Reducing the computational footprint on quantum hardware by a correlated wavefunction Ansatz

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MAX PLANCK INSTITUTE
FOR SOLID STATE RESEARCH

PostDoc in Prof. Rahm's group at Chalmers University
Development of **quantum algorithms** to enable accurate and efficient **quantum chemistry** calculations on current and near-term quantum computers

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PhD in theoretical chemistry at
MPI Stuttgart and University of Stuttgart
Method Development for Quantum Chemistry
under Supervision of Profs. Alavi and Werner
Development of FCIQMC methods for strongly correlated electron systems
Subsequent 2 years of PostDoc at MPI

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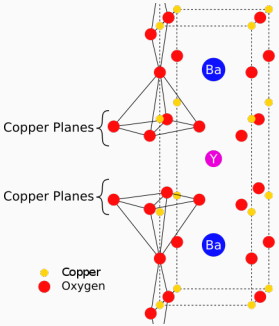
BSc/MSc Studies in Physics at TU Graz
Specialization: **Computational Physics**
Supervision Prof. von der Linden
Application of the Full Configuration Interaction Quantum Monte Carlo method to the Hubbard Model

Outline

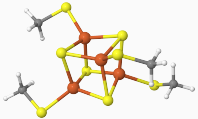
- Motivation
- Correlated Ansatz to reduce the computational footprint on quantum hardware
- Conventional Results
- Outlook

Motivation

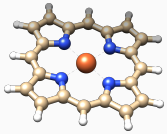
Bottom-up Design of Quantum Materials



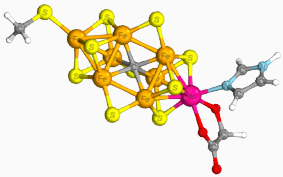
YBCO*: Unconventional high-T_c superconductivity



Iron-Sulfur clusters: electron transfer proteins



(Iron) Porphyrins: oxygen and electron transport



FeMoCo†: primary cofactor of nitrogenase

Interesting systems usually challenging systems for computational approaches.

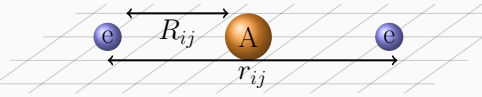
Accurate theoretical understanding at nano-scale for bottom-up materials design!

Images from: *commons.wikimedia.org, †G. Li Manni

Ab Initio Quantum Chemistry – Electronic Structure Theory

All necessary information of a quantum system contained in electronic molecular Hamiltonian (Born-Oppenheimer approx., atomic units and first quantization)

$$\hat{H} = \underbrace{-\sum_i \nabla^2_{\mathbf{r}_i}}_{\text{kinetic energy of e}^-} + \underbrace{\frac{1}{2}\sum_{i\neq j}\frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}}_{\text{e}^- - \text{e}^- \text{ repulsion}} \underbrace{-\sum_{I,j}\frac{Z_I}{|\mathbf{R}_I - \mathbf{r}_j|}}_{\text{Potential}}$$



Electronic properties: Ground- and low-lying excited state properties, energy differences, polarization, response functions, ...

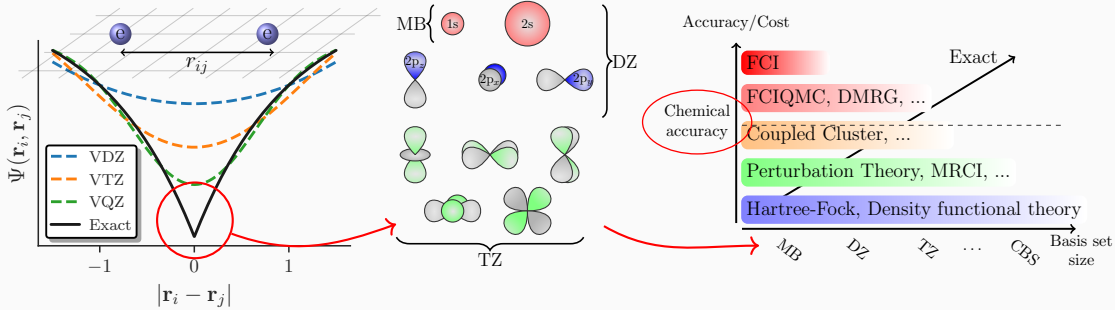
Target: High / chemical accuracy to ensure predictability, interpretability and comparison with experimental results

Task: Solve the Schrödinger equation derived from first principles

$$\hat{H}\,|\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_n)\rangle = E\,|\Psi(\mathbf{r}_1,\ldots,\mathbf{r}_n)\rangle$$

Problems for accurate description: Cusp condition and hierarchy of methods and basis set size

Cusp condition: Singularity of Coulomb potential, $\frac{1}{r_{ij}}$, for $r_{ij} = 0 \rightarrow$ non-differentiable behavior of $\Psi(\{\mathbf{r}\})$ at electron coalescence, so kinetic energy, $-\nabla_{\mathbf{r}}$, cancels

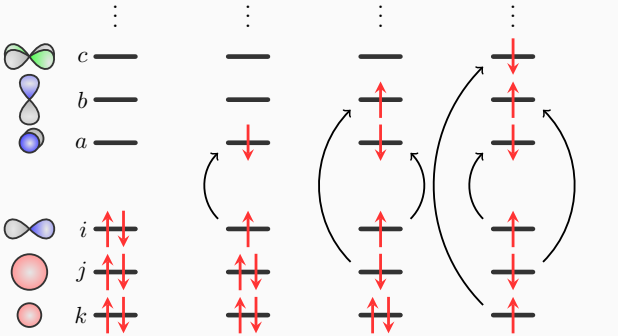


Short range behavior → dynamic correlation. Important for quantitatively accurate quantum chemistry. Necessitates large basis set expansion → costly for highly accurate methods. Adds another axis of complexity!

