Energy Spectrum and Quantum Hall Effect in Twisted Bilayer Graphene

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(Dated: March 2, 2012)

We investigate the electronic structure and the quantum Hall effect in twisted bilayer graphenes with various rotation angles in the presence of magnetic field. Using a low-energy approximation which incorporates the rigorous interlayer interaction, we computed the energy spectrum and the quantized Hall conductivity in a wide range of magnetic field from the semi-classical regime to the fractal spectrum regime. In weak magnetic fields, the low-energy conduction band is quantized into electron-like Landau levels and hole-like Landau levels at energies below and above the van Hove singularity, respectively, and the Hall conductivity sharply drops from positive to negative when the Fermi energy goes through the transition point. In increasing magnetic field, the spectrum gradually evolves into a fractal band structure called Hofstadter’s butterfly, where the Hall conductivity exhibits a non-monotonic behavior as a function of Fermi energy. The typical electron density and magnetic field amplitude characterizing the spectrum monotonically decrease as the rotation angle is reduced, indicating that the rich electronic structure may be observed in a moderate condition.

I. INTRODUCTION

The electronic structure of bilayer graphene is highly sensitive to the stacking geometry between the two layers. The interlayer interaction in bilayer graphene with regular AB stacking modifies the linear dispersion of monolayer graphene into the quadratic dispersion, where an electron behaves as a massive particle. On the other hand, the recent epitaxial growth technique realized twisted bilayer graphene (TBG) in which two layers are stacked with a random rotation angle. The unit cell area of TBG can be more than a thousand times as large as that of monolayer graphene, due to slightly misoriented lattice vectors of two layers. Such an atomic configuration was observed as Moiré pattern in the scanning tunneling microscopy and TBG was also fabricated in different methods such as folding of mechanically exfoliated graphenes and unzipping of carbon nanotube.

The electronic structure of TBG shows a linear band dispersion near Dirac points rather than the massive dispersion of AB stacked bilayer, suggesting relatively weak interlayer interaction. In strong magnetic fields, however, it is predicted that the spectrum exhibits a fractal structure called Hofstadter’s butterfly, in which a series of energy gaps appears in a self-similar fashion. The fractal band structure generally occurs in periodic systems when the magnetic flux per a unit cell is comparable to \( \frac{h}{e} \), and this condition is realized in TBG in a reasonable magnetic-field range owing to the large unit cell. The fractal band structure and the quantum Hall effect were theoretically studied for TBG in the strong magnetic field regime using a continuous interlayer coupling model. Experimentally, the energy spectrum of the twisted graphene stacks in magnetic field was probed in the transport measurements and the magneto-optical absorption, while the fractal band structure has not yet been observed.

In this paper, we investigate the electronic spectrum and the quantum Hall effect in TBG with various rotation angles and magnetic fields. We calculate the spectrum by including a limited number of bases which are significant in the low-energy spectrum, while rigorously taking account of transfer integrals between lattice points on the different layers. Using this method, we describe the spectral evolution in a wide range of magnetic field, from the semiclassical Landau levels in the weak field regime to the fractal band structure in the strong field regime.

In weak magnetic fields, we find that the low-energy conduction band is quantized into electron-like Landau levels and hole-like Landau levels at energies below and above the van Hove singularity, respectively, in accordance with the topological change of the Fermi surface from electron-type to hole-type at the band saddle point. As a consequence, the quantized Hall conductivity abruptly jumps from positive to negative when the Fermi energy goes through the transition point. In increasing magnetic field, the electron and hole Landau levels begin to be mixed and gradually evolve into the fractal band structure. We calculate the quantized Hall conductivity for each single gap, and demonstrate that it changes non-monotonically as a function of Fermi energy and magnetic field.

II. THEORETICAL METHODS

A. Atomic structure

TBG is characterized by the relative rotation angle \( \theta \) and the relative translation vector between two graphene layers. When the lattice structures of the two layers are commensurate, we can define the primitive lattice vectors \( \mathbf{L}_1 \) and \( \mathbf{L}_2 \) as the least common multiples of the unit vectors on the two layers. \( \mathbf{L}_1 \) is written by integers \( m, n, m', n' \) as

\[
\mathbf{L}_1 = m \mathbf{a}_1^{(1)} + n \mathbf{a}_2^{(1)} = m' \mathbf{a}_1^{(2)} + n' \mathbf{a}_2^{(2)},
\]

