

Optical Properties of Graphene in Magnetic and Electric Fields

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Preface

This book intends to illustrate the development of the generalized tight-binding model in graphene systems. The contents cover the AA, AB, ABC, AAB and sliding configurations and discuss many factors affecting their magneto-electronic properties. The absorption spectra of multilayer graphene systems are investigated in detail under the various stacking configurations, and their competition and cooperation with the applied electric and magnetic field are also studied. The main spectral characteristics focus on the prominent structures, number, intensity and frequency of the absorption peaks. In chapter 2, we introduce the Peierls tight-binding model under electric and magnetic fields. The optical response, which is used to describe the spectral properties, is calculated based on Fermi's golden rule. We start with monolayer graphene in chapter 3, and subsequently discuss AA-stacked trilayer graphene. Symmetry-preserved multi-Dirac-cone structures are responsible for the optical absorption spectra. Intra-Dirac-cone excitations comprise the whole frequency region of the spectra. Chapters 4 and 5 are devoted to AB- and ABC-stacked trilayer graphene. The excitations of massless and massive Dirac quasi-particles dominate the former system, while the vertical transitions between any two inter- or intragroup Landau levels (LLs) are permitted for the latter. In particular, the phenomenon of LL anticrossings in the energy spectrum is discussed and explained using a clear physical picture, especially for the energy width of the sombrero-shaped subbands. This leads to the corresponding non-monotonic dependence of the spectral frequency and intensity on the magnetic field. In chapters 6 and 7, we discuss the optical response under the effects of broken inversion symmetry in AAB-stacked trilayer and sliding bilayer graphene. The low-symmetry stacking configurations trigger more significant LL anticrossings and cause distinct features for the K- and K'-splitting LLs.

Chapter 1

Introduction

Graphene is a 2D material made up of hexagonal carbon lattices [1, 2]. Since mono- and few-layer graphene sheets were first fabricated in 2004 [1, 2], low-dimensional graphene-related systems have been of great interest in experimental and theoretical studies. The stacking order of graphene sheets includes the essential sequences of AA [3, 4], AB [5–9], ABC [5, 8–11] stacking. While the AA stacking configuration has only been made artificially from intercalated graphite compounds, the AB and ABC configurations are common orders in natural graphite, with their respective estimated volume fractions of 80% and 14% [15, 16]. The remaining ~6% consists of haphazardly stacked graphene sheets in a turbostratic configuration [17–19]. Few-layer graphene with the desired specific stacking configuration can be exfoliated from highly orientated pyrolytic graphite [1, 2] as well as chemically and electrochemically reduced from graphene oxide [20–27]. Nevertheless, the chemical vapor deposition method has the advantage of producing large-scale, high-quality graphene sheets. Recently, a large area of graphene with high mobility and highly symmetric configurations, e.g. AA, AB and ABC, was found in chemical vapour deposition (CVD)-grown samples [28–40]. The improved quality is adequate for research experiments and industrial applications [41–49]. In addition, AAB stacking and intermediate bilayer configurations, with relative layer-layer shifts or twists, have also been found and are becoming interesting subjects [59–67]. Synthesized graphene is generally comprised of different kinds of domain due to the tiny difference between the total energies of the different configurations. Infrared spectroscopy, transmission electron microscopy (TEM) [3–5, 34], scanning tunneling microscopy (STM) and spectroscopy (STS) [6–9, 11, 35–37], angle-resolved photoemission spectroscopy (ARPES) [68–74], and Raman spectroscopy [10, 75, 76], can be used to visualize the stacking domains with nanometric resolution for cases of tri-, tetra- and pentalayer graphene with typical or intermediate configurations.

