Stacking Domains and Dislocation Networks in Marginally Twisted Bilayers of Transition Metal Dichalcogenides

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We apply a multiscale modeling approach to study lattice reconstruction in marginally twisted bilayers of transition metal dichalcogenides (TMD). For this, we develop density functional theory parametrized interpolation formulae for interlayer adhesion energies of MoSe₂, WSe₂, MoS₂, and WS₂, combine those with elasticity theory, and analyze the bilayer lattice relaxation into mesoscale domain structures. Paying particular attention to the inversion asymmetry of TMD monolayers, we show that $3R$ and $2H$ stacking domains, separated by a network of dislocations develop for twist angles $\theta^\circ < \theta_p^\circ \sim 2.5^\circ$ and $\theta^\circ < \theta_{AP}^\circ \sim 1^\circ$ for, respectively, bilayers with parallel (P) and antiparallel (AP) orientation of the monolayer unit cells and suggest how the domain structures would manifest itself in local probe scanning of marginally twisted P and AP bilayers.

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Layer-by-layer assembly of van der Waals (vdW) heterostructures of two-dimensional crystals became a popular method of creating new hybrid materials [1]. The underlying physics of optoelectronic properties of such systems includes the interlayer hybridization of electronic states of the layers and the superlattice effects, produced by the periodic moiré patterns characteristic to pairs of mutually twisted or slightly incommensurate lattices. Such effects have been extensively investigated in graphene-hBN heterostructures [2–6] and twisted graphene bilayers [7–9], and these studies have identified two distinct structural forms of bilayers. One corresponds to larger twist angles, θ , and a stronger lattice mismatch, δ , for which the periodic variation of local stacking of the atoms (moiré pattern with period $\ell = a/\sqrt{\delta^2 + \theta^2}$) is due to the superposition of rigid crystalline lattices of the two layers [2–9]. The other, “marginally twisted bilayers” [10] regime is peculiar to a very small misalignment in graphene bilayers, which reconstruct into large-area Bernal stacking domains, separated by networks of domain walls [11–18].

Beyond graphene and hBN, moiré superlattice effects have been observed in $MX_2/M'X'_2$ heterostructures of transition metal dichalcogenides (TMD) [19–25], and it has been suggested theoretically [26–28] that twisted bilayers of TMD can undergo lattice reconstruction. Here, we determine parametric conditions for the formation of and the types of domain structures in twisted TMD homo- and heterobilayers, enriched by the lack of inversion symmetry of the individual 2D crystals. Different domain wall networks form for bilayers with parallel (P) and antiparallel (AP) orientations of unit cells in the two layers (shown on top of Fig. 2), and we find crossover angles, $\theta_{AP}^\circ \sim 1.0^\circ$ and $\theta_p^\circ \sim 2.5^\circ$, for the marginality of the twist and, then, discuss how the resulting domain structures can be observed in scanning tunneling experiments.

In this study we employ the multiscale modeling: a combination of density functional theory (DFT) leading to interpolation formulae for adhesion energy, $W_{P/AP}(\mathbf{r}_0, d)$, between the layers at a distance d from each other and lateral offset $\mathbf{r}_0$, and elasticity theory for the lattice relaxation [27]. We perform this analysis for small misalignment angles, $\theta \ll 1$ (i.e., $\theta^\circ < 5^\circ$), and lattice mismatch $\delta \ll 1$. In this case energetics of local stacking can be described in terms of a lateral offset, $\mathbf{r}_0(\mathbf{r}) = \theta \hat{z} \times \mathbf{r} + \delta \mathbf{r} + \mathbf{u}^{(t)} - \mathbf{u}^{(b)}$, between two aligned commensurate lattices, which varies across the moiré supercell and includes the effect of local deformations, $\mathbf{u}^{(b/t)}(\mathbf{r})$, in the bottom and top layers. This multiscale approach enables us to

