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Insulating behavior in metallic bilayer graphene: Interplay between density inhomogeneity and temperature

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We investigate bilayer graphene transport in the presence of electron-hole puddles induced by long-range charged impurities in the environment. We explain the insulating behavior observed in the temperature-dependent conductivity of low mobility bilayer graphene using an analytic statistical theory taking into account the non-mean-field nature of transport in the highly inhomogeneous density and potential landscape. We find that the puddles can induce, even far from the charge neutrality point, a coexisting metallic and insulating transport behavior due to the random local activation gap in the system.

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Recent experiments[1–4] have revealed an intriguingly strong (and anomalous) "insulating" temperature dependence in the measured electrical conductivity of bilayer graphene (BLG) samples, not only at the charge neutrality point (CNP) where the electron-hole bands touch each other (with vanishing average carrier density), but also at carrier densities as high as 10^{12} cm^{-2} or higher. [Insulating temperature dependence of conductivity $\sigma(T)$ simply means an increasing σ with increasing temperature at a fixed gate voltage, which is, in general, considered unusual in a nominally metallic system where the resistivity, not the conductivity, should increase with temperature.] Such an anomalous insulating temperature dependence of $\sigma(T)$ is typically not observed in monolayer graphene (MLG) away from the CNP although the gate voltage (or equivalently, the density) dependence of MLG and BLG conductivities are similar.[5,6] To be specific we mention the recent measurements by Zhu *et al.*,[1] but other groups[2–4] have made very similar observations. In Ref. 1, BLG $\sigma(T)$ increases by 20–40 % for carrier density in the $3.19\times10^{12}-7.16\times10^{12}$ cm^{-2} range as T increases from 4 to 300 K compared with the corresponding MLG case where $\sigma(T)$ decreases, as is customary for metallic transport, by 10–30 % in the same temperature and similar density range. The metallic nature of the BLG system is verified by the fact that $k_F l \gtrsim 5-10$ in Ref. 1, and is even higher in other BLG samples studied by other groups.[3,4] In addition, the temperature range (4–300 K) satisfies the low temperature $T \ll T_F$ criterion, making the insulating behavior very mysterious.

In this Rapid Communication we theoretically establish that this anomalous insulating BLG $\sigma(T)$ behavior is likely to be caused by the much stronger BLG density inhomogeneity[7] (compared with MLG) which gives rise to a qualitatively new type of temperature dependence in graphene transport, namely, the intriguing coexistence of both metallic and activated transport, hitherto not discussed in the literature. We therefore predict that the observed temperature dependence of BLG $\sigma(T)$ arises from the same charged impurity-induced puddles in the system which are responsible for the minimum conductivity plateau at the CNP.[8] We provide an analytic theory which appears to be in excellent qualitative agreement with the existing experimental results. One direct prediction of our theory, the suppression of the anomalous insulating temperature dependence in high mobility samples with lower disorder, seems to be consistent with experimental observations.

Our theory is based on a physically motivated idea: in the presence of large potential fluctuations $V(\mathbf{r})$, the local Fermi level, $\mu(\mathbf{r})=E_F-V(\mathbf{r})$, would necessarily have large spatial fluctuations [particularly when $E_F \lesssim s$, where $s=V_{\rm rms}$ is the root-mean-square fluctuations or the standard deviation in $V(\mathbf{r})$], leading to a complex temperature dependence of transport since both metallic and activated transport would be present due to random local gap. Below we carry out an analytical theory implementing this physical idea. We will see that this physical idea leads to the possible coexistence of metallic and activated transport, which explains the observed temperature dependence of BLG transport.

