Moiré induced topology and flat bands in twisted bilayer WSe\(_2\): A first-principles study

Sudipta Kundu, 1 Mit H. Naik, 1,* H. R. Krishnamurthy, 1 and Manish Jain 1

1 Center for Condensed Matter Theory, Department of Physics, Indian Institute of Science, Bangalore 560012, India

We study the influence of strong spin-orbit interaction on the formation of flat bands in relaxed twisted bilayer WSe\(_2\). Flat bands, well separated in energy, emerge at the band edges for twist angles (\(\theta\)) near 0° and 60°. For \(\theta\) near 0°, the interlayer hybridization together with a moiré potential determines the electronic structure. The bands near the valence band edge have nontrivial topology, with Chern numbers equal to +1 or −1. We propose that the nontrivial topology of the first band can be probed experimentally for twist angles less than a critical angle of 3.5°. For \(\theta\) near 60°, the flattening of the bands arising from the K point of the unit cell Brillouin zone is a result of atomic rearrangements in the individual layers. Our findings on the flat bands and the localization of their wavefunctions for both ranges of \(\theta\) match well with recent experimental observations.

Twisted bilayer transition metal dichalcogenides (TMD) have recently gained attention as potential platforms for hosting correlated phases and novel excitonic properties [1–21]. The absence of a ‘magic angle’ in twisted TMDs [22–26] makes the experimental realization of flat bands [1, 2] easier compared to twisted bilayer graphene. Large spin-orbit coupling can strongly influence the electronic structure of the moiré superlattice (MSL), and cause the bands to have nontrivial topological character. Within the TMD family, WSe\(_2\) is a prototypical example with large spin-orbit coupling, making twisted WSe\(_2\) bilayers (tWSe\(_2\)) especially interesting.

Recent spectroscopic imaging and transport measurements in tWSe\(_2\) show evidence of flat bands [1, 2, 27]. As we discuss in detail in this letter, the flat bands at the valence band (VB) edge in the MSL of tWSe\(_2\) arise from the bands near the K points of the unit cell Brillouin zone (UBZ), unlike in other TMDs like MoS\(_2\). The origin and electronic structure of the flat bands arising from the K point are different from those due to the Γ point; the latter are primarily determined by the interlayer hybridization, whereas the former are a result of the “moiré potential”, the additional effective potential generated due to the relaxation of the atoms upon twisting. The spin character of the flat bands is determined by the spin-valley locking in WSe\(_2\). Due to the large spin-orbit splitting and the spin-valley locking at K/K’-valley, tWSe\(_2\) is a good platform for exploring opportunities for spintronics as well as valleytronics.

Most theoretical studies on tWSe\(_2\) to date [28–33] have been based on continuum models [34, 35]. In this letter, we study flat bands in relaxed MSL of tWSe\(_2\) using density functional theory (DFT). We focus our study on twist angles \(\theta\) near 0°, ranging from 7.3° to 1.89°, and near 60°, ranging from 52.7° to 58.11°. We find several well-separated flat bands near the band edges in tWSe\(_2\) for both ranges of \(\theta\). The spin-orbit splitting of the monolayer bands is preserved in tWSe\(_2\). Both the moiré potential and the interlayer hybridization govern the electronic structure of tWSe\(_2\) with \(\theta\) near 0°; in contrast, the flat bands in tWSe\(_2\) with \(\theta\) near 60° primarily result from the moiré potential alone. Flat bands near the VB edge for \(\theta\) near 0° are topologically nontrivial [34]. This stems from the nonzero Berry curvature at the K-valleys of the UBZ. We fit the bands to the continuum model [34]; the obtained parameters thus include the effect of structural relaxation of the MSL. We also find flat bands arising from the Γ point of the UBZ inside the VB continuum for both ranges of \(\theta\). The spatial localization of these flat bands is in excellent agreement with recent scanning tunnelling measurements (STM) [1].

We note that larger deviations of \(\theta\) from 0° or 60° lead to smaller moiré supercells. In such systems, the effect of kinetic energy is more prominent compared to the moiré effect, and the bands in the moiré Brillouin zone (MBZ) are primarily a result of the band folding. Conversely, the closer \(\theta\) is to 0° or 60°, the larger the moiré supercell, and the flatter the bands, increasing the possibilities for interesting strong correlation phenomena, with scope for additional richness in the presence of bands with nontrivial topology. Hence, we focus in this letter on tWSe\(_2\) with \(\theta\) near 0° and 60°. It is to be noted that 30° tWSe\(_2\) can give rise to quasicrystalline order [36, 37], and we hope to study such systems in the future.

Our electronic structure calculations were performed using the atomic-orbital basis as implemented in the SIESTA package [38]. The commensurate tWSe\(_2\) structures are generated using the Twister code [22] and relaxed using classical force fields as implemented in LAMMPS [39–42] (for other details, see Supplemental material (SM) [43]). The relaxation patterns agree well with previous studies on other TMDs [23, 44–46]. MSLs with \(\theta\) near 0° consist of regions with three high symmetry stackings: AA, AB, and BA (also known as 3R, B\(\!\!\!\!\!\!\!\!\!\!\!/\)Se, and B\(\!\!\!\!\!\!\!\!\!\!\!/\)W respectively), as well as bridge (Br) regions (the transition regions from one such stacking to another) (see SM [43]). MSLs with \(\theta\) near 60° consist of three other high symmetry stackings: AA′, A′B and AB′.
Electronic structure of tWSe$_2$ with $\theta$ near 0°

Fig. 1 presents our results for the band structures of tWSe$_2$ for $\theta$ near 0°. For such $\theta$ values, bands near the K$_M$ point of the moiré MBZ (subscript M denotes k-points in the MBZ) arise from the monolayer bands near the K and K$'$ points of the UBZ of the two layers. The zone folding relation is illustrated in Fig. 2(a). As the monolayer VB maximum at the K and K$'$ points have antiparallel spins, the VB maximum at the K$_M$ point is doubly degenerate, with antiparallel spins. Fig. 1(a) shows the bands for the 2.28° tWSe$_2$. Two-fold degenerate bands with opposite spins are evident near the VB edge (V1, V2, V3). These first few VBs are shown along the path $\text{K}_M - \text{M}_M - \text{K}'_M$ in Fig. 1(b). There is always a gap between V1 and V2, even at the M$_M$ point, due to the interlayer hybridization (Fig. 1(b)); this is clear from the fact that the bands for the relaxed-MSL monolayer (i.e., without interlayer interaction but with the structural relaxation effects of the MSL included) along the same path (pink dashed line in Fig. 1(b)) show a band crossing at M$_M$.

