

# Accurate Electronic and Magnetic Properties of 2D Materials Using Quantum Monte Carlo Methods

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NIST, Materials Science and Engineering Division

QMCPACK Workshop: December 13, 2023

<https://jarvis.nist.gov>



Joint Automated Repository for Various Integrated  
Simulations

12/13/2023

# Acknowledgement and Collaboration



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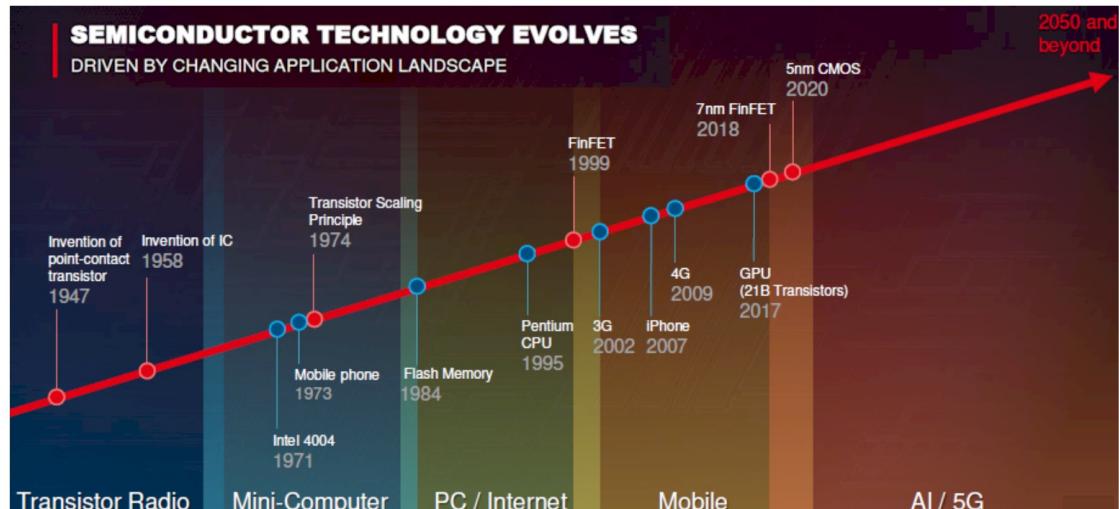
# Outline

- Overview of 2D Materials
- Limitations of DFT for 2D Materials
- QMC Background
- QMC Applied to 2D Systems:
  1. Electronic and Optical Properties of 2D GaSe
  2. 2D  $\text{GaS}_x\text{Se}_{1-x}$  Alloys: Constructing Energy Hull
  3. Magnetic properties of 2D  $\text{CrI}_3$ ,  $\text{CrBr}_3$  and  $\text{MnO}_2$
  4. Structural and magnetic properties of 2D T- and H-VSe<sub>2</sub>\*
- Overview of JARVIS at NIST
- JARVIS Leaderboard
- Conclusions and Future Perspectives

# QMCPACK

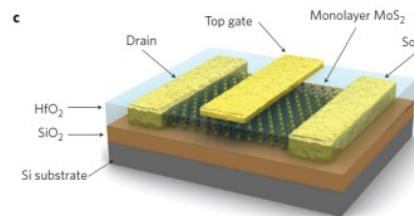
# 2D Materials: Applications

NIST



<https://www.nextplatform.com/2019/09/13/tsmc-thinks-it-can-uphold-moores-law-for-decades/>

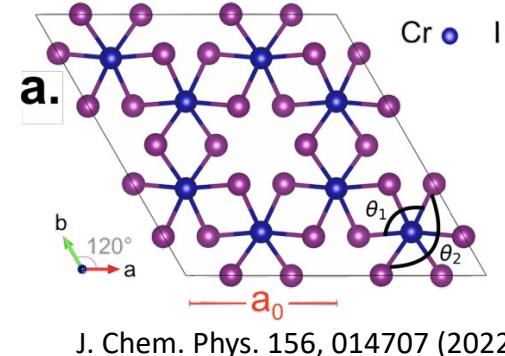
- Denser (More transistors)
- Energy Efficient
- Cheaper (per transistor)



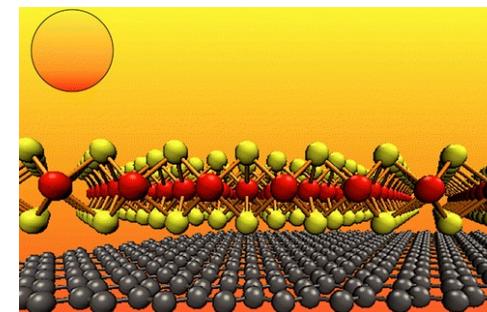
Nature Nanotechnology 6, 147–150 (2011)

## Smaller Transistors

- LEDs
- Flexible electronics



J. Chem. Phys. 156, 014707 (2022)

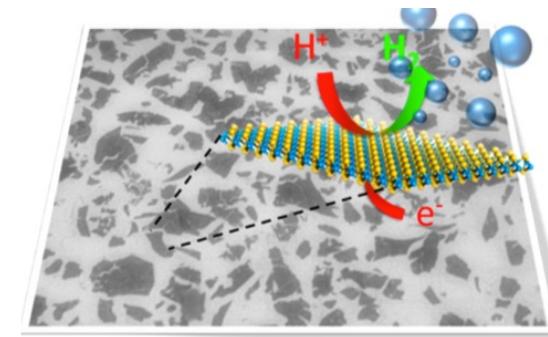


## Efficient energy conversion

Nano Lett. 13, 3664–3670 (2013)

## H<sub>2</sub> generation

Nano Lett. 13, 6222–6227 (2013)



## 2D Magnets

- Spintronics
- Magnetic Storage

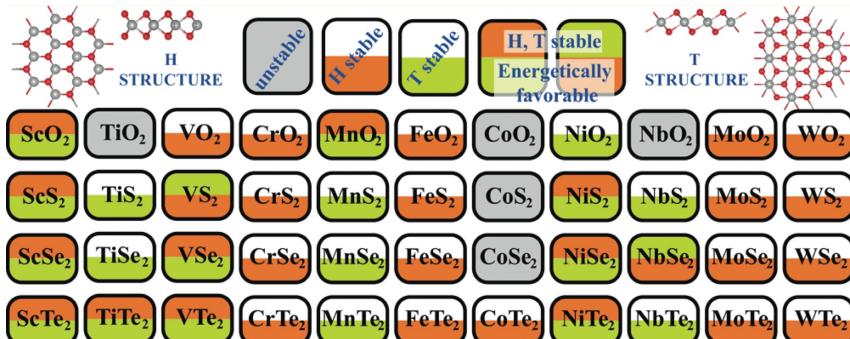
# 2D Materials: Rising Stars

NIST

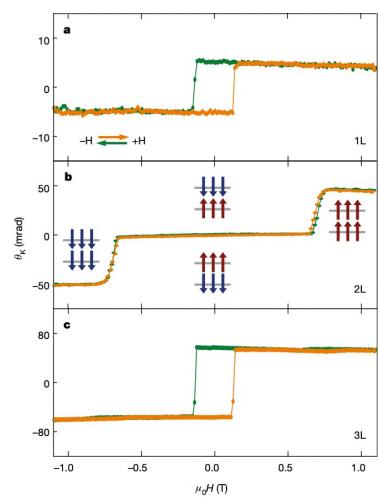
week ending  
24 SEPTEMBER 2010

## Rising Stars:

- Monoelemental
- Transition Metal Dichalcogenides
- Post-Transition Metal Chalcogenides
- Transition Metal Halides
- Transition Metal Oxides
- MXenes



J. Phys. Chem. C 116, 8983 (2012)



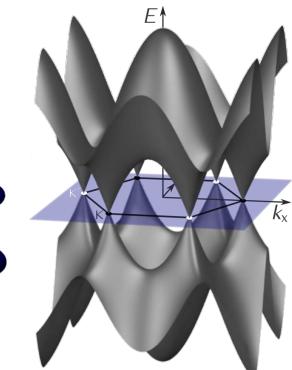
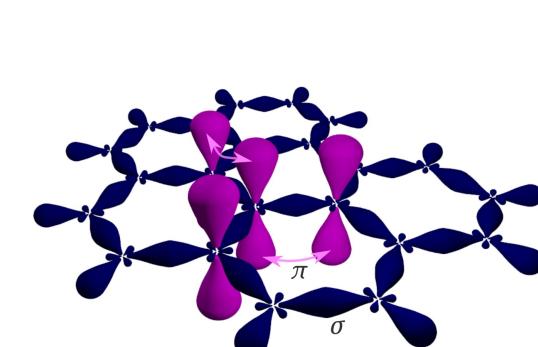
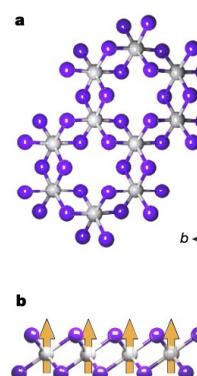
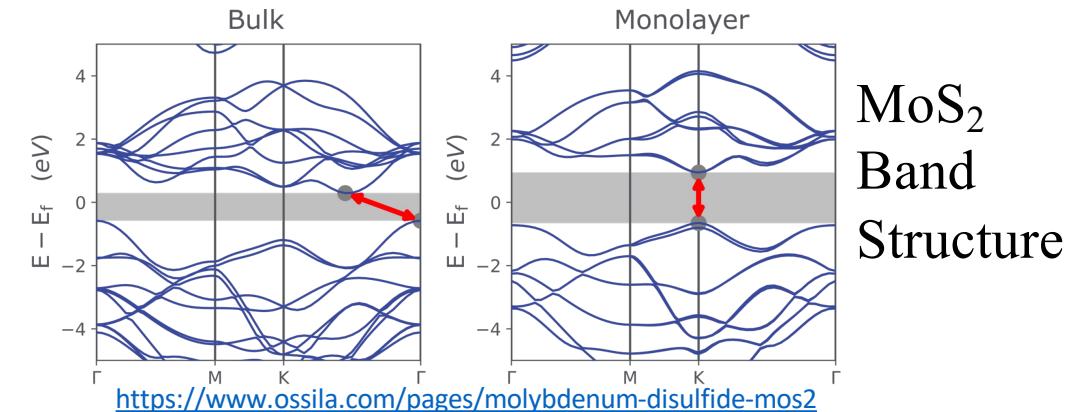
Nature 546, 270–273 (2017)

PRL 105, 136805 (2010)

PHYSICAL REVIEW LETTERS

### Atomically Thin MoS<sub>2</sub>: A New Direct-Gap Semiconductor

PRL 105, 136805 (2010)



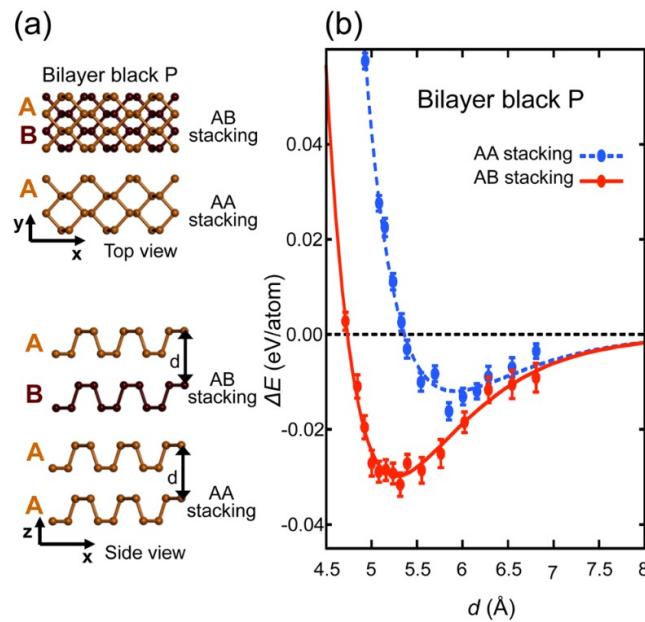
https://en.wikipedia.org/wiki/Electronic\_properties\_of\_graphene

# Why are 2D Materials difficult?

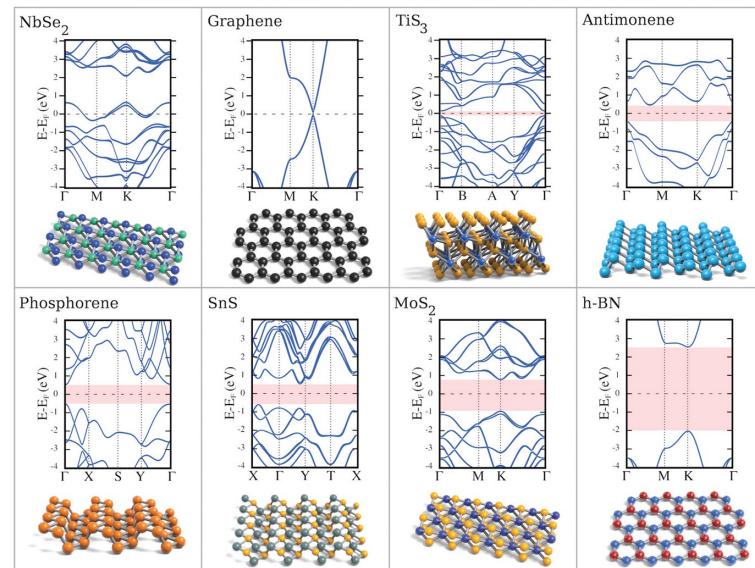
NIST

## Difficulties with modeling 2D/layered materials

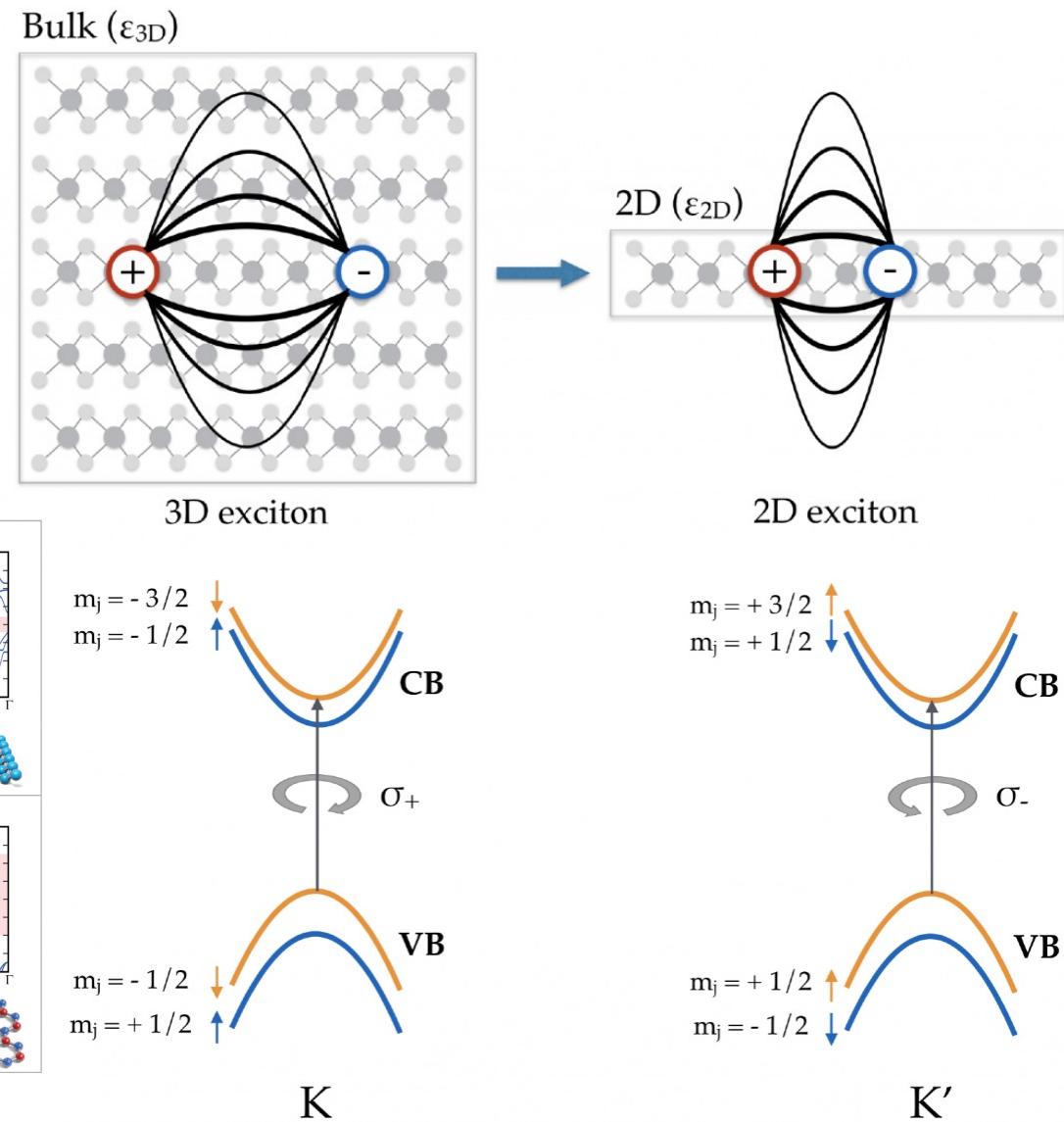
- vdW interactions
- Correlated electronic structure (*d* and *f* electrons)
- Reduced screening (strong excitonic effects)
- Spin-orbit coupling (band splitting, topological properties)
- Many 2D materials are transition metal-based



*Nano Lett.* 2015, 15, 12, 8170–8175



*Chem. Soc. Rev.*, 2017, 46, 4387–4399



<https://www.femtosecond.fisi.polimi.it/2dmaterials/>

# DFT: Success and Limitations

NIST



## DFT Successes

- Reduces  $3N$ -dimensional problem to 3
- Good balance between computational efficiency and accuracy

## DFT Shortcomings

- Results depend directly on which XC functional is used
  - van der Waals interactions (corrections)
  - Systems with strongly localized and correlated electrons (DFT+U)
  - Band gaps (underestimated)

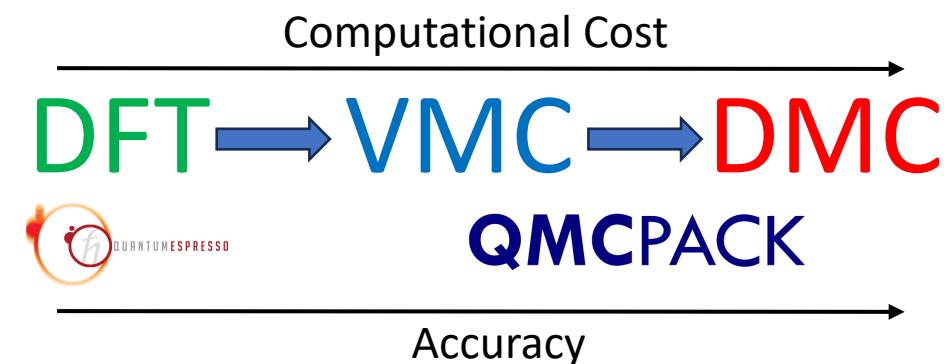
## Proposed Solutions

- Post DFT methods (**many-body perturbation theory**)
- Stochastic methods (**Quantum Monte Carlo**)

# Computational Metrology: Quantum Monte Carlo



- A class of algorithms that apply **MC integration** to solve quantum problems (many-body)
- **Variational MC (VMC)** and **Diffusion MC (DMC)** are most common for studying crystals
- Scales  $\sim N_e^{3-4}$  (similar to DFT), accuracy beyond DFT
- Current state of the art software: QMCPACK



©Nicholas Michalec Photography

# QMC: Diffusion Monte Carlo (DMC)

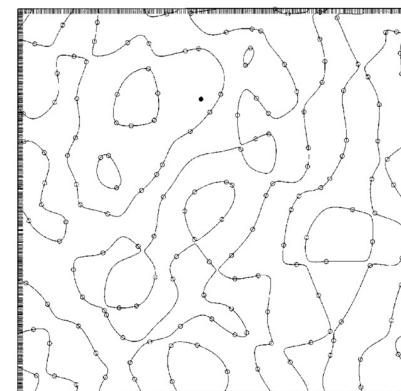
NIST

$$-\frac{\partial_t \Phi(\mathbf{R}, t)}{\partial t} = (\hat{H} - E_T)\Phi(\mathbf{R}, t),$$

Imaginary-time Schrödinger Eq.

