The screening of charged impurities in bilayer graphene

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Abstract. Positively charged impurities were introduced into a bilayer graphene (BLG) transistor by \textit{n}-doping with dimethylformamide. Subsequent exposure of the BLG device to moisture resulted in a positive shift of the Dirac point and an increase of hole mobility, suggesting that moisture could reduce the scattering strength of the existing charged impurities. In other words, moisture screened off the ‘effective density’ of charged impurities. At the early stage of moisture screening the scattering of hole carriers is dominated by long-range Coulomb scatter, but an alternative scattering mechanism should also be taken into consideration when the effective density of impurities is further lowered on moisture exposure.

Bilayer graphene (BLG) has attracted much attention because of its tunable gap\textsuperscript{1}–\textsuperscript{4}. It has quadratic low-energy dispersion and finite density of states near the Fermi level\textsuperscript{5, 6}, distinguishing it from single layer graphene (SLG) that exhibits a linear electronic dispersion and vanished density of states near the Fermi level\textsuperscript{7}–\textsuperscript{9}. However, experimentally, the two systems show very similar carrier density-dependent conductivity $\sigma(n)$; for $n \sim 0$ the conductivity is almost constant (formation of a minimum conductivity plateau) and $\sigma$ is proportional to $n$ away from the charge neutrality point (CNP)\textsuperscript{9, 10}. Therefore, significant efforts, for transport theory considerations, have been put into understanding the similarities and differences between these two systems. In SLG, theoretical calculations have shown that the screened Coulomb disorder behaves as an unscreened Coulomb interaction. SLG carrier transport is dominated by Coulomb disorders from the CNP to the linear conductivity.

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area [11]. The short-range disorder dominates impurity scattering only in the high carrier density area [11]–[13]. By contrast, the screened Coulomb disorders in BLG behave similarly to two-dimensional screened Coulomb interactions. It is suggested that BLG carrier transport receives contributions from both screened Coulomb disorders and a short-range disorder all the way from CNP to the linear conductivity area [14]–[17].

Recently, Xiao et al [10] studied the charged-impurity scattering in BLG by potassium doping. On the intentional addition of charged impurities to BLG, the minimum conductivity $\sigma_{\text{min}}$ decreases in proportion to $(n_{\text{imp}})^{-1/2}$, where $n_{\text{imp}}$ is the density of a charged impurity. These results suggest the validity of charged impurity scattering and are in good agreement with theoretical predictions based on Thomas–Fermi screening approximation (TFSA) [15]. Interestingly, they also show that the magnitude of charged-impurity scattering in BLG is similar to that of SLG, but pristine BLG typically shows a lower mobility than SLG on nominally identical SiO$_2$ substrates, indicating that short-range disorder may be needed to interpret their experimental results. Also, Jang et al [18] have suggested that the measurement of the change in mobility with dielectric constant can potentially discriminate between charged impurity and short-range disorders. Here, we introduce positively charged impurities into a BLG device by $n$-doping with dimethylformamide. Subsequent exposure of BLG devices to moisture leads to a lowered scattering strength of the existing charged impurities. In other words, moisture is able to screen off the effective density of a charged impurity. We find that the constant $C$, defined as $C = \mu n_{\text{imp}}$ ($\mu$ is the carrier mobility), decreases with the effective impurity density $n_{\text{imp}}$. In the early stage of screening during which the effective density of charged impurities is still high, the conductivity is dominated by long-range scatter. However, when the effective density of charged impurities is low subsequent to an extended screening period, the TFSA is not able to describe the transport behavior and the short-range scattering effect should be taken into account in addition to the long-range.

BLG films were mechanically exfoliated from natural graphite and identified using Raman spectra (488 nm excitation) and atomic force microscopy (AFM), as previously described [19]. BLG field effect transistors were prepared by evaporating Au as the source and drain electrodes directly on top of the BLG films using hardmasks without photo-lithography processes. The device structure is shown in figure 1(a). Figure 1(b) shows a typical Raman spectrum taken at the center of the transistor channel. An as-prepared BLG transistor was treated with DMF vapors for 24 h and then sealed in a small chamber. This chamber allows for electrical probing under various gas environments. The sample was purged with Ar for 1 h in the chamber. A source–drain voltage (15 V) was then applied to electrically anneal the sample and then the BLG transistor became stably $n$-doped. Figure 1(c) presents the transfer curves measured before treatments and after soaking in DMF vapor and after stably $n$-doping with electrical annealing. For effective charge screening with moisture, Ar gas (50 ml min$^{-1}$) was bubbled into de-ionized water and sent to the chamber. All electrical measurements were made at ambient pressure at room temperature. It should be noted that the observed adsorption of water on graphene can enhance their hysteresis [20]. The enhancement of hysteretic behaviors after DMF soaking could be related to the adsorption of residual moisture in DMF liquid or to the adsorption of DMF molecules themselves.

We start with an $n$-doped BLG device whose CNP is at $-33$ V, indicating that the residual charged impurities are positive. We note that the $p$-channel mobility ($\mu_p$) is always larger than the $n$-channel mobility ($\mu_n$). Because the scattering cross section of carriers is larger in the case when they are attracted to a charged impurity than when they are repelled from it, the larger
Figure 1. (a) Schematic illustration of the device structure. (b) Optical image and Raman spectrum of a BLG device. (c) Transfer characteristics of the as-prepared BLG device before treatments and those after the following treatments: soaking in DMF vapor and \textit{n}-doping with electrical annealing. The transfer curves before and after treatments were from the same device.

