1 Introduction to quantum algorithmic approaches

In this section we will attempt to construct quantum algorithms for calculating three of the GFN2-xTB energy terms: E^{Γ}, E^{γ} and E^{EHT}. We will showcase three different approaches to doing a calculation as a building block of a larger circuit.

The conceptually simplest approach is to directly translate classical mathematical circuits to the quantum world using ancillary qubits to ensure reversibility. Here all the computation happens in the state, and the result is readable in the bits of the measurement output. This approach has seen some development beyond this simple translation resulting in some very qubit efficient primitives for multiplication and addition in particular. This approach will be applied to the E^{Γ} and E^{γ} terms, and be referred to as Quantum Digital Arithmetic in this report.

Our second approach is Quantum Amplitude Arithmetic. In this approach we try to prepare the desired result not as a easily read measurement result, but as part of the amplitude of a state. We will use this approach for the E^{Γ} term to prepare a qubit in the state $w|0\rangle + \alpha|0\rangle$ where we can choose α to be proportional to the E^{Γ} of the molecule. Alternatively we can choose α to be proportional to $E^{\Gamma} - E^{\Gamma}_H$ where E^{Γ}_H is the E^{Γ} energy for some known high energy isomer. This is not something we imagine being a common thing to want, however it is something which we want for the total energy! The issue that is solved by subtracting a known high energy is the following. Say we know all the energies, and we want to do something with those states that have a low energy. If all the energies lie between 100 and 101 (units not important), which may make a large difference, the relative difference is not large. If we subtract a known high energy of say 100.9 we get much larger relative differences where the low energy isomers will have a much larger α than the high energy isomers.

The final algorithmic approach we will explore uses quantum singular value transformations. In this approach the calculations are being carried out in the singular values of block encoded matrices. We will use this approach to calculate the E^{ETH} term, as it involves a lot of matrix multiplications. This is well suited for this approach.

For all of these approaches we will assume that we have access to some pretty intricately prepared states. We will not go into how the are prepared other than the fact that classically we can generate the geometries and so for entire isomer spaces without any other information. As any classical computation in theory also can be applied to a quantum computer after modifications it is a possibility to prepare these states.

2 GFN2-xTB E^{Γ}

2.1 Quantum Digital Arithmetic

$$E^{\Gamma} = \frac{1}{3}\sum_{A}\sum_{\mu\in A}(q_{A,\mu})^3\Gamma_{A,\mu} \tag{1}$$

where $q_{A,\nu} = \sum_B \sum_{\nu\in B} P_{\mu\nu}S_{\mu\nu}$ is the partial charge of shell μ associated with atom A. P, S are the density and overlap matrices. $\Gamma_{A,\mu} = \Gamma_A K_\mu$ is just the product of an element specific constant and a shell specific constant, for our purposes the element is always carbon and the shell is either the first or second in GFN2 thus we have 2 numbers $\Gamma_{\text{Carbon},0(1)}$ henceforth referred to as $\mathit{\Gamma}_{0(1)}$.

Let us first rewrite the inner expression a bit given our new definition and knowledge of the atoms we are working with.

$$\sum_{\mu\in A}(q_{A,\mu})^3\Gamma_{A,\mu} = \sum_{\mu\in\{0,1\}}(q_{A,\mu})^3\mathit{\Gamma}_{\mu} \tag{2}$$

We now need a unitary which computes this function on a given state $|q_{A,\mu}\rangle_Q |\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \rightarrow |q_{A,\mu}\rangle_Q |\mathit{\Gamma}_\mu\rangle_\Gamma |acc + (q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_E$. Consider having access to the following addition (multiplication) unitary $|A\rangle|B\rangle \rightarrow |A\rangle|A + (*)B\rangle$. We can now apply the following unitary $E^{\Gamma}_{i\ (Q,\Gamma,E)} = \text{MULT}^{\dagger^3}_{Q,\Gamma}\text{ADD}_{\Gamma,E}\text{MULT}^{3}_{Q,\Gamma}$.

$$E^{\Gamma}_{i\ (Q,\Gamma,E)} |q_{A,\mu}\rangle_Q |\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \tag{3}$$

$$= \text{MULT}^{\dagger^3}_{Q,\Gamma}\text{ADD}_{\Gamma,E}\text{MULT}^{3}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \tag{4}$$

$$= \text{MULT}^{\dagger^3}_{Q,\Gamma}\text{ADD}_{\Gamma,E}\text{MULT}^{2}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |q_{A,\mu}\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \tag{5}$$

$$= \text{MULT}^{\dagger^3}_{Q,\Gamma}\text{ADD}_{\Gamma,E}\text{MULT}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |(q_{A,\mu})^2\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \tag{6}$$

$$= \text{MULT}^{\dagger^3}_{Q,\Gamma}\text{ADD}_{\Gamma,E} |q_{A,\mu}\rangle_Q |(q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_\Gamma |acc\rangle_E \tag{7}$$

$$= \text{MULT}^{\dagger^3}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |(q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_\Gamma |acc + (q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_E \tag{8}$$

$$= \text{MULT}^{\dagger^2}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |(q_{A,\mu})^2\mathit{\Gamma}_\mu\rangle_\Gamma |acc + (q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_E \tag{9}$$

$$= \text{MULT}^{\dagger}_{Q,\Gamma} |q_{A,\mu}\rangle_Q |q_{A,\mu}\mathit{\Gamma}_\mu\rangle_\Gamma |acc + (q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_E \tag{10}$$

$$= |q_{A,\mu}\rangle_Q |\mathit{\Gamma}_\mu\rangle_\Gamma |acc + (q_{A,\mu})^3\mathit{\Gamma}_\mu\rangle_E \tag{11}$$

