

A Variational Quantum Eigensolver Based on the Measurement Scheme Tailored to Multiband Tight-Binding Simulations

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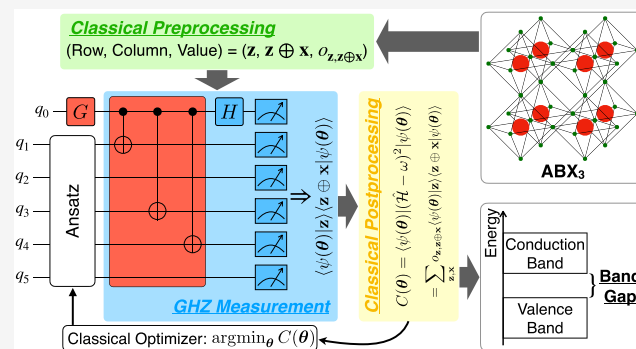
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ABSTRACT: We propose a cost-efficient measurement scheme of the variational quantum eigensolver (VQE) for atomistic simulations of electronic structures based on a tight-binding (TB) theory. Leveraging the lattice geometry of a material domain, the sparse TB Hamiltonian is constructed in a bottom-up manner and is represented as a linear combination of the standard-basis (SB) operators. The cost function is evaluated with an extended version of the Bell measurement circuit that can simultaneously measure multiple SB operators and therefore reduces the number of circuits required by the evaluation process. The proposed VQE scheme is applied to find band gap energies of metal-halide-perovskite supercells that have finite dimensions with closed boundaries and are described with an sp^3 TB model. Experimental results confirm that the proposed scheme gives solutions that follow well the accurate ones but, more importantly, has computing efficiency that is obviously superior to the commutativity-based Pauli grouping methods. Extending the application scope of VQE to three-dimensional confined atomic structures, this work can serve as a practical guideline for handling TB simulations and, more generally, for calculations of sparse Hermitian matrices in the noise-intermediate-scale quantum devices.



1. INTRODUCTION

Quantum computers have been expected to efficiently simulate diverse many-body quantum systems, having the potential to surpass the performance of classical computers.¹ It was first proposed that the eigenvalue problem of a given Hamiltonian can be tackled with a universal quantum computer based on the quantum phase estimation (QPE) algorithm.² Though the QPE algorithm shows an exponential speedup that is not easy to be secured with classical computers,^{3,4} it requires many controlled unitary gates to solve large-scale systems so the fault-tolerant quantum computer is essential. Despite of ongoing significant development in quantum hardware and software, QPE is not yet feasible in near-term quantum computers that lie in the noisy intermediate-scale quantum (NISQ) regime,⁵ motivating strong needs for quantum-classical hybrid algorithms for leveraging NISQ devices. Ever since its initial proposal for tackling small-size molecular Hamiltonians,⁶ the variational quantum eigensolver (VQE) has been the most extensively utilized and developed algorithms over the past decade for simulations of electronic structures in NISQ computers with support of classical computing.^{7–9} Although the VQE algorithm has challenges such as barren plateaus¹⁰ and NP-hardness of the classical optimization process,¹¹ alternative or breakthrough strategies tailored to specific conditions have been devised to circumvent these

difficulties,¹² so VQE still belongs to practical applications that can exploit NISQ computers.

Many studies on quantum algorithms involving VQE and QPE so far have predominantly investigated properties of molecular systems or lattice models by encoding qubits with second quantization.^{8,9} However, application studies based on encoding with first quantization still remain relatively rare in the context of VQE,⁸ though, from a resource perspective, the use of first quantization can be beneficial for computing electron systems in certain cases.^{4,13} A good candidate for computation based on first quantization is the empirical simulation of nanoscale materials,^{14–19} whose focal characteristic is to describe a single-electron Hamiltonian with a fixed set of parameters. In particular, simulations with the atomistic tight-binding (TB) model have been extensively employed to study electrical properties of realistically sized solid crystalline structures that is in principle hard to be done with ab initio simulations, such as wavelength engineering of quantum dot

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devices,^{20,21} transport characteristics of nanowire transistors,^{22,23} and designs of novel electronic devices like atomic-scale interconnects or single electron transistors.^{24–26}

Recently, TB simulations have also gained attention as a sound application of VQE.^{27–29} Two previous works focused on the calculation of single-band TB Hamiltonians with second quantization that eliminates the need for intricate Fermion-to-Pauli mappings such as the Jordan–Wigner and the Bravyi–Kitaev transformations.^{28,29} They however require $O(D)$ qubits to tackle Hamiltonians of dimension D so challenge in scalability may exist in NISQ devices. Cerasoli et al.²⁷ computed the Silicon bulk band structure with $O(\log D)$ qubits, by representing the TB Hamiltonian with first quantization. But, the target problem is rather small (a single primitive unit cell consisting of two atoms), and, more importantly, the Hamiltonian is mapped with the most primitive Pauli decomposition method that can in principle require the computation of up to $4^{O(\log D)}$ coefficients.

So in this work, we propose a new variant of VQE, focusing on cost-efficient handling of TB Hamiltonians with a set of the first-quantized atomic orbitals. By leveraging the geometric information on three-dimensional (3D) finite-size supercells, we first show that the bottom-up construction of the Hamiltonian matrix can be done with a set of standard-basis (SB) operators that is known to be useful for description of any vectors and matrices.³⁰ Then, we discuss a measurement scheme that computes the cost function designed to determine the band gap energy of a confined supercell. To reduce the number of quantum circuits required to evaluate the cost function, we group SB operators obtained with decomposition of the TB Hamiltonian and employ the Greenberger–Horne–Zeilinger (GHZ) measurement that is a generalized version of the Bell measurement.^{31–33} Results reveal that our measurement scheme remarkably outperforms Pauli-based approaches because the cost of SB decomposition and grouping is linearly proportional to the number of nonzero elements in the Hamiltonian matrix, and our measurement circuit, requiring at most N CNOT gates (N = the number of qubits in the circuit), can be more concise than the one that simultaneously measures commuting Pauli strings.³⁴ Finally, we verify the proposed method by conducting numerical simulations to find the band gap energy of 3D metal-halide-perovskite supercells that are described with a sp^3 TB model.^{19,21}

2. METHODS

2.1. TB Model and Hamiltonian. The TB model is one of the theoretical frameworks used to describe electronic structures of crystalline materials. It is based on the assumptions that a single electron occupies localized atomic orbitals on individual atoms residing in a material structure and that the overlap of atomic orbitals occurs only between nearest neighboring atoms. The TB Hamiltonian in the first-quantized basis $\{|n, i\rangle\}$, where n and i indicate the orbital index and atomic site, respectively, can be written as

$$\hat{H} = \sum_{n', n, i, i'} t_{n'n}^{i'i} |n', i'\rangle \langle n, i| \quad (1)$$

where $t_{n'n}^{i'i} = \langle n', i' | \hat{H} | n, i \rangle$ is the single-electron integral involving the on-site energy of each atoms and the overlap energies between two nearest atoms. The element $t_{n'n}^{i'i}$ is then given as

$$t_{n'n}^{i'i} = \int d\mathbf{r} \psi_n^*(\mathbf{r}, \mathbf{R}_i) \hat{H}(\mathbf{r}) \psi_n(\mathbf{r}, \mathbf{R}_i) \quad (2)$$

where $\psi_n(\mathbf{r}, \mathbf{R}_i)$ denotes the basis function that describes a single electron in n th localized atomic orbital with the lattice vector $\mathbf{R}_i$. Since atoms are assumed to be tightly coupled, the overlap integral $t_{n'n}^{i'i}$ is zero when $i' \neq i$ except the case that i' th atom belongs to nearest neighbors of the i th atom, and therefore $\hat{H}$ becomes a sparse matrix. Due to the translational symmetry of crystalline solid atomic structures, $\mathbf{R}_i$ for all atomic indices i in the lattice can be spanned with the primitive translation vectors. Usually, the $t_{n'n}^{i'i}$ terms of the TB Hamiltonian in eqs 1 and 2 are empirically secured by fitting the bulk band structure that is known by either experiments or ab initio calculations. Since the on-site and coupling overlap integrals are dictated by atomic types and positions, we can easily identify nonzero elements and their row and column indices in the TB Hamiltonian by examining the types and primitive vectors of all the atoms from the geometric information (atomic species and positions) of the supercell composed of a given material.

