Effective band structures of multilayer graphene quasicrystals

Guodong Yu,1,2 Zewen Wu,1 Zhen Zhan,1 Mikhail I. Katsnelson,2 and Shengjun Yuan1,2,*

1Key Laboratory of Artificial Micro- and Nano-structures of Ministry of Education and School of Physics and Technology, Wuhan University, Wuhan 430072, China
2Institute for Molecules and Materials, Radboud University, Heijendaalseweg 135, NL-6525 AJ Nijmegen, Netherlands

In this paper, the electronic properties of several multilayer graphene quasicrystals are studied by means of the tight-binding method. The multilayer graphene quasicrystals are stacked in AA, AB or their combined orders, and the quasi-periodicity is introduced by rotating at least one layer by 30°. The periodic approximants of several multilayer graphene quasicrystals are proposed, and their effective band structures are derived by unfolding the band structures of approximants. The 30° twisted interface separates the system into subsystems, and our results show that these subsystems are coupled by the interlayer interactions crossing the interface but will retain their own band structures at low-energies. The band structures of all the subsystems with the same orientation are combined at the corners of the Brillouin zone, and scattered into their mirror-symmetric points inside the Brillouin zone, with weak and anisotropic spectral functions. Such mirror symmetry and combination effects are robust even if an external electric field is applied. Our results suggest that the 30° twisted quasicrystal interface can be used to engineer the band structures of multilayer graphene or other layered two-dimensional materials.

I. INTRODUCTION

In theory, a twisted bilayer graphene (TBG) can transform from a crystalline (commensurate configuration) to quasi-crystalline (incommensurate configuration) depending on the twist angle. The commensurate configuration with small twist angle as a model system of strongly correlated electrons has drawn much attention due to the novel electronic properties, such as the flat band[1–3], unconventional superconductivity[4–6], correlated insulator phases[7–11], etc. Recently, the 30° TBG (decagonal bilayer graphene quasicrystal), an incommensurate configuration, has been grown successfully on H-SiC(0001)[12], Pt(111)[13], and Cu-Ni(111)[14] surfaces. As the first two dimensional quasicrystal based on graphene, 30° TBG has received increasing attention in both experiment and theory[15–17]. A method to grow high-quality 30° TBG epitaxially on SiC using borazine as a surfactant has been proposed[18]. A number of Dirac cones, especially the mirror-symmetric ones, were observed by the angle resolved photoemission spectroscopy (ARPES) measurement[18]. The quasicrystalline order in 30° TBG can induce unique localization of electrons without any extrinsic disorder[18,19]. All these peculiar properties make 30° TBG much different from graphene monolayer, although some studies claimed that TBG with large twist angle (> 15°) should behave like two decoupled graphene monolayers[20,21].

The twisted multilayer graphenes (TMGs) consist of several subsystems with different orientations, and each subsystem can be graphene monolayer or multilayer. Comparing with the TBG consisting of only graphene monolayer, the electronic properties of TMGs can be tuned by not only twist angle but also stacking order in subsystems and even electric field. Recently, one kind of TMG, twisted double bilayer graphene (TDBG) consisting of two AB-stacked bilayers, has received much attention especially on the properties[22,23] associated with the strongly correlated electrons in the electrically tunable flat band[24,25], such as the superconductivity, magnetic phase transition, correlated insulating state, etc. Besides, the AB-BA twisted double bilayer graphene under the electric field was found to be a valley Hall insulator[25]. A generic TMG, M-layer graphene on top of N-layer graphene with a twist angle, possesses two topologically nontrivial flat bands, which exhibit a Chern-number hierarchy[26]. Similar to the 30° TBG, 30° TMGs, namely multilayer graphene quasicrystals (MGQCs), are expected to possess some striking properties. More importantly, the successful fabrication of 30° TBG and accurate determination of some key structural parameters (such as twist angle, stacking order and interlayer spacing) in experiment[27,28] ensure the realization of MGQCs in the near future. Therefore, in this paper, we study the effect of the stacking order and electric field on the electronic properties of several MGQCs.

