Charge and spin response functions on a quantum computer: applications to molecules

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We propose a scheme for the construction of charge and spin linear-response functions of an interacting electronic system via quantum phase estimation and statistical sampling on a quantum computer. By using the unitary decomposition of electronic operators for avoiding the difficulty due to their non-unitarity, we provide the circuits equipped with ancillae for probabilistic preparation of qubit states on which the necessary non-unitary operators have acted. We perform simulations of such construction of the response functions for C\(_2\) and N\(_2\) molecules by comparing with the accurate ones based on the full configuration interaction calculations. It is found that the accurate detection of subtle structures coming from the weak poles in the response functions requires a large number of measurements.

I. INTRODUCTION

Since the information carrier of a programmable quantum computer is a set of qubits that exploits the principle of superposition, essentially parallel algorithms can exist and perform computation for classically formidable problems.[1, 2] Quantum chemistry[3] is believed to be one of the most suitable research fields for quantum computation since its problem setting is quantum mechanical by definition. Indeed, a quantum computer can treat a many-electron state composed of lots of Slater determinants as it is in a sense that the electronic state is encoded as a superposition of qubit states via an appropriately chosen map such as the Jordan–Wigner (JW)[4] and Bravyi–Kitaev (BK)[5] transformations.

The quantity which a quantum chemistry calculation is asked to first provide is the total energy of a target system.[6] One of the most widespread methods for obtaining the total energy is the variational quantum eigen-solver (VQE), in which a trial many-electron state is prepared via a parametrized quantum circuit. The parameters are optimized iteratively with the aid of a classical computer aiming at the ground state. This approach was first realized[7] by using a quantum photonic device, after which the realizations by superconducting[8, 9] and ion trap[10] quantum computers have been reported. There exist algorithms for obtaining the energy spectra of excited states.[11–16]

Not only academic interest but also industrial demands for accurate explanations and predictions of material properties make it an urgent task to develop methodologies on quantum computers for various electronic properties other than energy levels. The one-particle Green's functions (GFs) are important in correlated electronic systems[17, 18] since they are often used in explanation of spectra measured in photoemission and its inverse experiments. Recently we proposed a method for the GFs via statistical sampling[19] by employing quantum phase estimation (QPE).[1] Endo et al.,[20] on the other hand, proposed a method for the GFs by focusing on noisy intermediate-scale quantum (NISQ) devices.

The charge and spin response functions \(\chi\), formulated in the linear-response theory,[21, 22] describe the leading contributions to the electric and magnetic excitations when perturbation fields are applied to a target system. Since the response functions are the fundamental building blocks in constructing the elaborated methods for correlated electrons such as GW theory,[21, 23] the accurate calculation of them is needed.

Given the recent rapid development of fabrication techniques for quantum hardware and the growing demands for quantum computation in material science, it is worth making tools for analyses on correlation effects. In this study, we propose a scheme for the construction of the response functions of an interacting electronic system via statistical sampling on a quantum computer. For examining the validity of our scheme, we perform simulations of such construction of the response functions of diatomic molecules by referring to the full configuration interaction (FCI) results.

This paper is organized as follows. In Section II, we explain the basic ideas and our scheme in detail by providing the quantum circuits for obtaining the response functions via statistical sampling. In Section III, we describe the computational details for our simulations on a classical computer. In Section IV, we show the simulation results for C\(_2\) and N\(_2\) molecules. In Section V, we provide the conclusions.

II. METHODS

A. Definitions

I. Charge and spin response functions

We work with the \(n_{\text{orbs}}\) orthonormalized spatial orbitals for each spin direction in a target \(N\)-electron system, which we assume is in the ground state \(|\Psi_{\text{gs}}\rangle\) at zero temperature. The formalism described below can be easily extended to a system with multiple ground states.
and/or a nonzero temperature.

By using the creation $a^\dagger_{p\sigma}$ and annihilation $a_{p\sigma}$ operators for the $p$th spatial orbital of $\sigma$ spin ($\sigma = \alpha, \beta$), the electron number operator is given by $n_{p\sigma} = a^\dagger_{p\sigma}a_{p\sigma}$. The spin operator is given by $\mathbf{s}_p = \sum_{\sigma} a^\dagger_{p\sigma} \sigma_{\sigma\sigma'} / 2 a_{p\sigma'}$, where $\sigma^{el}$ is the Pauli matrix for the electronic state.

The response functions in terms of charge and spin in time domain for $\mu, \mu' = n, x, y, z$ are defined as \cite{21]

$$\chi_{pp',\mu'\mu}(t, t') \equiv -i\delta(t - t') \langle [O_{pp}(t), O_{p'\mu'}(t')] \rangle \quad (1)$$

$O_{pp}(t)$ is the operator in the Heisenberg picture for $O_{pn} \equiv \sum_n n_{p\sigma}$ and $O_{pj} \equiv s_{pj} (j = x, y, z)$. To rewrite the expression for the response functions into a more tractable form, we define the charge-charge transition matrix element

$$N_{\lambda\sigma,\sigma',\lambda'} \equiv \langle \Psi_{\lambda}\rangle n_{p\sigma}\langle \Psi_{\lambda'}|n_{p'\sigma'}\rangle \langle \Psi_{\lambda}\rangle \quad (2)$$

for the $\lambda$th energy eigenstate $|\Psi_{\lambda}\rangle$ of the $N$-electron states. We define similarly the spin-spin one

$$S_{\lambda\lambda'} \equiv \langle \Psi_{\lambda}|s_{pj}\langle \Psi_{\lambda'}|s_{p'j'}\rangle \langle \Psi_{\lambda}\rangle \quad (3)$$

for $j, j' = x, y, z$ and the spin-charge one

$$M_{\lambda\lambda',\sigma,\sigma'} \equiv \langle \Psi_{\lambda}\rangle s_{pj}\langle \Psi_{\lambda'}|n_{p'\sigma'}\rangle \langle \Psi_{\lambda}\rangle \equiv M_{\lambda\lambda',\sigma,\sigma'}^\dagger \quad (4)$$

From these matrix elements, we define the following Hermitian matrices $L_{\lambda}$:

$$L_{\lambda\sigma,\sigma',\lambda'} \equiv \sum_{\sigma,\sigma'} N_{\lambda\sigma,\sigma',\lambda'} \quad (5)$$

$$L_{\lambda\lambda'} \equiv S_{\lambda\lambda'} \quad (6)$$

$$L_{\lambda\lambda',\sigma,\sigma'} \equiv \sum_{\sigma} M_{\lambda\lambda',\sigma,\sigma'} \quad (7)$$

which are involved in the Lehmann summation over the $N$-electron energy eigenstates

$$R_{pp',\mu'\mu}(z) \equiv \sum_{\lambda} \frac{L_{\lambda p p', \mu' \mu}}{z - (E_{\lambda}^N - E_{\text{gs}}^N)} \quad (8)$$

for a complex frequency $z$. $E_{\text{gs}}^N$ is the ground-state energy and $E_{\lambda}^N$ is the energy eigenvalue of $|\Psi_{\lambda}\rangle$. Since the response functions defined in eq. (1) depend on time only via $t - t'$ in this case, their expressions in frequency domain are written as

$$\chi_{pp',\mu'\mu}(\omega) = R_{pp',\mu'\mu}(\omega + i\delta) + R_{pp',\mu'\mu}(\omega - i\delta) \quad (9)$$

for a real frequency $\omega$. The positive infinitesimal constant $\delta$ appears due to the retarded nature of the response functions, rendering all the poles immediately below the real axis. It is clear that the real part of $\chi_{pp',\mu'\mu}(\omega)$ is even with respect to $\omega$, while the imaginary part is odd.

