Curie Temperature Prediction Models of Magnetic Heusler Alloys

Using Machine Learning Methods Based on First-Principles Data From Ab-initio KKR-GF Calculations

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Question:
Can we compute complex magnetic material properties easier and faster using Machine-Learning?

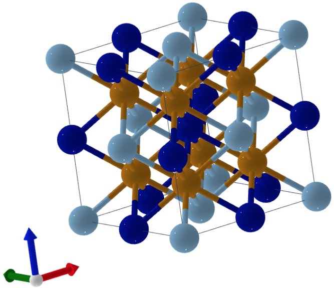


Figure: FeFeCrAl Heusler structure

Example: Curie-Temperature

- Major magnetic quantity
- Complex computing (Ab-initio, Exchange params, MC)
- Application requires $T_c > T_{Room}$

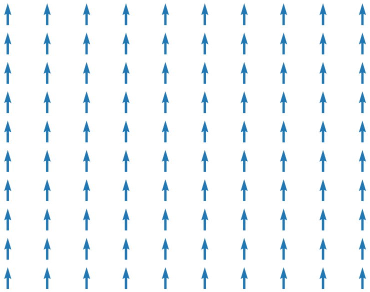
Example material class: Heusler alloys

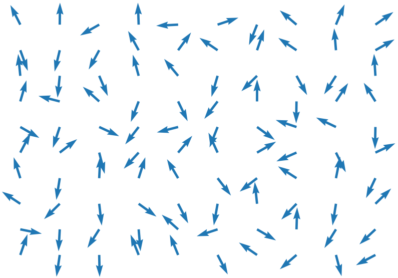
- Subclass of materials
- Structural homogeneity

Curie Temperature

$T < T_c$: $\langle M \rangle = \frac{\sum_i m_i}{V}$

$T > T_c$: $\langle M \rangle = 0$
without external fields





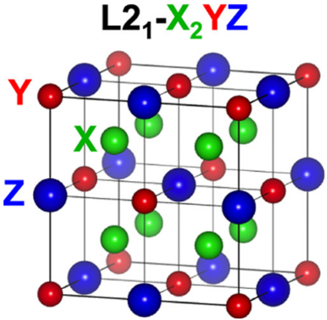


Figure: Heusler structure, from [Kojima et al., 2017]

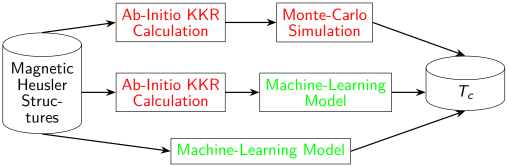
Heuslers properties:

- Magnetism
- Thermoelectricity
- Superconductivity
- etc.

X, Y $\in$ transition metals
Z $\in$ main-groups 3-5

Machine Learning

- Computing T_c is very expensive
- Criterion $T_c > T_{Room}$ makes applicability classifiable
- High-Throughput screening possible with ML



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- 776 Systems incl. disordered
- Experimental and theoretical T_c
- Theoretical based on ab-initio + MC
- Publicly available database under CC-by 4.0 [Kováčik et al., 2022]

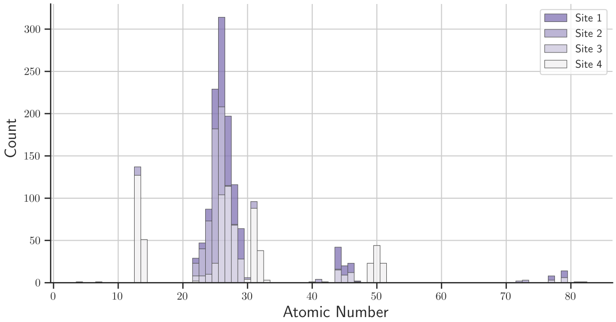


Figure: Distribution of atomic numbers post-processing

Data Properties

- Post-processing: 408 structures
- Visible outlier in T_c distribution removed
- 118 possible descriptors:
  - Structural
  - Electronic
  - Magnetic
  - Atomic

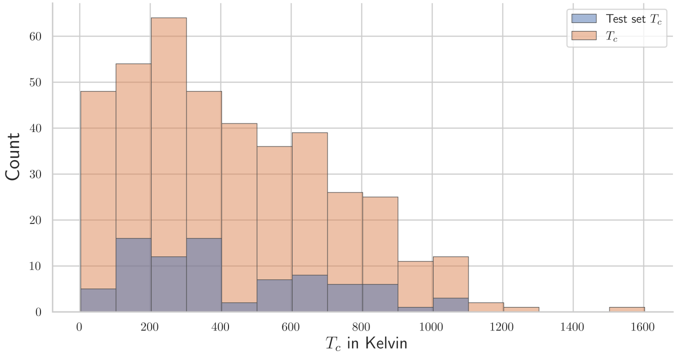
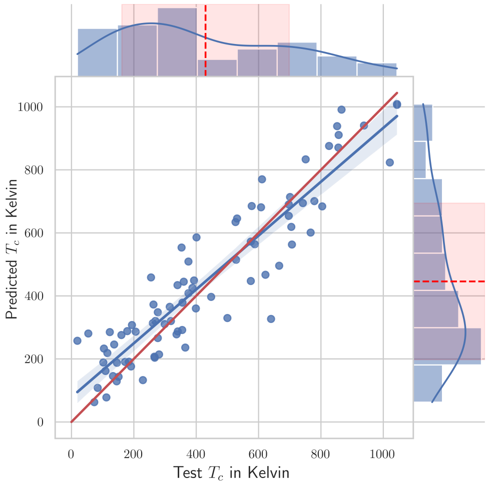


