Coupled Cluster Downfolding Theory: towards efficient many-body algorithms for dimensionality reduction of composite quantum systems

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The recently introduced coupled cluster (CC) downfolding techniques for reducing the dimensionality of quantum many-body problems recast the CC formalism in the form of the renormalization procedure allowing, for the construction of effective (or downfolded) Hamiltonians in small-dimensionality sub-space, usually identified with the so-called active space, of the entire Hilbert space. The resulting downfolded Hamiltonians integrate out the external (out-of-active-space) Fermionic degrees of freedom from the internal (in-the-active-space) parameters of the wave function, which can be determined as components of the eigenvectors of the downfolded Hamiltonians in the active space. This paper will discuss the extension of non-Hermitian (associated with the unitary CC formulations) and Hermitian (associated with the unitary CC approaches) downfolding formulations to composite quantum systems. The non-Hermitian formulation can provide a platform for developing local CC approaches, while the Hermitian one can serve as an ideal foundation for developing various quantum computing applications based on the limited quantum resources. We also discuss the algorithm for extracting the semi-analytical form of the inter-electron interactions in the active spaces.

I. INTRODUCTION

The coupled cluster (CC) methodology\textsuperscript{1–9} is a driving engine of high-precision simulations in physics, chemistry, and material sciences. Several properties of CC made it especially efficient in capturing correlation effects in many-body quantum systems ranging from quantum field theory,\textsuperscript{10–14} quantum hydrodynamics,\textsuperscript{15,16} and nuclear structure theory\textsuperscript{17–19} to quantum chemistry\textsuperscript{20–27} and material sciences.\textsuperscript{28–35} In this article, we will mainly focus our attention on the application to quantum chemistry. Many appealing features of the single-reference (SR) CC formalism (which will be the main focus of the present discussion) in applications to chemical systems originate in the exponential parametrization of the ground-state wave function and closely related linked cluster theorem.\textsuperscript{36–38} The last feature assures the so-called additive separability of the calculated energies in the non-interacting sub-system limit, which plays a critical role in the proper description of various chemical transformations such as chemical reactions that include bond breaking and bond-forming processes. The linked cluster theorem also plays a crucial role in designing formalisms that can provide the chemical accuracy needed for predicting spectroscopic data, reaction rates, and thermochemistry data. The best-known example of such class of methods is the CCSD(T) formalism,\textsuperscript{39} which combines the iterative character of the CCSD formalism\textsuperscript{3} (CC with single and double excitations) with perturbative techniques for determining CC energy corrections due to connected triple excitations. Over the last few decades, the CCSD(T)-type formulations have been refined to provide accurate description of bond-breaking processes. Among several formulations that made it possible were the method of moments of coupled cluster equations and renormalized approaches\textsuperscript{40–49} perturbative formulations based on the A-operator (defining the left eigenvectors of the similarity-transformed Hamiltonians)\textsuperscript{50–58} and other techniques.\textsuperscript{59–62}

One should also mention the tremendous effort in formulating reduced-scaling or local formulations of the CC methods to extend the applicability of the CC formalism across spatial scales\textsuperscript{63–75} In several cases, the extension of local formulations was possible for linear response CC theory\textsuperscript{76} and excited state CC formulations based on the equation-of-motion formalism.\textsuperscript{77,78}

Recently, interesting aspects of SR-CC were discussed using the sub-system embedding sub-algebras (SES) approach,\textsuperscript{79,80} where we demonstrated that the CC energy can be calculated in an alternative way to the standard CC energy formula. Instead of using standard energy expression, one can obtain the same energy by diagonalizing the downfolded/effective Hamiltonian in method-specific active space(s) generated by appropriate sub-system embedding sub-algebras. The SR-CC theory provides a rigorous algorithm for how to construct these Hamiltonians using the external, with respect to the active space, class of cluster amplitudes.\textsuperscript{79} Shortly after this discovery, these results for static SR-CC formulations were extended to the time domain.\textsuperscript{81} Following similar concepts as in the static case and assuming that the external time-dependent cluster amplitudes are known or can be effectively approximated, it was shown that the quantum evolution of the entire system can be generated in the active space by time-dependent downfolded Hamiltonian. Another interesting aspect of CC SES downfolding is the possibility of integrating several SES CC eigenvalue problems corresponding to various active spaces into a computational flow or quantum flow as discussed in Ref.\textsuperscript{80}, where we demonstrated that the flow equations are fully equivalent to the standard approximations given by cluster operators defined by unique internal excitations involved in the active-space problems defining the flow. This feature provides a natural language for expressing the sparsity of the system. In contrast to other local CC approaches, the CC quantum flow equations can effectively embrace the concept of localized orbital pairs at the level of effective Hamiltonian acting in the appropriate
active space. The SES CC downfolded Hamiltonians are non-Hermitian operators, which limits their utilization in quantum computing. Instead, using double unitary CC (DUC) Ansatz, one can derive the active-space many-body form of Hermitian downfolded Hamiltonians. In contrast to the SR-CC, the DUC-based effective Hamiltonians are expressed in terms of non-terminating expansions involving anti-Hermitian cluster operators defined by external type excitations/de-exitations. Several approximate forms of DUC Hamiltonians have been tested in the context of quantum simulations, showing the potential of DUC downfolding in reproducing exact ground-state energy in small active spaces. In particular, the downfolded Hamiltonians have been integrated with various quantum solvers, including Variational Quantum Eigensolvers (VQE) and Quantum Phase Estimation (QPE), to calculate ground-state potential energy surfaces corresponding to breaking a single chemical bond.

