

QMC Workshop 2021

5 October – 23 November

Week 1 Kickoff: Paul Kent, kentpr@ornl.gov

https://github.com/QMCPACK/qmc_workshop_2021

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.

Welcome

We have >200 registrants for this workshop from across US, Europe, Asia

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



2019 QMCPACK Users Workshop

Goals for Today

Motivation: Why quantum Monte Carlo?

Logistics for the workshop

Using the workshop VirtualBox image

Covering any questions

Next week: Theory of real space Quantum Monte Carlo

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 Anouar Benali @ Argonne who is watching Slack & the chat today.

Acknowledgements

Thanks to the presenters:

Chandler Bennett, Jaron Krogel @ Oak Ridge National Lab, Anouar Benali @ Argonne National Lab, Cody Melton, Joshua Townsend @ Sandia National Labs, Gani Annaberdiyev, Lubos Mitas @ North Carolina State University

Website & administrative help @ ORNL: Laura Becker, Makysha Mitchell

Current QMCPACK developers and users

Goals for Workshop

1. Cover the fundamentals of Quantum Monte Carlo
2. Provide foundation for practical molecular & solid-state calculations using QMCPACK in combination PySCF, Quantum ESPRESSO
3. Cover some new developments & recent research
4. Provide routes for follow-on support after the workshop

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

Schedule Notes

Tuesdays 11am Eastern Time for up to 2 hours, covering presentations and worked examples. Work through these and make your own experiments during each week.

“Out of hours” support via Slack.

In case of “bad network weather” we will reschedule later the same week based on speaker availability to stay on our overall schedule.

Caution! Daylight savings ends on 7 November in USA (-1hr). Check with e.g. timeanddate.com for correct start time.

Schedule

Week 1 / 5 October	Workshop Introduction and Kickoff (Paul Kent)
Week 2 / 12 October	Fundamentals of Quantum Monte Carlo (Paul Kent)
Week 3 / 19 October	Computational workflows with NEXUS. Statistical Analysis (Jaron Krogel)
Week 4 / 26 October	Molecular Calculations (Anouar Benali)
Week 5 / 2 November	Solid-State Calculations (Joshua Townsend)
Week 6 / 9 November	Pseudopotentials in QMC. Observables (Lubos Mitas, Gani Annaberdiyev, Chandler Bennett)
Week 7 / 16 November	Guest lecture and Research Presentations (Claudia Filippi and others)
Week 8 / 23 November	Spin-orbit Calculations. Workshop wrap-up and next steps (Cody Melton and Paul Kent)

QMC Workshop Slack

Short link to register: <https://bit.ly/qmcworkshopsslack>

Pick appropriate channels:

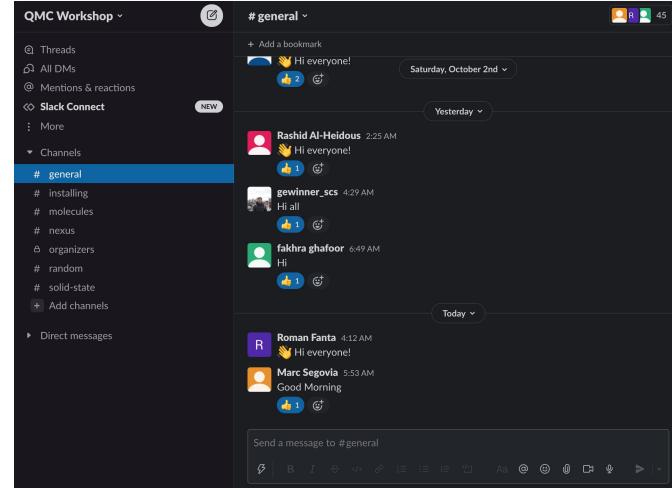
#general

#installing

#nexus

#molecules

#solid-state



We encourage an interactive workshop

Mute or leave enabled Slack notifications as you prefer

Support after the Workshop

Slack channel will remain open

Use the QMCPACK Google Group

Open an issue on QMCPACK GitHub for more technical issues

Contact us directly

This is a screenshot of the QMCPACK Google Group interface. At the top, there's a header with 'Groups' and a search bar. Below that, a pinned message from Paul R. C. Kent about the QMCPACK Workshop 2021 is shown. The main area displays a list of messages from various users, including Hung Q., Michele Rugg., zen.an..., and Andrea G. The messages cover topics like 2RDM from a AFQMC BP, problems with multideterminant wavefunction, and issues with layered materials.

