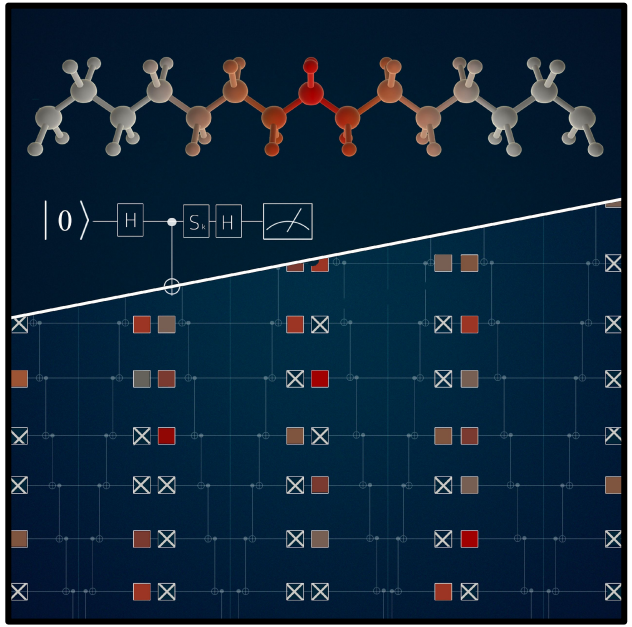
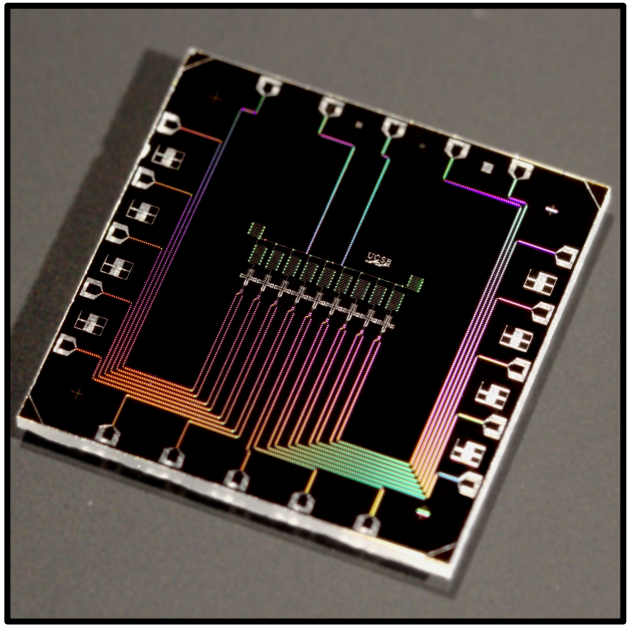
# Simulating Chemistry with Realistic Quantum Computers




Ryan Babbush
June, 2018

Google

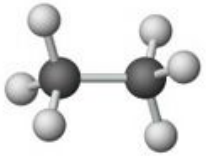


# The world is made of atoms. Chemistry arises from interactions of their electrons.

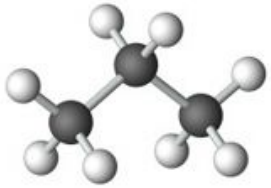
**Systems of interacting electrons are near ideal application for quantum simulation**

They're classically hard to simulate but extremely important

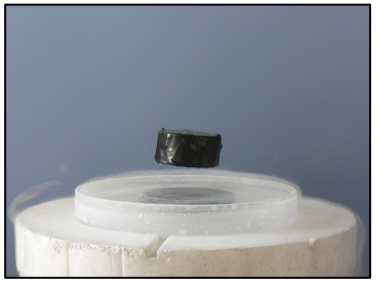
CPU seconds



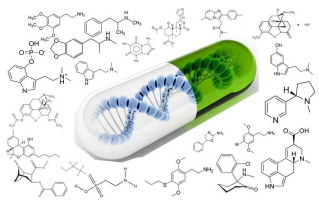
CPU minutes



CPU days



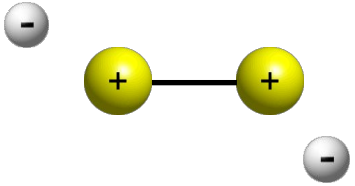
The prospect of more efficient simulations is both scientifically exciting and valuable



# The molecular electronic structure problem

**Goal is to solve for the energy of a molecule:**

$$H = \hat{T}_{\text{nuc}} + \hat{T}_{\text{elec}} + \hat{V}_{\text{nuc-nuc}} + \hat{V}_{\text{nuc-elec}} + \hat{V}_{\text{elec-elec}}$$

