

Accelerating Grain-Boundary Calculations in Solid Electrolytes with Fine-Tuned Machine-Learning Interatomic Potentials



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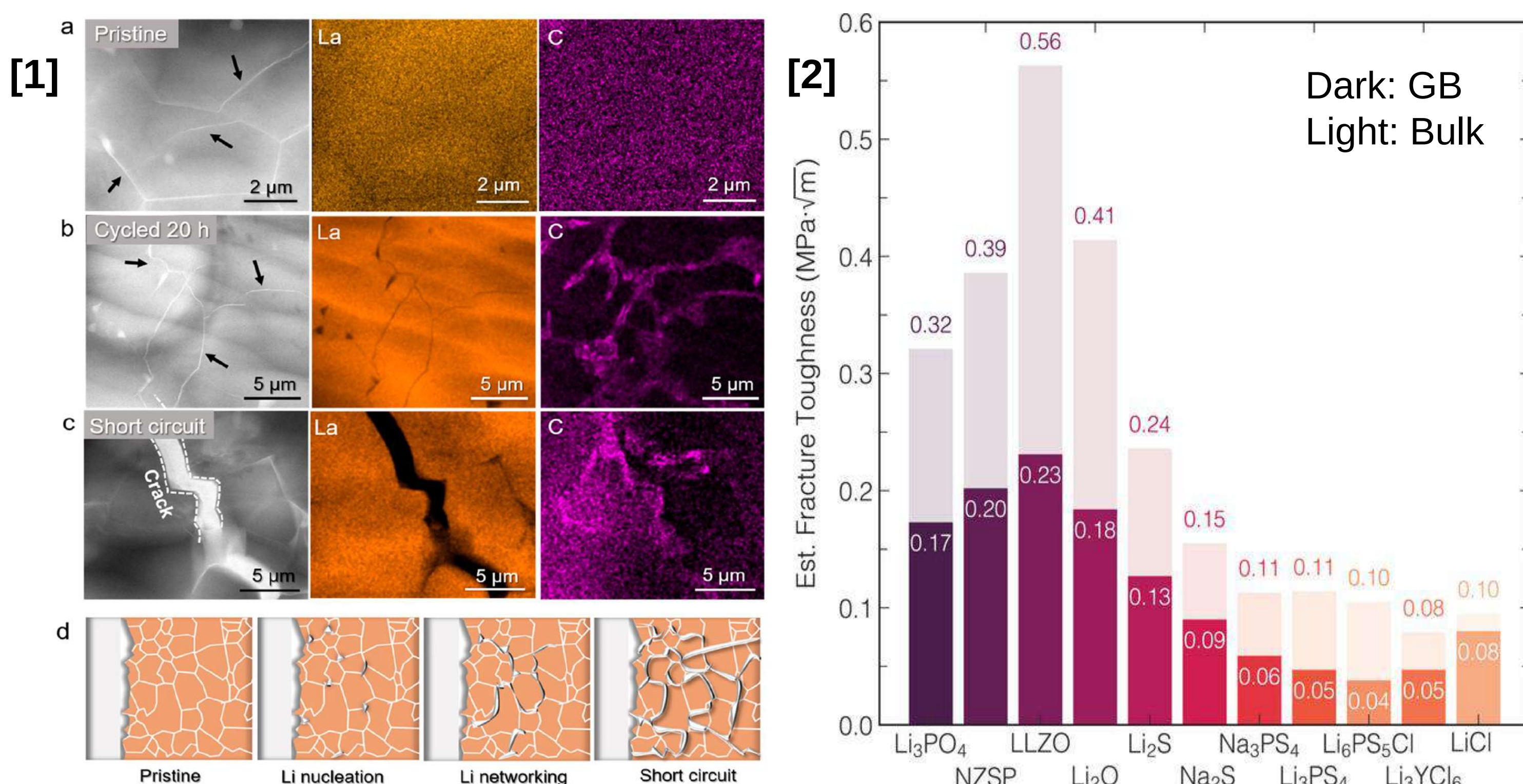


Introduction

All-solid-state batteries offer improved safety and high-energy-density potential. However, ceramic electrolytes are generally polycrystalline and inevitably contain grain boundaries, whose distinct local structures may influence mechanical stability. In LLTO, Li/La/vacancy disorder and different grain-boundary types give rise to numerous possible structures, making large-scale first-principles calculations computationally expensive. This study combines first-principles calculations with machine-learning interatomic potentials to efficiently explore LLTO structures, with the ultimate goal of predicting grain-boundary mechanical responses. After validation against first-principles calculations and experimental measurements, this framework can be extended to more complex grain-boundary systems and material modification strategies, such as doping, to explore and optimize LLTO mechanical properties.

