



# Benchmark study of symmetry-adapted ML-DFT models for magnetically doped topological insulators

September 14, 2023 | **Johannes Wasmer<sup>1</sup>** Rubel Mozumder<sup>2,1</sup> Philipp Rüßmann<sup>3,1</sup> Stefan Blügel<sup>1</sup> | <sup>1</sup>Forschungszentrum Jülich | <sup>2</sup>Humboldt University Berlin | <sup>3</sup>University of Würzburg

# Outline

Introduction

Atomistic machine learning

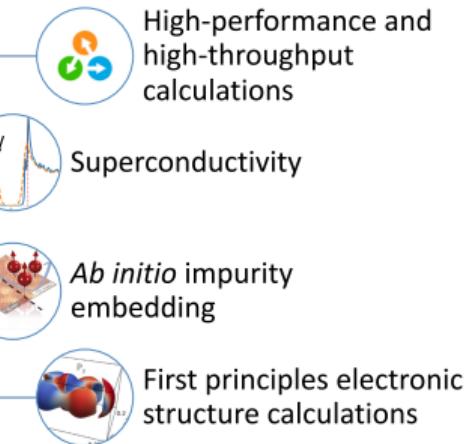
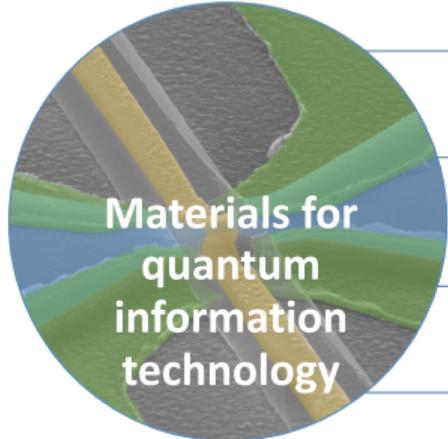
My PhD project

Talk held at WE-Heraeus Workshop 2023, “First-principles Green function formalisms”,  
September 4-7 2023, in Athens, Greece ([go.fzj.de/gf2023](http://go.fzj.de/gf2023)).

Latest version of slides are [here](#).

# Affiliations

## My research group



**juDFT**



[judft.de](http://judft.de)

## My graduate school

**HDSLEE**

**HELMHOLTZ**  
School for Data Science  
in Life · Earth · Energy

Exchange between data science students across domains

# A high-level perspective

Learning simulation systems<sup>1</sup>

1st Paradigm



Observation

2nd Paradigm

$$H\Psi = E\Psi$$

Theory

3rd Paradigm



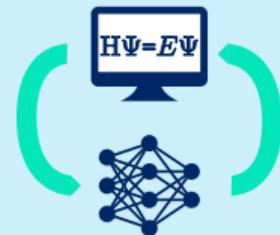
Simulation

4th Paradigm



Data Science

5th Paradigm

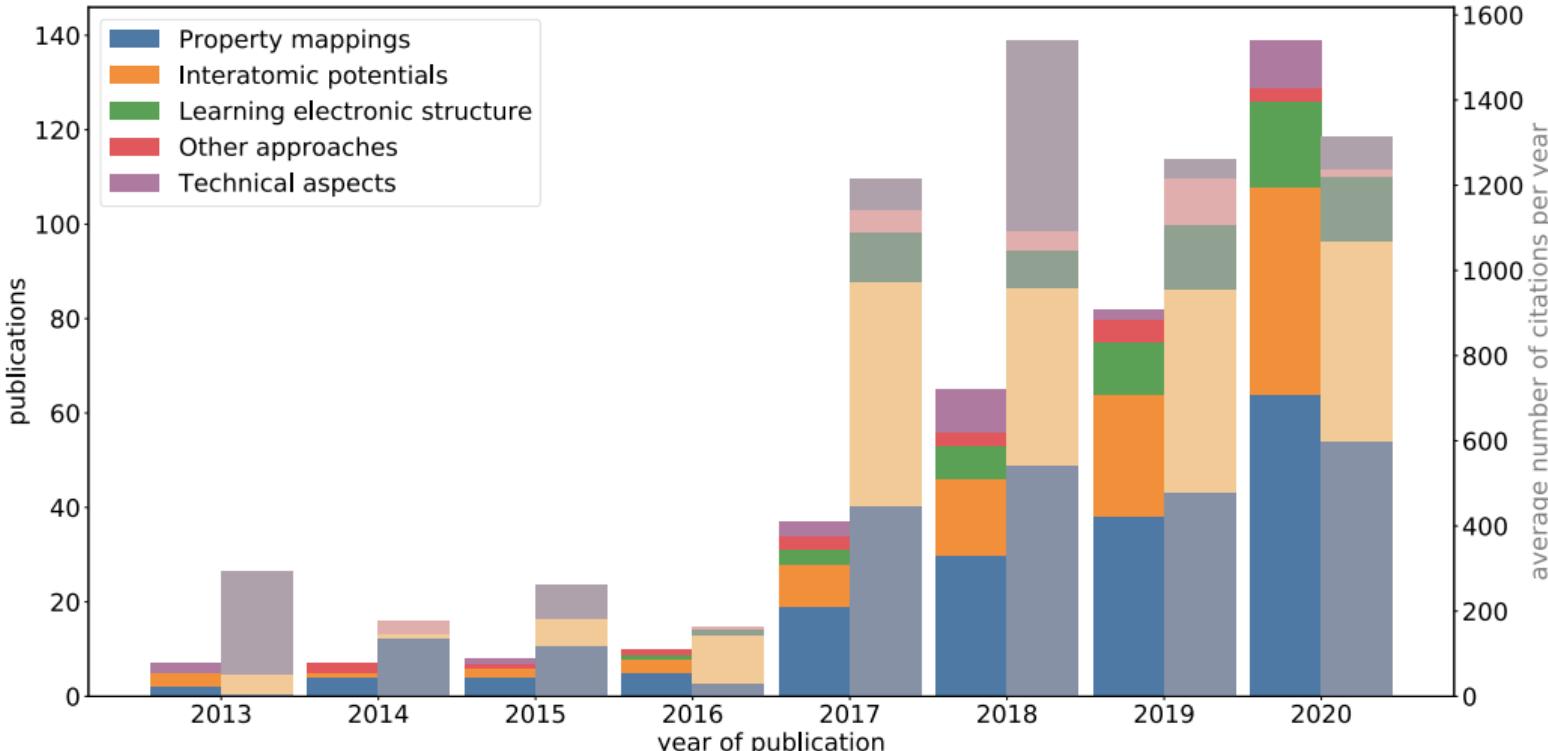


Emulation

<sup>1</sup>Bishop, Welling, and Llorens 2022.

