Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information for

Moiré-Templated Strain Patterning in Transition-Metal Dichalcogenides and Application in Twisted Bilayer MoS_2

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This supporting file contains the following:

1. Probing moiré pattern in bilayer TMDs

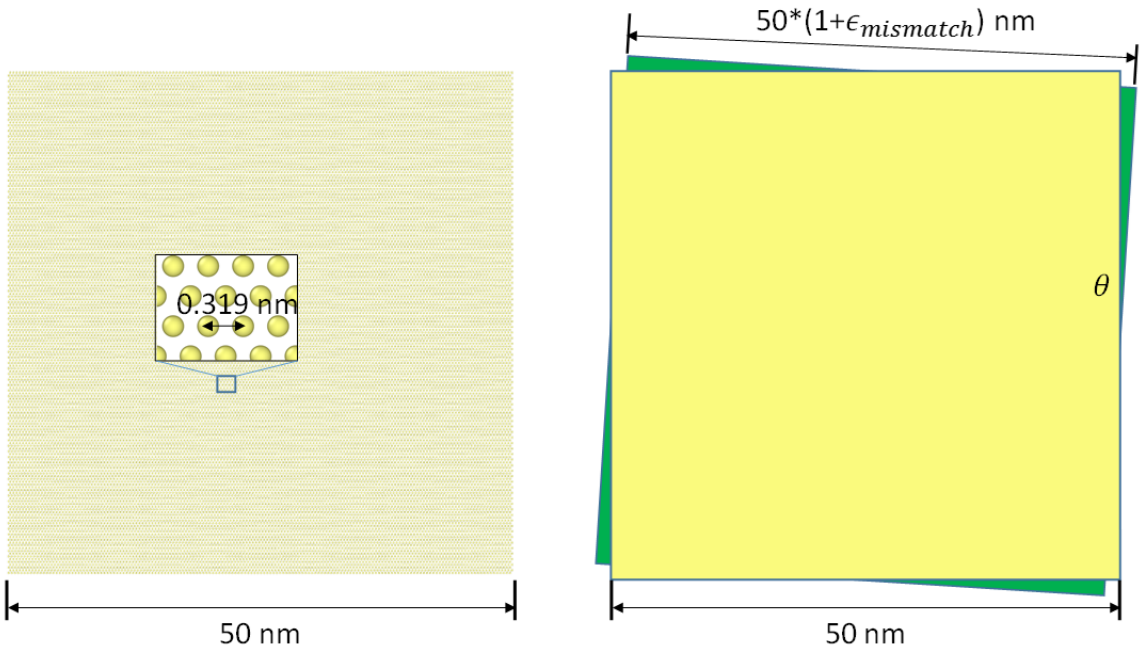

Figure S1: schematic of constructing moiré of TMD bilayers using interfacial chalcogenide atoms with twist angle and mismatch.

