

Boson Sampling Enhanced Quantum Chemistry

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In this work, we give a hybrid quantum-classical algorithm for solving electronic structure problems of molecules using only linear quantum optical systems. The variational ansatz we proposed is a hybrid of noninteracting boson dynamics and classical computational chemistry methods, specifically, the Hartree-Fock method and the configuration interaction method. The boson part is built by a linear optical interferometer, which is easier to realize compared with the well-known unitary coupled cluster (UCC) ansatz composed of quantum gates in conventional variational quantum eigensolver, and the classical part is merely classical processing acting on the Hamiltonian. The appearance of permanents in the boson part has its physical intuition to provide different kinds of resources from commonly used single, double, and higher excitations in classical methods and the UCC ansatz to explore chemical quantum states. Such resources can help enhance the accuracy of methods used in the classical parts. We give a scalable hybrid homodyne and photon-number measurement procedure for evaluating the energy value, which has intrinsic abilities to mitigate photon loss errors, and discuss the extra measurement cost induced by the no Pauli exclusion principle for bosons with its solutions. To demonstrate our proposal, we run numerical experiments on several molecules and obtain their potential energy curves reaching chemical accuracy.

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I. INTRODUCTION

Linear quantum optical systems (LQOSs) [1] have a distinct advantage: the photons are exceptionally clean and robust against noise, especially when compared to other architectures [2–4]. However, this advantage of cleanness comes at a cost: LQOS lacks natural interactions between photons, which are essential for achieving universal quantum computing. To address this limitation, researchers have explored methods to introduce nonlinearity based

on feedforward measurements [5,6]. Unfortunately, implementing these techniques in practice remains challenging.

It is well established that passive LQOSs, which lack feedforward measurements, cannot serve as universal quantum computers. However, can they exhibit advantages in specific tasks that are classically hard? Aaronson and Arkhipov [7] gave a positive answer. Specifically, they demonstrated that sampling the outputs of a linear optical interferometer (LOI) with Fock state inputs—known as the boson sampling (BS) problem—is unlikely to be efficiently simulated by classical computers. The BS problem has become a promising platform for showcasing early-stage quantum advantages due to its experimental feasibility. Zhong *et al.* [8] recently achieved a milestone in this direction by using Gaussian boson sampling, a variant of BS. Subsequent works [9–11] further improved upon this demonstration of quantum advantage. However, it remains true that passive LQOS is nonuniversal. The next significant challenge is to identify specific problems of practical interest that passive LQOS can solve and where

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they hold potential advantages. While proposals exist for applications such as molecular docking [12], molecular vibrational spectra [13], and graph theory [14], further efforts are needed to explore additional use cases.

When we look at other platforms in the NISQ era [15], solving the electronic structure problem (ESP) [16] using the variational quantum eigensolver (VQE) [17,18] is one of the most anticipated applications. For ESP, we aim to solve the ground-state energy of the corresponding chemical Hamiltonian to help predict the chemical reaction rates. This can be done by VQE where a classical optimizer is used to optimize the energy expectation measured from the output of a shallow quantum circuit ansatz. The workflow of VQE has some degree of resilience to the noise, which makes solving ESP using NISQ computers possible. Since solving ESP exactly requires exponential resources [full configuration interaction (FCI)] using classical computers, quantum computing may manifest potential advantages. However, VQE is not friendly to passive LQOS since there is no natural correspondence from multimode boson Fock space to multi-qubit space, and the quantum circuit language cannot be used to describe general operations of linear interferometers.

In this work, we give a passive LQOS-based VQE algorithm for solving ESP. The circuit ansatz in this algorithm is a hybrid of noninteracting boson dynamics and classical computational chemistry methods. In the following, we will introduce this algorithm from its physical intuition to its workflow and implementation. Specifically, we will talk about how to measure the cost function efficiently by a combination of homodyne and photon-number detections. We will also present several numerical results to show the properties and the performance of this algorithm.

II. BACKGROUND

In quantum chemistry [16], we are interested in the electronic structures of molecules. Under the Born–Oppenheimer approximation, treating the nuclei as classical point charges, the Hamiltonian of a molecule only has the kinetic energy terms of the electrons, the electrons’ Coulomb interaction with the nuclei, and the electron–electron Coulomb repulsion left. For ESP, we aim to find the ground-state energy of this electronic structure Hamiltonian. However, this Hamiltonian is currently in the first quantization picture, and we have to take care of the Pauli exclusion principle explicitly. To simplify the situation, we typically take the Hamiltonian to the second quantization picture where electrons can be created or annihilated from a set of orbitals (e.g., the STO-3G basis set used in our numerical examples),

$$H^F = \sum_{pq} h_{pq} f_p^\dagger f_q + \frac{1}{2} \sum_{pqrs} h_{pqrs} f_p^\dagger f_q^\dagger f_r f_s, \quad (1)$$

where $f_p^\dagger$ and f_p are the fermion creation and annihilation operations respectively and the h_{pq} and the h_{pqrs} correspond to the one- and two-electron integrals. The form of the Hamiltonian H^F naturally preserves the number of electrons. If we consider a basis set containing M orbitals and the molecule contains N electrons, then solving ESP corresponds to finding the smallest eigenenergy in the electron subspace of dimension $\binom{M}{N}$. Note that, to be more accurate, we can also consider the spin symmetry to further reduce the subspace dimension. However, in the following discussions, we will just relax this restriction.

The Hartree-Fock (HF) [16] method is a well-known method to give an approximate solution to the problem. The HF method can be understood as a VQE procedure using the noninteracting fermion dynamics $U_F(\vec{\beta}) = \exp(-i \sum_{pq} \beta_{pq} f_p^\dagger f_q)$ as an ansatz to minimize the energy: $|\text{HF}\rangle = U_F(\vec{\beta}_{\text{hf}})|\phi_0\rangle$, $\vec{\beta}_{\text{hf}} = \text{argmin}_{\vec{\beta}} \langle \phi_0 | U_F^\dagger(\vec{\beta}) H^F U_F(\vec{\beta}) | \phi_0 \rangle$, where $|\phi_0\rangle$ is an initial product state with N electrons. U_F belongs to the fermion Gaussian operations that are classically tractable due to their equivalence with the match gate circuits [19]. Since U_F only works as a basis rotation of orbitals and contains no electron–electron interactions, the HF method can only give a single-determinant solution, which is insufficient to accurately characterize the ground state, especially in the dissociation area.

To get more accurate results, one typically uses $|\text{HF}\rangle$ as the reference state $|\Phi_{\text{ref}}\rangle$ for post-HF methods such as configuration interaction (CI) [16] and coupled cluster (CC) [16] methods to give multideterminant solutions [20]. Specifically, CI wave function is $(1 + T)|\Phi_{\text{ref}}\rangle$ and CC wave function is $e^T|\Phi_{\text{ref}}\rangle$ with $T = T_1 + T_2 + \dots$ contains single, double, and higher order excitations. When only considering $T = T_1 + T_2 = \sum_{pq} \beta_{pq} f_p^\dagger f_q + \sum_{pqrs} \beta_{pqrs} f_p^\dagger f_q^\dagger f_r f_s$, we have the CI singles and doubles (CISD) and CC singles and doubles (CCSD). For quantum computing, the famous unitary coupled cluster (UCC) ansatz [21,22] used for VQE is the unitary version of CCSD whose dynamics can be expressed as $e^{T-T^\dagger}$. To implement commonly used UCC singles and doubles (UCCSD) on quantum computers, we usually use the Trotter decomposition to approximate the full dynamics in terms of local unitary operations, which gives rise to deep circuits intractable in NISQ devices. The idea of these methods is to introduce additional resources of interacting fermion terms to explore states beyond a single determinant in the electron subspace. As the expressivity power of the ansatz increases by including more degrees of freedom, a higher accuracy may be achieved.

Let us briefly introduce BS and make a comparison with fermion’s version. The general operations of a multimode LOI are noninteracting boson dynamics that can be expressed as $U_B(\vec{\alpha}) = \exp(-i \sum_{pq} \alpha_{pq} b_p^\dagger b_q)$ where $b_p^\dagger$

and b_p are the boson creation and annihilation operations, respectively. The (anti-)commutation relations of $b^\dagger$ and b are different from $f^\dagger$ and f , which reflects the different behaviors between bosons and fermions. This also leads to the difference in the complexity between the BS and the fermion sampling (FS) [23]. More specifically, the transition amplitude from an initial product state $|S\rangle = |s_1 s_2 \cdots s_M\rangle$ to an output product state $|T\rangle = |t_1 t_2 \cdots t_M\rangle$ (s_p and t_p are natural numbers for bosons and 0, 1 for fermions) under the noninteracting boson and the fermion dynamics, respectively, can be expressed as

$$\begin{aligned} \langle T|U_B(\vec{\alpha})|S\rangle &= \frac{\text{Per}(U_{B,ST}(\vec{\alpha}))}{\sqrt{\Pi_p s_p! \Pi_p t_p!}}, \\ \langle T|U_F(\vec{\beta})|S\rangle &= \text{Det}(U_{F,ST}(\vec{\beta})), \end{aligned} \quad (2)$$

where Per and Det denote the matrix permanent and the matrix determinant and $U_{B,ST}(\vec{\alpha})$ ($U_{F,ST}(\vec{\beta})$) is generated from the mode transformation matrix $e^{i\alpha} (e^{i\beta})$ corresponding to $U_B(\vec{\alpha})$ ($U_F(\vec{\beta})$) [23]. Since the computation of permanent is **#P-hard** and computation of determinant is in **P** complexity, we believe BS is classically intractable but FS is classically tractable [7]. This difference is the key inspiration for our proposal.

