A quantum advantage for Density Functional Theory?

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The technological revolution brought about by quantum computers promises to solve problems with high economical and societal impact that remain intractable on classical computers. While several quantum algorithms have been devoted to solve the many-body problem in quantum chemistry, the focus is on wavefunction theory that is limited to relatively small systems, even for quantum computers, i.e., the size of tractable systems being roughly limited by the number of qubits available. Computations on large systems rely mainly on mean-field-type approaches such as density functional theory, for which no quantum advantage has been envisioned so far. In this work, we question this a priori by investigating the benefit of quantum computers to scale up not only many-body wavefunction methods, but also mean-field-type methods, and consequently the all range of application of quantum chemistry.

I. INTRODUCTION

Quantum computers have shown promises to solve problems that are currently intractable on classical computers for specific problems, but they are still in their infancy to solve problems of industrial interest faster than classical computers [1]. One of the nearest-term application of quantum computers is quantum chemistry (see Refs. [2–4] and references therein), where the focus is on wavefunction theory (WFT) that targets a numerically exact solution of the electronic structure problem. While quantum phase estimation (QPE) algorithms are in principle capable of solving the problem in its entirety [5–9], the required circuit-depth precludes their application in the noisy intermediate-scale quantum (NISQ) era [10]. More efficient algorithms have therefore been developed such as the quantum stochastic drift protocol [11], or the direct simulation of the Hamiltonian using linear combination of unitaries and the qubitization formalism [12–15]. More adapted to the NISQ era, several variational quantum algorithms (hybrid quantum-classical algorithms) have been specifically designed to prepare ground states [16–20] and, more recently, excited states [21–23], and to calculate of atomic forces and molecular properties [24–27].

However, despite the exponential speed-up announced by quantum computers, it remains unclear when quantum advantage will really be met in practice, and expecting any high impact applications in a near future is difficult [28–30]. Indeed, the applications of quantum algorithms for quantum chemistry remain limited in terms of size of affordable systems, as the size of the system – i.e., the size of the basis-set – dictates the number of required qubits. Even though the number of qubits on quantum devices is expected to increase rapidly, stable machines able to tackle real quantum chemistry systems are not expected in the next few years. Within noisy quantum computers of the NISQ era, a high precision outcome is illusive and the quest to chemical precision remains a long journey.

Currently, classical computations on large system, from chemistry, condensed matter physics and even biology, rely mainly on density functional theory (DFT) [31, 32], for which no quantum advantage is envisioned as they only scale cubically with respect to the system size. Few attempts have been done to perform mean-field approximations on quantum computers, such as Hartree–Fock with the experimental milestone on a 12 qubits platform [33], or the calculation of the one-particle density matrix on quantum annealers [34]. In both cases, no quantum advantage has been envisioned.

In this work, we investigate the benefit of using digital quantum computers to scale up mean-field-type methods such as DFT. A possible quantum advantage is discussed in terms of a counter-intuitive mapping between the Kohn–Sham Hamiltonian and an auxiliary interacting one, expressed in the computational basis, at the opposite of what has been done for decades. With such new encoding, mean-field type Hamiltonians can, in some ideal cases, be solved with an exponential speed-up on quantum computers, in analogy with interacting Hamiltonians. Besides, because DFT has its own level of approximations that leads to errors of orders of magnitudes higher than chemical accuracy, the impact of the noise of quantum computers plays a much less significant role. Hence, less efforts in applying quantum error correction codes and error mitigation techniques should be required to correct the noise of quantum computers. This is a key observation that could bring DFT to center stage as the nearest-term application of quantum computers for molecular, material and biological science.

