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GATE-TUNABLE BANDGAP IN BILAYER GRAPHENE

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The tight-binding model of bilayer graphene is used to find the gap between the conduction and valence bands, as a function of both the gate voltage and the doping by donors or acceptors. The total Hartree energy is minimized and an equation for the gap is obtained. This equation for the ratio of the gap to the chemical potential is determined only by the screening constant. Therefore, the gap is strictly proportional to the gate voltage or the carrier concentration in the absence of donors or acceptors. But in the case where the donors or acceptors are present, the gap demonstrates an asymmetric behavior on the electron and hole sides of the gate bias. A comparison with experimental data obtained by Kuzmenko et al. demonstrates a good agreement.

1. INTRODUCTION

Bilayer graphene has attracted much interest partly due to the opening of a tunable gap in its electronic spectrum by an external electrostatic field. Such a phenomenon was predicted in Refs. [1, 2] and can be observed in optical studies controlled by applying a gate bias [3–10]. In Refs. [11, 12], within the self-consistent Hartree approximation, the gap was derived as a near-linear function of the carrier concentration injected in the bilayer by the gate bias. Recently, this problem was numerically considered in Ref. [13] using the density functional theory (DFT) and including the external charge doping due to impurities. The DFT calculation gives the gap that is roughly half the gap obtained in the Hartree approximation. This disagreement was explained in Ref. [13] as a result of both the inter- and intralayer correlations.

In this work, we study this problem within the same Hartree approximation as in Refs. [11, 12], but including the effect of external doping. We calculate the carrier concentration on both sides of the bilayer in the case where the carrier concentration in the bilayer is less than 10^{13} cm^{-2}. We then minimize the total energy of the system and self-consistently find both the chemical potential and the gap induced by the gate bias. Our results completely differ from those in Refs. [11, 12], where the external doping is disregarded. In the presence of dopants, the dependence of the gap on the carrier concentration, i. e., on the gate voltage, exhibits an asymmetry at the electron and hole sides of the gate bias.

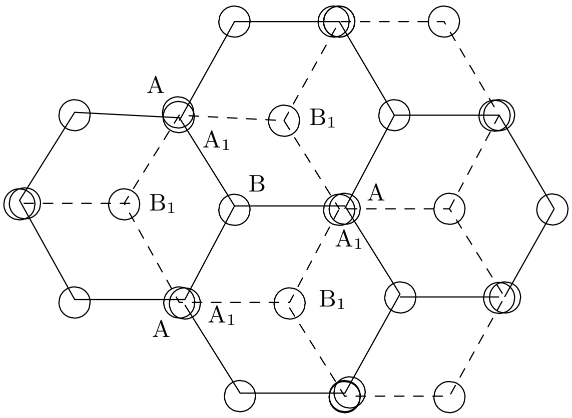

Fig. 1. Bilayer lattice

2. TIGHT-BINDING MODEL OF BILAYER GRAPHENE

The graphene bilayer lattice is shown in Fig. 1. Atoms in one layer, i. e., A and B in the unit cell, are connected by solid lines, and in the other layer, e. g.,

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Table. Parameter values of the electron spectrum in electronvolts

Parameter	Experiment [10]	DFT calculation [16]
γ_0	3.16 ± 0.30	2.598 ± 0.015
γ_1	0.381 ± 0.003	0.34 ± 0.02
γ_3	0.38 ± 0.06	0.32 ± 0.02
γ_4	0.14 ± 0.03	0.177 ± 0.025
Δ	0.011 ± 0.002	0.012 ± 0.05

A_1 and B_1, by dashed lines. An atom A (A_1) differs from B (B_1) because it has a neighbor just below it in the adjacent layer, whereas the atom B (B_1) does not.

We recall the main results of the Slonchewski–Weiss–McClure model [14, 15]. In the tight-binding model, the Bloch functions of the bilayer are written as

$$\begin{gathered}
\psi_a = \frac{1}{\sqrt{N}} \sum_j \exp(i\mathbf{k} \cdot \mathbf{a}_j) \psi_0(\mathbf{a}_j - \mathbf{r}), \\
\psi_b = \frac{1}{\sqrt{N}} \sum_j \exp\left(i\mathbf{k} \cdot \mathbf{a}_j\right) \psi_0(\mathbf{a}_j + \mathbf{a} - \mathbf{r}), \\
\psi_{a1} = \frac{1}{\sqrt{N}} \sum_j \exp(i\mathbf{k} \cdot \mathbf{a}_j) \psi_0(\mathbf{a}_j + \mathbf{c} - \mathbf{r}), \\
\psi_{b1} = \frac{1}{\sqrt{N}} \sum_j \exp(i\mathbf{k} \cdot \mathbf{a}_j) \psi_0(\mathbf{a}_j + \mathbf{c} + \mathbf{a} - \mathbf{r}),
\end{gathered} \tag{1}$$

where the sums are taken over the lattice vectors $\mathbf{a}_j$ and N is the number of unit cells. The vectors $\mathbf{a}$ and $\mathbf{c}$ respectively connect the nearest atoms in the layer and in the neighbor layers.