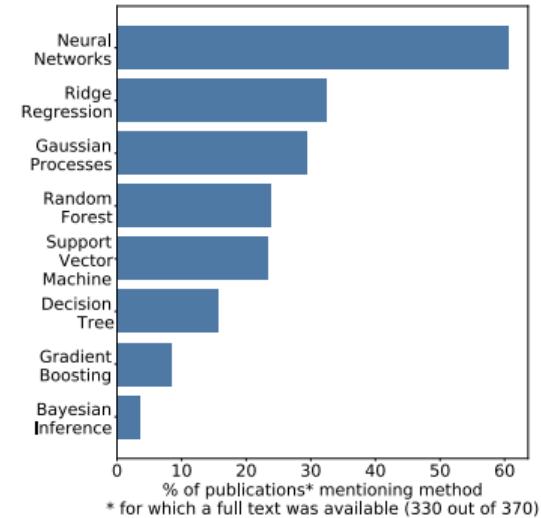
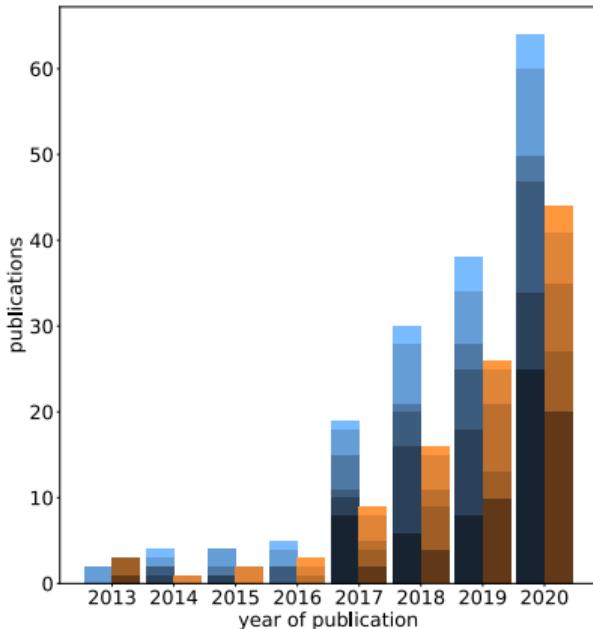
# Literature analysis

## Atomistic machine learning



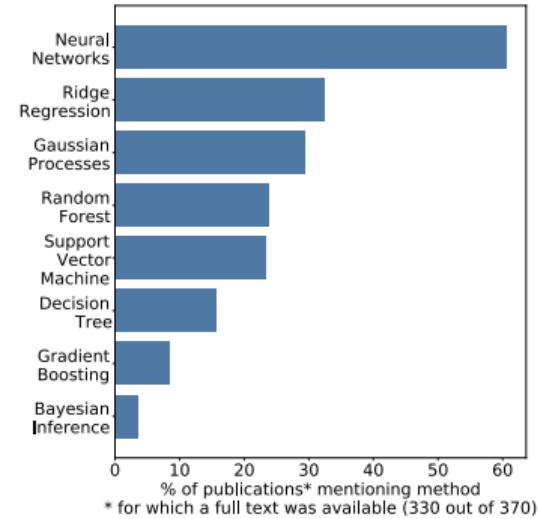
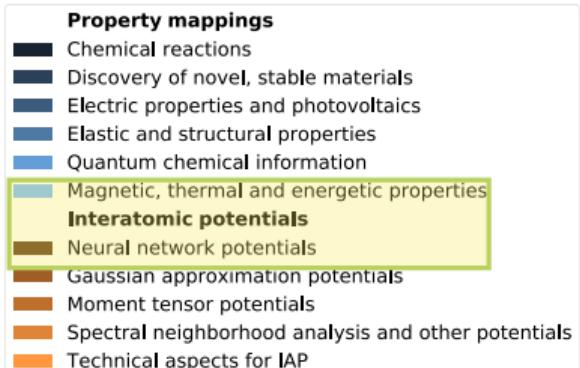
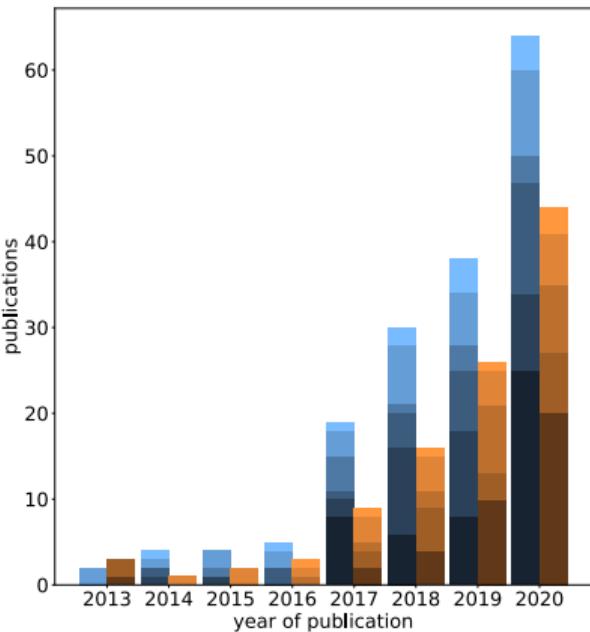
# Literature analysis

