Optical transitions between Landau levels: AA-stacked bilayer graphene

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Abstract

The low-frequency optical excitations of AA-stacked bilayer graphene are investigated by the tight-binding model. Two groups of asymmetric LLs lead to two kinds of absorption peaks resulting from only intragroup excitations. Each absorption peak obeys a single selection rule similar to that of monolayer graphene. The excitation channel of each peak is changed as the field strength approaches a critical strength. This alteration of the excitation channel is strongly related to the setting of the Fermi level. The peculiar optical properties can be attributed to the characteristics of the LL wave functions of the two LL groups. A detailed comparison of optical properties between AA-stacked and AB-stacked bilayer graphenes is also offered. The compared results demonstrate that the optical properties are strongly dominated by the stacking symmetry. These optical properties could be verified by optical measurements.
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1 Introduction

Few-layer graphenes (FLGs) are very exotic nanomaterials owing to their nanoscale inter-layer distance, hexagonal symmetry, and stacking configurations. FLGs have attracted numerous investigations on band structures,\textsuperscript{1−8} optical spectra,\textsuperscript{1,9−17} electronic excitations,\textsuperscript{5,18,19} and transport properties.\textsuperscript{20,21} The presented properties seem to make FLG-based materials excellent candidates for application in electronic and photonic devices. Since the physical properties of FLGs are strongly affected by the stacking configurations, FLGs with different stacking configurations have attracted considerable experimental and theoretical research. Monolayer (MG), AA-stacked bilayer (AABG) and AB-stacked bilayer (BBG; bilayer Bernal) graphenes are three prototypical FLGs. For the low-lying energy dispersions, MG exhibits isotropic linear bands near the Fermi level ($E_F = 0$); these bands become gradually anisotropic parabolic bands as the energy exceeds the region of $\pm0.5$ eV.\textsuperscript{2} In AABG, the linear bands in MG change into two pairs of linear subbands with slightly different slopes.\textsuperscript{5} These two pairs cross at $E_F = 0$ and are asymmetric about $E_F = 0$. For BBG, two pairs of parabolic subbands exist that are asymmetric about $E_F = 0$.\textsuperscript{8,14} The conduction and valence bands of the first pair slightly overlap about $E_F = 0$.

In the presence of a uniform perpendicular magnetic field $\mathbf{B} = B_0 \hat{z}$, the zero-field energy bands of FLGs become the dispersionless Landau levels (LLs).\textsuperscript{2−4,6−8} In MG, the low-lying LLs are characterized by the special relation $E_{n}^{c,v} \propto \sqrt{n^{c,v}B_0}$, where $n^{c} (n^{v})$ is the quantum number of the conduction (valence) LLs. This special relation is broken as the energy exceeds the region of $\pm0.5$ eV since the linear dispersions become gradually parabolic as the energy exceeds this region.\textsuperscript{2} For AABG and BBG, two groups of asymmetric LLs exist.\textsuperscript{8,14} These LLs are distributed away from a certain energy value in each group. The LL energies of both bilayer graphenes do not exhibit a simple relation similar to that of MG. These main features of FLGs would be reflected in the magneto-optical absorption
spectra. MG and BBG display different optical properties, e.g., different field-dependent absorption frequencies and distinct optical selection rules.$^{3,14}$

In this work, the magneto-optical absorption spectra of AABG are calculated by gradient approximation$^{1,11,14,22}$ within the tight-binding model (TB). Two groups of LLs are divided based on the characteristics of the LL wave functions. The LL wave functions are clearly depicted and utilized to explain the main features of the optical absorption spectra. The results show that two kinds of absorption peaks exist in the absorption spectra. Each peak obeys a single selection rule. The excitation channel associated with each peak varies with a changing field strength. The optical properties can be reasonably comprehended by the LL spectra and the characteristics of the LL wave functions. A detailed comparison between AABG and BBG reveals that they possess different magneto-optical absorption spectra, reflecting the influences of stacking configurations on the electronic properties. In other words, this method offers another way to distinguish AABG from BBG in addition to the STM images. Moreover, it may also be used to discriminate AABG from MG, which can be hardly done by STM.$^{23}$

