Signatures of Disorder in the Minimum Conductivity of Graphene

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ABSTRACT: Graphene has been proposed as a promising material for future nanoelectronics because of its unique electronic properties. Understanding the scaling behavior of this new nanomaterial under common experimental conditions is of critical importance for developing graphene-based nanoscale devices. We present a comprehensive experimental and theoretical study on the influence of edge disorder and bulk disorder on the minimum conductivity of graphene ribbons. For the first time, we discovered a strong nonmonotonic size scaling behavior featuring a peak and saturation minimum conductivity. Through extensive numerical simulations and analysis, we are able to attribute these features to the amount of edge and bulk disorder in graphene devices. This study elucidates the quantum transport mechanisms in realistic experimental graphene systems, which can be used as a guideline for designing graphene-based nanoscale devices with improved performance.

KEYWORDS: Graphene, minimum conductivity, disorder, edge states, evanescent, scaling

Conductivity of a material offers insights into the energy band structure and exhibits different behaviors depending on the scattering mechanisms involved. Conductivity is independent of geometry for conventional bulk materials, but scales with length for low-dimensional materials under ballistic transport conditions (e.g., carbon nanotubes), where no scattering occurs within the channel. The situation becomes more intriguing for graphene at the Dirac point, where the density of states (DOS) is zero. Despite zero carrier density, the conduction takes place by the evanescent modes tunnelling through the Dirac point and leads to a counterintuitive finite minimum conductivity ($\sigma_{\text{min}}$). In the ideal disorder-free case, $\sigma_{\text{min}}$ scales monotonically with the width-to-length ratio ($W/L$) of the channel. In reality, disorder in experimental graphene structures has been found to be inevitable and profoundly impacts carrier transport, particularly for the off-state. However, there has not been a comprehensive understanding on the scaling of $\sigma_{\text{min}}$ over a broad width and length parameter space, where disorder dictates the transport behavior. Here, we report an experimental investigation of graphene ribbons in which $\sigma_{\text{min}}$ is governed mainly by the impacts of edge and bulk disorder. In distinct contrast to the disorder-free case, our investigation reveals a strong nonmonotonic $W/L$ dependence along with an overall enhancement in $\sigma_{\text{min}}$ characterized by a peak and saturation. Our model for the disordered graphene system agrees quantitatively with the experiments. In addition, our framework accommodates previous findings on minimum conductivity for graphene ribbons in different geometrical limits. This study probes into the behavior of disordered graphene system, elucidates the transport mechanism, and provides insights for improving the performance of graphene nanoelectronics.

Over the years, experiments have provided evidence that disorder significantly impacts the off-state carrier transport in graphene structures. Minimum conductivity of graphene, initially thought to be a universal constant, is only so when external perturbations such as disorder and most notably contacts are absent. However, perturbations are ubiquitous for experimental graphene devices, and their impact on the finite size scaling of $\sigma_{\text{min}}$ is unclear due to the lack of experimental data in the disorder dominated regime. To address this issue, we have fabricated bottom-gated graphene devices with dimensions larger than typical length scales of disorder and covered a wide range of width-to-length ratio ($W/L = 0.1 - 2.6$). A two-probe setup rather than four-probe is employed since only the former explicitly accounts for the contacts, which are the origins of the evanescent modes and responsible for the observed width scaling behavior.

Throughout this entire work, we focus on single-layer graphene devices on SiO$_2$/Si substrates. Figure 1a shows a scanning electron microscopy (SEM) micrograph of the structure under investigation. Typical transfer characteristics at different temperatures are shown in Figure 1b, where $\sigma_{\text{min}}$ shows little temperature dependence. The width-to-length ratio is used as a metric to investigate the scaling behavior of $\sigma_{\text{min}}$ (discussed in Supporting Information).

