

Dislocation Networks in Marginally Twisted Bilayer MoS₂Byunghyun Kim,[▽] Ayoung Yuk,[▽] Yunyeong Chang,[▽] Dongsin Kim,[▽] Daesung Park, Young Woo Choi,*
Miyoung Kim,* Moon Sung Kang,* and Hyobin Yoo*Cite This: *ACS Materials Lett.* 2026, 8, 1382–1388

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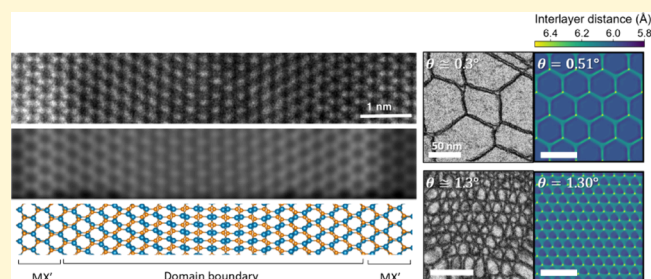


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ABSTRACT: In marginally twisted van der Waals homo- and heterostructures, spontaneous atomic reconstruction forms commensurate stacking domains separated by domain boundaries, which can be regarded as dislocations. Here, we employ dark field transmission electron microscopy combined with diffraction-contrast analysis to identify the Burgers vectors of domain boundaries within reconstructed domain boundary networks. Owing to the atomically thin nature of two-dimensional (2D) materials, diffraction contrast can be interpreted within a kinematical approximation, enabling simple and accessible analysis of dislocation character. In marginally twisted bilayer MoS₂ with antiparallel stacking, large hexagonal MX' domains are enclosed by networks of screw-type perfect dislocations. As the twist angle increases, these hexagonal domains evolve into triangular MX' domains, accompanied by a reorganization of the domain boundary from hexagonal networks of perfect dislocations to triangular networks of partial dislocations. Our work establishes an accessible framework for resolving dislocation networks and their twist-angle-driven evolution in reconstructed twisted 2D materials.



Manipulating the twist angle in two-dimensional (2D) van der Waals (vdW) homo- and heterostructures provides a powerful route to engineer moiré superlattices with tunable length scales.^{1–10} In marginally twisted vdW bilayers, competition between interlayer stacking energy and intralayer strain drives spontaneous atomic reconstruction, forming commensurate stacking domains separated by interconnected domain boundary networks.^{11–17} From a crystallographic perspective, these domain boundaries can be regarded as dislocations, whose atomic structure and network topology critically influence the emergent functionalities of reconstructed vdW interfaces.^{17–19}

In marginally twisted bilayer transition metal dichalcogenides (TMDs), reconstructed domain patterns, domain boundaries, and the functionalities they enable depend sensitively on the relative stacking alignment. Bilayers twisted near parallel (3R-like) alignment host alternating MX and XM domains connected by triangular networks of domain boundaries,^{20–24} whose structural dynamics give rise to moiré-domain antiferroelectric responses.^{22,23} In contrast, bilayers twisted near antiparallel (2H-like) alignment form MX' domains enclosed by hexagonal networks of domain boundaries,^{24–27} which host unconventional electronic responses, including piezoelectric responses,^{24,26,28} asymmetric tunneling conductivity across domain boundaries,²⁵ and electronic states distinct from those of bulk 2H crystals.^{29–31}

Despite their functional importance, how domain boundary networks evolve with twist angle remains poorly understood. While atomic reconstruction strength is known to weaken with

increasing twist angle, how this reduction reshapes the structure and dislocation character of domain boundary networks has not been systematically explored, largely due to the field-of-view limitation of conventional atomic scale structural probes including scanning transmission electron microscopy (STEM)^{24,32} and scanning tunneling microscopy (STM).^{30,33}

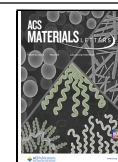
Here, we investigate marginally twisted bilayer MoS₂ (MTBM) with stacking configurations near antiparallel alignment using dark-field transmission electron microscopy (DF TEM) (Figure 1a). Interpreted within a kinematical diffraction framework,^{17,23,27,32,34–37} DF TEM enables direct identification of the dislocation character of individual domain boundaries while simultaneously visualizing their organization over extended length scales. Using this approach, we directly track the twist-angle-dependent structural evolution of reconstructed domain boundary networks from a large-scale dislocation perspective. Atomic-resolution annular dark-field scanning TEM (ADF STEM) is further employed to validate local stacking configurations. Together, this approach provides a robust framework for elucidating the twist-angle-dependent

