Inhomogeneous screening of gate electric field by interface states in graphene FETs

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Abstract
The electronic states at graphene-SiO$_2$ interface and their inhomogeneity is investigated using the back-gate-voltage dependence of local tunnel spectra acquired with a scanning tunneling microscope. The conductance spectra show two, or occasionally three, minima that evolve along the bias-voltage axis with the back gate voltage. This evolution is modeled using tip-gating and interface states. The energy dependent interface states’ density, $D_{it}(E)$, required to model the back-gate evolution of the minima, is found to have significant inhomogeneity in its energy-width. A broad $D_{it}(E)$ leads to an effect similar to a reduction in the Fermi velocity while the narrow $D_{it}(E)$ leads to the pinning of the Fermi energy close to the Dirac point, as observed in some places, due to enhanced screening of the gate electric field by the narrow $D_{it}(E)$. Finally, this also demonstrates STM as a tool to probe the density of interface states in various 2D Dirac materials.

Keywords: graphene, interface States, scanning tunneling spectroscopy

Supplementary material for this article is available online

(Some figures may appear in colour only in the online journal)

1. Introduction

Monolayer graphene is the first experimentally accessible two-dimensional material [1], which, together with its linear Dirac-Fermion-like dispersion near Fermi energy, offers access to very exciting physics and applications, such as high speed electronics and photonic devices [2–5]. With the objective to investigate the exotic state of electrons in graphene driven by inter-electron interactions, graphene field-effect-transistors (Gr-FETs) of extremely high mobility and free path of tens of microns have been realized [6–8]. However, one still cannot access the Dirac point (DP) in graphene with enough energy resolution due to residual disorder and inhomogeneities [9].

In commonly made Gr-FETs with SiO$_2$ gate, the disorder and residual doping are mainly attributed to the interface, defect and/or trap states at graphene-SiO$_2$ interface [10–14]. The exact nature of these states depend on the detailed surface structure of amorphous-SiO$_2$ [15–20] together with the species, such as O$_2$ and H$_2$O, adsorbed on it [12]. In fact, the nature of doping has been controlled by interface engineering [21]. The trapping/detrapping of electrons in these interface states is responsible for noise [22] as well as hysteresis in resistance [10–14] and capacitance [23, 24] in Gr-FETs. A direct probe of the disorder due to interface states is quantum capacitance [25–28]. As a result of this disorder in SiO$_2$ based graphene devices several research groups are now using either suspended graphene or the one encapsulated between hexagonal boron nitride layers [7, 8]. However, the disorder, like interface or trap states, and related physics is common to all graphene devices [29] although in the later devices the amount of disorder is much less.

In 2D materials, scanning tunneling microscope (STM) can directly access the electronic states, by local tunneling spectroscopy, which eventually control the bulk electronic transport properties. Electronic inhomogeneities in graphene as arising from charge disorder due to interface and defect states have been investigated by several STM groups [30–32]. Since the carrier density in graphene is small near DP, the tip-gating effect on the spectra is significant [32, 33]. Additional tip-gating related effects such as ionization of impurities on graphene [34], quantum confinement effects [35], have also
been reported. In addition, as discussed here, the interface states also affect the local tunnel spectra through their weak interaction with graphene.

In this paper, we present a systematic study of local tunnel spectra on several atomically resolved single layer graphene (SLG) surfaces with back-gate voltage ($V_g$). Local tunnel spectra show multiple minima that move along the tip-bias axis as a function of $V_g$. This evolution of the minima is modeled using tip-gating and an energy dependent interface states’ density. The later is found to be spatially inhomogeneous with a narrow energy-width in some places pinning the graphene Fermi energy and broad width in other places leading to an apparent reduction in Fermi velocity. Finally, we discuss the possible origin and implications of these inhomogeneous interface states.

