Research Article

Flake Electrical Conductivity of Few-Layer Graphene

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Received 24 August 2013; Accepted 20 October 2013; Published 16 January 2014

Academic Editors: K. Liu, S. K. Yang, and T. Zhou

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The Kubo formula for the electrical conductivity of per stratum of few-layer graphene, up to five, is analytically calculated in both simple and Bernal structures within the tight-binding Hamiltonian model and Green’s function technique, compared with the single-layer one. The results show that, by increasing the layers of the graphene as well as the interlayer hopping of the nonhybridized $p_z$ orbitals, this conductivity decreases. Although the change in its magnitude varies less as the layer number increases to beyond two, distinguishably, at low temperatures, it exhibits a small deviation from linear behavior. Moreover, the simple bilayer graphene represents more conductivity with respect to the Bernal case.

Graphene is an atom thick allotrope of carbon in two-dimensional (2D) hexagonal honeycomb lattice [1]. Electrons in a single-layer graphene exhibit a characteristic linear dispersion relation between energy and momentum near the $K$ point of the first Brillouin zone (FBZ) [2–4]. The overall electronic structure changes sensitively with increasing crystallographic stacking sequence. The sequence of graphene sheets brings about the various 3D graphite crystals [5–8], that is, AA-stacked bilayer graphene (hexagonal simple bilayer graphene), AB-stacked bilayer graphene (Bernal bilayer graphene), and ABC-stacked trilayer graphene (rhombohedral trilayer graphene). The interlayer interactions, due to the weak overlap of the nonhybridized $p_z$ orbitals, result in the anisotropic band structure along the stacking direction. Some theoretical studies [9–12] have predicted in two or more layers of graphene that the linearly dispersing bands are either replaced or augmented by split hyperbolic bands. Experimental investigations have also been considered to single- and bilayer graphene [13–15]. In a single-layer graphene transistor, the current is modulated by a gate voltage but it cannot be switched off due to lack of a band gap in the energy dispersion. Bilayer graphene is the only known semiconductor with a gate tunable band gap [16]. Opposed to the case of single- and bilayer, the trilayer material is a semimetal with a gate tunable band overlap between the conduction and the valence bands [16]. The variety of electronic properties found in different few-layer graphene (FLG) is the true strength of these newly discovered materials.

In this study, the electrical conductivity (EC) of FLG in {AA, AAA, AAAA, AAAAA, AB, ABA, ABAB, ABABA} structures is investigated within the tight-binding (TB) Hamiltonian model and Green’s function method. Using band representation of Green’s function, we calculate the EC of the systems by Kubo formula [17–19]. Then, the temperature-dependent EC of a monolayer graphene will be compared to that for per sheet of these systems, hereinafter mentioned as “flake EC” (FEC). In second quantization form, the Hamiltonian of the TB model for FLG lattice reads as follows [20]:

$$
\mathcal{H} = -\sum_{\alpha,\beta, i,j=1}^{N_c} \sum_{p,q=1}^{N_p} t_{ipjq} e^{\alpha\beta}_{ij} e^{\alpha\beta}_{jq},
$$

where $\alpha$ and $\beta$ refer to the $A$ or $B$ subsites inside the Bravais lattice unit cells (Figure 1) in each plane of the system, $i$ and $j$ denote the position of the Bravais unit cell in the lattice, $p$ and $q$ describe plane’s indexes, $N_c$ shows the number of the Bravais lattice unit cell, $N_p$ implies the number of the layers, $t_{ipjq}$ presents the amplitude for a $\pi$ electron to hop from the subsite $\alpha$ of the Bravais lattice site $i$ in plane $p$ to the subsite $\beta$ of the nearest-neighbor (NN) site $j$ in plane $q$, and $e^{\alpha\beta}_{ij} (e^{\beta\alpha}_{jq})$. 

