Study of Density of States in Bilayer Graphene

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Abstract

This report investigates the electronic properties of graphene structures using the tight-binding model. The first section introduces the tight-binding model, detailing its formulation through first and second quantization formalisms. The subsequent section provides an overview of graphene's structural aspects, covering monolayer and bilayer configurations with emphasis on AA and AB stacking patterns. The main focus of the report lies in the following three subsections, which analyze the density of states (DOS) under open boundary conditions for monolayer AA and AB bilayer structures. The results reveal distinct features in the DOS, highlighting differences between monolayer and bilayer graphene configurations.

1 Introduction

Graphene, an atomically thin material composed of carbon atoms arranged in a hexagonal lattice, has captivated researchers for its unique electronic properties[2]. These properties are fundamentally governed by the behavior of electrons within the material's structure. To comprehensively understand and analyze these electronic behaviors, theoretical models such as the tight-binding model are indispensable.The tight-binding model provides a robust framework for studying graphene's electronic structure, employing both first and second quantization formalisms.

Graphene exists in monolayer structural configurations, with different types of shapes i.e., Nanotubes and fullrenes. The stacking pattern between layers—such as AA and AB (Bernal) stacking—greatly influences the material's electronic properties.

In this report, we focus on studying the density of states (DOS) for two specific configurations: bilayer graphene with AA stacking and AB stacking. By comparing and contrasting the DOS profiles of monolayer AA and bilayer AB graphene, we aim to discern unique electronic characteristics that arise from their distinct structural arrangements. This investigation contributes to advancing our understanding of graphene's electronic behavior and its potential applications in future technologies.

2 Tight Binding Approximation

The tight binding approximation is a method used in solid-state physics to model the electronic structure of solids[1]. According to this approach, the overlap of the atomic wave function is enough to require correction of the picture of an isolated atom. The approximation is most useful for describing the energy band that arises from the partially filled d-shells of transition metal atoms and for describing the electronic structure of the insulator.

In crystals, atoms are sitting in a periodic lattice. The periodicity potential leads naturally to Bloch's theorem and the prediction of energy bands.

Therefore, one is left with a picture of (approximately) independent electrons moving in a periodic potential generated by the ionic cores. The wave functions of those independent electrons can be written as linear combinations of the orbitals of isolated atoms. This approximation can be justified by assuming that each electron is strongly bound to a single core, with some probability of tunneling to neighboring sites in the lattice. Such approximation is thus known as a tight-binding (TB) model.

2.1 First Quantization formalism

The TB model consists of writing the electron's wave function as a linear combination of atomic orbitals, considering the whole sites of a given crystal. We assume that the electron is strongly bound to the nucleus in these orbitals, It is confined to a region of small dimensions compared to internuclear distances.

We are interested in Energy-dispersion calculation using the first quantization of a 1-D linear chain:

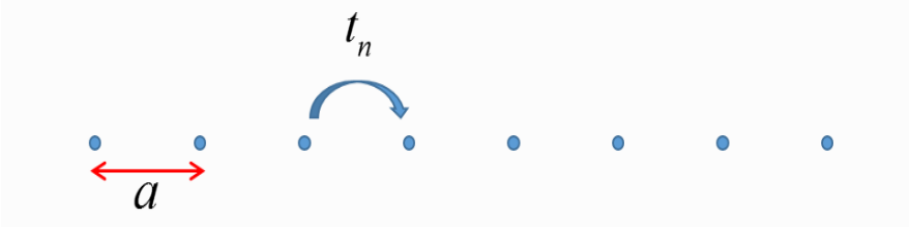

Figure 1: 1-D linear chain

where 'a' is the atomic distance in lattice and $'t_n'$ is the hopping parameter(related to the interaction energy of electrons located at different sites).

The wave function that describes the electrons in the crystal similar to the Bloch function, given by :

$$\phi_k(x) = \frac{1}{\sqrt{N}} \sum_n (e^{i(k*t_n)} \phi_a(x - t_n), \qquad n = 1, 2, 3, \ldots \tag{1}$$