II. A QUANTUM ALGORITHM FOR THE DENSITY FUNCTIONAL THEORY

The undeniable success of DFT relies on the transformation of the complex many-body quantum elec-
tronic problem into an effective and electronic density-dependent one-body problem [31, 32]. More precisely, the ground-state energy and ground-state electronic density of a given complex system can be obtained exactly by solving the Kohn–Sham equation [32]:

$$\left( \hat{T} + \mathbf{v}^{KS}[\mathbf{n}] \right) |\Phi^{KS}\rangle = \mathcal{E}^{KS} |\Phi^{KS}\rangle,$$  \hspace{1cm} (1)

where $\hat{T}$ is the kinetic energy operator, $|\Phi^{KS}\rangle$ is a single Slater determinant and $\mathbf{v}^{KS}[\mathbf{n}] = \mathbf{v}^{\text{ext}} + \mathbf{v}^{H}[\mathbf{n}] + \mathbf{v}^{xc}[\mathbf{n}]$ is the density-dependent Kohn–Sham potential. It contains the external potential $\mathbf{v}^{\text{ext}}$, i.e., the ion-electron interaction, the trivial Hartree potential $\mathbf{v}^{H}[\mathbf{n}]$ and finally the so-called exchange and correlation (XC) potential $\mathbf{v}^{xc}[\mathbf{n}]$. The latter contains all non-trivial contributions arising from the electron-electron interaction.

The KS equation (1) is complemented by the self-consistent condition to obtain the electronic density

$$\mathbf{n}(\vec{r}) = \sum_{k=1}^{N_{\text{occ}}} \langle \vec{r} | \varphi_k \rangle \langle \varphi_k | \vec{r} \rangle$$  \hspace{1cm} (2)

where $\{\varphi_k\}$ denotes the KS orbitals in $|\Phi^{KS}\rangle$. Note that the occupations for a closed-shell system are recovered from the $N_{\text{occ}}$ occupied KS spatial orbitals. The electronic density is then used to compute a new KS potential, so that Eqs. (1) and (2) are self-consistently and iteratively solved until convergence is reached. Providing the exact XC functional leads to equivalent electronic density between the non-interacting auxiliary KS system and the ground state of the physical system $\mathbf{n}_0$, i.e. $\mathbf{n}^{\text{KS}} = \mathbf{n}_0$. The exact ground-state energy of the physical system is also recovered as follows:

$$E_0 = \mathcal{E}^{KS} + E^{\text{Hxc}}[\mathbf{n}] - \left( \mathbf{v}^{\text{Hxc}}[\mathbf{n}] |\Phi^{KS}\rangle \langle \Phi^{KS}| \right).$$

Despite plenty of different flavors of approximations, the exact XC functional is in general not known thus leading to subsequent density- and functional-driven errors [35].

### A. A possible quantum advantage? From Kohn–Sham to auxiliary interacting Hamiltonian mapping

The key point under any quantum algorithm is to encode the problem in a comprehensive language for the quantum computer. In the context of WFT, the encoding of an interacting system described in a basis set of $L$ spin-orbitals generally requires a quantum computer with $L$ qubits (for instance using the Jordan–Wigner (JW) transformation [36]). It implies a one-to-one correspondence between the state of the qubits and the occupation of the spin-orbitals. More precisely, a qubit in state $|0\rangle$ (respectively $|1\rangle$) is interpreted as an empty (respectively occupied) spin-orbital. Hence, each of the $2^L$ bitstrings generated by the $L$ qubits corresponds to a given electronic configuration (Slater determinant) in the entire Fock space. While this encoding proves itself very efficient for an interacting system, it seems counterproductive for a non-interacting system which can be described by a single Slater determinant. In practice, the non-interacting Hamiltonian is expressed in the monoparticle basis of dimension $L$ for which diagonalization typically scales as $\mathcal{O}(L^3)$, instead of the many-body basis of dimension $2^L$ that is infamous for its exponential scaling.

