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

RESEARCH FOR GRAND CHALLENGES

Magnetic Multilayers: From High-Throughput Ab-initio Calculations to Predictive Machine Learning

Boosting DFT with Integrated ML

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Magnetic Multilayers: Spintronics Platform

Magnetic Multilayers properties are tuneable by:

- Choice of thickness
- Choice of layer composition
- Choice of growth conditions
- Choice of FM or AF coupling strength
- Possibility to modify *e.g.* DMI
- Device scalability

=> Convenient for materials design, but lack of systematic high-throughput studies

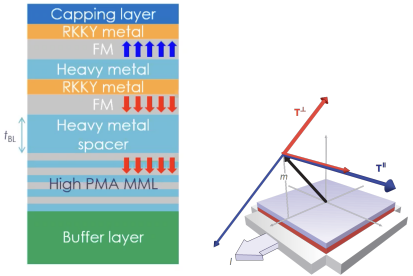

Figure: W. Legrand et al, Nature Materials 19, 34 (2020), Garello et al., Nature Nano. '13, Freimuth, Geranton, et al., '14–'17

Methodological Overview

Methods used:

- First-principles based structural relaxations using the FLEUR code
- Systematic high-throughput study supported by the AiiDA framework
- Necessary AiiDA-fleur plugin
- XGBoost ML model

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fleur

However, relaxing magnetic films can be challenging as:

- The convergence is strongly dependent on the initial state and structure
- Magnetic solutions can be non-unique
- Converged solutions can be meta-stable

2D Layered Film Structures

- Relaxing the A, B, and C layers in z-direction...
- ...using high-throughput DFT...
- ...for a total of 6660 films, allowing the outer layers to be unoccupied.
- Initial ILD's are set using average bond-lengths from the Materials Project database.
- Relaxation => Minimizing the interatomic-force

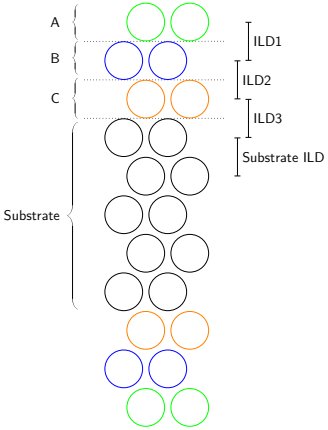

Figure: A, B,C $\in$ {Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn}, Substrate $\in$ {Pt, Au, Ag, Ir, Pd, Rh}.

Magnetic-Moment

A general problem in high-throughput DFT is input parameter estimation and tuning. Let's discuss the methodology we used in the following for the magnetic moment:

Magnetic Moments m_A, m_B, m_C:

- Magnetic films are examined => Ferromagnetic initialization
- Default initial moment for all atoms of 1 μ_B

However:

- Magnetic moments initialization choice can impact the output state...
- ...hence could introduce a bias...
- ...and affect convergence.

Results with ILD & Magnetic Moment Guesses

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Convergence rate:

- From 6660 films only 4316 converged
- Corresponding to 64.8 % convergence rate
- Too much, even for high-throughput

Errors occurring:

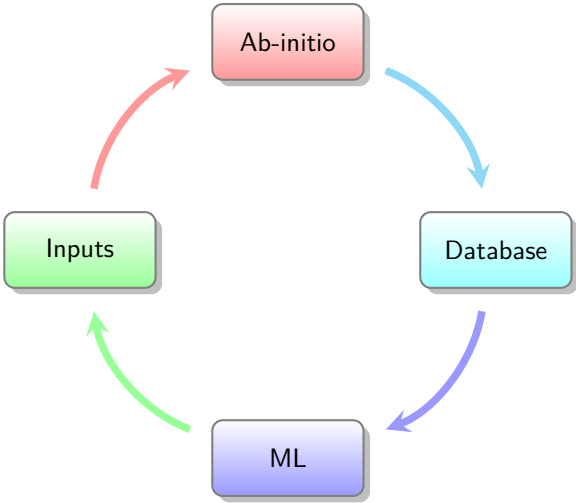
- Structural (*e.g.* layers repelling each other entirely)
- Suboptimal starting state (*e.g.* SCF loop did not converge)

How to fix this in a reasonable way? (Fixing $\approx$*2000* calculations manually is not a viable option!)

