# QMCDB:
# A Living Database to Accelerate Worldwide Development & Usage of Quantum Monte Carlo Methods

Elif Ertekin, Ray Plante, Lucas K. Wagner

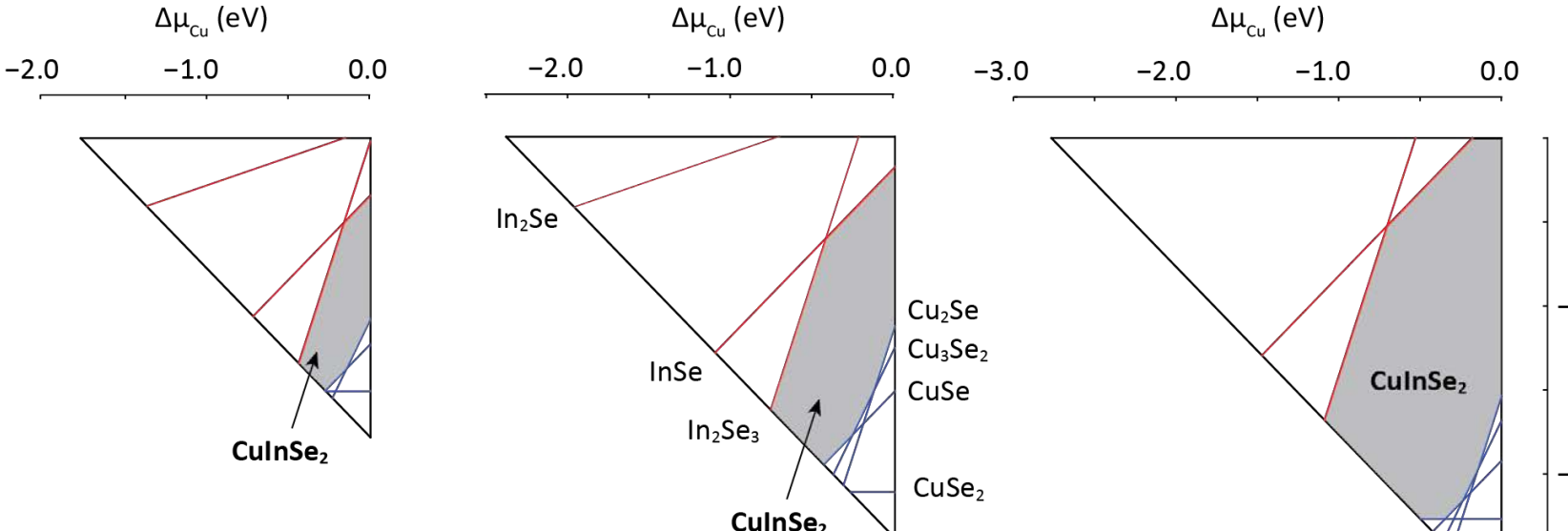
tional materials:
ng materials with
ified properties

d to know: where do
ut the atoms to get
ed effect? And can
ut the atoms there?