## Atomistic machine learning



# Literature analysis

## Atomistic machine learning



# Success stories

## Molecular dynamics

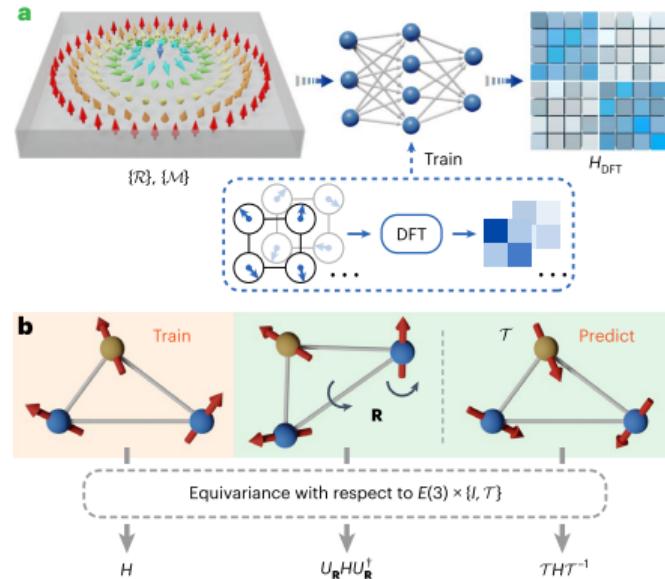
- Linear-scaling simulation of full HIV capsid (44M atoms) at DFT accuracy over nanoseconds on 5'000 A100 GPUs<sup>a</sup>
- Trained on 1M structures  $\leq 100$  atoms, single GPU, 7 days
- Gordon Bell Prize 2023 finalist



<sup>a</sup>Musaelian et al. 2023.

## Electronic structure

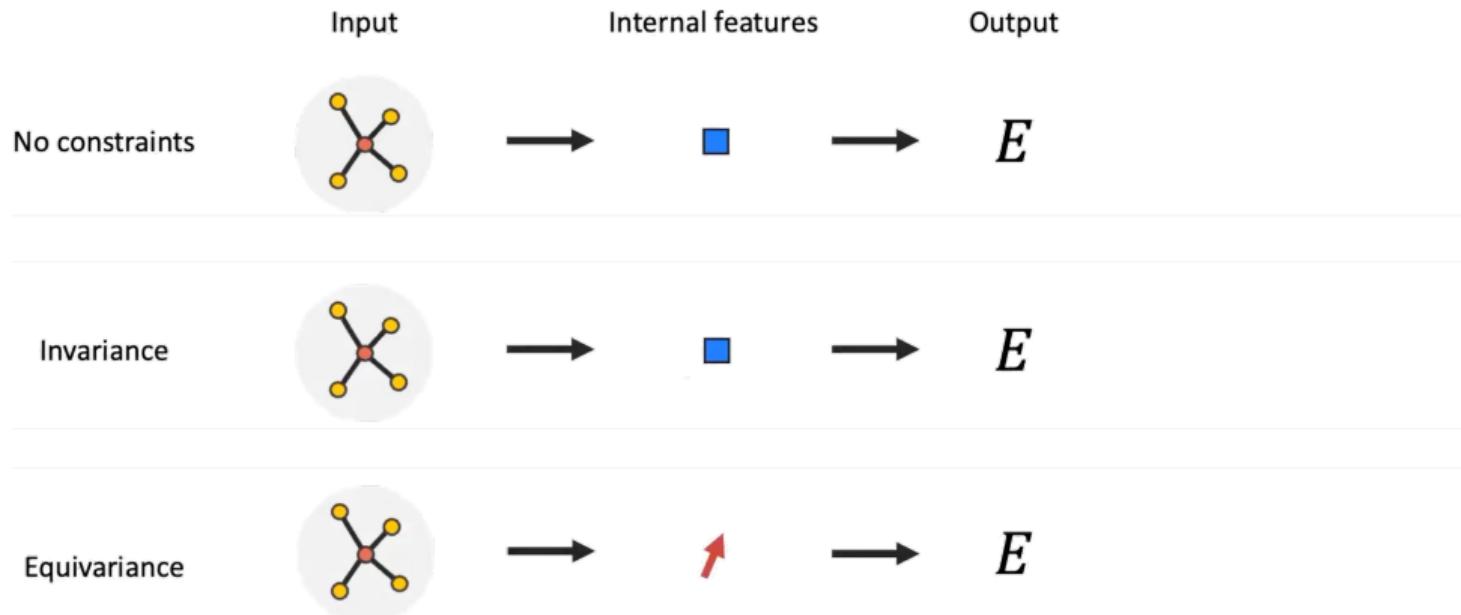
- Prediction of Hamiltonian for non-collinear DFT<sup>a</sup>



<sup>a</sup>Li et al. 2023.

