Misalignment instability in magic-angle twisted bilayer graphene on hexagonal boron nitride

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Abstract
We study the stability and electronic structure of magic-angle twisted bilayer graphene on the hexagonal boron nitride (TBG/BN). Structural relaxation has been performed for commensurate supercells of the heterostructures with different twist angles ($\theta'$) and stackings between TBG and BN. We find that the slightly misaligned configuration with $\theta' = 0.54^\circ$ and the AA/AA stacking has the globally lowest total energy due to the constructive interference of the moiré interlayer potentials and thus the greatly enhanced relaxation in its $1 \times 1$ commensurate supercell. Gaps are opened at the Fermi level ($E_F$) for small supercells with the stackings that enable strong breaking of the $C_2$ symmetry in the atomic structure of TBG. For large supercells with $\theta'$ close to those of the $1 \times 1$ supercells, the broadened flat bands can still be resolved from the spectral functions. The $\theta' = 0.54^\circ$ is also identified as a critical angle for the evolution of the electronic structure with $\theta'$, at which the energy range of the mini-bands around $E_F$ begins to become narrower with increasing $\theta'$ and their gaps from the dispersive bands become wider. The discovered stabellest TBG/BN with a finite $\theta'$ of about 0.54° and its gapped flat bands agree with recent experimental observations.

1. Introduction
The recently realized magic-angle twisted bilayer graphene (TBG) has inspired great interest in exploring its peculiar electronic structure [1–5]. Superconductivity and correlated-insulator phases associated with the low-energy flat bands have been observed in TBG with twist angles around the first magic angle ($\theta_m$) of about 1.1° [1–8]. In the TBG devices, the hexagonal boron nitride (BN) not only acts as the ideal atomically flat van der Waals substrates but also can facilitate the realization of the quantized anomalous Hall (QAH) effect in TBG nearly aligned with BN (TBG/BN) [9]. Due to the lattice-constant mismatch between graphene and BN, the largest moiré supercell of graphene on BN occurs when the layers are perfectly aligned [10–12]. In contrast, the twist angle ($\theta'$) between TBG and BN in the experimental devices with the QAH effect were observed to be about 0.6° [9]. The energetics mechanism behind this remains to be revealed. For the pristine TBG with $\theta_m$, the flat bands around the Fermi level ($E_F$) already have the minimum widths. Our previous study demonstrated that the effects of BN on the electronic structure associated with the flat bands in TBG/BN can be beyond the perturbation regime as BN induces both the direct modification of the Hamiltonian and strong structural deformation in the graphene layer adjacent to BN that break the $C_2$ symmetry of TBG, while only the minimum commensurate supercell of TBG/BN with one specific $\theta' = 1.64^\circ$ was considered [13]. Other previous studies also focused on specific configurations of TBG/BN or only considered the rigid superlattices [13–18]. Therefore, it is important to systematically explore the evolution of the energetic and electronic properties of relaxed TBG/BN with $\theta'$ to understand the stability and band structures of the experimentally observed configurations.

Here in-plane and out-of-plane relaxation of the commensurate supercells of magic-angle TBG on BN with different $\theta'$ and stacking configurations is performed and their electronic structures are acquired based on the effective Hamiltonian taking into account the relaxation effect and the full moiré
Hamiltonian induced by BN. We find that the slightly misaligned configuration with \( \theta' = 0.54^\circ \) and the AA/AA stacking has the globally lowest total energy due to the constructive interference of the moiré interlayer potentials, and gaps are opened at \( E_F \) for small supercells with the stacking that enables strong breaking of the \( C_2 \) symmetry in the atomic structure of TBG. The \( \theta' = 0.54^\circ \) is also demonstrated to be a critical angle for the evolution of the electronic structure with \( \theta' \), at which the energy range of the minigaps around \( E_F \) begins to become narrower with increasing \( \theta' \) and their gaps from the dispersive bands become wider.

