Effects of site asymmetry and valley mixing on Hofstadter-type spectra of bilayer graphene in a square-scatter array potential

Danhong Huang¹,²,⁶, Andrii Iurov¹, Godfrey Gumbs⁴,⁵ and Liubov Zhemchuzhna³

¹ Air Force Research Laboratory, Space Vehicles Directorate, Kirtland Air Force Base, NM 87117, United States of America
² Department of Electrical & Computer Engineering, University of New Mexico, Albuquerque, NM 87131, United States of America
³ Center for High Technology Materials, University of New Mexico, Albuquerque, NM 87106, United States of America
⁴ Department of Physics and Astronomy, Hunter College of the City University of New York, 695 Park Avenue, New York, NY 10065, United States of America
⁵ Donostia International Physics Center (DIPC), P de Manuel Lardizabal, 4, 20018, San Sebastian, Basque Country, Spain

E-mail: danhong.huang@us.af.mil

Received 15 October 2018, revised 19 December 2018
Accepted for publication 9 January 2019
Published 30 January 2019

Abstract
Under a magnetic field perpendicular to a monolayer graphene, the existence of a two-dimensional periodic scatter array can not only mix Landau levels of the same valley for displaying split electron–hole Hofstadter-type energy spectra, but also couple two sets of Landau subbands from different valleys in a bilayer graphene. Such a valley mixing effect with a strong scattering strength has been found observable and studied thoroughly in this paper by using a Bloch-wave expansion approach and a projected $2 \times 2$ effective Hamiltonian including interlayer effective mass, interlayer coupling and asymmetrical on-site energies due to a vertically-applied electric field. For bilayer graphene, we find two important characteristics, i.e. mixing and interference of intervalley scatterings in the presence of a scatter array, as well as a perpendicular-field induced site-energy asymmetry which deforms severely or even destroys completely the Hofstadter-type band structures due to the dependence of Bloch-wave expansion coefficients on the applied electric field.

Keywords: bilayer graphene, valley mixing, Hofstadter, site energy

(Some figures may appear in colour only in the online journal)
A comprehensive theoretical study of the LL degeneracy and quantum Hall effect for bilayer graphene in Bernal stacking was reported in [15]. Based on an effective 2D Hamiltonian, it was concluded that the low-energy spectrum of bilayer graphene can be characterized as parabolic dispersion of chiral quasi-particles with a Barry phase $2\pi$. Meanwhile, its magnetic-field dependent energy spectrum is found consisting of a set of nearly equidistant four-fold degenerate LLs. In this paper, we will employ such an effective-Hamiltonian approach to establish theoretical formalism for modulated LLs in the presence of a square-scatter array potential in section 2.

One of the most unusual and fascinating phenomena related to the electronic spectrum under a perpendicular quantizing magnetic field is the so-called Hofstadter butterfly [16, 17], theoretically predicted in 1976. Here, a recursive fractal electron spectrum was obtained as a function of prime ratio of the magnetic flux passing through a lattice unit cell, and these degenerate electronic subbands split and clustered themselves into different patterns corresponding to the value of a given magnetic-flux ratio. By performing first-principles calculations for hexagonal 2D graphene-type lattice, tight-binding approximation resulted in a fractal electron spectrum was obtained as a function of prime ratio of the magnetic flux passing through a lattice unit cell through which the magnetic flux was measured [16]. Inside a van der Waals heterostructure sample [24], the main idea involved in such experiments is that an elementary lattice-unit cell through which the magnetic flux was measured [16] will be replaced by a much bigger supercell of the Moire lattice [25–27], so that the butterfly is to be seen at a much lower magnetic field. Additionally, the theory for such butterfly structures in twisted bilayer graphene was proposed in [28], in which long-period spatial patterns can be created precisely at small twist angles. Later, the coexistence of both fractional-quantum-Hall and integral-quantum-Hall states associated with fractal Hofstadter spectrum was confirmed experimentally within such twisted-bilayer structures [29]. Moreover, specific subband gaps of a Hofstadter’s butterfly were also found for interacting Dirac fermions in graphene [30].

On the other hand, in the absence of a magnetic field, a periodic electrostatic field gives rise to new zero-energy states with minigaps and chirality [31], and their composite wave functions can still satisfy the required Bloch periodic condition. Apart from this, new massless Dirac fermions with strong anisotropic properties [32] are realized in graphene subjected to a slowly-varying periodic potential [33]. In contrast, a spatially-uniform interaction of Dirac electron with an off-resonant optical field can lead to the formation of either gapped [34–36] or anisotropic dressed [37] states depending on polarizations of an imposed irradiation.

Very interestingly, two unique features associated with bilayer graphene systems have been found. The first property is the interval valley mixing and the quantum interference effect coming from two valleys in the presence of a 2D scattering-lattice potential, while the second property results from a site-energy asymmetry induced by a perpendicular electric field. Here, the latter factor is able to destroy the Hofstadter-type fractal band structures established by an in-plane scattering-lattice potential and an out-of-plane quantizing magnetic field, resulting in strongly deformed self-repeated patterns. Such a phenomena is attributed to the dependence of Bloch-wave expansion coefficients on an applied electric field, leading to an electro-modulation of the Hofstadter-type subband splittings.

