

QMCPACK Users Workshop 2023

12-14 December, Argonne National Laboratory

Introduction

Paul Kent, kentpr@ornl.gov

https://github.com/QMCPACK/qmcpack_workshop_2023

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.

Outline

Workshop Goals

Meeting logistics & housekeeping

Note on tomorrow's discussion session

Building QMCPACK

Getting support

Recent developments

Contributing to QMCPACK

Questions

Workshop Goals

Hear about the latest science done with QMC

Learn about new features and upcoming code changes

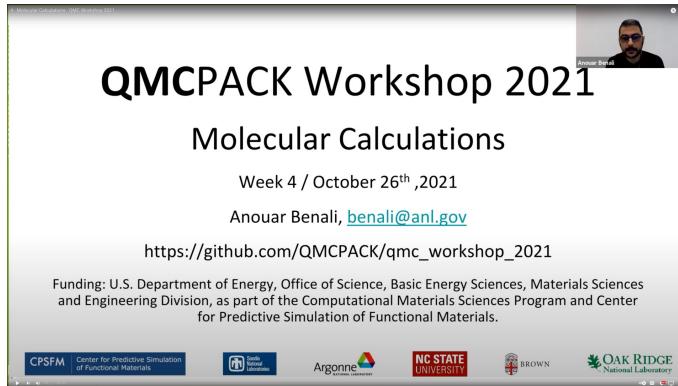
Connect with developers and users

2021 QMC Tutorial

Comprehensive introduction to QMC, molecular and solid-state calculations, statistics, workflows (etc.) with recorded tutorials on YouTube.

https://github.com/QMCPACK/qmc_workshop_2021

Includes virtual machines (x86, mac) with QMCPACK, Quantum ESPRESSO, PySCF, examples, and more, ready to run.

A screenshot of a Linux desktop environment showing a terminal window titled "Imported QMC Workshop Image based on Ubuntu 20.04 [Running]". The terminal shows the following command and output:

```
qmouser@WorkshopVMX: ~/apps/qmcpack/qmcpack/examples/molecules/H2O$ qmcpack simple-H2O.xml
Rank = 0 Free Memory = 2286 MB
Input file(s): simple-H2O.xml

=====
QMCPACK 3.11.9
(c) Copyright 2003- QMCPACK developers
Please cite:
J. Kim et al. J. Phys. Cond. Mat. 30 195901 (2018)
https://doi.org/10.1088/1361-648X/aab9c3

Git branch: develop
Last git commit: 928edfb1b36c1f1f31d0106fc16743d9f7bbf8d66
Last git commit date: Wed Sep 22 16:50:35 2021 -0400
Last git commit subject: Merge pull request #5417 from Hyondeok-Shin/deterministic_test |
```

Global options

```
Total number of MPI ranks = 1
Number of MPI groups      = 1
MPI group ID              = 0
Number of ranks in group  = 1
MPI ranks per node        = 1
OMP 1st level threads     = 4
OMP nested threading disabled or only 1 thread on the 2nd level

Precision used in this calculation, see definitions in the manual:
Base precision      = double
Full precision       = double
```

CPU only build

Agenda

Tuesday (Building 240, Room 1501)	12.00 Lunch
9.00 Welcome, Introduction, Background	13.00 Features & Progress Towards QMCPACK v4.0
Anouar Benali, ANL	Paul Kent, ORNL
Paul Kent, ORNL	Jaron Krogel, ORNL
9.50 Research Talk	Ye Luo, ANL
Andrea Zen, Universita di Napoli, Italy	14.45 Break
10.35 Break	15.00 Poster session and open collaboration session to test batched code, discuss with presenters & developers
10.50 Research Talk	17.15 Bus to guest house
Fernando Reboreda, ORNL	
11.35 Short Research Talk	
David Ceperley, UIUC	

Agenda

Wednesday (Building 240, Room 1501)

9.00 All participant discussion on improving capabilities and scientific productivity of QMCPACK

14.40 Orbital Optimization

Joshua Townsend, SNL

10.30 Group Photo

Amanda Dumi, SNL

10.45 Break

15.10 Surrogate Hessian for Geometry Optimization

11.00 Research Talk

Jaron Krogel, ORNL

Daniel Wines, NIST

15.30 Fermi Surface Calculation

11.50 Lunch

Jaron Krogel, ORNL

13.00 Spin-orbit coupling in QMCPACK

16.00 Open collaboration session: 1-1 discussion, help with projects

Cody Melton, SNL

17.30 Bus to guest house

13.50 New correlation consistent ECPs

18.30 Dinner at "Bar Louie" (Car Pool, <10 miles)

Benjamin Kincaid, NCSU

14.20 Break

Agenda

Thursday (Building 241, Room D172)

