Predictive computing for condensed matter

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Peter Abbamonte & Wen-Mei W. Hwu (UIUC)

Strategic goal

Suite of programs that can predict the properties of materials with unique and highly correlated electronic structure

- High accuracy
- Material specificity

Quantum mechanics of materials

$$\hat{H}|\Psi_i\rangle = E_i|\Psi_i\rangle$$

$\hat{H}$ Linear operator (matrix or differential operator)

$|\Psi_i\rangle$ Function or vector, state of the system

For N particles:

Dimensionality $\mathbb{R}^N$ or $N_{lattice}^N$

The wave function

$|\Psi_i\rangle = c_1 |$ $\rangle + c_2 |$ $\rangle$

Two cats: curse of dimensionality

$|\Psi_i\rangle = c_1 |$ $\rangle$

$+ c_2 |$ $\rangle$

$+ c_3 |$ $\rangle$

$+ c_4 |$ $\rangle$

Fermionic solution

$|\Psi_i\rangle$ = | ⟩ - | ⟩

Not an extreme eigenvector of the Hamiltonian

$$\binom{\text{states}}{\text{particles}}$$ combinations

Strongly correlated condensed matter

DFT and others: separate into effective single particle problems

Many materials in which this approach seems difficult or impossible:

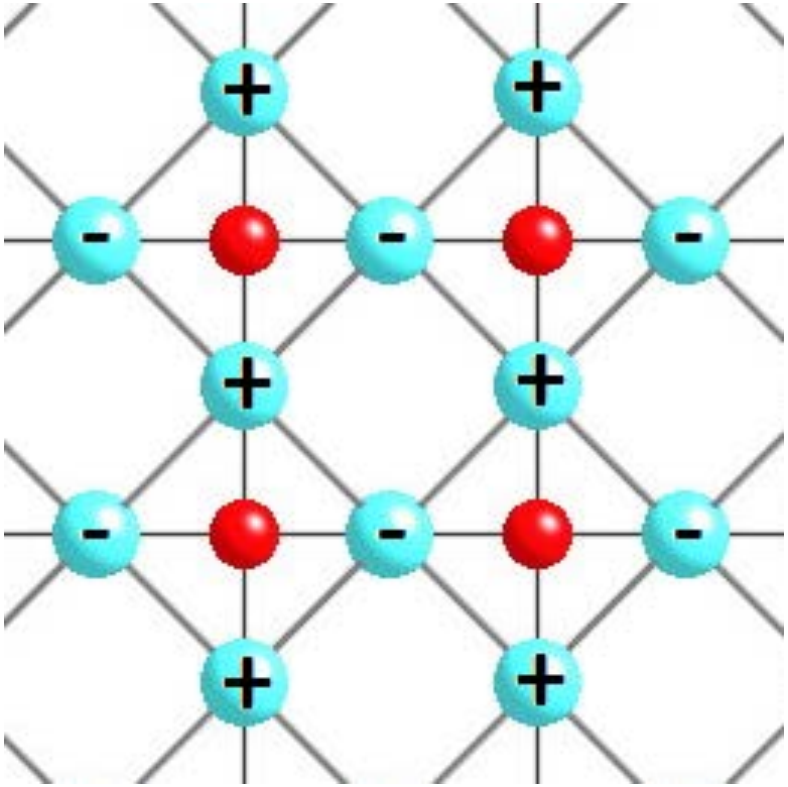
- High-Tc superconductors
- Metal-insulator transition in VO_2
- Collossal magnetoresistance
- Excited states in many materials
- Ice and liquid water

Our strategy

Reduce the size of the vector space in a controllable way

Quantum Chemistry	**Density matrix methods**	**Quantum Monte Carlo**
Systematically expand in terms of orders of determinants	Assume short-ranged correlations	Sample entire space with constraint
$N^{4\text{-}7}$	Various scaling	N^3
Well-established, very accurate on molecules	Established for 1D systems	Any system, often accurate
Scales to ~1000 cores	Parallelism limited by linear algebra	Scalability to >100k cores

Need for large-scale computing
Example case: quantum Monte Carlo



Number of electrons: ~300

900 dimensional function!