In this paper, we will briefly review the current status of the downfolding methods and provide further extension of the CC downfolding methods to multi-component systems. As a specific example, we choose a composite system defined by Fermions of type A and Fermions of type B. This is a typical situation encountered for certain classes of non-Born-Opennheimer dynamics and nuclear structure theory. The discussed formalism can be easily extended to other types of systems composed of Fermions and Bosons as encountered in the descriptions of polaritonic systems. We believe that these formulations will pave the way for more realistic quantum simulations of multi-component systems.

II. CC THEORY

The SR-CC theory utilizes the exponential representation of the ground-state wave function |Ψ⟩,

$$|Ψ⟩ = e^{T} |Φ⟩ ,$$

where T and |Φ⟩ represent the so-called cluster operator and single-determinant reference function. The cluster operator can be represented through its many-body components $T_k$

$$T = \sum_{k=1}^{M} T_k ,$$

where individual component $T_k$ takes the form

$$T_k = \frac{1}{(k!)^2} \sum_{l_1,...,l_k} e^{l_1,...,l_k} E^{l_1,...,l_k} ,$$

where indices $l_1, l_2, ..., (a_1, a_2, ...) \in \mathbb{N}$ refer to occupied (unoccupied) spin orbitals in the reference function |Φ⟩. The excitation operators $E^{l_1,...,l_k}$ are defined through strings of standard creation ($a_p$) and annihilation ($a_p^\dagger$) operators

$$E^{l_1,...,l_k} = a_{l_1}^\dagger a_{l_2}^\dagger \cdots a_{l_k}^\dagger a_{l_1} \cdots a_{l_k} ,$$

where creation and annihilation operators satisfy the following anti-commutation rules:

$$[a_p, a_q^\dagger]_+ = [a_p^\dagger, a_q]_+ = 0 ,$$

When $M$ in the summation in Eq. (2) is equal to the number of correlated electron ($N_e$) then the corresponding CC formalism is equivalent to the FCI method, otherwise for $M < N_e$ one deals with the standard approximation schemes. Typical CC formulations such as CCSD, CCSDT, and CCSDTQ correspond to $M = 2, M = 3$, and $M = 4$ cases, respectively.

The equations for cluster amplitudes $t_{l_1,...,l_k}$ and ground-state energy $E$ can be obtained by introducing Ansatz (1) into the Schrödinger equation and projecting onto $P + Q$ space, where $P$ and $Q$ are the projection operator onto the reference function and the space of excited Slater determinants obtained by acting with the cluster operator onto the reference function $|Φ⟩$, i.e.,

$$\langle P + Q | H e^{T} | Φ⟩ = E \langle P + Q | e^{T} | Φ⟩ ,$$

where $H$ represents the electronic Hamiltonian. The above equation is the so-called energy-dependent form of the CC equations, which corresponds to the eigenvalue problem only in the exact wave function limit when $T$ contains all possible excitations. Approximate CC formulations do not represent the eigenvalue problem. At the solution, the energy-dependent CC equations are equivalent to the energy-independent or connected form of the CC equations:

$$Q e^{-T} H e^{T} | Φ⟩ = 0 ,$$

$$⟨Φ| e^{-T} H e^{T} | Φ⟩ = E .$$

Using Baker-Campbell-Hausdorff (BCH) formula and Wick’s theorem one can show that only connected diagrams contribute to Eqs. (8) and (9). For notational convenience, one often uses the similarity transformed Hamiltonian $\tilde{H}$, defined as