To study the topological aspects of the band structure, we have calculated the Chern number ($C_n$) for the first few bands near the VB edge. As the bands are doubly degenerate with antiparallel spin, we differentiate between the flat bands by following the spin. For this purpose we calculate the expectation value of the Pauli matrices for the first few bands at the VB edge and find that $\langle \sigma_z \rangle \sim \pm 1$, and $\langle \sigma_y \rangle$ is nearly zero. We consider the band with $\langle \sigma_z \rangle = +1 (-1)$ as the spin-up (spin-down) band. We generate the Bloch wavefunctions using DFT on a regular k-grid in the MBZ and compute the $C_n$'s [43, 60, 61]. The larger $\theta$ is, the denser is the k-grid we have used. For $\theta = 1.89°$, we compute the $C_n$ using a 6×6 k-grid in the MBZ and the $C_n$ are +1, +1, +1 respectively, for the V1, V2 and V3 spin-up bands. For 2.28° $\geq \theta > 4.4°$, $C_n$ are +1, +1, −1 for V1, V2 and V3 respectively. For $\theta \geq 4.4°$ we report $C_n$ for the first two isolated bands (+1 for both V1 and V2) only. The spin-down bands have $C_n$’s with opposite signs. It is important to note that the first band is topologically trivial when calculated for unrelaxed tWSe$_2$, and the 2nd band overlaps with other bands. The $\theta$ variation of the bandwidth of V1 (E$_W$) and the minimum energy gap between V1 and V2 (E$_{12}$) are shown in Fig. 1(c). While E$_W$ increases monotonically with $\theta$, E$_{12}$ becomes negative at 3.5°. While nonzero $C_n$ is found for a large range of $\theta$, the negative E$_{12}$ limits the range of angles for observing quantum spin-Hall insulating states of the first band in tWSe$_2$ [62, 63]. Furthermore, the quantum spin-Hall state can be probed for the first two bands till 4.4°. We have compared our DFT results to the continuum model [34]. While the latter gives the Chern number of the first two bands consistent with DFT for all $\theta$, it is unable to describe the DFT-calculated $C_n$ of the third band [43].

Fig. 1(a) also shows six split-off bands (C1) near the conduction band (CB) edge. These bands are degenerate...
at $\Gamma_M$ with three having spin-up and other three spin-down. The Q valley of the UBZ gives rise to these bands near the CB minima in the MBZ. The bands near the CB minima at alternate Q points of the UBZ have antiparallel spins. This determines the spin-character of C1 as well as of the next set of bands near the CB minimum in the MBZ, which are also 6-fold degenerate at $\Gamma_M$.

In order to clarify the nature of the spin-orbit splitting in the MSL, we have calculated the matrix elements of the spin-raising/lowering operator $S^{\pm} (= S_x \pm i S_y)$: $S_{ij} = \langle \psi^I_{K_M} | S^{\pm} | \psi^J_{K_M} \rangle$ where $\psi^I_{K_M}$, $\psi^J_{K_M}$ are two wavefunctions at the $K_M$ point of MBZ. We calculate the $S_{ij}$ for $i$ corresponding to the two highest energy states of VB edge (at the $K_M$ point) and $j$ to the states within the energy range 0–500 meV below. We find states 436 meV below the VB maximum for which $S_{ij}$ is large (Fig. 1(d)). This implies that the atomic spin-orbit splitting remains essentially unchanged in MSLs.

Wavefunction localization: Next, we discuss the localization of the wavefunctions of the bands mentioned above, using false color plots of the probability $|\psi_k(r)|^2$ inside the MSL, for special choices of $k$. (The localization characteristics vary with the choice of $k$.) Figs. 3(a) and (b) depict the localization of the wavefunctions corresponding to V1 and V2 respectively at the $\Gamma_M$ point; the V1 wavefunction shows a hexagonal pattern avoiding AA stacking regions and occupying the AB and BA regions, while the V2 wavefunction is localized in the AA regions. Figs. 3(d) and (e) show the localization of V1 and V2 at $K_M$ for one spin orientation (say spin-down); V1 localizes on AA and AB regions, while V2 does so on BA regions. At $K_M$, the localization of the other spin (spin-up, not shown) is complementary; V1 localizes on AA and BA regions and V2 on AB regions. Figs. 3(c) and (f) depict the localization of the C1 wavefunctions at $\Gamma_M$ and $K_M$ respectively. In both cases C1 localizes on AB and BA regions as the CB minimum at the Q valley in the UBZ has the lowest energy for AB and BA stackings.

A complimentary picture of the localization is provided by the coarse-grained “moiré potential” ($V_M$) [23] of a relaxed MSL monolayer. The coarse-grained potential is obtained by averaging the potential in a Voronoi cell centered at the W atoms ($x_W, y_W$) in each layer:

$$V_M(x_W, y_W) = \frac{\int_{\Omega_{VW}} V(x, y, z) d\mathbf{r}}{\Omega_{VW}} - \bar{V}. \tag{1}$$

Here $V(x, y, z)$ is the local potential obtained from the DFT calculation and $\bar{V}$ is its mean. Fig. 3(g) depicts the moiré potential of one of the layers. The variation of the potential is driven by the strain and relaxation patterns. The resulting $V_M$ shows an alternate arrangement of maxima and minima in a hexagonal shape centred on the AA regions. The other layer has a similar pattern with the positions of maxima and minima interchanged. The moiré potential along with the interlayer hybridization determines the electronic structure of the relaxed MSL of tWSFe2 for $\theta$ near 0°.

Comparison with experiment: STM images of 3° tWSFe2 [1] showed signatures of flat bands derived from the $\Gamma$ point of the UBZ and the local density of states (LDOS) corresponding to these bands . We find flat bands arising from the $\Gamma$ point of UBS inside the VB continuum [43]. The LDOS associated with these $\Gamma$-derived states localizes strongly on AB, BA and Br regions, forming a hexagonal pattern (Fig. 3(h)). This is in excellent agreement with the STM image of the $\Gamma$-derived flat band [1]. In contrast, the LDOS of V1, which arises due to the K point of the UBZ, and is shown in Fig. 3(i), is delocalized in the moiré unit cell, also consistent with the experimental findings [1].

Electronic structure of tWSFe2 with $\theta$ near 60°: We next discuss our findings for the electronic structure and the wavefunctions of the relaxed MSLs obtained for $\theta$ near 60°.

Fig. 4(a) depicts the band structure for the 57.72° MSL, with the bands near the VB and CB edges being shown more clearly in Figs. 4(b) and 4(c). Several well-separated flat bands emerge at the VB edge (Fig. 4(b)). These bands are four-fold degenerate at the $\Gamma_M$ point of the MBZ, and split into two sets of two-fold degenerate bands at the $K_M$ point, with the spins of each set being parallel (inset of Fig. 4(a)) but antiparallel to the spins.
of the other set. This is because the first set of bands at the $K_M$ point of the MBZ arise from the VB maximum at the $K$ points of the UBZ of the two layers, which have the same spin. The second set of bands with the opposite spin at the $K_M$ point arises from the VBs at the $K'$ (modulo a reciprocal lattice vector of the MSL) point of the UBZ of the two layers. This folding relation is shown in Fig. 2(b). The bands at the VB edge for $\theta$ near $60^\circ$ are topologically trivial. The flat bands at CB edge are nearly 12-fold degenerate (Fig. 4(c)) (two sets of 6-fold degenerate bands with $\sim 4\text{meV}$ separation), and originate from the doubly-degenerate bands with antiparallel spins at the Q valley of UBZ.

To further clarify the spin-character of the bands, the bands $V_1$ and $V_2$ near the VB edge are shown along the path $K_M - M_M - K'_M$ in the MBZ in the inset in Fig. 4(a). As can be seen, the two sets comprising the $V_1$ bands cross each other at the $M_M$ point of the MBZ and have opposite ordering at the $K'_M$ point. Fig. 4(d) shows the variation with $\theta$ of the energy gap ($E_W$) at the $K_M$ point between these two sets of bands (brown line), and of the minimum energy gap between $V_1$ and $V_2$ ($E_{12}$) (purple line). As can be seen from the figure, $E_{12}$ becomes negative for $\theta \geq 54.9^\circ$, and $E_W$, which also denotes the bandwidth of $V_1$ as both the maximum and minimum value of $V_1$ are at $K_M$, decreases with increasing MSL size. It is worth emphasizing that $E_W$ is not a spin-orbit splitting but arises due to band folding. This becomes evident if we look for the atomic spin-orbit partner of the $V_1$ state at the $K_M$ point by calculating the matrix elements $S_{ij}$ as discussed before. We find large $S_{ij}$ for states $\sim 440\text{meV}$ below the corresponding states at the VB edge (Fig. 4(e)).
with respect to the dispersion of the bands, as well as
the separation between them. This implies that the flat
bands near the VB edge originate primarily from the in-
plane strains and not from the interlayer hybridization.
The first three wavefunctions at the VB edge of the re-
axed MSL monolayer are depicted in Figs. 5(d)–(f) and
they look the same as those of the MSL bilayer. On
the other hand, the flat bands at the CB edge are derived
from the Q valley of the UBZ. Although the interlayer
hybridization is nonzero at the Q valley, the bands near
the CB edge of bilayer MSL and of the relaxed MSL
monolayer also agree very well. Furthermore, the CB
edge wavefunction at the $\Gamma_M$ point of the relaxed monolayer localizes at the same stacking regions as that of the full MSL.

