Reconstruction of moiré lattices in twisted transition metal dichalcogenide bilayers

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Abstract

The introduction of a small twist angle (θ) between two layers of two dimensional materials gives rise to a large-scale lattice (moiré), which can significantly alter its electronic properties. These moiré lattices can provide a simple platform to study strongly correlated electron physics [1–3]. An important step in understanding these electronic properties is the inclusion of the effects of structural relaxation on the un-relaxed twisted structures. Here, we propose candidate structures for twisted bilayer of transition metal dichalcogenides (TMDs). For θ ≥ 58.4°, we find dramatic reconstruction of the moiré lattices. The moiré lattice constant of the reconstructed structure is \sqrt{3} times that of the un-relaxed twisted structure. We show that the development of curved domain walls due to three-fold symmetry of stacking energy landscape, is responsible for such reconstruction.
The formation of flat bands in the electronic band structure of twisted bilayer of two-dimensional materials is central to understanding the observed exotic electronic phases\[^{1-4}\]. Twisted bilayer TMDs, an important class of materials, can possess flat bands for a continuum of twist angles\[^{5-11}\]. To accurately calculate the electronic band structure, incorporation of structural relaxation effects is crucial \[^{5, 6, 12, 13}\]. Typically, these relaxations are performed by starting from a configuration and \textit{only} allowing downhill motion in the potential energy landscape using local search algorithms (standard minimization protocol). Since the number of local minima in the potential energy landscape increases exponentially with the number of atoms, standard minimization protocols are often insufficient to find stable structures\[^{14-16}\]. Irrespective of the details of relaxation, the moiré lattice periodicity of the un-relaxed twisted structures is generally believed to remain intact for any $\theta$.

We obtain candidate structures using simulated annealing with classical molecular dynamics and demonstrate reconstruction of moiré lattices of twisted bilayer (tBL) of TMDs as $\theta \gtrsim 58.4^\circ$. We show that, the assumption of moiré lattice constant of un-relaxed twisted structures remaining intact is not always true. Such lattice reconstructions are absent in standard minimization approaches. We discuss the details of lattice reconstruction for tBL of MoS$_2$ and confirm our conclusions for MoSe$_2$, WSe$_2$, WS$_2$ (see Supplementary Information (SI), Sec. I).

Due to the presence of different sub-lattice atoms (Mo/W, S/Se) in TMD, tBL possesses distinct high-symmetry stacking regions for $\theta$ near $0^\circ$ (AA, AB, BA) and near $60^\circ$ (AA’, AB’, A’B)\[^{17}\]. However, the lattice constant of the un-relaxed tBL is identical for $\theta$ and $60^\circ - \theta$ (e.g. $1^\circ$ and $59^\circ$). Among these stackings, AB is energetically the most favourable stacking as $\theta \to 0^\circ$ ($E_{AB} = E_{BA} < E_{AA}$, six-fold symmetric around AA) and AA’ for $\theta \to 60^\circ$ ($E_{AA'} < E_{AB'} < E_{A'B}$, reduced three-fold symmetric around A’B)\[^{18, 19}\]. Relaxation of the tBL leads to significant increment of the area of the most favourable stacking regions \[^{20}\].

In Fig. 1 we show the interlayer separation (ILS) landscape for tBL of MoS$_2$ for several $\theta$ using both standard relaxation (SR) and simulated annealing (SA) with $3 \times 3 \times 1$ superlattice. As a representative of $\theta \to 0^\circ$, we show the ILS landscape for $\theta = 1^\circ$. With SR we find straight domain walls, which separate AB and BA stacking regions and form a hexagonal network (Fig. 1a, top panel). On the other hand, the ILS landscape computed with SA shows curling of domain walls near AA region (both clockwise and counter-clockwise curling are present, Fig. 1a bottom panel). Although, the number of clockwise and counter-
FIG. 1: Interlayer separation landscape using standard relaxation (top panel) and simulated annealing (bottom panel) for tBL of MoS$_2$ (using Mo atoms). The smallest repetitive cell in the top panel is a moiré lattice. The scales of the associated colorbar are in Å and denotes interlayer separation. The curling of domain walls near AA, A′B are marked (only few).

clockwise curling are equal, they do not always form a checkerboard-like pattern. While the checkerboard pattern is the lowest in energy, the total energy difference between the checkerboard pattern and random distribution of curling are small (few meV per moiré lattice). Nevertheless, the AA stacking always form a triangular lattice for any $\theta$ close to $0^\circ$.