The rest of the paper is organized as follows. In section 2 we present theoretical formalism and acquire a set of characteristic equations, describing electron energy spectrum and corresponding eigenstates for bilayer graphene in the presence of both a perpendicular quantizing magnetic field and a 2D periodic electrostatic modulation potential. These results expand the previously studies for a 2D electron gas (2DEG) [38] and for a monolayer graphene [39, 40]. In section 3, we display and discuss our numerical results demonstrating fractal Hofstadter band-structures in different ranges of magnetic field of interest and with various modulation strengths in a close up view for separate LLs and self-repeated superstructures as well. Finally, a brief summary with remarks is given in section 4.

### 2. Model and theory

By considering $K$ and $\tilde{K}$ valleys, where $K = (\frac{2\pi}{a}, \frac{2\pi}{\sqrt{3}a}, 0)$, $\tilde{K} = (-\frac{2\pi}{2a}, \frac{2\pi}{\sqrt{3}a}, 0)$ and $a \approx 2.46\,\text{Å}$, and including sublattices $A$ and $B$ as well as bilayer structure, the four-component wave functions for each valley can be formally written as [15]

$$\Psi_K = \begin{pmatrix} \phi_A^K \\ \phi_B^K \\ \phi_A^{\tilde{K}} \\ \phi_B^{\tilde{K}} \end{pmatrix}, \quad \Psi_{\tilde{K}} = \begin{pmatrix} \phi_A^{\tilde{K}} \\ \phi_B^{\tilde{K}} \\ \phi_A^K \\ \phi_B^K \end{pmatrix},$$

where $A$ and $B$ label the bonds in the bottom layer and $\tilde{A}$ and $\tilde{B}$ label the bonds in the top layer. For each valley, the $4 \times 4$ graphite tight-binding Hamiltonian matrix within the $xy$-plane for Bernal-stacking [12] bilayer takes the form

$$\hat{H}_{TB}^{xy} = v_F \begin{bmatrix} V + \xi \nu/2 & \xi \nu(\tilde{p}_x + \tilde{p}_y) & 0 & \xi \nu(\tilde{p}_x - \tilde{p}_y) \\ \xi \nu(\tilde{p}_x - \tilde{p}_y) & V - \xi \nu/2 & \xi \nu(\tilde{p}_x + \tilde{p}_y) & 0 \\ 0 & \xi \nu(\tilde{p}_x - \tilde{p}_y) & V - \xi \nu/2 & \gamma_1 \\ \xi \nu(\tilde{p}_x + \tilde{p}_y) & 0 & \gamma_1 & V + \xi \nu/2 \end{bmatrix},$$

where $\xi = \pm$ represents the $K$ $(+)$ or $\tilde{K}$ $(-)$ valley, $v_F = \sqrt{3}a \gamma_0 \equiv v_F \approx 3 \times 10^6 \,\text{cm} \, s^{-1}$ is the intralayer Fermi velocity, $\gamma_1 = 2m^*c^2 < \gamma_0$ characterizes the effective mass of electrons in the parabolic band, $v_T = \sqrt{3}a \gamma_1 < v_F$ measures the strength of the interlayer coupling, $m^* \approx \frac{m}{2}$ represents the bias-induced on-site energies of bilayer, $u = e\mathcal{E}_D D$ with electric field $\mathcal{E}_D$ and bilayer separation $D$, and $u = 0$ corresponds to a symmetrical bilayer. In addition, we have introduced canonical momentum operators $\hat{p}_x \equiv -i\hbar \frac{\partial}{\partial x} + e\mathcal{E}_D y$ and $\hat{p}_y \equiv -i\hbar \frac{\partial}{\partial y}$, where the Landau...
gauge \( \mathbf{A} = (-B_0 y, 0, 0) \) is chosen for a uniform magnetic field \( \mathbf{B}_0 \) along the vertical z direction. The potential of a 2D scatter array in equation (2) is assumed as

\[
V \equiv V(x, y) = V_0 \left[ \cos \left( \frac{\pi x}{d_x} \right) \cos \left( \frac{\pi y}{d_y} \right) \right]^{2N}.
\]  
(3)

where \( N \) is an integer, \( V_0 \) stands for the scattering-potential strength, \( d_x \) and \( d_y \) are the two array periods in the x and y directions, respectively.

Even in the absence of the scatter potential (i.e. \( V_0 = 0 \)), the eigen-energies and eigen-states correspond to the Hamiltonian in equation (2) can only be calculated numerically. For low-energy states of electrons (with kinetic energy less than \( \gamma_1/4 \)), however, the \( 4 \times 4 \) Hamiltonian in equation (2) can be projected onto a \( 2 \times 2 \) one. For such a situation, the wave functions in equation (1) for each valley also reduce to a two-component form

\[
\Psi^K = \begin{bmatrix} \phi^K_x \\ \phi^K_y \end{bmatrix}, \quad \tilde{\Psi}^K = \begin{bmatrix} \phi^B_x \\ \phi^B_y \end{bmatrix},
\]

and the projected \( 2 \times 2 \) effective Hamiltonian matrix becomes

\[
\tilde{\mathcal{H}}^\xi_{eff} = -\frac{1}{2m} \left[ \begin{array}{cc} 0 & (\hat{p}_x - i\hat{p}_y)^2 \\ (\hat{p}_x + i\hat{p}_y)^2 & 0 \end{array} \right] + \xi \xi_0 \left[ \begin{array}{cc} 0 & \hat{p}_x + i\hat{p}_y \\ \hat{p}_x - i\hat{p}_y & 0 \end{array} \right] + \frac{\xi u}{2} \left[ \begin{array}{cc} 1 & 0 \\ 0 & 1 \end{array} \right] \\
- \frac{\xi \omega_c^2}{\gamma_1} \left[ \begin{array}{cc} (\hat{p}_x - i\hat{p}_y)(\hat{p}_x + i\hat{p}_y) & 0 \\ 0 & -(\hat{p}_x + i\hat{p}_y)(\hat{p}_x - i\hat{p}_y) \end{array} \right] + V(x, y) \hat{h},
\]

where \( \hat{h}_0 \) is the number term in the \( 2 \times 2 \) identity matrix, the first, second and the rest two terms represents the intralayer, interlayer and bias effects, respectively.

