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Influence of single vacancy defect at varying length on electronic properties of zigzag graphene nanoribbons

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ABSTRACT

Graphene, identified in 2004, is now an established two-dimensional (2D) material due to its outstanding physical and electronic characteristics namely its superior electrical conductivity. Graphene is a zero-gap material that has linear dispersion with electron-hole symmetry. As pristine sheet, it cannot be utilized in digital logic application without the induction of a band gap inside the band structure. In our work, the modeling and simulation of graphene nanoribbons (GNRs) are carried out to determine its electronics properties that are benchmarked with other published simulation data. A 4-Zigzag GNRs (4-ZGNRs) under different length are utilized. A single vacancy defects is introduced at various positions inside the atomic structure. The theoretical model is implemented based on single-neighbour tight binding technique coupled with a non-equilibrium Green's function formalism. The single vacancy defects are represented by the elimination of tight binding energies in the Hamiltonian matrix. Subsequently, these matrix elements are utilized to compute dispersion relation and density of states (DOS) through Green's function. It is found that single vacancy defects at different positions in 4-ZGNRs' atomic structure under varying length has no significant impacts on the sub-band structure but these vacancies impact the DOS that are computed throught Green's function approach.

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1. INTRODUCTION

Graphene Nanoribbons (GNRs) or strips of graphene are theoretically introduced by Mitsutaka Fujita in 1997 [1]. GNRs are one of the potential material that can complement and ultimately replace Silicon [2] because of its remarkable electronic properties that can be engineered by controlling its width, length and edge orientation as well as defects [3, 4]. Defects can be a beneficial advantage that can alter the electronics properties to enhance their applicability for a distinctive applications [5]. In tight binding approach, Hamiltonian matrix that is a solution of Schrödinger's equation is obtained to compute electronic properties of 4-ZGNRs with single vacancy defects at different locations inside the band structure [6-8]. The nanoribbon system can be described by Hamiltonian matrix the elements [9, 10]. A tight binding 4-ZGNR model is produced by disintegrating 4-ZGNRs' quasi-2D structure into an equivalent one-dimensional (1D) matrix system with alpha and beta matrices [11, 12]. Figure 1 depicts an imperfect 4-ZGNRs at length of 3 unit cells with single vacancy defect at intersection of first row and third column.

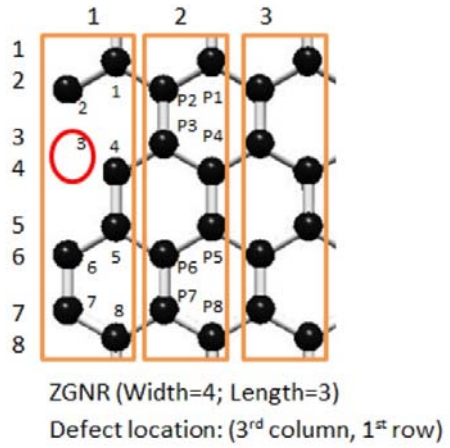


Figure 1. (a) Atomic structure of 4-ZGNRs at length of 3 unit cells
for defect location at first row of third column intersection

2. RESEARCH METHOD

Non-ideal ZGNRs with single vacancy defect is implemented through single neighbor tight-binding approach within non-equilibrium Green's function formalism in MATLAB. Tight-binding method uses semi-empirical model for simple and fast computation. The construction of alpha matrix, α that consists of the interaction among atoms within the unit cell, is shown in (1). The (2) shows Beta matrix, β that encapsulates the interaction between unit cell [13, 14]. Parameter t is the tight binding energy between carbon atoms and -2.7eV are used for nearest neighbout [15]. The red zeroes in Equation 1 highlights the affected tight-binding energies that are eliminated when a carbon atom is vacated. The (3) shows the Hamiltonian operator with the inclusion of α and β matrices. The α matrix of each unit cell is located at the diagonal of Hamiltonian matrix whereas the β matrix is located at the top of the diagonal matrix. Parameter β' matrix is the transpose of β matrix located at the bottom of diagonal.

