Measurement on quantum devices with applications to time-dependent density functional theory

Jun Yang, James Brown, James Daniel Whitfield
Dartmouth College, Department of Physics and Astronomy, Hanover NH, 03755
(Dated: September 10, 2019)

Quantum algorithms are touted as a way around some classically intractable problems such as the simulation of quantum mechanics. At the end of all quantum algorithms is a quantum measurement whereby classical data is extracted and utilized. In fact, many of the modern hybrid-classical approaches are essentially quantum measurements of states with short quantum circuit descriptions. Here, we compare and examine three methods of extracting the time-dependent one-particle probability density from a quantum simulation: direct Z-measurement, Bayesian phase estimation and harmonic inversion. We have tested these methods in the context of the potential inversion problem of time-dependent density functional theory. Our test results suggest that direct measurement is the preferable method. We also highlight areas where the other two methods may be useful and report on tests using Rigetti’s quantum device. This study provides a starting point for imminent applications of quantum computing.

Simulating quantum systems on a classical computer is a difficult problem even for a supercomputer due to the fact that the Hilbert space grows exponentially with the system size. A universal quantum computer is believed to be the solution of the difficulty, where it is known that a wide class of physical systems can be simulated efficiently on a quantum computer.

Density functional theory (DFT) is a powerful tool in simulating condensed matter systems. DFT converts the problem of solving a many-particle system to the problem of solving a non-interacting system with a new scalar potential. The additional potential term in the non-interacting system is known as the Kohn-Sham potential. In the Kohn-Sham (K-S) system, the calculation of an exchange-correlation term is required, which can only be obtained by approximation methods given the computational complexity of its computation. In the article, a method of combining simulations on a quantum computer and classical DFT calculation is proposed, the exchange-correlation term can be obtained from a quantum computer to enhance the accuracy of DFT calculation.

As an extension of DFT, time-dependent density functional theory (TDDFT) is widely used in finding the dynamics of the system when time-dependent potentials are present. Similar to DFT, TDDFT uses the time dependent K-S system where again the key hurdle is constructing the required K-S potentials. We call the task of constructing such K-S potentials when given the time-evolution of the on-site probability density, the K-S potential inversion problem. In article, a scheme of solving the K-S potential inversion problem utilizing a quantum computer was proposed. We have recently returned to this proposal with improved numerical methods for inverting the potential. To obtain the K-S potential, we need to get the density of the time evolved many-particle system using a quantum computer. In this paper, we will present three different methods of measuring the density operator on a quantum computer and compare the performance of the methods.

An outline for the remainder of the article is as follows: Next, we discuss the phase estimation approach to measurement. Then we describe the circuit implementation for measuring the on-site fermionic density. Qubit descriptions for the fermionic operator are explained before turning to the illustration of a two-electron test. The three schemes for extracting the density are tested numerically and compared. We have also performed preliminary tests of these techniques on Rigetti’s quantum computer and report briefly on our results. Finally, we draw conclusions to close the manuscript.

Phase estimation.— The clearest understanding of the quantum simulation paradigm is given by the view that spectra are Fourier transforms of auto-correlation functions e.g. $f(t) = \langle \psi | \psi(t) \rangle$. In a time-dependent approach to quantum mechanics, the spectrum is given by

$$\varepsilon(\omega) \propto \int dt \ f(t) e^{i\omega t} = \int dt \ \langle \psi | U(t) | \psi \rangle e^{i\omega t} \propto \sum |c_j|^2 \int dt \ e^{i(\omega-E_j)t} \propto \sum |c_j|^2 \delta(\omega - E_j) \quad (1)$$

Here $U(t)$ is a unitary operator given by $U(t) = e^{-iHt} = \sum e^{-iE_k t} |k\rangle \langle k|$ and $\psi = \psi(0) = \sum c_k |k\rangle$.

