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Atomistic Dynamics Simulations of Complex Materials and Interfaces with Machine-Learned Force Fields

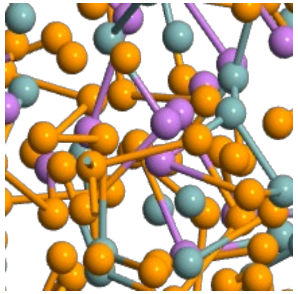
Outline

- Why do we need machine-learned force fields (ML FFs)?
- A few details on moment tensor potentials (MTPs)
- How to use automated training protocols to fit ML FFs to solve realistic problems
- Example applications of ML FFs for complex materials and interfaces

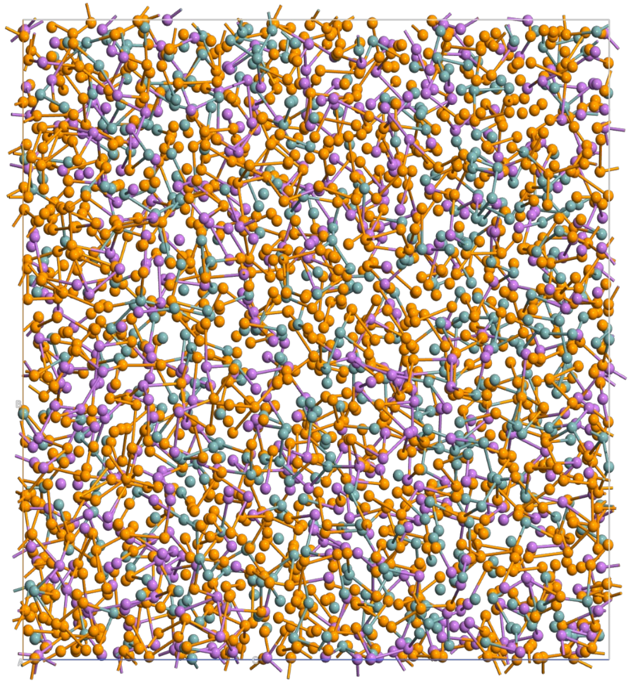
Why Do We Need Machine-Learned Force Fields?

Why Do We Need Machine-Learned Force Fieds?

Accurate dynamical modeling is limited to ~100 atoms due to TAT limitations



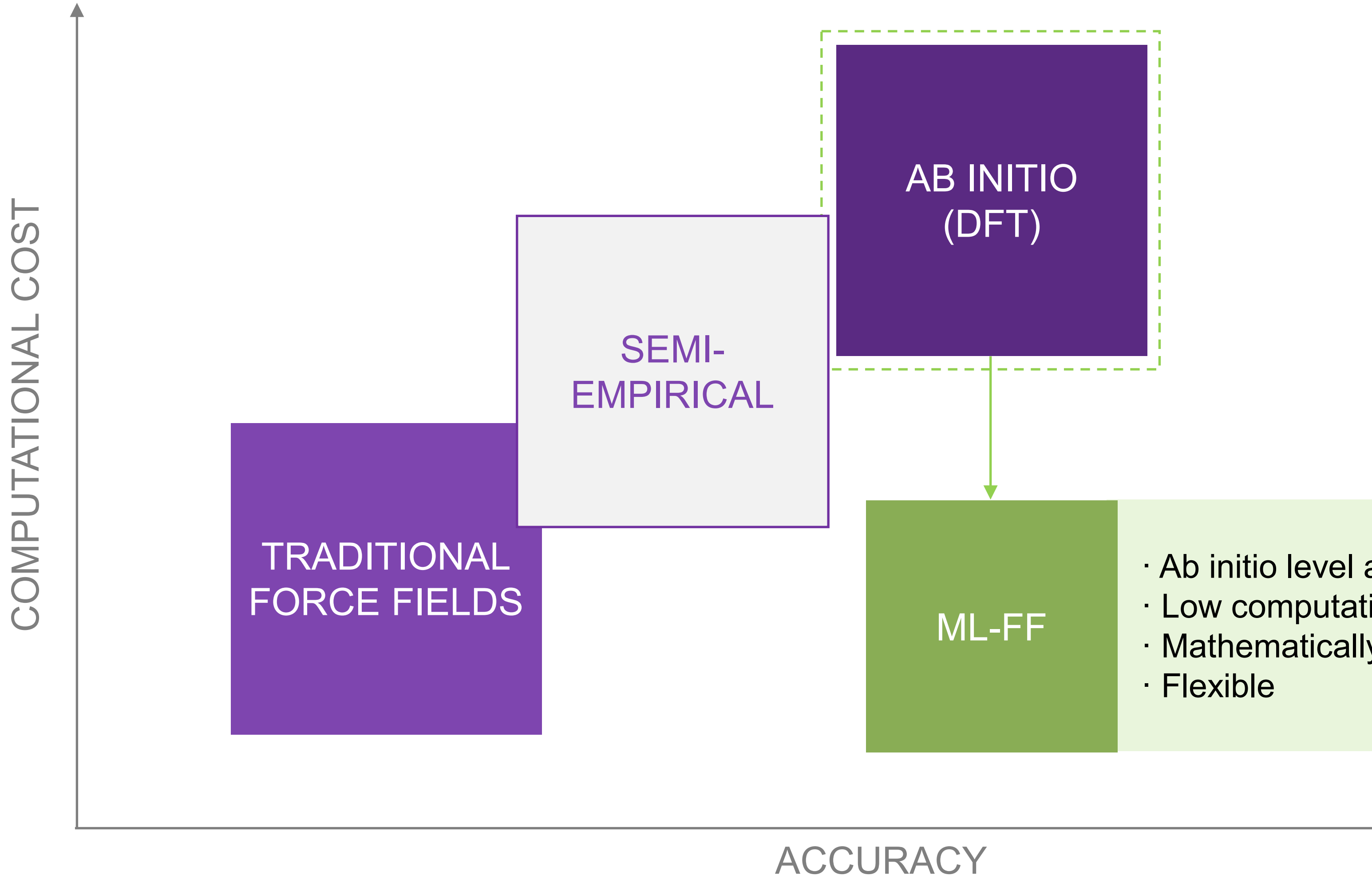
Yet many problems of interest to the industry require 10,000+ atoms to be simulated



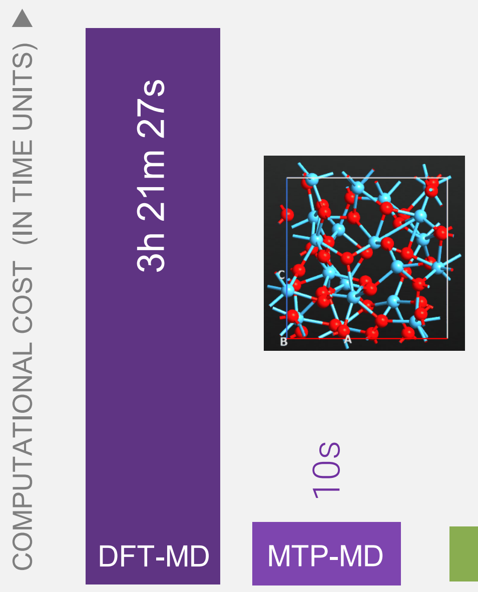
<u>Not practical</u> with quantum-based methods (TAT) or traditional force fields (QoR)

- Structure generation for further DFT studies
- Mechanical properties (fracture)
- Thermal properties
- Electron-phonon scattering
- Diffusion
- Process simulations

Goal of Machine-Learned Force Fieds



· Ab initio level accuracy
· Low computational cost
· Mathematically inspired
· Flexible



· am-HfO_2
· 96 atoms
· 50 MD steps
· 16 cores

3.5 Key Ingredients to Be Successful with ML (in General)

1. Smart generation of training data
2. Effective training & retraining when more data is added
3. Robust validation protocols

