# $\frac{d\mu}{dn}$ in suspended bilayer graphene: The interplay of disorder and band gap

D. S. L. Abergel, H. Min,* E. H. Hwang, and S. Das Sarma
*Condensed Matter Theory Center, Department of Physics, University of Maryland, College Park, Maryland 20742, USA*
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We present an interpretation of recent experimental measurements of $d\mu/dn$ in suspended bilayer graphene samples. We demonstrate that the data may be quantitatively described by assuming a spatially varying interlayer potential asymmetry (which generates a band gap) induced by local electric fields resulting from charged-impurity disorder in the graphene environment. We demonstrate that the fluctuations in the interlayer potential asymmetry and density vary between different samples, and that the potential asymmetry fluctuations increase in magnitude as the density fluctuations increase. This indicates that the mechanism causing this effect is likely to be extrinsic. We also provide predictions for the optical conductivity and mobility of suspended bilayer graphene samples with small band gaps in the presence of disorder.

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Recently, the fabrication of suspended bilayer graphene (BLG) devices[1,2] has allowed the direct local measurement of the thermodynamic quantity $K \equiv d\mu/dn$ using scanning single-electron transistor spectroscopy. This is an important quantity to study since it is linked directly to thermodynamic observables such as the thermodynamic density of states, electronic compressibility, and the quantum capacitance of the interacting electron liquid in the BLG. In samples mounted on a $SiO_2$ substrate, the level of disorder is generally too high for the intrinsic physics near the charge neutrality point to be seen because of the existence of strong inhomogeneity (electron-hole puddles) induced by the disorder.[3] However, suspended samples provide a setting where the effective strength of the impurities is reduced as reflected, for example, in the enhanced mobility[4,5] and it is likely that the low-density physics is more readily accessible. In the experiments,[1,2] a region of decreased compressiblilty (or equivalently a peak in $K$) is seen near the charge-neutrality point in zero magnetic field, indicating the possible presence of a small gap in the low-energy band structure. It is claimed[1] that the size of the gap that generates this peak is approximately 2 meV, and that the magnitude of charge inhomogeneity is of the order of $10^{10}$ cm$^{-2}$. The authors of Ref. 1 state that Castro *et al.*[6] show that this level of in-plane charge inhomogeneity by itself would create a gap which is a factor of ten smaller than that observed, ruling out disorder-induced fluctuations as a mechanism for the generation of this gap. Instead, they claim[1] that this peak is evidence for a correlated ground state of the interacting electron liquid, such as one of those proposed in the recent literature.[7–15] Some of these proposed states may induce a spontaneous many-body charge transfer instability in the bilayer system that could open a small band gap at zero density, and the region of decreased compressibility seen in the experiments may be evidence for the formation of one of these charge-transfer-instability states.[1] However, we believe that this interpretation of the experimental results is problematic because the out-of-plane charge imbalance, which would drive a disorder-induced gap is not the same quantity as the in-plane charge inhomogeneity, which Martin *et al.* use[1] to estimate the gap size. Therefore it is important to look more closely at the role of disorder in the suspended bilayer graphene system to investigate in depth the issue of the possible existence (or absence) of a many-body quantum phase transition creating a spontaneous band gap. The goal of the current work is to carry out such a phenomenological investigation to see whether random charge impurities in the bilayer environment could produce a signature in the compressibility consistent with the observations of Refs. 1 and 2.

Transport measurements suggest that there are some residual impurities in the environment of the suspended graphene, each of which will have an electric field associated with it, and it is well established that the electrons in the BLG order themselves into puddles of electrons and holes in order to screen these fields.[16] Additionally, it was shown recently[17] that these electric fields also cause a spatial variation in the interlayer potential asymmetry that is highly correlated with the disorder profile. This is a different effect from the in-plane reorganization of charge, and the two effects may in principle be of quite different magnitudes, and both are likely to be present in disordered bilayer graphene. Although this interlayer effect was reported for bilayer graphene supported on a substrate,[17] the general principle applies to suspended samples as well.

In this article, we revisit the issue of density fluctuations induced by charged impurity disorder, and show that our calculation of $K$ by averaging over the density of the electrons in the puddles[18] can be applied in two different phenomenological ways to this situation and gives results for the gap fluctuations, density fluctuations, and required charge imbalance that are self-consistent. By fitting this theory to measurements of $K$ from two different devices,[1,2] we show that the gap and charge inhomogeneity are device dependent, which is, of course, not unexpected, since disorder in different devices is likely to be different leading to different interlayer potential asymmetry in each sample. We also demonstrate that the transport characteristics associated with our phenomenological disorder fits are reasonable, and predict that in an optical spectroscopy experiment, the gap will still be obscured by disorder in these samples.[19] Since our results are internally consistent, we claim that the mechanism of local density fluctuations induced by charged impurity disorder cannot be ruled out as a cause of the observed[1,2] peak in $K$ at low carrier density. Our work by no means eliminates the possible occurrence of a many-body BLG instability, it just establishes the qualitative and quantitative importance of disorder in thinking about the quantitative

aspects of the observed effects, and points out that there is a possible single-particle extrinsic cause for the observed effective gap, namely, the interlayer potential asymmetry induced by the random charged impurities in the BLG environment. In reality, both interaction and disorder may be present in a nonperturbative manner, making a microscopic theoretical analysis quite challenging, particularly since the graphene landscape becomes inhomogeneous due to the formation of electron-hole puddles in the presence of the charged impurity disorder.