Kato, Communications on Pure and Applied Mathematics **10** (2), 151 (1957)

Exponential scaling of Full Configuration Interaction

To make the electronic structure problem tractable: map it to atomic/molecular orbitals in quantum chemistry. Post-HF/DFT: Multi-determinant wave function: $|\Psi\rangle = |\Phi_{HF}\rangle + \sum_i c_i |\Phi_i\rangle$. FCI $\Rightarrow$ exact solution in a given basis set



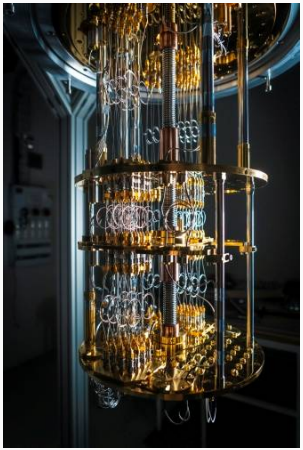
$$|\Phi_{HF}\rangle + |\Phi_i^a\rangle + |\Phi_{ij}^{ab}\rangle + |\Phi_{ijk}^{abc}\rangle + \cdots$$

All possible excitations from HF determinant

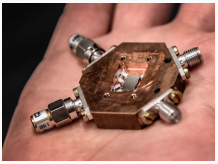
Number of possible states for given number of electrons, N, and orbitals, n, $\sim \binom{N}{n}$

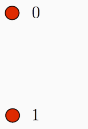
#orbitals	#electrons	#states
2	2	4
4	4	36
8	8	4900
12	12	$\sim 8 \cdot 10^5$
16	16	$\sim 16 \cdot 10^6$
18	18	$\sim 2 \cdot 10^9$

Quantum Computing

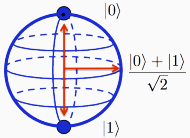


@ Chalmers





Classical Bit



Qubit

0011
0101
0110
1001
1010
1100

Remember: exponentially scaling FCI $|\Psi\rangle \sim \binom{N}{n}$.
$>$ 20 electrons and 20 orbitals can not even store wf.
Due to **entanglement** and **superposition**: n qubits can encode $\sim 2^n$ states:

$$\overbrace{(|0\rangle_1 + |1\rangle_1)}^{\text{qubit1}} \otimes \overbrace{(|0\rangle_2 + |1\rangle_2)}^{\text{qubit2}} = |00\rangle + |01\rangle + |10\rangle + |11\rangle$$

$$|q_1\rangle \otimes |q_2\rangle \otimes |q_3\rangle = \\ = |000\rangle + |001\rangle + |010\rangle + |100\rangle \\ + |011\rangle + |101\rangle + |110\rangle + |111\rangle$$

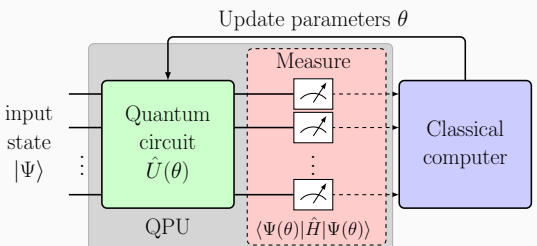
"Nature isn't classical, dammit, and if you want to make a simulation of nature, you'd better make it quantum mechanical" – R. Feynman 1981

Benioff, J. Stat. Phys. **22** (5), 563 (1980); Feynman, Int. J. Theo. Phys. **21** (6/7), 467 (1982); Preskill, Quantum **2**, 79 (2018)

Quantum Chemistry on Quantum Computers

Current quantum hardware has many problems still: noise, decoherence and **limited number of qubits** – noisy intermediate-scale quantum (NISQ) era

Quantum chemistry / electronic structure problem potential use-case / killer-application of NISQ devices and "quantum primacy" for relevant systems



Current hybrid quantum-classical approach:

Use quantum processor (QPU) combined with classical resources. **Basis set scaling** a problem as **every spin-orbital needs a qubit** for representation on quantum hardware.

Minimal basis far from CBS results!

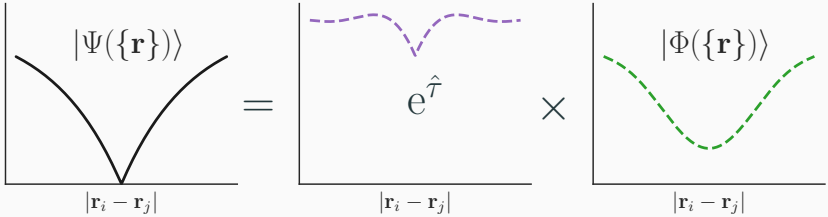
IBM Quantum Roadmap*

Year	2019	2020	**2021**	2022	2023	beyond
# qubits	27	65	**127**	433	1121	...

