Electronic and optical properties of monolayer and bilayer graphene

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The electronic and optical properties of monolayer and bilayer graphene are investigated to verify the effects of interlayer interactions and external magnetic field. Monolayer graphene exhibits linear bands in the low-energy region. Then the interlayer interactions in bilayers change these bands into two pairs of parabolic bands, where the lower pair is slightly overlapped and the occupied states are asymmetric with respect to the unoccupied ones. The characteristics of zero-field electronic structures are directly reflected in the Landau levels. In monolayer and bilayer graphene, these levels can be classified into one and two groups, respectively. With respect to the optical transitions between the Landau levels, bilayer graphene possesses much richer spectral features in comparison with monolayers, such as four kinds of absorption channels and double-peaked absorption lines. The explicit wave functions can further elucidate the frequency-dependent absorption rates and the complex optical selection rules. These numerical calculations would be useful in identifying the optical measurements on graphene layers.

Keywords: graphene; band structure; magneto-optical property; Landau level

1. Introduction

Graphene is a densely packed single layer of carbon atoms as well as the basic building block of bulk graphite. In the past few years, few-layer graphene sheets have attracted a considerable amount of experimental and theoretical interest, mainly owing to the experimental achievements in the epitaxial growth on silicon carbide (Ohta et al. 2006; Berger et al. 2007) and the mechanical exfoliation from highly oriented pyrolytic graphite (Novoselov et al. 2004, 2005, 2006; Zhang et al. 2005). They are regarded as ideal layered materials to explore the quantum phenomena in two-dimensional electron systems. The one-atom-thick crystal of carbon atoms is constructed by the strong covalent bonds between sp² hybrid orbitals. Its hexagonal structure means that monolayer graphene possesses linear...
and isotropic energy dispersions in the low-energy region (Castro Neto et al. 2009). However, these energy bands are substantially altered by the coupling between layers (Latil & Henrard 2006; Lu et al. 2006; Novoselov et al. 2006; Ohta et al. 2006, 2007; Partoens & Peeters 2006; Abergel & Fal’ko 2007; Gruneis et al. 2008). Bilayer graphene with Bernal stacking is one of the simplest coupled layers. Its interlayer atomic hoppings effectively convert the linear bands into two pairs of parabolic bands, and induce asymmetry between valence and conduction bands (Latil & Henrard 2006; Novoselov et al. 2006; Ohta et al. 2006, 2007; Partoens & Peeters 2006; Abergel & Fal’ko 2007; Gruneis et al. 2008). These subband characteristics can be directly observed through the measurements of angular-resolved photoemission spectroscopy (Ohta et al. 2006, 2007), which is capable of identifying the layer number and the stacking consequences of graphene layers. Furthermore, an external electric or magnetic field can effectively modulate these electronic properties (Novoselov et al. 2005, 2006; Zhang et al. 2005, 2009; Lu et al. 2006; Ohta et al. 2006; Li & Andrei 2007; Ho et al. 2008a). In this work, numerical calculations of electronic and optical properties are performed for monolayer and bilayer graphene. Special emphasis is given to the effects of interlayer interactions and uniform magnetic field.

In the presence of an external magnetic field, the planar motions of electrons are effectively quantized to form dispersionless Landau levels. These energy levels exhibit more pronounced spectral features than those at zero field, which gives rise to a better understanding of intrinsic physical properties in graphene systems, as well as the estimation of hopping intensity between atomic sites (Nakao 1976; Matsui et al. 2005; Novoselov et al. 2005; Deacon et al. 2007; Li & Andrei 2007; Orlita et al. 2008). Unlike the conventional electron gas of metals, monolayer graphene possesses Landau levels with nearly square-root dependence on field. These levels are not equally spaced; in particular, one of them is located right at the charge neutrality point (Novoselov et al. 2005; Zhang et al. 2005; Sadowski et al. 2006; Jiang et al. 2007; Deacon et al. 2007). With regard to bilayer graphene, the Landau levels derived from parabolic bands are close to being linearly dependent on field (Novoselov et al. 2006; Abergel & Fal’ko 2007; Li & Andrei 2007; Henriksen et al. 2008; Lai et al. 2008). The interlayer interactions dominate these quantized electronic states, which can be examined through measurements of quantum Hall conductivity (Zhang et al. 2005; Novoselov et al. 2006, 2005; Berger et al. 2007), scanning tunnelling spectroscopy (Matsui et al. 2005; Li & Andrei 2007) and cyclotron resonance (Sadowski et al. 2006; Deacon et al. 2007; Jiang et al. 2007; Henriksen et al. 2008; Orlita et al. 2008; Plochocka et al. 2008).