where \( \mathbf{a}_1^{(1)} \) and \( \mathbf{a}_2^{(1)} \) are the lattice vectors of the layer 1, while \( \mathbf{a}_1^{(2)} \) and \( \mathbf{a}_2^{(2)} \) are the lattice vectors of the layer 2 defined in Fig. 1(a). \( \mathbf{L}_2 \) is obtained by rotating
L_1 by 60°. By appropriate choice of lattice vectors a_i^{(l)}, the indices (m', n') can be made equal to (n, m), and thus TBG is specified by a single pair of integers (m, n). The rotation angle θ is related to (m, n) by

\[ \cos \theta = \frac{1}{2} \left( \frac{m^2 + n^2 + 4mn}{m^2 + n^2 + mn} \right) \]

and the lattice constant of TBG by

\[ L = |L_1| = a \sqrt{m^2 + n^2 + mn}, \]

where \( a = |a_1| = |a_2| \approx 0.234 \text{ nm} \) is the lattice constant of monolayer graphene. The area of TBG unit cell is given by \( S = |L_1 \times L_2| = (\sqrt{3}/2)L^2 \).

Figure 1(a) shows the atomic structure of TBG with (m, n) = (1, 2) and \( \theta = 21.8° \). Throughout the paper, we set the coordinates \((x, y)\) on graphene plane so that \( y\) axis is parallel to \( L_2 \), and \( z \) to the direction perpendicular to the plane. We ignore the relative translation between two layers, which makes a minor difference in the electronic structure when the unit cell is large enough.

Figure 1(b) shows the extended Brillouin zone of TBG with \( \theta = 21.8° \). The two large hexagons represent the first Brillouin zones of layer 1 and 2, respectively. \( K^{(l)} \) and \( K^{(l')} \) denote the two inequivalent corners of layer \( l \), which are Dirac points in the single-layer band structure. The four Dirac points of \( K^{(1)} \), \( K^{(1')} \), \( K^{(2)} \), and \( K^{(2')} \) are folded back to two Dirac points, \( K \) and \( K' \), in the reduced Brillouin zone \( \Gamma \). Figure 2 shows the atomic structures of four different TBGs to be considered in following sections. They are specified by \((m, n) = (3, 4), (8, 9), (12, 13), \) and \((22, 23)\), and the rotation angles \( \theta = 9.43°, 3.89°, 2.65°, \) and \( 1.47° \), respectively. As the angle \( \theta \) decreases, the size of the unit cell enlarges and the Moiré pattern becomes evident.

### B. Tight-binding model

In a tight-binding model in terms of \( p_z \) atomic orbitals, the Hamiltonian of TBG at zero magnetic field is written as

\[ H_{\text{TBG}}^{B=0} = - \sum_{(i,j)} t(R_i, R_j) |\Psi_i \rangle \langle \Psi_j| + h.c. \]

where \( R_i \) and \( |\Psi_i \rangle \) represent the lattice point and the atomic state at site \( i \), respectively, and \( t(R_i, R_j) \) is the transfer integral between the sites \( i \) and \( j \). We adopt an approximation \( \delta \approx 0.1 \).

\[ t(R_i, R_j) = V_{pp\pi} \left[ 1 - \left( \frac{d \cdot e_z}{d} \right)^2 \right] + V_{pp\sigma} \left( \frac{d \cdot e_z}{d} \right), \]

\[ V_{pp\pi} = V_{pp\pi}^0 \exp \left( -\frac{d - a_0}{\delta} \right), \]

\[ V_{pp\sigma} = V_{pp\sigma}^0 \exp \left( -\frac{d - d_0}{\delta} \right), \]

where \( d = R_i - R_j, \) and \( e_z \) is the unit vector parallel to \( z \) axis. \( V_{pp\pi}^0 \) is the transfer integral between the nearest-neighbor atoms of monolayer graphene which are located at distance \( a_0 = a/\sqrt{3} \approx 0.142 \text{ nm} \), and \( V_{pp\sigma}^0 \) is the interlayer transfer integral between vertically located atoms at the interlayer distance \( d_0 \approx 0.335 \text{ nm} \). Here we take \( V_{pp\pi}^0 \approx -2.7 \text{ eV}, V_{pp\sigma}^0 \approx 0.48 \text{ eV}, \) to fit the low-energy dispersion of bulk graphite. \( \delta \) is the decay length of the transfer integral, and is chosen as 0.184a so that the next nearest intralayer coupling becomes 0.1V_{pp\pi}^{20.32}. 

