Predictive computing for condensed matter

2014-2015 report

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Princeton University

SciDAC team:
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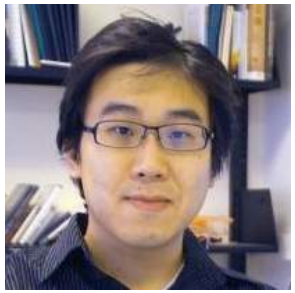
Peter Abbamonte, Bryan Clark, Garnet Chan, David Ceperley, Shinsei Ryu, Shiwei Zhang

Team

Hirata

Wagner

Ceperley



Chan

Clark

Ryu

Zhang

UIUC

Princeton

William and Mary

tame exptalist

Abbamonte

Expertise: quantum chemistry, quantum Monte Carlo, embedding and DMFT, DMRG and tensor networks, continuum field theories

44 publications to date

SciDAC objective: predictive computation

Dirac

The fundamental laws necessary for the mathematical treatment of a large part of physics and **the whole of chemistry** are thus completely known …

the difficulty lies only in the fact that application of these laws leads to equations that are too complex to be solved.

accurately solve **many-particle** Schrödinger equation for solids

$$H\Psi(r_1, \ldots, r_N) = E\Psi(r_1, \ldots, r_N)$$

partial differential equation

N = O(10^{23}) dimensions

hopeless?

Maybe!

[The Schrodinger equation] cannot be solved accurately when the number of particles exceeds about 10. No computer existing, or that will ever exist, can break this barrier because it is a catastrophe of dimension ...

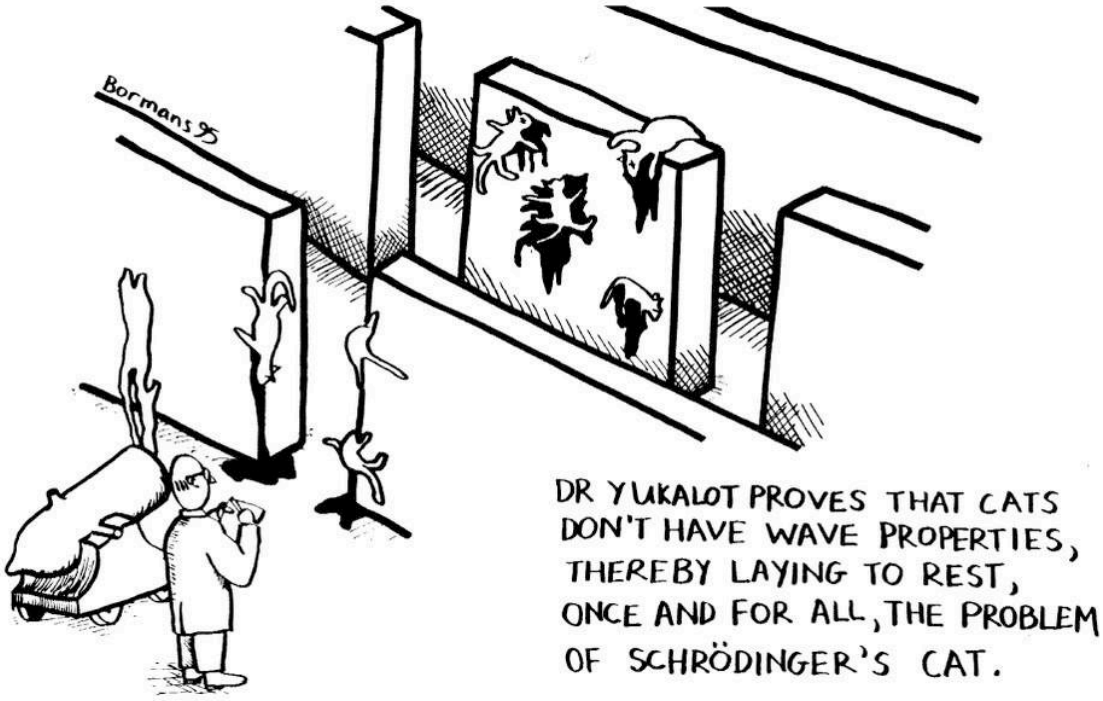
Pines and Laughlin (2000)

in general the many-electron wave function Ψ … for a system of N electrons is not a legitimate scientific concept [for large N]

Kohn (Nobel lecture, 1998)

illusion of complexity

nature does not explore all possibilities



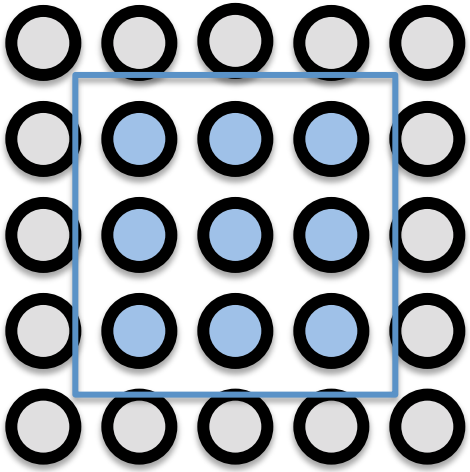
exponential complexity of wavefunctions: not real
if **cleverly** exploit structure of physical reality

intellectual basis of this SciDAC

Examples

Electrons interact only **pairwise**

foundation of pair approximations in QC,
Jastrow factors in quantum Monte Carlo



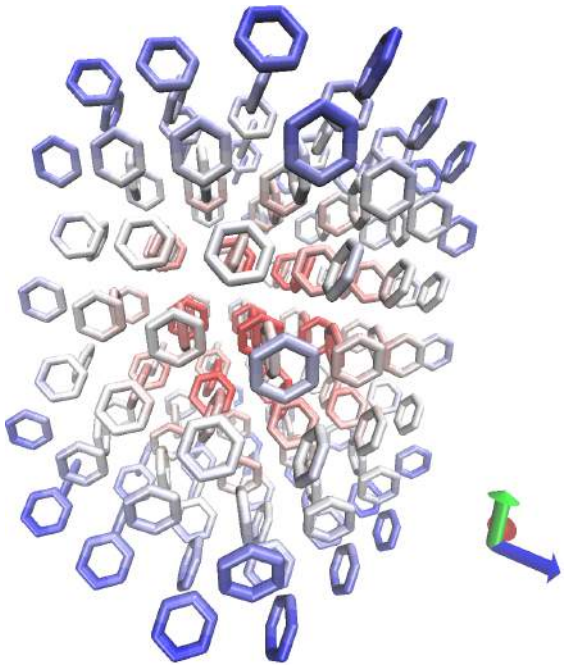
If divide system into inner and outer regions, only degrees of freedom on the boundary are "entangled".
"Area Law" of entanglement.