In order to model the clean BLG system, we consider the continuum limit of the tight-binding Hamiltonian. In this approach, the wave functions are described by Bloch functions modulated by a spinor, the elements of which correspond to the four sites in the BLG unit cell.[20] We make this assumption despite the fact that the inhomogeneity in the disorder potential breaks translational symmetry because we assume that the changes in the potential landscape are spatially slowly varying enough that the physics associated with $p$-$n$ junctions or quantum dots do not manifest. We also point out that this theory has been very successful in describing capacitance-based compressibility experiments in samples which are much dirtier than those we currently discuss.[18] The band structure is determined by the relative strength of electron hopping between the four lattice sites. To simulate the experimental situation as accurately as possible, we go beyond the standard nearest-neighbor tight-binding formalism (where $v_F$ is the Fermi velocity of monolayer graphene which comes from the intralayer hopping between adjacent atoms, and $\gamma_1$ is the energy associated with the interlayer dimer bond, which generates the finite effective mass at low energy) by including the next-nearest-neighbor interlayer hops with energy $\gamma_4$, the onsite energies given by the lattice site asymmetry $\Delta$, and a potential asymmetry $u$ between the two layers. This potential asymmetry directly generates a band gap at the K point of the band structure, and we shall therefore use the terms "interlayer potential asymmetry" and "gap" synonymously in this paper. The Hamiltonian for one valley is then

$$H = \begin{pmatrix} \frac{u}{2} & 0 & -\hbar v_4 k e^{-i\phi} & \hbar v_F k e^{-i\phi} \\ 0 & -\frac{u}{2} & \hbar v_F k e^{i\phi} & -\hbar v_4 k e^{i\phi} \\ -\hbar v_4 k e^{i\phi} & \hbar v_F k e^{-i\phi} & -\frac{u}{2}+\Delta & \gamma_1 \\ \hbar v_F k e^{i\phi} & -\hbar v_4 k e^{-i\phi} & \gamma_1 & \frac{u}{2}+\Delta \end{pmatrix}, \tag{1}$$

where $v_4 = \sqrt{3}\gamma_4 a/2\hbar$ with the lattice constant $a$, and $k$ and $\phi$ are respectively the radial and angular part of the wave vector measured from the K point. The spectrum is valley symmetric and isotropic. The effect of the $\gamma_4$ and $\Delta$ terms is to introduce an asymmetry between the low-energy conduction and valence bands, and has previously been used to explain an observed asymmetry in capacitance measurements.[21] Using the results collated in Ref. 20, we choose representative parameters $\gamma_1 = 0.4$ eV, $\gamma_4 = 0.15$ eV, $\Delta = 0.018$ eV, $v_F = 10^6$ ms$^{-1}$, and $a = 0.246$ nm. We describe the band structure and $K$ in the absence of disorder in the Appendix.

The existence of puddles of carriers in graphene is a well established phenomenon both in experiment and in theory.[22] In a previous paper,[18] we introduced the macroscopic averaging over density fluctuations and applied this theory to measurements of the capacitance of BLG devices finding good agreement with the experimental data. The idea of the theory is that the potential fluctuations associated with charged impurity disorder generates a spatially varying onsite term in the tight-binding Hamiltonian. This in turn leads to a local fluctuation in the Fermi energy and hence in the carrier density. The statistics of this fluctuation can be encapsulated within a distribution function $P$. The coupling between the tip of the SET and the electron liquid in the bilayer graphene is capacitative, so we assume that each region with a given carrier density has a local capacitance associated with it. Then, the total capacitive coupling of the SET to the sample is the parallel coupling of all of the areas, which equates to the geometrical average over the region sampled by the SET. Therefore the areal average can be replaced by an average over the charge density distribution $P$. We note that the dimension of the region sampled by the SET in the experiments of Ref. 1 is $\sim$100 nm, whereas the puddle size seen in Ref. 17 is $\sim$10 nm so that the averaging should be reasonable in this case. We stress that this philosophy deliberately retains the inhomogeneity of the system and treats it as the primary manifestation of disorder. This is a fundamentally different approach from, for example, a diagrammatic expansion of the electron-impurity interaction, which contains within it an average over realizations of disorder, which explicitly restores the translational symmetry. We believe that the important disorder physics to be included in the low-density regime of graphene near the charge neutrality point is the formation of electron-hole puddles in the system around the discrete quenched charged impurity centers leading to strong spatial inhomogeneity modeled by the distribution function $P$ mentioned above, which has been calculated in the literature in a few instances[3] and measured in experiment for a bare $SiO_2$ substrate.[23]