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FIG. 1. (a) Sketch of lattice relaxation across moiré supercell of marginally twisted AP-bilayers ($\theta < \theta_{AP}$). Insets show local stacking configurations in $2H$ domain and MM' seed. $X(X')$ and $M(M')$ label chalcogen and metal atoms, respectively, in the bottom (top) layer; (b) the left (right) panel reveals side view of $3R$ - ($2H$ -) stacked TMD bilayers with the size of dots reflecting the layer asymmetry of the electronic states at the conduction band edge. Central panel: the first Brillouin zone of the TMD bilayers with marked conduction (CB_Q , CB_K) and valence band (VB_Γ , and VB_K) extrema.

overcome the system-size limitations of molecular dynamics simulations [26,28].

For adhesion energy, we use the form, $W_{P/AP}(\mathbf{r}_0, d) = \sum_n f_n^{(P/AP)}(d) e^{i\mathbf{G}_n \mathbf{r}_0}$, where $\mathbf{G}_n$ are the reciprocal lattice vectors of TMD. We truncate this sum at the first star of reciprocal lattice vectors, $\pm \mathbf{G}_{1,2,3}$ ($|\mathbf{G}_{1,2,3}| = G$, Fig. 1) and set $\mathbf{r}_0 = 0$ at the XX' stacking for both P and AP bilayers. This choice—together with the D_{3h} lattice symmetry of TMD monolayers—suggests [29] that $W_P = f(d) + f_1(d) \sum_{n=1}^3 \cos(\mathbf{G}_n \mathbf{r}_0)$ and $W_{AP} = \tilde{f}(d) + \sum_{n=1}^3 [\tilde{f}_1(d) \cos(\mathbf{G}_n \mathbf{r}_0) + \tilde{g}_1(d) \sin(\mathbf{G}_n \mathbf{r}_0)]$. Then, we inspect the adhesion energies for various bilayers, computed using vdW DFT with the OPTB88 functional [30] implemented in QUANTUM ESPRESSO [43] for stacking configurations shown in Fig. 2. For P bilayers, the most energetically favorable are configurations MX' [$\mathbf{r}_0 = (0, -a/\sqrt{3})$] and XM' [$\mathbf{r}_0 = (0, a/\sqrt{3})$], which have equal energies [44] and correspond to twins of a $3R$ bulk phase of a TMD. For AP bilayers, $2H$ stacking [$\mathbf{r}_0 = (0, -a/\sqrt{3})$] has the lowest energy (in agreement with the $2H$ bulk phase of these TMDs), rather than MM' stacking [configuration 5, $\mathbf{r}_0 = (0, a/\sqrt{3})$] suggested in Ref. [27]. Configurations 6, $\mathbf{r}_0 = -a/3(1, \sqrt{3})$ and $\mathbf{r}_0 = -a/3(1, 0)$, are such that $W_P = f$ and $W_{AP} = \tilde{f}$. The remaining two (2 and 4) have

FIG. 2. VdW-DFT data for $\text{MoSe}_2/\text{MoSe}_2$ bilayers and their fits using $W_{P/AP}$ in Eq. (1) (for $\text{WSe}_2/\text{WSe}_2$, MoS_2/WS_2 , $\text{MoS}_2/\text{MoS}_2$, WS_2/WS_2 , and MoS_2/WS_2 bilayers see the Supplemental Material [30]). Fitting parameters are listed in Table S1 in the Supplemental Material [30].

offsets $\mathbf{r}_0 = (0, -a/2\sqrt{3})$ [$\mathbf{r}_0 = (0, a/2\sqrt{3})$] and $\mathbf{r}_0 = (0, a/2\sqrt{3})$ [$\mathbf{r}_0 = (0, a\sqrt{3}/2)$] for AP(P) bilayers.

