

# APPLIED QUANTUM ALGORITHMS

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## 1. STRONGLY-CORRELATED SYSTEMS

*In computational chemistry and physics, we are interested in predicting the outcome of experiments, which involve systems that may be defined by the action of a Hamiltonian.*

*A strongly-correlated, or strongly interacting quantum system is a quantum system of many particles acting in an external force-field, where the interactions between particles (described by their Hamiltonian) cannot be treated perturbatively.*

- The quantum state of an  $n$  particle system is a function of the positions  $\vec{r}_i \in \mathbb{R}^3$  of the individual particles and (if necessary) their internal states  $s_i = \{0, \dots, N_S - 1\}$  (also known as quantum numbers).

$$|\Psi\rangle \equiv \Psi(\vec{r}_1, s_1, \vec{r}_2, s_2, \dots) \quad (1.1)$$

- In quantum computing we like to think of the vector  $|\Psi\rangle$  — remember that normalized wavefunctions are also vectors. As a simple example, consider a single particle in a cube of length  $L$  with an internal spin  $s = \pm 1$ .

**Notation 1.** *Note here we have switched between  $s = 0, 1$  and  $s = +1, -1$ . This occurs very often throughout the literature — be aware! Note also that when this switch is made, we always associate  $0 \leftrightarrow +1$  and  $1 \leftrightarrow -1$ . This can be achieved as  $s_{+1,-1} = -1^{s_{0,1}}$ , or  $s_{+1,-1} = 2s_{0,1} - 1$ , and backwards as  $s_{0,1} = (1 - s_{+1,-1})/2$ .*

Then, we may write

$$\Psi(\vec{r}, s) = \frac{1}{N} \sum_{\lambda_x, \lambda_y, \lambda_z=1}^{\infty} \sum_{\sigma=\pm 1} a_{\lambda_x, \lambda_y, \lambda_z, \sigma} \delta_{s, \sigma} \sin(\pi r_x \lambda_x / L) \sin(\pi r_y \lambda_y / L) \sin(\pi r_z \lambda_z / L), \quad (1.2)$$

and our function may be represented as well by the vector

$$|\Psi\rangle \equiv \vec{a} = (a_{1,1,1,-1}, a_{1,1,1,1}, a_{1,1,2,-1}, \dots, a_{\lambda_x, \lambda_y, \lambda_z, s} \dots). \quad (1.3)$$

- An interacting Hamiltonian is one that contains particle-particle interactions (equivalently, its action on one particle depends on another).
- For example, the Hamiltonian

$$H_0 = \sum_i \frac{-\hbar^2}{2m_i} \nabla_i^2 + \sum_i V(\vec{r}_i), \quad (1.4)$$

but in the presence of electrostatic repulsion

$$H = H_0 + \sum_{i,j} \frac{Z_i Z_j}{4\pi\epsilon_0 |r_i - r_j|}, \quad (1.5)$$

this becomes interacting.

- Non-interacting Hamiltonians may typically be solved quite well on classical computers for any potential  $V(\vec{r}_i)$  — namely an efficient representation (or good approximation) may be written down by solving the Schrödinger equation for individual particles, and combining:

$$a_{\lambda_1^1 \lambda_2^1 \lambda_3^1 s_1 \lambda_1^2 \lambda_2^2 \lambda_3^2 s_2} = a_{\lambda_1^1 \lambda_2^1 \lambda_3^1 s_1} a_{\lambda_1^2 \lambda_2^2 \lambda_3^2 s_2}. \quad (1.6)$$

- Interacting Hamiltonians, by comparison, are typically incredibly difficult to solve, as the above equation no longer holds.
- Note that the number of indices in  $a$  grows with the number of particles, and so the number of coefficients grows exponentially.

*Quantum computers are finite, so we must truncate an infinite-dimensional Hilbert space to (just) an exponentially large one.*

- We cannot store an infinite dimensional Hilbert space in any computer, so we have to truncate the above Hilbert space by removing some basis vectors.
- This can be done by projection

**Definition 1.** *A projector is an operator  $R$  that satisfies  $R^2 = R$ . When this is true, note  $(1 - R)^2 = (1 - R)$  also.*

- We can approximate the error in our Hamiltonian eigenstates  $PHP|\tilde{E}_j\rangle = \tilde{E}_j|\tilde{E}_j\rangle$  by considering the effect of re-adding the missing terms in the Hamiltonian as a perturbation  $\lambda V = PH(1 - P) + (1 - P)HP$ .
- Assume that we have removed all basis vectors above some  $K$ , and let  $|E_k\rangle$  be the eigenstates on the projected-out system (with Hamiltonian  $(1 - R)H(1 - R)$ ), then

$$|E_0\rangle - |\tilde{E}_0\rangle = \lambda \sum_{k>K} \frac{\langle \tilde{E}_k | V | \tilde{E}_0 \rangle}{\tilde{E}_k - \tilde{E}_0} + O(\lambda^2). \quad (1.7)$$

- We see the error in the representation (commonly called basis-set error) scales inversely with the energy of the states that we are removing. Typically, as long as this is done carefully it is ok, but as always, be careful.

### 1.1. First and second quantization

*In a first-quantized representation, one stores the position of individual particles on separate quantum registers.*

- The above representation of a  $\eta$ -particle state  $|\Psi\rangle$  was in first-quantized notation — each quantum register stored data about individual particles.
- Basis states of an  $\eta$ -particle system take the form

$$|\phi_1\rangle|s_1\rangle|\phi_2\rangle|s_2\rangle \dots |\phi_\eta\rangle|s_\eta\rangle, \quad (1.8)$$

where here,  $\phi_i$  is a label of our basis — i.e. it might convey some information about the system, but we don't require it to. We could say have  $\phi_i = (\lambda_x^{(i)}, \lambda_y^{(i)}, \lambda_z^{(i)})$ , or it could just be an integer.

