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Theoretical Approach to Investigate the Band Structure of Multilayers Armchair Graphene Nanoribbons (MLAGNRs)

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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.

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, 2]. The experimental efforts emphasized that the monolayer graphene, bilayer graphene and the multilayers graphene can be devoted in the manufacturing of electronic devices [3-6]. It has become known that graphene is a two-dimensional structure consists of a single layer of the carbon atoms in the form of a hexagonal lattice. 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

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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 [7]. 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 [8]. 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 level and the electrons in the π bond behave as massless fermions. The valence (hole) and conduction (electron) bands of this crystal structure converge at the unequal Dirac points, which is called (K_+, K_-) in the form of two opposite cones [9]. The electronic features of graphene based on the structure of the edges in the ribbons. Thus, the evedence of edges in the graphene has strong effects at low-energy approach of the electrons [10]. There are two types of the edges in the pristine graphene: armchair and zigzag, which determined the characteristics of graphene nanoribbons. Zigzag has an edges which possess localized states with energies near the Fermi energy level. Correspondingly, in the armchair nanoribbon, the edges states are absent, because the zigzag edges have a strong response to the stimulated magnetic field [11-15], numerous effort has been achieved to investigating the effect of edges in carbonous nanomaterials. While considerable theoretical studies has been devoted to studying the electronic structure and the essential properties of a single layer graphene [16-20]. Relatively, the studies on a bilayer, trilayer and multilayer graphene have been achieved [9]. The energy band gap in the bilayer graphene has been studied in the presence of an electric field, it's found that it can be tuned for a wide range of energies (up to 250 meV) [21]. The electronic structure of multilayer graphene is treated in several ways, the tight bonding model, effective mass approximation, function density theory, etc. [22-25]. The energy band gap inauguration only in the ABC stacked layers of graphene, where an external electric field has been applied in the perpendicular orientation to the layers has been studied, and the effect of the electron-electron interactions in multilayer structures due to the emergence of the flat bands close to the Fermi level in Ref. [26]. The electronic properties of twisted multilayer structure of graphene has been investigated theoretically in Ref. [27]. In this study, we generalize our previous studies on electronic band structure in a monolayer graphene by proposing the generalized Hamiltonian operator to investigate the behavior of the band structure for bilayer and trilayer armchair graphene nanoribbon taking into account the tight binding model.

2. Computational Details

It is known that there are two structures of the edge of graphene nanoribbon; armchair and zigzag. These two edges possess a 30◦ difference with the graphene sheet in their orientation [28]. As mentioned above, a zigzag edge possess localized energy states, whereas an armchair edge does not include such localized states. By analogy to the carbon nanotubes, it is possible visualize the energy band structure of the armchair graphene nanoribbons by taking the transversal wave number as a quantized values. This analogy cannot be represents in 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 [29].

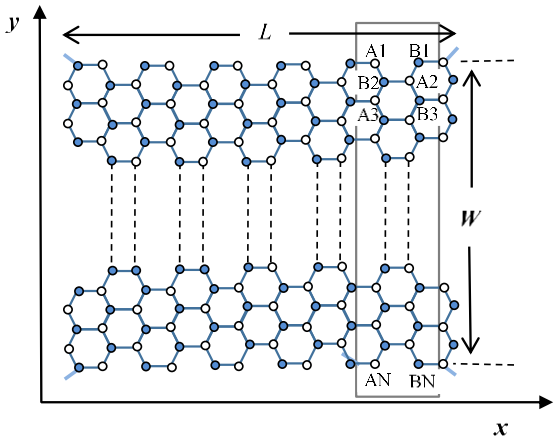

Fig.1. Schematic structure of armchair graphene nanoribbon (AGNR), W and L refer to the width and length of the graphene nanoribbon respectively [29].

