

Research Article

Capacitance Variation of Electrolyte-Gated Bilayer Graphene Based Transistors

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Quantum capacitance of electrolyte-gated bilayer graphene field-effect transistors is investigated in this paper. Bilayer graphene has received huge attention due to the fact that an energy gap could be opened by chemical doping or by applying external perpendicular electric field. So, this extraordinary property can be exploited to use bilayer graphene as a channel in electrolyte-gated field-effect transistors. The quantum capacitance of bi-layer graphene with an equivalent circuit is presented, and also based on the analytical model a numerical solution is reported. We begin by modeling the DOS, followed by carrier concentration as a function V in degenerate and nondegenerate regimes. To further confirm this viewpoint, the presented analytical model is compared with experimental data, and acceptable agreement is reported.

1. Introduction

Recently, graphene, an atomic layer of carbon atoms arranged in a two-dimensional (2D) honeycomb lattice, has drawn researchers' attention due to its exceptional mechanical and electrical properties for new semiconductor materials and devices [1–3]. Bilayer graphene (BLG), the stacking of two-monolayer graphene, as a new material with outstanding electrical and physical properties holds the great promise to be used as a conducting channel in FETs [4, 5]. The energy dispersion relation of conductance and valence bands of each graphene layer is linearly touching on Dirac points [6]. As shown in Figure 1, the two layers are arranged in Bernal stacking which its lattice constant within a layer is given by $a = 0.246$ nm and the layer spacing by $d = 0.334$ nm. BLG is typically arranged in AA-stacked and AB-stacked arrangements. A bandgap in BLGs can be created by applying a perpendicular electric field and incorporating the inversion symmetry

breaking between double layers in the atomic structure. Two different stacking shapes of the BLG (AA, AB) in layers result from interlayer coupling effects in low energy, which shows a different band structure [7]. The study had shown that AA-stacked BLG is gapless metallic [8], whereas AB-stacked BLG is semiconducting with a bandgap, and the bandgap is tunable in the presence of external electric field perpendicular to the BLG [9].

Nowadays, electrolyte-gated bilayer graphene field-effect transistors (EGFETs) have caught much attention due to their advantages such as small size and the possibilities for mass production. Their short and consistent response times are very favorable to the electronics industry. EGFETs introduced new features such as the integration of compensation and data processing circuits in the same circuit for this type of sensors [10]. Recently, microelectronic advances have been exploited and applied to improve EGFETs fabrication. Because of the electrolyte ionic properties, electrical parts of EGFETs cannot

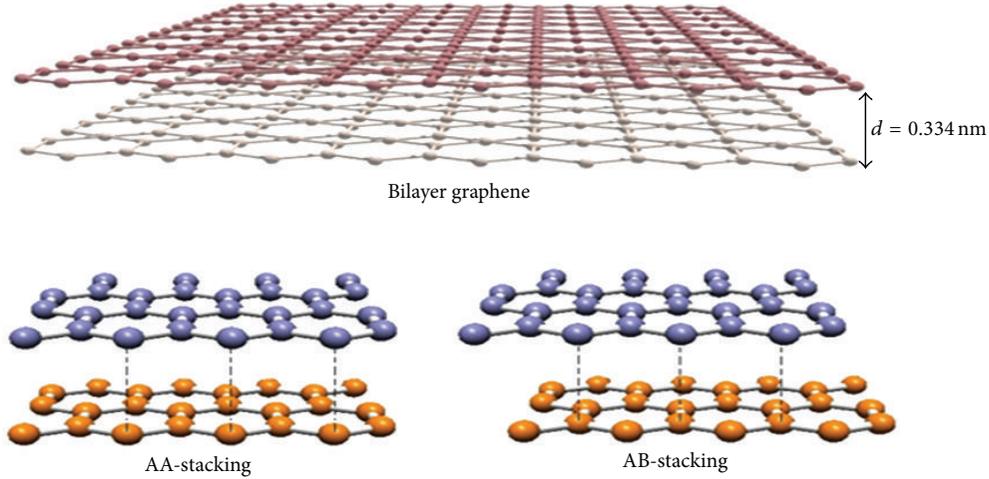


FIGURE 1: Schematic of bilayer graphene, AA- and AB-stacking.

have contact with liquid; only the gate area is open. EGFETs can be based on many materials as their detectors such as membrane and graphene [11]. On the other hand, in 2011, Ye et al. presented a comparative study of high carrier density transport in ion-gated mono-, bi-, and trilayer graphenes using electric double-layer transistors [12].