From the theoretical aspect, previous calculations of graphene Landau levels were mainly based on the effective-mass model (Zheng & Ando 2002; Abergel & Fal’ko 2007; Koshino & Ando 2008) and Peierls tight-binding model (Chang et al. 2004, 2005; Ho et al. 2008b; Lai et al. 2008). The former is a kind of approximation method with respect to the Fermi-momentum states (K-point), which enables one to properly evaluate the Landau levels of monolayer and bilayer graphene. The interlayer atomic hoppings are treated as perturbations, with some of them being neglected in calculation. This simplified model successfully reproduces the linear- and parabolic-band associated Landau levels. Nevertheless, it is not suitable for higher Landau levels since the actual zero-field subbands gradually deviate from the ideal linear or parabolic dispersions with the increment of state energy. In contrast, the Peierls tight-binding model is relatively reliable for Landau levels.
over a much wider energy range while its calculation takes account of all interlayer interactions and field-induced Peierls phase simultaneously (Chang et al. 2004, 2005; Ho et al. 2008b; Lai et al. 2008). The resulting Landau levels can directly reflect the zero-field subband features. In addition, the explicit distribution of wave function over the constituent sublattices is also attainable, which can be adopted to clearly define the quantum index of individual Landau levels. The wave function is an essential physical quantity that gives access to further transport and excitation properties.

In this work, the tight-binding model is employed to calculate the electronic and optical properties of monolayer and bilayer graphene. The effects of both interlayer interactions and magnetic field are investigated in detail. The former effectively brings about the second-group Landau levels, lifts the electron–hole symmetry, and modifies the distribution of energies and wave functions. Accordingly, bilayer graphene is expected to possess much richer optical properties than monolayers. The optical spectra with linearly polarized light are studied within the gradient approximation (Johnson & Dresselhaus 1973; Lin & Shung 1994; Chang et al. 2004). The transitions between Landau levels are revealed as pronounced peaks, where their evolution with field closely relates to hopping integrals. In bilayers, the appearance of the second-group Landau levels leads to four kinds of absorption peaks. And the subband asymmetry appears as double-peak absorptions. The way the wave functions are constituted by sublattices provides a direct way to resolve the optical selection rules and the absorption rates in response to photon energy and field strength. The distinct spectral features between monolayer and bilayer graphene are examined in detail. A comparison between zero-field and low-field limited spectral features is also made. The numerical results would be useful in identifying magneto-optical measurements (Sadowski et al. 2006; Deacon et al. 2007; Jiang et al. 2007;
Henriksen et al. 2008; Orlita et al. 2008; Plochocka et al. 2008). In particular, the absorption frequencies in the low field limit would be useful to estimate the value of certain hopping parameters.

2. Electronic structures

The geometry of monolayer and bilayer graphene is sketched in figure 1. The hexagonal crystal is composed of inequivalent \( A \) and \( B \) atoms. The primitive unit cell is shaded in grey, which contains two atoms for monolayer (\( A, B \)) and four atoms for bilayer (\( A_1, B_1, A_2, B_2; \) the \( A \) and \( B \) on respective layers) graphene. At \( B_0 = 0 \), the tight-binding Hamiltonian is a \( 2 \times 2 \) (\( 4 \times 4 \)) square matrix for monolayer (bilayer), where its elements are given by

\[
\langle k_f | H | j_l \rangle = \gamma_s (\vec{R}_{jl}, \vec{R}_{kl}) \sum \exp [ik \cdot (\vec{R}_{jl} - \vec{R}_{kl})],
\]

