Evaluating the efficiency of ground-state-preparation algorithms

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In recent years, substantial research effort has been devoted to quantum algorithms for ground-state-energy estimation (GSEE) in chemistry and materials. Given the many heuristic and nonheuristic methods being developed, it is challenging to assess what combination of these methods will ultimately be used in practice. One important metric for assessing utility is the runtime, which depends on the ground-state preparation (GSP) for most GSEE algorithms. Towards assessing the utility of various combinations of GSEE and GSP methods, we asked under which conditions a GSP method should be accepted over a reference method, such as the Hartree-Fock method. We introduce a criterion for accepting or rejecting a GSP method for the purposes of GSEE. We consider different GSP methods ranging from heuristics to algorithms with provable performance guarantees and perform numerical simulations to benchmark their performance on different chemical systems, starting from small molecules like the hydrogen molecule to larger systems like jellium. In the future, this approach may be used to abandon certain variational quantum eigensolver (VQE) ansatzes and other heuristics. Yet our findings do not provide evidence against using VQE and more expensive heuristic methods, like the low-depth booster. This work sets a foundation from which to further explore the requirements to achieve quantum advantage in quantum chemistry.

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I. INTRODUCTION

Quantum computation promises to unlock new computational capabilities for certain tasks such as the ground-state-energy estimation (GSEE) for molecules and materials [1–3]. However, realizing quantum advantage for the task of GSEE requires improvements in the quantum algorithms that will reduce the resource requirements needed, such as the circuit depth [4]. This has led to extensive research on the development of algorithms with modest circuit depths [4,5].

However, the performance of GSEE algorithms also strongly depends on the overlap of the true ground state of the Hamiltonian and the initial state generated by a ground-state-preparation (GSP) method [6–8]. For quantum chemistry applications, the Hartree-Fock (HF) Slater determinant state is widely used for GSP, since the cost (in terms of circuit depth) of implementing it on quantum hardware is insignificant compared to GSEE algorithms [9] and it provides satisfactory results for many molecules and materials [10]. However, in some important cases the overlap is relatively small (for example, molecules with a bond distance out of equilibrium [10,11]), which creates a need for methods that can provide a larger initial overlap.

To this end, different quantum GSP algorithms have been developed to provide a higher overlap and improve the performance of GSEE algorithms. These include quantum algorithms that prepare multideterminant states [10] and numerous GSP algorithms with a provable performance guarantee [5,12,13]. Adiabatic-state preparation using digital quantum computing [14,15] is another approach to preparing ground states and in some works [16,17] has been considered the de facto method for state preparation. More recent heuristic approaches, such as the variational quantum eigensolver (VQE) [18,19] can also provide an approximation to the ground state, while using relatively little circuit depth. Recent work on low-depth boosters has introduced a method with provable performance guarantees on reliably converting circuit depth into ground-state overlap and goes beyond heuristic parametrized quantum circuits [20]. They showed that any function f that satisfies the monotonicity suppresses the high-energy eigenstates of a Hamiltonian H and hence boosts the low-energy states [20]. However, the choice of the function f and the function parameters is heuristic. Finally, classical machine-learning techniques from generative modeling have been applied to the task of generating approximations to the ground state [11].

Even though we have a plethora of methods for ground-state preparation, we are still missing a reliable way to benchmark their performance. The notion of "good overlap," as usually referred to, is vague and does not explore a performance to resource cost ratio as a benchmarking tool [8,10]. We desire benchmark tools that address the trade-off between the resource cost and performance improvement of the GSP and GSEE subroutines. Such a tool could be used to answer questions like the following: Is it worth the high circuit depth cost to use a GSP algorithm that provides almost perfect overlap values? Or is it better to settle for a heuristic method like VQE with smaller ground state overlap even though it increases the runtime of the GSEE subroutine?

The efficiency of a GSP algorithm gives the right tools to understand the appropriate balance of resource cost and performance of GSP and GSEE algorithms. To evaluate the

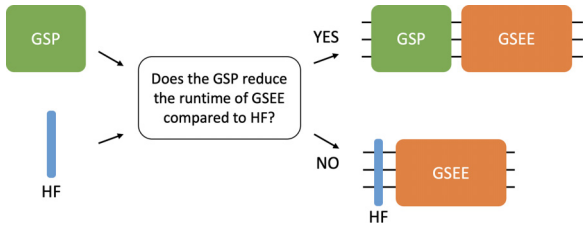

FIG. 1. The acceptability criteria are used to benchmark the given GSP method over the HF reference with the goal of reducing the total runtime of the GSEE algorithm.