Idea: ML for Magnetic Moment Optimization

Using the converged compounds data to...

- .. train a ML model (in this case XGBoost) to predict the magnetic moments from the composition alone.
- .. predict m_A, m_B, and m_C for failed films.
- .. use the ML prediction as input to restart.



Intermediary Result: Improved Convergence Rate

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- 570 more systems converged
- Increased convergence rate to 73.3 %
- Better convergence rate by simply optimizing the magnetic starting point
- Still large error rate

ILD

Interlayer Distances d_{AB}, d_{BC}, d_{CSub}:

- Interlayer-Distance (ILD) estimation by mean average bond lengths
- Computed pairwise for all atom pairs
- Based on all bonds in the Materials Project structures database

Optimization could assist with:

- Reducing number of relaxation steps required
- Assisting with method specific problems which emerge from bad ILD estimates

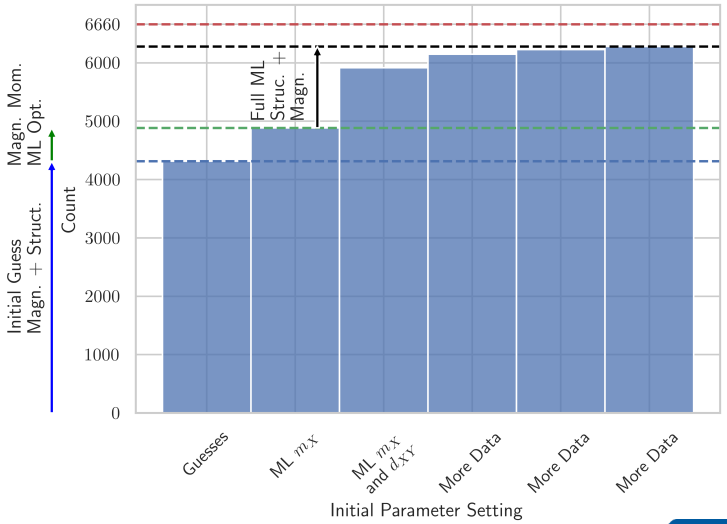
2nd Idea: ML for ILD Optimization

Using the converged compounds data to...

- .. train a ML model (in this case XGBoost) to predict the magnetic moments from the composition alone.
- .. predict d_{AB}, d_{BC}, and d_{CSub} for failed films.
- .. use the ML prediction as input to restart.

=> This time also structural optimization and replacement of the ILD guess!

Accumulated Convergence Rate



Guess Improvement (When to start with ML?)

Posterior analysis:

- From about 200 structures, ML beats classical estimate
- Ideally, we should've started earlier with this method
- Best case: Continuous model integration

Additional Benefits of DFT Integrated ML

Beyond the convergence rate improval up to 94 % advantages are:

- Optimizing Structure and Magnetism => Reduced needed relaxation steps by 27 %
- ILD prediction has 70 % lower error - compared to the ab-initio results - than the bond length estimate guess.
- Easy to implement on a **high-throughput scale**.
- This method is not restricted to magnetic moments and ILD's. **Any quantity** which is in-/ **and** output of an ab-initio calculation can be learned and predicted.

Conclusion & Acknowledgements

- Integrating ML into high-throughput ab-initio studies can benefit both convergence rate, as well as computational efficiency
- ML puts existing data to a use, enabling it to accelerate the scientific discovery process instead of just being accumulated
- ML complements ab-initio methods in high-throughput applications by making input parameter tuning feasible on a large scale

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