We start by assuming that the disorder-induced potential-energy fluctuations in the BLG are described by a distribution function $P(V)$ which $V=V(\mathbf{r})$ is the fluctuating potential energy at the point $\mathbf{r}\equiv(x,y)$ in the two-dimensional (2D) BLG plane. We approximate the probability $P(V)dV$ of finding the local electronic potential energy within a range dV about V to be a Gaussian form, i.e., $P(V)=\frac{1}{\sqrt{2\pi s^2}} \times\exp(-V^2/2s^2)$, where s is the standard deviation (or equivalently, the strength of the potential fluctuation). Then in the presence of electron-hole puddles the density of states (DOS) is reduced by the allowed electron region fraction and given by $D(E)=\int_{-\infty}^{E} D_0 P(V)dV = D_0\,\mathrm{erfc}(-E/\sqrt{2}s)/2$, where erfc$(x)$ is the complementary error function and $D_0 = g_s g_v m/(2\pi\hbar^2)$ is the DOS in a homogeneous system, where m is the band effective mass, $g_s=2$ and $g_v=2$ are the spin and valley degeneracies, respectively. We have $D_0=2.8 \times 10^{10}$ cm^{-2}/meV with the effective mass $m=0.033 m_e$ (where m_e is the bare electron mass). Note that the tail of the DOS is determined by the potential fluctuation strength s and the DOS in the presence of disorder is no longer a constant.

Since BLG is a gapless semiconductor the electron density at finite temperature increases due to the direct thermal excitation from valence band to conduction band, and this thermal excitation is an important source of temperature-dependent transport. Thus, we first consider the temperature dependence of thermally excited electron density. The total electron density is given by

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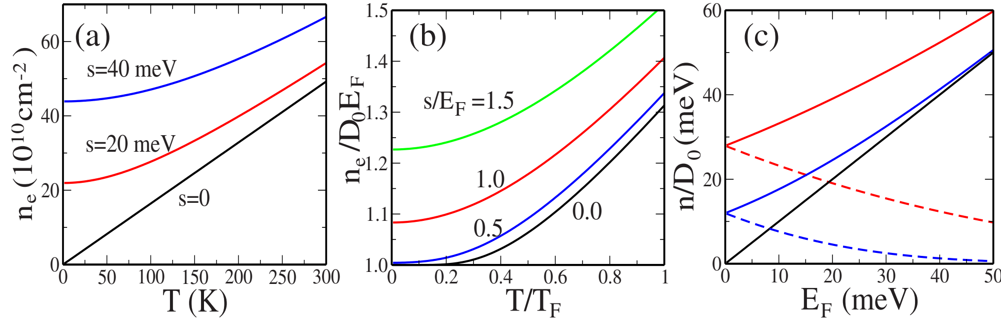

FIG. 1. (Color online) (a) The electron density at CNP as a function of temperature for different s. At $T=0$ the density is given by $n_0=D_0 s/\sqrt{2\pi}$. (b) The temperature-dependent electron density at finite E_F for different s. For $s/E_F\neq 0$ the leading-order behavior is quadratic while at $s=0$ the density is exponentially suppressed. (c) Total electron densities (solid lines) and hole densities (dashed lines) as a function of E_F for two different $s=30$ and 70 meV. The linear line represents the density difference $n=n_e-n_h=D_0E_F$, which linearly depends on the Fermi energy. The densities at the band tails are given by $n_e(E_F=0)=n_h(E_F=0)=D_0 s/\sqrt{2\pi}$.

$$n_e = \int_{-\infty}^{\infty} D(E)\frac{dE}{e^{\beta(E-E_F)}+1}, \tag{1}$$

where $\beta=1/k_BT$ and E_F is the Fermi energy. When the Fermi energy is zero (or at CNP) all electrons are located in the band tail at $T=0$ and the electron density in the band tail is given by $n_0=n_e(E_F=0)=D_0 s/\sqrt{2\pi}$. Note that the electron density in the band tail is linearly proportional to the standard deviation s. At finite temperatures we find the asymptotic behavior of $n_0(T)$. The low-temperature ($k_BT/s \ll 1$) behavior of electron density at CNP becomes

$$n_e(T) = n_0\left[1+\frac{\pi^2}{6}\left(\frac{k_BT}{s}\right)^2\right]. \tag{2}$$

Thus, the electron density increases quadratically in low-temperature limit. For homogeneous BLG with the constant DOS the electron density at finite temperatures is given by $n_e(T)=D_0 \ln(2)k_BT$. The presence of the band tail suppresses the thermal excitation of electrons and gives rise to the quadratic behavior. However, at high temperature the density increases linearly with the same slope as in the homogeneous system, i.e.,

$$n(T) \sim D_0\left[\ln(2)k_BT + \frac{1}{8}\frac{s^2}{(k_BT)^2}\right]. \tag{3}$$

In Fig. 1(a) we show the temperature-dependent electron density at CNP for different standard deviations.

