Flat Bands in Twisted Bilayers of Two-Dimensional Polar Materials

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The existence of Bloch flat bands provides an facile pathway to realize strongly correlated phenomena in materials. Using density-functional theory and tight-binding approach, we show that the flat bands can form in twisted bilayer of hexagonal boron nitride (hBN). However, unlike the twisted graphene bilayer where a magic angle is needed to form the flat band, for the polar hBN, the flat bands can appear as long as the twisted angle is less than certain critical values. Our simulations reveal that the valence band maximum (conduction band minimum) states are predominantly resided in the regions of the moiré superlattice where the anion N (cation B) atoms in both layers are on top of each other. The preferential localization of these valence and conduction states originate from the chemical potential difference between N and B and is enhanced by the stacking effects of N and B in both layers, respectively, as demonstrated by an analysis of the energy level order of the hBN bilayers with different stacking patterns. When these states are spatially localized because regions with a specific stacking pattern are isolated for moiré superlattices at sufficient small twist angle, completely flat bands will form. This mechanism is applicable to other twisted bilayers of two-dimensional polar crystals.

In condensed matters, an unusual characteristic of Bloch electrons is the existence of flat bands. Being weakly dispersive, a flat band has a vanishingly small band width ($W$) and accordingly, high density of states, inducing strong Coulomb interactions ($U$) between electrons, with $U \gg W$. If the flat band is at the Fermi level, the kinetic energy of the electrons that is confined by $W$ is substantially suppressed to be much smaller than the Coulomb interaction. Thus, the associated system may exhibit pronounced correlation effects [1] as already seen in various exotic quantum states. These include superconductivity [2], ferromagnetism [3], Wigner crystal [4], and zero-magnetic field fractional quantum Hall effects (QHE) of Bloch states [5–9]. Since flat bands provide a route to accessing correlated electronic states, searching for new materials with flat bands is important and currently under active investigations.

Recent theoretical and experimental advances have shown that such flat bands could be obtained in twisted van der Waals (vdW) heterostructures assembled from atomically thin two-dimensional (2D) crystals. Due to the twist-induced misalignment between constituent layers, a twisted vdW heterostructures will develop complex lateral morphologies usually showing as a moiré pattern with periodicity much longer than the interatomic distance. This special moiré superlattice creates strong modulation on the electronic interlayer coupling, leading to interesting physics such as fractional QHE [10–12], gap opening [13], and moiré excitons [14–17]. Particularly, the electronic structure of twisted graphene bilayers (TGB) can be tailored to develop isolated flat bands at some magic angles [18–21]. Furthermore, it has also been shown that the value of magic angles relies on the interlayer coupling that can be tuned by varying the interlayer spacing with hydrostatic pressure [22, 23]. Experiments have shown that these flat bands are the key to achieve the correlated insulating and superconductive phases in graphene systems [24–26]. Nearly flat bands and possible correlated effects are also predicted for twisted bilayers of transition-metal dichalcogenides (TMDs) at specific twist angles [27, 28].

In this Letter, we investigate the electronic properties of bilayers and twisted bilayers of hexagonal boron nitride (hBN), a typical two-component 2D polar crystals constituting cation (B) and anion (N) atoms with quantunmechanical simulations. Our results reveal a new mechanism for the formation of Bloch flat bands other than the known mechanism for twisted graphene bilayer that needs a magic angle. We show that in the bilayer form of hBN, the band edge states are sensitive to the stacking patterns of cation atoms and anion atoms between layers. Namely, the valence band edge will shift upwards if the anion atoms in one layer are on the top of those in the other layer; while the conduction band edge will shift downwards if the cation atoms in one layer are on the top of those in the other layer. Accordingly, in the twisted bilayer form of hBN, the valence band maximum (VBM) states are confined to the regions where anion atoms in both layers are on top of each other; while the conduction band minimum (CBM) states are confined to the regions where cation atoms in both layers are on top of each other. Because regions with a specific stacking pattern are spatially separated in the moiré superlattice, especially at sufficiently small twist angle, the VBM and CBM states are actually localized, forming the expected flat bands.

Our simulations are carried out with a density-functional based tight-binding (DFTB) theory with a local orbital basis [29, 30], which has been widely applied successfully to various forms of hBN including sheets [31] and nanotubes [32]. A direct comparison of the electronic structures calculated by DFTB and by first-principles ap-
To illustrate the stated idea to find flat bands in 2D, it is appropriate to use twisted hexagonal boron nitride (hBN) bilayer as a representative system because hBN is a typical 2D polar crystal and its electronic property of monolayer hBN has been well investigated experimentally and theoretically. Different from graphene, monolayer hBN adopts an energy gap due to the symmetry breaking of $A - B$ sublattice of the hexagon lattice, where the large polarization between cation $B$ and anion $N$ atoms enlarges the energy gap, driving hBN into an insulator. The ground state of hBN bilayer assumes an $AB$ stacking where B atoms of one layer are on top of N atoms of the other layer and vice versa, Fig. 1(a)[left]. If twisted for a small angle $\theta$, the resulting moiré superlattice as shown in Fig. 1(b) can also develop two additional Bernal stackings, having N atoms (B atoms) of one layer on the top of N atoms (B atoms) of the other layer, labeled as $B^{NN}$ ($B^{BB}$) stacking, see Fig. 1(a)[middle] and [right].