2. Commensurate supercells of TBG/BN

We study the trilayer heterostructures with the top magic-angle TBG nearly aligned with the bottom BN layer. The top graphene layer (G3) and the bottom BN layer are rotated by \( \theta \) and \( \theta' \) counterclockwise with respect to the fixed middle graphene layer (G2), respectively. The B and N atoms are represented by blue and red circles, respectively. The high-symmetry local stackings between G3 and G2 and those between G2 and BN are labeled. \( \mathbf{L}_j \) (\( j = 1, 2 \)) are the basis vectors of the moiré superlattice in G3/G2, and \( \mathbf{L}_j' \) are those in G2/BN. (b) Variations of the length \( L_j' \) and the angle \( \phi \) from \( L_j' \) to \( L_j \), as a function of \( \theta' \) for \( \phi = 1.08^\circ \). \( \tilde{L} \) is the length of \( L_j \). The filled circles represent the strictly commensurate supercells. The four small supercells are displayed schematically and their \( \theta' \) are labeled. The gray, blue, and green lines represent the supercell of TBG/BN, the moiré cell of G3/G2, and that of G2/BN, respectively. \( \mathbf{L}_j \) are the supercell vectors. (c) The length \( L_j' \) of \( \mathbf{L}_j \) for the supercells at different \( \theta' \).

Figure 1. The geometry of the commensurate supercells of TBG/BN. (a) The schematic view of the TBG/BN trilayer. The top graphene layer (G3) and the bottom BN layer are rotated by \( \theta \) and \( \theta' \) counterclockwise with respect to the fixed middle graphene layer (G2), respectively. The B and N atoms are represented by blue and red circles, respectively. The high-symmetry local stackings between G3 and G2 and those between G2 and BN are labeled. \( \mathbf{L}_j \) (\( j = 1, 2 \)) are the basis vectors of the moiré superlattice in G3/G2, and \( \mathbf{L}_j' \) are those in G2/BN. (b) Variations of the length \( L_j' \) and the angle \( \phi \) from \( L_j' \) to \( L_j \), as a function of \( \theta' \) for \( \phi = 1.08^\circ \). \( \tilde{L} \) is the length of \( L_j \). The filled circles represent the strictly commensurate supercells. The four small supercells are displayed schematically and their \( \theta' \) are labeled. The gray, blue, and green lines represent the supercell of TBG/BN, the moiré cell of G3/G2, and that of G2/BN, respectively. \( \mathbf{L}_j \) are the supercell vectors. (c) The length \( L_j' \) of \( \mathbf{L}_j \) for the supercells at different \( \theta' \).
We consider the strictly periodic moiré superlattices in G3/G2. The hexagonal superlattice is spanned by the basis vectors \( \mathbf{L}_1 = N\mathbf{a}_1 + (N + 1)\mathbf{a}_2 \) and \( \mathbf{L}_2 = T_{60}\mathbf{L}_1 = -(N + 1)\mathbf{a}_1 + (2N + 1)\mathbf{a}_2 \). The twist angle \( \theta \) is given by \( \cos \theta = (1 + 6N + 6N^2)/(2 + 6N + 6N^2) \). We focus on the \( N = 30 \) case with \( \theta = 1.0845^\circ \) which is closest to the experimentally observed \( \theta_m \). In G2/BN, the spanning vectors of the moiré superlattices are taken as \( (S^{-1} - I)\mathbf{L}_1 = -2\mathbf{a}_2 \) and \( (S^{-1} - I)\mathbf{L}_2 = \mathbf{a}_1 - 2\mathbf{a}_2 \) with \( \mathbf{L}_1 = T_{60}\mathbf{L}_1 \). Starting from the aligned TBG on BN, commensurate supercells of the double superlattices arise when rotating TBG to BN. For \( \theta' = 0^\circ \), the length \( (L') \) of \( \mathbf{L}_1' \) is larger than that of \( \mathbf{L}_1 \), while \( L' \) decreases with \( \theta' \) and becomes equal to \( L \) at \( \theta' \approx 0.54^\circ \), as shown in figure 1(b). More importantly, \( \mathbf{L}_1' \) rotates clockwise with increasing \( \theta' \) and the angle \( \phi \) changes from \( 30^\circ \) at \( \theta' = 0^\circ \) to zero at \( \theta' \approx 0.54^\circ \). Then the two superlattices coincide at \( \theta' \approx 0.54^\circ \) with the minimum commensurate supercell. At larger \( \theta' \), \( L' \) becomes smaller than \( L \) and the sign of \( \phi \) changes.

