QMC & **QMC**PACK

Summer School 2025

Tuesdays and Thursdays, 1 through 24 July

Session 1 Introduction: Paul Kent, kentpr@ornl.gov

https://github.com/QMCPACK/qmc_summer_school_2025

Funding: U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division, as part of the Computational Materials Sciences Program and Center for Predictive Simulation of Functional Materials.

CPSFM Center for Predictive Simulation of Functional Materials

Sandia National Laboratories

Argonne NATIONAL LABORATORY

NC STATE UNIVERSITY

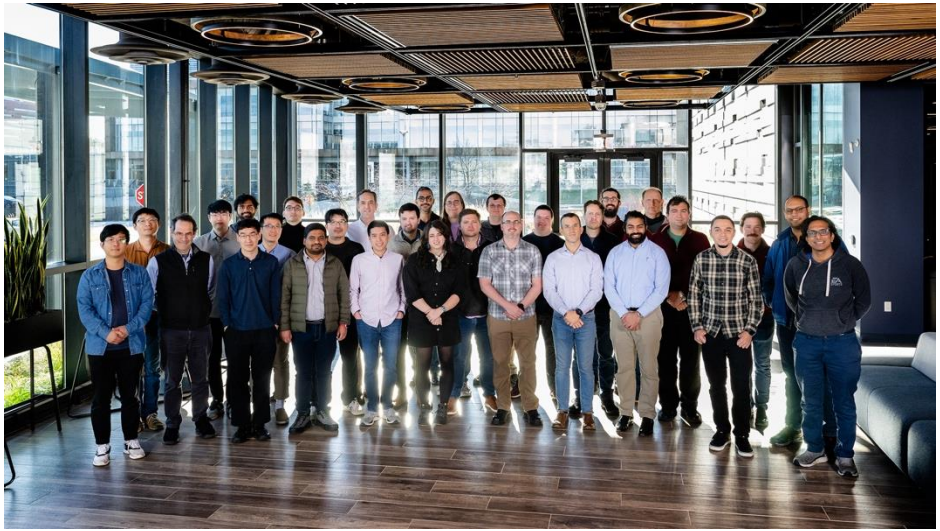
BROWN

OAK RIDGE National Laboratory

Welcome

We have >200 registrants for this summer school from across the globe.

To help with time zone differences, recordings will be uploaded to YouTube.



2023 QMCPACK Users Workshop

Outline

Motivation: Why quantum Monte Carlo?

Logistics for the workshop

Using the workshop virtual machines (VirtualBox & UTM)

Quick example

Questions

Thursday: Theory of real space Quantum Monte Carlo. Foundation for what follows!

Zoom Practicalities

We are recording these presentations for upload to YouTube

Questions and interaction improves the talks! Use the chat feature or Slack for questions. Indicate if you wish to ask the question “live”.

Slack will remain accessible after each presentation while the chat will not.

Thanks to Jaron Krogel @ ORNL who is watching Slack & the Zoom chat.

Acknowledgements

Thanks to the presenters:

Hyeondeok Shin, Ye Luo @ Argonne National Lab

Lubos Mitas @ North Carlina State University

Jaron Krogel, Kayahan Saritas @ Oak Ridge National Lab

Ray Clay, Amanda Dumi @ Sandia National Labs

Juha Tiihonen @ Tampere University

Current QMCPACK users and previous workshop attendees.

Goals for Summer School

1. Cover the fundamentals of real space Quantum Monte Carlo

2. Provide foundation for practical molecular & solid-state calculations using QMCPACK in combination with PySCF and Quantum ESPRESSO

3. Illustrate some real-world research level calculations

4. Provide routes for follow-on support after the workshop

The above topics were most requested. More advanced topics can be covered at our office hours – put what you would like to see in the survey.

Schedule

Tuesdays & Thursday 11am Eastern Time for up to 2 hours, covering presentations and worked examples. Work through examples and make your own experiments afterwards.

“Out of hours” support via Slack.

Thursday 24th July is reserved as a backup session in case of technical issues

Schedule

Session 1, 1 July	Introduction and Kickoff (Paul Kent)
Session 2, 3 July	Fundamentals of Quantum Monte Carlo (Paul Kent)
Session 3, 8 July	Pseudopotentials (Lubos Mitas). Computational workflows with NEXUS, Statistical Analysis (Jaron Krogel)
Session 4, 10 July	Molecular Calculations (Amanda Dumi, Paul Kent)
Session 5, 15 July	Solid-State Calculations (Raymond Clay)
Session 6, 17 July	Real-world Research Calculations (Paul Kent, Kayahan Saritas, Hyeondeok Shin)
Session 7, 22 July	Running on GPUs (Ye Luo), Geometry Optimization with STALK (Juha Tiihonen), Wrap Up (Paul Kent)
Session 8, 24 July	Back session. Only if needed

QMC Workshop Slack

Short link to register: https://bit.ly/qmcworkshopslack

Pick appropriate channels:

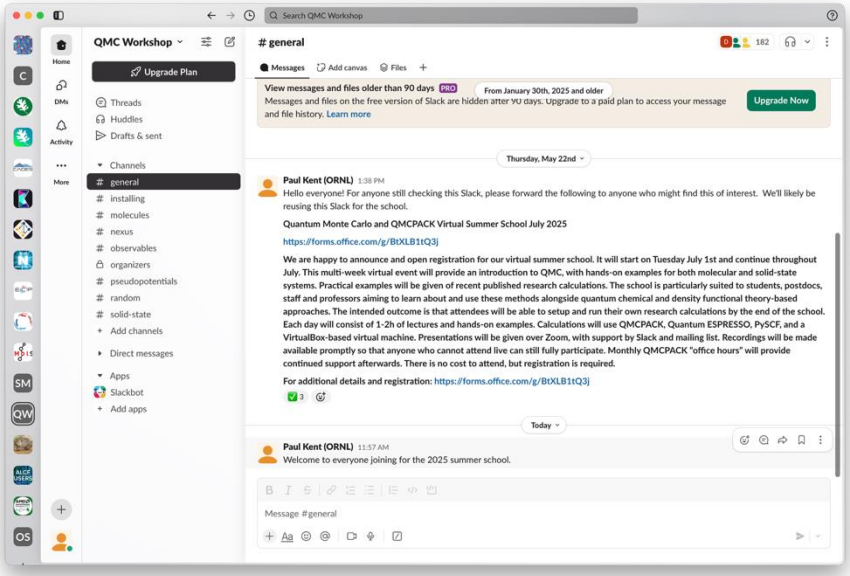
#general

#installing

#nexus

#molecules

#solid-state

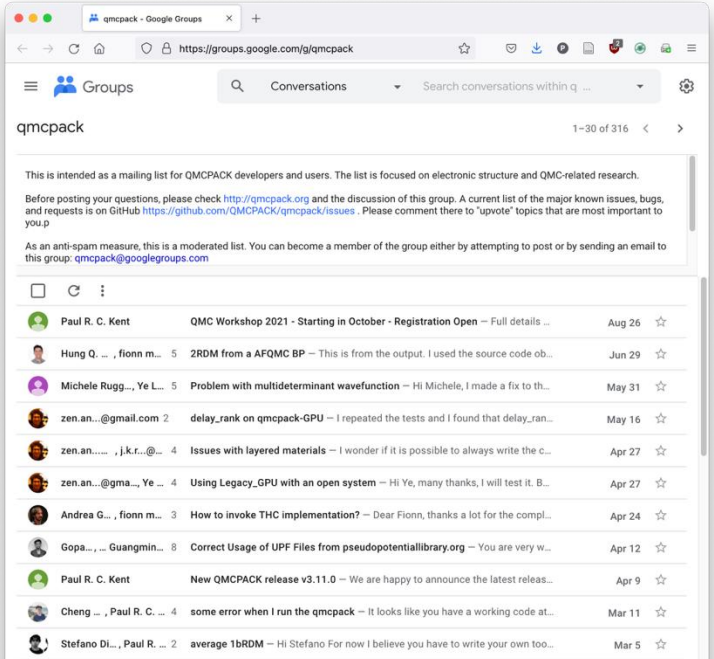


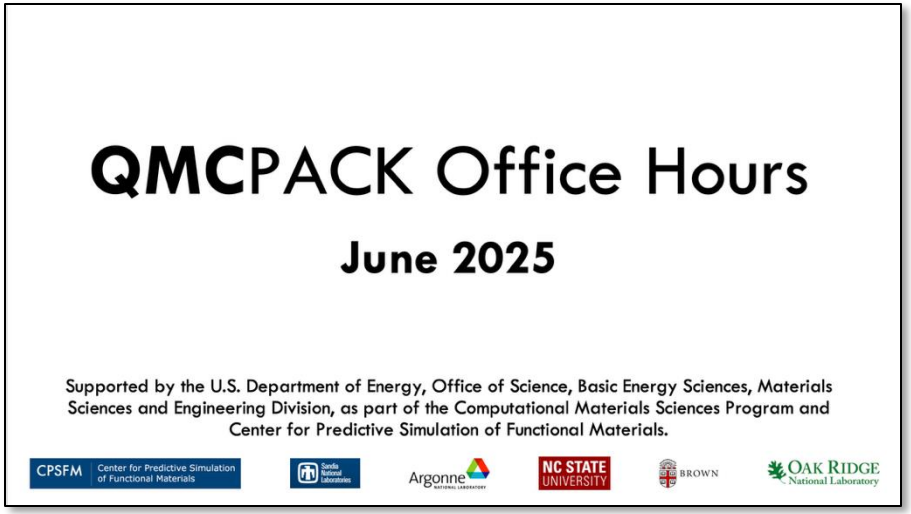
We encourage an interactivity! Questions & discussion welcomed!

Mute or leave Slack notifications enabled as you prefer

Support after the Summer School

Slack channel will remain open. Open an issue on QMCPACK GitHub. Use the QMCPACK Google Group. Attend our monthly “office hours”, typically on Thursdays in the last week of each month. See pinned issue on GitHub.

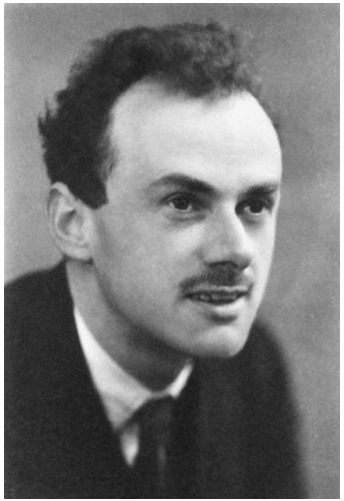




Why Quantum Monte Carlo?

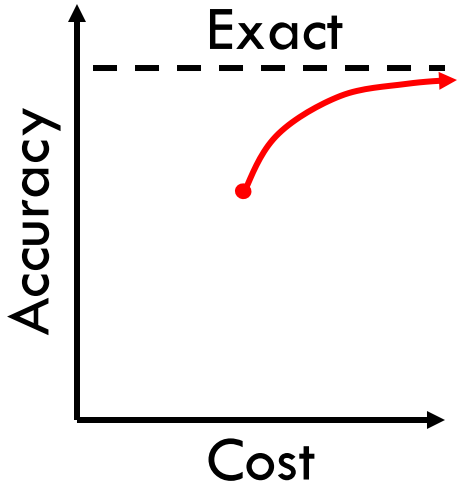
The Electronic Structure Problem

We seek accurate and reliable solutions of the Schrödinger equation. Workhorse DFT based methods are computationally affordable but approximate in practice. We need additional methods where the approximations can be made smaller and applied to general systems: molecules, insulators, metals, surfaces, interfaces, and nanostructures…



"The underlying physical laws necessary for the mathematic theory of a large part of physics and the whole of chemistry are thus completely known, and the difficulty is only that **the exact application of these laws leads to equations much too complicated to be soluble**. It therefore becomes desirable that **approximate practical methods of applying quantum mechanics should be developed**…"

Paul Dirac, Proc. Roy. Soc. (1929)



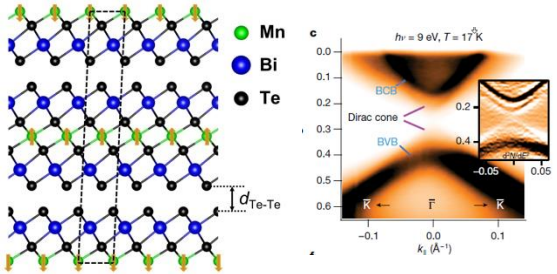
Key Features of Quantum Monte Carlo

1. The methods are very accurate even in their simplest forms.
2. The methods are increasingly affordable and usable for timely and interesting science problems. Scaling $O(N^2\text{-}N^4)$.
3. The few approximations made are known, testable and increasingly able to be converged.
4. The methods provide access to the full many-body wavefunction and many-body physics; choice of input trial wavefunctions is highly flexible.

