A data-centered approach to understanding quantum behaviors in materials

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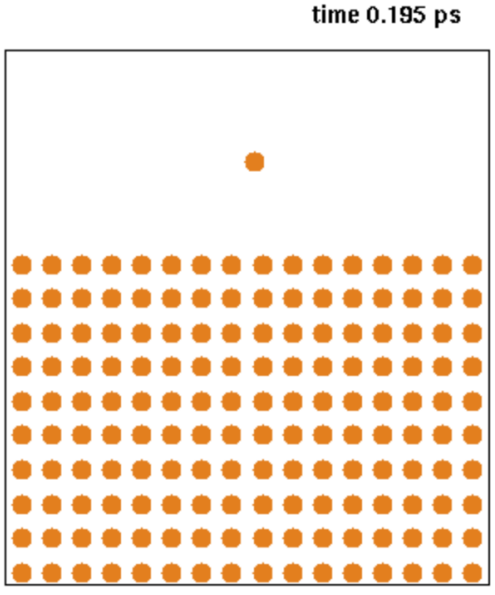
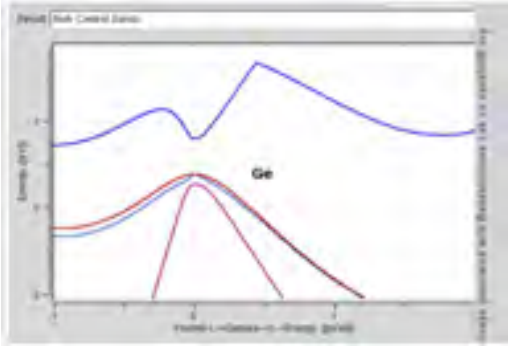
June 5, 2018

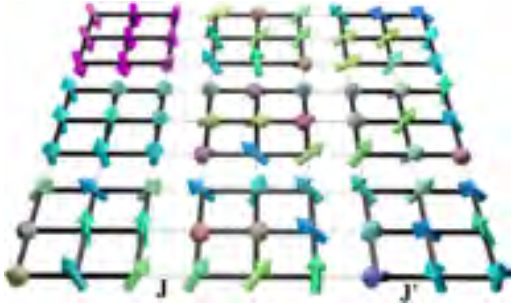
Effective models in materials physics



Interacting balls with potentials



Band structure



Interacting spins

Leblanc, Whitehead, Plumer. J. Phys. Cond. Mat. **25** 296004

The challenge

Can we systematically build **interacting quantum** models based on fine-grained simulations?

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Can we systematically build **interacting quantum** models based on fine-grained simulations?

Brian Busemeyer

Hitesh Changlani

Huihuo Zheng

Joao Nunes Rodrigues

Kiel Williams

Thanks to: Blue Waters, Simons foundation, Department of Energy EFRC Center for Emergent Superconductivity (DEAC0298CH1088) , DOE FG02-12ER46875 , NSF Grant No. DMR 1206242

Outline

Quantum mechanics of electrons in materials

Effective models as a data analysis problem

Applications to real materials

Quantum mechanics of electrons in materials

Basics of quantum mechanics

The state of a system is described by a wave function.

For a collection of particles at a given time t, $\Psi(r_1, r_2, r_3, \ldots, t)$.

$|\Psi(r_1, r_2, r_3, \ldots, t)|^2$ gives the probability of each particle being at the given position at the given time.

Expectation values:

$$\langle Q \rangle = \int \Psi^*(r_1, r_2, \ldots) \hat{Q} \Psi(r_1, r_2, \ldots)$$

The Hamiltonian

Hamiltonian $\hat{H}$ is an operator.

For nuclei and electrons, it is

$$-\frac{1}{2}\sum_i \nabla_i^2 + \sum_{i<j}\frac{1}{r_{ij}} - \sum_{\alpha,i}\frac{Z_\alpha}{r_{i\alpha}} + \sum_{\alpha<\beta}\frac{Z_\alpha Z_\beta}{r_{\alpha\beta}}$$

Special wave functions are eigenfunctions:

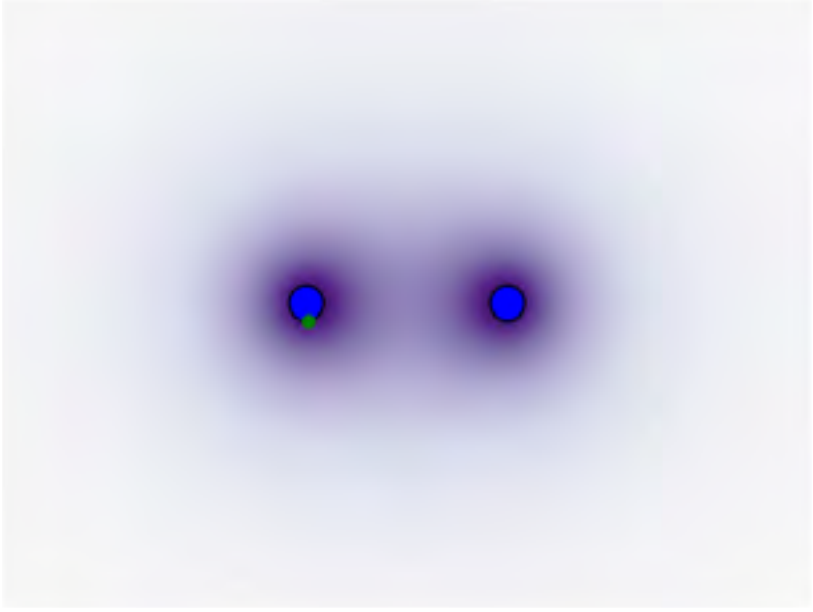
$$\hat{H}\Phi_k = E_k\Phi_k$$

If you know the eigenfunctions and their eigenenergies, then you know the dynamics of the system.

An example: H_2

An H_2 molecule: two atoms and two electrons. Electrons exist in a continuum.

$$\Psi(r_1, r_2) = \Phi(r_1)\Phi(r_2)$$



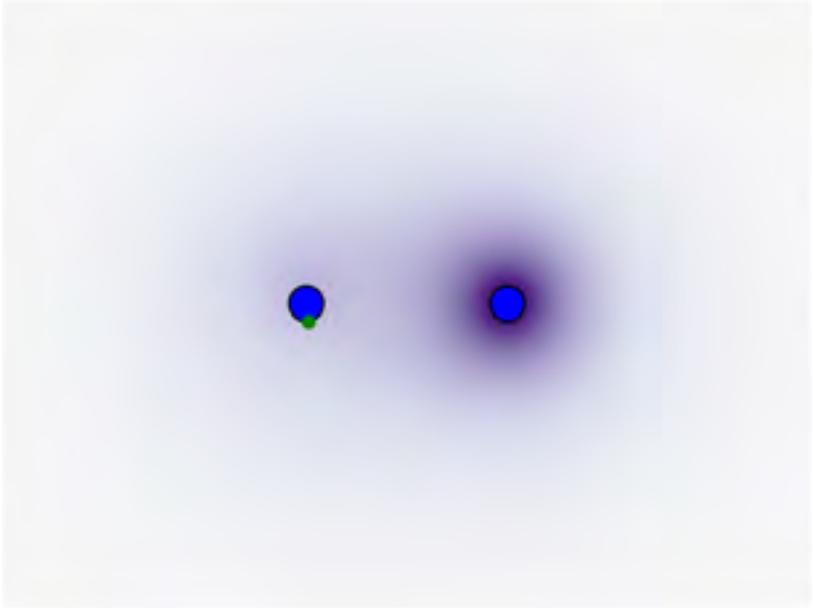
Color is wave function of electron 1 given that electron 2 is at the green dot.

No correlation here! Not an eigenfunction.

An example: H_2

Better approximation to the lowest energy eigenstate

$$\Psi(r_1, r_2) \neq \Phi(r_1)\Phi(r_2)$$



Electron 1 avoids the atom where electron 2 is nearby.

