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 Fields?

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.

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

Not practical with quantum-based methods (TAT) or traditional force fields (QoR)
Goal of Machine-Learned Force Fields

- **Ab initio level accuracy**
- **Low computational cost**
- **Mathematically inspired**
- **Flexible**

**Traditional Force Fields**

**Ab Initio (DFT)**

**Semi-Empirical**

**Machine-Learned Force Fields (ML-FF)**

**Computational Cost (in time units):**
- DFT-MD: 3h21m27s
- MTP-MD: 10s
- FF-MD: 9s

- am-HfO₂
- 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

MTP

- Solve realistic problems
- Make ML FFs easy and efficient
A Few Details on Moment Tensor Potentials (MTPs)
MTP Is One of the Most Accurate and Efficient ML Potentials

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
- Active learning
Compute training data for initial geometries and train an MTP
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

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

Select specific material or interface

Training configurations → Training data with DFT → Machine Learning

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 =

- Si(100)/GaAs(100)
- Si(110)/GaAs(100)
- Si(111)/GaAs(100)
- Si(100)/GaAs(110)
- Si(110)/GaAs(110)
- Si(111)/GaAs(110)
- Si(100)/GaAs(111)
- Si(110)/GaAs(111)
- Si(111)/GaAs(111)
Active Learning

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.

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

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

$c$-Ge$_2$Sb$_2$Te$_5$ thermal transport simulation using reverse non-equilibrium MD and ML-MTP

<table>
<thead>
<tr>
<th>Crystalline GST</th>
<th>Amorphous GST</th>
</tr>
</thead>
<tbody>
<tr>
<td>DFT-MD (2d9h)</td>
<td>Force-Bias-MC (1d5h)</td>
</tr>
<tr>
<td>MTP (1h)</td>
<td></td>
</tr>
</tbody>
</table>

Heat sink

Heat source

Temperature gradient

**Accurate thermal conductivities.**

- $k_{\text{in-plane}} = 1.44 \text{ W} / \text{K} / \text{m}$
- $k_{\text{out-of-plane}} = 0.35 \text{ W} / \text{K} / \text{m}$

<table>
<thead>
<tr>
<th>Method</th>
<th>$k_{\text{out-of-plane}}$</th>
<th>$k_{\text{in-plane}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kooi</td>
<td>0.34</td>
<td>1.59</td>
</tr>
<tr>
<td>Persson</td>
<td>0.59</td>
<td>2.10</td>
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</table>

<table>
<thead>
<tr>
<th>Method</th>
<th>$\kappa_{\text{eff}}$</th>
<th>$\kappa_{\text{c}}$</th>
<th>$\kappa_{\text{s}}$</th>
<th>$\kappa_{\text{f}}$</th>
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<tbody>
<tr>
<td>Exact</td>
<td>1.20</td>
<td>0.34</td>
<td>1.51</td>
<td>1.12</td>
</tr>
<tr>
<td>SMA</td>
<td>1.60</td>
<td>0.58</td>
<td>2.00</td>
<td>1.53</td>
</tr>
</tbody>
</table>
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:

<table>
<thead>
<tr>
<th>RMSE</th>
<th>Energy/atom (eV)</th>
<th>Force (eV/Å)</th>
<th>Stress (eV/Å^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Training</td>
<td>0.0249859393</td>
<td>0.4436314418</td>
<td>0.0484427052</td>
</tr>
<tr>
<td>Testing</td>
<td>0.0264453277</td>
<td>0.4638423203</td>
<td>0.0486505346</td>
</tr>
</tbody>
</table>

- 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 amorphous 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.
- Quickly 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:
  - 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.

✓ All crystals very well reproduced
Interfaces between Multiple Layers in Magnetic Tunneling Junctions (MTJs) for MRAM Applications

3 different layers with Delta-MTP:
- MgO / FeCo / W:
  - 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 $(\text{Na}_2\text{O})_2(\text{SiO}_2)_{40000}$, i.e., sodium silicate glass only containing a few Na atoms
- Train MTP by active-learning MDs of $(\text{Na}_2\text{O})_2(\text{SiO}_2)_{50}$ (Fig. 1) $\rightarrow$ MTP-MDs of $(\text{Na}_2\text{O})_2(\text{SiO}_2)_{40000}$
- **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) $(\text{Na}_2\text{O})_2(\text{SiO}_2)_{50}$ used to train MTP by active-learning MDs
(b) $(\text{Na}_2\text{O})_2(\text{SiO}_2)_{40000}$ 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)

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