$$\tilde{H} = e^{-T} H e^{T} .$$

III. NON-HERMITIAN CC DOWNFOLDING

The main idea of SR-CC non-Hermitian downfolding hinges upon the characterization of sub-systems of a quantum system of interest in terms of active spaces or commutative sub-algebras of excitations that define corresponding active space. This is achieved by introducing sub-algebras of algebra $g^{N}$ generated by $E_{ij} = a_{i}^\dagger a_{j}$ operators in the particle-hole representation defined with respect to the reference $|Φ⟩$. As a consequence of using the particle-hole formalism, all generators commute, i.e., $[E_{ij}, E_{kl}^*] = 0$, and algebra $g^{N}$ (along with all sub-algebras considered here) is commutative. The CC SES approach utilizes class of sub-algebras of commutative $g^{N}$ algebra, which contain all possible excitations $E^{l_1,...,l_k}$ needed to generate all possible excitations from a subset of active occupied orbitals (denoted as $R$, $\{ R_i, i = 1, ..., x_R \}$) to a subset of active virtual orbitals (denoted as $S$, $\{ S_i, i = 1, ..., y_S \}$) defining active space. These sub-algebras will be designated as $g^{N}(R,S)$. Sometimes it is convenient to use alternative notation $g^{N}(x_R, y_S)$ where numbers of active orbitals in
when a localized orbital basis set is used, this procedure we call where the CC equations given by Eqs. (8) and (9) can be replaced by quantum flow equations composed of non-Hermitian operators. This is can be reproduced when one uses trivial sub-algebra, which contains no excitations (i.e., active space contains \(|\Phi\rangle\) only).

The existence of alternative ways of calculating CC energy opens alternative ways of constructing new classes of approximations. For example, if one integrates several eigenvalues problems corresponding to SESs \(h_i\), \((i = 1, \ldots, M)\) into a quantum flow equations (QFE) discussed in Ref. 80, i.e.,

\[
H^{\text{eff}}(h_i) e^{T_{\text{int}}(h_i)} |\Phi\rangle = E e^{T_{\text{int}}(h_i)} |\Phi\rangle \quad (i = 1, \ldots, M) .
\] (15)

In Ref. 79 we demonstrated that at the solution, the solution of the QFE is equivalent to the solution of standard CC equations in the form of Eqs. (8) and (9) defined by cluster operator \(T\) which is a combination of all unique excitations included in \(T_{\text{int}}(h_i)\) \((i = 1, \ldots, M)\) operators. This is can be symbolically expressed as

\[
T = \bigcup_{i=1}^{M} T_{\text{int}}(h_i) .
\] (16)

These two equivalent representations allows one also to form the following important corollary:

**Corollary (or the equivalence theorem)** For certain forms of cluster operator \(T\), the standard connected form of the CC equations given by Eqs. (8) and (9) can be replaced by quantum flow equations composed of non-Hermitian eigenvalue problems, Eq. (15).

The above corollary plays an important role in defining reduced-scaling formulations. This is a consequence of the fact that each sub-problem corresponding to sub-algebra \(h_i\) has the associated form of the effective Hamiltonian \(H^{\text{eff}}(h_i)\), which allows to define one body-density matrix for the sub-system and select the sub-set of the most important cluster amplitudes in the \(T_{\text{int}}(h_i)\) operator. For example, when a localized orbital basis set is used, this procedure can be used to define the so-called orbital pairs at the level of the effective Hamiltonian, which is a significant advantage compared to the existing local CC approaches. This procedure can also be extended to other systems driven by different types of interactions such as in nuclear structure theory or quantum lattice models, where the extension of the standard local CC formulations as used in quantum chemistry is not obvious.

**IV. HERMITIAN CC DOWNFOLDING**

In order to employ downfolding methods in quantum simulations, one has to find a way to construct Hermitian effective Hamiltonians. This goal can be achieved by employing the double unitary coupled Ansatz (DUCC)\(^\ddagger\) where the ground-state wave function is represented as

\[
|\Psi\rangle = e^{\sigma_{\text{ext}}(h)} e^{\sigma_{\text{int}}(h)} |\Phi\rangle ,
\] (17)

where \(\sigma_{\text{ext}}(h)\) and \(\sigma_{\text{int}}(h)\) are general-type anti-Hermitian operators

\[
\sigma_{\text{int}}^\dagger(h) = -\sigma_{\text{int}}(h) ,
\] (18)

