Pressure induced gap modulation and topological transitions in twisted bilayer and double bilayer graphene

Xianqing Lin,1,* Haotian Zhu,1 and Jun Ni2

1College of Science, Zhejiang University of Technology, Hangzhou 310023, People’s Republic of China
2State Key Laboratory of Low-Dimensional Quantum Physics and Frontier Science Center for Quantum Information, Department of Physics, Tsinghua University, Beijing 100084, People’s Republic of China
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We study the electronic and topological properties of fully relaxed twisted bilayer (TBG) and double bilayer (TDBG) graphene under perpendicular pressure. An approach has been proposed to obtain the equilibrium in-plane structural deformation and out-of-plane corrugation in moiré superlattices under pressure. We find that the in-plane relaxation becomes much stronger under higher pressure, while the corrugation height in each layer is maintained. The comparison between band structures of relaxed and rigid structures demonstrates that not only the gaps on the electron and hole sides ($\Delta_e$ and $\Delta_h$) are significantly underestimated without relaxation but also the detailed dispersions of the middle bands of rigid structures are rather different from those of relaxed systems. $\Delta_e$ and $\Delta_h$ in TBG reach maximum values around critical pressures with narrowest middle bands. Topological transitions occur in TDBG under pressure with the middle valence and conduction bands in one valley touching and their Chern numbers transferred to each other. The pressure can also tune the gap at the neutrality point of TDBG, which becomes closed for a pressure range and reopened under higher pressure. The behavior of electronic structure of superlattices under pressure is sensitive to the twist angle $\theta$ with the critical pressures generally increase with $\theta$.

I. INTRODUCTION

Realization of magic-angle twisted bilayer graphene (TBG) has recently intrigued great interest in exploring their peculiar electronic structure associated with the nearly flat bands around the Fermi level ($E_F$)\textsuperscript{1–4}. In TBG with twist angles ($\theta$) around the first magic $\theta$ of about 1.1\textsuperscript{5,6}, experiments observed correlated-insulator and nonconventional-superconductivity phases\textsuperscript{1–4}, which were found to be extremely sensitive to $\theta$. With $\theta$ away from the magic angle, the middle bands around $E_F$ become much wider\textsuperscript{5–12}, while the perpendicular pressure has been demonstrated experimentally to be able to flatten these bands again\textsuperscript{13}. Pressure thus provides an efficient way of tuning TBG into the magic regime besides the precise control of $\theta$. The narrowing of the middle bands under pressure\textsuperscript{13} has also been confirmed by theoretical calculations\textsuperscript{14,15}. However, these studies only considered rigid TBG or some relaxation effect in TBG with one empirical parameter and mainly focused on the widths of middle bands\textsuperscript{14,15}, while the rigid superlattices undergo spontaneous in-plane relaxation and out-of-plane corrugation due to the energy gain from the larger domains of energetically favorable stacking configurations\textsuperscript{16–27}. The detailed electronic structure of TBG around $E_F$ can be greatly affected by the structural deformation\textsuperscript{21,22,24,26,27}, especially the gaps on the electron and hole sides and the explicit dispersions of the nearly flat bands. Therefore, it is important to obtain the energetically stable structures of TBG to study the evolution of their electronic structure under pressure. First, the variation of realistic structural parameters with pressure can be provided by full relaxation of compressed TBG. Second, the relaxation effect can be taken into account in calculations of the electronic structure of TBG under pressure so that the critical pressures into the magic regime and the pressure induced modulations of electronic properties corresponding to experimental systems can be identified.

Besides TBG, twisted double bilayer graphene (TDBG) with relative rotation between the top and bottom graphene bilayers has been realized recently\textsuperscript{28–30}. In addition to the nearly flat middle bands in TDBG with a small $\theta$, a gap at $E_F$ is opened, and among various stacking arrangements between the bilayers AB-BA TDBG can become valley Hall insulators\textsuperscript{31–35}. Then the perpendicular pressure may be employed to tune the electronic\textsuperscript{32} and topological properties of TDBG, and the structural relaxation\textsuperscript{31} can be important to predict these properties under pressure. It is also noted that the pressure effect on topological properties of TDBG remains to be revealed.

Here we propose an approach to fully relax TBG and AB-BA TDBG under perpendicular pressure. We find that the in-plane relaxation becomes stronger under higher pressure. The gaps on the electron and hole sides, the dispersions of middle bands of TDBG and the neutrality-point gap of TDBG can be effectively modulated by pressure. Pressure induced topological transitions can be observed in TDBG.

The outline of this paper is as follows: In Sec. II we study the full relaxation of moiré superlattices in TBG and TDBG under perpendicular pressure. The band structures of systems with and without relaxation are compared in Sec. III. For the fully relaxed structures, we show the gap modulation and evolution of bands in TBG under pressure in Sec. IV and the topological and electronic transitions in TDBG induced by pressure in Sec. V. Section VI presents the summary and conclusions.
FIG. 1. (Color online) The full relaxation of TBG and TDBG under perpendicular pressure. (a) The out-of-plane atomic corrugation of TBG with \( \theta = 1.248^\circ \) under pressure of 3.2 GPa along the line shown in (b). The z direction is perpendicular to the layers. \( h_0 \) is the average interlayer distance and the high-symmetry local stackings are labeled. (b) The spatial distribution of the local interlayer interaction energy \( V \) as a function of the position in the superlattice of TBG under pressure of 3.2 GPa. \( V \) is in units of meV per graphene unit cell (UC). (c) The corrugation (solid lines) and the elastic energy density (dashed lines) in the top layer of TBG under zero and a high pressure. (d) The variation of \( E_{\text{tot}} \) of a TBG supercell with \( h_0 \) for \( \theta \) from 1° to 2°. \( E_{\text{tot}} \) is in units of meV per graphene unit cell (UC). The black line represents the equilibrium \( h_0 \) of free TBG under zero pressure. (e) The \( h_0 \) as a function of the calculated pressure and \( \theta \). (f) The average interlayer distances (solid lines) and the elastic energy (dashed lines) of one layer in the supercell of TDBG with \( \theta = 1.538^\circ \) under increasing pressure. The average interlayer distances between the two middle layers (\( h_0 \)) and between the two top layers (\( h'_0 \)) and the displacement fields in the two top layers (\( u(r) \) and \( u'(r) \)) are labeled in the inset.