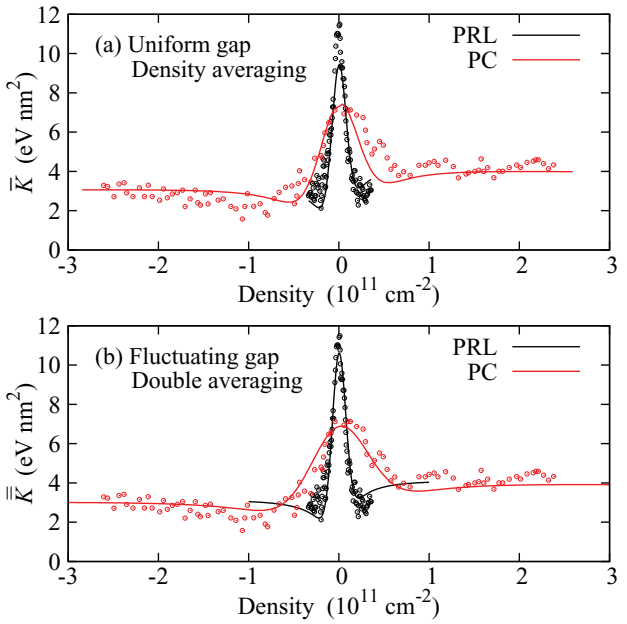
For density fluctuations associated with puddles of carriers in graphene systems, we write the local density as $n(r) = n_e + \tilde{n}(r)$, where $n_e$ is the average density induced by external gates and the fluctuations are encapsulated within $\tilde{n}$. We parametrize these fluctuations by their standard deviation $\delta n$, and computing the average of $K$ over the probability distribution for the density results in the following expression for the average inverse compressibility:

$$\bar{K}(n_e,\delta n,u) = \int K(n,u)P(n,n_e,\delta n)dn. \tag{2}$$

The distribution function $P$ is taken as the following Gaussian:

$$P(x,x_0,\delta x) = \frac{1}{\sqrt{2\pi}\delta x}\exp\left[-\frac{(x-x_0)^2}{2(\delta x)^2}\right], \tag{3}$$

since there is compelling theoretical[16] and experimental[23] evidence that this is approximately the correct form. In this theory, the layer asymmetry $u = u_c$ is constant in space (hence the subscript "c") and is generated by either external electric fields (such as those from gates) or by an asymmetry of charge between the two layers of the BLG generated by the intrinsic electron-electron interactions. Analytical evaluation of this procedure is not possible since, for finite $\gamma_4$ and $\Delta$, the eigenvalues of the Hamiltonian (1) are the roots of a quartic polynomial. Therefore the expression for $\mu$ as a function of $n$, its derivative, and the integration in Eq. (2) must be calculated numerically.




| | Density average | | Double average | |
|---|---|---|---|---|
| | $u_c$ (meV) | $\delta n$ ($10^{10}$cm$^{-2}$) | $\delta u$ (meV) | $\delta n$ ($10^{10}$cm$^{-2}$) |
| PRL | 1.7 | 0.8 | 2.2 | 0.75 |
| PC | 3.3 | 2.1 | 5.2 | 3.2 |

FIG. 1. (Color online) (a) Fits of the density averaged $\bar{K}$ from Eq. (2) to the experimental data. Note that this theory includes a spatially uniform band gap $u_c$. The black lines are for published data[1] and the red lines for the unpublished data.[2] (b) Fits of the double averaged $\bar{\bar{K}}$ from Eq. (4) with $u_c = 0$ to the experimental data. Note that in this theory, the band gap $u = \tilde{u}(r)$ fluctuates in space, and the distribution of these fluctuations is parameterized by $\delta u$. The table shows a comparison of the fitting parameters for the two samples and two procedures.

Figure 1(a) shows the results of finding the best fit between Eq. (2) and the experimental data.[1,2] We find that the published data[1] (which we shall denote by the label PRL) are quantitatively described by a constant gap $u_c = 1.7$ meV with density fluctuations characterized by $\delta n = 0.8 \times 10^{10}$ cm$^{-2}$, which is very close to the values claimed by Martin *et al.* in their paper.[1] The unpublished data[2] (denoted by PC, as in "private communication") is described by $u_c = 3.3$ meV and $\delta n = 2.1 \times 10^{10}$ cm$^{-2}$. Therefore a different degree of in-plane charge homogeneity and different size of gap is required to describe the experimental data in each sample, which is perfectly reasonable, since we expect the disorder to have sample-to-sample variations. Also, the band gap required to fit the data is larger for the more strongly disordered sample. The opposite would be true if the gap was the result of an intrinsic interaction-driven effect, since in this case, the gap due to interactions would be universal, but disorder would work to reduce its size.[19] Therefore these results, particularly the fact that the more strongly disordered sample exhibits the larger effective experimental band gap, indicate that there is some role being played by extrinsic effects in these experiments.

