Quantum chemistry on near-term devices

Anton Nykänen, Walter Talarico, Roberto Di Remigio Eikås I QAS2023, Göteborg I 27.10.2023

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Bringing quantum to life

Our mission is to revolutionise life sciences by exploiting the potential of quantum computing to solve currently inaccessible problems

Quantum Network Medicine

Algorithmiq's multiscale approach to tackle the complexity of the cell biology

1 Classical & Quantum - inspired *(proprietary)*

Data-driven identification of the relevant network biology

Hazardous targets
(Leading to toxicity if targeted)

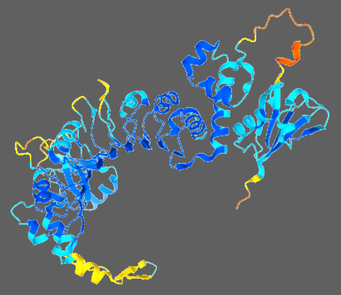
Safe Targets
(With therapeutic effects)

aurora
Network Medicine

2 Classical

Modelling protein structures

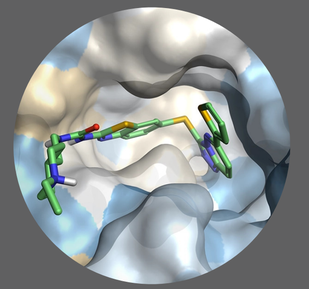
machine learning and crystallographic data



3 Quantum *(proprietary)*

Ab-initio molecular simulations

Quantum computer simulation



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Chemistry

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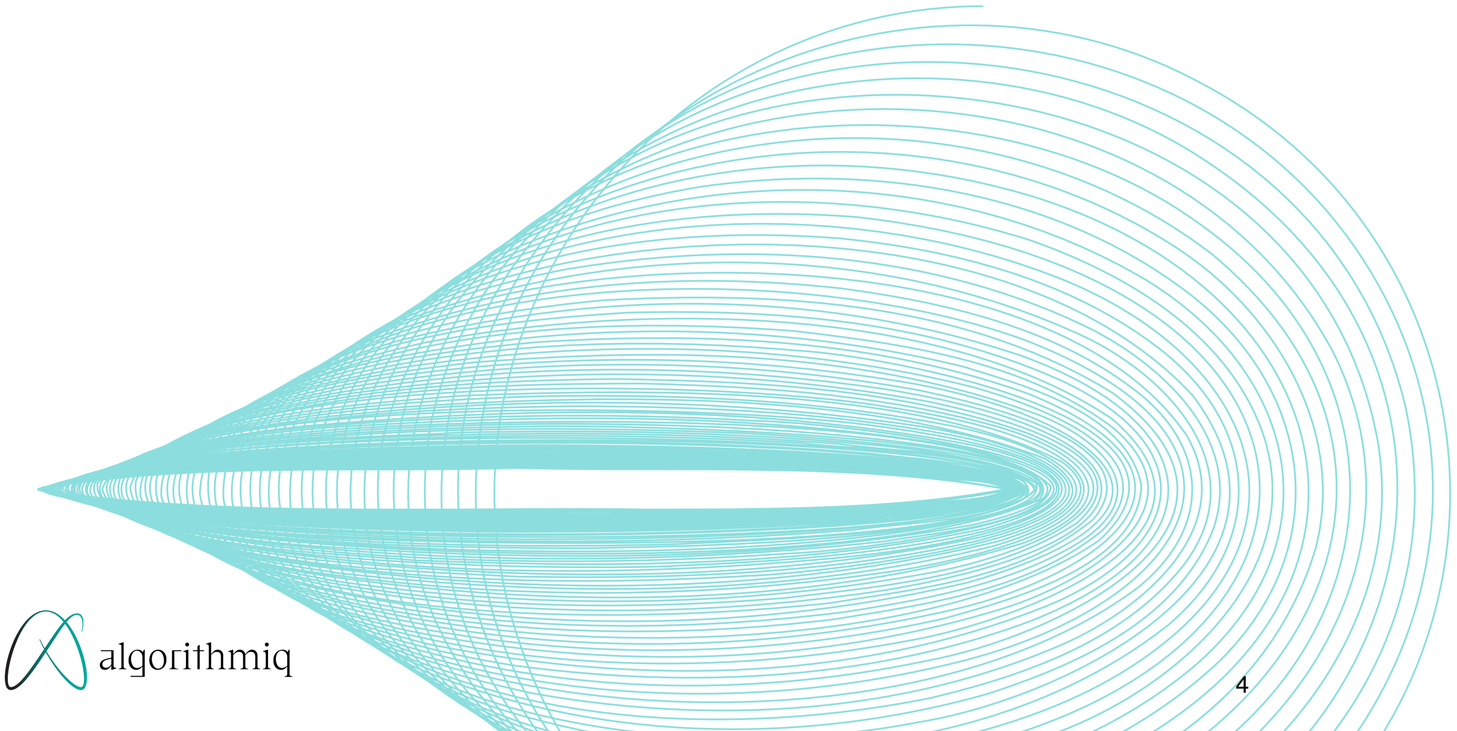
Quantum computing is evolving very quickly

Billions are being invested

Hardware is improving fast

Drug discovery is emerging as a promising area of application



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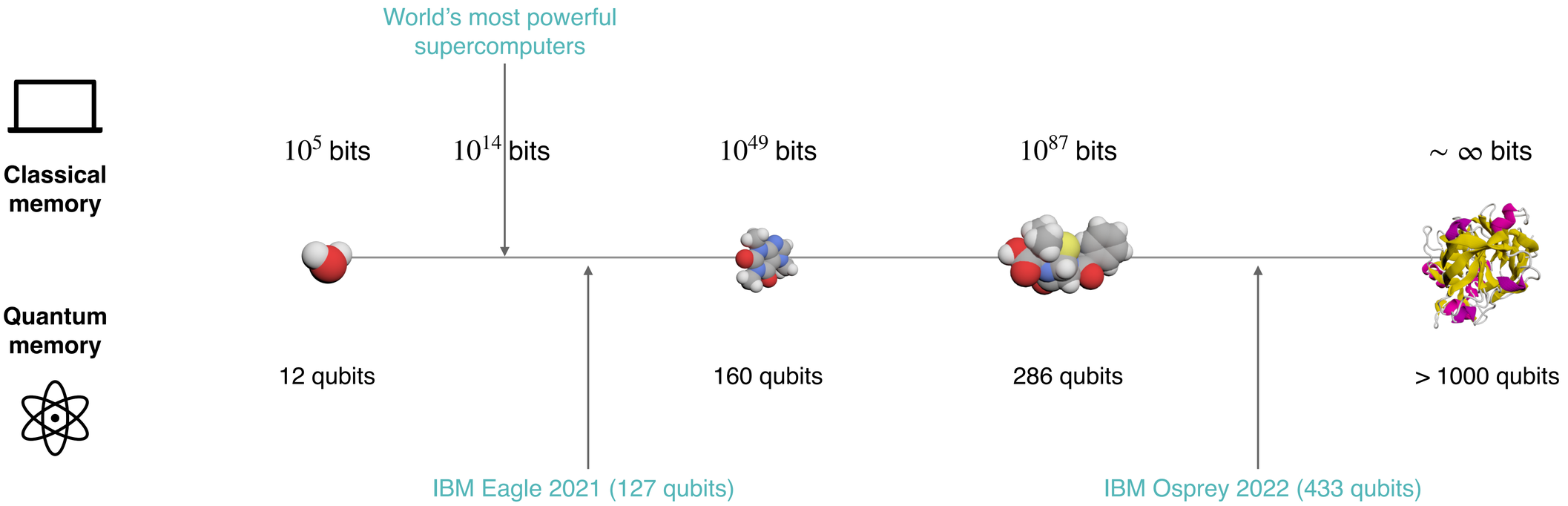
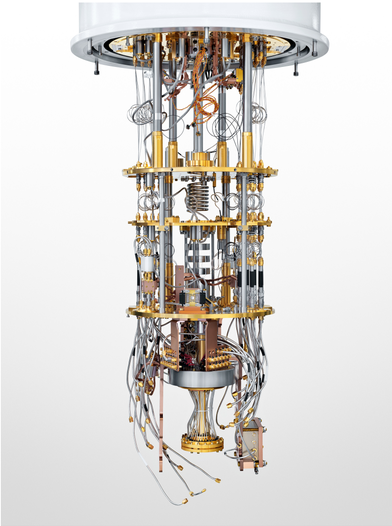
Despite advances in high performance computing it is challenging to model complex molecules on classical computers



Using a quantum computer

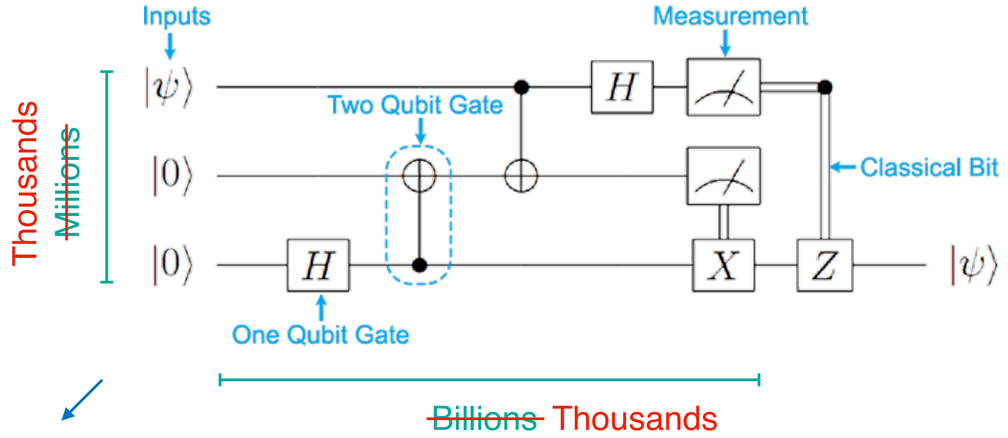
As a quantum physics simulator



Arbitrary state of its quantum bits (qubits)

$$|\Psi\rangle = \sum_{\vec{f}} \alpha_{\vec{f}} |f_1 f_2 \dots f_M\rangle$$

A universal quantum computer can solve problems beyond quantum simulation (e.g. factorisation)



We are in the era of the Noisy Intermediate-Scale Quantum computers: soon useful for simulation!

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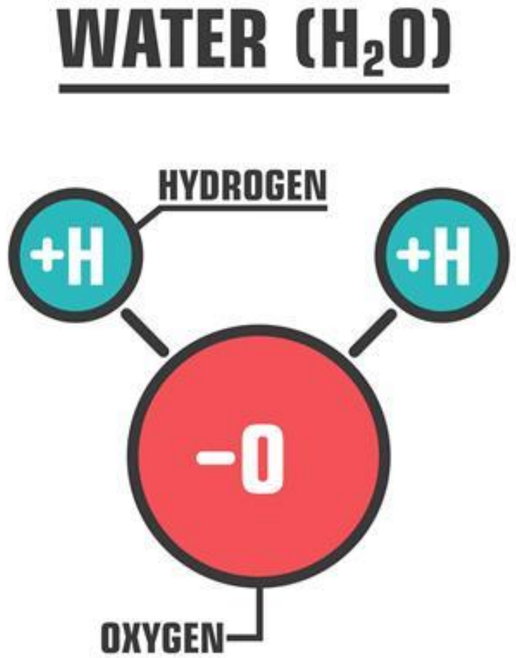
Variational Quantum Eigensolver