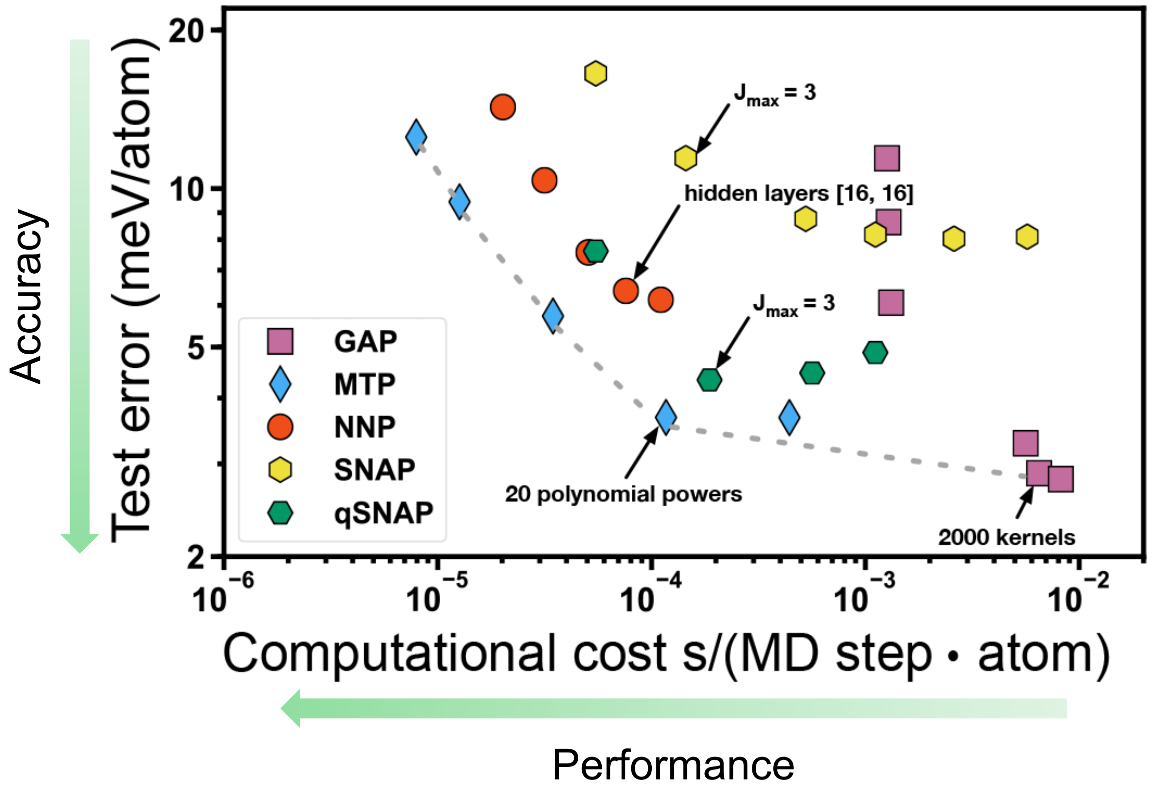
3.5 Efficient execution

- Solve realistic problems
- Make ML FFs easy and efficient

MTP

A Few Details on Moment Tensor Potentials (MTPs)

MTP Is One of the Most Accurate and Efficient ML Potentials



A. V. Shapeev: "Moment Tensor Potentials: A Class of Systematically Improvable Interatomic Potentials", Multiscale Modeling & Simulation (2016)
Y. Zuo et al.: "A Performance and Cost Assessment of Machine Learning Interatomic Potentials", J. Phys. Chem. A, 124, 731, (2020)

Advantages of MTP

- Ideal balance between efficiency and accuracy
- Natural descriptors for atomistic models
  - Many-body descriptors for effective structure property relationship
- Linear regression model for fitting
  - Fast to evaluate
  - Training data can be increased without performance loss during prediction
- Systematically improvable
- Advantages for multi-element systems
  - Global parameters and element-dependent parameters separated
  - Number of parameters scales favorably with the number of elements

How to Use Automated Training Protocols to Fit ML FFs to Solve Realistic Problems

MTP Training Stages

(1) Choose reference method

Choose reference calculator for the system (LCAO-DFT, plane-wave DFT, DFTB, even other FF)

(2) Generate initial geometries

Use basic protocol to generate initial geometries

(3) MTP Training

- **Batch learning** — Compute training data for initial geometries and train an MTP

or

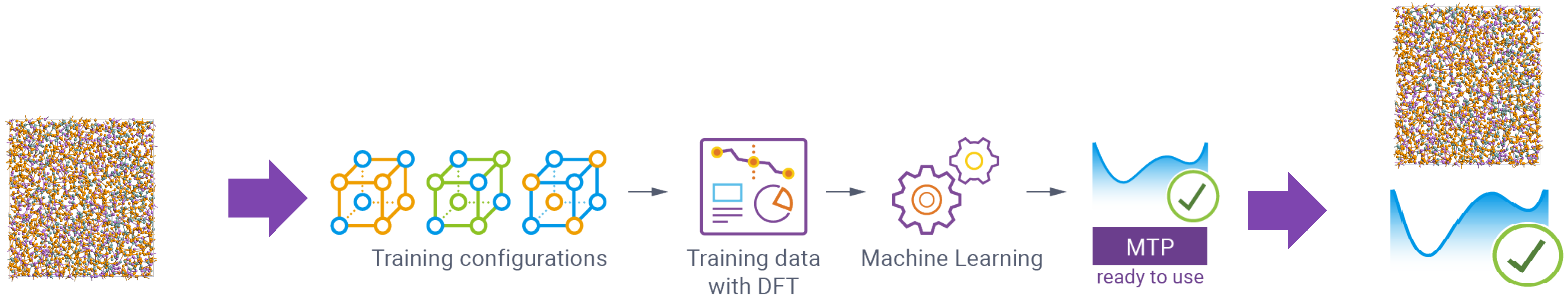
- **Active learning** — Augment training data by dynamically including new atomic environments while running validation MD

(4) MTP tuning (optional)

Optimize hyper parameters – non-linear coefficients, cutoff radii and number of basis functions

Batch Learning

<u>Pre-defined basic training protocols</u>
- Crystals and crystal-like materials
- Interfaces
- Alloys
- Surface processes (molecules)



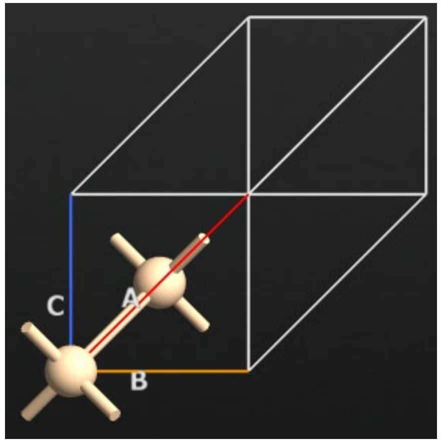
Select specific material or interface

Use Machine Learning to generate a
Force Field called a Moment Tensor Potential (MTP)

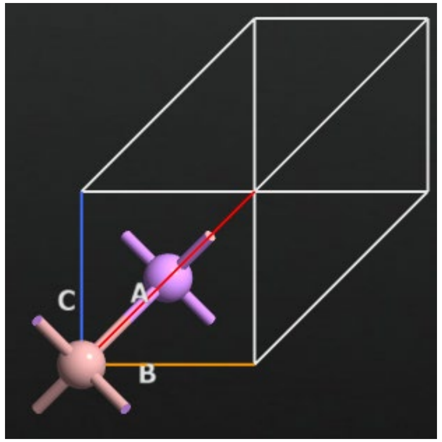
Production dynamical simulations for simple cases

