U.S. Department of Energy SciDAC Partnership

Predictive Computing for condensed matter

University of Illinois at Urbana-Champaign | Princeton University | College of William & Mary

So Hirata (Chemistry, UIUC) - Lead PI

Hirata is a theoretical/computational chemist and an expert in electron-correlation theories for molecules and solids. He is the primary author of the computer-generated, high-rank electron-correlation modules in DOE's massively parallel NWCHEM suite of software, implemeting several of his original methods.

Lucas Wagner (Physics, UIUC) - Co-lead PI

Wagner is the principal author of the quantum Monte Carlo program, QWALK, with which he has performed predictively accurate calculations on strongly correlated systems.

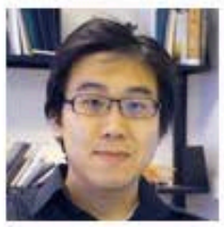
Peter Abbamonte (Physics, UIUC)

Abbamonte, an experimental condensed-matter physicist, brings an invaluable experimental insight into the project. He is one of the originators of resonant soft x-ray scattering, with which he discovered a Wigner crystal in doped spin ladders and the charged stripes in copper-oxide superconductors.

David Ceperley (Physics, UIUC)

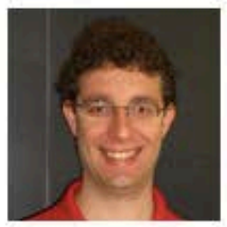
Ceperley is a theoretical/computational physicist and an authority of quantum Monte Carlo (QMC). He invented a number of QMC algorithms and is the author of massively parallel QMCPACK software.

Garnet K.-L. Chan (Chemistry, Princeton)



Chan is a recognized expert in strong correlation theories including density matrix renormalization group (DMRG), tensor networks, and density matrix functional theory. He has, in particular, established DMRG as a practical, powerful tool for strongly correlated molecular electronic structures.

Bryan Clark (Physics, UIUC)



Clark has considerable experience in both conventional quantum Monte Carlo (QMC) and novel extensions such as QMC in the Hilbert space. He has developed a large-scale parallel algorithm of QMC in PIMC++.

Shinsei Ryu (Physics, UIUC)

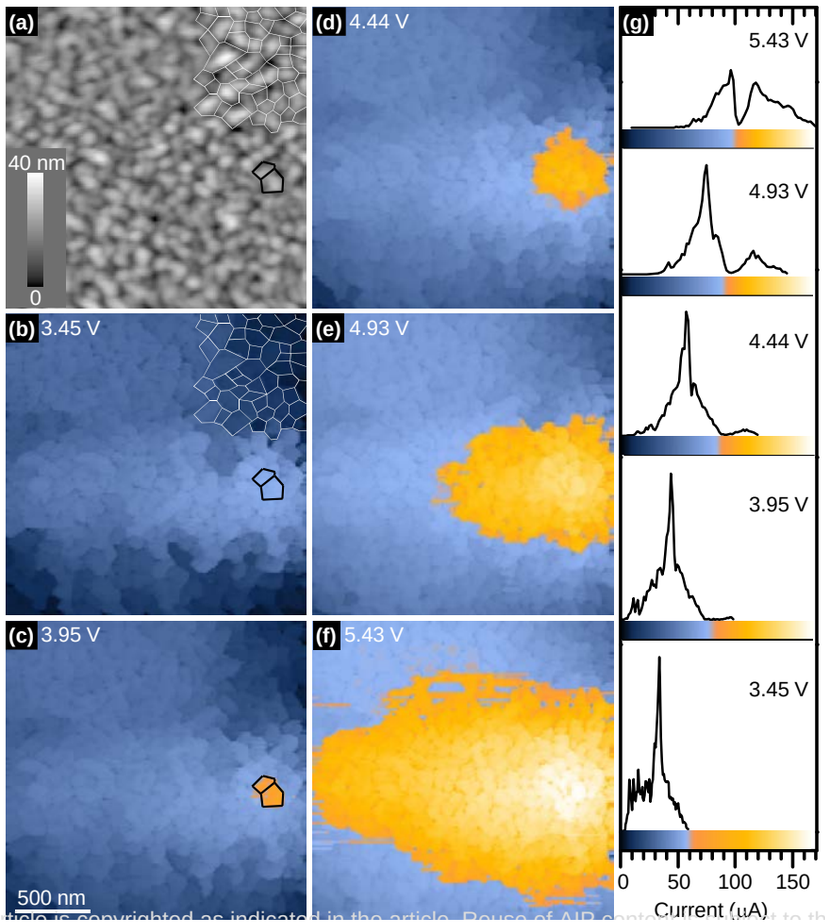
Ryu specializes in mathematical theories of strong correlation and other condensed phase electronic structures. Ryu is a pioneer in the use of entanglement entropy in classifying topological phases of matter and has predicted the fractional topological insulator in two dimension.

Shiwei Zhang (Physics, W&M)

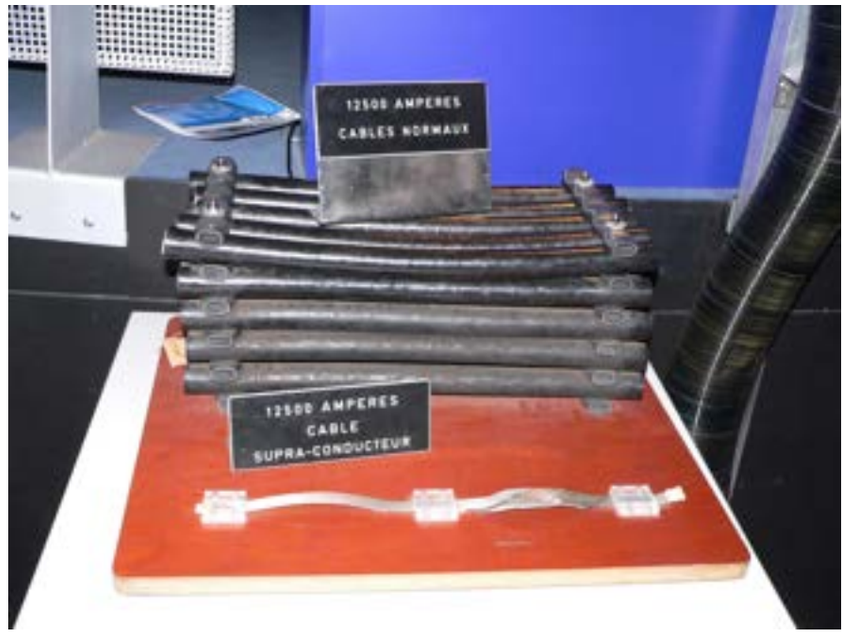
Zhang specializes in computational condensed matter physics and materials science. Zhang is a pioneer in the use of quantum Monte Carlo (QMC) in the Hilbert space for strong correlation. He is the inventor of the phaseless auxiliary field QMC.

Financial support of this project is provided through Scientific Discovery through Advanced Computing (SciDAC) program funded by U.S. Department of Energy, Office of Science, Advanced Scientific Computing Research and Basic Energy Sciences under award number DE-FG02-12ER46875.

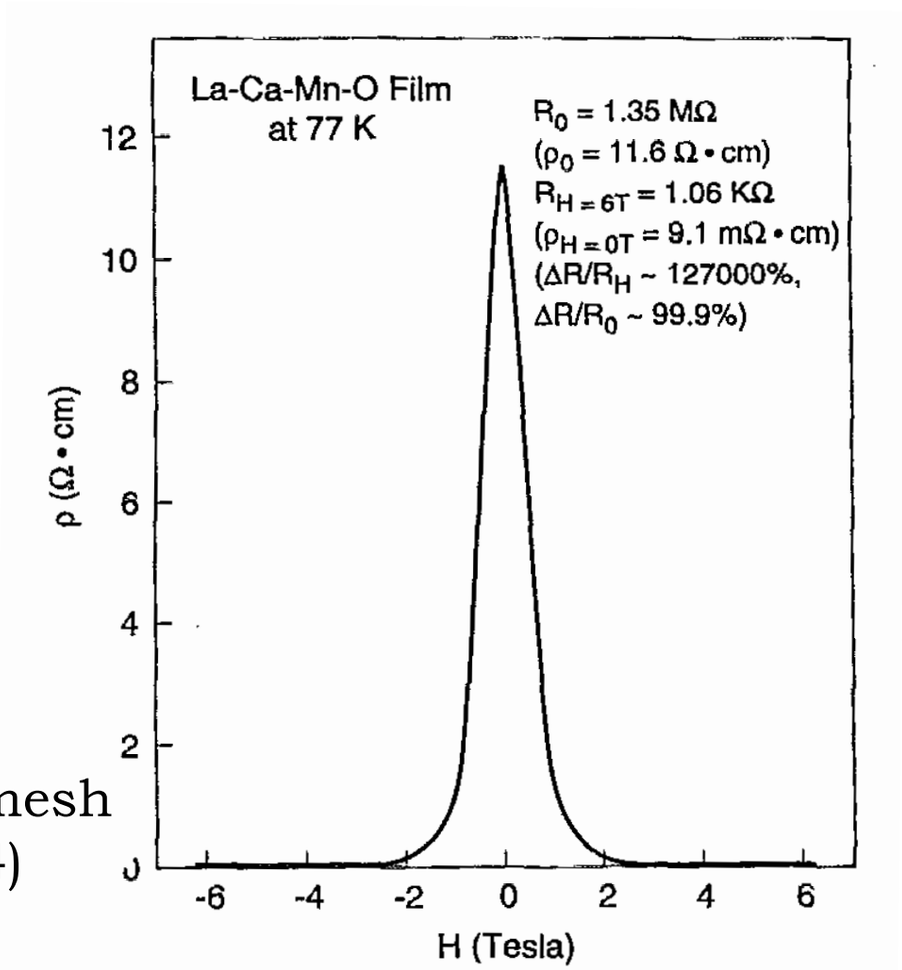
Strongly correlated condensed matter



Kim, Ko, Frenzel, Ramanathan, Hoffman
Applied Physics Letters 96, 213106 (2010)

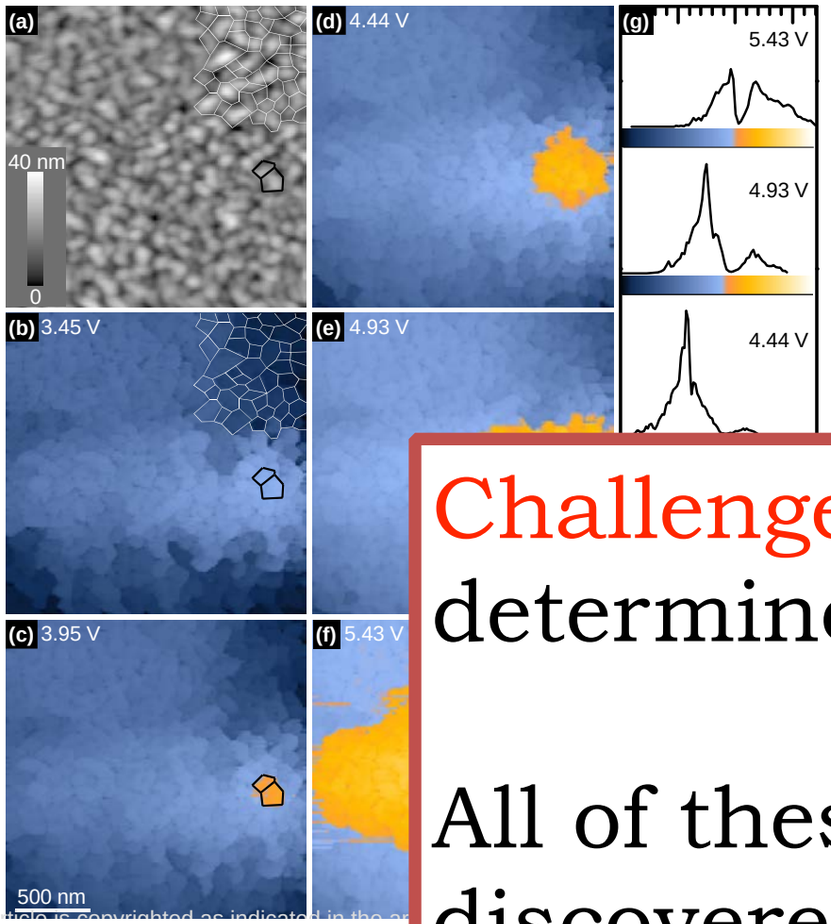


CERN



Jin, McCormack, Tiefel, Ramesh
J. App. Phys. 76 6929 (1994)

Strongly correlated condensed matter

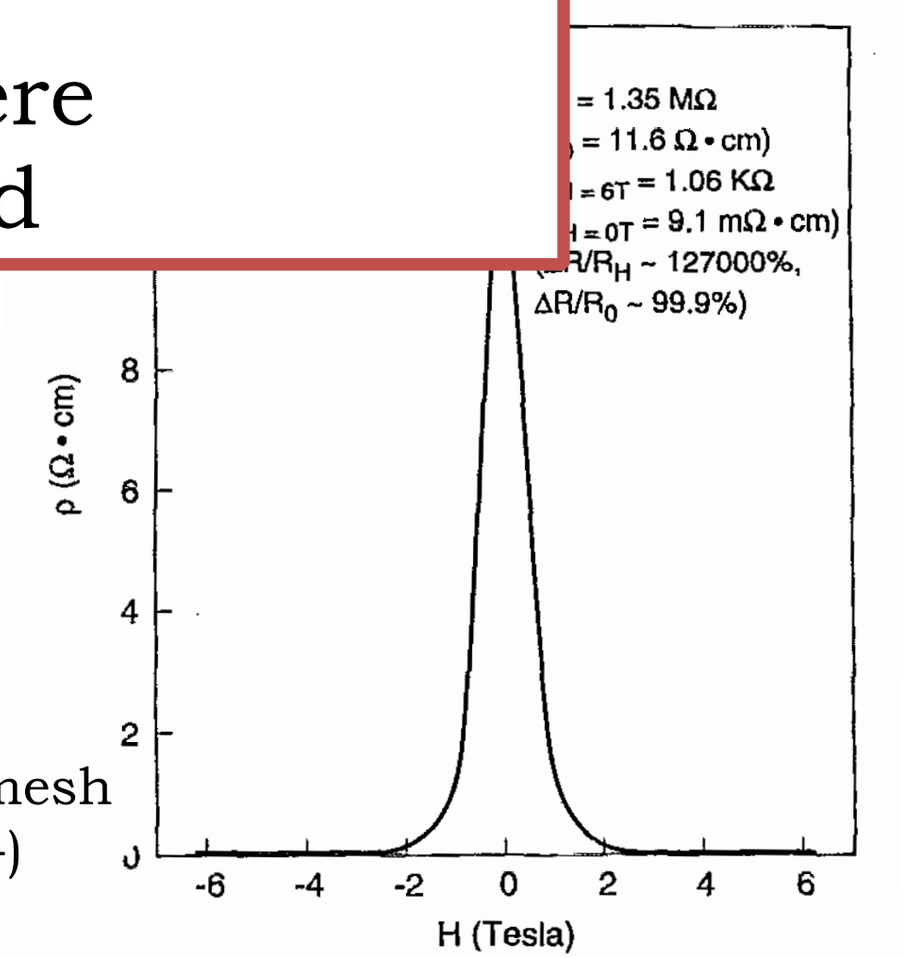


CERN

Challenge: electronic state is subtly determined by electron correlations!

