Transport Properties and Local Imaging
of Graphene Quantum Dots

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Abstract

In this thesis, we present transport and scanning-gate microscopy experiments on graphene nanostructures at cryogenic temperatures. Moreover, an analytic expression for the quantum states of a circular graphene quantum dot in a perpendicular magnetic field is derived.

Graphene is a semi-metal showing the electric field effect and thus is an interesting material for the mesoscopic semiconductor community. In graphene quantum dots, we observed clear indications of transport through confinement-induced excited states. These showed up in resonant sequential tunneling and in inelastic co-tunneling in the Coulomb-blockaded regime using tunneling spectroscopy. To deepen our understanding of the confining potential, we looked at the magnetic-field dependence and identified a transition from confinement-induced to magnetic-field induced behavior.

Motivated by these experimental results, we could derive an analytic mathematical expression which qualitatively describes both the confinement-induced and the magnetic-field induced effects as well as the transition between both. As a side-effect, a strategy for determining experimentally the electron-hole crossover in graphene quantum dots was developed.

A second graphene quantum dot was studied with our dilution refrigerator scanning-gate microscope. We could clearly resolve Coulomb resonances of the quantum dot. More than 35 Coulomb resonances were recorded in some measurements where a spacing of around 20 nm of adjacent Coulomb resonances could clearly be resolved. This proved the exceptional quality of both the sample and the scanning sensor. Our measurements also gave direct proof of the existence of localized states in graphene nanostructures: We could image and locate a single localized puddle in each constriction connecting the quantum dot to source and drain. Furthermore, our technique enabled us to deduce a radius of around 10 to 13 nm for such a localized state. Despite the good spatial resolution, we could not resolve quantum confinement effects in the quantum dot or in one of the localized puddles.

Finally, we analyzed experimentally how a change in the electrostatic environment can influence the outcome of a scanning-gate measurement. We identified
the possible underlying processes and supported our arguments with numerical simulations.

As a basis for future scanning-tunneling experiments on graphene, a description of the feedback loop in linear response including bandwidth and response function was established. A comparison with the measured response showed good agreement.
Zusammenfassung

In der vorliegenden Arbeit präsentieren wir Transportmessungen und Scanning-Gate-Experimente an Graphen nanostrukturen und bei Temperaturen unter 4.2 K. Zudem wird ein analytischer Ausdruck für die Quantenzustände eines runden Graphenquantenpunkts in einem senkrechten Magnetfeld hergeleitet.


Abschliessend analysierten wir experimentell, wie eine Änderung in der elek-
Zusammenfassung

Die Antwortfunktion des Regelkreises für den Betrieb des Aufbaus im Rastertunnelmodus für potentielle Rastertunnelexperimente auf Graphen wurde in linearer Antworttheorie hergeleitet. Der Vergleich mit der gemessenen Antwortfunktion zeigte eine gute Übereinstimmung.
List of Acronyms, Constants, and Variables

Acronyms

<table>
<thead>
<tr>
<th>acronym</th>
<th>explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>2DEG</td>
<td>two-dimensional electron gas</td>
</tr>
<tr>
<td>SET</td>
<td>single-electron transistor</td>
</tr>
<tr>
<td>AFM</td>
<td>atomic-force microscope/microscopy</td>
</tr>
<tr>
<td>STM</td>
<td>scanning-tunneling microscope/microscopy</td>
</tr>
<tr>
<td>SGM</td>
<td>scanning-gate microscope/microscopy</td>
</tr>
<tr>
<td>CNP</td>
<td>charge-neutrality point</td>
</tr>
<tr>
<td>PIC</td>
<td>proportional-integral controller</td>
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</table>

Constants

<table>
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<tr>
<th>constant</th>
<th>value</th>
<th>explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$e =</td>
<td>e</td>
<td>$</td>
</tr>
<tr>
<td>$m_e$</td>
<td>$9.1094 \times 10^{-31} \text{kg}$</td>
<td>(free) electron mass</td>
</tr>
<tr>
<td>$k_B$</td>
<td>$1.3807 \times 10^{-23} \text{J/K}$</td>
<td>Boltzmann’s constant</td>
</tr>
<tr>
<td>$\hbar = h/2\pi$</td>
<td>$1.0546 \times 10^{-34} \text{Js}$</td>
<td>Planck’s constant</td>
</tr>
<tr>
<td>$\varepsilon_{\text{vac}}$</td>
<td>1</td>
<td>relative dielectric constant of vacuum</td>
</tr>
<tr>
<td>$\varepsilon_{\text{SiO}_2}$</td>
<td>3.9</td>
<td>relative dielectric constant of $\text{SiO}_2$</td>
</tr>
<tr>
<td>$c_*$</td>
<td>$\sim 10^6 \text{m/s}$</td>
<td>effective velocity of charge carriers in graphene</td>
</tr>
<tr>
<td>$\alpha_* = e^2/(4\pi\varepsilon_0\hbar c_*)$</td>
<td>$\sim 1$</td>
<td>effective fine-structure constant for graphene</td>
</tr>
</tbody>
</table>
# List of Acronyms, Constants, and Variables

## Variables

<table>
<thead>
<tr>
<th>variable</th>
<th>explanation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_{bias}$</td>
<td>bias voltage across a device, e. g. a quantum dot</td>
</tr>
<tr>
<td>$\mu_N$</td>
<td>electrochemical potential of a quantum dot with $N$ charge carriers</td>
</tr>
<tr>
<td>$\Delta E_C$</td>
<td>charging energy of a quantum dot</td>
</tr>
<tr>
<td>$C_\Sigma$</td>
<td>total capacitance of a quantum dot</td>
</tr>
<tr>
<td>$\alpha_i$</td>
<td>lever arm of gate $i$ acting on a quantum dot</td>
</tr>
<tr>
<td>$T$</td>
<td>temperature</td>
</tr>
<tr>
<td>$E_F$</td>
<td>Fermi energy</td>
</tr>
<tr>
<td>$\mathcal{D}(E) = 2E/\left(\pi\hbar^2c_\Sigma^2\right)$</td>
<td>density of states of graphene</td>
</tr>
<tr>
<td>$N$</td>
<td>number of charge carriers trapped on a quantum dot</td>
</tr>
<tr>
<td>$B$</td>
<td>magnetic field</td>
</tr>
<tr>
<td>$\Delta$</td>
<td>single-particle level spacing</td>
</tr>
<tr>
<td>$A$</td>
<td>area of a quantum dot</td>
</tr>
<tr>
<td>$I_{tc}$</td>
<td>tunneling current in STM-mode</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

“Graphene” denotes the two-dimensional arrangement of carbon atoms on a hexagonal lattice. It has been known for a long time that graphite consists of stacked and weakly-bound graphene layers. In his seminal paper of 1947, Wallace derived the dispersion relation for graphene in order to deduce properties of bulk graphite \cite{1}. The dispersion displayed close analogies to massless, relativistic particles. However, since the fabrication of graphene remained impossible, this material system served as a theoretical playground for a condensed-matter system mimicking relativistic quasi-particles \cite{2}.

The situation changed drastically when A. Geim, K. Novoselov, and co-workers succeeded in the fabrication of single-layer graphene in 2004 using a surprisingly simple approach coined the “scotch-tape technique” \cite{3}. Their experiment showed the existence of the electric-field effect for graphene and thus made it interesting for the semiconductor community. Since then, publications on graphene – both theoretical and experimental in nature – exploded; it is fair to say that graphene is one of the hottest topics in current physics research. Intriguing properties and experimental results like the highest room-temperature mobility of all known materials, the unconventional quantum Hall effect \cite{4, 5}, and Klein tunneling \cite{6, 7} – just to name few – make it a popular object of research in both applied and fundamental sciences.

The theory and the experiments described in this thesis apply to graphene nanostructures. We are still far away from completely understanding the electronic properties of graphene. This thesis will try to contribute one jigsaw piece to the big graphene puzzle. The employed experimental techniques are tunneling spectroscopy for conventional transport experiments and scanning-gate microscopy as a localprobe technique. The composition of the thesis is the following:

- Chapter 1 serves as an introduction into the basic electronic properties and

\footnote{Fabrication details will be given below.}
Chapter 1. Introduction

the fabrication of graphene. It motivates transport and scanning-probe experiments on graphene. It finishes with a review on scanning-probe experiments performed so far on graphene.

• Chapter 2 presents the theoretical background of quantum-dot physics as far as it is necessary for this thesis. Derivations will be given in case they cannot be found in modern textbooks. This includes in particular the derivation of the energy spectrum of a graphene quantum dot in a magnetic field.

• Chapter 3 is of technical nature because it shortly summarizes the experimental setup and explains the scanning-gate principle. Additionally, it describes how the setup can be used in scanning-tunneling mode and how feedback parameters have to be optimized accordingly.

• Chapter 4 describes tunneling spectroscopy on a graphene quantum dot where excited states and co-tunneling were observed. The transition from confinement-dominated to magnetic-field dominated behavior is also investigated.

• Chapter 5 presents a scanning-gate experiment carried out on a graphene quantum dot where Coulomb resonances of the dot and localized states in the constrictions were imaged in real space.

• Chapter 6 delves into the complications arising when the electrostatic environment in the sample due to e. g. gate sweeps changes. This can lead to an apparent shift of the quantum-dot location. Spurious additional Coulomb rings are created by gate electrodes due to screening effects.

• Chapter 7 gives a summary of this thesis and an outlook.

In the course of the experimental work, severe problems with the KELVINOX 100 dilution refrigerator used for the experiments occurred. The insert of the cryostat was sent back to the manufacturer for repair. After re-assembling the cryostat, system parameters were determined. These together with the causal chain leading to the problems are discussed in appendix D.

In this introductory chapter, we will first give an overview over the basic electronic properties of graphene and shortly describe how pristine graphene is obtained and graphene nanostructures are fabricated. The second section will argue why conventional transport experiments and in particular scanning-probe techniques are of special interest. It also includes a review of scanning-probe experiments already carried out on graphene.
1.1 Graphene – Basic Electronic Properties and Fabrication

1.1.1 Electronic Properties

Graphene is a monolayer of carbon atoms, arranged in a hexagonal lattice as shown in Fig. 1.1(a). This leads to the identification of a primitive cell made up of two atoms A and B. Each carbon atom is bound to its three neighbours via $\sigma$-bonds creating an $sp^2$-hybridization. The fourth valence electron forms a $p_z$-orbital which sticks out of the graphene plane. It is this valence electron which is important for electronic transport since it covalently binds to neighboring $p_z$-orbitals forming a $\pi$-band. A tight-binding calculation including only nearest-neighbor hopping yields the dispersion relation of graphene [1]

$$E(q) = \pm \gamma \sqrt{1 + 4 \cos^2 (q_x a/2) + 4 \cos (q_x a/2) \cos (\sqrt{3} q_y a/2)}$$  (1.1)

with the nearest-neighbor hopping energy $\gamma \approx 2.8$ eV and the lattice constant $a = 2.46$ Å. Conduction and valence band are denoted by the plus and the minus sign. The corresponding energy bands are shown in Fig. 1.1(b). In the ground state, the two valence electrons of the primitive cell fill the valence band whereas the conduction band is completely empty so that the Fermi energy is at $E_F = 0$ eV.