We can apply this same view to phase estimation. The first Hadamard followed the controlled unitary gate is used to impart a relative phase such that the initial state $|0\rangle \langle \psi|$ which then evolves into $\frac{1}{2} (|0\rangle \langle \psi| + |\psi(t)\rangle) + \frac{i}{2} (|1\rangle \langle \psi| - |\psi(t)\rangle)$. Before measurement, the final Hadamard transform creates a state proportional to $|0\rangle (|\psi| + |\psi(t)|) + |1\rangle (|\psi| - |\psi(t)|)$. The bracketed terms are the $k = 0$ and $k = 1$ points of the two-bit discrete Fourier transform respectively. Measurement probabilities are then given by

$$P(0|t) = \frac{1}{2} + \frac{1}{2} (\langle \psi(t) | \psi \rangle + \langle \psi | \psi(t) \rangle) \quad (2)$$
where operators \{a_j\} follow \(a_j^\dagger a_p + a_p a_j^\dagger = \delta_{pq}\) and \(a_p a_q = -a_q a_p\). The circuit in Fig. 1 does not explicitly include the state preparation but can be included as part of \(U(t)\) if required. The latter half is a phase estimation circuit \[12\] where \(U_2(\tau) = e^{-iH_2\tau}\) where \(H_2\) is the observable to be measured.

For a general state, the phase estimation algorithm yields the eigenvalues \(E_j\)'s with probability \(|c_j|^2\). To be precise, for a general state \(|\psi\rangle = \sum_k c_k |k\rangle\), where \(|k\rangle\)'s are the eigenvectors of the Hamiltonian \(H\), the probability of measuring zero on the top register

\[
P(0|\tau, t) = \sum_k |c_k(\tau)|^2 \cos^2 \left( \frac{E_k \tau}{2} \right)
= \frac{1}{2} + \frac{1}{4} \sum_k |c_k(\tau)|^2 (e^{iE_k \tau} + e^{-iE_k \tau})
\]

In this article, we only consider \(H_2 = n_j = a_j^\dagger a_j\) in order to measure the local on-site density at site \(j\). The eigenvalues of \(n_j = a_j^\dagger a_j\) are \(\lambda = 0, 1\) so that the wave function after evolution under \(U(t)\) is given by \(\psi(t) = c_0(t) |\psi_{n_j=0}\rangle + c_1(t) |\psi_{n_j=1}\rangle\). The expectation value of the density is given by

\[
\langle n_j(t) \rangle = \langle \psi(t) | a_j^\dagger a_j | \psi(t) \rangle = |c_1(t)|^2
\]

**Qubit Encoding.** To implement the evolution and phase estimation algorithm on a quantum computer, we need to encode the Hamiltonians into qubits. Jordan-Wigner (JW) transformation is a standard way to encode a fermionic system of \(M\) orbitals into \(M\) qubits using a tensor product of Pauli operators

\[
a_p = \frac{1}{2} (X_p + iY_p) Z_1 Z_2 \ldots Z_{p-1}
\]

\[
a_p^\dagger = \frac{1}{2} (X_p - iY_p) Z_1 Z_2 \ldots Z_{p-1}
\]

With these transformations, the fermionic Hamiltonian can be encoded into qubit representations. Thus, the Hamiltonian can be written as \(H = \sum_i h_i\), where all the \(h_i\)'s are in terms of qubit operators.

There is not an easy way to construct arbitrary unitary operators on a quantum computer. In order to simulate the propagator \(U(t)\), we applied Trotter decomposition.

\[
U(t) = e^{-iHt} \approx \left( e^{-iH_1t/N} e^{-iH_2t/N} \ldots e^{-iH_Nt/N} \right)^N
\]

Each term in the decomposition above is straightforward to simulate on a quantum computer \[13\].