+/- 0.02 eV :100 million function evaluations

Many independent-> excellent parallel scaling >100k processors

A brief introduction to our team

So Hirata
UIUC

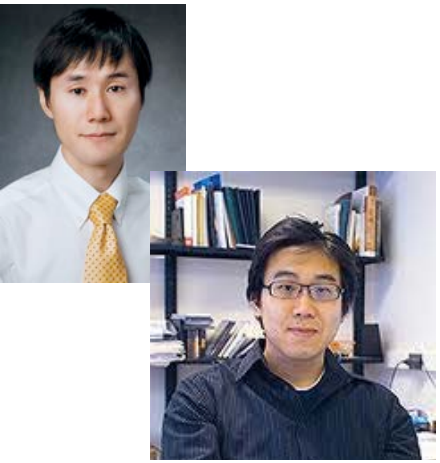
Extend quantum chemistry to solids
Monte Carlo and parallelization methods

Shiwei Zhang
William and Mary

Excited states using AFQMC
Coarse-grained models

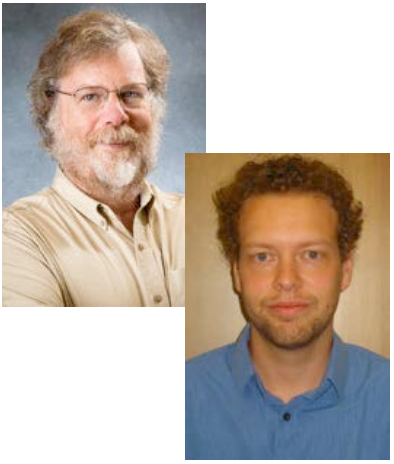
Garnet Chan
Bryan Clark
Princeton

Improved solvers for coarse grained models



Shinsei Ryu
UIUC
Garnet Chan
Princeton

Development of a new form for correlated wave functions based on tensor networks



David Ceperley
Lucas Wagner
UIUC

Improved accuracy and speed in continuum quantum Monte Carlo methods

Peter Abbamonte
UIUC

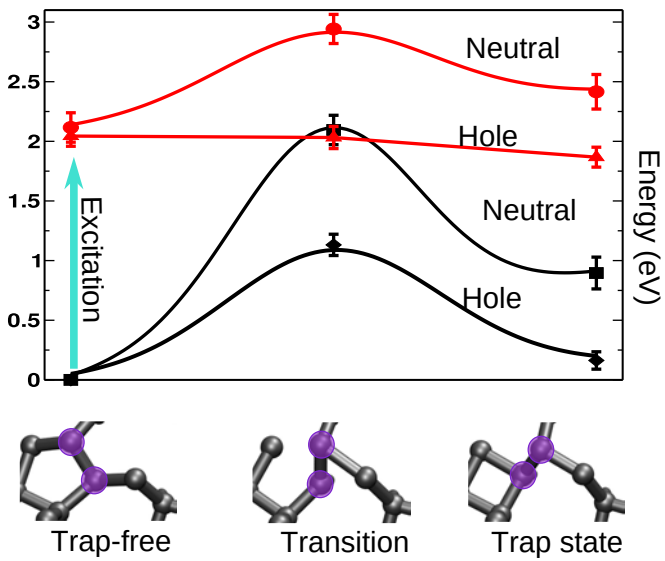
Iterative computation/ theory and experimental cycles