$$\alpha_{4-ZGNR} = \begin{array}{c} \\ 1 \\ 2 \\ 3 \\ 4 \\ 5 \\ 6 \\ 7 \\ 8 \end{array} \begin{array}{c} \begin{array}{cccccccc} 1 & 2 & 3 & 4 & 5 & 6 & 7 & 8 \end{array} \\ \begin{bmatrix} 0 & t & 0 & 0 & 0 & 0 & 0 & 0 \\ t & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & t & 0 & 0 & 0 \\ 0 & 0 & 0 & t & 0 & t & 0 & 0 \\ 0 & 0 & 0 & 0 & t & 0 & t & 0 \\ 0 & 0 & 0 & 0 & 0 & t & 0 & t \\ 0 & 0 & 0 & 0 & 0 & 0 & t & 0 \end{bmatrix} \end{array} \quad (1)$$

$$\beta_{4-ZGNR} = \begin{array}{c} \\ 1 \\ 2 \\ 3 \\ 4 \\ 5 \\ 6 \\ 7 \\ 8 \end{array} \begin{array}{c} \begin{array}{cccccccc} p1 & p2 & p3 & p4 & p5 & p6 & p7 & p8 \end{array} \\ \begin{bmatrix} 0 & t & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & t & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & t & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & t & 0 \end{bmatrix} \end{array} \quad (2)$$

$$H_{4-AGNR/ZGNR,L=3} = \begin{bmatrix} \alpha_{11} & \beta_{12} & 0 \\ \beta'_{21} & \alpha_{22} & \beta_{23} \\ 0 & \beta'_{32} & \alpha_{33} \end{bmatrix} \quad (3)$$

The periodic boundary condition is the applied to obtain the the Hamiltonian operator matrix in (3). Following that, the energy and wave number relationship is given by the dispersion relation in (4) where α and β matrices of (1) and (2) are used respectively to yield the system energy sub-band structure [13, 16].

$$E = \alpha + \beta' e^{-ika} + \beta e^{ika} \tag{4}$$

The availability of energy states can be shown through the density of states (DOS) in (5) [17] that required non-equilibrium Green's function (NEGF) formalism

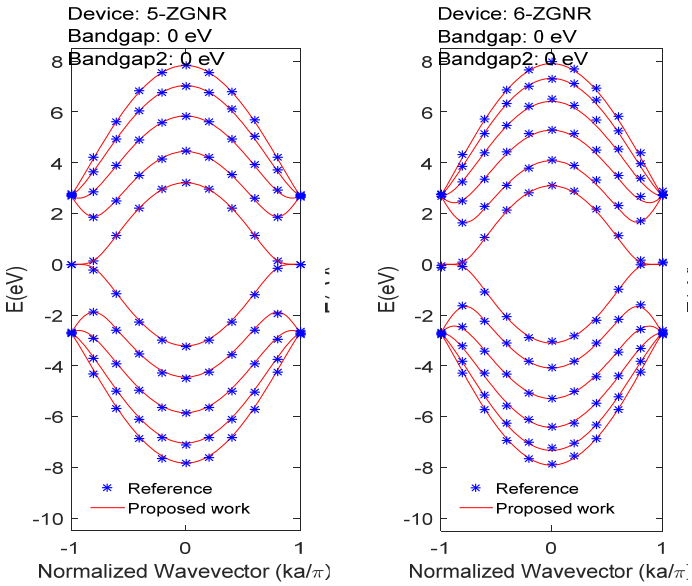
$$DOS(E) = \frac{1}{\pi} \mathrm{Im}\left[Trace\left[G_F \right] \right] \tag{5}$$

$$G_F = \left[\left(E + i\eta \right) * I - H \right]^{-1} \tag{6}$$

where G_F is retarded Green's function, η is a very small imaginary value to block divergence of the inverse matrix, I is an identity matrix, and H is the Hamiltonian operator matrix [18].

