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Fock-Space Schrieffer–Wolff Transformation: Classically-Assisted Rank-Reduced Quantum Phase Estimation Algorithm

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Abstract: We present an extension of many-body downfolding methods to reduce the resources required in the quantum phase estimation (QPE) algorithm. In this paper, we focus on the Schrieffer–Wolff (SW) transformation of the electronic Hamiltonians for molecular systems that provides significant simplifications of quantum circuits for simulations of quantum dynamics. We demonstrate that by employing Fock-space variants of the SW transformation (or rank-reducing similarity transformations (RRST)) one can significantly increase the locality of the qubit-mapped similarity-transformed Hamiltonians. The practical utilization of the SW-RRST formalism is associated with a series of approximations discussed in the manuscript. In particular, amplitudes that define RRST can be evaluated using conventional computers and then encoded on quantum computers. The SW-RRST QPE quantum algorithms can also be viewed as an extension of the standard state-specific coupled-cluster downfolding methods to provide a robust alternative to the traditional QPE algorithms to identify the ground and excited states for systems with various numbers of electrons using the same Fock-space representations of the downfolded Hamiltonian. The RRST formalism serves as a design principle for developing new classes of approximate schemes that reduce the complexity of quantum circuits.

Keywords: quantum computing; practical quantum algorithms; quantum chemistry

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1. Introduction

The coupled-cluster (CC) theory [1–11] has assumed a preeminent role in providing a high-accuracy description of diversified classes of many-body systems [6,12–18], quantum field theory [19–23], quantum hydrodynamics [24,25], nuclear structure theory [26–28], quantum chemistry [29–36], and material sciences [37–47]. Many strengths of the single-reference CC formalism (SR-CC) originate in the exponential parametrization of the ground-state wave function and closely related linked cluster theorem [48,49].

The standard CC downfolding techniques [50–57] provide a many-body form of the effective (downfolded) Hamiltonians that can be used to calculate ground-state energies in reduced-dimensionality active spaces as long as the so-called external amplitudes defining the ground-state out-of-active-space correlation effects are known or can be effectively approximated. Although these methods have originated in the context of single-reference CC theory leading to active-space representations of non-Hermitian effective Hamiltonians, it became clear that the utilization of the double unitary CC (DUCC) Ansatz can provide Hermitian formulations for downfolded/effective active-space Hamiltonians, which thereafter have intensively been tested and validated in the context of quantum simulations based on the utilization of various quantum solvers. For example, the quantum phase estimation (QPE) [58–64] and variational quantum eigensolvers (VQE) [65–82] were invoked to obtain ground-state energies of molecular systems. These tests demonstrated that DUCC-based downfolded Hamiltonians and corresponding dimensionality reduction could accurately reproduce the electronic energies for basis sets of the sizes that are currently beyond the reach of the most advanced quantum algorithms and quantum hardware [81–85]. The

Hermitian CC downfolding procedures were also discussed and tested in the context of quantum dynamics and excited-state simulations [52,53,55]. Due to the state-specificity of the downfolding procedures, the latter attempts require detailed knowledge of the external Fermionic degrees of freedom for excited states and the construction of separate effective Hamiltonians for each excited state. Although extraction of excited-state external correlation effects is possible for some classes of excited states, which can be captured by approximate Equation-of-Motion CC (EOMCC) methods [86–88], the generalization of this formalism to general-type excited states may be numerically challenging. A part of the problem is also associated with translating the EOMCC Ansatz defined by commuting operators into a language of unitary CC expansions involving non-commuting operators.

Instead of following this strategy, in this paper, we discuss the class of Fock-space Schrieffer–Wolff (SW) transformation-inspired downfolding procedures designed to simplify the many-body form of the Hamiltonian. In analogy to the standard DUCC-based techniques, the SW-transformation-based (or rank-reducing similarity transformation (RRST)) formulations utilize the partitioning of one-electron states (spin–orbitals) into active and external spin–orbitals. Although the RRST cannot eliminate all components of the Hamiltonian that involve creation/annihilation operators carrying the external spin–orbital indices (the so-called external component of the Hamiltonian), the RRST is designed in a way that leads to a simple form that involves only local actions of qubits in corresponding quantum algorithms such as QPE. In the context of QPE methodology, this form not only makes qubit mappings simpler but can enable more efficient utilization of the Trotter formulas. In contrast to the standard DUCC downfolding, the proposed approach and related approximations eliminate its state-specific character and provide a description of multiple electronic states corresponding to the ground and excited states at least well approximated by the set of active orbitals. An exciting feature of the discussed framework is its universal character (in the sense of Fock space) in describing many-body systems with various numbers of electrons (particles), where the number of particles is specified when the action of the Hamiltonian (or the corresponding quantum evolution operator) on specific states takes place. The discussed development is primarily motivated by impressive progress in developing Fock-space generalization of CC formulations [89–93].

In analogy to all existing algorithms, the RRST formalism can be viewed as a platform for developing broad classes of approximations. In this paper, we will outline the hybrid algorithm that combines the classical-computing part associated with the determination of the RRST and the quantum-computing part, which provide a mean for modeling time evolution generated by the RRST downfolded Hamiltonian.