In order to further understand the wavefunction local-
ization, we have computed the moiré potential ($V_M$) of
the relaxed MSL monolayer (Fig. 5(h)). $V_M$ has max-
ima at the AA' and $\Lambda'B$ regions and minima at the AB' regions, which is consistent with the holes localizing at AA' and $\Lambda'B$ and the electrons localizing at AB'. Hence, in the case of tWSe$_2$ with $\theta$ close to 60°, a relaxed MSL monolayer is sufficient to account for the electronic properties of the MSL if one is interested in the first few bands at the band edges.

**Flat bands arising from the $\Gamma$ point of the UBZ:** The monolayer band states near the $\Gamma$ point of the UBZ also give rise to flat bands in the MBZ. The flat band at 129 meV below the VB maximum (marked as $V_\Gamma$ in Fig. 4(b)) is the topmost such band. This band has a bandwidth < 1 meV, and its wavefunction localizes strongly on the AA' regions, similar to the findings in twisted MoS$_2$ [22, 23]. The localization is consistent with the ordering of the VBs at the $\Gamma$ point in the UBZ of the different high symmetry stacking bilayers [43]. This $V_\Gamma$ band is two-fold degenerate and has charge density in the interlayer region, unlike the states arising from the K point of the UBZ. The LDOS corresponding to the state at the $\Gamma_M$ point of this band is shown in Fig. 5(i). STM studies on 57.5° tWSe$_2$ find $\Gamma$-derived bands localized on AA' stacking [1] regions. Our findings on the nature and localization of the $\Gamma$-derived flat band are thus in excellent agreement with the STM results.

**Concluding comments:** In this letter we have presented an extensive study of the formation of flat bands in tWSe$_2$ for $\theta$ near 0° as well as 60°, including the localization characteristics of the wavefunctions of these bands, using DFT. Our study includes the strong spin-orbit coupling present in this system, as well as the atomic re-arrangements arising from the relaxation of the rigidly rotated bilayer system. We find topologically nontrivial bands for tWSe$_2$ near 0° where interlayer hybridization together with the moiré potential determines the electronic structure. For $\theta$ near 60°, the moiré potential of the relaxed MSL monolayer alone can account for the electronic structure and the localization characteristics of the first few flat bands at the band edges. Furthermore, we have identified another set of flat bands arising from the $\Gamma$ point of the UBZ. Our findings are in excellent agreement with recent STM experiments. We believe that our study provides generic insights into the nature of flat bands in twisted TMD bilayers with strong spin-orbit interactions. The K-derived flat bands at the VB edge should prove especially valuable for investigating the spin-valley physics in twisted homobilayer TMD systems.

We thank the Supercomputer Education and Research Centre (SERC) at IISc for providing the computational resources. S. K. and M. J. acknowledge discussions with Johannes Lischner and Valerio Vitale. M. J. gratefully acknowledges financial support through grant no. DST/NSM/R&D_HPC_Applications/2021/23 from the National Supercomputing Mission of the Department of Science and Technology, India. H. R. K. gratefully acknowledges the Science and Engineering Research Board of the Department of Science and Technology, India for financial support under grant No. SB/DF/005/2017.