Our model for the effective 2D Hamiltonian in the presence of a strong (or intermediate) magnetic field is based on the corresponding derivations in [15]. All the Hamiltonian parameters, as well as the corresponding derivations in [15], are measured in terms of the cyclotron energy \( \hbar \omega_c \). \( \omega_c = eB_0/m^* \) is proportional to the applied magnetic field \( B_0 \). The physical picture behind estimating each Hamiltonian coefficient is built on the two possible types of interlayer hopping between the equivalent sites \( \hat{A} \) and \( \hat{B} \) (\( \hat{A} \) and \( \hat{B} \)), which connect to atomic bonds in the top (bottom) layer, respectively. In the Bernal (\( A - B \)) stacking, all the other coupling types could be effectively neglected since they represent insignificant tunneling. In addition, interlayer (\( A - B \)) coupling appears to be much weaker compared to the intralayer dimer-state hopping. Two more Hamiltonian terms (\( \propto \xi_0 u \)) are included due to the site asymmetry between two graphene layers. The amplitude of electrostatic modulation \( V_0 \) is chosen to display comparable effect in comparison with the applied magnetic field on the electron dynamics.

By taking \( V = 0 \) in equation (5) as a start, in the strong-field limit, i.e. \( m^* v_3^2 \ll \hbar \omega_c < m^* v_3^2 \) with a cyclotron frequency \( \omega_c = eB_0/m^* \), we can formally set \( v_3 \to 0 \) in equation (5).

Based on this simplification, we obtain the analytical form of the eigen-energy levels for each valley (\( \xi = \pm \))

\[
E_{\xi = \pm} = \left\{ \begin{array}{ll} \pm \hbar \omega_c \sqrt{n(n-1)} - \xi \delta/2, & \text{for } n \geq 2, \\
\xi u/2 - \xi \delta, & \text{for } n = 1, \\
\xi u/2, & \text{for } n = 0. \end{array} \right.
\]

where \( \delta = u \hbar \omega_c / \gamma_l \). \( E_{\xi = \pm} \) and \( E_{\xi = \mp} \) correspond to electron and hole energy levels at each valley, respectively, each energy level is spin degenerate, and the lowest two energy levels are four-fold degenerate with respect to both spins and electron–hole pseudospins. If \( u = 0 \), we get \( E_{\xi = \pm,0} = E_{\xi = \pm,1} = 0 \) from equation (6), which becomes eight-fold degenerate now. The corresponding eigen-states to these electron (hole) energy levels (\( n \geq 2 \)) are calculated as

\[
\Psi_{\xi = \pm,n,k_x}(x, y) = C_n^\pm(\xi) e^{i(k_x+k_y)x} \sqrt{\frac{L_y}{D_n^\pm(\xi)}} \left[ \frac{\phi_{n,k_x+k_y}(y)}{\sqrt{n(n-1)}} \right] \left[ \frac{\phi_{n-2,k_x+k_y}(y)}{\sqrt{n(n-1)}} \right],
\]

where \( L_y \to \infty \) is the sample length in the x direction, \( \xi = K_x = K_y = \tilde{K} \) for \( \xi = + \). \( \phi_{n,k}(y) \) is the harmonic-oscillator wave functions with a guiding center \( y_0 = 0 \), \( \xi = K = \tilde{K} \) is the magnetic length, and two coefficients

\[
D_n^\pm(\xi) = \frac{E_{\xi = \pm}^n - \xi u/2 + \xi n \delta}{\hbar \omega_c \sqrt{n(n-1)}}, \quad C_n^\pm(\xi) = \frac{1}{\sqrt{1 + \left| D_n^\pm(\xi) \right|^2}}.
\]

Assuming \( u = 0 \), we have \( D_n^\pm(\xi) = \pm 1 \) and \( C_n^\pm(\xi) = 1/\sqrt{2} \) for \( n \geq 2 \), which becomes independent of \( \xi \) and \( n \). On the other hand, for \( n = 0 \) and \( n = 1 \) we obtain

\[
\Psi_{\xi = \pm,0,k_x}(x, y) = \frac{e^{i(k_x+k_y)x}}{\sqrt{L_y}} \left[ \frac{\phi_{0,k_x+k_y}(y)}{0} \right] \left[ \frac{\phi_{0,k_x+k_y}(y)}{0} \right],
\]