For the Hamiltonian, we get the linear combination:

$$\Psi_{nk} = \sum_i C_{ik}\phi_{ik}(x) \tag{2}$$

We are interested in calculating :

$$E(k) = \frac{\langle\psi|H|\psi\rangle}{\langle\psi|\psi\rangle} \tag{3}$$

where

$$H = -\frac{\hbar^2}{2m}(\partial_x^2) + V(x) \tag{4}$$

Putting the value of Hamiltonian from Eq.(3) and using inner product property, the energy-dispersion relation is obtained as :

$$E_k = E_0 - 2tcos(ka) \tag{5}$$

2.2 Second Quantization formalism

In second quantization, the Hamiltonian for a linear chain with one site per unit cell can be written as

$$\hat{H} = \sum_i \varepsilon_o \hat{c}_i^\dagger \hat{c}_i + \sum_{i,j} t(\hat{c}_j^\dagger \hat{c}_i + \hat{c}_i^\dagger \hat{c}_j) \tag{6}$$

where '$\hat{c}_i^\dagger$' and '$\hat{c}_i$' are the creation and annihilation operators that act upon a basis of orthogonal states, 'ε_0' represents onsite energy and 't' is the hopping energy associated with the neighboring sites.

Using Fourier transformation, operators can be expressed in momentum space as

$$\hat{c}_i = \frac{1}{\sqrt{N}} \sum_k e^{ikx_i} \hat{c}_k \tag{7}$$

$$\hat{c}_i{}^\dagger = \frac{1}{\sqrt{N}} \sum_k e^{-ikx_i} \hat{c}_k{}^\dagger \tag{8}$$

Using Eq.(7) and (8) in Eq.(6), we can easily get,

$$\hat{H} = \sum_k \hat{c}_k{}^\dagger \hat{c}_k (te^{ika} + te^{-ika}) + h.c = \sum_k \hat{c}_k{}^\dagger \hat{c}_k E(k) + h.c \tag{9}$$

which gives us dispersion relation :

$$E(k) = 2tcos(ka) \qquad \text{for 't' is real} \tag{10}$$

3 Graphene

In Graphene, carbon atoms are arranged in a two-dimensional honeycomb lattice, where each carbon atom is sp^2 hybridized, forming strong covalent bonds with three neighboring carbon atoms and creating a hexagonal pattern. The remaining unhybridized p orbitals overlap to form a delocalized electron cloud, contributing to graphene's remarkable electrical conductivity. When multiple layers of graphene are stacked, they form graphite, held together by weak van der Waals forces. Graphene's unique properties, such as high electrical conductivity, mechanical strength, flexibility, and transparency, stem from its atomic arrangement and make it a material of great interest for various technological applications.

Graphene has 2 sub-lattices 'A' type which connects to type 'B' and vice-versa. This is responsible for its bipartite nature. When we go to bilayer graphene, AA does not maintain the bipartite nature because it breaks the distinct separation of sublattices. Each atom in one sublattice has a corresponding atom directly above or below it, leading to direct overlap and interaction between the same types of atoms (A-A and B-B). In AB-stacked bilayer graphene, atoms in one layer are positioned such that half of the atoms in the top layer (sublattice A) are directly above the atoms in the bottom layer's sublattice B, while the other half (sublattice B) is in the center of the hexagons formed by the bottom layer hence this configuration maintains the bipartite nature.

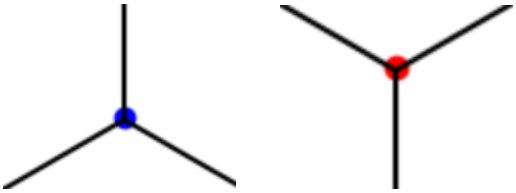

Figure 2: A type and B type sublattice

3.1 Monolayer Graphene

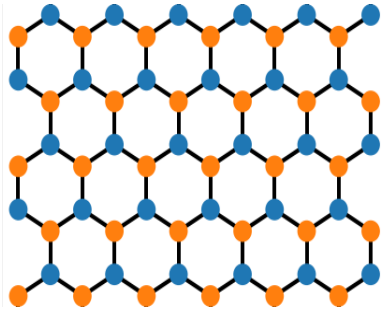

Figure 3: Graphene monolayer with A and B sublattice.

Tight Binding Hamiltonian: We will confine our study to analyzing electrons in p-orbitals. Assume the electron hopping is limited to nearest neighbors (NN) only. As discussed in section (2.2), the tight-binding Hamiltonian in second quantization formalism for this system will be:

$$\hat{H} = -t \sum_{<i,j>} \left(a_i^{\dagger} b_j + h.c \right) \tag{11}$$

Where a(b) labels sublattice 'A'('B'), and the operator ${a_i}^{\dagger}\,(a_i)$ creates(annihilates) an electron at $\vec{x}_{i,A}$. Similarly, for ${b_i}^{\dagger}\,(b_i)$. We assumed the onsite potential to be zero and only allowed nearest neighbor interactions, where 't' represents the nearest neighbor hopping amplitude. When formulating this Hamiltonian for a system with N sites, we obtain an NxN matrix with only off-diagonal entries. The Hamiltonian matrix will appear as follows:

$$H = \begin{pmatrix} a^{\dagger} & b^{\dagger} \end{pmatrix} \begin{pmatrix} 0 & -t \\ -t & 0 \end{pmatrix} \begin{pmatrix} a \\ b \end{pmatrix} \tag{12}$$

Where a and b are the sublattices.