How do grain boundaries affect the mechanical stability of LLTO solid electrolytes?

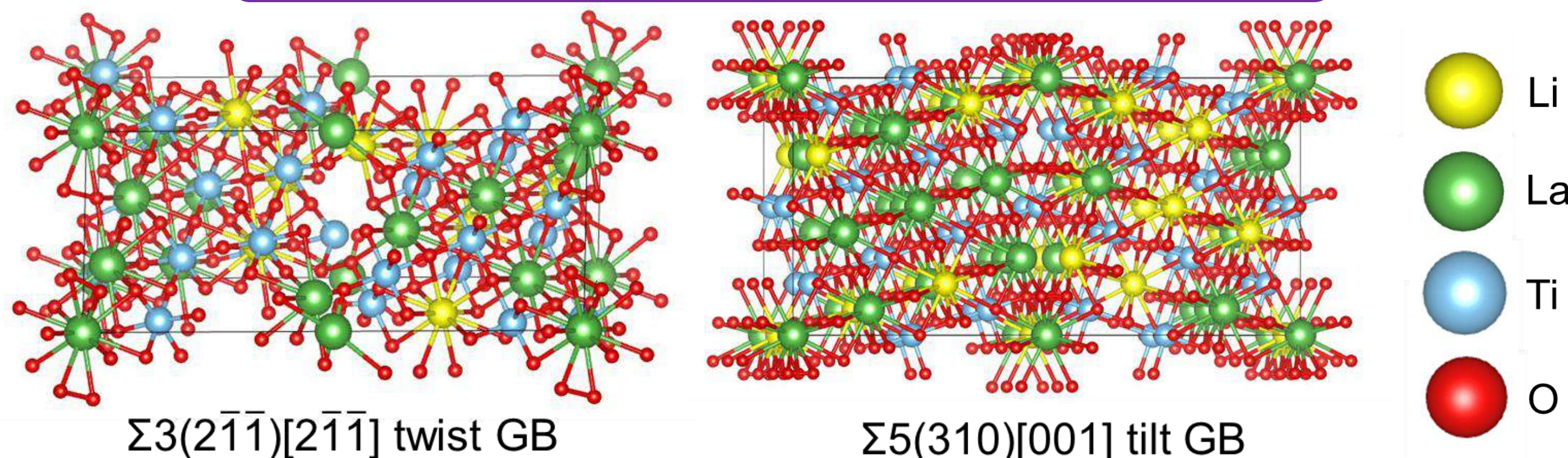
Why investigate GB mechanical properties?



GBs can act as mechanically vulnerable regions in solid electrolytes

How can we efficiently investigate the mechanical response of LLTO grain boundaries?

