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

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

Outline

Introduction

Atomistic machine learning

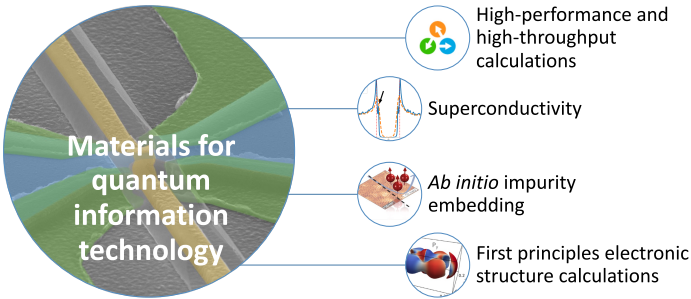
My PhD project

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

Latest version of slides are here.

Affiliations

My research group



juDFT

spirit

AiiDA

judft.de

My graduate school

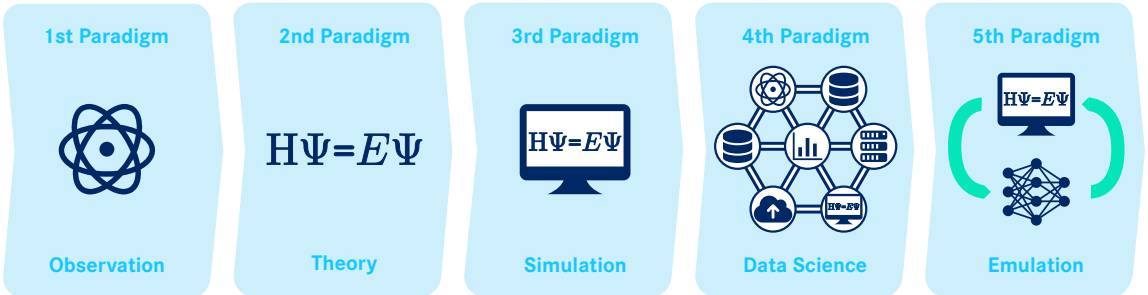
HDSLEE

HELMHOLTZ
School for Data Science
in Life · Earth · Energy

Exchange between data science students across domains

A high-level perspective

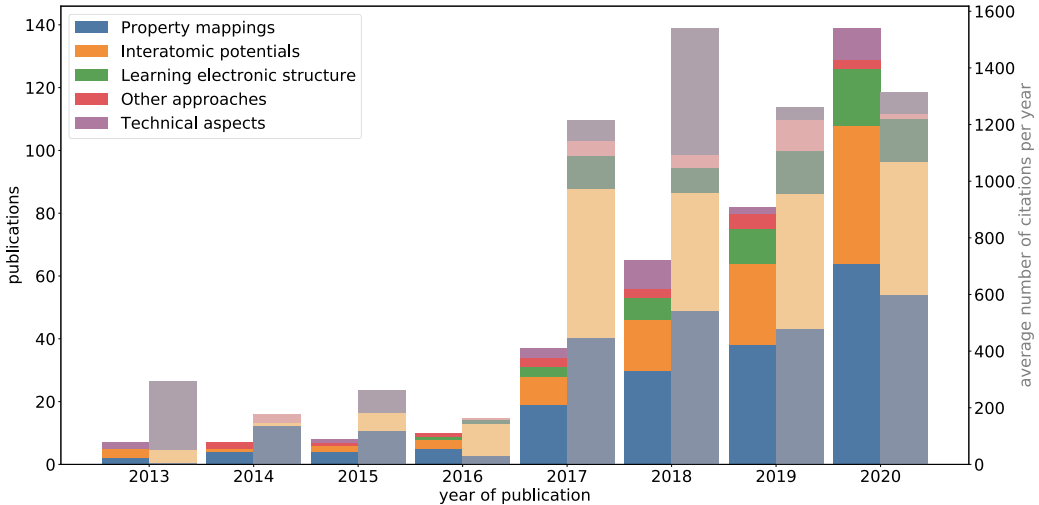
Learning simulation systems[1]



[1] Bishop, Welling, and Llorens 2022.