(General) problem statement

Molecular ground-state energy

- Composition of the molecule is given



(General) problem statement

Molecular ground-state energy

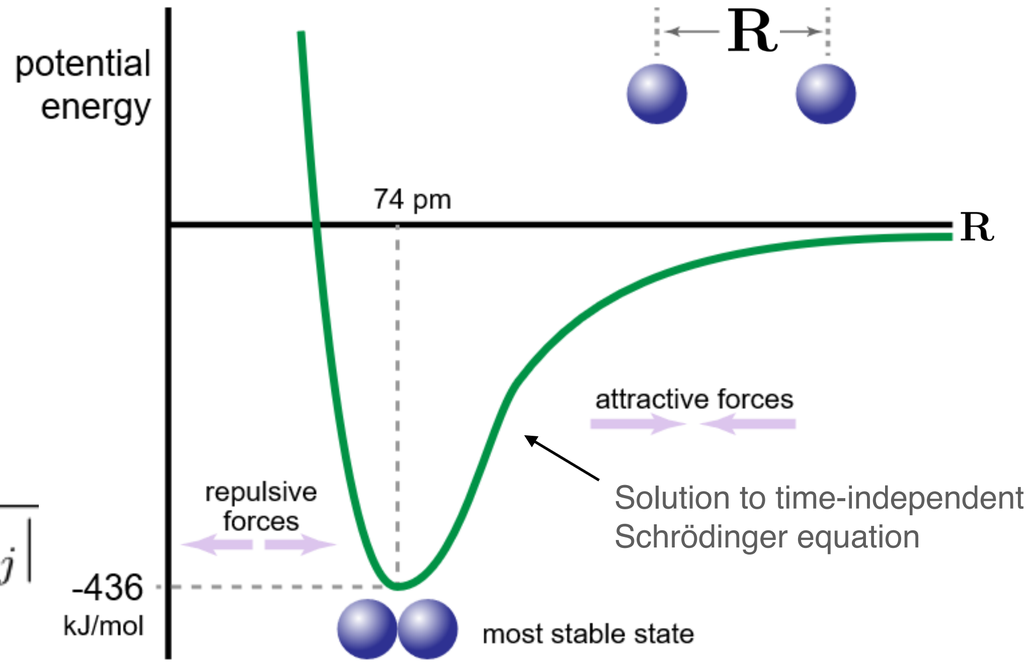
- Composition of the molecule is given
- Must determine ground-state energy as a function of positions of nuclei (potential energy surface)

$$\hat{H}_e = -\frac{1}{2}\sum_i \nabla_i^2 - \sum_i \sum_\alpha \frac{Z_\alpha}{|\mathbf{R}_\alpha - \mathbf{r}_i|} + \frac{1}{2}\sum_{i \neq j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

$$\hat{H}_e|\Psi\rangle = \boxed{E}|\Psi\rangle$$

Well-known from QM theory
Efficient classical representation

High-dimensional vector
in Hilbert space



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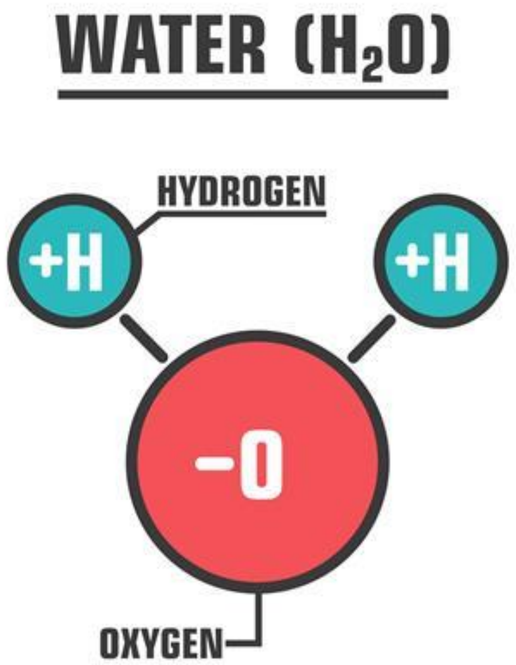
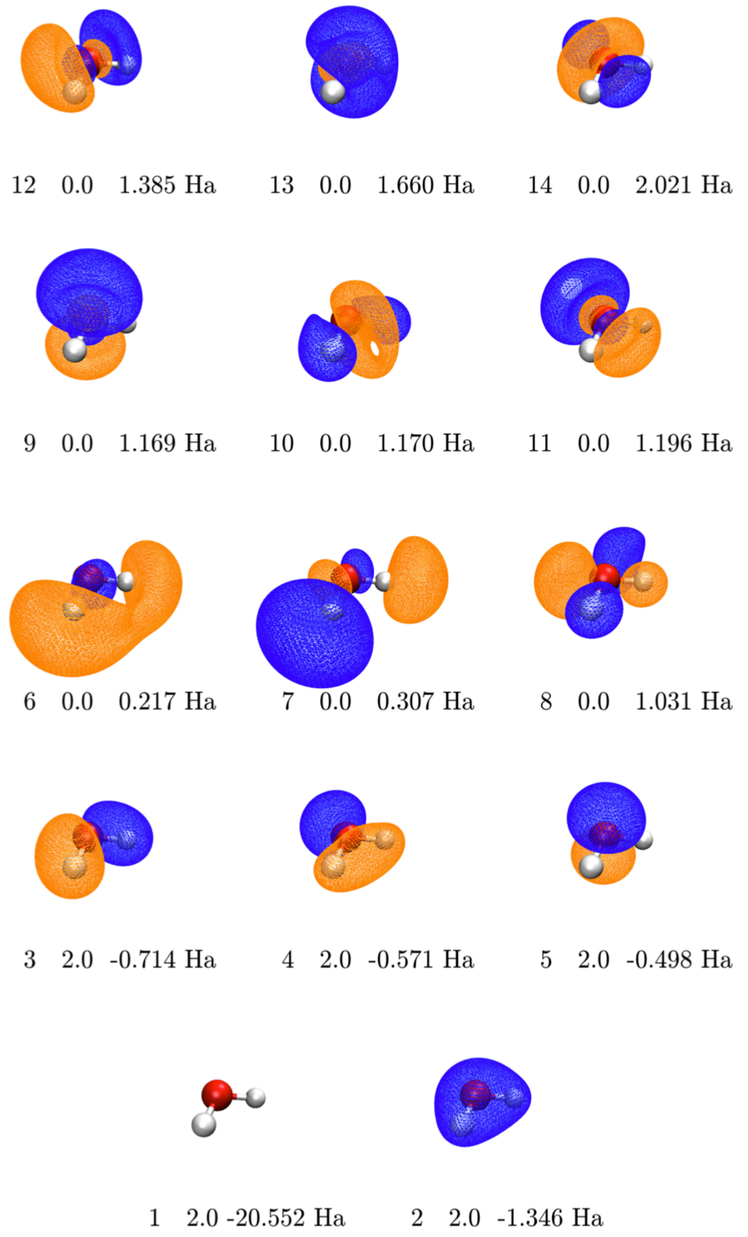
Why is this difficult?

Dimensionality of many-body QM





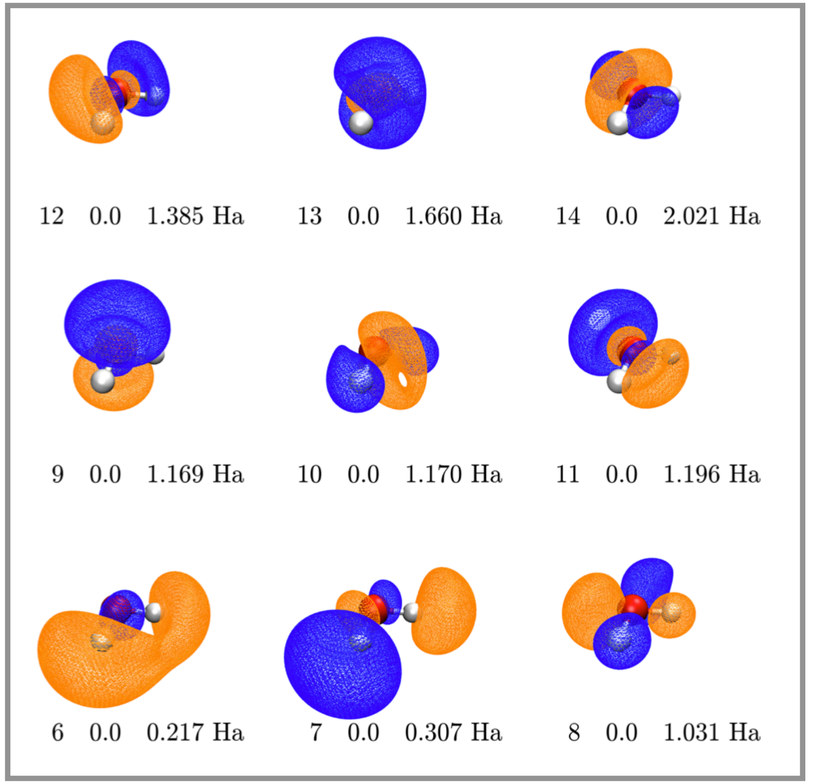
Why is this difficult?

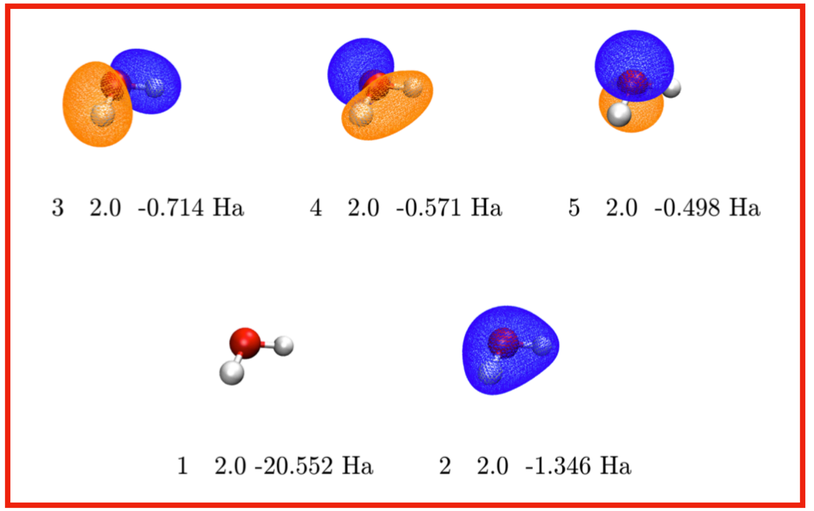
Dimensionality of many-body QM

$$|\Psi\rangle = |f_1 f_2 \dots f_M\rangle$$

Equal to 1 if spin-orbital occupied
Otherwise 0

$$|\Psi\rangle = |1 \cdots 1 0 \cdots 0\rangle$$





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Why is this difficult?

Dimensionality of many-body QM

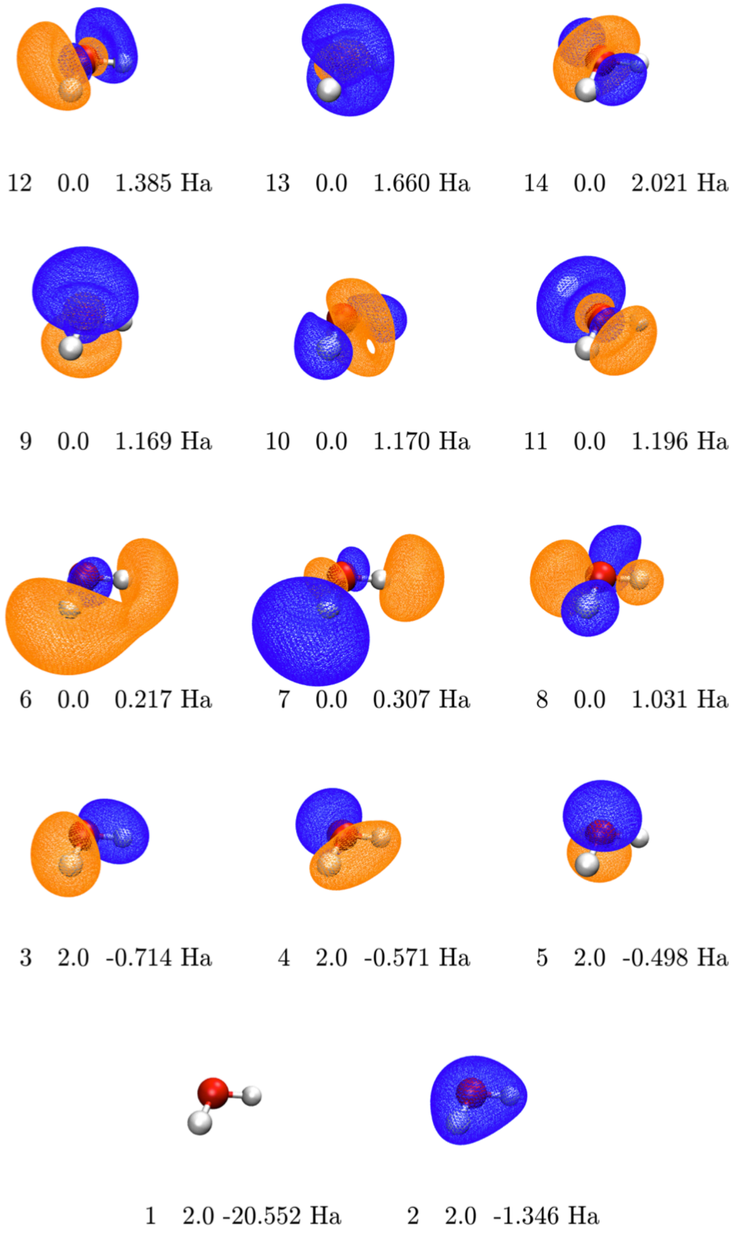
Superposition principle: states are linear combinations of basis states

