In a strong perpendicular magnetic field, the energy spectrum of a two-dimensional electron gas (2DEG) splits into a series of Landau levels. If a periodic potential is also added to this system, the noninteracting energy spectrum then displays an intricate fractal pattern known as the Hofstadter’s butterfly [1]. In the quantum Hall effect regime, the Coulomb interaction between electrons plays an important role both in the ground state and in the low-energy excitations. Theoretically, the Coulomb interaction effects on the butterfly states were investigated in the Hartree or mean field approximation [2–4], or with the electron correlations [5, 6]. In the Hartree approximation, the electrons are classical particles with negative charge and repel each other. This approximation is unable to deal with a spin/valley system such as graphene if the spin is not polarized, since the exchange interaction is not taken into consideration. In the exact diagonalization scheme, the electron correlations are completely included, and hence more adaptable to the fractional quantum Hall effect [7], but then only the finite-size systems can be handled in this scheme. Here we consider the Hartree–Fock approximation (HFA) to deal with the Coulomb interaction and the spin/valley system in graphene. Due to the crystallized structure of the density of the butterfly states [2], we are able to work in the momentum space to study an infinite system.

In contrast to the conventional 2DEG in a semiconductor, electrons in graphene must be described by the Dirac equation with an extra valley degree of freedom [19–21]. In this case, the Hartree approximation may not be enough to describe the behavior of the electrons if we consider both the spin and the valley. Indeed, the excitation gap measured in a recent experiment [13] cannot be explained either in the noninteracting picture or in the Hartree approximation. Here, we develop a theory based on the Hartree–Fock approximation to evaluate the energy gaps for integer filling factors. We also employ a method involving the crystalline state to explain the experimental results. The crystalline method establishes a picture of the electron gas in the thermodynamic limit, so that we could study the system at any magnetic flux, while only...
some discrete flux can be studied in a finite-size system with electron-electron interaction.

The Hofstadter states are not affected much by the geometry of the external potential. For simplicity and without loss of generality we introduce in the graphene Hamiltonian a square periodic potential \([23]\), \(V_{\text{ext}}(x,y) = V_0[\cos(q_0x) + \cos(q_0y)]\), where \(V_0\) is the amplitude of the potential and \(q_0 = 2\pi/a_0\) with a period of the potential \(a_0\). The Hamiltonian of the 2DEG in such a system is then given by \(H = H_0 + V_{\text{ext}}\). The non-interacting Hamiltonian \([19–22]\) without the external potential is

\[
H_0 = v_F \begin{pmatrix} 0 & P_x - i e A_y & P_y \\ P_x + i e A_y & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},
\]

where \(\eta = \pm 1\) for \(K\) and \(K'\) valley respectively, \(v_F\) is the Fermi velocity, and \(P = p + e A\) is the canonical momentum. We choose the Landau gauge to define the vector potential \(A = (0,B_x)\), and \(V_{\text{ext}}\) is the electron–electron interaction.

The period of the external potential is large enough experimentally \([12, 13]\) (larger than 10 nm), so that the valley mixing can be neglected. The energy bandwidth is narrow \([12, 13]\) (larger than 10 nm), so that the valley mixing can be neglected. The energy bandwidth is narrow. The Landau level (LL) mixing is also very weak and is not considered here. We first diagonalize the noninteracting Hamiltonian with the external potential, \(\tilde{H}_0 = H_0 + V_{\text{ext}}\) by using the basis \(\{\phi_{\sigma K}^n\}\), where \(\phi_{\sigma K}^n\) is the wave function of the \(n\)-th LL with valley-spin index \(\sigma\) and the guiding center \(X\) \([19–22]\). Then we could obtain \(4N_0\) eigenvectors:

\[
\begin{pmatrix} c_{1,x}^0 \phi_{1,x}^1 & \ldots & c_{1,x}^0 \phi_{1,x}^{N_0} & \ldots & c_{4,x}^0 \phi_{4,x}^1 & \ldots & c_{4,x}^0 \phi_{4,x}^{N_0} \end{pmatrix},
\]

where \(i = 1, \ldots, 4N_0\) and \(4N_0\) is the degeneracy of the Landau level in the finite sample. The coefficients \(c_{i,x}^0\) satisfy the normalization condition \(\sum_{\sigma} \sum_{j=1}^{N_0} |c_{i,x}^0|^2 = 1\). Note that \(i\) is also the index of the corresponding eigenenergies with ascending order and \(j\) is the guiding center index for the \(N_0\) states in a single LL.