II. RELAXATION OF MOIRÉ SUPERLATTICES UNDER PERPENDICULAR PRESSURE

We consider moiré superlattices in TBG and AB-BA TDBG, where the top layer in TBG has been rotated by \( \theta \) counterclockwise with the bottom layer fixed and the top BA-stacked bilayer in TDBG is twisted with respect to the bottom AB-stacked bilayer. The mathematical formulation of the moiré superlattice and its reciprocal lattice are detailed in the Appendix.

The rigid moiré superlattices undergo spontaneous in-plane relaxation due to the energy gain from the larger domains of energetically favorable stacking configurations. Each layer is also corrugated to reach the optimal interlayer distances \( (h) \) of the varying stacking configurations across the superlattice. Here we propose a method to fully relax TBG under perpendicular pressure. The TBG with a fixed average interlayer distance \( (h_0 \) shown in Fig. 1(a)) is first relaxed to calculate the variation of its total energy \( (E_{\text{tot}}) \) with \( h_0 \). Then the pressure is obtained by differentiation of \( E_{\text{tot}} \) with respect to \( h_0 \). TDBG under pressure can be relaxed in a similar way.

Among all configurations of the shifted graphene bilayer, the AB and BA stacked structures have the lowest energy, and the AA stacking is the most unfavorable. In view of the rotation and inversion symmetry relations of shifted bilayers, the calculated energy of a bilayer with the same \( h \) as a function of the shift vector \( \delta \) can be expressed as \( V(\delta, h) = V_0(h) + V(h) \sum_{j=1}^{3} \cos(G_j \cdot \delta) \), where the sum is limited to three shortest vectors, \( G_1 = b_1 \), \( G_2 = -b_1 + b_2 \), and \( G_3 = -b_2 \), \( b_i \) (i=1,2) are basis vectors of the reciprocal lattice of graphene, and components with longer \( G_j \) are negligible \(^{24,36} \). The dependence of \( V_0 \) and \( V \) on \( h \) is detailed in the Appendix. We note that \( V(h) \) is positive for \( h \) smaller than 4.22 Å and increases exponentially with decreasing \( h \), which indicates that the high perpendicular pressure applied to TBG with small average \( h \) tends to enhance the energy differences among configurations with different stackings. It can thus be anticipated that superlattices under higher pressure may have much stronger in-plane structural deformation and also reduced out-of-plane corrugation, which may indicate stronger influence of relaxation on the electronic structure.

We have adopted the continuum elastic theory to express \( E_{\text{tot}} \) of a moiré supercell. \( E_{\text{tot}} \) as the sum of the
elastic energy \((E_{el})\) in each layer and interlayer interaction energy \((E_{int})\) is a functional of the displacement fields \(u_j^j(r)\) with \(j = 1\) for the bottom layer and \(j = 2\) for the top layer and the interlayer distance field \(h(r)\). The elastic energy in a layer with the displacement field \(u(r)\) is given by \(^{37}\)

\[
E_{el}[u(r)] = \int dr \left\{ \frac{\lambda + \mu}{2} \left( \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} \right)^2 + \mu \left( \left( \frac{\partial u_x}{\partial x} + \frac{\partial u_y}{\partial y} \right)^2 + \left( \frac{\partial u_y}{\partial x} - \frac{\partial u_x}{\partial y} \right)^2 \right) \right\}, \tag{1}
\]

where the integral extends over a moiré supercell. We use \(\lambda = 4.23\) eV/Å\(^2\) and \(\mu = 9.04\) eV/Å\(^2\) for the 2D elastic Lamé factors\(^{23}\) of graphene. \(E_{int}\) is given by the integral of the local interlayer interaction energy

\[
E_{int} = \int V[\delta(r), h(r)] dr, \tag{2}
\]

where \(\delta(r) = r - T_{\theta}r + u^{(2)}(r) - u^{(1)}(r)\) for a relaxed superlattice with \(T_{\theta}\) denoting the clockwise rotation by \(\theta\). The \(\tilde{u}^{(n)}(r)\) and \(h(r)\) have been expanded in Fourier series as

\[
u^{(n)}(r) = \sum_{G} \tilde{u}^{(n)}(G^{(s)}) e^{iG^{(s)} \cdot r} \tag{3}
\]

and

\[
h(r) = h_0 + \Delta h(r) = h_0 + \sum_{G^{(s)}} \Delta \tilde{h}(G^{(s)}) e^{iG^{(s)} \cdot r}, \tag{4}
\]

where the summation is over nonzero reciprocal lattice vectors \(G^{(s)}\) of the supercell and only \(|G^{(s)}| < 4|b_1^{(s)}|\) are used as \(\tilde{u}^{(n)}(r)\) and \(h(r)\) vary smoothly across the supercell. The average \(h(r)\) over the supercell is just \(h_0\). In the following, we have minimized the total energy functional for a fixed \(h_0\) with respect to \(u^{(n)}(r)\) and \(\Delta h(r)\). With the obtained \(E_{tot}\) as a function of \(h_0\), the perpendicular pressure applied to TBG can be obtained

\[
P = -\frac{1}{\Omega^{(s)}} \frac{dE_{tot}}{dh_0}, \tag{5}
\]

where \(\Omega^{(s)}\) is the supercell area and \(\Omega^{(s)} = \sqrt{3}a^2/[8\sin^2(\theta/2)]\) for the considered moiré superlattices. We have extended the method proposed by Nam and Koshino\(^{21}\) to relax TBG with a fixed \(h_0\) as detailed in the Appendix.