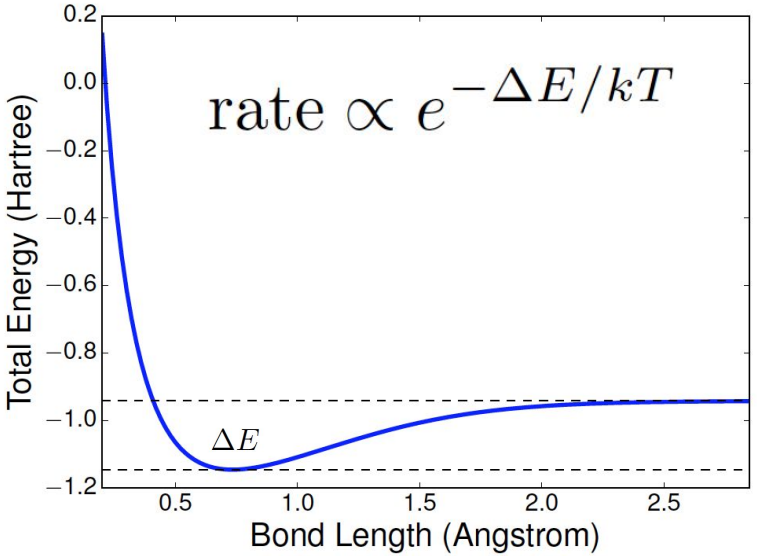



**Clamp nuclei under Born-Oppenheimer approximation and solve for energy surface**

**Energy surfaces allow us to understand reactions**
Need chemical accuracy (1 kcal/mol) for rates

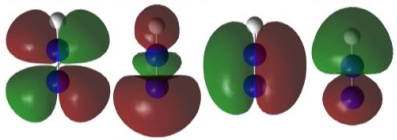
**Such accuracy is often classically intractable**
Especially for systems with strong correlation

**Should only use quantum comp for active space**



# The second quantized Galerkin discretization

**To represent wavefunctions on computer one must discretize space (confine to a basis)**



$$|\psi\rangle = a_1 |0011\rangle + a_2 |0101\rangle + a_3 |1001\rangle + a_4 |0110\rangle + a_5 |1010\rangle + a_6 |1100\rangle$$

If η electrons confined to N locations, there are (N choose η) configurations!

**Galerkin discretization in MOs leads to O($N^4$) Hamiltonian terms at all sizes**

$$H = \frac{1}{2} \sum_{pqrs} \langle pq| \hat{V}_{\text{elec-elec}} |sr\rangle \, a_p^\dagger a_q^\dagger a_r a_s + \sum_{pq} \langle p| \hat{T}_{\text{elec}} + \hat{V}_{\text{nuc-elec}}(R) |q\rangle \, a_p^\dagger a_q$$

**2nd quantization = antisymmetry in operators**

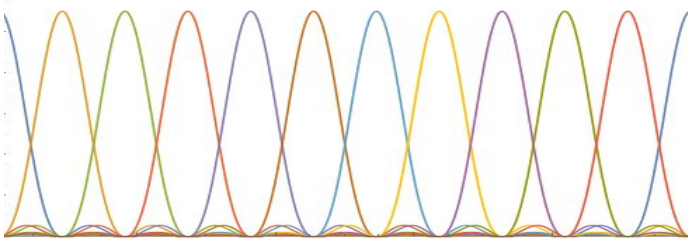
$$\{a_p, a_q^\dagger\} = a_p a_q^\dagger + a_q^\dagger a_p = \delta_{pq} \mathbb{1}$$
$$\{a_p, a_q\} = \{a_p^\dagger, a_q^\dagger\} = 0$$

**Jordan-Wigner transformation:**

$$a_p^\dagger = (X_p - iY_p) \, Z_{p-1} Z_{p-2} \cdots Z_0$$
$$a_p = (X_p + iY_p) \, Z_{p-1} Z_{p-2} \cdots Z_0$$

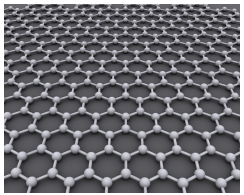
# Basis sets with quadratically fewer terms (1706.00023)

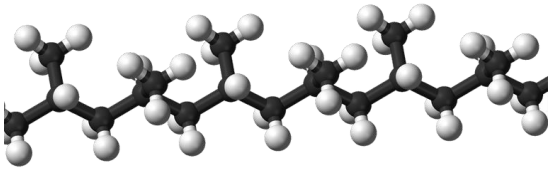
$$H = \sum_{pq} T_{pq} a_p^\dagger a_q + \sum_p U_p n_p + \sum_{p \neq q} V_{pq} n_p n_q$$



**Derived from unitary rotation of plane wave basis**

Good for periodic systems!