Literature analysis

Atomistic machine learning



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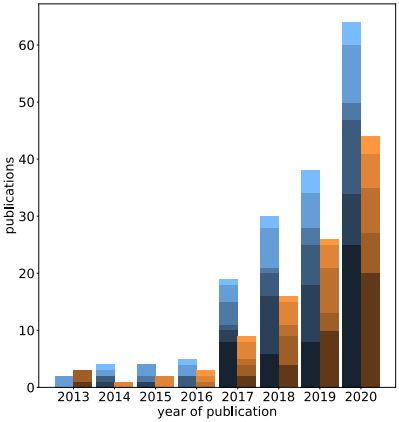
[1] 370 AML papers 2023-2020, figures from (Fiedler et al. 2022)

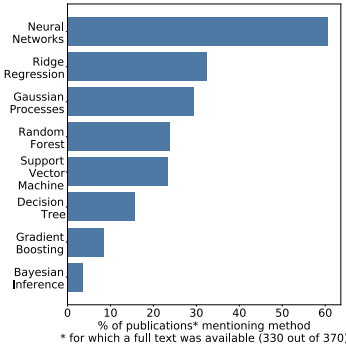
Literature analysis

Atomistic machine learning



Property mappings
- Chemical reactions
- Discovery of novel, stable materials
- Electric properties and photovoltaics
- Elastic and structural properties
- Quantum chemical information
- Magnetic, thermal and energetic properties

Interatomic potentials
- Neural network potentials
- Gaussian approximation potentials
- Moment tensor potentials
- Spectral neighborhood analysis and other potentials
- Technical aspects for IAP



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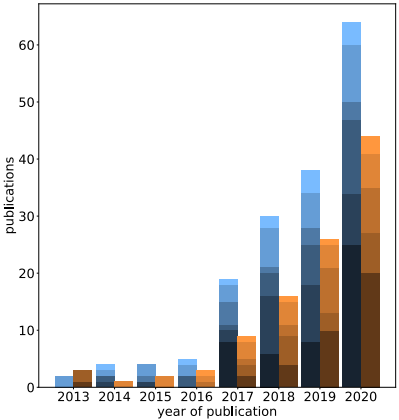
Literature analysis

Atomistic machine learning



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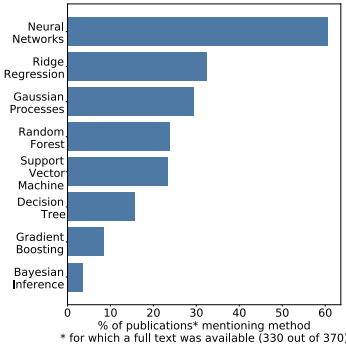

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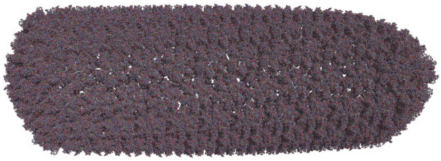
Success stories

Molecular dynamics

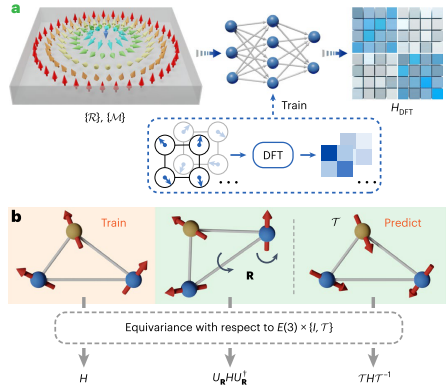
- Linear-scaling simulation of full HIV capsid (44M atoms) at DFT accuracy over nanoseconds on 5'000 A100 GPUs[a]
- Trained on 1M structures $\leq$ 100 atoms, single GPU, 7 days
- Gordon Bell Prize 2023 finalist



[a]Musaelian et al. 2023.

Electronic structure

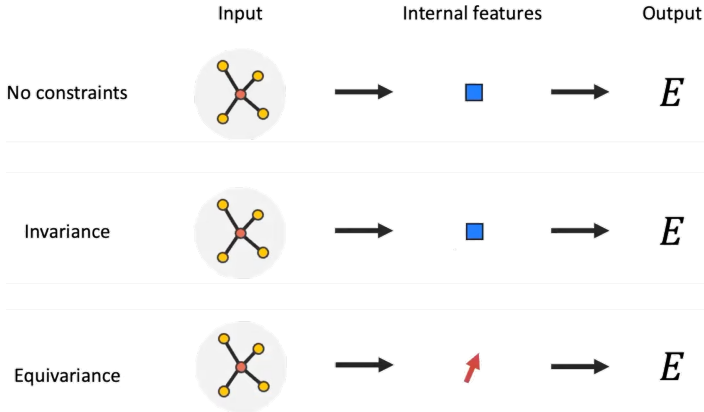
- Prediction of Hamiltonian for non-collinear DFT[a]



[a]Li et al. 2023.

