Microsoft ETH zürich

Quantum computing enhanced computational catalysis

arXiV:2007.14460

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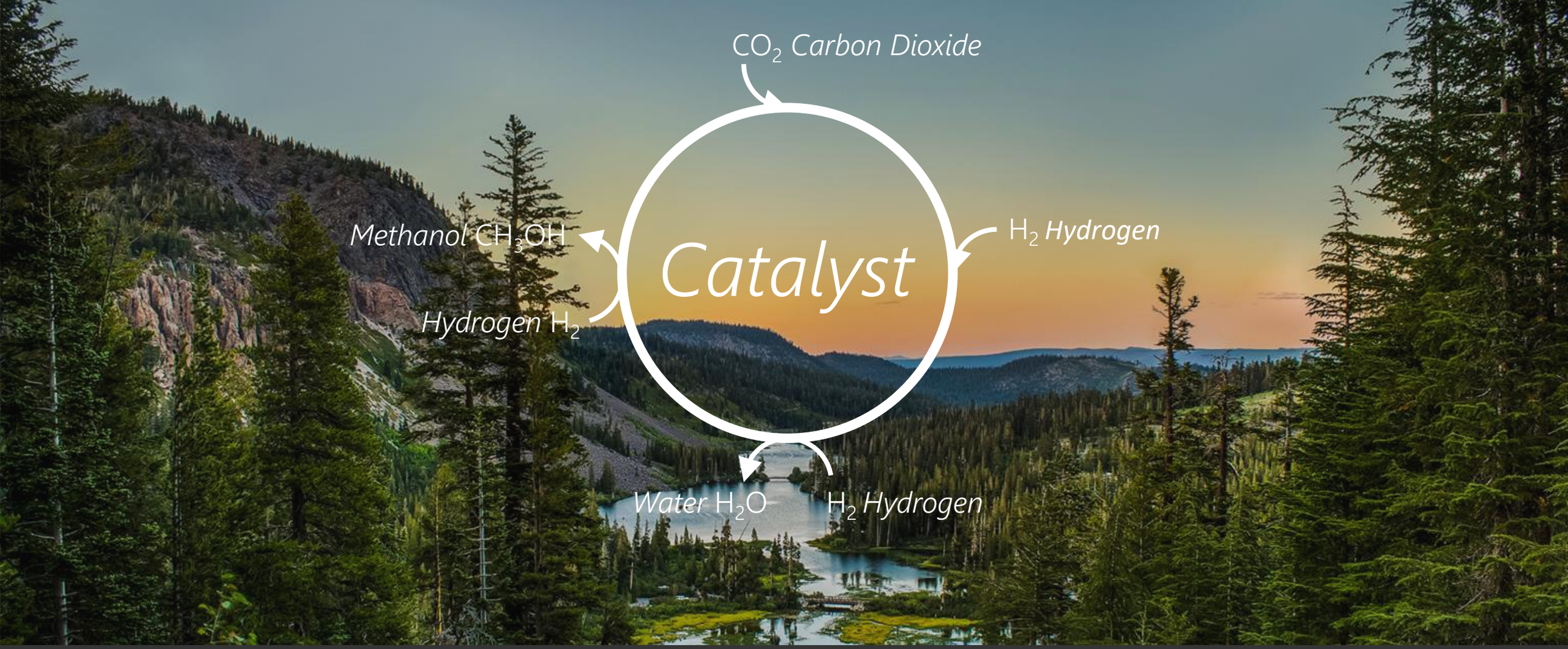
Quantum Information Processing 2021



Understanding Carbon Dioxide Fixation

Outline

1. Requirements of computational catalysis on quantum algorithms[1]

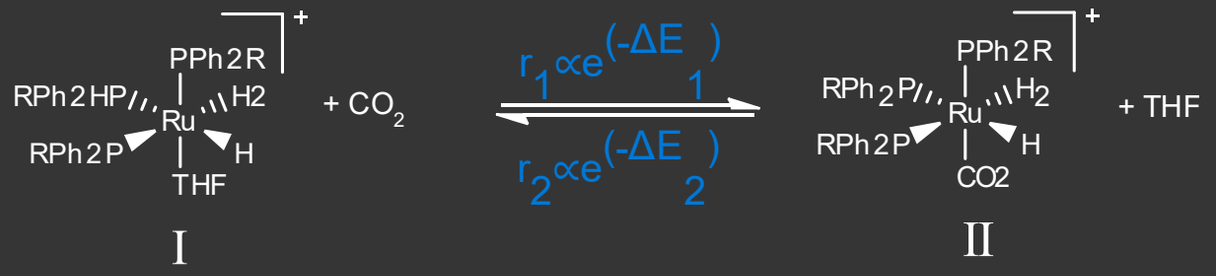
2. Obtaining a suitable input for quantum phase estimation

