

Supplementary Information

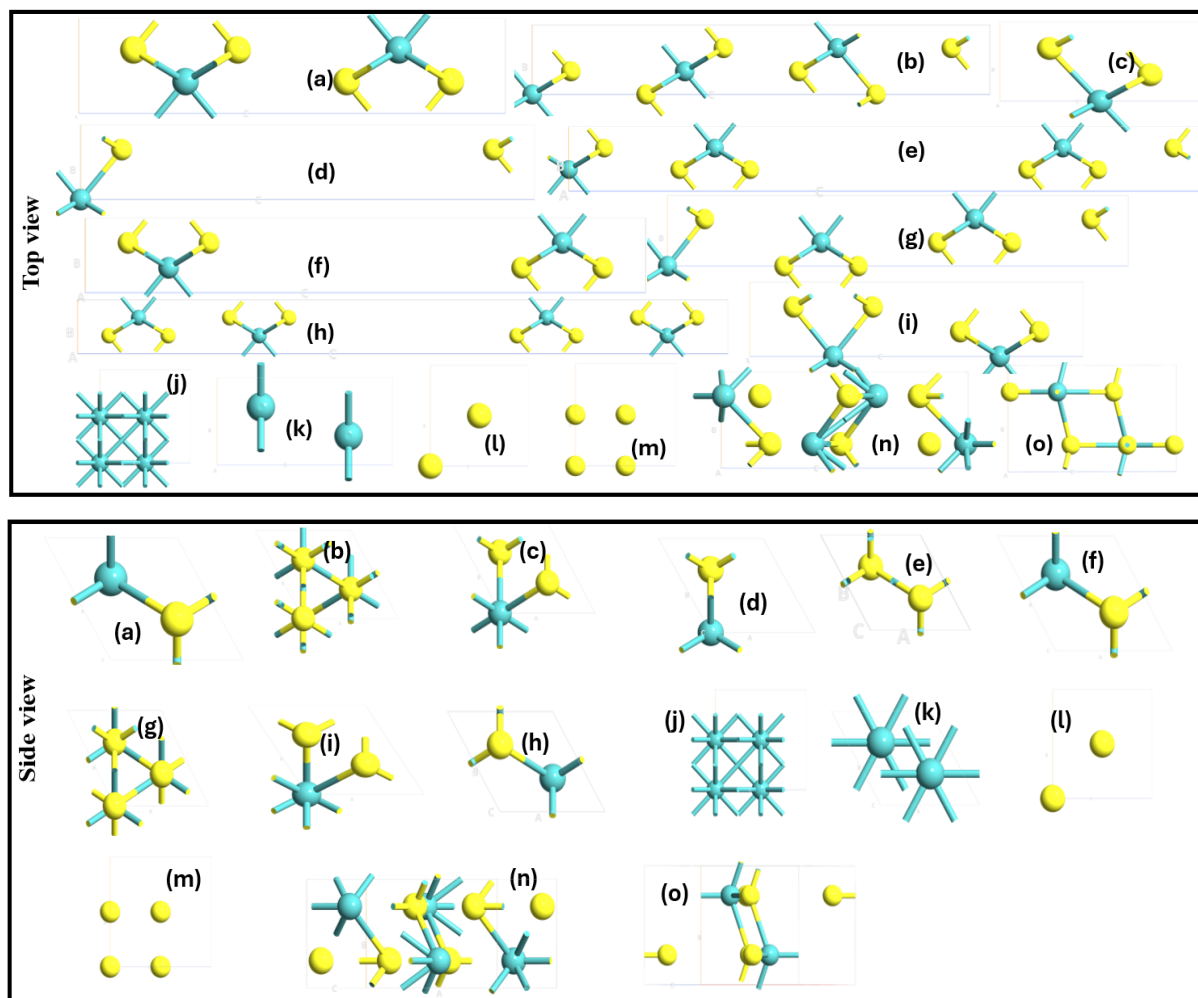


Figure S1. Representative crystal structures used for i-MTP training. Top views (a-o) of pristine MoS₂, elemental Mo (blue spheres), elemental S (yellow spheres), and their strained variants. Side views (a-o) of pristine MoS₂, elemental Mo, elemental S, and their strained variants.