**Definition 2.** *Remember that individual states  $|\phi_i\rangle$  correspond to single-particle wavefunctions  $\phi_i(\vec{r})$ . These are known in the literature as ‘spin-orbitals’, ‘basis vectors’, ‘single-particle wavefunctions’, or ‘orbitals’. For simplicity, we will use the term ‘orbital’.*

- If the above system has  $N_b$  possible orbitals for each particle (including both  $\phi$  and  $s$  variables), the total number of states is  $N_b^\eta$  — this requires  $\eta \log_2(N_b)$  qubits to store.
- Hamiltonians of the above system tend to take the form

$$H = \sum_{\phi_1, s_1, \dots, \phi_\eta, s_\eta} \sum_{\phi'_1, s'_1, \dots, \phi'_\eta, s'_\eta} H_{\phi_1, s_1, \dots, \phi'_1, s'_1, \dots} |\phi_1\rangle |s_1\rangle \dots |\phi_\eta\rangle |s_\eta\rangle \langle s'_\eta| \langle \phi'_\eta| \dots \langle s'_1| \langle \phi'_1|. \quad (1.9)$$

- This representation is a bit cumbersome, but luckily it is sparse — interactions between only  $k$  particles will only change at most  $k$  indices, so only  $O\left(\binom{\eta}{k} N_b^k\right)$  of the  $H$  coefficients will be non-zero.
- In physical systems,  $\eta, N_b$  grow with the size of the system, but  $k$  remains constant; polynomial scaling of the representation of the Hamiltonian (this is an example of something critical to check in any given problem).
- Hamiltonian simulation may be performed e.g. by LCU methods or qubitization.
- Oracular cost here is highly non-trivial — this is probably not available for the near-term.
- Need to consider particle-exchange symmetry (to be discussed later) —  $a_{\phi_1, \phi_2} = \pm a_{\phi_2, \phi_1}$ .

*In the second-quantized representation, one stores the occupation of a given single-particle wavefunction. This necessitates that particles are identical.*

- It makes just as much sense to ask ‘which particles lie in this orbital’ as opposed to ‘which orbital does this particle lie in’.
- If particles are identical, we can further reduce the question by asking ‘how many particles lie in this orbital’.
- Basis states take the form

$$|n_1\rangle|n_2\rangle|n_3\rangle \dots |n_{N_b}\rangle, \quad (1.10)$$

where  $n_i$  is the number of particles in the  $i$ th orbital.

- Need to truncate  $n_i$  to a maximum number of particles per orbital,  $\chi \leq \eta$ .
- Particle symmetry/antisymmetry automatically implicit (shows up in commutation relations of operators) — note that antisymmetry fixes  $\chi = 2$ .
- One advantage of this rephrasing — no longer need to conserve particle number.
- Number of states  $\chi^{N_b}$  — need  $N_b \log_2(\chi)$  qubits ( $= N_b$  for fermionic systems).
- Terms in the Hamiltonian are now no longer  $k$ -body (i.e. involving up to  $k$  particles), but  $2k$ -local (i.e. involving up to  $2k$  orbitals). The factor of 2 here comes as each term in the Hamiltonian removes a particle from at most one orbital and places it in at most one orbital.
- This implies that the total number of terms in the Hamiltonian scales as  $O(N_b^{2k})$ .
- If we need  $\chi = \eta$  (e.g. for bosonic systems), at large  $N_b$  and low  $\eta$  the first-quantized representation is far more compressed than the second — but number of terms in the Hamiltonian are still large.

*Spin systems may be considered either in the first or second quantized representation*

- If the position of all particles is fixed, then states in the first-quantized representation reduce to

$$|s_1\rangle|s_2\rangle \dots |s_\eta\rangle, \quad (1.11)$$

where  $s_i$  is the internal (i.e. spin) degree of freedom.

- This representation is equivalent to the above, but with  $N_b \rightarrow \eta$ , and the maximum number of particles  $\chi$  set equal to the number of internal states  $N_S$  defined before.
- Physically, in the second representation we are considering a spin flip to be an ‘excitation’, rather than considering the spin as a property of a fixed particle.
- In this situation, if  $\chi = N_S = 2$  the problem maps identically to the dynamics of an interacting set of qubits, making representing the system on  $\eta$  qubits trivial — we can use our old favourite Pauli matrices  $\{I, X, Y, Z\}^{\otimes N}$ .

## 1.2. Ground and excited state energies

*Ground and excited state energies are a critical property to calculate for many quantum systems.*

- One of the most important quantities to obtain from a quantum system is the low-energy spectra; the ground and low-level excited state energies.
- Energy differences  $\Delta E$  are immediately correlated to spectroscopy peaks.
- Reaction rates scale by the Arrhenius law:  $k = Ae^{-E_a/RT}$ , where  $E_a$  is the reaction energy barrier — the difference between the ground state energy and the barrier as a molecule rearranges.
- Note that for molecular problems atomic nuclei are typically fixed and treated classically (so this requires obtaining energies of two different Hamiltonians).
- Similarly, the difference between the energy of individual atoms and a molecular ground state energy will tell us whether or not the molecule is stable.