…

Recent Applications of QMC

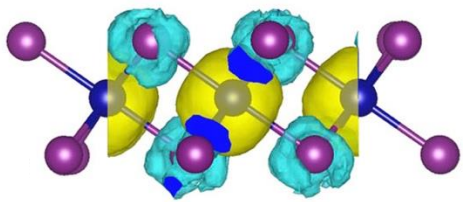
Topology & nature of surface states in $MnBi_2Te_4$



Variation in reported topology of states and band gap explained via stacking faults.

J. Ahn et al.
JPC Letters **14** 9052 (2023)

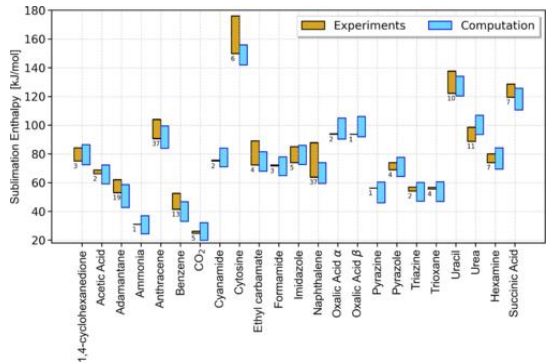
Magnetic and phononic properties of CrI_3



CrI_3 geometry from DMC in agreement with independent STM expt. Novel magnetic structure.

D. Staros et al. JCP **157** 014707 (2022); D. Staros et al. npj Spintronics **2** 4 (2024)

Lattice Energies of Molecular Crystals



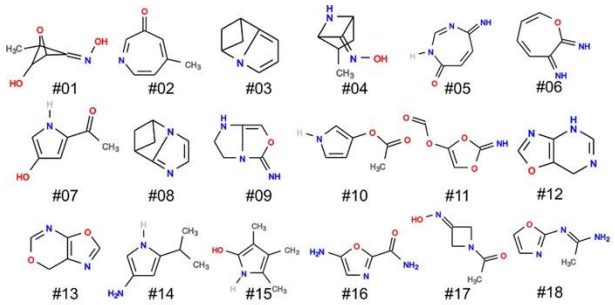
DMC for X23 dataset finds "same or better" reliability than expt (!).

[CASINO] Pia et al.
PRL **133** 046401 (2024)

Computational Cost

Small molecular calculations may take only a few node hours. Many of the earliest nanostructure and solid-state defect calculations can now be run on workstations! Many calculations are in scope using departmental level resources. Heavier elements & large systems still need HPC and careful planning.

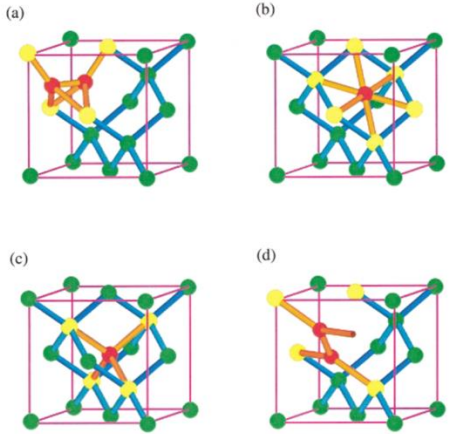
1175 QM9 test set molecules used for delta learning instead of CCSD(T)



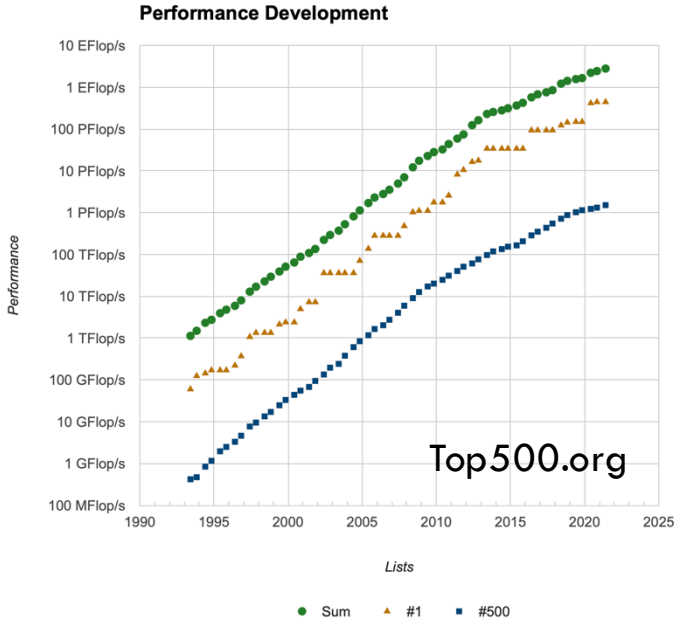
"32 node hours per molecule"

B. Huang et al.
JCTC **19** 1711 (2023)

Si self-interstitial defects



W. K. Leung et al.
PRL **83** 2351 (1999)



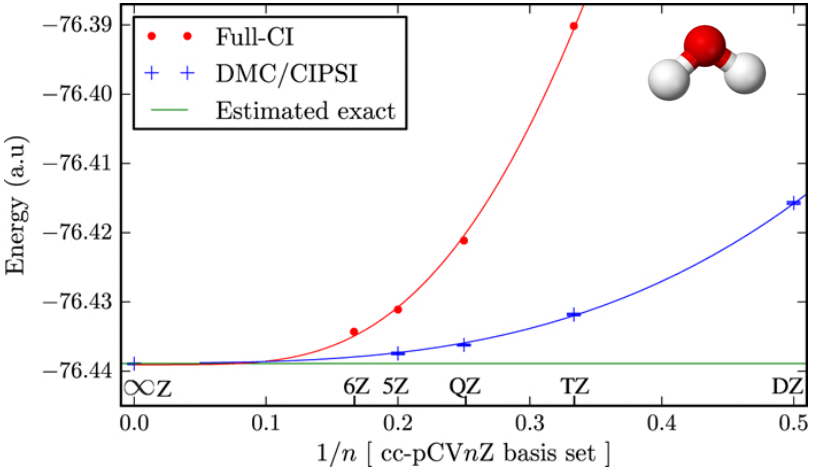
Improvability (rare/unique capability)

While highly accurate, QMC methods make a few approximations in solving the Schrodinger equation, as typically applied.