Considering the non-interacting KS Hamiltonian (1) described in finite computational basis-set $\{\chi_i\}$ composed of $L$ spin-orbitals, it reads in second quantization:

$$\hat{h}^{\text{KS}} = \sum_{i,j=1}^{L} h^{\text{KS}}_{ij} \left( \hat{c}_i \hat{c}_j + h.c. \right)$$  \hspace{1cm} (3)

where $\hat{c}_i^\dagger$ (\hat{c}_i) refers to the creation (annihilation) operator of an electron in the spin-orbital $\chi_i$, respectively. $h^{\text{KS}}_{ij}$ contains both the kinetic contributions and the KS potential. In the following we propose a transformation to solve this problem with only $M = \mathcal{O}(\log_2 L)$ qubits by mapping $\hat{h}^{\text{KS}}$ of dimension $L \times L$ onto a $M$-qubit system, in the spirit of the Harrow–Hassidim–Lloyd algorithm to solve linear systems of equations [37]. In complete analogy with the usual encoding of an interacting problem onto a quantum computer, this $M$-qubit system can be interpreted as an auxiliary system of $M$ pseudo-spin-orbitals. This means that each state of the computational basis denoted by $\{|I\rangle = \eta_I^1, \ldots, \eta_I^L \rangle \}$ corresponds to a configuration where each pseudo spin-orbital $\mu$ is either empty ($\eta_I^\mu = 0$) or occupied ($\eta_I^\mu = 1$). Note again that the number of configurations $\{|I\rangle \}$ corresponds to the number of orbitals $L = 2^M$. This leads to a direct mapping between the non-interacting Hamiltonian $\hat{h}^{\text{KS}}$ to an auxiliary interacting Hamiltonian $\hat{H}^{\text{aux}}$.

$$\hat{h}^{\text{KS}} \rightarrow \hat{H}^{\text{aux}} = \sum_{I=0, J=0}^{2^M-1} H^{\text{aux}}_{IJ} |I\rangle \langle J|.$$  \hspace{1cm} (4)

where $I$ and $J$ are the integers corresponding to the binary strings $|I\rangle$ and $|J\rangle$ such that $H^{\text{aux}}_{IJ} = h_{(I+1)(J+1)}$. In other words, each spin-orbital of the KS computational basis-set $\{|\chi_i\rangle\}$ is associated to a binary string $|I\rangle$ of the Fock-space of $H^{\text{aux}}$. Note that this mapping is actually arbitrary and that one can map any computational basis state to any orbital. Hence, depending on the architecture of the quantum device, one mapping could be more appropriate than another to reduce the circuit complexity and mitigate the effect of noise.

Let us now introduce the JW transformation for phaseless fermions, for $1 \leq \mu \leq M$,

$$\hat{b}_\mu = \Gamma^{\otimes \mu-1} \otimes S^- \otimes \Gamma^{\otimes M-\mu} \quad \text{with} \quad S^- = \frac{1}{2} \left( X + iY \right)$$  \hspace{1.3cm} (5)

$$\hat{b}_\mu^\dagger = \Gamma^{\otimes \mu-1} \otimes S^+ \otimes \Gamma^{\otimes M-\mu} \quad \text{with} \quad S^+ = \frac{1}{2} \left( X - iY \right)$$  \hspace{1.3cm} (6)

where $\hat{b}_\mu^\dagger$ ($\hat{b}_\mu$) is the creation (annihilation) operator of a phaseless fermion in the auxiliary orbital $\mu$. $X$, $Y$ and $Z$
are the usual Pauli matrices and \( I \) is the identity matrix. The projector in Eq. (4) reads

\[
|I\rangle\langle J| = \prod_{\mu=1}^{M} \left( \hat{b}_{\mu} \hat{b}_{\mu}^\dagger \right) (1-\eta_{\mu}^I)(1-\eta_{\mu}^J) \left( \hat{b}_{\mu}^\dagger \right) (1-\eta_{\mu}^I^*) \eta_{\mu}^J^*.
\]

The Hamiltonian in Eq. (4) is now written as a linear combination of Pauli strings, which expectation values can be measured on a quantum computer. As readily seen in Eqs. (2) and (3), the occupied KS orbitals and associated energies are required. At each iteration of the KS self-consistent procedure, they are solutions of the Schrödinger equation

\[ \hat{H}^{\text{aux}} |\varphi_k\rangle = \varepsilon_k |\varphi_k\rangle \]  

for which quantum computers are known to provide an exponential speed-up compared to classical computers.

