

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

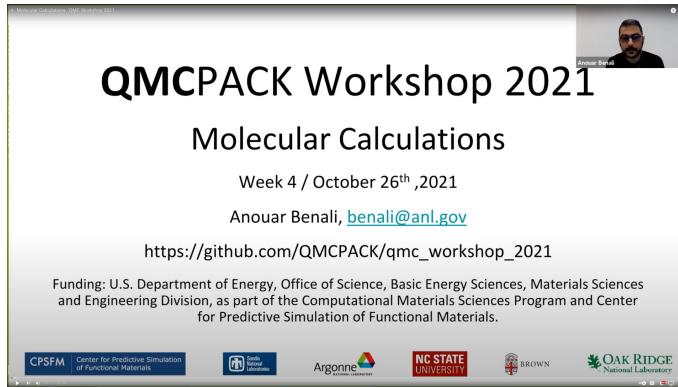
Connect with developers and users

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 command "qmcpack simple-H2O.xml" being run, followed by the QMCPACK copyright notice and global options. A red arrow points from the top of the terminal window towards the right edge of the slide.

Agenda

Tuesday (Building 240, Room 1501)

9.00 Welcome, Introduction, Background

Anouar Benali, ANL

Paul Kent, ORNL

9.50 Research Talk

Andrea Zen, Universita di Napoli, Italy

10.35 Break

10.50 Research Talk

Fernando Reboredo, ORNL

11.35 Short Research Talk

David Ceperley, UIUC

12.00 Lunch

13.00 Features & Progress Towards QMCPACK v4.0

Paul Kent, ORNL

Jaron Krogel, ORNL

Ye Luo, ANL

14.45 Break

15.00 Poster session and open collaboration session to test batched code, discuss with presenters & developers

17.15 Bus to guest house

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

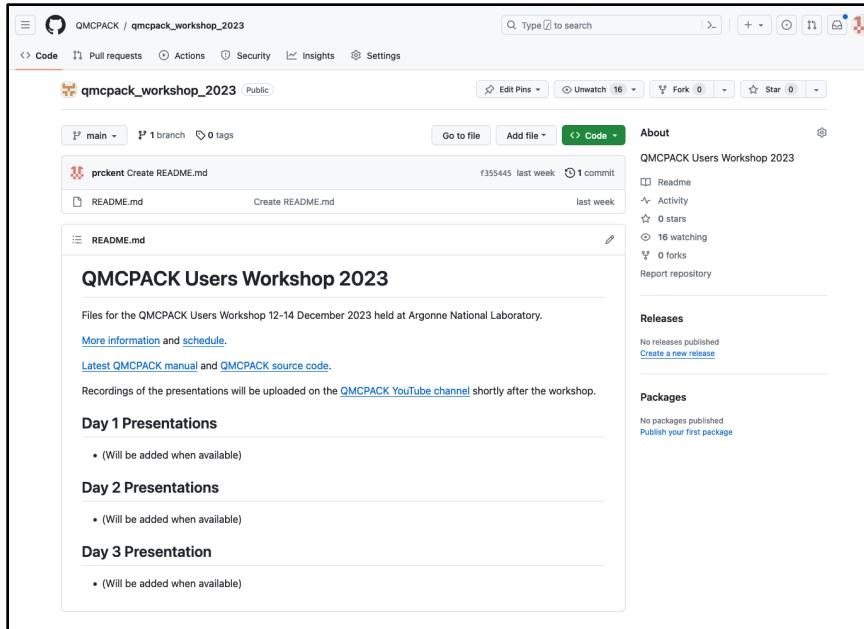
Support in General

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

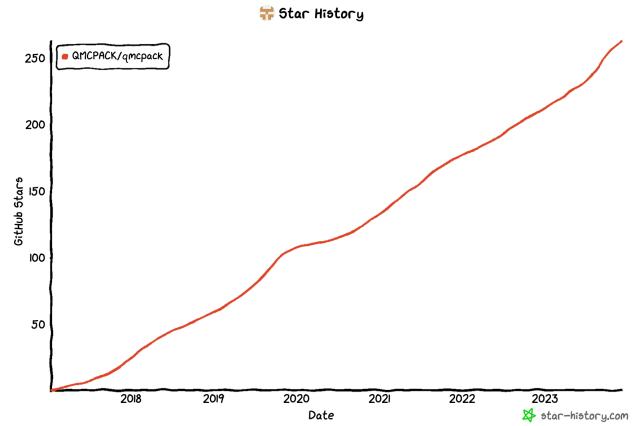
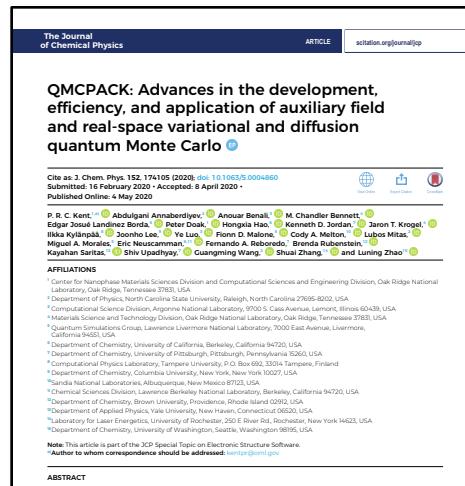
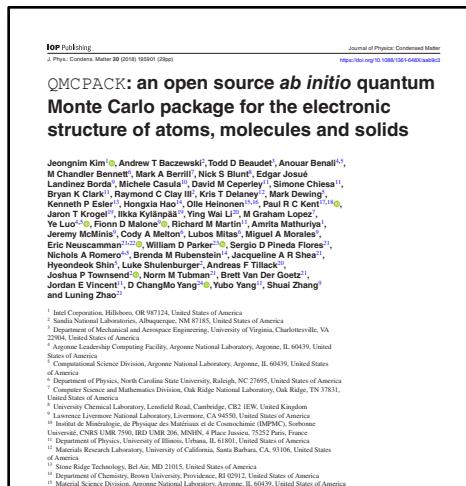
op 2023

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

CrI_3 Staros JCP **156** 014707 (2022), H phase diagram Niu PRL **130** 076102 (2023), $>10^3$ 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 CrI_3

Cite as: J. Chem. Phys. **156**, 014707 (2022); doi: 10.1063/0021-9606/156/1/014707
Submitted: 11 October 2021 • Accepted: 7 December 2021 • Published Online: 7 December 2022