This is a screenshot of the QMCPACK GitHub Issues page. The header shows 'Issues - QMCPACK/qmcpack'. Below the header, there are sections for 'Pinned issues' and '296 Open'. One pinned issue is 'Welcome' (#3368) and another is 'Tests wanted / List of untested functionality' (#1061). The main list of issues includes entries like 'Convert4qmcp crashes while processing PySCF h5' (#3503), 'BUILD_AFQMC_WITH_NCCL=ON generates undefined references' (#3500), and 'Test for expected executables in bin directory' (#3494).

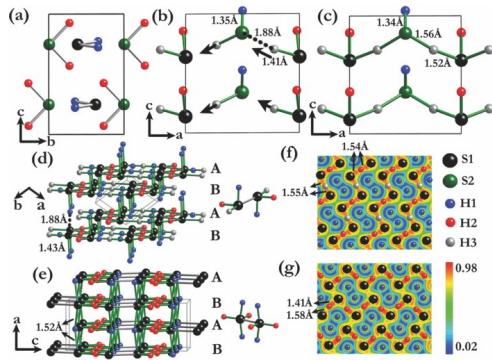
Why Quantum Monte Carlo?

Successes of Electronic Structure Methods & Quantum Chemistry

Aiding discovery of high-temperature superconductors

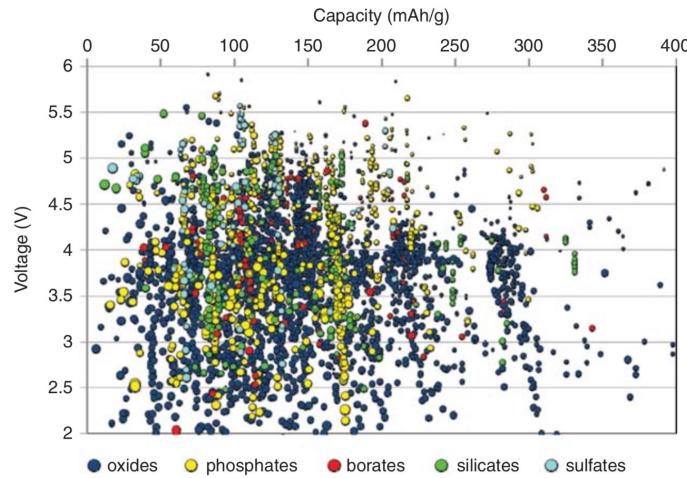


Snider et al. Nature
586 373 (2020)
A “carbonaceous sulfur hydride”



Li et al. JCP **140** 174712 (2014)
“unravels a superconducting potential of metallic H₂S”

High throughput screening of battery materials. Cheap & reliable enough to guide synthesis, and elucidate trends.
Databases, AI, ML.



Ceder MRS Bulletin (2010)

Our research uses a range of methods each with their own strengths and weaknesses: tight binding, GW, DMFT, CI, CASSCF, CCSD(T), DFT, QMC

Key Features of Quantum Monte Carlo

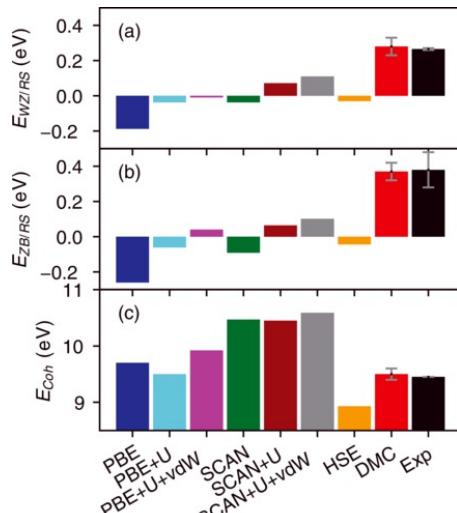
1. The methods are very accurate even in their simplest forms.
2. The few approximations made are known, testable and increasingly able to be converged.
3. The methods provide access to the full many-body wavefunction and many-body physics.
4. The methods are increasingly affordable and usable for timely and interesting science problems.