Photovoltaic cells in Urba

*Case study of chalcopyrite material $CuInSe_2$*

ky' case,
to predict
lity



uperconductivity
nd magnetism—
o strange
uantum effects
e see day to day

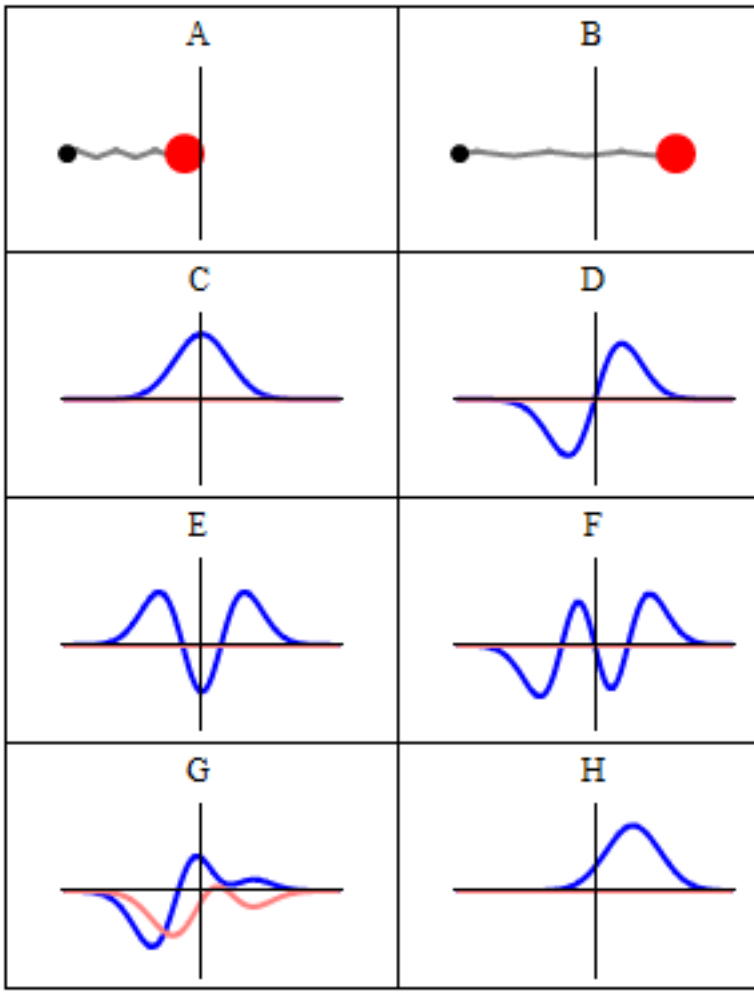
ntum mechanics: wave
tion

are is probability density

hematically we want to
e

$$\hat{H}\Psi_i(r,t) = i\frac{d}{dt}\Psi_i(r,t)$$

ifferential operator



function
nposition

$$\hat{H}\Psi_i(r) = E_i\Psi_i(r)$$

particle
function

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

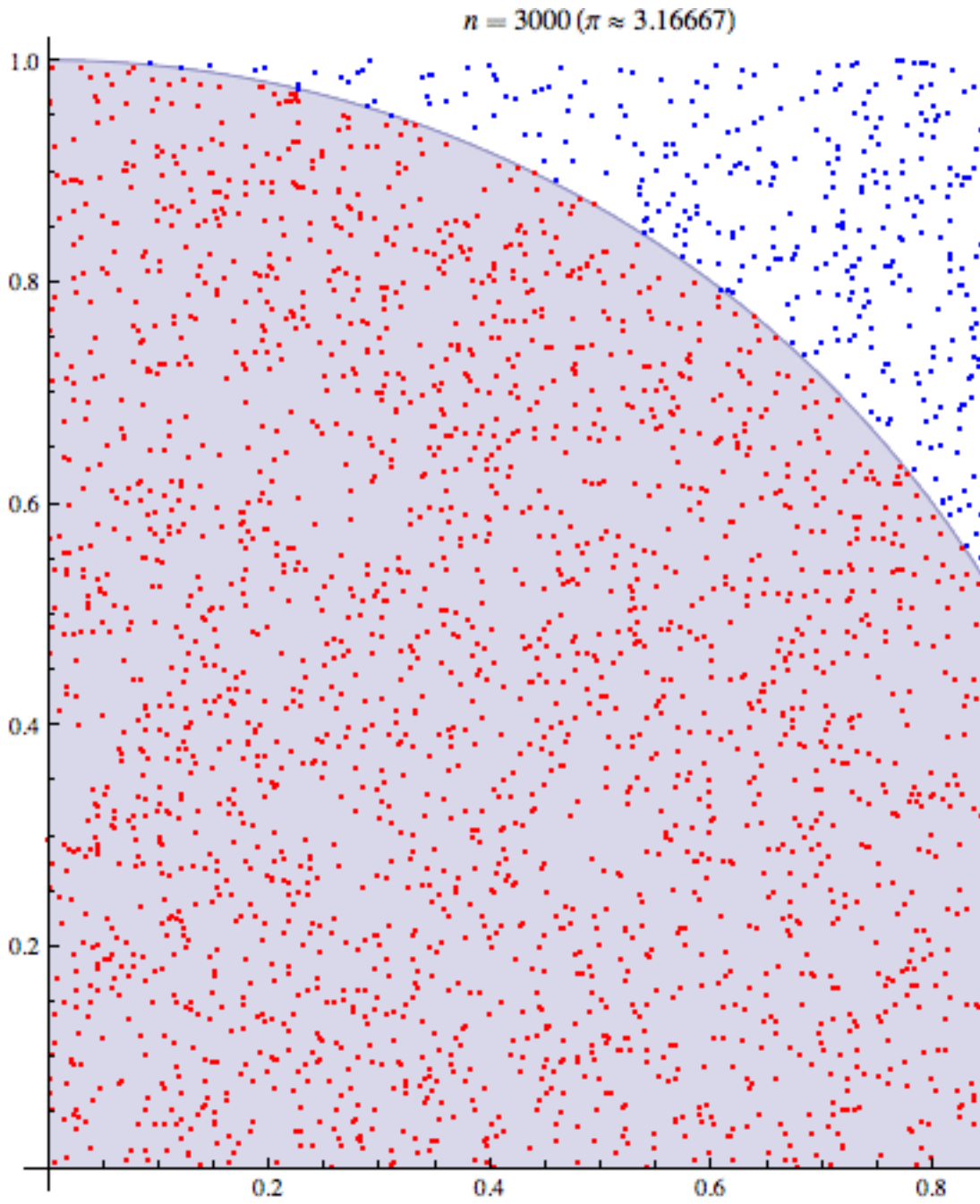
etry
ement

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

high dimensional partial differential equation!