We find that the layers in TBG under high pressure are still corrugated and the corrugation height in each layer is just slightly decreased compared with the free structure, as shown in Figs. 1(a) and 1(c). In contrast, the in-plane relaxation is greatly enhanced by the high pressure with the AB-like and BA-like parts becoming larger, as seen in Figs. 1(b) and 1(c). For \(\theta = 1.248^\circ\), the highest elastic energy density increases by 156\% with pressure from 0.0 to 3.2 GPa. This is due to the decrease in \(h_0\) from 3.44 to 3.21 Å, as the potential energy of the AB-stacking part is much lower than that of the AA-stacking part for the smaller \(h_0\).

Figure 1(d) shows the variation of \(E_{tot}\) of TBG with \(h_0\) for \(\theta\) from 1° to 2°, from which the pressure \(P\) is calculated by Eq. (5). Then the \(h_0\) of systems with given \(\theta\) and \(P\) can be solved, as seen in Fig. 1(e). Under the same \(P\), \(h_0\) increases slowly with \(\theta\). With the same \(h_0\), \(P\) also increases with \(\theta\).

For TDBG, similar analysis as TBG shows that the displacement fields in the two bottom layers are opposite to those in the two top layers \((u(r)\) and \(u'(r)\), and the profile of the interlayer distance between the two bottom layers is the same as that between the two top layers \((\tilde{h}'(r))\), as shown in Fig. 1(f). The profile of the interlayer distance between the two middle layers is denoted by \(h(r)\). The total energy of a TDBG supercell is given by

\[
E_{tot} = 2E_{el}[u(r)] + 2E_{el}[u'(r)] + \int \left[ V[\delta(r), h(r)] + 2V[\delta'(r), h'(r)] \right] dr,
\]

where \(\delta(r) = r - T_{\theta}r + 2u(r), \delta'(r) = \delta_{AB} + u'(r) - u(r), h(r) = h_0 + \Delta h(r)\) and \(h'(r) = h_0 + \Delta h'(r)\). Here \(h_0\) and \(h_0'\) are the average values of \(h(r)\) and \(h'(r)\), respectively. The TDBG with given \(h_0\) and \(h_0'\) is also relaxed by solving Euler-Lagrange equations using the method similar to that for TBG. The perpendicular pressure applied to TDBG can be obtained

\[
\text{FIG. 2. (Color online). The band structures of relaxed (solid lines) TBG (a and b) and TDBG (c and d) under zero and a moderate pressure as well as the bands of rigid superlattices (dashed lines) with interlayer distances the same as the average ones of the relaxed superlattices.}
\]
1.67 GPa
1.09 GPa
0.50 GPa
0.50 GPa
1.67 GPa
|ψ|2 (10−4)
(a) (b) (c)
(d) (e)

FIG. 3. (Color online) The evolution of bands with pressure for TBG with θ = 1.248°. (a-c) The widths (a) of middle valence (W_v) and conduction bands (W_c), the gaps (b) on the electron (Δ_e) and hole (Δ_h) sides, and the energy (c) of the valence band state at Γ^(s) with respect to that at K^(s) under increasing pressure. The data of relaxed TBG are represented by solid lines and the dashed lines denote rigid superlattices with interlayer distances the same as the average ones of the relaxed structures. (d) The band structures of relaxed TBG (solid lines) under pressures of 0.50, 1.09 and 1.67 GPa and those of corresponding rigid structures (dashed lines). (e) The spatial distribution at the sublattice-A sites of the bottom layer of the probability density of the middle valence band state at Γ^(s) for relaxed TBG under pressures of 0.50 and 1.67 GPa.

With the relaxed structures of TBG and TDBG under pressure, their band structures can be obtained based on the tight-binding Hamiltonian. We will demonstrate below that relaxation is important to describe the pressure induced gap modulation and topological transitions in TBG and TDBG.

III. COMPARISON BETWEEN BAND STRUCTURES OF SYSTEMS WITH AND WITHOUT RELAXATION

To calculate band structures of relaxed TBG and TDBG under pressure, we extend the Hamiltonian of p_z orbitals for graphene bilayers proposed in Refs. [39,24] to take into account the effect of the pressure dependent in-plane structural deformation and out-of-plane atomic corrugation. The hopping between intralayer nearest neighbors with their distance d deviated from that of the pristine graphene d_0 = a/√3 is given by V_{ppπ}(d) = −V_{0ppπ}e^{−(d−d_0)/λ_π} with λ_π = 0.47 Å, V_{0ppπ} = 3.09 eV reproduces the experimental Fermi velocity v_F ≈ 1 x 10^6 m/s in the graphene layer. The expression of the interlayer hopping between sites with in-plane projection r
and out-of-plane projection \( h \) is

\[
V_{pp\sigma}(r, h) = V_{pp\sigma}^0 e^{-(h-h_0)/\lambda} e^{-(\sqrt{r^2+h^2}-h)/\lambda} \frac{\hbar^2}{r^2+h^2},
\]

where \( V_{pp\sigma}^0 = 0.453 \text{ eV}, h_0 = 3.38 \text{ Å}, \lambda = 0.58 \text{ Å}, \lambda = 0.27 \text{ Å}. \) All interlayer hopping terms with \( r \leq 5.0 \text{ Å} \) are included in the calculations. We note that these Hamiltonian parameters can reproduce the observed magic angle of free TBG.