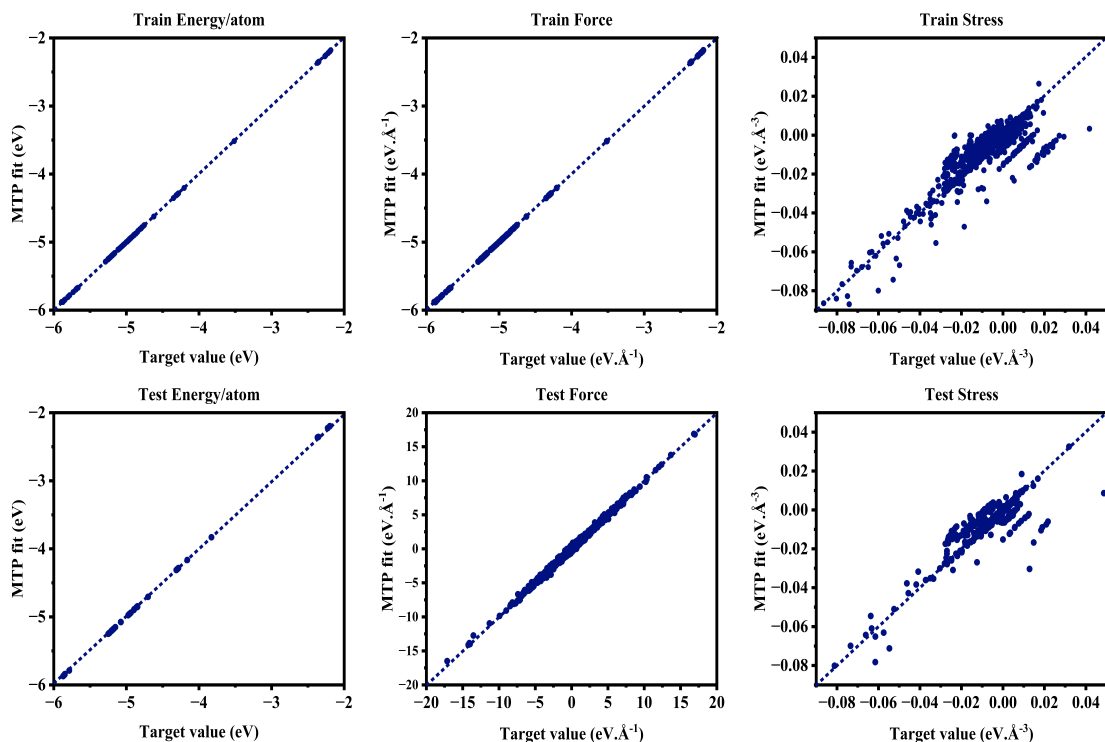


Figure S2. Scatter plots of training- and test-set performance for the selected i-MTP. Energy, force, and stress predictions from the MTP are compared with the DFT reference values. The near-linear distribution of points demonstrates excellent agreement and minimal fitting error.

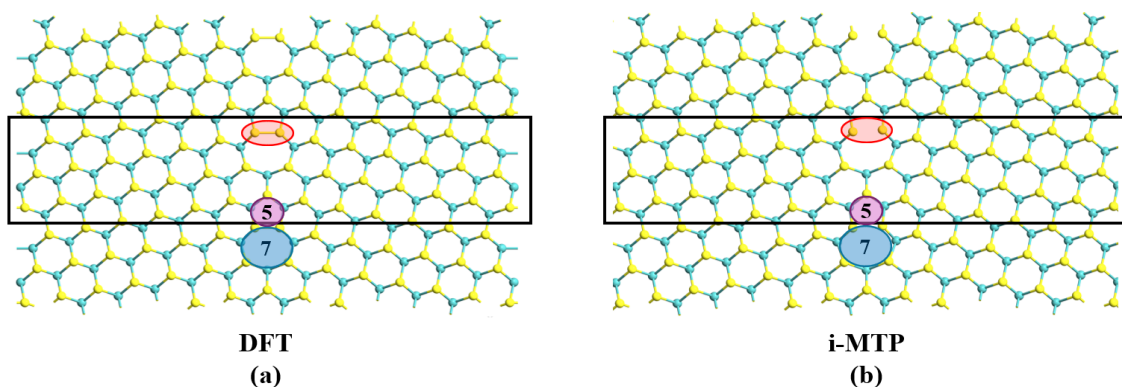


Figure S3. Comparison of (a) DFT- and (b) i-MTP-optimized 5|7-22-S GB structures in MoS_2 . Blue spheres represent Mo atoms, and yellow spheres represent S atoms. The black rectangular box indicates the simulation unit cell, which is periodic along the y-axis.