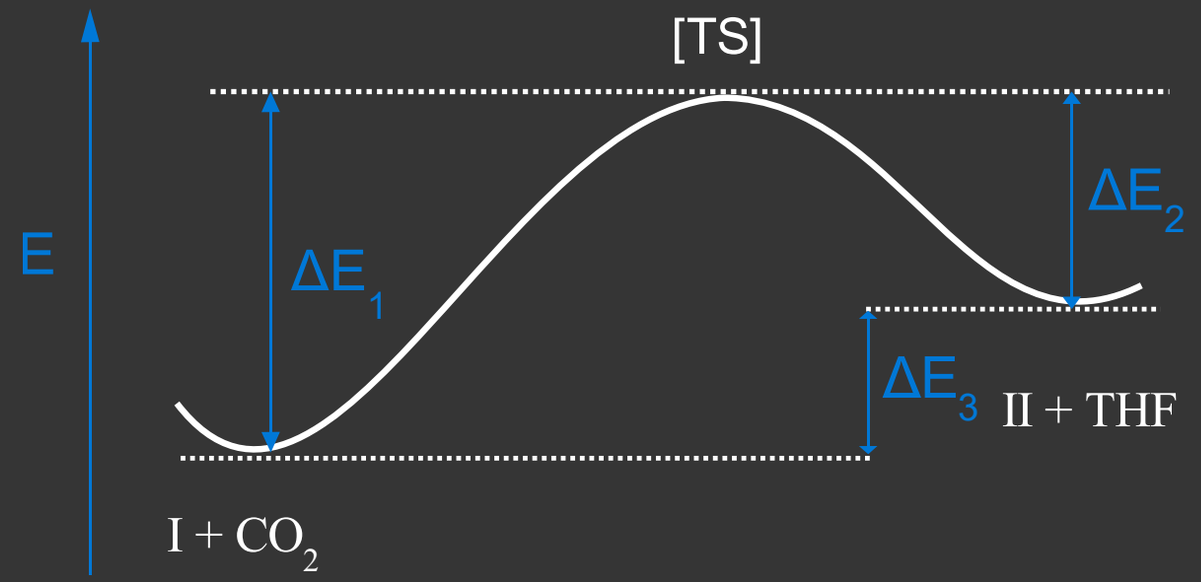
3. Low-rank approximation algorithm & resource estimation[2]

[1]PNAS, 114 (2017), 755.

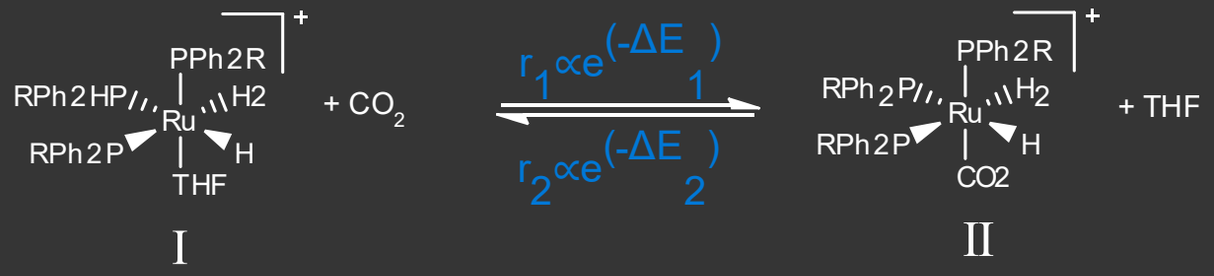
[2]arXiV:2007.14460 (this work).

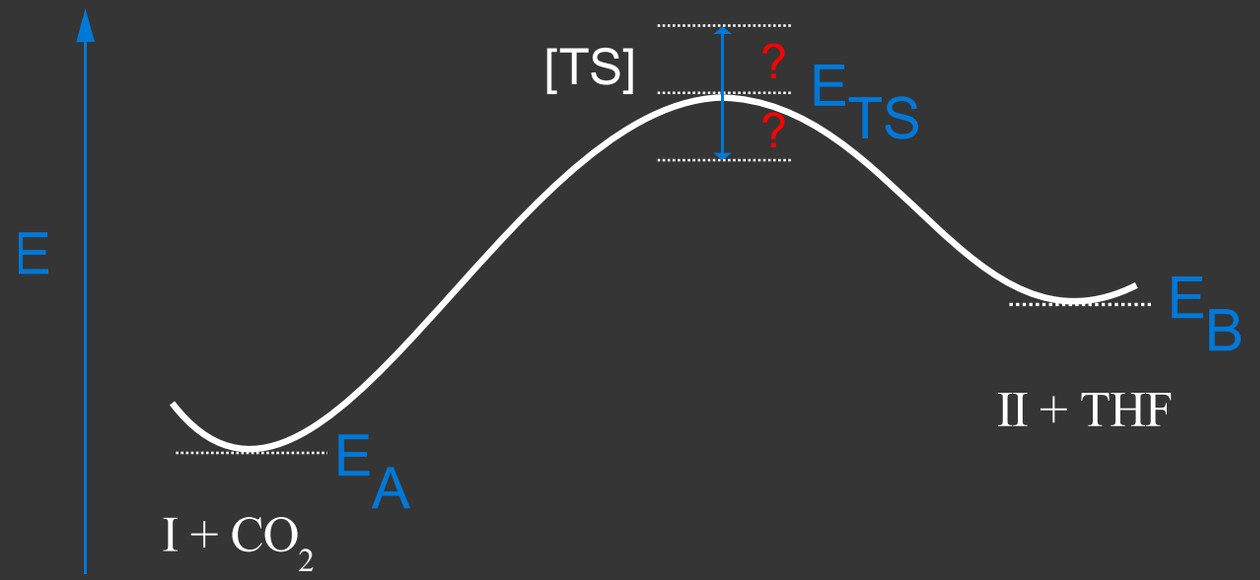
Relative molecular energies determine stability and reactivity