Using the DFT data for bilayers shown in Fig. 2 and S1 in the Supplemental Material [30], we find the d dependence of the factors $f, \tilde{f}, f_1, \tilde{f}_1$, and $\tilde{g}_1$, and plot them in Fig. S2. By inspection, we find that $f \approx \tilde{f}$ and $f_1 \approx \tilde{f}_1 + \tilde{g}_1$, over the broad interval of interlayer distances that covers the minima of $W_{P/AP}(\mathbf{r}_0, d)$, and, then, consider the following factors to make an informed choice of functions, $\tilde{f}, \tilde{f}_1$, and $\tilde{g}_1$. (i) Coulomb potential of a lattice of $\pm q$ ions, whose potential decays exponentially, $\sum_n \alpha_n e^{-G_n d} e^{i\mathbf{G}_n \mathbf{r}_0}$, with the distance from their plane ($\alpha_0 = 0$ is due to the electroneutrality of each layer). This suggests a choice of $\tilde{g}_1 = A_2 e^{-Gd}$ for the first star of reciprocal lattice vectors $\pm \mathbf{G}_{1,2,3}$. (ii) The overlap of atomic orbitals, strongest for chalcogen atoms in the outer (top and bottom) sublattices in each layer is determined by the exponential decay of atomic wave functions away from the plane, $|\psi(z)|^2 \propto e^{-|z|/\rho}$. Also, tunneling between the layers is suppressed for electrons with a larger in-plane wave number [45], $\sum_n \beta_n e^{-d\sqrt{G_n^2 + \rho^{-2}}} \cos(\mathbf{G}_n \mathbf{r}_0)$, so that we choose $\tilde{f}_1 = A_1 e^{-d\sqrt{G^2 + \rho^{-2}}}$. (iii) Finally, following the earlier studies [46] of vdW interaction of TMDs, which has a long-distance asymptotic, $\propto -C/d^4$, we combine the short range repulsion and long-range vdW attraction into $\tilde{f} = -\sum_{n=1}^3 C_n/d^{4n}$. Then, we use

$$W_{P/AP}(\mathbf{r}_0, d) = \sum_{n=1}^3 \left[-\frac{C_n}{d^{4n}} + A_1 e^{-\sqrt{G^2 + \rho^{-2}} d} \cos(\mathbf{G}_n \mathbf{r}_0) + A_2 e^{-Gd} \sin(\mathbf{G}_n \mathbf{r}_0 + \varphi_{P/AP}) \right], \quad (1)$$

with $\varphi_P = \pi/2$, $\varphi_{AP} = 0$, and fit the values of parameters $C_{1,2,3}$, $A_{1,2}$, ρ to the DFT data listed in Table S1 in the Supplemental Material [30].

Lattice reconstruction in bilayers happens when energy gain from the formation of favorable stacking overcomes

elastic energy cost of strain produced by the local mutual adjustment of the two lattices, $U = \sum_{l=t,b} [(\lambda_l/2)(u_{ii}^{(l)})^2 + \mu_l(u_{ij}^{(l)})^2]$. Here, $\lambda_{l/b}$, $\mu_{l/b}$, and $u_{ij}^{(t/b)} = \frac{1}{2}(\partial_j u_i^{(t/b)} + \partial_i u_j^{(t/b)})$, are the first Lamé coefficient, shear modulus, and strain tensors related to the local in-plane displacements in top and bottom layers. Values of Young's moduli and Poisson ratios determining λ and μ coefficients for TMD crystals under consideration are listed in Table S3 in the Supplemental Material [30]. We neglect the energy cost of flexural deformations [47], see Sec. S3 in the Supplemental Material [30] allowing for the out-of-plane bending of the layers towards the optimal interlayer distance, $d_{P/AP}(\mathbf{r}_0)$, for each offset $\mathbf{r}_0$. We describe the latter by expanding $W_{P/AP}$ into Taylor series about the minimum point, d_0 , of the zeroth Fourier harmonic term, $f(d) \approx f(d_0) + \varepsilon(d - d_0)^2$ (Table S1 in the Supplemental Material [30]), and, then, find $\mathbf{u}^{(t/b)}(\mathbf{r})$ that minimize energy,

