Extremely flat band in antiferroelectric bilayer $\alpha$-In$_2$Se$_3$ with large twist-angle

C F Li, W J Zhai, Y Q Li, Y S Tang, J H Zhang, P Z Chen, G Z Zhou, X M Cui, L Lin, Z B Yan, K H Huang, X P Jiang and J-M Liu

1 Laboratory of Solid State Microstructures, Nanjing University, Nanjing 210093, People’s Republic of China
2 School of Materials Sciences, Jingdezhen Ceramic Institute, Jingdezhen, People’s Republic of China
3 Institute for Advanced Materials, South China Normal University, Guangzhou, People’s Republic of China

* Author to whom any correspondence should be addressed.

E-mail: zbyan@nju.edu.cn

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Abstract

The moiré-pattern with slightly interlayer-twisted bilayer two-dimensional (2D) materials has recently been receiving substantial attention. One of the major characters for these intriguing structures is the appearance of low-energy ultra-flat bands and thus a package of new physics associated with strong electron correlation emerges. However, such new physics may become vague unless the twist-angle $\theta$ is sufficiently small such as $\theta \sim 1^\circ$, making practical applications and control-flexibility hard to handle. In this work, we explore the possible flat-band moiré physics in recently concerned 2D bilayer $\alpha$-In$_2$Se$_3$ which is antiferroelectric with sublayer out-of-plane (OP) polarizations, addressing the potential role of polarization-bound charges in modulation of electron–electron correlation and interlayer hybridization, based on the extensive first-principles calculations. On one hand, it is found that the low energy band becomes extremely flat in the bilayer $\alpha$-In$_2$Se$_3$ moiré-pattern with relatively large twist-angle, e.g. $\theta = 13.17^\circ$, which is more easily accessible experimentally. On the other hand, the impact of the sublayer OP polarizations on the band structure is asymmetric, and the flattening effect is much more remarkable for the end-to-end polarization alignment but weaker for the head-to-head alignment. This work thus opens a broad roadmap for technological access to artificial fabrication of novel moiré-patterned 2D materials by means of low-dimensional ferroelectricity.

1. Introduction

When two identical layers with periodic atomic structure are stacked with a small in-plane (IP) twist-angle $\theta$, e.g. the so-called magic angle $\theta \sim 1.1^\circ$ [1, 2], certain moiré pattern will be developed, constituting an emergent class of novel materials in the catalogue of highly concerned two-dimensional (2D) systems [1, 3–6]. Such a twisting can destroy the original lattice periodicity of bilayer structure [7, 8]. While this operation can be extended to multilayer systems where different layers may be independently twisted or stacked with a small period mismatch [9, 10], we focus on the bilayer systems and their twisted structures, without losing the generality. Roughly speaking, the periodic length scale of these newly developed moiré structures is proportional to $\theta^{-1}$ as $\theta$ is small [11, 12], thus likely generating rich emergent structures.

This strategy was recently employed in graphene and a number of 2D van der Waals (vdW) systems by means of amazing and challenging technical handling, and a number of emergent quantum phenomena have been reported [7, 13–16]. For such vdW systems, the interlayer coupling is usually weak, and likely a bilayer system would exhibit no much difference from a monolayer system. Nevertheless, if the two layers are inter-twisted for a twist-angle $\theta$, usually an ordered moiré pattern will be developed due to the broken short-range lattice periodicity [7, 8]. Consequently, some additional interlayer hybridization occurs, leading to renormalized low-energy electronic structure with relatively flat low-energy bands [17, 18]. For such flat bands, it is easy to imagine that the kinetic energy is small relative to the Coulomb energy (potential) and
the electronic states in the flat bands would be localized, driving the system into the so-called strongly correlated phases [19, 20].

Indeed, such 2D moiré patterns do bring about surprising electronic properties, including superconductivity and nontrivial topological states [15, 16, 20–24]. These emergent phenomena constitute the main ingredients of so-called moiré physics. While most of so far synthesized 2D vdW materials are weak in electronic correlation, this moiré physics expands substantially the scope of strongly correlated materials. As an example, the twisted bilayer graphene (TBG) is the most known system [7, 20–22]. It was predicted theoretically and then confirmed experimentally that a TBG system with a small $\theta \sim 1.1^\circ$, the so-called ‘magic angle’, does show an extremely isolated flat band near the Fermi energy and the typical energy scale for the entire bandwidth of this band is $w = 5–10$ meV only, an unbelievably small value indicating the ultra-flat band [1, 8, 20]. In this case, the Fermi velocity at the Dirac point likely drops to almost zero, enabling the Coulomb energy $U$ to greatly exceed the kinetic energy, even though the magnitude of $U$ could be small with respect to those typical correlated electron systems [21]. This relatively clear physics results in appearance of gate-tunable superconductivity and correlated insulating states in various TBG systems [16, 21, 22].

