

Nonrelativistic spin splittings and altermagnetism in twisted bilayers of centrosymmetric antiferromagnets

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Magnetism-driven nonrelativistic spin splittings (NRSS) are promising for highly efficient spintronics applications. Although 2D centrosymmetric (in four-dimensional spacetime) antiferromagnets are abundant, they have not received extensive research attention owing to symmetry-forbidden spin polarization and magnetization. Here, we demonstrate a paradigm to harness NRSS by twisting the bilayer of centrosymmetric antiferromagnets with commensurate twist angles. We observe *i*-wave altermagnetism and spin-momentum locking by first-principles simulations and symmetry analysis on prototypical MnPSe₃ and MnSe antiferromagnets. The strength of NRSS (up to 80 meVÅ) induced by twisting is comparable to SOC-induced linear Rashba-Dresselhaus effects. The results also demonstrate how applying biaxial strain and a vertical electric field tune the NRSS. The findings reveal the untapped potential of centrosymmetric antiferromagnets and thus expand the material's horizons in spintronics.

Spin splittings in the electronic structure of crystalline solids play a pivotal role in spintronics applications (e.g., spin transistor) [1, 2]. The conventional spin-orbit coupling (SOC) induced Rashba-Dresselhaus [3–6] in nonmagnetic and Zeeman effects in ferromagnetic (FM) materials create spin splittings under certain crystalline (i.e., inversion [$\mathcal{P}$]) and time-reversal symmetry ($\mathcal{T}$) breaking [7], respectively. SOC-induced spin splitting and resulting spin polarization engender spin-orbit torques [8], while FM spin polarization has been widely known for spin generation and detection [1]. However, the SOC effect introduces spin dephasing mechanisms [9–11], limiting the practical application. In addition, materials with heavy elements having significant SOC imparts additional challenges, including scarcity, toxicity, and instability. Therefore, nonrelativistic spin splitting (NRSS) is an important avenue to pursue.

Recently, antiferromagnetic (AFM) materials have emerged as viable substitutes for nonmagnetic and FM materials, benefiting from resilience towards stray fields, ultrafast dynamics, and magnetotransport effects [12–16]. The coupling of spin to lattice degrees of freedom via a staggered collinear compensated magnetism leads to alternating NRSS, termed altermagnetism [17–19]. Numerous efforts have been undertaken to investigate NRSS in AFM materials by breaking combined $\mathcal{PT}\tau$ and/or $U\tau$ symmetries, where U and τ are spinor and translation symmetry, respectively [20–26]. Nevertheless, the majority of AFM spin splittings are limited to bulk materials (e.g., MnF₂ [21, 27], LaMnO₃, and MnTiO₃ [23]), require SOC (e.g., MnS₂ [23] and ZnV₂O₄ [28]), or external perturbation [29–31].

Since the experimental revelation of 2D magnetic ordering, 2D vdW magnetic materials have garnered significant attention in scientific research, emerging as promising contenders for future information technology. Inter-

estingly, two recent works focus on spin splitting in FM NiCl₂ [32] and FeBr₂ (although “hidden”) [33] monolayers vdW stacked antiferromagnetically. In contrast, antiferromagnetism-induced spin splitting among centrosymmetric materials with AFM order within each layer is not achieved due to $\mathcal{PT}$ symmetry-enforced spin-degeneracy. Despite being abundant in nature, this impedes practical applications of 2D centrosymmetric AFM materials [34–38].

This study generates NRSS and altermagnetism in $\mathcal{PT}$ -symmetric AFM monolayers vdW stacked with a relative twist. We perform density functional theory (DFT) simulations on twisted bilayer (tb-) MnPSe₃ and MnSe as prototypical candidates. The *i*-wave spin-momentum coupling arises in the 2D BZ for θ ($\neq 0^\circ, 60^\circ$) tb-MnPSe₃ and MnSe. Based on the symmetry analysis, we find that the strengths of NRSS along specific crystallographic k -paths are comparable to the conventional SOC-induced Rashba-Dresselhaus effects. Moreover, external perturbations (i.e., electric and strain fields) provide exceptional tunability to NRSS.