$$\begin{aligned} \mathcal{E} = \int d^2\mathbf{r} \Big\{ & U - \varepsilon Z_{P/AP}^2 \\ & + \sum_{n=1}^3 [A_1 e^{-\sqrt{G^2 + \rho^{-2}} d_0} \cos(\mathbf{g}_n \mathbf{r} + \mathbf{G}_n [\mathbf{u}^{(t)} - \mathbf{u}^{(b)}]) \\ & + A_2 e^{-G d_0} \sin(\mathbf{g}_n \mathbf{r} + \mathbf{G}_n [\mathbf{u}^{(t)} - \mathbf{u}^{(b)}] + \varphi_{P/AP})] \Big\}; \\ Z_{P/AP} = \frac{1}{2\varepsilon} \frac{\partial}{\partial d} [f(d) - W_{P/AP}(\mathbf{r}, d)] \Big|_{d=d_0}. \end{aligned} \quad (2)$$

Here, $\mathbf{g}_n = \delta \mathbf{G}_n - \theta \hat{z} \times \mathbf{G}_n$ are reciprocal lattice vectors of moiré superlattice, which we also use to expand $\mathbf{u}^{(t/b)}(\mathbf{r})$ in Fourier harmonics up to the 80th reciprocal space star. Then, we minimize $\mathcal{E}$ with respect to those Fourier amplitudes numerically and obtain the displacements in each layer of the reconstructed bilayer. Using this method, we study moiré structure with $\theta^\circ \geq 0.2^\circ$.

Representative examples of lattice reconstructions in P and AP bilayers are shown in Fig. 3. For a larger angle, $\theta^\circ = 4^\circ$ [Figs. 3(a) and 3(b)] adhesion-induced displacements are small and the two layers behave as rigid lattices. For $\theta^\circ < 1^\circ$, twisted bilayers reconstruct into domain structures. For P bilayers [Fig. 3(d)], each reconstructed moiré supercell comprises two equal area triangular domains of $3R(XM'/MX')$ stacking, separated by partial dislocations, with XX' regions squeezed to the nodes on that partial dislocations network [Fig. 3(g)]. For AP bilayers [Fig. 3(c)], the reconstructed lattice features a honeycomb array of $2H$ domains separated by a dislocation network, where one half of the nodes are the seeds of the quasiequilibrium MM' phase (stacking configuration AP-5). As a quantitative measure for domain formation, we use a lateral distance, $\mathfrak{N}$, between the closest metal and sulfur atoms in top and bottom layers (for ideal $2H$ and $3R$

FIG. 3. (a)–(d) Reconstructed lattices of $\text{WSe}_2/\text{WSe}_2$ AP(P) bilayer with $\theta^\circ = 4^\circ > \theta_{AP/P}$ and $0.6^\circ (0.7^\circ) < \theta_{AP/P}$ twist (for $\theta^\circ = 0.2^\circ$ see the Supplemental Material [30]). For $\theta > \theta_{AP/P}$ the two layers behave as rigid, for marginally twisted bilayers ($\theta \rightarrow 0$) $2H$ for AP and $3R$ for P stacking domains emerge, separated by dislocations. (e)–(g) Intersections of dislocations, drawn over on the color maps of the varied interlayer distance. (h) Crossover from rigid twist to the fully developed domain structure in P and AP bilayers, quantified using average value of parameter $\mathfrak{N}^2$ described in the text.

domains, $\mathfrak{N} = 0$) and compute its mean square, $\overline{\mathfrak{N}^2}/a^2$, over the supercell normalized by the TMD lattice constant, a . For rigidly rotated monolayers, $\overline{\mathfrak{N}^2}_{AP} \approx 0.14a^2$ and $\overline{\mathfrak{N}^2}_P \approx 0.08a^2$. Upon the formation of $2H$ and $3R$ domains, main contribution to $\overline{\mathfrak{N}^2}$ comes from domain boundaries so that the asymptotic behavior, $\overline{\mathfrak{N}^2} \propto 1/\ell \propto \theta$, in Fig. 3(h) signals the formation of a domain structure at $\theta < \theta_p \approx 0.044 (2.5^\circ)$