Nevertheless, the above highlighted moiré physics hints some drawbacks either. On one hand, not all of the moiré structures (called moiré superlattices) exhibit the flat-band characters and most of those amazing emergent phenomena may not occur unless the twist angle $\theta$ is very small [1, 6, 8, 25]. In other words, the property of a moiré system depends critically on the magnitude of angle $\theta$. For a TBG system, the first-principles calculations show that the low-energy flat band with $w < 20$ meV appears only as $\theta < 3^\circ$ and $w$ increases remarkably with $\theta$ [13]. This deficiency makes practical applications challenging in terms of technical handling of so small $\theta$ and so high $\theta$-sensitivity. On the other hand, it is understandable that the smaller the angle $\theta$, the periodicity of the moiré superlattice becomes larger. A simple estimation shows that the moiré supercell of twisted bilayer black phosphorus at $\theta = 3.6^\circ$ contains more than 2000 atoms [25]. This ultra-large moiré superlattice makes realistic devices technologically non-scalable and electronic structure computations/designs either inaccessible.

It becomes clear that additional interlayer-twisted 2D materials should be searched for, and they can access the ultra-flat band at large $\theta$ or the bandwidth is insensitive to the magnitude of $\theta$. In other words, additional mechanism that can extend the ‘magic angle’ to large angle range is needed. While various strategies may be employed from different viewpoints, one comes back to the Coulomb energy $U$ that can be enhanced via some different schemes. An immediate motivation comes from the recently discovered antiferroelectric (AFE) bilayer systems such as bilayer $\alpha$-In$_2$Se$_3$ [26–29]. This motivation comes from the simple fact that electric polarization associated ferroelectric (FE) and AFE systems carries bound charges. As a model bilayer in which the two sublayers are both FE with an out-of-plane (OP) polarization, this ultra-large moiré superlattice usually exhibits low lattice symmetry [7, 8]. This hints some connection between the moiré pattern and polar symmetry which favors ferroelectricity in some cases. In fact, the interlayer twisting does reduce the original symmetry of bilayer structure. At least, it destroys easily the mirror and inverse symmetries, likely leading to polar moiré superlattice. Recent experiments did unveil switchable ferroelectricity in Bernal-stacked bilayer graphene sandwiched by two boron nitride layers, suggesting that the moiré superlattice offers a roadmap for modulating or generating 2D ferroelectricity [30]. In this framework, FE 2D insulators, semiconductors, or even metals may be possible by twisting those non-FE bilayer structures.

Interestingly, it was indeed confirmed that the 2D vdW $\alpha$-In$_2$Se$_3$ monolayer, bilayer and multilayer, all have simple crystal structures and exhibit intrinsically OP polarization for each layer/sublayer [27, 28]. In particular, the bilayer $\alpha$-In$_2$Se$_3$ in its energy-lowest structural mode accommodates the AFE state with antiparallel-stacked sublayer OP polarizations [29, 31]. While the sublayers are polar, the bilayer belongs to the $P$-3$m1$ nonpolar space group (No. 164) and shows AFE order where the two sublayer polarizations are either head-to-head or end-to-end aligned. Consequently, the bound charges must accumulate inside the interlayer and additional Coulomb energy $\Delta U$, depending on the head-to-head or end-to-end alignment, is expected. As a side-effect, this $\Delta U$ may allow a large twist-angle at which the moiré physics maintains remarkable yet.

In this article, we intend to investigate the emergent moiré physics of bilayer $\alpha$-In$_2$Se$_3$ in the large $\theta$-range, using the first-principles calculations. The major attention is paid to the electronic structure evolution associated with electric polarization and twist-angle, taking the high-temperature paraelectric structure (bilayer $\beta$-In$_2$Se$_3$) as the reference. It is demonstrated that the AFE state does contribute to the band flattening remarkably. On one hand, on the other hand such flat band effect remains remarkable at quite large twist-angle (e.g. $\theta = 13.17^\circ$ and $9.43^\circ$). Given the fact that electric polarization can be switched
upon external stimuli, the emergent moiré physics in such FE/AFE 2D materials would be highly interested for both fundamental physics and application potentials.

2. Twisted structural models and computational method

This work mainly relies on the *ab initio* calculations to investigate the electronic structure of IP twisted bilayer α-In₂Se₃ with various alignments of the two sublayer OP polarizations. As discussed earlier, the moiré superlattice at small $\theta$ would be extremely big for practical calculations, while the moiré pattern would be smaller and include much less atoms at large angle. For example, the unit cells at $\theta = 13.17^\circ$ and $9.43^\circ$ for high-temperature nonpolar β-In₂Se₃ phase contain 190 atoms and 370 atoms respectively, far smaller than thousands of atoms at $\theta < 3^\circ$. We thus focus mainly on the systems in the large $\theta$ range.