- Quantum phase transitions correspond to gap closings as parameters in the system are tuned.
- Thermodynamic properties of a system depend on the number of states (density of states) near to 0 energy.
- Construction of thermal states is possible via the quantum Metropolis algorithm — this is an active area of research and not covered in this course.

*Energies may be estimated via quantum phase estimation or direct state tomography of a prepared state  $|\Psi\rangle$*

- Recall from earlier that state tomography requires  $O(\epsilon^{-2})$  measurements to estimate the expectation value  $\langle H \rangle$ .
- A useful mathematical object to store physically-relevant tomographic information is the (particle) reduced density matrix, or RDM.

**Definition 3.** *The  $k$ -local qubit RDM, or qubit  $k$ -RDM, is the collection*

$$D_{i_1, \dots, i_k}^{A^{(1)}, \dots, A^{(k)}} = \langle \Psi | A_{i_1}^{(1)} \dots A_{i_k}^{(k)} | \Psi \rangle. \quad (1.12)$$

*of expectation values of all  $k$ -local qubit operators.*

*For an  $N$ -qubit system, this may be measured in  $\log(N)e^{O(k)}$  or even constant time (in  $N$  — the  $e^{O(k)}$  factor remains), depending on what extra resources are needed.*

- Note that this is a reduction over the number of particles, not the number of sites.
- By comparison, QPE can obtain energies with an error scaling as  $O(\epsilon^{-1})$ .
- QPE has the additional advantage of returning eigenstates of the Hamiltonian (up to simulation error); state tomography only tells the energy of the state you have.
- However, QPE requires far longer coherence times than state tomography.
- QPE further does not return the entire RDM (which may be of further use itself).



### 1.3. Beyond eigenvalues

*Energy derivatives correspond to various physical properties, and are crucial in mapping parameter spaces.*

- Many properties of interest may be written as derivatives of the energy as a function of parameters of the Hamiltonian

$$\frac{\partial^{n_1+\dots+n_{N_i}} E}{\partial \lambda_1^{n_1} \dots \partial \lambda_{n_{N_i}}^{N_i}}. \quad (1.13)$$

- For example, the  $i$ th component of the electric dipole of a system may be calculated as  $\frac{\partial E}{F_i}$ , where  $F_i$  is the  $i$ th component of an applied electric field.
- First derivatives may be easily calculated by the Hellman-Feynman theorem, for eigenstates  $|E_j\rangle$  of the system

$$\frac{\partial}{\partial \lambda} \langle E_j | H | E_j \rangle = \left( \frac{\partial \langle E_j |}{\partial \lambda} \right) H | E_j \rangle + \langle E_j | \frac{\partial H}{\partial \lambda} | E_j \rangle + \langle E_j | H \frac{\partial | E_j \rangle}{\partial \lambda} \quad (1.14)$$

$$= \langle E_j | \frac{\partial H}{\partial \lambda} | E_j \rangle + E \frac{\partial}{\partial \lambda} \langle E_j | E_j \rangle \quad (1.15)$$

$$= \langle E_j | \frac{\partial H}{\partial \lambda} | E_j \rangle. \quad (1.16)$$

- Terms in  $\partial H \partial \lambda$  may be determined classically, and so the quantity may be calculated via state tomography.
- Indeed, if  $H$  may be tomographed from a  $k$ -RDM,  $\frac{\partial H}{\partial \lambda}$  almost always may be also (with similar cost estimates).
- Higher-order derivatives cannot be estimated so easily, and must be approximated through perturbation theory or similar. However, these retain polynomial scaling for their estimation.

*Many properties of material systems may be written in terms of response/correlation functions.*

- Many properties of interest in physical systems involve static correlations — e.g. magnetism, which can be calculated as  $\langle \sum_i Z_i \rangle$ .
- Local static correlations may be extracted from the qubit  $k$ -RDM, making it a useful mathematical object here as well.
- Some properties of interest involve correlations in time of the form

$$\langle \Psi(t) | E | \Psi(0) \rangle = \langle \Psi(0) | e^{iHt} E | \Psi(0) \rangle, \quad (1.17)$$

which may be generated by time evolution and subsequent measurement of  $|\Psi(0)\rangle$ , conditional on an ancilla qubit, using the following circuit (these are not contained in the RDM).

- Alternatively, some properties of interest involve response measurements

$$\langle \Psi(0) | O(t) | \Psi(0) \rangle, \quad (1.18)$$

Which may be generated by time evolution and subsequent measurement of  $|\Psi(0)\rangle$  using the following circuit. Note that there is no ancilla needed, but these are similarly not contained in the RDM, unless  $|\Psi(0)\rangle$  is an eigenstate.

## 2. SPIN SYSTEMS

### 2.1. Lattices

*A common set of systems to study are lattice systems with local interactions*

- A  $d$ -dimensional lattice is series of points, or sites, in  $d$ -dimensional space with translational symmetry defined by  $d$  lattice vectors  $\vec{a}_i$ : if there exists a site at point  $\vec{p}$ , there also exists a site at point  $\vec{p} + \vec{a}_i$ .
- As an example, a square lattice with a lattice size 1 has lattice vectors  $(1, 0)$  and  $(0, 1)$ .