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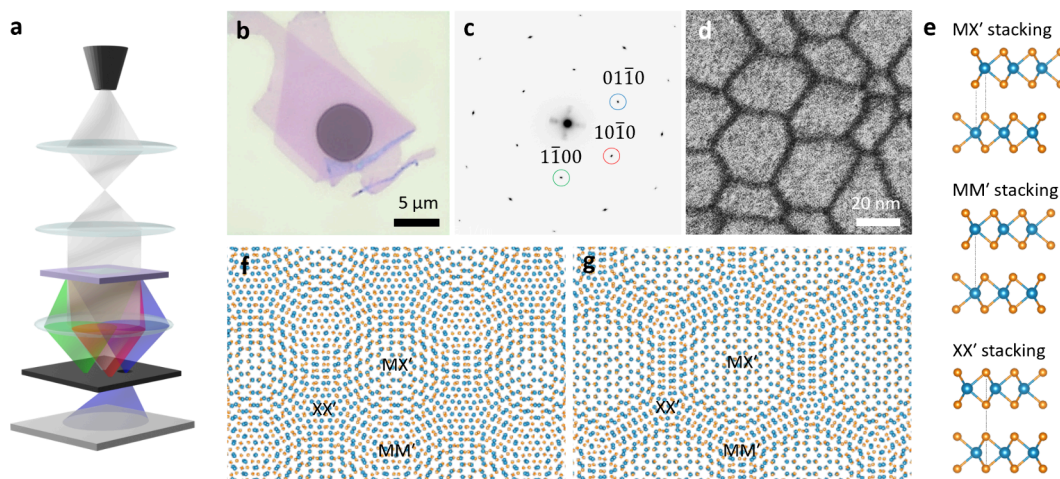


Figure 1. Atomic reconstruction in marginally twisted bilayer 2H TMDs. (a) Schematic DF TEM imaging process. A ray diagram shows electron beam (expressed in RGB colors) diffracted by crystal planes in the specimen. DF image is obtained by selecting a Bragg peak in the diffraction plane with an objective aperture (schematically represented as a dark perforated plate). (b) Optical microscope image of the MTBM suspended on a micropore ($6\ \mu\text{m}$ diameter) membrane. (c) SAED pattern of the MTBM. Bragg peaks marked with colored circles are used to form the DF image in (d). (d) Overlaid DF image obtained along the $[0001]$ zone axis, by integrating DF images from individual Bragg peaks in (c), corresponding to three sets of zigzag family planes. The image clearly shows hexagonal MX' domains and domain boundaries. (e) Side-view schematic diagrams of three high-symmetry stacking orders in bilayer 2H TMDs. Blue and orange atoms correspond to transition metal and chalcogen atoms, respectively. (f) Plan-view schematic diagram of the atomic arrangement in twisted bilayer 2H TMDs before reconstruction. Moiré superlattice exhibits continuously varying stacking orders with locally commensurate MX' , MM' and XX' stackings. (g) Plan-view schematic diagram of the atomic arrangement in twisted bilayer 2H TMDs after reconstruction. Energetically favorable MX' stacking expands to form hexagonal domain, while MM' and XX' stackings are localized at the vertices. Domain boundaries emerge between hexagonal domains.

evolution of reconstructed domain boundary networks from a large-scale dislocation perspective.

The tear-and-stack technique³⁸ was utilized to fabricate MTBM with the controlled twist angle. A half piece of a monolayer MoS_2 layer is stacked onto the other half piece with a small twist angle offset to the antiparallel alignment. The MTBM specimen is suspended on a microporous SiN membrane chip for TEM and STEM analysis (Figure 1b, see Supporting Information S1 for methods).

