Faculty and Staff Publication:

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Citation for Final Published Work:


Disorder density of states in supported graphene

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(Received 14 March 2014; accepted 7 August 2014; published online 21 August 2014)

Transport in graphene is impacted by disorder. Disorder, which can occur on supported graphene, manifests as a significant shift in the Fermi level position from the charge neutrality point (Dirac point) and leads to carrier scattering. Here, we provide a direct measurement of the disorder density of states (DOS). We show that the disorder is extrinsic to graphene and is characterized by a continuum of DOS located at the graphene-substrate interface. A key feature is a Gaussian-like DOS that causes pinning of the Fermi level and the shift in the Dirac point.

I. INTRODUCTION

Atomically thin two-dimensional (2D) materials have generated a high level of interest on account of their promise for future applications.1–5 A wide range of 2D materials has been developed since the discovery of the prototypical 2D material, the graphene.6 These materials may find applications in flexible electronics,7 high frequency electronics,8 photo detectors,9 and many other electronic and photonic applications.10

Despite the impressive breakthroughs in research concerning the electronic properties of 2D materials, there are fundamental issues impeding their use in applications. A common feature for all 2D materials is the large hysteresis in the transport characteristics.8,10,17,18 Even in a gapless material like graphene, a poor combination of synthesis and transfer technique can induce disorder, which causes hysteresis. Recent studies identified the existence of random charged impurities at the interface of graphene and the supporting substrate.19 Such impurity-induced disorders cause carrier trapping and other effects, including shift in the charge neutrality point $V_{\text{CNP}}$, scattering, and hysteresis.20–22 The nature of the disorder, however, is still not well understood. In this work, we provide a direct measure of the disorder density of states (DOS) that arise from graphene-substrate interaction. The spectroscopic signature of the interface is suggestive of an ideal graphene which is in intimate contact with a topologically disordered network characterized by a continuum of DOS. Specifically, we ascribe a Gaussian-like DOS to the pinning of the Fermi level, which causes the shift in $V_{\text{CNP}}$.

II. EXPERIMENTAL

Here, we use transient current analysis to probe the filling into and emission out of the disorder DOS. These measurements use graphene samples configured in FET geometry. By fitting the experimental data to a universal escape time which is related to energy, we extract the DOS. We fabricate FETs in a back-gated configuration by growing a high quality 100 nm of thermal SiO$_2$ on a P$^+$ Si substrate, which serves as the gate. Next, graphene grown via chemical vapor deposition is transferred onto SiO$_2$ surface. We fabricated two types of devices, type I and II. In type I devices, a water-assisted transfer technique is used, using the procedure described in Ref. 23, that results in different $V_{\text{CNP}}$ devices. We examine two such devices: type I-A and I-B. The water-assisted technique results in good adhesion of graphene with high yield. Raman spectroscopy confirms that the transferred film has properties of single-layer graphene films and is of high quality with no detectable D-band in the spectra. As a comparison to and a validation of our results from type I devices, we fabricated type II devices using isopropyl alcohol (IPA) as the transfer medium.24 The resulting films, however, consisted of large voids due to poor adhesion and produced devices with much lower yield. Next, photolithography is used to pattern graphene into an area of $\sim2.5 \times 10^{-4}$ cm$^2$, which is large enough to provide transient signals up to 1000 s. The channel is contacted with 7/70 nm thick Cr/Au contacts as source and drain terminals, which is used to measure both the charge neutrality point and the transient currents. We fabricated over 60 identical devices on several die, and the data we present are typical of the results we measure.

III. RESULTS AND DISCUSSION

Fig. 1 shows typical $I_D$ – $V_{GS}$ curves for type I-A, B and II devices measured in vacuum. Type I-A, B devices show

![Graph showing typical transfer curves for type I-A and type II devices, measured in high vacuum to eliminate any possibilities of hysteretic behavior arising from adsorption of gaseous molecules in ambient.](image-url)
large hysteresis and large off-sets in the charge neutral point ($V_{\text{CNP}}$) that range from $+45$ to $+75$ V and from $+10$ to $+25$ V for type I-A and I-B devices, respectively. Mobility for type I-A and I-B devices are $\sim 1120$ and $\sim 2450$ cm$^2$/Vs, respectively. Type II devices, on the other hand, proved to have negligible hysteresis, $V_{\text{CNP}} \sim 0$ V and mobility of $\sim 3700$ cm$^2$/Vs, and helped to confirm that hysteresis is not intrinsic to graphene. In the analysis below, we focus on type I-A and B devices and show that charge transfer occurs from graphene to a continuum of disorder DOS, which hitherto has not been demonstrated.