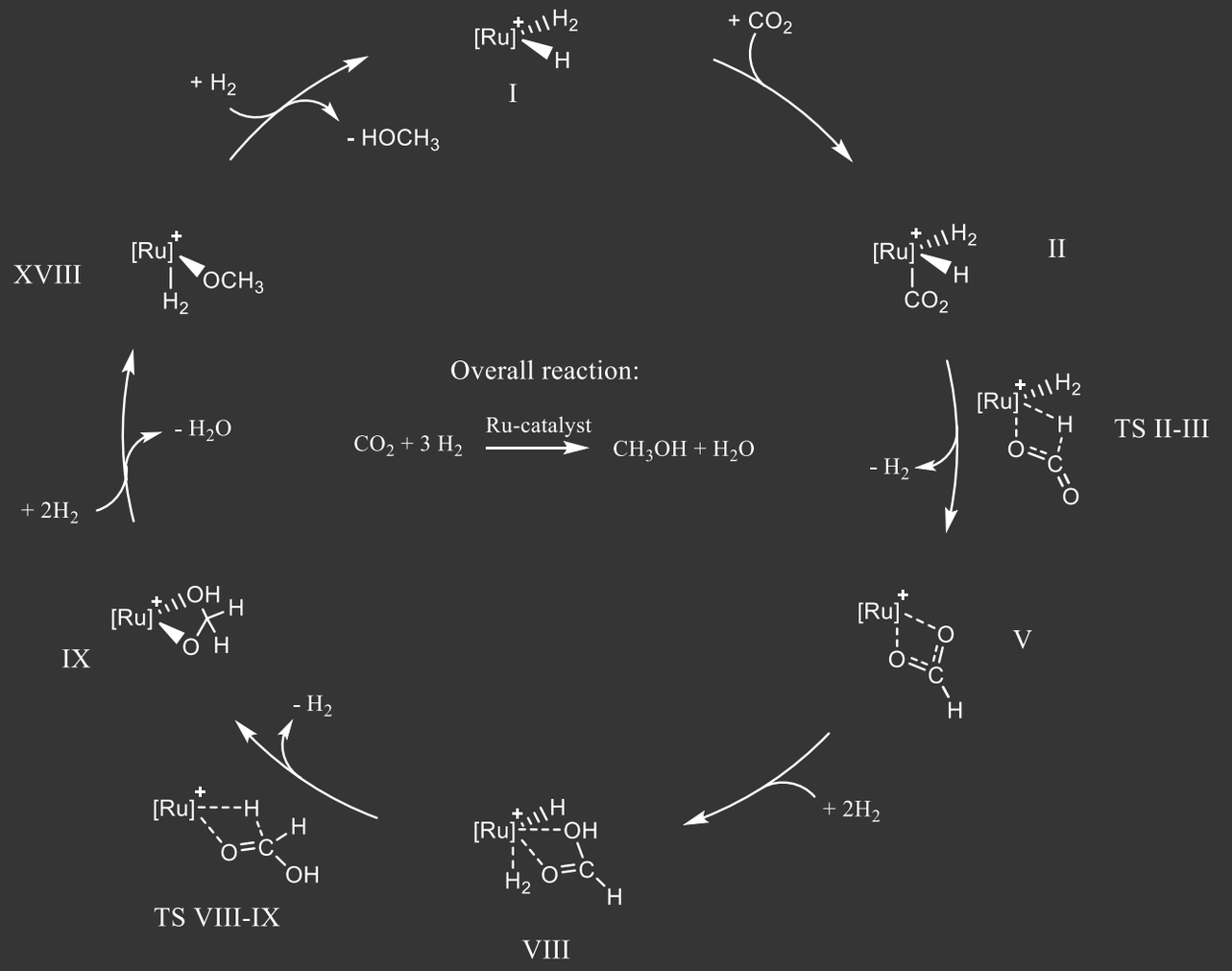


Relative molecular energies determine stability and reactivity





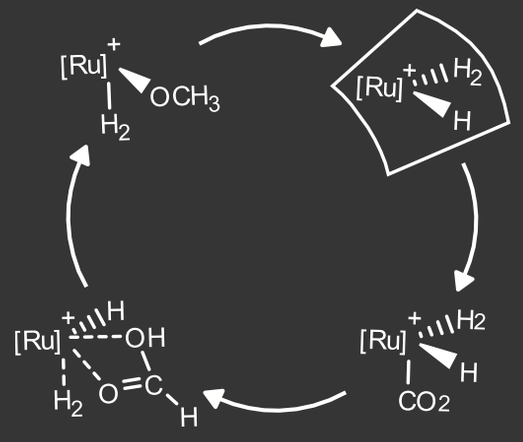
From molecular reactivity to mechanism elucidation



Chem. Sci., 6 (2015), 693.