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denoted by $\ell=\text{null}$ are utilized that the carbon atoms have been fitted as zero. Besides, such units zero corresponding to the contribution of one electron per lines illustrate the Bravais lattice unit cell. Each cell includes orbital in the system. We note that the on-site energy of the trilayers graphene, the Green function could be represented as

\[ G(i,j;\tau) = \{\Phi_i^\alpha, \Phi_j^\beta, \phi_i^\alpha, \phi_j^\beta, \phi_i^\beta, \phi_j^\alpha\}, \]

so the trilayer Green function could be represented as

\[
G(i,j;\tau) = \begin{pmatrix}
G_{11}^{AA} & G_{12}^{AB} & G_{13}^{BB} \\
G_{21}^{AA} & G_{22}^{AB} & G_{23}^{BB} \\
G_{31}^{AA} & G_{32}^{AB} & G_{33}^{BB}
\end{pmatrix}
\]

with $c_{\ell p}^{\alpha\beta}(i,j;\tau) = -\langle T c_{\ell p}^{\alpha}(\tau)c_{p}^{\beta}(0)\rangle$, in which $\tau = it$ remarks imaginary time and $T$ hints the time ordering operator. Here, $\langle \cdots \rangle$ exhibits ensemble averaging on the ground state of the system. Using Green's function formalism for the Hamiltonian in (1), the equation of motion for electrons in AAA structure can be written as

\[
\sum_{\ell} \begin{pmatrix}
E & t_{\ell1}^{AA} & 0 & t_{\ell1}^{AB} & 0 & 0 \\
0 & E & t_{\ell1}^{AB} & 0 & t_{\ell1}^{BB} & 0 \\
0 & 0 & E & t_{\ell1}^{BB} & 0 & 0 \\
0 & 0 & 0 & E & t_{\ell1}^{AB} & 0 \\
0 & 0 & 0 & 0 & E & t_{\ell1}^{AB} \\
0 & 0 & 0 & 0 & 0 & E
\end{pmatrix} G(\ell,j;E) = \text{I} \delta_{ij},
\]

and that for ABA case is given by

\[
\sum_{\ell} \begin{pmatrix}
E & 0 & 0 & t_{\ell1}^{AB} & 0 & 0 \\
0 & E & 0 & t_{\ell1}^{AB} & 0 & t_{\ell1}^{BB} \\
0 & 0 & E & 0 & 0 & t_{\ell1}^{AB} \\
0 & 0 & 0 & E & 0 & 0 \\
0 & 0 & 0 & 0 & E & 0 \\
0 & 0 & 0 & 0 & 0 & E
\end{pmatrix} G(\ell,j;E) = \text{I} \delta_{ij},
\]

where $E = \varepsilon + \delta$, the index $\langle \cdots \rangle$ shows NN sites, $I$ plays the role of a $6 \times 6$ unit matrix, and $\delta_{ij}$ notifies the Kronecker symbol. The $k$-space Fourier transformation of (4) and (5) leads to the following relations:

\[
G(k;E) = \begin{pmatrix}
E & t_{11} & 0 & \epsilon_k & 0 & 0 \\
t_{11} & \tilde{E} & t_{11} & 0 & \epsilon_k & 0 \\
0 & t_{11} & E & 0 & 0 & \epsilon_k \\
0 & \epsilon_k & 0 & E & t_{11} & \tilde{E} \\
0 & 0 & \epsilon_k & 0 & t_{11} & \tilde{E} \\
0 & 0 & 0 & \epsilon_k & 0 & E
\end{pmatrix}^{-1},
\]

\[
G(k;E) = \begin{pmatrix}
E & 0 & 0 & \epsilon_k & 0 & 0 \\
0 & E & 0 & t_{11} & \epsilon_k & 0 \\
0 & 0 & E & 0 & 0 & \epsilon_k \\
0 & \epsilon_k & 0 & E & 0 & 0 \\
0 & 0 & \epsilon_k & 0 & E & 0 \\
0 & 0 & 0 & \epsilon_k & 0 & E
\end{pmatrix}^{-1},
\]

in which $\epsilon_k$ is defined as

\[
\epsilon_k = t_{1} \left[ 1 + 2 \exp \left( t \sqrt{3} k_x a \alpha \right) \cos \left( k_y \frac{a}{2} \right) \right],
\]

where $k = (k_x, k_y)$ points a 2D wave vector in the FBZ, $a = |a_1| = |a_2| = \sqrt{3} \delta_0$, in which $a_0$ displays interatomic distance, and $\{a_1, a_2\}$ perform as primitive vectors (Figure 1). In (6)-(7), the intraplane hopping to the NN sites and the interplane ones are denoted by $t_1$ and $t_{11}$, respectively (Figure 2).