**Two-electron test system.** The two-electron system that we tested on is the four-spin-orbital HeH\(^+\) model with parameters matching the example found in Ref. \[14\]. The basis functions are orthogonalized and then transformed such that the one-body Coulomb matrix is diagonal. This transformation was chosen so that a corresponding scalar time-dependent Kohn-Sham potential could be calculated using for this system using the method of Reference \[9\]. The initial state at \(t = 0\) places two electrons in the first two modes of opposite spin. This state is obtained by employing two X-gates to prepare \(\psi(0) = |1100\rangle\).

Using Rigetti’s quantum virtual machines \[15\], we then evolve the system under its Hamiltonian for times less than three atomic units. The propagation is implemented via first-order Trotterization with three time steps. To reduce the Trotter error in evolution, either a shorter Trotter step or a higher order Trotter approximation must be used \[16\]. This means more quantum gates are needed, making it hard to be implemented on a near term device. Additional sources of error are associated with finite sampling from the binomial distribution and the error associated with the inference steps. To make the virtual machine slightly closer to a real quantum computer, in all methods below, measurement noise was added into the system, giving \(1\%) probability of flipping the qubit. It should be noted that the quantum noise found on the actual device was much higher.

In Figs. 2 and 3, we used 3000 quantum measurement samples per time point. For the plot of harmonic inversion depicted in Fig. 3, a total of 120,000 quantum measurements occur for the density at each time point. This is because there were 40 equally spaced \(\tau\)-points and 3000 quantum measurements were used per fixed \(\tau\). In all three of these plots, the density according to the Trotterize evolution is plotted in pink with a fading color region representing the decay of the binomial distribution with parameters \(p = n(t), n_{\text{trials}} = 3000\).

**Method 1: Z-basis Measurement.** In the first of the three methods investigated, we rely on the fact that the Jordan-Wigner transformation of the on-site density operator has a simple form \(a_p^\dagger a_p = (1 - Z_p)/2\). Thus, we can directly measure the local density operator by measuring

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**Figure 1.** The circuit for measuring the density matrix. The half before the dashed line is used for evolving the state, the half after is used for doing the density measurement.
For an arbitrary wave function $|\psi(t)\rangle = c_0(t) |\psi_n_p=0\rangle + c_1(t) |\psi_n_p=1\rangle$, where $\psi_n_p$ denotes the state projected into the subspace where the $p$-th qubit is in state $n_p$. Both amplitudes can be obtained from the measurement giving $\langle Z_p(t) \rangle = |c_0(t)|^2 - |c_1(t)|^2$.

By repeating the measurement many times at each value of $0 \leq t \leq 3$, we obtain the expectation value of the density. The results based on 15 equally spaced time-points with 3000 measurements at each fixed time are shown in Fig. 2. The exact time evolution of the density is also shown in the figure for comparison along with error bars of 2$\sigma$ reflective of the $N=3000$ sample variance of the binomial distribution.

The simplicity of this measurement approach reduces the classical runtime to the lowest of the three methods compared, and the convergence of the error bars is faster than the Bayesian measurement discussed later.

Method 2: Harmonic Inversion.— Harmonic inversion is a technique of extracting the amplitudes $A_j$, frequencies $f_j$, phases $\phi_j$ and exponential decay constants $\alpha_j$ out of a signal,

$$f(\tau) = \sum_j A_j e^{-i(2\pi f_j \tau - \phi_j)} e^{-\alpha_j \tau}$$

which is evenly sampled [17] [18]. The signal reconstructed from harmonic inversion has the same form as the probability $P(0|\tau,t)$ except for the decaying term which is negligible in real implementation. The decay of the signal could be used to represent the decaying fidelity of the Trotter approximation with the target evolution operator but this decay is a priori unknown. By comparing the form of the reconstructed signal with the probability, we can obtain the density from the reconstructed signal.

The results of density measurement through harmonic inversion are shown in Fig. 3. Each point in Fig. 3 was computed through harmonic inversion using the HarmInv package [19]. Because the local density operator $a_p^\dagger a_p$ only has eigenvalues zero and one, the measurement outcome has a simple form

$$P(0|\tau,t) = A_0(t) + A_1(t) \left(e^{-i2\pi f \tau} + e^{i2\pi f \tau}\right)$$

where $A_0(t) = \frac{1}{2}(2 - |c_1(t)|^2)$, $A_1(t) = |c_1(t)|^2/4$, and $f = 1/2\pi$. One example of the reconstruction is shown in Fig. 4.