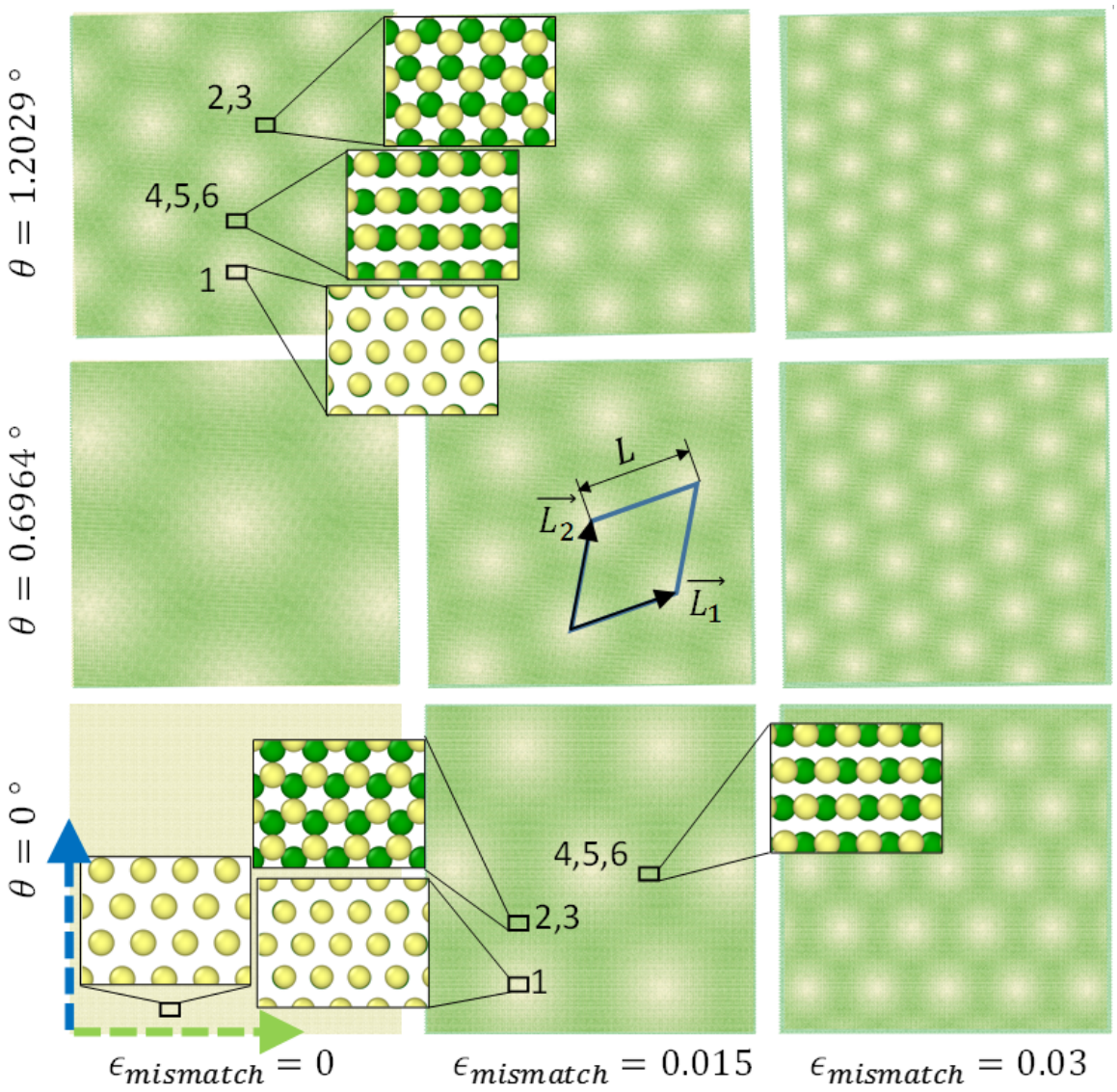

Figure S2: moiré patterns and representative stacking pattern, following the notation in Figure 1 of main text. Dashed arrows mark the orientation of the moiré (same as in Figure S3).

To calculate structure-energy relationship, the interaction between the X atom and the X' atom is described in standard 12-6 potential

$$V(r) = 4 * \epsilon(\frac{s^{12}}{r^{12}} - \frac{s^6}{r^6})$$

Where $\epsilon = 1\ eV$

$d_0 = 0.339\ nm$ in the maintext is the equilibrium distance of the above potential.