III. ALGORITHM

A. Ansatz

To enable passive LQOS for solving ESP, we use the single-rail encoding [1], where 0, 1 photon states $|0\rangle_B$ and $|1\rangle_B$ represent 0, 1 electron states $|0\rangle_F$ and $|1\rangle_F$. When we use the Jordan-Wigner (JW) mapping [16] to transform fermion Hamiltonians to qubit Hamiltonians, this encoding also means $|0\rangle_B$ and $|1\rangle_B$ represent qubit 0, 1 states $|0\rangle_Q$ and $|1\rangle_Q$. The conservation of photon number in passive LQOS naturally corresponds to the conservation of electron number. As mentioned above, to go beyond a single determinant and create superpositions in the electron subspace, it is important to have resources different from the HF method based on noninteracting fermion dynamics.

The comparison between BS and FS tells us that permanents in noninteracting boson dynamics under the single-rail encoding exactly provide such a resource and are different from the higher-order excitations in CI, CC, and UCC. Also, this resource is classically intractable. This observation helps us create the following ansatz, which we named boson sampling-classic (BS-C) for VQE,

$$|\Phi_{\text{BS-C}}\rangle = V_C(\vec{\beta})|\Phi(\vec{\alpha})\rangle = V_C(\vec{\beta})U_B(\vec{\alpha})|\Phi_{\text{ref}}\rangle. \quad (3)$$

The $U_B(\vec{\alpha})$ part is the noninteracting boson dynamics that serves as the additional resources to enhance the performances of classical operations $V_C(\vec{\beta})$, such as $V_{\text{HF}}(\vec{\beta}) = U_B(\vec{\beta})$ in HF that leads to BS-HF and $V_{\text{CISD}}(\vec{\beta}) = 1 + T_1 + T_2$ in CISD that leads to BS-CISD.

The realization of BS-C and the implementation of the measurement of energy require a detailed discussion. The initial single-determinant reference state $|\Phi_{\text{ref}}\rangle$ directly corresponds to an initial photon state. The occupied orbitals correspond to optical modes with single-photon inputs, and the virtual orbitals correspond to modes with vacuum inputs. $U_B(\vec{\alpha})$ is a physical part realized by a parameterized LOI whose universal realization can be found in Ref. [24]. $V_C(\vec{\beta})$ is a virtual part that merely corresponds to classical processing acting on the Hamiltonian H^F . Similar ideas have recently been investigated in Refs. [25,26]. Specifically, we need to calculate the transformed Hamiltonian $H_T^F(\vec{\beta}) = V_C^\dagger(\vec{\beta})H^F V_C(\vec{\beta})$ which is used to replace H^F for JW transformation to get a qubit Hamiltonian $H_T^{\text{JW}}(\vec{\beta})$ for measurements. [In current encoding, $H_T^{\text{JW}}(\vec{\beta})$ has the same matrix elements as $H_T^F(\vec{\beta})$. The reason we add JW mapping is that the Pauli expression of $H_T^{\text{JW}}(\vec{\beta})$ can be easier to formulate the measurement procedure.]

Calculating $H_T^F(\vec{\beta})$ is efficient for proper $V_C(\vec{\beta})$ like HF and CISD since H^F in Eq. (1) is a local fermion Hamiltonian and $V_{\text{HF}}(\vec{\beta})$ is merely a basis rotation for HF [19] and $V_{\text{CISD}}(\vec{\beta})$ is also a local operator for CISD. The resulting $H_T^F(\vec{\beta})$ is also local, which means there is only a polynomial number of fermion terms in $H_T^F(\vec{\beta})$ and also a polynomial number of Pauli terms in $H_T^{\text{JW}}(\vec{\beta})$. The polynomial number of terms in $H_T^{\text{JW}}(\vec{\beta})$ is crucial for the measurement procedure to be scalable, which will be discussed later. Note that other classical blocks can also be used as the classical part, as long as they satisfy the conditions on efficient measurements shown later.

B. Cost function and hybrid measurement

For the cost function, we should realize that the value $\langle \Phi(\vec{\alpha})|H_T^{\text{JW}}(\vec{\beta})|\Phi(\vec{\alpha})\rangle$ does not correspond to the true expectation value of energy, and two corrections are needed. One direct correction is inherited from $V_C(\vec{\beta})$. When $V_C(\vec{\beta})$ is non-unitary, the norm of $|\Phi_{\text{BS-C}}\rangle$ will be changed. The other correction is due to the fact that $|\Phi(\vec{\alpha})\rangle$ may have populations outside the encoding space since there is no Pauli exclusion principle for photons, which will also influence the length of $|\Phi_{\text{BS-C}}\rangle$. Thus, the true energy value should be

$$E(\vec{\alpha}, \vec{\beta}) = \frac{\langle \Phi(\vec{\alpha})|H_T^{\text{JW}}(\vec{\beta})|\Phi(\vec{\alpha})\rangle}{\langle \Phi(\vec{\alpha})|V_C^\dagger(\vec{\beta})V_C(\vec{\beta})|\Phi(\vec{\alpha})\rangle}. \quad (4)$$

We should remember that both $H_T^{\text{JW}}(\vec{\beta})$ and $V_C(\vec{\beta})$ are defined in the electron subspace, which has zero matrix element values outside this subspace, where the Pauli exclusion principle is unsatisfied. For BS-HF, $V_{\text{HF}}(\vec{\beta})$ is unitary in this subspace, as a result, $V_{\text{HF}}^\dagger(\vec{\beta})V_{\text{HF}}(\vec{\beta}) = Q$ becomes the projection operator from the whole photon Hilbert state to electron subspace with $E(\vec{\alpha}, \vec{\beta}) =$

$\langle \Phi(\vec{\alpha}) | H_T^{\text{JW}}(\vec{\beta}) | \Phi(\vec{\alpha}) \rangle / \langle \Phi(\vec{\alpha}) | Q | \Phi(\vec{\alpha}) \rangle$. We name the value of $\langle \Phi(\vec{\alpha}) | Q | \Phi(\vec{\alpha}) \rangle$ the projection ratio. When considering the whole measurement cost of $E(\vec{\alpha}, \vec{\beta})$, the value of the projection ratio is crucial, which we will discuss later.

Having the definition of the cost function in Eq. (4), the question is how to measure it. According to the transformation rule of the JW mapping, the terms we need to measure will only be composed of I , Z , σ_+ , and σ_- . Under the single-rail encoding, different quantum states correspond to different photon numbers; thus, conventional photon-number basis measurement in dual encoding is not universal [1] for measuring σ_- and σ_+ . On the other hand, the homodyne measurement [27] (also see Appendix A) based on continuous variables in phase space which can be done experimentally by mixing the target quantum mode with a local oscillator by a balanced beam splitter and measuring two output modes using two photodetectors with postprocessing [28] can only be efficient for evaluating local terms which is not compatible with the JW mapping due to the existence of Z . Here, a term is local means it has Z , σ_+ , and σ_- acting on a limited number of qubits. To resolve this obstacle, we give a hybrid measurement strategy where for qubits with I and Z , we use the photon-number measurements, and for qubits with σ_+ and σ_- , we use the homodyne measurements. We want to mention that the introduction of Gaussian measurements will not affect the hardness of BS [29].

To illustrate the workflow, we can consider a term with the form $H_i = Z_1 Z_2 \sigma_{+,3} \sigma_{-,4}$, the value $\text{Tr}(H_i \rho)$ with ρ a four-mode optical state can be expressed as

$$\text{Tr}(H_i \rho) = \sum_{jk} q_{jk} \langle j | Z_1 | j \rangle \langle k | Z_2 | k \rangle \int_0^\pi \frac{d\phi_3 d\phi_4}{\pi^2} \int_{-\infty}^\infty dx_3 dx_4 p_{jk}(\phi_3, \phi_4, x_3, x_4) K(\phi_3, \phi_4, x_3, x_4, \sigma_{+,3} \sigma_{-,4}), \quad (5)$$

where $q_{jk} p_{jk}(\phi_3, \phi_4, x_3, x_4) = \langle jk | \langle x_{\phi_3} x_{\phi_4} | \rho | jk \rangle | x_{\phi_3} x_{\phi_4} \rangle$ is the joint probability of getting j photons and k photons on the first two modes and obtaining the results x_{ϕ_3} and x_{ϕ_4} when measuring the quadratures X_{3,ϕ_3} and X_{4,ϕ_4} where $X_{p,\phi_p} = (a_p^\dagger e^{i\phi_p} + a_p e^{-i\phi_p})/2$ and $K(\phi_1, \dots, \phi_M, x_1, \dots, x_M, \beta)$ the kernel function associated with $\sigma_{+,3} \sigma_{-,4}$ which can be efficiently calculated. Equation (5) is a typical Monte-Carlo integration, which means we can do repeated samplings to estimate the value. From the operational level, to get a sample, Eq. (5) means we can first do a photon-number measurement on the first two qubits and then do a homodyne measurement on the last two qubits. For the homodyne measurement, we need to get ϕ_3 and ϕ_4 from a uniform distribution, and then measure the corresponding X_{3,ϕ_3} and X_{4,ϕ_4} and get the results

x_{ϕ_3} and x_{ϕ_4} which are then used to calculate the value of the corresponding kernel function.

