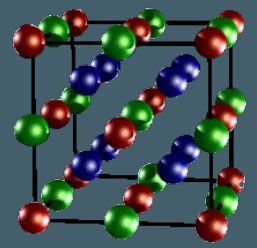
Los Alamos NATIONAL LABORATORY

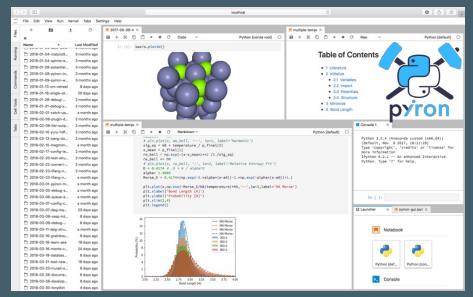
MAX PLANCK INSTITUTE FOR SUSTAINABLE MATERIALS

Machine Learned Interatomic Potentials for Extreme Environments

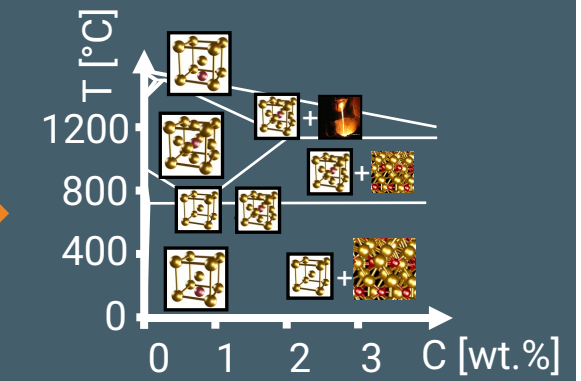
Jan Janssen – Group Leader for Materials Informatics



Simulation



Machine Learning



Experiment

May 27th - Models and Data for Plasma-Material Interactions in Fusion Devices

Artificial Intelligence

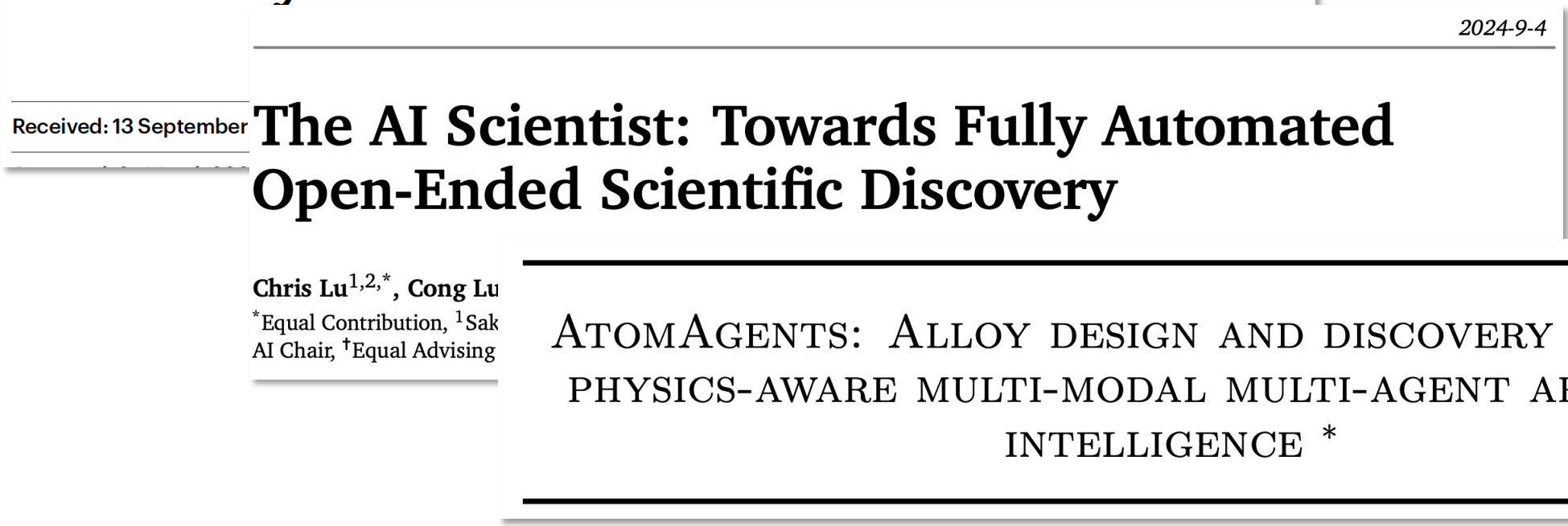
Can a Large Language Model replace a scientist?

nature machine intelligence

Article

https://doi.org/10.1038/s42256-024-00832-8

Augmenting large language models with chemistry tools



2024-9-4

Received: 13 September

The AI Scientist: Towards Fully Automated Open-Ended Scientific Discovery

Chris Lu[1,2,*], Cong Lu

*Equal Contribution, [1]Sak
AI Chair, †Equal Advising

ATOMAGENTS: ALLOY DESIGN AND DISCOVERY THROUGH PHYSICS-AWARE MULTI-MODAL MULTI-AGENT ARTIFICIAL INTELLIGENCE *

M.C. Ramos et al., arXiv 2407.01603 (2024).

Artificial Intelligence

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Article

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Augmenting large language models with chemistry tools

Recent publications claim that
Large Language Models can predict novel materials.

Chris Lu[1,2,*], Cong Lu
*Equal Contribution, [1]Sak
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AtomAgents: Alloy design and discovery through physics-aware multi-modal multi-agent artificial intelligence *

M.C. Ramos et al., arXiv 2407.01603 (2024).

State of the Art - ChatGPT 4o Model

Hands-on: Let's give it a try!

Calculate the bulk modulus for aluminium with ASE using the EMT simulation code.

https://chatgpt.com

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Physical Parameters,

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Physical Parameters, Technical Parameters

https://chatgpt.com

State of the Art - ChatGPT 4o Model

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Calculate the bulk modulus for aluminium with ASE using the EMT simulation code.

Answer form ChatGPT:

Physical Parameters, Technical Parameters

Thought for 12 seconds

To calculate the bulk modulus of aluminum using the Atomic Simulation Environment (ASE) with the Effective Medium Theory (EMT) potential, you can follow these steps:

1. **Set up the aluminum bulk structure.**
2. **Assign the EMT calculator to the structure.**
3. **Compute the energies at different volumes by scaling the unit cell.**
4. **Fit the calculated energies and volumes to an equation of state (EOS).**
5. **Extract the bulk modulus from the EOS fit.**

https://chatgpt.com

State of the Art - ChatGPT 4o Model

Hands-on: Let's give it a try!

Calculate the bulk modulus for aluminium with ASE using the EMT simulation code.

Answer form ChatGPT:

[1]:
```
from ase.build import bulk
from ase.calculators.emt import EMT
from ase.eos import EquationOfState
import numpy as np
```

[2]:
```
# Create an aluminum bulk structure
al_bulk = bulk('Al', 'fcc', a=4.05, cubic=True)
```

[3]:
```
# Attach the EMT calculator
al_bulk.calc = EMT()
```

[4]:
```
# Define a range of scaling factors for volume
scaling_factors = np.linspace(0.94, 1.06, 10)  # Scale between 94% and 106%
volumes = []
energies = []
```


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Technical Parameter:
Atomic Simulation Environment (ASE)

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Physical Parameter:
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State of the Art - ChatGPT 4o Model

Hands-on: Let's give it a try!

Calculate the bulk modulus for aluminium with ASE using the EMT simulation code.

Answer form ChatGPT:

[5]:
```python
# Loop over scaling factors, calculating energy for each
for factor in scaling_factors:
    al_bulk.set_cell(al_bulk.get_cell() * factor, scale_atoms=True)
    volumes.append(al_bulk.get_volume())
    energies.append(al_bulk.get_potential_energy())
```

[6]:
```python
# Fit the Equation of State (EOS)
eos = EquationOfState(volumes, energies)
v0, e0, B = eos.fit()  # Fit returns equilibrium volume, energy, and bulk modulus
```

[7]:
```python
# Print results
print(f"Equilibrium volume: {v0:.2f} Å³")
print(f"Equilibrium energy: {e0:.2f} eV")
print(f"Bulk modulus: {B:.2f} GPa")
```

```
Equilibrium volume: 62.50 Å³
Equilibrium energy: -0.03 eV
Bulk modulus: 0.26 GPa
```


https://chatgpt.com

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```
Equilibrium volume: 62.50 Å³
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```

Experiment: 62 GPa

https://chatgpt.com

State of the Art - ChatGPT 4o Model

Hands-on: Let's give it a try!

Calculate the bulk modulus for aluminium with ASE using the EMT simulation code.

Debugging:

```
[9]: eos.fit??
```

```
Signature: eos.fit(warn=True)
Source:
    def fit(self, warn=True):
        """Calculate volume, energy, and bulk modulus.

        Returns the optimal volume, the minimum energy, and the bulk
        modulus.  Notice that the ASE units for the bulk modulus is
        eV/Angstrom^3 - to get the value in GPa, do this::

          v0, e0, B = eos.fit()
          print(B / kJ * 1.0e24, 'GPa')

        """
```

With Correction: 40 GPa Experiment: 62 GPa

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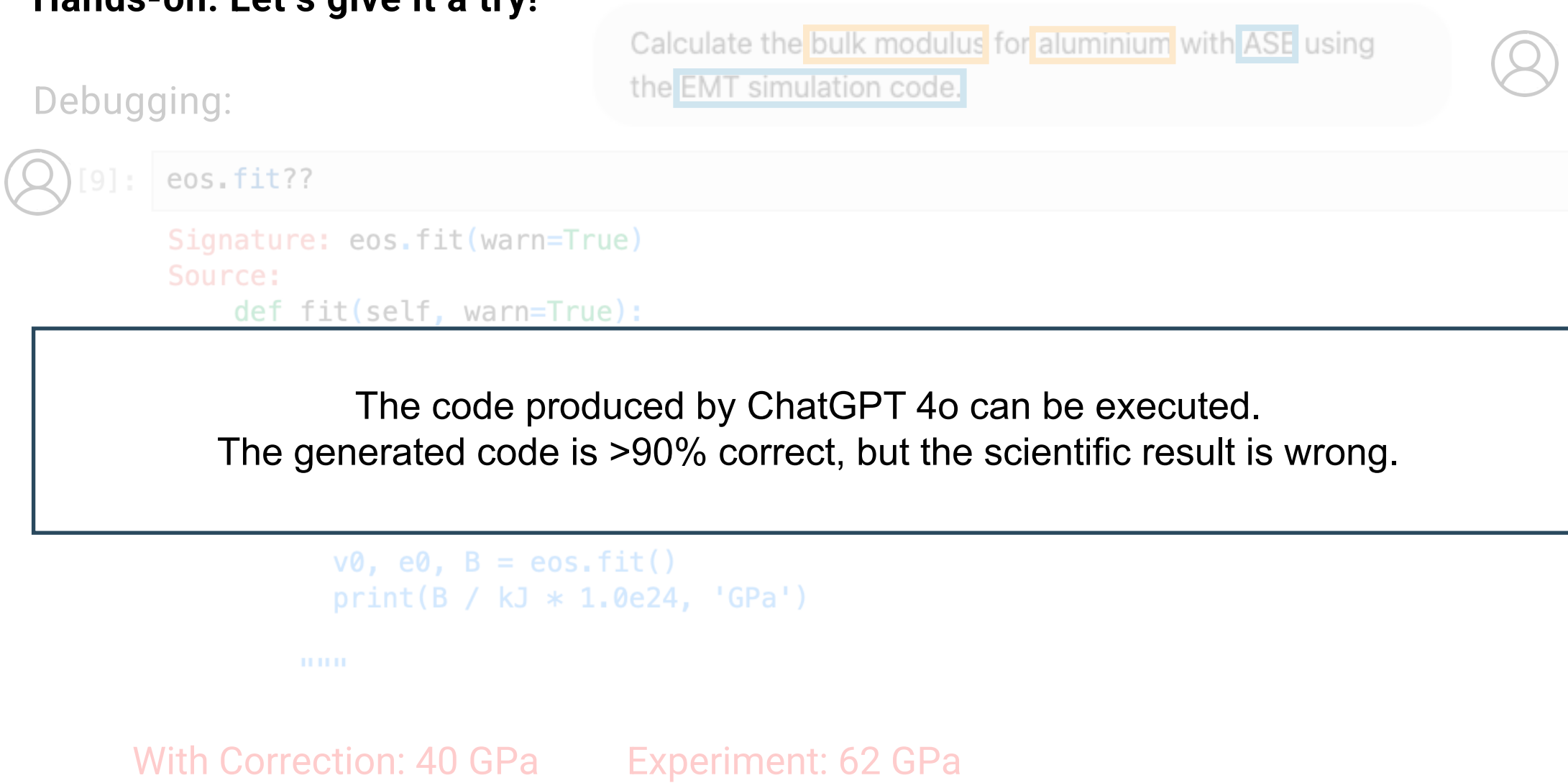
State of the Art - ChatGPT 4o Model

Hands-on: Let's give it a try!

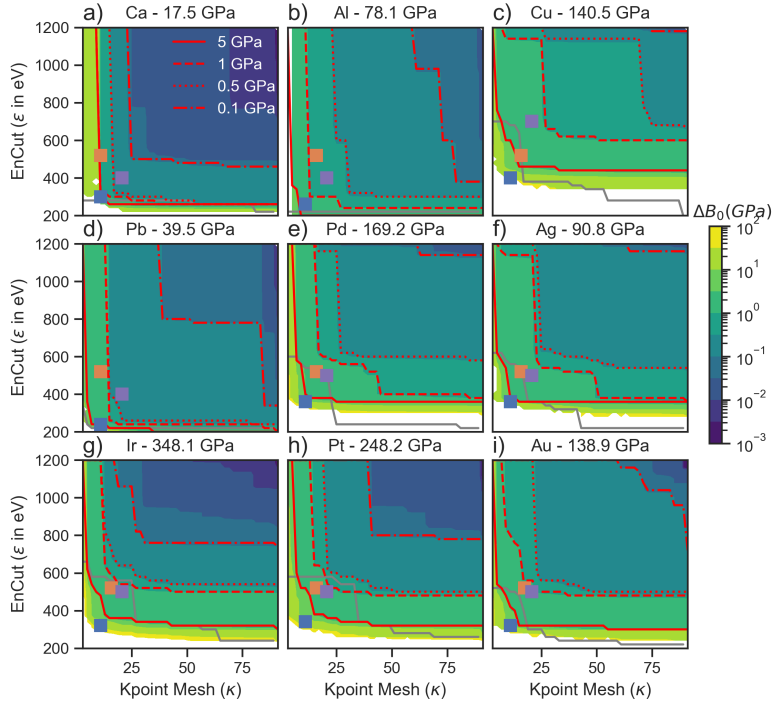


The code produced by ChatGPT 4o can be executed.
The generated code is >90% correct, but the scientific result is wrong.

Materials Informatics Group

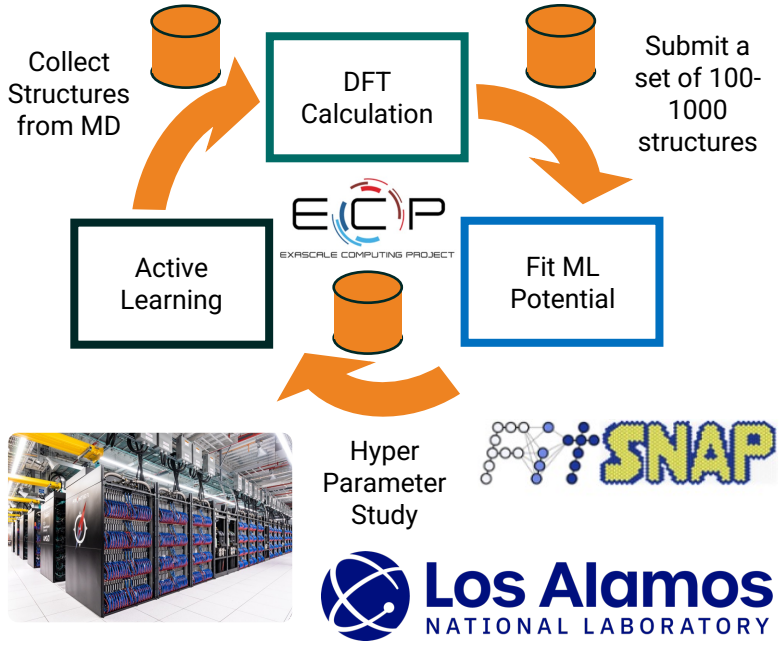
Our Expertise: Workflows for Sustainable Materials

Uncertainty Propagation for Density Functional Theory



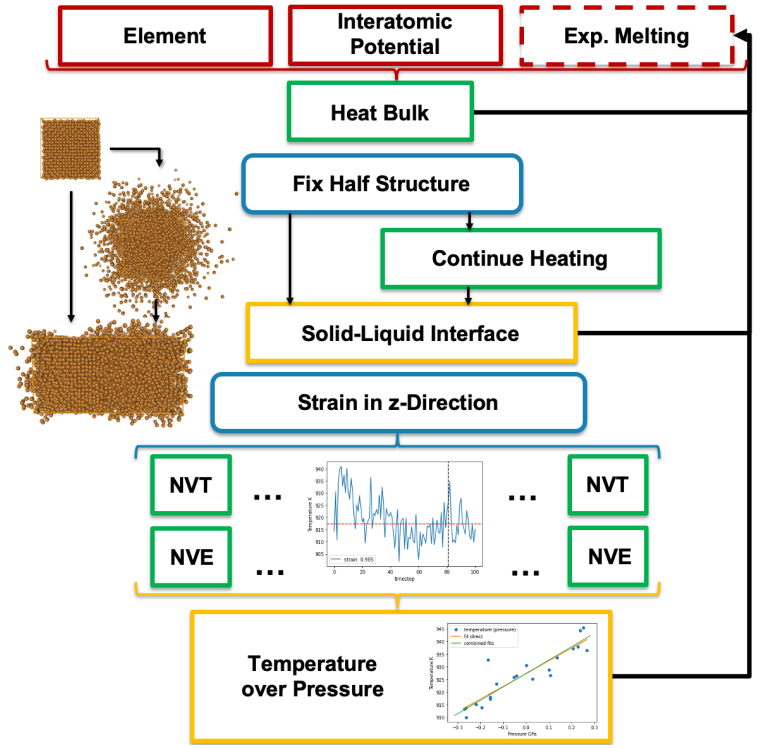
J. Janssen, et al.,npj Comput. Mater., 10, 263 (2024)

Machine-Learned Interatomic Potentials for Extreme Environments



A. Rohskopf, C. Sievers, N. Lubbers, M.a. Cusentino, J. Goff, J. Janssen, et al. JOSS, 8 (2023)

Automated Workflow for Calculating Melting Temperatures

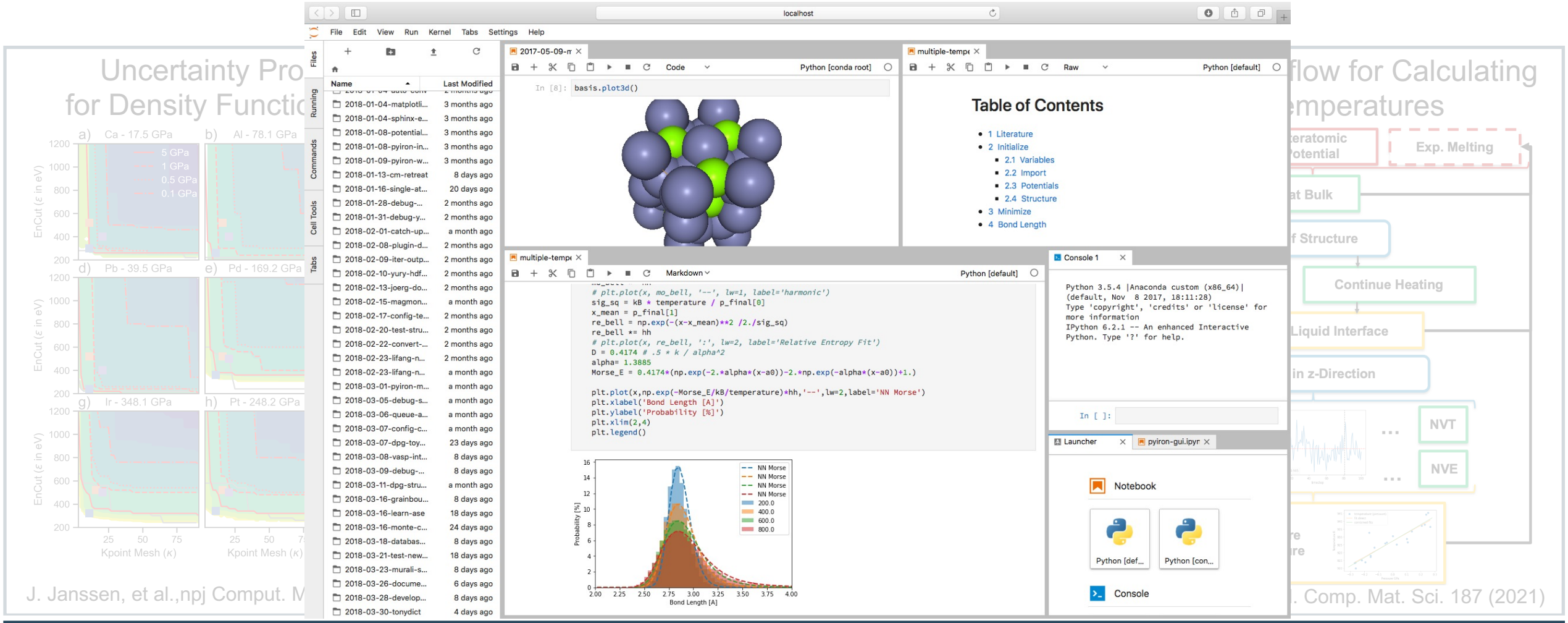


L.F. Zhu, J. Janssen, et al. Comp. Mat. Sci. 187 (2021)

pyiron Workflow Framework

Materials Informatics Group

Our Expertise: Workflows for Sustainable Materials



pyiron Workflow Framework

J. Janssen, et al., Comp. Mat. Sci. 161 (2019) - https://pyiron.org

Materials Informatics Group

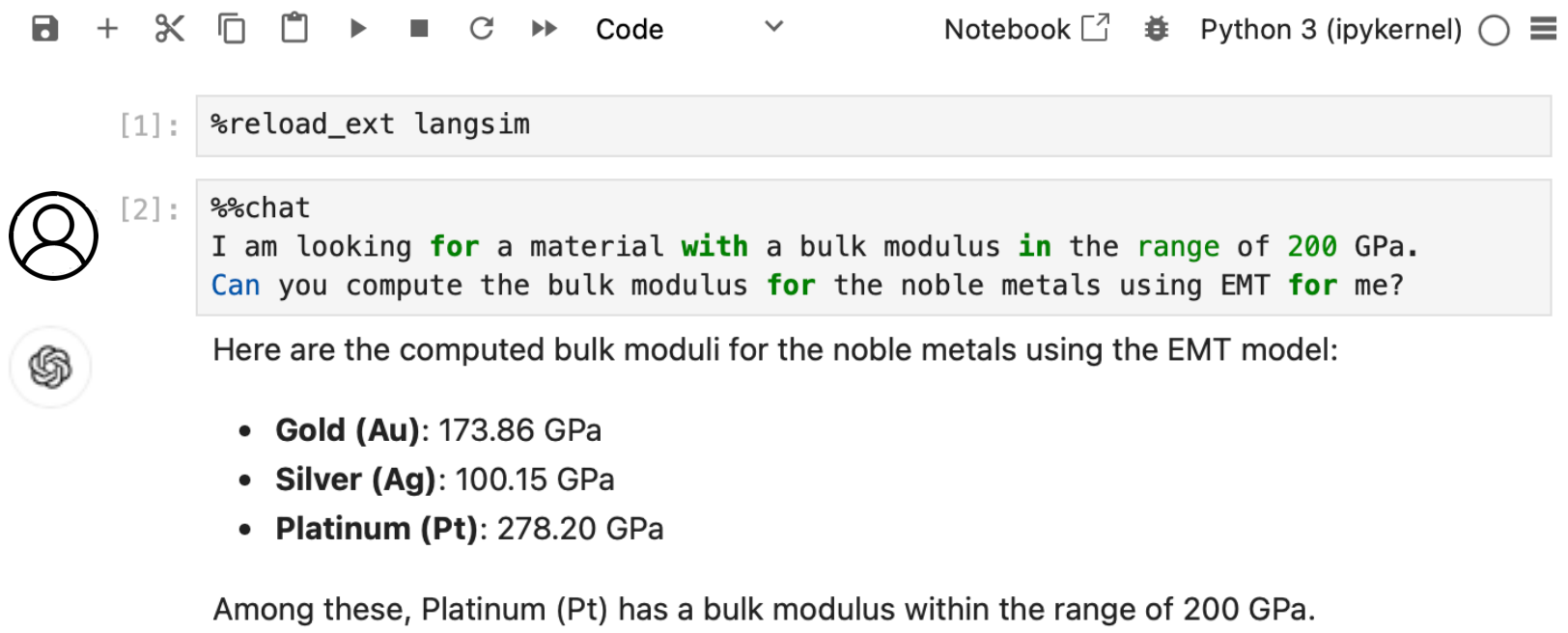
Our Expertise: Workflows for Sustainable Materials

The pyiron workflow framework enables data-driven materials science, by moving away from the terminal-/shell-based interface towards a programmatic Python-based interface.

pyiron Workflow Framework

LangSim

pyiron based Large Language Model Interface for Atomistic Simulation



https://jan-janssen.com/LangSim - arXiv 2505.03049 (2025).

LangSim

pyiron based Large Language Model Interface for Atomistic Simulation

Code Notebook Python 3 (ipykernel)

[1]:
```
%reload_ext langsim
```

[2]:
```
%%chat
I am looking for a material with a bulk modulus in the range of 200 GPa.
Can you compute the bulk modulus for the noble metals using EMT for me?
```

Here are the computed bulk moduli for the noble metals using the EMT model:

- **Gold (Au)**: 173.86 GPa
- **Silver (Ag)**: 100.15 GPa
- **Platinum (Pt)**: 278.20 GPa

Among these, Platinum (Pt) has a bulk modulus within the range of 200 GPa.

[3]:
```
%%chat
How does this compare to experiment?
```

Here are the experimental bulk moduli for the noble metals:

- **Gold (Au)**: 180 GPa
- **Silver (Ag)**: 100 GPa
- **Platinum (Pt)**: 230 GPa

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> Entering new AgentExecutor chain...

Invoking: `get_atom_dict_bulk_structure` with `{'chemical_symbol': 'Au'}`
numbers=[79] positions=[[0.0, 0.0, 0.0]] cell=[[0.0, 2.04, 2.04], [2.04, 0.0,
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       Step     Time          Energy          fmax
LBFGS:    0 19:55:11        0.002606        0.308859
LBFGS:    1 19:55:11        0.000032        0.077808
LBFGS:    2 19:55:11       -0.000135        0.003099
LBFGS:    3 19:55:11       -0.000135        0.000029
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Using specialized agents, the Large Language Model (LLM) can interface with atomistic simulation codes and utilities.

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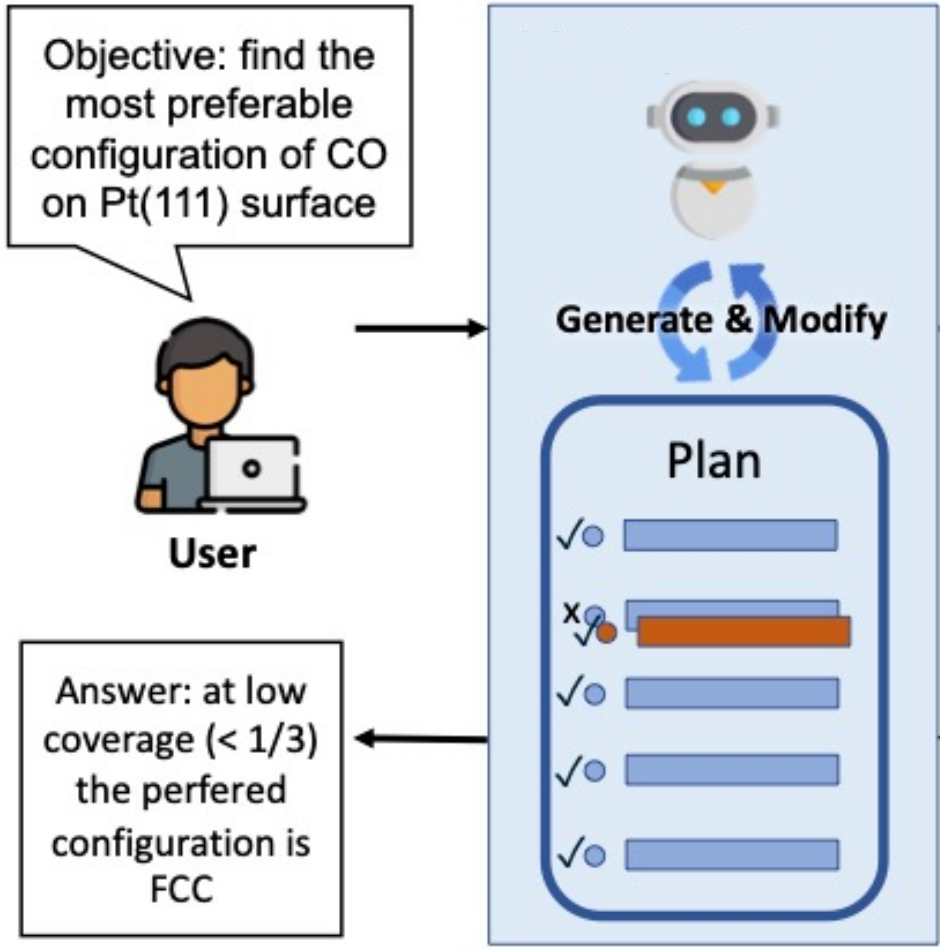
DFT-based Research Engine for Automated Materials Screening

Collaborative Team of Large Language Model Agents



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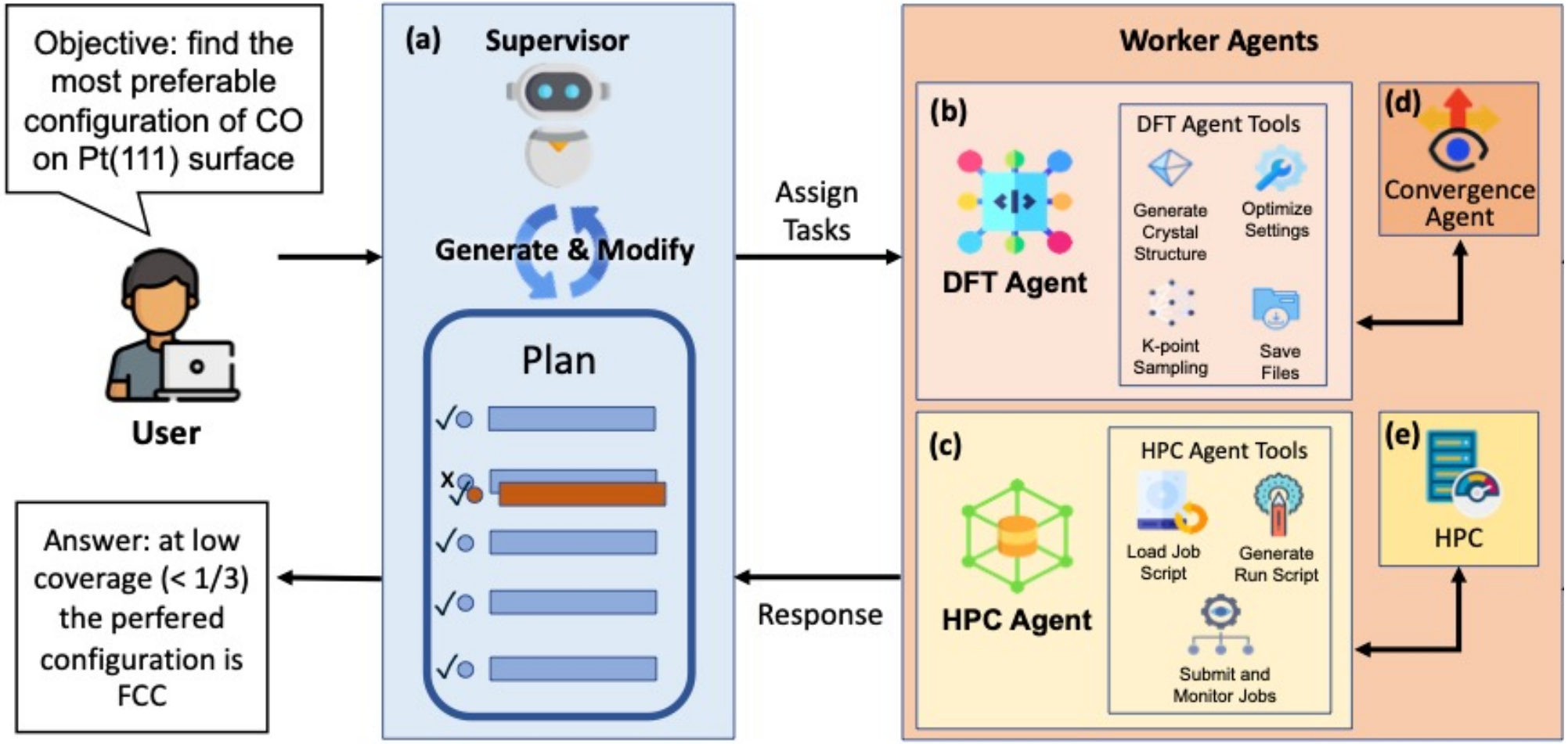