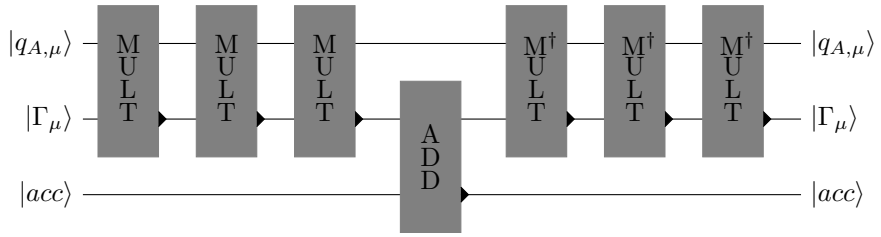
$$\tag{12}$$

This circuit to compute the inner sum in E^{Γ} could be called $E^{\Gamma}_{i\ (Q,\Gamma,E)}$. Let us say we are given a circuit, SDA, for encoding a molecule from its ID in the following manner.

$$\text{SDA}\,|ID\rangle_{ID}\,|0\rangle = |ID\rangle_{ID}\,|0\rangle_E\,|0\rangle_C\,|0\rangle_W \bigotimes_{A_n\in\text{Atoms}} \left(|q_{A_n,0}\rangle_{Q_{n,0}}\,|q_{A_n,1}\rangle_{Q_{n,1}}\right)|\mathit{\Gamma}_0\rangle_{\Gamma_0}\,|\mathit{\Gamma}_1\rangle_{\Gamma_1} \tag{13}$$

We can now apply $\mathrm{DA} = \prod_{\mu\in\{0,1\}} \prod_{A_n} E_{i\ (Q_{n,\mu},\Gamma_\mu,E)}^{\Gamma}$ to get

$$\begin{aligned}\mathrm{DA}\,|ID\rangle_{ID}\,|0\rangle_E\,|0\rangle_C\,|0\rangle_W \bigotimes_{A_n}\left(|q_{A_n,0}\rangle_{Q_{n,0}}\,|q_{A_n,1}\rangle_{Q_{n,1}}\right)|\Gamma_0\rangle_{\Gamma_0}\,|\Gamma_1\rangle_{\Gamma_1} = \\ |ID\rangle_{ID}\,|E^{\Gamma}\rangle_E\,|0\rangle_C\,|0\rangle_W \bigotimes_{A_n}\left(|q_{A_n,0}\rangle_{Q_{n,0}}\,|q_{A_n,1}\rangle_{Q_{n,1}}\right)|\Gamma_0\rangle_{\Gamma_0}\,|\Gamma_1\rangle_{\Gamma_1}\end{aligned} \tag{14}$$



2.2 Sampling using Quantum Amplitude Arithmetic

Assume that we are given an equal superposition of all the canonical IDs of the fullerenes in an isomer-space. We can apply SDA to set up the encoding and then apply DA. We now have computed the E^{Γ} energies for every isomer. However we can only sample once! Let us say that we are interested in the isomers with the lowest energies. We then would like the probability of sampling an isomer to be proportional to E^{Γ}. We can achieve this using Quantum Amplitude Arithmetic, not to be confused with Quantum Amplitude Amplification, both shortened as QAA but in this writing as QA-Arithmetic and QA-Amplification.

The paper on QA-Arithmetic uses the introduced addition and multiplication primitives to construct a circuit which transforms the state $|x\rangle_D\,|0\rangle_C\,|0\rangle_W \rightarrow \frac{1}{2}\frac{x}{2^n}\,|x\rangle_D\,|0\rangle_C\,|1\rangle_W + \alpha\,|\omega\rangle_{D\otimes C\otimes W}$ where α is some normalization factor, and $|\omega\rangle$ is some state with no overlap with the state containing all 0's in the control register, C, and 1 in the work register, W.

When using this circuit we can treat the E register containing our resulting E^{Γ} term as the data register, D. Let us take a look at that.

$$\begin{aligned}\sum_{ID\in\text{isomers}}\left[|ID\rangle_{ID}\,|E^{\Gamma}_{ID}\rangle_E\,|0\rangle_C\,|0\rangle_W \bigotimes_{A_n}\left(|q_{A_n,0}\rangle_{Q_{n,0}}\,|q_{A_n,1}\rangle_{Q_{n,1}}\right)\right]|\Gamma_0\rangle_{\Gamma_0}\,|\Gamma_1\rangle_{\Gamma_1} \rightarrow \\ \sum_{ID\in\text{isomers}}\left[|ID\rangle_{ID}\left(\frac{1}{2}\frac{E^{\Gamma}_{ID}}{2^n}\,|E^{\Gamma}_{ID}\rangle_E\,|0\rangle_C\,|1\rangle_W + \alpha_{ID}\,|\omega_{ID}\rangle\right)\bigotimes_{A_n}\left(|q_{A_n,0}\rangle_{Q_{n,0}}\,|q_{A_n,1}\rangle_{Q_{n,1}}\right)\right]|\Gamma_0\rangle_{\Gamma_0}\,|\Gamma_1\rangle_{\Gamma_1}\end{aligned} \tag{15}$$

If we now sample from this superposition and postselect for $C = 0$ and $W = 1$ we are more likely to sample a low energy fullerene. The likelihood of sampling a given canonical fullerene ID is proportional with E^{Γ} for that fullerene.

