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Absorption spectra of AA-stacked graphite

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Abstract. AA-stacked graphite shows strong anisotropy in geometric structures and velocity matrix elements. However, the absorption spectra are isotropic for the polarization vector on the graphene plane. The spectra exhibit one prominent plateau at middle energy and one shoulder structure at lower energy. These structures directly reflect the unique geometric and band structures and provide sufficient information for experimental fitting of the intralayer and interlayer atomic interactions. On the other hand, monolayer graphene shows a sharp absorption peak but no shoulder structure; AA-stacked bilayer graphene has two absorption peaks at middle energy and abruptly vanishes at lower energy. Furthermore, the isotropic features are expected to exist in other graphene-related systems. The calculated results and the predicted atomic interactions could be verified by optical measurements.

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1. Introduction

Two-dimensional (2D) monolayer graphene is made up of carbon atoms in a planar sheet with a hexagonal lattice. Due to such hexagonal symmetry, there exist linear bands intersecting at the Fermi level. The density of states (DOS) vanishes at \( E_F = 0 \). Consequently, monolayer graphene is a zero-gap semiconductor. As for stacked systems, a 3D graphite crystal is a stack of graphene layers along the graphene plane vector. The weak interlayer \( \pi \) bonding, as well as the strong intralayer \( sp^2 \) bonding, characterizes the electronic structures of layers. Because of the interlayer interactions, graphite is a semimetal. It can form simple hexagonal systems with AA stacking, orthorhombic systems with AB stacking and rhombohedral systems with ABC stacking \([1]–[5]\). In general, orthorhombic graphite may be the most stable material among them \([5]\). Meanwhile, the other two types also have unique structures resulting in some special physical properties, which have attracted many theoretical and experimental studies \([6]–[10]\).

Other graphene-related materials, such as carbon nanotubes \([11]\), nanographene ribbons \([12]–[17]\) and few-layer graphene \([18]–[21]\), have also drawn great attention since their discovery. As for the nanotube, it can be regarded as a monolayer graphene rolled into a cylinder. Depending on its diameter and chirality, a nanotube can be either a semiconductor or a metal \([22]–[28]\). Geometric structures dominate the electronic structures and hence the physical properties. There have been numerous theoretical and experimental studies of optical excitations \([29]–[33]\). The nanographene ribbon \([34]–[36]\) is a strip of a graphene layer, while the multiribbon \([36]–[38]\) is a stack of graphene strips. Electronic properties \([37]\), optical properties \([38]\) and magnetic properties \([39]\) of nanographene ribbons are greatly dependent on the edge effects, the number of ribbons and the stacking sequence. The edge effects have been observed by scanning tunneling microscopy \([40]–[42]\), scanning tunneling spectroscopy \([41, 43]\) and Raman spectra \([43]\). In terms of few-layer graphene \([18]–[21], [44, 45]\), a stack of graphene layers with thickness less than 50 nm \([18]\), this is a semimetal with slight overlap near the Fermi level between the conduction and valence bands. Theoretical approaches have been used in the investigation of magnetic properties \([46]\), the quantum Hall effect \([46, 47]\), phonon spectra \([47]\), electronic properties \([48, 49]\) and optical excitations \([50, 51]\).
AA-stacked graphene layers used to be produced from lithium intercalated graphite half a century ago [52]. Recently, AA-stacked graphite has been successfully synthesized from a high-density dc plasma in hydrogen–methane mixtures, in which diamond is used as the growth substrate [53]. The structure and thickness of AA-stacked graphite may be found by high-resolution transmission electron microscopy and electron diffraction [54]. Owing to advances in sample growth, the study of its essential physical properties has become more popular. In this work, we mainly study the optical excitations in AA-stacked graphite. The dependence on polarization directions is investigated in detail, and a comparison with related systems is also made.

2. Electronic properties

The tight-binding model is used to study the electronic properties of AA-stacked graphite, e.g. the energy dispersions and the DOS. Its unique geometric structure results in some special electronic properties, which help us to further understand this material.

