# Resource efficient method for representation and measurement of constrained electronic structure states with a quantum computer

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**Abstract**

We present a novel method for improving the quantum simulation of the ground state energy of molecules. We perform a pre-processing step classically, which reduces the dimensionality of the problem by generating a custom mapping which excludes states which violate problem constraints. Subsequently, a specialized measurement scheme is used to extract the expectation value of the problem Hamiltonian through this mapping. We demonstrate that this method reduces the amount of quantum resources needed to run a Variational Quantum Eigensolver (VQE) algorithm without making any approximations to the physics of the quantum chemistry problem.

## Introduction

$$E_g = \min_\theta \{\langle\psi|U^\dagger(\theta)\hat{H}U(\theta)|\psi\rangle\}$$

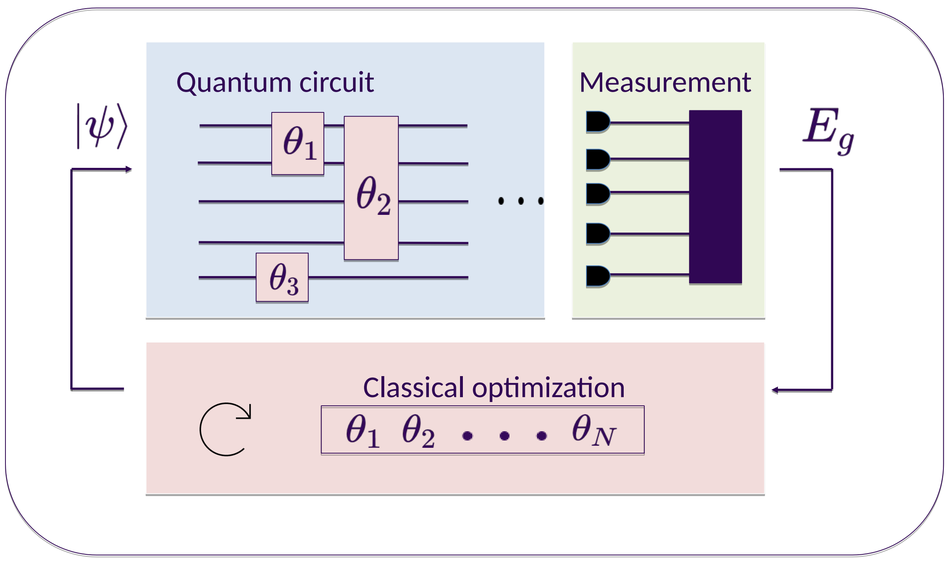



**Figure 1:** Schematic of the VQE algorithm, used to determine the ground state of a molecule.

On Noisy Intermediate Scale Quantum (NISQ) hardware, the performance of VQE algorithms is limited by three main factors:

**1) Number of available qubits**. For every molecular orbital included in the calculation, two qubits are required for the quantum computer to be able to represent all possible electronic states.

**2) Size and number of executable circuits**. Each gate in the circuit accumulates errors and decoherence limits the possible depth for quantum circuits. Executing more circuits takes more time (and therefore cost for the user).

**3) Ability to enforce constraints.** The design of the quantum circuit or the nature of the optimization algorithm may cause VQE to waste time and efficiency searching states with wrong spin, electron number or spatial symmetry.

## Method

We have developed and filed a patent [1] for a **method for mapping** a constrained electronic structure problem to a quantum computer.

- Pre-processing step **performed on a classical computer**, before any quantum computations are made, visualized in Figure 2.
- **One time calculation**, regardless of the details of the VQE algorithm
- **Makes no approximations.** The mapping takes advantage of mathematical redundancies.
- **Compatible with state-of-the-art VQE methods**. Puts no limitation on which circuits can be used to prepare the state. Also retains the possibility for further resource requirement reduction through approximation methods.
- Employing this method has **multiple desirable consequences**, directly addressing the problems outlined in the introduction:

