Unitary partitioning approach to the measurement problem in the Variational Quantum Eigensolver method

Artur F. Izmaylov\textsuperscript{a,b}, Tzu-Ching Yen\textsuperscript{a}, Robert A. Lang\textsuperscript{a,b}, and Vladyslav Verteletskyi\textsuperscript{a,b,c}

\textsuperscript{a}Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Toronto, Ontario, M5S 3H6, Canada; \textsuperscript{b} Department of Physical and Environmental Sciences, University of Toronto Scarborough, Toronto, Ontario, M1C 1A4, Canada; \textsuperscript{c}Department of Quantum Field Theory, Taras Shevchenko National University of Kyiv, Kyiv, 03022, Ukraine

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To obtain estimates of electronic energies, the Variational Quantum Eigensolver (VQE) technique performs separate measurements for multiple parts of the system Hamiltonian. Current quantum hardware is restricted to projective single-qubit measurements, and thus, only parts of the Hamiltonian which form mutually qubit-wise commuting groups can be measured simultaneously. The number of such groups in the electronic structure Hamiltonians grows as $N^4$, where $N$ is the number of qubits, and thus puts serious restrictions on the size of the systems that can be studied. Using a partitioning of the system Hamiltonian as a linear combination of unitary operators we found a circuit formulation of the VQE algorithm that allows one to measure a group of fully anti-commuting terms of the Hamiltonian in a single series of single-qubit measurements. Compared to previously used grouping of Hamiltonian terms based on their qubit-wise commutativity, the unitary partitioning provides at least an $N$-fold reduction in the number of measurable groups.

I. INTRODUCTION

The variational quantum eigensolver (VQE) method\textsuperscript{1–5} provides a practical approach to solving the eigen-value problem for many-body interacting Hamiltonians on current and near-future universal quantum computers. Solving this problem for fermionic systems such as molecules and solids opens numerous possibilities for developing new materials and pharmaceutical compounds.

VQE is a hybrid quantum-classical approach based on the variational theorem and a mapping of the electronic structure problem

$$\hat{H}_e(\mathbf{R}) |\Psi(\mathbf{R})\rangle = E_e(\mathbf{R}) |\Psi(\mathbf{R})\rangle$$

(1)

to its qubit counterpart

$$\hat{H}_q(\mathbf{R}) |\Psi_q(\mathbf{R})\rangle = E_e(\mathbf{R}) |\Psi_q(\mathbf{R})\rangle.$$  

(2)

Here, $\hat{H}_e(\mathbf{R})$ is the electronic Hamiltonian, $\mathbf{R}$ is the nuclear configuration of interest, $E_e(\mathbf{R})$ is the electronic energy, $\hat{H}_q(\mathbf{R})$ is the qubit Hamiltonian obtained from a second quantized form of $\hat{H}_e(\mathbf{R})$\textsuperscript{6} using one of the fermion-qubit mappings,\textsuperscript{7–11} and $|\Psi_q(\mathbf{R})\rangle$ is the corresponding qubit wave-function. For notational simplicity, in what follows, we will skip the nuclear configuration but will always assume its existence as a parameter.

In VQE, the quantum computer prepares a trial qubit wavefunction $|\Psi_q\rangle$ and then does measurements to accumulate statistics for the expectation value of the qubit Hamiltonian. The classical computer completes the VQE cycle by suggesting a new trial wavefunction based on previous expectation values of energy. The two steps, on classical and quantum computers, are iterated until convergence. One of the strengths of the VQE approach is ability to use relatively short-depth quantum circuits to construct qubit wavefunction $|\Psi_q\rangle$ that is a good approximation for the true eigenstate of the problem. Note though that the VQE scheme cannot measure the whole system Hamiltonian at once, because the system Hamiltonian is not the Hamiltonian of qubits and is not physically implemented in the quantum computer. This is one of the differences between universal quantum computing and quantum simulation.\textsuperscript{12,13}