2.1. Tight-binding Hamiltonian

The geometric structure of AA-stacked graphite is shown in figure 1(a). It is formed from periodically stacked monolayer graphene along the z-direction. The coordinates \([x, y]\) of carbon atoms in each layer are the same, and the interlayer distance is \(I_c = 3.35\ \text{Å}\). A primitive cell consists of two carbon atoms. The nearest-neighboring carbon atoms have the same chemical environment. The tight-binding \(\pi\)-band calculations are similar to those done for 2D monolayer graphene. The two tight-binding functions \([\Psi^A_k(r)\text{ and } \Psi^B_k(r)]\) are built from atomic \(2p_z\) orbitals \(\phi_z(r')\): 

\[
\Psi^A_k(r) = C \sum_{R_n} \exp[-i \mathbf{k} \cdot (\mathbf{R}_n + \tau_{A(B)})] \phi_z(r - \mathbf{R}_n - \tau_{A(B)}),
\]

where \(\mathbf{k}\) is the 3D wavevector, \(C\) is the normalization factor and \(\mathbf{R}_n\) is the lattice vector. \(\tau_A\) and \(\tau_B\), denoted by circles and dots in figure 1(a), define the positions of atoms \(A\) and \(B\) in a unit cell. The wave function is described by the linear combination of the two Bloch functions, 

\[
\Psi_k(r) = \lambda \Psi^A_k(r) + \lambda' \Psi^B_k(r).
\]

The Hamiltonian matrix in the subspace spanned by the tight-binding functions is

\[
H = \begin{pmatrix}
\alpha_1 \Gamma + \alpha_2 (\Gamma^2 - 2) & f(k_x, k_y)(\alpha_0 + \alpha_3 \Gamma) \\
\alpha_1 \Gamma + \alpha_2 (\Gamma^2 - 2) & 2 \alpha_2 (\alpha_0 + \alpha_3 \Gamma)
\end{pmatrix}.
\]

where 

\[
f(k_x, k_y) = \sum_{l=1}^2 \exp(i \mathbf{k} \cdot \mathbf{b}_l) = h + 2g \quad \text{and} \quad \mathbf{b}_l \text{ corresponds to the three nearest neighbors with the magnitude } |\mathbf{b}_l| = b = 1.42\ \text{Å}, \quad \text{where} \quad h = \exp(i b k_x) \quad \text{and} \quad g = \exp(-i b k_x/2) \cos(\sqrt{3} b k_x/2)
\]

\(\Gamma = 2 \cos(k_c I_c)\) and \(\mathbf{k} (k_x, k_y, k_z)\) is confined within the first Brillouin zone (BZ) (the inset of figure 1(b)). \(\alpha_0 = 2.569\ \text{eV}\) and \(\alpha_3 = -0.032\ \text{eV}\) \((\alpha_1 = 0.361\ \text{eV}\) and \(\alpha_2 = 0.013\ \text{eV}\), respectively, represent the interactions between two nearest-neighboring atoms (between two atoms of the same projection on the \(xy\) plane), from the same plane and two neighboring planes (from two neighboring and next-neighboring planes) (figure 1(a)) [5]. By diagonalizing the Hamiltonian, the energy dispersions and the Bloch functions are

\[
E_{\pm}(k_x, k_y, k_z) = \alpha_1 \Gamma + 2 \alpha_2 [\Gamma^2/2 - 1] \pm (\alpha_0 + \alpha_3 \Gamma) |f(k_x, k_y)|
\]

and

\[
\Psi_{\pm, \mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{2}} \begin{pmatrix}
\Psi^A_k(\mathbf{r}) \pm f^*(k_x, k_y) \Psi^B_k(\mathbf{r}) \\
|f(k_x, k_y)| \Psi^B_k(\mathbf{r})
\end{pmatrix}.
\]

Figure 1. (a) The geometric structure, (b) electronic structure and (c) DOS of AA-stacked graphite. The inset in (b) is the first BZ, and the red lines are for the case of monolayer graphene.