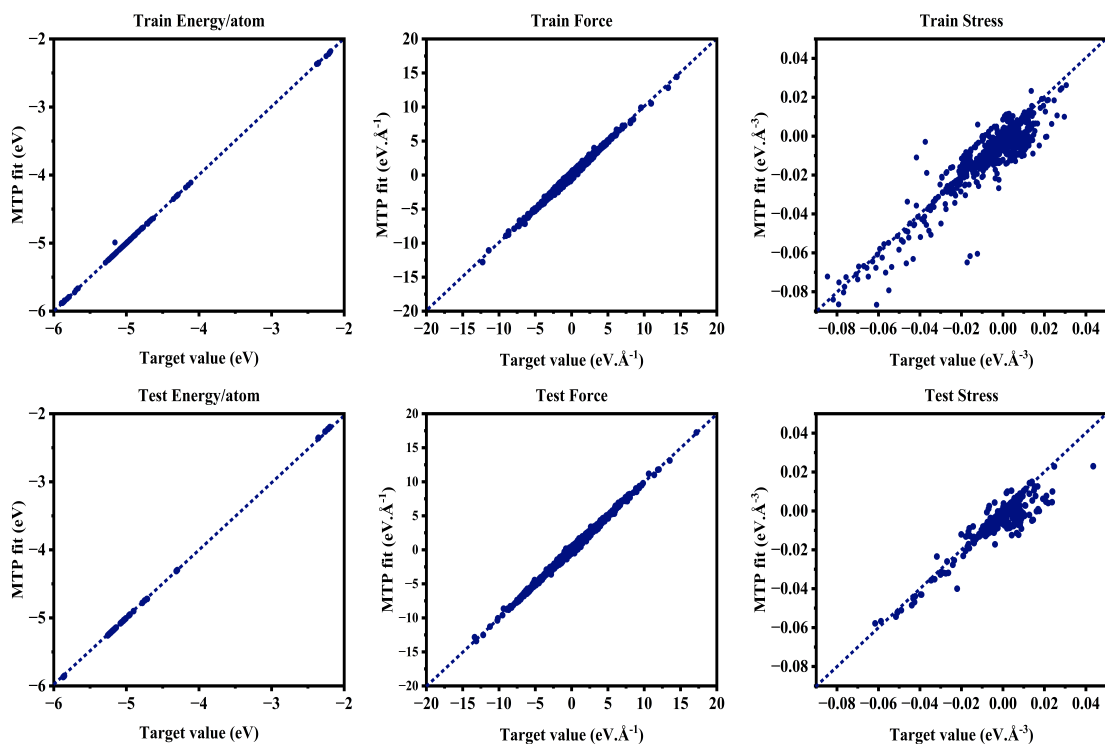


Figure S4. Improved accuracy with A-MTP. Scatter plots of training- and test-set performance for the selected A-MTP, comparing MTP-predicted energies, forces, and stresses with DFT reference values.

