Electronic states in two-dimensional bilayer quasicrystals and the effects of interlayer and sublattice symmetries

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We study the electronic structure of quasicrystals composed of incommensurate stacks of atomic layers. We consider two systems: a pair of square lattices with a relative twist angle of $\theta = 45^\circ$ and a pair of hexagonal lattices with a relative twist angle of $\theta = 30^\circ$, with various interlayer interaction strengths. This constitutes every two-dimensional bilayer quasi-crystal system. We investigate the resonant coupling governing the quasicrystalline order in each quasicrystal symmetry, and calculate the quasi-band dispersion. We find that some quasicrystalline states, which are usually obscured by additional weakly coupled states, are more prominent, i.e., "exposed", in the systems with strong interlayer interaction. We also show that we can switch the states between quasicrystalline configuration and its layer components, by turning on and off the interlayer symmetry. On the other hand, hexagonal lattices with sublattice potential asymmetry, e.g., transition metal dichalcogenide of hexagonal Boron Nitride, give the quasicrystalline states quite similar to those in the system in the absence of sublattice asymmetry.

I. INTRODUCTION

When two honeycomb lattices are overlapped on top of the other at a twist angle $\theta = 30^\circ$, the atomic arrangement is mapped on to a quasicrystalline lattice, which is ordered but not periodic, with a 12-fold rotational symmetry. Recently, it has been demonstrated that bilayer graphene with a precise rotation angle of $30^\circ$ exhibits the atomic structures satisfying the quasicrystalline tiling as well as the spectrum respecting the 12-fold rotational symmetry. Similar structures have also been realized by growing bilayer graphene on top of the Ni or Cu surface, and also by a transfer method. The conventional moiré effective theory, which is based on the period of the moiré pattern arising from the interference between the lattice periods, cannot describe the electronic structures of such quasicrystals composed of incommensurate stack of atomic layers (hereafter "vdW-QCs") since the rotational symmetry of quasicrystals does not commute with translation. In our previous work, we developed a momentum-space tight-binding model which can describe the electronic structures of atomic layers stacked at any configuration without relying on the moiré periodicity. This model enabled us to reveal the quasi-band dispersion and the emergence of the electronic states having the quasicrystalline order in the vdW-QC composed of two graphene layers stacked at $30^\circ$ by fully respecting the rotational symmetry of quasicrystals as well as the translational symmetry of constituent layers. While conventional quasicrystals can be viewed as intrinsic quasicrystals where all the atomic sites are intrinsically arranged in the quasiperiodic order, vdW-QCs are regarded as extrinsic quasicrystals, in that they are composed of a pair of perfect crystals having independent periodicities, and the quasiperiodic nature appears only in the perturbational coupling between the two sub-systems. Thus, vdW-QCs provide a unique opportunity to design quasicrystalline states by using atomic layers with various symmetries and interaction strength.

In this paper, we numerically investigate the electronic structures of vdW-QCs for every possible rotational symmetry in two-dimensional space. Since a periodic two-dimensional atomic layer can have 2-, 4-, 6-fold rotational symmetry, we can make only 8-fold [octagonal, Fig. 1(a)] or 12-fold [dodecagonal, Fig. 1(d)] vdW-QCs by stacking two square lattices at $45^\circ$ or by stacking two hexagonal lattices at $30^\circ$, respectively. We first find the resonant condition, which gives quasicrystalline order to the electronic states, in each system, and calculate the quasi-band dispersion and density of states (DOS) for various interlayer interaction strength. We identify the features which arise from the quasicrystalline order as opposed to those arising from the interaction common to any other $\theta$ in the spectrum of vdW-QCs. In addition, we show that some quasicrystalline states, which are usually obscured by additional weakly coupled states, are more prominent in vdW-QCs with strong interlayer interaction.

We also investigate the effects of lifting both interlayer and sublattice symmetry on the electronic structure. Since the quasicrystalline order arises from the resonant interaction between the states in both layers, interlayer potential asymmetry results in a dramatic change in the electronic structure. In addition, we can switch between states respecting quasicrystal symmetry and those satisfying only half the symmetry by turning on and off the interlayer symmetry. On the other hand, we show that sublattice potential asymmetry, such as that in transition metal dichalcogenide, does not make a dramatic difference - it results only in a constant energy shift. We also analytically interpret the mixing between the
quasicrystalline states, which may influence the physical properties (such as optical selection rules), and changes to the location of the band edge via interlayer or sublattice potential asymmetry.

The paper is organized as follows. In Sec. II, we present the atomic structures and tight-binding model for vdW-QCs, and introduce the dual tight-binding approach in the momentum space. In Sec. IIIA we find the resonant interaction which gives the quasicrystalline electronic states in vdW-QCs. In Sec. IIIB and IIIC we derive the minimal Hamiltonian and calculate the band dispersion and wave functions of octagonal and dodecagonal vdW-QCs, respectively. We also investigate the effects of various interlayer interaction strengths, the features arising from 2-wave mixing, and the effects of the lifting of interlayer and sublattice potential asymmetry. A brief conclusion is given in Sec. IV.

II. THEORETICAL METHODS

A. Atomic structure and Brillouin zones of quasicrystalline twisted bilayers

We define the atomic structure of the octagonal vdW-QCs by starting from two perfectly overlapping square lattices and rotating the layer 2 around the center of the square by $\theta = 45^\circ$ [Fig. I(a)]. We set $xy$ coordinates parallel to the layers and $z$ axis perpendicular to the plane. The system belongs to the symmetry group $D_{4d}$, and it is invariant under an improper rotation $R(\pi/4)M_x$, where $R(\theta)$ is the rotation by an angle $\theta$ around $z$ axis, and $M_x$ is the mirror reflection with respect to $xy$ plane. The primitive lattice vectors of layer 1 are taken as $a_1 = a(1,0)$ and $a_2 = a(0,1)$, where $a$ is the lattice constant, and those of the layer 2 as $\tilde{a}_1 = R(\pi/4)a_1$. In this paper, we model the square lattices by a minimal, one orbital model with one sublattice site. Then, the atomic positions are given by

$$R_X = n_1a_1 + n_2a_2 + \tau_X \quad \text{layer 1},$$

$$\tilde{R}_X = \tilde{n}_1\tilde{a}_1 + \tilde{n}_2\tilde{a}_2 + \tau_{\tilde{X}} \quad \text{layer 2},$$

where $n_i$ and $\tilde{n}_i$ are integers, $X$ ($\tilde{X}$) denotes the sublattice site (only one in this case) of layer 1 (layer 2) of which position in the unit cell is defined by $\tau_X = (a/2, a/2)$ and $\tau_{\tilde{X}} = R(\pi/4)\tau_X + de_z$. Here, $d$ is the interlayer spacing between the two layers and $e_z$ is the unit vector normal to the layer. The reciprocal lattice vectors of layer 1 are given by $a^*_1 = (2\pi/a)(1,0)$ and $a^*_2 = (2\pi/a)(0,1)$, and layer 2 by $\tilde{a}^*_2 = R(\pi/4)\tilde{a}^*_2$.

Likewise, we define the atomic structure of the dodecagonal vdW-QCs by starting from two perfectly overlapping honeycomb lattices (i.e., AA-stacked bilayers) and rotating the layer 2 around the center of the hexagon by $\theta = 30^\circ$ [Fig. I(d)]. The system belongs to the symmetry group $D_{6d}$, and it is invariant under an improper rotation $R(\pi/6)M_z$. The primitive lattice vectors of layer 1 are taken as $a_1 = a(1,0)$ and $a_2 = a(1/2, \sqrt{3}/2)$, where $a$ is the lattice constant, and those of the layer 2 as $\tilde{a}_1 = R(\pi/6)a_1$. The atomic positions are given by $X\equiv R = (a/2, a/2)$, where $X = A,B$ ($\tilde{X} = \tilde{A}, \tilde{B}$) denotes the sublattice site of layer 1 (2), and $\tau_X$ and $\tau_{\tilde{X}}$ are the sublattice positions in the unit cell, defined by $\tau_A = -\tau_1$, $\tau_B = R(\pi/6)\tau_1 + de_z$, $\tau_{\tilde{A}} = -R(\pi/6)\tau_1$, and $\tau_{\tilde{B}} = R(\pi/6)\tau_1 + de_z$ with $\tau_1 = (0,a/\sqrt{3})$, where $d$ is the interlayer spacing between the two layers. The reciprocal lattice vectors of layer 1 are given by $a^*_1 = (2\pi/a)(1,-1/\sqrt{3})$ and $a^*_2 = (2\pi/a)(0,2/\sqrt{3})$, and layer 2 by $\tilde{a}_2 = R(\pi/6)a^*_2$. The red and blue squares (hexagons) in Fig. I(c) [Fig. I(f)] show the Brillouin zones of layer 1 and 2 in octagonal (dodecagonal) vdW-QCs, respectively.