FIG. 4. (a),(b) Dependence of domain wall (DW) energy per unit length (blue) and width (red) on the angle between DW axis and zigzag direction (x axis) in TMD layer, α , for AP and P bilayers, respectively. (c) and (d) describe DW (dislocation) profile for AP and P bilayers, respectively. The calculated profiles for various TMDs (shown by different colors) almost coincide. Here, $\xi = a\sqrt{\mu/2A_1} \exp[d_0\sqrt{G^2 + \rho^{-2}}/2]$.

and $\theta < \theta_{AP} \approx 0.017(1^\circ)$. These quantitative estimates also explain why the molecular dynamics simulations performed in Refs. [26,28] for $\theta > 3^\circ$ failed to establish the full picture of the lattice reconstruction in heterostructures of 2D crystals, having interpreted MM' areas in almost rigid AP bilayers [Fig. 3(a)], as fully developed domains. Note that superlattice patterns—perfect domains and the dislocation network—also appear in TMD heterobilayers with the same chalcogens (WS₂/MoS₂ or WSe₂/MoSe₂), which have lattice mismatch $\delta \lesssim 0.3\%$, whereas bilayers with $\theta > \theta_{P/AP}$, or MS₂/M'Se₂ heterostructures with $\delta \approx 3.8\%$ behave as almost rigid crystals.

The formation of domain structures takes place when the energy gain from the expanded 2H (for AP) or 3R (for P) areas overcomes the energy cost of domain walls. The latter are nothing but dislocations: screw dislocations in 2H and

partial screw dislocations in 3R bilayers. The properties of such linear defects, analyzed using energy functional (2), are shown in Fig. 4. Here, we set $\theta = 0$ in Eq. (2) and replace $u^t - u^b$ by $\mathfrak{N}$, such that $\mathfrak{N}(-\infty) = 0$ and $\mathfrak{N}(+\infty) = b_{AP/P}$, where $b_{AP} = a(-1, 0)$ is a Burgers vector of a dislocation in 2H TMD ($b_{AP} = a$), and $b_P = a(0, 1/\sqrt{3})$ is a partial dislocation Burgers vector in 3R bilayer ($b_P = a/\sqrt{3}$). A vector $\mathbf{n} = (\cos \alpha, \sin \alpha)$ determines the orientation of the dislocation line with respect to zigzag axis in the crystal. We find (see Fig. 4) that the energy of a dislocation in 2H homobilayers is the lowest and the cross sectional width narrowest for a zigzag orientation of the defect line (screw dislocation); for 3R homobilayers, the most favorable orientation of the partial dislocation axis is along the armchair direction (partial screw dislocation). These choices coincide with the orientations of domain boundaries shown in Fig. 3. In addition, in Fig. S8, we show the profile of edge dislocations that would form in perfectly aligned ($\theta < \delta$) heterobilayers MS₂/M'Se₂ and MSe₂/M'Se₂ [48].

As a consequence of the domain formation, optoelectronic properties of marginally twisted bilayers would be dominated by the areas of 2H and 3R stacking. While 2H bilayers have been deeply explored both theoretically [49–55] and experimentally [56–67], 3R bilayers were studied less [57,64,68–76], despite that the latter have interesting features resulting from their lack of inversion symmetry. Table I gives the atomic decompositions of the states at the edges of conduction and valence bands, computed using DFT for 3R bilayers (see the Supplemental Material for details [30]), highlighting the interlayer asymmetry of the states near band edges marked in Fig. 1(b). Such an asymmetry would make tunnelling characteristic of MX' and XM' domains different, leading to a manifestation of domain formation in conductive atomic force microscopy [77] as well as to a linear Stark shift, $[d_z(\text{CB}_Q) - d_z(\text{VB}_\Gamma)]E$, for the lowest-energy exciton transition in the bilayer, with the opposite sign of the shift in MX' and $M'X$ stacking domains [78].

