Using quantum computers to solve quantum chemistry problems

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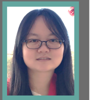
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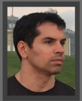
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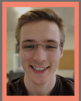
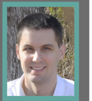

George Barron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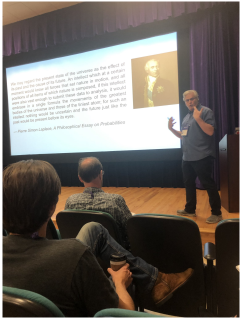
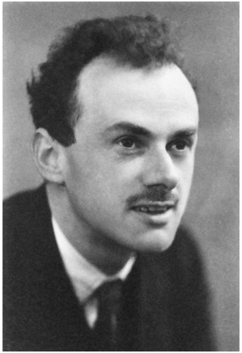


The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble.

P.A.M. Dirac
Proc. Roy. Soc. (London), 123 714 (1929).



- Consider H_2 ground state
- $|\sigma\bar{\sigma}\rangle$ is Aufbau principle – approximation
- Exact state:

$$|\psi\rangle = a\,|\sigma\bar{\sigma}\rangle + b\,|\sigma\bar{\sigma}^*\rangle + c\,|\sigma^*\bar{\sigma}\rangle + d\,|\sigma^*\bar{\sigma}^*\rangle$$

- To get $|\psi\rangle$, solve for values of a, b, c, d and store in a vector
- Number of configurations increases rapidly with system size:

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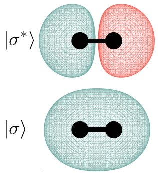

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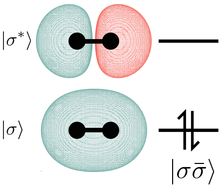

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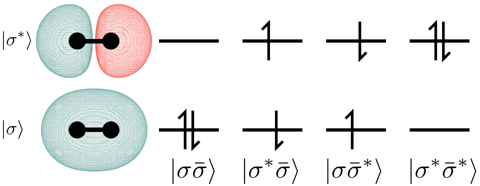

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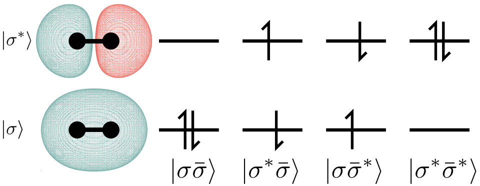

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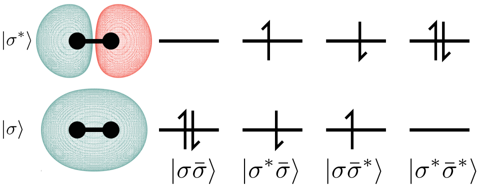

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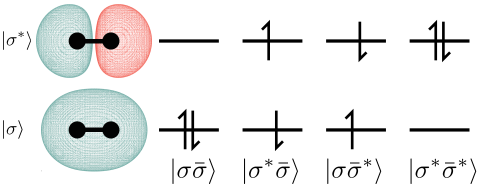

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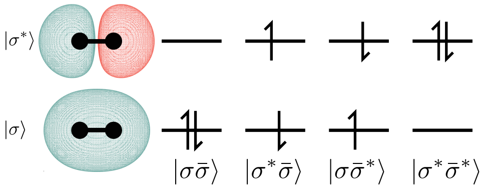

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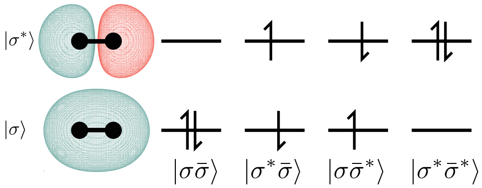

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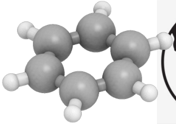

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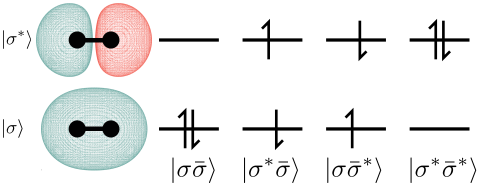

* Hard drive with mass of earth - estimated using 5Tb / Kg

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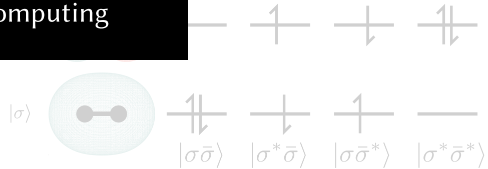

- Because of this, approximations are needed, e.g. CCSD, MP2, (and even DFT)
- However, often these approximations aren't accurate enough to solve a given problem
- This is why chemists have started thinking about quantum computing

What is a quantum computer?

Quantum computing is the use of quantum-mechanical phenomena such as **superposition** *and* **entanglement** *to perform computation.*

– Wikipedia

Quantum systems are fully characterized by a wavefunction: $|\psi\rangle$

- In general, this state can be a **superposition** of any number of states:

$$|\psi\rangle = c_1 |\phi_1\rangle + c_2 |\phi_2\rangle + \cdots$$

- Not a strange idea - think of the σ bond in H_2
- This can also illustrate **entanglement** which means “not factorizable into product form”
  - Consider occupation number basis - not separable:

$$|\sigma\rangle = \frac{1}{\sqrt{2}}\left(|10\rangle + |01\rangle\right)$$

  - Superposition but **not** entangled:

$$\left(|0\rangle + |1\rangle\right) \otimes \left(|0\rangle + |1\rangle\right) = |00\rangle + |10\rangle + |01\rangle + |11\rangle$$

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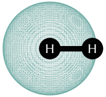
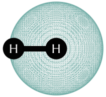


$|A\rangle$



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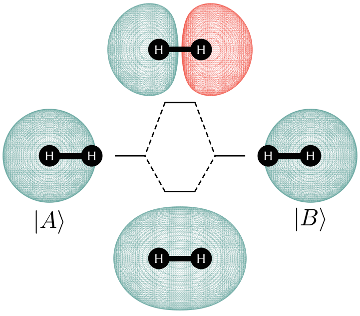

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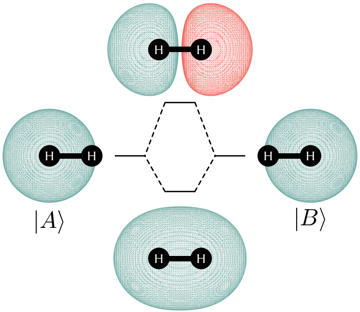