Automatic Interface Geometry Generation

Bulk Si



Bulk GaAs



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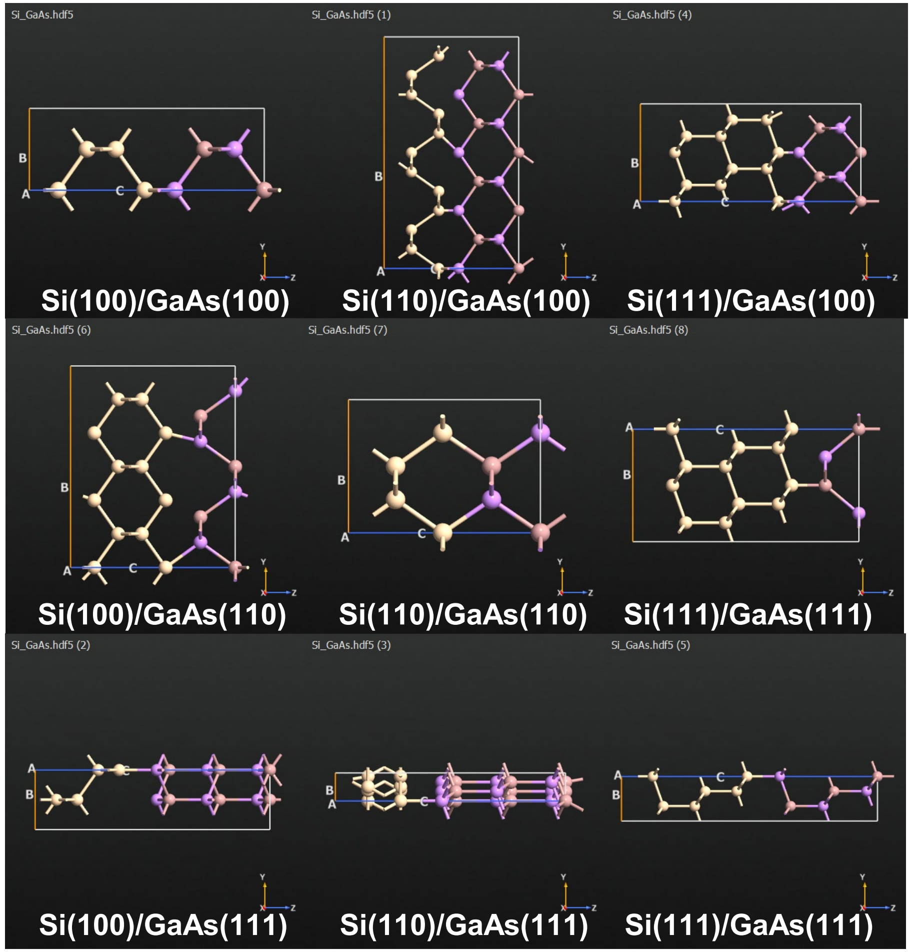
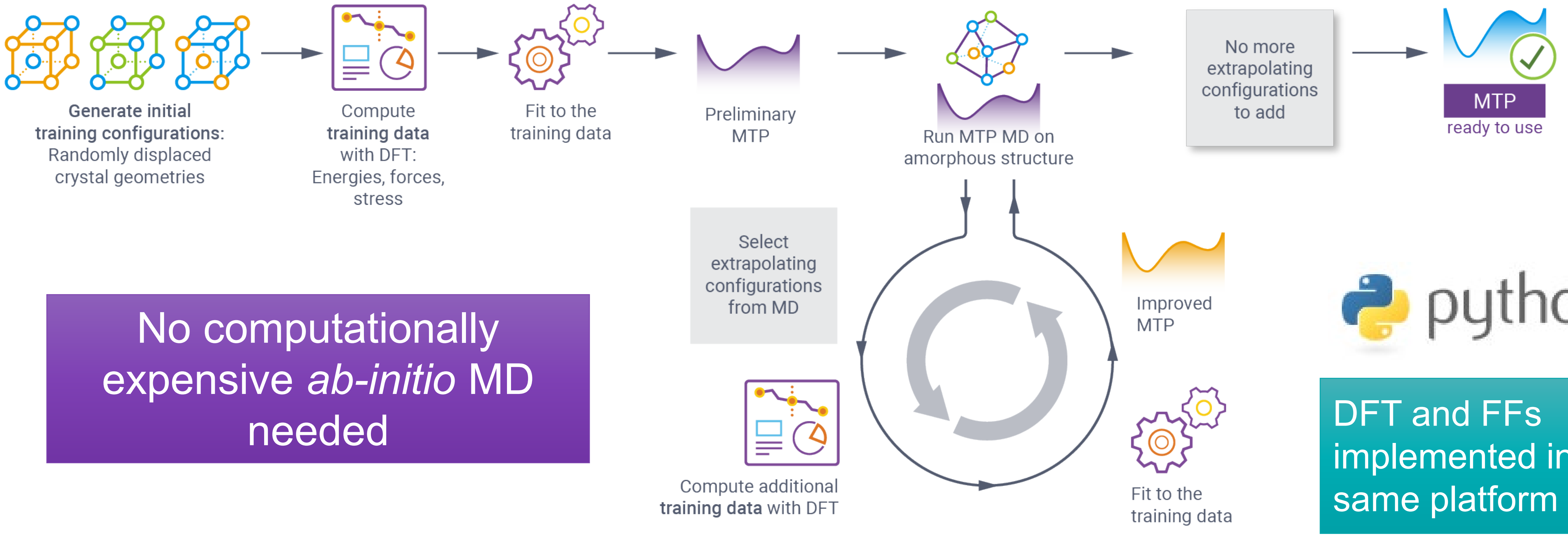


Active Learning

Active Learning MD recommended:
- Amorphous systems
- Interfaces
- High-temperature
- Surface processes

Initial training dataset is improved on-the-fly by actively adding **missing training configurations** and **DFT training data** during MD, meta-dynamics, force-bias Monte Carlo or NEB simulations



No computationally expensive *ab-initio* MD needed

python

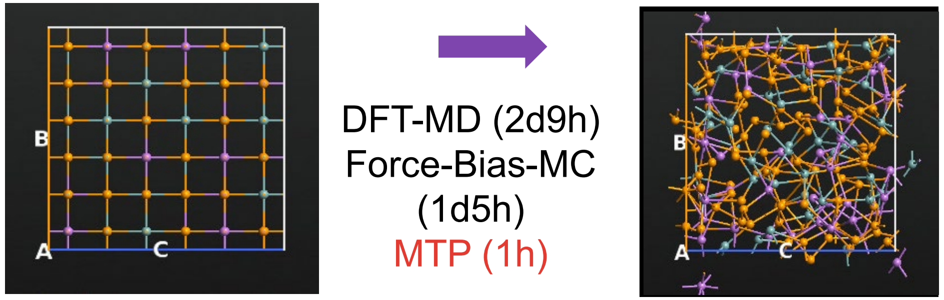
DFT and FFs implemented in the same platform

Example Applications of ML FFs (MTPs) for Complex Materials and Interfaces

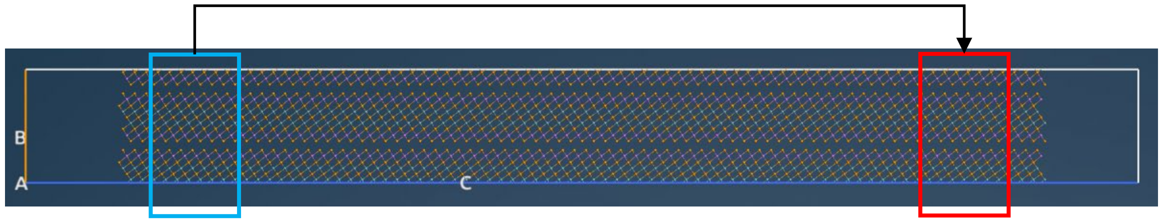
Thermal Transport in Ge-Sb-Te Phase-Change Materials

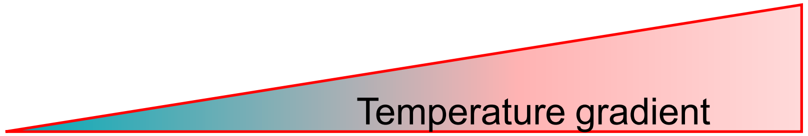
No Conventional Force Fields Exist for GST Materials

Fast amorphization of GST



c-$Ge_2Sb_2Te_5$ thermal transport simulation using reverse non-equilibrium MD and ML-MTP





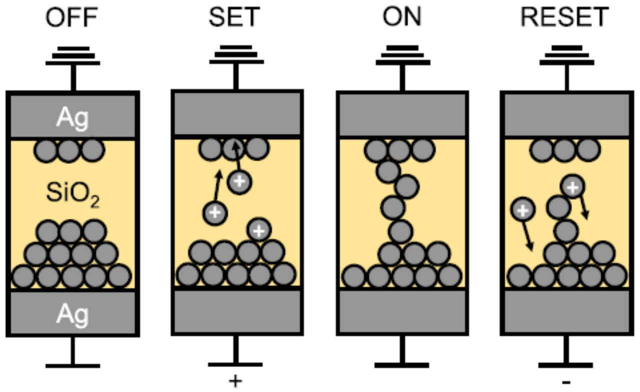
- Accurate thermal conductivities.
- Fast alternative to expensive simulations based on Boltzmann transport equation.