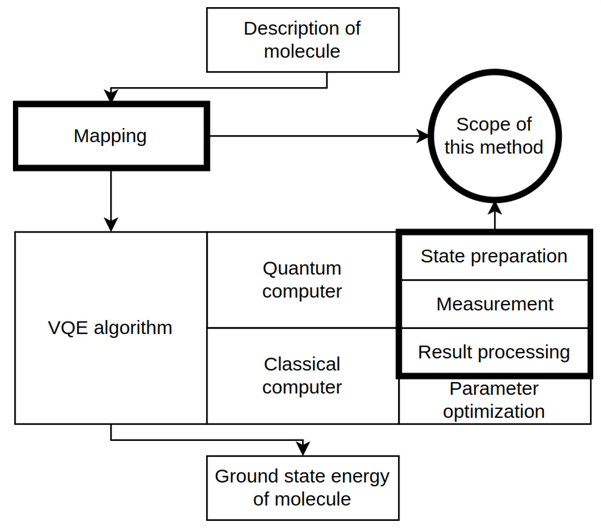



**Figure 2:** Scope of our method within a calculation

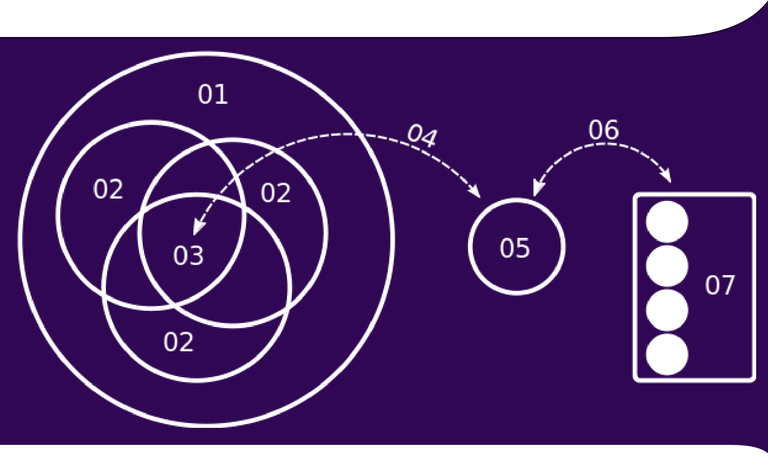
**1) Fewer qubits needed for simulation**. The number of qubits required by the quantum computer is reduced, because the representable space (05) is smaller than the original (01), as shown on Figure 3. This enables performing calculations that would have otherwise been too big for available hardware. Additionally, the width of the circuits needed to parameterize this space is also reduced, thus incurring fewer errors when executed during VQE.

**2) Reduced sources of errors**. States which violate constraints are entirely excluded from the calculation, making it impossible for the VQE to attempt to evaluate those states. This makes the optimization procedure more efficient and the final answer more reliable. Regardless of errors incurred on physical hardware, the answer is guaranteed to satisfy problem constraints.

**3) Access to more calculations with VQE.** We obtain the ability to choose which constraints to enforce. This enables us to search for other states besides the ground state, such as excited states with particular characteristics. No additional post-processing or need to rely on an accurate ground state estimation, as is required by existing methods such as QSE [2] and VQD [3].

After the mapping, we use a **custom measurement scheme**, filed in a separate patent [4], which prescribes a recipe for the quantum computer to extract the expectation value of the original problem Hamiltonian. This scheme can also be used independently of the mapping and in some cases, reduces the number of circuits required for the measurement.

**Figure 3:** Reduction of the original space size of the problem (01) by identifying the subspaces of valid states defined by the problem constraints (02). The intersection of these subsapces (03) is mapped (04) to a newly defined Hilbert space (05), which can then be represented (06) on a quantum comptuer (07).




## Performance and analysis

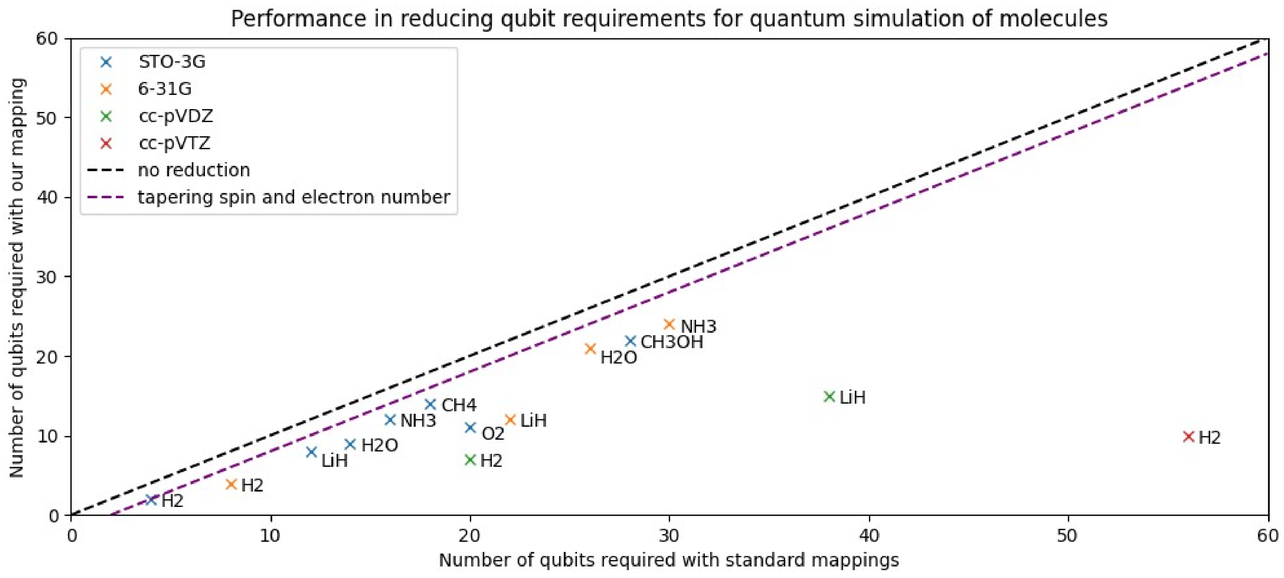



**Figure 4:** Performance of method in terms of qubit requirement reduction. Different colors of crosses correspond to different basis sets used for the quantum chemistry calculation.

