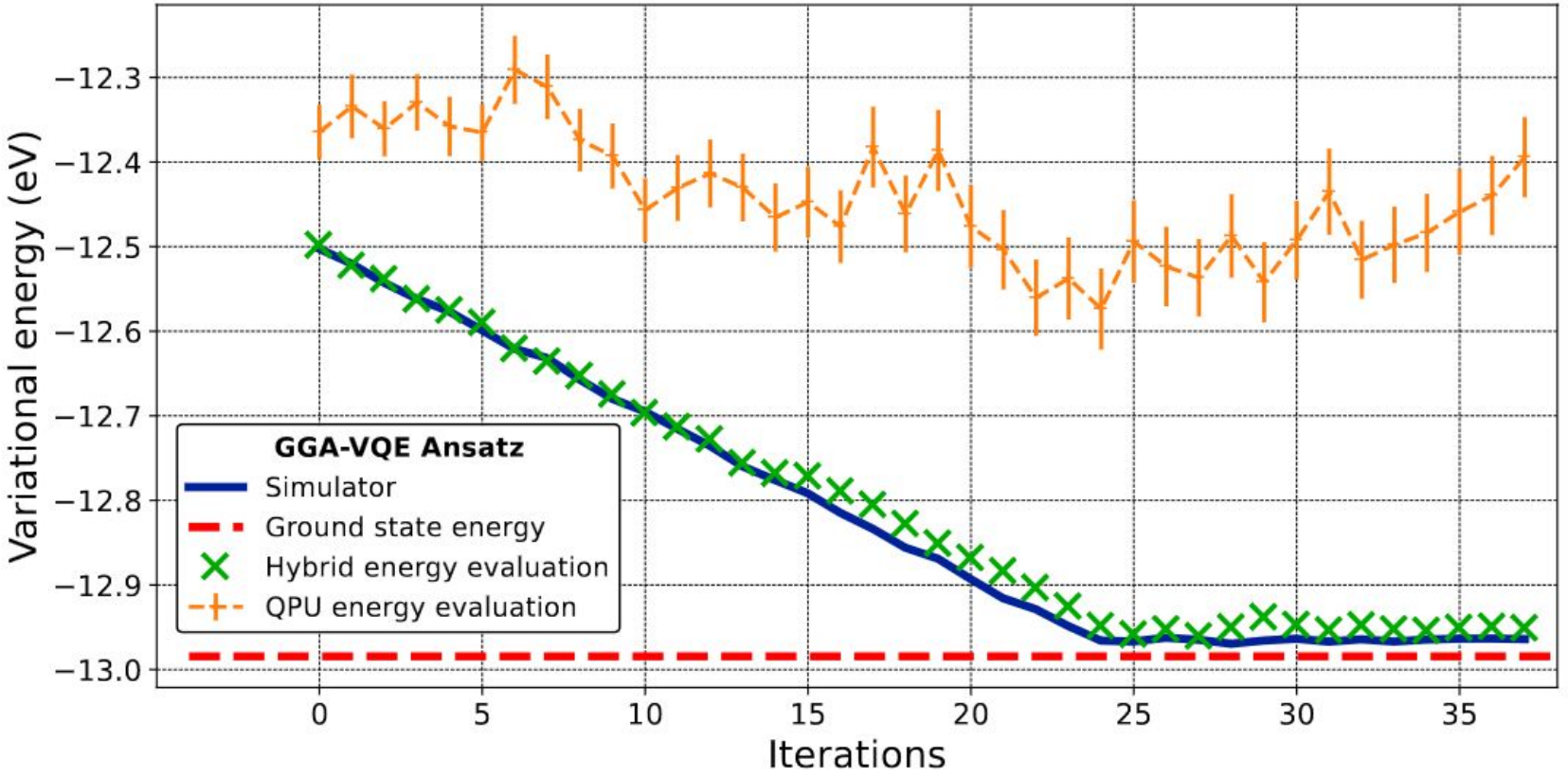



**The original QPE algorithm**


width
depth

# Quantum computational chemistry




Variational energy (eV)
−12.3
−12.4
−12.5
−12.6
−12.7
−12.8
−12.9
−13.0
0
5
10
15
20
25
30
35
Iterations
GGA-VQE Ansatz
Simulator
Ground state energy
Hybrid energy evaluation
QPU energy evaluation