Method 3: Bayesian inference.— Bayesian inference can be used to estimate the density as well. As a powerful tool of making inferences, Bayesian inference has
wide applications. We applied Bayesian inference to infer the unknown parameters in a quantum system which, in our case, is the on-site density. The density estimation was implemented via sequential Monte Carlo (SMC) \[20\]. This method requires the most communication between the classical and quantum processors since the SMC suggests each \(\tau\)-point based on the previous outcomes. The Bayesian experimental design is based on the implementation found in the QInfer package \[21\]. Bayesian inference gives the probability distribution of a parameter over the parameter space. The final decision is made according to the posterior probability \(P(\theta|d_1, d_2 \ldots d_N)\), where \(\theta\) is the parameter we want to estimate, \(d_i\)'s are the outcome of each measurement. In the present application \(\theta \equiv \langle n_j(t) \rangle\).

Recall the Bayesian rule, the posterior probability is updated by carrying out experiments sequentially,

\[
P(\theta|d_1, d_2, \ldots d_N) \propto \prod_{i=1}^{N} P(d_i|\theta)P(\theta)
\]

where \(P(\theta)\) is the prior probability, \(P(d_i|\theta)\) is the likelihood function.

The likelihood function is the information we know about the parameter before conducting any experiments. Because we know nothing before the experiment, we can initialize the prior with a uniform distribution over the parameter space. For the phase estimation circuit of Fig. 1, the likelihood function is given by

\[
P(d_1 \langle n_j(t) \rangle \tau) = \frac{1}{2} + \frac{(-1)^d}{4} \langle \psi(t)|[U_2(\tau) + U_2^\dagger(\tau)]|\psi(t)\rangle
\]

(12)

where \(U_2(\tau) = \exp(-i\tau a_j^\dagger a_j)\) and \(d = 0\) or 1. Note, when \(d = 0\) we recover Eq. (2). With this we can rewrite the likelihood function as

\[
P(d_1 \langle n_j(t) \rangle \tau) = \delta_{d,0} + \frac{(-1)^d}{2} (\cos \tau - 1) \langle n_j(t) \rangle
\]

(13)

This can be compared with Eq. (4) in the case that \(d = 0\). The results of Bayesian inference are shown in Fig. 5. Bayesian inference has good performance within a wide range of the time domain except at the boundary of the estimate domain e.g. when the density is one or zero. This is based on numerical evidence since the majority of the points at near the boundary of the estimation domain needed to be discarded when cleaning the data as discussed below.

Unlike harmonic inversion, \(\tau\) in the phase estimation circuit is not required to be evenly spaced. Another advantage of Bayesian inference is that we do not need to know the exact form of the function to be estimated a priori. Bayesian inference could also be applied to estimate more general parameters.

\textit{Comparison}

Figure 5. The expectation values of \(a_1^\dagger a_1\) measured via Bayesian inference versus exact Trotter solution. The first point shows a large deviation from the Trotter solution typical of behavior whenever the exact density’s value is close to one. Error bars are twice that of the standard errors.

To quantify the accuracy of these density extraction methods, we employ the \(L_1\) norm to measure the deviation from the Trotter solution. For discrete data points, the deviation is given by the loss function on the density at the first site: \(L = \sum_k |\tilde{n}_1(t_k) - n_1(t_k)|/N\).