where \( j(k) \) stands for the inequivalent \( A \) or \( B \) site and \( l(l') = 1, 2 \) for the layer index. The tight-binding wave function is a linear combination of atomic basis \( \psi = \sum_{j,l} c_j | j_l \rangle \), where \( | j_l \rangle = \sum (1/N) \exp \{ik \cdot \vec{R}_{jl}\} \). The atomic basis \( | j_l \rangle \) symbolizes \( | A \rangle \) and \( | B \rangle \) for monolayer graphene and \( | A_1 \rangle, | B_1 \rangle, | A_2 \rangle \) and \( | B_2 \rangle \) for bilayer graphene. \( \gamma_s (\vec{R}_{jl}, \vec{R}_{kl}) \) contains information about the relative hopping integrals between atoms (Nakao 1976; Partoens & Peeters 2006). Only the nearest-neighbouring \( A-B \) hopping (\( \gamma_0 = -3.12 \text{eV} \)) is taken into account for the in-plane interactions. The coupling between layers includes the direct \( A_1-A_2 \) hopping (\( \gamma_1 = 0.38 \text{eV} \)), the \( B_1-B_2 \) hopping (\( \gamma_3 = 0.28 \text{eV} \)), the \( A_1-B_2 \) hopping (\( \gamma_4 = 0.12 \text{eV} \)), and the enhancement of site energy on \( A_1 \) and \( A_2 \) atoms (\( \gamma_6 = 0.016 \text{eV} \)).

In the presence of a perpendicular magnetic field \( \vec{B} = B_0 \hat{z} \), the vector potential \( \vec{A} = (0, B_0 x, 0) \) makes the Peierls phase \( \Delta G(\vec{R}_{jl}, \vec{R}_{kl}) = \int_0^1 (\vec{R}_{kl} - \vec{R}_{jl}) \cdot \vec{A} [\vec{R}_{jl} + \lambda (\vec{R}_{kl} - \vec{R}_{jl})] \text{d} \lambda \) accumulate in the Bloch wave function (Luttinger 1951; Chang et al. 2004, 2005; Ho et al. 2008b; Lai et al. 2008). The corresponding Hamiltonian elements are expressed as

\[
\langle k_f | H | j_l \rangle = \gamma_s (\vec{R}_{jl}, \vec{R}_{kl}) \sum \exp \left[ ik \cdot (\vec{R}_{jl} - \vec{R}_{kl}) + i \frac{e}{\hbar} \Delta G(\vec{R}_{jl}, \vec{R}_{kl}) \right].
\]

The Peierls phase term is described as a path integral of vector potential, where its variation period closely relates to the penetrating magnetic flux \( \phi \). A flux quantum \( \phi_0 \) through hexagonal crystalline graphene is of the order of 79 000 T, which is far beyond the experimental reach. With the diminishing \( \phi \), the primitive cell has to be \( 2R_B \)-times larger (\( R_B = \phi_0/\phi \)), in order to satisfy the whole periodic boundary condition. Meanwhile, the Hamiltonian matrix turns into \( 2R_B \times 2R_B \)-times larger than the zero-field cases. To solve the huge matrix problem, we rearrange the matrix elements, convert the matrix into a band-like one and reduce its dimension based on the localized features of wave functions. In this manner, both the eigenvalues \( E^{c,v} \) and eigenvectors \( \psi^{c,v} \) are efficiently resolved, even for a field down to 0.5 T. For each Landau state, its wave function can be represented as constituent sublattices; \( A_i \) and \( B_i \) for monolayers and \( A_{1,i}, B_{1,i}, A_{2,i} \) and \( B_{2,i} \) for bilayers (\( i = 1 \sim 2R_B \)). For \( B_0 < 60 \text{T} \), the spin-splitting effect is insignificant enough to be neglected in our calculation.