For the nearest neighbors, the effective Hamiltonian in the space of functions (1) can be written as

$$H(\mathbf{k}) = \\ = \begin{pmatrix} U+\Delta & \gamma_0 f^* & \gamma_1 & \gamma_4 f \\ \gamma_0 f & U-\Delta & \gamma_4 f & \gamma_3 f^* \\ \gamma_1 & \gamma_4 f^* & -U+\Delta & \gamma_0 f \\ \gamma_4 f^* & \gamma_3 f & \gamma_0 f^* & -U-\Delta \end{pmatrix}, \tag{2}$$

where

$$f = \gamma_0 \left[\exp(ik_x a) + 2\exp(-ik_x a/2) \cos\left(k_y a\sqrt{3}/2\right) \right].$$

The values of hopping integrals γ_0, γ_1, γ_3, γ_4, and Δ are given in the Table. The largest of them, γ_0, determines the band dispersion near the K point in the Brillouin zone, where the matrix element $\gamma_0 f$ can be expanded as

$$\gamma_0 f = v(ik_x - k_y),$$

with a velocity parameter $v = 3\gamma_0 a/2$. The parameters γ_3 and γ_4 giving a correction to the dispersion are less than γ_0 by a factor of 10. The parameters γ_1 and Δ result in the position of levels at K, but Δ is much less than γ_1. In addition, the parameter U induced by the gate voltage is associated with the asymmetry of two layers in the external electrostatic field. This parameter plays a role of the potential energy $-e\,dE$ between two layers, where d is the interlayer distance and E is electric field induced both by the gate voltage and the external dopants in the bilayer.

The parameter U and the chemical potential μ should be self-consistently calculated for the given gate voltage. For this, we can keep only the parameters γ_0 and γ_1, neglecting the small effect of γ_3, γ_4, and Δ on the gap U. In this approximation, the effective Hamiltonian can be written in the simple form

$$H(\mathbf{k}) = \begin{pmatrix} U & vk_+ & \gamma_1 & 0 \\ vk_- & U & 0 & 0 \\ \gamma_1 & 0 & -U & vk_- \\ 0 & 0 & vk_+ & -U \end{pmatrix}, \tag{3}$$

where $k_\pm = \mp ik_x - k_y$ in the vicinity of the K points. The Hamiltonian gives four energy bands:

$$\begin{gathered}
\varepsilon_{1,4}(q) = \pm\left(\frac{\gamma_1^2}{2} + U^2 + q^2 + W\right)^{1/2}, \\
\varepsilon_{2,3}(q) = \pm\left(\frac{\gamma_1^2}{2} + U^2 + q^2 - W\right)^{1/2},
\end{gathered} \tag{4}$$

where

$$W = \left(\frac{\gamma_1^4}{4} + (\gamma_1^2 + 4U^2)q^2\right)^{1/2}$$

and we set $q^2 = (vk)^2$.

The band structure is shown in Fig. 2. The minimal value of the upper energy ε_1 is $\sqrt{U^2 + \gamma_1^2}$. The ε_2 band takes the maximal value $|U|$ at $q = 0$ and the minimal value $\tilde{U} = \gamma_1|U|/\sqrt{\gamma_1^2 + 4U^2}$ at $q^2 = 2U^2(\gamma_1^2 + 2U^2)/(\gamma_1^2 + 4U^2)$. Because the value of U is much less than γ_1, the distinction between U and $\tilde{U}$ is small and the gap between the bands ε_2 and ε_3 approximately takes the value $2|U|$.