2.3 An alternative circuit using exclusively Quantum Amplitude Arithmetic.

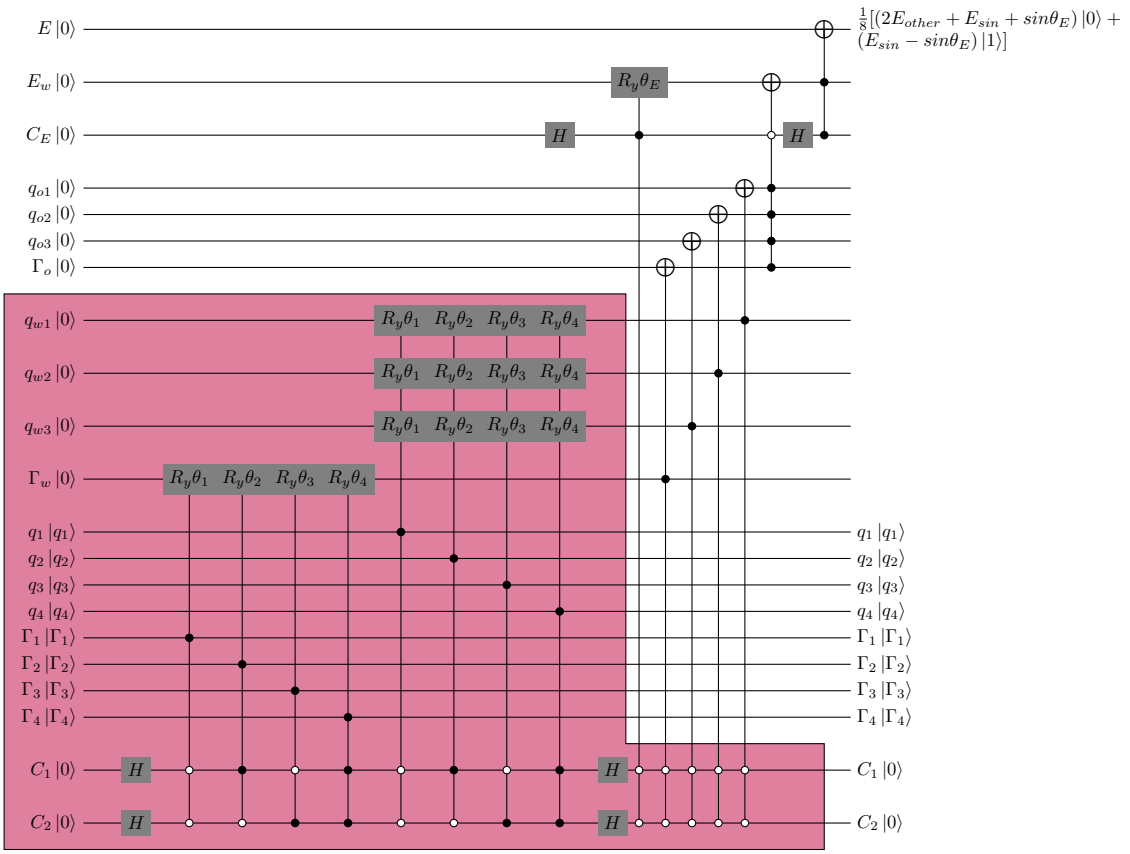
An alternative strategy would be to go all in on QA-Arithmetic and do all the arithmetic in the amplitudes. Here we would encode a molecule as follows

$$\text{SAA}\,|ID\rangle_{ID}\,|0\rangle_{\Gamma\otimes Q\otimes C\otimes W} = |ID\rangle_{ID}\sum_{\mu\in\{0,1\}}|\Gamma_\mu\rangle_\Gamma\sum_{A_n}|q_{A_n,\mu}\rangle_Q\,|0\rangle_C\,|0\rangle_W \quad (16)$$

Where $C = C_1 \otimes C_2 \otimes C_3 \otimes C_4$ and $W = W_1 \otimes W_2 \otimes W_3 \otimes W_4$. We can now apply the QA-Arithmetic papers transformation 4 times. This would be on the $(\Gamma, C_1, W_1), (Q, C_2, W_2), (Q, C_3, W_3), (Q, C_4, W_4)$ registers. This would, if we collect all terms involving ω into one term, yield

$$\alpha_{ID}\,|\omega_{ID}\rangle + \frac{1}{2^4}\,|ID\rangle\sum_{\mu\in\{0,1\}}\frac{\Gamma_\mu}{2^n}\,|\Gamma_\mu\rangle_\Gamma\sum_{A_n}\frac{q^3_{A_n,\mu}}{2^{n^3}}\,|q_{A_n,\mu}\rangle_Q\,|0\rangle_C\,|1\rangle_W \quad (17)$$

If we apply this to a superposition of such molecule encodings and postselect on $C = 0$ and $W = 1$ we would sample a molecule with ID with probability $\propto \sum_{\mu\in\{0,1\}}\sum_{A_n}\frac{\Gamma_\mu}{2^n}\frac{q^3_{A_n,\mu}}{2^{n^3}} = \frac{E^\Gamma_{ID}}{2^{n^4}} \propto E^\Gamma_{ID}$. We can then compute the energy classically.