In Fig. 1, we represent the possible improvement in computational cost by using the Kohn–Sham formalism together with quantum computers. Note that each method in Fig. 1 is in principle exact, although approximate functionals are used in practice in DFT.

**B. Kohn–Sham self-consistent conditions on a quantum device**

Given the KS equation mapped onto an auxiliary interacting Hamiltonian in Eq. (8), we propose to use a NISQ-adapted variational quantum eigensolver (VQE) [16, 17] to efficiently extract the occupied KS orbitals energies on quantum computers. The case of phase estimation algorithms is discussed in appendix A. Because in our context, one has to solve Eq. (8) for the first \( N_{\text{occ}} \) lowest-energy states, we naturally turn to the ensemble extension of VQE, so-called subspace-search VQE [22] (SS-VQE) or state-averaged VQE [23]. Within SS-VQE, we consider an ensemble of \( N_{\text{occ}} \) states,

\[ |\varphi_k(\theta)\rangle = U(\theta) |\varphi_k\rangle, \quad 1 \leq k \leq N_{\text{occ}}, \]  

where \( \{|\varphi_k\rangle\} \) is an orthonormal basis issued from the computational basis and simple to prepare. \( U(\theta) \) is usually referred to the circuit ansatz, i.e. a unitary transformation composed of \( \theta \)-parametrized quantum gates. The set of parameters \( \theta \) are classically optimized by minimizing the ensemble energy,

\[ E^{\text{SS-VQE}} = \min_{\theta} \left\{ \sum_k w_k \langle \varphi_k(\theta) | \hat{H} | \varphi_k(\theta) \rangle \right\}. \]  

Considering \( w_1 > w_2 > \cdots > w_N \), it results that the optimized states are by construction approaching the KS orbitals, \(|\varphi_k(\theta^*)\rangle \approx |\varphi_k\rangle \) [22] (up to the error of the classical optimizer and the expressibility of the ansatz), where \( \theta^* \) are the parameters that minimize Eq. (10). Note that in contrast to the usual WFT problems studied within VQE, the auxiliary many-body problem described in this paper is not physical anymore, and no constraints on the spin or the number of particles have to be considered. Consequently, hardware efficient ansatz can be used for \( U(\theta) \) instead of physically motivated ansatz such as the unitary coupled cluster ansatz which requires very deep circuits.

At this stage it is important to highlight that we have a direct access to the occupation number of orbitals in the computational basis. Indeed, repeated measurements of the state \(|\varphi_k\rangle = \sum_I |\varphi_k(I)\rangle |I\rangle \) give access to \(|\varphi_k(I)|^2 \), the probability to measure \(|I\rangle \), and thus to the computational basis orbitals occupations

\[ n_I = 2 \sum_k |\varphi_k(I)|^2. \]  

Within the lattice version of DFT, known as Site-Occupation Functional Theory (SOFT) [38, 39], the orbital dependent XC potential \( v_{\text{xc}}[n] \), that only depends on orbital occupations \( n = \{n_1, \ldots, n_I, \ldots, n_L\} \), is then straightforwardly accessible to loop the KS protocol. This flavor of DFT appears optimal for quantum computers. However SOFT remains mainly applied to lattice Hamiltonians and model systems [40–49], even though recent promising works aim to extend it to real quantum chemistry problems [50, 51].