Furthermore, from figure 1 it is clear that there exist multiple rotation axes (each call a twist-axis) for twisting the two sublayers in order to form a moiré superlattice. The difference among these as-generated moiré superlattices remains less explored. We choose two representative twist-axes. By taking one twist-axis, the as-generated moiré superlattice belongs to the P3 polar space group (No. 143), and by taking the other one, a moiré superlattice belonging to the P312 nonpolar space group (No. 149) will be obtained. Therefore, a proper choice of twist-axes would be concerned, noting that the non-twisted bilayer α-In₂Se₃ is nonpolar in the AFE ground state (space group P-3m1).

2.1. Moiré superlattice of ground state bilayers α-In₂Se₃

We start from a monolayer α-In₂Se₃, which belongs to the P3m1 polar space group (No. 156). It has a quintuple layer (QL) containing five triangular atomic units stacked in the sequence of Se–In–Se–In–Se via covalent bonding, as shown in figure 1(a), side-view. Clearly, the middle Se sublayer at the asymmetric position (dashed red line AP) spontaneously breaks the center-symmetry, resulting in the OP polarization $P_{op}$, as marked by the arrow [27]. As a comparison, a monolayer β-In₂Se₃ phase has the similar structure, as shown in figure 1(b), the Se atoms occupy the symmetric positions (dashed green line SP), satisfying the P-3m1 nonpolar space group (No. 164).

Earlier work revealed that the bilayer α-In₂Se₃ has the AFE ground state with also the P-3m1 space group (No. 164), identical to the monolayer β-In₂Se₃ [31]. The fully relaxed bilayer α-In₂Se₃ with the end-to-end polarization alignment consists of two α-In₂Se₃ QLs with opposite $P_{op}$, as shown in figure 1(c), side view. The trigonal crystal structure has the IP threefold and mirror symmetries and a global symmetry
center. We choose two representative twist-axes. One is along the [001] by passing across the In/Se atoms (denoted as [001]$_{In}$) and the other one is along the [001] by only passing across the Se atoms (denoted as [001]$_{Se}$). The two twist-axes both have the threefold-axis of symmetry (as see from the top view, figure 1(c)). The two axes and one global symmetry center (red-cross dot) are also denoted in figure 1(c), side view.

In particular, as seen in the top view of figure 1(c), the plane marked by the orange line contains no twofold axis and the plane marked by the violet line contains one twofold axis parallel to the ab plane. The twofold axis and the global center link the atoms in the top layer with the corresponding atoms in the bottom layer one by one, forming the OP AFE structure. Consequently, we rotate the top sublayer around the [001]$_{In}$ and [001]$_{Se}$ axes respectively, by angle $\theta$ relative to the bottom layer. Eventually, a bilayer $\alpha$-In$_2$Se$_3$ moiré superlattice is developed.

Apparentely, such rotation operations break all the mirror symmetries. The rotation around [001]$_{In}$ breaks both the central inversion symmetry and the twofold symmetry, leading to the P3 polar space group (No. 143) with the [001]$_{P3}$ polar axis. The rotation around [001]$_{Se}$ breaks the central inversion symmetry but does not break the twofold symmetry, leading to the P312 nonpolar space group (No. 149). The two types of moiré patterns generated at angle $\theta = 13.17^\circ$ are shown in figures 2(a) and (d) respectively. One of new twofold axes is marked by the violet dash–dotted line in figure 2(d), for the P312 space group. We call these structures that exhibit the end-to-end polarization alignment as the AFE-1 structures (figures 1(c) and 2(a) and (d)).

2.2. Moiré superlattice of non-ground state bilayers $\alpha$-In$_2$Se$_3$

While it is known that the bilayer $\alpha$-In$_2$Se$_3$ prefers the AFE ground state with the end-to-end OP polarization alignment (AFE-1 structure), as shown in figure 1(c), for the present motivation of study it is of high interest to check the consequence of the head-to-head OP polarization alignment in contrast to the end-to-end alignment. This head-to-head state is a non-ground AFE state and the fully relaxed structure is shown in figure 1(d), called the AFE-2 state.