The Hartree–Fock Coulomb interaction shall be determined self-consistently by the coefficients \(c_{i,x}^0\). Let us consider the Hartree term \(V_{\text{H}}\) and the Fock term \(V_{\text{F}}\) separately. The Hartree term \([2, 4, 23]\) is

\[
\langle \sigma', X | V_{\text{H}} | \sigma, X \rangle = \frac{\hat{\phi}_{\sigma'}}{N_0} \sum_{k} \sum_{\ell} \sum_{\kappa} \sum_{l} \frac{1}{G\ell} \times c_{k,x}^0 c_{l,x}^0 M_{k,\ell}(-G) M_{X,\ell}(G),
\]

where \(\kappa\) is the dielectric constant, the summation with a prim factor is defined by \(c_{k,x}^0\) to compute the Hartree–Fock Coulomb interaction. Then the full Hamiltonian \(H\) is diagonalized and a new group of coefficients \(c_{i,x}^0\) is obtained. By repeating this process the coefficients \(c_{i,x}^0\) and the energy spectrum can be evaluated self-consistently. The energy gap is then obtained from \(\Delta = E_{N_0+1} - E_0\). It is the gap between the highest occupied state and the lowest unoccupied state. This gap may be observed in transport or capacitance measurements.

The parameter \(\alpha\) defines the units of the magnetic flux \(\Phi\) per unit cell of the periodic potential, \(\alpha = \Phi/\Phi_0\), where \(\Phi_0\) is the magnetic flux quantum. Hence, \(\alpha\) also describes the magnetic field if the periodic potential is fixed. Moreover, the size of the sample is related to the period and \(\alpha = N_0 = L_y/L_x/(\alpha a_0^2)\), where \(L_x\) and \(L_y\) are the length of the sample in the \(x\) and \(y\) directions. In what follows, we would like to study the energy gaps in different magnetic fields or \(\alpha\) for a fixed filling factor \(\nu\). However, the size of the sample is not fixed \((L_x = L_y\) are not constant\) when we study the system for a continuous \(\alpha\).

**Crystal phase in an infinite sample:** If we calculate the energy spectrum for continuous \(\alpha\) in a fixed-size sample, the energy gaps may not be reliable since both the energy spectrum and the gaps are perhaps size-dependent. To calculate the energy spectrum in an infinite sample, we could work in the momentum space. To be consistent with the finite-size study, we consider a crystal phase of the electron gas with the same geometry as the periodic potential. The lattice constant of this electron crystal \(a\) is given by \(a = a\ell = \sqrt{2}\pi n_e/\nu\), where \(n_e\) is the electron number per crystal site. If the lattice constant of the electron crystal is identical to the period of the potential \(\alpha = a0\), then \(n_e = \nu/\alpha\).

The Hamiltonian of the 2DEG in the Hartree–Fock approximation is written

\[
H = \sum_{\sigma} E_{\sigma}(\rho_{\sigma}(q) = 0) + \sum_{q,\sigma} V_{\text{ext}}(q) \rho_{\sigma}(q) \\
+ \sum_{q,\sigma} U_{\sigma}(q) \rho_{\sigma}(-q) + \sum_{q,\sigma} U_{\sigma}(q) \rho_{\sigma}(q). 
\]

The density matrix operator is

\[
\langle \sigma', X | V_{\text{F}} | \sigma, X \rangle = \frac{e^2}{\pi \ell N_0} \sum_{k} \sum_{\ell} \sum_{\kappa} \sum_{l} \frac{1}{G\ell} \times c_{k,x}^0 c_{l,x}^0 M_{k,\ell}(-G) M_{X,\ell}(G),
\]

with a Laguerre polynomial \(L\) (we define \(L_{n<0} = 0\) and the magnetic length \(\ell = \sqrt{\mu/eB}\). The Fock term is

\[
\langle \sigma', X | V_{\text{F}} | \sigma, X \rangle = \frac{e^2}{\pi \ell N_0} \sum_{k} \sum_{\ell} \sum_{\kappa} \sum_{l} \frac{1}{G\ell} \times c_{k,x}^0 c_{l,x}^0 M_{k,\ell}(-G) M_{X,\ell}(G). 
\]
where operators $c_{\sigma,X,\epsilon}$ and $c_{\sigma,X,\epsilon}^\dagger$ are the annihilation and creation operators of electrons in valley-spin $\sigma$ and the guiding center $X$. The Hartree and Fock interaction functions, $U_H$ and $U_X$, are defined by

$$U_H(q) = \frac{e^2}{\kappa \ell} \frac{1}{q^2} [M(q) \epsilon_{q,1/2} - q \epsilon_{q,1/2}](q)^2,$$

$$U_X(q) = \frac{e^2}{\kappa \ell} \int dp \left[ \frac{M(p)}{p} \right] J_0(pq\ell),$$

where $J_0$ is the Bessel function. The external potential in such an electron crystal is

$$V_{\text{ext}}(q) = \frac{V_0}{2} [M(q) \epsilon_{q,1/2} - q \epsilon_{q,1/2}](q)^2.$$  