Yongwei Niu (牛永伟)^{a,*}, Yubo Yang (杨博源)^{a,2,3}, Scott Jensen^{b,3}, Markus Holzmann^c, Carlo Pierleoni^d, and David M. Ceperley^{e,f}

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^f(Received 12 September 2022; revised 29 November 2022; accepted 12 January 2023; published 17 February 2023)

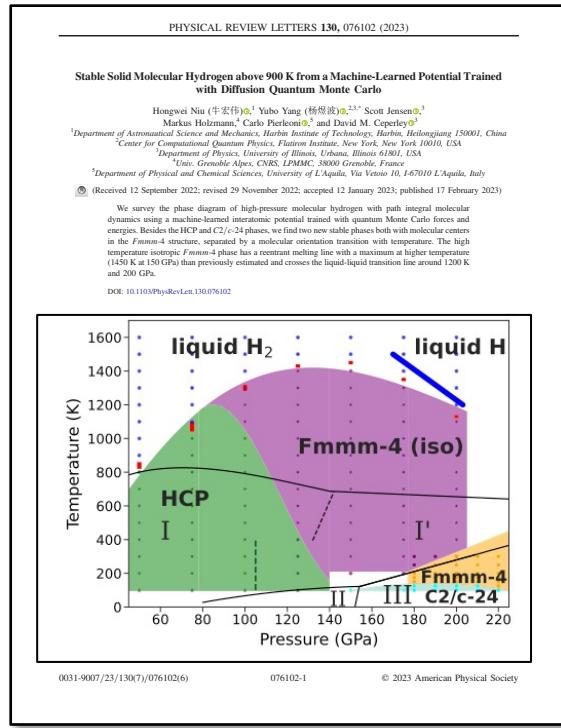
FIG. 1. Geometry of monolayer CrI_3 cleaved from the bulk structure reported in Ref. 45. (a) Top view depicting a lattice constant of $a_0 = 6.867 \text{ \AA}$ and the bond angles θ_1 and θ_2 computed in this work. (b) Side view depicting the $\text{Cr}-\text{I}$ bond distance of 2.728 \AA and the $\text{Cr}-\text{Cr}_2$ bond distance of 3.965 \AA .
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ABSTRACT
For the first time, we report the structural, magnetic, and phonon properties of monolayer CrI_3 using first-principles calculations. Our results show that monolayer CrI_3 exhibits a ferromagnetic ground state with a magnetic moment of $1.0 \mu_B/\text{f.u.}$ and a Curie temperature of $T_C = 1200 \text{ K}$. The magnetic moments are found to be localized on the Cr atoms, while the iodine atoms have zero magnetic moment. The calculated band gap is 0.65 eV , which is larger than the previous DFT-derived value of 0.45 eV . Our results also show that monolayer CrI_3 is a semimetal with a direct band gap at the Γ -point. We find that ML CrI_3 has key properties, which are different from those of their 3D counterparts. Due to the lack of dimensionality, the electronic properties of monolayer CrI_3 are more sensitive to the atomic arrangement than their 3D counterparts. Two-dimensional superconductivity, and exotic spin and charge density waves.