2.2. Measurement Scheme for Expectation Value. To determine eigenpairs of a given matrix, VQE needs the expectation value of the observable that involves the target matrix. To evaluate the expectation value with quantum circuits, the observable needs to be decomposed into a weighted sum of operators that can be readily manipulated with gate operations. Hamiltonians describing electronic structures based on a set of orthogonal basis are always Hermitian, and can be typically expressed as a linear combination of Pauli operators with approaches like the Jordan–Wigner transformation.³⁵ As the Hamiltonian becomes larger in size, however, the number of associated Pauli strings rapidly increase,³⁶ so strategies for grouping Pauli strings have been adopted to reduce the effective number of Pauli strings with the general commutativity (GC), qubit-wise commutativity (QWC), and unitary partitioning.^{37–43} A cost-efficient circuit that can simultaneously measure k commuting Pauli strings with up to kN CNOT gates, has been also reported.³⁴ In this subsection, we present the methodological details of our measurement scheme and grouping strategy that are well-suited for sparse TB Hamiltonian matrices with better performance than existing approaches.

2.2.1. Representation of TB Hamiltonian with SB Operators. The SB in linear algebra, a set of L -dimensional vectors having a single nonzero element of 1, can span the entire vector space $\mathbb{R}^L$ or $\mathbb{C}^L$. The computational basis $\{|z\rangle\}$ for N qubits in quantum computing is nothing but the SB of the pure quantum state, where $z \equiv z_1 \cdots z_N$ for each $z_j \in \{0, 1\}$. Adopting this basis in the quantum circuit, we can define a SB operator $\{|z\rangle\langle z'|$ whose functionality is to map one computational basis state to another. Notably, any N -qubit SB operator can be factorized as a tensor product of single-qubit SB operators, which is given by

$$|z\rangle\langle z'| = \bigotimes_{j=1}^N |z_j\rangle\langle z'_j| \quad (3)$$

where z_j denotes the binary value of the j th qubit and $|z_j\rangle\langle z'_j|$ represents the SB for single-qubit operators at the j th site. These SB operators can obviously facilitate the representation of an arbitrary observable $\hat{O}$ as follows

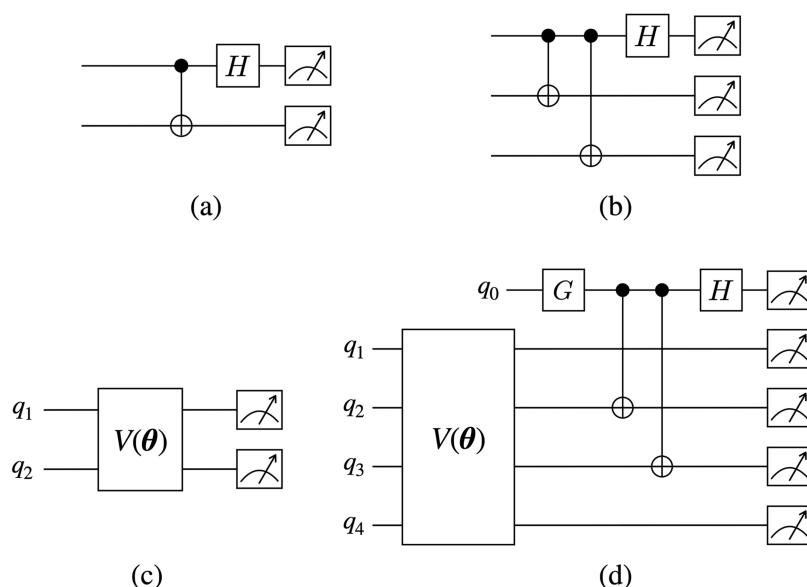


Figure 1. Circuits for the (a) Bell measurement and (b) three-qubit GHZ measurement, and circuits for estimation of (c) $\langle \psi | z_1 z_2 \rangle \langle z_1 z_2 | \psi \rangle$ and (d) $\langle \psi | z_1 z_2 z_3 z_4 \rangle \langle z_1 z_2 z_3 z_4 | \psi \rangle$. Non-Hermitian operators can be measured by applying the GHZ measurement to the tensor product of $G|0\rangle$ and $|\psi\rangle = V(\theta)|0\rangle$, where $G \in \{H, S\cdot H\}$ and $V(\theta)$ is an ansatz circuit.

$$\hat{O} = \sum_{\mathbf{z}, \mathbf{z}'} o_{\mathbf{z}, \mathbf{z}'} |\mathbf{z}\rangle \langle \mathbf{z}'| \quad (4)$$

where $o_{\mathbf{z}, \mathbf{z}'}$'s are complex coefficients. Representation of sparse TB Hamiltonians with eq 4 is then straightforward since the row and column index of a specific nonzero element can be directly mapped to bit strings $\mathbf{z}$ and $\mathbf{z}'$ by converting indices to binary values, and the coefficient $o_{\mathbf{z}, \mathbf{z}'}$ indicates the corresponding nonzero value. Following this scheme, we only need to map up to M terms if a given Hamiltonian has M nonzero elements. Consequently, in general, the SB-based strategy in eqs 3 and 4 can be much more efficient in terms of computing cost than the Pauli decomposition whose cost grows exponentially with the number of qubits needed to describe the target Hamiltonian.³⁶

2.2.2. GHZ Measurement Circuit. The Bell measurement, being also known as the Bell state measurement, is a joint measurement scheme that projects two qubits onto one of the four Bell states: $\{|00\rangle \pm |11\rangle, |01\rangle \pm |10\rangle\}$. In a quantum circuit, the Bell measurement can be implemented using a sequence of a CNOT gate, a Hadamard (H) gate, and Z-basis measurements, as shown in Figure 1(a). The Bell measurement can be extended to multiqubit (>2) joint measurements, analogous to the way that Bell states are generalized to multiqubit GHZ states that have the genuine multipartite entanglement. The sequence of quantum gates needed to implement a circuit for the GHZ measurement can be expressed as follows

$$U_{\text{GHZ}} = H_0 \left(\prod_{j \in X} CX_{0,j} \right) \quad (5)$$

where $CX_{0,j}$ represents the CNOT gate with the (control, target) qubit at the (0th, j th) site, and X denotes the set of qubits employed as target qubits of CNOT gates. Conducting Z-basis measurements at the end of an N -qubit U_{GHZ} yields one of 2^N different binary strings that determines the type of projection driven by the entire circuit ($U_{\text{GHZ}} + \text{Z-basis}$

measurement). In the three-qubit circuit shown in Figure 1(b) as an example, an outcome of 010 leads to the GHZ measurement being performed as $U_{\text{GHZ}}|010\rangle \langle 010| U_{\text{GHZ}}^\dagger = (|010\rangle + |101\rangle) \langle 010| + \langle 101|) / 2$.

2.2.3. Expectation Value of SB Operator. The GHZ measurement can be used to evaluate the expectation value of a SB operator with an n -qubit ansatz $v(\theta)$ and a state $|\psi\rangle \equiv v(\theta)|0\rangle$. According to eq 3, it is easy to get the expectation value of SB operators that consist solely of projective measurements $|z_j\rangle \langle z_j|$ on individual qubits, as depicted in Figure 1(c) that indicates the expectation value of the SB operator $|z\rangle \langle z|$ is equal to the probability $p(\mathbf{z})$ of the outcome $\mathbf{z}$.