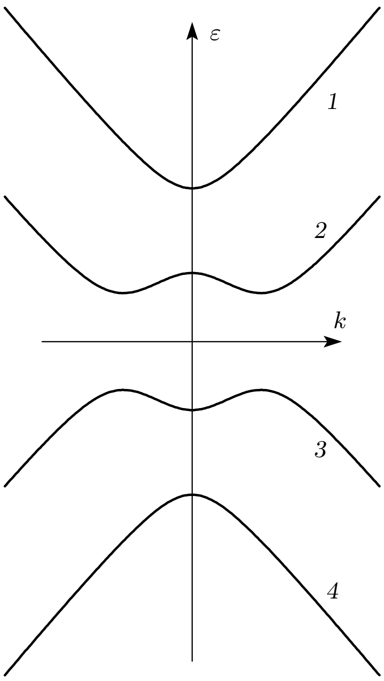

Fig. 2. Band structure of a bilayer

3. EIGENFUNCTIONS AND CARRIER CONCENTRATION

The four eigenfunctions **C** corresponding to eigenvalues (4) of Hamiltonian (3) are given by

$$\mathbf{C} = \frac{1}{C}\begin{pmatrix} (U-\varepsilon_n)[(\varepsilon_n+U)^2-q^2] \\ -q_-[(\varepsilon_n+U)^2-q^2] \\ \gamma_1(U^2-\varepsilon_n^2) \\ \gamma_1 q_+(U-\varepsilon_n) \end{pmatrix}, \qquad (5)$$

where the **C** norm squared is

$$C^2 = [(\varepsilon_n+U)^2-q^2]^2[(\varepsilon_n-U)^2+q^2] + \\ + \gamma_1^2(\varepsilon_n-U)^2[(\varepsilon_n+U)^2+q^2] .$$

It follows from Eqs. (1) that the probability p_1 to find an electron, for instance, on the first layer is

$$p_1 = |C_1|^2 + |C_2|^2 ,$$

where the subscript in C_i numerates the elements of column (5).

We assume that carriers occupy only the bands $\varepsilon_{2,3}$, so the chemical potential μ and the gap $2|U|$ are less than the distance between the bands ε_1 and ε_2, i.e., $(|\mu|, 2|U|) < \gamma_1$. The electron dispersion for the $\varepsilon_{2,3}$ bands can be expanded in powers of q^2:

$$\varepsilon_n^2(q) = U^2 - 4\frac{U^2}{\gamma_1^2}q^2 + \frac{q^4}{\gamma_1^2} , \qquad (6)$$

where $n = 2$ stands for the electron conductivity and $n = 3$ for the hole conductivity. Then, for $q^2 \gg 4U^2$, we can omit the second term here and use the simple relations

$$q^2 = \gamma_1(\varepsilon_n^2 - U^2)^{1/2}, \qquad (7)$$

neglecting the small effect of the "mexican hat".

Keeping only the leading terms, we find from Eq. (5) that the probabilities $p_{1,2}$ to find an electron on the layers are

$$p_1 = |C_1|^2 + |C_2|^2 \propto q^6 = \gamma_1^3(\varepsilon_n^2 - U^2)^{3/2} ,$$

$$p_2 = |C_3|^2 + |C_4|^2 \propto q^2\gamma_1^2(U-\varepsilon_n)^2 = \\ = \gamma_1^3(\varepsilon_n^2-U^2)^{1/2}(U-\varepsilon_n)^2 .$$

Therefore, the normalized probability to find an electron, for instance, on the first layer can be written as

$$p_1 = \frac{(\varepsilon_n^2-U^2)^{3/2}}{(\varepsilon_n^2-U^2)^{3/2} + (\varepsilon_n^2-U^2)^{1/2}(U-\varepsilon_n)} = \\ = (\varepsilon_n + U)/2\varepsilon_n . \qquad (8)$$

Within the approximation in (7)–(8), many observable effects can be evaluated analytically for the intermediate carrier concentration, $4U^2 \ll \gamma_1\sqrt{\mu^2-U^2} \ll \gamma_1^2$.

At zero temperature, the carrier concentration on the sides of the bilayer is found with the help of Eq. (8) as

$$n_{1,2} = \frac{2}{\pi\hbar^2 v^2}\int p_{1,2}\, q\, dq = \\ = \frac{n_0 U}{2\gamma_1}\left[\sqrt{x^2-1} \pm \ln\left(x + \sqrt{x^2-1}\right)\right] , \qquad (9)$$

where the integration limits are given by $q = 0$ and the chemical potential μ, and we set

$$n_0 = \gamma_1^2/\pi\hbar^2v^2 = 1.03\cdot 10^{13}\ \text{cm}^{-2} , \\ x = |\mu/U|. \qquad (10)$$