\(\mu_p\) corroborates that the residual charged impurities are positive [21]. Figure 2(a) shows that the \(\sigma-V_g\) curve shifts from \(-33\) V to 0 V with increasing moisture exposure time. Figure 2(b) shows that hole mobility (extracted from the linear conductivity region) increases with exposure to moisture. Figure 2(c) shows that the width of the minimum conductivity plateau (\(\Delta V_m\)) decreases with the right-shift of \(V_{\text{CNP}}\), where \(\Delta V_m\) is obtained from the intersection of minimum conductivity with the two fitting linear regimes in \(p\) and \(n\) channels [11, 19]. In BLG, electron–hole puddles dominate the BLG properties around the charge neutral point and this
Figure 2. (a) Transfer curves for a DMF-doped BLG device recorded at various moisture exposure times ($V_{\text{CNP}}$ right-shifts with increasing exposure time); (b) $p$-channel mobility; and (c) the plateau width of minimum conductivity $\Delta V_m$ plotted as a function of $V_{\text{CNP}}$.  

will give rise to a finite ‘minimum conductivity’ [15, 17, 21], and the $\Delta V_m$ is proportional to the density of electron–hole puddles ($n^*$). The electron–hole puddles are induced by Coulomb impurities, and $n^*$ decreases with the decreasing density of charged impurities ($n_{\text{imp}}$). Thus, figures 2(b) and (c) both suggest that moisture can effectively reduce (screen off) the charged impurity density. Meanwhile, in BLG, theoretical calculations based on TFSA has shown that the density of charged impurities ($n_{\text{imp}}$) is proportional to $V_{\text{CNP}}$ [15], which implies that a decrease in charged impurities can result in a right-shift of $V_{\text{CNP}}$ for $n$-doped devices.  

It has been proved by experiments and theory that the effects of dielectric screening on short-range and long-range potentials differ in two-dimensional systems [18], [22]–[24]. Screening off long-range potentials will reduce the Coulomb interactions between carriers and charged impurities, which means that the conductivity dominated by charged impurities shall increase. However, dielectric screening does not modify the atomic-scale potential of short-range scatter, and the major effect is to reduce the screening by charged carriers; thus, the effect of scattering is enhanced, which will result in a lower conductivity at the high-density region. Our experimental results in figure 3 show that the conductivity increases monotonically at various carrier densities, e.g. $|V_g - V_{\text{CNP}}| = -70, -50, -30$ and 0 V. Therefore, the major effect of moisture exposure is to effectively screen off charged impurities, leading to a conductivity increase.
Figure 3. Conductivity versus \(V_{\text{CNP}}\) for different carrier densities.

Figure 4. (a) \(1/\mu_p\) as a function of the estimated \(n_{\text{imp}}\) with increasing exposure time (lowered \(n_{\text{imp}}\)). (b) \(\sigma_{\text{min}}\) as a function of \(n_{\text{imp}}^{-1/2}\), where the linearity in regime A agrees with long-range Coulomb scattering.

We further analyze the data quantitatively using the TFSA model, where the dominant scattering mechanism is over-screened Coulomb impurities at low carrier density [15]. The density of a charged impurity can be estimated by using \(n_{\text{imp}} = (c_g/e)V_{\text{CNP}}\), where \(e\) is the electric charge and \(c_g = 1.15 \times 10^{-8}\) F cm\(^{-2}\) is the gate capacitance per unit area for 300 nm thick SiO\(_2\). As shown in figure 4(a), we plot \(1/\mu_p\) as a function of the estimated \(n_{\text{imp}}\), where we can extract the constant \(C\) from the inverse slope based on the equation \(\mu_p = C/n_{\text{imp}}\). The constant \(C\) is \(1.5 \times 10^{16}\) V\(^{-1}\) s\(^{-1}\) in the early stage of moisture screening (before CNP reaches about \(-19\) V). Moreover, \(\sigma_{\text{min}}\) is also linearly related to \((n_{\text{imp}})^{-1/2}\) at the same stage as shown in figure 4(b). These two results are consistent with the prediction from the TFSA model, suggesting that the charged impurities dominate the scattering in linear conductivity when the effective \(n_{\text{imp}}\) is higher than \(~1.5 \times 10^{16}\) m\(^{-2}\) (in the first stage of screening). Surprisingly, when the effective \(n_{\text{imp}}\) is further lowered (\(V_{\text{CNP}}\) further shifts to the right), the constant \(C\) for the hole becomes significantly larger \((1.1 \times 10^{17}\) V\(^{-1}\) s\(^{-1}\)) as shown in figure 4(a). The change in \(C\) indicates that a different scattering mechanism is required to explain the observation. Also,
the nonlinear relationship between $\sigma_{\text{min}}$ and $(n_{\text{imp}})^{-1/2}$ at the second stage of screening urges the reconsideration or inclusion of other scattering mechanisms [10, 19].

In summary, we observed that moisture was able to reduce effectively the scattering strength of the charged impurities we intentionally introduced into BLG. This allowed us to observe the gradual change in transport behavior by using controlled screening with moisture soaking. We conclude that only when the effective charged impurity is high enough can the TFSA model describe the carrier transport properties of BLG. However, when the effective charged impurity is lower, the experimental results deviate from the prediction. Our experimental results are a powerful indication for us to also take into consideration short-range scattering.

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