- It is not necessary to be able to get from every point on a lattice to every point by using the lattice vectors; just every equivalent point. E.g. a hexagonal lattice has two inequivalent points.
- We define a quantum system on a lattice by defining a quantum degree of freedom (i.e. a Hilbert space) on each site.
- As before, we may think of a lattice system in both a first or second quantized representation, and have the freedom to define the symmetry under exchange / commutation relations between different sites.
- For example, a qubit lattice is defined by attaching a copy of the Pauli operators  $I_{\vec{p}}, X_{\vec{p}}, Y_{\vec{p}}, Z_{\vec{p}}$  to each site, and combining via the tensor product.
- A Hamiltonian is typically defined locally on a lattice; using either nearest-neighbour ( $\langle\langle i, j \rangle\rangle$ ) or next-nearest-neighbour ( $\langle\langle\langle i, j \rangle\rangle\rangle$ ) coupling terms (or similar).
- Such Hamiltonians are geometrically local, which is advantageous for simulating on a quantum computer.

## 2.2. Symmetries and order parameters

*Following the theory of Landau, different phases of matter may be identified by the emergence of a symmetry, or conserved quantity.*

- A (unitary) symmetry of a system is an operator  $\hat{S}$  that commutes with the Hamiltonian  $\hat{H}$ .
- This implies that if  $|E_j\rangle$  is an eigenstate of  $\hat{H}$ , so is  $\hat{S}|E_j\rangle$ , and  $\hat{S}|E_j\rangle$  has the same energy as  $|E_j\rangle$ .
- This in turn means that either  $\hat{S}|E_j\rangle = s|E_j\rangle$ , or  $E_j$  is degenerate.
- In the first case,  $\langle E_j|\hat{S}|E_j\rangle = s \neq 0$ , but in the second case,  $\langle E_j|\hat{S}|E_j\rangle = 0$  may be achieved.

- The ‘may’ here is important; we are usually concerned with the response of the expectation value as a perturbation breaking the symmetry is added (i.e. the susceptibility). This requires measuring first derivatives  $\frac{\partial E}{\partial F}$  (and second derivatives  $\frac{\partial^2 E}{\partial F^2}$ ) for some external field  $F$ . As  $F \rightarrow 0$  (recall the last lecture).
- As symmetries themselves are static correlations they may be measured themselves by direct tomography (recall the last lecture).
- As we can study finite systems only, need to be careful to study a large enough system (and worry about boundary conditions).

### 2.3. Ising model

*By adding a transverse magnetic field, we make the Ising model quantum*

- We have already encountered the Hamiltonian for the Ising model in previous lectures:

$$H = - \sum_{\langle i,j \rangle} J_{i,j} Z_i Z_j - \sum_i h_i Z_i. \quad (2.1)$$

- With arbitrary choice of  $J_{i,j}$  this problem is QMA-hard (maps to 3-SAT); we do not expect a quantum computer to provide a speedup in finding the ground state.
- With  $J_{i,j} = J$  and  $h_i = h$ , in three-dimensions at finite temperature this model is still not yet solved, but numerics are accurate to below  $10^{-6}$  (and system does not have a sign problem).
- Ground state at  $J_{i,j} = J$ ,  $h_i = h$  is either  $|\vec{0}\rangle$  or  $|\vec{1}\rangle$  (depending on sign of  $h$ ); magnetic ordering!
- With  $J_{i,j} = -J$ , system is either anti-ferromagnetic or frustrated, depending on the lattice used.
- For example, a square lattice is anti-ferromagnetic with symmetry  $\sum_{i \in A} Z_i - \sum_{i \in B} Z_i$ , but a triangular lattice is a spin liquid.

- Note that as the total spin is still a good quantum number, in both cases the ground states must be degenerate.
- To make the model quantum, we can add a transverse field instead of an in-line field:

$$H = -J \sum_{\langle i,j \rangle} Z_i Z_j - h \sum_i X_i. \quad (2.2)$$

- Although the  $Z$  magnetism is no longer conserved,  $\prod_i X_i$  is!
- When  $h \gg J$ , the ground state is a non-degenerate eigenstate of  $\prod_i X_i$ .
- When  $J \gg h$ , the ground state is a degenerate non-eigenstate of  $\prod_i X_i$ .

#### 2.4. Heisenberg model

*The Heisenberg model is a quantum model for magnetism*

- The Heisenberg model is known as the ‘quantum Ising model’, as we measure the correlation of the spin in all three directions

$$H = - \sum_{\langle i,j \rangle} (J_x X_i X_j + J_y Y_i Y_j + J_z Z_i Z_j) - h \sum_i Z_i. \quad (2.3)$$

- Like the Ising model, the Heisenberg model has an anti-ferromagnetic and a ferromagnetic phase. It has two symmetries -  $\prod_i Z_i$  is always a symmetry, while the magnetism  $\sum_i Z_i$  is a symmetry when  $J_x = J_y$ .
- However, all excitations in the Ising model require finite energy, while the minimum excitation energy of the ferromagnetic Heisenberg model decreases with system size (spin wave).
- Exact solutions for the Heisenberg model are not known in more than 1 dimension.
- Depending on the values of the couplings, and on the lattice, this model has a rich phase diagram which is still a very active area of study.