In contrast, the behaviour of the ILS landscape show intriguing features for $\theta \rightarrow 60^\circ$. We categorize the $\theta$ dependence into two regions. Region I ($\theta < 58.3^\circ$) : With SR both the AA′ and AB′ occupy comparable area of the moiré lattice, with each forming an approximate equilateral triangle (Fig. 1b). Similar to $\theta \rightarrow 0^\circ$, the ILS landscape obtained with SA shows curling of domain walls near A′B stacking (Fig. 1b, bottom panel). Region II ($\theta \gtrsim 58.4^\circ$) : The most favorable AA′ stacking increases in area significantly and evolves from Reuleaux triangle to approximate hexagonal structures obtained with SR (Fig. 1c-1e, top panel), consistent with previous report [19]. In this case, the domain walls connecting A′B stacking regions are significantly curved and never straight-lines. We find that, these structures show notable reconstruction with SA. In particular, a triangular lattice is formed with three A′B stacking regions trimerizing to form a motif (Fig. 1c-1e, bottom panel). Moreover, the domain walls connecting different A′B stacking are almost straight in the reconstructed
structures. The relaxed structures obtained using SA, are always energetically more stable than those obtained using SR. Also, the number of A′B stacking and domain walls connecting them are always conserved.

FIG. 2: (a)-(b) Radial distribution function, \( g_m(r) \) computed (using minimum image convention) with standard relaxation (SR) and simulated annealing (SA). The moiré lattice constants are marked (blue triangle pointing up for SR, red triangle pointing down for SA). (c) Schematics of tBLMoS\(_2\) as \( \theta \rightarrow 60^\circ \) with SR. (d) Change of \( p/a_m \) with \( a_m \). Several ideal geometric structures are marked with dashed lines.

In order to investigate the structural long-range order of tBL we compute the radial distribution function with both SR and SA. In the un-relaxed tBL there are two distinct length scales, one for the individual TMD layer with lattice constant \( a \), and another for the moiré lattice with \( \theta \) dependent lattice constant, \( a_m = a/(2 \sin(\theta/2)) \). Therefore, one can define two separate radial distribution functions, one for atoms of individual TMD layer and another for stackings of the moiré lattice. As an example, we compute the moiré scale radial distribution function, \( g_m(r) \) using the AA/A′B stacking regions of tBLMoS\(_2\) (Fig. 2a, 2b). Each AA/A′B region represents a moiré lattice point (MLP). Identifying the MLPs from the ILS landscape, we calculate the \( g_m(r) \) defined as, \( \langle N(r+\delta r) \rangle \) with \( \langle N(r+\delta r) \rangle \) representing the average number of MLP within a ring of radius \( r \), width \( \delta r \) and the area of the ring \( A(\delta r) \).

For \( \theta \rightarrow 0^\circ \), \( g_m(r) \) are similar for both SR and SA (Fig. 2a). The average number of nearest neighbour MLPs is always 6, calculated by integrating the first peak of \( g_m(r) \). This confirms the existence of the hexagonal network formed by domain walls (Fig. 1a). Furthermore, the moiré lattice constant calculated from \( g_m(r) \) is identical to that of un-relaxed tBLMoS\(_2\).
Therefore, the long-range order of the un-relaxed structures remains intact as $\theta \to 0^\circ$. As $\theta \to 60^\circ$ for Region I, the moiré lattice constants are identical for un-relaxed and relaxed structures, $a_m = a_{m}^{SR} = a_{m}^{SA}$ (Fig. 2a), and the number of nearest neighbour MLP is always 6. The reconstruction of the moiré lattices leads to the formation of a triangular lattice with modified lattice constant, $a_{m}^{SA} = \sqrt{3}a_m$ in Region II (Fig. 2a, 2b). The first peak in the $g_m(r)$ ($\approx 7.5$ nm) denotes the motif of the triangular lattice. The motif consists of 3 A'B regions. By integrating the first peak of $g_m(r)$ we find that, the number of nearest neighbour of A'B region is 2.

