Unraveling correlated materials’ properties with noisy quantum computers:

Natural-orbitalized variational quantum eigensolving

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How to handle strongly correlated materials with a classical computer?

- Holy grail: High-temperature superconductors

- “Spherical cow”: the Hubbard model (see David’s talk)

\[ H = -t \sum_{i,j,\sigma} c^+_i c_j + U \sum_i n_{i\uparrow} n_{i\downarrow} \]

- Failure of mean field (Hartree Fock)!

State-of-the-art classical method: Embedding

Map lattice problem to local impurity problem:

Boil problem down to “quantum quintessence”

Similar to active space selection: pick correlated degrees of freedom
Increase $N_c$ (∼active space size) to reach convergence

Problems with increasing $N_c$:

- Exponential size of Hilbert space (if using FCI/exact diagonalization)
- (or) Monte-Carlo sign problem
- (or) too much entanglement for tensor network methods (MPS... see Alberto’s talk)

“Control”: nothing changes when $N_c \rightarrow N_c + 1$
Solving impurity models with quantum computers: first attempts
To reach larger impurity sizes: use a quantum computer!?

Early proposals: Bauer '16, Kreula '16
Embedding: Dynamical mean field theory
Impurity model solved with QC!
All the rest is classical

Key quantity: impurity Green’s function

\[ G_{ij}(t) = \langle \psi_{GS} | c_i(t) c_j^+(0) | \psi_{GS} \rangle \]

with \( |\psi_{GS}\rangle \): ground state of impurity model

Quantum algorithm:

- (classical) Truncate infinite bath (finite #qubits!)
- Prepare ground state \( |\psi_{GS}\rangle \)
- Time-evolve \( |\psi_{GS}\rangle \) via Trotterization to measure \( G(t) \)

As expected:
- With no or “few” errors, it works: exponential speedup.
  - “few”: error levels compatible with quantum error correction.

But what with today’s QCs?
NISQ computers... and their issues

Limitations:

▶ Few qubits + Short coherence times

Expected issues:

▶ Truncate infinite bath
  – Few qubits: truncation error?

▶ Prepare ground state \( |\psi_{GS}\rangle \)
  – Adiabatic preparation... too long circuit?

▶ Time-evolve \( |\psi_{GS}\rangle \) via Trotterization to measure \( G(t) \)
  – Implementation of \( e^{iHt} \): too long circuit?
A hybrid variational approach for shorter circuits: VQE

Variational Quantum Eigensolving, Peruzzo ’14

- Prepare ground state $|\psi_{GS}\rangle$
  - Adiabatic preparation... too long circuit?

Find ground state of $H$:

- An old method:
  Variational principle: $\min_\theta \langle \psi_\theta | H | \psi_\theta \rangle \geq E_0$

- A new idea:
  Use QC to compute $\langle \psi_\theta | H | \psi_\theta \rangle$

- Hybrid!

**Bonus 1**: QPU better at preparing and measuring quantum states
**Bonus 2**: Can choose ansatz circuit $U_\theta$ to accommodate QPU constraints

Caveat: no speedup guarantee!
Variational Quantum Algorithms on the Atos QLM

- QLM: powerful stack for variational algorithms
- VQE for the Heisenberg model $H = XX + YY + ZZ$:

```
H_XY = Observable(2, pauli_terms=[Term(1, term, [0, 1]) for term in ['XX', 'YY', 'ZZ']])

prog = Program()
qbits = prog.qalloc(2)
theta = [ prog.new_var(float, '{theta:.3f}') for i in range(3) ]
H(qbits[0])
RY(theta[0])(qbits[1])
CNOT(qbits)
RX(theta[1])(qbits[0])
RY(theta[2])(qbits[1])
circ = prog.to_circ()

optimizer_scipy = ScipyMinimizePlugin(method="COBYLA", x0=[0.4, -0.3, 0.6])
qpu = optimizer_scipy + LinAlg()
job = circ.to_job(job_type="OBS", observable=H_XY)
result = qpu.submit(job)
print("Minimum energy =", result.value)
```

Minimum energy = -2.999999851052666

Download+install: myqlm.github.io
from qat.core import Observable, Term
from qat.lang.AQASM import Program, RY, CNOT
from qat.qpus import get_default_qpu
from qat.plugins import ScipyMinimizePlugin

# we instantiate the Hamiltonian we want to approximate the ground state
hamiltonian = Observable(nqbits=2, pauli_terms=[Term(1, op, [0, 1])])

# we construct the variational circuit (ansatz)
prog = Program()
reg = prog.qalloc(2)
theta = [prog.new_var(float, r'\theta_%s' % i) for i in range(2)]
RY(theta[0])(reg[0])
RY(theta[1])(reg[1])
CNOT(reg[0], reg[1])
circ = prog.to_circ()

# construct a (variational) job with the variational circuit and
job = circ.to_job(observable=hamiltonian,
                  nbshots=100)

# we now build a stack that can handle variational jobs
stack = ScipyMinimizePlugin()
stack.add_job(job)
stack.run
Run on an actual QPU!