performance of quantum algorithms, recent work has proposed a resource efficiency metric as the ratio of the success metric over the resource cost [21]. Instead of defining efficiency metrics, in this work we introduce a criterion to evaluate whether to accept or reject a given GSP algorithm. The recent work [22] explores the conditions under which the improved performance is worth the extra resource cost of the GSP algorithm introduced in Ref. [13] for the task of GSEE. Here we propose a systematic way to benchmark different GSP methods for the problem of GSEE. We use the HF method as a reference and explore under which conditions a GSP method will be accepted over HF. The benchmarking criteria incorporate both the reduction of the total runtime for GSEE and the resource cost of the GSP algorithm (see Fig. 1). We perform numerical simulations to showcase how to use the criteria in practice and provide a resource estimation of the maximum allowed depth of a GSP to be acceptable over HF.

The paper is organized as follows. In Sec. II we introduce the criteria for acceptability of GSP that do not require repetitions, while we incorporate repetitions of GSP in Sec. III. In these two sections, the choice of GSP is arbitrary. In Sec. IV, we present numerical simulations on how to use the benchmarking criteria set in the first two sections by using VQE and low-depth boosters as GSP methods. Also, we include a resource estimation of the maximum allowed GSP circuit depth for solid-state materials. Section V contains the conclusions and future research directions.

II. CRITERIA FOR ACCEPTABILITY OF STATE-PREPARATION METHODS

Here we discuss the criteria under which a state-preparation method is acceptable for the purposes of GSEE. Any GSEE method has a runtime that depends on the features of the GSP: the GSP circuit depth D and the overlap γ between the prepared state and the true ground state. The runtime also depends on the target accuracy ϵ. For a given energy estimation algorithm [7], the runtime can be formulated as

$$\begin{aligned}\mathcal{T}(D,\gamma;\epsilon) &= (\text{number of repetitions})\\ &\quad\times(\text{total circuit depth of each repetition})\\ &= (\text{number of repetitions})\\ &\quad\times(\text{GSP depth}+\text{GSEE depth})\\ &= \tilde{O}\left[\frac{1}{\gamma^{\alpha}}\left(D+\frac{1}{\epsilon\gamma^{\beta}}\right)\right].\end{aligned} \tag{1}$$

A recent review on the different GSP and GSEE algorithms has been discussed in Refs. [7,23]. According to the table given in Ref. [7], example GSEE algorithms have values $\alpha\in\{0,2,4\}$ and $\beta\in\{0,1,2\}$ and it depends on the studied GSEE algorithms which pairwise values of α and β are used. The units for the GSP and GSEE circuit depths need to match, but as it will become evident from the numerical simulations in the next section different choices for the units could be used, such as the circuit depth and the T-gate count. Note that the GSP and GSEE algorithms could belong to different quantum regimes, for example, noisy intermediate scale quantum (NISQ) and fault-tolerant quantum computing (FTQC) as traditionally referred to, but eventually the depth units should match. Also, we are ignoring constant factors and logarithmic dependence on the parameters for now to simplify the introduction of this technique, but these should ultimately be included to set a more accurate benchmark. In the numerical simulations in Sec. IV we discuss cases where this simplification is valid. Finally, some state-preparation methods require a number of repetitions to ensure their success (with high probability). In Sec. III, we discuss the runtime cost that includes repetitions of the GSP algorithms.

To establish the concept of an acceptable state preparation, we must assume that the ground-state-energy-estimation algorithm has a reference or default method to compare with. A reference initial state for the task of GSEE could be simply a product state of the measurement basis, or the solution to a mean-field-level method, such as the Hartree-Fock ground state. The circuit depth for preparing a reference state can usually be neglected in cases such as the second quantization, thus we label the depth and overlap as $D_0=0$ and γ_0. In this work, we focus on the HF method as a reference exactly due to the zero depth cost. Other methods, such as adiabatic-state preparation [1,24], which have nonzero depth cost, could be used as reference in future work. The runtime of GSEE using the HF-reference-state preparation is

$$\mathcal{T}_0(D_0,\gamma_0;\epsilon)=\tilde{O}\left(\frac{1}{\epsilon\gamma_0^{\alpha+\beta}}\right). \tag{2}$$

The condition for a state-preparation method to be acceptable over the reference is that the total runtime of the GSEE with the GSP ($\mathcal{T}$) is smaller than the total runtime of the GSEE with HF ($\mathcal{T}_0$), i.e., $\mathcal{T}<\mathcal{T}_0$. This puts constraints on the state-preparation parameters:

$$\frac{1}{\gamma^{\alpha}}\left(D+\frac{1}{\epsilon\gamma^{\beta}}\right)<\frac{1}{\epsilon\gamma_0^{\alpha+\beta}}. \tag{3}$$