The $E = 0$ plane is crossed at six points, the inequivalent $K$- and $K'$-points of the first Brillouin zone. If we expand the above dispersion around these points by e. g. writing $q = K + k$, where $K$ is the position of the $K$-point and $|k| \ll |K|$, we end up with a linear dispersion relation shown in Fig. 1.1(c), namely

$$E(k) = \pm \hbar c_s |k|$$  (1.2)

with the effective velocity of charge carriers in graphene $c_s = \sqrt{3} \gamma a / 2 \hbar \approx 10^6$ m/s. The corresponding Hamiltonian is given by $H_K = \hbar c_s k \cdot \sigma$ and similar for the $K'$-point with the Pauli matrices $\sigma = (\sigma_x, \sigma_y)$. This Hamiltonian is closely related to Dirac’s equation for massless particles: Electrons and holes are in analogy to electrons and positrons and valleys $K$ and $K'$ take the role of the spin of Dirac’s equation. If we also consider the spin of charge carriers in graphene, the full wave function will consist of eight components: spin up and down for electrons and holes living in the $K$- or $K'$-valley [3]. Employing the linear dispersion relation at the $K$- and $K'$-points and the four-fold degeneracy due to spin and valley, it is straightforward to derive the density of states for graphene, $D(E) = 2E / (\pi \hbar^2 c_s^2)$. Contrary to conventional two-dimensional electron gases with a parabolic dispersion relation, it does depend on energy.
Intrinsic graphene without any doping is not a typical metal because the Fermi energy does not lie in the conduction band. On the other hand, it is also not a semiconductor because of the missing band gap. However, when graphene is deposited on an insulating substrate like SiO$_2$ followed by highly doped silicon, this conductive layer can be used as a back gate. Graphene then shows an electric-field effect \cite{3}. In fact, it is possible to tune the charge-carrier type from valence-band holes through the so-called charge-neutrality point (CNP) or Dirac point with nominally zero-carrier density to conduction-band electrons according to $n = \alpha (V_{BG} - V_{CNP})$. Here $n$ is the induced charge-carrier density, $V_{CNP}$ is the position of the CNP, $V_{BG}$ is the applied back gate voltage, and $\alpha \approx 7.3 \times 10^{10} \text{cm}^{-2}\text{V}^{-2}$ is a proportionality constant. Surprisingly, there is a finite maximal resistivity of around $\rho_{\text{max}} \sim \hbar/4e^2$ for $V_{BG} = V_{CNP}$ \cite{4}. The origin of this behavior is still not completely clear but it is known that electrons and holes form so-called puddles co-existing at the CNP \cite{9}. Then, charge transport can take place by tunneling of

\footnote{Because of unintentional doping by adsorbates on the graphene sheet, the CNP can be shifted by several tens of volts off zero.}
1.1. Graphene – Basic Electronic Properties and Fabrication

carriers through the puddles and a complete suppression of current is not possible.

If graphene is used for creating more complex nanostructures like quantum dots, the lack of a band gap is a challenge when confining carriers. Carriers with normal incidence on an electrostatic barrier will cross the barrier with perfect transmission due to the relativistic character and Berry’s phase of $\pi$ of the carriers \cite{6, 10, 11}. In general, back scattering is strongly suppressed in graphene because most scatterers do not lead to $K$-$K'$-scattering. A transport gap in graphene is obtained by cutting the graphene flake into small stripes, so-called nanoribbons or constrictions (for short nanoribbons). Transport can then be strongly suppressed; however, the physical origin of the (transport) gap is still debated \cite{12, 19}. Similarly, quantum dots are formed by cutting the graphene flake into the desired shape. It is not yet possible to control the edge of a graphene structure on an atomic scale and it is not clear what happens at the edge in an experiment. Theoretically, the formation of edge channels for specific edge structures is predicted \cite{20}. Therefore, gaining a better understanding of the role of the edges is an important task.

There are many more aspects related to the electronic properties of graphene. Those mentioned above shall serve as an introduction to the fascinating transport physics of graphene as well as necessary background information for the next chapter. We refer to Ref. \cite{8} for an extensive review on the electronic properties of graphene.

1.1.2 Fabrication

The way of producing graphene used for the samples described in this thesis is called mechanical exfoliation: Graphite powder is spread over adhesive tape\textsuperscript{3} which is then folded and pulled apart several times. In such a way, graphite flakes are mechanically cleaved. Multilayer, few-layer, and eventually bilayer and single-layer graphene flakes will stick to the tape now. In a next step, the flakes have to be transferred onto a chip. The chip consists of highly doped silicon covered with 300 nm of insulating SiO\textsubscript{2}. The transfer is achieved by simply pressing the chip onto the tape with the flakes. Due to Van-der-Waals forces, some flakes will detach from the adhesive tape and stick to the chip.

On the chip, there will be both multilayer- and single-layer graphene flakes. A preliminary identification is obtained using optical microscopy under illumination with white or green light. Since the optical transmission coefficient of few-layer graphene changes depending on the number of layers, potential single-layers can be determined with high probability by their contrast. Their position on the chip is noted. An unambiguous identification of a single-layer flake is finally obtained by either measuring the step height of the flake with an atomic-force microscope

\textsuperscript{3}Therefore, this technique is often called the scotch-tape technique.
(below 1 nm) or – even more reliably – by Raman spectroscopy [21, 22].

Single-layer flakes have typical extensions from few µm’s up to some tens of µm’s for big flakes. After their identification, electron-beam lithography in combination with 90 nm of polymethyl methacrylate (PMMA) is used for patterning an etch mask of the desired shape followed by Ar/O\(_2\) reactive ion etching. In particular, self-aligned graphene in-plane gates can be patterned in the same etching step as a quantum dot. An atomic-force micrograph of a quantum-dot structure with three in-plane gates and a nearby graphene ribbon after lift-off is shown in Fig. 2.1(b). A second electron-beam and lift-off step is performed to pattern the metallic electrodes (Ti/Au with 2 nm/50 nm), contacting the gates, source, and drain.

### 1.2 Motivation for Research on Graphene Nano-structures

#### 1.2.1 Transport Experiments

As pointed out above, the extraordinary electronic properties of graphene make it a promising candidate for future applications but also for fundamental research. In particular, research into the direction of quantum information processing employing graphene could be very promising. In their seminal paper of 1998, D. Loss and D. P. DiVincenzo described how quantum dots can be used as spin qubits for quantum computation [23]. They suggested to use electric and magnetic fields for spin manipulation; transport measurements are used for read-out. This paved the way for impressive experimental breakthroughs in quantum-dot physics in the last ten years [24–27]. However, all these results were obtained with GaAs-structures in which spin-coherence times are limited due to rather strong hyperfine and spin-orbit interaction. Graphene, on the other hand, is expected to have comparatively long coherence times because of the reduced hyperfine interaction\(^4\) and the small nuclear charge of carbon leading to a weak spin-orbit interaction [28].

With the big goal of using graphene nanostructures for information processing, many research groups worldwide perform transport experiments on these devices. Important steps along the way are the measurement of Coulomb blockade, excited states, identification of spin states, fabrication of double dots, spin blockade, identification of singlet and triplet states, charge detection etc. Some of these steps were achieved by now, others still await their successful demonstration. All steps are challenging but full of interesting physics.

\(^4\)The isotope \(^{12}\)C has a nuclear spin of zero, whereas \(^{13}\)C has a nuclear spin 1/2. However, the natural abundance of 1.1% of \(^{13}\)C is very small.
1.2.2 Scanning-Probe Experiments

The measured variable in conventional transport experiments, i.e. typically the current through the device, is a macroscopic quantity. Consequently, spatial information is difficult to extract from these experiments. Scanning-probe experiments, where a physical quantity is recorded as a function of probe position, are therefore an essential addition to the existing tools for semiconductor research: They give access to the underlying microscopic properties of nanostructures.

With the advent of graphene as a new material system, it is obvious to employ scanning probes in graphene research as well. First of all, there is a technical reason: Since the two-dimensional electron gas sits at the surface, the probe-sample separation can be reduced to nominally zero. This will result in a better spatial resolution compared to conventional two-dimensional electron gases. There, the minimal probe-sample separation is given by the thickness of the dielectric capping layer. A second technical reason is that graphene allows for the implementation of an additional imaging technique which has been impractical so far for semiconductor heterostructures, namely scanning-tunneling microscopy/spectroscopy. With these technical advantages and possibilities, the “holy grail” of local-probe techniques – namely wave-function mapping – might come feasible. This would not only be restricted to bulk graphene, but could also be applied to nanostructured graphene, e.g. graphene quantum dots. Completely new insights into quantum-dot physics could be obtained!

However, this is the prospect for the future. For now, theoretical proposals regarding scanning-probe experiments on graphene are scarce and propose to test graphene’s special properties like its chirality [29] or the appearance of zitterbewegung (trembling motion) [30]. Albeit interesting proposals, they are hard to realize experimentally. On the other hand, very different probing techniques have already been applied to graphene as will be discussed below. Nanostructured graphene, however, has not been studied with these techniques as it was done with nanostructures based on different materials [31–35]. For a better understanding, improvement of the design, and the local manipulation, it is highly desirable to perform scanning-probe experiments on graphene nanostructures as well.

Regarding quantum dots with typical dimensions of the order of 100 nm, a scanning-gate microscope is a particularly well-suited tool since it offers the necessary spatial resolution (in contrast to e.g. existing scanning-SET setups). In scanning-gate microscopy, a tip acts as a local gate as it will be discussed in chapter 3.2. Transport through the quantum dot therefore depends on the position of the tip. Wave-function mapping is in principle also possible with this technique; primarily, it allows for the detection of localized states in real space. In this thesis, we report on the first application of scanning-gate microscopy to a graphene quantum-dot structure.
Overview over Scanning-Probe Experiments on Graphene

A first scanning-probe study, namely scanning-tunneling spectroscopy on bulk graphene\(^5\) was performed in the group of Michael Crommie by V. Brar et al. Later experiments carried out by Y. Zhang et al. in the same group confirmed their previous result, namely the appearance of a gap-like feature in the tunneling spectrum \(^37\). The authors attribute the gap to a phonon-mediated inelastic tunneling process. A later study of this group used a new technique called “Dirac-point mapping” to show that variations of the charge density in graphene is not caused by topographical variations, i.e. ripples, but rather by impurities below the graphene \(^38\). In the same publication, the authors also present images of charge puddles as shown in Fig. 1.2(a). The puddles have a characteristic length of \(\sim 20 \text{ nm}\). Similar results were also obtained by A. Deshpande et al. using a related technique for measuring the shift of the Dirac point \(^39\). G. Li et al. performed spectroscopic experiments of the same type on graphite where Landau-level spectroscopy proved that single layers can detach from the bulk and show graphene physics \(^40\). A recent experiment used tunneling spectroscopy to measure Landau levels in graphene “nanobubbles” in strain-induced pseudo-magnetic fields. The nanobubbles form when graphene is grown on a platinum surface. The deduced pseudo-magnetic fields were as high as 300 T \(^41\).

The existence of electron-hole puddles had been confirmed by J. Martin et al. in the group of A. Yacoby before the spectroscopic experiments took place. They used a scanning single-electron transistor to map the local inverse compressibility from which they could calculate the spatial charge-density fluctuations \(^9\). In their case, typical extension of puddles are about 150 nm as seen in Fig. 1.2(b) but the authors argue that this length scale is limited by the spatial resolution of their probe. The same group also looked at the quantum Hall regime. J. Martin et al. could show that – despite strong disorder – localization is not governed by single-particle physics, but instead by an interplay of the disorder potential and the repulsive Coulomb interaction leading to screening \(^42\).

Scanning-gate measurements on graphene were realized later than the experiments described before. J. Berezovsky et al. in the group of R. Westervelt used a tip as one additional scatterer to probe coherent transport and universal conductance fluctuations in bulk graphene \(^43\). A subsequent experiment focused on weak localization: The tip created a controllable local change to the disorder potential and thus changed the coherent backscattering. Using this technique, the authors determined the diffusive coherence length to be about 500 nm at 4.2 K \(^44\). Scanning-gate microscopy was also used to look at the effects of current-annealing of graphene on the local conductivity \(^45\). Furthermore, the same group

\(^{5}\)This study was done on epitaxially grown graphene whereas the later ones were done on mechanically cleaved graphene.
1.2. Motivation for Research on Graphene Nanostructures

Figure 1.2: (a) $dI/dV$-map of a $60 \times 60 \text{nm}^2$ area taken from Ref. 38 Fig. 2b and obtained with scanning-tunneling spectroscopy. Differently charged areas, the so-called charge puddles, show up as either yellow or blue areas and possess a characteristic length of $20 \text{ nm}$. (b) Charge-density plot of a $3 \times 1.8 \mu\text{m}^2$ area taken from Ref. 9 Fig. 3a and obtained with a scanning single-electron transistor. Blue regions correspond to holes whereas red regions depict electrons. Typical extensions of the puddles in this measurement are about $150 \text{ nm}$, a value which is limited by the resolution of the probe.

performed scanning-gate experiments on a graphene nanoribbon. Their results indicated that a localized state in the nanoribbons showed quantum-dot behavior induced by potential inhomogeneities [46]. R. Jalilian et al. in the group of Y. P. Chen did extensive scanning-gate studies on graphene field-effect transistors at room temperature. Moreover, they approached their sample as close as possible by using contact-mode SGM with a dielectric-coated metallic nanowire as a tip. Images of puddles showed typical extensions of more than $200 \text{ nm}$, indicating that their resolution is – as in the case of the scanning SET – limited by the size of their tip. More interestingly, the authors predominantly found puddles close to the contacts, at the graphene edges, and around scratches in the graphene flake [47].