# Inductive bias

Example:  $O(3)$  symmetry

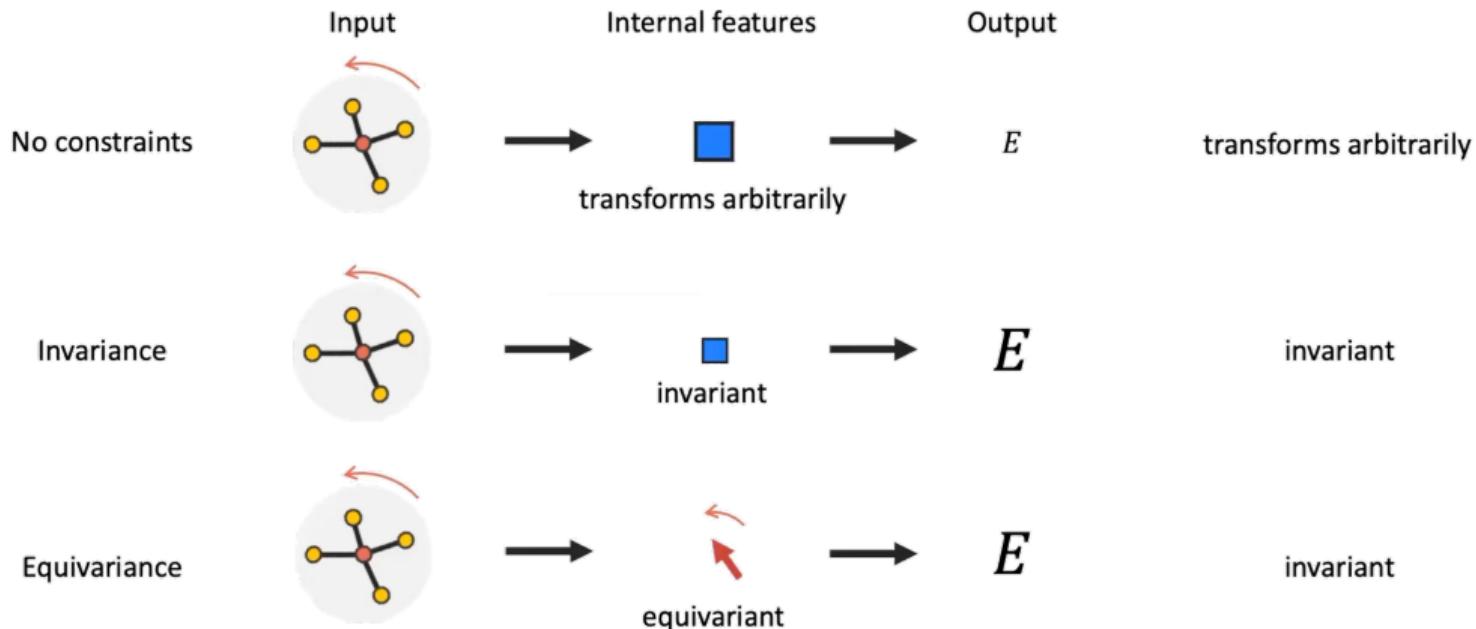


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<sup>1</sup> Adapted from (Musaelian 2023)

# Inductive bias

## Example: $O(3)$ symmetry



1

<sup>1</sup> Adapted from (Musaelian 2023)

# Feature engineering

Example: O(3)-invariant SOAP

## Descriptor: 3-body

Smooth overlap of atomic positions (SOAP)<sup>a</sup>

Feature vector  $\phi(A) = \mathbf{p} \in \mathbb{R}^n$

## Model: Kernel regression

Kernel  $\kappa(A, A') = \langle \phi(A), \phi(A') \rangle_{\mathcal{F}} = \langle \mathbf{p}, \mathbf{p}' \rangle_{\mathcal{F}} \in \mathbb{R}$

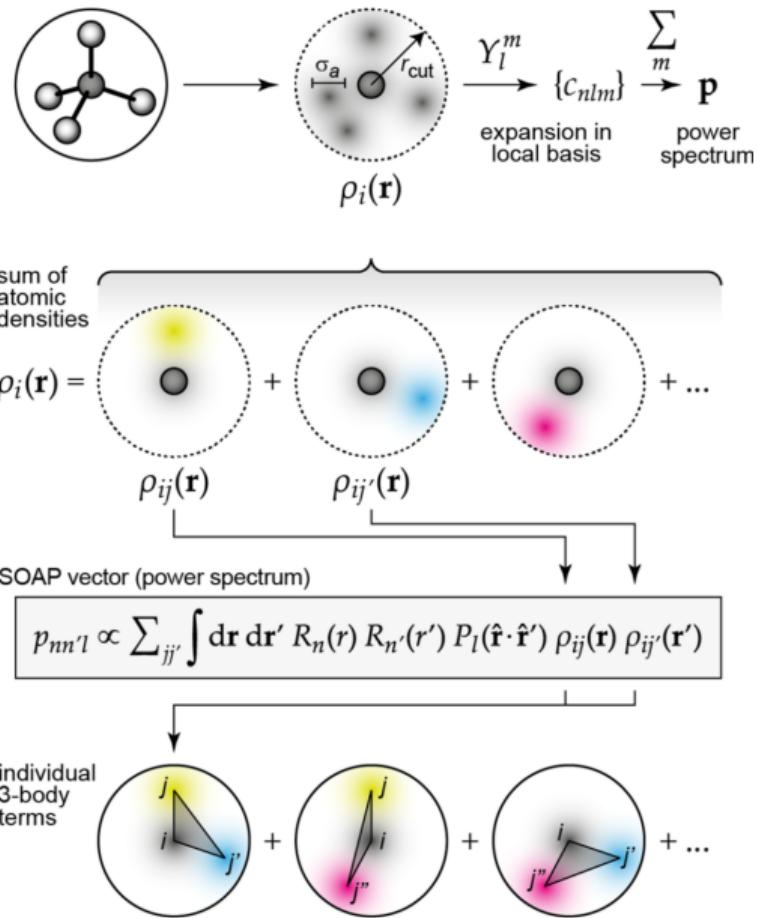
Gram matrix  $\mathbf{K} = \left( \kappa(A^{(i)}, A^{(j)}) \right)_{i,j} \in \mathbb{R}^{m \times m}$

Cost function  $\mathcal{C}(\mathbf{y}, \hat{\mathbf{y}}) = \|\mathbf{y} - \hat{\mathbf{y}}\|^2 + \alpha \mathbf{c}^\top \mathbf{K} \mathbf{c}$

Training  $\mathbf{c} = (\mathbf{K} + \alpha \mathbf{I}_m)^{-1} \mathbf{y}$

Inference  $\hat{y}(A) = \sum_i^m c_i \kappa(A, A^{(i)})$

<sup>a</sup>Adapted from (Deringer et al. 2021)