2.2. Energy dispersions

The $\pi$-electronic structure of monolayer graphene is simply reviewed (the red curves in figure 1(b)). The energy dispersions reduced from equation (2) at $\alpha_1 = \alpha_2 = \alpha_3 = 0$ are

$$E^{c,v}(\mathbf{k}) = \pm \alpha_0 |f(k_x, k_y)|$$

$$= \pm \alpha_0 \left| 1 + 4 \cos \left( \frac{3bk_x}{2} \right) \cos \left( \frac{\sqrt{3}bk_y}{2} \right) + 4 \cos^2 \left( \frac{\sqrt{3}bk_y}{2} \right) \right|^{1/2}.$$  \hspace{1cm} (4)

The first BZ is a hexagon, which can be considered as the projection of the first BZ of AA-stacked graphite on the $k_z = 0$ plane (the red hexagon in figure 1(b)). The conduction and the valence bands are symmetric about the Fermi energy ($E_F = 0$) along $K \to \Gamma \to M \to K$ in

the first BZ. The low-energy bands, with \(|E^{c,v}| \leq 0.2\alpha_0\), own two linear bands intersecting at the Fermi level. Such energy dispersions are isotropic near the K point and can be described by \(|E^{c,v}| = 3\alpha_0bk/2\). The Fermi momentum is located exactly at the K point \((k_F = k_F = [2\pi/(3b), 2\pi/(3\sqrt{3}b), 0])\). The middle energy dispersion along the \(\Gamma M (MK)\) direction has a local minimum (maximum) \(|E^{c,v}_{M}\) at the M point, which makes M a saddle point in the energy-wave-vector space. The energy spacing \(E^c_M = E^c_M - E^c_M = 2\alpha_0\) remains unchanged along each MM’ direction, inducing a special structure in the DOS. As for the high-energy dispersion, the maximum and minimum energies are ±3\(\alpha_0\) at the \(\Gamma\) point, and the energy spacing \(E^c_\Gamma = E^c_\Gamma - E^c_\Gamma = 6\alpha_0\) describes the \(\pi\)-band width. Different from monolayer graphene, the AA-stacked bilayer graphene owns four carbon atoms in a unit cell. There exist two sets of energy bands exhibiting band asymmetry about \(E_F = 0\). One of them is higher, while the other is lower, than the band structure of monolayer graphene. It shows two pairs of linear bands at low energy and an overlap \((\sim 2\alpha_1)\) between the conduction and valence bands at the K point (the inset of figure 3(c)).

The energy dispersions of AA-stacked graphite along \(K \rightarrow \Gamma \rightarrow M \rightarrow K (k_z = 0)\) in the first BZ are similar to those of monolayer graphene (figure 1(b)). However, owing to the interlayer interactions, there are several important differences. The band symmetry about \(E_F (=0.0208\text{ eV} \sim 0.0081\alpha_0)\) is broken. For \(k_z = 0\), the intersection of the linear bands at the K point shifts upwards to \(2(\alpha_1 + \alpha_2)\), and the state energies of the M and \(\Gamma\) points are \(E^c_{\Gamma} = 2(\alpha_1 + \alpha_2) \pm (\alpha_0 + 2\alpha_3)\) and \(E^c_{\Gamma} = 2(\alpha_1 + \alpha_2) \pm 3(\alpha_0 + 2\alpha_3)\). When \(k_z\) becomes \(\pi/I_z\), the \((K, M, \Gamma)\) points change into the \((H, L, A)\) points with the state energies \(E^c_H = -2(\alpha_1 - \alpha_2)\), \(E^c_L = -2(\alpha_1 - \alpha_2) \pm (\alpha_0 - 2\alpha_3)\) and \(E^c_A = -2(\alpha_1 - \alpha_2) \pm 3(\alpha_0 - 2\alpha_3)\). Furthermore, the energy spacing at the saddle points with the same \((k_z, \alpha_3)\) projection exhibits obvious variations, e.g. \(E^c_M - E^c_\Gamma = 8\alpha_3\). It is also noted that \(E^c[k_F(k_z = 0)] = 2[2(\alpha_1 + \alpha_2) - E_F] \sim E^c[k_F(k_z = \pi/I_z)] = 2[2E_F + 2(\alpha_1 - \alpha_2)] \sim 0.56\alpha_0\).