Our starting point for the EC tensor is the well-known Kubo formula [17–19],

\[
\sigma_{\mu\nu}(T) = \int_{-\infty}^{+\infty} d\delta \left[ -\partial_{\delta} f(\delta,T) \right] \langle \xi_{\mu\nu}(\delta) \rangle,
\]
where \( \{\mu, \nu\} \) indicate Cartesian components, \( T \) shows temperature, and \( f_\varepsilon (\varepsilon, T) \) refers to Fermi-Dirac distribution function, \( f_\varepsilon (\varepsilon, T) = \left[ 1 + \exp (\varepsilon / T) \right]^{-1} \). In band representation, energy-dependent EC, \( \xi_{\nu \mu} (\varepsilon) \), is defined as

\[
\xi_{\nu \mu} (\varepsilon) = \frac{1}{\pi N_b N_c \Omega_z} \sum_{b=1}^{N_b} \sum_{k} v^{(b)}_{\nu} (k) v^{(b)}_{\mu} (k) \left[ 3 G^{(b)} (k; E) \right]^2,
\]

in which \( b \) serves as band index, \( N_b = N_a N_p \) equals the number of the bands in the system, \( v^{(b)}_{\nu} (k) = \partial_{k_{\nu}} \xi^{(b)}_0 (k) \) describes a \( b \) band Cartesian component of the velocity operator, and \( \xi^{(b)}_0 (k) \)'s express eigenvalues of the Hamiltonian of the system. We point out that, in band representation, the Hamiltonian of the system has a diagonal form, so (6) get the shape as

\[
G (k; E) = \begin{pmatrix}
\zeta^{(1)} (k) & 0 & 0 & 0 & 0 & 0 \\
0 & \zeta^{(2)} (k) & 0 & 0 & 0 & 0 \\
0 & 0 & \zeta^{(3)} (k) & 0 & 0 & 0 \\
0 & 0 & 0 & \zeta^{(4)} (k) & 0 & 0 \\
0 & 0 & 0 & 0 & \zeta^{(5)} (k) & 0 \\
0 & 0 & 0 & 0 & 0 & \zeta^{(6)} (k)
\end{pmatrix}
\]

(10)

where \( \zeta^{(b)} (k) = E - \xi^{(b)}_0 (k) \). For monolayer graphene, \( N_p = 1 \), \( \xi^{(b)}_0 (k) \)'s are calculated as

\[
\xi^{(1)}_0 (k) = -\xi^{(2)}_0 (k) = |\epsilon_k|,
\]

(11)

so that

\[
|\epsilon_k| = t_1 \left[ 1 + 4 \left[ \cos \left( \sqrt{3} k_x \frac{a}{2} \right) + \cos \left( k_y \frac{a}{2} \right) \right] \cos \left( k_y \frac{a}{2} \right) \right]^{1/2}.
\]

(12)

For FEG, up to five layers, the eigenvalues could also be analytically found. Using (10), \( G^{(b)} (k; E) \) turns out to be

\[
G^{(b)} (k; E) = \frac{1}{E - \xi^{(b)}_0 (k)}.
\]

(13)

Moreover, the velocity operator could be represented by

\[
v_{\nu} (k) = \begin{pmatrix}
v_{\nu}^{(1)} (k) & 0 & 0 & 0 & 0 & 0 \\
0 & v_{\nu}^{(2)} (k) & 0 & 0 & 0 & 0 \\
0 & 0 & v_{\nu}^{(3)} (k) & 0 & 0 & 0 \\
0 & 0 & 0 & v_{\nu}^{(4)} (k) & 0 & 0 \\
0 & 0 & 0 & 0 & v_{\nu}^{(5)} (k) & 0 \\
0 & 0 & 0 & 0 & 0 & v_{\nu}^{(6)} (k)
\end{pmatrix}
\]

(14)