INTRODUCTION
2D materials as research. Due to the lack of dimensionality, the electronic properties of monolayer CrI_3 are more sensitive to the atomic arrangement than their 3D counterparts. Two-dimensional superconductivity, and exotic spin and charge density waves.

An exciting recent development in the field of new magnetic 2D materials.²⁴ While the Fermi theorem predicts finite-temperature magnetism for the $\text{Cr}-\text{I}$ system, it is still not clear whether the magnetic moments are localized on the Cr atoms or delocalized throughout the system. This uncertainty motivates our interest in understanding the electronic structure and ordering of their magnetic moments. In this regard, the discovery of the Mermin-Wagner theorem is the key to understanding the magnetic properties of monolayer CrI_3 .

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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^b, and Anouar Benali^{a*}

^aCite this: https://doi.org/10.1021/acs.jctc.3d1058 | 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 solids by numerically solving the electron Schrödinger equation. However, the computational cost of DMC scales exponentially with the number of electrons N , making DMC for smaller molecules to be 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 is important to validate the accuracy of the many-body theory used in QMCPACK. We show that when coupled with quantum machine learning (QML)-based surrogate methods, the computational burden can be alleviated such that quantum Monte Carlo (QMC) shows clear signs of convergence formation of high-quality descriptions of small systems. We discuss theoretical approaches to achieve this with the δ -function approximation, universal and accurate references for chemical bond dissociation energies, and scalable minimal atom-based QML (AQML) models. Numerical evidence is provided for the accuracy of AQML models for small organic molecules with up to five heavy atoms used as atoms and 50 medium-sized organic molecules with nine heavy atoms used as nuclei to validate the AQML predictions. Numerical evidence collected for Δ -QML models suggests that already modestly sized QMC training data sets of atoms suffice to predict total energies with near chemical accuracy throughout chemical space.

Downloaded for an OAK RIDGE NATIONAL LABORATORY on March 2, 2023 at 01:12:47 (UTC).

INTRODUCTION
The development of quantum machine learning (QML) based on quantum chemistry and used for the navigation of chemical compound space (CCS) is inherently limited by the predictive accuracy of the approximation used within the underlying quantum theory. Consequently, in order to make significant progress in the field, it is necessary to improve the accuracy ($\sim 1 \text{ kcal/mol}$ average deviation of calculated values from experimental measurements of dissociation energies), it is necessary to rely on training data sets of at least at the $\text{CCSD}(T)/\text{CBS}$ level. The current state-of-the-art (CSA) QML models, such as the $\text{CCSD}(T)/\text{CBS}$ generalizes considerably computational cost due to steep prefactors and scaling of $\sim 10^{10}$ to 10^{12} atoms per system size.¹ Thus, the routine generation of high-quality quantum training data sets is still elusive, even for relatively small organic molecules with only four or five “heavy” atoms. Here we demonstrate for an example of $\text{CCSD}(T)/\text{CBS}$ that the accuracy of the model is more useful than currently implemented and numerically more efficient quantum Monte Carlo (QMC) methods for computing QML training data. The subset of training data sets assesses the quality of the generated QML models, which serve as the foundation for the study of larger databases. Our numerical evidence indicates the possibility to routinely train QMCPACK models that are at least as powerful 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

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ACS Publications A

https://doi.org/10.1021/acs.jctc.3d1058

J. Chem. Theory Comput. XXXX, XXX, XXX–XXX

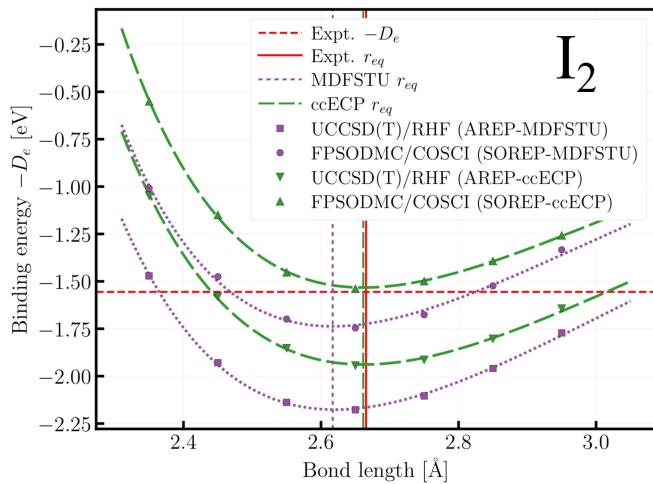
Pseudopotentiallibrary.org

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.