## 3. FERMIONIC SYSTEMS

## 3.1. Fermionic operators

*Fermions are antisymmetric particles, so local operators need different commutation relations*

- A fermionic wavefunction on  $\eta$  (identical) particles must satisfy the equation

$$\Psi(\vec{R}_1, \dots, \vec{R}_i, \dots, \vec{R}_j, \dots, \vec{R}_\eta) = -\Psi(\vec{R}_1, \dots, \vec{R}_j, \dots, \vec{R}_i, \dots, \vec{R}_\eta). \quad (3.1)$$

- Given a set of  $\eta$  orthogonal single-particle wavefunctions  $\psi_i(\vec{R})$ , a Slater determinant

$$\Psi(\vec{R}_1, \dots, \vec{R}_\eta) = \frac{1}{\sqrt{\eta!}} \text{Det} \begin{pmatrix} \psi_1(\vec{R}_1) & \psi_1(\vec{R}_2) & \dots & \psi_1(\vec{R}_\eta) \\ \psi_2(\vec{R}_1) & \psi_2(\vec{R}_2) & \dots & \psi_2(\vec{R}_\eta) \\ \vdots & \vdots & & \vdots \\ \psi_\eta(\vec{R}_1) & \psi_\eta(\vec{R}_2) & \dots & \psi_\eta(\vec{R}_\eta) \end{pmatrix}, \quad (3.2)$$

has the correct antisymmetry relations required.

- Note here that the index  $i$  contains both the spatial details of the orbital and the spin of the electron — these are often known as ‘spin-orbitals’.
- If we are restricted to just  $\eta$  particles in  $\eta$  orbitals, the above is the only allowed wavefunction, as two fermionic particles cannot occupy the same spin-orbital.
- If we have  $\eta$  particles in  $N > \eta$  orbitals  $\{\psi_1, \dots, \psi_N\}$ , then for each unique combination of  $\eta$  orbitals  $\{\psi_{i_1}, \dots, \psi_{i_\eta}\}$  we may define a Slater determinant  $|\Psi_{i_1, \dots, i_\eta}\rangle$ .
- (Note that following the above,  $|\Psi_{i_1, \dots, i_p, \dots, i_q, \dots, i_\eta}\rangle = -|\Psi_{i_1, \dots, i_q, \dots, i_p, \dots, i_\eta}\rangle$ .)
- This gives a total of  $\binom{N}{\eta}$  basis states, which form an orthonormal basis on the entire allowed anti-symmetric Hilbert space, as long as the initial spin-orbitals are similarly orthonormal.

- The single Slater determinant with the lowest energy is known as the Hartree-Fock solution, and may be calculated classically relatively quickly; this is both used as a common starting state for a quantum computer and a (relatively easy) target to beat for many quantum and classical methods.
- We may map between Slater determinants via creation and annihilation operators in the second-quantized picture:  $\hat{c}_j$  removes the index  $j$  from a Slater determinant  $|\Psi_{i_1, \dots, i_\eta}\rangle$  (and sends the state to 0 if it does not contain orbital  $j$ ).
- (Note that having a term in a Hamiltonian that ‘sends an orbital to 0’ is not an issue physically — i.e. for normalization — as we have to exponentiate  $H$  to get a unitary. If  $H|\Psi\rangle = 0$ ,  $e^{iHt}|\Psi\rangle = |\Psi\rangle$ .)
- However, to preserve the anti-symmetry, we need the action of  $\hat{c}_j$  to account for the sign of the removed operator:

$$\hat{c}_j |\Psi_{i_1, \dots, i_\eta}\rangle = \sum_{p=1}^{\eta} \delta_{i_p, j} (-1)^p |\Psi_{i_1, \dots, i_{p-1}, i_{p+1}, \dots, i_\eta}\rangle. \quad (3.3)$$

- One can check that this implies the  $\hat{c}_i$  operators must satisfy the anti-commutation relations

$$\{\hat{c}_i, \hat{c}_j\} = 0, \quad \{\hat{c}_i^\dagger, \hat{c}_j^\dagger\} = 0, \quad \{\hat{c}_i, \hat{c}_j^\dagger\} = \delta_{i, j}, \quad (3.4)$$

where  $\{A, B\} = AB + BA$ .

- Each operator  $\hat{c}_i^\dagger$  has a corresponding number operator  $\hat{n}_i = \hat{c}_i^\dagger \hat{c}_i$ , which counts the number of particles in orbital  $i$ .
- Typically (at low energies), systems conserve total particle number  $\sum_i \hat{n}_i$ ; this is ensured by making sure that every term in a Hamiltonian contains an equal number of creation and annihilation operators.
- The above set of operators allow us to define the fermionic  $k$ -body RDM:

$$D_{j_1, \dots, j_k}^{i_1, \dots, i_k} = \langle \Psi | \hat{c}_{i_1}^\dagger \dots \hat{c}_{i_k}^\dagger \hat{c}_{j_1} \dots \hat{c}_{j_k} | \Psi \rangle \quad (3.5)$$

*The fermionic equivalent of the qubit operator is the Majorana operator*

- The creation and annihilation operators  $\hat{c}_j^\dagger$  and  $\hat{c}_j$  provide great physical intuition. However, they are not so nice for quantum computing — they are not unitary, nor Hermitian, and indeed, they are nilpotent (square to 0)!
- A ‘nicer’ set of operators are the Majorana operators

$$\gamma_{2j} = \hat{c}_j + \hat{c}_j^\dagger, \quad \gamma_{2j+1} = i(\hat{c}_j - \hat{c}_j^\dagger). \quad (3.6)$$

- One can check that these operators are Hermitian, unitary, and traceless, implying that their eigenvalues are either  $+1$  or  $-1$ .
- This is similar to the Pauli operators, however distant Majoranas anti-commute rather than commuting:  $\{\gamma_i, \gamma_j\} = 2\delta_{i,j}$ .
- Note that this implies a product of  $k$  Majorana operators is not necessarily Hermitian; we require that  $k = 0, 1 \pmod{4}$ .
- Also note that symmetries such as number conservation in this representation become more difficult to observe.