**Need a constant factor more plane waves than Gaussians for single-molecules**

There are bases with similar properties that are better for small molecules (e.g. 1709.08059)

**After mapping to qubits the Hamiltonian is highly structured with $O(N^2)$ terms**

$$H = \sum_{p \neq q} \tilde{T}_{pq} \left( X_p Z_{p+1} \cdots Z_{q-1} X_q + Y_p Z_{p+1} \cdots Z_{q-1} Y_q \right) + \sum_{p \neq q} \tilde{V}_{pq} Z_p Z_q + \sum_p \tilde{U}_p Z_p$$

Can simulate Trotter steps on linear array in linear depth (see 1711.04789)

# Molecular eigenstates from phase estimation (quant-ph/0604193)

**The key to this approach is leveraging the ability to prepare good initial states**

$$H\left|k\right\rangle = E_k\left|k\right\rangle \qquad \left|\left\langle\psi|k\right\rangle\right|^2 = \text{Something reasonable}$$

**Then, construct a quantum circuit encoding spectrum of *H***

$$U = e^{-iHt} \qquad U = e^{i\arccos(H/\lambda)}$$

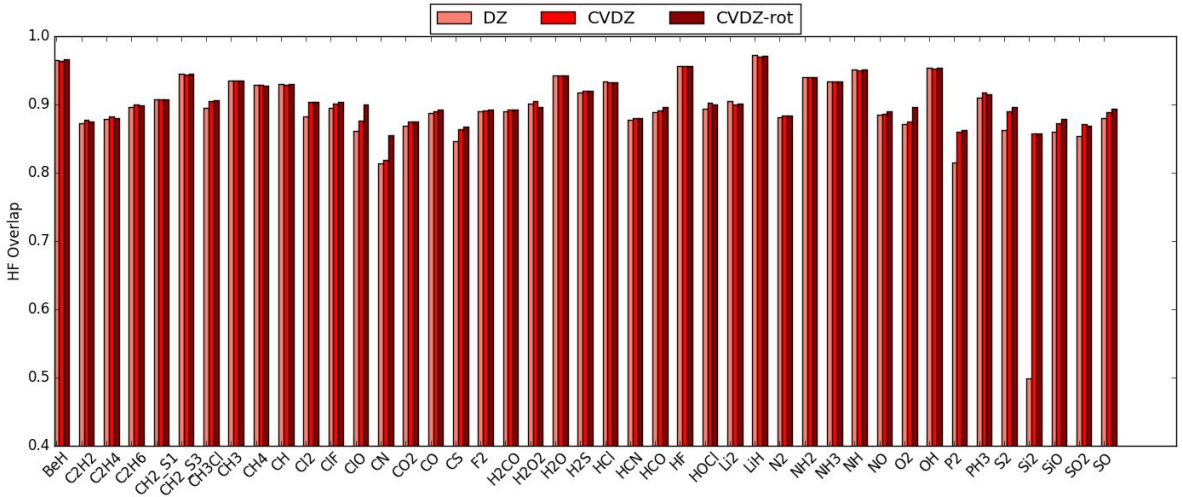
**Apply the quantum circuit to accumulate phases on eigenstates of the Hamiltonian**

$$e^{-iHt}\left|\psi\right\rangle = \sum_{k=0}^{2^N} \underbrace{\left\langle k|\psi\right\rangle}_{a_k} e^{-i\theta_k(t)}\left|k\right\rangle \qquad \theta_k(t) = E_k t$$

**Use quantum phase estimation algorithm to measure $\theta_k$ with probability $a_k^2$**

**O(1/$\epsilon$) coherent repetitions of *U* for phase estimation *will* require error-correction**

# Preparing initial states with strong support

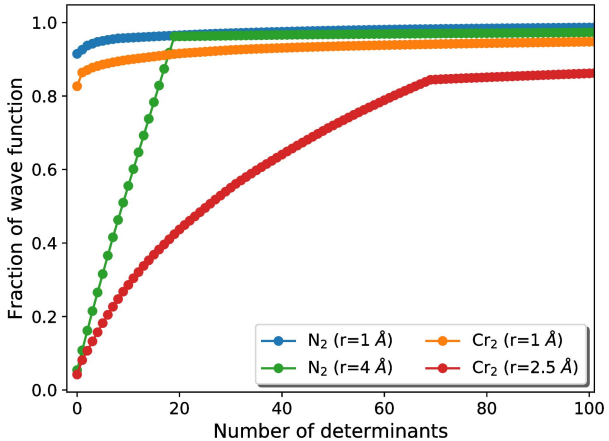



**data to appear soon in paper by**
Norm Tubman, Carlos Mejuto-Zaera, Jeffrey Epstein, Diptarka Hait, Daniel Levine, William Huggins, Zhang Jiang, Jarrod McClean, Ryan Babbush, Martin Head-Gordon, and K. Birgitta Whaley