TABLE I. Orbital decomposition of DFT states at conduction (CB_Q) and valence (VB_Γ) band edges of 3R homobilayers of MX₂ [as labeled in Fig. 1(b)], the corresponding out-of-plane electric dipole moment, d_z , and ratio between tunneling currents into MX' and XM' stacking domains.

	$ \psi ^2[\%]$	$\tilde{X}$	M	X	X'	M'	$\tilde{X}'$	d_z (eÅ)	$I_{MX'}:I_{XM'}$
MoS ₂	CB _Q	4.2	30.6	3.3	5.5	49.6	6.8	0.73	1:1.6
	VB _Γ	10.6	41.8	0.9	0.7	36.0	10.0	−0.20	1:1
MoSe ₂	CB _Q	6.8	29.7	4.3	6.5	42.6	10.0	0.58	1:1.6
	VB _Γ	14.9	36.6	1.2	1.0	31.9	14.3	−0.17	1:1
WS ₂	CB _Q	4.3	28.6	4.0	6.8	48.9	7.4	0.81	1:1.7
	VB _Γ	10.3	41.5	1.6	1.3	35.5	9.9	−0.21	1:1
WSe ₂	CB _Q	7.0	24.4	6.2	10.3	40.6	11.5	0.80	1:1.6
	VB _Γ	14.5	36.3	2.2	1.8	31.4	13.7	−0.19	1:1

FIG. 5. (a),(b) Distribution of piezoelectric charge density and pseudomagnetic field induced in AP bilayers MoS₂/MoS₂ (θ = 0.2°).

Lacking inversion symmetry, TMD monolayers are piezoelectric crystals. The inhomogeneous strain concentrated in each layer at domain walls results in piezocharges with density, $\rho_{\text{piezo}}^{(t/b)} = e_{11}[2\partial_x u_{xy}^{(t/b)} + \partial_y(u_{xx}^{(t/b)} - u_{yy}^{(t/b)})]$. In P bilayers, the opposite signs of strain fields in the top and bottom layers leads to the mutual compensation of their piezoelectric charges. In AP bilayers the inversion of the sign $e_{11}^t = -e_{11}^b$ of piezoparameter in the two layers adds up charges, to the values shown in Fig. 5(a), computed after taking into account partial screening of piezoelectric field by dielectric polarizability of TMD layers (see in the Supplemental Material [30] for details), with opposite signs in MM' to XX' corners of the domain wall network. Then, the MM' regions and $2H/2H$ DWs will attract electrons in n -doped AP homobilayers, giving rise to a larger tunnel current response than inside the $2H$ domains [77]. These spots of piezocharges, with the values up to $\sim 10^{12} \text{ cm}^{-2}$, can be viewed as “quantum dots” for electrons and holes in the vicinity of the band edges of the bilayers. The same areas are also the hot spots of pseudomagnetic fields [$B_* \propto \rho_{\text{piezo}}^{(t/b)}$, Fig. 5(b)], generated by inhomogeneous strain for electrons in multivalley semiconductors [79], which makes the dislocation network sites (MM' and XX' regions) interesting objects for optoelectronic studies.

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- [29] This is because aligned P-oriented TMD bilayers with opposite in-plane offsets represent mirror copies of each other resulting in the same interaction value for $\pm r_0$; whereas for AP orientation, the lack of inversion symmetry in each layer allows for both even and odd terms.
- [30] See the Supplemental Material at <http://link.aps.org/supplemental/10.1103/PhysRevLett.124.206101> for details of DFT calculations, values of fitted parameters, and piezocharge density calculations, which includes Refs. [31–42].
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