The controlled gate notation here is the following, t is the target register and $c1, c2, c3, \ldots$ are the control registers. $a, b, c, \ldots$ are all 1 except if there is a bar over the corresponding $c1, c2, c3, \ldots$ in which case it is 0.

$$CU_t^{c1,c2,c3,\cdots} = (U_t - I_t) \otimes |a\rangle\langle a|_{c1} \otimes |b\rangle\langle b|_{c2} \otimes |c\rangle\langle c|_{c3} \otimes \cdots + \sum_{\alpha,\beta,\zeta,\cdots \in \{0,1\}} I_t \otimes |\alpha\rangle\langle\alpha|_{c1} \otimes |\beta\rangle\langle\beta|_{c2} \otimes |\zeta\rangle\langle\zeta|_{c3} \otimes \ldots. \tag{18}$$

Let us follow the execution of the above diagram. We neglect writing out the $q_{1,2,3,4}, \Gamma_{1,2,3,4}$ as they never change throughout the calculation, we also neglect the $_{o,output}$ registers for now. We begin by applying the Hadamard gates.

$$\begin{aligned} H_{C1}H_{C2}\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w}\,|00\rangle_{C_{(1,2)}} \rightarrow \\ \frac{1}{2}(\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w}\,|00\rangle_{C_{(1,2)}} + \\ |0000\rangle_{q_{w(1,2,3)},\Gamma_w}\,|01\rangle_{C_{(1,2)}} + \\ |0000\rangle_{q_{w(1,2,3)},\Gamma_w}\,|10\rangle_{C_{(1,2)}} + \\ |0000\rangle_{q_{w(1,2,3)},\Gamma_w}\,|11\rangle_{C_{(1,2)}}) \end{aligned} \tag{19}$$

We then apply the conditional rotation gates to some register like Γ_w we do the following

$$\begin{aligned}
&CRy_{\Gamma_w}^{\Gamma_4,C_1,C_2}(2\theta_4)CRy_{\Gamma_w}^{\Gamma_3,\bar{C_1},C_2}(2\theta_3)CRy_{\Gamma_w}^{\Gamma_2,C_1,\bar{C_2}}(2\theta_2)CRy_{\Gamma_w}^{\Gamma_1,\bar{C_1},\bar{C_2}}(2\theta_1)\\
&\qquad\qquad \frac{1}{2}\sum_{x_1,x_2\in\{0,1\}} |0000\rangle_{q_{w(1,2,3)},\Gamma_w} |x_1x_2\rangle_{C_{(1,2)}} \rightarrow\\
&\qquad \frac{1}{2}(CRy_{\Gamma_w}^{\Gamma_1}(2\theta_1)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |00\rangle_{C_{(1,2)}} +\\
&\qquad\quad CRy_{\Gamma_w}^{\Gamma_2}(2\theta_2)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |01\rangle_{C_{(1,2)}} +\\
&\qquad\quad CRy_{\Gamma_w}^{\Gamma_3}(2\theta_3)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |10\rangle_{C_{(1,2)}} +\\
&\qquad\quad CRy_{\Gamma_w}^{\Gamma_4}(2\theta_4)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |11\rangle_{C_{(1,2)}}) \rightarrow\\
&\frac{1}{2}(|000\rangle\,[\Gamma 1(cos\theta_1\,|0\rangle + sin\theta_1\,|1\rangle) + (1-\Gamma_1)\,|0\rangle]_{q_{w(1,2,3)},\Gamma_w} |00\rangle +\\
&\quad |000\rangle\,[\Gamma 2(cos\theta_2\,|0\rangle + sin\theta_2\,|1\rangle) + (1-\Gamma_2)\,|0\rangle]_{q_{w(1,2,3)},\Gamma_w} |01\rangle +\\
&\quad |000\rangle\,[\Gamma 3(cos\theta_3\,|0\rangle + sin\theta_3\,|1\rangle) + (1-\Gamma_3)\,|0\rangle]_{q_{w(1,2,3)},\Gamma_w} |10\rangle +\\
&\quad |000\rangle\,[\Gamma 4(cos\theta_4\,|0\rangle + sin\theta_4\,|1\rangle) + (1-\Gamma_4)\,|0\rangle]_{q_{w(1,2,3)},\Gamma_w} |11\rangle)
\end{aligned} \tag{20}$$