...

Accuracy in standard forms

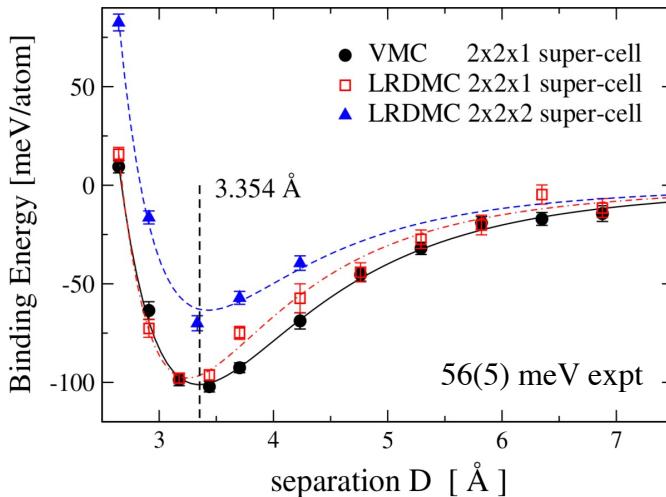
QMC methods naturally avoid double counting & self interaction errors, and accurately account for the van der Waals interaction. This avoids key problems with most density functional theory approximations.

Scandium oxide polymorphs



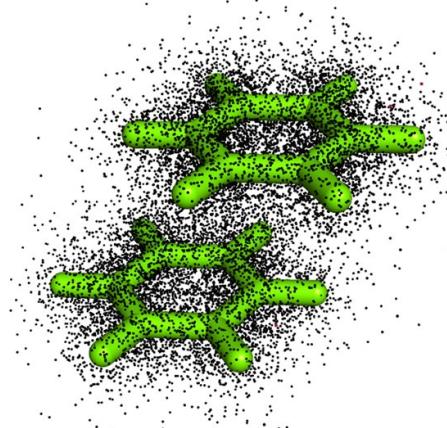
K. Saritas et al.
PRB **98** 155130 (2018)

Graphite



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

Molecular Systems



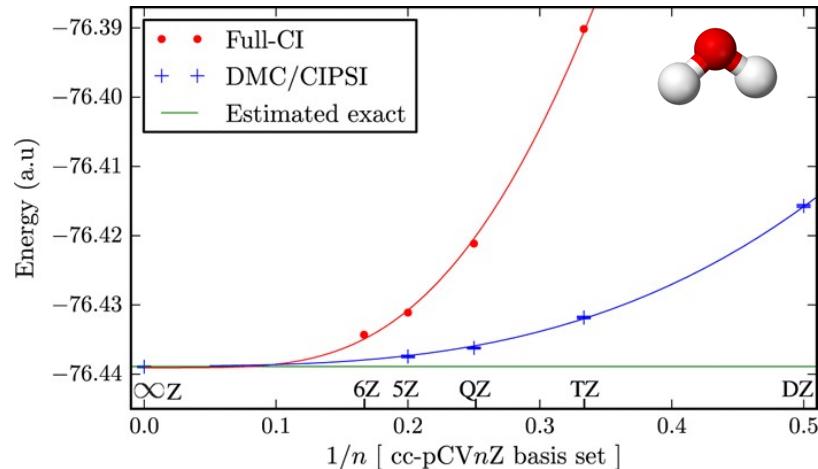
M. Dubecky et al.
Chem Rev. **116** 5188 (2016)

Improvability (rare/unique capability)

QMC methods are not “magically” accurate, and as typically applied, make a few approximations in solving the Schrodinger equation.