F. Giannazzo et al. report on a fourth technique used to locally study graphene, namely scanning-capacitance spectroscopy. With this tool at hand, the screening length of graphene and – deduced from that – the local quantum capacitance of graphene was measured [48]. The lateral variations of the density of states they found are in agreement with previous scanning-probe studies.
Chapter 2

Theoretical Background of Quantum Dots

The expression *quantum dot* refers to the geometry of such a device, namely an island – the *dot* – which is coupled to leads via highly resistive elements, e. g. tunnel barriers. Under suitable experimental conditions as low temperature and strong confinement of charge carriers, these islands show quantum behavior.

The discreteness of the electronic charge leads to a phenomenon known as *Coulomb blockade*. In the regime of Coulomb blockade, quantum dots show the behavior of a *single-electron transistors* (SET). Hence, the chip industry is interested in this kind of research since SETs might be a complement to the existing zoo of transistor types. However, additional quantum mechanical properties of quantum dots make them also interesting for fundamental research. Especially the possibility of processing information quantum-mechanically with interacting quantum dots is one of the driving forces in quantum-dot research [23].

In this chapter, we will introduce the *constant-interaction model* for describing the discrete energy levels of a quantum dot. Quantum confinement effects will not be specified further in the first section so that the description does not differ from those of conventional semiconductors. We will give a short overview and refer to the literature for derivations. The quantum behavior of graphene quantum dots, on the other hand, is expected to be different because of graphene’s unique bandstructure. In the second and third section, the quantum-mechanical confinement effects will be investigated theoretically – first without and later with an external magnetic field.
2.1 The Constant-Interaction Model

In Fig. 2.1(a), we present a schematic of a quantum-dot device. The dot itself is coupled to source and drain leads with electrochemical potential $\mu_{S,D}$ via tunnel barriers with tunnel coupling rates $\Gamma_{S,D}$. A real graphene quantum-dot device is presented in Fig. 2.1(b). A bias voltage $V_{\text{bias}}$ can be applied between source and drain contacts so that $eV_{\text{bias}} = \mu_S - \mu_D$. Under suitable experimental conditions, the phenomenon of Coulomb blockade can be observed. In this regime, the electrochemical potential $\mu_N$ of the quantum dot is discrete. In the constant-interaction model [49], it can be expressed as

$$\mu_N = \epsilon_N + \frac{e^2}{C_\Sigma} \left( N - \frac{1}{2} \right) + e \sum_{i=1}^{n} \frac{C_{0i}}{C_\Sigma} \left( V_i - V_i^{(0)} \right),$$

(2.1)

where the summation runs over all gates of the device to which a voltage $V_i$ is applied; $V_i^{(0)}$ is an offset voltage of that gate. The quantity $N$ is the number of charge carriers trapped on the dot, and $C_\Sigma := -\sum_{i=1}^{n} C_{0i}$ is the total capacitance of the quantum dot to its environment. Quantum mechanical contributions to $\mu_N$ are summarized in $\epsilon_N$; this will be discussed in more detail in the next section. The capacitance coefficients $C_{0i}$ are the capacitances between gate $i$ and the quantum dot; the ratio $\alpha_i := -C_{0i}/C_\Sigma$ is called the lever arm of gate $i$. Current flows whenever $\mu_N$ is in the bias window of the electrochemical potentials of source and
2.1. The Constant-Interaction Model

Figure 2.2: Energy diagram of a quantum dot for finite bias $eV_{\text{bias}} = \mu_S - \mu_D$ when current is flowing (a) or when current is blocked (b). The light grey shaded areas are the bias windows; whenever one of the discrete energy states $\mu_N$ lies within, current through the dot can flow. This is depicted with blue arrows in (a). In (c), the zero-bias case is shown when the dot is in resonance with source and drain. Current can then flow which results in a Coulomb-resonance peak in the current $I_{\text{dot}}$ through the dot. An experimental trace of $I_{\text{dot}}$ as a function of gate voltage and for zero bias is depicted in (d). The distance between two Coulomb resonances is proportional to the charging energy with the lever arm of that gate being the proportionality constant, $\Delta E_C = e\alpha_{BG}V_{BG}$.
Chapter 2. Theoretical Background of Quantum Dots

drain, i.e. $\mu_D \leq \mu_N \leq \mu_S$ (or reversed bias). This situation is schematically depicted in Fig. 2.2(a). Otherwise, the current is blocked due to Coulomb repulsion, Fig. 2.2(b).

The energy difference $\mu_{N+1} - \mu_N$ between two discrete energy states of the quantum dot is called the charging energy $\Delta E_C$. It can easily be determined in transport experiments as it will be explained below and it is given by

$$\Delta E_C = \mu_{N+1} - \mu_N = \frac{e^2}{C_\Sigma} + \Delta(N),$$

where $\Delta(N) = \epsilon_{N+1} - \epsilon_N$. The capacitance $C_\Sigma$ cannot be calculated analytically in general. For a rough estimate, the quantum dot can be considered as a metallic disc embedded in a material with relative dielectric constant $\varepsilon$ and infinite extent. Then, $C_\Sigma$ is approximately equal to the self-capacitance of a disc, $C_\Sigma \approx 8\varepsilon\varepsilon_0 r$. Typical charging energies are in the range of a few meV’s.

The charging energy is also the crucial energy scale setting the experimental conditions for observing Coulomb blockade. A finite temperature $T$ of the electronic system smears out the Fermi edge in the source and drain leads and leads to thermal broadening of experimental signatures like Coulomb resonances (see below). Therefore, a necessary requirement is

$$k_B T \ll \Delta E_C.$$

Charging the quantum dot with an additional charge carrier takes the time $\Delta t = R_{tb} C_\Sigma$ with $R_{tb}$ being the resistance of a tunnel barrier. Heisenberg’s uncertainty principle $\Delta E_C \Delta t \geq \hbar$ must be respected if the charging event is experimentally resolvable. It follows that

$$R_{tb} > \frac{\hbar}{e^2} \approx 25.8 \text{ k}\Omega.$$

The resistance $R_{tb}$ of the tunnel barriers is mainly determined by the tunnel coupling rates $\Gamma_{S,D}$. Hence, they have to be tuned such that the above condition holds. Otherwise, the charge on the quantum dot is not quantized.

According to Eq. (2.1), the electrochemical potential of the quantum dot can be continuously swept by tuning a plunger gate. If the current through the quantum dot is recorded as a function of plunger gate voltage $V_{PG}$ and for small bias, $eV_{\text{bias}} < k_B T$, Coulomb resonances occur whenever the electrochemical potentials of source, dot, and drain are in resonance, $\mu_S = \mu_N = \mu_D$. This situation is depicted in Fig. 2.2(c); an experimental trace is shown in Fig. 2.2(d).

This section provides an overview over the constant-interaction model as far as it is relevant for this thesis. Extensive discussions can be found in Refs. [49–51].
2.2. Graphene Quantum Dots and Quantum Confinement

Figure 2.3: Energy diagram of a quantum dot where current transport takes place through excited states. (a) Current through an excited state which is in resonance with the source lead. (b) One possible inelastic co-tunneling process. Here the quantum dot remains in a virtual state after process 1. In order to fulfill Heisenberg’s uncertainty relationship for energy and time, process 2 has to occur on a corresponding time scale. The whole process is called inelastic because the final state is higher in energy than the initial one by an amount $\Delta$.

2.2 Graphene Quantum Dots and Quantum Confinement

Whenever a quantum mechanical particle is confined to a certain region in space, discrete energy states are a consequence of the wave nature of the particle. This of course is also true for charge carriers trapped on a quantum dot and leads to the quantum-mechanical contribution $\epsilon_N$ to the electrochemical potential in Eq. (2.1).

If we neglect electron-electron interactions, the total confinement energy can be approximated by

$$E_{\text{conf}}(N) = A \int_0^{E_F} dE \mathcal{D}(E) = \frac{2}{3} \hbar c_s \sqrt{\frac{\pi}{A}} N^{3/2},$$

where $A$ is the area of the quantum dot and $\mathcal{D}(E) = 2E/ (\pi \hbar^2 c_s^2)$ is the density of states of graphene. The contribution to the electrochemical potential can then be calculated as the difference of the confinement energies for $N+1$ and $N$ charge carriers,

$$\epsilon_N = E_{\text{conf}}(N+1) - E_{\text{conf}}(N) = \frac{2}{3} \hbar c_s \sqrt{\frac{\pi}{A}} \left( N^{3/2} \left( 1 + \frac{3}{N} + \frac{3}{N^2} \right)^{1/2} - N^{3/2} \right)$$

$$\approx \hbar c_s \sqrt{\frac{\pi N}{A}}.$$
Chapter 2. Theoretical Background of Quantum Dots

The approximation done in the last step is valid for \( N \gg 1 \). In other words, the above expression must not be used very close to the CNP where one expects \( N \) to be around zero. The expression for \( \epsilon_N \) is the same as for the Fermi energy in bulk graphene if we identify \( N/A \) with the charge carrier density \( n \). However, one has to keep in mind that densities in the bulk and on the dot are in general not the same. The single-particle level spacing which is the relevant quantity for transport experiments and which also enters the charging energy \( E_C \), Eq. (2.2), is then

\[
\Delta(N) = \epsilon(N+1) - \epsilon(N) \approx \hbar c_s \sqrt{\frac{\pi}{A}} \left( (N+1)^{1/2} - N^{1/2} \right)
\]

\[
\approx \frac{\hbar c_s}{2} \sqrt{\frac{\pi}{A}} \frac{1}{\sqrt{N}}, \tag{2.3}
\]

where \( N \gg 1 \) as above.

In the previous section, we described how current can flow through a quantum dot as charge carriers hop on and off the discrete states of the dot without explicitly considering the quantum-mechanical contributions. Analogously, charge transport can also occur through the excited states spaced by \( \Delta(N) \) due to quantum confinement. In Fig. 2.3(a), one possibility how this can happen is depicted. Inelastic co-tunneling is a second-order process which can also take place and which is shown in Fig. 2.3(b): Initially, the quantum-dot state with electrochemical potential \( \mu_N \) is occupied. Borrowing “energy from the vacuum”, the charge carrier tunnels out into the drain (process 1) and remains in a virtual state. In the next step, a charge carrier tunnels from the source into an excited state (process 2). Process 1 and 2 have to occur on a time scale given by Heisenberg’s uncertainty relationship to not violate energy conservation. The complete cycle is referred to be inelastic because the final state is higher in energy by an amount \( \Delta \) than the initial state. This energy is provided by the source-drain bias. Therefore, inelastic co-tunneling is only observable when \( \Delta \leq eV_{\text{bias}} \).