## 2 Landau level spectrum

The geometric configuration of AABG is shown in Fig. 1(a). The primitive unit cell consists of four sublattices, $A_1$, $B_1$, $A_2$, and $B_2$. The subscripts 1 and 2 are, respectively, the indices of the first and second layer. Three atomic hopping integrals,$^{24}$ $\alpha_0 (=2.569$ eV), $\alpha_1 (=0.361$ eV), and $\alpha_3 (=0.032$ eV), are taken into account in this work. The first integral is the nearest-neighbor hopping integral on the same layer and the second and third are the interlayer interactions, as indicated in Fig. 1(a). $\mathbf{B}$ induces a periodic Peierls phase related to the vector potential $\mathbf{A}(\mathbf{r}) = (0, B_0 x, 0)$. The Hamiltonian under a magnetic field is

$$H_B = \left( \mathbf{P} - e \mathbf{A}(\mathbf{r})/c \right)^2/2m + V(\mathbf{r}),$$

where $m$, $\mathbf{P} (= \hbar \mathbf{k})$, and $V(\mathbf{r})$ are the electron mass, the
crystal momentum, and the lattice potential, respectively. Under the periodic condition, the primitive unit cell is enlarged\(^2\) and composed of four effective sublattices (denoted \(A_1, B_1, A_2,\) and \(B_2\) for convenience, similar to the symbols of the zero-field wave functions) including \(2R_B \ A_1, 2R_B \ B_1, 2R_B \ A_2,\) and \(2R_B \ B_2\) atoms, respectively. \(R_B\) is defined by 
\[R_B \equiv \Phi_0 / (\frac{3\sqrt{3}}{2}B_0 b^2) = \frac{79000 T}{B_0},\]
where \(\Phi_0\) is the flux quantum and \(b\) is the C-C bond length. \(R_B\) is inversely proportional to \(B_0\) and related to the dimension of \(H_B;^2\) for example, \(R_B\) is 1975 for \(B_0 = 40 T.\) That is to say, each LL wave function is the linear combination of the four magnetic TB functions associated with the four effective sublattices. The Hamiltonian matrix elements in the presence of a magnetic field are
\[
\langle R_{i,M} | H_B | R_{i',M'} \rangle = \gamma_s(R_{i,M}, R_{i',M'}) \sum_{1}^{N} \exp[i \mathbf{k} \cdot (R_{i',M'} - R_{i,M})] e^{i\mathbf{\bar{h}} \int_{0}^{1} (R_{i',M'} - R_{i,M}) \cdot A[R_{\xi',M'} + \lambda(R_{\xi',M'} - R_{i,M})] d\lambda], \tag{1}
\]
where \(R_{i,M}\) is the position vector of \(A_{i,M}\) or \(B_{i,M}.\) \(\gamma_s(R_{i,M}, R_{i',j'})\) indicate the atomic interactions between the atoms at \(R_{i,M}\) and \(R_{i',M'},\) i.e., they are \(\alpha_0, \alpha_1,\) and \(\alpha_3\) in this work. The representation \(A_{i,M} (B_{i,M})\) indicates the \(M\)th \((M = 1, 2...2R_B)\) \(A (B)\) atom on the \(i\)th \((i = 1, 2)\) layer.

The first Brillouin zone of AABG is shown in Fig. 1(b). For the low-energy electronic structure of AABG, the linear subbands of MG change into two pairs of linear subbands owing to the AA-stacking configuration and interlayer interactions, as shown in Fig. 1(c). The analytical solution of the energy dispersions of the first (second) pair can be described as
\[
E^{c,v} = -\alpha_1 \pm \frac{3}{2} (\alpha_0 - \alpha_3) bk + \frac{\alpha_1 \alpha_3}{\alpha_0} (E^{c,v} = \alpha_1 \pm \frac{3}{2} (\alpha_0 + \alpha_3) bk + \frac{\alpha_1 \alpha_3}{\alpha_0}),\tag{5}
\]
where \(E^c\) and \(E^v\) are respectively associated with the conduction and valence bands and are crossing at \(-\alpha_1 + \frac{\alpha_1 \alpha_3}{\alpha_0} \simeq -0.366 eV (\alpha_1 + \frac{\alpha_1 \alpha_3}{\alpha_0} \simeq 0.357 eV),\) as indicated by the black (red) lines. \(k\) is the wave vector measured from the \(K\) point. The conduction and valence bands of each pair are symmetric about the crossing energy. Furthermore, the two pairs of subbands intersect at \(E_F = 0\) and their slopes are slightly different. This causes the occupied and unoccupied
states to be asymmetric about \( E_F = 0 \). For the wave functions of zero-field subbands (not shown), the first and second pairs show the special relations, \( A_1/A_2 = B_1/B_2 = -1 \) and \( A_1/A_2 = B_1/B_2 = 1 \), respectively. These relationships mainly result from the nearest-neighbor interlayer interaction \( \alpha_1 \), which leads to the separation of the first pair from the second pair. These main features of the zero-field subbands would be reflected in a LL spectrum.