foundation of DMRG, tensor networks
density matrix embedding theory

SciDAC: predictive computing

simple condensed phase systems: can solve many-particle Schrödinger equation with accuracy rivalling or exceeding experiment

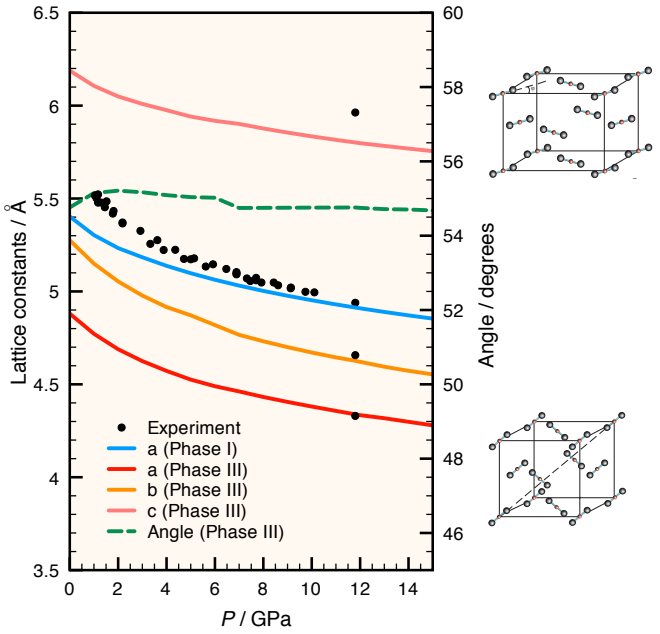
thermodynamics of benzene crystal



theory revises experiment

Chan group, Science (2014)

phase diagram of solid CO_2

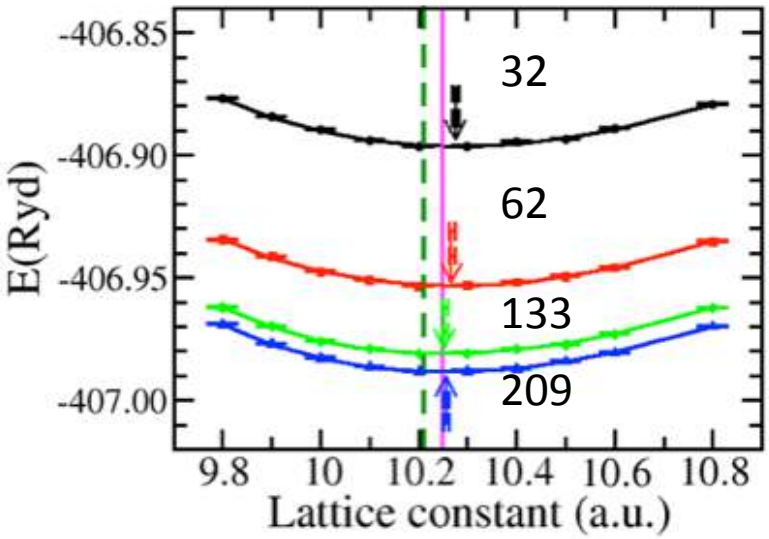


structures involved in
solid-solid transition elucidated by theory

Hirata group, Nature Comm (2013)

SciDAC: predictive computing

Bulk silicon equation of state

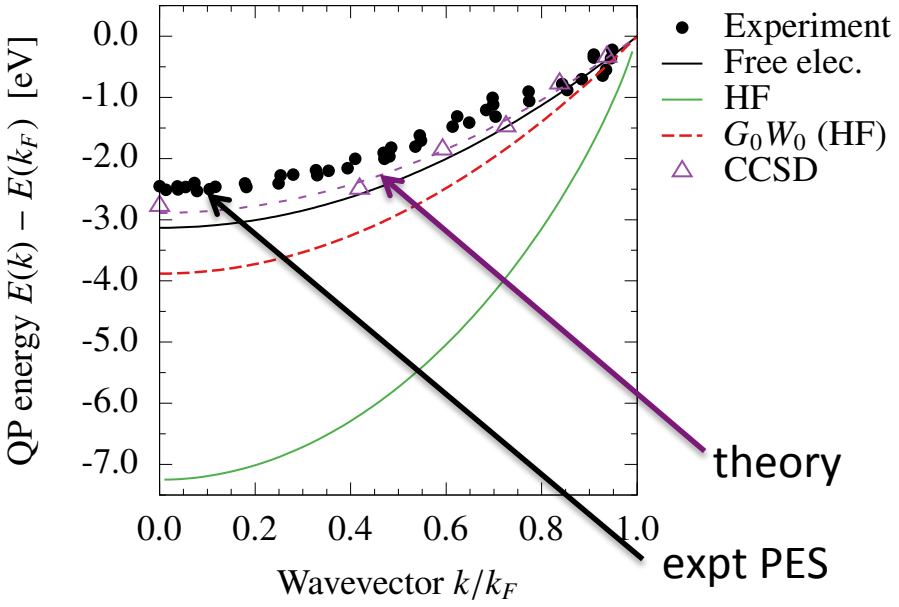


	a (a.u.)	B (Gpa)
expt	10.25	99.2
AFQMC	10.256(6)	97.5(1.8)

auxiliary field quantum Monte Carlo systematically nails Si equation of state

Zhang group, Phys Rev Lett (2015)