In the case of finite doping (or gate voltage), i.e., the Fermi level away from CNP, $E_F\neq 0$, the electron density of the homogeneous BLG for $s=0$ is given by

$$n_{0e}(T) = D_0E_F[1+t\ln(1+e^{-1/t})], \tag{4}$$

where $t=T/T_F$ and $T_F=E_F/k_B$. At low temperatures ($T \ll T_F$) the thermal excitation is exponentially suppressed due to the Fermi function but at high temperatures ($T\gg T_F$) it increases linearly. In the presence of electron-hole puddles ($s\neq 0$) we have the electron density at zero temperature for the inhomogeneous system,

$$n_e(0) = D_0E_F\left[\frac{1}{2}\mathrm{erfc}\left(\frac{-1}{\sqrt{2}\tilde{s}}\right)+\frac{\tilde{s}}{\sqrt{2\pi}}e^{-1/2\tilde{s}^2}\right], \tag{5}$$

where $\tilde{s}=s/E_F$. At low temperatures ($T\ll T_F$) the asymptotic behavior of the electron density is given by

$$n_e(T) = n_e(0) + D_0E_F\frac{\pi^2}{12\sqrt{2}}\frac{e^{-1/2\tilde{s}^2}}{\tilde{s}}\left(\frac{T}{T_F}\right)^2. \tag{6}$$

The leading-order term is the same quadratic behavior as in undoped BLG ($E_F=0$) but the coefficient is strongly suppressed by fluctuation. In the case of $s>E_F$, the existence of electron-hole puddles gives rise to a notable quadratic behavior [see Fig. 1(b)]. At high temperatures ($T\gg T_F$) we find

$$n_e(T) = n_{0e}(T) + \frac{D_0E_F}{(1+e^{\beta E_F})^2}\frac{\tilde{s}^2}{2}\frac{T_F}{T}. \tag{7}$$

At CNP ($E_F=0$) electrons and holes are equally occupied. As the Fermi energy increases, more electrons occupy increasingly larger proportion of space [see Fig. 1(c)]. For $E_F\gg s$ nearly all space is allowed to the electrons and the conductivity of the system approaches the characteristic of the homogeneous materials. In the presence of electron-hole puddles, there is a possible coexistence of metallic and thermally activated transport. When electron puddles occupy more space than hole puddles, most electrons follow the continuous metallic paths extended throughout the system, but it is possible at finite temperature that the thermally activated transport of electrons persists above the hole puddles. On the other hand, holes in hole puddles propagate freely but when they meet electron puddles activated holes conduct over the electron puddles. Carrier transport in each puddle is characterized by propagation of weak scattering transport theory.[7] The activated carrier transport of prohibited regions, where the local potential energy is V less (greater) than Fermi energy for electrons (holes), is proportional to the Fermi factor. If σ_e and σ_h are the average conductivity of electron and hole puddles, respectively, then the activated conductivities are given by

$$\sigma_e^{(a)}(V) = \sigma_e \exp[\beta(E_F - V)], \tag{8a}$$

$$\sigma_h^{(a)}(V) = \sigma_h \exp[\beta(V - E_F)], \tag{8b}$$

where the density and temperature-dependent average conductivities (σ_e and σ_h) are given within the Boltzmann transport theory[7] by $\sigma_e = n_e e^2 \langle \tau \rangle / m$ and $\sigma_h = n_h e^2 \langle \tau \rangle / m$, where n_e and n_h are average electron and hole densities, respectively, and $\langle \tau \rangle$ is the transport relaxation time which depends explicitly on the scattering mechanism.[7]