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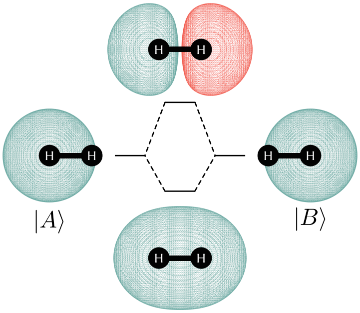

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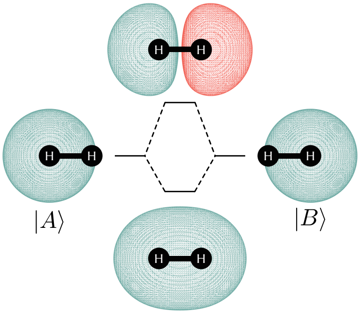

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- Quantum bit (Qubit): any superposition of 0 *and* 1
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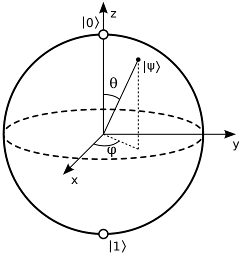

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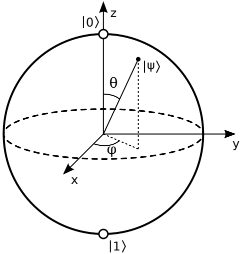

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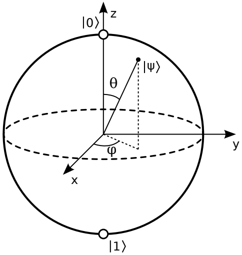

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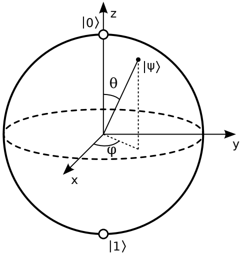

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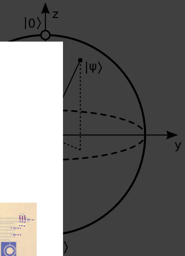

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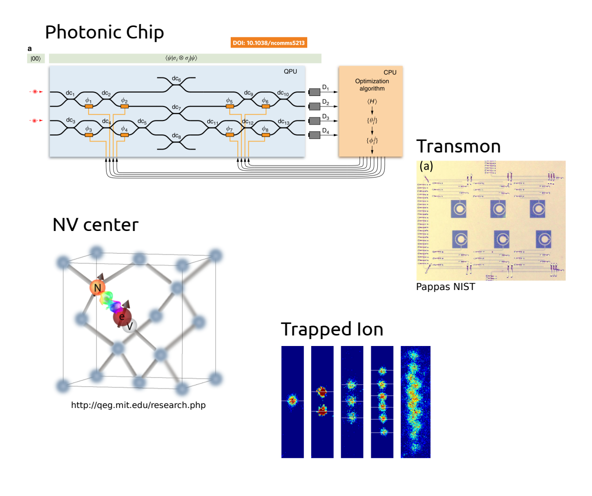
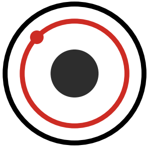


How to study chemistry with qubits?

- Think of a “trapped-ion” in it’s ground state
- Excite it with light to it’s first excited state (these two states will define the qubit)
- Now map or “associate” a single qubit to a molecular orbital for some system you wish to study
- A $|0\rangle$ qubit means the MO is unoccupied. A $|1\rangle$ qubit means the MO is occupied.

$|0\rangle$  Atom in Ground State

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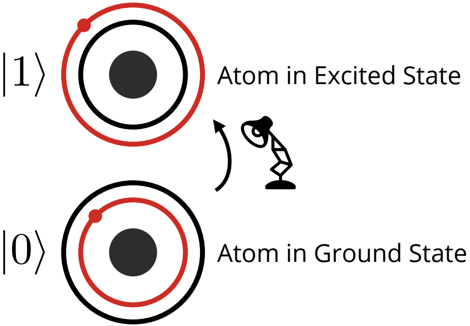
- Now map or “associate” a single qubit to a molecular orbital for some system you wish to study
- A $|0\rangle$ qubit means the MO is unoccupied. A $|1\rangle$ qubit means the MO is occupied.



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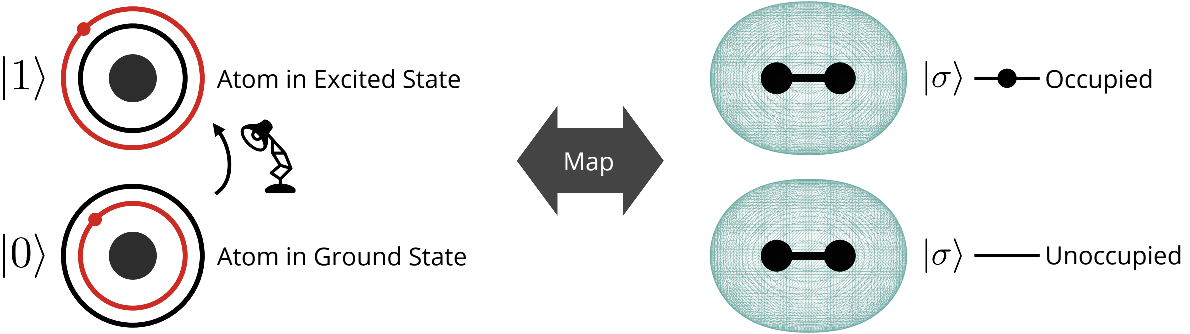

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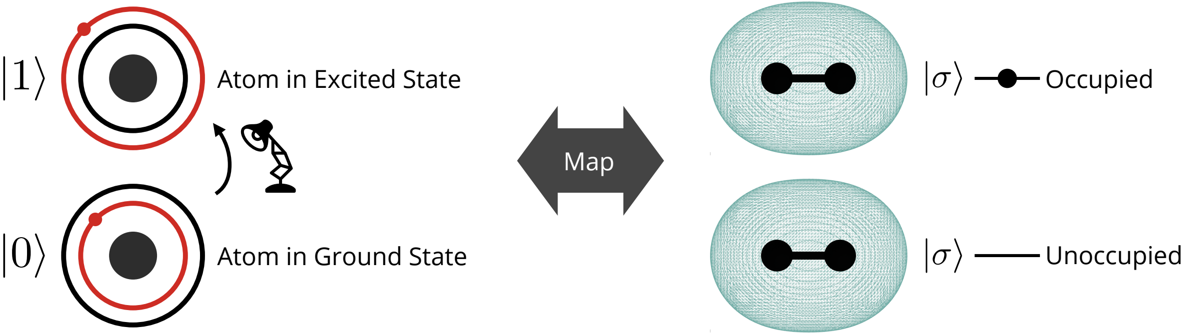
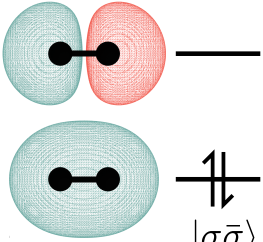


