Vital Annex: International Journal of Novel Research in Advanced Sciences
ISSN: 2751-756X
Volume 04 Number 02 (2025)
https://innosci.org/IJNRAS

Article

Theoretical Study of The Band Structure of Multilayer Armchair Graphene Nanoribbons (MLGNRs)

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Abstract: The electronic band structure for the stacked multilayer armchair graphene nanoribbon (MLAGNRs) is presented theoretically by using the generalized effective long-wave Hamiltonian and the tight-binding approximation. The relation between the energy gap and the number of layers in a wide range of energies around Fermi's energy level is calculated numerically. The energy of the electron depends on the momentum is investigated for an arbitrary number of layers for the armchair graphene nanoribbon having number of layers' n = 1,2 and 3 with the stacking ABC. We find, in agreement with previous calculations, that MLAGNRs are changeable from conducting to semiconducting according to the number of stacked layers and the width of the armchair graphene nanoribbons. Our results revealed the behavior of the flat electronic bands for ABC-stacked multilayer armchair graphene nanoribbon at the K-point around Fermi's energy level. This study may be useful in various forms of graphene's physics. Thus, it emphasized the possibility of controlling the electronic properties as required by the techniques based on these nanomaterials.

Keywords: Multilayers Armchair Graphene Nanoribbon (MLAGNRs); Band Structure; Energy Gap; Tight Binding Approximation.

Citation: Qetheth, M. K. Theoretical Study of The Band Structure of Multilayer Armchair Graphene Nanoribbons (MLGNRs). Vital Annex: International Journal of Novel Research in Advanced Sciences 2025, 4(2), 1-8.

Received: 10th Mar 2025
Revised: 21th Mar 2025
Accepted: 02th Apr 2025
Published: 09th Apr 2025

1. Introduction

Recently, graphene and the material derived from it such as carbon nanotubes or nanoribbons have become one of the most important systems due to its distinctive electronic, mechanical and thermal properties [1]. The experimental efforts emphasized that the monolayer graphene, bilayer graphene and the multilayers graphene can be devoted in the manufacturing of electronic devices [2]. Graphene is known as a two-dimensional structure of carbon atoms arranged in a hexagonal honeycomb pattern. Carbon has four valence electrons, one electron in the $2s$ plane and three electrons in $2p$, where the hybridization is sp^2. The electrons in $2p$ form a strong δ - bond between carbon atoms forming honeycomb cells with a spacing of about $0.142nm$. The fourth electron in $2p$ forms a π bond perpendicular to the rest of the electrons, this bond is weak compared to δ [3]. The energy gap in the δ bond is about 11 ev, which is very small compared to the π bond, so most of the effect of the low-energy behavior is determined by the π bond, The overlap between the $(2s, 2p_x, 2p_y)$ electron orbitals is zero due to symmetry and the $(2p_z)$ electrons which form the π bond that can be treated independently from the rest of the valence electrons [4]. The crystal structure of the graphene lattice contains two sub lattices, usually denoted as α and β lattice, that are unequal with an electronic π bond near the Fermi energy level and the electrons in the π bond behave as massless fermions. The conduction and valence bands of this crystal structure converge at the unequal Dirac points, called (K_+, K_-) in the form of two opposite cones [5]. The electronic properties of graphene based on the effect of the

edges in the ribbons. Thus, the presence of edges in the graphene has strong effects at low-energy spectrum of the electrons [6]. There are two types of graphene edges: armchair and zigzag, that determine the characteristics of graphene nanoribbons. Zigzag has edges which possess localized edge states with energies near the Fermi energy level. By contrast, armchair ribbon where edge states are absent, because the zigzag edges have a strong response to the stimulated magnetic field [7], numerous efforts have been achieved to investigating the effect of edges in carbonous nanomaterials.

While considerable theoretical studies has been devoted to studying the electronic structure and related properties of monolayer graphene [8]. Relatively, fewer studies on bilayer, trilayer and multilayer graphene have been performed. The energy gap in the bilayer graphene has been studied in the presence of a electric field, they found that it can be tunable for a wide range of energies (up to 250 meV) [9].