* Present address: Department of Physics, University of California at Berkeley, California 94720, USA and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA.
[1] Z. Zhang, Y. Wang, K. Watanabe, T. Taniguchi, K. Ueno, E. Tutuc, and B. J. LeRoy, Nature Physics 16, 1093 (2020).
[2] L. Wang, E.-M. Shih, A. Ghiotto, L. Xian, D. A. Rhodes, C. Tan, M. Claassen, D. M. Kennes, Y. Bai, B. Kim, K. Watanabe, T. Taniguchi, X. Zhu, J. Hone, A. Rubio, A. N. Pasupathy, and C. R. Dean, Nature Materials 19, 861 (2020).
[3] G. Scuri, T. I. Andersen, Y. Zhou, D. S. Wild, J. Sung, R. J. Gelly, D. Bérubé, H. Heo, L. Shao, A. Y. Joe, A. M. Mier Valdivia, T. Taniguchi, K. Watanabe, M. Lončar, P. Kim, M. D. Lukin, and H. Park, Phys. Rev. Lett. 124, 217403 (2020).
[4] S. Brem, K.-Q. Lin, R. Gillen, J. M. Bauer, J. Maultzsch, J. M. Lupton, and E. Malic, Nanoscale 12, 11088 (2020).
[5] T. I. Andersen, G. Scuri, A. Sushko, K. D. Greve, J. Sung, Y. Zhou, D. S. Wild, R. J. Gelly, H. Heo, K. Watanabe, T. Taniguchi, P. Kim, H. Park, and M. D. Lukin, “Moiré excitons correlated with superlattice structure in twisted wse2/wse2 homobilayers,” (2019), arXiv:1912.06955 [cond-mat.mes-hall].
[6] S. Brem, K.-Q. Lin, R. Gillen, J. M. Bauer, J. Maultzsch, J. M. Lupton, and E. Malic, Nanoscale 12, 11088 (2020).
[7] A. M. van der Zande, J. Kunstmann, A. Chernikov, D. A. Chenet, Y. You, X. Zhang, P. Y. Huang, T. C. Berkelbach, L. Wang, F. Zhang, M. S. Hybertsen, D. A. Muller, D. R. Reichman, T. F. Heinz, and J. C. Hone, Nano Letters 14, 3869 (2014).
[8] S. Carr, D. Massatt, S. Fang, P. Cazeaux, M. Luskin, and E. Kaxiras, Phys. Rev. B 95, 075420 (2017).
[9] M.-L. Lin, Q.-H. Tan, J.-B. Wu, X.-S. Chen, J.-H. Wang,
Y.-H. Pan, X. Zhang, X. Cong, J. Zhang, W. Ji, P.-A. Hu, K.-H. Liu, and P.-H. Tan, ACS Nano 12, 8770 (2018).
[10] J. Kang, J. Li, S.-S. Li, J.-B. Xia, and L.-W. Wang, Nano Letters 13, 5485 (2013).
[11] I. Maity, M. H. Naik, P. K. Maiti, H. R. Krishnamurthy, and M. Jain, Phys. Rev. Research 2, 013335 (2020).
[12] R. Deb Nath, I. Maity, R. Biswas, V. Raghunathan, M. Jain, and A. Ghosh, Nanoscale (2020).
[13] D. Sorian and J. L. Lado, Journal of Physics D: Applied Physics 53, 474001 (2020).
[14] M. Forg, A. S. Baimuratov, S. Y. Kruchinin, I. A. Vovk, J. Scherzer, J. Förste, V. Funk, K. Watanabe, T. Taniguchi, and A. Hangle, “Moiré excitons in mose2-wse2 heterobilayers and heterotrilayers,” (2020), arXiv:2006.09105 [cond-mat.mes-hall].
[15] Z. Li, X. Lu, D. F. C. Leon, J. Hou, Y. Lu, A. Kaczmarek, Z. Lyu, T. Taniguchi, K. Watanabe, L. Zhao, L. Yang, and P. B. Deotare, “Exciton transport under periodic potential in mose2/wse2 heterostructures,” (2020), arXiv:2002.01501 [cond-mat.mes-hall].
[16] J. Wang, Q. Shi, E.-M. Shih, L. Zhou, W. Wu, Y. Bai, D. A. Rhodes, K. Barmak, J. C. Hone, C. R. Dean, and X.-Y. Zhu, “Excitonic phase transitions in mose2/wse2 heterobilayers,” (2020), arXiv:2001.03812 [cond-mat.mes-hall].
[17] X. Lu, X. Li, and L. Yang, Phys. Rev. B 100, 155416 (2019).
[18] A. Y. Joe, L. A. Jäuregui, K. Pustanova, Z. Lu, D. S. Wild, G. Scur, K. D. Greve, R. J. Gelly, Y. Zhou, J. Sung, A. M. M. Valdivia, A. Sushko, T. Taniguchi, K. Watanabe, D. Smirnov, M. D. Lukin, H. Park, and P. Kim, “Electrically controlled emission from triplet charged excitons in atomically thin heterostructures,” (2019), arXiv:1912.07678 [cond-mat.mes-hall].
[19] L. le Zhang, Z. Zhang, F. Wu, D. Wang, R. Gogna, S. Hou, K. Watanabe, T. Taniguchi, K. Kulkarni, T. S. Kuo, S. R. Forrest, and H. Deng, “Moiré lattice-induced formation and tuning of hybrid dipolar excitons in twisted ws2/mose2 heterobilayers,” (2019), arXiv:1911.10069 [cond-mat.mes-hall].
[20] H. Li, S. Li, M. H. Naik, J. Xie, X. Li, J. Wang, E. Regan, D. Wang, W. Zhao, S. Zhao, S. Kahn, K. Yunigeta, M. Blei, T. Taniguchi, K. Watanabe, S. Tongay, A. Zettl, S. G. Louie, F. Wang, and M. F. Crommie, Nature Materials (2021).
[21] Y. Luo, R. Engelke, M. Mattheakis, M. Tamagnone, S. Carr, K. Watanabe, T. Taniguchi, E. Kaxiras, P. Kim, and W. L. Wilson, Nature Communications 11, 4200 (2020).
[22] M. H. Naik and M. Jain, Phys. Rev. Lett. 121, 266401 (2018).
[23] M. H. Naik, S. Kundu, I. Maity, and M. Jain, Phys. Rev. B 102, 075413 (2020).
[24] M. Angeli and A. H. MacDonald, “γ-valley transition-metal-dichalcogenide moiré bands,” (2020), arXiv:2008.01735 [cond-mat.str-el].
[25] L. Xian, M. Claassen, D. Kiese, M. M. Scherer, S. Trebst, D. M. Kennes, and A. Rubio, “Realization of nearly dispersionless bands with strong orbital anisotropy from destructive interference in twisted bilayer mos2,” (2020), arXiv:2004.02964 [cond-mat.mes-hall].
[26] Y. Zhang, T. Liu, and L. Fu, Phys. Rev. B 103, 155414 (2021).
[27] M. Huang, Z. Wu, J. Hu, X. Cai, E. Li, L. An, X. Feng, Z. qing Ye, N. Lin, K. T. Law, and N. Wang, “Giant nonlinear hall effect in twisted wse2,” (2020), arXiv:2006.05615 [cond-mat.mes-hall].
[28] Z. Bi and L. Fu, “Excitonic density wave and spin-valley superfluid in bilayer transition metal dichalcogenide,” (2019), arXiv:1911.04493 [cond-mat.str-el].
[29] J. Hu, C. Zhang, Y. Xie, and K. T. Law, “Nonlinear hall effects in strained twisted bilayer wse2,” (2020), arXiv:2004.14140 [cond-mat.mes-hall].
[30] H. Pan, F. Wu, and S. Das Sarma, Phys. Rev. Research 2, 033087 (2020).
[31] H. Pan, F. Wu, and S. Das Sarma, Phys. Rev. B 102, 201104 (2020).
[32] H. Pan and S. D. Sarma, “Interaction-driven filling-induced metal-insulator transitions in 2d moiré lattices,” (2020), arXiv:2012.04554 [cond-mat.str-el].
[33] T. Devakul, V. Crépel, Y. Zhang, and L. Fu, Nature Communications 12, 6730 (2021).
[34] F. Wu, T. Lovorn, E. Tutuc, I. Martin, and A. H. MacDonald, Phys. Rev. Lett. 122, 086402 (2019).
[35] H. Tang, S. Carr, and E. Kaxiras, “Geometric origins of topological insolation in twisted layered semiconductors,” (2021), arXiv:2101.04867 [cond-mat.mtrl-sci].
[36] P. Stampfl, Helvetica Physica Acta 59, 1260 (1986).
[37] S. J. Ahn, P. Moon, T.-H. Kim, H.-W. Kim, H.-C. Shin, E. H. Kim, H.-W. Cha, S.-J. Kahng, P. Kim, M. Koshino, Y.-W. Son, C.-W. Yang, and J. R. Ahn, Science 361, 782 (2018).
[38] J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, Journal of Physics: Condensed Matter 14, 2745 (2002).
[39] S. Plimpton, Journal of Computational Physics 117, 1 (1995).
[40] F. H. Stillinger and T. A. Weber, Phys. Rev. B 31, 5262 (1985).
[41] A. N. Kolmogorov and V. H. Crespi, Phys. Rev. B 71, 235415 (2005).
[42] M. H. Naik, I. Maity, P. K. Maiti, and M. Jain, The Journal of Physical Chemistry C 123, 9770 (2019).
[43] See supplemental material for more details.
[44] V. V. Enaldiev, V. Zólyomi, C. Yelgel, S. J. Magorrian, and V. I. Falko, Phys. Rev. Lett. 124, 206101 (2020).
[45] I. Maity, P. K. Maiti, H. R. Krishnamurthy, and M. Jain, Phys. Rev. B 103, L121102 (2021).
[46] S. Carr, S. Fang, Z. Zhu, and E. Kaxiras, Phys. Rev. Research 1, 013001 (2019).
[47] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. D. Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Scaccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, Journal of Physics: Condensed Matter 21, 395502 (2009).
[48] J. P. Perdew and A. Zunger, Phys. Rev. B 23, 5048 (1981).
[49] H. J. Monkhorst and J. D. Pack, Phys. Rev. B 13, 5186 (1976).
[50] V. R. Cooper, Phys. Rev. B 81, 161104 (2010).
[51] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).
[52] J. Deslippe, G. Samsonidze, D. A. Strubbe, M. Jain,
M. L. Cohen, and S. G. Louie, Computer Physics Communications 183, 1269 (2012).
[53] M. S. Hybertsen and S. G. Louie, Phys. Rev. B 34, 5390 (1986).
[54] S. Ismail-Beigi, Phys. Rev. B 73, 233103 (2006).
[55] J. Deslippe, G. Samsonidze, M. Jain, M. L. Cohen, and S. G. Louie, Phys. Rev. B 87, 165124 (2013).
[56] Z. Gong, G.-B. Liu, H. Yu, D. Xiao, X. Cui, X. Xu, and W. Yao, Nature Communications 4 (2013).
[57] G.-B. Liu, D. Xiao, Y. Yao, X. Xu, and W. Yao, Chem. Soc. Rev. 44, 2643 (2015).
[58] M. H. Naik and M. Jain, Phys. Rev. B 95, 165125 (2017).
[59] V. Vitale, K. Atalar, A. A. Mostofi, and J. Lischner, “Flat band properties of twisted transition metal dichalcogenide homo- and heterobilayers of mos₂, mose₂, ws₂ and wse₂,” (2021), arXiv:2102.03259 [cond-mat.mtrl-sci].
[60] T. Fukui, Y. Hatsugai, and H. Suzuki, Journal of the Physical Society of Japan 74, 1674 (2005).
[61] R. Zhao, G.-D. Xie, M. L. N. Chen, Z. Lan, Z. Huang, and W. E. I. Sha, Opt. Express 28, 4638 (2020).
[62] C. L. Kane and E. J. Mele, Phys. Rev. Lett. 95, 226801 (2005).
[63] C. L. Kane and E. J. Mele, Phys. Rev. Lett. 95, 146802 (2005).
Supplemental material for moiré induced topology and flat bands in twisted bilayer WSe$_2$: a first-principles study