3. RESULTS AND DISCUSSION

First, the band structure of pristine ZGNRs is simulated and compared with previously reported data on the band structure. Figure 2 illustrates the band structure of 4-ZGNRs and 5-ZGNRs with previously reported studies that also adopted tight binding approach [19, 20]. It is demonstrated that our results are in good agreement with the previously published data. Figure 3 shows the band structure of 8-ZGNRs generated using tight-binding method versus Density Functional Theory (DFT) method [21]. As expected, there are differences in band structure as our 8-ZGNR does not have any bandgap whereas DFT shows a small bandgap. DFT method are used to determine the ground state properties of an electronic system from the electron density and suitable for a large and complex systems. Nevertheless, tight binding is sufficient for a simple system and less computationally expensive than DFT approaches.



(a) 5-ZGNRs (b) 6-ZGNRs

Figure 2. Band structure of pristine (a) 5-ZGNRs; (b) 6-ZGNRs using tight binding method versus previously reported data [19, 20]

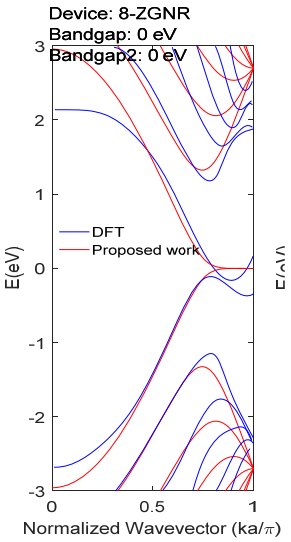
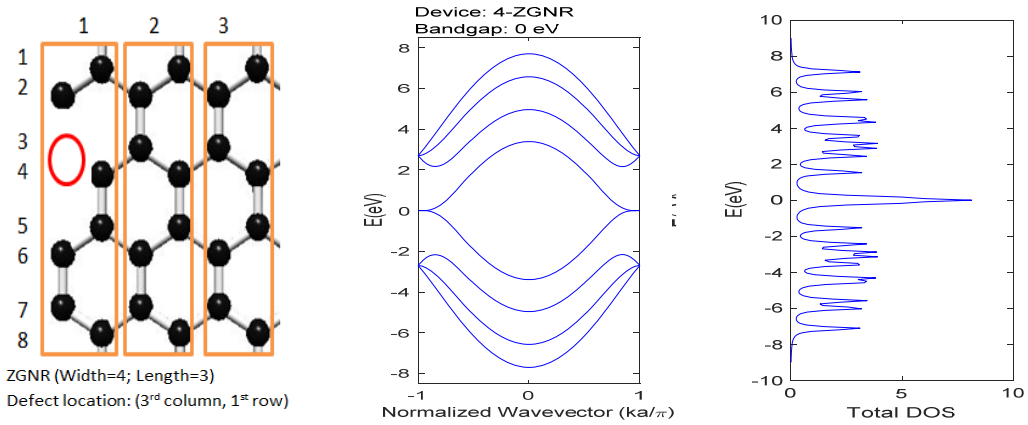


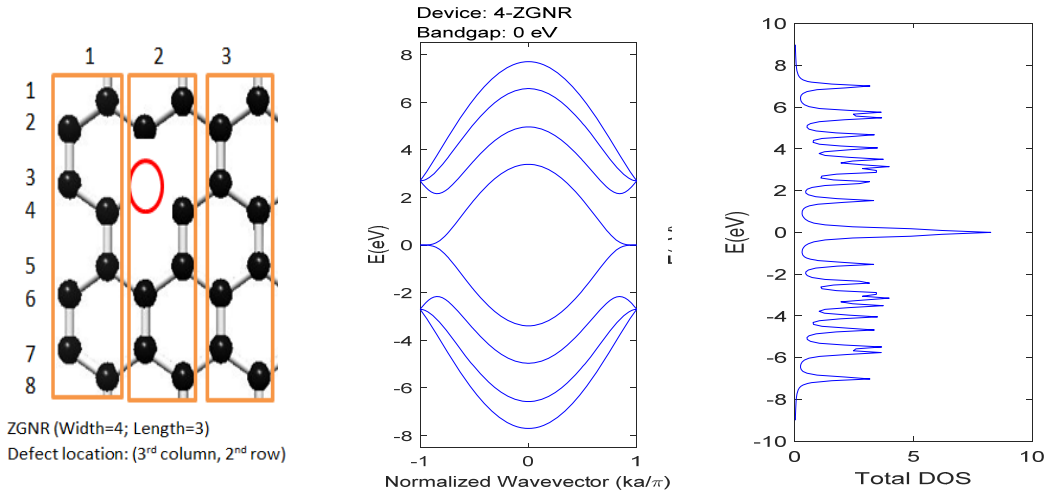
Figure 3. Band structure of pristine 8-ZGNRs using tight binding method versus DFT method [21]

