Understanding Domain Reconstruction of Twisted Bilayer and Heterobilayer Transition Metal Dichalcogenides through Machine Learned Interatomic Potentials

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In the study of twisted bilayer 2D materials, a detailed picture of the relaxations and layer-corrugations that occur due to interlayer interaction is crucial to predicting how their electronic and optical properties depend on twist angle and the resulting large-scale Moiré pattern [1]. As the relative twist angle between the layers approaches 0º, referred to as parallel (P) stacking, or 60º, referred to as antiparallel (AP) stacking, reconstructions occur to maximise the area of low-energy stacking domains, with a lattice of solitons of high-energy stacking connected by domain walls (DWs). We show that Machine Learned Interatomic Potentials (MLIPs) can provide the combination of accuracy and scaling required to obtain atomistic insight into this behaviour. In contrast to empirical potential methods, MLIPs based on higher-order equivariant message passing, as implemented in MACE [2], can provide very precise energetics of stacking, strain, shear and varying interlayer distances to exactly reproduce vdW-corrected DFT for systems dramatically larger than can be treated with ab initio methods. We predict, explain and quantify the domain reconstruction patterns for all like-chalcogen combinations of the Transition Metal Dichalcogenides MoS_2, $MoSe_2$, WS_2 and WSe_2 down to twist angles approaching 1º. We demonstrate effects including DW-bending in AP systems, and the “twirling” that occurs at the solitons in heterobilayers.

References

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[2] I. Batatia, D. P. Kovacs, G. N. C. Simm, C. Ortner, and G. Csanyi, MACE: Higher order equivariant message passing neural networks for fast and accurate force fields, *Adv Neural Inf Process Syst* (2022).

Figures

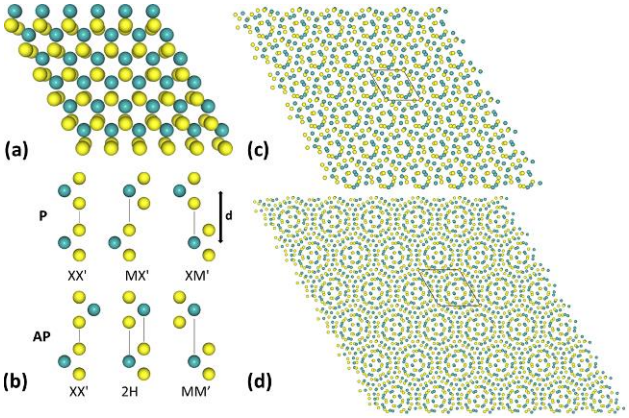

Figure 1. Schematics of the systems providing training data for the MLIP developed in this work (a) Monolayer MoS_2; (b) high-symmetry stacking configurations of bilayer TMDs, showing the three configurations in the P and AP cases; (c) the Moiré unit cell of the largest accessible twist angle (21.79º) and (d) the Moiré unit cell of the second-largest accessible twist angle (13.17º).

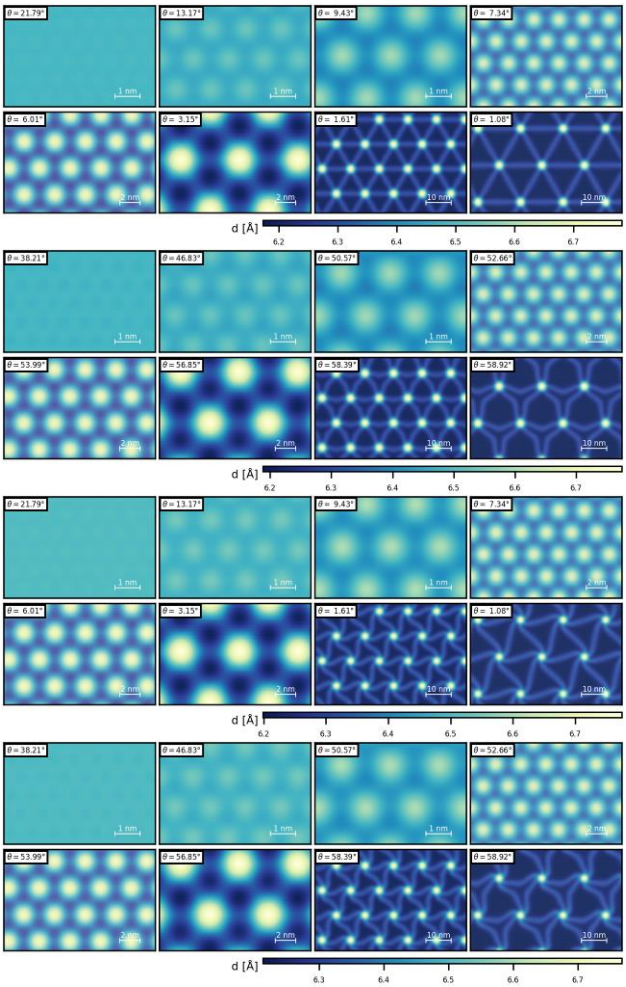

Figure 2. Interlayer distance maps for (top to bottom): P-MoS_2/MoS_2, AP-MoS_2/MoS_2, P-MoS_2/WS_2 and AP-MoS_2/WS_2 at a range of twist angles.