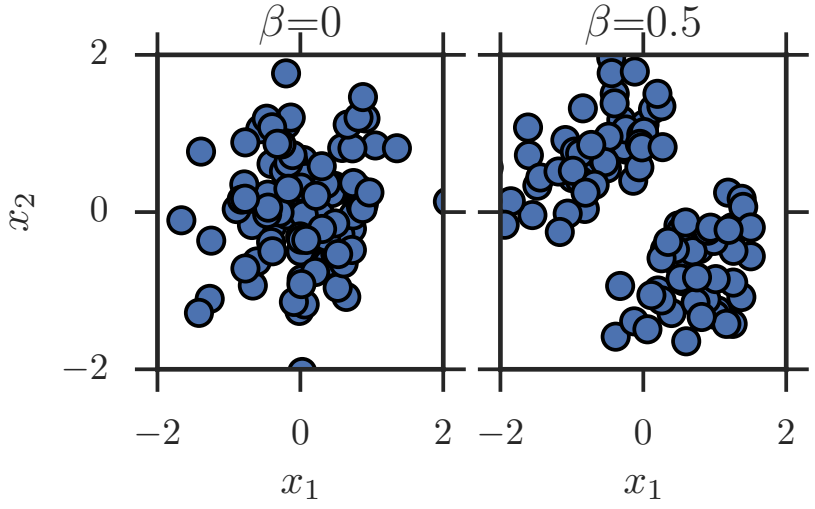
Competes with quantum "spreading out" effect.

Quantum Monte Carlo

Expectation values:

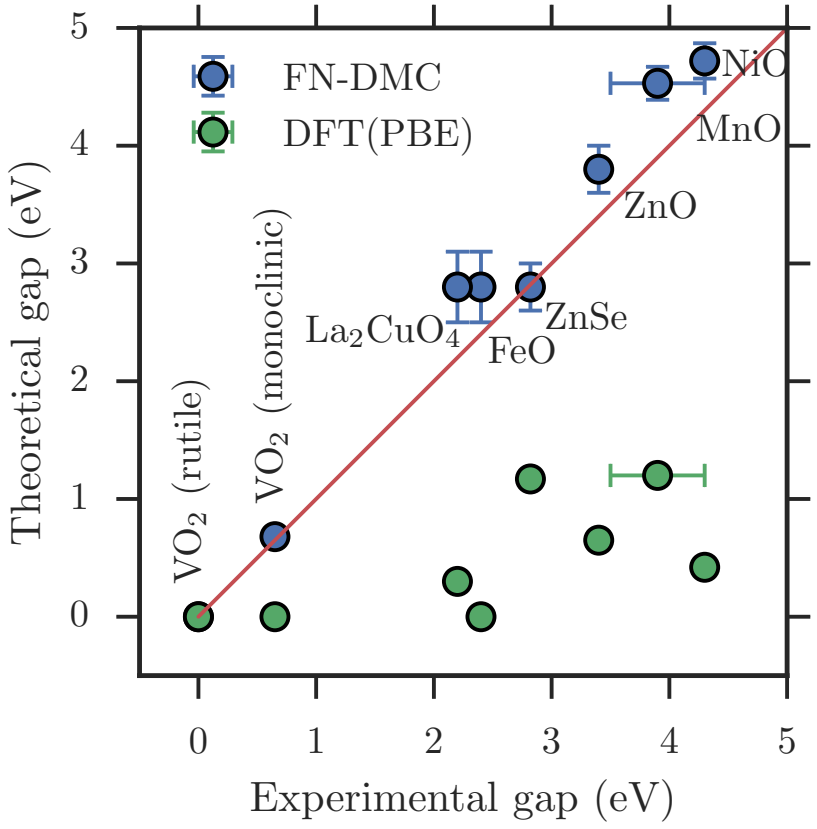
$$\langle E \rangle = \int \Psi^*(r_1, r_2, \ldots) \hat{H} \Psi(r_1, r_2, \ldots)$$

If Ψ is not factorizable..use Monte Carlo!



We technically use projection methods for higher accuracy but it doesn't affect the point here.

Quantum Monte Carlo



Can create wave functions that are very close (but not quite exactly equal) to the ground state and some excited states.