### 3.2. The electronic structure problem

*Particles in molecules and material systems obey the Coloumb Hamiltonian*

- Recall the Coulomb Hamiltonian from a few lectures previous in the first-quantized representation

$$H = \sum_i \frac{-\hbar^2}{2m_i} \nabla_i^2 + \sum_i V(\vec{r}_i) + \sum_{i,j} \frac{Z_i Z_j}{4\pi\epsilon_0 |\vec{r}_i - \vec{r}_j|}. \quad (3.7)$$

- Here,  $\vec{r}_i$  are the positions of both electrons and atomic nuclei (which may be considered point particles).



- Atomic nuclei tend to be quite large, and electrons quite small, so we can treat the atomic particles classically; i.e. consider their positions as fixed numbers, and study the problem of the electrons around them.
- Let us rewrite the electronic positions using capital  $\vec{R}_i$  to emphasize the difference:

$$H = H_{\text{nuc}}(\vec{r}_i) + \sum_i \frac{-\hbar^2}{2m_e} \nabla_i^2 + \sum_{i,j} \frac{eZ_j}{4\pi\epsilon_0|\vec{R}_i - \vec{r}_j|} + \sum_{i,j} \frac{e^2}{4\pi\epsilon_0|\vec{R}_i - \vec{R}_j|} = H_{\text{nuc}} + T + U + V \quad (3.8)$$

- This is known as the electronic structure Hamiltonian.

*In second quantization form, the Coulomb Hamiltonian becomes a sum of one-body (two-operator) and two-body (four-operator) terms.*

- To encode the above Hamiltonian on a quantum computer, we should start by choosing a basis set of single-particle wavefunctions  $\psi_i$ , and combine them to form Slater determinants as before.
- In the new representation, the Hamiltonian takes the form

$$H = \sum_{i,j} t_{i,j} \hat{c}_i^\dagger \hat{c}_j + \sum_{i,j,k,l} V_{i,j,k,l} \hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_k \hat{c}_l. \quad (3.9)$$

- Here, the  $t_{i,j}$  and  $V_{i,j,k,l}$  are given by the integrals

$$t_{i,j} = \int d^3\vec{R} \psi_i^*(\vec{R})(T + U)\psi_j(\vec{R}), \quad V_{i,j,k,l} = \int d^3\vec{R}_1 d^3\vec{R}_2 \psi_i^*(\vec{R}_1)\psi_j^*(\vec{R}_2)V\psi_k(\vec{R})\psi_l(\vec{R}). \quad (3.10)$$

- These integrals may be calculated to high accuracy classically once we have chosen the  $\psi_i$ .
- Note that the expectation value of the Hamiltonian

$$\langle \Psi | H | \Psi \rangle = \sum_{i,j} t_{i,j} \langle \Psi | \hat{c}_i^\dagger \hat{c}_j | \Psi \rangle + \sum_{i,j,k,l} V_{i,j,k,l} \langle \Psi | \hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_k \hat{c}_l | \Psi \rangle, \quad (3.11)$$

may be calculated entirely from the fermionic 2-RDM of a given state.

- Note also that when  $V_{i,j,k,l} = 0$  the ground state of the Hamiltonian may be found in polynomial time on a classical computer by diagonalizing the matrix  $t_{i,j}$ .

### 3.3. The Hubbard model

*The Hubbard model is possibly the most famous example of a strongly-correlated fermionic lattice model*

- The terms in the electronic structure problem have a physical interpretation.  $\hat{c}_i^\dagger \hat{c}_j$  involves an electron ‘hopping’ from orbital  $j$  to  $i$ , while  $\hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_k \hat{c}_l$  terms are ‘scattering’ processes — electrons from orbitals  $k$  and  $l$  bounce off each other and into orbitals  $i$  and  $j$ .
- If  $i = k$  and  $j = l$  (or  $i = l$  and  $j = k$ ), this is known as ‘repulsion’; electrons do not scatter off each other as they do not move, but an electron in orbital  $i$  incurs a penalty due to the presence of an electron in orbital  $j$ .
- If orbitals are separated in space then these terms will dominate —  $\psi_i$  and  $\psi_k$  need some spatial overlap in order for a particle to scatter from  $k$  to  $i$ .
- Moreover, the size of  $V_{ijij}$  decreases quadratically with the separation between  $\psi_i$  and  $\psi_j$ .
- This implies that the dominant interaction are the terms  $V_{ijij}$  where  $i$  and  $j$  correspond to the same spatial function, but with different spin.
- Taking these terms and the hopping terms leads to the Hubbard Hamiltonian — with spin explicitly specified:

$$H = \sum_{i,j,\sigma} t_{i,j} \hat{c}_{i\sigma}^\dagger \hat{c}_{j\sigma} + \sum_i U_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} \quad (3.12)$$

- (Here,  $\uparrow = +1$  and  $\downarrow = -1$  are chosen to make the labels prettier — this is very common in the literature.)

- The number of terms in the Hubbard model Hamiltonian is reduced to  $O(N^2)$ , making simulation easier.
- The Hubbard model is additionally often studied on a lattice, in which case the Hamiltonian is reduced to  $O(N)$ .
- Despite its relative simplicity, the Hubbard model is both interesting and challenging to solve; in two-dimensions or more no good solution is known, but it is thought to be a good model for high-temperature superconductivity.