B. Tight-binding model for van der Waals bilayers

We model both systems by the tight-binding model with a single orbital of arbitrary atomic species. Although we use $p_z$ orbitals in this paper, just like the model of graphene and hexagonal Boron Nitride, but it can be any other orbital optimal for each system; e.g., in transition metal dichalcogenides, we can use $d$-orbital and so on. The Hamiltonian is spanned by the Bloch bases of each sublattice,

$$|\mathbf{k}, X\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_X} e^{i\mathbf{k}\cdot\mathbf{R}_X} |\mathbf{R}_X\rangle \quad \text{layer 1},$$

$$|\mathbf{k}, \tilde{X}\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_{\tilde{X}}} e^{i\mathbf{k}\cdot\mathbf{R}_{\tilde{X}}} |\mathbf{R}_{\tilde{X}}\rangle \quad \text{layer 2},$$

where $|\mathbf{R}_X\rangle$ is the atomic $p_z$ orbital at the site $\mathbf{R}_X$, $\mathbf{k}$ and $\mathbf{\tilde{k}}$ are the two-dimensional Bloch wave vectors and $N = S/S_{tot}$ is the number of the unit cells with an area $S = a^2$ for square lattices and $S = (\sqrt{3}/2)a^2$ for honeycomb lattices in the total system area $S_{tot}$.

We use a Slater-Koster parametrization \cite{Slater} for the transfer integral between any two $p_z$ orbitals,

$$-T(\mathbf{R}) = V_{pp\sigma} \left[ 1 - \left( \frac{\mathbf{R} \cdot \mathbf{e}_z}{|\mathbf{R}|} \right)^2 \right] + V_{pp\pi} \left( \frac{\mathbf{R} \cdot \mathbf{e}_z}{|\mathbf{R}|} \right)^2,$$

where $\mathbf{R}$ is the relative vector between two atoms, and

$$V_{pp\sigma} = V_{pp\sigma}^0 e^{-|\mathbf{R} - a|/\delta_1},$$

$$V_{pp\pi} = V_{pp\pi}^0 e^{-|\mathbf{R} - 3a|/\delta_2},$$

for square lattices and

$$V_{pp\sigma} = V_{pp\sigma}^0 e^{-|\mathbf{R} - a/\sqrt{3}|/\delta_1},$$

$$V_{pp\pi} = V_{pp\pi}^0 e^{-|\mathbf{R} - 1.36a|/\delta_2},$$

for honeycomb lattices so that the first-nearest neighbor interlayer coupling becomes $V_{pp\pi}^0 (< 0)$. In both systems, we take the decay length of the transfer integral as $\delta_1 = 0.184a$ \cite{Pol}.
The total tight-binding Hamiltonian is expressed as
\[ \mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{U} + \mathcal{H}_V + \mathcal{H}_\Delta \]  \hspace{1cm} (6)
where \( \mathcal{H}_1 \) and \( \mathcal{H}_2 \) are the Hamiltonian for the intrinsic square or honeycomb lattices of layer 1 and 2, respectively, \( \mathcal{U} \) is for the interlayer coupling, \( \mathcal{H}_V \) is for the interlayer potential asymmetry, and \( \mathcal{H}_\Delta \) is for the sublattice potential asymmetry. Note that the square lattices with the minimal, one orbital model considered in this work does not have the \( \mathcal{H}_\Delta \) term as there is only one sublattice in this case. The intralayer matrix elements of layer 1 are given by
\[ \langle \mathbf{k}' X' | \mathcal{H}_1 + \mathcal{H}_V + \mathcal{H}_\Delta | \mathbf{k}, X \rangle = [h_{X',X}(\mathbf{k}) + \{V/2 + s_\Delta \Delta/2\} \delta_{X',X}] \delta_{\mathbf{k}',\mathbf{k}}, \]  \hspace{1cm} (7)
where \( \mathbf{L} = n_1 a_1 + n_2 a_2, \tau_{X',X} = \tau_{X'} - \tau_X, V \) is the magnitude of the interlayer potential asymmetry, \( s_\Delta \) is +1 and −1 for \( X = A \) and \( B \), respectively, and \( \Delta \) is the magnitude of the sublattice potential asymmetry. Similarly, the matrix for \( \mathcal{H}_2 \) is given by replacing \( \mathbf{k} \) with \( R(-\pi/4)\mathbf{k} \) in a square lattice and \( R(-\pi/6)\mathbf{k} \) in a honeycomb lattice, \( V/2 \) with \( -V/2 \), and \( s_\Delta \) by +1 and −1 for the sublattice \( A \) and \( B \), respectively. The interlayer matrix element between layer 1 and 2 is written as
\[ \langle \mathbf{k}, \tilde{X} | U | \mathbf{k}, X \rangle = -\sum_{\mathbf{G}, \tilde{\mathbf{G}}} t(\mathbf{k} + \mathbf{G}) e^{-i\mathbf{G} \cdot \tau_X + i\tilde{\mathbf{G}} \cdot \tau_{X'}} \delta_{\mathbf{k} + \mathbf{G}, \mathbf{k} + \tilde{\mathbf{G}}}, \]  \hspace{1cm} (8)
where \( \mathbf{G} = n_1 a_1 + n_2 a_2 \) and \( \tilde{\mathbf{G}} = \tilde{n}_1 \tilde{a}_1 + \tilde{n}_2 \tilde{a}_2 \) \((n_1, n_2, \tilde{n}_1, \tilde{n}_2 \in \mathbb{Z})\) run over all the reciprocal points.
of layer 1 and 2, respectively. Here

$$t(q) = \frac{1}{S} \int T(r + z_X c_e) e^{-i q \cdot r} dr$$

(9)

is the in-plane Fourier transform of the transfer integral, where $z_X = (\tau_X - \tau_X) \cdot e_z$. For $H_1$ and $H_2$, we consider only the nearest neighbor interactions in octagonal vdW-QCs to keep the symmetric and simple cosine bands, and all interactions of $|R|$ within 3a in dodecagonal vdW-QCs to make it consistent with previous work \cite{2,8,10}.

C. Effects of using different atomic layers and interlayer interaction

Different vdW-QCs exhibit different electronic structures, since different atomic layers have different interlayer interaction. In addition, we can also tune the interlayer interaction in a given vdW-QC with the interlayer distance $d$, e.g., by applying an external pressure or intercalation. However, if two different vdW-QCs have the same sublattice configuration and orbitals, the interactions [Eq. (3)] in the two systems have the same phase and the same direction $|q|/|q|$ dependence, and differ only in the magnitude of the interaction $t(q)$. In most van der Waals systems with $d > a$, $t(q)$ scales with $V_{pp\sigma}$ and exponentially decays with $d$. Thus, we can investigate the electronic structures of various vdW-QCs by simply scaling $t(q)$. In Sec. III B and III C, we will first investigate the electronic structures of a vdW-QC with $t(q)$ obtained at a specific combination of $(V_{pp\sigma}, d)$, then investigate the effects of using different materials and interlayer strength by tuning $t(q)$.

D. Dual tight-binding model in momentum space

In real-space, since quasicrystals do not have periodicity, we need infinitely many bases to solve Eq. (6). Although some conventional approximations with a finite number of bases, such as a periodic approximant or a finite-size model, can give an energy spectrum quite similar to the actual spectrum, the resulting wave functions lose their long-range quasicrystalline nature.

Instead, we can solve Eq. (6) rigorously by using a tight-binding model in $k$ space, which is the dual counterpart of the original tight-binding Hamiltonian in the real space. Equation (6) shows that the interlayer interaction occurs between the states satisfying the generalized Umklapp scattering condition $k + G = k + G$. It is straightforward to show that the entire subspace spanned by $H_1 + H_2 + U$ from a layer 1’s Bloch state at $k$ is given by $\{ |k, X\rangle | k = k + G, \forall G \}$ and $\{ |k, X\rangle | k = k + G, \forall G \}$. According to Eq. (6), the interaction strength between $k = k + G$ and $k = k + G$ is given by $t(q)$ where $q = k + G = k + G = k + k - k$. Then, the interaction strength can be visualized by the diagram Figs. III (b) and (e), where all the layer 2’s wave points $\tilde{k}$ are inverted to $\tilde{k} - k$, and overlapped with the layer 1’s wave points $k$. In the map, the quantity $|q| = |k + k - k|$ is the geometrical distance between two points, and the interaction takes place only between the points located in close distance, since $t(q)$ decays in large $q$. Therefore, in practical calculation, we only need a limited number of states around $k$ inside a certain cut-off circle $k_c$. Hence, we can obtain the energy eigenvalues at $k$ by diagonalizing the Hamiltonian matrix within a finite set of bases. If the $k$ points are viewed as “sites”, the whole system can be recognized as a tight-binding lattice in $k$ space, which is dual to the original Hamiltonian in the real space. This enables us to calculate the electronic structures of almost every possible stack of atomic layers without relying on moiré periodicity. The method as well as its validity are explained in detail in Ref. 8.

Below, we will first study the eigenstates of $H_1 + H_2 + U$ by using the dual-tight binding method, then investigate the effects of $H_V$ and $H_\Delta$ in terms of those eigenstates.