lon’t know an efficient and exact way to solve for t

ple high dimensionality.
ead of worrying about
points, worry about
ance.



$$\hat{H}\Psi_i(r) = E_i\Psi_i(r)$$

ctrons, 300 K is cold

ns spend almost all their time in the lowest energy

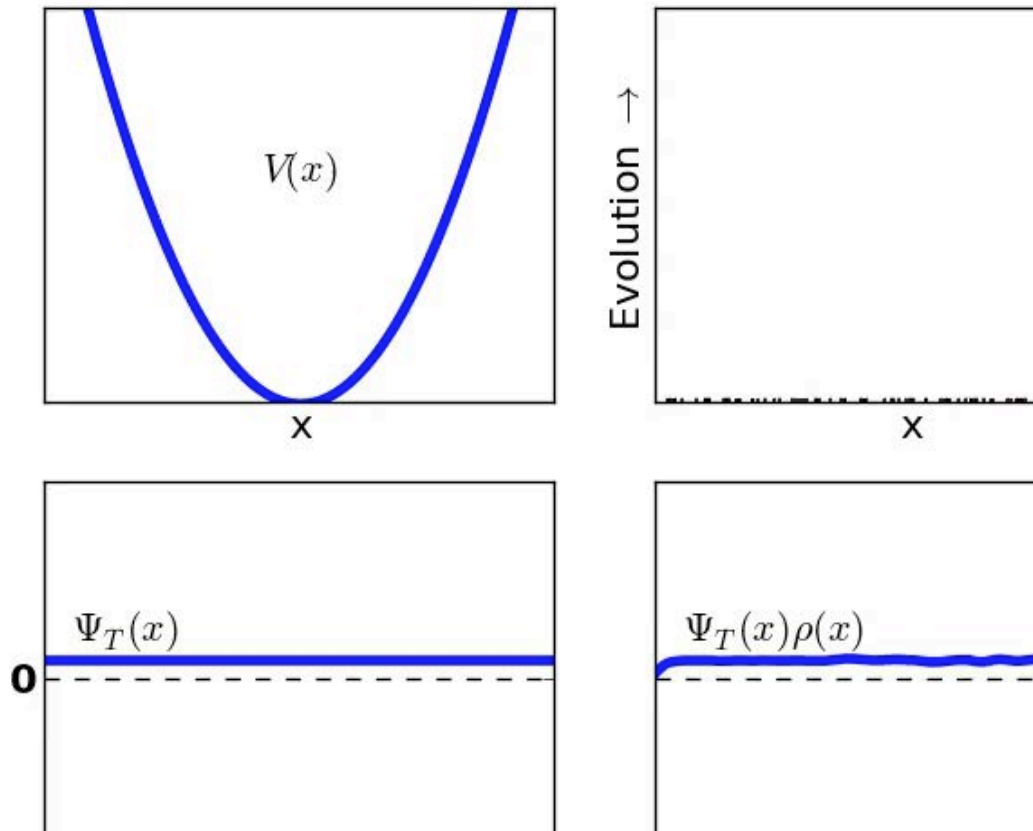
$$\frac{d}{dt}\Psi_i(r,t) = \hat{H}\Psi_i(r,t)$$

to ground state

nto a stochastic process

y of walkers <-> wave

on value



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

a constrained optimization!

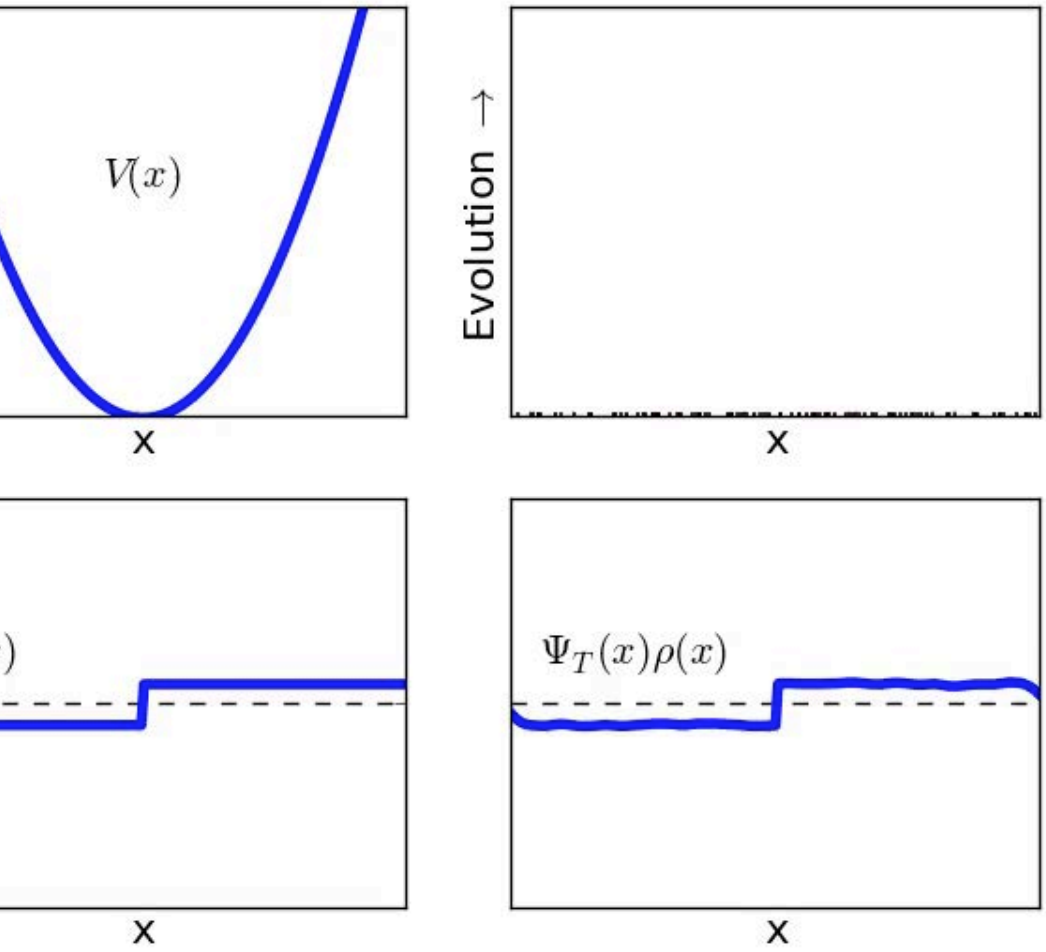
netric state lies below in the spectrum, must proje
e antisymmetric state.

l/noise goes to zero.

pts to get around this only work in very specific
s.

e must approximate!