We also examine the atomic-scale radial distribution function for individual MoS$_2$ layer (considering the Mo atoms). For both $\theta \to 0^\circ$ and $\theta \to 60^\circ$, the long-range order is preserved at the unit-cell MoS$_2$ level. This clearly establishes that, the aforementioned reconstruction in Region II is an emergent phenomenon arising at the moiré scale.

In order to pinpoint the onset of lattice reconstruction geometrically, we consider the ratio of perpendicular bisector, $p$ to $a_m$ of twisted structures obtained using SR (Fig. 2c). Interestingly, as $p/a_m$ becomes $\gtrsim 1$ we notice reconstruction of the moiré lattices (Fig. 2d). When $p/a_m = 1$ (for $\theta \sim 58.5^\circ$), the AA' stacking represents a Reuleaux triangle with the domain walls occupying it’s perimeter. When one considers perimeter$^2$ to area ratio, the Reuleaux triangle is a local maximum [21]. Since the domain walls are energetically unfavourable than the AA' region, the Reuleaux triangle is expected to undergo rearrangements to minimize total energy. The shortest distance between two A'B region is $\approx d_{A'B} + d_{AB'} \approx 3.3 + 4.5 = 7.8$ nm, where $d_{A'B}, d_{AB'}$ denote linear dimension of the corresponding stackings. This explains the occurrence of first peak in $g_m(r)$ in Region II at $\approx 7.5$ nm.

Now, we investigate the origin of these reconstructions from energetics and correspondingly, the greater stability of structures obtained with SA. The total energy of tBL of MoS$_2$ is a sum of the intra-layer elastic energy, which is a combination of strain and bending energy[22] and interlayer stacking dependent energy i.e. $E = E_{\text{intra}} + E_{\text{inter}}$. For $\theta \to 60^\circ$, the interlayer energy per MLP evaluated with respect to AA' stacking can be approximated as,

$$E_{\text{inter}} - E_{\text{inter}}^{AA'} = \delta E_{\text{inter}}^{A'B} S_{A'B} + \delta E_{\text{inter}}^{DW} S_{DW} + \delta E_{\text{inter}}^{AB'} S_{AB'}$$

(1)

Here, $\delta E_{\text{inter}}^{\alpha}$ represents the interlayer energy of stacking $\alpha$, evaluated with respect to AA' and $S^{\alpha}$ denotes the occupied area. For small twist angles, $S_{A'B}, S_{AB'}$ and the width of the domain wall (DW), $w$ becomes constant ($S_{DW} = wl$). Therefore, the interlayer energy as in
Eqn. (1) becomes linear with domain wall length, $l$ and is repulsive. Moreover, the intralayer strain energies are concentrated on the domain walls, also referred to as solitons and scales as $\propto l/w$ \cite{23, 24}. Thus, the minimization of $l$ will minimize both the interlayer and intralayer energies. In Fig. 3a we show the scaling of the total energy measured with respect to the stable stacking, with $a_m$ using SR for both $\theta$ near $0^\circ$ and $60^\circ$. The domain walls obtained with SR are always significantly curved for $\theta > 58.4^\circ$. The length of these curved domain walls can be minimized by reconstruction of the moiré lattices such that the domain walls become straight-lines (those in Fig. 1c-e, bottom panel). On the other hand, the domain walls are straight-lines for identical set of twist angles near $0^\circ$ with SR. Thus, $l$ per moiré lattice is already minimized. As a result, we do not find lattice reconstruction with SA as $\theta \rightarrow 0^\circ$. It should be noted that, the domain walls are always curled near AA, A’B stacking with SA, irrespective of lattice reconstruction. This originates from a buckling instability, primarily localized at AA, A’B (see below). Taking these into account, $l^{SA}$ is expected to be greater than $l^{SR}$ in the absence of lattice reconstruction. Such small curling of domain walls, however, does not change overall long-range order of moiré lattices (Fig 1a,1b). In Fig. 3b we show the estimate of $(l^{SA} - l^{SR})$ per MLP as $\theta \rightarrow 0^\circ$, $\rightarrow 60^\circ$ (SI, Sec. II for details). As $\theta \gtrsim 58.4^\circ$, the difference becomes negative, indicating reduction in domain wall length for the reconstructed lattice. The reduction in $l$, disregarding the curling of the domain walls with SA is (shown in Fig. 3b) large in Region II. Fig 3c shows the gain in total energy with SA relative to SR. The gain in total energy near $0^\circ$ arises from curling of the domain walls near AA, whereas the gain in Region II arises predominantly from lattice reconstruction. The energy gain per MLP in Region II is remarkably larger compared to identical set of $\theta$ near $0^\circ$.