Next, vacancy defect is then introduced in the atomic structure of ZGNRs and the electronics properties of non-ideal nanoribbon is explored. The position of the single vacancy defect inside atomic structure is defined in terms of a row versus column plane. Figure 4 (a) and Figure 5 (a) depict a non-ideal 4-ZGNRs at length of 3 unit cells with single vacancy defect at different locations. A pristine 4-ZGNR with length of 3 unit cells has 24 carbon atoms inside the atomic structure. The single vacancy defect decreases the number of carbon atoms to 23. In Figure 4 (a), a single carbon atom is absent at the first row of third column intersection along the edge of 4-ZGNR atomic structure. In Figure 5 (a), a single carbon atom is missing at the second row of third column intersection within the bulk of 4-ZGNR atomic structure. Red circle in the figure indicates the absence of atoms inside the atomic structure. The interaction to previously neighbouring carbon atoms no longer exist thus the tight bonding energies are completely removed from the 4-ZGNR Hamiltonian matrix elements. Finally, dispersion relation and Green's function are computed to study the non-ideal 4-ZGNRs' electronic properties at varying defect locations particularly the sub-band structure in Figure 4 (b) and Figure 5 (b) and Green's function DOS in Figure 4 (c) and Figure 5 (c) respectively.



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

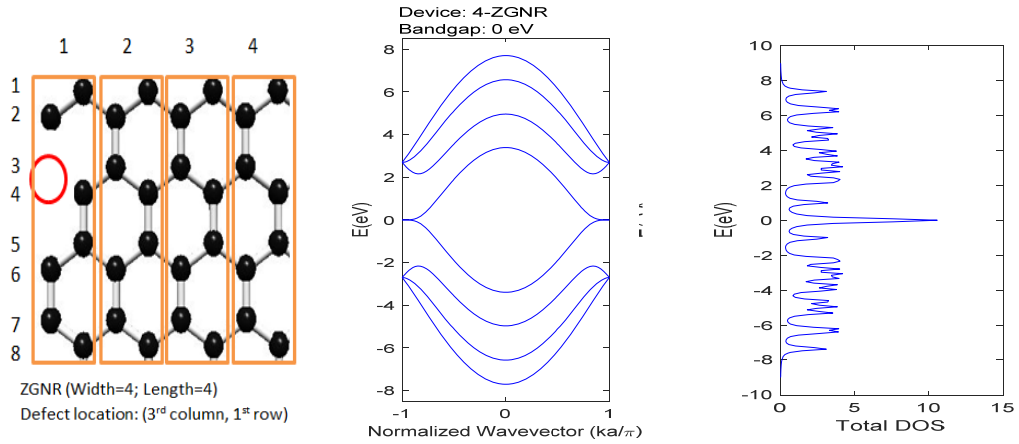
Figure 4. (a) Atomic structure of 4-ZGNRs at length of 3 unit cells for defect location at first row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