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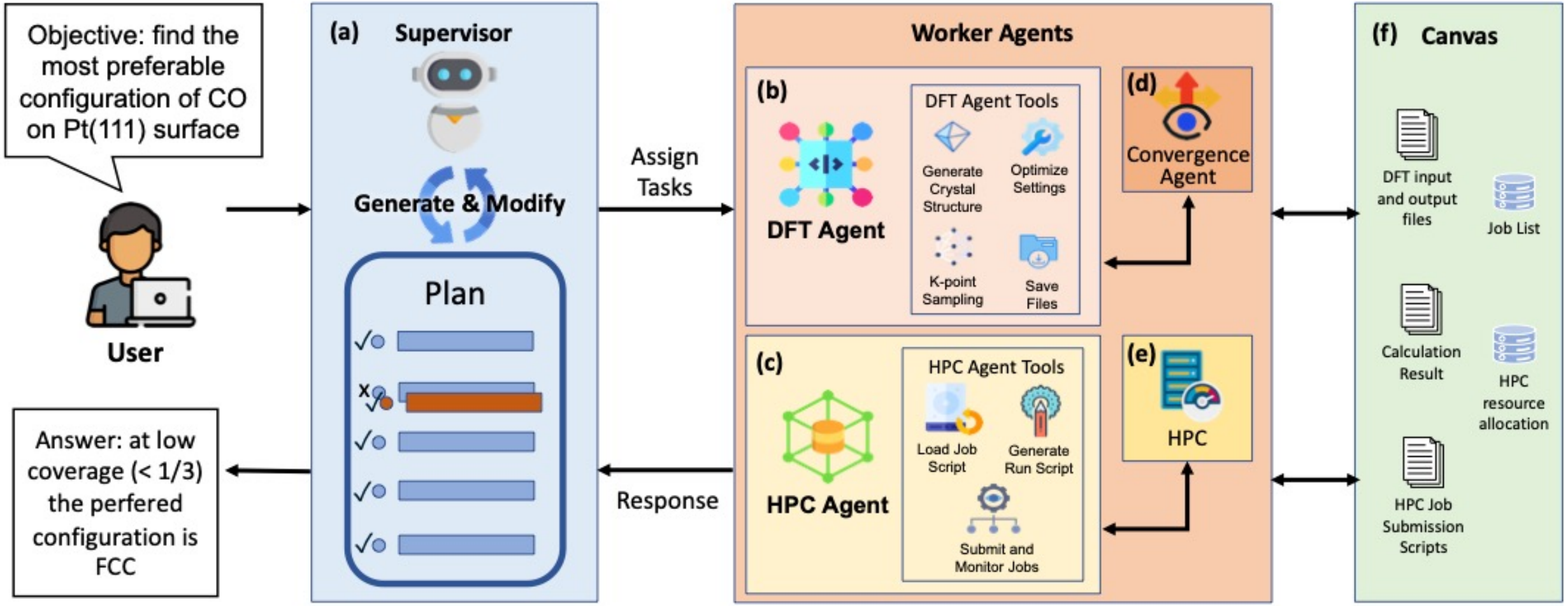

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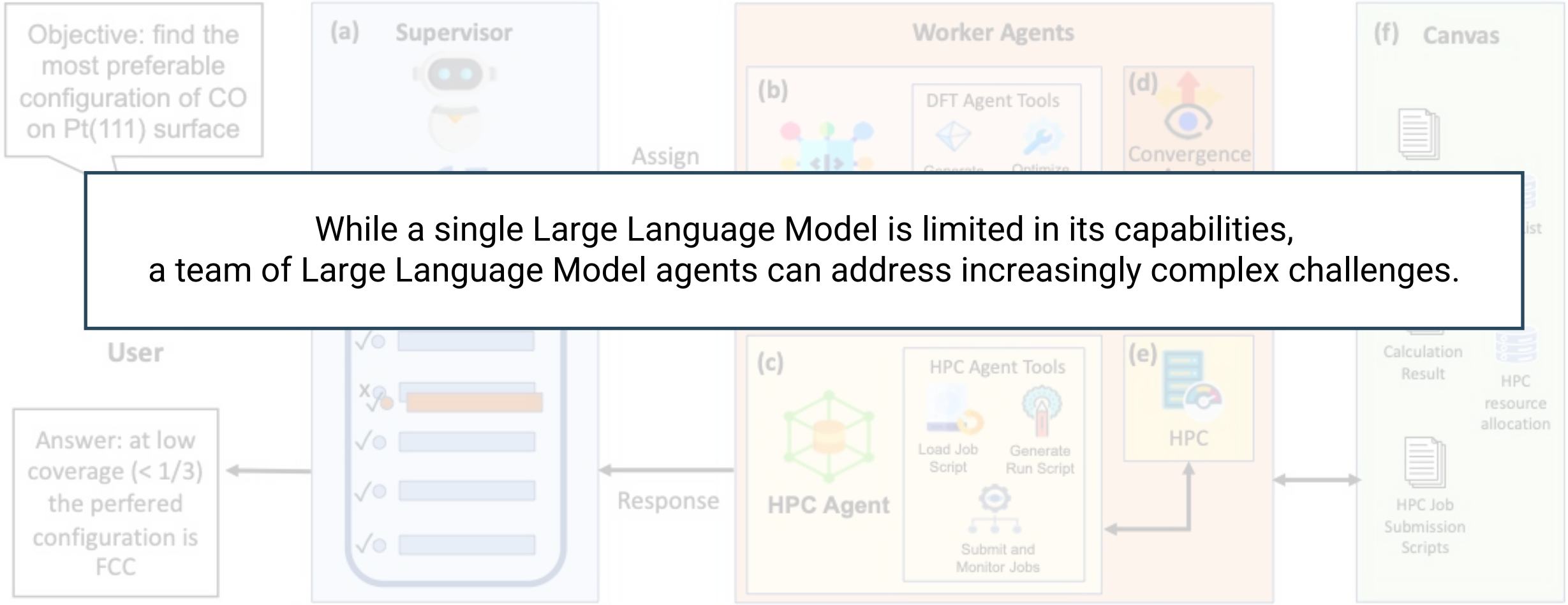

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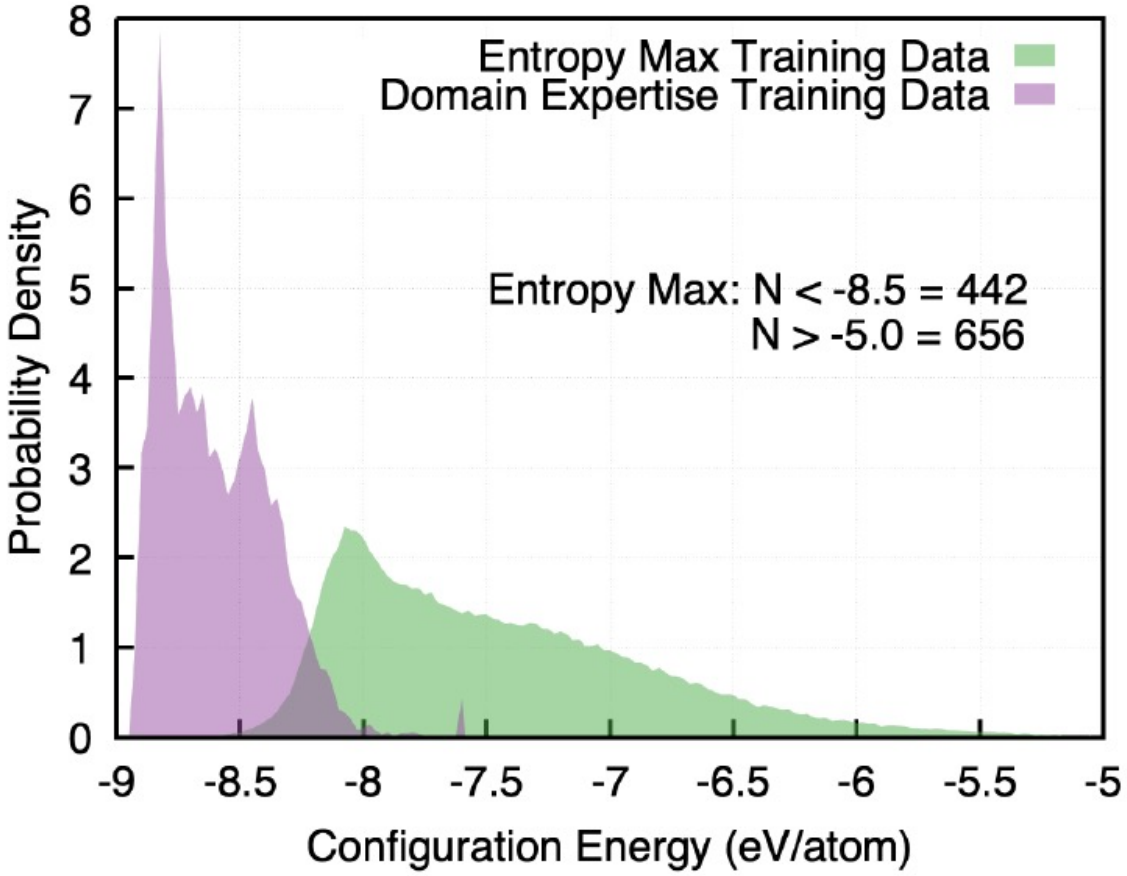
Transferable Machine-Learned Interatomic Potentials

Generate a diverse training set based on maximizing the informational entropy



M. Karabin, et. al. J. Chem. Phys. 153, 094110 (2020)

Montes de Oca Zapiain, D., et al. npj Comput Mater 8, 189 (2022).

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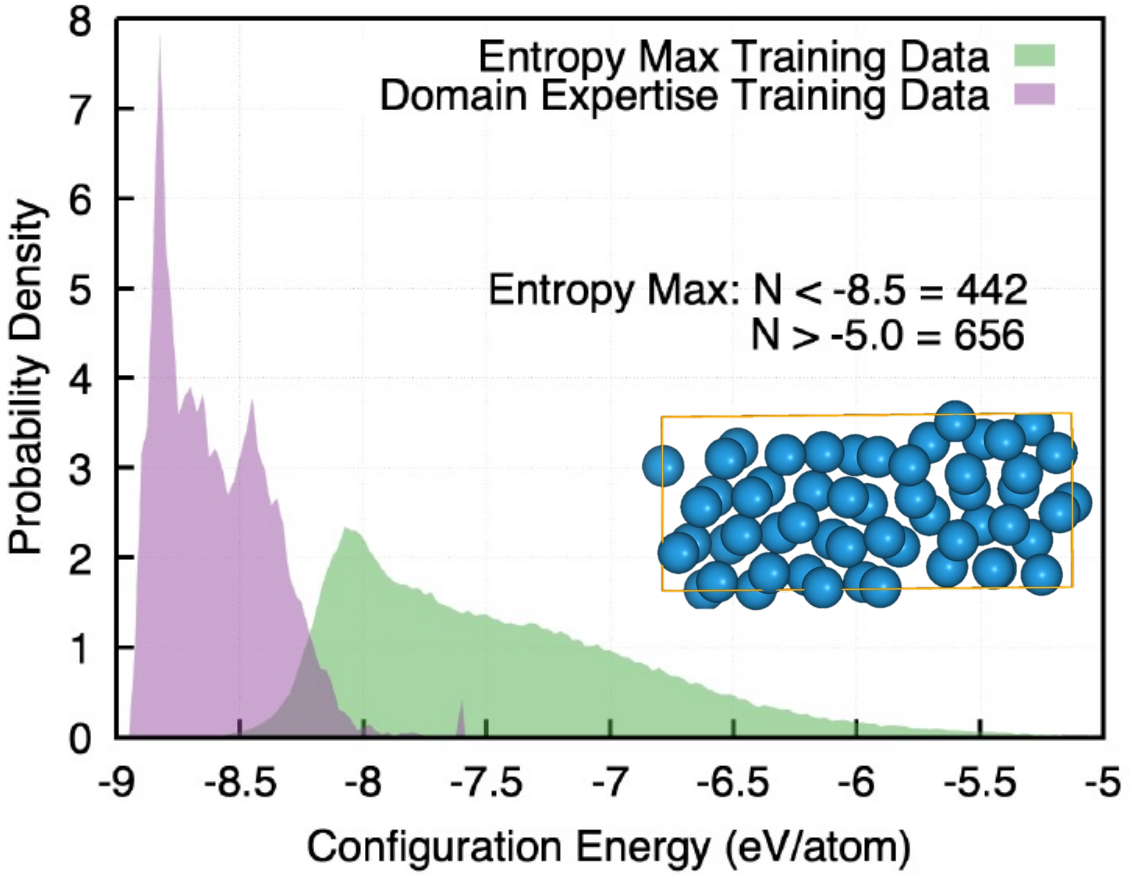

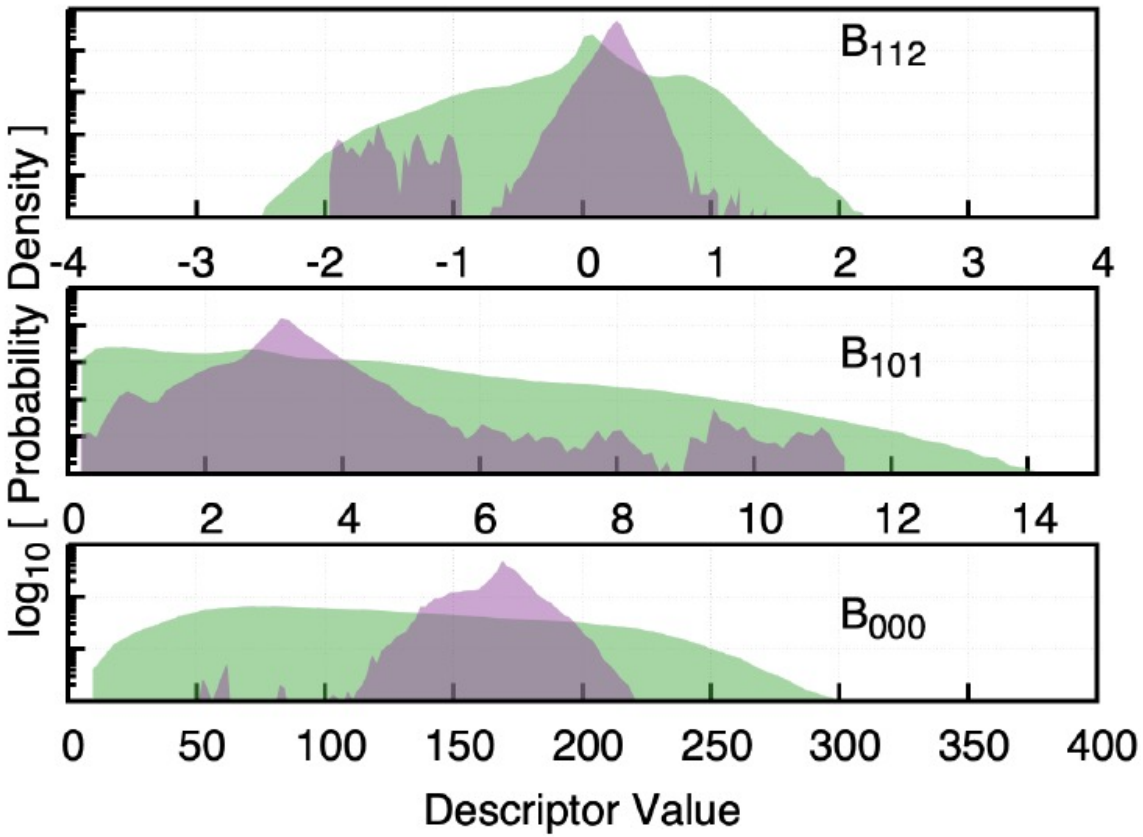

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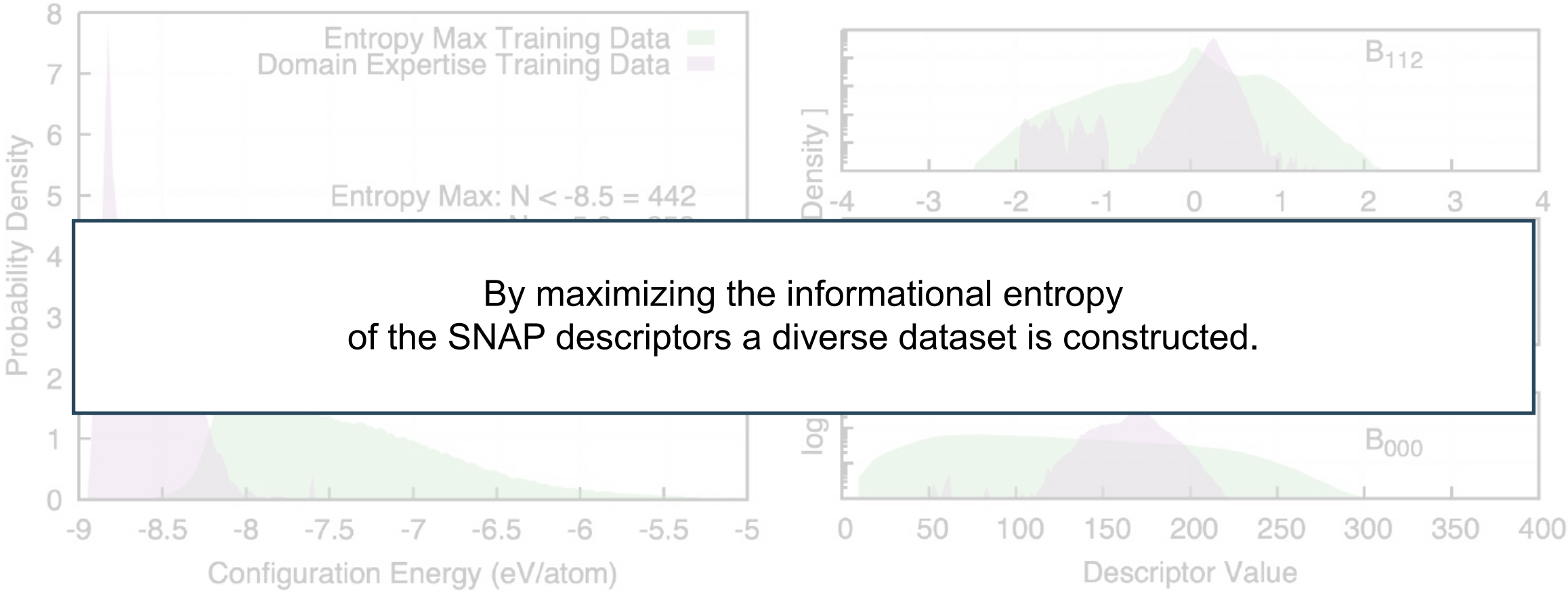


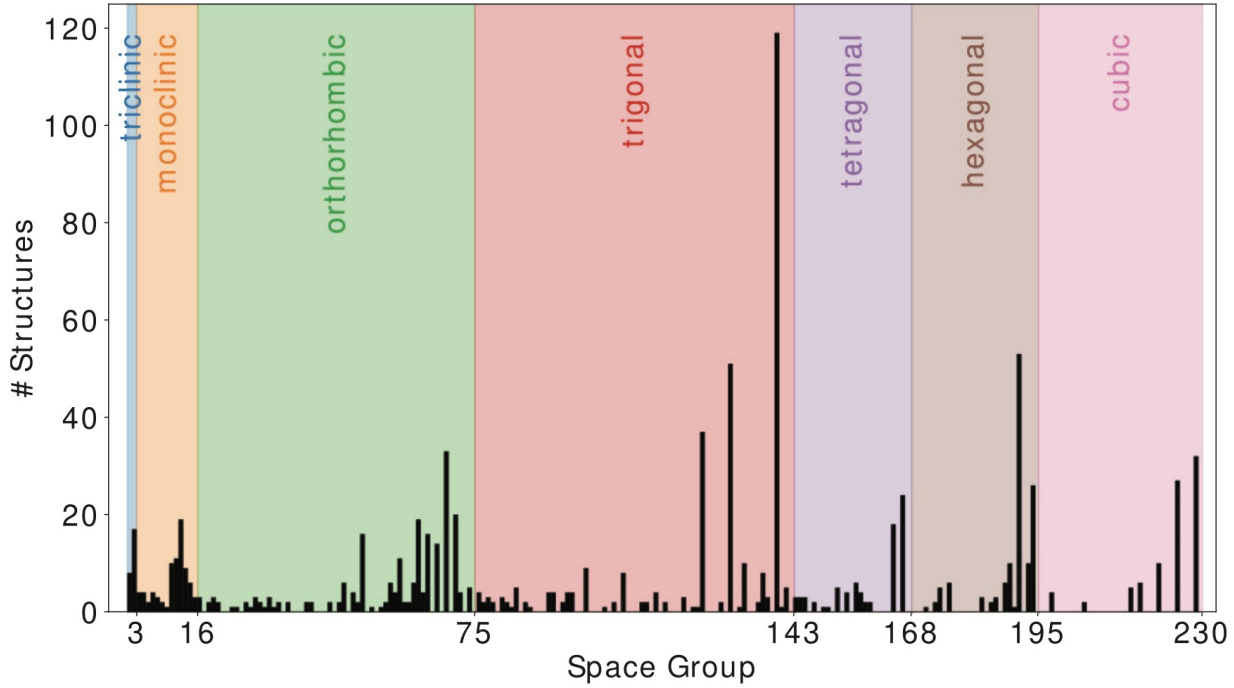
By maximizing the informational entropy
of the SNAP descriptors a diverse dataset is constructed.

M. Karabin, et. al. J. Chem. Phys. 153, 094110 (2020)
Montes de Oca Zapiain, D., et al. npj Comput Mater 8, 189 (2022).

Divers Training Sets

Automated Small SYmmetric Structure Training (ASSYST)

Space Group Symmetry based Structures



M. Poul, L. Huber, E. Bitzek and J. Neugebauer., Phys. Rev. B 107 (2023)

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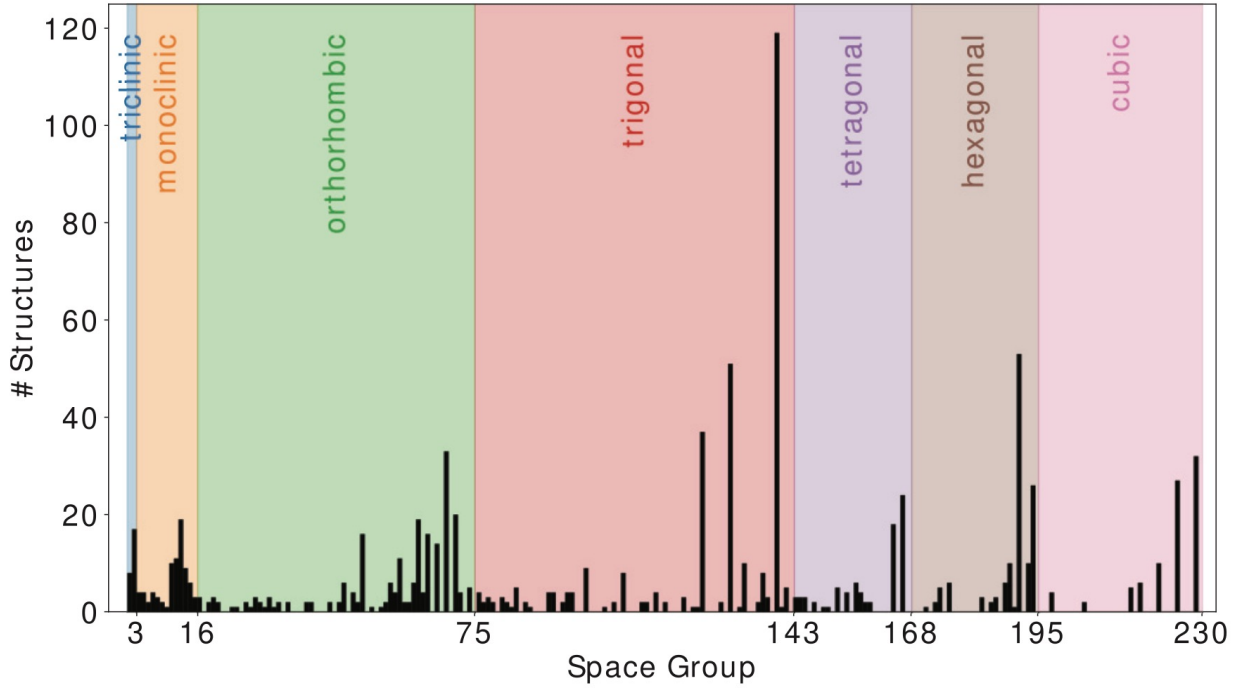

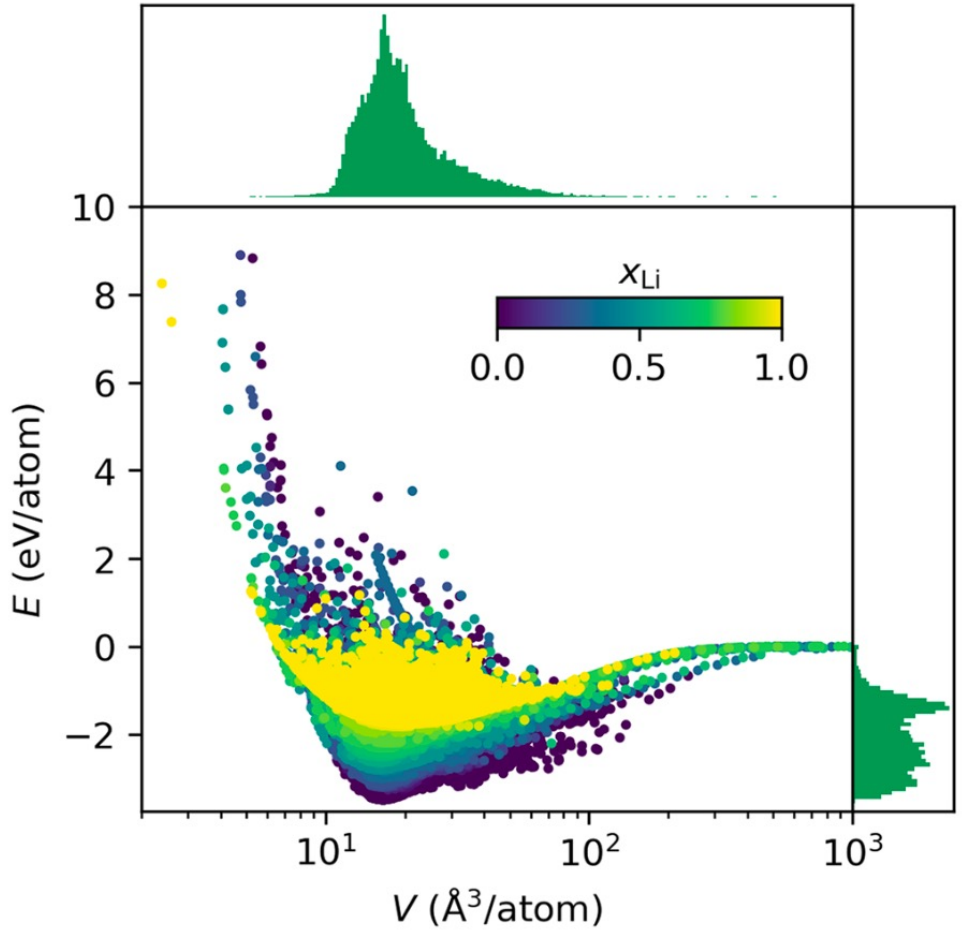

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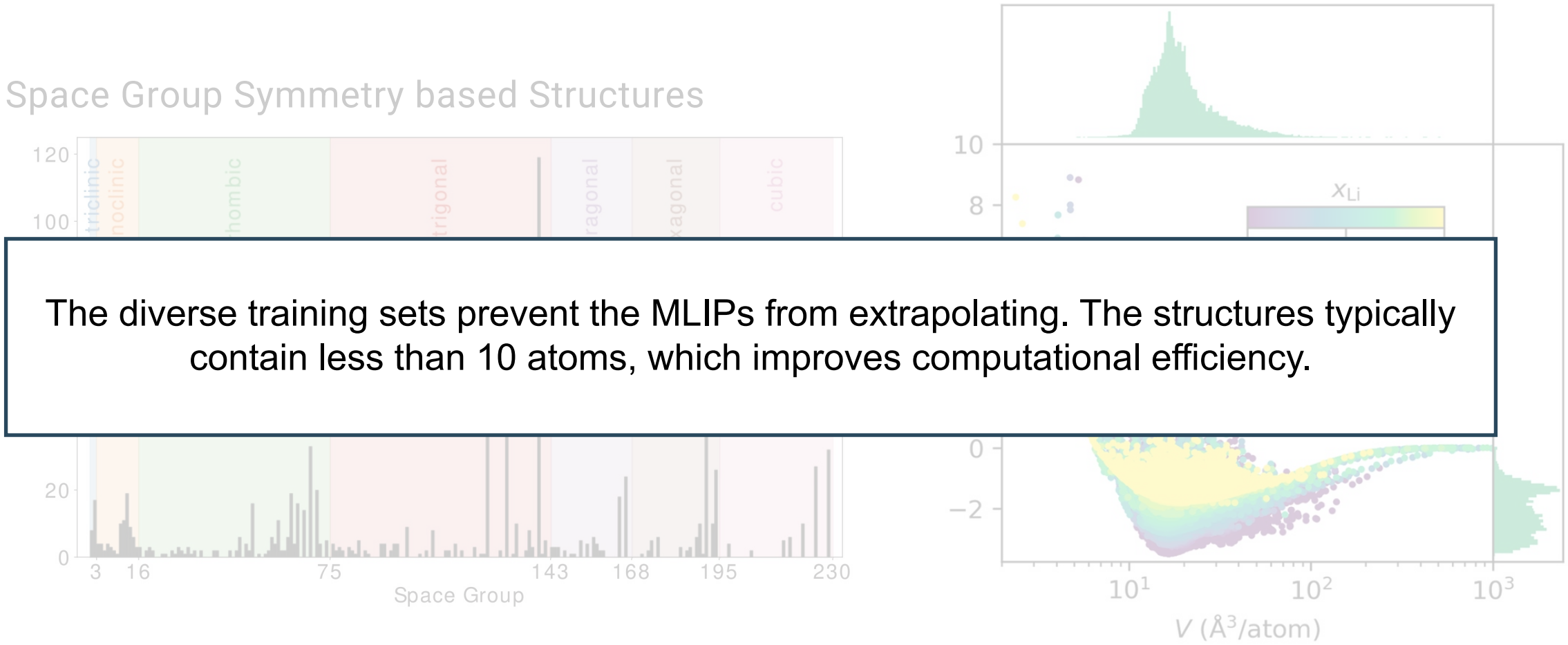
Divers Training Sets

Automated Small SYmmetric Structure Training (ASSYST)

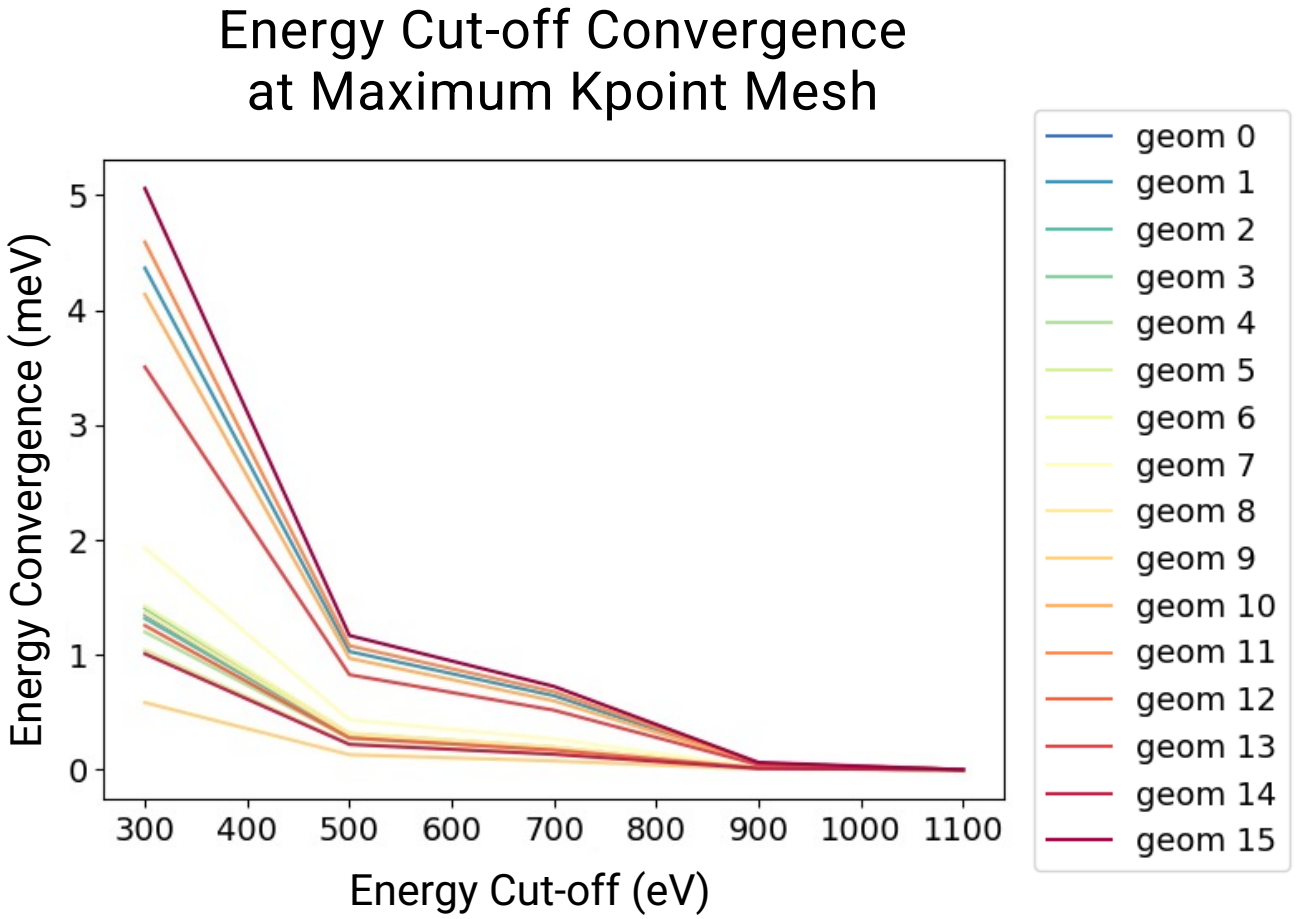


The diverse training sets prevent the MLIPs from extrapolating. The structures typically contain less than 10 atoms, which improves computational efficiency.