For the total carrier concentration n in the bilayer, we obtain

$$n = \frac{\gamma_1}{\pi\hbar^2v^2}\sqrt{\mu^2-U^2} = \frac{n_0 U}{\gamma_1}\sqrt{x^2-1} . \qquad (11)$$

4. MINIMIZATION OF THE TOTAL ENERGY

To find the chemical potential μ and the gap $2|U|$ at the given gate voltage

$$eV_g = -e\,dE - e\,d_w E_w\,, \tag{12}$$

where d is the interlayer distance and d_w is the wafer thickness, we minimize the total energy containing both the energy $V^{(c)}$ of the carriers and the energy $V^{(f)}$ of the electrostatic field. Within the Hartree approximation, when no electron correlations are taken into account, the filled bands do not contribute to the energy of the system, because the electron charge of the filled bands is compensated by the ion charge and this energy has to be regarded as the ground state energy. The excitation energy owes its origin to the carriers in unfilled bands. Electrons in the ε_2 band or holes in the ε_3 band contribute the energy

$$V^{(c)} = \frac{2}{\pi\hbar^2 v^2}\int |\varepsilon_n(q)|q\,dq = \\ = \frac{n_0 U^2}{2\gamma_1}\left[x\sqrt{x^2-1} + \ln\left(x+\sqrt{x^2-1}\right)\right] \tag{13}$$

to the total energy of the system. The energy of the electrostatic field (Fig. 3)

$$V^{(f)} = \frac{1}{8\pi}(dE^2 + \epsilon_w d_w E_w^2) \tag{14}$$

can be written in terms of the carrier concentrations with the help of relations

$$4\pi e(n_1 - N_1) = E, \quad 4\pi e(n - N) = \epsilon_w E_w\,, \tag{15}$$

where ϵ_w is the dielectric constant of the wafer, and N_1 and N_2 are the respective concentrations of the acceptor or donor impurities on the left and right layers, with the total dopant concentration on the bilayer being $N = N_1 + N_2$. All these numbers are supposed to be positive or negative for the electron or hole doping respectively. We emphasize that the dielectric constant ϵ of bilayer graphene depends on the substrate. For simplicity, we put $\epsilon = 1$ in definition (14).

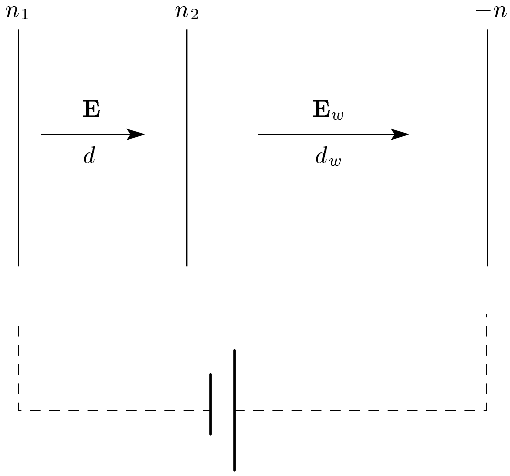

Fig. 3. Electrostatic model; d is the interlayer distance and d_w is the wafer thickness

We seek the minimum of the total energy

$$V^{(f)} + V^{(c)} + \lambda(eV_g + edE + ed_w E_w)$$

as a function of U, μ, and the Lagrange multiplier λ. Differentiation with respect to λ gives the gate voltage constraint (12). Minimization with respect to U and x gives

$$4\pi e^2\left[(n_1 - N_1)n_{1u}d + (n - N)n_u d_w/\epsilon_w\right] + \\ + V_u^{(c)} + 4\pi e^2\lambda(n_{1u}d + n_u d_w/\epsilon_w) = 0$$

and a similar equation with the substitution $u \to x$, where the subscripts u and x denote the derivatives with respect to the corresponding variables. The Lagrange multiplier λ can be eliminated from these two equations. Then, the equation obtained should be expanded in d/d_w, since the thickness d of the bilayer is much less than the thickness d_w of the substrate.

Thus, we obtain the equation

$$4\pi e^2 d\,(n_2 - N_2)\left(\frac{n_{1x}}{n_x} - \frac{n_{1u}}{n_u}\right) = \frac{V_x^{(c)}}{n_x} - \frac{V_u^{(c)}}{n_u}\,. \tag{16}$$

We emphasize that this equation is invariant under the simultaneous sign change in $n_{1,2}$ and $N_{1,2}$, which expresses the charge invariance of the problem. At the fixed external doping $N_{1,2}$, the gap on the electron and hole sides of the gate bias is not symmetric.

