



Electronic transport in graphene nanostructures on SiO₂

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ABSTRACT

We report two experiments on graphene nanostructures. The first was performed on a graphene nanoribbon, where the nature of electronic transport was investigated in detail. Electrons or holes are found to localize in pockets of the potential along the ribbon. Transport is governed by the joint action of localization and Coulomb interaction. The temperature-dependence of the conductance shows activated behavior at temperatures above a few Kelvin. The activation energy retraces the edges of Coulomb blockade diamonds found in nonlinear transport. In the second experiment the metallic tip of a low-temperature scanning force microscope was scanned above a graphene quantum dot. In addition to the familiar Coulomb blockade fringes, localized states are detected forming in the constrictions connecting the dot to source and drain.

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1. Introduction

Experiments on semiconductor nanostructures have allowed researchers to investigate tailored quantum mechanical systems. In the past, a number of novel phenomena ranging from the quantum Hall effects [1,2] to conductance quantization [3,4], the Aharonov–Bohm effect [5–8] and the Coulomb blockade [9,10] were found. Progress since these early days has been enormous, such that today the quantum states of individual electrons and their spins can be coherently controlled [11,12]. The availability of graphene as a new material for nanostructure research has led to a new and fascinating playground for investigating quantum phenomena in a two-dimensional solid consisting of only one single atomic layer. The unusual band structure of this material and its resulting exceptional electronic properties have sparked hope for new physics and applications. In this paper we review a small selection of research on graphene nanostructures recently performed in our group.

2. Fabrication of nanostructures on SiO₂ substrates

We fabricate graphene nanostructures starting from flakes of natural graphite crystals. Atomic monolayer sheets are peeled from the crystal by mechanical exfoliation using the ‘sticky-tape method’ [13]. The tape with the graphene sheets is subsequently pressed on the substrate surface by which some of the material is

deposited there. The substrate is a highly doped silicon wafer with a 290 nm thick SiO₂ layer covering the surface. This thickness of the oxide leads to a unique contrast of single-layer flakes [14] and therefore allows us to preselect them with an optical microscope. The highly doped silicon remains conducting even at the temperature of liquid helium and below. It can therefore be used as a back gate of the graphene flake in low-temperature experiments that allow us to exploit the field effect for changing the carrier density in the graphene sheet. The specific fingerprint in Raman spectroscopy [15,16] is used to confirm that the preselected flakes are indeed single-layer. Imaging the flake with a scanning force microscope gives information about the cleanliness of the graphene sheet.

Nanostructures are then defined in polymethylmethacrylate (PMMA) resist on the single-layer flakes using electron beam (e-beam) lithography. Very fine structures down to 20 nm can be achieved using single-pixel lines in a resist of 45 nm thickness. The pattern is transferred into the graphene sheet by reactive ion etching (RIE) for 10 s with argon and oxygen. The power used to ignite the plasma in the RIE chamber is kept at a minimum. This avoids additional cross-links of the PMMA which make the resist removal more difficult. The parts of the graphene sheet protected by the resist are not damaged during etching [17,18]. Ohmic contacts are made by defining the contact pattern in resist using another e-beam lithography step, evaporating 2 nm of chromium and 40 nm of gold, and lifting off the excess metal.

We usually obtain devices with more tuning flexibility by integrating graphene in-plane gates with the structures, as shown in Fig. 1. In-plane gates have the advantage over the back gate that they allow us to tune the devices locally [17]. With this