M. Poul, L. Huber, E. Bitzek and J. Neugebauer., Phys. Rev. B 107 (2023)

Uncertainty Propagation from Density Functional Theory

Can we leverage computationally more efficient less complete basis sets?



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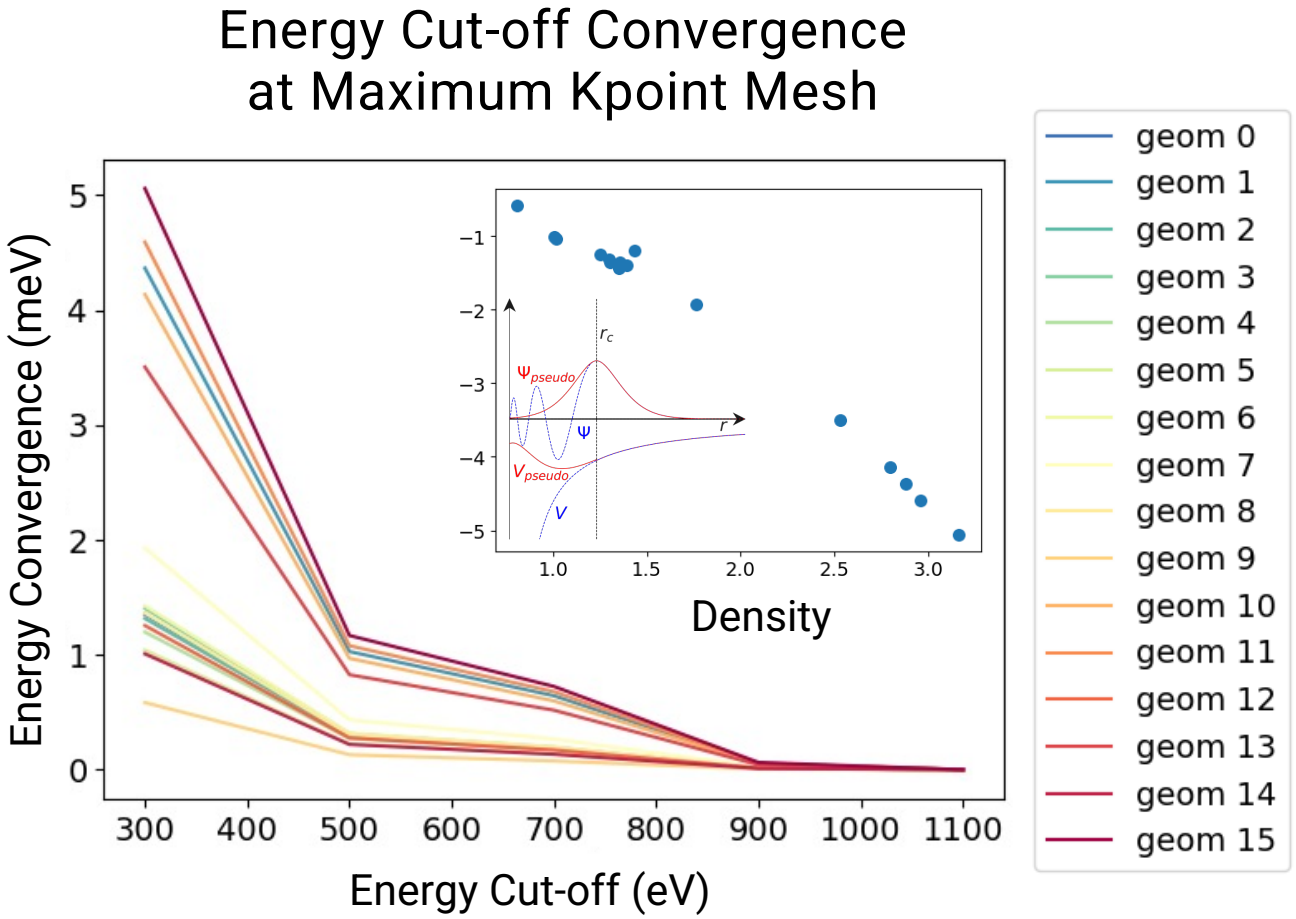
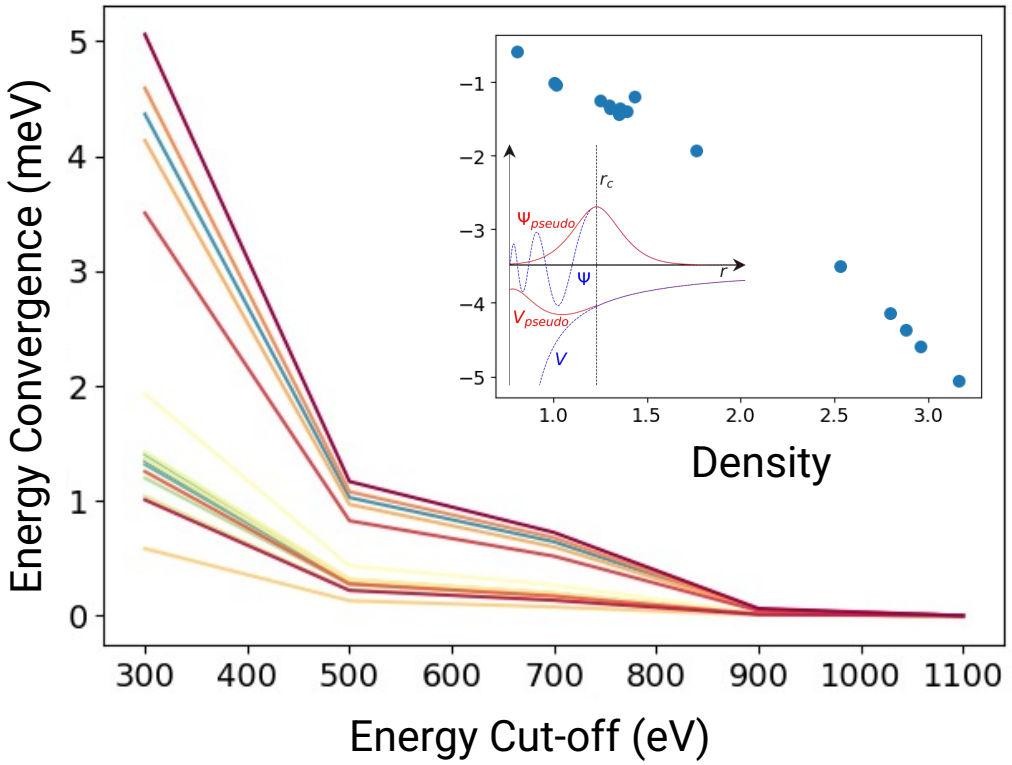


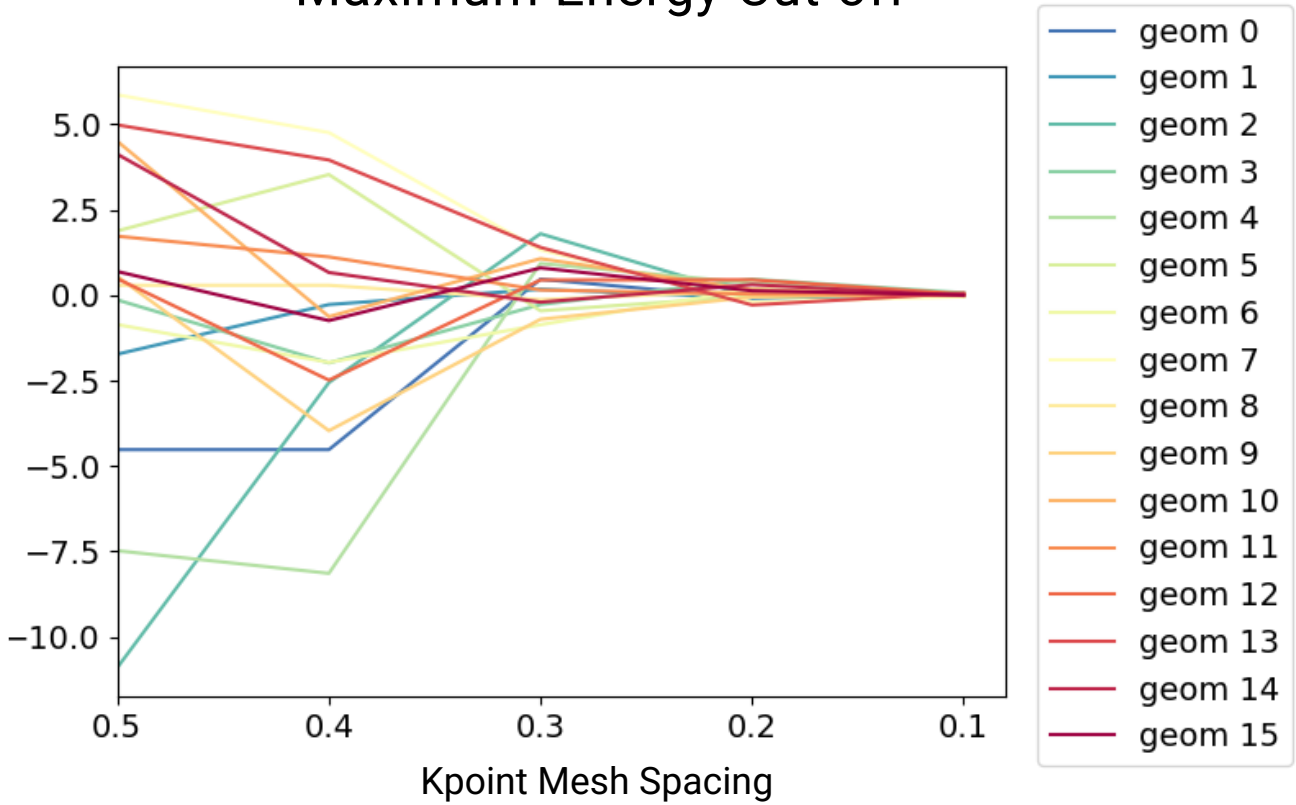
Uncertainty Propagation from Density Functional Theory

Can we leverage computationally more efficient less complete basis sets?

Energy Cut-off Convergence at Maximum Kpoint Mesh



Kpoint Mesh Convergence at Maximum Energy Cut-off



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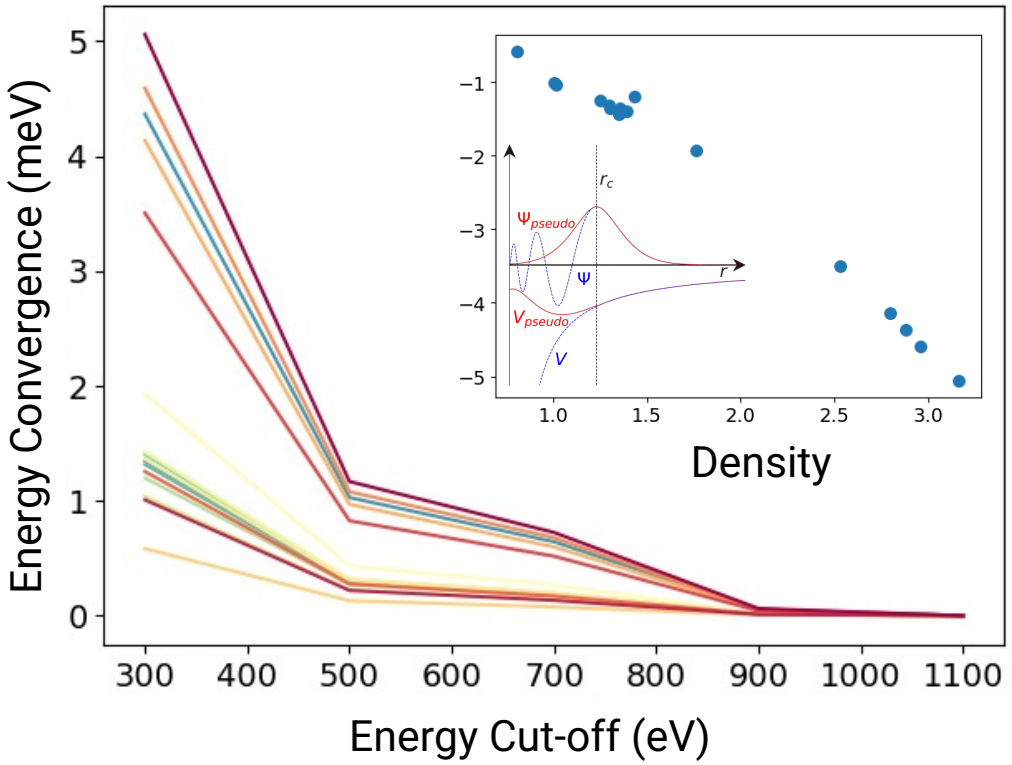

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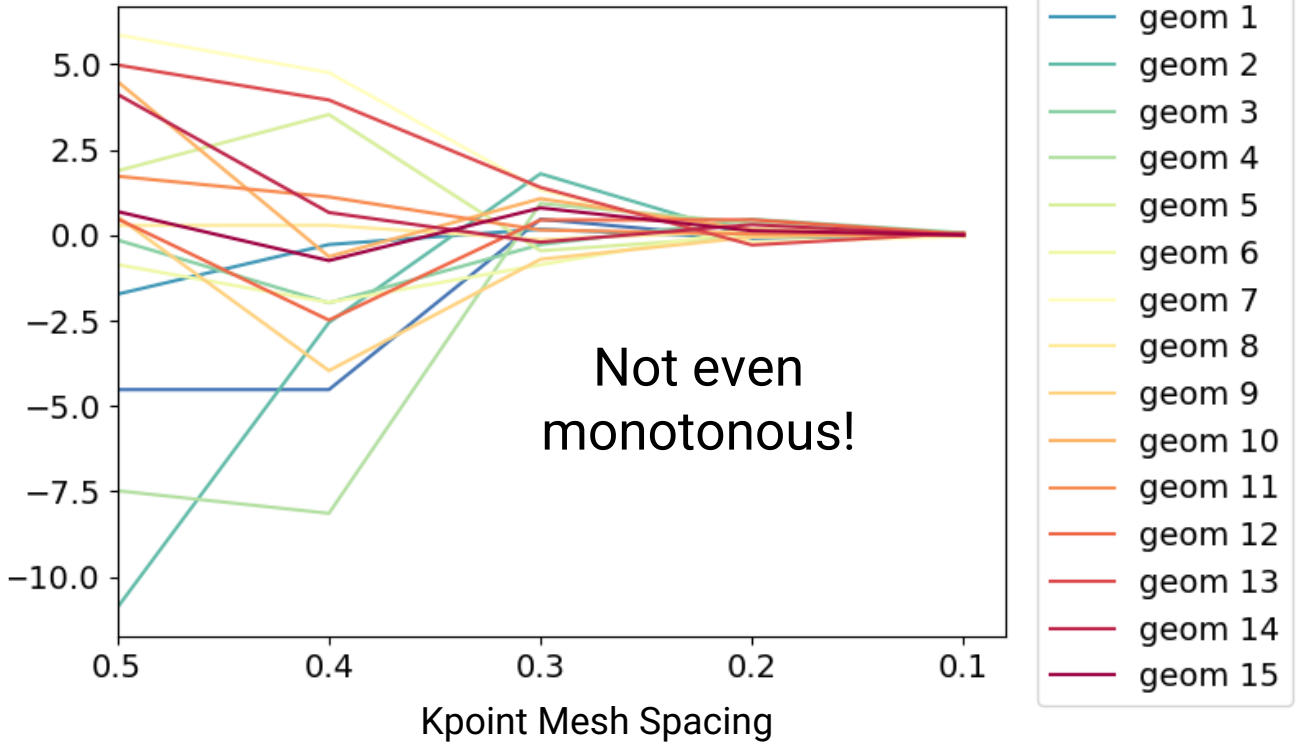

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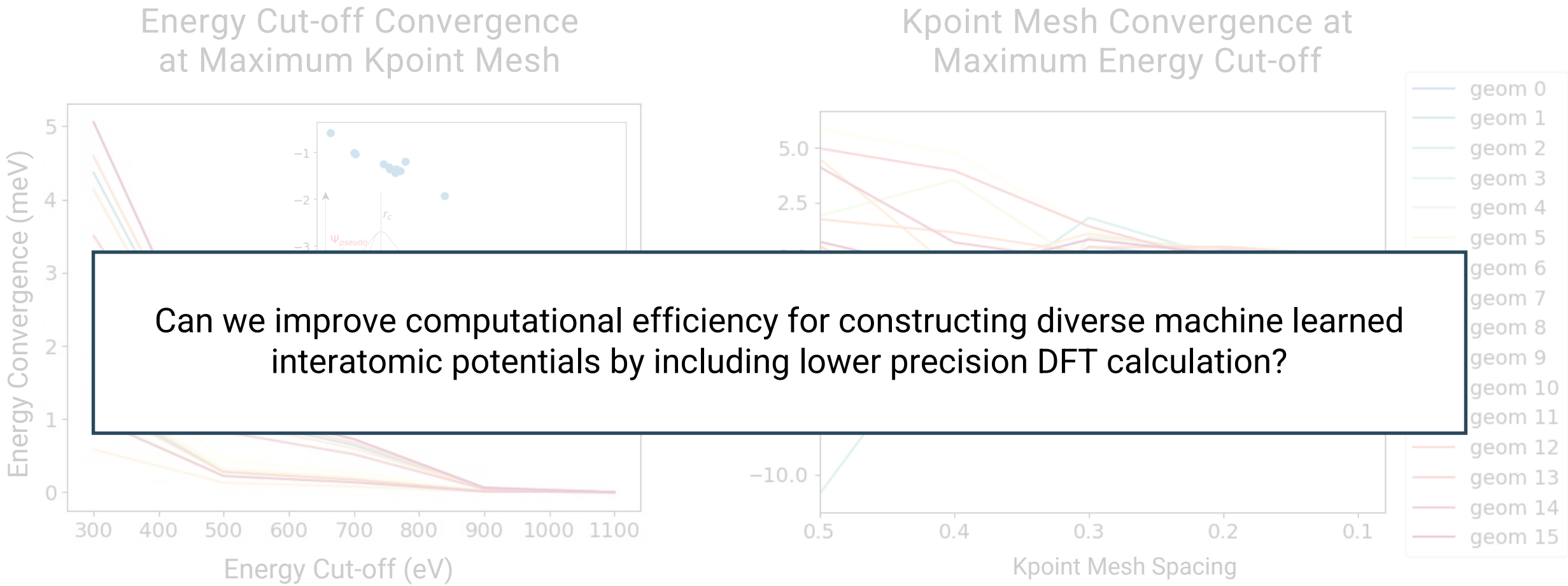


Can we improve computational efficiency for constructing diverse machine learned interatomic potentials by including lower precision DFT calculation?

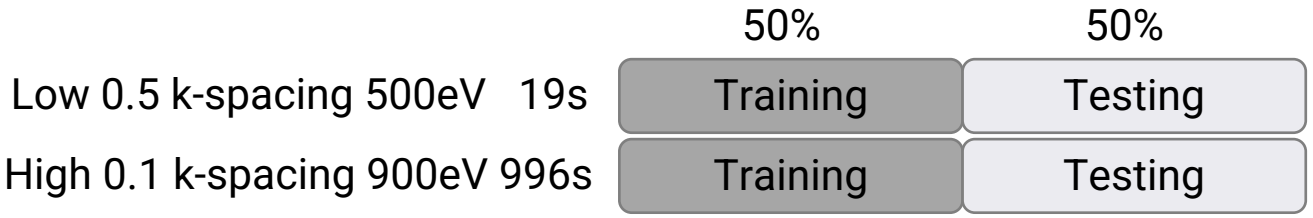
Predict From Low Precision to High Precision

For Fitting a Machine Learned Interatomic Potential

Low 0.5 k-spacing 500eV 19s	20 000 configuration
High 0.1 k-spacing 900eV 996s	20 000 configuration

Predict From Low Precision to High Precision

For Fitting a Machine Learned Interatomic Potential



RMS difference of DFT Energies (meV)	5.60

Predict From Low Precision to High Precision

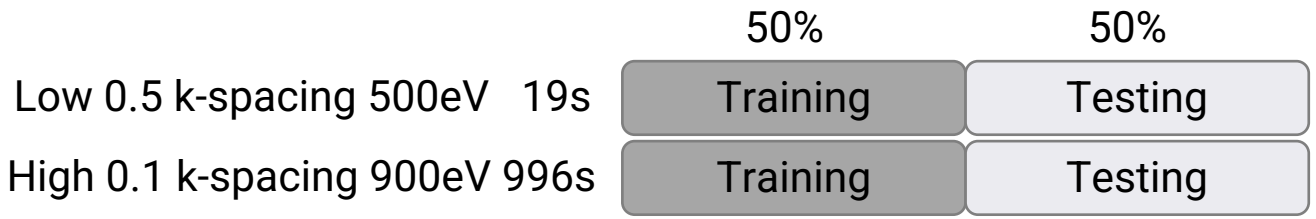
For Fitting a Machine Learned Interatomic Potential

SNAP:

r_{cutoff} = 4.81

$2j_{max}$ = 10

		Train Energy RMSE	Test Energy RMSE	
			low	high
Train on	low	**6.20**	**6.58**	
	high	**4.77**		**5.05**



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	high	**4.77**	7.33	**5.05**

	50%	50%
Low 0.5 k-spacing 500eV 19s	Training	Testing
High 0.1 k-spacing 900eV 996s	Training	Testing

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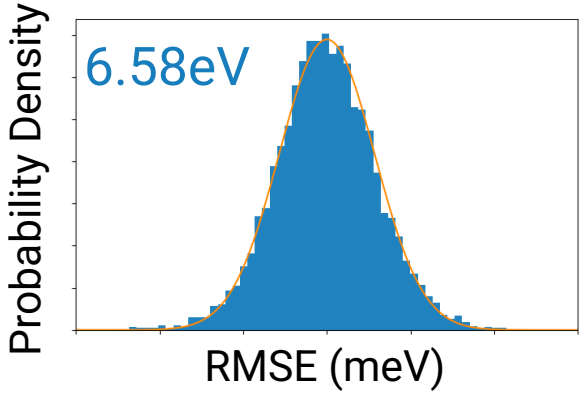
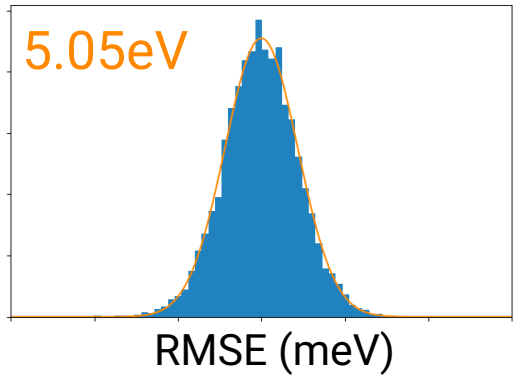
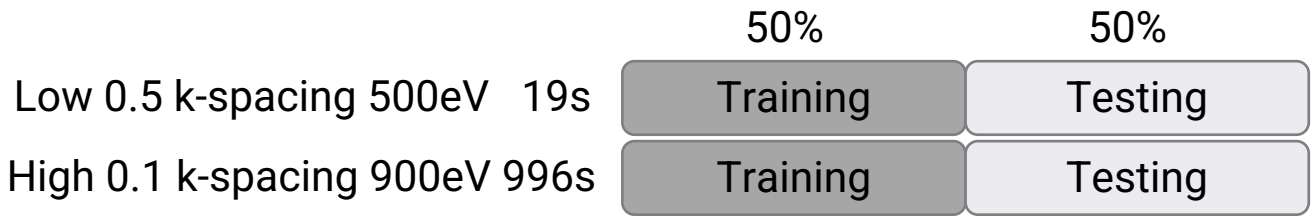
Error on low fitted on low



Error on high fitted on high





RMS difference of DFT Energies (meV)	5.60

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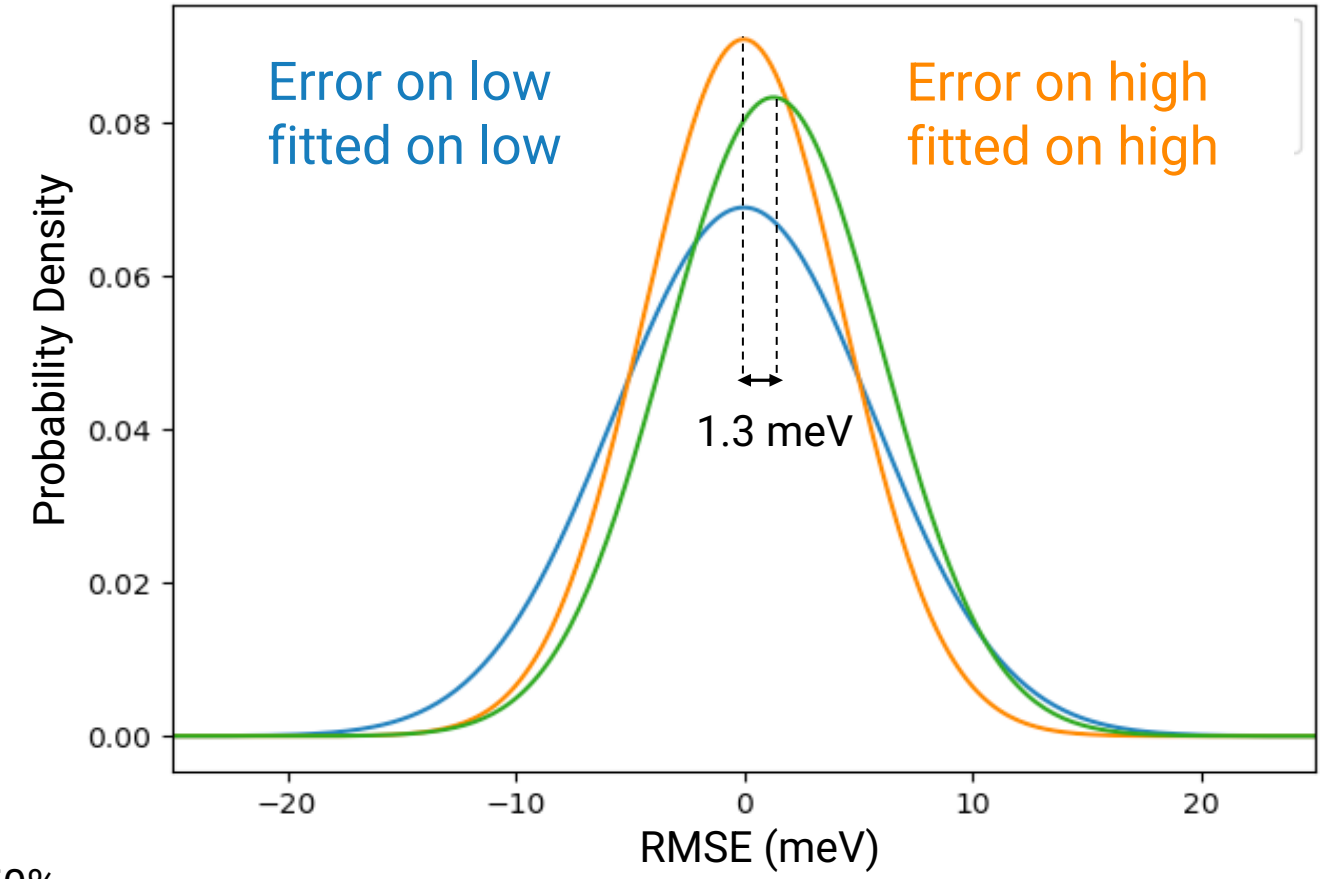

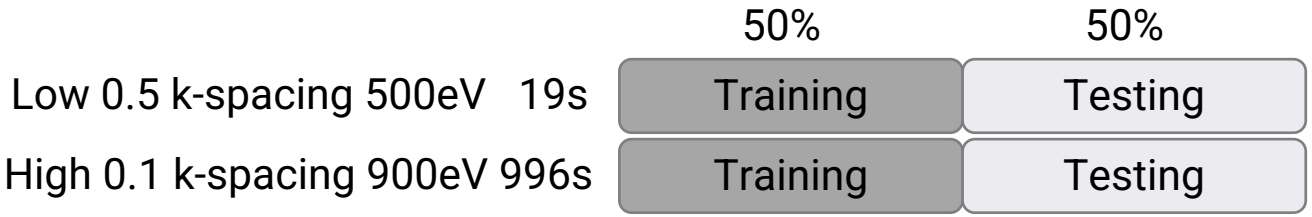

RMS difference of DFT Energies (meV)	5.60

Predict From Low Precision to High Precision

For Fitting a Machine Learned Interatomic Potential

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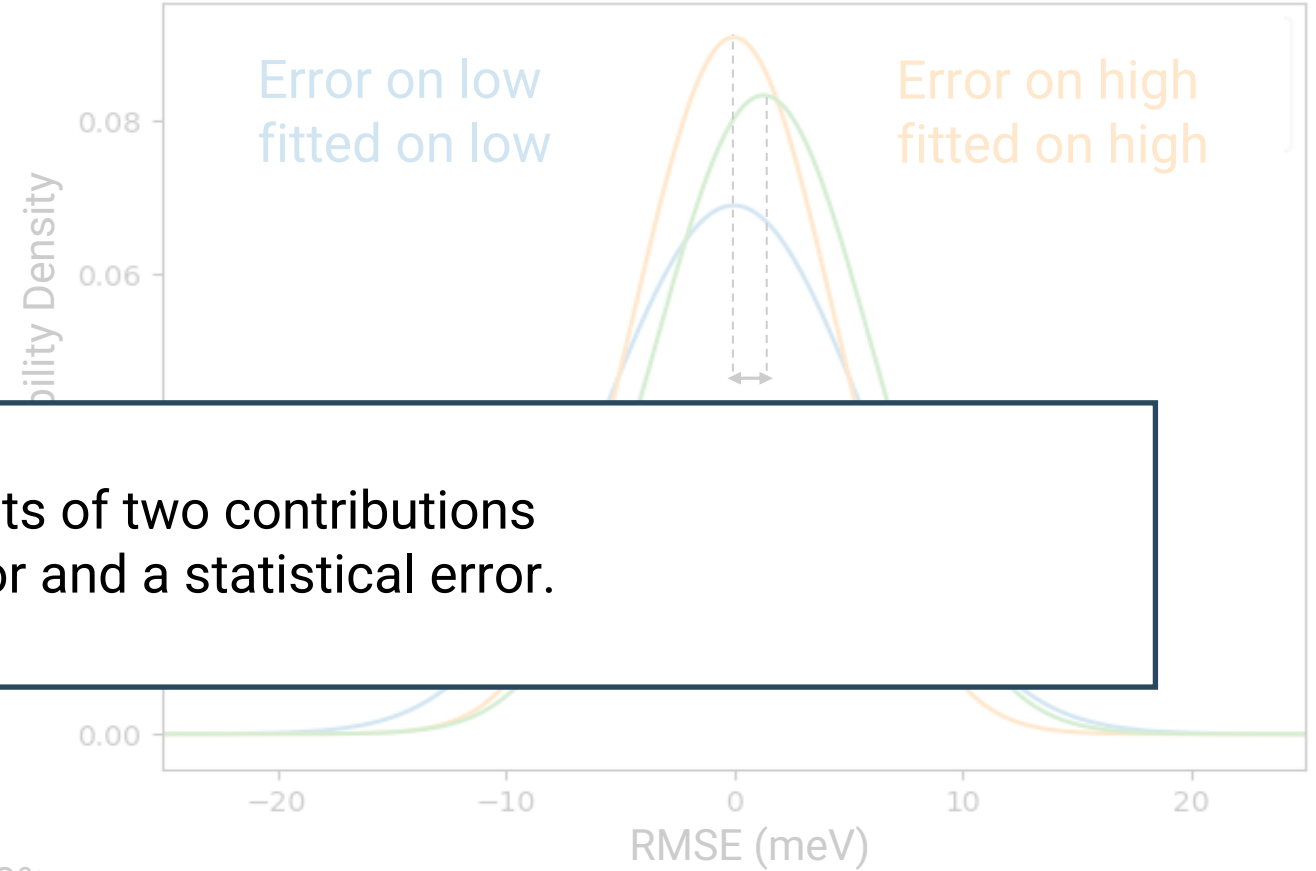