For the measurement cost, we prove that the required number of samples following the procedure of Eq. (5) to estimate Eq. (4) is proportional to $\{2.07317^k, \epsilon^{-2}, m_H, m_V, \chi^{-2}\}$ with m_H the number of terms in $H_T^{\text{JW}}(\vec{\beta})$, m_V the number of terms in $V_C^\dagger(\vec{\beta}) V_C(\vec{\beta})$ [Note that when $V_C^\dagger(\vec{\beta}) V_C(\vec{\beta})$ is reduced to Q , we can simply do pure photon-number measurements to estimate the projection ratio.], k the maximum number of qubits with σ_- and σ_+ in terms, ϵ the desired precision, and χ the lower bound of the projection ratio. Note that k has restricted values for HF and CISD methods. See also Appendix B.

C. Error mitigation against photon loss

Currently, the above discussions treat LQOS to be ideal, which neglects the photon loss error in real devices. Photon loss as the prominent noise channel is non-negligible, especially for our proposal, where photon loss will lead to BS-C VQE searching outside the legal subspace. Thus, besides the corrections in Eq. (4), we also need corrections for photon loss. Since we use a hybrid measurement strategy, which seems unable to correct the photon loss error by commonly used post selections at first glance, we now show a procedure to correct the photon loss error, whose reason can be found in Appendix C.

To illustrate the concrete correction procedure, without loss of generality, we can consider an H_i that only has one σ_+ and one σ_- as an example. We have N single-photon sources as input to match the correct number of electrons. First, we do the mentioned hybrid measurements. Here, since $\sigma_+ \sigma_- = |10\rangle\langle 01|$, we require detecting $N - 1$ photons on I and Z modes with at most one photon on each mode to proceed with homodyne detections. We can do repeated such measurements to obtain a raw expectation value of H_i denoted as $\langle H_i \rangle_{\text{raw}}$. Next, we need a normalization factor, which can be obtained by doing repeated pure photon-number measurements on all modes many times.

Suppose among these measurements, we have n_1 times of getting N photons in total on all modes with at most one photon on each mode and n_2 times of getting $N - 1$ photons in total on I and Z modes with at most one photon on each mode. And among those n_1 events, we have n_3 times where there is one photon in two nondiagonal modes and $N - 1$ photons in diagonal modes. The true value is then corrected by $(n_3/n_1)(n_2/n_3) \langle H_i \rangle_{\text{raw}}$. This value has already taken the projection ratio into consideration. For other H_i , this procedure can be easily generalized. The efficiency of this error mitigation method merely depends on the level of photon loss and has no additional nonscalable cost.

IV. ADVANTAGE DISCUSSION

We have introduced all components of our proposal and the whole workflow of the algorithm, which we named

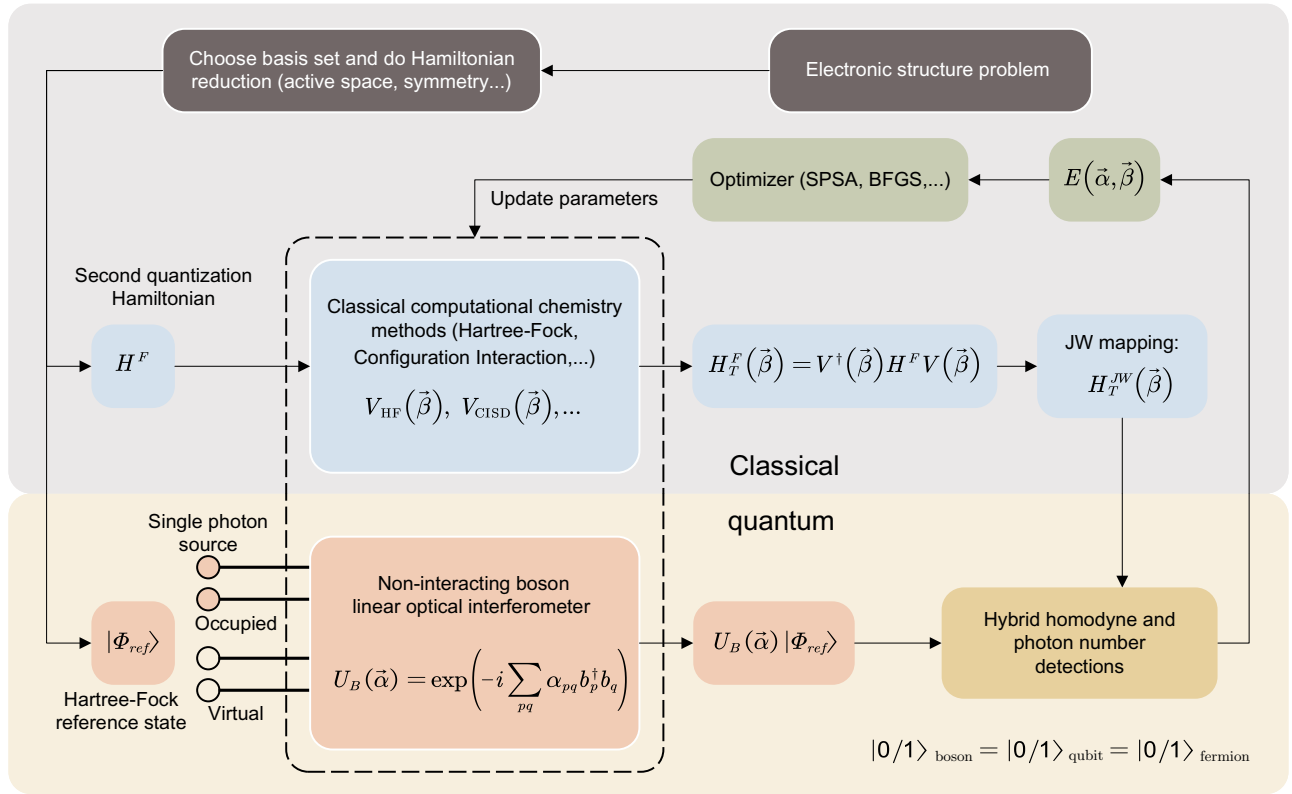


FIG. 1. The framework of boson sampling-classic variational quantum eigensolver. In boson sampling-classic variational quantum eigensolver encoding, the 0, 1 occupation of the boson, the 0, 1 occupation of the qubit, and the 0, 1 occupation of the fermion are equivalent. Given the electronic structure Hamiltonian of a molecule, the task is to obtain its ground-state energy. Typically, we choose a suitable basis set and do Hamiltonian reduction if necessary to obtain the second quantization Hamiltonian H^F and the Hartree-Fock reference state $|\Phi_{ref}\rangle$. In boson sampling-classic variational quantum eigensolver, H^F will be transformed by classical operations like Hartree-Fock and configuration interaction singles and doubles. The transformed Hamiltonian is then mapped to a qubit Hamiltonian by Jordan-Wigner mapping. The reference state gives the information on the initial photon source preparation. The occupied orbitals correspond to optical modes with single-photon input, and the virtual orbitals correspond to modes with vacuum input. Such an initial state is then passed to parametrized noninteracting boson dynamics, i.e., linear optical interferometer. The output state is used for the hybrid measurements to obtain the energy expectation value, which, as the cost function, is fed to the classical optimizer to update the parameters $\vec{\alpha}$ and $\vec{\beta}$ for the next iteration until converged.

BS-C VQE, has been summarized in Fig. 1. We also explain the physical intuitions of BS-C VQE in Figs. 2(a) and 2(b). Here, we want to have several discussions on the advantages of our methods over classical and other quantum methods.

Under our encoding, since BS and FS are different, the noninteracting boson dynamics in LOI lead to a different transition probability distribution from noninteracting fermion dynamics since permanents and determinants in Eq. (2) are fundamentally different. Thus, unlike HF, LOI belongs to the multideterminant type. This “interacting” part can be understood as a multideterminant reference state [20] generator for the following classical part, and the bosonic nature of these resources indicates they have small overlaps with those classical methods originating from electron excitations (see Appendix F). Thus, when the optimization procedure is converged, we should expect to obtain more accurate ground-state energy estimations

than using the classical methods alone. Also, since the circuit depth for a universal quantum BS circuit has a depth $\mathcal{O}(M)$ [24], it might also have the advantage over classical multireference methods whose scaling is typically proportional to the dimension of the electron subspace [32,33].

The numerator of $E(\vec{\alpha}, \vec{\beta})$ can be expanded as

$$\begin{aligned} & \langle \Phi(\vec{\alpha}) | H_T^{JW}(\vec{\beta}) | \Phi(\vec{\alpha}) \rangle \\ &= \sum_{ij} H_T^{JW}(\vec{\beta})_{ij} \text{Per}(U_{B,0i}(\vec{\alpha})) \text{Per}(U_{B,0j}(\vec{\alpha})), \end{aligned} \quad (6)$$

where 0 denotes $|\Phi_{ref}\rangle$. The permanents that appeared in this expression provide evidence for our approach to be classically intractable, which is similar to UCCSD, whose unitarity makes it classically intractable. We want to mention that while the hardness of calculating the permanents