Iterative refinement of reaction mechanism hypothesis

Structure generation & optimization



Classical computation

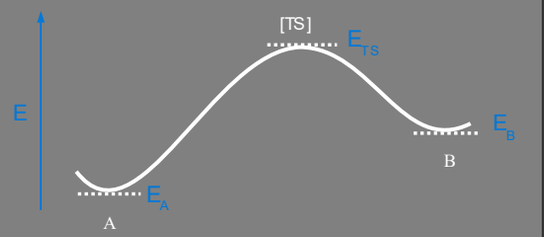
Obtain chemically faithful representation

Kinetic modeling

Add corrections

Quantum computer

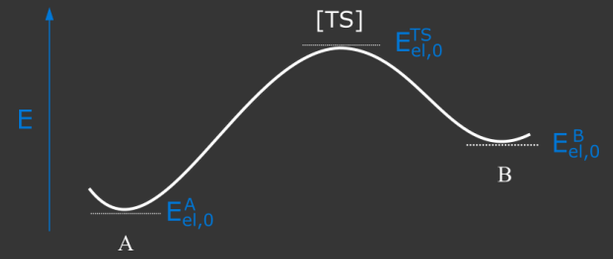
Compute accurate & reliable electronic energies



Requirement on computational energy determination

We need to obtain energies of **multiple structures** with **high accuracy** (1 mHa) and **reliable error estimates** to be able to answer chemical questions

Solving the electronic Schrödinger equation to assign energies



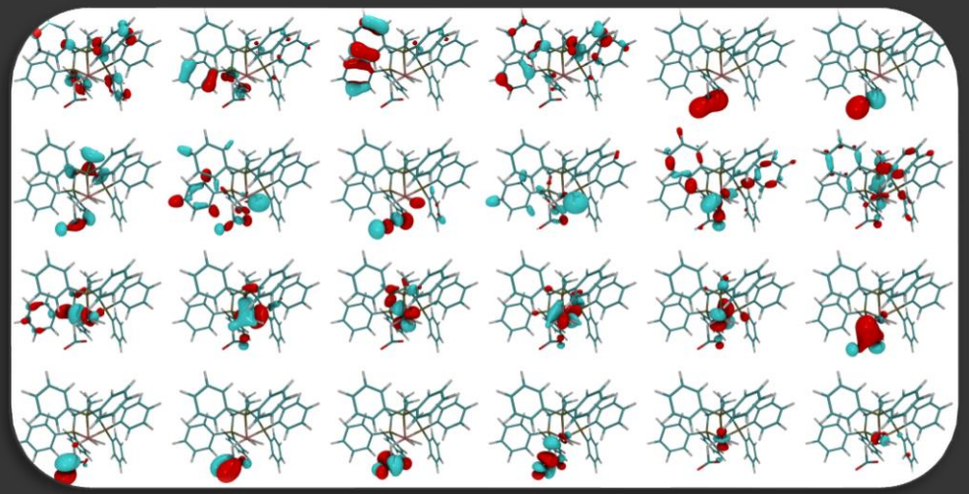
$$H^A_{el}\,|\Psi_n\rangle = E^A_{el,n}\,|\,\Psi_n\rangle$$

$$H^A_{el} = T_e + V_{eN} + V_{ee} + V_{NN}$$

$$= \sum_i \left(-\frac{\nabla_i^2}{2}\right) - \sum_{iJ} \frac{Z_J}{|\boldsymbol{x}_i - \boldsymbol{R}_J|} + \frac{1}{2}\sum_{ij} \frac{1}{|x_i - \boldsymbol{x}_j|} + \frac{1}{2}\sum_{IJ} \frac{Z_I Z_J}{|\boldsymbol{R}_I - \boldsymbol{R}_J|}$$

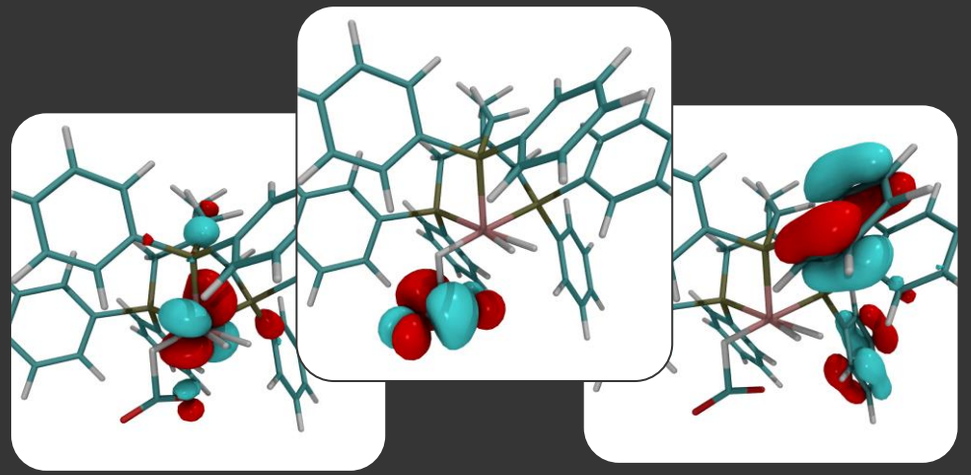
Obtaining a chemically faithful representation

Introduce finite set of basis functions (orbitals)