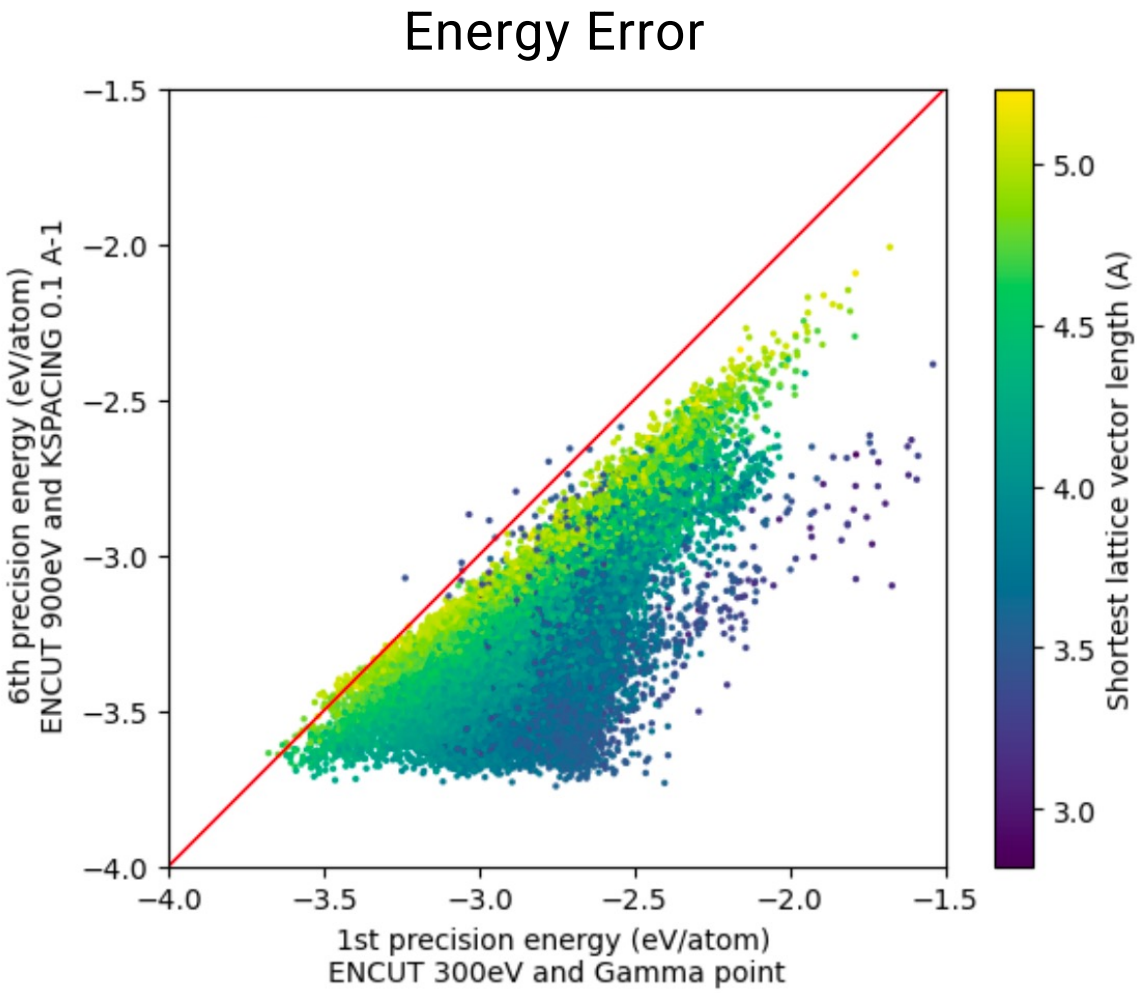
The error consists of two contributions
a systematic error and a statistical error.

	50%	50%
Low 0.5 k-spacing 500eV 19s	Training	Testing
High 0.1 k-spacing 900eV 996s	Training	Testing

RMS difference of DFT Energies (meV)	5.60

DFT Uncertainty in the Training Data

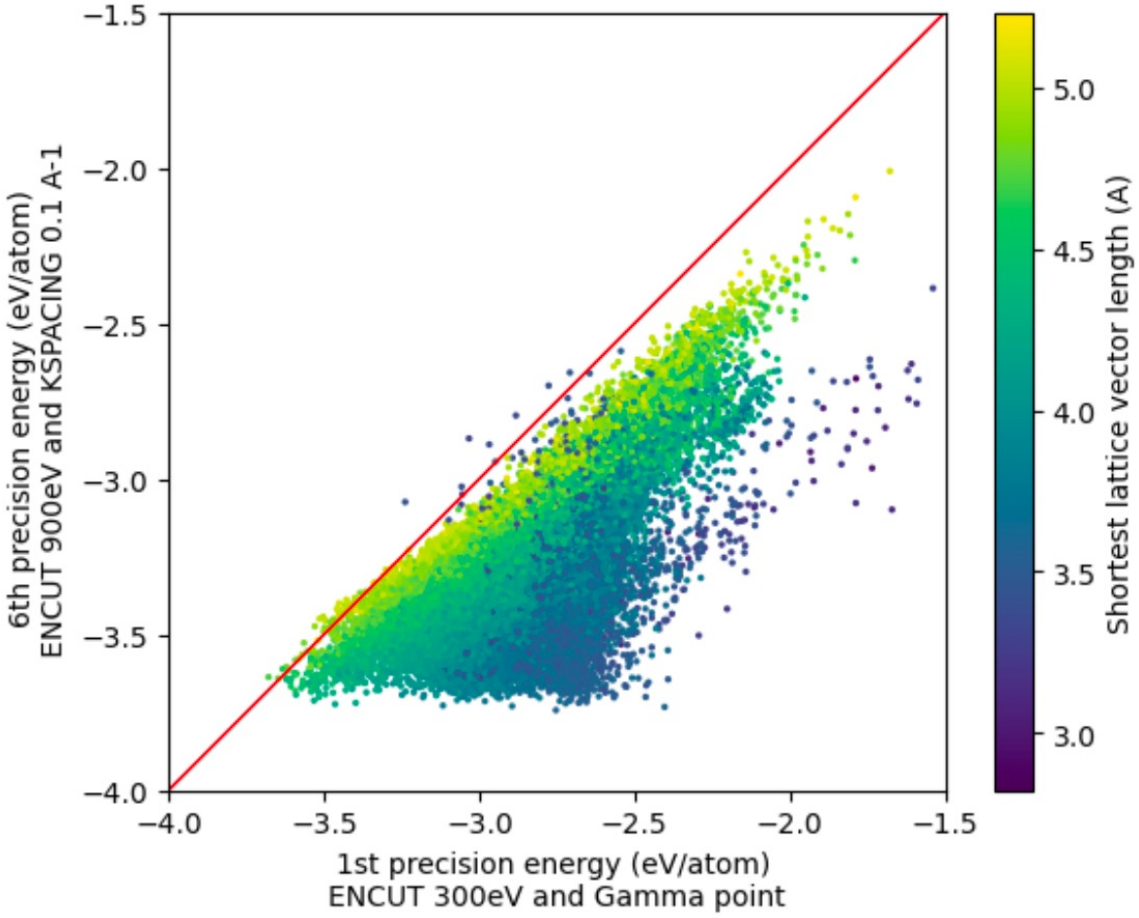
For Energies and Forces



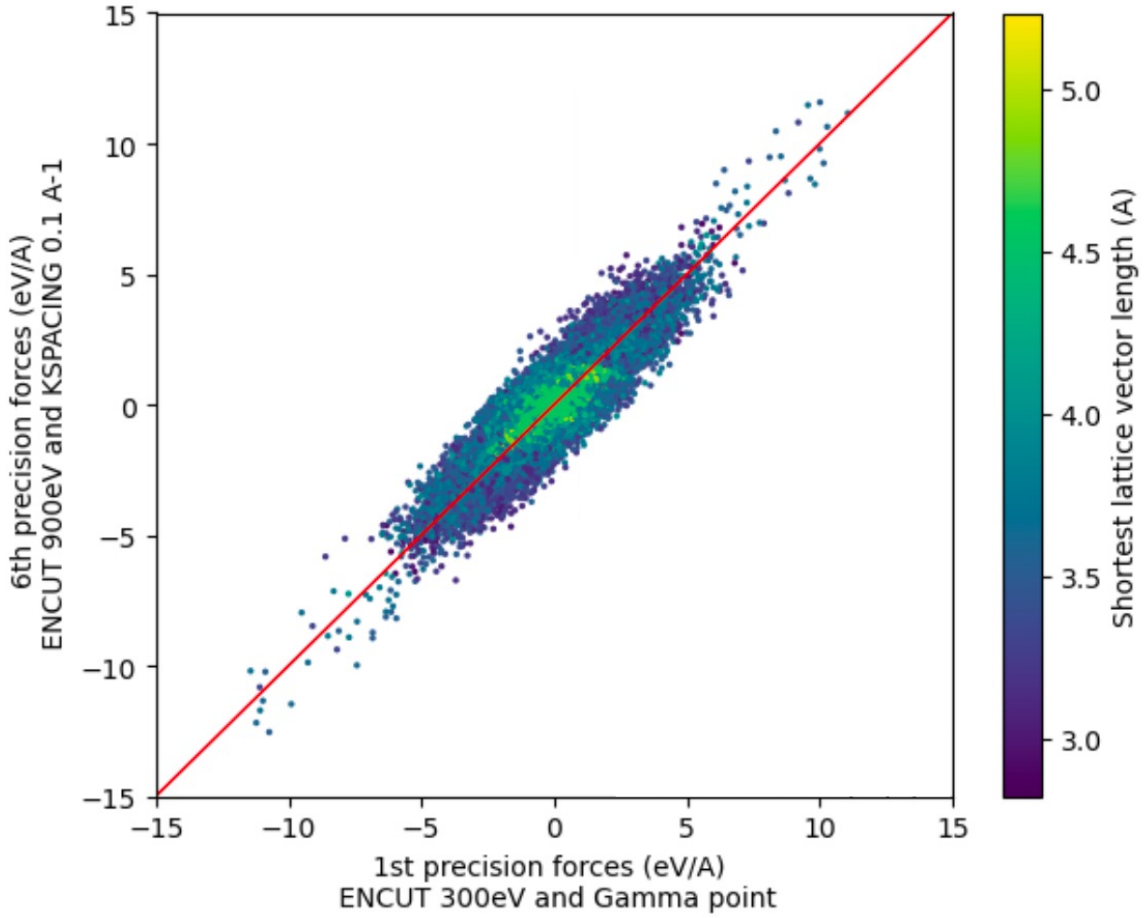
DFT Uncertainty in the Training Data

For Energies and Forces

Energy Error

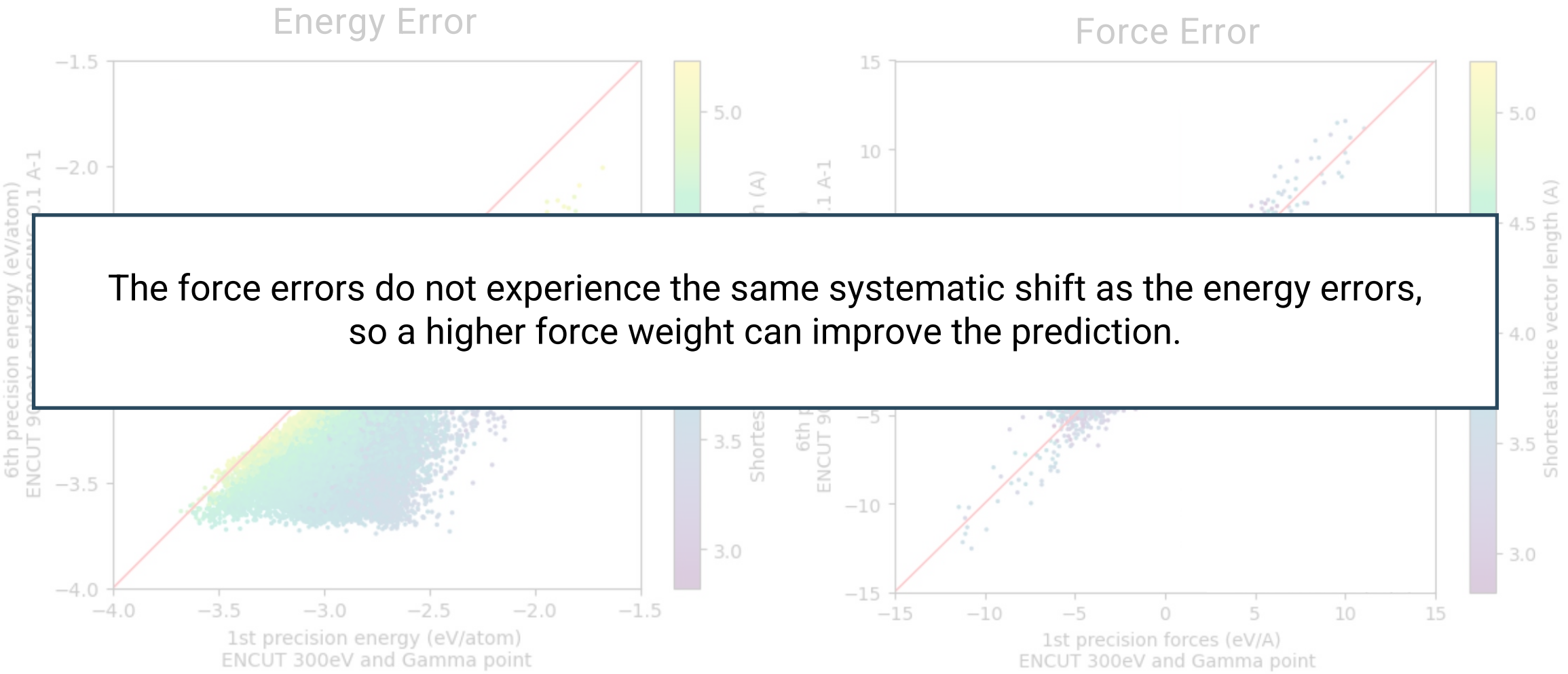


Force Error



DFT Uncertainty in the Training Data

For Energies and Forces

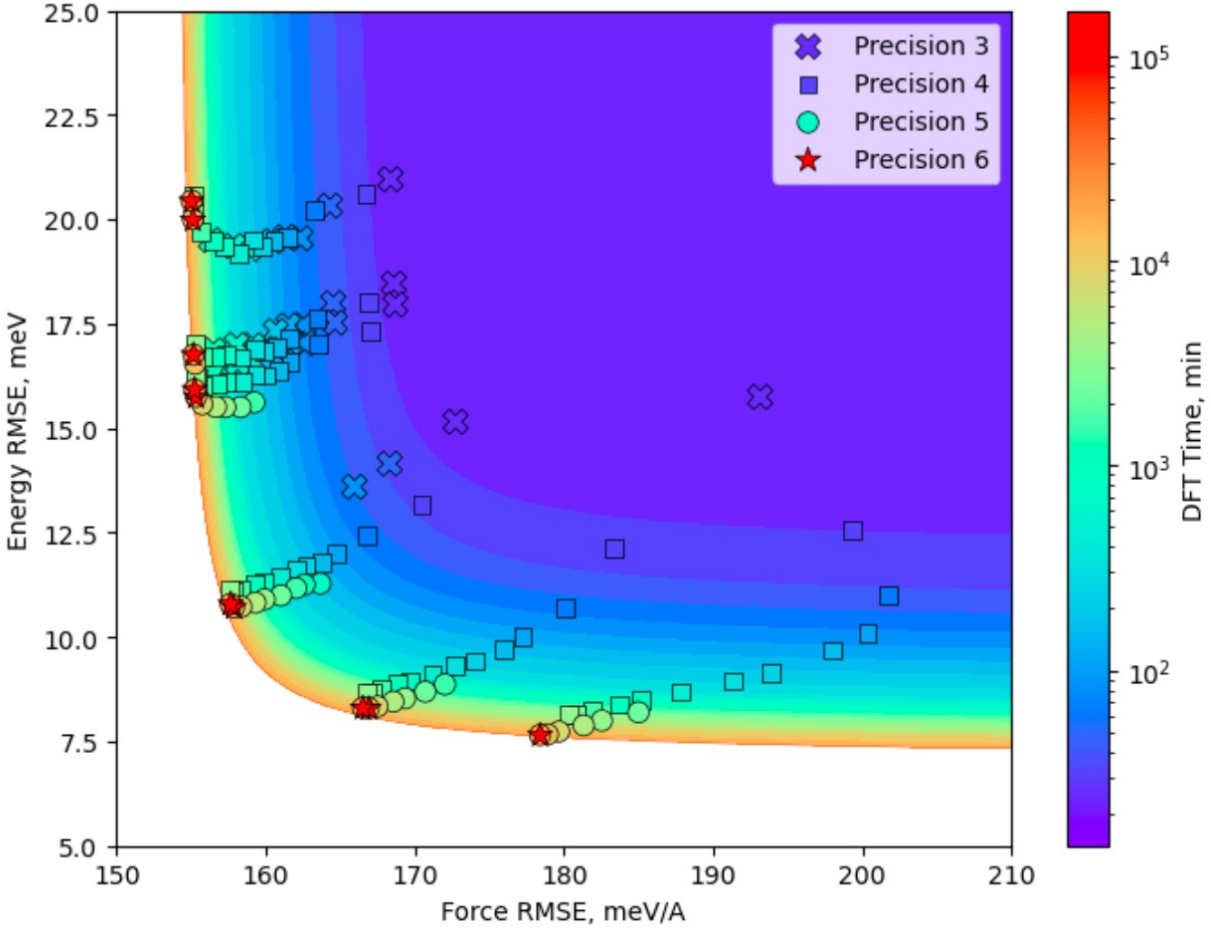


The force errors do not experience the same systematic shift as the energy errors, so a higher force weight can improve the prediction.

Impact of MLIP Complexity

Coupling of MLIP Precision and DFT Precision

Less Complex MLIP (2JMax = 6)



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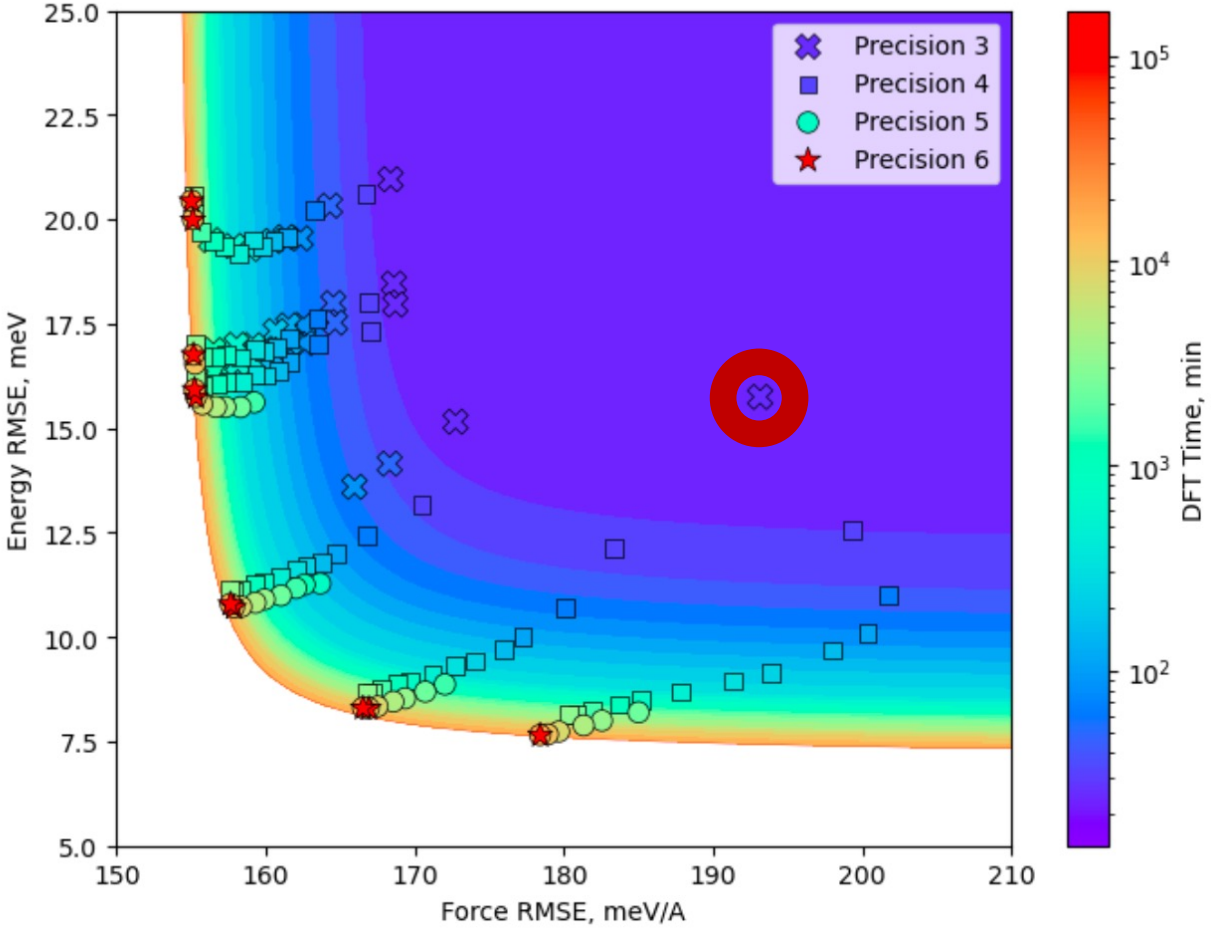

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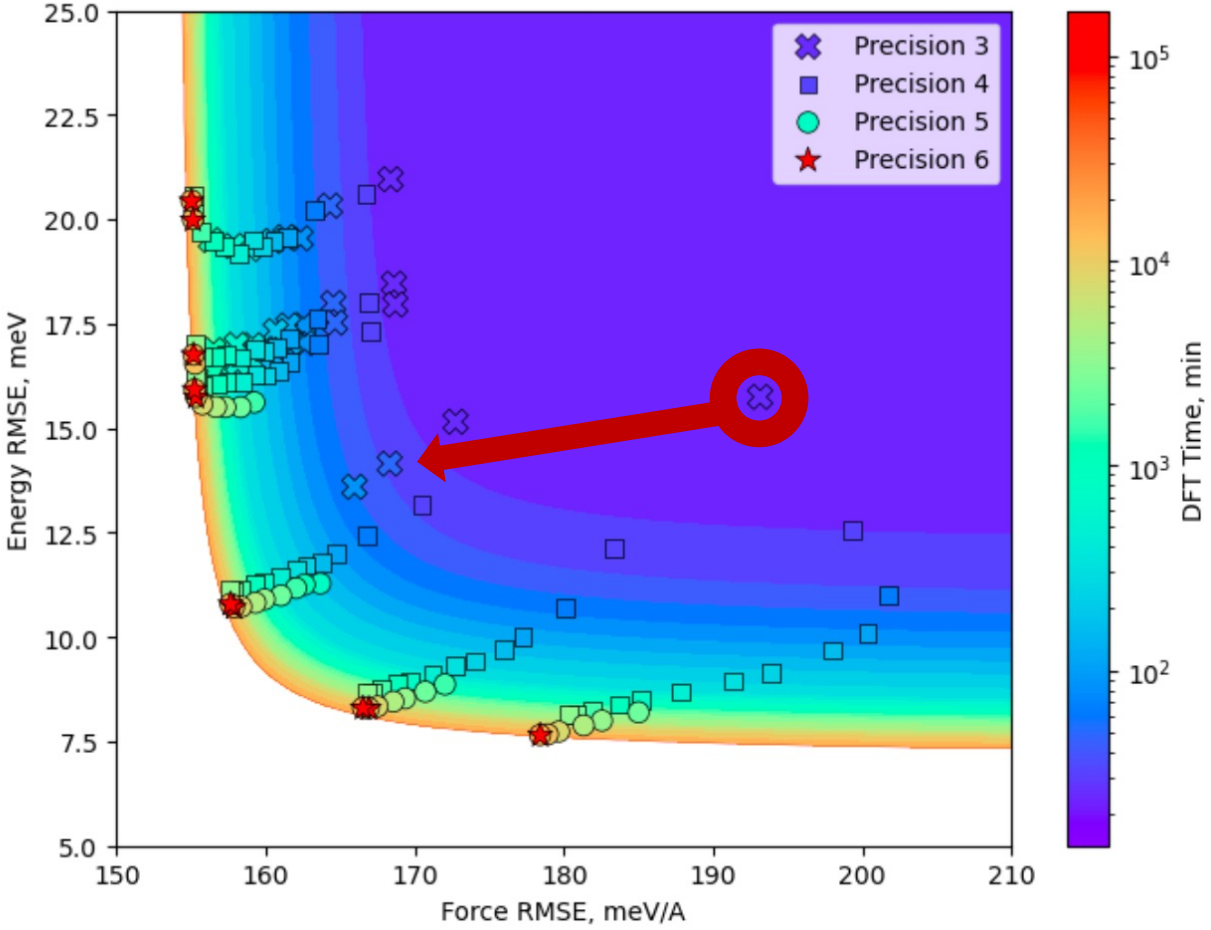

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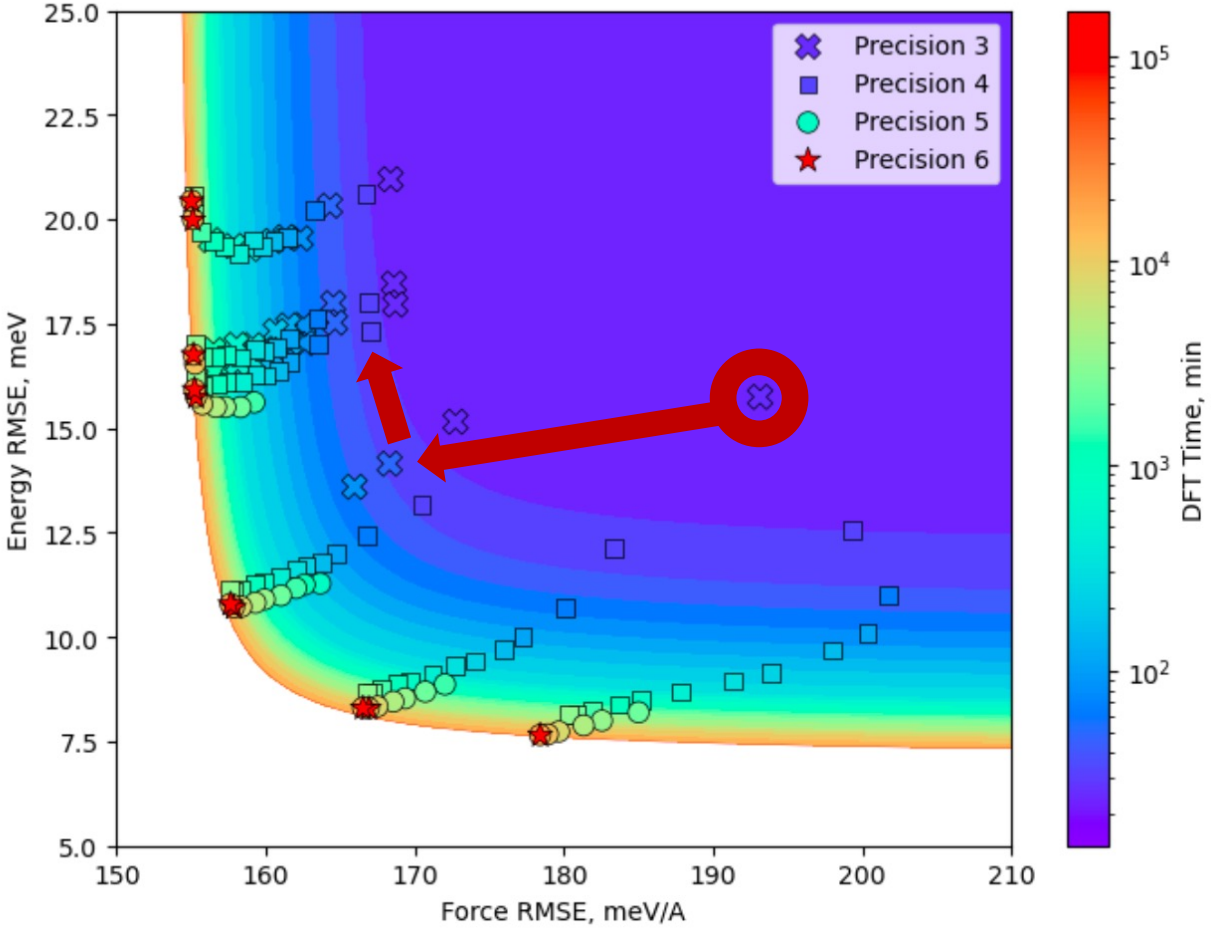

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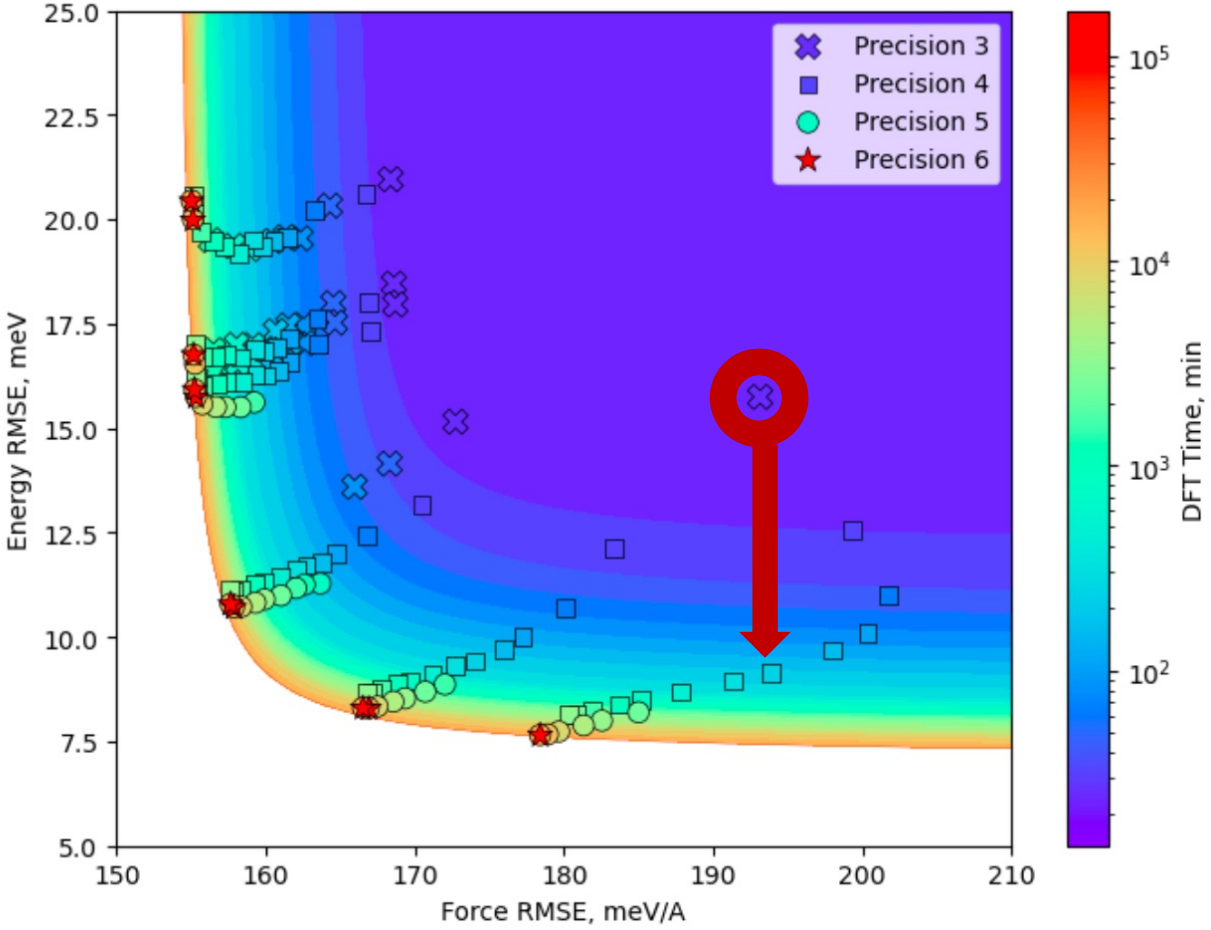