Inductive bias

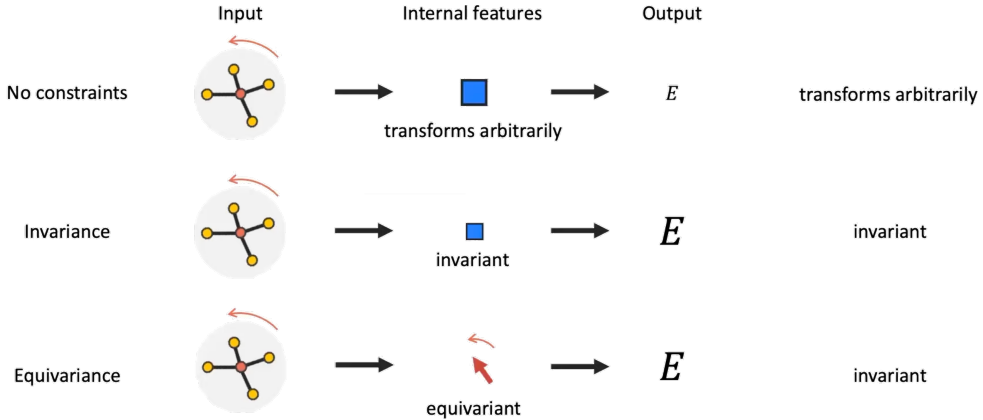
Example: O(3) symmetry



[1] Adapted from (Musaelian 2023)

Inductive bias

Example: O(3) symmetry



[1] Adapted from (Musaelian 2023)

Feature engineering

Example: O(3)-invariant SOAP

Descriptor: 3-body

Smooth overlap of atomic positions (SOAP)[a]

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

Model: Kernel regression

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

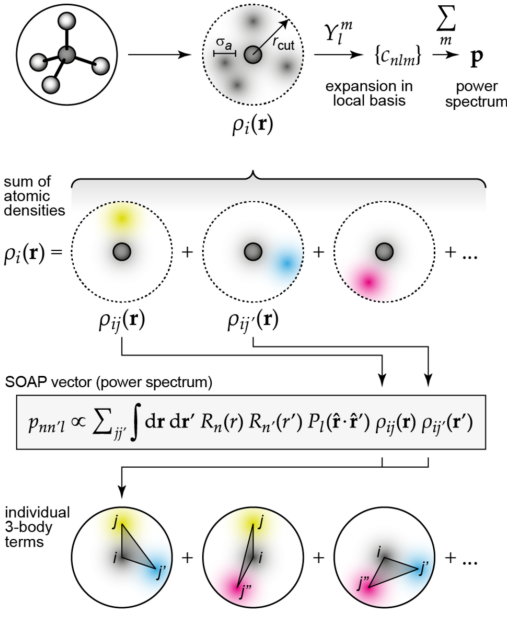
Gram matrix $\mathbf{K} = \left(\kappa(A^{(i)}, A^{(j)}) \right)_{i,j} \in \mathbb{R}^{m \times m}$

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

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

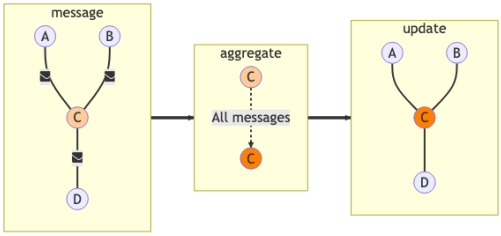
Inference $\hat{y}(A) = \sum_i^m c_i\, \kappa(A, A^{(i)})$

[a] 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, ϕ update function.