Occupied band structure of sodium



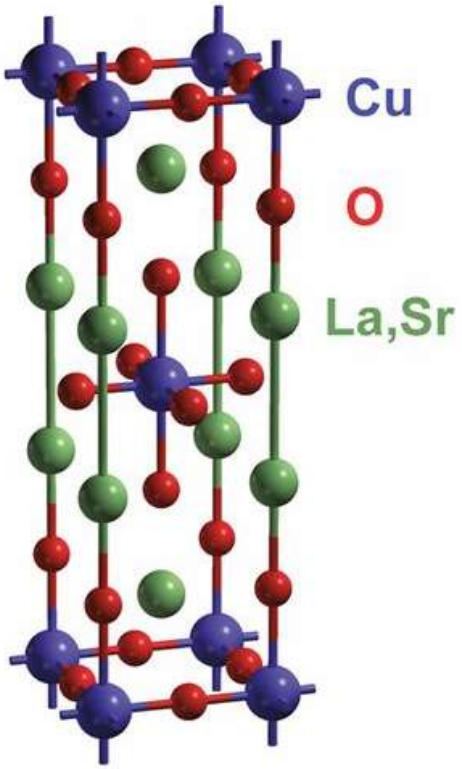
systematic quantum chemistry Green functions yield near-exact solid band-structure

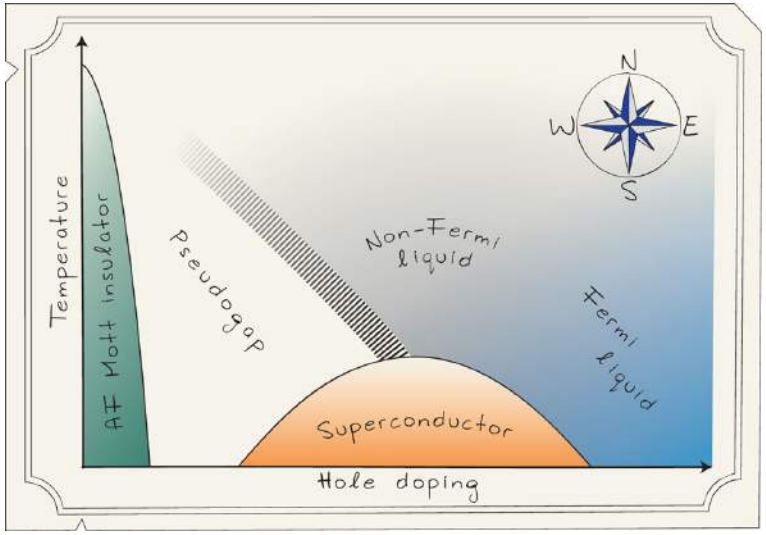
Chan group, in preparation(2015)

SciDAC: predictive computing

complex condensed phase systems:
non-empirical computation of properties becoming a reality

correlated solids, e.g. **high temperature cuprate superconductors**





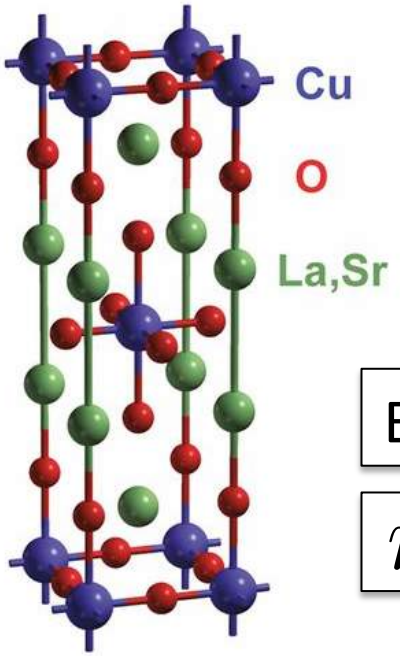
fantasy?

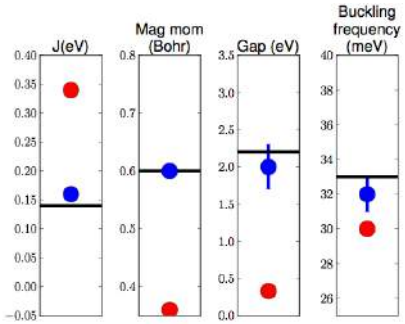
SciDAC: predictive computing

no DFT, no U, no fitting, no empirical parameters

normal (undoped) state of cuprates

La_2CuO_4





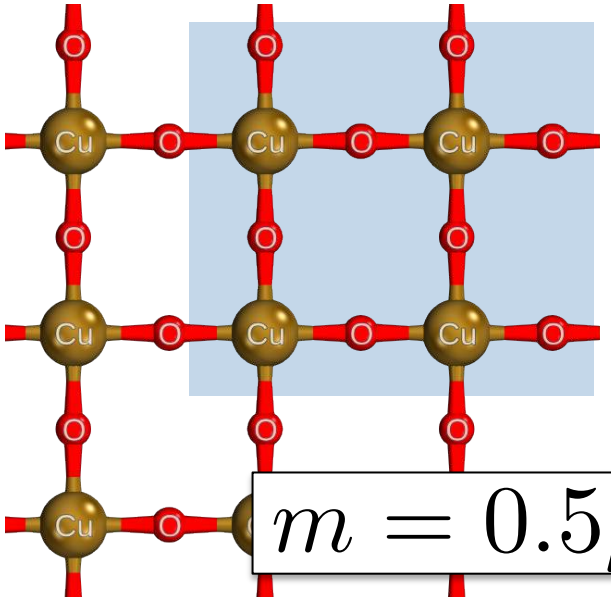
Expt $0.61\mu_B$

$m = 0.68\mu_B$

real-space Diffusion Monte Carlo

Wagner group, Phys Rev B (2014)