Measuring parts of the system Hamiltonian is a very time-consuming task. Experimentally, one can only measure single-qubit Pauli operators, $\hat{\sigma}_i = \hat{x}_i, \hat{y}_i$ or $\hat{z}_i$. A regular qubit Hamiltonian

$$\hat{H}_q = \sum_i C_i \hat{P}_i$$

(3)

is a linear combination of products of Pauli operators $\hat{P}_i$ (Pauli “words”) for different qubits,

$$\hat{P}_i = \prod_{i=1}^N \hat{\sigma}^{(i)}_i,$$

(4)

where $\hat{\sigma}^{(i)}_i$ is one of the $\hat{x}, \hat{y}, \hat{z}$ Pauli operators or the identity $\hat{1}$ operator for the $i^{th}$ qubit, and $N$ is the total number of qubits. For single-qubit measurements one can group only those terms that share a common tensor product eigen-basis. Thus, during the measurement, the system wavefunction can collapse to a set of entangled eigenstates common to all Pauli operators in the group. A simple criterion for grouping terms based on shared tensor product eigen-basis is their mutual commutativity within single-qubit subspaces or qubit-wise commutativity.\textsuperscript{14}

The total number of terms in the qubit Hamiltonian scales as the fourth power of the number of qubits needed to represent the electronic wavefunction. Even though
Recently we have proposed an efficient grouping technique based on a mapping of the qubit Hamiltonian to a graph, the best grouping technique can only reduce the total number of simultaneously measurable parts by factor of 3 from the total number of terms in Eq. (3), which still leaves a large number of groups to be measured.\textsuperscript{14}

Another way to reduce the number of separately measured groups has been suggested recently in Ref. 15, where the idea of the single-qubit measurement was generalized to the case when the result of one qubit measurement was used to determine what single-qubit operator needs to be measured next. Partitioning of the qubit Hamiltonian into fragments that can be measured with such feed-forward measurement procedures increased the number of terms that can be grouped together and thus reduced the number of separately measured groups. However, even though such feed-forward measurements were demonstrated in some experiments\textsuperscript{16–19} they have not yet become available in mainstream quantum computing hardware available to the public. Another difficulty with this approach is that a procedure for ensuring the optimality of this partitioning has yet to be found.

Here we explore a different route to the Hamiltonian partitioning, which is based on an idea that if the Hamiltonian were unitary operator \( \hat{U}_H \) its expectation value could be obtained in one set of single-qubit measurements. Although the qubit Hamiltonian is not a single unitary operator, its individual Pauli products in Eq. (3) are unitary operators and it is possible to combine them under certain conditions to larger groups of unitary operators that each can be measured as a single set. Optimal grouping of such unitary fragments is possible through solving a minimum clique cover problem for a specially constructed graph of the qubit Hamiltonian.

The rest of the paper is organized as follows. In Sec. II A we develop a partitioning of the qubit Hamiltonian to a minimal number of unitary fragments. Section II B details quantum computing circuit for measuring expectation values for these unitary fragments. Assessment of the new scheme is done on a set of molecular systems with the number of terms in \( H_q \) up to fifty thousands (Sec. III). Section IV summarizes main results.

II. THEORY

A. Unitary Partitioning

Here we will discuss how to partition the qubit Hamiltonian into a linear combination of the minimum number of unitary operators

\[ \hat{H}_q = \sum_n d_n \hat{U}_n, \tag{5} \]

where \( d_n \) is a set of real coefficients, and \( \hat{U}_n \) are \( M \) unitary operators.

