Quantum simulations employing connected moments expansions

Karol Kowalski1, a) and Bo Peng1, b)

Physical & Computational Sciences Division, Pacific Northwest National Laboratory, Richland, WA 99354, USA

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Further advancement of quantum computing (QC) is contingent on enabling many-body models that avoid deep circuits and excessive use of CNOT gates. To this end, we develop a QC approach employing finite-order connected moment expansions (CMX) and affordable procedures for initial state preparation. We demonstrate the performance of our approach employing several quantum variants of CMX through the classical emulations on the H2 molecule potential energy surface and the Anderson model with a broad range of correlation strength. The results show that our approach is robust and flexible. Good agreements with exact solutions can be maintained even at the dissociation and strong correlation limits.

I. INTRODUCTION

The quantum computing (QC) techniques attract much attention in many areas of mathematics, physics, and chemistry by providing a means to address insurmountable computational barriers for simulating quantum systems on classical computers. One of the best illustrations of this fact is associated with solving Schrödinger equations for many-electron systems in quantum chemistry, where the attempt of including all configurations spanning Hilbert space (or at least configurations from the subspace relevant to a problem of interest) quickly evolves into numerical problems characterized by exponentially growing numerical cost. Unfortunately, for a large class of the problems commonly referred to as the strongly correlated systems, the inclusion of all configurations is necessary to obtain a desired level of accuracy. To bypass these problems, several classes of many-body methods targeting the so-called full configuration interaction (FCI) accuracy have been developed including high-rank coupled-cluster (CC), selected CI, density matrix renormalization group (DMRG), stochastic and semi-stochastic CI, hybrid stochastic CI and CC, and virtual orbitals many-body expansions and virtual orbitals many-body expansions. Recently, accuracies of these formalisms have been evaluated on the example of benzene system, where considerable differences in the calculated energies with high-accuracy formalisms were reported.

Although quantum computing has not reached its maturity yet, intensive effort has been directed towards developing algorithms for quantum simulations that can take advantage of early quantum registers. Among several algorithms that are being developed for this purpose one should mention variational quantum eigensolver (VQE), quantum phase estimation (QPE), and more recently quantum algorithms for imaginary time evolution (ITE). These algorithms draw heavily on the utilization of Trotter formulas for evaluating the exponential forms of the operators corresponding to unitary coupled cluster ansatzes, unitary time evolution of the system, and the solution of the imaginary-time Schrödinger equation, respectively. For example, for the unitary time evolution defined by Hamiltonian $H$, where $H$ is sum of multiple (generally non-commuting) terms $H_j$, $H = \sum_{j=1}^{M} H_j$ (here $H_j = h_j P_j$ with $h_j$ defining interactions of the system and $P_j$ representing tensor product of Pauli matrices and/or identity matrices), it is hard to encode $e^{-\imath H t}$ directly into quantum gates. Instead in quantum simulations one resorts to the Trotter formula with $K$ Trotter steps:

$$e^{-\imath H t} \simeq U(\Delta t)^K$$

where $\Delta t = t/K$ and $U(\Delta t) = \prod_{j=1}^{M} e^{-\imath h_j P_j \Delta t}$. Note that even in its simplest case ($K = 1$) Eq. (1) results in arbitrary connectivity between qubits. With number of terms in Hamiltonian being proportional to $O(N^4)$ (where $N$ stands for the number of one-particle basis functions employed) the optimization of the corresponding circuits plays a crucial role in the quantum simulations (especially when employing the techniques related to the fermionic swap network). Nevertheless, even for formulations using reduced number of terms (e.g. proportional to $O(N^2)$) in many-body Hamiltonian, the resulting gate depth still depends polynomially on $N$ and the total number of CNOT gates is still proportional to $O(N)$ (letting alone the multiple CNOT gates for multi-qubit operators needed for example in QPE).

In this letter we propose an alternative approach for the quantum simulations of quantum systems, where the number of instances for using the Trotter formula is significantly reduced (or eliminated at all). For this purpose we utilize connected moments expansion introduced by Horn and Weinstein and further advanced by Cioslowski and others. The energy of a quantum system can be calculated using trial wave function $|\Phi\rangle$ (non-orthogonal to the exact ground state $|\Psi\rangle$) and the expectation values of the powers of the Hamiltonian operator $\langle \Phi | H^n | \Phi \rangle$. Here, $\langle \Phi | H^n | \Phi \rangle$’s are calculated using simple variants of Hadamard test, where CNOT gates are used to control the products of the unitaries $P_j$. Additional CNOT gates are optimally required for strongly correlated systems where rudimentary static correlation effects need to be captured in the trial state preparation, which in a realistic setting requires only the inclusion of a relatively small number of parameters in the VQE formulations.