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

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



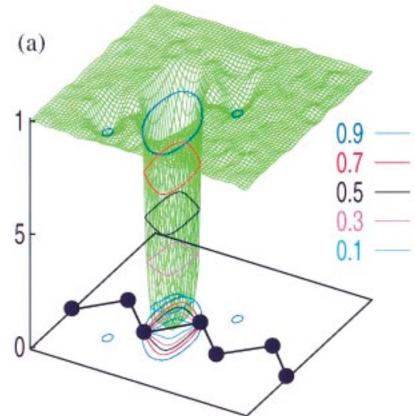
H₂O molecule
-76.438 94(12) a.u. CIPSI-DMC
vs -76.438 9 a.u. Experiment

Most accurate water calculation to-date

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

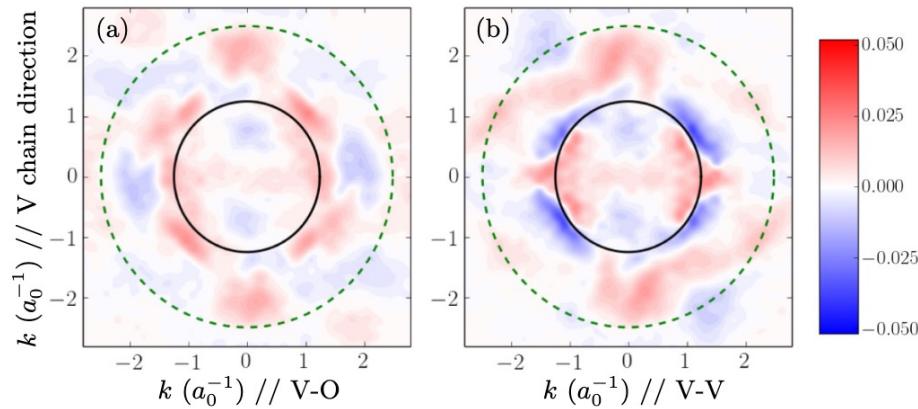
Many-body wavefunctions

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 in my opinion!



$g(\mathbf{r}, \mathbf{r}')$ spin parallel, one electron on bond center, Si (110) plane

R. Q. Hood et al. PRL **78** 3350 (1997)

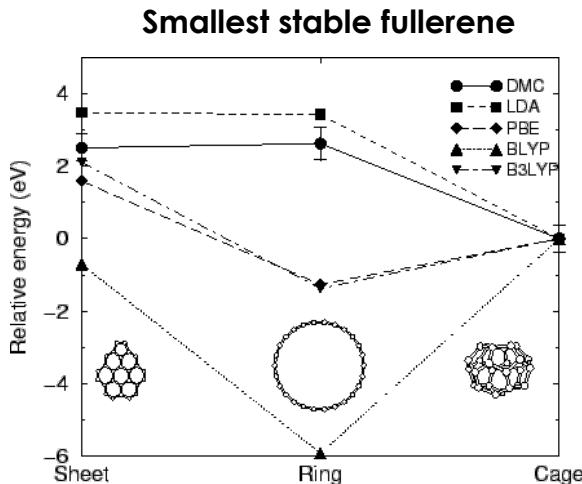


Difference in momentum distribution $n(\mathbf{k})$ in VO_2 through R-M1 phase transition

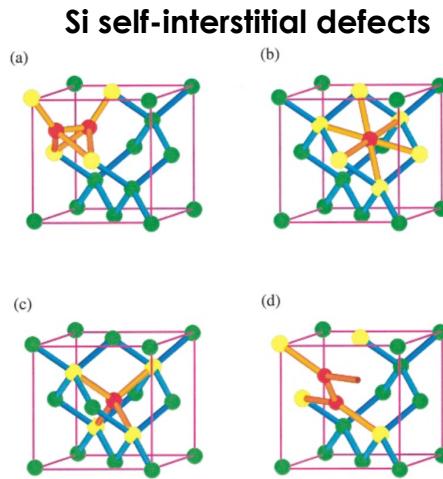
I. Kylanpaa et al. PRB **99** 075154 (2019)