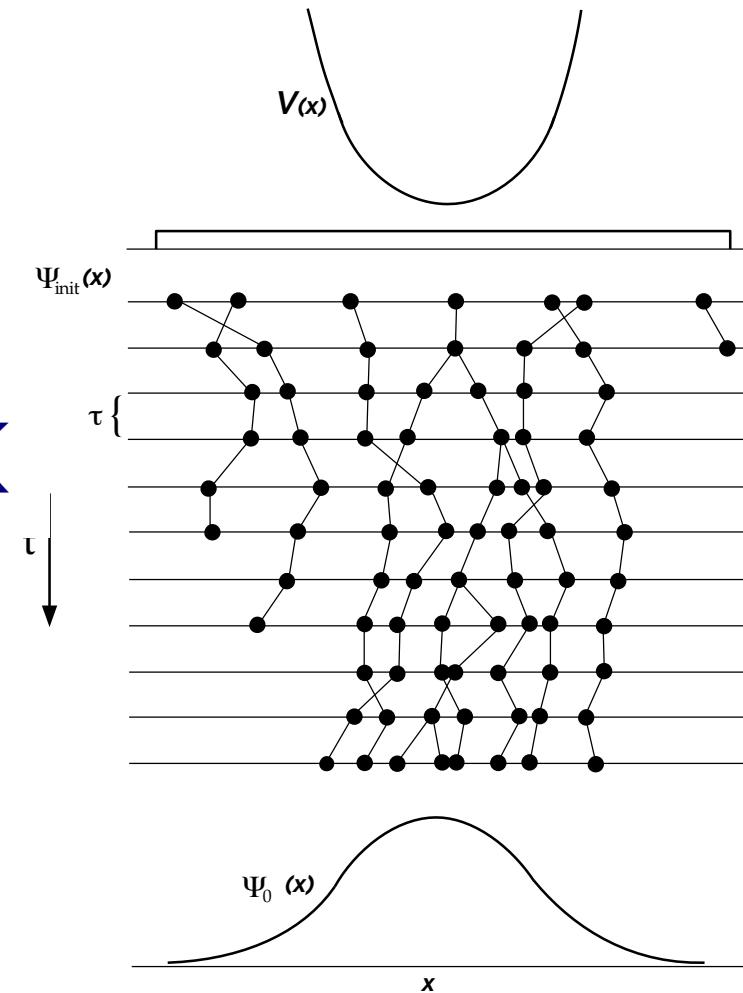
- Projector (Green's function)-based method
- Simulate **diffusion** of walkers in **imaginary-time** until you reach **steady state**
- Timestep **errors**
- Finite size **errors**

**QMCPACK**



Rev. Mod. Phys., 73, 1, (2002)

Fixed-nodal surface



Rev. Mod. Phys., 73, 1, (2002)

Diffusion of walkers in imaginary time

# QMCPACK on Small Machines

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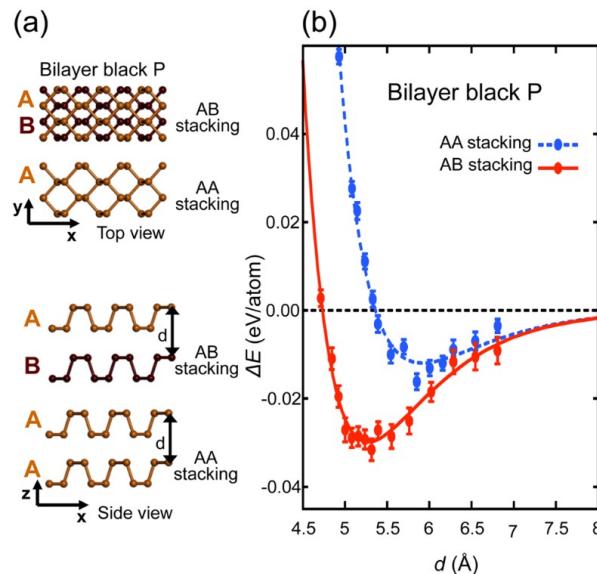


UMBC

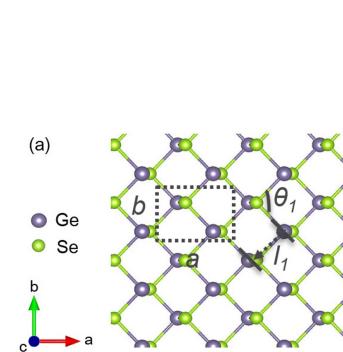
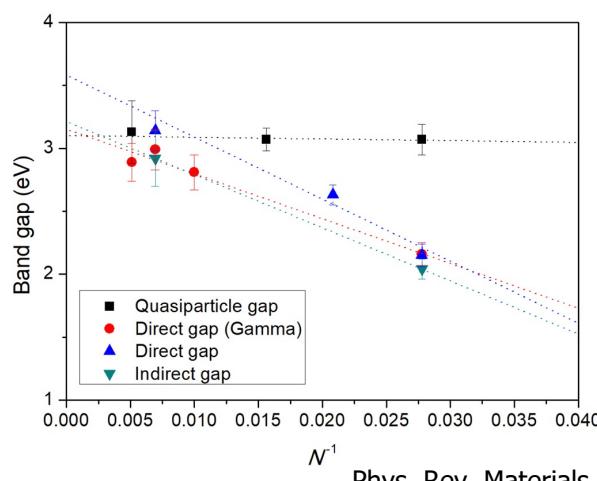
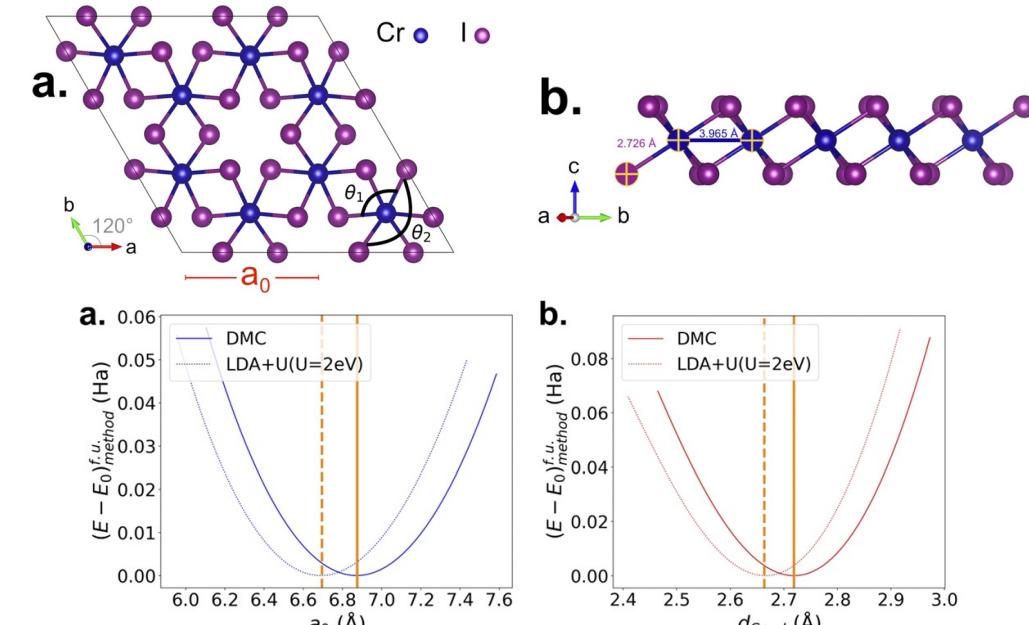
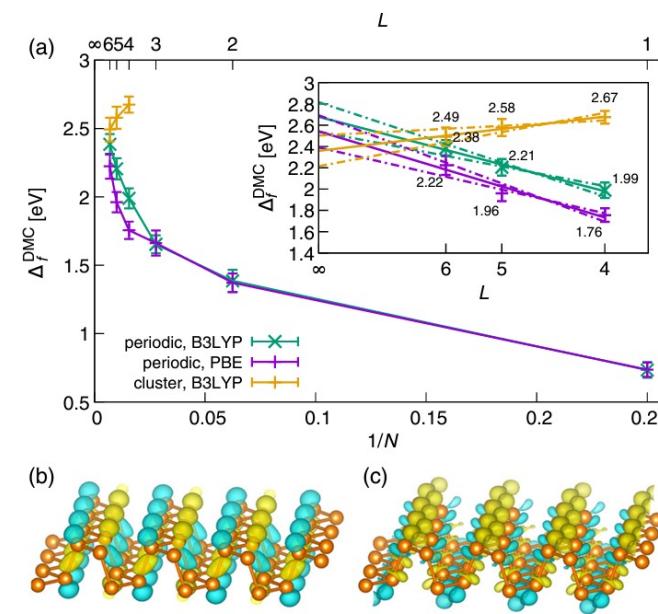
Kisir, est. 2019 at UMBC

# DMC for 2D Materials

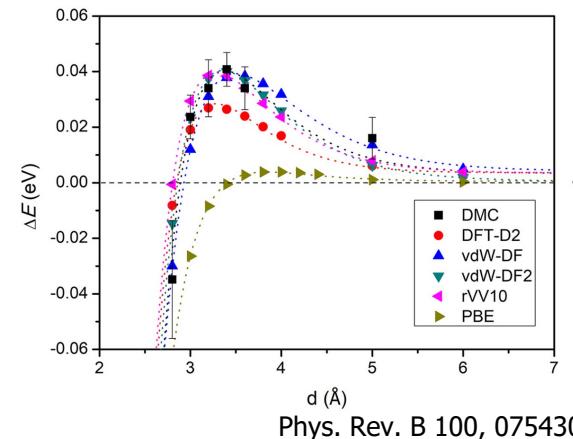
NIST



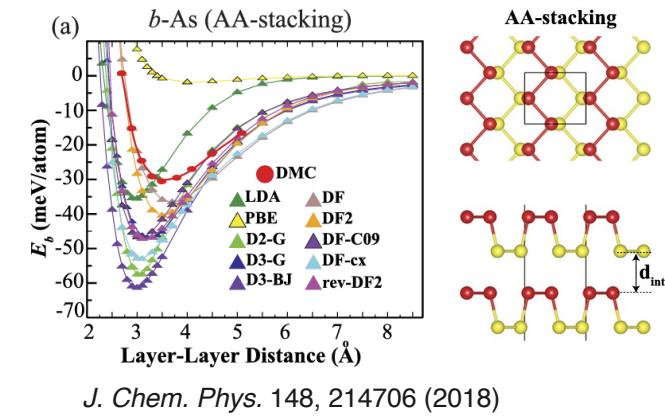
Nano Lett., 15, 12, 8170–8175 (2015)



Phys. Rev. Materials 5, 024002 (2021)

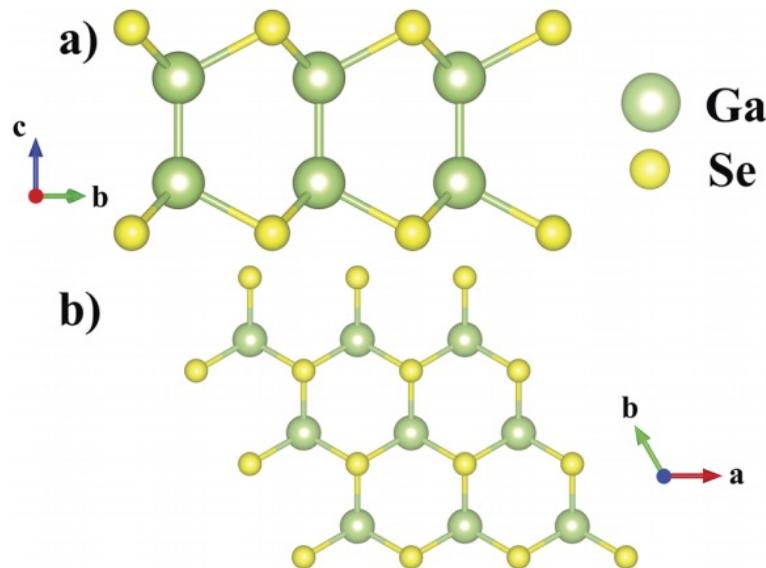


Phys. Rev. B 100, 075430 (2019)

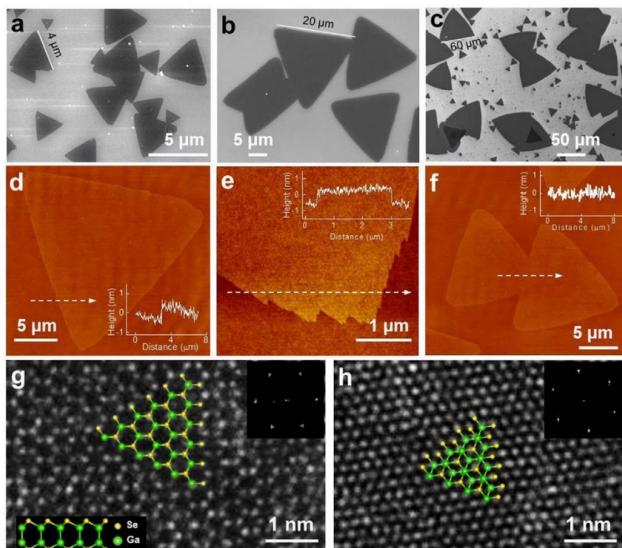


J. Chem. Phys. 148, 214706 (2018)

# Motivation: 2D GaSe



D. Wines, K. Saritas, C. Ataca. *J. Chem. Phys.*, 153, 154704 (2020)

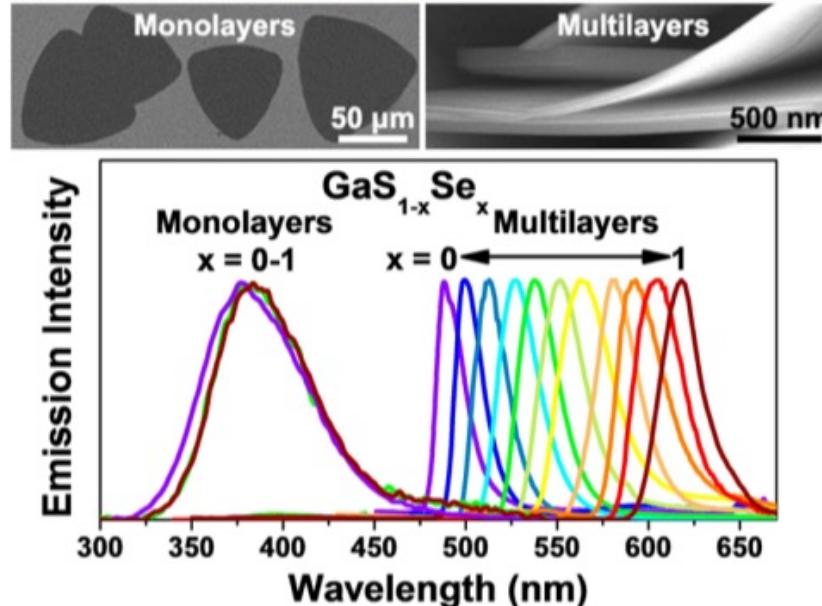


Scientific Reports, 4, 5497 (2014)

## Motivation

- **2D GaSe** is a semiconductor suitable for photovoltaics, transistors, water-splitting, adatoms also binds strongly to surface (energy applications)
- Post transition metal chalcogenides predicted to have **lower exciton binding energy** than transition metal dichalcogenides
- Reliably synthesized
- DFT functionals **disagree** and results are **off from experiment**

# Monolayer GaSe



ACS Nano 9, 9585–9593 (2015).

## 2D GaSe in Experiment

- $a = b = 3.74 \text{ \AA}$  [1]
- $E^{\text{QP}} = 3.5 \text{ eV}$  (on graphene) [2]
- $E^{\text{opt}} = 3.3 \text{ eV}$  (on  $\text{SiO}_x/\text{Si}$ ) [3]
- **Alloys** have been synthesized in few-layer form, **band gap** is **tunable** [3]

## Goal: Use QMC for 2D GaSe

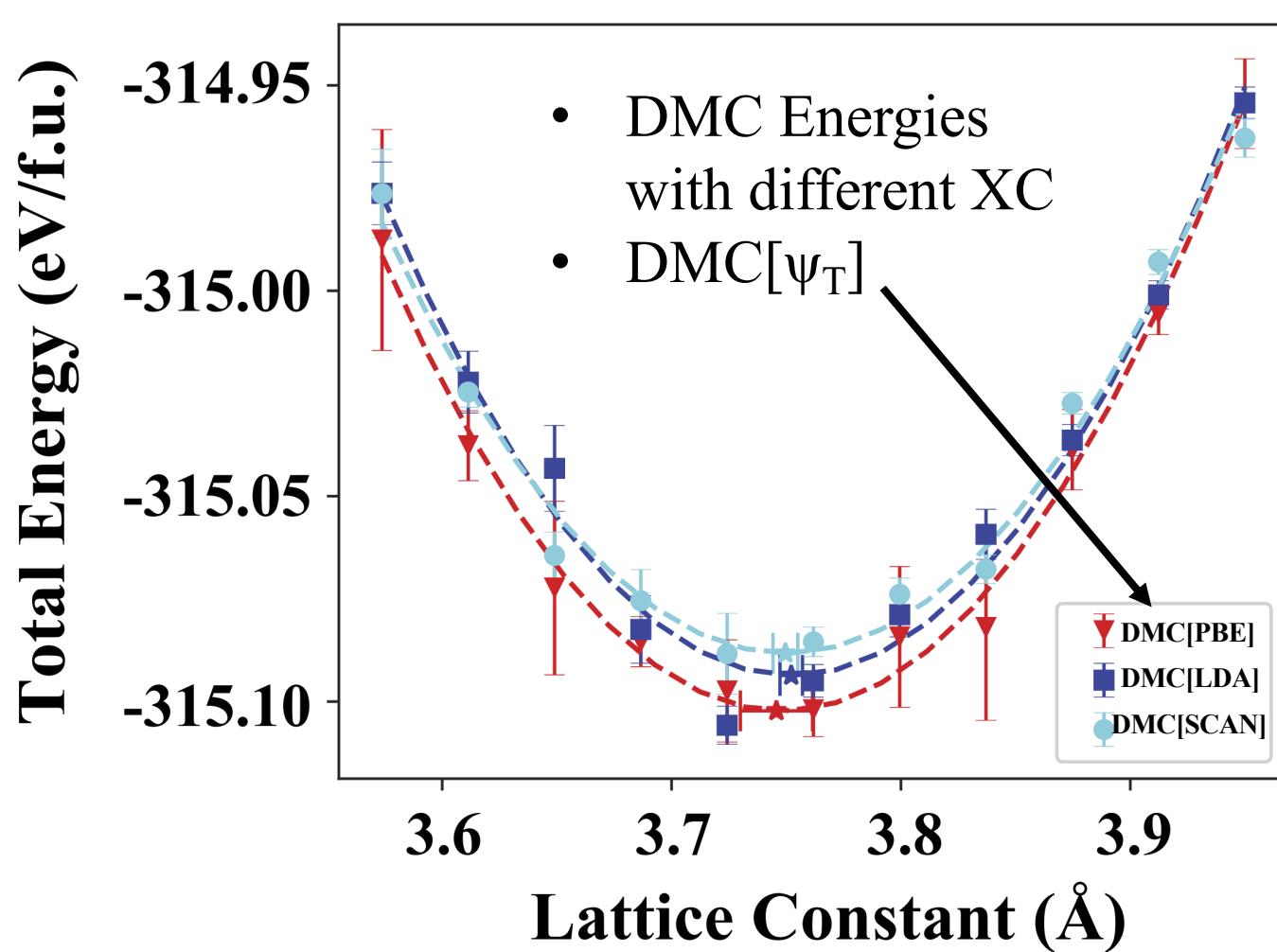
- Obtain “chemically accurate” **results for GaSe** from diffusion Monte Carlo (**DMC**) since DMC has weak dependence on starting wavefunction (WF)
- Calculate lattice parameters, band gap
- **Benchmark** with DFT and GW results (**multiple functionals**) and **experiment**

[1] Sci. Rep. 4, 5497 (2014).