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The electronic density of states per atom per unit area is written as

$$D(\omega; B_0) = \sum_n \int_{\text{1st BZ}} \frac{\partial k_x \partial k_y}{4\pi^3} \frac{\Gamma}{[\omega - E_n^{c,v}(B_0; k_x, k_y)]^2 + \Gamma^2},$$

(2.3)

where $\Gamma = 1\text{ meV}$ is the broadening parameter. The characteristics of energy bands would be revealed as specific spectral signatures in the density of states. In the case of an external magnetic field, the quantized Landau levels appear as the delta-function peaks, where the peak positions and intensities provide a simple way to understand the intrinsic electronic properties of graphene layers.

At zero field, monolayer graphene exhibits isotropic and linear bands in the low-energy region, as shown in figure 1a. These bands intersect exactly at $E_F$ which corresponds to the special point $K$ in the reciprocal space of the hexagonal lattice. The energy dispersions would gradually deviate from the linear relation in the higher energy region. In the density of states, the Dirac-cone-like energy bands result in the V-shaped structure with vanishing carrier density at $E_F$ (the bottom curve in figure 4a). Accordingly, monolayer graphene is a kind of zero-gap semiconductor. With respect to the coupled bilayer system, the interlayer interactions convert the linear bands into two pairs of parabolic bands, as depicted in figure 1b. Each pair is composed of the occupied valence and unoccupied conduction bands. They are asymmetric with respect to $E_F$. The first pair (black curves) slightly overlaps around $E_F$, and thus results in the finite density of states at the charge neutral point. This also implies semimetallic behaviour. The second pair (red curves) is in the higher energy region, where its band edges maintain at $K$ point with energies $0.396\text{ eV} (\gamma_1 + \gamma_6)$ and $-0.364\text{ eV} (-\gamma_1 + \gamma_6)$. The two onset energies are revealed as the cusp-like singularities in the density of states (the bottom curve in figure 4b). Also, with the increment of state energy, the two pairs of parabolic bands would gradually change over into linear-like dispersions.

The perpendicular magnetic field can effectively quantize the two-dimensional electronic states into dispersionless Landau levels. These energy levels are momentum-independent, and their energies against field strength are illustrated in figures 2c and 3e for monolayers and bilayers. Each Landau level is composed of fourfold degenerate states with similar characteristics. The wave functions of the four states are, respectively, located around $x = 1/6, 1/3, 2/3$ and $5/6$ of the extended unit cell relative to the $\Gamma$ point ($k_x = k_y = 0$). In the case of monolayer graphene, the wave function is composed of two inequivalent sublattices ($A_i$ and $B_i$), where those of $x = 1/6$ at 40 T are depicted in figure 2a,b. Each curve can be approximated by the product of the Hermite polynomial of order $n$ and the Gaussian function. In particular, the node number is $n$ for $A$ site with respect to $n - 1$ for $B$. Each Landau state is almost equally governed by $A$- and $B$-sublattices, except for the $n = 0$ state governed by $A$ only. Accordingly, the node number of $A$-sites is chosen to define the quantum index $n^{c,v}$ of individual Landau levels, where $v$ stands for electrons and $c$ for holes. The distribution widths correspond to the magnetic lengths; they broaden with the increment of Landau energy, as do their node numbers $n$. In addition to clearly define level indices, how the wave function distributes over its constituent sublattices deserves a closer examination for further excitation and transport properties.
Turning our attention to the Landau energies, the valence and conduction states are symmetric with respect to $E_F$. The particular $n = 0$ level is located right at $E_F$ which is made up of both electron and hole carriers. Landau energy is a function of quantum number $n_{c,v}$ and field strength $B_0$, which is characterized by $E_{n_{c,v}}^{c,v} \propto \pm \sqrt{n_{c,v}B_0}$. This specific dependence can be ascribed to the linear bands at zero field, where the level spacings correlate to the in-plane hopping integral $\gamma_0$. However, Landau levels in the higher energy region or at a stronger field would gradually deviate from this simple field dependence since the corresponding electronic states at $B_0 = 0$ exceed the linear dispersion relation. Meanwhile, Landau levels have neither $\sqrt{B_0}$ nor $B_0$ dependence.