### 3.4. Mapping fermions to qubits

*Fermion-to-qubit transforms must preserve the commutation relations of all relevant operators.*

- On a single orbital/qubit, we have no particle exchange statistics, and one can see that  $\gamma_0 \sim X$ ,  $\gamma_1 \sim Y$ .
- To map a fermionic system onto a qubit system we can either map the basis states or the operators (one will follow from the other).
- When mapping operators, we need to make sure that we preserve the algebra between them; it suffices to make sure we preserve the eigenspectrum and the commutation relations between different operators.
- This implies that a mapping is simply an association from each Majorana operator  $\gamma_i$  to a Pauli operator in  $\mathbb{P}^N$  such that the anti-commutation relations are conserved.

*The Jordan-Wigner transformation tracks anti-commutation relations by attaching a ‘Jordan-Wigner’ string to each operator*

- The Jordan-Wigner transformation is defined by the mapping

$$\gamma_{2j} \rightarrow \otimes_{k<j} Z_k X_j, \quad \gamma_{2j+1} \rightarrow \otimes_{k<j} Z_k Y_j. \quad (3.13)$$

- For those of you more used to creation and annihilation operators, this corresponds to a mapping

$$\hat{c}_j \rightarrow \otimes_{k<j} Z_k (X_j + iY_j), \quad \hat{c}_j^\dagger \rightarrow \otimes_{k<j} Z_k (X_j - iY_j) \quad (3.14)$$

- On the states, the Slater determinant  $|\Psi_{i_1, i_2, \dots, i_\eta}\rangle$  is mapped to  $|\vec{i}\rangle$ .
- The ‘Jordan-Wigner string’  $\prod_{k<j} Z_k$  can be thought of as keeping track of the parity of adding/removing a fermion to site  $j$  (or of keeping track of the commutation relations).
- This string extends the locality of terms in most Hamiltonians to  $O(N)$  (minimizing the additional cost of Hamiltonian simulation and tomography from this has been a research topic for a lot of the last few years).

*The Bravyi-Kitaev transformation and JKMN transformation compress the JW string into a log-depth encoding, but at the cost of requiring non-locality*

- Let us consider how to minimize the locality of Majoranas following a transform onto qubits. One way to do this is to start by assigning a nominal qubit to each orbital, mapping  $\gamma_{2j} \rightarrow X_j P_{j,0}$ ,  $\gamma_{2j+1} \rightarrow Y_j P_{j,1}$ , and choosing the Pauli strings  $P_{j,0}$  and  $P_{j,1}$  so that everything anti-commutes.
- Non-local Pauli strings always commute, so there must be some overlap. One way to enforce this overlap is by drawing a tree, and connecting everything up to the origin (say by adding a set of  $X_i$  operators to each  $P_{j,0}$  and  $P_{j,1}$ ).
- Now all  $\gamma_{2j+1}$  operators anti-commute with Majoranas from all of their children. If we add  $Z_k$  to all  $P_{j,0}$  whenever  $k$  is a child of  $j$  then all  $\gamma_{2j}$  operators also anti-commute with all Majoranas from their children.
- Finally, for each  $j$ , as we go up the string to the root node, let us add a  $Z_k$  to all children of every node that we pass that are to the right of our current node. This

guarantees that we commute with everything to the right (and the left nodes will similarly anti-commute with us)! Problem solved.

- We are free to set up our tree as we see fit for this problem; it turns out that the optimal construction to minimize the weight of the resulting Majoranas is known as a Fenwick tree, although others have been studied.
- The resulting operator weight is  $\lceil \log_2(N) + 1 \rceil$ .
- Note that under this transformation the occupation of an orbital is mapped non-locally across its children —  $i\gamma_{2j}\gamma_{2j+1} = Z_j \otimes_{k \text{ child of } j} Z_k$ .
- Another tree-based method for constructing a set of non-commuting Pauli operators is the JKMN transform, which relies on constructing a ternary tree (where each node has 3 children).
- In this method, each node with children corresponds to a qubit  $j$ , and each edge below this node to the operators  $X_j, Y_j, Z_j$ .
- Nodes without children correspond to Majoranas; each Majorana is assigned the tensor product of Pauli operators along the path leading to the root node.
- This method achieves a weight  $\log_3(N)$ , which is provably optimal; however, occupancies are mapped to highly non-local objects.

#### 4. STATE PREPARATION FOR STRONGLY CORRELATED SYSTEMS

- We have previously discussed what properties to measure of quantum states that are interesting in strongly-correlated physics and chemistry problems, and how to map those problems onto a physical system.
- However, actually preparing/ obtaining a representation of the states on a quantum computer is a critical challenge for quantum computing.
- Critical point: preparation of ground states of arbitrary Hamiltonians is QMA-hard. Even arbitrary 2-local Hamiltonians — even even geometrically-local 2-local

Hamiltonians. We don't know about the type of Hamiltonians which I've described earlier in the text. So, we *expect* each of these methods to fail at some point.

#### 4.1. Quantum phase estimation

*As previously mentioned, QPE has the ability to prepare eigenstates*

- Performing QPE with a full ancilla register projects a starting state  $\sum_j a_j |E_j\rangle$  into an eigenstate  $|E_j\rangle$  with probability  $|a_j|^2$ .
- (In many cases the full projection is not necessary, as terms such as  $\langle E_j | V | E_k \rangle$  may be obtained in post-processing.)
- If the largest time evolution is for  $e^{iHT}$ , separate a window of approximately  $\frac{1}{T}$  states - need to ensure either this is sufficiently small or  $a_j$  is chosen appropriately in starting state for required purpose.
- Can 'choose'  $a_j$  via e.g. symmetry or particle number constraints (if respected by  $H$ ).
- Critical failure - if we can't find a starting state with *some* overlap with the ground state, QPE won't help us!