[2] Phys. Rev. B 96, 035407 (2017).

[3] ACS Nano 9, 9585 (2015).

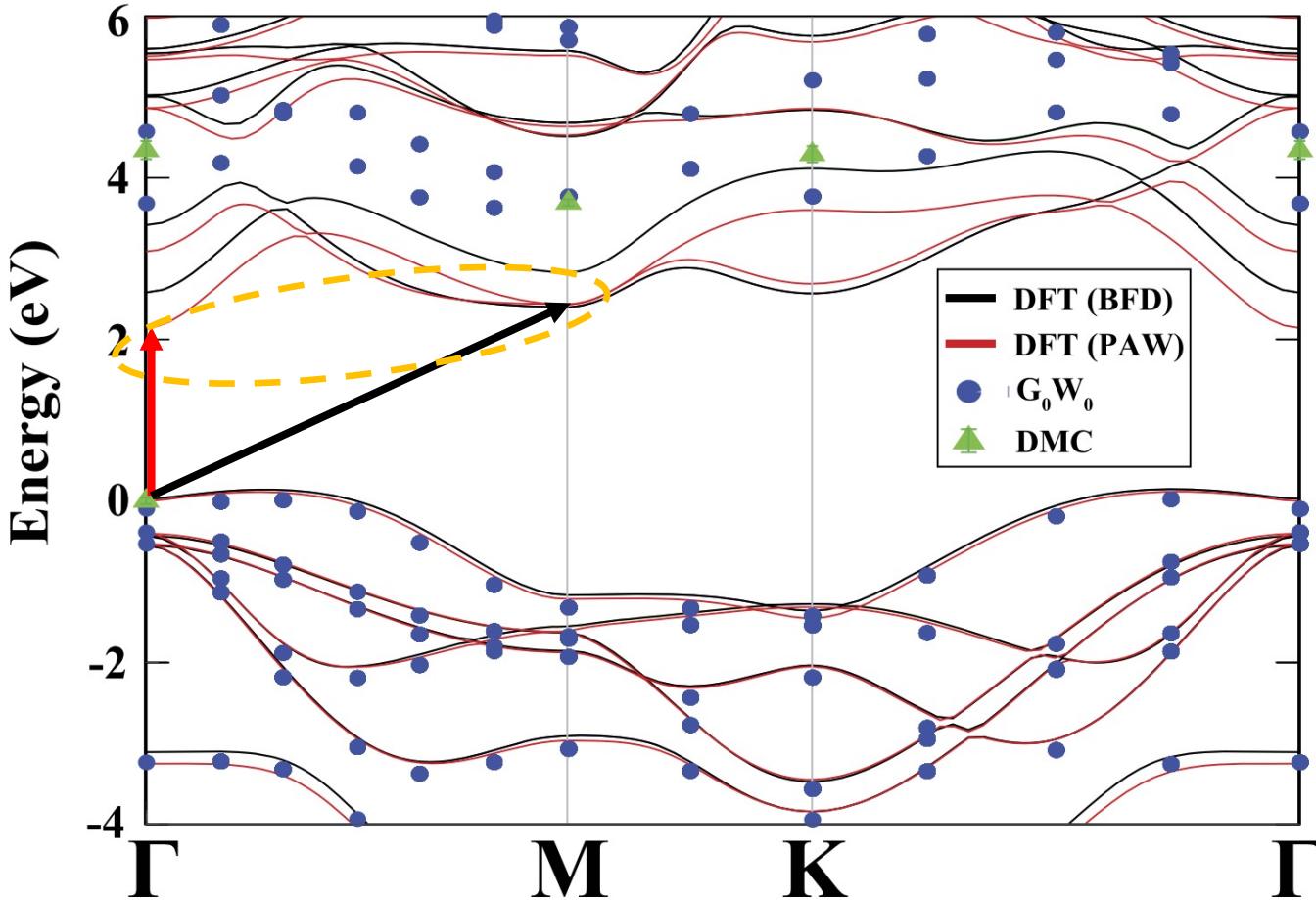
# 2D GaSe: Optimal Lattice Constant



PBE: 3.81 Å  
LDA: 3.71 Å  
SCAN: 3.76 Å  
Exp: 3.74 Å

- Lattice scaled isotropically Using a trial wavefunction ( $\Psi_T$ ) from PBE, LDA, and SCAN yields in similar results at the DMC level
- DMC[PBE]: 3.74(2) Å
- DMC[LDA]: 3.75(1) Å
- DMC[SCAN]: 3.75(1) Å

# Discrepancies in Band Structure: DMC Corrections



D. Wines, K. Saritas, C. Ataca. *J. Chem. Phys.*, 153, 154704 (2020)

- Small energy difference between each high symmetry point at the conduction band edge
- Results in incorrect band gap prediction and direct/indirect discrepancy
- DFT and GW band gaps are heavily dependent on functional and pseudopotential choice
- Benchmarked with PBE, SCAN, HSE06,  $G_0W_0$  and QMC pseudopotentials (BFD)

\*PBE functional used for DFT,  $G_0W_0$  and DMC (above)

# 2D GaSe: Band Gap Summary

Functional	Kohn-Sham Gap	Direct (D) or Indirect (I)
PBE (PAW)	2.14	D
PBE (BFD)	2.37	I
SCAN (PAW)	2.53	D
SCAN (BFD)	2.78	I
HSE (PAW)	3.11	D

Method	Quasiparticle Gap	D or I
$G_0W_0$ –PBE (PAW)	3.81	D
$G_0W_0$ –SCAN (PAW)	3.14	D
DMC (BFD)	3.69(5)	I

- 2D GaSe is an **indirect** semiconductor, QP gap and in **good agreement** with experiment (3.5 eV)
- **Upper bound** of **80 meV** for **exciton binding** energy (from 3.70(4) eV optical gap)
- Have in depth **theoretical benchmarks** for this material, which will aid future studies involving DMC and 2D materials

# Motivation and Goal: 2D $\text{GaS}_x\text{Se}_{1-x}$ Alloys

Alloy  
Automated  
Theoretic Toolkit

## Motivation

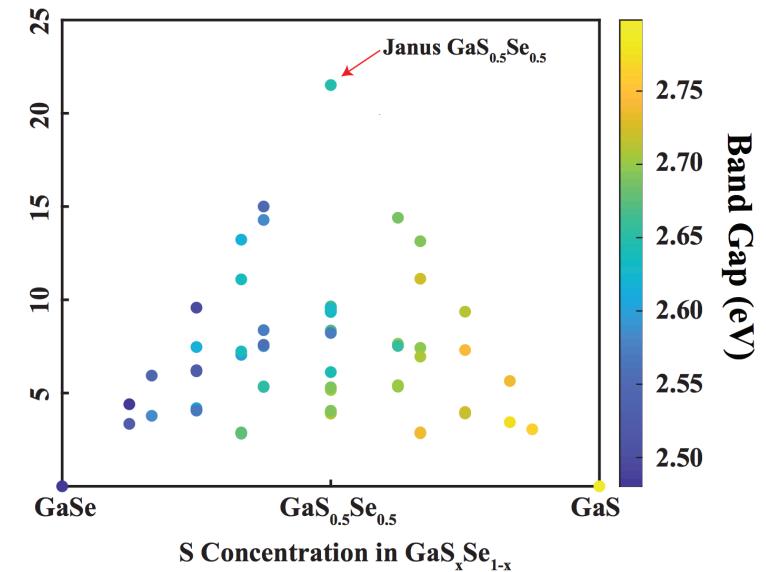
- Alloying is an effective way to **tune** 2D material properties
- 2D **Janus**  $\text{GaS}_x\text{Se}_{1-x}$  and  $\text{GaS}_x\text{Se}_{1-x}$  alloys have been studied at the DFT level

## Goal

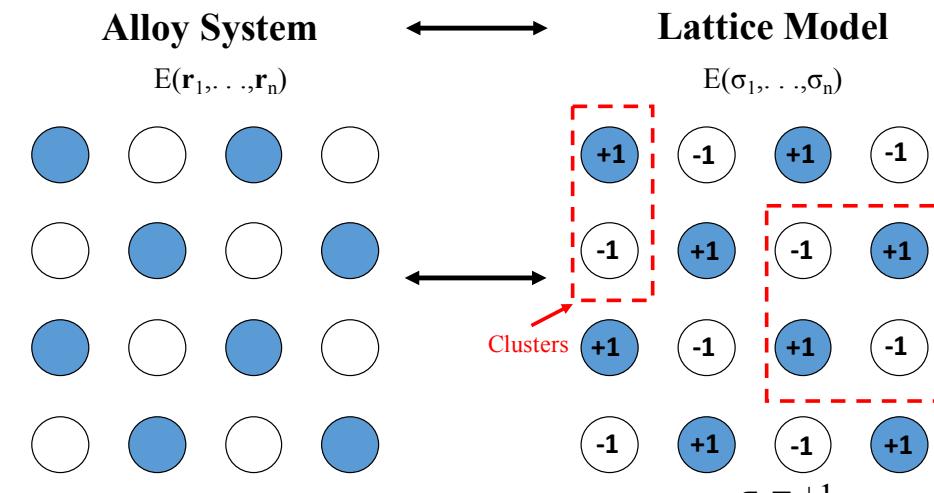
- Develop **procedure** to study **2D alloys** with similar atomic environments using **DMC**
- Study effect of **different Jastrow parameters** on the **VMC and DMC energies**
- **Minimize** the number of **optimizations** needed (*Jastrow sharing*) while reducing error



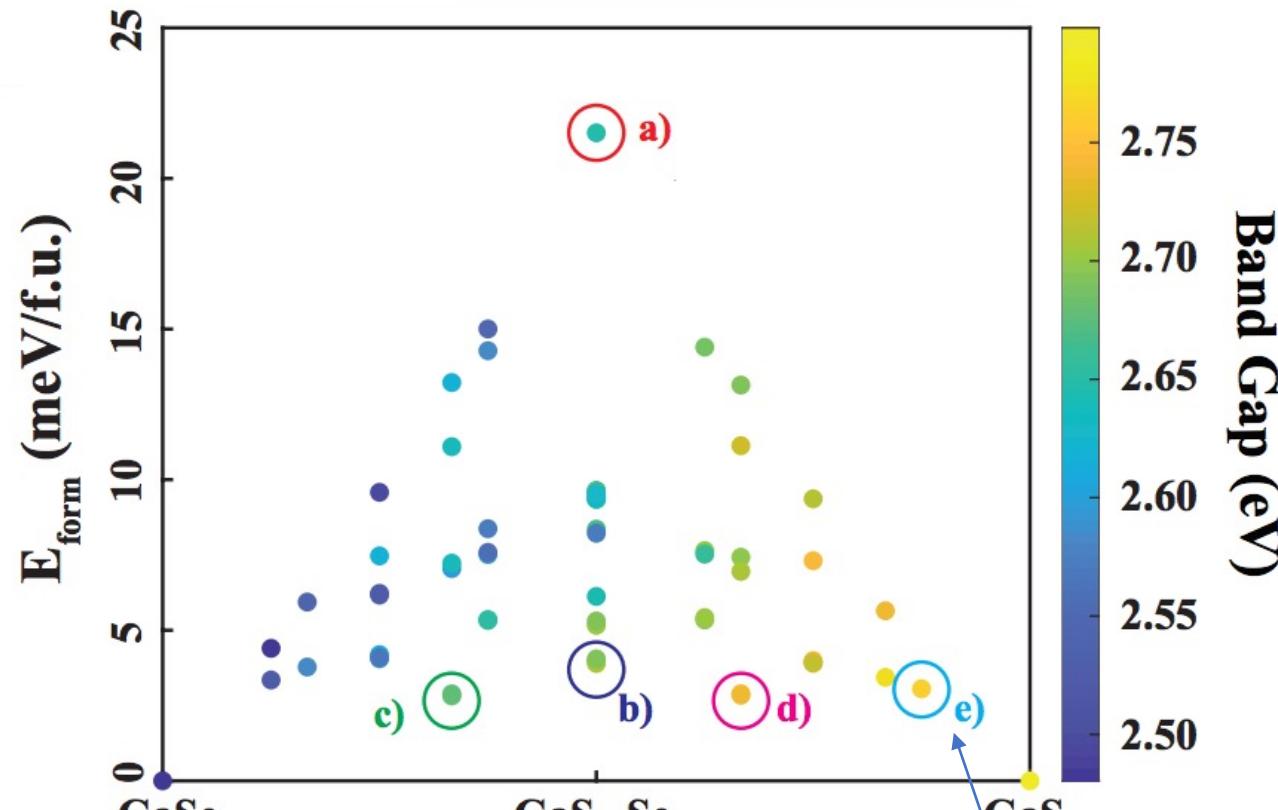
<https://en.wikipedia.org/wiki/Janus>



D. Wines, K. Saritas, C. Ataca. *J. Chem. Phys.*, 155, 194112 (2021)

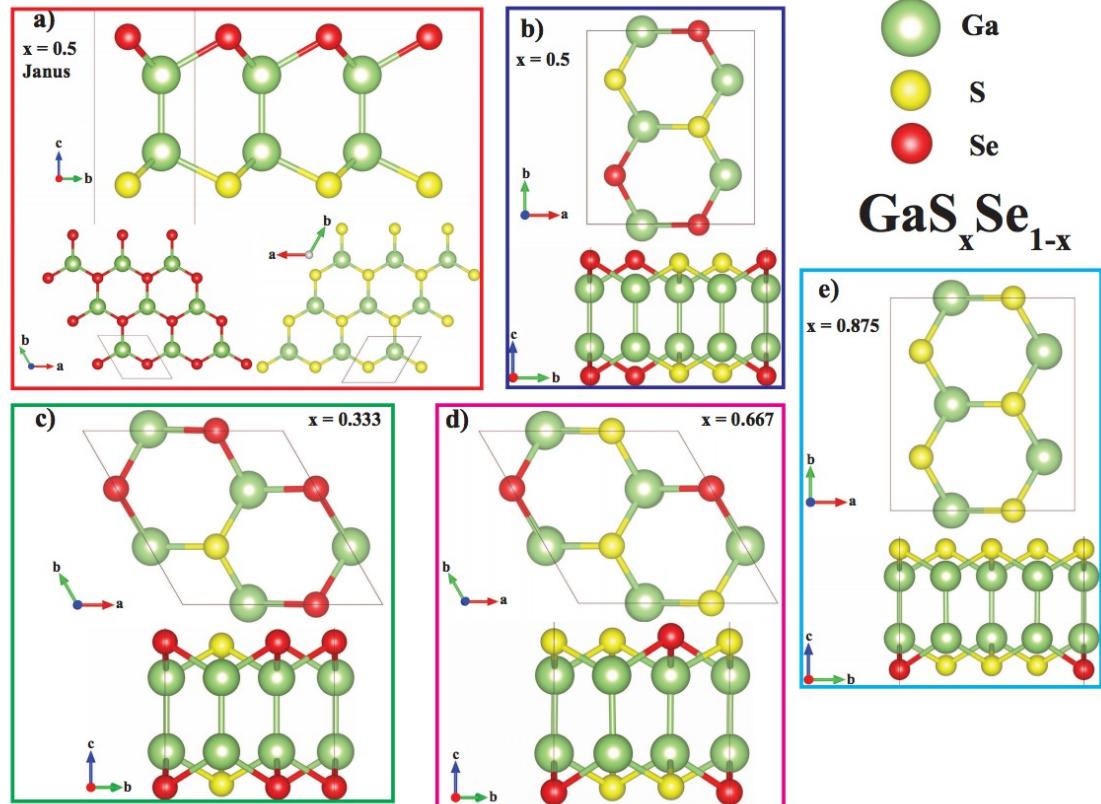


# 2D $\text{GaS}_x\text{Se}_{1-x}$ Alloys: DFT



D. Wines, K. Saritas, C. Ataca. *J. Chem. Phys.*, 155, 194112 (2021)

Smallest  
Wigner-Seitz  
radius

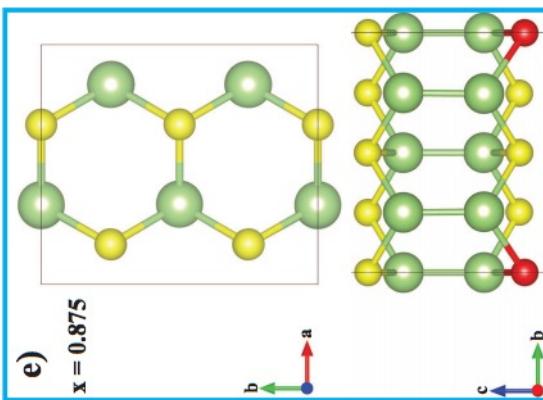
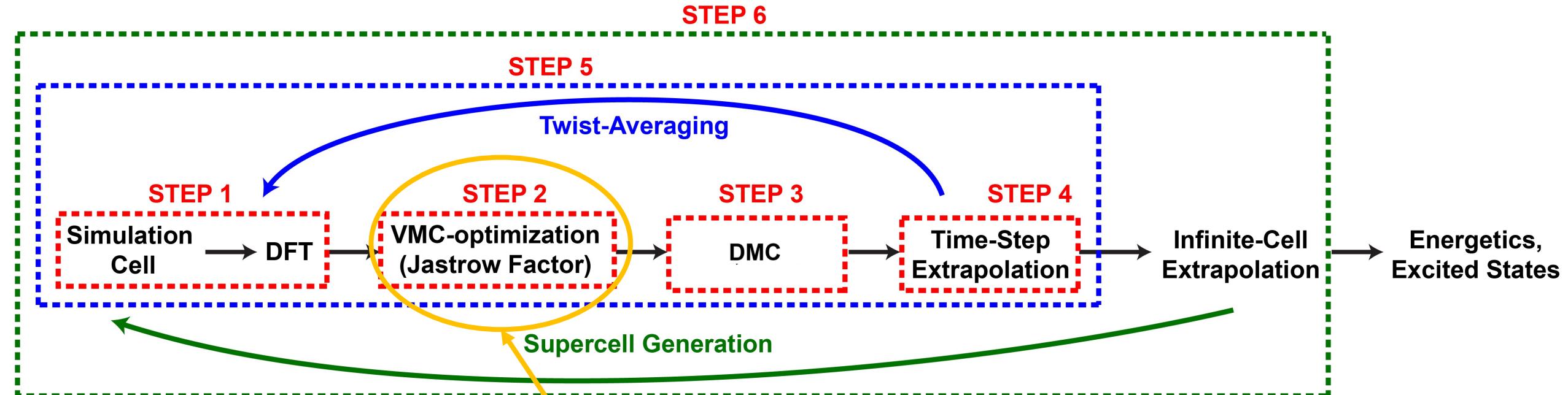