Focusing on the electronic structures, we first calculate the energy bands of hBN bilayer considering $AB$, $B^{NN}$, and $B^{BB}$ stackings. Results are shown in Figs. 1(c), (d) and (e). Note that for the convenience to compare with twisted bilayer form, these calculations are performed with a cell size comparable with the moiré superlattice at the twist angle $\theta = 3.89^\circ$, see Fig. 1(b). Interestingly, the electronic states for different stacking patterns display special energy orders. Compared to $AB$ and $B^{BB}$ stackings, the valence band edge of $B^{NN}$ stacking is of higher energy, Fig. 1(d); while compared to $AB$ and $B^{NN}$ stackings, the conduction band edge of $B^{BB}$ stacking is lower, Fig. 1(e).

To understand these intriguing phenomena caused by the different stacking patterns between layers, we plot schematically the evolution of the band edge states of various layered hBN, Fig. 2(a)[left] shows the VBM state and the CBM state of monolayer hBN. When forming bilayer, the reduced quantum confinement (without considering the stacking effects) causes slightly an upshift of VBM states and a downshift of CBM states, considering the stacking effects) causes slightly a upshift of VBM states and a downshift of CBM states, Fig. 2(b)[middle]. Next, to show the stacking effect, we employ the Hamiltonian

$$H_{12} = \begin{pmatrix} \varepsilon_1 & U_{12} \\ U_{12}^* & \varepsilon_2 \end{pmatrix}$$

where $\varepsilon_1$ and $\varepsilon_2$ denote the states of layer 1 and layer 2, respectively. $U_{12}$ depicts the interlayer coupling. It is worth noting that the energy levels of valence states of cation $B$ atom is much higher than those of the anion $N$ atom. Thus, for hBN, the occupied VBM states are around $N$ atoms and the empty CBM states reside on $B$ atoms and it is the same for the hBN bilayer. For example, inserts of Figs. 1(g) and (h) show that for the $B^{NN}$ stacking, the VBM states are situated at $N$ atoms and
for the $B^{BB}$ stacking the CBM states are situated at B atoms. We further note that in the $B^{NN}$ stacking, the interlayer coupling $U_{12}$ essentially relies on the interaction between the N atoms in one layer and the N atoms in other layer, and $\varepsilon_1 = \varepsilon_2$ are VBM states for both layers. This indicates,

$$\varepsilon_{\pm} = \varepsilon_1 \pm U_{12}. \quad (2)$$

The result is schematically shown in Fig. 2(b)[lower panel], giving rise to higher energy of the bilayer VBM states, Fig. 2(a)[right]. On the other hand, for the $B^{BB}$ stacking, the interlayer coupling ($U_{12}$) is between the CBM states on B atoms in both layers. Again, $\varepsilon_1 = \varepsilon_2$. Hence, similar result of Eq. (2) can be obtained, Fig. 2(b)[upper panel], indicating a downshift to lower energy of the bilayer CBM states, Fig. 2(a)[right]. However, for the AB stacking, the stacking effect on CBM and VBM states is much less, where the interlayer coupling ($U_{12}$) is between the CBM states on B atom in one layer ($\varepsilon_1$) and the VBM states on N atoms in other layer ($\varepsilon_2$). Notice that $U_{12} \ll \varepsilon_1 - \varepsilon_2$, we have,

$$\varepsilon_+ = \varepsilon_1 + U^2_{12}/(\varepsilon_1 - \varepsilon_2); \varepsilon_- = \varepsilon_2 - U^2_{12}/(\varepsilon_1 - \varepsilon_2), \quad (3)$$

showing that the variations in the CBM and VBM states are small.

Because the $B^{NN}$ stacking has the highest VBM states and the $B^{BB}$ stacking has the lowest CBM states than other stacking patterns, the VBM and CBM states of the twisted hBN bilayer are expected to reside within the $B^{NN}$ stacking region and the $B^{BB}$ stacking region, respectively, of the moiré superlattice. As a demonstration, Figs. 1(i) and (j) displaying the charge density distributions of both VBM and CBM states at high symmetry $k = K$ point for the considered twisted hBN bilayer, Fig. 1(f), show that indeed the VBM states are within the $B^{NN}$ stacking region and the CBM states are confined to the $B^{BB}$ stacking region.