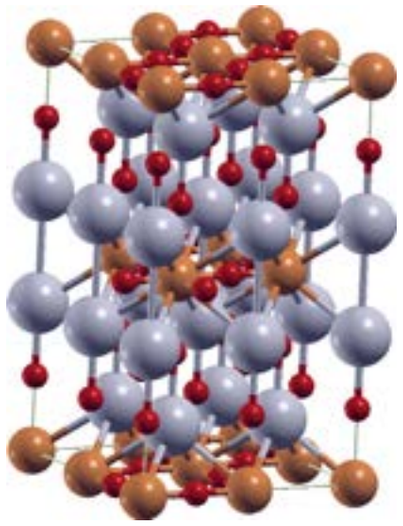
All of these materials were discovered, not predicted

Kim, Ko, Frenzel, Ramanathan, Hoffman
Applied Physics Letters 96, 213106 (2010)



Jin, McCormack, Tiefel, Ramesh
J. App. Phys. 76 6929 (1994)

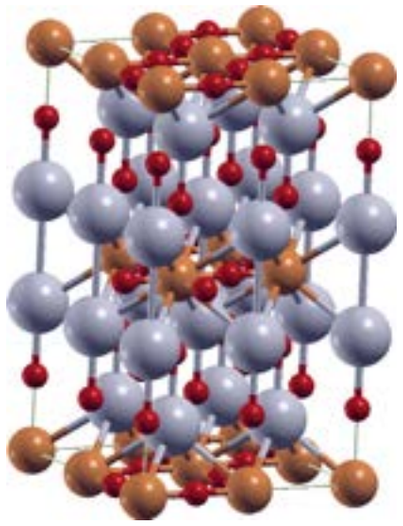
Objective

Start with  + $H = -\sum_i \frac{1}{2}\nabla_i^2 - \sum_{\alpha i} \frac{Z_\alpha}{r_{i\alpha}} + \sum_{\alpha\beta} \frac{Z_{\alpha\beta}}{r_{\alpha\beta}} \sum_{ij} \frac{1}{r_{ij}}$

Solve $E_i \Psi_i(r_1, r_2, \ldots) = \hat{H} \Psi_i(r_1, r_2, \ldots)$

minimal approximations including electron correlations explicitly

Objective

Start with  + $H = -\sum_i \frac{1}{2}\nabla_i^2 - \sum_{\alpha i} \frac{Z_\alpha}{r_{i\alpha}} + \sum_{\alpha\beta} \frac{Z_{\alpha\beta}}{r_{\alpha\beta}} \sum_{ij} \frac{1}{r_{ij}}$

No U, no J, no t

Solve $E_i \Psi_i(r_1, r_2, \ldots) = \hat{H} \Psi_i(r_1, r_2, \ldots)$

minimal approximations including electron correlations explicitly

Fundamental equation

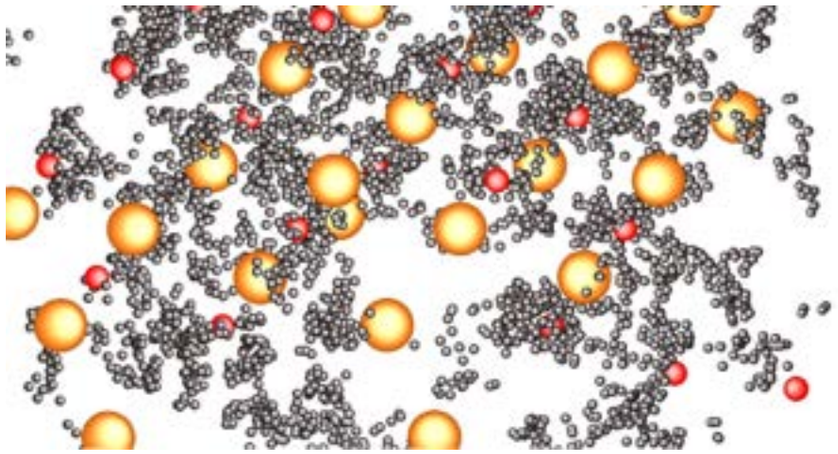
$$\hat{H}|\Psi_i\rangle = E_i|\Psi_i\rangle$$

$$\hat{H} = -\sum_i \frac{1}{2}\nabla_i^2 - \sum_{\alpha i} \frac{Z_\alpha}{r_{i\alpha}} + \sum_{\alpha\beta} \frac{Z_{\alpha\beta}}{r_{\alpha\beta}} \sum_{ij} \frac{1}{r_{ij}}$$

$$\Psi(r_1, r_2, r_3, \ldots)$$

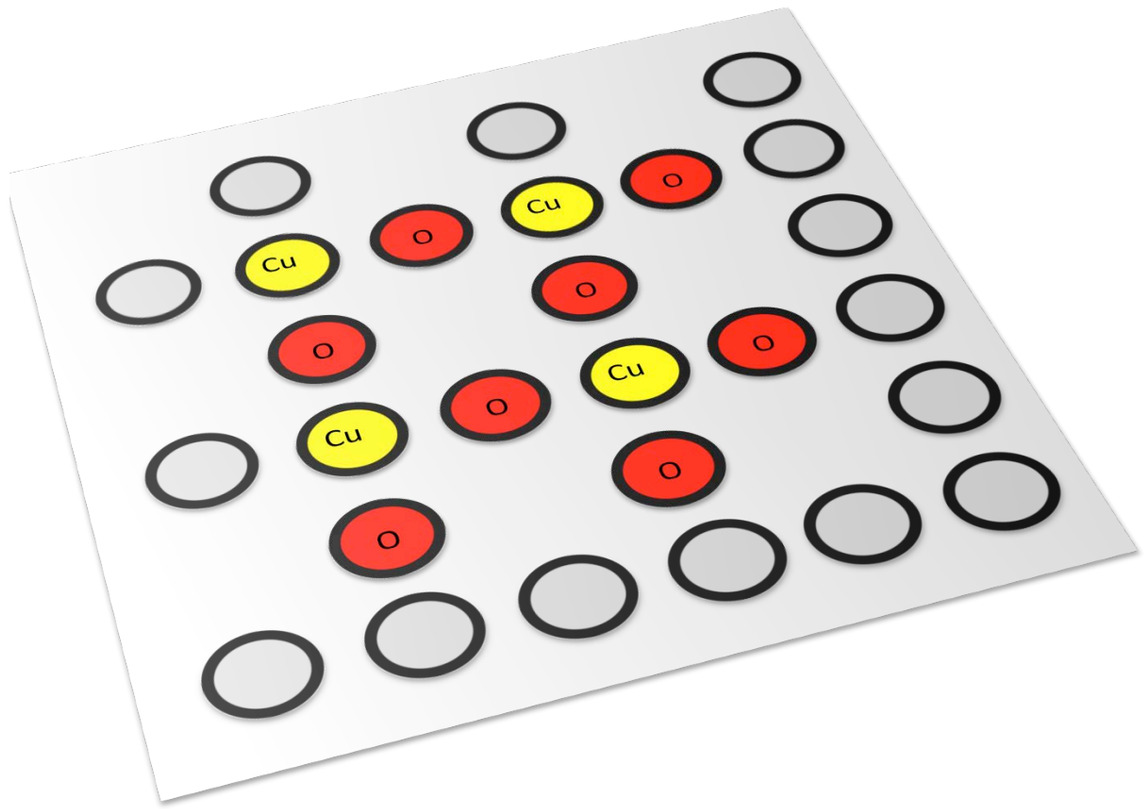
This is not a theoretical problem; this is a computational problem!

PDE in many dimensions (thousands).



Start with sub-angstrom resolution of electron correlations

To be able to describe mesoscale correlated physics



Method	Type of Hilbert space	Major approximation	Variational principle	Investigator	Code
FN-DMC	Continuum	Node of wave function	Yes	Ceperley, Wagner	QWalk, QMCPACK
AF-QMC	Discrete	Phase of wave function	Somewhat	Zhang	development
DMRG/ DMET	Discrete	Local matrix product states	Yes	Chan	available
MERA	Discrete	Ansatz	Yes	Ryu	development
MP perturbation theory	Discrete	Perturbation /interaction strength	No	Hirata	development

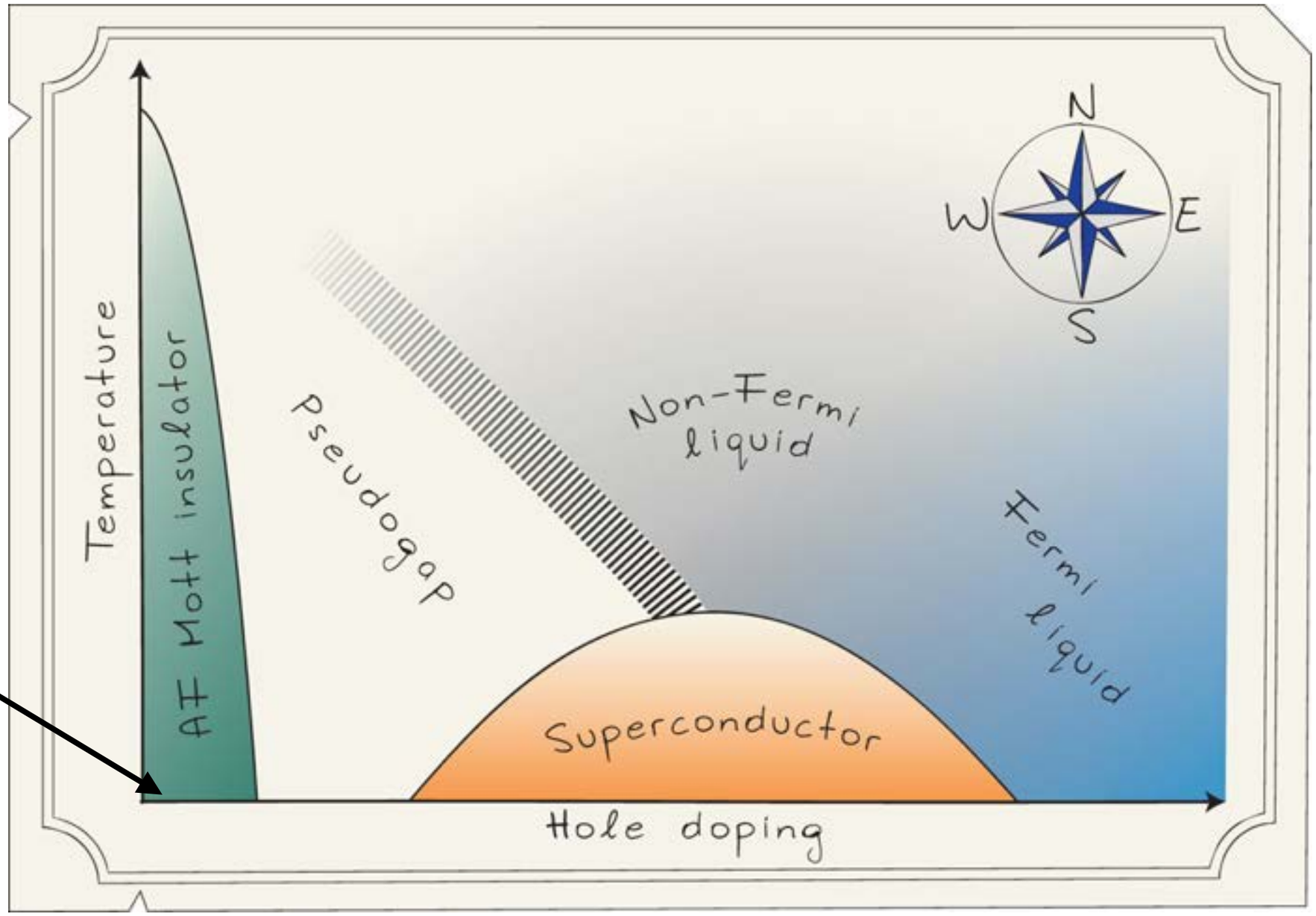
Benchmarking

The methods are all highly accurate compared to traditional electronic structure techniques like density functional theory.

How accurate?

Where do they fail?