Map Aufbau configuration to QPU

$|\sigma^*\rangle$

$|\sigma\rangle$



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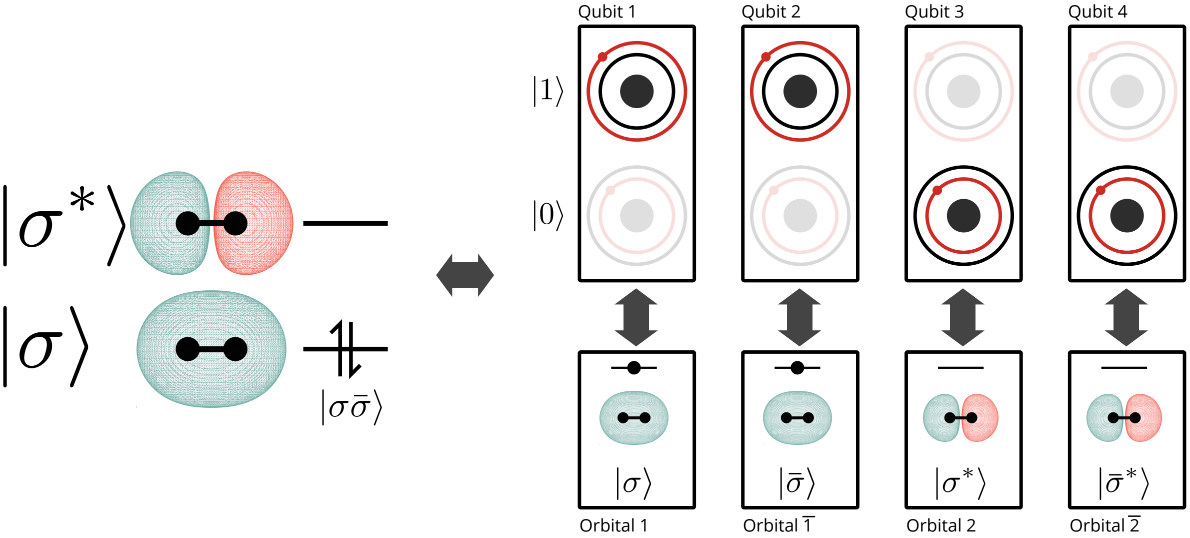
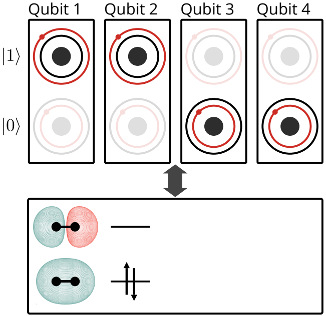
Map Aufbau configuration to QPU



1. Create with Aufbau state
2. Create superposition of $|0\rangle + |1\rangle$ on qubit 3 (Hadamard): $|1100\rangle \rightarrow \frac{1}{\sqrt{2}}(|11(1+0)0\rangle)$
3. Excite qubit 1 only when qubit 3 is excited (CNOT): $\frac{1}{\sqrt{2}}(|11(1+0)0\rangle) \rightarrow \frac{1}{\sqrt{2}}(|1100\rangle + |0110\rangle)$
4. Continue to entangle other qubits to create $|\psi\rangle = a|1100\rangle + b|1010\rangle + c|0101\rangle + d|0011\rangle$
5. Measure any operator on the QPU to get info about molecule



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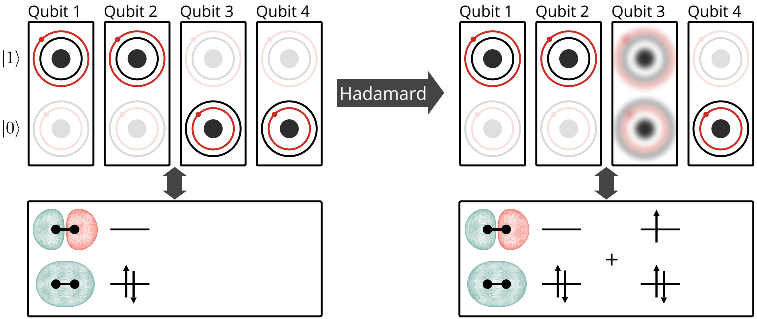

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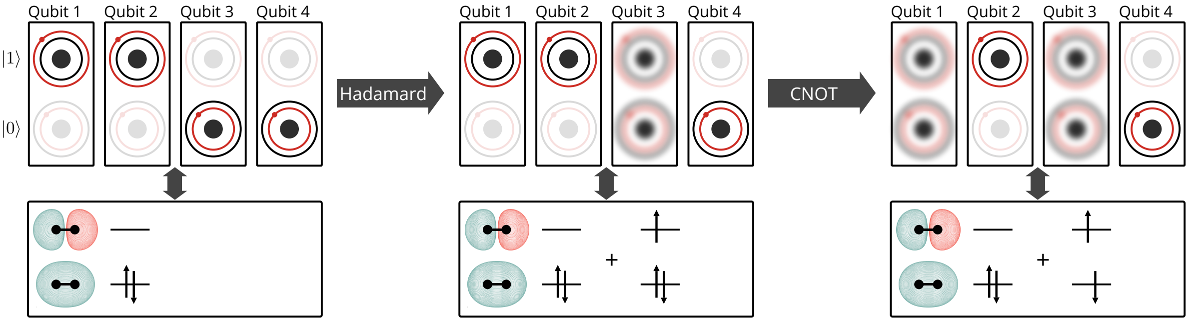