↓

$$|\Psi\rangle = \sum_{\vec{f}} \alpha_{\vec{f}} |f_1 f_2 \dots f_M\rangle$$

↓

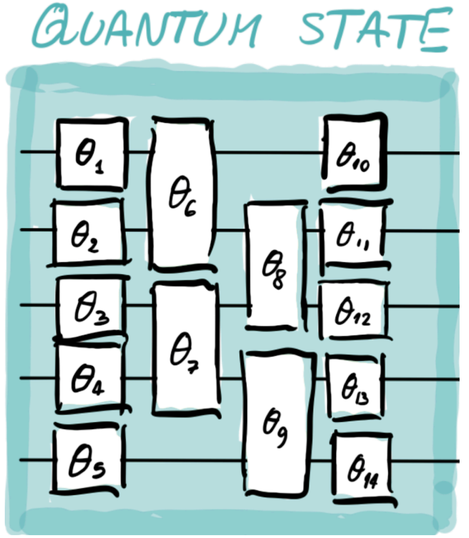
The dimensionality of state space is classically intractable



Solving the problem

The Variational Quantum Eigensolver

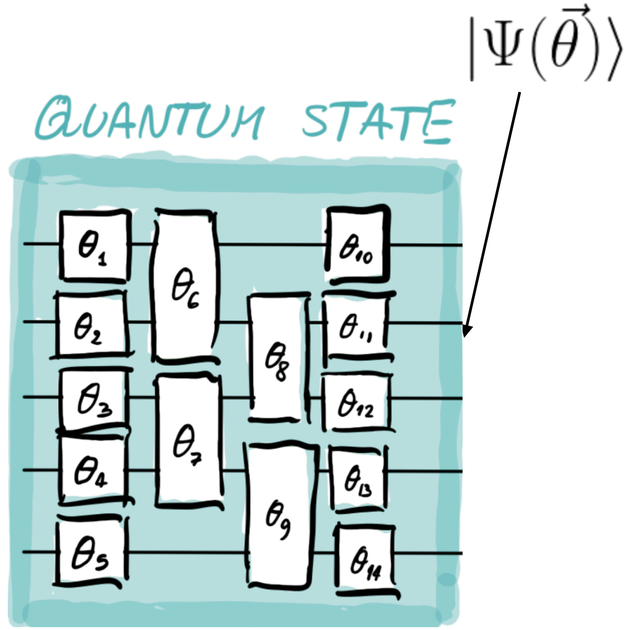
- Prepare *some* quantum state using a so-called variational form (ansatz)



Solving the problem

The Variational Quantum Eigensolver

- Prepare *some* quantum state using a so-called variational form (ansatz)
- Gates in the ansatz have free parameters



Solving the problem

The Variational Quantum Eigensolver

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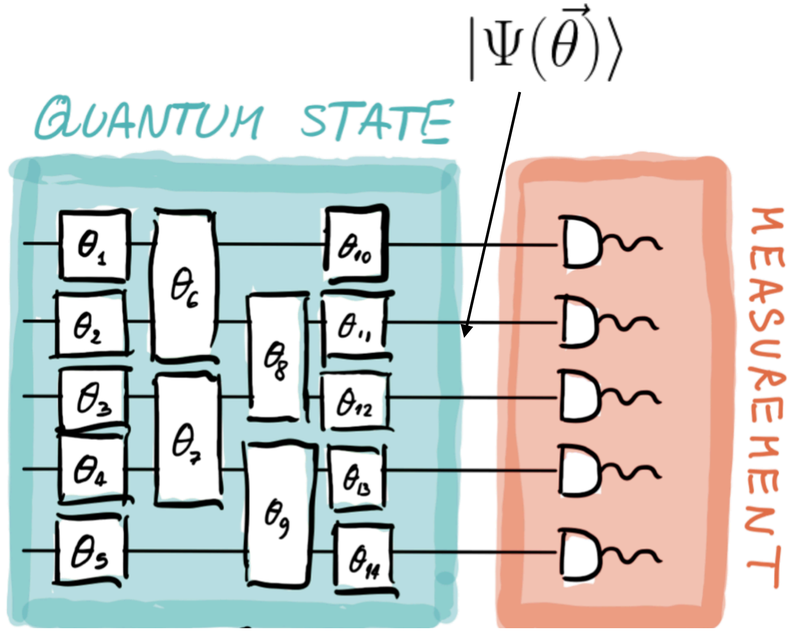
- For each value of the parameters the resulting state has some mean energy

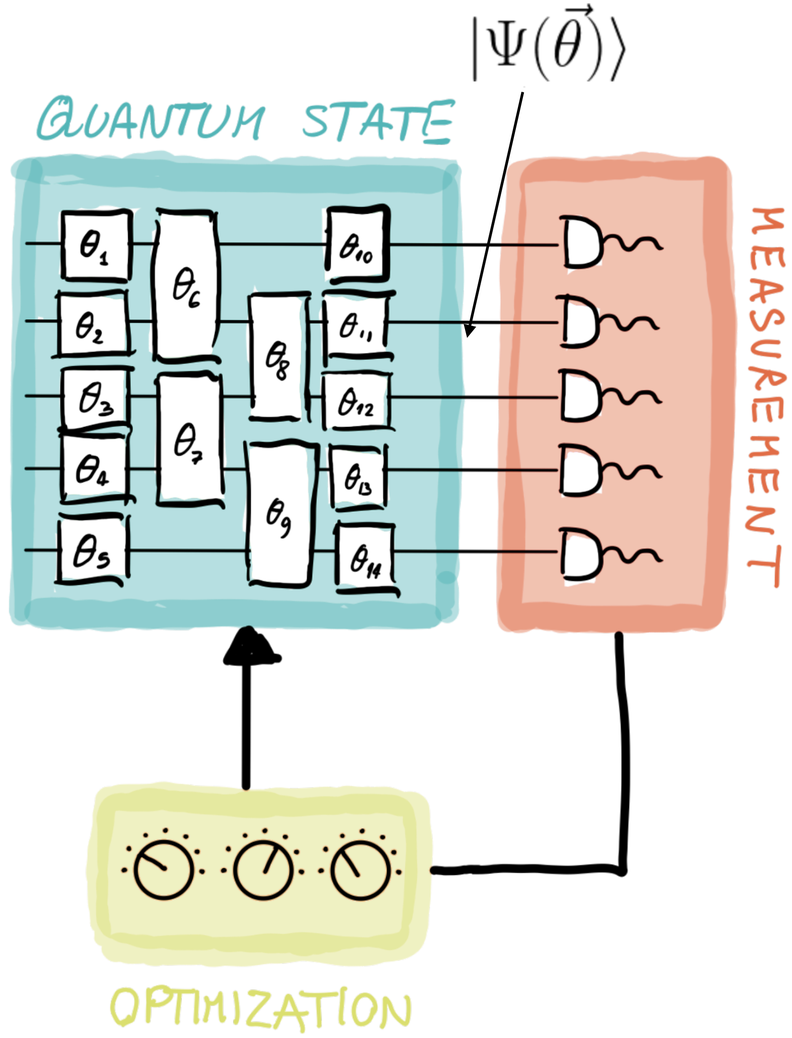


$$\langle E \rangle = \langle \Psi(\vec{\theta}) | \hat{H}_e | \Psi(\vec{\theta}) \rangle \geq E_{\text{ground}}$$

Solving the problem

The Variational Quantum Eigensolver

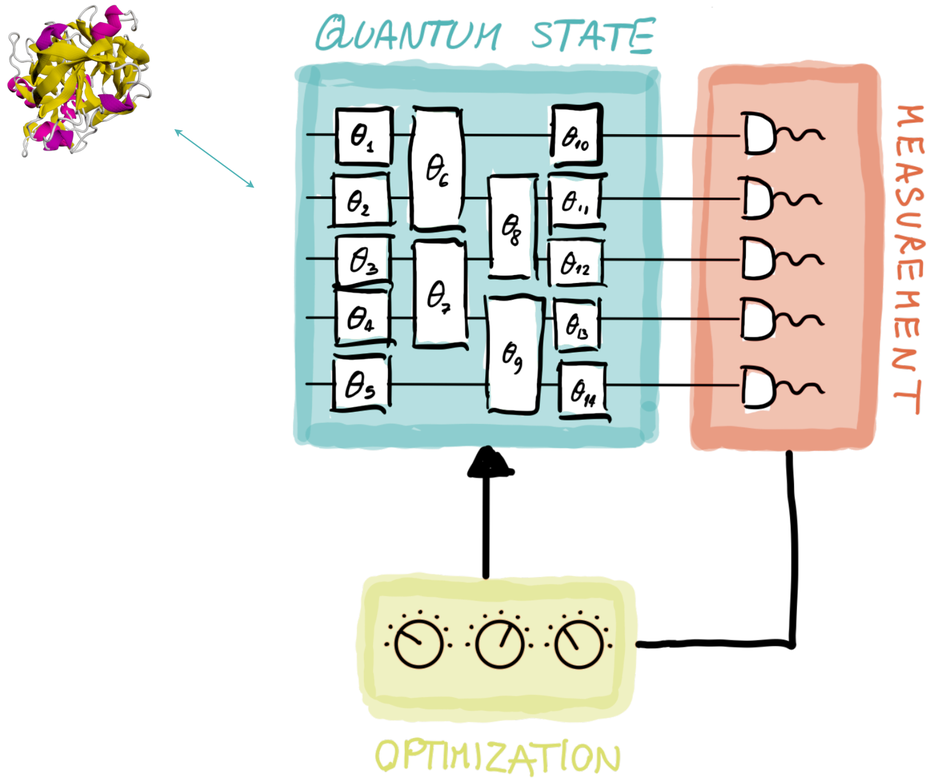
- Prepare *some* quantum state using a so-called variational form (ansatz)
- Gates in the ansatz have free parameters
- For each value of the parameters the resulting state has some mean energy
- Find the ground state variationally, that is, minimising over the parameters



$$\langle E \rangle = \langle \Psi(\vec{\theta}) | \hat{H}_e | \Psi(\vec{\theta}) \rangle \geq E_{\text{ground}}$$