With regard to the Landau levels in bilayer graphene, their wave functions consist of four sublattices, $A_1$, $B_1$, $A_2$ and $B_2$. Those at 40 T are shown in figure 3a–d. Based on the relative distribution among atomic sites, Landau levels can be classified into two groups. The first group (black curves) is mainly dominated by the $B_{1,2}$ atoms, while the second group (red curves) by $A_{1,2}$. Accordingly, the node numbers of $B_{2}$- and $A_{1,2}$-sublattices are, respectively, adopted to define the quantum index of the first- and second-group Landau levels, $n_1^{c,v}$ and $n_2^{c,v}$ ($n_{c,v}^{c,v} = n_{c,v,eff}^{c,v}$ in Lai et al. (2008)). With the increment of state energy, the minor sites would gradually increase their proportion in the entire

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**Figure 2.** Landau levels of monolayer graphene: the wave functions at 40 T in terms of (a) $A$- and (b) $B$-sublattices. (c) Landau energies against field strength for $n_{c,v}^{c,v} \leq 15$.
Figure 3. Landau levels of bilayer Bernal graphene: the wave functions at 40T in terms of (a) $A_1$, (b) $B_1$, (c) $A_2$- and (d) $B_2$-sublattices. (e) Landau energies against field strength for $n_{c,v}^1 \leq 15$ and $n_{c,v}^2 \leq 5$. Those owing to the first-group Landau levels are represented by black and the second-group by red.

Figure 4. Density of states in (a) monolayer and (b) bilayer graphene. Curves from the bottom to the top correspond to 0–60T. The red circles label signatures of the second-group Landau levels.

wave function; they are $A_{1,2}$ for $n_{c,v}^1$ and $B_{1,2}$ for $n_{c,v}^2$. The node number between inequivalent atomic sites is $A_1:B_1:A_2:B_2 = n + 1 : n : n + 1 : n + 2$, except for the two lowest levels $n_{c,v,eff}^1 = 0, 1$ dominated by $B_2$ only. As to the positions of

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Landau levels, the first group $n_1^{c,v}$ (black curves) distributes away from $E_F = 0$ while the second group $n_2^{c,v}$ (red curves) appears in the higher energy region (approx. 0.4 eV).

Figure 3 illustrates the dependence of bilayer Landau levels on field strength for $n_1^{c,v} \leq 15$ and $n_2^{c,v} \leq 5$. As a result of interlayer interactions, the distribution of Landau levels in bilayers is quite different from that in monolayers. The level spacings evidently decrease, and the zero-energy level splits into $n_1^{c,v,\text{eff}} = 0, 1$ that gives rise to a small field-dependent energy gap. The symmetry between electron and hole states is absent; more specifically, the Landau levels are denser in occupied states than in unoccupied states for the first group levels, and vice versa for the second group ones. The Landau levels converted from the zero-field parabolic bands scale nearly linearly with field, in contrast to being proportional to $\sqrt{B_0}$ in monolayers. More precisely, they are close to being linearly dependent for low-lying levels and in the weaker field only, but involved as a combination of $\sqrt{B_0}$ and $B_0$ elsewhere. The absence of simple $B_0$ dependence can be easily explained in that the corresponding zero-field subbands are out of the parabolic band region. In the limit of low field, the first group of Landau levels $E_c^{v}(n_1)$ eventually falls to the charge neutral point, which is similar to the monolayer Landau levels. However, the second group $E_c^{v}(n_2)$ converges towards finite energies, $0.396 \text{eV} (= \gamma_1 + \gamma_6)$ for conduction states and $-0.364 \text{eV} (= -\gamma_1 + \gamma_6)$ for valence states. The appearance of second-group Landau levels is identified as the quantization of the second-pair parabolic bands, where the two onset energies reflect the parabolic band edges.

Landau levels are revealed as symmetric divergences in the density of states, as shown in figure 4a, b for monolayer and bilayer. This is one of the proper Landau-level spectra which clearly reveals information about Landau energies and Landau-level degeneracy simultaneously. The reducing $B_0$ results in the less degenerate states accommodated in the denser Landau levels. In principle, these divergences should be equal in intensity, except for the broadening-induced overlap which probably occurs in the higher energy region. However, in bilayer spectra, several peaks are evidently more pronounced than the others. These peaks are ascribed to the overlap of the two groups of Landau levels with the approaching state energy. Peaks due to the second-group Landau levels $(n_2^{c,v} = 0, 1, 2)$ in both valence and conduction states are marked with red circles in figure 4b.