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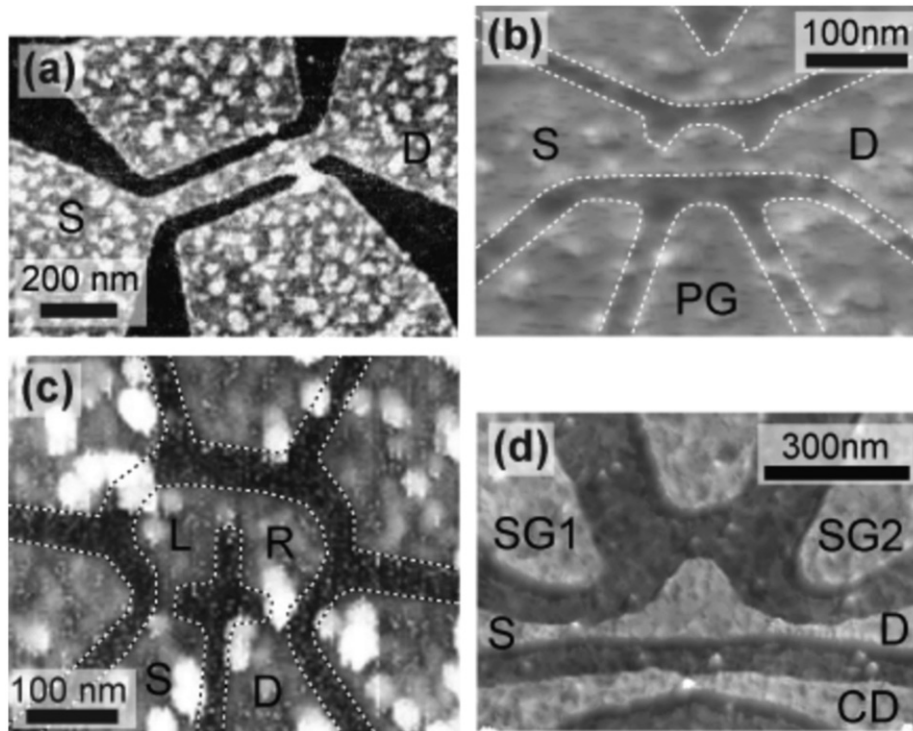


Fig. 1. Various nanostructures made from graphene imaged with a scanning force microscope. (a) A nanoribbon (length 500 nm, width 85 nm) connected via source (S) and drain (D) contacts. Two in-plane gates give tunability of the ribbon in addition to the back gate. (b) A single-electron transistor (SET) device with source (S) and drain (D) contacts and a plunger gate. Additional in-plane gates allow us to fine-tune the constrictions and the SET. (c) A double quantum dot device (left dot L and right dot R) connected to source (S) and drain (D) reservoirs and combined with a number of in-plane gates. (d) An SET connected to source (S) and drain (D) electrodes. In-plane gates used for tuning the constrictions are labeled SG1 and SG2. An additional nanoribbon (CD) is coupled capacitively to the quantum dot.

fabrication technique, a range of different device types has been made in the past. In Fig. 1(a) we show an example of a graphene nanoribbon [19,20]. Such devices are of interest for studying the transport characteristics of graphene with strong influence of graphene edges. Furthermore their properties have to be known, because they are building blocks of quantum dot structures [see Fig. 1(b–d)] and can be used as on-chip charge detectors [Fig. 1(d)]. Quantum dot structures [21] give access to single-electron electronics including opportunities for the investigation and manipulation of single spins. Steps in the direction of the latter are the double quantum dot in Fig. 1(c) [22] and the integrated charge read-out in Fig. 1(d) [23].

3. Constrictions

Interest in the detailed transport characteristic of graphene constrictions has led to the publication of a remarkable number of publications in the past. The interest was triggered by a number of theoretical predictions made before single-layer graphene was experimentally available. It was suggested that the dispersion relation and the density of states in perfectly clean and long ribbons depend severely on the width of the ribbon and on the orientation of its edges [24–26]. Similar to carbon nanotubes, where the properties depend on the diameter and the chirality [27], ribbons with pure zig-zag edges should have a gapless dispersion and form edge states at zero magnetic field. If edges are pure arm-chair the dispersion is gapless if the number n of dimer rows within the width of the ribbon fulfills $n=3m-1$, otherwise a confinement band gap is expected to form. If both, arm-chair and zig-zag edges, are present and alternate randomly, localized states are predicted to form along the ribbon edges [24,26]. These results for the dispersion relations of clean ribbons

also lead to the prediction of peculiar conductance quantization in graphene nanoribbons [28].