2. Rank-Reducing Unitary Similarity Transformations of Many-Body Hamiltonians

The dynamics of the quantum system are given by the evolution operator $\Omega(t)$

$$\Omega(t) = e^{-itH} \tag{1}$$

where we assume that the Hamiltonian H is time-independent and takes the following second-quantized form in the basis of N spin–orbitals

$$H = \sum_{p,q=1}^{N} h^p_q a^\dagger_p a_q + \frac{1}{4} \sum_{p,q,r,s=1}^{N} v^{pq}_{rs} a^\dagger_p a^\dagger_q a_s a_r \ , \tag{2}$$

where p, q, r, s are spin–orbital indices and h^p_q and v^{pq}_{rs} are one- and two-electron (antisymmetrized) integrals defining H_1 and H_2 operators, respectively. The $a^\dagger_p$ (a_p) operator corresponds to a creation (annihilation) operator for the electron in p-th spin–orbital. In quantum computing applications, especially in quantum phase estimations, various representations of the electronic Hamiltonian (induced, for example, by the unitary transformations) can be used. This is a consequence of the fact that the spectrum of the Hamiltonian remains unchanged upon these transformations. Let us denote a general unitary transformation U as

$$U = e^{A(t)} \,, \tag{3}$$

where the operator $A(t)$ is anti-Hermitian

$$A(t)^{\dagger} = -A(t) \,, \tag{4}$$

and similarity-transformed Hamiltonian $\bar{H}(t)$ is defined as

$$\bar{H}(t) = e^{-A(t)} H(t) e^{A(t)} \,. \tag{5}$$

In analogy to the H operator, the similarity-transformed Hamiltonian is also Hermitian. In consequence, probing the phase with the $\bar{H}$ and corresponding time evolution operator $\bar{\Omega}(t)$

$$\bar{\Omega}(t) = e^{-it\bar{H}(t)} = e^{-A(t)} \Omega(t) e^{A(t)} \tag{6}$$

should detect the same values of energy/phase (subject to the various choices of initial state). A typical illustration of the above techniques is the interaction and Heisenberg pictures widely used in quantum mechanics, which have recently been explored in the context of quantum computing simulations. The interaction-picture-based approach has recently been studied in the context of quantum computing [94–96].

In this paper, we will pursue a slightly different goal associated with the design of *time-independent* unitary transformation generated by the time-independent anti-Hermitian operator B ($B^{\dagger} = -B$) such that

$$H = e^{-B} G e^{B} \,, \tag{7}$$

where the properties of the G operator and the form of the B operator assuring these properties will be discussed later. For the sake of the following discussion, let us introduce the partitioning of the orbitals (spin–orbitals) into active (with corresponding first $2(n-k)$ qubits) and external (with qubits enumerated as $2(n-k)+1, \ldots, 2n$) as shown in Figure 1. Additionally, we will assume that all spin–orbitals are arranged in a way that spin-up (↑) and spin-down (↓) spin–orbitals occupying the same orbitals P (isoenergetic spin–orbitals) are neighboring as shown in the following scheme.

$$\ldots [Q \uparrow][Q \downarrow] \ldots [P \uparrow][P \downarrow] \ldots \tag{8}$$

If spin–orbitals p and q are isoenergetic we will denote it by $q = e(p)$ (or $p = e(q)$).

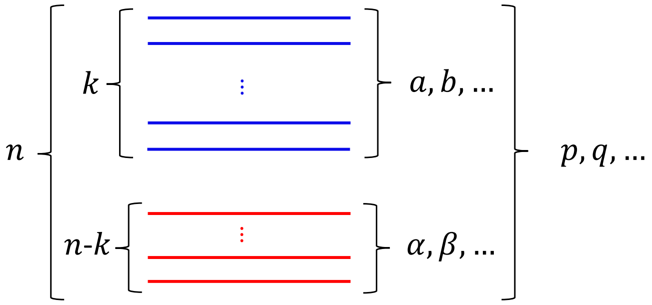


Figure 1. The orbital (spin–orbital) domain is partitioned into $n-k$ active and k external orbitals (n stands for the total number of orbitals). The active spin–orbitals are denoted as $\alpha, \beta, \ldots$, the external spin–orbitals as $a, b, \ldots$, and generic spin–orbitals as $p, q, \ldots$. In general, the active spin–orbitals do not have to be defined as the "lowest" lying spin–orbitals using some energy-related criteria. At this moment, the nature of active/external spin–orbitals remains unspecified. The total number of spin–orbitals N is defined as $N = 2n$. For simplicity, in this paper, we assume that the orbital energies are non-degenerate.

The two classes of spin–orbitals induce the partitioning of the second-quantized operators into internal and external parts that are defined by creation-annihilation operator strings (CAOSs) carrying only active spin–orbital indices (active part) and strings that contain at least one creation/annihilation operator carrying external spin–orbital index (external part), respectively. The internal and external parts of arbitrary operator X can be symbolically denoted as

$$\check{\mathcal{P}}(X) \tag{9}$$

for the internal part and

$$\widehat{\mathcal{P}}(X) \tag{10}$$

for the external part. Typical examples of CAOSs entering internal and external parts are $E^{\alpha}_{\beta} = a^{\dagger}_{\alpha} a_{\beta}$ and $E^{\alpha}_{b} = a^{\dagger}_{\alpha} a_{b}$, respectively, where the general form of the excitation operator $E^{pq\ldots}_{rs\ldots}$ is defined as $E^{pq\ldots}_{rs\ldots} = a^{\dagger}_{p} a^{\dagger}_{q} \ldots a_{s} a_{r}$ (the $E^{pq\ldots}_{rs\ldots}$ operator is antisymmetric with respect to swapping adjacent spin–orbital indices). Furthermore, it is convenient to decompose the external part into its diagonal part ($\widehat{\mathcal{P}}_{d}(X)$), iso-energetic off-diagonal ($\widehat{\mathcal{P}}_{ie}(X)$), and energetically distinct off-diagonal ($\widehat{\mathcal{P}}_{eod}(X)$) that is defined by the following classes of CAOSs (in all $E^{pq\ldots}_{rs\ldots}$ below, we assume that $p < q < \ldots$ and $r < q < \ldots$):

$$\begin{aligned}
\widehat{\mathcal{P}}_{d}(X) &\rightarrow \{E^{a}_{a}, E^{\alpha a}_{\alpha a}, E^{ab}_{ab}, \ldots\}\,, & (11)\\
\widehat{\mathcal{P}}_{ie}(X) &\rightarrow \{E^{a}_{e(a)}, E^{\alpha a}_{e(\alpha)e(a)}, E^{ab}_{e(a)e(b)}, \ldots\}\,, & (12)\\
\widehat{\mathcal{P}}_{eod}(X) &\rightarrow \{E^{a}_{b}, E^{ac}_{bd}, \ldots\} \;\; \text{(all off-diagonal CAOSs that are not isoenergetic).} & (13)
\end{aligned}$$

