# Supplementary Material for: Switching the moiré lattice models in the twisted bilayer $WSe_2$ by strain or pressure

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# Contents

# 1 Computational methods

The first-principles calculations are performed within the density functional theory (DFT) framework using the projected augmented-wave (PAW) method[1] as implemented in the Vienna ab initio simulation package (VASP).[2,3] The exchange-correlation term is treated in the generalized gradient approximation (GGA) of Perdew-Burke-Ernzerhof (PBE).[4] The Kohn-Sham orbitals are expanded in a plane wave basis set with an energy cutoff of 550 eV. The $15 \times 15 \times 1$ k-point grid and the $\Gamma$ point are used for the ground state and relaxation calculations of the $1 \times 1$ unitcell and moiré supercell, respectively. All the structures are optimized until the Hellman-Feynman forces on each atom are smaller than 0.01 eV/Å and the energy convergence is reached when the energy difference is below $10^{-6}$ eV between two consecutive self-consistent steps. A vacuum space of more than 15 Å along the $z$ direction is added to decouple artificial interactions between periodic neighboring slabs. Van der Waals corrections are taken into account using the Tkatchenko-Scheffler method.[5] The VESTA[6] package is used to visualize the geometry structures and the real space illustration of the wave functions.The real space illustrations of wave functions are constructed with pseudoatomic orbital using the first-principles calculation package OpenMX. Where the norm-conserving pseudo-potentials are used for the discretion of the core electron.[7,8] The twisted bilayer $WSe_2$ supercells with twist angles of $5.1^\circ/54.9^\circ$, $4.4^\circ$, and $3.9^\circ$ are generated along $[2, -1]$ to obtain the minimal cells and contain 762, 1014, and 1302 atoms, respectively. A negligible tensile strain ($\approx 0.0013\%$ ) is applied on the hBN layers (the fully relaxed lattice constant is 2.509Å) to satisfy the commensurate periodic boundary condition in the ABA sandwich stacking heterostructure.

## 2 Squared wave function distribution of t5.1° $WSe_2$

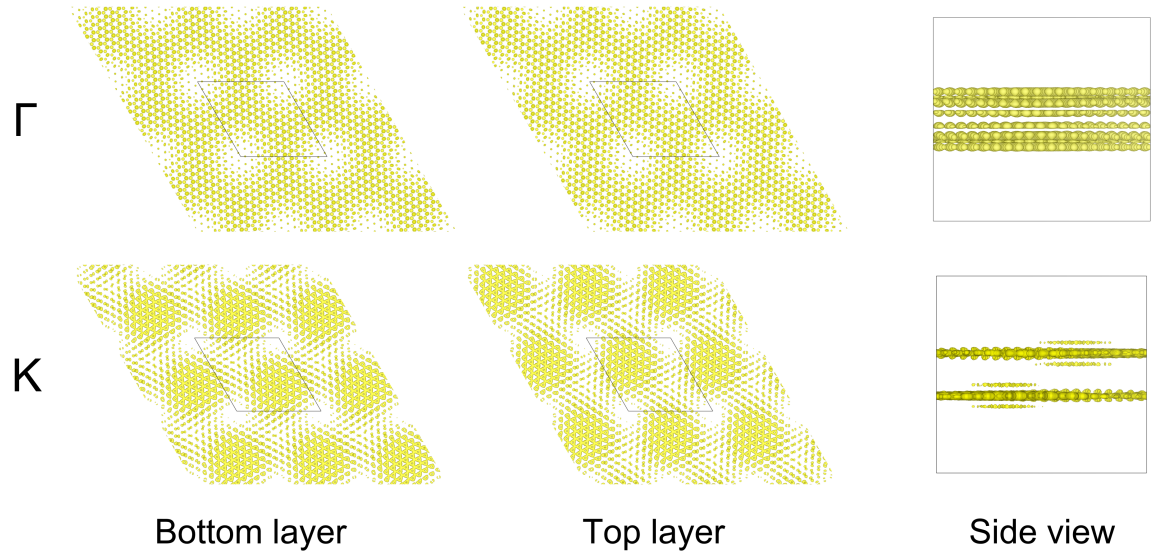


Figure S1: Real space illustration of the squared wave functions $|\psi_k|^2$ of Γ (upper panel) and $K$ valleys (lower panel) at $\gamma$ for t5.1° $WSe_2$.