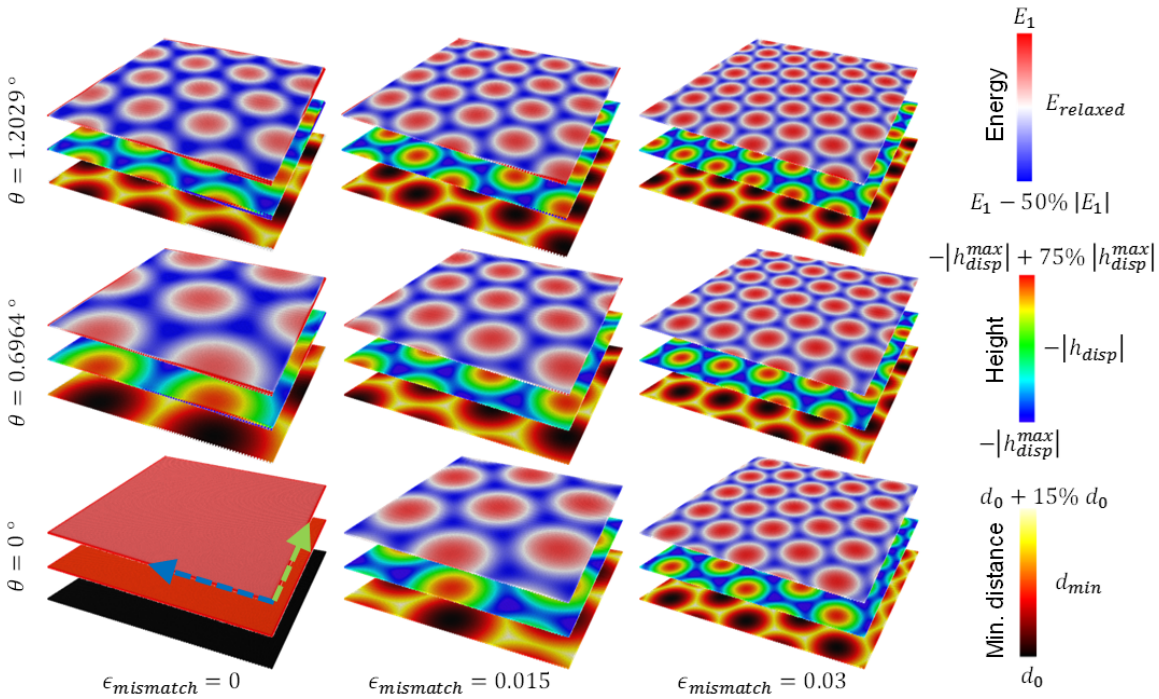

Figure S3: Energy, height and minimum distance distribution of the X layer. Refer to detailed definition in Figure 2 of main text. Dashed arrows mark the orientation of the moiré (same as in Figure S2).

2. Geometry of moiré pattern in twisted bilayer MoS2

Following the notation in Figure 1 of main text, the moire pattern should satisfy:

$$\overrightarrow{L_1} = m\overrightarrow{a_1^{(1)}} + n\overrightarrow{a_2^{(1)}} = m\overrightarrow{a_2^{(2)}} + n\overrightarrow{a_1^{(2)}}$$

The twist angle is then $\theta = \arccos\left(\frac{1}{2}\frac{m^2+n^2+4mn}{m^2+n^2+mn}\right)$

3. Generic modeling of substrate adhesion in MD simulation

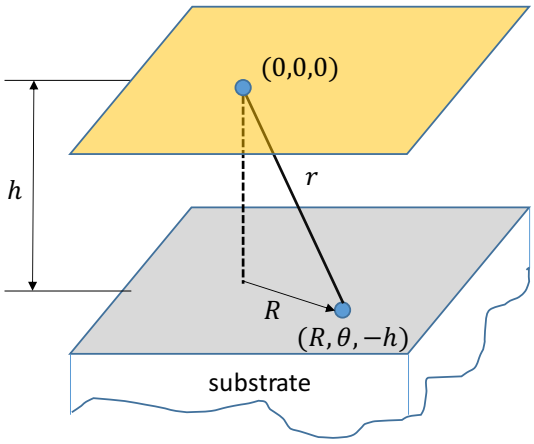

Figure S4: schematic to calculate the interaction energy between a point in the 2D layer and a point in the bulk substrate (atom-to-atom pair interaction energy). Represented in cylinder coordinates.

The atom to atom pair energy is calculated with standard 12-6 potential

$$V(r) = 4 * \epsilon(\frac{\sigma_{eq}^{12}}{r^{12}} - \frac{\sigma_{eq}^{6}}{r^{6}})$$

For substrate, denote ρ_p as atom density per volume; for 2D layer, denote ρ_p as atom density per area. Thus the total energy per volume in substrate per area in 2D layer is calculated as

$$\rho_g dA_g \int V(r)\rho_p dV_p = \rho_g dA_g \rho_p \int V(r) dV_p = \rho_g dA_g \rho_p \int_{-\infty}^{-h} dz \int_0^{\infty} V(r) 2\pi R dR$$

The cohesive energy Φ induced by substrate is energy per unit area of 2D layer

$$\Phi = 2\pi\, \rho_g \rho_p \int_{-\infty}^{-h} dz \int_0^{\infty} V(r) R dR$$

$$= 2\pi\, \rho_g \rho_p \int_{-\infty}^{-h} dz \int_0^{\infty} 4\,\epsilon\, (\frac{\sigma_{eq}^{12}}{(z^2+R^2)^6} - \frac{\sigma_{eq}^{6}}{(z^2+R^2)^3}) R dR$$

$$= 2\pi \rho_g \rho_p\, \epsilon\, \sigma_{eq}^6 \frac{-15h^6 + 2\,\sigma_{eq}^6}{45\, h^9} = \frac{2\pi}{3} \rho_g \rho_p\, \epsilon\, \sigma_{eq}^6 \frac{-15h^6 + 2\,\sigma_{eq}^6}{15\, h^9}$$

$$\Phi = \frac{2\pi}{3} \rho_g \rho_p\, \epsilon\, \sigma_{eq}^6 \left(\frac{2\,\sigma_{eq}^6}{15\, h^9} - \frac{1}{h^3} \right)$$