We define the Green’s function $G_{\sigma,s}(X, X', \tau) = \langle \rho_{\sigma,s}(X', \tau) \rho_{\sigma,s}(X, \tau) \rangle$, where $T$ is the time order operator. Then at zero temperature,

$$G_{\sigma,s}(q, \tau = 0^+) = \langle \rho_{\sigma,s}(q) \rangle,$$  

where $G_{\sigma,s}(q, \tau)$ is the Fourier transform of $G_{\sigma,s}(X, X', \tau)$. In the Matsubara frequency $\omega_n$, the equation of motion of the Green's function

$$\left(i\hbar \omega_n - E_d\right)G_{\text{d},d}(q, \omega_n) = h_{\text{d},d}(q, 0) + \sum_q V_{\text{ext}}(q) e^{i q \cdot X} \epsilon_{q,1/2} G_{\text{d},d}(q + q', \omega_n)$$

$$+ \sum_{\sigma} U_H(q) \langle \rho_{\sigma,s}\rangle e^{i q \cdot X} \epsilon_{q,1/2} G_{\text{d},d}(q + q', \omega_n)$$

$$- \sum_{\sigma} U_X(q) \langle \rho_{\sigma,s}\rangle e^{i q \cdot X} \epsilon_{q,1/2} G_{\text{d},d}(q + q', \omega_n),$$

where $d, \epsilon$ are valley-spin indices, can be solved self-consistently (see [26] for details). Then all the elements of the density matrix can be obtained according to equation (11).

The density matrix completely describes the system with the Hamiltonian in equation (6). The energy spectrum is thus obtained by solving the equation of motion in equation (12). The Fermi level is given by the sum rule of fixing the filling factor [26]. When we estimate the energy gap, we need to subtract the energy of the lowest unoccupied state by the energy of the highest occupied state in the density of states (DOS). The relation between the DOS $g$ and the retarded Green’s function is [27]

$$g(\omega) = -\frac{N_\nu}{\pi} \sum_\sigma \Im \left[ G_{\sigma,\sigma}^R(0, \omega_n \to \omega + i0^+) \right].$$

The crystalline electron gas was studied in monolayer graphene [25] and in graphene bilayer [27] without the external potential. Wigner crystals and skyrmion crystals can be found in those systems for non-integer filling factors. Moreover, the skyrmion crystals were found in bilayer [28] and trilayer graphene [29] even for integer filling factors, due to the Dzyaloshinskii–Moriya (DM) interaction. We employ a similar method to study the electron gas in the presence of the external potential. For a strong potential the 2DEG may be crystallized with the same geometry as the potential for integer filling factors even without the DM interaction.

In a recent experiment [13], there is about $1^\circ$ misalignment between graphene and the BN substrate with the dielectric constant $\kappa = 8$. The period of the moiré pattern is about 100 times larger than the lattice constant of graphene. Here we fix the period of the potential, $a_0 = 30$ nm. We could then neglect the weak Landau level mixing. The period is large enough to avoid the valley mixing. Hence, we could set the valley the same as spin. The valley pseudo-spin is conserved in the HFA. In order to be consistent with the observed results, we consider $\alpha \in [1, 2]$, so $1/\alpha \in [0.5, 1]$. (In [13] $1/\alpha$ is in this paper).

Because of the fractal pattern, the noninteracting energy spectrum in the region $\alpha \in [1, 2]$ is similar to that in the region $\alpha \in [0, 1]$.

Finite-size study for $\nu = -1$: For the finite-size study of the energy spectrum in the HFA, we fix the size of the sample and vary the magnetic field (or the parameter $\alpha$). The amplitude of the potential, is fixed, $V_0 = 20$ meV. The size of the sample is $6a_0 \times 6a_0$. For the filling factor $\nu = -1$ in the Landau level $N = 0$, there is $4N_\nu$ degeneracy if we do not consider the Zeeman coupling, but there are only $N_\nu$ electrons in this LL, $N_\nu = N_\nu$. So in this system, $N_\nu = [36/\alpha]$, where $[x]$ is the largest integer not exceeding $x$.