\[
\sigma_{\text{ext}}^\dagger(h) = -\sigma_{\text{ext}}(h) .
\] (19)
In analogy to the SR-CC case, all cluster amplitudes defining \( \sigma_{\text{int}} \) cluster operator carry active indices only (or indices of active orbitals defining given \( \hbar \)). The external part \( \sigma_{\text{ext}}(\hbar) \) is defined by amplitudes carrying at least one inactive orbital index. However, in contrast to the SR-CC approach, internal/external parts of anti-Hermitian cluster operators are not defined in terms of excitations belonging explicitly to a given sub-algebra, but rather by indices defining active/inactive orbitals specific to a given \( \hbar \). Therefore \( \hbar \) will be used here in the context of CAS’s generator. Another difference with the SR-CC downfolding lies in the fact that while for the SR-CC cases components of cluster operators \( T_{\text{int}}(\hbar) \) and \( T_{\text{ext}}(\hbar) \) were commuting as a consequence of particle-hole formalism employed, in the unitary case, the operators forming \( \sigma_{\text{int}}(\hbar) \) and \( \sigma_{\text{ext}}(\hbar) \) are non-commuting.

Employing DUCC Ansatz, Eq. (17), one can show that in analogy to the SR-CC case, the energy of the entire system (once the exact form of \( \sigma_{\text{ext}}(\hbar) \) operator is known) can be calculated through the diagonalization of the effective/downfolded Hamiltonian in SES-generated active space, i.e.,

\[
H_{\text{eff}}(\hbar)e^{\sigma_{\text{int}}(\hbar)}\Phi = E e^{\sigma_{\text{int}}(\hbar)}\Phi, \tag{20}
\]

where

\[
H_{\text{eff}}(\hbar) = (P + Q_{\text{int}}(\hbar))H_{\text{ext}}(\hbar)(P + Q_{\text{int}}(\hbar)) \tag{21}
\]

and

\[
H_{\text{ext}}(\hbar) = e^{-\sigma_{\text{ext}}(\hbar)}He^{\sigma_{\text{ext}}(\hbar)}. \tag{22}
\]

The above results means that when the external cluster amplitudes are known (or can be effectively approximated), in analogy to single-reference SES-CC formalism, the energy (or its approximation) can be calculated by diagonalizing the Hermitian effective/downfolded Hamiltonian, given by Eq. (21), in the active space using various quantum or classical diagonalizers.

The analysis of the many-body structure of the \( \sigma_{\text{int}}(\hbar) \) and \( \sigma_{\text{ext}}(\hbar) \) operators\(^\text{81}\) shows that they can be approximated in a unitary CC manner:

\[
\sigma_{\text{int}}(\hbar) \simeq T_{\text{int}}(\hbar) - T_{\text{int}}(\hbar)^\dagger, \tag{23}
\]

\[
\sigma_{\text{ext}}(\hbar) \simeq T_{\text{ext}}(\hbar) - T_{\text{ext}}(\hbar)^\dagger, \tag{24}
\]

where \( T_{\text{int}}(\hbar) \) and \( T_{\text{ext}}(\hbar) \) are SR-CC-type internal and external cluster operators.

To make a practical use of Hermitian downfolded Hamiltonians, Eq. (20), in quantum calculations one has to deal with non-terminating expansions of Eq. (22) and determine approximate form of the \( T_{\text{ext}}(\hbar) \) operator to approximate its anti-Hermitian counterpart \( \sigma_{\text{ext}}(\hbar) \) according to Eq. (24). In recent studies, we demonstrated the feasibility of approximations based on the finite commutator expansion. We also demonstrated that \( T_{\text{ext}}(\hbar) \), provided by the CCSD formalism, can efficiently be used in building approximate form of the downfolded Hamiltonians. In particular, in this paper we will consider two approximate representations of the downfolded Hamiltonians \( (A \text{ and } B) \) defined by the following expressions for \( \bar{H}_{\text{ext}}(\hbar) \):

\[
\bar{H}_{\text{ext}}^{(A)} = H + [H, \sigma_{\text{ext}}(\hbar)] + \frac{1}{2}[[F_N, \sigma_{\text{ext}}(\hbar)], \sigma_{\text{ext}}(\hbar)], \tag{25}
\]

\[
\bar{H}_{\text{ext}}^{(B)} = H + [H, \sigma_{\text{ext}}(\hbar)] + \frac{1}{2}[[H, \sigma_{\text{ext}}(\hbar)], \sigma_{\text{ext}}(\hbar)] + \frac{1}{6}[[[F_N, \sigma_{\text{ext}}(\hbar)], \sigma_{\text{ext}}(\hbar)], \sigma_{\text{ext}}(\hbar)], \tag{26}
\]

where \( F_N \)-dependent commutators were introduced to provide perturbative consistency of single- \( (C1) \) and double-commutator \( (C2) \) expansions.

