Molecular Quantum Circuit Design

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Science is rich in abstract concepts capturing the essence of complex processes in astonishingly simple ways. In Chemistry, one of the most prominent examples is the reduction of molecules to simple graphs with the atomic nuclei as vertices connected by edges representing so-called chemical bonds.[1] Developing similar abstract concepts is crucial to efficiently construct and analyze parametrized circuits which prepare approximations to molecular (ground) states. Despite various efforts, there is still no method to efficiently construct such circuits.[2] In addition, the importance and exact arrangement of the individual building blocks of existing construction schemes is often based on heuristics with a hard to predict range of applicability.[3] This work introduces a design principle for parametrized quantum circuits based on chemical graphs, providing a way forward in three major obstacles in quantum circuit design for molecular systems: Operator ordering, parameter initialization and initial state preparation. Here, the chemical graph is key for physical interpretation of each individual component of the circuit, and it provides an heuristic to qualitatively estimate the difficulty of preparing ground states for individual instances of molecules.
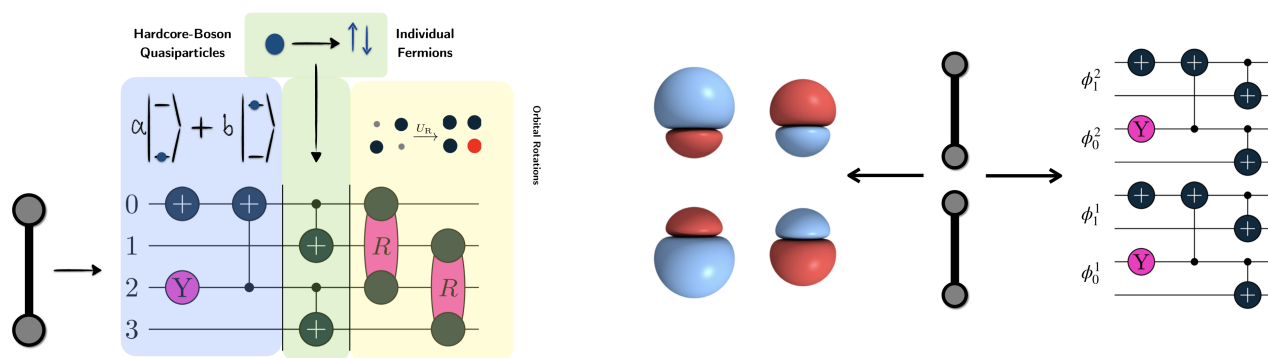

Figure 1: **High-Level Concepts**:
Left: Circuit that prepares a two-electron function representing a single edge in a molecular graph leveraging physical concepts. Here the orbital rotations are explicitly incorporated into the circuit. Right: Initial assumptions about optimal orbitals and a corresponding circuit topology are directly determined from a chemical graph.

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[1]Although there has been - and up to this day still is - a vibrant discussion on what a chemical bond is, it is usually depicted as an electron pair localized between two atomic nuclei. In the most simplistic picture this can be reduced to an edge connecting two vertices.

[2]Existing methods either suffer from non-beneficial gate counts/circuit depth, parameter counts, convergence properties or classical overheads.

[3]Explicit examples are discussed in Section II of [1].

Single Edges - Elementary Building Blocks: An individual edge is the basic building block of a molecular graph. This gives us a natural starting point for the design of quantum circuits. Some of the most prominent model systems in the literature on variational quantum algorithms are molecules representable by a single edge. In particular those are the hydrogen molecule H_2, the lithium hydride molecule LiH and the H_3^+ cation. The construction of quantum circuits for a single edge therefore not only functions as building block for larger graphs but also shows how to efficiently prepare ground states for these prominent model systems - explicitly showing that they can be efficiently solved with classically tractable quantum circuits. A single edge represents a two-electron wavefunction in N_o orbitals which under most encodings results in a $2N_o$ qubit representation.[4] A quantum circuit for a single edge can be constructed from three parts:

1. A hard-core Boson representation where the electron pair is treated as a quasiparticle occupying one of the N_o orbitals. This representation allows an efficient construction of shallow-depth and reasonably local quantum circuits, expressible enough to reach all possible superpositions of the N_o states of the quasiparticle.[5]

2. A unitary transfering from the hard-core Boson to a Fermionic representation. If Fermions are represented via Jordan-Wigner this is a simple sequence of local CNOT operations.

3. A series of Given's rotations corresponding to orbital rotations.[6] Note that through this correspondence the last block of Given's rotations can be absorbed into the Hamiltonian by simply constructing an updated electronic Hamiltonian with rotated orbitals.

Without explicit representation of the orbital rotations in the circuit this leaves us with a linear-depth and local quantum circuit. As the orbital rotations can be absorbed into the Hamiltonian there are only N_o possible states left to represent, rendering the corresponding wavefunction classically simulable. The circuit parameters of the individual edges can therefore always be (pre-)optimized classically.