We find that the calculated energy gap oscillates with $\alpha$ as the magnetic field increases. We however note that the finite-size study cannot deal with the liquid phase. If the external potential is not strong enough or $\alpha$ is not small enough, the broadening of a LL is not large, then the Coulomb energy could force the ground state to stay in a liquid phase in which the density of the 2DEG is homogeneous everywhere. Hence, the finite-size study may not be very reliable. In fact, the solution of equation (11) is predicated on the size of the sample being infinite. Moreover, it is easier to estimate the energy of the system in the crystal method. So we could compare the energy of the fractal phase with the liquid phase. We find that the energy of the fractal phase is higher than that of the liquid phase when $\alpha > 1$. In this case, the fractal phase which basically contains a crystal density profile, is only a metastable state of the system. Nevertheless, we could calculate the energy gap of the metastable fractal phase in the crystal method to compare with the finite-size results. The energy gap also oscillates with $1/\alpha$. However, the amplitude and the peaks of the oscillation are changed. This might be because the system is size-dependent when the size is finite. Generally, the results of the two different calculations are similar.

**Effects of the Fock interaction**: We set the same parameters as in Ref. [2], $V_0 = 4$ meV, $a_0 = 50$ nm, and $\kappa = 12.4$ for GaAs, but only at zero temperature. For $\alpha = 1/3$ and $\nu = 1/3$ in the lowest LL and without LL mixing we calculate the energy spectrum of the butterfly state, which is essentially displayed by the DOS of the electron gas. We can turn off the Hartree or Fock interaction in equation (6) to study the energy spectrum of the electron gas in different approximations. In figure 1(a), the noninteracting DOS shows the width of the energy spectrum to be 6.4 meV, while in figure 1(b), the energy spectrum in the Hartree approximation is 3.8
meV. These results agree very well with figure 8 of [2]. For other \( \eta \), our numerical results all agree well with figure 8 of [2]. The Hartree interaction decreases the band width of the spectrum. However, in figure 1(c), when we take the Fock interaction into account the bandwidth broadens significantly. The bandwidth is even larger than that of the noninteracting case. Note that the lower energy gap is larger than the higher one in the HFA, while the case is reversed in the Hartree approximation.

Energy gaps in an infinite sample for \( \nu = -1, 0 \): We now focus on the filling factors \( \nu = -1, 0 \), in the \( N = 0 \) LL. As what we have mentioned above, the liquid phase (or say the quantum Hall ferromagnetic state) of which the energy per electron is \( E_0 + U (0) \) still exists if the potential is not strong or \( \alpha \) is not small, e.g. when \( V_0 < 21 \) meV, the ground state is always a liquid phase for \( \alpha \gg 1 \). Generally, the fractal phase only exists when the external potential is strong, i.e. \( V_0 \) or the period \( d_0 \) is large. In order to compare our results with a recent experiment [13], we assume \( V_0 = 23 \) meV hereafter.

Figures 2(a) and (b) show the energy gaps for the filling factors \( \nu = -1, 0 \), respectively. These two curves are similar: the gaps are not monotonous. Energy gaps increase with \( 1/\alpha \), and then decrease to almost zero for \( \alpha = 1 \). In both cases there is a phase transition between the liquid phase and the fractal phase. Figure 2(c) indicates the comparison of energies of the two phases around the phase transition at \( \nu = 0 \). The spin is polarized in the liquid phase, but is unpolarized in the fractal phase. The potential amplitude \( V_0 \) is larger, the phase transition occurs at larger \( \alpha \). The gap for \( \nu = -1 \) is larger than that of \( \nu = 0 \) since the gap of \( \nu = 0 \) involves the Zeeman gap. Our numerical results show very similar behavior as observed in the experimental measurements [13]. We note that the energy gap calculated in the liquid phase is only due to the electron-hole excitation. This excitation is affected by the external potential since the gap curve is nonmonotonic and is not proportional to \( \sqrt{B} \). Another candidate, the skyrmion excitation [24, 30] may be degenerate to a quasi-particle in the presence of the potential, which is beyond the scope of this work. We can not obtain a skyrmion crystal ground state in the fractal phase either. It should be because the electron density is much higher than what the skyrmion crystal was found numerically [25]. The (pseudo-) spin textures are suppressed by the high density electron gas: (pseudo-) spin flipping can not decrease enough energy to create a (pseudo-) spin texture. The reason why our numerical results for \( \nu = 0 \) is not exactly the same as in the experiment is because perhaps the ground state in the \( N = 0 \) LL is not spin polarized without the external potential [31], and we only consider a spin polarized liquid ground state here.