MnPSe₃ and MnSe (space group #162, $P\bar{3}1m$) represent two distinct classes of vdW materials that possess exceptional exfoliation properties [34–38]. Unlike the majority of other 2D magnetic materials, they exhibit an AFM arrangement, conforming to the conventional collinear Néel order on the honeycomb lattice [Fig. 1]. This in-plane antiferromagnetism is different from the A-type antiferromagnetism observed in various other 2D vdW compounds, i.e., MnBi₂Te₄ [39], CrI₃ [40], and CrSBr [41], where individual layers exhibit FM order but stack antiferromagnetically. The antiferromagnetism of MnPSe₃ is “truly” in-plane and differs from that of MnSe, where Mn ions with opposite magnetic moments (Mn_A and Mn_B) form unusual out-of-plane ordering within the individual layer. Note that the orientation of on-site magnetic moments concerning the lattice only matters if SOC is included. Therefore, nonrelativistic spin-group formalism is described as the symmetry transformation of decoupled real and spin space [18, 42–

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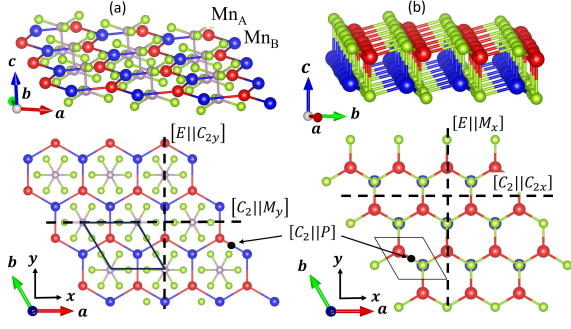


FIG. 1. Crystal structure of the monolayer (a) MnPSe_3 and (b) MnSe . The red and blue spheres indicate Mn atoms with the opposite collinear magnetic densities. The brown and green spheres represent P and Se atoms, respectively. The Cartesian (x, y, z) coordinate system and the hexagonal unit cell (with solid black lines) are shown for each case. The nontrivial spin-group symmetries are also highlighted. E and C_2 represent identity and two-fold rotation (about an axis perpendicular to spins) in spin-space, respectively. M_i and C_{2j} denote the mirror reflection perpendicular to the i axis and the two-fold rotation parallel to the j axis in real-space, respectively. $\mathcal{P}$ represents the real-space inversion.

45]. The spin-symmetry operations $[R_i||R_j]$ of monolayer MnPSe_3 and MnSe are indicated in Fig. 1, where the transformation on the left (right) of the double vertical bar acts on the only spin (real) space. In addition, collinear magnets always have additional symmetry $[\bar{C}_2||T]$ arising from spin-only groups, where $\bar{C}_2$ is the two-fold rotation perpendicular to the collinear spin axis, followed by spin-space inversion. Mn_A and Mn_B sublattices are connected through $[C_2||\mathcal{P}]$ symmetry in monolayer MnPSe_3 and MnSe . $[C_2||\mathcal{P}][\bar{C}_2||T] (\equiv \mathcal{PT}^1)$ symmetry transforms energy eigenstate $E(k, \sigma)$ as $[C_2||\mathcal{P}][\bar{C}_2||T]E(k, \sigma) = [C_2||\mathcal{P}]E(-k, \sigma) = E(k, -\sigma)$, leading to spin degeneracy throughout the Brillouin zone (BZ). We have verified that through DFT+U calculations performed on the projector augmented wave method [46] based VASP [47] code (methods are detailed in Sect. I of supplemental material (SM) [48]). DFT simulated energy bands for monolayer MnPSe_3 and MnSe are doubly degenerate (see Sect. II in SM [48]). The semiconducting MnPSe_3 and MnSe have a magnetic moment of $\sim 4.5\mu_B/\text{Mn}$ with weak interlayer coupling. In addition, $[C_2||\tau]$ can also enforce spin-degeneracy by connecting opposite spin sublattices by translation (τ) as $[C_2||\tau]E(k, \sigma) = E(k, -\sigma)$. Since 2D systems have only in-plane components of momentum $k_{||}$, nonrelativistic Hamiltonian for 2D systems may have symmetries other than $[C_2||\mathcal{P}]$ and $[C_2||\tau]$ enforcing spin degeneracy. For example, $[C_2||M_z]$ symmetry also enforces spin degeneracy throughout BZ in 2D materials, with $M_z : M_z k_{||} = k_{||}$