We compare the ratio of energy scales \( \Delta(N)/(e^2/C_\Sigma) \) to the case of a conventional 2DEG with parabolic dispersion. In a conventional 2DEG, \( \Delta(N)/(e^2/C_\Sigma) \propto 1/r \). The quantization energy becomes dominating for \( r < 10 \) nm in gallium arsenide and \( r < 30 \) nm in indium arsenide \([50]\). The situation is completely different for graphene: Assuming a disk with radius \( r \), \( A = \pi r^2 \), we find \( \Delta(N)/(e^2/C_\Sigma) = 1/(\pi c_s \sqrt{N}) \). The ratio is independent of dot size but depends on the number of charge carriers. Interestingly, the transition to the regime where the quantization energy dominates is given by graphene’s fine-structure constant, namely for \( N < 1/(\pi c_s)^2 \). Since this is of order 1, this regime is experimentally hardly accessible. The measurements presented in chapter 4 support these considerations.
Bias Spectroscopy

We can experimentally deduce the relevant energy scales of a quantum dot when performing bias spectroscopy, i.e. when the bias voltage is swept and the plunger gate is stepped (or vice versa). A schematic measurement outcome is shown in Fig. 2.4. The white regions of suppressed current are called Coulomb-blockade diamonds. Their extent in bias direction is a direct measure of the charging energy, $\Delta E_C = e\Delta V_{\text{bias}}$. The lever arm of the plunger gate is given by the ratio $\alpha_{\text{PG}} = \Delta V_{\text{bias}}/\Delta V_{\text{PG}}$. Excited states spaced by the single-particle level spacing can be seen as lines of high conductance running in parallel to the edge of Coulomb diamonds. They are depicted as dashed, black lines outside the diamond with $N$ charge carriers. Transport within the diamond-shaped region is forbidden for first-order processes. However, transport employing virtual quantum states is possible as discussed above. The onset of inelastic co-tunneling is shown in diamond $N$ with a dashed, black...
line where the condition $\Delta = eV_{\text{bias}}$ is fulfilled. The inelastic co-tunneling onset is aligned with an excited-state line at the edge of the diamond.

## 2.3 Graphene Quantum Dots in a Perpendicular Magnetic Field

In the previous sections, we saw that the spacing of Coulomb peaks in a particular gate voltage is proportional to the charging energy where the proportionality constant is essentially the lever arm of that gate. What happens if the quantum dot is exposed to a perpendicular magnetic field? The contribution to the charging energy which is determined by the geometry of the quantum dot, $e^2/C_{\Sigma}$, stays constant. The single-particle level spacing $\Delta$, on the other hand, does depend on the magnetic field. This dependence $\Delta(B)$ can be calculated analytically in the case of quantum dots with a parabolic dispersion and a parabolic confinement potential. The so-called Fock-Darwin spectrum shows how energy levels evolve as a function of magnetic field \[52, 53\] and has been verified under suitable experimental conditions \[54\].

In the following, we want to establish a corresponding energy spectrum for graphene quantum dots in a magnetic field and look at the transition from confinement-dominated to magnetic field-dominated behavior of the charge carriers. Especially, we expect that the hole energy levels shift in the opposite direction as the electron energy levels because of the electron-hole symmetry. This would enable experimentalists to determine the electron-hole crossover by employing magnetic-field spectroscopy.

In Ref. \[55\] the authors just consider positive energy states and electrostatic potential barriers. This circumvents the problem of Klein tunneling. However, it is a theoretical trick which is experimentally not realizable. More realistic calculations have been carried out by employing other boundary conditions or by doing numerical tight-binding calculations \[56–59\].

Here, we will present an analysis based on the infinite-mass boundary condition which allows us to determine the energy spectrum analytically \[60\]. The infinite-mass boundary condition was originally derived as a toy model for two-dimensional neutrinos \[61\]. We will extend the discussion by coupling an external perpendicular magnetic field $B$ in symmetric gauge to the Dirac equation expressed in cylindrical coordinates. The vector potential then reads $A = B/2(-y, x, 0) = B/2(-r \sin \phi, r \cos \phi, 0)$ with $\phi$ being the polar angle. The Hamiltonian is given by

$$H = c_\alpha (\mathbf{p} + eA) \cdot \mathbf{\sigma} + \tau V(r)\sigma_z.$$ \hspace{1cm} (2.4)

The Dirac equation is $H\psi(r, \phi) = E\psi(r, \phi)$ with the wave function being a two-
component spinor, \( \psi(r, \phi) = (\psi_1(r, \phi), \psi_2(r, \phi)) \). The vector \( \boldsymbol{\sigma} = (\sigma_x, \sigma_y) \) contains Pauli’s spin matrices in the basis of the two sublattices of A- and B-atoms as its components. The electron spin is neglected in our analysis. A mass-related potential energy \( V(r) \) is coupled to the Hamiltonian via the \( \sigma_z \) Pauli matrix. The mass in the dot is zero, \( V(r) = 0 \) for \( r < R \), but tends to infinity at the edge of the dot, \( V(R) \to \infty \), for a circular dot of radius \( R \). In this way, charge carriers are confined inside the quantum dot. This leads to the infinite-mass boundary which yields the simple condition that

\[
\psi_2/\psi_1 = \tau i \exp[i\phi]
\]

for circular confinement \cite{61}. The parameter \( \tau \) takes the two values \( \pm 1 \) for distinguishing the two valleys \( K \) and \( K' \). We can therefore set \( V(r) = 0 \) in the following and we will respect the different energy spectra of the \( K \)- and \( K' \)-valleys via the boundary condition. The derivation of the wave function \( \psi(r, \phi) = (\psi_1(r, \phi), \psi_2(r, \phi)) \) will be carried out in appendix A. Employing Eq. (2.5), we finally arrive at the following implicit formula describing the energy spectrum

\[
(1 - \tau kl_B/R/l_B) L\left(\frac{k^2l_B^2}{2} - (m + 1), m, \frac{R^2}{2l_B^2}\right) + L\left(\frac{k^2l_B^2}{2} - (m + 2), m + 1, \frac{R^2}{2l_B^2}\right) = 0.
\]

The magnetic length is defined according to \( l_B = \sqrt{\hbar/eB} \). The functions \( L(a, b, x) \) are generalized Laguerre polynomials which are oscillatory functions. Hence, there is an infinite number of wave vectors \( k_n \) for given \( B \), angular momentum quantum number \( m \), and \( \tau \) which fulfill the above equation. This defines the radial quantum number \( n \) which labels the roots of the left part of Eq. (2.6). The energy spectrum \( E(n, m, \tau) = \hbar c k_n \) of charge carriers confined to a circular graphene quantum dot which is exposed to a perpendicular magnetic field is determined through Eq. (2.6). The relation \(-E(n, m, \tau) = E(n, m, -\tau)\) is a manifestation of the electron-hole symmetry.

In appendix A we will also derive the approximate solutions for the two limits \( B \to 0 \) and \( R/l_B \to \infty \) for Eq. (2.6). For \( B \to 0 \), we retrieve the result from Ref. \cite{61} namely

\[
\tau J_m(kR) = J_{m+1}(kR),
\]

where \( J_m(x) \) is a Bessel function of the first kind. It can be used to estimate the number of charge carriers confined on a graphene dot when the energy of an excited state is measured. This is an alternative to the density of states argument derived

\footnote{The potential is positive for sublattice A and negative for sublattice B. This does not seem to be very physical. On the other hand, the edge structure cannot be controlled experimentally and the chemical structure at the graphene edge is unknown. Hence, every calculation relies on assumptions which cannot be verified experimentally \cite{57, 62, 63}.}
Figure 2.5: Energy spectrum of a quantum dot with \( R = 70 \) nm which was numerically calculated from Eq. (2.6). The formation of the lowest Landau levels can be seen as predicted by Eq. (2.8). Energy states for \( \tau = +1 \) are drawn with black, solid lines, those for \( \tau = -1 \) with green, dashed lines.

In the previous section. Moreover, we can deduce that \( E(n, m, \tau) = E(n, -m - 1, -\tau) \) for \( B = 0 \), derived from the property \( J_m(x) = (-1)^m J_{-m}(x) \). Consequently, pairs of states are degenerate at zero magnetic field. There is no state at zero magnetic field and zero energy [61]. This leads to an energy gap between negative and positive energy states.

In contrast, in the limit \( R/l_B \rightarrow \infty \) we retrieve

\[
E_m = \hbar c_s k_m = \pm c_s \sqrt{2e\hbar B(m + 1)}
\] (2.8)

which are the well-known Landau levels for graphene. Therefore, there will be a transition, governed by the parameter \( R/l_B \), from a regime where the energies of the charge carriers are dominated by confinement (Eq. (2.7)) to Landau levels (Eq. (2.8)). This transition including these two limiting cases is described by Eq. (2.6).

We evaluate Eq. (2.6) for a dot of radius \( R = 70 \) nm which is about the same size as the device presented in chapter 4. The energy spectrum as a function of magnetic field is shown in Fig. 2.5 for \( m = -4, \ldots, 4 \) and \( n = 1, \ldots, 6 \). For \( B = 0 \), the energy states are not equidistant. For higher magnetic fields, we can see the formation of Landau levels according to the previous discussion. The zero-energy
2.3. Graphene Quantum Dots in a Perpendicular Magnetic Field

Landau level is formed by states with quantum number \( \tau = -1 \) and \( E > 0 \) and those with \( \tau = +1 \) and \( E < 0 \).

The lowest positive energy state has an energy of about 4 meV for \( B = 0 \). This yields an energy gap of around 8 meV between electron and hole states. Since the energy gap to the next excited state is much lower, the electron-hole transition may be detected experimentally by a confinement-enhanced energy gap. Additionally, the first resonances on both sides of the electron-hole crossover have opposite slopes and merge into the zeroth Landau level. The experimental observation of this effect would clearly identify the crossover.
Chapter 3

Experimental Setup, Scanning-Probe Techniques

From the previous chapter, it follows that low temperatures are an essential ingredient in order to observe phenomena like Coulomb blockade or other aspects of quantum-dot physics. Typical charging energies of quantum dots are in the meV-range which corresponds to a temperature of around 10 K. Therefore, measurements on Coulomb blockade are usually carried out at the temperature of liquid helium, $T \approx 4.2$ K, or below. If e.g. spin phenomena are in the experimentalist’s focus, then much lower temperatures are required. For example, the Zeeman splitting at $B = 1$ T and gyromagnetic factor $g = 2$ (expected for charge carriers in graphene) corresponds to a temperature $T \approx 1$ K. Today, cryostats are commercially available which reliably obtain temperatures well below 100 mK – so-called dilution refrigerators. The experiments presented here were carried out in a dilution refrigerator.

Local properties of graphene structures are one focus of this thesis. In a conventional semiconductor heterostructure, the 2DEG is buried some tens of nanometers below the surface. Electrostatic interactions between a sharp, metallic tip scanned above the surface and the 2DEG below can reveal local properties of the electronic system. For this kind of experiment, called scanning-gate microscopy (SGM), we equipped a dilution refrigerator with a home-built atomic-force microscope (AFM) and use its metallic tip as the scanning gate. The 2DEG is at the surface in graphene and directly accessible by the AFM tip. It is therefore a promising candidate for a better resolution in SGM compared to semiconductor heterostructures.

For scanning-tunneling microscopy (STM), the tunneling current between a conducting tip and the conducting system is measured. With our scanning-gate setup and graphene as the object of investigation, STM is also possible. STM can yield information which is not available through SGM like the local density of
states. The combination of both techniques is therefore highly desirable but also challenging.

In the first section of this chapter, we will give a short introduction to our setup. Then, the principle of SGM will be explained. The final section is devoted to STM, namely to the key idea of STM and then to the optimization of feedback parameters in the STM mode.

3.1 The Dilution Refrigerator Atomic Force Microscope

The dilution refrigerator used for the experiments presented in this thesis is a Kelvinox MX 100 manufactured by Oxford Instruments. It has a nominal cooling power of around 100 \( \mu \text{W} \) at a temperature of 100 mK at the mixing chamber. The base temperature of our setup is around 36–40 mK. At the mixing chamber plate, a sample holder and a home-built AFM were assembled. A home-built slip-stick motor allows the coarse lateral positioning of the sample holder with respect to the AFM tip. A second slip-stick motor moves the scan piezo and the AFM-sensor including the tip in \( z \)-direction for approaching the sample. The approach can be monitored with a capacitively-working positioning sensor \[64\]. The scan piezo itself is rather long (63.5 mm) which gives a scan range of roughly \( 7 \times 7 \mu \text{m}^2 \) at base temperature.