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Supplement I) with the experimental findings shown in Figure 1c. Both the transfer length ($L_T$) and the contact resistance ($R_C$) are considered when evaluating $L$ and $\sigma_{\text{min}}$. (Supporting Information Supplement II). The following distinct features are immediately noticed: (i) $\sigma_{\text{min}}$ has a nonmonotonic dependence on $W/L$ with $\sigma_{\text{peak}} \sim 8 \ (4e^2/\pi h)$ at $W/L \approx 0.5$; (ii) $\sigma_{\text{min}}$ saturates for large $W/L$, denoted as $\sigma_{\text{sat}} \sim 4 \ (4e^2/\pi h)$. Very few previous reports can be found on the geometrical scaling of $\sigma_{\text{min}}$. Figure 1c includes previous published data$^{10,10}$ reproductions from a previous report. The red solid curve indicates theory for disorder-free graphene ribbons$^5$ with the enhancement from bulk disorder.

First, it is useful to recall that there are two types of states in a graphene ribbon, namely edge states and bulk states. Edge states are highly conductive propagating states residing along nonarmchair edges.$^{16-18}$ The conductance from edge states is independent of $W$, and therefore would dominate as $W/L$ approaches zero and lead to a diverging $\sigma_{\text{min}}$. Bulk states on the other hand are less conductive evanescent states with the number of states proportional to $W$. In the large $W/L$ regime, bulk states dominate the transport and lead to a constant $\sigma_{\text{min}}$ of $4e^2/\pi h$ in the disorder-free case.$^5$ Combining these two types of states is the key to understand the experimental observations. Note that both types of states are involved in transport through zigzag ribbons, whereas only bulk states are responsible for transport in armchair ribbons.

Edge disorder is a result of the inevitable edge roughness from imperfect cleaving or top-down fabrication. It is known to cause gaplike transport behavior due to localization or quantum dot formation.$^{19-22}$ A zigzag edge is more common for graphene ribbons than an armchair edge since ribbons with edges “between” zigzag and armchair tend to acquire zigzag-type boundaries.$^{16}$ In the absence of disorder, $\sigma_{\text{min}}$ is predicted to increase or decrease monotonically with $W/L$ for armchair or zigzag ribbons$^6$ (black curves in Figure 2a). We have performed quantum transport simulations using a non-equilibrium Green’s function (NEGF) formalism to model the edge and bulk disorder in 2D graphene sheets with the numerical implementations described in Supporting Information Supplement IV and V. All simulations are performed for $T = 0 \ \text{K}$ (see Supporting Information Supplement III). Indeed, edge disorder immediately gives rise to a nonmonotonic behavior for zigzag ribbons, while $\sigma_{\text{min}}$ for armchair ribbons remains monotonic as a function of $W/L$, albeit with a lower conductivity (Figure 2a). Interestingly, our experimental devices (Figure 1c) exhibit a scaling behavior that is consistent only with edge-disordered zigzag ribbons, and none of the devices fall into the “armchair” trend as predicted by the green circles in Figure 2a. This observation can be viewed as a manifestation of the fact that the zigzag boundary condition, where the edge is characterized by a majority sublattice and hence accommodates edge states, is generic for experimental graphene ribbons.

An analytical description for the impact of edge disorder can be obtained by considering a width-dependent transport gap. $\sigma_{\text{min}}$ for a zigzag ribbon with edge roughness can be described as follows$^6$

$$\sigma_{\text{min}} = \frac{\pi L}{W} \sum_{n=2}^{\infty} \cosh^{-2} \left( \frac{n\pi}{W} L \right) + \frac{1}{2} \cosh^{-2} \left( \frac{\varepsilon_{\text{loc}}}{h v_f} L \right)$$