The isoenergetic part, $\widehat{\mathcal{P}}_{ie}(X)$, is defined by off-diagonal external excitation operators where the sums of energies corresponding to all upper spin–orbitals and energies corresponding to lower spin–orbital indices are equal according to the spin–orbital energy ordering shown in Figure 1. For example, this includes situations when each upper spin–orbital index (p) has a corresponding isoenergetic lower spin–orbital index ($e(p)$) according to the spin–orbital energy ordering shown in Figure 2. We will also define external off-diagonal part, $\widehat{\mathcal{P}}_{od}(X)$, defined as

$$\widehat{\mathcal{P}}_{od}(X) = \widehat{\mathcal{P}}_{ie}(X) + \widehat{\mathcal{P}}_{eod}(X). \tag{14}$$

For example, using these decompositions, the electronic Hamiltonian H (for simplicity, in this manuscript, we focus on the spin-independent Hamiltonians) can be decomposed as

$$H = \check{\mathcal{P}}(H) + \widehat{\mathcal{P}}(H) = \check{\mathcal{P}}(H) + \widehat{\mathcal{P}}_{d}(H) + \widehat{\mathcal{P}}_{ie}(H) + \widehat{\mathcal{P}}_{eod}(H) \tag{15}$$

where

$$\begin{aligned}
\check{\mathcal{P}}(H) &= \sum_{\alpha\beta} h^{\alpha}_{\beta} E^{\alpha}_{\beta} + \sum_{\alpha<\beta;\gamma<\delta} v^{\alpha\beta}_{\gamma\delta} E^{\alpha\beta}_{\gamma\delta} & (16)\\
\widehat{\mathcal{P}}_{d}(H) &= \sum_{a} h^{a}_{a} E^{a}_{a} + \sum_{\alpha a} v^{\alpha a}_{\alpha a} E^{\alpha a}_{\alpha a} + \sum_{a<b} v^{ab}_{ab} E^{ab}_{ab} & (17)\\
\widehat{\mathcal{P}}_{ie}(H) &= \sum_{a} h^{a}_{e(a)} E^{a}_{e(a)} + \sum_{\alpha a} v^{\alpha a}_{\alpha a} E^{\alpha a}_{e(\alpha)e(a)} + \sum_{a<b} v^{ab}_{ab} E^{ab}_{e(a)e(b)} & (18)\\
\widehat{\mathcal{P}}_{eod}(H) &= \overline{\sum_{\alpha a}} h^{\alpha}_{a} E^{\alpha}_{a} + \overline{\sum_{a\alpha}} h^{a}_{\alpha} E^{a}_{\alpha} + \overline{\sum_{ab}} h^{a}_{b} E^{a}_{b} + \overline{\sum_{\alpha b;\gamma<\delta}} v^{\alpha b}_{\gamma\delta} E^{\alpha b}_{\gamma\delta} \\
&\quad + \overline{\sum_{\alpha<\beta;\gamma d}} v^{\alpha\beta}_{\gamma d} E^{\alpha\beta}_{\gamma d} + \overline{\sum_{\alpha<\beta;c<d}} v^{\alpha\beta}_{cd} E^{\alpha\beta}_{cd} \overline{\sum_{a<b;\gamma<\delta}} v^{ab}_{\gamma\delta} E^{ab}_{\gamma\delta} + \overline{\sum_{\alpha b;\gamma d}} v^{\alpha b}_{\gamma d} E^{\alpha b}_{\gamma d} \\
&\quad + \overline{\sum_{a<b;\gamma d}} V^{ab}_{\gamma d} E^{ab}_{\gamma d} + \overline{\sum_{\alpha b;c<d}} V^{\alpha b}_{cd} E^{\alpha b}_{cd} + \overline{\sum_{a<b;c<d}} V^{ab}_{cd} E^{ab}_{cd} & (19)
\end{aligned}$$

and $\bar{\sum}$ symbol represents the summation over off-diagonal non-isoenergetic terms. Assuming that $n-k<k$, one can see that $\hat{\mathcal{P}}_{eod}(H)$ includes the largest number of terms of all components defining the decomposition (15). It is also obvious that, in general, the $\hat{\mathcal{P}}_d(X)$ of an arbitrary operator X can be expressed in terms of particle number operators, n_p,

$$n_p = a_p^\dagger a_p \;, \tag{20}$$

which can symbolically be denoted as

$$\hat{\mathcal{P}}_d(X) = f_X(\{n_p\}_{p=1}^N). \tag{21}$$

For example,

$$\hat{\mathcal{P}}_d(H) = f_H(\{n_p\}_{p=1}^N) = \sum_a h^a_a n_a + \sum_{\alpha a} v^{\alpha a}_{\alpha a} n_\alpha n_a + \sum_{ab} v^{ab}_{ab} n_a n_b \;. \tag{22}$$

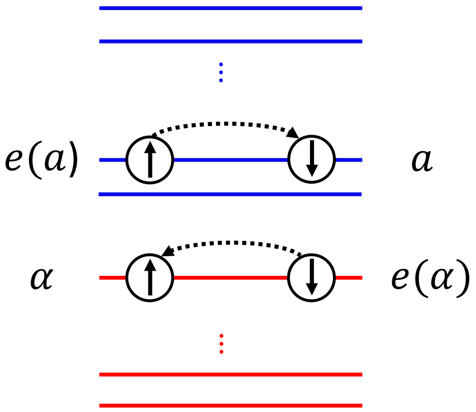


Figure 2. A typical example of the external off-diagonal isoenergetic excitation operator $E^{\alpha a}_{e(\alpha)e(a)}$.