We now introduce another step in this theory, motivated by recent observations of spatial fluctuations in the interlayer potential asymmetry in bilayer graphene.[17] The interlayer asymmetry was measured in electron and in hole puddles of a bilayer graphene flake, which was mounted on an $SiO_2$ substrate. The difference in the asymmetry of approximately 70 meV was seen between the two puddles at zero back gate voltage, and an associated band gap was seen in the spectrum of Landau levels measured via STM. Although we expect that the magnitude of this effect will be smaller in suspended samples, the presence of disorder-induced puddles (as demonstrated above by our fitting to the appropriate theory) means that it is sensible to assume that the same disorder may also induce (albeit smaller in magnitude) local fluctuations in the potential asymmetry just as it does in graphene on substrates in Ref. 17. In this case, the potential asymmetry becomes a function of position such that $u \equiv u(r) = u_c + \tilde{u}(r)$. The fluctuations are contained within $\tilde{u}$, and we assume that they can be described by the distribution $P$ with standard deviation $\delta u$ (just as the density fluctuations can) because the same disorder is generating both types of fluctuation. Note that it is the in-plane component of the electric field associated with disorder that causes the formation of puddles, while the spatially varying interlayer potential asymmetry is driven by the out-of-plane component of the electric field. Therefore, while we expect the two types of fluctuation to be correlated, it is not obvious exactly what relationship should exist between them. However, on a phenomenological level, we can treat $\delta n$ and the potential asymmetry fluctuations parameterized by $\delta u$ as independent parameters. We can therefore perform an average over the gap fluctuations in the same way we previously did for the density. The spatially varying part of the band gap $\tilde{u}$ is described by $\delta u$, and the Gaussian is centered around $u_c$. The spatial average then corresponds to the following average over the distribution function:

$$\bar{\bar{K}}(n_e,\delta n,u_c,\delta u) = \int \bar{K}(n_e,\delta n,u) P(u,u_c,\delta u) du. \qquad (4)$$

We call this procedure "double averaging." In order to assess if the observed gap might be entirely due to disorder-induced fluctuations, we fit this expression with $u_c = 0$ to the experimental data. Therefore the fitting parameters are now $\delta u$ and $\delta n$, so we have two parameters as was the case for the density averaging procedure. The results are displayed in Fig. 1(b). We find that the PRL data are fitted by $\delta u = 2.2$ meV and $\delta n = 0.75 \times 10^{10}$ cm$^{-2}$ and the PC data by $\delta u = 5.2$ meV and $\delta n = 3.2 \times 10^{10}$ cm$^{-2}$. The fit of the double-averaged theory is actually slightly better than that which is averaged only over density (especially in the region of the peak), but both averaging procedures give results which are in reasonable agreement with the experiment, and therefore it is not possible to distinguish between a spatially fluctuating disorder-induced gap and a uniform intrinsic gap in the current data.[24] Therefore our work convincingly demonstrates the key importance of disorder, particularly the density and potential fluctuations associated with the inhomogeneous puddles, in determining the experimental compressibility measurements of Refs. 1 and 2.

Using the parameters from these fits, we can make the following observation. In principle, if there was truly no disorder then both $\delta n$ and $\delta u$ should be zero. However, if we take the two pairs extracted from the double-averaging fitting procedure and assume a linear extrapolation to $\delta n = 0$, we find a residual $\delta u \approx 1.3$ meV. This may indicate the presence of

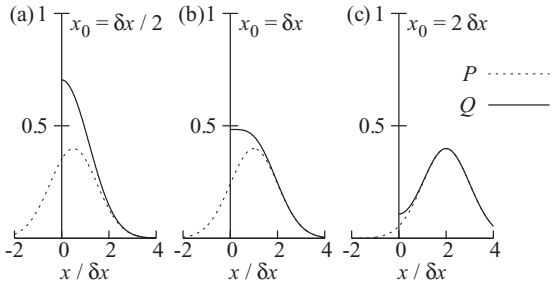



FIG. 2. (Color online) Plots of distributions $P$ and $Q$ for (a) $x_0 < \delta x$, (b) $x_0 = \delta x$, and (c) $x_0 > \delta x$. In all cases, we have $\delta x = 1$.

an intrinsic (possibly, many-body) gap not generated by the fluctuations, so we examine that issue now. The experimental measurements of $K$ in the PRL data suggest that the size of the uniform gap, if it exists, is approximately 2 meV. Since this is of the same order as the gap predicted by extrapolating to the clean limit, we have run fits of our double-averaged theory with a constant gap of this size to the experimental data. We find that there is no improvement in the fitting for any value of $\delta n$ or $\delta u$. To explain this, we need to analyze the distribution function for the gap fluctuations. We know that when $\gamma_4 = \Delta = 0$ that $K$ is an even function of the gap size $u$, and therefore the absolute value $|u|$ is the key for determining the distribution of $K$. Therefore, although we average over the Gaussian $P$ in Eq. (4), the effective distribution of $K$ is non-Gaussian, and given by

$$Q(x,x_0,\delta x) = P(x,x_0,\delta x) + P(-x,x_0,\delta x), \tag{5}$$

where $x > 0$ and $P(x,x_0,\delta_x)$ is defined in Eq. (3). Figure 2 shows the distribution $Q$ for the cases $x_0 < \delta x$, $x_0 = \delta x$, and $x_0 > \delta x$. We see that in all but the last case, the distribution is dominated by small values of $x$ because of the significant contribution from the $P(-x)$ term, and it is only when $\delta x > x_0$ that the Gaussian shape is revealed in $Q$. In the two sets of data that we have studied, we see that $u_c$ found from the single averaging procedure and the $\delta u$ from the double averaging obey the relation $u_c < \delta u$. This suggests that the disorder is too strong to determine if a spontaneous gap exists in the absence of disorder. In order to measure the spontaneous gap, samples with levels of disorder such that $\delta u < u_c$ are needed. This necessitates the development of samples with much higher quality (i.e., less disorder), perhaps mobilities that are a factor of five or so larger, to decisively establish the existence of a spontaneous many-body BLG gap. In short, the disorder-induced gap needs to be a factor of two or more smaller than the many-body gap for an unambiguous conclusion on this issue.