Impact of MLIP Complexity

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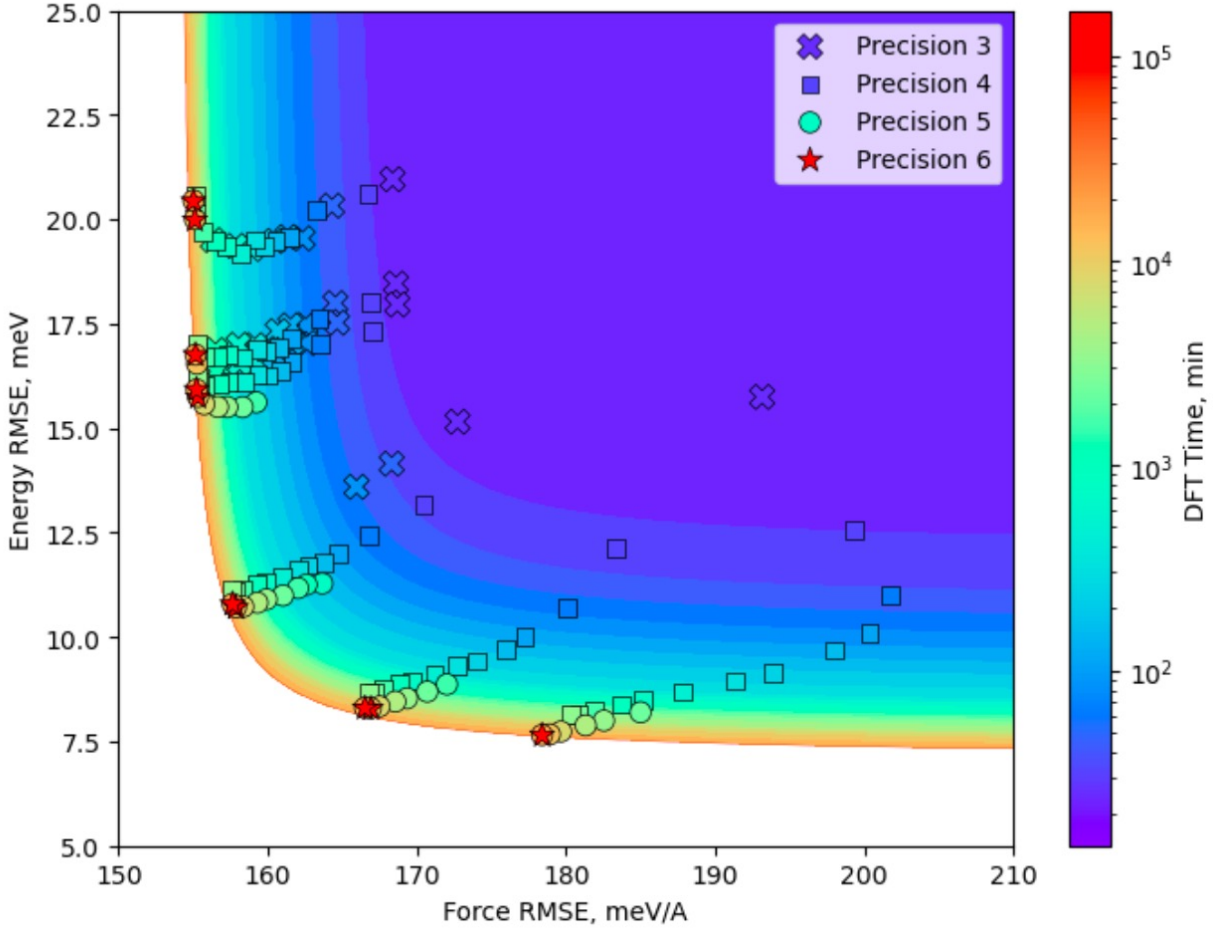
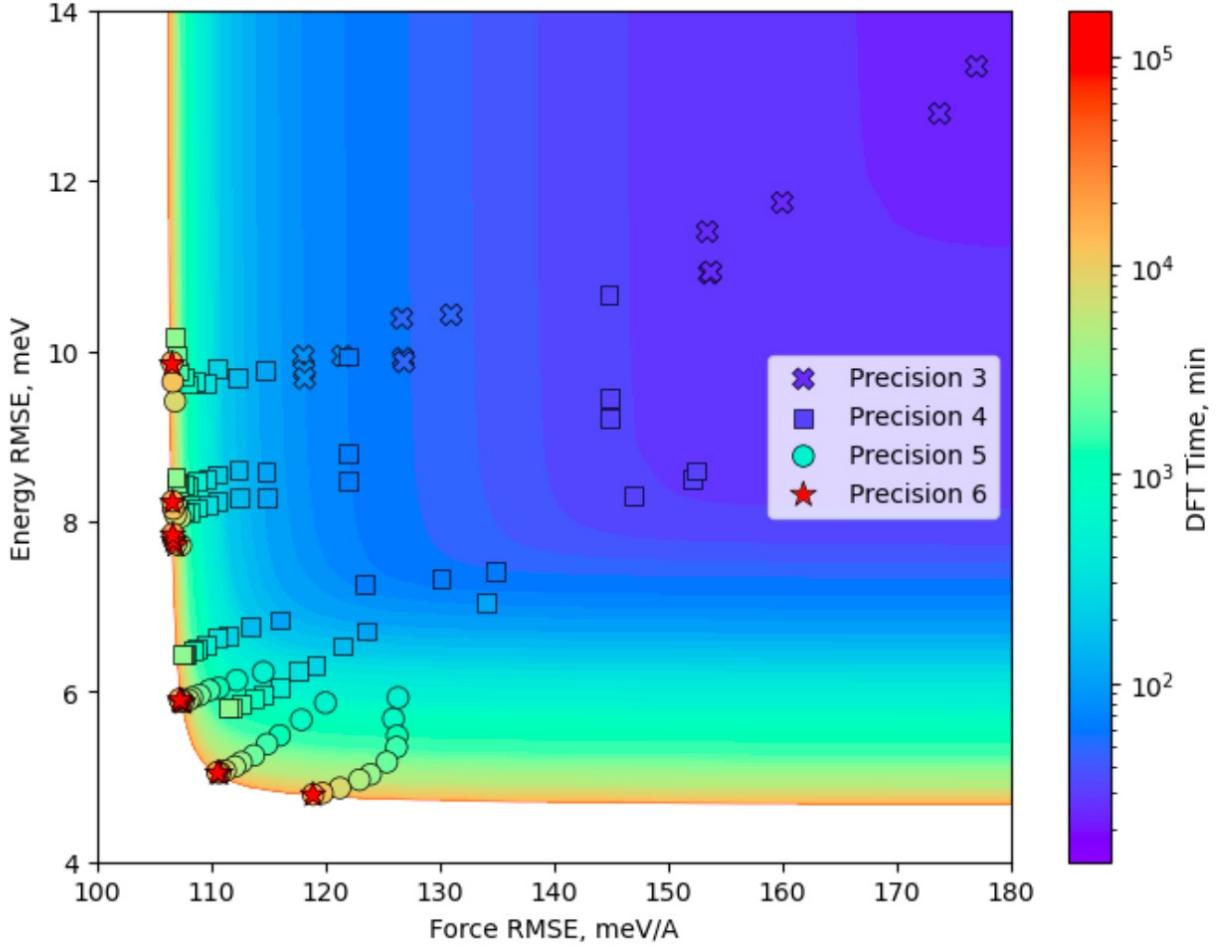
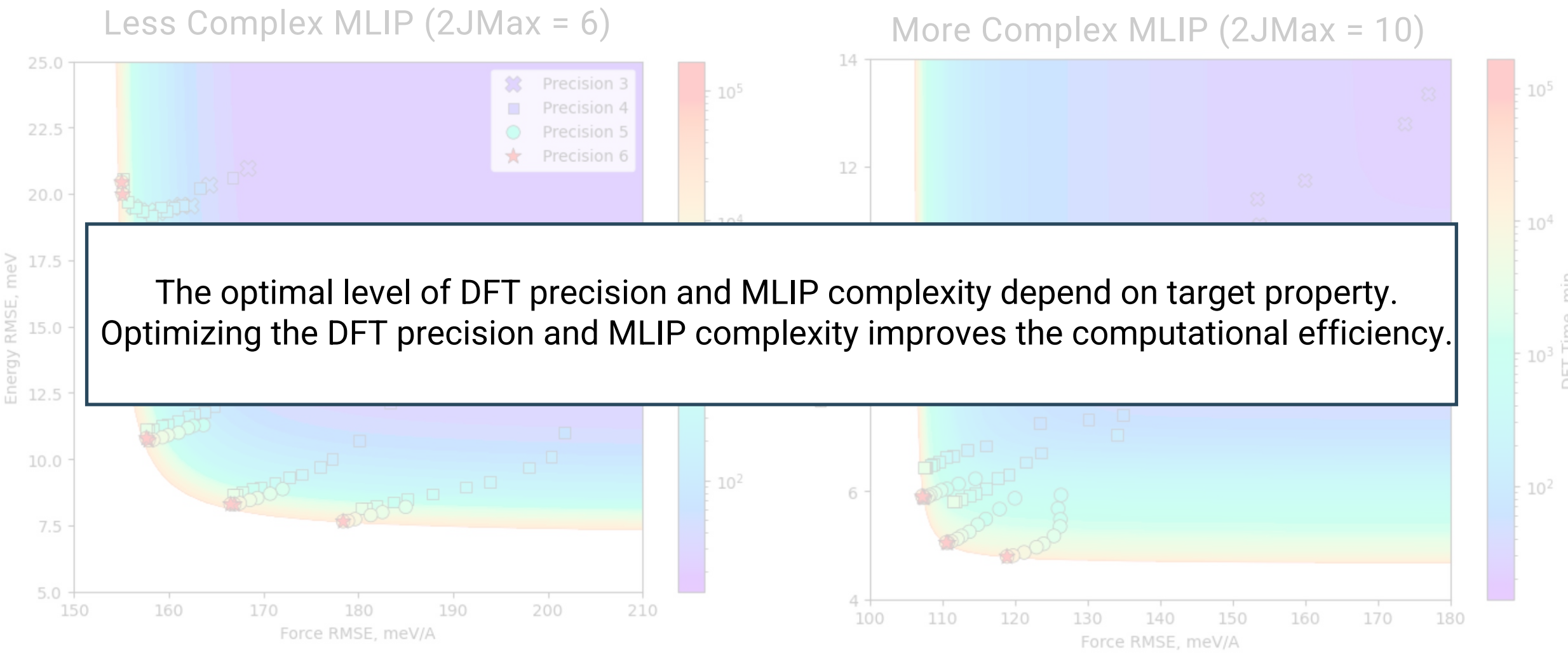


More Complex MLIP (2JMax = 10)



Impact of MLIP Complexity

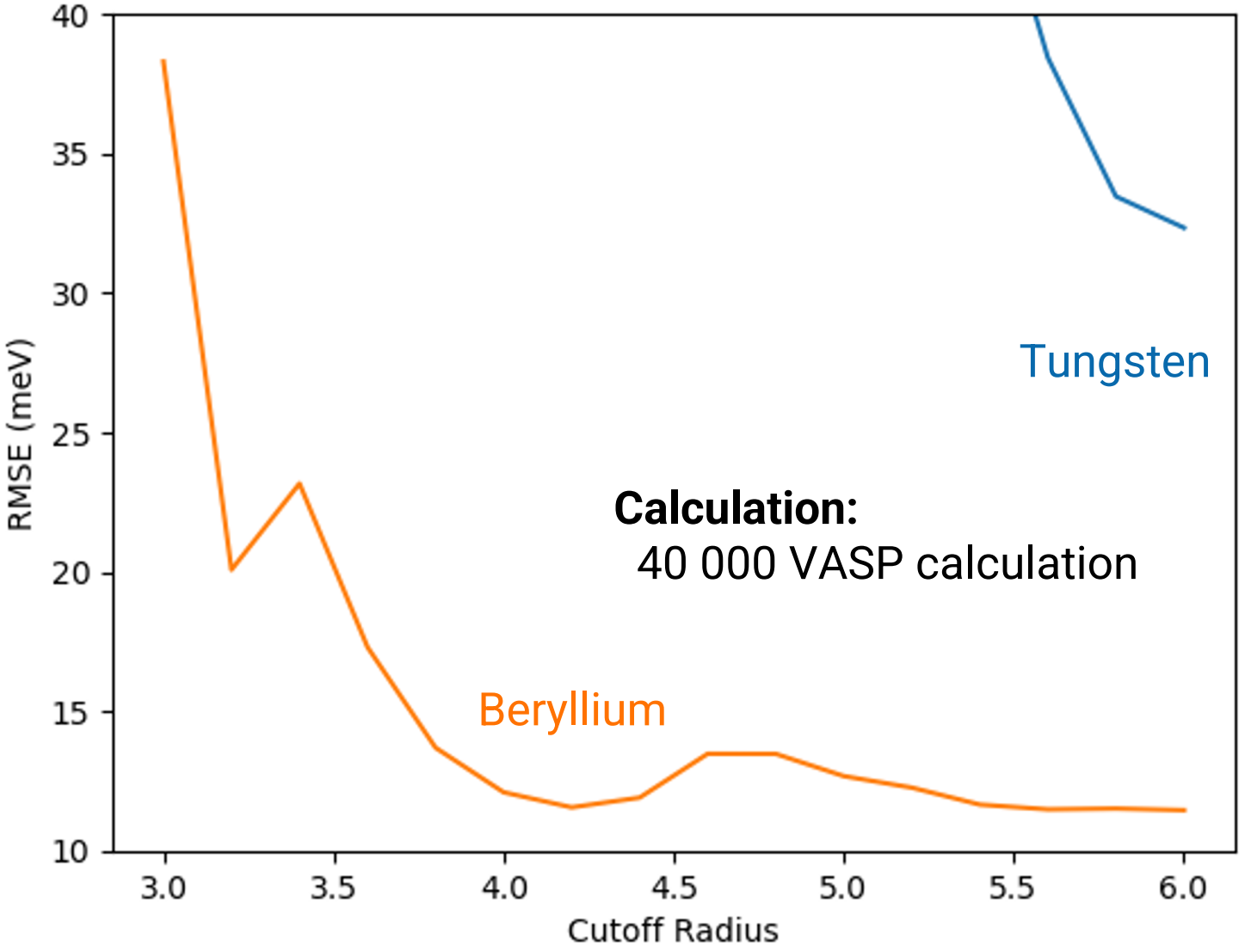
Coupling of MLIP Precision and DFT Precision



The optimal level of DFT precision and MLIP complexity depend on target property.
Optimizing the DFT precision and MLIP complexity improves the computational efficiency.

Compare Chemical Elements

Beryllium and Tungsten

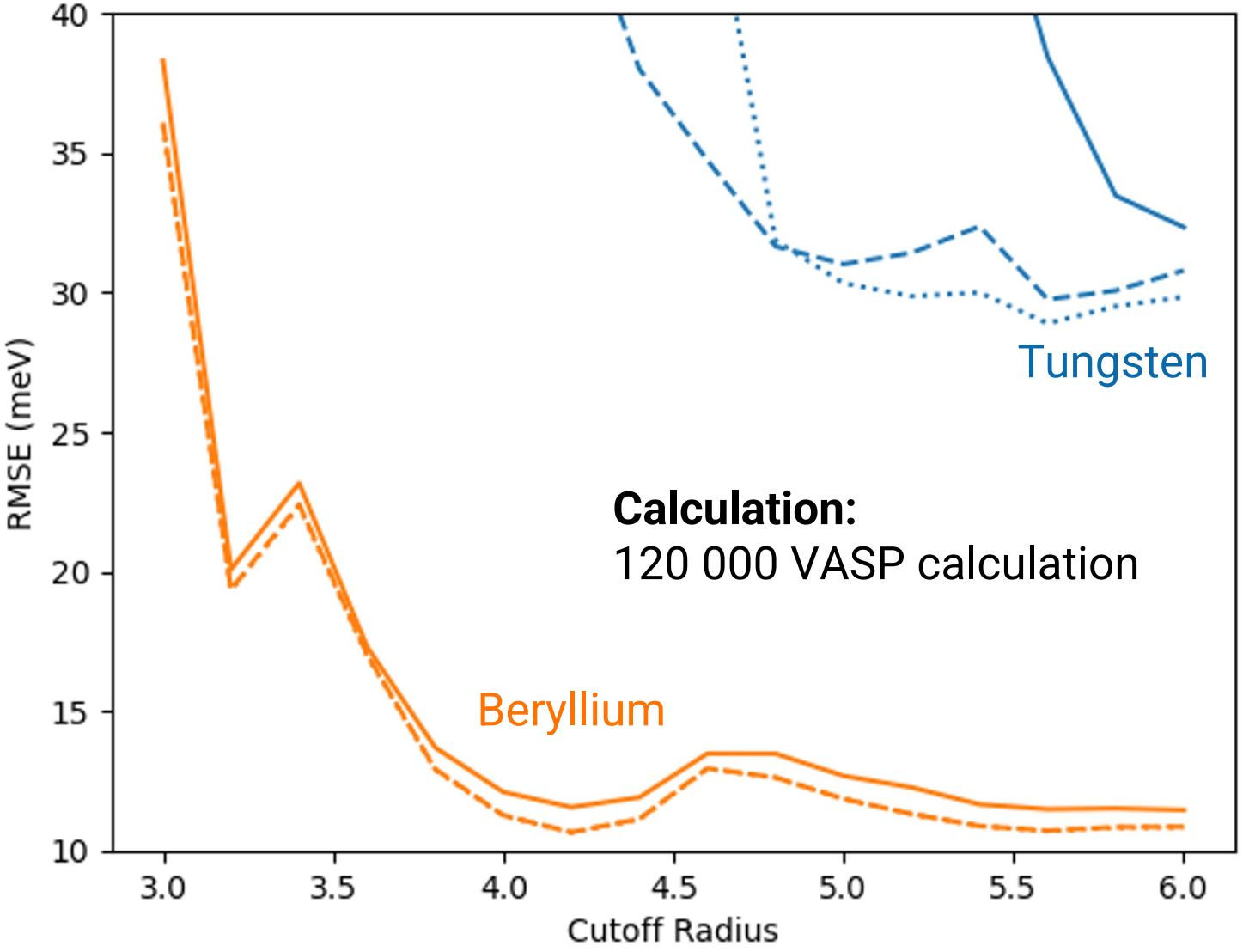


Calculation:
40 000 VASP calculation

DFT precision:
solid: 0.5 kspacing 500 eV encut

Compare Chemical Elements

Beryllium and Tungsten



DFT precision:
solid: 0.5 kspacing 500 eV encut
dashed: 0.25 kspacing 700 eV encut
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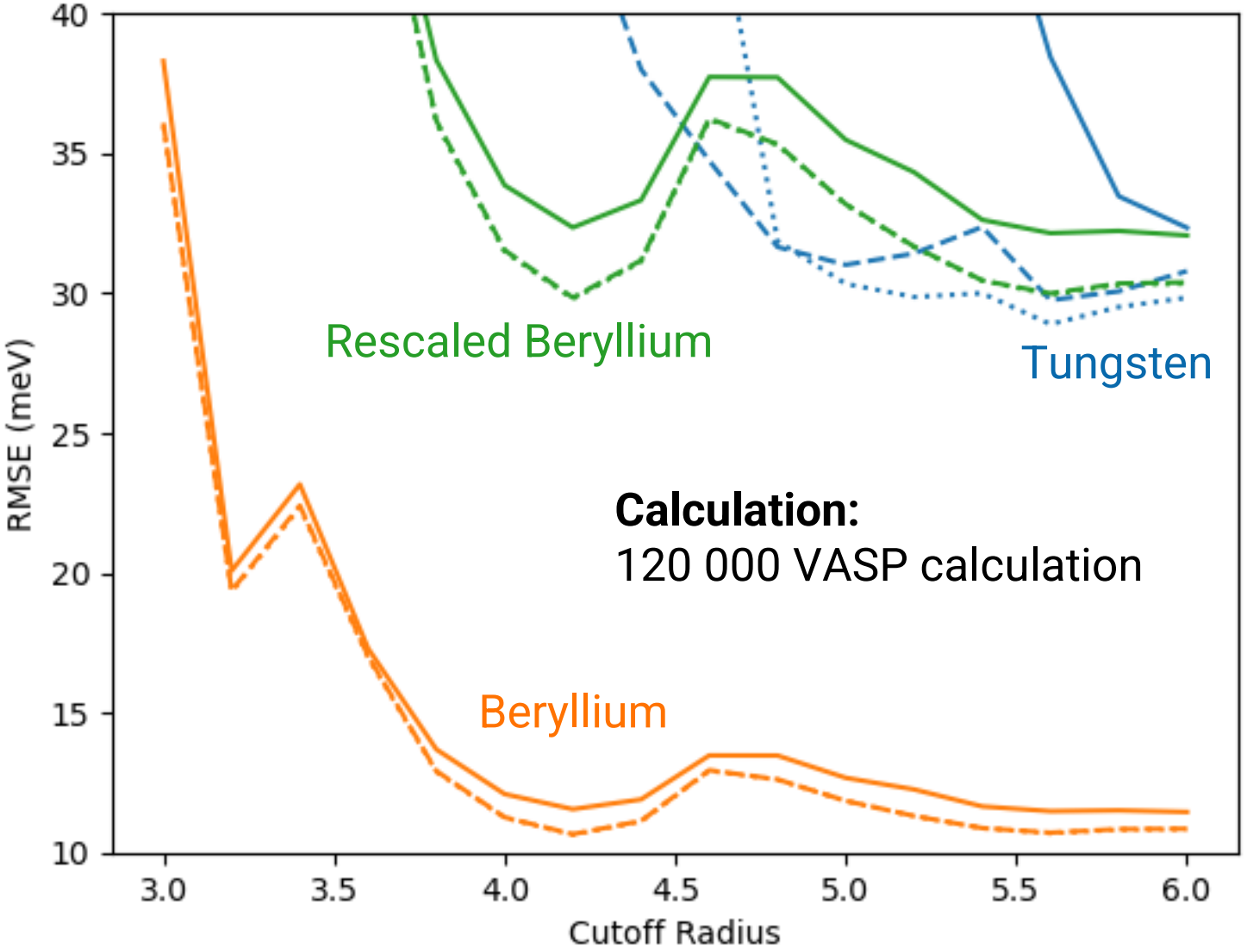
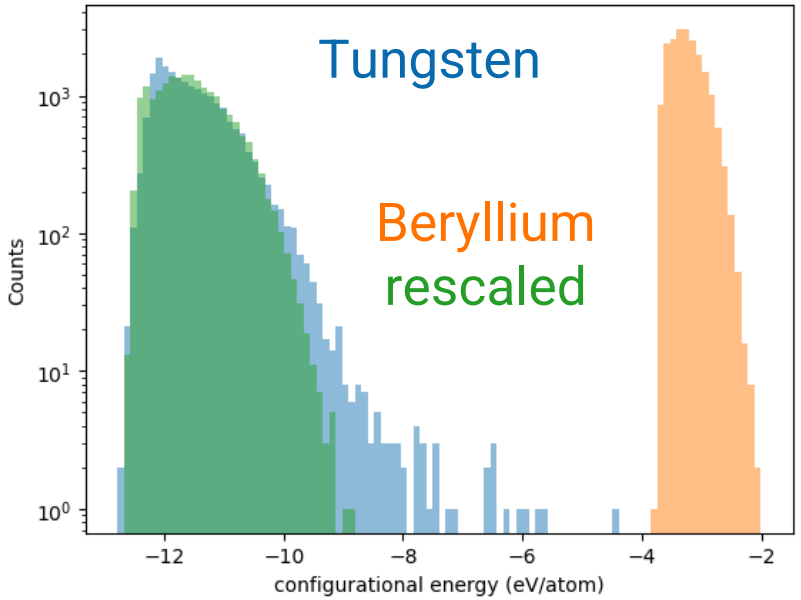
Compare Chemical Elements

Beryllium and Tungsten



Calculation:
120 000 VASP calculation



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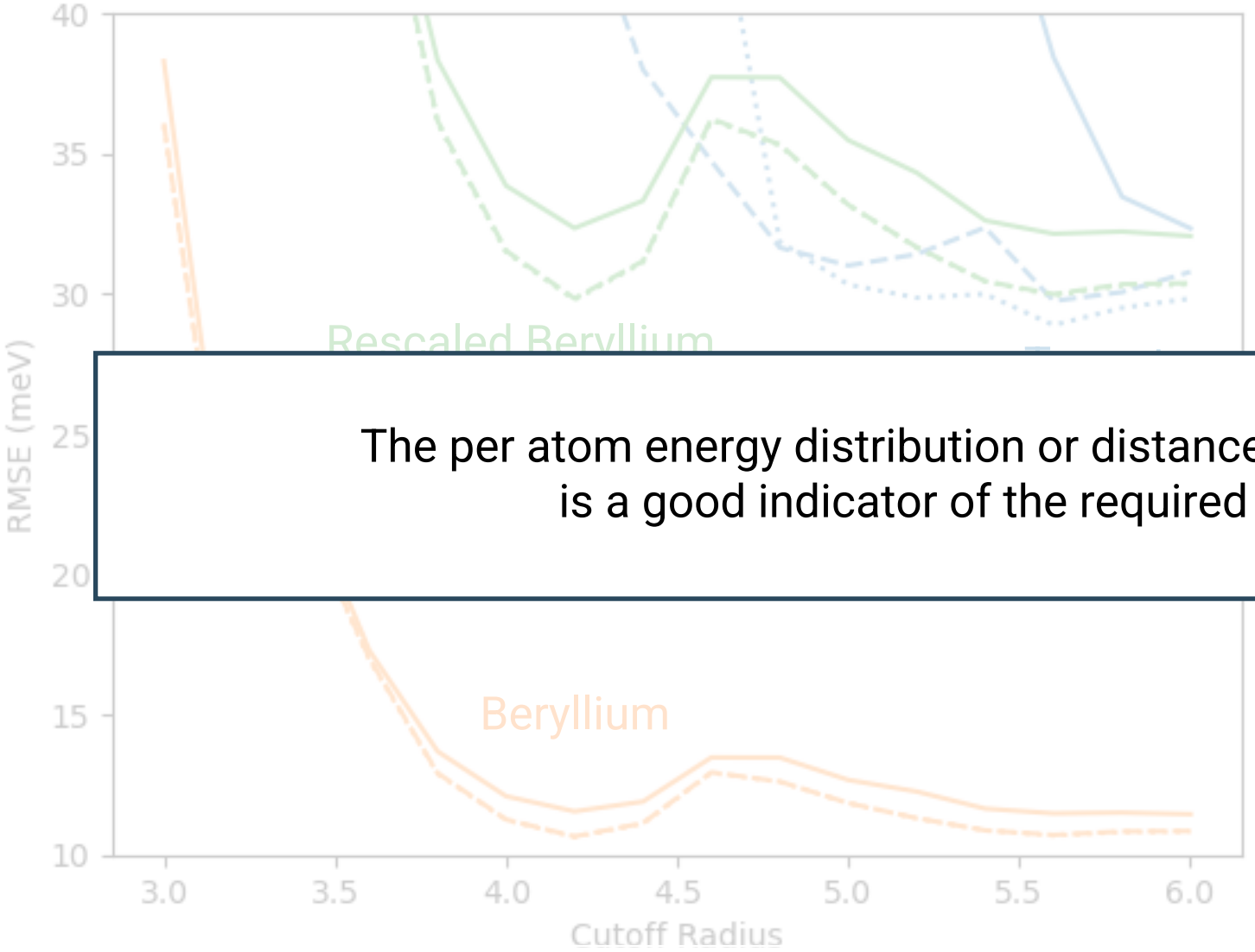
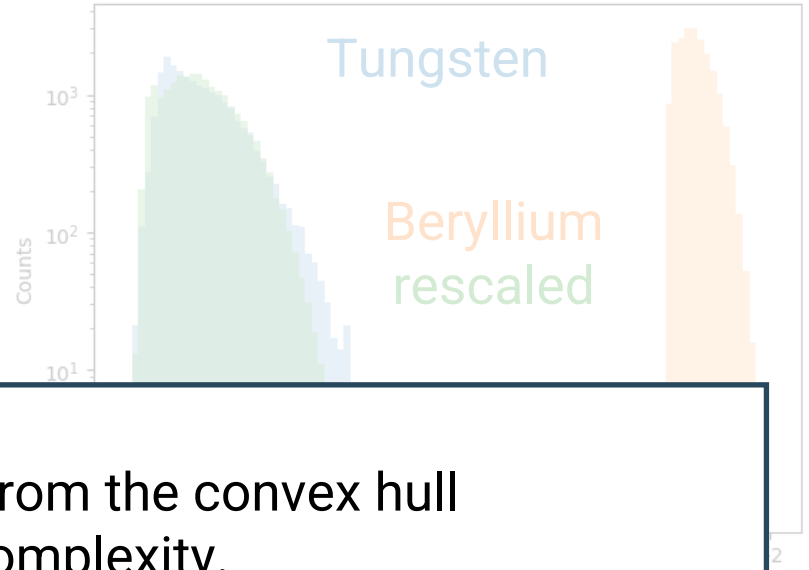


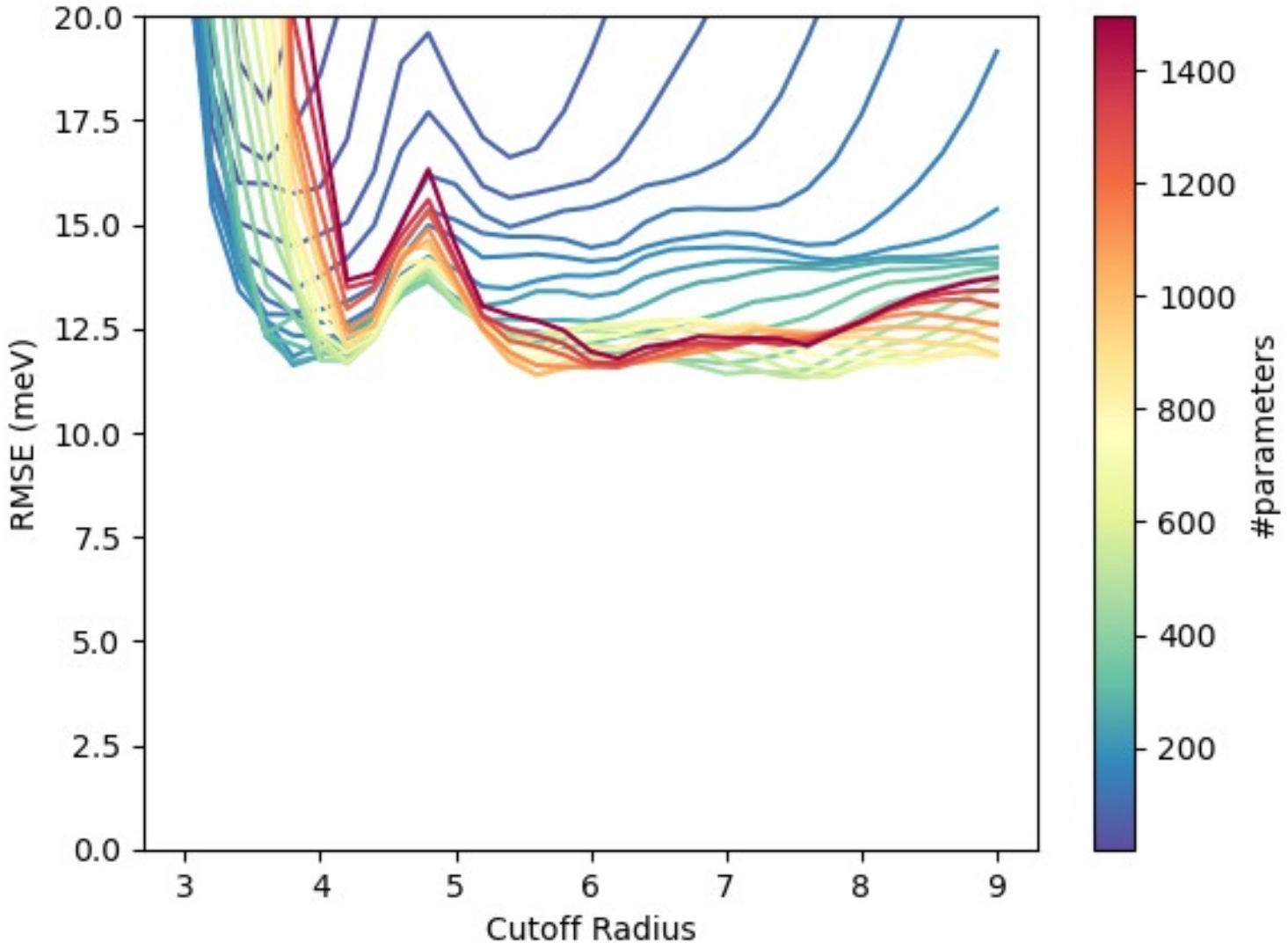


The per atom energy distribution or distance from the convex hull is a good indicator of the required complexity.