The large hysteresis provides a clue to how the disorder DOS can be measured. The Fermi energy $E_F$ in graphene is positioned significantly below its $V_{\text{CNP}}$ due to charge transfer, as depicted in Fig. 3(b). In the absence of disorder, the application of a gate voltage induces charges entirely in the graphene and moves $E_F$ according to the density of states of graphene. With disorder, however, the induced charges...
divide between graphene and disorder states, and the time to reach equilibrium between them is a function of the disorder DOS. Therefore, the lag in thermalization between band states in graphene and disorder states depends on time and energy, which we analyze to provide the spectroscopic signature of the disorder DOS. We implicitly assume that transport is only through graphene, and the transient current reflects the features in the disorder DOS. This is reasonable since in disordered regions transport is through the less efficient hopping between localized states. Therefore, the ideal devices for measuring the transient currents are devices with a significant shift in the Dirac point, which are provided in type I devices.

To measure the transient currents, we tie the source and drain contacts together, as shown in Fig. 2(a), so that no drift component contributes to the transient current. Here, we explain the trapping and the subsequent emission of carriers for a positive gate pulse. A detailed analysis for a negative gate pulse is provided in Appendix A. When the gate is pulsed to a positive bias, as Fig. 2(b) shows, disordered states above the equilibrium Fermi level can be probed. The polarity of the gate bias directly determines the polarity of the emission current so that either a hole current or an electron current is generated, even though the equilibrium Fermi level is established well into the valence band. The reversal in the emission current with pulse polarity is governed by the occupancy of the impurity states by either electrons or holes.

In one set of study, we pulse the gate to a positive voltage for 60 s. This time is sufficiently long so that carriers in graphene and disordered states reach equilibrium with the contact chemical potential. The contacts to graphene, which are held at 0 V for the entire duration of the measurement, serve as the reference potential. For a positive gate pulse, the Fermi level in the graphene shifts toward the charge neutrality point, filling the impurity states with electrons. We vary the pulse from +10 to +30 V. The transient current measurements begin the moment the gate pulse is set back to 0 V. The sampling time for the current is much longer than the RC time constant of the circuit, which we estimate to be in the nanosecond range. The bandwidth of the setup is capable of measuring current transients above 100 μs, but it depends on the magnitude of the transient current. The gate pulse induces carriers both in the graphene and the disordered states. The carriers in the graphene react within several RC time constants, while the carriers in the disorder states can respond very slowly, since they are thermally activated.

Figs. 2(c)–2(h) show the general features of the transient currents. For type I devices, the transient currents follow a power law. Specifically, they follow a 1/t dependence for both positive and negative gate pulses as shown in Figs. 2(c) and 2(d), respectively. For type II devices, negligible current is observed, showing that the transient currents observed in type I devices are not intrinsic to graphene. The transient currents are largely independent of gate voltage, but their sign changes with gate bias polarity. We show below that these characteristics can be described by a single parameter that relates time to the energy of the trap level. Furthermore, we show that the 1/t dependence correlates to a specific form of the DOS based on our model. The solid lines are fits to the data using the analysis given below. The weak gate bias dependence seen in Figs. 2(c) and 2(d) and the weak temperature dependence seen in Figs. 2(e) and 2(f) at short times can be modeled accurately with our model using disorder states that exhibit a largely constant DOS.