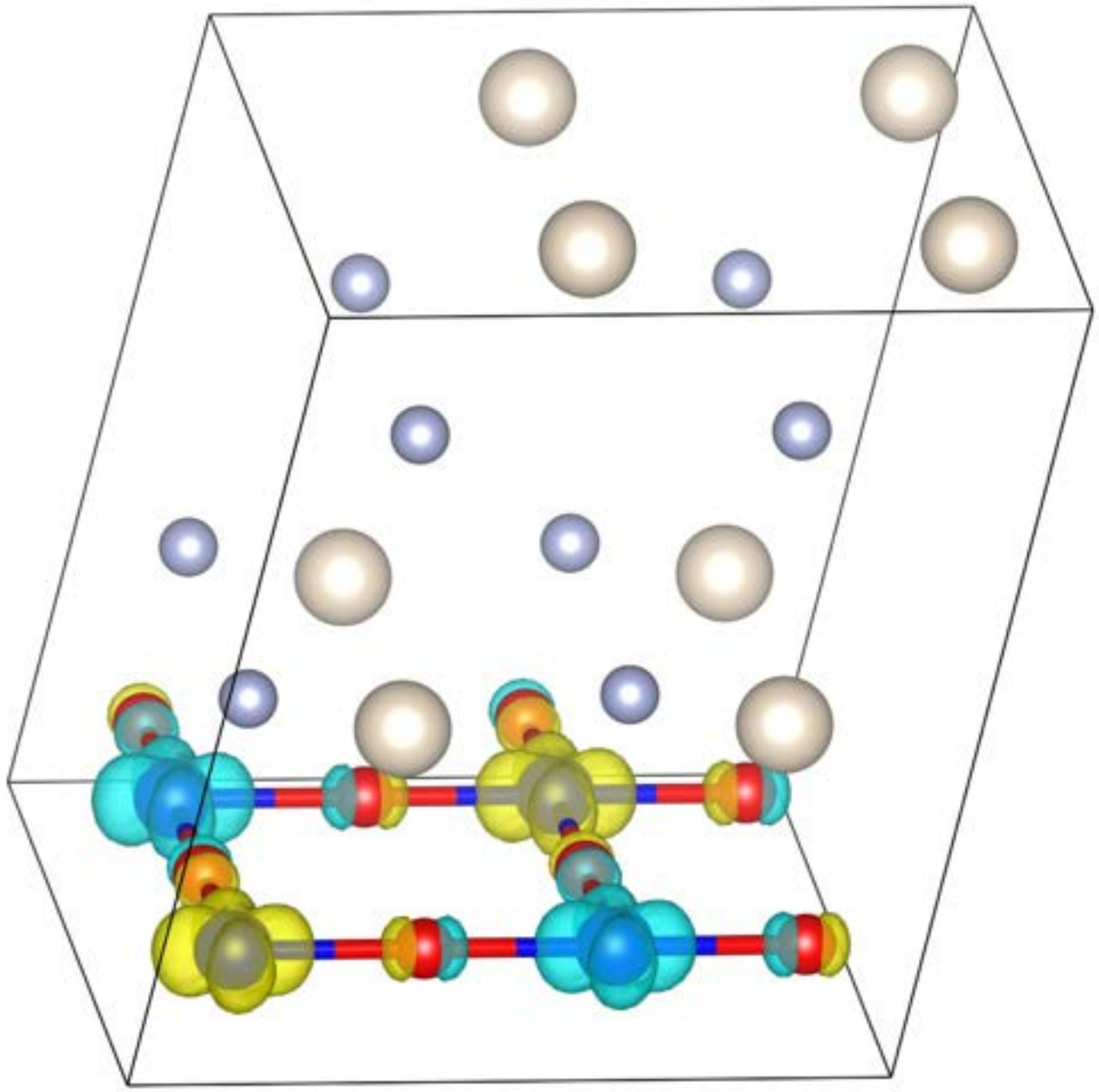
Electronic structure of the cuprates



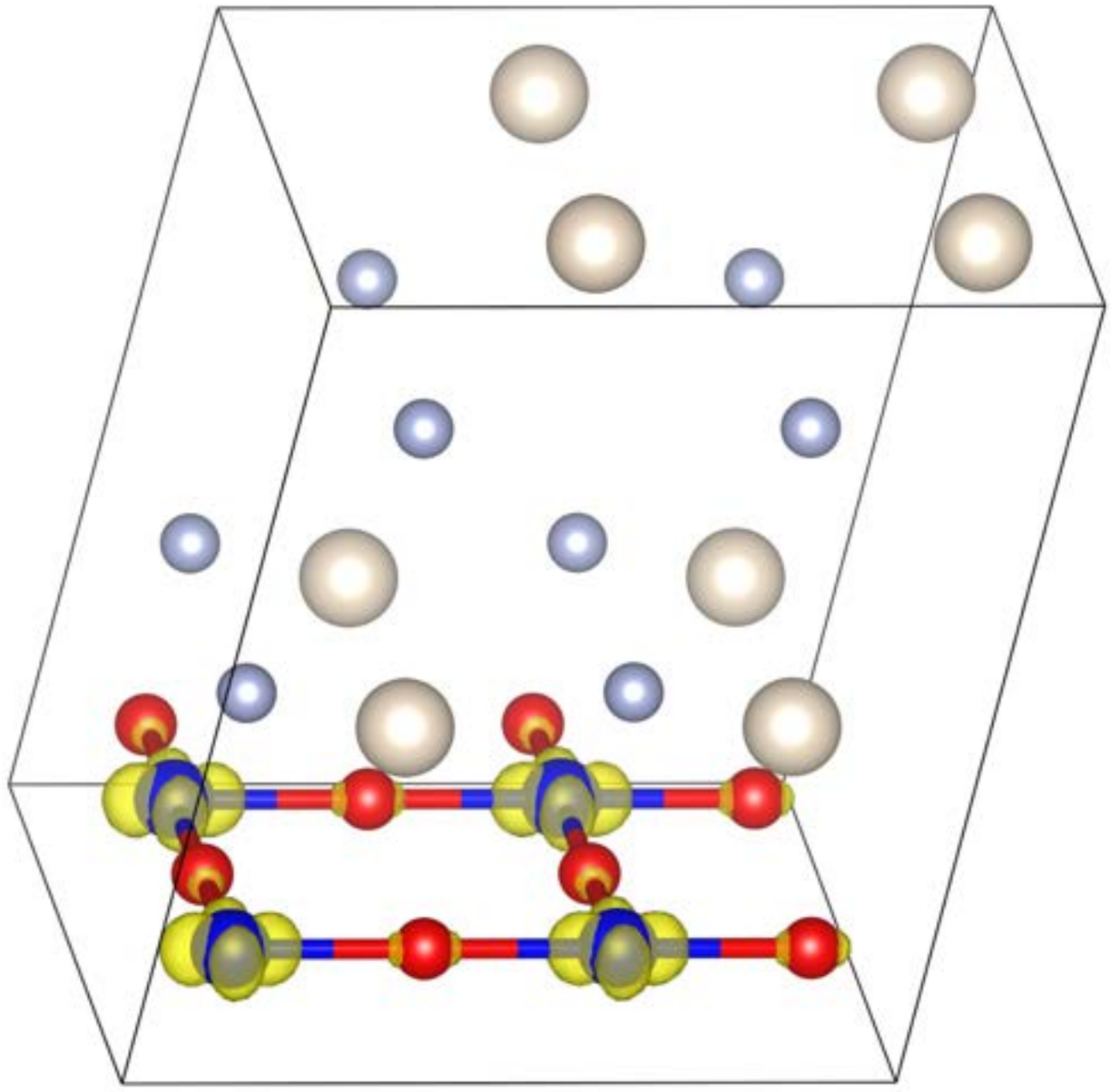
Test here

Standard methods fail to describe any of this!

FN-DMC: magnetic properties

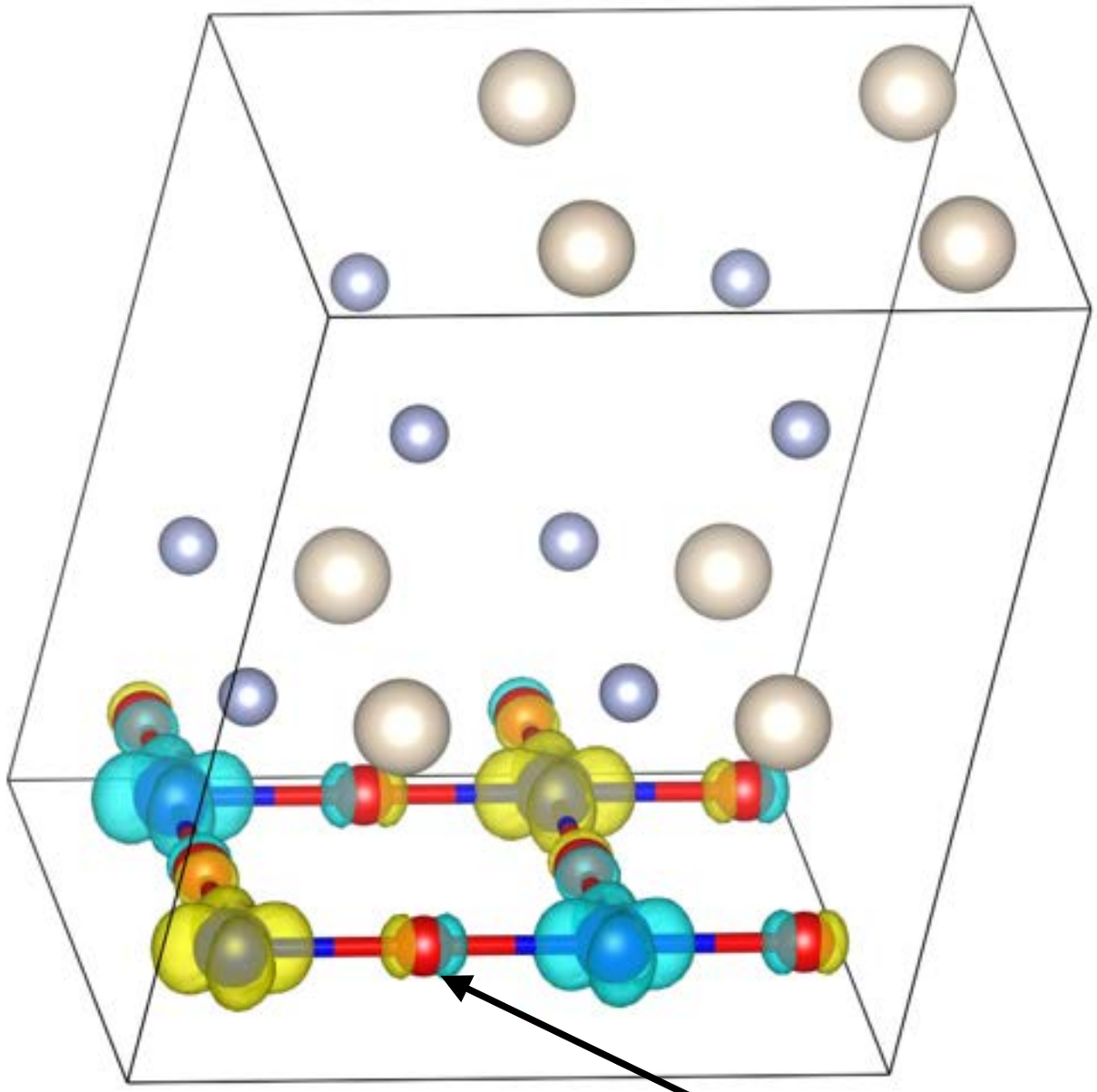


AFM (S_z=0)

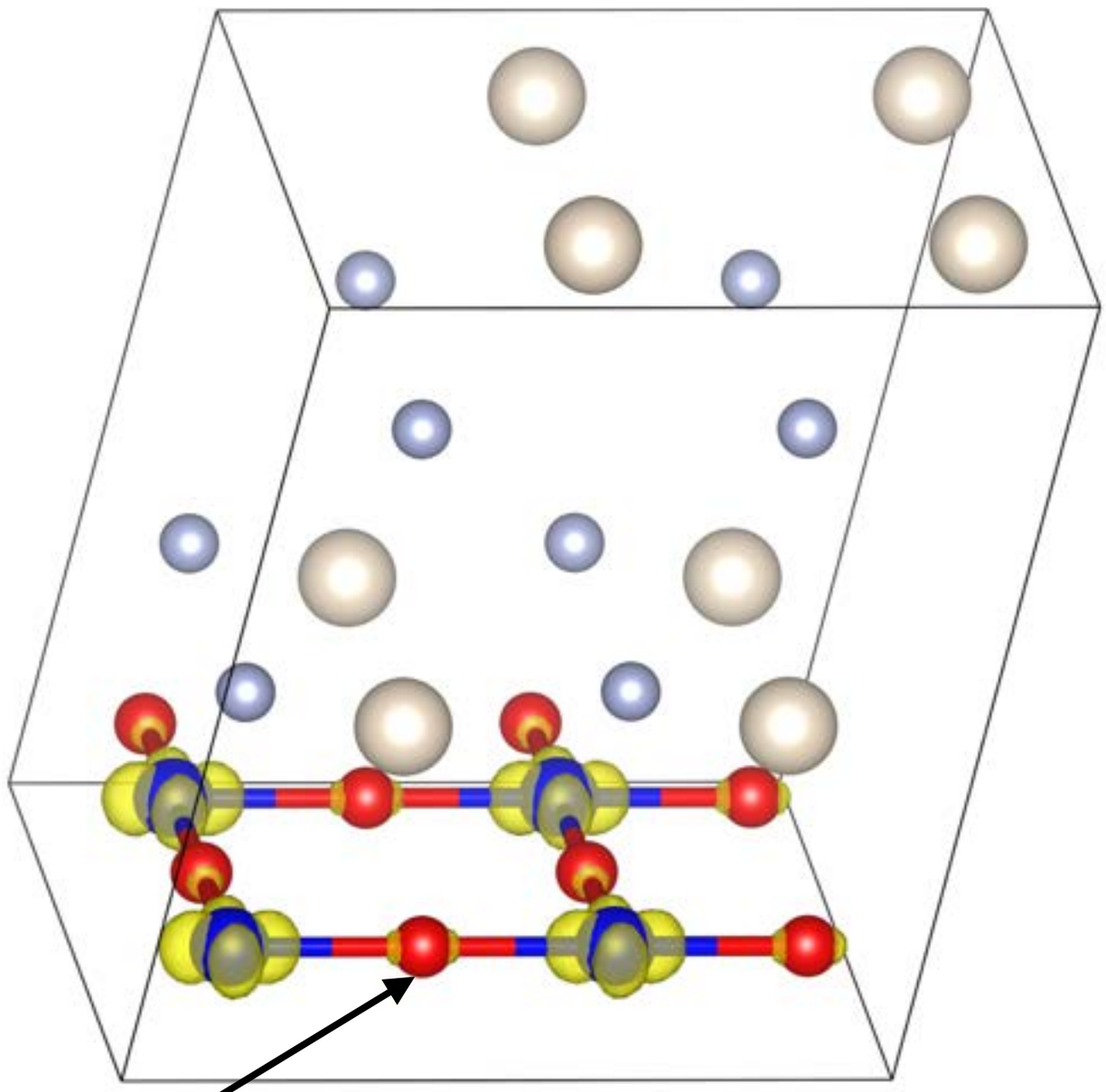


FM (S_z=2)

FN-DMC: magnetic properties



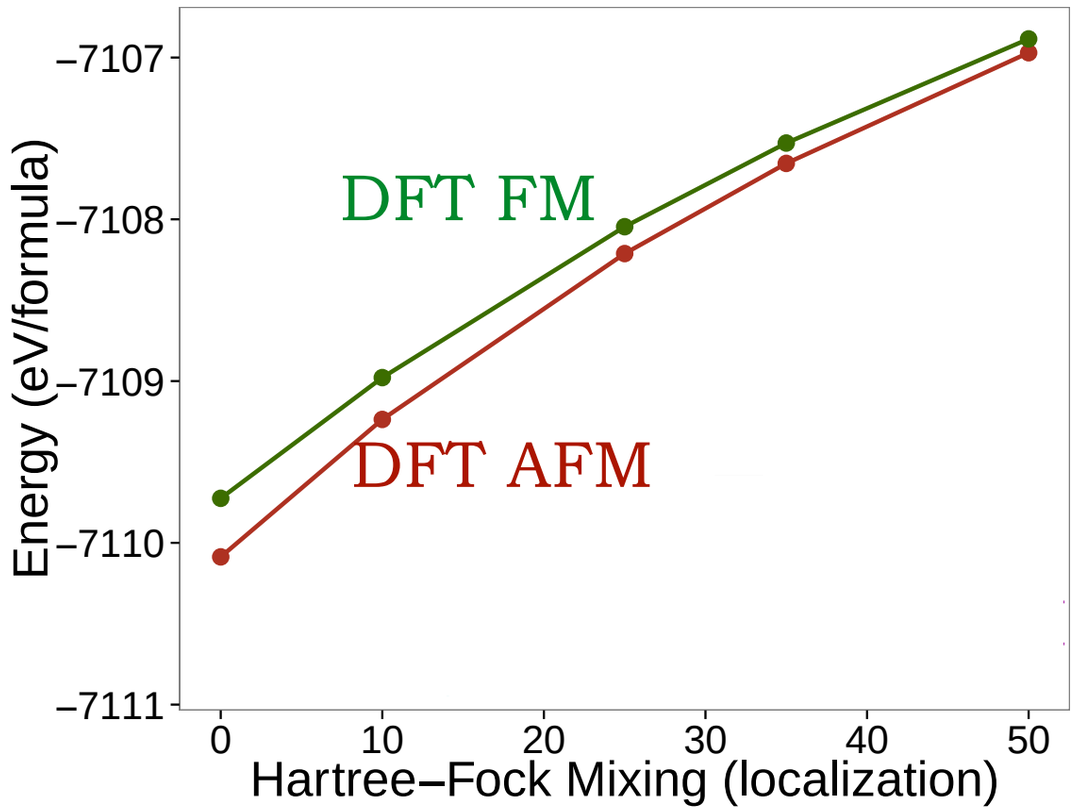
AFM (S_z=0)



FM (S_z=2)