8.55 Best Poster Award Announcement

9.00 "Using Quantum Monte Carlo to understand hydrogen storage in metal decorated graphene" - Yasmine Al-Hamadani, UCL

9.20 "VMC Wave Function Optimization for Excited States in Molecules and Solids" - Leon Otis, University Of Chicago

9.40 "Force-free identification of minimum-energy pathways and transition states using quantum Monte Carlo methods" - Gopal Iyer, Brown U.

10.00 Break

10.15 "Modeling Correlated Two-Dimensional Magnets with QMC"- Can Ataca, University of Maryland, Baltimore County

10.35 "Enhanced Twist-Averaging Technique for Magnetic Metals: Applications using Quantum Monte Carlo" - Gani Annaberdiyev, ORNL

10.55 Open collaboration session

12.00 Workshop ends

Zoom Practicalities

We are recording these presentations (with permission) for upload to YouTube

Questions and interaction improves the talks!

Keep muted when not presenting or asking a question

Participants should be able to unmute

Be careful not to unmute by accident!

Poster Session & Prize

Thanks to the presenters

Poster session is this afternoon ~3pm+

Look out for voting email

Winner announced Thursday

Tomorrow: Open Discussion on Improving Capabilities and Productivity of QMCPACK

How could we improve QMCPACK to most help your research?

What would you like QMCPACK to do in 6 months? 1 year?

What is the top thing we need to fix?

...

To aid a full discussion, this session will not be recorded.

Possible topics

Science Capabilities

Ease of Use

Installation

Workflows

Support for different sources of trial wavefunction

Documentation & Examples

Development questions

Introductory Tutorials

Onboarding New Users

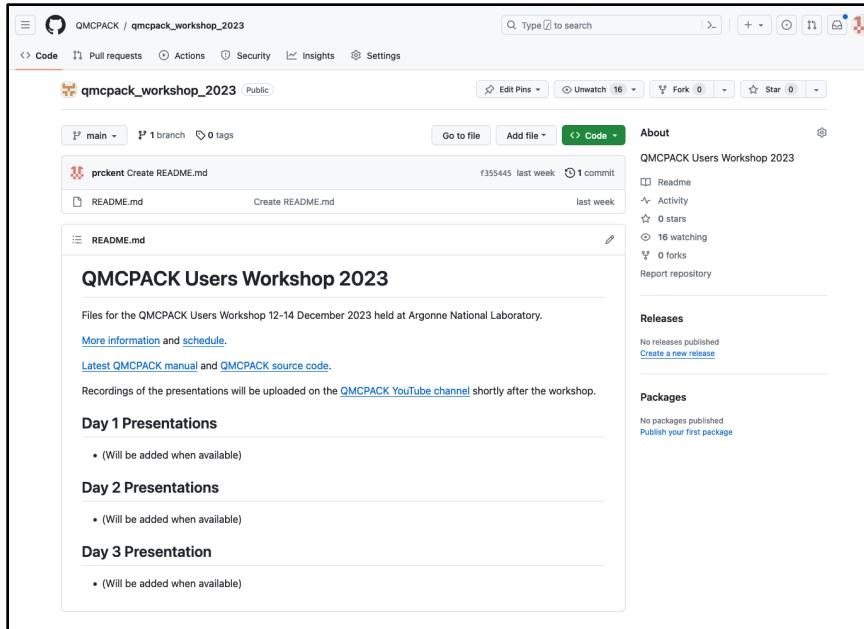
Support in General

The success of this session depends on you!
Help determine the direction of QMCPACK

Workshop GitHub Repository

https://github.com/QMCPACK/qmcpack_workshop_2023

Will have examples, data files, presentations, and links to recordings as they become available.



Building QMCPACK

For the development version:

```
git clone https://github.com/QMCPACK/qmcpack.git
```

```
cd qmcpack/build
```

```
cmake -DCMAKE_C_COMPILER=mpicc -DCMAKE_CXX_COMPILER=mpicxx ..
```

```
make -j 16; ctest -j 16 -R deterministic
```

More details and examples at

<https://qmcpack.readthedocs.io/en/develop/installation.html> and in config directory

Support

Open an issue on QMCPACK GitHub (preferred)

Use the QMCPACK Google Group

Contact us directly – talk to or email a developer

This is intended as a mailing list for QMCPACK developers and users. The list is focused on electronic structure and QMC-related research.