2. Unitary decomposition of electronic operators

Although there are alternatives for mapping the electronic operators of a target system to the qubit ones such as JW\cite{4} and BK\cite{5} transformations, we do not distinguish between an electronic operator and its corresponding qubit representation in what follows since no confusion will occur for the readers.

For each combination of a spatial orbital $p$ and a spin $\sigma$, we can define the following operators for the qubits:\cite{19]

$$U_{0\sigma} = a_{p\sigma} + a^\dagger_{p\sigma} \quad (10)$$

and

$$U_{1\sigma} = a_{p\sigma} - a^\dagger_{p\sigma} \quad (11)$$

which are unitary regardless of the adopted qubit representation thanks to the anti-commutation relation between the electronic operators and can thus be implemented as logic gates in the quantum computer. This means that we can prepare at least probabilistically an electronic state on which an arbitrary product of the creation and annihilation operators has acted, similarly to the case for GFs.\cite{19}

In what follows, we assume that the many-electron ground state $|\Psi_{\text{gs}}\rangle$ is already known and can be prepared on a quantum computer.

B. Charge-charge contributions

Let us first consider the determination of the charge-charge transition matrices $N_{\lambda}$.

1. Circuits for diagonal components

From the unitary operators in eqs. (10) and (11) for a combination of a spatial orbital $p$ and a spin $\sigma$, we define

$$U_{\kappa\kappa'\sigma} = U_{\kappa\sigma}U_{\kappa'\sigma} \quad (12)$$

for $\kappa, \kappa' = 0, 1$, which are also unitary. With them, the electron number operator is written as

$$n_{p\sigma} = U_{0\sigma}^\dagger + U_{1\sigma}^\dagger - U_{0\sigma}^\dagger - U_{1\sigma}^\dagger \quad (13)$$

while the hole number operator is written as

$$\tilde{n}_{p\sigma} \equiv 1 - n_{p\sigma} = \frac{U_{0\sigma}^\dagger - U_{0\sigma}^\dagger + U_{1\sigma}^\dagger - U_{1\sigma}^\dagger}{4} \quad (14)$$

We construct a circuit $C_{p\sigma}$ equipped with two ancillary qubits $|q_0^A\rangle$ and $|q_1^A\rangle$ by implementing the controlled operations of $U_{\kappa\kappa'\sigma}$, as depicted in Fig. 1. The whole system consists of the ancillae and an arbitrary input
probabilistic preparation of the register as a circuit. The partial measurement is defined as follows:

\[ \text{prob.} \left| \left| n_{\text{par}} \right| \right| ^2 = P_{n_{\text{par}}}, \]

since \( a_{\text{par}}^2 \) and \( a_{\text{par}}^2 \) vanish due to the Fermi statistics. The projective measurement on \( \left| q_0^A \right\rangle \) is represented by the two operators \( P_q = I \otimes |q\rangle \langle q| \otimes I \) \((q = 0, 1)\). The state of the whole system collapses immediately after the measurement as follows:

\[ \text{prob.} \left| \left| n_{\text{par}} \right| \right| ^2 = \frac{1}{2}, \]

\[ \text{prob.} \left| \left| n_{\text{par}} \right| \right| ^2 = \frac{1}{2}, \]

2. Circuits for off-diagonal components

For mutually different combinations \((p, \sigma)\) and \((p', \sigma')\) of spatial orbitals and spins, we define the four non-Hermitian auxiliary operators

\[ n_{\text{par}, p' \sigma'}^\pm = \frac{n_{\text{par}} \pm e^{i\pi/4} n_{p' \sigma'}}{2}, \]

and

\[ \tilde{n}_{p, \sigma'}^\pm = \frac{n_{p} \pm e^{i\pi/4} n_{p' \sigma'}}{2}. \]

Unnormalized auxiliary states \( |\Psi_{p, \sigma'}^\pm \rangle = \frac{1}{\sqrt{2}} \left( |\Psi_{p, \sigma'}^\pm \rangle \pm |\Psi_{\text{gs}} \rangle \right) \) can have overlaps \( T^{\pm}_{\lambda p, \sigma' \sigma} \equiv |\langle \Psi_{\text{gs}} | \Psi_{p, \sigma'}^\pm \rangle|^2 \) with the energy eigenstates, from which the charge-charge matrix elements in eq. (2) can be calculated as

\[ N^{\pm}_{\lambda p, \sigma' \sigma} = e^{-i\pi/4} \left( T^{\pm}_{\lambda p, \sigma' \sigma} - T^{-\pm}_{\lambda p, \sigma' \sigma} \right) + e^{i\pi/4} \left( T^{\pm}_{\lambda p', \sigma' \sigma} - T^{-\pm}_{\lambda p', \sigma' \sigma} \right). \]

We construct a circuit \( C_{\text{par}, p' \sigma'} \) equipped with three ancillary qubits by using the controlled operations of the partial circuits \( \mathcal{U}^{(p)}_{\text{para}} \) and \( \mathcal{U}^{(q)}_{\text{para}} \), defined in Fig. 1, as depicted in Fig. 2. The whole system consists of the ancillary and an arbitrary input register \( |\psi\rangle \). Its state changes by undergoing the circuit as

\[ |q_0^A = 0 \rangle \otimes |q_0^A = 0 \rangle \otimes |\psi\rangle \]

The projective measurement on \( q_0^A \) is represented by the four operators \( P_{qq'} = |q\rangle \langle q| \otimes |q'\rangle \langle q'| \otimes I \) \((q, q' = 0, 1)\). The two outcomes among the possible four are of our interest, immediately after which the whole system collapses as follows:

\[ \text{prob.} \left| \left| n_{\text{par}} \right| \right| ^2 = \frac{1}{2}, \]

\[ \text{prob.} \left| \left| n_{\text{par}} \right| \right| ^2 = \frac{1}{2}, \]

3. Transition matrices via statistical sampling

Given the result of a measurement on the ancillary bits for a diagonal or a off-diagonal component, we have the register \(|\tilde{\psi}\rangle\) different from the input \(N\)-electron state. Then we perform QPE for the Hamiltonian \( \mathcal{H} \) by inputting \(|\tilde{\psi}\rangle\) to obtain one of the energy eigenvalues in the
Hilbert subspace for the N-electron states. A QPE experiment inevitably suffers from probabilistic errors that depend on the number of qubits and the various parameters for the Suzuki–Trotter decomposition of \( \mathcal{H} \). We assume for simplicity, however, that the QPE procedure is realized on a quantum computer with ideal precision as well as in our previous study.\(^{[19]}\) We will thus find the estimated value to be \( E_{\lambda}^N \) with a probability \( |\langle \Psi_{\lambda}^N | \psi \rangle|^2 \).\(^{[1]}\)

If we input \(|\Psi_{gs}\rangle\) to the diagonal circuit \( C_{pa} \) in Fig. 1 and observe the ancillary bit \(|q^a = 0\rangle\) for QPE, the energy eigenvalue \( E_{\lambda}^N \) will be obtained with a probability [see eq. (16)]

\[
\langle \Psi_{\lambda}^N | \frac{n_{pa}}{\sqrt{p_{pa}}} | \Psi_{gs} \rangle \|^2 \equiv \mathbb{P}_{pa} = N_{\lambda p a, p a}.
\]

This means that we can get the diagonal components of \( N_{\lambda} \) from eq. (20) via statistical sampling for a fixed combination of \( p \) and \( \sigma \).