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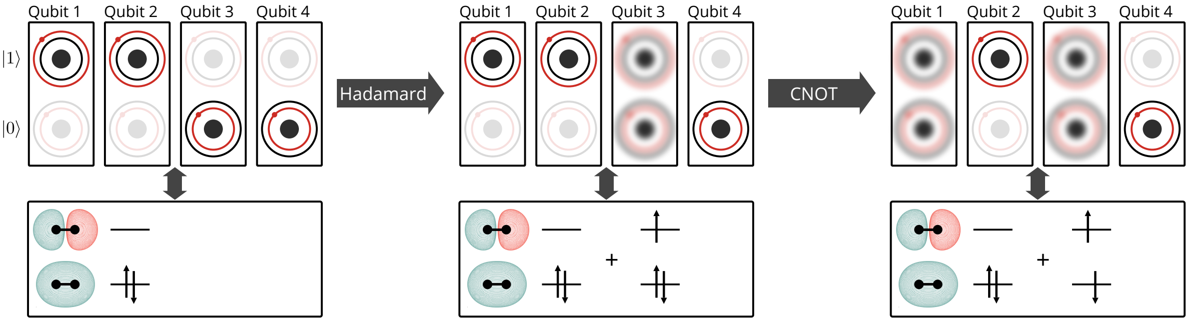

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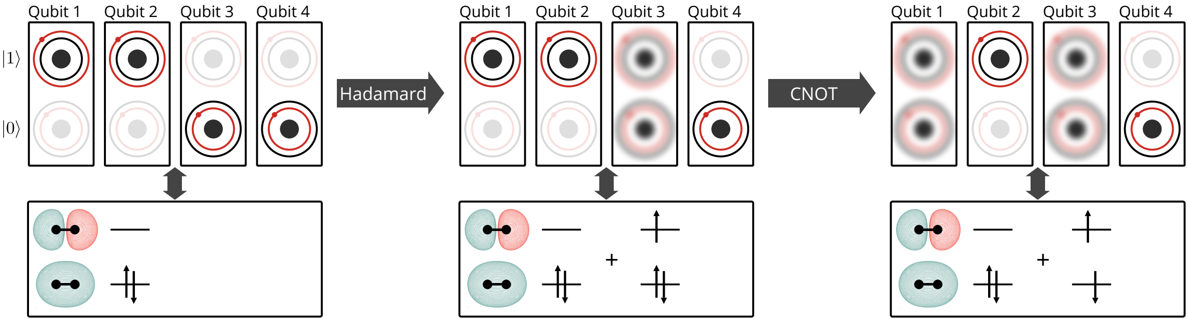

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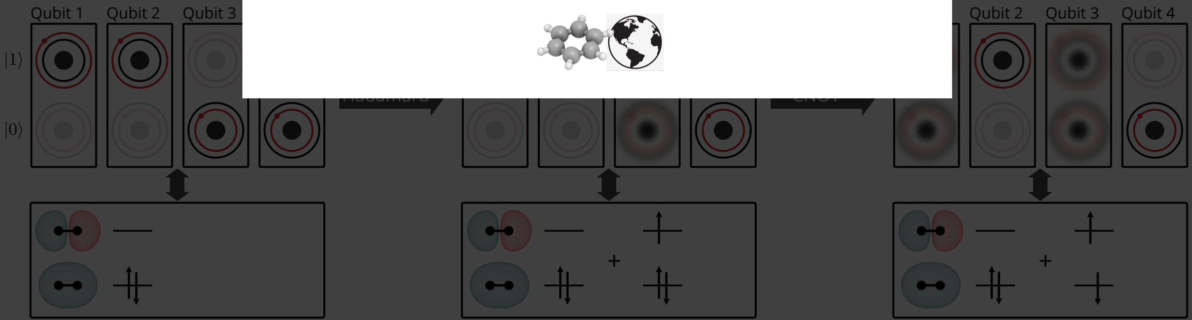
- Every spin orbital requires 1 new qubit
- 64 orbitals could be *exactly* treated with only 128 qubits



Challenges with realizing useful quantum computers

- Many errors/noise with current devices:
  - Entangled superpositions only last for a short period of time
  - Related to T_1 and T_2 in NMR
  - Operations take a fixed amount of time, limiting the types of computations that can be performed
  - Qubits are rarely perfect 2-level systems - operations can have errors

- Long-term: *error correction* – Until then NISQ devices
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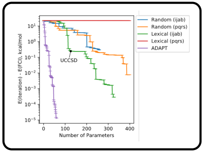
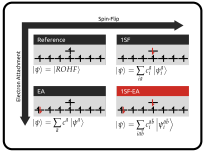
Quantum chemistry and quantum computing

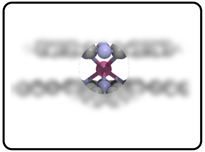
Improved Quantum
Algorithms: ADAPT-VQE



Quantum
Computing

Computational
Chemistry





New electron-
correlation methods

New WFT-DFT
embedding methods

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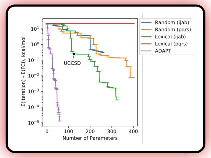

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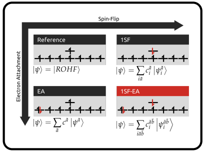

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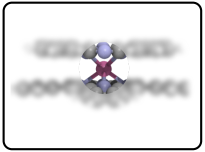

New WFT-DFT
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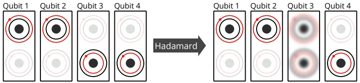
- **Problem:** Every operation adds noise - how to minimize # of operations?
- **Solution:** Don't define operations before computation - grow dynamically: ADAPT-VQE*



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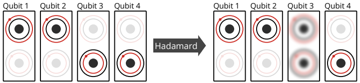

*Grimsley, Barnes, Economou, Mayhall. *Nature Communications* 10, 3007, (2019)

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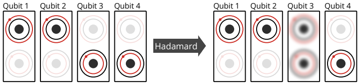

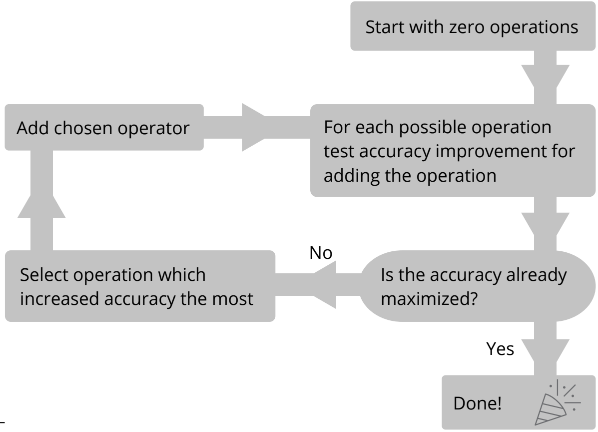

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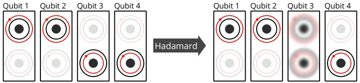