# Feature learning

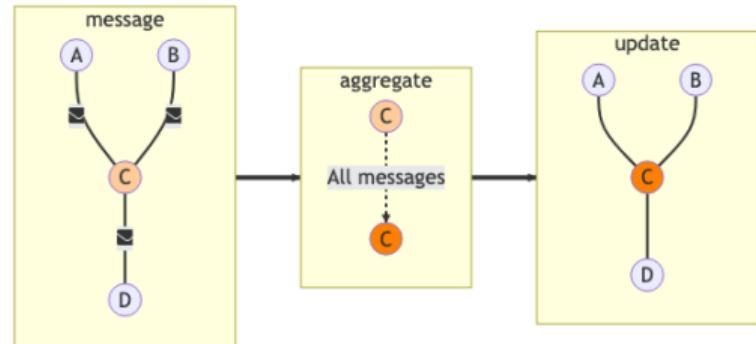
Example: O(3)-equivariant MPNN

## Model: Graph neural network

### Message-passing formalism (MPNN)

1. Message  $\mathbf{m}_{ij}^{(t)} = \mathcal{M} \left( \mathbf{h}_i^{(t)}, \mathbf{h}_j^{(t)}, \mathbf{e}_{ij} \right)$
2. Aggregation  $\hat{\mathbf{m}}_i = \bigoplus_{j \in \mathcal{N}_i} \mathbf{m}_{ij}$
3. Update  $\mathbf{h}_i^{(t+1)} = \phi \left( \mathbf{h}_i^{(t)}, \hat{\mathbf{m}}_i^{(t)} \right)$

Where  $\mathbf{h}_i^{(t)}$  node features at step  $t$ ,  $\mathbf{e}_{ij}$  edge features,  $\mathcal{N}_i$  neighborhood of node  $i$ ,  $\phi$  update function.



## Equivariance

Internal features  $\mathbf{h}_i$  transform under rotations  $Q \in \text{O}(3)$ .

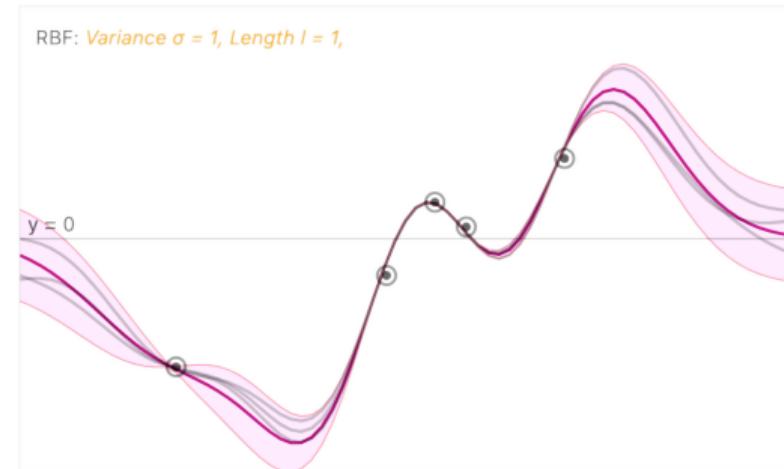
$$h_{i,klm}^{(t)}(Q \cdot \mathbf{R}) = \sum_{m'} D_{mm'}^l(Q) h_{i,klm'}^{(t)}(\mathbf{R})$$

Where  $k$  feature index,  $\mathbf{R}$  atom positions,  $D^l$  Wigner D-matrix,  $l = 0$  invariant scalars.

# Active learning

Materials space **too large** for true generalizability → Couple data generation & model training

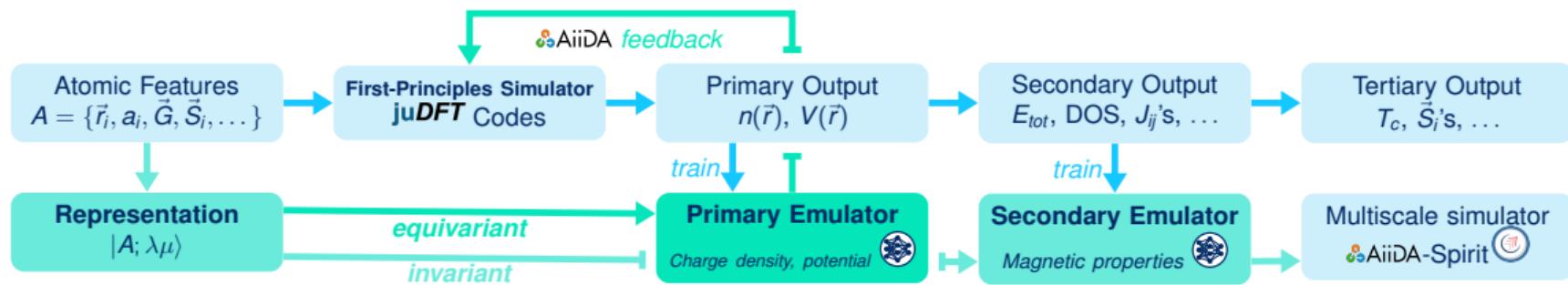
- AL improves data efficiency drastically<sup>a</sup>
- Offline AL: Test on simulations, manual.  
Online AL: Model has built-in uncertainty prediction → automatable retraining.
- Uncertainty methods: Ensemble learning (indirect), Bayesian (direct, e.g. Gaussian process)
- Sampling criteria: uncertainty (min. error), dissimilarity measure (max. diversity), ...



<sup>a</sup>Jinnouchi et al. 2019.

# My PhD project

[go.fzj.de/wasmer](http://go.fzj.de/wasmer)