Since TBG and TDBG with small \( \theta \) have large moiré supercells, their Hamiltonian can be diagonalized using the planewave-like basis functions. In this approach, the atomic positions of the rigid graphene lattice in each layer are used to label the hopping sites in the Hamiltonian. At a k-point \( k^{(s)} \) in the supercell BZ, a low-energy basis function for the sublattice \( \alpha \) (\( \alpha = \Lambda, B \) in layer \( n \) with momentum close to one Dirac point of the layer is defined as

\[
|n_\alpha, k^{(s)} + \mathbf{k}_0 + \mathbf{G}^{(s)}\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{r}_{n\alpha}} e^{i(k^{(s)} + \mathbf{k}_0 + \mathbf{G}^{(s)}) \cdot \mathbf{r}_{n\alpha}} |\mathbf{r}_{n\alpha}\rangle,
\]

where \( \mathbf{k}_0 \) is the center of one of the supercell BZs containing the Dirac points (\( K_\xi \) and \( K'_\xi \) with \( \xi = \pm 1 \) the valley index) of the fixed and twisted layers at their corners and \( \mathbf{r}_{n\alpha} \) is the rigid position of a sublattice-\( \alpha \) atom in layer \( n \). \( \mathbf{G}^{(s)} \) is a reciprocal lattice vector of the supercell and the used \( \mathbf{k}_0 \) can be seen in the schematic reciprocal lattice of a moiré superlattice in Fig. 7(a) of the Appendix. \( \mathbf{G}^{(s)} \) is a reciprocal lattice vector of the superlattice and is given by \( \mathbf{G}^{(s)} = j_1 \mathbf{b}_1^{(s)} + j_2 \mathbf{b}_2^{(s)} \) with small integers \( j_1 \) and \( j_2 \) typically in the range of \(-4 \sim 4 \). The Hamiltonian element between two basis functions is given by

\[
\langle n_\alpha, \mathbf{k}_1 | H | m_\beta, \mathbf{k}_2 \rangle = \frac{1}{N_0} \sum_{\mathbf{r}_{n\alpha} \in SC} \sum_{\mathbf{r}_{m\beta}} e^{-i\mathbf{k}_1 \cdot \mathbf{r}_{n\alpha} + i\mathbf{k}_2 \cdot \mathbf{r}_{m\beta}} \langle \mathbf{r}_{n\alpha} | H | \mathbf{r}_{m\beta} \rangle,
\]

where \( \mathbf{k}_j = \mathbf{k}^{(s)} + \mathbf{k}_0 + \mathbf{G}_j^{(s)} \) \( (j = 1, 2) \), the summation over \( \mathbf{r}_{n\alpha} \) is done in a supercell, \( N_0 \) is the number of graphene unit cells in one layer of the supercell, and only a small number of large hopping terms \( \langle \mathbf{r}_{n\alpha} | H | \mathbf{r}_{m\beta} \rangle \) are required for each \( \mathbf{r}_{n\alpha} \). Since the Hamiltonian between states from the two different valleys is negligible for large moiré superlattices, each obtained band can be characterized by the valley index \( \xi \), and this is an advantage of diagonalization of the Hamiltonian using the planewave-like basis functions.

Figure 2 shows the band structures of relaxed TBG and TDBG under zero and a moderate pressure as well as the bands of rigid superlattices with interlayer distances the same as the average ones of the relaxed superlattices. The bands without relaxation could give similar band widths as those with relaxation for both TBG and TDBG. However, the energy gaps above and below the flat bands are significantly underestimated without relaxation for both TBG and TDBG, and the gaps at the neutrality point of relaxed TDBG under pressure are rather different from those without relaxation. Moreover, around the critical pressure with the minimum band width of the middle valence bands, the dispersions of the middle bands without relaxation are also different from those with relaxation for both systems. It is also noted that the middle valence and conduction bands touch each other in relaxed TDBG under a moderate pressure, which may indicate the occurrence of a topological transition. Therefore, relaxation must be considered to describe the pressure induced gap modulation and topological transitions in TBG and TDBG. In comparison, previous studies only focused on pressure induced narrowing of bands in TBG as full relaxation was not considered there.

IV. GAP MODULATION AND EVOLUTION OF BANDS IN TBG BY PRESSURE

At the first magic angle of around 1.1°, the middle bands at \( E_F \) in free TBG flatten. For \( \theta \) larger than the magic angle with wider middle bands, the perpendicular pressure narrows down these bands. We find that the pressure can also tune the band gaps on the electron and

![Figure 4](image-url)
When \( P \) is larger than the critical pressures, \( \Delta_c \) and \( \Delta_h \) decrease with \( P \) and become closed under \( P \approx 2.8 \) GPa. In contrast, the \( \Delta_e \) and \( \Delta_h \) of rigid superlattices are already closed with \( h_0 \) around that of the relaxed structure under \( P \approx 1.0 \) GPa.