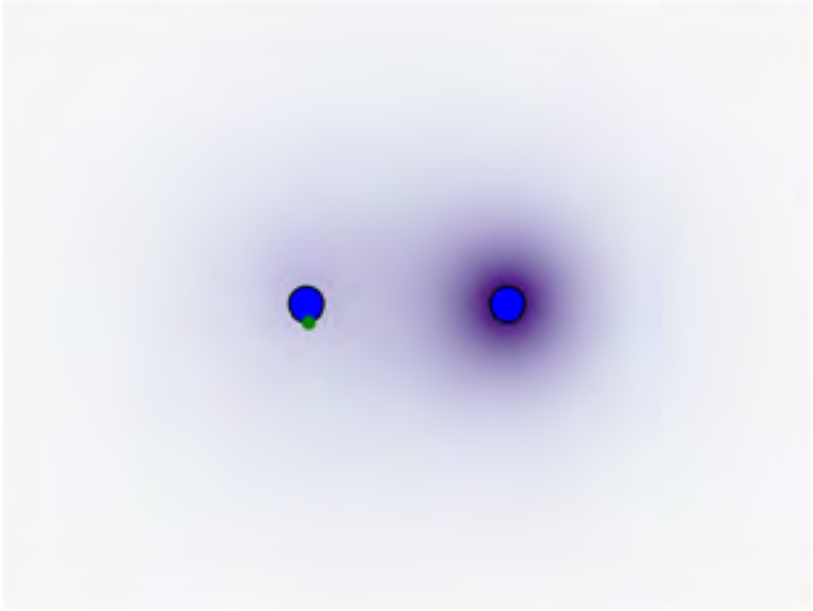
Open-source code QWalk: `http://qwalk.org`

Effective models as a data analysis problem

Effective quantum models

	Detailed	Coarse-grained
Wave function	$\Psi(r_1, r_2, r_3, \ldots)$	[0.1,-0.1,...]
Hamiltonian	$-\frac{1}{2}\sum_i \nabla_i^2 + \sum_{i<j} \frac{1}{r_{ij}} + \ldots$	Matrix
Expectation values	integral	$\Psi^T M \Psi$

Model for H_2



Reduced wave function representation

$$\begin{bmatrix} (1,1) \\ (1,2) \\ (2,1) \\ (2,2) \end{bmatrix}$$

(1,1) means that both electrons are near atom 1.

Model Hamiltonian

Want to find matrix that operates on the state vector, such that the eigenstates are the same as the original one.

$$\Psi = \begin{bmatrix} (1,1) \\ (1,2) \\ (2,1) \\ (2,2) \end{bmatrix}, \hat{H}_{\text{eff}} = \begin{bmatrix} U & t & t & 0 \\ t & 0 & 0 & t \\ t & 0 & 0 & t \\ 0 & t & t & U \end{bmatrix}$$

How to map (Hitesh Changlani)

$$\langle E \rangle = \begin{bmatrix} a & b & c & d \end{bmatrix} \begin{bmatrix} U & t & t & 0 \\ t & 0 & 0 & t \\ t & 0 & 0 & t \\ 0 & t & t & U \end{bmatrix} \begin{bmatrix} a \\ b \\ c \\ d \end{bmatrix}$$

$$= U \underbrace{(a^2 + d^2)}_{\text{double occupancy}} + t \underbrace{(ab + ac + \ldots)}_{\text{hopping}}$$

How to map (Hitesh Changlani)

$$\langle E \rangle = \begin{bmatrix} a & b & c & d \end{bmatrix} \begin{bmatrix} U & t & t & 0 \\ t & 0 & 0 & t \\ t & 0 & 0 & t \\ 0 & t & t & U \end{bmatrix} \begin{bmatrix} a \\ b \\ c \\ d \end{bmatrix}$$

$$= U \underbrace{(a^2 + d^2)}_{\text{double occupancy}} + t \underbrace{(ab + ac + \ldots)}_{\text{hopping}}$$

$$\langle E \rangle = U(\text{double occupancy}) + t(\text{hopping})$$

All quantities in blue can be evaluated using detailed (real space) simulations.