The magnetic field quantizes the two pairs of zero-field linear bands into fourfold degenerate LLs, as shown in Fig. 2(a) for 40 T. Based on the characteristics of the wave functions (Fig. 2(b)) discussed below, these LLs can be further divided into two groups of LLs, \( 1^{st} \)LLs and \( 2^{nd} \)LLs. The first (second) group corresponds to the first (second) pair of zero-field subbands and is distributed away from the onset energy -0.366 eV (0.357 eV), as shown in Fig. 2(a) by the black (red) lines. The onset energy of the first (second) group is located at the crossing energy of the first (second) pair of zero-field subbands. The LLs of each group are symmetric about the onset energy of this group. However, the occupied and unoccupied LLs are asymmetric about \( E_F = 0 \). The LL energies of the two groups can be written as

\[
E_{n_1,c,v} \simeq -\alpha_1 + \frac{\alpha_1 \alpha_3}{\alpha_0} \pm \frac{3}{2} (\alpha_0 - \alpha_3) b \sqrt{2 e B_0 n_1^{c,v,eff} / \hbar} 
\]

(2a)

\[
E_{n_2,c,v} \simeq \alpha_1 + \frac{\alpha_1 \alpha_3}{\alpha_0} \pm \frac{3}{2} (\alpha_0 + \alpha_3) b \sqrt{2 e B_0 n_2^{c,v,eff} / \hbar}, \quad (2b)
\]

where \( n_1^{c,v,eff} \) and \( n_2^{c,v,eff} \) are effective quantum numbers of the LLs in the first and second groups and defined below. The low-lying LL energies in each group linearly depend on \( \sqrt{B} \) similar to the relationship of MG.\(^{2,3}\)

The four degenerate wave functions of a LL are similar and thus only one of them is discussed here. Each LL wave function of AABG is linearly combined by four TB functions associated with the four effective sublattices. These TB functions display oscillatory modes and localized features. The four TB functions localized at a certain position are chosen
Through appropriate fitting, the wave functions of the \( n \)th 1st LL (2nd LL) can be expressed as \( A_1 = -A_2 \propto \varphi_{n-1}(x) \) and \( B_1 = -B_2 \propto \varphi_{n-2}(x) \) (\( A_1 = A_2 \propto \varphi_{n-1}(x) \) and \( B_1 = B_2 \propto \varphi_{n-2}(x) \)). \( \varphi_n(x) \) is the product of the \( n \)th-order Hermite polynomial and Gaussian function,\(^3\)\(^,\)\(^8\)\(^,\)\(^11\)\(^,\)\(^14\) where \( n \) is the number of zero points of \( \varphi_n(x) \) and chosen to define the quantum number of a LL.\(^{ref}\) For convenience, the zero-point numbers of the \( A \) atoms are chosen as effective quantum numbers of the LLs. \( n_{eff}^1 \)'s and \( n_{eff}^2 \)'s are, respectively, the effective quantum numbers of the LLs in the first and second groups, as shown in Fig. 2(a). Thus, the effective quantum numbers of the \( n \)th conduction (valence) LLs in both groups are \( n-1 \), which are similar to those of the LL wave functions of MG.\(^3\) Furthermore, the LL wave functions in Fig. 2(b) show a special relationship similar to those of the zero-field wave functions, i.e., \( A_1/A_2 = B_1/B_2 = -1 \) and \( A_1/A_2 = B_1/B_2 = 1 \) for the first and second groups, respectively. These relations mainly originate from the specific stacking configuration and should strongly affect the optical properties, e.g., the optical-absorption peak structure and the optical selection rules. The LL wave functions in both groups exhibit features similar to those of MG, i.e., they display similar oscillation modes, localization features, and a combination of the \( A \) and \( B \) atoms in each layer. Obviously, the characteristics of the LLs reflect those of the zero-field subbands, i.e., the existence of the two LL groups, the specific onset energies of the two groups, the symmetric structure about the onset energy of each group, the asymmetry of the occupied and unoccupied LLs about \( E_F = 0 \), and the specific relations of the LL wave functions of the two groups.

3 Magneto-optical properties

The main features of the LL spectra would be reflected in optical excitations. At zero temperature, there exist only excitations from the occupied to the unoccupied states. Based
on Fermi’s golden rule, the optical absorption function is given by

\[ A(\omega) \propto \sum_{h,h'} \int_{1st BZ} \frac{dk}{(2\pi)^2} \langle \psi^{h'}(n_{1,2}^{h',\text{eff}}, k) | \hat{E} \cdot \mathbf{P} | \psi^{h}(n_{1,2}^{h,\text{eff}}, k) \rangle^2 \]

\[ \times \text{Im} \left\{ \int_{1st BZ} \frac{dk}{(2\pi)^2} \left( f\left( E_{h}^{h'}(k, n_{1,2}^{h',\text{eff}}) \right) - f\left( E_{h}^{h}(k, n_{1,2}^{h,\text{eff}}) \right) \right) \right\}. \]  

