Efficient quantum computation of chemistry through tensor hypercontraction

Joonho Lee,[*] Dominic W. Berry,[†] Craig Gidney,[‡] William J. Huggins,[‡] Jarrod R. McClean,[‡] Nathan Wiebe,[§] and Ryan Babbush[‡]

We show how to achieve the highest efficiency yet for simulations with arbitrary basis sets by using a representation of the Coulomb operator known as tensor hypercontraction (THC). We use THC to express the Coulomb operator in a non-orthogonal basis, which we are able to block encode by separately rotating each term with angles that are obtained via QROM. Our algorithm has the best complexity *scaling* for an arbitrary basis, as well as the best complexity for the specific case of FeMoCo. By optimising the surface code resources, we show that FeMoCo can be simulated using about 4 million physical qubits and 3.5 days of runtime, assuming 1 μs cycle times and physical gate error rates no worse than 0.1%.

Background and contribution: The computation of quantum chemistry is regarded as one of the most promising applications of quantum computers [1–3]. This is because there are many applications of quantum chemistry that pertain to the development of practical technologies and, for at least some of these applications, quantum algorithms appear to provide an exponential scaling advantage relative to the best known classical approaches [4]. Specifically, most algorithmic work in this area focuses on the construction of quantum circuits enabling precise estimation of the ground state energy, which is important to predict properties such as reaction barrier heights and non-covalent interaction energies [5]. The holy grail of quantum chemistry is to have a generally applicable electronic structure method that yields relative energies with errors less than 1.6 mHa.

An important challenging problem in chemistry is understanding the mechanism of biological nitrogen fixation [6]. The key to understanding this is believed to be in the electronic structure of the primary cofactor, FeMoCo, which is currently intractable to classically simulate. Reiher *et al.* [7] proposed a crude active space model of FeMoCo, deployed a Trotter-based approach and required more than 10^{14} T gates to simulate the model. Work in this area focuses on counting the required number of non-Clifford (T or Toffoli) gates, because within the surface code [8] these gates require significantly more resources. With 0.1% physical error rates, just distilling the magic states needed for the Reiher *et al.* FeMoCo calculation would require a decade with 4 million qubits.

More recent work employs a qubitisation based approach [9, 10], and improves considerably over the Trotter-based methods of [7]. In [11], presented at QIP 2020, we gave two methods. One used a low-rank factorisation of the Coulomb operator resulting in lower asymptotic complexity, and the other exploited sparsity in the Hamiltonian, yielding 3 orders of magnitude improvement for FeMoCo. Work by von Burg *et al.* [12] further improved upon our complexity by performing a second factorisation of the Coulomb operator, resulting in a crucial reduction in λ (the 1-norm of Hamiltonian coefficients) which governs the complexity. The challenge with second factorisation is to apply the basis rotations for each component of the factorisation, which they achieve by using QROM to output a database of angles, then performing Givens rotations from that database.

A drawback to the von Burg *et al.* approach is that it requires a large number of angles to be output by the QROM. We overcome that drawback by using the THC representation. Via the THC representation we are able to express the Coulomb operator in terms of number operators for a larger non-orthogonal basis. We overcome the problem of non-orthogonality by performing individual rotations in a superposition for each of the individual terms in the decomposition. We employ a QROM to output the rotation angles in a similar way as von Burg *et al.*, but the number of rotations needed is orders of magnitude smaller. At the same time we obtain a value of λ

[*] Department of Chemistry, Columbia University, New York, NY, USA
[†] Department of Physics and Astronomy, Macquarie University, Sydney, NSW 2109, Australia
[‡] Google Research, Venice, CA 90291, United States
[§] Department of Physics, University of Washington; Pacific Northwest National Laboratory

that is similar for FeMoCo, and smaller asymptotically, resulting in lower overall complexity. Our approach also considerably simplifies the state preparation, which needs multiple steps in [12].

A further improvement we make in our analysis is to more carefully account for the bits of precision needed for Hamiltonian coefficients and rotations. Our insight is that the THC representation of the Hamiltonian, as well as finite precision of the coefficients and rotations, all contribute to the approximation of the Hamiltonian. Therefore, the net contribution to the error can be found by computing the Hamiltonian with all these approximations and comparing to the exact Hamiltonian. To estimate the error in the energy from this approximation, we then compute the energy for both the exact and approximate Hamiltonians using the CCSD(T) approach [13].