ASCI method converges support of most important configurations much faster than total energy

Some molecules, especially out of equilibrium, cannot be described by one configuration

Can prepare superposition of L arbitrary configurations at O(L) T gate complexity

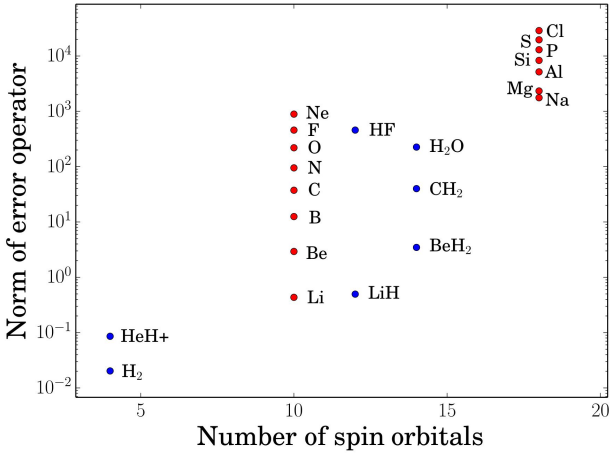


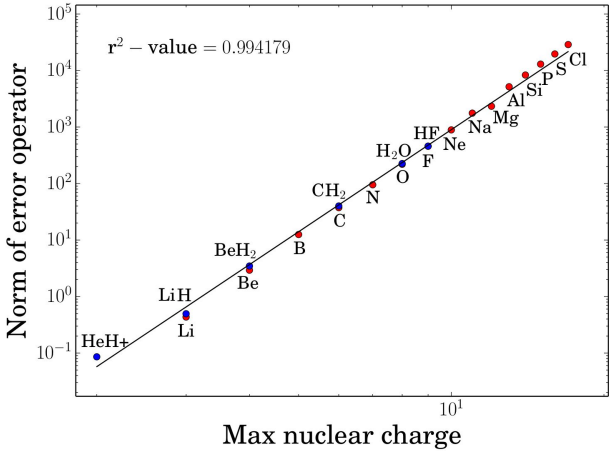
# Encoding electronic spectra in quantum circuits with Trotter

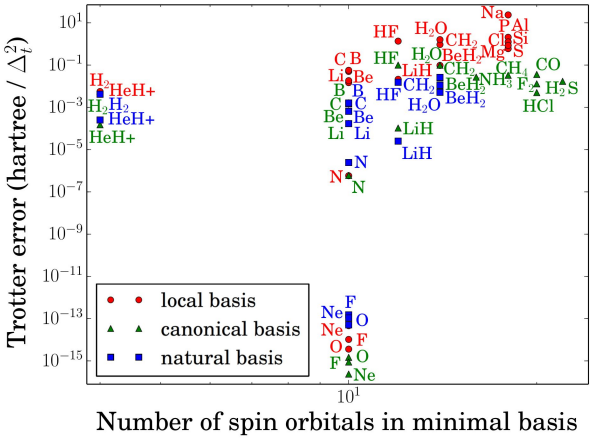
**Time-evolution based on Trotterization scales polynomially in number of terms and 1/$\epsilon$**

$$H = \sum_{\ell=0}^{L-1} \alpha_\ell H_\ell \qquad\qquad e^{-iHt} \approx \left( \prod_{\ell=0}^{L-1} e^{-i\alpha_\ell H_\ell t/\rho} \right)^\rho$$

**Bounds on $\rho$ suggest poor scaling, but are loose / instance specific (1406.4920, 1410.8159)**




**Expect Trotter methods to be used in "unsafe heuristic mode" on first devices**

# Post-Trotter methods: more precise, more interesting, and often faster

**Linear combination of unitary methods (1202.5822), e.g. qubitization (1610.06546), can scale sublinear in both number of Hamiltonian terms and 1/$\epsilon$**

$$H = \sum_\ell \alpha_\ell U_\ell \qquad U|\ell\rangle|\psi\rangle \mapsto |\ell\rangle U_\ell|\psi\rangle \qquad G|0\rangle \mapsto \sum_\ell \sqrt{\frac{\alpha_\ell}{\lambda}}|\ell\rangle$$

With 1 query to *U* and 2 queries to *G* one can *exactly* implement $e^{i \arccos(H/\lambda)}$

**(1506.01020) showed that one can implement *U* and *G* with O(N) gate complexity**
Method relied on on-the-fly integration to compute coefficients - very impractical!