If we input \(|\Psi_{gs}\rangle\) to the off-diagonal circuit \( C_{pa,p'a'm'} \) in Fig. 2 and observe the ancillary bits \(|q^a = 0\rangle \otimes |q'^{a'} = 1\rangle \) or \(|q^a = 1\rangle \otimes |q'^{a'} = 1\rangle \) for QPE, the energy eigenvalue \( E_{\lambda}^N \) will be obtained with probabilities [see eqs. (22) and (23)]

\[
\langle \Psi_{\lambda}^N | \frac{n_{pa,p'a'}}{\sqrt{p_{pa,p'a'}}} | \Psi_{gs} \rangle \|^2 \equiv \mathbb{P}_{pa,p'a'} = T_{\lambda p a, p a'}.
\]

This means that we can get the off-diagonal components of \( N_{\lambda} \) from eq. (20) via statistical sampling for a fixed combination of \( p, p', \sigma, \) and \( \sigma' \).

C. Spin-spin contributions

Let us next consider the determination of the spin-spin transition matrix elements \( S_{\lambda p j, p' j'} \) for \( j, j' = x, y \). Those involving the \( z \) components of spins can be calculated from \( N_{\lambda} \) by using the relation \( s_{pz} = (n_{pa} - n_{p\beta})/2 \).

1. Circuits for diagonal components

From the unitary operators in eqs. (10) and (11) for a combination of a spatial orbital \( p \) and a spin \( \sigma \), we define the following four unitary operators:

\[
U_{0x}^{(p)} \equiv U_{0p} U_{1p}, \quad U_{1x}^{(p)} \equiv -U_{1p} U_{0p}
\]

and

\[
U_{0y}^{(p)} \equiv -i U_{0p} U_{0p}, \quad U_{1y}^{(p)} \equiv i U_{1p} U_{1p}.
\]

With them, the spin operators for the \( x \) and \( y \) directions are written as

\[
s_{pj} = \frac{U_{0j}^{(p)} + U_{1j}^{(p)}}{4}.
\]

![FIG. 3. Spin-spin diagonal circuit \( C_{pj} (j = x, y) \) for probabilistic preparation of \( s_{pj} |\psi\rangle \) and \( \tilde{s}_{pj} |\psi\rangle \) from an arbitrary input state \( |\psi\rangle \) and an ancillary qubit. We define the partial circuit \( U_{j}^{(p)} \) by enclosing it with dashed lines.](image)

For \( j = x, y \). We define

\[
\tilde{s}_{pj} \equiv \frac{U_{0j}^{(p)} - U_{1j}^{(p)}}{4}
\]

for later convenience.

We construct a circuit \( C_{pj} \) equipped with an ancillary qubit by implementing the controlled operations of \( U_{0j}^{(p)} \) and \( U_{1j}^{(p)} \), as depicted in Fig. 3. The whole system consists of the ancilla and an arbitrary input register \( |\psi\rangle \). Its state changes by undergoing the circuit as

\[
|q^A = 0\rangle \otimes |\psi\rangle \rightarrow |0\rangle \otimes 2 s_{pj} |\psi\rangle + |1\rangle \otimes 2 \tilde{s}_{pj} |\psi\rangle \equiv |\Phi_{pj}\rangle.
\]

The projective measurement on \( |q^A\rangle \) is represented by the two operators \( \mathcal{P}_q = |q\rangle \langle q| \otimes I \) (\( q = 0, 1 \)). The state of the whole system collapses immediately after the measurement as follows:

\[
|\Phi_{pj}\rangle \begin{cases} \text{observed} \rightarrow |0\rangle \otimes \frac{2 s_{pj}}{\sqrt{p_{pj}}} |\psi\rangle \\ \text{prob.} \ |2 s_{pj} |\psi\rangle|^2 \equiv \mathbb{P}_{pj}, \end{cases}
\]

\[
|\Phi_{pj}\rangle \begin{cases} \text{observed} \rightarrow |1\rangle \otimes \frac{2 \tilde{s}_{pj}}{\sqrt{p_{pj}}} |\psi\rangle \\ \text{prob.} \ |2 \tilde{s}_{pj} |\psi\rangle|^2 \equiv \mathbb{P}_{pj}. \end{cases}
\]

2. Circuits for off-diagonal components

For mutually different combinations \( (p, j) \) and \( (p', j') \) of spatial orbitals and spin components \( j, j' = x, y \), we define the four non-Hermitian auxiliary operators

\[
s_{pj}^{\pm,p'j'} \equiv s_{pj} \pm e^{i \pi / 4} s_{p'j'}
\]

and

\[
\tilde{s}_{pj}^{\pm,p'j'} \equiv \tilde{s}_{pj} \pm e^{i \pi / 4} \tilde{s}_{p'j'}.
\]

Unnormalized auxiliary states \( |\Psi_{pj,p'j'}^\pm\rangle \equiv s_{pj}^{\pm,p'j'} |\Psi_{gs}\rangle \) can have overlaps \( T_{\lambda p j, p' j'}^{\pm} \equiv \langle \Psi_{\lambda} | \Psi_{pj,p'j'}^{\pm} \rangle^2 \) with the
energy eigenstates, from which the spin-spin transition matrix elements in eq. (3) can be calculated as

\[ S_{\lambda p_j, p_j'} = e^{-i\pi/4} (T_{\lambda p_j, p_j'}^+ - T_{\lambda p_j, p_j'}^-) + e^{i\pi/4} (T_{\lambda p_j', p_j}^+ - T_{\lambda p_j', p_j}^-). \]  

(35)

We construct a circuit $C_{p_j, p_j'}$ equipped with two ancillary qubits by using the controlled operations of the partial circuits $U_{ij}^{(p)}$ and $U_{ij}^{(p')}$, depicted in Fig. 4. The whole system consists of the ancillae and an arbitrary input register $|\psi\rangle$. Its state changes by undergoing the circuit as

\[ |q_1^A = 0\rangle \otimes |q_0^A = 0\rangle \otimes |\psi\rangle \quad \Rightarrow \quad |0\rangle \otimes |0\rangle \otimes 2s_{p_j, p_j'}^+|\psi\rangle \quad + |1\rangle \otimes |0\rangle \otimes 2s_{p_j, p_j'}^-|\psi\rangle \quad + |1\rangle \otimes |1\rangle \otimes 2s_{p_j', p_j'}^+|\psi\rangle \quad + |1\rangle \otimes |1\rangle \otimes 2s_{p_j', p_j'}^-|\psi\rangle \equiv |\Phi_{p_j, p_j'}\rangle. \]  

