Supplementary Materials for manuscript "Interplay of interlayer distance and in-plane lattice relaxations in encapsulated twisted bilayers"

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S1. Derivation of equation (5) in the main text

Throughout the Supplementary Materials we exploit designations introduced in the main text. Using Eq. (1) of the main text with

$$f(d) \approx f(d_0) + (d-d_0)^2,$$
$$e^{-qd} \approx e^{-qd_0}\left[1 - q(d-d_0)\right],$$
$$e^{-Gd} \approx e^{-Gd_0}\left[1 - G(d-d_0)\right],$$

we find internal pressure created by the TMD bilayer on up and down encapsulating slabs:

$$P_{\rm in}(\boldsymbol{r}_0, d_0+u_z^t+u_z^b) \equiv -\frac{\partial W_{\rm ad}(\boldsymbol{r}_0,d)}{\partial d}\bigg|_{d=d_0+u_z^t+u_z^b} = -2\varepsilon(u_z^t+u_z^b)+\sum_{l=1,2,3}\left\{\mathcal{A}Q\cos\left(\boldsymbol{G}_l\boldsymbol{r}_0\right)+\mathcal{B}G\sin\left(\boldsymbol{G}_l\boldsymbol{r}_0+\phi\right)\right\}. \quad \text{(S1)}$$

The internal pressure (S1) is balanced by pressures emerging between interface and inner layers of up and down encapsulating slabs that reads as

$$P_{\rm out}(d_* - u_z^t) = -\frac{\partial W_{\rm u}}{\partial d}\bigg|_{d=d_*-u_z^t} = k_{\rm u}u_z^t, \quad \text{(S2)}$$
$$P_{\rm out}(d_* - u_z^b) = -\frac{\partial W_{\rm d}}{\partial d}\bigg|_{d=d_*-u_z^t} = k_{\rm d}u_z^b. \quad \text{(S3)}$$

At equilibrium $P_{\rm in}(d_0+u_z^t+u_z^b) = P_{\rm out}(d_*-u_z^t)$ and $P_{\rm in}(d_0+u_z^t+u_z^b) = P_{\rm out}(d_*-u_z^b)$, which results in

$$u_z^t = \frac{k_{\rm u}}{(k_{\rm u}+k_{\rm d})}\frac{\sum_{l=1,2,3}\left\{\mathcal{A}Q\cos\left(\boldsymbol{G}_l\boldsymbol{r}_0\right)+\mathcal{B}G\sin\left(\boldsymbol{G}_l\boldsymbol{r}_0+\phi\right)\right\}}{2\varepsilon+\frac{k_{\rm u}k_{\rm d}}{k_{\rm u}+k_{\rm d}}} \quad \text{(S4)}$$
$$u_z^b = \frac{k_{\rm d}}{(k_{\rm u}+k_{\rm d})}\frac{\sum_{l=1,2,3}\left\{\mathcal{A}Q\cos\left(\boldsymbol{G}_l\boldsymbol{r}_0\right)+\mathcal{B}G\sin\left(\boldsymbol{G}_l\boldsymbol{r}_0+\phi\right)\right\}}{2\varepsilon+\frac{k_{\rm u}k_{\rm d}}{k_{\rm u}+k_{\rm d}}}. \quad \text{(S5)}$$
$$\text{(S6)}$$

Therefore, summing Eqs. (S4) and (S5) we obtain Eq. (5) in the main text.

S2. Relaxation order parameter dependence on twist angle for different sizes of XX stacking area

Figure S1 demonstrates that crossover twist angles from rigid to relaxed moiré pattern and difference between them for suspended and rigidly-encapsulated twisted P WSe_2 bilayers are independent on choice of the exact value of ν in Eq. (8) of the main text.

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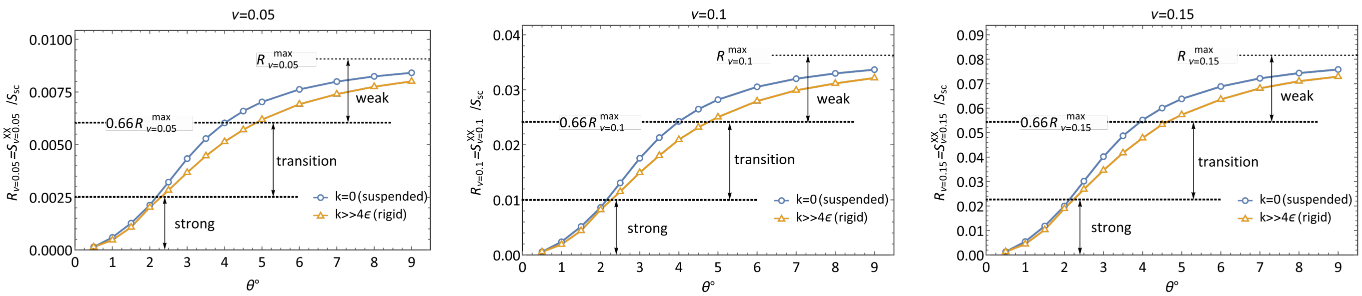

Figure S1. Dependences of the relaxation strength parameter as a function of twist angle for suspended and rigidly-encapsulated twisted P WSe_2 bilayers for three values of parameter $\nu = 0.05, 0.1, 0.15$ characterising three sizes of the XX stacking area (see Eq. (8) of the main text).