Quantum chemistry in the near term

The variational quantum eigensolver

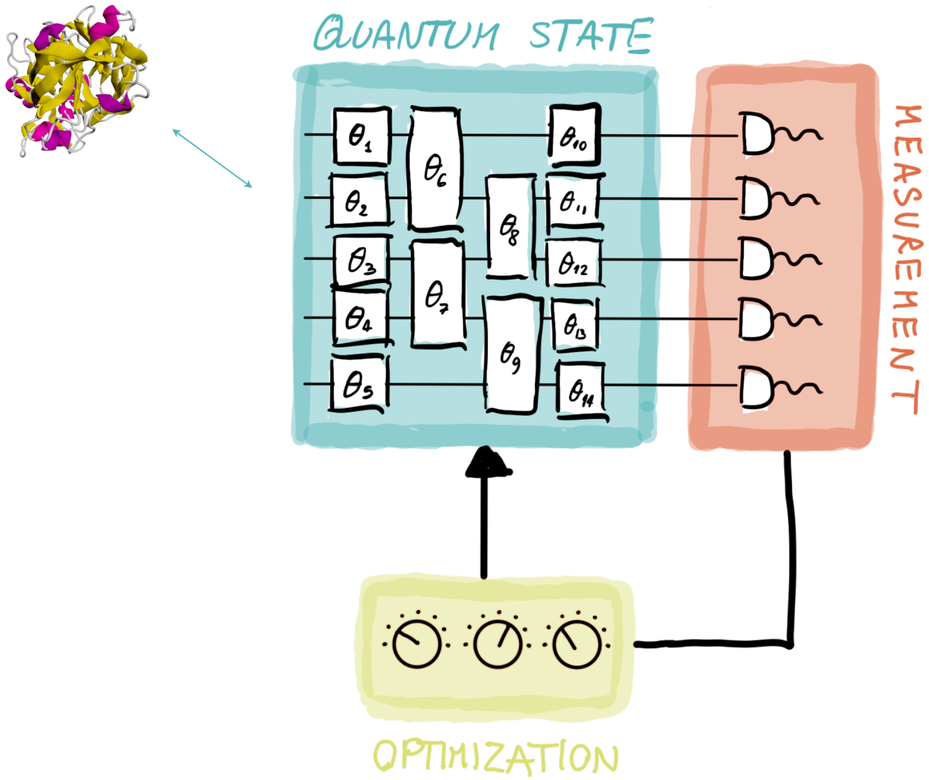


Challenges

- Hilbert space is a big space
- Quantum computers are error-prone
- Not many qubits available
- Simulations can be time-consuming

Quantum chemistry in the near term

The variational quantum eigensolver



Challenges

- Hilbert space is a big space
- Quantum computers are error-prone
- Not many qubits available
- Simulations can be time-consuming

Identifying challenges towards practical quantum advantage through resource estimation: the measurement roadblock in the variational quantum eigensolver

Jérôme F. Gonthier,[1] Maxwell D. Radin,[1] Corneliu Buda,[2]
Eric J. Doskocil,[2] Clena M. Abuan,[3] and Jhonathan Romero[1]
[1]*Zapata Computing, Inc., 100 Federal St., Boston, MA 02110, USA*

Molecule	H_2O	CO_2	CH_4	CH_4O	C_2H_6	C_2H_4	C_2H_2	C_2H_6O	C_3H_8	C_3H_6	C_3H_4
N_{el}	8	16	8	14	14	12	10	20	20	18	16
N_q	104	208	104	182	182	156	130	260	260	234	208
$K \cdot 10^{-3}$	1.9	16	1.6	8.4	8.5	6.6	3.1	24	16	23	18
$M \cdot 10^{-9}$	3.9	32	3.2	17	17	13	6.2	48	31	46	36
t (days)	2.3	39	1.9	18	18	12	4.6	71	47	62	44

TABLE IV. Estimated runtimes t in days for a single energy evaluation using the number of measurements M from extrapolated

State preparation

in a VQE simulation

- Borrow from the coupled cluster (CC) method in computational chemistry

$$|\mathrm{CC}\rangle = \mathrm{e}^{\hat{T}}|0\rangle \qquad \hat{T} = \sum_k \hat{T}_k = \sum_k \frac{1}{(k!)^2} \left(\sum_{i_1 \cdots i_k} \sum_{a_1 \cdots a_k} t^{i_1 \cdots i_k}_{a_1 \cdots a_k} a^\dagger_{a_1} a^\dagger_{a_k} a_{i_k} \cdots a_{i_1} \right)$$

- Use its ***unitary variant*** $|\mathrm{UCC}\rangle = \mathrm{e}^{\hat{T} - \hat{T}^\dagger}|0\rangle$
- ***Truncated*** to single and double excitations $\hat{T} = \hat{T}_1 + \hat{T}_2$
- Apply in ***Trotterized form***

Measuring the energy

in a VQE simulation

- The Hamiltonian is given as a linear combination of Pauli strings

$\hat{H}_e = \sum c_{\mathbf{k}} \hat{P}_{\mathbf{k}}$ ⟵ Each term is a product of local operators $P_{\mathbf{k}} = \bigotimes_{i=1}^{N} \sigma_{k_i}^{(i)}$

- We can calculate expectation values on the QC

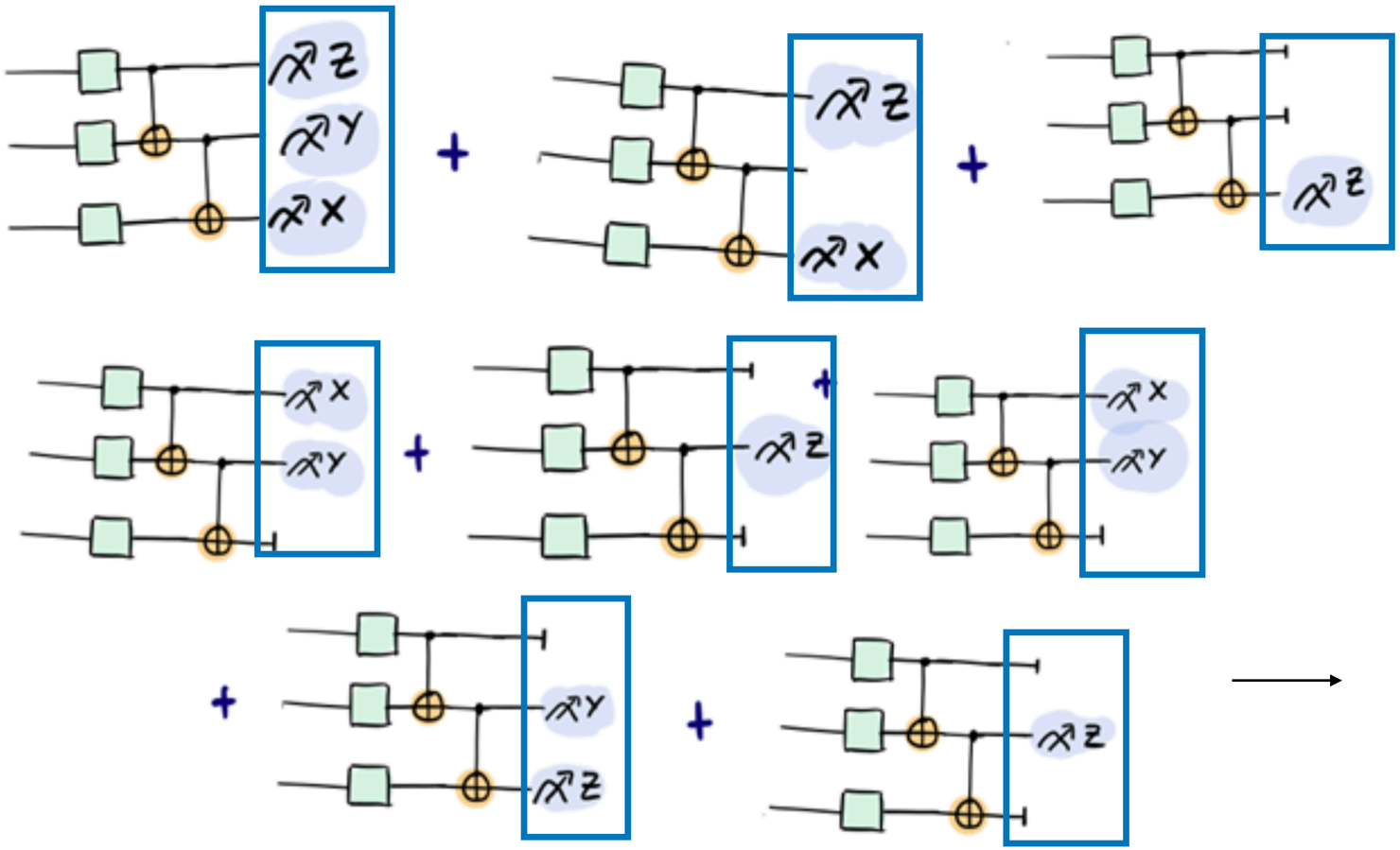
$$|\Psi\rangle = \sum_{\vec{f}} \alpha_{\vec{f}} |f_1 f_2 \dots f_M\rangle \longrightarrow \langle\Psi|\hat{H}_e|\Psi\rangle = \sum c_{\mathbf{k}} \langle\Psi|\hat{P}_{\mathbf{k}}|\Psi\rangle$$

Cannot even write down on a classical computer

Easy on a quantum computer: only requires measuring Pauli strings

(Specific) problem statement

Measurement cost in VQE



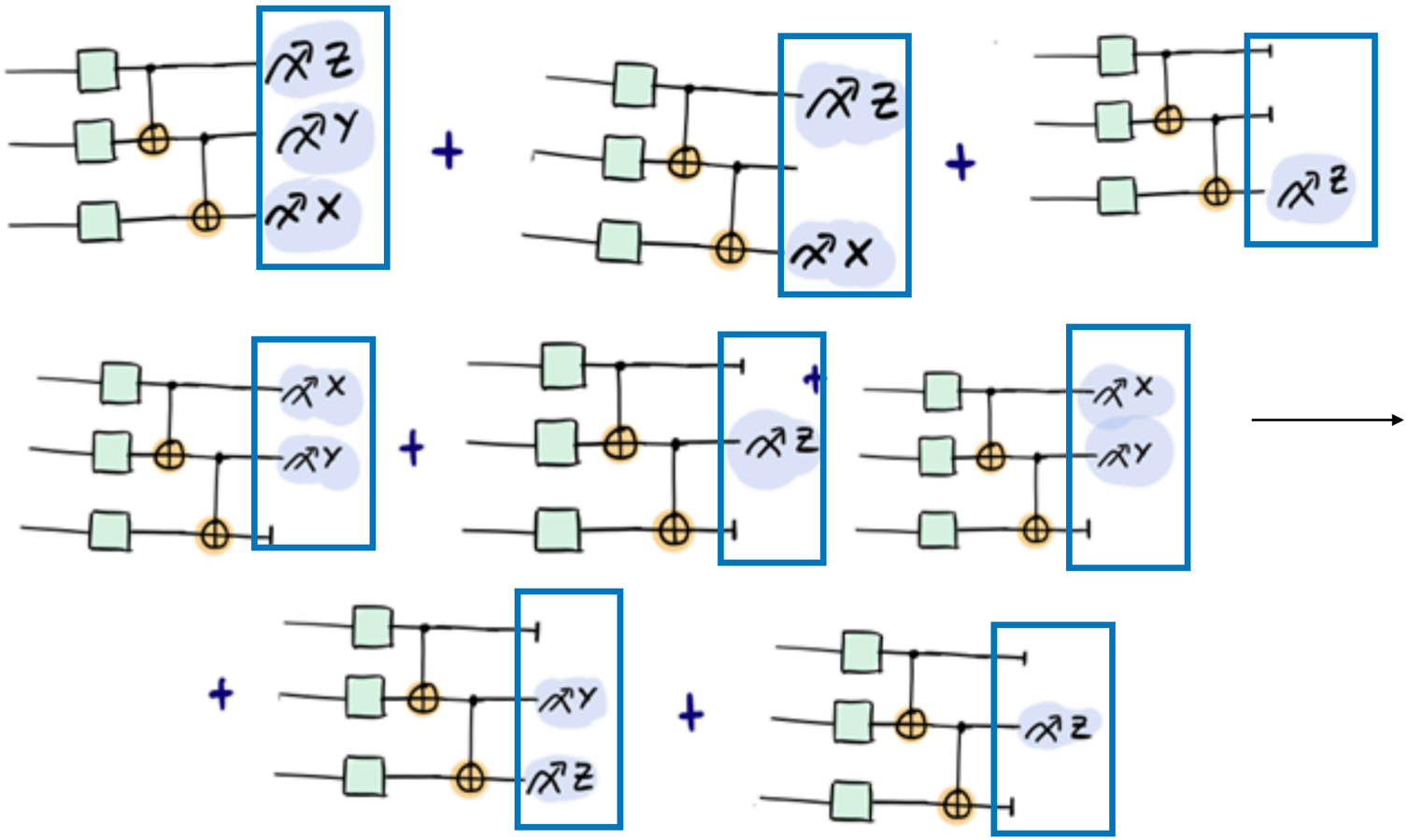
$$\langle \Psi | \hat{H}_e | \Psi \rangle = \sum c_{\mathbf{k}} \langle \Psi | \hat{P}_{\mathbf{k}} | \Psi \rangle$$