Note that all Pauli words are hermitian unitary operators, \( \hat{P}_I \hat{P}_I = \hat{P}_I^2 = 1 \). However, a general sum of unitary operators is non-unitary

\[ \left( \sum_n C_n \hat{P}_n \right) \left( \sum_n C_n \hat{P}_n \right) \neq 1. \tag{6} \]

To make \( \sum_n C_n \hat{P}_n \) unitary, it is sufficient to impose the following three additional conditions: 1) \( \text{Im}(C_n^* C_J) = 0 \), 2) \( \sum_n |C_n|^2 = 1 \), and 3) \( \{ \hat{P}_I, \hat{P}_J \} = 2 \delta_{IJ} \) (where \( \{,\} \) is the anti-commutator). The first two conditions are easy to satisfy for any partial sum of the Hamiltonian in Eq. (3) because all coefficients are real so only their renormalization is required

\[ \sum_n C_n \hat{P}_n = C \sum_n C_n \hat{P}_n = C \left( \sum_n C_n^2 \right)^{1/2}, \tag{7} \]

then the first two conditions for unitarity will be satisfied for the sum with coefficients \( C_n/C \).

To satisfy the third condition, one needs to partition the Hamiltonian into groups of Pauli matrices that mutually anti-commute. To reduce the number of unitary operators needed to represent \( H_q \) in Eq. (5), we would like to maximize the number of mutually anti-commuting terms in each group. Recently, it was found that a similar problem of finding minimum partitioning into groups of mutually qubit-wise commuting terms can be solved using a graph representation for the Hamiltonian.\textsuperscript{14} There, every Pauli word was considered as a graph vertex and edges were put between the terms that qubit-wise commute. The grouping problem is equivalent to the very well-known minimum clique cover problem. For the anti-commuting sets, one can also build a graph representation of the Hamiltonian where two Pauli word vertices are connected if the corresponding operators anti-commute. Since two Pauli words always either commute or anti-commute, the anti-commutativity graph is complementary for the commutativity graph. Thus, for the further discussion we will assume that solving the minimum clique cover problem for the Hamiltonian anti-commutativity graph provides the minimum number of \( \hat{U}_n \) operators

\[ \hat{U}_n = \frac{1}{d_n} \sum_n C_I^{(n)} \hat{P}_I^{(n)}, \tag{8} \]

\[ d_n = \left( \sum_n (C_I^{(n)})^2 \right)^{1/2}, \tag{9} \]

where \( \{ \hat{P}_I^{(n)}, \hat{P}_J^{(n)} \} = 2 \delta_{IJ} \).

B. Unitary Operator Measuring Circuit

Partitioning of the \( H_q \) in Eq. (5) allows us to rewrite the energy expectation value as

\[ \bar{E} = \langle \Psi | \hat{H}_q | \Psi \rangle = \sum_n d_n \langle \Psi | \hat{U}_n | \Psi \rangle. \tag{10} \]
Accounting for a unitary preparation of the wavefunction $|\Psi\rangle = U |\bar{0}\rangle$, where $|\bar{0}\rangle$ is $N$ qubit vacuum or initial all-qubits-up state. For measuring, it is convenient to rewrite $\hat{E}$ in a symmetric form as

$$\hat{E} = \frac{1}{2} \sum_n d_n (\langle \Psi | U_n | \Psi \rangle + \langle \Psi | U_n^\dagger | \Psi \rangle). \quad (11)$$

By introducing $|\Phi_n\rangle = \hat{U}^\dagger U_n |\bar{0}\rangle$ states the energy estimate can be written as

$$\hat{E} = \frac{1}{2} \sum_n d_n (\langle 0 | \Phi_n \rangle + \langle \Phi_n | 0 \rangle). \quad (12)$$

In what follows we will discuss how to measure the individual components

$$\langle 0 | \Phi_n \rangle + \langle \Phi_n | 0 \rangle = 2 \text{Re} \langle 0 | \Phi_n \rangle, \quad (13)$$

which are directly connected to the energy estimate:

$$\hat{E} = \sum_n d_n \text{Re} \langle 0 | \Phi_n \rangle. \quad (14)$$

To measure the real part of the overlap $\langle 0 | \Phi_n \rangle$ we will not use the swap test because this test produces the absolute value of the overlap instead of its real part. Our approach to evaluating $\text{Re} \langle 0 | \Phi_n \rangle$ will be as follows (see Fig. 1). The initial state is a tensor product $|0\rangle \otimes |a\rangle$ of one ancilla and $N$ target qubits. First, the Hadamard gate $H = (\hat{x} + \hat{z})/\sqrt{2}$ is applied to an ancilla qubit

$$|\Psi_1\rangle = |\bar{0}\rangle \otimes (|0\rangle_a + |1\rangle_a)/\sqrt{2}. \quad (15)$$