The approximant method is adopt to study the electronic properties of these MGQCs. The method to construct the approximant of 30° TBG[27] still apply to MGQCs. The proposed approximants can be confirmed by comparing their electronic properties, such as the density of states (DOS). So it is necessary to calculate accurately the electronic properties of these MGQCs. However to calculate the electronic properties of a quasicrystal is a big challenge because the band theory is invalid here due to the lack of the translational symmetry in the system. In order to solve the problem, it is a natural idea to simulate these MGQCs by using the large enough round disks, which ensures that the edge states have minor influence on the electronic properties so that they can be ignored safely. In this paper, the round disks with ten million atoms for all MGQCs are adopted. To calculate such big systems is obviously beyond the abilities of density functional theory and even the diagonalization of
FIG. 1: (a) The multilayer graphene quasicrystals (side and top views) with the point group in bracket. They are named by the sequence of layers from bottom to top. $\tilde{A}$ (red) and $\tilde{B}$ (orange) layers are obtained by rotating $A$ (black) and $B$ (grey) layers by $30^{\circ}$, respectively, so the armchair direction of $A$ and $B$ layers is along the zigzag direction of $\tilde{A}$ and $\tilde{B}$ layers. (b) The Brillouin zones of the $0^{\circ}$- and $30^{\circ}$-subsystems, which are plotted in black and red, respectively. Six $\tilde{K}_1$ and six $K_1$ represented by red and black dots inside the Brillouin zones are the mirror-symmetric points of $K$ (and $K'$) and $\tilde{K}$ (and $\tilde{K}'$) points, respectively. (c) Scattering path from $K'$ to $\tilde{K}_1$. (d) The elementary unit cell of 15/26 approximant of $BA\tilde{A}$. The rotation center is at the four corners. For the 15/26 approximants of all multilayer graphene quasicrystals, the $\tilde{A}$ and $B$ layers are slightly compressed, which makes the lattice constant change from $a = 2.456$ Å of a pristine graphene to $\tilde{a} = 2.454$ Å.

the tight-binding Hamiltonian due to the enormous CPU time and memory costs. We will use the tight-binding propagation method (TBPM) to overcome such computational difficulties. The TBPM is based on the numerical solution of time-dependent Schrödinger equation without any diagonalization, and both memory and CPU costs scale linearly with the system size.

II. METHODS

A. Structures

Each MGQC consists of the $0^{\circ}$- and $30^{\circ}$-subsystems with $0^{\circ}$ and $30^{\circ}$ standing for the orientations. In this paper, only monolayer, AA or AB stacked bilayers are considered as the subsystem. Besides, the tilde notation is used to distinguish the layers in $30^{\circ}$-subsystems ($\tilde{A}$ and $\tilde{B}$) from the layers ($A$ and $B$) in $0^{\circ}$-subsystems. The schematic diagrams of the MGQCs under study are given in Fig. 1(a). They are named according to the sequence of the layers from bottom to top. $\tilde{A}$ (red) and $\tilde{B}$ (orange) layers are obtained by rotating $A$ (black) and $B$ (grey) layers by $30^{\circ}$, respectively, so the armchair direction of $A$ and $B$ layers is along the zigzag direction of $\tilde{A}$ and $\tilde{B}$ layers. (b) The Brillouin zones of the $0^{\circ}$- and $30^{\circ}$-subsystems, which are plotted in black and red, respectively. Six $\tilde{K}_1$ and six $K_1$ represented by red and black dots inside the Brillouin zones are the mirror-symmetric points of $K$ (and $K'$) and $\tilde{K}$ (and $\tilde{K}'$) points, respectively. (c) Scattering path from $K'$ to $\tilde{K}_1$. (d) The elementary unit cell of 15/26 approximant of $BA\tilde{A}$. The rotation center is at the four corners. For the 15/26 approximants of all multilayer graphene quasicrystals, the $\tilde{A}$ and $B$ layers are slightly compressed, which makes the lattice constant change from $a = 2.456$ Å of a pristine graphene to $\tilde{a} = 2.454$ Å.
which are plotted in black and red, respectively. For the MGQCs in Fig. 1(a), the first five structures consist of just one 0°-subsystem and one 30°-subsystem, which are divided by the AÅ stacking interface. But the last one is composed of two 0°-subsystems and one 30°-subsystem.