All the possible hexagonal commensurate supercells are obtained as follows. The basis vector of the supercell \( \mathbf{l}_1 = n_1\mathbf{L}_1 + n_2\mathbf{L}_2 \) satisfies

\[
(S^{-1} - I)\mathbf{L}_1 = n_1(-\mathbf{a}_2) + n_2(\mathbf{a}_1 - \mathbf{a}_2)
\]

with integer values of \( n_1 \) and \( n_2 \) so that it is also a lattice vector of the superlattice in G2/BN. The other basis vector is \( \mathbf{l}_2 = T_{60}\mathbf{L}_1 = -(n_1 + n_2)\mathbf{L}_1 \). The two superlattices coincide when \( (S^{-1} - I)\mathbf{L}_1 = -\mathbf{a}_2 \), which gives \( \varepsilon = N/\sqrt{1/3 + N + N^2} - 1 \) and \( \cos \theta' = (3 + 6N)/(2\sqrt{3} + 9N + 9N^2) \). For \( N = 30 \) with \( \theta = 1.0845^\circ \), \( \varepsilon = -1.6437^\circ \) and \( \theta' = 0.5423^\circ \). This \( \varepsilon \) is just close to the ab-initio value of \(-1.70^\circ \). For \( N = 29 \) with \( \theta = 1.1213^\circ \), the obtained \( \varepsilon \) can also be close to \(-1.70^\circ \), while for \( \theta \) quite away from \( \theta_m \), the required \( \varepsilon \) becomes rather deviated from \(-1.70^\circ \), then the minimum commensurate supercell can not occur. In the following, we focus on cases with \( \theta = 1.0845^\circ \) which is closest to the \( \theta_m \). For other \( \theta' \) from \( 0^\circ \) to \( 2^\circ \), the \( \mathbf{l}_1 \) satisfying equation (2) exactly is searched numerically by varying \( \varepsilon \) around \(-1.6437^\circ \) very slightly for \( \theta = 1.0845^\circ \). The obtained strictly periodic supercells are listed in table S1 in the SM. We note that to study the energetic stabilities of these supercells, the differences between \( \varepsilon \) should be extremely small, which are all smaller than 0.026% here. For most cases in table S1, the supercell vector can be expressed as \( \mathbf{l}_1 = n\mathbf{L}_1 \) with \( n = 1 \sim 9 \). Since these supercells are comprised of \( n^2 \) TBG moiré cells, they are denoted by \( n \times n \). Their structural parameters can be computed analytically as given in the SM.

Figure 1(c) shows the length \( (L) \) of the supercell basis vectors as a function of \( \theta' \). Four cases have \( L \leq 2L \), including the one with almost perfect alignment between G2 and BN, as shown in figure 1(b). The \( L \) of the largest considered supercell reaches \( 9L \) (116.3 nm). The \( 1 \times 1 \) supercell is comprised of 16 564 atoms, then the \( 9 \times 9 \) supercell contains more than one million atoms. The reciprocal space of the TBG/BN supercell is shown schematically in figure S1.