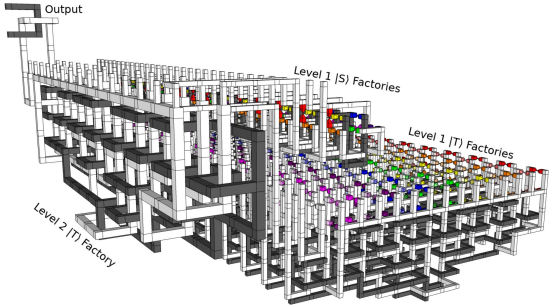
**(1805.03662) showed practical linear gate complexity for plane wave Hamiltonian**
But one must repeat phase estimation $\lambda = \sum_\ell |\alpha_\ell|$ times to extract phase

**(1805.00675) performs simulation in interaction picture where H = T + V**
Instead of $\lambda = \lambda_T + \lambda_V$ queries, one can take either $O(\lambda_T \log(\lambda_V))$ or $O(\lambda_V \log(\lambda_T))$ queries

# T gates: a good cost model for error-correction

**T gates mostly come from arbitrary rotations or Toffolis**
Implementation requires T factory for magic state distillation

**First error-corrected devices will have only one T factory**
State distillation will occur in series, cause bottleneck



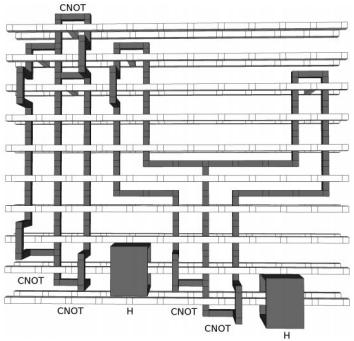

| Year | arXiv | Basis | Algorithm | Oracle T Gates | Queries | Total T Gates |
|---|---|---|---|---|---|---|
| 2005 | quant-ph/0604193 | Arbitrary | Trotterization | $\mathcal{O}(\text{poly}(N/\epsilon))$ | $\mathcal{O}(\text{poly}(N/\epsilon))$ | $\mathcal{O}(\text{poly}(N/\epsilon))$ |
| 2010 | 1001.3855 | Arbitrary | Trotterization | $\mathcal{O}(N^4 \log(1/\epsilon))$ | $\mathcal{O}(\text{poly}(N/\epsilon))$ | $\mathcal{O}(\text{poly}(N/\epsilon))$ |
| 2013 | 1312.1695 | Arbitrary | Trotterization | $\mathcal{O}(N^4 \log(1/\epsilon))$ | $\mathcal{O}(N^6/\epsilon^{1/2})$ | $\mathcal{O}(N^{10} \log(1/\epsilon)/\epsilon^{1/2})$ |
| 2014 | 1407.7863 | Arbitrary | Trotterization | $\mathcal{O}(\sim N^2 \log(1/\epsilon))$ | $\mathcal{O}(N^6/\epsilon^{1/2})$ | $\mathcal{O}(\sim N^8 \log(1/\epsilon)/\epsilon^{1/2})$ |
| 2014 | 1406.4920 | Arbitrary | Trotterization | $\mathcal{O}(N^4 \log(1/\epsilon))$ | $\mathcal{O}(\sim N^2/\epsilon^{1/2})$ | $\mathcal{O}(\sim N^6 \log(1/\epsilon)/\epsilon^{1/2})$ |
| 2014 | 1410.8159 | Arbitrary | Trotterization | $\mathcal{O}(N^4 \log(1/\epsilon))$ | $\mathcal{O}(\sim N/\epsilon^{1/2})$ | $\mathcal{O}(\sim N^5 \log(1/\epsilon)/\epsilon^{1/2})$ |
| 2015 | 1506.01020 | Arbitrary | Taylorization | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}\left(\frac{N^4 \log(N/\epsilon)}{\log\log(N/\epsilon)}\right)$ | $\mathcal{O}(N^5\text{polylog}(N/\epsilon))$ |
| 2016 | 1610.06546 | Arbitrary | Qubitization | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}\left(N^4 + \frac{\log(N/\epsilon)}{\log\log(N/\epsilon)}\right)$ | $\mathcal{O}(N^5\text{polylog}(N/\epsilon))$ |
| 2017 | 1706.00023 | Plane Waves | Taylorization | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}\left(\frac{N^{8/3} \log(N/\epsilon)}{\log\log(N/\epsilon)}\right)$ | $\mathcal{O}(N^{11/3}\text{polylog}(N/\epsilon))$ |
| 2017 | 1711.10460 | Plane Waves | Qubitization | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}(N^{8/3})$ | $\mathcal{O}(N^{11/3}\text{polylog}(N/\epsilon))$ |
| 2018 | 1805.03662 | Plane Waves | Qubitization | $\mathcal{O}(N + \log(1/\epsilon))$ | $\mathcal{O}(N^2)$ | $\mathcal{O}(N^3 + N^2 \log(1/\epsilon))$ |
| 2018 | 1805.00675 | Plane Waves | Interaction Pic. | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}(N\text{polylog}(N/\epsilon))$ | $\mathcal{O}(N^2\text{polylog}(N/\epsilon))$ |