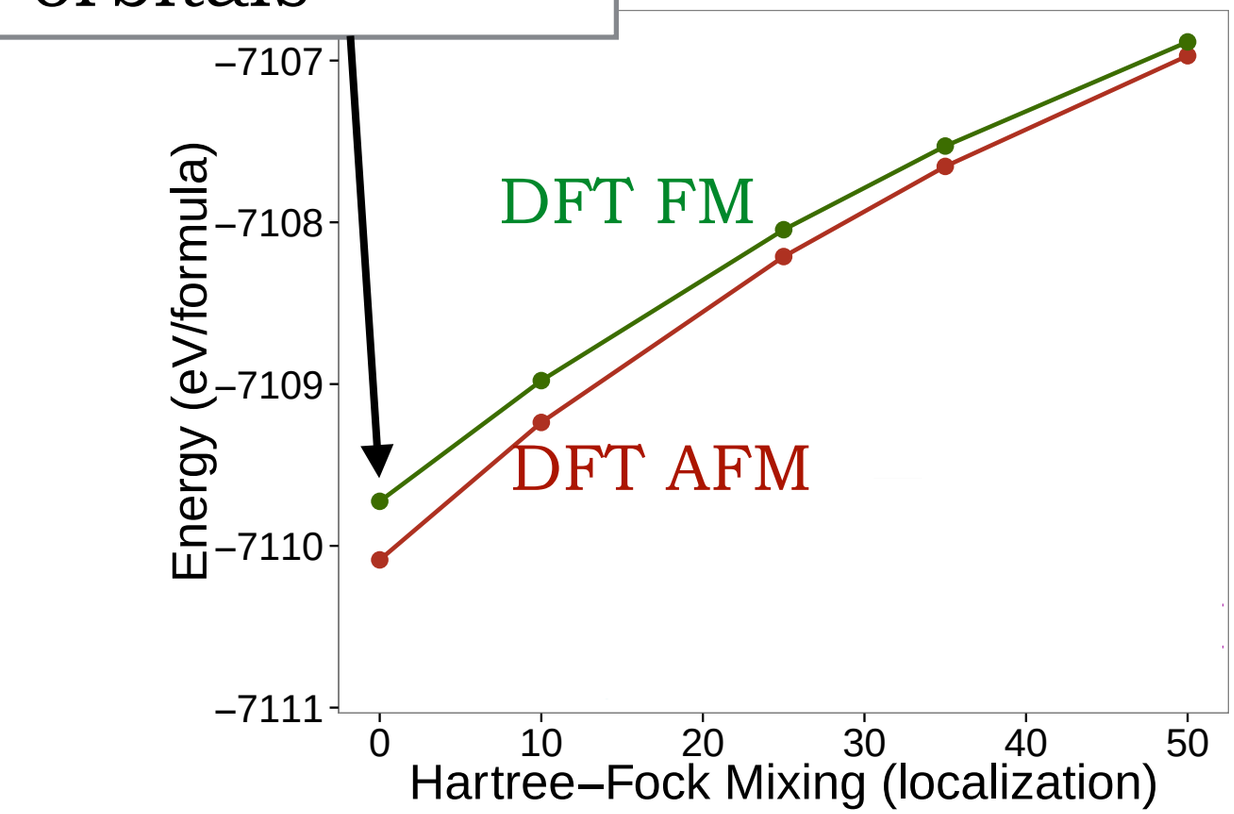
Hybridization

How energies change with nodes: $Ca_2CuO_2Cl_2$



How energies change with nodes: $Ca_2CuO_2Cl_2$

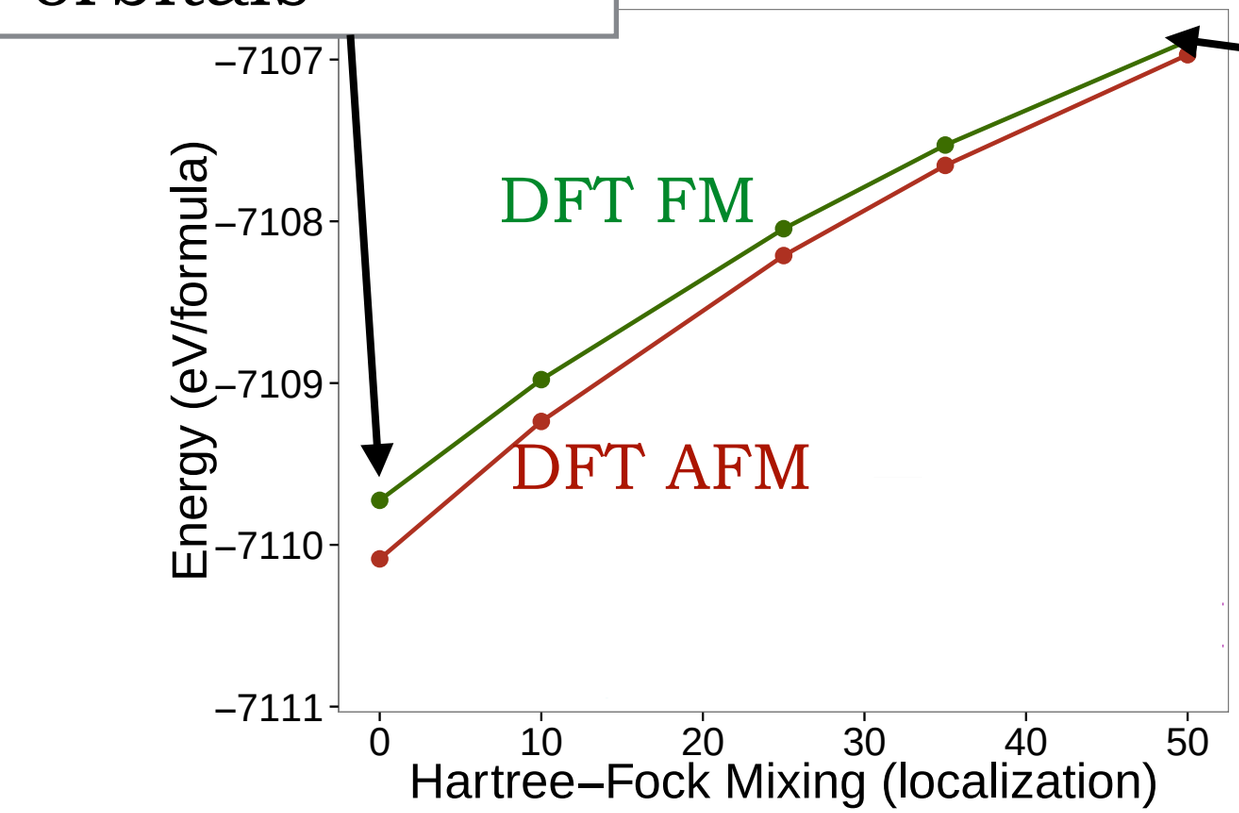
Very hybridized orbitals



How energies change with nodes: $Ca_2CuO_2Cl_2$

Very hybridized orbitals

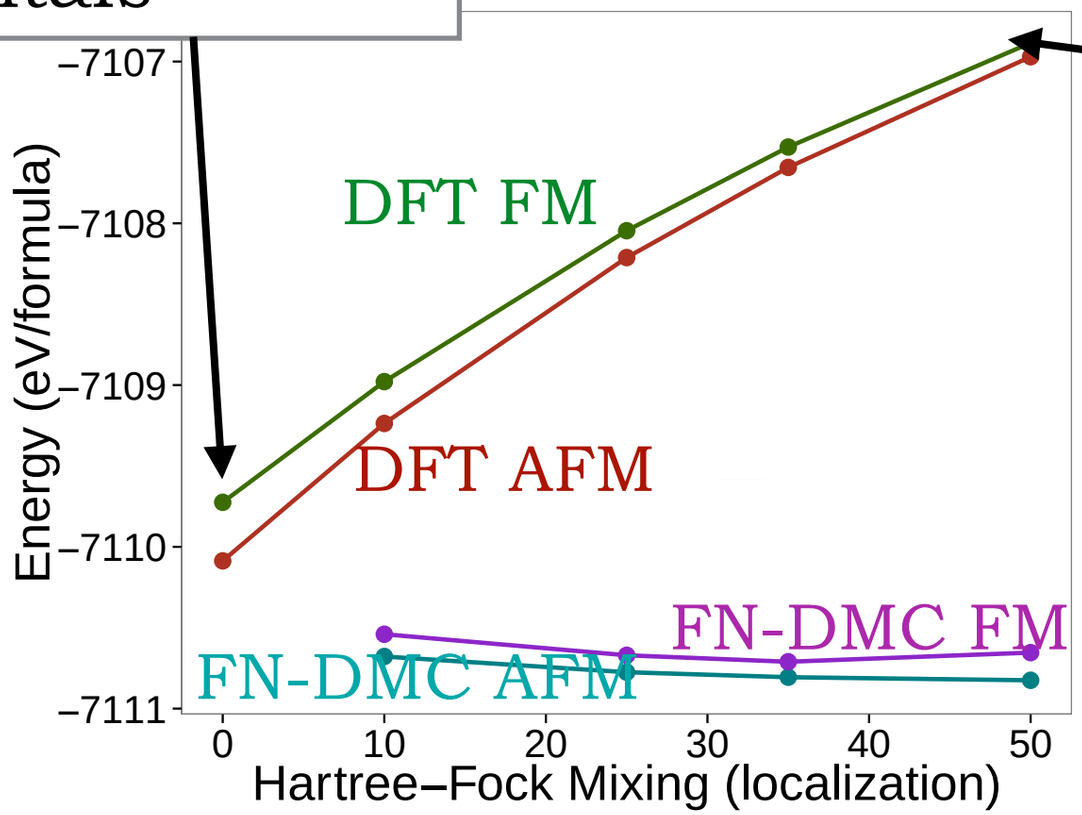
Very localized orbitals



How energies change with nodes: $Ca_2CuO_2Cl_2$

Very hybridized orbitals

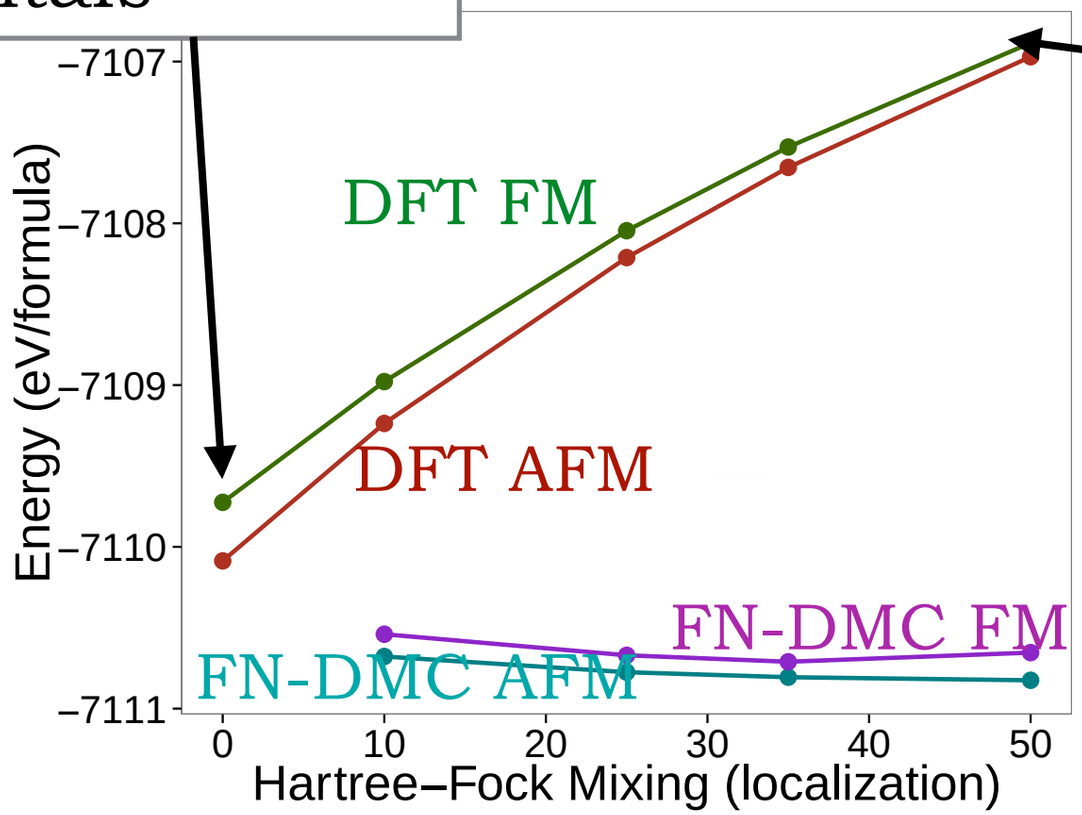
Very localized orbitals

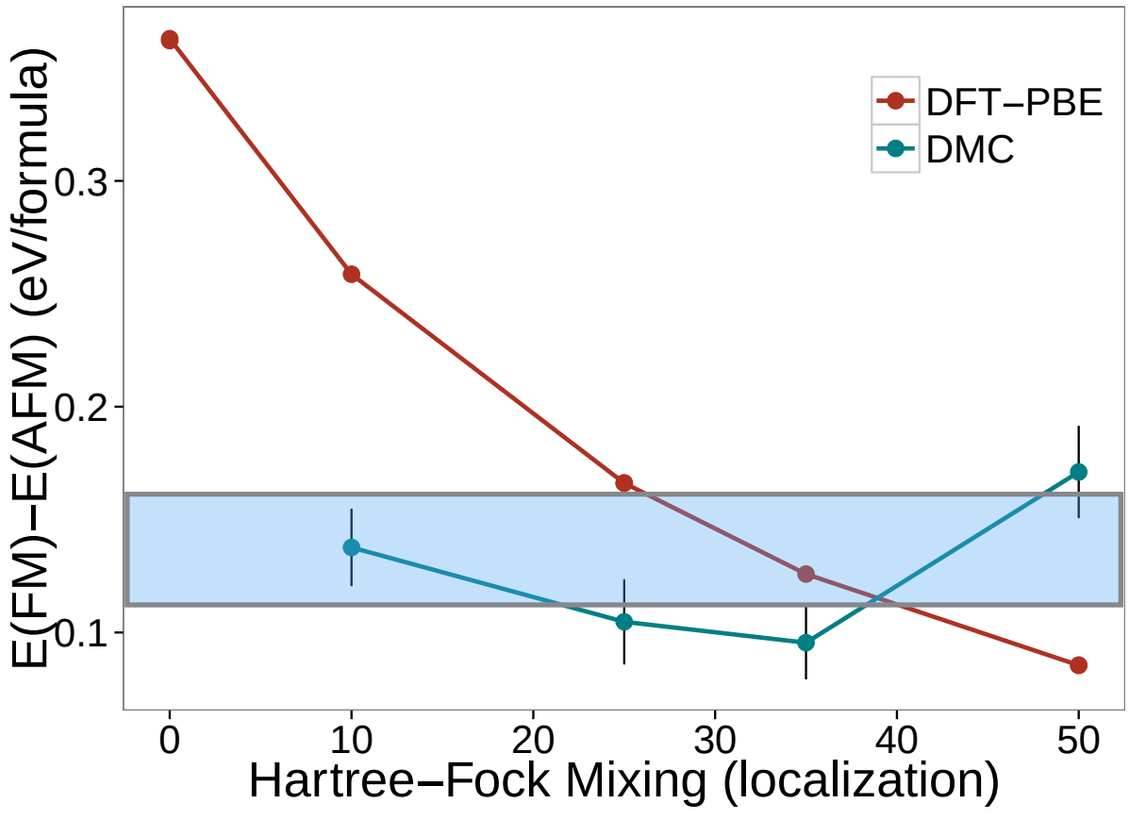


How energies change with nodes: $Ca_2CuO_2Cl_2$