The sublattice-A and sublattice-B atoms in a unit cell of graphene are located at \((\mathbf{a}_1 + \mathbf{a}_2)/3\) and \((2\mathbf{a}_1 + 2\mathbf{a}_2)/3\), respectively. In BN, the lattices formed by the boron and nitrogen atoms are labeled as sublattice-A and sublattice-B, respectively. Besides the relative twist between BN and TBG, BN can be shifted with respect to TBG forming different stacking configurations. For the configurations with the \( C_3 \) symmetry, the stackings of G3/G2 and G2/BN at the origin are one of AA, AB, and BA, as shown in figure 1(a), which are used to denote the stacking between TBG and BN like AA/AA with the left one for G3/G2. Among the nine possible symmetric stackings, only three are inequivalent. The local stackings between adjacent layers vary continuously and are characterized by the relative shift vectors. At an in-plane position \( r \) in the rigid superlattice, the shift vector between G3 and G2 is taken to be \( \delta = (I - T_{-\theta})r + \tau_{32} \), and that between G2 and BN is given by \( \delta' = (S^{-1} - I)r + \tau_{32} \), with \( \tau_{32} \) the shift vectors at the origin.

### 3. Energetics of relaxed supercells

The rigid moiré superlattices undergo spontaneous in-plane relaxation due to the energy gain from the larger domains of energetically favorable local stackings [20–31]. Each layer is also corrugated to reach the optimal interlayer distances of the varying local stackings across the superlattices. We have performed structural relaxation of TBG/BN with different \( \theta' \) and stackings employing the continuum elastic theory. The interlayer potentials as a function of the local shift vectors are represented by a Fourier expansion over the graphene lattice [32]. The total energy \( (E_{tot}) \) of TBG/BN as a functional of the displacement field in each layer is minimized by Euler–Lagrange equations with the displacement fields expanded in Fourier series, as detailed in the SM.

The variation of \( E_{tot} \) with \( \theta' \) and stackings shows that the energetically stables configuration occurs at \( \theta' = 0.54^\circ \) and has the AA/AA stacking, as shown in figure 2(a). The elastic and interlayer interaction energies that sum to \( E_{tot} \) can be seen in figure S2. At \( \theta' = 0.54^\circ \) with the \( 1 \times 1 \) supercell, the three stackings have distinct \( E_{tot} \), while the \( E_{tot} \) for different stackings at other \( \theta' \) are rather similar. The general trend of \( E_{tot} \) with \( \theta' \) suggests that \( E_{tot} \) reaches minimum at about \( 0.54^\circ \) even without strict commensurability. To realise the stables AA/AA stacking at \( \theta' = 0.54^\circ \), there should not be high energy barriers across the three symmetric stackings. An energy barrier indeed exists when continuously shifting TBG relative to BN from AA/AB to AA/AA, while it is just 0.03 meV UC\(^{-1}\), much smaller than the difference of
$E_{\text{tot}}$ between the two symmetric stackings, as shown in figure 2(b). Therefore, TBG/BN tends to have the configuration with $\theta' = 0.54^\circ$ and the AA/AA stacking when the $\theta'$ of the initial assembled structure is close to this angle and is allowed to be relaxed by, for example, annealing. Other structures with $\theta'$ rather away from this angle may still exist. In experiments, TBG/BN with $\theta' \approx 0.6^\circ$ was realized [9]. Such a slightly larger $\theta'$ than $0.54^\circ$ can be due to a larger $\theta$ within the experimental samples and the ambiguity of the experimental determination of $\theta'$. The calculated $E_{\text{tot}}$ for other two $\theta'$ quite away from $\theta_{\text{min}}$ indeed show that the $\theta'$ with minimum $E_{\text{tot}}$ for a larger (smaller) $\theta$ becomes larger (smaller) than $0.54^\circ$, as seen in figure S3.