Experimentally, the field was triggered by two experiments in 2007. The experiments by Kim and coworkers [29] showed suppressed conductance of ribbons compared to large area graphene flakes close to the Dirac point. The strength of the suppression was found to increase with decreasing ribbon width. Based on these results, the presence of an energy band gap in ribbons was claimed. At the same time Avouris and coworkers [30] published similar results and suggested a quantum confinement gap opening in narrow ribbons. However, both groups were aware of the possible influence of the localized states at the ribbon edges that would form given the fabrication technique used. Indeed, theoretical work performed at the same time suggested that localized edge state could transform graphene nanoribbons into Anderson insulators [31].

The question of the nature of the insulating behavior in narrow ribbons remained open and attracted more groups to perform experiments. While a later controversial publication by Avouris and coworkers [32] continued to spread the idea of lateral quantum confinement and even claimed indications of conductance quantization, detailed studies by other groups revealed novel aspects of the problem. It was found that even short constrictions show pronounced transmission resonances that can modulate the conductance over orders of magnitude [33]. In addition, the observation of Coulomb blockade diamonds in ribbons with a length of more than 100 nm [19,20,34] made clear that interaction effects do indeed play an important role for transport in ribbons, as anticipated by theoretical work [35]. Many more experiments by different groups [36–39] supported the notion that graphene nanoribbons are no band insulators, and that transport is governed by localization caused by disorder and electron–electron interactions.

Studies of the temperature dependence of the conductance were reported in Refs. [40,41]. These experiments confirmed the importance of the Coulomb interaction for transport, and found a temperature dependence consistent with Shklovskii–Efros variable-range hopping conduction [42]. However, it is not clear if this theory can be applied to ribbons of finite length, because it requires averaging over a macroscopic number of localized sites. We will show details here about recent measurements on the Coulomb gap in graphene nanoribbons [43] that avoids this interpretation, and aims at an understanding of the temperature dependent conductance of nanoribbons based on a microscopic picture instead.

The inset of Fig. 2 shows a nanoribbon, 60 nm in width and 200 nm in length. The dependence of its conductance on the back-gate voltage is shown in the main panel of Fig. 2 on a logarithmic scale. The conductance changes by about three orders of magnitude while the Fermi-energy is shifted from the valence band (very negative V_{BG}) to the conduction band (very positive V_{BG}). There is a range around zero back gate voltage, where the conductance is suppressed well below the conductance quantum e^2/h . Such a strong suppression is indicative of strong localization in the ribbon. This gate-voltage range, gray-shaded in the figure, is called the transport gap. It has been found in other experiments that the transport gap depends only weakly on the ribbon length [38], but is inversely proportional to the ribbon width [29,37]. The conductance is seen to fluctuate strongly within the transport gap, an effect which is characteristic and reproducible. It reminds us of conductance fluctuations, for example those that have been observed in disordered mesoscopic silicon wires [44], with the important difference that silicon has a large energy band gap such that transport cannot be tuned easily from the valence to the conduction band.

The lower panel in Fig. 3 shows a color plot of the differential conductance of the same ribbon measured in the transport gap as a function of back gate voltage and source–drain voltage. Very pronounced diamonds of suppressed conductance indicate the importance of Coulomb blockade. Charging energies vary between only a few and more than 10 meV, with the trend to be largest in the center of the transport gap. In other experiments it has been found that this Coulomb gap (usually taken to be the largest observed charging energy in the transport gap) increases with increasing ribbon length [38] and decreasing ribbon width [37]. In some regions of gate voltage, the diamond pattern looks regular like that of a single quantum dot. However, in other regions, neighboring diamonds overlap indicating that several dots limit the current flow. The value of the charging energy allows us to estimate the size of the localized puddles in the ribbon to be comparable to the width of the ribbon. The microscopic view of the ribbon is therefore a quasi-one-dimensional

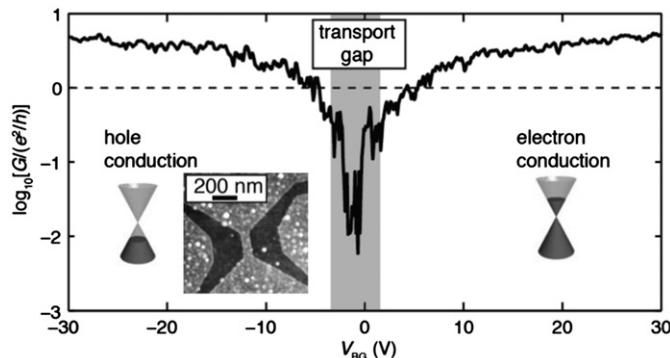


Fig. 2. Conductance of the nanoribbon depicted in the inset (width 60 nm, length 200 nm) as a function of the back gate voltage. The transport gap is indicated as a gate-voltage interval shaded in gray.