(36)

The projective measurement on $|q_1^A\rangle$ and $|q_0^A\rangle$ is represented by the four operators $P_{qq'} = |q\rangle\langle q| \otimes |q'\rangle\langle q'| \otimes I (q, q' = 0, 1)$. The two outcomes among the possible four are of our interest, immediately after which the whole system collapses as follows:

\[ \text{prob. } 2\left| s_{p_j, p_j'}^+|\psi\rangle \right|^2 \equiv P_{p_j, p_j'}^+, \]  

(37)

\[ \text{prob. } 2\left| s_{p_j, p_j'}^-|\psi\rangle \right|^2 \equiv P_{p_j, p_j'}^-. \]  

(38)

3. Transition matrices via statistical sampling

We can get the transition matrices $S_{\lambda}$ via statistical sampling similarly to the charge-charge ones. If we input $|\Psi_{gs}\rangle$ to the diagonal circuit $C_{p_j}$ in Fig. 3 followed by a measurement and QPE for $\mathcal{H}$, the energy eigenvalue $E_{\lambda}$ will be obtained with a probability $4S_{\lambda p_j, p_j'}$. [See eqs. (3) and (31)] If we use the off-diagonal circuit $C_{p_j, p_j'}$ in Fig. 4, on the other hand, the energy eigenvalue $E_{\lambda}$ will be obtained with probabilities $4\left| s_{p_j, p_j'}^\pm \right|^2\lambda_{p_j, p_j'}$ depending on the measurement outcome. [See eqs. (37) and (38)] The off-diagonal components of transition matrices are then calculated from eq. (39).

D. Spin-charge contributions

Having found ways to determine the charge-charge and spin-spin contributions, let us consider the determination of the spin-charge transition matrices $M_{\lambda}$.

1. Circuits for off-diagonal components

For combinations $(p, j)$ and $(p', j')$ of spatial orbitals and spin components $j = x, y$ with $\sigma' = \alpha, \beta$, we define the two non-Hermitian auxiliary operators

\[ v_{p_j, p'}^{\pm} \equiv s_{p_j} + e^{\pm i\pi/4} n_{p'}^{\pm} \sigma'. \]  

(39)

Unnormalized auxiliary states $|\Psi_{p_j, p'}^{\pm}\rangle \equiv v_{p_j, p'}^{\pm} |\Psi_{gs}\rangle$ can have overlaps $T_{\lambda p_j, p'}^{\pm} \equiv \langle |\Psi_{\lambda}\rangle |\Psi_{p_j, p'}^{\pm}\rangle$ with the energy eigenstates, from which the spin-charge transition matrix elements in eq. (4) can be calculated as

\[ M_{\lambda p_j, p'}^{\pm} = e^{-i\pi/4} T_{\lambda p_j, p'}^{+} + e^{i\pi/4} T_{\lambda p_j, p'}^{-} - \sqrt{2} S_{\lambda p_j, p_j} - \frac{N_{p_j}^{\pm} n_{p'}^{\pm} \sigma'}{2\sqrt{2}}. \]  

(40)

We construct a circuit $C_{p_j, p'}^{\pm}$, equipped with three ancillary qubits by using the controlled operations of the partial circuits $U_{ij}^{(p)}$ in Fig. 3 and $U_{ij}^{(p')}$ in Fig. 1, as depicted in Fig. 5. The whole system consists of the ancillae and an arbitrary input register $|\psi\rangle$. Its state changes by undergoing the circuit as

\[ |q_2^A = 0\rangle \otimes |q_1^A = 0\rangle \otimes |q_0^A = 0\rangle \otimes |\psi\rangle \quad \Rightarrow \quad |0\rangle \otimes |0\rangle \otimes |0\rangle \otimes v_{p_j, p'}^{\pm}|\psi\rangle \quad + |0\rangle \otimes |1\rangle \otimes |1\rangle \otimes s_{p_j}|\psi\rangle \quad + |1\rangle \otimes |0\rangle \otimes |1\rangle \otimes |1\rangle \otimes e^{\pm i\pi/4} n_{p'}^{\pm} |\psi\rangle \equiv |\Phi_{p_j, p'}^{\pm}\rangle. \]  

(41)

The projective measurement on $|q_2^A\rangle$ and $|q_1^A\rangle$ is represented by the four operators $P_{qq'} = |q\rangle\langle q| \otimes |q'\rangle\langle q'| \otimes I (q, q' = 0, 1)$. Only one of the four possible outcomes is of our interest, immediately after which the whole system...
for the followed by a measurement and QPE for $q = 0$ and other states from an arbitrary input state $|\psi\rangle$ and three ancillary qubits. The partial circuits defined in Figs. 1 and 3 are contained as the controlled subroutines.

collapses as follows:

$$
|\Phi_{p_j,p'\sigma'}^{\pm}\rangle \otimes |0\rangle_{\text{observed}} \rightarrow |0\rangle \otimes |0\rangle \otimes |v_{p_j,p'\sigma'}^{\pm}\rangle \sqrt{p_{p_j,p'\sigma'}} |\psi\rangle
$$

$$
\text{prob. } \left| v_{p_j,p'\sigma'}^{\pm} |\psi\rangle \right| ^2 \equiv p_{p_j,p'\sigma'}^{\pm}.
$$

2. Transition matrices via statistical sampling

We can get the transition matrices $M_A$ via statistical sampling similarly to the charge-charge and spin-spin ones. If we input $|\Psi gauge\rangle$ to the off-diagonal circuit $C_{p_j,p'\sigma'}^{\pm}$ in Fig. 5 followed by a measurement and QPE for $H$, the energy eigenvalue $E_A$ will be obtained with a probability $T_{p_j,p'\sigma'}^{\pm}$. [See eq. (42)] The off-diagonal components of transition matrices are then calculated from eq. (40).

We provide the pseudocodes in Appendix A for the calculation procedures of response functions explained above.

III. COMPUTATIONAL DETAILS

We adopted STO-6G basis sets as the Cartesian Gaussian-type basis functions[24] for all the elements in our quantum chemistry calculations. The two-electron integrals between the atomic orbitals (AOs) were calculated efficiently.[25] We first performed restricted Hartree–Fock (RHF) calculations to get the orthonormalized molecular orbitals (MOs) in the target systems and calculated the two-electron integrals between them, from which we constructed the second-quantized electronic Hamiltonians.

In the FCI calculations for the large target Hilbert subspaces, we performed exact diagonalization of the electronic (not in qubit representation) Hamiltonians by using the Arnoldi method.[26] We can take the $z$ axis as the quantization axis for spins without loss of generality since our calculations are non-relativistic.

We calculated the FCI response functions simply by substituting the necessary quantities into eq. (9). For the simulations of response functions from statistical sampling, we generated random numbers according to the matrix elements between the FCI energy eigenstates to mimic the measurements on ancillae and the ideal QPE procedures. We set $\delta$ in eq. (9) to 0.01 a.u. throughout the present study.