To model the emission current, we begin by analyzing the trapping and emission from a single localized energy level $E_t$ within a continuum of disorder states, as shown in Fig. 3(b). The energy is in reference to the charge neutrality point. Its occupancy is dictated by the gate bias and has an occupancy probability between 0 and 1. We use subscripts $t_1$, $t_2$, and $t_3$ in Fig. 3(b) to relate the Fermi level positions to the timing of the gate pulse shown in Fig. 3(a). $E_{t_1}$ serves as a reference level and we are able to probe the states above and below this level, depending on the polarity of the pulse. Time $t_3$ here corresponds to the start of current sampling time $t > 0$ in Fig. 2. As stated earlier, the width of the pulse is sufficiently long to establish equilibrium between graphene and the disorder states, which we denote to occur at $t_2$. When the gate pulse switches back to 0 V from a positive value, states beginning at $E_{t_2}$ empty to the conduction band (Fig. 3(b), right). The emission is to the conduction band states as these are the only states to which the trapped carriers could emit, and is consistent with transport of electrons that give rise to positive currents in Fig. 2(c). The minimum energy difference between a trap level $E_t$ to the conduction band is $2E_s$, as depicted by the vertical arrow in Fig. 3(b), right. The form for the occupancy function at $E_t$ for $t > 0$ is given as $p = p_0 e^{(-t/\tau)}$; $p_0$ is 1 to denote that we are dealing with an initially occupied state. $\tau$ depends on energy and attempt-to-escape frequency $\Omega$. It is given by $\tau = \frac{k}{e^{\frac{E_t}{kT}}}$, where $k$ is Boltzmann constant and $T$ is temperature. When the gate pulse is negative, however, holes trap below $E_{t_1}$ and the proper expression is $\tau = \frac{1}{\Omega} e^{(-E_{t_1} - E_{t_2})}$, since trapping and emission occur entirely within the valence band (see Appendix A). The emission of holes, which occurs when the negative gate pulse is set back to 0 V, is measured as a

FIG. 3. Electron emission when a positive gate pulse is applied. The band diagram shown in (b) demonstrates emission from traps as a function of channel potential, in correspondence to the potential changes in the gate, shown in (a). The shaded regions denote filled states, for both graphene and impurity states.
negative current in Figs. 2(d), 2(f), and 2(h). A detailed discussion analyzing this distinction of $\tau$ with gate polarity is included in Appendix A. Now, the emission current due to $N_t$ number of states at energy $E_t$ is given as $I = -qN_t \frac{dE_t}{dt}$, where $q$ is the charge on an electron.

If the disorder is characterized by a continuum of disorder states in energy, the expression for the electron emission current corresponding to a positive gate pulse is given as

$$I = \int_{E_t}^{E_{t1}} qN_t \frac{1}{E_t} e^{-\frac{t}{\tau}} dE_t,$$  \hspace{1cm} (1)

Equation (1) can be simplified if we consider that $N_t$ is constant in energy and the measurement times are between the emission times that correspond to $E_{t1}$ and $E_{t2}$, i.e., $\tau(E_{t2}) < t < \tau(E_{t1})$ for a positive gate pulse (the limits of the integral reverse for a negative gate pulse). Equation (1) then reduces to $I = \frac{q}{2\tau E_0} D_{CN}$ (see Appendix A), which is the functional form we observe in Figs. 2(c) and 2(d). This behavior also predicts a relatively weak temperature dependence. We confirmed the weak temperature dependence in Figs. 2(e) and 2(f) for type I-A device and in Figs. 2(g) and 2(h) for type I-B device by varying the temperature between 300 and 380 K. The solid lines, which are fits to Eq. (1), are in good agreement with the measured results. The precipitous drop in the currents at long times in Figs. 2(d), 2(f), and 2(h) occurs because the measured times are close to the time constant of the upper integration limit (see Appendix A). The resulting expression from Eq. (1) is rather striking and different than had the emission occurred from either a single or just a few localized levels. In both cases, one would have seen a much stronger exponential dependence in temperature for all times. On the other hand, the integrated behavior from a continuum of disorder states in energy, one that is constant in energy in particular, tends to strongly weaken its functional form. Simply, the $1/\tau$ dependence results from a sum of exponentials with a continuum of decay times.

We now discuss the limits of the integral for Eq. (1). These limits can be explained by a qualitative illustration of filling and emission of electrons, as described in Fig. 3. The increase in electron density as a result of a positive gate bias is pictured as lowering of graphene bands and an increase in Fermi energy from $E_{t1}$ to $E_{t2}$ (Fig. 3(b), center). When the Fermi energy is re-established at the new equilibrium level $E_{t2}$, the disorder states within the energy range $\Delta E = E_{t2} - E_{t1}$ are occupied. When the gate is set back to 0 V, the trapped electrons within $\Delta E$ emit.