Computational Cost

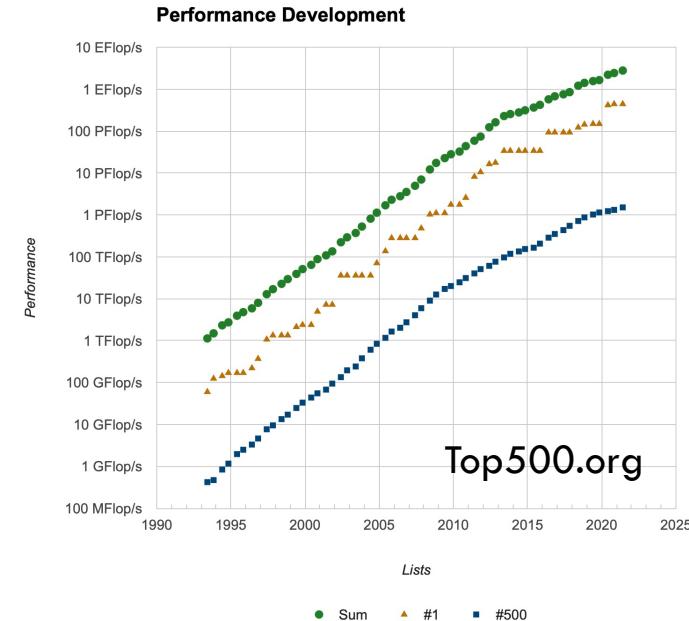
Many of the earliest-but-still-accurate nanostructure and solid-state defect calculations can now be run on workstations! Similar calculations & light element systems are in scope using departmental level resources. Heavier element systems & delicate properties still need HPC and careful planning.



P. R. C. Kent et al.
PRB **62** 15394 (2000)



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



Using the workshop Virtual Machine

Workshop Virtual Machine

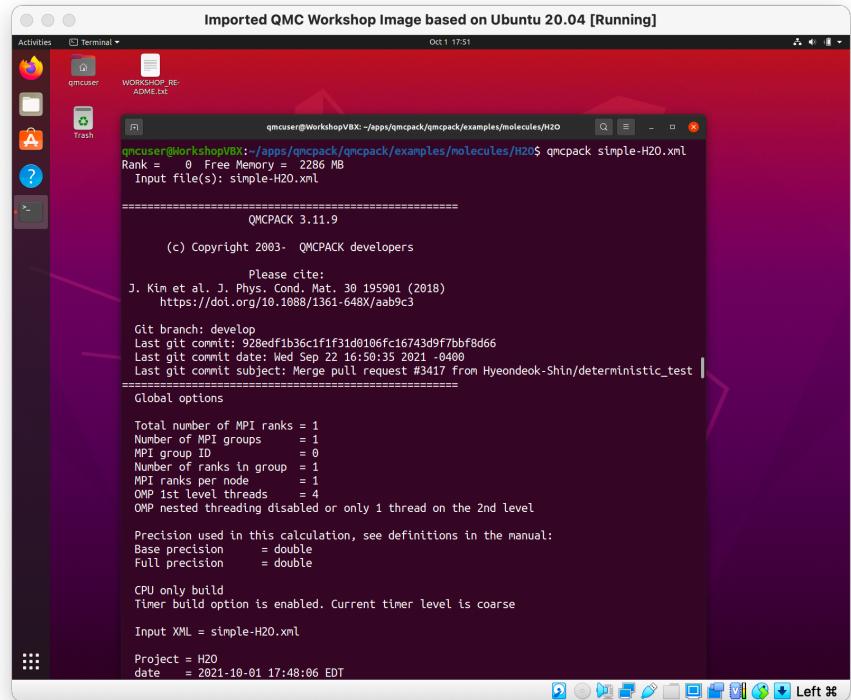
The VirtualBox Virtual Machine image is a complete desktop for running QMC, DFT and quantum chemical calculations.

The VM is a Ubuntu 20.04 desktop with QMCPACK, NEXUS, Quantum ESPRESSO (QE), PySCF, DIRAC, VESTA, and xcrysden installed, as well as the workshop files.

We support running the provided VirtualBox image on x86 architectures. Problems are sized to be runnable on a recent/strong laptop.

Workarounds & alternatives will be covered after these setup instructions.

While the Virtual Machine is only really needed by week 3, please try earlier so that any problems can be debugged. Feedback welcomed: we hope to make a live QMCPACK image going forward.