III. RESULTS AND DISCUSSION

A. Resonant states respecting the rotational symmetry of quasicrystals

The rotational symmetry of the quasicrystal as well as the translational symmetries of the constituent atomic layers [accordingly, the generalized Umklapp scattering condition Eq. (6)] reveal the most dominant interaction, which comes from the resonance between degenerate states, in each vdW-QC. In octagonal vdW-QCs, we see that the eight symmetric points $Q_n = [-\pi/a, -\pi/a] + (a'/\sqrt{2})[\cos(n\pi/4), \sin(n\pi/4)] (n = 0, 1, 2, \cdots, 7)$ form a circular chain in the dual tight-binding lattice with $k = Q_0$. The chain has a radius of $a'/\sqrt{2} = |a'|/\sqrt{2} = \sqrt{2}\pi/a$, and is indicated by the dashed ring in Fig. 1(b). Noting that the layer 2’s wave points are inverted, these points are associated with layer 1’s Bloch wave numbers $k = Q_n$ for even $n$’s and layer 2’s $k = Q_n - Q_0$, for odd $n$’s. Figure III (c) shows the original positions of $k$ (layer 1) and $k$ (layer 2) associated with $Q_n$ in the first Brillouin zone, $C_n = \sqrt{2}a' \sin 22.5^\circ[\cos(3\pi/8 + 5n\pi/4), \sin(3\pi/8 + 5n\pi/4)] (n = 0, 1, 2, \cdots, 7)$. Each intrinsic square lattice has a single cosinusoidal band with a band maximum and minimum at M- and Γ-points of the Brillouin zone, respectively. Due to the symmetry, the Bloch states of the intrinsic lattices at the eight points are all degenerate in energy, and therefore the interlayer coupling hybridizes them to make resonant states. Here the coupling is only relevant between the neighboring sites of the ring, and it is given by $t_0 = t(|C_n|)$. The interaction to other neighboring states in the dual tight-binding lattice can be safely neglected since the interaction strength is much less than $t_0$ and the two states are not degenerate in most cases.
Likewise, in dodecagonal vdW-QCs, we see that the twelve symmetric points \( Q_n = a^* \cos(n\pi/6), \sin(n\pi/6) \) form a circular chain in the dual tight-binding lattice with \( \mathbf{k} = 0 \) and the radius is \( a^* \equiv |a^*| = 4\pi/(\sqrt{3}a) \) [dashed ring in Fig. 1(e)]. These points are associated with layer 1’s Bloch wave numbers \( \mathbf{k} = \mathbf{Q}_n \) for even \( n \)’s and layer 2’s \( \mathbf{k} = -\mathbf{Q}_n \) for odd \( n \)’s, and Fig. 1(e) shows the original positions of \( \mathbf{k} \) (layer 1) and \( \mathbf{k} \) (layer 2) associated with \( \mathbf{Q}_n \) in the first Brillouin zone, \( \mathbf{C}_n = 2a^* \sin(15\pi/2 + n\pi/6), \sin(5\pi/6 + n\pi/6) \) \( (n = 0, 1, 2, \ldots, 11) \). Again, the Bloch states of each intrinsic honeycomb lattice at the twelve points are all degenerate in energy and hybridized to form resonant states by interlayer coupling with \( t_0 = t(|C_n|) \).

It should be noted that these states are not the only set of states which show the resonant coupling in each system. As we shown in Appendix A there are more sets of states, with different wave numbers, showing the resonant interaction respecting the rotational symmetry of the quasicrystals. However, the sets in Figs. 1(b) and (e) give the strongest interaction, i.e., largest energy separation, since these states form the rings with the shortest distance between neighboring states in the dual tight-binding lattices.

### B. Octagonal quasicrystal

#### 1. Hamiltonian

In octagonal vdW-QCs, the strongest resonant interaction occurs at \( \mathbf{k} = \mathbf{Q}_0 \). Thus, by replacing \( \mathbf{k} \) with \( \mathbf{Q}_0 + \mathbf{k}_0 \), we can express the Hamiltonian

\[
\mathcal{H} = \mathcal{H}_{\text{ring}} + \mathcal{H}_V,
\]

in the vicinity of \( \mathbf{k}_0 = 0 \), in the bases of \( \{|k(0)\rangle, |k(1)\rangle, \ldots, |k(7)\rangle\} \), where \( |k(n)\rangle \equiv |\mathbf{k}_0 + \mathbf{C}_n, X\rangle \) for even \( n \) (layer 1) and \( |\mathbf{k}_0 + \mathbf{C}_n, \bar{X}\rangle \) for odd \( n \) (layer 2). Here,

\[
\mathcal{H}_{\text{ring}}(\mathbf{k}_0) = \begin{pmatrix}
H(0) & -t_0 & -t_0 & \cdots & -t_0 \\
-t_0 & H(1) & -t_0 & \cdots & -t_0 \\
-t_0 & -t_0 & H(2) & -t_0 & \cdots \\
\vdots & \cdots & \cdots & \cdots & \cdots \\
-t_0 & \cdots & \cdots & -t_0 & H(6) \\
-t_0 & \cdots & \cdots & \cdots & -t_0 & H(7)
\end{pmatrix},
\]

is the Hamiltonian of the resonant ring in the absence of interlayer potential asymmetry, where \( H(n)(\mathbf{k}_0) = h_{X,X} [R(-5\pi/4)\mathbf{Q}_0 + \mathbf{C}_0] \), and we neglect the \( \mathbf{k}_0 \) dependence of the interlayer matrix element \( t(\mathbf{q}) \). The diagonal elements \( H(n) \) represent monolayer’s Hamiltonian at \( \mathbf{k} = \mathbf{k}_0 + \mathbf{C}_n \) for even \( n \) and \( \mathbf{k} = \mathbf{k}_0 + \mathbf{C}_n \) for odd \( n \). Note that \( h_{X,X} \) in \( H(n) \) is same for any \( n \), and the dependence of the diagonal elements on \( n \) solely comes from \( R(-5\pi/4)\mathbf{k}_0 \) in the argument of \( h_{X,X} \). Consequently, the ring Hamiltonian \( \mathcal{H}_{\text{ring}} \) is obviously symmetric under rotation by a single ring of the ring (i.e., moving \( \mathbf{C}_n \) to \( \mathbf{C}_{n+1} \)), which actually corresponds to the operation \( [R(\pi/4)\mathbf{M}]^5(225^\circ \text{ rotation and swapping layer 1 and 2}) \) in the original system. In addition, \( \mathcal{H}_{\text{ring}} \) has a particle-hole symmetry with respect to the energy \( E = h_0 \), where \( h_0 = h_{X,X}(\mathbf{C}_0) \approx -2V_{pp}\sin(\sqrt{2}\pi + 1) \), up to the first order to \( \mathbf{k}_0 \) (Appendix B).

\( \mathcal{H}_V \) is the Hamiltonian representing the interlayer potential asymmetry,

\[
\mathcal{H}_V = \frac{V}{2} \begin{pmatrix}
\sigma_z & \sigma_z & \sigma_z & \sigma_z & \cdots & \sigma_z & \sigma_z
\end{pmatrix},
\]

where \( V \geq 0 \) represents the difference in the electrostatic energies between the two layers, and \( \sigma_z \) is the Pauli matrix. With \( \mathcal{H}_V \), the Hamiltonian \( \mathcal{H} \), which was originally in the form of one-dimensional monatomic chain in the dual-tight binding lattice, becomes that of diatomic chain with alternating on-site potential.

#### 2. Band structures and wave functions

Figure 2(a) shows the band structures of the octagonal vdW-QCs near \( \mathbf{C}_n \), in the absence of interlayer potential asymmetry (i.e., \( V = 0 \)), plotted as a function of \( \mathbf{k}_0 \), and Fig. 2(b) shows its closer view near \( \mathbf{k}_0 = 0 \). We choose \( (V_{pp}^0, d) = (-0.48V_{pp}^0, 2.97a) \), which gives the interaction strength between the neighboring sites of the circular chain in the dual-tight binding lattice \( t_0 = 0.1V_{pp}^0 \). The eight parabolic bands are arranged on a circle with a radius \( \Delta k = (2 - \sqrt{2})\pi/a \) by the Umklapp scattering [Eq. (8)], and they are strongly hybridized near \( \mathbf{k}_0 = 0 \). As a result, the originally degenerate eight states of the square lattices split into different energies, and exhibit the characteristic dispersion including parabolic bandbottoms at this high energy regime and the frilled band edge, which is flat up to the first order to \( \mathbf{k}_0 \).

At \( \mathbf{k}_0 = 0 \), \( \mathcal{H}_{\text{ring}} \) can be analytically diagonalized to obtain a set of energies

\[
E_m = h_0 - 2t_0 \cos q_m,
\]

which have the energy span of \( 4t_0 \), where \( q_m = (5\pi/4)m \) with \( m = 0, \pm 1, \pm 2, \pm 3, 4 \) is the wave number along the chain. Each element of the eigenvectors \( \mathbf{v}_m = (\mu_{m_3}, \mu_{m_2}, \mu_{m_1}, \ldots, \mu_{m})/\sqrt{8} \) (\( \mu_{m} = e^{i\theta_m} \)) is the coefficient to the Bloch bases \( |k(0)\rangle, |k(1)\rangle, \ldots, |k(7)\rangle \). Here the states with \( m = \pm s \) \( (s = 1, 2, 3) \) form twofold doublets, and belong to two-dimensional \( E_s \) irreducible representation of \( D_{6d} \) point group, while the \( m = 0 \) and 4 are non-degenerate, and belong to \( A_1 \) and \( B_2 \), respectively. If we disregard the \( z \)-position difference, the index \( m \) can be regarded as quantized angular momentum. The fact that there are 8 unique values for \( m \) as well as the fact
Fig. 2. (a) Electronic structures of octagonal vdW-QCs calculated by the 8-ring effective model. Blue and red arrows show the band opening by the interlayer and intralayer 2-wave mixing, respectively (see Sec. III B 3). (b) Detailed band structures near \( k = C_n \) [the region encircled by the black dashed rectangle in (a)] with index \( m \) indicating quantized angular momentum in 8-fold rotational symmetry. (c) LDOS at \( k = C_n \), characterized by \( m \), where the area of the circle is proportional to the squared wave amplitude, and red and blue circles represent the states in the upper and the lower layers, respectively.

that the eigenvalue of \( R(\pi/4)M_z \) is given by \( e^{-i\pi m/4} \) are the evidence that the quasicrystalline electronic states respect an 8-fold rotational symmetry.