Before posting your questions, please check <http://qmcpack.org> and the discussion of this group. A current list of the major known issues, bugs, and requests is on GitHub <https://github.com/QMCPACK/qmcpack/issues>. Please comment there to "upvote" topics that are most important to you.p

As an anti-spam measure, this is a moderated list. You can become a member of the group either by attempting to post or by sending an email to this group: qmcpack@googlegroups.com

Paul R. C. Kent QMC Workshop 2021 - Starting in October - Registration Open - Full details ... Aug 26

Hung Q. ..., fionn m... 2RDM from a AFQMC BP - This is from the output. I used the source code ob... Jun 29

Michele Rugg..., Ye L... 5 Problem with multideterminant wavefunction - Hi Michele, I made a fix to th...

zen.an...@gmail.com 2 delay_rank on qmcpack-GPU - I repeated the tests and I found that delay_ran...

zen.an...., j.k.r...@... 4 Issues with layered materials - I wonder if it is possible to always write the c...

zen.an...@gmail.com, Ye L... 4 Using Legacy_GPU with an open system - Hi Ye, many thanks, I will test it. B...

Andrea G., fionn m... 3 How to invoke THC implementation? - Dear Fionn, thanks a lot for the compl...

QMCPACK / qmcpack Public

Issues 296

Pinned issues

- Welcome #3368 opened on Aug 18 by prkent
- Tests wanted / List of untested functionality #1061 opened on Sep 11, 2018 by jtkrogel

Filters Labels 26 Milestones 2 New issue

Author	Label	Projects	Milestones	Assignee	Sort
prkent					
jtkrogel					
quantumstevie					
cricket					

296 Open 739 Closed

Convert4qmcp crashes while processing PySCF h5 bug #3503 opened 16 hours ago by prkent

BUILD_AFQMC_WITH_NCCL=ON generates undefined references #3500 opened 2 days ago by quantumstevie

Test for expected executables in bin directory enhancement testing #3494 opened 4 days ago by cricket

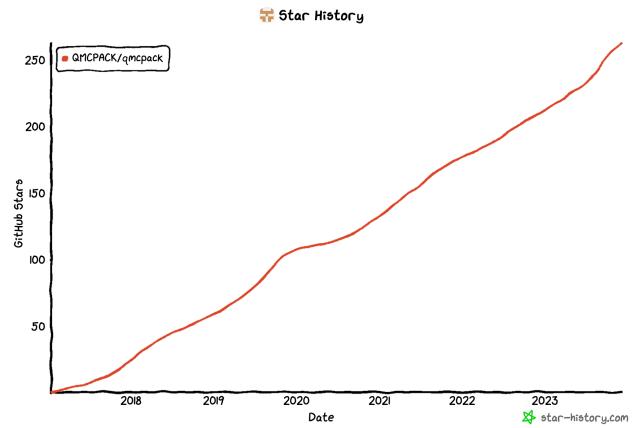
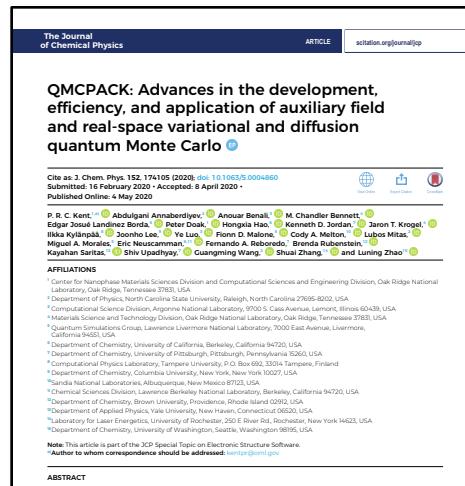
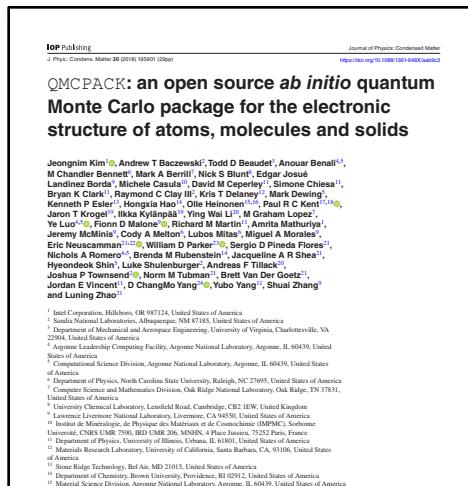
op 2023

Citing QMCPACK

Please cite the following and star the GitHub repo! Citations help funding!