The interaction energy per atom is

$$\mathrm{E} = \Phi / \rho_g = \frac{2\pi}{3} \rho_p\, \epsilon\, \sigma_{eq}^3 \left(\frac{2\,\sigma_{eq}^9}{15\, h^9} - \frac{\sigma_{eq}^3}{h^3} \right)$$

In MD simulation, the effect of substrate can be thus modeled using the standard 9-3 potential with a virtual wall

$$V_{93}(r) = \epsilon_{93}(\frac{2\,\sigma_{eq}^9}{15\,h^9} - \frac{\sigma_{eq}^3}{h^3})$$

The buckling initiation sites in bilayer TMDs does not depend on the substrate adhesion. Without losing generality, in the MD simulations of bilayer MoS_2, we use parameters $\sigma_{eq} = 3.5\,Ang, \epsilon_{93} = 0.2\,Kcal/mole.$

The density of number of atoms per unit area of MoS_2 (e.g., for triangular lattice, the outer layer of S) is

$$\rho_g = 3 * \frac{\frac{1}{2}}{\frac{1}{2} l_0 * l_0 * \frac{\sqrt{3}}{2}} = \frac{6}{\sqrt{3} l_0^2}$$

Where l_0 is equilibrium distance between points in a triangular lattice. The factor of 3 is caused by that in TMD, there are three layers of atoms.

4. MD simulation of buckling of twisted bilayer MoS_2

The substrate effect is modeled using a virtual wall (see Section 3 in supporting information). Before deformation, energy minimization is performed. The deformations for X, Y and equi-biaxial is carried in a quasi-static mode. After each incremental time step, energy minimization is performed.

5. MD simulation of stacking domains in bilayer MoS_2

The substrate effect is modeled using a virtual wall (see Section 3 in supporting information). The simulation cell of each stacking domain is shown in Figure 5 of main text. Before deformation, energy minimization is performed. Then the out-of-plane displacement is disabled, only allowing in-plane relaxation. The further equi-biaxial deformations is carried in a quasi-static mode. After each incremental time step, energy minimization is performed.

6. Continuum mechanics model to predict the wrinkling morphology

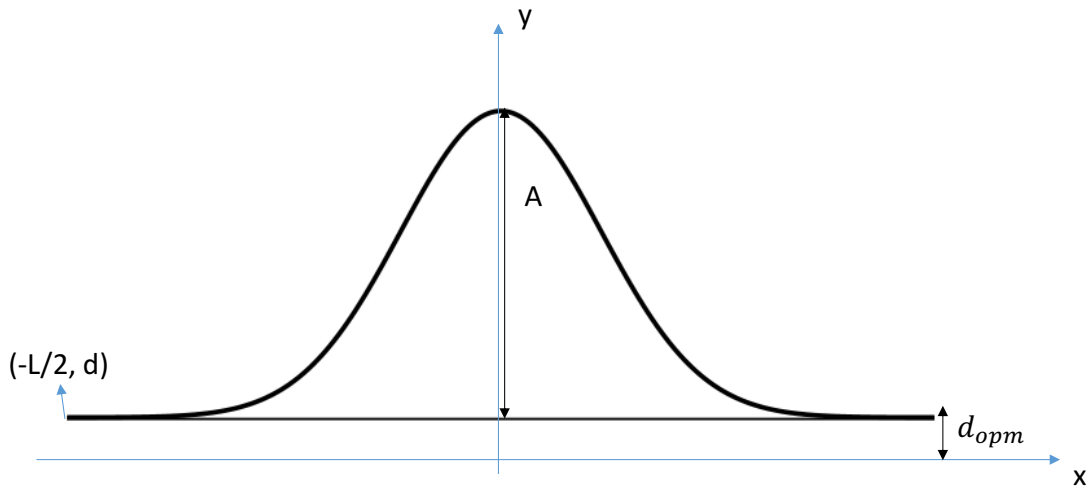

Figure S5: Schematic of characterizing the geometry (Amplitude A, half periodicity L, and separation distance to the substrate)

As already discussed in main text, due to the decoupled nature of the deformation of bilayer, we can model just one layer (Figure S5). The wrinkled shape have half periodicity $L/2$, amplitude A, asymptotic separation distance to substrate d_{opm}.