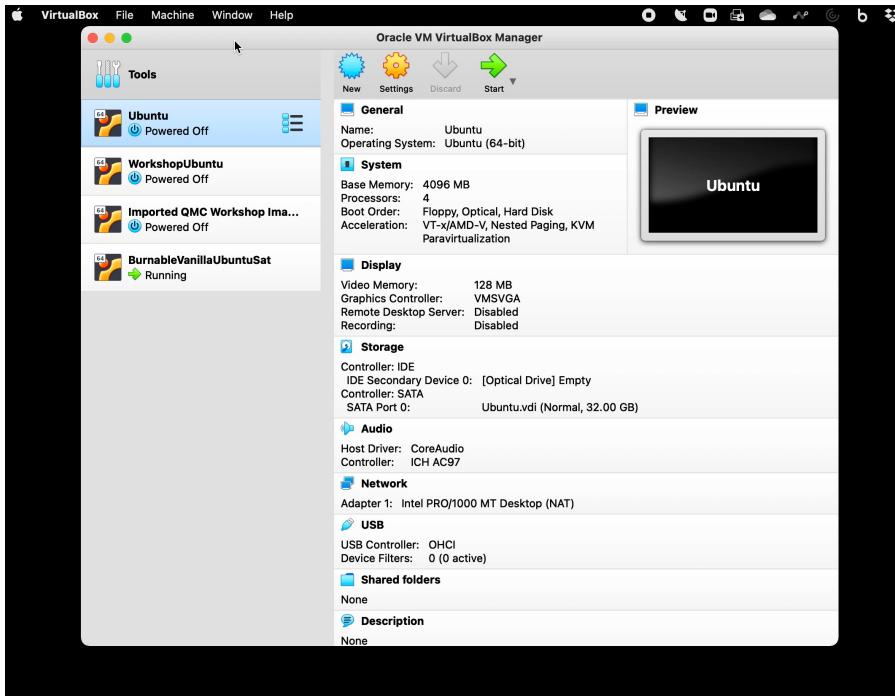


Using the Workshop VM

- Install the latest version of VirtualBox (free), currently v6.1.26. <https://www.virtualbox.org/> or package manager.
- Download the workshop image file <https://bit.ly/qmcworkshopimage>
- The file is 5GB in size, so will take several minutes to download even on the fastest networks, and potentially several hours. Be patient and try 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 the menu File->Import Appliance. Import the QMCWorkshopImage.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. Consider renaming the machine.