Going back to standard DFT, the knowledge of both the KS and computational basis-set occupation numbers is not sufficient to access the real-space electronic density in Eq. (2). Indeed, inserting closure relations in Eq. (2) leads to

\[ n(r) = \sum_{ij} \sum_{k=1}^{N_{\text{occ}}} \langle \hat{r} | \chi_i \rangle |\chi_i\rangle \langle \varphi_k | \chi_j \rangle (\chi_j | \hat{r} \rangle, \]  

\[ = \sum_{ij} \langle \hat{r} | \chi_i \rangle |\chi_j\rangle \sum_{k=1}^{N_{\text{occ}}} \varphi_k(I) \varphi_k(J), \]  

where

\[
\frac{\varepsilon_n^I}{\varepsilon_n^J} = \frac{1}{\mu} \left[ \frac{1}{\hat{b}_{\mu}^\dagger} \hat{b}_{\mu} \right] \left( \hat{b}_{\mu}^\dagger \right) \left( \hat{b}_{\mu} \right) \left( \hat{b}_{\mu}^\dagger \right)^2
\]
where we recall that the KS computational basis set \( \{ |\psi_i \rangle \} \) is translated to quantum computers by the mapping \( |\psi_i \rangle \rightarrow |I \rangle \). Hence, inferring the real-space density requires the ability to extract the sign of the amplitudes (the KS orbital coefficients) on the quantum computer. To access to the amplitude and the sign of these coefficients, one can apply the Grover diffusion operator \[ U_G = H^{\otimes M} (2 |0 \rangle \langle 0 | - 1) H^{\otimes M}, \] (13) where \( |0 \rangle \) denotes the M-qubit zero-state in the computational basis. The role of this transformation is to realize an inversion about mean, where the mean refers to the average of the coefficients \( \overline{\varphi}_k = (1/\sqrt{2^M}) \sum_I \varphi_k(I) \) [53]. Applying this transformation on the prepared state \( |\varphi_k \rangle \) allows to infer the probabilities \( 2\overline{\varphi}_k - \varphi_k(I) \) for all \( I \) by repeated measurements. Within the direct measure of \( |\varphi_k(I)|^2 \), the correct value of \( \overline{\varphi}_k \) is identified by classically evaluating the four different values \( \pm \{ (1/2) (\pm |\varphi_k(I)|) \pm [2\overline{\varphi}_k - \varphi_k(I)] \} \), for different \( I \). From all these values, half of them are identical and correspond to \( \overline{\varphi}_k \). All together, the amplitude and the sign of the wavefunction coefficient \( \varphi_k(I) \) is given by \[ \varphi_k(I) = \frac{4\overline{\varphi}_k^2 + |\varphi_k(I)|^2 - [2\overline{\varphi}_k - \varphi_k(I)]^2}{4\overline{\varphi}_k}, \] (14) up to a unimportant global phase factor.

### III. RESULTS AND DISCUSSIONS

After the transformation of the non-interacting Hamiltonian into the auxiliary interacting one [see Eqs. (4) and (7)], we classically simulate the SS-VQE algorithm [22, 23] using the Qiskit package [54] and a state-vector simulation (without noise). The hardware efficient ansatz, the \( R_y \) ansatz, is used for \( U(\theta) \) in Eq. (9), and is given by [55]

\[
U(\theta) = \prod_{m=1}^{M} R_{y,m}(\theta_{m}^0) \prod_{n=1}^{\Gamma} U_n^{\text{ENT}}(\theta^n).
\] (15)

for a number of layers \( \Gamma \) and a number of qubits \( M \). The entanglement unitary blocks read

\[
U_n^{\text{ENT}}(\theta^n) = \prod_{m=1}^{M-1} \text{CNOT}_{m(m+1)} \prod_{m=1}^{M} R_{y,m}(\theta_{m}^{n}).
\] (16)

As a proof a concept, we study the one-dimensional Hubbard model and hydrogen chains in the following, using SOFT and regular DFT, respectively.

#### A. One-dimension Hubbard model

Starting with the one-dimensional Hubbard model, the Hamiltonian reads

\[
\hat{H} = -t \sum_{i=1}^{L} \sum_{\sigma} (\hat{c}_{i\sigma}^\dagger \hat{c}_{i+1 \sigma} + \text{h.c.}) + U \sum_{i=1}^{L} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \sum_{i=1}^{L} v_i \hat{n}_i,
\] (17)

where \( \hat{n}_i = \hat{n}_{i\uparrow} + \hat{n}_{i\downarrow} \) is the occupation number operator, with \( \hat{n}_{i\sigma} = \hat{c}_{i\sigma}^\dagger \hat{c}_{i\sigma} \), and \( t \) and \( U \) are the hopping integral and the on-site electron-electron interaction, respectively.