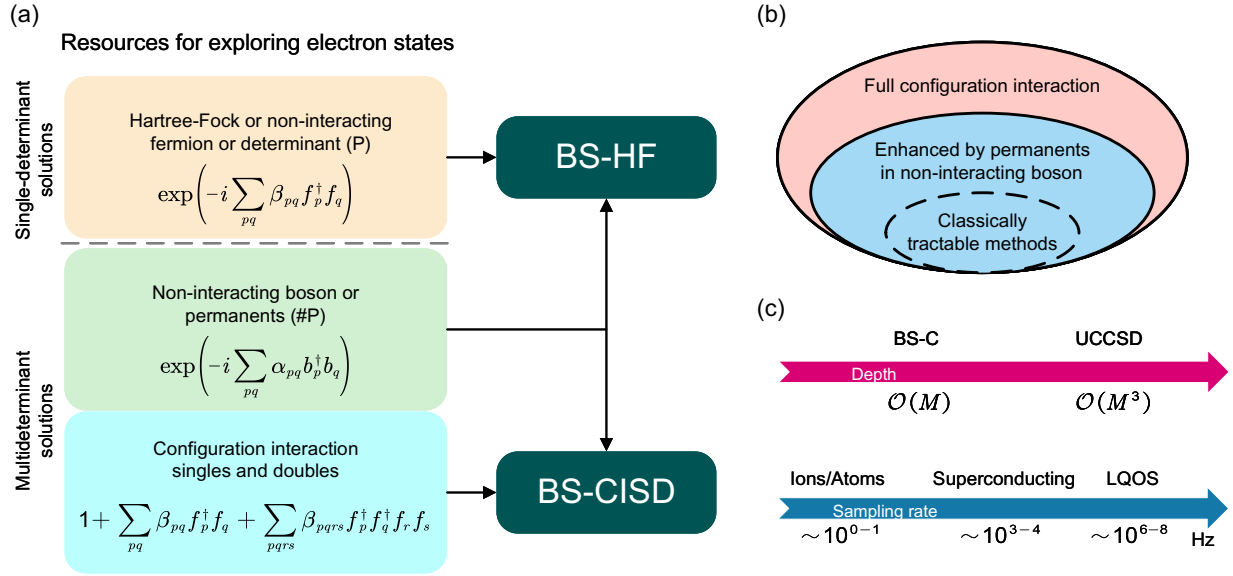


FIG. 2. Properties of boson sampling-classic ansatz. (a) and (b) In the Schrödinger picture, we can see both the noninteracting boson dynamics and the classical operations act on the reference state. Different operations give different resources to explore the electron subspace. Unitary coupled cluster singles and doubles and classical methods like Hartree-Fock and configuration interaction singles and doubles give resources for electron excitations with various orders. In contrast, permanents appearing in noninteracting boson dynamics are a different type of resource that results from the differences between bosons and fermions. Also, since permanent is classically intractable, it is a quantum resource that may manifest potential advantages. Thus, the main idea of boson sampling-classic is to use such quantum resources to enhance the accuracy of classical methods. When the classical method is Hartree-Fock, we have boson sampling Hartree-Fock. When the classical method is configuration interaction singles and doubles, we have boson sampling-configuration interaction singles and doubles. Note that the classical methods are not restricted to these two. (c) Boson sampling-classic has the attractive advantage of requiring much shallower depth than unitary coupled cluster singles and doubles. Also, the linear quantum optical system for our proposal can have a much higher sampling rate than other platforms.

does not necessarily indicate that the summation of the permanents in Eq. (6) is also classically intractable, to the best of our knowledge, no existing classical algorithm can efficiently simulate our approach. There is a recent work [34] showing how to classically efficiently simulate the expectation value of observables in linear optics; however, their algorithm has the requirement that the Frobenius norm should be polynomially large in N and M , which does not meet our cases, where due to the JW transformation, observables we considered can have exponentially large norms. At the same time, just like UCCSD, since our method is designed for the chemical problem, which is more problem specialized and restricted, our approach might also be helpful to mitigate the barren plateau problem [35] according to the expressibility-trainability trade-off [36].

Finally, when considering the realizations, BS-C VQE has its advantage over the UCCSD. Unlike UCCSD where we need a locally connected quantum circuit of depth around $\mathcal{O}(M^3)$ [22] to build in the general case where all excitations are allowed, BS-C only needs a $\mathcal{O}(M)$ -time LOI [24] which has the same scaling as the quantum implementation of HF ansatz $U_F(\beta)$ of depth around $\mathcal{O}(M)$ [37] but is with the speed of light as the propagation speed. Thus, combined with the efficient error mitigation

method we will introduce below against the photon loss, BS-C VQE is much more hardware efficient. What is more, the LQOS for our proposal has the advantage of the sampling rate compared with other platforms, which is highly demanded for variational quantum algorithms since massive repeated measurements are required. Current state-of-the-art experiments show that the sampling rate of LQOS can reach around 10^{6-8} Hz [38] whereas the sampling rate is around 10^{3-4} Hz for superconducting qubits [39] and 10^{0-1} Hz for ion traps [40] and neutral atoms [41]. We summarize these in Fig. 2(c).

V. NUMERICAL EXPERIMENTS

A. Solving ESP of small molecules

We demonstrate the performance of BS-C VQE by running numerical experiments on several molecules and drawing their potential energy curves in Fig. 3. We test molecules including LiH, BeH₂, and H₄ with line geometry and H₄ with square geometry. All molecules use the STO-3G basis set, and the active space reduction method [16] is used for LiH, BeH₂. The resulting JW Hamiltonians are 6, 8, 8, and 8 qubits. We add data of FCI, HF, and CISD (with HF state as input) as references. We also show the projection ratios corresponding to the BS-C VQE solutions.

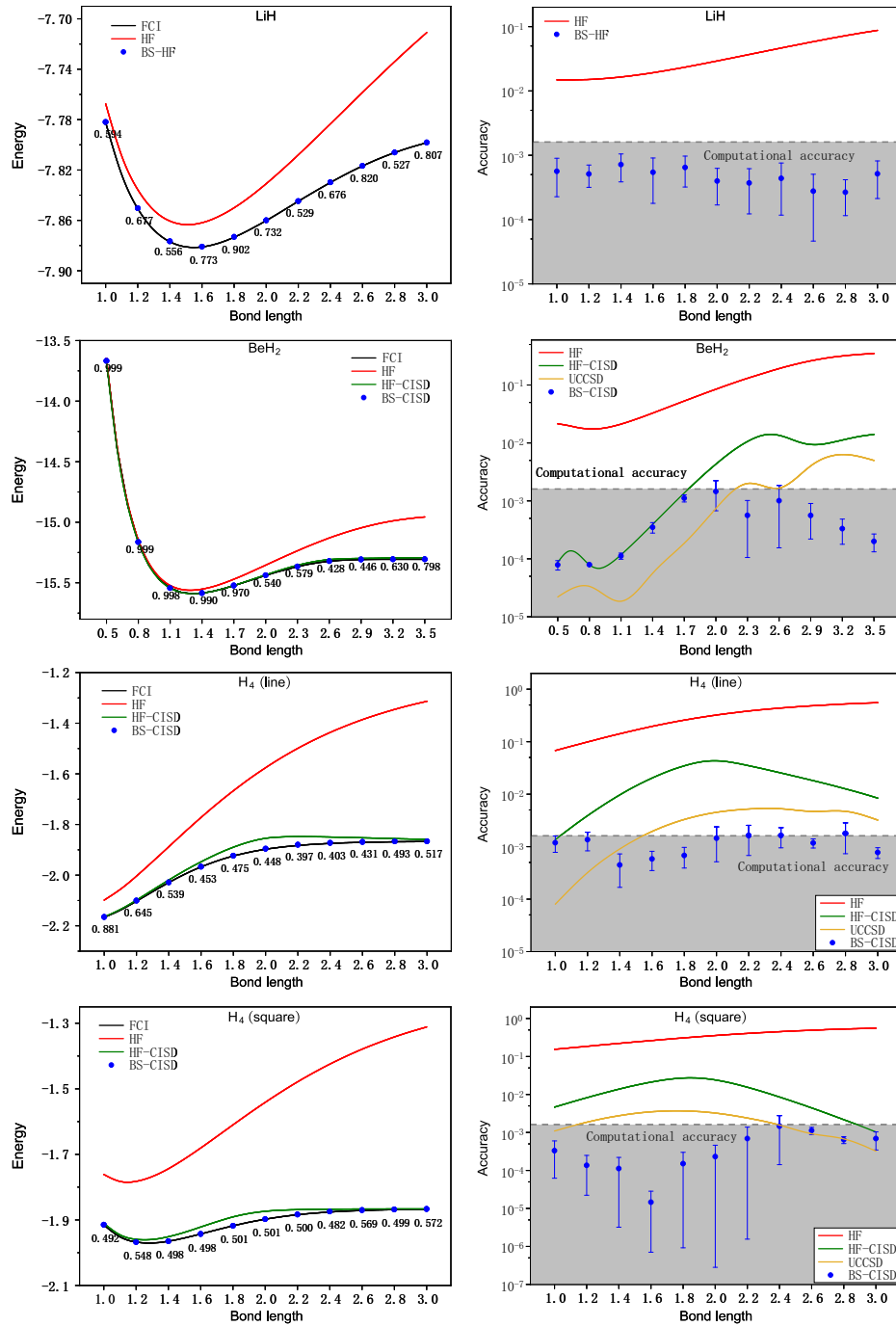


FIG. 3. Numerical experiments on several molecules (LiH, BeH₂, and H₄ with line geometry and H₄ with square geometry). The potential energy curves of molecules as functions of bond length (Li–H bond, all Be–H bonds, all H–H bonds, and all H–H bonds) are shown on the left, and the accuracy (energy differences) from the full configuration interaction results are shown on the right. Here, full configuration interaction is defined in the considered basis set (Appendix E), where for LiH, full configuration interaction is equivalent to configuration interaction singles and doubles. And Hartree-Fock configuration interaction singles and doubles refers to the configuration interaction singles and doubles method with the Hartree-Fock state as input. We run boson sampling Hartree-Fock for LiH, and boson sampling-configuration interaction singles and doubles for the others. Ten experiments for each molecule, and to have large projection ratios, we only allow occupied-to-virtual excitations and virtual-to-virtual excitations in our ansatz. The numbers attached to blue points are the corresponding average projection ratios. For molecules BeH₂, H₄ (line), and H₄ (square), we also show the results of the unitary coupled cluster singles and doubles method on the right. The shaded gray region represents the area within the computational accuracy [30] or chemical accuracy (1.6×10^{-3} hartree). The classical optimizer we used is limited-memory Broyden–Fletcher–Goldfarb–Shanno algorithm for bound-constrained optimization (L-BFGS-B) [31]. The bond lengths are reported in Å, and energies are reported in hartree.