IV. RESULTS AND DISCUSSION

A. $C_2$ molecule

We used the experimental bond length of 1.242 Å[27] for a $C_2$ molecule in the RHF calculation and obtained the total energy $E_{\text{RHF}} = -2045.2939$ eV. This system contains six electrons per spin direction which occupy the lowest six MOs, as shown in Fig. 6(a). We found via the subsequent FCI calculation with $E_{\text{FCI}} = -2052.6918$ eV that the major electronic configuration in the non-degenerate many-electron ground state, denoted by $X^1\Sigma^+_g$ in spectroscopic notation[28], is the same as in the RHF solution. The ground state was found via the exact diagonalization of the Hilbert subspace for $n_\alpha = n_\beta = 6$, from which we obtained the lowest 2000 among the 44100 energy eigenvalues. We calculated the response functions $\chi_{\text{FCI}}$ exact within the FCI solution, from which the components $\chi_{1\pi_n,1\pi_n}^{\text{FCI}}$ and $\chi_{1\pi_\alpha,3\pi_\alpha}^{\text{FCI}}$ are plotted in Fig. 7. We also performed simulations of statistical sampling for the construction of response function $\chi_{\text{FCI-stat}}$ based on our scheme and plotted those for $N_{\text{meas}} = 10000$ and 40000. It is seen that $\chi_{1\pi_n,1\pi_n}^{\text{FCI-stat}}$ in Fig. 7(a), which involves only the HOMO, is well reproduced by $\chi_{1\pi_n,1\pi_n}^{\text{FCI-stat}}$ with $N_{\text{meas}} = 10000$. On the other hand, $\chi_{1\pi_\alpha,3\pi_\alpha}^{\text{FCI-stat}}$ in Fig. 7(b) is not accurately reproduced by $\chi_{1\pi_\alpha,3\pi_\alpha}^{\text{FCI-stat}}$ with $N_{\text{meas}} = 10000$. These results mean that the response involving a weak excitation channel requires a large number of measurements for its accurate reproduction, just like the situation for the GFs.[19]

B. $N_2$ molecule

We used the experimental bond length of 1.098 Å[29] for an $N_2$ molecule in the RHF calculation and obtained the total energy $E_{\text{RHF}} = -2953.5952$ eV. This system contains seven electrons per spin direction which occupy the lowest seven MOs, as shown in Fig. 6(b). We found via the subsequent FCI calculation with $E_{\text{FCI}} = -2957.9124$ eV that the major electronic configuration in the non-degenerate many-electron ground state is $X^1\Sigma^+_g[28]$, the same as in the RHF solution. The ground state was found via the exact diagonalization of the Hilbert subspace for $n_\alpha = n_\beta = 7$, from which we obtained the lowest 2000 among the 14400 energy eigenvalues.
FIG. 6. Schematic illustration of RHF orbitals and their electronic occupancies for (a) a C₂ molecule and (b) an N₂ molecule. The descriptions beside the energy levels represent the orbital characters. Those with asterisks are the anti-bonding orbitals. The 1σ and 1σ⁺ MOs, coming from the 1s AOs of the constituent atoms, are not shown. The 1π and 1π⁺ MOs come mainly from the π bonding of 2p AOs, while the 3σ and 3σ⁺ MOs from the σ bonding of 2p AOs. The non-degenerate many-electron ground state for each system consists mainly of the same electronic configuration as the RHF one.

We calculated the response functions from the FCI solution, from which the components $\chi_{3\sigma n, 3\sigma n}^{\text{FCI}}$ and $\chi_{3\sigma n, 1\pi^+ n}^{\text{FCI}}$ are plotted in Fig. 8. We also performed simulations for the construction of $\chi_{\text{FCI-stat}}$ and plotted those for $N_{\text{meas}} = 10000$ and 40000. It is seen that $\chi_{3\sigma n, 3\sigma n}^{\text{FCI}}$ in Fig. 8(a), which involves only the HOMO, is not well reproduced by $\chi_{3\sigma n, 3\sigma n}^{\text{FCI-stat}}$ with $N_{\text{meas}} = 10000$, in contrast to the C₂ molecule case. Since the strength of $\chi_{3\sigma n, 1\pi^+ n}^{\text{FCI}}$ in Fig. 8(b) is similar to that of $\chi_{3\sigma n, 3\sigma n}^{\text{FCI}}, N_{\text{meas}} = 10000$ is not sufficient for the accurate reproduction of correct values as well. We found that $\chi_{3\pi z, 1\pi^+ z}^{\text{FCI}}$ (not shown) is much weaker and even $N_{\text{meas}} = 40000$ is insufficient for obtaining good $\chi_{3\pi z, 1\pi^+ z}^{\text{FCI-stat}}$.

V. CONCLUSIONS

We proposed a scheme for the construction of charge and spin linear-response functions of an interacting electronic system via QPE and statistical sampling on a quantum computer. By using the unitary decomposition of electronic operators for avoiding the difficulty due to their non-unitarity, we provided the circuits equipped with at most three ancillae for probabilistic preparation of qubit states on which the necessary non-unitary operators have acted. We performed simulations of such construction of the response functions for C₂ and N₂ molecules by comparing with the accurate ones based on the FCI calculations. It was found that the accurate detection of subtle structures coming from the weak poles in the response functions requires a large number of measurements.

Since the unitary decomposition of electronic operators is applicable regardless of the adopted qubit representation, an electronic state on which an arbitrary product of the creation and annihilation operators has acted can be prepared at least probabilistically. The approach described in this study thus enables one to access not only the response functions and GFs but also various physical quantities on a quantum computer. Invention and enrichment of tools for such properties will enhance the practical use of quantum computers for material simulations.

FIG. 7. The response function (a) $\chi_{1\pi n, 1\pi n}^{\text{FCI}}$ and (b) $\chi_{1\sigma z, 3\sigma z}^{\text{FCI}}$ exact within the FCI solution for a C₂ molecule. The simulated ones $\chi_{\text{FCI-stat}}$ for for the number of measurements $N_{\text{meas}} = 10000$ and 40000 are also shown.
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Appendix A: Pseudocodes

Here we provide the pseudocodes for the calculation of response functions proposed in the present study. We assume that all the energy eigenvalues of $N$-electron states have been obtained before entering the main procedure given just below.

1. Main procedure

Here is the main procedure. It calls directly or indirectly all the other procedures provided in this Appendix.
Procedure 1 Calculation of charge and spin response functions via statistical sampling

Input:
Hamiltonian $\mathcal{H}$, number of spatial orbitals $n_{\text{ orbs}}$, N-electron ground state $|\Psi_{\text{ gs}}\rangle$ with its energy $E_{\text{ gs}}^N$, energy eigenvalues $E^N_\lambda$, real frequency $\omega$, small positive constant $\delta$, number of measurements $N_{\text{ meas}}$ for each component.