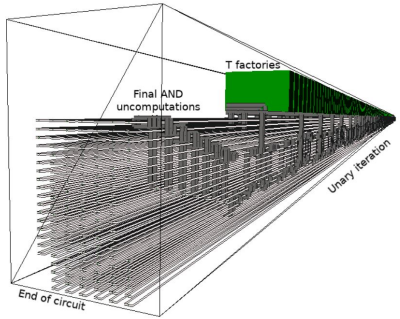
**Asymptotic complexity can be misleading - must count T gates!**

# Finite size T counts for chemistry

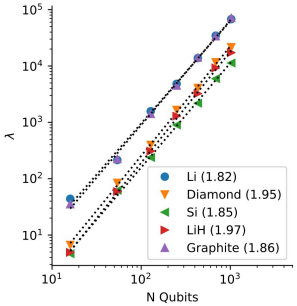
**(1605.03590) estimates $FeS_2$ (108 qubits) requires $10^{15}$ T gates**
Uses Gaussian orbitals + Trotterization with formal $O(N^8)$, empirical $O(N^6)$ scaling

**(1805.03662) uses diagonalizing basis (e.g. plane waves) + qubitization to obtain $O(N^3)$ scaling**
Compiled all bottlenecks to surface code gates both by hand and with automated system




T count is relatively insensitive to nuclear potential for LCU methods



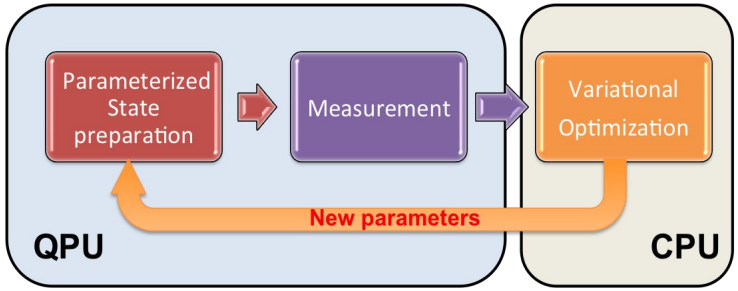

**Layouts limited by existing software tools**
We believe can reduce physical qubits by 10x

**We can use this approach to simulate $FeS_2$**
But we should develop better basis functions

| instance size | | physical qubits | | execution time (hours) | |
|---|---|---|---|---|---|
| spin-orbitals | T gates | $p = 10^{-3}$ | $p = 10^{-4}$ | $p = 10^{-3}$ | $p = 10^{-4}$ |
| 54 | $1.8 \times 10^7$ | $1.4 \times 10^6$ | $3.9 \times 10^5$ | 0.82 | 0.43 |
| 128 | $1.9 \times 10^8$ | $2.4 \times 10^6$ | $8.1 \times 10^5$ | 9.9 | 5.6 |
| 250 | $1.1 \times 10^9$ | $4.4 \times 10^6$ | $1.2 \times 10^6$ | 58 | 30 |
| 1024 | $4.3 \times 10^{10}$ | $2.0 \times 10^7$ | $4.8 \times 10^6$ | $2.7 \times 10^3$ | $1.4 \times 10^3$ |

# Quantum variational eigensolver, the NISQ heuristic (1304.3061)

$$\frac{\langle\varphi(\vec{\theta})|\, H \,|\varphi(\vec{\theta})\rangle}{\langle\varphi(\vec{\theta})|\varphi(\vec{\theta})\rangle} \geq E_0$$




1. ***Cleverly*** **parameterize short quantum circuit with a polynomial number of variables**