The microstructural properties of the MTBM were probed using DF TEM (Figure 1a). In this setup, electron beams are diffracted by the specimen and are separated by their in-plane momentum at the back focal plane, forming an electron diffraction pattern. DF images are subsequently obtained by selecting a specific Bragg peak with the objective aperture. Figure 1c shows the selected area electron diffraction (SAED) pattern obtained from the MTBM with the twist angle of 0.7° . Bragg peaks marked with colored circles in Figure 1c, corresponding to three distinct crystallographic planes in the $\{10\bar{1}0\}$ family, were selected to construct three DF images. These images were subsequently summed to obtain an integrated DF image (Figure 1d), which exhibits contrast sensitive to variation in the stacking configuration. Consequently, an array of hexagonal commensurate domains with MX' stacking orders is visualized with high intensity, while their boundaries are revealed as darker regions.

The tessellation of hexagonal domains in the MTBM arises from interfacial atomic reconstruction. The moiré superlattice formed near antiparallel alignment features three symmetric commensurate stacking orders, MX' , MM' , and XX' (Figure 1e), among which MX' represents the ground-state configuration.^{11,21,24} At sufficiently small twist angle, the moiré superlattice undergoes atomic reconstruction that expands the energetically favorable MX' stacking order to form hexagonal domains, while minimizing the unfavorable MM' and XX'

stacking orders, as schematically represented in Figures 1f,g.^{11,24–26} The resulting hexagonal domain array is distinct from triangular domain arrays observed in the marginally twisted bilayer TMDs near parallel alignment (3R-like) or marginally twisted bilayer graphene, where the energetic degeneracy between two ground-state configurations (MX and XM in TMDs, or AB and BA in graphene) leads to tessellation of the two degenerate triangular domains.

We further investigated the structure of domain boundaries in MTBM. From a crystallographic perspective, these domain boundaries correspond to dislocations, each defined by a characteristic Burgers vector that describes the relative translational shift between the top and bottom layers. Because a domain boundary involves a well-defined interlayer translational shift, its diffraction contrast in DF TEM is governed by the relative orientation between the diffraction vector $\mathbf{g}$ and the Burgers vector $\mathbf{b}$ of the boundary.

To exploit this relationship, we performed a systematic contrast analysis by imaging the same specimen area while varying only the diffraction vector $\mathbf{g}$. Specifically, DF images were acquired using first-order Bragg peaks (purple dashed circles in Figure 2a) and second-order Bragg peaks (orange dashed circles in Figure 2a), enabling direct comparison of the contrast of identical domain boundaries under different diffraction vectors.

The appearance of domain boundaries strongly depends on the selected diffraction vector (Figures 2b–g). In DF images formed using first-order Bragg peaks, the boundaries appear as single dark lines or become completely invisible for specific $\mathbf{g}$ vectors (white dashed boxes in Figures 2b–d and corresponding magnified views). In contrast, DF images obtained from second-order Bragg peaks reveal either single-line or double-line contrast, again depending on $\mathbf{g}$. This systematic contrast variation reflects the interplay between the interlayer translational shift across the domain boundary and the selected

