Supporting information

Orientation Dependent Interlayer Stacking Structure in Bilayer MoS_2 domains

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S1. Atomic models with corresponding ADF-STEM simulations and intensity measurements for Mo zigzag step edges and 100% S-covered Mo Klein step edges in AA' stacked bilayer MoS_2

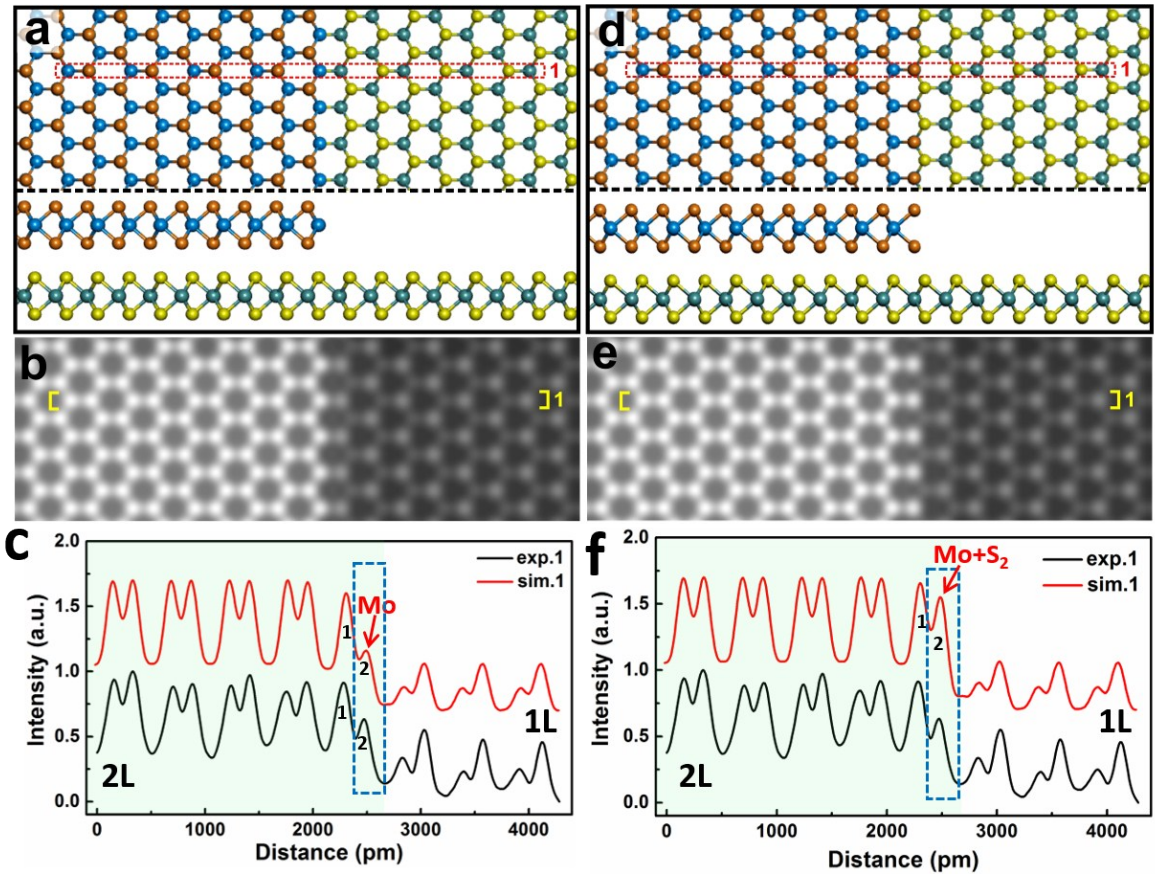

Figure S1. (a) Atomic model of one possible 2nd-layer MoS_2 step edge configuration having a Mo-terminated zigzag edge. (b) Multislice ADF-STEM image simulations based on the atomic model in (a) (see simulation parameters in Methods). (c) Intensity line profiles across the step edge region marked 1 in the experimental image in figure 2c and the simulated image in panel (b) of this figure, respectively. (d) Atomic model of an alternative 2nd-layer MoS2 step edge configuration having a 100% S-covered Mo Klein edge. (b) Multislice ADF-STEM image simulations based on the atomic model in (d). (c) Intensity line profiles across the step edge region marked 1 in the experimental image in Figure 2c and the simulated image in panel (e) of this figure, respectively.