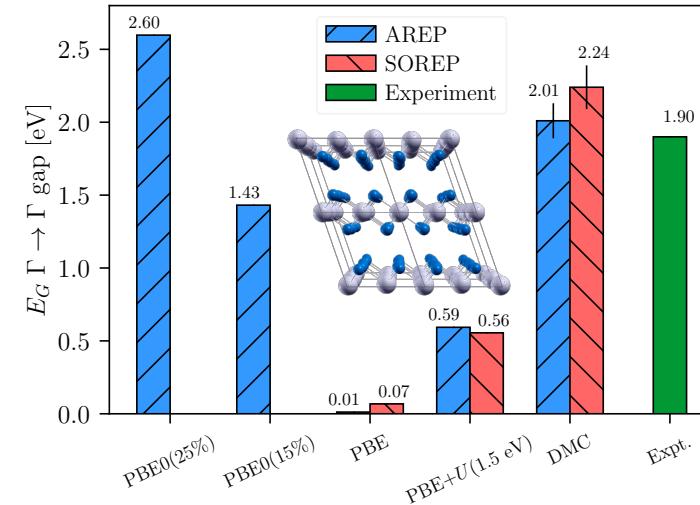
A screenshot of a web browser displaying the Pseudopotential Library website. The page title is "Pseudopotential Library". Below the title is a subtitle: "A community website for pseudopotentials/effective core potentials developed for high accuracy correlated many-body methods such as quantum Monte 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 Lithium (Li) and Beryllium (Be), orange for Sodium (Na) and Magnesium (Mg), red for Potassium (K) and Calcium (Ca), purple for Rb and Sr, blue for Cs and Ba, pink for Fr and Ra, and grey for the transition metals Sc-Ti, V-Cr, Mn-Fe, Co-Ni, Cu-Zn, Ga, Ge, As, Se, Br, Kr, and the lanthanides Ce-Th, Pr-Pm, Nd-Np, Sm-Pu, Eu-Am, Gd-Cm, Tb-Bk, Dy-Cf, Ho-Es, Er-Fm, Tm-Md, Yb-No, Lu-Lr. Below the grid, a footer message reads: "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.



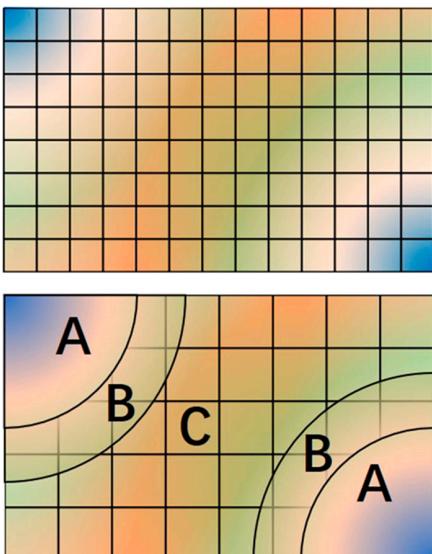
SO DMC binding energy & bond length are most accurate compared to experiment.
G. Wang et al. JCP **157** 054101 (2022)



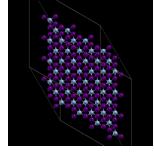
Role of SO on gap of RuCl_3
A. Annaberdiyev et al. PRB **106** 075127 (2022)

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).