The gap size may also be extracted from measurements of the optical conductivity via absorption or reflection experiments.[25–28] To demonstrate that this may be an unreliable process in the case of the small band gaps (of the order of a few meVs) we are discussing in this context, we plot in Fig. 3 the optical conductivity derived from the Kubo formula with $u = 5$ meV in panel (a) and with $u = 2$ meV in panel (b). By way of comparison, in the linear (high density) part of the hyperbolic spectrum of bilayer graphene, the density fluctuations $\delta n$ correspond to fluctuations in the Fermi level of approximately $\hbar v_F \sqrt{\pi \delta n}$, and with $\delta n = 3 \times 10^{10}$ cm$^{-2}$, this gives an energy scale of approximately 20 meV. When

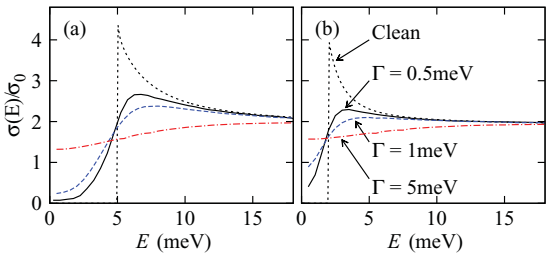



FIG. 3. (Color online) Optical conductivity for (a) $u_c = 5$ meV and (b) $u_c = 2$ meV for various degrees of disorder broadening. The color of the lines is the same in both panels. Dashed lines indicate the clean case.

the Fermi energy is in the sombrero (low-density) region with $u = 5$ meV, the flatness of the bands means that this density fluctuation corresponds to a fluctuation of less than 1 meV. We include the disorder through a constant broadening of the bands parameterized by the energy $\Gamma$, which appears in the Green's function as

$$G_{k\lambda}(E) = \frac{1}{E - E_{k\lambda} + i\Gamma}. \tag{6}$$

The optical conductivity is calculated from this Green's function from the following expression:[30]

$$\sigma(E) = 2g_s g_v \frac{e^2}{h} \sum_{\lambda,\lambda'} \int \frac{k'\,dk'}{2\pi} \int_{E_F - E}^{E_F} \frac{dE'}{E} \times M^2_{\lambda\lambda'}(k') \mathrm{Im}\, G_{k'\lambda}(E') \mathrm{Im}\, G_{k'\lambda'}(E' + E), \tag{7}$$

where $g_s$ and $g_v$ are the spin and valley degeneracies, and

$$M^2_{\lambda\lambda'}(k) = \int_0^{2\pi} \frac{d\phi}{2\pi} |\langle \lambda,k,0|\hbar \hat{v}_x|\lambda',k,\phi\rangle|^2. \tag{8}$$

Figure 3 shows that for $\Gamma = 1$ meV, the peak due to the onset of the intraband conductivity is already significantly blurred. By the time $\Gamma > u_c$, the peak is completely smeared, as shown by the $\Gamma = 5$ meV line. We mention in this context that the same conclusion can also be reached by considering an inhomogeneous broadening effect rather than a homogeneous broadening as used in Eq. (6), for example, we can introduce a density fluctuation leading to an uncertainty in the Fermi energy by 1 meV, leading to exactly the same conclusion that the optical gap measurement would be unable to discern the small band gap in the presence of this inhomogeneous broadening effect.

We also predict the transport properties of suspended bilayer graphene with disorder so that independent characterization of the samples in which $K$ was measured can be done and compared with our theory incorporating inhomogeneous puddle effects of density fluctuations. We use the highly successful Boltzmann-RPA formalism, which has been used to study the transport in monolayer graphene and BLG systems.[22] This theory incorporates both long-range Coulomb impurities and short-range scatterers. In Fig. 4(a), we show the mobility of suspended BLG as a function of charged impurity density $n_i$, calculated at a carrier density $n = 10^{11}$ cm$^{-2}$ for different short-ranged disorder potentials, $n_d V_0^2$, where $n_d$ is the density of short-ranged impurities and $V_0$ is the strength of the potential. If we assume a charged impurity density $n_i \approx \delta n$,