The electronic structure of multilayer graphene is treated in several ways, the tight bonding model, effective mass approximation, function density theory, etc. [10]. Where the energy gap was controlled in the ABC-type stacking of graphene layers, by applying an external electric field perpendicular to the surface of the stacked graphene layers. The electronic interactions in the multilayer structure of graphene are due to the appearance of flat bands near the Fermi surface. [11]. The electronic properties of twisted multilayer graphene have been investigated theoretically in Ref. [12].

In the current study, previous studies on the electronic band structure of graphene were generalized from a single layer to several layers. In this study, we reached three layers with different values of the width of the graphene nanoribbon, using the tight-binding theory.

2. Materials and Methods

As is known, there are two types of graphene nanoribbons, one is the armchair type and the other is the one under study. These two edges possess a 30° difference with the graphene sheet in their orientation [13]. As mentioned above, a zigzag edge possesses localized states, whereas an armchair edge does not include such localized states. By analogy to the carbon nanotubes, it is possible visualize the energy spectrum of the armchair graphene nanoribbons by taking the transversal wave number as discrete. Its analogy cannot be applied for the zig-zag graphene nanoribbons, where the longitudinal wave number *kx* in the zig-zag graphene nanoribbons depends on the transversal wave number *kn* and the ribbon width N simultaneously [14] (Figure 1).

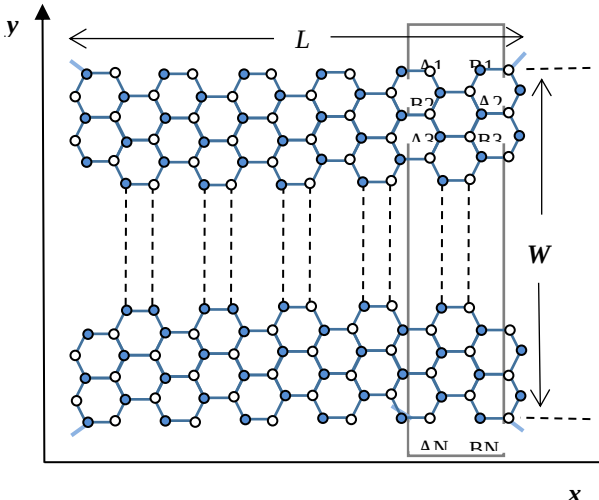

Figure 1. The figure represents a schematic diagram of an armchair-type graphene nanoribbon with length ***L*** and width ***W***, respectively.

Figure 1 shows a graphene nanoribbon armchair (AGNRs) where length is in the x-axis and width is in the y-axis. The width of the nanoribbon represents the number of dimer lines along the *x*-axis. The width of the nanoribbon is calculated as a function of the unit cell number of carbon atoms *N*, $W = \frac{N}{4}\sqrt{3}a_o$, a_o is the lattice constant of graphene.

Depending on the tight-binding approximation to the nearest neighbors, we can obtain the energy of the armchair-type graphene nanoribbon as follows [30]:

$$E(k,k_n) = Sv_F\hbar\xi \qquad where\ \xi = \sqrt{1 + 4\cos(k_n)\cos\left(\frac{k}{2}\right) + 4(\cos(k_n))^2} \tag{1}$$

The boundary condition of the armchair graphene nanoribbon (AGNR) edge, the transverse wave vector k_n is related to N as follows: $k_n = \frac{2}{\sqrt{3}a}\frac{n}{N+1}\pi,\ n = 1,2,3,\ldots\ldots,N$, a is the bond length of the lattice of graphene sheet.