Let us adopt the notation $|\Psi_i^t\rangle = t(cos\theta_i\,|0\rangle + sin\theta_i\,|1\rangle) + (1-t)\,|0\rangle$ before redoing the application using our new notation. We also apply the rotation gates for the q_w registers:

$$\begin{aligned}
&CRy_{\Gamma_w}^{\Gamma_4,C_1,C_2}(2\theta_4)CRy_{\Gamma_w}^{\Gamma_3,\bar{C_1},C_2}(2\theta_3)CRy_{\Gamma_w}^{\Gamma_2,C_1,\bar{C_2}}(2\theta_2)CRy_{\Gamma_w}^{\Gamma_1,\bar{C_1},\bar{C_2}}(2\theta_1)\\
&CRy_{q_{w1}}^{q_4,C_1,C_2}(2\theta_4)CRy_{q_{w1}}^{q_3,\bar{C_1},C_2}(2\theta_3)CRy_{q_{w1}}^{q_2,C_1,\bar{C_2}}(2\theta_2)CRy_{q_{w1}}^{q_1,\bar{C_1},\bar{C_2}}(2\theta_1)\\
&CRy_{q_{w2}}^{q_4,C_1,C_2}(2\theta_4)CRy_{q_{w2}}^{q_3,\bar{C_1},C_2}(2\theta_3)CRy_{q_{w2}}^{q_2,C_1,\bar{C_2}}(2\theta_2)CRy_{q_{w2}}^{q_1,\bar{C_1},\bar{C_2}}(2\theta_1)\\
&CRy_{q_{w3}}^{q_4,C_1,C_2}(2\theta_4)CRy_{q_{w3}}^{q_3,\bar{C_1},C_2}(2\theta_3)CRy_{q_{w3}}^{q_2,C_1,\bar{C_2}}(2\theta_2)CRy_{q_{w3}}^{q_1,\bar{C_1},\bar{C_2}}(2\theta_1)\\
&\qquad\qquad \frac{1}{2}\sum_{x_1,x_2\in\{0,1\}} |0000\rangle_{q_{w(1,2,3)},\Gamma_w} |x_1x_2\rangle =\\
&\frac{1}{2}(CRy_{q_{w1}}^{q_1}(2\theta_1)CRy_{q_{w2}}^{q_1}(2\theta_1)CRy_{q_{w3}}^{q_1}(2\theta_1)CRy_{\Gamma_w}^{\Gamma_1}(2\theta_1)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |00\rangle +\\
&\quad CRy_{q_{w1}}^{q_2}(2\theta_2)CRy_{q_{w2}}^{q_2}(2\theta_2)CRy_{q_{w3}}^{q_2}(2\theta_2)CRy_{\Gamma_w}^{\Gamma_2}(2\theta_2)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |01\rangle +\\
&\quad CRy_{q_{w1}}^{q_3}(2\theta_3)CRy_{q_{w2}}^{q_3}(2\theta_3)CRy_{q_{w3}}^{q_3}(2\theta_3)CRy_{\Gamma_w}^{\Gamma_3}(2\theta_3)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |10\rangle +\\
&\quad CRy_{q_{w1}}^{q_4}(2\theta_4)CRy_{q_{w2}}^{q_4}(2\theta_4)CRy_{q_{w3}}^{q_4}(2\theta_4)CRy_{\Gamma_w}^{\Gamma_4}(2\theta_4)\,|0000\rangle_{q_{w(1,2,3)},\Gamma_w} |11\rangle) \rightarrow\\
&\qquad \frac{1}{2}(|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle_{q_{w(1,2,3)},\Gamma_w} |00\rangle +\\
&\qquad\quad |\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle_{q_{w(1,2,3)},\Gamma_w} |01\rangle +\\
&\qquad\quad |\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle_{q_{w(1,2,3)},\Gamma_w} |10\rangle +\\
&\qquad\quad |\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle_{q_{w(1,2,3)},\Gamma_w} |11\rangle)
\end{aligned} \tag{21}$$

Let us now apply the second set of Hadamard:

$$
\begin{aligned}
H_{C_1}H_{C_2}\frac{1}{2}(&|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle|00\rangle+\\
&|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle|01\rangle+\\
&|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle|10\rangle+\\
&|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle|11\rangle)\rightarrow\\
\frac{1}{4}(&|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle[|00\rangle+|01\rangle+|10\rangle+|11\rangle]+\\
&|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle[|00\rangle-|01\rangle+|10\rangle-|11\rangle]+\\
&|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle[|00\rangle+|01\rangle-|10\rangle-|11\rangle]+\\
&|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle[|00\rangle-|01\rangle-|10\rangle+|11\rangle])=\\
\frac{1}{4}&\left[|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle+|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle+|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle+|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle\right]|00\rangle+\\
\frac{1}{4}\Bigg(&\left[|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle-|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle+|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle-|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle\right]|01\rangle+\\
&\left[|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle+|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle-|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle-|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle\right]|10\rangle+\\
&\left[|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle-|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle-|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle+|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle\right]|11\rangle\Bigg)\\
=\frac{1}{4}&\left[|\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{q_1}\Psi_1^{\Gamma_1}\rangle+|\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{q_2}\Psi_2^{\Gamma_2}\rangle+|\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{q_3}\Psi_3^{\Gamma_3}\rangle+|\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{q_4}\Psi_4^{\Gamma_4}\rangle\right]|00\rangle+|M\rangle\\
=&|N\rangle+|M\rangle
\end{aligned}
\tag{22}
$$