After the scatter array has been included in the strong-field limit, the wave function of the system can be expanded as

\[
\Phi_{\xi = \alpha,n,k_y}(x, y) = \frac{1}{\sqrt{N_y}} \sum_{\gamma = -\infty}^{\infty} e^{i\gamma(k_x+k_y)x} \psi_{\xi = \alpha,n,k_x}(y, x),
\]

where \( \xi = \pm \), \( \alpha = \pm \) corresponds to electron and hole states, \( k_x = (k_x, k_y), \) \( \gamma = \pm \) \( n/d_x \), \( K_1/2 \), and \( k_y = \pm \pi/4d_y \) for the first magnetic Brillouin zone, \( N_y = L_y/(4d_y) \) is the number of unit cells spanned by \( b_1 = (d_x, 0) \) and \( b_2 = (0, 4d_y) \) in the y direction, \( L_y \to \infty \) is the sample length in the y direction, \( K_1 = 2\pi/d_x \) is the reciprocal lattice vector in the x direction, and \( \ell = 1, 2, \cdots, p \) is a new quantum number for labeling split p subbands from a \( k_y \)-degenerated LL in the absence of scatters. Importantly, the above constructed wave function satisfies the usual Bloch condition, i.e.

\[
\Phi_{\xi = \alpha,n,k_y}(x + d_x, y + qd_y) = e^{i\gamma d_x} e^{i\gamma d_y} \Phi_{\xi = \alpha,n,k_y}(x, y) \Phi_{\xi = \alpha,n,k_y}(x, y). \]

(11)

Substituting the expression for wave function at each valley in equations (7) into (9), we find
where \( n \neq \ell \).

Now, by taking into account of the \( V(x,y) \) term in equation (5), a tedious but straightforward calculation leads to an explicit expression for the matrix elements of the potential \( V(x,y) \), yielding

\[
\Phi_{\ell,0,n,k}^{\xi}(x,y) = \frac{1}{\sqrt{N_l L_s}} \sum_{s=-\infty}^{\infty} \left\{ e^{\imath (p+\ell) \xi} C_n^0(\xi) \right\},
\]

\[
\times e^{\imath (k_x+K_{\ell})(-\ell+\xi)} \left\{ \phi_{n,K_{\ell}}(x,y) \right\} + \phi_{n,K_{\ell}}(x,y) \right\},
\]

where \( D_{n}^{\ell}(\xi) = D_{n}^{\ell}(\xi) = 0 \) and \( C_n^{\ell}(\xi) = C_n^{\ell}(\xi) = 1 \).

The energy dispersion \( \epsilon_{\nu}(k_{||},\xi) \) of the \( \nu \)th magnetic band around each valley for this modulated system is a solution of the eigenvector problem \( \hat{M}(k_{||},\xi) \cdot \mathbf{A}(k_{||},\xi) = 0 \) with elements of the coefficient matrix \( \hat{M}(k_{||},\xi) \) given by

\[
\{ \hat{M}(k_{||},\xi) \}_{\nu',\nu} = \left[ E_{n,n}^{\nu',\nu} - \epsilon(k_{||},\xi) \right] \delta_{\nu',\nu} \delta_{\ell,\ell'} \delta_{\alpha,\alpha'} + V_{\ell',\alpha'}^{\nu'}(k_{||},\xi),
\]

where \( \delta_{\nu',\nu} = 1 \) for \( n = 0, 1 \) (i.e., degenerate electron-hole levels) and \( \delta_{\ell,\ell'} = \delta_{\alpha,\alpha'} \) for \( n \geq 2 \).

\subsection{3.1 2DEG and monolayer graphene}

As a starting point, we first briefly discuss the effect of a 2D periodically-modulated scattering-lattice potential in equation (3) on a 2DEG under a perpendicular quantizing magnetic field \( B_0 \). In the absence of this scattering-lattice potential, 2DEG will be quantized into a series of discrete LLs: \( \varepsilon_n^{(0)} = (n + 1/2) h\omega_c \) with \( n = 0, 1, 2, \ldots, \omega_c = eB_0/m^* \) as the cyclotron frequency, and \( m^* \) as the effective mass of electrons. These uncoupled LLs are highly degenerate with respect to their guiding centers \( \nu_0 = k_x \theta_B \) (with different cyclotron orbits), where \( \theta_B = \sqrt{\hbar/eB_0} \) is the magnetic length. In the presence of the scattering-lattice potential, however, these

\[
m_{n,n} = \frac{m!}{n!(m-n)!}.
\]

Finally, we have introduced in equation (14) the following three self-defined functions

\[
A_1^{(BA)}(r, s|\xi', \xi) = D_{n,n}^{(BA)} T_{\ell,0,\nu'}^{(\xi', \xi)} \delta_{\ell,\ell'},
\]

\[
A_2^{(BA)}(r, s|\xi', \xi) = D_{n,n}^{(BA)} \left\{ \delta_{\ell,\ell'} \left[ \epsilon_{n,n}(\xi') - \epsilon_{n,n}(\xi) \right] \right\},
\]

\[
A_3^{(BA)}(r, s|\xi', \xi) = D_{n,n}^{(BA)} \left\{ \delta_{\ell,\ell'} \left[ \epsilon_{n,n}(\xi') - \epsilon_{n,n}(\xi) \right] \cos(\Theta_{n,n}^{(\xi', \xi)} \xi') \right\} + \delta_{\ell-\ell'} \left[ \epsilon_{n,n}(\xi') - \epsilon_{n,n}(\xi) \right] \cos(\Theta_{n,n}^{(\xi', \xi)} \xi'),
\]

where \( \beta = |n - n'| \).
Figure 1. Distributions of magnetically-quantized energy levels $\varepsilon_n(k_x)$ of a 2DEG as a function of magnetic flux $\Phi/\Phi_0 = p/q$ under a 2D scattering-lattice potential given by equation (3) with parameters $V_0/\hbar\omega_c = 1$, $N = 3$, $d_s = d_h$, $\omega_c = eB_0/m^*$, and $m^*$ as the effective mass of electrons. Here, we have chosen $k_x = k_y = 0.3 K_1$. Panel (a) displays the distributions of the lowest four bands, and panel (b) shows the close-up view of the self-similar pattern of the $n = 3$ band at lower $B_0$.