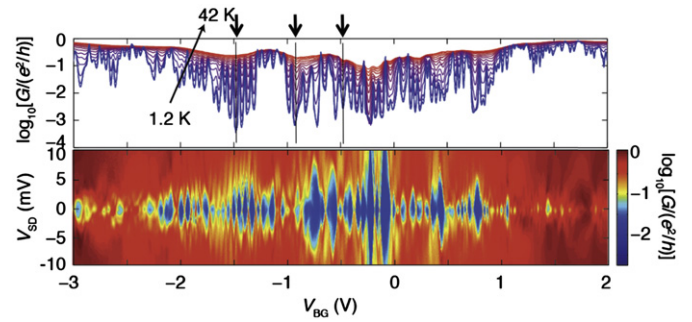


Fig. 3. (Color online) Bottom panel: color plot of the differential conductance of the nanoribbon, measured as a function of back gate voltage and source–drain bias voltage. Top panel: temperature dependence of the linear response conductance measured as a function of back gate voltage between 1.2 K and 42 K.

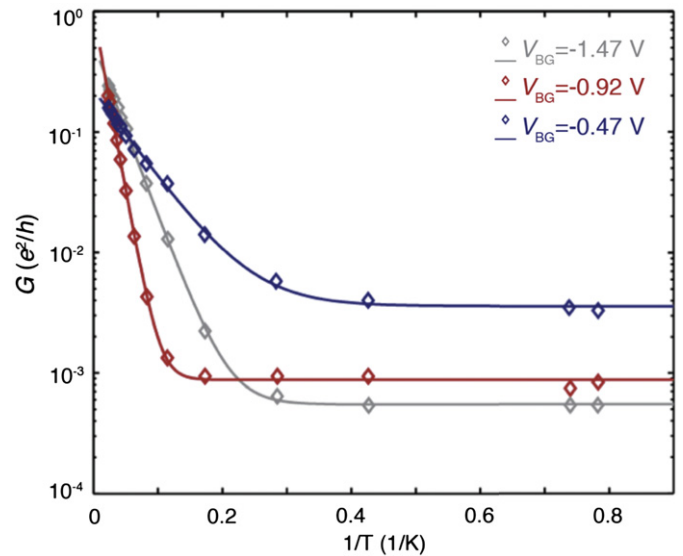


Fig. 4. (Color online) Temperature-dependence of the conductance in valleys between conductance resonances at three selected gate voltages.

chain of randomly placed pockets containing strongly localized electronic states. This situation is reminiscent of stochastic Coulomb blockade [45,46].

The nature of electronic transport in this system can be inferred from the temperature dependence of the conductance in the linear response regime, where the source–drain voltage $|eV_{SD}|$ is smaller than the temperature $k_B T$. The result of such a measurement is shown in the top panel of Fig. 3. One can see that increasing temperature increases the conductance in the valleys between the resonances observed at the lowest temperature. Even most of the resonances themselves increase their peak value, when the temperature is raised. Only occasionally, a conductance peak decreases its height with increasing temperature. The temperature-dependence of the resonances may be a signature of multilevel transport in the relevant quantum dots [47].

A more detailed temperature dependence in the valleys between resonances is shown in Fig. 4. We observe activated behavior at high temperatures, which levels off to an approximately constant value at the lowest temperatures. A detailed analysis of the data in the full gate voltage range [43] reveals that the activation energy that can be extracted as a function of back gate voltage retraces the edges of the Coulomb blockade diamonds seen in the bottom panel of Fig. 3. The thermal activation allows carriers to overcome the Coulomb gap created by the

localized electron pockets dominating transport. At the lowest temperatures, the thermal energy is insufficient to excite carriers above the Coulomb gap. The only way they get past the weak links in the ribbon is by cotunneling, which has a very weak temperature dependence. This leads to the saturation of the activated behavior seen at the lowest temperatures in Fig. 4.