B. Approximants

In order to calculate the electronic properties of these MGQCs, we need to construct the qualified periodic approximants to reproduce accurately the electronic properties of the MGQCs. Then, the EBS corresponding to the primitive unit cell of each layer can be derived by unfolding the band structure of the approximant. The approximant method has been widely used to understand the physics of a quasicrystal. In this paper, we follow the procedure proposed for 30° TBG to construct the approximants of all MGQCs. That is, A and B layers keep the lattice constant of a pristine graphene $a = 2.456$ Å, but A and B layers are slightly compressed with the lattice constant changing to be $\tilde{a} = 2.454$ Å, which makes the layers in 0°- and 30°-subsystems the commensurate lattices. The corresponding periodic pattern is used as the approximant, which is named as 15/26 approximant TBG. Our results (shown below) indicate that the 15/26 approximants can reproduce the DOS of these MGQCs accurately. Therefore, the EBSs derived by unfolding the band structures of these 15/26 approximants can be used to further investigate the electronic properties of the MGQCs.

C. Tight-binding model

All MGQCs are simulated by the tight-binding approximation based on $p_z$ orbitals, where the hopping energy between site $i$ and $j$ is

$$t_{ij} = n^2V_{pp\sigma}(|r_{ij}|) + (1 - n^2)V_{pp\pi}(|r_{ij}|).$$

(1)

Here, $n$ is the direction cosine of relative position vector $r_{ij}$ with respect to $e_z$. The Slater and Koster parameters $V_{pp\sigma}$ and $V_{pp\pi}$ follow the form:

$$V_{pp\sigma}(|r_{ij}|) = -\gamma_0e^{2.218(b-|r_{ij}|)}F_c(|r_{ij}|),$$

(2)

$$V_{pp\pi}(|r_{ij}|) = \gamma_1e^{2.218(h-|r_{ij}|)}F_c(|r_{ij}|).$$

(3)

The interlayer distance $h$ and nearest carbon-carbon distance $b$ are 3.349 and 1.418 Å, respectively. $\gamma_0$ and $\gamma_1$ are 3.12 and 0.48 eV, respectively, which fit well the experimental results of 30° TBG. $F_c(r)$ is a smooth function

$$F_c(r) = (1 + e^{(r - 0.265)/3})^{-1}.$$ 

(4)

This tight-binding model has been proved by comparing results with several experiments. It has been implemented in our home-made code of tight-binding propagation simulator (Tipsi) and reproduces successfully experimentally observed pseudo-magnetic fields in low-angle twisted bilayer graphene.

D. Density of states

The DOS of the MGQCs and the 15/26 approximants are calculated by TBPM, where a random superposition of the $p_z$ orbitals at all sites is used as the initial state $|\phi_0\rangle$ with $\langle \phi_0 | \phi_0 \rangle = 1$. DOS is calculated as Fourier transform of the time-dependent correlation function

$$d(\epsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\epsilon\tau} \langle \phi_0 | e^{-iH\tau/\hbar} | \phi_0 \rangle \, d\tau. \quad (5)$$

The MGQCs simulated by a huge enough round disk containing more than 10 million atoms are calculated in TBPM. In such large enough system, the edge states have a negligible effect to the electronic properties. Similarly, the elementary unit cell of the 15/26 approximants is repeated to obtain a supercell which contains also 10 million atoms. Moreover, the open and periodic boundary conditions are applied to the round disk samples of the MGQCs and the approximants, respectively. In fact, in such huge systems, only one initial state is enough to obtain the accurate DOS.