2.3. Density of states (DOS)

The DOS (defined in [9]) reflects the features of band structures. It is associated with the number of excitation channels and is useful in explaining the optical absorption spectra. The DOS of AA-stacked graphite is shown in figure 1(c). For most values of \(k_z\), the linear conduction and valence bands do not intersect with each other at \(E_F\). The intersection at the K (H) point is above (below) the Fermi level. There are holes and electrons near the K and H points, respectively, and the DOS is nonzero at \(\omega = 0\). The DOS shows asymmetry about \(\omega = 0 [\text{DOS}(\omega) \neq \text{DOS}(-\omega)]\), different from monolayer graphene. When \(\omega\) departs gradually from zero, the DOS grows quadratically and forms several special structures \((s_1, s_2, s_3 \text{ and } s_4)\). The line between \(s_1\) and \(s_2\) \((s_3 \text{ and } s_4)\) is linear, resulting from the saddle points, which change from M to L. Therefore, the state energy of \((s_1, s_2, s_3, s_4)\) corresponds to \((E^c_\Gamma, E^c_M, E^c_\Gamma, E^c_M)\). According to the selection rules for the optical transitions, the high DOS from these structures will make a great contribution to absorption spectra.

3. Optical excitations

The features of the \(\pi\)-electronic structure are directly reflected in the optical excitations. The optical absorption functions are evaluated for AA-stacked graphite, bilayer graphene and monolayer graphene. The cause of the special spectrum structure, the atomic interaction

obtained, the dependence on dimensionality and layer number, and the anisotropic properties are discussed in the following.

3.1. Absorption functions

At zero temperature, electrons in the presence of electromagnetic fields will be excited from the occupied valence band to the unoccupied conduction band. The optical absorption function of AA-stacked graphite is given by

\[ A(\omega) \propto \sum_{h, h'} \int_{1stBZ} \frac{d|k|}{(2\pi)^3} \left| \langle \Psi_k^f | \hat{E} \cdot \mathbf{P} | \Psi_k^i \rangle \right|^2 \text{Im} \left[ \frac{n_F(E^h(k)) - n_F(E^b(k))}{E^h(k) - E^b(k) - \omega - i\delta} \right], \tag{5} \]

where \( n_F(E(k)) \) is the Fermi–Dirac distribution function. \( h \) and \( h' \) represent the initial (final) state. \( \delta \) \((= 0.025\alpha_0)\) is the energy width due to various de-excitation mechanisms, and it is treated as a free parameter in the calculations. The unit vector of electric polarization is denoted by \( \hat{E} \). Due to the zero transferred momentum of the photon, the optical transitions must obey the optical selection rule \( \Delta k = 0 \). \( A(\omega) \) is determined by \( |M^{cv}|^2 = |\langle \Psi_k^f | \hat{E} \cdot \mathbf{P} / m_e | \Psi_k^i \rangle|^2 \) and the number of optical transitions. The polarization along the armchair and the zigzag directions \( \hat{E}\|\hat{x} \) and \( \hat{E}\|\hat{y} \) is taken into account. Analysis of these two directions gives sufficient information about polarization dependence.

