Comment on "Screening in gated bilayer graphene"

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We analyze the response of bilayer graphene to an external transverse electric field using a variational method. A previous attempt to do so in a recent paper by Falkovsky [Phys. Rev. B **80**, 113413 (2009)] is shown to be flawed. Our calculation reaffirms the original results obtained by one of us [E. McCann, Phys. Rev. B **74**, 161403(R) (2006)] by a different method. Finally, we generalize these original results to describe a dual-gated bilayer graphene device.

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The physics of monolayer and bilayer graphenes (BLG) has been a subject of much interest recently.[1] A unique feature of BLG is its tunable band structure: its band gap depends on the external electric field, which can be controlled by doping or gating. This effect was first analyzed theoretically[2–4] and recently studied experimentally.[3,5–18]

The subject of this Comment is the value of the gap at the Brillouin zone (BZ) corners, which we denote by $2|U|$. Within the conventional mean-field Hartree theory,[2,3] $2U$ coincides with the electrostatic energy difference per electron in the two layers,

$$2U = eE_m d_m, \quad d_m = 0.33 \text{ nm}, \tag{1}$$

where E_m is the component of the electric field inside the BLG directed from the bottom to the top layer and d_m is the interlayer spacing. In experimental practice, this field depends not only on the external gate voltages but also on the induced electron densities n_t and n_b of the layers, see Fig. 1. For a given chemical potential μ, these densities, and thus the total electron density of BLG $n=n_t+n_b$ are nonlinear functions of U. In general, they can be calculated only numerically.[3,4,19–23] However, in a range $|U| \ll |\mu| \ll \gamma_1$ an asymptotic analytical formula for the interlayer bias was derived by one of us[4]

$$2U \simeq \gamma_1 \frac{N - 2N_{dt}}{\Lambda_{\mathrm{Mc}}^{-1} + |N| - \frac{1}{2}\ln|N|}, \quad N \equiv \frac{n}{n_*}. \tag{2}$$

The derivation was done within the commonly used approximation that neglects certain small electronic structure parameters γ_3, γ_4, and Δ' (for their physical meaning and a discussion of their numerical values, see Ref. 23). The parameters that are retained include the interlayer hopping energy $\gamma_1=0.41$ eV and the nearest-neighbor in-plane hopping $\gamma_0=3.0$ eV, which define the characteristic density scale in the problem

$$n_* = \frac{4}{3\pi a^2}\frac{\gamma_1^2}{\gamma_0^2} = 1.2 \times 10^{13} \text{ cm}^{-2}, \tag{3}$$

where $a=2.44$ Å is the lattice constant. The remaining notations in Eq. (2) are

$$N_{dt} = n_{dt}/n_*, \tag{4}$$

which is the scaled background density of positive charge n_{dt} on the top layer (or above it, see below) and

$$\Lambda_{\mathrm{Mc}} = \frac{2\pi e^2 d_m n_*}{\kappa_m \gamma_1} \sim 1, \tag{5}$$

which is the dimensionless strength of the interlayer screening, with $\kappa_m \approx 1$ being the effective dielectric constant of the medium between the layers. The screening is particularly significant in the narrow gap regime. If $N_{dt}=0$, it is realized in the limit $n \to 0$. According to Eq. (2), the screened field E_m is suppressed compared to the external field E_b by a divergent logarithmic factor [see Fig. 1 and also Eq. (30) below]. As a result, $2U$ has a superlinear dependence on n.

Formula (2) was recently challenged by Falkovsky.[24] Under the same conditions and assumptions, he obtained the simple linear law for $N_{dt}=0$,

$$2U = c\gamma_1 N, \quad c = \frac{2}{\sqrt{6.2784^2 - 1}} = 0.3227. \tag{6}$$

The goal of this Comment is to show that Ref. 24 contains egregious mistakes. Once they are corrected, Eq. (2) is recovered.

These mistakes are apparent on physical grounds. One key formula, Eq. (8) of Ref. 24, states (in our notations) that the BLG polarization

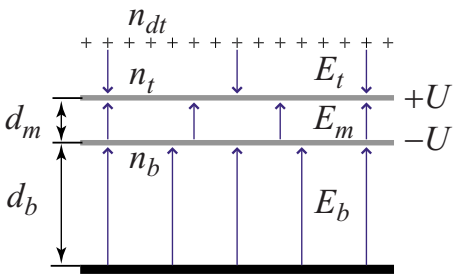

FIG. 1. (Color online) Device schematic. The BLG is shown as two horizontal lines in the middle. The thicker line at the bottom represents the gate. The arrows show the electric field in the system and the crosses depict positive charge density n_{dt} on (or on and above) the top layer of the BLG.

$$\Delta n \equiv n_t - n_b \tag{7}$$

is positive for $U, \mu > 0$, i.e., the BLG is polarized against, not along the electric field. In principle, such a phenomenon can arise if a system has a negative compressibility[25–27] due to exchange and correlations (see the discussion at the end). However, neither exchange nor correlation effects are included in the theory of Ref. 24. To expose the mistakes in Ref. 24, we carry out below our own derivation using the method proposed by Falkovsky in Ref. 24. Its idea is to treat U and μ as variational parameters and get their actual values by minimizing the total energy density ϵ of the system at a fixed n.