To obtain a more detailed form for the DOS in Eq. (1), we perform a global fit using a single attempt-to-escape frequency $\Omega$ for both electron and hole emissions, over the temperatures examined. The procedure and the methodology adopted for performing this global fit are discussed in detail in Appendix A. We obtain a relatively low attempt-to-escape frequency $\Omega = 1 \times 10^5 \text{s}^{-1}$. This is in contrast to what is observed for localized states in bulk materials, which typically range between $10^{11}$ and $10^{13} \text{s}^{-1}$.\footnote{27} We attribute the discrepancy to the fact that the disordered region is physically separate from graphene. In addition, we obtain $E_{t1} = -0.3 \text{eV}$, $N_t(E) = 2.5 \times 10^{12} + 1.6 \times 10^{13} e^{-\frac{1}{3}(\frac{E-E_{t1}}{32})^2} \text{cm}^2 \text{eV}^{-1}$ for type I-A device and $E_{t1} = -0.26 \text{eV}$, $N_t(E) = 2 \times 10^{11} + 3 \times 10^{12} e^{-\frac{1}{3}(\frac{E-E_{t1}}{32})^2} \text{cm}^2 \text{eV}^{-1}$ for type I-B device. The values for $N_t$ are indeed large, but are consistent with the large shift in $V_{\text{CNP}}$. Fig. 6 (see Appendix C) illustrates our finding that the center of the Gaussian is positioned near the equilibrium Fermi level $E_{t1}$, suggesting pinning of the equilibrium Fermi level at the Gaussian-like disorder states.

The values for $\Delta E$ we use to fit the data are somewhat larger than what we would expect strictly from the capacitance model we provide in Appendix B. We explain this discrepancy by noting that $\Delta E$ shown in Fig. 3 and in Fig. 4 (see Appendix A) are defined under equilibrium conditions, where the occupancy is determined by the chemical potential of the contacts. During transient conditions, however, since the disorder states can have response times much longer than the RC time constant, the surface potential can deviate from the contact potential long after the gate has switched. This results in a larger $\Delta E$, as our model shows. Finally, the Gaussian terms in $N_t$ are necessary to fit the data for the negative gate pulse. Interestingly, we find that the center of the Gaussian terms is positioned near the equilibrium Fermi level $E_{t1}$, suggesting pinning of the Fermi level at the Gaussian-like DOS determines $V_{\text{CNP}}$.

IV. CONCLUSIONS

In summary, we study the disorder density of states of supported graphene using transient current analysis. We find a continuum of disorder density of states at the interface. These states are characterized by long time constants and explain the large hysteresis in the current-voltage characteristics. Furthermore, we find that the offset in the charge neutrality point is caused by the pinning of the Fermi level at a Gaussian-like disorder states.

APPENDIX A: FORMULATING A MODEL FOR ELECTRON AND HOLE EMISSION FROM A CONTINUUM OF TRAP STATES

When a positive gate pulse is applied, the barrier height is $2E_t$ for the emission of electrons from a trap level $E_t$. This is the minimum energy difference between $E_t$ and the continuum of the conduction band (Fig. 3 in the main text). Each localized state $E_i$ is described as a delocalized state in k-space and can make an emission only to the graphene’s conduction band by an electronic transition over the barrier height $2E_i$. The vertical transition (illustrated in Fig. 3(b) right) depicts an emission from each localized state through multi phonon processes where the k-dependence can be ignored.\footnote{28} Here, each individual trapped states describes a local density of states, which make up the total DOS that we calculate.

When the gate pulse is negative, however, the trapping and emission occur within an energy range $\Delta E$ in the valence band, as shown in Fig. 4. Thus, the barrier energy is $E_i - E_{t2} < \Delta E$, or proportional to $E_i$. Using these barrier heights, we define the emission times as $\tau = \frac{1}{\Omega} e^{-\frac{E_{t1}}{\tau}}$ for a
positive gate pulse and \( \tau = \frac{1}{\Omega} e^{-\frac{E_{\text{hi}}-E_0}{kT}} \) for a negative gate pulse. Here, \( k \) is Boltzmann constant, \( T \) is temperature, and \( \Omega \) is attempt-to-escape frequency.