Sudipta Kundu,$^1$ Mit H. Naik,$^{1,*}$ H. R. Krishnamurthy,$^1$ and Manish Jain$^1$

$^1$Center for Condensed Matter Theory, Department of Physics, Indian Institute of Science, Bangalore 560012, India
COMPUTATIONAL DETAILS AND METHODOLOGY:

We use the Twister code [1] to generate the twisted bilayer WSe$_2$ (tWSe$_2$). As the twist angle ($\theta$) approaches 0° or 60°, the number of atoms in the moiré supercell becomes very large (Table 1). Relaxing the twisted structure is important to obtain the correct electronic structure and localization. Owing to the large number of atoms, relaxation of these large systems using standard density functional theory (DFT) calculations is challenging. Here, we employ a multiscale approach to relax our systems and calculate the electronic structure of the relaxed systems, as discussed below.

The unit cell lattice parameter of WSe$_2$ is 3.25 Å in all our calculations. We relax our twisted structures using classical force fields, employing LAMMPS package [2]. The intralayer interaction is described by the Stillinger-Weber force field [3] and the interlayer interaction by the Kolmogorov-Crespi forcefield [4, 5] which is fitted to van der Waals (vdW) corrected DFT calculations. The forces are minimized using the conjugate gradient method till the force on each atom becomes less than $10^{-8}$ eV/Å.

With the relaxed moiré structure, we perform DFT calculations to obtain the electronic structure. Our DFT calculations are performed using the atomic-orbital based code:

| Twist angle($\theta$) | No. of atoms | Length (Å) |
|-----------------------|--------------|------------|
| 7.3°, 52.7°            | 366          | 25.39      |
| 6.0°, 54.0°            | 546          | 31.01      |
| 5.1°, 54.9°            | 762          | 36.63      |
| 4.4°, 55.6°            | 1014         | 42.26      |
| 3.9°, 56.1°            | 1302         | 47.89      |
| 3.5°, 56.5°            | 1626         | 53.51      |
| 3.14°, 56.86°          | 1986         | 59.14      |
| 2.88°, 57.12°          | 2382         | 64.77      |
| 2.65°, 57.35°          | 2814         | 74.80      |
| 2.28°, 57.72°          | 3786         | 81.66      |
| 1.89°, 58.11°          | 5514         | 98.55      |
SIESTA [6]. We use the polarized-double-zeta basis set to expand the wavefunctions. We use norm-conserving pseudopotentials and the exchange-correlation is treated within the local density approximation (LDA) [7]. As vdW correction in DFT only affects the relaxation of the structure, we do not use vdW correction further in our electronic structure calculations. 10Å vacuum is added to restrict interaction between periodic images in the out-of-plane direction. We use a 12×12×1 Monkhorst-Pack k-point sampling [8] to converge the ground state charge density for the bilayer unit cell. For the moiré cell, we accordingly scale the k-grid, and the large moiré cells are only sampled at the Γ M point of their moiré Brillouin zone (MBZ). All DFT calculations include spin-orbit interaction.

We have performed GW calculations on the untwisted bilayer systems corresponding to different high symmetry stackings using the BerkeleyGW package [9] with a 12×12×1 Monkhorst-Pack k-grid. The mean-field wavefunctions for the GW calculations are generated using Quantum Espresso [10]. To calculate the interlayer separation for the unit cells, we used the DF-C09 [11] van der Waals functional and the exchange-correlation was treated using LDA. The dielectric (epsilon) matrix is expanded in a plane-wave basis with energy up to 15 Ry and extended to finite frequencies using the generalized plasmon pole model [12]. The Coulomb interaction is truncated to compute the dielectric matrix, and the self-energy [13]. We use 1000 bands (without spin-orbit coupling) to construct the epsilon matrix and 1000 spinors for calculating the self-energy. We use the static remainder technique [14] to accelerate the convergence of the self energy with respect to empty bands. While the number of bands is insufficient to obtain the absolute convergence of the band edges with respect to vacuum, it is sufficient to calculate the relative position of the valence band (VB) maximum at the Γ point and the K point in the unit cell Brillouin zone. To check for the convergence of the epsilon matrix with respect to the number of bands, we have done calculations for the AB stacking with 5000 bands (without spin-orbit coupling) and 1000 spinors for the construction of the epsilon matrix. Our calculations show that using 1000 bands for computing the epsilon matrix is sufficient to converge the relative energy difference. Table 2. shows our results for the different high symmetry stackings. In all stackings, the VB maximum at the K point of the unit cell Brillouin zone (UBZ) is higher in energy than at the Γ point of the same.

Furthermore, we have optimized the AB stacking structure with the generalized gradient approximation [15] and the DF-C09 [11] van der Waals functional. Even in this case, our
GW calculations show that the VB maximum at the K point is higher than at the Γ point by 191 meV.

![Graph showing variation of Δ with interlayer separation for three different lattice constants (a).](image)

**FIG. 1.** Variation of Δ with interlayer separation for three different lattice constants (a). The vertical dashed lines represent the equilibrium interlayer separation at a particular lattice constant. The equilibrium interlayer separation for a = 3.30 Å is 6.43 Å at which Δ is 2.2 meV. Hence, the blue dashed line is not visible.

It is to be noted here that the energy difference between the VB maximum at the K and Γ points (Δ ≡ E_K - E_Γ) depends strongly on the interlayer separation or the distance between the inner Se atoms. Furthermore, the distance between W and Se also influences Δ. The interlayer separation and Δ are very sensitive to strain. In Fig 1, we show the variation of Δ with the interlayer separation for three different lattice constants (a). We find that the Δ decreases with increasing lattice constant for a given value of interlayer separation. Δ changes sign at a smaller value of interlayer separation with increasing lattice constant. The changes in sign of Δ for a = 3.25 Å and a = 3.35 Å occur outside the range shown in Fig. 1. Furthermore, the W-Se distance and Se-Se distance are also dependent on the functional used. A different functional, for example Grimme’s D2 functional, leads to smaller interlayer separation, which can lead to different ordering of the VB edge at K and Γ.