Results: Using CCSD(T) we estimated the number of bits needed to keep the error due to approximation of the Hamiltonian below 0.6 mHa, leaving 1 mHa for the phase estimation error. We computed the Toffoli complexity for the THC method for FeMoCo for the Reiher *et al.* Hamiltonian, as well as for the more physically sound FeMoCo active space model Hamiltonian introduced by Li *et al.* [14]. We also recomputed the Toffoli complexity for the von Burg *et al.* double factorisation approach, as well as the single factorisation and sparse approaches from [11]. For these other approaches we used the same number of bits, as well as allowing the same 1 mHa error for the phase estimation, in order to give the fairest possible comparison.

The results are given in Table I, where it can be seen that our new THC approach provides the lowest complexity for both FeMoCo Hamiltonians, and about half that for the double factorisation approach. That is even after we have reduced the number of Toffoli gates for the von Burg *et al.* approach below what was quoted in [12]. For the Reiher Hamiltonian, the THC method would bring the computation time with four million qubits down to only 3.5 days. An interesting feature of these results is that for the more accurate Li Hamiltonian, the double factorisation approach actually requires *more* Toffolis than our previous sparse approach from [11]. That is because the double factorisation provides only a modest improvement to λ in that case.

Our THC method provides the best asymptotic scaling of complexities out of prior methods as well. The scalings all depend on the value of λ, which depends on the system and is different for the different methods making them difficult to compare. To more accurately compare them, we performed numerics on hydrogen systems of a range of sizes, fitting to determine the scaling of λ with N. As a result, we found that the THC method improves on double factorisation by a factor of $N^{0.7}$ in the continuum limit, or $N^{1.3}$ in the thermodynamic limit.

Algorithm	Reiher *et al.* FeMoCo [7]		Li *et al.* FeMoCo [14]	
	logical qubits	Toffoli count	logical qubits	Toffoli count
Berry *et al.* [11] (single factorisation)	3,320	9.5×10^{10}	3,628	1.2×10^{11}
Berry *et al.* [11] (sparse)	2,190	8.8×10^{10}	2,489	4.4×10^{10}
von Burg *et al.* [12] (double factorisation)	3,725	1.0×10^{10}	6,404	6.4×10^{10}
this work (tensor hypercontraction)	2,142	5.3×10^{9}	2,196	3.2×10^{10}

TABLE I. Here we report the finite resources required for various recent algorithms to quantum phase estimate two FeMoCo active space models to within chemical accuracy (1.6 mHa).

Methods: The Hamiltonian in an arbitrary second-quantised basis can be expressed as

$$H = T + V, \qquad T = \sum_{\sigma\in\{\uparrow,\downarrow\}} \sum_{p,q=1}^{N/2} T_{pq} a^\dagger_{p,\sigma} a_{q,\sigma}, \qquad V = \frac{1}{2} \sum_{\alpha,\beta\in\{\uparrow,\downarrow\}} \sum_{p,q,r,s=1}^{N/2} V_{pqrs} a^\dagger_{p,\alpha} a_{q,\alpha} a^\dagger_{r,\beta} a_{s,\beta}, \quad (1)$$

where N is the number of spin-orbital basis functions used to discretise the Hamiltonian, T and V are the one- and two-body operators respectively, and $a^\dagger_{p,\sigma}$ and $a_{p,\sigma}$ are fermionic raising and

lowering operators. The THC representation [15–17] is

$$V \approx G = \frac{1}{2} \sum_{\alpha,\beta\in\{\uparrow,\downarrow\}} \sum_{p,q,r,s=1}^{N/2} G_{pqrs} a^\dagger_{p,\alpha} a_{q,\alpha} a^\dagger_{r,\beta} a_{s,\beta}, \qquad G_{pqrs} = \sum_{\mu,\nu=1}^{M} \chi^{(\mu)}_p \chi^{(\mu)}_q \zeta_{\mu\nu} \chi^{(\nu)}_r \chi^{(\nu)}_s, \quad (2)$$

where $\chi^{(\mu)}_p$ and $\zeta_{\mu\nu} = \zeta_{\nu\mu}$ are real scalars, and empirical studies [15–37] suggest that error $\epsilon_{\rm THC}$ can be obtained with $M = \mathcal{O}\left(N \,\mathrm{polylog}\left(1/\epsilon_{\rm THC}\right)\right)$.