Now we denote the electron (hole) puddle as region "1" (2). In region 1 electrons occupy more space than holes when $E_F > 0$. The fraction of the total area occupied by electrons with Fermi energy E_F is given by $p = \int_{-\infty}^{E_F} P(V) dV$. Then the total conductivity of region 1 can be calculated,

$$\sigma_1 = \frac{1}{p} \int_{-\infty}^{E_F} (\sigma_e + \sigma_h^{(a)}) P(V) dV$$

$$= \sigma_e + \frac{\sigma_h}{2p} e^{(\beta^2 s^2/2) - \beta E_F} \operatorname{erfc}\left(-\frac{E_F}{\sqrt{2}s} + \frac{\beta s}{\sqrt{2}} \right). \tag{9}$$

At the same time the holes occupy the area with a fraction $q = 1 - p$ and the total conductivity of region 2 becomes

$$\sigma_2 = \frac{1}{q} \int_{E_F}^{\infty} (\sigma_e^{(a)} + \sigma_h) P(V) dV$$

$$= \sigma_h + \frac{\sigma_e}{2q} e^{(\beta^2 s^2/2) + \beta E_F} \operatorname{erfc}\left(\frac{E_F}{\sqrt{2}s} + \frac{\beta s}{\sqrt{2}} \right). \tag{10}$$

The σ_1 and σ_2 are distributed according to the binary distribution. The conductivity of binary system can be calculated by using the effective-medium theory of conductance in mixtures.[9] The result for a 2D binary mixture of components with conductivity σ_1 and σ_2 is given by[9]

$$\sigma_t = \left(p - \frac{1}{2} \right) \left[(\sigma_1 - \sigma_2) + \sqrt{(\sigma_1 - \sigma_2)^2 + \frac{4\sigma_1 \sigma_2}{(2p-1)^2}} \right]. \tag{11}$$

This result can be applied for all Fermi energy. For a large doping case, in which the hole puddles disappear, we have $p = 1$ and $\sigma_2 = 0$, then Eq. (11) becomes $\sigma = \sigma_1$, i.e., the conductivity of electrons in the homogeneous system.

We first consider the conductivity at CNP ($E_F = 0$). The conductivities in each region are given by

$$\sigma_1 = \sigma_e \left[1 + \frac{\eta}{2p} e^{\beta^2 s^2/2} \operatorname{erfc}(\beta s/\sqrt{2}) \right], \tag{12a}$$

$$\sigma_2 = \sigma_h \left[1 + \frac{1}{2q\eta} e^{\beta^2 s^2/2} \operatorname{erfc}(\beta s/\sqrt{2}) \right], \tag{12b}$$

where $\eta = n_h / n_e$ is the ratio of the hole density to the electron density. Since the electrons and holes are equally populated we have $p = q = 1/2$ and $\sigma_e = \sigma_h$, then the total conductivity becomes $\sigma_t = \sqrt{\sigma_1 \sigma_2} = \sigma_1$. The asymptotic behavior of the conductivity at low temperatures ($k_B T \ll s$) becomes

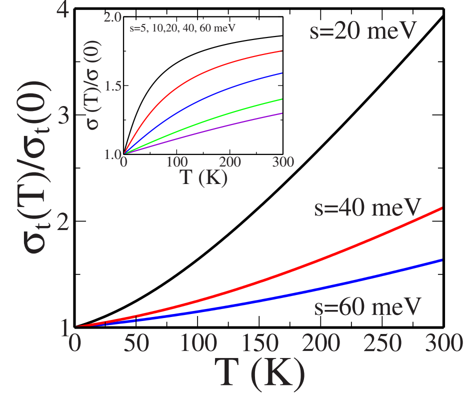

FIG. 2. (Color online) $\sigma_t(T)$ at charge neutral point for different s. Inset shows the thermally activated conductivity as a function temperature.

$$\sigma_t(T) = \sigma_e \left[1 + \sqrt{\frac{2}{\pi}} \frac{k_B T}{s} - \frac{2}{\sqrt{\pi}} \frac{(k_B T)^3}{s^3} \right]. \tag{13}$$