From (9), (10), and (13) and definition of velocity, the \( x \)-component of the energy-dependent EC, \( \xi_{xx} (\varepsilon) \), of the simple structures \( (N_p = 1, 2, 3, 4, 5) \) can be written as

\[
\xi_{xx} (\varepsilon) = \xi_0 \sum_k \left[ \frac{\sin^2 \left( \sqrt{3} k_x \frac{a}{2} \right) \cos^2 \left( k_y \frac{a}{2} \right)}{|\epsilon_k|^2} \right] \\
\times \sum_{b=1}^{N_b} \left[ \mathfrak{I} \left( \frac{1}{E - \xi^{(b)}_0 (k)} \right) \right]^2,
\]

(15)

while that of the Bernal structure with \( N_p = 2 \) is determined by

\[
\xi_{xx} (\varepsilon) = \xi_0 \sum_k \frac{\text{FRZ}}{k} \left[ \frac{\sin^2 \left( \sqrt{3} k_x \frac{a}{2} \right) \cos^2 \left( k_y \frac{a}{2} \right)}{|\epsilon_k|^2 + 1} \right] \\
\times \sum_{b=1}^{N_b} \left[ \mathfrak{I} \left( \frac{1}{E - \xi^{(b)}_0 (k)} \right) \right]^2;
\]

(16)

for \( N_p = 3 \), the result is

\[
\xi_{xx} (\varepsilon) = \xi_0 \sum_k \left[ \frac{\sin^2 \left( \sqrt{3} k_x \frac{a}{2} \right) \cos^2 \left( k_y \frac{a}{2} \right)}{|\epsilon_k|^2} \right] \\
\times \left[ \frac{1}{1/2} \right] \times \left[ \mathfrak{I} \left( \frac{1}{E - \xi^{(b)}_0 (k)} \right) \right]^2.
\]

(17)

When \( N_p = 4 \), it is found that

\[
\xi_{xx} (\varepsilon) = \xi_0 \sum_k \left[ \frac{\sin^2 \left( \sqrt{3} k_x \frac{a}{2} \right) \cos^2 \left( k_y \frac{a}{2} \right)}{|\epsilon_k|^2} \right] \\
\times \left[ \frac{1}{1/2} \right] \times \sum_{b=1}^{N_b} \left[ \mathfrak{I} \left( \frac{1}{E - \xi^{(b)}_0 (k)} \right) \right]^2.
\]

(18)

and \( N_p = 5 \) leads to

\[
\xi_{xx} (\varepsilon) = \xi_0 \sum_k \left[ \frac{\sin^2 \left( \sqrt{3} k_x \frac{a}{2} \right) \cos^2 \left( k_y \frac{a}{2} \right)}{|\epsilon_k|^2 + 1} \right] \\
\times \left[ \frac{1}{1/2} \right] \times \sum_{b=1}^{N_b} \left[ \mathfrak{I} \left( \frac{1}{E - \xi^{(b)}_0 (k)} \right) \right]^2.
\]
Figures 3: The FEC of mono-, bi-, tri-, tetra-, and pentalayer graphene plane for simple structure (a) and Bernal case (b). In (a) and (b), the interplane hopping term is chosen to be $t_\perp = t_\parallel/7$. (c) shows the FEC of the bilayer Bernal graphene for four values of interplane hopping term, $t_\perp = t_\parallel/14$, $t_\parallel/7$, $t_\parallel/4.67$, and $t_\parallel/3.5$.

In summary, using Green's function method and the Kubo-Greenwood formula through the TB Hamiltonian model, the EC of FLG is analytically found for single-layer and {AA, AAA, AAAA, AAAAA} simple cases as well as {AB, ABA, ABAB, ABABA} Bernal structures. The aim is to compare the EC of the single-layer graphene and FLG (see (8) and (11)–(19)). We set the intraplane hopping to the NN and interplane ones as $t_\parallel \simeq 2.8$ eV and $t_\perp = t_\parallel/7 \simeq 0.4$ eV [21–23], respectively. Figures (3) and (4) show the results. In Figure 3(a), the FECs of mono-layer graphene and FLG in simple structure are plotted, while, in Figure 3(b), those of the Bernal cases are classified. The latter are investigated and summarized in Figure 3(c) for $N_p = 2$ and different values of interplane term; that is, $t_\perp = t_\parallel/14$, $t_\parallel/7$, $t_\parallel/4.67$, and $t_\parallel/3.5$. Also, $\sigma_{xx}(T)$ is illustrated for simple and Bernal bilayer graphene in Figure 4. We mention that, for a monolayer graphene, $\Omega_c$ is just the area of the graphene unit cell.