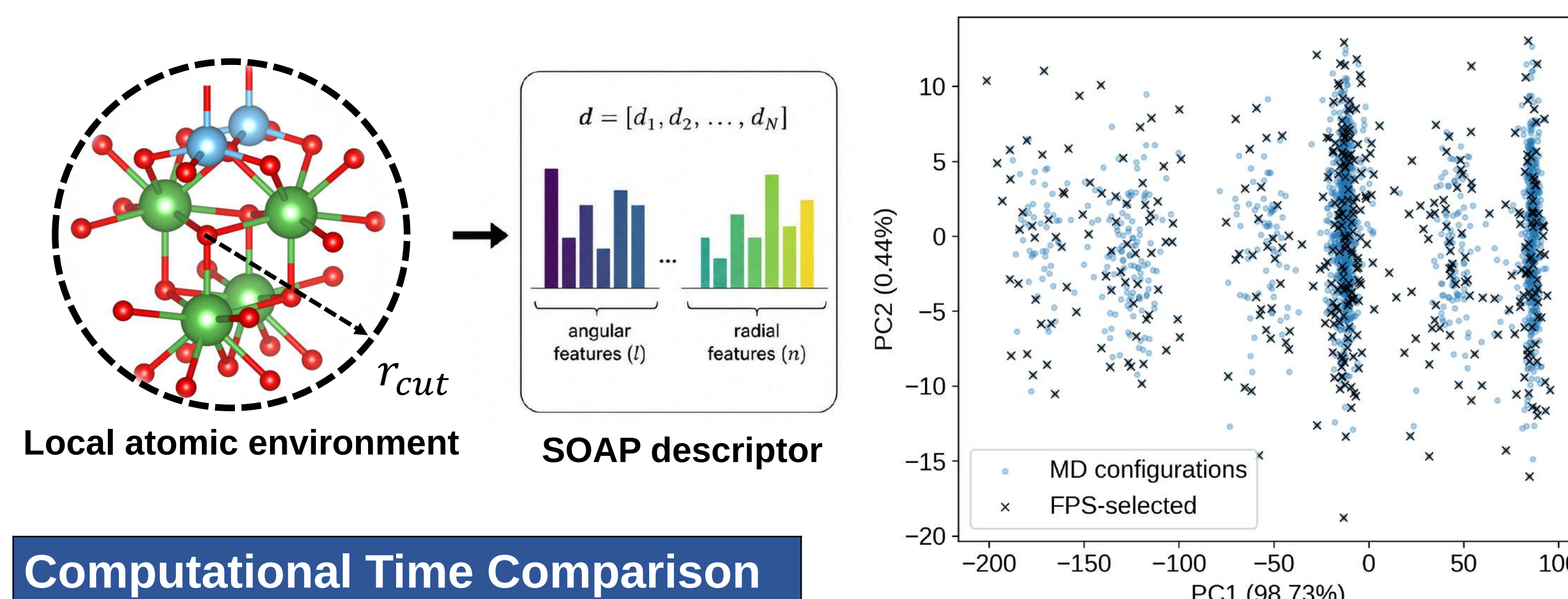
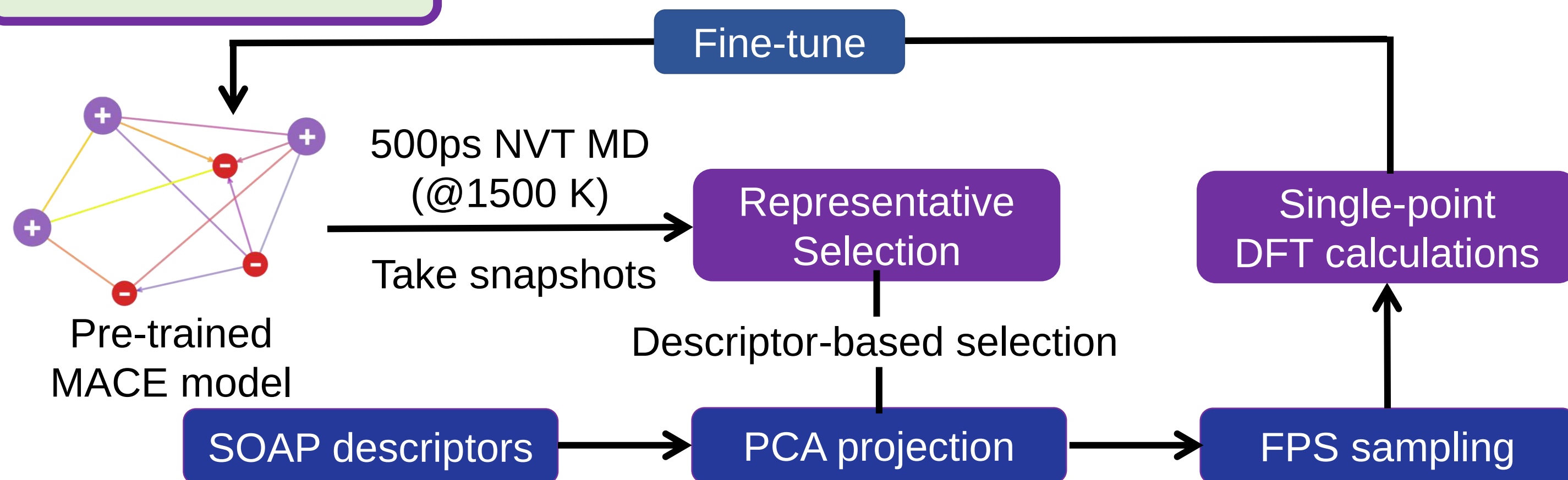
Why use MLIPs for GB mechanics?