**HIGH SYMMETRY STACKINGS IN MOIRÉ SUPERLATTICE**

Moiré superlattices (MSLs) in transition metal dichalcogenides have different sets of high symmetry stackings depending on whether θ is close to 0° or 60°. For θ close to 0°, the MSL has three high symmetry stackings: AA (W on top of W and Se on top of Se), AB
TABLE II. The VB energies at the Γ and K points are reported. The first entry for each stacking is for the first VB, and the second one is for the next VB. All energies are in eV. The structures are optimized with LDA (except for the last row), and the interlayer interaction is described by vdW-DFC09. The epsilon matrix for the GW calculations is constructed with 1000 bands, and the quasiparticle energies are calculated using 1000 spinors for the first five rows. The sixth and seventh rows are calculated with 5000 bands and 1000 spinors respectively in the epsilon calculations. The epsilon matrix of the last row is again calculated with 1000 bands.

|     | DFT       |       | GW       |       |
|-----|-----------|-------|----------|-------|
|     | E_Γ       | E_K   | Δ(=E_K - E_Γ) | E_Γ   | E_K   | Δ(=E_K - E_Γ) |
| AA' (2-H) | 1.96 2.06 0.10 | -0.12 | 0.0012 0.12 |
|       | 1.96 2.06 0.10 | -0.12 | -0.0005 0.12 |
| AB' (BW-W) | 1.89 2.07 0.18 | -0.21 | 0.02 0.23 |
|       | 1.89 2.07 0.18 | -0.21 | 0.02 0.23 |
| A'B (BSe-Se) | 1.72 2.07 0.35 | -0.42 | 0.0052 0.42 |
|       | 1.72 2.07 0.35 | -0.42 | 0.0042 0.42 |
| AA (3-R) | 1.71 2.10 0.39 | -0.15 | 0.28 0.43 |
|       | 1.71 2.03 0.32 | -0.15 | 0.23 0.38 |
| AB (BW-Se) | 1.96 2.10 0.14 | -0.13 | 0.06 0.19 |
|       | 1.96 2.03 0.07 | -0.13 | -0.0058 0.13 |
|       | 5000 bands for constructing epsilon matrix |
| AB (BW-Se) | 1.96 2.10 0.14 | -0.20 | -0.0027 0.20 |
|       | 1.96 2.03 0.07 | -0.20 | -0.07 0.13 |
|       | 1000 spinors for constructing epsilon matrix |
| AB (BW-Se) | 1.96 2.10 0.14 | 0.21 | 0.41 0.20 |
|       | 1.96 2.03 0.07 | 0.21 | 0.34 0.13 |
|       | The structure is optimized with GGA and vdW-DFC09. 1000 bands are used to construct epsilon matrix. |
| AB (BW-Se) | 0.96 1.08 0.12 | 0.0006 0.19 0.19 |
|       | 0.96 1.02 0.06 | 0.0006 0.12 0.12 |
(W on top of Se) and BA (Se on top of W) (Figs. 2 (a–b)). The staggered stackings AB and BA are of lower energy than the eclipsed stacking AA. In AA stacking, the Se atoms from both layers face each other, and due to steric hindrance, the interlayer separation is the maximum, at 7.1 Å. In the moiré supercells, the interlayer separation is minimum (6.5 Å) for the AB and BA stackings. The interlayer separation of any given stacking in WSe$_2$ is larger than that of MoS$_2$ because of the denser electron cloud in Se compared to S. In Figs. 3 (a–c), we show band structures of these high-symmetry structures. Additionally, we show the band structure of the WSe$_2$ monolayer in Fig. 3(b) (red dashed line). The VB maxima and conduction band (CB) minima of all these bilayer structures are located at the K and Q points, respectively. The VB maximum at the K point and the next band have parallel spins (Fig. 3(a)), and there is a splitting of 30–50 meV between them depending on the stacking. The split due to spin-orbit coupling is 445 meV which is also present in the monolayer (Fig. 3(b)). Spin-orbit splitting is also present at the CB minimum at the Q point. Additionally, there is interlayer splitting due to hybridization in the bilayers, which is absent in the monolayer [16]. In this regard, it is to be noted that the interlayer splitting of the VB maximum at the K point is very small as the bands at the K point have very localized wavefunctions. On the other hand, the CB minimum wavefunctions at the Q point and the VB maximum wavefunctions at the Γ point are more delocalized, leading to larger interlayer splittings at these points.

![FIG. 2. Relaxed MSL of tWSe$_2$ with θ values of 2.28° (a) and 57.72° (c). The rectangular boxes show different high symmetry stackings present in these structures. (b) and (d): Side views of (a) and (c) respectively. W and Se atoms are shown in red and green colours respectively.](image)
The other set of MSL structures with $\theta$ close to 60° contains AA$'$ (W on top of Se and Se on top of W), A$'$B (Se on top of Se) and AB$'$ (W on top of W) high symmetry stackings (Figs. 2 (c–d)). AA$'$ (also known as 2H stacking) is the most stable structure of bilayer WSe$_2$ and has an interlayer separation of 6.5 Å. In A$'$B stacking, Se atoms from both layers face each other, leading to an increased interlayer separation of 7.1 Å due to electron-electron repulsion. A$'$B stacking hence has the highest energy among these three high symmetry stackings. Figs. 3 (d–f) show band structures of AA$'$, AB$'$ and A$'$B, respectively. Due to the presence of inversion symmetry in these high symmetry stackings, the VB maximum are two-fold degenerate at the K point and have antiparallel spin (Fig. 3(d)). The CB minima at the Q point are doubly degenerate.

FIG. 3. Band structures of (a) AA, (b) AB, (c) BA, (d) AA$'$, (e) AB$'$ and (f) A$'$B high symmetry stacked bilayers. The insets in (a) and (d) show the spin configuration of the top two bands at the VB edge at the K point. The red dashed lines in (b) represent the band structure of the monolayer.
STRUCTURAL ASPECTS

In this section, we discuss the relaxed MSLs. As the eclipsed stacking (AA) region has higher energy compared to the staggered stacking regions (AB and BA, which are degenerate), upon relaxation, the AB, BA regions expand equally while the AA region shrinks, giving rise to a hexagonal pattern in MSL with $\theta$ close to 0°. The relaxation effect becomes more evident as $\theta$ approaches 0°. The relaxation pattern can be depicted by the variation of the interlayer separation. We plot the distribution of the interlayer separation in Fig. 4(a). The interlayer separation varies between 6.5–7.1 Å. AB and BA have the minimum interlayer separation. The relaxation process also induces local strain and corrugation in the MSL. As a result, the structure of the MSL is determined by the competition between strain-energy cost and stacking-energy gain. Figs. 4(b) and (c) show the strain distribution in layer 1 (L1) and layer 2 (L2), respectively. We plot the strain for every W atom along the direction of its six nearest W neighbours. In both layers, the strains are zero at the high symmetry stackings and are localized in the domain walls. The strain distribution in each layer is conjugate to the other i.e. at a given point in the MSL along a certain direction, if the strain is compressive in L1, it becomes tensile in L2.

![Figure 4](image-url)

**FIG. 4.** (a): Distribution of the interlayer separation of tWSe$_2$ for $\theta$=2.28°. (b) and (c): Strain distributions in the two layers.

The MSL with $\theta$ close to 60° has AA’, AB’ and A’B. The fact that AA’ has lower energy than AB’ manifests in the reduced symmetry of the relaxed MSL, and gives rise to Reuleaux triangles in the moiré patterns. The highest energy stacking A’B occupies the smallest area. The interlayer separation varies between 6.5–7.1 Å. The relaxation pattern can be visualized...
FIG. 5. (a): Distribution of the interlayer separation of tWSe$_2$ for $\theta=57.72^\circ$. (b) and (c): Strain distributions in the two layers.

by plotting the variation of interlayer separation in the MSL (Fig. 5(a)). We also plot the distribution of strain induced by relaxing the MSL in Figs. 5(b) and (c). We again find that the strains are localized on the domain walls and are zero at the high symmetry stackings. It is to be noted that lattice reconstruction, where the lattice constant of the MSL changes, is not observed in the angle ranges we have explored [17].