DFT precision:
solid: 0.5 kspacing 500 eV encut
dashed: 0.25 kspacing 700 eV encut
dotted: 0.1 kspacing 900 eV encut

Optimal Cut-off Radius

For Spectral Neighbour Analysis Potential (SNAP)



Calculation:

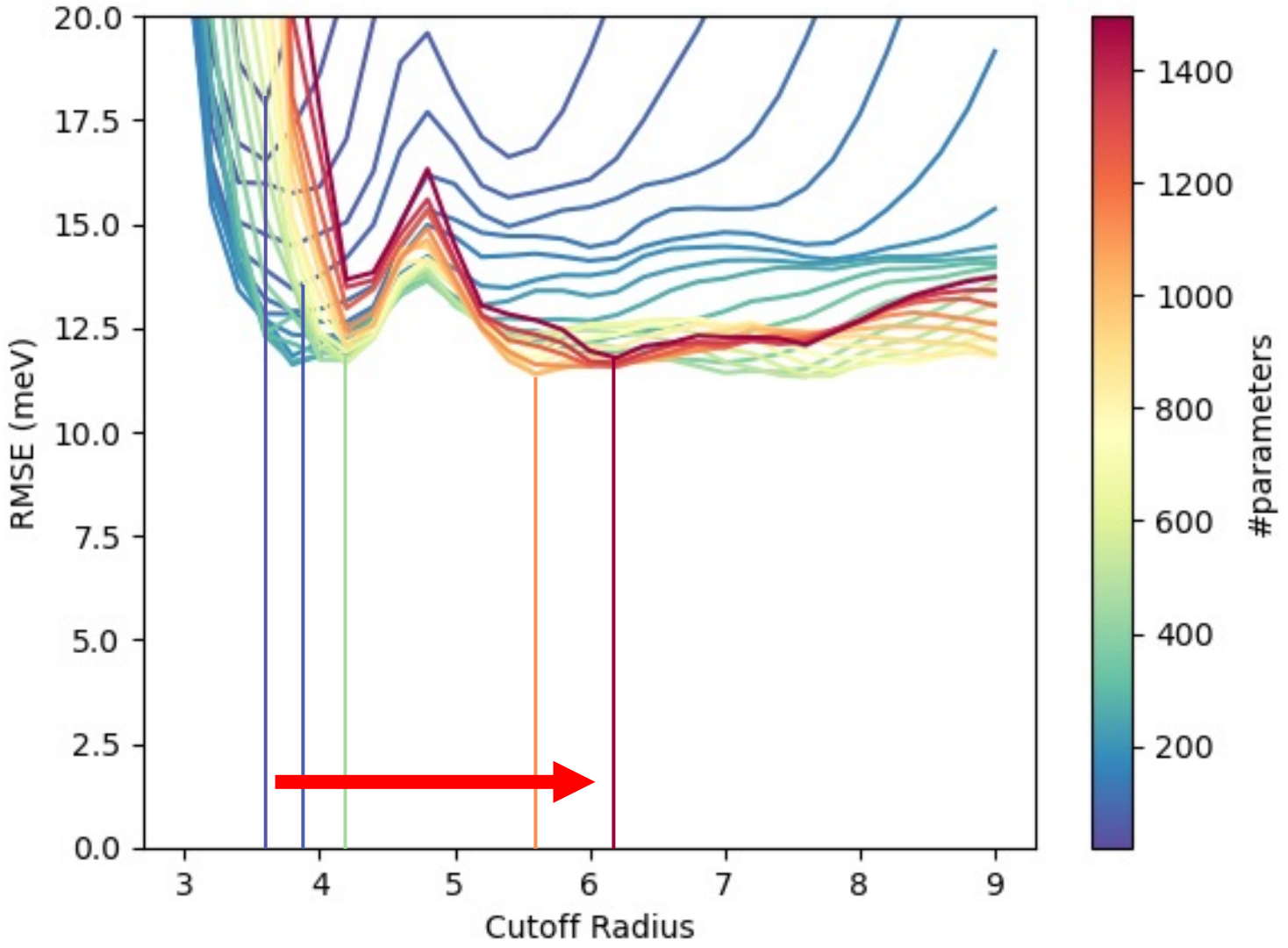
- 26 $2j_{max}$
- 31 radii

Total:

806 calculation

Optimal Cut-off Radius

For Spectral Neighbour Analysis Potential (SNAP)



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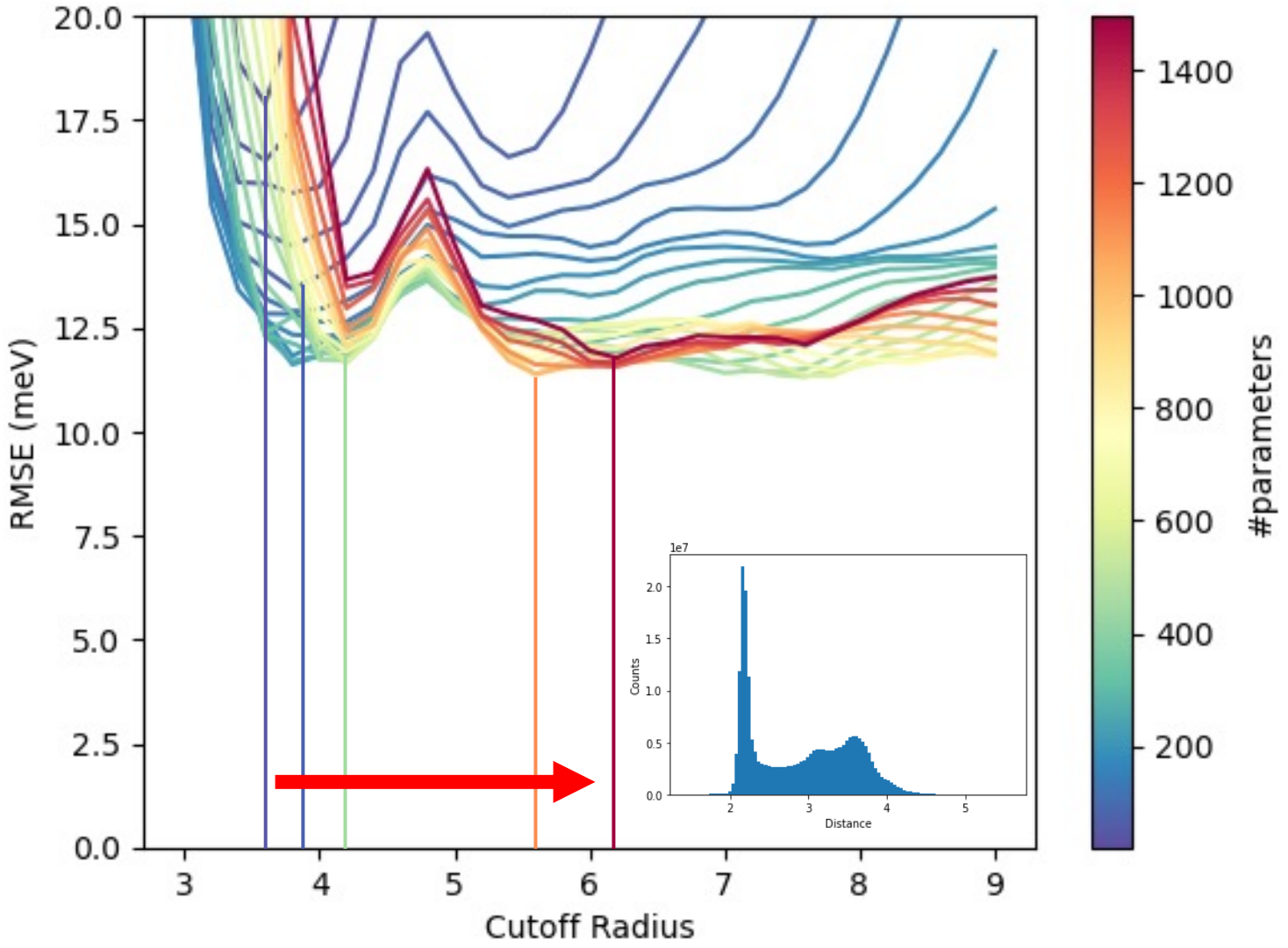

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806 calculation

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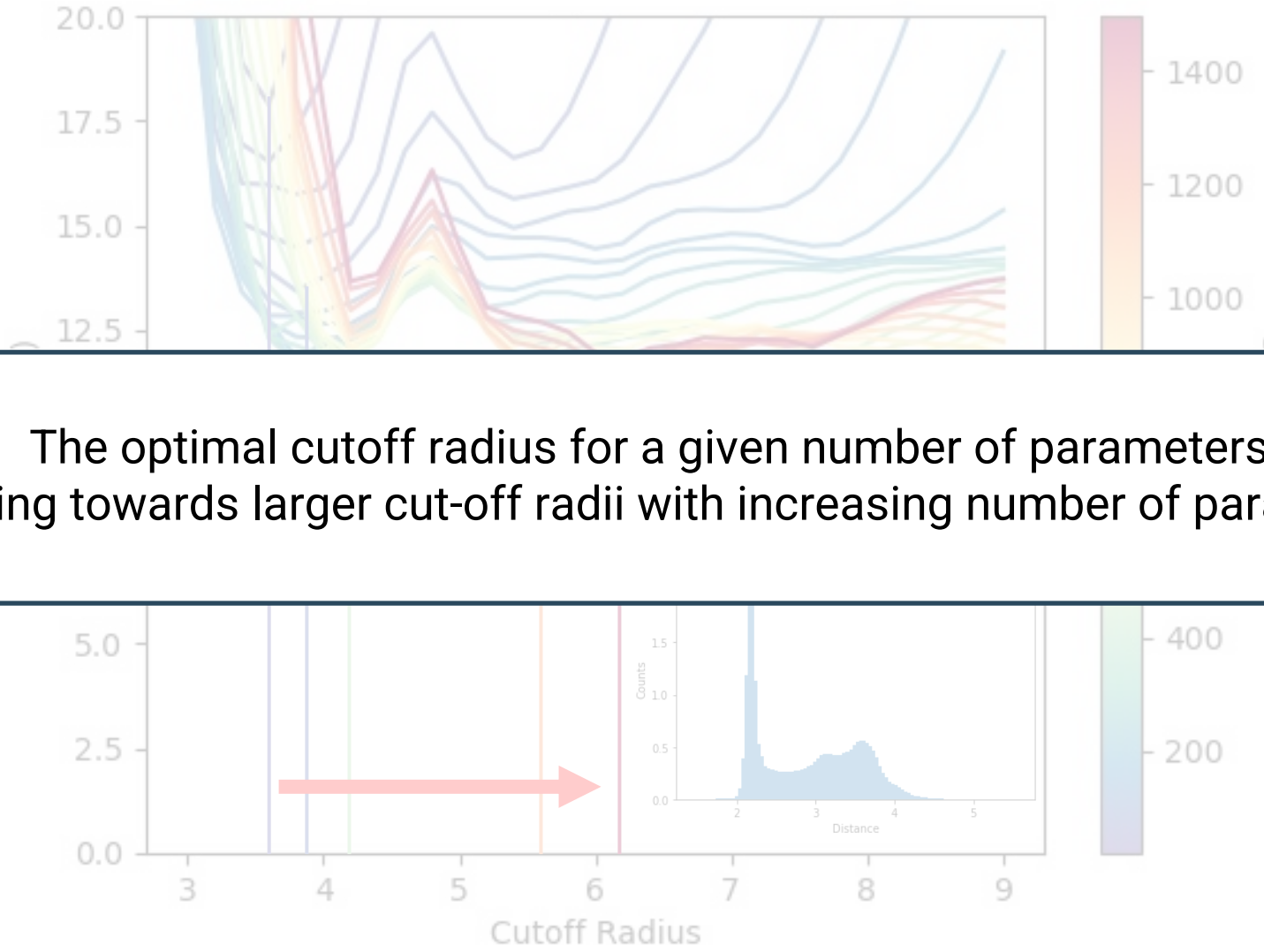
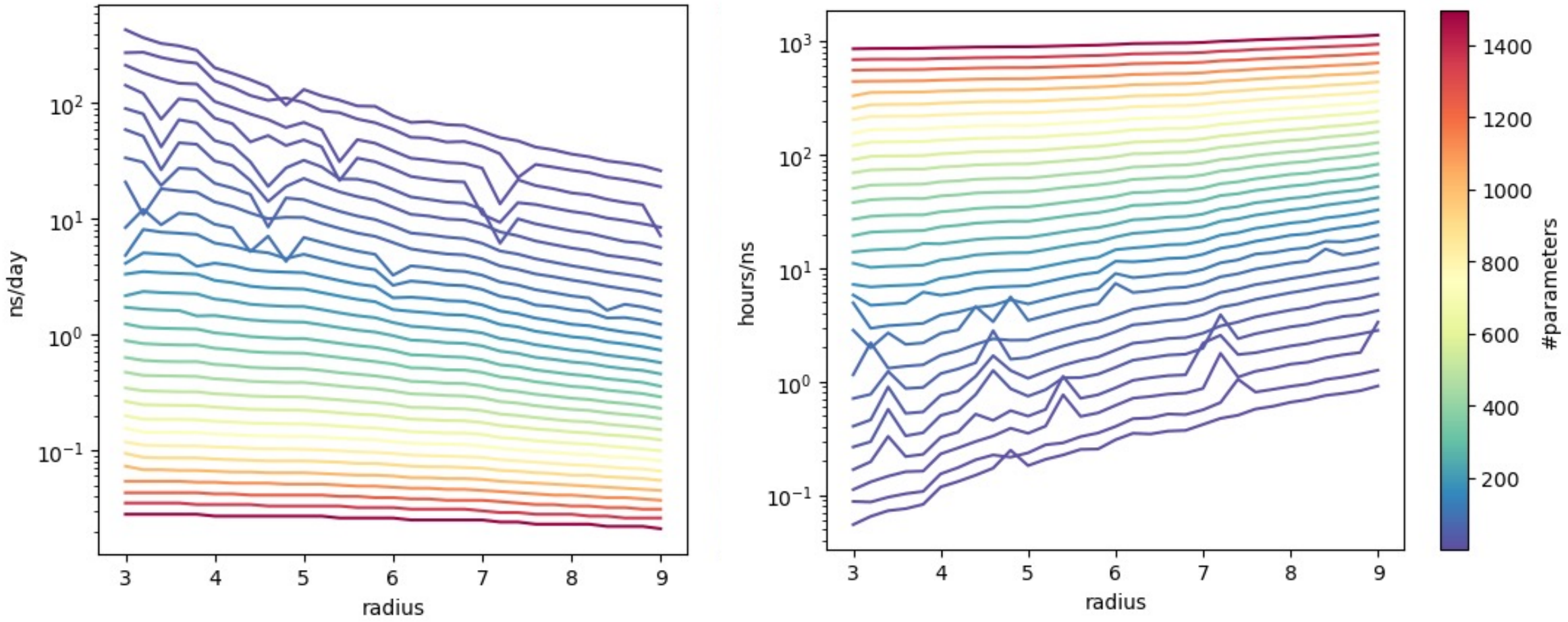


The optimal cutoff radius for a given number of parameters
is shifting towards larger cut-off radii with increasing number of parameters.

Computational Cost calculating Molecular Dynamics

For Spectral Neighbour Analysis Potential (SNAP)



Computational Cost calculating Molecular Dynamics

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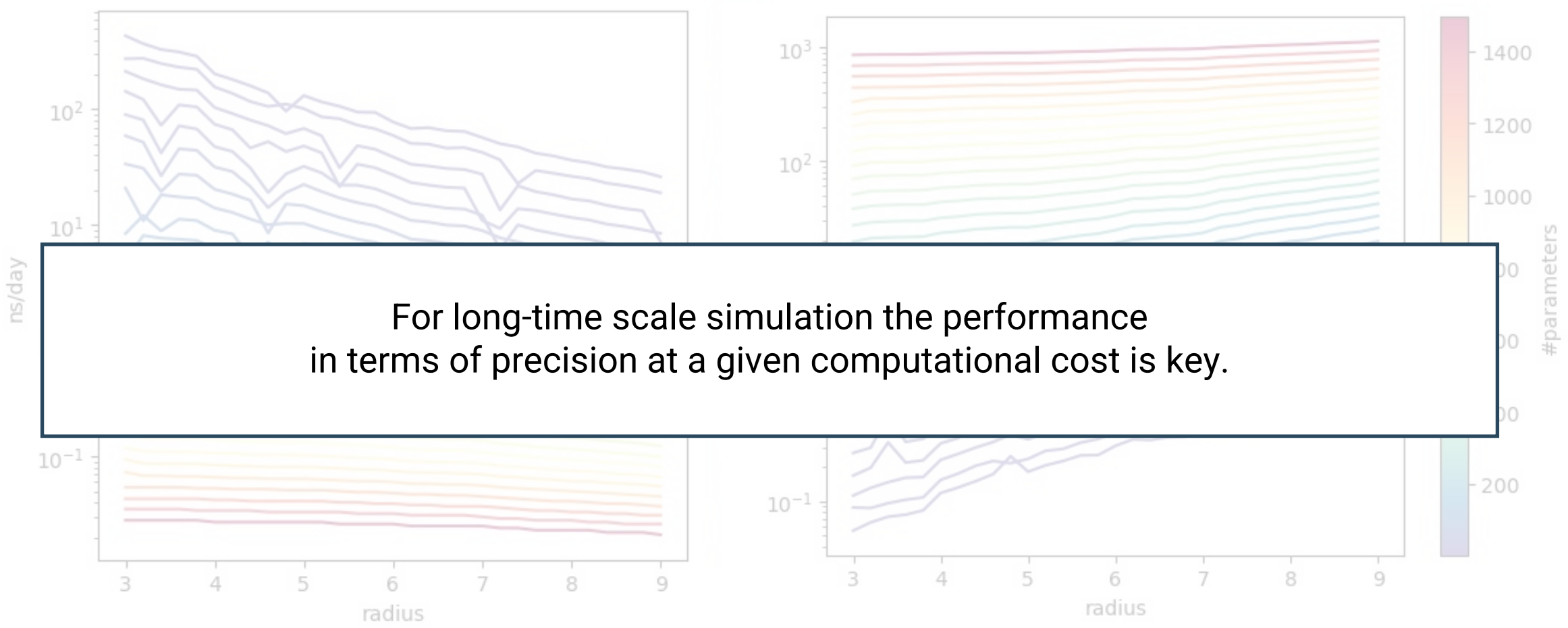
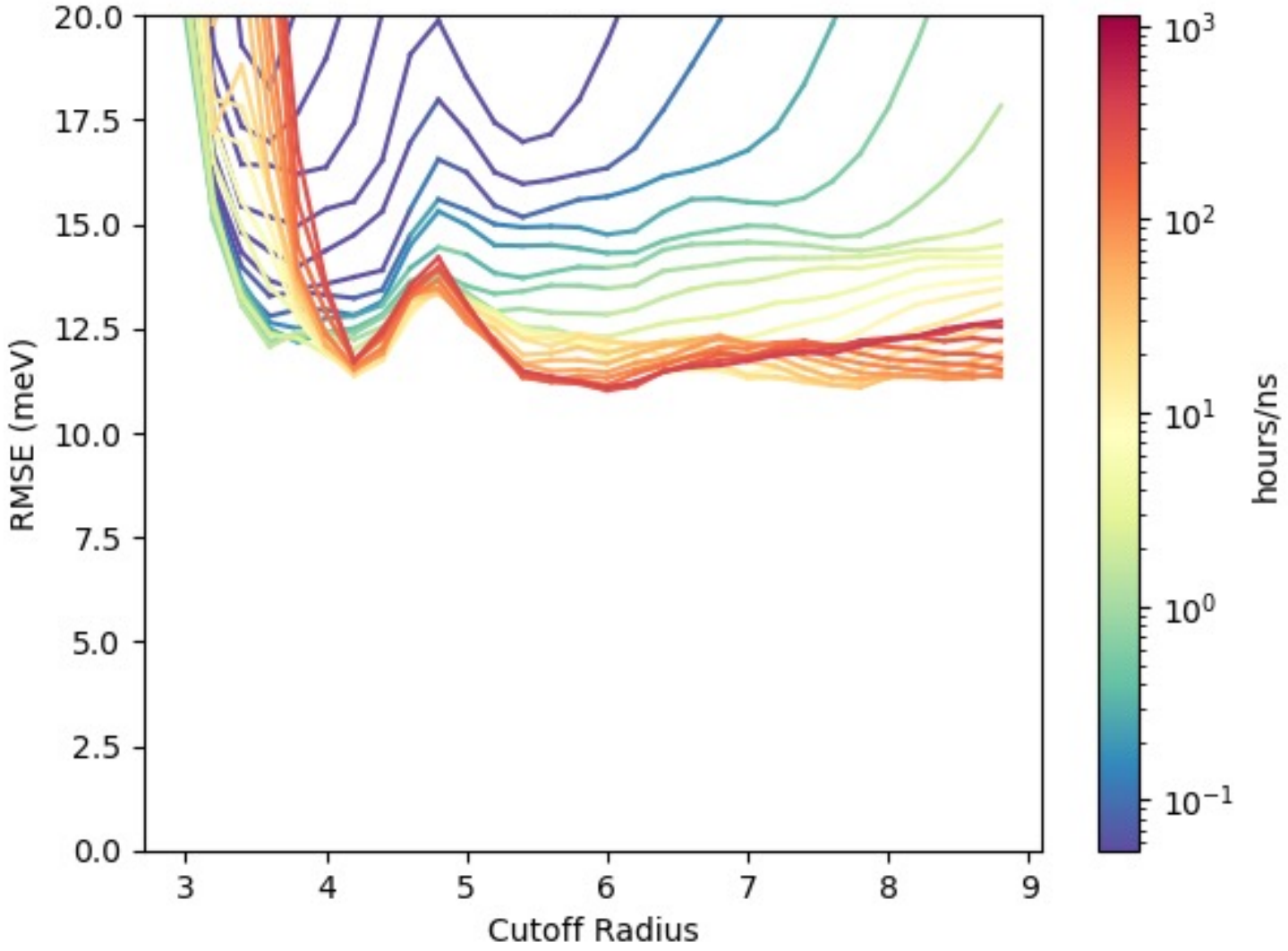


For long-time scale simulation the performance
in terms of precision at a given computational cost is key.