The activated conductivity increases linearly with a slope $\sqrt{2/\pi} k_B / s$ as temperature increases. Because s is typically smaller in higher mobility sample, the high mobility samples show stronger insulating behavior at low temperatures. The next order temperature correction to conductivity arises from the thermal excitation given in Eq. (2) which gives T^2 corrections. Thus in low-temperature limit the total conductivity at CNP is given by

$$\sigma_t(T) = \sigma(0) \left[1 + \sqrt{\frac{2}{\pi}} \frac{k_B T}{s} + \frac{\pi^2}{6} \left(\frac{k_B T}{s} \right)^2 \right]. \tag{14}$$

At high temperatures ($k_B T \gg s$) we have

$$\sigma_t = \sigma_e \left[2 - \sqrt{\frac{2}{\pi}} \frac{s}{k_B T} + \frac{s^2}{2(k_B T)^2} \right]. \tag{15}$$

The total conductivity due to the activation behavior approaches a limiting value and all temperature dependence comes from the thermal excitation through the change in carrier density given in Eq. (3). Thus at very high temperatures ($T \gg s/k_B$) the BLG conductivity at the charge neutral point increases linearly with a universal slope ln(2) regardless of the sample quality. In Fig. 2 we show the calculated temperature-dependent conductivity at charge neutral point.

At finite doping ($E_F > 0$) the temperature-dependent conductivities are very complex because three energies (E_F, s, and $k_B T$) are competing. Especially when $k_B T \ll s$, regardless of E_F, we have the asymptotic behavior of conductivities in region 1 and 2 from Eqs. (9) and (10), respectively,

$$\sigma_1 = \sigma_e \left[1 + \frac{\eta}{2p} e^{-1/2\tilde{s}^2} \sqrt{\frac{2}{\pi}} \frac{1}{\tilde{s}/t - 1/\tilde{s}} \right], \tag{16a}$$

$$\sigma_2 = \sigma_h \left[1 + \frac{1}{2q\eta} e^{-1/2\tilde{s}^2} \sqrt{\frac{2}{\pi}} \frac{1}{\tilde{s}/t + 1/\tilde{s}} \right], \tag{16b}$$

where $\tilde{s} = s/E_F$ and $t = T/T_F$. The leading-order correction is linear but the coefficient is exponentially suppressed by the

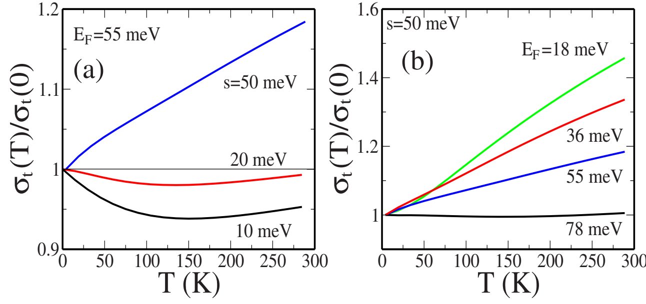

FIG. 3. (Color online) (a) $\sigma_t(T)$ for $E_F=55$ meV and for different s. (b) $\sigma_t(T)$ for $s=50$ meV and for several $E_F=18$, 36, 55, and 78 meV, which correspond to the densities $n=0.5\times10^{12}$, 1.0 $\times10^{12}$, 1.5×10^{12}, and 2.0×10^{12} cm^{-2}.

term $\exp(-E_F^2/2s^2)$. This fact indicates that in the high mobility sample with small s, the activated conductivity is weakly temperature dependent except around CNP, i.e., $E_F < s$. Since the density increase by thermal excitation is also suppressed exponentially by the same factor [see Eq. (6)] the dominant temperature-dependent conductivity arises from the scattering time.[7] On the other hand, for a low mobility sample with a large s, the linear temperature dependence due to thermal activation can be observed even at high densities $E_F \gtrsim s$.

In Fig. 3 we show the total conductivities (a) for a fixed E_F and several s and (b) for a fixed s and several E_F. In total conductivity the activated insulating behavior competes with the metallic behavior due to the temperature-dependent screening effect. When s is small the activated behavior is suppressed. As a result the total conductivity manifests the metallic behavior.[7] However, for large s the activated temperature dependence behavior overwhelms the metallic temperature dependence and the system shows insulating behavior.