QPE: Kitaev, arXiv:quant-ph/9511026 (1995), Nielsen and Chuang, *Quantum computation and quantum information* (2001); **VQE:** Peruzzo *et al.*, Nature Comm., **5**, 4213, (2014), McClean, *et al.*, New J. Phys. **18**, 023023 (2016); * https://research.ibm.com/blog/ibm-quantum-roadmap

Correlated Ansatz to reduce the computational footprint on quantum hardware

Cusp Condition – Explicitly Correlated Ansatz



Describe the cusp exactly and capture part of correlation with a **correlated wavefunction Ansatz** (Jastrow Ansatz):

$$|\Psi(\{\mathbf{r}\})\rangle = \mathrm{e}^{\hat{\tau}}\,|\Phi(\{\mathbf{r}\})\rangle\,,$$

with an explicit function of two electron coordinates

$$\hat{\tau}(\{\mathbf{r}\}) = \sum_{i<j} J_{ij} u(\mathbf{r}_i, \mathbf{r}_j)$$

Jastrow, Phys. Rev. **98**, 1479 (1955)

Other Approaches

Transcorrelated approach of Boys and Handy: optimize Slater-Jastrow form, orbitals $\{\phi\}$ of a single Slater determinant $|\Phi_0\rangle$ and Jastrow parameters J_{ij} in $\hat{\tau}$

$$|\Psi_{BH}\rangle = \mathrm{e}^{\hat{\tau}} |\Phi_0(\{\phi\})\rangle$$

Problematic because on non-Hermitian nature of $\bar{H}$

Variational quantum Monte Carlo: minimize variational energy, by optimizing trial-wf. parameters (accuracy limited by trial-wf.) $\rightarrow$ our **starting point**

$$E_{VMC} = \min \frac{\langle \Phi_0 | \mathrm{e}^{\hat{\tau}} \, \hat{H} \, \mathrm{e}^{\hat{\tau}} | \Phi_0 \rangle}{\langle \Phi_0 | \mathrm{e}^{2\hat{\tau}} | \Phi_0 \rangle}, \quad |\Phi_T\rangle = \mathrm{e}^{\tau} |\Phi_0\rangle$$

Explicitly correlated methods (R12/F12): use correlating functions of the interelectronic distance to describe electronic cusp

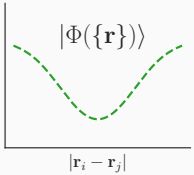
$$|\Psi_{F12}\rangle = (1 + \lambda \hat{Q}_{12} f(r_{12})) |\Phi_{HF}\rangle + \sum_{ijab} c_{ij}^{ab} |\Phi_{ij}^{ab}\rangle, \quad \text{with } f(r_{12}) \sim \mathrm{e}^{-\gamma r_{12}}$$

Boys, Handy 1969; Tsuneyuki, Prog. Theor. Phys. Supp., **176**, 134 (2008); Scuseria et al., PRB, **91**, 041114 (2015) **R12:** Kutzelnigg, Theor. Chem. Acc. **68**, 335 (1985); Klopper, Kutzelnigg, Chem. Phys. Lett. **134**, 17 (1987); **F12:** S. Ten-no, Chem. Phys. Lett. **398**, 56 (2004)

Similarity Transformation – Transcorrelated Method

Instead of $\hat{H}\,|\Psi\rangle = E\,|\Psi\rangle$ solve the similarity transformed (ST) problem

$$\begin{aligned} \hat{H}\,|\Psi\rangle &= E\,|\Psi\rangle \\ \mathrm{e}^{-\hat{\tau}} \to |\quad \hat{H}\,\mathrm{e}^{\hat{\tau}}\,|\Phi\rangle &= E\,\mathrm{e}^{\hat{\tau}}\,|\Phi\rangle \\ \left(\mathrm{e}^{-\hat{\tau}}\,\hat{H}\,\mathrm{e}^{\hat{\tau}}\right)|\Phi\rangle &= \bar{H}\,|\Phi\rangle = E\,|\Phi\rangle \end{aligned}$$



Baker-Campbell-Hausdorff (BCH) exp. to obtain s.t. Hamiltonian:

$$\bar{H} = \mathrm{e}^{-\hat{\tau}}\,\hat{H}\,\mathrm{e}^{\hat{\tau}} = \hat{H} + [\,\hat{H},\hat{\tau}\,] + \frac{1}{2}\,[\,[\,\hat{H},\hat{\tau}\,],\hat{\tau}\,] + \ldots$$

terminates at 2nd order. Only kinetic energy operators in $\hat{H}$ do not commute with $\hat{\tau}$

$$\begin{aligned} \bar{H} &= \hat{H} - \sum_i \left(\frac{1}{2}\nabla_i^2\hat{\tau} + (\nabla_i\hat{\tau})\nabla_i + \frac{1}{2}(\nabla_i\hat{\tau})^2\right) \\ &= \hat{H} - \sum_{i<j}\hat{K}(\mathbf{r}_i,\mathbf{r}_j) - \sum_{i<j<k}\hat{L}(\mathbf{r}_i,\mathbf{r}_j,\mathbf{r}_k) \end{aligned}$$

- Exact transformation
- Rapid basis set convergence
- 3-body terms and non-Hermitian!

Hirschfelder, JCP, **39**, 3145 (1963), Boys and Handy, Proc. R. Soc. A (**1969**)

Consequences: 3-body terms

Transcorrelated Hamiltonian in 2nd quantised form

$$\bar{H} = \sum_{pq,\sigma} h_q^p a_{p,\sigma}^\dagger a_{q,\sigma} + \frac{1}{2} \sum_{pqrs} (V_{rs}^{pq} - K_{rs}^{pq}) \sum_{\sigma,\tau} a_{p,\sigma}^\dagger a_{q,\tau}^\dagger a_{s,\tau} a_{r,\sigma}$$
$$- \frac{1}{6} \sum_{pqrstu} L_{stu}^{pqr} \sum_{\sigma\tau\lambda} a_{p,\sigma}^\dagger a_{q,\tau}^\dagger a_{r,\lambda}^\dagger a_{u,\lambda} a_{t,\tau} a_{s,\sigma}$$

with $K_{rs}^{pq} = \langle \phi_p \phi_q | \hat{K} | \phi_r \phi_s \rangle$

$L_{stu}^{pqr} = \langle \phi_p \phi_q \phi_r | \hat{L} | \phi_s \phi_t \phi_u \rangle$ (48-fold symmetry in L for real orbitals)