Output:
Response functions $\chi(\omega)$

1. function CALCRESPFUNCS($\mathcal{H}, n_{\text{ orbs}}, |\Psi_{\text{ gs}}\rangle, E_{\text{ gs}}^N, E^N_\lambda, \omega, \delta, N_{\text{ meas}}$)

2. for $\lambda$

3. $d_{\pm} := \pm (\omega + i \delta) - (E^N_\lambda - E_{\text{ gs}}^N)$

4. $\chi_{\text{ tmp}} := 0$

5. for $p = 1, \ldots, n_{\text{ orbs}}$

6. for $\sigma = \alpha, \beta$

7. $N_{\text{ par}, \sigma} := \text{AmplsChargeOffDiag}(\mathcal{H}, |\Psi_{\text{ gs}}\rangle, E^N, p, \sigma, N_{\text{ meas}})$

8. for $\lambda$

9. $\chi_{\text{ tmp}}^{\text{ par}, \sigma} := N_{\text{ par}, \sigma}/d_{\lambda} + N_{\lambda_{\text{ par}, \sigma}}/d_{\lambda}$

10. for $\lambda$

11. $S_{p_j, p_j} := \text{AmplsSpinDiag}(\mathcal{H}, |\Psi_{\text{ gs}}\rangle, E^N, p, \sigma, N_{\text{ meas}})$

12. for $\lambda$

13. $\chi_{\text{ tmp}}^{\text{ spin}, \sigma} := S_{\lambda_{p_j, p_j}}/d_{\lambda} + S_{\lambda_{p_j, p_j}}/d_{\lambda}$

14. for $p, p' = 1, \ldots, n_{\text{ orbs}} (p \neq p')$

15. for $\sigma, \sigma' = \alpha, \beta$

16. if $p > p'$ or $(p = p' \text{ and } \sigma = \beta \text{ and } \sigma' = \alpha)$ then

17. $N_{\lambda_{p, p', \sigma', \sigma}} := \text{AmplsChargeOffDiag}(\mathcal{H}, |\Psi_{\text{ gs}}\rangle, E^N, p, p', \sigma, \sigma', N_{\text{ meas}})$

18. for $\lambda$

19. $\chi_{\text{ tmp}}^{\text{ par}, \sigma, \sigma'} := N_{\lambda_{p, p', \sigma', \sigma}}/d_{\lambda}$

20. for $j, j' = x, y$

21. if $p > p'$ or $(p = p' \text{ and } j = y \text{ and } j' = x)$ then

22. $S_{p_j, p_j'} := \text{AmplsSpinOffDiag}(\mathcal{H}, |\Psi_{\text{ gs}}\rangle, E^N, p, p', \sigma, \sigma', N_{\text{ meas}})$

23. for $\lambda$

24. $\chi_{\text{ tmp}}^{\text{ spin}, \sigma, \sigma'} := S_{\lambda_{p_j, p_j'}}/d_{\lambda}$

25. for $j = x, y, \sigma' = \alpha, \beta$

26. $M_{p_j, \sigma', \sigma} := \text{AmplsSpinChargeOffDiag}(\mathcal{H}, |\Psi_{\text{ gs}}\rangle, E^N, p, p', j, \sigma, \sigma', N_{\text{ meas}})$

27. for $\lambda$

28. $\chi_{\text{ tmp}}^{\text{ spin}, \sigma, \sigma'} := M_{\lambda_{p_j, \sigma'}}/d_{\lambda} + M_{\lambda_{p_j, \sigma'}}/d_{\lambda}$

29. for $p, p' = 1, \ldots, n_{\text{ orbs}} (p \neq p')$

30. $\chi_{\text{ par}, p', \sigma}^{\text{ tmp}} := \chi_{\text{ par}, p', \sigma}^{\text{ tmp}} + \chi_{\text{ par}, p', \sigma}^{\text{ spin}}$ $\chi_{\text{ tmp}}^{\text{ par}, \sigma, \sigma'} := (\chi_{\text{ par}, p', \sigma}^{\text{ spin}} + \chi_{\text{ par}, p', \sigma}^{\text{ spin}})$

31. $\chi_{p', p'}^{\text{ tmp}} := (\chi_{p', p'}^{\text{ spin}} + \chi_{p', p'}^{\text{ spin}})/2$

32. for $j = x, y$

33. $\chi_{p, p'}^{\text{ tmp}} := \chi_{p, p'}^{\text{ spin}} + \chi_{p, p'}^{\text{ spin}}$ $\chi_{p, p'}^{\text{ tmp}} := (\chi_{p, p'}^{\text{ spin}} + \chi_{p, p'}^{\text{ spin}})/2$

34. for $j, j' = x, y$

35. $\chi_{p, p'}^{\text{ tmp}} := \chi_{p, p'}^{\text{ spin}}$ $\chi_{p, p'}^{\text{ tmp}} := (\chi_{p, p'}^{\text{ spin}} + \chi_{p, p'}^{\text{ spin}})/2$

36. return $\chi$

2. Charge-charge contributions

The following procedures calculate the matrix elements in the manner described in Subsection II.B.
**Procedure 2** Calculation of diagonal components of transition matrices

1: function `AmplsChargeDiag(H, |Ψgs⟩, E^N, p, σ, N_{meas})`
2: \[ N_{pσ, pσ} := 0 \]
3: for \( m = 1, \ldots, N_{\text{meas}} \) do
4:   Input \(|Ψgs⟩\) to \( C_{pσ} \) and measure the ancilla \(|q_0^A⟩\)
5:   if \(|q_0^A⟩ = |0⟩\) then
6:     \[ E := \text{QPE}(|Ψ⟩, H) \]
7:     Find \( E \) among \( \{E^N_λ\}_λ \)
8:   \[ N_{pσ, pσ} += 1 \]
9: end for
10: return \( N_{pσ, pσ} \)

**Procedure 3** Calculation of off-diagonal components of transition matrices

1: function `AmplsChargeOffDiag(H, |Ψgs⟩, E^N, p, p', σ, σ', N_{meas})`
2: \[ T^±_{pσ, p'σ'} := 0 \]
3: for \( m = 1, \ldots, N_{\text{meas}} \) do
4:   Input \(|Ψgs⟩\) to \( C_{pσ}, p'σ' \) and measure the ancillae \(|q_{A1}⟩ \) and \(|q_{A2}⟩ \)
5:   if \(|q_{A1}⟩ \otimes |q_{A2}⟩ = |0⟩ \otimes |1⟩\) then
6:     \[ E := \text{QPE}(|Ψ⟩, H) \]
7:     Find \( E \) among \( \{E^N_λ\}_λ \)
8:   \[ T^±_{pσ, p'σ'} += 1 \]
9: else if \(|q_{A1}⟩ \otimes |q_{A2}⟩ = |1⟩ \otimes |1⟩\) then
10:    \[ E := \text{QPE}(|Ψ⟩, H) \]
11:   Find \( E \) among \( \{E^N_λ\}_λ \)
12: \[ T^±_{pσ, p'σ'} += 1 \]
13: end if
14: end for
15: return \( T^±_{pσ, p'σ'} \)

