Dirac-Point Shift by Carrier Injection Barrier in Graphene Field-Effect Transistor Operation at Room Temperature

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ABSTRACT
A positive shift in the Dirac-point in graphene field-effect transistors was observed with Hall-effect coupled with Kelvin-probe measurements at room temperature. This shift can be explained by the asymmetrical behaviour of the contact resistance by virtue of the electron injection barrier at the source contact. As an outcome, an intrinsic resistance is given to allow a retrieval of an intrinsic carrier mobility found to be decreased with increasing gate bias, suggesting the dominance of short-range scattering in a single layer graphene field-effect transistor. These results analytically correlate the field-effect parameters with intrinsic graphene properties.
Ever since graphene was identified as a promising electronic material for newly emerging applications, there have been numerous studies on its material properties and associated device performance \(^1\text{-}^8\). This has led to a study of new device architectures, and in particular, attempts to identify the role of contact materials with the graphene layer \(^9\text{-}^{11}\), suggesting that contact properties significantly limit the performance of graphene devices. At the same time, there has been equally important emphasis on the study of the intrinsic electronic properties of “bulk” graphene \(^4\text{-}^6\). For example, it has been reported that the Dirac cones of a suspended graphene layer are reshaped by carrier interaction effects associated with the carrier density in graphene \(^8\text{-}^{12}\). However, when used as the channel in a field-effect device, it is important to understand how the graphene material properties correlate with device characteristics. On this, it has been reported that the Dirac point can be shifted due to contact properties at the source and drain electrodes, which was observed either by a four-point probe measurement at low temperatures \(^13\), e.g. 60 K, or by employing different work-function metals for the source and drain electrodes \(^14\text{-}^{15}\). Although there have been previous studies on the effects of contacts on graphene field-effect devices, these have not been used to understand how the intrinsic properties, such as contact effects, Dirac point, carrier density, and intrinsic carrier mobility are influenced at room temperature by an orthogonal electric field, and their inter-relation in a field-effect structure. This constitutes the focus of the investigations presented in this work.

Using a combination of Hall-effect and Kelvin probe measurements at room temperature, we examine the symmetry in behaviour of the intrinsic resistance as a function of gate bias, retrieved from the incongruity in the contact and extrinsic resistances of the device. The resulting positive shift in the extrinsic Dirac point, from that of the intrinsic counterpart is explained by the asymmetrical behaviour of the contact resistance as a function of gate bias, suggesting the
presence of an injection barrier for electrons by virtue of the higher work-function of the nickel (source contact) electrode compared to the graphene layer. In addition, Dirac voltage is observed to be the common intercept for the linear variation in electron and hole carrier densities. From the extracted intrinsic resistance and carrier density, the intrinsic carrier mobility is retrieved showing its decrease with increasing gate bias. This suggests dominance of short-range scattering, the rate of which can increase as more carriers are induced by a higher gate bias, and has been observed previously in single layer graphene. These results provide analytical insight

Figure 1. Hall-effect and Kelvin probe measurements on the graphene field-effect transistor: (a) Micro-photo of the fabricated structure and its (b) 3-D view along with the electrical measurement set-up. Here, the channel length (L), width (W), and vertical separation (x_{12}) are 250µm, 50µm, and 120µm, respectively. (c) Measured drain current (I_{DS}) at V_{DS} = 10mV and the equivalent resistance (R_{ext} = V_{DS} / I_{DS}) as a function of V_{GS} for B = 0.45 T. (d) Measured V_{x1} and V_{x2} as a function of V_{GS}, and (e) measured V_{y1} and V_{y2} as a function of V_{GS}. Here, the measured data in Figs.1c to e have a 5% error-bar.
into the underlying physics across the field-dependent intrinsic parameters, while maintaining a consistency with earlier reports.

Fig. 1 shows representative results from Hall-effect coupled with Kelvin probe measurements of a graphene field-effect transistor; the detailed fabrication process has been published elsewhere \(^{17,18}\), and can also be found in the Supporting Information. The photomicrograph of the measured test structure is shown in Fig. 1a. The electrodes on the x-axis are used to measure the internal voltage drops and that on the y-axis are for measuring the Hall-voltage (\(V_H\)). The source (S) and drain (D) electrodes are on an oxidized high-doped silicon wafer which serves as the gate (G). The gate bias (\(V_{GS}\)) is swept at a constant drain bias (\(V_{DS}\)) in the presence of a magnetic field (\(B\)) applied orthogonally (i.e. along the z-axis) and uniformly over the graphene layer. Along with this, a drain current (\(I_{DS}\)) at a small drain bias (\(V_{DS}\)) and the equivalent extrinsic resistance (i.e. \(R_{ext} = V_{DS}/I_{DS}\)) are measured as a function of \(V_{GS}\) for \(B = 0.45 \, T\), as seen in Fig. 1c. Here, \(V_{DS}\) is fixed at 10mV which is sufficiently smaller than the gate bias and thermal voltage (about 26mV at room temperature, i.e. 300K) to satisfy the gradual channel approximation \(^{19}\). In addition, the voltage levels (\(V_{x1}, V_{x2}\)) at the nodes of \(x_1\) and \(x_2\) are measured while measuring the voltage levels (\(V_{y1}, V_{y2}\)) at the nodes \(y_1\) and \(y_2\) as a function of \(V_{GS}\), respectively (see Figs. 1d and e). Here, the extrinsic Dirac voltage (i.e. \(V^{ext}_{Dirac}\)) is retrieved at the minimum point of \(I_{DS}\) (i.e. the peak of \(R_{ext}\)), which is consistent with \(V_{GS}\) at which \(V_{y1} = V_{y2}\). Note that the extrinsic Dirac voltage can be shifted due to atmospheric absorption \(^{19}\).