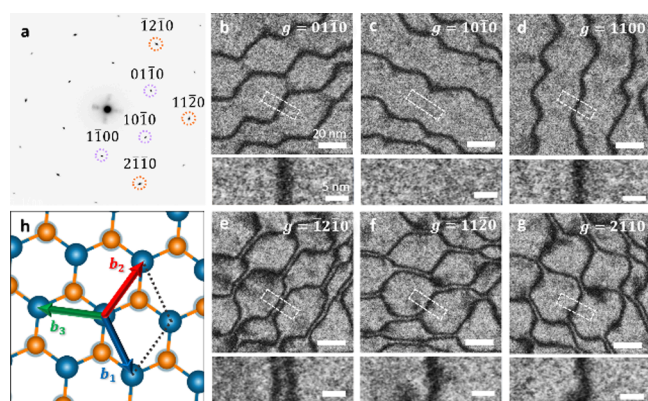


Figure 2. Domain boundary characterization. (a) SAED pattern of MTBM acquired along the [0001] zone axis. Three diffraction peaks of the {1010} family are indicated by purple, and three diffraction peaks of {1120} family are marked in orange. (b–d) Series of DF images acquired from the same region using the {1010} diffractions along the [0001] zone axis. The bottom panels show magnified views of the regions indicated by white dashed boxes in the top panels, where a domain boundary invisible at $g = 10\bar{1}0$ becomes visible at $g = 01\bar{1}0$ and $g = 1\bar{1}00$. (e–g) Series of DF images acquired from the same region using the {1120} diffractions along the [0001] zone axis. The bottom panels show magnified views, where a domain boundary exhibiting double-line contrast at $g = 1\bar{1}20$ appears as a single line at $g = 1\bar{1}20$ and $g = 2\bar{1}10$. (h) Schematic illustration of the Burgers vectors $\mathbf{b}_1 = \frac{1}{3}[2\bar{1}\bar{1}0]$, $\mathbf{b}_2 = \frac{1}{3}[\bar{1}2\bar{1}0]$ and $\mathbf{b}_3 = \frac{1}{3}[\bar{1}\bar{1}20]$ of MoS_2 .

diffraction vector, providing a robust experimental basis for identifying the dislocation character of the domain boundaries.^{32,34}

At the domain boundaries, relative translational shifts between the top and bottom layers can introduce a phase difference between scattered electron waves from the two layers. Within a kinematical diffraction framework, the resulting DF contrast is governed by how this phase difference modulates the coherent addition of diffracted waves.

To quantitatively examine this relationship and unambiguously determine the dislocation character of the domain boundaries, we analyzed the DF contrast using a kinematical diffraction theory (see Supporting Information S2 for a

detailed discussion of the applicability of the kinematical theory). Owing to the atomically thin nature of vdW bilayers, multiple scattering effects are strongly suppressed, making the kinematical approximation well suited for describing DF contrast in this system.

Within the kinematical diffraction approximation, the phase shift induced by an interlayer translational shift $\mathbf{b}$ across a domain boundary is proportional to the scalar product $\mathbf{g} \cdot \mathbf{b}$. Under first-order Bragg conditions, this relationship naturally categorizes domain boundaries into two distinct cases: boundaries for which $\mathbf{g} \cdot \mathbf{b} = 0$, resulting in zero phase shift and complete suppression of DF contrast, and boundaries for which $\mathbf{g} \cdot \mathbf{b} \neq 0$, where a finite phase shift gives rise to detectable DF contrast. Accordingly, the observed visibility or invisibility of domain boundaries in first-order DF images (Figures 2b–d) can be directly interpreted in terms of the orientation relationship between $\mathbf{g}$ and $\mathbf{b}$.

For $\text{MX}'\text{--MX}'$ domain boundaries in MTBM, two dislocation configurations are possible. A single perfect dislocation is characterized by a Burgers vector along the zigzag family directions, $\mathbf{b}_{\text{perfect}} = \frac{1}{3}\langle 2\bar{1}\bar{1}0 \rangle$, corresponding to a one-step translational shift between adjacent MX' domains. Alternatively, a pair of two closely spaced partial dislocations would involve two successive armchair-oriented Burgers vectors, $\mathbf{b}_{\text{partial}} = \frac{1}{3}\langle 1\bar{1}00 \rangle$, whose vector sum is equivalent to that of a perfect dislocation but follows a distinct two-step translation pathway. Although both configurations yield the same net MX' to MX' stacking transition, they involve fundamentally different atomic translation pathways and dislocation characters.

Crucially, the contrast behavior observed in first-order DF images provides a decisive experimental test for distinguishing between the two dislocation scenarios. Under specific first-order Bragg conditions, certain MTBM domain boundaries become completely invisible (Figure 2c), indicating that the Burgers vector of the boundary satisfies $\mathbf{g} \cdot \mathbf{b} = 0$.^{34,35,39,40} Such complete suppression of DF contrast can occur only for a single perfect dislocation with a zigzag-oriented Burgers vector, $\mathbf{b}_{\text{perfect}} = \frac{1}{3}\langle 2\bar{1}\bar{1}0 \rangle$. In contrast, paired partial dislocations,