Second, using a controlled unitary operator $\hat{U}^\dagger U_n \hat{U}$ the following superposition is created

$$|\Psi_2\rangle = (|\bar{0}\rangle \otimes |0\rangle_a + |\Phi_n\rangle \otimes |1\rangle_a)/\sqrt{2}. \quad (16)$$

Third, another Hadamard gate rotates the $|\Psi_2\rangle$ state into

$$|\Psi_3\rangle = \frac{1}{2} [|\Phi_{n+}\rangle \otimes |0\rangle_a + |\Phi_{n-}\rangle \otimes |1\rangle_a], \quad (17)$$

where $|\Phi_{n\pm}\rangle = |\bar{0}\rangle \pm |\Phi_n\rangle$. Then, the expectation values of all $\hat{z}_n$ operators are measured on $|\Psi_3\rangle$. The measurement of $\hat{z}_n$ for the ancilla qubit collapses the wavefunction to the $|\Phi_{n\pm}\rangle$ superpositions for the target qubits with the equal probabilities. Based on the ancilla qubit result $(\pm 1)$ we can separate outcomes of the operator $\hat{Z} = \sum_{n=1}^N \hat{z}_n$ measurements for states $|\Phi_{n\pm}\rangle$ and obtain the expectation values

$$Z_{n\pm} = \frac{\langle \Phi_{n\pm} | \hat{Z} | \Phi_{n\pm} \rangle}{\langle \Phi_{n\pm} | \Phi_{n\pm} \rangle}. \quad (18)$$

Both numerator and denominator of $Z_{n\pm}$ contain $\text{Re} \langle 0 | \Phi_n \rangle$, by doing some simple algebra involving relations like $\hat{Z} |\bar{0}\rangle = N |\bar{0}\rangle$ one can express $\text{Re} \langle 0 | \Phi_n \rangle$ as

$$\text{Re} \langle 0 | \Phi_n \rangle = \frac{Z_{n+} - Z_{n-}}{2N - Z_{n+} - Z_{n-}}. \quad (19)$$

FIG. 1. Circuit for extracting values of $\text{Re} \langle 0 | \Phi_n \rangle$: it requires $N+1$ qubits and $M$ series of $N+1$ single-qubit measurements.

C. Circuit depth analysis

How can one implement the controlled $\hat{U}^\dagger U_n \hat{U}$ transformation on a quantum computer? Any $\hat{U}$ can be presented as a product of one- and two-qubit operators for a regular VQE circuit to generate a trial wavefunction. If $\hat{U}_n = \sum_{k=1}^L c_k \hat{R}_k$, where $\sum_{k=1}^L c_k^2 = 1$, using the anticommutativity of terms, this sum can be presented as a product of $2L - 1$ exponents of Pauli words (entanglers)

$$\hat{U}_n = \prod_{k=1}^{L-1} (\hat{e}^{i\theta_k \hat{R}_k/2}) = \hat{e}^{i\theta_L \hat{P}_L} \prod_{k=L-1}^1 e^{i\theta_k \hat{P}_k/2}, \quad (20)$$

where $\theta_k$’s can be connected with $c_k$’s as

$$\theta_k = \arcsin \frac{c_k}{\sqrt{\sum_{j=1}^L c_j^2}}. \quad (21)$$

This connection is easy to understand from a geometric point of view for $c_k$’s as Cartesian coordinates of a point on a unit $L - 1$-dimensional sphere and $\theta_k$’s as corresponding hyper-spherical coordinate components. Therefore, compare to $\hat{U}$, the new transformation $\hat{U}_n^\dagger U_n \hat{U}$ in the worst case (no significant cancellation between terms in the product) will have twice as many terms in addition to $2L - 1$ terms generated from $\hat{U}_n$. The $2L - 1$ entanglers are not necessarily one- and two-qubit operators, but they can be expanded as products of those.