3. Schrieffer–Wolff Rank-Reducing Similarity Transformations

In this Section, we discuss the possibility of designing unitary transformation generated by the time-independent anti-Hermitian operator B (see Equation (7)).

$$G = e^B H e^{-B} \;, \tag{23}$$

that assures specific properties of the G operator and consequently simplifies qubit mapping of the evolution operator $\Omega(t)$. In particular, we will prove the following Theorem:

Theorem 1. *There exist conditions for anti-Hermitian B-operator that render the G operator in the following form:* $G = \check{\mathcal{P}}(G) + \hat{\mathcal{P}}_d(G) + \hat{\mathcal{P}}_{ie}(G)$. *Moreover, the action of* $\hat{\mathcal{P}}_d(G) + \hat{\mathcal{P}}_{ie}(G)$ *on the qubits corresponding to external spin–orbitals (assuming the ordering of spin–orbitals and qubits in accordance with Equation (8)) is local.*

Proof of Theorem 1. To eliminate the $\hat{\mathcal{P}}_{eod}(G)$ we impose the condition for the B operator

$$\hat{\mathcal{P}}_{eod}(G) = \hat{\mathcal{P}}_{eod}(e^B H e^{-B}) = 0 \;, \tag{24}$$

which can explicitly be expanded in terms of multi-commutator expansion using Hausdorff expansion

$$\hat{\mathcal{P}}_{eod}(H) - \hat{\mathcal{P}}_{eod}([H,B]) + \frac{1}{2}\hat{\mathcal{P}}_{eod}([[H,B],B]) - \ldots = 0. \tag{25}$$

Solving the above equations in the Fock space requires some attention. One should realize that the dimensionality of Equations (25) is much bigger than the number of non-zero terms contributing to $\widehat{\mathcal{P}}_{eod}(H)$ in Equation (25) and is equal to the number of all excitation-type operators spanning $\widehat{\mathcal{P}}_{eod}$ space in Equation (13). The perturbative analysis of the solution of Equation (24) (see Appendix A) shows that at the first order of perturbation theory, the number of amplitudes defining B is precisely equal to the number of non-zero elements of $\widehat{\mathcal{P}}_{eod}(H)$. However, higher orders of many-body perturbation theory generate higher many-body components in space $\widehat{\mathcal{P}}_{eod}$. For this reason, in the general case, the B operator is expressed in terms of all operators in set (13) and satisfy the condition

$$B = \widehat{\mathcal{P}}_{eod}(B) \, , \tag{26}$$

which guarantees that the number of equations and unknowns are equal. As will be discussed later, various approximate techniques can be used to approximate the B operator using a smaller number of variables. If the solution of Equation (24), labeled as $B^{\star}$, can be found (or effectively approximated by limiting the rank of the multi-commutator expansion) on classical computers, then the G operator with the desired properties is given by

$$G = \check{\mathcal{P}}(e^{B^{\star}} H e^{-B^{\star}}) + \widehat{\mathcal{P}}_{d}(e^{B^{\star}} H e^{-B^{\star}}) + \widehat{\mathcal{P}}_{ie}(e^{B^{\star}} H e^{-B^{\star}}) \, . \tag{27}$$

The parts of G contributing to action on the qubits corresponding to external spin–orbitals are $\widehat{\mathcal{P}}_{d}(e^{B^{\star}} H e^{-B^{\star}})$ and $\widehat{\mathcal{P}}_{ie}(e^{B^{\star}} H e^{-B^{\star}})$. These two classes of elements are advantageous components of G when designing quantum circuits with low complexity. The diagonal part, according to Equation (21), obviously involves the particle number operators that are qubit-local. On the other hand, the encoding of general operators contributing to the $\widehat{\mathcal{P}}_{ie}(e^{B^{\star}} H e^{-B^{\star}})$ requires encoding a chain of $E^{a}_{e(a)}/E^{\alpha}_{e(\alpha)}$ and their Hermitian conjugates. It turns out, however, that these operators involve gates only on the pairs of adjacent qubits $[Q\uparrow][Q\downarrow]$ in the representation given by scheme (8) (assuming a non-degenerate character of orbital energies as shown in Figure 1). It can be easily inspected by using for example Jordan–Wigner (JW) qubit encoding [97].

$$\begin{aligned} a_q^{\dagger} \quad &\rightarrow \quad Q_q^{+} \otimes Z^{\rightarrow}_{q-1} \, , \qquad (28) \\ a_q \quad &\rightarrow \quad Q_q^{-} \otimes Z^{\rightarrow}_{q-1} \, , \qquad (29) \end{aligned}$$

where

$$\begin{aligned} Q_q^{+} \quad &= \quad \frac{1}{2}(\sigma_q^x - i\sigma_q^y) \, , \qquad (30) \\ Q_q^{-} \quad &= \quad \frac{1}{2}(\sigma_q^x + i\sigma_q^y) \, , \qquad (31) \\ Z^{\rightarrow}_{q-1} \quad &= \quad \sigma^z_{q-1} \otimes \ldots \otimes \sigma^z_1 \, , \qquad (32) \end{aligned}$$

and σ_q^x, σ_q^y, and σ_q^z represent Pauli gates on q-th qubit, that the general $a^{\dagger}_{Q\uparrow} a_{Q\downarrow}$ can be expressed as local two-qubit action

$$I^{\leftarrow}_{Q\uparrow+1} \otimes Q^{+}_{Q\uparrow} \otimes Q^{-}_{Q\downarrow} \otimes I^{\rightarrow}_{Q\downarrow-1} \, . \tag{33}$$

where $I^{\leftarrow}_{Q\uparrow+1}$ and $I^{\rightarrow}_{Q\downarrow-1}$ symbolically represent tensor products of the unit operator to the left and to the right of the $Q\uparrow$ and $Q\downarrow$ qubits, respectively. □

An interesting consequence of Equation (24) is the fact that the solution $B^{\star}$ cannot commute with the Hamiltonian H. If it was the case, i.e.,

$$[B^{\star}, H] = [e^{B^{\star}}, H] = 0 \tag{34}$$

then the corresponding equation (Equation (24)), which becomes

$$\widehat{\mathcal{P}}_{eod}(H) = 0 \,, \tag{35}$$

has no solutions (where we assumed the non-trivial case of a Hamiltonian $\widehat{\mathcal{P}}_{eod}(H) \neq 0$).