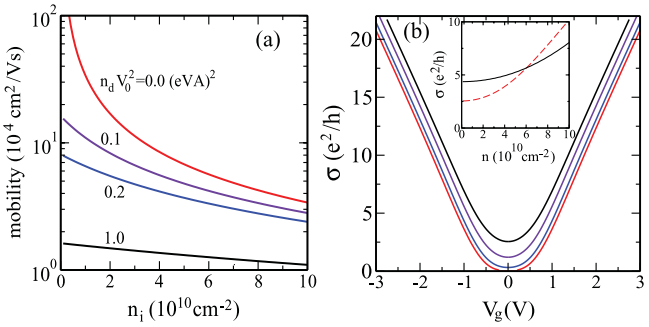



FIG. 4. (Color online) (a) Calculated mobility of suspended bilayer graphene as a function of impurity density for different strengths of the short-ranged impurities. (b) Conductivity calculated within effective medium theory as a function of gate voltage $V_g$ for a charged impurity density, $n_i = 2 \times 10^{10}$ cm$^{-2}$, and a fixed short-range potential $n_d V_0^2 = 0.3$ (eVÅ)$^2$. Different curves correspond to the band gap $E_g = 0$, 0.15, 0.3, 0.33 eV (from top to bottom). Inset shows the calculated conductivities as a function of carrier density for two different charged impurity densities $n_i = 2 \times 10^{10}$ cm$^{-2}$ (dashed line) and $n_i = 4 \times 10^{10}$ cm$^{-2}$ (solid line) with zero band gap.

then we have $\mu \sim 4$–$5 \times 10^4$ cm$^2$/Vs with a fixed short-range potential $n_d V_0^2 = 0.3$ (eVÅ)$^2$. However, we note that even though the carrier density fluctuation is related to the impurity density, the theoretical relation between two densities is not yet known. In Fig. 4(b), we show the conductivity calculated within effective medium theory[16] as a function of gate voltage $V_g$ in the presence of an energy band gap. We assume that the energy dispersion of carriers is given by $E_{k\nu} = \nu\hbar^2 k^2/(2m) + \nu E_g/2$, where $\nu = \pm 1$ indicates the electron ($\nu = 1$) or hole ($\nu = -1$) band, $m$ is the effective mass, and $E_g$ is the band gap. In this figure, a charged impurity density $n_i = 2 \times 10^{10}$ cm$^{-2}$ and a fixed short-range potential $n_d V_0^2 = 0.3$ (eVÅ)$^2$ are used. Our calculation of Fig. 4(b) shows that due to the electron-hole puddles, the transport gap (i.e., the region of zero conductivity) does not occur even though there is a band gap (top three curves in the figure). Only large enough energy gaps (the lowest curve) reflect the transport gap. However, our calculation also shows that for small density fluctuations (i.e., shallow electron-hole puddles), the transport gap can be induced by the smaller energy gap. In the inset to Fig. 4(b), the conductivity of suspended BLG calculated within the effective medium theory[22] is shown for two different charged impurity densities, $n_i = 2$ and $4 \times 10^{10}$ cm$^{-2}$ for a fixed short-range potential $n_d V_0^2 = 0.3$ (eVÅ)$^2$. Our calculation shows that the minimum conductivity at the charge neutral point is higher for a low-mobility (more strongly disordered) sample.

In conclusion, we have demonstrated that even though suspended graphene samples are known to be significantly less disordered than those mounted on substrates, the effect of disorder in recent measurements of $d\mu/dn$ cannot be ruled out. Using phenomenological averaging procedures, which specifically incorporate the effects of strong inhomogeneous broadening arising from electron-hole puddle induced spatial density and potential fluctuations, we have demonstrated that current experimental data cannot distinguish between an intrinsic, spatially uniform band gap generated by electron-electron interactions and a spatially fluctuating band gap induced by disorder. Specifically, these disorder-induced fluctuations can account for the observed data without invoking the presence of an intrinsic gap.[24] In order to be confident about the origin of the band gap, higher quality samples where $u_c > \delta u$ must be fabricated. One possibility is to carry out measurements on BLG samples fabricated on BN substrates, which typically have much lower charged impurity disorder.[29,31]

The theory we develop involves exactly one uncontrolled approximation, assuming the density fluctuations and the fluctuations in $u$ to be uncorrelated. Going beyond this approximation would require knowledge about the details of the local electric fields producing the fluctuations in $n$ and $u$, which is currently unavailable experimentally. It would be straightforward to include the correlation in our theory if microscopic information about the correlator between fluctuations in $n$ and $u$ is available from experiments or some other microscopic calculations. In the absence of such information, we assumed them to be independent.

We also emphasize that the theory presented in this article is phenomenological and is a first step toward the understanding of the role of disorder in the experiments of Refs. 1 and 2. What we have achieved in this paper is a demonstration that spatial density and potential fluctuations are capable of producing a spatially fluctuating gap consistent with the experimental data. Many questions of details could be raised with respect to our phenomenological procedure, for example, the averaging procedure we employ is certainly not unique, but in the presence of real spatial fluctuations, which explicitly break the translational invariance in graphene near the Dirac point, our procedure is physically motivated as it averages precisely over the quantities that fluctuate according to some distribution function, which has been calculated in the literature.[3,16,22,32] A better (purely numerical) method to tackle the problem would be to use the techniques developed in Refs. 3, 16, and 32 to explicitly and self-consistently calculate the real-space electronic structure in the presence of the electron-hole puddles generated by the random charge impurities, and then to use