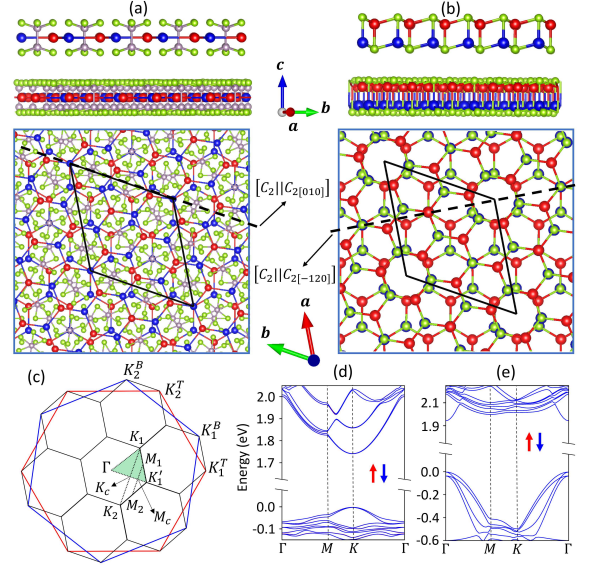


FIG. 2. The Moiré superlattices formed by twisting bilayer of (a) MnPSe_3 and (b) MnSe by 21.79° . (c) The Moiré BZ construction uses BZs of the top and bottom layers. The large red and blue hexagons are the first BZ of the top and bottom layers, respectively, and black hexagons represent the BZ corresponding to the Moiré superlattice. Spin-polarized band structure of (d) tb-MnPSe_3 and (e) tb-MnSe at the PBE level. The red and blue bands denote spin-up and spin-down states, respectively.

as a planer mirror reflection [see Sec. II of SM [48] for details]. That makes achieving NRSS even more difficult for 2D materials. In the case of MnPSe_3 and MnSe monolayers, $[C_2||M_z]$ is already broken [Figs. 1(a) and 1(b)], whereas type-III Shubnikov MSG ensures $[C_2||\tau]$ symmetry-breaking [49]. The only symmetry preserving spin degeneracy is $[C_2||\mathcal{P}]$ symmetry in monolayers MnPSe_3 and MnSe .

Bilayer MnSe and MnPSe_3 are obtained from monolayers with various high-symmetry stackings as used in Ref [30]. Spin-up and spin-down states are degenerate for AA, AA', AB, and BA stackings [see Sec. II of SM [48]]. The $[C_2||\mathcal{P}]$ symmetry enforces double degeneracy in AA, AB, and BA, whereas double degeneracy in AA' stacking is protected by the $[C_2||M_z]$. Therefore, high-symmetry stackings are not an ideal for SOC-unrelated spin splitting in 2D $\mathcal{PT}$ -symmetric antiferromagnets.