For ABC - stacked multilayer AGNR, as illustrated in Fig. 1(a), the effective Hamiltonian in the first Brillouin zone near the K point, can be expressed as the follows [28]:

$$H_n = \hbar v_F \begin{pmatrix} \sigma.k & \tau\cdots & 0 \\ \tau^+ & \sigma.k\ldots & 0 \\ 0 & \tau^+ & \sigma.k \end{pmatrix} \tag{2}$$

where τ represent the 2×2 coupling matrix as follows:

$$\tau = \frac{1}{\hbar v_F}\begin{pmatrix} 0 & \gamma \\ 0 & 0 \end{pmatrix} \tag{3}$$

where $\gamma = 377\ meV$ which represent the inter layer hopping parameter. As evident that the diagonal in Eq. (2) corresponds to the monolayer graphene Hamiltonian. In the low-energy approximate (i.e. $|E| \ll \gamma$), The generalized Hamiltonian can be rewritten as follows:

$$H_n(k) = \frac{(\hbar v_F k)^n}{\gamma^{n-1}}\begin{pmatrix} 0 & e^{-in\phi} \\ e^{in\phi} & 0 \end{pmatrix} \tag{4}$$

where ϕ represent the 2-dimension polar angle in momentum space, n refer to the number of layers. In this case, the eigenstate is given as $2n$ component of the wave function. The boundary conditions at the edges of the armchair graphene nanoribbon are:

$$\psi_A(x=0) = \psi_B(x=0) = \psi_A(x=L) = \psi_B(x=L) = 0 \tag{5}$$

Then, the spinor wave function of the Hamiltonian is given as follows:

$$\psi(r) = e^{ik_y y}\begin{pmatrix} \phi_A(x) \\ \phi_B(x) \end{pmatrix} \tag{6}$$

Finally, for multilayer graphene nanoribbon with width N, the energy spectrum is given as follows: Fig (2) illustrates the schematic structure of trilayer armchair graphene nanoribbon [15] (Figure 2).

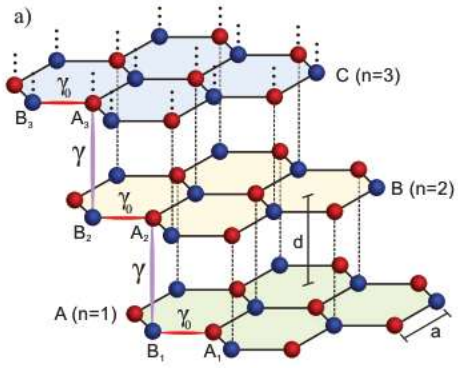

Figure 2. Rhombohedral (*ABC*) stacking in trilayer armchair graphene nanoribbon [28]

The generalized formula of the energy spectrum for n- layer (*ABC*) stacking armchair graphene nanoribbon can be approximated as follows:

$$E_n = S\hbar v_F \sqrt{1 + 4\cos(k_q^n)\cos\left(\frac{k^n}{2}\right) + 4\frac{\left[\cos(k_q^n)\right]^2}{\gamma^{n-1}}} \quad (7)$$

where $S = \pm 1$ represent the values of the valence and conduction bands, respectively, in graphene. The wavenumber of the transverse wave(k_n)is given by the following relationship:

$k_n = \frac{m\pi}{N+1}$, $m = 1,2,3,4, \ldots.$
(8)

This approximation is valid only for the low-energy limit of energy and becomes more significant with increasing the number of layers n. For Armchair graphene nanoribbons, the direct energy gap always appears at k = 0.

3. Results

Here the electronic band structure of armchair graphene nanoribbons of different layers and different values of nanoribbon width will be discussed and the relationship between multilayer and width with band structure will be found with an attempt to find some physical explanations for the results. We considered in our study that the nanoribbons have the widths 4,5,10 and 15. On the other hand, it is assumed that van der Waals interaction neglected, although it contributes to the total energy by the weak influence of the interlayer distance on overall energy band structure [16].

Table (1) present the values of the energy band gap for single, double and trilayer armchair graphene nanoribbone with various widths. It is clearly seen from table (1), that the energy gap decreases with the increase in the number of layers for AGNR with N=4, this coincides with previous theoretical results for all semiconducting AGNR [17].

Table 1. The table represents the energy gap values in units of (eV) for different width values for a multilayer armchair-type graphene nanoribbon.