(3)

\[ \hat{E} \] is the unit vector of an electric polarization and \( \hat{E} \parallel \hat{x} \) is taken into account in this work. \( \Gamma (=1 \text{ meV}) \) is a broadening parameter and often affected by temperature and defect effects. \( h \) and \( h' \) represent the occupied and unoccupied states, respectively. \( \langle \psi^{h'}(n_{1,2}^{h',\text{eff}}, k) | \hat{E} \cdot \mathbf{P} / m_e | \psi^{h}(n_{1,2}^{h,\text{eff}}, k) \rangle \) is the velocity matrix element (denoted \( M^{h'h} \)) derived from the dipole transition and calculated by gradient approximation.\(^{11,14} \) Through detailed calculations, \( M^{h'h} \) is expressed as

\[ \sum_{i,j=1,2}^{2R_B} \sum_{M,M'}^{M_H} [A_i^{h',s} \times B_j^{h}] \nabla_k \langle A_{i,Mk} | H_E | B_{j,M'k} \rangle + h.c. \]  

(4)

Eq. (4) corresponds to the two hopping integrals \( \alpha_0 \) (the terms for \( i = j \)) and \( \alpha_3 \) (the terms for \( i \neq j \)), and the terms associated with \( \alpha_0 \) dominate the value of Eq. (4). For the sake of convenience, the dominant term \( A_i^{h',s} \times B_j^{h} \) is represented by \( M^{h'h}_{ii} (\alpha_0) \) in the following discussions.

The low-frequency optical absorption spectra for \( B_0 = 0, 20 \text{ T}, \) and \( 40 \text{ T}, \) are shown in Fig. 3(a). The zero-field spectrum does not show any peak structure and the absorption rate is zero as the energy is below 0.71 eV. The vanishing absorption rate corresponding to the intersubband excitations is owing to the special relations of the wave functions in the two subbands. The emergence frequencies of the intrasubband excitations associated with the first and second pairs are nearly 0.73 eV \( (\simeq 2\alpha_1 - 2\frac{\alpha_3}{\alpha_0}; \text{ indicated by the black dot}) \) and 0.71 eV \( (\simeq 2\alpha_1 + 2\frac{\alpha_3}{\alpha_0}; \text{ indicated by the red dot}) \), respectively. For \( B_0 = 20 \text{ T} \) and \( 40 \text{ T}, \) absorption peaks basically result from the excitations between two LLs in the same group, i.e., only intragroup excitations exist in the absorption spectrum. Each peak can clearly be identified. The excitation channels of the first (second) kind of absorption
peaks, \(n_1^{c,v,\text{eff}} \rightarrow n_1^{c,v,\text{eff}} (n_2^{c,v,\text{eff}} \rightarrow n_2^{c,v,\text{eff}})\), are indicated by black (red) dots in Fig. 3(a). \(n_1^{c,v,\text{eff}} \rightarrow n_1^{c,v,\text{eff}} (n_2^{c,v,\text{eff}} \rightarrow n_2^{c,v,\text{eff}})\) represents the excitation channel from the occupied 1st LL with \(n_1^{c,v,\text{eff}} (2\text{nd LL with } n_2^{c,v,\text{eff}})\) to the unoccupied 1st LL with \(n_1^{c,v,\text{eff}} (2\text{nd LL with } n_2^{c,v,\text{eff}})\). The mth absorption peak frequency of the first (second) kind is denoted \(\omega_{11}^m (\omega_{22}^m)\). For \(B_0 = 20 \text{ T (40 T)}\), \(\omega_{11}^1 \text{ and } \omega_{22}^1\) originate from \(7^c \rightarrow 8^c \text{ and } 8^v \rightarrow 7^v \text{ (3}^c \rightarrow 4^c \text{ and } 4^v \rightarrow 3^v)\) respectively, i.e., they originate in the excitations from the occupied conduction to the unoccupied conduction LLs and from the occupied valence to the unoccupied conduction LLs, respectively. \(\omega_{11}^1 \text{ and } \omega_{22}^1\) are merged owing to their almost identical frequencies and strongly affected by the setting of Fermi energy (the Fermi energy is set to \(E_F = 0\)). Except for \(\omega_{11}^1 \text{ and } \omega_{22}^1\), the other peaks, \(\omega_{11}^m \text{ and } \omega_{22}^m \text{ for } m \geq 2\), come from the excitations between the occupied valence and unoccupied conduction LLs. For \(B_0 = 20 \text{ (40) T}\), the excitations \(7^v \rightarrow 8^c \text{ and } 8^v \rightarrow 7^c \text{ (3}^v \rightarrow 4^c \text{ and } 4^v \rightarrow 3^c)\) result in \(\omega_{11}^2 \text{ and } \omega_{22}^2\) respectively. For 20 T, the other channels, \((m + 5)^v \rightarrow (m + 6)^c \text{ and } (m + 6)^v \rightarrow (m + 5)^c\) possess the same frequency in the first (second) group which result in \(\omega_{11}^m \text{ and } \omega_{22}^m\). However, \(\omega_{11}^m (\omega_{22}^m)\) for 40 T originates from \((m + 1)^v \rightarrow (m + 2)^c \text{ and } (m + 2)^v \rightarrow (m + 1)^c\) possessing the same frequency in the first (second) group. Obviously, the excitation channels of the mth peaks related to the different field strengths may be different. \(\omega_{11}^m \text{ and } \omega_{22}^m\) are also slightly different. The former is higher than the latter, and they form a pair-like structure. This reflects the asymmetric structure of the LLs. The vanishing peak structure in the low-frequency region below \(\omega_{22}^2\), except for \(\omega_{11}^1 \text{ and } \omega_{22}^1\), can be ascribed to the characteristics of the LL wave functions and is discussed below. Simply said, the magneto-optical absorption spectra reflect the main features of the zero-field spectrum, i.e., two kinds of absorption peaks exist and only intragroup excitations are allowed.