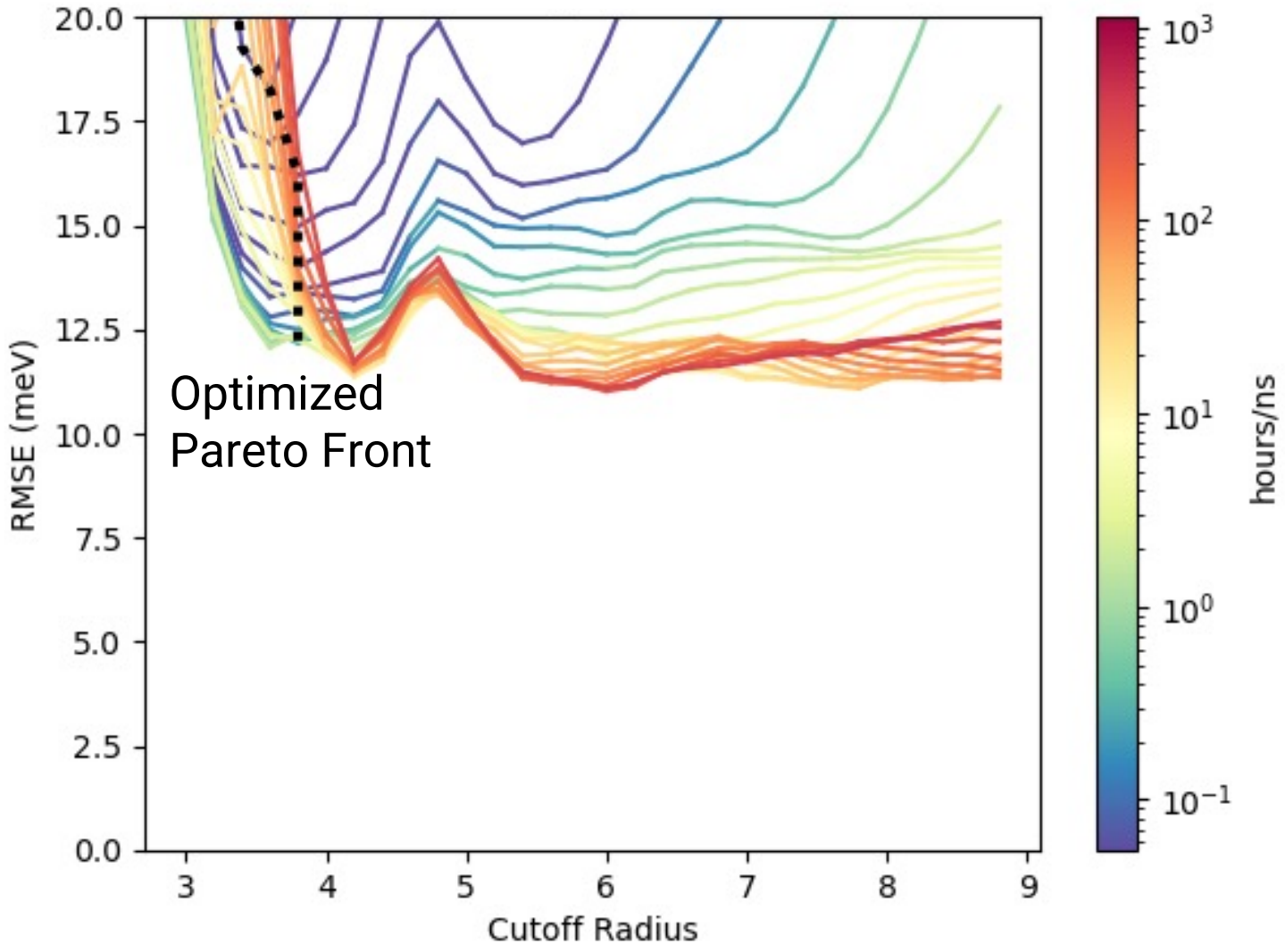
Pareto Front

Including Computational Cost



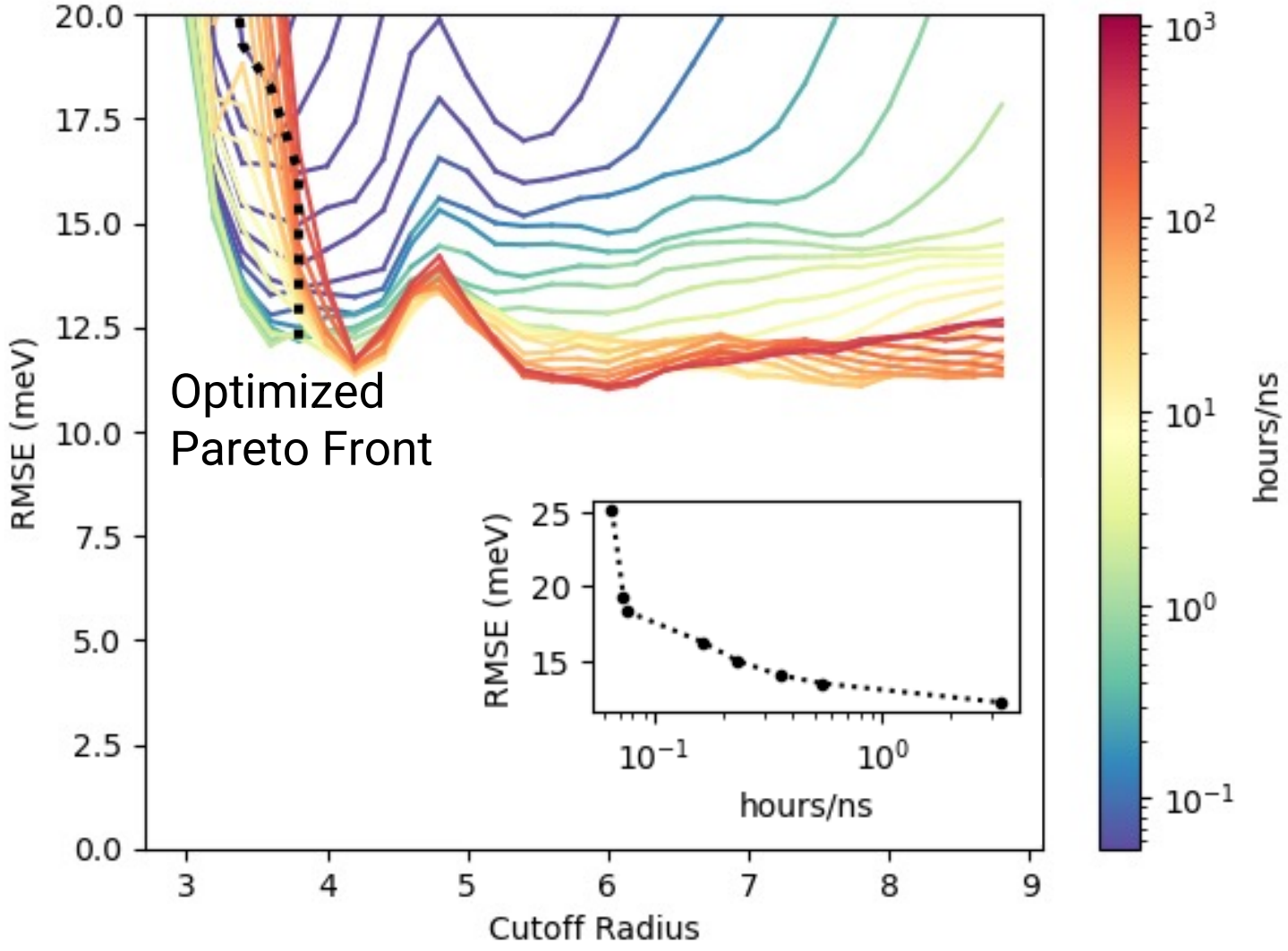
Pareto Front

Including Computational Cost



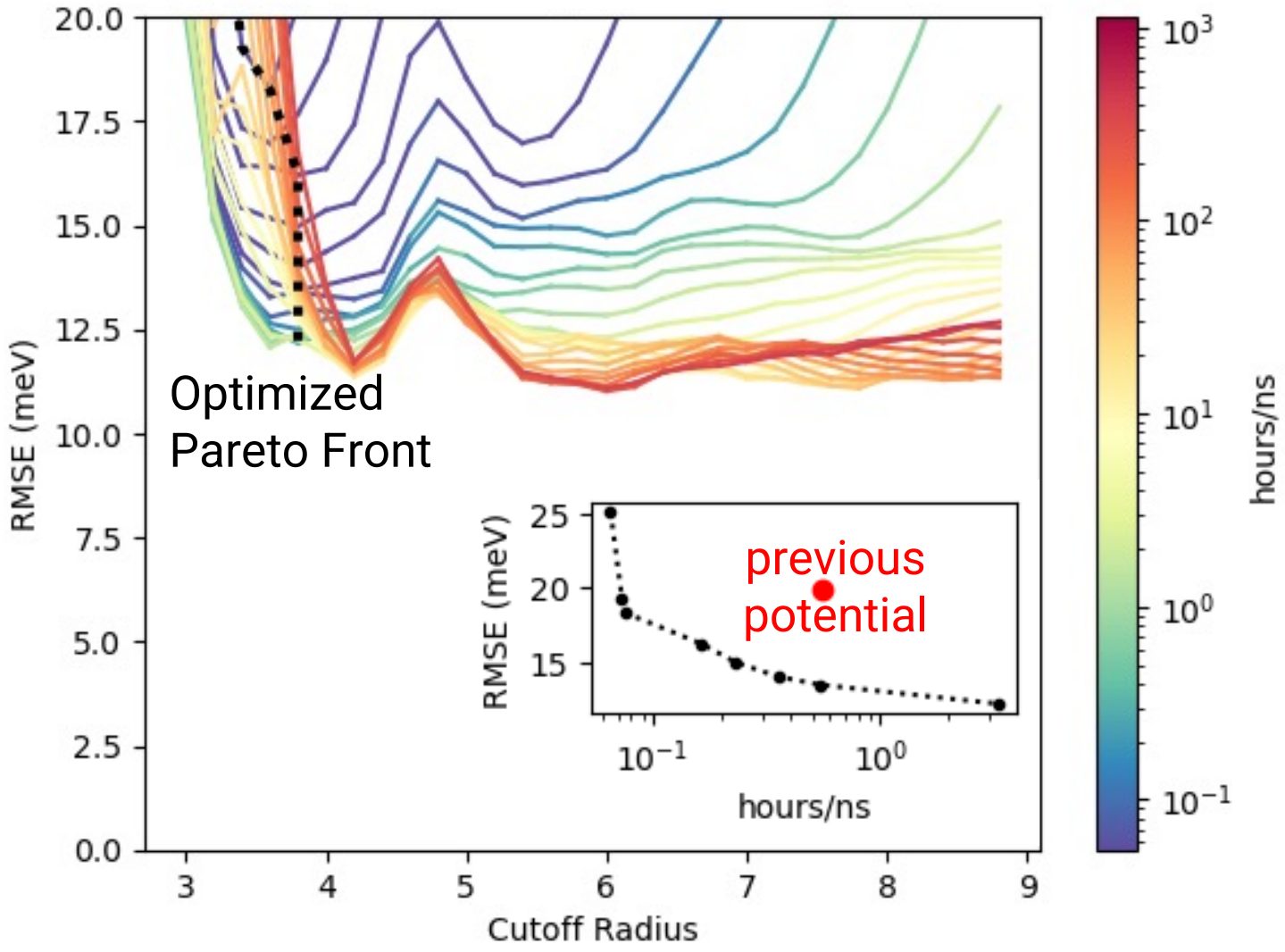
Pareto Front

Including Computational Cost



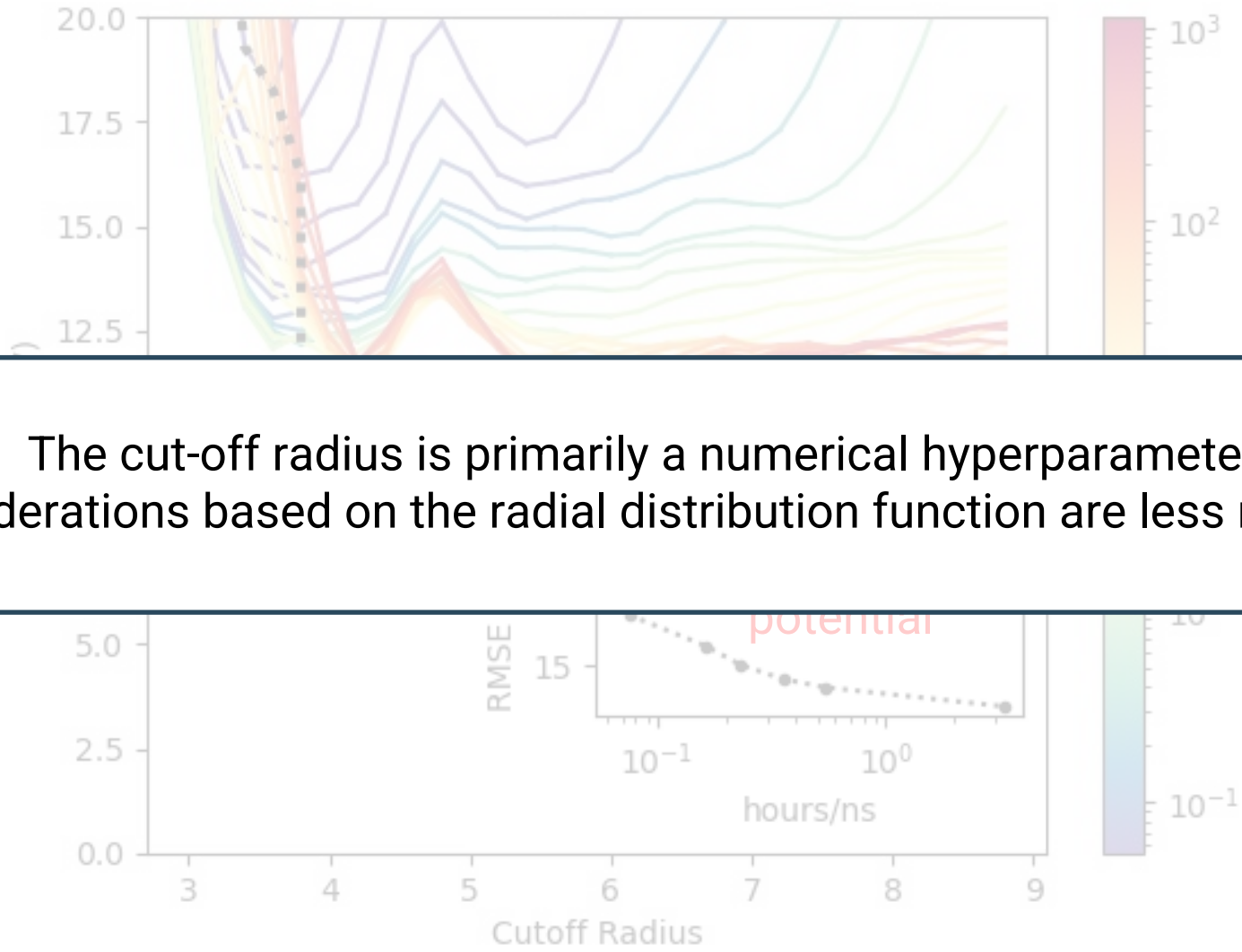
Pareto Front

Including Computational Cost



Pareto Front

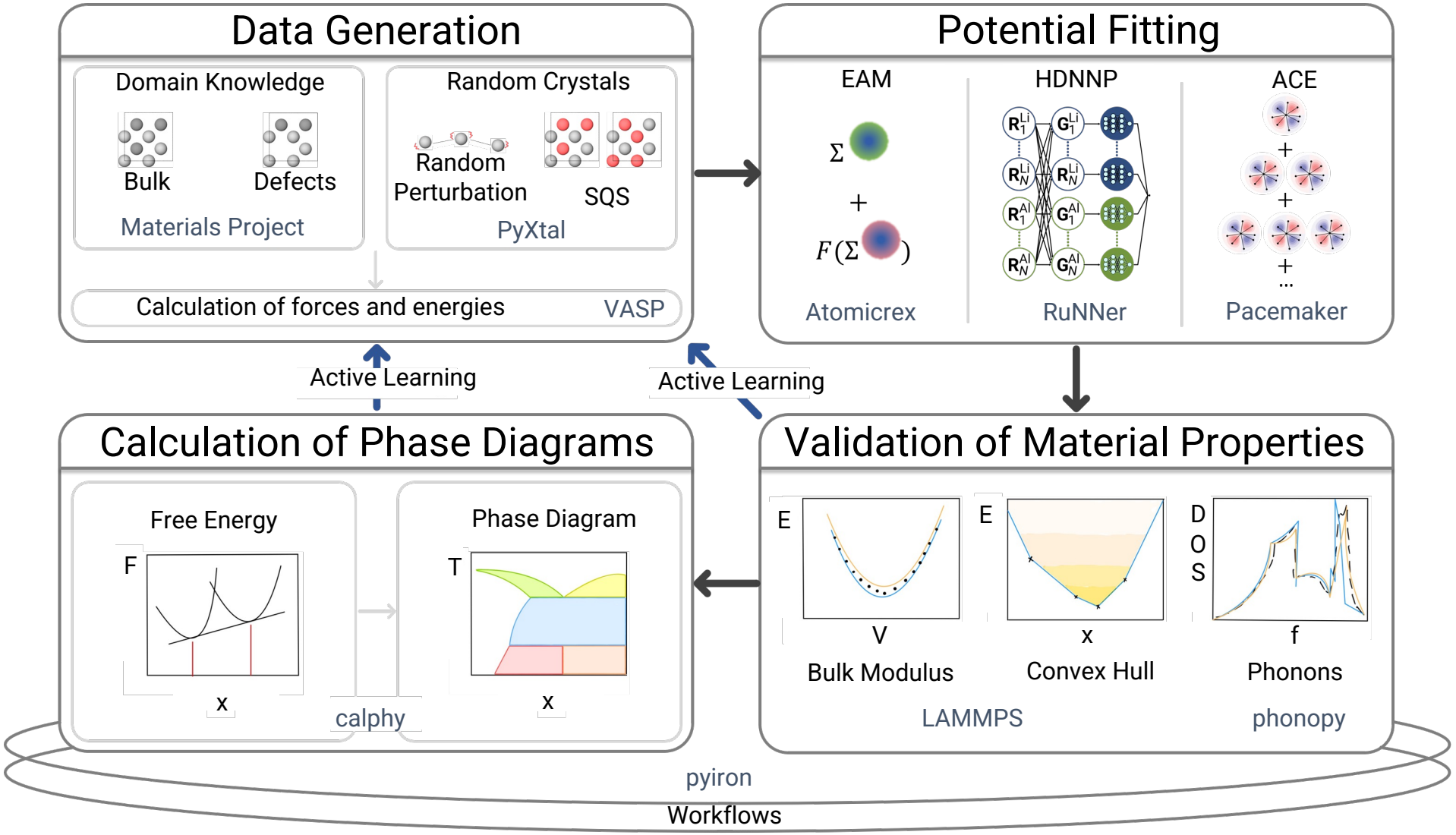
Including Computational Cost



The cut-off radius is primarily a numerical hyperparameter,
considerations based on the radial distribution function are less relevant.

Temperature Concentration Phase Diagram

Ab-initio Thermodynamics



S. Menon, Y. Lysogorskiy, A. L. M. Knoll, N. Leimeroth, M. Poul, M. Qatar, J. Janssen, M. Mrovec, J. Rohrer, K. Albe, J. Behler, R. Drautz and J. Neugebauer. npj Comput. Mater., 10, 261 (2024)

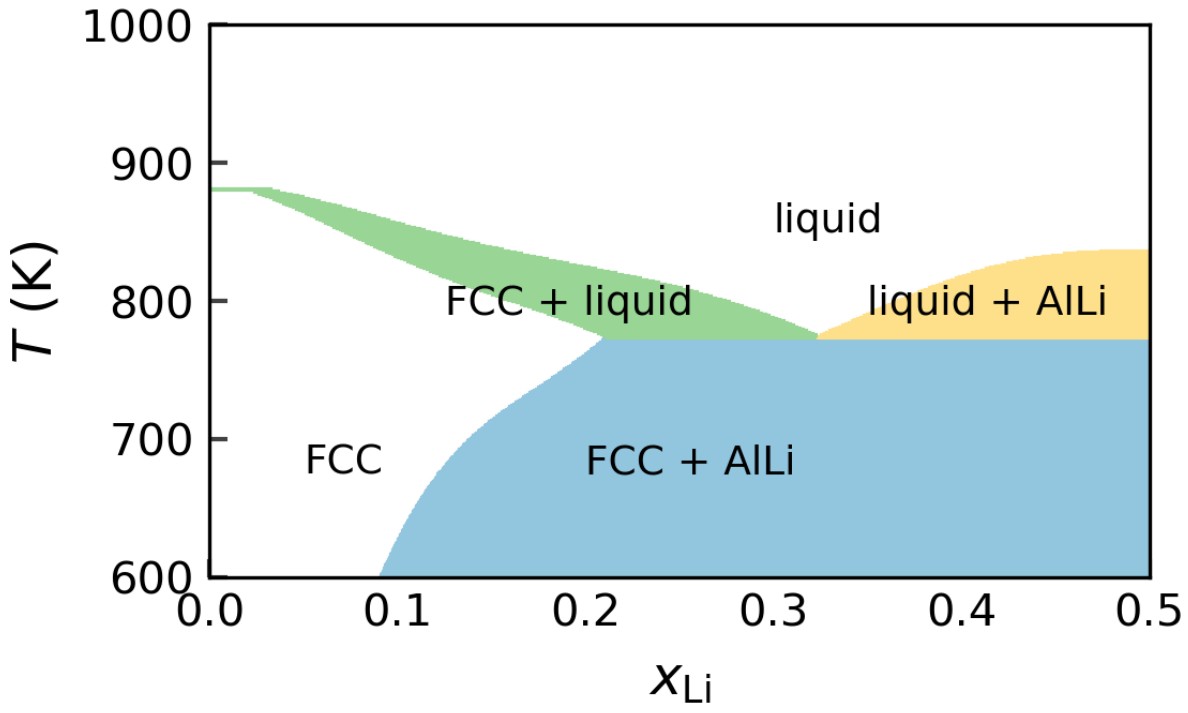
Temperature Concentration Phase Diagram

Ab-initio Thermodynamics

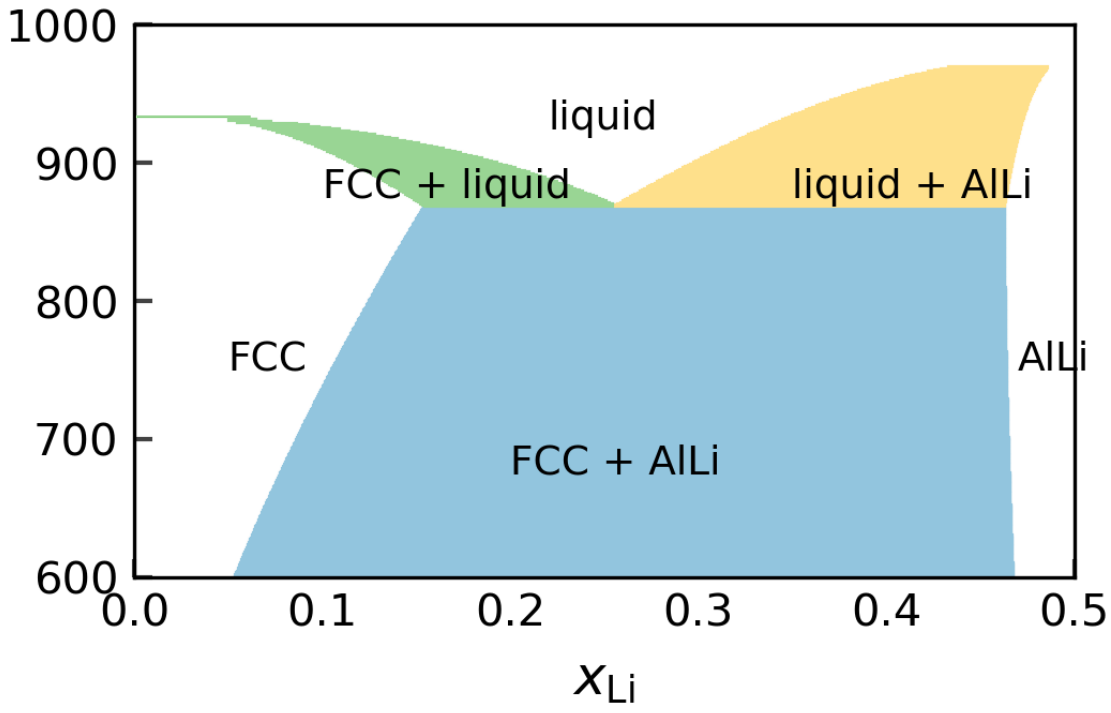
Data Generation

Potential Fitting

Machine-Learned Interatomic Potential



Experiment (CALPHAD)



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Workflows

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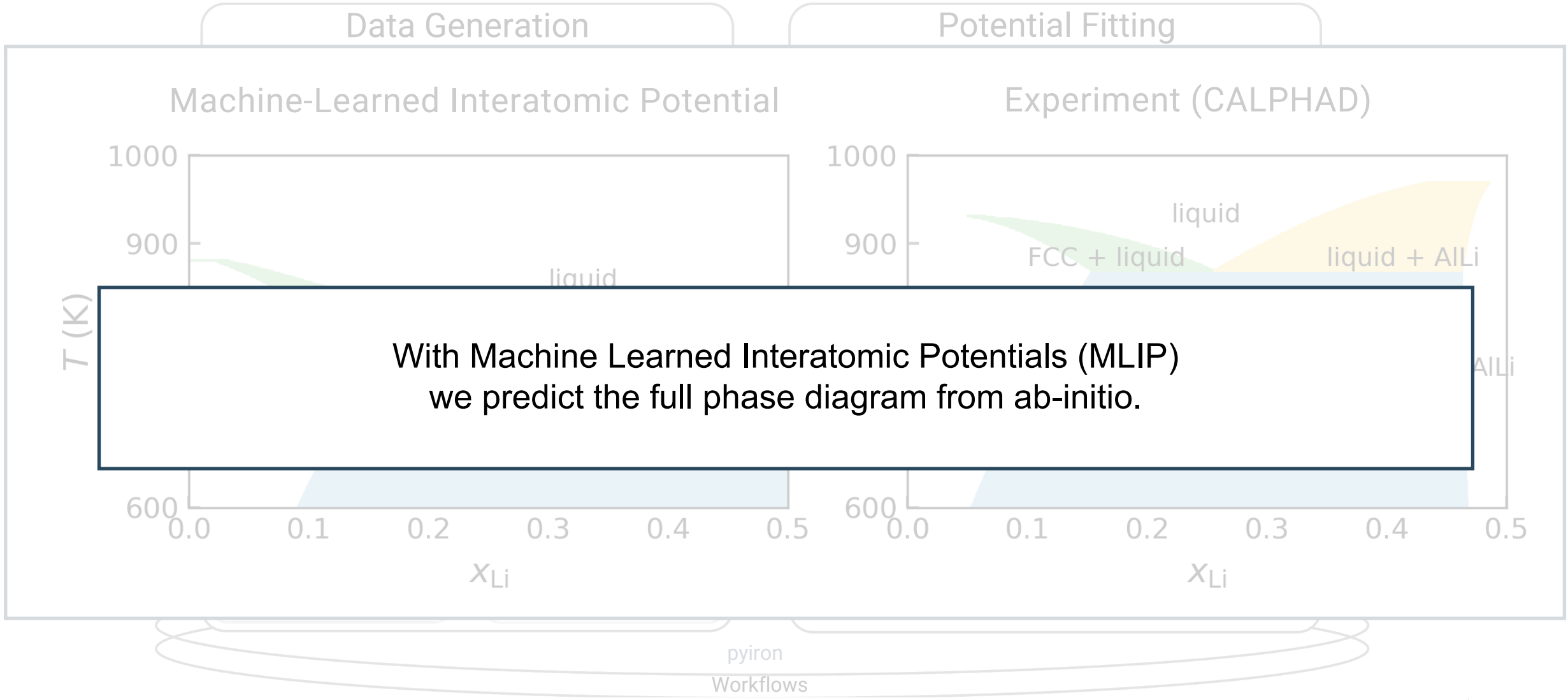
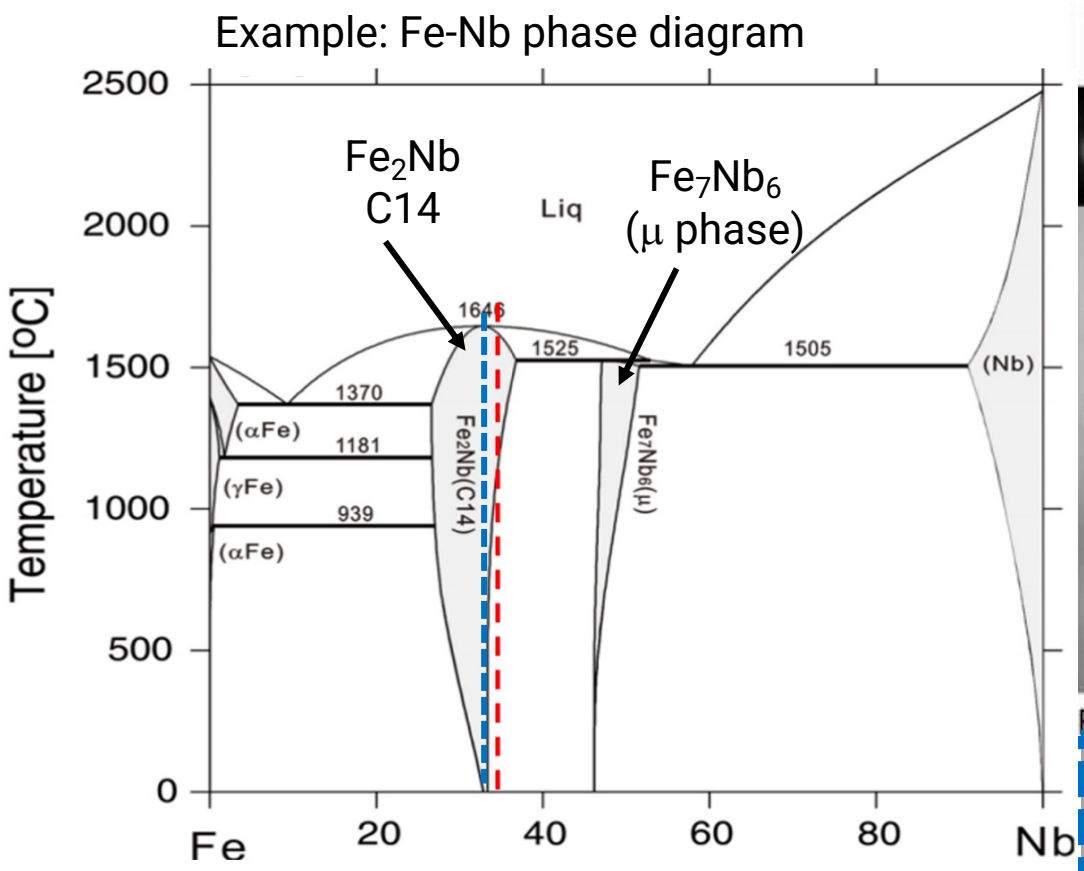


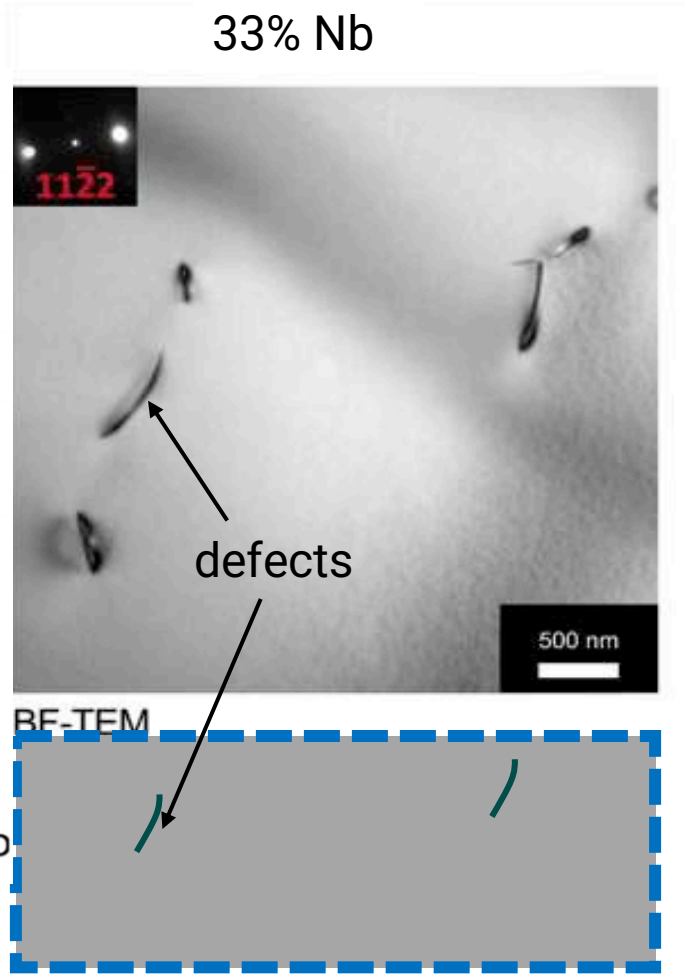
With Machine Learned Interatomic Potentials (MLIP)
we predict the full phase diagram from ab-initio.

S. Menon, Y. Lysogorskiy, A. L. M. Knoll, N. Leimeroth, M. Poul, M. Qatar, J. Janssen, M. Mrovec, J. Rohrer, K. Albe, J. Behler, R. Drautz and J. Neugebauer. npj Comput. Mater., 10, 261 (2024)

Defect Phase Diagram

Can we apply the same concepts to understand and design microstructure?





M. Slapakova, A. Zendegani, C.H. Liebscher, T. Hickel, J. Neugebauer, T. Hammerschmidt, A. Ormeci, J. Grin, G. Dehm, K.S. Kumar, F. Stein, Acta Mater 183, 362 (2020)

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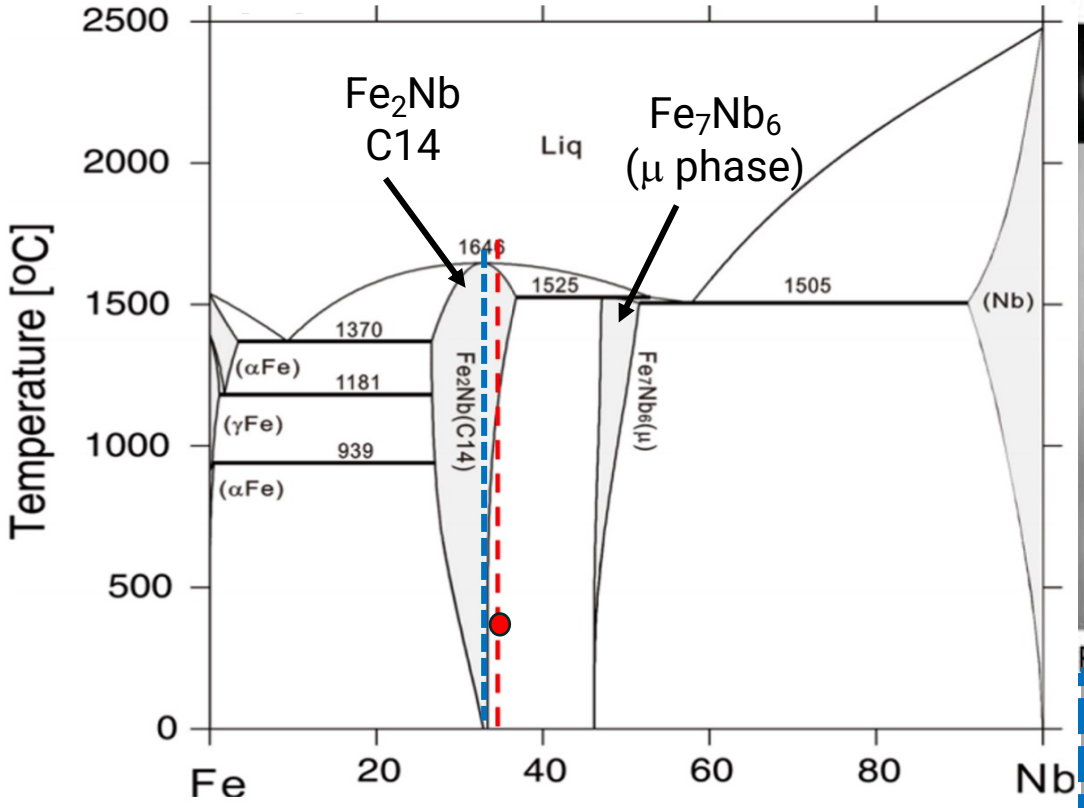
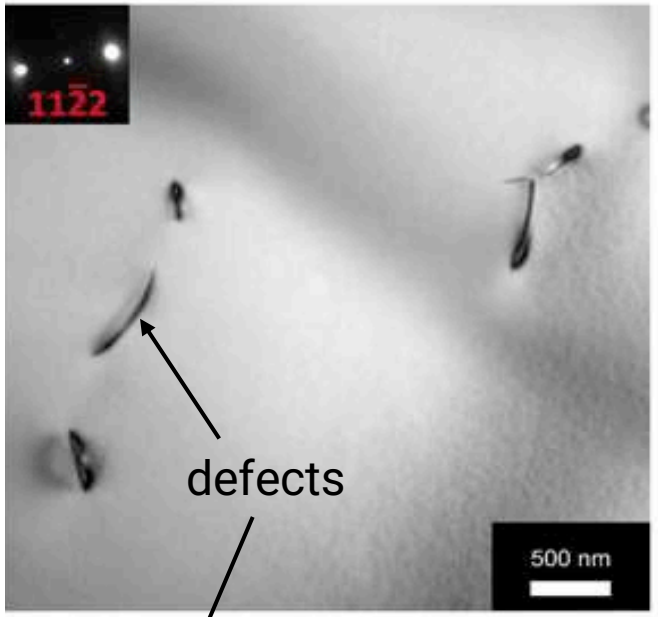
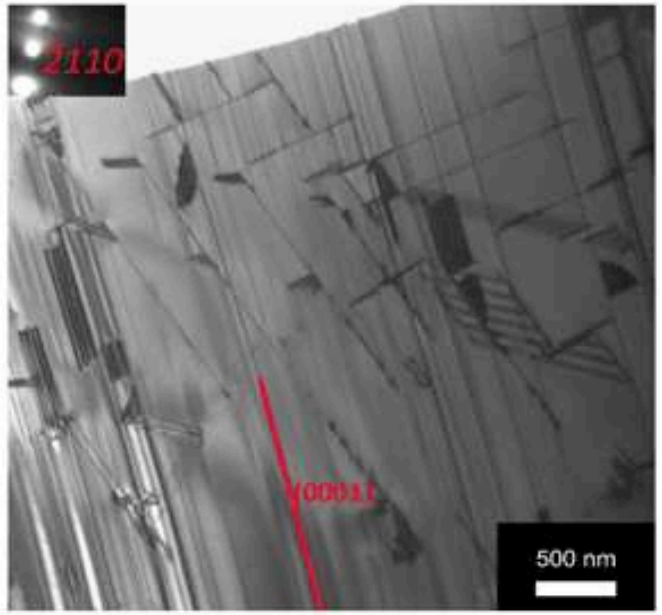
Example: Fe-Nb phase diagram