Very hybridized orbitals

Very localized orbitals

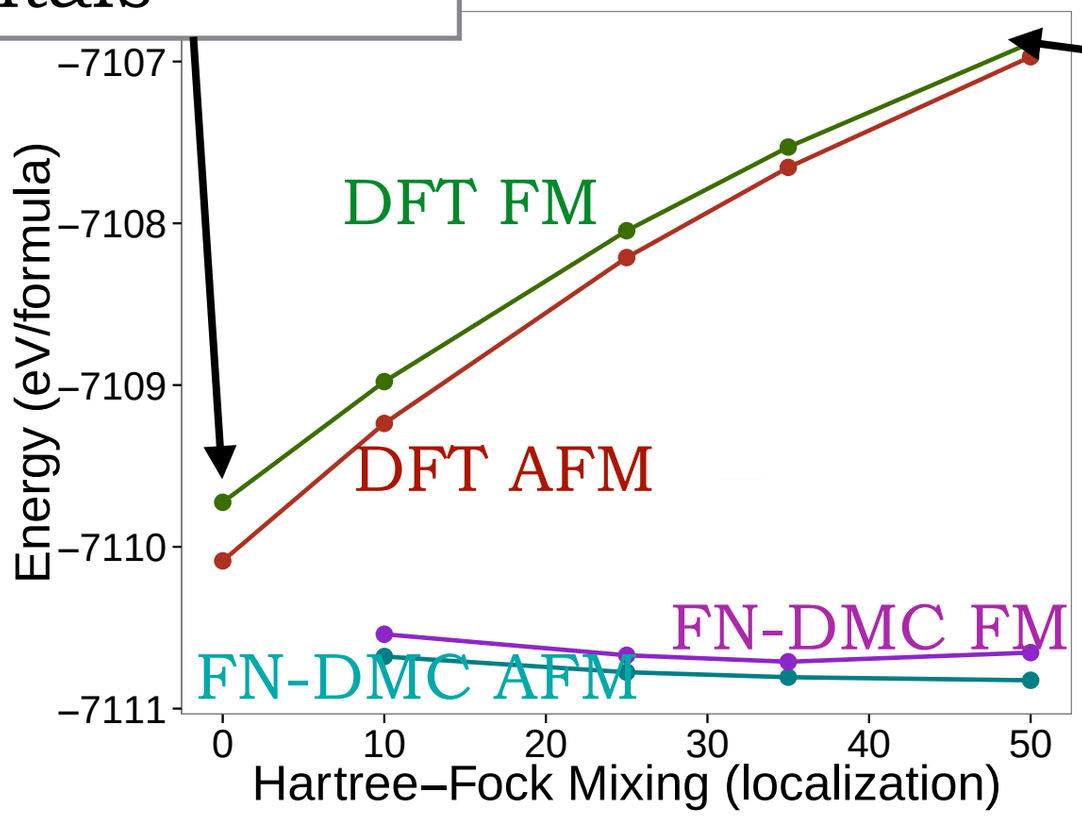


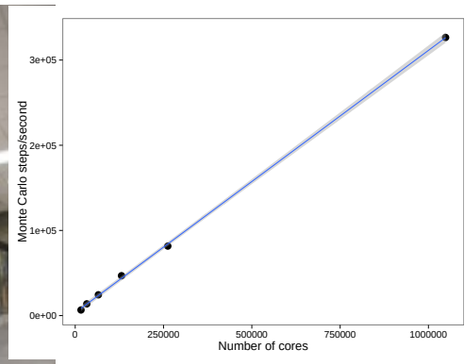


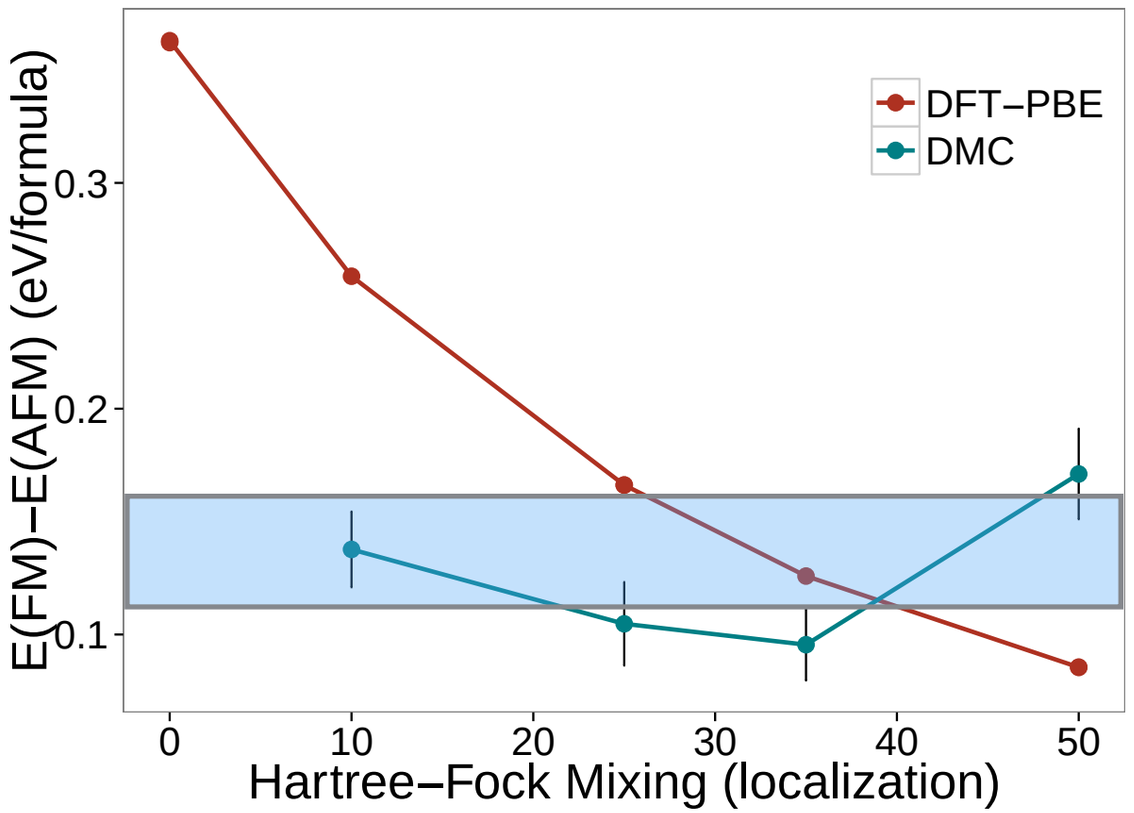
How energies change with nodes: $Ca_2CuO_2Cl_2$

Very hybridized orbitals

Very localized orbitals

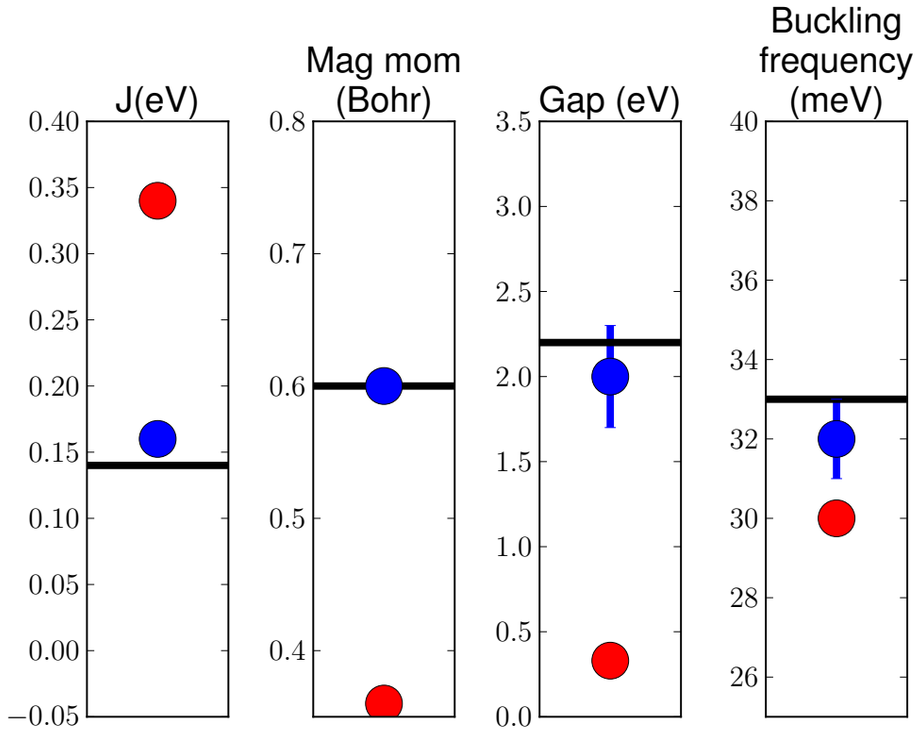




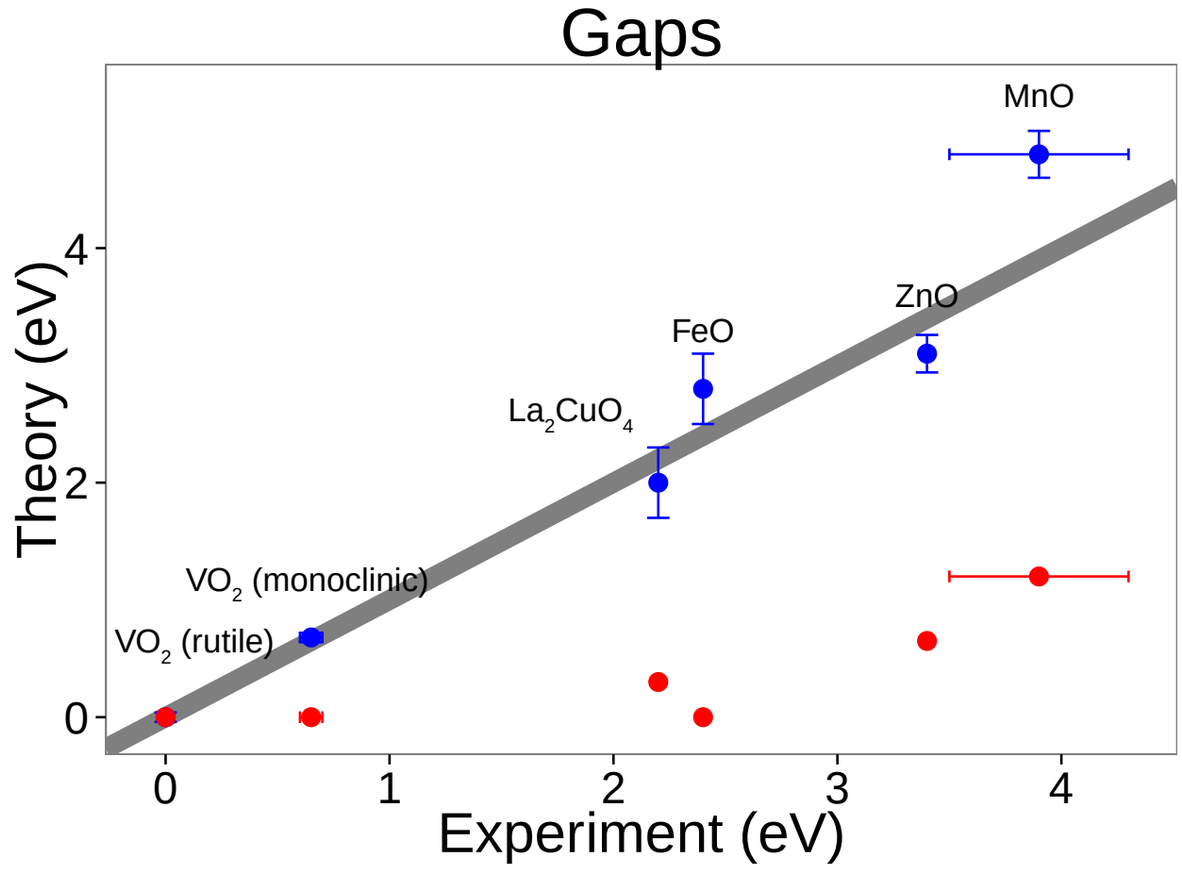


INCITE, 1 million cores

High accuracy on challenging systems from FN-DMC



Experiment
FN-DMC
DFT(PBE)

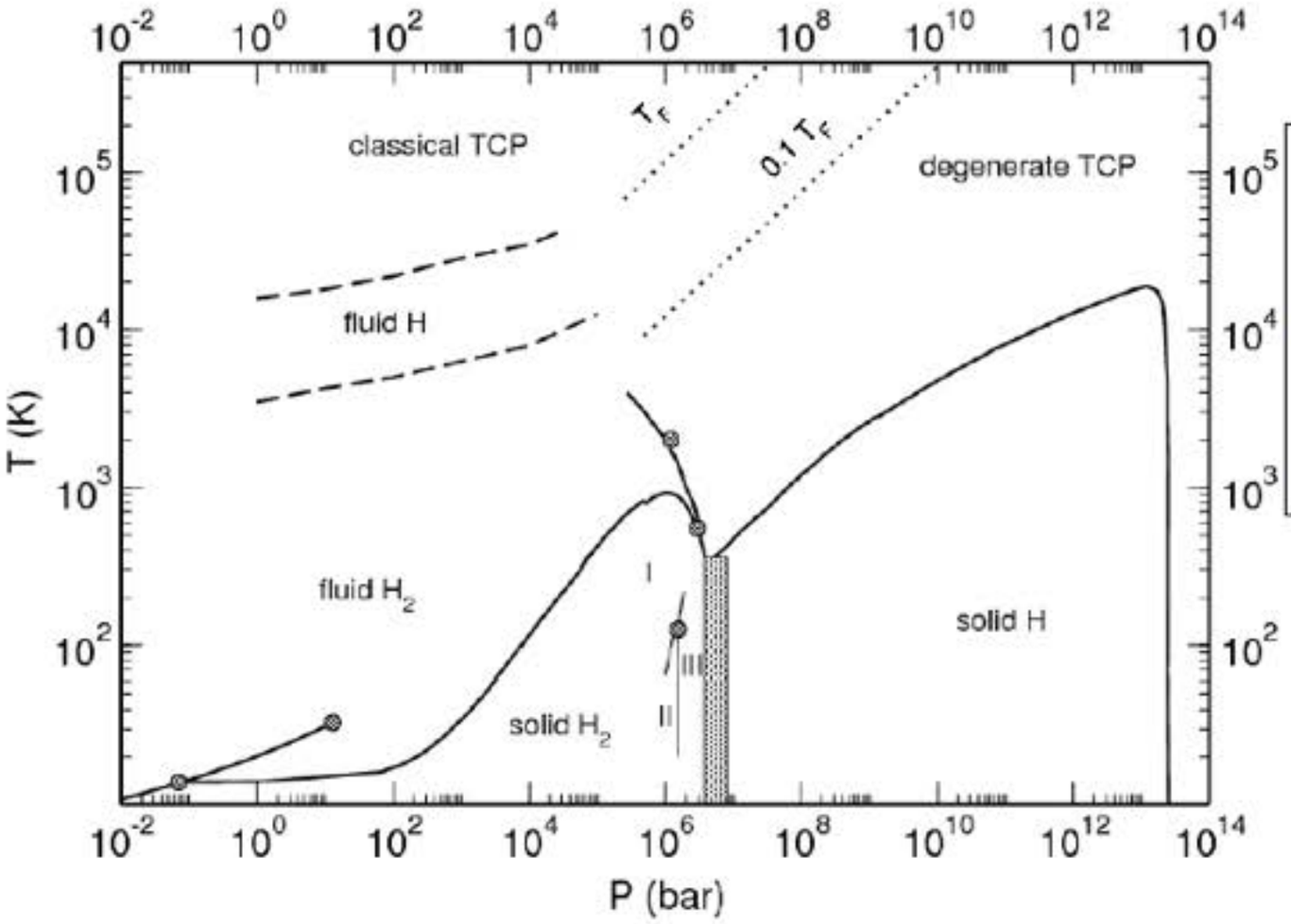


Accurate results on the cuprates and other challenging materials

What functional to use for dense hydrogen?