The derivatives in Eq. (16) are calculated with the help of Eqs. (9)–(15). As a result, Eq. (16) becomes

$$2\frac{\gamma_1 N_2}{Un_0} = \sqrt{x^2-1} \pm \\ \pm\left\{f(x) + \frac{xf(x)}{\Lambda[xf(x) - \sqrt{x^2-1}]}\right\} \tag{17}$$

with the function $f(x) = \ln\left(x + \sqrt{x^2-1}\right)$ and the dimensionless screening constant

$$\Lambda = \frac{e^2\gamma_1 d}{(\hbar v)^2}\,. \tag{18}$$

For the parameters of graphene $d = 3.35\,\text{Å}$, $\gamma_1 = 0.381\,\text{eV}$, and $v = 1.02{\cdot}10^8\ \text{cm/s}$, we obtain $\Lambda = 0.41$.

5. THE GAP IN UNDOPED AND DOPED BASED BILAYER

5.1. Undoped bilayer

We first consider an ideal undoped bilayer with $N_1 = N_2 = 0$. We obtain a nonzero solution for U if the right-hand side of Eq. (17) vanishes. This condition is fulfilled only for the minus sign in Eq. (17), which defines the polarity of the layers (see Eq. (9)). We obtain the solution $x = x_0 = 6.61$. According to Eq. (11), the gap as a function of the carrier concentration takes a very simple form:

$$2\left|\frac{U}{n}\right| = \frac{2\gamma_1}{n_0\sqrt{x_0^2-1}} = 1.13\cdot 10^{-11}\ \text{meV}\cdot\text{cm}^2\,, \qquad (19)$$

where the right-hand side does not depend at all on the gate bias, but depends only on the screening constant Λ. This dependence is shown in Fig. 4 in dashed lines; it is symmetric on the electron and hole sides.

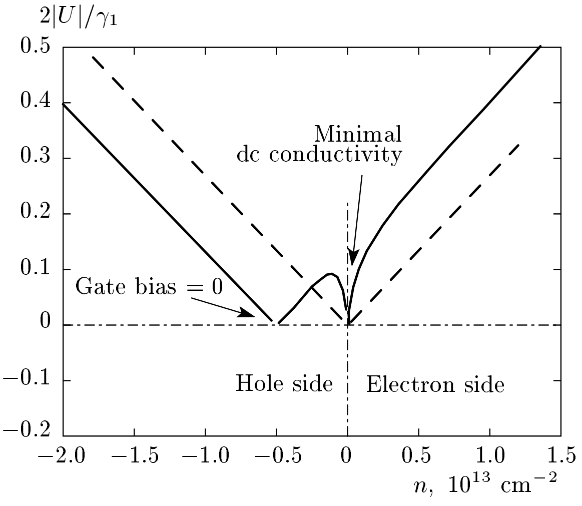

Fig. 4. The gap in units of $\gamma_1 = 0.381$ eV versus the carrier concentration in the absence of doping (dashed line) and for the hole doping level $N_2 = -2.5\cdot 10^{12}$ cm^{-2} (solid line); the positive (negative) values of n correspond to the electron (hole) conductivity. The difference between values of n marked as "gate bias = 0" and "minimal dc conductivity" is $2N_2$

With the help of Eq. (10), we obtain the chemical potential as a linear function of the carrier concentration:

$$\mu = \frac{\gamma_1 x_0}{n_0\sqrt{x_0^2-1}}\,n\,, \qquad (20)$$

where n is positive (negative) for the electron (hole) conductivity.

We can compare Eq. (19) with the corresponding result in Ref. [11]:

$$2\left|\frac{U}{n}\right| = \frac{e^2 d}{2\epsilon_0}\left[1 + 2\Lambda\frac{|n|}{n_0} + \Lambda\ln\frac{n_0}{|n|}\right]^{-1}. \qquad (21)$$

Both equations give approximately the same results at $|n| \approx 0.1 n_0 \approx 10^{12}\ \text{cm}^{-2}$. But unlike Eq. (19), Eq. (21) contains the carrier concentration in the right-hand side, giving rise to a more rapid increase in the gap with $|n| \ll n_0$. This increase also contradicts the DFT calculations [13].

Two reasons can result in the disagreement of our theory with Ref. [11]. First, in Ref. [11], the filled bands are supposed to create the electric field in the bilayer, which is incorrect, as was explained in the previous section. Second, the minimization should be done with respect to two variables μ and U, but only one of them seems to be used in Ref. [11].

