

## (Supplementary Materials)

### Intrinsic setting of exciton state in MoS<sub>2</sub> monolayer via tailoring Moiré correlation with a sapphire substrate

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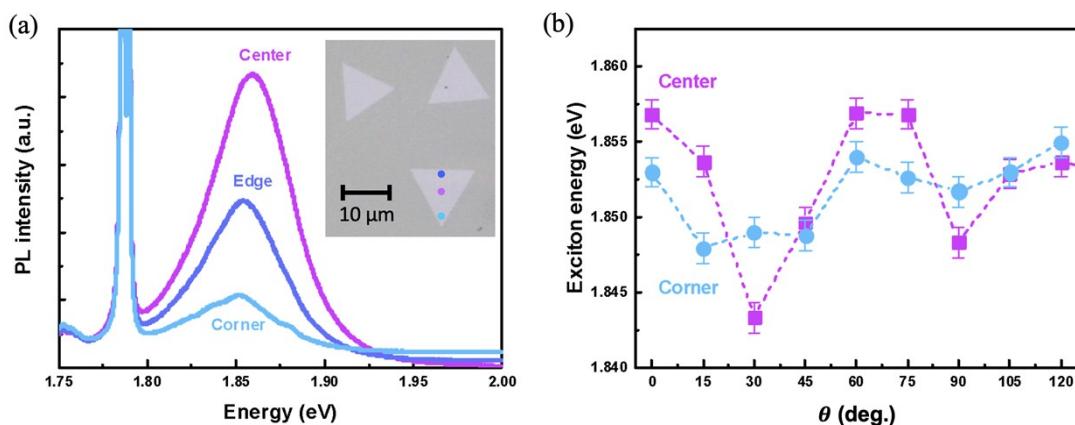
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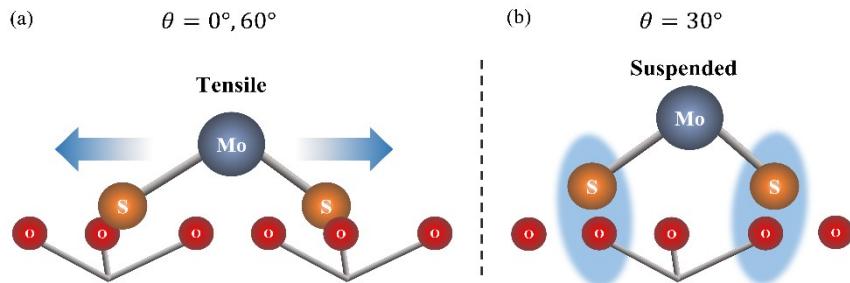
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**Supplementary Materials 1|**Site-dependent PL spectra on an MoS<sub>2</sub> monolayer domain with  $\theta$ -dependency, which shows significant PL redshift together with intensity drop while collecting the PL spectra at the edge of the MoS<sub>2</sub> domains.



**Fig. S1** (a) Site-selected PL spectra of MoS<sub>2</sub> monolayer acquired at center (violet), edge (deep blue), and corner (blue), as highlighted in the optical image in the inset. (b) Plots of exciton energy taken at the center and corner of MoS<sub>2</sub> monolayers as a function of  $\theta$ . Note: the plots of  $\theta > 60$  deg were arbitrarily selected to extend the 6-fold periodicity.

**Supplementary Materials 2|**Conceptual demonstration for the tensile stress induced via the closely packed stack.



**Fig. S2** Atomic stacking configuration for the case of (a)  $\theta = 0^\circ/60^\circ$  and (b)  $\theta = 30^\circ$ . In **Fig. S2(a)**, the preferred population of S atoms at the pseudo-interstitial sites, locating at the pyramid tops formed by three O atoms, induces tensile strain in the  $\text{MoS}_2$  monolayer. In contrast, in **Fig. S2(b)**, the mismatched stacking between  $\text{MoS}_2$  and  $\text{Al}_2\text{O}_3$  may drive S atoms to primarily occupy the positions directly above O atoms, resulting in a pseudo-suspended configuration.