D. Ceperley *University of Illinois*

Benchmark various DFT functionals on data sets of realistic 54/96 hydrogen atom systems using accurate QMC energies and forces.



INCITE award for computer time

WHO DID THE WORK?

Miguel Morales *Livermore*

Ray Clay *U of Illinois*

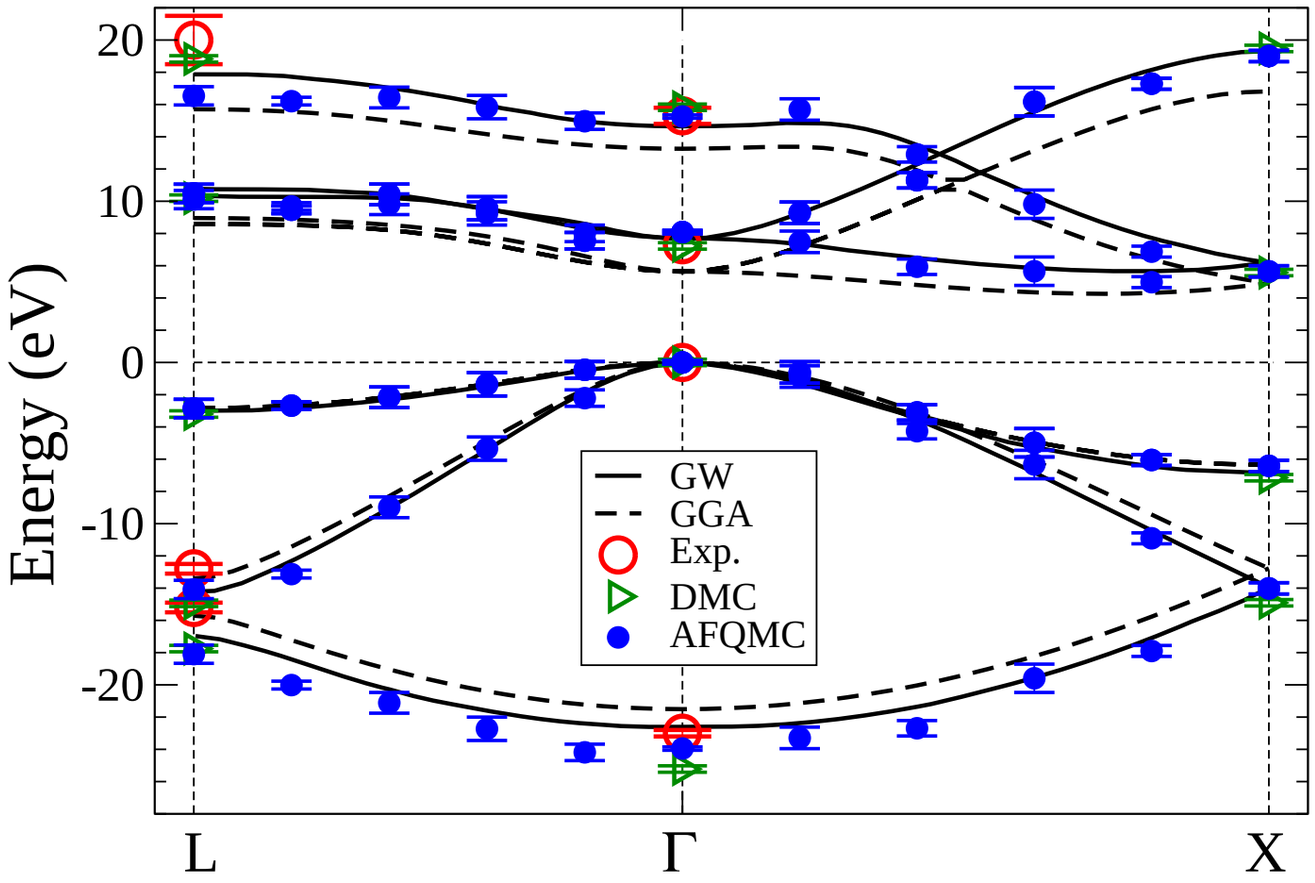
Phys. Rev. B **89**, 184106 (2014)

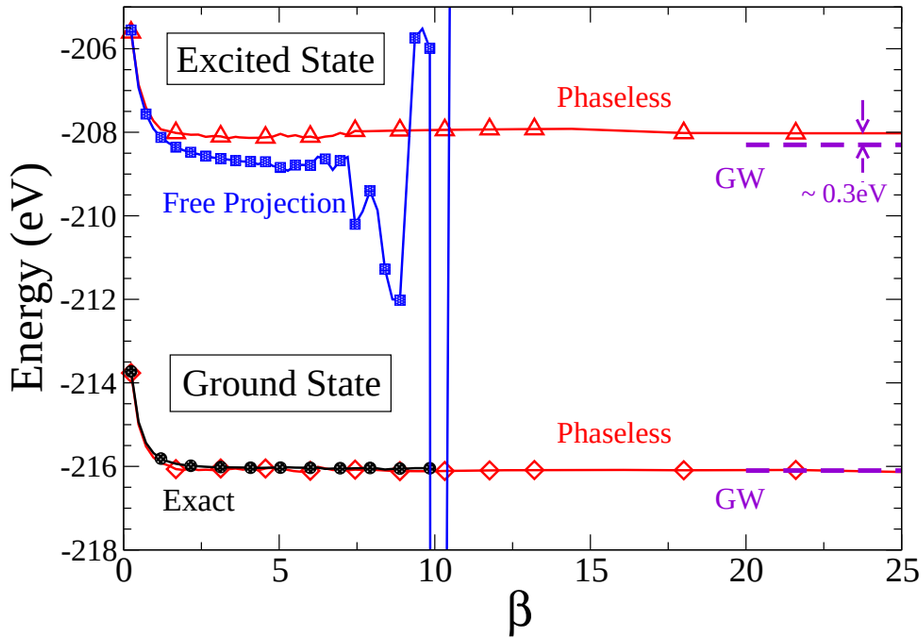
AF-QMC approach for excited states

AFQMC with virtual orbital orthogonalization

- Ma, SZ, Krakauer: NJP 2013

Band structure of diamond





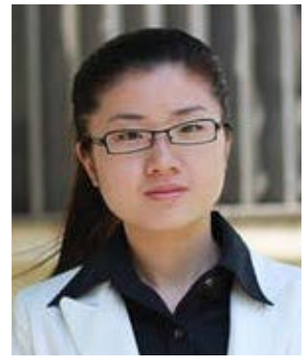
Band gap in wurtzite ZnO

	GGA	Hybrid	*GW*	AFQMC	Exp.
Δ_{gap}	0.77	3.32,2.90	3.4,3.6,2.56	3.26(16)	3.30,3.44,3.57

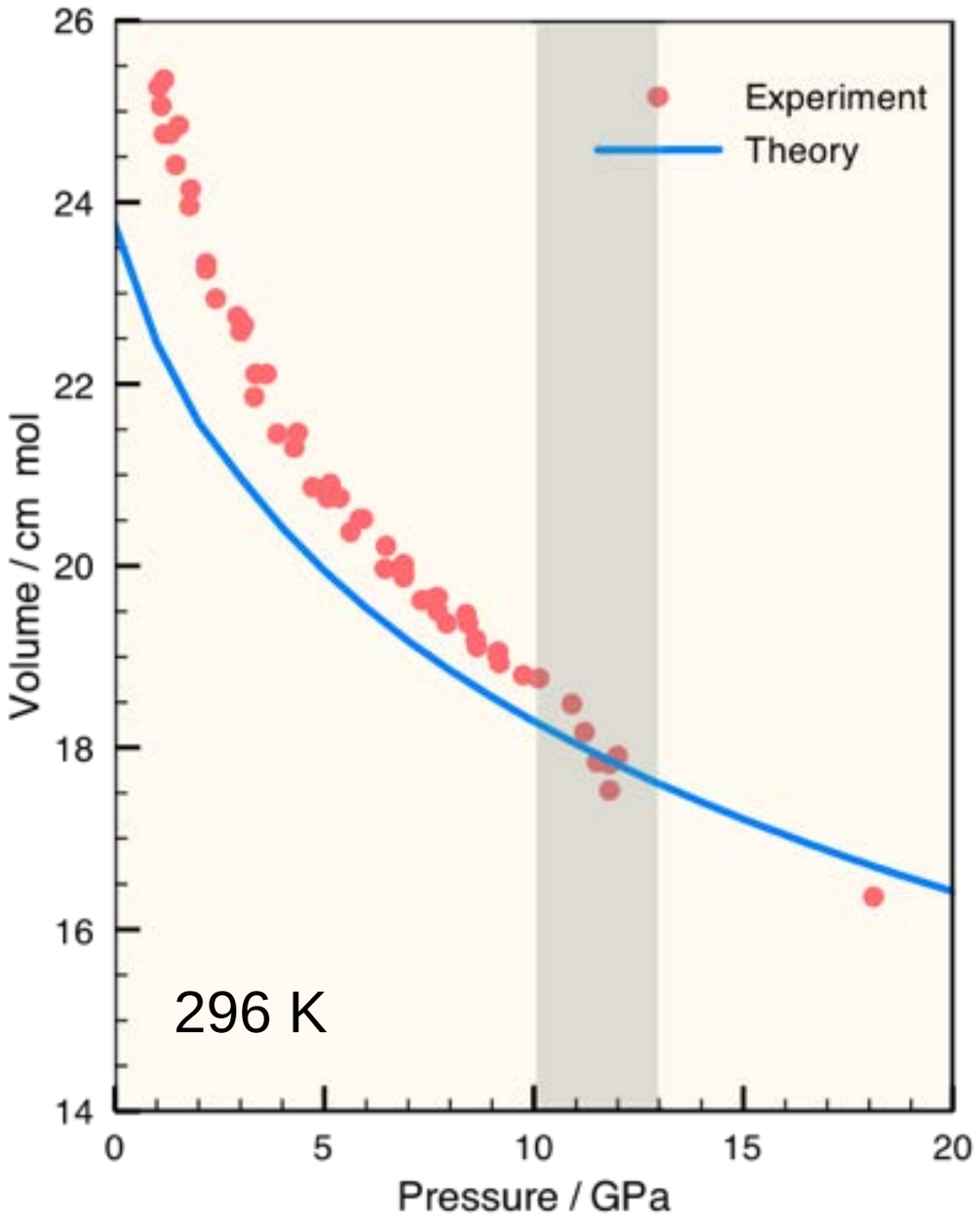
MP2: Phase transition in solid CO_2

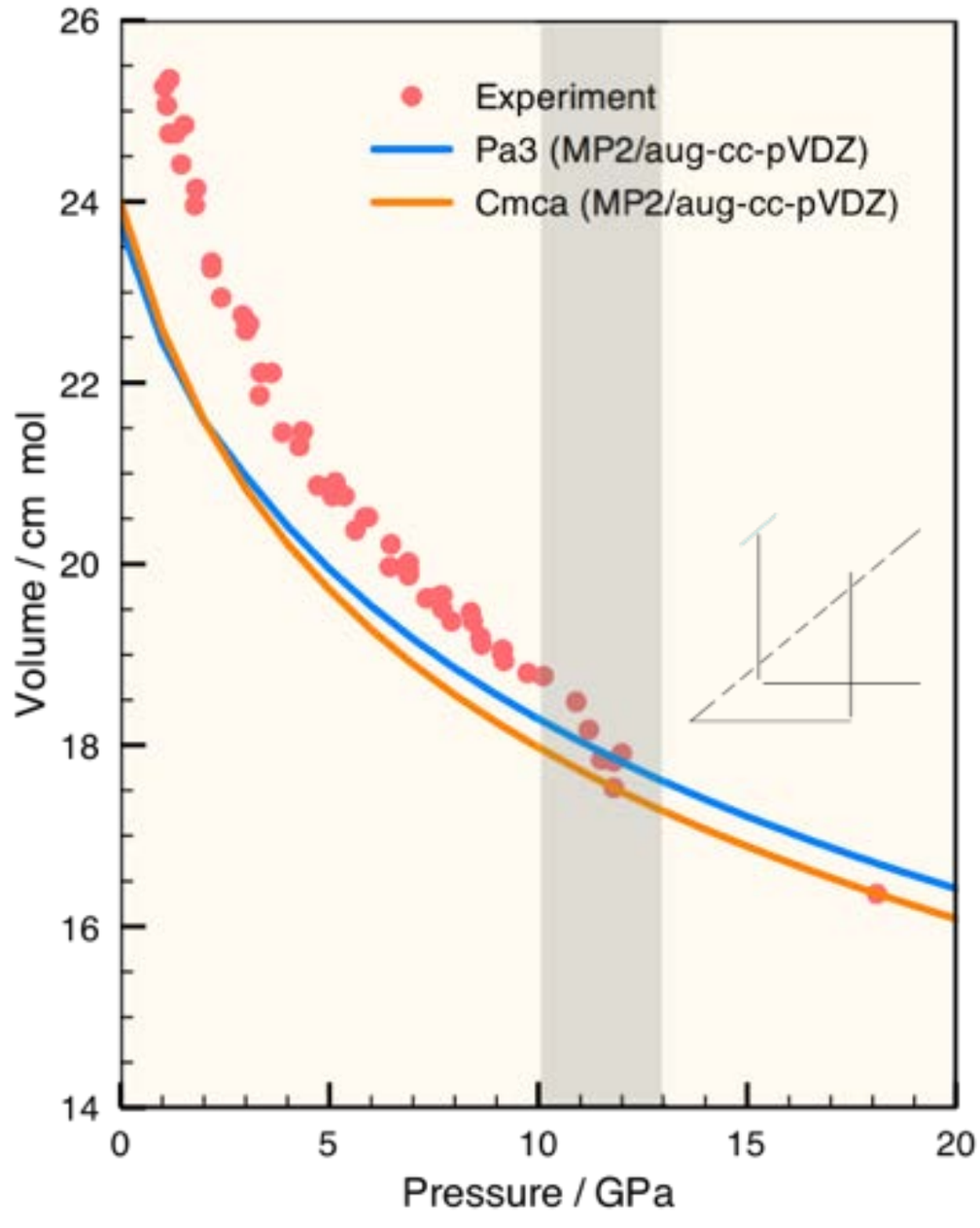
Sode, Keceli, Yagi, and Hirata, *JCP* (2013);

Li, Sode, Voth and Hirata, *Nature Comm* (2013)