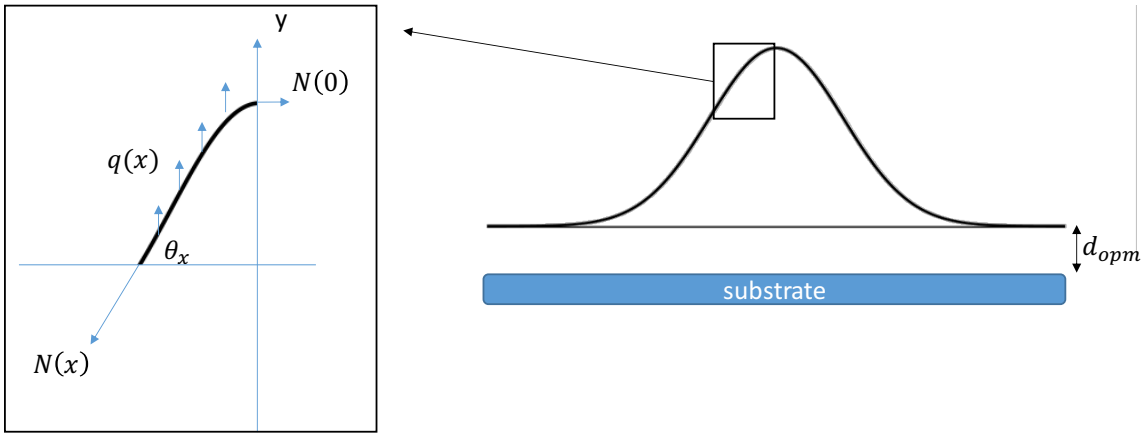

Figure S6: Schematic of deriving the normal strain in the wrinkled monolayer.

By symmetry, consider the left half of the wrinkled layer (Figure S6). Required by force equilibrium, the normal strain in the layer $N(x)$ is

$$N(x) = N(0)cos\theta_x + \int_{s(x)}^{s(0)} q(x)\, sin\theta_x ds$$

Where $q(x)$ is the vdW force exerted from the substrate (positive sign if pointing upward).

Denote the profile as $y(x)$, then we obtain

$$N(x) = N(0)\frac{1}{\sqrt{1+y'(x)^2}} + \frac{y'(x)}{\sqrt{1+y'(x)^2}}\int_{s(x)}^{s(0)} q(x)\ ds$$

$$N(x) = N(0)\frac{1}{\sqrt{1+y'(x)^2}} + \frac{y'(x)}{\sqrt{1+y'(x)^2}}\int_{x}^{0} q(x')\sqrt{1+y'(x')^2}dx'$$

Denote Et as the in-plane stiffness of the layer, then the normal strain can be expressed as

$$\epsilon(x) = \epsilon_0\frac{1}{\sqrt{1+y'(x)^2}} + \frac{1}{Et}S(x)$$

Where

$$S(x) = \frac{y'(x)}{\sqrt{1+y'(x)^2}}\int_{x}^{0} q(x')\sqrt{1+y'(x')^2}dx'$$

The solution of $y(x)$ can be obtained by minimize the total energy $U_{total} = U_{bending} + U_{compression} + U_{adhesion}$.

The bending energy can be expressed as

$$U_{bending} = \int_{-L/2}^{0}\frac{D}{2}k(x)^{\,2}ds$$

Where $k(x)$ is the curvature of $y(x)$.

The normal strain energy can be expressed as

$$U_{compression} = \frac{1}{2}Et\int_{-\frac{L}{2}}^{0}\big(\epsilon(x)\big)^2 ds$$

The adhesion energy can be expressed as

$$U_{adhesion} = \int_{-\frac{L}{2}}^{0} V_{93}(y(x)) * \rho'_g ds$$

Where

$$\rho'_g = \rho_g\frac{1}{1+\epsilon(x)}$$

to account for the change of atom density per area in the layer. The expression for $V_{93}(r)$ is described in Section 3 of supporting information. d_{opm} is the separation distance of minimim $V_{93}(r)$.