degenerate LLs are strongly coupled to each other and expand into a set of split Landau bands, as shown in figure 1(a). Furthermore, a close-up view in figure 1(b) reveals that a self-similar pattern occurs within the fourth ($n = 3$) Landau band at low $B_0$, just as predicted early by Hofstadter in his seminal work [16].

The density-of-states (DoS) for a given valley can be calculated from

$$\text{DoS}(E|B_0) \approx \sum_\nu \int_{-\pi/d_v}^{\pi/d_v} dk_v \int_{-\pi/(qd_v)}^{\pi/(qd_v)} dk_y \delta \left\{ \varepsilon_\nu(k_x) - E \right\},$$

where the summation is performed over all magnetic bands $\nu$ corresponding to each accessible LL $0, \pm 1, \ldots$. We know, on the other hand, that the DoS is directly related to the subband locations and dispersions. For example, scanning-tunneling spectroscopy, which has been employed to investigate the DoS of newly synthesized germanene, demonstrated a perfectly linear-shaped DoS, corresponding to the Dirac cone dispersions of a free-standing layer. The only visible difference in this paper stems from the fact that the DoS basically reproduces the Hofstadter fractal structure, after having averaged over all allowed values of the wave vector $k_y$. Consequently, experimentally obtained self-repeated structures from the density-of-states measurements become the only required evidence for verification of the corresponding behavior of the low-energy band structure.

If the 2DEG is replaced by a monolayer graphene, a different set of LLs $\varepsilon_n^{(0)} = \pm \frac{\hbar\omega_c}{2\sqrt{n}}$ with $n = 0, \pm 1, \pm 2, \cdots$ appears in the absence of a scattering-lattice potential, where $\omega_c = \sqrt{2}v_F/L_p$, $v_F$ is the Fermi velocity of graphene, and $+$ ($-$) corresponds to electrons (holes), respectively. In this case, we find that the $n = 0$ LL sits at the zero-energy Dirac point instead of $\hbar\omega_c/2$ for 2DEG, and $\varepsilon_n^{(0)} \propto \sqrt{|n|B_0}$ but not proportional to $(n + 1/2)B_0$ for 2DEG. After the scattering-lattice potential in equation (3) has been employed, these guiding-center degenerated energy levels also expand into a Landau band through mutual couplings, as seen in figure 2(a). However, the mirror symmetry with respect to the band center is lost in figure 2(b) for monolayer graphene, as discussed in details recently by us [40]. Here, we prepared the second difference between 2DEG and monolayer graphene is the LL separation $(\sqrt{n + 1} - \sqrt{n})\hbar\omega_c$ for graphene, in contrast with a uniform one, $\hbar\omega_c$, for 2DEG. Consequently, the graphene energy-level separation will decrease with increasing $n$, and therefore, overlaps of many Hofstadter butterflies will show up for higher $n$ values as in figure 2(a).

In figure 2, $p/q$ represents the ratio of magnetic flux $\Phi = B_0d_1d_s$ through an elemental 'supercell' to the magnetic-flux quantum $\Phi_0 = 2.068 \times 10^{-15}$ Wb. Therefore, $p/q = 0.1$ in our study corresponds to a magnetic filed $B_0 = 34.17$ T, where $d_s = d_h = 10 \text{ Å}$ is assumed and $a = 2.46 \text{ Å}$ is the carbon–carbon interatomic distance in bilayer graphene. Also, we would like to emphasize that the minimal ratio $p/q = 0.01$ taken in this paper, for which the Hofstadter structure can still be resolved, gives rise to a technologically-accessible field strength $B_0 \approx 3 \text{ T}$. On the other hand, the calculated electron energies in this study are on the level of $\hbar\omega_c$, where $\omega_c = eB_0/m^* = 9.77 \times 10^{13}$ Hz is the cyclotron frequency of electrons, and $m^* = 0.054m_e = 4.9 \times 10^{-32}$ kg is the effective mass of electrons. This gives rise to $\hbar\omega_c = 6.42$ meV.

3.2. Bilayer graphene

Now, let us turn our attention to discussions on the development of Landau bands in a bilayer graphene. For bilayer graphene subjected to a scattering-lattice potential given by equation (3) and under a perpendicular quantizing magnetic...
field $B_0$ at the same time, our numerical solutions for the eigenvalue equation in equation (22) are presented in figures 3–5 with various scattering strengths $V_0$. As a whole, we find that degenerate LLs with different guiding centers tend to couple to each other and lead to band-center asymmetric Landau bands within which a fractal Hofstadter structure is seen for high magnetic fields $B_0$. Furthermore, the developed Landau bands for two valleys ($\xi = \pm$) are coupled to each other in a bilayer graphene through an Umklapp scattering process across whole magnetic Brillouin zones, which is in contrast with the case for a monolayer graphene where the Landau bands are found independent of a valley.