Observe that if $D=0$, then the acceptability criteria reduce to the condition $\gamma>\gamma_0$. We can rewrite the general condition (3) as

$$D<\frac{1}{\epsilon\gamma^{\beta}}\left[\left(\frac{\gamma}{\gamma_0}\right)^{\alpha+\beta}-1\right]. \tag{4}$$

This shows that if ϵ is decreased then a state preparation with larger D will be accepted. In other words, for less demanding GSEE algorithms with a worse target accuracy ϵ, a more costly GSP algorithm could be accepted over the HF. Finally,

TABLE I. Comparison of quantum algorithms for ground-state-energy estimation along with the scaling parameters α and β that are relevant to the efficiency calculation.

Algorithm	GSEE depth $\tilde{O}(\epsilon^{-1}\gamma^{-\beta})$	Repetitions $\tilde{O}(\gamma^{-\alpha})$	$\alpha+\beta$
LT20 [13]	$\tilde{O}(\epsilon^{-1}\gamma^{-1})$	$\tilde{O}(1)$	1
DLT22 [7]	$\tilde{O}(\epsilon^{-1})$	$\tilde{O}(\gamma^{-2})$	2
QPE [26,27]	$\tilde{O}(\epsilon^{-1}\gamma^{-2})$	$\tilde{O}(\gamma^{-2})$	4

we could write the above inequality as the following:

$$\frac{D+1/\epsilon\gamma^{\beta}}{1/\epsilon\gamma^{\beta}} < \left(\frac{\gamma}{\gamma_0}\right)^{\alpha+\beta}. \tag{5}$$

Therefore, the acceptability criterion in the more strict case when $\alpha+\beta=1$ could be expressed in words as

$$\frac{\text{total depth}}{\text{GSEE depth}} < \frac{\text{GSP overlap}}{\text{HF overlap}} \tag{6}$$

or

$$\frac{\text{total depth}}{\text{GSEE depth}} < \frac{N_{\text{reps}} \text{ from HF}}{N_{0,\text{reps}} \text{ from GSP}}, \tag{7}$$

where N_{reps} is the number of repetitions due to the GSP overlap value, i.e., $N_{\text{reps}}=1/\gamma$ and $N_{0,\text{reps}}=1/\gamma_0$.

Next, we discuss the simple case when the GSP query depth is much smaller than the GSEE query depth. Then, we have

$$\frac{\text{GSP depth}}{\text{GSEE depth}} \ll 1, \tag{8}$$

$$D\gamma^{\beta} \ll \frac{1}{\epsilon}. \tag{9}$$

For typical values of $\epsilon \simeq 10^{-3}$ [4,25], the condition becomes $D\gamma^{\beta} \ll 10^3$. Since $\beta \in \{0,1,2\}$ [7] and $\gamma \leqslant 1$, the more strict condition is $D \ll \frac{1}{\epsilon}$.

Then, the acceptability criterion for any $\alpha \in \{0,2,4\}$ and $\beta \in \{0,1,2\}$ [7] is simplified to

$$1 < \left(\frac{\gamma}{\gamma_0}\right)^{\alpha+\beta}, \tag{10}$$

which simply states that the acceptance of the GSP over HF is determined by the respective overlap values ratio.

Next, we compare the acceptability criteria for two different GSEE algorithms presented in the table of GSEE performance [7], the quantum phase estimation semiclassical algorithm (here referred to as QPE) [26,27], and the GSEE algorithm developed in Ref. [13] (here referred as LT20) for a given GSP algorithm whose depth depends on the lower bound of the spectral gap Δ and the overlap γ_0. We choose the QPE and LT20 algorithms since $\alpha+\beta$ takes the maximum (4) and the minimum (1) value, respectively, as shown in Table I. For a more detailed discussion on these GSEE algorithms, we refer the reader to the aforementioned references. For QPE we have $\alpha=\beta=2$ and

$$\frac{\epsilon\gamma^2+\Delta\gamma_0}{\Delta\gamma_0} < \left(\frac{\gamma}{\gamma_0}\right)^4, \tag{11}$$

while for LT20 we have $\alpha=0$, $\beta=1$, and

$$\frac{\epsilon\gamma+\Delta\gamma_0}{\Delta\gamma_0} < \left(\frac{\gamma}{\gamma_0}\right). \tag{12}$$