Both integrals K and L are computed numerically using standard DFT grids over gaussian orbitals. The main problem is the storage of L. Current limit $\approx$ 100 orbitals

$\Rightarrow$ 3-body terms need more measurements / deeper circuits on quantum hardware

Consequences: Non-Hermitian → loss of variational principle

Variational methods like VQE not applicable. Solve for the **right** eigenvector of non-Hermitian $\bar{H}$ by projection / imaginary-time evolution

$$i\frac{\partial\,|\Psi\rangle}{\partial t} = \hat{H}\,|\Psi\rangle \quad \overset{\tau=it}{\rightarrow} \quad \frac{\partial\,|\Psi\rangle}{\partial \tau} = -\hat{H}\,|\Psi\rangle \quad \rightarrow \quad |\Psi(\tau)\rangle = N(\tau)\,\mathrm{e}^{-\hat{H}\tau}\,|\Psi(0)\rangle$$

Conventional: FCIQMC, Quantum: **Ansatz-based QITE*** allows to formulate non-unitary time evolution as a minimization

$$\frac{\partial\,|\Psi(\tau)\rangle}{\partial \tau} = -(\hat{H} - E(\tau))\,|\Psi(\tau)\rangle\,, \quad \text{with} \quad E(\tau) = \langle\Psi(\tau)|\hat{H}|\Psi(\tau)\rangle,$$

Use McLachlan's variational principle

$$\delta||\frac{\partial}{\partial \tau} + \hat{H} - E(\tau)\,|\Psi(\tau)\rangle\,|| \overset{!}{=} 0 \quad \text{and an Ansatz:} \quad |\Psi(\tau)\rangle \approx |\Phi(\vec{\theta}(\tau))\rangle$$

Booth, Thom, and Alavi, JCP, **131**, 054106 (2009); Guther, ..., **WD**, ..., Alavi, JCP, **153**, 034107 (**2020**) *McArdle, *et al.*, npj Quantum Information **5**, 75, 2019; Motta *et al.*, Nature Physics **16**, 205, 2020; Current work with **Igor Sokolov**

Workflow

1. Starting guess $|\Phi_0\rangle$: HF in a basis set (e.g. VDZ)

pyscf

2. VMC optimization of $\hat{\tau}(\{\mathbf{r}\}) = \sum_{i<j} J_{ij} u(\mathbf{r}_i, \mathbf{r}_j)$
$E_{VMC} = \min_{J_{ij}} \langle \Phi_0 | e^{\hat{\tau}} \hat{H} e^{\hat{\tau}} | \Phi_0 \rangle$

CASINO*

3. Compute 2- and 3-body integrals in real-space
$\sum_i \left(\frac{1}{2} \nabla_i^2 \hat{\tau} + (\nabla_i \hat{\tau}) \nabla_i + \frac{1}{2} (\nabla_i \hat{\tau})^2 \right)$
and project onto chosen basis (VDZ)

TCHINT†

4. Solve for right eigenvector and energy of $\bar{H}$
by projection / imaginary-time evolution
$|\Psi(\tau)\rangle = N(\tau)\, e^{-\hat{H}\tau} |\Phi(0)\rangle$

NECI‡

*https://vallico.net/casinoqmc/ †https://gitlab.com/kguther/tchint ‡https://github.com/ghb24/NECI_STABLE

Results: Total energies of first-row atoms

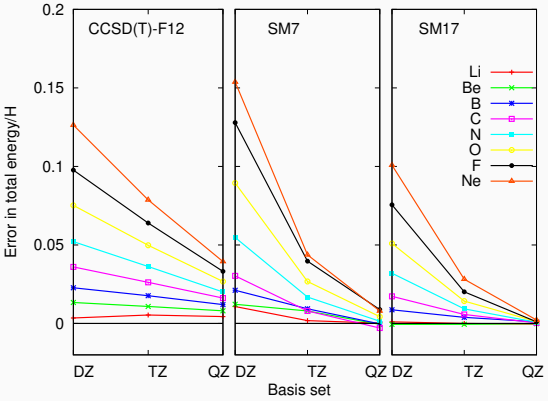
Boys-Handy form of $u(\mathbf{r}_i, \mathbf{r}_j)$ (1969)[†]:

$$u(\mathbf{r}_i, \mathbf{r}_j) = \sum_{\substack{mno \\ m+n+o \leq 6}} c_{mno}(\bar{r}_i^m \bar{r}_j^n + \bar{r}_j^m \bar{r}_i^n)\bar{r}_{ij}^o,$$

- $\bar{r}_i^m$: **distance of electrons from nuclei**
- $\bar{r}_{ij}^o$: **relative distance between electrons**
- Includes $e-e$, $e-n$ and $e-e-n$ terms
- Parameters obtained by VMC*

$\Rightarrow$ CBS limit results at QZ level

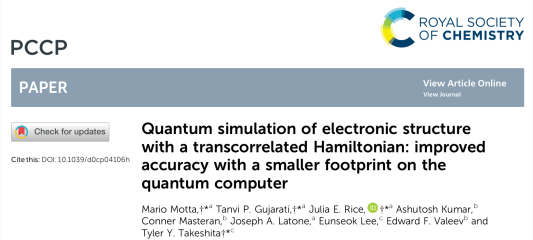
$\Rightarrow$ No need for core functions



SM7: u without $e-e-n$ terms; SM17: full parametrization in cc-pVnZ basis set **without core functions**!