The optical selection rules for \(\omega_{11} \text{ and } \omega_{22}\) can be represented by \(|\Delta n_{11}| (= |n_1^{h,c,\text{eff}} - n_1^{h,c,\text{eff}}|) = |\Delta n_{22}| (= |n_2^{h,c,\text{eff}} - n_2^{h,c,\text{eff}}|) = 1\). These rules are similar to those of the LLs in MG. The
selection rules can be comprehended by the characteristics of the LL wave functions. In Eq. (3), \( M_{n}^{h/h}(\alpha_0) \) dominates the excitations of the absorption peaks. \( M_{n}^{h/h}(\alpha_0) = A_{i}^{h'*} \times B_{i}^{h} \) has non-zero values only when \( A_1 \) and \( B_1 \) \((A_2 \text{ and } B_2)\) own the same \( \varphi_n(x) \) because of the orthogonality of \( \varphi_n(x) \). Since \( A_1 \) \((A_2)\) in the \( n \)th LL and \( B_1 \) \((B_2)\) in the \( n + 1 \)th LL for both groups own a same \( \varphi_n(x) \), the selection rules \( |\Delta n_{11}| = |\Delta n_{22}| = 1 \) can be easily obtained. Furthermore, the disappearance of the intergroup excitations can be ascribed to the fact that the two products, \( A_1 \times B_1 \) and \( A_2 \times B_2 \), in the intergroup excitations cancel each other out due to the special relationship of the wave functions.

The field-dependent absorption frequencies associated with the first and other \( m \)th peaks are shown in Figs. 3(b) and 3(c), respectively. In Fig. 3(b), at \( B_0 = 60 \) T, the excitation channel of \( \omega_{11}^1 \,(\omega_{22}^1) \) comes from \( 2^c \to 3^c \,(3^v \to 2^v) \). With decreasing field strength, the frequencies of \( \omega_{11}^1 \) and \( \omega_{22}^1 \) approach each other and merge at a sufficiently small field strength. Furthermore, the excitation channels of the first peak are altered as the field strength decreases to a critical field strength. For example, the channel \( 2^e \to 3^e \,(3^v \to 2^v) \) changes into \( 3^c \to 4^c \,(4^v \to 3^v) \) and \( 4^c \to 5^c \,(5^v \to 4^v) \) at \( B_0 = 47 \) T and 35 T (indicated by two yellow lines), respectively. As \( B_0 \) is reduced, the excitation channels become those associated with the LLs possessing larger effective quantum numbers. The main reason for this is that the setting of the Fermi level strongly affects which LL is considered the highest occupied (lowest unoccupied) one. The discontinuity of field-dependent absorption frequencies can be used in optical experiments to determine the excitation channels. Moreover, the excitation frequency of each excitation channel in its existent region is proportional to \( B_0 \), a behavior similar to the excitation frequencies of MG. The other excitation channels shown in Fig. 3(c) show features similar to those in Fig. 3(b). The convergent frequencies of \( \omega_{11} \) and \( \omega_{22} \) at the weak field strength are approximately 0.73 eV and 0.71 eV and correspond to the two emergence frequencies in the zero-field absorption
spectrum, respectively. Since the absorption peak intensities related to the LLs are strong, optical measurements can reasonably determine the values of $\alpha_1$ and $\alpha_3$ through observing the convergent frequencies of absorption peaks.