$Ca_2CuO_2Cl_2$



$m = 0.5\mu_B$

ab-initio density matrix embedding

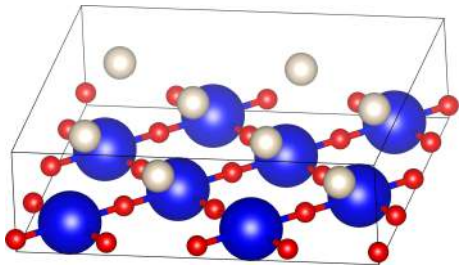
Chan group (2015), in preparation

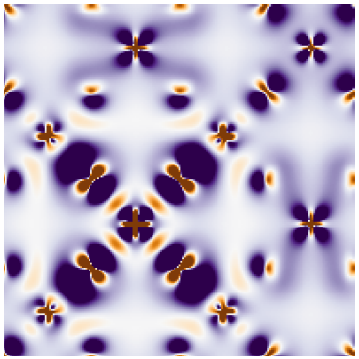
SciDAC: predictive computing

towards cuprate phase diagram with many-particle wavefunctions

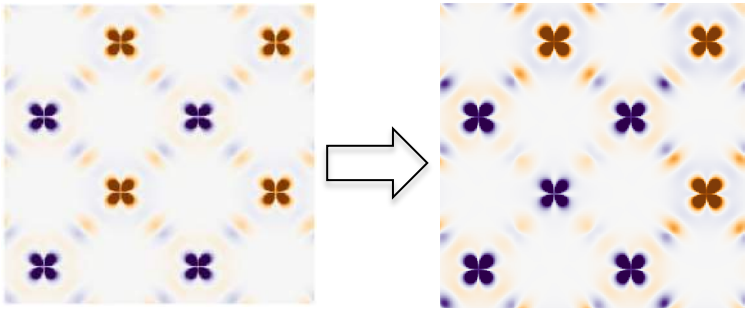
fixed-node DMC simulation of doped $CaCuO_2$

1 hole in 2x2 unit cell (1/8 doping)





charge density
hole localized on oxygen



spin densities
Cu flips spin after doping

single hole forms spin polaron (c.f. Emery-Reiter)

See poster!

Wagner group, arxiv (2015)

SciDAC: predictive computing

2D Hubbard model is **fundamental model** of high Tc superconductivity
long debate over ground-state phase diagram

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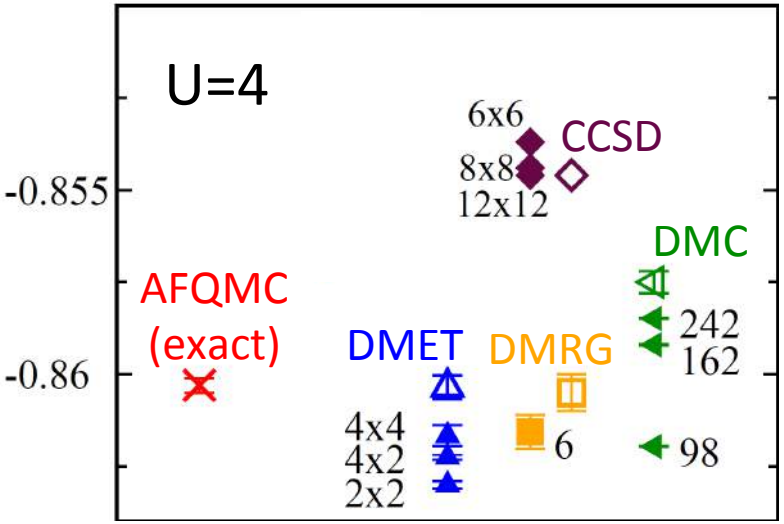
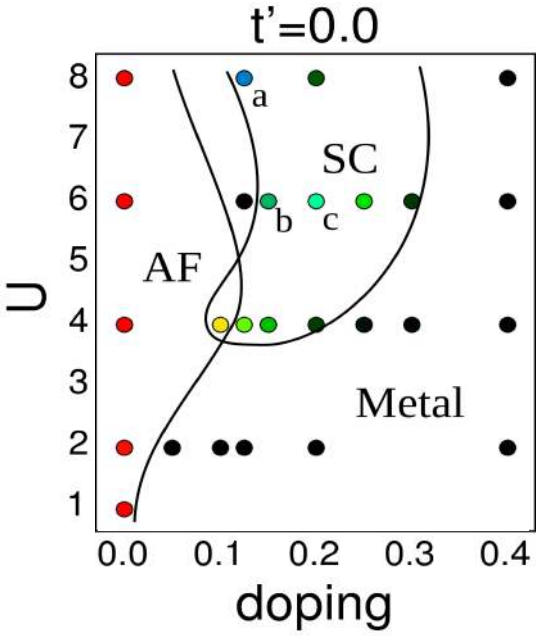
ground-state 2D Hubbard phase diagram to < 20K accuracy from DMET



accuracy of DMET
extensively benchmarked
(Simons Collaboration)



complete phase diagram from DMET
uncertainty **below 20K**

Chan group, arxiv (2015)