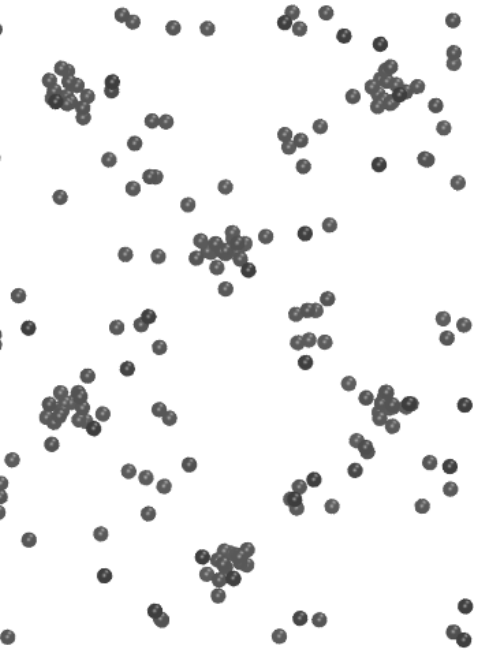
# One approximation to many body S.E. solutions



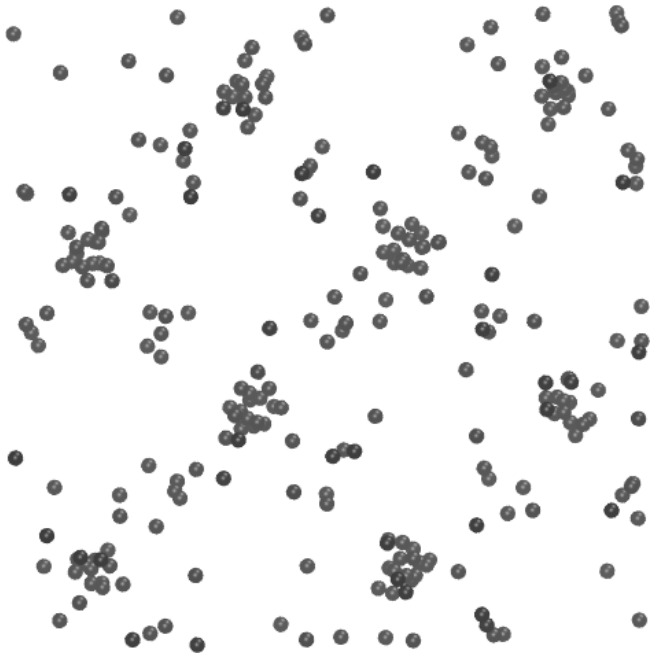

Energy obtained is an u
bound to exact ground

Can vary input wave fu
to get best upper boun

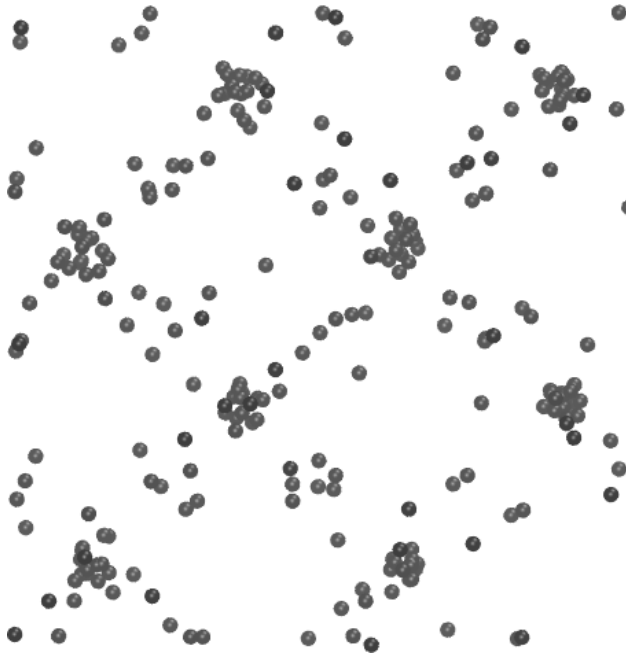
Sample 1



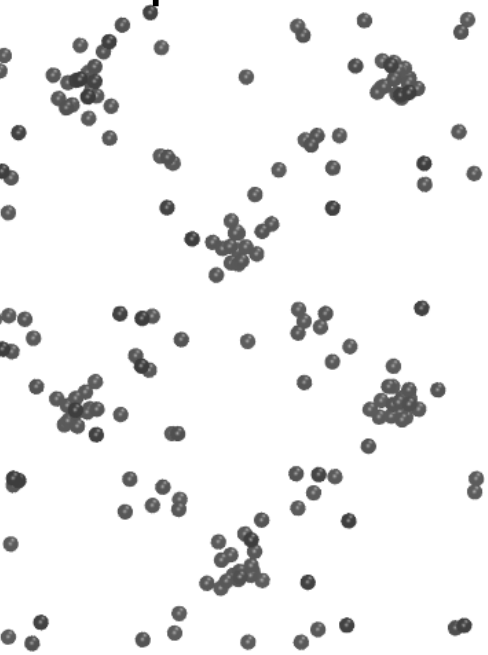
Sample 2



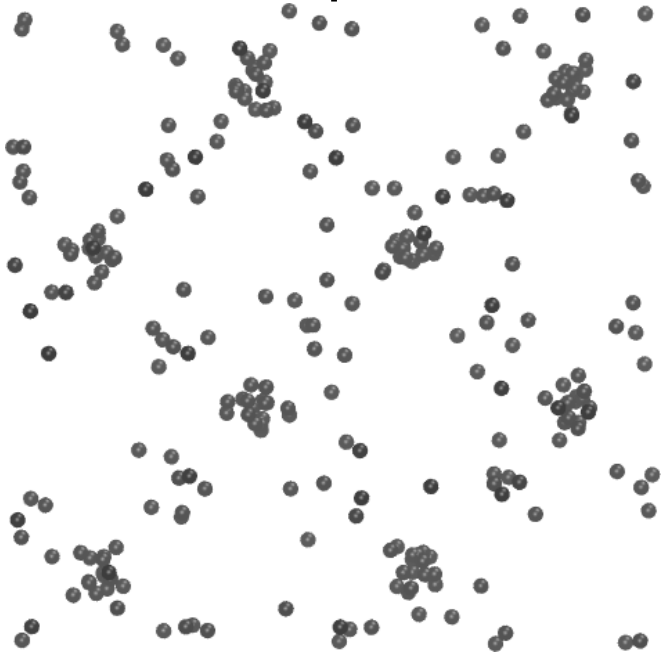
Sample 3