#### 4.2. Adiabatic state preparation

*By deforming a system's Hamiltonian from a Hamiltonian with a known ground state to a target, one evolves the known ground state to the ground state of the target — if the evolution is slow enough.*

- Let us consider evolving a system by the time-dependent Hamiltonian  $H(t) = (T-t)H_0 + tH_1$ , as  $t$  goes from 0 to  $T$ . Let  $|E_j(t)\rangle$  be the instantaneous eigenstate of  $H(t)$  at time  $t$ .
- The ground state  $|E_0(0)\rangle$  is not necessarily an eigenstate of the Hamiltonian  $H(t)$  for  $t \neq 0$ , so it will evolve in time; let us define  $|\tilde{E}_0(t)\rangle$  as the evolution after time

*t.* Formally

$$|\tilde{E}_j(t)\rangle = \mathcal{T} e^{i \int_0^t dt' H(t')} |E_j(0)\rangle, \quad (4.1)$$

where  $\mathcal{T}$  is the time-ordering operator.

- The adiabatic theorem states (roughly) that  $|\langle E_j(t) | \tilde{E}_j(t) \rangle|^2 \sim 1 - \frac{\delta E}{T}$ , where  $\delta E$  is the minimum gap between  $|E_j(t)\rangle$  and  $|E_{j\pm 1}(t)\rangle$  as  $t : 0 \rightarrow T$ . (This is only true in the small  $\delta E/T$  limit.)
- If  $H_0$  is a Hamiltonian for which we know the ground state, and  $H_1$  is the target Hamiltonian, if we simulate slow enough time evolution we can prepare approximate states with reasonable overlap with the ground state.
- Cost estimates for realistic systems scale as  $O(N^{5.5} \Delta E^{-3})$  (with some assumptions); very costly. Provable upper bounds scale as  $O(N^{12+o(1)} \Delta E^{-2})$ ; even worse (ArXiv:1605.03590).
- Critical failure - orthogonality catastrophe, gap size.

### 4.3. The unitary coupled cluster ansatz

*VQE recap*

- Recall that a variational algorithm/variational quantum eigensolver consists of
  - (1) A parameterized quantum circuit  $U(\vec{\theta})$ , or ‘variational ansatz’; a circuit with input to some gates left as classical free parameters  $\vec{\theta}$ .
  - (2) A starting state  $|\vec{0}\rangle$  on which to act the quantum circuit.
  - (3) A classical cost function  $E(\theta)$ . In our case  $E(\theta)$  is the expectation value of a target Hamiltonian  $H$ :

$$E(\vec{\theta}) = \langle \vec{0} | U^\dagger(\vec{\theta}) H U(\vec{\theta}) | \vec{0} \rangle \quad (4.2)$$

- (4) A classical optimization algorithm to minimize the cost function  $E(\theta)$  in terms of the parameters  $\theta$ .

(5) A protocol to estimate  $E(\vec{\theta})$  (and optionally derivatives  $\nabla_{\theta}E(\vec{\theta})$ ) from the quantum hardware at each required point  $\theta$ .

- VQEs are attractive for the near term as they can be made as low-depth as possible + adapted to any given hardware.
- Unlike QPE or other methods, VQEs do not typically promise to outperform classical algorithms / solve BQP-complete problems (or even problems not in P!).

*The cluster operator generates excitations from the Hartree-Fock state to low-lying non-interacting higher energy states.*

- As discussed previously, we typically expect the ground state of an interacting problem to be a linear combination of low-energy excitations of the corresponding non-interacting problem.
- In the electronic structure problem, this corresponds to excitations of a few particles from the Hartree-Fock state  $|\Psi_{\text{HF}}\rangle$ .
- These excitations may be captured by the cluster operator

$$T = T^{(1)} + T^{(2)} + T^{(3)} + \dots \quad (4.3)$$

$$T^{(1)} = \sum_{i \text{ empty}, a \text{ filled}} \theta_a^i \hat{c}_i^\dagger \hat{c}_a \quad (4.4)$$

$$T^{(2)} = \sum_{i,j \text{ empty}, a,b \text{ filled}} \theta_{a,b}^{i,j} \hat{c}_i^\dagger \hat{c}_j^\dagger \hat{c}_a \hat{c}_b. \quad (4.5)$$

- Here,  $\theta_a^i$  and  $\theta_{a,b}^{i,j}$  are the free parameters of the problem.
- $e^T|\Psi_{\text{HF}}\rangle$  may be evaluated classically as the series terminates quickly (we may only excite each electron once). This is a powerful classical chemistry method known as coupled cluster.



- By comparison,  $e^{T-T^\dagger}$  is a unitary operator, and  $e^{T-T^\dagger}|\Psi_{\text{HF}}\rangle$  cannot be simply truncated — terms allow for the excitation and subsequent de-excitation of electrons.
- However,  $e^{T-T^\dagger}$  may be implemented (approximately) on a quantum computer — e.g. by Trotterization.
- The key issue here is truncation. In theory, using all  $T^{(n)}$  operators up to  $n = \eta$  (where  $\eta$  is the number of electrons) will allow the ansatz to capture the exact ground state (both via UCC and CC); however the gate-count for implementing  $T^{(n)}$  for a system with  $N$  spin-orbitals scales as

$$O\left(\binom{\eta}{n}\binom{N-\eta}{n}\right) \sim O(N^{2n}). \quad (4.6)$$

In practice, we expect to need aggressive truncation of the cluster operator (even as much as taking only a few operators within  $T^{(1)}$  and  $T^{(2)}$ ).

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