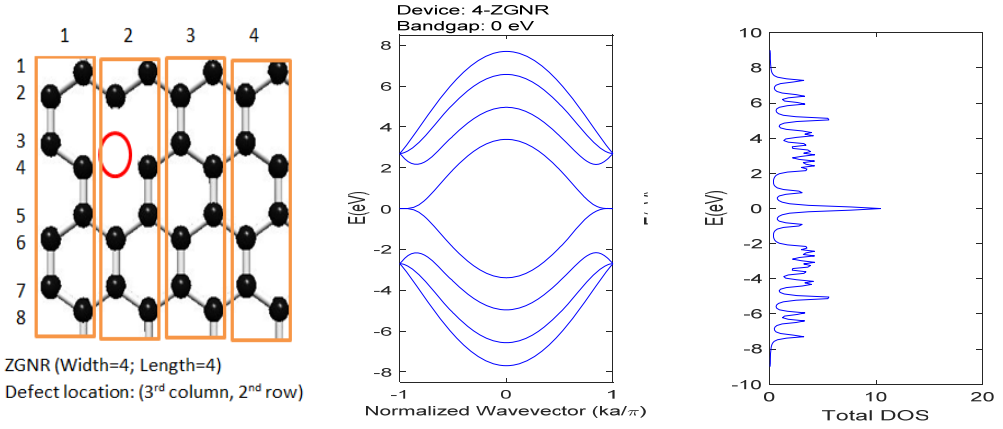
Figure 5. (a) Atomic structure of 4-ZGNRs at length of 3 unit cells for defect location at second row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)

A zero band gap is observed of non-ideal 4-ZGNR at length of 3 unit cells and thus behaves like a metal. Figure 4 (b) and Figure 5 (b) reveals that the sub-band structure is unaffected by varying positions of single vacancy defect in 4-ZGNRs' atomic structure. The peak values for the sub-band are consistent across all configurations which are around ±8eV. However, vacancies directly affect the Green's function DOS of Figure 4 (c) and Figure 5 (c) where each DOS has distinct number of peaks and shapes. For instance, the peak numbers for DOS inside Figure 4 (c) and Figure 5 (c) are 19 and 20 respectively. In addition, whenever there is a vacancy defect along the edges or inside the bulk of 4-ZGNRs, there is a peak arising at 0eV. A non-ideal 4-ZGNRs at length of 4 unit cells with single vacancy defect at different locations inside atomic structure are shown in Figure 6 (a) and Figure 7 (a). A pristine 4-ZGNR with length of 4 unit cells has 32 carbon atoms inside the atomic structure but reduces to 31 when vacancy is introduced. A carbon atom is missing at the intersection of first row of third column in Figure 6 (a) and at the intersection of second row and third column in Figure 7 (a). The absence of atom is represented by the red circle. Dispersion relation and Green's function DOS of a non-ideal 4-ZGNRs are shown in Figure 6 (b) and Figure 7 (b) as well as Figure 6 (c) and Figure 7 (c) respectively.



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

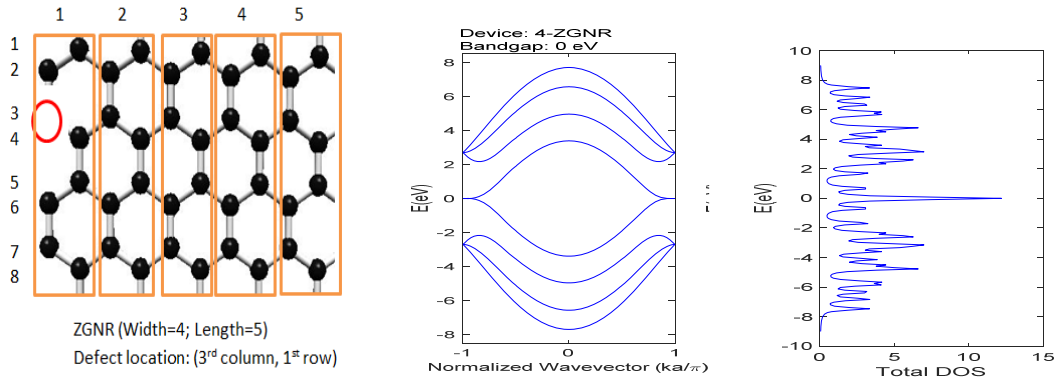
Figure 6. (a) Atomic structure of 4-ZGNRs at length of 4 unit cells for defect location at first row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