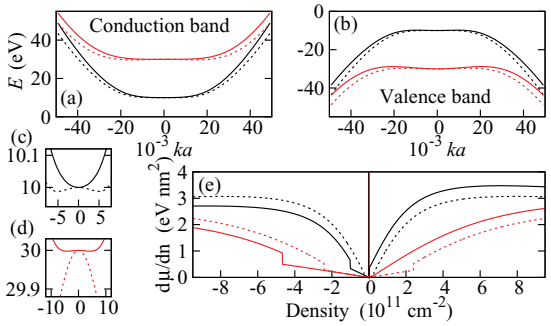



FIG. 5. (Color online) (a) and (b) The band structure of clean bilayer graphene with a band gap. The dashed lines correspond to the electron-hole symmetric band structure found with $\gamma_4 = \Delta = 0$ [see Eq. (A1)] and the solid line to the full band structure with $\gamma_4 = 150$ meV and $\Delta = 18$ meV. Black lines are for $u = 20$ meV and red lines for $u = 60$ meV. (c) The bottom of the conduction band for $u = 20$ meV showing the absence of sombrero structure for finite $\gamma_4, \Delta$. (d) The bottom of the conduction band for $u = 60$ meV showing the restoration of the sombrero structure for finite $\gamma_4, \Delta$. (e) $d\mu/dn$ for clean bilayer graphene. The lines correspond to the same as in (a).

this real-space electronic structure to directly numerically calculate the compressibility. Such a calculation is obviously numerically prohibitively difficult, and our work is a shortcut (albeit a rather simple one) in carrying out such a numerical procedure. Since the Gaussian probability distribution function we use is numerically well verified in Refs. 3, 16, 22 and 32 through self-consistent calculations, we believe that our results, although phenomenological, should be qualitatively and quantitatively valid. The eventual theory involving a microscopic calculation of the compressibility in the presence of long-range Coulomb disorder and electron-electron interaction including the realistic band structure of bilayer graphene is very far in the future, since no one knows how to approach such a nonperturbative problem even at a formal level.

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## APPENDIX: CLEAN BILAYER GRAPHENE

The band structure associated with the Hamiltonian presented in Eq. (1) is shown in Fig. 5. In the symmetric case with $\gamma_4 = \Delta = 0$, the low-energy bands have the dispersion

$$E_{\pm} = \pm\sqrt{\frac{\gamma_1^2}{2} + \frac{u^2}{4} + \kappa^2 - \sqrt{\frac{\gamma_1^4}{4} + \kappa^2\left(\gamma_1^2 + u^2\right)}}, \tag{A1}$$

where $\kappa = \hbar v_F k$. Writing this as a function of density and differentiating yields the following analytical expression for $d\mu/dn$ in this limit in the conduction band:

$$K = \begin{cases} \frac{\hbar^2 v_F^2 \pi}{2} \frac{\lambda}{\sqrt{u^2+\gamma_1^2}} \frac{1}{\sqrt{\lambda^2+\gamma_1^2 u^2}}, & \lambda < u^2, \\ \frac{\hbar^2 v_F^2 \pi}{2} \frac{1-(u^2+\gamma_1^2)/(2\alpha)}{\sqrt{\lambda+\frac{u^2}{4}+\frac{\gamma_1^2}{2}-\alpha}}, & \lambda \geqslant u^2, \end{cases} \tag{A2}$$

where $\lambda = \hbar^2 v_F^2 \pi n$ and $\alpha = \sqrt{\lambda(u^2+\gamma_1^2) + \frac{\gamma_1^4}{4}}$. In contrast, when $\gamma_4$ and $\Delta$ are included in the analysis, analytical solutions of the eigenvalue problem are not possible. The energy spectrum is found as the roots of the following quartic polynomial equation for $E$:

$$\begin{aligned} E^4 - 2\Delta E^3 &+ \left[\Delta^2 - \gamma_1^2 - \frac{u^2}{2} - 2\kappa^2(1+\chi^2)\right] E^2 \\ &+ \left[\frac{\Delta u^2}{2} + 2\kappa^2(\Delta + \chi^2\Delta + 2\chi\gamma_1)\right] E \\ &+ \frac{u^2}{4}\left(\gamma_1^2 - \Delta^2\right) + \left[\frac{u^2}{4} - \kappa^2(1-\chi^2)\right]^2 = 0, \end{aligned} \tag{A3}$$

where $\chi = v_4/v_F$. The Fermi surface may be ring shaped for finite values of $u$ so that the wave vectors corresponding to the inner and outer Fermi surfaces $k_{\pm}$ are given by the equation $\pi n = k_+^2 - k_-^2$ with the constraint that $k_+$ and $k_-$ must give the same energy for the appropriate band when substituted into Eq. (A3). When the Fermi energy is above the sombrero region, $k_- = 0$. The extracted value of $k_+$ can be substituted into Eq. (A3) to find $\mu$. The differentiation to obtain $K$ can then be carried out numerically. Evaluations of these equations are shown in Fig. 5 for the band structure and $K$ for two different values of the band gap and with and without the tight-binding parameters $\gamma_4$ and $\Delta$. The step feature in $K$ is a result of the change in topology of the Fermi surface from a ring to a disk as the Fermi energy leaves the sombrero region. For small values of $u$, the conduction band does not show the sombrero shape when $\gamma_4, \Delta$ are finite and this step is absent. The spike at the origin is a $\delta$ function resulting from the discontinuity of the Fermi energy at zero density as it jumps from the valence band to the conduction band.