The approximations can now be tested and converged. E.g. Using large multideterminant expansions to converge the nodal error due to the Fermion sign problem. Importantly, most QMC methods are variational in the energy.

All technical convergence parameters also need to be handled: time step, cell size etc.

"Pay more, get more"



H_2O molecule
−76.438 94(12) a.u. CIPSI-DMC
vs −76.438 9 a.u. Experiment

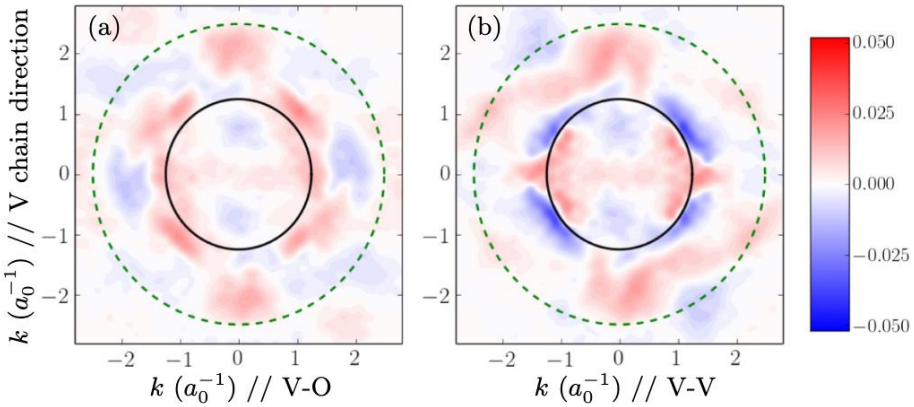
Most accurate water calculation to-date

Caffarel et al. JCP **144** 151103 (2016)

Solids: Benali et al. JCP **153** 1841 (2020)

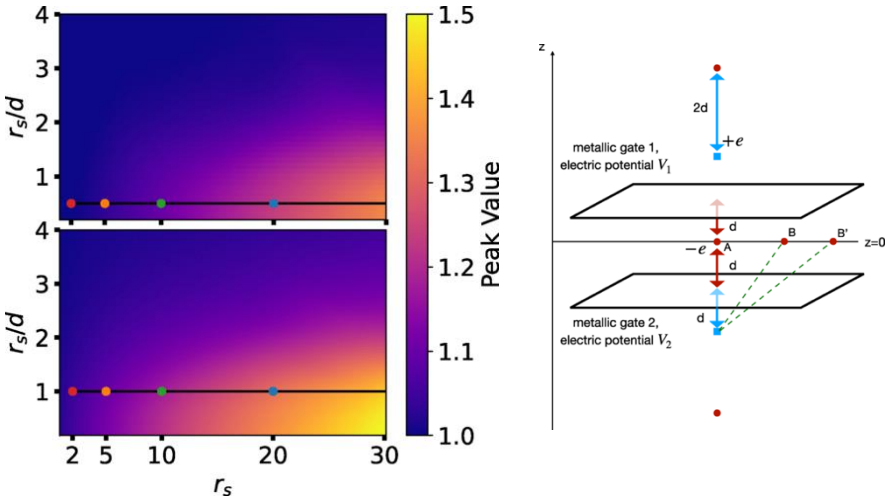
Many-body physics

QMC methods give access to the full many-body quantum mechanical wavefunction. Can compute any property! Density matrices, momentum distributions, pair-correlation functions, and 'beyond single particle physics' can be studied in general. Under exploited!



Difference in momentum distribution n(**k**) in VO_2 through R-M1 phase transition

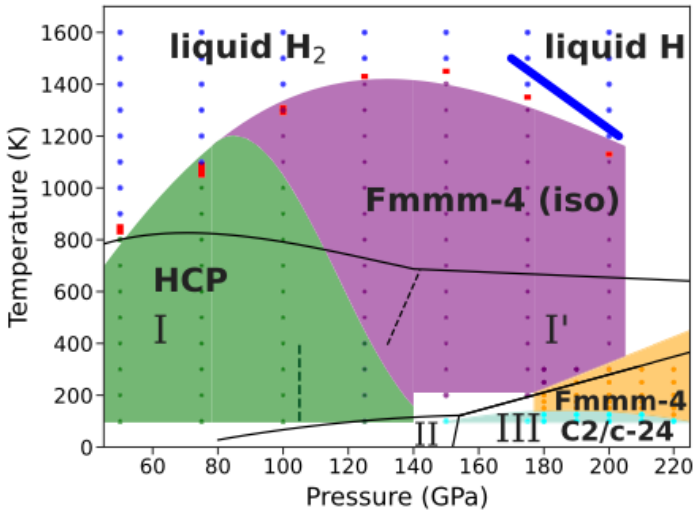
I. Kylanpaa et al. PRB **99** 075154 (2019)



Analysis of structure factor S(k) in 2D Homogeneous electron gas under dual gate screening.
DFT functional also created from correlation energy.
Y. Yang et al. PRB **111** 045136 (2025)

Use for Machine Learning

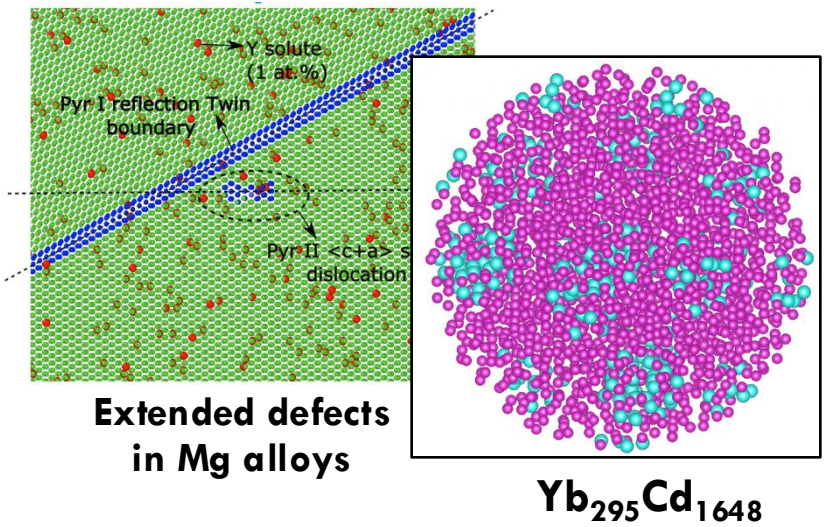
MLIPs from QMC data



Niu et al. PRL **130** 076102 (2023), UIUC group

Hydrogen phase diagram using machine learned potential from QMCPACK data. Testable approximations required for progress.