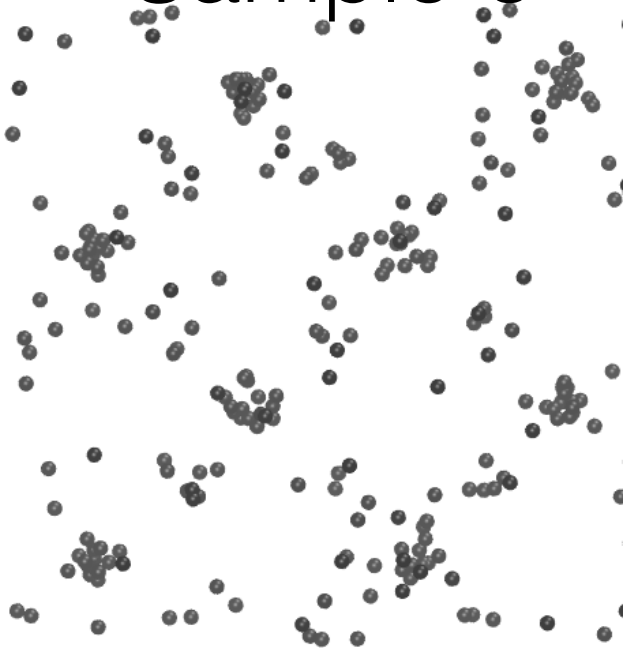
Sample 4



Sample 5



Sample 6



Density functional theory

Slater determinant

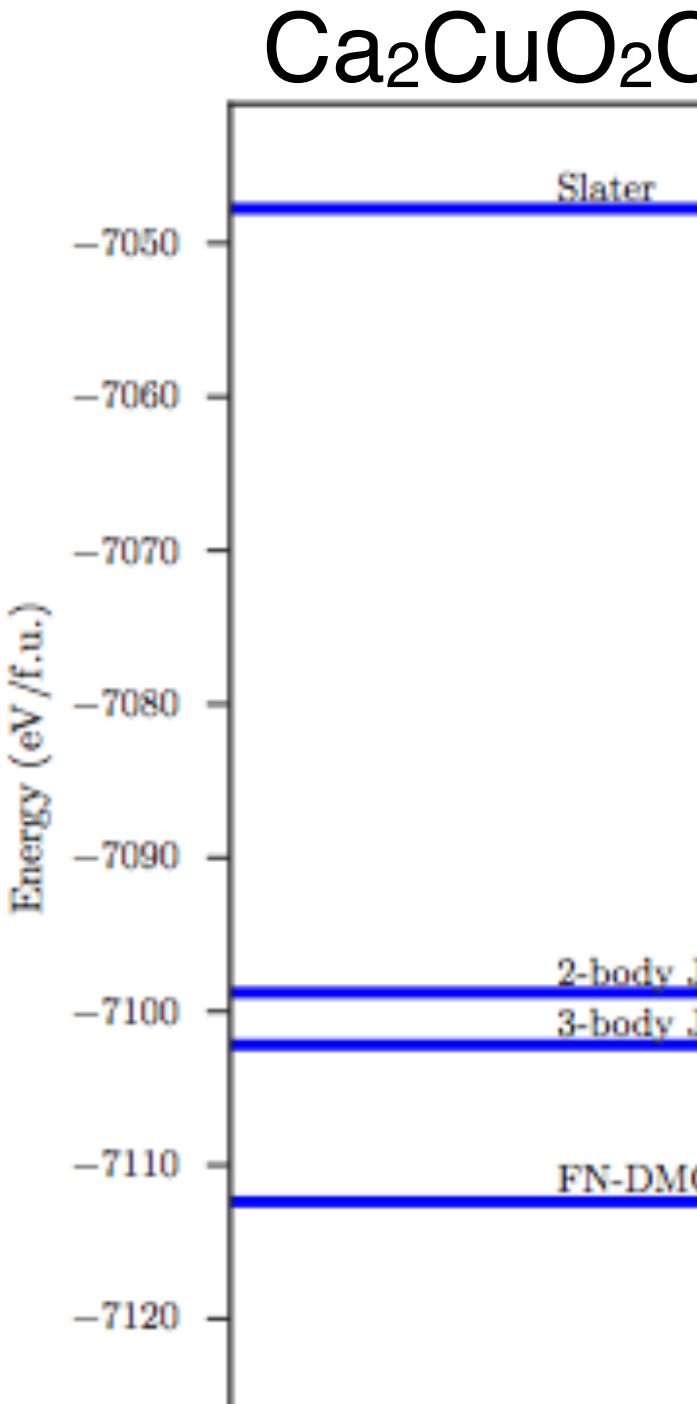
Variational Monte Carlo

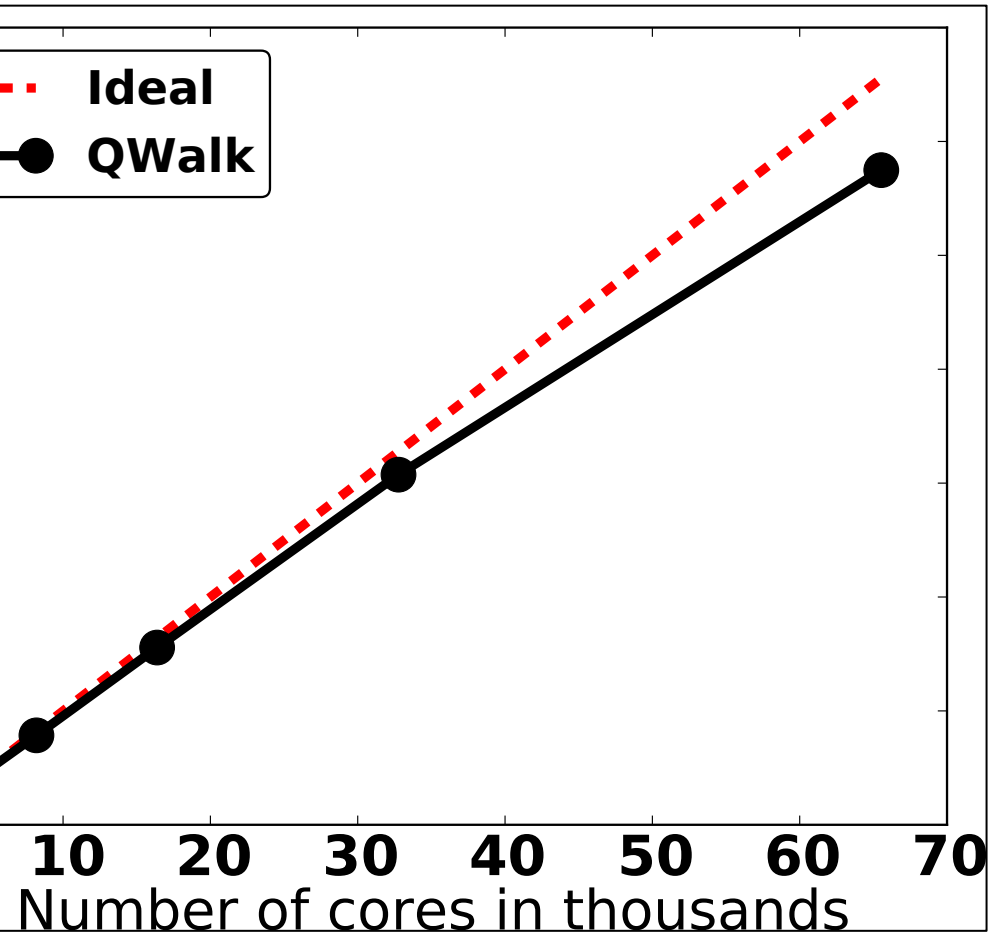
Slater-Jastrow

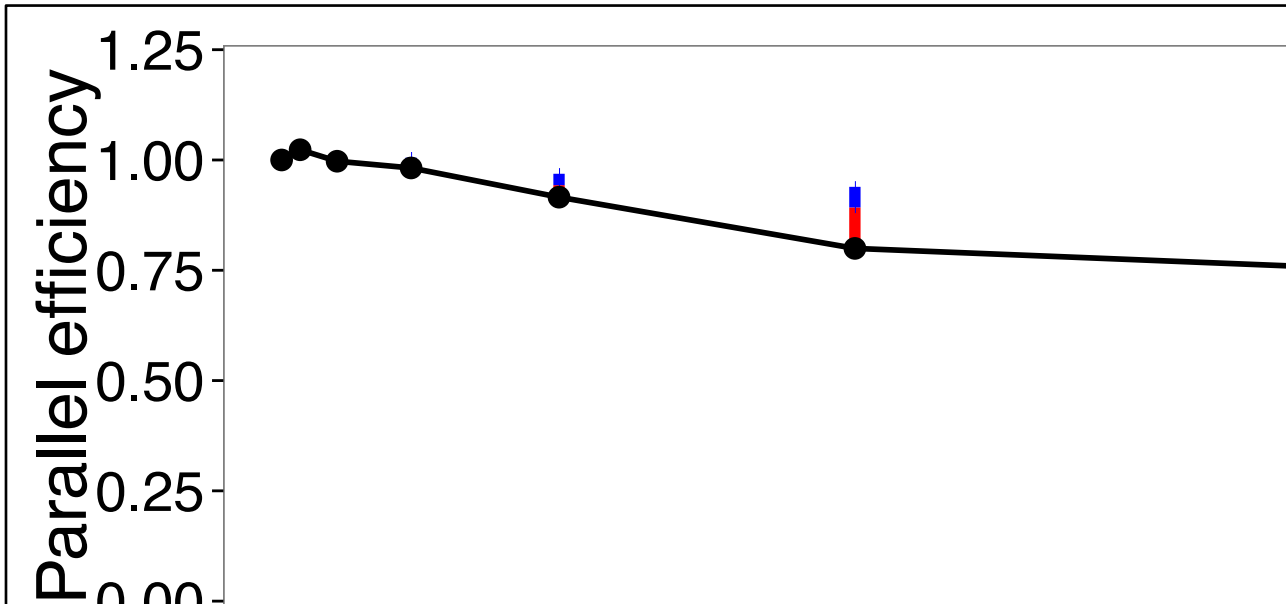
Fixed node diffusion Monte Carlo

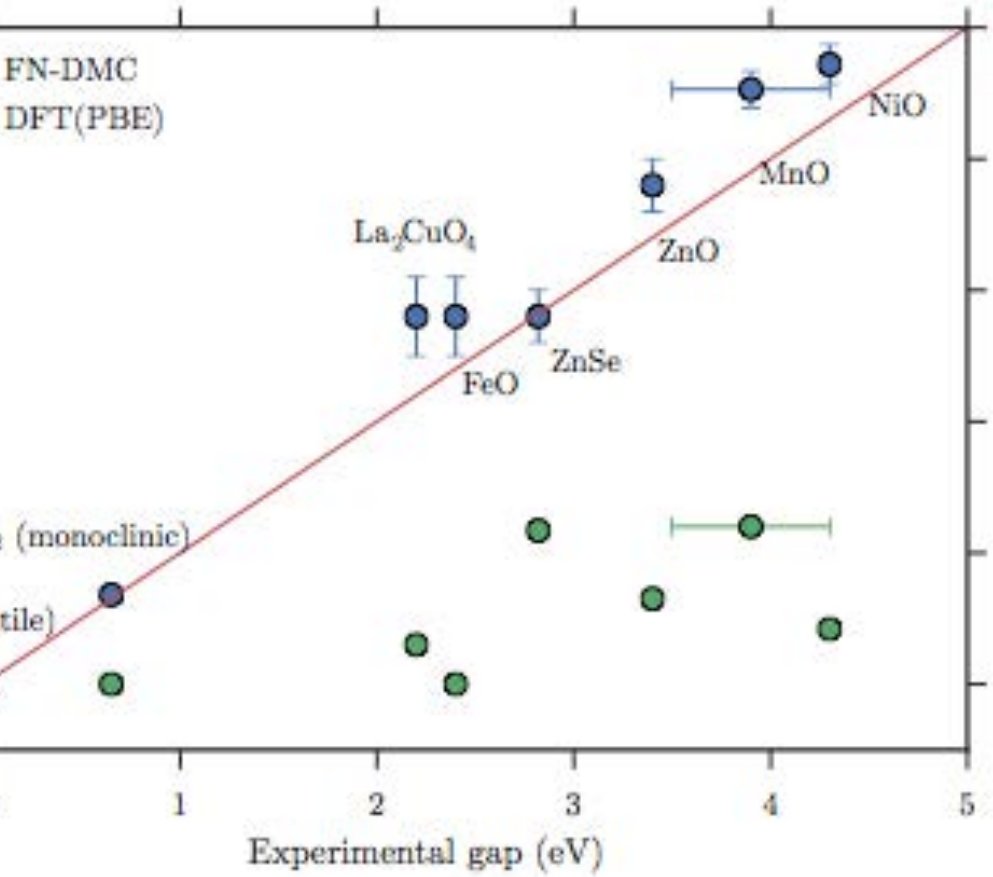
$$\langle \Psi_T | \mathcal{O} | \Phi_{FN} \rangle$$