If non-Hermitian operators $|z_j\rangle \langle z_j \oplus 1|$ act on certain qubits in a given SB operator, the expectation value of the operator can be computed with an additional ancilla qubit followed by a single-qubit logic $G \in \{H, S\cdot H\}$ (a single H gate or a sequence of the H & S gate) as shown in Figure 1(d). The GHZ measurement is then applied to this $(N+1)$ -qubit circuit where the ancilla qubit acts as a control qubit for up to N CNOT gates in U_{GHZ} and some of the remaining N qubits are used as target qubits, belonging to the set X defined in eq 5. Here, the set X is determined by the SB operator whose expectation value is to be estimated: $X \equiv \{j | x_j = 1 \text{ for } \mathbf{x}, 1 \leq j \leq N\}$ for a given SB operator $|z\rangle \langle z'|$, where $\mathbf{z}' \equiv \mathbf{z} \oplus \mathbf{x}$ with a bit string $\mathbf{x} \equiv x_1 \cdots x_N$. If a SB operator is $|0101\rangle \langle 0011|$, for example, $\mathbf{x} = 0110$ contributes to the set $X = \{2, 3\}$ so U_{GHZ} has two CNOT gates (see Figure 1(d)). Performing the GHZ measurement on this $(N+1)$ -qubit circuit where the ancilla qubit is the zeroth qubit, we get the probabilities that the $(N+1)$ -bit outcome is $0z_1 \cdots z_N$ ($0\mathbf{z}$) or $1z_1 \cdots z_N$ ($1\mathbf{z}$). These probabilities are used to obtain the expectation value of the SB operator $|z\rangle \langle z'|$ as follows

$$\langle \psi | z \rangle \langle z' | \psi \rangle = [p_H(0\mathbf{z}) - p_H(1\mathbf{z})] - i[p_{SH}(0\mathbf{z}) - p_{SH}(1\mathbf{z})] \quad (6)$$

where p_H and p_{SH} are the probabilities obtained with $G = H$ and $S\cdot H$, respectively. The difference between $p_G(0\mathbf{z})$ and $p_G(1\mathbf{z})$ can be evaluated with a single circuit as follows

$$p_G(0\mathbf{z}) - p_G(1\mathbf{z}) = \langle G|0\rangle\langle 1|G\rangle \cdot \langle \psi|\mathbf{z}\rangle\langle \mathbf{z}'|\psi\rangle + \langle G|1\rangle\langle 0|G\rangle \cdot \langle \psi|\mathbf{z}'\rangle\langle \mathbf{z}|\psi\rangle \quad (7)$$

where $|G\rangle = G|0\rangle$. We note that the detailed derivation of eqs 6 and 7 is provided in Appendix A.

2.2.4. Grouping SB Operators. Equations 4 and 6 clearly indicate the expectation value of a TB Hamiltonian having M nonzero elements can be estimated with $O(2M)$ circuits. This number of quantum circuits, however, can be further reduced with a proper partition of the M SB operators that can be achieved by means of the GHZ measurement circuit. Let us consider a set of $(N + 1)$ -qubit circuits that consist of an N -qubit ansatz, a single-qubit G logic on the ancilla qubit, and a GHZ measurement, as illustrated in Figure 1(d). Then, as eq 6 indicates, all the possible outcomes $\mathbf{z}$ with probability $p(\mathbf{z})$ can be leveraged to get the expectation value of corresponding SB operators $|\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}|$, provided that the GHZ measurement is performed with a set of $\mathcal{X}$ that is determined by $\mathbf{x}$. In other words, a single GHZ measurement dedicated to a specific $\mathcal{X}$ can simultaneously measure SB operators $|\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}|$ for all $\mathbf{z}$, where $x_j = 1$ for $j \in \mathcal{X}$ and $x_j = 0$ otherwise. Consequently, the expectation value of a TB Hamiltonian can be efficiently estimated by grouping associated SB operators based on $\mathbf{x}$, which reduces the number of required circuits to the number of distinct $\mathbf{x}$'s. The detailed procedure for grouping SB operators is presented in Algorithm 1.

Algorithm 1: Grouping SB operators based on the GHZ measurement for estimation of $\langle \hat{O} \rangle$

```

input :  $M$  nonzero coefficients  $o_{\mathbf{z},\mathbf{z}'}$  and pairs of binary indices  $\mathbf{z}, \mathbf{z}'$  ( $\mathbf{z} \leq \mathbf{z}'$ )
output:  $\mathcal{G}_x^R$  and  $\mathcal{G}_x^I \forall \mathbf{x}$ 
forall nonzero  $o_{\mathbf{z},\mathbf{z}'}$  do
   $\mathbf{x} \leftarrow \mathbf{z} \oplus \mathbf{z}'$ 
   $\mathcal{X} \leftarrow \{j | x_j = 1 \text{ for } \mathbf{x}, 1 \leq j \leq N\}$ 
  if  $\text{Re}[o_{\mathbf{z},\mathbf{z}'}] \neq 0$  then
    if  $\mathcal{G}_x^R$  does not exist then  $\mathcal{G}_x^R = \emptyset$ ;
     $\mathcal{G}_x^R \leftarrow \mathcal{G}_x^R \cup \{\mathbf{z}\}$ 
  else if  $\text{Im}[o_{\mathbf{z},\mathbf{z}'}] \neq 0$  then
    if  $\mathcal{G}_x^I$  does not exist then  $\mathcal{G}_x^I = \emptyset$ ;
     $\mathcal{G}_x^I \leftarrow \mathcal{G}_x^I \cup \{\mathbf{z}\}$ 
  end
end
end
```

Two features of TB Hamiltonians can make the grouping procedure in Algorithm 1 be more efficient. First, due to their Hermiticity, all the Hermitian conjugate pairs $|\mathbf{z}\rangle\langle \mathbf{z}'|$ and $|\mathbf{z}'\rangle\langle \mathbf{z}|$ can be grouped, and the search space $\{\mathbf{z}, \mathbf{z}'\}$ in Algorithm 1 can be further narrowed down to the cases that satisfy the condition $\mathbf{z} \leq \mathbf{z}'$. Second, not all the elements in TB Hamiltonians have both real and imaginary parts. So, the number of circuits required to estimate the expectation value can be reduced by half for matrix elements ($o_{\mathbf{z},\mathbf{z}'}$) that are purely real or imaginary, as can be supported by eqs 4 and 6.

Our strategy for grouping SB operators with the GHZ measurement has nontrivial advantages over Pauli-based methods. First, SB grouping does not require optimization processes based on the commutativity of operators that are essential for Pauli grouping, and incurs only an $O(M)$ cost (M = the number of nonzero elements). Second, the measurement circuit for simultaneously estimating k commuting Pauli strings requires roughly at most $O(kN/\log k)$ CNOT gates.³⁴ The GHZ measurement circuit, however, can evaluate the expectation value of SB operators with a single H and at most N CNOT gates.

Finally, we note that Algorithm 1 is specialized for TB simulations upon the work reported by Kondo et al.³² Compared to the original approach, ours treats nonzero

elements with the condition $\mathbf{z} \leq \mathbf{z}'$ to handle Hermitian matrices with lower cost, and can additionally reduce the number of required circuits by up to half depending on the type of elements of TB Hamiltonians.

2.3. Optimal Number of Shots. Due to the probabilistic nature of quantum systems, the expectation value of any observables needs to be evaluated with a set of samples that are obtained by repeated measurements, so the error driven by finite sample sizes (sampling errors) inevitably affects the preciseness of any quantum algorithms. The sampling error has been analyzed in the context of VQE where the Hamiltonian is expressed as a linear combination of Pauli strings,^{44,45} and, in the same vain, here we discuss the sampling error involved in estimating the expectation of an observable $\hat{O}$ in eq 4 with our grouping method presented in Algorithm 1.