~1000-2000 orbitals

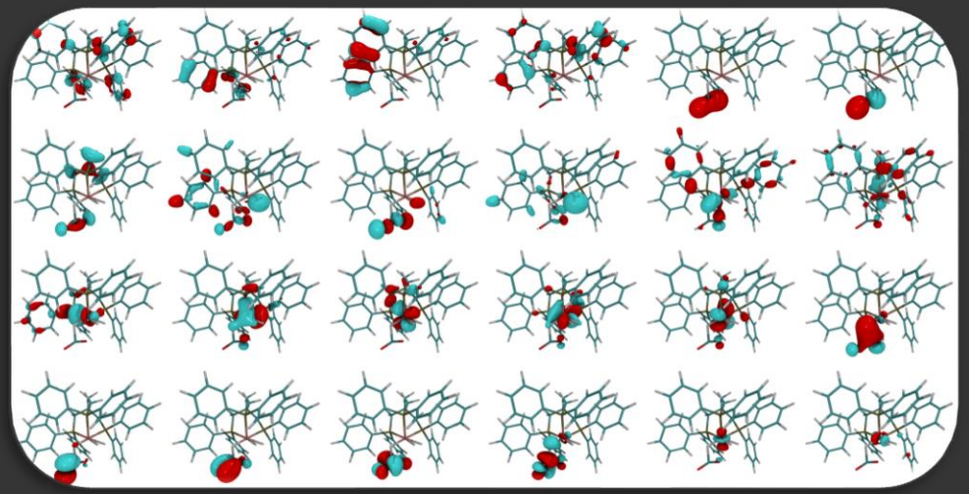
Select orbital subspace providing essential chemistry



~10-60 orbitals

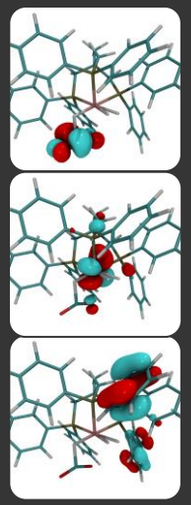
Obtaining a chemically faithful representation

Introduce finite set of basis functions (orbitals)



~1000-2000 orbitals

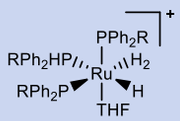
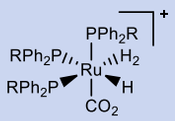
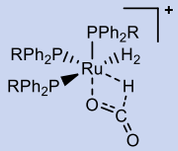
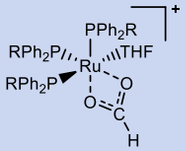
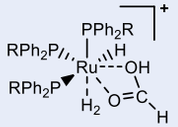
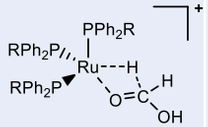
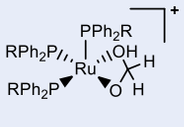
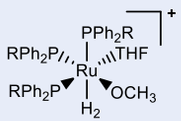
Select orbital subspace providing essential chemistry



1. Compact (~10 orbitals)
2. Broad (~20 orbitals)
3. Ample (~60 orbitals)

~10-60 orbitals

Active space sizes at the classical limit

Catalyst	I	II	II-III	V
				
AS (N_e,N_o)	(48,52)	(70,62)	(74,65)	(68,60)
Catalyst	**VIII**	**VIII-IX**	**IX**	**XVIII**
				
AS (N_e,N_o)	(76,65)	(72,59)	(68,62)	(64,56)

Hamiltonian represented in this basis set choice

$$H = \sum_{pq,\sigma} h_{pq} a^{\dagger}_{(p,\sigma)} a_{(p,\sigma)} + \frac{1}{2} \sum_{pqrs,\sigma\rho} h_{pqrs} a^{\dagger}_{(p,\sigma)} a^{\dagger}_{(q,\rho)} a_{(r,\rho)} a_{(q,\sigma)}$$

$$h_{pq} = \int \phi_p{}^*(x_1) \left(-\frac{\nabla_1^2}{2} - \sum_I \frac{Z_I}{|x_1 - R_I|} \right) \phi_q(x_1)\, \mathrm{d}^3 x_1$$

$$h_{pqsr} = \int \phi_p^*(x_1) \phi_q(x_1) \left(\frac{1}{|x_1 - x_2|} \right) \phi_r^*(x_2) \phi_s(x_2) \mathrm{d}^3 x_1 \mathrm{d}^3 x_2$$

→ Input to Quantum Phase Estimation

Quantum phase estimation

Unitary $U|\psi_j\rangle = e^{iE_j/\alpha}|\psi_j\rangle$

- Estimate E_j to error σ
- E.g. time-evolution $U = e^{-iH/\alpha}$

- Queries to U: $\sim \frac{\alpha\pi}{\sigma}$
  - $\times \frac{1}{2}$ by controlling on $|0\rangle\langle 0| \otimes U^\dagger + |1\rangle\langle 1| \otimes U$

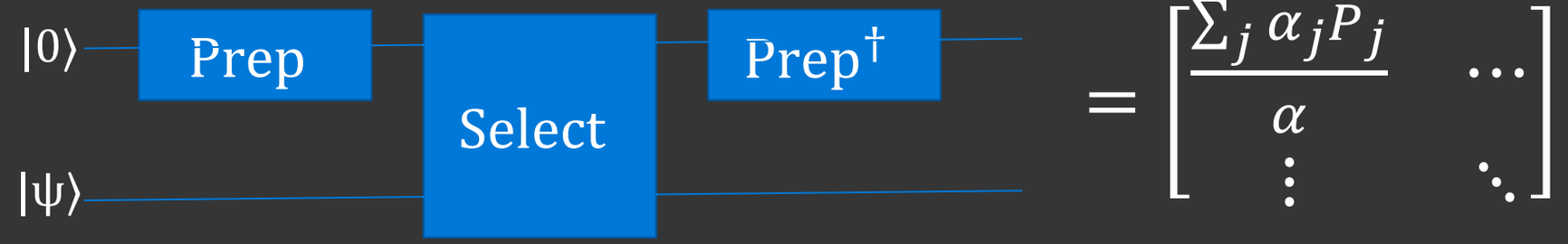
Qubitization[1]