E. Effective band structure

First of all, the spectral function at wavevector $k$ and energy $\epsilon$ can be calculated by

$$A(k, \epsilon) = \sum_{l k_{SC}} P_{l k_{SC}}(k) \delta(\epsilon - \epsilon_{l k_{SC}}). \quad (6)$$

where $\epsilon_{l k_{SC}}$ is the energy for $l^{th}$ band at wavevector $k_{SC}$ for the approximant. Actually, only one $k_{SC}$, namely $k_{SC} = k + G$ being $G$ the reciprocal lattice vector of the approximant, contributes to the spectral function. The spectral weight is defined as

$$P_{l k_{SC}}(k) = \sum_{s} \sum_{i} \left| \langle \psi^{PC}_{i k} | \Psi_{l k_{SC}}^{S} \rangle \right|^2 = \sum_{s} P_{l k_{SC}}^{s}(k), \quad (7)$$

where $\left| \psi^{PC}_{i k} \right>$ and $\left| \Psi_{l k_{SC}}^{S} \right>$ are the eigenstates of layer $s$ and the approximant, respectively. Under the tight-binding method, the spectral weight contributed from layer $s$ can be described by

$$P_{l k_{SC}}^{s}(k) = \frac{1}{n_s} \sum_{\alpha} \sum_{l_s l_s'} e^{i(k - l_s l_s')U_{l s l_s'}^{l_{SC}^{\alpha} l_{SC}^{\alpha}}}, \quad (8)$$

Here, $n_s$ is the number of primitive unit cell of layer $s$ in one elementary unit cell of the approximant. $U_{l s l_s'}^{l_{SC}^{\alpha} l_{SC}^{\alpha}}$ is the
projection of \(|\Psi_{k_{SC}}^{\alpha}\rangle\) (the eigenstate of the approximant) on \(|k_{SC}l_{x}\alpha\rangle\) (the Bloch basis function of approximant), which can be constructed by all atomic orbitals

\[
|k_{SC}l_{x}\alpha\rangle = \frac{1}{N} \sum_{L} e^{i k_{SC} \cdot L} \phi(r - L - l_{x} - t_{\alpha}),
\]

where \(N\) is a normalization constant, \(\phi(r - L - l_{x} - t_{\alpha})\) is the \(p_{z}\) orbital located at \(L + l_{x} + t_{\alpha}\). Equation (9) indicates that only the eigenstates of approximant are necessary to obtain the spectral function.

Then, the effective band structure can be obtained by

\[
\delta N(k, \epsilon) = \int_{\epsilon - \delta\epsilon/2}^{\epsilon + \delta\epsilon/2} A(k, \epsilon') d\epsilon',
\]

where \(\delta\epsilon\) is the bin width in energy sampling.

### III. RESULTS AND DISCUSSION

![Figure 2: The comparison of density of states of MGQCs (QC) with their 15/26 approximants. Each MGQC contains more than ten million atoms. The complete agreement verifies the validation of these approximants.](image)

The DOS comparisons between the large round sample of each MGQC and its corresponding 15/26 approximant are given in Fig. 2. The full agreement indicates that these 15/26 approximants are accurate enough to further understand the electronic properties of these MGQCs, for instance the EBS which is the main quantity investigated in this paper.

#### A. Mirror symmetry effect

Firstly, we have a brief review of the electronic structure of the 30° TBG before the discussion of MGQCs. Experimental [38, 39] and theoretical studies [40, 41] indicated that, the Dirac cones occur not only at the BZ corners (all \(K\), \(K'\), \(\bar{K}\) and \(\bar{K}'\) points), but also at all their mirror-symmetric points \(K_{1}\) and \(\bar{K}_{1}\) (see Fig. 1(b) for the positions of these points.). This phenomenon can be explained qualitatively by the generalized Umklapp scattering theory [39, 42]. Graphene monolayers \(A\) and \(\bar{A}\) are crystallines, which have Bloch functions

\[
|k, X\rangle = \frac{1}{\sqrt{n}} \sum_{R} e^{i k \cdot R} |\varphi(r - R - \tau_{X})\rangle
\]

and

\[
|\tilde{k}, \tilde{X}\rangle = \frac{1}{\sqrt{n}} \sum_{\tilde{R}} e^{i \tilde{k} \cdot \tilde{R}} |\varphi(r - \tilde{R} - \tau_{\tilde{X}})\rangle,
\]

respectively. The quantities without and with tilde notations correspond to layer \(A\) and \(\bar{A}\) respectively. \(n\) (\(\tilde{n}\)) is the normalization factors, \(\tau_{X}\) (\(\tau_{\tilde{X}}\)) is the position of sublattice \(X\) (\(\tilde{X}\)), and \(\varphi(r - R - \tau_{X})\) (\(\varphi(r - \tilde{R} - \tau_{\tilde{X}})\)) is the \(p_{z}\) orbital locating at sublattice \(X\) (\(\tilde{X}\)) in unit cell \(R\) (\(\tilde{R}\)).