In fact, this AFE-2 state can be formed by exchanging the top and bottom sublayers of the AFE-1 state. By running the identical procedure as the AFE-1 state, the as-generated moiré superlattices by twisting around the two axes ([001]$_{In}$ and [001]$_{Se}$) respectively belong to the P3 polar space group (No. 143) and P312 nonpolar space group (No. 149), and are shown in figures 2(b) and (e). The two artificially assigned AFE-2 moiré patterns allow us to compare the calculated band structures with the AFE-1 ground states.

2.3. Moiré superlattice of bilayers $\beta$-In$_2$Se$_3$

Similarly, the fully relaxed bilayer $\beta$-In$_2$Se$_3$ with the P-3m1 space group (No. 164) consists of two nonpolar $\beta$-In$_2$Se$_3$ QLs, as shown in figure 1(e). One can also choose two typical twist-axes: [001]$_{In}$ with the threefold-axis of symmetry and [001]$_{Se}$ with the threefold-axis of symmetry and one global symmetry...
Figure 3. (a)–(c) The calculated band structures of the relaxed AFE-1-α-P-3m1, PE-β-P-3m1, and AFE-2-α-P-3m1 respectively. (d)–(f) The amplified bands near the Fermi level in (a)–(c) respectively. The band gap $\Delta = 0.69$ eV, 0.29 eV and 0.62 eV, and the bandwidths of the top valence bands (red lines) $w = 0.20$ eV, 0.38 eV and 0.45 eV, respectively.

center (red cross), as shown in figure 1(e). In particular, as seen in the top view of figure 1(e), the plane marked by the orange line contains no twofold axis and the plane marked by the violet line contains one twofold axis parallel to the $ab$ plane. We can take the [001]$_{In}$ and [001]$_{Se}$ axes and rotate the top layer by angle $\theta$ relative to the bottom layer, leading to a bilayer $\beta$-In$_2$Se$_3$ moiré superlattice. The two types of rotation operations break all the mirror symmetries. The rotation around the [001]$_{In}$ axis breaks both central inversion symmetry and the twofold symmetry, leading to the $P$3 polar space group (No. 143) in which the polar axis is [001], noting that the structure is paraelectric in spite of being polar. The rotation around the [001]$_{Se}$ twist-axis breaks the central inversion symmetry but not the twofold symmetry, leading to the $P312$ nonpolar space group (No. 149). The two moiré patterns as generated at $\theta = 13.17^\circ$ are shown in figures 2(c) and (f) respectively, and one of new twofold axes marked by the violet dash–dotted line in the $P312$ space group is shown in figure 2(f). For the AFE-1 and AFE-2 bilayer $\alpha$-In$_2$Se$_3$ as well as non-polar bilayer $\beta$-In$_2$Se$_3$ moiré superlattices, each of them can be slide the two sublayers each other so that the interconversion between the $P$3 polar space group (No. 143) structure and $P312$ nonpolar space group (No. 149) structure occurs.

To this end, we have totally the six moiré patterns as shown in figure 2 for our calculations. For convenience of expression, we name the ‘AFE-1 bilayer $\alpha$-In$_2$Se$_3$ moiré superlattice with the polar $P$3 space group’ as ‘AFE-1-α-P3’ structure, and other abbreviations are shown in figure 2 for reference (e.g. AFE-1-α-P312, AFE-2-α-P3, PE-β-P3 etc).

2.4. Details of calculations

Starting from each of the six units, we employ the DFT calculations based on the projected augmented wave (PAW) pseudopotentials as implemented in the Vienna $ab$ initio simulation package (VASP) [32–34]. The electron interactions are described using the PBE parametrization of the GGA [35]. The plane-wave cutoff is set to 400 eV, which is sufficient because it is already 1.5 times larger than the default values for In (239 eV) and Se (212 eV) in the PAW pseudo-potential. The $\Gamma$-centered Monkhorst–Pack $k$-point mesh is $12 \times 12 \times 1$ for the untwisted In$_2$Se$_3$ bilayer, as well as $3 \times 3 \times 1$ and $2 \times 2 \times 1$ for the moiré superlattices with $\theta = 13.17^\circ$ and $9.43^\circ$ respectively.

For the details of computation, we take the following procedure. First, for all the systems, the interlayer vdW interaction is considered via the Grimme’s DFT-D3 method [36]. Second, we take the standard
The calculated band structures of the moiré superlattices of initial AFE-1-α-P3, PE-β-P3, and AFE-2-α-P3 respectively, with twist angle $\theta = 13.17^\circ$. (d)-(f) The amplifying bands near the Fermi level in (a)-(c) respectively. The bandgaps $\Delta$ are 0.46 eV, 0.27 eV and 0.46 eV, and the bandwidths $w$ of the top valence bands (red lines) are 2.66 meV, 33.65 meV and 98.88 meV, respectively.

procedure and insert a vacuum space of more than 2.5 nm in thickness into periodically repeated bilayer lattice, noting that the $c$-axis lattice constant for these bilayers is fixed to be 4.5 nm. A relaxation of this $c$-axis constant is not allowed due to the insertion of vacuum space, referring to earlier reports [37] and the 2.5 nm thick or more vacuum space would be sufficient. Third, for the non-twisted structures, the lattice relaxations start from the structures shown in figures 1(c)–(e) the initial structures respectively, and the measured lattice constants of the bulk systems are used as the initial IP lattice constants ($a = b$) [38].