As a numerical example illustrating the efficiency of approximations \( C1 \) and \( C2 \) we use the LiF molecule at 1.0\( R_e \), 2.0\( R_e \), and 5.0\( R_e \) Li-F distances where \( R_e = 1.5639 \) Å. All calculations were performed using the cc-pVTZ basis set\(^\text{104} \) (employing spherical representation of \( d \) orbitals). The calculations using downfolded Hamiltonians \( C1 \) and \( C2 \) were performed employing restricted Hartree-Fock (RHF) orbitals and active spaces composed of 13 lowest-lying orbitals (6 occupied and 7 virtual). The results of the diagonalization of the downfolded Hamiltonians are shown in Table I. The \( C1 \) and \( C2 \) energies are compared with the CCSD, CCSDT, and CCSDTQ\(^\text{105} \) energies obtained with all orbitals correlated and the CCSDTQ formalism in the active space, which represent nearly exact diagonalization of the electronic Hamiltonian in the active space.

A comparison of the RHF and CCSDTQ-in-active-space results indicates that the active space used reproduces only a very small part of the total correlation energy approxi-

| Geometry | CCSD | CCSDT | CCSDTQ |
|----------|------|-------|--------|
| 1.0R_e   | 9.99 | 19.70 | 4.53   |
| 2.0R_e   |     |       |        |
| 5.0R_e   |     |       |        |

This deficiency in the active space choice, the C2 DUCC approximation yields 9.99, 19.70, and 4.53 milliHartree of error with respect to the CCSDTQ\(^\text{10} \) energies for 1.0\( R_e \), 2.0\( R_e \), and 5.0\( R_e \) geometries, respectively. These errors should be collated with the errors of the CCSDTQ-in-active-space approach of 310.97, 311.20, and 299.49 milliHartree. As seen from Table I, the inclusion of double commutator \( (C2 \text{ approximation}) \) results in a significant improvements of the energies obtained with the \( C1 \) scheme.

In analogy to the equivalence theorem of Section III, similar quantum flow algorithms can also be defined in the case of the Hermitian downfolding (see Ref. 80). Although, due to non-commutative character of generators defining anti-Hermitian \( \sigma_{\text{int}}(\hbar)_i \) and \( \sigma_{\text{ext}}(\hbar)_i \) \( (i = 1, \ldots, M) \), certain approximations has to be used (mainly associated with the use of the Trotter formula), similar flow can be defined for the Hermitian case (see Fig.1). In this flow, we couple Hermitian eigenvalue problems corresponding to various
active spaces (defined by sub-algebras \( h_i \) and corresponding effective Hamiltonians \( H_{\text{eff}}(h_i) \) \( (i = 1, \ldots, M) \)). The main advantage of this approach is the fact that larger subspaces of the Hilbert space can be sampled by a number of small-dimensionality active-space problems. This feature eliminates certain problems associated with (1) the need of using large qubits registers to represent the whole system, (2) qubit mappings of the basic operators, and (3) assuring anti-symmetry of the wave function of the entire system. For example, problem (3) is replaced by procedures that assure the anti-symmetry of the wave-functions of subsystems defined by the active space generated by various \( h_i \) \( (D_i \ll N \) as shown in Fig.1). This approach is ideal for developing quantum algorithms that take full advantage of the sparsity (or the local character of the correlation effects) of the system and uses only a small fraction of qubits \( (D_i, \) see Fig. 1) needed to describe the system represented by \( N \) spin orbitals.

V. MULTI-COMPONENT CC DOWNFOLDING

The development of computational algorithms for composite quantum systems keeps attracting a lot of attention in the field of quantum computing. Typical examples are related to quantum electrodynamics, nuclear physics, and quantum chemistry. In quantum chemistry, this effort is related to the development of methods for non-perturbative coupling of electronic degrees of freedom with strong external fields\(^{106,107} \) and formulations going beyond Born-
Oppenheimer approximation.\textsuperscript{108–111} Given the current status of quantum computing technology, it is important to provide techniques for compressing the dimensionality of these problems or finding an effective potential experienced by the one type of particles.