We can see BS-C has enhancements over its classical part and can reach the chemical accuracy (1.6×10^{-3} hartree) at all regions. Note that since chemical accuracy sometimes refers to the accuracy from the complete basis set of a molecule, to avoid confusion, from now on, we will use the word computational accuracy [30] instead. We use BS-HF for LiH, where we can see that while we only use noninteracting bosons and noninteracting fermions, the results are still rather accurate. For the other three molecules, we test the BS-CISD method, and we also show the results of using UCCSD for comparisons. Here, the ansatz designs do not consider the spin symmetry, and one can always introduce this symmetry to further reduce the number of parameters.

From the numerical results, we can find clear differences between noninteracting boson (BS) and noninteracting fermion (HF). For LiH, the performance of BS-HF demonstrates that a combination of noninteracting boson and fermion dynamics will manifest the interactions between electrons. For the other three molecules, we can also see the advantage of using noninteracting bosons as the multireference input state over noninteracting fermions as the single-reference input state for the CISD methods. For these three molecules, we can also find the advantages of BS-CISD over UCCSD. UCCSD is unable to reach the computational accuracy under all bond lengths, while BS-CISD can consistently reach the computational accuracy, even with shallower quantum circuits.

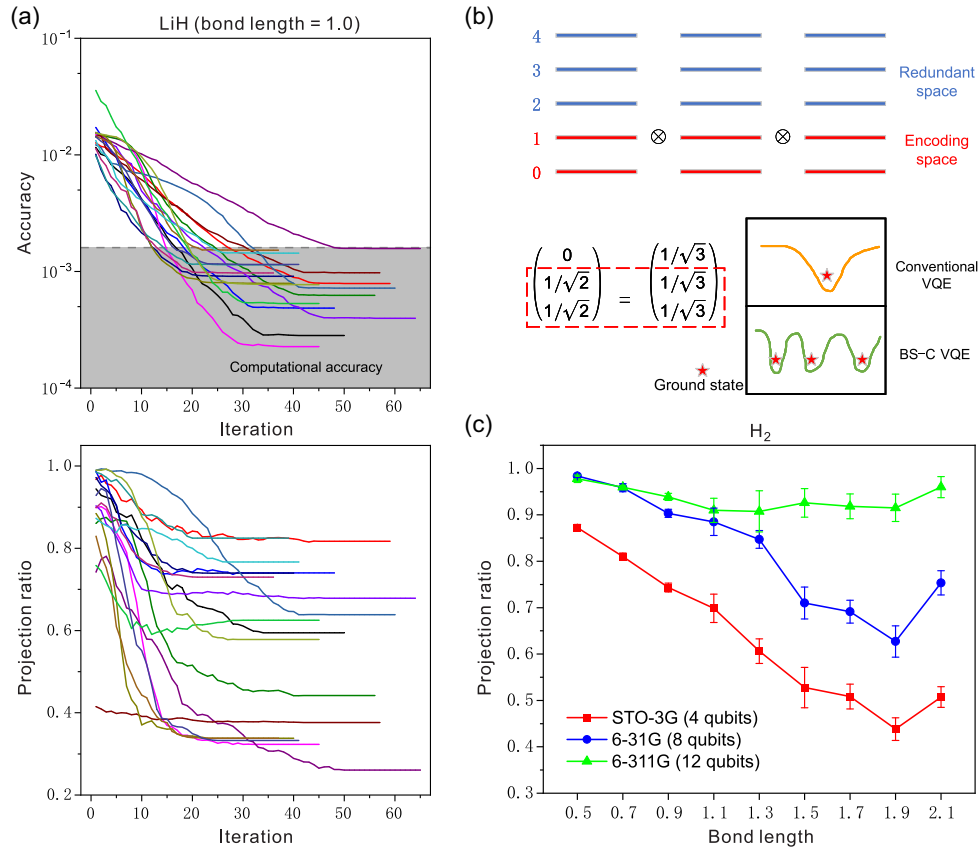


FIG. 4. Projection ratio of boson sampling-classic variational quantum eigensolver. (a) LiH molecule as an example. We fix the bond length as 1.0 and repeatedly run boson sampling-classic variational quantum eigensolver. We see that all presented tests have achieved computational accuracy. However, as explained, their corresponding projection ratios are totally different. (b) In boson sampling-classic variational quantum eigensolver encoding, we only utilize 0 and 1 occupation dimensions, and all higher photon occupations are redundant. However, after the action of linear optical interferometer, part of the output state will unavoidably jump to these redundant dimensions. This leads to an interesting point of boson sampling-classic variational quantum eigensolver that two physically different states can be treated as the same states in the encoding space. Thus, when optimizing boson sampling-classic variational quantum eigensolver, it is possible that we can get multiple sets of parameters that correspond to the ground state. (c) The number of orbitals and the number of electrons have a crucial influence on the projection ratio. The smaller the number of electrons compared to the number of orbitals, the higher the projection ratio will appear. We test the H₂ molecule with different choices of basis set to show this phenomenon. The number of electrons is 2 and the orbital number is 4 for STO-3G, 8 for 6-31G, and 12 for 6-311G. The error bars are collected from the tests that achieve computational accuracy. We can easily see that the larger number of orbitals gives higher projection ratios. We use boson sampling Hartree-Fock for LiH and H₂.

B. Projection ratio

Now, we want to talk about the projection ratio. The projection ratio $\langle \Phi(\vec{\alpha}) | Q | \Phi(\vec{\alpha}) \rangle$ has great influence on the total measurement cost of estimating $E(\vec{\alpha}, \vec{\beta})$. Our statistical analysis shows that the mean-squared error containing both bias and variance of estimating $E(\vec{\alpha}, \vec{\beta})$ will increase by $\mathcal{O}(\langle Q \rangle^{-2})$ times compared with the ideal case with a projection ratio equal to one. Thus, to make the measurements of $E(\vec{\alpha}, \vec{\beta})$ truly scalable, the projection ratio cannot be exponentially small, which is also the reason for the existence of the sign problem [42] in quantum Monte-Carlo methods.

We can increase the projection ratios from two aspects. The first can be understood from Figs. 4(a) and 4(b). We use BS-HF for the LiH molecule as a direct demonstration where we can see that while all experiments finally give energy reached the computational accuracy, they have totally different projection ratios. This means there may be physically different states $|\Phi(\vec{\alpha})\rangle$ which have similar components in the electron space, but can have different components outside the electron subspace and resulting in different projection ratios. Thus, unlike the conventional VQE, where there is only one minimum energy, there may be multiple solutions in our algorithm. To help the optimization converge to those with high projection ratios, a penalty term $-\lambda \langle Q \rangle$ can be introduced to the cost function, which helps solutions with higher projection ratios have lower cost function values. Another method is to only allow occupied-to-virtual photon excitations and virtual-to-virtual photon excitations to reduce the populations outside the encoding space, which is the method we used in numerical experiments. Note that this can be further adjusted by only allowing those to preserve the spin and orbital symmetries [22].

These methods, however, are not enough since there may be situations where even the highest projection ratio among the solutions is still small. We can understand this by defining the ratio $r(M, N) = \binom{M}{N} / \binom{M+N-1}{N}$ between the dimension of the fermion Hilbert space and the dimension of the boson Hilbert space. Obviously, $r(M, N)$ is a fair judge for the projection ratio. If we fix the number of orbitals M and enlarge the number of electrons N , $r(M, N)$ will decrease. In contrast, if we fix the number of electrons N and enlarge the number of orbitals M , $r(M, N)$ will increase. Thus, to avoid low projection ratios, M should be large enough compared to N (also see Appendix D). Luckily, while N depends on the number of electrons in the molecule, the number of orbitals has no restrictions and can be arbitrarily large.

In Fig. 4(c), we run BS-HF for H2 with different choices of basis sets, including STO-3G, 6-31G, and 6-311G. The resulting number of qubits (spin orbitals) is 4, 8, and 12, respectively, with the number of electrons fixed as 2. We can see that as the number of orbitals gets larger, the average projection ratios of the experiments that reach the

computational accuracy get larger as well. More rigorously, we proved that if $M = \eta N^2$, $r(M, N)$ will have a lower bound $e^{1/\eta}$, which indicates the empirical requirements to make the measurements scalable. Note that such a relation between M and N is also a believed requirement for BS to be classically intractable [7,43]. Thus, if the hardware allows, we should choose a basis set as large as possible, since this can bring three benefits at the same time: more accurate modeling for the molecules, more expected classical hardness of BS, and higher projection ratio.

VI. SUMMARY AND OUTLOOK

In summary, we give a novel hybrid quantum-classical ansatz using only passive LQOS for running VQE to solve ESP. The permanents that appeared in noninteracting boson dynamics are a new type of resource for exploring the electron subspace, which can help enhance the accuracy of quantum chemistry. We use BS-HF and BS-CISD as two concrete settings and show their performances by numerical results. The introduction of the hybrid measurement strategy with the methods against photon loss error and low projection ratios helps to have a scalable measurement cost. Both the theory and the simulations showed that our proposal may open a road for quantum applications using passive LQOS in the NISQ era.