The unitary transformation generated by the B operator was used to eliminate the most non-local, in the sense of qubit utilization, $\widehat{\mathcal{P}}_{eod}(G)$ part of the G operator. In a similar fashion, one can also eliminate the $\widehat{\mathcal{P}}_{ie}(G)$ contribution to the external excitations. However, an added layer of complexity is associated with the numerical nature of the problem that must be considered. For example, certain classes of solvers, like many-body perturbations theory, may stumble into numerical problems associated with the vanishing denominators for isoenergetic components of B. These problems can be remedied (in addition to using zeroth order Hamiltonians that break symmetries of the system) by using additional unitary "gauge" transformation and breaking the energetic symmetry of the external isoenergetic off-diagonal terms (*vide infra*). If the numerical issues can be effectively handled, then the "domain" of the B operator can be extended to the external isoenergetic off-diagonal excitations, and the following Corollary holds:

Corollary 1. *If equations can be solved for the B operator with the solution $B^\star$ in the extended excitations domain involving isoenergetic external off-diagonal excitations, i.e.,*

$$\widehat{\mathcal{P}}_{ie}(e^{B}He^{-B}) + \widehat{\mathcal{P}}_{eod}(e^{B}He^{-B}) = \widehat{\mathcal{P}}_{od}(e^{B}He^{-B}) = 0 \tag{36}$$

then the operator G takes the form

$$G = \check{\mathcal{P}}(e^{B^\star}He^{-B^\star}) + \widehat{\mathcal{P}}_{d}(e^{B^\star}He^{-B^\star}) \,, \tag{37}$$

where are $\widehat{\mathcal{P}}_{d}(e^{B^\star}He^{-B^\star})$ is expressed solely in terms of the particle number operators, i.e., $\widehat{\mathcal{P}}_{d}(e^{B^\star}He^{-B^\star}) = f_G(\{n_p\}_{p=1}^{N})$.

To date, we have been discussing the application of the single unitary transformation generated by the unitary operator e^B (or e^{-B} in the context of Equation (23)). However, there exists flexibility in choosing the form of the unitary transformation. For example, the product of two unitary transformations, e.g.,

$$U = e^{B}e^{C} \,, \tag{38}$$

where $C^\dagger = -C$, can transform the H operator to the Γ operator in an analogous way in Equation (23),

$$\Gamma = e^{B}e^{C}He^{-C}e^{-B} \tag{39}$$

with the same spectral properties as the original Hamiltonian H. However, the purpose of the additional transformation generated by the anti-Hermitian operator C is to produce the form of the auxiliary Hamiltonian $\bar{H}_C$,

$$\bar{H}_C = e^{C}He^{-C} \,, \tag{40}$$

that eases the process of solving analogs of Equation (24)

$$\widehat{\mathcal{P}}_{eod}(e^{B}\bar{H}_C e^{-B}) = 0 \,, \tag{41}$$

or Equation (36)

$$\widehat{\mathcal{P}}_{ie}(e^{B}\bar{H}_C e^{-B}) + \widehat{\mathcal{P}}_{eod}(e^{B}\bar{H}_C e^{-B}) = \widehat{\mathcal{P}}_{od}(e^{B}\bar{H}_C e^{-B}) = 0 \,. \tag{42}$$

For this reason, we can view the e^C operator as an auxiliary transformation of the Hamiltonian H. The idea behind using the auxiliary transformation is to employ as robust (or even postulated) form of the C operator as possible. In particular, the C operator can include different types of excitation operators than the B operator. For example, it can contain many-body effects in the active space only. The utilization of the auxiliary transformation offers us flexibility in exploring various scenarios, including the possibility of spatial/spin symmetry breaking in the simulations of molecular systems, without altering the spectral properties of the original Hamiltonian H.

4. QPE Formulations Based on the SW-RRST Representation of Many-Body Hamiltonians

The main idea of the QPE is in the controlled execution of the powers of the $\Omega(t)$ operator according to the progression

$$\Omega(t)^{2^0} \rightarrow \Omega(t)^{2^1} \rightarrow \ldots \Omega(t)^{2^j} \ldots\ldots \Omega(t)^{2^{m-1}} \tag{43}$$

where m designates the number of ancilla qubits used to read the phase(s) of the unitary evolution operator (see Figure 3a). When a standard representation of the Hamiltonian H is used, the qubit encoding of the 2^j-th power of $\Omega(t)$ operator,

$$\Omega(t)^{2^j} = (e^{-itH})^{2^j} , \tag{44}$$

utilizes all N "physical" qubits. When representation (7) is invoked, the same operator power can be expressed as

$$\Omega(t)^{2^j} = (e^{-B} e^{-itG} e^{B})^{2j} = e^{-B} (e^{-itG})^{2^j} e^{B} , \tag{45}$$

where B and G operators are given by Equations (7) or (23). The main difference between (44) and (45) (see Figure 3 instes (b) and (c)) is that the sequence $(e^{-itG})^{2^j}$ can be executed using localized qubit gates in the sense of earlier discussion of G-operator properties. The additional advantages stem from the fact that the external part of the G operator $\widehat{\mathcal{P}}(G)$ involves a simpler form of the gates and large classes of operators that commute with the internal part of the G operator, $\breve{\mathcal{P}}(G)$, which simplifies the form of the Trotter formula. We will explain it on the example of the G operator discussed in Corollary 1, where G is decomposed into internal $\breve{\mathcal{P}}(G)$ and external $\widehat{\mathcal{P}}(G)$ parts, and where the external part is expressed in terms of number operators $\widehat{\mathcal{P}}(G) = f_G(\{n_p\}_{p=1}^N)$ since $\widehat{\mathcal{P}}(G)$ only contains diagonal elements as a result of Corollary 1. Let us further decompose $\widehat{\mathcal{P}}(G)$ into the part that mixes particle number operators for active and external spin–orbitals ($\widehat{\mathcal{P}}_M(G)$) and the part that is solely expressed in terms of the particle number operator corresponding to external spin–orbitals only ($\widehat{\mathcal{P}}_E(G)$). Therefore the following commutation relations holds