Fig. (1) Illustrate the scheme of the armchair graphene nanoribbon (AGNR) along the longitudinal x-direction with width W and length L. The width of the graphene nanoribbon define by the number of dimer lines in the direction of x-axis and perpendicular to the y-axis in the graphene nanoribbon, it's represent as function of the number of carbon atoms at the unit cell N, $w = \frac{N}{4}\sqrt{3}a_o$, a_o is the graphene lattice constant. Based on the tight-binding model for the nearest-neighbors, the energy for the armchair graphene nanoribbon is given as follows [30]:

$$E(k,k_n) = S\upsilon_F \hbar\xi \ , \ \xi = \sqrt{1+4\cos(k_n)\cos\left(\frac{k}{2}\right)+4\left(\cos(k_n)\right)^2} \quad (1)$$

The boundary condition of the armchair graphene nanoribbon (AGNR) edge, the transversal component 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]:

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

Where $\boldsymbol{\tau}$ represent the $\mathbf{2\times 2}$ coupling matrix as follows:

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

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:

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

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

$$\psi_A(x=0) = \psi_B(x=0) = \psi_A(x=L) = \psi_B(x=L) = 0 \ (5)$$

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

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

Finally, For multilayer graphene nanoribbon of width *N* , Fig. (2) illustrate the schematic structure of trilayer armchair graphene nanoribbon [29].

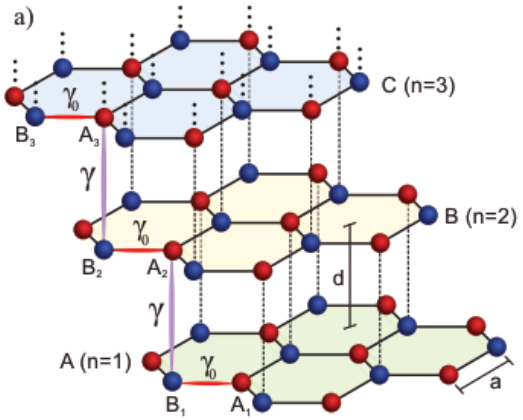

Fig.2. Rhombohedral (*ABC*) stacking in trilayer armchair graphene nanoribbon [28]

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

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

where $S = \pm 1$ ($S = +1$ and $S = -1$) represent the conduction and valence energy band, respectively. The transversal wave number k_n is given by:

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

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 and discussion

In this section, the results of the electronic band structure for the multilayer armchair graphene ribbons , the number of layers and the width dependence of energy gap has been presented and compared with previous studies [31]. It is 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 contribute to the total energy by the weak influence of the interlayer distance on overall energy band structure. 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 coincide with previous theoretical results for all semiconducting AGNR [32].

Table 1. The energy gap values for multilayers armchair graphene nanoribbon (MLAGNR)

N	Energy Gap [eV]		
	n=1	***n=2***	***n=3***
4	**0.76394**	**0.44906**	**0.0276**
5	$\mathbf{6.44 \times 10^{-7}}^{*}$	**0.0468**	**0.03828**
10	**0.33834**	**0.0169***	**0.05804**
15	**0.22228**	**0.07526**	**0.02908***

The zero energy gap in AGNR with *N*=5 increases in the double layer structure to be 0.0468 *eV* and it increases even more when using the tri-layer structure, and thus, it turns from a conductor to a semiconductor. 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. Next, the band structure variation as a function of 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. It is evident that the band structure varies obviously with increasing the layers number n, causing bands near Fermi energy level to become flatter. Correspondingly , the flatness feature extends into the first Brillouin Zone when the number of layer *n* increases, which appear as an the density of states increased due to the denser band structure caused by parallel bands.