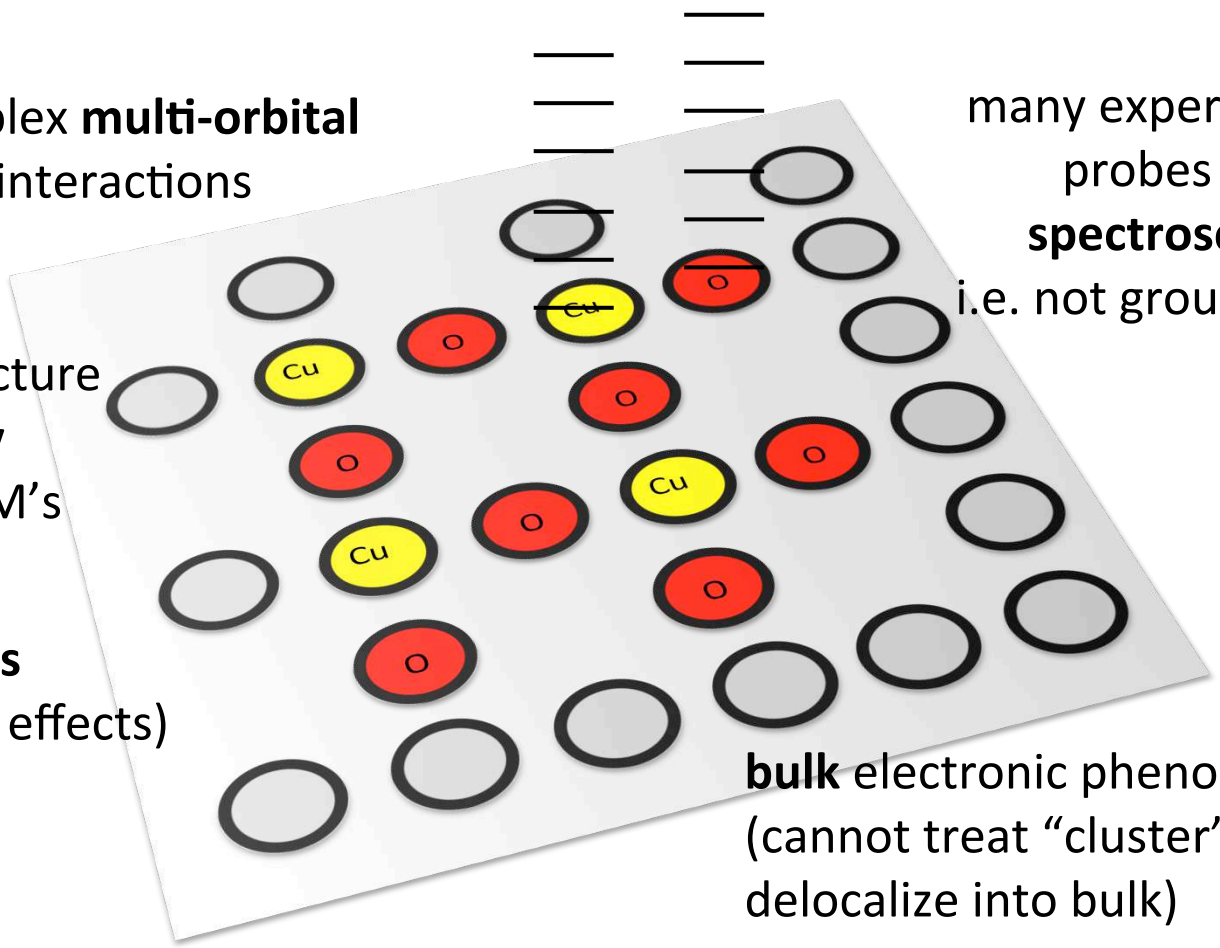
SciDAC: methods

ingredients to simulate a high temperature superconductor

complex **multi-orbital** interactions

many experimental probes are **spectroscopic** i.e. not ground-state

electronic structure of **strongly correlated** TM's



phonons (non-adiabatic effects)

bulk electronic phenomenon (cannot treat "cluster", states delocalize into bulk)

no silver bullet: multiple methods necessary

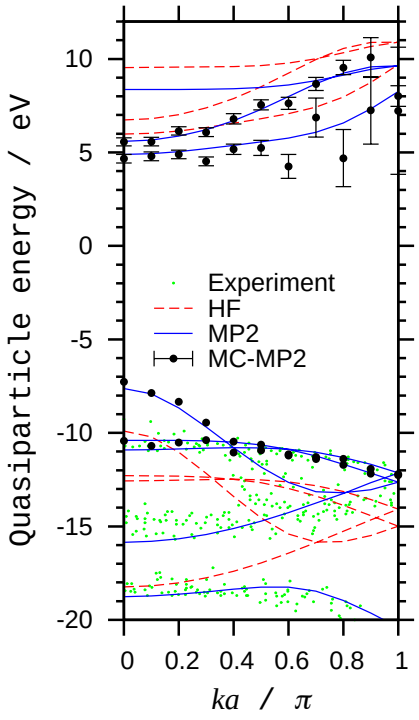
SciDAC: methods

Method	Pros	Cons	ab-initio solids	investigators
quantum Monte Carlo	scalable	sign problem or constraint	✔	Ceperley, Clark, Hirata, Wagner, Zhang
density matrix embedding	cheap	cluster size	✔	Chan
DMRG	flexible	expensive	✔ (with DMET)	Chan, Ryu
tensor networks	v. flexible	v. expensive	not yet	Chan, Clark, Ryu
quantum chemistry	systematic	no strong correlation	✔	Chan, Hirata

SciDAC: method developments

traditional *ab initio* quantum chemistry implemented as matrix-algebra, scales poorly with respect to system size and computer size.

Hirata: reformulate quantum chemistry via Monte Carlo



$$E^{(2)} = \sum_{i,j}^{\text{occ.}} \sum_{a,b}^{\text{vir.}} \frac{\langle ab|ij\rangle\langle ij|ab\rangle}{\varepsilon_i + \varepsilon_j - \varepsilon_a - \varepsilon_b}$$

highly scalable

$$E^{(2)} = -\int \cdots \int \int_0^\infty \frac{G^-(\mathbf{r}_1,\mathbf{r}_3,\tau)G^-(\mathbf{r}_2,\mathbf{r}_4,\tau)G^+(\mathbf{r}_1,\mathbf{r}_3,\tau)G^+(\mathbf{r}_2,\mathbf{r}_4,\tau)}{r_{12}r_{34}} d\mathbf{r}_1 \cdots d\mathbf{r}_4 \, d\tau$$