J. Kim et al. J. Phys. Cond. Mat. **30** 195901 (2018),
<https://doi.org/10.1088/1361-648X/aab9c3>

P. Kent et al. J. Chem. Phys. **152** 174105 (2020),
<https://doi.org/10.1063/5.0004860>



QMCPACK Users Workshop 2023

Recent QMC Applications using QMCPACK

Crl₃ Staros JCP **156** 014707 (2022), H phase diagram Niu PRL **130** 076102 (2023), >10³ molecules Huang JCTC **19** 1711 (2023).

The Journal
of Chemical Physics

ARTICLE

scitation.org/journal/jcp

A combined first principles study of the structural, magnetic, and phonon properties of monolayer CrI₃

Cite as: J. Chem. Phys. 156, 014707 (2022); doi: 10.1063/5.0074848

Submitted: 11 October 2021 • Accepted: 7 December 2021 •

Published Online: 7 January 2022

Daniel Stares,^a Guoxiang Hu,^b Juha Tiilinen,^b Ravindra Nanguneri,^c Jaron Krogel,^c M. Chandler Bennett,^c Olli Heinonen,^{a,c} Panchakakesan Ganesh,^{a,c} and Brenda Rubenstein^{a,c}

AFFILIATIONS

^a Department of C
Department of C
Center for Nano
Materials Science
Materials Science
Northwestern Uni

Note: This paper is
"Authors to whom"

ABSTRACT

The first magnetic, However, because, simulated, recently developed parameter in good spin moments are accurate than previous DFT a liquid suggests that ML key properties, materials.

Published under an

FIG. 1. Geometry of monolayer CrI₃ cleaved from the bulk structure reported in Ref. 45. (a) Top view depicting a lattice constant of $a_0 = 6.67 \text{ \AA}$ and the bond angle θ_1 and θ_2 computed in this work. (b) Side view depicting the Cr-I bond distance of 2.728 \AA (purple) and the Cr-I-Cr bond distance of 3.965 \AA (blue).

2D materials as research; Despite tend to exhibit that the monolayer may give rise to some new properties, phase behavior, including Moiré patterns, two-dimensional superconductivity,

At cooling rate development in monolayer is the discovery of new magnetic 2D materials.²¹ While the Mermin-Wagner theorem^{22,23} prohibits finite-temperature

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C20B, USA
jcp.sciencedirect.com

state ferromagnetism CrI₃'s structural, electronic, Monte Carlo (DMC). CrI₃ exhibits a curvy, latticing lattice's variability in previous ML CrI₃'s magnetic revealed by more than one both theory¹³ and other¹⁴ suggested in its 2D limit. We many of ML CrI₃'s magnetic and other 2D

and just by stacking.¹⁵⁻¹⁸ This versatility may be due to the arrangement and ordering of their

I. INTRODUCTORY

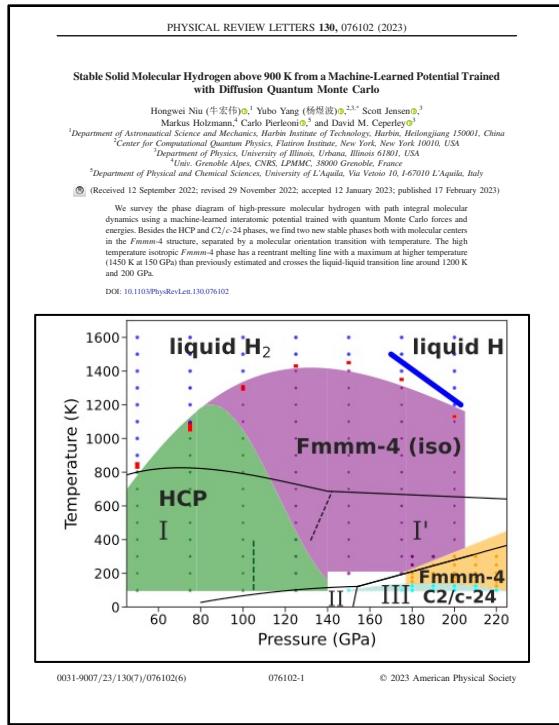
2D materials as research; Despite tend to exhibit that the monolayer may give rise to some new properties, phase behavior, including Moiré patterns, two-dimensional superconductivity,