$$[\breve{\mathcal{P}}(G), \widehat{\mathcal{P}}_E(G)] = [\widehat{\mathcal{P}}_M(G), \widehat{\mathcal{P}}_E(G)] = 0 , \tag{46}$$

therefore

$$\begin{aligned} e^{-itG} &= e^{-it[\breve{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]} e^{-it\widehat{\mathcal{P}}_E(G)} \\ &= e^{-it\widehat{\mathcal{P}}_E(G)} e^{-it[\breve{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]} , \end{aligned} \tag{47}$$

where the $e^{-it[\breve{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]}$ term requires a Trotter formula to be implemented. At the same time the $e^{-it\widehat{\mathcal{P}}_E(G)}$ terms as depending only on the particle number operators (or their products) can be calculated exactly, which is a consequence of the fact that all n_p's operators defining $\widehat{\mathcal{P}}_E(G)$ commute. For example, the JW qubit mapping of n_p and $n_p n_q$ can be represented by a simple local circuits

$$n_p \quad \rightarrow \quad \frac{1}{2}(1_p - \sigma_p^z)\,, \tag{48}$$

$$n_p n_q \quad \rightarrow \quad \frac{1}{4}(1_p - \sigma_p^z) \otimes (1_q - \sigma_q^z)\,, \tag{49}$$

where 1_p and σ_p^z are identity and Pauli Z matrices (gates) on p-th qubit. We also used a simplified notation, where

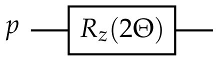
$$(1_p - \sigma_p^z) \otimes (1_q - \sigma_q^z) \equiv \ldots \otimes 1_i \otimes \ldots \otimes (1_p - \sigma_p^z) \otimes \ldots \otimes 1_k \otimes \ldots \otimes (1_q - \sigma_q^z) \otimes \ldots \otimes 1_l \otimes \ldots\,. \tag{50}$$

The corresponding qubit representations of

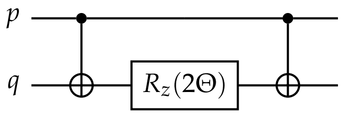
$$e^{-it\alpha_p n_p} \quad \rightarrow \quad e^{-i\frac{t\alpha_p}{2}} e^{i\frac{t\alpha_p}{2}\sigma_p^z}\,, \tag{51}$$

$$e^{-it\alpha_{pq} n_p n_q} \quad \rightarrow \quad e^{-i\frac{t\alpha_{pq}}{4}(1_p - \sigma_p^z)\otimes(1_q - \sigma_q^z)}\,, \tag{52}$$

where α_p and α_{pq} are scalars, require two types of circuits (shown below, respectively) that encode general $e^{-i\Theta\sigma_p^z}$ and $e^{-i\Theta\sigma_p^z \otimes \sigma_q^z}$ operators



and



which demonstrates the locality of the qubit encoding of the $e^{-it\widehat{\mathcal{P}}_E(G)}$ operator (or its approximate form defined by a truncated form of the $\widehat{\mathcal{P}}_E(G)$ operator).

Further analysis of qubit encoding of (45) requires explicit expansion of $\breve{\mathcal{P}}(G)$ and $\widehat{\mathcal{P}}_M(G)$ in terms of Pauli strings P_i, i.e.,

$$\breve{\mathcal{P}}(G) \quad = \quad \sum_i \breve{g}_i P_i\,, \tag{53}$$

$$\widehat{\mathcal{P}}_M(G) \quad = \quad \sum_j \hat{h}_j P_j\,. \tag{54}$$

where $\breve{g}_i$ and $\hat{h}_j$ are scalars. There is a simple way how Trotter formula can be utilized to expand $e^{-it[\breve{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]}$:

$$e^{-it[\breve{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]} \simeq (X(r)Y(r))^r\,, \tag{55}$$

where

$$X(r) \quad = \quad \prod_i e^{-i\frac{t\breve{g}_i}{r}P_i}\,. \tag{56}$$

$$Y(r) \quad = \quad \prod_j e^{-i\frac{t\hat{h}_j}{r}P_j}\,, \tag{57}$$

According to the definition of $\widehat{\mathcal{P}}_M(G)$ all Pauli strings in expansion (54) are defined through the Z-gates, hence

$$Y(r) = e^{-i\frac{t}{r}\sum_j \hat{h}_j P_j}\,. \tag{58}$$

Now, let us assume that r is even and notice that for sufficiently large r, the ordering of the factors $X(r)$ and $Y(r)$ in (55) can be arbitrary, i.e., it can be rewritten as

$$
\begin{aligned}
e^{-it[\check{\mathcal{P}}(G)+\widehat{\mathcal{P}}_M(G)]} &\simeq (X(r)Y(r)Y(r)X(r))^{\frac{r}{2}} &(59)\\
&= (X(r)e^{-i\frac{2t}{r}\sum_j \hat{h}_j P_j}X(r))^{\frac{r}{2}} \,. &(60)
\end{aligned}
$$

Combining (45) and (47) with (60) leads to the further simplifications of the $\Omega(t)^{2^j}$:

$$\Omega(t)^{2^j} \simeq e^{-B}[(X(r)e^{-i\frac{2t}{r}\sum_j \hat{h}_j P_j}X(r))^{\frac{r}{2}}]^{2^j} e^{-it2^j\widehat{\mathcal{P}}_E(G)} e^{B} \,. \quad (61)$$

for sufficiently large values of r parameter (see Figure 3 insets (c) and (d)).