Since $\gamma \leqslant 1$, we have

$$\frac{\epsilon\gamma^2+\Delta\gamma_0}{\Delta\gamma_0} \leqslant \frac{\epsilon\gamma+\Delta\gamma_0}{\Delta\gamma_0} < \left(\frac{\gamma}{\gamma_0}\right) < \left(\frac{\gamma}{\gamma_0}\right)^4. \tag{13}$$

This suggests that the acceptability criterion for LT20 is more strict than that for QPE. LT20 has a smaller GSEE query depth compared to QPE, so it is harder to accept a GSP algorithm with nonzero depth over HF. Therefore, the better the GSEE algorithms becomes in terms of query depth reduction, the more strict the criterion for the acceptance of a GSP method over HF is. In other words, as the query depth of the GSEE becomes smaller, the GSP depth becomes more relevant than the GSP overlap.

Finally, the acceptability criteria allow us to explore the maximum values of the GSP depth that enable the given GSP method to be acceptable over the HF state. Given a specific GSEE algorithm and the value of γ_0, and assuming that the GSP provides a specific value of γ, i.e., $\gamma=1$, we find the corresponding maximum acceptable depth of a GSP method. To this end, Eq. (5) can be written as

$$D < \frac{\gamma-\gamma_0}{\gamma_0} D_{\text{GSEE}}, \tag{14}$$

for more demanding GSEE algorithms with $\alpha+\beta=1$ and depth D_{GSEE}. The above equation can be expressed as

$$\text{GSP depth} < \frac{\text{performance gain}}{\text{HF performance}} \text{ GSEE depth}. \tag{15}$$

III. CRITERIA FOR ACCEPTABILITY OF STATE-PREPARATION METHODS WITH REPETITIONS

In this section, we discuss the criteria under which a GSP method that requires repetitions to reach an overlap γ is acceptable for a given GSEE algorithm. The runtime Eq. (1) discussed in the previous section becomes

$$\begin{aligned}\mathcal{T}(D,\gamma;\epsilon) &= (\text{number of repetitions of GSEE}) \\ &\quad \times [(\text{number of repetitions of GSP}) \\ &\quad \times (\text{circuit depth of GSP}) \\ &\quad + (\text{circuit depth of GSEE})] \\ &= \tilde{O}\left[\frac{1}{\gamma^{\alpha}}\left(\frac{1}{P_{\text{succ}}}\times D+\frac{1}{\epsilon\gamma^{\beta}}\right)\right].\end{aligned} \tag{16}$$

As explained earlier, the runtime for the GSEE using the HF-reference-state preparation $\mathcal{T}_0$ is given by Eq. (2). Then, the condition, i.e., $\mathcal{T} < \mathcal{T}_0$, for a state-preparation method being acceptable over the reference becomes

$$\frac{D/P_{\text{succ}}+1/\epsilon\gamma^{\beta}}{1/\epsilon\gamma^{\beta}} < \left(\frac{\gamma}{\gamma_0}\right)^{\alpha+\beta}, \tag{17}$$

which can be expressed as

$$\frac{\text{total query depth}}{\text{GSEE query depth}} < \frac{N_{\text{reps}} \text{ from HF}}{N_{\text{reps}} \text{ from GSP}}.$$

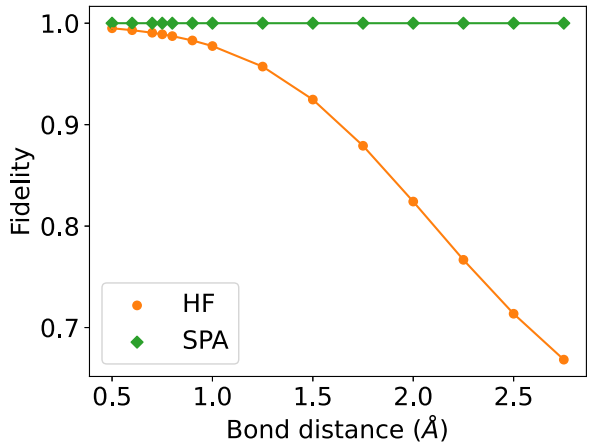

FIG. 2. Fidelity as a function of bond distance of the H_2 molecule for the HF method and the SPA algorithm as GSP methods, respectively. The value of α and β is set to be $\alpha + \beta = 1$, which corresponds to a GSEE method that has a more strict acceptability criterion.

IV. NUMERICAL SIMULATIONS

In this section, we apply the acceptability criteria and benchmark different GSP methods over the HF state for different Hamiltonians of molecules and solid-state materials, starting from small molecules (H_2 molecule) and moving on to larger molecules (N_2 molecule). These systems have also been used in other recent works [10,11,18,20,28,29]. Finally, we perform a resource estimation of the maximal acceptable circuit depth of GSP for different solid-state materials over HF state based on the recent work [25].