An interesting question remaining after this study is about the causes of the disorder potential in the ribbons. In particular, it would be interesting to differentiate between effects of edge disorder and bulk or substrate disorder. The measurement shown in Fig. 5 shows the behavior of the conductance traces as a function of back gate, before and after a thermal cycle to room temperature. At room temperature the sample was not exposed to air, but kept in vacuum. It is seen that the details of the conductance resonances (position, amplitude, distribution) are very different in the two cool downs. It is probably safe to assume that the geometric details of the ribbon including the shape and disorder of the edges remain unchanged by such a thermal cycle. In contrast, it is conceivable that the exact way how impurities in the SiO₂ substrate freeze out during cooldown is different, and the position of adsorbed impurities on the surface (or at the edges) of the graphene ribbon may also have changed during the time the sample was kept at room temperature. Also the warping of the flake on top of the substrate may have changed. While this reasoning does not allow us to make a conclusive statement about the importance of edge disorder versus bulk disorder, it seems likely that rigid edge disorder alone (in particular the disordered edge structure formed by chemical bonds) cannot be the cause of the observed changes between cooldowns.

We do not finish the discussion of graphene nanoribbons without mentioning very interesting recent measurements by van Wees and coworkers which indicate the observation of quantized conductance in suspended graphene nanoribbons [48]. These observations were made on a high-mobility suspended graphene device after annealing the ribbon by applying

high current. Even in this ribbon the microscopic structure of the ribbon edges is unknown. However, removing the ribbon from the substrate has obviously improved the electronic properties of the device to such an extent that localization and strong backscattering could be avoided completely.

4. Quantum dots

Coulomb blockade in graphene nanostructures can not only be measured in disordered nanoribbons, but also in geometrically well-defined and electrostatically well-controlled quantum dot devices [21,49]. After the first experiments, rapid progress has been made in reducing the size of devices, in observing excited states by dI/dV_{SD} -spectroscopy [50] and the crossover between electron- and hole transport [51], and even in identifying Zeeman-split spin states in quantum dots [52]. The main driving force behind these investigations is the proposal that graphene could be advantageous for realizing spin-qubits compared to other materials, such as Ga[Al]As. While in the latter hyperfine-coupling of the electron spins in a quantum dot to the nuclear spins in the underlying host lattice seriously limits spin-coherence, the situation looks more promising in graphene-based quantum dots, because 98.9% of the carbon atoms in natural graphite are ¹²C with zero nuclear spin, and only 1.1% of the atoms are ¹³C with a nuclear spin of 1/2. In addition, also spin-orbit interaction effects are expected to be small in graphene owing to the small nuclear charge of the carbon atoms. These advantages led to the proposal to use graphene quantum dots for implementing spin-qubits [53].

Here we briefly focus on an experiment that was able to map the conductance properties of a graphene quantum dot with spatial resolution [54]. The inset at the top right of Fig. 6 shows the investigated quantum dot sample. The quantum dot in the center is connected via narrow constrictions with the source (S) and drain (D) contact. Voltages applied to the two in-plane gates B1 and B2 selectively tune the transparency of the constrictions. A voltage applied to the plunger gate (PG) mainly changes the number of electrons residing on the quantum dot. The long ribbon labeled CD in the figure is not used in the present experiment. The conductance of this structure is investigated with a scanning gate, a sharp metallic tip that can be scanned at a distance of only a few 10 nm above the surface of the structure. The voltage applied to this scanning gate induces a local potential in the quantum dot device and thereby changes its conductance. Raster-scanning this gate above the surface results in a spatially resolved conductance image.