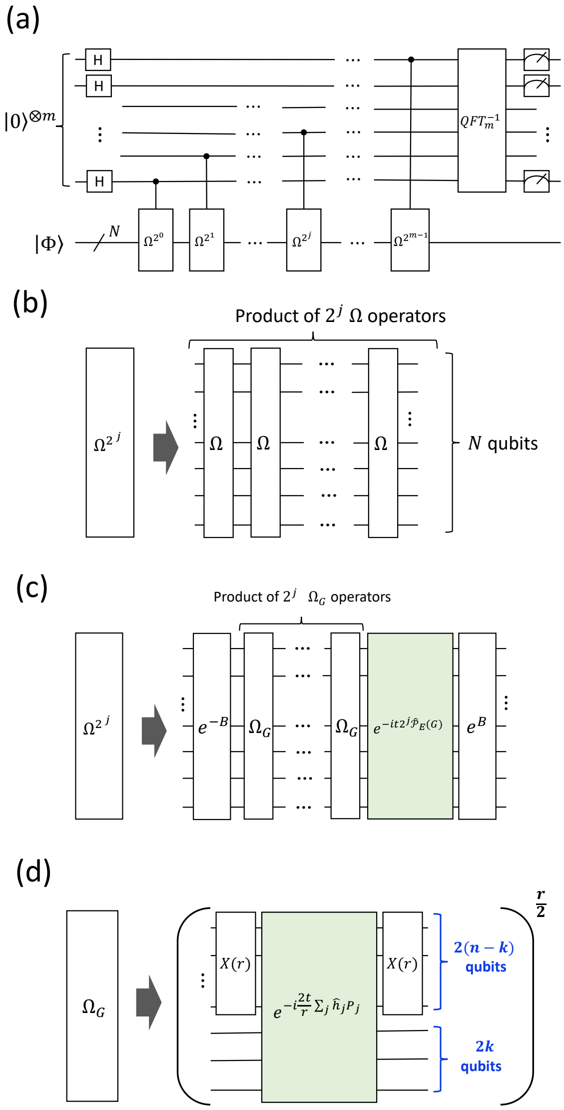


Figure 3. Schematic representations of the QPE algorithm (inset (**a**)) and algorithm for the evaluation of the $2j$-th power of the $\Omega(t)$ operator (insets (**b**,**c**)) in the representation involving G operator given by Equation (7). See the text for descriptions of each diagram.

If the operator B can be effectively calculated/approximated, the above formula offers several interesting properties:

- all terms depending on the $\widehat{\mathcal{P}}_M(G)$ and $\widehat{\mathcal{P}}_E(G)$ can be evaluated exactly,

- the $\breve{\mathcal{P}}(G)$ is $2(n-k)$-local (in the sense of Ref. [98]), i.e., is defined interactions involving at most $2(n-k)$ qubits (assuming qubit ordering defined in scheme (8)),
- for $(n-k) \ll k$, if the operator B can be approximated by single and double excitations (approach consistent with the first order of perturbation theory), then the number of terms that need to be included in the B operator is proportional to $(n-k) \times k^3$ (smaller number of terms compared to the original Hamiltonian H, which is proportional to n^4). Additionally, there are only two instances when the e^{-B}/e^{B} transformations have to be performed to encode $\Omega(t)^{2^j}$ (see, Equation (45)).
- expansion (61) is reminiscent of the QAOA (Quantum Approximate Optimization Algorithm) Ansatz [99–103] consisting of alternating sequence of cost ($\widehat{\mathcal{P}}_M(G)$-dependent part) and mixer ($\breve{\mathcal{P}}(G)$-dependent part) layers.

Furthermore, in analogy to the Fock-space coupled-cluster formulations (for example, the valence-universal (VU-CC) theories), the B and G operators along with the corresponding time evolution operator $\Omega(t)$ can act on the states $|\Theta(n_e)\rangle$ corresponding to various number of electron (n_e), i.e.,

$$\Omega(t)^{2^j}|\Theta(n_e)\rangle \,, \tag{62}$$

providing access to energies of neutral, ionized, doubly-ionized, etc., electronic states having non-zero overlap with $|\Theta(n_e)\rangle$. A similar algorithm can be used to evaluate phase using representation (39) based on the auxiliary unitary transformation.

5. Approximations

The potential of reducing hardware requirements discussed in the previous Section is inextricably connected to our ability to approximate solutions of non-terminating expansions (15) using their finite-rank commutator expansion (16). We envision this step to be entirely performed using conventional computers. This step is justified because the process of solving equations for finite-rank commutator expansions is associated with polynomial scaling.

In analogy to the existing quantum algorithms, including a broad class of VQE and QPE formulations, classes of approximations are indispensable to translate the problem into the form of circuits for quantum computers or their classical emulators. For example, the broad class of Trotter-based approximations is needed for realizing unitary CC-driven variants of the VQE formalism and evolution operators in the QPE formalism. It does not come as a surprise that for the SW-RRST approximation, the situation is no different. The main factors that need to be taken into account when defining approximate SW-RRST formulations are as follows:

1. **The excitation-rank of the many-body form of the transformed G Hamiltonian.** Our experience with Hilbert-space-type downfolding indicates that one- and two-body effective interactions can provide satisfactory results when active space is adequately defined [56].
2. **The rank of the many-body effects in the B operator.** The elementary perturbative analysis for the case discussed in Corollary 1 indicates that the 0-th order of B is equal to zero, the 1-st order contributed to one- and two-body terms, while the 2-nd order introduces higher-rank effects (see the analysis in Appendix A). It suggests that simple models based on the inclusion of one- and two-body effects in the B operator are justified.
3. **The working equations for B-amplitudes.** Due to their non-terminating nature, the algebraic form of Equations (24) or (36) has to be approximated due to their non-terminating nature. A numerically feasible way of introducing sufficiency conditions for B-operator amplitudes is to use finite commutator-rank expansion in