CALCULATION OF CHERN NUMBER

The moiré Brillouin zones (MBZ) studied in our work are very small compared to the UBZ. The flat bands at the VB edge in MBZ hence arise from monolayer bands so close to the K and K’ points of UBZ that they can be associated with up and down spins, respectively, with great accuracy. (For example, in the 2.28° twisted WSe$_2$ (tWSe$_2$) the flat bands at the VB edge arise from K, K’ points and k-points within a circle around K, K’ point in UBZ with the radius of 3.6% of the reciprocal lattice vector of UBZ.) Due to the large spin-orbit splitting at the K and K’ points, bands associated with the complementary spin will be $\sim 436$ meV below and contribute only to bands inside the VB continuum in the MBZ. In particular, the flat bands at the VB edge in twisted WSe$_2$ with twist angle close to 0° arise from the K point of the top layer and K’ point of the bottom layer. As a result, the bands are doubly degenerate with antiparallel spin, and we can differentiate between the flat bands arising from the K and K’ points by following the spin.

More specifically, we discretize the MBZ into a regular k-grid (in terms of the reciprocal
lattice vectors of the MBZ); for example, we sample the MBZ of 2.28° tWSe2 with a 6×6 k-grid. We compute the expectation value of the Pauli matrices at each grid point for the first six bands at the VB edge. Fig. 6 depicts the k-grid in the hexagonal MBZ as well as the expectation value of \( \langle \sigma_z \rangle \) and \( \langle \sigma_x \rangle \) for the first two bands at the VB edge for 2.28° tWSe2. As can be seen, the value of \( \langle \sigma_z \rangle \) is either close to +1 or −1 (|\( \langle \sigma_z \rangle \) | > 0.9) and \( \langle \sigma_x \rangle \) is very small (|\( \langle \sigma_x \rangle \) | < 0.03). This implies that spin is essentially a good quantum number for the bands arising from the K/K’ points of the UBZ. Hence we can identify the bands according to the \( \langle \sigma_z \rangle \) value in the whole MBZ and calculate the Chern number for the up spin (\( \langle \sigma_z \rangle \) close to +1) and down spin (\( \langle \sigma_z \rangle \) close to −1) separately. We find that the Chern numbers for the up and down spin bands to be opposite, making the total Chern number of the system to be zero. It is to be noted that purely energetically, the first band is spin-up at some k-points in the MBZ and spin-down at the rest of the k-points (Fig. 6(a)). The other band has the opposite spin to that of the first band (Fig. 6(b)).

To calculate the Chern numbers, we generate the Bloch wavefunctions for the various bands on the above mentioned k-grid using density functional theory (DFT) calculations. As the MBZ is small, the 6×6 sampling turns out to be sufficient for 2.28° tWSe2 and divides the MBZ into 36 plaquettes [18, 19]. The Chern number for each spin (see discussion above) is then calculated following the equation:

\[
C_{n,s} = \frac{1}{2\pi} \sum_{k \in MBZ} \text{Im} \{ \ln[U_{k_1 \rightarrow k_2} U_{k_2 \rightarrow k_3} U_{k_3 \rightarrow k_4} U_{k_4 \rightarrow k_1}] \}
\]

where \( U_{k_{\alpha} \rightarrow k_{\beta}} \equiv \frac{\langle u_{n,s,k_{\alpha}} | u_{n,s,k_{\beta}} \rangle}{|\langle u_{n,s,k_{\alpha}} | u_{n,s,k_{\beta}} \rangle|}, \quad \alpha, \beta = 1, 2, 3, 4. \) \( u_{n,s,k} \) is the periodic part of the Bloch
FIG. 7. (a): The PDOS contributions of AA, AB, Br and BA. (b): Enlarged view of PDOS near the VB edge. The violet dashed line marks the first peak due to $\Gamma$. (c)-(f): The contribution of Orb$_z$ (green line) and Orb$_{xy}$ (red line) to the PDOS (black line) of AB, AA, Br and BA respectively. Orb$_z$ consists of the $d_{z^2}$ orbital of W and the $p_z$ orbital of Se, while Orb$_{xy}$ represents the $d_{x^2-y^2}$ and $d_{xy}$ orbitals of W atom and the $p_x$, $p_y$ orbitals of Se atom.

wavefunction $(\psi_{n,s,k} = u_{n,s,k} e^{i k \cdot r})$ and $n, s$ refer to the band number and spin of the band. $k_1, k_2, k_3, k_4$ are the four vertices of each plaquette ordered in an anti-clockwise fashion. It is to be noted that we have employed different k-grids for different values of $\theta$ as the MBZ becomes smaller with decreasing $\theta$; for example, the MBZ of 2.88° tWSe$_2$ is sampled with an 8×8 k-grid. For 5.1° tWSe2, we checked the convergence of the $C_n$ by computing the $C_n$ with 12×12 and 30×30 k-grids and the $C_n$ are same for both.

**COMPARISON WITH EXPERIMENTS**

Scanning tunnelling measurements show flat bands coming from the $\Gamma$ point of the UBZ for both kinds of MSL [20]. We have shown the local density of states (LDOS) plots corresponding to those states in the main manuscript. To further compare with experimental findings, we calculate the projected density of states (PDOS). We first consider a 2.88° tWSe$_2$ to compare with the STM on 3° tWSe$_2$. We plot the PDOS of 2.88° tWSe$_2$ in Fig.
7(a). Fig. 7(b) shows the zoomed in view of the PDOS near the VB edge. The first few states (up to 178 meV below the VB maximum of MSL) arise from the K point of the UBZ. It is evident from Fig. 7(b) that the first peak has a non-zero contribution from AA, AB, BA and Br (domain walls between AB and BA). Hence, the LDOS calculated in the energy range of the first set of doubly degenerate bands (V1 of Fig. 1(a) in main manuscript) at the VB edge is delocalized in the whole MSL. The violet dashed line in Fig. 7(b) marks the first peak due to the band arising from the the Γ point of the UBZ. The states of this band are primarily localized in the AB, BA and Br regions. The wavefunction at the Γ point of the UBZ consists of the $d_z^2$ orbital of the W atom and $p_z$ orbitals of the Se atoms. We will refer to these orbitals as Orb$_z$. The component along the out-of-plane direction of these orbitals makes the interlayer interaction stronger, and hence larger interlayer splitting is found at the Γ point. On the other hand, the $d_{x^2-y^2}$ and $d_{xy}$ orbitals of the W atom and the $p_x$, $p_y$ orbitals of the Se atoms (Orb$_{xy}$) contribute to the K point wavefunctions of the UBZ. We plot the contributions of Orb$_z$ (green line) and Orb$_{xy}$ (red line) to the PDOS (black line) of the AA and Br regions in Figs. 7(d) and (e), respectively. The peak marked by the violet dashed line has the main contribution from Orb$_z$, confirming the fact that this peak arises from the Γ point of the UBZ. We also show the contribution of Orb$_z$ and Orb$_{xy}$ to the PDOS of the AA and Br regions in Figs. 7(d) and (e), respectively.

Next, we consider the 57.72° tWSe$_2$ to compare with the STM on the 57.5° tWSe$_2$. We plot the PDOS in Fig. 8(a). Fig. 8(b) shows the enlarged view of PDOS near the VB edge. The peak marked by the violet dashed line in Fig. 8(b) arises from the Γ point of the UBZ and has contributions from AA’ stacked regions, which implies that the Γ derived flat band states localize on AA’. We plot the contributions of Orb$_z$ (green line) and Orb$_{xy}$ (red line) to the PDOS (black line) of AA’, AB’, Br (bridge region between AA’ and AB’) and A’B in Figs. 8(c)–(f). The large contribution of Orb$_z$ to the peak marked by the violet dashed line (Fig. 8(c)) shows the onset of the Γ derived state. Furthermore, this peak has a large contribution from the AA’ stacking and negligible contributions from other stackings.