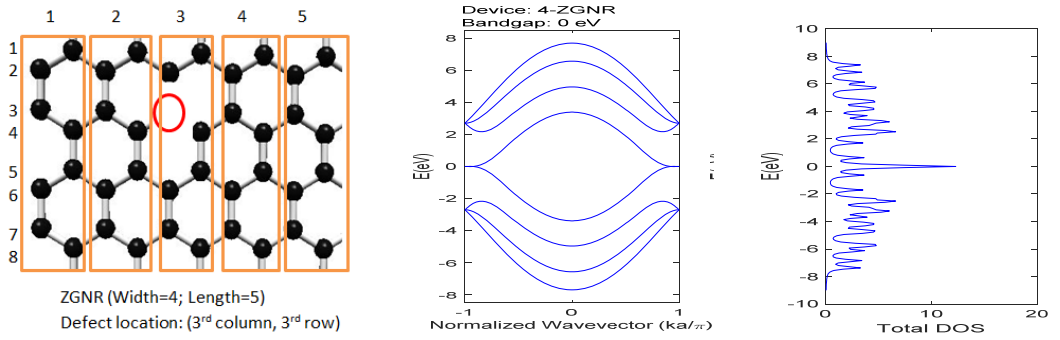
Figure 7. (a) Atomic structure of 4-ZGNRs at length of 4 unit cells for defect location at second row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)

The DOS of Figure 7 (c) has 4 extra number of peaks compared with DOS inside Figure 6 (c). Moreover, when vacancy defect is located at the edge of 4-ZGNR, the peak value of highest DOS peak of Figure 6 (c) is lower by 5% than the highest peak of Figure 7 (c) where the single vacancy defect is located in the bulk. The non-ideal 4-ZGNRs at length of 5 unit cells with single vacancy defect at different locations inside atomic structure are illustrated in Figure 8 (a) and Figure 9 (a) where there are 39 carbon atom. A carbon atom is missing at the intersection of first row of third column in Figure 8 (a) and at the intersection of third row of third column in Figure 9 (a). Dispersion relation and Green's function DOS of a non-ideal 4-ZGNRs are shown in Figure 8 (b) and Figure 9 (b) as well as Figure 8 (c) and Figure 9 (c) respectively.



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

Figure 8. (a) Atomic structure of 4-ZGNRs at length of 5 unit cells for defect location at first row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)



(a) Atomic structure (b) Sub-band plot (c) Green's function DOS plot

Figure 9. (a) Atomic structure of 4-ZGNRs at length of 5 unit cells for defect location at third row of third column intersection; (b) Sub-band plot; (c) Green's function DOS plot of 4-ZGNRs as in (a)

For 4-ZGNR with single vacancy defect located at the edge but under varying length, the disclosure of the findings from Figure 4 (b), Figure 6 (b) and Figure 8 (b) reveal the sub-band structure is not at all affected by varying length. In fact, varying length only affects the Green's function DOS. Non-ideal 4-ZGNR with single vacancy defect under varying length shows metallic properties as band gap is. The peak value for these sub-band occur at ±8eV. Furthermore, Figure 5 (b), Figure 7 (b) and Figure 9 (b) of 4-ZGNR with single vacancy defect located in the bulk but under varying length, also show similar characteristics as previously mentioned. Key differences are noticeable within the DOS for 4-ZGNR with single vacancy defect located at the edge and in the bulk. The highest peaks inside DOS of Figure 4 (c) to Figure 9(c) rise with length. Likewise, the number of peaks also increased.

4. CONCLUSION

In this paper, the electronic properties of a tight binding model of GNR with single vacancy is studied. The tight binding models is generated for 4-ZGNRs under varying length containing a single vacancy defect at different atomic structure locations. The Hamiltonian matrix was modified by identifying the location where defects occurred and eliminating tight binding energies. Non-ideal 4-ZGNRs' electronic properties such as Green's function DOS and sub-band structure are computed. Varying the length and position of the single vacancy defect on 4-ZGNRs' atomic structure has no effect on the sub-band structure but does have an profound implication on DOS as the Hamiltonian operator is used for Green's function computation.

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