2. Experimental details

Graphene was mechanically exfoliated from Kish graphite using adhesive tape on n-doped Si substrate with 300 nm thick thermal oxide. The substrate was first cleaned either with oxygen plasma (50 W) for 5 min or by dipping into the freshly prepared piranha solution for 5 min followed by rinsing in de-ionized water and blow drying. Exfoliation was done within 30 min of the cleaning process. The Raman spectrum of a sample, in figure 1(c), shows characteristic Raman features, i.e. G and 2D bands, of SLG. The absence of D-peak indicates lack of defects. Single Lorentzian fit of the 2D peak (see inset figure 1(c)) with I(2D)/I(G) = 3.4 confirms SLG [36]. Initially, to avoid contamination from wet chemical process and resist used in lithography steps, a mechanical shadow masking method [37] was followed for electrical contacts. Cr(10 nm)/Au(50 nm) was deposited twice after masking the masking method [37] was followed for electrical contacts.

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After deposition of Cr/Au lift-off followed by Ar/H₂ (5% H₂ in contacts is negligible [38]. Later on we found that electron beam of aligning graphene with STM tip under optical microscope. were kept perpendicular to each other in the two steps for ease of aligning graphene with STM tip under optical microscope.

Several freshly prepared as well as stored, in vacuum desiccators over several months time period, SLG samples were studied using a homemade vacuum-STM at room-temperature with an integrated 2D-positioner [39] for coarse sample movement. The STM tip was aligned with the gold pad near graphene in ambient conditions followed by transfer of STM to the vacuum chamber. The chamber was then pumped to a pressure lower than $5 \times 10^{-4}$ mbar using a cryopump attached to the chamber. Guided by the STM images, the sample was coarse-adjusted in situ to align graphene in front of the STM tip and also to explore larger area. As shown in figure 1(d), $V_b$ was applied to the Si substrate with 270 kΩ series resistance. The STM bias voltage ($V_b$) was applied on graphene while the tip was kept at (virtual) ground potential. Electrochemically etched and hydrofluoric acid treated tungsten wire (0.25 mm diameter) was used as the STM tip. The apex radius of the tip was found to be in 30–50nm range from electron microscopy. The tunnel conductance spectra were acquired by using 20 mV amplitude ac-modulation with the dc bias voltage. The general reproducibility of the spectra was confirmed on different regions of several samples with a number of tungsten tips.

3. Experimental results

We performed STM/S on twelve different SLG devices and transport on a few devices prepared by either piranha or oxygen-plasma cleaning. The sheet resistance of a device made using piranha cleaning, in figure 1(e), shows the expected [40] n-type doping with DP occurring at $V_g = -14$ V. SLG device made by oxygen plasma cleaning, in figure 1(f), shows large p-type doping with DP occurring at $V_g = 53$ V. Oxygen-plasma treatment increases the silanol group density by removing hydrocarbon contaminants on SiO₂ and hence p-dopes graphene [15].

The STM/S was done at lateral distances of more than 1 μm from the metal-graphene interface so as to avoid the influence of the metal contact. Large area topographic image of SLG in figure 2(a) shows 0.4nm rms roughness due to underlying SiO₂ [30, 33]. The inset shows a zoomed-in image showing atomically resolved surface with honeycomb structure. Figures 2(b)–(d) show the evolution of the tunnel spectra with $V_g$ acquired on three different samples with different dopings.

Figure 2(c) shows tunnel spectra on a SLG device with large p-doping, where the two minima move towards each other when $V_g$ is increased from $-39$ V and the two eventually meet near $V_g = 40$ V. The tunnel spectra in figure 2(d) also show two minima but only $V_b^2$ changes with $V_g$ while $V_b^1$ remains fixed near $V_b = 0$ for $V_g$ values from $-53$ to $46$ V. From our study on these SLG devices we find that the tunnel spectra on devices prepared by piranha cleaning show mostly n-type doping (i.e. $V_b^1$ at positive bias voltage for $V_g = 0$) consistent with their transport behavior while those on oxygen plasma processed devices show mostly p-type doping, again consistent with the respective transport behavior. The evolution of the two minima with $V_g$ in local spectra shows significant variation even on a given SLG device, which cannot be modeled by simple tip-gating effect [33] as discussed further.