If the collection of the Bloch functions \(\{|k, X\rangle, |\tilde{k}, \tilde{X}\rangle\}\) is used as the basis functions of 30° TBG, the interlayer interaction \(U\) couples the \(k\) state of layer \(A\) and \(\tilde{k}\) state of layer \(\bar{A}\), if the condition \(k + G = \tilde{k} + \tilde{G}\) is satisfied, where \(G\) and \(\tilde{G}\) are the reciprocal lattice vectors of \(A\) and \(\bar{A}\) layers, respectively. Moreover, the smaller length of \(|q| = |k + G|\) corresponds to the stronger scattering (coupling) strength \(|\langle k, X|U|\tilde{k}, X\rangle|\). The strongest scattering processes scatter the Dirac cones at the \(K\), \(K'\), \(\bar{K}\) and \(\bar{K}'\) to their mirror-symmetric points \(K_{1}\) and \(\bar{K}_{1}\). As an example, one of the strongest scattering processes for \(K'\) point is shown in Fig. 1(c). That is the reason why the Dirac cones exist not only at \(K\), \(K'\), \(\bar{K}\) and \(\bar{K}'\) but also at their mirror-symmetric points \(K_{1}\) and \(\bar{K}_{1}\).

In order to check whether the mirror symmetry effect still remains in the MGQCs, the EBSs of these MGQCs are shown in Fig. 3. The detailed EBSs around \(K\), \(K_{1}\), \(\bar{K}\) and \(\bar{K}_{1}\) along four different directions are given in Fig. 4. Our results show that the EBSs around \(K_{1}\) and \(\bar{K}_{1}\) are always the mirror symmetry of the EBSs at BZ corners (\(K\) and \(\bar{K}\), respectively). Moreover, like 30° TBG [43], the spectral functions at mirror-symmetric points become also anisotropic and much weaker. For any MGQC, the EBSs at \(\bar{K}_{1}\) and \(\bar{K}_{1}\) are expected to be able to be detected by ARPES measurements because the EBSs of the same magnitude in 30° TBG have been detected successfully [39].

For the MGQCs composed of only one 0°-subsystem and one 30°-subsystem, for instance, the first five in Fig. 1(a), our results in Fig. 4 indicate that in the low-energy region the two subsystems always retain their own band structures. For example, in \(BA\), the EBSs at \(K\) and \(\bar{K}\) are just the band structures of \(AB\) stacked bilayer graphene and graphene monolayer, which are just the 0°- and 30°-subsystems, respectively. However, different
FIG. 3: The effective band structures of the MGQCs. The spectral functions around $K_1$ and $\tilde{K}_1$ are multiplied by 300. The path for effective band structure is along the arrowed dashed blue line in Fig. 1(b).

from separating the subsystems in space, the EBSs at BZ corners are always scattered to their mirror-symmetric points. Besides, similar to 30° TBG, the band valleys from any BZ corner ($K$ or $\tilde{K}$) and its mirror-symmetric point ($\tilde{K}_1$ or $K_1$) also hybrid strong near their mid-point ($\tilde{M}$ or $M$).

The EBSs of the MGQCs in the low-energy region can also be explained by the generalized Umklapp scattering theory. Let’s take $AA\tilde{A}$ as an example, where the 0°- and 30°-subsystems are $AA$ and $\tilde{A}\tilde{B}$ bilayers, respectively. The Hamiltonian can be written as

$$H = H_{AA} + H_{A\tilde{A}} + H_{\tilde{A}B} + U_{AA} + U_{A\tilde{A}} + U_{\tilde{A}B},$$

(13)

where, $H_{AA} = H_A + H_{\tilde{A}} + U_{AA}$ and $H_{\tilde{A}B} = H_{\tilde{A}} + H_B + U_{\tilde{A}B}$, which are the Hamiltonians of isolated $AA$ and $AB$ bilayers, respectively. If the collection of the Bloch functions of the two subsystems $\{ |k, X\rangle, |\tilde{k}, \tilde{X}\rangle \}$ is used as the basis, where $X$ and $\tilde{X}$ are the sublattices of $AA$ and $AB$ bilayers, respectively. Then the interaction between the two subsystems $U_{A\tilde{A}}$ as the perturbation scatters the band structures at BZ corners to their mirror-symmetric points.