The 8-wave resonant coupling also gives rise to a characteristic pattern in the wave function. Figure 2(c) shows the wave functions at \( k_0 = 0 \) where the hybridization is the most prominent. We can see that the wave amplitude is distributed on a limited number of sites in a 8-fold rotationally symmetric pattern.

3. Effects of interlayer interaction strength and other kinds of interaction

As discussed in Sec. II C we can calculate the quasicrystalline states of various octagonal vdW-QCs, which are either composed of other materials or different interlayer distance \( d \), by simply scaling the magnitude of the interlayer interaction \( t(q) \). Figure 3(a) shows the band structures of an octagonal vdW-QC with \( t(q) \) 2 times larger than that in Fig. 2. Although the interaction strength varies with \( q \), hereafter we label each system with \( t(q) \) at \( q \) showing the strongest quasicrystalline interaction, \( t_0 \) [\( = t(C_n) \)]. The stronger \( t_0 \) makes the 8-waves interact over a much wider area in the Brillouin zone, and the energy spacing between the quasicrystalline states larger. Accordingly, the \( m = \pm 2 \) states become flatter and the band curvature of the other states increases.

The density map in Fig. 3(b) shows the DOS of octagonal vdW-QCs with various \( t_0 \), where the white dashed line corresponds to the DOS for the system considered in Fig. 2. The white arrows show the pseudogaps by the interlayer and intralayer 2-wave interaction. (c) Three representative interaction residing in octagonal vdW-QCs; quasicrystalline interaction by 8-wave mixing (left), interlayer 2-wave mixing (middle), and intralayer 2-wave mixing (right). Red and blue contours show the Fermi surfaces of layer 1 and 2, respectively, black dashed circles show the wave numbers where the interaction occurs, and the blue dashed line shows the reciprocal lattice vector of layer 2 which mediates the interaction between the states in layer 1.

FIG. 2. (a) Electronic structures of octagonal vdW-QCs with the interlayer interaction \( |t_0| \) 2 times larger than that in Fig. 2. Blue and red arrows show the band opening by the interlayer and intralayer 2-wave mixing, respectively. (b) Density map of DOS calculated by using 32-waves and band edges of the quasicrystalline states [black dashed lines, Eq. (13)] with various \( t_0 \). The white dashed line corresponds to the DOS for the system considered in Fig. 2. The white arrows show the pseudogaps by the interlayer and intralayer 2-wave interaction. (c) Three representative interaction residing in octagonal vdW-QCs; quasicrystalline interaction by 8-wave mixing (left), interlayer 2-wave mixing (middle), and intralayer 2-wave mixing (right). Red and blue contours show the Fermi surfaces of layer 1 and 2, respectively, black dashed circles show the wave numbers where the interaction occurs, and the blue dashed line shows the reciprocal lattice vector of layer 2 which mediates the interaction between the states in layer 1.
in wider energy range, we used more (32 waves) bases to calculate the DOS in Fig. 3. We also plot the band edges of the quasicrystalline states [Eq. (10)] by black dashed lines. As \(|t_0|\) increases, the energy spacing between the edges increases and the height of the DOS peaks also grows rapidly. It should be noted that some band edges (e.g., \(m = \pm 2\)) lead to a series of characteristic spiky peaks in DOS and dips (pseudogaps) in between, while other edges are buried in the DOS of weakly coupled states. Thus, quasicrystalline features, such as local density of states (LDOS) with 8-fold rotational symmetry and relevant physical properties, are most prominent at the energies where the band edges coincide with the spiky peaks in DOS. As changing \(t_0\) does not break the symmetry of the Hamiltonian, it neither changes the symmetry nor the degeneracy of quasicrystalline states.

In addition to the features from the quasicrystalline 8-wave mixing, Fig. 3(b) shows the peaks and pseudogaps associated with other kinds of interaction. We plot the wave numbers associated with these interactions in Fig. 3(c), together with the Fermi surfaces. The middle panel shows the 2-wave mixing between the states in different layers \(\tilde{\Psi}_{m}^{0}\) which occurs when the Fermi surfaces of the two layers meet, while the right panel shows the 2-wave mixing between the states in the same layer assisted by the potential of the opposite layer \(\tilde{\Psi}_{s}^{0}\). Blue and red arrows in Figs. 2(a) and 3(a) show the band opening by the interlayer and intralayer 2-wave mixing, respectively, whose size also increases with \(|t_0|\). The interlayer interaction strength \(t(q)\) involved in the interlayer and intralayer 2-wave mixing is 0.472 and 1.49 times the interaction \(t_0\) for the 8-wave interaction. However, the intralayer mixing exhibits a band opening smaller than the interlayer mixing partly due to the two successive interlayer interaction and partly due to the energy difference between the states in opposite layers. At \(t_0 \to 0\), the 8-wave, interlayer 2-wave, and intralayer 2-wave mixing emerge at the energies \(E = \cos \sqrt{2} \pi + 1 (\approx 1.47)\), \(-2\cos \frac{\sqrt{2} \pi - 1}{2} (\approx 1.59)\), \(-4\cos \frac{\pi}{\sqrt{2}} (\approx 2.42)\) in unit of \(|V_{pp\pi}|\). It should be noted that the states and band opening arising from these three mixing are continuously connected to each other in the Brillouin zone [Figs. 3(a) and (c)]. Unlike the quasicrystalline 8-wave interaction, both the 2-wave mixing processes, labelled \(\alpha\) and \(\beta\) in Fig. 3(c), can occur in bilayer square lattices stacked at any rotation angle \(\theta\), i.e., at usual moiré superlattices. However, it is straightforward to show that \(\alpha\) and \(\beta\) occur at different energies \(\theta\) in the systems with \(\theta\) other than \(45^\circ\). And at small \(\theta\), the interlayer mixing appears at the energies closer to the band maximum at \(M\) than the intralayer mixing, while the order is reversed as \(\theta\) approaches \(45^\circ\).

4. Effects of interlayer potential asymmetry

Figure 4(a) shows the band dispersion near \(C_n\) of octagonal vdW-QCs under three different interlayer potential asymmetry, \(V = 0, 0.1|V_{pp\pi}|, 0.2|V_{pp\pi}|\). Since

\[
\Sigma^{-1}(H-h_0)\Sigma' = -(H-h_0) \quad \text{at} \quad k_0 = 0
\]

for \(\Sigma' = \text{diag}(i\sigma_y, i\sigma_y, i\sigma_y, i\sigma_y)\) and \(I\) is an 8 \(\times\) 8 unit matrix, regardless of the presence of the interlayer potential asymmetry, \(H\) has a particle-hole symmetry with respect to the energy \(E = h_0\). As \(V\) increases, however, the states with \(m = \pm 2\) at \(k_0 = 0\) lose the degeneracy, and all the band edges move away from \(E_{m=\pm 2} = h_0\).

We can obtain further insight on the effects of the interlayer potential asymmetry from the analytic expression of the energies at \(k_0 = 0\). The interlayer potential asymmetry couples the eigenstates of \(H_{\text{ring}}(k_0 = 0)\) that have angular momenta that differ by \(\pm 4\),

\[
\langle v_m | H_V | v_{m'} \rangle = \left\{ \begin{array}{ll} -V/2 & (m - m' \equiv 4 \mod 8), \\ 0 & \text{(otherwise)} \end{array} \right.
\]

since the diagonal elements of \(H_V\) work as a staggered potential with 1/4 period of the ring in the dual tight-binding lattice. Thus, the Hamiltonian matrix in the bases of the quasicrystalline states is reduced to four 2 \(\times\) 2 matrix.
for \((m, m') = (0, 4), (1, -3), (2, -2), (3, -1)\). As the quasicrystalline states, \(v_m\), originate from the resonant interaction between the degenerate states \(|k(n)\rangle\) in the two layers, the interlayer potential asymmetry breaks the 8-fold rotational symmetry of the states by lifting the degeneracy of \(|k(n)\rangle\). This reduces the allowed angular quantum numbers to \(s = 0, \pm 1, 2 \equiv m \equiv m' (\mod 4)\), which indicates a 4-fold rotational symmetry. We obtain the following energies and wave functions

\[
E_s = h_0 \pm \sqrt{4h_0^2 \cos^2(5\pi s/4) + V^2/4},
\]

\[
\Psi_s = c_m v_m + c_{m'} v_{m'},
\]

where \((c_m, c_{m'})\) is \((\sin(\phi/2), -\cos(\phi/2))\) for the upper band and \((\cos(\phi/2), \sin(\phi/2))\) for the lower band with \(\phi = \tan^{-1}(V/(4h_0 \cos q_m))\), and plot \(E_s\) against \(V\) in Fig. 4(b). The states with \(s = 1\) and \(s = -1\) are always degenerate due to the \(v_m = v_{-m}\) symmetry of the wave functions. At small \(V\), the interlayer interaction \(t_0\) suppresses the energy shift of \(s = 0, \pm 1, 2\) states \([\text{Eq. (16)}]\), in a similar manner to the way the interaction suppresses the Dirac point shift in twisted bilayer graphene with a small twist angle\([23]\). On the other hand, the two states with \(s = 2 (m = \pm 2)\) are composed of two degenerate quasicrystalline states, \(m = 2\) and \(m = -2\). Thus, their band edges shift as much as the applied bias in opposite directions and are not affected by the interlayer interaction \(t_0\). As \(V\) increases, the overall energy span of these resonant states increases, while the energy spacing between the adjacent states decreases.