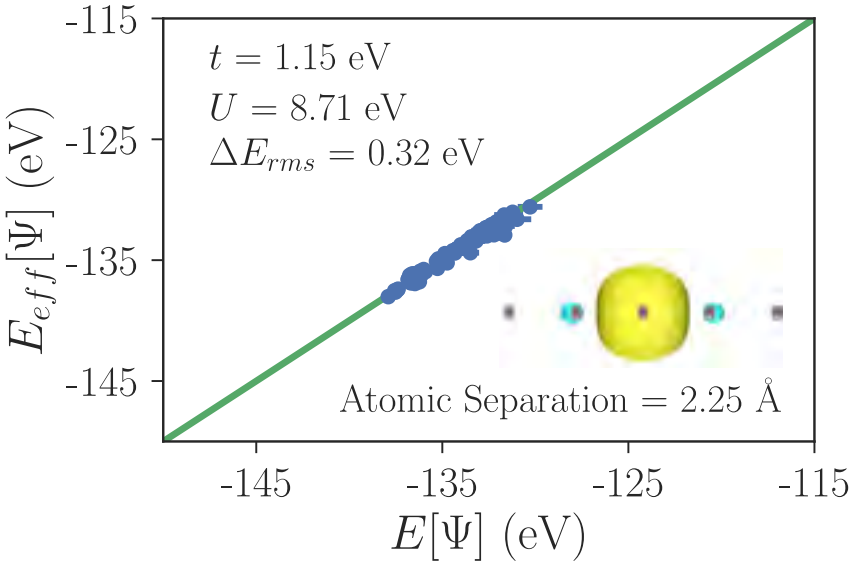
The algorithm

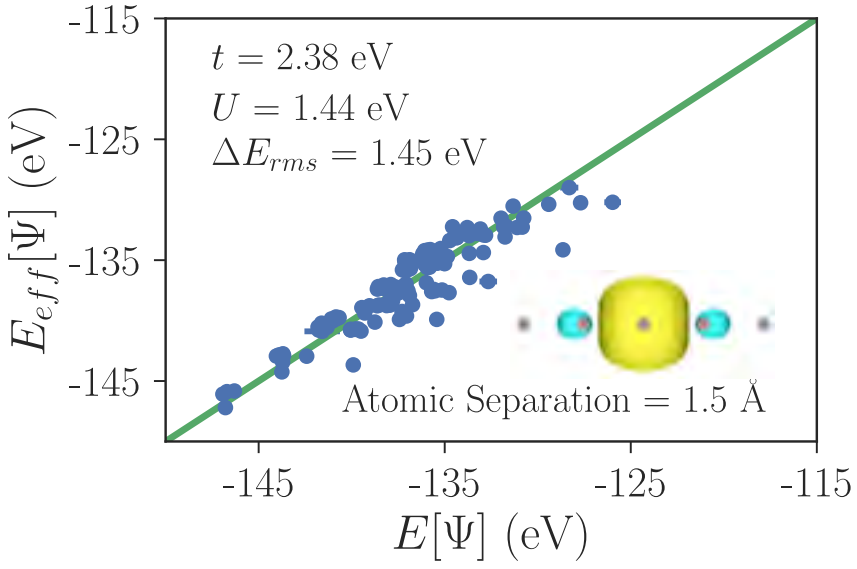
$$\langle H \rangle = U(\text{double occupancy}) + t(\text{hopping})$$

1. Generate wave functions in the low-energy space of interest
2. Accumulate expectation value of energy and "descriptors" (e.g. double occupancy and hopping)
3. $E_k \simeq \sum_i c_i d_k[\Psi_k]$; find c_i to minimize deviation

Proofs that this is the right thing to do:
Frontiers in Physics. DOI:10.3389/fphy.2018.00043

A chain of H atoms (Kiel Williams)





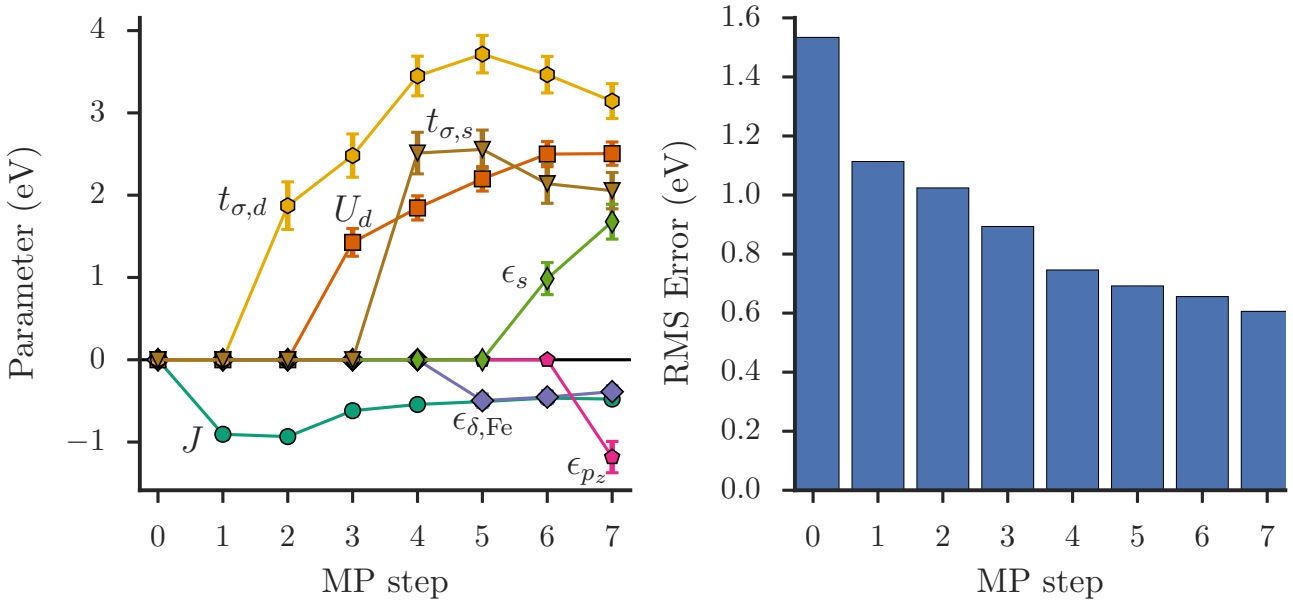
Good model when atoms are well-separated, poor when the atoms are too close (need more long-range terms)