Previous calculations indicated that the interlayer binding energies in $AB$ and $AA$ stacked bilayer graphene are $17.7 \sim 70$ and $10.4 \sim 31.1$ meV/atom depending on calculation methods, respectively. The binding energy in 30° TBG is 1.6 meV/atom smaller than that in the $AB$ stacked bilayer graphene. It means that the stability of the $AA$ stacking should be comparable to the $AA$ stacking. As a consequence, in principle, for the MGQC, it is not definitely convincing to treat the interlayer interaction across $A\tilde{A}$ stacking interface as a perturbation when comparing with the $AA$ stacking interface. However, all of our calculated EBSs of the MGQCs indicate that, comparing with the $AA$ and $AB$ stacking interfaces, to treat the interlayer interaction across the $A\tilde{A}$ stacking interface as the perturbation can always gives the accurate band structure in the low-energy region.

B. Under the electric field

Because the band structure of the subsystem in MGQCs, such as $AA$ and $AB$ stacked bilayers, is sensitive to the electric field, the application of the electric filed is expected to be a valid method to turn the the EBSs of the MGQCs. The calculated EBSs of the MGQCs under the 0.05 eV/Å electric field along $z$ direction are shown in Fig. 5. The results indicate that in the low-energy region all subsystems still perform like isolated systems and keep their own band structures at the BZ corners. For in-
FIG. 4: The effective band structures (EBSs) of the MGQCs around $K$, $K_1$, $\bar{K}$ and $\bar{K}_1$ along four directions $x$, $y$, $x+y$ and $x-y$. The EBSs around $\bar{K}_1$ and $K_1$ are multiplied by 300, which are obviously anisotropic along different directions. The mirror symmetry effect, namely the EBSs around $\bar{K}_1$ and $K_1$ being identical to the EBSs around their mirror-symmetric points $K$ and $\bar{K}$, respectively, remains for all systems. For the first five systems composed of one 0$^\circ$-subsystem and one 30$^\circ$-subsystem, the EBSs around $K$ and $\bar{K}_1$ ($\bar{K}$ and $K_1$) are just the band structures of 0$^\circ$-subsystem (30$^\circ$-subsystem). For the case of $AA\bar{A}B$, the EBSs at $K$ and $\bar{K}_1$ are the simple combination of the band structures of two 0-subsystems (an $AA$ and an $AB$ stacked bilayer graphene), and the EBSs at $\bar{K}$ and $K_1$ are just the band structure of the 30$^\circ$-subsystem, namely the graphene monolayer ($\bar{A}$).

In $AA\bar{A}\bar{B}$, the EBSs at $K$ and $\bar{K}$ are just the band structures of the 0$^\circ$- and 30$^\circ$-subsystems, namely the $AA$ and $AB$ stacked bilayers, respectively, under an external electric field. Importantly, the mirror symmetry effect of the EBS still remains. Therefore, relative to the $AA$ and $AB$ stacking interfaces, it is still reliable to deal with the interlayer interaction across the $AA$ stacking interface as the perturbation even under the electric field. Besides, the charge transfer from top to bottom subsystem can be obviously detected from these calculated EBSs after the application of the electric field, which results in the p- and n-type doping in two subsystems, respectively. Although the screen effect are not included in our calculations, the weaker charge transfer should still exist. Therefore, the application of the electric field is an effective method to engineer the EBSs of MGQCs due to the
FIG. 5: The effective band structures (EBSs) of the 30° twisted multilayer graphenes around $K$, $K_1$, $\bar{K}$ and $\bar{K}_1$ along four directions $x$, $y$, $x+y$ and $x-y$ under an electric field of 0.05 eV/Å. The EBSs around $\bar{K}_1$ and $K_1$ are multiplied by 300. The mirror symmetry effect still remains. For the first five systems, any of which can be divided into two parts by 30° interface, the EBSs around $K$ and $\bar{K}_1$ (\bar{K} and $K_1$) are just the band structures of the bottom (top) part under the electric field with extra electrons (holes) doping. For the case of $AAA\bar{A}$, the EBSs at $K$ and $\bar{K}_1$ are the combination of the band structures of an $AA$ and an $AB$ bilayer graphene under the electronic field with extra electrons and holes doping, respectively, and the EBSs at $\bar{K}$ and $K_1$ are just the band structure of p-type doped graphene monolayer ($\bar{A}$).