**FIG. 1:** (Color online) (a) Atomic structure of TBG with rotation angle \( \theta = 21.8° \). Dashed (red) and solid (green) lines represent the lattices of layer 1 and 2, respectively. (b) Brillouin zone of TBG with \( \theta = 21.8° \). Dashed (red) and solid (green) large hexagons correspond to the first Brillouin zone of layer 1 and 2, respectively, and thick small-hexagon to the reduced Brillouin zone of TBG. Open and filled hexagon are two inequivalent valleys \( K \) and \( K' \) of TBG.
The transfer integral for $d > 4a_0$ is exponentially small and can be safely neglected. The band velocity of the Dirac cone in monolayer graphene is given by

$$v \approx \frac{\sqrt{3} V_{pp}}{2} \frac{1}{\hbar}.$$  

(6)

We plot the band structures of four TBGs with the different rotation angles in Fig. 3. The band structure near the Dirac point exhibits a linear dispersion analogous to monolayer’s $7,16–19$ while the interlayer interaction strongly influences the band structure in the higher energy region, giving the van Hove singularity to $M$ point $10,12,20,21$ and a hole-like pocket to $\Gamma$ point. As the rotation angle decreases, the energy span between the saddle point and Dirac point is shortened, and the Fermi velocity decreases accordingly. $8,14,17$ In the smallest rotation angle $\theta = 1.47^\circ$, in particular, the lowest energy band is highly distorted and nearly flat dispersion appears near zero energy. $22,35$ A different choice of parameters slightly modifies the band structure of TBG but does not make qualitative differences.

C. Electronic structures in magnetic fields

We consider TBG in a uniform magnetic field $B = (0, 0, B)$ perpendicular to the layer. For simplicity, we neglect spin Zeeman splitting throughout the paper. The system is characterized by the number of magnetic flux per a unit cell, $\Phi = BS$, measured in units of the flux quantum $\Phi_0 = \hbar/e$. In magnetic field, the Hamiltonian is no longer translationally-symmetric because of the spatial dependence of the vector potential. When $\Phi/\Phi_0$ is a rational number $p/q$ ($p$ and $q$ are coprime integers), however, we can introduce a magnetic unit cell with lattice vectors $\vec{L}_1 = q\vec{L}_1$ and $\vec{L}_2 = L_2$, and construct the eigenstates so as to satisfy the magnetic Bloch condition. $36,37$

By choosing the vector potential as $A = (0, Br, 0)$ and taking $y$ axis parallel to $L_2$, the magnetic Bloch condition for TBG is written as

$$\Psi_k(r + \vec{L}_1) = e^{i k \cdot \vec{L}_1} e^{-i (e/\hbar) (A - B \times r) \cdot \vec{L}_1} \Psi_k(r),$$

$$\Psi_k(r + \vec{L}_2) = e^{i k \cdot \vec{L}_2} \Psi_k(r),$$  

(7)

where $k$ is the Bloch wavenumber defined in the magnetic Brillouin zone spanned by reciprocal vectors of $\vec{L}_1$ and $\vec{L}_2$. Since the magnetic unit cell is $q$ times as large as the unit cell in the absence of magnetic field, the magnetic Brillouin zone is $q$-fold of the original, and each energy band at zero magnetic field splits into $q$ subbands. $20$

The tight-binding Hamiltonian under a magnetic field is obtained by adding a phase factor to the transfer integral in Eq. (3). This is written as

$$H_{TBG} = - \sum_{(i,j)} t(R_i, R_j) e^{i \phi_{ij}} |\Psi_i\rangle \langle \Psi_j| + h.c.,$$

$$\phi_{ij} = -\frac{e}{\hbar} \int_{R_i}^{R_j} A(r) \cdot dr.$$  

(8)

It is, however, not practical to calculate the energy spectrum of TBG by diagonalizing this Hamiltonian, since the number of atoms in a magnetic unit cell is huge in feasible magnetic fields. Instead, we construct the basis from the effective mass wavefunctions for Landau levels of monolayer graphene, which approximate the eigenstates in the absence of the interlayer coupling. We then truncate the bases far from the Dirac point, and compose the Hamiltonian matrix by writing $H_{TBG}$ in terms of the reduced basis.