We construct the moiré superlattices following the scheme presented by Lopes dos Santos, Peres and Castro Neto [3], and starting from the relaxed but non-twisted bilayer lattices. Then we calculate the band structures of the constructed moiré superlattices by proper interlayer twisting. It should be mentioned that additional structural relaxation may be needed after such twisting operation, while such a structural relaxation is very time-consuming due to the super-large supercells associated with the moiré superlattices. As a well-accepted compromise, we fully optimize the IP lattice constants and atomic positions of the AFE-1-α-P3 and AFE-1-α-P312 structures at twist-angle $\theta = 13.17^\circ$ until the Hellman–Feynman forces are converged down to 10.0 meV Å$^{-1}$ and below. Then we compare the calculated band structures of the non-relaxed and fully relaxed systems, and it is found that the optimization-induced difference is negligible for the bands near the Fermi level, although the deep-level bands show some variation.

Schematic illustrations of crystal structures and the isosurfaces of charge density distributions are drawn using the program VESTA [39].
3. Results and discussion

3.1. The band structures of untwisted bilayers

For reference and comparison, we first calculate the band structures of the three bilayer structures in the non-twisted case (i.e. $\theta = 0$). They are the AFE-1-$\alpha$-P$_3$12, PE-$\beta$-P$_3$12, and AFE-2-$\alpha$-P$_3$12 respectively, with the same non-polar P-$3m1$ space group. The results are plotted in figures 3(a)–(c) respectively. It is seen that for all the three bilayers, the obtained band structures exhibit relatively remarkable band dispersions with wide bandwidths $w$, as shown in figures 3(d)–(f) respectively by amplifying the bands near the Fermi level. For a quantitative estimation, the obtained bandgap is $\Delta =$ 0.69 eV, 0.29 eV, and 0.62 eV respectively, consistent with earlier calculated results for the AFE-1 bilayer $\alpha$-In$_2$Se$_3$ and monolayer $\beta$-In$_2$Se$_3$, indicating that they are all narrow-gap insulators [29, 31, 40]. The top valence band that is the closest to the Fermi level, as indicated by the red line in figure 3, shows the bandwidth $w = 0.20$ eV, 0.38 eV, and 0.45 eV respectively. It is noted that relatively strong delocalization appears if $w > 0.1$ eV, implying that the electron correlation is not strong if any in these non-twisted bilayers.

Furthermore, it is interesting to compare quantitatively the bandwidths of the three systems. The AFE-1 bilayer $\alpha$-In$_2$Se$_3$ has the smallest $w = 0.20$ eV, noting that its two sublayer OP polarizations are end-to-end aligned, i.e. the interlayer bound charges are negative (electrons). The existence of these negative charges must enhance effectively the Coulomb energy $U$, i.e. the electron correlation, and thus flat the bands to some extent. For the AFE-2 bilayer $\alpha$-In$_2$Se$_3$, the situation is opposite and the electron correlation is suppressed to some extent, explaining why its bandwidth $w$ is the largest.

3.2. Band structures of moiré superlattices

Based on the preliminary results on non-twisted systems, now we can check the band structures of the as-generated moiré superlattices. Given the twist-angle $\theta = 13.17^\circ$, the band structures of the AFE-1-$\alpha$-P3, PE-$\beta$-P3, and AFE-2-$\alpha$-P3 moiré superlattices are plotted in figures 4(a)–(c) respectively, noting that the P3 space group (No. 143) is polar (the twist axis is the [001]$_{In}$). While details of the band structure for each moiré superlattice seem to be complicated, we discuss the difference of overall features between these.
superlattices. It is seen that the interlayer twisting of each system does generate strong band-flattening consequence, at least for the bands near the Fermi level. This flattening effect becomes particularly strong for the top valence band of AFE-1-α-P3 and its bandwidth shrinks down to a few meV from 200 meV. For the PE-β-P3, the band flattening effect is also remarkable but weaker than the case of AFE-1-α-P3 while the flattening effect of the AFE-2-α-P3 is the weakest, as re-shown in figures 4(d)–(f) respectively by amplifying the bands (red lines) near the Fermi level.