D. Wines, K. Saritas, C. Ataca. *J. Chem. Phys.*, 155, 194112 (2021)

- Energy hull diagram calculated with **DFT**
- Screen most **stable** structures from hull

$$E_{\text{form}} = E_{\text{GaSSe}} - x^*E_{\text{GaSe}} - (1 - x)^*E_{\text{GaS}}$$

# Change in QMC Workflow



- **Alloys:** must perform **Steps 1 – 6** for **several materials**
- Modify **Step 2** to **save cost**
- Most convenient to **reuse** Jastrows from cell with **smallest Wigner-Seitz radius**

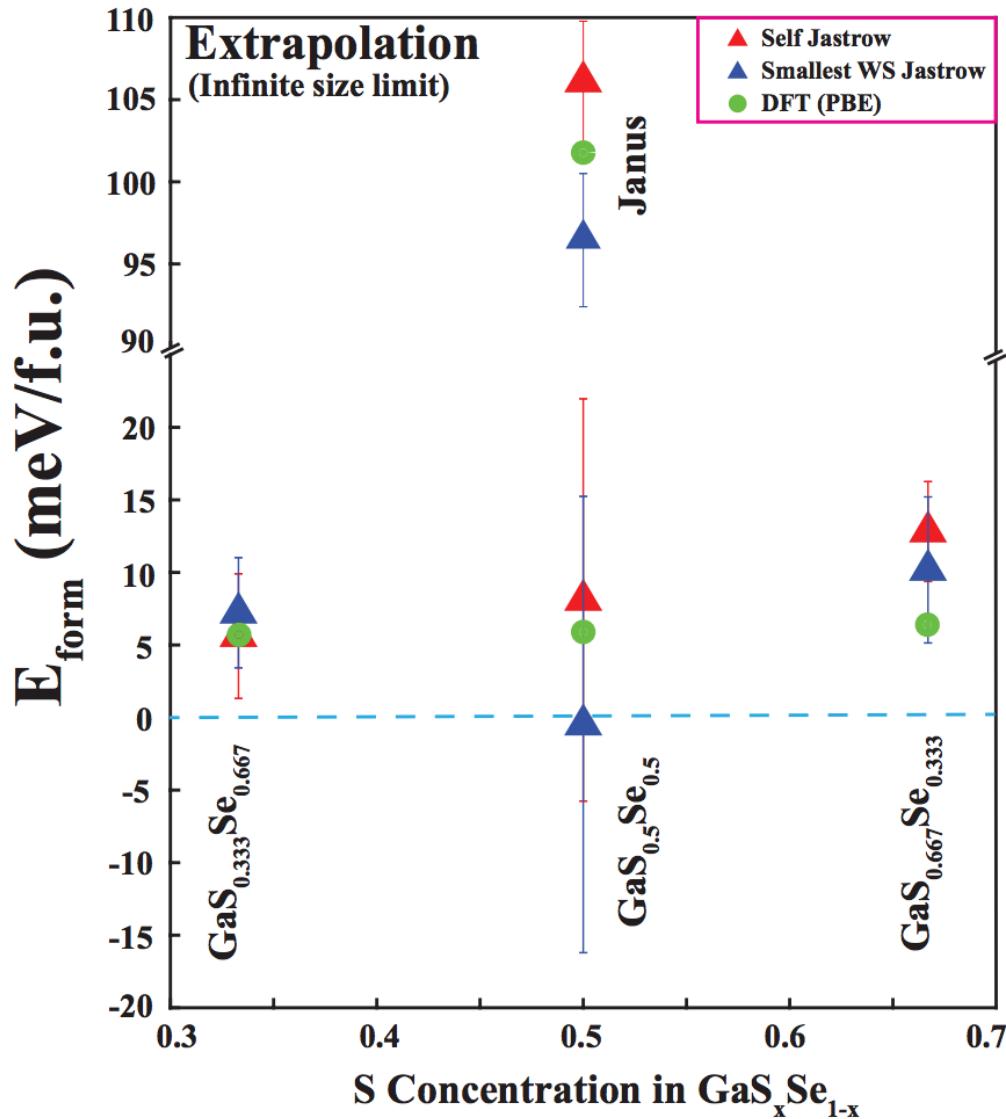
# Jastrow Sharing: Testing

<b>DMC-J2</b>	<b>Janus</b>	<b>x = 0.5</b>	<b>x = 0.333</b>	<b>x = 0.667</b>
Janus Jastrow	0	-3(7)	-9(7)	-6(7)
x = 0.5 Jastrow	3(4)	0	5(7)	-9(8)
x = 0.333 Jastrow	-7(5)	4(9)	0	6(9)
x = 0.667 Jastrow	-9(7)	-2(8)	-2(6)	0
x = 0.875 Jastrow	-8(4)	-5(8)	-2(6)	-3(7)

\*Energy differences  
(in meV) between  
DMC with self  
Jastrows and shared  
Jastrows)

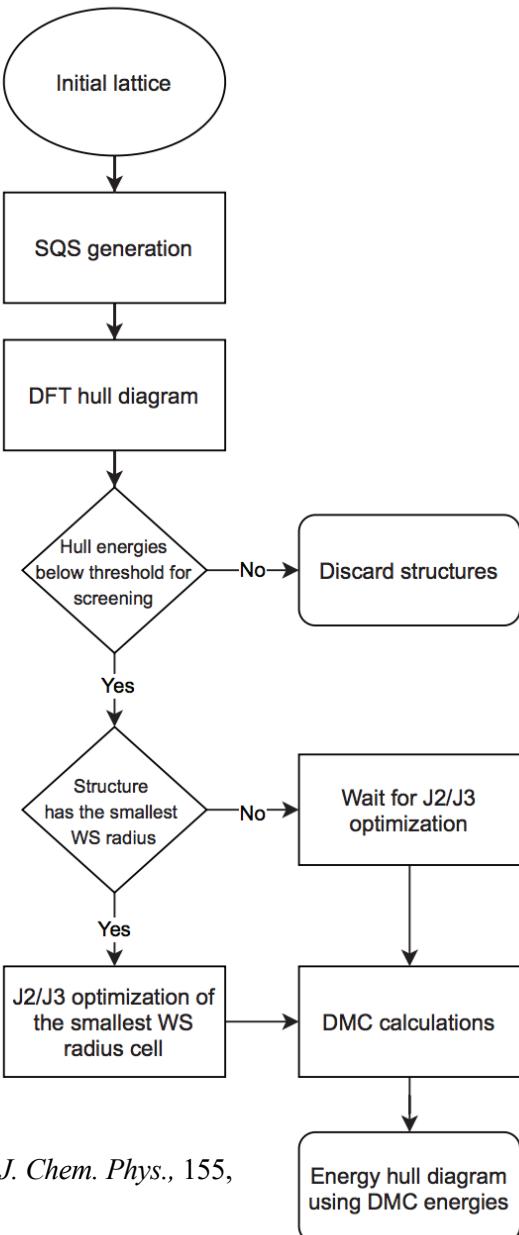
- Sharing Jastrows nearly **identical** at the DMC level (same simulation settings)
- Error is still decreased no matter which Jastrows are used
- Most convenient to **reuse** Jastrows from cell with **smallest Wigner-Seitz radius**

# DMC Energy Hull of $\text{GaS}_x\text{Se}_{1-x}$



- DMC calculated energy **hull diagram**
- Comparison of DMC energies calculated with alloy's **self Jastrows** versus **Jastrows** of the **smallest WS** cell, **nearly identical**
- DFT and DMC are in **close agreement**, beneficial for benchmarking our hull diagram for **low** and **high**  $\text{E}_{\text{form}}$  structures

# Workflow and Cost Analysis



VMC Computational Time (seconds)

	1 Supercell	Extrapolation	Energy Hull
Self Jastrow	755	7,124	24,043
WS Jastrow	134	134	134

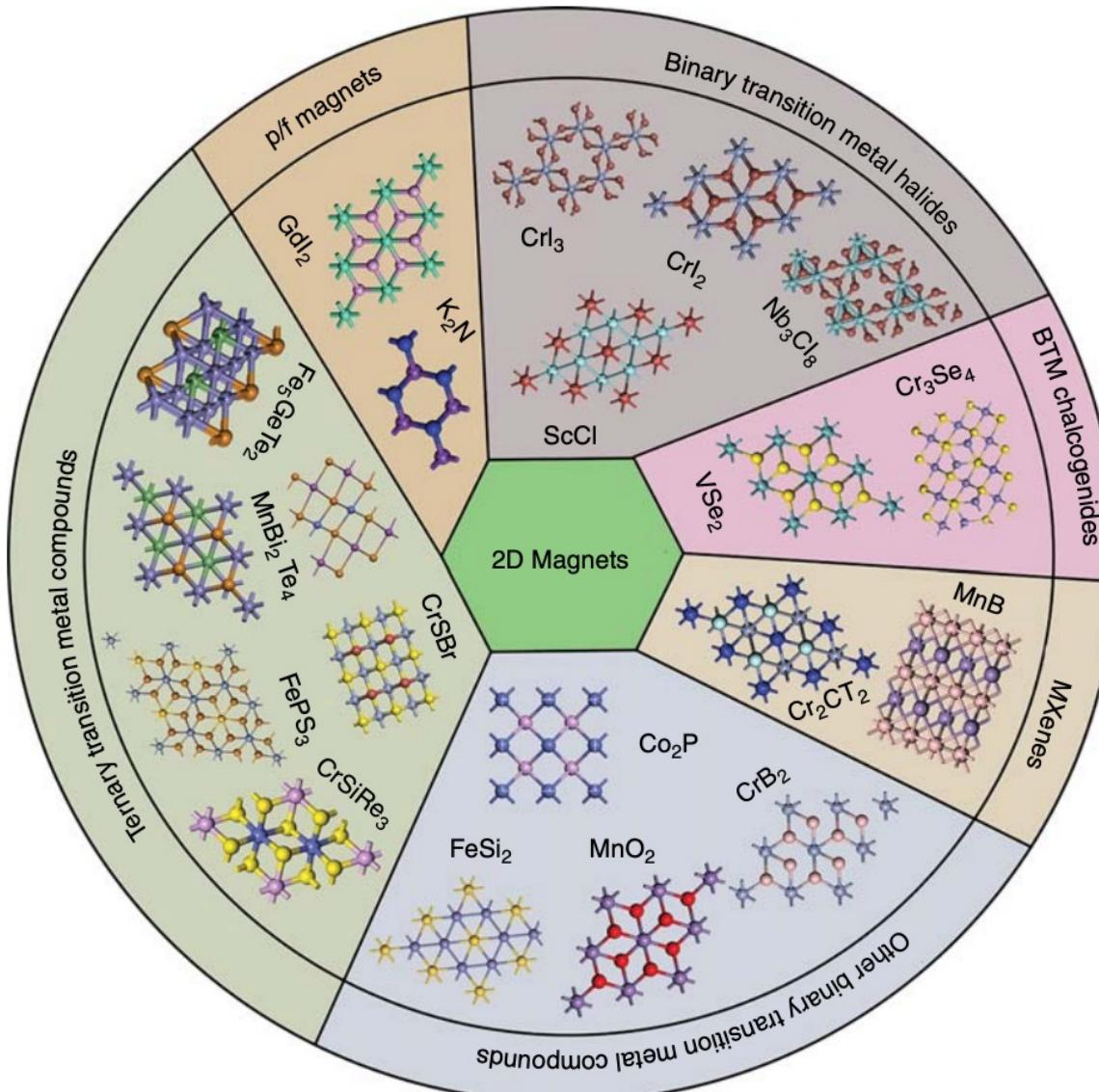
DMC Computational Time (seconds)

	1 Supercell	Extrapolation	Energy Hull
Self Jastrow	2,143	32,899	93,066
WS Jastrow	2,109	33,259	93,170

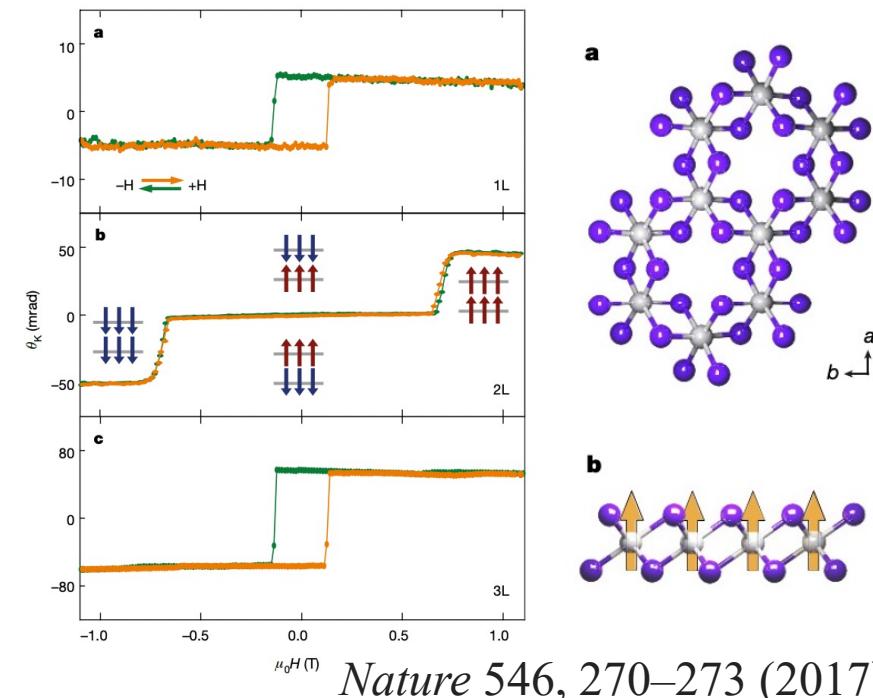
- By using WS Jastrow, **extra VMC is avoided** (additional supercell sizes, stoichiometries)
- When self Jastrows are used, VMC accounts for **substantial amount of total cost (~1/4)**, cannot be neglected
- Jastrow **does not change the DMC time** needed to **achieve target error bar**

# 2D Magnetic Materials

NIST

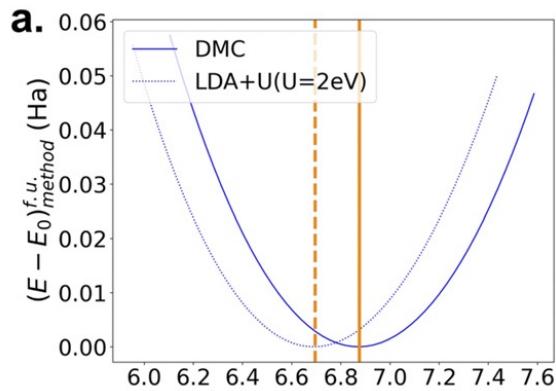
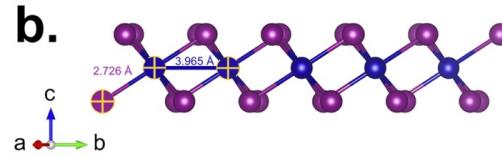
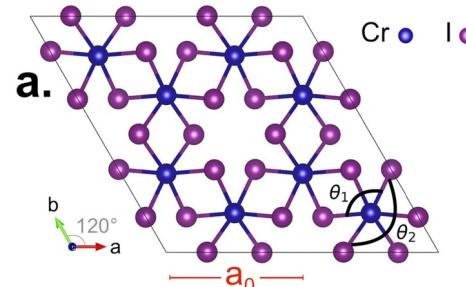


- **2D magnets** are important class of materials for **spintronic** devices and **information storage**
- **Correlated** systems that are **difficult** to accurately model (**DFT+U**)
- Calculations can be **revisited** with accurate **many-body techniques**

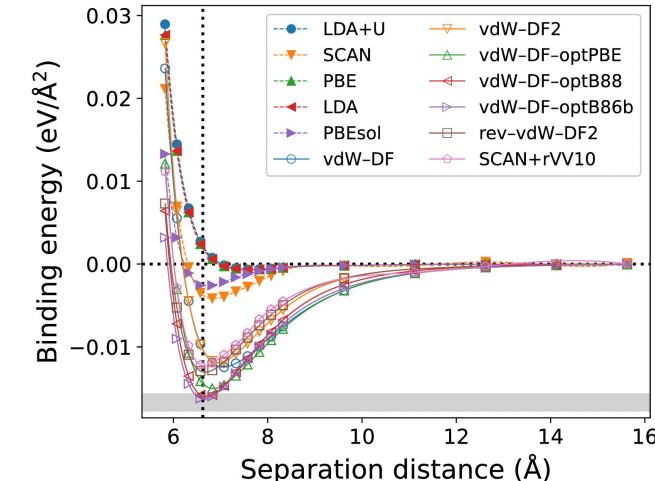
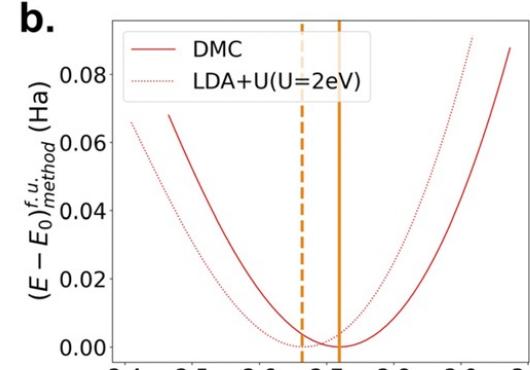


# QMC for Magnetic Materials

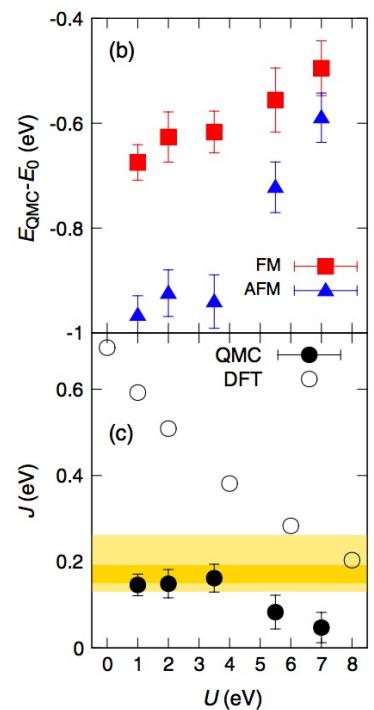
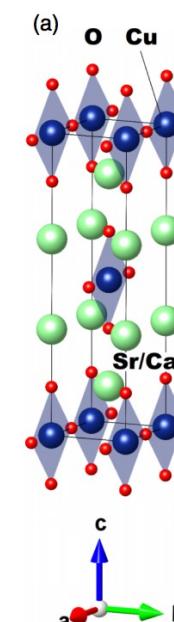
NIST



Staros et al. *J. Chem. Phys.* 156, 014707 (2022)



Ichibha et al.  
*Phys. Rev. Materials* 5, 064006 (2021)



- **Magnetic Materials** have been studied with many-body **QMC** methods
- Magnetic (moment,  $T_c$ ,  $J$ ) structural, interlayer binding

Foyevtsova et al., *Phys. Rev. X* 4, 031003 (2014)

# 2D Model Hamiltonian

$$\mathcal{H} = - \left( \sum_i \textcolor{blue}{D} (S_i^z)^2 + \frac{\textcolor{red}{J}}{2} \sum_{i,i'} \vec{S}_i \cdot \vec{S}_{i'} + \frac{\textcolor{green}{\lambda}}{2} \sum_{i,i'} S_i^z S_{i'}^z \right)$$