In monolayer graphene under magnetic field, the eigenstates are labeled by $(v, n, k_y)$ with the valley index
\( v = K, K' \), the Landau level index \( n = 0, \pm 1, \ldots \), and the wave vector \( k_y \) along \( y \) direction. The eigenenergy depends only on \( n \) as

\[
\varepsilon_n = \hbar \omega_B \text{sgn}(n)\sqrt{|n|}, 
\]

with \( \hbar \omega_B = \sqrt{2\hbar e^2/\epsilon B} \). The effective wavefunctions are written as

\[
\begin{align*}
\mathbf{F}_{K\nk_y}(\mathbf{r}) &= \frac{C_n}{\sqrt{L}} e^{ik_y y} \begin{pmatrix} \text{sgn}(n) (-i) \phi_{|n|-1, k_y}(x) \\ \phi_{|n|, k_y}(x) \\ 0 \\ 0 \end{pmatrix}, \\
\mathbf{F}_{K'\nk_y}(\mathbf{r}) &= \frac{C_n}{\sqrt{L}} e^{ik_y y} \begin{pmatrix} \phi_{|n|, k_y}(x) \\ \text{sgn}(n) (-i) \phi_{|n|-1, k}(x) \\ 0 \\ 0 \end{pmatrix}
\end{align*}
\]

where \( \eta^{(l)} \) is the angle of \( a_i^{(l)} \) to \( x \) axis. We define \( \Psi_{vnk_y}^{(l)} \) as the tight-binding wavefunction on the layer \( l \) generated from \( \mathbf{F}_{vnk_y} \).

We then combine the bases of different \( k_y \) so as to satisfy the magnetic Bloch condition, Eq. (7). We define

\[
\Psi_{vnkm}^{(l)} = \sum_{j=-\infty}^{\infty} \alpha^j \exp \left[ i \pi (j + 1) \right] \Psi_{vnk_y(m)}^{(l)},
\]

\[
\alpha = \exp \left[ i (\mathbf{k} - \mathbf{K}^{(l)}) \cdot \left( \mathbf{L}_1 - \frac{1}{2} \mathbf{L}_2 \right) \right],
\]

\[
k_y^{(m)} = k_y - (\mathbf{K}^{(l)}_v)_y - \frac{2\pi}{L_y}(pj + m),
\]

where \( \mathbf{k} \) is the Bloch wavenumber defined in the magnetic Brillouin zone, \( m = 0, 1, \ldots, p-1 \), and \( \mathbf{K}^{(l)}_v \) represent \( \mathbf{K}^{(l)}_v, \mathbf{K}^{(l)}_v \) for \( v = K, K' \), respectively. It is straightforward to show that this satisfies the condition of Eq. (7).

An eigenstate of TBG is written as a linear combination of single-layer eigenstates \( \Psi_{vnk_y}^{(l)} \) belonging to the same \( \mathbf{k} \). We only include single-layer bases within \( -E_{\text{max}} < \varepsilon_n < E_{\text{max}} \), to discard the bases which do not much affect the low-energy spectrum. The eigenenergies are obtained by diagonalizing the Hamiltonian matrix within the reduced bases,

\[
H_{\mathbf{k}}[(l, v, n, m), (l', v', n', m')] \equiv \langle \Psi_{vnk_y}^{(l)} | H_{\text{TBG}} | \Psi_{v'n'k_{y(m')}}^{(l')} \rangle,
\]

for each \( \mathbf{k} \) in the magnetic Brillouin zone. The cut-off energy should be sufficiently larger than the interlayer-coupling energy, which is of the order of \( V_{\text{per}} \) at most, and tends to decrease in small twisting angles. Here we take \( E_{\text{max}} = 1.5 \text{ eV} \) for \( \theta = 9.43^\circ \) and \( 3.89^\circ \), and \( 1.0 \text{ eV} \) for \( 2.65^\circ \) and \( 1.47^\circ \). To avoid undesired effects caused by a discrete change in the number of bases in varying magnetic field, we adopt a soft cut-off which gradually

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**FIG. 3:** Band structure of TBG with rotation angles of (a) 9.43°, (b) 3.89°, (c) 2.65°, and (d) 1.47°, in the absence of magnetic field. Dirac point energy is set to zero.
reduces the matrix elements associated to the single-layer bases beyond $\pm E_{\text{max}}$.

We calculate the matrix elements [Eq. (13)] between different layers by evaluating the transfer integral for each pair of carbon atoms up to the cut-off distance $d = 4a_0$. The matrix elements within the same layer can be replaced with a diagonal matrix composed of the effective-mass eigenenergies in monolayer graphene,

$$H_{k}[(l, v, n, m), (l, v', n', m')] = \varepsilon_n \delta_{v,v'} \delta_{n,n'} \delta_{m,m'}. \quad (15)$$

This treatment is valid in low energies, as long as the magnetic field is not too strong, or $l_B \gg a$.