In this paper, EGFET has been employed to study the electron transport of graphene [13]. As shown in Figure 2, the bilayer graphene based electrolyte-gated transistor consists of source and drain (gold), in which an AB stacked bilayer graphene sheet which is employed as a conducting channel on an oxidized silicon/SiO₂ substrate is proposed. Therefore, a back gate controlled the current through the graphene. A photoresist layer as an insulator has been employed which results in the creation of a small window, which exposes the electrolyte [14]. The interfacial capacitance of the graphene was measured by the standard three-electrode electrochemical cell using a potentiostat, which the potential of graphene is controlled with respect to a reference electrode (a platinum electrode) [15]. By assuming that the source and substrate terminals are held at ground potential, the channel region has the characteristics of the resistor in small voltage between the source and drain (V_{DS}) [16]. It is notable that, by applying this configuration, the background capacitance can be minimized, and the graphene edges do not expose to the electrolyte. To control the gate voltage precisely, a platinum counter electrode is included to form the standard three-electrode electrochemical configuration.

The focus of this paper is to model the quantum capacitance of bilayer graphene based EGFET. To address this possibility, two interfacial capacitances which arise from the double layer formed by ions at the graphene ionic liquid interface and the quantum capacitance of graphene have a strong influence on the measurement of the total capacitance. To understand the electrical response of the bilayer graphene based EGFET device, an electrical equivalent circuit for the mentioned structure is developed and discussed in Figure 3.

It can be seen in Figure 3 that the equivalent circuit is composed of an ohmic resistance of a solution (R_s) in series with the double-layer capacitance and the quantum

capacitance of bilayer graphene. In order to present the accumulation of a layer of counterions on a charged electrode, $C_{\text{double layer}}$ is defined [17]. The capacitance versus potential of a double-layer graphene device was measured in ionic liquid in 1-butyl-3-methylimidazolium hexafluorophosphate (BMIMPF₆) which can be expressed as $C_{\text{double layer}} = \epsilon_0 \epsilon / t$ where $\epsilon_0 = 8.85 \times 10^{-12} \text{ Fm}^{-1}$ is the dielectric constant of the ionic liquid and it is the radius of the counterions [18]. For BMIM-PF₆ ionic liquid, $\epsilon \approx 7$ and $t \approx 0.3 \text{ nm}$, which leads to double-layer capacitance of $\approx 21 \mu\text{Fcm}^{-2}$ [16]. It could be stated that the quantum capacitance of the bilayer graphene based electrolyte-gated FETs can be modeled, holding the fact that the smaller of the two capacitances dominates the total capacitance [19]. On one hand, the double-layer capacitance can be neglected compared to the predicted quantum capacitance of bilayer graphene, while, on the other hand, the double-layer capacitance is not strongly dependent on the potential, making it straightforward to determine the quantum capacitance of bilayer graphene. In 2009, the quantum capacitance of bilayer graphene in an ionic liquid electrolyte was measured by Xia et al. [18]. He found out that in his measurements Debye ionic screening length of the ionic liquid is virtually zero which makes the quantum capacitance a dominant source of the measured capacitance. The aim of this study is to evaluate the quantum capacitance of bilayer graphene sheet as a function of voltage and validate theoretical predictions with the experimental results [18].