As indicated in section 2, the valley mixing and interference effect contained in the modulation potential $V_{\alpha}(\xi)\delta$ and $\xi$ in equation (13) are described explicitly by the wave number $k_0(\xi',\xi) = k_0 + (K_x - K_y)$, where $K_y = -K_\xi = 20.94 d_{c}^{-1}$, and $d_{c} = d_{f} = 10a = 2.46$ nm. The integer power $N$, which measures the peak sharpness of the scattering potential in equation (3), is selected as $N = 3$. In the absence of the 2D scattering-lattice potential, each LL under the magnetic flux ratio $\Phi/\Phi_0 = p/q$ has a $p$-fold degeneracy for magnetic subbands. We have taken $p = 13, 17$ and 11, respectively, in figures 3–5. For all three graphs, we only show the lowest four Landau bands for both electrons and holes. All the numerical results which display self-repeated Hofstadter butterfly structures are presented as a function of $\Phi/\Phi_0 = p/q$. Here, all energy levels, except for $n=0$ and $n=1$, are shifted upwards by a fixed energy offset $4^{-N} [(2N)!/N!]^2 V_0 = 0.146 V_0$ for $N = 3$. Therefore, we had to make an adjustment to our plots in figures 3–5 so that the electron–hole symmetry could be restored with respect to the zero-energy point. With fixed lattice period $d_{c} = d_{f}$, a magnetic-flux ratio $\Phi/\Phi_0 = p/q$ can be uniquely related to a magnetic-field strength $B_0$. The upper bound of $p/q$ in figures 3–5 for observing Hofstadter spectra is found within the range of $B_0 = 5–10$ T.

The unperturbed LL spectrum is shown in equation (6). In our numerical calculations, we have set $\delta/h\omega_c \approx 0.001$ and $u/h\omega_c \approx 0.003$ so that the LL structure consists of a few pairs of extremely closely-located levels, corresponding to $\xi, \xi' = \pm$ for two valley indexes. This on-site energy-level separation ($\sim 10^{-3} h\omega_c$) depends on $B_0$ or $p/q$. Additionally, two groups of LLs associated with $n = 0$ and $n = \pm 1$ are nearly degenerate due to their very small separations $\delta$, as found from the inset of figure 3(a). Furthermore, the spin degeneracy in these LLs is kept since none of them depends on spin index. All $h\omega_c$-scaled higher levels staring for $n \geq 2$ have the same $\alpha = \sqrt{n(n-1)}$ dependence which becomes nearly equidistant as $n \gg 1$ and in contrast with the monolayer graphene. Here, the pseudospin index $\alpha = \pm 1$ hints a complete electron/hole symmetry for these $n \geq 2$ LLs. After the scattering-lattice potential given by equation (3) has been introduced to bilayer graphene, the previously uncoupled and highly-degenerate LLs expand into many magnetic bands with self-similar structures, as can be verified directly from figure 3(b). Since the higher LLs become almost equally separated in bilayer graphene, we expect similar self-repeated structures within a magnetic band for large $n$ values.

Because the mixing of LLs depends on $V_0$, we present comparisons in figures 4 and 5 for strong and intermediate scattering strengths $V_0/h\omega_c$. When the strong scattering strength is $V_0/h\omega_c = 10$, the mixing of $n = 2$ and $n = 3$ Landau bands is severe, as seen in figure 4(a). In addition, the band mixing is found to increase with magnetic field $B_0$ in this case. If the scattering strength, $V_0/h\omega_c = 1$, is weak, on the other hand, no band mixing appears, as can be verified from figure 4(b).

For intermediate scattering strength $V_0/h\omega_c = 2.5$ in figure 5(a), we find the band mixing still happen, but it occurs...
a repeated butterfly in the Hall conductivity map. The Hall conductivity associated with each miniband in the fractal spectrum is quantized according to the parameters of a simple Diophantine relation [21]. In all these relatively recent experimental studies mentioned above, graphene flakes and bilayers were used to couple with a boron-nitride substrate, which is another strong motivation for our theoretical investigation of bilayer graphene. Our results demonstrate the existence of Hofstadter self-similarity in electro-modulated bilayer graphene under specific conditions and its distortion once these conditions are not satisfied. Our investigation is expected to stimulate further experimental (and, obviously, theoretical and numerical) research into the transport properties and the quantum Hall effect in such modulated structures.

It is also known that the low-energy wave functions of electrons are crucial for fully understanding the induced non-uniform magnetization in the bilayer-graphene system. Therefore, we present in figure 6 the calculated spatial dependence of eigenfunctions for some low-energy electronic states similar to those seen in figure 4(a). As shown by equation (12), these eigenfunctions correspond to a superposition of different harmonic-oscillator wave functions in the y direction. Here, the physical meaning of these 2(2N_L + 1) p components in a solved eigenvector represents the expansion coefficients in this superposition, where N_L is the number of LLs considered in numerical computations. As displayed in figure 6, the y dependence of the eigenfunction contains many displaced harmonic-oscillator wave functions, while its x dependence just presents a sinusoidal oscillation. As B_0 increases, more and more displaced harmonic-oscillator wave functions show up in the y dependence of the eigenfunction, along with a shorter spatial period for sinusoidal oscillations in its x dependence.