given in units of $4e^2/\pi h$. $\varepsilon_{\text{loc}} = \alpha/W^d$ is the Anderson localization-induced transport gap with $\alpha \sim 1 \ \text{ÅeV}^d$ is the Anderson localization-induced transport gap with $\alpha \sim 1 \ \text{ÅeV}^d$ as estimated in ref 20. Although temperature should have an effect on $\varepsilon_{\text{loc}}$ due to dephasing, however, the thermal broadening due to Fermi function.
smearing is negligible as discussed in Supporting Information Supplement III. Since \( \varepsilon_{\text{loc}} \) increases with decreasing width, \( \sigma_{\text{min}} \) for narrow ribbons is more suppressed than for wide ribbons, resulting in decreasing \( \sigma_{\text{min}} \) values for small \( W/L \). Furthermore, the edge states wave function decays exponentially from the edges with a characteristic length proportional to \( W \) at a given Fermi energy \( (E_F) \), making the narrower ribbons more susceptible to edge disorder. Another way to form a transport gap in graphene nanoribbons (GNRs) is to combine small confinement-induced energy gaps with potential inhomogeneities.19,21,22 We believe that the transport gap in our case is mainly due to edge localization for the following reasons: First, the smallest \( W (\approx 100 \text{ nm}) \) is much larger than the effective diameter of a charging island \((\approx 20 \text{ nm})\) near the Dirac point\(^8\) and is unlikely to form isolated quantum dots. Second, the confinement-induced energy gap for \( W \geq 100 \text{ nm} \) is too small to block the band-to-band tunnelling current. Therefore, it is likely that edge roughness scattering is responsible for \( \sigma_{\text{min}} \) reduction for zigzag ribbons of small \( W/L \). The length scaling of \( \sigma_{\text{peak}} \) is found to follow a power-law relationship, as shown in Figure 2b. Extrapolation to \( L = 1 \mu \text{m} \) yields \( \sigma_{\text{peak}} \approx 4.2 \,(4e^2/h) \) for reasonable assumptions of root-mean-square edge roughness \((\text{rms} = 7.5 \text{ nm})\) and autocorrelation length \((\text{AL} = 2 \text{ nm})\),24,25 Note that since the peak position \((\text{not the peak height})\) is roughly a constant when the rms roughness is varied, we deduce that \( \sigma_{\text{peak}} \approx W^{0.46} \) (as \( W/L \approx \text{constant at the peaks} \)) , a finding that will become relevant in subsequent discussions.

Bulk disorder in graphene arises from charge inhomogeneity at the graphene–substrate interface, with a typical potential fluctuation of \( \pm 40 \text{ meV} \), the origin of which could be due to charge impurities on \( \text{SiO}_2 \). Local electron- and hole-rich regions, called electron- and hole puddles, are induced in graphene with typical diameters of \( \approx 150 \text{ nm} \).\(^4\) For any given \( E_F \) line-up in the off-state, percolation paths exist from the source to the drain through connected electron and hole puddles. As a result, current flows preferably through the regions of unperturbed potential to avoid the additional resistance associated with the pn interfaces at puddle boundaries,\(^26\) as demonstrated by Figure 3a. Therefore, bulk disorder has a strong influence on \( \sigma_{\text{min}} \).

Figure 3b shows the NEGF simulation results for \( \sigma_{\text{min}} \) versus \( W/L \) for ribbons with bulk disorder. Adding bulk disorder to graphene leads to higher conductivity;\(^27\) which is exactly the opposite trend compared to other material systems. The reason for this unique behavior lies in the fact that graphene is a gapless material, where carriers can tunnel almost unimpeded from the conduction band into the valence band. Different \( E_F \) line-ups with respect to the Dirac point lead to a variation of DOS and thus conductivity in different puddles. The average DOS in the puddles increases as the magnitude of bulk disorder increases due to the linear energy dispersion of graphene. This leads to an enhancement in \( \sigma_{\text{min}} \) with increasing bulk disorder over the entire \( W/L \) range (Figure 3b). Effective medium theory is able to capture the main features of this phenomenon (Figure 3c), with equations in Supporting Information Supplement VI. It is a remarkable coincidence that the saturation feature of the bulk states conductivity persists for both disorder-free and bulk-disordered ribbons, despite different mechanisms involved. The former is due to evanescent transport, while the latter is dominated by percolation. For smaller devices (e.g., \( L < 500 \text{ nm} \)) that only contain a few puddles, the potential is relatively uniform across the channel and the impact of bulk disorder is minimal. As a result, \( \sigma_{\text{min}} \) is markedly lower than for large devices, just like the disorder-free case\(^8\) (e.g., “+” in Figure 1c). Using the effective medium model, we obtain an effective conductivity \( \sigma_{\text{eff}} \approx W^{0.16} \) for \( W/L < 1 \), assuming \( \sigma_{\text{f}}/\sigma_0 = 0.01 \) and \( L \approx 30 \, \mu \text{m} \) (puddle radius). Rigorous NEGF simulations assuming reasonable bulk disorder parameters extrapolate to \( \sigma_{\text{eff}} \approx 4.3 \,(4e^2/h) \) for \( L = 1 \mu \text{m} \). Quantitative agreement between experiments and NEGF simulations for the peak and saturation \( \sigma_{\text{min}} \) is reached with details in Supporting Information Supplement VII.