$V_g$ dependence of the local tunnel-spectra has been studied and modeled by several STM groups [32, 33, 35] using tip-gating effect. In this model the primary minima position shows $v_F \sqrt{|V_g - V_g^D|}$ dependence on $V_g$ with $v_F$ as the Fermi velocity of graphene and $V_g^D$ is a local constant dependent on local doping as described later. In order to fit the $V_g$-dependence of the two minima as arising from tip-gating effect we will need...
1.0 \times 10^5 \text{ m s}^{-1} \leq v_F \leq 7.5 \times 10^5 \text{ m s}^{-1}. From the Friedel oscillations, seen using STM, near atomic defects in Ar\(^+\) ion-irradiated graphene, Tapaszto \textit{et al} [41] found three times reduction in \(v_F\) and attributed it to induced disorder in hopping amplitudes. However, we cannot understand the origin of this ten times reduction in \(v_F\) as our exfoliated graphene is unlikely to have such atomic defects. Such hard defects should also give rise to the defect mediated D-peak in the Raman spectra which we do not see at all, see figure 1(c). Another drawback of \(v_F\) reduction in the simple tip-gating model is the disappearance of secondary minima. The later can be recovered but only with significant reduction in tip-sample distance, which again cannot be justified. Thus we conclude that the simple tip-gating model cannot explain various tunnel spectra observed in our experiments. On the other hand, interface states have been invoked for understanding various experimental results as discussed earlier, which we incorporate in the simple tip-gating model to understand our local spectra as follows.

4. Effect of interface states on tunnel spectra

As discussed earlier, the interface states between graphene and SiO\(_2\) can arise from the detailed SiO\(_2\) surface structure and adsorbates. The STM tip has two interactions with graphene, namely, electrostatic (tip-gating) and tunneling. The former directly affects the filling of the graphene states and the interface states, which weakly interact with graphene, while the electron tunneling happens only between graphene and the tip states. The interface states will not directly affect the tunnel conductance if the equilibration rate of the tunneling electrons is much larger than the electron transfer rate between graphene and the interface states. The electron equilibration time within graphene is expected to be sub-ns while typical electron transfer times for the later process are more than \(\mu\text{s}\) order. This is in contrast to the tunnel spectra directly probing the defect’s states [34] when the defect lies on top of graphene the exchange of electron between tip and graphene.
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either directly or via graphene states, happens much faster. The other measurement times in the STM/S are much larger. Thus in this quasi-static equilibrium limit the occupancy of the interface and graphene states will be governed by the same Fermi distribution function. Here, we discuss a model that incorporates the effect of energy dependent interface-state density $D_{it}(E)$ (defined as the number of states per unit area and per unit energy) and tip-gating to find $V_b$ and $V_g$ dependent tunnel conductance.

The density of states of SLG near the DP is given by,

$$N(E) = 2|E + E_F|/\pi(hv_F)^2$$

with $E_F$ as the Fermi energy of graphene measured from the DP. A change in $V_g$ by $dV_g$ causes a change in $E_F$ by $dE_F$ and a change in total charge density by $d\sigma$. The latter change occurs due to change in filling of the graphene, as well as the interface trap, states. $dV_g$ is shared between $dE_F$ and the potential drop across the gate oxide giving $dV_g = -(d\sigma/C_{ox}) + (dE_F/e)$. Here $e$ is the magnitude of electronic charge and $C_{ox} = \kappa \varepsilon_0/d_{ox}$ with $\kappa \approx 4$ as the dielectric constant of SiO$_2$, $\varepsilon_0$ as the free space permeability and $d_{ox}$ as the SiO$_2$ thickness. With $d\sigma$, due to graphene and interface states, as $d\sigma = -e[N(E_F) + D_{it}(E_F)]dE_F$ we get $dV_g = (e/C_{ox})[N(E_F) + D_{it}(E_F)]dE_F + (dE_F/e)$. On integration this gives, $e(V_g - V_g^0) = \text{Sgn}(E_F)[e^2E_F^2/C_{ox}\pi(hv_F)^2] + (e^2/C_{ox}) \int_0^{E_F} D_{it}(E)dE + E_F$. Here, $V_g^0$ is a local constant, whose value depends on various contact potentials and the interface trap density. It can be found by $V_g$ required to make $E_F$ coincide with the DP. In addition, incorporating the effect of tip-gating [33], we get,

$$e(V_g - V_g^0 - \beta V_b) = \text{Sgn}(E_F)[e^2E_F^2/C_{ox}\pi(hv_F)^2] + E_F$$

$$+ e^2/C_{ox} \int_0^{E_F} D_{it}(E)dE.$$  

Here, $\beta = d_{ox}/z\kappa$ with $z$ as the tip-sample separation. Right hand side of equation (2) is a monotonically increasing