A. Molecules

We explore the acceptability criteria for a small molecule (H_2) with four spin-orbitals or qubits in an adapted basis [30]. We compare the HF method to the separable pair approximation (SPA) approach introduced in the recent work of Kottmann and Aspuru-Guzik [18] as the GSP method. For the numerical simulations on H_2 and N_2 molecules with the SPA algorithm, we followed the notebook [18] from Tequila [31] and the molecular data [30].

According to the work of Kottmann and Aspuru-Guzik [18], the circuit depth of SPA for the H_2 molecule equals 3. Since the depth is $D_{\mathrm{GSP}} = 3 \ll 10^3$ for typical values of chemical accuracy $\epsilon = 10^{-3}$, we are in the simple case discussed in Sec. II where the criteria are simplified to the overlap values ratio [Eq. (10)]. We assume a more demanding GSEE algorithm with $\alpha + \beta = 1$ and the criterion is given by

$$1 < \frac{\gamma}{\gamma_0}. \tag{18}$$

In Fig. 2, we plot the fidelity of the two different GSP methods. For all bond distances presented in Fig. 2, the criterion is satisfied. Specifically, at bond distance $d = 0.5$ the ratio $\frac{\gamma}{\gamma_0}$ is 1.005, leading up to the value of 1.5 for $d = 2.6$. This suggests that initially the SPA method is comparable to the HF method, and as we increase the bond distance (Å), the SPA method is acceptable over the HF method. For a less demanding GSEE algorithm (i.e., with $\alpha + \beta$ possessing different values than 1), the criteria would be satisfied and the SPA method would be acceptable over the HF method.

Next, we explore the larger molecule N_2 considering 6 active electrons in 12 spin-orbitals or qubits in an adapted basis [30] at a bond distance of $d = 2.0$. Initially, we benchmark the SPA method over the HF method. Since $D_{\mathrm{GSP}} = 3$ [18], we could apply the criterion of Eq. (18), which gives

$$1 < \frac{\gamma}{\gamma_0} = \frac{0.85}{0.72} = 1.18. \tag{19}$$

Therefore, the SPA is acceptable over the HF method. In this case, the prefactors in the acceptability criteria could be disregarded since the SPA resource cost is much smaller than that of the GSEE.

Next, for the same molecule (N_2 at bond distance $d = 2.0$), we benchmark a more costly heuristic algorithm: the low-depth booster from the recent work of Want *et al.* [20]. Details on the simulations on the low-depth-booster algorithm can be found in Ref. [20]. Following the aforementioned work [20], we change the unit of depth from the circuit depth to the accumulations of the controlled time evolution $\exp(2i\pi H)$ operations, where H is the Hamiltonian of the system. Also, we have $D_{\mathrm{GSP}} = 10^3$ with $\gamma \approx 1$, while $D_{\mathrm{GSEE}} = 2 \times 10^4$ and $\gamma_0 = 0.72$. The success probability of the low-depth-booster GSP algorithm applied with the linear combination of unitaries (LCU) method is $P_{\mathrm{succ}} \approx 0.5$ [20]. Therefore, the criterion of Eq. (17) becomes

$$\frac{D/(P_{\mathrm{succ}}) + 1/\epsilon\gamma^{\beta}}{1/\epsilon\gamma^{\beta}} < \left(\frac{\gamma}{\gamma_0}\right)^{\alpha+\beta} \Rightarrow$$

$$\frac{2.2}{2} < \left(\frac{1}{0.72}\right)^{\alpha+\beta} \Rightarrow 1.1 < (1.39)^{\alpha+\beta},$$

which is satisfied for any values of α and β of the GSEE algorithms. In this case, since both the low-depth booster and the GSEE are implemented with the LCU method, we could follow the simplification of neglecting the prefactors in the acceptability criteria.

B. Solid-state materials

Here, based on the resource estimation on the T-gate count for the problem of GSEE given in the recent work [25] and implemented with the LCU method, we explore the maximal acceptable depth of the GSP methods for different solid-state materials.

Recent work [25] estimates the T gates needed for quantum simulation of three-dimensional (3D) spinful jellium (or the homogeneous electron gas). It focuses on the T count since applying a T gate requires a lot of logical qubits and takes much longer than any other operation in a quantum circuit [32]. The 3D spinful jellium is in the dual basis at a Wigner-Seitz radius of 10 Bohr radii assuming the system is at half filling. For 54 spin-orbitals and a target chemical accuracy of $\Delta E = 0.0016$ Hartree, the depth is equal to 1.8×10^7 T count.