We now derive the emission current from a continuum of disorder DOS \( N_s(E) \). We begin by considering the functional form for the occupancy of a single level \( E_i \) for times \( t > 0 \). It is given as \( p = p_0 \exp(-t/\tau); p_0 = 1 \) to denote that we are dealing with an initially occupied state. To generalize, we use \( \tau = \frac{1}{\Omega} e^{-\frac{E_1}{kT}} \), and \( E_{i1} \) and \( E_{i2} \) define the limits of the integral. The emission current due to \( N_s \) number of states at energy \( E_i \) is \( I = -qN_s \frac{d}{dt} \), where \( q \) is the charge on an electron. Now, for a continuum of disorder states in energy, the emission current is expressed as

\[
I = \int_{E_{i1}}^{E_{i2}} qN_s e^{-\frac{E_i}{kT}} dE_i. \tag{A1}
\]

Substituting for \( \tau \) we have

\[
I = \int_{E_{i1}}^{E_{i2}} qN_s \frac{1}{\Omega} e^{-\frac{E_i}{kT}} e^{-\frac{E_i}{kT}} dE_i. \tag{A2}
\]

For a constant disorder density of states \( N_s \), we have

\[
I = q \frac{kT}{t} \int_{E_{i1}}^{E_{i2}} \frac{d}{dE_i} \left[ e^{-\frac{E_i}{kT}} - e^{-\frac{E_{i2}}{kT}} \right] dE_i
\]

\[
= -q \frac{kT}{t} e^{-\frac{E_{i1}}{kT}} E_{i2}
\]

\[
= -q \frac{kT}{t} \left[ e^{-\frac{E_{i1}}{kT}} - e^{-\frac{E_{i2}}{kT}} \right]
\]

or

\[
I = -q \frac{kT}{t} \left[ e^{-\frac{E_{i1}}{kT}} - e^{-\frac{E_{i2}}{kT}} \right]. \tag{A2}
\]

For times \( \tau(E_{i1}) \ll t \ll \tau(E_{i2}) \), Eq. (A2) simplifies to

\[
I = \frac{qkT}{t}, \tag{A3}
\]

which is the functional form for the emission currents that we observe in Figs. 2(c), 2(e), and 2(g). In addition, Eq. (A3) captures the relatively weak temperature and bias dependence that we observe. In general, \( \tau(E_{i1}) \ll 1 \) ns, which is beyond our measurement capabilities. But the long time scales can be measured. In that case, Eq. (A2) has the form

\[
I = \frac{kTn_{i}}{t} e^{-\alpha t}.
\]

This is the origin for the precipitous drop in the current we observe at long times, as seen in Figs. 2(d), 2(f), and 2(h), when the gate pulse is negative. It also correctly predicts the strong temperature dependence we observe in this time range. For a negative gate pulse and at short times, we observe a stronger temperature dependence than would be allowed strictly from a constant \( n_i \), as predicted by Eq. (A3) (see Figs. 2(f) and 2(h)). This regime can be modeled by the inclusion of a Gaussian function to the constant \( n_i \).

Interestingly, the equilibrium Fermi levels \( E_{i1} \) that we calculate are close to the peak positions of the Gaussian functions in the disorder DOS for both type I-A and B devices. The implication is remarkable—the origin for \( E_{i1} \), is due to pinning in the Fermi level caused by the enhanced DOS from the Gaussian terms.

To calculate the DOS, we use Eq. (A1) to fit both electron and hole emissions for the times and temperatures we examined. We use the expressions for the two \( \tau \) defined above, and the fitting parameter are \( \Omega, E_{i1}, E_{i2}, \) and \( N_s \). \( E_{i2} \) is the only parameter that changes when the pulse polarity is switched. These parameters, including the independent variable \( t \), depend in a complex way in Eq. (A1) and cannot be linearized by applying a logarithmic transformation. In order to solve these parameters numerically, we use the following procedure for each device type:

(i) Starting with a positive gate pulse, we guess the functional form of \( N_s \), and provide a range of values for the function. We provide a range for the numerical parameters \( \Omega, E_{i1}, \) and \( E_{i2} \). We take the log of the data and minimize the residual sum of squares. This is applied to all the data taken over the temperature range we examined.