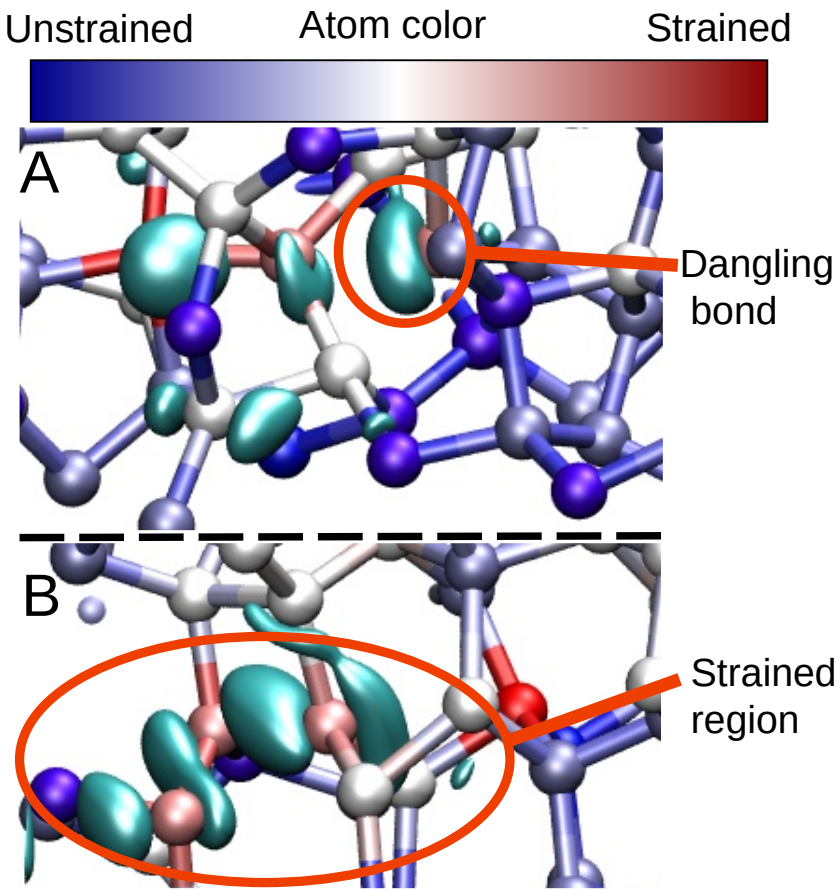
Some historical successes with materials related to energy