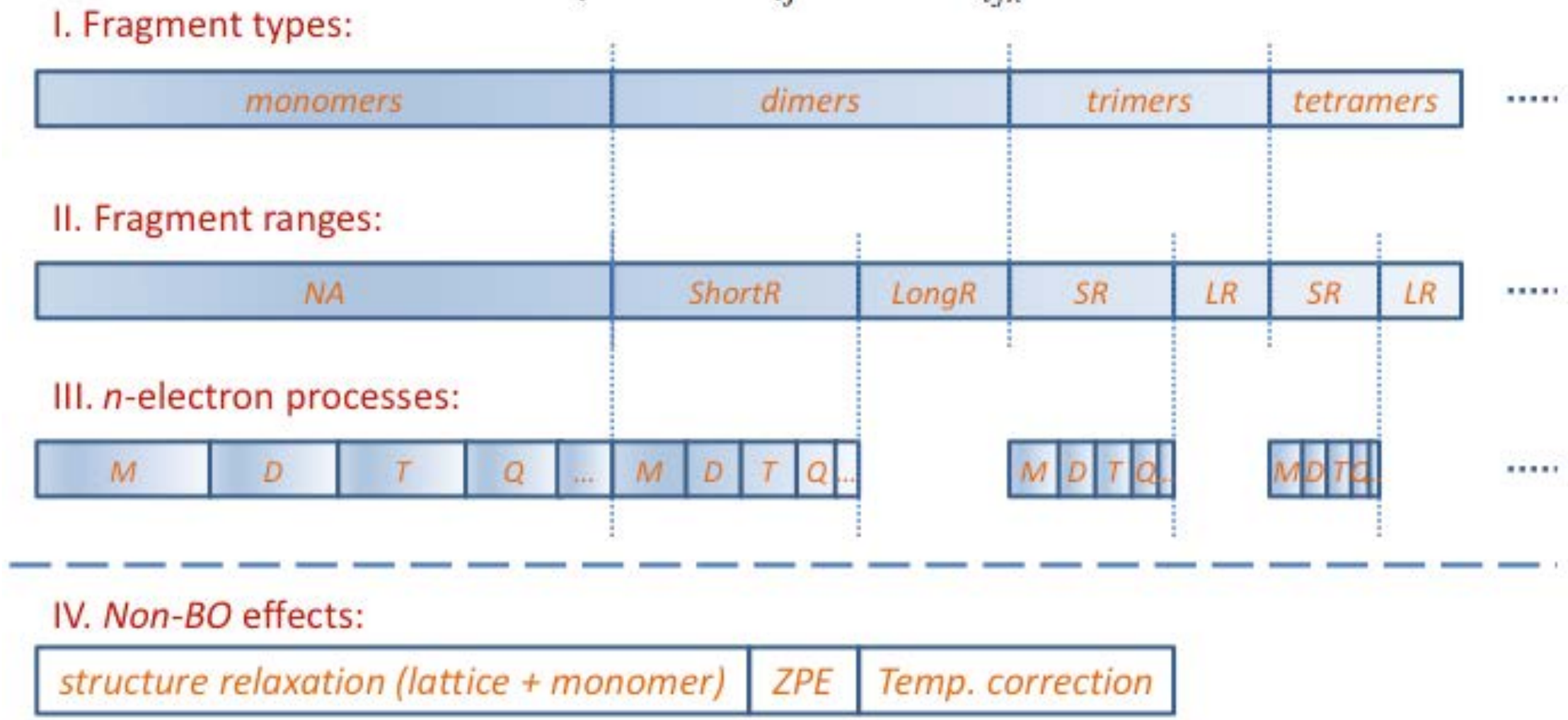
Jinjin Li





Multi-scale: nailing down the benzene crystal

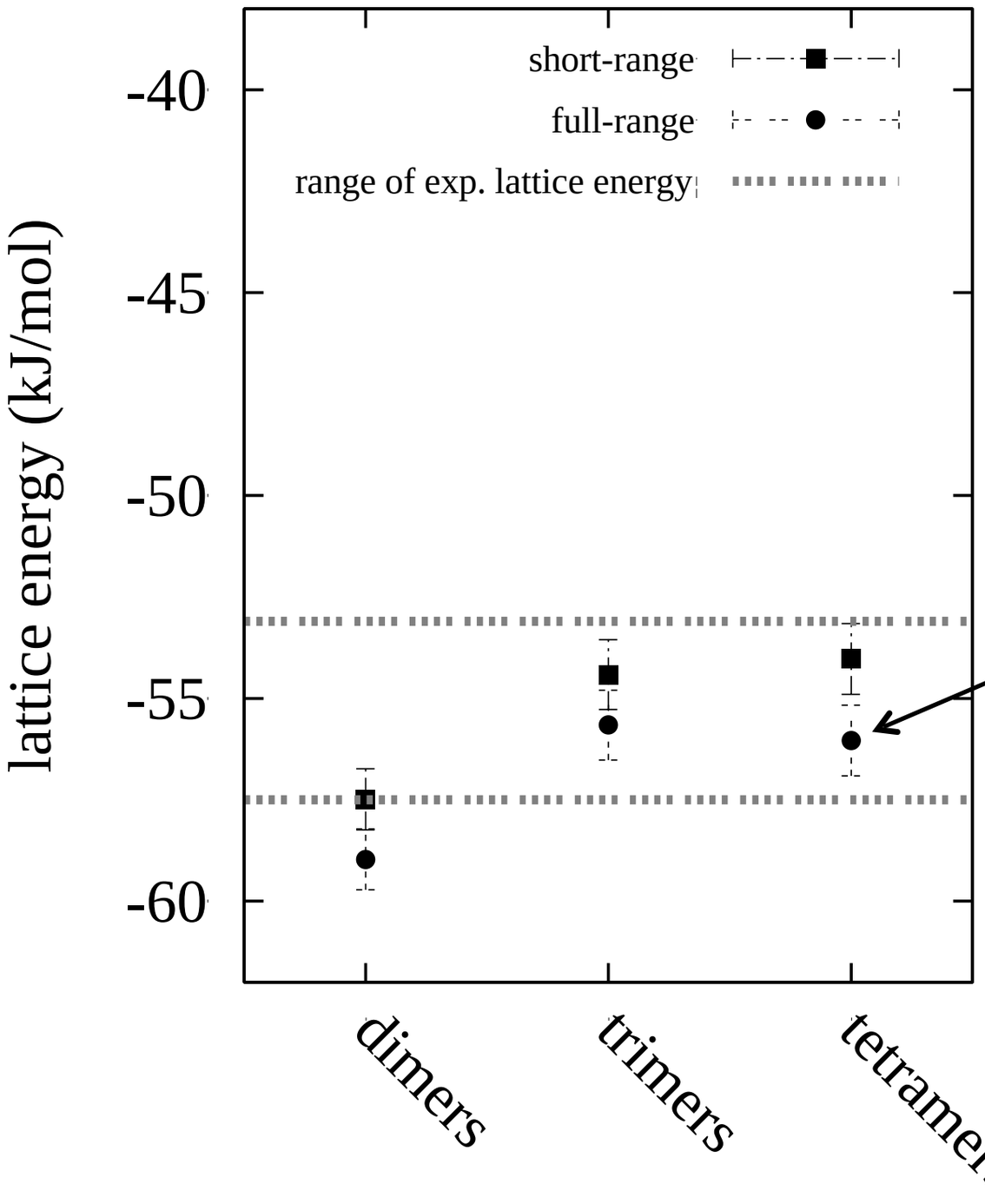
$$E_{\text{tot}} = \sum_i \Delta E_i + \sum_{ij} \Delta E_{ij} + \sum_{ijk} \Delta E_{ijk} + \ldots$$



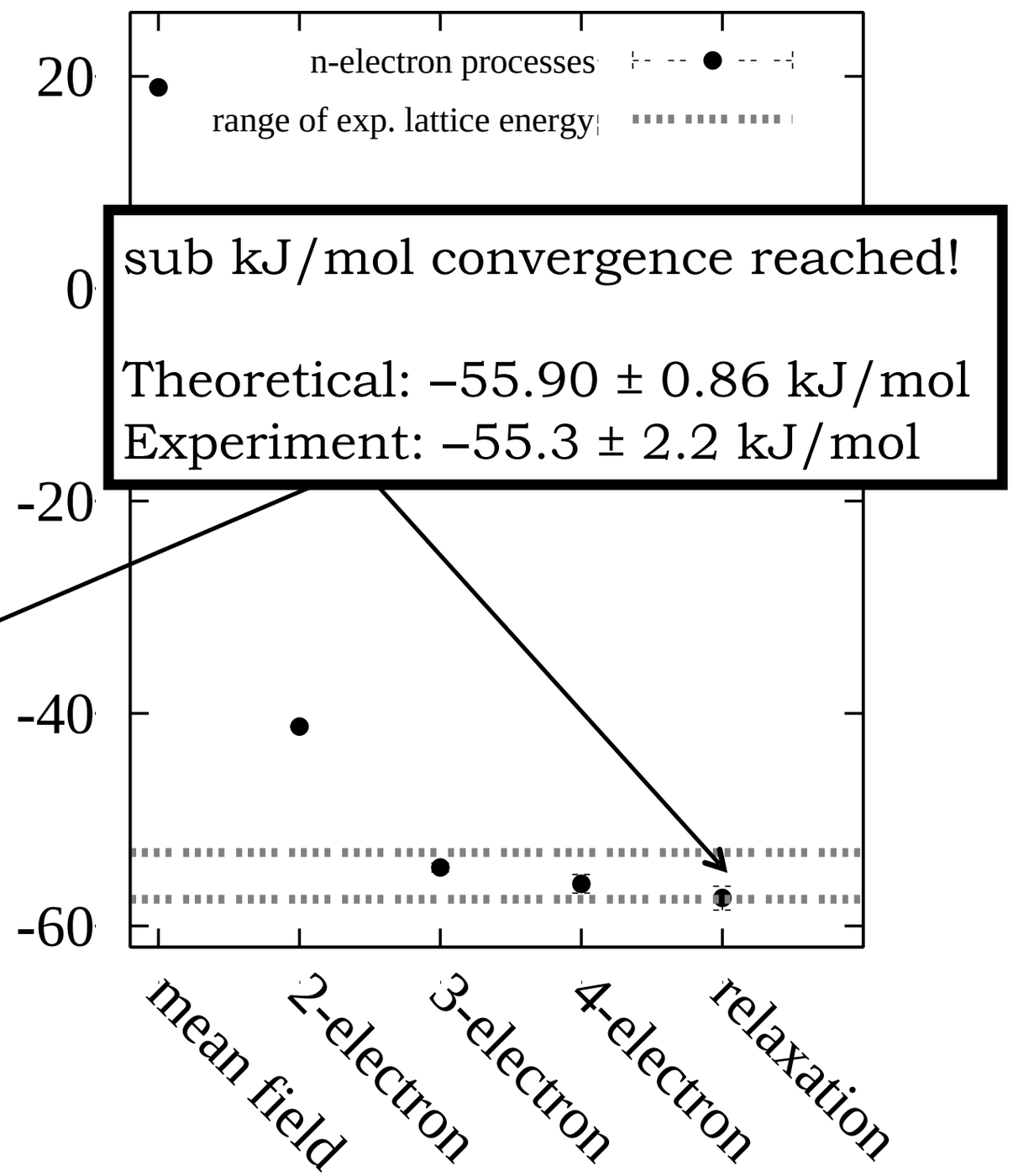
Computations made possible by methods (e.g. local CC) developed with DOE support

Nailing down the benzene crystal

Fragment types and ranges



Correlation levels



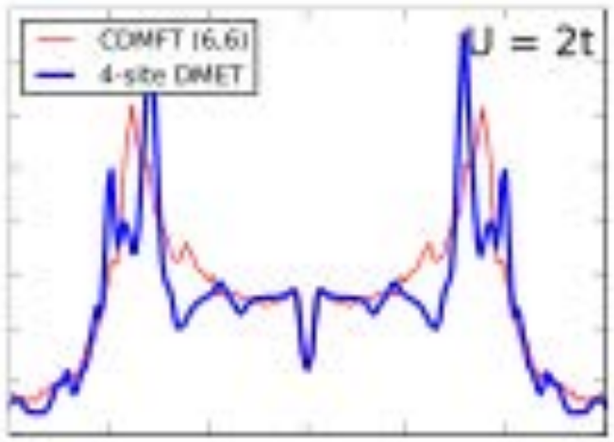
Jun Yang, GKC et al., Science, in press (2014)

New extensions to methods

Obtaining new quantities or higher accuracy than before.

Connecting two methods that were previously disparate.

Highlights in DMET



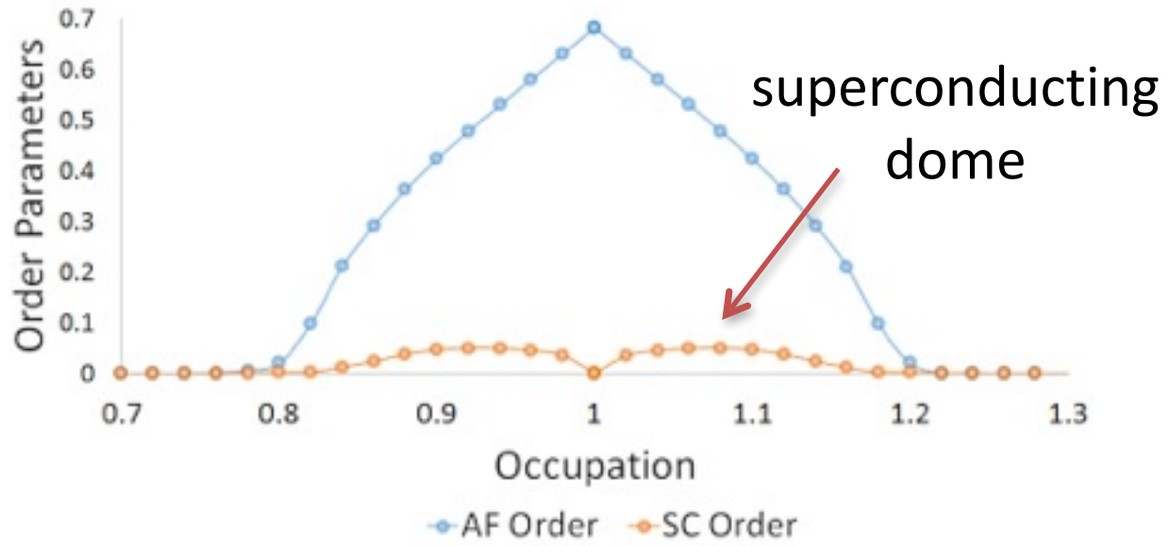
spectral functions from DMET
(a "better and cheaper" DMFT)

Booth, GKC, submitted to Phys. Rev. Lett.

---

phase diagram of 2D Hubbard model
(resembles phase diagram of cuprates)

Zheng, GKC, in preparation

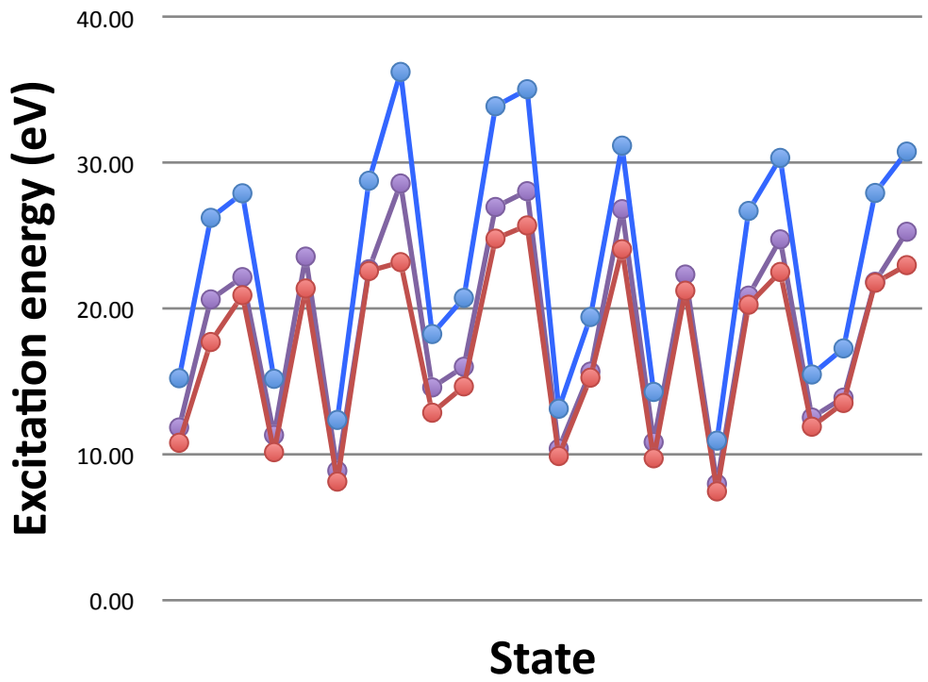


In addition

Intermediate phases of the honeycomb Hubbard model: Chen, Sharma, Booth, Knizia, GKC, submitted to Phys. Rev. Lett.