Equivariance

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

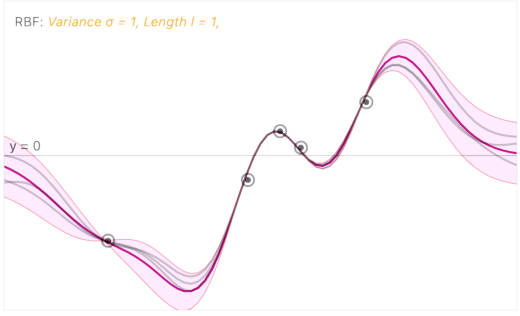
$$h_{i,klm}^{(t)}(Q \cdot \mathbf{R}) = \sum_{m'} D_{mm'}^{l}(Q) h_{i,klm'}^{(t)}(\mathbf{R})$$

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

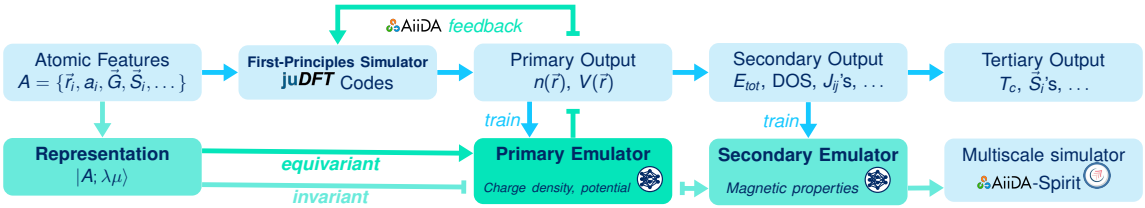
- AL improves data efficiency drastically[a]
- Offline AL: Test on simulations, manual. Online AL: Model has built-in uncertainty prediction ➔ automatable retraining.
- Uncertainty methods: Ensemble learning (indirect), Bayesian (direct, e.g. Gaussian process)
- Sampling criteria: uncertainty (min. error),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



[a] Jinnouchi et al. 2019.

My PhD project

go.fzj.de/wasmer



Electronic structure emulator
for fast SCF convergence

Magnetic property prediction
for spin dynamics simulation

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Magnetic property prediction

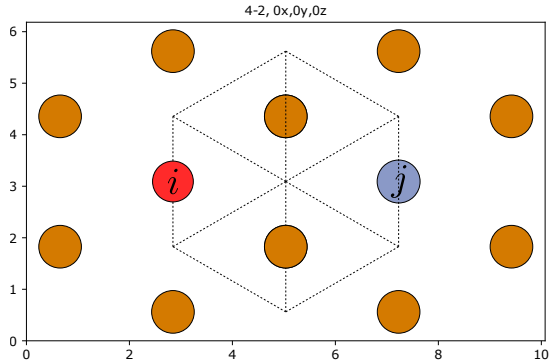
Example: Transition metal dimer impurities, embedded into Bi_2Te_3.

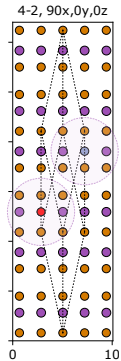
Magnetic interactions, classical model:

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

Coupling constants from the KKR Green function:

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





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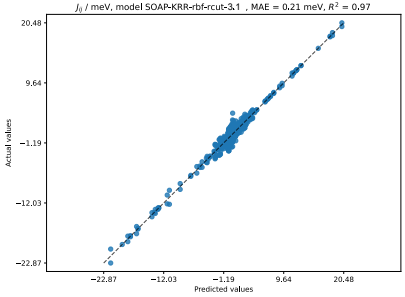
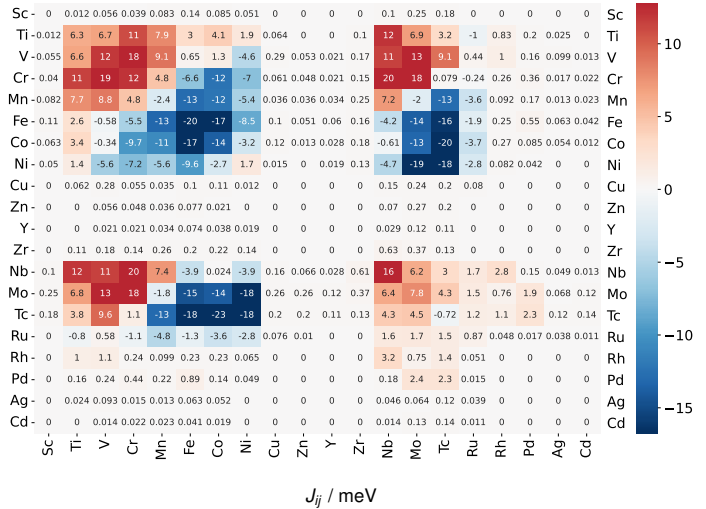
Magnetic property prediction