2. Proposed Model

An important quantity in the design of nanoscale devices is the quantum capacitance. To model a theoretical prediction of quantum capacitance for ideal bilayer graphene based electrolyte-gated FETs, the expression for quantum capacitance is used [20] as follows:

$$C = \frac{\partial Q}{\partial V}, \quad (1)$$

where $\partial Q = e \cdot \partial n$ is the charge measured in coulombs, e is the charge of the electron, n is the BLG's intrinsic carrier

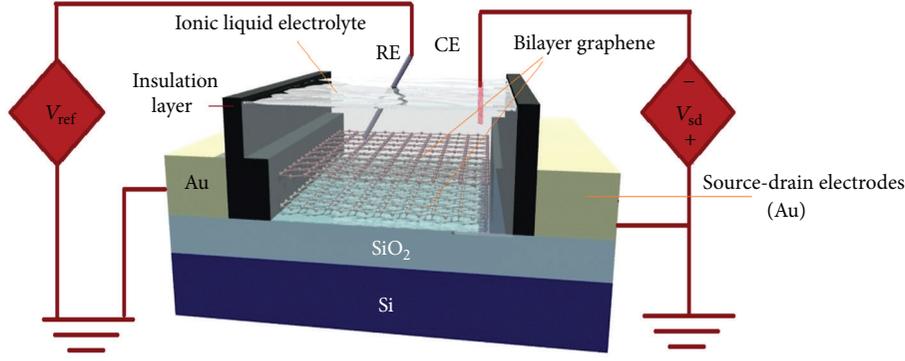


FIGURE 2: Schematics of the proposed structure and the electrical circuit of the bilayer graphene based EGFET.

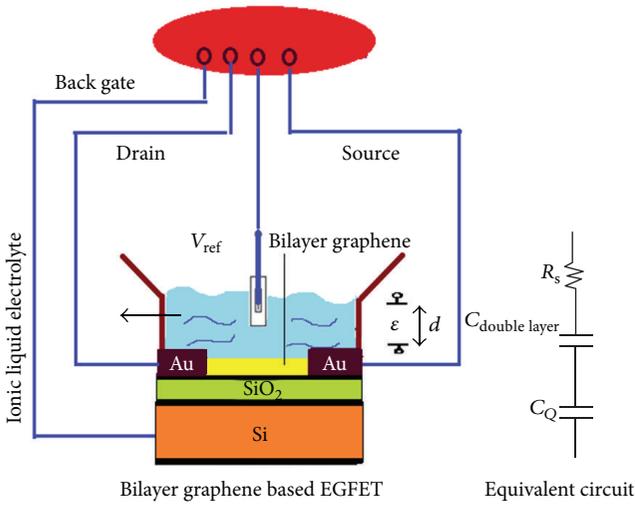


FIGURE 3: Equivalent circuit of bilayer graphene based electrolyte-gated field-effect transistor.

concentration, and $\partial V = \partial E/e$ is the voltage applied to the devices, where E is energy. By the substitution of the quantum capacitance model, (2) can be expressed as

$$C = e^2 \frac{\partial n}{\partial E}. \quad (2)$$

According to (2), the carrier concentration and the energy dispersion of BLG is necessary to be calculated for modeling the quantum capacitance. Understanding the electronic structure of bilayer graphene starts with looking at its band structure. It has been shown that the bandgap of BGLs can be varied by means of an external perpendicular electric field and induced significant bandgap between the valence and conduction bands from a zero-gap semiconductor to an insulator [21–24]. The energy band structure of AB which is stacking biased BGL using tight-binding method has been studied in [25]. Equation (3) illustrates energy (E) and biased

voltage (V) relationship of BGL. The energy dispersion of BLG is expressed as

$$E(k) = \frac{V_1 + V_2}{2} \pm \sqrt{\varepsilon_k^2 + \frac{V^2}{4} + \frac{t_{\perp}^2}{2} \pm \frac{1}{2} \sqrt{4(V^2 + t_{\perp}^2)\varepsilon_k^2 + t_{\perp}^4}}, \quad (3)$$