The mechanism behind the minimum $E_{\text{tot}}$ with $\theta' = 0.54^\circ$ and the AA/AA stacking is that the moiré interlayer potentials in G3/G2 and G2/BN interfere constructively so that the most favorable BA and AB stackings in G3/G2 are located at the same position as the most favorable BA stacking and less favorable AB stacking in G2/BN, as seen in figure 2(d). In contrast, the completely destructive interference of the moiré potentials for the AA/AB stacking at $\theta' = 0.54^\circ$ leads to the highest $E_{\text{tot}}$ among all $\theta'$ and stackings. Upon relaxation from the rigid superlattices, the parts with favorable local interlayer stackings tend to increase their sizes. For the AA/AA case, the favorable BA-like stackings in G3/G2 and G2/BN share the same positions in G2 so that these positions can be strained to increase the sizes of both favored stackings. However, the complete separation of the positions in G2 with favorable stackings of G3/G2 from those of G2/BN for AA/AB leads to opposite forces exerted by G3 and BN in some positions of G2 so that the in-plane relaxation of G2 is greatly suppressed. The interlayer potentials for the almost perfect alignment of TBG on BN ($\theta' = 0.01^\circ$) are also exhibited in figure 2(e) for comparison. The supercell of these configurations is comprised of four moiré cells of G3/G2 and three moiré cells of G2/BN. Among the three BA-stacked positions of G2/BN in one supercell, two coincide with the BA and AB stackings in G3/G2 for the AA/BA case so that it has a lower energy, while the BA-stacked positions in G2/BN are still located in the vicinity of some BA and AB stackings in G3/G2 for the other two cases. Then the $E_{\text{tot}}$ of the three stackings are similar at $\theta' = 0.01^\circ$. For other $\theta'$ with larger supercells, more local stacking configurations between the two moiré superlattices are included in the supercells so that the $E_{\text{tot}}$ is less sensitive to the global shift between G2 and BN. Moreover, some of
the BA-stacked positions of G2/BN are also separated from the BA and AB stackings of G3/G2 in these larger supercells, leading to a higher energy than that at $\theta' = 0.54^\circ$ with the AA/AA stacking.

The constructive interference of the moiré potentials results in the strongest atomic relaxation for the AA/AA stacking at $\theta' = 0.54^\circ$, in contrast to the suppressed relaxation for the AA/BA stacking, as demonstrated in figure 3. Upon relaxation, the regions with favorable BA-like and AB-like stackings in G3/G2 increase in size, and they are much larger for AA/AA than that for AA/BA. The stronger relaxation for AA/AA is also reflected evidently in the spatial distributions of the elastic-energy density, whose highest value for AA/AA is much higher than that for AA/BA. When $\theta'$ is away from $0.54^\circ$ with a larger value, the relaxation also becomes much weaker than that at smaller $\theta'$, as reflected in the more suppressed structural deformation in G2 at larger $\theta'$ (see figure S4).

4. Electronic structure of TBG/BN

For the relaxed TBG/BN, we have built an effective Hamiltonian $\hat{H}$ for the moiré superlattice in G3/G2 by extending the Hamiltonian of $p_z$ orbitals for graphene bilayers and the effective Hamiltonian of monolayer graphene on BN [13]. It is noted that all the electronic effect of BN has been taken into account in the effective Hamiltonian by the spatially varying intralayer hopping terms in G2 [33]. This effective Hamiltonian reads

$$\hat{H} = \sum_{n=2}^{3} \sum_{i,j} \epsilon_{n,i,j} \hat{c}_{n,i,j}^\dagger \hat{c}_{n,i,j} + \sum_{n=2}^{3} \sum_{(i,j)} \epsilon_{n,i,j}^{(n,n)} \hat{c}_{n,i,j}^\dagger \hat{c}_{n,i,j} + h.c.$$

$$+ \sum_{i,j} \epsilon_{i,j}^{(2,3)} \hat{c}_{i,j}^\dagger \hat{c}_{i,j} + h.c.,$$

where $\hat{c}_{n,i,j}^\dagger (n = 2, 3)$ is the creation and $\hat{c}_{n,i,j}$ is the annihilation operator of a $p_z$-like orbital at the site $i$ in the Gn layer and $\epsilon_{n,i,j}$ denotes the intralayer nearest neighbors. The on-site energies, intralayer and interlayer hopping terms are represented by $\epsilon_{n,i,j}$, $\epsilon_{i,j}^{(n,n)}$, and $\epsilon_{i,j}^{(2,3)}$, respectively, and are given in the SM. These Hamiltonian terms are obtained taking into account the relaxation effect and the full moiré Hamiltonian induced by BN, and $\hat{H}$ is diagonalized using the plane-wave-like basis functions as the supercells can have a huge number of atoms, as detailed in the SM.