**Procedure 4** Calculation of transition amplitudes for auxiliary states

1: function `AmplsChargeAux(H, |Ψgs⟩^N, E^N, p, p', σ, σ', N_{meas})`
2: \[ T^±_{pσ, p'σ'} := 0 \]
3: for \( m = 1, \ldots, N_{\text{meas}} \) do
4:   Input \(|Ψgs⟩\) to \( C_{pσ}, p'σ' \) and measure the ancillae \(|q_2^2⟩ \) and \(|q_1^4⟩ \)
5:   if \(|q_2^2⟩ \otimes |q_1^4⟩ = |0⟩ \otimes |1⟩\) then
6:     \[ E := \text{QPE}(|Ψ⟩, H) \]
7:     Find \( E \) among \( \{E^N_λ\}_λ \)
8:   \[ T^±_{pσ, p'σ'} += 1 \]
9: else if \(|q_2^2⟩ \otimes |q_1^4⟩ = |1⟩ \otimes |1⟩\) then
10:    \[ E := \text{QPE}(|Ψ⟩, H) \]
11:   Find \( E \) among \( \{E^N_λ\}_λ \)
12: \[ T^±_{pσ, p'σ'} += 1 \]
13: end if
14: end for
15: return \( T^±_{pσ, p'σ'} \)

**Procedure 4** Calculation of transition amplitudes for auxiliary states

1: function `AmplsChargeAux(H, |Ψgs⟩^N, E^N, p, p', σ, σ', N_{meas})`
2: \[ T^±_{pσ, p'σ'} := 0 \]
3: for \( m = 1, \ldots, N_{\text{meas}} \) do
4:   Input \(|Ψgs⟩\) to \( C_{pσ}, p'σ' \) and measure the ancillae \(|q_2^2⟩ \) and \(|q_1^4⟩ \)
5:   if \(|q_2^2⟩ \otimes |q_1^4⟩ = |0⟩ \otimes |1⟩\) then
6:     \[ E := \text{QPE}(|Ψ⟩, H) \]
7:     Find \( E \) among \( \{E^N_λ\}_λ \)
8:   \[ T^±_{pσ, p'σ'} += 1 \]
9: else if \(|q_2^2⟩ \otimes |q_1^4⟩ = |1⟩ \otimes |1⟩\) then
10:    \[ E := \text{QPE}(|Ψ⟩, H) \]
11:   Find \( E \) among \( \{E^N_λ\}_λ \)
12: \[ T^±_{pσ, p'σ'} += 1 \]
13: end if
14: end for
15: return \( T^±_{pσ, p'σ'} \)

3. Spin-spin contributions

The following procedures calculate the matrix elements in the manner described in Subsection II C.
a. Diagonal components

**Procedure 5** Calculation of diagonal components of transition matrices

1. function AmplsSpinDiag(\(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, j, N_{\text{meas}}\))
2. \(S_{pj,pj} := 0\)
3. for \(m = 1, \ldots, N_{\text{meas}}\)
4. Input \(|\Psi_{gs}\rangle\) to \(C_{pj}\) and measure the ancilla \(|q^\Lambda\rangle\)
5. if \(|q^\Lambda\rangle \equiv |0\rangle\) then \(E := \text{QPE}(|\Psi\rangle, \mathcal{H})\), Find \(E\) among \(\{E^N\}_\lambda\)
6. \(S_{\lambda pj, pj} \pm := 1\)
7. \(S_{pj, pj} := 1/(4N_{\text{meas}})\)
8. return \(S_{pj, pj}\)

b. Off-diagonal components

**Procedure 6** Calculation of off-diagonal components of transition matrices

1. function AmplsSpinOffDiag(\(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, p', j, j', N_{\text{meas}}\))
2. \(T_{pj, p'j'} := \text{AmplsSpinAux}(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, p', j, j', N_{\text{meas}})\)
3. \(T^\pm_{p'j',pj} := \text{AmplsSpinAux}(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p', p, j', j, N_{\text{meas}})\)
4. for \(\lambda\)
5. \(S_{\lambda pj, p'j'} := e^{-i\pi/4}(T^+_{\lambda pj, p'j'} - T^-_{\lambda pj, p'j'}) + e^{i\pi/4}(T^+_{\lambda p'j', pj} - T^-_{\lambda p'j', pj})\) \(\triangleright\) See eq. (35)
6. return \(S_{pj, p'j'}\)

**Procedure 7** Calculation of transition amplitudes for auxiliary states

1. function AmplsSpinAux(\(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, p', j, j', N_{\text{meas}}\))
2. \(T_{pj, p'j'}^\pm := 0\)
3. for \(m = 1, \ldots, N_{\text{meas}}\)
4. Input \(|\Psi_{gs}\rangle\) to \(C_{pj,p'j'}\) and measure the ancilla \(|q^\Lambda\rangle\) and \(|q^\sigma\rangle\)
5. if \(|q^\Lambda\rangle \otimes |q^\sigma\rangle \equiv |0\rangle \otimes |0\rangle\) then \(E := \text{QPE}(|\Psi\rangle, \mathcal{H})\), Find \(E\) among \(\{E^N\}_\lambda\)
6. \(T^\pm_{\lambda pj, p'j'} := 1\)
7. \(E := \text{QPE}(|\Psi\rangle, \mathcal{H})\), Find \(E\) among \(\{E^N\}_\lambda\)
8. else if \(|q^\Lambda\rangle \otimes |q^\sigma\rangle \equiv |1\rangle \otimes |0\rangle\) then \(E := \text{QPE}(|\Psi\rangle, \mathcal{H})\), Find \(E\) among \(\{E^N\}_\lambda\)
9. \(T^\pm_{\lambda pj, p'j'} := 1\)
10. \(T_{pj, pj}^\pm := 1/(4N_{\text{meas}})\)
11. return \(T_{pj, p'j'}^\pm\)

4. Spin-charge contributions

The following procedures calculate the matrix elements in the manner described in Subsection IID.

**Procedure 8** Calculation of off-diagonal components of transition matrices

1. function AmplsSpinChargeOffDiag(\(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, p', j, j', \sigma', S_{pj, pj}, N_{p', p', \sigma', \sigma', N_{\text{meas}}}\))
2. \(T_{pj, p'j'}^\pm := \text{AmplsSpinChargeAux}(\mathcal{H}, |\Psi_{gs}\rangle, E^N, p, p', j, j', \sigma', N_{\text{meas}})\)
3. for \(\lambda\)
4. \(M_{\lambda pj, p'j'} := e^{-i\pi/4}T^+_{\lambda pj, p'j'} + e^{i\pi/4}T^-_{\lambda pj, p'j'} - \sqrt{2S_{\lambda pj, pj} - N_{\lambda p', p', \sigma', \sigma'}}\) \(\triangleright\) See eq. (40)
5. return \(M_{pj, p'j'}\)
Procedure 9 Calculation of transition amplitudes for auxiliary states

1:  function AmpsSpinChargeAux($H$, $|\Psi_{gs}\rangle$, $E^N$, $\nu, p, p', j, j'$, $N_{meas}$)
2:    $T^{\nu}_{pj,p'j} := 0$
3:    for $m = 1, \ldots, N_{meas}$
4:      Input $|\Psi_{gs}\rangle$ to $C^{\nu}_{pj,p'j}$, and measure the ancillae $|q^A\rangle$ and $|q^B\rangle$
5:      if $|q^A\rangle \otimes |q^B\rangle = |0\rangle \otimes |0\rangle$ then
6:        $E := \text{QPE}(\Psi, H)$, Find $E$ among $\{E^N\}_\lambda$
7:        $T^{\nu}_{pj,p'j} := 1$
8:      end if
9:    end for
10:   return $T^{\nu}_{pj,p'j}$