Each transformation involved in the product $\hat{U}_n^\dagger U_n \hat{U}$ needs to be implemented as a controlled operation, which increases the gate qubit-count by one. Still, products of two- and three-qubit entanglers can be always expressed as products of one- and two-qubit operators.

To implement the controlled $\hat{U}_n^\dagger U_n \hat{U}$, all one-qubit can be replaced by operators with controlled-U gates. For the two-qubit operators, we can find decompositions in CNOT and one-qubit gates, which are then replaced by Toffoli and controlled-U gates. Hence, implementing the controlled $\hat{U}_n^\dagger U_n \hat{U}$ is not asymptotically more expensive than implementing $\hat{U}_n^\dagger U_n \hat{U}$.

D. Application to the projection formalism

To impose physical symmetries one can construct projectors on irreducible representations of the symmetry
All these heuristics except BKT have polynomial computational scaling with respect to the number of graph vertices.

\[ \hat{P} = \sum_k a_k \hat{U}_k, \]

and can be applied in the expectation values of the projected Hamiltonian

\[ \bar{E} = \frac{\langle \Psi | \hat{P}^\dagger \hat{H}_q \hat{P} | \Psi \rangle}{\langle \Psi | \hat{P}^\dagger \hat{P} | \Psi \rangle} \]

\[ \bar{E} = \frac{\langle \Psi | \hat{H}_q \hat{P} | \Psi \rangle}{\langle \Psi | \hat{P} | \Psi \rangle}. \]

Here, the last equation used hermiticity, idempotency, and commutativity with the Hamiltonian for the symmetry projector. The expansions in unitary transformations for the projector [Eq. (22)] and the Hamiltonian [Eq. (5)] can be easily combined because a product of two unitary operators is unitary. Even though introducing the projector expansion will increase the number of terms for the measurement, it allows us to reduce the complexity of the unitary transformation for the preparation of |\Psi\rangle by satisfying symmetry requirements by construction.\(^22\)

### III. NUMERICAL STUDIES AND DISCUSSION

To assess our developments we apply them to several small molecule Hamiltonians (Tables I and II). Details of generating these Hamiltonians are given in Supplementary Information. Some of these systems were used to illustrate performance of quantum computing techniques previously.\(^20,23,24\)

To solve the minimum clique cover problem we have used several heuristic algorithms based on either reformulating the problem as graph coloring or approximating it as finding and removing maximum cliques.\(^14\) The description of used heuristics can be found in Ref. 14 and original papers: Greedy Coloring (GC),\(^25\) Largest First (LF),\(^26\) Smallest Last (SL),\(^27\) DSATUR,\(^28\) Recursive Largest First (RLF),\(^29\) Dutton and Brigham (DB),\(^30\) COSINE,\(^31\) Ramsey,\(^32\) Bron-Kerbosch-Tomita (BKT).\(^33\) All these heuristics except BKT have polynomial computational scaling with respect to the number of graph vertices.

Table I summarizes results of the anti-commuting partitioning and compares it with previously used qubit-wise commutativity (QWC) partitioning. Fitting Table I data in the double log-scale reveals \(N^3\) scaling of the total number of terms in Hamiltonians and the number of QWC groups with the number of qubits, \(N\). Deviation from asymptotic \(N^4\) scaling is attributed to insignificantly large size of systems and thus a non-negligible contribution of the one-electron integral part that has \(N^2\) scaling. The number of anti-commuting groups scales only as \(N^2\). The advantage of partitioning to anti-commuting groups can be rationalized from the graph connectivity point of view.