3.2. Velocity matrix element

The square of the velocity matrix element \( |M^{cv}|^2 \) is proportional to the optical absorption intensity. It is evaluated within the gradient approximation. Similar approximations have also successfully explained the optical spectra of graphite, graphite intercalation compounds and carbon nanotubes. The velocity matrix elements for \( \hat{E}\|\hat{x} \) and \( \hat{E}\|\hat{y} \) are given by

\[
M_x^{cv}(k) = \left\langle \Psi_k^f | \frac{P_x}{m_e} | \Psi_k^i \right\rangle = \frac{1}{2} \left( \begin{array}{cc} 1 & \frac{f(k)}{|f(k)|} \end{array} \right) \begin{pmatrix} \frac{\partial H_{11}}{\partial k_x} & \frac{\partial H_{12}}{\partial k_x} \\ \frac{\partial H_{21}}{\partial k_x} & \frac{\partial H_{22}}{\partial k_x} \end{pmatrix} \begin{pmatrix} 1 \\ -f^*(k) \end{pmatrix} \\
\frac{1}{2} \left( \begin{array}{cc} 1 & \frac{f(k)}{|f(k)|} \end{array} \right) \begin{pmatrix} 0 & q(h-g) \\ q^*(h^*-g^*) & 0 \end{pmatrix} \begin{pmatrix} 1 \\ -f^*(k) \end{pmatrix} \\
-\text{i} \text{Im} \left[ \frac{q(h-g)}{|f(k)|} f^*(k) \right] \tag{6} \]

and

\[
M_y^{cv}(k) = \frac{1}{2} \left( \begin{array}{cc} 1 & \frac{f(k)}{|f(k)|} \end{array} \right) \begin{pmatrix} 0 & p g^* \\ p^* g & 0 \end{pmatrix} \begin{pmatrix} 1 \\ -f^*(k) \end{pmatrix} = -\text{i} \text{Im} \left[ \frac{p g f^*(k)}{|f(k)|} \right]. \tag{7} \]

where \( q = (\alpha_0 + \alpha_3 \beta) \gamma_0 b \) and \( p = -\sqrt{3}(\alpha_0 + \alpha_3 \beta) b \tan(\sqrt{3}b k_x) \). The contour plots of \( |M_x^{cv}|^2 \) and \( |M_y^{cv}|^2 \) are shown in figure 2 for the first BZ. They are symmetric about the \( \Gamma \) point, and the values are very small in most areas. At \( k_z = 0 \) (figures 2(a) and (b)), \( |M^{cv}|^2 \) vanishes near
Figure 2. Contour plots of $|M^{cv}|^2$ for the first BZ. Panels (a) and (b) are for the $k_z = 0$ case and (d) and (e) are for $k_z = \pi/2I_c$, with the polarization along the armchair and zigzag directions, respectively. Panel (c) is shown for $|M^{cv}|^2$ depicted along the energy contour line.

the K points, because the Fermi level forbids optical transition channels. The wavevector of the edges of the vanishing $|M^{cv}|^2$ corresponds to $k_F$. The greatest values are located at $M_3$ and its symmetry point for $\hat{E}||\hat{x}$ ($M_1$ and $M_2$ and their symmetry points for $\hat{E}||\hat{y}$) (figures 2(a) and (b)). There is no simple relationship between the cases of $\hat{E}||\hat{x}$ and $\hat{E}||\hat{y}$. However, the integrals of $|M^{cv}|^2$ over the area of the same energy are equal between the two cases. For example, the areas within the two curves along the energy contour line ($M_1 \rightarrow M_2 \rightarrow M_3$) equal each other (figure 2(c)), that is, $\int |M^{cv}|^2 \, dk_x \, dk_y$ is the same for the two cases when $\omega = E_M^x$, $|M^{cv}|^2$ of each $k_z$ shows similar results except for the edges of the vanishing $|M^{cv}|^2$. When $k_z = \pi/2I_c$ (figures 2(d) and (e)), these edges disappear because of the intersections of the linear bands falling on the Fermi level. The same value of $\int |M^{cv}|^2 \, dk_x \, dk_y$ is obtained for each $k_z$ at all photon frequencies between $\hat{E}||\hat{x}$ and $\hat{E}||\hat{y}$, causing the polarization isotropy in the absorption spectra. Such properties may exist in other graphene-related systems for the graphene plane’s hexagonal symmetry.
Both the lattice structure and the band structure near the M points of a graphene layer have the rotational symmetry with the period of $\pi/3$, and so do those of AA-stacked graphite. However, the symmetric period of $|M^{cv}|^2$ is $\pi$. The different symmetric periods indicate that the lattice structure and $|M^{cv}|^2$ have no direct relation in the symmetry. Furthermore, there are no translational and rotational symmetries in $|M^{cv}|^2$ between the two cases of $\hat{E}\parallel\hat{x}$ and $\hat{E}\parallel\hat{y}$. It is hard to conceive the consistency of $|M^{cv}|^2$ of the two cases. On the other hand, when the interlayer interactions and the $k_z$ dependence are neglected, the 3D AA-stacked graphite could be considered as a 2D monolayer graphene for calculations. Therefore, the comparison of $|M^{cv}|^2$ between the former with a given $k_z$ and the latter is meaningful. In Grüneis’s study [55],