We want to emphasize that this proposal is not restricted to what we presented here. For example, the classical methods are not restricted to HF and CI and can be generalized. Also, with the developments of LQOS, to get better accuracy, one can use entangled reference states [20] or introduce intermediate measurements for more freedom and higher accuracy. It is worth noticing that there are still several open questions on this algorithm that need to be further investigated such as the level of performance enhancements of this algorithm on larger molecules and better measurement strategies such as developing efficient tomography methods with pure homodyne or photon-number measurements [44] and adopting the ideas of recently developed quantum learning methods [45].

Finally, we briefly discuss the feasibility of an experimental demonstration. Our protocol requires a standard boson sampling (and its variants) setup where the single-photon quantum light sources are injected into a tunable multimode interferometer and the outputs are resorted to hybrid homodyne and photon-number detections. All the required components have been demonstrated in previous experiments, although separately. For example, Gaussian boson sampling has been tested up to 255 output photons [11] and 216-mode reconfigurable optical circuits [10]. High-quality single-photon sources [46,47] and high-fidelity homodyne measurements have been realized in Refs. [48,49]. Therefore, our scheme is within the reach of current technologies.

We used Python packages including Qiskit [31], OpenFermion [50], PySCF [51], and The Walrus [52] for parts of our simulations.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are openly available [53].

APPENDIX A: HOMODYNE MEASUREMENT

First, consider a single mode [27]. An operator H can be expressed in the coherent state basis,

$$\begin{aligned} H &= \int \frac{d^2\alpha}{\pi} \text{Tr}[H D^\dagger(\alpha)] D(\alpha) \\ &= \int_0^\pi \frac{d\phi}{\pi} \int_{-\infty}^\infty dr \frac{|r|}{4} \text{Tr}(H e^{irX_\phi}) e^{-irX_\phi}, \end{aligned} \quad (\text{A1})$$

where $D(\alpha) = e^{(\alpha b^\dagger - \alpha^* b)}$ is the displacement operator and $\alpha = -ir e^{i\phi}/2$. The quadrature operator X_ϕ is defined as $X_\phi = (b^\dagger e^{i\phi} + b e^{-i\phi})/2$. The expectation value of H under an optical state ρ can then be expressed as

$$\begin{aligned} \langle H \rangle &= \text{Tr}(H\rho) \\ &= \text{Tr} \left(\int \frac{d^2\alpha}{\pi} \text{Tr}(H D^\dagger(\alpha)) D(\alpha) \rho \right) \\ &= \int_0^\pi \frac{d\phi}{\pi} \int_{-\infty}^\infty dr \frac{|r|}{4} \text{Tr}(H e^{irX_\phi}) \text{Tr}(\rho e^{-irX_\phi}) \\ &= \int_0^\pi \frac{d\phi}{\pi} \int_{-\infty}^\infty dx \int_{-\infty}^\infty dr \frac{|r|}{4} \text{Tr}(H e^{irX_\phi}) p(\phi, x) e^{-irx} \end{aligned}$$

$$\begin{aligned} &= \int_0^\pi \frac{d\phi}{\pi} \int_{-\infty}^\infty dx p(\phi, x) \int_{-\infty}^\infty dr \frac{|r|}{4} \text{Tr}(H e^{ir(X_\phi - x)}) \\ &= \int_0^\pi \frac{d\phi}{\pi} \int_{-\infty}^\infty dx p(\phi, x) K(\phi, x, H). \end{aligned} \quad (\text{A2})$$

The probability $p(\phi, x) = \langle x_\phi | \rho | x_\phi \rangle$ is obtained by measuring $\langle X_\phi | \rho | X_\phi \rangle$. We see that this is a standard Monte-Carlo estimation. The kernel function $K(\phi, x, H)$ is defined as

$$K(\phi, x, H) = \int_{-\infty}^\infty dr \frac{|r|}{4} \text{Tr}(H e^{ir(X_\phi - x)}). \quad (\text{A3})$$

When $H = |n + \lambda\rangle\langle n|$, the kernel function has an analytic expression,

$$\begin{aligned} K(\phi, x, |n + \lambda\rangle\langle n|) &= 2e^{-i\lambda\phi} \sqrt{\frac{n!}{(n + \lambda)!}} e^{-x^2} \sum_{v=0}^n \frac{(-1)^v}{v!} \binom{n + \lambda}{n - v} \\ &\quad \times (2v + \lambda + 1)! \text{Re} \left[(-i)^\lambda \mathcal{D}_{-(2v + \lambda + 2)}(-2ix) \right], \end{aligned} \quad (\text{A4})$$

where $\mathcal{D}_l(x)$ the parabolic cylinder function that can be easily calculated. In our algorithm, we will need the kernel functions of σ_+ , σ_- , and Z . According to Eq. (A4), we can calculate the range of the kernel functions of σ_+ and σ_- , which are among $[-2.07317, 2.07317]$ and the kernel functions of Z , which are among $[-2.92345, 5.33333]$.

For multimode situations, the generalization is natural,

$$\begin{aligned} \langle H \rangle &= \int_0^\pi \frac{d\phi_1 d\phi_2 \cdots d\phi_M}{\pi^M} \int_{-\infty}^\infty dx_1 dx_2 \cdots dx_M \\ &\quad p(\phi_1, \phi_2, \dots, \phi_M, x_1, x_2, \dots, x_M) \\ &\quad K(\phi_1, \phi_2, \dots, \phi_M, x_1, x_2, \dots, x_M, H), \end{aligned} \quad (\text{A5})$$

where

$$\begin{aligned} &K(\phi_1, \phi_2, \dots, \phi_M, x_1, x_2, \dots, x_M, H) \\ &= \int_{-\infty}^\infty dr_1 dr_2 \cdots dr_M \prod_{m=1}^M \frac{|r_m|}{4} \text{Tr}(H e^{ir_m(X_{\phi_m} - x_m)}). \end{aligned} \quad (\text{A6})$$

We can see from Eq. (A6) that the multimode kernel function is a simple product of the single-mode kernel function if H is a product operator, which makes the complexity of calculating the kernel function scalable. Also, we do not need to calculate all values of all kernel functions in advance. We only need to do a measurement followed by a kernel function calculation. Thus, the resource for calculating the kernel function also has linear scaling with the number of measurements.

The variance of using Eq. (A2) to estimate the expectation value of H depends on the kernel function. When a kernel function has the range $[a, b]$, then the variance of Eq. (A2) is bounded by $((b - a)^2)/4$. Since the kernel functions of σ_+ , σ_- , and Z have ranges larger than $[-2, 2]$ and terms we want to evaluate in the cost function obtained by the JW mapping is global, the kernel functions of these terms can have exponentially large ranges and thus pure homodyne measurements will be nonscalable, which leads to the hybrid measurement strategy introduced in the main text. On the other hand, since $p(\phi_1, \phi_2, \dots, \phi_M, x_1, x_2, \dots, x_M)$ has no dependence on the concrete form of H , if all terms in a Hamiltonian are local, then the same set of samples can be used to estimate all expectation values of these terms and will greatly reduce the sampling complexity.

APPENDIX B: MEASUREMENT COMPLEXITY

In this Appendix, we will estimate the measurement complexity of estimating,

$$E(\vec{\alpha}, \vec{\beta}) = \frac{\langle \Phi(\vec{\alpha}) | H_T^{\text{JW}}(\vec{\beta}) | \Phi(\vec{\alpha}) \rangle}{\langle \Phi(\vec{\alpha}) | V_C^\dagger(\vec{\beta}) V_C(\vec{\beta}) | \Phi(\vec{\alpha}) \rangle}, \quad (\text{B1})$$

assuming there is no photon loss error. Before the analysis, we will introduce the estimator that will be used.

- (a) Estimator for $E[X]/E[Y]$: When X and Y are two independent random variables, the value of the ratio of their expectations $E[X]/E[Y]$ can be estimated by an asymptotically unbiased estimator $\bar{X}/\bar{Y}$ where $\bar{X}$ and $\bar{Y}$ are the averages of X and Y . It has been shown in Ref. [54] that the expectation and the variance of this estimator are

$$E\left[\frac{\bar{X}}{\bar{Y}}\right] \approx \frac{E[X]}{E[Y]} + \frac{E[X]}{E[Y]^3} \text{Var}[\bar{Y}], \quad (\text{B2})$$

$$\text{Var}\left[\frac{\bar{X}}{\bar{Y}}\right] \approx \frac{E[X]^2 \text{Var}[\bar{Y}] + E[Y]^2 \text{Var}[\bar{X}]}{E[Y]^4}. \quad (\text{B3})$$

In our case, the numerator X corresponds to $\langle H_T^{\text{JW}} \rangle$ short for $\langle \Phi(\vec{\alpha}) | H_T^{\text{JW}}(\vec{\beta}) | \Phi(\vec{\alpha}) \rangle$ and the denominator Y corresponds to $\langle V_C^\dagger V_C \rangle$ short for $\langle \Phi_{\text{ref}} | V_C^\dagger(\vec{\beta}) V_C(\vec{\beta}) | \Phi_{\text{ref}} \rangle$. For the numerator, we assume $H_T^{\text{JW}}(\vec{\beta})$ has the form $H_T^{\text{JW}}(\vec{\beta}) = \sum_{i=1}^{m_H} h_i H_i$ with H_i containing σ_+ , σ_- , Z , and I . For each P_i , we do the photon-number measurements on qubits with Z and I and homodyne measurements on qubits with σ_+ and σ_- . Suppose each term has at most k_h σ_+ and k_h σ_- , since the kernel functions of σ_+ and σ_- have ranges among $[-2.07317, 2.07317]$, the remaining Z and I have diagonal elements -1 and 1 , the whole variance is then bounded by 2.07317^{4k_h} . If we do repeated measurements for N_H/m_H times for P_i , the resulting variance is bounded

by $m_H 2.07317^{4k_h}/N_H$. Following this, the total variance of the numerator has the bound

$$\text{Var}[\overline{\langle H_T^{\text{JW}} \rangle}] \leq \sum_{i=1}^{m_H} \frac{2.07317^{4k_h} m_H h_i^2}{N_H}. \quad (\text{B4})$$