Magnetic atoms

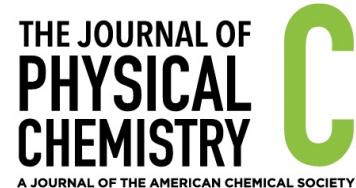
**$J$**  Isotropic Heisenberg Exchange

**$D$**  Easy Axis Single Ion Anisotropy

**$\lambda$**  Anisotropic Symmetric Exchange

# 2D CrX<sub>3</sub> Magnets

NIST



[pubs.acs.org/JPCC](https://pubs.acs.org/JPCC)

JARVIS  Beyond DFT

Article

## Systematic DFT+U and Quantum Monte Carlo Benchmark of Magnetic Two-Dimensional (2D) CrX<sub>3</sub> (X = I, Br, Cl, F)

Daniel Wines,\* Kamal Choudhary, and Francesca Tavazza

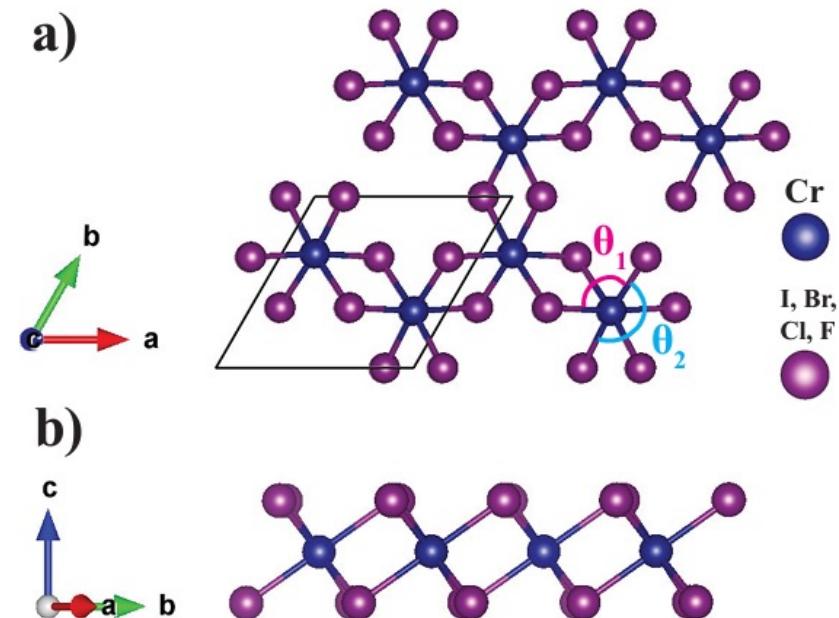


Cite This: *J. Phys. Chem. C* 2023, 127, 1176–1188



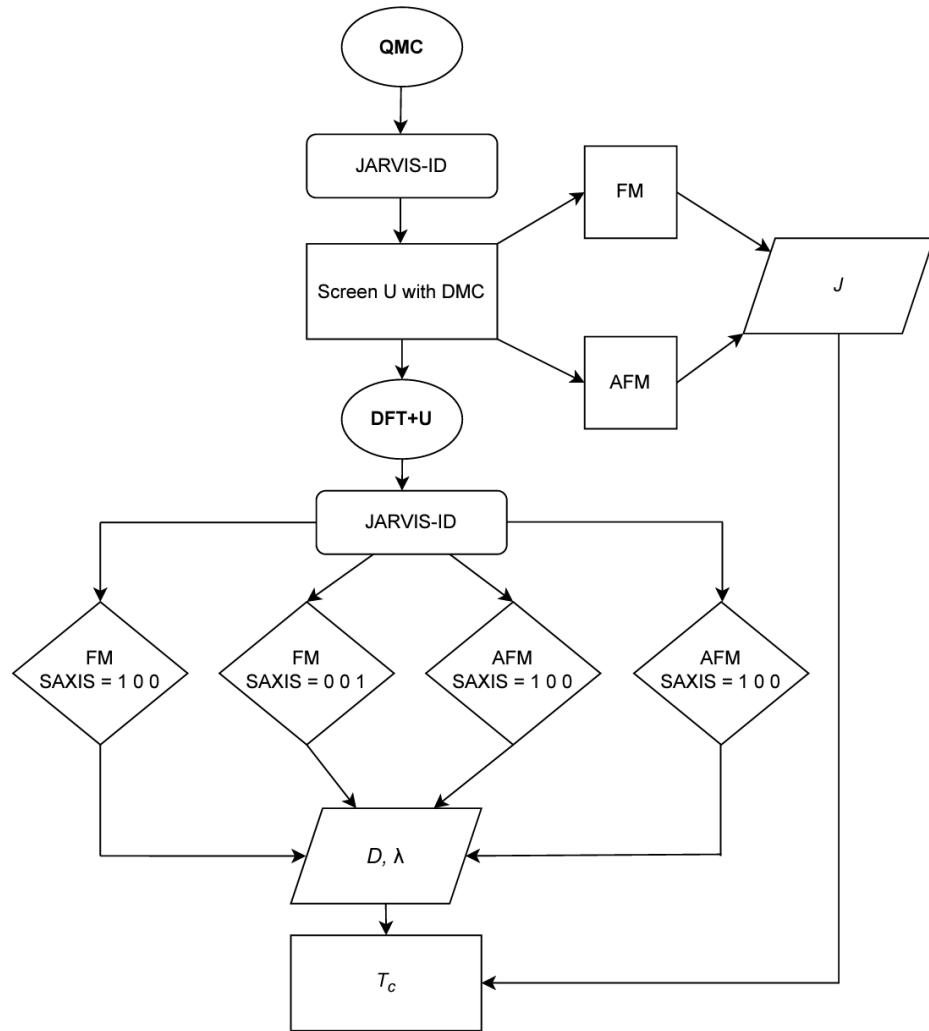
Read Online

- Case study of 2D correlated magnets with CrX<sub>3</sub> stoichiometry
- QMC added to JARVIS framework



# 2D CrX<sub>3</sub> Magnets

NIST



2D Model Spin Hamiltonian:

$$\mathcal{H} = - \left( \sum_i \textcolor{blue}{D} (S_i^z)^2 + \frac{\textcolor{red}{J}}{2} \sum_{i,i'} \vec{S}_i \cdot \vec{S}_{i'} + \frac{\textcolor{green}{\lambda}}{2} \sum_{i,i'} S_i^z S_{i'}^z \right)$$

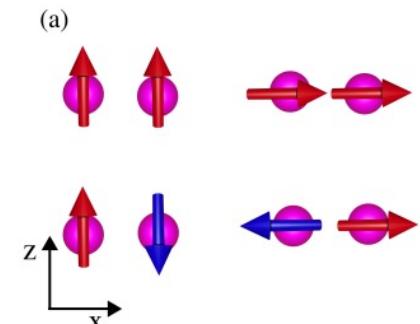
$\textcolor{red}{J}$  Isotropic Heisenberg Exchange

$\textcolor{blue}{D}$  Easy Axis Single Ion Anisotropy

$\textcolor{green}{\lambda}$  Anisotropic Symmetric Exchange

CrI <sub>3</sub>	$J$ (meV)	$D$ (meV)	$\lambda$ (meV)	$T_c$ (K)
PBE	2.83	0.192	0.173	38.33
PBE+U	3.70	0.075	0.161	43.60
LDA	1.98	0.139	0.126	27.09
LDA+U	2.80	0.054	0.147	34.21
SCAN	3.24	-0.129	0.019	-
SCAN+U	4.06	-0.069	0.216	46.23
r2SCAN	2.57	0.145	0.049	29.00
r2SCAN+U	3.08	0.146	0.044	32.91
HSE06	4.22	0.068	0.173	48.63

Strong variability  
in DFT results



©Nicholas Michtzsch

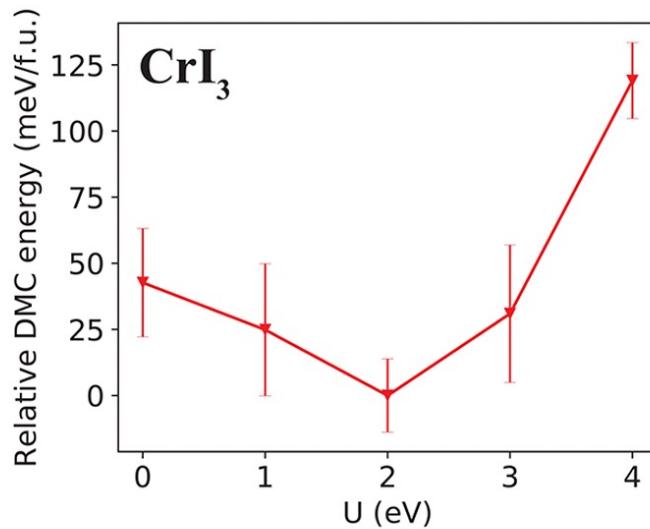
\* $T_c$  (Curie Temperature)  
estimated by method of  
Torelli and Olsen

2D Materials, 6, 015028 (2019)

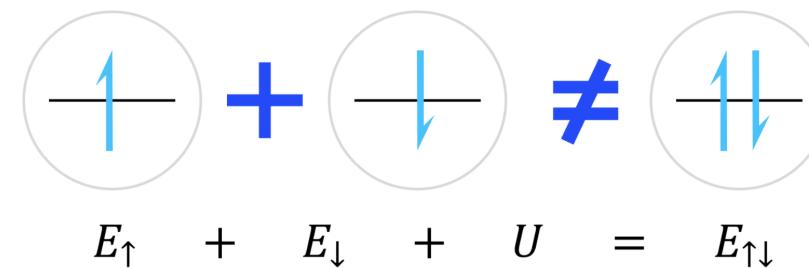
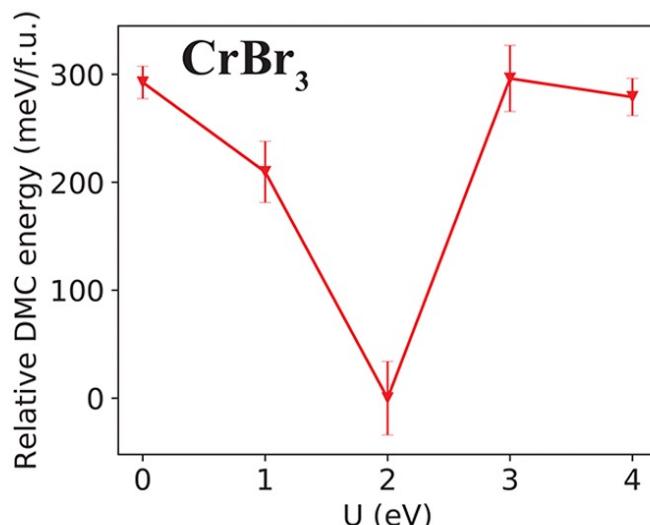
# 2D CrX<sub>3</sub> Magnets

NIST

a)



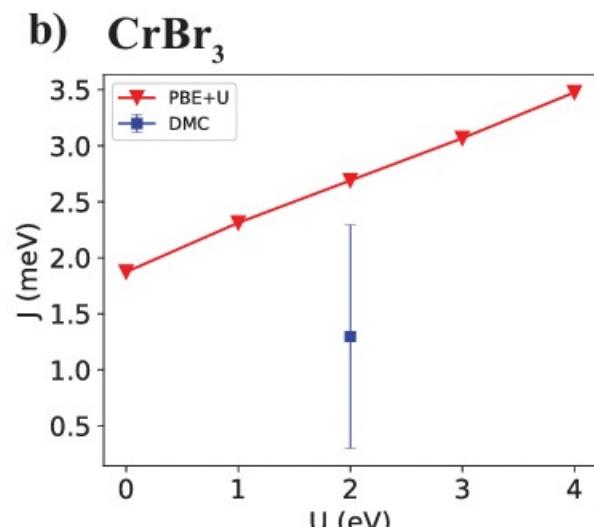
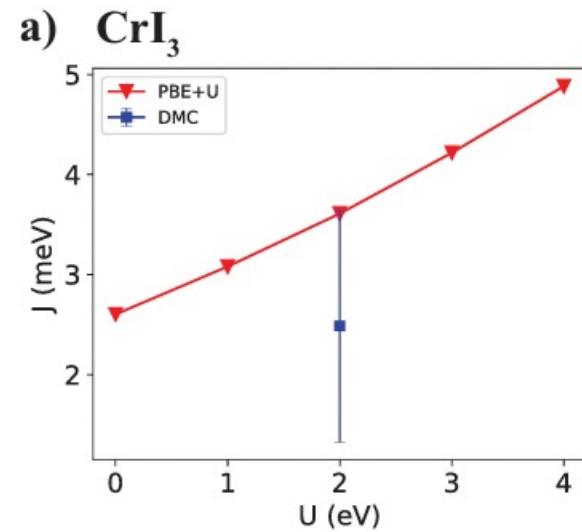
b)



- Optimal trial WF can be created by tuning U parameter
- U = 2 eV variationally yields optimal WF

# 2D CrX<sub>3</sub> Magnets

NIST



- Accurate **statistical bound** on magnetic exchange and Curie Temperature
- Maximum T<sub>c</sub>: **43.56 K** for CrI<sub>3</sub> and **20.78 K** for CrBr<sub>3</sub>
- **Less dependence** on starting functional and Hubbard (U) parameter
- Same workflow can be applied to other 2D ferromagnets
- **Goal: JARVIS-QMC database**

# JARVIS-QMC: 2D CrX<sub>3</sub> Magnets

NIST

CrI<sub>3</sub>

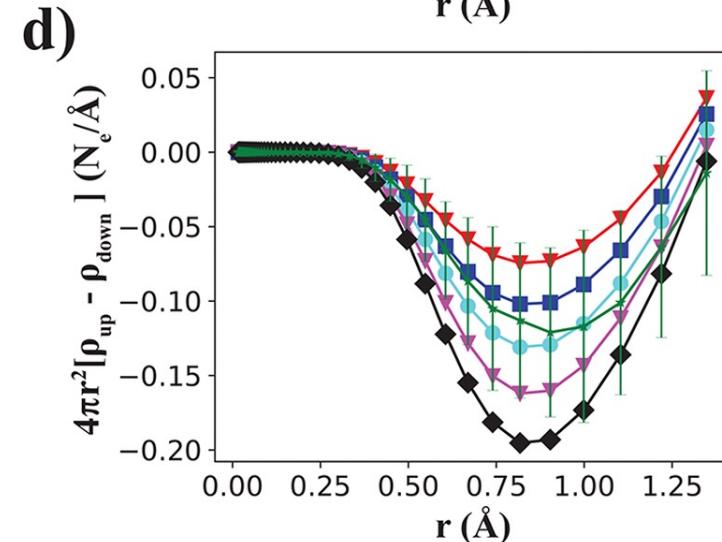
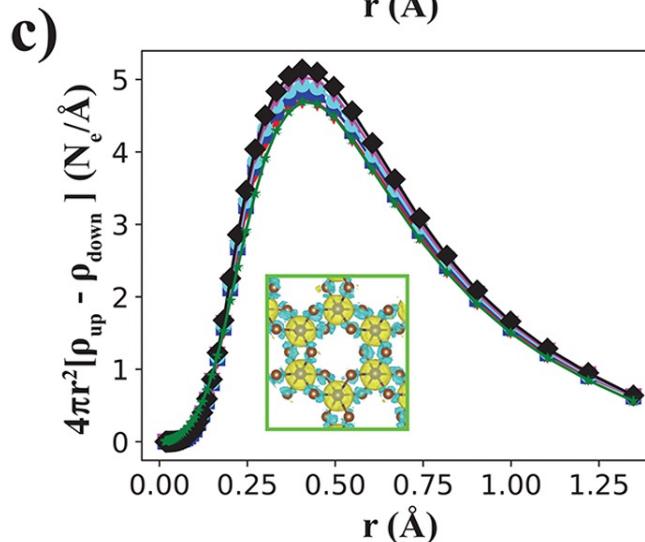
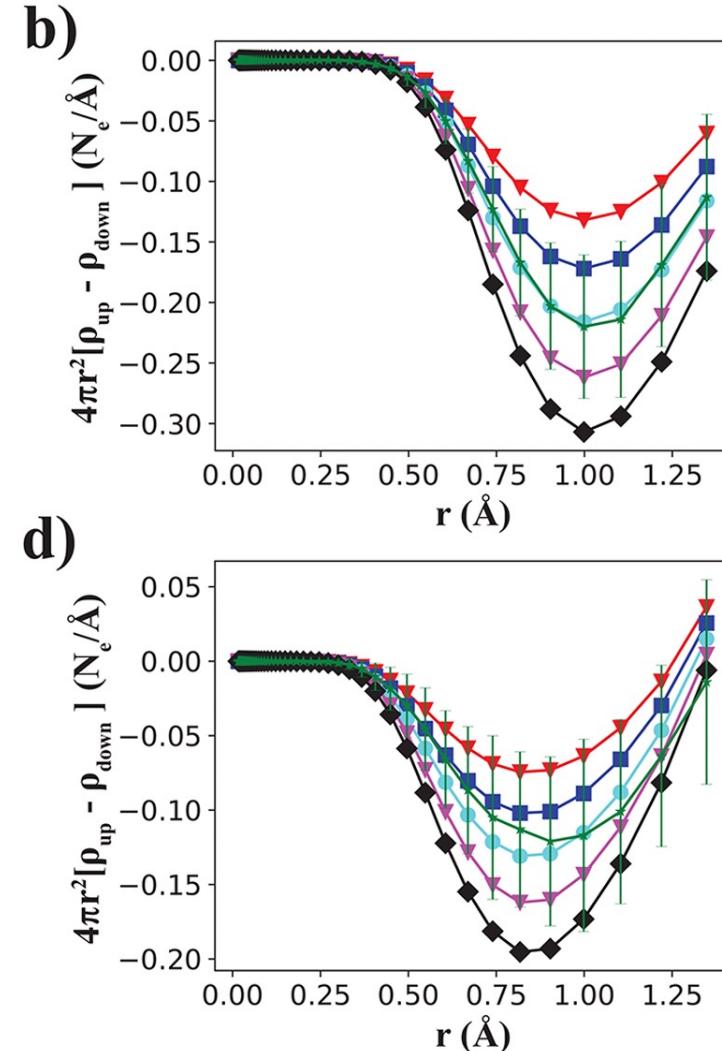
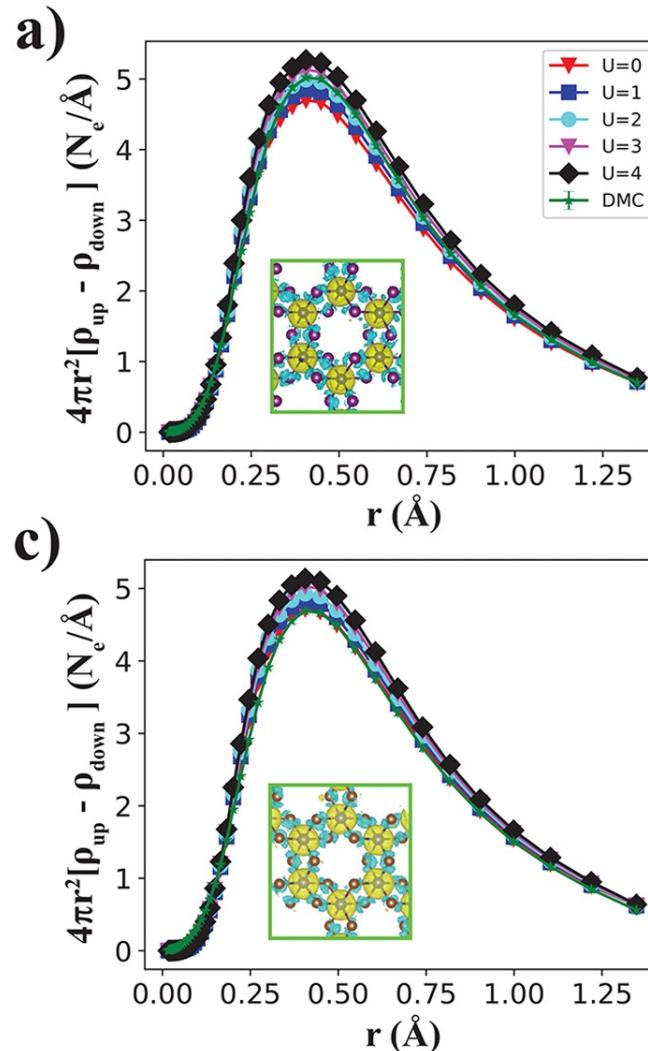
Method	$J$ (meV)	$T_c^{\text{Ising}}$ (K)	$T_c^{\text{PBE+2}}$ (K)	$T_c^{\text{LDA+2}}$ (K)	$T_c^{\text{SCAN+2}}$ (K)	$T_c^{\text{r2SCAN+2}}$ (K)	$T_c^{\text{HSE06}}$ (K)
PBE+0	2.60	103.26	33.46	32.40	33.10	29.00	33.81
PBE+1	3.08	122.23	37.98	36.78	37.57	32.91	38.38
PBE+2	3.61	143.23	42.79	41.42	42.32	37.07	43.23
PBE+3	4.22	167.39	48.10	46.57	47.58	41.67	48.60
PBE+4	4.88	193.84	53.70	51.99	53.12	46.52	54.26
DMC	2.49(1.16)	98.65(46.02)	32.34(10.78)	31.31(10.43)	31.98(10.66)	28.02(9.33)	32.67(10.89)