The dashed arrows in Fig. 4(b) show the interplay between quasicrystalline states between \(v_m\) and \(v_{m'}\) by \(\mathcal{H}_V\), and Fig. 4(c) shows the degree of mixing in \(\Psi_{s=\pm 1}\), which we defined as \((1 - |c_m|^2 - |c_{m'}|^2) \times 100\%\). Systems with \(|t_0| < V\) exhibit stronger mixing, which will influence the transition behavior, such as the optical selection rule, in vdW-QCs. The states with \(s = 0\) exhibit a similar, but slightly weaker, mixing owing to the larger energy difference between \(E_m\) and \(E_{m'}\) in \(\Psi_{s=0}\). However, the states with \(s = 2\) are special in that the constituent states \(v_2\) and \(v_{-2}\) are always fully mixed, i.e., \(c_{m'} = -c_m\) for the upper band and \(c_{m'} = c_m\) for the lower band, regardless of the values of \(t_0\) and \(V\). Again, this is due to the degeneracy between the constituent states \(m = 2\) and \(m' = -2\).

We plot the LDOS of the upper and lower bands with \(s = 0, \pm 1, 2\) at \(V = 0.2|V_{pp}|\) in the top and bottom panels in Fig. 4(d), respectively. Due to the interlayer potential asymmetry, the wave functions \(\Psi_s\) are more or less spatially polarized to either layer. And the stronger the mixing, the more the wave functions are layer polarized; for example, \(\Psi_{s=\pm 1}\) exhibit more polarization than \(\Psi_{s=0}\). This is because

\[
\Psi_s = \frac{1}{2} (c_m + c_{m'}) (v_m + v_{m'}) + \frac{1}{2} (c_m - c_{m'}) (v_m - v_{m'}),
\]

where \((c_m, c_{m'}) \in \mathbb{R}\), and \(v_m + v_{m'}\) and \(v_m - v_{m'}\) are perfectly polarized to layer 2 and 1, respectively, since \(\mu_{m'} = -\mu_m\). Thus, as the mixing becomes stronger, the upper bands \((c_{m'} \approx -c_m)\) consist mostly of \(|k(n)\rangle\) with even \(n\) (i.e., layer 1) while the lower bands \((c_{m'} \approx c_m)\) consist mostly of \(|k(n)\rangle\) with odd \(n\) (i.e., layer 2). Again, the states with \(s = 2\) are special in that their wave functions \(\Psi_{s=2}\) are perfectly polarized to either layer regardless of the values of \(t_0\) and \(V\) because the constituent states \(v_2\) and \(v_{-2}\) are always fully mixed. This is similar to the case of an one-dimensional diatomic chain whose sublattices stop completely at the acoustic and optical modes.

The \(C_n\) in Fig. 1(c), which are associated with \(|k(n)\rangle\), remain the same since the interlayer potential asymmetry does not change the Umklapp scattering paths. Thus, the LDOS profile of each layer-polarized state, which is associated with \(C_n\) for even \(n\) (layer 1) or \(C_n\) for odd \(n\) (layer 2), is exactly consistent with the profile of each layer in the absence of the potential asymmetry [Fig. 2(c)]. Therefore, we can switch between the quasicrystalline states and their layer components by applying an electric field.

C. Dodecagonal quasicrystal

1. Hamiltonian

In dodecagonal vdW-QCs, the strongest resonant interaction occurs at \(k = 0\). Thus, by replacing \(k\) with \(k_0\), we can express the Hamiltonian of the resonant ring \(\mathcal{H}_{\text{ring}}\) in the absence of the interlayer and sublattice potential asymmetry by a \(24 \times 24\) matrix

\[
\mathcal{H}_{\text{ring}}(k_0) = \begin{pmatrix}
H^{(0)} & W^\dagger \\
W & H^{(1)} & W^\dagger \\
& W & H^{(2)} & W^\dagger \\
& & W & H^{(3)} & W^\dagger \\
& & & W & H^{(4)} \\
& & & & W & H^{(5)} \\
& & & & & W & H^{(6)} \\
& & & & & & W & H^{(7)} \\
& & & & & & & W & H^{(8)} \\
& & & & & & & & W & H^{(9)} \\
& & & & & & & & & W & H^{(10)} \\
& & & & & & & & & & W & H^{(11)} \\
\end{pmatrix},
\]

\[
H^{(n)}(k_0) = \begin{pmatrix}
\tilde{h}_A^{(n)} & \tilde{h}_B^{(n)} \\
\tilde{h}_A^{(n)} & \tilde{h}_B^{(n)} \\
\end{pmatrix}, \quad W = -t_0 \begin{pmatrix}
\omega & 1 \\
1 & \omega^* \\
\end{pmatrix},
\]

in the bases of \((|k^{(0)}\rangle, |k^{(1)}\rangle, |k^{(2)}\rangle, |k^{(3)}\rangle, \ldots, |k^{(11)}\rangle)\). Here, \(k^{(n)} = k_0 + Q_n\) for even \(n\) (layer 1) and \(k^{(n)} = k_0 - Q_n\) for odd \(n\) (layer 2), where \(|k^{(n)}\rangle\) and \(|k^{(n)}\rangle\) are \((|k^{(n)}\rangle, X), |k^{(n)}\rangle\rangle\) and \((|k^{(n)}\rangle, X')\rangle\rangle\) with the sublattices \(X\) and \(X'\) are arranged in the order of \((A, B)\) or \((A, B)\) for \(n = 0, 3\) in modulo of 4, and \((B, A)\) or \((B, A)\)
for \( n \equiv 1, 2 \). And \( h^{(n)}_{VX}(\mathbf{k}_0) = h_{VX} [R(-7n\pi/6)\mathbf{k}_0 + \mathbf{Q}_0] \), 
\( \omega = e^{2\pi i/3} \), and we neglect the \( \mathbf{k}_0 \) dependence of the interlayer matrix element \( t(\mathbf{q}) \).

In the given bases order, the Hamiltonian representing the interlayer and sublattice potential asymmetry are expressed by

\[
\mathcal{H}_V = \frac{V}{2} \text{diag}(I, -I, I, -I, I, -I, -I, I, -I, -I),
\]

and

\[
\mathcal{H}_\Delta = \frac{\Delta}{2} \text{diag} (\sigma_z, -\sigma_z, -\sigma_z, \sigma_z, \sigma_z, -\sigma_z, -\sigma_z, \sigma_z),
\]

respectively, where \( V \) represents the difference in the electrostatic energies between the two layers, \( \Delta \) is the difference between the on-site potentials between two sublattices, and \( I \) is a 2 \( \times \) 2 unit matrix. Then, the Hamiltonian of general dodecagonal vdW-QCs is given by

\[
\mathcal{H} = \mathcal{H}_{\text{ring}} + \mathcal{H}_V + \mathcal{H}_\Delta.
\]

### 2. Band structures and wave functions

We plot the band structures near \( \mathbf{C}_n \) of the dodecagonal vdW-QCs with \((V_{pp\pi}, d) = (-0.48V_{pp\pi}, 1.36a)\) and \( V = 0, \Delta = 0 \) in Fig. 5(a), and their closer view in (b). The twelve Dirac cones are arranged on a circle with a radius \( \Delta k = 4(2 - \sqrt{3})\pi/3a \), and they are strongly hybridized near \( \mathbf{k}_0 = 0 \) with \( t_0 = 0.046V_{pp\pi} \) to exhibit the characteristic dispersion including flat band bottoms, the Mexican-hat edges, and the frilled band edges.

At \( \mathbf{k}_0 = 0 \), \( \mathcal{H}_{\text{ring}} \) can be analytically diagonalized to obtain a set of energies (neglecting the constant energy)

\[
E_m \pm = t_0 \cos q_m \pm \sqrt{3t_0^2 \sin^2 q_m + (h_0 - 2t_0 \cos q_m)^2},
\]

where \( h_0 = h_{AB}(\mathbf{Q}_0) = h_{BA}(\mathbf{Q}_0) = -0.682V_{pp\pi} \), and \( q_m = (\pi/6)m \) with \( m = -5, -4, \cdots, 5, 6 \) is the wave number along the chain. Unlike the octagonal vdW-QCs in the minimal model (Sec. III B), which have one set of the hybridized states, the dodecagonal vdW-QCs show hybridization both in the conduction band and valence bands, which correspond to \pm in Eq. (23), respectively.

The energy scaling in conduction band is, however, much smaller than that in the valence band since the wave function of the conduction band of intrinsic graphene, having the same phases between the sublattices, suppresses the interlayer interaction by a factor of 3. The index \( m \) is a quantized angular momentum respecting the 12-fold rotational symmetry. The states with \( m = \pm s \) \((s = 1, 2, 3, 4, 5)\) form twofold doublets, while the \( m = 0 \) and \( 6 \) are non-degenerate. Note that the interaction responsible for the formation of the quasicrystalline states only weakly affects the spectrum at energies away from the momentum matching conditions; e.g., in a quasicrystalline twisted bilayer graphene there is no meaningful change on the Fermi velocity, since \( E_m^\pm \) are far from the Dirac point. Further detailed analysis on \( \mathcal{H}_{\text{ring}} \) can be found in Ref. 8.