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J. Chem. Phys. 156, 014707 (2022); doi: 10.1063/5.0074848

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156, 014707-1



JCTC Journal of Chemical Theory and Computation

pubs.acs.org/JCTC Article

Toward DMC Accuracy Across Chemical Space with Scalable Δ -QML

Bing Huang,^a O. Anatole von Lilienfeld,^a Jaron T. Krogel,^a and Anouar Benali^b

Cite This: https://doi.org/10.1021/acs.jctc.2c01058 Read Online

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ABSTRACT: In the past decade, quantum diffusion Monte Carlo (DMC) has been demonstrated to successfully predict the energetics and properties of a wide range of molecules and materials by numerically solving the electronic many-body Schrödinger equation. While DMC is considered the gold standard for QM calculations, it is also a reference method for larger systems that are not accessible to more traditional methods such as CCSD(T). Assessing the accuracy of DMC for smaller molecules becomes the stepping stone in making the method a reference for larger systems. We show that by combining quantum machine learning (QML) and DMC calculations, the computational burden can be alleviated such that quantum Monte Carlo (QMC) shows clear potential to undergo the formation of high-quality descriptions across chemical space. By using a QML model to predict the energy of molecules with this fixed-node approximation, universal and accurate set-based QML (AQML) models were generated. These AQML models were able to predict the energy of small organic molecules with up to five heavy atoms cited as among the most difficult to validate the QMC predictions. Numerical evidence collected for Δ -AQML models in training data sets of amino acids to predict total energies with near chemical space.

INTRODUCTION

The predictive power of quantum machine learning (QML) models trained on quantum chemistry data and used for the navigation of chemical compound space (CCS) is inherently limited by the predictive accuracy of the approximations used to calculate the underlying quantum theory. Consequently, in order for QML models to be truly competitive, they must achieve accuracy (~ 1 kcal/mol average deviation of calculated values from experiments or measurements of atomization energies), it is necessary to rely on training datasets generated at the best possible Hartree-Fock level (e.g., CCSD(T)) to ensure that the "gold standard" in the field, CCSD(T)/CC, generally impossible to obtain computationally, is met before predicting and scaling up QMC (whereas the quality of the training generation of large-quality quantum data sets has remained elusive, even for relatively small organic molecules with only four or five "heavy" (second-row) atoms). Here we demonstrate for an example of a dipeptide that the predictive power and usefulness of recent implemented and numerically more efficient quantum Monte Carlo (QMC) models for computing QM properties can be greatly improved by combining one of the approximations used in the method, setting the foundation for study of larger databases. Our numerical evidence indicates the possibility to trainfully route QM models that achieve predictive power similar to QMC but at much reduced computational cost.

QMC approaches solve the many-body electronic Schrödinger equation stochastically. QMC is general and applicable to

a wide range of physical and chemical systems in any dimension, boundary conditions etc. Among the most widely used fates for electronic structure are variational Monte Carlo (VQC)^{1–4} and diffusion Monte Carlo (DMC).^{5–7} Both VQC and DMC are variational methods and allow the energy and properties of a given system to be calculated by performing a stochastic computation of the matrix elements, posing no restriction on its functional form. Using the VQC algorithm, through a stochastic many-body integration scheme, the expected value of the energy for a given set of basis functions can be obtained by averaging the local energy over an ensemble of configurations distributed as w_i , sampled during a random walk in the configuration space. The fluctuations of the energy depend on the quality of the trial wavefunction, and they are the exact weight factors in a user's (azimuthal) principle. The DMC algorithm is very similar to the sampling technique beyond the distribution function by solving the Schrödinger equation in imaginary time $\tau = i$ using a projector or Green's function-based method. Any initial state $|\psi_0\rangle$ that is not orthogonal to the ground state $|\psi_0\rangle$ will evolve to the ground state in the long time limit.

Received: October 25, 2022

^a 2022 Elsevier Inc. All rights reserved. This article is an open access publication. Published by Elsevier Inc. on behalf of the American Chemical Society.