Electronic structure emulator  
for fast SCF convergence

Magnetic property prediction  
for spin dynamics simulation

# Magnetic property prediction

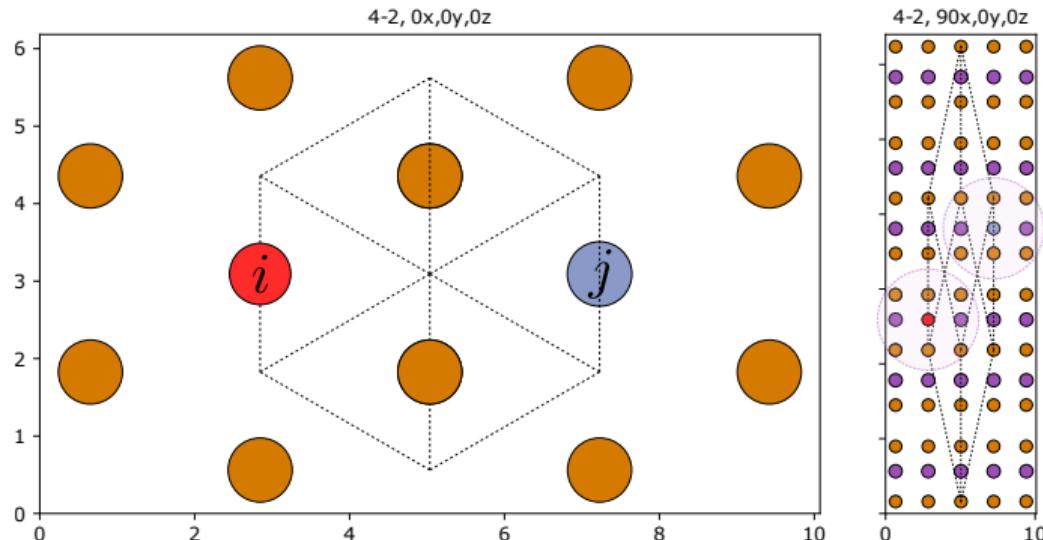
Example: Transition metal dimer impurities, embedded into  $\text{Bi}_2\text{Te}_3$ .

Magnetic interactions, classical model:

$$\mathcal{H} \approx - \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j - \sum_{\langle ij \rangle} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

Coupling constants from the KKR Green function:

$$\mathcal{J}_{ij} = -\frac{1}{\pi} \text{Im} \int_{-\infty}^{E_F} dE \text{Tr}[\delta t_i G_{ij} \delta t_j G_{ji}]$$



# Magnetic property prediction

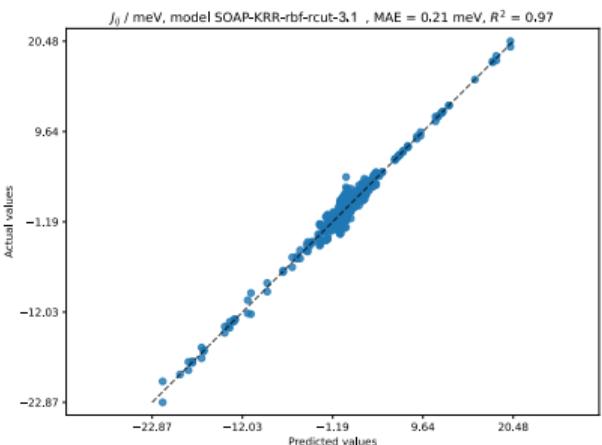
Data 2'000 dimer embeddings

Model SOAP+KRR

Performance MAE = 0.21 meV,  $R^2 = 0.97$

Next steps

- Compare with MPNN models
- Offline AL: Test model predictions in spin dynamics simulations



	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	Sc
Sc	0	0.012	0.056	0.039	0.083	0.14	0.085	0.051	0	0	0	0	0.1	0.25	0.18	0	0	0	0	0	Ti
Ti	-0.012	6.3	6.7	11	7.9	3	4.1	1.9	0.064	0	0	0.1	12	6.9	3.2	-1	0.83	0.2	0.025	0	V
V	-0.055	6.6	12	18	9.1	0.65	1.3	-4.6	0.29	0.053	0.021	0.17	11	13	9.1	0.44	1	0.16	0.099	0.013	Cr
Cr	-0.04	11	19	12	4.8	-6.6	-12	-7	0.061	0.048	0.021	0.15	20	18	0.079	-0.24	0.26	0.36	0.017	0.022	Mn
Mn	-0.082	7.7	8.8	4.8	-2.4	-13	-12	-5.4	0.036	0.036	0.034	0.25	7.2	-2	-13	-3.6	0.092	0.17	0.013	0.023	Fe
Fe	-0.11	2.6	-0.58	-5.5	-13	-20	-17	-8.5	0.1	0.051	0.06	0.16	-4.2	-14	-16	-1.9	0.25	0.55	0.063	0.042	Co
Co	-0.063	3.4	-0.34	-9.7	-11	-17	-14	-3.2	0.12	0.013	0.028	0.18	-0.61	-13	-20	-3.7	0.27	0.085	0.054	0.012	Ni
Ni	-0.05	1.4	-5.6	-7.2	-5.6	-9.6	-2.7	1.7	0.015	0	0.019	0.13	-4.7	-19	-18	-2.8	0.082	0.042	0	0	Cu
Cu	0	0.062	0.28	0.055	0.035	0.1	0.11	0.012	0	0	0	0.15	0.24	0.2	0.08	0	0	0	0	0	Zn
Zn	0	0	0.056	0.048	0.036	0.077	0.021	0	0	0	0	0.07	0.27	0.2	0	0	0	0	0	0	Y
Y	0	0	0.021	0.021	0.034	0.074	0.038	0.019	0	0	0	0	0.029	0.12	0.11	0	0	0	0	0	Zr
Zr	0	0.11	0.18	0.14	0.26	0.2	0.22	0.14	0	0	0	0.63	0.37	0.13	0	0	0	0	0	0	Nb
Nb	0.1	12	11	20	7.4	-3.9	0.024	-3.9	0.16	0.066	0.028	0.61	16	6.2	3	1.7	2.8	0.15	0.049	0.013	Mo
Mo	-0.25	6.8	13	18	-1.8	-15	-14	-18	0.26	0.26	0.12	0.37	6.4	7.8	4.3	1.5	0.76	1.9	0.068	0.12	Tc
Tc	-0.18	3.8	9.6	1.1	-13	-18	-23	-18	0.2	0.2	0.11	0.13	4.3	4.5	-0.72	1.2	1.1	2.3	0.12	0.14	Ru
Ru	0	-0.8	0.58	-1.1	-4.8	-1.3	-3.6	-2.8	0.076	0.01	0	0	1.6	1.7	1.5	0.87	0.048	0.017	0.038	0.011	Rh
Rh	0	1	1.1	0.24	0.099	0.23	0.23	0.065	0	0	0	3.2	0.75	1.4	0.051	0	0	0	0	0	Pd
Pd	0	0.16	0.24	0.44	0.22	0.89	0.14	0.049	0	0	0	0.18	2.4	2.3	0.015	0	0	0	0	0	Ag
Ag	0	0.024	0.093	0.015	0.013	0.063	0.052	0	0	0	0	0.046	0.064	0.12	0.039	0	0	0	0	0	Cd
Cd	0	0	0.014	0.022	0.023	0.041	0.019	0	0	0	0	0.014	0.13	0.14	0.011	0	0	0	0	0	Sc