CrBr<sub>3</sub>

Method	$J$ (meV)	$T_c^{\text{Ising}}$ (K)	$T_c^{\text{PBE+2}}$ (K)	$T_c^{\text{LDA+2}}$ (K)	$T_c^{\text{SCAN+2}}$ (K)	$T_c^{\text{r2SCAN+2}}$ (K)	$T_c^{\text{HSE06}}$ (K)
PBE+0	1.87	74.31	17.70	17.84	-	15.98	14.52
PBE+1	2.31	91.76	20.74	20.90	-	18.72	17.01
PBE+2	2.69	106.74	23.23	23.41	-	20.97	19.05
PBE+3	3.07	121.72	25.63	25.84	-	23.14	21.03
PBE+4	3.48	137.97	28.16	28.38	-	25.42	23.10
DMC	1.30(1.00)	51.45(39.52)	13.43(7.17)	13.54(7.23)	-	12.13(6.47)	11.02(5.88)

# JARVIS-QMC: 2D CrX<sub>3</sub> Magnets

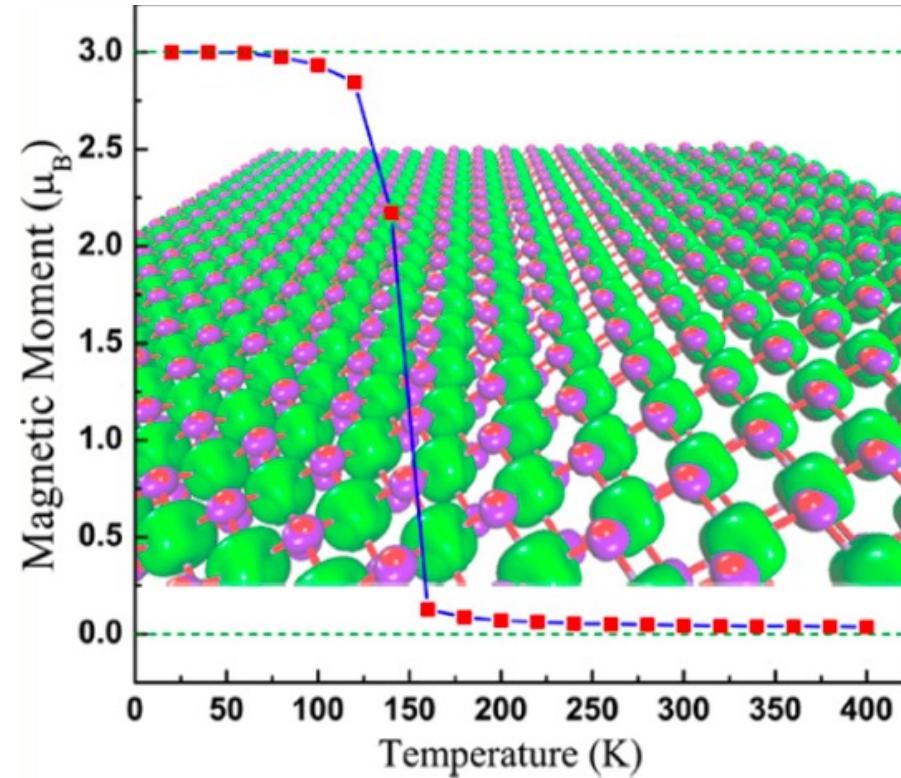
NIST



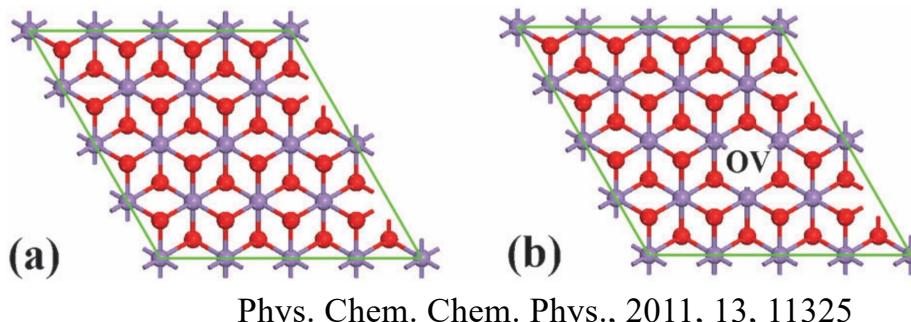
CrI <sub>3</sub>		
Method	$M_{\text{Cr}} (\mu_B)$	$M_{\text{I}} (\mu_B)$
PBE+0	3.06	-0.08
PBE+1	3.15	-0.1
PBE+2	3.25	-0.13
PBE+3	3.34	-0.16
PBE+4	3.43	-0.18
DMC	3.21(5)	-0.13(5)
CrBr <sub>3</sub>		
Method	$M_{\text{Cr}} (\mu_B)$	$M_{\text{Br}} (\mu_B)$
PBE+0	3.01	-0.03
PBE+1	3.07	-0.05
PBE+2	3.14	-0.07
PBE+3	3.2	-0.09
PBE+4	3.26	-0.11
DMC	2.96(5)	-0.07(5)

- Can obtain accurate estimates for **spin density** and **magnetic moment** with **DMC**

# Motivation: 2D MnO<sub>2</sub> with QMC



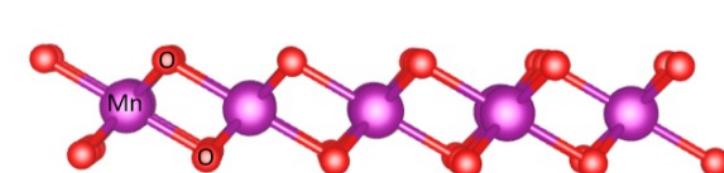
J. Phys. Chem. Lett. 2013, 4, 3382



Phys. Chem. Chem. Phys., 2011, 13, 11325

## Motivation

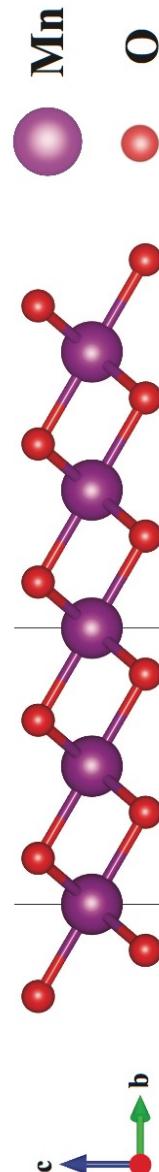
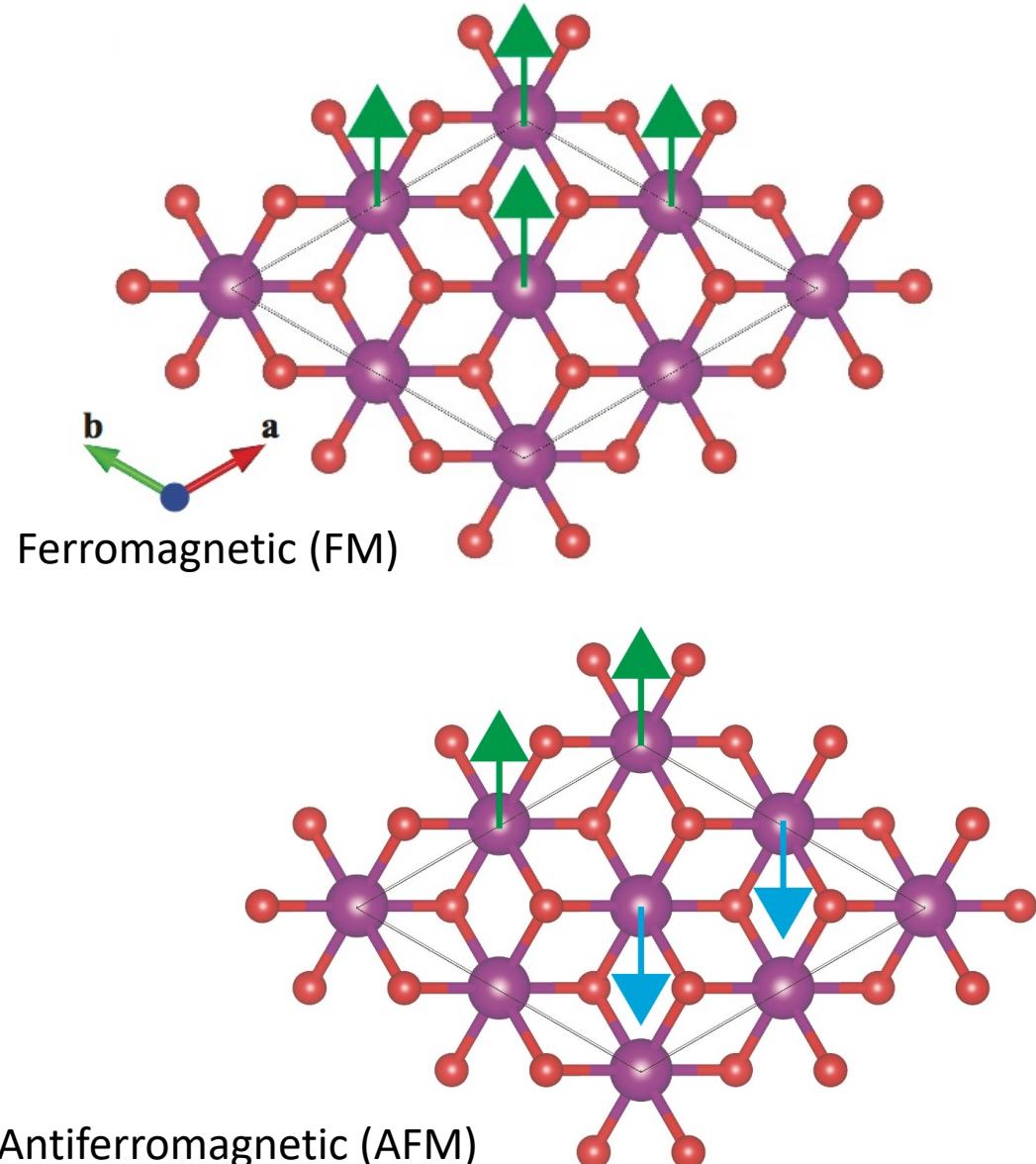
- 2D MnO<sub>2</sub> has been reliably synthesized
- Semiconducting
- **Predicted ferromagnetic**
- T and H phase predicted stable



[https://www.2dsemiconductors.com/2D\\_mno2/](https://www.2dsemiconductors.com/2D_mno2/)



# 2D MnO<sub>2</sub>: DFT Benchmark

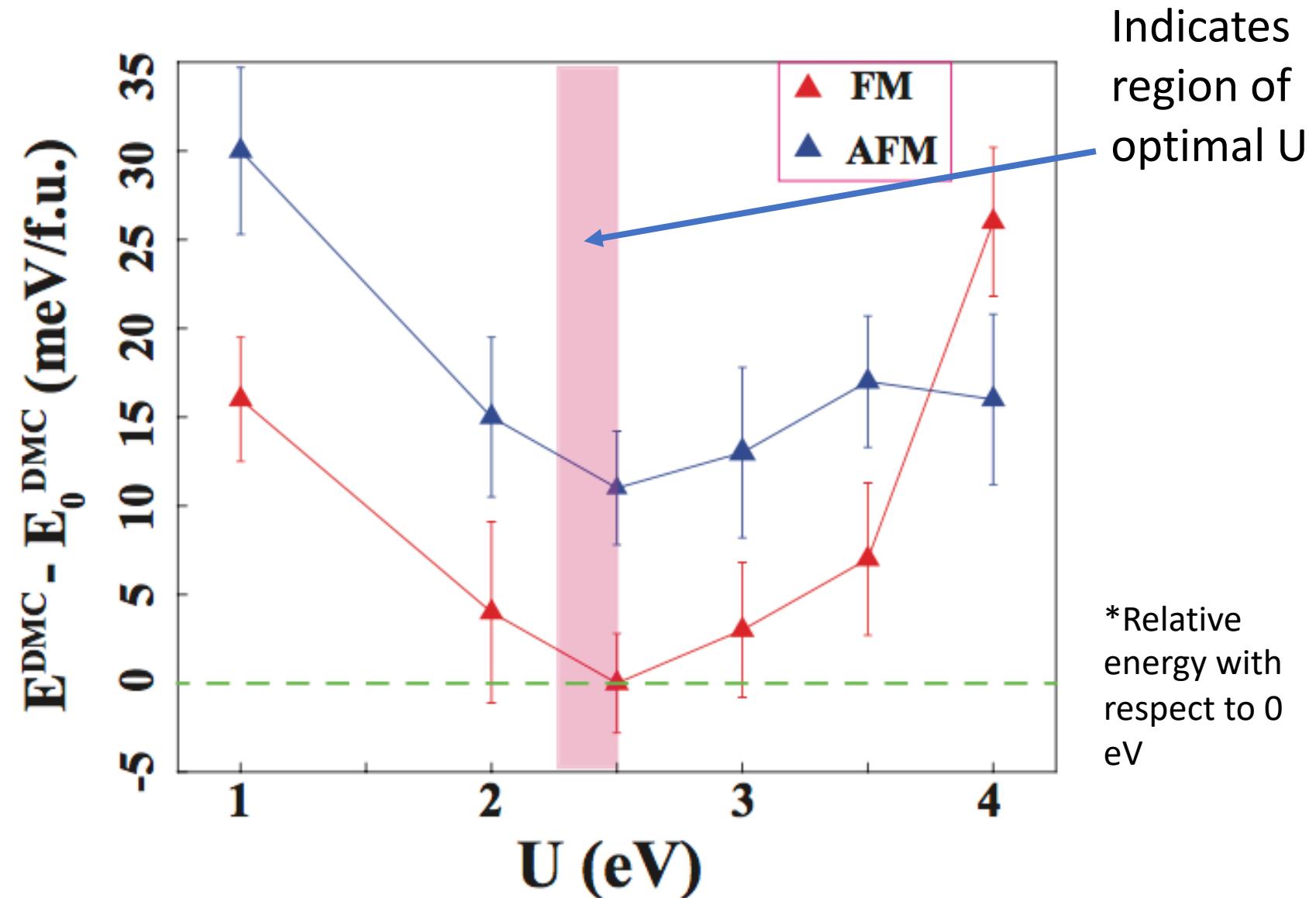


Functional	$E_{FM} - E_{AFM}$ (meV)
PBE, U=0	-8.2
PBE, U=2	-36.6
PBE, U=3.5	-55.0
SCAN, U=0	-3.1
SCAN, U=2	-16.0
SCAN, U=3.5	-28.7
LDA, U=0	18.0
LDA, U=2	-25.4
LDA, U=3.5	-44.1

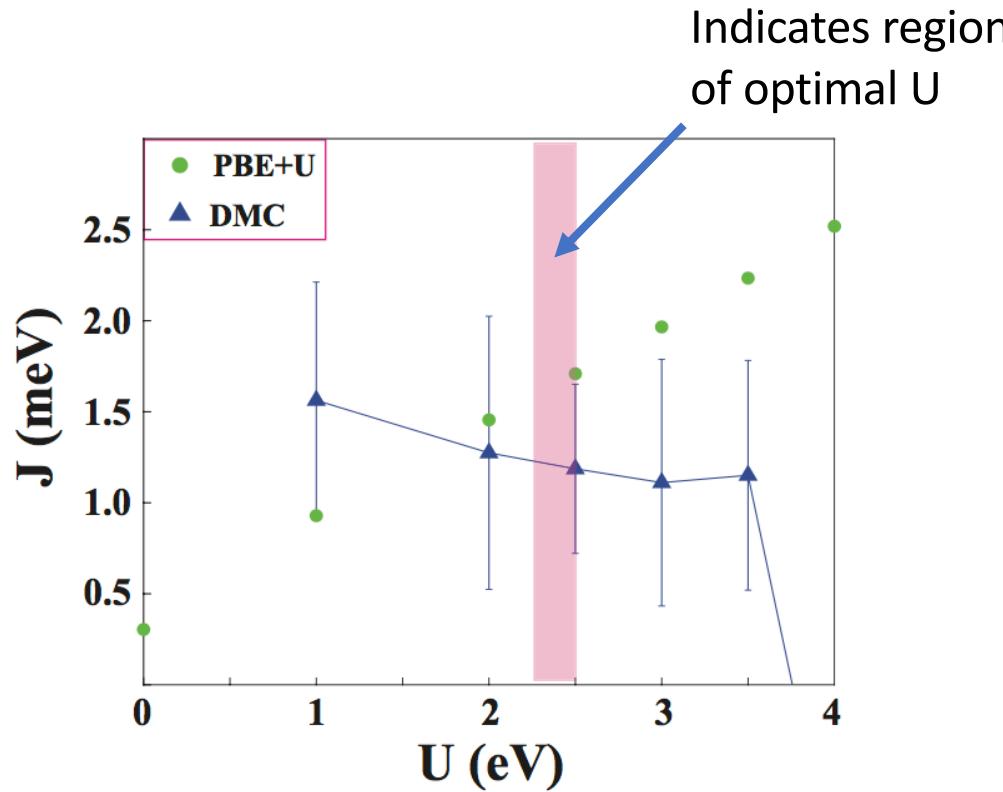
Discrepancies  
in FM/AFM  
favorability  
with DFT  
functionals  
and U