The sampling error ϵ stemming from the estimation of $\langle \hat{O} \rangle$ is the statistical error that arises due to the nonzero variance of samples, $\epsilon^2 \equiv \text{Var}(\langle \hat{O} \rangle)$. Since its N -qubit SB operators are grouped based on the bit string $\mathbf{x}$ and the type of the coefficient $o_{\mathbf{z},\mathbf{z}'}$, the observable $\hat{O}$ in eq 4 can be reorganized as follows

$$\hat{O} = \sum_{\mathbf{x}} \sum_{\mathbf{z}} \text{Re}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] \cdot |\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}| + \sum_{\mathbf{x} \neq 0} \sum_{\mathbf{z}} \text{Im}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] \cdot i|\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}| \quad (8)$$

Each term in eq 8 corresponds to a single GHZ measurement for each bit string $\mathbf{x}$ that is performed on the state $|\psi\rangle|G\rangle$ with $G = H$ and $S \cdot H$ for the real and imaginary part of the coefficient, respectively. By applying the formula for the sample variance ($\text{Var}(\langle \hat{A} \rangle) = \text{Var}(\hat{A})/n$ with n shots) to eq 8, the variance of the sample mean for $\langle \hat{O} \rangle$ can be expressed as follows

$$\epsilon^2 = \sum_{\mathbf{x}} \frac{\text{Var}(\sum_{\mathbf{z}} \text{Re}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] |\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}|)}{m_{R,\mathbf{x}}} + \sum_{\mathbf{x} \neq 0} \frac{\text{Var}(\sum_{\mathbf{z}} i \text{Im}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] |\mathbf{z}\rangle\langle \mathbf{z} \oplus \mathbf{x}|)}{m_{I,\mathbf{x}}} \quad (9)$$

where $m_{R(I),\mathbf{x}}$ denotes the number of shots obtained from the repeated GHZ measurements that are associated with the bit string $\mathbf{x}$ and the real(imaginary) part of $o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}$. With Lagrange multiplier, the total number of samples ($m = \sum_{\mathbf{x}} m_{R,\mathbf{x}} + \sum_{\mathbf{x} \neq 0} m_{I,\mathbf{x}}$) can be minimized with the constraint of eq 9. The upper bound of this minimum sample number is given as

$$\frac{1}{\epsilon^2} \left(\sum_{\mathbf{x}} \max_{\mathbf{z}} \left| \text{Re}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] \right| + \sum_{\mathbf{x} \neq 0} \max_{\mathbf{z}} \left| \text{Im}[o_{\mathbf{z},\mathbf{z} \oplus \mathbf{x}}] \right| \right)^2 \quad (10)$$

which can roughly tell us how many shots are required to estimate $\langle \hat{O} \rangle$ with error ϵ using our grouping method. We note that the detailed derivation of eq 10 is provided in Appendix B.

3. RESULTS AND DISCUSSION

Now, here we rigorously examine the feasibility of the VQE solver that incorporates the grouping strategy and measurement scheme presented in the Section 2.2, to find band gap energies of 3D-confined atomic structures that are described

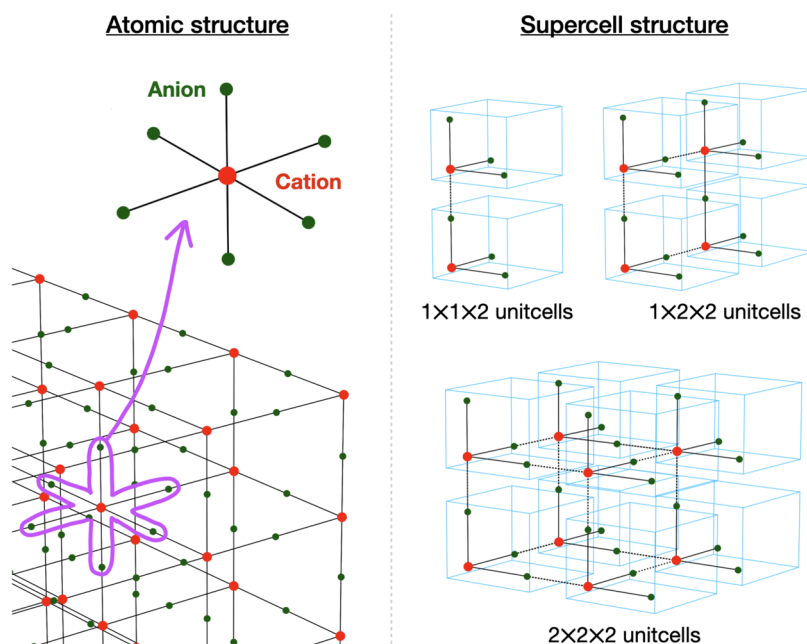


Figure 2. (Left) The atomic structure of MHP that is effectively described with a BX_3 crystalline, where each cation (B, red circle) is surrounded by six anions (X, green circles). (Right) Illustration of three different MHP supercells that are different in sizes, being made up of $1 \times 1 \times 2$, $1 \times 2 \times 2$, and $2 \times 2 \times 2$ unit cells. Note that each unit cell containing 4 atoms is outlined with a sky-blue box.

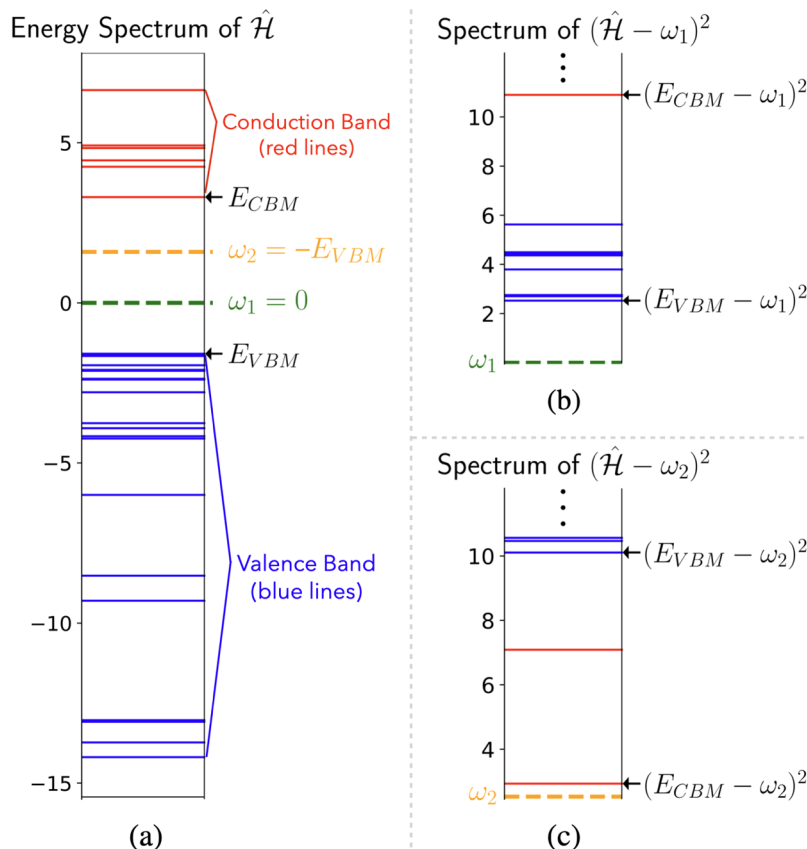


Figure 3. Illustration of the folded-spectrum method used in the cost function (eq 11) for a $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercell consisting of $1 \times 1 \times 2$ unit cells: (a) The energy spectrum of a sp^3 TB Hamiltonian $\hat{\mathcal{H}}$ and the corresponding folded spectrum of $(\hat{\mathcal{H}} - \omega I)^2$ with (b) $\omega = 0$ and (c) $\omega = -E_{\text{VBM}}$. Results show that E_{VBM} and E_{CBM} are located near zero, and E_{VBM} is closer to zero than E_{CBM} . This observation supports the validity of selecting reference values as $\omega = 0$ and $-\omega = E_{\text{VBM}}$.

with a TB model. For the case problem, we consider metal-halide-perovskite (MHP) supercells that are based on the

ABX_3 crystal structure and can flexibly generate visible lights of various colors by engineering the supercell size and the

composition of halogen atoms.^{19,21} In the particular case where A and B are Methylammonium (CH_3NH_3) and Lead (Pb), respectively, the molecule A is weakly coupled and the ABX_3 crystal can be effectively described with a BX_3 configuration as Figure 2 shows, enabling an empirical representation of the corresponding electronic structure with a sp^3 TB model.¹⁹ In consequence, $\text{CH}_3\text{NH}_3\text{PbX}_3$ supercells of finite sizes can be computationally constructed with repeated placements of the unit cell consisting of one cation (B(Pb)) and three anions (X) as illustrated in the right subfigures of Figure 2.