N	Energy Gap [eV]		
	n=1	***n=2***	***n=3***
4	**0.76**	**0.45**	**0.03**
5	$\mathbf{6.44 \times 10^{-7}}^{*}$	**0.05**	**0.04**
10	**0.34**	**0.02***	**0.06**
15	**0.22**	**0.08**	**0.03***

4. Discussion

The zero-energy gap in AGNR with $N = 5$ increases in the double layer structure to be 0.04 eV and it increases even more when using the tri-layer structure, and thus, it turns from a conductor to a semiconductor [18]. This behavior seems clear for all widths of AGNR, except in the case of AGNR with $N = 10$, where, the energy gap of the trilayer is larger than that of the double layer AGNR [19].

Next, the variation in the band structure with the number of layers n has been illustrated by presenting the band structure in the low energy and near Fermi level of AGNR with $N = 4$ as shown in the Fig. 3 - a, b and c for single layer, double layer and trilayer respectively [20]. We noticed that there is a clear change in the electronic band structure of graphene nanoribbons with increasing the number of layers n. This means that these bands close to the Fermi surface became flatter [21]. Correspondingly, the feature of flatness extends more into Brillouin Zone as n increases, which appear as an increase in the density of states due to a denser band structure caused by parallel bands [22][23][24] (Figure 3).

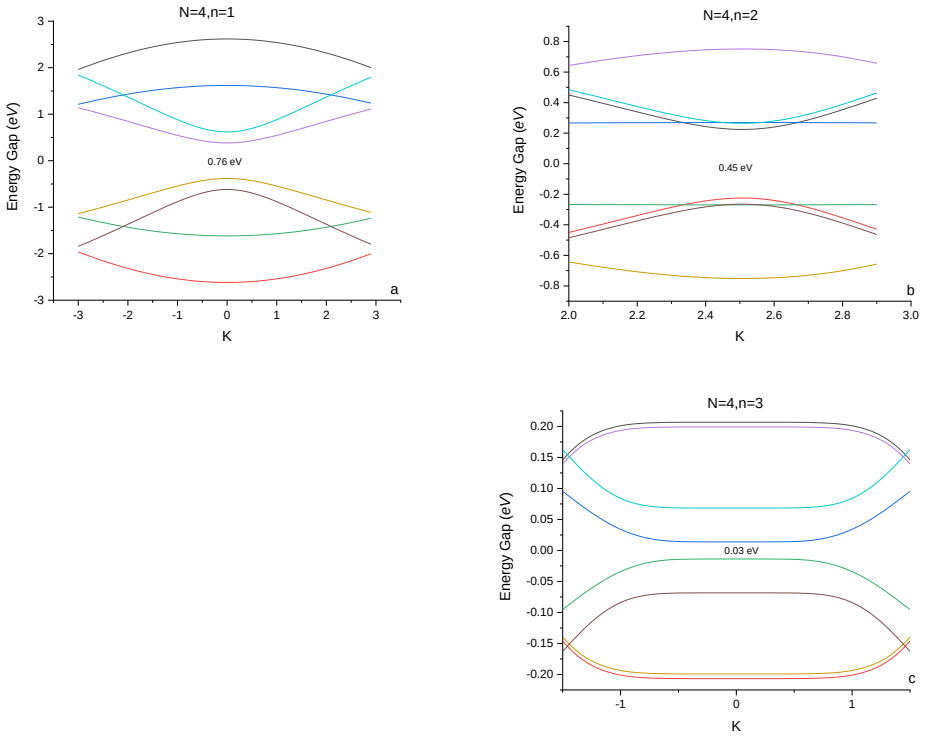

Figure 3. Energy spectrum of multilayers armchair graphene nanoribbon N=4, n=1,2,3

Fig. 4 display the variation in the band structure with the number of layers n for of AGNR with $N = 5$. It also seems clear that the band structure changes from the feature of the conductor to the semiconductor with increasing of n. Note that for the single layer AGNR with $N = 5$ the metallic behavior has a linear band, but for a double and trilayer, a quadratic band appear near Fermi level [25][26].