IV. CONCLUSIONS

We have introduced and studied a new method for partitioning of the qubit Hamiltonian to a linear combination of unitary transformations. This unitary partitioning allows us to reduce the number of separate measurements required in the VQE approach to the electronic structure problem. The grouping produces \(N\)-fold reduction in the number of operators that require separate measurements. The unitary partitioning scheme has increased depth of quantum circuits. For measuring an anti-commuting group of terms containing \(L\) elements on a trial wavefunction prepared using \(K\) entanglers, the depth of a new circuit becomes at least \(2K + 2L - 1\) entanglers.

The partitioning of the qubit Hamiltonian is done by representing it as a graph where every vertex corresponds to a single Pauli word and the edges are connecting the terms that are anti-commuting. In this representation, the problem of grouping terms that can form a unitary operator corresponds to finding a fully connected subgraphs (cliques). To obtain optimal partitioning the number of groups should be the fewest. This is a well-known problem in discrete math, the minimum clique cover problem, which is solved using polynomial heuristic algorithms.

Among various tested heuristics, the RLF approach is found to be the most efficient polynomial algorithm producing the lowest number of fully anti-commuting groups. Hamiltonians produced using different fermion-qubit transformations (JW and BK) had similar compression rates due to the unitary partitioning.

Another advantage of the unitary partitioning is its straightforward incorporation of the symmetry projections that can always be presented as linear combinations of unitary operators.

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TABLE I. The number of qubits \((N)\), Pauli words in qubit Hamiltonians (Total), QWC groups \((M_{QWC})\), and anti-commuting groups \((M)\) produced by different heuristics for systems with up to 14 qubits. The STO-3G basis has been used for all Hamiltonians unless specified otherwise.

| Systems | \(N\) | Total | \(M_{QWC}\) | GC | LF | SL | DSATUR | RLF | DB | COSINE | Ramsey | BKT |
|---------|------|-------|-------------|----|----|----|--------|-----|----|--------|--------|-----|
| \(\text{H}_2\) (BK) | 4 | 15 | 3 | 11 | 11 | 11 | 11 | 11 | 11 | 11 | 11 | 11 |
| \(\text{LiH}\) (Parity) | 4 | 100 | 25 | 33 | 33 | 11 | 11 | 11 | 11 | 11 | 11 | 11 |
| \(\text{H}_2\)O (6-31G, BK) | 6 | 165 | 34 | 41 | 41 | 41 | 41 | 41 | 41 | 41 | 41 | 41 |
| \(\text{BeH}_2\) (BK) | 14 | 666 | 172 | 141 | 130 | 135 | 121 | 119 | 119 | 119 | 119 | 119 |
| \(\text{BeH}_2\) (JW) | 14 | 666 | 203 | 139 | 135 | 121 | 119 | 110 | 110 | 110 | 110 | 110 |
| \(\text{H}_2\)O (BK) | 14 | 1086 | 308 | 176 | 147 | 154 | 127 | 127 | 127 | 127 | 127 | 127 |
| \(\text{H}_2\)O (JW) | 14 | 1086 | 322 | 181 | 197 | 159 | 153 | 127 | 127 | 127 | 127 | 127 |

TABLE II. Comparison of RLF results for BK and JW transformed Hamiltonians: the number of anti-commuting cliques \((M)\), their maximum size (Max Size), and standard deviation of their size distributions (STD). The total number of Hamiltonian terms (Total) is almost everywhere the same for JW and BK; for the last two systems, JW numbers are in parenthesis.