Data 2'000 dimer embeddings
Model SOAP+KRR
Performance $\mathrm{MAE} = 0.21$ meV, $\mathrm{R}^2 = 0.97$

Next steps

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





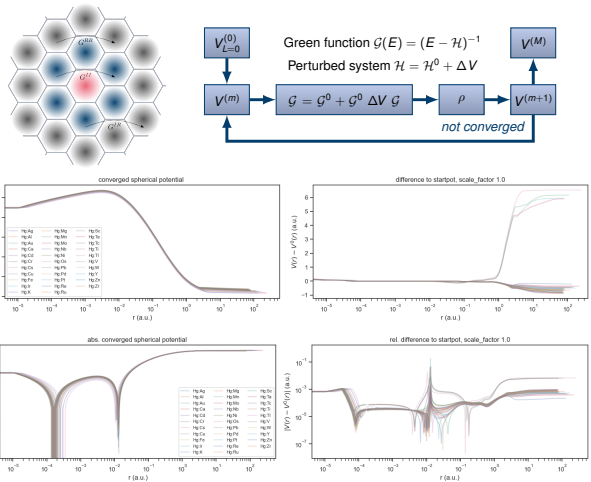
Electronic structure emulator

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

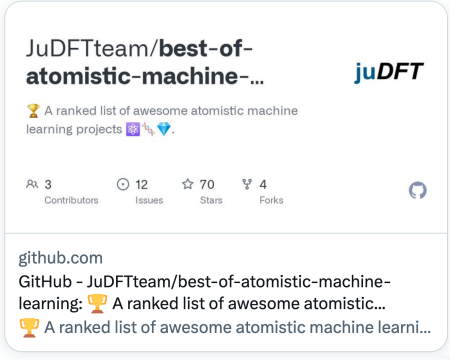
- **Collaboration** Adapt the linear Jacobi-Legendre Charge Density Model (JLCDM)[a] to all-electron DFT
- Large chemical feature space
- From radial to full equivariant potential
- Intermediate SCF results
- Should be code-agnostic
- From collinear to noco magnetism



Spherical impurity potentials in first Voronoi cell of Hg:X embeddings, left upper to bottom right: V, $V - V^0$, $|V|$, $|V - V^0|$.

[a] Focassio et al. 2023.

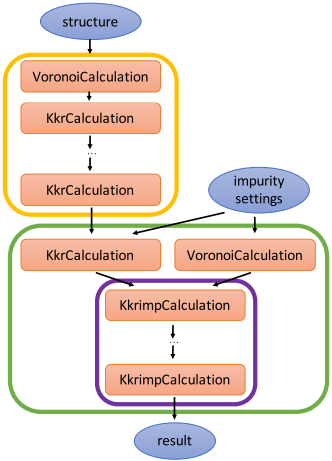
Tools & Infrastructure



↗ **Tools** • **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 →

Discussion slides



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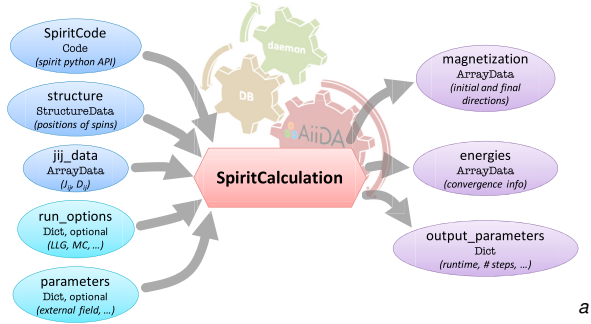

a

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

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

[a] Rüßmann, Bertoldo, and Blügel 2021.

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

[a] Rüßmann, Ribas Sobreviela, et al. 2022.

References I

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References III

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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. URL: https://www.frontiersin.org/articles/10.3389/fmats.2022.825043 (visited on 08/11/2022).