New DFT approximations from QMC?



Extended defects in Mg alloys

$Yb_{295}Cd_{1648}$

2023 Gordon Bell Prize, V. Gavini et al. (U. Michigan)

Mg alloys and quasicrystals using machine-learned DFT approximation; called for QMC data.
See also: Skala, DM21 functionals

Machine learning provides several routes to systematic (non-ad hoc) upscaling

Using the workshop Virtual Machine

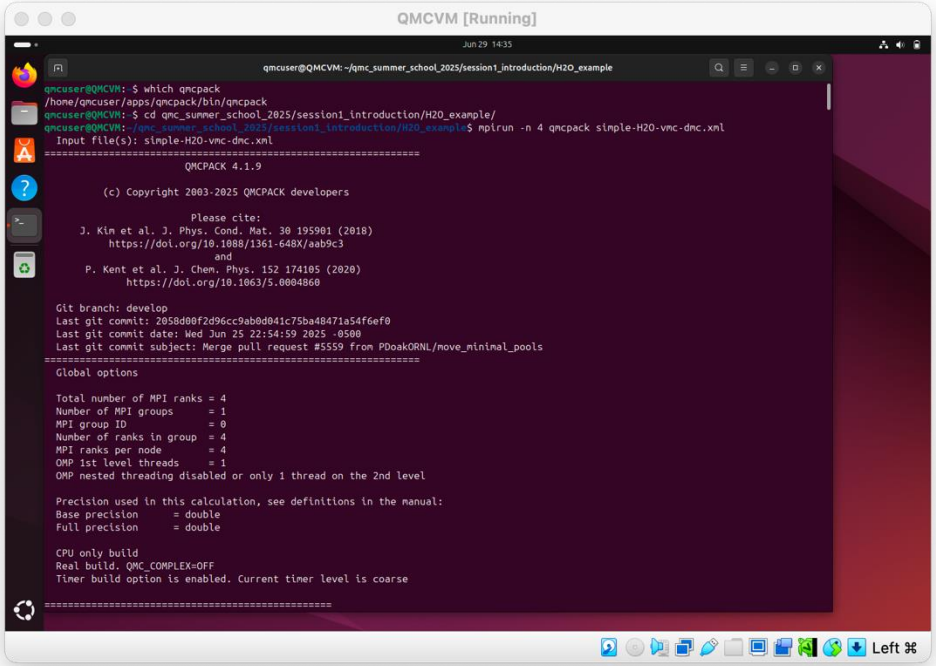
Workshop Virtual Machine

The VirtualBox & UTM Virtual Machine image is a complete desktop for running QMC, DFT and quantum chemical calculations.

The VM is an Ubuntu 24.04 or 25.04 desktop with QMCPACK, NEXUS, Quantum ESPRESSO (QE), PySCF installed, as well as the workshop files.

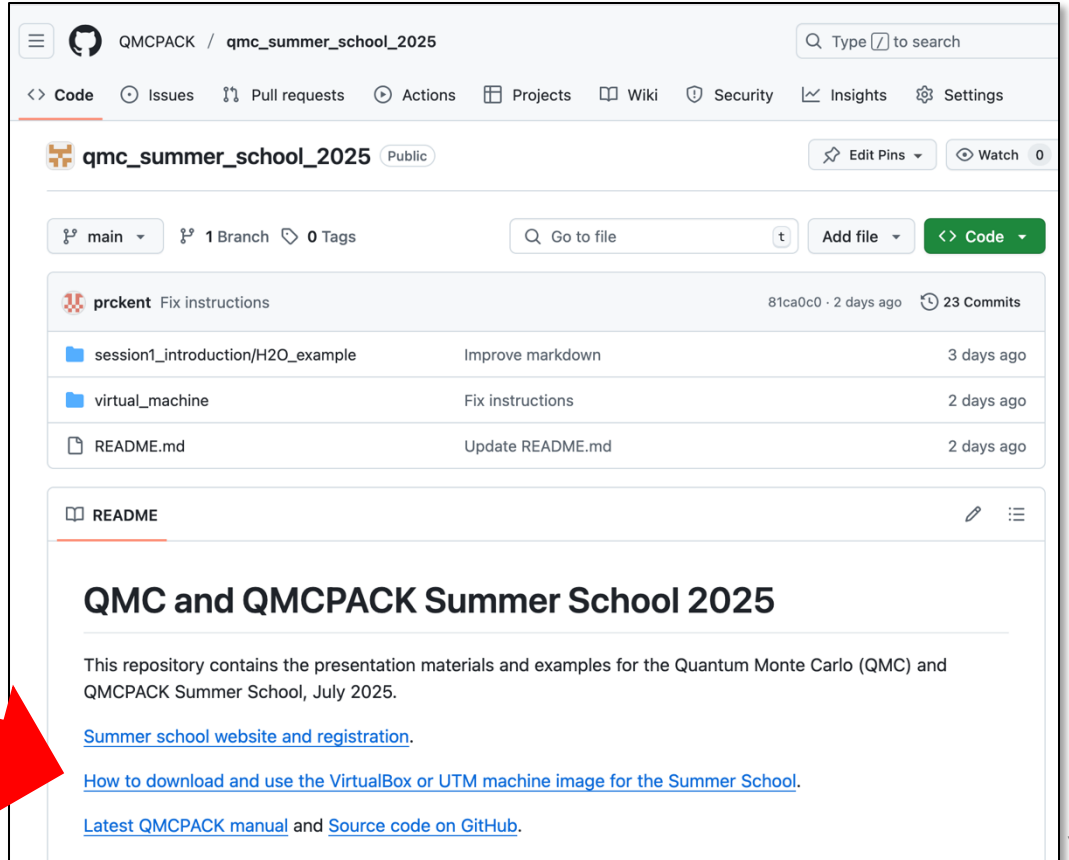
- Use VirtualBox on x86 (Windows, Linux, MacOS)
- Use UTM on Apple Silicon (MacOS)
- We assume 4 cores, 8GB memory

While the Virtual Machine is only really needed by Session 3, please try earlier so that any problems can be debugged.