3.3. Effect of breaking down of inversion symmetry

For a monolayer graphene, the group of wavevectors associated with the K or K’ point within the crystal first Brillouin zone is found isomorphic to the point group [41] D_{3h}. For a bilayer graphene with a Bernal stacking, on the other hand, this D_{3h} point group is downgraded to D_3 with a lower symmetry. Furthermore, in the presence of a vertical bias field, these two point groups [41] become C_{3v} and C_3, respectively, for a gated monolayer graphene and a biased bilayer graphene. The loss of an inversion symmetry for a bilayer graphene under a vertical electric field has a profound effect on the formation of fractal Landau subbands in the presence of a square-scatter array potential, as can be seen from equations (8) and (13) where both LL-coupling coefficients C_n^α(ξ) and D_n^α(ξ) are ξ dependent and the intervalley coupling also becomes possible.

Compared with a monolayer graphene, a bilayer graphene can bring into additional valley mixing and site asymmetry after a perpendicular electric field E_0 has been applied. Such intervalley interference and electro-modulation effects can be seen clearly from equation (13) for the matrix elements of the scattering potential, i.e. the summation over ξ for fixed ξ and changing coefficients C_n^α(ξ) and D_n^α(ξ) with u and δ.
8

for \( n \geq 2 \). In figures 3–5, only a negligible electric field is employed (\( \delta \sim 10^{-3} \hbar \omega_c \)), and therefore, no visible distortions of the Hofstadter butterfly, which results from a square 2D periodically-modulated scattering-lattice potential, can be resolved. However, as \( \delta / \hbar \omega_c \) is slightly increased from \( 1 \times 10^{-3} \) to \( 2.5 \times 10^{-3} \) in figure 7 for a very weak modulation with \( V_0 / \hbar \omega_c = 0.5 \) and \( N = 1 \), we find from figure 7(b) that the previously found self-similar pattern within the third Landau band under \( \delta \sim 10^{-3} \hbar \omega_c \) is very strongly distorted, and therefore disappears.

Moreover, as \( \delta / \hbar \omega_c \) is further increased from \( 2.5 \times 10^{-3} \) to \( 10^{-1} \) in figure 8 for \( V_0 / \hbar \omega_c = 0.5 \) but \( N = 3 \), we find from a direct comparison between figures 7(b) and 8(b) that the previously observed self-repeated pattern within the fourth Landau band under \( \delta \sim 10^{-3} \hbar \omega_c \) is destroyed completely. Meanwhile, the mixing of the third and fourth Landau bands is seen clearly even for such a small modulation amplitude \( V_0 / \hbar \omega_c = 0.5 \) in contrast with the result in figure 4(b) for \( V_0 / \hbar \omega_c = 1.0 \).

4. Brief summary

In conclusion, we have developed a theoretical formalism to demonstrate the Hofstadter-type fractal band structure for bilayer graphene in the presence of a 2D periodic electrostatic modulation. The current work can be viewed as...
a generalization of the previous reported results based on Bloch-wave expansion approach applied to both a 2D electron gas [38] and a monolayer graphene [40]. As in previous studies [38, 40], this work includes explicitly deriving a non-perturbative eigenvalue equation, finding numerical solutions which display self-repeated split Landau subbands as a function magnetic flux and periodic subband dispersions as a function of electron wave number in a full magnetic Brillouin zone. Both Hofstadter butterflies and band mixing effects can be displayed simultaneously for a strong scattering strength which further reduces a required magnetic field for such observations to an accessible level.

Interestingly, we find two unique features for the bilayer-graphene system in this study. The first one is related to a bias-modulated mixing of and an interference from two valleys (i.e. non-vanishing intervalley scattering with $\xi, \xi' = \pm 1$) in the presence of a scattering-lattice potential. The second one, however, is associated with a lost inversion symmetry due to a perpendicular electric field, which tends to distort and even destroy the Hofstadter-type fractal band structures established by this scattering-lattice potential, as seen from figures 7 and 8.

The dependence of Bloch-wave expansion coefficients on the applied electric field directly leads to an electro-deformation of the Hofstadter-type subband splittings, resulting in strongly distorted or even destroyed self-repeated patterns.

We believe that apart from tremendous calculational and computational effort, our work represents substantial novelty and a scientific advance in studying magnetic properties of low-dimensional materials. First, our technique allows for a substantial tuning of $B_0$ required for achieving a visible Hofstadter band structure. The magnetic flux through a ‘supercell’, defining an effective 2D period for an electrostatic potential, is a flux ratio to the elementary magnetic-flux quantum. The size of the supercell could be up to hundred times larger than the atomic cell considered at all previous Hofstadter butterfly studies, and therefore, the required $B_0$ could be achieved in a usual lab setup. More importantly, such kind of studies have never been done for bilayer graphene.

By taking valley-coupling into consideration due to site asymmetry, this makes our study of bilayer-graphene unique and very distinguished from all other previous calculations. We have found a bias-modulated mixing of the two $K$ and $K'$ valleys, as well as their interference associated with different scattering paths. We have also shown the loss of inversion symmetry due to a perpendicular electric field, which tends to affect and eventually destroy the fractal band structures induced by this scattering-lattice potential. We should...
also mention that in contrast to all previous studies featuring Hofstadter band structure, our current work is a rare example demonstrating how the Hofstadter butterfly is distorted. In particular, it happens due to the application of an electric field, leading to a loss of sublattice and interlayer symmetry, which is completely absent in monolayer graphene.