S3. Definition of $R_{\mathrm{MIN},\nu}$ in eq. (9) of the main text

We define $r_{\mathrm{min},\nu}$, that characterizes area of XX stacking area in the regime of strong relaxation of moiré pattern (i.e. $\theta^\circ < 2^\circ$), as a root of the following equation:

$$W_{\mathrm{adh}}(\boldsymbol{r}_0, d_{\mathrm{enc}}(\boldsymbol{r}_0))|_{|\boldsymbol{r}_0|=\nu a} = W_{\mathrm{adh}}(\boldsymbol{r}_0(\boldsymbol{r}_{\mathrm{min},\nu})), d_{\mathrm{enc}}(\boldsymbol{r}_0(\boldsymbol{r}_{\mathrm{min},\nu})|_{\boldsymbol{r}_0(\boldsymbol{r})=\theta z\times\boldsymbol{r}+\boldsymbol{u}^t(\boldsymbol{r})-\boldsymbol{u}^b(\boldsymbol{r})}\,. \tag{S7}$$

Graphical solution of Eq. (S7) is shown in Fig. S2.

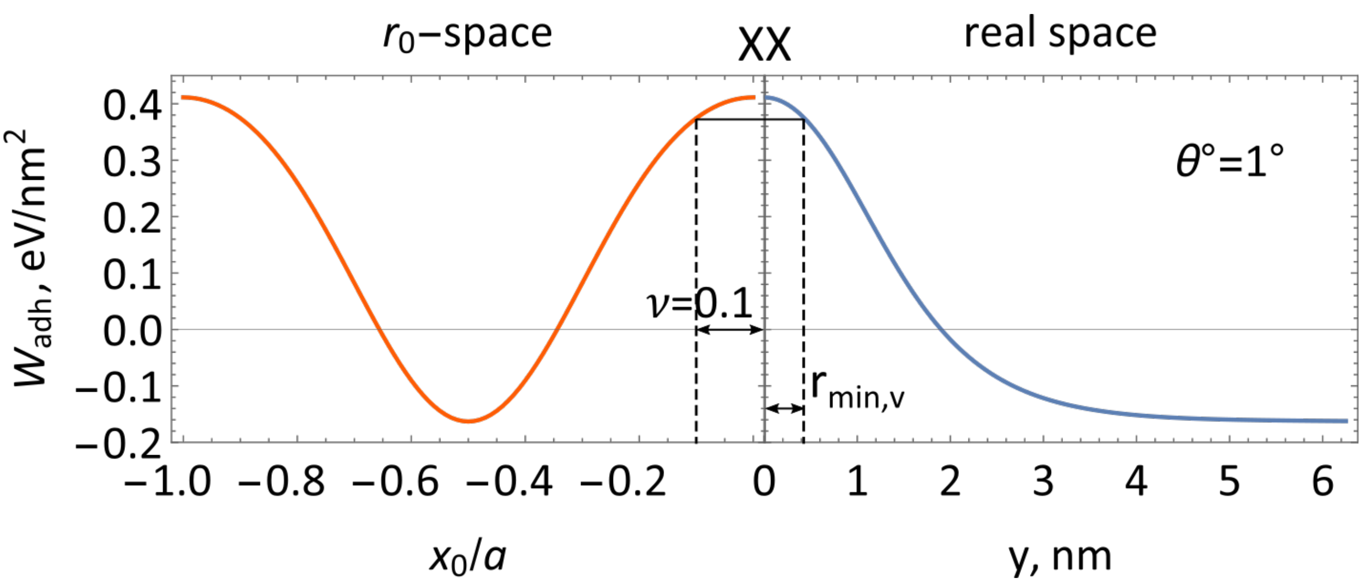

Figure S2. Adhesion energy for P WSe_2 bilayers in vicinity of XX stacking area, with the middle at the coordinate origin ($x_0 = y = 0$), as a function of $\boldsymbol{r}_0 = (x_0, 0)$ (left panel) and real space coordinate (right panel) for moiré superlattice at $\theta^\circ = 1^\circ$. Setting $\nu = 0.1$ we define $r_{\mathrm{min},\nu}$ as a root of Eq. (S7).