Finally, we discuss the implications of this work for nanoscale graphene devices. It is a major hurdle to realize high-performance graphene transistors due to the absence of a bandgap. Our study provides valuable insights in this context. Figure 4a shows the width scaling of the on- and off-state conductance \((G_{\text{on}} \text{ and } G_{\text{off}})\). Interestingly, while \( G_{\text{on}} \propto W \), \( G_{\text{off}} \) follows an unconventional trend \( G_{\text{off}} \propto W^{2.1} \). We attribute this stronger suppression in \( G_{\text{off}} \) as \( W \) decreases to the following mechanisms: (i) the number of rough edges per unit width is increased, thereby suppressing the highly conductive edge states; (ii) some percolation paths are terminated. The particular \( G_{\text{off}} \propto W \) dependence can be rationalized knowing \( \sigma_{\text{min}} \approx W^{0.16-0.46} \) for \( W/L < 0.5 \) and \( G_{\text{eff}} = \sigma_{\text{min}} (W/L) \approx W^{1.16-1.46} \). Width reduction without compromising the on-current can be accomplished by cutting nanometer-scale incisions along the transport direction (Figure 4b). We have observed at least a two- to three-fold improvement in on/off ratio by moderately reducing the effective width from \( 1 \mu \text{m} \) to \( 100 \text{ nm} \), as demonstrated in Figure 4a. This also allows us to scale the channel width without modifying the on/off ratio to facilitate circuit level design requirements. In a similar fashion, the recently demonstrated graphene nanomesh works under the same principle and was found to achieve an on/off ratio of \( \sim 160 \).\(^28\)

Exploiting these geometries will allow designing better...
graphene-based nanoelectronics such as RF devices and biosensors.\textsuperscript{29,30}

Note: Our study should have relevance to the recently demonstrated atomically flat graphene on boron nitride system.\textsuperscript{31} In fact, we expect a stronger nonmonotonic dependence of $\sigma_{\text{mob}}$ as a function of $W$ due to suppression of bulk disorder in such a system.

**Methods.** Graphene flakes are mechanically exfoliated onto 90 nm SiO$_2$/Si substrates from highly oriented pyrolytic graphite (HOPG, NT-MDT). Source and drain contacts are patterned by e-beam lithography, followed by e-beam evaporation and lift-off of a stack of Ti/Pd/Au (10 nm/30 nm/20 nm) metals. To prevent damages to the graphene ribbons from the e-beam patterning process, we choose graphene flakes of various widths and lengths as naturally cleaved from the exfoliation process. In this way, there are no further modifications on the graphene ribbons widths.

Devices with $W = 0.1$–4.9 $\mu$m and physical lengths ($L_{\text{SEM}}$) of 1.0–1.5 $\mu$m are created from single-layer graphene flakes. The thickness of the graphene flakes is determined by optical analysis and atomic force microscopy (AFM). Electrical measurements of the devices are carried out at $T = 4$–300 K under vacuum ($1 \times 10^{-7}$ Torr). The field-effect mobility of typical devices is $\sim 4000$ cm$^2$/V·s as determined from room temperature transfer characteristics.

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