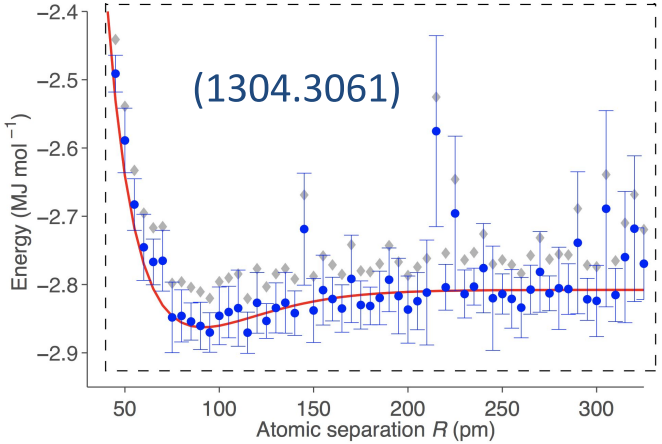
$$U(\vec{\theta}) \equiv U_1(\theta_1)U_2(\theta_2)\cdots U_n(\theta_n)$$

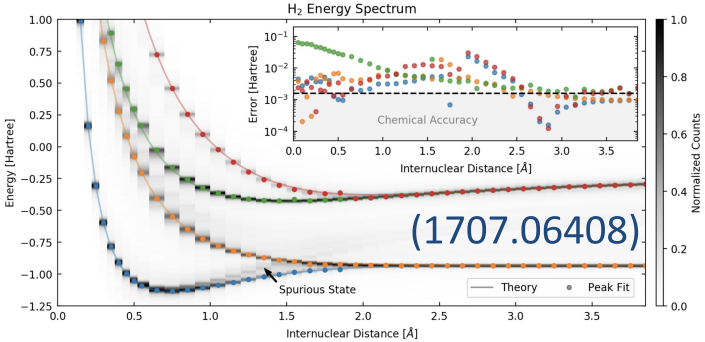
2. **Apply circuit to guess state (e.g. Hartree-Fock state) and measure energy**

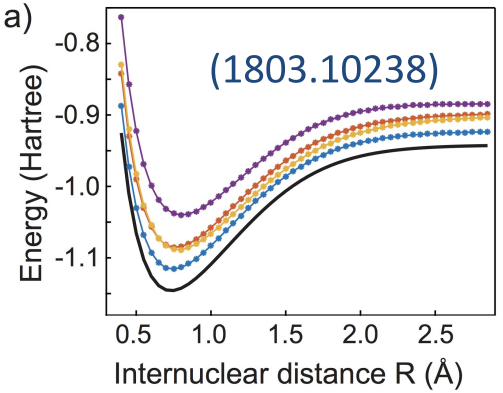
$$|\varphi(\vec{\theta})\rangle \equiv U(\vec{\theta})\,|\phi\rangle \qquad E(\vec{\theta}) = \sum_{\gamma} g_\gamma \,\langle\varphi(\vec{\theta})|\, H_\gamma \,|\varphi(\vec{\theta})\rangle$$

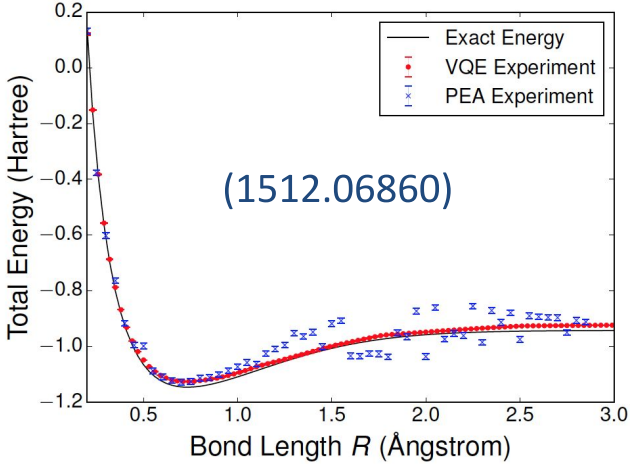
3. **Use classical optimizer (e.g. gradient descent) to suggest new parameters**