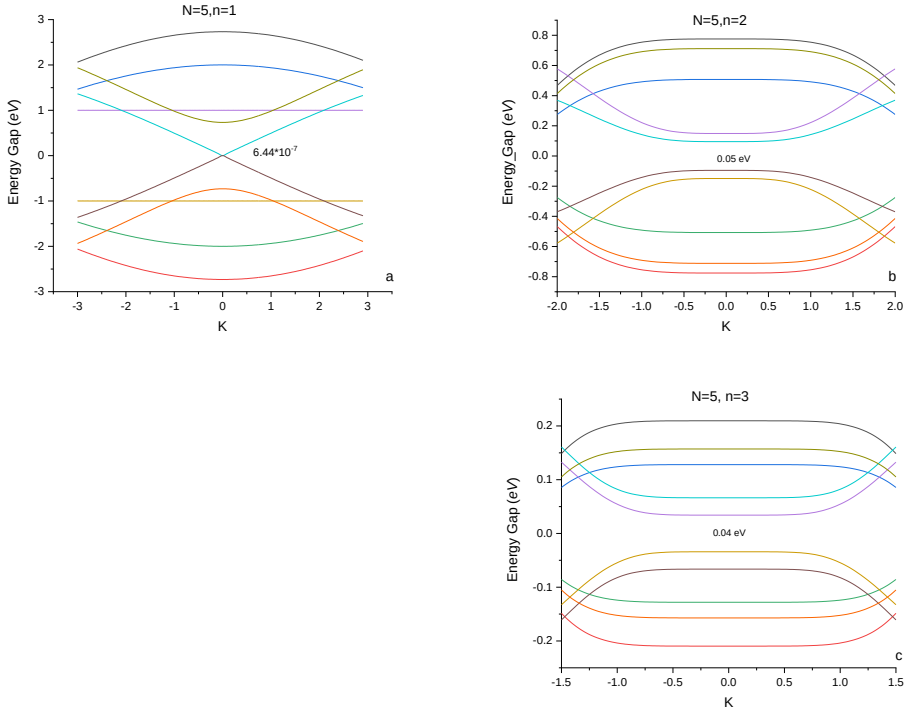

Figure 4. Energy spectrum of multilayers armchair graphene nanoribbon N=*5*, n=*1,2,3*

In Fig. 5, which represents the energy spectrum of $N = 10$, we also find a change in the energy gap when the number of layers increases, as it changes from a semiconductor at $n = 1$ to a semi-metallic with an energy gap of 0.02 $e\,V$ similar to the case $N = 1$ and $n = 4$ In Fig. 5 (a1), this is a strange case unlike the rest of the results that we obtained when reviewing the table. We note that the lowest values of the energy gap are at $(N = 5, n = 1)$ $(N = 10, n = 2)$ $(N = 15, n = 3)$ indicated by* [27][28].