Circuit in Fig. 5 for the register $|\Psi\rangle$ coming out of the circuit

[1] M. A. Nielsen and I. L. Chuang, Quantum Computation and Quantum Information: 10th Anniversary Edition, 10th ed. (Cambridge University Press, New York, NY, USA, 2011).
[2] F. Arute, K. Arya, R. Babbush, D. Bacon, J. C. Bardin, R. Barends, R. Biswas, S. Boixo, F. G. S. L. Brandao, D. A. Buell, B. Burkett, Y. Chen, Z. Chen, B. Chiaro, R. Collins, W. Courtney, A. Dunsworth, E. Farhi, B. Foxen, A. Fowler, C. Gidney, M. Giustina, R. Graff, K. Guerin, S. Habegger, M. P. Harrigan, M. J. Hartmann, A. Ho, M. Hofmann, T. Huang, T. S. Humble, S. V. Isakov, E. Jeffrey, Z. Jiang, D. Kafri, K. Kechedzhi, J. Kelly, P. V. Klimov, S. Knysh, A. Korotkov, F. Kostritsa, D. Landhuis, M. Lindmark, E. Lucero, D. Lyakh, J. M. Martinis, P. M. Mewes, X. Mi, K. Michielsen, M. Mohseni, J. Mutus, O. Naaman, N. Neeley, C. Neill, M. Y. Niu, E. Ostby, A. Petukhov, J. C. Platt, C. Quintana, E. G. Rieffel, P. Roushan, N. C. Rubin, D. Sank, K. J. Satzinger, V. Smelyanskiy, K. J. Sung, M. D. Treutli, B. Villalonga, T. White, Z. J. Yao, P. Yeh, A. Zalcman, H. Neven, and J. M. Martinis, Nature 574, 505 (2019).
[3] S. McArdle, S. Endo, A. Aspuru-Guzik, S. Benjamin, and X. Yuan, arXiv e-prints, arXiv:1808.10402 (2018), arXiv:1808.10402 [quant-ph].
[4] P. Jordan, and E. Wigner, Zeitschrift für Physik 47, 631 (1928).
[5] S. B. Bravyi and A. Y. Kitaev, Annals of Physics 298, 210 (2002).
[6] A. Aspuru-Guzik, A. D. Dutoi, P. J. Love, and M. Head-Gordon, Science 309, 1704 (2005), https://science.sciencemag.org/content/309/5741/1704.full.pdf
[7] A. Peruzzo, J. McClean, P. Shadbolt, M.-H. Yung, X.-Q. Zhou, P. J. Love, A. Aspuru-Guzik, and J. L. O’Brien, Nature Communications 5, 4213 EP (2014), article.
[8] P. J. J. O’Malley, R. Babbush, I. D. Kivlichan, J. Romero, J. R. McClean, R. Barends, J. Kelly, P. Roushan, A. Trantner, N. Ding, B. Campbell, Y. Chen, Z. Chen, B. Chiaro, A. Dunsworth, A. G. Fowler, E. Jeffrey, E. Lucero, A. Megrant, J. Y. Mutus, M. Neely, C. Neill, C. Quintana, D. Sank, A. Vainsencher, J. Wenner, T. C. White, P. V. Coveney, P. J. Love, H. Neven, A. Aspuru-Guzik, and J. M. Martinis, Phys. Rev. X 6, 031007 (2016).
[9] A. Kandala, A. Mezzacapo, K. Temme, M. Takita, M. Brink, J. M. Chow, and J. M. Gambetta, Nature 549, 242 EP (2017).
[10] C. Hempel, C. Maier, J. Romero, J. McClean, T. Monz, H. Shen, P. Jurcevic, B. P. Lanyon, P. Love, R. Babbush, A. Aspuru-Guzik, R. Blatt, and C. F. Roos, Phys. Rev. X 8, 031022 (2018).
[11] J. R. McClean, E. Kimchi-Schwartz, J. Carter, and W. A. de Jong, Phys. Rev. A 95, 042308 (2017).
[12] K. M. Nakanishi, K. Mitarai, and K. Fujii, arXiv e-prints, arXiv:1810.09434 (2018), arXiv:1810.09434 [quant-ph].
[13] O. Higgott, D. Wang, and S. Brierley, arXiv e-prints, arXiv:1805.08138 (2018), arXiv:1805.08138 [quant-ph].
[14] R. Santagati, J. Wang, A. A. Gentile, S. Paesani, N. Wiebe, J. R. McClean, S. Morley-Short, P. J. Shadbolt, D. Borneau, J. W. Silverstone, D. P. Tew, X. Zhou, J. L. O’Brien, and M. G. Thompson, Science Advances 4 (2018), 10.1126/sciadv.aap9646, https://advances.sciencemag.org/content/4/1/eaap9646.full.pdf.
[15] J. I. Colless, V. V. Ramasesh, D. Dahlen, M. S. Blok, M. E. Kimchi-Schwartz, J. R. McClean, J. Carter, W. A. de Jong, and I. Siddiqi, Phys. Rev. X 8, 011021 (2018).
[16] T. Jones, S. Endo, S. McArdle, X. Yuan, and S. C. Benjamin, Phys. Rev. A 99, 062304 (2019).
[17] A. Damascelli, Physica Scripta 2004, 61 (2004).
[18] T. Kosugi and Y.-i. Matsushita, Journal of Physics: Condensed Matter 30, 435604 (2018).
[19] T. Kosugi and Y.-i. Matsushita, arXiv e-prints, arXiv:1908.03902 (2019), arXiv:1908.03902 [quant-ph].
[20] S. Endo, I. Kurata, and Y. O. Nakagawa, arXiv e-prints, arXiv:1909.12250 (2019), arXiv:1909.12250 [quant-ph].
[21] G. Stefanucci and R. van Leeuwen, Nonequilibrium Many-Body Theory of Quantum Systems (Cambridge University Press, 2013).
[22] T. Kosugi and Y.-i. Matsushita, The Journal of Chemical Physics 147, 114105 (2017), https://doi.org/10.1063/1.4994720.
[23] L. Hedin, Phys. Rev. 139, A796 (1965).
[24] T. Helgaker, P. Jørgensen, and J. Olsen, Molecular Electronic-Structure Theory (Wiley, 2000).
[25] J. T. Fermann and E. F. Valeev, “Libint: Machine-generated library for efficient evaluation of molecular integrals over Gaussians,” (2003), freely available at
http://libint.valeyev.net/ or one of the authors.

[26] R. Lehoucq, D. Sorensen, and C. Yang, 
*ARPACK Users’ Guide* (Society for Industrial and Applied Mathematics, 1998) 
http://epubs.siam.org/doi/pdf/10.1137/1.9780898719628.

[27] M. Douay, R. Nietmann, and P. Bernath, 
*Journal of Molecular Spectroscopy* **131**, 250 (1988).

[28] L. Bytautas and K. Ruedenberg, 
*The Journal of Chemical Physics* **122**, 154110 (2005), 
https://doi.org/10.1063/1.1869493.

[29] L. Sutton, *Tables of Interatomic Distances and Configuration in Molecules and Ions* (The Chemical Society, London, 1958).