As explained in Ref. [25], the jellium is a good proxy for different solid-state materials, such as diamond, graphite, silicon, metallic lithium, and crystalline lithium hydride. For

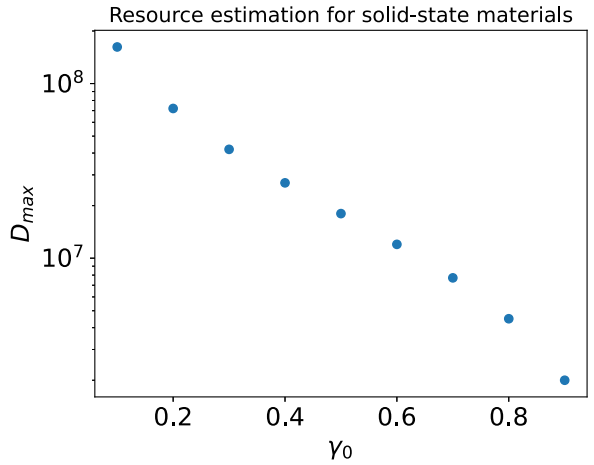

FIG. 3. Maximum acceptable depth $D_{\max}$ of the GSP algorithm with $\gamma = 1$ and $D_{\text{GSEE}} = 1.8 \times 10^7$ for solid-state materials as a function of the HF overlap γ_0.

these materials, the HF overlap could range from smaller to larger values as presented in Fig. 3. Assuming that the GSP method gives $\gamma \approx 1$, we have a resource estimation of the maximum depth allowed for the GSP method to be acceptable over that of the HF method (see Fig. 3) given by Eq. (14).

V. DISCUSSION

We introduced a method to assess when to accept or reject a ground-state-preparation (GSP) method over the Hartree-Fock (HF) reference for the task of ground-state-energy estimation (GSEE) by introducing acceptability criteria. The criteria are defined through the total runtime of the GSEE algorithm that incorporates both the number of repetitions needed and the total circuit depth of each repetition—i.e., the GSP and the GSEE depth. If the inequality introduced in Eq. (5) is satisfied, then the GSP method is acceptable over the HF method—i.e., provides a speedup in the total runtime of the GSEE algorithm. The criteria explore the trade-off of both the resource cost and the performance of GSP and GSEE subroutines.

We explored under which conditions the acceptability criteria could be simplified and also established them for GSP methods that require repetitions to reach an overlap γ. Comparing the acceptability criteria for two different GSEE algorithms with a GSP, we found that the better the GSEE algorithms become in terms of query depth reduction, the more strict the criteria to accept a GSP over the HF method must be. This could be due to the fact that the number of repetitions introduced by the GSP overlap becomes less significant as the GSEE query depth becomes smaller. The ability to trade circuit depth with runtime is also suggested in recent works [4,5]. In agreement, the resource estimation performed in this work suggests that a GSP method with a circuit depth larger than that of the GSEE could be accepted for total runtime reduction.

Next, we showed that the separable-pair-approximation (SPA) method is acceptable over the HF method for the hydrogen molecule for different bond lengths, which suggests that even for a simple molecule there exists GSP that could offer an improved performance to the GSEE algorithm over using the HF method. We also evaluated the more expensive low-depth-booster GSP algorithm for the nitrogen molecule, which is widely used for benchmarking quantum chemistry simulations, in particular, when the bond is stretched [10,18,20]. These results suggest that more expensive GSP methods could reduce the total runtime, thus being acceptable over the HF reference. In accordance, the resource estimation of the maximum allowed depth of a GSP does not provide evidence against the use of VQEs and more expensive heuristic methods. Further numerical and theoretical work is needed to draw a more definitive conclusion.

The proposed acceptability criteria are a first attempt to explore the trade-off between the resource cost and performance of the GSP algorithm used and those of the GSEE algorithm. The resource cost is evaluated in terms of the circuit depth, while the GSP and GSEE performances are evaluated in terms of the overlap γ given by GSP and the overall runtime needed for both algorithms. The proposed criteria drop the big-O notation, which means that the prefactors of the studied GSP and GSEE algorithms are neglected. It will be the subject of future work to update the criteria to incorporate the prefactors of the GSP and GSEE algorithms. For certain cases though, like the ones studied in Sec. IV, the prefactors could be neglected, for example, when the GSP depth is much smaller than that of the GSEE or when both the GSP and GSEE are implemented with the same method, i.e., linear combination of unitaries. Then, the prefactors do not play a role in the ratio that defines the acceptability criteria.