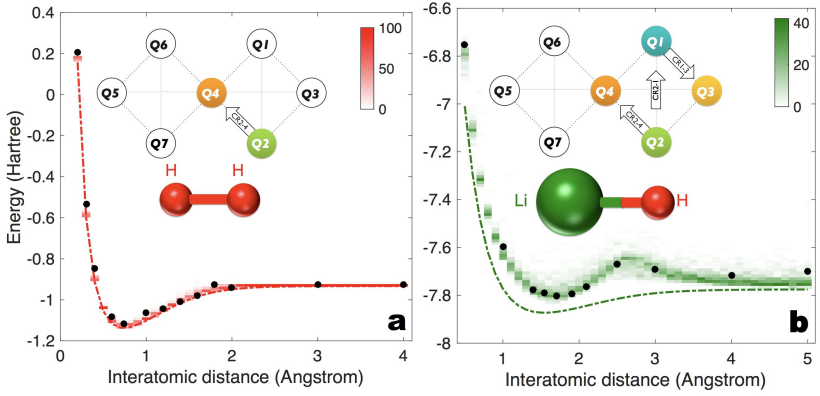
# Experimental data on small molecules



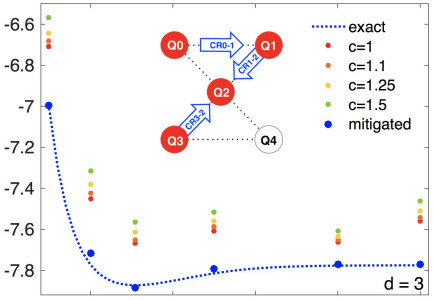




(1704.05018)



# How to parameterize the variational ansatz?

**Random circuits have concentration of measure, exponential scaling (1803.11173)**

**Unitary coupled cluster (1701.02691)** $U(\vec{\theta}) = e^{T(\vec{\theta}) - T(\vec{\theta})^\dagger}$

$$T(\vec{\theta}) = \mathbb{1}\theta_0 + \underbrace{\sum_{\substack{i \in \text{occ} \\ p \in \text{virt}}} \theta_i^p a_p^\dagger a_i}_{^{(1)}T(\vec{\theta})} + \underbrace{\sum_{\substack{ij \in \text{occ} \\ pq \in \text{virt}}} \theta_{ij}^{pq} a_q^\dagger a_j a_p^\dagger a_i}_{^{(2)}T(\vec{\theta})}$$

Variational / multireference version of classical theory

Extremely powerful approach but also quite costly

**Trotterized adiabatic state preparation (1507.08969)**

Based on real time evolution, similar to QAOA

Works well if adiabatic path is gapped

$$H = \sum_\ell \alpha_\ell H_\ell \qquad U\left(\vec{\theta}\right) = \prod_k \left( \prod_\ell e^{-i\theta_{k,\ell} H_\ell} \right)$$

**Good cost model for variational algs is circuit depth in terms of two-qubit gates**

| Year | arXiv | Basis | Variational Ansatz | Layout | Two-Qubit Depth | Measurements |
|---|---|---|---|---|---|---|
| 2013 | 1304.3061 | Arbitrary | Unitary Coupled Cluster | Arbitrary | $\mathcal{O}(N^5)$ | $\mathcal{O}(\text{poly}(N))$ |
| 2014 | 1403.1539 | Arbitrary | Unitary Coupled Cluster | Arbitrary | $\mathcal{O}(N^4)$ | $\mathcal{O}(\text{poly}(N))$ |
| 2015 | 1507.08969 | Arbitrary | Trottized Adiabatic State Prep | Arbitrary | $\mathcal{O}(N^4)$ | $\mathcal{O}(N^8)$ |
| 2016 | 1509.04279 | Arbitrary | Unitary Coupled Cluster | Arbitrary | $\mathcal{O}(\eta^2 N^2)$ | $\mathcal{O}(N^8)$ |
| 2017 | 1706.00023 | Plane Waves | Trottized Adiabatic State Prep | Planar | $\mathcal{O}(N)$ | $\mathcal{O}(N^4)$ |
| 2017 | 1711.04789 | Plane Waves | Trottized Adiabatic State Prep | Linear | $\mathcal{O}(N)$ | $\mathcal{O}(N^4)$ |

# OpenFermion

The Electronic Structure Package for Quantum Computers

pyscf

www.openfermion.org/
(1710.07629)

Psi4
OPEN-SOURCE QUANTUM CHEMISTRY

**OpenFermion is an Apache 2 open source project for quantum simulation:**

- Generate chem Hamiltonians, Hubbard, jellium, mean-field, etc.
- Compute orbitals, integrals, active spaces, etc. at arbitrary geometries, basis sets
- Map fermions to qubits (e.g. Jordan-Wigner, Bravyi-Kitaev, Verstraete-Cirac)
- Analyze and compile variety of simulation algorithms and more!
- Google software engineering standards enforced; ~50K lines of code at 99.9% test coverage

**OpenFermion is a community! Over two dozen contributors from over fifteen institutions**

Google rigetti quantum computing ZAPATA QUSIM D:wave The Quantum Computing Company™ XANADU QUTech NASA HARVARD BERKELEY LAB

**Framework and platform agnostic**

- Works with Microsoft LIQUID, IBM QISKit, Google Cirq, Xandu Strawberry, Rigetti Forest, etc.

**PULL REQUESTS WELCOME!**