Under pressure between \( P_{c1} \) and \( P_{c2} \), we find that the nearly flat valence and conduction bands become overlapped as indicated by the higher energy of the valence band state at \( \Gamma^{(e)} \) than the degenerate valence and conduction states at \( K^{(e)} \), as shown in Figs. 3(c) and 3(d). The varying band structures with \( P \) can also be seen in video 1 of the Supplemental Material (SM)\(^\text{1}\). With pressure increasing from \( P_{c1} \) to \( P_{c2} \), the flat valence bands around \( \Gamma^{(e)} \) move up while the flat conduction bands around \( \Gamma^{(s)} \) move down, and they switch at \( P = 1.09 \) GPa. Such evolution of band dispersions may suggest a transition of the electronic structure. It is noted that under pressure beyond \( P_{c1} \) and \( P_{c2} \), the bands around \( E_F \) are just those with linear dispersions around \( K^{(s)} \) (see Fig. 3(d)), so the density of state (DOS) at \( E_F \) is zero. However, when the flat valence and conduction bands overlap, the DOS at \( E_F \) can be very large. In addition, the middle bands under a large pressure may be similar to those under a small pressure (see Fig. 3(d)), while the wavefunctions of states around \( K^{(s)} \) are distinct from each other, especially the positions with highest probability density, as shown in Fig. 3(e).

Since the electronic properties of TBGs are sensitive to \( \theta \), the effect of pressure on tuning their band structures also depends closely on \( \theta \). \( P_{c1} \) and \( P_{c2} \) increase rapidly with \( \theta \), and only for \( \theta \) smaller than about 1.54° \( W_V \) can reach the minimum under \( P \leq 3.2 \) GPa, as shown in Fig. 4(a). In addition, the pressure range \( P_{c2} − P_{c1} \) with

![Graphs](image_url)
FIG. 6. (Color online). (a) The Chern numbers of the middle valence ($C_{V}$) and conduction ($C_{C}$) bands for free TDBGs with varying $\theta$ from 1.0° to 2.0°. (b-d) The $\Delta_{c}$ (b), $\Delta_{h}$ (c) and $W_{V}$ (d) as functions of pressure for strictly periodic TDBG with $\theta$ from 1.0° to 2.0°. The black squares in (b) show the $P_{c1}$ and $P_{c2}$ for each $\theta$. The black areas in (c) denote the pressure range with closed $\Delta_{0}$.

overlapped valence and conduction bands increases with $\theta$. For $\theta$ larger than about 1.3°, $\Delta_{v}$ and $\Delta_{h}$ remain open under $P \leq 3.2$ GPa, and the maximum $\Delta_{h}$ increases with $\theta$ though $\Delta_{h}$ of free systems decreases with $\theta$, as shown in Fig. 4(b). With $\theta$ from 1.08° to 1.54°, the maximum $\Delta_{h}$ increases from 42 to 59 meV.

V. TOPOLOGICAL AND ELECTRONIC TRANSITIONS IN TDBG UNDER PRESSURE

Free AB-BA TDBGs were found to be valley Hall insulators with a gap ($\Delta_{0}$) opened at the charge neutrality point31,32. We observe topological transitions in TDBG induced by the perpendicular pressure with the Chern number of the middle valence band transferred to the middle conduction band in a valley. Electronic transitions with $\Delta_{0}$ closed and reopened also occur under pressure.

The topological properties of TDBG are characterized by Chern numbers of the bands in each valley, which are defined as $C_{n} = 1/2\pi \int F_{n,k^{(s)}} d^{2}k^{(s)}$, where $F_{n,k^{(s)}}$ is the Berry curvature of the state at $k^{(s)}$ in the band with index $n$.42 Due to the absence of inversion symmetry, states in TDBG can have nonzero Berry curvature. We have calculated $F_{n,k^{(s)}}$ of a relaxed superlattice by42

$$F_{n,k^{(s)}} = -2 \text{Im} \sum_{n' \neq n} \langle n,k^{(s)} | \frac{\partial H_{k^{(s)}}}{\partial k^{(s)}} | n',k^{(s)} \rangle \langle n',k^{(s)} | \frac{\partial H_{k^{(s)}}}{\partial k^{(s)}} | n,k^{(s)} \rangle (\varepsilon_{nk^{(s)}} - \varepsilon_{n'k^{(s)}})^{2},$$

(10)

where the band state $| n,k^{(s)} \rangle$ and $\partial H_{k^{(s)}}/\partial k^{(s)} (\gamma = x, y)$ are represented in the Bloch basis $| \nu k^{(s)} \rangle = 1/\sqrt{N_{s}} \sum_{R^{(s)}} e^{i k^{(s)} \cdot (R^{(s)} + r_{\nu})} | R^{(s)} + r_{\nu} \rangle$ with $r_{\nu}$ denoting the rigid position of a site in the supercell and $R^{(s)}$ a superlattice vector. The band state $| nk^{(s)} \rangle$ expanded as $\sum_{\mu s} G^{(s)}(\psi_{\mu s},G^{(s)} | \mu s, k^{(s)} + k_{0} + G^{(s)} \rangle$ can be represented in the basis of $| \nu k \rangle$ as

$$\sum_{\nu} \left( \sum_{\mu s} G^{(s)}(\psi_{\mu s},G^{(s)} | \mu s, k^{(s)} \rangle 1/\sqrt{N_{s}} e^{i k^{(s)} \cdot (k_{0} + G^{(s)} \cdot r_{\nu})} | \mu k \rangle \right) | \nu k \rangle.$$  

(11)

The matrix representation of $\partial H_{k^{(s)}}/\partial k^{(s)}$ is

$$\left( \frac{\partial H_{k^{(s)}}}{\partial k_{\gamma}^{(s)}} \right)_{\mu \nu} = \sum_{R^{(s)}} e^{i k^{(s)} \cdot (R^{(s)} + r_{\nu} - r_{\mu})} \langle R^{(s)} + r_{\nu} - r_{\mu} | H | R^{(s)} + r_{\nu} \rangle$$

$$\langle r_{\mu} | H | R^{(s)} + r_{\nu} \rangle$$

(11)

with $\langle r_{\mu} | H | R^{(s)} + r_{\nu} \rangle$ the Hamiltonian between sites at $r_{\mu}$ and $R^{(s)} + r_{\nu}$.