Figure 5(c) shows the LDOS of the quasicrystalline states, where we can see that the wave amplitude distribute selectively on a limited number of sites in a characteristic 12-fold rotationally symmetric pattern. The wave functions for \( E_m^\pm \) are \( \mathbf{v}_m^\pm = (1/\sqrt{12})(\mu_m^{-5}, \mu_m^{-4}, \mu_m^{-3}, \cdots, \mu_m^0) \otimes (c_{m,1}^\mp, c_{m,2}^\pm) \) \((\mu_m = e^{i\phi_m})\), where \((c_{m,1}^+, c_{m,2}^+) = (\sin(\phi_m/2), \cos(\phi_m/2))\) and \((c_{m,1}^-, c_{m,2}^-) = (\cos(\phi_m/2), -\sin(\phi_m/2))\) are the coefficients of the sublattices arranged in the order of the bases of Eq. (18), and \( \phi_m = \tan^{-1}(h_0 - 2t_0 \cos q_m)/(\sqrt{3}t_0 \sin q_m) \). Since the Hamiltonian has a symmetry of

\[
\Sigma = \text{diag}(\sigma_x, \sigma_x, \sigma_x, \sigma_x, \sigma_x) \equiv \mathcal{H}_{\text{ring}}
\]

at \( \mathbf{k}_0 = 0 \), the states with angular momentum \( m \) and \( -m \) are degenerate and \( \mathbf{v}_m^\pm = \sigma_x (\mathbf{v}_m^0)^\dagger \), and it is straightforward to show that their LDOS profiles are exactly the same to each other. Figure 5(c) also shows that the states with \( \pm m \) exhibit LDOS profiles which look similar to those of \( \mathbf{6} \mp \mp \); \( \mathbf{v}_m \) clearly shows that the states with \( m = 0 \) and \( 6 \), and also the states with \( m = 3 \) and \( -3 \) have LDOS that are exactly the same as each other, while the LDOS profiles of the other states (i.e., \( m = \pm 1 \) and \( \pm 5 \), and also \( m = \pm 2 \) and \( \pm 4 \)) become different as \( |t_0| \) grows. Likewise, the \( \pm m \) states in the conduction band exhibit LDOS profiles which look similar to the \( 3 \mp \pm \) ones in the valence band in the systems with a small \( |t_0/V_{pp\pi}| \).

### 3. Effects of interlayer interaction strength and other kinds of interaction

Figure 6(a) shows the valence band structures of a dodecagonal vdW-QC with an interlayer interaction \( t(\mathbf{q}) \) that is 2 times larger than the one in Fig. 5. The energy spacing between the quasicrystalline states becomes larger, and the flat band area of \( m = 6 \) \((m = \pm 4)\) state in the valence band in Fig. 6(a) is approximately 2-times (5-times) as large as that in Fig. 5(a), and it is 28-times (70-times) bigger than the flat band area of magic-angle twisted bilayer graphene. As a greater number of the electronic states are involved in the flat bands, we expect to see stronger electron-electron interacting effect. The density map in Fig. 6(b) shows the DOS of dodecagonal vdW-QCs with various \( t_0 \) calculated by using 182-wave bases. The white dashed line corresponds to the DOS.
for the system considered in Fig. 5 and the black dashed lines show the band edges of the quasicrystalline states [Eq. (23)].

The systems with larger $|t_0|$ exhibit higher DOS peaks owing to the increase of the flat band area in the $k$ space. Not every quasicrystalline state leads to a DOS displaying spiky peaks interspersed with pseudogaps, so quasicrystalline features would be most prominent at the energies where the band edges coincide with the spiky peaks in DOS, especially at the $m = 5, 6, \pm 4, \pm 3$ states in the valence band and the $m = 6, \pm 1, \pm 4$ states in the conduction band. Again, changing $t_0$ neither changes the symmetry nor degeneracy of quasicrystalline states. In most practical parameter ranges, a system with larger $|t_0|$ exhibits a larger energy spacing between the quasicrystalline states in both the conduction and valence bands. Note that, in the systems with extremely strong interlayer interaction ($|t_0| > h_0/2$), the energy spacing in the conduction band may decrease as $|t_0|$ increases (Appendix C). Such a condition, however, is hard to be achieved in the most practical systems. Thus, hereafter, we will consider the systems with $|t_0| < h_0/2$ only.

The two DOS peaks and the dips in between, which originate at $E = -0.5|V_{pp\pi}|$ when $t_0 = 0$, show to the band edges and pseudogaps arising from intralayer 2-wave mixing. We plot the band opening associated the interlayer and intralayer mixing as the blue and red arrows in Figs. 5(a) and 5(b), respectively, and visualize these interactions in Fig. 5(c). The band opening by the

FIG. 5. Plots similar to Fig. 2 for dodecagonal vdW-QCs calculated by the 12-ring effective model. Blue and red arrows in (a) show the band opening by the interlayer and intralayer 2-wave mixing, respectively. The top and bottom panels in (b) show the quasicrystalline states in the conduction band and valence band, respectively.

FIG. 6. (a) Electronic structures of dodecagonal vdW-QCs with the interlayer interaction $|t_0|$ 2 times larger than that in Fig. 4. Blue and red arrows show the band opening by the interlayer and intralayer 2-wave mixing, respectively. (b) Density map of DOS calculated by using 182-waves and band edges of the quasicrystalline states [black dashed lines, Eq. (23)] with various $t_0$. The white dashed line corresponds to the DOS for the system considered in Fig. 5. (c) Plots similar to Fig. 3(c) in dodecagonal vdW-QCs.
2-wave mixing in the conduction band is much smaller than that in the valence band for the same reason as the band opening via the 12-wave mixing (Sec. III C 2). Although the interlayer interaction strength \( t(|q|) \) involved in the intralayer 2-wave mixing is about 1.50 times the interaction strength in the interlayer 2-wave mixing, the intralayer mixing exhibits smaller band opening since it involves two successive interlayer interaction between the states with different energies. At sufficiently large \( |t_0| \) (> 0.08\( |V_{pp\pi}| \)), however, the intralayer interaction gives band opening throughout the entire Brillouin zone in the valence band except in the vicinity of the quasicrystalline states. This makes the quasicrystalline states with \( m = 6, \pm 1 \) be easily observable in a specific energy window as the quasicrystalline states remain after the weakly coupled bands become gapped. At \( t_0 \rightarrow 0 \) limit, the interlayer 2-wave mixing emerges at an energy that is the same as the 12-wave mixing, while the intralayer 2-wave mixing emerges at the energy much closer to \( E = 0 \). Again, the states and band opening arising from these three different mixings are continuously connected to each other in the Brillouin zone [Figs. 4(a) and (c)]. The 2-wave mixing can occur in bilayer honeycomb lattices stacked at any rotation angle \( \theta \), and \( \alpha \) and \( \beta \) occur at different energies when \( \theta \neq 30^\circ \). At small \( \theta \), the interlayer mixing appears at energies closer to the band edge at \( K \) (and \( K' \)) than the intralayer mixing, while the order is reversed as \( \theta \) approaches 30°.

4. Effects of interlayer potential asymmetry

Figures 7(a) and (d) show the dispersion in the conduction band and valence band near \( C_n \) of dodecagonal vdW-QCs under three different interlayer asymmetric potential, \( V = 0, 0.1 |V_{pp\pi}|, 0.2 |V_{pp\pi}| \). Again, the states with \( m = \pm 3 \) at \( k_0 = 0 \) lose their degeneracy, and all the band edges, in both the conduction band and valence band, move away from \( E_{m=\pm 3} \) as \( V \) increases.

At \( k_0 = 0 \), the interlayer potential asymmetry couples the eigenstates of \( \mathcal{H}_{\text{ring}} \), \( |v_m^b \rangle \) (\( b \) is + for conduction band and − for valence band), having their angular momenta differ by \( \pm 6 \),

\[
\langle v_{m'}^b | \mathcal{H}_V | v_{m}^b \rangle = \begin{cases} 
-\frac{1}{V} (\epsilon_{m',1}^b c_{m,1}^b + c_{m',2}^b c_{m,2}^b) \\
0 
\end{cases} \quad (m-m' \equiv 6 \text{ (mod 12)}),
\]

(27)

since the diagonal elements of \( \mathcal{H}_V \) work as a staggered potential with 1/6 period of the ring in the dual tight-binding lattice. Thus, the Hamiltonian matrix is reduced to 4 \( \times \) 4 matrices

\[
\mathcal{H}_{m,m'} = \begin{pmatrix} 
C_m & -\frac{V}{2} R^{-1}\left(\frac{\phi_{m'} - \phi_{m}}{2}\right) \\
-\frac{V}{2} R\left(\frac{\phi_{m'} - \phi_{m}}{2}\right) & C_{m'} 
\end{pmatrix},
\]

\[
C_m = \begin{pmatrix} 
E_m & 0 \\
0 & E_m \end{pmatrix},
\]

(26)
in the bases of \( (v_{m}^+, v_{m}^-, v_{m'}^-, v_{m'}^+) \) for \( (m, m') = (0, 6), (1, -5), (2, -4), (3, -3), (4, -2), (5, -1) \), where \( R(\phi) \) is a rotation matrix. The electronic states lose the 12-fold rotational symmetry, since \( \mathcal{H}_V \) lifts the degeneracy of \( |k^{(0)}\rangle \) in different layers, and are characterized by the angular quantum number \( s = 0, \pm 1, \pm 2, 3 \) \( m \equiv m' \pmod{6} \) for a 6-fold rotational symmetry. We obtain two band edges \( E_{\pm}^V \) in the conduction band and another two \( E_{\pm}^V \) in the valence band by diagonalizing Eq. (26). These are plotted against \( V \) in Figs. 7(b) and (e), for the conduction and valence bands, respectively. It is straightforward to show that the states with a quantum number \( s \) and \( -s \) \( (s = 1, 2) \) are degenerate, since the reduced Hamiltonian satisfies

\[
\Sigma^{-1}\mathcal{H}_{m,m'}\Sigma = \mathcal{H}_{-m,-m'},
\]

\[
\Sigma = \text{diag}(1, -1, 1, -1),
\]

(27)

In most practical cases, Eq. (26) can be further reduced to two 2 \( \times \) 2 matrices, since the interaction between the state in the conduction band and the state in valence band is almost negligible. This is partly due to the large energy difference and partly due to \( (\phi_m - \phi_{m'})/2 \approx 0 \) (since \( \phi_m \approx \phi_{m'} \approx 90^\circ \)).