*Current address: Department of Physics and Astronomy, Seoul National University, Seoul 151–747, Korea.

[1] J. Martin, B. E. Feldman, R. T. Weitz, M. T. Allen, and A. Yacoby, Phys. Rev. Lett. **105**, 256806 (2010).
[2] A. Yacoby (private communication).
[3] E. Rossi and S. Das Sarma, Phys. Rev. Lett. **101**, 166803 (2008).
[4] K. I. Bolotin, K. J. Sikes, Z. Jiang, M. Klima, G. Fudenberg, J. Hone, P. Kim, and H. L. Stormer, Solid State Commun. **146**, 351 (2008).
[5] S. Adam, S. Das Sarma, Solid State Commun. **146**, 356 (2008).
[6] E. V. Castro, K. S. Novoselov, S. V. Morozov, N. M. R. Peres, J. M. B. Lopes dos Santos, J. Nilsson, F. Guinea, A. K. Geim, and A. H. Castro Neto, Phys. Rev. Lett. **99**, 216802 (2007).
[7] J. Nilsson, A. H. Castro Neto, N. M. R. Peres, and F. Guinea, Phys. Rev. B **73**, 214418 (2006).
[8] H. Min, G. Borghi, M. Polini, and A. H. MacDonald, Phys. Rev. B **77**, 041407(R) (2008).
[9] F. Zhang, H. Min, M. Polini, and A. H. MacDonald, Phys. Rev. B **81**, 041402 (2010).
[10] R. Nandkishore and L. Levitov, Phys. Rev. B **82**, 115124 (2010).
[11] O. Vafek and K. Yang, Phys. Rev. B **81**, 041401 (2010).
[12] Y. Lemonik, I. L. Aleiner, C. Toke, and V. I. Falko, Phys. Rev. B **82**, 201408 (2010).
[13] R. Nandkishore and L. Levitov, Phys. Rev. Lett. **104**, 156803 (2010).
[14] J. Jung, F. Zhang, and A. H. MacDonald, Phys. Rev. B **83**, 115408 (2011).
[15] F. Zhang, J. Jung, G. A. Fiete, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. **106**, 156801 (2011).
[16] S. Das Sarma, E. H. Hwang and E. Rossi, Phys. Rev. B **81**, 161407 (2010).
[17] G. M. Rutter, S. Jung, N. N. Klimov, D. B. Newell, N. B. Zhitenev, and J. A. Stroscio, Nat. Phys. **7**, 649 (2011).
[18] D. S. L. Abergel, E. H. Hwang, and S. Das Sarma, Phys. Rev. B **83**, 085429 (2011).

[19]H. Min, D. S. L. Abergel, E. H. Hwang, and S. Das Sarma, Phys. Rev. B **84**, 041406 (2011).
[20]D. S. L. Abergel, V. A. Apalkov, J. Berashavich, K. Ziegler, and T. Chakraborty, Adv. Phys. **59**, 261 (2010).
[21]E. A. Henriksen and J. P. Eisenstein, Phys. Rev. B **82**, 041412(R) (2010).
[22]S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi, Rev. Mod. Phys **83**, 407 (2011).
[23]K. Burson *et al*. (unpublished).
[24]A typical disorder-induced charge imbalance between the two BLG layers as small as $1.6 \times 10^{10}$ cm$^{-2}$ (which is of the same order as the disorder-induced in-plane density fluctuations $\delta n$ in the system, although they are independent physical quantities) is sufficient to produce the band gap necessary for interpreting the observed peak in $K$ in the experiment.[1,2] Although this is suggestive that the experimental peak is arising entirely from extrinsic effects, we cannot, however, rule out a small many-body spontaneous gap underlying the disorder effects.
[25]Y. Zhang, T.-T. Tang, C. Girit, Z. Hao, M. C. Martin, A. Zettl, M. F. Crommie, Y. Ron Shen, F. Wang, Nature (London) **459**, 820 (2009).
[26]K. F. Mak, C. H. Lui, J. Shan, and T. F. Heinz, Phys. Rev. Lett. **102**, 256405 (2009).
[27]A. B. Kuzmenko, I. Crassee, D. van der Marel, P. Blake, and K. S. Novoselov, Phys. Rev. B **80**, 165406 (2009).
[28]Z. Q. Li, E. A. Henriksen, Z. Jiang, Z. Hao, M. C. Martin, P. Kim, H. L. Stormer, and D. N. Basov, Phys. Rev. Lett. **102**, 037403 (2009).
[29]S. Das Sarma and E. H. Hwang, Phys. Rev. B **83**, 121405 (2011).
[30]D. S. L. Abergel, H. Min, E. Hwang, and S. Das Sarma, e-print arXiv:1109.5702 (unpublished).
[31]C. R. Dean, A. F. Young, I. Meric, C. Lee, L. Wang, S. Sorgenfrei, K. Watanabe, T. Taniguchi, P. Kim, K. L. Shepard, and J. Hone, Nat. Nano. **5**, 722 (2010).
[32]E. Rossi and S. Das Sarma, Phys. Rev. Lett. **107**, 155502 (2011).