# 2D MnO<sub>2</sub>: FM and AFM with QMC

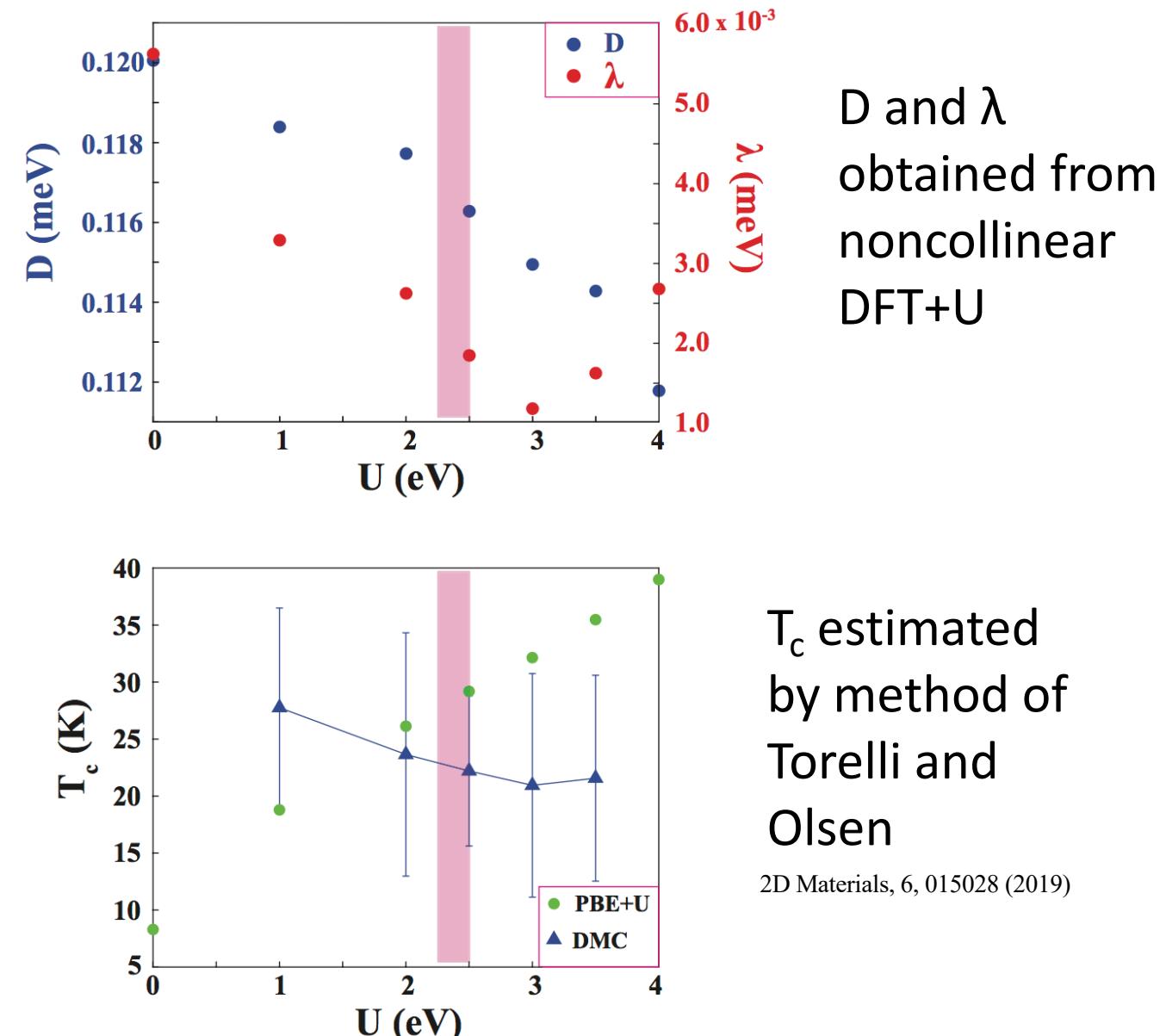
- Optimal trial WF for DMC can variationally be determined by changing U
- U = 2.5 eV fitted to be optimal U value
- DMC has much weaker dependence on U value than DFT



# 2D MnO<sub>2</sub>: Magnetic Properties



- 2D MnO<sub>2</sub> confirmed to be 2D ferromagnet with out of plane anisotropy
- Upper bound on T<sub>c</sub> : 28.8 K

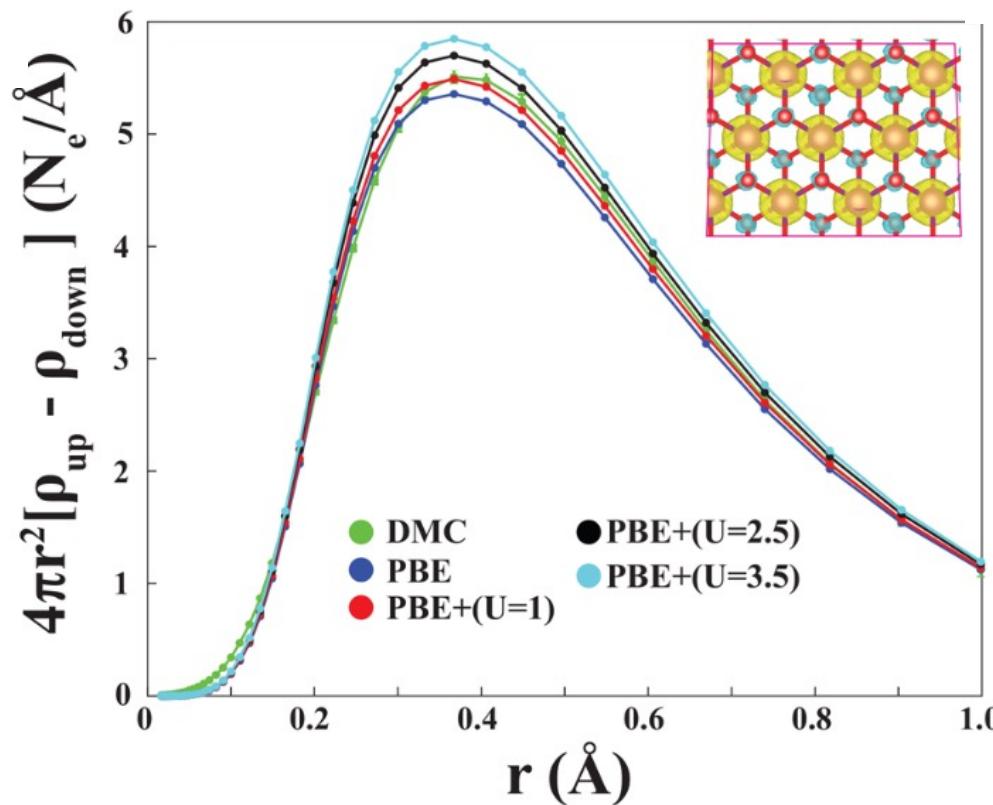


D and  $\lambda$   
obtained from  
noncollinear  
DFT+U

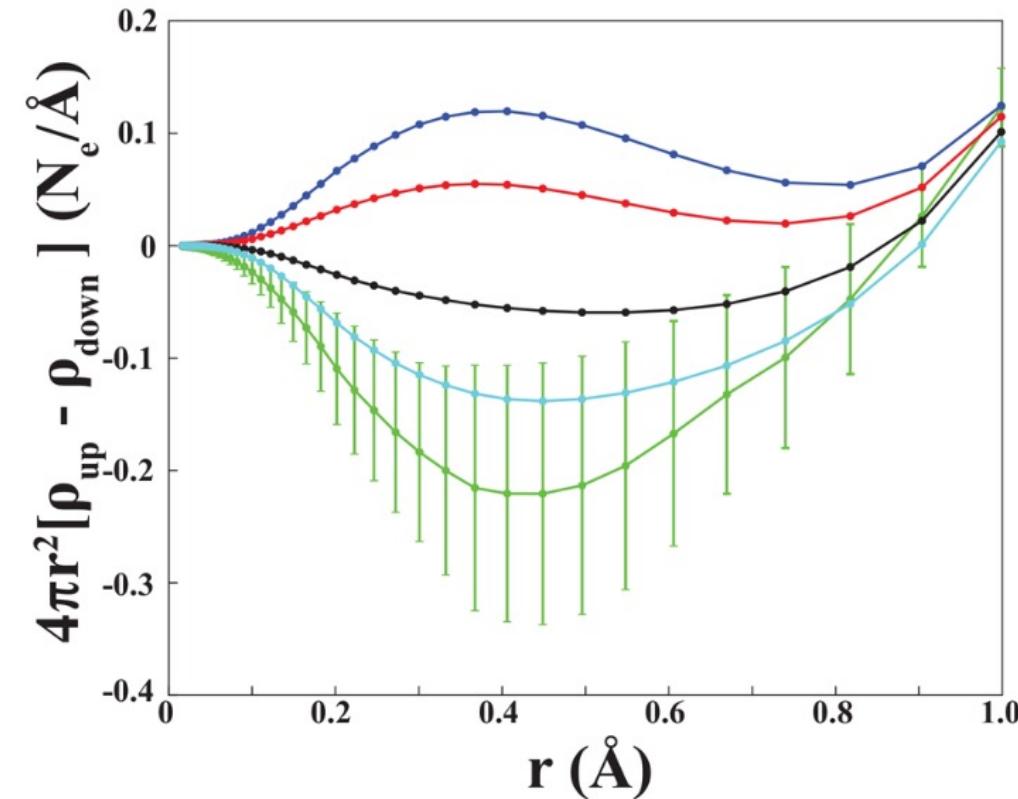
T<sub>c</sub> estimated  
by method of  
Torelli and  
Olsen

2D Materials, 6, 015028 (2019)

# 2D MnO<sub>2</sub>: Magnetic Properties



method	$M_{\text{Mn}} (\mu_B)$	$M_{\text{O}} (\mu_B)$
DMC	2.77(1)	-0.10(2)
PBE, $U = 0$	2.69	0.06
PBE, $U = 1$	2.76	0.03
PBE, $U = 2.5$	2.86	-0.03
PBE, $U = 3.5$	2.93	-0.07



- Spin density of O polarized antiparallel with respect to Mn with DMC

# QMC: 2D T- and H-VSe<sub>2</sub>

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Letter

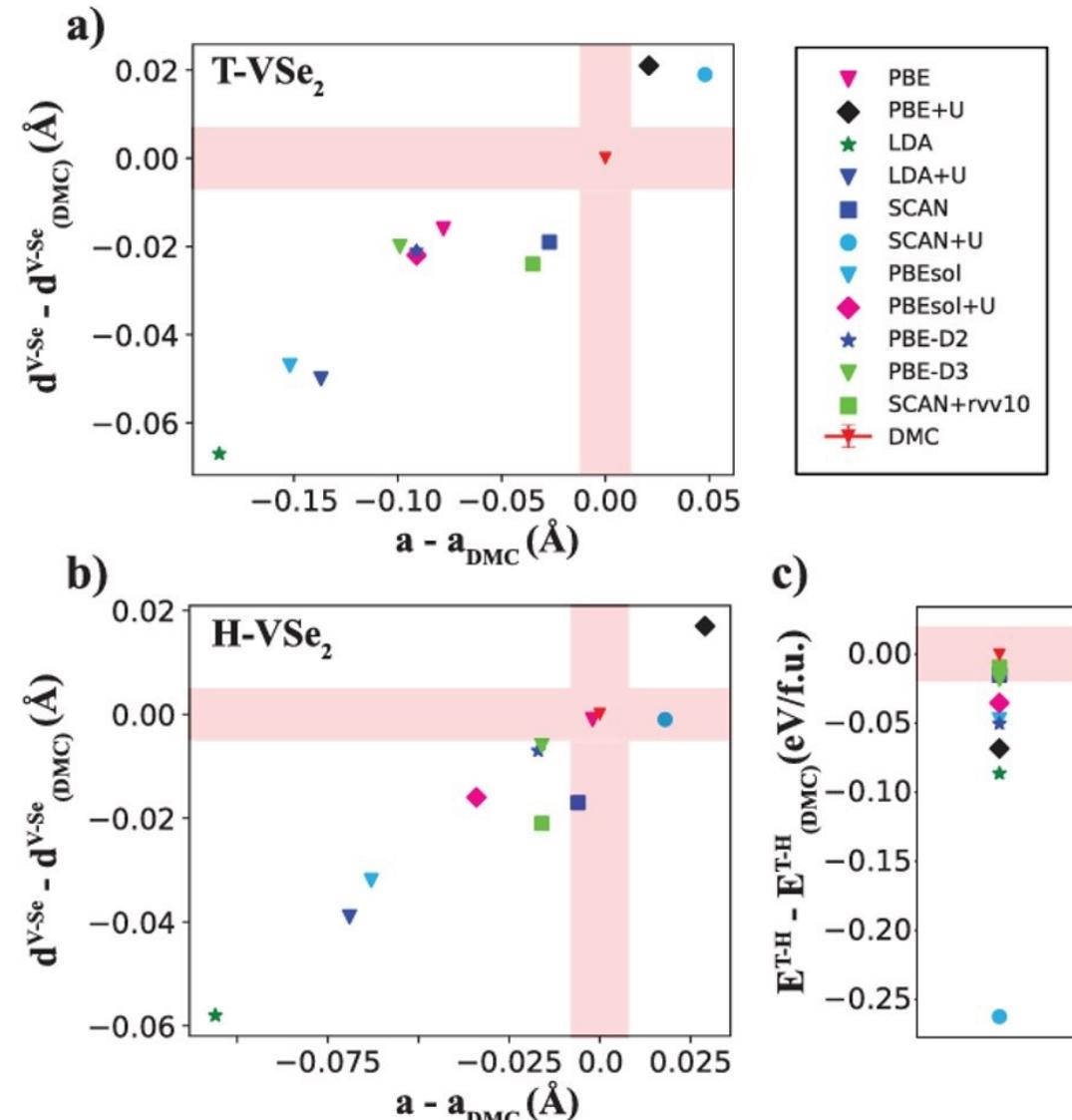
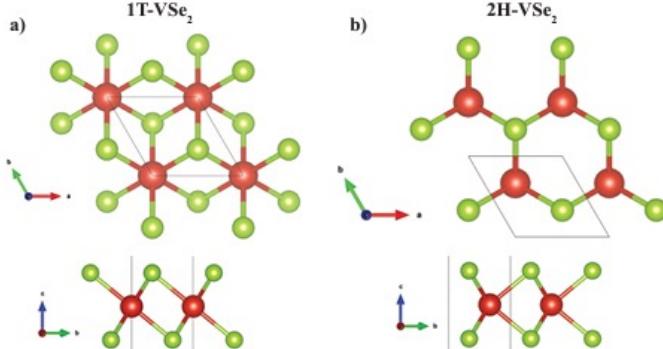
## A Quantum Monte Carlo Study of the Structural, Energetic, and Magnetic Properties of Two-Dimensional H and T Phase VSe<sub>2</sub>

Daniel Wines, Juha Tiihonen, Kayahan Saritas, Jaron T. Krogel, and Can Ataca\*

Cite This: *J. Phys. Chem. Lett.* 2023, 14, 3553–3560

Read Online

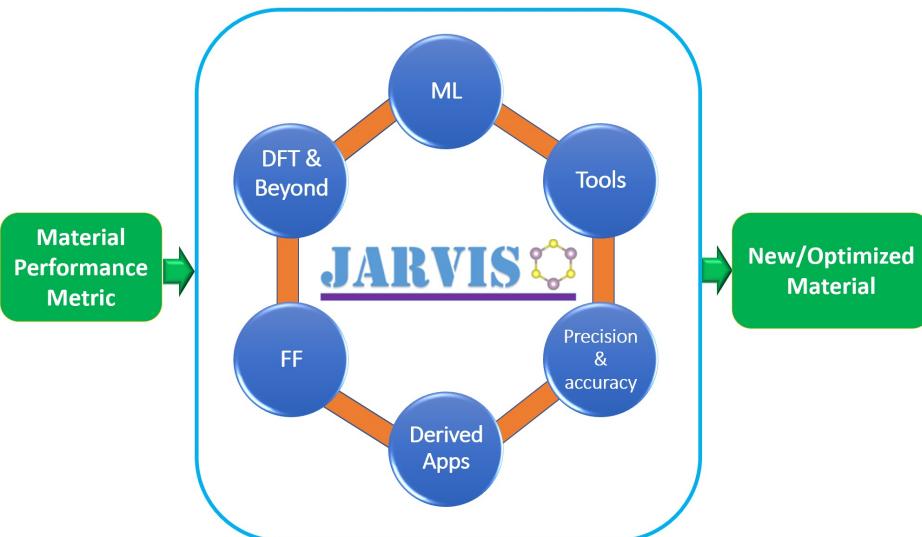
- Structural optimization using QMC methods
- Accurate magnetic properties of T-VSe<sub>2</sub>
- **Covered in detail by Prof. Ataca (12/14)**



# JARVIS: Databases, Tools, Events, Outreach



<https://jarvis.nist.gov>



Screenshots of the JARVIS website sections: Resources, Density functional theory, Force-field, JARVIS-QETB, JARVIS-ALIGNN, Machine learning (CFID), Wannier tight-binding, Databases, and Help.

**Established:** January 2017

**Published:** >40 articles

**Users:** >20000+ users worldwide

**Materials:** >80000, millions of properties

**Events:**

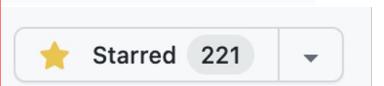
- Quantum Matters in Materials Science (QMMS)
- Artificial Intelligence for Materials Science (AIMS)
- JARVIS-School



[usnistgov / jarvis](https://usnistgov/jarvis)

User-comments:

- “There are many different theoretical levels on which you can approach the field. JARVIS is unusual in that it spans more levels than other databases.”
- “A pure gold-mine for the data-quality effort...”
- “Thanks for your generous sharing. Your works inspire me a lot.”
- “You guys are doing something really beneficial...”
- “I find JARVIS-DFT very useful for my research...”



downloads 220k

# Recent Updates to JARVIS

NIST

Applied Physics Reviews

REVIEW

[pubs.aip.org/aip/are](https://pubs.aip.org/aip/are)

## Recent progress in the JARVIS infrastructure for next-generation data-driven materials design

Cite as: Appl. Phys. Rev. **10**, 041302 (2023); doi: [10.1063/5.0159299](https://doi.org/10.1063/5.0159299)

Submitted: 22 May 2023 · Accepted: 18 September 2023 ·

Published Online: 18 October 2023



View Online



Export Citation



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Daniel Wines,<sup>1</sup> Ramya Gurunathan,<sup>1</sup> Kevin F. Garrity,<sup>1</sup> Brian DeCost,<sup>1</sup> Adam J. Biacchi,<sup>2</sup> Francesca Tavazza,<sup>1</sup> and Kamal Choudhary<sup>1,a)</sup>

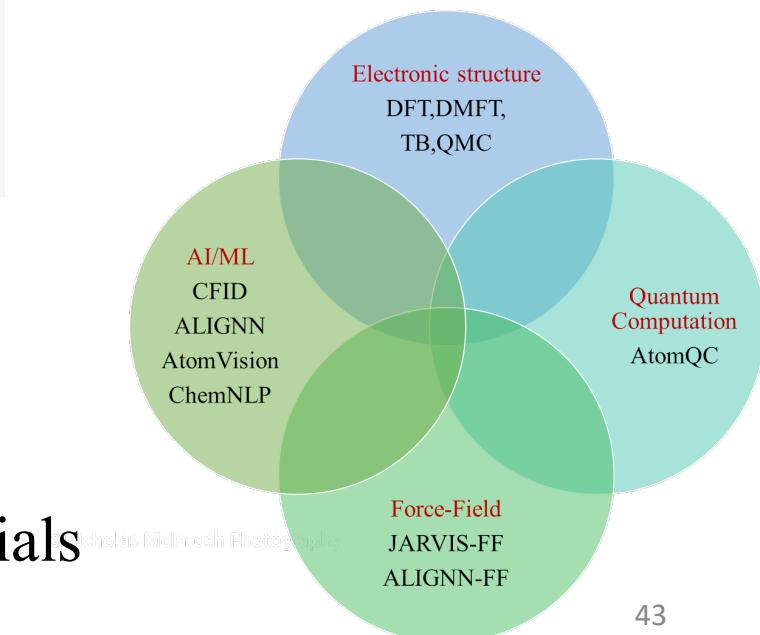
### AFFILIATIONS

<sup>1</sup>Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

<sup>2</sup>Physical Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA

## Updates

- 80,000 materials
- **QMC**, tight binding, ALIGNN, ALIGNN-FF, AtomVision
- Quantum Computation algorithms
- Superconductors (bulk and 2D), magnetic topological materials
- JARVIS-Leaderboard



# JARVIS-Leaderboard: Large Scale Benchmark

## Challenges in materials science community:

- Reproducibility
- Transparency
- Validation
- Fidelity
- Data vs. metadata
- What is the ground truth/reference to compare our models to?  
How does this change depending on the model?
- Synergy of computational and experimental databases



[https://pages.nist.gov/jarvis\\_leaderboard/](https://pages.nist.gov/jarvis_leaderboard/)

Community  
effort to tackle  
challenges:

The screenshot shows the JARVIS Leaderboard website. The header is teal with the "jarvis\_leaderboard" logo and version information (v2023.05.13). The main navigation bar has links for Home, AI, ES, EXP, FF, and QC. The page title is "JARVIS Leaderboard (Introduction)". The content area explains the project's purpose: "This project provides benchmark performances of various methods for materials science applications using the datasets available in JARVIS-Tools databases. Some of the categories are: Artificial Intelligence (AI), Electronic Structure (ES), Force-field (FF), Quantum Computation (QC) and Experiments (EXP). There are a variety of properties included in the benchmark. In addition to prediction results, we attempt to capture the underlying software, hardware and instrumental frameworks to enhance reproducibility. This project is a part of the NIST-JARVIS infrastructure."