We first show the evolution of topological and electronic properties with pressure for TDBG with $\theta = 1.538°$, whose total Chern number of the valence bands is 2 in the $\xi = +$ valley31.

Since the topological transition occurs when the middle valence and conduction bands touch each other, the minimum direct gap ($\Delta_{d}$) is calculated to identify the transitions. The Chern numbers of the middle valence and conduction bands of the free system in the $\xi = +$ valley are 3 and -1, respectively. Under the critical pressures $P_{c1} = 1.03$ GPa and $P_{c2} = 1.51$ GPa, the valence and conduction bands touch and their Chern numbers are transferred to each other, as shown in Figs. 5(a), 5(b), 5(f), and 5(g). Then under pressure between $P_{c1}$ and $P_{c2}$, the total Chern number of the valence bands becomes -2. In the $\xi = -$ valley, the Chern number of each band is just the opposite of that in the $\xi = +$ valley. For the rigid superlattice, the valence and conduction bands only touch under a very high pressure, as shown in Fig. 5(a).

Besides topological properties, $\Delta_{0}$ and the gaps on the electron and hole sides ($\Delta_{c}$ and $\Delta_{h}$) are also tuned by the pressure. Under pressure from 0 to $P_{c1}$, $\Delta_{0}$ decreases to zero. $\Delta_{0}$ becomes negative with overlapped valence and conduction bands under pressure slightly larger than $P_{c2}$, the system thus becomes metallic in this pressure range. Then $\Delta_{0}$ becomes positive under increasing pressure, as shown in Figs. 5(a) and 5(h). The varying band structures with pressure can also be seen in video 2 of the Supplemental Material (SM).41 The trend of $\Delta_{c}$ and $\Delta_{h}$ with pressure is similar to that of TBG. Under pressure
of about 3.2 GPa, the middle valence bands touch the lower valence bands, while \( \Delta_0 \) becomes reopened under increasing pressure, as shown in Figs. 5(c) and 5(h).

The trend of \( W_V \) and \( W_C \) with pressure is shown in Fig. 5(d). At about \( P_{c1} \), \( W_V \) reaches the minimum value of 10 meV, while \( W_C \) only becomes minimum under a much higher pressure of 1.91 GPa. In contrast to the increasing \( W_V \) beyond the critical pressures in TBG, the \( W_V \) of TDBG can become small under high pressure.

Similar topological transitions as those under increasing pressure also occur in free TDBGs with varying \( \theta \) from 2.0° to 1.0°, as shown in Fig. 6(a). For \( \theta \) from 1.248° to 1.890°, topological transitions can be induced by pressure smaller than 3.2 GPa, as shown in Fig. 6(b). In particular, the critical pressures are close to 0 GPa for \( \theta = 1.248° \). For \( \theta \) smaller than 1.248°, no topological transitions are observed. The semiconductor to metal transition under pressure can be observed for \( \theta \gg 1.203° \), while \( \Delta_0 \) remains positive and decreases slowly for a smaller \( \theta \), as shown in Fig. 6(c). Figure 6(d) shows variations of \( W_V \) with pressure for different \( \theta \). For small \( \theta \), the profile of \( W_V \) can be rather complicated. We note that the pressure induced minimum \( W_V \) tends to decrease with \( \theta \) from 2.0° toward 1.0°. For \( \theta = 1.050° \), the minimum \( W_V \) is 3.89 meV, while it becomes 7.65 meV for \( \theta = 1.538° \).

VI. SUMMARY AND CONCLUSIONS

An approach has been proposed to fully relax TBG and TDBG under perpendicular pressure with both in-plane structural deformation and out-of-plane corrugation considered. The moiré superlattices with fixed average interlayer distances \( h_0 \) are first relaxed and the pressure is obtained through the variation of the total energy with \( h_0 \). We find that the in-plane relaxation is greatly enhanced by the high pressure in TBG and in the middle layers of TDBG, while the corrugation height in each layer is just slightly decreased by high pressure. The electronic and topological properties of relaxed superlattices under pressure have been explored by diagonalizing the tight-binding Hamiltonian using the plane-wave-like basis functions, so that each band can be conveniently characterized by the valley index \( \xi \). The comparison between band structures with and without relaxation demonstrates that relaxation is required to describe the pressure dependent band gaps and detailed band dispersions. Only band widths without relaxation are similar to those of relaxed systems.

In TBG, the gaps on the electron and hole sides reach maximum values around the critical pressures \( P_{c1} \) and \( P_{c2} \) with narrowest middle bands and become closed only under a very high pressure. The nearly flat valence and conduction bands become overlapped between \( P_{c1} \) and \( P_{c2} \), and the wavefunctions of band states under higher pressures than \( P_{c2} \) exhibit rather different spatial distributions from those under smaller pressures. Topological transitions occur in AB-BA TDBGs under pressure with the middle valence and conduction bands in one valley touching and their Chern numbers transferred to each other. The pressure can also tune the gap \( \Delta_0 \) at the neutrality point of TDBG, which becomes closed for a pressure range and reopened under higher pressure. The behavior of the electronic structure of superlattices under pressure is sensitive to the twist angle \( \theta \) with the critical pressures generally increasing with \( \theta \). Our study thus provides a systematic description of electronic and topological properties of fully relaxed TBG and TDBG under pressure, and the proposed approach to relax moiré superlattices under pressure can be applied to various twisted bilayers and multilayers under pressure including heterostructures.