For simplicity, in this section we will consider a fictitious system composed of two types of Fermions A and B, defined by two sets of creation/annihilation operators \( \{a_\alpha, a_\alpha^\dagger\}_{\alpha=1}^{N_A} \) and \( \{b_\beta, b_\beta^\dagger\}_{\beta=1}^{N_B} \) (these operators should not be confused with the notation used in Section II) satisfying typical Fermionic anti-commutation relations \((\ldots,\cdot)\) and commuting \((\ldots,\cdot)\) between themselves
\[
[a_\alpha, b_\beta] = [a_\alpha^\dagger, b_\beta^\dagger] = 0, \quad (27)
\]
\[
[a_\alpha, b_\beta] = [a_\alpha^\dagger, b_\beta^\dagger] = 0. \quad (28)
\]

We will also assume specific form of the Hamiltonian
\[
H_{AB} = H_A + H_B + V_{AB} \quad (29)
\]
where \( H_A, H_B, \) and \( V_{AB} \) describe sub-systems A, B, and interactions between A and B, respectively. We will also assume that the interaction part commutes with the particle number operators \( n_A \) and \( n_B \) for systems A and B, i.e.,
\[
[V_{AB}, n_A] = [V_{AB}, n_B] = 0, \quad (30)
\]
\[
n_A = \sum_{\alpha=1}^{N_A} a_\alpha^\dagger a_\alpha, \quad n_B = \sum_{\beta=1}^{N_B} b_\beta^\dagger b_\beta. \quad (31)
\]

This situation is typically encountered in models relevant to non-Born-Oppenheimer approaches in electronic structure theory\textsuperscript{108–111} and nuclear structure theory.\textsuperscript{17}

Let us assume that the correlated ground-state wave function can be represented in the form of single reference CC wave function
\[
|\Psi_{AB}\rangle = e^{T_{AB}}|\Phi_{AB}\rangle, \quad (32)
\]
where cluster operator contains excitations correlating sub-system A (\( T_A \)) and sub-system B (\( T_B \)) as well as collective excitations involving both sub-systems (\( S_{AB} \)), i.e.,
\[
T_{AB} = T_A + T_B + S_{AB}. \quad (33)
\]

The reference function \( |\Phi_{AB}\rangle \) is a reference function for the composite system which is assume to be represented as
\[
|\Phi_{AB}\rangle = \Omega_A \Omega_B |0\rangle, \quad (34)
\]
where \(|0\rangle\) represents physical vacuum and \( \Omega_A \) and \( \Omega_B \) are string of \( a_\alpha^\dagger b_\beta^\dagger \) operators distributing electrons among occupied levels of sub-systems A and B, respectively.

The energy-dependent CC equation for the composite system takes the form
\[
(\langle p_{AB} + Q_{AB}|H_{AB}e^{T_{AB}}|\Phi_{AB}\rangle = E_{AB}e^{T_{AB}}|\Phi_{AB}\rangle, \quad (35)
\]
where \( E_{AB} \) is the energy of the composite system, \( p_{AB} \) is a projection operator onto the reference function \( |\Phi_{AB}\rangle \), and projection operator \( Q_{AB} \) can be decomposed as follows:
\[
Q_{AB} = Q_A + Q_B + Z_{AB}, \quad (36)
\]
where \( Q_A \) and \( Q_B \) are the projection operators onto excited Slater determinants obtained by exciting particles within sub-system A and B from \( |\Phi_{AB}\rangle \), respectively, and \( Z_{AB} \) corresponds to the projection operator onto sub-space spanned by excited Slater determinants where fermion particles of type A and B are excited simultaneously.

By projecting Eq. (35) onto \( (p_{AB} + Q_A) \) and introducing the resolution of identity \( e^{T_B + S_{AB}}e^{-T_B - S_{AB}} \) one obtains
\[
(p_{AB} + Q_A)e^{T_B + S_{AB}}(H_{AB,ext} - E_{AB})e^{T_A}|\Phi_{AB}\rangle = 0, \quad (37)
\]
where
\[
H_{AB,ext} = e^{-T_B - S_{AB}}H_{AB}e^{T_B + S_{AB}}. \quad (38)
\]

In analogy to analysis in Ref. 79 the role of \( e^{T_B + S_{AB}} \) in Eq. (37), reduces to the unit operator. This is a consequence of the fact that the operator \( T_B + S_{AB} \) produces excitations within sub-system B, which are subsequently eliminated by the \( Q_A \) projection operator. Consequently, Eq. (37) takes the form:
\[
H^{\text{eff}}(A)e^{T_A}|\Phi_{AB}\rangle = E_{AB}e^{T_A}|\Phi_{AB}\rangle, \quad (39)
\]
where the downfolded/effective Hamiltonian \( H^{\text{eff}}(A) \) is defined as
\[
H^{\text{eff}}(A) = (p_{AB} + Q_A)H_{AB,ext}(p_{AB} + Q_A). \quad (40)
\]

The above result shows that once \( T_B \) and \( S_{AB} \) amplitudes are know (or can be effectively approximated) the energy of the entire system can be calculated performing simulations on the sub-system A using effective Hamiltonian \( H^{\text{eff}}(A) \).