Every Pauli string evaluated independently through repeated measurements

Repeat each many times to estimate the mean $\langle \Psi | \hat{P}_{\mathbf{k}} | \Psi \rangle$

(Specific) problem statement

Measurement cost in VQE



$$\langle\Psi|\hat{H}_e|\Psi\rangle = \sum c_{\mathbf{k}}\langle\Psi|\hat{P}_{\mathbf{k}}|\Psi\rangle \quad \sim O(N^4)$$

Every Pauli string evaluated independently through repeated measurements

Repeat each many times to estimate th$\langle\Psi|\hat{P}_{\mathbf{k}}|\Psi\rangle$

Estimation error:

$$\epsilon = \sqrt{\sum_{\mathbf{k}} |c_{\mathbf{k}}|^2 \mathrm{Var}(P_{\mathbf{k}})/S_{\mathbf{k}}}$$

Number of shots needed to reach given precision: S = $O(N^{5\div6})$

References

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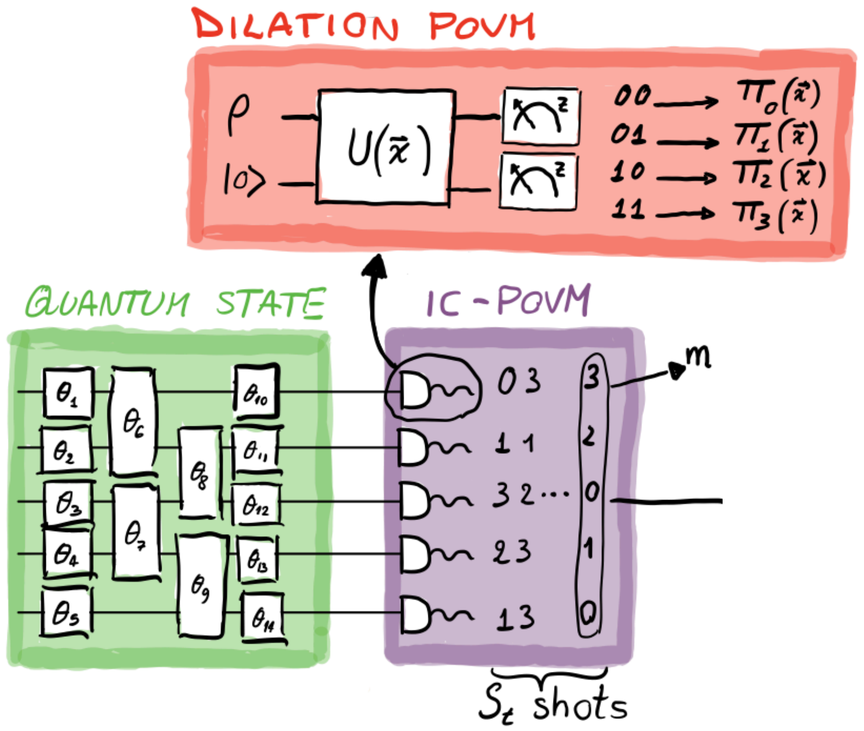
Informationally Complete Measurements

Tackling the measurement problem

with generalised measurements

Using generalised quantum measurements

- Add ancillary qubit in a known state
- Apply a two-qubit transformation
- Measure both in the computational basis



Tackling the measurement problem

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$$\Pi_i > 0$$

$$\sum_i \Pi_i = \mathbb{I}$$

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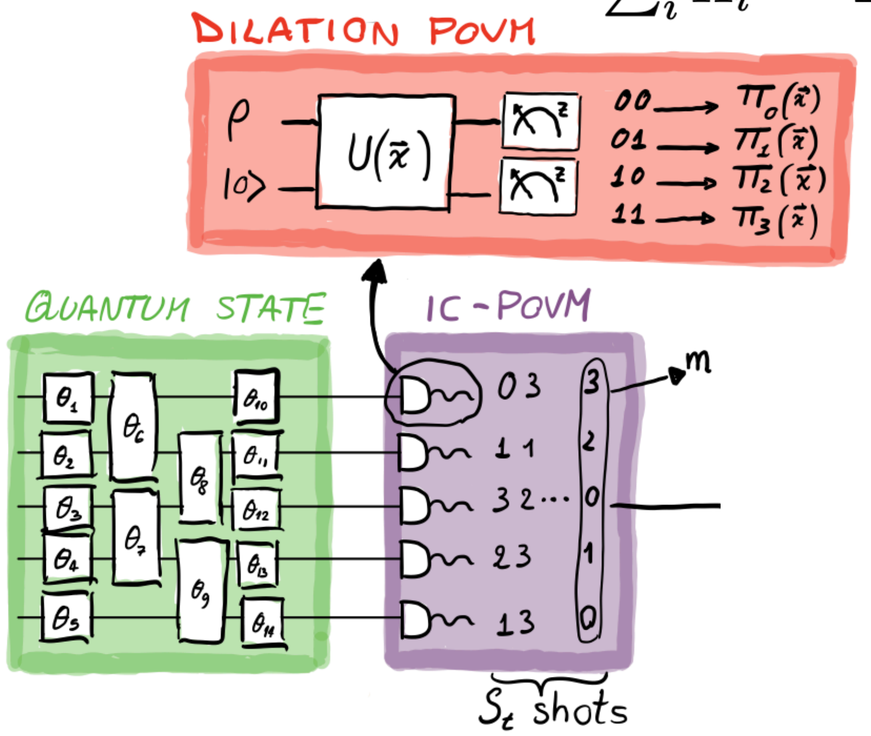

Probability of outcome **m**: $\mathrm{Tr}[\rho\Pi_{\mathbf{m}}]$ where $\Pi_{\mathbf{m}} = \bigotimes_{i=1}^{N} \Pi_{m_i}^{(i)}$

Tackling the measurement problem

with generalised measurements

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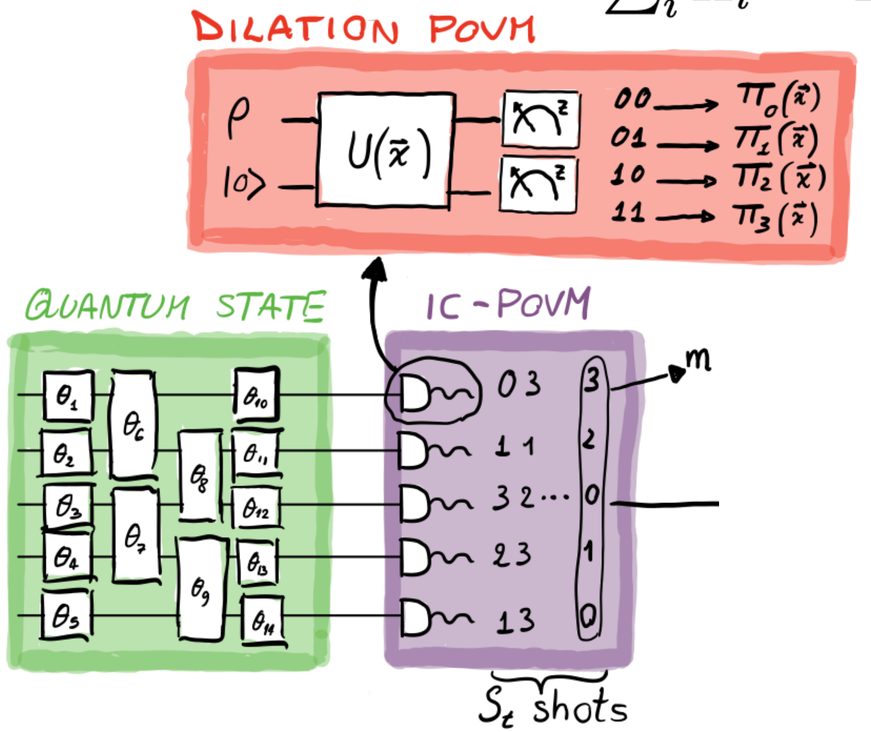

Using generalised quantum measurements

- Add ancillary qubit in a known state
- Apply a two-qubit transformation
- Measure both in the computational basis
- informationally complete

$$\mathcal{O} = \sum \omega_{\mathbf{m}} \Pi_{\mathbf{m}} \qquad \langle \mathcal{O} \rangle = \sum \omega_{\mathbf{m}} Tr[\rho \Pi_{\mathbf{m}}]$$

Probability of outcome **m**: $\mathrm{Tr}[\rho \Pi_{\mathbf{m}}]$ where $\Pi_{\mathbf{m}} = \bigotimes_{i=1}^{N} \Pi_{m_i}^{(i)}$

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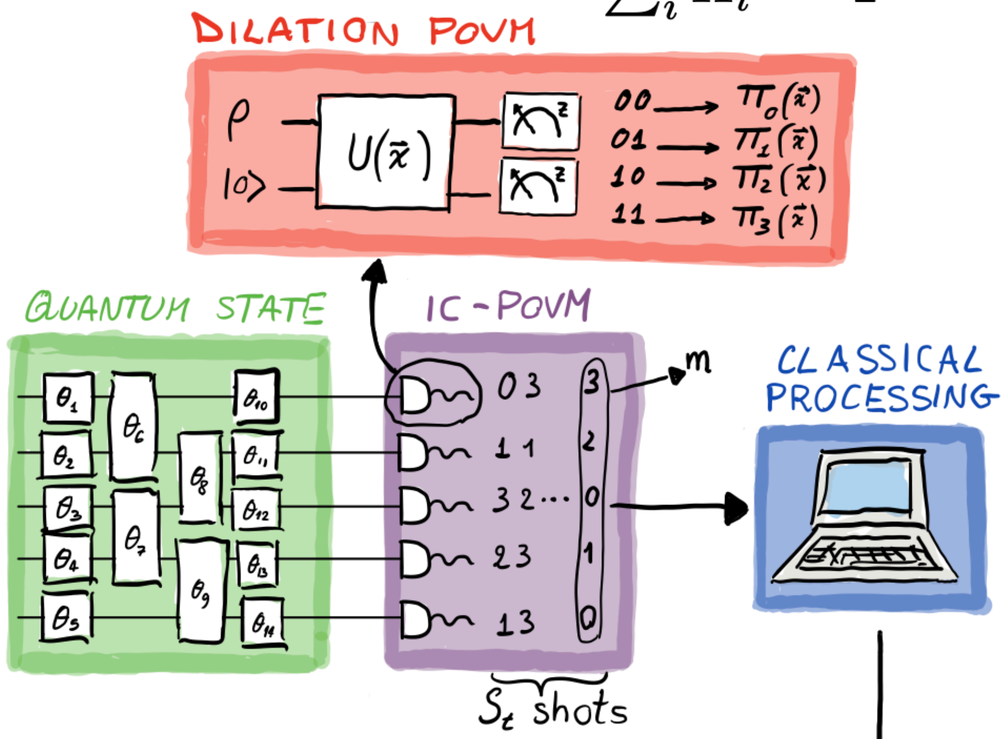

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Energy → $(\overline{\mathcal{O}}_t, \overline{V}_t)$