Figure 2. (a) STM image of SLG (0.6 V and 0.1 nA, 0.8 × 0.8 µm$^2$) with a line-cut along the marked line. The inset shows a zoomedin image (0.4 V and 0.1 nA, 7.9 × 7.9 nm$^2$) showing honeycomb graphene lattice. (b)–(d) show $V_g$ variation of the local tunnel spectra on three different SLG devices having moderate n-, large p-, and large n-type doping, respectively. All the spectra were taken with same set-point current of 0.1 nA but at $V_b =$ 0.6, 0.8, 0.8 V, respectively. The black and red arrows mark the location of primary and secondary minima, respectively. See the supplementary material for more detail (stacks.iop.org/JPhysCM/29/385302/mmedia).
single valued function of $E_F$. Thus the tunnel conductance at given $V_b$ and $V_g$ and at zero temperature will be given by $G(V_b, V_g) \propto |eV_b + E_F(V_g, V_b)|$.

We take an energy-localized $D_n(E)$ with Gaussian shape, i.e.

$$D_n(E) = \frac{D_{n0}}{\sqrt{2\pi}\delta}\exp\left(-\frac{(E - \epsilon_c)^2}{2\delta^2}\right). \tag{3}$$

Here $D_{n0}$ represents total defect state density, $\epsilon_c$ is the center energy and $\delta$ represents the energy-width. In this case equation (2) gives,

$$e(V_g - V_D^0 - \beta V_b) = \frac{\text{Sgn}(E_F) e^2 E_F^2}{C_{ox}\pi(h\nu_F)^2} + E_F + \frac{e^2D_{n0}d_{ox}}{2\epsilon_0} \left[ \text{erf}\left( \frac{\epsilon_c}{\sqrt{2}\delta}\right) + \text{erf}\left( \frac{E_F - \epsilon_c}{\sqrt{2}\delta}\right) \right]. \tag{4}$$

$v_F$ is no longer a fitting parameter and we take it as $1.0 \times 10^6 \text{ m s}^{-1}$. Figure 3(a) shows $E_F$ as a function of $V_g - V_D^0$ at $V_b = 0$ for two different $D_n(E)$ (shown in inset). $D_{i1}(E)$ is broad in energy and $D_{i2}(E)$ is narrow. For $D_{i1}(E)$, dependence of $E_F$ on $V_g - V_D^0$ shows qualitatively similar behavior as $D_{i2}(E) = 0$ except the reduced movement of the primary minima with $V_g$. The narrow $D_{i2}(E)$ shows pinning of $E_F$. The $V_g$-range over which $E_F$ remains pinned depends on $D_{i0}$ and $\delta$ while pinning-energy depends on $\epsilon_c$. Curves 1, 2 and 3 of figure 3(b) show the calculated tunnel conductance for $D_{i2}(E)$ at different $V_g$ voltages. Here $V_g^0$ moves with $V_g$ while $V_g^c$ remains pinned near $V_g = 0$. At negative $V_g$ there is a feature showing the possibility of another minima and moving away from $V_g^0$ when $V_g$ changes from 10V to −20V. Similar features and movement with $V_g$ have been seen in experiments, see figure 2(d).