The constraints used in Figure 4 are electron number and total spin. Results which also incorporate molecular point group symmetries are not shown here, but we have confirmed that it is possible, and automation and benchmarking of this feature is ongoing work.

The computational capabilities are limited by the size of the output, rather than the size of the input, which enables some particularly drastic reductions in qubit requirement to be made. For example, for the hydrogen molecule (H2) in the cc-pvtz basis set, we are able to reduce the problem from 56 qubits to 10 qubits, bringing an otherwise inaccessible system within the range of NISQ devices.

To verify that the mapping is successful and makes no approximations:
**1) FCI calculation**. If both input and output are small enough for exact diagonalization, we can calculate the ground state for both cases and make a direct comparison.
**2) CIS and CISD calculations**. If the input too large for FCI calculation, but output sufficiently small to diagonalize, then we can calculate the ground state for the output and compare it to CIS and CISD values provided in public quantum chemistry databases.
**3) VQE**. Regardless of size, we may attempt to use VQE on both cases and observe whether they approach the same value or a value similar to those expected from other quantum chemistry calculations.

- We report an exact match for all molecules which fall under the first case of verification
- Example for second case: Ground state energy of H2 in the cc-pvtz basis set after exact diagonalization of reduced problem: -1.1723366673753 Hartree. This is equal to the CISD value up to the amount of digits provided by NIST [5].
- Ongoing work to establish a rigorous basis for verifying the results belonging to the third case.

## Benchmarking

Below we present tables with comparisons to other existing methods, which perform a similar task of reducing the amount of quantum resources without making approximations.

| LiH (STO-3G) | $N_Q$ | $N_C$ | HF | VQE | FCI |
|---|---|---|---|---|---|
| Our method (mapping and measurement) | 7 | 123 | -7.861 | -7.880 | -7.882 |
| Our method (measurement only) | 12 | 84 | -7.861 | N/A | -7.882 |
| No preprocessing | 12 | 630 | -7.861 | N/A | -7.882 |
| Qubit tapering | 8 | 557 | -7.861 | -7.880 | -7.882 |
| Pauli grouping | 12 | 154 | -7.861 | N/A | -7.882 |
| Tapering + Grouping | 8 | 104 | -7.861 | -7.880 | -7.882 |

| BeH2 (STO-3G) | $N_Q$ | $N_C$ | HF | VQE | FCI |
|---|---|---|---|---|---|
| Our method (mapping and measurement) | 8 | 256 | -15.560 | -15.569 | -15.595 |
| Our method (measurement only) | 14 | 94 | -15.560 | N/A | -15.595 |
| No preprocessing | 14 | 665 | -15.560 | N/A | -15.595 |
| Qubit tapering | 9 | 595 | -15.560 | -15.576 | -15.595 |
| Pauli grouping | 14 | 208 | -15.560 | N/A | -15.595 |
| Tapering + Grouping | 9 | 107 | -15.560 | -15.576 | -15.595 |

- $N_Q$ – Number of qubits required for simulation. We use spin, electron number and spatial symmetries for both our method and tapering. We use the OpenFermion [6] implementation for tapering [7].
- $N_C$ – Number of unique circuits required for measurement. We use the Pennylane [8] implementation for Pauli grouping [9].
- HF is the energy of the Hartree-Fock state from which the VQE is initiated for all cases.
- VQE is obtained with 5 iterations of Qubit-ADAPT-VQE [10], run on a state vector simulator. Some results are not available due to issues explained in ref [11].
- FCI is the exact energy to which the VQE should eventually converge.

[1] EP Patent Application No. 22177733.1
[2] Nakanishi, Ken M., Kosuke Mitarai, and Keisuke Fujii. "Subspace-search variational quantum eigensolver for excited states." Physical Review Research 1.3 (2019): 033062.
[3] Higgott, Oscar, Daochen Wang, and Stephen Brierley. "Variational quantum computation of excited states." Quantum 3 (2019): 156.
[4] EP Patent Application No. 22177733.7
[5] http://cccbdb.nist.gov/
[6] McClean, Jarrod R., et al. "OpenFermion: the electronic structure package for quantum computers." Quantum Science and Technology 5.3 (2020): 034014.
[7] Bravyi, Sergey, et al. "Tapering off qubits to simulate fermionic Hamiltonians." arXiv preprint arXiv:1701.08213 (2017).
[8] Ville Bergholm et al. PennyLane: Automatic differentiation of hybrid quantum-classical computations. 2018. arXiv:1811.04968
[9] Gokhale, Pranav, et al. "Minimizing state preparations in variational quantum eigensolver by partitioning into commuting families." arXiv preprint arXiv:1907.13623 (2019).
[10] Tang, Ho Lun, et al. "qubit-adapt-vqe: An adaptive algorithm for constructing hardware-efficient ansätze on a quantum processor." PRX Quantum 2.2 (2021): 020310.
[11] Shkolnikov, V. O., et al. "Avoiding symmetry roadblocks and minimizing the measurement overhead of adaptive variational quantum eigensolvers." arXiv preprint arXiv:2109.05340 (2021).