For a polar 2D crystal, its twisted bilayer form can be also obtained by applying a small twist angle to the bilayer form with AA stacking, where the B atoms of one layer are on top of the B atoms of the other layer and the same for N atoms, see Fig. 3(a)[left]. The resulting moiré superlattice as shown in Fig. 3(b) develops a new Bernal stacking besides the AA stacking, having N atoms (B atoms) of one layer are on the top of B atoms (N atoms) of the other layer, labeled as $B^{NN}$, see Fig. 3(a)[right].

Figs. 3(c) and (d) displaying the energy bands of hBN bilayer for these two stacking patterns show that compared to the $B^{BB}$ stacking, the valence (conduction) band edge of AA stacking is higher (lower). These results can also be understood by using the Hamiltonian of Eq. (2). For the AA stacking, the interlayer interaction is not only in between the B atoms of both layers but also in between the N atoms of both layers. Therefore, for the VBM states that reside on N atoms in both monolayers, as shown in Fig. 3(f), the coupling between the N states shifts the bilayer VBM states up. Similarly, CBM states reside on B atoms, as shown in Fig. 3(g), the coupling between B states push down the bilayer CBM states. Note that the stacking of the $B^{BN}$ pushes the CBM up and VBM down in energy, as inferred by Eq. (3), so it has little contribution to the charge distribution at the CBM and VBM edges of the twisted hBN bilayer. That is, both the CBM and VBM states of the twisted hBN bilayer shown in Fig. 3(e) reside within the AA stacking region, as demonstrated by Figs. 3(h) and (i).

Because for the moiré superlattice, regions for a specific stacking pattern are spatially separated, it is thus possible that the electronic states (CBM states and/or VBM states) resided in these regions form flat energy bands when the separation of the regions is large. Indeed, as shown in Fig. 4(a), the charge density distribution of
the CBM state at \( k = K \) of the twisted hBN bilayers displays a gradual localization around the \( AA \) stacking regions with the decrease of the twist angle \( \theta \). Similar trend can be also identified for VBM states as shown in Fig. S2 of the Supplemental Material. This mainly happens because that the distance between the centers of regions for each stacking pattern is larger at smaller \( \theta \). Notice that the space between regions for a particular stacking pattern is not as perfect as that in the bilayer form, which is computationally demanding, could be avoided. This mechanism brings up two benefits. (i) Similar flat bands are expected to emerge in the twisted bilayer structure, Bloch flat bands emerge as long as the twist angle is sufficiently small. By analyzing the evolution of the valence and conduction band edges of the bilayer with different stacking patterns, a mechanism attributed to the splitting of the band edge states induced by different stacking patterns is revealed. This mechanism differs from that for twisted graphene bilayers where a magic angle is needed. This new mechanism brings up two benefits. (ii) The possibility of the formation of the flat bands for a twisted bilayer can be inferred by just examining the energy order of the VBM and CBM states of the various bilayers forming the stacking patterns of the twisted bilayer. (iii) For example, we have studied the electronic proprieties of the bilayer and twisted bilayers of polar 2D crystals such as transition-metal dichalcogenides \([28, 36]\), and 2D oxides \([37]\) at small twist angles \( \theta \). A separation hints that the coupling between electronic states residing in these regions is weak, giving rise to flat bands. The obtained flat bands in twisted hBN bilayers are important and the above results reveal several important aspects which should be common for polar 2D crystals.

**FIG. 4:** (color online) (a) Twisted hBN bilayers with different twist angle \( \theta \). The circles with radius \( R \) highlight the \( AA \) stacking regions. Electronic density distribution of the conduction band edge state at \( k = K \), as marked by circles in (b). (b) Energy bands of twisted hBN bilayers at different twist angle \( \theta \). The dashed horizontal line indicates the energy zero.

In summary, using the DFTB method and taking hBN as an example, we have studied the electronic proprieties of the bilayer and twisted bilayers of polar 2D crystals and shown that in the twisted bilayer structure, Bloch flat bands emerge as long as the twist angle is sufficiently small. By analyzing the evolution of the valence and conduction band edges of the bilayer with different stacking patterns, a mechanism attributed to the splitting of the band edge states induced by different stacking patterns is revealed. This mechanism differs from that for twisted graphene bilayers where a magic angle is needed. This new mechanism brings up two benefits. (i) Similar flat bands are expected to emerge in the twisted bilayer of other polar 2D crystals such as transition-metal dichalcogenides \([28, 36]\), and 2D oxides \([37]\) at small twist angles \( \theta \). (ii) The possibility of the formation of the flat bands for a twisted bilayer can be inferred by just examining the energy order of the VBM and CBM states of the various bilayers forming the stacking patterns of the twisted bilayer. The direct simulation of the twisted bilayer, which is computationally demanding, could be avoided. These findings, thus pave a new route to explore Bloch flat bands and associated many-body physics in 2D materials.
This work was supported by NSFC under Grants Nos. 11674022, 51672032, 11634003, and U1530401. D.-B.Z. was supported by the Fundamental Research Funds for the Central Universities. X.-J.Z. was supported by Postdoctoral innovative talents support program (BX201700025).

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