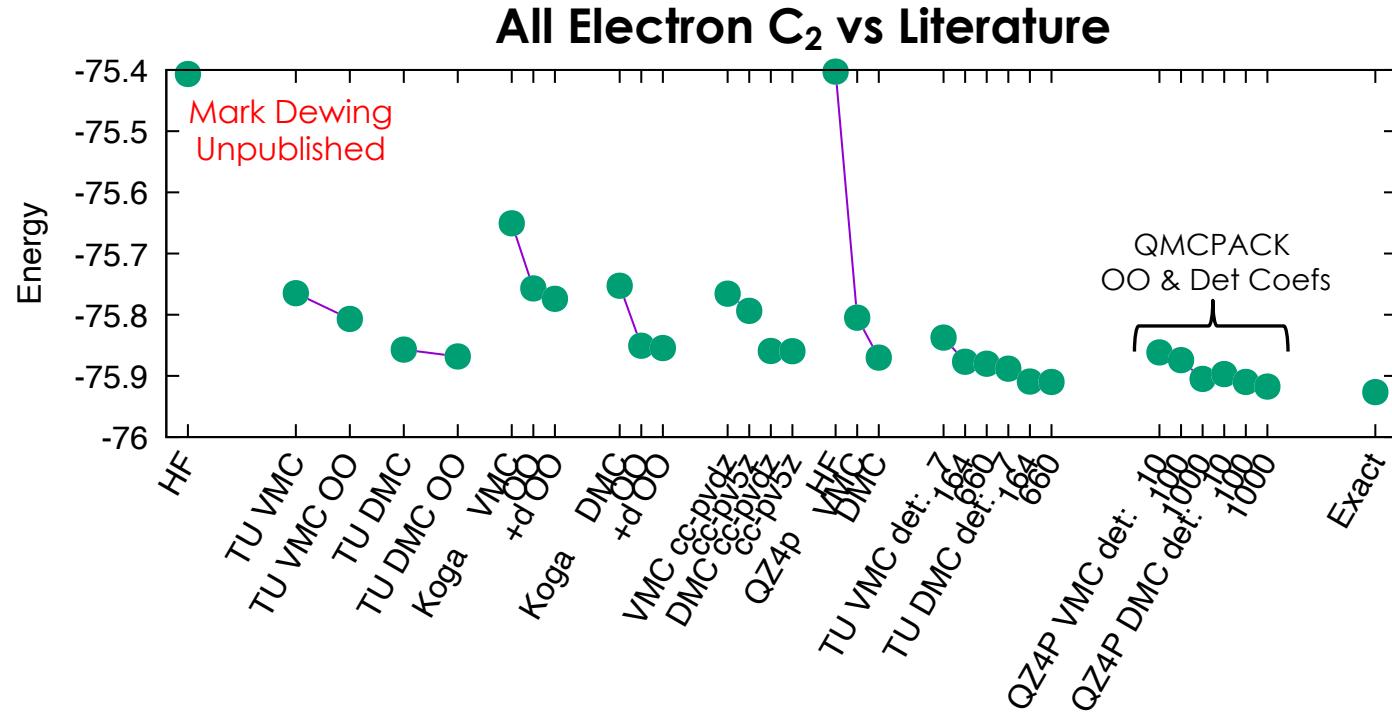


CrI_3 Monolayer Analysis (Saritas & Krogel)

	# 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

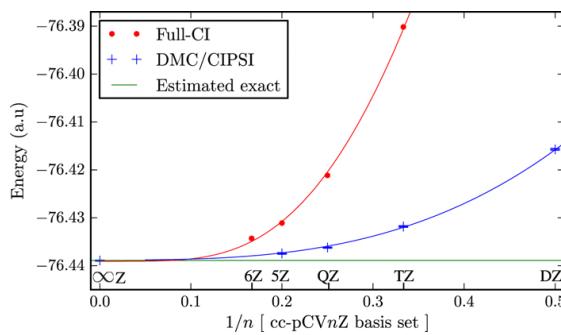
Orbital Optimization

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.**

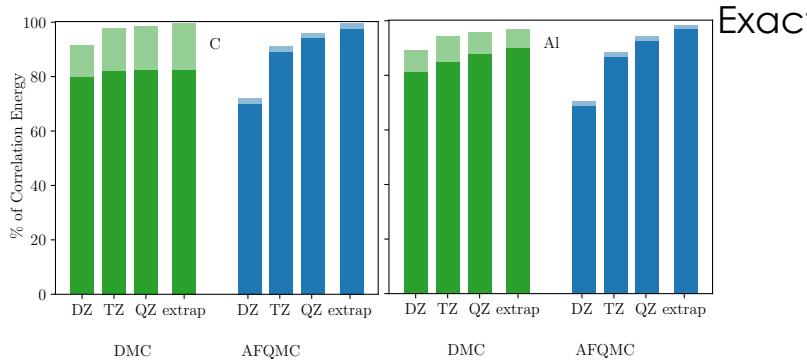


Selected Configuration Interaction

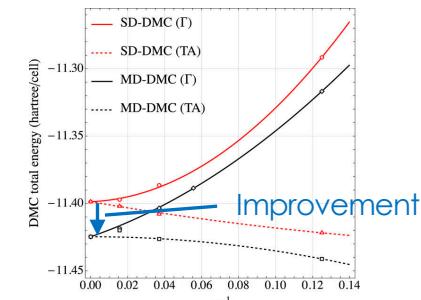
Discuss with Anouar Benali. **Systematic and deterministic reduction** of nodal error via large configuration interaction expansions. Workflow via Quantum Package both for molecules and solid-state. Many efficiency improvements in LCAO and multideterminants recently. **Low memory usage periodic LCAO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

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?