recent developments

MP2, MP3, MP2-F12,
periodic systems

Polyethylene
band structure

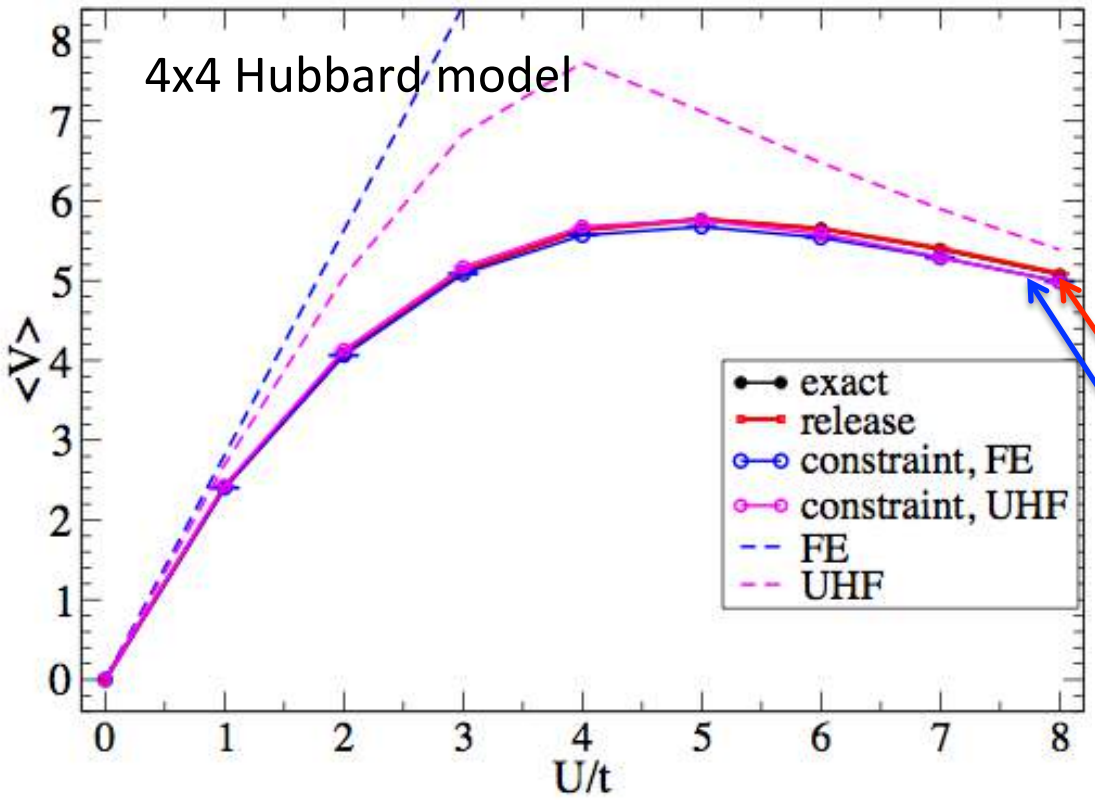
Blue Waters

Hirata group, J. Chem. Phys. (2014), Phys. Rev. B (2014)

SciDAC: method developments

Zhang: auxiliary field Quantum Monte Carlo
random walk in space of non-orthogonal determinants

recent developments



improved trial wfs (with **Chan**)
constraint release
excited states
many-body band structures
spin-orbit coupling
embedding (with **Chan**)
downfolded Hamiltonians

constraint release removes small error of phaseless approximation

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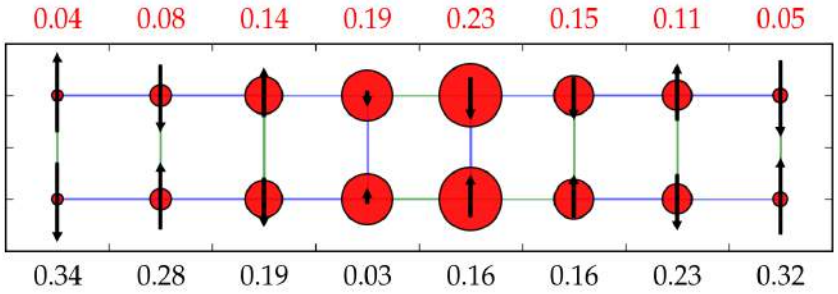
other Monte Carlo developments in PIMC, DMC by **Ceperley**, **Clark**

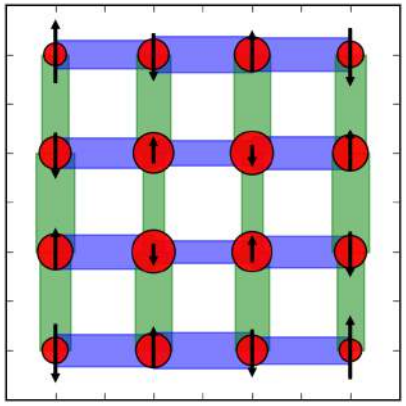
SciDAC: method developments

Chan: density matrix embedding theory: map **infinite** bulk problem to **finite problem** using entanglement theory

recent developments



8x2 DMET impurity order



4x4 DMET impurity order

superconductivity (see earlier)
ab-initio condensed phase (see earlier)
electron-phonon problems
spectral functions
disorder
AFQMC solvers (with **Zhang**)

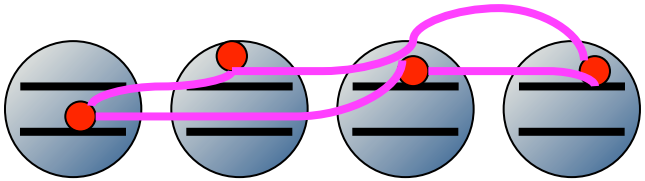
in underdoped region of cuprates,
observed order highly sensitive to cluster shape

Current solver: DMRG, largest cluster ~ 16 sites

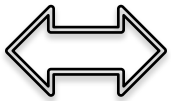
AFQMC solver: greatly increase cluster size

SciDAC: interpretation

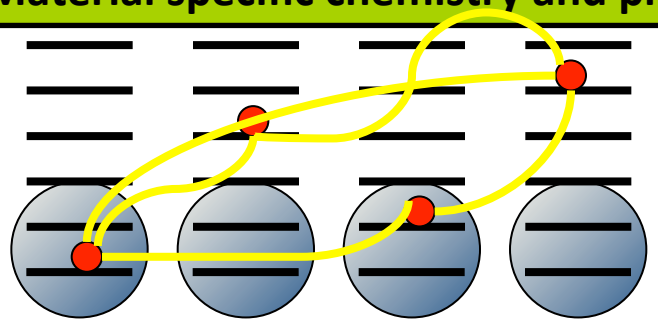
Model (Universal physics)



atoms and interactions are simplified



Material specific chemistry and physics



each atom has many levels, complex interactions

downfolding and **deriving models**

Wagner: fitting QMC data to model Hamiltonians

Zhang: improving AFQMC basis from Kohn-Sham calculations

Chan: deriving models by canonical transformations

Ryu: extracting field theories from low-energy physics of lattices

SciDAC: interpretation

standard procedure: fit model Hamiltonians to a few energies

which energies? ill-conditioned fit? non-unique parameters?