Based on the experimental results discussed with Fig. 1, the gate voltage-dependence of the intrinsic parameters of the graphene device is shown in Fig. 2. Firstly, the contact resistance (\(R_C\)) is found from the difference between \(R_{ext}\) and the intrinsic resistance (\(R_{int}\)), i.e. \(R_C = R_{ext} - R_{int}\), which are as functions of \(V_{GS}\), respectively. Here, \(R_{int}\) is described by the following relation,
Figure 2. Gate voltage-dependence of intrinsic parameters of the graphene transistor and underlying band diagram illustrating electron and hole injection: (a) Retrieved $R_C$ from the difference between $R_{ext}$ and $R_{int}$. (b) Extracted concentrations of the free electrons (n) and holes (p) as a function of $V_{GS}$, respectively. (c) Conceptual band diagram to describe the blocking of electron injection at $V_{GS} = V^\text{int}_{Dirac}$. (d) Conceptual band diagram at $V_{GS} = V^\text{ext}_{Dirac}$.

$$R_{int} = \left( \frac{V_{x1} - V_{x2}}{I_{DS}} \right) \frac{L}{x_{12}},$$

where $x_{12}$ is the distance between the electrodes $x_1$ and $x_2$, as indicated in Fig.1a. It is observed that the peak point of $R_{ext}$ (i.e. extrinsic Dirac voltage, $V^\text{ext}_{Dirac}$) is right-shifted by 4 volts from that of $R_{int}$ (i.e. intrinsic Dirac voltage, $V^\text{int}_{Dirac}$) (see Figs.2a and 2b). This suggests the presence of an electron injection barrier, corresponding to the asymmetrical characteristics of $R_C$ as a function of $V_{GS}$, as can be seen in Fig.2a. Note that, if there was a hole injection barrier, this would be shifted the other way. Here, the sign of $V^\text{int}_{Dirac}$ is positive, as seen in Fig.2a, suggesting the graphene layer is p-type as its initial polarity. Note that $V^\text{ext}_{Dirac}$ is widely used as a signature of the
polarity of the graphene layer. However, it can be wrong depending on the extent of the shift voltage, i.e. $V_{\text{Shift}} \equiv V_{\text{Dirac}}^{\text{ext}} - V_{\text{Dirac}}^{\text{int}}$. For example, if $V_{\text{Shift}} > V_{\text{Dirac}}^{\text{ext}}$ (i.e. $V_{\text{Dirac}}^{\text{int}} < 0$), the graphene layer would be n-type.

To explain this theoretically, a conceptual band diagram is shown in Figs.2c and d. Firstly, Fig.2c shows the impeded electron injection at $V_{GS} = V_{\text{Dirac}}^{\text{int}}$, before $R_{\text{ext}}$ reaches its peak, in which the injected electron density ($n_{\text{inj}}$) is still less than the injected hole density ($p_{\text{inj}}$) even when $n = p = p_{\text{inj}}$ in the graphene layer at $V_{GS} = V_{\text{Dirac}}^{\text{int}}$. Here, $n$ and $p$ are the respective free electron and hole densities within the graphene layer. In contrast, $R_{\text{ext}}$ at $V_{GS} = V_{\text{Dirac}}^{\text{ext}}$ now exhibits its peak when there is sufficient electron injection due to higher gate bias, resulting in $n_{\text{inj}} = p_{\text{inj}} = p < n$, as indicated in the conceptual band diagram seen in Fig.2d. Here, a higher gate bias induces more electrons in the channel, making the Schottky barrier narrower at the source contact eluding to higher electron injection ($n_{\text{inj}}$). When $n_{\text{inj}}$ is balanced with injected holes ($p_{\text{inj}}$), the extrinsic resistance ($R_{\text{ext}}$) exhibits its peak at $V_{GS} = V_{\text{Dirac}}^{\text{ext}}$.