Commensurate twisted bilayers are obtained using coincidence lattice theory [50] by taking the AA bilayer as the untwisted limit to break $\mathcal{PT}$ symmetry. A periodic lattice structure, including Moiré superlattice, can form with special twist angle θ , $\cos \theta = \frac{n^2 + 4mn + m^2}{2(m^2 + mn + n^2)}$, where m, n are whole numbers [51]. We only considered twist angles that resulted in reasonably sized commensurate supercells with the number of atoms in unit-cell fewer than 350. Figures 2(a) and 2(b) show relaxed crystal structures and Moiré patterns in $\theta = 21.79^\circ$ tb-MnPSe_3 and tb-MnSe [see Moiré BZ in Fig. 2(c)]. Different pos-

¹ Therefore, we use “ $[C_2||\mathcal{P}]$ ” and “ $\mathcal{PT}$ ” interchangeably.

sible interlayer and intralayer magnetic couplings $\uparrow\uparrow\uparrow$, $\uparrow\downarrow\downarrow$, $\uparrow\downarrow\uparrow$, and $\uparrow\uparrow\downarrow$ were considered to determine the preferred magnetic ordering [here, up and down arrows represent the relative magnetic moment direction on Mn atoms]. The most stable magnetic structure is $\uparrow\downarrow\uparrow\downarrow$, where magnetic order is intralayer and interlayer AFM [Figs. 2(a) and 2(b)]. Twist angle leads to small variation in the magnitude of local magnetic moments from 4.441 to 4.448 μ_B/Mn in tb-MnSe. The tb-MnPSe₃ and tb-MnSe are antiferromagnetic with opposite spin sublattices connected through the rotation symmetries ($[C_2||C_{2[010]}]$ and $[C_2||C_{2[-120]}]$, respectively) with net zero magnetization [Figs. 2(a) and 2(b)]. In addition to Mn atoms, nonmagnetic ligands also contribute to $\mathcal{PT}$ symmetry breaking in tb-MnPSe₃ and tb-MnSe.

Firstly, we compute the spin-polarized band structures of tb-MnPSe₃ and tb-MnSe along the high-symmetry paths (HSPs) [Figs. 2(d),(e)]. The bands are spin degenerate along HSPs due to special symmetries arising at arbitrary k -point on HSP. For instance, $[C_2||C_{2[010]}]$ in tb-MnPSe₃ transforms spin-up to spin-down state along the Γ - K path, enforcing degeneracy between them [see Sec. III of SM [48]]. However, this is not the case for any generic k -point. No symmetry transform transform spin-up to spin-down at generic k -point, leading to the lifting of Kramers degeneracy. Therefore, the full BZ analysis of spin splitting is required. We plot spin splitting energy $\delta E [=E_{\uparrow}(k) - E_{\downarrow}(k)]$ of valence bands in tb-MnSe as a function of k [Fig. 3(a)]. The δE is invariant under real-space inversion $[\delta E(k) = \delta E(-k)]$ due to spin-only symmetry $[\bar{C}_2||T]$, which transforms energy eigenstates $[\bar{C}_2||T]E(k, \sigma) = E(-k, \sigma)$. Therefore, leading to 6-fold symmetric ($[E||C_6]$) planar i -wave spin-momentum coupling, which is different from the 3-fold symmetry of SOC-induced δE observed in well-known monolayer MoS₂ [52]. Similar patterns are also observed for δE of CB in tb-MnSe and VB in tb-MnPSe₃ (see Sect. III in SM). The maximum NRSS is observed at the orthocenter (H/H') of the triangle formed by Γ , M , and K_1/K'_1 points. Maximum splitting observed is 20.4, 4.2, and 5.1 meV for VB of tb-MnSe, CB of tb-MnSe, and VB of tb-MnPSe₃, respectively. Maximum δE is smaller than well-known bulk antiferromagnets, i.e., MnF₂ [21], Fe₂TeO₆ [31], and LaMnO₃ [23]. The δE observed in CB of tb-MnPSe₃ is negligible and beyond the accuracy of our calculations.