### B. Hydrogen chains

Let us turn to the quantum simulation of DFT (that we refer to as Q-DFT) to the \( \text{H}_8 \) linear chain in the STO-3G basis with the local density functional SVWN. This system is composed of 16 spin-orbitals and should in principle be described by 16 qubits with the Jordan–Wigner transformation. Similarly as in the previous section, this closed-shell system can be described exactly by considering the spin-\( \alpha \) or spin-\( \beta \) block of \( L = 8 \) spin-orbitals only, such that the non-interacting KS system is now mapped onto an auxiliary system of only \( M = \log_2(L) = 3 \) interacting qubits. The same ensemble weights definition is used, as well as the \( R_y \) ansatz with \( \Gamma = 4 \) layers, thus leading to \( M(\Gamma+1) = 15 \) ansatz-parameters that are also optimized with L-BFGS-B and basinhopping. The initial states in Eq. (9) are taken to be the first states in the computational basis, i.e. \( \{ |0000 \rangle, |0001 \rangle, |0010 \rangle, |0011 \rangle \} \).

In Fig. 3, we compare the ground-state energy obtained...
FIG. 2: Top panel: Relative errors in energy $E_0$. Our notation implies that the relative error in between two methods $X$ and $Y$ (referred to as $(X/Y)$ in the legend) is given by $\frac{||X|| - ||Y||}{||Y||}$ where $||X||$ denotes the Euclidean norm of $X$. Bottom panel: Ground-state energy of the uniform one-dimensional Hubbard model as a function of the interaction strength $U/t$, in units of $t$.

by Q-DFT and the ones obtained by regular DFT (on classical computer) and the full configuration iteration (FCI) method of Psi4 in the same basis, together with the relative error. As expected, the Q-DFT is on top of the DFT curve, proving that our exact mapping in Sec. II A is correct. Using the L-BFGS-B optimizer together with basinhopping led to an excellent fidelity of states compared to the occupied KS orbitals, with an error inferior to $10^{-5}$.

One of the key important aspect of this paper relies on the accuracy that one needs to achieve with our quantum algorithm. For now on, quantum computers have been promised to help solving the many-body problem with chemical accuracy (1.6 milliHartree). This goal is extremely challenging, to the point that quantum supremacy for chemistry might only be attainable in several decades. On the other hand, DFT is itself subject to approximations coming from the XC functional. Hence, the accuracy that Q-DFT has to reach is bounded by this functional-driven error. As seen by the logarithmic scale on the top panel of Fig. 3, the relative error between Q-DFT and DFT is three- or four-order of magnitude below the error of DFT compared to FCI. Note also that bigger is the system, larger is the error between the DFT and FCI total energy. This functional-driven error allows to bypass the burden of reaching chemical accuracy, such that Q-DFT appears much more achievable with noisy devices than quantum algorithms for wavefunction theory.