# 3 Untwisted bilayers

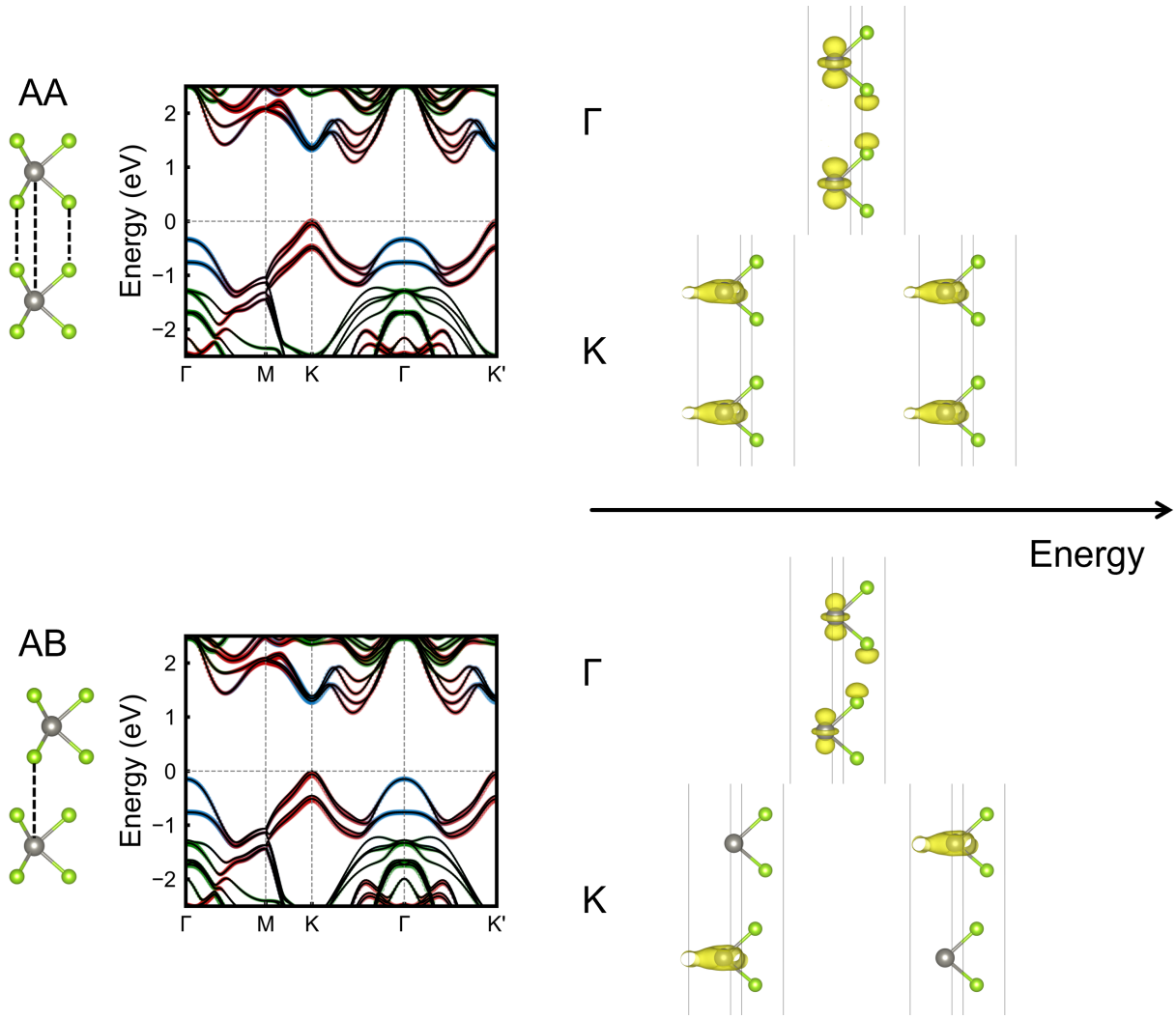


Figure S2: Left panel: the band structure of AA(upper) and AB(lower) stacked bilayer $WSe_2$ with orbital projections. The color description is the same as the main text; Right panel: Side views of the squared wave functions $|\psi_k|^2$ at $\Gamma$ and $K$ points in the order of increasing energy from left to right.