In addition to AABG, the magneto-optical properties of BBG are also discussed in a previously published work. AABG and BBG show similar LL spectra, e.g., two groups of LLs and an asymmetric structure. However, these two prototypical bilayer graphenes display totally different optical properties. AABG exhibits two kinds of absorption peaks, and only intragroup excitations that follow a single optical selection rule take place. BBG produces four kinds of absorption peaks, and both intra- and inter-group excitations that follow complex optical selection rules take place. These optical properties can be comprehended by obtaining the characteristics of the LL wave functions. The differences between the optical absorption spectra of AABG and BBG imply that the optical properties can reflect the influences of different stacking configurations. The dissimilarities between these two graphenes are helpful for distinguishing AABG from BBG via optical measurements. Furthermore, both the magneto-optical properties of AABG and BBG reflect the main features of zero-field optical properties.

4 Conclusions

In summary, the TB calculations show that the LLs are asymmetric about $E_F = 0$ and can be divided into two groups based on the characteristics of the wave functions. These two groups lead to two kinds of optical-absorption peaks $\omega_{11}$’s and $\omega_{22}$’s associated with only intragroup excitations of the first and second groups, respectively. The absorption frequencies of $\omega_{11}^m$’s and $\omega_{22}^m$’s are slightly different and form pair-like structures, which originate from the asymmetry of LLs. The optical selection rules can be reasonably explained by the characteristics of the LL wave functions. The selection rules of the two kinds of peaks are
$\Delta n_{11} = \Delta n_{22} = \pm 1$ similar to that of MG. The similar selection rules of AABG and MG mainly originate from the resembling characteristics of their LL wave functions. Furthermore, each $\omega_{11}^m$'s ($\omega_{22}^m$'s) corresponds to different excitation channels within the different field strength region. The field-dependent absorption frequencies for each excitation channel are linearly dependent on $\sqrt{B_0}$, and resemble the absorption frequencies of MG. The convergent absorption frequencies at the weak field strength might be helpful and reliable in determining the interlayer atomic interactions $\alpha_1$ and $\alpha_3$. The different magneto-optical properties of AABG and BBG reflect the influences of different stacking configurations. The differences between the two prototypical bilayer graphenes can help experimental researchers discriminate AABG from BBG. The above-mentioned magneto-optical properties could be confirmed by magneto-absorption spectroscopy measurements.
5 Appendix

In the absence of external fields, the Hamiltonian is $H_0 = P^2/2m + V(r)$, where $m$, $P$ ($= \hbar k$), and $V(r)$ are the electron mass, the crystal momentum, and the lattice potential, respectively. In the presence of a uniform magnetic field $B = B_0\hat{z}$, the Hamiltonian is $H_B = (P - eA(r)/c)^2/2m + V(r)$, where $A(r) = (0, B_0x, 0)$ is the vector potential. A Peierls phase induced by the magnetic field leads to a periodic condition and thus cause the primitive unit cell to be enlarged, i.e., the primitive unit cell in the absence of external fields is composed of four atoms, $A_1$, $B_1$, $A_2$ and $B_2$, while the enlarged primitive unit cell in the presence of $B$ comprises $2R_B A_1$, $2R_B B_1$, $2R_B A_2$, and $2R_B B_2$ atoms. $R_B \equiv \Phi_0/(\frac{3\sqrt{3}}{2}B_0b^2) = \frac{79000}{B_0}$ is associated with the dimension of the Hamiltonian matrix. The Peierls is defined by

$$\Delta G = \int_0^1 (R_{i',j'} - R_{i,j}) \cdot A[R_{i',j'} + \lambda(R_{i',j'} - R_{i,j})]d\lambda, \quad (A.1)$$

where $R_{i,j}$ is the position vector of $A_{i,j}$ or $B_{i,j}$ for $i = 1, 2$ and $j = 1, 2, \ldots, 2R_B$. In the sequence of the bases: $|A_1^1\rangle$, $|A_2^1\rangle$, $|B_1^1\rangle$, $|B_2^1\rangle$; $|A_1^2\rangle$, $|A_2^2\rangle$, $|B_1^2\rangle$, $|B_2^2\rangle$; $\ldots$; $|A_1^{2R_B}\rangle$, $|A_2^{2R_B}\rangle$, $|B_1^{2R_B}\rangle$, $|B_2^{2R_B}\rangle$, the Hamiltonian matrix related to the enlarged unit cell is expressed by