Before the next step let us define:

$$
\begin{aligned}
q_{sin} =& q_1 sin\theta_1 + q_2 sin\theta_2 + q_3 sin\theta_3 + q_4 sin\theta_4 && (23)\\
q_{other} =& q_1 cos\theta_1 + q_2 cos\theta_2 + q_3 cos\theta_3 + q_4 cos\theta_4 + 4 - q_1 - q_2 - q_3 - q_4 && (24)\\
\Gamma_{sin} =& \Gamma_1 sin\theta_1 + \Gamma_2 sin\theta_2 + \Gamma_3 sin\theta_3 + \Gamma_4 sin\theta_4 && (25)\\
\Gamma_{other} =& \Gamma_1 cos\theta_1 + \Gamma_2 cos\theta_2 + \Gamma_3 cos\theta_3 + \Gamma_4 cos\theta_4 + 4 - \Gamma_1 - \Gamma_2 - \Gamma_3 - \Gamma_4 && (26)\\
E_{sin} =& \Gamma_{sin}(q_{sin})^3 && (27)\\
E_{other} =& \Gamma_{other}(q_{other}^3 + 3q_{other}^2 q_{sin} + 3q_{other}q_{sin}^2 + q_{sin}^3)\\
&+ \Gamma_{sin}(q_{other}^3 + 3q_{other}^2 q_{sin} + 3q_{other}q_{sin}^2) && (28)\\
|\sigma_t\rangle =& t_{other}|0\rangle + t_{sin}|1\rangle && (29)\\
& && (30)
\end{aligned}
$$

Let us now add in the $_o$ registers and apply the first 4 conditional not gates:

$$CX_{q_{o1}}^{q_{w1},\bar{C}_1,\bar{C}_2}CX_{q_{o2}}^{q_{w2},\bar{C}_1,\bar{C}_2}CX_{q_{o3}}^{q_{w3},\bar{C}_1,\bar{C}_2}CX_{\Gamma_o}^{\Gamma_w,\bar{C}_1,\bar{C}_2}\left|0000\right\rangle_{E_o,q_{o(1,2,3)},\Gamma_o}(\left|N\right\rangle+\left|M\right\rangle)\rightarrow$$
$$\left(CX_{q_{o1}}^{q_{w1}}CX_{q_{o2}}^{q_{w2}}CX_{q_{o3}}^{q_{w3}}CX_{\Gamma_o}^{\Gamma_w}\left|0000\right\rangle\left|N\right\rangle\right)+\left|0000\right\rangle\left|M\right\rangle\rightarrow$$
$$\left|\sigma_q\sigma_q\sigma_q\sigma_\Gamma\right\rangle_{q_{o(1,2,3)},\Gamma_o}\left|N\right\rangle+\left|0000\right\rangle\left|M\right\rangle \tag{31}$$

Now let us disregard everything below the $_o$ registers in the diagram except the C_1, C_2 registers and do the final gates involving the $_o$ registers:

$$H_{C_E}CX_{E_w}^{\bar{C}_E,q_{o1},q_{o2},q_{o3},\Gamma_o}CRy_{E_w}^{C_E,\bar{C}_1,\bar{C}_2}(\theta_E)H_{C_E}\left|000\right\rangle_{E_o,E_w,C_E}\frac{1}{4}\Bigg(\left|\sigma_q\sigma_q\sigma_q\sigma_\Gamma\right\rangle_{q_{o(1,2,3)},\Gamma_o}\left|00\right\rangle_{C_1,C_2}$$
$$+\left|0000\right\rangle_{q_{o(1,2,3)},\Gamma_o}[\left|01\right\rangle+\left|10\right\rangle+\left|11\right\rangle]_{C_1,C_2}\Bigg)\rightarrow$$
$$H_{C_E}\frac{1}{4}\Bigg(\left|0\right\rangle\frac{1}{\sqrt{2}}\Bigg[CX_{E_w}^{q_{o1},q_{o2},q_{o3},\Gamma_o}\left|0\right\rangle\left|0\right\rangle+Ry_{E_w}(\theta_E)\left|0\right\rangle\left|1\right\rangle\Bigg]\left|\sigma_q\sigma_q\sigma_q\sigma_\Gamma\right\rangle_{q_{o(1,2,3)},\Gamma_o}\left|00\right\rangle_{C_1,C_2}$$
$$+\left|00+0000\right\rangle_{E_o,E_w,C_E,q_{o(1,2,3)},\Gamma_o}[\left|01\right\rangle+\left|10\right\rangle+\left|11\right\rangle]_{C_1,C_2}\Bigg)\rightarrow$$
$$H_{C_E}\frac{1}{4}\Bigg(\left|0\right\rangle\frac{1}{\sqrt{2}}\Bigg[\left|\sigma_E\right\rangle_{E_w}\left|0\right\rangle_{C_E}+(cos\theta_E\left|0\right\rangle+\sin\theta_E\left|1\right\rangle)_{E_w}\left|1\right\rangle_{C_E}\Bigg]\left|\sigma_q\sigma_q\sigma_q\sigma_\Gamma\right\rangle_{q_{o(1,2,3)},\Gamma_o}\left|00\right\rangle_{C_1,C_2} \tag{32}$$
$$+\left|00+0000\right\rangle_{E_o,E_w,C_E,q_{o(1,2,3)},\Gamma_o}[\left|01\right\rangle+\left|10\right\rangle+\left|11\right\rangle]_{C_1,C_2}\Bigg)\rightarrow$$
$$\frac{1}{4}\Bigg(\left|0\right\rangle\frac{1}{\sqrt{2}}\Bigg[\left|\sigma_E\right\rangle_{E_w}\left|+\right\rangle_{C_E}+(cos\theta_E\left|0\right\rangle+\sin\theta_E\left|1\right\rangle)_{E_w}\left|-\right\rangle_{C_E}\Bigg]\left|\sigma_q\sigma_q\sigma_q\sigma_\Gamma\right\rangle_{q_{o(1,2,3)},\Gamma_o}\left|00\right\rangle_{C_1,C_2}$$
$$+\left|000000\right\rangle_{E_o,E_w,C_E,q_{o(1,2,3)},\Gamma_o}[\left|01\right\rangle+\left|10\right\rangle+\left|11\right\rangle]_{C_1,C_2}\Bigg)$$