Applications to real materials

More complex materials (Brian Busemeyer)

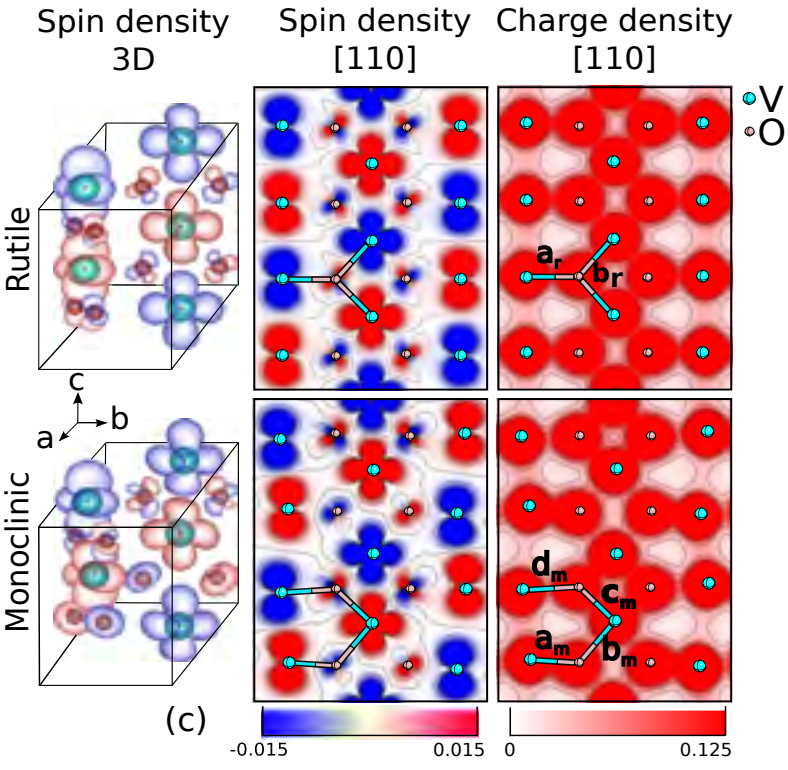
Fe=Se molecule

21 symmetry-allowed parameters



Use matching pursuit to select best parameters

Vanadium dioxide (Huihuo Zheng)



Metal-insulator transition at 340 K.

Insulating state not well understood. Spin excitation?