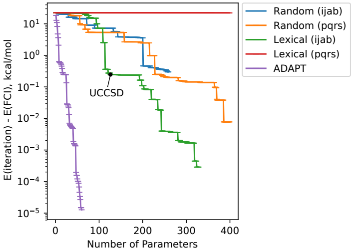

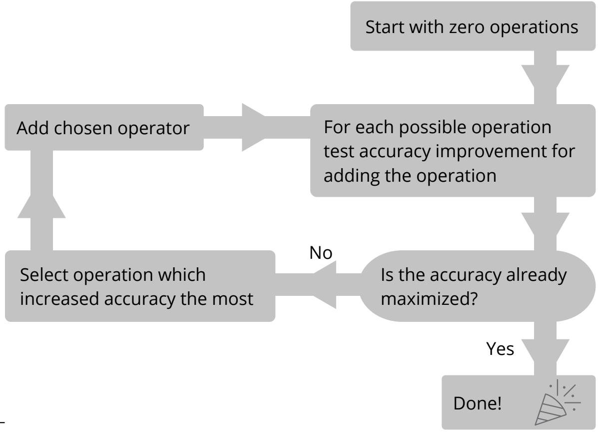

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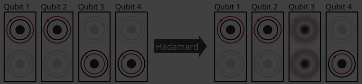
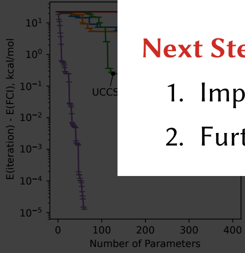
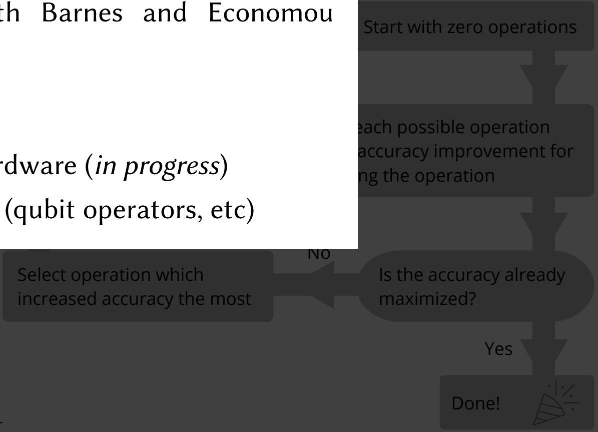


Collaborative Projects with Barnes and Economou groups in Physics Dept

Next Steps:

1. Implement on IBM hardware (*in progress*)
2. Further improvements (qubit operators, etc)





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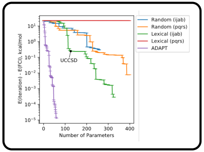

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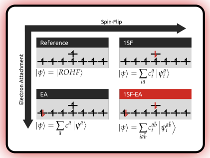

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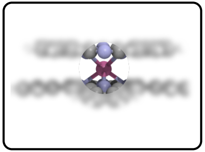

New WFT-DFT
embedding methods

TbPc2: A special Single Molecule Magnet

- SMM: as natural 2-level systems - obvious candidates for qubits - but electronic spins quickly decohere
- Nuclear spins, better isolated, longer coherence, but difficult to control/couple (magnetic fields)
- Recently,[a] Wernsdorfer demonstrated that the **nuclear** spin in SMM $TbPc_2$ could be **electronically** controlled

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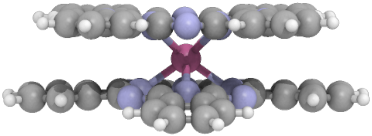
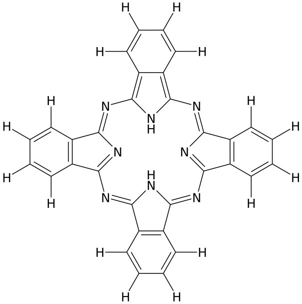


$TbPc_2$



Phthalocyanine "Pc"

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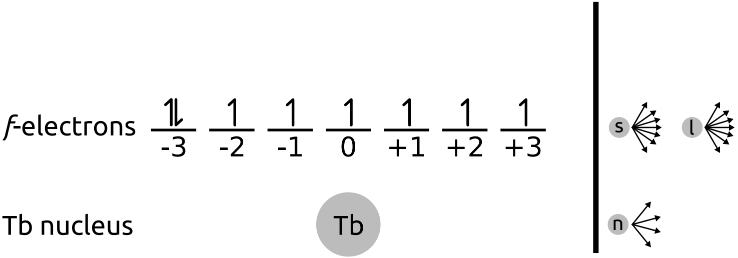

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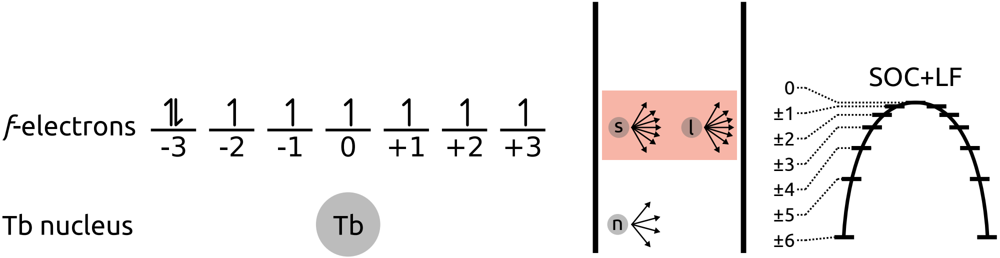

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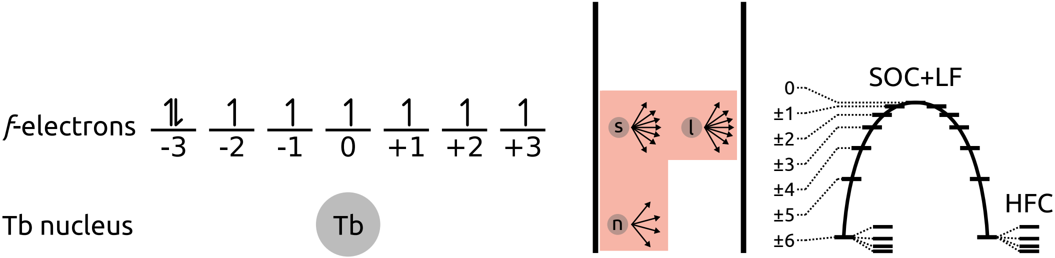