The most desirable consequence of the near alignment of TBG with BN for the electronic structure is that the flat conduction and valence bands can become separated by a gap at $E_F$ due to the broken $C_2$ symmetry in TBG by BN. Our calculations show that such a gap is only present in systems with small commensurate supercells, as shown in figure 4. Moreover, among the three stackings, the flat bands could be gapped at $E_F$ for the AA/AA and AA/AB stackings, while they overlap at $E_F$ for AA/BA. For AA/AA and AA/AB, the $C_2$ symmetry is strongly broken in the relaxed atomic structure due to the evident absence of the inversion symmetry with respect to the origin in the interlayer potential between G2 and BN (see figure 2(d)). In contrast, the interlayer potential between G2 and BN is roughly inversion symmetric with respect to the origin for AA/AB so that the $C_2$ symmetry is only weakly broken by the structural relaxation, and the avoided crossings of the flat bands can be attributed to the electronic contribution of BN. Although the gap at $E_F$ only arises in specific configurations, it can be readily observed in experiment [9] as the globally stabelst system with $\theta' = 0.54^\circ$ and the AA/AA stacking just has a rather large gap, as shown in figure 4(a).
For the $2 \times 2$ supercells at $\theta' = -0.01^\circ$ and $1.09^\circ$, the mini-bands on the electron or hole side could also be separated from each other, as shown in figures 4(c) and 4(d). However, the band separation depends on the stacking. For example, at $\theta' = -0.01^\circ$, the first two conduction bands are gapped from the higher bands for AA/AA and AA/AB, while only the lowest conduction band is gapped from the other three overlapped conduction bands for AA/BA. Such sensitivity of band structures to stackings of small supercells suggests complicated mini-bands in large supercells.

Figure 5 shows the band structures of TBG/BN with the $7 \times 7$ or $9 \times 9$ supercell at four different $\theta'$. The bands of such large supercells are no longer sensitive to stackings. Most middle mini-bands around $E_F$ become completely flat lines and are well separated from the dispersive bands, while no gaps can be observed at $E_F$. For the two smaller $\theta'$, the energy range ($W$) of the middle mini-bands is rather large, while it decreases with the two larger $\theta'$. The gaps ($\Delta_e$) between the dispersive conduction bands and the middle mini-bands are all rather large, while the middle mini-bands approach the dispersive valence bands with a vanishing gap ($\Delta_h$) between them at $\theta' = 0.42^\circ$. The variations of $W$, $\Delta_e$, and $\Delta_h$ with $\theta'$ are exhibited in figure 5(b), which clearly infer that $\theta' = 0.54^\circ$ is the critical angle for the evolution of the electronic structure of TBG/BN with $\theta'$. At $\theta' < 0.54^\circ$, $W$ remains large and $\Delta_h$ can reach near zero, which can be attributed to the significant contribution of BN to the structural relaxation and electronic perturbation in TBG as the moiré cell of G2/BN is larger than that of G3/G2. At $\theta' > 0.54^\circ$, $W$ decreases rapidly with $\theta'$ and the increasing $\Delta_h$ becomes rather large. We also notice that small dips occur for $\Delta_h$ and $\Delta_e$ at $\theta' = 1.64^\circ$ with the $1 \times 1$ supercell, which could be due to the abrupt changes of the local stacking configurations around this minimum commensurate supercell. In comparison with $\theta' = 0.54^\circ$, the moiré superlattice of G2/BN at $\theta' = 1.64^\circ$ is much smaller so that the trend of the gaps is only slightly affected by the occurrence of the $1 \times 1$ commensurate supercell at this angle.