# 4 Band structures of other twisted angles

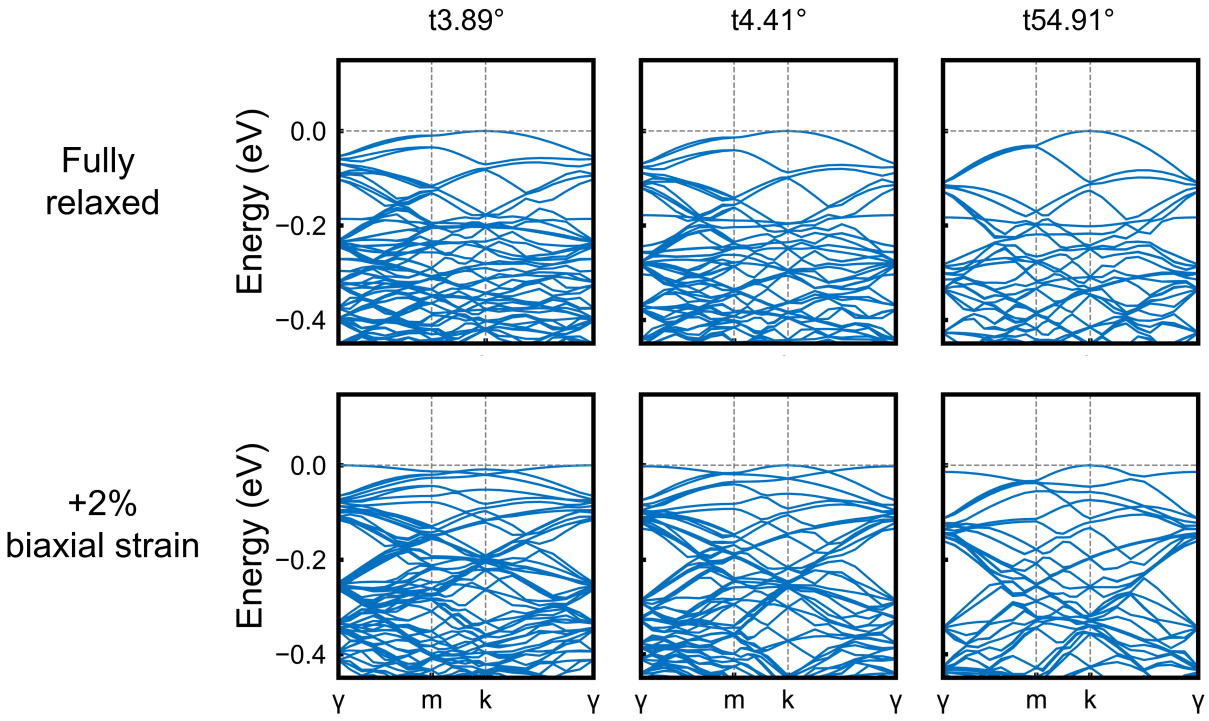


Figure S3: The DFT calculated band structures of twisted bilayer $WSe_2$ near Fermi level with different twisted angles (54.9°, 4.4°and 3.9°, from left to right, respectively).

# 5 5.1° $tWSe_2$ under out-of-plane pressure with/without BN layers

The pressure is calculated by:

$$p = \frac{\sum f_{z-BN_{top}} - f_{z-BN_{bottom}}}{2|\vec{a} \times \vec{b}|} \tag{1}$$

where $f_z$ describes the calculated atomic force on each atom in the top/bottom layer after geometric relaxation. It turns out that:

Table S1: Calculated results for out-of-plane pressure for different initial layer distance $d$.

| $d$ (Å) | Pressure (GPa) |
|---|---|
| 2.5 | 7.935 |
| 3 | 2.175 |
| 3.5 | -0.562 |

To reduce computational cost, we remove the BN layers after relaxation and calculate the band structure. To exclude unexpected interactions between BN layers and $tWSe_2$, the density of states projected on BN layers and the $tWSe_2$ is plotted in Fig. S4(a). There seems no contribution of BN layers near the Fermi level. Besides, the band structure with BN layers is also similar to that without BN layers, as shown in Fig. S4(b).

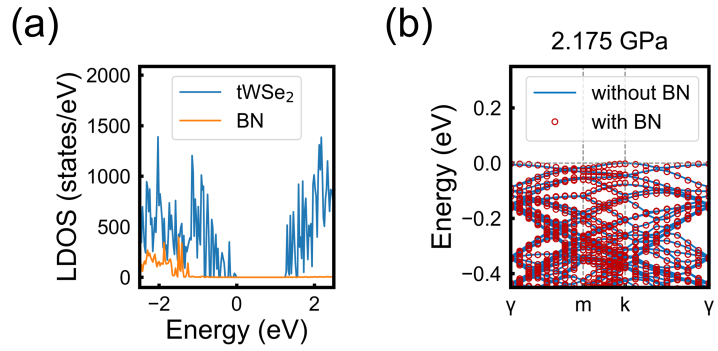


Figure S4: (a) Projected density of states of BN−$tWSe_2$−BN sandwiched structure. (b) The band structure with and without BN layers, as plotted with red circles and blue lines, respectively.

# References


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