To provide more insight into the origin of lattice reconstruction, we compute the vibrational modes of $1 \times 1 \times 1$ moiré lattice. The phason modes associated with tBL of TMDs originates from relative displacement of the constituent layers and their velocities ($d\omega/dq$ away from $\Gamma$) determine the rigidity of the moiré lattice\cite{20}. Although, at large $\theta$ the phason modes are similar for $\theta$ near $0^\circ$ and $60^\circ$\cite{20}, we find notable differences in the dispersion of these modes near the onset of lattice reconstruction. As an example, we highlight the dispersion of phason modes for $\theta = 1.5^\circ$ and $\theta = 58.5^\circ$ for tBL of MoS$_2$ computed with SR (Fig. 3d, 3e). One of the phason modes softens significantly and becomes nearly dispersion-less with attributes of zero mode\cite{25} for $\theta = 58.5^\circ$. Such modes are expected to cause reconstruction
FIG. 3: (a) Change of total energy computed with respect to stable stacking, $\alpha$ (AB near $0^\circ$, AA’ near $60^\circ$) using SR with $a_m$ (corresponding $\theta$ are marked). (b) Change in domain wall length with SA with respect to SR including (dashed lines) and excluding (solid lines) curling near AA/A’B. The error bar denotes standard deviation of the estimated change. (d) Total energy gain with SA compared to SR. (d),(e) Phonon dispersion with SR and SA for a moiré lattice for 1.5°, 58.5°, respectively. The solid blue, dashed red, solid red lines represent the acoustic, phason, buckling mode localized at AA/A’B, respectively. (f),(g) Schematics near the topological defect for tBL of TMDs for $\theta = 0.5^\circ$, 59.5°, respectively. The order parameter is shown with arrows.

Furthermore, with SR we find a soft mode with imaginary frequency for both $\theta = 1.5^\circ$ and 58.5° (Fig. 3d, 3e). The corresponding eigenvector at $\Gamma$ is localized on AA/A’B stacking and denotes a buckling instability. In this case, at AA (A’B) stacking both the constituent MoS$_2$ layers buckle out-of-phase with maximum ILS. However, the lattice reconstruction do not arise from such instability. To show this, we compute the phonon dispersion with SA of single moiré lattice, where no lattice reconstruction can arise. The buckling instability can be removed without lattice reconstruction (Fig. 3e).

We characterize the domain walls using the order-parameter, defined as the shortest displacement vector to take any stacking to the most unfavourable high-symmetry stacking (AA for $\theta \to 0^\circ$, A’B for $\theta \to 60^\circ$)\[^5\][^12][^23]. Irrespective of $\theta$, we find the domain walls to be
shear solitons (change in order parameter is along the domain wall as we go from AB → BA for θ → 0° and AA′ → AA′ for θ → 60°). For θ → 0°, the width of the domain walls are ≈ 2.9 nm for tBL of MoS₂. In the reconstructed lattice in Region II, two domain walls come close together and the effective width of the combination becomes ≈ 4.3 nm. Moreover, the order parameter rotates by 2π at the node where six domain walls meet (AA'/A'B), indicating its topological nature (Fig. 3f, 3g). For θ → 0° (θ → 60°), the width of the domain walls are: 2.9 (3.8) for tBL MoSe₂, 3.7 (4.7) for tBL WSe₂, 3.5 (4.5) for tBL WS₂, in nm.