Installation

Follow the instructions linked at https://github.com/QMCPACK/qmc_summer_school_2025

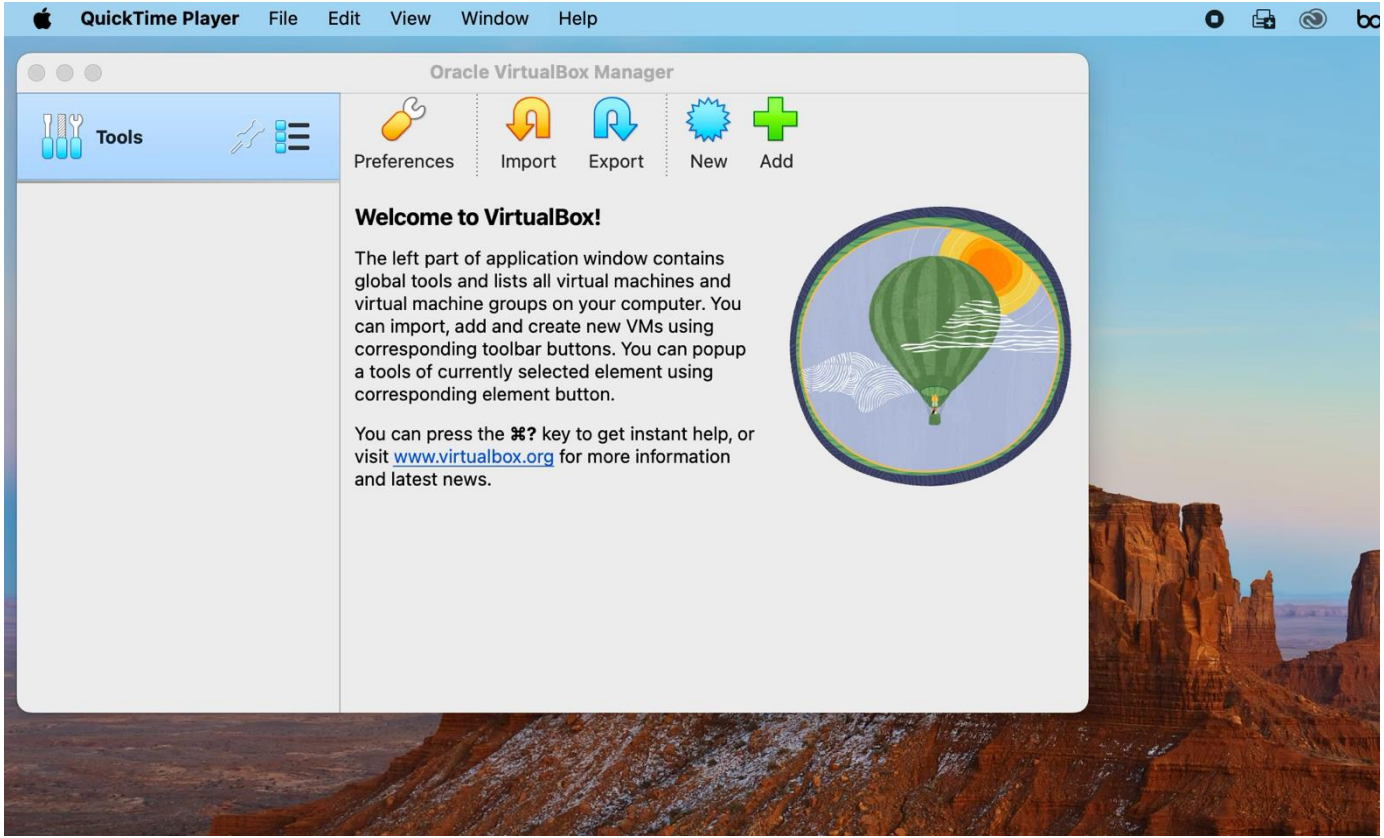


Using the Virtual Machine – Virtual Box

- Install VirtualBox (free). https://www.virtualbox.org/ or via package manager.
- Download the virtual machine image file https://bit.ly/qmcsummerschool2025vbximage (~6GB). Be patient and try downloading again at a different time or using a different network if initially unsuccessful.
- Run VirtualBox and import the image to setup the workshop virtual machine:
- In VirtualBox, select File->Import Appliance. Import QMCSummerSchool2025Image.ova.
- Customize the "Appliance" Settings during Import to suit your computer. e.g. Increase the CPU count to match the machine you are running on (maybe 4 for laptop, 16-128 for a workstation), Increase the memory (4GB should be OK on laptops). Not setting the core count and memory sufficiently large will result in poor performance.

Virtual Box Setup

Use the latest/most powerful machine you can. Customize the "Appliance" Settings during Import to suit your computer. To help performance, increase the CPU count to match the machine you are running on (maybe 4 for laptop, 16-128 for a workstation), and increase the memory.



(Movie)

Using the Workshop VM

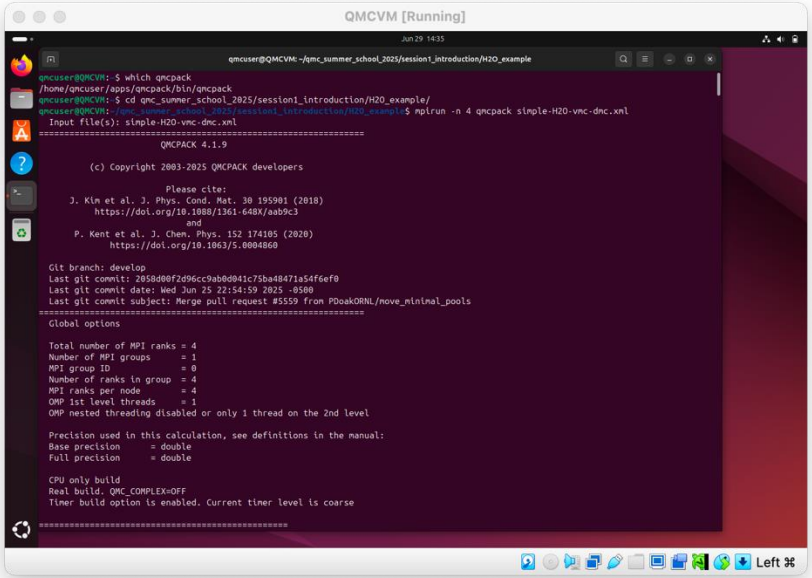
Select the workshop VM, then "Start" (green arrow). Ubuntu will boot and a standard desktop should appear.