Variance

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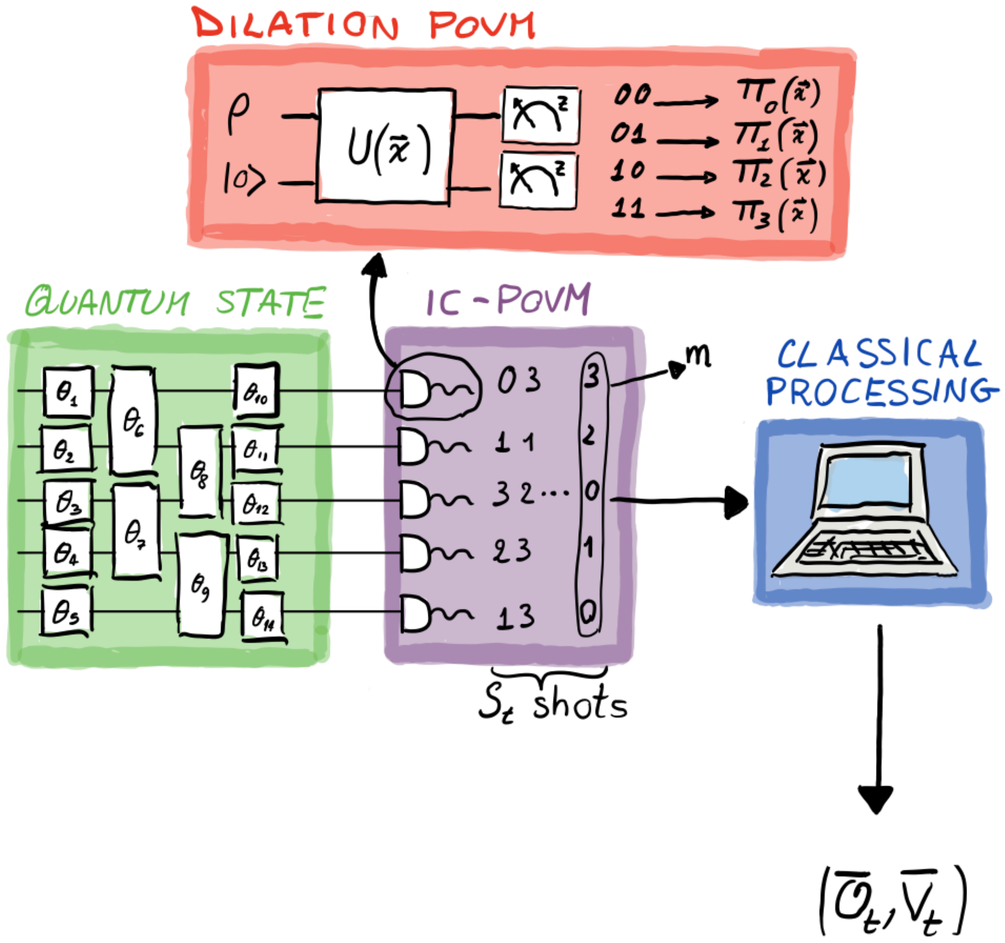
Tackling the measurement problem

with adaptive generalised measurements

Optimising measurements on the fly

- Qubit-ancilla interaction is parametrised

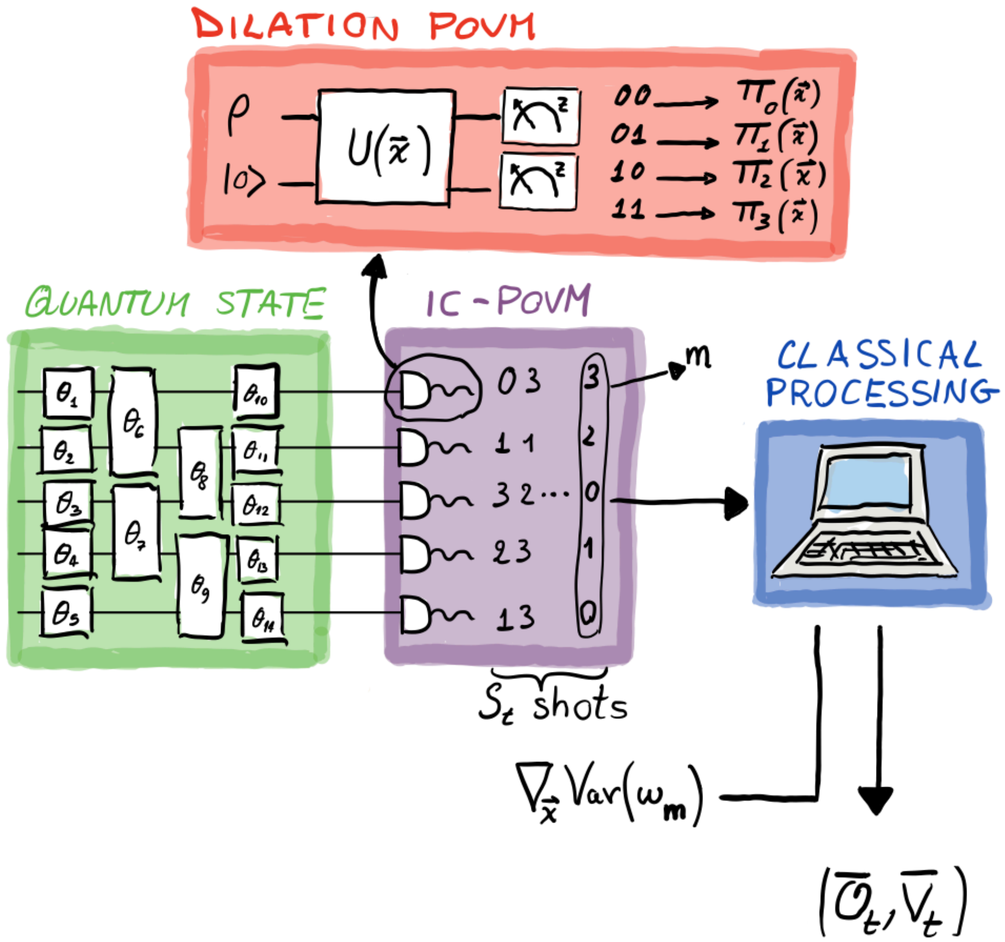


Tackling the measurement problem

with adaptive generalised measurements

Optimising measurements on the fly

- Qubit-ancilla interaction is parametrised
- Can use informationally complete data to calculate gradient of measurement error



Tackling the measurement problem

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- Qubit-ancilla interaction is parametrised
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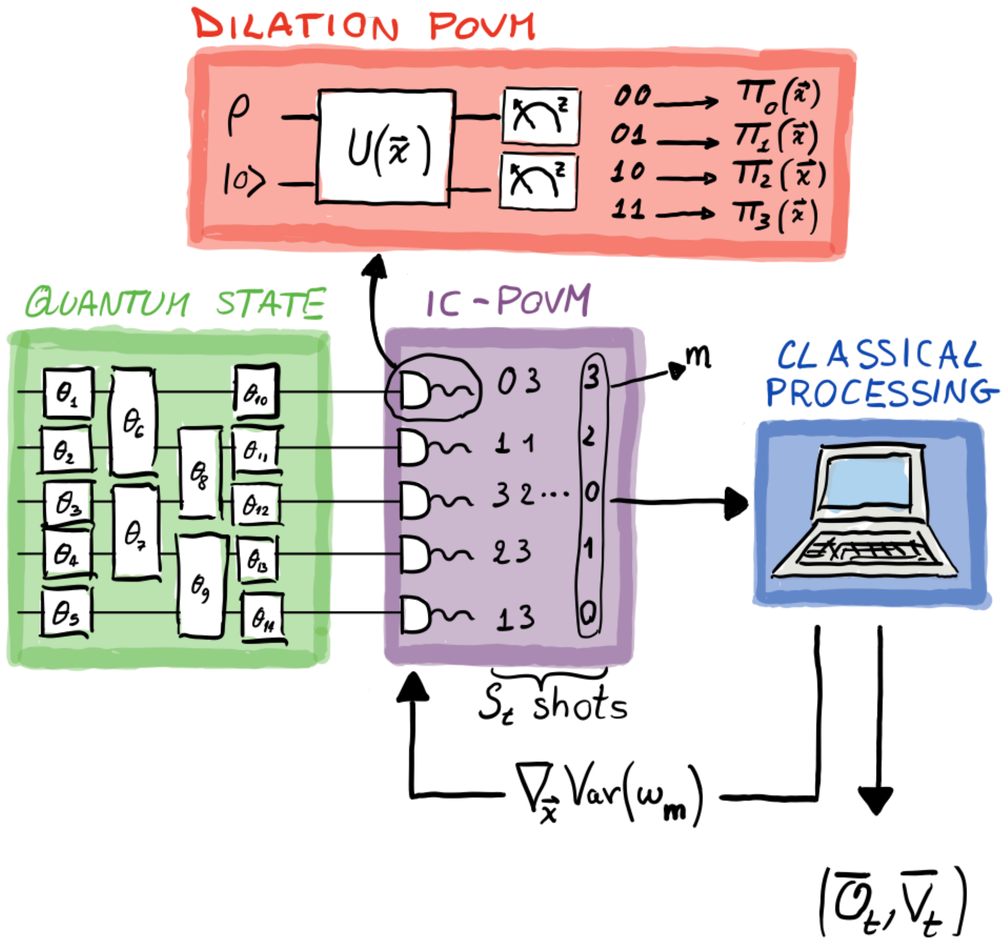
- At every iteration, we use a better POVM than in the previous one



Tackling the measurement problem

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Optimising measurements on the fly

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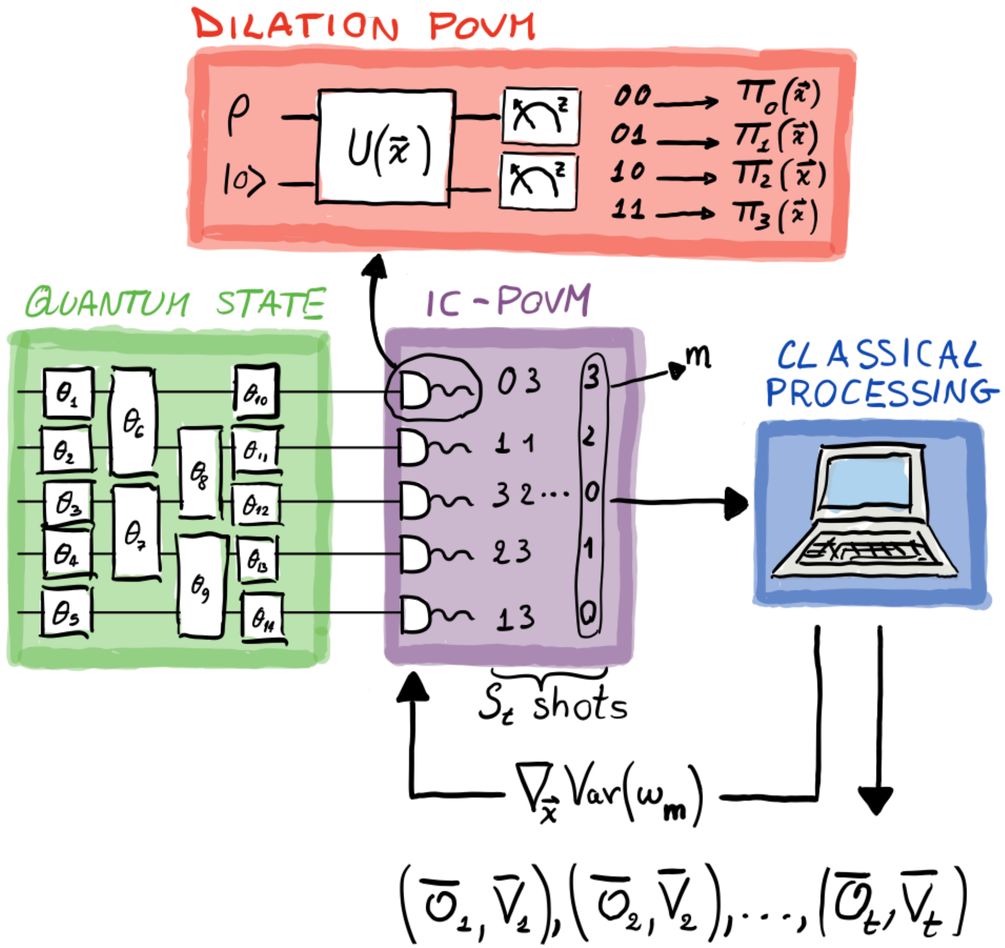
- At every iteration, we use a better POVM than in the previous one
- We produce many estimators of the mean with different statistical error