Metal-insulator transitions across the transition metal oxides: Chen et al., Millis, GKC in preparation (CMCSN collaboration)

Generating effective models

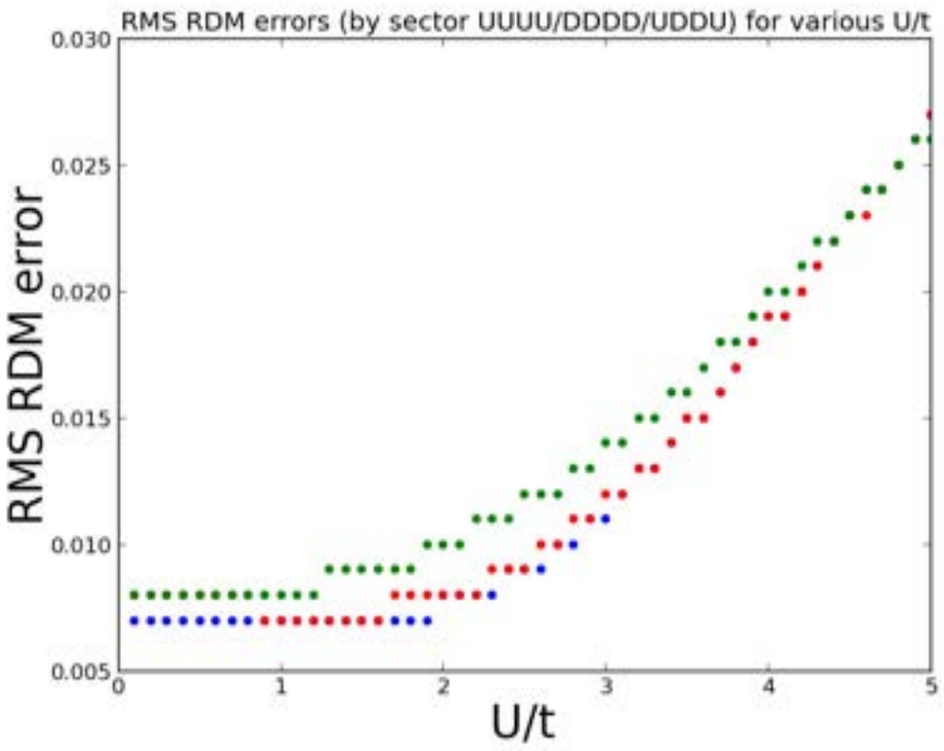


Ab-initio effective Hamiltonians from canonical transformations
works well in molecules,
ready to be applied to solids

Watson, GKC, in preparation

Effective Hamiltonians fitted to FN-DMC
Applied to graphene, small effective U

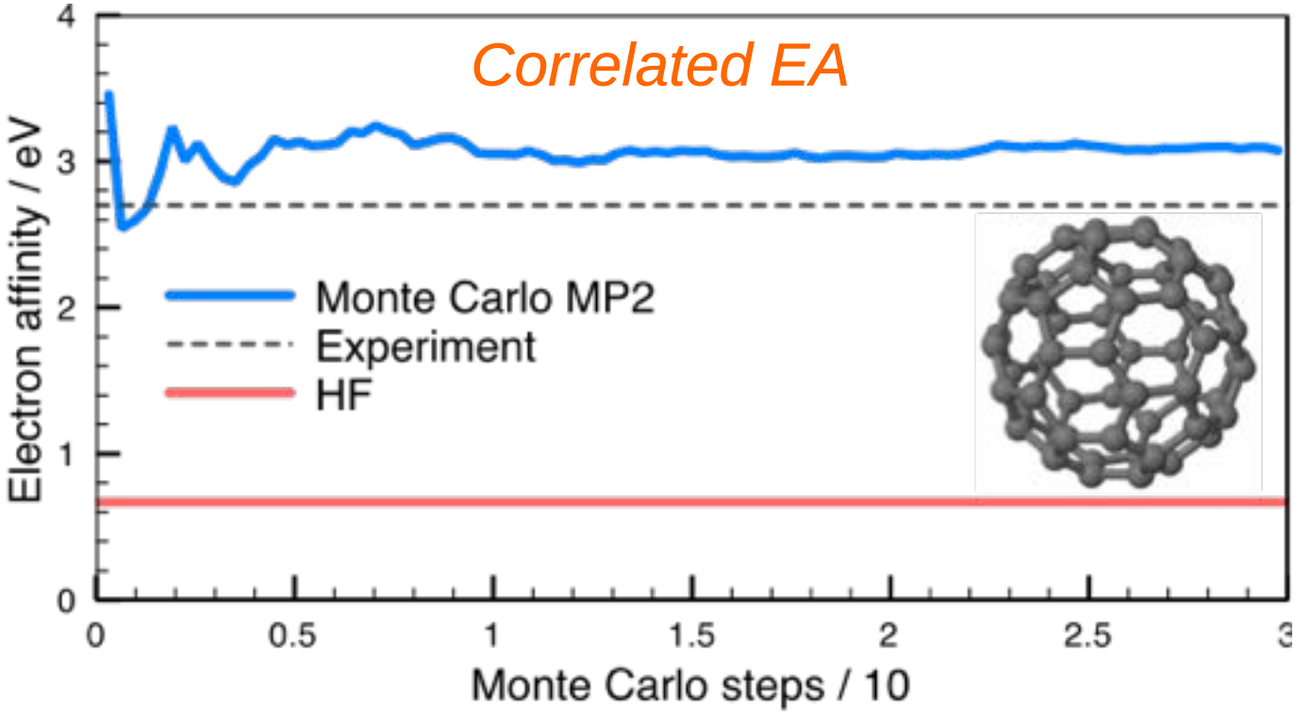
Connects continuum with discrete models
Changlani, Zheng, Wagner



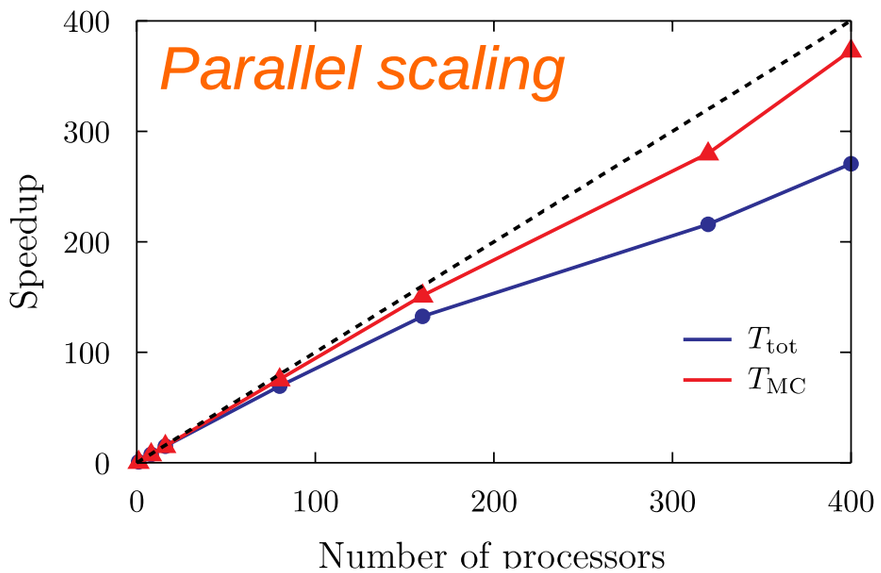
Parallel MC-MP2, MP3, MP2-R12

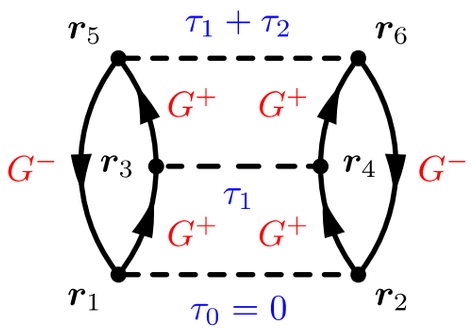
Willow, Hermes, Kim and Hirata, *JCTC* (2013); Willow and Hirata, *JCP* (2014); Willow, Zhang, Valeev, and Hirata, *JCP Comm* (2014)

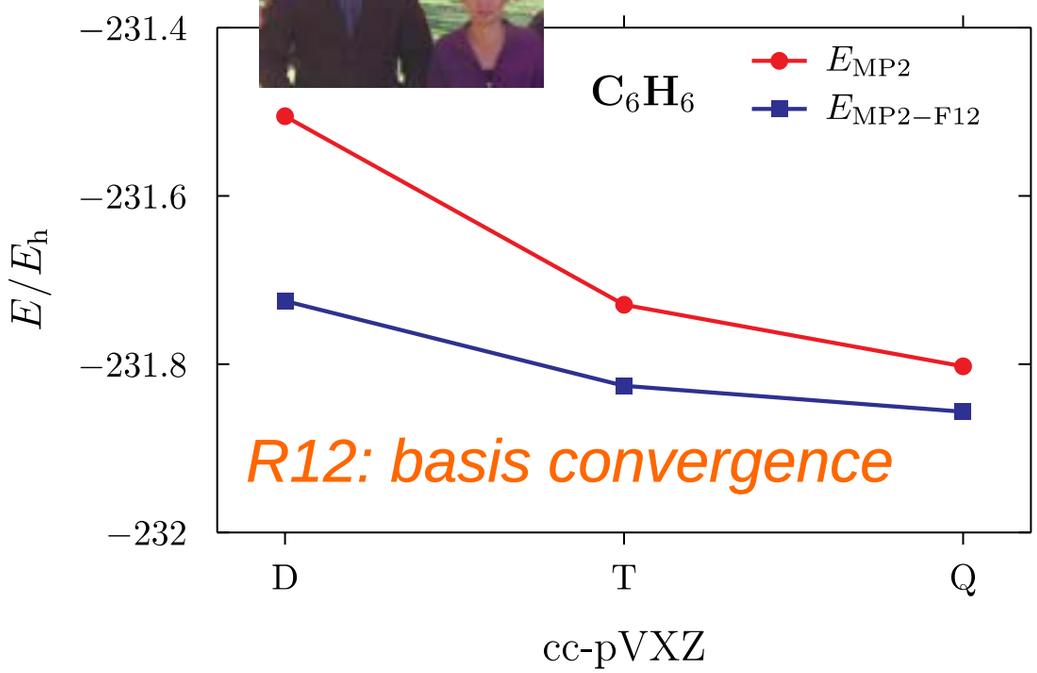
C_{60} cc-pVDZ on 320 processors of Blue Waters



Edward Valeev
Jinmei Zhang







Challenges: data

Many-body wave function

Most compact representation for a complicated wave function. How can we store this systematically for many systems?

Data analysis: high dimensional feature extraction; any takers?

Computational development

- Methods are are computationally demanding (three INCITE awards among our collaboration)

- Monte Carlo methods (example from cuprates):
  one step: ~1 ms
  perform **many millions** of times

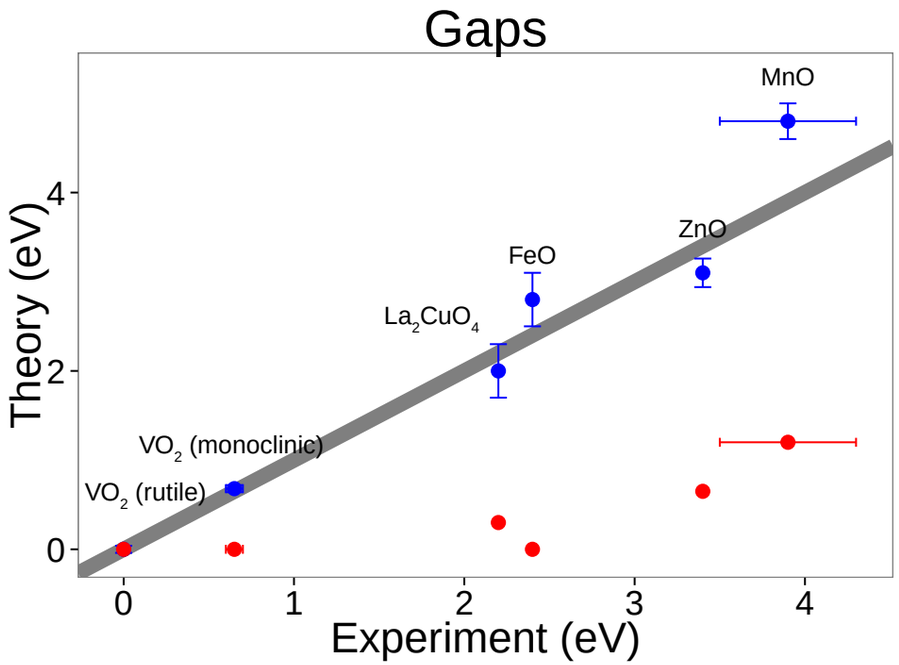
- **TAU** performance tools used:
  2x speedup of QWalk on Mira

Outlook

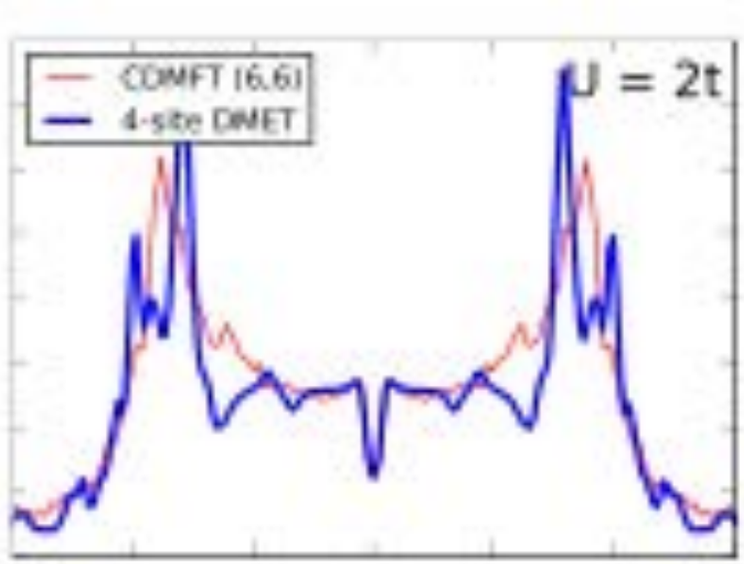
A suite of methods
http://www.predictive-scidac.org

Now partnered with QUEST

Method	Type of Hilbert space	Major approximation	Variational principle	Investigator	Code
FN-DMC	Continuum	Node of wave function	Yes	Ceperley, Wagner	QWalk, QMCPACK
AF-QMC	Discrete	Phase of wave function	Somewhat	Zhang	development
DMRG/ DMET	Discrete	Local matrix product states	Yes	Chan	available
MERA	Discrete	Ansatz	Yes	Ryu	development
MP perturbation theory	Discrete	Perturbation / interaction strength	No	Hirata	development



High accuracy for correlated electron systems



New properties