II. CONNECTED MOMENTS EXPANSION

The connected moments expansion is derived from Horn-Weinstein (HW) theorem that relates the function $E(\tau)$ to

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a)Electronic mail: karol.kowalski@pnnl.gov
b)Electronic mail: peng398@pnnl.gov
a series expansion in $\tau$ with coefficients being represented by connected moments $I_k$,
\[
E(\tau) = \frac{\langle \Phi | H e^{-\tau H} | \Phi \rangle}{\langle \Phi | e^{-\tau H} | \Phi \rangle} = \sum_{k=0}^{\infty} \frac{(-\tau)^k}{k!} I_{k+1} \ .
\]
Here the connected moments $I_k$ are defined through a recursive formula (see Ref. 
38 for details)
\[
I_k = \langle \Phi | H^k | \Phi \rangle - \sum_{i=1}^{k-2} \left( \frac{k-1}{i} \right) I_{i+1} \langle \Phi | H^{k-i-1} | \Phi \rangle.
\]
It can be shown that $E(\tau)$ is monotonically decreasing and
\[\lim_{\tau \to \infty} E(\tau) \] corresponds to the exact ground-state energy $E_0$, which is a consequence of the fact that $e^{-\tau H}$ contracts any trial wave function $|\Phi\rangle$ towards the true ground state $|\Psi\rangle$ (assuming $\langle \Phi | \Psi \rangle \neq 0$). In practical applications based on the truncated moments expansion, Padé approximants need to be used to reproduce the proper behavior in the $\tau = \infty$ limit. The behavior of this expansion and some techniques for maintaining size-extensivity were discussed in Ref. 41.

By applying re-summation techniques to Eq. (2) Cioslowski \cite{39} derived an analytical form for exact energy in the $\tau = \infty$ limit
\[
E_0 = I_1 - \sum_{n=1}^{\infty} \frac{S_{2,n}}{S_{3,n}} (1 + \frac{S_{2,2}}{S_{2,1} S_{3,2}} \ldots (1 + \frac{S_{2,m}}{S_{2,m-1} S_{3,m}})) \ldots)
\]
where $S_{k,1} = I_k$ ($k = 2, 3, \ldots$) and $S_{k,i+1} = S_{k,1} S_{k+2,j} - S_{k+1,j}$. The truncation of the CMX series (4) after the first K terms leads to the CMX(K) approximations. For example, CMX(2) and CMX(3) energies are defined as follows,
\[
E_{0}^{\text{CMX(2)}} = I_1 - \frac{I_2}{I_3},
\]
(5)
\[
E_{0}^{\text{CMX(3)}} = I_1 - \frac{I_2}{I_3} - \frac{(I_2 I_4 - I_3^2)}{I_3 I_5 - I_4^2}.
\]
(6)
The CMX formalism provides a trade-off between the rank of the connected moments included in the approximation and the quality of the trial wave function. In this formalism one can naturally introduce the multi-configurational nature of sought-for-state \cite{39} or even use its correlated representations in the form of truncated CI or CC wave functions. \cite{48}

The analytical properties of CMX expansions and their relations to Lanczos methods have been discussed in the literature. \cite{40,44,45,49-52} Several techniques have been introduced to counteract possible problems associated with singular behavior of the CMX expansions (as pointed out by Mancini there exists an infinite number of valid CMX expansions) \cite{45} including Knowles’s generalized Padé approximation, \cite{40} alternate moments expansion (AMX), \cite{45} and generalized moments expansion. \cite{51} Additionally, as suggested by Noga et al. in Ref. 48 a proper definition of the trial wave function may significantly alleviate possible issues associated with singular behavior. An interesting yet less known CMX formulation has been proposed by Peeters and Devreese \cite{53} (further analyzed by Soldatov \cite{54}) where the upper bound of the ground-state energy in $n$-th order approximation is calculated as a root of the polynomial,
\[
P_n(x) = \sum_{n=1}^{\infty} a_n x^n - 1,\] where coefficients $a_n$’s (forming vector $a$) are obtained by solving linear equations, $M_a = -b$, with matrix element $M_{ij} = \langle \Phi | H^{2n-2(i+j)} | \Phi \rangle$ and vector component $b_i = \langle \Phi | H^{2n-i} | \Phi \rangle$. Remaining roots of the polynomial correspond to the upper bounds of the excited-state energies.

Recently, quantum computing algorithms for imaginary time evolution and quantum Lanczos algorithm \cite{33,34,55} have attracted a lot of attention. One of the most appealing features of these methods is the avoidance of a large number of ancillae qubits and complex circuits. In this context, we believe that the CMX framework provides an alternatively interesting platform for the utilization of near-term quantum architectures, where one can almost entirely eliminate Trotter steps and significantly reduce the number of CNOT gates. Below we will describe a simple quantum algorithms for calculating $\langle \Phi | H^n | \Phi \rangle$ for an arbitrary trial state.