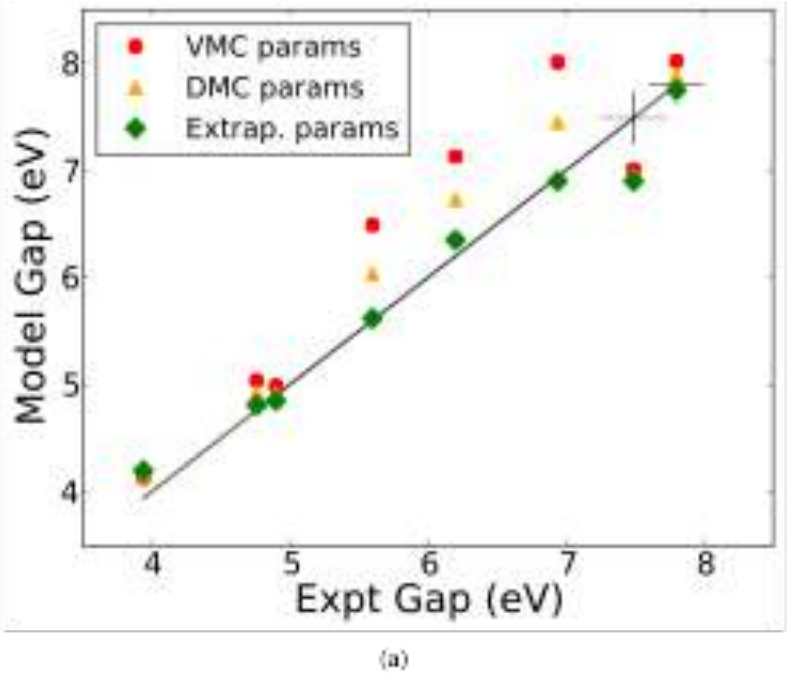
Changlani and **Wagner**

fit using **correlation function** information

$$\begin{pmatrix} E_1 \\ E_2 \\ E_3 \\ \cdots \\ \cdots \\ \cdots \\ \cdots \\ E_M \end{pmatrix} = \begin{pmatrix} 1 & \langle c_i^\dagger c_j \rangle_1 & .. & \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_1 & .. \\ 1 & \langle c_i^\dagger c_j \rangle_2 & .. & \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_2 & .. \\ 1 & \langle c_i^\dagger c_j \rangle_3 & .. & \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_3 & .. \\ 1 & \langle c_i^\dagger c_j \rangle_4 & .. & \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_4 & .. \\ 1 & .... & .. & .. & .. \\ 1 & .... & .. & .. & .. \\ 1 & \langle c_i^\dagger c_j \rangle_M & .. & \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_M & .. \end{pmatrix} \begin{pmatrix} C \\ t_{ij} \\ .. \\ V_{ijkl} \\ .. \end{pmatrix}$$

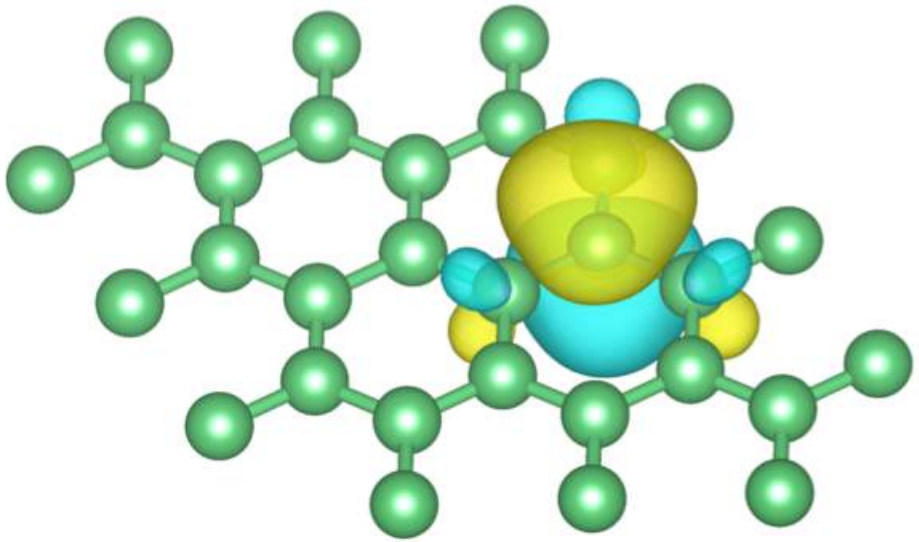
Changlani, **Wagner** group, arxiv (2015)

SciDAC: interpretation



(a)

carbon systems: derived model reproduces experimental gaps over wide range



Graphene

strong or weak correlation?

$$\frac{U}{t} = 1.1(1)$$

weak correlation!

Changlani, **Wagner** group, arxiv (2015)

SciDAC: discovering new physics

ultimate goal: predict **new** phases, new materials, new physics

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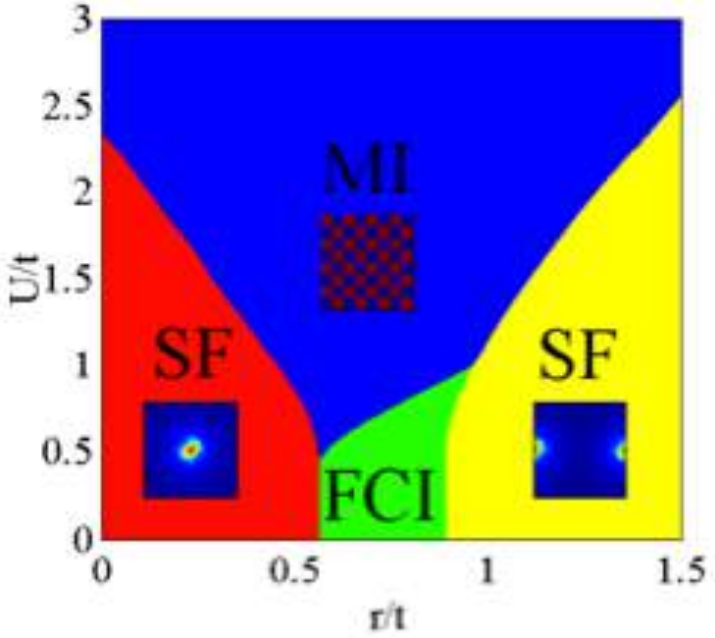
Clark