multilayer structure of the subsystems.

C. Combination effect

For the MGQC composed of two 0°-subsystems and one 30°-subsystem, namely $AAA\bar{A}$. The two 0°-subsystems, $AB$ and $AA$ stacked bilayers, share the same BZ (the black one in Fig. 4(b)). The 30°-subsystem is a graphene monolayer $\bar{A}$, whose BZ is the red one in Fig. 1(b). The result shown in Fig. 4(f) indicates that the EBS around $K$ is the combination of the band structures of the $AA$ and $AB$ stacked bilayers, which is also scattered to $\bar{K}_1$. Such a result opens up a way to engineer the band structure of the graphene multilayer. If the combination of the band structures of two different structures is required, we can replace the two 0°-subsystems with corresponding structures. For example, if the two 0°-
subsystems are replaced with graphene and AA stacked bilayer graphene, namely AA\AA, the combination of the band structures of graphene and AA stacked bilayer graphene occurs at K point (see Fig. 6(a)). For BAA\AA, the EBS at K point is the combination of the band structures of graphene and AB stacked bilayer graphene (see Fig. 6(b)). Such a strategy can also be verified by comparing the EBS of AAAAB with the band structures of AAB and AAAB, both of which are crystalline. All the three structures contain both AA and AB stacking interfaces. However, the interactions among the layers in AAB and AAAB make the band structures totally different from the combination of the band structures of AA and AB stacked bilayers (see Fig. 6(c)). Besides, if the distance between the two middle layers in AAAB is increased to be large enough, the band structure of the system would show the combination, while these two bilayers are separated totally in not only structure but also electronic properties.

The EBS of a general MGQC composed of several 0°- and 30°-subsystems can be envisioned. According to our calculated EBSs of the MGQCs with one and two subsystems with the same orientation, it can be expected that each subsystem in the general MGQC should keep its own band structure in the low-energy region at the BZ corners. Accordingly, the combination of the band structures of all 0°- and 30°-subsystems should appear at corresponding BZ corners. Moreover, these EBSs should be scattered into the their six mirror-symmetric points, respectively.

FIG. 6: (a) and (b) are effective band structures of AA\AA and BAA\AA, respectively. (c) and (d) are the band structures of multilayer AAB and AAAB, respectively. All results are around K point along x direction.

IV. CONCLUSION

By means of the tight-binding approximation, we systematically study the electronic properties of the multilayer graphene quasicrystals (MGQCs), which are composed of some subsystems, such as graphene monolayer, AA and AB stacked bilayer graphenes, with some of the subsystems twisted by 30°. The approximants of all MGQCs are proposed, and the validations of these approximants are verified by comparing the density of states (DOS) by using the tight-binding propagation method. In order to calculate the DOS of a MGQC, the MGQC is simulated by the round disk with large enough size (containing 10 million atoms) because of the failure of the band theory. The effective band structures of these MGQCs are derived by unfolding the band structures of these approximants. For the MGQCs composed of only one 0°-subsystem and one 30°-subsystem, in the low-energy region each subsystem performs like an isolated system and keeps its own band structures at the corners of the Brillouin zone. More importantly, all the effective band structures at the corners are scattered into the mirror-symmetric points.

DATA AVAILABILITY

All data generated and/or analyzed during this study are included in this article.

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AUTHOR CONTRIBUTIONS

Z. Wu performed the TBPM calculations. G. Yu performed all other calculations and wrote the
manuscript. All authors analyzed the results and revised the manuscript. S. Yuan conducted the project.

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