In order to resolve the effective energy dispersions for large supercells with complicated mini-bands around $E_F$, the spectral functions unfolded to the BZ of the pristine TBG have been computed, as shown in figure 6. We find that the flat bands corresponding to those of the pristine TBG could still be identified for systems close to the $1 \times 1$ commensurate configurations (see figures 6(b)–(d)), while no energy dispersions could be resolved from the spectral function for the system close to the $2 \times 2$ configuration (see figure 6(a)). The broadened flat bands could be well separated (see figure 6(b)), while they could also overlap in the vicinity of $\Gamma^0$ (see figures 6(c) and (d)). These features of the spectral functions may be observed in future ARPES measurement of TBG/BN.

**Figure 4.** The band structures of TBG/BN with the $1 \times 1$ supercell at $\theta' = 0.54^\circ$ (a) and $\theta' = 1.64^\circ$ (b) and with the $2 \times 2$ supercell at $\theta' = -0.01^\circ$ (c) and $\theta' = 1.09^\circ$ (d) for the three inequivalent symmetric stackings. The superimposed bands of the pristine TBG without BN are represented by dashed lines. The Fermi levels are set to be zero.
Figure 5. (a) The band structures of TBG/BN with large supercells at four different $\theta'$. (b) The energy range ($W$) of the middle mini-bands, the gap ($\Delta$) between the dispersive conduction bands and the middle mini-bands, and the gap ($\Delta_h$) separating the middle mini-bands from the dispersive valence bands for all considered configurations with different stackings and $\theta'$.

Figure 6. The spectral functions unfolded to the BZ of the pristine TBG corresponding to the band structures in figure 5(a) for large supercells at four $\theta'$. The $\theta' = 0.07^\circ$ (a) is close to $\theta' = -0.01^\circ$ that has a $2 \times 2$ supercell, $\theta' = 0.42^\circ$ (b) and $\theta' = 0.66^\circ$ (c) are close to $\theta' = 0.54^\circ$ that has a $1 \times 1$ supercell, and $\theta' = 1.52^\circ$ (d) is close to $\theta' = 1.64^\circ$ that has a $1 \times 1$ supercell.
5. Summary and conclusions

A series of commensurate supercells of TBG/BN with magic angle within TBG and with varying $\theta'$ and stackings between TBG and BN have been constructed. Structural relaxation of the supercells has been performed, which shows that the energetically stabllest configuration is slightly misaligned between TBG and BN with $\theta' = 0.54^\circ$ and has the AA/AA stacking. This is due to the completely constructive interference of the moiré interlayer potentials and thus the greatly enhanced relaxation in the 1×1 supercell of this configuration. In contrast, in-plane relaxation can be partially suppressed in the graphene layer adjacent to BN for other configurations. The band structures of the supercells are acquired based on the effective Hamiltonian taking into account the relaxation effect and the full moiré Hamiltonian induced by BN. As the supercells have a huge number of atoms, the Hamiltonian is diagonalized using the plane-wave-like basis. Gaps are opened at $E_F$ for small supercells with stackings that enable strong breaking of the $C_2$ symmetry in the atomic structure of TBG. For large supercells with $\theta'$ close to those of the 1×1 supercells, the broadened flat bands can still be resolved from the spectral functions. The $\theta' = 0.54^\circ$ is also identified as a critical angle for the evolution of the electronic structure with $\theta'$, at which the energy range of the mini-bands around $E_F$ begins to become narrower with increasing $\theta'$ and their gaps from the dispersive bands become wider. The discovered stabllest TBG/BN with a finite $\theta'$ of about 0.54$^\circ$ and its gapped flat bands agree with recent experimental observations.

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