What we aim to find is the band gap energy of a MHP supercell, which can be obtained with the difference in energy between the minimum energy-level in conduction band and the maximum energy-level in valence band that are embedded in the middle of the dense energy spectrum of a TB Hamiltonian as shown in the left subfigure of Figure 3. Although numerous VQE studies have focused on finding excited energy states beyond the ground state of a given Hamiltonian, they mainly rely on the fact that eigenvectors of different states are orthogonal, to seek for states in higher energy with the information on states in lower energy that are already secured.^{8,9} Such strategies however would not be suitable in our case since the valence band of solid systems normally has many energy levels and we may need to compute too many redundant eigenvalues until the band gap energy is secured. We therefore strategically employ the so-called *folded-spectrum* method,⁶ targeting to find a specific energy level with a cost function defined as follows

$$C(\theta) = \langle \psi(\theta) | (\hat{\mathcal{H}} - \omega I)^2 | \psi(\theta) \rangle \quad (11)$$

where ω is the user-defined reference value around which the eigenvalue spectrum of a given Hamiltonian $\hat{\mathcal{H}}$ is folded. The cost function given in eq 11 is constructed against TB Hamiltonians with the SB grouping and GHZ measurement, and, though the folded-spectrum method requires the circuit decomposition of a squared Hamiltonian, the SB grouping still works in a cost-efficient manner as discussed in the Section 2.2. Through minimization of eq 11, we can find the square of an eigenvalue of $\hat{\mathcal{H}}$ that is closest to ω , and the selection of an appropriate ω for securing the band gap energy is straightforward since the two energy levels of our interest—the valence band maximum (VBM) and the conduction band minimum (CBM)—normally lie in the vicinity of zero energy, having a negative and a positive value, respectively. Additionally, the VBM is usually closer to zero than the CBM as the hole effective mass in solid systems is generally much larger than the electron effective mass. So, the band gap energy can be determined with the following two steps: we first set ω to zero to find the energetic position of VBM (E_{VBM}) by minimizing $C(\theta)$, and then determine the energetic position of CBM (E_{CBM}) by minimizing $C(\theta)$ with $\omega = -E_{\text{VBM}}$ as illustrated in the right subfigure of Figure 3.

To secure the ansatz circuit $V(\theta)$ that is realizable in NISQ devices with high customizability, which is essential to generate a variety of eigenvectors with a limited set of parameters θ , we exploit a quantum circuit consisting of two-qubit $\text{SU}(4)$ unitary blocks that are arranged in a brick-wall pattern^{46,47} as depicted in Figure 4. The number of layers (depth) of the ansatz circuit (see Figure 4 for the definition of a single layer), which also serves as a control factor of the expressivity of $V(\theta)$ for eigenvectors, is tuned from 4 to 8 depending on the perovskite supercell to be computed. The entire workflow of a

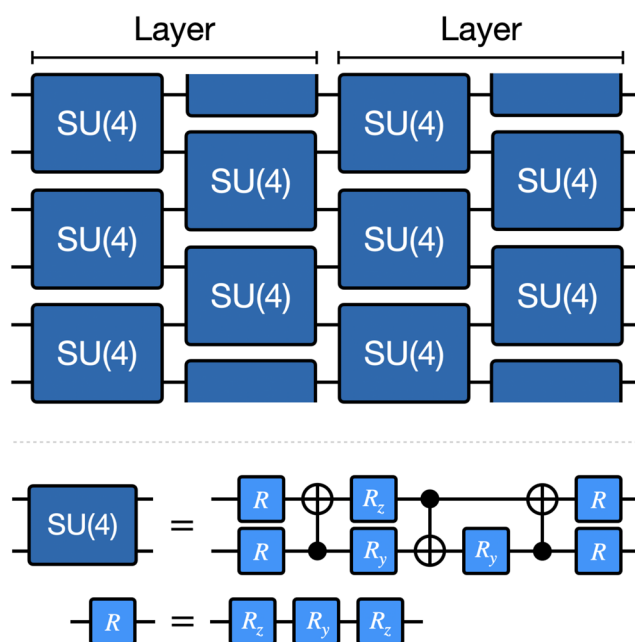


Figure 4. $\text{SU}(4)$ -based ansatz circuit in a brick-wall pattern with two layers that has been proposed by Anselmetti et al.⁴⁶ The structure of the two-qubit logic block $\text{SU}(4)$ is implemented based on the work of Shende et al.⁴⁷ The optimal number of layers, which is used for actual computation, is empirically determined for each supercell whose band gap energy is to be evaluated.

VQE calculation, including the minimization process of the cost function given in eq 11, is implemented with the PennyLane software development kit in conjunction with the ADAM optimizer and JAX library.⁴⁸

With the VQE scheme that has been described so far in this section and the Section 2.2, now we solve three finite $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercells (the halogen atom X is set to the Iodine(I)) that are different in sizes, being composed of $1 \times 1 \times 2$, $1 \times 2 \times 2$, and $2 \times 2 \times 2$ unit cells. Since we describe atomic structures with a sp^3 TB model, every atom in supercell domains is represented with 4 bases (s, p_x, p_y, p_z orbital) if no spin-orbit coupling is included (8 bases with spin-orbit coupling). Since a single unit cell of $\text{CH}_3\text{NH}_3\text{PbI}_3$ has four atoms as depicted in Figure 2, the corresponding TB Hamiltonian matrices become 64×64 , 128×128 , and 256×256 , and can be represented with 7 ($= 6 + 1$), 8 ($= 7 + 1$), and 9 ($= 8 + 1$) qubits, respectively, as our proposed method requires one extra qubit for implementation of the GHZ measurements.

Results of VQE calculations, which are summarized in Figure 5, demonstrates how the evaluated cost function for each supercell changes with respect to the iteration number of the optimization loop in an ideal condition where realistic effects such as gate errors, decoherence, and finite sampling errors are not considered. The top subfigures show the first iterative processes for securing E_{VBM} that are conducted with $\omega = 0$, while the bottom ones show the second processes conducted with $\omega = -E_{\text{VBM}}$ to get E_{CBM} . Starting with randomly determined variational parameters, all the calculations are conducted with the ansatz circuit that has 4 ($1 \times 1 \times 2$ unit cells), 7 ($1 \times 2 \times 2$ unit cells), and 8 ($2 \times 2 \times 2$ unit cells) layers, and the optimization process continues until the evaluated cost function fluctuates within 1.5×10^{-2} . We note that, for each supercell, the optimal number of ansatz layers is