# Categories of benchmarks

The screenshot shows a red header with the arXiv logo and navigation links. Below it, the title 'Large Scale Benchmark of Materials Design Methods' is displayed, along with the names of over 30 authors. The main content area contains a table with columns for 'Category', 'General name', and 'Method Specification'. The table lists various software tools and their specifications across categories like Electronic Structure (ES), Artificial Intelligence (AI), Force Field (FF), Quantum Computation (QC), and Experiment (EXP).

Category	General name	Method Specification
ES	DFT <sup>64</sup>	VASP <sup>52,53</sup> (PBE <sup>54</sup> , LDA <sup>64</sup> , OptB88vdW <sup>65</sup> , Opt86BvdW <sup>66</sup> , TBmBJ <sup>67,68</sup> , SCAN <sup>69</sup> , r2SCAN <sup>70</sup> , HSE06 <sup>71</sup> ) QE <sup>72</sup> (PBE <sup>54</sup> , PBEsol <sup>73</sup> ) ABINIT <sup>74-76</sup> (PBE <sup>54</sup> ) GPAW <sup>77</sup> (PBE <sup>54</sup> , LDA <sup>64</sup> , GLLB-sc <sup>78</sup> )
	QMC <sup>61</sup>	QMCPACK <sup>79</sup> (DMC <sup>61</sup> )
	GW <sup>63</sup>	VASP <sup>52,53</sup> ( $G_0W_0$ <sup>63</sup> , GW <sub>0</sub> <sup>63</sup> )
	TB <sup>57</sup>	ThreeBodyTB.jl <sup>59</sup> (Wannier90 <sup>80</sup> )
AI	Descriptor	CFID <sup>81</sup> , MagPie <sup>82</sup> , MatMiner <sup>55,83</sup> , crystal feature model <sup>84</sup> , ELEMNet <sup>85-87</sup> , IRNet <sup>88-90</sup> , BRNet <sup>91,92</sup> , SNAP <sup>93</sup>
	Graph-based	ALIGNN <sup>94</sup> , CGCNN <sup>95</sup> , SchNet <sup>96</sup> , AtomVision <sup>97</sup> , ChemNLP <sup>98</sup> , DimeNet <sup>99,100</sup> , CHGNet <sup>101</sup> , M3GNET <sup>102</sup> , kgcnn_coGN <sup>103</sup> , Potnet <sup>104</sup> , Matformer <sup>105</sup>
	Transformers	OPT <sup>106</sup> , GPT <sup>15</sup> , T5 <sup>107</sup>
FF	LJ <sup>108</sup>	LAMMPS <sup>109</sup> (2D-Liquid)
	EAM <sup>110</sup>	LAMMPS <sup>109</sup> (FCC-Al)
	REBO <sup>111</sup>	LAMMPS <sup>109</sup> (Diamond-Si)
	AMBER99sb-ildn <sup>112</sup>	GROMACS <sup>113</sup> (Alanine dipeptide)
	CHARMM36m <sup>114</sup>	GROMACS <sup>113</sup> ( $\alpha$ -aminoisobutyric acid)
QC	Algorithms	Qiskit <sup>115</sup> (VQE <sup>116</sup> , VQD <sup>117</sup> ) PennyLane <sup>118,119</sup> (VQE <sup>116</sup> , VQD <sup>117</sup> )
	Circuits	Qiskit <sup>115</sup> (PauliTwo Design <sup>115</sup> , SU(2) <sup>115</sup> )
EXP	Diffraction	XRD (Bruker D8)
	Manometry	CO <sub>2</sub> adsorption FACT lab <sup>120</sup>
	Vibroscopy	Kevlar FAVIMAT <sup>121</sup>
	Magnetometry	Susceptibility (PPMS) <sup>121</sup>

- 1) Electronic Structure
- 2) Artificial Intelligence
- 3) Force Field
- 4) Quantum Computation
- 5) Experiment

## Types of Data:

- **Atomic structure** (Molecule, Crystal)
- **Material Property** (Bandgap, bulk modulus)
- **Images** (Microscopy: SEM, TEM, STM)
- **Spectra** (Diffraction: X-ray, Neutron, PL)
- **Text** (Research articles, notebooks, blogs)
- **Eigensolver** (Quantum Computation algorithms)



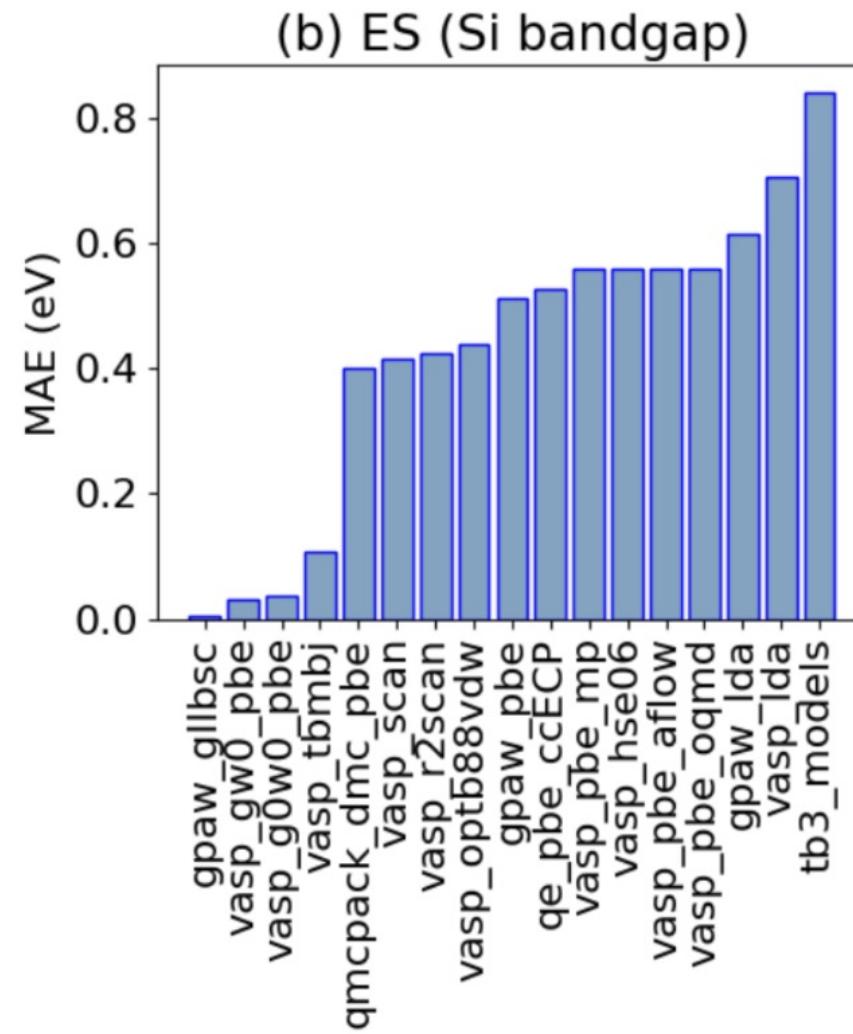
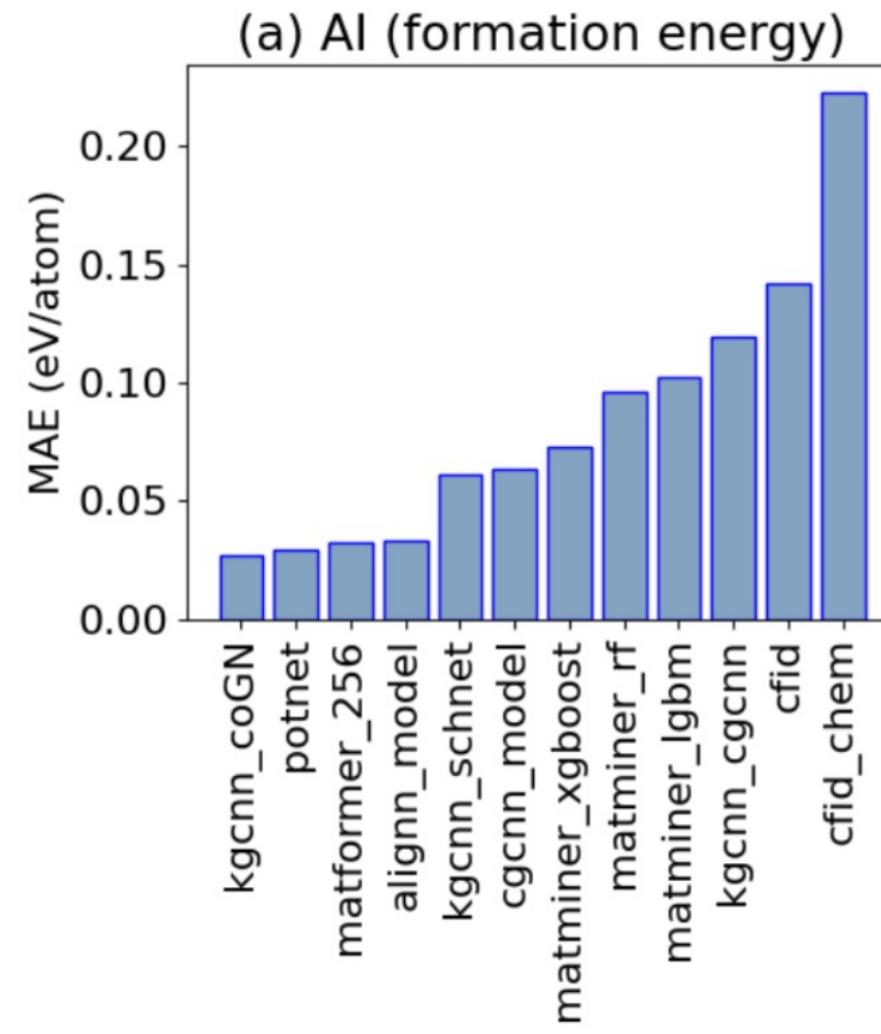
# Model for bulk\_modulus\_JVASP\_890\_Ge

## Model benchmarks

Model name	Dataset	MAE	Team name	Dataset size	Date submitted	Notes
qmcpack_dmc_pbe	dft_3d	2.1838	JARVIS	1	04-05-2023	CSV, JSON, run.sh, Info, JVASP-890
qe_pbe_ccECP	dft_3d	16.7	JARVIS	1	04-05-2023	CSV, JSON, run.sh, Info, JVASP-890
vasp_pbe_mp	dft_3d	16.8	MP	1	01-14-2023	CSV, JSON, run.sh, Info, JVASP-890
vasp_optpbe	dft_3d	19.4	JARVIS	1	01-14-2023	CSV, JSON, run.sh, Info, JVASP-890
vasp_optcx13	dft_3d	9.4	JARVIS	1	01-14-2023	CSV, JSON, run.sh, Info, JVASP-890
vasp_optb88vdw	dft_3d	17.73	JARVIS	1	01-14-2023	CSV, JSON, run.sh, Info, JVASP-890
vasp_opt86b	dft_3d	12.7	JARVIS	1	01-14-2023	CSV, JSON, run.sh, Info, JVASP-890

- Why do our methods **succeed or fail?**
- There is **no avenue** to report **if/where things went wrong** (details not reported in papers)
- **Reproducibility** and **Transparency** are **essential**
- JARVIS-Leaderboard **requires metadata** and **run.sh** file for each **contribution**
- **run.sh:** A script with details to **reproduce** the contributed data
- **metadata:**
  - Team name, URL, DOI, software/hardware, software version, computational timing, uncertainty, specific calculation details (i.e. choice of pseudopotential, hyperparameters, etc.)

# JARVIS-Leaderboard: Visualizing the Benchmarks



- Contributions/benchmarks can be **visualized** using **Google Colab** notebooks
- Assess accuracy of different methods
- **Validate** methods or **identify areas** where existing methods are not sufficient (motivation to **develop newer/more accurate models**)
- **Identify materials/material classes** which require **high fidelity methods**

# Conclusions and Outlook

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pubs.acs.org/JPCL

Letter

## A Quantum Monte Carlo Study of the Structural, Energetic, and Magnetic Properties of Two-Dimensional H and T Phase VSe<sub>2</sub>

Daniel Wines, Juha Tiihonen, Kayahan Saritas, Jaron T. Krogel, and Can Ataca\*

THE JOURNAL OF  
PHYSICAL  
CHEMISTRY C  
A JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

pubs.acs.org/JPCC

Article

## Systematic DFT+U and Quantum Monte Carlo Benchmark of Magnetic Two-Dimensional (2D) CrX<sub>3</sub> (X = I, Br, Cl, F)

Daniel Wines,\* Kamal Choudhary, and Francesca Tavazza

THE JOURNAL OF  
PHYSICAL  
CHEMISTRY C  
A JOURNAL OF THE AMERICAN CHEMICAL SOCIETY

pubs.acs.org/JPCC

Article

## Intrinsic Ferromagnetism of Two-Dimensional (2D) MnO<sub>2</sub> Revisited: A Many-Body Quantum Monte Carlo and DFT+U Study

Daniel Wines, Kayahan Saritas, and Can Ataca\*

Cite This: *J. Phys. Chem. C* 2022, 126, 5813–5821

Read Online

RESEARCH ARTICLE | NOVEMBER 18 2021

## A pathway toward high-throughput quantum Monte Carlo simulations for alloys: A case study of two-dimensional (2D) GaS<sub>x</sub>Se<sub>1-x</sub>

Daniel Wines\*; Kayahan Saritas; Can Ataca

Check for updates

*J. Chem. Phys.* 155, 194112 (2021)

<https://doi.org/10.1063/5.0070423>

CHORUS

- 2D materials often times have a complicated electronic structure and require higher accuracy methods
- DMC can be used to accurately predict 2D material properties
- DMC can be combined with DFT/+U to extract useful measurable quantities
- There is a community need to incorporate high accuracy methods into the traditional materials workflows/databases

RESEARCH ARTICLE | OCTOBER 19 2020

## A first-principles Quantum Monte Carlo study of two-dimensional (2D) GaSe

Special Collection: *Frontiers of Stochastic Electronic Structure Calculations*

Daniel Wines\*; Kayahan Saritas; Can Ataca

Check for updates

*J. Chem. Phys.* 153, 154704 (2020)

<https://doi.org/10.1063/5.0023223>

**JARVIS** Beyond DFT

Applied Physics Reviews

REVIEW

[pubs.aip.org/aip/are](https://pubs.aip.org/aip/are)

arXiv > cond-mat > arXiv:2306.11688

Condensed Matter > Materials Science

[Submitted on 20 Jun 2023]

Large Scale Benchmark of Materials Design Methods

Kamal Choudhary, Daniel Wines, Kangming Li, Kevin F. Garrity, Vishu Gupta, Aldo H. Romero, Jaron T. Krogel, Kayahan Saritas, Addis Fuhr, Panchapakesan Ganesh, Paul R. C. Kent, Keqiang Yan, Yuchao Lin, Shuiwang Ji, Ben Blaiszik, Patrick Reiser, Pascal Friederich, Ankit Agrawal, Pratyush Tiwary, Eric Beyerle, Peter Minch, Trevor David Rhone, Ichiro Takeuchi, Robert B. Wexler, Arun Mannodi-Kanakkithodi, Elif Ertekin, Avanish Mishra, Nithin Mathew, Sterling G. Baird, Mitchell Wood, Andrew Dale Rohskopf, Jason Hatrick-Simpers, Shih-Han Wang, Luke E. K. Achenie, Hongliang Xin, Maureen Williams, Adam J. Biacchi, Francesca Tavazza

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## Recent progress in the JARVIS infrastructure for next-generation data-driven materials design

Cite as: *Appl. Phys. Rev.* **10**, 041302 (2023); doi:10.1063/5.0159299

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Daniel Wines,<sup>1</sup> Ramya Gurunathan,<sup>1</sup> Kevin F. Garrity,<sup>1</sup> Brian DeCost,<sup>1</sup> Adam J. Biacchi,<sup>2</sup> Francesca Tavazza,<sup>1</sup> and Kamal Choudhary<sup>1(a)</sup>



# Openings in Ataca Group: Students and Postdocs

NIST

## Openings

We are always looking for hardworking candidates with strong communication skills and creativity, who are capable of working both independently and in collaborative environments.

Post-doctoral candidates should send their CVs to [Professor Ataca](#). Prospective graduate students should apply to the PhD Program in Physics at UMBC before they can start working in our group; however, they are encouraged to contact [Prof. Ataca](#) beforehand to discuss opportunities & possibilities. Interested undergraduate students should directly contact [Prof. Ataca](#).



Bachelor

Master

PhD

Postdoc

PI



[ataca@umbc.edu](mailto:ataca@umbc.edu)



# Quantum Matters in Materials Science Workshop

NIST



- Workshop based on emerging quantum materials, hosted by NIST
- Experiment and theory
- Academia, government and industry (28 invited speakers)
- Poster session: if interested email abstract to [daniel.wines@nist.gov](mailto:daniel.wines@nist.gov)
- \$100 registration fee, \$60 for students (includes lunch/coffee for both days)

## Registration

<https://www.nist.gov/news-events/events/2024/02/quantum-matters-materials-science-workshop>

# Postdoctoral Opportunities with JARVIS

NRC Postdoc Opportunities:  
Many project opportunities for recent PhDs interested in quantum materials, machine learning, computation, and materials design.

- Open to U.S. citizens
- Proposal deadlines: Feb. 1st and Aug. 1st

## CHIPS Positions (~5)

- Multiscale modeling of semiconductor defects and interfaces
- Electronic structure/DFT
- Tight Binding
- Machine learning, ML-force field development
- Transport properties (non-equilibrium Green's function)
- Device modeling (i.e. TCAD)