Figure 3. Optical absorption spectra of (a) AA-stacked graphite, (b) monolayer graphene and (c) AA-stacked bilayer graphene. The inset in (c) is the low-energy band structure of AA-stacked bilayer graphene, and the allowed (disallowed) optical excitations are denoted by circles (crosses).
$|M^c|^2$ of monolayer graphene is multiplied by a delta function to discuss the contributions from the specific laser frequency. Here, $|M^c|^2$ is analyzed to understand the absorption spectra with continuously varying frequency, so the delta function is not needed.

3.3. Optical absorption spectra

$A(\omega)$, shown for AA-stacked graphite in figure 3(a), does not reflect the anisotropy of the geometric structures and the velocity matrix elements, whereas it exhibits independence of the polarization direction $\hat{E}$ on the graphene plane. A shoulder structure exists at lower energy and a prominent symmetric plateau is at middle energy. For any given $k_z$, the excitations are forbidden when half of their excitation energy is smaller than the energy ($E_{th}$) between the Fermi level and the intersection of the linear bands. The intersection moves downwards from $2(\alpha_1 + \alpha_2)$ to $-2(\alpha_1 - \alpha_2)$ as $k_z$ grows from zero to $\pi / I_c$, and thus the maximum of $2E_{th}$ is about $E[k_f(k_z = 0)] \sim E[k_f(k_z = \pi)] \sim 0.56\alpha_0$. Therefore, the low-frequency $A(\omega)$ declines significantly when $\omega < 0.56\alpha_0$, forming the shoulder structure. Concerning the plateau structure, the width of $8\alpha_3$ arises from the changing $E^s$ of the saddle points. The left boundary corresponds to $E^s_M [= 2(\alpha_0 + 2\alpha_3)]$ and the distance between $s_2$ and $s_4$ in the DOS, and the right one relates to $E^s_B [= 2(\alpha_0 - 2\alpha_3)]$ and the $s_1s_3$ distance. The position and width of the plateau structure is a function of $\alpha_0$ and $\alpha_3$, and the position of the shoulder structure and $E_F$ are associated with $\alpha_1$ and $\alpha_2$. Therefore, analysis of optical experiment data could be another way to obtain the intralayer ($\alpha_0$) and interlayer ($\alpha_1$, $\alpha_2$, and $\alpha_3$) atomic interactions.

The prominent structure near $2\alpha_0$ in the optical spectra results from the $\pi$-electronic excitations and is a general feature in the carbon-related materials with sp² binding. It can be found at $\sim 5-7$ eV in many systems with different forms. For example, an asymmetric absorption peak exists in the form of square-root divergence in both 1D carbon nanotubes [26] and graphene nanoribbons [38]. On the other hand, orthorhombic graphite with AB stacking has been investigated through optical measurements. One prominent peak is observed experimentally at $\omega \sim 2\alpha_0$, but no special structure at low energy [56]. Due to the much stronger interlayer interactions of AA-stacked graphite, the predicted plateau structure is even more obvious, and one shoulder structure is induced at low energy.