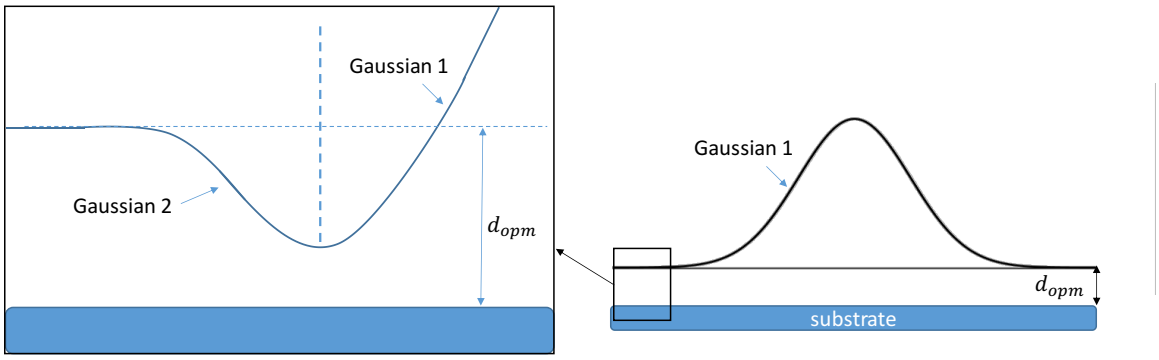

Figure S6: Schematic of deriving the normal strain in the wrinkled monolayer.

To obtain a simplified solution, we assume the shape as a piecewise function with two Gaussians (Figure S6).

$$y(x) = \begin{cases} d + A\, e^{-\frac{x^2}{2\,\sigma^2}}, -x_0 \le x \le 0 \\ d_{opm} - \left(d_{opm} - y(-x_0)\right) e^{-\frac{(x+x_0)^2}{2\,\sigma_2^2}}, -\frac{L}{2} \le x \le -x_0 \end{cases}$$

We further drop the vdW term in calculating the normal strain $\epsilon(x)$

$$\epsilon(x) \cong \epsilon_0 \frac{1}{\sqrt{1 + y'(x)^2}}$$

ϵ_{app} denote the globally applied strain to the undeformed layer. The original length of the sheet before buckling is

$$L_0 = \frac{L}{1 + \epsilon_{app}}$$

After deformation, the contour length of the layer is

$$L_{contour} = 2 \int_{-\frac{L}{2}}^{0} ds$$

We take an estimated assumption for simplicity

$$\epsilon_0 = \frac{L_{contour} - L_0}{L_0}$$

The total energy is then a five-variable function as $U_{total}(A, \sigma, x_0, \sigma_2, d)$, for every set of geometrical, mechanical, and material constants L(feature spacing), ϵ_{app} (applied

compression), ϵ_{93}(substrate adhesion), Et(in-plane stiffness). Minimize U_{total} gives the five variables that determines the spatial profile of the wrinkle.

The amplitude A is well-defined. The delamination length is reported as 2σ.

7. DFT calculation

Due to limited model size that DFT calculations can handle, we use a unit cell described in Figure S7. We first use the same MD simulation setup (e.g., substrate, deformation) as described in Section 5. We then take the structure snapshot at several deformations from MD simulation as input for DFT calculation. Slab model is used to ensure enough vacuum space in the out-of-plane direction.

The generalized gradient approximation (GGA) in the framework of Perdew-Burke-Ernzerhof (PBE) is adopted for the exchange-correlation potential. Numerical atomic orbitals with double zeta plus polarization are used for the basis set, with a plane-wave energy cutoff of 500 Ry. Geometric structures are relaxed until the force on each atom is less than 0.01 eV $A^{\circ-1}$ and the convergence criteria for energy is 10^{-5} eV. Monkhorst-Pack k-points is 2x9x1. For optical absorption calculation (the imaginary part of dielectric function), five times denser K-points were used and self-consistent field tolerance is 10^{-6}. The electronic smearing temperature during all calculations is 300 K.

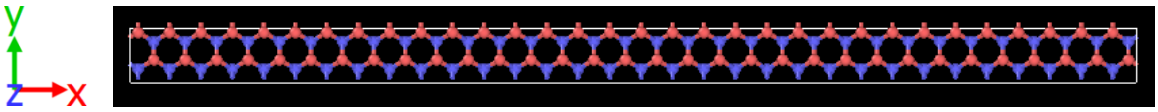

Figure S7: Schematic of unit cell of monolayer MoS_2 for DFT calculation.