$$H = \begin{pmatrix}
H_{s,j=1} & H_{a,j=2} & 0 & 0 & \cdots & 0 & H_d \\
H_{a,j=2} & H_{s,j=2} & H_{a,j=3} & 0 & \cdots & 0 & 0 \\
0 & H_{a,j=3} & H_{s,j=3} & H_{a,j=4} & \cdots & 0 & 0 \\
0 & 0 & H_{a,j=4} & H_{s,j=4} & \cdots & 0 & 0 \\
\vdots & \vdots & \ddots & \ddots & \ddots & \ddots & 0 \\
0 & 0 & 0 & H_{a,j=2R_B-1} & H_{s,j=2R_B-1} & H_{a,j=2R_B} & 0 \\
H_d & 0 & 0 & 0 & H_{a,j=2R_B} & H_{s,j=2R_B} & H_{s,j=2R_B} \\
\end{pmatrix}, \quad (A.2)$$

13
where

\[
H_{s,j} = \begin{pmatrix}
H_{A_1^j A_1^j} & H_{A_1^j A_2^j} & H_{A_1^j B_1^j} & H_{A_1^j B_2^j} \\
H_{A_2^j A_1^j} & H_{A_2^j A_2^j} & H_{A_2^j B_1^j} & H_{A_2^j B_2^j} \\
H_{B_1^j A_1^j} & H_{B_1^j A_2^j} & H_{B_1^j B_1^j} & H_{B_1^j B_2^j} \\
H_{B_2^j A_1^j} & H_{B_2^j A_2^j} & H_{B_2^j B_1^j} & H_{B_2^j B_2^j}
\end{pmatrix} = \begin{pmatrix}
0 & \beta_1 & \beta_0 t_{1,j}^* & \beta_3 t_{1,j}^* \\
\beta_1 & 0 & \beta_3 t_{1,j}^* & \beta_0 t_{1,j}^* \\
\beta_0 t_{1,j} & \beta_3 t_{1,j} & 0 & \beta_1 \\
\beta_3 t_{1,j} & \beta_0 t_{1,j} & \beta_1 & 0
\end{pmatrix},
\]

(A.3)

\[
H_{a,j} = \begin{pmatrix}
H_{A_{1-1}^0 A_1^0} & H_{A_{1-1}^0 A_2^0} & H_{A_{1-1}^0 B_1^0} & H_{A_{1-1}^0 B_2^0} \\
H_{A_{2-1}^0 A_1^0} & H_{A_{2-1}^0 A_2^0} & H_{A_{2-1}^0 B_1^0} & H_{A_{2-1}^0 B_2^0} \\
H_{B_{1-1}^0 A_1^0} & H_{B_{1-1}^0 A_2^0} & H_{B_{1-1}^0 B_1^0} & H_{B_{1-1}^0 B_2^0} \\
H_{B_{2-1}^0 A_1^0} & H_{B_{2-1}^0 A_2^0} & H_{B_{2-1}^0 B_1^0} & H_{B_{2-1}^0 B_2^0}
\end{pmatrix} = \begin{pmatrix}
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & \beta_0 q & \beta_3 q & 0 \\
0 & \beta_3 q & \beta_0 q & 0
\end{pmatrix},
\]

(A.4)

and

\[
H_d = \begin{pmatrix}
H_{A_1^0 A_{2R}^0} & H_{A_1^0 A_{2R}^i} & H_{A_1^0 B_{1R}^0} & H_{A_1^0 B_{2R}^0} \\
H_{A_1^0 A_{2R}^i} & H_{A_1^0 A_{2R}^i} & H_{A_1^0 B_{1R}^i} & H_{A_1^0 B_{2R}^i} \\
H_{B_1^0 A_{2R}^0} & H_{B_1^0 A_{2R}^i} & H_{B_1^0 B_{1R}^0} & H_{B_1^0 B_{2R}^0} \\
H_{B_1^0 A_{2R}^i} & H_{B_1^0 A_{2R}^i} & H_{B_1^0 B_{1R}^i} & H_{B_1^0 B_{2R}^i}
\end{pmatrix} = \begin{pmatrix}
0 & 0 & \beta_0 q^* & \beta_3 q^* \\
0 & 0 & \beta_3 q^* & \beta_0 q^* \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix},
\]

(A.5)

and

\[
t_{1,j} = \exp\{i[-k_x \frac{b}{2} - k_y \frac{\sqrt{3}b}{2} + \frac{\phi}{\phi_0} (j - 1 + \frac{1}{6})]\}
+ \exp\{i[-k_x \frac{b}{2} + k_y \frac{\sqrt{3}b}{2} - \frac{\phi}{\phi_0} (j - 1 + \frac{1}{6})]\},
\]

(A.6(a))

\[
q = \exp[ik_z b].
\]

(A.6(b))