5. Analysis and discussion

Figure 4 shows the evolution of $V_g^0$ and $V_g^c$ of experimentally observed tunnel spectra, on three representative devices, together with the fitting. The plots of figures 4(a)–(c) correspond to those in figures 2(b)–(d), respectively. Figures 4(c) and (d) are from spectra taken at two different locations of the same sample. We note that the slope of $V_g^0$ versus $V_g$ line does not change much between spectra while the $V_g$-dependence of $V_g^c$ has significant variation. The former depends on $\beta = d_{ox}/2\epsilon_0$, which does not change significantly between spectra while the later depend on $\delta$, which has significant variation. Small $\delta$ leads to a pinned $E_F$ and a large $\delta$ only slows down the $V_g$ dependence of $E_F$. Inhomogeneity in $\delta$, see (figures 4(c) and (d)), for a given sample is common to all our studied devices and indicates an inhomogeneous screening of the gate electric field by the interface states. Further evidence of interface-defects changing their state can be seen from the evolution of the STS maps with gate voltage [42].

We have mainly focussed on the agreement between broad spectral features, i.e. two minima, while the spectra as a whole, particularly near the primary minima has significant rounding as compared to what is expected, see figure 3(b). This rounding is significantly more than that expected from thermal smearing. The graphene DOS near the Dirac point is extremely small and the extrinsic effects, such as defect states, potential disorder (which makes a precise access to Dirac point impossible even in bulk graphene [9]), or even tip-doping, can influence the spectra. At this point we have not attempted to incorporate these effects in the simulated spectra.

The DFT as well as ab initio calculations show broad or a constant $D_n(E)$ arising from silanol group on the SiO2 surface and narrow $D_{i}(E)$ just above DP as arising from a partially occupied state below the conduction band minimum of SiO2 [19, 20]. The later can donate electrons and pin $E_F$ above DP. The narrow states can come from the formation of three-fold coordinated O-atoms in amorphous-SiO2 [20] due to higher local concentration of Si-atoms or nearby O-vacancies. The energy-localized trap/interface states with a Gaussian distribution have also been invoked recently [29] to understand the transport in graphene constrictions.

Figure 3. (a) shows the effect of $D_n(E)$ on movement of $E_F$ as a function of $(V_g - V_D^0)$ at $V_b = 0$ with the inset showing $D_{i1}$ ($D_{i0} = 5 \times 10^{12} \text{ cm}^{-2}$, $\epsilon_c = 0.0 \text{ eV}$ and $\delta = 0.2 \text{ eV}$), $D_{i2}$ ($D_{i0} = 5 \times 10^{12} \text{ cm}^{-2}$, $\epsilon_c = 0.04 \text{ eV}$ and $\delta = 0.02 \text{ eV}$) and graphene density of states (N(E)) in $10^{13} \text{ cm}^{-2} \text{ eV}^{-1}$ units. (b) shows the calculated tunnel conductance at $V_g^D = -58 \text{ V}$ and $\epsilon = 0.75 \text{ nm}$ for gaussian $D_{i2}(E)$ (curve-1, 2 and 3). The inset in (b) depicts experimental spectra (with small offset for clarity) showing two minima with the primary minimum remaining pinned and the secondary minimum shifting with $V_g$. 

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6. Conclusions

In conclusion our STM/S study on several atomically resolved SLG surfaces with back-gate show multiple minima in local spectra that move along the tip-bias axis as a function of $V_g$. The evolution of the minima is successfully modeled using tip-gating and an energy dependent interface states' density. The later is found to be inhomogeneous and even leads to a pinning of the graphene Fermi energy in some places. Such inhomogeneous screening of the gate electric field by the interface states will lead to a non-linear change in the carrier density with gate-voltage together with a mobility change due to the change in potential-landscape seen by graphene due to the change in filling, with $V_g$, of the interface/trap states. Finally, the dependence of tunnel spectra on the density of the interface states that exchange electrons with graphene can be a useful tool for probing such interface states in different 2D Dirac materials.

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Figure 4. Variation of the $V_b^1$ (squares) and $V_b^2$ (rhombus) with $V_g$ with the solid lines showing the calculated position of the two minima. The plots in (c) and (d) are from two locations of the same sample while those in (a) and (b) are from two other samples. The plot in (b) is from O$_2$-plasma cleaned sample, while the other three are from piranha cleaned ones. The plots in (a), (b) and (c) are derived from (b), (c) and (d), respectively, of figure 2. The fitting parameters $z$, $V_b^1$, as well as used in $D_{\text{it}}(E)$ (see equation (3)) are shown in respective plots.

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