\[ \times \sum_{b=1}^{4'} \left[ 3 \left( \frac{1}{E - \sigma_{0}^{(b)} (k)} \right) \right]^2 + \left[ \frac{1}{\xi_k^2 + (t_\parallel/2)^2} \right] \times \sum_{b=5}^{8'} \left[ 3 \left( \frac{1}{E - \sigma_{0}^{(b)} (k)} \right) \right]^2 + \left( \frac{1}{\xi_k^2} \right) \sum_{b=9}^{10'} \left[ 3 \left( \frac{1}{E - \sigma_{0}^{(b)} (k)} \right) \right]^2, \]  

in which $\xi_0 = 3\alpha^2 t_\parallel^4/(\pi N_b N_c \Omega_c)$ and $\sum'$ implies sum over just some bands not all.

\[(19)\]
It is also resulted that the FEC depends on the amount of the interplane hopping integrals. In Figure 3(c), the temperature-dependent FEC of bilayer Bernal structure is plotted for four values of $t_{\perp}$. Obviously, the more $t_{\perp}$ increases, the more $\sigma_{xx}(T)$ decreases. This could be interpreted that the procedure of increasing the interlayer hopping transforms the interlayer interactions towards the limit of somehow covalence-like bonds, which resemble the carbonic system as an insulator with four strong bonds and consequently weak EC.

We have also compared $\sigma_{xx}(T)$ of bilayer both simple and Bernal graphene in Figure 4. It is known that a finite DOS at zero energy appears in the simple case [25] in contrast to the single layer whose DOS vanishes. Therefore, because of appearing allowed states close to Fermi energy, the temperature-dependent FEC of the simple case is more than that of the Bernal one.

Totally, it is concluded that the FEC decreases by increasing the layers of the graphene due to overlapping of the nonhybridized $p_z$ orbitals perpendicular to the sheets. But, the variation in the magnitude of the FEC varies less as the layer number increases to beyond two as a result of changes in the low energy band structures. Besides, a deviation from the linear behavior of the FEC is observable at low temperatures originated from changes in relevant dispersion, especially for the simple case. It is found that more increase in interplane term causes more decrease in the temperature-dependent FEC because of transforming the interlayer interactions towards the limit of covalence-like bonds. Finally, it is resulted that the FEC of simple structure is more than that of Bernal one.

**Conflict of Interests**

The authors declare that there is no conflict of interests regarding the publication of this paper.

**References**

[1] K. S. Novoselov, A. K. Geim, S. V. Morozov et al., “Two-dimensional gas of massless Dirac fermions in graphene,” *Nature*, vol. 438, no. 7065, pp. 197–200, 2005.

[2] E. McCann and V. I. Fal’ko, “Landau-level degeneracy and quantum hall effect in a graphite bilayer,” *Physical Review Letters*, vol. 96, no. 8, Article ID 086805, 2006.

[3] H. K. Min and A. H. MacDonald, “Origin of universal optical conductivity and optical stacking sequence identification in multilayer graphene,” *Physical Review Letters*, vol. 103, no. 6, Article ID 067402, 4 pages, 2009.

[4] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, “The electronic properties of graphene,” *Reviews of Modern Physics*, vol. 81, no. 1, pp. 109–162, 2009.

[5] J.-C. Charlier, X. Gonze, and J.-P. Michenaud, “First-principles study of the electronic properties of graphite,” *Physical Review B*, vol. 43, no. 6, pp. 4579–4589, 1991.