^b https://doi.org/10.1021/acs.jctc.2c01058 J. Chem. Theory Comput. XXXX, XXX, XXX–XXX

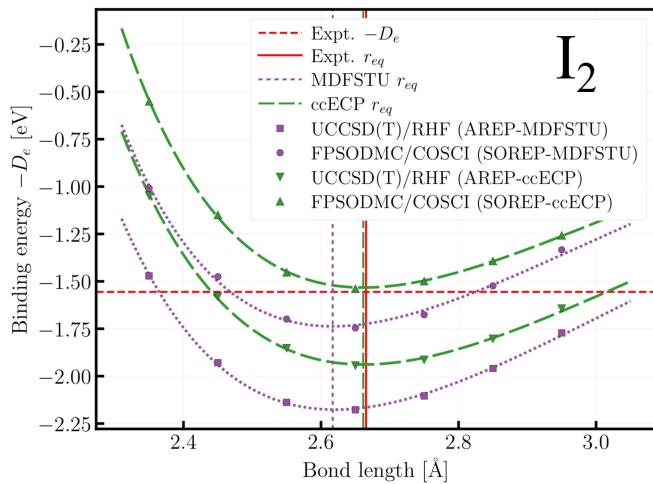
Pseudopotentiallibrary.org

See Benjamin Kincaids's talk Wednesday. Many new ccECPs added including softer ECPs. Requests taken. It is still a lot of work to create & test each ccECP, quantum chemistry and QE formats etc. Please cite the appropriate papers.

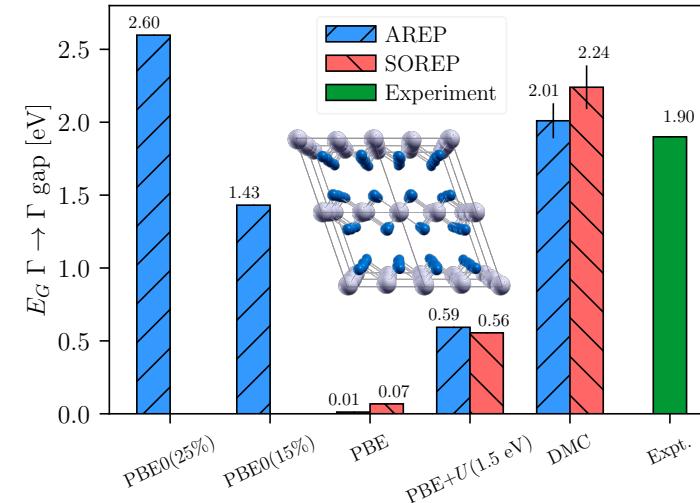
A screenshot of a web browser displaying the Pseudopotential Library website. The page title is "Pseudopotential Library". A subtitle below it reads: "A community website for pseudopotentials/effective core potentials developed for high accuracy correlated many-body methods such as quantum Monte Carlo and quantum chemistry." The main feature is a large periodic table grid where each element is represented by a colored square containing its symbol. The colors follow a gradient: green for Hydrogen (H), yellow for the first two groups (Li, Be, Na, Mg), orange for the next two groups (K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn), red for the transition metals (Pd, Ag, Cd, In, Sn, Sb, Te, I), blue for the next two groups (B, C, N, O, F, Ne, Al, Si, P, S, Cl, Ar, Ge, As, Se, Br, Kr), purple for the next two groups (Rb, Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Cs, Ba, La, Hf, Ta, W, Re, Os, Ir, Pt, Au, Hg, Tl, Pb, Bi, Po, At, Rn, Fr, Ra, Ac, Rf, Db, Sg, Bh, Hs, Mt, Ds, Rg, Cn, Nh, Fl, Mc, Lv, Ts, Og), and gold for the lanthanides (Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu) and actinides (Th, Pa, U, Np, Pu, Am, Cm, Bk, Cf, Es, Fm, Md, No, Lr). Below the table, a footer note says: "Click an element above to view available recipes."

Relativistic effects & Spin-orbit

See Cody Melton's talk on Wednesday. Spin-orbit, spinor wavefunction support, some observables for both LCAO and spline wavefunctions.



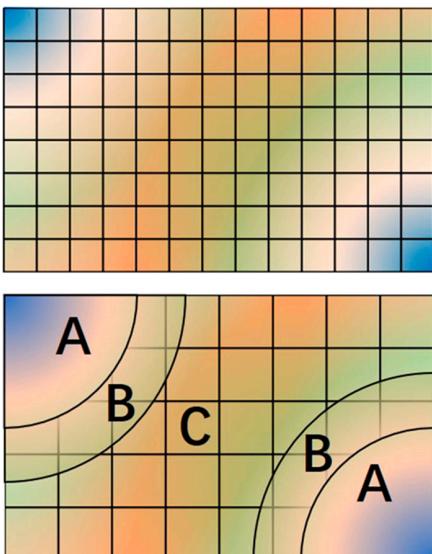
SO DMC binding energy & bond length are most accurate compared to experiment.
G. Wang et al. JCP **157** 054101 (2022)



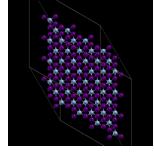
Role of SO on gap of RuCl₃
A. Annaberdiyev et al. PRB **106** 075127 (2022)

Hybrid Basis Set

Modified augmented-plane wave style representation to reduce grid density needed for spline wavefunctions. LARGE memory savings for hard ccECPs and for materials with heavier elements. Can be the difference between running or not.
Ye Luo et al. JCP **149** 084107 (2018).