While equilibration of molecular dynamics simulations we notice metastable structures, such as, distorted hexagon, kagome etc, which evolves to form structures shown in Fig 1c-e (SI, Sec. III). However, in experiment due to substrate or external constraints, this transformation may be slow. In order to find more candidate structures, we also simulate larger superlattice allowing significantly large degrees of freedom for lattice reconstruction. We find that, metastable structures with motifs of > 3 A'B stacking are also possible (SI, Sec. IV). However, the candidate structures have an abundance of the motifs shown in Fig. 1.

In conclusion, we propose novel structures for twisted bilayer TMDs. For θ ≳ 58.4°, the structures possess different long-range moiré periodicity than the un-relaxed twisted structures. These structures can be probed using electron microscopy[13, 23], optical imaging[26, 27] etc. and expected to be generic for materials with different sub-lattice atoms. Such reconstruction of moiré lattices is expected to significantly alter the electronic properties.

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Methods: Using the Twister code[5] we construct a superlattice of tBL of TMDs (0.2° ≤ θ ≤ 59.8° with ≥ 10⁶ atoms). We use Stillinger-Weber and Kolmogorov-Crespi potential to capture the intra and interlayer interaction of TMD layers respectively [18, 28]. We relax the rigidly twisted superlattice with target pressure, P = 0 bar in LAMMPS using standard minimization protocol[29–31], denoted as standard relaxation (SR). We perform classical molecular dynamics simulations using canonical ensemble with Nosé-Hoover thermostat at T = 1 K and cool down snapshots to 0 K, which is followed by an energy minimization. We find that, heating to higher temperature for annealing produces similar results. We refer to this approach as simulated annealing (SA). The radial distribution function is computed with 9 × 9 × 1 superlattice and averaged over 30 configurations. The phonon frequencies are
calculated using modified PHONOPY\cite{10} code.

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FIG. S1: Interlayer separation landscape using standard relaxation (top panel for all materials) and simulated annealing (bottom panel) computed with $3 \times 3 \times 1$ superlattice. The scales of the associated colorbar are in Å and denotes interlayer separation.
II: ESTIMATION OF DOMAIN WALL LENGTH

The estimation of domain wall length, \( l \) are done in four steps: (i) Identifying approximate region of only domain walls (ii) Defining a polygon that encloses all the points for a representative domain wall (iii) A suitable representation of the thick domain wall as a line. (iv) Averaging over the superlattice to get statistically significant result. We illustrate this in Fig. S2.

FIG. S2: (a) Interlayer separation landscape using simulated annealing approach for \( \theta = 59^\circ \). The scales of the associated colorbar are in Å and denotes interlayer separation. (b) Identification of the domain wall region from ILS landscape (c) Defining the polygon (solid black line) and finding a suitable representation of the domain wall (dashed blue line).
III : METASTABLE STRUCTURES AS $\theta \rightarrow 60^\circ$

FIG. S3: Interlayer separation landscape using simulated annealing approach for some metastable structures realized while equilibration of molecular dynamics simulations of a $3 \times 3 \times 1$ superlattice of tBL MoS$_2$. Similar results are obtained for other TMDs, as well. The scales of the associated colorbar are in Å and denotes interlayer separation. (a), (b) Kagome lattice (star of David), (c) distorted hexagon. These structures relax further and become a triangular lattice with 3 A'B stacking regions trimerizing to form a motif.
IV : INTERLAYER SEPARATION LANDSCAPE WITH LARGE SUPERLATTICE

FIG. S4: (a) Interlayer separation landscape using simulated annealing approach for \( \theta = 59^\circ \) with 10 \( \times \) 10 \( \times \) 1 moiré lattices (not a multiple of 3 A'B stackings) in the superlattice, (b) with 15 \( \times \) 15 \( \times \) 1 moiré lattices. The scales of the associated colorbar are in Å and denotes interlayer separation. Because of large degrees of freedom the lattice reconstruction can lead to such metastable structures. The annealed structures with 3 A'B stackings are energetically most favourable (shown in the main text). The energy difference between the structure shown in (b) and ordered structure with motif of 3 A'B is \( \sim 520 \) meV per moiré lattice.