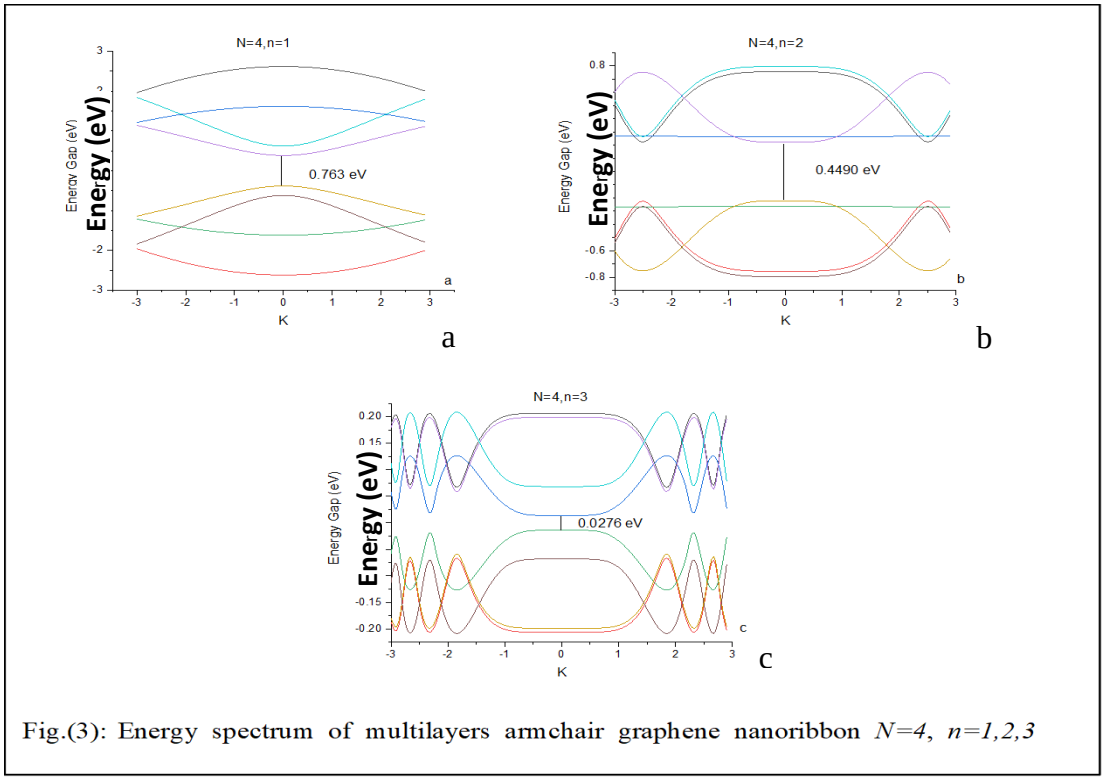

Fig.(3): Energy spectrum of multilayers armchair graphene nanoribbon *N=4, n=1,2,3*

Fig. 4 display the band structure variation as a function of 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.

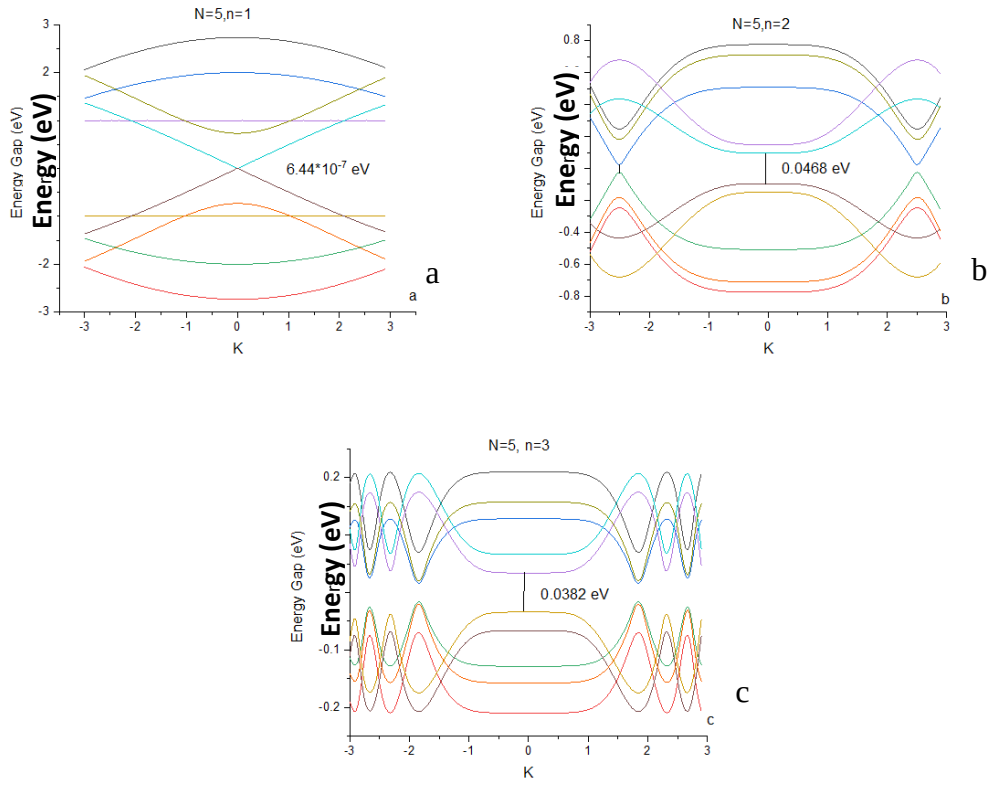

Fig.(4): Energy spectrum of multilayers armchair graphene nanoribbon *N=5, n=1,2,3*

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 0.016 *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.