**MOIRÉ HAMILTONIAN**

The electronic structure at the VB edge of the relaxed MSL can be fit to a continuum model [21] which can be used to go beyond the one-particle physics in these systems. The
FIG. 8. (a) ((b)): The PDOS (PDOS near the VB edge) due to $AA'$, $A'B$, $Br$ and $AB'$. The violet dashed line in (b) corresponds to the peak arising due to the $\Gamma$ point. (c): The contribution of $\text{Orb}_z$ (green line) and $\text{Orb}_{xy}$ (red line) to the PDOS (black line) of $AA'$ to identify the $\Gamma$ and $K$ derived peaks. (d), (e) and (f): The orbital resolved PDOS for $A'B$, $Br$ and $AB'$. 

The moiré Hamiltonian at the $K$ valley for a particular spin state can be written as:

$$H = \begin{pmatrix} \frac{-\hbar^2(k-\kappa)^2}{2m^*} + \Delta_+(r) & \Delta_T(r) \\ \Delta_T^*(r) & \frac{-\hbar^2(k+\kappa)^2}{2m^*} + \Delta_-(r) \end{pmatrix}$$ (2)

Here, $\kappa_{\pm}$ are located at the MBZ corners and correspond to the $K_M$ and $K'_M$ points. For $\theta$ close to $0^\circ$, the layer dependent moiré potential $(\Delta_{\pm})$ is given by:

$$\Delta_{\pm}(r) = \sum_{j=1,3,5} 2V\cos(G_j.r \pm \psi)$$ (3)

$V$ represents the potential amplitude and $\psi$ describes the spatial pattern. $G_j$ is the $j^{th}$ moiré reciprocal lattice vector in the first shell obtained by an anti-clockwise rotation of the first vector $G_1$ by $(j-1)\pi/3$. The interlayer tunneling $(\Delta_T)$ is given by:

$$\Delta_T(r) = w(1 + e^{-iG_2.r} + e^{-iG_3.r})$$ (4)

where $w$ is the interlayer tunneling strength.
FIG. 9. (a) and (b): Band structures of 3.5° and 2.28° tWSe₂ with no interlayer tunneling (i.e. of the relaxed MSL monolayer). The blue solid and black dashed lines show the model band structure and the DFT result respectively. (c) and (d): Band structures of 5.1° and 2.28° MSLs. The nearly-doubly-degenerate DFT bands have been averaged. (e) and (f) ((g) and (h)): Localization of wavefunctions of V1 and V2 (marked in Fig. (d)) at the Γₚ (Kₚ) point.

First, we fit the model with no interlayer tunneling to the DFT results of the relaxed MSL monolayer. Figs. 9(a) and (b) (blue lines) show the band structures obtained by solving the model with w = 0 for 3.5° and 2.28°. For V = 21 meV and ψ = 94°, the model band structures are in good agreement with DFT results. Fitting the model band structures including the interlayer tunneling, we find w = 30 meV. The band structures with (V, ψ, w) = (21 meV, 94°, 30 meV) for 5.1° and 2.28° are shown in Figs. 9(c) and (d). The Chern numbers for the first two bands are consistent with the DFT results. We depict the localization of the wavefunctions of the first two spin-up bands of the 2.28° MSL (V1 and V2 in Fig. 9(d)) at the Γₚ and Kₚ points in Figs. 9(e)–(h) and the results are in good agreement with the localization of the DFT wavefunctions (Fig. 3 in the main manuscript). Hence, the model is able to describe the first two bands. A limitation of the model would seem to be that neither the Chern number nor the localization characteristics of the third band agrees with the DFT results. It is to be noted that F. Wu et al. [21] gives a different set of parameters obtained from the DFT calculation on bilayer unit cells. Although the parameters are very different, the Chern numbers and the localization of the first two bands are consistent with
FIG. 10. (a): Band structures of $57.72^\circ$ tWSe$_2$. Blue solid line shows the model band structure. (b)–(d): Localization of V1, V2 and V3 at $\Gamma_M$.

our DFT and model calculations. Widely different parameters can describe the same band structure because of the fact that the bands are mostly determined by kinetic energy. The intralayer and interlayer potentials act as perturbations to split and modulate the bands.

For $\theta$ close to $60^\circ$, the intralayer potential becomes:

$$\Delta(r) = \sum_{j=1,3,5} 2V \cos(G_j \cdot r + \psi)$$

and the effective interlayer tunneling is zero. Hence, fitting the model with DFT results for the $57.72^\circ$ tWSe$_2$ (Fig. 10(a)), we find the parameters to be $(V, \psi, w) = (26 \text{ meV}, 46^\circ, 0)$. Furthermore, we show the localization of the wavefunctions in Figs. 10(b)–(d) and they are in excellent agreement with the DFT results.

* Present address: Department of Physics, University of California at Berkeley, California 94720, USA and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA.

[1] M. H. Naik and M. Jain, Phys. Rev. Lett. 121, 266401 (2018).
[2] S. Plimpton, Journal of Computational Physics 117, 1 (1995).
[3] F. H. Stillinger and T. A. Weber, Phys. Rev. B 31, 5262 (1985).
[4] A. N. Kolmogorov and V. H. Crespi, Phys. Rev. B 71, 235415 (2005).
[5] M. H. Naik, I. Maity, P. K. Maiti, and M. Jain, The Journal of Physical Chemistry C 123, 9770 (2019).
[6] J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, Journal of Physics: Condensed Matter 14, 2745 (2002).

[7] J. P. Perdew and A. Zunger, Phys. Rev. B 23, 5048 (1981).

[8] H. J. Monkhorst and J. D. Pack, Phys. Rev. B 13, 5188 (1976).

[9] J. Deslippe, G. Samsonidze, D. A. Strubbe, M. Jain, M. L. Cohen, and S. G. Louie, Computer Physics Communications 183, 1269 (2012).

[10] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. D. Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, Journal of Physics: Condensed Matter 21, 395502 (2009).

[11] V. R. Cooper, Phys. Rev. B 81, 161104 (2010).

[12] M. S. Hybertsen and S. G. Louie, Phys. Rev. B 34, 5390 (1986).

[13] S. Ismail-Beigi, Phys. Rev. B 73, 233103 (2006).

[14] J. Deslippe, G. Samsonidze, M. Jain, M. L. Cohen, and S. G. Louie, Phys. Rev. B 87, 165124 (2013).

[15] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996).

[16] M. H. Naik and M. Jain, Phys. Rev. B 95, 165125 (2017).

[17] I. Maity, P. K. Maiti, H. R. Krishnamurthy, and M. Jain, Phys. Rev. B 103, L121102 (2021).

[18] T. Fukui, Y. Hatsugai, and H. Suzuki, Journal of the Physical Society of Japan 74, 1674 (2005).

[19] R. Zhao, G.-D. Xie, M. L. N. Chen, Z. Lan, Z. Huang, and W. E. I. Sha, Opt. Express 28, 4638 (2020).

[20] Z. Zhang, Y. Wang, K. Watanabe, T. Taniguchi, K. Ueno, E. Tutuc, and B. J. LeRoy, Nature Physics (2020).

[21] F. Wu, T. Lovorn, E. Tutuc, I. Martin, and A. H. MacDonald, Phys. Rev. Lett. 122, 086402 (2019).