Idea: Circuit for $e^{i \sin^{-1}(H/\alpha)}$ is more efficient than time-evolution

- Encodes same spectral information as e^{-iHt}
- No approximation error; different cost tradeoff

Subroutine: Block-encode H/α into unitary $U = \begin{bmatrix} H/\alpha & \cdots \\ \vdots & \ddots \end{bmatrix}$

LCU Lemma [Childs, Wiebe 2012], [Berry, Childs, Kothari 2015]



[1]Hamiltonian simulation by qubitization, Low, Chuang Quantum 3, 163 (2019)

Compressed representation

Overall cost: $\sqrt{\#\text{ terms}} \times \frac{\alpha\pi}{\sigma}$

Low-rank (data ↓) & partially diagonalized (α ↓)

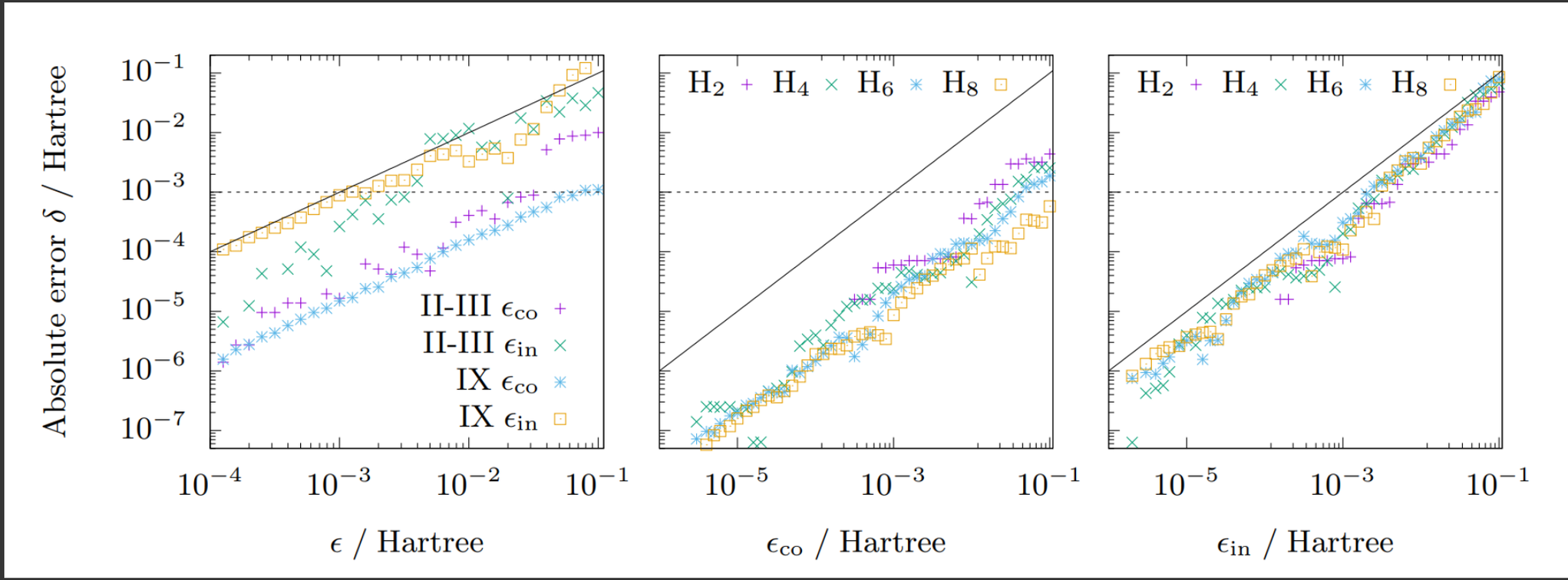
	Hamiltonian $\sum_{pq} h_{pq} a_p^\dagger a_q +$	# terms	$\alpha = \alpha_{\text{free}} + O(\cdot)^*$
Unfactorized	$\sum_{pqrs} h_{pqrs} a_p^\dagger a_q a_r^\dagger a_s$	$O(N^4)$	$\sum_{pqrs} \lvert h_{pqrs} \rvert$ ~10,600
Single-factorized	$\sum_{r\in[R]} \left(\sum_{pq} L_{pq}^{(r)} a_p^\dagger a_q\right)^2$	$O(RN^2)$	$\sum_r \sum_{pq} \left\lvert L_{pq}^{(r)} \right\rvert^2$ ~42,200
Double-factorized	$\sum_{r\in[R]} \left(\sum_{j\in[\Xi],p,q} \lambda_j^{(r)} u_p^{(r,j)} u_q^{(r,j)}\, a_p^\dagger a_q\right)^2$	$O(RN\Xi)$	$\sum_r \sum_{pq} \left\lvert \lambda_j^{(r)} u_p^{(r,j)} u_q^{(r,j)} \right\rvert^2$
Double-factorized (Givens rotated)	$\sum_{r\in[R]} G^{(r)} \left(\sum_{j\in[\Xi]} \lambda_j^{(r)} n_j\right)^2 G^{(r)\dagger}$		$\sum_r \sum_{pq} \left\lvert \lambda_j^{(r)} \right\rvert^2$ ~570