Cohen, Luo, Guther, **WD**, Tew and Alavi, JCP, **151**, 061101 (2019); [†]Boys and Handy, Proc. R. Soc. A (**1969**); *Schmidt and Moskowitz, JCP, **93**, 4172 (**1990**)

Ab initio systems – other work



PCCP

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Cite this: DOI: 10.1039/d0cp04106h

Quantum simulation of electronic structure with a transcorrelated Hamiltonian: improved accuracy with a smaller footprint on the quantum computer

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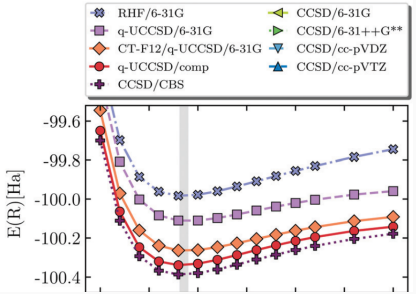
2 Methods

2.1 Canonical transcorrelated F12 Hamiltonian

In the CT-F12 method, two main approximations are employed in addition to the approximate BCH expansion of eqn (2): (a) the expansion is truncated to only include up to double commutators and (b) in the double commutator term, the full Hamiltonian $\hat{H}$ is replaced by its effective 1-body constituent, the Fock operator $\hat{F}$,

$$\hat{H}' \approx \hat{H} + [\hat{H}, \hat{A}]_{1,2} + \frac{1}{2}\Big[[\hat{F}, \hat{A}]_{1,2}, \hat{A}\Big]_{1,2}. \qquad (3)$$



Hydrogen Fluoride

Motta *et al.*, Phys. Chem. Chem. Phys. **22**, 24270, 2020

Ab initio systems – other work

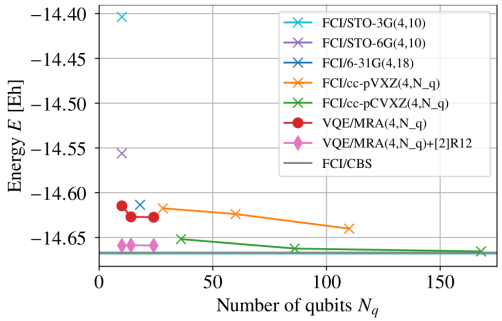
Improving the Accuracy of the Variational Quantum Eigensolver for Molecular Systems by the Explicitly-Correlated Perturbative $[2]_{\mathbf{R12}}$ -Correction

Philipp Schleich,[1,2,3,*] Jakob S. Kottmann,[1,4,†] and Alán Aspuru-Guzik[1,4,3,5,‡]

[1]*Department of Computer Science, University of Toronto, Canada.*
[2]*Applied and Computational Mathematics, Department of Mathematics, RWTH Aachen University, Aachen, Germany*
[3]*Vector Institute for Artificial Intelligence, Toronto, Canada.*
[4]*Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Canada.*
[5]*Canadian Institute for Advanced Research (CIFAR) Lebovic Fellow, Toronto, Canada*

(Dated: October 14, 2021)

On the contrary, we opt to use an *a posteriori* correction based on a perturbative *explicitly correlated* method, namely the spin-free variant of $[2]_{\mathrm{R12}}$ [18–20]. This approach is to be contrasted with other post-corrections in form of the family of quantum subspace expansion techniques [8, 9, 21]; and an application of VQE together with the transcorrelated approach, $[2]_{\mathrm{R12}}$ or a so called "CABS singles" correction [22] has yet been suggested in [23]. Brief comments on the distinction of our approach with this one will follow.

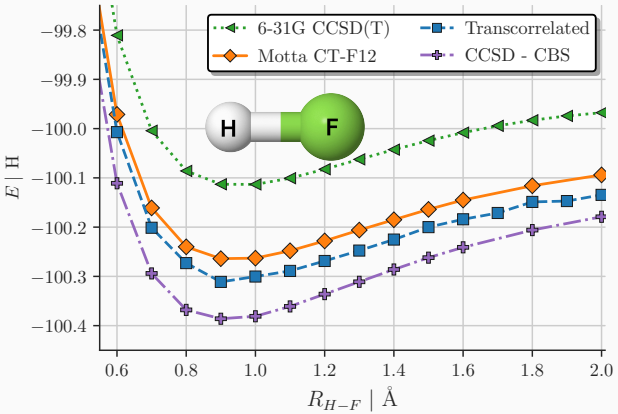


(b) Be, $E_{\mathrm{CBS}} = -14.667$ [96]

Conventional Results

Preliminary Conventional Results – HF

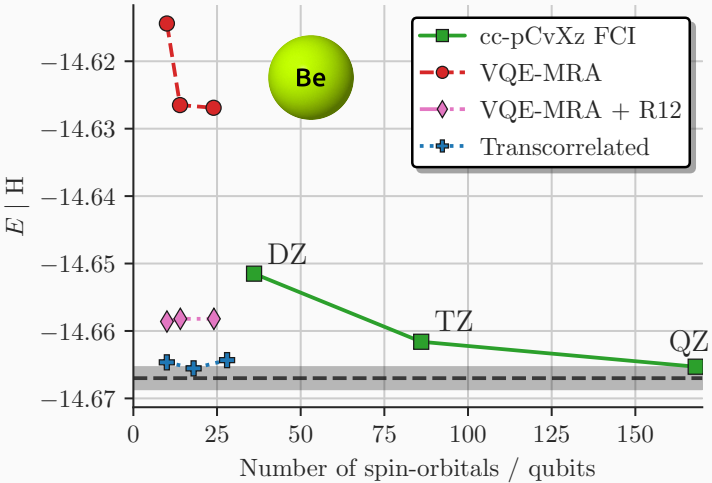
Hydrogen Fluoride in a 6-31G basis set – **22 spin-orbitals → 22 qubits**