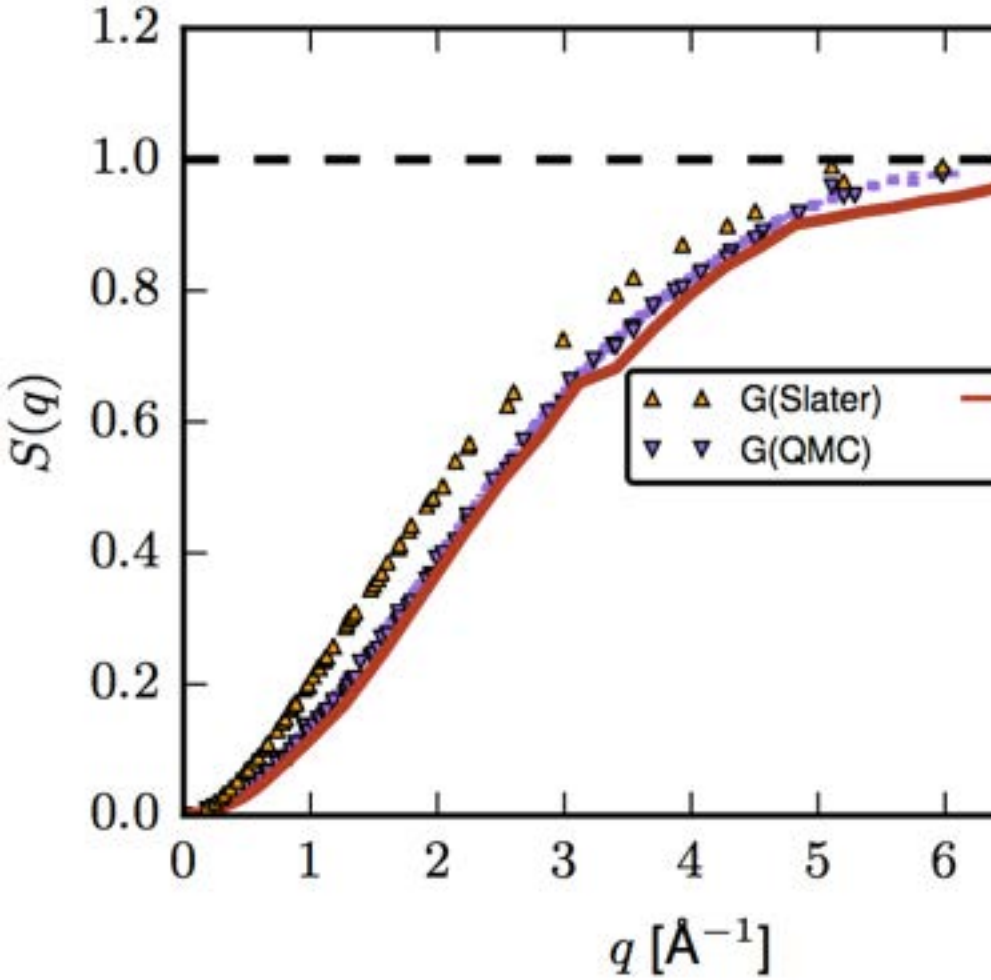
qwalk.org




Ideal
QWalk
10
20
30
40
50
60
70
Number of cores in thousands


Parallel efficiency
1.25
1.00
0.75
0.50
0.25
0.00




$$S(\mathbf{q}) = \frac{1}{N} \sum_{ij} \langle \exp\left[ i\mathbf{q} \cdot (\mathbf{r}_i \right.$$

| proximation | Controlled | Description | Assessment method |
|---|---|---|---|
| eudopotential | No | Replace the core electrons with an effective potential | Vary within the space of reasonable potentials |
| alization | No | Approximate the diffusion Monte Carlo projector in the presence of a nonlocal potential | Vary trial function and projector approximations |
| dal | No | Fix the zeros of the trial wave function when performing diffusion Monte Carlo | Vary the trial wave function, apply variational theorem. |
| estep | Yes | Approximate diffusion Monte Carlo projector | Reduce time step until quantities are converged. |
| te size | Yes | Finite size cells with periodic boundary conditions | Increase the supercell size and average over twisted boundary conditions |

rgy differences are tiny compared to total energy.

culations are tedious, but now thanks to Blue Wate

ed by human time rather than computer

antum

nte

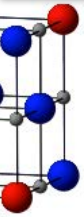
rlo

ta

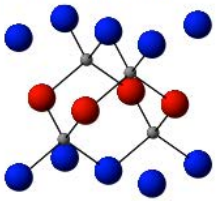
se

## WORKFLOW FOR AUTOMATED MATERIALS CALCULATIONS

**input: tallographic rmation file (CIF)**

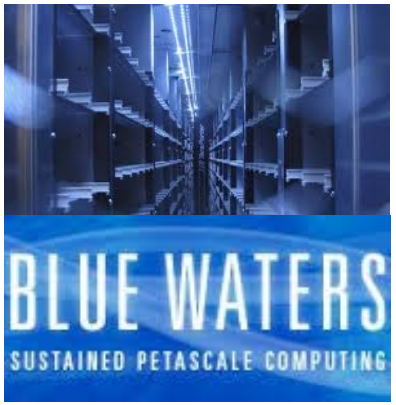


Zinc Blende



**Generate standard input for DFT/ QMC code**

**DFT/QMC simulations on HPC Platform (Blue Waters)**




**Gather provenance & results, generate database entry**

- Run ID
- Method Details
  - Algorithm
  - Software Package
  - Convergence parameters
  - Basis, Pseudopotential
- Total Energy
- Other calculated properties (*i.e.* band structure for DFT)

a computational tool for solid state chemistry and physics

CRYSTAL

QWalk

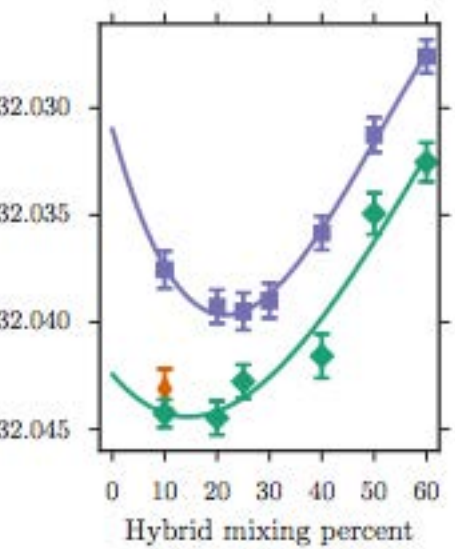
In collaboration with the National Data Service at the National Center for