Monolayer graphene belongs to a gapless system which possesses extremely high carrier mobility of up to $15\,000\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ [1]. P R Wallace discovered that the low-energy spectrum around the corners K and K' in the Brillouin zone is described

by isotropically conical dispersions in the framework of the nearest-neighbor tight-binding model [77]. The conical band structure and the vertex points at K and K' are known as the Dirac cone and Dirac points, respectively. The linearly dispersed quasi-particles in graphene behave as relativistic Dirac fermions with an effective speed of light $\simeq c/300$ (c is the speed of light). Few-layer graphene with specific stacking configurations, e.g. AA [78–80], AB [81–93], ABC [93–99] and AAB [64], are predicted to display unique electronic energy dispersions. This new material holds great promise for the development of next-generation electronic and optoelectronic nanodevices [41–47, 50–56] because the electronic and optical properties can be flexibly tuned by the application of external fields [64–74] as well as changes to the geometric structures [75–114] and dopants [115–122]. The stacking configurations have been experimentally and theoretically verified to have great significance for the magneto-electronic and optical properties [64–67, 108, 124–189], shedding some light on other intriguing structure-specific physical properties, e.g. the Coulomb excitations and the quantum Hall transport properties [190–221]. The essential properties under previous investigations are rather different from those in other layered materials, conventional metals, semiconductors and topological insulators [222–235]; they are dominated by the interplay between the external fields and the specific geometric symmetries. In this paper, we thoroughly review the electronic and optical properties of multilayer graphene to explore its response under external fields with respect to the specific AA, AB, ABC, AAB and sliding configurations.

In AA-stacked graphene, all the graphene sheets are identically stacked along the \hat{z} direction [126, 153]. The low-energy band structure consists of several monolayer-like massless Dirac cones [78–80, 153], which are exactly located at the two valleys K and K', while the energies of the Dirac points shift away from the Fermi energy due to the vertically projected geometry. The absorption spectrum is, in general, a superposition of monolayer-like spectra, because only the vertical excitations of intra-Dirac-cones are permitted, regardless of the external electric and magnetic fields [153, 183]. However, the stacking effect gives rise to an optical gap for the even-layer configurations, which is a forbidden optical transition zone resulting from free carriers in the Dirac cones. The middle-frequency spectrum shows symmetric peaks as a result of the saddle-point excitation channels around the M point. Such peaks are also observed for other stacked graphene, as well as being a critical characteristic of sp^2 -bonding carbon systems. In a uniform perpendicular magnetic field $B = B_0\hat{z}$, the Landau level (LL) spectrum behaves in the same way as several groups of monolayer-like spectra [126, 153, 183]. Responsible for each Dirac cone, the quantized LLs of each group are characterized by a set of single quantum modes, and their energies can be described by a square root dependence on B_0 . Therefore, the magneto-absorption spectrum consists of several monolayer-like spectra with delta-function-like absorption peaks following the optical selection rule of $\Delta n = \pm 1$ [153, 183]. A forbidden transition region and a discontinuous threshold frequency would be revealed for all the bilayer-like intragroup LL transitions, as the Fermi level ($E_F = 0$) deviates from the Dirac point. Moreover, the preservation of the optical and electronic properties of the massless Dirac fermions indicate the unchanged spatial symmetry of the chemical environment under the external fields.

AB-stacked graphene is a fascinating material which has attracted intense attention due to the nature of its massless and/or massive Dirac fermions, depending on whether the numbers of their stacking layers are even or odd [82–93]. Of special interest is the electronic structure, which can be essentially characterized by the superposition of monolayer-like and bilayer-like structures. Optical excitation channels accessible to the monolayer-like and bilayer-like subbands construct the optical spectrum [81, 100, 154–157]. Under an electric field, bilayer graphene shows a sharp peak due to new transition channels within the energy gap (up to 100 meV) [102, 103, 107, 109], and for cases in which there are more than three layers, displays enhanced semimetal characteristics attributed to the significant overlap of the low-energy subbands [101, 109, 110, 114]. In particular, as a result of the nearly isotropic energy dispersions around the K point, quasi-1D asymmetric peaks are revealed for the vertical excitations between two constant energy loops. Electric-field-induced characteristic peaks are also observed in other stacking systems. On the other hand, a magnetic field gives rise to interesting Landau quantization phenomena in the electronic and optical properties of odd-layer symmetry-broken graphene and even-layer inversion-symmetry graphene [126, 128–131, 141, 142]. Massless and massive Dirac quasi-particles reveal single- and twin-peak structures, respectively, in the monolayer-like and bilayer-like spectra [65, 158]. The corresponding ratio of the peak intensities is estimated to be about 2:1. In the presence of an electric field, the magneto-excitation channels around K and K' are separated from each other due to LL splitting, so that the channel numbers become double those under zero electric field [126]. In addition, very few LLs with a special quantum mode relationship avoid crossing each other during the variation of the magnetic or electric field; these are characterized by the main quantum mode and the side modes under the hybridization of the LLs [126, 131, 149]. Extra optical selection rules are induced for such LLs with multi-quantum-modes in addition to the ordinary $\Delta n = \pm 1$. In certain regions of the absorption spectra, the effects of geometry breaking and the anticrossings of intergroup LLs would trigger some special peaks without the simple relationships between the intensity, frequency and field strength.