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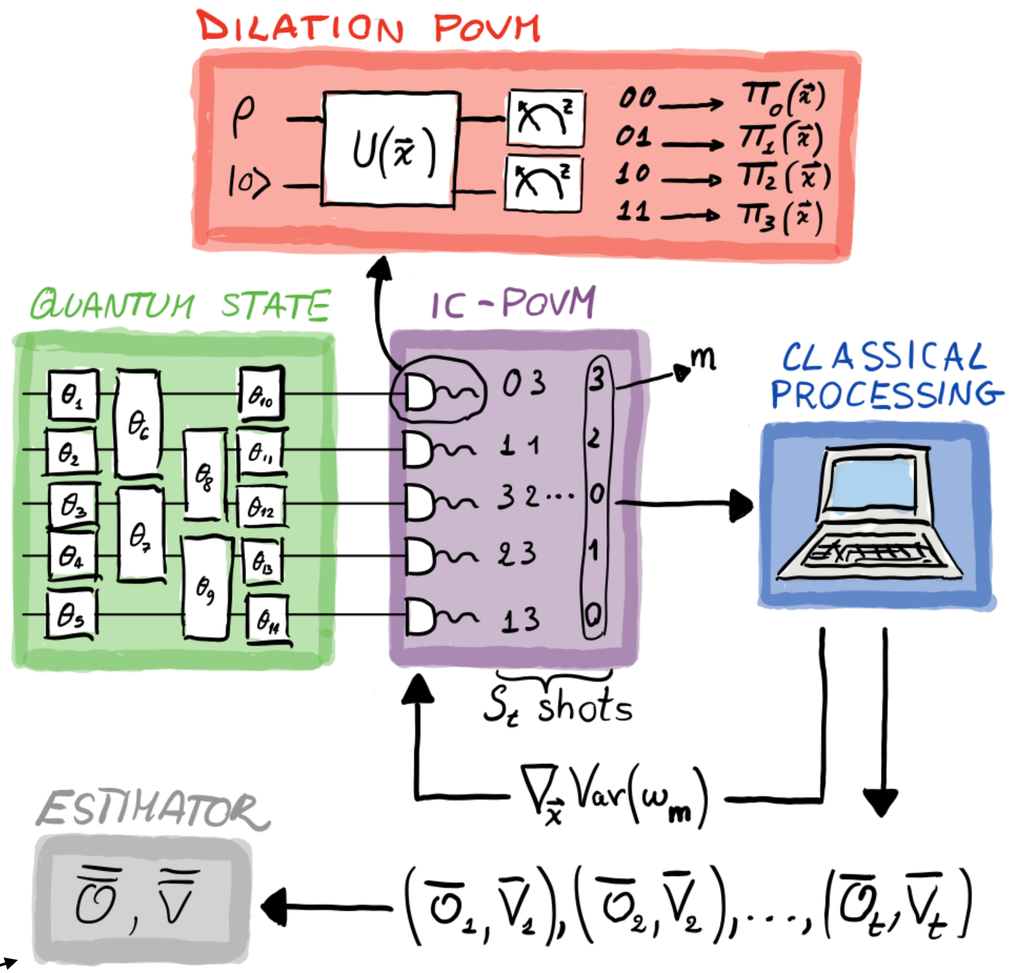
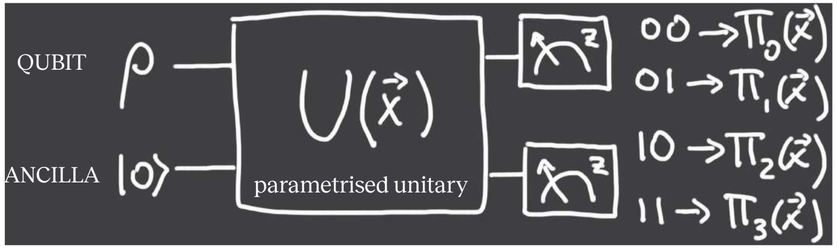


Final estimation combining intermediate results

POVM implementations

1. Dilation POVM [García-Pérez te al. PRX Quantum **2**, 040342 (2021)]
2. Physical dilation POVM [Fischer et al. PRR 4 (2022)]
3. Randomized unitaries [Glos et al., arXiv:2208.07817]



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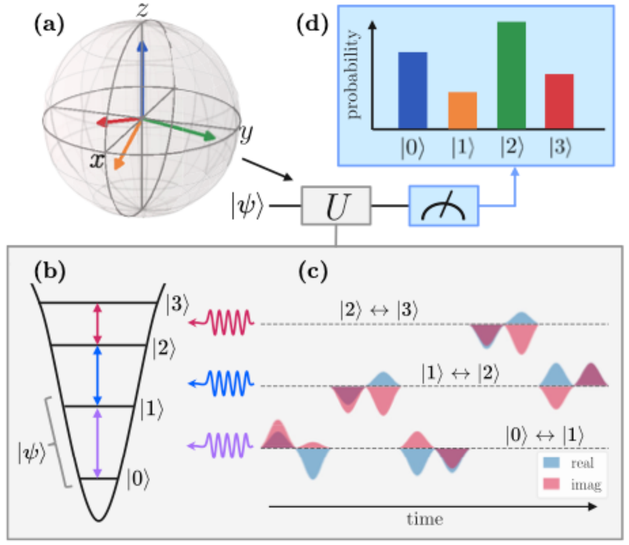
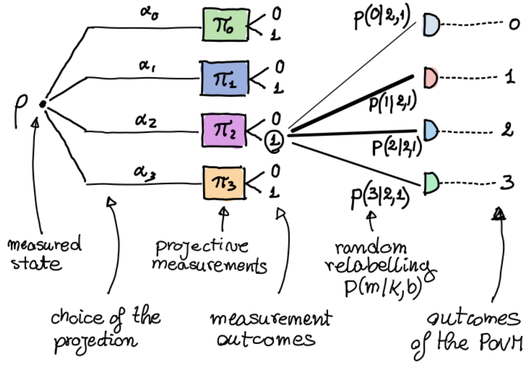


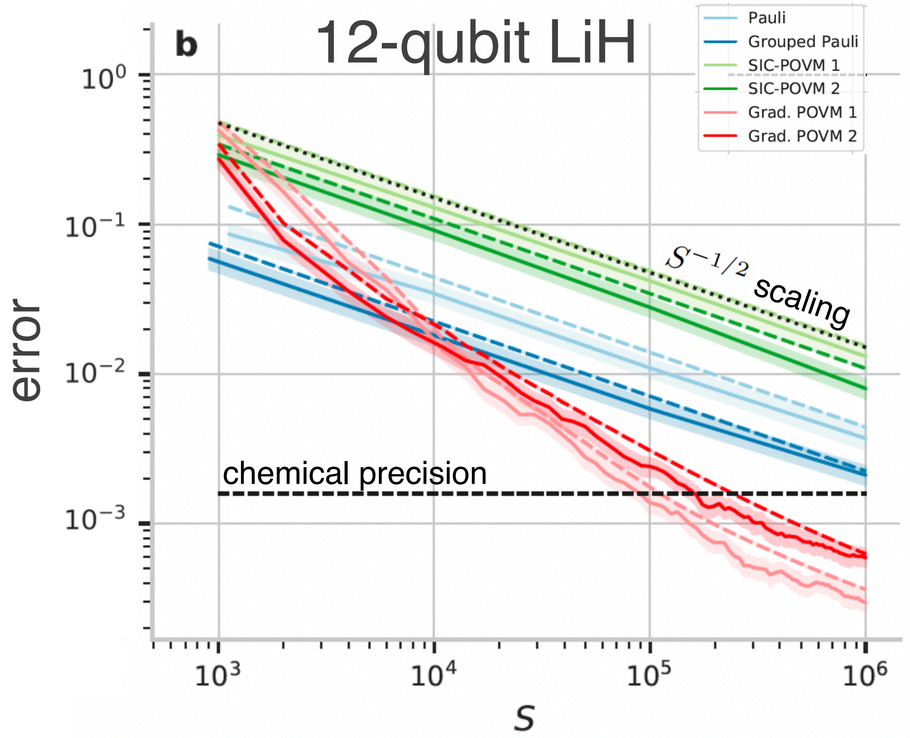
[2]

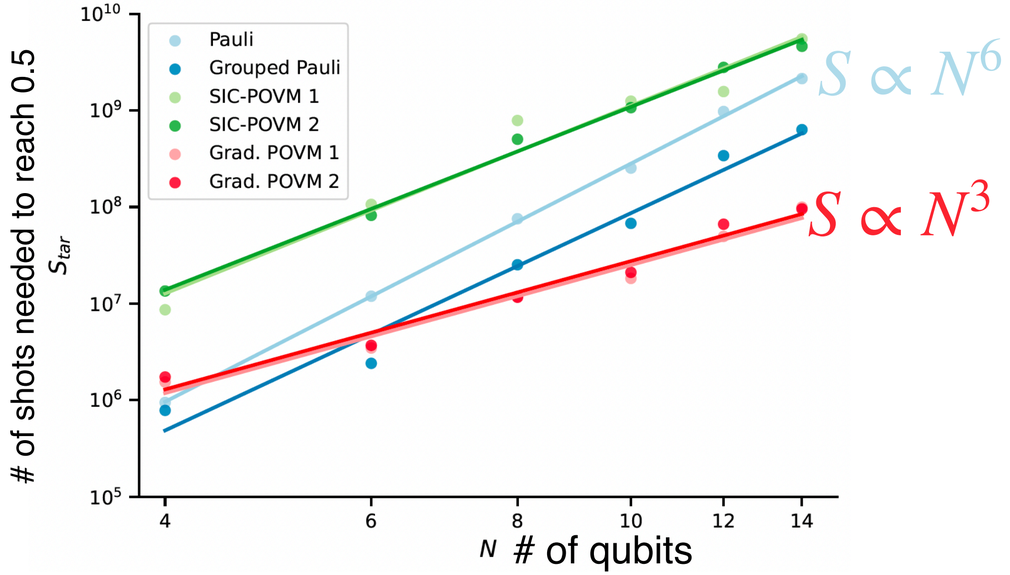


[3]

Benefits of IC-POVMs

- Can be adapted to improve estimation precision for a given state and observable
- Provide better scaling of the number of measurements vs number of qubits
- Allow to estimate other observables with the same data:
  - RDMs
  - Commutators
  - Noise mitigation





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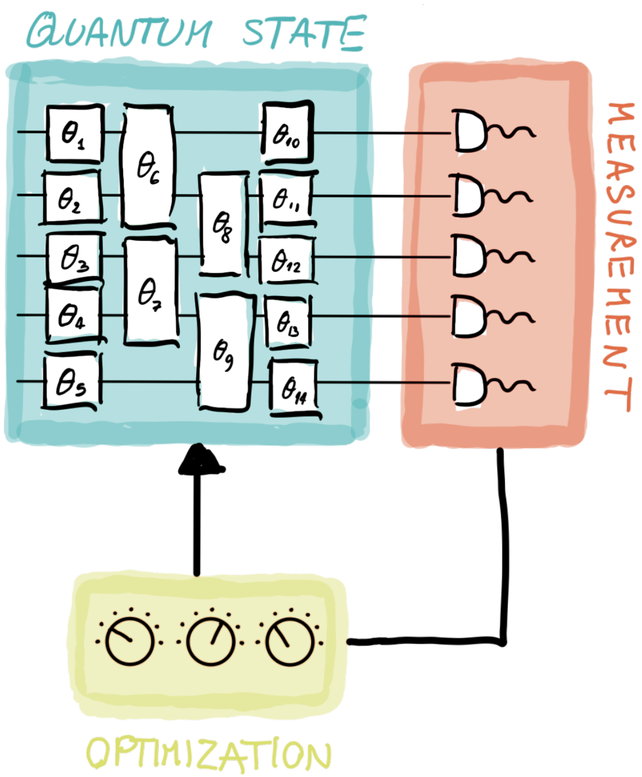
Advanced State Preparation Techniques

Ansatz preparation

Efficient representation of state

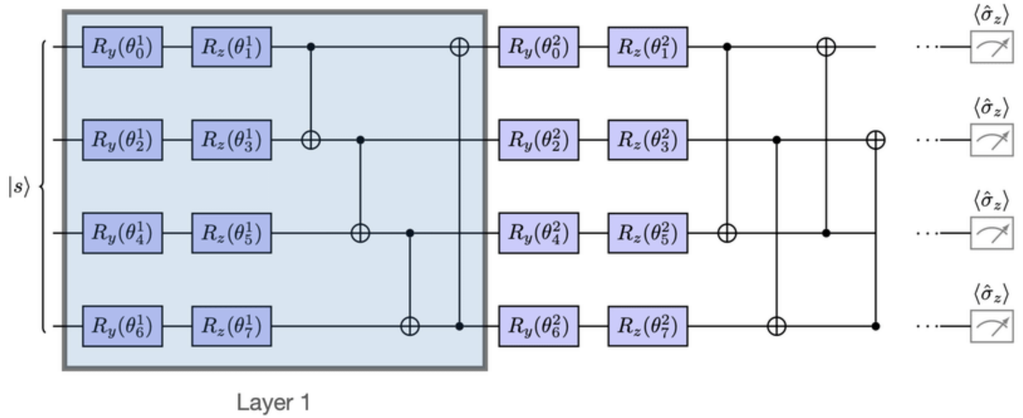
- Design a quantum circuit, which efficiently covers the part of Hilbert space that contains the target quantum state