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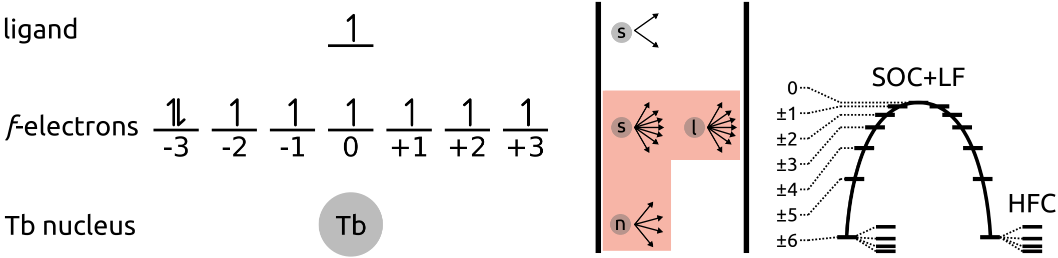

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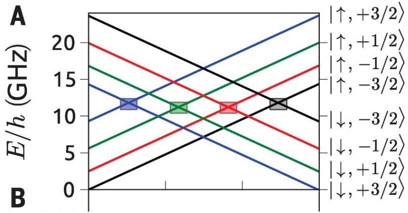

[a] Wernsdorfer and coworkers, Science, 334, 1135 (2014)
[b] Ishikiwa et al., JACS., 125, 8694 (2003)

TbPc2: A special Single Molecule Magnet

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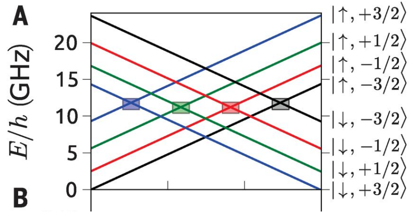

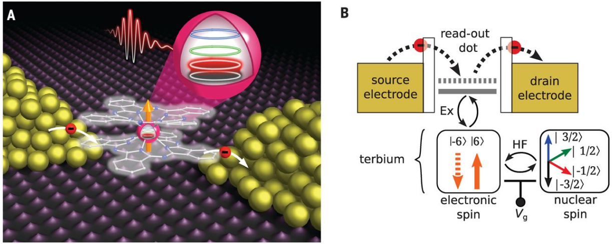

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- Our goal is to understand how the chemical properties relate to the physical properties

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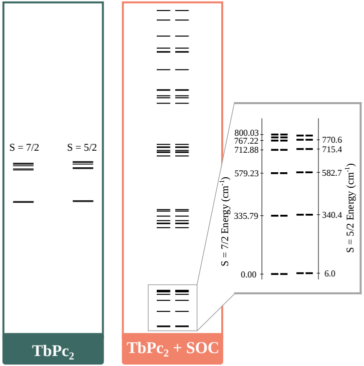
Ryan Pederson Alex Wysocki Kyunghwa Park

- RASSCF/SO-RASSI/ANO-RCC-VDZ (VDZP on Tb)
- Spin-orbit effects are **huge**



* Pederson, Wysocki, Mayhall, Park. arXiv:1905.10635 (2019)

Ab initio spectrum of $TbPc_2$

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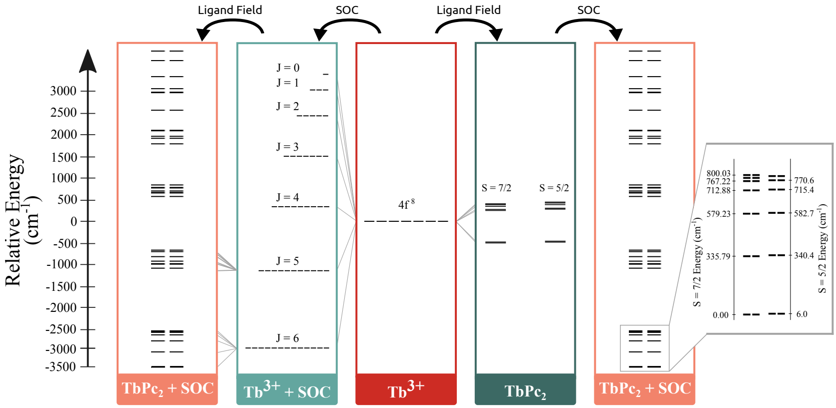

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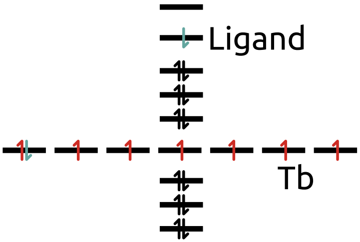
New method to simplify calculations: SF-EA

- Multireference methods (CASPT2, MRCI) expensive and difficult to use

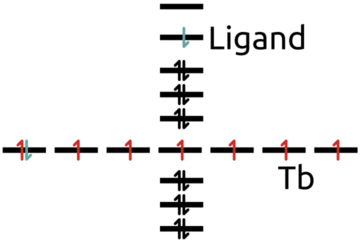


- Spin degeneracy – **Spin-flip methods** (Anna Krylov):

- Spatial degeneracy – **IP/EA methods**:

- $TbPc_2$ has both spin- and spatial- degeneracy → SF-EA or SF-IP*

New method to simplify calculations: SF-EA



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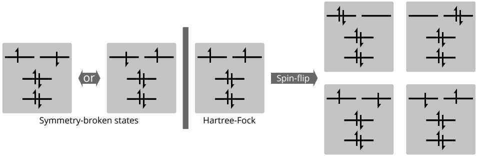

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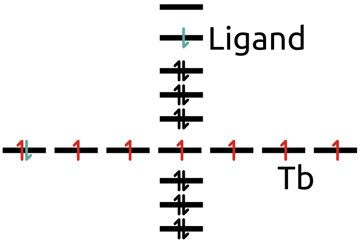

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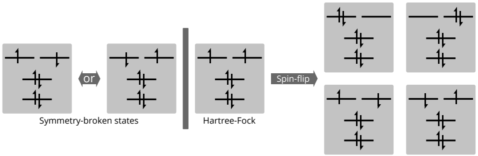

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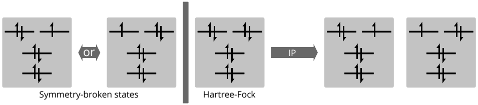

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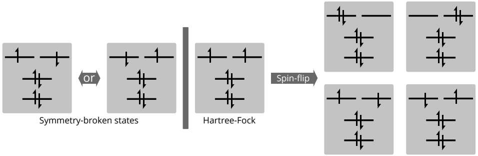