Let us proceed. In the mean-field approximation ϵ is the sum of the kinetic and the Hartree interaction terms, $\epsilon = \epsilon_{\rm kin} + \epsilon_{\rm H}$. The Hartree term is straightforward,

$$\epsilon_{\rm H} = \frac{e^2}{2c_b}(n - n_{dt} - n_{db}^0)^2 + \frac{2\pi e^2 d_m}{\kappa_m}(n_t - n_{dt})^2, \tag{8}$$

which can be understood as the energy of two parallel-plate capacitors in series. Here $c_b = \kappa_b/(4\pi d_b)$ is the capacitance per unit area between the BLG and the bottom gate, κ_b is the dielectric constant below the BLG, and n_{db}^0 is the density of additional positive background charge on the bottom layer (not shown in Fig. 1). For $n_{db}^0 = n_{dt}$, Eq. (8) agrees with Eq. (10) in Ref. 24.

The kinetic term $\epsilon_{\rm kin}$ requires a little care. The author of Ref. 24 seems to assume that it coincides with the sum of the occupied single-particle energies

$$\epsilon_{\rm sum} = \frac{1}{A}\sum_\alpha \Theta(\mu - E_\alpha)E_\alpha, \tag{9}$$

where α is a short-hand notation for all quantum numbers, $\Theta(x)$ is the unit step function, and A is the area of the system. In fact, each energy E_α, which is an eigenvalue of the BLG Hamiltonian $H = H_{\rm kin} + \Phi$ also contains a potential term: $E_\alpha = \langle\alpha|H_{\rm kin}|\alpha\rangle + \langle\alpha|\Phi|\alpha\rangle$. Here Φ stands for the electrostatic energy that is equal to $\pm U$ in the top (bottom) layer. Lumping together $\epsilon_{\rm sum}$ and $\epsilon_{\rm H}$ is incorrect as it leads to double counting of the interaction energy. Instead, the proper formula is

$$\epsilon_{\rm kin} = \frac{1}{A}\sum_\alpha \Theta(\mu - E_\alpha)\langle\alpha|H_{\rm kin}|\alpha\rangle = \epsilon_{\rm sum} - U\Delta n. \tag{10}$$

Next, using the Hellmann-Feynman theorem, we get

$$\Delta n = \sum_\alpha \frac{\Theta(\mu - E_\alpha)}{A}\langle\alpha|\frac{\partial H}{\partial U}|\alpha\rangle = \frac{\partial(\epsilon_{\rm sum} - \mu n)}{\partial U},$$

which entails

$$n_{t,b} = \frac{n \pm \Delta n}{2} = \frac{1}{2}\left[n \pm \frac{\partial}{\partial U}(\epsilon_{\rm sum} - \mu n)\right]. \tag{11}$$

The correct variational functional to minimize is

$$\Omega = \epsilon_{\rm H} + \epsilon_{\rm sum} - U\frac{\partial}{\partial U}(\epsilon_{\rm sum} - \mu n) - \xi n, \tag{12}$$

where ξ is a Lagrange multiplier. The sought minimum is determined by the equations $\partial\Omega/\partial\mu = \partial\Omega/\partial U = 0$, which leads to

$$2U = 4\pi e^2(d_m/\kappa_m)(n_t - n_{dt}). \tag{13}$$

This equation is obviously correct because it follows from Gauss's law. Indeed, it is the starting equation [Eq. (2)] of Ref. 4. The closest to Gauss's law in Ref. 24 is Eq. (11) of that paper but the electric field is defined in the wrong direction in that equation, as can be seen by comparison with Fig. 3 in Ref. 24.