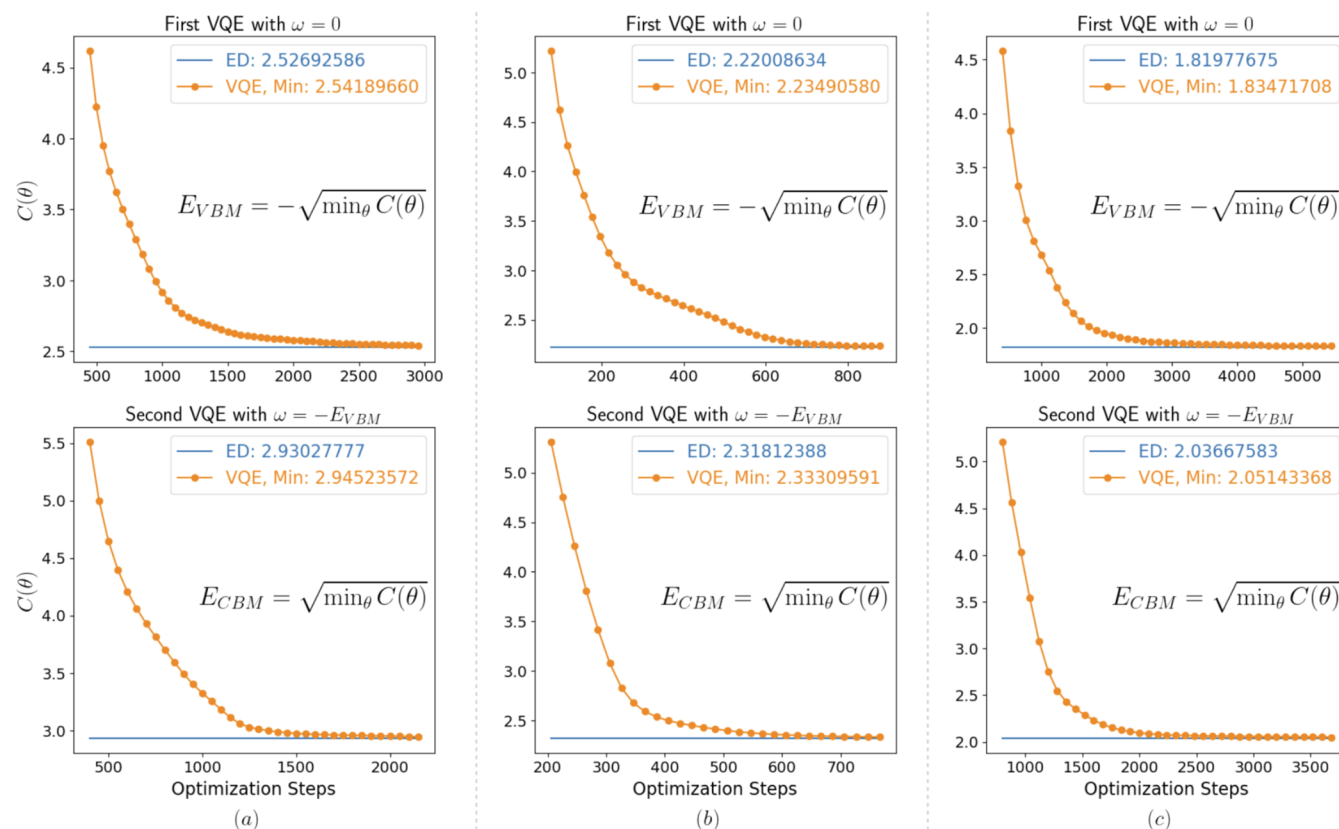


Figure 5. Results of VQE calculations that are conducted to find the band gap energy of a $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercell consisting of (a) $1 \times 1 \times 2$ unit cells, (b) $1 \times 2 \times 2$ unit cells, and (c) $2 \times 2 \times 2$ unit cells. For all the cases, the cost function given in eq 11 is minimized with the folded-spectrum method to obtain the minimum energy-level in conduction band (E_{CBM}) and the maximum energy-level in valence band (E_{VBM}). Optimization processes are conducted in an iterative manner until the cost function converges within 1.5×10^{-2} . Note that the exact value of the cost function, which is calculated with the exact diagonalization (ED) method, is plotted with a blue line.

empirically determined by conducting VQE simulations repeatedly with increasing the number of layers until the desired solution is secured. When compared to the reference value of the cost function that is obtained with the exact diagonalization (ED) method (marked with a blue line in every subfigure of Figure 5), the overall experimental results here clearly confirm that our VQE scheme works reasonably well, indicating the solid potential for empirical simulations of electronic structures based on a TB model.

To examine how the working performance of our VQE scheme is affected by sampling errors that arise in reality during the measurement process, we conduct additional calculations against the $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercell consisting of $1 \times 1 \times 2$ unit cells. We consider two error rates, $\epsilon_1 = 0.5$ and $\epsilon_2 = 0.1$ that drive the optimal shot number of $\sim 6.15 \times 10^5$ and $\sim 1.53 \times 10^7$, respectively, with eq 10. Since the cost function in this case can be evaluated with a total of 51 circuits, the optimal number of shots for each circuit, in principle, could be determined as discussed in the Section 2.3 and Appendix B. However, here we assume an equal number of measurement shots across all quantum circuits, and, under this assumption, the number of required shots per circuit becomes $\sim 1.3 \times 10^4$ and $\sim 3.01 \times 10^5$ for ϵ_1 and ϵ_2 , respectively. The early behavior of the optimization process in our VQE scheme is shown in Figure 6, where the value of the cost function is plotted with a 2ϵ confidence interval that is centered on the error-free value. As the optimization process evolves, the evaluated cost function fluctuates beyond the exact value since the sampling

error would affect gradients as well as updated parameters. Nevertheless, the cost function evaluated under sampling errors generally resides within the confidence interval, supporting the utility of eq 10 as a rule of thumb for determination of required shot numbers. Apparently, the number of shots cannot be increased blindly, and thus it must be determined appropriately at the sacrifice of the precision of solutions.

Though our VQE scheme cannot be free from the sampling issue as other quantum algorithms do, its strength—the cost efficiency driven with the SB grouping and the GHZ measurement—is quite obvious compared to VQEs employing other Pauli-based approaches. To examine the efficiency of our method for TB simulations rigorously, we construct the cost function given in eq 11 with various mapping strategies for Hamiltonians of $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercells whose dimensions range from $1 \times 1 \times 2$ (6 qubits) to $4 \times 4 \times 4$ unit cells (12 qubits). Results given in Figure 7(a) clearly show that, in terms of the number of quantum circuits required to construct the cost function, the performance of our approach surpasses that of the QWC Pauli grouping method and closely approaches that of the GC Pauli grouping method. We note that these results are well aligned to the underlying grouping conditions: the measurement circuit coupled to the GC Pauli grouping method has up to $O(kN/\log k)$ CNOT gates whereas the QWC Pauli grouping method does not require CNOT gates for measurements. The SB grouping method, which employs up to N CNOT gates for measurements, would be thus

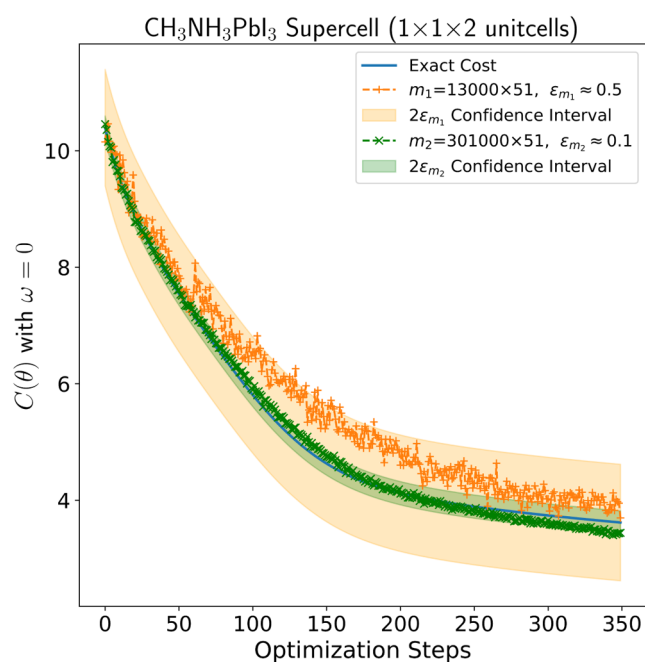


Figure 6. Early stage of the optimization process for VQEs in the presence of finite sampling errors. The cost function targets the lowest eigenvalue of the folded spectrum of the TB Hamiltonian with $\omega = 0$. Numerical simulations are conducted with two cases of sampling m_1 (orange dashed line with circles) and m_2 (green dashed line with crosses), and each shaded area represents the corresponding 2ϵ confidence interval. The blue line corresponds to the value of the cost function obtained with no sampling errors.

positioned between the two Pauli-based methods. In spite of the similarity in the required number of circuits, the advantage of the SB grouping over the GC Pauli grouping is still clear in terms of the computing speed as Pauli-based grouping methods must involve an optimization problem that is NP-hard,^{40,41} whereas the SB grouping method just requires up to $O(M)$ runtimes as outlined in Algorithm 1. Figure 7(b), which shows the time required to complete mapping Hamiltonians into quantum circuits in an Apple Mac Studio (M2Max processor and 64GB memory), clearly supports the computational advantage of our method over the GC and the QWC Pauli grouping. Though heuristic approaches are often exploited for GC Pauli grouping, they in principle cannot guarantee the quality of solutions particularly as the problem size grows. Moreover, these results clearly underscore the advantage of the proposed method for estimation of the cost function under finite sampling error ϵ . As described in Appendix B, the sampling variance of the cost function (ϵ^2) becomes S^2/m^* , where S is the sum of standard deviations across grouped observables and m^* denotes the total number of shots. The reduced number of quantum circuits, combined with shorter runtime, ultimately contributes to a smaller S , thereby diminishing the number of shots required to secure a given precision. As Figure 7(c) shows, the SB grouping intriguingly yields the lowest S in most cases for a fixed random ansatz state, indicating the superior sampling efficiency that our method has compared to the Pauli-based ones. In consequence, our method involving the SB grouping offer a more practical and scalable route for handling TB Hamiltonians, and arbitrary sparse matrices in a more broad sense.