For the denominator, similarly, if we assume $V_C^\dagger V_C = \sum_{i=1}^{m_V} g_i V_i$ and each term has at most k_v σ_+ and k_v σ_- . Then, if we do measurements N_V/m_V times for each P_i , the total variance of the denominator has the bound

$$\text{Var}[\overline{\langle V_C^\dagger V_C \rangle}] \leq \sum_{i=1}^{m_V} \frac{2.07317^{4k_v} m_V g_i^2}{N_V}. \quad (\text{B5})$$

Assume the projection ratio $\langle Q \rangle$ short for $\langle \Phi(\vec{\alpha}) | Q | \Phi(\vec{\alpha}) \rangle$ has a lower bound: $\langle Q \rangle \geq \chi$ and define $\langle H_T^{\text{JW}} \rangle_r = \langle H_T^{\text{JW}} \rangle / \langle Q \rangle$ and $\langle V_C^\dagger V_C \rangle_r = \langle V_C^\dagger V_C \rangle / \langle Q \rangle$. By putting Eqs. (B5) and (B4) into Eqs. (B2) and (B3), we obtain that the bias of estimating $E(\vec{\alpha}, \vec{\beta})$ is bounded by

$$\begin{aligned} \text{Bias}[E(\vec{\alpha}, \vec{\beta})]^2 &\leq \left(\frac{\langle H_T^{\text{JW}} \rangle}{\langle V_C^\dagger V_C \rangle} - \frac{\overline{\langle H_T^{\text{JW}} \rangle}}{\overline{\langle V_C^\dagger V_C \rangle}} \right)^2 \\ &\approx \left(\frac{\langle H_T^{\text{JW}} \rangle}{\langle V_C^\dagger V_C \rangle^3} \text{Var}[\overline{\langle V_C^\dagger V_C \rangle}] \right)^2 \\ &= \left(\frac{E(\vec{\alpha}, \vec{\beta})}{\langle V_C^\dagger V_C \rangle^2} \sum_{i=1}^{m_V} \frac{2.07317^{4k_v} m_V g_i^2}{N_V} \right)^2 \\ &= \left(\frac{E(\vec{\alpha}, \vec{\beta})}{\chi^2 \langle V_C^\dagger V_C \rangle_r^2} \frac{2.07317^{4k_v} m_V}{N_V} \sum_{i=1}^{m_V} g_i^2 \right)^2 \\ &\leq \frac{2.07317^{4k_v} m_V^2 (\sum_{i=1}^{m_V} g_i^2)^2 \|H_T^{\text{JW}}\|_2^2}{\chi^4 N_V^2}, \end{aligned} \quad (\text{B6})$$

and the variance of estimating $E(\vec{\alpha}, \vec{\beta})$ is bounded by

$$\begin{aligned} \text{Var}[E(\vec{\alpha}, \vec{\beta})] &\leq \frac{\langle H_T^{\text{JW}} \rangle^2 \text{Var}[\overline{\langle V_C^\dagger V_C \rangle}] + \langle V_C^\dagger V_C \rangle^2 \text{Var}[\overline{\langle H_T^{\text{JW}} \rangle}]}{\langle V_C^\dagger V_C \rangle^4} \\ &= \frac{E(\vec{\alpha}, \vec{\beta})^2 \sum_{i=1}^{m_V} \frac{2.07317^{4k_v} m_V g_i^2}{N_V} + \sum_{i=1}^{m_H} \frac{2.07317^{4k_h} m_H h_i^2}{N_H}}{\langle V_C^\dagger V_C \rangle^2} \\ &= \frac{E(\vec{\alpha}, \vec{\beta})^2 \frac{2.07317^{4k_v} m_V}{N_V} \sum_{i=1}^{m_V} g_i^2 + \frac{2.07317^{4k_h} m_H}{N_H} \sum_{i=1}^{m_H} h_i^2}{\langle V_C^\dagger V_C \rangle^2} \\ &\leq \frac{2.07317^{4k_v} m_V (\sum_{i=1}^{m_V} g_i^2) \|H_T^{\text{JW}}\|_2^2}{\chi^2 N_V} \\ &\quad + \frac{2.07317^{4k_h} m_H \sum_{i=1}^{m_H} h_i^2}{\chi^2 N_H}. \end{aligned} \quad (\text{B7})$$

In the above derivation, we use the fact that for HF and CISD, $\langle V_C^\dagger V_C \rangle_r \geq 1$. The mean squared error is defined as the sum of Eqs. (B6) and (B7). From these results, we can see that to make the measurement scalable, we need χ not to be exponentially small, and that k_v and k_h are restricted to specific values, which is true for HF and CISD.

APPENDIX C: ERROR MITIGATION AGAINST PHOTON LOSS ERRORS

Consider an optical state with N photons in M modes ($M > N$). Photon loss can be regarded as a photon-number exchange between the system and the environment, as shown below:

$$|\psi\rangle = \sum_{i=0}^N c_i |\psi_{N-i}\rangle |i\rangle_e. \quad (\text{C1})$$

In this equation, $|\psi\rangle$ represents the whole state, including the system and environment. $|\psi_{N-i}\rangle$ represents the $N - i$ photon state component in the system with i photons escaping to the environment, and c_i is a coefficient determined by the efficiency of the experimental system. Since the number of electrons in a molecule is fixed, we need to postselect the state $|\psi_N\rangle$. Note that $|\psi_N\rangle = d_F |\psi_{N,F}\rangle + \dots$ where $|\psi_{N,F}\rangle$ is the component living in the encoding fermion space. Thus, the true expectation value of a term H_i should be

$$\langle \psi_{N,F} | H_i | \psi_{N,F} \rangle. \quad (\text{C2})$$

Given the feature of the Hamiltonian after the JW mapping, as explained in the main text, terms of the Hamiltonian have nondiagonal σ_+ and σ_- for certain modes and diagonal Z and I for other modes. Nondiagonal terms, for example, $\sigma_+ \sigma_- = |10\rangle\langle 01|$, strongly restrict the form of state that gives rise to a nonzero contribution to the expectation of the term of the Hamiltonian. This can be seen by an elaborate example where H_i has k σ_- and k σ_+ . Nonzero contributions can only be given by an optical state that has k photons in nondiagonal modes, and due to the reason of the electron number conservation, $N - k$ photons should be in the diagonal modes. Therefore, in the operational level, we can first estimate $\langle \phi_{N-k,T} | H_i | \phi_{N-k,T} \rangle$ where $|\phi_{N-k,T}\rangle$ means $N - k$ photons in diagonal modes and T means the true state where there is no more than 1 photon in each mode according to the fermion encoding. In hybrid measurements, we first directly measure the photon number in diagonal modes, so, for this condition, only when $N - k$ photons are measured with no more than 1 photon in these modes, we are allowed to go to the homodyne measurement for nondiagonal modes. After this postselection procedure, we are measuring the state $|\phi_{N-k}\rangle = \alpha |\phi_{N-k,T}\rangle + \beta |\phi_{N-k,W}\rangle$, which includes the state we desired and also includes the

component in the wrong subspace, including those with more than one photon in a nondiagonal mode and those with less than k photons in all $2k$ nondiagonal modes. Luckily, for the undesired state $|\phi_{N-k,W}\rangle$, it has no nonzero contributions to the expectation value, and the hybrid measurements let us estimate $\langle H_i \rangle_{\text{raw}} = \langle \phi_{N-k} | H_i | \phi_{N-k} \rangle = |\alpha|^2 \langle \phi_{N-k,T} | H_i | \phi_{N-k,T} \rangle$. Thus, we only need a correction factor $1/|\alpha|^2$. Currently, $|\phi_{N-k,T}\rangle$ is not equal to $|\psi_{N,F}\rangle$ since it has other N -photon components. Suppose we have $|\psi_{N,F}\rangle = \gamma |\phi_{N-k,T}\rangle + \sum_i \kappa_i |\phi_{i \neq N-k,T}\rangle$, since other components also have zero contributions to the expectation value, we can simply add another correction factor $|\gamma|^2$. Thus, the true result is

$$\langle \psi_{N,F} | H_i | \psi_{N,F} \rangle = \frac{|\gamma|^2}{|\alpha|^2} \langle \phi_{N-k} | H_i | \phi_{N-k} \rangle. \quad (\text{C3})$$

To get these factors, we can add an additional measurement procedure where we do pure repeated photon-number measurements many times. Suppose during these measurements, we recorded n_1 times of events that detected N photons in all and no more than 1 photon in each mode, n_2 times of events that detect $N - k$ photon in diagonal modes with no more than 1 photon in these modes, and n_3 events among those n_1 ones where $N - k$ photons in the diagonal modes and k photons in the nondiagonal modes. Then we have $|\gamma|^2 = n_3/n_1$ and $|\alpha|^2 = n_3/n_2$ which can be used for the true expectation value estimations,

$$\langle \psi_{N,F} | H_i | \psi_{N,F} \rangle \approx \frac{n_3}{n_1} \frac{n_2}{n_3} \langle H_i \rangle_{\text{raw}}, \quad (\text{C4})$$

whose total sampling complexity can be estimated by a similar procedure as above in the measurement complexity section.