We also constructed atomic models for another two possible 2nd-layer step edge configurations in the AA' stacked bilayer MoS_2 and used these to calculate additional ADF-STEM image simulations. One model is a Mo-terminated zigzag edge and the other is 100% S-covered Mo Klein edge, as shown in Figures S1 a and d. It can be seen that the intensity profiles at the step edge region for simulated images corresponding to these two models do not match those the experimental image, as indicated by the blue dashed boxes in Figures S1 c and f. The ratio of

intensity between peak 1 and 2 for the image simulation in Figure 3c and 3f are measured to be 1: 0.51 and 1: 0.89, both of which are not consistent with the intensity ratio between these two peaks in the experimental image, which is 1: 0.68. Therefore, these models are excluded as candidates for step edge structures for AA' stacked bilayer MoS_2. The intensity ratio between these two peaks for the image simulation in Figure 2f is 1:0.69, which agrees with experiments very well.

S2. Statistical analysis of AA' and AB stacked MoS_2 second layers on monolayer MoS_2

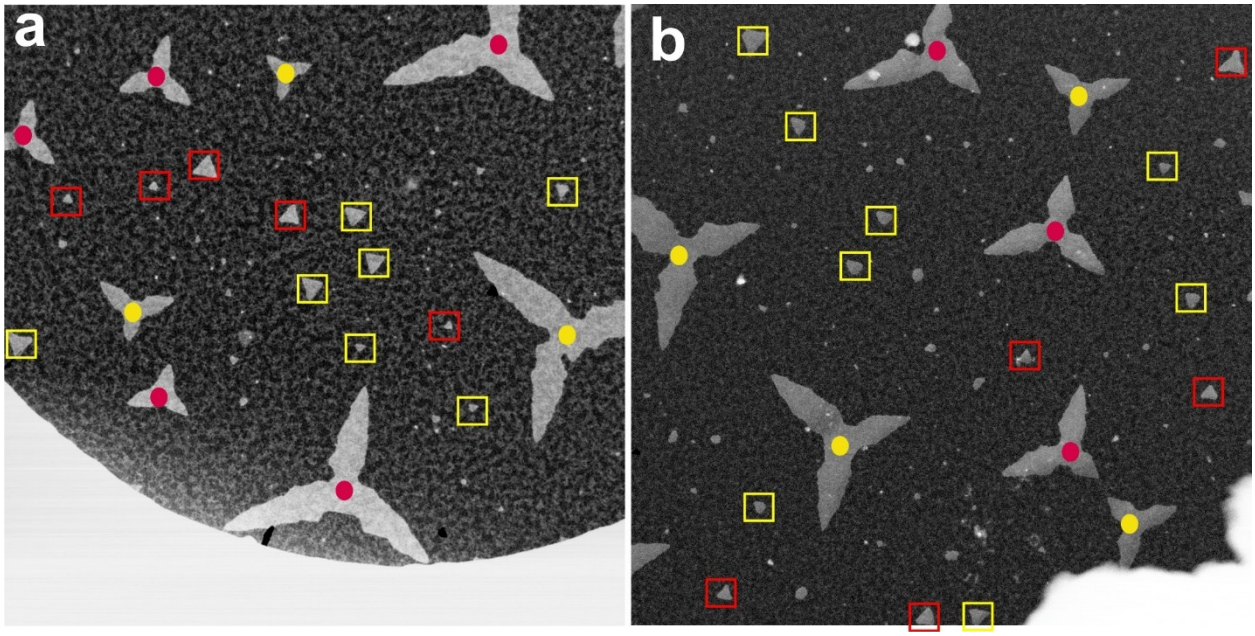

Figure S2. Low magnification ADF-STEM images used for the statistical analysis of domain orientations shown in Figure 4b. Red and yellow markers represent AA' and AB stacked bilayer MoS_2, respectively, while dots and squares indicate the larger shuriken-shaped 2nd-layer domains and smaller nuclei, respectively.