The calculations in the framework of the Peierls tight-binding model are feasible for $\pi$-electron-associated Landau levels since they take account of the magnetic field and interlayer interactions simultaneously. Compared with a flux quantum, the field strength available nowadays in laboratories is extremely small; hence, the field acts as the quantization of zero-field states with approximate energy into dispersionless Landau states. The atomic hoppings between layers essentially alter the two-dimensional electronic structures and consequently the quantized Landau levels at finite field. All specific hopping parameters play a significant role in the Landau level structures. In bilayer Bernal graphene, $\gamma_1$ results in the nearly linear field dependence and the appearance of the second-group Landau levels. $\gamma_3$, $\gamma_4$ and $\gamma_6$ substantially modify the state energies and break the simple relation between energy and field strength. Moreover, $\gamma_4$ and $\gamma_6$ are also responsible for the electron–hole asymmetry and the field-modulated energy gap.

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3. Optical absorption spectra

The optical transitions between occupied and unoccupied states can be realized through the absorption of incident photons. Based on the Fermi golden rule, the optical spectrum at zero temperature is given by (Mahan 1990)

\[
A(\omega; B_0) \propto \sum_{n,n'} \left\{ \frac{\partial k_x \partial k_y}{(2\pi)^2} \right\} |(\psi_n^c(B_0; k_x, k_y)\frac{\hat{E} \cdot \hat{p}}{m_e} \psi_{n'}^v(B_0; k_x, k_y))|^2 \times \left\{ \frac{\Gamma}{[\omega - \omega_{n,n'}(B_0; k_x, k_y)]^2 + \Gamma^2} - \frac{\Gamma}{[\omega + \omega_{n,n'}(B_0; k_x, k_y)]^2 + \Gamma^2} \right\},
\]

(3.1)

where \( \omega_{n,n'}(B_0; k_x, k_y) = E_n^c(B_0; k_x, k_y) - E_{n'}^v(B_0; k_x, k_y) \) is the energy difference between occupied and unoccupied states. The polarization of electromagnetic radiation is linear and parallel to the graphene plane. The gradient approximation is used to evaluate the velocity matrix element \( \hat{E} \cdot \hat{p}/m_e = v_H/v_k \), which is suitable for carbon-related nanosystems to interpret the interactions between the electrons and photons (Johnson & Dresselhaus 1973; Lin & Shung 1994; Chang et al. 2004). It is expressed as the first-order derivative of Hamiltonian matrix elements. Also, the wave functions of occupied and unoccupied states would play a dominant role in optical absorption rate. That is, how the wave function distributes over its sublattices would offer an important insight into the possible excitation channels.

The low-frequency optical spectra of monolayer graphene are illustrated in figure 5a. At zero field, the absorption rate monotonically increases with incident photon energy, shown as the bottom curve. With the presence of a magnetic field, the magneto-optical response is governed by transitions between Landau levels. They are uniform in intensity but not equally spaced. The optical transition rate is primarily determined by the velocity matrix elements, as well as the product of initial- and final-state wavefunctions, as depicted in equation (3.1). The dominant contributions of the former are owing to the terms of in-plane hopping \( \gamma_0 \). That is, the way the wave function expands in terms of its sublattices, \( A \) and \( B \), enables one to determine the selection rules between Landau levels. In accordance with the orthogonal relation of Hermite polynomials, transitions are permitted only if the two sublattices have the same node number. For individual Landau levels in monolayer graphene, the node number of \( A \) and \( B \) steadily differs by one; hence, the selection rule is concluded to be \( \Delta n = n^c - n^v = \pm 1 \). The \( i \)th absorption is recognized as transitions from \( n^v = i - 1 \) to \( n^c = i \) and \( n^v = i \) to \( n^c = i - 1 \). Each Landau wave function is contributed almost equally by \( A \)- and \( B \)-sublattices, which results in the equal intensity of these pronounced peaks.