The dashed arrows in Figs. 7(b) and (e) show the interaction between quasicrystalline states between \( v_{m}^+ \) and \( v_{m}^- \) by \( \mathcal{H}_V \). The states in the conduction band exhibit larger mixing between the constituent quasicrystalline states than those in the valence band, due to the smaller energy difference (not shown). And, similar to the octagonal vdW-QCs [Fig. 4(c)], materials with weaker \( |t_0| \) under larger \( V \) experience larger energy shift, mixing, and accordingly larger spatial layer-polarization because of Eq. (17) (Appendix D). Again, the states with \( s = 3 \) are special in that the constituent states \( v_{m=3}^+ \) and \( v_{m=-3}^- \) are always fully mixed regardless of the values of \( t_0 \) and \( V \), due to the degeneracy between \( E_{m=3}^+ \) and \( E_{m=-3}^- \).

We plot the LDOS of the states in the conduction band and valence band with \( s = 0, \pm 1, \pm 2, 3 \) at \( V = 0.2 |V_{pp\pi}| \) in Figs. 7(c) and (f), respectively, where the top and bottom panels in each figure show the LDOS of the upper and lower bands, respectively. Again, the stronger the mixing, the more the wave functions are layer polarized, and the wave functions \( \Psi_{s=3}^\pm \) are mostly polarized to either layer even at very weak \( V \). The LDOS profile of each layer-polarized state is exactly consistent with the profile of each layer in the absence of the potential asymmetry [Fig. 4(c)], since \( \mathcal{H}_V \) does not change the Umklapp scattering paths.

In dodecagonal vdW-QCs with sublattice symmetry (i.e., \( \Delta = 0 \)), an interlayer potential asymmetry does not open a gap at the Dirac point. This is because the coexistence of the time reversal symmetry and the in-place \( C_2 \) (180°) rotation symmetry requires vanishing of the Berry curvature at any nondegenerate point the energy band [18], and this guarantees the robustness of band touching points in two-dimensional systems [20]. Just like
5. Effects of sublattice potential asymmetry

We plot the conduction band and valence band near \( C_n \) of a dodecagonal vdW-QC with sublattice potential asymmetry of \( \Delta = -0.60 V_{ppr} \) in Figs. 7(b) and (e), respectively, and plot the bands in the absence the asymmetry in (a) and (d) as a reference. As usual in most transition-metal dichalcogenides, \( \Delta \neq 0 \) in the current model makes a band opening as large as \( \Delta \) at the energy range centered at \( E = 0 \). Unlike the interlayer potential asymmetry, however, we can see that breaking the sublattice symmetry does not make dramatic change to the band structures near the quasicrystalline states.

The potential which breaks the sublattice symmetry, \( \mathcal{H}_\Delta \), couples the eigenstates of \( \mathcal{H}_{\text{ring}} \), \( \psi^b_m \), whose angular momenta differ by \( \pm 3 \),

\[
\langle \psi^b_{m'} | \mathcal{H}_\Delta | \psi^b_m \rangle = \begin{cases} 
\frac{(1-i)\Delta}{4} (c^b_{m',1} c^b_{m,1} - c^b_{m,2} c^b_{m',2}) & (m - m' \equiv -3 \, (\text{mod } 12)), \\
\frac{(1+i)\Delta}{4} (c^b_{m',1} c^b_{m,1} - c^b_{m,2} c^b_{m',2}) & (m - m' \equiv 3 \, (\text{mod } 12)), \\
0 & \text{(otherwise)},
\end{cases}
\]

since the diagonal elements of \( \mathcal{H}_\Delta \) work as a potential with 1/3 period of the ring in the dual tight-binding lattice. Thus, \( \mathcal{H}_\Delta \) couples quasicrystalline states with four different \( m \), \( (m_1, m'_1, m_2, m'_2) = (0, 3, 6, -3), (1, 4, -5, -2), (-1, 2, 5, -4) \), and the Hamil-
The Hamiltonian matrix is reduced to $8 \times 8$ matrices

$$\hat{\mathcal{H}}_{m_1,m'_1,m_2,m'_2} = \begin{pmatrix} C_{m_1} & D_{m_1,m'_1} & 0 & D_{m_2,m_1}^\dagger \\ D_{m_1,m'_1}^\dagger & C_{m'_1} & D_{m_1,m_2} & 0 \\ 0 & D_{m'_1,m_2}^\dagger & C_{m_2} & D_{m_2,m'_2} \\ D_{m_2,m_1} & 0 & D_{m_2,m'_2}^\dagger & C_{m'_2} \end{pmatrix},$$

$$D_{m_a,m_b} = \frac{(1 + i)\Delta}{4} \begin{pmatrix} \cos \frac{1}{2}(\phi_{m_a} + \phi_{m_b}) & \sin \frac{1}{2}(\phi_{m_a} + \phi_{m_b}) \\ \sin \frac{1}{2}(\phi_{m_a} + \phi_{m_b}) & \cos \frac{1}{2}(\phi_{m_a} + \phi_{m_b}) \end{pmatrix}.$$

in the bases of $(\mathbf{v}_{m_1}, \mathbf{v}_{m'_1}, \mathbf{v}_{m_2}, \mathbf{v}_{m'_2}, \mathbf{v}_{m_1}', \mathbf{v}_{m'_1}', \mathbf{v}_{m_2}', \mathbf{v}_{m'_2}')$. The electronic states lose the 12-fold rotational symmetry, and are characterized by the angular quantum number $s(\equiv 0, \pm 1) \equiv m_a, (\mod 3)$ $(m_1 = m_1, m_2, m_1', m_2')$ for a 3-fold rotational symmetry. Again, the states with a quantum number $s = 1$ and $s = -1$ are degenerate, due to the symmetry of the Hamiltonian. And in most practical cases, Eq. (29) can be further reduced to four $2 \times 2$ matrices, since the interaction between the state in the conduction band and the state in valence band is almost negligible due to the large energy difference.

Figures 8(c) and (f) show the energies $E_s^\pm$ in the unit of $|V_{pp\pi}|$ plotted against $\Delta$. Unlike the interlayer potential asymmetry, $\Delta$ merely shifts the energies slightly away from the Dirac point and does not make dramatic change to the quasicrystalline states, which is consistent with the band structures in Figs. 8(b) and (f). This is because, $|c_{m,1}^b| \approx |c_{m,2}^b|$ in most practical systems with $|t_0| < \hbar \omega / 5 (\phi_m \approx 90^\circ)$ at this high energy regime. Thus, the potential, which has the opposite sign between the sublattices, is almost cancelled in the intraband interaction $(b = b')$, due to the phase cancellation. On the other hand, although materials with much higher $|t_0|$ have finite contribution from the sublattice phases in the intraband interaction, the overall interaction is still very weak.
weak since the energy difference between the quasicrystalline states increases as $|t_0|$ grows. In any system, the interband interaction ($b \neq b'$) is always negligible due to the large energy difference. Thus, dodecagonal vdW-QCs composed of transition metal dichalcogenides will also exhibit the quasicrystalline states and DOS analogous to the quasicrystals composed of bilayer graphene. If the two layers have different $\Delta$, other than a simple sign difference, the degeneracy of the $m = \pm 3$ states is lifted but the other states remain almost the same.

IV. CONCLUSIONS

We investigated the electronic structures of quasicrystals composed of the incommensurate stack of atomic layers (vdW-QCs) with various rotational symmetry and various interlayer interaction strength. We found the resonant coupling of the monolayer states which gives quasicrystalline order in each quasicrystal symmetry, and calculate the quasi-band dispersion and wave functions respecting the rotational symmetry of quasicrystal. Although the quasicrystalline states coexist in energy with weakly coupled states (e.g., the states from 2-wave mixing) in general, we showed that some quasicrystalline states, which are usually obscured by additional weakly coupled states, are more prominent in quasicrystals with strong interlayer interaction. Besides, we showed that we can switch the states between quasicrystalline configuration and its layer components, and also mix the states by lifting the layer degeneracy, e.g., by applying electric field. We analytically showed that the states in the middle are always fully mixed and 100% layer polarized regardless of the magnitude of the interlayer asymmetry. On the other hand, hexagonal lattices with sublattice potential asymmetry, e.g., transition metal dichalcogenide or hexagonal Boron Nitride, give the quasicrystalline order quite similar to those in the system in the absence of sublattice asymmetry. This is the first theoretical work which investigated the formation of quasicrystalline states in vdW-QCs for every possible rotational symmetry and sublattice symmetry in two-dimensional system, which will lead to extended exploration of rich quasicrystal physics in designer quasicrystals.