To understand the nature of NRSS, we plot band structures along both K_1 - K_c - K_2 and M_1 - M_c - M_2 directions [Figs. 3(b)-(e)]. Interestingly, linear NRSS is observed around K_c and M_c for VB and CB of tb-MnSe and VB of tb-MnPSe₃. The spin splittings exhibit contrasting characteristics at the H and H' points, featuring distinct valleys and maximum strength, suggesting the potential for valleytronics applications in twisted bilayers of antiferromagnets [53, 54]. Note that the spin splittings around the Γ point, along the Γ - H/H' direction, exhibit cubic characteristics, which result in their being relatively small and, as such, are excluded from the cur-

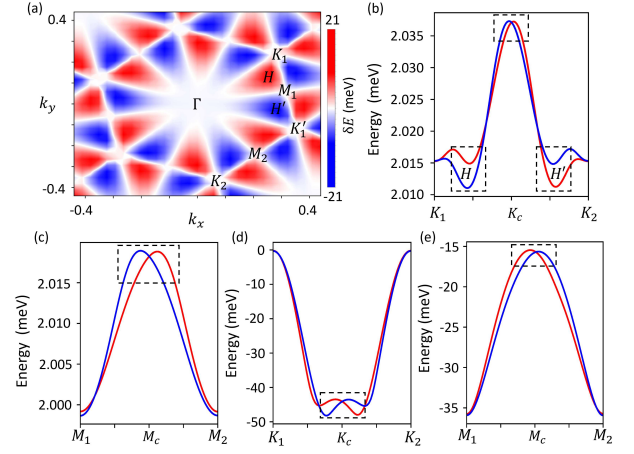


FIG. 3. (a) Spin splitting energy $[\delta E = E_{\uparrow}(k) - E_{\downarrow}(k)]$ distribution of valence band in 21.79° tb-MnSe. The units of k_x and k_y are $\text{\AA}^{-1}$. Conduction bands of tb-MnSe along the (b) K_1 - K_c - K_2 and (c) M_1 - M_c - M_2 paths [see Fig 2(c) for paths]. Valence bands of tb-MnPSe₃ along the (d) K_1 - K_c - K_2 and (e) M_1 - M_c - M_2 paths. The red and blue curves denote spin-up and spin-down bands, respectively. Black dashed squares represent prominent spin splittings. Fermi energy is set to valence band maximum.

rent discussion [55].

Spin splittings around M_c and K_c points are further analyzed using the symmetry-based model Hamiltonian, deduced using the “method of invariants” [55, 58]. The symmetry element (besides identity) of M_c/K_c point is $[C_2||C_{2[010]}]$ and $[C_2||C_{2[-120]}]$ for tb-MnPSe₃ and tb-MnSe, respectively [see Figs. 2(a) and 2(b)]. The symmetry invariant terms include $\alpha q_y \sigma_z$ and $q_y q_i^2 \sigma_z$ ($i = x', y'$), where $q = k - M_c/K_c$ are the momenta measured from M_c/K_c [see Sec. III of SM [48] for notation, derivation, and discussion]. Therefore, splitting is absent

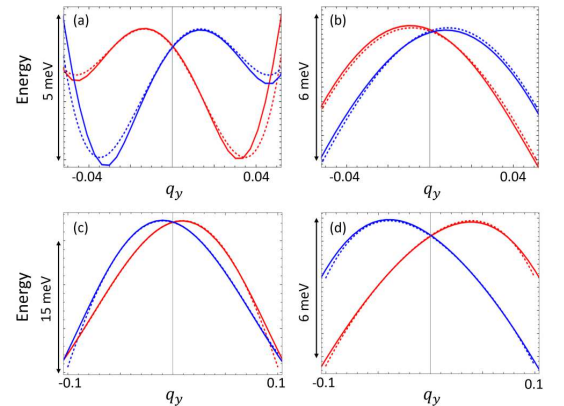


FIG. 4. Band structures of tb-MnPSe₃ around (a) K_c and (b) M_c along q_y direction. (c) and (d) are counterparts of (a) and (b), respectively, obtained for tb-MnSe. The solid and dotted lines are band structures obtained by DFT and the model described by Eq. 1, respectively.

TABLE I. Classification of 2D materials based on the MSG type, magnetic order and their impact on the NRSS. The relevant spin-group symmetry is also indicated in the case of spin-degeneracy at generic k .