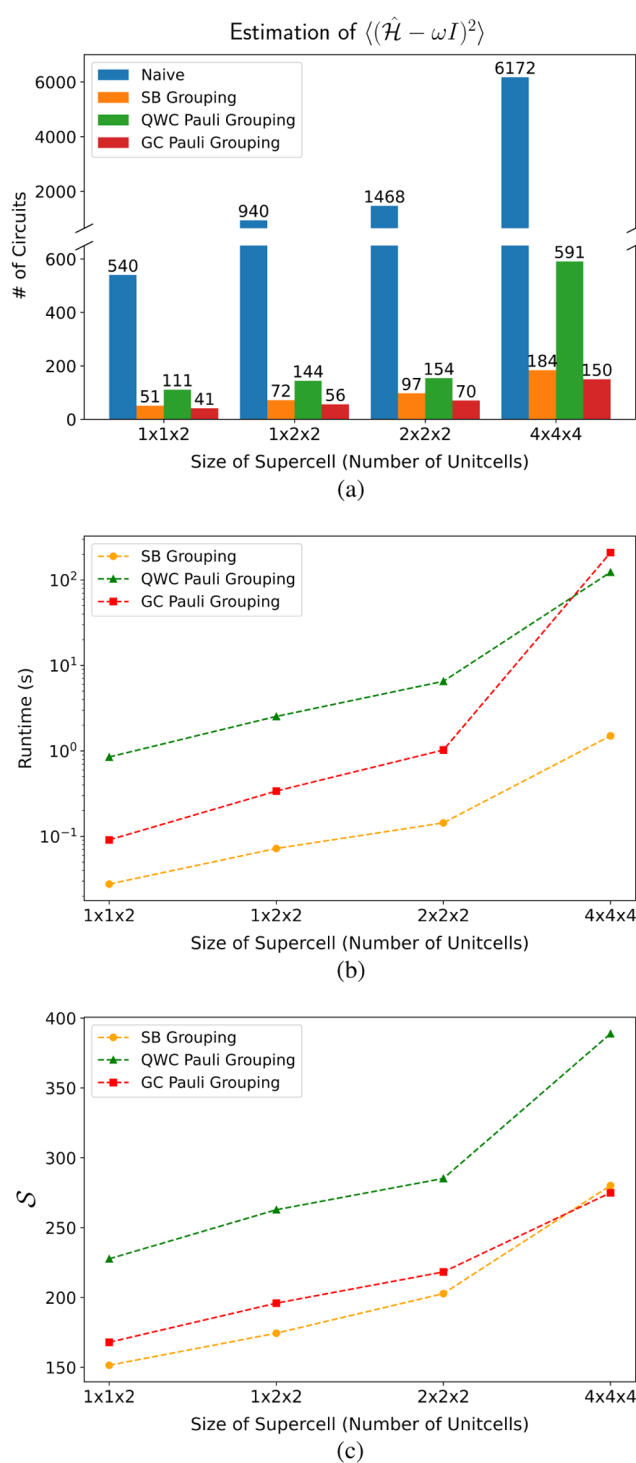


Figure 7. Superiority of the SB grouping to the QWC and GC Pauli grouping for handling the cost function given in eq 11. (a) The number of required circuits is shown as a function of supercell sizes, where the “Naive” case indicates the results secured with the raw Pauli decomposition. (b) The time required to complete the mapping process of TB Hamiltonians clearly supports the superiority of our approach based on the SB grouping to the Pauli-based grouping methods that require NP-hard optimization processes. (c) S (the sum of standard deviations across grouped observables) is plotted as a function of supercell size for a fixed ansatz state against the three grouping strategies. The graph shows that S evaluated with SB grouping is generally smaller than those obtained with Pauli-based methods, indicating that our method either achieves a smaller error

Figure 7. continued

with the same shot number or requires a less number of shots to drive the same error.

4. CONCLUSIONS

We have presented a variant of VQE tailored for finding the band gap energy of the confined atomic system whose electronic structure is described with the empirical tight-binding (TB) model. Employing a set of standard-basis (SB) operators to build the sparse Hamiltonian directly from the lattice geometry of the target atomic system, the proposed VQE scheme evaluates the expectation value of the TB Hamiltonian by grouping SB operators in conjunction with the so-called Greenberger–Horne–Zeilinger (GHZ) measurement circuit that is an extension of the Bell measurement circuit. The superiority of our method to Pauli-based approaches in terms of computational efficiency is quite obvious particularly for TB simulations, because the SB-based expression of the Hamiltonian can be done with $O(M)$ (M = the number of nonzero elements in the Hamiltonian) while the cost becomes $O(4^N)$ (N = the qubit size of circuits) with the Pauli decomposition. Our grouping strategy coupled to the

GHZ measurement evaluates the expectation value of the Hamiltonian with fewer circuits than the case of the Pauli grouping based on the qubit-wise commutativity, and also outperforms the Pauli grouping based on the general commutativity as it involves no NP-hard optimization processes that are essential in Pauli-based methods. Results of experiments that have been conducted to determine band gap energies of $\text{CH}_3\text{NH}_3\text{PbI}_3$ supercells, clearly support the computational benefit of our scheme, reinforcing the potential of VQE for empirical simulations of electronic structures that are described with sparse Hermitian system matrices.

■ A DERIVATION OF EQUATION 7

Let us consider an N -qubit quantum state $|\psi\rangle$. In order to directly estimate the expectation value of N -qubit SB operators in the state $|\psi\rangle$, we prepare an ancillary qubit in the state $|G\rangle = |G0\rangle$ with $G \in \{H, S\cdot H\}$ and apply a single GHZ measurement to the composite $(N + 1)$ -qubit state $|G\rangle|\psi\rangle$. The GHZ measurement is expressed as $U_{\text{GHZ}}^\dagger |z_0 z\rangle \langle z_0 z| U_{\text{GHZ}}$, where the unitary U_{GHZ} is defined as eq 5 and $z_0 z$ is $z_0 z_1 z_2 \cdots z_N$. The probability difference $p(0z)$ and $p(1z)$ from repeated GHZ measurements yields the expectation value of a Hermitian conjugate pair of SB operators as follows

$$p_G(0z_1 \cdots z_N) - p_G(1z_1 \cdots z_N) = \langle G | \langle \psi | U_{\text{GHZ}}^\dagger (\hat{Z}_0 \otimes |z\rangle \langle z|) U_{\text{GHZ}} | G \rangle | \psi \rangle = \langle G | \langle \psi | \left(\prod_{j \in \mathcal{X}} CX_{0,j} \right) (\hat{X}_0 \otimes |z\rangle \langle z|) \left(\prod_{j \in \mathcal{X}} CX_{0,j} \right) | G \rangle | \psi \rangle \quad (12)$$

$$= \langle G | \langle \psi | \left(|0\rangle \langle 1| \otimes |z\rangle \langle z| \prod_{j \in \mathcal{X}} \hat{X}_j \right) + h.c. \rangle | G \rangle | \psi \rangle \quad (13)$$

$$= \langle G | \langle \psi | (|0\rangle \langle 1| \otimes |z\rangle \langle z| \oplus \mathbf{x} | + h.c.) | G \rangle | \psi \rangle \quad (14)$$

$$= \langle G | 0 \rangle \langle 1 | G \rangle \cdot \langle \psi | z \rangle \langle z \oplus \mathbf{x} | \psi \rangle + \langle G | 1 \rangle \langle 0 | G \rangle \cdot \langle \psi | z \oplus \mathbf{x} \rangle \langle z | \psi \rangle \quad (15)$$

$$= \begin{cases} \frac{1}{2} (\langle \psi | z \rangle \langle z \oplus \mathbf{x} | \psi \rangle + \langle \psi | z \oplus \mathbf{x} \rangle \langle z | \psi \rangle) & \text{if } G = H \\ \frac{i}{2} (\langle \psi | z \rangle \langle z \oplus \mathbf{x} | \psi \rangle - \langle \psi | z \oplus \mathbf{x} \rangle \langle z | \psi \rangle) & \text{if } G = S \cdot H \end{cases} \quad (16)$$

Here, $\mathbf{x}$ is the N -bit string corresponding to the set $\mathcal{X}$, where $x_j = 1$ if $j \in \mathcal{X}$ and $x_j = 0$ otherwise. Thus, the configuration of the CNOT gates in U_{GHZ} determines the bit string $\mathbf{x}$. Conversely, if the expectation value of a specific SB operator $|z\rangle \langle z'|$ is to be estimated, we calculate $\mathbf{x} = \mathbf{z} \oplus \mathbf{z}'$ and define the set $\mathcal{X} = \{j | x_j = 1, \text{ for } 1 \leq j \leq N\}$.