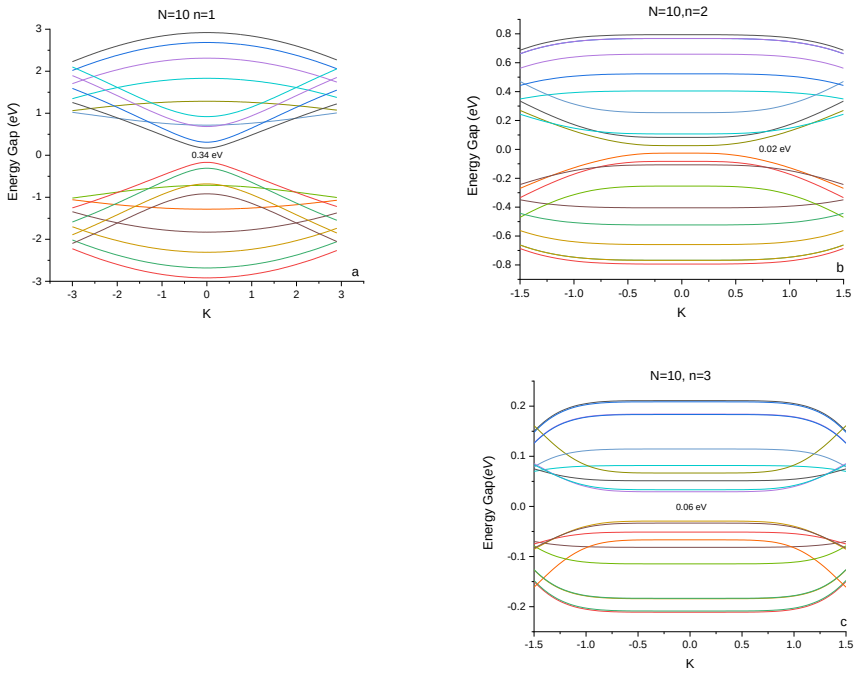

Figure 5. Energy spectrum of multilayers armchair graphene nanoribbon N=*10*, n=*1,2,3*

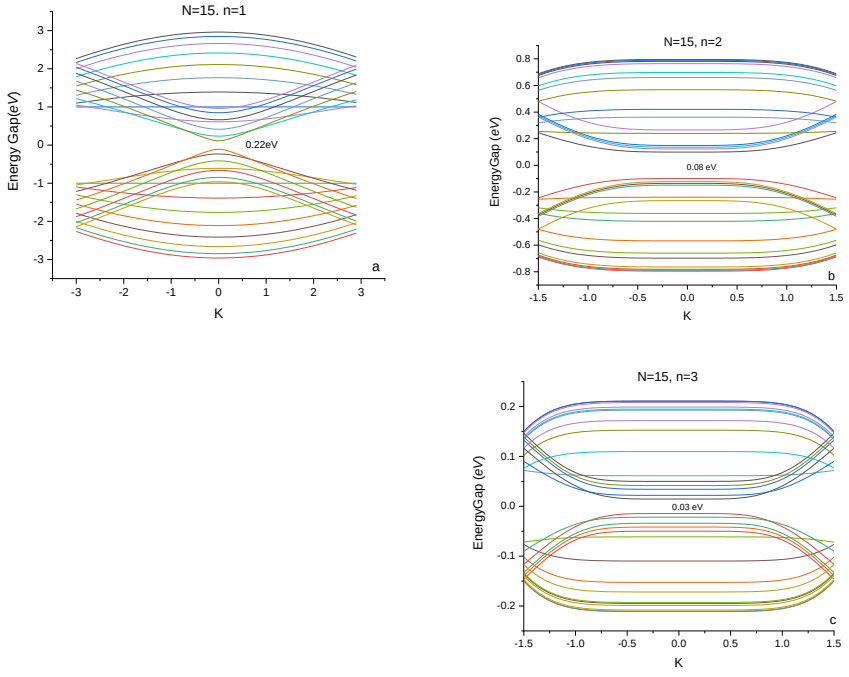

Figure 6. Energy spectrum of multilayers armchair graphene nanoribbon N=*15*, n=*1,2,3*

Band structure of multilayer armchair graphene ribbon with $N = 15$ as shown in Fig 6. It is noted that the behavior of the energy spectrum changes with the increase of the number of layers, as the energy gap decreases with the increase of n, as previously shown with AGNR with $N = 4$.When comparing the energy gap values of single-, double- and triple-layer graphene nanoribbons, we find that the energy gap value decreases with increasing number of layers. This may be due to the interference occurring between the layers near the Fermi level [29][30].

5. Conclusion

We have studied the band structure of a multilayer armchair graphene nanoribbons using tight binding approximation. The studied armchair graphene nanoribbons exhibit three pattern, one metallic and two semiconducting depending on the width of the ribbon. The energy gap in multilayer armchair graphene nanoribbons, for a semiconducting pattern, is found to be smaller than in the corresponding single layer nanoribbons, but it reveals to be larger than in the corresponding single layer nanoribbons for a metallic pattern. In view of encouraging advances in the controlling and fabrication of graphene nanoribbons, we emphasized that our study of the multi-layering effects will be significant for developing the graphene based nanodevices.

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