The AFM-sensor is the heart of the AFM and consists of a commercial quartz tuning fork and the tip connected to the upper prong of the tuning fork. Force detection in AFM-mode is not done by laser deflection, but by measuring the resonance frequency shift of the tuning fork when a force gradient is acting on it \[65\]. The tuning fork is always excited at its resonance frequency of initially around \( f_{\text{res}} = 2^{15} \text{ Hz} \) using a phase-locked loop (PLL). Laser deflection detection is not applicable for two reasons: First of all, many of our samples are optically active. Secondly, the laser power would heat up the environment and topographic scans at base temperature would be impossible. Tuning forks, on the other hand, allow for an all-electric force detection. Moreover, their high \( Q \)-factor of around 8000 under ambient conditions and up to 500000 at base temperature minimizes power dissipation. At room temperature, we can estimate the mechanical energy stored in the tuning fork to be of the order of \( E_{\text{mech}} \sim k \Delta x^2 \approx 3 \text{ pJ} \) for a spring constant \( k \approx 30 \text{kN/m} \) and an oscillation amplitude \( \Delta x \approx 10 \text{ nm} \) \[66\]. This relates to a power dissipation of \( P = f_{\text{res}} E_{\text{mech}} / Q \approx 10 \text{ pW} \). At base temperature, the power dissipation should even be less because of the higher \( Q \)-factor. Correspondingly, topographic scans should not lead to an increase of the temperature. This is, indeed, not observed in the experiments. On the other hand, a large \( Q \)-factor
3.2. Scanning-Gate Microscopy

reduces the bandwidth of the AFM. The PLL helps to increase the bandwidth again at the expense of a higher noise level \cite{67}.

A Pt80/Ir20-wire with a diameter of 15 \(\mu\text{m}\) forms the tip. It is sharpened by electrochemical etching. Tip diameters of around 50 nm can be obtained in that way. However, this is a highly non-reproducible step leading to different tip shapes and aspect ratios for every sensor. The wire is glued to the upper prong of the tuning fork. We want to apply a voltage to the tip for SGM and use it as a local gate. Therefore, it is crucial to use a conducting tip. A positive side effect is that these tips can also be employed for STM.

In total, 66 cables connect the microscope and the sample holder to the outside world. Twelve of them are superconducting cables for high voltages which drive the slip-stick motors and the scan piezo. 24 twisted-pair constantan cables are used for thermometry and heating of the sample. The remaining 30 cables are semi-rigid stainless steel coaxial cables. 24 of them connect to the sample holder whereas the others are used for the AFM-sensor and the positioning sensor. Thermal anchoring of the coaxial cables takes place at two stages, namely at the 1K-pot plate and at the mixing chamber plate. The base temperature of below 40 mK measured at the mixing chamber of the dilution refrigerator proves the success of the thermal anchors.

The AFM is controlled by Nanonis hard- and software (SPECS Zurich GmbH). The optimization of feedback parameters is implemented in the software. Initially, the \(Q\)-factor of the tuning fork is determined by measuring its resonance curve. This sets the integral part of the feedback parameters of the proportional-integral controller (PIC) of the PLL. The \(z\)-controller holds the tip at a constant height above the surface such that the resonance frequency shift is kept constant. Its integral part is also determined through the \(Q\)-factor. The only free parameters which have to be set by the experimentalist are the proportional parts of both the PLL and the \(z\)-controller. Their values are a trade-off between a high bandwidth and a low noise level.

The details of the microscope, the AFM-sensor, and the optimization of the feedback parameters are described in Refs. \cite{67} \cite{68}.

3.2 Scanning-Gate Microscopy

The idea behind scanning-gate microscopy is straightforward: We take the conducting tip of an AFM and apply a voltage to it. Then, the tip is scanned at constant height over the nanostructure while we record the current through the structure as a function of tip position. In such a way, the tip acts as a gate which induces – depending on the aspect ratio of the tip and the voltage applied to it – a local potential perturbation in the sample. This is used to probe the sample
locally \[31, 34, 69\]. For SGM on a quantum dot, we can employ the capacitance model and rewrite Eq. (2.1) as \[70\]

$$
\mu_N (r_{\text{tip}}) = \epsilon_N (r_{\text{tip}}) + \frac{e^2}{C_{\Sigma} (r_{\text{tip}})} \left( N - \frac{1}{2} \right) + e \sum_{i=1}^{n} \frac{C_{0i} (r_{\text{tip}})}{C_{\Sigma} (r_{\text{tip}})} V_i + e \frac{C_{\text{tip}} (r_{\text{tip}})}{C_{\Sigma} (r_{\text{tip}})} V_{\text{tip}},
$$

(3.1)

where the tip is treated as an additional gate with tip-dot capacitance $C_{\text{tip}} (r_{\text{tip}})$. Offset voltages of the gates and the tip are neglected. All capacitances depend on the tip position because of mutual screening effects. In particular, this implies that the charging energy of the quantum dot depends on the tip position which has been confirmed by experiments on a GaAs-dot and a metallic SET \[35, 70\].

Coulomb resonances occur for $\mu_{S,D} = \mu_N$ in the low-bias case. Therefore, Coulomb resonances are contour lines of the electrochemical potential $\mu_N$ and the tip-induced potential in a scanning-gate image \[68, 69\].

Wave-function mapping, i.e. measuring the spatial distribution of the dot’s wave function $|\Psi_{\text{dot}} (r)|^2$, is the holy grail of SGM on quantum dots. In first-order perturbation theory, the tip-induced shift of the dot’s energy levels is the convolution of the tip-induced potential and the wave function. The shift of the energy level is obtained by SGM; the tip-induced potential can be approximated by a Lorentzian function or also experimentally determined. Then, the unperturbed wave-function amplitude $|\Psi_{\text{dot}} (r)|^2$ can be obtained by deconvolution \[71\]. So far, this goal has not been achieved in any group worldwide performing SGM because the experimental conditions were not favorable enough. Prerequisites for wave-function mapping are: (i) a tip-induced potential with a FWHM of the order of the Fermi wave length or smaller, (ii) a tip-sample separation as small as possible, and (iii) an as least-invasive probe as possible, i.e. applying only small voltages to the tip \[70, 71\]. Fermi wave lengths in our nanostructures are of the order of 50 nm.

The first requirement is therefore hard to fulfill as we will discuss in Chap. 6. The second one is easily accomplishable with samples where the electron gas is at the surface without a dielectric capping layer, e.g. InAs-nanowires or graphene. The third requirement, however, is again hard to implement although a lot of efforts have been done \[34, 69, 71\]. Related issues will also be discussed in Chap. 6.

### 3.3 Scanning-Tunneling Microscopy

Historically, STMs were developed before the AFM \[72, 73\]. The key idea is to use the tunneling current between the sample and a conducting tip as the input signal for topographic imaging. Since the tunneling current depends exponentially on the tip-sample separation $z$, small changes in $z$ are comparably easy to detect. STMs
have the best lateral and vertical spatial resolutions of all scanning-probe microscopes; atomic resolution is commonly achieved. A big disadvantage is that STM can only be used in combination with conducting surfaces or substrates. Hence, SGMs were used instead of STMs to investigate semiconductor nanostructures.

Graphene, on the other hand, is a unique material system where the two-dimensional electron gas is directly accessible. Consequently, STM on graphene is possible and has already been performed \([37,40]\). The advantage of our system is that both STM and AFM are possible. In principle, it is possible to record a topographic AFM image first in order to locate the edges of the graphene structure. Then, the STM-measurements can be performed. Alternatively, AFM and STM are performed simultaneously. In both cases, the danger of tip crashes by crossing the edge of the graphene flake can be minimized. STM on small graphene nanostructures is therefore challenging but possible. Interesting experiments could be for example: Tunneling into edge channels in the quantum Hall regime, tunneling into a quantum dot and detection of the tunneling current with a nearby charge detector \([74]\), scanning-tunneling spectroscopy on nanostructures to determine the local density of states, etc.

STM is a challenge with our setup since it has not been optimized for scanning on an atomic scale. First of all, the scan piezo is rather long and atomic resolution in lateral direction is at its noise limit. Secondly, mechanical vibrations due to normal laboratory work are visible in the tunneling current and obscure the desired signal. However, the operation in STM-mode with NANONIS is possible and has been successfully tested. The optimization of feedback parameters is crucial and will be discussed in the next section.

### 3.3.1 Optimization of Feedback Parameters for the STM Mode

We will model our system in STM-mode in linear response. The approach is similar as in Refs.\([66,68]\) where analogous studies are carried out for the AFM with a PLL. In the case of STM, the analysis is simpler because the control loop consists of only one feedback loop, namely the one which keeps the tunneling current \(I_{tc}\) at a constant value and varies the height of the STM-tip for that purpose.

The STM control circuit consists of two main elements, namely the IV-converter which converts the tunneling current \(I_{tc}\) into a voltage. This is then fed into the NANONIS hardware where a PIC varies the tip-sample separation \(z\) in order to keep the tunneling current at a fixed value \(I_{set}\). In appendix\([3]\) we derive an expression for the response function and the corresponding bandwidth of our STM using linear-response theory and starting from these two elements.

From this, it follows that the STM shows low-pass behavior with response
function and bandwidth given by

\[
Z_{\text{STM}}(f) = \left(1 + i \frac{f}{f_{\text{STM}}} \right)^{-1},
\]

\[
f_{\text{STM}} = \mu \beta P f_{\text{PI}}.
\]  

(3.2)

Here, \(\mu\) is a proportionality constant defined in appendix B. The frequency \(f_{\text{PI}}\) is the bandwidth of the PI-controller which equals the bandwidth of the IV-converter, \(f_{\text{PI}} = f_{\text{IV}}\), according to the discussion in appendix B. The only parameter which can be varied in the software directly is the proportional gain \(P\) of the PI-controller.

The tunneling current \(I_{tc}\) can be expressed as \(I_{tc}(z, V_{\text{bias}}) \propto D_{\text{sample}}(E_F) D_{\text{tip}}(E_F + eV_{\text{bias}}) V_{\text{bias}} e^{-2\kappa z}\)

\[
(3.3)
\]

where the densities of states of the sample and the tip and the applied bias \(V_{\text{bias}}\) between tip and sample enter. The decay constant \(\kappa\) is related to the height of the tunnel barrier \(\Phi\) via \(\kappa = \sqrt{2m_e \Phi / \hbar^2}\). The quantity \(\beta\) is defined according to Eq. (B.1): It is the slope of the tunneling current for given \(V_{\text{bias}}\) and at height \(z\).

Therefore, there are three possibilities to increase the bandwidth \(f_{\text{STM}}\):

- increase of the proportional gain \(P\) (disadvantage: higher noise in \(z\)),
- increase \(V_{\text{bias}}\) which will lead to a larger \(\beta\) (disadvantage: lower resolution),
- increase \(I_{\text{set}}\) which will lead to a lower \(z\) and larger \(\beta\) (disadvantage: increased danger of tip crashes).

Typical values are \(V_{\text{bias}} \sim 10 \text{ meV} - 100 \text{ meV}\) and \(I_{\text{set}} \sim 1 \text{ nA}\).

### 3.3.2 Comparison with the Experiment

We measure the response function of the STM-feedback by applying an AC-voltage \(V_{\text{AC}}(f)\) to the tip so that \(V_{\text{bias}}(f) = V_{\text{DC}} + V_{\text{AC}}(f)\). Since the tunneling current is kept constant at the set point \(I_{\text{set}}\), this will induce oscillations in \(z\) with an amplitude \(A_z\), the phase \(\phi\), and the frequency \(f\). As long as \(f < f_{\text{STM}}\), the feedback is fast enough to keep the amplitude \(A_z\) constant. However, when \(f > f_{\text{STM}}\), the amplitude \(A_z\) will decrease. According to our considerations in the previous section, the relations

\[
A_z \propto |Z_{\text{STM}}(f)| = \left(1 + \frac{f^2}{f_{\text{STM}}^2}\right)^{-1/2},
\]

\[
\phi = \arctan \frac{\text{Im} Z_{\text{STM}}(f)}{\text{Re} Z_{\text{STM}}(f)} = - \arctan \frac{f}{f_{\text{STM}}}
\]
3.3. Scanning-Tunneling Microscopy

Figure 3.1: (a) Response functions of a typical IV-converter (OPA 111 with a subsequent INA 118) for three different feedback resistances $R_f$. The bandwidths which are extracted from a numerical fit to the experimental data are also indicated. Deviations of the experimental data points from the theoretical line above $\sim 2\, \text{kHz}$ are due to problems with the sine-generator of the NANONIS electronics.