First implementation

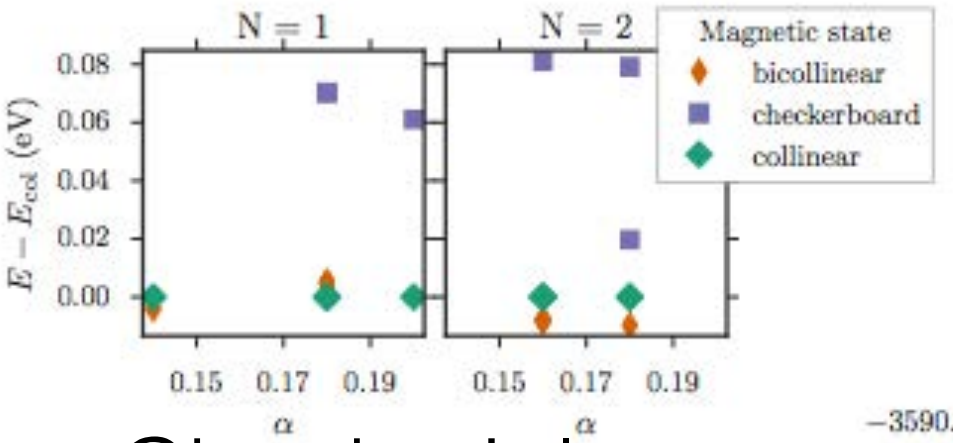
Testing

Hackathon

ub.com/qwalk

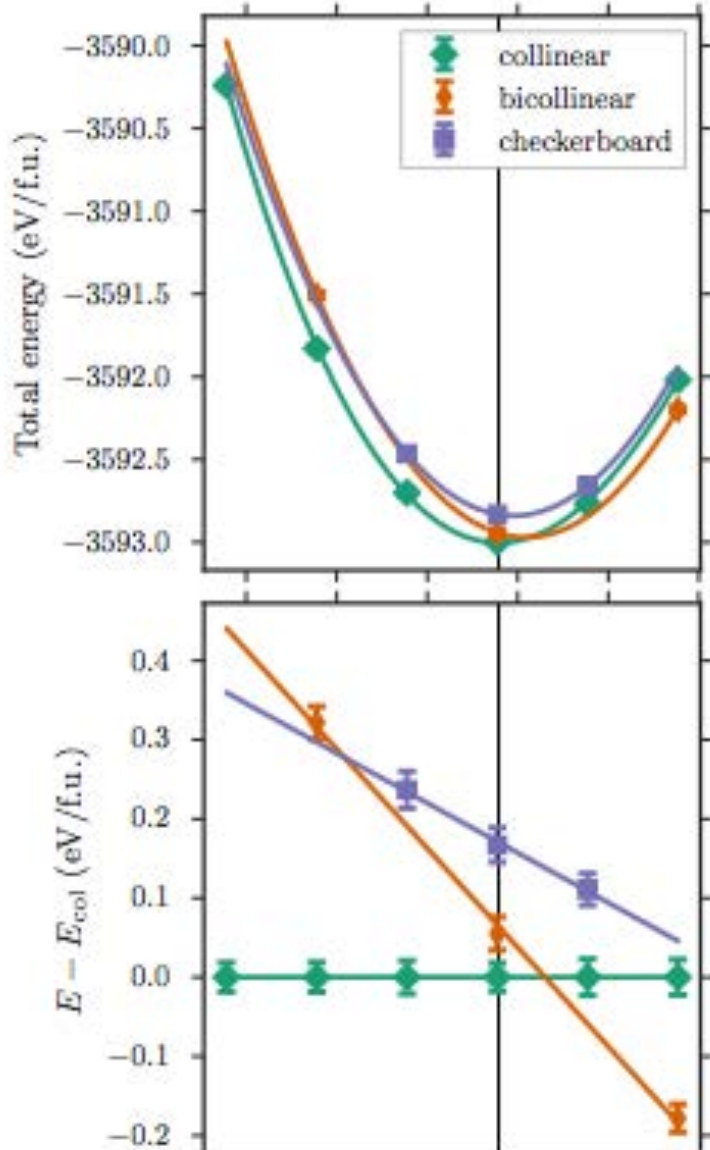



nd best
odal surface



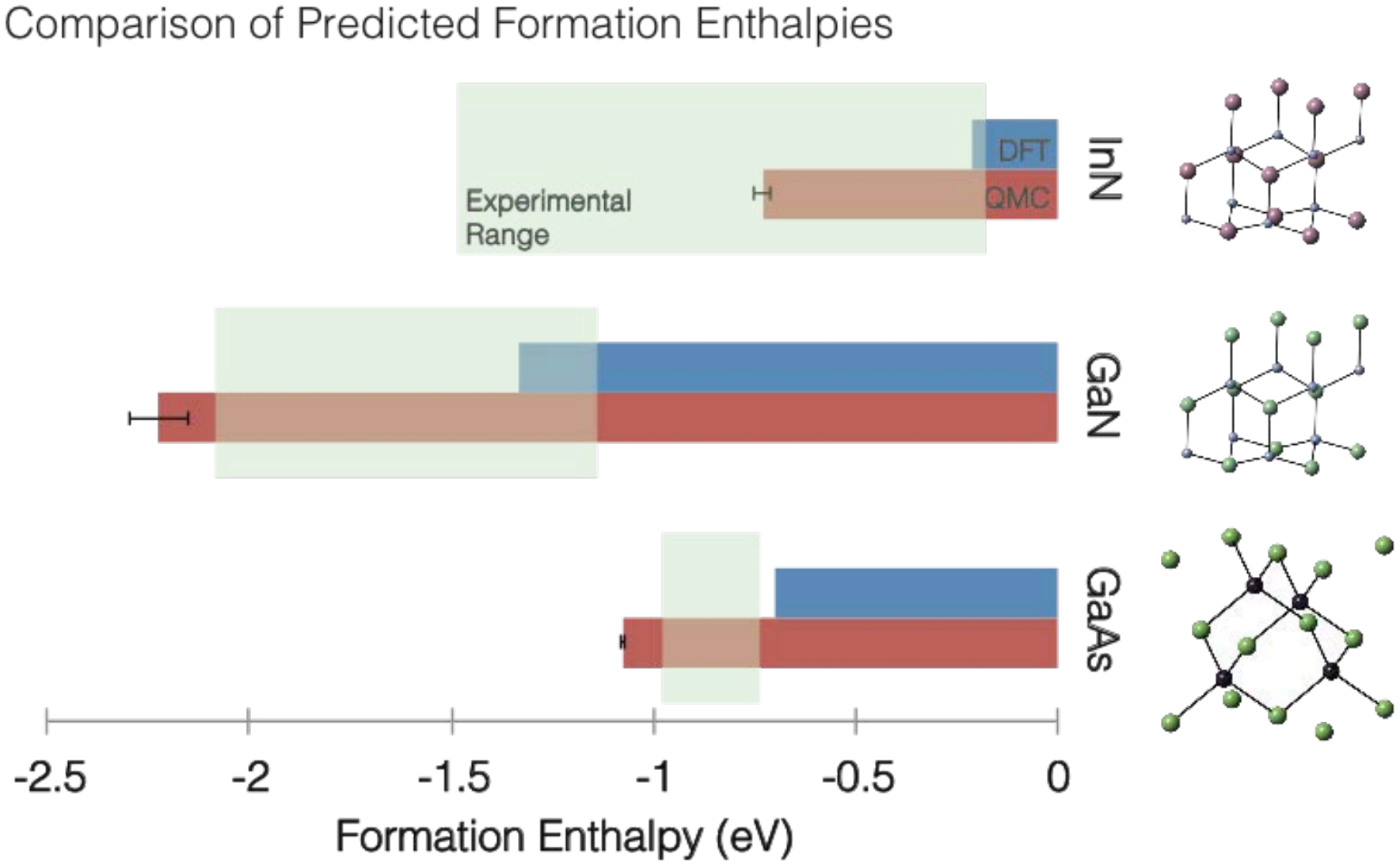

Check trial
function basis

*Br*




erate a bunch of data for
lysis

hout QMCDB (FeSe): 1.5 years
n QMCDB (FeTe): 3 months

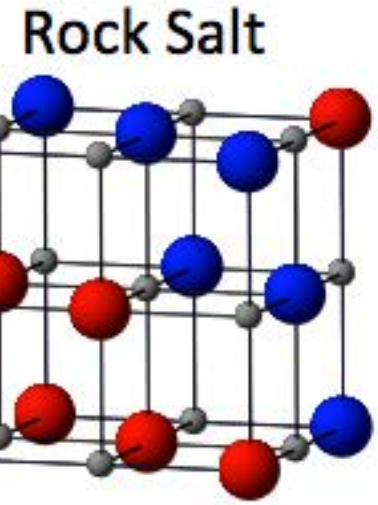



tabase of energies, all systematic

ows comparison of energies -> formation energy

cessary to figure out if we can make a new materi

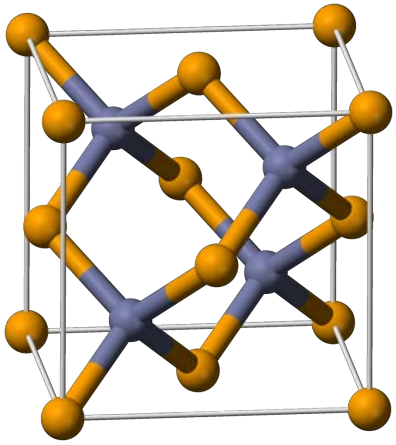
## ganese Oxide

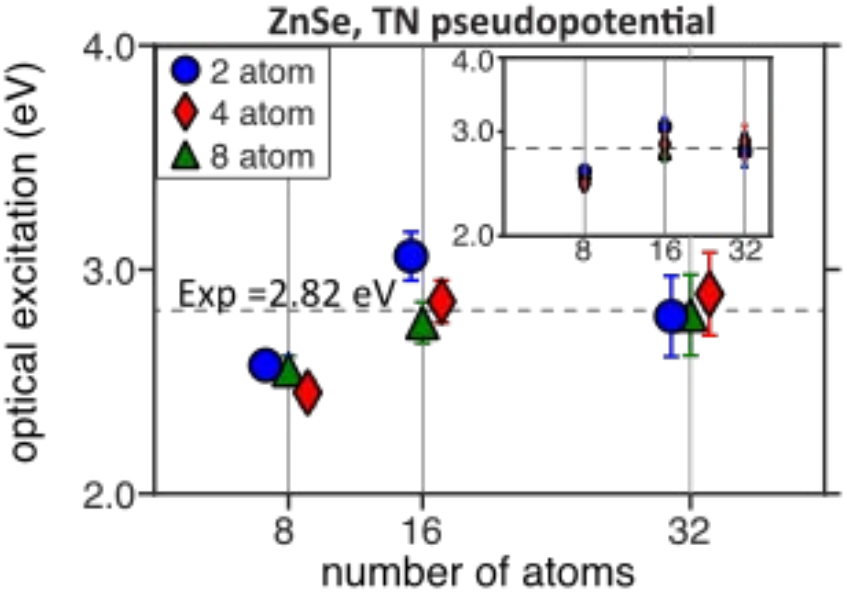



| | Gap (eV) |
|---|---|
| ity | 3.8-4.2 |
| n | 3.6-3.8 |
| ssion<br>opy | 3.9 ± 0.4 |
| ent<br>nents | 3.4 |
| gap) | 4.53(0.14) |