Spin splitting prototype	Monolayer			Twisted bilayer	Examples
	Magnetic order	MSG type	NRSS at generic k	NRSS at generic k	
SST-1	Nonmagnetic	II	$\times([C_2 E])$	$\times([C_2 E])$	MoS ₂ [56], PtSe ₂ [57]
SST-2	Ferromagnetic	I/III	$\checkmark$	$\checkmark$	NiCl ₂ , CrI ₃ , CrN, CrSBr [32]
SST-3	Antiferromagnetic	III	$\times([C_2 \mathcal{P}]/[C_2 \mathcal{M}_z])$	$\checkmark$	MnPSe ₃ , MnSe [This work]
SST-4	Altermagnetic	I/III	$\checkmark$	-	
SST-5	Antiferromagnetic	IV	$\times([C_2 \tau])$	-	

along the $q_{x'}$ (K_c - K'_1 and M_c - K'_1) direction, whereas it is present along the $q_{y'}$ direction (K_c - $K_{1/2}$ and M_c - $M_{1/2}$). To understand the NRSS along the $q_{y'}$ direction, it is possible to write an effective Hamiltonian (H_{eff}), up to third-order in k :

$$H_{eff} = \alpha q_{y'} \sigma_z + \eta q_{y'}^3 \sigma_z \quad (1)$$

α and η are the constants determining the strength of NRSS. The primary linear term in Eq. 1 leads to the linear splitting of spin-up and spin-down energy bands around M_c and K_c points, similar to the linearly split bands by SOC-induced Rashba and Dresselhaus effects. Note that spin splitting in Eq. 1 originates from altermagnetic ordering and is completely nonrelativistic. On the other hand, the Rashba-Dresselhaus effect is induced by the spin-orbit field originating from noncentrosymmetric sites and is of relativistic origin. We fit the energy levels around M_c and K_c along the $q_{y'}$ direction to obtain spin-splitting parameters. The fits are obtained by minimization of the summation, $S = \sum_{i=1}^2 \sum_q f(q) |\text{Det}[H_{eff}(q) - E^i(q)I]|^2$ over the i^{th} energy eigenvalues $[E^i(q)]$ as training sets. We have also included a weight function $f(q)$ with normal distribution to get a better fit near the spin-degenerate point and avoid overfitting. The obtained fits to the DFT energy levels of tb-MnPSe₃ and tb-MnSe are shown in Figs. 4(a)-4(d). The Hamiltonian in Eq 1 with $\alpha=58.6$ meVÅ and $\eta=34.2$ eVÅ³ provide the best fit to the VBs of tb-MnPSe₃ around the K_c point [Fig. 4(a)]. Whereas $\alpha=39.8$ meVÅ and $\eta=3.5$ eVÅ³ are observed for VBs of tb-MnPSe₃ around the M_c point, respectively [Fig. 4(b)]. Similarly, linear splitting strength of 35.4 and 60.7 meVÅ is observed in CBs of tb-MnSe around the K_c and M_c points, respectively [Figs. 4(c)-3(d)]. The NRSS is comparable to those experimentally reported in the literature (e.g., 10 meVÅ for KTaO₃ [59], 4.3 meVÅ for LaAlO₃/SrTiO₃ interface [60], ~70 meVÅ in InGaAs/InAlAs interface [61], and 77 meVÅ for MoS₂ monolayer [62]). The growing field of twistronics makes NRSS observed in tb-MnPSe₃ and tb-MnSe experimentally accessible.

The 2D magnetic materials can be classified into five prototypes depending on the magnetic order, MSG, and whether NRSS is absent or present in a monolayer [Table I]. Spin degeneracy in nonmagnetic materials (SST-1) is enforced by $[C_2||E]$ and remains preserved under twisting operations. In contrast, FM materials (SST-2) show NRSS in both monolayer limits and two layers

stacked antiferromagnetically with a twist [32]. Altermagnetic materials (SST-4) have opposite-spin sublattices connected through mirror-rotation symmetries with opposite-spin electronic states separated in the momentum space. MSG type-IV always has AFM order with $[C_2||\tau]$ symmetry (SST-5) and necessitates SOC to induce spin splitting [23]. 2D AFM materials with MSG type-III containing $[C_2||\mathcal{P}]$ ($\mathcal{PT}$) or $[C_2||\mathcal{M}_z]$ (SST-3) are unique, as NRSS is absent in the monolayer and presented in twisted bilayer. Therefore, the twisting operation generates splittings in SST-3 type materials, the most common magnetic ordering found in nature.