Single Graphs - Classically Tractable Initial States: A quantum circuit U_G corresponding to a single chemical graph $G = (V, E)$ can be constructed as a tensor product of the circuits U_e representing the individual edges $e \in E$ in the graph

$$U_G = \bigotimes_{e \in E} U_e. \tag{1}$$

As this is a product of electron pairs, this type of circuit is also referred to as separable pair approximation. Since the individual components are classically tractable with linear memory requirement, this property also holds for the assembled U_G circuit. In the same way, the resulting circuits remain local and shallow in depth.[7] In practice these circuits give accurate results whenever the chemical description of a molecule in terms of a single graph is valid.[8] For each instance of a molecule, one can draw several possible graphs whose relative importance usually cannot be quantified explicitly. A qualitative assessment is however possible in most cases.[9] One might argue, that in fact the wavefunction prepared by the circuit U_G quantifies the graph[10] and not the other way around.

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[4] For simplicity, the Jordan-Wigner encoding is employed in the context of this work. The developed design principles are however independent of the qubit encoding.

[5] See Eq.(11-12) in [1] for a circuit with $N_o > 2$ orbitals, and Section II as well as Fig. 1 in [2] for details on the construction.

[6] See Appendix B of [1] for more details.

[7] See Tab.II in [2].

[8] See Figs. 5 and 7 in [2].

[9] For explicit examples see Section II of [1].

[10] For example through its fidelity with respect to the exact ground state, or via the energy expectation value

Multiple Graphs - Challenging Molecular Systems: Ground states for instances of molecular systems that are not described accurately by a single molecular graph are more challenging to prepare by simple quantum circuits. An intuitive example are the two graphs shown in Fig. 2 that can represent different molecules such as BeH_2, H_4, or the π-system of a C_2H_4. Take for example BeH_2: Around the equilibrium Be-H bond distance, the description of the wavefunction is dominated by a single graph representing the two Be-H bonds. The same holds true for the dissociated molecule where the dominating graph represents the individual atoms. In between these two scenarios there is however a small regime where both graphs become important for the accurate description of the molecule. In this case, a multi-Graph circuit can be constructed by adding further operations to the circuit U_G representing the first graph

$$U = U_G U_R U_C U_R^\dagger, \tag{2}$$

with U_C assembled from electronic correlators[11] and U_R consisting of a series of Given's rotations that rotate the orbitals such that they represent the second graph.' The structure of the second graph is hereby crucial as it provides the essential information on how to place and initialize the individual components of the U_R and U_C blocks. The effect can be seen by comparing the optimized energies of the graph-based circuits with standard methods as k-UpCCGSD that are built from the same elementary operations. Here the graph based construction is outperforming this conventional method in several metrics.[12] In the same way an adaptive approach with an operator pool constructed from those elementary operations is not able to produce similar results to the graph based construction either.[13] An intuitive explanation is, that an adaptive procedure circuit can not locally detect the $U_R U_C U_R^\dagger$ pattern, as for example even the full set of individual rotations in U_R alone will not affect the energy.
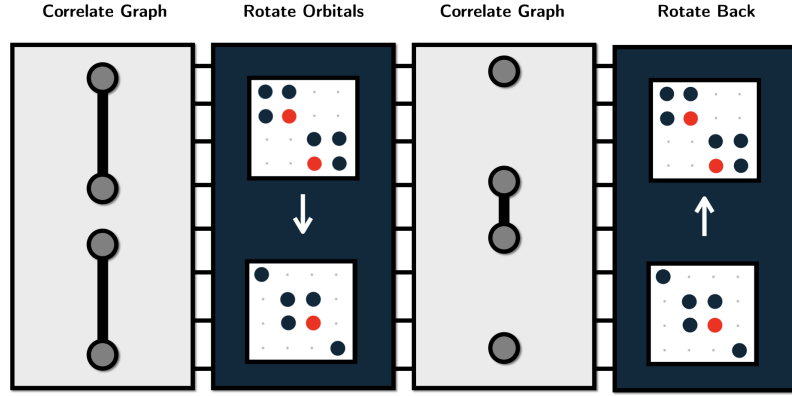

Figure 2: **Multi-Graph Approach**: The description of challenging molecules is not dominanted by a single molecular graph. An example are the equidistant linear H_4 and BeH_2 molecules as well as the π-system of C_2H_4, all represented by the two graphs in the figure. The topology of the quantum circuit should be designed to reflect correlation within the individual graphs. A series of unitary Given's rotations (corresponding to orbital rotations) transfers between the two graph representation. In the figure the orbitals are abstractly represented as blue and red dots placed on the atomic positions. Here the colors represent relative signs similar to the orbitals depicted in Fig. 1

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[11]The simplest correlator would be an electron-pair correlator that excites spin-paired electrons between doubly occupied orbitals. See Eq.(22) in [1].

[12]Such as energy, fidelity, depth, number of variables and iterations. See Tabs. I and II in [1] for explicit results.

[13]See Tab. I in [1]

References

[1] Jakob S. Kottmann. Molecular Quantum Circuit Design: A Graph-Based Approach. 2022. doi: 10.48550/arxiv.2207.12421. URL https://arxiv.org/abs/2207.12421.

[2] Jakob S. Kottmann and Alán Aspuru-Guzik. Optimized low-depth quantum circuits for molecular electronic structure using a separable-pair approximation. *Phys. Rev. A*, 105:032449, Mar 2022. doi: 10.1103/PhysRevA.105.032449. URL https://link.aps.org/doi/10.1103/PhysRevA.105.032449.