where $\varepsilon_k^2 = (v^4/(4 + v^2 t_{\perp}^2/2))/(v^2 + t_{\perp}^2)$ is electron's dispersion in monolayer graphene [26], $t_{\perp} = 0.35$ eV is an interlayer hopping energy [6], V_1 and V_2 are the potential energy of first and second layers, respectively, and V is the potential energy difference between first and second layers ($V = V_1 - V_2$). By using the energy dispersion of BLG, density of state (DOS) as a fundamental parameter of BLG indicates available energy states that can be defined as

$$\text{DOS} = \left(\frac{\hbar^2 (k - k_g)}{m^* k} 2\pi \right)^{-1}, \quad (4)$$

where \hbar is the reduced Planck's constant and m^* is the effective mass of electron in the BLG. k_g is defined as $k_g = (V/2v_F\hbar)\sqrt{(V^2 + 2t_{\perp}^2)/(V^2 + t_{\perp}^2)}$ where $v_F = (\sqrt{2}/3)$ at $\hbar \approx 1 \times 10^6$ m · s⁻¹ is the Fermi Velocity. k is defined as $k = \pm[2m^*(E - E_c)/\hbar^2]^{1/2} + k_g$ where $E_c = E_g/2 + (V_1 + V_2)/2$ and E_g is the energy gap. DOS indicates available energy states. It is notable that electrical property of materials from metallic to semiconducting is changing by the gradient of DOS near the Dirac point. In the next step, the carrier concentration of BGL is given by

$$n = \int \text{DOS} \cdot f(E) dE, \quad (5)$$

where $f(E) = 1/(1 + e^{(E-E_f)/k_B T})$ is the Fermi-Dirac distribution function which gives the probability of occupation of a state at any energy level. In this function, E_f is the Fermi level, k_B is Boltzmann's Constant, and T is the temperature. Using DOS calculated from (4), the carrier concentration reads

$$n = \frac{m^*}{\hbar^2 2\pi} \int_0^{+\infty} \frac{k}{k'} \frac{1}{1 + e^{(E-E_f)/k_B T}} dE, \quad (6)$$

where $k' = (k - k_g)$. As depicted in Figure 4, the carrier concentration is plotted for different values of V .

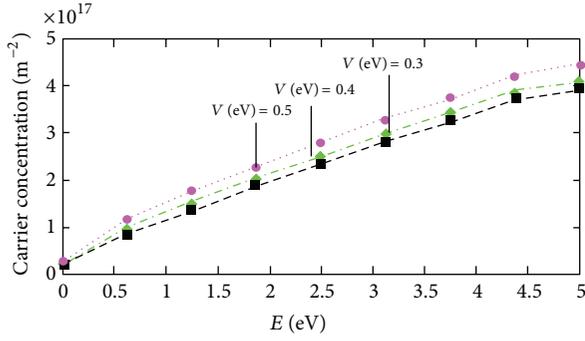


FIGURE 4: Carrier concentration versus energy (E).

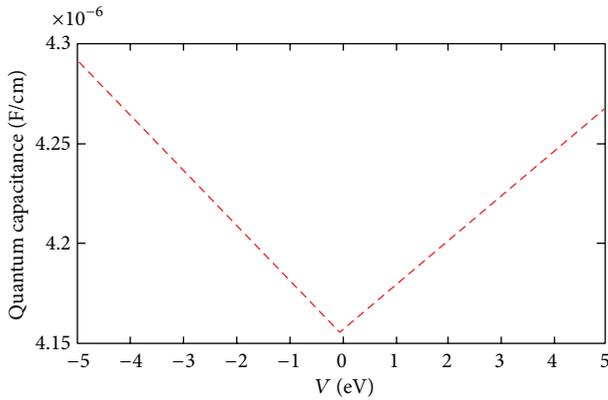


FIGURE 5: The quantum capacitance of bilayer graphene based electrolyte-gated FETs.

It is apparently seen that the calculated carrier concentration model points out the strong dependence of voltage showing that the voltage increment effect will influence the carrier concentration. Substituting the carrier concentration in (2), the quantum capacitance of BLG is expressed as

$$C = \frac{q^2 m^* k}{\hbar^2 2\pi k'} \frac{1}{1 + e^{(E-E_f)/k_B T}}. \quad (7)$$

Equation (7) provides a quantitative description of the quantum capacitance of bilayer graphene in terms of Fermi velocity [27], carrier density, temperature, and fundamental physical quantities. According to the relation between energy band structure and the graphene potential, the quantum capacitance-voltage characteristic of bilayer graphene is depicted as shown in Figure 5.