Large GB supercells × Multiple GB types → High DFT computational cost

MLIPs enable efficient exploration of large and diverse GB configuration spaces

MLIP workflow



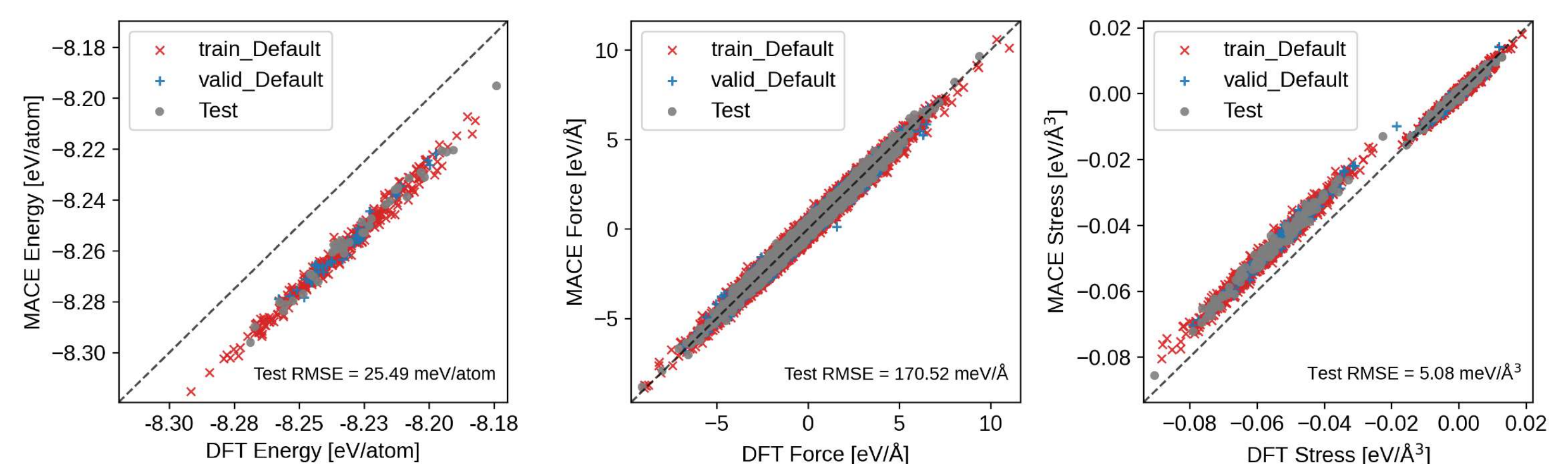
Computational Time Comparison

DFT 14.2hr/config. → 7700x faster → MLIP 6.6s/config.

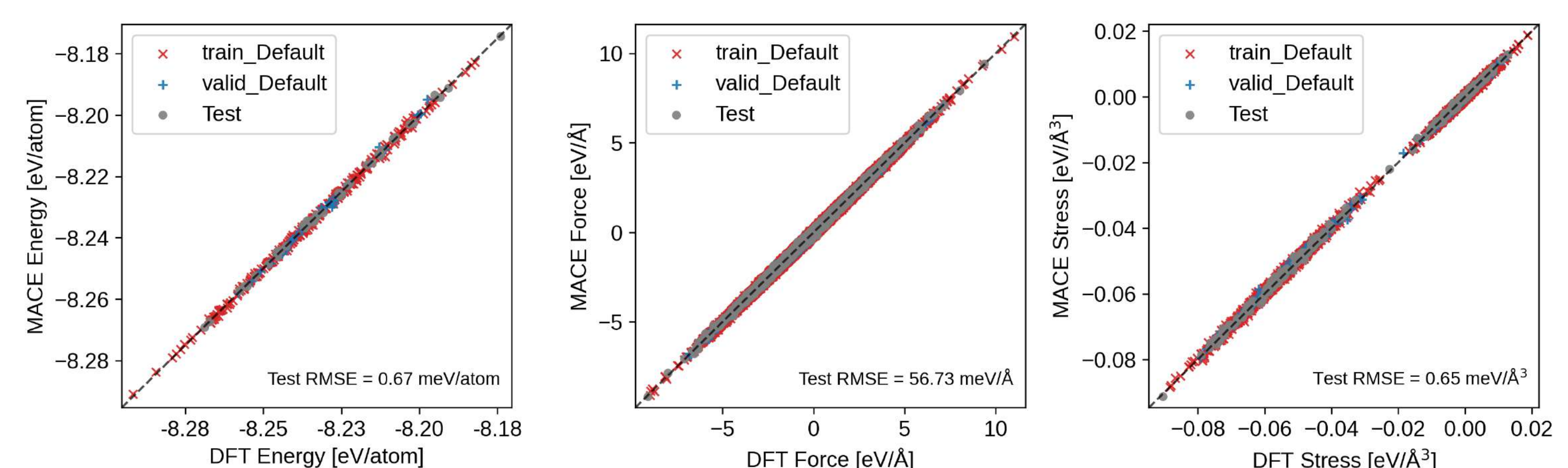
Can MLIPs accurately reproduce DFT calculations?