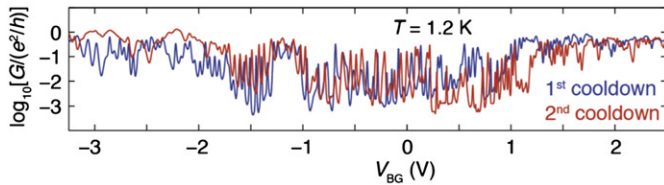


Fig. 5. (Color online) Back gate voltage dependence of the ribbon conductance in different cooldowns. Between them, the sample was not exposed to air, but only cycled from 1.2 K to room temperature and back.

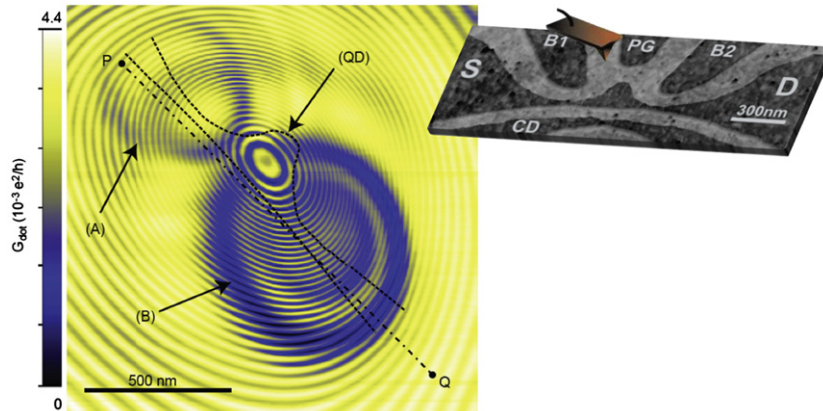


Fig. 6. (Color online). Scanning gate image of the graphene quantum dot structure is discussed in the text in detail. Inset: Real space image of the investigated structure. The scanning tip is schematically indicated.

Such an image measured at a temperature of 2.6 K is shown in Fig. 6. The scanned area is $1.4 \times 1.4 \mu\text{m}^2$. The position and orientation of the quantum dot device is indicated in the figure with black dashed lines marking the edge of the relevant structure. The quantum dot is labeled QD. The color-coded conductance image gives clear information about the location of the quantum dot in the image. The center of the dot is the origin of a large number of concentric fringes of alternating enhanced and suppressed conductance. The suppression between some fringes is strong enough to result in essentially zero current. This is the signature of Coulomb blockade in the quantum dot. When the metallic tip approaches the dot center from any direction, the number of electrons in the dot is reduced one-by-one, because the repulsive tip-induced potential has a stronger influence with decreasing distance. The quality of this image is comparable with the best previous results on quantum dots [55–60].

The interesting observation seen in Fig. 6 is the modulation of the Coulomb-blockade resonance amplitudes labeled (A) and (B). Each of the two modulations has the shape of concentric fringes with a center sitting in one of the two constrictions. The origin of these superimposed fringes is transmission resonances of localized electronic states in the constrictions. Such constriction resonances had been observed previously in pure transport experiments [21,33], but the direct observation of their location in real space has remained elusive so far in standard transport experiments employing fixed local in-plane gates. These resonances are reminiscent of the localized electron pockets observed in the nanoribbons discussed above in this paper. Further measurements and a detailed analysis allowed us to estimate the spatial extent of the localized states in the constriction to be 20–30 nm, comparable to the smallest width of the constrictions [54].

5. Conclusions

In this paper we have reviewed transport in graphene nanoribbons, and we have demonstrated an imaging technique that allowed us to locate localized states forming in the narrow constrictions of a quantum dot device. These studies are exemplary for the type of physics encountered in graphene nanostructures. It is highly desirable to reduce the effects of disorder in the material in future. Promising directions that many research groups follow at the moment are suspended devices and graphene devices on boron nitride. Large area graphene sheets fabricated with these techniques have shown much higher mobilities than devices on silicon substrates. In addition, bilayer graphene attracts a lot of attention. The option to open a band gap by using top and bottom gates is promising for fabricating nanostructures with smooth electrostatic confinement potentials. All these developments show that graphene nanostructure research is still at an early stage with excellent prospects for new achievements in physics and electronics.

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