Few-layer ABC-stacked graphene is a semimetal [93–99]. The electronic structure is characterized by one pair of partially flat subbands near $E_F = 0$, and pairs of sombrero-shaped subbands near the energy of the vertical atomic interactions between the nearest-neighboring layers [93–99]. Contributed by the surface-localized states [97–99], the partially flat subbands give rise to a prominent peak in the density of states (DOS) and play an important role in the low-frequency optical spectrum [110, 112]. An electric field can separate the two partially flat bands and trigger a new kind of optical channel responsible for a gap transition [110, 112]. Under a magnetic field, the intergroup and intragroup excitation channels among well-behaved LLs comprise the main part of the absorption spectrum [146, 174–176], in which the absorption peaks show a twin-peak structure because the LL energy spectrum is asymmetric about $E_F = 0$ [175, 176]. Moreover, it is interesting to study the quantization effects of any two constant energy loops in the sombrero-shaped structure, where the quantized energies of the LLs are proportional or inversely proportional to the magnetic field strength [126, 146, 147]. This leads to many

anticrossings of intragroup LLs in the ABC stacking configuration. By varying the electric field strength, one can suppress or enhance the degree of LL anticrossings and change the components of the main mode and side modes, that is, one can induce and modulate optical channels for progressive LLs during the variation of the field strength. Furthermore, the absorption spectrum is no longer dominated by a twin-peak structure but rather by a double-peak structure under a sufficiently large electric field—a transition which is attributed to the spatial symmetry breaking of the LL subenvelope functions.

AAB-stacked graphene exhibits an extraordinary band structure compared to the AA, AB and ABC systems, mainly due to the low symmetry of its stacking geometry [64]. In the case of the trilayer system, AAB-stacked graphene is a narrow-gap semiconductor which presents a pair of subbands with strong oscillatory energy dispersions near $E_F = 0$. With increasing energy, pairs of sombrero-shaped and parabolic bands appear in substantial amounts. In the low-frequency region, rich optical excitation channels cause many absorption peaks, some of which show quasi-1D asymmetric divergence due to the vertical excitations between two constant energy loops. The LLs coming from the two valleys K and K' are obviously separate in the mirror symmetry broken system [64]. The results present certain important differences for the state degeneracy, spacing, quantum number and spatial distribution of the wavefunctions, as compared with the highly symmetric configurations. Furthermore, the low-symmetry geometry of the AAB configuration implies the ease of triggering complex patterns of LL anticrossings and the richness of the magneto-absorption spectra. At low energies, the oscillatory subbands can induce triple-degenerate Landau states under the condition of three constant energy contours. In the B_0 -dependent LL spectrum, there are many prominent multicrossings and anticrossings, i.e. the existence of both well-behaved and perturbed subenvelope functions, and sometimes abnormal ones resulting from severe LL hybridizations. Accordingly, intergroup and intragroup LL anticrossings are both shown for various regions, and they even coexist in a narrow region. The optical characteristics resulting from either the intragroup or intergroup inter-LL excitations are remarkably different for the K- and K'-valley splitting states. The main reason for this is the phase and amplitude of the LL wavefunctions.