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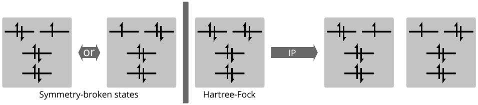

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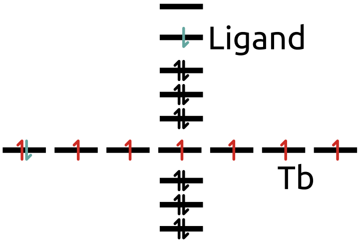

*Shannon Houck, Mayhall (2019). JCTC. 15, 2278

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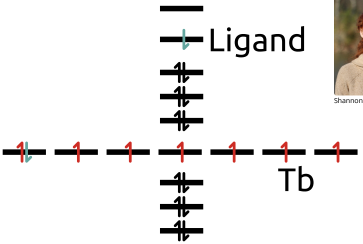

Shannon Houck

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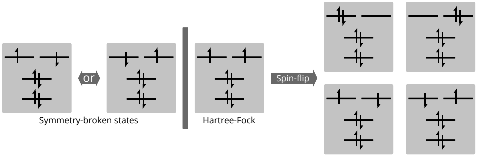

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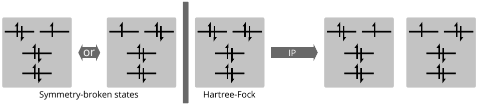

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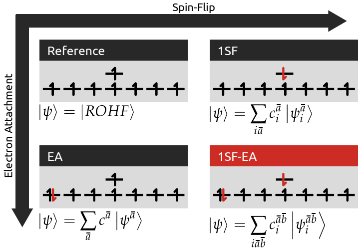

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- $[Fe_2(OH)_3(NH_3)_6]^{2+}$ is a simpler example of mixed spin/spatial degeneracy (Double Exchange)
- Oxidized/High-spin gives well-defined ground state
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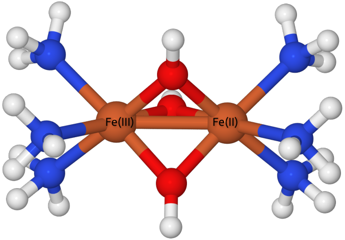
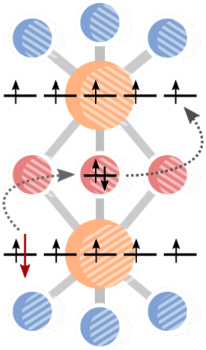
Shannon Houck





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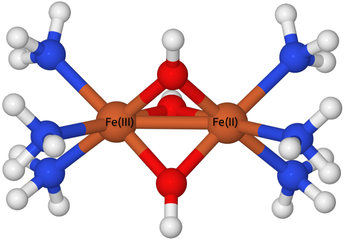

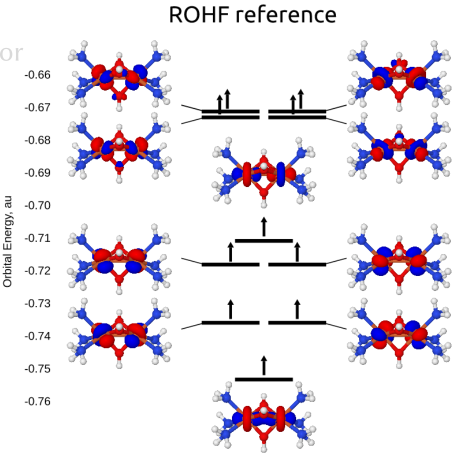

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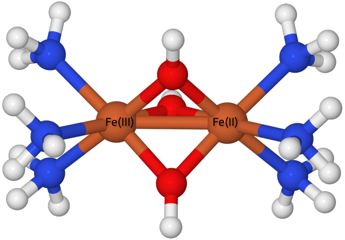

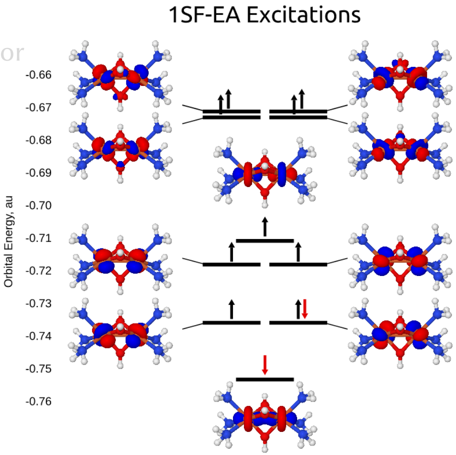

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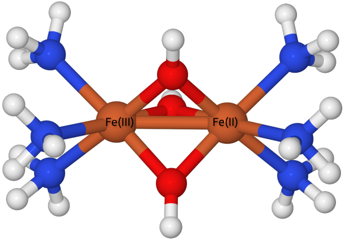

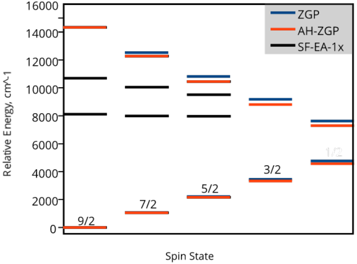

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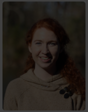
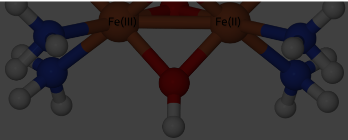
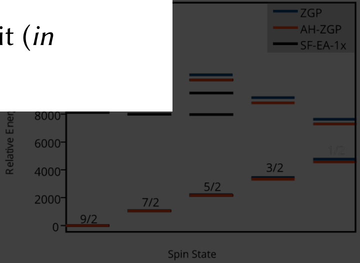


Shannon Houck

Next Steps:

1. Faster implementation (Shannon currently at QChem internship doing this!)
2. Ready for $TbPc_2$ after adding spin-orbit (*in progress*: Oinam Meitei)





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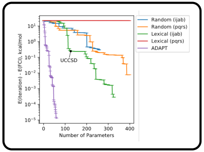
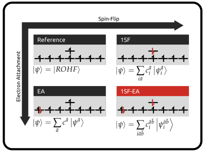
Quantum chemistry and quantum computing