(b) Response functions of the STM for three different values of the proportional gain $P$ and $R_f = 10\, \text{M}\Omega$. The solid lines correspond to fits to the experimental data; the extracted bandwidths are also indicated in the figure.

should hold. Fitting the experimental with the theoretical curve yields the bandwidth of the whole system.

In Fig. 3.1(a), we show the response function of a typical IV-converter used in our labs (OPA 111 with a subsequent INA 118) for three different feedback resistances $R_f = 10\, \text{M}\Omega$, $R_f = 100\, \text{M}\Omega$, and $R_f = 1\, \text{G}\Omega$. The numerical fit yields the bandwidth of the IV-converter.

In the following STM-measurements, we use a feedback resistance of $R_f = 10\, \text{M}\Omega$ resulting in a bandwidth $f_{IV} \approx 1900\, \text{Hz}$. According to our considerations above, we set $\tau_{PI} = \tau_{IV} = 1/2\pi f_{IV} \approx 100\, \mu\text{s}$ in the NANONIS software. We carried out the described procedure for three different proportional gains $P = 45\, \text{pm}$, $P = 150\, \text{pm}$, and $P = 450\, \text{pm}$ as shown in Fig. 3.1(b). The solid lines are fits to the experimental data from which we can extract the STM-bandwidths $f_{STM}$. The fit for $P = 45\, \text{pm}$ is very good whereas in the other cases the higher proportional gain leads to overshooting. This is not included in the theoretical model. However, the overall trend is still reasonably well described.

3.3.3 A Recipe for Adjusting the Feedback Parameters

It is crucial to set the feedback parameters in advance so that tip crashes can be avoided when approaching the surface. The following recipe is a summary of the
previous sections:

• tip-sample voltage: \( V_{\text{bias}} \sim 10 \text{ mV} - 100 \text{ mV} \),

• tunneling current setpoint: \( I_{\text{set}} \sim 1 \text{ nA} \),

• time constant of PI-controller: \( \tau_{\text{PI}} = \tau_{\text{IV}} \sim 0.1 \text{ ms} - 1 \text{ ms} \) (depending on feedback resistance),

• proportional gain of PI-controller: \( P \sim 100 \text{ pm} - 1 \text{ nm} \) (trade-off between low noise and large bandwidth).

### 3.3.4 Noise Considerations

As mentioned above, the scanning-gate setup presented in this chapter was not intended for STM. Albeit STM is possible as it will be shown below, two main problems will limit the performance, i.e. the resolution of the microscope. Firstly, the microscope is not sufficiently mechanically decoupled from the environment. Mechanical vibrations, e.g. due to people working close-by, closing of doors etc., show up as additional noise on the tunneling current. Secondly, the scan piezo is rather long for reasons discussed above. This inevitably leads to less lateral and vertical resolution. From the piezo specifications, we can estimate the minimal resolution. It is of the order of 1 nm laterally; atomic resolution in lateral direction is therefore not possible. For nanostructures, however, this is not necessary.

The lateral noise has not been determined experimentally yet because topographic scans have not been successful so far. However, the noise in \( z \)-direction could be estimated from experiments measuring the tunneling current as a function of tip-sample separation. We write the tip-sample separation as \( z = z_0 + \delta z \) with the “ideal” separation \( z_0 \) plus a noise term \( \delta z \). From Eq. (3.3), it follows that the tunneling current is then

\[
I_{\text{tc}}(z_0 + \delta z) = C e^{-2 \kappa (z_0 + \delta z)} = C e^{-2 \kappa z_0} e^{-2 \kappa \delta z}
\]

with some \( z \)-independent constant \( C \).

The noise in \( z \) yields a noise in the current which we define as \( \delta I_{\text{tc}} = I_{\text{tc}}(z_0 + \delta z) - I_{\text{tc}}(z_0) \). The normalized current noise reads

\[
\frac{\delta I_{\text{tc}}}{I_{\text{tc}}(z_0)} = e^{-2 \kappa \delta z} - 1.
\]

The dependence on the local density of states and the bias voltage vanishes because these parameters are included in the prefactor \( C \) which drops out. Solving this expression for \( \delta z \) yields the noise of the scan piezo,

\[
\delta z = -(2 \kappa)^{-1} \ln \left( \frac{\delta I_{\text{tc}}}{I_{\text{tc}}(z_0)} + 1 \right).
\]
According to these considerations, the \( z \)-noise is independent of the tip-sample separation \( z \) as it is expected to be. It can be deduced from an experiment where the tunneling current is measured as a function of tip-sample separation. Such a measurement for tunneling into a gold contact under ambient conditions is shown in Fig. 3.2(a). We fit the experimental data with an exponential curve. This allows us to extract the relevant parameters, essentially \( \kappa = 1.81 \times 10^9 \text{ m}^{-1} \) which corresponds to \( \Phi = 125 \text{ meV} \).

We can now calculate the normalized current noise in two ways: i) By taking the difference between the experimental data points and smoothed data points and ii) by taking the difference between the experimental data points and the fitted curve for every value of \( z_0 \). The result for both possibilities is shown in Fig. 3.2(b).\(^1\) In both cases, \( \delta I_{tc}/I_{tc} \) is of the order of 1 corresponding to \( \delta z \approx 2 \text{ Å} \). We emphasize that this is a very crude estimation where we assumed that the noise in the current is dominated by the effects of the noise of the scan piezo. Since this is not necessarily true, this estimation gives an upper bound for \( \delta z \).

\(^1\)The plot points for the fitted curve are only valid up to \( z_0 \approx -717.5 \text{ nm} \); this is due to the way the fit was done.
Chapter 4

Excited States in Graphene Quantum Dots

A quantum dot is a single-electron transistor which additionally shows quantum-confinement effects. Because of the special properties of graphene, it is desirable to look at spin physics, especially spin-dephasing times, in graphene quantum dots. There are several important steps along this way: demonstration of Coulomb blockade in single and double dots, observation of excited states, spin states, spin blockade, and fabrication of a single-charge read-out. While some of these steps have been realized, the realization of others is still ongoing research. For example, Coulomb blockade was already measured in single and double dots \[76\text{–}80\] and also single-charge read-out using a charge detector was successfully demonstrated \[74\]. On the other hand, the observation of spin blockade in double dots has not been successful for unknown reasons so far \[79\text{,} 80\]. Nevertheless, indications for spin states in single dots were found \[81\]. In this chapter, we report on the observation of excited states in a graphene quantum dot \[82\] – in some sense precursor measurements to the determination of spin states. Similar effects were also observed by other groups \[76\].

4.1 Coulomb Diamonds and Excited States

An atomic-force microscope image of the quantum dot under ambient conditions is shown in Fig. 4.1a. The quantum dot was fabricated with the standard procedure described in section 1.1.2. The single-layer quality was experimentally verified with Raman spectroscopy \[22\]. The quantum-dot device consists of two about 60 nm and 70 nm wide graphene constrictions separating source and drain contacts from the graphene island (diameter 140 nm). The island can be tuned by a nearby plunger gate (PG), whereas the overall Fermi level is adjusted with a highly doped
Figure 4.1: (a) Atomic-force micrograph of the quantum dot. It can be tuned by a nearby plunger gate (PG). The central island is connected to source (S) and drain (D) contacts by two constrictions. The scale bar is 200 nm. (b) A back-gate sweep shows a transport gap from roughly $V_{BG} = -8$ V to 8 V ($V_{bias} = 3$ mV). (c) Coulomb blockade measured with the back gate at an electronic temperature of 200 mK and a bias voltage of $V_{bias} = 16$ $\mu$V (same figure as Fig. 2.2(d)).

silicon back gate (BG). The sample was annealed for about 24 hours in vacuum at 400 K directly before cool down. The experiments were carried out in a dilution refrigerator at a base temperature of 40 mK. Measuring the current $I_{dot}$ through the quantum dot as a function of back gate voltage $V_{BG}$ allows us to identify a transport gap extending roughly from $V_{BG} = -8$ V to 8 V (Fig. 4.1(b)). Since the gap is centered around zero back-gate voltage, we have little doping of our graphene device. Characteristic peaks in the gap region were identified as Coulomb peaks (Fig. 4.1(c)). From their width, we extracted an upper bound for the electronic temperature of around 200 mK [50]. In the following measurements, we set the back-gate voltage to zero in order to tune the device close to the CNP.

Coulomb-diamond measurements as described in section 2.2, i.e. plots of the differential conductance $G_{dot} = dI_{dot}/dV_{bias}$ as a function of bias voltage $V_{bias}$ and plunger-gate voltage $V_{PG}$, are shown in Fig. 4.2. Within this plunger-gate voltage range, no charge rearrangements were observed and the sample was stable for more than two weeks. We extract typical charging energies of the order of...
Figure 4.2: (a) Differential conductance $G_{\text{dot}}$ as a function of source-drain voltage $V_{\text{bias}}$ and plunger-gate voltage $V_{\text{PG}}$. (b) The lower panel is a zoom of the enframed area in (a). An excited state is clearly visible (white arrow). A cut along the dashed line at $V_{\text{bias}} = -2.87$ mV is shown in the upper panel (here $G_{\text{dot}}$ is measured in units of $10^{-3} e^2/h$ and was smoothed over 4 points). (c) Stability diagram in a different plunger-gate regime. Several excited states are visible as shown by dashed lines. In the upper part of the right diamond, regions of higher conductance can be seen. This is interpreted as the onset of inelastic co-tunneling in a graphene quantum dot (see arrow). In all measurements, the back gate voltage was set to $V_{\text{BG}} = 0$ V and the electronic temperature was around 200 mK as deduced from the Coulomb-peak width.
Chapter 4. Excited States in Graphene Quantum Dots

$\Delta E_C \approx 10 \text{ meV}$. This agrees reasonably well with the energy estimated from a disk model according to Eq. (2.2), $\Delta E_C \approx 12 \text{ meV}$ \[1\]. A strong fluctuation of the size of the Coulomb diamonds over the plunger gate voltage range $-0.1 \text{ V} < V_{PG} < 1.2 \text{ V}$ (full data range not shown), corresponding to an energy range of around $100 \text{ meV}$ is observed, indicating the importance of quantum-confinement effects.

This is supported by the observation of excited states which appear in Fig. 4.2(a) as distinct lines of increased conductance running in parallel to the edge of the Coulomb diamonds. Figure 4.2(b) – showing a close up of Fig. 4.2(a) – allows to extract a single-particle level spacing of $\Delta \approx 1.6 \text{ meV}$ as marked by the white arrow. A line cut at $V_{bias} = 2.78 \text{ mV}$ (dashed line) presented in the upper panel of Fig. 4.2(b) shows the peak of the excited state at finite bias (arrow). The broadening of the peak significantly exceeds thermal broadening and might be due to the energy-dependent coupling of the excited state to the graphene leads.

Figure 4.2(c) shows two Coulomb diamonds at a lower plunger-gate voltage where more than one excited state is observed as a function of increasing energy, as shown by pairs of dashed lines. These excitations are found at energies of around $1.6 \text{ meV}$ and $3.3 \text{ meV}$ (black dashed lines) and $2.1 \text{ meV}$ and $4.2 \text{ meV}$ (white dashed lines), respectively. The observation of excitations at finite source-drain voltage finds support by the detection of inelastic co-tunneling onsets at lower bias. Inside the right Coulomb diamond, we distinguish between regions of suppressed and slightly elevated conductance separated by the dotted line. The edge of this conductance step is aligned with the first excited state outside the diamond at an energy of $1.6 \text{ meV}$ as highlighted by an arrow. This is a direct evidence for inelastic co-tunneling as we explained in section 2.2.