III. QUANTUM ALGORITHMS FOR CONNECTED MOMENTS EXPANSION

The main difference between our algorithm and ITQ algorithms (for example QITE approach of Ref. 34) is the fact that in the latter approach the infinitesimal imaginary time evolution generated by unitary $H_j$, i.e. $e^{-\Delta t H_j}$, is mirrored by a unitary evolution generated by the $e^{-i\Delta t C_j A_j}$ operator acting on properly normalized states, while in the former method the moments $I_n = \langle \Phi | H^n | \Phi \rangle$ are directly calculated.

Figure 1. A schematic representation of the Hadamard test employed for calculating real part of $\langle \Phi | U_j | \Phi \rangle$.

Quantum CMX algorithm works as follows: (1) preparing an initial state $|\Phi\rangle$. In our numerical tests we used a simple single Slater determinant for $|\Phi\rangle$. Alternatively, one can envision the use of "non-aggressive" variant of VQE where a small number of amplitudes that define basic static correlation effects in the sought-for-wave-function is used. (2) Performing Hadamard test to evaluate $p_j = \langle \Phi | U_j | \Phi \rangle$, where $U_j = \prod_{k=1}^{J} P_{b_k}$ (for CMX(K) formulation $l < K$) (see Fig. 1) and cumulative index $J$ designates $k$-tuple $\{j_1, \ldots, J\}$. The contribution from $p_j$ to $I_l$ is equal to $p_j \times \prod_{b_k} b_k$. (3) Once all $I_l$ ($l = 1, \ldots, K$) are known we choose the optimal form of the CMX expansion. For example, if $I_3$ is close to 0, then we choose a CMX form that is not using $I_3$ in the denominator (this will provide an optimal utilization of the information obtained
from quantum computing). Additionally, using identity
\( \sigma_i \sigma_j = \delta_{ij} I + i \epsilon_{ijk} \sigma_k \) one can reduce entire
\( U_I \) to an effective unitary \( \bar{P}_I \) corresponding to a tensor
product of Pauli matrices and/or identity matrices with an
appropriate product of phase factors (Fig. 2). It means that
the gate depth is exactly the same irrespective of the rank
\( l \) of the calculated moment \( I_l \) (of course, the number of terms to be evaluated in this way increases with the rank
increasing). Summarizing, the gate depth of the quantum
CMX algorithm is mainly determined by the wave function
preparation step and the practical realization of multi-qubit
CNOT in the Hadamard test. In the case where the state
preparation corresponds to flipping states from \( |0\rangle \) to \( |1\rangle \)
using \( X \) operators (for quantum registers corresponding to
occupied spin-orbitals), the resulting gate depth is extremely shallow. The utilization of VQE for \( \Phi \) state
preparation increases the gate depth. Even though, if it
includes only a small number of amplitudes, the resulting
gate depth should not be excessive.

IV. SIMULATIONS AND RESULTS

For building quantum circuits to calculate the moments,
\( I_l \)'s, we used Qiskit software.\(^{56}\) As benchmark systems
we chose \( H_2 \) molecule in minimum basis (for various
geometries corresponding to situations characterized by
weak and strong correlation effects)\(^{57}\) and two-site single-
impurity Anderson model (for a broad range of hybridization
strength \( V \)) described by model Hamiltonians. In our
studies we used various orders of Cioslowski CMX(K),
Knowles CMX(K),\(^{40}\) and K-th order Peeters, Devreese,
Soldatov expansion, PDS(K). The results of quantum simu-
lations are shown in Figs. 3 and 4.

In Fig. 3, all three CMX variants with up to fourth
order expansions are able to reproduce the FCI energy in
at least mHartree level for \( R_{H-H} \) being up to 1.50 Å.
The only exception is PDS(1), which corresponds to
\( I_1 = \langle \Phi | H | \Phi \rangle \). The higher-order PDS expansions perform
exceptionally well, and the deviations between the PDS(K)
(\( K = 2 \) - 4) and FCI energy are below \( 10^{-14} \) a.u. The original
Cioslowski CMX results start to diverge and show
singularity on the potential energy surface when \( R_{H-H} \) is
approaching dissociation limit (\( R_{H-H} > 1.50 \) Å).

The singularity problem originates from the near-zero
connected moments defining the denominators in the CMX
energy expansion (see Eqns. (5) and (6)). Mancini et
al.\(^{45}\) have shown how to mitigate these singularities by re-
summing the CMX expansion (i.e. the AMX approach) and
introducing new class of denominators corresponding to
non-zero higher order moments. By a similar substitution
derived from a Padé approximant, Knowles\(^{40}\) has shown that
the singularity could be largely eliminated. This can be
observed from \( H_2 \) results as shown in Fig. 3a,b, where at
the dissociation limit, the Knowles’s approach (Fig. 3b) is
able to provide a smooth curve and move towards the FCI
limit in an oscillatory manner, while divergences appears
in the original Cioslowski’s CMX formalism after the sec-
ond order, and additionally the singularity emerges in the
original CMX(4) for \( R_{H-H} \) being \( \sim 1.75 \) Å and \( \sim 2.50 \) Å.