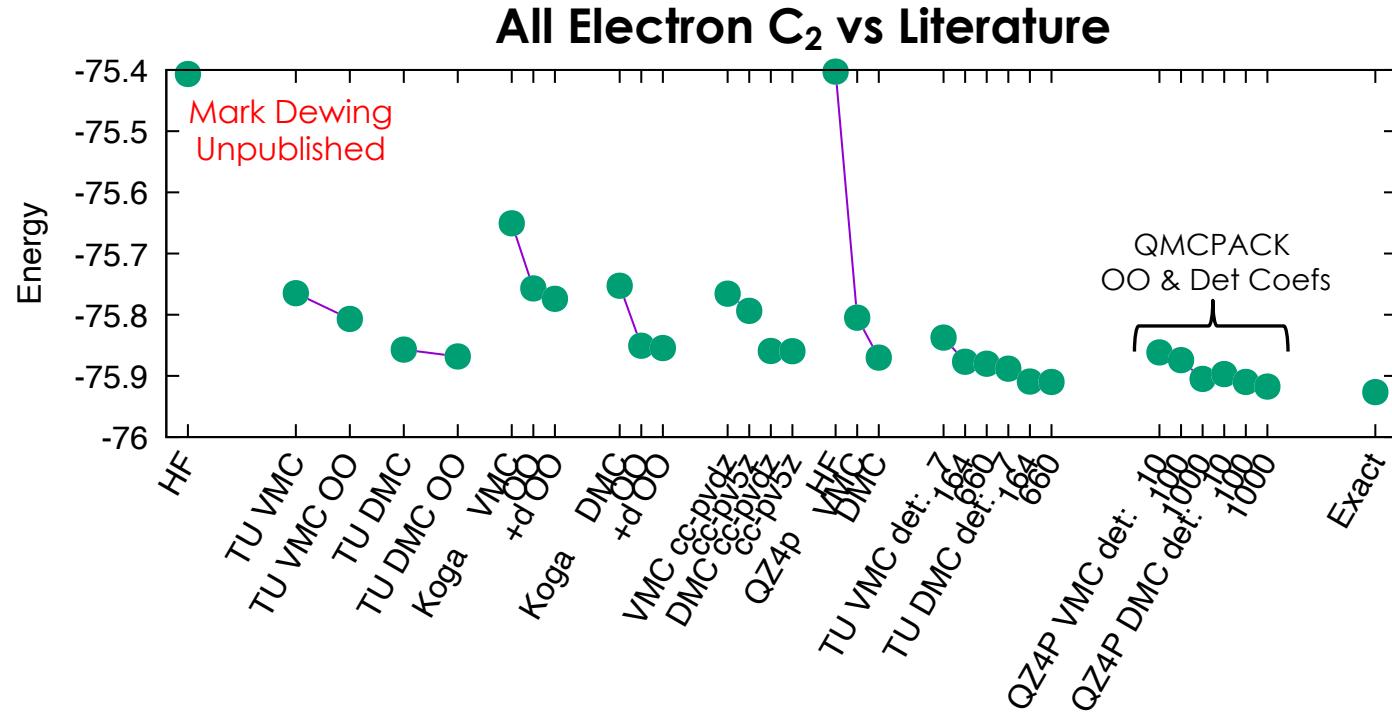


CrI_3 Monolayer Analysis (Saritas & Krogel)

	# elec	# atoms	Std. spline mem	Hybrid basis mem	Walker mem
2x2x1	560	64	176 GB	22 GB	19 MB
3x3x1	1260	144	396 GB	49 GB	95 MB
4x4x1	2240	256	704 GB	88 GB	298 MB