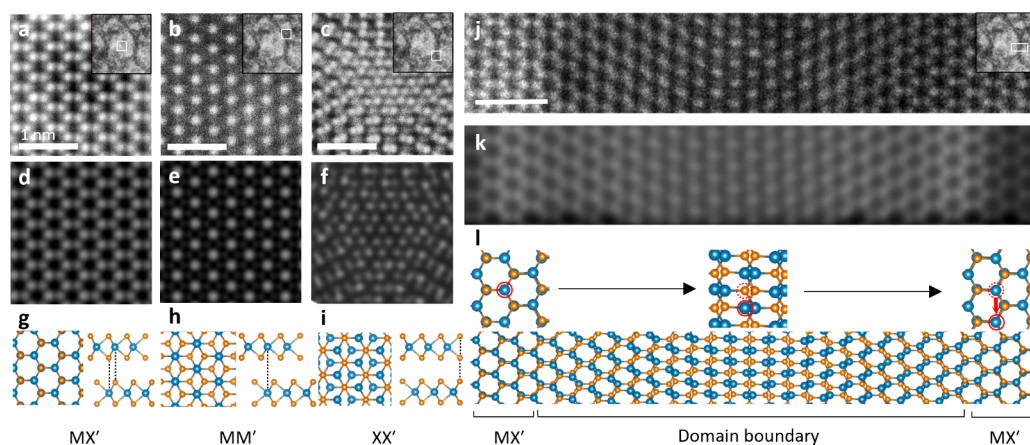


Figure 3. Atomic structures of domains and domain boundary in MTBM. (a–c) Atomic-resolution ADF STEM images of (a) MX' , (b) MM' , and (c) XX' domains. Insets show low-magnification images, with the probed locations marked by white boxes. (d–f) STEM simulations of (d) MX' , (e) MM' , and (f) XX' domains. (g–i) Corresponding atomic models of (g) MX' , (h) MM' , and (i) XX' domains viewed from the top (left) and side (right). (j) ADF STEM image of the domain boundary between MX' domains. (k) Simulated ADF STEM image and (l) atomic model of the domain boundary, exhibiting translation from MX' to the adjacent MX' domains, corresponding to a screw-type perfect dislocation.