User is "qmcuser". Should you need to authenticate the word you need is "qmc2025" (no quotes).

You can get a Terminal (or other application) by clicking the circle icon at the bottom left and either typing or selecting the application you want.

"qmcpack", "qmcpack_complex", "pw.x", are on the path. NEXUS and PySCF are available through python. These are installed in $HOME/apps

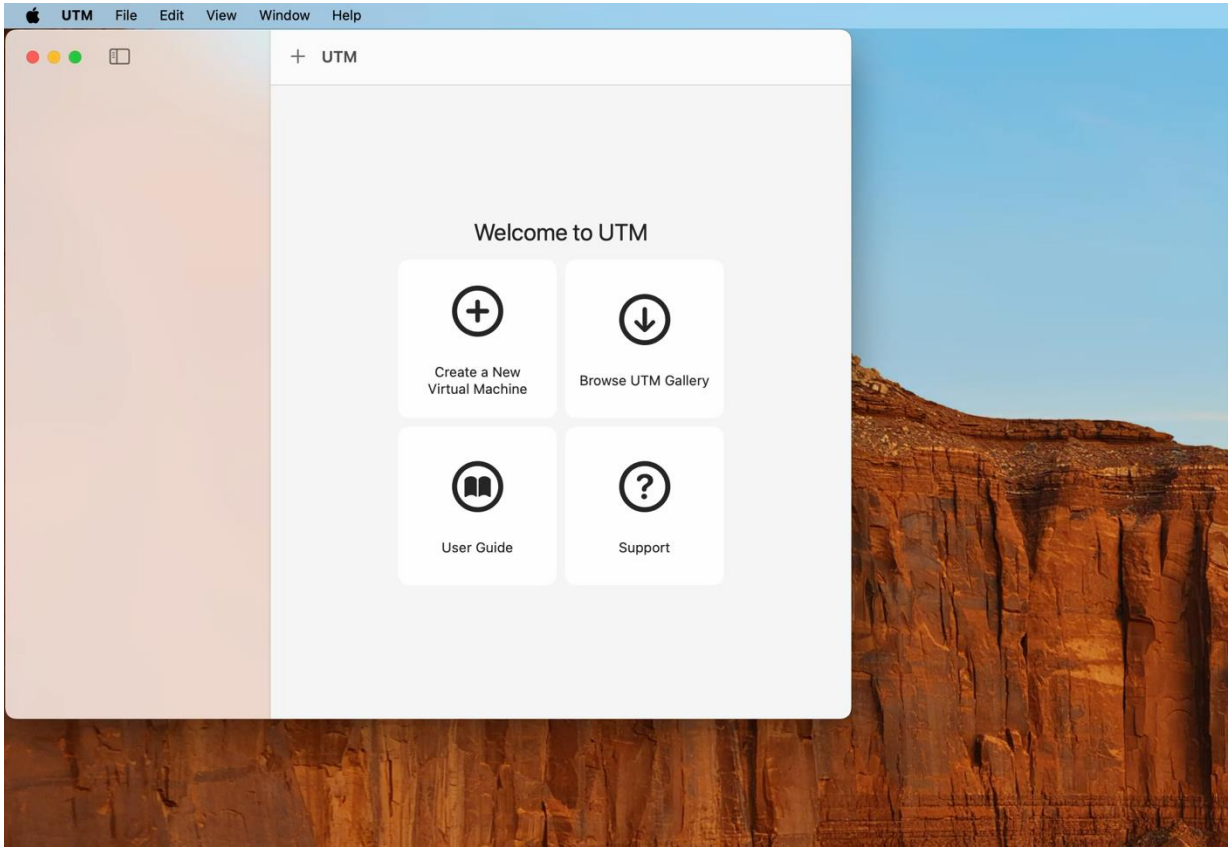
"sudo apt-get install your_favorite_editor" to install software



Using the Virtual Machine – Apple Silicon

- Install UTM (free). https://mac.getutm.app/ or support development via App Store.
- Download the workshop image file https://bit.ly/qmcsummerschool2025utmimage (~6GB). Be patient and try downloading again at a different time or using a different network if initially unsuccessful.
- Run UTM and setup the workshop virtual machine:
- In UTM, select "Create a New Virtual Machine", then "Open.." an existing image. Select QMCSummerSchool2025Image.ova.
- Edit the settings to suit your computer. e.g. Increase the CPU count to match the machine you are running on, increase the memory etc.

UTM Setup



(Movie)

Alternatives

1. Install VirtualBox on a remote workstation. Better than using a weak laptop.
2. On an Ubuntu 24+ machine, run the setup script directly.
3. On a Windows machine, use WSL2 and Ubuntu 24. Run the setup script directly.
4. Install the software directly. Adopt the setup script for other UNIXes by changing the package manager. Best route if available. Every workshop, several people choose this route and are successful.

Workshop image setup script and instructions are in https://github.com/QMCPACK/qmc_summer_school_2025/ "virtual_machine" directory. See QMCPACK installation instructions at https://qmcpack.readthedocs.io

Ask on Slack for advice!

Updating Files

Workshop example files and slides (expect regular updates):

```
cd $HOME/qmc_summer_school_2025
git pull
```

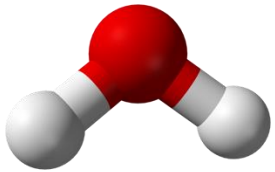
QMCPACK & NEXUS python updates:

```
cd $HOME/apps/qmcpack/qmcpack
git pull
```

We will try to minimize full rebuilds or redownloads!