The overall quantum capacitance versus gate potential for the bilayer graphene is similar to that of a single layer graphene. To get a greater insight into the quantum capacitance of bilayer graphene based electrolyte-gated FETs devices, several outstanding features of the C - V characteristics are embarked. First of all, the quantum capacitance possesses a minimum value at the Dirac point which is close to zero. On the other hand, the linear increase of the capacitance with a voltage which is symmetric with respect to the Dirac point can be considered as the other perceptible feature of the quantum capacitance model. In order to validate the proposed model,

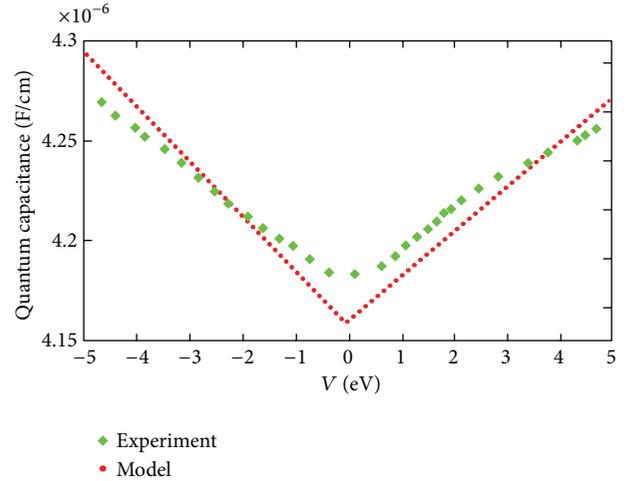


FIGURE 6: Comparison between bilayer graphene quantum capacitance model with experimental extracted data.

the comparison between the quantum capacitance model of bilayer graphene based electrolyte-gated FETs with extracted experiment data is done as illustrated in Figure 6 [18].

It is noteworthy that the theoretical slope is greater than the measured slope for bilayer graphene. A possible explanation for this might be that the suggested model is based on the pure and ideal bilayer graphene in which, in reality, various impurities and defects exist. It seems possible that these results are due to the fact that the impurities can drastically lower the slope for bilayer graphene. However, unlike single layer graphene, a theory for treating impurities in bilayer graphene has not yet been developed [18]. These findings further support the idea of the effects of impurities on both the first and second layers, which requires a self-consistent theory to include the screening of the impurity field by the carriers in both layers. This is further complicated by the possibility of impurities intercalated between the two layers. Further experimental studies should be done to investigate the quantitative understanding of bilayer graphene. It is apparently seen that there is a favorable agreement between the proposed model of bilayer graphene based electrolyte-gated FETs and experimental results. It can be concluded that the presented model can be applied as a powerful tool to optimize the bilayer graphene based electrolyte-gated FETs performance.

3. Conclusion

BLGs can be employed in digital electronics because their bandgap can be varied by means of an external perpendicular electric field and can induce a significant bandgap between the valence and conduction bands turning it from a zero-gap semiconductor to an insulator. Carrier mobility values as high as $200,000 \text{ cm}^2 (\text{Vs})^{-1}$ (200 times higher than in silicon) can be achieved by graphene, which are increased by increasing carrier density [22]. According to the graphene structure, it can satisfy our major requirement of a channel in electrolyte-gated FETs due to its large surface-to-volume ratio, high conductivity, high mobility, and strong mechanical and elasticity properties. The aim of this paper was to model the

quantum capacitance of bilayer graphene based electrolyte-gated FETs with an equivalent circuit of the proposed device. For purposes of verification, a comparison study between the model and experimental data was done and notable agreement is reported which shows that bilayer graphene based electrolyte-gated FETs model can be used to predict bilayer graphene behavior in graphene based devices.

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