We now extract the free electron ($n$) and hole ($p$) densities within the graphene layer from the Hall-effect measurements (see Fig.1e), as a function of $V_{GS}$, respectively, using the following relation $^{20-22}$,

$$n, p = \left| \frac{I_{DS} B}{q(V_{y1} - V_{y2})} \right|,$$

where $q$ is the elementary charge. As seen in Fig.2b, the extrapolation lines for electrons and holes, converge at $V_{\text{Dirac}}^{\text{int}}$ rather than at $V_{\text{Dirac}}^{\text{ext}}$. This further confirms that the peak of $R_{\text{int}}$ is the intrinsic point for $n = p$. Here, the slope of the extrapolation line is proportional to the gate-capacitance ($C_G$), as labelled in Fig.2b.
Using the results in Figs.2a and 2b, a field-effect mobility of the graphene transistor can be retrieved. And there are two types of the field-effect mobility depending on whether the contact effect resides. One of them is the intrinsic carrier mobility (\(\mu_{\text{int}}\)), where the contact effect is removed. And it can be extracted with the following macroscopic definition \(^19\),

\[
\mu_{\text{int}} = \frac{1}{R_{\text{int}} Q_{n,p}},
\]

where \(Q_{n,p} = C_G \left| V_{GS} - V_{\text{Dirac}}^{\text{int}} \right| = qn \) or \(qp\). Similarly, the extrinsic mobility (\(\mu_{\text{ext}}\)) with the contact effect, as the other type, can be defined as \(\mu_{\text{ext}} = (R_{\text{ext}} Q_{n,p})^{-1}\). Fig.3a shows the extracted \(\mu_{\text{int}}\) and \(\mu_{\text{ext}}\) for electrons and holes, respectively. As can be seen, \(\mu_{\text{int}}\) is higher than \(\mu_{\text{ext}}\). In particular, the difference between \(\mu_{\text{int}}\) and \(\mu_{\text{ext}}\) for electrons is much higher than the case of the hole mobility. This reflects the asymmetrical behaviour of the contact resistance as a function of gate bias, as discussed earlier with Fig.2a. As another observation from Fig.3a, the \(\mu_{\text{int}}\) for both electrons and holes is found to be decreased with increasing the gate bias. Since the carrier density is linearly proportional to the gate voltage in the regimes A and B, i.e. \(qn, qp = C_G \left| V_{GS} - V_{\text{Dirac}}^{\text{int}} \right|\), where each carrier mobility decays (see also Fig.2b), \(\mu_{\text{int}}\) can be re-plotted as a function of the carrier density, as shown in Fig.3b. From this, we find that the \(\mu_{\text{int}}\) is inversely proportional to the carrier density. This behaviour can be explained with the short-range scattering model \(^{23-25}\). This trend is consistent with the expected behavior of a single-layer graphene transistor \(^{24,25}\).

In order to explain the intrinsic mobility behaviour, the following microscopic definition of the intrinsic mobility based on the short-range scattering model is employed \(^{24}\),

\[
\mu_{\text{int}} = \frac{q V_F^2 r_s}{|E_F|},
\]
where $v_F$ is the Fermi velocity, $\tau_S$ is the short-range scattering time, and $E_F$ is the Fermi energy. From Eqs.3 and 4, the carrier density $(n,p)$ is represented as a function of $|E_F|/\tau_S$,

$$n, p = \frac{1}{q^2 v_F^2 R_{int}} \frac{|E_F|}{\tau_S}. \quad (5)$$

With Eq.5, the correspondence between the carrier density and $|E_F|/\tau_S$ is computed for $v_F = 10^8$ cm/s, as seen in Fig.3c, clearly confirming their proportionality $^{24,25}$. These results indicate that the dominance of the short-range scattering which is proportional to the carrier density in a single-layer graphene-based field-effect transistor.

Figure 3. Field-Effect Mobility and Carrier Density: (a) Mobility ($\mu_{int}$ and $\mu_{ext}$) as a function of $V_{GS}$ calibrated with $V_{Dirac}^{int}$. Here, the regime A and B show the mobility decays. (b) Intrinsic mobility ($\mu_{int}$) vs. the carrier density $(n, p)$. (c) Fermi energy per scattering time ($|E_F|/\tau$) as a function of the carrier density $(n, p)$. Here, the dotted lines denote the fitted trends.
A combination of Hall-effect and Kelvin probe measurements shows a shift in the extrinsic Dirac point from the intrinsic counterpart, suggesting the presence of an electron injection barrier and hence asymmetrical contact resistance as a function of gate bias. This indicates that the Dirac point is shifted by not only the polarity of the graphene layer but also by the contact properties at the source and drain junctions. In addition, the intrinsic carrier mobility is found to be decreased with increasing the gate bias, which is due to an increased short-range scattering of induced carriers in a single layer graphene-based field effect transistor. These results provide analytical physical insight into the correlation between the field-effect parameters and intrinsic material properties.

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Author Contributions:

S. L. and A. N. designed the experiment and performed analysis. J. A.-W., P. B.-W., A. A. S., and S. H. prepared the samples. D. H checked the theory and analysis. S. L. and A. N. wrote the manuscript while getting feedback from all other authors.

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