An intermediate bilayer configuration is formed from AA to AB, and then to AA', as the two graphene sheets shift relative to each other in the armchair direction [66, 148]. The two Dirac cones transform into parabolic bands from AA to AB configurations, and for a further layer-layer shift, they tilt towards each other at a small angle, restoring the AA' configuration. It should be noted that the two Dirac points are located at different wave vectors and energies once the stacking configuration deviates from the AA configuration. Furthermore, an eye-shaped stateless region is formed near $E_F = 0$ as a result of the strong hybridization of Dirac cones. The dramatic transformation between the Dirac cone structure and the parabolic band structure leads to drastic changes to the optical response under external electric and magnetic fields. The layer-layer shift has a significant impact on the LLs in sliding bilayer systems. In addition to the well-behaved and perturbed LLs stemming from the massless and massive Dirac fermions in the highly symmetric AA and AB

configurations, some undefined LLs appear to be promising in the case of a significant mixture of LLs, as the geometry deviates from the typical configurations. However, there is an absence of any regular optical selection rule for such undefined LLs, because their wavefunctions display extremely irregular oscillations without a dominating quantum mode. On the other hand, when the AA configuration is built, the LLs become well-behaved again, while additional optical channels and selection rules are accessible for intragroup LL excitations within the tilted Dirac cones. In the case of different bilayer configurations, it would be worthwhile to investigate the considerable shift-induced absorption peaks, which appear in a wide range of anticrossing patterns in the field-dependent energy spectrum.

The electronic properties are studied by use of STM, STS [6–9, 11] and ARPES [10, 68–76]. The band structures based on the tight-binding model agree with the first principles calculations [64, 66, 77, 78, 80–84, 89, 90, 93, 94, 96, 148]. The isolated Dirac cones are clearly seen in the monolayer graphene [69, 71, 73, 74], and the stacking effects for the typical configurations, AA, AB and ABC, are also identified by the main characteristics of the band structures [78, 80, 82, 126]. The former two show multiple Dirac cone dispersions and the superposition of monolayer-like and bilayer-like dispersions respectively [8], while the latter is not analogous to the AA and AB configurations [68, 70]. On the other hand, the deformation of Dirac cones for the AAB- and sliding bilayer configurations [64, 66, 148] can be verified by using ARPES. The aforementioned electronic characteristics have also been confirmed in absorption [164, 166–168, 173, 174, 177], transmission [180, 181], reflection [136], Raman scattering and Rayleigh scattering spectroscopy [75, 76, 138, 139, 171, 172, 236–243]. The evidence of Dirac cones has been observed for monolayer graphene in terms of the zero threshold frequency and the linear dependence of the intensity on the frequency. The low-frequency spectral features have also been verified for the AB- and ABC-stacked samples on SiO₂/Si substrates. Furthermore, the results provide spectroscopic evidence of a tunable gap in biased AB-stacked bilayer and ABC-stacked trilayer graphene through the observation of new gap-induced absorption peaks, with a blue or red shift at different gate voltages [109, 110]. The gap size is compatible with the theoretical calculations based on the tight-binding model and *ab initio* theory [100–106]. Transport and STS measurements also give the modified electronic properties under the electric field, including the enhanced semimetal properties of AB-stacked trilayer graphene [108, 112–114]. The electric-field-induced excitations rely on the geometric structure; that is to say, verifications of the spectral structure would be worthwhile in further research to identify the interlayer atomic interactions and geometric structures.

The quantized LL energies directly correspond to the prominent DOS peaks measured in STS, where the LLs near $E_F = 0$ have been identified for monolayer, AB-stacked bilayer and trilayer as well as ABC-stacked trilayer graphene [244–254]. Quantum Hall transport measurements also observe the sequence of the LLs [203–220]. Furthermore, low-energy optical transitions with specified selection rules are verified for the intergroup LLs near $E_F = 0$ in the infrared transmission, absorption and magneto-Raman spectra [75, 76, 136, 138, 139, 164, 166–168, 171–174,

177, 180, 181, 236–243]. The spectral features of the equivalent monolayer-like and bilayer-like Landau quantization are, respectively, identified by the square root and linear dependence of the energies on the magnetic field strength in the infrared transmission of AB-stacked trilayer graphene and in the magneto-Raman spectroscopy of AB-stacked graphene up to five layers [171]. For ABC-stacked trilayer graphene, while quantum Hall effect (QHE) transport measurements show the low-lying LLs [217–219], experimental evidence on the inter-LL excitations—distinct from the massless and massive Dirac fermion excitations—is needed for further verification. The crossings and anticrossings of LLs enrich the essential magneto-electronic and optical properties [64–66, 126, 131, 135, 146, 147, 175]. The former has been confirmed by the shift of plateaus in QHE measurements due to the increase in the LL degeneracy [214–220]. Regarding the latter, one can use optical spectroscopy to identify the non-monotonic changes in the structures, frequencies, intensities and numbers of the absorption peaks [175]. This phenomenon is expected to be more significant in low-symmetry AAB-stacked and sliding bilayer graphene [65, 66]. In sufficiently low magnetic fields, the spectral features reflect the band structures, such as the symmetry, energy dispersions and atomic interactions, as well as the broken mirror symmetry under the electric field. These experimental results can be used to identify the stacking configurations and the tight-binding parameters.