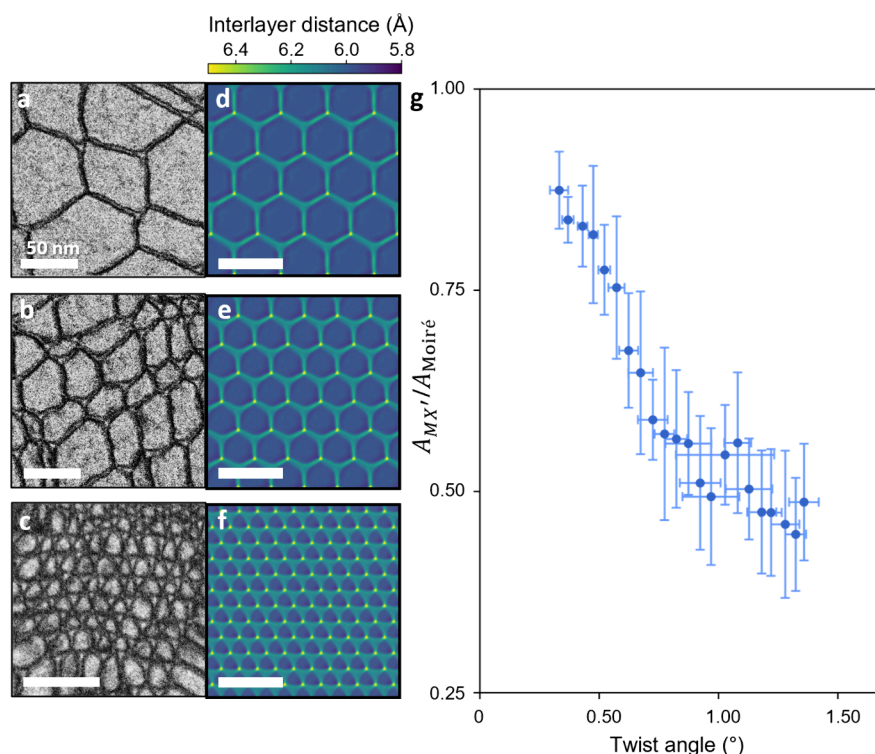


Figure 4. Twist-angle-dependent structural evolution. (a–c) Overlaid DF images obtained from three Bragg peaks of $\{11\bar{2}0\}$ family planes for regions with average twist angles of (a) 0.3°, (b) 0.7°, and (c) 1.3°. (d–f) Interlayer distance maps of fully relaxed 2H-like twisted bilayer MoS₂ obtained from atomistic simulations for twist angles of (d) 0.51°, (e) 0.71°, (f) 1.30°. MX' domains are identified as regions where the interlayer distance is below the cutoff value of 6.05 Å. As the twist angle increases from 0.3°, MX' domains with a hexagonal shape gradually decrease in size and evolve into a round triangular shape. (g) The fraction of the area of MX' domain ($A_{MX'}$) to the moiré half unit cell area ($A_{Moiré}$). Blue circles denote areal fraction averaged over 0.05° twist-angle bins. Horizontal and vertical error bars indicate the standard deviations of the twist angle and $A_{MX'}/A_{Moiré}$ within each bin, respectively (see Supporting Information S5 for details on the binning).

composed of two noncollinear armchair-oriented Burgers vectors, $\mathbf{b}_{\text{partial}} = \frac{1}{3}\langle 1\bar{1}00 \rangle$, would necessarily retain residual contrast under all first-order DF imaging conditions. The experimental observation of fully disappearing boundaries therefore unambiguously excludes paired partial dislocation configuration.

On this basis, we identify the domain boundaries separating adjacent MX' domains in MTBM as single perfect dislocations with a screw character. The three symmetry-equivalent Burgers vectors are determined to be $\mathbf{b}_1 = \frac{1}{3}[2\bar{1}\bar{1}0]$, $\mathbf{b}_2 = \frac{1}{3}[\bar{1}2\bar{1}0]$ and $\mathbf{b}_3 = \frac{1}{3}[\bar{1}\bar{1}20]$ (Figure 2h), corresponding to the three crystallographically equivalent zigzag directions of the hexagonal lattice.

Second-order DF images provide an independent validation of this assignment. Double-line contrast emerges under specific second-order Bragg conditions when $\mathbf{g}\cdot\mathbf{b} = \pm 2$ (Figure 2e; see Supporting Information S3), whereas single-line contrast is observed when $\mathbf{g}\cdot\mathbf{b} = \pm 1$ (Figures 2f, g).³⁵ Compared to the case of $\mathbf{g}\cdot\mathbf{b} = \pm 1$, the phase shift accumulated across the domain boundary is doubled for $\mathbf{g}\cdot\mathbf{b} = \pm 2$, leading to a characteristic splitting of the DF contrast into two parallel lines. Importantly, this contrast behavior is fully consistent with the same Burgers vector assignment and conclusively identifies the domain boundary network in MTBM as a network of screw-type perfect dislocations.

The resulting hexagonal network of screw-type perfect dislocations in MTBM is therefore fundamentally distinct from the triangular domain boundary networks observed in

marginally twisted bilayer TMDs near parallel alignment, which are composed of partial dislocations separating alternating stacking domains. This contrast demonstrates a key difference in both geometric symmetry and dislocation character between antiparallel- and parallel-aligned TMDs bilayers.

To further clarify the atomic structure of domains and their boundaries, we employed aberration-corrected annular dark-field scanning transmission electron microscopy (ADF STEM). The experimental images (Figures 3a–c, see Supporting Information S1 and S4 for details), together with corresponding ADF STEM simulations (Figures 3d–f) based on atomic models (Figures 3g–i), directly confirm the reconstructed stacking configurations inferred from DF TEM. In the hexagonal domain, the MX' stacking (where one transition metal atom aligns with two chalcogen atoms) occupies the largest area, confirming it as the energetic ground state (Figures 3a, d, g). In contrast, the MM' and XX' stacking configurations appear as localized vortices (Figures 3b, e, h and Figures 3c, f, i, respectively). Quantitative analysis shows that the metastable MM' domains have a characteristic size of approximately 7 nm, whereas the unstable XX' regions are highly confined, with sizes of about 1 nm.