(ii) We follow the same procedure as in (i) for the opposite polarity, but here we only vary the parameter \( E_{i2} \) while keeping all the other parameters from (i). \( E_{i1} \) serves as a reference level and we are able to probe the states above and below this level, depending on the polarity of the pulse.

(iii) Procedures in (i) and (ii) lead to the optimal parameters we provide in the main text.
APPENDIX B: CAPACITANCE MODEL

We use the band diagram shown in Fig. 5(a) to develop a small signal equivalent capacitance model. The model is used to estimate the equilibrium Fermi level $E_t$. For analyzing the transient response of a large pulse, however, the capacitance model does not apply since a time dependent capacitance model is required. Instead, the transient analysis we provide in Appendix A is more appropriate.

To simplify, we consider the situation where the bias on the semiconductor is positive ($V_G > 0$), causing accumulation of holes at the p-type semiconductor/SiO$_2$ interface. With the accumulation of holes in semiconductor, most of the voltage is dropped across the oxide and the graphene, which we label as $V_{ox}$ and $V_{Graphene}$. $V_{Graphene}$ is the Fermi level in the graphene defined with respect to the charge neutrality point.

Using the band diagram shown in Fig. 5(a), we define the small-signal equivalent capacitance as

$$C = \left. \frac{d(Q_{Graphene} + Q_{it})}{d(V_{oxide} + V_{Graphene})} \right|_{V=V_G}$$

The incremental gate charge $dQ_{Silicon}$ resulting from an incremental AC gate voltage $dV_G$ is defined as $dQ_{Silicon} = (dQ_{Graphene} + dQ_{it})$, where $dV_G = (dV_{oxide} + dV_{Graphene})$. $dQ_{Graphene}$ and $dQ_{it}$ are incremental charges that reside in the graphene and at the interface trap states, respectively. Rewriting, we have

$$C = \frac{1}{dQ_{Silicon}} + \frac{1}{dQ_{Graphene} + dQ_{it}}$$

$$C = \frac{1}{C_{oxide} + \frac{1}{C_{oxide}} + \frac{1}{C_{Graphene} + C_{it}}}$$

(C)Graphene is the quantum capacitance of graphene, and for a constant disorder DOS $C_{it} = q^2 N_i$. Fig. 5(b) shows the equivalent circuit based on Eq. (B1). This model is equivalent to the small-signal capacitance from others.29

Because of the thick oxide we use, $C_{Graphene} \gg C_{it}$, $C_{oxide}$. Thus, the equivalent capacitance is just that of the oxide capacitance. Therefore, we can calculate the position of the equilibrium Fermi level $(E_t)$ using the following equation:

$$V_G - V_{CNP} \approx \frac{q}{C_{oxide}} \int_{E_t}^{0} N_i(E) dE$$

$$+ \frac{q}{C_{oxide}} \int_{E_t}^{0} DOS(E)_{Graphene} dE. \quad \text{(B2)}$$

Using Eq. (B2), we calculate $E_t = -0.4$ eV and $-0.23$ eV for type I-A and I-B devices, respectively. These values are close to the values we arrive from the fit, which are $-0.3$ eV and $-0.26$ eV for type I-A and I-B devices, respectively. Some discrepancy is expected due to the uncertainty in defining $V_{CNP}$ when large hysteresis is present.

APPENDIX C: DISTRIBUTION OF DISORDER DENSITY OF STATES

Because of the thick oxide we use, $C_{Graphene} \gg C_{it}$, $C_{oxide}$. Thus, the equivalent capacitance is just that of the oxide capacitance. Therefore, we can calculate the position of the equilibrium Fermi level $(E_t)$ using the following equation:

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APPENDIX C: DISTRIBUTION OF DISORDER DENSITY OF STATES

![Graph](image)

FIG. 6. The distribution of disorder density of states (DDOS) for type I-A and type I-B devices is illustrated against band structure of graphene, over an energy range close to the Dirac point. The position of $E_t$ in these devices is suggestive of the equilibrium Fermi level pinned at the center of Gaussian-like disorder states.