Measurement: 460 meV[a]

Calculation: 440(24) meV[b]

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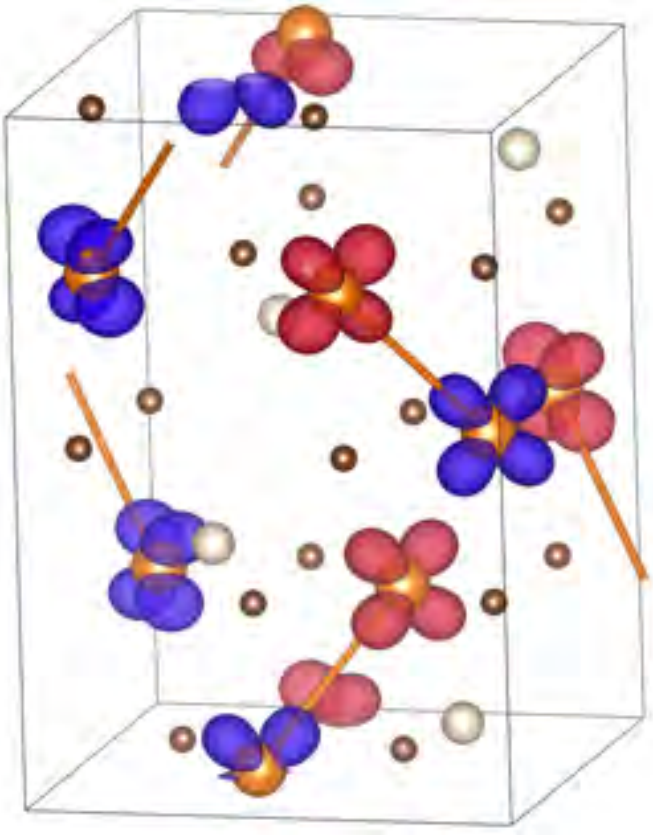
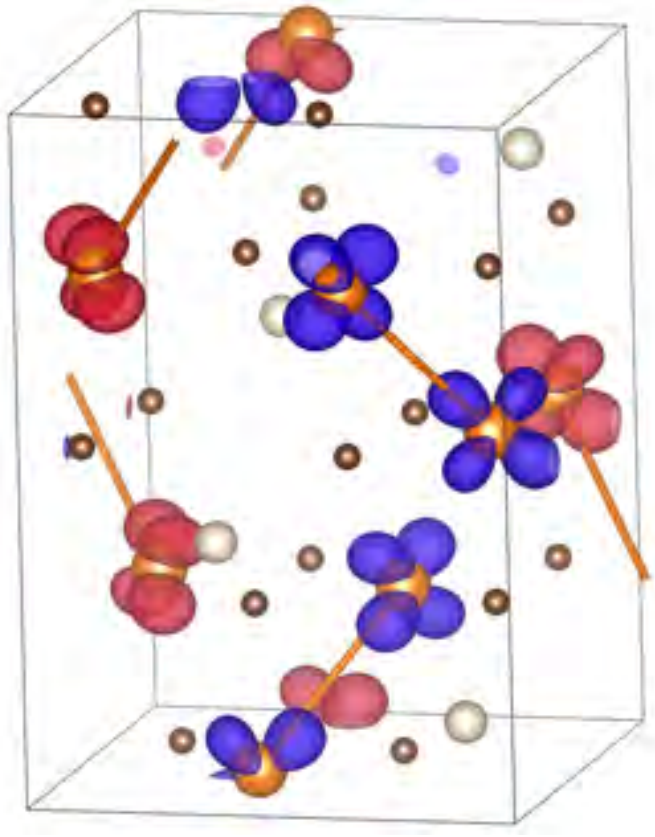
[a] He et al. PRB **94**, 161119 (2016).

[b] Zheng, Wagner PRL **120**, 059901(E) (2018)