For nonheuristic GSP algorithms, the γ value might be estimated *a priori*, but this is challenging to do with heuristic methods. In principle, one might be able to establish bounds for the performance of the low-depth-booster algorithm [13]. Moreover, after a careful numerical analysis and resource estimation on a studied system, the results could potentially be extrapolated to systems with similar properties as discussed in Ref. [33] or to an increased system size. Extrapolation seems to be one of the most trustworthy approaches to assessing the efficiency of heuristic methods [8], but of course, due to the nature of heuristic methods, one should carefully evaluate such an analysis.

This work sets a foundation to further explore resource efficiency metrics for GSP and GSEE algorithms. As already mentioned, the criteria could be adjusted to incorporate logarithmic dependencies on the parameters or the recent GSEE algorithm with an exponential improvement in the circuit depth [4]. It would also be interesting to apply the criteria introduced here to molecules and materials of industrial relevance [34] and further use them for resource estimations. Our follow-up work performs a careful numerical analysis and resource estimation to benchmark two GSP algorithms with the same system under study [33]. Finally, other GSP methods could play the role of the reference method instead of the HF method. It is challenging to assess what combination of methods will ultimately be used in practice, and further research will help evaluate the utility of various combinations of GSEE and GSP methods.

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[1] A. Aspuru-Guzik, A. D. Dutoi, P. J. Love, and M. Head-Gordon, Simulated quantum computation of molecular energies, Science **309**, 1704 (2005).

[2] J. J. Goings, A. White, J. Lee, C. S. Tautermann, M. Degroote, C. Gidney, T. Shiozaki, R. Babbush, and N. C. Rubin, Reliably assessing the electronic structure of cytochrome P450 on today's classical computers and tomorrow's quantum computers, Proc. Natl. Acad. Sci. USA **119**, e2203533119 (2022).

[3] V. E. Elfving, B. W. Broer, M. Webber, J. Gavartin, M. D. Halls, K. P. Lorton, and A. Bochevarov, How will quantum computers provide an industrially relevant computational advantage in quantum chemistry? arXiv:2009.12472.

[4] G. Wang, D. Stilck-França, R. Zhang, S. Zhu, and P. D. Johnson, Quantum algorithm for ground state energy estimation using circuit depth with exponentially improved dependence on precision, Quantum **7**, 1167 (2023).

[5] Z. Ding and L. Lin, Even shorter quantum circuit for phase estimation on early fault-tolerant quantum computers with applications to ground-state energy estimation, PRX Quantum **4**, 020331 (2023).

[6] R. Zhang, G. Wang, and P. Johnson, Computing ground state properties with early fault-tolerant quantum computers, Quantum **6**, 761 (2022).

[7] Y. Dong, L. Lin, and Y. Tong, Ground-state preparation and energy estimation on early fault-tolerant quantum computers via quantum eigenvalue transformation of unitary matrices, PRX Quantum **3**, 040305 (2022).

[8] S. Lee, J. Lee, H. Zhai, Y. Tong, A. M. Dalzell, A. Kumar, P. Helms, J. Gray, Z.-H. Cui, W. Liu, M. Kastoryano, R. Babbush, J. Preskill, D. R. Reichman, E. T. Campbell, E. F. Valeev, L. Lin, and G. K.-L. Chan, Evaluating the evidence for exponential quantum advantage in ground-state quantum chemistry, Nat. Commun. **14**, 1952 (2023).

[9] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information*, 10th anniversary ed. (Cambridge University, England, 2011).

[10] N. M. Tubman, C. Mejuto-Zaera, J. M. Epstein, D. Hait, D. S. Levine, W. J. Huggins, Z. Jiang, J. R. McClean, R. Babbush, M. Head-Gordon, and K. B. Whaley, Postponing the orthogonality catastrophe: efficient state preparation for electronic structure simulations on quantum devices, Bull. Am. Phys. Soc. (2018).

[11] J. Ceroni, T. F. Stetina, M. Kieferova, C. O. Marrero, J. M. Arrazola, and N. Wiebe, Generating approximate ground states of molecules using quantum machine learning, arXiv:2210.05489.

[12] D. Poulin and P. Wocjan, Preparing ground states of quantum many-body systems on a quantum computer, Phys. Rev. Lett. **102**, 130503 (2009).

[13] L. Lin and Y. Tong, Near-optimal ground state preparation, Quantum **4**, 372 (2020).

[14] S. Jansen, M.-B. Ruskai, and R. Seiler, Bounds for the adiabatic approximation with applications to quantum computation, J. Math. Phys. **48**, 102111 (2007).