Amorphous silicon

New mechanism for light-induce degradation in amorphous silicon

Barriers in excited states by QMC

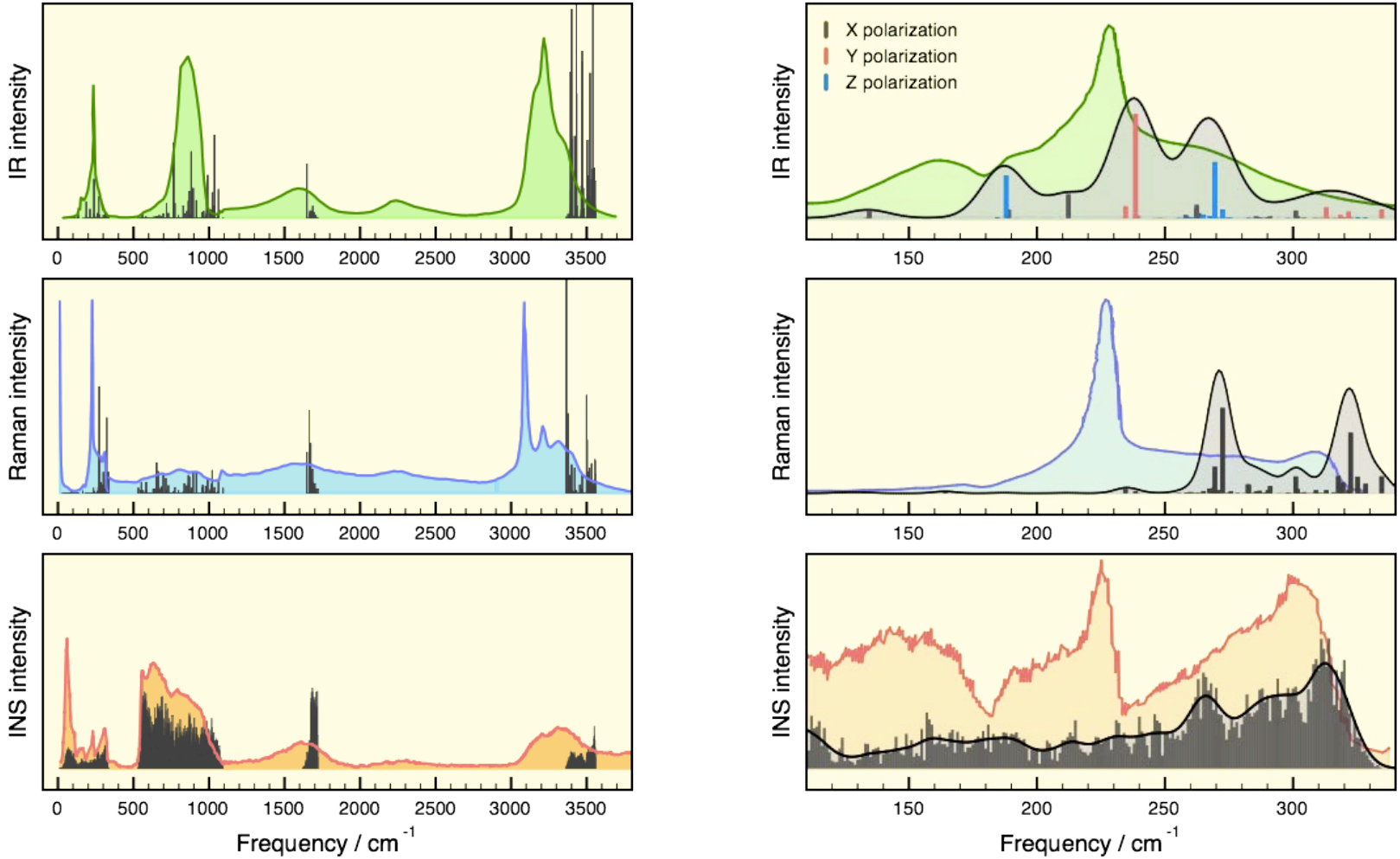




L.K. Wagner and J.C. Grossman
Phys. Rev. Lett. 101, 265501 (2008)

Ice

He, Sode, Xantheas, and Hirata (2012)



Excited states in solids

- AFQMC approach for excited states
- First attempt in solids: ***preliminary results***
- Excellent agreement with experiment and FN-DMC

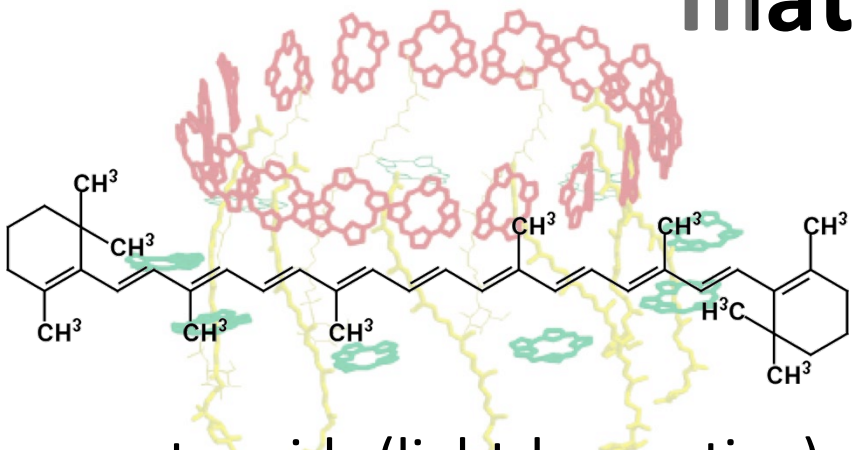
Band gap in ZnO (wurtzite)

method	***Band gap*** (eV)
LDA	0.6
LDA+*U*	1.0
Hybrid functionals	3.3; 2.9
GW	2.4
AFQMC*	**3.20(13)**
FN-DMC&	**3.1(2)**
experiment	**3.2-3.4**