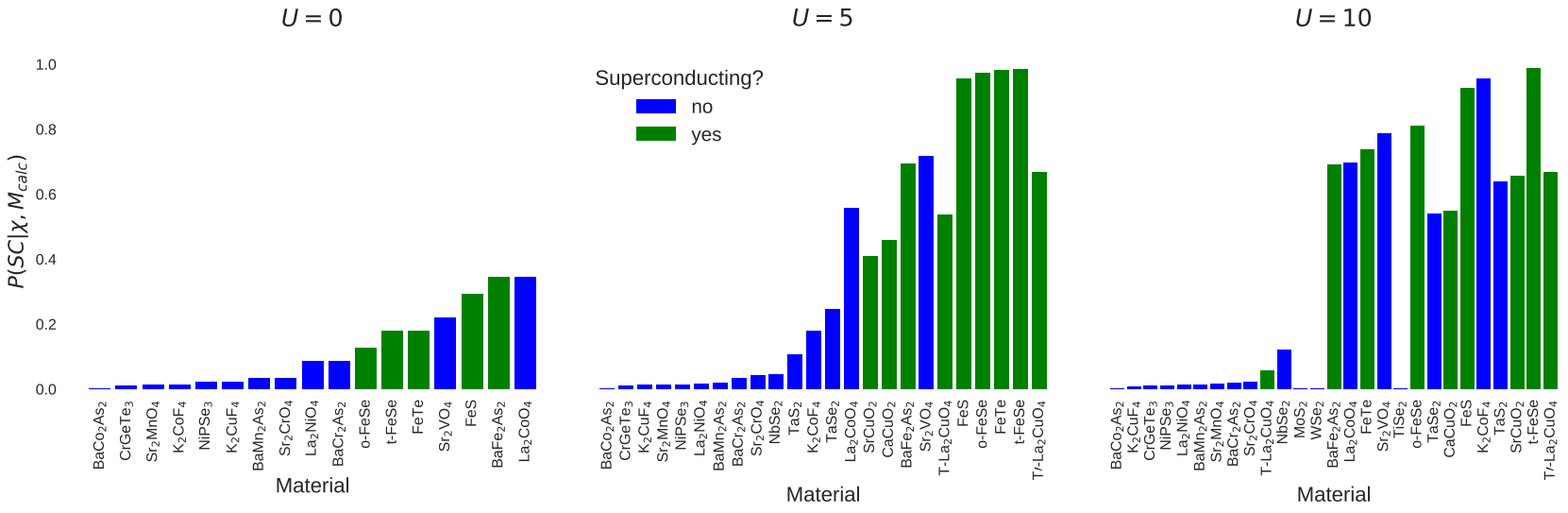
$MgTi_2O_4$ (Brian Busemeyer)





Another spin excitation: used Blue Waters to make a prediction of an excitation at 350(50) meV. Hoping experiments will test this!

Predicting superconductivity classes (Joao Nunes Rodrigues)



Predictor derived through the model fitting technique.

Separates superconductors from non-superconductors with high fidelity.

Relevance of Blue Waters

Need to compute expectation values of many many-particle wave functions via Monte Carlo:

$$\langle O \rangle = \int \Psi^*(r_1, r_2, \ldots) \hat{O} \Psi(r_1, r_2, \ldots)$$

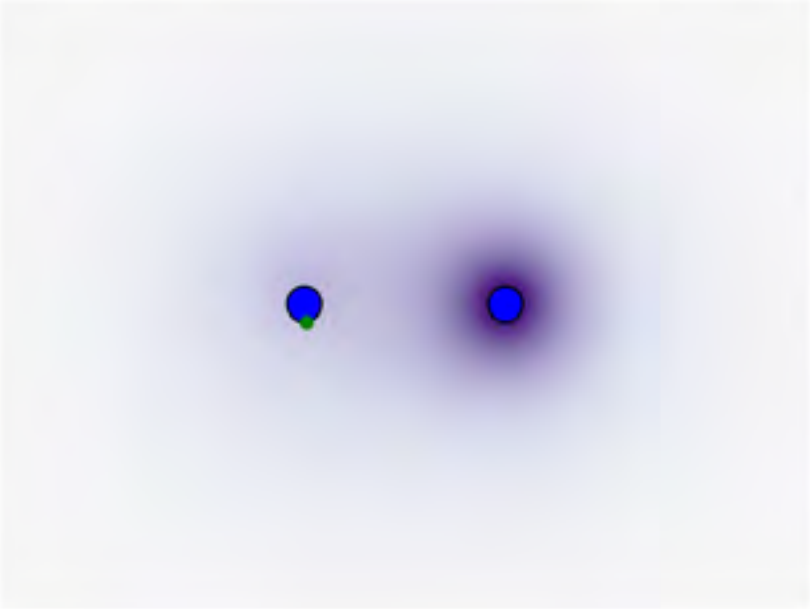
Massively parallel and high throughput.

Need to generate a moderate amount of high-cost data.

Open questions

- Can we automatically sample good quality wave functions? Usually we rely a lot on physical understanding.
- Can we make QMC faster? (probably algorithms)
- Best representation of coarse-grained model: finding basis.

The end



$$\rightarrow \begin{bmatrix} U & t & t & 0 \\ t & 0 & 0 & t \\ t & 0 & 0 & t \\ 0 & t & t & U \end{bmatrix}$$