Example Calculation

See qmc_summer_school_2025/session1_introduction/H2O_example



Instructions in README.md

Compute the energy of a single water molecule by VMC and DMC. O(1) minute.

```
mpirun –n 4 qmcpack simple-H2O-vmc-dmc.xml # Run

qmca -q ev --equilibration=20 --units=ev H2O.s00*.scalar.dat # Analyze

qmca -t -q e --equilibration=20 --units=ev H2O.s00*.scalar.dat # Plot trace
```

Reference output files are in “results”

Try varying the # blocks at the end of simple-H2O-vmc-dmc.xml ; Increasing the statistics by 4x should halve the error bar.

Ubuntu25Test

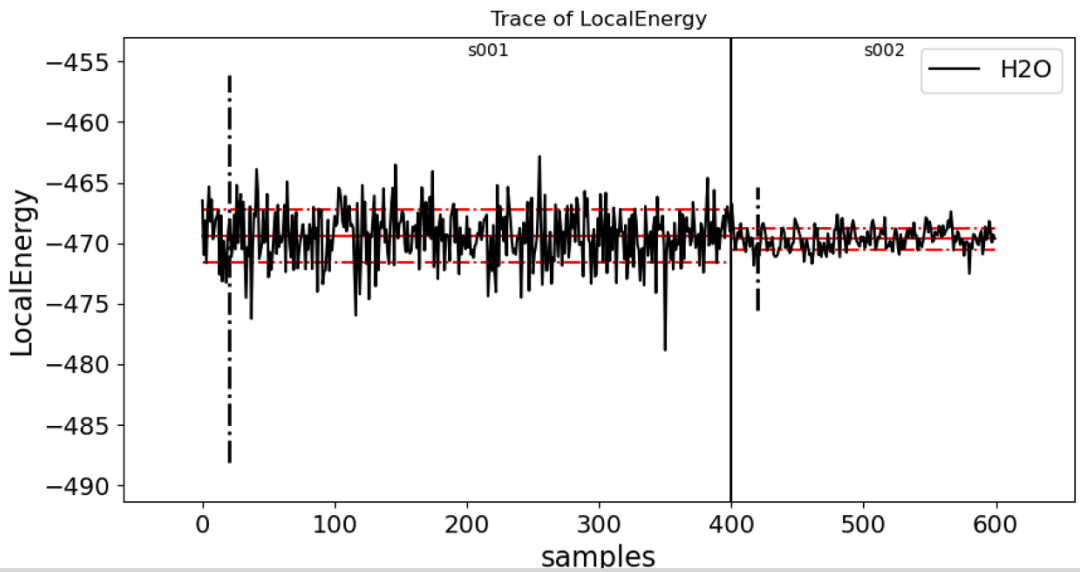
Jun 26 14:24

qmcuser@qmcvm: ~/H2O_example

```
qmcuser@qmcvm:~/H2O_example$ qmca -t -q e --equilibration=20 --units=ev H2O.s00*
.scalar.dat

H2O  series 1  LocalEnergy           =  -469.391489 +/- 0.113465
H2O  series 2  LocalEnergy           =  -469.648041 +/- 0.103980
```

Figure 1



WORKSHOP_README.txt

Home

Things to do

1. Register on Slack: https://bit.ly/qmcworkshopslack
2. Install virtual machine using instructions linked at

   https://github.com/QMCPACK/qmc_summer_school_2025/
3. Try running the water molecule example

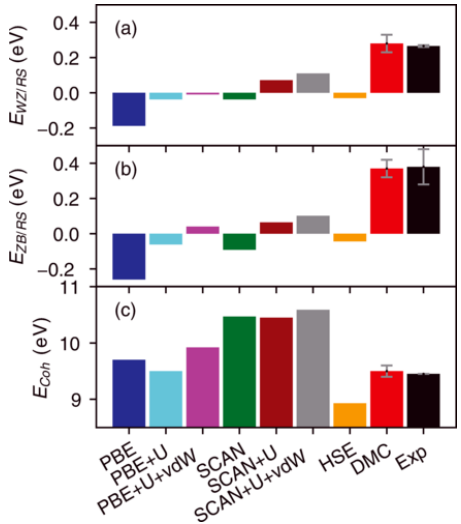
Questions?

This Thursday: Theory of real space Quantum Monte Carlo

Accuracy in standard forms

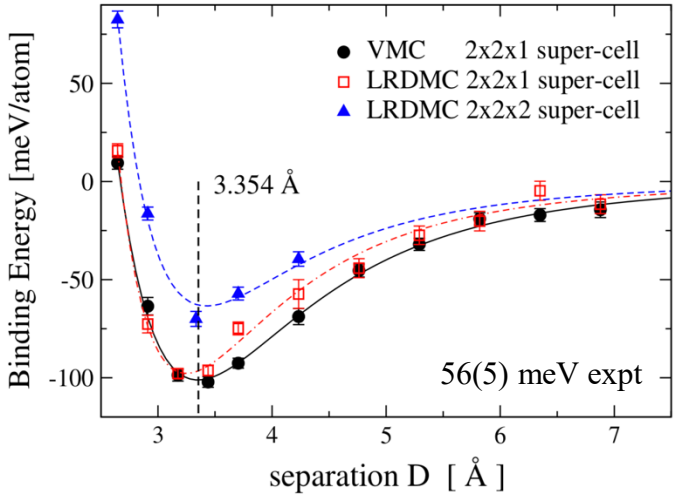
QMC methods naturally avoid double counting & self interaction errors, and accurately account for the van der Waals interaction. This avoids key problems with most density functional theory approximations.

Scandium oxide polymorphs



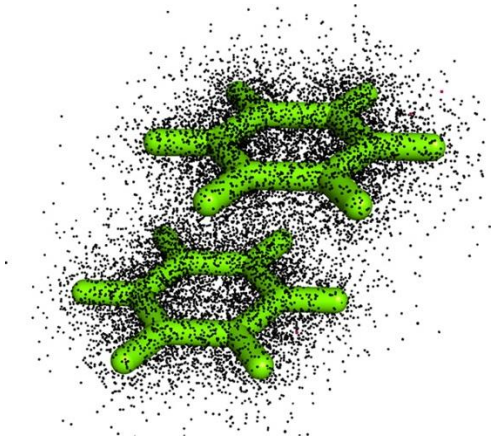
K. Saritas et al.
PRB **98** 155130 (2018)

Graphite



L. Spanu et al.
PRL **103** 196401 (2009)

Molecular Systems



M. Dubecky et al.
Chem Rev. **116** 5188 (2016)