Download+install: myqlm.github.io

Connecting to a QPU/Backend

myQLM can be used to connect to a Qiskit Backend. This module is composed of three main classes:

- **BackendToQPU**: Synchronous QPU, capable of running in a Qiskit backend

```python
from qat.interop.qiskit import BackendToQPU

# Declare your IBM token
MY_IBM_TOKEN = "..."

# Wrap a Qiskit backend in a QPU
qpu = BackendToQPU(token=MY_IBM_TOKEN, ibmq_backend="ibmq_

# Submit a job to IBMQ
result = qpu.submit(job)
```
First attempts on NISQ computers: summary

<table>
<thead>
<tr>
<th>Keen ’20</th>
<th>Rungger ’19</th>
<th>Jaderberg ’20</th>
<th>Yao ’21</th>
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<tbody>
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<td>One bath site (‘two site DMFT’)</td>
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<td>One bath site (slave boson)</td>
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<tr>
<td>VQE (‘hardware efficient ansatz’)</td>
<td>VQE with excited states</td>
<td>VQE (with machine learning techniques)</td>
<td>VQE (unitary coupled cluster ansatz)</td>
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<td>Trotterization.</td>
<td>Via computation of excited states.</td>
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Number $N_c$ of impurities

Our goal: reach larger impurity models ($N_c$) with same noise
Step 1/2: pick the right embedding method

Several embedding methods on the market:

<table>
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<tr>
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<th>Dynamical Mean Field Theory</th>
<th>Rotationally-Invariant Slave Bosons (Gutzwiller)</th>
<th>Density Matrix Embedding Theory</th>
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<td><strong># bath levels</strong></td>
<td>infinite</td>
<td>$N_c$</td>
<td>$N_c$</td>
</tr>
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<td><strong>Impurity observable</strong></td>
<td>Green’s function</td>
<td>1-RDM</td>
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<td>$\langle \psi_GS</td>
<td>c_i(t) c_j^+(0)</td>
<td>\psi_GS \rangle$</td>
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<tr>
<td><strong>Final output</strong></td>
<td>Freq-dependent self-energy</td>
<td>Low-energy self-energy</td>
<td>Static self-energy</td>
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Most NISQ-compatible choice: RISB

- Well-defined bath truncation
- Simpler observable
- Access to quasiparticle renormalization factor

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TA, Lee, Kotliar '17
Step 2/2: pick the right ansatz

Goal: compute \( \langle \psi_{GS} | c_i f_j^+ | \psi_{GS} \rangle \) with VQE. How to choose the ansatz \( |\psi_{\theta} \rangle \)?

**Key requirement: “expressivity”**

- **Gold standard: Unitary coupled-cluster (UCC)**

  \[
  |\text{UCCSD}\rangle = e^{T - T^+} |HF\rangle
  \]

  with cluster operator

  \[
  T = \sum_{i \in \text{occ}, a \in \text{virt}} \theta_i^a c_a^+ c_i + \sum_{i > j \in \text{occ}, a > b \in \text{virt}} \theta_{ij}^{ab} c_a^+ c_b^+ c_i c_j
  \]

  **Issue: too many terms, too deep (long) circuit!**

- **A shallower ansatz: the Low-Depth Circuit Ansatz (LDCA)**

  Dallaire-Demers et al ’20

  **Key insight:**

  - pick mostly “Gaussian operations” (\( c_a^+ c_i \) terms) that create single-Slater determinant states
  - ... and few selected non-Gaussian (\( c_a^+ c_b^+ c_i c_j \)) terms to include correlations (\( \sim \) multi reference)

  **Issue: still too deep!**
Step 2/2: pick the right ansatz

Our ansatz: the Multi-Reference Excitation-Preserving (MREP) ansatz

Start from multi-reference state
Spread fermionic excitations

How to read such a circuit?