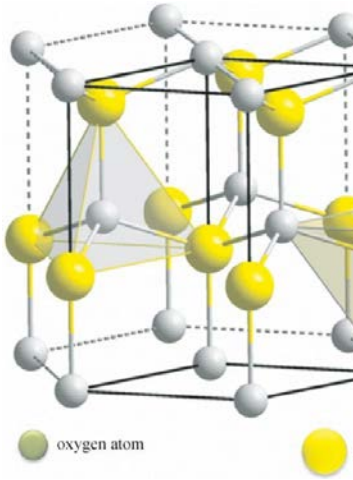
*re*: Kolorenc & Mitas

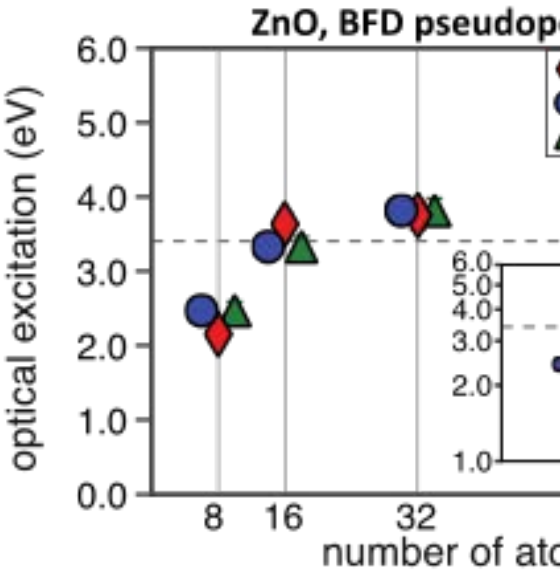
## Zinc Selenide






## Zinc Oxid




Gaps are within few tenths of an eV on very challenging

Some gaps may be over-estimated.

- Finite size effects for excited states?

e able to compute highly accurate reference data
rials more systematically and with much less huma
than before.

s enabling scientific understanding of how electro
act in materials, and how that results in the proper
ee.

materials database of properties calculated using
.

# Related Work

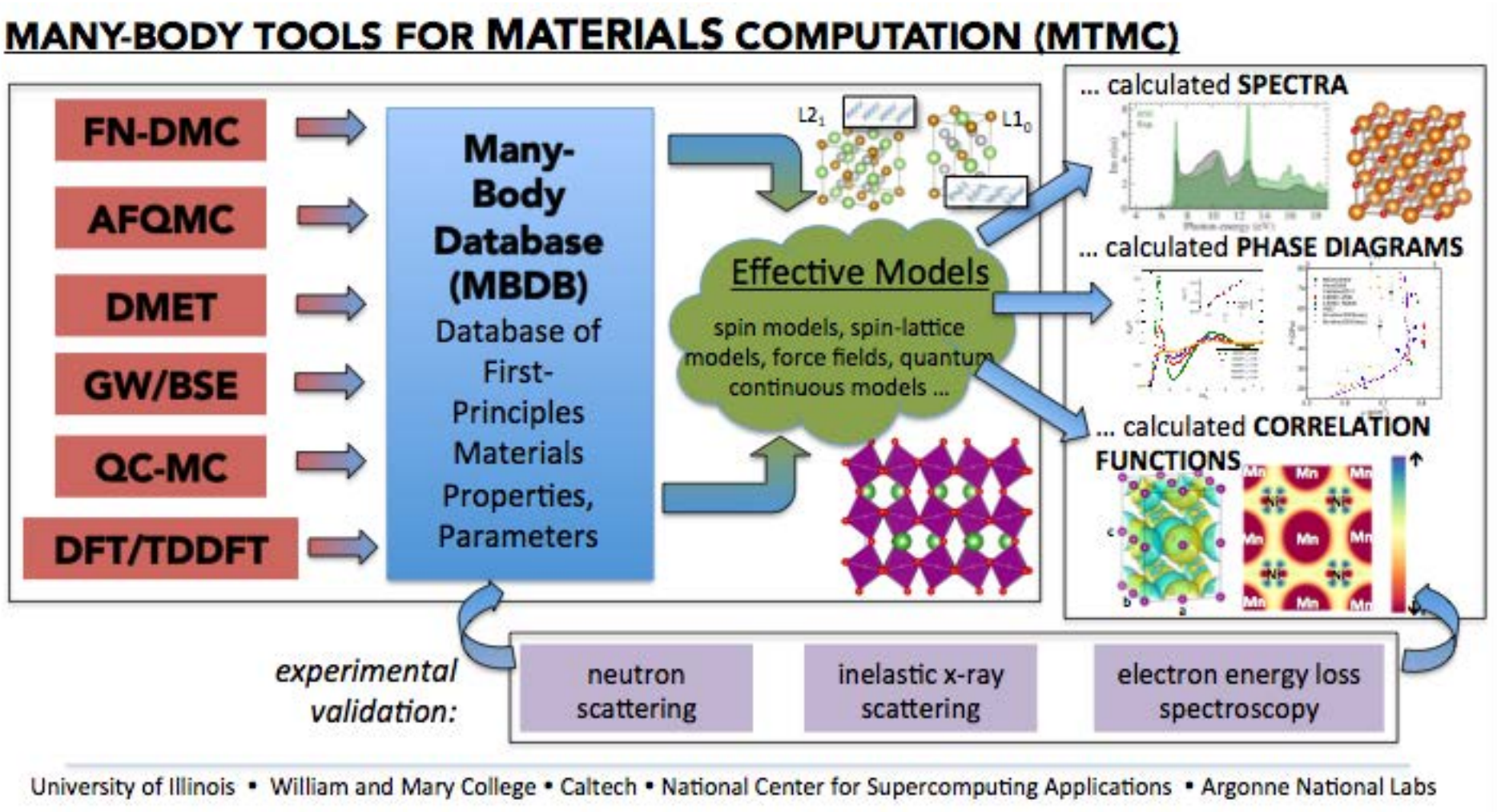
oshua A. Schiller, Lucas K. Wagner, Elif Ertekin, "Phase Stability roperties of Manganese Oxide Polymorphs: Assessment and Ins om Diffusion Monte Carlo", Phys. Rev. B 92, 235209 (2015) OI: 10.1103/PhysRevB.92.235209

aehyung Yu, Lucas K. Wagner, Elif Ertekin, "Towards a systemat ssessment of errors in diffusion Monte Carlo calculations of emiconductors: case study of zinc selenide and zinc oxide", J. C hys. 143 224707 (2015) DOI: 10.1063/1.4937421

aehyung Yu, Lucas K. Wagner, Elif Ertekin, “Quantum Monte Ca mulations of nitrogen defects in zinc oxide”, manuscript submitt r publication.

oshua A. Schiller, Ray Plante, Lucas K. Wagner, Elif Ertekin, Quantum Monte Carlo Database and Autogen: a shared commu ol for many-body statistical simulations of materials”, manuscrip reparation

aehyung Yu, Elif Ertekin, “Exciton binding energy of two-dimensi oS, and WS, from diffusion Monte Carlo”, manuscript in prepara




but in large proposal that uses these ideas

ce to resolve many body quantum problems (at le


# NSF Graduate research fellowships

*Josh Schiller*

*Brian B*