MLIP fine-tuning and validation

Pre-trained MACE model

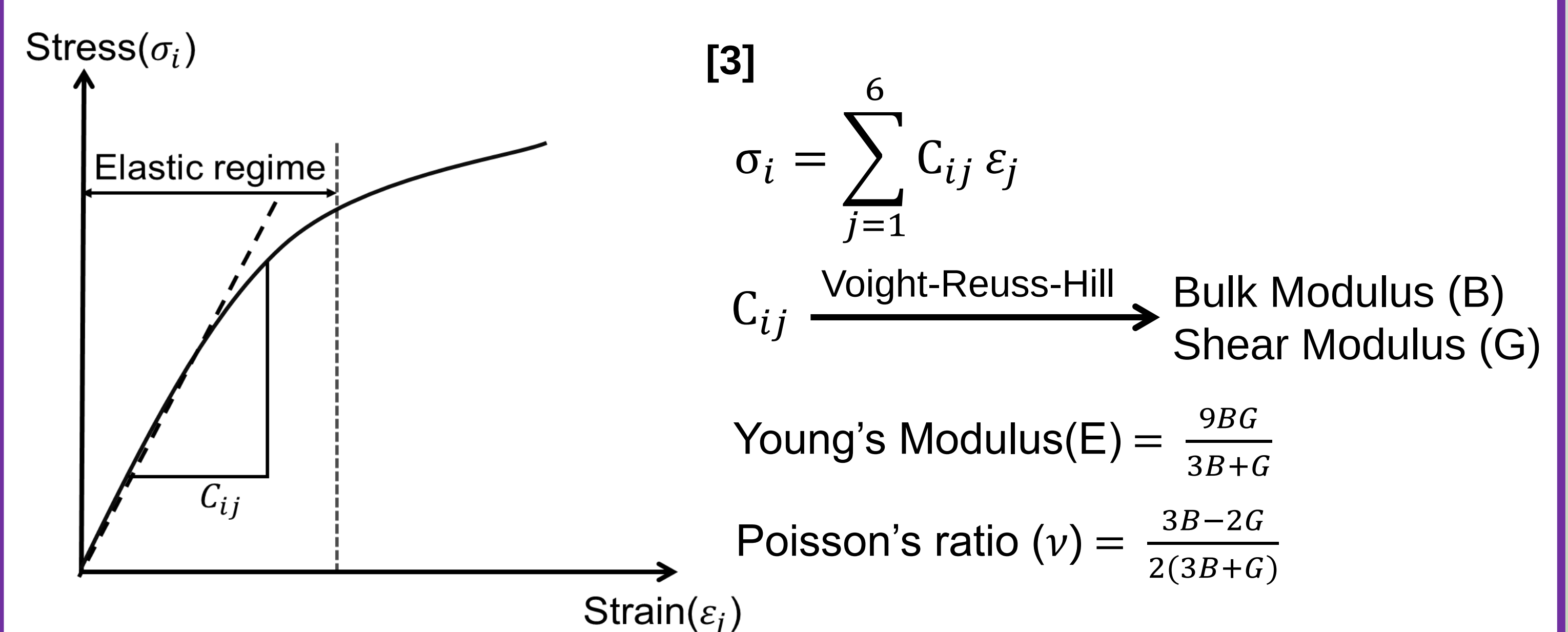


Fine-tune MACE model



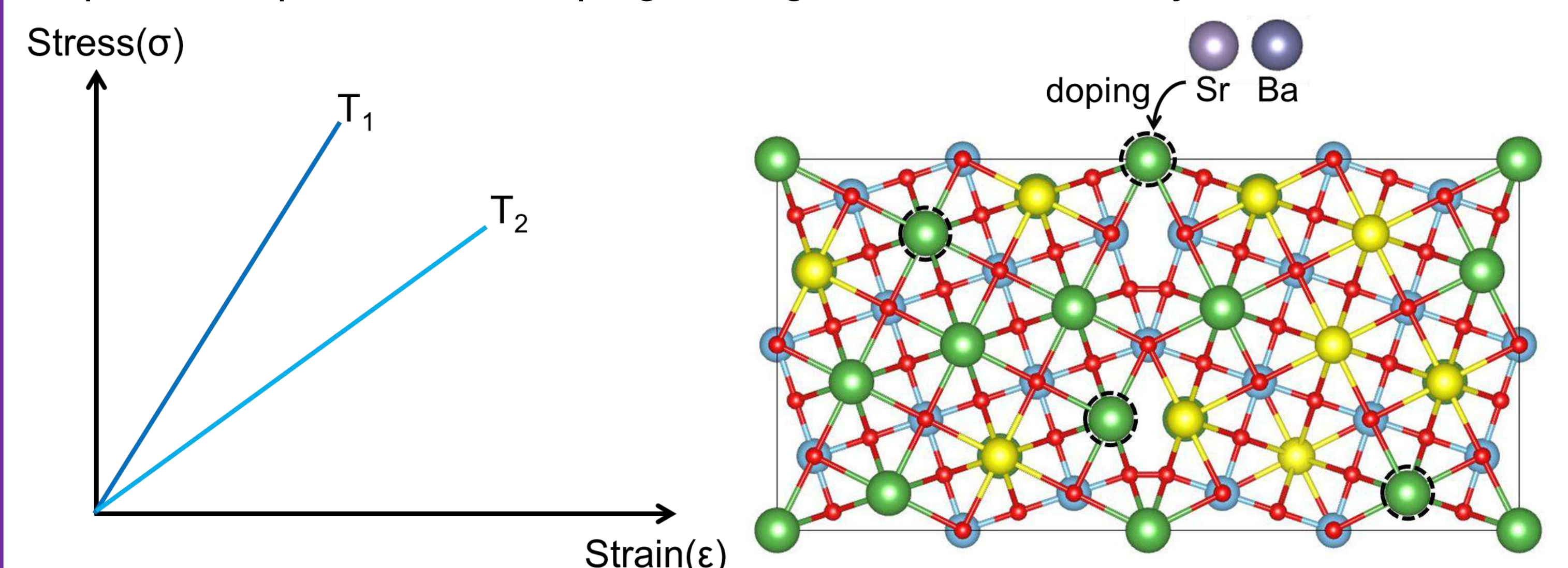
Fine-tuning improves agreement with DFT

From stress-strain response to elastic properties



Next Steps

- **Finite-Temperature Mechanical Properties**
Predict LLTO elastic properties at finite temperatures using the validated MLIP.
- **GB Engineering and Optimization**
Explore composition and doping strategies for mechanically stable GBs.



References

- [1] Xu, Lei, et al. "Structural heterogeneity induced Li dendrite growth in Li_{0.33}La_{0.56}Ti_{0.3}O₃ solid-state electrolytes." *ACS Applied Energy Materials* 5.3 (2022): 3741-3747.
- [2] Xie, Weihang, et al. "Effects of grain boundaries and surfaces on electronic and mechanical properties of solid electrolytes." *Advanced Energy Materials* 14.17 (2024): 2304230.
- [3] Yao, Hongzhi, Lizhi Ouyang, and Wai-Yim Ching. "Ab initio calculation of elastic constants of ceramic crystals." *Journal of the American Ceramic Society* 90.10 (2007): 3194-3204.