Atomic-resolution imaging of the boundary between adjacent MX' domains (Figure 3j) shows a continuously varying atomic configuration, in good agreement with the simulated image (Figure 3k) derived from an atomic model of a single perfect dislocation (Figure 3l). Across this boundary, atoms undergo a relative translational displacement along the

zigzag direction, forming a screw-type perfect dislocation. This direct real-space observation is consistent with our kinematical analysis of the DF TEM contrast, as well as with previous theoretical predictions^{26,41} and experimental observations.²⁴

The quantitative agreement between wide-field DF TEM analysis and atomic-resolution ADF STEM validation establishes kinematical analysis of DF TEM as a robust and unambiguous approach for identifying the dislocation character of domain boundaries over large length scales. Unlike atomic-resolution probes, which are inherently limited in field of view, this DF-based methodology enables systematic mapping of dislocation networks while retaining direct sensitivity to the underlying Burgers vectors. This validated capability provides a necessary foundation for examining how reconstructed domain boundary networks evolve with twist angle, a question that has remained largely inaccessible using conventional techniques.

Building on this validated large-scale dislocation analysis method, we investigate the twist-angle-dependent evolution of reconstructed domain boundary networks, in conjunction with atomistic simulations (see [Supporting Information S1](#) for details). With increasing twist angle, the MX' domains progressively shrink, while MM' regions occupy a larger fraction of the moiré unit cell, gradually disrupting the hexagonal tessellation ([Figure 4a-f](#)).

To quantify the evolution of domain morphology within the reconstructed regime, we measured the areal fraction of MX' domains, $A_{MX'}/A_{Moiré}$, as a function of twist angle ([Figure 4g](#)). Notably, the MX' areal fraction decreases approximately linearly with increasing twist angle up to $\sim 1.5^\circ$, a range in which the atomic reconstruction strength has been reported to remain robust in previous study.⁴² This linear trend can be qualitatively understood from the geometric scaling between the moiré supercell area and the domain-wall area (see [Supporting Information S6](#) for details).

Based on this linear behavior, we define the transition angle, θ_b , as the twist angle at which the MX' stacking occupies half of the moiré supercell ($A_{MX'}/A_{Moiré} = 0.5$). This characteristic angle corresponds to the point at which the dislocation network undergoes a gradual structural transition from a hexagonal network of screw-type perfect dislocations separating adjacent MX' domains to a triangular network of partial dislocations separating MX' and MM' domains. From linear fitting of the areal-fraction data extracted from the experimental measurements, θ_t is determined to be approximately 1.1° .

Notably, this transition reflects a reorganization of the dislocation-network topology within the reconstructed regime, rather than the disappearance of atomic reconstruction itself. As the twist angle approaches θ_b , the balance between interlayer interaction and intralayer lattice distortion energies causes the domain sizes of the ground-state MX' stacking and metastable MM' stacking to become increasingly comparable, leading to a reconfiguration of the dislocation network while preserving distinct stacking domains. At larger twist angles, where the interfacial reconstruction amplitude becomes insufficient to stabilize well-defined commensurate domains, the system gradually crosses over into a weakly reconstructed regime, consistent with previous reports.^{17,42–44}

Such a twist-angle-driven dislocation-network transition is a distinctive feature of 2H-like TMDs bilayers; in contrast, in 3R-like TMDs, comparable transitions between two distinct dislocation networks are typically induced by external electric fields rather than by twist-angle variation.^{22,23}

We expect that the twist-angle-driven transition characterized by θ_t is a general feature of 2H-like TMDs moiré superlattices. This behavior fundamentally originates from the energetic hierarchy among the three local stacking configurations (MX', XX', and MM'), where the MX' stacking is energetically favored over the other two. As a result, an increase in the energetic preference for MX' stacking shifts θ_t to larger twist angles, whereas an increase in layer stiffness reduces θ_t due to the increased in-plane strain energy cost.

In summary, by combining DF TEM with kinematical diffraction analysis and atomic-resolution ADF STEM, we establish a validated, multiscale framework for resolving dislocation networks in marginally twisted bilayer TMDs. Our method reveals that small twist angles stabilize a hexagonal network of screw-type perfect dislocations, while increasing the twist angle drives a transition to a triangular network of screw-type partial dislocations through a reorganization of the domain boundary structure. The ability to directly link twist-controlled dislocation network topology to reconstructed domain morphology enables systematic exploration of structure–property relationships in twisted vdW bilayers, including piezoelectric charge accumulation,^{24,28} enhanced tunneling conductivity,²⁵ and quantum-dot-like electronic states at domain boundary junctions,^{15,31} as well as more complex twisted multilayer systems.