- Things to consider
  - Expressibility
  - Optimisability
  - Scalability
  - Depth
  - Gate count
  - Hardware layout
  - Native gate set



Ansatz types

- Hardware efficient Ansatz
  - Expressible circuits, but too much -> Hard to optimize
  - Barren plateaus -> Finding gradient direction requires very precise measurements
  - Breaks Symmetries (Spin and Particle number)



- Unitary Coupled-Cluster Singles and Doubles (UCCSD)
  - Chemically inspired, ansatz consists of fermionic single and double excitations
  - Long circuits, too many parameters and gate ordering not optimal
  - Improvements exist, but tradeoffs between depth and accuracy

$$\hat{\tau}_p^q = a_q^\dagger a_p - a_p^\dagger a_q$$
$$\hat{\tau}_{rs}^{pq} = a_p^\dagger a_q^\dagger a_r a_s - a_s^\dagger a_r^\dagger a_q a_p$$

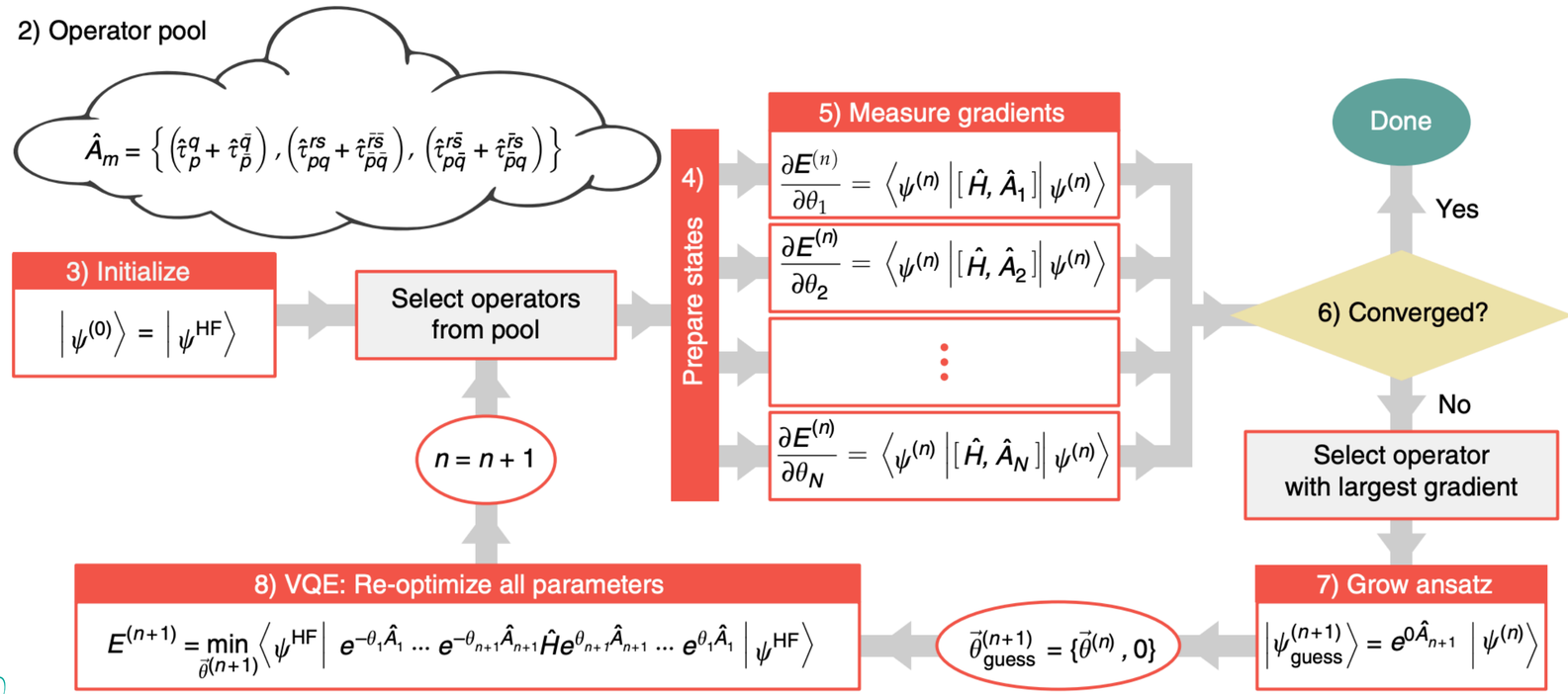
Adaptive Ansätze

ADAPT = Adaptive Derivative-Assembled Pseudo-Trotter ansatz

- Adaptively build a problem-tailored ansatz
  - Expressive enough to contain the ground state
  - Not too expressive to make it easily optimisable

- Operators chosen by their energy gradient

$$\left.\frac{\partial E}{\partial \theta_i}\right|_{\theta_i=0} = \left[\frac{\partial}{\partial \theta_i}\left\langle \psi^{(n)} \left| e^{-\theta_i A_i} H e^{\theta_i A_i} \right| \psi^{(n)} \right\rangle\right]\Bigg|_{\theta_i=0}$$
$$= \left\langle \psi^{(n)} \left| [H, A_i] \right| \psi^{(n)} \right\rangle$$



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Gates in ADAPT

- Fermionic single and double excitations
  - Chemical motivation -> Good convergence
- Qubit Excitation Based (QEB) operators (only Jordan-Wigner)
  - Remove Z-chains -> More hardware-efficient in all-to-all connectivity
  - Negligible effect on convergence for small molecules
- qubit-ADAPT
  - Split generators into separate terms
  - More parameters, but more hardware-efficient
  - Breaks symmetries more

$$A_{ik}(\theta) = e^{\theta T_{ik}} = \exp\left[\theta(a_i^\dagger a_k - a_k^\dagger a_i)\right] \quad \text{and}$$

$$A_{ijkl}(\theta) = e^{\theta T_{ijkl}} = \exp\left[\theta(a_i^\dagger a_j^\dagger a_k a_l - a_k^\dagger a_l^\dagger a_i a_j)\right]$$

JW-mapping

$$A_{ik}(\theta) = \exp\left[\mathrm{i}\frac{\theta}{2}(X_iY_k - Y_iX_k)\prod_{r=i+1}^{k-1} Z_r\right] \quad \text{and}$$

$$A_{ijkl}(\theta) = \exp\left[\mathrm{i}\frac{\theta}{8}(X_iY_jX_kX_l + Y_iX_jX_kX_l + Y_iY_jY_kX_l + Y_iY_jX_kY_l\right.$$
$$\left.-X_iX_jY_kX_l - X_iX_jX_kY_l - Y_iX_jY_kY_l - X_iY_jY_kY_l)\prod_{r=i+1}^{j-1} Z_r \prod_{r'=k+1}^{l-1} Z_{r'}\right].$$

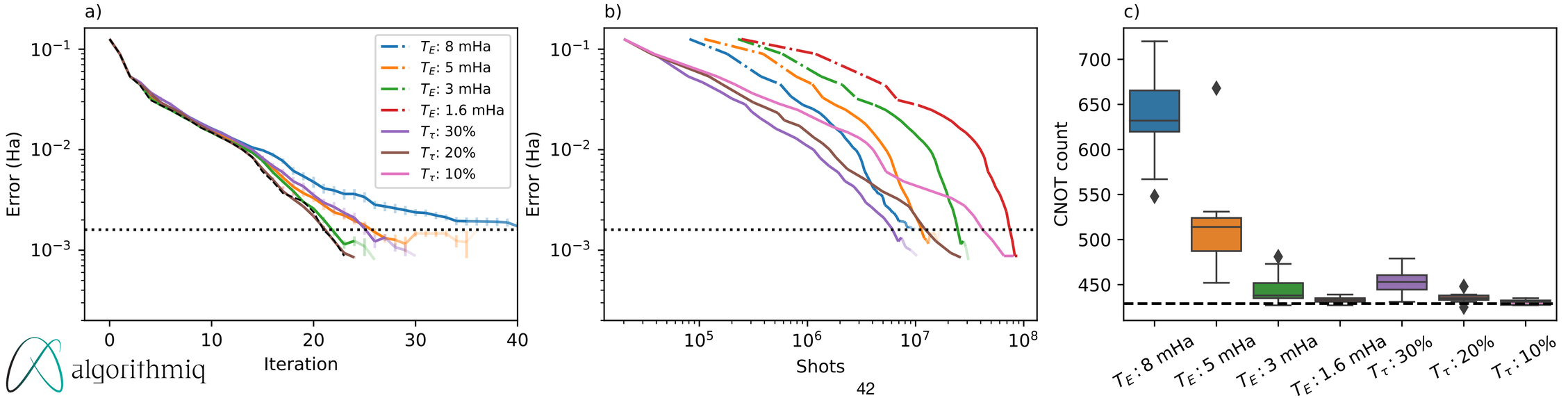
$$A_{ijkl,1}(\theta_1) = \exp(i\theta_1 X_iY_jX_kX_l)$$
$$A_{ijkl,2}(\theta_2) = \exp(i\theta_2 Y_iX_jX_kX_l)$$
$$\vdots$$

Main limitation of ADAPT

Measurement overhead

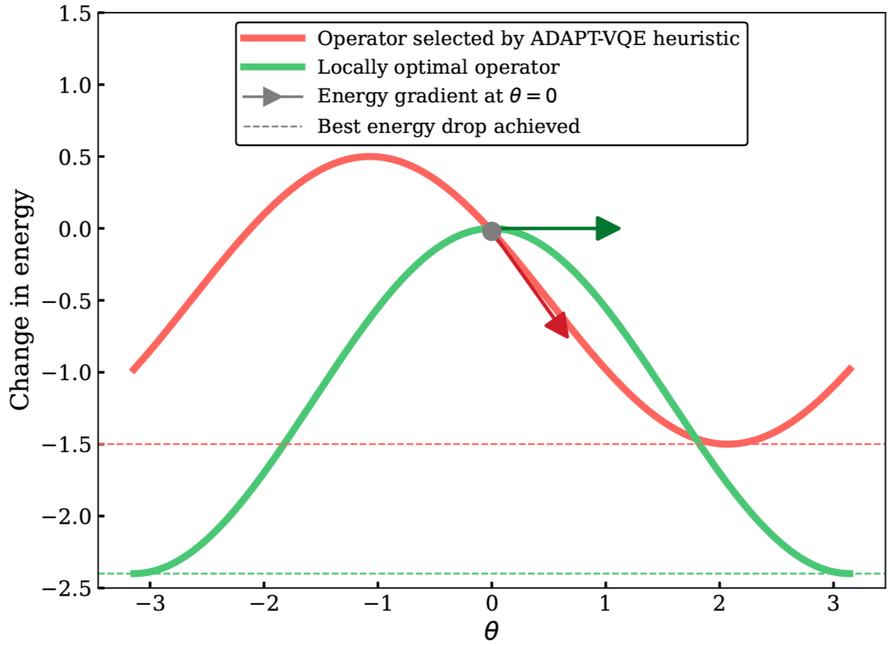
- To find the operator with the highest gradient, one needs to measure a commutator for each operator in the operator pool
- IC-POVMs solve the problem. Just measure the energy and use the same measurement data for the evaluations of the commutators.

N2 1.089Å



Selection criteria

- Gradient selection is robust but not necessarily the most efficient
  - The gradient is measured at parameter 0.0, so low gradient doesn’t necessarily mean small effect on energy.
  - Alternatively, select the operator which lowers the energy the most
- Use overlap selection and optimisation if the target state or an approximation of it is known
  - Not easily implemented on a quantum computer



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