$k_{in\text{-}plane}$ = 1.44 W / K / m

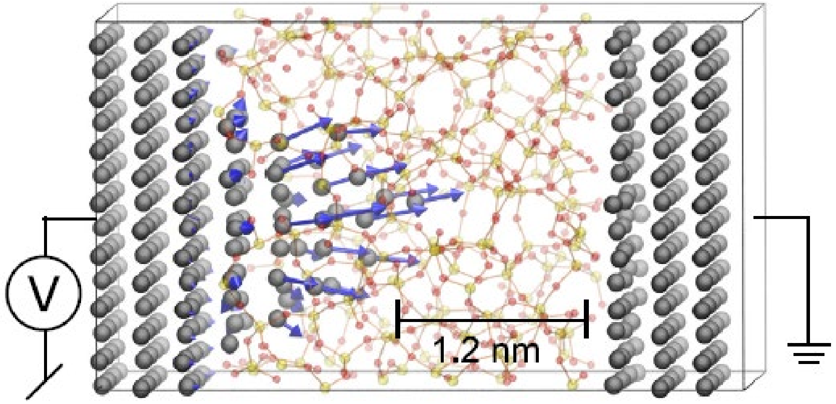
$k_{out\text{-}of\text{-}plane}$ = 0.35 W / K /m

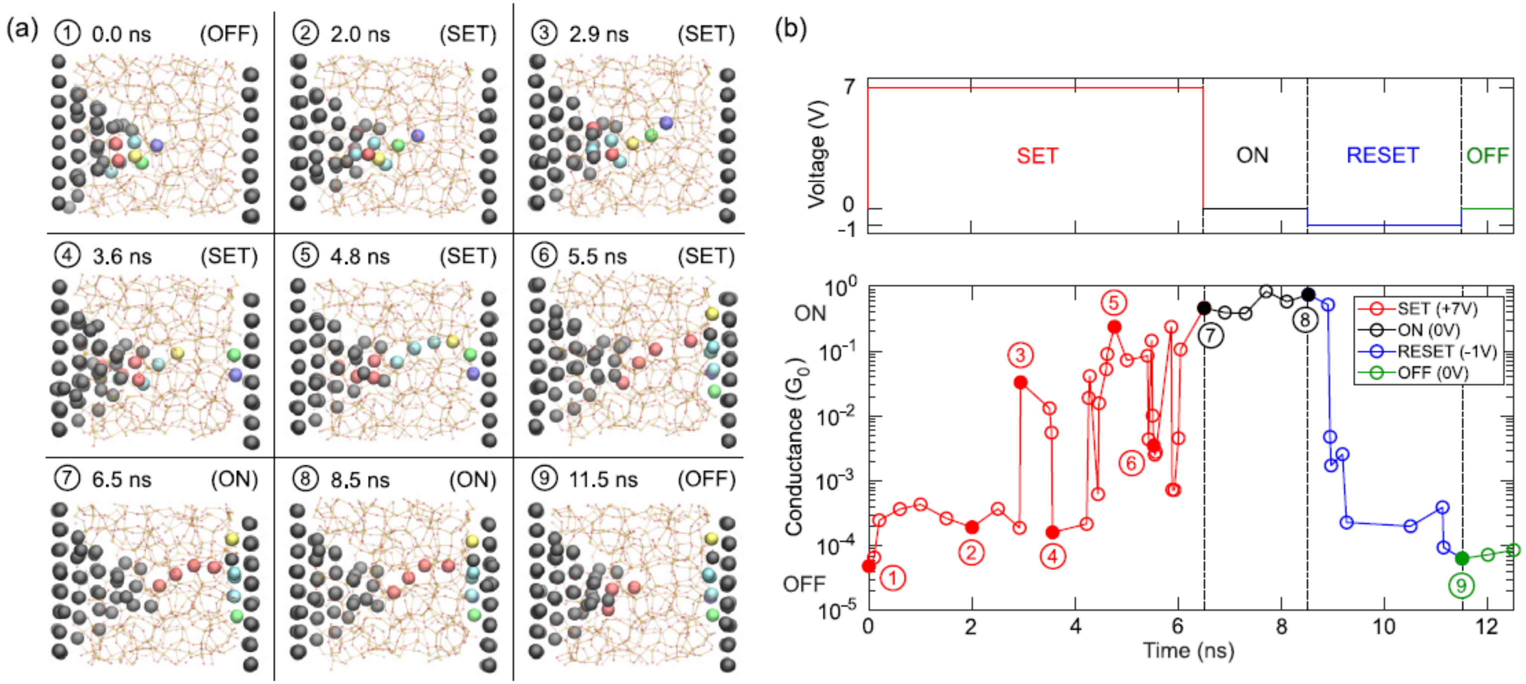
	Exact			SMA		
	κ_z	κ_x	κ_{av}	κ_z	κ_x	κ_{av}
Kooi	0.34	1.59	1.20	0.34	1.51	1.12
Petrov	0.59	2.10	1.60	0.58	2.00	1.53

Structural Changes under E-field in CBRAM (Ag/a-SiO_2)