Fractal phase in an infinite sample for \( \nu = 4 \): For \( \nu = 4 \), our numerical results shown in figure 3 are also very similar to those observed in the experiment [13]. In the higher LLs the external potential is easier to convert the quantum Hall state to a fractal phase. We find that the ground state is a fractal state when \( \alpha < 2.2 \) for \( \nu = 4 \). When \( \alpha = 1.5 \), which is equivalent to \( \alpha = 0.5 \), a previous study [4] have shown that the gap between the two bands would be open when the spin is polarized and one valley is half-filled in the Hartree approximation. In this work, we take the spin into consideration. The ground state is no longer spin polarized. The Zeeman coupling is very weak (about 1 meV) while the amplitude of the potential is 23 meV. The potential is strong enough to mix different spins. The gap for \( \alpha = 1.5 \) is very small in figure 3. This is because, intuitively, the two spins are mixed and the corresponding four bands (two for each spin) in one valley are overlapped to close the Hartree gap. In the DOS we clearly see the energy band structures. The DOS for \( \bar{\sigma} \in [1, \sqrt{2}] \) is similar to the DOS for \( \alpha = 3 - \bar{\sigma} \). For simplicity, we show the DOS for \( \alpha = 1.4 \) only in figure 4. Note that all the bands mix both spins. A spinless picture is not satisfied in this spin/valley system.

We now define the spin field [32] in valley \( \eta (K \text{ or } K') \)

\[
\delta_{\eta x} + i \delta_{\eta y} = \langle \rho_{\eta x}(\bar{\sigma}, \eta)| \rho_{\eta y}(\bar{\sigma}, \eta)| \rangle,
\]

(14)
The spin field contains no texture at (b). The spin field, in units of \( \frac{1}{(2\pi)^2} \), is also crystallized. Note that the maximum points of electron crystal are not a skyrmion crystal. The electron gas tends to be in a liquid phase, but the strong external potential crystallizes it. The HFA is reliable in the integer quantum Hall regime and the studies of the Wigner and skyrmion crystals. The transport properties given in the HFA are reliable at least qualitatively. Moreover, the electron crystal may be observed in a scan tunelling measurement [27, 33]. We propose that this method is able to study the interacting (in the HFA) Hofstadter’s butterfly states with different periodic potentials in an infinite system conveniently and efficiently, not only in graphene, but also in other Dirac materials.

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Figure 3. Energy gaps for \( \nu = 4 \) in LL \( N = 1 \).

Figure 4. The DOS for \( \nu = 4 \), and for \( \alpha = 1.4 \). The arrow indicates the Fermi level location.

\[
S_{\alpha z} = \langle \rho_{(\eta,1),1}(r) \rangle - \langle \rho_{(\eta,1),1}(r) \rangle.
\]

The density in valley \( \eta \) is given by \( n_{\eta}(r) = \sum_{s} \langle \rho_{(\eta,s),1}(r) \rangle \), where \( s \) is the spin index. The two valleys are completely equivalent in our numerical results, so only the order parameters in the \( K \) valley are shown in figure 5. The density profiles have the same geometry as that of the external potential. There is no valley coherence and is not shown in figure 5, i.e. \( \langle \rho_{(K,\alpha),1}(K,x) \rangle = 0 \). The spin field contains no texture at all, \( S_{\alpha \alpha} = S_{\alpha \beta} = 0 \), so the electron crystal is not a skyrmion crystal. Only the \( z \)-components are nonzero, \( S_{z z} \neq 0 \), and \( S_{z z} \) is also crystallized. Note that the maximum points of \( S_{\alpha z} \) do not match the maximum points of the density, where the minimum points of the external potential are. At these points the potential decreases the kinetic energy for both spins. The electrons with both spins overcome the repulsive interaction to be localized by the potential. The density of electrons is minimum at the sites where the energy of the potential is maximum. At the points where the density of electrons is maximum or minimum, the spin field \( S_{\alpha z} \) is minimum (the blue dots in figure 5(b)).

In conclusion, inspired by a recent experiment [13], we have studied the interacting Hofstadter’s butterfly states in the HFA in order to study the spin/valley systems such as graphene. We show that the Fock interaction plays an important role by comparing with our numerical results with previous work in figure 1. In graphene, with the Coulomb interaction in the HFA, we observe a phase transition from a quantum Hall state to a fractal state with the increase of the magnetic field.
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