Besides the AFE-1-α-P3, PE-β-P3, and AFE-2-α-P3 moiré superlattices, we also investigate the band flattening effect of the AFE-1-α-P312, PE-β-P312, and AFE-2-α-P312 moiré superlattices which are obtained by twisting around the [001]Se axis, noting that the P312 space group is nonpolar. The calculated band structures at θ = 13.17° are plotted in figures 5(a)–(c) respectively. It is seen clearly that the band flattening effect is also very remarkable, as re-shown in figures 5(d)–(f) respectively by amplifying the bands near the Fermi level, even though these superlattices are nonpolar.

A comparison of the band structures shown in figures 4 and 5 with those shown in figure 3 makes clear at least three issues. First, the interlayer twisting by an angle even as large as θ = 13.17° is sufficient to seriously flatten the band near the Fermi level remarkably, resulting in the bandwidth shrinking down to a few meV from hundreds of meV. It can be expected that this flattening effect would be even more remarkable if the twist-angle is smaller. Second, the twisting operations around the [001]In axis and [001]Se axis show no large difference in terms of the band flattening. Third and more importantly, it is found that the band flattening effect also depends on the sublayer OP polarization alignment or FE behavior of the system under investigation. The strongest band-flattening effect appears in the AFE-1-α-P3 and AFE-1-α-P312 moiré superlattices where the two sublayer polarizations are end-to-end aligned, while the weakest effect appears in the AFE-2-α-P3 and AFE-2-α-P312 moiré superlattices where the two sublayer polarizations are head-to-head aligned.

To this stage, all of our discussion go to the top valence band near the Fermi level. It is found that the flattening effect of conduction band above the Fermi level is much weaker against the interlayer twisting and electric polarization, although the conduction bands are also slightly flattened in comparison with the non-twisted bilayers. The bandwidths remain to be \( w \sim 0.5 \text{ eV} \), remarkably larger than the seriously flattened valence band near the Fermi level. This effect may be understood from the fact that there actually
Figure 7. The calculated band structures including the SOC effect for all the six initial moiré superlattices with twist angle $\theta = 13.17^\circ$. (a) AFE-1-$\alpha$-$P3$. (b) PE-$\beta$-$P3$. (c) AFE-2-$\alpha$-$P312$. (d) AFE-1-$\alpha$-$P312$. (e) PE-$\beta$-$P312$. (f) AFE-2-$\alpha$-$P312$. Comparing with the valence band near the Fermi level shown in figures 4 and 5, for each of all the six moiré superlattices, the top valence band is split into two sub-bands (red and blue lines respectively), and the bandwidths $w$ of the two sub-bands are labelled separately.

is no charge occupation in these conduction bands and thus no charges bound with the electric polarizations.

3.3. Reduced band gap in connection with reduced bandwidth
It should be mentioned that the band flattening effect in response to the interlayer twisting is accompanied with an additional consequence, i.e. the band gap $\Delta$ also decreases with the decreasing $w$, an unusual effect. In common scenario, it is believed that the strong electron correlation would result in carrier localization and metal–insulator transition, thus lead to the enlarged band gap, i.e. $\Delta$ should increase with reduced bandwidth $w$.

To illustrate the calculated results, we present in figure 6 the band structures of the AFE-1-$\alpha$-$P$-3$m$1, polar $P$-3, and polar $P$-3 structures respectively. While the bandwidth is $\sim 0.2$ eV for the non-twisted AFE-1-$\alpha$-$P$-3$m$1 structure, its band gap is $\sim 0.7$ eV. They become $w = 2.66$ meV and $\Delta = 0.46$ eV for the twisted AFE-1-$\alpha$-$P$-3 moiré superlattice with $\theta = 13.17^\circ$ and $w = 0.36$ meV and $\Delta = 0.16$ eV for the twisted AFE-1-$\alpha$-$P$-3 moiré superlattice with $\theta = 9.43^\circ$. Here, a band gap as small as $\Delta = 0.16$ eV is sufficiently small that the structure becomes highly conductive, although the top valence band is almost free of dispersion, i.e. of strongly electron correlation. It can be expected that a further reduction of the twist-angle from 9.43$^\circ$, a metallic moiré superlattice would be reached due to the merged band gap. This seems to be an unusual state with strong electron correlation and FE/AFE order simultaneously.

It should be mentioned that such unusual concomitant reduction of bandwidth $w$ and band gap $\Delta$ is not strange, and earlier investigations revealed similar effect in other moiré superlattices with small twist-angle, although the underlying physics seems to be complicated without a simple scenario [6, 25]. Certainly, such a state may exhibit some specific properties deserved for further exploration.