When the Fermi energy $\varepsilon_F$ is inside a band gap of the spectrum, the Hall conductivity $\sigma_{xy}$ is evaluated by the formula $^{13,34}$

$$\sigma_{xy} = -e \left( \frac{\partial n_F}{\partial B} \right) \varepsilon_F,$$  

where $n_F$ is the electron density per unit area below the gap.

**III. RESULTS AND DISCUSSION**

We show the energy spectrum (left) and quantized Hall conductivity (middle) against the magnetic field amplitude, for $\theta = 9.43^\circ$, $3.89^\circ$ in Fig. 4 and for $2.65^\circ$, $1.47^\circ$ in Fig. 5. In the right-most panel, we show the zero-field band structure in the same energy range. The energy spectrum of $\theta = 9.43^\circ$ [Fig. 4(a)] is almost equivalent to monolayer’s Landau level, suggesting that two layers are nearly decoupled in this energy region.

The sequence of the Hall conductivity, 4, 12, 20, · · · in units of $-e^2/h$ is exactly twice as large as the monolayer’s $^{12,30,31}$ Each Landau level is eight-fold degenerate due to the number of layers as well as the spin and valley degeneracies.

In contrast, the energy spectrum of $\theta = 3.89^\circ$ [Figs. 4(d) and 4(e)] exhibits a complicated structure which is clearly distinguished from monolayer graphene. In weak magnetic fields of $\Phi/\Phi_0 < 0.1$, the low-energy spectrum below 0.2 eV shows monolayer-like Landau levels and Hall conductivity of 4, 12, 20, · · ·. In the higher energy region above 0.2 eV, on the other hand, we observe hole-like Landau levels moving downward in energy, and the negative Hall conductivity of 0, −4, −8, −12, · · ·. When the electron density increases from the charge neutrality point, the Hall conductivity rises in a sequence of 4, 12, 20, · · · with a step of 8, then abruptly drops to a negative extremum, and increases with a step of 4 all the way to zero.

Those spectral features in weak magnetic field perfectly coincide with the zero-field band structure in Fig. 4(f). The electron-like Landau levels are regarded as the quantized orbits accommodated in electron pockets at $K$ and $K'$ points, while the hole-like Landau levels are those in a hole pocket at $\Gamma$ point. The transition from electron-like levels to hole-like levels corresponds to topological change of the Fermi surface at the saddle point ($M$ point), which is responsible for the van Hove singularity at 0.2 eV. The step of the Hall conductivity reflects the number of electron and hole pockets in the first Brillouin zone, i.e., the degeneracy of an electron-like level is twice as large as that of a hole-like level, because there are inequivalent $K$ and $K'$ points whereas there is only one $\Gamma$ point. Note that the lowest conduction band is composed of a pair of nearly degenerate energy bands as shown in Fig. 4(f), and this doubling contributes to the degeneracy of two in addition to the spin degeneracy. These two bands are not completely degenerate but mirror-symmetric with respect to the lines of $K - \Gamma$, $K' - \Gamma$ and $K - K'$, and thus give the identical Landau level energies. The corresponding states of the two bands are related by $C_2$ symmetry in the real-space lattice structure with respect to the sides of the unit cell in Fig. 2.

The electron-like and hole-like Landau levels are alternatively explained by a nearly-free electron model, without mentioning the rigorous zero-field band structure. In Fig. 6 we illustrate semi-classical electron trajectories at several different Fermi energies for a “free” TBG with interlayer coupling neglected. In the limit of a small Fermi energy [Fig. 6(a)], electrons move along closed orbits around $K$ and $K'$, and those motions are quantized into monolayer-like Landau levels. Since each of $K$ and $K'$ points include two original $K$ points from top and bottom monolayers, the Hall conductivity yields 4, 12, 20, · · ·, i.e., double of monolayer’s sequence. For large Fermi energies, the electron orbits around the $K$ and $K'$ valleys cross each other as shown in Fig. 6(c). A finite interlayer coupling interchanges the orbits at each crossing point, and generates a single hole-like trajectory moving around $\Gamma$ point in the opposite direction. The corresponding hole-like Landau levels are four-fold degenerate due to spin and the Fermi circle doubling, and thus the Hall conductivity takes 0, −4, −8, −12, · · ·. The middle panel [Fig. 6(b)] is for the intermediate energy region between two regimes. There, the different semiclassical orbits are strongly mixed by the magnetic breakdown due to a small $k$-space separation, resulting in broadening of Landau levels near the van Hove singularity in Fig. 4(d).