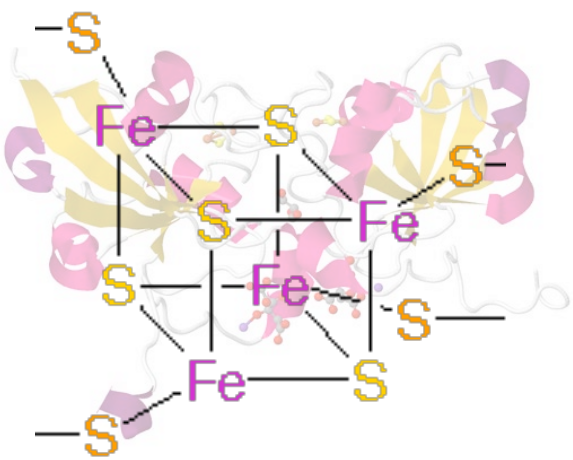
*Ma, Zhang, Krakauer, to be published (2012)

&Ertekin, Wagner, Grossman, in preparation (2012)

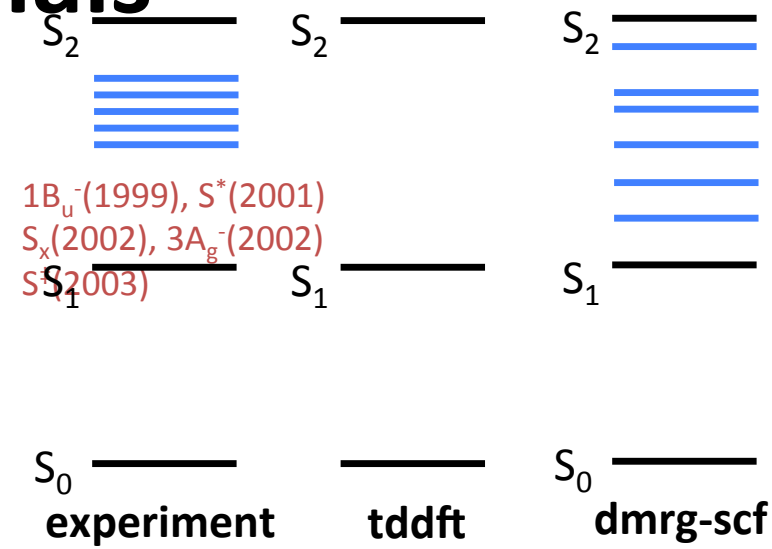
DMRG calculations of light-harvesting materials

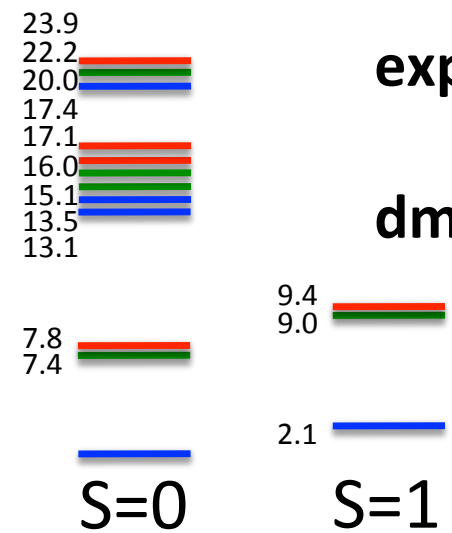


carotenoids (light-harvesting)



iron-sulfur complexes (life!)