3.4. The spin–orbit coupling effect
Until now, we focus on only the polarization dependence of the top valence band flattening effect. Certainly, the SOC effect should be considered since it is believed to have non-negligible influence on the band structure [29, 40]. While the In and Se atoms with compound In$_2$Se$_3$ are in the IIIA group of the fifth
Figure 8. Top and side view of the charge density distributions of the top valence bands (blue lines shown in figure 7) with inclusion of SOC for all the six initial moiré superlattices with twist angle $\theta = 13.17^\circ$. (a) AFE-1-$\alpha$-P3. (b) PE-$\beta$-P3. (c) AFE-2-$\alpha$-P3. (d) AFE-1-$\alpha$-P312. (e) PE-$\beta$-P312. (f) AFE-2-$\alpha$-P312. The yellow area shows the isosurface and the isosurface value is set at 0.001 e Å$^{-3}$.

Comparing the band structures without inclusion of the SOC shown in figures 4 and 5 with those with inclusion of the SOC effect shown in figure 7, we highlight several aspects of the SOC effect from the calculations while details of the underlying physics are beyond the scope of this work and will be investigated in future. First, the SOC does show influences on the band structure, and these influences are different depending on the different structures. Inclusion of the SOC enlarges the band gap for the PE-$\beta$ structures. However, the band gap is only weakly affected by the SOC for the AFE-1-$\alpha$ structures, while the influence seems to be complicated for the AFE-2-$\alpha$ structures. Second, for all the six moiré superlattices, the SOC inclusion leads to the splitting of top valence bands each into two sub-bands (as shown by the red and blue lines in the figure 7). Such splitting is remarkable for both the PE-$\beta$ and AFE-2-$\alpha$ structures, but much weaker for the AFE-1-$\alpha$ structures. Third, the very flat top valence bands (small bandwidths) for the AFE-1-$\alpha$ structures suggest the very strong electron–electron correlation. It is thus clear that the SOC effect becomes negligible when the electron–electron correlation is strong, while it becomes much more remarkable when the bandwidth is large or one says, the correlation is relatively weak.

The values of bandwidth for these structures with inclusion of the SOC effect are marked in figure 7 and their dependences of the SOC and electron–electron correlation are complicated. The details will be presented elsewhere in future after much more investigations are carried out.
3.5. Charge density distribution

Previous theoretical results had shown that one of the physical characteristics corresponding to the flat band is the localization of charge density distribution in real space [6, 25]. The charge density distribution in the presence of FE polarization may be different from those non-FE systems or systems with different polarizations. Thus we study the real space distributions of the charges taken from the top valence bands for all the six moiré superlattices with twist angle $\theta = 13.17^\circ$. By doing this, one needs to consider the influence of the SOC. The results presented below are from the calculations with SOC inclusion, noting that the results without inclusion of SOC show no remarkable difference.

The calculated charge density distributions of the top valence bands for all the six moiré superlattices are shown in figure 8 where the isosurface value is set at 0.001 e Å$^{-3}$ and the yellow area shows the isosurface in which the charge density is larger than the isosurface value. First for the AFE-1-$\alpha$ moiré superlattices, the charge density distributions are localized in both the IP and OP directions. The electrons are largely localized near the bilayer interface, obviously due to the end-to-end polarization alignment. Unfortunately, a comprehensive understanding of the IP charge localization remains to be an issue to date, and most likely this localization is the consequence of the strong electron–electron correlation. Second for the PE-1-$\alpha$ moiré superlattices, the charge density distributions of the top valence bands are also localized in the IP direction. Along the OP direction, no charge localization is seen due to the absence of FE polarization. Third for the AFE-2-$\alpha$ moiré superlattices, the charge density distributions of the top valence bands are IP homogeneous and localized in the OP direction only. This could be understood from the fact that the IP correlation is weak. For the OP distribution, it is also clear that the charges tend to be localized away from the bilayer interface due to the head-to-head polarization alignment along the OP direction.

The results indicate that the polarization indeed leads to additional charge density localization thus adjusts the band flattening effect. While there are other differences of the charge density distributions and the SOC effect has less remarkable on the charge density distributions, the details of discussion are beyond the scope of this work and will be presented elsewhere in future after much more investigations are carried out.

3.6. Discussion

To this end, two major facts with the In$_2$Se$_3$ moiré superlattices constructed by the interlayer twisting are: (1) the band flattening effect depends largely on the OP polarization alignment, i.e. the AFE-1-$\alpha$ or AFE-2-$\alpha$ structure; (2) the concomitant reduction of the bandwidth $w$ and band gap $\Delta$ with decreasing twisting angle. A comprehensive understanding of the underlying physics with these facts is challenging while a simplified and qualitative explanation may be possible.