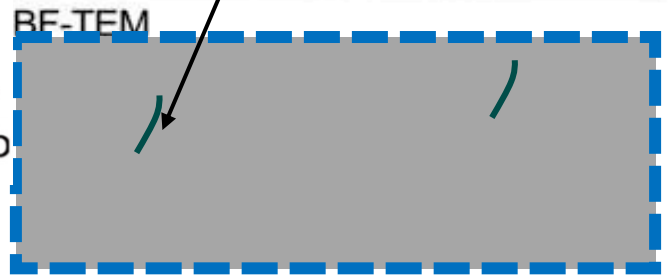


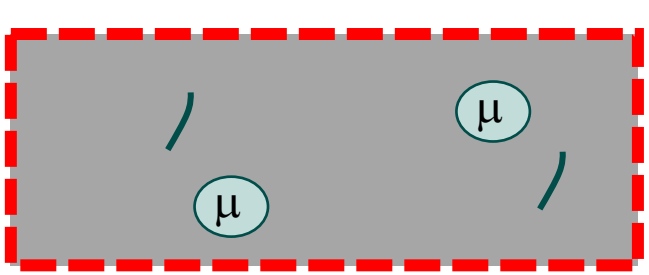
33% Nb



35% Nb







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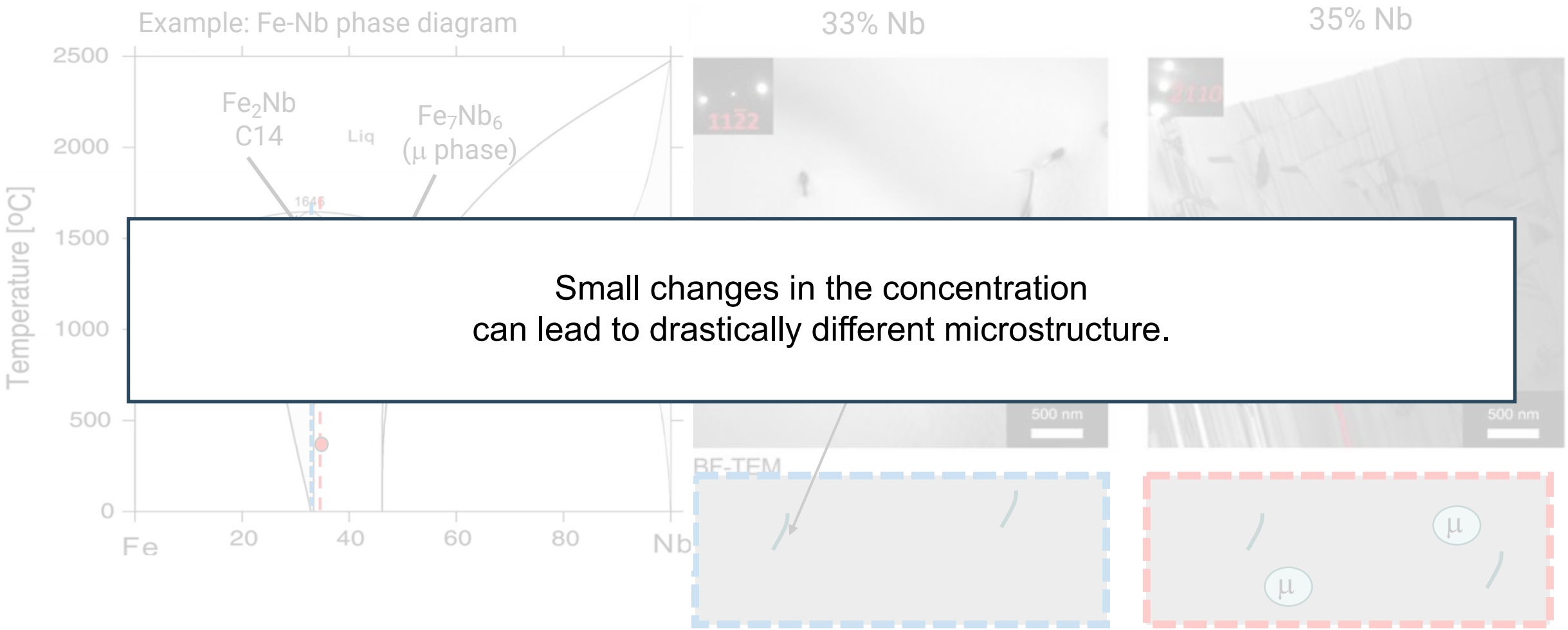

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Defect Formation Energies and Phase Diagram

Defect formation energy: $$E_\sigma^f = F_\sigma^{DFT} - \sum_i n_i^\sigma \mu_i$$

σ - defect state
i - chemical species
n_i - number of species i atoms

S. Korte-Kerzel, T. Hickel, L. Huber, D. Raabe, S. Sandlöbes-Haut, M. Todorova, and J. Neugebauer. Int. Mat. Rev., 67(1), 89–117. (2021)

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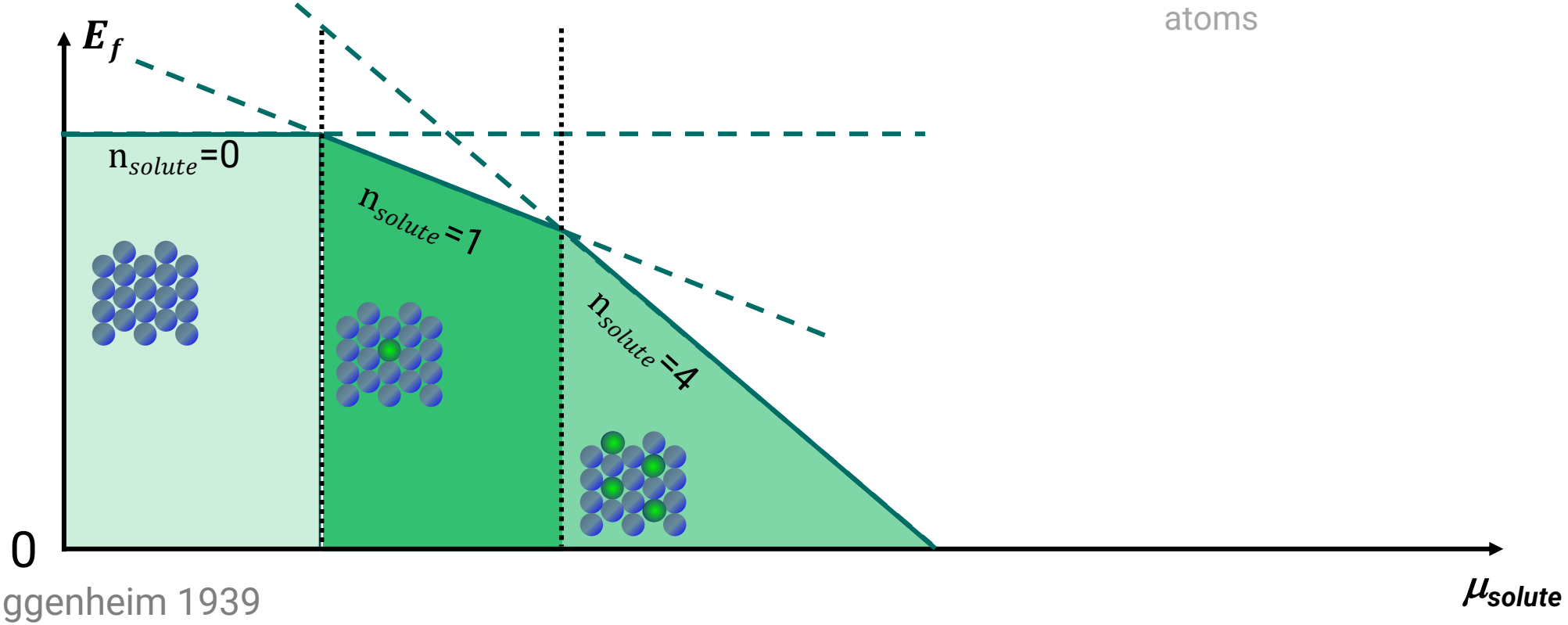

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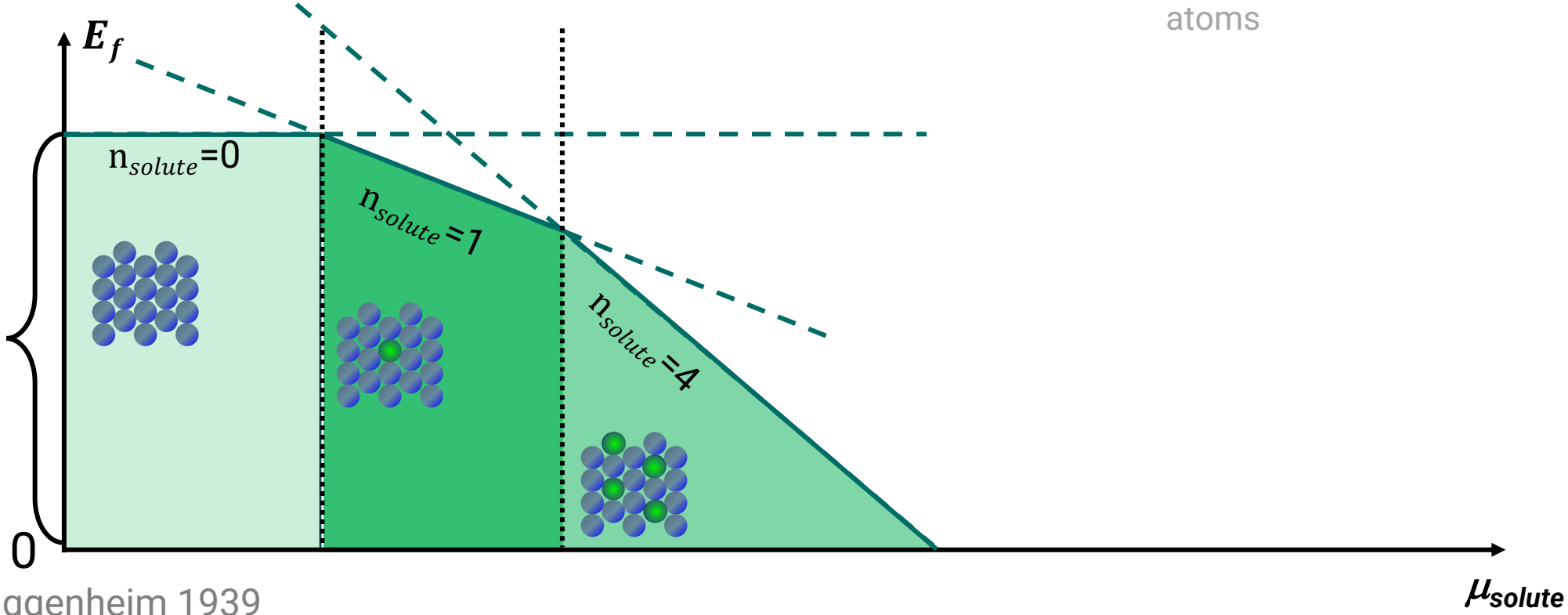

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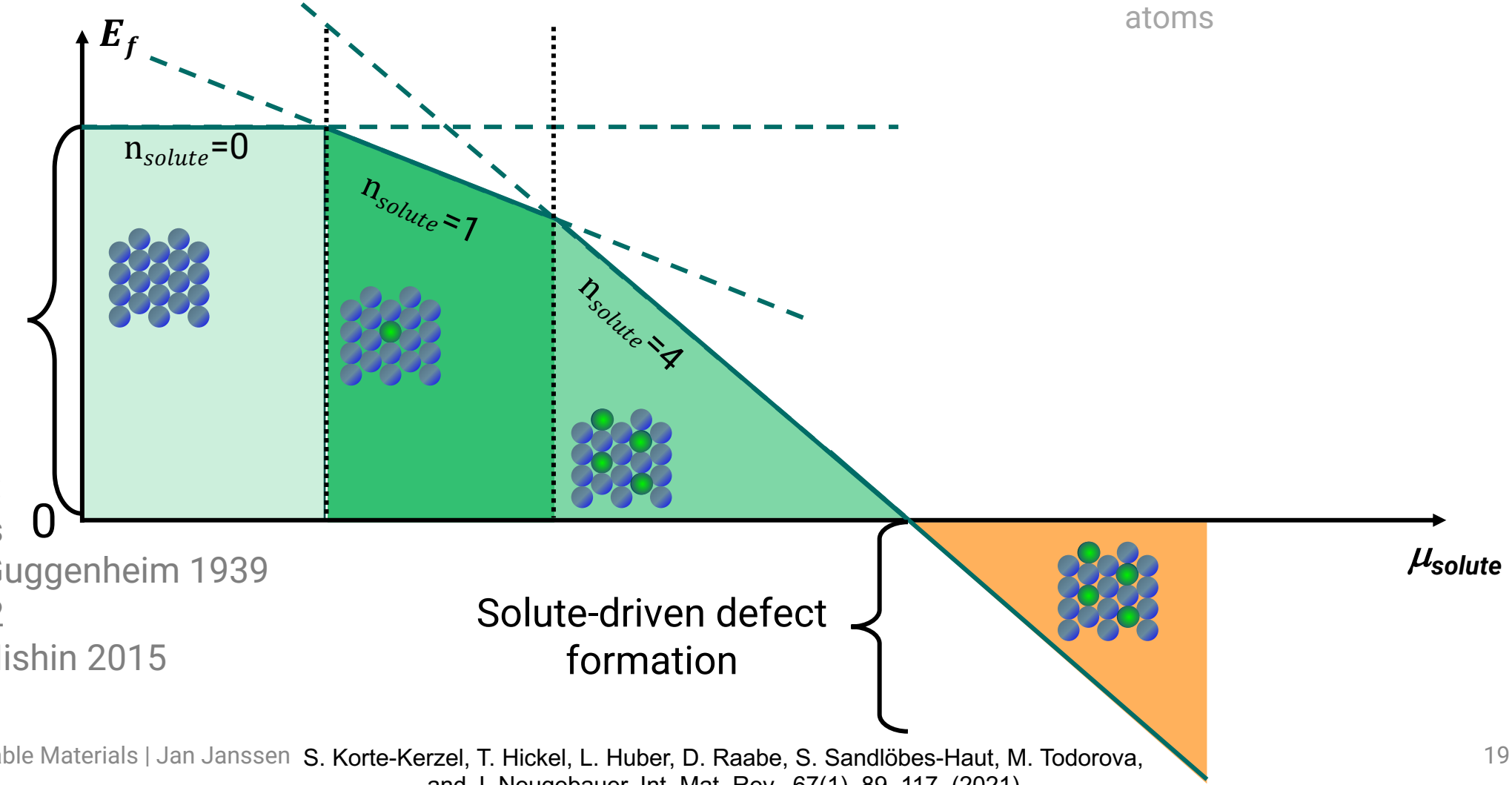

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Solute-driven defect formation

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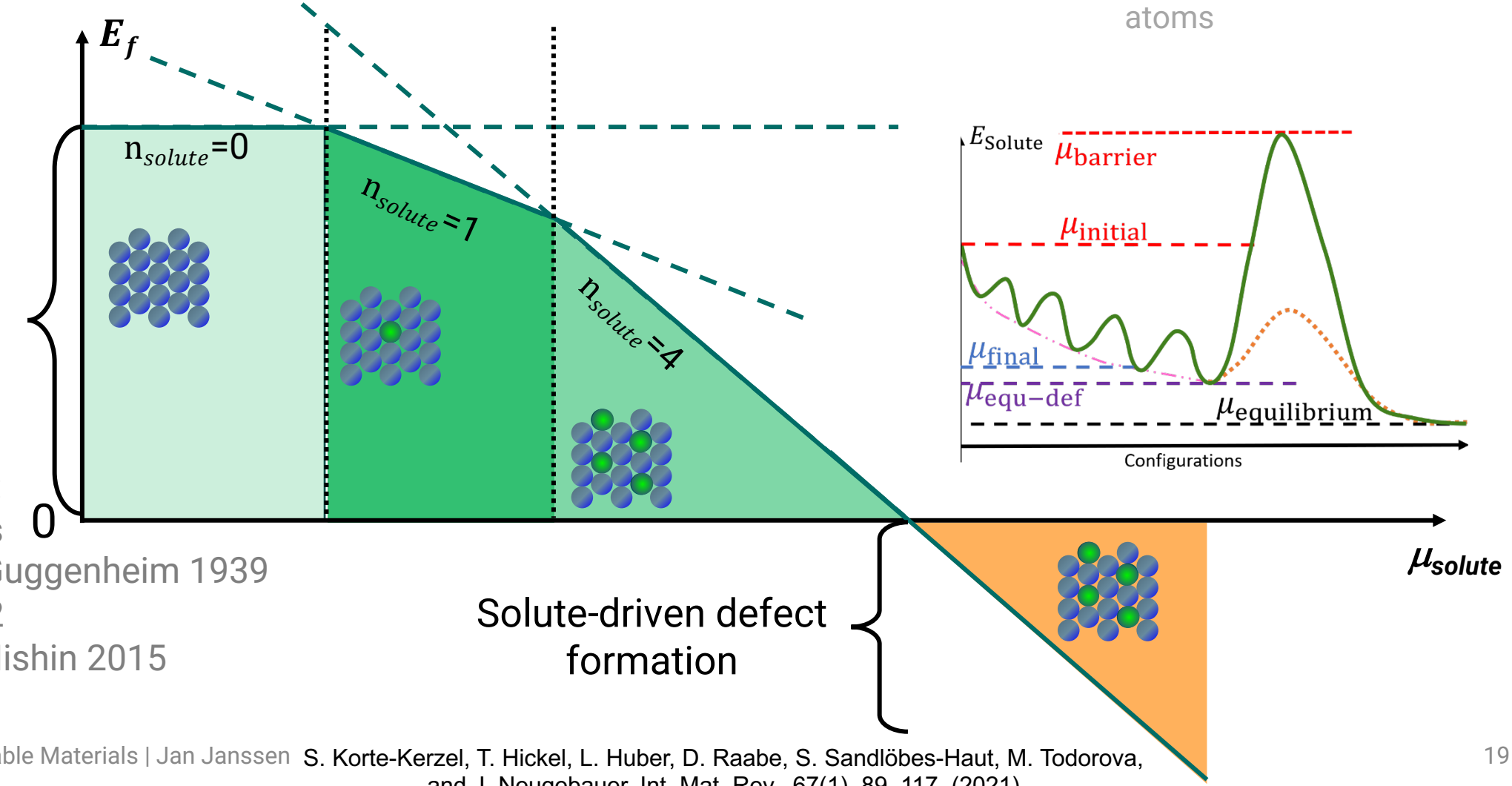

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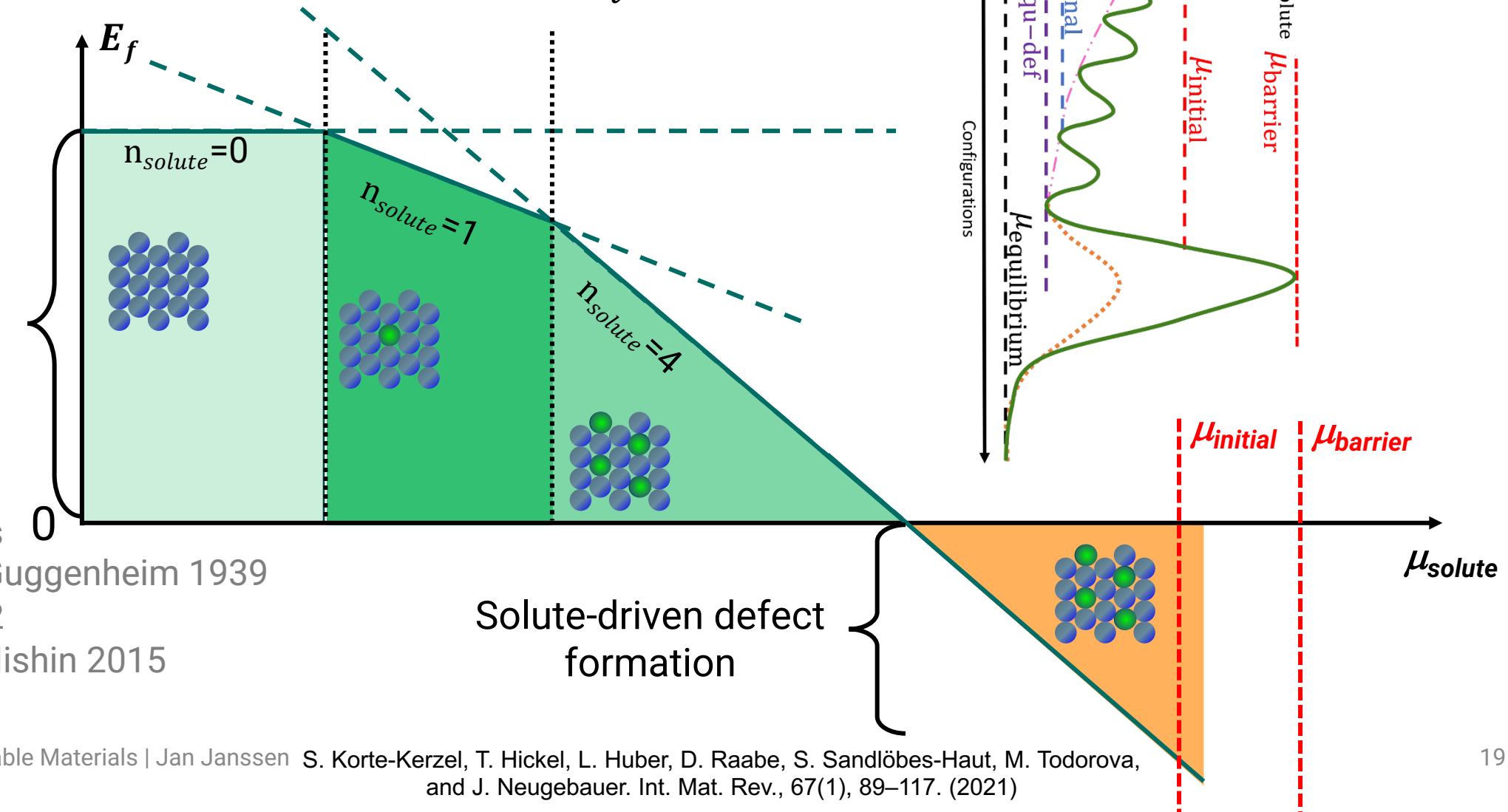

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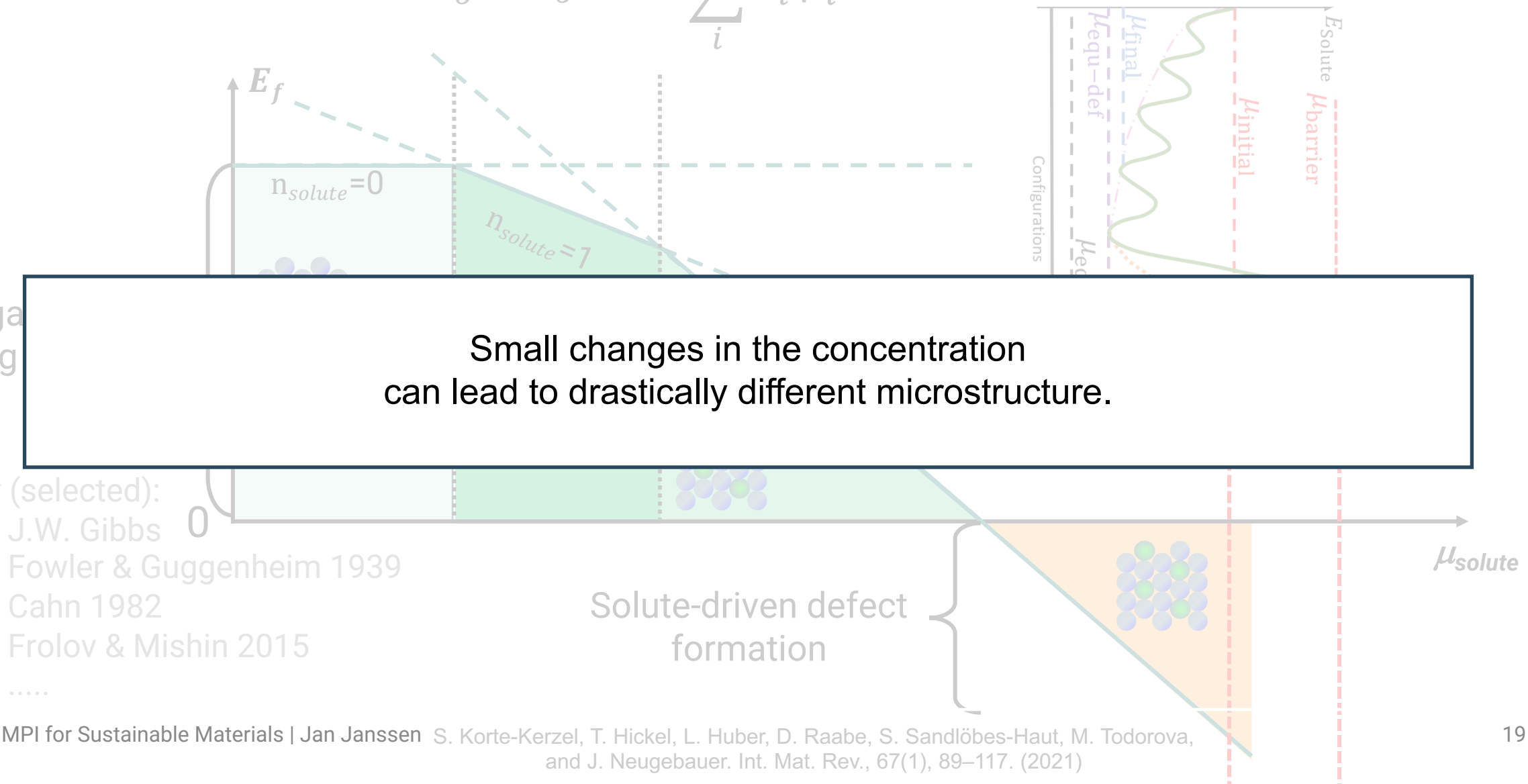

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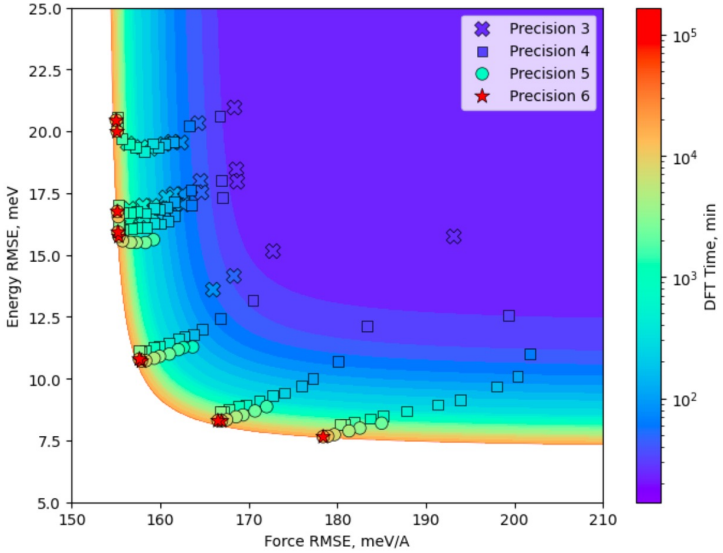
Workflows to Accelerate Materials Discovery

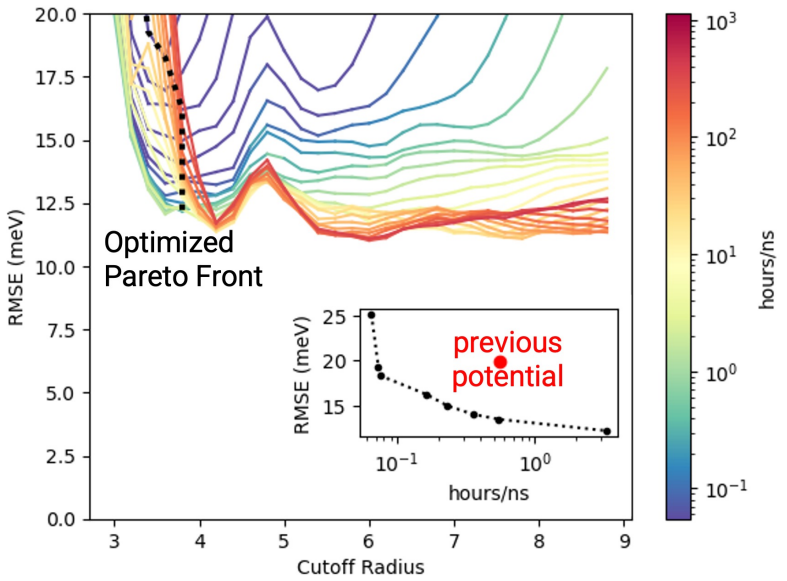
Inspiration from Functional Programming

Density Functional Theory Uncertainty Quantification



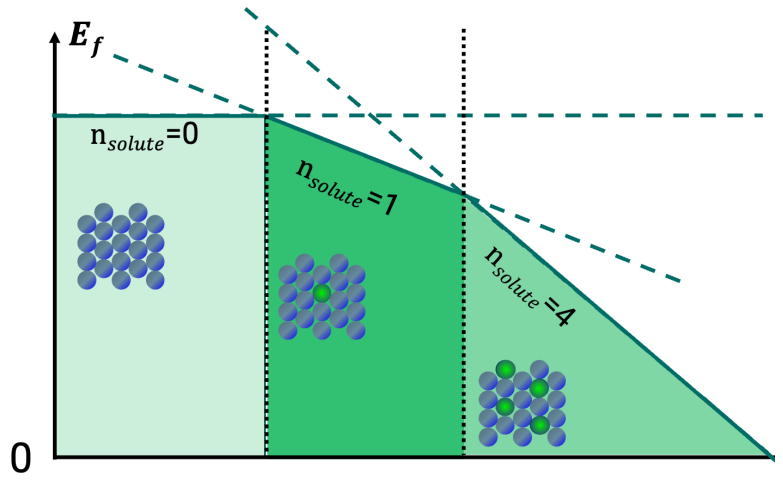
The desired application of the machine learned interatomic potential limits the benefit of high precision training data.

Computational Efficient Machine Learned Interatomic Potentials



The cut-off radius is primarily a numerical hyperparameter for optimizing potential efficiency.

Defect Phase Diagrams to Predict Microstructure



Materials Acceleration Platforms (MAP) combine simulation and experiment in the same workflow.

Workflows to Accelerate Materials Discovery – Thank you

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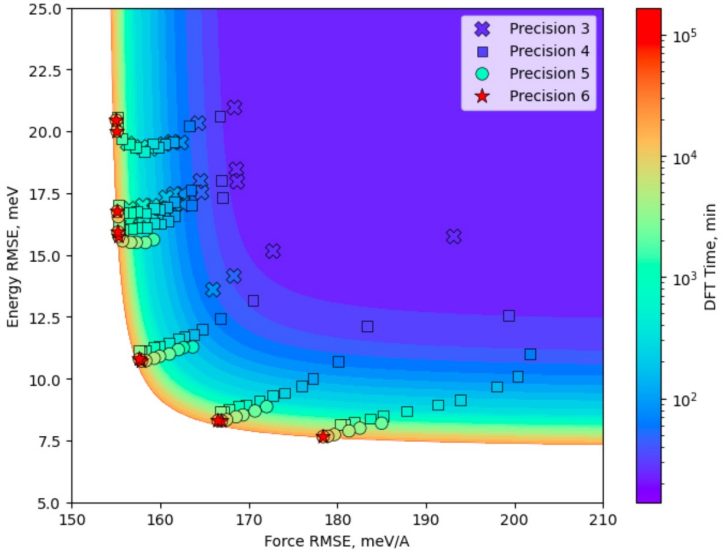

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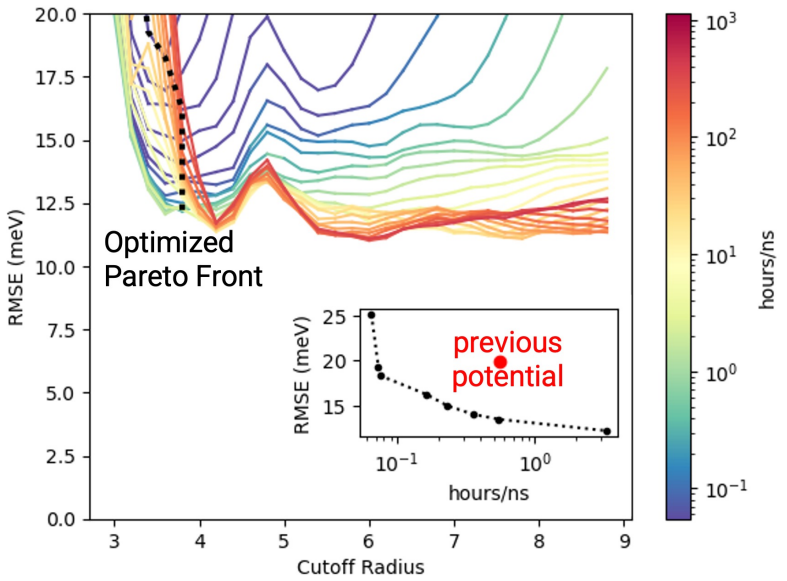

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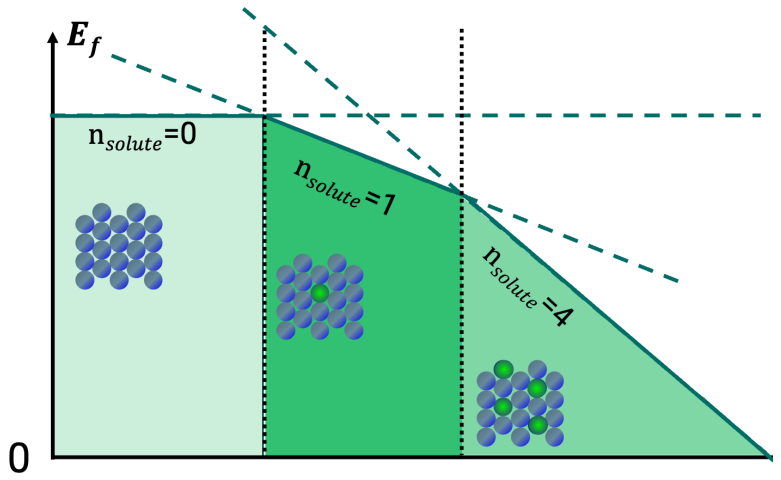

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