The validity and applicability of the concept being proved, the “numerical” efficiency and potential computational gain should be discussed. In SOFT and regular DFT, the number of probability amplitudes $|\psi_k(I)|^2$ or $\varphi_k(I)$, respectively, to measure is exponential in the number of qubits and linear in the number of spin-orbitals of the original system, i.e. it scales as $O(2^M) = O(L)$. In addition, the number of expectation values to estimate on the quantum computer is proportional to the number of terms in Eq. (4), thus scaling as $O(4^M) = O(L^2)$. Owing the sparse nature of the KS Hamiltonian, with a sparsity that is at minima higher than 90% [57], this estimation represent a largely overestimated bound. Although these scalings question a quantum advantage for SOFT or regular DFT, in principle the evaluations of the expectation values and probability amplitudes are completely independent and straightforwardly parallelisable on multiple quantum computers. Within this strong assumption and the large overestimation of scaling bounds, these operations can be omitted in the estimation of the complexity, and the limiting step to implement SOFT and DFT on a quantum computer will be dictated by the gate complexity. Finally, one could also argue that the number of parameters within SSVQE should also scale as $O(2^M)$ to prepare the states exactly. However, similarly to standard VQE for molecules, it is sufficient to prepare approximate states as long as the results of the approximation remain within a given accuracy (i.e., the functional-driven error). All together, Q-DFT can certainly be advantageous providing optimizations of both quantum and classical algorithms.

Let us now focus on the gate complexity, that is related to the number of orbitals and the ansatz operator in the
SS-VQE simulation. Considering a system of $M$ interacting orbitals, the different variants of the UCC ansatz would typically scale as $O(M^4)$ (related to the number of terms in the ansatz operator). However, as our auxiliary system of $M$ interacting pseudo-orbital is not physical anymore, no physically-inspired ansatz is required and one can focus on the hardware efficient ansatz described in Eq. (9) instead, which features shallower circuit at the expense of more variational parameters given to the classical optimizer. The $R_g$ ansatz features $\Gamma(M-1)$ two-qubit gates, thus leading to a gate complexity that scales as $O(M^2)$ (assuming that the number of layers grows linearly with $M$). Turning to regular DFT, the Grover diffusion circuit only add one multi-controlled qubit gate to the full circuit, for which efficient implementations can be found in Refs. [58, 59]. Summing up, as $L = 2^M$, the gate complexity required to diagonalize the KS Hamiltonian on quantum computers is about $O(\log_2(L)^2)$ using hardware efficient ansatz within SS-VQE. In comparison with the $O(L^3)$ complexity on classical computers for this operation, a substantial speed-up can yet be envisioned, in particular for SOFT. For regular DFT, still remains an $O(L^3)$ complexity operation to be performed on classical computers at each KS iteration to compute the real-space density matrix, see Eq. (12). Nevertheless, a quantum advantage can still be envisioned as the number of $O(L^3)$ operations can be drastically reduced. Given the potential lower complexity of Q-DFT compared to and DFT, especially for their lattice version, and the fact that one doesn’t aim to reach chemical accuracy, it might be the nearest-NISQ-era application of quantum computers for quantum chemistry or condensed matter physics.

IV. CONCLUSIONS

In this work, we show that mean-field-type approaches such as DFT are also expected to benefit from the advance of quantum technologies, opening the door for quantum simulations of systems having millions of orbitals. As a proof of concept, we simulated the one dimensional Hubbard model with a quantum algorithm implementation of site occupation functional theory, the lattice version of DFT, for which an exponential speed-up can be envisioned, assuming that parallel quantum computing is accessible. Then, the quantum analog of regular DFT was successfully applied to hydrogen chains. Though an exponential speed-up is not expected due to the reconstruction of the real-space density, solving the KS self-consistent equation on a quantum device might still lead to quantum advantage. Moreover, a significant advantage of performing DFT on quantum computer compared to WFT is the level of accuracy that one needs to achieve. Because, WFT target chemical accuracy, the quantum advantage for WFT and quantum chemistry in the NISQ era is currently surrounded by skepticism. However, DFT has its own level of approximations coming from the approximate xc functional. This error being orders of magnitude larger than chemical accuracy, we can expect DFT on quantum computers to be much more resilient to noise than WFT and to be the nearest-term application of quantum computer for not only quantum chemistry, but also condensed-matter physic and even biology.