5.2. Doped bilayer

For a bilayer with acceptor or donor dopants, Eq. (17) has the solution $w = 2\gamma_1 N_2/U n_0$ as a function of x. We obtain, evidently, small values of w for x close to $x_0 = 6.61$. Since $x_0 \gg 1$, we can expand the function in the right-hand of Eq. (17) in $1/x$. In this region of relatively large $|U|$, Eqs. (11) and (17) again yield the linear dependence

$$\begin{aligned} 2|U| &= |n - 2N_2|\frac{2\gamma_1}{n_0 x_0} = \\ &= 1.13\,|n - 2N_2|\cdot 10^{-11}\ \ \text{meV}\cdot\text{cm}^2. \qquad (22)\end{aligned}$$

The value of the carrier concentration $n = 2N_2$ corresponds to the zero bias voltage, where $U = 0$ (see Fig. 4). Therefore, in contrast to the undoped case, the gap demonstrates an asymmetric behavior on the electron and hole sides. If the bilayer contains acceptors with a concentration N_2, the gap decreases linearly with the hole concentration and vanishes when the gate bias is not applied and the hole concentration equals $2N_2$ ($-5\cdot 10^{12}\ \text{cm}^{-2}$ in Fig. 4). Starting from this point, the gap increases and, thereafter, becomes small again (equal to zero in Fig. 4) at the carrier concentration corresponding to the minimal value of the dc conductivity, where $n = 0$. Therefore, the difference

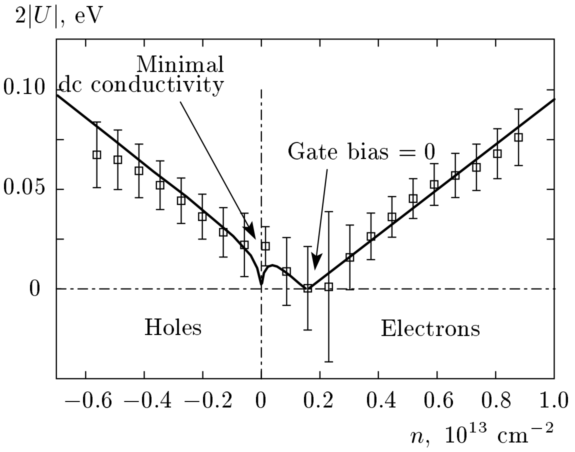

Fig. 5. The gap in electronvolts versus the carrier concentration for the electron doping with the concentration $N_2 = 0.78 \cdot 10^{12}$ cm^{-2} (our theory); the positive (negative) values of n correspond to the electron (hole) conductivity; squares are experimental data [10]

($1.56 \cdot 10^{12}$ cm^{-2} in Fig. 5) observed in Refs. [9, 10] between these two values of carrier concentrations, at the zero bias and at the minimal conductivity, directly gives the donor/acceptor concentration $2N_2$ on the layer close to the substrate. Then, for the gate bias applied in order to increase the electron concentration, the gap rapidly opens as electrons appear.

We see that an asymmetry arises between the electron and hole sides of the gate bias. This asymmetry can simulate a result of the hopping integral Δ in the electron spectrum [17]. To obtain the gap dependence in the case of electron doping, $N_2 > 0$, the reflection transformation $n \to -n$ has to be made. This case is shown in Fig. 5, where the experimental data from Ref. [10] are displayed.

The gap in the vicinity of the minimal conductivity value could indeed reach a finite value due to several reasons. One of them is the form of the "mexican hat" shown in Fig. 2. Second, the trigonal warping is substantial at low carrier concentrations. Finally, the graphene electron spectrum is unstable with respect to the Coulomb interaction at low momentum values. For a graphene monolayer, as shown in Ref. [18], logarithmic corrections occur at small momenta. In the case of a bilayer, the electron self-energy contains linear corrections, as can be found using the perturbation theory. Similar linear terms resulting in a nematic order were also obtained in the framework of the renormalization group [19].

6. CONCLUSION

The gap opening in the gated graphene bilayer has an intriguing behavior as a function of carrier concentration. In the presence of an external doping charge, i. e., donors or acceptors, this function is asymmetric on the hole and electron sides of the gate bias and it is linear only for a large gate bias. The difference between two values of carrier concentrations, i. e., at the zero bias and at the minimal conductivity, directly gives the sign and concentration of the charged dopants on the bilayer.

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