Dispersion-corrected Machine Learning Potentials and a Systematic Database for 2D van der Waals Materials

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Machine-learned interatomic potentials (MLPs) based on message passing neural networks hold promise to enable large-scale atomistic simulations of complex materials with *ab initio* accuracy. A number of MLPs trained on energies and forces from density functional theory (DFT) calculations employing semi-local exchange-correlation (xc) functionals have recently been introduced [2]. In the present work, we benchmark the performance of six dispersion-corrected MLPs on a dataset of van der Waals heterobilayers containing between 4 and 300 atoms in the moiré cell. Using various structure similarity metrics, we compare the relaxed heterostructures to the ground truth DFT results. With some notable exceptions, the model precisions are comparable to the uncertainty on the DFT results stemming from the choice of xc-functional. We further explore how the structural inaccuracies propagate to the electronic properties, and find that in most cases the effect on the band energies is below 0.01 eV. Our results demonstrate that several MLP models are on par with DFT for vdW heterostructures, and thus justify their application to complex and experimentally relevant 2D materials. Finally, we present the groundwork for a systematic database of vdW heterostructures, *HetDB*, starting with the 336 structures presented in this benchmark.

References

[1] Antoine Loew, Dewen Sun, Hai-Chen Wang, Silvana Botti, Miguel A. L. Marques, arXiv:2412.16551 (2024) [cond-mat.mtrl-sci]
[2] Janosh Riebesell, Rhys E. A. Goodall, Philipp Benner, Yuan Chiang, Bowen Deng, Gerbrand Ceder, Mark Asta, Alpha A. Lee, Anubhav Jain, Kristin A. Persson, arXiv:2308.14920 (2023) [cond-mat.mtrl-sci]

Figures

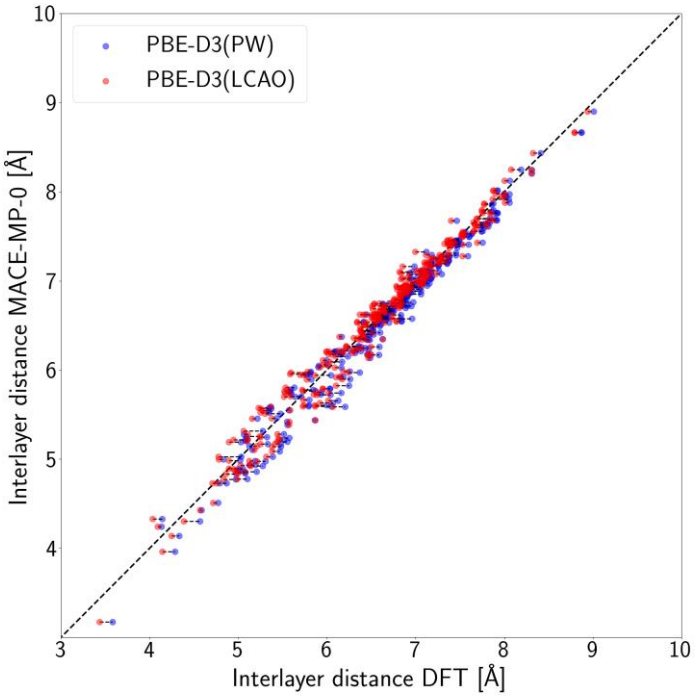

Figure 1: Interlayer distances for 336 vdW heterostructures relaxed using the large MACE-MP-0 MLP versus DFT calculations with wavefunctions represented in either a plane wave (PW) or linear combination of atomic orbitals (LCAO) basis. All results include a D3 dispersion correction.