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APPENDIX

A. Mathematical formulation of the moiré superlattices

In the TBG moiré superlattice, the top layer has been rotated by \( \theta \) counterclockwise with respect to the fixed bottom layer, and one sublattice-A atom in the bottom layer is placed just below one sublattice-A atom in the top layer at the origin. In the considered AB-BA TDBG, the top BA-stacked bilayer is twisted with the bottom AB-stacked bilayer fixed, and one sublattice-A atom in each of the two middle layers is placed at the origin.

The unit cell of a fixed layer is spanned by the basis vectors \( \mathbf{a}_1 = a(\sqrt{3}/2, -1/2) \) and \( \mathbf{a}_2 = a(\sqrt{3}/2, 1/2) \), where \( a = 2.46 \) Å is the lattice constant of graphene. Then the basis vectors of a twisted layer become \( \mathbf{a}_1' = T_0 \mathbf{a}_j \) \((j = 1, 2)\), where \( T_0 \) denotes the rotation. The sublattice A atom in a unit cell of each layer is located at the origin of the cell, and the positions of the sublattice B atoms in the unit cell of the fixed and twisted layers are \((\mathbf{a}_1 + \mathbf{a}_2)/3\) and \((\mathbf{a}_1' + \mathbf{a}_2')/3\), respectively.

We consider strictly periodic moiré superlattices with basis vectors \( \mathbf{a}_1^{(s)} = N \mathbf{a}_1 + (N + 1) \mathbf{a}_2 \) and \( \mathbf{a}_2^{(s)} = -(N + 1) \mathbf{a}_1 + (2N + 1) \mathbf{a}_2 \), where \( N \) is an integer. The relation between \( \theta \) and \( N \) can be expressed as \( \cos \theta = (1 + 6N + 6N^2)/(2 + 6N + 6N^2) \). For \( N \) from 16 to 32, \( \theta \) takes values from 2.005° to 1.018°. The reciprocal lattice of such a moiré superlattice is spanned by the vectors \( \mathbf{b}_1 = \mathbf{b}_1' - \mathbf{b}_2' \), and \( \mathbf{b}_2'' = (\mathbf{b}_1' + \mathbf{b}_2') - (\mathbf{b}_1 + \mathbf{b}_2) \), where \( \mathbf{b}_1 \) and
Large hexagons are BZs of the fixed layers (blue), spanned by \( \mathbf{b}_1^{(s)} \) and \( \mathbf{b}_2^{(s)} \). Large hexagons are BZs of the twisted layers (red), spanned by \( \mathbf{b}_1' \) and \( \mathbf{b}_2' \). The Dirac points of the fixed \((K_\xi)\) and twisted \((K'_\xi)\) layers are located at corners of the supercell BZs, with \( \xi = \pm 1 \) the valley index. \( \mathbf{k}_0 \) is the center of one of the supercell BZs containing \( K_\xi \) and \( K'_\xi \) at their corners. (b) The high-symmetry \( k \)-points in the supercell BZ.

\( \mathbf{b}_i' \) with \( i = 1, 2 \) are reciprocal lattice vectors of the fixed and twisted layers, respectively, as shown in Fig. 7(a). The Dirac points of the fixed and twisted layers \((K_\xi, K'_\xi)\) are just located at two adjacent corners of a supercell Brillouin zone (BZ), whose center is denoted by \( \mathbf{k}_0 \). The \( \mathbf{k}_0 \) insider the large BZs in Fig. 7(a) is just \(-N\mathbf{b}_2^{(s)}\). The high-symmetry \( k \)-points in the supercell BZ are labeled in Fig. 7(b).

In a large moiré superlattice, the local lattice structure around a position \( \mathbf{r} \) is similar to that of a shifted graphene bilayer, where the shift vector \( \mathbf{\delta} \) is defined as the in-plane displacement vector from an atom in the bottom layer to the atom in the top layer which sits just above the bottom-layer atom for the AA stacked bilayer with \( \mathbf{\delta} = 0 \). Then we find that the local \( \mathbf{\delta} \) between the twist and fixed layers at \( \mathbf{r} \) in the superlattice can be taken as \( \mathbf{\delta} = (I - T_{-\theta})\mathbf{r} \). The \( \mathbf{\delta} \) at \( \mathbf{r} = \mathbf{a}_1^{(s)} \) and \( \mathbf{a}_2^{(s)} \) are just \( \mathbf{a}_2 - \mathbf{a}_1 \) and \(-\mathbf{a}_1 \), respectively, which is consistent with the AA stacking at the superlattice vectors of TBG.

B. Dependence of energies of shifted graphene bilayers on the interlayer distance

The dependence of \( \bar{V}_0 \) and \( \bar{V} \) on \( h \) can be obtained from the variation of \( V \) with \( h \) for the AA- and AB-stacked bilayers \((V_{AA}(h) \text{ and } V_{AB}(h))\) with \( \mathbf{\delta}_{AA} = 0 \) and \( \mathbf{\delta}_{AB} = (\mathbf{a}_1 + \mathbf{a}_2)/3 \) as \( V_{AA}(h) = |V_{AA}(h) + 2V_{AB}(h)|/3 \) and \( \bar{V}(h) = 2|V_{AA}(h) - V_{AB}(h)|/9 \). By \textit{ab initio} calculations of energies of bilayers, \( V_{AA}(h) \) and \( V_{AB}(h) \) are fitted as \( V_{AA}(h) = 0.0322 e^{-(h-3.2)/0.316} - (1.443/h)^4 + 0.0205 \) and \( V_{AB}(h) = 0.0259 e^{-(h-3.2)/0.344} - (1.488/h)^4 + 0.0222 \) with \( V \) and \( h \) in units of eV/Å² and Å, respectively, where the first terms represent the short-range repulsion and the second terms represent the long-range van der Waals interaction. From \( V_{AA}(h) \) and \( V_{AB}(h) \), the estimated corrugation height in free TBG is about 0.12 Å, which is slightly smaller than that of about 0.14 Å from direct \textit{ab initio} relaxation of magic-angle TBG.