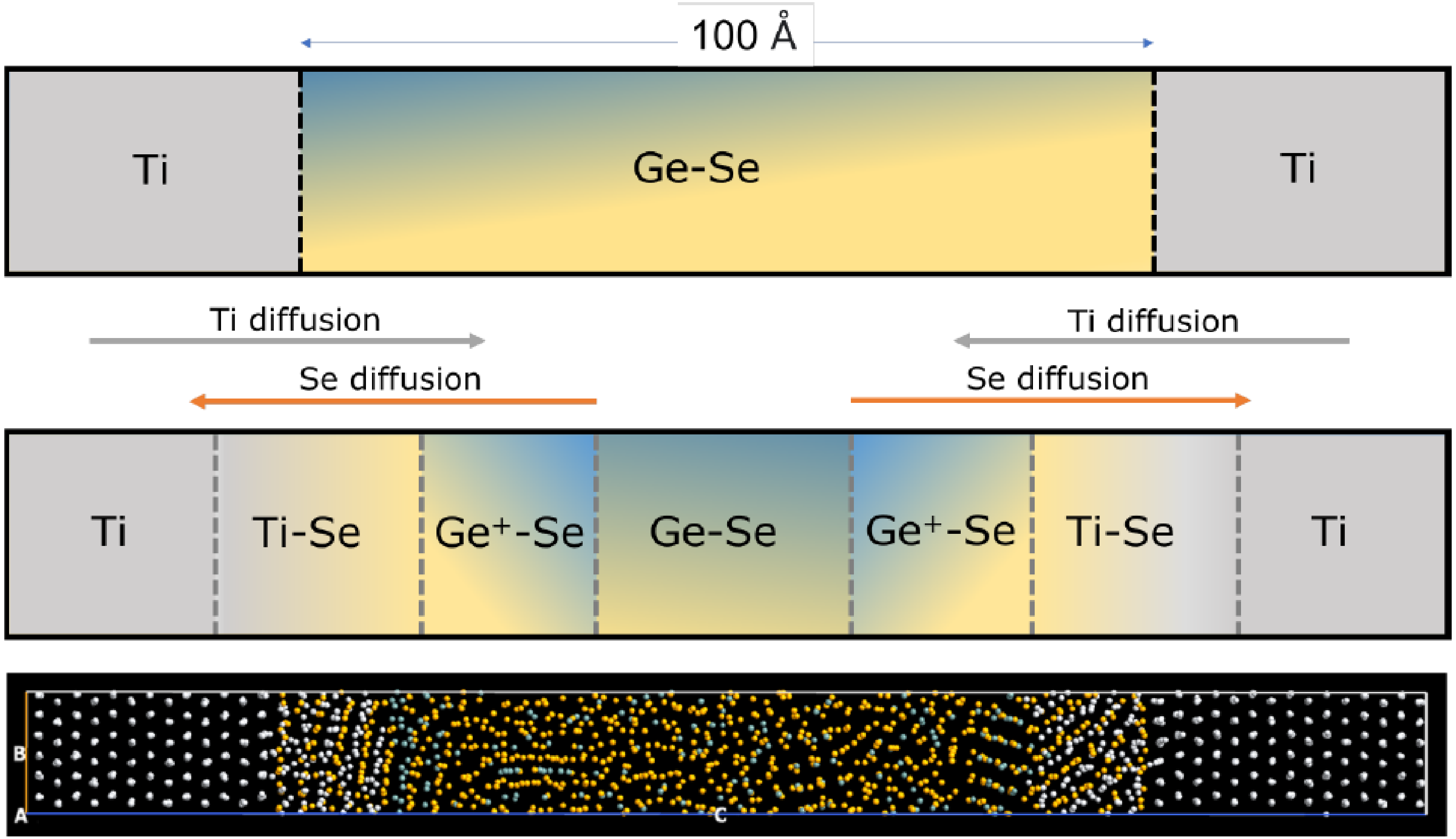
MD-FF simulation with applied E-field drives structural changes which in turn influence the conductance





J. Aeschlimann et al. Solid State Electronics 199 (2023) 108493

Interdiffusion at Metal−Chalcogenide Interfaces



S. K. Achar et al., ACS Appl. Mater. Interfaces (2022)

Automatic Interface Training Tool

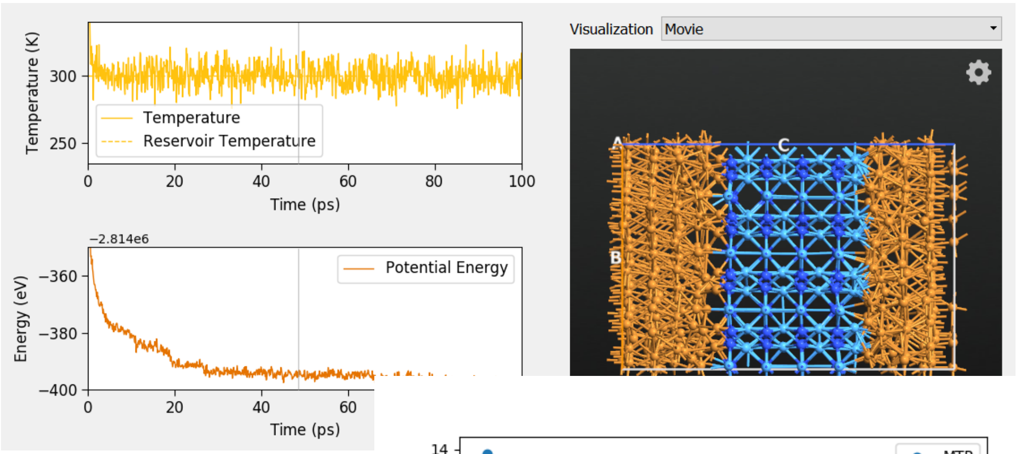
Cu/TaN

- Automatic set up of all possible interface combination between low index surfaces.
- Different terminations (Ta, N) considered.
- Optimize each interface configuration.
- Apply random displacements of different magnitude to sample different energies and forces for the optimized interfaces.

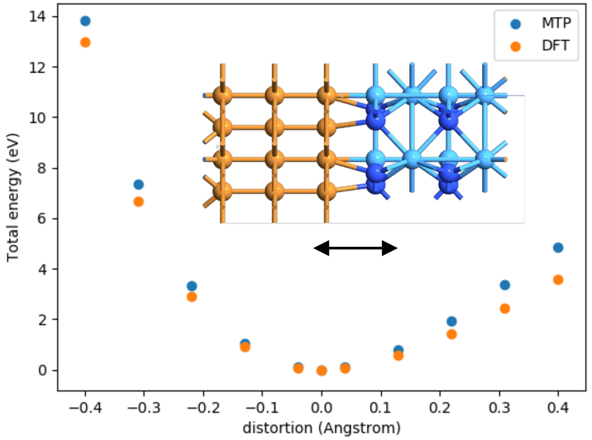
- Training errors:

RMSE	Energy/atom (eV)	Force (eV/Å)	Stress (eV/Å^3)
Training	0.0249859393	0.4436314418	0.0484427052
Testing	0.0264453277	0.4638423203	0.0486505346

- MD is stable at 300K for an interface manually generated in the interface builder.



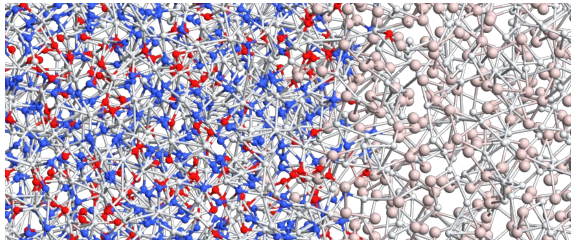
Separation energies are well reproduced.



Diffusion across Ti-Based Amorphous Interfaces

Training MTP for Aluminum Diffusion in TiN-based Materials

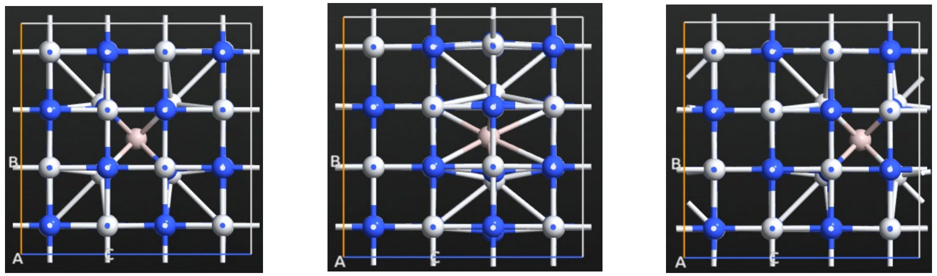
MTP for amorphous interfaces between Ti-based materials (4 elements):

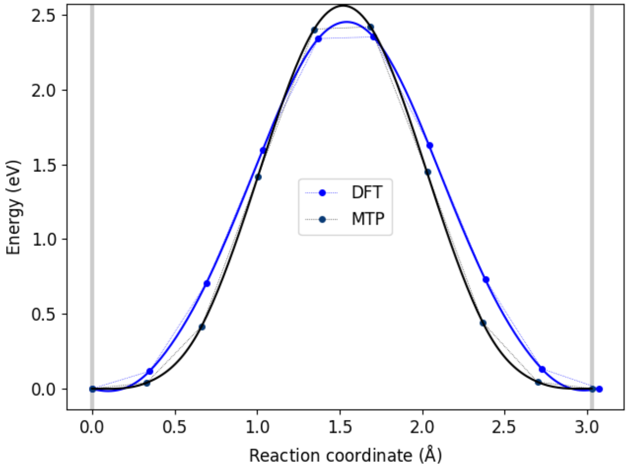


Training data consists of:

- Displaced crystal structures of different compositions
- Active learning of amoprhous materials of different composition.
- No explicit interface configurations in the training data.