$J_{jj}$  / meV

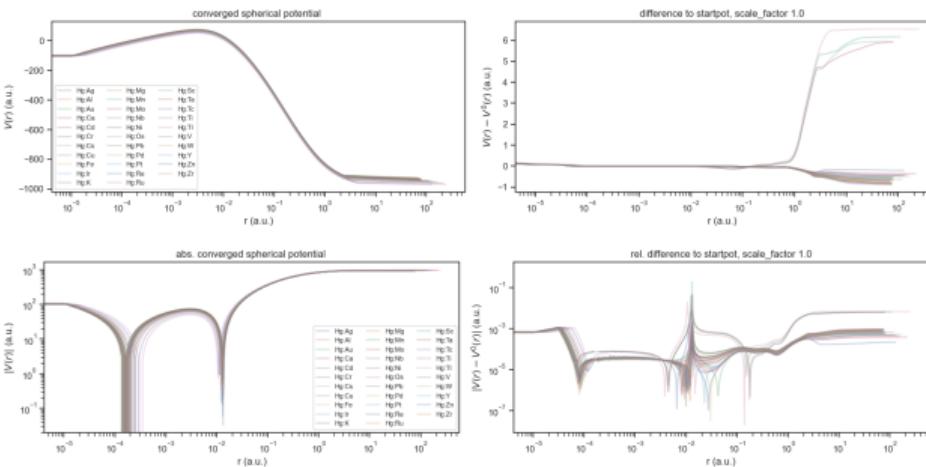
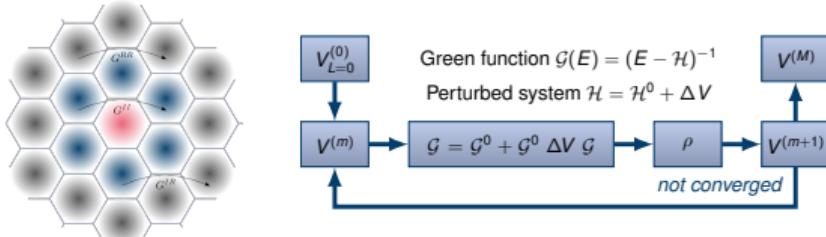
# Electronic structure emulator

**Task** Predict electron potential difference  $\Delta V_{imp}(\vec{r})$  & use as initial guess for AiiDA-KKR

**Data** 10'000 single impurity in elemental crystal combinations ( $60 \times 40$  elements) from DFT

## Challenges

- **Collaboration** Adapt the linear Jacobi-Legendre Charge Density Model (JLCDM)<sup>a</sup> to all-electron DFT
- Large chemical feature space
- From radial to full equivariant potential
- Intermediate SCF results
- Should be code-agnostic
- From collinear to no-co magnetism



<sup>a</sup>Focassio et al. 2023.

Spherical impurity potentials in first Voronoi cell of Hg:X embeddings,

left upper to bottom right:  $V$ ,  $V - V^0$ ,  $|V|$ ,  $|V - V^0|$ .

# Tools & Infrastructure

JuDFTteam/best-of-atomistic-machine-...

juDFT

🏆 A ranked list of awesome atomistic machine learning projects 🎮💎.

3

Contributors

12

Issues

70

Stars

4

Forks



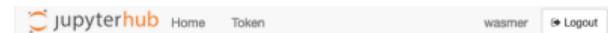
github.com

GitHub - JuDFTteam/best-of-atomistic-machine-learning: 🏆 A ranked list of awesome atomistic...

🏆 A ranked list of awesome atomistic machine learn...

↗ Tools • [go.fzj.de/best-of-aml](https://go.fzj.de/best-of-aml) • Largest list of atomistic ML tools on the web (300+), regular updates

Infrastructure • iffAiiDA • High-throughput & machine learning without the setup cost, portable ➡



## Server Options

Select an image:

alida-v2:latest

Use a previous image version

Select a previous image version:

alida-common-workflows:2022.07.27

Start

## iffAiiDA User Documentation

iffAiiDA lets you use the JuDFT team's [plugins](#) for the AiiDA workflow engine in your browser. No setup required! It also comes with pre-installed machine learning frameworks. Each user gets 64 CPU cores.

Have a look at the [tutorial](#) to get started. If you have questions, please [contact us](#).

[Bookmark this page](#).

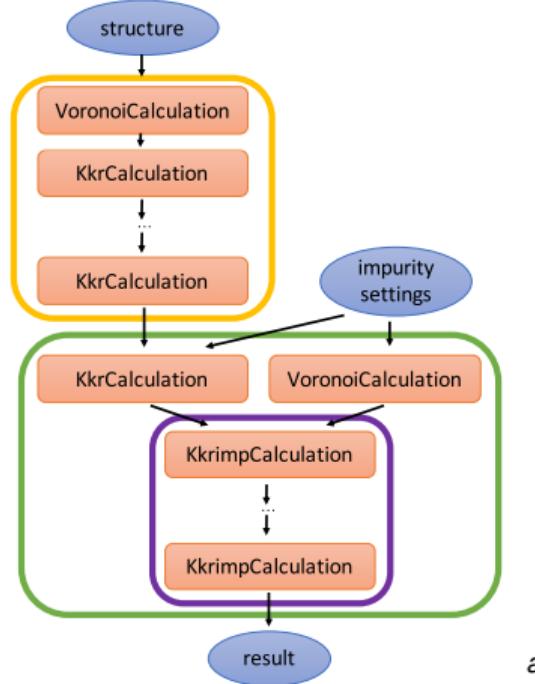


## Main Images

alida-v2 . Contains AiiDA in version 2. Contains the plugins alida-fleur, alida-kkr, alida-spirit, mascotools in their latest versions. Contains the iffdata command line tool for loading codes for the cluster iffSLURM.

alida-v1 . Contains AiiDA in version 1. Contains the plugin alida-fleur.