Motta *et al.*, Phys. Chem. Chem. Phys. **22**, 24270, 2020

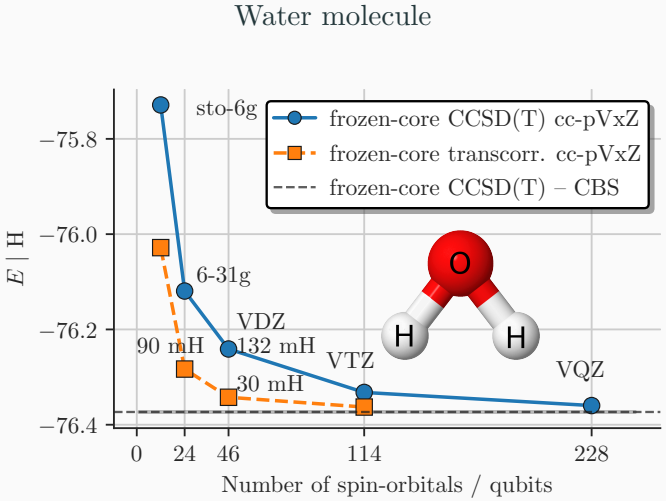
Preliminary Conventional Results – Be

Beryllium atom study. VQE – basis set free, TC: sto-6g, 6-31g and cc-pvdz basis



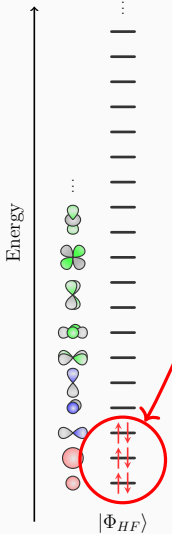
Schleich *et al.*, arXiv:2110.06812, 2021

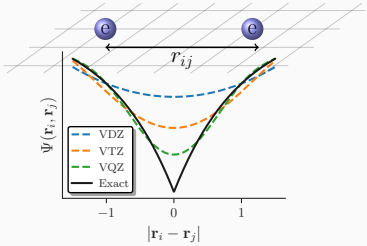
Preliminary Conventional Results – H_2O



Outlook

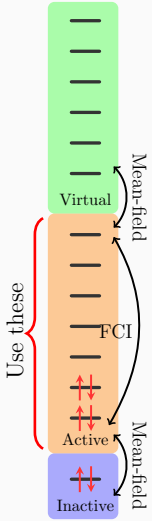
Outlook: (Virtual) orbital optimization



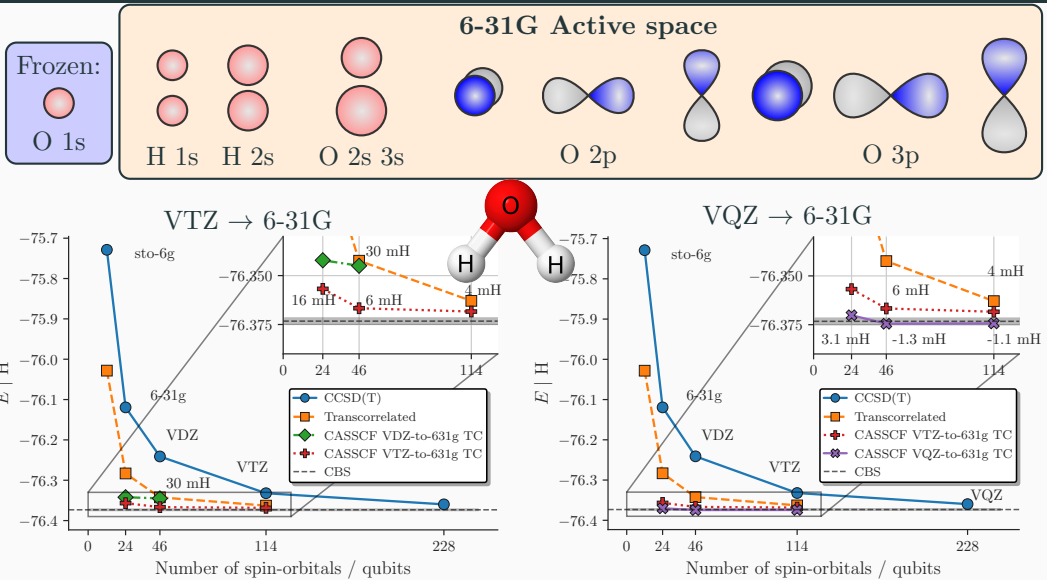


$$E_{VMC} = \min \frac{\langle \Phi_{HF} | \mathrm{e}^{\hat{\tau}} \, \hat{H} \, \mathrm{e}^{\hat{\tau}} | \Phi_{HF} \rangle}{\langle \Phi_{HF} | \mathrm{e}^{2\hat{\tau}} | \Phi_{HF} \rangle}$$

Standard orbitals not optimal for the TC method → include effect of virtuals (e.g. polarization) through orbital optimization / downfolding → e.g. complete active space self-consistent field (CASSCF)



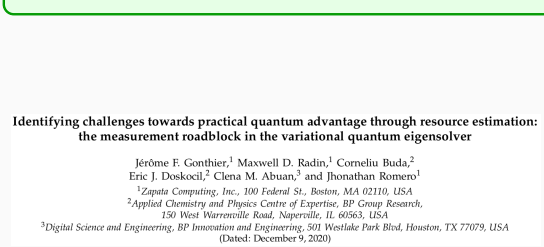
Water revisited – CASSCF orbitals



Outlook: Virtual orbital optimization

CASSCF is scaling exponentially with the active space size!