In addition to the simplest downfolding procedure described above, there are several other possible scenarios how downfolding procedures can be defined for the composite system:

- the utilization of second downfolding procedure to the \( H^{\text{eff}}(A) \) in reduced-size active space for sub-system A,
- the utilization of the composite active space that is represent by tensor product of active spaces for sub-systems A and B.

These techniques are especially interesting for the the explicit inclusion of nuclear degrees of freedom (for Fermionic nuclei) in the effective Hamiltonians describing electronic degrees of freedom in the non-Born-Oppenheimer formulations.

A Hermitian extension of the downfolding procedure can be accomplished by utilizing DUCC Ansatz for the composite system given by the expansion
\[
|\Psi_{AB}\rangle = e^{\sigma_A + \rho_{AB}}e^{\sigma_A}|\Phi_{AB}\rangle, \quad (41)
\]
where \( \sigma_A, \sigma_B, \) and \( \rho_{AB} \) are the anti-Hermitian operators defined by the cluster amplitudes with indices belonging to sub-systems A, B, and amplitudes defined by a mixed indices involving basis functions on A and B, respectively. As in the non-Hermitian case of downfolding discussed in this Section, we will focus on the downfolding of the entire sub-system B into the effective Hamiltonians for sub-system
A. Since creation/annihilation operators correspond to the
sub-systems A and B, the exactness of the above expansion
can be obtained as a generalization of the procedure based
on the elementary Givens rotations discussed in Ref. 112.
For the specific case discussed in this Section (based on the
downfolding of the entire B sub-system) one should assume
that all basis functions defining sub-system A are defined
as active indices (see Ref.81 for details).

Substituting Eq. (41) into the Schrödinger equations and
projecting onto \((P_{AB} + Q_A)\), one arrives the following form
of the equations

\[
H_{DUCC}^{\text{eff}}(A)e^{\sigma_1}|\Phi_{AB}\rangle = E_{AB}e^{\sigma_1}|\Phi_{AB}\rangle ,
\]

where

\[
H_{DUCC}^{\text{eff}}(A) = (P_{AB} + Q_A)H_{AB,\text{ext}}(P_{AB} + Q_A) ,
\]

and

\[
H_{AB,\text{ext}} = e^{-\sigma_B - \rho_{AB}}He^{\sigma_B + \rho_{AB}}.
\]

Again, the energy of the full system can be probed by
sub-system A using effective Hamiltonian \(H_{DUCC}^{\text{eff}}(A)\). For
example, one can envision the utilization of Eq. (42) in the
context of coupling nuclear and electronic degrees of free-
dom. In this case, sub-system A is represented by electrons
while system B corresponds do nuclei obeying Fermi statis-
tics. If \(\sigma_B\) and \(\rho_{AB}\) can be effectively approximated then the
\(H_{DUCC}^{\text{eff}}(A)\) Hamiltonian describes the behavior of electron
in the presence of “correlated” nuclei. The intensive de-
velopment of the CC models beyond Born-Oppenheimer
approximatos\(^{108,110,111}\) provides a reference for building
approximate, for eaxmple, perturbative, form of \(\sigma_B\) and
\(\rho_{AB}\) according formula analogous to Eqs. (24), which
requires the knowledge of \(T_B\) and \(S_{AB}\) to determine \(\sigma_B\)
and \(\rho_{AB}\), respectively.