The layer number also affects the absorption spectra drastically. For monolayer graphene (figure 3(b)), the low-energy bands have an intersection at $E_F = 0$, and the state energy is exactly $\pm \alpha_0$ along the MM' direction. It leads to the fact that $A(\omega)$ grows linearly from zero with increasing frequency, and has a sharp absorption peak at $\omega = 2\alpha_0$. As for AA-stacked bilayer graphene, the layer number results in two sets of energy bands (the inset of figure 3(c)). There are two pairs of linear bands near $E_F = 0$, and the intersections of sets 1 and 2 (blue and red curves) show an energy spacing of about $2\alpha_1$. Furthermore, due to the symmetry of the wave function of atoms A and B in the 1st and 2nd layers ($\Psi_{k,1}^{A_{1st}} / \Psi_{k,1}^{A_{2nd}} = \Psi_{k,1}^{B_{1st}} / \Psi_{k,1}^{B_{2nd}} = 1$ and $\Psi_{k,2}^{A_{1st}} / \Psi_{k,2}^{A_{2nd}} = \Psi_{k,2}^{B_{1st}} / \Psi_{k,2}^{B_{2nd}} = -1$), only transitions between the occupied and unoccupied states in the same set are allowed, and they do not take place between the intersections of sets 1 and 2 (the inset of figure 3(c)). Accordingly, $A(\omega)$ of AA-stacked bilayer graphene has two absorption peaks coming from the $E^s_M$ of sets 1 and 2, but abruptly vanishes [57] when $\omega < 2\alpha_1$ (figure 3(c)). The two sets of linear bands will also cause two steps near $2\alpha_1$. However, their slopes are so close that these two steps are hard to distinguish from each other. The identical projection of each layer results in this special relationship of wave functions between the initial and the final states. Similarly, it may be expected that the peak number in the
absorption spectra equals the layer number of the few-layer AA-stacked graphene. Nevertheless, the wave functions of AB- and ABC-stacked graphenes have no such special relationship, and their absorption spectra are more complicated. In general, the intensity and number of peaks, as well as the low-frequency structure, are associated with the dimensionality and layer number. These special structures in the spectra could provide information about the geometric structure.

The same independence of the polarization direction has also been seen in the absorption spectra of both monolayer and AA-stacked bilayer graphenes (figures 3(b) and (c)). The velocity matrix element decides the polarization dependence. Each graphene layer in these systems has hexagonal symmetry and hence the integral of $|M^{cv}|^2$ of all frequencies presents isotropic property when the polarization vector lies on the graphene plane (figure 2). For a layer number greater than two, AA-stacked graphenes still show the same isotropic feature (not shown), which indicates that the polarization-direction independence of the absorption spectra might occur in other layered systems, such as AB-stacked few-layer graphene and graphite.

4. Concluding remarks

In this work, the tight-binding model and the gradient approximation are, respectively, used to study the band structures and the optical properties of AA-stacked graphite. The geometric structures and the velocity matrix elements exhibit strong anisotropy. Also, the band structures, except for those at low energy, are anisotropic. Nevertheless, the optical absorption spectra show isotropic features for the polarization vector on the graphene plane, because the integrals of the square of velocity matrix elements are equal for all frequencies between different polarization directions. There is one prominent plateau at middle energy and one shoulder structure at lower energy in the spectra. This plateau results from the saddle points with the changing energy spacing of $\sim 2\alpha_0$ ($M \rightarrow H$), and the shoulder is caused by the $k_z$-dependent intersection of the low-energy linear bands. The positions and width of those special structures are directly associated with the interlayer and intralayer atomic interactions. Experimental methods may be utilized to determine these atomic interactions. Moreover, for monolayer graphene, the spectra grow linearly from zero with an increase in frequency, and show a sharp peak at $2\alpha_0$. As for AA-stacked bilayer graphene, the spectra own two sharp peaks near $2\alpha_0$ and abruptly vanish at low frequency. Both the dimensionality and the layer number play an important role in the optical absorption spectra. These calculated results can be verified by optical measurements.

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