↳ Sum of squares of free-fermion Hamiltonians

*Ruthenium catalyst configuration VIII with 130 spin-orbitals

Improved truncation

Triangle inequality ($\epsilon_{\mathbf{co}}$) vs. sum-of-squares ($\epsilon_{\mathbf{in}}$)



Block-encoding strategy

1. Input: B.E. of free fermion $H_r = \sum_{pq} L_{pq}^{(r)} a_p^\dagger a_q = G^{(r)} \left(\sum_{j \in [\Xi]} \lambda_j^{(r)} n_j \right) G^{(r)\dagger}$

$$U_r = \begin{bmatrix} H_r/\lambda_r & \cdots \\ \vdots & \ddots \end{bmatrix}$$

2. Square w/ 2 queries (2nd Chebyshev polynomial $T_2(x) = 2x^2 - 1$)

$$V_r = \begin{bmatrix} T_2(H_r/\lambda_r) & \cdots \\ \vdots & \ddots \end{bmatrix}$$

3: Interacting fermion Hamiltonian by LCU

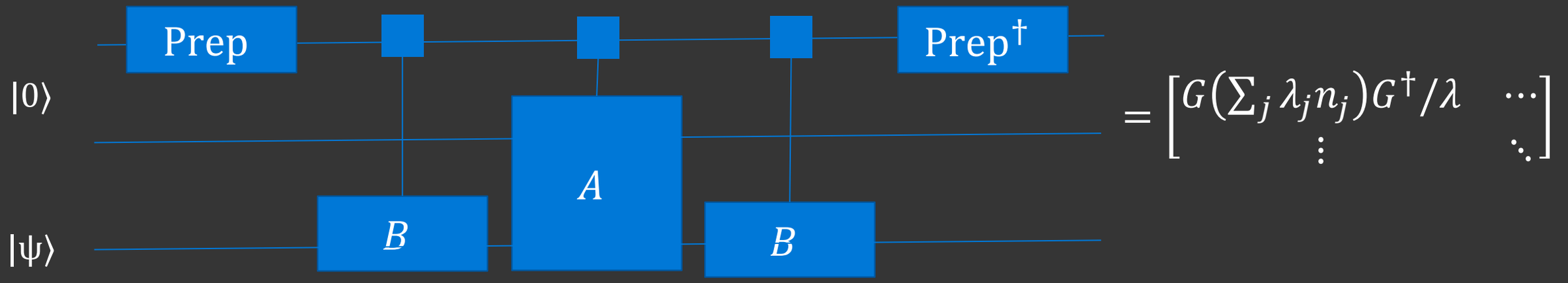
$$V = \begin{bmatrix} \dfrac{\sum_r \lambda_r^2 T_2(H_r/\lambda_r)}{\sum_r \lambda_r^2 /2} & \cdots \\ \vdots & \ddots \end{bmatrix}$$

B.E. of free-fermion Hamiltonian

Idea: LCU of rotated number operators

$$G\left(\sum_j \lambda_j n_j\right)G^\dagger = \sum_j \lambda_j (G_j n_j G_j^\dagger)$$

1. Block-Encode $A = \sum_j |j\rangle\langle j| \otimes \begin{bmatrix} \sum_j n_j & \cdots \\ \vdots & \ddots \end{bmatrix}$
2. Multiplexed Givens rotation $B = \sum_j |j\rangle\langle j| \otimes G_j$
3. Prepare quantum state $\sum_j \sqrt{\lambda_j/\lambda}|j\rangle$

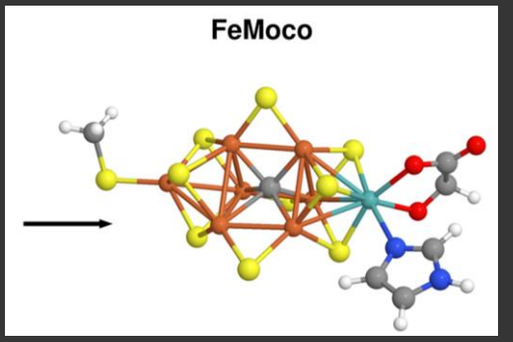


Key block-encoding techniques

1. Ancilla-assisted dim-$R\Xi$ state preparation
   - $O(\sqrt{R\Xi})$ Non-Clifford gates vs $O(R\Xi)$ Low, Kliuchnikov, Schaffer arXiv:1812.00954

2. Local vs. global Multiplexed Givens rotation
   - $\sum_{r\in[R],j\in[\Xi]} |r\rangle\langle r| \otimes |j\rangle\langle j| \otimes G_j^{(r)}$ w/ $O\left(\sqrt{R\Xi} + N\right)$ Non-Clifford gates
   - vs $\sum_{r\in[R]} |r\rangle\langle r| \otimes G$ w/ $O\left(\sqrt{RN^2} + N\right)$ Non-Clifford gates