[6] J.-C. Charlier, J.-P. Michenaud, and X. Gonze, “First-principles study of the electronic properties of simple hexagonal graphite,” *Physical Review B*, vol. 46, no. 8, pp. 4531–4539, 1992.
[7] J.-C. Charlier, J.-P. Michenaud, and P. Lambin, “Tight-binding density of electronic states of pregraphitic carbon,” Physical Review B, vol. 46, no. 8, pp. 4540–4543, 1992.

[8] J.-C. Charlier, X. Gonze, and J.-P. Michenaud, “First-principles study of the stacking effect on the electronic properties of graphite(s),” Carbon, vol. 32, no. 2, pp. 289–299, 1994.

[9] N. M. R. Peres, A. H. Castro Neto, and F. Guinea, “Dirac fermion confinement in graphene,” Physical Review B, vol. 73, no. 24, Article ID 241403, 2006.

[10] M. Koshino and T. Ando, “Orbital diamagnetism in multilayer graphenes: systematic study with the effective mass approximation,” Physical Review B, vol. 76, no. 8, Article ID 085425, 11 pages, 2007.

[11] M. Aoki and H. Amawashi, “Dependence of band structures on stacking and field in layered graphene,” Solid State Communications, vol. 142, no. 3, pp. 123–127, 2007.

[12] H. K. Min and A. H. McDonald, “Chiral decomposition in the electronic structure of graphene multilayers,” Physical Review B, vol. 77, no. 15, Article ID 155416, 5 pages, 2008.

[13] E. Wang, Y. Zhang, C. Tian et al., “Gate-variable optical transitions in graphene,” Science, vol. 320, no. 5873, pp. 206–209, 2008.

[14] Y. Zhang, T.-T. Tang, C. Girit et al., “Direct observation of a widely tunable bandgap in bilayer graphene,” Nature, vol. 459, no. 7248, pp. 820–823, 2009.

[15] Z. Q. Li, E. A. Henriksen, Z. Jiang et al., “Band structure asymmetry of bilayer graphene revealed by infrared spectroscopy,” Physical Review Letters, vol. 102, no. 3, Article ID 037403, 2009.

[16] S. Russo, M. F. Craciun, T. Khodkov, M. Koshino, M. Yamamoto, and S. Tarucha, Graphene-Synthesis, Characterization, Properties and Applications, chapter 9, InTech, 2011.

[17] S. F. Edwards, “A new method for the evaluation of electric conductivity in metals,” Philosophical Magazine, vol. 3, no. 33, pp. 1020–1031, 1958.

[18] S. F. Edwards, “The statistical thermodynamics of a gas with long and short-range forces,” Philosophical Magazine, vol. 4, pp. 1171–1182, 1959.

[19] B. Velicky, “Theory of electronic transport in disordered binary alloys: coherent-potential approximation,” Physical Review Online Archive, vol. 184, pp. 614–627, 1969.

[20] H. Mousavi, “The impact of gas molecule adsorption on the orbital magnetic susceptibility of graphene,” Journal of Magnetism and Magnetic Materials, vol. 322, no. 17, pp. 2533–2536, 2010.

[21] C. L. Lu, C. P. Chang, Y. C. Huang, R. B. Chen, and M. L. Lin, “Influence of an electric field on the optical properties of few-layer graphene with AB stacking,” Physical Review B, vol. 73, no. 14, Article ID 144427, 7 pages, 2006.

[22] Y. Takane, “Tunneling density of states in multilayer graphene deposited on a superconductor,” Journal of the Physical Society of Japan, vol. 79, no. 12, Article ID 124706, 2010.

[23] E. V. Castro, K. S. Novoselov, S. V. Morozov et al., “Electronic properties of a biased graphene bilayer,” Journal of Physics Condensed Matter, vol. 22, no. 17, Article ID 175503, 2010.

[24] C. L. Lu, C. P. Chang, Y. C. Huang, J. H. Ho, C. C. Hwang, and M. F. Lin, “Electronic properties of AA- and ABC-stacked few-layer graphites,” Journal of the Physical Society of Japan, vol. 76, Article ID 024701, 7 pages, 2007.

[25] E. McCann, D. S. L. Abergel, and V. I. Fal’ko, “Electrons in bilayer graphene,” Solid State Communications, vol. 143, no. 1-2, pp. 110–115, 2007.