We further examine the performance of the CMX vari-
ants in terms of computing the ground state energy rang-
ing from weak to strong correlation in the context of a
single-impurity Anderson model (SIAM). As can be seen
from Fig. 4, for fixed Hubbard repulsion \( U (U = 8) \), the
effect correlation energy curve monotonically declines as
the hybridization \( V \) becomes large. Among the low order
CMX(K) (\( K = 1, 2 \)) results, all the curves behave very similarly
showing the same trend as the FCI curve (except the
PDS(1) curve where the correlation contribution from the
single-particle \( V \) terms operating on the present trial wave
function is zero). Among high order CMX(K) (\( K > 2 \)) re-
sults, the original CMX diverges as the \( V \) becomes large,
while the Knowles’s approach improves the original CMX
results by showing slow convergence as the expansion or-
der increases. On the other hand, PDS approach shows fast
convergence, and basically reproduces the FCI results after
the second order.

Note that in all the CMX calculations shown in Figs. 3
and 4, the simple trial wave function (i.e. single determin-
ate wave function) is universally applied in either weak
results can be obtained with alternative parametrizations of $U(\theta)$ given by $\exp(i\theta X_0 Y_1 Z_3)$ or $\exp(i\theta Y_0 X_1)$.

The striking advantage offered by quantum CMX algorithms is its “insensitivity” with respect to the choice of initial state. As long as the initial state is non-orthogonal to the exact state, the CMX expansions (especially the PDS approach) are capable of reproducing exact or near-to-exact energies. This feature could largely eliminate the challenging state-preparation process in quantum algorithms. As we can see from the performance of the CMX variants in Figs. 3 and 4, instead of constructing a better trial wave function, the moments matrix $M$ can be constructed to avoid singularity problems for quasi-degenerate situations.

or strong correlation scenarios, and the commonly used unitary coupled-cluster ansatze, or in general the unitary operations for the state preparation as discussed in the previous practices (see e.g. Refs. 57–61), were totally skipped. In other word, a crude trial wave function may be still useful by employing the proposed quantum algorithm to obtain highly accurate energies. This does not defy the accuracy of the ground state energy computed at the low and strong correlation limits. Therefore, the PDS can further work with some unitary parameterization to improve the accuracy of the ground state energy computed at the low expansion order. As shown in Fig. 5, a unitary parameterization as suggested by Ref. 60, $U(\theta) = \exp(i\theta Y_0 X_1 Z_2 X_3)$, can help the PDS(2) to well reproduce the FCI curve. As can be seen in Fig. 5d, the energy deviation can be reduced on average by about three orders of magnitude from employing only PDS(2) to employing a mixed scheme, where the PDS(2) energies are minimized through rotating the trial wave function using the selected unitary operation $U(\theta)$ over the studied range of $V$ values. Note here that employing the selected unitary parameterization alone cannot produce FCI energy for the four-qubit, 2-electron problem (here $E_U(\theta) = \langle \Phi | U(\theta) \hat{H}_{\text{SIAM}} | \Phi \rangle = -4$ for $\langle \Phi \rangle = |0110\rangle$ and arbitrary $\theta$, see Fig. 5(b,c)).

V. CONCLUSIONS

We demonstrated the feasibility of a new quantum computing algorithm based on calculating various types of CMX expansions. The discussed algorithm is robust in the sense of possible gate depth reduction and could be highly scalable with the increasing number of available qubits. Since the CMX algorithms offer a trade-off between the quality of the trial wave function and the rank of the moments required to achieve a high level of accuracy, we believe that its combination with the VQE approach, that is utilizing the unitary coupled-cluster representation of the trial wave function together with the quantum CMX algorithms, may provide a much needed way for improving the quality of the VQE energies. Additionally, the flexibility of CMX re-summation techniques allows one
to define customized (or optimal) form of the expansion that avoids possible singularities associated with the near-zero values of the calculated connected moments. In our studies, we demonstrated that the PDS expansions provide superior results compared to other CMX expansions for the systems considered here, especially for the strongly correlated situations (characterized by large $R_{1}-H$ distance or large hybridization strength) studied with a relatively poor choice of trial wave function corresponding to a single Slater determinant. In our future studies we will explore the applicability of the PDS expansions for describing multi-configuration states and excited states. Of special interest will be integrating quantum CMX algorithms with multi-configuration states and excited states. Of special interest will be integrating quantum CMX algorithms with multi-configuration states and excited states.

VI. ACKNOWLEDGEMENT

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