The electron density to fill the lowest conduction band is given by

$$n_0 = \frac{2g_s}{S}, \quad (17)$$

where $g_s$ is the spin degeneracy and 2 is the band doubling. $n_0$ characterizes the order of the electron density required to reach the van Hove singularity and the hole-like Landau levels. We have $n_0 = 3.5, 1.6$ and 0.5 in units of $10^{13}$ cm$^{-2}$ for $\theta = 3.89^\circ, 2.65^\circ$ and $1.47^\circ$, respectively. The folded band structure and the corresponding Landau level spectrum of TBG are quite analogous to the whole $\pi$ band in monolayer graphene, while the electron density to access the hole-like levels is of the order of $10^{15}$ cm$^{-2}$. 


FIG. 4: (Color online) Energy spectrum and quantum Hall effect in TBG in magnetic field with rotation angles of 9.43° (above) and 3.89° (below). In each row, the left and middle panels display the energy spectrum and the quantized Hall conductivity as functions of magnetic field strength, respectively, and the right panel shows the band structure in the absence of magnetic field. In (b) and (e), the quantized values of Hall conductivity inside energy gaps are indicated by numbers as well as colors filling the gaps. The Hall conductivity of the gray area cannot be determined by the present calculation.

When the magnetic field amplitude becomes as large as $\Phi/\Phi_0 \sim 1$, the uncertainty in electron momentum is comparable to the size of the Brillouin zone, and the semiclassical picture breaks down. There the energy spectrum exhibits a fractal band structure.\textsuperscript{20,21} In Fig. 1 we observe that the electron and hole Landau levels gradually evolve into the fractal structure as $\Phi/\Phi_0$ approaches 0.5, and the Hall conductivity in the fractal regime behaves non-monotonically as a function of Fermi energy.\textsuperscript{23,26}

The energy spectra of TBG with $\theta = 2.65^\circ$ and $1.47^\circ$ exhibit similar fractal structures to that of $\theta = 3.89^\circ$, as expected from the resemblance between the band structures in the absence of magnetic field. The field amplitude corresponding to the same $\Phi/\Phi_0$ is lower for smaller $\theta$, so the actual magnetic field strength needed to observe a fractal band structure becomes more feasible in smaller rotation angle. The fractal feature becomes conspicuous roughly when $\Phi/\Phi_0 \sim 0.3$, which corresponds to 100 T, 50 T, and 15 T in $\theta = 3.89^\circ, 2.65^\circ$, and $1.47^\circ$, respectively.
FIG. 5: (Color online) Plots similar to Fig. 4 for TBG with rotation angles of 2.65° (above) and 1.47° (below).

In the case of θ = 1.47°, the spectrum is strongly compressed in the vicinity of Dirac points, in accordance with the band width reduction which occurs in small rotation angles. Although the band structure near Dirac points is almost flat, Γ point still has a finite band velocity. As a consequence, the energy gaps between the hole-like Landau levels are much wider than those between the electron-like levels.

FIG. 6: Fermi circle and electron trajectories of TBG in a nearly free electron picture, for three different Fermi energies (a) in the vicinity of Dirac points, (b) near van Hove singularity at the saddle point, and (c) hole-like band at Γ point.
IV. CONCLUSION

We investigated the electronic structure and the quantum Hall effect in TBG with various rotation angles in the presence of magnetic field. We calculated the energy spectrum and quantized Hall conductivity in a wide magnetic-field range, and described the evolution from the semi-classical Landau levels to the fractal band structure. In weak magnetic field, those semiclassical levels gradually evolve into Hofstadter’s butterfly, where the Hall conductivity exhibits a non-monotonic behavior as a function of Fermi energy. The typical electron density and magnetic field amplitude characterizing the spectrum monotonically decrease as the rotation angle is reduced, indicating that the rich electronic properties may be observed in a moderate condition for TBG with small angle less than 5°.

ACKNOWLEDGMENTS

This work is supported by JST-EPSRC Japan-UK Cooperative Programme Grant No. EP/H025804/1. P. M. acknowledges the support from Grant-in Aid for Research Activity Start-up (23840004), and appreciate the support from Korea Institute of Science and Technology Information Supercomputing Center through the strategic support program for the supercomputing application research (Grant No. KSC-2009-S02-0009), and the Supercomputer Center, Institute for Solid State Physics, University of Tokyo for the use of the facilities (project ID: H23-D-0009).

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