We use \textit{ab initio} density functional theory (DFT) as implemented in the VASP code to calculate energies of shifted graphene bilayers with different interlayer distances. We use projector augmented-wave pseudopotentials and the SCAN+RVV10 exchange correlation functional, which provides a proper description of the van der Waals interaction. We use 800 eV as the kinetic energy cutoff for the plane-wave basis and the tolerance for the energy convergence is \( 10^{-6} \) eV. The BZ sampling is done using a \( 36 \times 36 \times 1 \) Monkhorst-Pack grid, and vacuums in the \( z \) direction are larger than 17 Å.

C. Relaxation of TBG with a fixed average interlayer distance by Euler-Lagrange equations

We have extended the method proposed by Nam and Koshino to relax TBG with a fixed \( h_0 \). The \( E_{el} = \sum_{n=1}^{2} E_{el}(u^{(n)}) + E_{int} \) of a supercell can be expressed as \( E_{tot} = \int L(u^{(1)}, u^{(2)}, h_0 + \Delta h) \). The minimization of \( E_{tot} \) as a functional of \( u^{(n)} \) and \( \Delta h \) leads to a series of Euler-Lagrange equations

\[
\frac{\partial}{\partial x} \left[ \frac{\partial L}{\partial (\partial u^{(n)} / \partial x)} \right] + \frac{\partial}{\partial y} \left[ \frac{\partial L}{\partial (\partial u^{(n)} / \partial y)} \right] - \frac{\partial L}{\partial u^{(n)}} = 0 \quad (A1)
\]

and

\[
\frac{\partial L}{\partial \Delta h} = \frac{\partial V(\mathbf{\delta}, h_0 + \Delta h)}{\partial h_0} = 0, \quad (A2)
\]

where \( n = 1, 2 \) and \( \nu = x, y \). With \( \partial L/\partial u^{(2)} = -\partial L/\partial u^{(1)} = \partial V/\partial \mathbf{\delta} \) expanded as \( \sum_{G^{(s)}} \mathbf{f}(G^{(s)}) e^{iG^{(s)} \cdot \mathbf{r}} \), substitution of the Fourier expansion of \( u^{(n)}(\mathbf{r}) \) into Eq. (4) leads to

\[
\left( (\lambda + 2\mu) \mathbf{q}_x^2 + \mu \mathbf{q}_y^2 \right) \left( (\lambda + \mu) \mathbf{q}_x \mathbf{q}_y \right) \left( \frac{\partial u^{(n)}(\mathbf{q})}{\partial \mathbf{q}} \right) = \left( \frac{\partial u^{(n)}(\mathbf{q})}{\partial \mathbf{q}} \right) \left( \frac{\partial S_{G^{(s)}}}{\partial \mathbf{q}} \right) \quad (A3)
\]

where \( \mathbf{q} \) takes each \( G^{(s)} \). It can be inferred from this equation that \( u^{(1)}(\mathbf{r}) = -u^{(2)}(\mathbf{r}) \). Expanding \( \partial V(\mathbf{\delta}, h_0 + \Delta h)/\partial \Delta h \) as \( \sum_{G^{(s)}} \tilde{g}(G^{(s)}) e^{iG^{(s)} \cdot \mathbf{r}} \), Eq. (7) gives rise to

\[
\tilde{g}(G^{(s)}) = 0. \quad (A4)
\]

In view of the real-valued fields and the 120°-rotation symmetry of the superlattice, \( \mathbf{u}^{(s)}(3\gamma_3, G^{(s)}) = \)
\(C_{2g} \tilde{u}(n)(G^{(s)}) - \tilde{u}(n)(-G^{(s)}) = \tilde{u}(n)(G^{(s)})\),
\(\Delta h(C_{2g} G^{(s)}) = \Delta \tilde{h}(G^{(s)})\),
\(\Delta h(-G^{(s)}) = \Delta \tilde{h}(G^{(s)})\).
So only Fourier components for one sixth of the considered \(G^{(s)}\) are independent in these equations.
We have solved Eqs. (8) and (9) self consistently to obtain converged \(\tilde{u}^{(0)}(G^{(s)})\) and \(\Delta \tilde{h}(G^{(s)})\) using zero as their initial values.
During the self-consistent iteration, all \(\Delta \tilde{h}(G^{(s)})\) remain real so \(\Delta h(r)\) can be expressed as
\(\sum_{G^{(s)}} \Delta \tilde{h}(G^{(s)}) \cos(G^{(s)} \cdot r)\), which indicates that the AA-stacked part at \(r = 0\) has the largest \(h\). Since \(\partial V / \partial \Re\) remains an odd function of \(r\), all \(\tilde{u}^{(n)}(G^{(s)})\) are purely imaginary and \(\tilde{u}^{(n)}(r)\) can be expressed as
\(-\sum_{G^{(s)}} \Im[\tilde{u}^{(n)}(G^{(s)})] \sin(G^{(s)} \cdot r)\).

* E-mail: xmlin@zjut.edu.cn

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