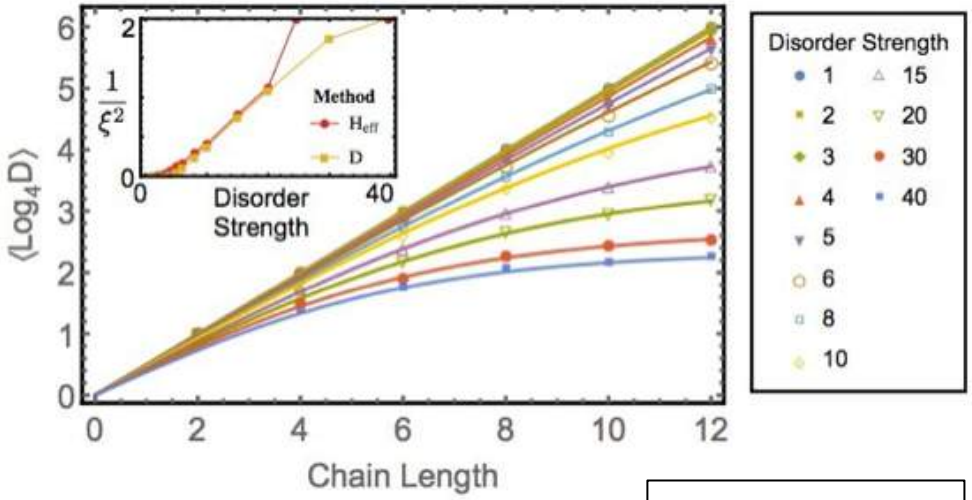
uncovering basic structure of topological and non-topological bosonic wavefunctions

simple wavefunctions for many-body localized phases

new spin-liquid phase in Bose-Hubbard models



representing many-body localized eigenstates through a matrix product operator



Clark, arxiv (2015)

See poster

Software

www.predictive-scidac.org

SciDAC supports **development** and maintenance of **leading packages**

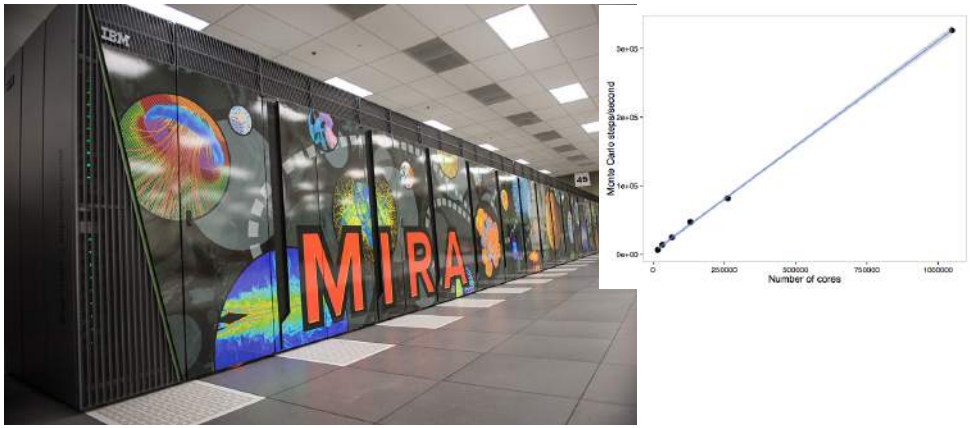
released software

Package	Method	PI
QMCPACK	QMC	Ceperley
PIMC++	PI-MC	Clark
QWalk	QMC	Wagner
Block	ab-initio DMRG	Chan
DMET	DMET	Chan
PySCF	Quantum Chem	Chan
CPMC_Lab	AFQMC	Zhang

Unreleased: POLYMER (Hirata), BIM (Hirata), CP-AFQMC (Zhang), G-AFQMC (Zhang)

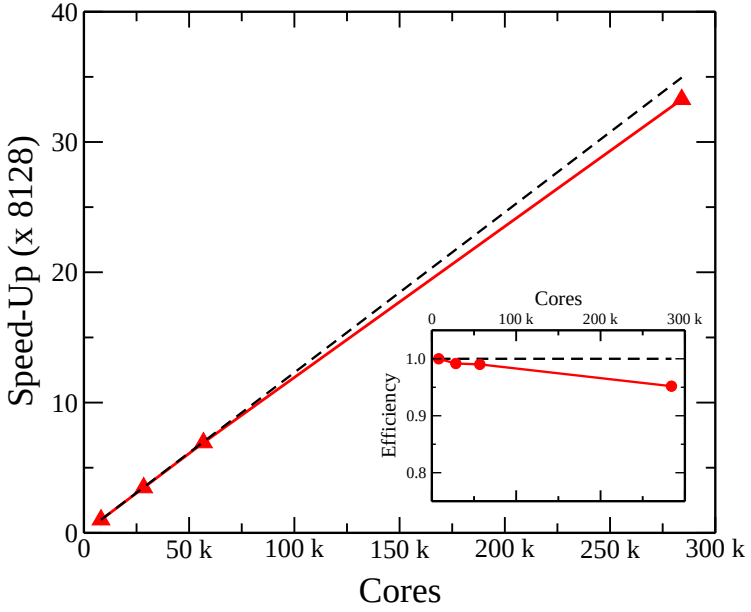
Software

much work spent to achieve **leading performance and scaling**



Wagner: Qwalk, INCITE, 1 million cores

Zhang: AFQMC, 284000 cores



Upcoming Intel MIC presents exciting paradigm, further optimization and **help needed**.

Perspectives

Predictive computation of condensed phase properties achievable in simple materials

Many advances across multiple algorithms

Non-empirical modeling of correlated materials becoming possible

Narrowing gap in capabilities between non-empirical theory and experiment in correlated materials is urgent goal

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Software is released: anyone can calculate!

Emerging architectures present another round of work!