In order to present all these features, we have developed a complete theoretical formalism for the electronic states and the band structure by taking into account 2D periodic electrostatic modulating field, which could now be used for a large number of device applications with an accessible magnetic-field strength, such as field controlled transport and optoelectronic studies. We believe the Hofstadter spectrum is closely related to electron conductivity, which implies that our novel results have both fundamental and practical impacts.

**Acknowledgments**

DH would like to acknowledge the financial support from the Laboratory University Collaboration Initiative (LUCI) program and from the Air Force Office of Scientific Research (AFOSR).

**ORCID IDs**

Danhong Huang https://orcid.org/0000-0001-6965-6786

**References**

[1] Novoselov K, Geim A K, Morozov S, Jiang D, Katsnelson M, Grigorieva I, Dubonos S and Firsov A 2005 Nature 438 197
[2] Geim A K and Novoselov K S 2007 Nat. Mater. 6 183
[3] Neto A C, Guinea F, Peres N, Novoselov K S and Geim A K 2009 Rev. Mod. Phys. 81 109
[4] Goerbig M 2011 Rev. Mod. Phys. 83 1193
[5] Zhang Y, Jiang Z, Small J, Purewal M, Tan Y W, Fazlollahi M, Chudov J, Jasczak J, Stormer H and Kim P 2006 Phys. Rev. Lett. 96 136806
[6] Zhang Y, Tan Y W, Stormer H L and Kim P 2005 Nature 438 201
[7] Novoselov K S, Jiang Z, Zhang Y, Morozov S, Stormer H L, Zeitler U, Maan J, Boebinger G, Kim P and Geim A K 2007 Science 315 1379
[8] Kane C L and Mele E J 2004 Phys. Rev. Lett. 95 226801
[9] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Science 306 666
[10] Oostinga J B, Heersche H B, Liu X, Morpurgo A F and Vandersypen L M 2008 Nat. Mater. 7 151
[11] Ohta T, Bostwick A, Seyller T, Horn K and Rotenberg E 2006 Science 313 1106
[12] Yan K, Peng H, Zhou Y, Li H and Liu Z 2011 Nano Lett. 11 438
[13] Novoselov K S,McCann E, Morozov S, Falco V I, Katsnelson M, Zeitler U, Jiang D, Schedin F and Geim A 2006 Nat. Phys. 2 177
[14] Henriksen E, Jiang Z, Tung L C, Schwartz M, Takita M, Wang Y J, Kim P and Stormer H 2008 Phys. Rev. Lett. 100 087403
[15] McCann E and Fal’ko V I 2006 Phys. Rev. Lett. 96 086805
[16] Hofstadter D R 1976 Phys. Rev. B 14 2239
[17] Azbel M Y 1964 Sov. Phys.—JETP 19 634
[18] Gumbs G and Fazlollahi M 1997 Phys. Rev. B 56 3787
[19] Nemec N and Cuniberti G 2006 Phys. Rev. B 74 165411
[20] Nemec N and Cuniberti G 2007 Phys. Rev. B 75 201404
[21] Dean C et al 2013 Nature 497 598
[22] Ponomarenko L et al 2013 Nature 497 594
[23] Woods C et al 2014 Nat. Phys. 10 451
[24] Hunt B et al 2013 Science 340 1427–30
[25] Schmidt H, Roel J C, Smirnov D and Haug R J 2014 Nat. Commun. 5 5742
[26] Yang W et al 2016 Nano Lett. 16 2387
[27] Wang L, Gao Y, Wen B, Han Z, Taniguchi T, Watanabe K, Koshino M, Hone J and Dean C R 2015 Science 350 1231
[28] Bistritzer R and MacDonald A 2011 Phys. Rev. B 84 035440
[29] Wang Z, Liu F and Chou M 2012 Nano Lett. 12 3833
[30] Apalkov V M and Chakraborty T 2014 Phys. Rev. Lett. 112 176401
[31] Brey L and Fertig H 2009 Phys. Rev. Lett. 103 046809
[32] Park C H, Yang L, Son Y W, Cohen M L and Louie S G 2008 Nat. Phys. 4 213
[33] Park C H, Yang L, Son Y W, Cohen M L and Louie S G 2008 Phys. Rev. Lett. 101 126804
[34] Kibis O 2010 Phys. Rev. B 81 165433
[35] Iurov A, Gumbs G, Roslyak O and Huang D 2011 J. Phys.: Condens. Matter 24 015303
[36] Iurov A, Gumbs G, Roslyak O and Huang D 2013 J. Phys.: Condens. Matter 25 135502
[37] Kibis O, Dini K, Iorsh I and Shelykh I 2017 Phys. Rev. B 95 125401
[38] Kühn O, Fessatidis V, Cui H, Selbmann P and Horing N 1993 Phys. Rev. B 47 13019
[39] Gumbs G, Iurov A, Huang D, Fekete P and Zhemchuzhna L 2014 AIP Conf. Proc. 1590 134–42
[40] Gumbs G, Iurov A, Huang D and Zhemchuzhna L 2014 Phys. Rev. B 89 241407
[41] Malard L M, Guimares M H D, Mafra D L, Mazzoni M S C and Jorio A 2009 Phys. Rev. B 79 125426