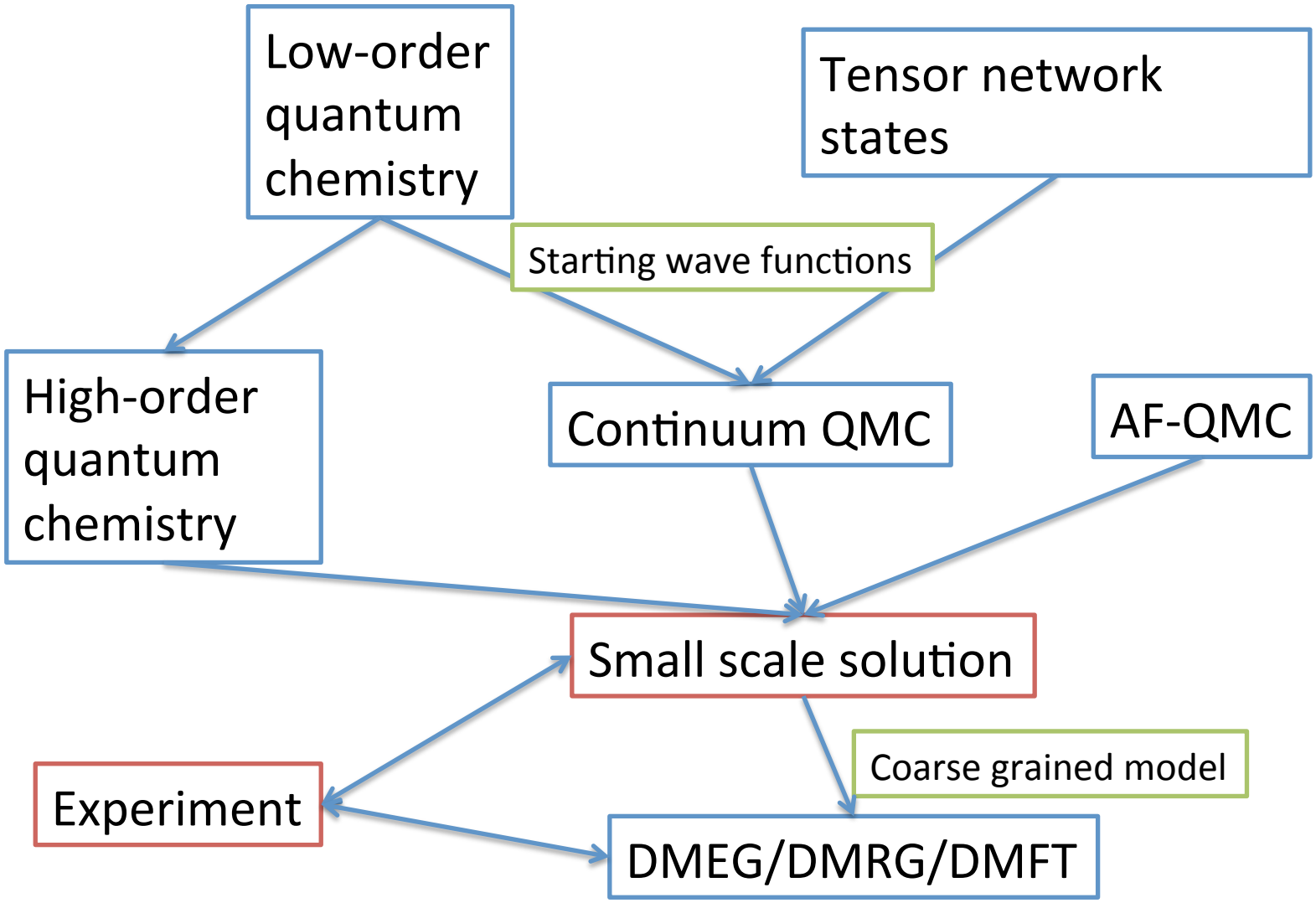
expt: J ~ 250 cm^{-1}

B unknown

dmrg: J ~ 240 cm^{-1}

B ~ 450 cm^{-1}

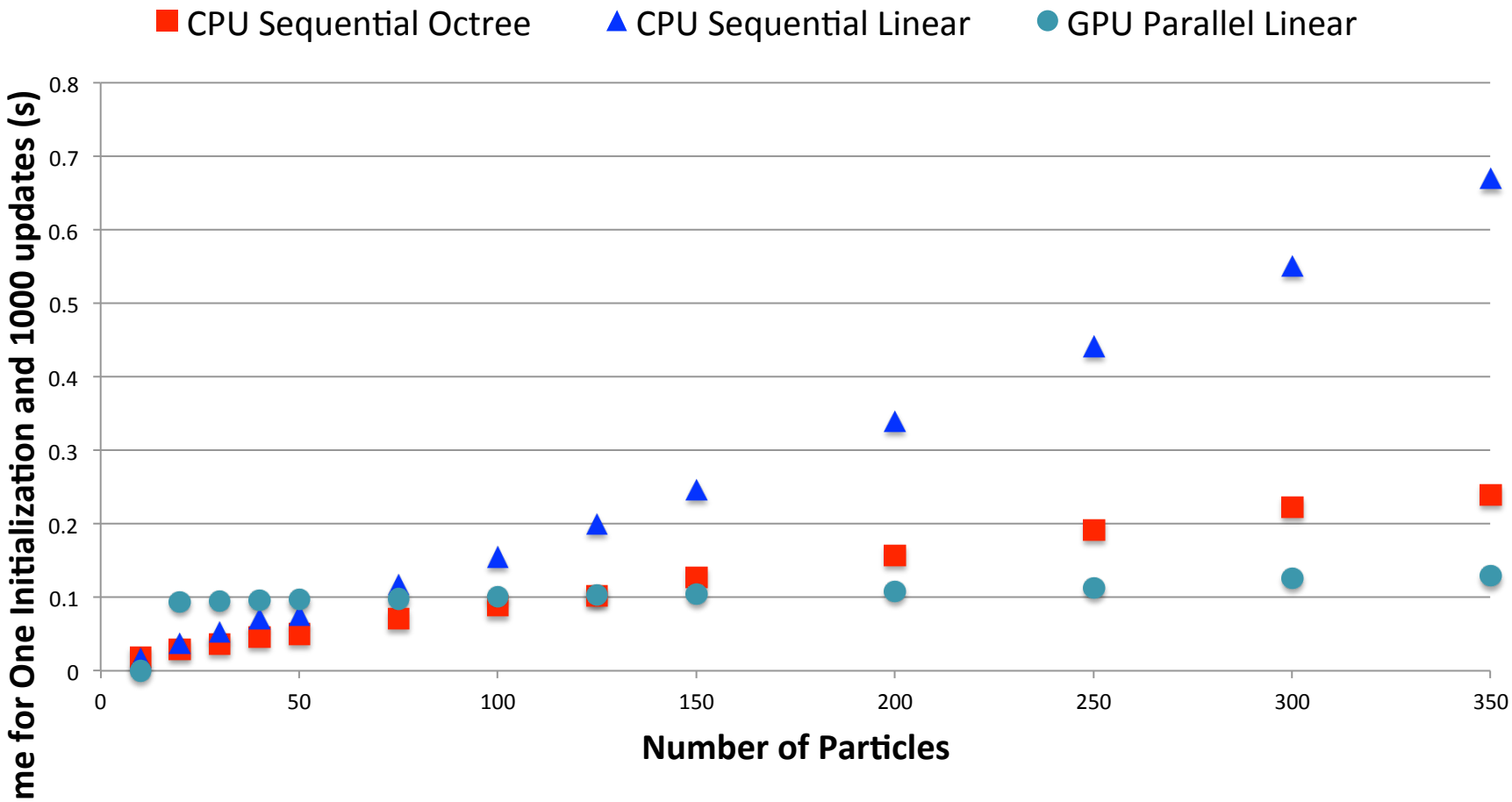
Interactions



Computational assessment

	Linear algebra	Function evaluation	Data storage and retrieval	Parameter set discovery & estimation
AFQMC		1-electron integrals		
Continuum QMC	Determinant updates	Several		
FCI-QMC			Matrix elements	
Density matrix methods	Sparse matrix eigenvalues			
Quantum chemistry	Several		Matrix elements	
Model building				Find minimal descriptive set at low energy

CPU vs GPU Initialization and Update Timings: Particle particle distances



Dillon Skeehan, Caroline Marcks, and LKW
POP REU 2012

Computational science team

- Collaboration with SUPER institute for code optimization
- Advising by W.M. Hwu at Illinois for GPU code

Potential interactions:

- FastMath for large eigenvalue problems
- QUEST for model fitting and verification

Software

QWalk: full-featured continuum quantum Monte Carlo package

- >100 users on the mailing list
- Works on all leadership computing platforms
- Scales to 100k processors or more
- Open source: http://www.qwalk.org

Lead developer: Lucas Wagner

For the other projects, new as of yet unnamed software will be developed

Expected Results

- A suite of software that produce accurate wave functions for strongly correlated systems and accurate coarse-grained models
- Multiple accurate (but approximate) methods that can be compared
- New understanding of electrons and electron-lattice interactions in strongly correlated systems