The interlayer interactions in bilayers significantly influence the low-frequency absorption spectra, as shown in figure 5b. The zero-field spectrum is sketched as the bottom curve which is composed of four specific spectral structures. The black and red circles denote two progressive increases of spectral intensity that initiate from 0 and 0.76 eV (= 2\( \gamma_1 \)). They are attributed to the optical transitions between the same pairs of parabolic bands, while the former is for the first pair and the latter is for the second pair. Besides, two abrupt increases occur at 0.364 eV (= \( \gamma_1 - \gamma_6 \)) and 0.396 eV (= \( \gamma_1 + \gamma_6 \)) marked with the green and blue circles,
Figure 5. Optical absorption spectra of (a) monolayer and (b) bilayer graphene. From the bottom to the top, curves correspond to 0–60 T. The black, green, blue and red circles indicate the first three excitations of \( n^v_1 \rightarrow n^c_1 \), \( n^v_2 \rightarrow n^c_1 \), \( n^v_1 \rightarrow n^c_2 \) and \( n^v_2 \rightarrow n^c_2 \), respectively.

which have their origin in the excitations between different pairs. The splitting of the two critical energies originates in the electron–hole asymmetry. In the case of finite field, the two pairs of parabolic bands are quantized into two groups of Landau levels with occupied and unoccupied states, \( n_{1,2}^{c,v} \). Consequently, the optical transitions between two groups give rise to four kinds of absorption peaks: \( n^v_1 \rightarrow n^c_1 \) (black circles), \( n^v_2 \rightarrow n^c_2 \) (red circles), \( n^v_1 \rightarrow n^c_2 \) (green circles) and \( n^v_2 \rightarrow n^c_1 \) (blue circles).

The explicit wave functions provide an intuitive way to interpret the selection rules and absorption rate of the four kinds of excitations. In the bilayer Landau states, the node numbers between sublattices are \( A_1 : B_1 : A_2 : B_2 = n + 1 : n : n + 1 : n + 2 \); particularly, \( B_2 \) and \( A_{1,2} \) sites are, respectively, adopted to define the quantum indices of the first- and second-group Landau levels. With this in mind, the selection rules are concluded to be \( \Delta n = \pm 1 \) for transitions between the same groups, \( n^v_1 \rightarrow n^c_1 \) and \( n^v_2 \rightarrow n^c_2 \). However, those between different groups are \( \Delta n = 0, -2 \) for \( n^v_2 \rightarrow n^c_1 \) and \( \Delta n = 0, 2 \) for \( n^v_1 \rightarrow n^c_2 \). These distinct selection rules primarily originate from the definition of Landau levels; nevertheless, all transitions are still identical from the point of view of sublattices. Transitions are permitted mainly between the same-layered \( A \) and \( B \) sublattices with the same node number.

The peak intensity in bilayer spectra is evidently suppressed and these peaks are mutually non-uniform, which can also be explained by the wave function distribution. Due to the difference in atomic chemical environment, wave functions are strongly localized on the \( B_{1,2} \) or \( A_{1,2} \) sublattices rather than uniformly distributed over the four sublattices, which qualitatively results in the overall weaker intensity. Such localization feature is gradually removed for higher energy levels, which can adequately elucidate the regular variation of intensity with photon energy in each kind of absorption. The absorption rate is determined by the product of major \( B_{1,2} \) sites and minor \( A_{1,2} \) sites (or major \( A_{1,2} \) and
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Figure 6. Field evolution of absorption lines for (a) monolayer and (b) bilayer graphene. The first five lines represent each kind of absorption peak.

minor $B_{1,2}$). For transitions between the same groups (black and red circles), the progressive development of minor sites leads to intensity growth with photon energy. On the contrary, those between different groups (green and blue circles) are stronger in the beginning and drop off with increasing photon energy.