Resource estimates

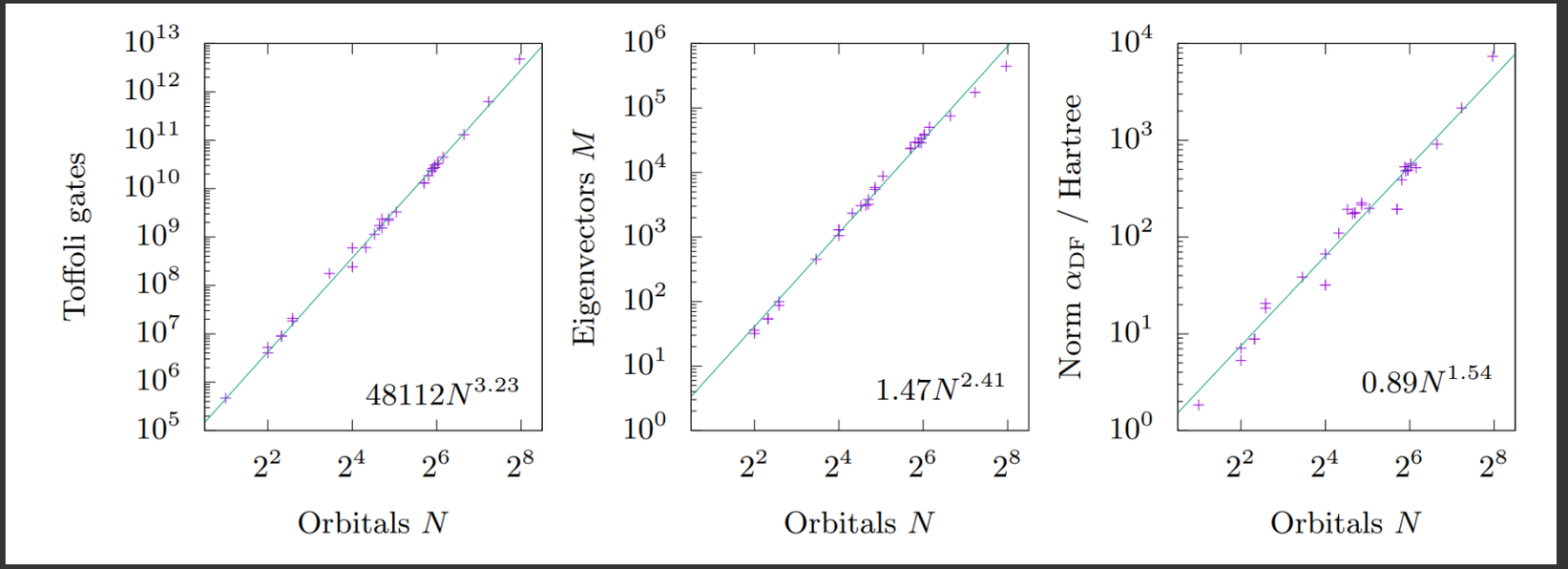


FeMoco Catalyst 1mHa precision	Reiher et al. 1605.03590		Low et al. 1904.01131	Berry et al. 1902.02134	von Burg et al. 2007.14460
Method	Trotter-Suzuki (Provable)	Trotter-Suzuki (Optimistic)	Qubitization (Provable)	Qubitization (Empirical)	Qubitization (Empirical)
T-gate cost	10^{21}	10^{14}	10^{14}	10^{12}	10^{10}

10^5 yrs @100Mhz

1 week @100kHz

Carbon capture
Leitner catalyst



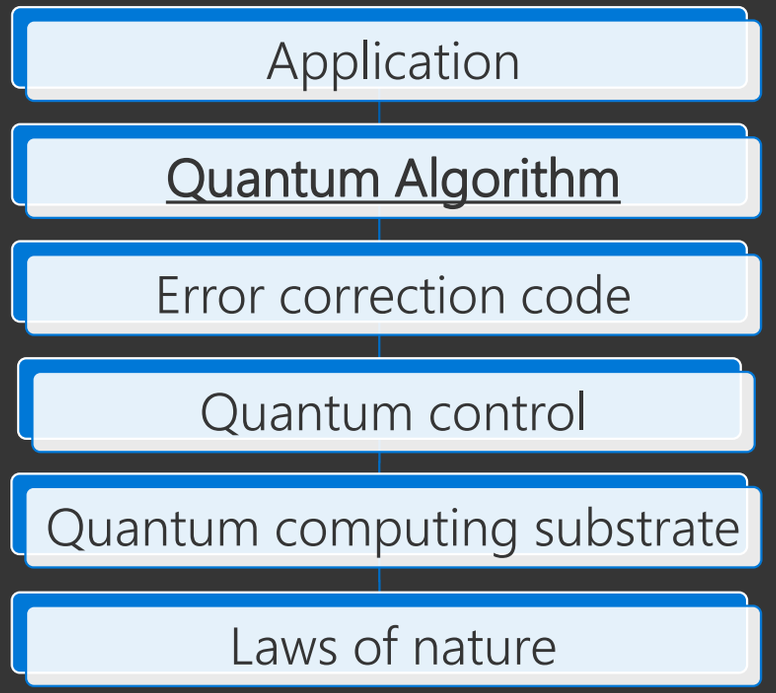
Towards useful quantum computing

Progress in Algorithms beats Moore's Law

Linear programming benchmark (1988-2003)[1]:
~1000x hardware, ~43000x algorithms

Physical qubits (2016-2020):
~10x from 5 to 53

Quantum alg. for Nitrogenase (2016-2020)[2,3]:
~10000x



Useful applications: ~100k physical qubits

[1]Report to the president and congress: Designing a digital future, 2010, pg 71
[2]Quantum computing enhanced computational catalysis, von Burg et al., arXiv:2007.14460
[2]Even more efficient quantum computations of chemistry through tensor hypercontraction, Lee et al., arXiv:2011.03494