We can provide a diagonal representation by re-expressing the Hamiltonian in a basis $c_{\mu,\sigma} = \sum_{p=1}^{N/2} \chi^{(\mu)}_p a_{p,\sigma}$ on a larger space of $2M$ spin-orbitals. Without loss of generality, we take $\chi^{(\mu)}$ to be a normalised vector for each μ. Using this, we rewrite Eq. (2) as

$$G = \frac{1}{2} \sum_{\alpha,\beta\in\{\uparrow,\downarrow\}} \sum_{\mu,\nu=1}^{M} \zeta_{\mu\nu} n_{\mu,\alpha} n_{\nu,\beta} \quad (3)$$

where $n_{\mu,\sigma} = c^\dagger_{\mu,\sigma} c_{\mu,\sigma}$ is the number operator in the larger basis.

In order to measure the energy, we apply the technique of constructing a quantum walk based on expressing the Hamiltonian as a linear combination of unitaries (LCU), which provides an operator with eigenvalues that are a function of those for the Hamiltonian [10]. Phase estimation on this quantum walk therefore provides an estimate of the eigenvalues of the Hamiltonian. We allow the phase estimation to be based on an arbitrary number of repetitions, rather than just a power of two. To achieve this, we perform unary iteration [38] on the control register.

To apply the block encoding of the Hamiltonian as a linear combination of unitaries, rewrite the Hamiltonian via a Jordan-Wigner transformation in the form

$$H = -\frac{1}{2} \sum_{\sigma\in\{\uparrow,\downarrow\}} \sum_{\ell=1}^{N/2} t_\ell U^\dagger_{T,\ell} Z_{1,\sigma} U_{T,\ell} + \frac{1}{8} \sum_{\alpha,\beta\in\{\uparrow,\downarrow\}} \sum_{\mu,\nu=1}^{M} \zeta_{\mu\nu} U^\dagger_\mu Z_{1,\alpha} U_\mu U^\dagger_\nu Z_{1,\beta} U_\nu \quad (4)$$

where the operations U_μ and $U_{T,\ell}$ are rotations into the appropriate basis, and $Z_{1,\alpha}$ is a Z operation on the qubit representing spatial orbital 1 for spin α. We are able to prepare the state efficiently using methods we introduced in [38], using QROM together with a quantum form of alias sampling. A further improvement we make is to compute the contiguous register more efficiently by summing triples of bits, similar to the Hamming weight sums in [39].

To perform the controlled "select" operations needed for block encoding, we use the fact that the basis rotations U_μ needed for the non-orthogonal basis can be performed independently for each μ, eliminating the difficulty involved with the basis being non-orthogonal. Each of these rotations only needs $N/2$ Givens rotations, which are output by a QROM in a similar way as for the method of von Burg *et al.*, but here there are only M values of μ, so this data can be output with Toffoli complexity M. With FeMoCo, we find that M can be taken to be 350 and 450 for the Reiher and Li Hamiltonians respectively, which means this QROM has a relatively small complexity.

The rotations U_μ can then be performed by using qubit rotations implemented by addition into a phase gradient register of the rotation angle output by the QROM. Then a Z gate is performed, and the rotations and QROM are inverted. This step needs to be performed twice, once for ν and once for μ in Eq. (4). A complication is that it is also necessary to apply the one-body term in Eq. (4). If that were performed in a separate step, as suggested in [12], it would significantly increase the complexity. Instead, we combine that part into the first part, $U^\dagger_\nu Z_{1,\beta} U_\nu$, for the two-body term (but not the second part $U^\dagger_\mu Z_{1,\alpha} U_\mu$). The only increase in the complexity is that there are $M + N/2$ sets of rotations that need to be output, instead of M. A further complication is that the LCU needs to be applied in a self-inverse way. We are able to do this with only a few additional Toffolis, despite the asymmetry resulting from including the one-body term.

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