Fig. 6 shows how the loss function scales with the number of trials for each of the three approaches. The con-
verge rate for determining the bias of a coin would be 0.5 but here additional measurement error has been introduced into the model which prevents \( L = 0 \) situation even with an infinite number of samples. Further, the Bayesian and harmonic inversion techniques sometimes reported anomalously poor estimates of the density at a given time. A single fluctuation of this type along the time trace of the density entirely dominates the loss function. For the sake of comparison, we have cleaned the data by removing anomalous density values that are too far from the exact solution. Although this is an \textit{ad hoc} procedure that requires knowing the exact answer, we have tested our data at various levels of cutoff finding that at any fixed cutoff harmonic inversion had the most points discarded and consistently displayed marginally faster convergence rates.

Regardless of the possible improvement in convergence, it should be reminded that the harmonic inversion technique uses many quantum computer queries to estimate \( P(0|\tau, t) \) at variable \( \tau \) before inferring the density at a fixed time \( t \). In comparing the three methods, all require time evolution of the system wave function to time \( t \). In the harmonic inversion and Bayesian estimation techniques, additional gates are needed for the \( \tau \) propagation under the observable for density. The difference between queries in harmonic inversion and Bayesian inference is the selection of the \( \tau \) parameter in \( U_\tau(\tau) \).

While the convergence rates are all approximately the same, it is clear that the \( Z \)-basis measurement has the best performance in terms of the number of queries of the quantum computer. In the case considered here, the direct \( Z \) measurements are convenient for the Jordan-Wigner encoding. In other circumstances with different fermion-to-spin transforms, the direct measurement technique may not be as fruitful. For existing and near-term quantum devices, the constraints of low circuit depth suggests direct measurement of the \( Z \) operators as the best path forward when using Jordan-Winger transformed qubit Hamiltonians.

The runtime of these three methods also varies. Since direct \( Z \)-measurements are the simplest from an inference point of view, the classical computation time is also the least. Bayesian inference requires many steps for the sequential Monte Carlo to converge \([20]\). Consequently, this method used the longest amount of classical computational time. Although harmonic inversion uses 40 times more measurement per time-point, it is interesting to note that it only took an intermediate amount of classical processing time.

\textit{Conclusions.—} We have tested three different methods of measuring the on-site density operator for a toy model inspired by TDDFT. We were able to conclude that direct \( Z \) measurements obtains the best estimates of the on-site density for a given number of quantum computer queries. This is based on the use of the Jordan-Wigner transform and simulated measurement noise. Of course, we could have considered other fermion-to-spin transforms which lead to different encodings of the \( a_i^\dagger a_i \).

For improving our noise models, we can do no better than testing our circuits on current and future quantum devices. We tested our circuits on Rigetti’s quantum device but found that the loss function depends heavily on which qubits are used as well as the permutation of qubit labels within the circuit. Time evolution under the full Hamiltonian did not return any signal even when using only one first-order Trotter step. We therefore resorted to using a truncated Hamiltonian which included the one-body Hamiltonian and only the Coulomb-like \((h_{ijji})\) terms of the two-body Hamiltonian. After encoding and exponentiation, this Hamiltonian results in 36 universal gates and compiled non-deterministically to approximately 200 allowable gates on the Rigetti device. Due to decoherence, only a weak signal was present where amplitudes recovered were between three and twenty percent of the exact solution. The recovered amplitude depended mostly on qubit selection but also changed run-to-run. The frequency and sinusoidal shape of the signal was recovered more reliably. In our present study, the eigenenergies were not interesting but we suspect that problems that depend on the frequencies may be more successfully calculated on the current Rigetti device.

We plan to continue our inquiry into the TDDFT potential inversion problem using existing and forthcoming quantum technology. Tasks that avoid QMA-hard state preparation problems will continue to be of interest to those looking for new applied areas of quantum computation.

\textbf{Acknowledgements.—} JY, JB and JDW were supported by the U.S. Department of Energy, Office of Science, Office of Advanced Scientific Computing Research, under the Quantum Computing Application Teams program (Award 1979657). JDW was also supported by the NSF (Grant 1820747) and additional funding from the DOE (Award A053685). Calculations were performed using Dartmouth’s Discovery Linux HPC Cluster.

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