Density of states(DOS) for monolayer graphene: It refers to the number of electronic states within the energy interval E to $E + \delta E$ that are available to be occupied by electrons. The DOS function g(E) provides information on how densely packed these states are at a given energy level E. It is a crucial concept in understanding the electronic properties of materials. Upon diagonalizing, the Hamiltonian will get the energy eigenvalues. Using this, we count the number of states at each energy E within a small energy window $(E–\delta E, E + \delta E)$ and plot the number with energy to get the DOS.

The density of states for monolayer graphene has shown below.

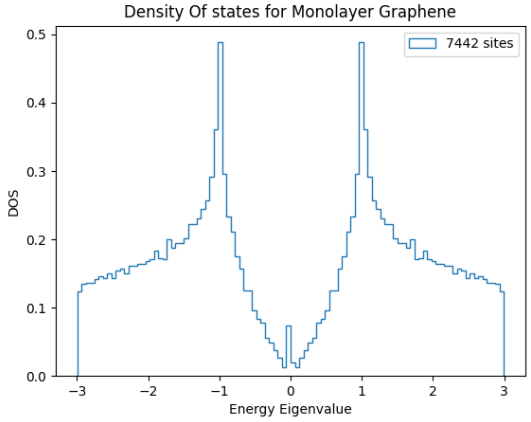

Figure 4: DOS vs Energy plot of Monolayer graphene with open boundaries.

3.2 Bilayer Graphene

Bilayer graphene consists of two monolayers stacked in either AA or AB (Bernal) configurations. In AA stacking, the carbon atoms of the top layer are directly aligned with those of the bottom layer.

On the other hand, AB stacking positions the carbon atoms of the top layer such that half of them align with the centers of the hexagons in the bottom layer, while the other half aligns with the carbon atoms below. The unit cell of bilayer graphene has four sub-lattices, two in the upper layer and two in the lower layer. We can name them 'A1', 'B1', and 'A2', 'B2'.

3.2.1 AA bilayer

Tight-binding Hamiltonian : Figure 5 illustrates that the 'A' sublattices of the upper layer are linked to the 'A' sublattices of the lower layer. The same applies to the 'B' sublattices. Consequently, the interlayer nearest neighbor hopping occurs between two layers, breaking the chiral symmetry while the chiral symmetry is preserved within the same layer. Below is the tight-binding Hamiltonian for the AA bilayer.

$$\hat{H} = -t\sum_{\alpha}\sum_{<i,j>} a^{\dagger}_{i,\alpha}b_{j,\alpha} + t_0\sum_{i}\left(a^{\dagger}_{i,1}a_{i,2} + b^{\dagger}_{i,1}b_{i,2}\right) + h.c \tag{13}$$

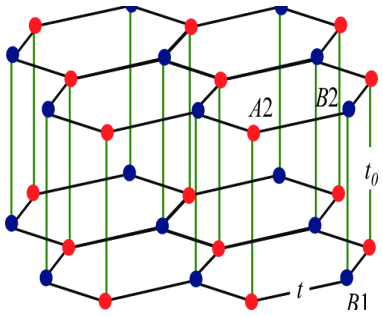

Figure 5: AA Bilayer structure[5]

Here 't_0' represents the interlayer hopping amplitude. For this study, we will assume that the interlayer hopping strength is equal to the intralayer hopping strength, i.e., $t_0 = t$. The variable 'α' denotes the layer index. As before, we have considered the onsite potential to be zero. The Hamiltonian matrix will appear as follows:

$$H = \begin{pmatrix} a_1{}^{\dagger} & a_2^{\dagger} & b_1{}^{\dagger} & b_2{}^{\dagger} \end{pmatrix} \begin{pmatrix} 0 & t_0 & -t & 0 \\ t_0 & 0 & 0 & -t \\ -t & 0 & 0 & t_0 \\ 0 & -t & t_0 & 0 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ b_1 \\ b_2 \end{pmatrix} \tag{14}$$

Where 'a1' and 'b1' are sublattices of the lower layer and 'a2' and 'b2' are the sublattices of the upper layer.

DOS of AA bilayer graphene: The graph for the DOS for the AA bilayer is shown below. We observe a significantly large number of states at zero energy, indicating a divergence at E=0 due to flat bands, which is not observed in other cases.

