Evaluating Machine Learning Workflows for Structural Relaxation and Strained Bandgap Modeling in Monolayer Transition Metal Dichalcogenides

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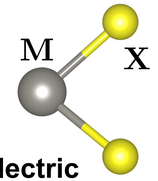
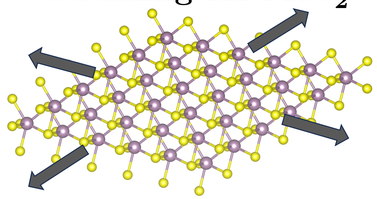
Background

- Transition metal dichalcogenides (TMDs) are a promising class of 2D materials with use cases for novel transistors, optoelectronics, and sensors
  - Monolayer TMDs follow an **MX_2** structure with M={Mo, W}, X={S, Se, Te} and are direct bandgap semiconductors



- TMDs are sensitive to **strain**, **alloying**, and **electric fields**; this allows for precise, continuous modulation of properties like bandgap[1]

Straining 1H MoS_2



VASP

- DFT is **computationally expensive** across TMD materials, large supercells (heterostructures, defects, alloying), and strain types
- ML workflows / MLIPs are a fast and promising DFT surrogate for TMDs! MatPES.ai

Methods

- VASP is used for **PBE** and **HSE06** calculations with consistent input parameters: ENCUT=700eV, 15Å vacuum spacing, and a $2\pi \times 0.03$ Å^{-1} k-space mesh
- Various MLIPs are evaluated with ASE, using a force convergence threshold of $|F_{max}| < 0.001$ eV/Å

Table 1: Summary of machine learning interatomic potentials (MLIPs), their training datasets, and highest level of theory for their training data.

MLIP Name	Training Dataset	Dataset Size	Level of Theory	Parameters
uma-s-1p2-OMat[2]	OMat24[3]	118M	PBE	6.6M/150M
uma-m-1p1-OMat[2]	OMat24[3]	118M	PBE	50M/1.4B
TensorNet-MatPES-PBE[4]	MatPES[4]	434,712	PBE	838,000
TensorNet-MatPES-r2SCAN[4]	MatPES[4]	387,897	r^{2}SCAN	838,000
PET-OMatPES-L[5]	OMat24[3] → MatPES[4]	118M + 287k	r^{2}SCAN	255M
PET-MAD-DOS[6]	MAD[7]	95,595	PBEsol	8,625,226

Structural Results

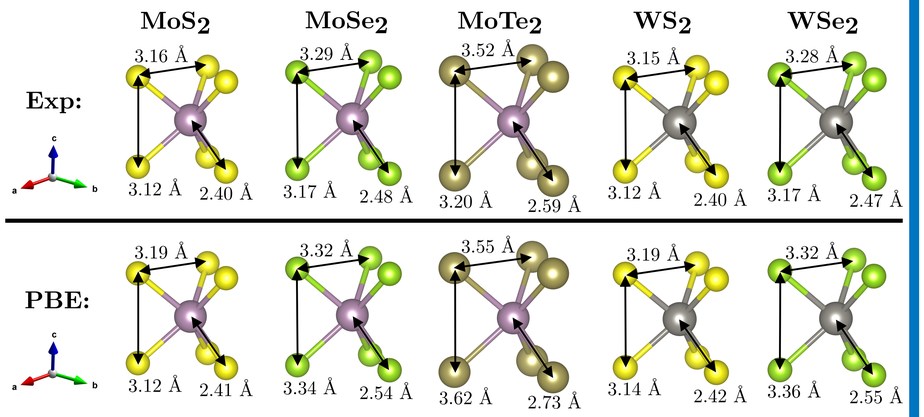
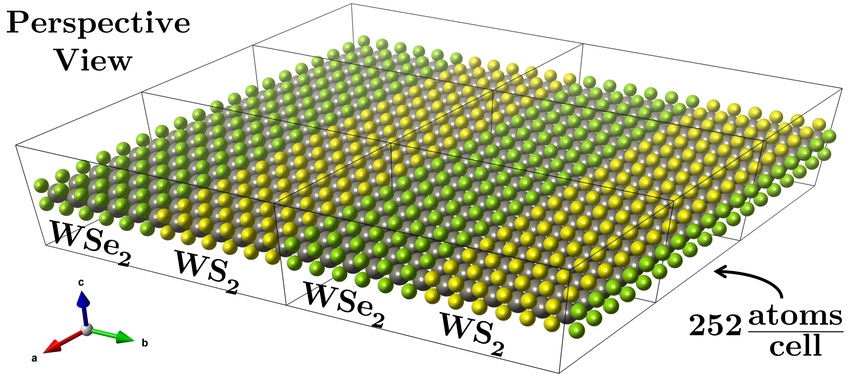