Let us now compute n_t by the present method to demonstrate where Ref. 24 contains another mistake. The total polarization $\Delta n = \Sigma_{s_1,s_2}\Delta n_{s_1 s_2}$ is the sum over all four bands of BLG, where, in keeping with notations of Ref. 28, we label the bands as follows: $s_1 = \pm$ distinguishes the conduction and valence bands while $s_2 = \pm$ denotes the outer (inner) bands. The band dispersions are given by

$$E_{s_1 s_2} = s_1\sqrt{\frac{\gamma_1^2}{2} + \epsilon^2(\mathbf{q}) + U^2 + s_2\Lambda^2[\epsilon(\mathbf{q})]}, \tag{14}$$

$$\Lambda^2(\epsilon) \equiv \sqrt{\frac{\gamma_1^4}{4} + \epsilon^2(\gamma_1^2 + 4U^2)}, \tag{15}$$

where $\mathbf{q}$ is the quasimomentum and $\epsilon(\mathbf{q})$ is the energy dispersion in monolayer graphene. Near a BZ corner $\mathbf{K}$, the Dirac approximation is valid $\epsilon(\mathbf{q}) = (\sqrt{3}/2)\gamma_0 a|\mathbf{k}|$, where $\mathbf{k} = \mathbf{q} - \mathbf{K}$.

For simplicity, we consider the case where the Fermi surface is singly connected and includes only the $\{+-\}$ states so that $n = k_F^2/\pi$, where k_F is the Fermi momentum measured from the nearest BZ corner and

$$\mu = E_{+-}(k_F) \simeq \sqrt{U^2 + \frac{\epsilon^4(k_F)}{\gamma_1^2}} \simeq \sqrt{U^2 + \gamma_1^2 N^2}. \tag{16}$$

As explained above, if U is positive, then Δn should be negative. In Ref. 24 however only the conduction band term,[4,24]

$$\Delta n_{+-} = \frac{n_* U}{\gamma_1}\ln\left(\frac{\mu}{U} + \sqrt{\frac{\mu^2}{U^2} - 1}\right), \tag{17}$$

is included while the much larger negative contribution $\Delta n_- \equiv \Delta n_{-+} + \Delta n_{--}$ of the completely filled valence bands is neglected. This important term is given by

$$\Delta n_- = \frac{\partial}{\partial U}\int_{\rm BZ}\frac{2d^2q}{(2\pi)^2}[E_{-+}(\mathbf{q}) + E_{--}(\mathbf{q})]\Big|_{U=0}^{U>0}. \tag{18}$$

Within the Dirac approximation, the above integral can be done exactly by extending the integration domain to infinity, with the result

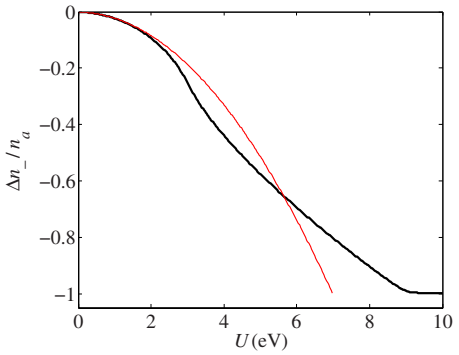

FIG. 2. (Color online) BLG polarization Δn_- due to the completely filled valence bands in the Dirac approximation [Eq. (19)] (thin red line) and the tight-binding model (thick black line). Parameter $n_a=4\sqrt{3}/3a^2$ is the atomic density.

$$\Delta n_- = -\frac{n_* u}{1+4u^2}\left[2u(u+\sqrt{1+u^2})^2 + \frac{1+2u^2}{\sqrt{1+4u^2}}\left(\sinh^{-1}\frac{1+2u^2}{u} + \sinh^{-1} 2u\right)\right], \quad (19)$$

where $u \equiv U/\gamma_1$. The leading term for $U \ll \gamma_1$ is

$$\Delta n_- \simeq -\frac{n_* U}{\gamma_1}\ln\frac{2\gamma_1}{U}, \quad (20)$$

in agreement with Ref. 4. The nonvanishing and, in fact, dominant contribution of the filled valence bands to the total polarization is not an artifact of the Dirac approximation. To prove that we repeated the calculation of Δn_- within the simplified tight-binding model, using

$$\epsilon(\mathbf{q}) = \gamma_0(e^{i\mathbf{q}\cdot\delta_1} + e^{i\mathbf{q}\cdot\delta_2} + e^{i\mathbf{q}\cdot\delta_3}) \quad (21)$$

in Eq. (14), with δ_j being the vectors connecting a lattice site to its three nearest neighbors. The integration in Eq. (18) was done numerically and the results are shown in Fig. 2. As expected, the Dirac approximation remains accurate up to $U \sim \gamma_0 = 3.0$ eV. At $\gamma_1 \ll U \ll \gamma_0$, the polarization varies quadratically with U because it is dominated by the properties of a single layer, where the density of states is linear in energy. Finally, when $2U$ exceeds the bandwidth $6\gamma_0 = \max \epsilon(\mathbf{q}) - \min \epsilon(\mathbf{q})$ of the monolayer dispersion, Δn_- quickly approaches its minimum possible value, which is equal to the total atomic density of BLG $n_a=4\sqrt{3}/3a^2$, with the minus sign.