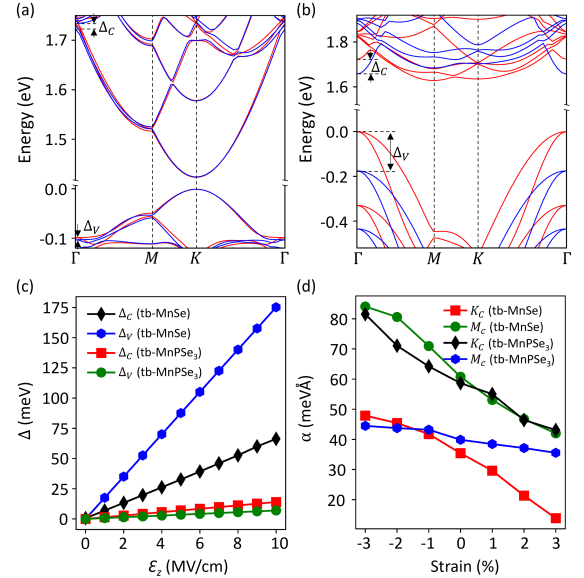


FIG. 5. Band structures of 21.79° (a) tb-MnPSe₃ and (b) tb-MnSe in the presence of the out-of-plane electric field (E_z) of strength 10 MV/cm. (c) The Zeeman spin splittings in the CB (Δ_C) and VB (Δ_V) of 21.79° tb-MnPSe₃ and tb-MnSe at Γ point as a function of E_z . (d) The variation in α (see Eq. 1) as a function of biaxial strain for 21.79° tb-MnPSe₃ and tb-MnSe.

Although controlling crystal symmetries in bulk materials is challenging, it has been shown that gating can effectively break the symmetries in 2D materials, including twisted bilayers [63–65]. In the following, we apply an out-of-plane electric field (E_z) to the tb-MnPSe₃ and tb-MnSe in DFT simulations self-consistently using the ap-

proach introduced by Neugebauer and Scheffler [66]. The electric field creates not only polarization but also magnetization by breaking opposite spin-sublattice transformation through the magnetoelectric coupling [67]. The Zeeman-like Hamiltonian under $\mathcal{E}_z$ are given by [31], $\hat{H}_Z = \lambda \mathcal{E}_z \sigma_z$, where λ is coefficient determining coupling strength. In the presence of $\mathcal{E}_z$, the spin degenerate levels at the high symmetry points (Γ , M , and K) and along HSPs will be split into two sublevels, $E_+ = \lambda \mathcal{E}_z$ and $E_- = -\lambda \mathcal{E}_z$ [Figs. 5(a)-5(c)]. We observe that the splitting induced by $\mathcal{E}_z$ in tb-MnPSe₃ and tb-MnSe exhibits markedly distinct characteristics. Specifically, an electric field $\mathcal{E}_z$ with a strength of 10 MV/cm results in nearly negligible splitting at the Γ point for tb-MnPSe₃, suggesting a small λ [Figs. 5(a), 5(c)]. In contrast, for tb-MnSe, the Γ point experiences a significantly larger Zeeman-type splitting (~ 175 meV) induced by an electric field $\mathcal{E}_z$ of 10 MV/cm [Figs. 5(b), 5(c)]. This disparity can be explained through structural analysis: in tb-MnPSe₃, Mn atoms with opposite magnetic moments lie within the same z -plane, while in tb-MnSe, they are situated in different z -planes, thus supporting magnetoelectric coupling when an electric field is applied along the z -direction. On the contrary, when we compare the Zeeman splittings induced in the CB and VB of tb-MnSe at the Γ point [Fig. 5(b)], it becomes evident that the splitting in the VB is significantly greater in magnitude compared to that in the CB. This pronounced splitting in the VB of tb-MnSe can be attributed to the in-plane orbitals, which have wave functions segregated on different z -planes and, as a result, are more susceptible to the $\mathcal{E}_z$. In addition, tunability in electronic states can be achieved by the strain engineering of 2D materials [68]. The in-plane biaxial strain preserves the crystal symmetry, thus creating no additional splittings. However, the strength of NRSS (α) around K_c/M_c for twisted bilayers are modified under biaxial in-plane strain [Fig. 5(d)]. α increases with compressive strain and increases with tensile strain, providing exceptional tunability.