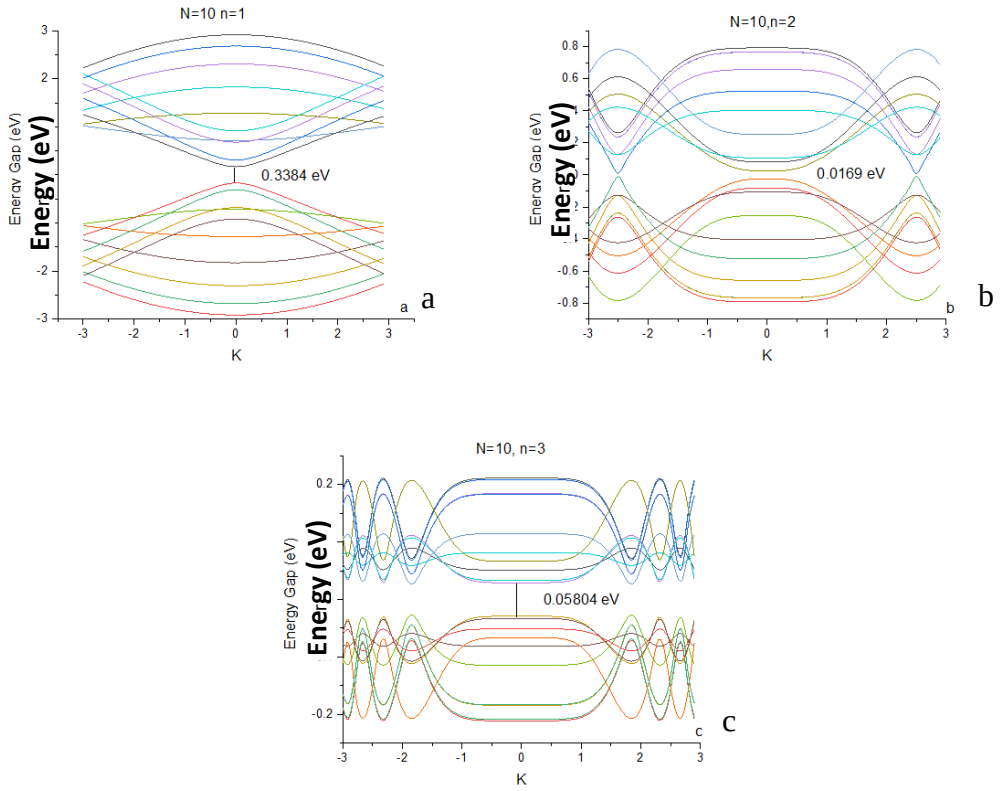

Fig.(5): Energy spectrum of multilayers armchair graphene nanoribbon *N=10, n=1,2,3*

The energy band structure of the multilayers AGNR with N=15 plotted in Fig. 6. It is noted that the behavior of the energy spectrum varies with increasing the number of the layers, as the energy gap decreases with the increase of n, as previously shown with AGNR with N= 4. The comparing between single, bilayer and trilayer armchair graphene nanoribbons, the energy gaps gradually decrease. When the number of the layers increases, new energy bands appear near the Fermi energy level as a result of the interference between these layers, which in turn reduces the value of the energy gap.