Now we can neglect the $q_{o(1,2,3),\Gamma_o,C_1,C_2}$ registers and do some preparatory manipulations before applying the final conditional not gate.

$$
\begin{aligned}
&\frac{1}{4}\Bigg(|0\rangle \frac{1}{\sqrt{2}}\Bigg[|\sigma_E\rangle_{E_w} |+\rangle_{C_E} + (cos\theta_E |0\rangle + \sin\theta_E |1\rangle)_{E_w} |-\rangle_{C_E} \Bigg] + 3\,|000\rangle \Bigg) = \\
&\quad \frac{1}{4}\Bigg(|0\rangle \frac{1}{2}\Bigg[|\sigma_E\rangle_{E_w} |0\rangle_{C_E} + |\sigma_E\rangle_{E_w} |1\rangle_{C_E} + (cos\theta_E |0\rangle + \sin\theta_E |1\rangle)_{E_w} |0\rangle_{C_E} \\
&\qquad -(cos\theta_E |0\rangle + \sin\theta_E |1\rangle)_{E_w} |1\rangle_{C_E} \Bigg] + 3\,|000\rangle \Bigg) = \\
&\qquad \frac{1}{4}\Bigg(|0\rangle \frac{1}{2}\Bigg[(|\sigma_E\rangle + cos\theta_E |0\rangle + \sin\theta_E |1\rangle)_{E_w} |0\rangle_{C_E} \\
&\qquad\qquad +(|\sigma_E\rangle - cos\theta_E |0\rangle - \sin\theta_E |1\rangle)_{E_w} |1\rangle_{C_E} \Bigg] + 3\,|000\rangle \Bigg) = \\
&\qquad\quad \frac{1}{8}\Bigg(|0\rangle[|\sigma_E\rangle + cos\theta_E |0\rangle + \sin\theta_E |1\rangle]_{E_w} |0\rangle_{C_E} \\
&\qquad\quad + |0\rangle[|\sigma_E\rangle - cos\theta_E |0\rangle - \sin\theta_E |1\rangle]_{E_w} |1\rangle_{C_E} + 6\,|000\rangle \Bigg) = \\
&\qquad\quad \frac{1}{8}\Bigg(|0\rangle[E_{other} |0\rangle + E_{sin} |1\rangle + cos\theta_E |0\rangle + \sin\theta_E |1\rangle]_{E_w} |0\rangle_{C_E} \\
&\qquad\quad + |0\rangle[E_{other} |0\rangle + E_{sin} |1\rangle - cos\theta_E |0\rangle - \sin\theta_E |1\rangle]_{E_w} |1\rangle_{C_E} + 6\,|000\rangle \Bigg) = \\
&\qquad\quad \frac{1}{8}\Bigg(|0\rangle[(E_{other} + cos\theta_E) |0\rangle + (E_{sin} + sin\theta_E) |1\rangle]_{E_w} |0\rangle_{C_E} \\
&\qquad\quad + |0\rangle[(E_{other} - cos\theta_E) |0\rangle + (E_{sin} - sin\theta_E) |1\rangle]_{E_w} |1\rangle_{C_E} + 6\,|000\rangle \Bigg) = \\
&\qquad\qquad \frac{1}{8}\Bigg((E_{other} + cos\theta_E) |000\rangle + (E_{sin} + sin\theta_E) |010\rangle \\
&\qquad\qquad +(E_{other} - cos\theta_E) |001\rangle + (E_{sin} - sin\theta_E) |011\rangle + 6\,|000\rangle \Bigg)
\end{aligned}
\tag{33}
$$

We now apply the final conditional not gate:

$$
\begin{aligned}
CX_{E_o}^{E_w,C_E}\frac{1}{8}\Big(&(E_{other}+cos\theta_E)\,|000\rangle+(E_{sin}+sin\theta_E)\,|010\rangle\\
&+(E_{other}-cos\theta_E)\,|001\rangle+(E_{sin}-sin\theta_E)\,|011\rangle+6\,|000\rangle\Big)\rightarrow\\
\frac{1}{8}\Big(&(E_{other}+cos\theta_E)\,|000\rangle+(E_{sin}+sin\theta_E)\,|010\rangle\\
&+(E_{other}-cos\theta_E)\,|001\rangle+X_{E_o}(E_{sin}-sin\theta_E)\,|011\rangle+6\,|000\rangle\Big)\\
\frac{1}{8}\Big(&(E_{other}+cos\theta_E)\,|000\rangle+(E_{sin}+sin\theta_E)\,|010\rangle\\
&+(E_{other}-cos\theta_E)\,|001\rangle+(E_{sin}-sin\theta_E)\,|111\rangle+6\,|000\rangle\Big)
\end{aligned}
\tag{34}
$$