NEB of Aluminum diffusion in c-TiN:



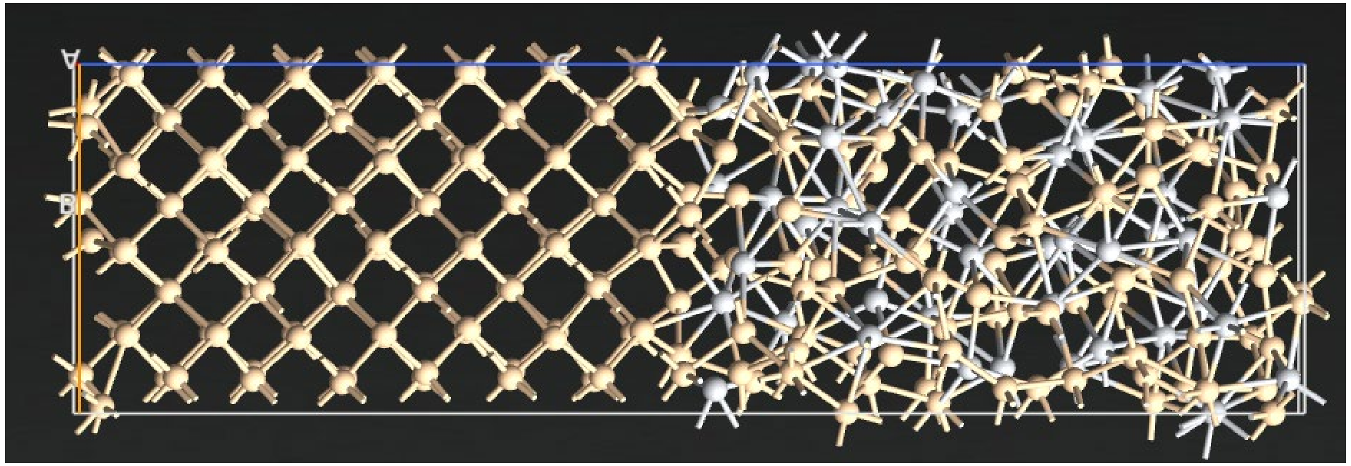


DFT barrier well reproduced, without explicitly training to diffusion events.

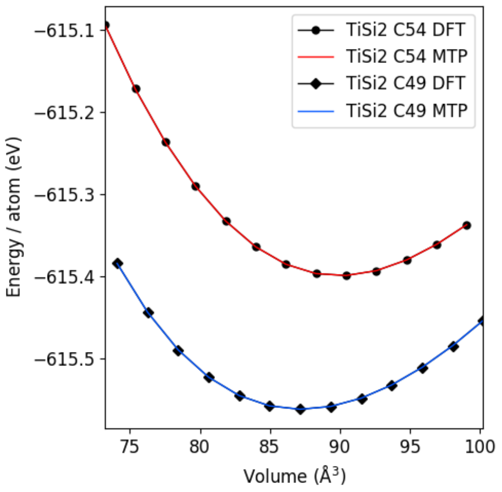
Structure of Ti / Si / TiSi Interfaces

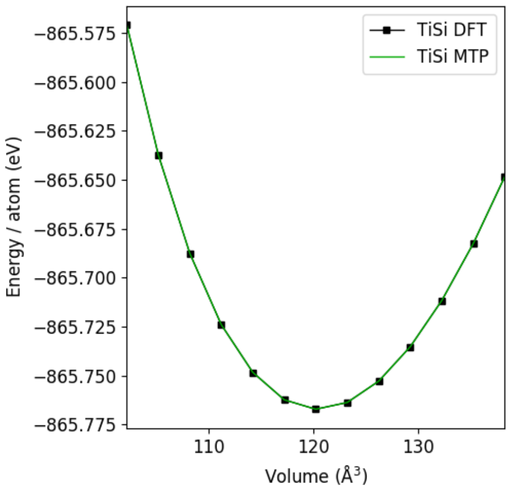
- Enables simulations of interfaces between Si / Ti and crystalline, poly-crystalline, or amorphous TiSi.

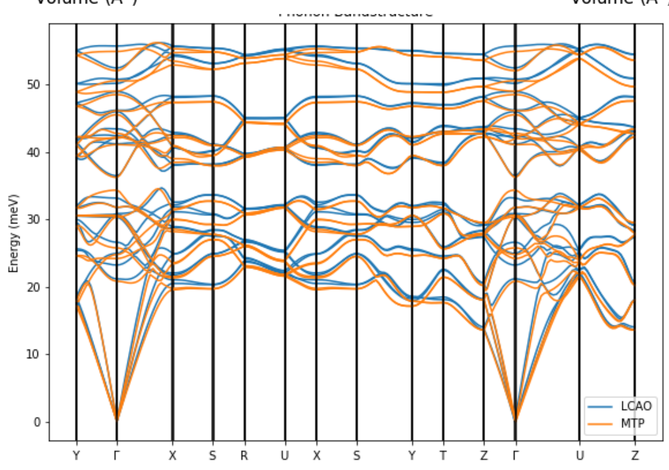
Crystalline Si Amorphous TiSi

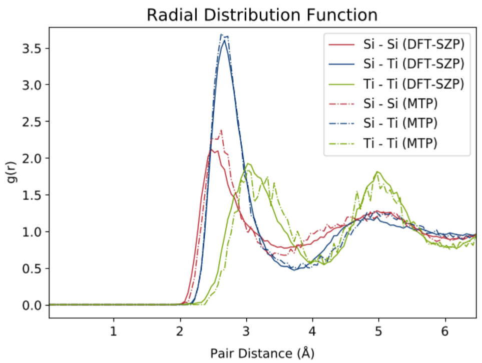


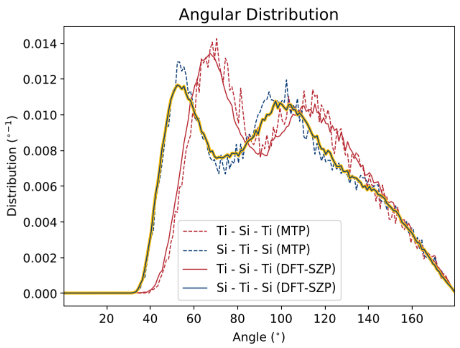
- Quicky generate realistic interface configurations, e.g. for DFT contact resistance calculations.
- Simulate Si / Ti interface, towards interdiffusion and onset of silicidation







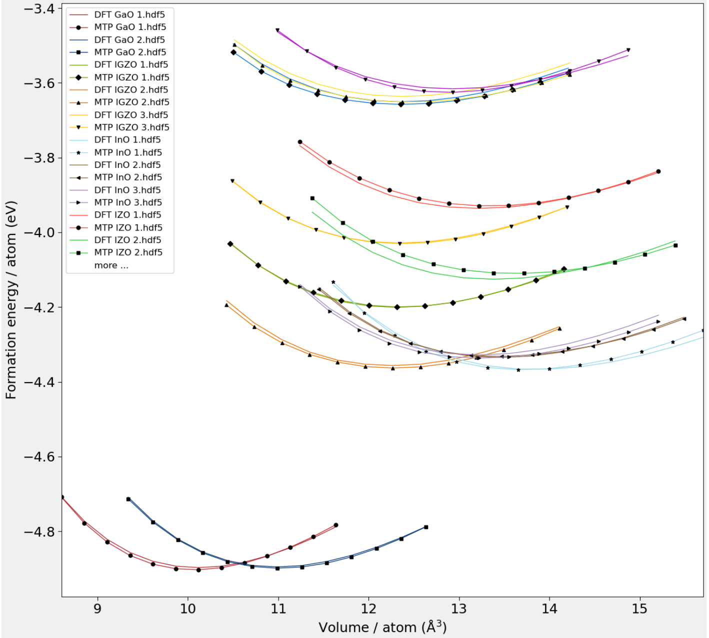




Structure of IGZO Materials

Crystal:

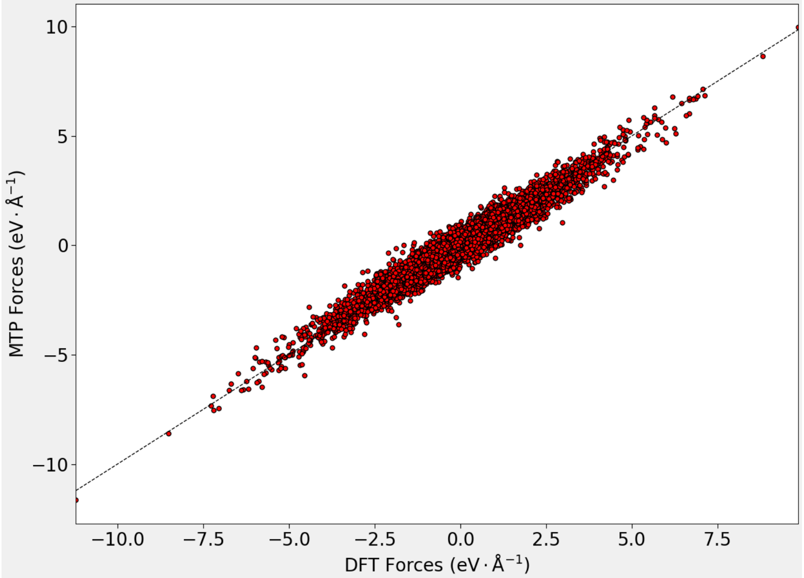
- Optimize all crystal structures with DFT and MTP and compare lattice constants:
  - Max. Deviation 2.5 %, most crystals have below 1% deviation in lattice constants.
- Calculate equation-of-state (EOS) with DFT and MTP:



✓ All crystals very well reproduced

Amorphous:

- Forces scatterplot for melt-quench of am. ZnO, InZnO, IGZO with MTP:



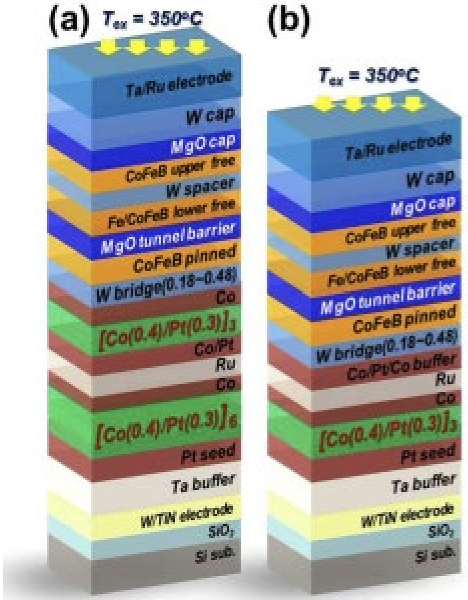
Energy RMSE 20 meV / Atom
Forces RMSE 0.35 eV / Ang

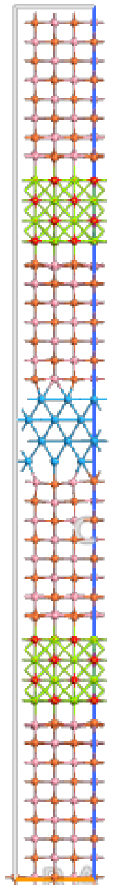
✓ Only slightly larger than training error due to many out-of-equilibrium configurations.

Interfaces between Multiple Layers in Magnetic Tunneling Junctions (MTJs) for MRAM Applications

3 different layers with Delta-MTP:

- MgO / FeCo / W:



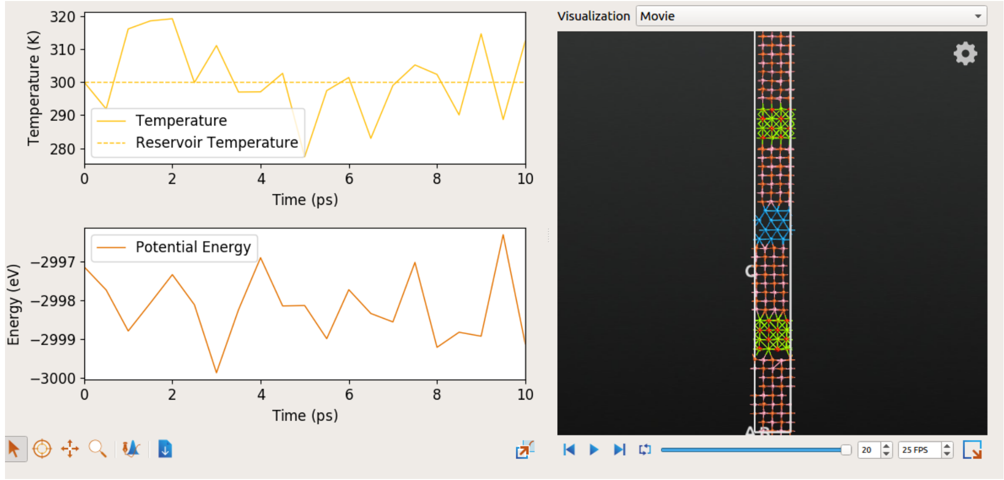


Delta-MTP: A | B

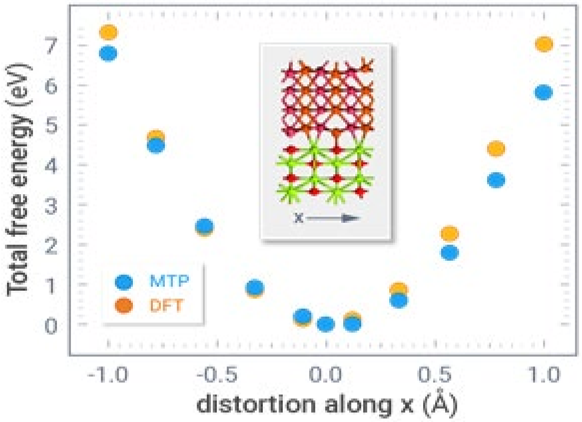
A + B + A | B

MTP_A MTP_B $MTP_{A/B}$

Fast and robust optimization and MD:

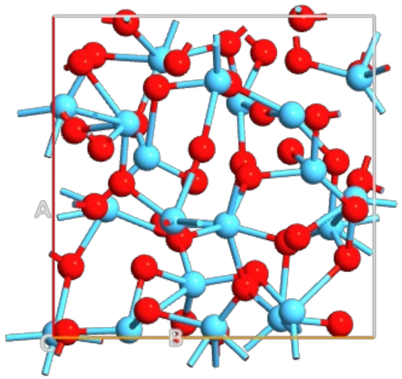


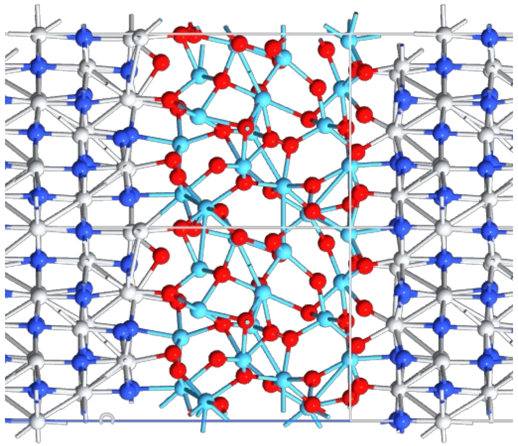
Interface mechanical properties accurately reproduced compared to DFT:

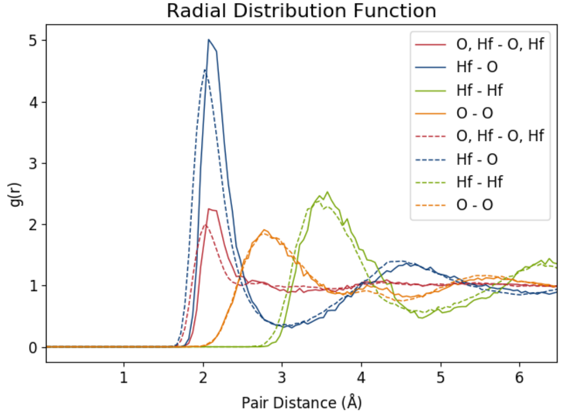


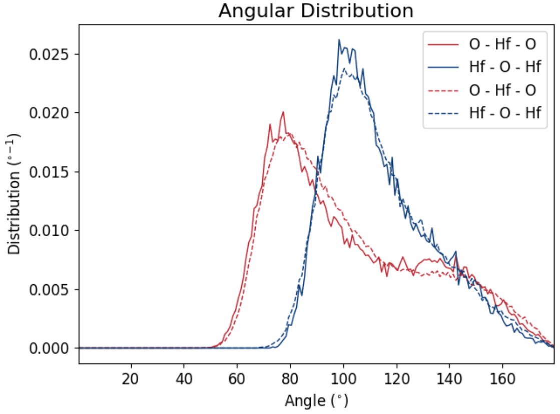
Structure of HfO_2 / TiN Interface and Glasses

MTP for crystal and amorphous HfO_2 with excellent reproduction of structural properties:

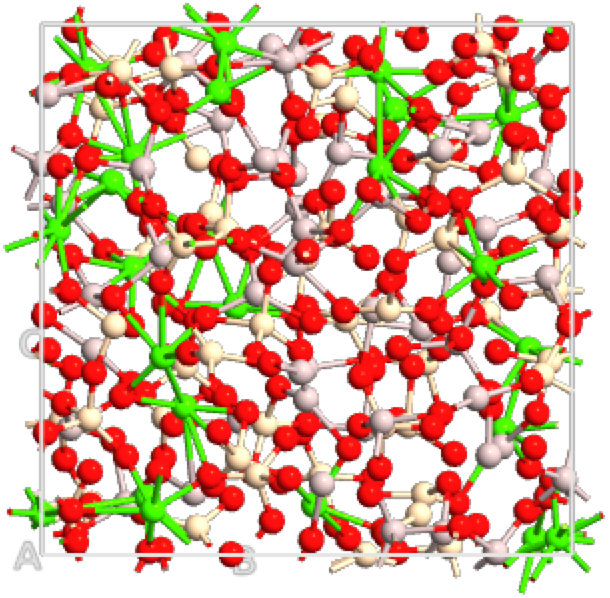








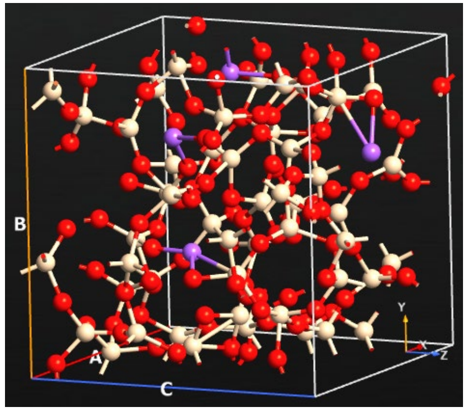
MTP for Calcium-Aluminum-Silicate glass:



- More accurate than the common conventional FFs for these materials

Structure of Sodium Silicate Glass with a Few Na Atoms

- Large-scale MTP-MDs of $(Na_2O)_2(SiO_2)_{40000}$, i.e., sodium silicate glass only containing a few Na atoms
- Train MTP by active-learning MDs of $(Na_2O)_2(SiO_2)_{50}$ (Fig. 1) → MTP-MDs of $(Na_2O)_2(SiO_2)_{40000}$
- **Results**: RDFs and ADFs obtained with MTP and FF [1] based MDs are in good agreement (Fig. 2 (a) and (b)).
- **Conclusion:** Active-learning MDs enable to train MTP applicable to large-scale MDs of glass containing a few impurity atoms



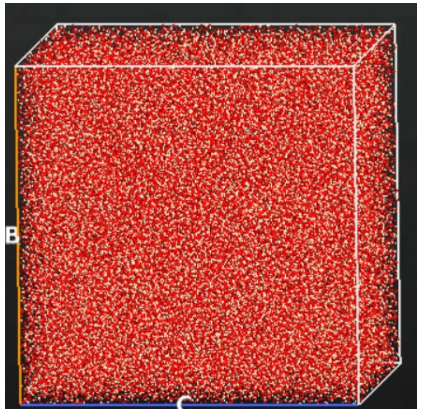
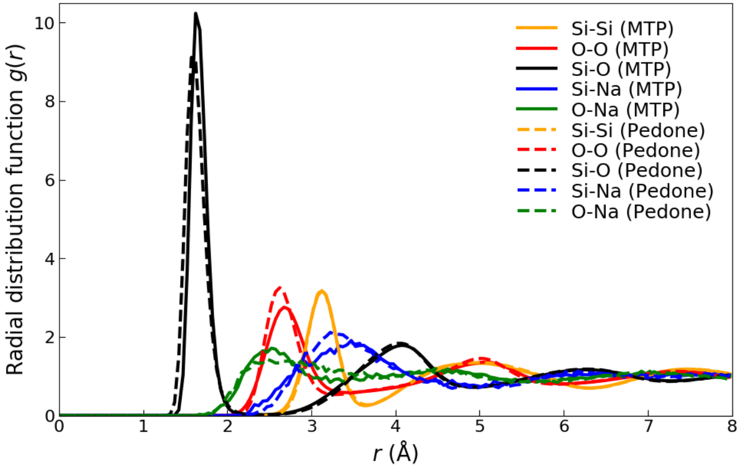


Fig.1: Systems of sodium silicate glass:
(a) $(Na_2O)_2(SiO_2)_{50}$ used to train MTP by active-learning MDs
(b) $(Na_2O)_2(SiO_2)_{40000}$ to which the trained MTP was applied



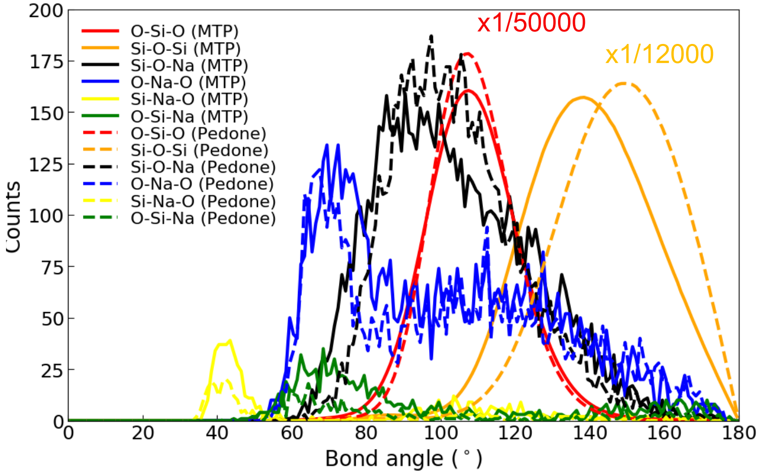


Fig. 2: Results of MTP-MD (at T=2500 K)
(a) RDF (Comparison with the results obtained using the Pedone potential)
(b) ADF (Comparison with the results obtained using the Pedone potential)

[1] A. Pedone *et al.*, Chem. Mater. **19**, 3144 (2007).

Summary

- An integrated Python-based platform combining DFT, force fields and ML algorithms extend the applicability of atomistic modeling
  - ML potentials can be used for MD, meta-dynamics, phonons, crystal structure prediction
  - Well trained ML FFs can even be accurate for reactions (NEB)
- By employing well-crafted protocols, ML FFs can be trained efficiently and robustly
  - Application-specific generation of small but relevant set of initial training structures
  - Active learning for difficult situations like interfaces and amorphous structures
- Demonstrated application examples for complex interfaces and multi-element structures
  - Advanced features like ZBL correction, dispersion corrections (D3, D4)

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Thank You

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