Regarding the error of using $(n_3/n_1)(n_2/n_3) = n_2/n_1$ to estimate $|\gamma|^2/|\alpha|^2$, suppose we do totally N_{qem} number of measurements and the probability of getting n_1 -associated event is p_1 and the probability of getting n_2 -associated event is p_2 , then we also have $|\gamma|^2/|\alpha|^2 = p_2/p_1$. Suppose we have the estimation $\mu_2 = n_2/N_{\text{qem}}$ for p_2 and the estimation $\mu_1 = n_1/N_{\text{qem}}$ for p_1 , then the variance of the estimation, according to the law of large numbers, becomes

$$\begin{aligned} \text{Var}[\mu_2/\mu_1] &= \frac{p_1^2 \text{Var}[\mu_2] + p_2^2 \text{Var}[\mu_1]}{p_1^4} \\ &= \frac{\text{Var}[\mu_2]}{p_1^2} + \frac{p_2^2 \text{Var}[\mu_2]}{p_1^4} \\ &\leq \frac{1}{p_1^2 N_{\text{qem}}} + \frac{p_2^2}{p_1^4 N_{\text{qem}}}. \end{aligned} \quad (\text{C5})$$

Therefore, to estimate within error ϵ , we require

$$N_{\text{qem}} \geq \left(\frac{1}{p_1^2} + \frac{p_2^2}{p_1^4} \right) \epsilon^{-2}. \quad (\text{C6})$$

For the concrete values of p_1 and p_2 , they are related to the noisy level of the concrete quantum device.

APPENDIX D: PROJECTION RATIO

The projection ratio highly depends on the ratio $r(M, N) = \binom{M}{N} / \binom{M+N-1}{N}$ between the dimension of the fermion Hilbert space and the dimension of the boson Hilbert space. $r(M, N)$ describes the portion of the encoding space (fermion) in physical space (boson). In the following, we will prove that if $M = \eta N^2$, $r(M, N)$ will have a lower bound $e^{1/\eta}$.

First, $r(M, N)$ can be re-expressed as

$$r(M, N) = \frac{M}{M+N-1} \frac{M-1}{M+N-2} \cdots \frac{M-N+1}{M}. \quad (\text{D1})$$

Thus, we have

$$r(M, N) > \left(\frac{M-N+1}{M} \right)^N. \quad (\text{D2})$$

If $M = \eta N^2$, we have

$$\left(\frac{M-N+1}{M} \right)^N = \frac{1}{\left(\frac{\eta N^2}{\eta N^2 - N + 1} \right)^N} = \frac{1}{\left(1 + \frac{N-1}{\eta N^2 - N + 1} \right)^N}. \quad (\text{D3})$$

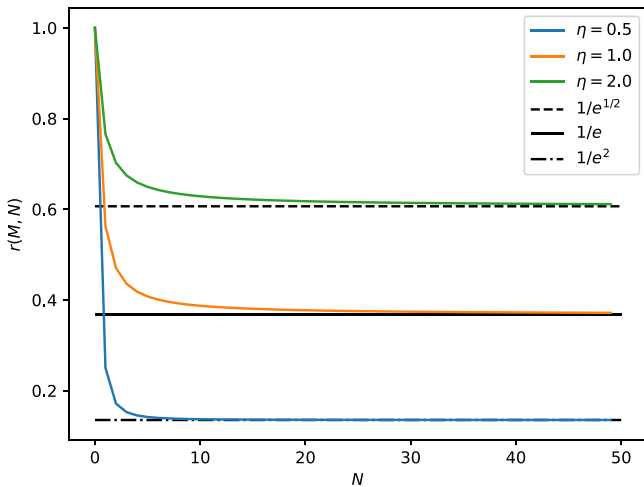


FIG. 5. $r(M, N)$ as functions of N . The number of orbitals is set to be $M = \eta N^2$ with $\eta = 0.5$, $\eta = 1$, and $\eta = 2$.

The RHS of Eq. (D3) has the limit,

$$\lim_{N \rightarrow \infty} \frac{1}{\left(1 + \frac{N-1}{\eta N^2 - N + 1} \right)^N} = \lim_{N \rightarrow \infty} \left(\frac{1}{\left(1 + \frac{1}{\eta N} \right)^{\eta N}} \right) = e^{\frac{1}{\eta}}. \quad (\text{D4})$$

Note that the RHS of Eq. (D3) decreases monotonically as N grows, which can be seen from Fig. 5. Thus, we have proved our conclusion.

APPENDIX E: MOLECULE INFORMATION IN FIG. 3

All molecules use the STO-3G basis set.

For the LiH molecule, we choose the active orbital $\{1, 2, 5\}$ with two active electrons. The orbital labels are ordered from low energy to high energy. Here, we sort the indices of the orbitals according to their energies from low to high.

For the BeH₂ molecule, the geometry is a line. We choose the active orbital $\{1, 2, 5, 6\}$ with four active electrons when the bond length is smaller than 1.9 and choose the active orbital $\{1, 2, 3, 6\}$ with four active electrons when the bond length is larger than 1.9. The orbital labels are ordered from low energy to high energy.

For the H₄ molecules with line geometry and square geometry, changing of bond length in the main text means changing all H–H bonds at the same time. There are a total of four orbitals and four electrons. No reduction method is used.

Each orbital above contains two spin orbitals.

For the systemic introduction of the active space methods, see Ref. [16].

The above orbital labels can be directly used in PySCF [51] and Qiskit [31] packages.

APPENDIX F: NONINTERACTING BOSONS AS INTERACTING FERMIONS

Here, we give a more formal explanation of why BS dynamics $U_B(\vec{\alpha})$ can be viewed as interacting fermions and its comparison with classical methods.

For noninteracting electrons or HF, we have

$$U_F(\vec{\beta}) = \exp \left(-i \sum_{pq} \beta_{pq} f_p^\dagger f_q \right). \quad (\text{F1})$$

For BS, since we only use the electron subspace with the projector $\mathcal{Q}$, the component we care about is

$$\mathcal{Q} U_B(\vec{\alpha}) |\Phi_{\text{ref}}\rangle = \left(\mathcal{Q} \exp \left(-i \sum_{pq} \alpha_{pq} b_p^\dagger b_q \right) \mathcal{Q} \right) |\Phi_{\text{ref}}\rangle. \quad (\text{F2})$$

We can do the Taylor expansion for $QU_B(\vec{\alpha})Q$,

$$\begin{aligned} QU_B(\vec{\alpha})Q &= Q \left(I - i \sum_{pq} \alpha_{pq} b_p^\dagger b_q - \frac{1}{2} \left(\sum_{pq} \alpha_{pq} b_p^\dagger b_q \right)^2 + \cdots \right) Q = A + B, \\ A &= \left(I - i \sum_{pq} \alpha_{pq} (Qb_p^\dagger Q)(Qb_q Q) - \frac{1}{2} \left(\sum_{pq} \alpha_{pq} (Qb_p^\dagger Q)(Qb_q Q) \right)^2 + \cdots \right), \\ B &= QU_B(\vec{\alpha})Q - A. \end{aligned} \quad (\text{F3})$$

For A , since QbQ and $Qb^\dagger Q$ are constrained in the electron subspace, due to their commutation relations, we have $QbQ = \sigma_-$ and $Qb^\dagger Q = \sigma_+$ become the qubit creation and annihilation operators. As a result, we will have

$$A = \exp \left(-i \sum_{pq} \alpha_{pq} \sigma_{+,p} \sigma_{-,q} \right). \quad (\text{F4})$$

Now, we can make a comparison with A and $U_F(\vec{\beta})$. A is essentially the noninteracting spin (qubits). Through the JW transformation, local qubit operators become nonlocal fermion operators (and vice versa), including many-body interactions, which noninteracting fermion dynamics cannot describe. More concretely, we can express A in terms of fermion operators,

$$A = \exp \left(-i \sum_{pq} \alpha_{pq} \left(f_p^\dagger e^{i\pi \sum_{j < p} f_j^\dagger f_j} \right) \left(f_q e^{-i\pi \sum_{j < q} f_j^\dagger f_j} \right) \right). \quad (\text{F5})$$

Therefore, $\sigma_{+,p} \sigma_{-,q}$ becomes a global operator under the JW transform (acting nontrivially on all electrons in $[p, q]$ if $q > p$ and in $[q, p]$ if $p > q$), and we must introduce the fermion interactions to describe them. Also, for extreme cases such as $p = 1$ and $q = n$, the interaction involves all fermions, which cannot be captured by simply introducing low-order electron excitations as in classical methods (e.g., CISD). Thus, this explains why $U_B(\vec{\alpha})$ can provide multireference resources beyond classical methods.

Note that, for now, we only consider the A part, or the noninteracting qubit part. However, we also have the B part, which captures the difference between noninteracting qubits and noninteracting bosons with a focus on the 0/1 Fock subspace. We now give an example to illustrate this difference. Consider a four-mode state $|1100\rangle$, there can exist terms like $O = Q(b_1 b_3^\dagger)(b_2 b_3^\dagger)(b_3 b_4^\dagger)Q$ in B , which gives $O|1100\rangle = |0011\rangle$. However, such a term in A will

have

$$\begin{aligned} &(Qb_1 Q)(Qb_2 Q)(Qb_3^\dagger Q)^2(Qb_3 Q)(Qb_4 Q) \\ &= \sigma_{-,1} \sigma_{-,2} \sigma_{+,3}^2 \sigma_{-,3} \sigma_{+,4} = 0. \end{aligned} \quad (\text{F6})$$

Thus, $U_B(\vec{\alpha})$ not only introduces fermion interactions but also cannot be captured by the noninteracting qubits (A) through JW transformation.

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