■ B DERIVATION OF EQUATION 10

Given an observable expressed in eq 8 and applying the sample variance formula, the variance of its expectation value $\text{Var}(\langle \hat{O} \rangle)$ can be written as eq 9. For each bit string $\mathbf{x}$ and the corresponding $o_{z, z \oplus \mathbf{x}}$, the GHZ measurement is performed on the composite state $|G\rangle|\psi\rangle$ with $m_{R(I), \mathbf{x}}$ shots, where the measurement configuration, specifically the set $\mathcal{X}$ in U_{GHZ} and the gate $G \in \{H, S\cdot H\}$, is determined by $\mathbf{x}$ and whether the coefficient is real or imaginary, respectively. The total number of shots in estimation of $\langle \hat{O} \rangle$ is then given by

$$m = \sum_{\mathbf{x}} m_{R, \mathbf{x}} + \sum_{\mathbf{x} \neq 0} m_{I, \mathbf{x}} \quad (17)$$

To minimize m subject to the constraint eq 9, we utilize the method of Lagrange multipliers. The Lagrangian $\mathcal{L}$ is constructed as

$$\mathcal{L} = m + \lambda \left(\sum_{\mathbf{x}} \frac{\sigma_{R, \mathbf{x}}^2}{m_{R, \mathbf{x}}} + \sum_{\mathbf{x} \neq 0} \frac{\sigma_{I, \mathbf{x}}^2}{m_{I, \mathbf{x}}} - \epsilon^2 \right) \quad (18)$$

where $\sigma_{R, \mathbf{x}}^2 = \text{Var}(\sum_z \text{Re}[o_{z, z \oplus \mathbf{x}}] |z\rangle \langle z \oplus \mathbf{x}|)$ and $\sigma_{I, \mathbf{x}}^2 = \text{Var}(\sum_z i \text{Im}[o_{z, z \oplus \mathbf{x}}] |z\rangle \langle z \oplus \mathbf{x}|)$. Taking the partial derivative of $\mathcal{L}$ with respect to m_i gives

$$\frac{\partial \mathcal{L}}{\partial m_i} = 1 - \lambda \frac{\sigma_i^2}{m_i^2} = 0 \quad (19)$$

which yields the optimal solution $m_i^* = \sqrt{\lambda} \sigma_i$ for each $i \equiv (R(I), \mathbf{x})$. By substituting this into eq 17 and eq 9, the minimal total number of shots, denoted as m^* , can be derived as follow

$$m^* = \frac{1}{\epsilon^2} \left(\sum_{\mathbf{x}} \sigma_{R,\mathbf{x}} + \sum_{\mathbf{x} \neq 0} \sigma_{I,\mathbf{x}} \right)^2 \quad (20)$$

If the variances σ_i^2 can be evaluated for all i , the number of shots required to estimate $\langle \hat{O} \rangle$ within a desired sampling error ϵ can be directly determined. Alternatively, we approximate them by deriving upper bounds, which allows for practical shot allocations without requiring prior knowledge of the quantum state. The upper bound on the variance is derived as follows

$$\begin{aligned} \sigma_i^2 &= \text{Var} \left(\sum_{\mathbf{z}} c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}} |\mathbf{z}\rangle \langle \mathbf{z} \oplus \mathbf{x}| \right) \leq \left\langle \left(\sum_{\mathbf{z}} |c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}|^2 |\mathbf{z}\rangle \langle \mathbf{z}| \right) \right\rangle \\ &= \sum_{\mathbf{z}} |c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}|^2 p(\mathbf{z}) \leq \max_{\mathbf{z}} |c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}|^2 \end{aligned} \quad (21)$$

where $c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}$ denotes either $\text{Re}[o_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}]$ or $\text{Im}[o_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}]$. This upper bound leads directly to the bound on the minimal total number of shots m^* stated in eq 10. Furthermore, eq 21, combined with the optimal shot allocation strategy, can provide an upper bound on the minimal number of shots for any circuit i , which can be written as follow,

$$\begin{aligned} m_i^* &\leq \frac{\max_{\mathbf{z}} |c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}|}{\epsilon^2} \left(\sum_{\mathbf{x}} \max_{\mathbf{z}} |\text{Re}[o_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}]| \right. \\ &\quad \left. + \sum_{\mathbf{x} \neq 0} \max_{\mathbf{z}} |\text{Im}[o_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}}]| \right) \end{aligned} \quad (22)$$

■ C SAMPLING VARIANCE ANALYSIS OF $\langle \hat{O} \rangle$ UNDER DIFFERENT OPERATOR BASES AND GROUPING STRATEGIES

To evaluate the effectiveness of the SB grouping method in terms of the minimum number of shots, we compare its sampling efficiency against two Pauli-based grouping strategies. As derived in Appendix B, the minimal total number of measurement shots can also be determined within the Pauli frameworks. An arbitrary observable $\hat{O}$ can be expressed in the Pauli basis as

$$\hat{O} = \sum_{i=1}^{N_g} \hat{P}_i = \sum_{i=1}^{N_g} \sum_{j=1}^{N_i} a_{ij} \hat{P}_{ij} \quad (23)$$

where $\hat{P}_{ij}$ is the j th Pauli string in the i th group, $a_{ij} \in \mathbb{R}$ is its coefficient, N_g is the number of groups of mutually commuting Pauli strings and N_i is the number of Pauli strings in the i th group. Each grouped observable $\hat{P}_i$ is a linear combination of mutually commuting Pauli strings. By applying the standard formula for sampling variance and using the method of Lagrange multipliers, the minimal total number of shots m_p^* needed to estimate $\langle \hat{O} \rangle$ with sampling error ϵ is given by

$$m_p^* = \frac{1}{\epsilon^2} \left(\sum_{i=1}^{N_g} \sqrt{\text{Var}(\hat{P}_i)} \right)^2 \quad (24)$$

where $\text{Var}(\hat{O}) = \langle \hat{O}^2 \rangle - \langle \hat{O} \rangle^2$ is the variance of the observable. Detail derivations of this expression can be found in refs 34,44,45

Consequently, following the form of eqs 20 and 24, the sampling variance for a given observable $\hat{O}$ can, in general, be expressed as $\epsilon^2 = S^2/m^*$, where S is defined as

$$S \equiv \sum_i \sqrt{\text{Var}(\hat{O}_i)} \quad (25)$$

with $\hat{O}_i$ denoting the grouped operators under any operator basis and any grouping strategy. When the Pauli decomposition and Pauli grouping methods are applied, $S = \sum_i \sqrt{\text{Var}(\hat{P}_i)}$. In contrast, for the SB decomposition and SB grouping method, $S = \sum_{\mathbf{x}} \sqrt{\text{Var}(\hat{O}_{\mathbf{x}})}$, where each SB-grouped operator is defined as $\hat{O}_{\mathbf{x}} \equiv \sum_{\mathbf{z}} c_{\mathbf{z}, \mathbf{z} \oplus \mathbf{x}} |\mathbf{z}\rangle \langle \mathbf{z} \oplus \mathbf{x}|$. Thus, S serves as a significant indicator for assessing which operator basis is more advantageous in reducing the total number of measurement shots or the sampling error when estimating the cost function.

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Notes

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