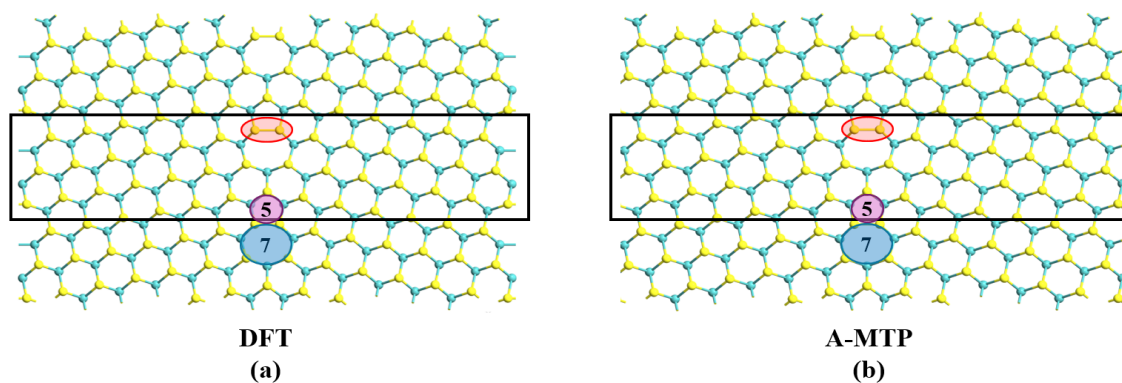


Figure S5. Comparison of (a) DFT- and (b) A-MTP-optimized 5/7-22-S GB structures in MoS₂. Blue spheres represent Mo atoms, and yellow spheres represent S atoms. The black rectangular box indicates the simulation unit cell, which is periodic along the y-axis.

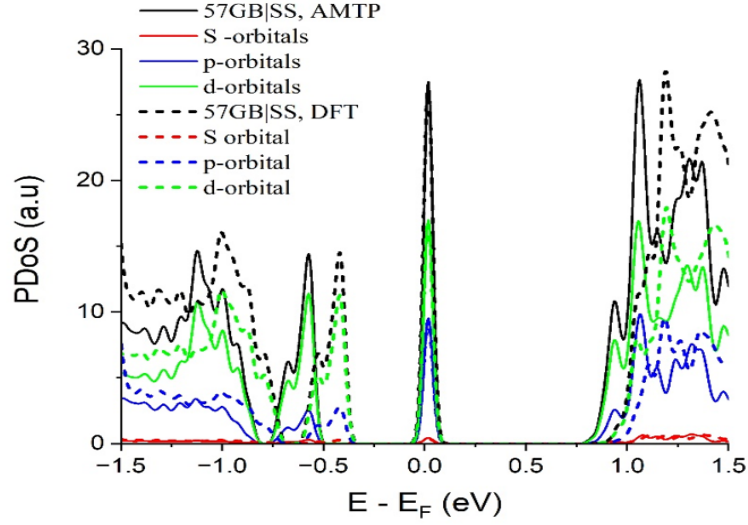


Figure S6. Orbital-resolved projected density of states (PDOS) at the grain boundary for the 5/7-22-S MoS₂ system, comparing DFT calculations performed using the DFT-relaxed geometry (dashed lines) and the A-MTP-relaxed geometry (solid lines). The calculation using the A-MTP-relaxed geometry reproduces the character of the defect-induced states with good agreement to the result obtained using the DFT-relaxed geometry.

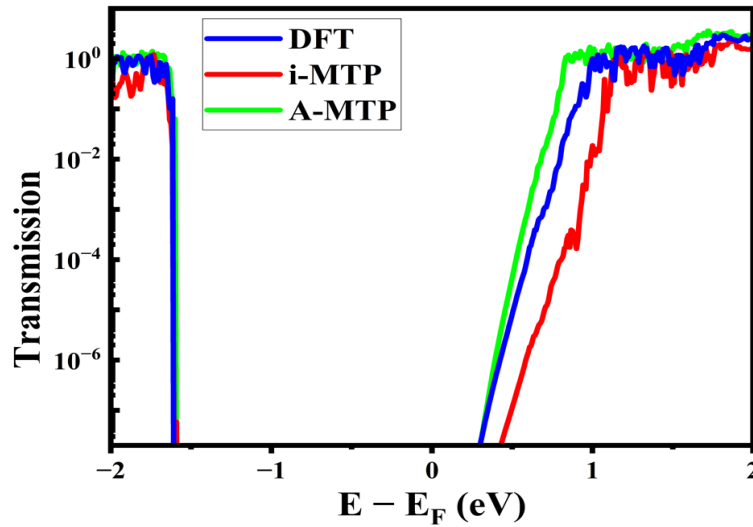


Figure S7. Transmission spectra calculated using the DFT-relaxed, i-MTP-relaxed, and A-MTP-relaxed 5/7-22-S geometries. Enriched training resolves earlier structural deviations, with the DFT-NEGF spectrum calculated using the A-MTP-relaxed geometry closely reproducing that obtained using the DFT-relaxed geometry.