■ ASSOCIATED CONTENT

Data Availability Statement

The DF TEM simulation codes used in this study are publicly available at: <https://github.com/HyunPhys/DislocationNetworksInMarginallyTwistedBilayerMoS2>. The repository contains atomic structure files, multislice simulations, kinematical simulations, and domain wall contrast models used to generate the results presented in this work. The atomistic calculation codes used in this study are publicly available at: <https://github.com/sogang-qmp/TwistedMoS2Relaxation>. The repository contains force constants of MoS₂ monolayer obtained from density functional perturbation theory, structural relaxation codes, and interlayer distance map plotting script used to generate the results presented in this work.

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsmaterialslett.5c01692>.

Sample preparation; DF TEM and ADF STEM imaging; Kinematical description of DF contrast at domain boundaries; identification of domain boundary character from DF contrast analysis; ADF STEM image acquisition and processing procedures ([PDF](#))

■ AUTHOR INFORMATION

Corresponding Authors

Young Woo Choi – Department of Physics and Center for Nano Materials, Sogang University, Seoul 04107, Korea; orcid.org/0000-0003-4725-1299; Email: ywchoi02@sogang.ac.kr

Miyoung Kim – Department of Materials Science and Engineering and Research Institute of Advanced Materials, Seoul National University, Seoul 08826, Korea; orcid.org/0000-0001-8632-6711; Email: mkim@snu.ac.kr

Moon Sung Kang – Department of Chemical and Biological Engineering and Institute of Emergent Materials, Sogang University, Seoul 04107, Korea; orcid.org/0000-0003-0491-5032; Email: kangms@sogang.ac.kr

Hyobin Yoo – Department of Materials Science and Engineering and Research Institute of Advanced Materials, Seoul National University, Seoul 08826, Korea; orcid.org/0000-0002-8039-9482; Email: hyobinyoo@snu.ac.kr

Authors

Byunghyun Kim – Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea

Ayoung Yuk – Department of Physics and Department of Chemical and Biological Engineering, Sogang University, Seoul 04107, Korea

Yunyeong Chang – Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea

Dongsin Kim – Department of Physics and Center for Nano Materials, Sogang University, Seoul 04107, Korea;

orcid.org/0009-0002-1838-2142

Daesung Park – Department of Materials Science and Engineering, Seoul National University, Seoul 08826, Korea; Department of Physics, Sogang University, Seoul 04107, Korea

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acsmaterialslett.5c01692>

Author Contributions

[†]B.K., A.Y., Y.C., and D.K. contributed equally to this work. H.Y. conceived the experiments. B.K., A.Y., and D.P. fabricated the samples. B.K., A.Y., and Y.C. performed the TEM experiments. D.K. and Y.W.C. performed theoretical calculations. B.K., A.Y., Y.C., D.P., M.K., M.S.K., and H.Y. analyzed the data. B.K., A.Y., Y.C., D.K., Y.W.C., M.K., M.S.K., and H.Y. wrote the manuscript with the inputs from all authors. All authors contributed to the overall scientific interpretation and edited the manuscript. CRediT: **Byunghyun Kim** data curation, formal analysis, investigation, methodology, writing - original draft, writing - review & editing; **Ayoung Yuk** data curation, formal analysis, investigation, methodology, writing - original draft, writing - review & editing; **Yunyeong Chang** data curation, formal analysis, investigation, methodology, writing - original draft, writing - review & editing; **Dongsin Kim** formal analysis, investigation, methodology, software, writing - review & editing; **Daesung Park** data curation; **Young Woo Choi** formal analysis, investigation, methodology, software, writing - original draft, writing - review & editing; **Miyoung Kim** formal analysis, investigation, writing - original draft, writing - review & editing; **Moon Sung Kang** formal analysis, investigation, writing - original draft, writing - review & editing; **Hyobin Yoo** conceptualization, funding acquisition, investigation, project administration, writing - original draft, writing - review & editing.

Notes

The authors declare no competing financial interest.

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ABBREVIATIONS

2D, two-dimensional; vdW, van der Waals; TMDs, transition metal dichalcogenides; STEM, resolution scanning transmission electron microscopy; STM, scanning tunneling microscopy; DF, dark field; TEM, transmission electron microscopy; ADF STEM, annular dark field scanning transmission electron microscopy; MTBM, marginally twisted bilayer MoS₂; SAED, selected area electron diffraction.

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