[15] N. Wiebe and N. S. Babcock, Improved error-scaling for adiabatic quantum evolutions, New J. Phys. **14**, 013024 (2012).

[16] D. Wecker, M. B. Hastings, N. Wiebe, B. K. Clark, C. Nayak, and M. Troyer, Solving strongly correlated electron models on a quantum computer, Phys. Rev. A **92**, 062318 (2015).

[17] M. Reiher, N. Wiebe, K. M. Svore, D. Wecker, and M. Troyer, Elucidating reaction mechanisms on quantum computers, Proc. Natl. Acad. Sci. USA **114**, 7555 (2017).

[18] J. S. Kottmann and A. Aspuru-Guzik, Optimized low-depth quantum circuits for molecular electronic structure using a separable-pair approximation, Phys. Rev. A **105**, 032449 (2022).

[19] H. G. A. Burton, D. Marti-Dafcik, D. P. Tew, and D. J. Wales, Exact electronic states with shallow quantum circuits through global optimisation, njp Quantum Inf. **9**, 75 (2023).

[20] G. Wang, S. Sim, and P. D. Johnson, State preparation boosters for early fault-tolerant quantum computation, Quantum **6**, 829 (2022).

[21] A. Auffèves, Quantum technologies need a quantum energy initiative, PRX Quantum **3**, 020101 (2022).

[22] S. Pathak, A. E. Russo, S. K. Seritan, and A. D. Baczewski, Quantifying t-gate-count improvements for ground-state-energy estimation with near-optimal state preparation, Phys. Rev. A **107**, L040601 (2023).

[23] A. Katabarwa, K. Gratsea, A. Caesura, and P. D. Johnson, Early fault-tolerant quantum computing, arXiv:2311.14814.

[24] T. Albash and D. A. Lidar, Adiabatic quantum computation, Rev. Mod. Phys. **90**, 015002 (2018).

[25] R. Babbush, C. Gidney, D. W. Berry, N. Wiebe, J. McClean, A. Paler, A. Fowler, and H. Neven, Encoding electronic spectra in quantum circuits with linear T complexity, Phys. Rev. X **8**, 041015 (2018).

[26] D. W. Berry, B. L. Higgins, S. D. Bartlett, M. W. Mitchell, G. J. Pryde, and H. M. Wiseman, How to perform the most accurate possible phase measurements, Phys. Rev. A **80**, 052114 (2009).

[27] D. W. Berry, A. M. Childs, and R. Kothari, Hamiltonian simulation with nearly optimal dependence on all parameters, in *2015 IEEE 56th Annual Symposium on Foundations of Computer Science* (IEEE, New York, 2015).

[28] J. S. Kottmann, Molecular quantum circuit design: A graph-based approach, Quantum **7**, 1073 (2023).

[29] P. Schleich, J. Boen, L. Cincio, A. Anand, J. S. Kottmann, S. Tretiak, P. A. Dub, and A. Aspuru-Guzik, Partitioning quantum chemistry simulations with Clifford circuits, J. Chem. Theory Comput. **19**, 4952 (2023).

[30] J. S. Kottmann, P. Schleich, T. Tamayo-Mendoza, and A. Aspuru-Guzik, Reducing qubit requirements while maintaining

numerical precision for the variational quantum eigensolver: A basis-set-free approach, J. Phys. Chem. Lett. **12**, 663 (2021).

[31] J. S. Kottmann, S. Alperin-Lea, T. Tamayo-Mendoza, A. Cervera-Lierta, C. Lavigne, T.-C. Yen, V. Verteletskyi, P. Schleich, A. Anand, M. Degroote, S. Chaney, M. Kesibi, N. G. Curnow, B. Solo, G. Tsilimigkounakis, C. Zendejas-Morales, A. F. Izmaylov, and A. Aspuru-Guzik, TEQUILA: a platform for rapid development of quantum algorithms, Quantum Sci. Technol. **6**, 024009 (2021).

[32] A. G. Fowler and S. J. Devitt, A bridge to lower overhead quantum computation, arXiv:1209.0510.

[33] K. Gratsea, J. S. Kottmann, P. D. Johnson, and A. A. Kunitsa, Comparing classical and quantum ground state preparation heuristics, arXiv:2401.05306.

[34] J. F. Gonthier, M. D. Radin, C. Buda, E. J. Doskocil, C. M. Abuan, and J. Romero, Measurements as a roadblock to near-term practical quantum advantage in chemistry: Resource analysis, Phys. Rev. Res. **4**, 033154 (2022).