Figure: T_c distributions

Regression

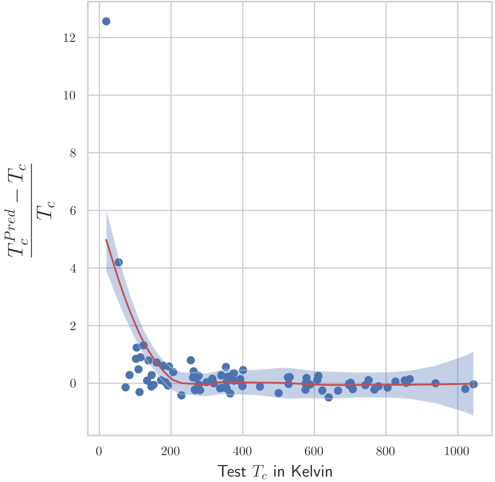
- Extra Trees Regression
- R^2 on test set ≈ 0.85
- Mean prediction deviations $\approx$ 60 K
  - Matches accuracy of DFT + MC approach compared to the experiment
- Linear regression deviates from the ideal (red)



Regression - Residuals

- $T_c < T_{Room}$ are overestimated
  - $T_c < T_{Room}$ is not relevant for application

- Besides that no systematic discrepancies are visible



Classification

- Using all data β error $< 3\%$
- Excluding magnetic + energy data from DFT β error $\approx 5\%$
  - Structural, electronic and atomic data allows applicability classification

Upcoming publication of all findings and approach details.

Full descriptor set:

Model	CV-Score	Test F1 Score	Test Accuracy
Extra Trees	0.82165	0.90625	0.92683
Logistic Reg.	0.82209	0.86154	0.89024

Descriptors excluding magnetic and energetic DFT results:

Model	CV-Score	Test F1 Score	Test Accuracy
Extra Trees	0.74196	0.83582	0.86585
Logistic Reg.	0.68224	0.75182	0.75362

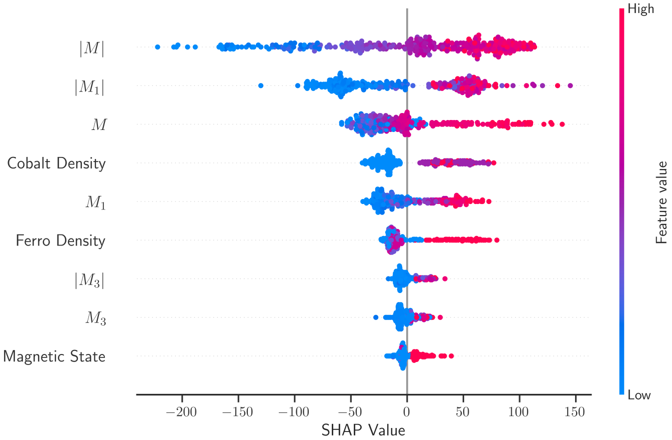
Shapley values originate from a game-theory approach and hence are interpretable. Implemented in the SHAP package [Lundberg et al., 2020]

SHAP is...

- optimized for scikit-learn
- model independent
- capable of visualizing Shapley values

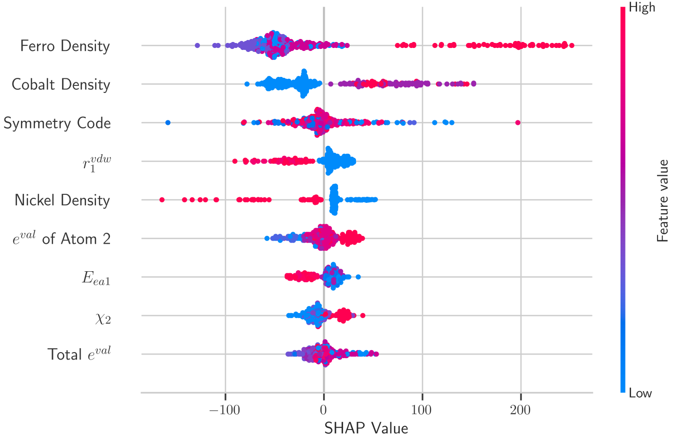
Feature Importance - Including DFT data

- SHAP beeswarm plots display feature values vs Shapley values
- Ordered by impact on prediction
- 9 most impactful all magnetic
- Most impactful: $|M| = \sum_i |m_i|$



Feature Importance - Without DFT data

- Same approach excluding DFT results
- Three magnetism related:
  - n_{Ferro}, n_{Cobalt}, n_{Nickel}
- Special interpretation: Symmetry code
- Surprise: Increased Nickel density has negative impact



Key Takeaways

- Reasonable predictions with small data set
- Physical interpretable XAI results
- Materials screening using ML is cheap and fast

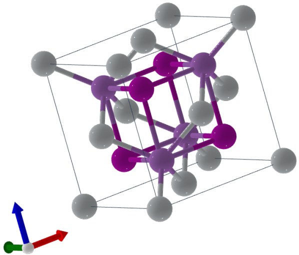


Figure: MnNiSb half-Heusler structure

Acknowledgements

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Thank you for your attention!

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