Finally, we discuss three important issues: (1) the same physics, of course, also applies to MLG graphene, but the quantitative effects of inhomogeneity (i.e., the puddles) are much weaker since simple estimates show that the dimensionless potential fluctuation strength $\tilde{s}(\equiv s/E_F)$ is much weaker in MLG than in BLG because of the linear (MLG) versus constant (BLG) DOS in the two systems. In particular, $\tilde{s}_{\text{BLG}}/\tilde{s}_{\text{MLG}} \sim 32\sqrt{\tilde{n}}$, where $\tilde{n}=n/10^{10}$, and therefore, $\tilde{s}_{\text{BLG}} \gg \tilde{s}_{\text{MLG}}$ up to $n=10^{13}$ cm^{-2}. Direct calculations[7] show that the self-consistent values of s tends to be much larger in BLG than in MLG for identical impurity disorder. In very low mobility MLG samples, where s is very large, the insulating behavior of temperature-dependent resistivity can be observed at high densities even in MLG samples.[10,11] (2) We have neglected all quantum tunneling effects in our consideration because they are unimportant except at very low temperatures. In particular, Klein tunneling is strongly suppressed in strong disorder.[12] (3) In the presence of a BLG gap (Δ_g), the situation becomes extremely complicated since four distinct energy scales (s, E_F, k_BT, and Δ_g) compete, and any conceivable temperature dependence may arise depending on the relative values of these four energy scales. It is, however, obvious that any experimental measurement of the activation gap (Δ_a) in such an inhomogeneous situation will produce $\Delta_a \ll \Delta_g$ unless $\Delta_g \gg s$. The system is now dominated by a random local gap arising from the competition among s, Δ_g, and E_F, and no simple activation picture would apply. This is precisely the experimental observations.[13–15] (4) Our assumption of the quadratic band dispersion is valid only at low ($\lesssim 5\times10^{12}$ cm^{-2}) carrier densities for BLG systems where most of the current transport experiments have been carried out. At higher densities the band dispersion is effectively linear and the disorder effects on $\sigma(T)$ are weaker.

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[1] W. Zhu, V. Perebeinos, M. Freitag, and P. Avouris, Phys. Rev. B **80**, 235402 (2009).
[2] B. Feldman, J. Martin, and A. Yacoby, Nat. Phys. **5**, 889 (2009).
[3] S. Nam, D. Ki, and H. Lee, arXiv:1005.4739 (unpublished).
[4] M. Fuhrer (private communication); J. Zhu (private communication); P. Jarillo-Herrero (private communication).
[5] S. V. Morozov, K. S. Novoselov, M. I. Katsnelson, F. Schedin, D. C. Elias, J. A. Jaszczak, and A. K. Geim, Phys. Rev. Lett. **100**, 016602 (2008).
[6] S. Xiao, J. Chen, S. Adam, E. Williams, and M. Fuhrer, Phys. Rev. B **82**, 041406 (2010).
[7] S. Das Sarma, E. H. Hwang, and E. Rossi, Phys. Rev. B **81**, 161407(R) (2010).
[8] S. Das Sarma, S. Adam, E. Hwang, and E. Rossi, arXiv:1003.4731 (unpublished).
[9] S. Kirkpatrick, Rev. Mod. Phys. **45**, 574 (1973).
[10] Y. W. Tan, H. S. Y. Zhang, and P. Kim, Eur. Phys. J. Spec. Top. **148**, 15 (2007).
[11] J. Heo *et al.* (unpublished).
[12] E. Rossi, J. H. Bardarson, P. W. Brouwer, and S. Das Sarma, Phys. Rev. B **81**, 121408 (2010).
[13] 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).
[14] J. B. Oostinga, H. B. Heersche, X. Liu, A. F. Morpurgo, and L. M. K. Vandersypen, Nature Mater. **7**, 151 (2008).
[15] K. F. Mak, C. H. Lui, J. Shan, and T. F. Heinz, Phys. Rev. Lett. **102**, 256405 (2009).