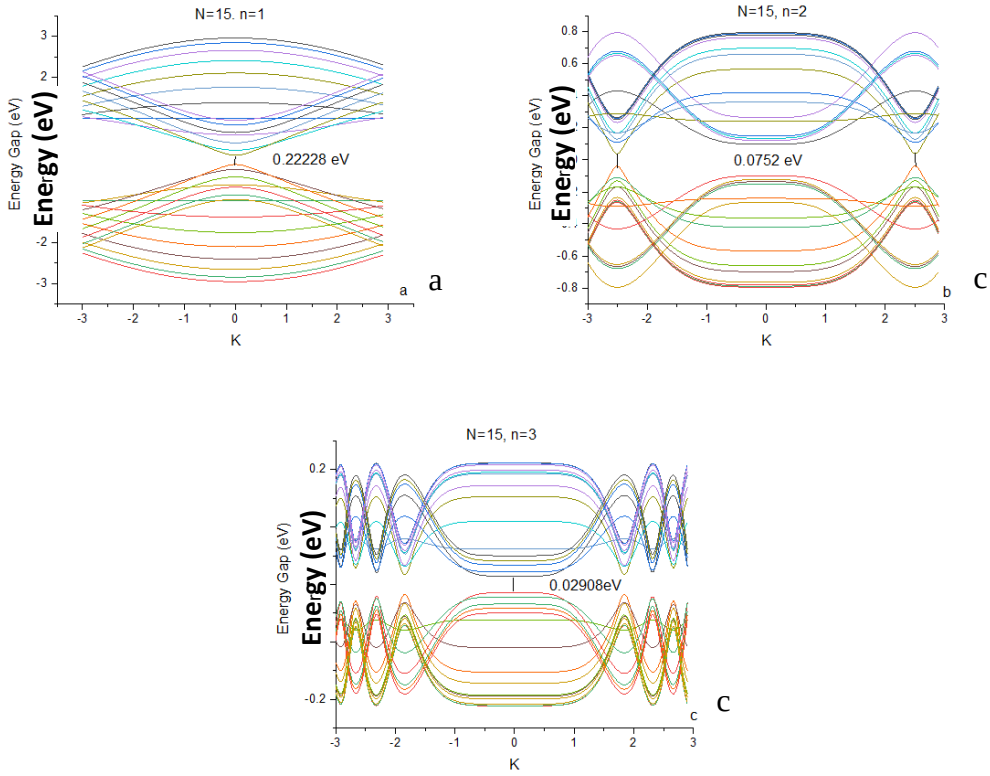

Fig.(6): Energy spectrum of multilayers armchair graphene nanoribbon *N=15*, *n=1,2,3*

4. 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 band gap in multilayer armchair graphene nanoribbons, for a semiconducting pattern, is found to be smaller comparing to that in the corresponding single layer nanoribbons, but it reveal to be larger than in the corresponding single layer nanoribbons for a metallic pattern. In view of encouraging advances in the controlling and innovation 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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دراسة نظريه لتركيب الحزم لأشرطة الجرافيين النانوية نوع ذراع الكرسي

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معلومات البحث

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الملخص

تم دراسة تركيب حزم الطاقة الإلكترونية للجرافين النانوي نوع ذراع الكرسي متعدد الطبقات (MLAGNRs) من الناحية النظرية باستخدام هاملوتون الفعال لتقريب الموجة الطويلة والربط المحكم. تم حساب العلاقة بين فجوة الطاقة وعدد الطبقات في نطاق واسع من الطاقات حول مستوى طاقة فيرمي عددياً. تعتمد طاقة الإلكترون على الزخم ويتم فحصها لعدد عشوائي من الطبقات لشريط الجرافين النانوي ذو الذراعين الذي يحتوي على عدد الطبقات n = 1 و2 و3 مع التراص .ABC لقد وجدنا، بالاتفاق مع الحسابات السابقة، أن MLAGNRs قابلة للتغيير من التوصيل إلى أشباه الموصلات وفقًا لعدد الطبقات المكدسة وعرض شرائط الجرافين النانوية ذات الكرسي بذراعين. كشفت النتائج التي توصلنا إليها عن سلوك الحزم الإلكترونية المسطحة لشريط الجرافين النانوي متعدد الطبقات ذو الذراعين ABC عند النقطة K حول مستوى طاقة فيرمي. قد تكون هذه الدراسة مفيدة في أشكال مختلفة من فيزياء الجرافين. وبالتالي تم التأكيد على إمكانية التحكم في الخواص الإلكترونية كما تتطلبها التقنيات المعتمدة على هذه المواد النانوية.

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