Cheaper methods, based on 2nd order perturbation theory (MP2):
Optimized virtual orbital space (OVOS) and **frozen natural orbitals** (FNO)
Correlation energy in the TC method usually smaller → more effective



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

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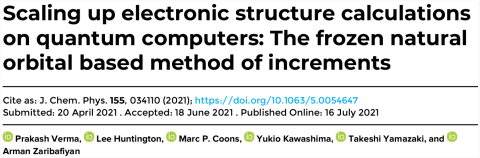
(Dated: December 9, 2020)



Scaling up electronic structure calculations on quantum computers: The frozen natural orbital based method of increments

Cite as: J. Chem. Phys. **155**, 034110 (2021); https://doi.org/10.1063/5.0054647
Submitted: 20 April 2021 . Accepted: 18 June 2021 . Published Online: 16 July 2021

Prakash Verma, Lee Huntington, Marc P. Coons, Yukio Kawashima, Takeshi Yamazaki, and Arman Zaribafiyan

OVOS: Adamowicz and Bartlett, JCP **86**, 6314 (1987); **FNO:** Sosa *et al.*, Chem. Phys. Lett. **159** (2-3) 148 (1989); Taube and Bartlett Collect. Czech. Chem. Commun. **70**, 837 (2005); **NISQ:** Gonthier *et al.* arXiv:2012.04001; (2020); Verma *et al.* JCP **155**, 034110 (2021)

Thank you for your attention!

Full Configuration Interaction Quantum Monte Carlo

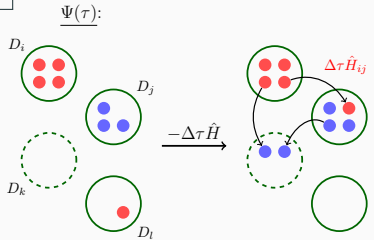
- *Projector MC method* based on the **imaginary-time Schrödinger equation**, stochastically sampling FCI wavefunction. Integration leads to an iterable equation:

$$i\frac{\partial\,|\Psi(t)\rangle}{\partial t} = \hat{H}\,|\Psi(t)\rangle \overset{\tau=it}{\rightarrow} \frac{\partial\,|\Psi(\tau)\rangle}{\partial\tau} = -\hat{H}\,|\Psi(\tau)\rangle \rightarrow |\Psi_{GS}\rangle \propto \lim_{\tau\rightarrow\infty} \mathrm{e}^{-\tau\hat{H}}\,|\Phi(\tau=0)\rangle$$

- *First order* Taylor expansion $\mathrm{e}^{-\Delta\tau\hat{H}} \approx 1 - \Delta\tau\hat{H}$ leads to the *working equation:*

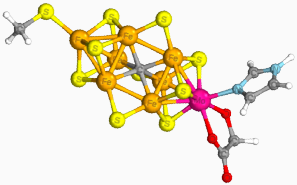
$$\boxed{c_i(\tau+\Delta\tau) = [1-\Delta\tau H_{ii}]\,c_i(\tau) - \Delta\tau\sum_{j\neq i} H_{ij}c_j(\tau)} \quad \text{for } c_i \text{ in } |\Psi(\tau)\rangle = \sum_i c_i\,|D_i\rangle$$

- Solved stochastically by the *population dynamics* of **"walkers"** in the discrete Slater determinant (SD) Hilbert space.
- **Multireference method** and highly accurate solutions for system sizes $> (50e, 50o)$ possible.



Booth, Thom, and Alavi, JCP, **131**, 054106 (2009); Guther, ..., **WD**, ..., Alavi, JCP, **153**, 034107 (**2020**)

Massively Parallel – Towards the Exascale



Markus Rampp @

MPCDF

MAX PLANCK
COMPUTING & DATA FACILITY

CAS(54e,54o) FeMoco molecule on 512 and 620 nodes @ Max-Planck Cobra HPC cluster

Walkers	Cores	Time/iteration	Ratio cores	Ratio time/iteration	Parallel efficiency (%)
32×10^9	19960	23.5	1.242	1.246	99.68
32×10^9	24800	18.8	–	–	–

Ongoing work in the European Center of Excellence
Targeting Real Chemical Accuracy at the EXascale (TREX)
Interfaced with `Molpro`, `OpenMolcas`, `PySCF` and `VASP`

William Jalby @

TREX
Targeting Real chemical accuracy at the EXascale

Guther, ..., **WD**, ..., Alavi, JCP, **153**, 034107 (**2020**)

Possible future project on Quantum Computing: Use spin-symmetry to reduce number of qubits/parameters/circuit depth

Based on the **spin-free** formulation of the non-relativistic Hamiltonian:

$$\hat{H} = \sum_{ij}^{n} t_{ij}\, \hat{E}_{ij} + \frac{1}{2} \sum_{ijkl}^{n} V_{ijkl} \left(\hat{E}_{ij}\hat{E}_{kl} - \delta_{jk}\hat{E}_{il} \right)$$

Use the (G)UGA Ansatz:

- Possible qubit reduction, due to n *spatial* orbitals instead of $2n$ spin-orbitals![*]
- Better overlap of Ansatz wavefunction with true groundstate beneficial for quantum phase estimation algorithm
- Reduce necessary Ansatz qubit number, due to conserved total spin (Serber-type Ansatz by Sugisaki *et al.*[‡])
- Reduce circuit depth, since no spin-contamination[†]

Possible collabs: Anyone interested in highly accurate results for spin systems?