The optical absorption rate is dominated by the velocity matrix \(M^{kh}\). Through the gradient approximation, the velocity matrix is simplified as the product of three matrices, the initial state (occupied state; \(\psi^h\)), final state (unoccupied state; \(\psi^{h'}\)), and the first-order differential of the Hamiltonian matrix element versus the wave vector \(k\) (\(\nabla_k H_{ij}\)). The last term corresponds to the direction of electric polarization, i.e., it is \(\partial H_{ij}/\partial k_x\) (\(\partial H_{ij}/\partial k_y\)) for polarization along the x-direction (y-direction). The elements in the third matrix are
non-zero only for \( H_{ij} \) associated with the hopping integrals, as are the velocity matrix elements. In other words, when the velocity matrix element does not vanish, the initial and final states in the product should be the two states in a hopping process corresponding to non-vanishing hopping integrals. In this work, the polarization is along the \( x \)-direction and \( M^{h' h} \) is expressed as

\[
M^{h' h} = \left\langle \psi^{h'} \left| \frac{\partial H}{\partial k_x} \right| \psi^h \right\rangle = \sum_{i,j=1,2} \left\langle \psi^{h'}_i \left| \sum_{M,M'=1}^{2R_B} \frac{\partial}{\partial k_x} \left( \psi^{h'}_i, M_K \right) \left| H_B \right| \psi^h_{j,M'} \right\rangle \psi^h_j \right\rangle + h.c., \quad (A.7)
\]

where \( \psi \) is the wave functions of the \( A \) or \( B \) atom. Eq. (A.7) is associated with the three hopping integrals \( \alpha_0, \alpha_1 \) and \( \alpha_3 \). The term related to \( \alpha_1 \) is vanishing since the relative differential value is zero. \( M^{h' h} \) can be simplified as

\[
\sum_{i=1,2} \left\langle A^h_i \left| \sum_{M=1}^{2R_B} \frac{\partial}{\partial k_x} \left( A_i, M_K \right) \left| H_B \right| B_i, M_K \right\rangle B^h_i \right\rangle + h.c. \\
+ \sum_{i=1,2; i\neq j} \left\langle A^h_i \left| \sum_{M,M'=1}^{2R_B} \frac{\partial H}{\partial k_x} \left( A_i, M_K \right) \left| H_B \right| B_{j,M'} \right\rangle B^h_j \right\rangle \delta_{M,M'\pm1} + h.c. \quad (A.8 (a))
\]

\[
= M^{h' h}_{ii}(\alpha_0) + M^{h' h}_{ij}(\alpha_3). \quad (A.8 (b))
\]

The first term, \( M^{h' h}_{ii}(\alpha_0) \), is associated with \( \alpha_0 \) and the second term, \( M^{h' h}_{ij}(\alpha_3) \), corresponds to \( \alpha_3 \); \( M^{h' h}_{ii}(\alpha_0) \) dominates the value of the velocity matrix.
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FIG. 1. (a) The geometric structure, (b) the first Brillouin zone, and (c) the low-lying sub-bands of AA-stacked graphene. $\alpha_0$ (2.598 eV) is the nearest-neighbor hopping integral and the two important interlayer interactions are $\alpha_1$ (0.361 eV) and $\alpha_3$ (−0.032 eV).

FIG. 2. (a) Landau levels and (b) Landau level wave functions of AA-stacked graphene at $B_0 = 40$ T. $n_{1,c,v}^{c,v}$s and $n_{2,c,v}^{c,v}$s are the effective quantum numbers of the first and second group of Landau levels, respectively.

FIG. 3. (a) The optical absorption spectra of AA-stacked graphene at $B_0 = 0$, 20, and 40 T. The field-dependent absorption frequencies associated with (a) the first and (b) other $m$th absorption peaks. The symbols $n \rightarrow n'$ describe the excitation channels of the absorption peaks.
(a) $A_2 \quad B_2$

$\alpha_1 \quad \alpha_3$

$\alpha_0$

$A_1 \quad B_1$

(b)

\[ \begin{array}{c}
\Gamma \\
K \\
M \\
\end{array} \]

\[\begin{array}{c}
\downarrow \quad k_y \\
\rightarrow \quad k_x \\
\end{array}\]

(c) $B_0=0$

\[\begin{array}{c}
\Gamma \\
K \\
M \\
\end{array} \]

$E_{\text{c,v}}$ (eV)

-0.6

-0.5

-0.4

-0.3

-0.2

-0.1

0.0

0.1

0.2

0.3

0.4

0.5

0.6

$E_F$
(a) $B_0 = 40 \, \text{T}$

$E^{c,v}$ (eV)

$k_x, k_y$

$\psi^{c,v}$ (arb. units)

$A_1 \quad B_1 \quad A_2 \quad B_2$