For the polarization dependence of the band flattening effect, one sees in figure 9(a) the values of bandwidth $w$ without inclusion of the SOC for the six systems under our investigation at the fixed twist-angle $\theta = 13.17^\circ$. While all these twisted systems do show remarkable band flattening effects, the corresponding OP polarizations of the two sublayers for the AFE-1-$\alpha$ and AFE-2-$\alpha$ structures are drawn schematically in figures 9(b) and (d) respectively. It is seen that for the AFE-1-$\alpha$ structures, the end-to-end polarizations induce negative bound charges in-between the two sublayers, implying the additional electron accumulation there which is equivalent to the enhanced electron–electron correlation and thus the enhanced Coulomb energy $U$, as shown in figure 9(b). For the AFE-2-$\alpha$ structures, the head-to-head polarizations induce ‘positive’ bound charges in-between the two sublayers, implying the additional electron deficiency there which is equivalent to the suppressed electron–electron correlation and thus the reduced Coulomb energy $U$, as shown in figure 9(d). Naturally, the PE-1-$\alpha$ structures have no net bound charges in-between the two sublayers, and thus the band flattening effect is between the AFE-1-$\alpha$ and AFE-2-$\alpha$ structures.

Second, the concomitant reduction of $w$ and $\Delta$ with decreasing $\theta$ represents a surprising consequence upon the interlayer twisting. A possible explanation is that the serious flattening of the top valence band just below the Fermi level is equivalent to remarkably enhance the local Coulomb energy within this band, and thus an outlet is needed to reduce the accumulated electron density. One potential outlet is the conduction band to which electrons jump from the valence band. In order to realize such jump in compensation of the large Coulomb energy, the system will restructure its band structure by reducing the band gap $\Delta$ so that the electron jump from the flat valence band to the conduction band.

Finally, we illustrate the remarkable impacting of the electric polarization on the band flattening effect by comparing the data on bandwidth $w$ for a series of different 2D bilayer materials with moiré superlattices, and the results are plotted in figure 10 where the obtained bandwidths $w$ as a function of twist-angle $\theta$ are plotted for bilayer graphene [13], bilayer black phosphorus [25], bilayer boron nitride [6], and our bilayer $\alpha$-In$_2$Se$_3$ (AFE-1-$\alpha$-P3). It is convincingly demonstrated that our AFE bilayer $\alpha$-In$_2$Se$_3$ has the smallest bandwidth at the largest twist-angle. This superior performance makes the twisted AFE bilayer
Figure 9. (a) The summary of the bandwidth $w$ of the top valence band for the six moiré superlattices with the fixed $\theta = 13.17^\circ$. (b)–(d) Sketch drawings for explaining the band flattening effect that largely depends on the OP polarization alignment in the moiré superlattices of AFE-1-$\alpha$, PE-$\beta$, and AFE-2-$\alpha$ respectively.

Figure 10. The bandwidth $w$ of the top valence band as a function of twist-angle $\theta$ for the moiré superlattices of bilayer graphene, bilayer black phosphorus, bilayer boron nitride, and bilayer $\alpha$-In$_2$Se$_3$ (AFE-1-$\alpha$-P3) respectively.

$\alpha$-In$_2$Se$_3$ the best candidate for practical fabrication of moiré superlattice materials and devices, because the twist-angle is large enough for realistic twisting operation on one hand, and on the other hand FE polarization is utilized as a control parameter for practical applications. These advantages seem to be highly favored but yet access-challenging for other 2D bilayer systems.

4. Conclusion

In conclusion, we have investigated the band structures of the interlayer twisting induced moiré superlattices from bilayer $\alpha$-In$_2$Se$_3$ systems of the end-to-end OP polarization aligned AFE ground state (AFE-1-$\alpha$) and head-to-head OP polarization aligned AFE state (AFE-2-$\alpha$) in order to explore the band flattening effect in such 2D FE systems, taking the paraelectric bilayer $\beta$-In$_2$Se$_3$ structure (PE-$\beta$) as the reference. First, a detailed discussion on the interlayer twisting model has been presented and two
representative twisting axes are chosen for constructing the moiré superlattices with polar and non-polar space groups respectively. Subsequently, it has been revealed that for the two types of moiré superlattices, the twisting with angles as large as 13.17° are already sufficient to induce remarkable flattening effect on the top valence band near the Fermi level, and the bandwidth can be suppressed down to ~1.0 meV from hundreds of meV. This flattening effect can be further enhanced if the smaller twist-angle is chosen. Simultaneously, the band flattening effect is also accompanied with the reduced band gap which may be hundreds of meV. This flattening effect can be further enhanced if the smaller twist-angle is chosen.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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