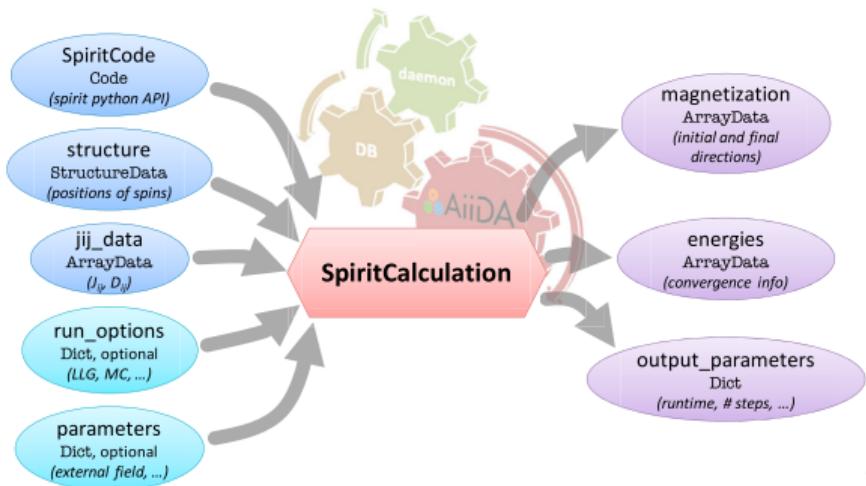
# Discussion slides



a

$$\mathcal{J}_{ij} = -\frac{1}{\pi} \operatorname{Im} \int_{-\infty}^{E_F} dE \operatorname{Tr}[\delta t_i G_{ij} \delta t_j G_{ji}]$$

<sup>a</sup>Rüßmann, Bertoldo, and Blügel 2021.



a

$$\mathcal{H} \approx - \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j - \sum_{\langle ij \rangle} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

$$\frac{\partial \mathbf{n}_i}{\partial t} = -\gamma' \mathbf{n}_i \times \mathbf{B}_i^{\text{eff}} - \lambda \mathbf{n}_i \times (\mathbf{n}_i \times \mathbf{B}_i^{\text{eff}})$$

<sup>a</sup>Rüßmann, Ribas Sobreviela, et al. 2022.

# References I

- Bishop, Christopher Michael, Max Welling, and Ashley Llorens (Oct. 17, 2022). Plenary: The Fifth Paradigm of Scientific Discovery. Microsoft Research Summit 2022. URL: <https://www.microsoft.com/en-us/research/video/plenary-the-fifth-paradigm-of-scientific-discovery/> (visited on 01/16/2023).
- Deringer, Volker L. et al. (Aug. 25, 2021). “Gaussian Process Regression for Materials and Molecules”. In: *Chemical Reviews* 121.16, pp. 10073–10141. ISSN: 0009-2665. DOI: [10.1021/acs.chemrev.1c00022](https://doi.org/10.1021/acs.chemrev.1c00022). URL: <https://doi.org/10.1021/acs.chemrev.1c00022> (visited on 06/03/2022).
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- Focassio, Bruno et al. (May 29, 2023). "Linear Jacobi-Legendre Expansion of the Charge Density for Machine Learning-Accelerated Electronic Structure Calculations". In: *npj Computational Materials* 9.1 (1), pp. 1–10. ISSN: 2057-3960. DOI: [10.1038/s41524-023-01053-0](https://doi.org/10.1038/s41524-023-01053-0). URL: <https://www.nature.com/articles/s41524-023-01053-0> (visited on 05/30/2023).
- Jinnouchi, Ryosuke et al. (June 7, 2019). "Phase Transitions of Hybrid Perovskites Simulated by Machine-Learning Force Fields Trained on the Fly with Bayesian Inference". In: *Physical Review Letters* 122.22, p. 225701. DOI: [10.1103/PhysRevLett.122.225701](https://doi.org/10.1103/PhysRevLett.122.225701). URL: <https://link.aps.org/doi/10.1103/PhysRevLett.122.225701> (visited on 09/05/2023).
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# References III

- Musaelian, Albert, director (June 13, 2023). Learning Local Equivariant Representations for Large-Scale Atomistic Dynamics. online. URL: <https://m2d2.io/talks/m2d2/learning-local-equivariant-representations-for-large-scale-atomistic-dynamics/> (visited on 09/05/2023).
- Musaelian, Albert et al. (Apr. 19, 2023). Scaling the Leading Accuracy of Deep Equivariant Models to Biomolecular Simulations of Realistic Size. DOI: [10.48550/arXiv.2304.10061](https://doi.org/10.48550/arXiv.2304.10061). arXiv: [2304.10061 \[physics, q-bio\]](https://arxiv.org/abs/2304.10061). URL: [http://arxiv.org/abs/2304.10061](https://arxiv.org/abs/2304.10061) (visited on 09/04/2023). preprint.
- Rüßmann, Philipp, Fabian Bertoldo, and Stefan Blügel (Jan. 26, 2021). “The AiiDA-KKR Plugin and Its Application to High-Throughput Impurity Embedding into a Topological Insulator”. In: *npj Computational Materials* 7.1 (1), pp. 1–9. ISSN: 2057-3960. DOI: [10.1038/s41524-020-00482-5](https://doi.org/10.1038/s41524-020-00482-5). URL: <https://www.nature.com/articles/s41524-020-00482-5> (visited on 05/13/2021).

# References IV

-  Rüßmann, Philipp, Jordi Ribas Sobreviela, et al. (2022). "The AiiDA-Spirit Plugin for Automated Spin-Dynamics Simulations and Multi-Scale Modeling Based on First-Principles Calculations". In: Frontiers in Materials 9. ISSN: 2296-8016. DOI: [10.3389/fmats.2022.825043](https://doi.org/10.3389/fmats.2022.825043). URL: <https://www.frontiersin.org/articles/10.3389/fmats.2022.825043> (visited on 08/11/2022).