The number of charge carriers on the quantum dot can be estimated from Eq. (2.3). Combined with the measured excitation energy and the lithographic dimension of the dot ($d = 140 \text{ nm}$), we estimate the number of charge carriers on the dot to be of the order of 10. This estimation obviously changes if the dot area is larger, e. g. if the dot was not defined by the two constrictions. The number of charge carriers would then be even lower. However, magnetic-field sweeps corroborate our estimation of the dot size as pointed out below. The current was below measurement resolution at $V_{PG} < -0.5 \text{ V}$ so that smaller charge-carrier numbers and the potential electron-hole crossover could not be studied. The Coulomb diamonds shown in Fig. 4.2(c) scatter significantly more strongly in size than those presented in Fig. 4.2(a). This might be a consequence of the lower number of charge carriers on the quantum dot.

\[1\] We assumed the effective dielectric constant including vacuum and the SiO$_2$ to be $\varepsilon_{\text{eff}} = (\varepsilon_{\text{vac}} + \varepsilon_{\text{SiO}_2})/2 = 2.5$. 

4.2 Magnetic Field Dependence

We further explore the excitation spectrum by measuring energy shifts of nine consecutive Coulomb peaks as a function of a perpendicular magnetic field. The measurement outcome where electrostatic contributions to the energy were subtracted as explained below is shown in Fig. 4.3. The plunger-gate voltage was converted into the vertical energy axis according to $\epsilon = e\alpha_{PG}V_{PG}$ with the measured lever arm $\alpha_{PG} = 0.075$.

In the capacitance model, the ground-state energy of an $N$-particle quantum dot can be written as the sum of the single-particle energies $\epsilon_i(B)$ plus the electrostatic contribution to the charging energy $Ne^2/C_\Sigma$ plus additional, gate-dependent terms, cf. Eq. (2.1). The ground-state energy is tuned by the gate voltage $V_{PG}$. The experiment was done in the zero-bias regime; hence we measured the chemical potential of the $N$th Coulomb resonance as explained in section 2.1 [60]. Experimentally, the single-particle energy $\epsilon_N(B)$ of the $N$th Coulomb resonance is then determined by $\epsilon_N(B) = e\alpha_{PG}V_{PG}^{res}(N, B) + Ne^2/C_\Sigma + \text{const.}$ with $V_{PG}^{res}$ being the plunger-gate voltage on the resonance. The constant part and the electrostatic

Figure 4.3: Experimental energy spectrum of the quantum dot in a perpendicular magnetic field. The typical magnetic-field scale at which a significant change is expected is approximately given by one flux quantum $\Phi_0 = h/e$ per dot area, i.e. $4\Phi_0/\pi d^2 = 270$ mT and is indicated by the black arrow. Starting around $B = 4$ T, a regular pattern with characteristic linear slopes evolves (see dashed lines) which shows the transition from single-particle fluctuations to $B$-field dependence.
contribution are subtracted such that consecutive peaks touch each other (alternatingly shown as red triangles and blue circles, respectively). Characteristic lines (see dashed lines in Fig. 4.3) linear in $B$ with slopes of around $\pm 2.5 \text{meV/T}$ can be seen. This strong $B$-field dependence cannot be explained by the Zeeman effect which would result in a slope of $g\mu_B = 116 \mu\text{eV/T}$, assuming a $g$-factor of $g = 2$. For higher magnetic fields, the Landau-level degeneracy increases and fewer Landau levels are filled. Consequently, the energy spectrum is expected to evolve from single-level fluctuations into a regular pattern. This transition can be seen at around 4 T. Theoretical calculations are in qualitative agreement with our experimental data [56, 58, 60].

Theoretically, this kind of measurement can unambiguously identify the electron-hole crossover in graphene as described in section 2.3. This has not been possible with the sample studied here. However, more recent experiments found strong indications for the crossover employing this method [83].
Chapter 5

Scanning-Gate Microscopy on a Graphene Quantum Dot

As explained in chapter [3], we want to employ SGM to retrieve local information about nanostructures. For example, the precise knowledge of local aspects of various nanostructures could help to improve their performance by optimizing their design. Graphene is a particularly interesting material system for SGM because the direct accessibility of the electron gas at the surface should yield a better resolution than in more conventional 2DEGs where the electron gas is buried some tens of nanometers below the surface. Moreover, important questions regarding graphene nanostructures still wait for an answer which may be found employing local techniques. The long debate on the localization mechanism in graphene constrictions is the most prominent example [12–19, 84, 85].

This chapter presents scanning-gate measurements on a graphene quantum dot which is connected to source and drain via two constrictions. The sample is characterized using conventional transport experiments in the first section. Then, we turn our attention to the scanning-gate experiments. We can clearly identify the quantum dot by observing Coulomb resonances in real space as well as localized states in both constrictions. Additionally, our method allows us to give a quantitative estimate for the size of these localized states.

5.1 Sample Characteristics

The graphene quantum dot under investigation in this chapter has been fabricated according to the standard procedure described in section [1.1.2]. Raman spectroscopy confirmed that the flake consisted of a single atomic layer. Atomic-force micrographs of the quantum dot (QD) and nearby in-plane gates (left and right side gate, LG and RG, and plunger gate PG) after etching and removing the
Figure 5.1: (a) Atomic-force micrograph of the graphene sample after reactive ion etching obtained under ambient conditions. The quantum dot (QD) is connected to source and drain via two constrictions. The nearby nanoribbon can be used as a charge detector but it was not connected in the measurements presented here. (b) In-situ atomic-force micrograph of the sample after cooldown at $T \approx 2.6$ K. This image was taken after positioning the tip above the sample. The outline of the dot and nearby in-plane gates is shown with white lines. The scale bar in (a) denotes 500 nm and 1 µm in (b). (c) Back-gate trace taken at $T \approx 2.6$ K and $V_{\text{bias}} = 500 \mu$V. The CNP is shifted to $V_{\text{BG}} \approx 30$ V. (d) Charge stability diagram of the quantum dot. The charging energy is found to be $\Delta E_C = 3.5$ meV at $V_{\text{BG}} = 15$ V and $T = 90$ mK.
5.2 Scanning-Gate Measurements

5.2.1 Scanning-Gate Microscopy of the Quantum Dot

We performed scanning-gate measurements of the quantum dot in the hole regime at $V_{BG} = 12 \text{ V}$. A representative result is shown in Fig. 5.2. The scan frame has an area of $1.4 \times 1.4 \mu\text{m}^2$ and the outline of the quantum dot, as obtained from topographic images (see Fig. 5.1(b)), is shown with dashed, black lines. We observe three sets of concentric rings which are marked by arrows labelled (QD), (A), and (B). The set (QD) is caused by Coulomb resonances of the quantum dot as verified by the presence of Coulomb-blockade diamonds when sweeping the tip and bias voltages (not shown here) and we refer to them as Coulomb rings. The conductance $G_{dot}$ does not drop to zero between two Coulomb rings because the measurements were done at the edge of the transport gap in back-gate voltage where the coupling of dot states to source and drain is rather strong.

Most strikingly, we observe two additional sets of rings (A) and (B). In the following, we will refer to them as resonances A and B, respectively. In all scanning-gate images taken on this sample, resonances A and B are manifest as amplitude-modulations of the Coulomb resonances of the quantum dot. They are centered around points in the constrictions connecting the quantum dot to source and drain. Their presence allows us to locate regions of localized charge carriers in the constrictions. The interpretation of rings A and B as being due to localized states will be corroborated below. As there is only one apparent set of resonances per constriction, we conclude that there is only one localized state in each constriction at this value of the Fermi energy.

In Fig. 5.3 we present a series of four SGM images recorded over more than 24 hours with the same parameters as Fig. 5.2 and using the left side gate to tune the...
Chapter 5. SGM on a Graphene Quantum Dot

4.4 $G_{\text{dot}} (10^{-3} e^2/h)$ (QD) (A) (B) P Q

Figure 5.2: Scanning-gate image in the hole regime, $V_{BG} = 12\, \text{V}$. The tip voltage was $V_{\text{tip}} = 2\, \text{V}$, left-gate voltage $V_{LG} = 0.15\, \text{V}$, and the scan frame has a size of $1.4 \times 1.4\, \mu\text{m}^2$. A symmetric bias of $V_{\text{bias}} = 300\, \mu\text{V}$ was applied across source and drain and the tip was scanned at a constant height of $\Delta z \approx 120\, \text{nm}$ above the sample. Coulomb resonances of the quantum dot show up as concentric rings denoted by arrow (QD). The center of the Coulomb resonances are offset from the topographic center of the dot by ca. $240\, \text{nm}$. The reason for that is of no importance here, but will be discussed in the next chapter. The outline of the quantum dot and its connection to source and drain via the two constrictions, depicted with dashed, black lines, is corrected for the offset, assuming that the Coulomb resonances are centered in the quantum dot (see also Fig. 5.3). Most striking, however, is the appearance of two more sets of concentric rings which are highlighted by arrows (A) and (B) and which are centered around points in the constrictions. The black, dashed-dotted line between points P and Q denotes the line along which the linescan of Fig. 5.4 was taken. The scale bar denotes $500\, \text{nm}$. 
5.2. Scanning-Gate Measurements

Figure 5.3: Scanning-gate image for the same parameters as in Fig. 5.2 apart from the left-gate voltage which is varied as denoted in panels (a) to (d). The topographic position of the quantum dot extracted from Fig. 5.1(b) is indicated with black, dashed lines in panel (a). The black, solid lines in all four panels trace the same resonance. Panel (d) shows the same measurement as Fig. 5.2 and is presented again here for completeness of this series of SGM images. The scale bar in (a) is 500 nm.
sample. Firstly, the series demonstrates the stability of the sample since no charge
rearrangement or the like is visible in the measurements. Secondly, the tunability
of not only the quantum dot but also of the localized states can been seen as an
increase in diameter of a given resonance: The black, solid lines in the four panels
trace the same resonance for the upper localized state.

Panel (a) of Fig. 5.3 indicates the topographic position of the quantum dot with
black, dashed lines as it is extracted from the topographic image Fig. 5.1(b). The
offset between topographic and scanning-gate image is apparent if the positions of
the quantum dot given on the one hand by the topography and on the other hand
by the centers of the Coulomb rings are compared. We corrected Fig. 5.2 for this
offset of 240 nm. Related issues will be discussed in detail in chapter 6.

The quality of the sensor, i.e. the PtIr-tip, is remarkably good. We can dis-
tinguish more than 35 Coulomb resonances, much more than in any measurement
on GaAs-structures. The spatial resolution is better than the minimal separation
of two Coulomb resonances of about 20 nm. This is of the order of the Fermi
wavelength!

5.2.2 Linescans and the Tip-Induced Potential

In electronic transport, resonances are in general localized in space (in SGM) and
sharp in energy. In order to confirm this for resonances A and B, we took linescans
between points P and Q in Fig. 5.2 and changed the energy of the localized states
by stepping the left side-gate voltage \( V_{LG} \) from 5 V up to 10 V. The result is shown
in Fig. 5.4(a). We can identify quantum-dot resonances (dashed, white line and
resonances parallel to it), resonances A (dotted, white lines and resonances parallel
to them), and resonances B (dashed-dotted line and resonance parallel to it).

The tip-induced potential at any fixed location in the graphene plane is changed
when moving the tip from P to Q. We stay on a particular resonance by compen-
sating for this change at the location of the resonance with \( V_{LG} \). This leads to the
characteristic slopes of resonances A and B and the quantum-dot resonances. In
other words, the shape of the resonances A, B, and of the quantum dot represent
the tip-induced potential sensed at the three different locations of the localized
states. The left side gate is closest to the center of resonance A; resonance B is
furthest away. Therefore, resonances A are strongly tuned by the left gate, whereas
resonances B are only slightly affected. Quantum-dot resonances are in between.
Below we will use this effect to deduce the lever-arm ratio \( \alpha_{local}^{LG}/\alpha_{dot}^{LG} \) of the lever
arms of the left gate on the dot, \( \alpha_{dot}^{LG} \), and on resonance A, \( \alpha_{local}^{LG} \).