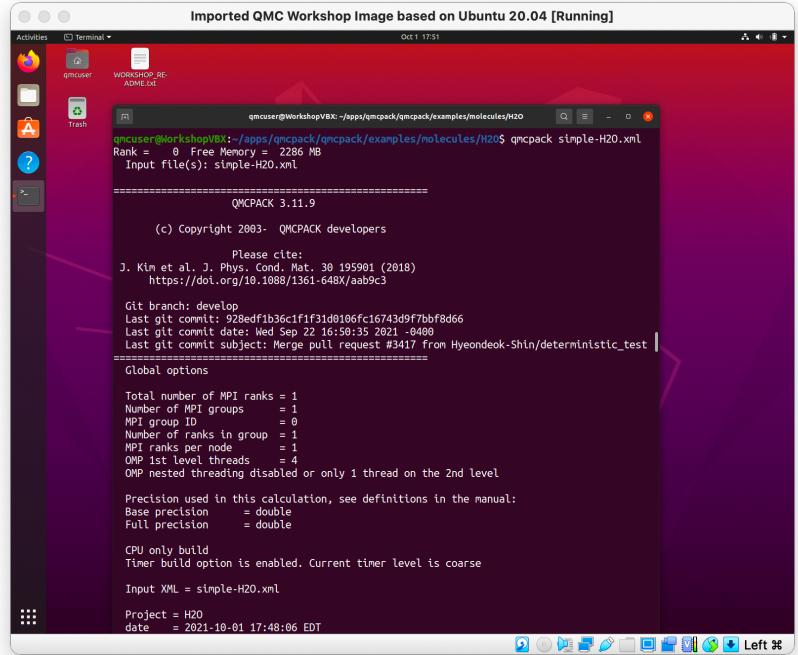
Using the Workshop VM

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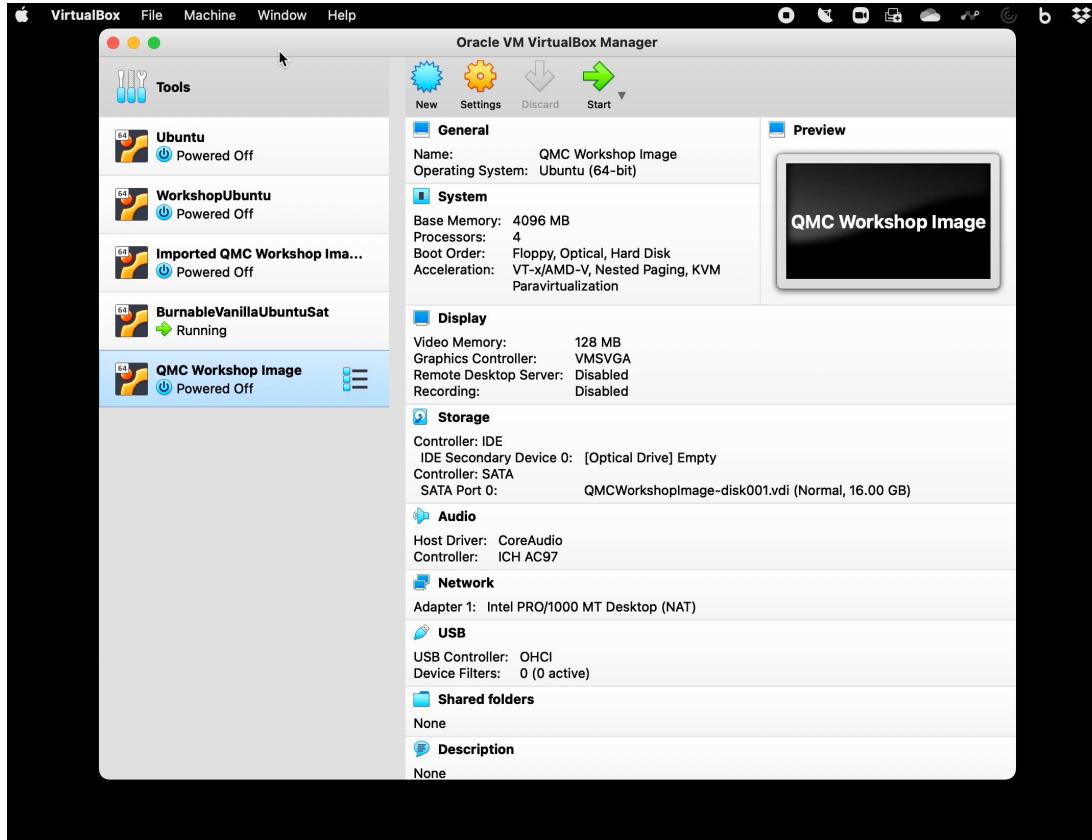


Using the Workshop VM

- 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 "workshop21" (no quotes).
- You can get a Terminal (or other application) by clicking the square of dots icon at the bottom left and either typing or selecting the application you want.
- "qmcpack", "qmcpack_complex", "pw.x", "pam-dirac", and "VESTA" 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
- Convenience tip: Right click on the Terminal icon & make a favorite to keep in the task bar.



Using the Workshop VM



Alternatives

1. Install VirtualBox on a remote workstation and access over X11. You will need root to install. Better than using a weaker laptop, highly advantageous if a powerful workstation.
2. Install the software directly: Adopt the setup script for other UNIXes by changing the package manager. Besides the simulation software, only standard Ubuntu packages are used.

Workshop image setup script and instructions are in
https://github.com/QMCPACK/qmc_workshop_2021

Updating Files

Workshop example files and slides (expect weekly updates):

```
cd $HOME/qmc_workshop_2021
```

```
git pull
```

QMCPACK & NEXUS python updates:

```
cd $HOME/apps/qmcpack/qmcpack
```

```
git pull
```

We will try to minimize full rebuilds or redownloads!

Things to do

1. Register on Slack: <https://bit.ly/qmcworkshopslack>
2. Install VirtualBox via <https://virtualbox.org> or package manager
3. Setup the workshop image <https://bit.ly/qmcworkshopimage> (5GB)

Questions?

Next week: Theory of real space Quantum Monte Carlo