Most pronounced peaks in bilayers are paired together as a result of the asymmetric behaviour between occupied and unoccupied states. For the transitions between same grouped Landau levels, the lower and higher peaks in each pair are identified as $\Delta n = -1$ and $\Delta n = 1$ for $n_1^v \rightarrow n_1^c$, while they are $\Delta n = 1$ and $\Delta n = -1$ for $n_2^v \rightarrow n_2^c$. With regard to the transitions between different groups, they are $\Delta n = -2$ and $\Delta n = 0$ for $n_1^v \rightarrow n_2^c$ while $\Delta n = 0$ and $\Delta n = 2$ for $n_2^v \rightarrow n_2^c$. Because of the absence of $n_1^c = 0$ and $n_2^c = 1$ levels and the wave-function localization of $n_1^c = 1$ and $n_2^c = 0$ levels on $B_2$ sites, the first several peaks of each kind of transitions might lack one of the double peaks. For instance, the threshold absorption peak involves transition $n_1^v = 2 \rightarrow n_1^c = 1$ only. The appearance of double peaks implies electron–hole asymmetry, which arises from the interlayer atomic hoppings $\gamma_4$ and $\gamma_6$. The width of these double peaks becomes narrower in higher energy region or at weaker field. Whether this signature is observable in experiments also depends on the broadening parameter $\Gamma$, which might be influenced by the temperature, defect scattering and other many-body effects.

The field evolution of absorption lines is plotted in figure 6a,b for monolayer and bilayer graphene. Those of monolayers are approximated as $\sqrt{B_0}$, which can be inferred from the Dirac linear fermions at zero field. They eventually fall towards zero in the limit of low field. In contrast, the bilayer spectra appear to be

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denser and richer than monolayers. Each kind of absorption lines are nearly linear with field but gradually deviate from the simple field dependence in the higher energy region. This also corresponds to the zero-field subband features: two pairs of parabolic bands right at the $K$-point with a transfer into linear-like ones away from $K$. The spacings between absorption lines correlate to all atomic hoppings ($\gamma_0$, $\gamma_1$, $\gamma_3$, $\gamma_4$ and $\gamma_6$). The four kinds of inter-Landau-level transitions are, respectively, illustrated as black, green, blue and red dots for $n^v_1 \rightarrow n^c_1$, $n^v_2 \rightarrow n^c_1$, $n^v_1 \rightarrow n^c_2$ and $n^v_2 \rightarrow n^c_2$. In the low-field limit, the four kinds of absorption lines fall off to $0$, $0.364$ eV ($\gamma_1 - \gamma_6$), $0.396$ eV ($\gamma_1 + \gamma_6$) and $0.76$ eV ($\gamma_1$). The convergent frequencies provide a direct way to determine the hopping parameters $\gamma_1$ and $\gamma_6$ in optical measurements. The spectral calculations are reliable since they can go back to zero-field characteristics. The magneto-optical spectra can be directly measured through the use of cyclotron resonance; it might be better than scanning tunnelling spectroscopy for layered materials since it can probe into several-atom-thick layers.

On the basis of the Peierls tight-binding model, except for the accurate Landau levels, the explicit wave functions are also available for the calculation of optical spectra. By means of wave-function characteristics, we can simplify the huge Hamiltonian matrix to access the small field problem that is attainable in experiments. The distribution over the sublattices enables one to clearly define the level indices. Moreover, it also provides a direct way to conclude the complex selection rules and the non-uniform absorption rate in bilayer optical spectra. The numerical method developed in this work can be expanded to other graphene layers (multilayer graphene and bulk graphite) and other physical properties (electronic excitations and transport properties).

4. Conclusion

The interlayer interactions in bilayers remarkably alter the zero-field electronic structures and thus the quantized Landau levels and inter-Landau-level optical excitations. The transfer from linear bands into two pairs of parabolic bands leads to two groups of Landau levels that scale nearly linearly with field. The optical transitions between the two groups result in four kinds of prominent peaks where the electron–hole asymmetry is responsible for the double-peaked absorptions. In particular, a close inspection of wave function distributions allows us to resolve the specific selection rules and energy-dependent absorption rates of the four kinds of transitions. The spectral characteristics in this work would be beneficial in identifying the measurements of few-layer graphene sheets.

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References