Note added. During the completion of this work, we became aware of recent theoretical works on the pressure and electric field dependence of quasicrystalline electronic states in 30° twisted bilayer graphene.\(^\text{[23]}\)

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Appendix A: Quasicrystalline states with weaker interaction

As mentioned in Sec. III A, the sets of the waves in Figs. 1(b) and (e) are not the only set of states which show the resonant coupling in each system. We can find more sets of states, with different wave numbers, showing the resonant interaction respecting the rotational symmetry of the quasicrystals. For example, the eight states $\mathbf{k}$ (red) and $\mathbf{k}'$ (blue) in Fig. 1(a) also form a circular chain in the dual tight-binding lattice. Note that $\hat{\mathbf{k}}$ for these states ($= 0$) is different from that for the states in Fig. 1(b). These states are mapped to $\mathbf{k}$ (red) and $\mathbf{k}$ (blue) in the first Brillouin zone, experience a resonant interaction, and form quasicrystalline states. Figure 9(c) shows the band structures near the quasicrystalline states arising from these eight states. It should be noted that, however, the strength of the resonant interaction, $|t'(\mathbf{q})|$, for the states in Fig. 1(a) is much weaker than that for the states in Fig. 1(b). This is because $|t'(\mathbf{q})|$ decays fast as $|\mathbf{q}|$ grows, and the former states have the chain with a longer segment length ($=|\mathbf{q}|$). Thus, the band opening in Fig. 9(c) is much smaller than that in Figs. 2(a) and (b). Dodecagonal vdW-QCs also have more sets of states showing the resonant interaction. In most systems, however, such states can be mostly neglected since their interaction strengths are very weak, and they are also mixed with other types of interaction (e.g., 2-wave mixing). Thus, the sets in Figs. 1(b) and (e) give the strongest interaction, i.e., largest energy separation and clear quasicrystalline order, since these states form the rings with the shortest distance between neighboring states in the dual tight-binding lattices.

Appendix B: Particle-hole symmetry of the Hamiltonian of octagonal quasicrystal

By considering only the nearest neighbor pairs in the intralayer interaction, the ring Hamiltonian $H_{\text{ring}}$ of the octagonal vdW-QCs [Eq. (1)], up to the first order to \(\mathbf{k}_0 = (k_{0,x}, k_{0,y})\), can be transformed to
FIG. 9. (a) and (b) Plots similar to Figs. 1(b) and (c) for the next strongest quasicrystalline interaction in octagonal vdW-QCs, where $\mathbf{k} = 0$. (c) Electronic structures near the second dominant quasicrystalline states of octagonal vdW-QCs calculated by the 8-ring effective model.

$$U^{-1} \mathcal{H}_\text{ring}(\mathbf{k}_0) U = \mathcal{H}'_\text{ring}(\mathbf{k}_0) = \begin{pmatrix} H^{(-3)} & C^* & C & \cdots \vphantom{C^*} \\ C & H^{(-2)} & C^* & \cdots \vphantom{C^*} \\ & \vdots & \ddots & \ddots \vphantom{C^*} \\ C^* & & \cdots & C & H^{(4)} \end{pmatrix}$$  \hspace{1cm} (B1)

with a transformation matrix

$$U = (v_{-3}, v_{-2}, v_{-1}, \cdots, v_3, v_4), \hspace{1cm} (B2)$$

where $v_m = \frac{1}{\sqrt{8}} (\mu_{m-1}^3, \mu_{m-2}^3, \cdots, \mu_m^3, \mu_4^3)^T$ ($\mu_m = e^{i5\pi m/4}$) is the eigenstate of the quasicrystalline state with a quantized angular momentum of $m$, $H^{(m)}_\text{ring} = h_0 - 2t_0 \cos(5\pi m/4)$, and $C(\mathbf{k}_0) = \sin(\sqrt{2\pi})a V_{pp\pi}(k_{0,x} - ik_{0,y})$. Then, it is straightforward to show that $\mathcal{H}'_\text{ring}$ has a particle-hole symmetry with respect to the energy $E = h_0$, $\Sigma^{-1}(\mathcal{H}'_\text{ring} - h_0 \mathbb{I}) \Sigma = -(\mathcal{H}'_\text{ring} - h_0 \mathbb{I})$, where $\mathbb{I}$ is an $8 \times 8$ unit matrix. This immediately demonstrates that if $\Psi(\mathbf{k}_0)$ is an eigenstate of $\mathcal{H}'_\text{ring}$ with an energy of $h_0 + E$, then $\Sigma^{-1} \Psi(\mathbf{k}_0)$ is an eigenstate of energy $h_0 - E$.

**Appendix C: Band edges of quasicrystalline states of dodecagonal vdW-QCs with various $t_0$**

Figure 10 shows the band edges of quasicrystalline states of dodecagonal vdW-QCs [Eq. (23)] with various interlayer interaction strength $t_0$ up to very strong interaction regime. We can see that the energy spacing between the quasicrystalline states increases as $|t_0|$ increases in most practical interaction strength, i.e., $|t_0| < h_0/2$ ($h_0 = -0.682 V_{pp\pi}$), while the energy spacing in the conduction band decreases as $|t_0|$ exceeds $h_0/2$.

**Appendix D: Energies and wave functions of dodecagonal quasicrystal with interlayer potential asymmetry**

Figures 7(b) and (e) show that the states with $s = 0$ determine the energy span of the resonant states in the presence of the interlayer potential asymmetry, in both the conduction band and valence band. Equation (26) shows the coupling between the quasicrystalline states with angular momentum $m$ by interlayer potential asymmetry. In most practical cases, the matrix can be further reduced to two $2 \times 2$ matrices,

$$\hat{\mathcal{H}}_{m,m'}^\pm = \begin{pmatrix} E_m^\pm & -V/2 \vphantom{-V/2} \\ -V/2 & E_m'^\pm \vphantom{-V/2} \end{pmatrix},$$

for the conduction band ($\hat{\mathcal{H}}_{m,m'}^+$) and valence band ($\hat{\mathcal{H}}_{m,m'}^-$), since the interaction between the state in the conduction band and the state in valence band is almost negligible due to the large energy difference. For the cou-
J. C. Slater and G. F. Koster, Phys. Rev. 10 (1958).  
G. Trambly de Laissardière, D. Mayou, and L. Magaud, Nano Lett. 10, 804 (2010).

The wave functions of the higher energy states in both the conduction band and valence bands are \( \Psi_{s=0}^{\pm} = \sin(\phi/2)v_{m=0}^{\pm} + \cos(\phi/2)v_{m=6}^{\pm} \), and the lower energy states are \( \Psi_{s=0}^{\pm} = \cos(\phi/2)v_{m=0}^{\pm} - \sin(\phi/2)v_{m=6}^{\pm} \), where \( \phi = \tan^{-1}(-V/(6t_0)) \) for valence band and \( \tan^{-1}(V/(2t_0)) \) for conduction band. As \( \phi \) becomes close to 90°, i.e., in materials with smaller \( |t_0| \) and \( |V| > |t_0| \), \( \Psi_{s=0}^{\pm} \) becomes \((1/\sqrt{2})(v_{6}^{\pm} + v_{0}^{\pm})\) for the upper state and \((1/\sqrt{2})(v_{6}^{\pm} - v_{0}^{\pm})\) for the lower state. Since \( v_{0}^{\pm} = (1/\sqrt{24})(1,1,1,1,\ldots,1) \otimes (1, \pm 1) \) and \( v_{6}^{\pm} = (1/\sqrt{24})(-1,1,-1,1,\ldots,1) \otimes (1, \pm 1) \), \( \Psi_{s=0}^{\pm} \) becomes polarized to either layer, i.e., the state is mostly composed of the Bloch bases with \( n \) of even (layer 1) or odd (layer 2) numbers, in the systems with small interlayer interaction strength \( |t_0| \). On the other hand, the states with \( s = 3 \) are always 100% polarized to either layer.

Note that the ratio between \( V_{pp\sigma}^{0} \) and \( V_{pp\pi}^{0} \) of the hexagonal lattices used in this work is different from that used in the previous works (\( V_{pp\sigma}^{0} \approx 0.48 \text{ eV} \) and \( V_{pp\pi}^{0} \approx -2.7 \text{ eV} \)) on the twisted bilayer graphene \(^{20,21,23}\) and graphene on hexagonal boron nitride \(^{20}\). In this work, we scaled \( V_{pp\sigma}^{0} \) by a factor of 1.25, while keeping \( V_{pp\pi}^{0} \approx 0.48 \text{ eV} \), to compensate the deviation of the Fermi velocity of a pristine graphene due to the summation over sites in the hopping range and make the band dispersion and the energies of the van Hove singularities consistent with the experimental result. \(^{21,23}\)

M. Koshino, Phys. Rev. B 88, 115409 (2013).

G. Yu, M. I. Katsnelson, and S. Yuan, Pressure and electric field dependence of quasicrystalline electronic states in 30° twisted bilayer graphene, arXiv preprint arXiv:2003.11879 (2020).

P. Moon and M. Koshino, Phys. Rev. B 90, 155406 (2014).