Appendix A: Phase estimation algorithms

The quantum phase estimation (QPE) algorithm is composed of two qubit-registers, one called the ancilla register and the other the system register. The first step is to initialize the system register to a state $|\varphi_{\text{ini}}\rangle = \sum_{k=0}^{M-1} a_k |\varphi_k\rangle$, where $\{|\varphi_k\rangle\}$ are the eigenvectors of a targeted unitary operator. In our case, this operator is the propagation operator $U = e^{-iH^{\text{aux}}t}$, where $H^{\text{aux}}$ is defined in Eq. (4). Hadamard gates are also applied to all ancilla qubits to create a equi-weighted superposition of the computational basis states, such that the total state of the QPE circuit reads

$$|\Psi^{\text{QPE}}\rangle = \frac{1}{\sqrt{n!}} \sum_{x=0}^{2^n-1} \sum_{k=0}^{M-1} a_k |x\rangle |\varphi_k\rangle,$$  \hspace{1cm} (A1)

where $x$ denote the integers associated to the binary strings formed by the $n$ ancilla qubits. Then, a phase-kickback is performed by applying a sequence of controlled-$U$ gates between the system and ancilla registers, such that the QPE state becomes

$$|\Psi^{\text{QPE}}\rangle = \frac{1}{\sqrt{n!}} \sum_{x=0}^{2^n-1} \sum_{k=0}^{M-1} a_k e^{ix\varepsilon_k t} |x\rangle |\varphi_k\rangle$$ \hspace{1cm} (A2)

Finally, the inverse quantum Fourier transform is applied on the ancilla register, thus leading to

$$|\Psi^{\text{QPE}}\rangle = \sum_{k=0}^{2^M-1} a_k |\varepsilon_k t \rangle |\varphi_k\rangle$$ \hspace{1cm} (A3)

Reading out the ancilla-register state gives an estimation of the $k$th KS orbital energy $|\varepsilon_k t\rangle$ with $t$ such that $\varepsilon_k t \in [0, 2\pi]$, and projects the system register to the KS orbital $|\varphi_k\rangle$ with probability $|a_k|^2$.

A more near-term algorithm is the so-called iterative phase estimation algorithm (IPEA) [5], a single-ancilla version of QPE where the phases $\varepsilon_k t$ and amplitudes $|a_k|^2$ are inferred by classical post-processing instead of being directly measured on the ancilla register. In this algorithm, the controlled-$U$ gate is applied $j$ times and the inverse quantum Fourier transform is replaced by a tomography gate on the ancilla qubit [9, 24]. Repeated tomography at various $j$ allows to construct the function

$$g(j, t) = \sum_{k=0}^{2^M-1} |a_k|^2 e^{ij\varepsilon_k t},$$ \hspace{1cm} (A4)

where $\varepsilon_k t$ and $|a_k|^2$ can be inferred indirectly through classical post-processing using Prony’s or Bayesian methods.
In both versions, the square of the amplitudes $|a_k|^2 = |\langle \phi_{\text{ini}} | \phi_k \rangle|^2$ are directly related to the square of the KS orbital coefficients by taking $|\phi_{\text{ini}}\rangle = |I\rangle$, such that $|a_k|^2 = |\varphi_k(I)|^2$. To calculate all the occupations in Eq. (11), one therefore has to repeat the QPE or IPEA algorithm for all computational states $\{|I\rangle\}$ as initial state.

Within QPE and IPEA, the gate complexity is related to the number of terms in the Hamiltonian which scales as $O(2^{2M}) = O(L^2)$ according to Eq. (4), though this Hamiltonian is generally very sparse [57] and the scaling can be reduced using low rank representation and factorization techniques [29, 60, 61]. Compared with the $O(L^3)$ scaling on classical computers, an improvement of only one order of magnitude is expected for algorithms based on Hamiltonian simulations for the lattice version of DFT (SOFT). For regular DFT, the Grover diffusion operator can be applied to the system register of the multiple-ancilla QPE circuit, as described in Sec. II B to extract the real-space density. Then, a classical operation that scales as $O(L^3)$ is required as readily seen in Eq. (12). Within IPEA, the system register is not projected to a state $|\varphi_k\rangle$ so that no straightforward implementation of Q-DFT is expected.

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