One line (qubit): one orbital
("Jordan-Wigner" fermion-qubit mapping)

- After X gates: state $|00001111\rangle$: single Slater det.
- After MR pattern: MR state $\alpha |00001111\rangle + \beta |11001100\rangle + \gamma |00110011\rangle + \delta |10101010\rangle$
- Action of "fSim" gate: $|01\rangle$ becomes $u|01\rangle + v|10\rangle$ (and dephase $|11\rangle$)

Cf Sugisaki ‘19: circuits for molecules with diradical character
Intermezzo: Demo
Beyond VQE with the natural orbitalization procedure
The failure of plain-vanilla VQE

- RISB embedding with MREP ansatz in VQE
  - Sanity check:
    without noise, works
  - With noise... no convergence!
    (too far from true impurity ground state)

- How to overcome this limitation?
  - Key idea: use freedom in choice of orbital basis
The role of the orbital basis

- Goal: minimize \( \langle \psi_{\tilde{\theta}} | H | \psi_{\tilde{\theta}} \rangle \).
- \( |\psi_{\tilde{\theta}} \rangle \) and \( H \): expressed in a given orbital basis. E.g:

\[
H = \sum_{p,q} h_{pq} c_p^+ c_q + \frac{1}{2} \sum_{p,q,r,s} h_{pqrs} c_p^+ c_q^+ c_r c_s
\]

where \( c_p^+ \) creates electron in orbital \( \phi_p(r) \).

- We can freely change basis:

\[
\tilde{c}_p^+ = \sum_q U_{pq} c_q^+
\]

- Equivalent \( |\psi_{\tilde{\theta}} \rangle \) and \( H \)!

- **How to exploit this freedom?**
  Although equivalent, different bases: more or less sparse representations

- E.g state \( |11 \rangle \) in \( c_0^+, c_1^+ \) basis
  ... becomes...

\[
\text{state } \frac{|00\rangle - |01\rangle + |10\rangle - |11\rangle}{2}
\]

in rotated basis:

- \( \tilde{c}_0^+ = (c_0^+ + c_1^+)/\sqrt{2} \)
- \( \tilde{c}_1^+ = (c_0^+ - c_1^+)/\sqrt{2} \)

- Different overhead to prepare on QC!
Aside: Hartree-Fock on a quantum computer

Arute et al, 2020

- Goal: rotate to “molecular-orbital” basis (orbital basis that minimizes energy of a single Slater determinant)

- Circuit:

- Accurate measured energies (not shown here)... w.r.t HF on classical computer...

- But... if rotation to MO basis on classical computer (instead of QC):
  - circuit would have been trivial!
    (a single Slater determinant: only a few X gates!)

- Orbital rotations: easy for classical computers... and potentially harmful on QCs!
  - Can we go (classically) to the “optimal” orbital basis?
Optimizing the orbital basis: Natural orbitals

▶ Natural orbitals (NOs): basis that diagonalizes the one-particle density matrix $D_{ij} = \langle \psi_{GS} | c_i^\dagger c_j | \psi_{GS} \rangle$

“Basis in which GS can be written with the least number of Slater determinants”

▶ Hence, basis in which preparation circuit should be the shortest!

▶ Comparison of original vs NO basis

NO leads to faster+more accurate convergence

On orbital optimization: see also Saad’s talk
In practice: iterative construction

- In practice:
  - the ground state $|\psi_{GS}\rangle$ is unknown
  - so the NO basis is unknown!

- Iterative procedure: “NOization”
  - Compute RDM for current approximation
  - Rotate to approximate NO basis
  - Repeat until convergence
NOization: results

- Iterative procedure converges to exact NO energy

- Noise-free case:
  - Ansatz reaches exact energy for \( U = 0 \) and \( U = 1 \)
  - Good (but not perfect) for \( U = 2 \)

- Noisy case
  - Bias is reduced by NOization
Putting everything together: self-consistent embedding loop

- Half-filled, paramagnetic phase of Hubbard model:
  Upon increasing interaction $U$, expect “Mott transition” (see David’s talk)

- Embedding: Rotationally-Invariant Slave Boson:
  - $N_c = 2$ (2 impurities, 2 bath sites: 8 qubits)
  - Solved by minimization procedure
- MREP ansatz

- **Realistic noisy simulations on Atos QLM:**
  - Depolarizing gate noise
  - Noise level to reproduce gate error rates: 0.6% (2-qubit gates), 0.16% (1-qubit gates).
Final results

Quasiparticle weight $Z = R^+ R$

$$\Sigma(\omega) = \omega (I - (R^+ R)^{-1}) + R^{-1} \lambda (R^+)^{-1}$$

Static self-energy $\lambda$

$$\bar{\lambda} = \lambda - \epsilon_{\text{loc}}$$
Conclusion

▶ Classical preprocessing matters
  – Embedding
  – NOization
  – VQE

▶ Algorithms for NISQ must be tested under noisy conditions
  – Noisy simulation on QLM

▶ NOization:
  – Here, used for impurity model... behavior for quantum chemistry problems?
  – With analog quantum simulators?
    (Variational Quantum Simulation, Kokail '19)
Thank you

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