Improved Quantum
Algorithms: ADAPT-VQE



Quantum
Computing

Computational
Chemistry



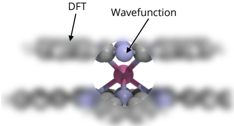
New electron-
correlation methods

New WFT-DFT
embedding methods

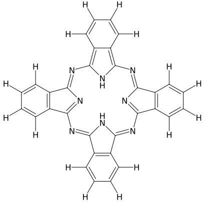
Better embedding methods

Daniel Claudino

- **Goal:** treat area directly interacting with Tb at high-level of theory, with low-level DFT for the rest
- **S**ubsystem **P**rojected **AO** **DE**composition (SPADE)*
  1. Perform full-system DFT calculation
  2. Project density onto active atoms
  3. SVD molecular orbital matrix
  4. Rotate orbitals into SVD basis
  5. Do high-level WF calculation only in embedded space
- SPADE is more robust than previous approaches
- We've recently made further improvements, reducing cost** by "concentric localization" of virtual orbitals



$TbPc_2$



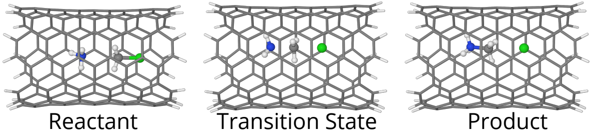
Phthalocyanine "Pc"

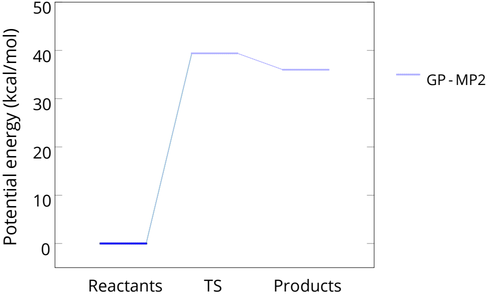
*Daniel Claudino, Mayhall (2019). JCTC. 15, 1053
**Daniel Claudino, Mayhall (2019). ChemRxiv. doi:10.26434/chemrxiv.8846108.v2

Better embedding methods

- As example, consider S_N2 inside Carbon Nano Tube (CNT)
- CNT environment has big impact on reaction
- DFT and MP2 exhibit large differences
- Embedding MP2 inside of DFT works really well

Daniel Claudino





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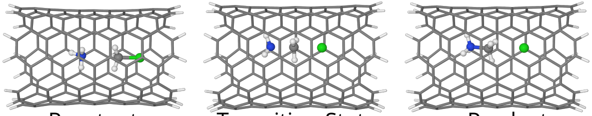

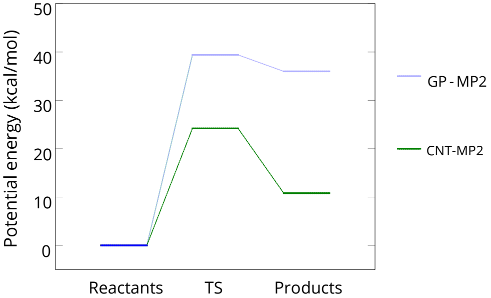

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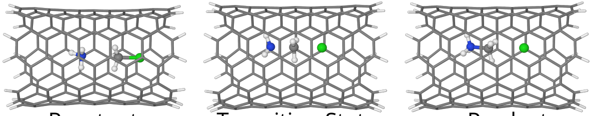
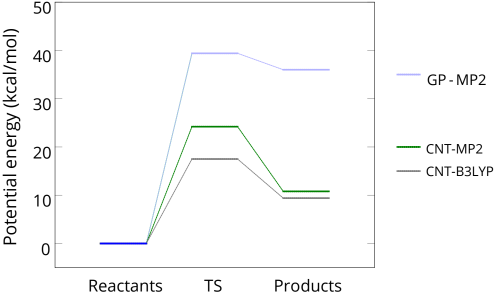


Reactant Transition State Product



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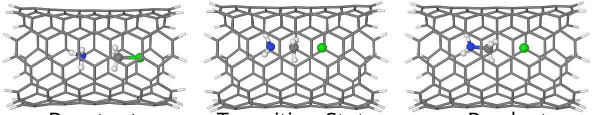

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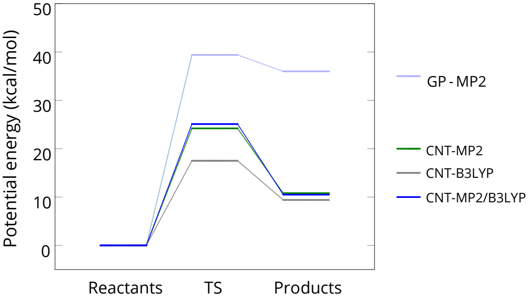

[*] Daniel Claudino, Mayhall (2019). JCTC. 15, 1053

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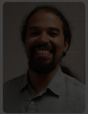
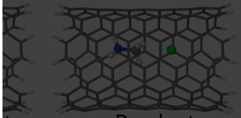
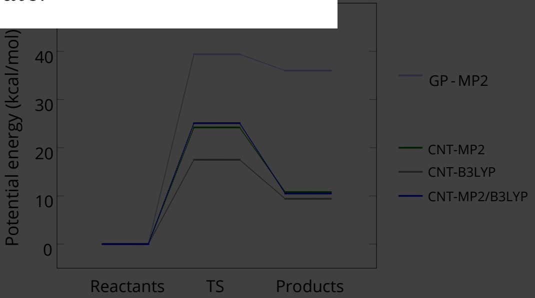




Next Steps:

1. Combine Shannon's SF-EA code with Daniel's embedding with Oinam's SOC integrals
2. Tackle $TbPc_2$ on a substrate!



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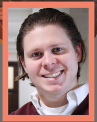
Team

Mayhall Group


Shannon Houck


Vibin Abraham


Harper Grimsley


Robert Smith


Daniel Claudino


Oinam Meitei

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NSF CAREER: 1752612
DOE: DE-SC0018326
DOE: DE-SC0019199
NSF: 1839136

VIRGINIA TECH

Theory Groups @ VT


Valerie Welborn


Daniel Crawford


Ed Valeev

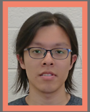

Diego Troya

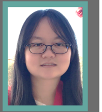

Nick Mayhall

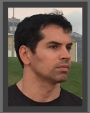
Collaborators

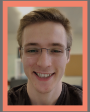
VT

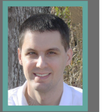

Sophia Economou


Ho Lun Tang


Linghua Zhu


Ed Barnes


George Barron


Bryan Gard


Kyungwha Park


Alex Wysocki

NIST


David Pappas


Junling Long