[*]Whitfield, JCP., **139**, 021105 (**2013**), [‡]Sugisaki *et al.*, CPL X, **1**, 100002 (**2019**), [†]Tsuchimochi, Mori, and Ten-no, PRR **2**, 043142 (**2020**), Grimsley, Economou, Barnes and Mayhall, Nature Comm. **10**, 3007 (**2019**)

Imaginary-time propagation with s.t. Hamiltonians

Why is the FCIQMC method applicable?

$$|\Psi(\beta)\rangle = \mathrm{e}^{-\beta(\hat{H}-E_0)}\,|\Psi(0)\rangle \quad \rightarrow \quad |\Psi_0\rangle = \lim_{\beta\to\infty} \mathrm{e}^{-\beta(\hat{H}-E_0)}\,|\Psi(0)\rangle$$

$$\text{with} \quad |\Psi(\beta)\rangle = \mathrm{e}^{\hat{\tau}}\,|\Phi(\beta)\rangle$$

$$|\Phi(\beta)\rangle = \mathrm{e}^{-\beta(\bar{H}-E_0)}\,|\Phi(0)\rangle \quad \rightarrow \quad |\Phi_0\rangle = \lim_{\beta\to\infty} \mathrm{e}^{-\beta(\bar{H}-E_0)}\,|\Phi(0)\rangle$$

Proof:

$$\mathrm{e}^{\hat{\tau}}\,|\Phi(\beta)\rangle = |\Psi(\beta)\rangle = \mathrm{e}^{-\beta(\hat{H}-E_0)}\,|\Psi(0)\rangle = \mathrm{e}^{-\beta(\hat{H}-E_0)}\,\mathrm{e}^{\hat{\tau}}\,|\Phi(0)\rangle$$

$$\Rightarrow |\Phi(\beta)\rangle = \mathrm{e}^{-\hat{\tau}}\,\mathrm{e}^{-\beta(\hat{H}-E_0)}\,\mathrm{e}^{\hat{\tau}}\,|\Phi(0)\rangle$$

$$= \lim_{m\to\infty} \mathrm{e}^{-\hat{\tau}} \left(1 - \frac{\beta}{m}(\hat{H}-E_0)\right)^m \mathrm{e}^{\hat{\tau}}\,|\Phi(0)\rangle$$

$$= \lim_{m\to\infty} \underbrace{\mathrm{e}^{-\hat{\tau}}}_{\rightarrow} \left(1 - \frac{\beta}{m}(\hat{H}-E_0)\right) \underbrace{\mathrm{e}^{\hat{\tau}}}_{\leftarrow} \mathrm{e}^{-\hat{\tau}} \left(1 - \frac{\beta}{m}(\hat{H}-E_0)\right) \ldots \mathrm{e}^{\hat{\tau}}\,|\Phi(0)\rangle$$

Other Work

Compact numerical solutions to the two-dimensional repulsive Hubbard model obtained via nonunitary similarity transformation

Werner Dobrautz, Hongjun Luo, and Ali Alavi
Phys. Rev. B **99**, 075119 – Published 8 February 2019

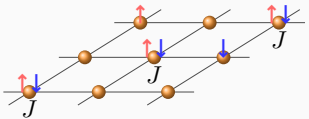
Improving the accuracy of quantum computational chemistry using the transcorrelated method

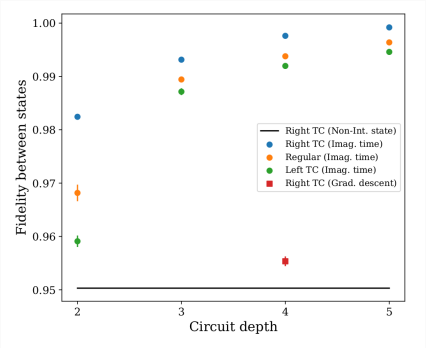
Sam McArdle[1,*] and David P. Tew[2]
[1]*Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, United Kingdom*
[2]*Physical and Theoretical Chemical Laboratory, University of Oxford, South Parks Road, Oxford OX1 3QZ, United Kingdom*
(Dated: June 22, 2020)

Suppress energetically unfavourable double occupancies via the *Gutzwiller* Ansatz:

$$|\Psi\rangle = e^{\hat{\tau}} |\Phi\rangle, \quad \hat{\tau} = J \sum_i n_{i\uparrow} n_{i\downarrow}$$

$$\bar{H} |\Phi\rangle = \left(-t \sum_{\langle i,j \rangle, \sigma} e^{-\hat{\tau}} a^\dagger_{i\sigma} a_{j\sigma} e^{\hat{\tau}} + U \sum_i n_{i\uparrow} n_{i\downarrow} \right) |\Phi\rangle = E |\Phi\rangle$$





McArdle and Tew, arXiv:2006.11181, 2020 Gutzwiller, PRL 10, **159** (1963); Tsuneyuki, Prog. Theor. Phys. Supp., **176**, 134 (2008); Scuseria et al., PRB, **91**, 041114 (2015); **WD**, Luo, and Alavi PRB **99**, 075119 (2019)

The Hubbard model