Similar effects were also investigated for other twist angles, including 9.43°, 13.17°, 27.79°, 32.20°, 38.21°, and 42.10° (see Sect. IV of SM [48]). The δE also depends upon the dispersiveness of energy bands, where δE increases with increasing band dispersion. The linear NRSS is more prominent for the twist angles around 30°, as the structure deviates from the $\mathcal{PT}$ -symmetric ($\theta = 0^\circ, 60^\circ$) counterparts by the highest amount. In addition, the strength of splitting is the same for twist angles θ and $60^\circ - \theta$ (see Sect. V of SM [48]). MnPSe₃

and MnSe contain relatively lighter elements with negligible SOC effects (see Sect. VI of SM [48]). The Zeeman splitting observed in bilayer MnSe with a twist angle of $\theta = 0^\circ$ is ~ 180 meV under 10 MV/cm of the vertical electric field [30], nearly similar to 21.79° tb-MnSe of ~ 175 meV with the same electric field. Similarly, the Zeeman effect in 0° tb-MnPSe₃ is negligible [29], like 21.79° tb-MnPSe₃. Therefore, the order of Zeeman spin splitting depends much on how opposite spin-sublattices are arranged in the monolayer concerning the electric field and has less to do with the twist angle. Note that the models in this study include only spin degrees of freedom, thus revealing spin splitting qualitatively. For quantitative analysis, other degrees of freedom (i.e., orbital and sublattice) through first-principles or multiband tight-binding model calculations need to be included.

To summarize, we have shown that NRSS can be induced in 2D $\mathcal{PT}$ -symmetric antiferromagnets by taking bilayers with a relative twist. By first-principles calculations and symmetry analysis, we further predict spin-moment coupling in 21.79° tb-MnPSe₃ and tb-MnSe that accommodate linear NRSS as large as ~ 90 meVÅ. The lateral electric field split otherwise spin degenerate bands along the HSPs through magnetoelectric coupling, with more prominent effects in tb-MnSe. In addition, NRSSs are tunable using the biaxial strain. The measurement of these spin splittings can be conducted through well-established optical [29] and electrical transport [69] techniques commonly used in the field of spintronics. Employing antiferromagnets featuring spin-split bands as described in the present study may obviate the necessity for a heavy-metal layer, given that the current AFM mechanism yields a substantial magnitude of spin-moment splitting, even with lighter elements. Moreover, the low-Z antiferromagnets with even larger NRSSs can be predicted by the inverse design approach with desired functionality [70]. In addition, NRSS in Moiré-induced flat bands ($\theta \lesssim 3^\circ$) can be an interesting avenue to prospect. We aspire to broaden the pool of available materials and enrich the field of AFM semiconductor spintronics [1, 2] through the complete realization of original devices.

Acknowledgments.—S.S. acknowledges CSIR, India, for the senior research fellowship [grant no. 09/086(1432)/2019-EMR-I]. S. B. acknowledges financial support from SERB under a core research grant (grant no. CRG/2019/000647) to set up High-Performance Computing (HPC) facility “Veena” at IIT Delhi for computational resources.

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