VI. EXTRACTION OF THE ANALYTICAL FORM OF
INTERACTIONS IN MANY-BODY SYSTEMS

In standard formulations of downfolding methods it is
assumed (see Refs. 82 and 113)) that downfolded Hamil-
tonians are dominated by one- and two-body effects, i.e.,
using the language of second quantization \(H_{\text{eff}}\) can be approximated as (for simplicity, let us assume that only virtual
orbitals are downfolded)

\[
H_{DUCC}^{\text{eff}} \approx \sum_{PQ} \chi_{Q}^{P} a_{P}^{\dagger} a_{Q} + \frac{1}{2} \sum_{P,Q,R,S} \chi_{R}^{PQ} a_{R}^{\dagger} a_{S}^{\dagger} a_{Q} a_{P} ,
\]

where \(P,Q,R,S\) indices, \(\chi_{Q}^{P}\) and \(\chi_{R}^{PQ}\) represent active spin
orbitals and effective one- and two-body interactions, re-
spectively (non-antisymmetrized matrix elements \(\chi_{R}^{PQ}\) are
employed in (45)). Once the set of \(\{\chi_{Q}^{P}, \chi_{R}^{PQ}\}\) is known (at
the end of flow procedure) this information can be further
used to derive an analytical form of effective inter-electron
interactions. This can be accomplished by fitting the gen-
eral form of one-body \(u\) and two-body \(g\) interactions defined
as functions of-to-be-optimized parameters \(\gamma/\delta\) as well
as

\[
u = u(\gamma, r_1, \nabla_1, \ldots) ,
\]

\[
g = g(\delta, r_1, r_2, \nabla_1, \nabla_2, \ldots) ,
\]

These effective interactions replace standard one- and
two-body interactions in non-relativistic quantum chemistry and
are defined to minimize the discrepancies with \(\{\chi_{Q}^{P}, \chi_{R}^{PQ}\}\)
for a given discrete molecular spin-orbital set, i.e.,

\[
\min_{\gamma} \{ \sum_{PQ} |\nu_{Q}^{P}(\gamma) - \chi_{Q}^{P}| \} ,
\]

\[
\min_{\delta} \{ \sum_{P,Q,R,S} |g_{RS}^{PQ}(\delta) - \chi_{RS}^{PQ}| \} ,
\]

We believe that the utilization of efficient non-linear opti-
mizers or machine learning techniques can provide an effec-
tive form of the interactions \(u\) and \(g\) defined in small-size
active spaces. These effective interactions can be utilized in
low-order methodologies, including Hartree-Fock (HF) and
density functional theories (DFT). In the latter case func-
tions \(u\) and \(g\) can be utilized to develop/verify new forms
of exchange-correlations functionals. The access to the
analytical form of the inter-electron interactions can also
enable affordable and reliable ab-initio dynamics driven by
low-order methods.
VII. CONCLUSIONS

In this paper, we briefly review the current state of two variants of CC downfolding techniques. While the non-Hermitian downfolding and resulting active-space Hamiltonians are not a primary target for quantum computing, the equivalence theorem opens new possibilities regarding forming systematic reduced-scaling frameworks based on the quantum flow equations. In contrast to the existing reduced scaling CC formulations, where the notion of electron pair is rather descriptive and is based on the partitioning of the correlation energy with respect to contributions that can be indexed by pairs of the occupied orbitals, the present formalism defines the pair through the corresponding effective Hamiltonian. This fact has a fundamental advantage over ad hoc localization procedures—it allows in a natural way to introduce the pair density matrix. It also allows for a more systematic way of introducing certain classes of higher-rank excitations. The double unitary CC Ansatz provides a natural many-body language to introduce Hermitian downfolded representation of many-body Hamiltonians in reduced-dimensionality active spaces. To approximate non-terminating commutator expansion of downfolded Hamiltonians, we use finite commutator expansions. On the LiF example, we demonstrated that the inclusion of double commutator terms leads to systematic improvements of the results obtained with single commutator expansion even in a situation when an active space is not providing a good zero-th order approximation of correlation effects. It should be also stressed that the downfolded Hamiltonians based on the double commutator expansion are capable of reducing the error of energies obtained by the diagonalization of the bare-Hamiltonian in the same-size active space by more than an order of magnitude (in fact, for the 1.0R_e and 5.0R_e one could witness 30- and 60-fold reduction in energy errors with respect to accurate CC results obtained by correlating all molecular orbitals).

In the second part of the paper, we extended non-Hermitian and Hermitian downfolding to multi-component quantum systems. As an example, we used the model system composed of two types of Fermions, which epitomize typical situations encountered in nuclear physics and for certain types of nuclei in non-Born-Oppenheimer electronic structure theory. We have also outlined an approximate procedure to extract the semi-analytical form of the one- and two-body inter-electron interactions in active space based on the minimization procedure utilizing one- and two-body interactions defining downfolded Hamiltonians. In the future, we will explore the usefulness of machine learning techniques for this procedure.

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