Table 2: Comparison of structural prediction accuracy across density functional theory (PBE) and machine-learned interatomic potentials (MLIPs) compared to reference experimental structures from Samadi *et al.*[1] Reported values are mean absolute percentage errors (MAE) for lattice constants, monolayer thickness, and transition metal to chalcogen bond-lengths, along with average RMSD in Å.

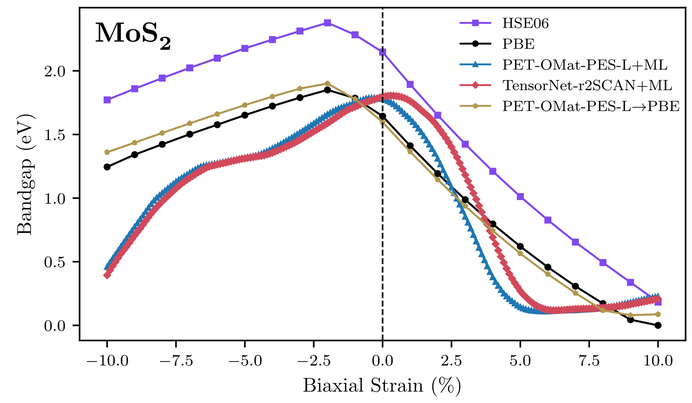
Metric	PBE	uma-s-1p2	uma-m-1p1	TensorNet -PBE	TensorNet -r2SCAN	PET-OMat -PES-L
Lattice Constant MAE (%)	1.031	1.227	1.009	0.629	0.729	1.008
Thickness MAE (%)	4.984	4.794	4.927	5.592	3.725	3.986
Bond-Length MAE (%)	2.626	2.660	2.591	2.649	1.926	2.212
Average RMSD (Å)	0.069	0.068	0.068	0.074	0.058	0.060

- All MLIPs attain *very similar* relaxations compared to the experimental unit-cells – however, MLIPs trained on r^{2}SCAN data (in blue) *slightly* outperform the PBE structural relaxations
- TensorNet-r^{2}SCAN is used to relax a periodic WS_2/WSe_2 in-plane heterostructure with viable results (below) despite the ~4% lattice mismatch, distorting the lattice & inducing non-uniform strain



Bandgap Modeling

- Traditional MLIPs are not capable of bandgap modeling: however, the PET-MAD-DOS model supports bandgap prediction given a relaxed structure using ML
- Unstrained, PET-MAD-DOS (denoted "+ML") closely reproduces PBE bandgaps (regardless of PBE vs. MLIP relaxed structures), underestimating true bandgaps by ~30%, compared to ~15% with HSE06



- Under strain, the MLIP + bandgap model workflow produces qualitatively correct behavior for all five TMDs but is quantitatively a poor match to PBE/HSE06 results

Conclusion

- MLIPs perform very well for structural relaxations of monolayer TMDs and heterostructures, which may then be used for more expensive static DFT calculations
- The bandgap model predicts correct trends, but it poorly predicts bandgaps outside of the zero-strain case
- For TMDs, MLIPs and ML workflows are primarily limited by the level-of-theory of training data!

References: **(1)** Samadi, M. *et al. Nanoscale Horizons* **2018**, *3*, 90-204. **(2)** Wood, B. M. *et al.* A Family of Universal Models for Atoms, **2025**. **(3)** Barroso-Luque, L. *et al.* Open Materials 2024, **2024**. **(4)** Kaplan, A. D. *et al.* A Foundational Potential Energy Surface Dataset for Materials, **2025**. **(5)** Bigi, F. *et al.* Pushing the limits of unconstrained machine-learned interatomic potentials, **2026**. **(6)** How, W. B. *et al.* A universal machine learning model for the electronic density of states, **2025**. **(7)** Mazitov, A. *et al. Scientific Data* **2025**, *12*, 1857.