Most relevant for current experiments is the case $U \ll \gamma_1$, where Eq. (20) applies. Combined with Eqs. (16) and (17), it yields the result of the correct sign for the total polarization,

$$\Delta n \simeq -\frac{n_* U}{\gamma_1}\ln\frac{2}{N+\sqrt{N^2+(U/\gamma_1)^2}}. \quad (22)$$

Substituting it into $n_t=(n+\Delta n)/2$ and using Eqs. (4), (5), and (13), we obtain the equation for the function $U(N)$,

$$2U(N) \simeq \gamma_1 \frac{N-2N_{dt}}{\Lambda_{\mathrm{Mc}}^{-1} + \frac{1}{2}\ln\frac{2}{N+\sqrt{N^2+(U/\gamma_1)^2}}}. \quad (23)$$

This formula is actually valid for arbitrary signs of U and N. In the region

$$2U(0)/\gamma_1 \ll N \ll 1 \quad (24)$$

it coincides with Eq. (2) within the accuracy of this calculation. Here

$$2U(0) \simeq -\frac{2\gamma_1 N_{dt}}{\Lambda_{\mathrm{Mc}}^{-1} + \frac{1}{2}\ln\frac{1}{|N_{dt}|}} \quad (25)$$

is the interlayer bias at $N=0$. Note that the gap $2|U|$ vanishes only at $N=2N_{dt}$, i.e., $n=2n_{dt}$, where

$$n_t = n_b = n_{dt}. \quad (26)$$

The gap remains nonzero at all other densities including $n=0$, in disagreement with the claim made in Ref. 24 but in agreement with the numerical results of Ref. 4. (A finite gap at zero density has indeed been observed in quantum Hall effect measurements[3] of doped BLG.)

As mentioned in the beginning of this Comment, the background positive charge density n_{dt} does not have to reside directly on the top layer. Equation (23) remains unchanged if some of this charge is located above the BLG, as sketched in Fig. 1. For example, for a dual-gated BLG

$$n_{dt} = n_{dt}^0 + \frac{c_t}{e^2}(eV_t - \mu + U), \quad (27)$$

where the first term is the fixed donor density on the top layer and the second term is the (tunable) charge density on the top gate. Parameter c_t is the capacitance between the top gate and the BLG per unit area. Similarly, the total density is given by

$$n = n_{dt}^0 + n_{db}^0 + \frac{c_t - c_b}{e^2}U + \sum_{a=t,b}\frac{c_a}{e^2}(eV_a - \mu), \quad (28)$$

where V_b is the voltage on the bottom gate. The chemical potential μ enters these equations because the measured "voltages" V_b and V_t of the two gates are not simply the electrostatic but instead the electrochemical potential differences between these gates and the BLG, cf. Refs. 17 and 27. However for distant gates,

$$d_a \gg d_m \frac{\kappa_a}{2\Lambda_{\mathrm{Mc}}\kappa_m}, \quad a=t,b, \quad (29)$$

one can in the first approximation neglect U and μ compared to eV_b and eV_t, leading to the simplified equations

$$2U = \frac{eE_0 d_m}{1 + \frac{\Lambda_{\mathrm{Mc}}}{2}\ln\frac{2}{N+\sqrt{N^2+(U/\gamma_1)^2}}}, \quad (30)$$

$$E_0 \simeq \frac{2\pi e}{\kappa_m}\left(n_{db}^0 - n_{dt}^0 + \frac{c_b V_b}{e} - \frac{c_t V_t}{e}\right), \tag{31}$$

$$N = \frac{n}{n_*} \simeq \frac{1}{n_*}\left(n_{db}^0 + n_{db}^0 + \frac{c_t V_t}{e} + \frac{c_b V_b}{e}\right), \tag{32}$$

which may be useful in experimental practice. Note however that unavoidable disorder creates additional corrections to these expressions,[23] which can be as large as 30%.

In closing, we comment on exchange and correlations effects. Our Eq. (2) is in qualitative agreement with density-functional theory (DFT) calculations,[20,29] which include some of these effects. On the other hand, spontaneous polarization $\Delta n \neq 0$ and the corresponding gap generation have been predicted to occur[19,30–34] even in the absence of the external electric field.[35] Such a symmetry breaking can be incorporated into either mean-field theory or the DFT by introducing a suitable self-energy difference between the layers, which is a function of N and U. In fact, we already have a similar parameter in our formalism—it is our N_{dt}. This interesting problem warrants further study.

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