In this work, the generalized tight-binding model is developed to investigate optical properties of graphene in magnetic and electric fields. The $2p_z$ -dominated Hamiltonian is built from the subspace spanned by a lot of tight-binding functions in an enlarged unit cell. Its dimension is determined by the commensurate period of the geometric structure and external field. This model can be utilized in cases where many kinds of external fields are applied, e.g. uniform/modulated magnetic fields [124, 125, 150], modulated electric fields and composite fields [151]. This is suitable for multi-layer graphene [126, 128, 143, 144] and bulk graphite [255, 256] with arbitrary stacking configurations. The most important interlayer atomic interactions and external fields are simultaneously taken into account without the need to treat either of them as a perturbation term. The Hamiltonian matrix for determining the magneto-electric properties becomes too large for the experimental field strengths. By using an exact diagonalization method, the eigenvalues and eigenfunctions of the Hamiltonian matrix are efficiently solved. The developed model can be further used to understand other essential material properties by combining the single- and many-particle theories, such as optical spectra and electronic excitations [158, 159, 162, 175, 176, 190–196]. On the other hand, the effective mass approximation is frequently adopted to comprehend the electronic and optical properties. The low-lying energy bands are expanded about the high-symmetry point, and then magnetic quantization is done for the approximate properties. The magneto-optical absorption spectra are calculated for the AA- [127] and AB-stacked graphene [165]. Detailed comparisons of these two methods will be made in later discussions.

This book broadly covers the fields related to multilayer graphene systems in the forms of AA, AB, ABC, AAB and sliding configurations, and discusses many of the factors which affect their magneto-electronic properties. The absorption spectra of multilayer graphene systems are discussed in detail under various stacking

configurations, and their competition and cooperation with the applied electric field and magnetic field are also discussed. The main spectral characteristics focus on the prominent structures, number, intensity and frequency of absorption peaks. In chapter 2, we introduce the Peierls tight-binding model under electric and magnetic fields. The optical response, used to describe the spectral properties, is calculated based on Fermi's golden rule. We start with monolayer graphene in chapter 3, and subsequently discuss AA-stacked trilayer graphene. Symmetry-preserved multi-Dirac-cone structures are responsible for the optical absorption spectra. Intra-Dirac-cone excitations comprise the whole frequency region of the spectra. Chapters 4 and 5 are devoted to AB- and ABC-stacked trilayer graphene. The excitations of the massless and massive Dirac quasi-particles dominate the former system, while the vertical transitions between any two inter- or intragroup LLs are permitted for the latter. In particular, the phenomenon of LL anticrossings in the energy spectrum is discussed and explained using a clear physical picture, especially for the energy width of the sombrero-shaped subbands. This leads to the corresponding non-monotonic dependence of the spectral frequency and intensity on the magnetic field. In chapters 6 and 7, we discuss the optical response under the effects of broken inversion symmetry in AAB-stacked trilayer and sliding bilayer graphene. The low-symmetry stacking configurations trigger more significant LL anticrossings and cause distinct features for the K- and K'-splitting LLs.

At the beginning of each chapter, we present the Peierls Hamiltonian and the spectral function with respect to each system. We focus on the mechanism of the optical excitations responsible for the spectral characteristics based on the LL spectra and wavefunctions. The electric-field-enriched optical properties are also discussed. Optical absorption experiments are available for the investigation of the layer and stacking dependence of the magneto-electronic properties. Meanwhile, a detailed comparison of these systems is also presented in chapter 7. Finally, chapter 8 contains the concluding remarks.

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