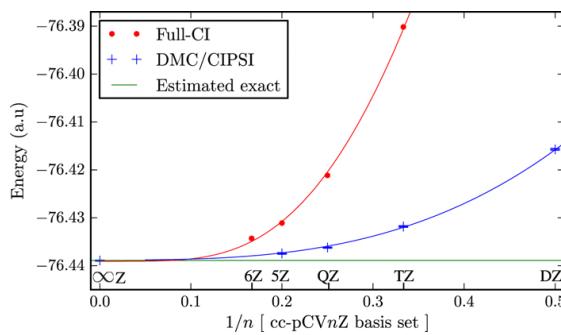
Orbital Optimization

See presentation by Joshua Townsend & Amanda Dumi, Wednesday. Reduction of nodal error for LCAO and spline wavefunctions (in process). Matches literature values and behaviors for molecular tests, single and multideterminant. **Requires latest development version.**

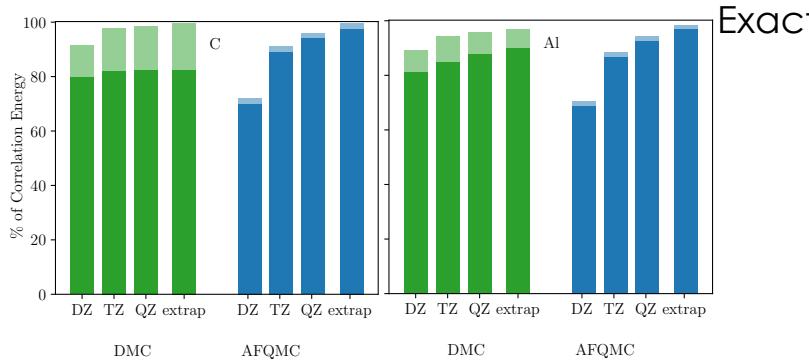


Selected Configuration Interaction

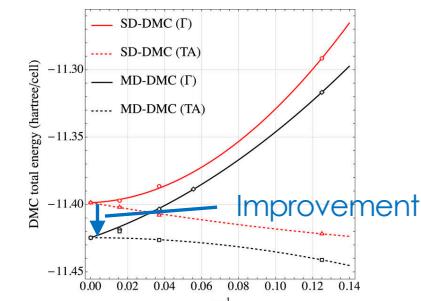
Discuss with Anouar Benali. **Systematic and deterministic reduction** of nodal error via large configuration interaction expansions. Workflow via Quantum Package both for molecules and solid-state. Many efficiency improvements in LCAO and multideterminants recently. **Low memory usage periodic LCAO available via PySCF trial wavefunctions.**



H₂O molecule
-76.438 94(12) a.u. CIPSI-DMC
vs -76.438 9 a.u. Experiment
Caffarel et al. JCP **144** 151103 (2016)



Convergence of C, Al, LiF, LiH solids (1x1x1).
DMC and AFQMC for single (dark bar) and
multideterminants (light bar). Malone et al.
PRB RC **102** 161104 (2020).



Thermodynamic limit of
C diamond
Benali et al. JCP **153**
184111 (2021)

Contributing

Contributions of any size are welcome

Make a pull request on GitHub

Small code updates are very welcome – e.g. tidy the output

Small documentation updates are very welcome – fix links, help with v4.0 transition

For feature contributions, we recommend to make a GitHub issue or discuss with a developer. Read the developer docs!

Contributors have the opportunity for coauthorship on the next citation paper

Releases & Recent Updates

Currently plan to continue at 3-4 releases per year

Fast development: ~334 merged pull requests in last 12 months

In general, we can release a new version on request

See CHANGELOG.md for list of notable changes, e.g. improved 2D support in v3.16.0, Paul Yang

Change Log

Notable changes to QMCPACK are documented in this file.

[Unreleased]

- Support for backflow optimization has been removed as part of refactoring and cleaning the codebase. QMC runs using backflow wavefunctions are still supported. This feature is expected to eventually be reimplemented in v4. Users needing backflow optimization can use previously released versions of QMCPACK or work towards its reimplementations in the modern code. [#4688](#)

[3.17.1] - 2023-08-25

This minor release is recommended for all users and includes a couple of build fixes and a NEXUS improvement.

- Improved HDF5 detection. Fixes cases where HDF5 was not identified by CMake, including on FreeBSD (thanks @yurivict for the report). [#4708](#)
- Fix for building with BUILD_UNIT_TESTS=OFF. [#4709](#)
- Add timer for orbital rotations. [#4706](#)

NEXUS

- NEXUS: Support for spinor inputs. [#4707](#)

[3.17.0] - 2023-08-18

Questions?