| Systems | \(N\) | Total | BK | \(M\) | Max Size | STD | JW | \(M\) | Max Size | STD |
|---------|------|-------|-----|-------|--------|-----|-----|-------|--------|-----|
| \(\text{BeH}_2\) / STO-3G | 14 | 666 | 112 | 10 | 2.0 | 110 | 11 | 2.1 |
| \(\text{H}_2\)O / STO-3G | 14 | 1086 | 127 | 13 | 2.0 | 127 | 15 | 2.2 |
| \(\text{NH}_3\) / STO-3G | 16 | 3609 | 251 | 25 | 3.9 | 251 | 25 | 3.8 |
| \(\text{N}_2\) / STO-3G | 20 | 2951 | 266 | 17 | 2.5 | 268 | 19 | 2.6 |
| \(\text{BeH}_2\) / 6-31G | 26 | 9204 | 556 | 26 | 4.5 | 558 | 29 | 4.6 |
| \(\text{H}_2\)O / 6-31G | 26 | 12732 | 767 | 33 | 5.2 | 779 | 32 | 5.2 |
| \(\text{NH}_3\) / 6-31G | 30 | 52758 (52806) | 1761 | 50 | 7.8 | 1781 | 50 | 7.8 |
| \(\text{N}_2\) / 6-31G | 36 | 34639 (34655) | 1402 | 43 | 5.9 | 1399 | 46 | 5.9 |

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SUPPLEMENTARY INFORMATION: HAMILTONIAN GENERATION

$H_2$ molecule: One- and two-electron integrals in the canonical restricted Hartree–Fock (RHF) molecular orbitals basis for $R$(H-H)$=1.5$ Å, were used in the Bravyi–Kitaev (BK) transformation to produce the corresponding qubit Hamiltonian. Spin-orbitals were alternating in the order $\alpha, \beta, \alpha, \ldots$.

LiH molecule: Using the parity transformation for the LiH molecule at $R$(Li – H)$=3.2$ Å, a 6-qubit Hamiltonian containing 118 Pauli words was generated. Spin-orbitals were arranged as “first all $\alpha$ then all $\beta$” in the fermionic form; since there are 3 active molecular orbitals in the problem, this leads to 6-qubit Hamiltonian. This qubit Hamiltonian has 3rd and 6th stationary qubits, which allowed us to replace the corresponding $\hat{z}$ operators by their eigenvalues, $\pm 1$, thus defining the different “sectors” of the original Hamiltonian. Each of these sectors is characterized by its own 4-qubit effective Hamiltonian. The ground state lies in the $z_3 = -1$, $z_6 = 1$ sector; the corresponding 4-qubit effective Hamiltonian ($\hat{H}_{\text{LiH}}$) has 100 Pauli words.

$H_2O$ molecule: 6- and 26-qubit Hamiltonians were generated for this system in the 6-31G basis, and the 14-qubit Hamiltonian was generated using the STO-3G basis. The geometry for all Hamiltonians was chosen to be $R$(O – H)$=0.75$ Å and $\angle\text{HOH}$ = 107.6$°. The 14- and 26-qubit Hamiltonians were obtained in OpenFermion using both JW and BK transformations without any modifications, while for the 6-qubit Hamiltonian we used several qubit reduction techniques detailed below.

Complete active space (4, 4) electronic Hamiltonian was converted to the qubit form using the BK transformation grouping spin-orbitals as “first all alpha than all beta”. The resulting 8-qubit Hamiltonian contained 185 Pauli terms. 4th and 8th qubits were found to be stationary; the ground state solution is located in the $z_3 = 1$, $z_7 = 1$ subspace. By integrating out $z_3$ and $z_7$, the 6-qubit reduced Hamiltonian with 165 terms was derived.

$N_2$, BeH$_2$, and NH$_3$ molecules: The BK and JW transformations of the electronic Hamiltonian in the 6-31G and STO-3G bases produced qubit Hamiltonians by OpenFermion. The nuclear geometry was fixed at $R$(N – N) = 1.1 Å(N$_2$); $R$(Be – H) = 1.4 Å, collinear geometry (BeH$_2$); $\angle\text{HNH}$ = 107$°$ and $R$(N – H) = 1.0 Å(NH$_3$).

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