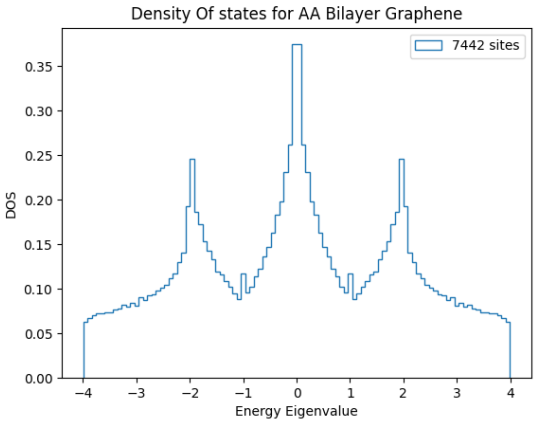

Figure 6: DOS vs Energy plot of AA bilayer graphene with open boundaries.

3.2.2 AB bilayer

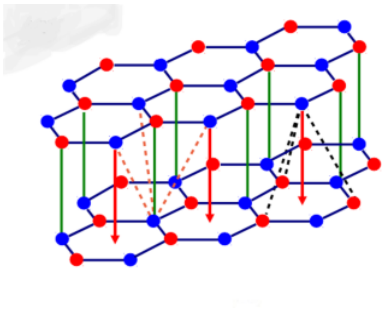

Figure 7: AB Bilayer structure[6]

Tight-binding hamiltonian: The tight-binding Hamiltonian remains the same for both configurations i.e., for AA and AB since the overlapping is identical. The tight-binding Hamiltonian is given as follows:

$$\hat{H} = -t\sum_{\alpha}\sum_{<i,j>} a^{\dagger}_{i,\alpha} b_{j,\alpha} + t_0 \sum_{<i,j>} \left(a^{\dagger}_{i,1} b_{2,j} + b^{\dagger}_{i,1} a_{2,j} \right) + h.c \tag{15}$$

Here, we observe that A-type sites are connected exclusively to B-type sites, thereby preserving the chiral symmetry. Also 't_0' represents interlayer hopping amplitude between A and B type sites while 't' is hopping amplitude within the layer. We will assume that interlayer and intralayer hoppings have equal strengths.
Furthermore, due to the onsite potential being zero, the Hamiltonian will result in an off-diagonal matrix form.

$$H = \begin{pmatrix} {a_1}^\dagger & a_2^\dagger & {b_1}^\dagger & {b_2}^\dagger \end{pmatrix} \begin{pmatrix} 0 & 0 & -t & 0 \\ 0 & 0 & t_0 & -t \\ -t & t_0 & 0 & 0 \\ 0 & -t & 0 & 0 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2 \\ b_1 \\ b_2 \end{pmatrix} \tag{16}$$

DOS of AB bilayer graphene: The graph for the DOS without disorder for the AB bilayer is shown below. We observe that the DOS exhibits a monolayer-like nature with two additional peaks.

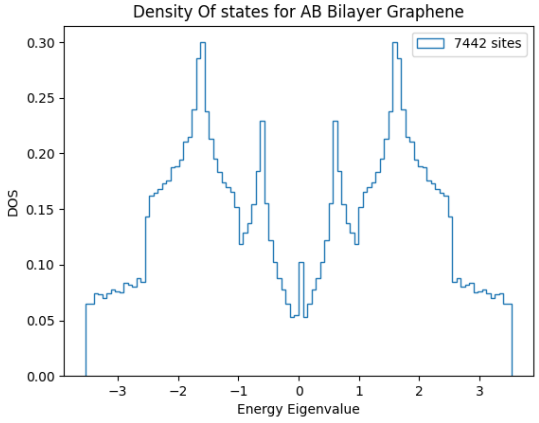

Figure 8: DOS vs Energy plot of AB bilayer graphene with open boundaries.

4 Conclusion

The analysis of the density of states (DOS) for monolayer AA and bilayer AB graphene under open boundary conditions without disorder reveals distinctive electronic characteristics inherent to each structure.
Overall, while both monolayer AA and bilayer AB graphene structures exhibit semimetallic behavior characterized by Dirac cones, the bilayer AB configuration introduces additional electronic states. These additional states arise from interlayer interactions, influencing the material's electronic properties and potentially enabling tailored applications in electronic and optoelectronic devices.
Understanding the density of states for these graphene configurations provides valuable insights into their electronic structures and guides future explorations into exploiting their unique properties for technological advancements.

References

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