Equations (24) or (36). The class of approximations termed SW-RRST(l) consist of retaining commutators up to the l-th rank. For formulation (24) we have

$$\widehat{\mathcal{P}}_{eod}(H+\sum_{i=1}^{l}(-1)^i\frac{1}{i!}[\ldots[H,B],\ldots],B]_i)=0. \tag{63}$$

whereas for (36) one gets

$$\widehat{\mathcal{P}}_{od}(H+\sum_{i=1}^{l}(-1)^i\frac{1}{i!}[\ldots[H,B],\ldots],B]_i)=0. \tag{64}$$

While the linear SW-RRST(1) is the simplest approximation consistent with the low-order perturbative analysis, one may have to deal with the singular or nearly singular forms of the equation. Similar problems have been observed in the early studies of the CC theory [104]. Several techniques akin to almost-linear CC approximations are discussed in Refs. [105–107], including the auxiliary transformation discussed earlier, have the potential to offset singular behavior. Another factor that was shown to play a key role in the removal of singularities of linear CC equation is associated with the inclusion of non-linear terms, which is the main motivation for the development of higher-rank SW-RRST(l) approximations ($l > 1$).

Another important aspect of the proposed approximation is the choice of the orbital basis that should be driven by the targeted quantum system.

6. Conclusions

In summary, we have discussed the extension of the traditional downfolding methods to the Fock-space formulation using the Schrieffer–Wolff-type transformations. We have presented the basic properties of this formalism that leads to a simplified form of the similarity-transformed Hamiltonian G that, in the context of the quantum circuit complexity, is dominated by its $2(n-k)$-local internal part. The remaining external part of the G operator, depending on the specific form of the approach, either given by conditions (24) or (36), is defined by simple operators (for example, particle number operators) that can be determined exactly. An additional advantage of the SW-RRST approach is that the external components of the G operator can be factored out and do not need to be handled by the Trotter expansion. While we have discussed the general features of the SW-RRST formalism and its approximation, in the following papers, we will analyze the numerical solutions to the SW-RRST(l) equations and the effect of non-linear terms on the amplitudes defining the anti-Hermitian B operator. An essential aspect of the numerical analysis is the determination of the feasibility of auxiliary similarity transformation that breaks symmetries of the targeted quantum system.

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Abbreviations

The following abbreviations are used in this manuscript:

ASET	Second-order Active-space Embedding
CAOS	creation-annihilation operator strings
CC	Coupled-Cluster
CCT	Canonical Transformation Theory
DUCC	Double Unitary CC
EOMCC	Equation-of-Motion CC
JW	Jordan–Wigner
NISQ	Noisy Intermediate-scale Quantum
QAOA	Quantum Approximate Optimization Algorithm
QPE	Quantum Phase Estimation
RRST	Rank-Reducing Similarity Transformation
RRST(*l*)	RRST approximation of *l*-th rank
SW	Schrieffer–Wolff
VQE	Variatonal Quantum Eigensolver
VU-CC	Valence-Universal CC

Appendix A. Perturbative Estimates of the *B* Operator

Our perturbative analysis of the B operator will utilize the following partitioning of the Hamiltonian (2) into 0-th order part H_0 (assumed to be diagonal) and perturbation W

$$H(\lambda) = H_0 + \lambda W \tag{A1}$$

where H_0 and W are generally defined as

$$H_0 = \sum_{p,q} (h^p_q - v^p_q) a^\dagger_p a_q = \sum_p \epsilon_p a^\dagger_p a_p \tag{A2}$$

and

$$\begin{aligned} W &= V_1 + V_2 \,, && \text{(A3)} \\ V_1 &= \sum_{p,q} v^p_q a^\dagger_p a_q \,, && \text{(A4)} \\ V_2 &= \frac{1}{4} \sum_{p,q,r,s} v^{pq}_{rs} a^\dagger_p a^\dagger_q a_s a_r \,. && \text{(A5)} \end{aligned}$$

At this point, we assume the diagonal form of the H_0 operator without specifying the form of the V_1 operator, which can generally be a spatial/spin symmetry-breaking operator. In particular, we are not assuming that ϵ_p's are Hartree-Fock spin–orbital energies, which would be challenging in situations when Hartree-Fock external orbitals are degenerate. These problems can be addressed by the proper definition of H_0 (or by the properly designed gauge transformation mentioned earlier).

For simplicity, we assume that the perturbative expansion for the B operator is convergent and takes the form

$$B = \sum_{i=0}^{\infty} \lambda^i B^{(i)}\,, \tag{A6}$$

where i refers to the order of perturbative expansion. Introducing (A6) into sufficiency conditions (24) (similar analysis is valid for the variant described in Corollary 1 or for the Hamiltonian $\bar{H}_C$ (40))

$$\widehat{\mathcal{P}}_{eod}(e^{\sum_{i=0}\lambda^i B^{(i)}}(H_0 + \lambda W)e^{-\sum_{j=0}\lambda^j B^{(j)}}) = 0\,, \tag{A7}$$

We get, using BCH expansion, equations for perturbative components of the B operator. For example,

- **0-th order:**

  $$\widehat{\mathcal{P}}_{eod}([H_0, B^{(0)}]) = 0\,, \tag{A8}$$

  where we utilized the fact that H_0 is a diagonal operator ($\widehat{\mathcal{P}}_{eod}(H_0) = 0$), which leads to $B^{(0)} = 0$.
- **1-st order:**

  $$\widehat{\mathcal{P}}_{eod}(W - [H_0, B^{(1)}]) = 0\,, \tag{A9}$$

  which yields one- and two-body components only.
- **2-nd order:**

  $$\widehat{\mathcal{P}}_{eod}(-[H_0, B^{(2)}] + \frac{1}{2}[[H_0, B^{(1)}], B^{(1)}] - [W, B^{(1)}]) = 0\,, \tag{A10}$$

  which generates lowest-order three-body interactions (stemming from the $[W, B^{(1)}]$ term).

As a consequence of the non-linear character of the expansion (A7), higher orders of perturbation theory generate higher-rank many-body components. It is interesting to notice that the rank of excitation vs. its perturbation order is essentially the same as in the standard SR-CC theory.

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