After applying those gates we see that the amplitude on $|1\rangle_{E_o}$ across the whole state is $\frac{1}{8}(E_{sin}-sin\theta_E)=(\Gamma_1 sin\theta_1+\Gamma_2 sin\theta_2+\Gamma_3 sin\theta_3+\Gamma_4 sin\theta_4)(q_1 sin\theta_1+q_2 sin\theta_2+q_3 sin\theta_3+q_4 sin\theta_4)^3-sin\theta_E$. Let us say we know the E^Γ energy of some high energy molecule in the isomer space $E_H^\Gamma=(0b0.\Gamma_H)(0b0.q_H)^3$ If we specify $\theta_i=arcsin\frac{1}{2^i},\theta_E=arcsin[(0b0.\Gamma_H)(0b0.q_H)^3]$ we get that $E_{sin}=(\frac{\Gamma_1}{2}+\frac{\Gamma_2}{2^2}+\frac{\Gamma_3}{2^3}+\frac{\Gamma_4}{2^8})(\frac{q_1}{2}+\frac{q_2}{2^2}+\frac{q_3}{2^3}+\frac{q_4}{2^8})^3=(0b0.\Gamma_1\Gamma_2\Gamma_3\Gamma_4)(0b0.q_1q_2q_3q_4)^3$ and that $\frac{1}{8}(E_{sin}-sin\theta_E)=\frac{1}{8}[(0b0.\Gamma_1\Gamma_2\Gamma_3\Gamma_4)(0b0.q_1q_2q_3q_4)^3-(0b0.\Gamma_H)(0b0.q_H)^3]$. This is proportional to $E^\Gamma-E_H^\Gamma$!

2.4 Complexity

The second algorithm has, in terms of big O notation, the same complexity as the state preparation introduced in the QA-Arithmetic paper, as it is simply 4 applications of this circuit. The state preparation can be achieved using $O(\log n)$ extra qubits and $O(n\log n)$ Toffoli gates where n is the number of bits used to represent $\Gamma_\mu, q_{A_n,\mu}$. Thus if we have a m bit canonical fullerene ID we end up using on the order of $m+2n+4O(\log n)=O(n+m)$ qubits and $4O(n\log n)=O(n\log n)$ Toffoli gates.

The first circuit on the other hand uses 4 mulitplication circuits, 2 squaring circuits which could just as well be multiplication circuits and an addition circuit. A QFT addition (multiplication) circuit uses $O(n^{2(3)})$ gates and no additional qubits. Thus if we have encoded $\Gamma_\mu, q_{A_n,\mu}, \mu\in\{0,..,l\}, A_n\in\{0,..,o\}$ each using n bits we will need to perform $6lo$ multiplications and lo additions, resulting in $6lo\cdot O(n^3)+lo\cdot O(n^2)=O(lon^3)$ gates, on $m+n+nl+lon=O(m+lon)$ qubits. Additionally we then have to run the state preparation circuit which adds $O(\log n)$ qubits and $O(n\log n)$ gates, however that is not enough to change the asymptotic runtime further.

2.5 Cleaning up ω

We would like to get rid of the $|\omega\rangle$ term in both algorithms to avoid having to post select. We can achieve this with amplitude amplification. To do amplitude amplification we first need to define what a 'good' state is, in our case we know all good states have $|0\rangle_C |1\rangle_W$. Second we need an oracle in terms of a unitary which flips the sign of the good state, i.e. reflecting the state around the bad state, this would be $I - 2|0\rangle_C \langle 0|_C |1\rangle_W \langle 1|_W$, which can be easily implemented with controlled rotations. We also need a circuit which would reflect around the initial state by flipping the sign of it, given that we have a circuit U for preparing the initial state that would be $I - 2U|0\rangle\langle 0|U^\dagger$. In our case the angle between the bad and initial states are $\theta_{DA} = arcsin(\frac{1}{2}\frac{E^\Gamma}{2^n}), \theta_{AA} = arcsin(\frac{1}{2^4}\frac{E^\Gamma}{2^{n4}})$ for the two algorithms. We have to do $\lfloor \frac{\pi}{4\theta} \rfloor$ repetitions to maximize the probability of measuring a good state.

2.6 Concentrating the probabilities on the best candidates

We now have a superposition where the probability of sampling a fullerene is proportional to the energy of that fullerene. But is that ideal? The energies might be quite close to each other in absolute terms. Therefore we would like to exaggerate the difference between them and then sample according to that difference. If we knew what the highest energy was we could just subtract that from every energy calculation thus getting probabilities proportional to how much lower an energy we are working with. Another option is if we expect the energies to be within $100(1-x)\%$ we could subtract ex from every energy calculation where e is the energy from a random fullerene in the isomer-space. This is of cause not as good but quite achievable. In both algorithms we can encode ex in a register and then use a digital subtraction or do a QAA state preparation addition but with all the R_y gates inverted, resulting in a counter-clockwise rotation, in effect subtracting ex.

2.7 Discussion

From the asymptotic resource use the second algorithm is clearly superior, even if some of the multiplications and additions can be run in parallel in the first one. We do have a factor 8 lower chance of getting a useful state out, but this again does not change the asymptotics, as we can just repeat it. QA-Amplification might be possible since we have a very clear "good" state in both algorithms. This would reduce the need for postselection and repetitions. Preparing the initial encoding of the isomer space seems less straight forwards in the second approach than in the first unfortunately.