Horizontal cuts in Fig. 5.4(a), i.e. cuts for fixed \( V_{LG} \), show that all resonances
eventually shrink to a single point in space. This allows us to identify them with
states localized in space at this point. Vertical cuts, on the other hand, show that
ey are reasonably sharp in energy. However, we notice a strong variation in the
5.2. Scanning-Gate Measurements

Figure 5.4: (a) Linescans along the dashed-dotted line between points P and Q depicted in Fig. 5.2 while the left gate was stepped from 5 V to 10 V. The other parameters are the same as in Fig. 5.2. We can clearly distinguish between features of resonances A and B and the quantum dot. Two resonances of A are highlighted with white, dotted lines labeled (1) and (2), a quantum-dot resonance is highlighted with a white, dashed line, and a resonance of B is denoted with a white, dashed-dotted line. The upper, broad resonance (1) of A does not show any charging effect, whereas the lower, sharp one (2) is accompanied with avoided crossings of the Coulomb resonances. (b) Shifting resonance (1) of A along the x-axis and scaling it by 1.67 lead to the determination of the ratio of lever arms of the quantum and resonance A as explained in the text.
Figure 5.5: (a) Model for explaining the essential features induced by localized states in the constrictions. For simplicity, we consider just one localized state (upper red puddle) which is coupled to the source lead via the tunnel coupling $\Gamma_{\text{loc}}$. $C_{\text{dot-loc}}$ denotes the capacitive coupling between the localized state and the quantum dot. (b) Scanning-gate measurements at $T \approx 90 \text{mK}$ measured at the mixing chamber. The image was taken at $V_{\text{BG}} = 12 \text{V}$, $V_{\text{bias}} = 35 \mu\text{V}$, $V_{\text{tip}} = -100 \text{mV}$, and a tip height of $\Delta z = 40 - 45 \text{nm}$; the scan frame covers an area of $0.3 \times 0.3 \mu\text{m}^2$. Compared to Fig. 5.2, Coulomb resonances are much sharper due to the lower temperature. Although this measurement and Figs. 5.2 and 5.4 were taken at the same back-gate voltages, a direct comparison of them is difficult because of several charge rearrangements in between. The crossings of Coulomb resonances and resonances of B lead to a modulation of $G_{\text{dot}}$ and no kinks in the Coulomb resonance for arrow (1) and to kinks for arrow (2). The scale bar denotes 100 nm.

width of the resonances; e.g., resonance A(2) is much sharper than resonance A(1). A closer inspection of the sharper resonances of A reveals that they are accompanied with kinks in the quantum-dot Coulomb resonances.

5.2.3 Model and Interpretation

We propose the model shown in Fig. 5.5(a) which is capable of capturing the essential details of our observations. It consists of a quantum dot coupled to source and drain via two tunnel barriers with tunnel coupling $\Gamma_{\text{dot}}^{l,r}$. We introduce an additional localized state located in the constriction and coupled to the lead via a tunnel barrier with coupling $\Gamma_{\text{loc}}$. The localized state interacts with the quantum-dot states by tunneling through the barrier and by a mutual capacitive coupling via $C_{\text{dot-loc}}$. 
Fig. 5.5(b) shows a scanning-gate image taken at the temperature \( T \approx 90 \text{ mK} \). Coulomb resonances are now much sharper than in Figs. 5.2 and 5.4(a). Resonances B lead to a modulation of the dot conductance \( G_{\text{dot}} \) as highlighted by arrows (1) and (2). The capacitive coupling between resonance B and the quantum dot leads to the kinks pointed at by arrow (2). They are more easily identified compared to Fig. 5.4(a) because of the lower temperature.

Whenever such a kink occurs, resonance A or B is charged with an additional charge carrier. Thus, resonances originate from localized states. The capacitive coupling \( C_{\text{dot-loc}} \) shifts the electrochemical potential in the dot when the localized state is charged. Consequently, the Coulomb ring in the scanning-gate image is shifted as well. Charging effects are not observed for all crossings of quantum-dot resonances with resonances A or B. Kinks in the Coulomb resonances occur only for narrow resonances A and B; the broader ones do not show the signature of charge quantization as inspection of Figs. 5.4 and 5.5 show. The tunnel coupling strength \( \Gamma_{\text{loc}} \) must therefore depend strongly on the Fermi energy. Then, charging of a localized state with discrete charges occurs only if \( \Gamma_{\text{loc}} \) is below a certain threshold such that the condition \( G_{\text{loc}}(\Gamma_{\text{loc}}) < e^2/h \) for the conductance of the localized state is fulfilled. The width of the resonance is determined by the tunnel coupling if \( \Gamma_{\text{loc}} > 4k_BT \). The strong fluctuations in width of the resonances as observed in Fig. 5.4 are explained in that way.

Whenever a localized state shows quantum dot-like behavior, we can infer its size from its charging energy. In order to do so, we pick just those resonances of A in Fig. 5.4(a) which show avoided crossings with quantum-dot resonances. We identify six resonances of this type in the regime denoted by “quantum-dot behavior of resonances A”. They have a spacing of \( \Delta V_{\text{LG}}^{\text{loc}} \approx 270 \text{ mV} - 370 \text{ mV} \) in left-gate voltage. In order to convert this voltage scale to an energy, we need to know the lever arm \( \alpha_{\text{LG}}^{\text{loc}} \) of the left gate onto the localized state. The desired lever arm is obtained from Fig. 5.4 by comparing resonances of the quantum dot with resonances of the localized state.

Coulomb resonances of the quantum dot are separated on average by \( \Delta V_{\text{LG}}^{\text{dot}} = 53 \text{ mV} \) in left-gate voltage. Combined with the charging energy \( \Delta E_C = 3.5 \text{ meV} \), this yields a lever arm of \( \alpha_{\text{LG}}^{\text{dot}} = 0.066 \). The two dotted lines in Fig. 5.4(a) denote how resonances of A evolve as a function of position. If we scale these lines by a factor of 1.67 in \( V_{\text{LG}} \)-direction and shift them along the x-axis (the “position”) such that their minima coincide with a minimum of a Coulomb resonance, they nicely fit onto each other over a range of 1 \( \mu \text{m} \) as shown in Fig. 5.4(b). (For tip positions > 1.1 \( \mu \text{m} \), screening effects lead to a different progression of the quantum-dot resonance and the resonances of localized state A.) The desired lever arm \( \alpha_{\text{LG}}^{\text{loc}} \) is given by \( \alpha_{\text{LG}}^{\text{loc}} = \alpha_{\text{LG}}^{\text{dot}} \cdot 1.67 = 0.110 \).

Consequently, the charging energy \( \Delta E_C^{\text{loc}} = \alpha_{\text{LG}}^{\text{loc}} \cdot e \Delta V_{\text{LG}}^{\text{loc}} \) of the localized state A
is between 30 meV and 41 meV. This corresponds to a radius of the localized state
of $r_{\text{loc}} = 10 \text{nm} - 13 \text{nm}$, assuming $\Delta E_C^{\text{loc}} / \Delta E_C = r / r_{\text{loc}}$ with $\Delta E_C = 3.5 \text{meV}$ and
lithographic size of the dot $r = 110 \text{nm}$. Previous experiments obtained similar
sizes by means of conventional transport experiments for graphene nanoribbons
[13, 14, 17]. However, our method allows to determine relative lever arms of
localized states in more complex structures such as the presented quantum-dot
device.

In Ref. [15], a variation of relative lever arms of localized states in nanoribbons by
up to 30% is reported. This was explained with a number of localized states spread
along the nanoribbon. Based on the geometry of the device used in Ref. [15], a rough
estimate yields a typical separation between localized states of more than 300 nm.
With our SGM, a separation of more than 100 nm should be easily detectable. In
our measurements on the quantum dot in the hole regime, we typically observed
one localized state per constriction at $T = 2.6 \text{K}$ and for a range of Fermi energies.
In some rare cases, we found indications of more than one resonance in a single
constriction separated on a scale of $\sim 100 \text{nm}$ (see Fig. 5.6 below). We speculate
that the observation of one localized state per constriction at a given Fermi energy
and at $T = 2.6 \text{K}$ is due to the short length of the constriction.

In Fig. 5.2, we can distinguish more than 30 Coulomb rings with a spacing
of around 20 nm for some pairs of rings. This corresponds to an energy gradient
$\Delta \epsilon_{\text{res}} = \Delta E_C / 20 \text{nm} \approx 0.2 \text{meV/nm}$. We define the spatial resolution $\Delta x_{\text{res}}$ as the
minimal separation of two localized states which we can resolve with the scanning-
gate tip. It is limited by the width $\Gamma_{\text{res}}$ of the resonances. We estimate the spatial
resolution from $\Delta x_{\text{res}} = \Gamma_{\text{res}} / \Delta \epsilon_{\text{res}}$. The best resolution is obtained for thermally
broadened resonances, $\Gamma_{\text{res}} \approx 4k_B T$, yielding $\Delta x_{\text{res}} \approx 5 \text{nm}$ for $T = 2.6 \text{K}$. However,
if the resonance is tunnel-coupling broadened by e. g. a factor of ten compared
to temperature, the spatial resolution decreases to $\Delta x_{\text{res}} \approx 50 \text{nm}$. In that case,
the potential existence of two localized sites within the constriction could not be
resolved with our SGM.

Because of the small extent of localized states, confinement effects will be very
pronounced. This property makes them to interesting candidates for observing
confinement effects in SGM. For this reason, preliminary scanning-gate images of
a localized state were taken at the base temperature of the cryostat of $T \approx 85 \text{mK}$.
Since the cryostat could not be operated at this temperature sufficiently long,
precise tuning of the sample was not possible. In Fig. 5.6, we present a detailed
scanning-gate image of the localized state sitting in the drain constriction. The
center of the quantum dot is still visible in the upper left corner. The SGM image

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1This expression holds if the typical width of the wave function in a localized state is small
compared to the characteristic length over which the tip-induced potential rises. This is true
here, given our experimental results.
Figure 5.6: Scanning-gate image of a localized state. The conductance of the sample is given in color-code with the brightest color denoting a maximal conductance of $G = 6.7 \times 10^{-3} e^2/h$. The center of the quantum dot is still visible in the upper left corner. For this measurement, the cryostat was cooled down to a temperature of $T \approx 85$ mK and the tip-sample separation was reduced to $\Delta z \approx 65$ nm. The other scanning-gate and sample parameters were: $V_{\text{bias}} = 35 \, \mu$V, $V_{\text{BG}} = 12$ V, $V_{\text{LG}} = -1$ V, and $V_{\text{tip}} = -0.5$ V. The red arrows point to substructures in the resonances of the localized state. The scan area is $500 \times 500$ nm$^2$ and the scale bar denotes 200 nm.

of the localized state shows a fan-like substructure pointed at with red arrows which goes beyond Coulomb rings of a single localized state. It is rather unlikely that this substructure is due to confinement effects; a second localized state sitting nearby is a more plausible explanation. However, it demonstrates that localized states are interesting objects in their own right. No substructure in Coulomb resonances of the quantum dot have been observed at this temperature and for similar scanning parameters.
Chapter 6

Relevance of Electrostatics for Scanning-Gate Microscopy

Scanning-probe techniques have been developed to extract local information from a given physical system. Looking from a broader perspective, the impulse of always looking more closely and more precisely is probably one of the main driving forces for our exploratory urge. In that sense, particle accelerators like the CERN fall in the same category as e. g. scanning-tunneling and scanning-gate microscopes in condensed-matter physics. When designing such an instrument, one of the main question is “What resolution do we need for the physical system we want to look at?” Typical length scales in mesoscopic physics are the Fermi wave length (some tens of nanometers), the phase coherence length (greater than a micrometer in high-quality samples and at low temperature), or the extent of a certain structure (e. g. the diameter of a quantum dot which is typically of the order of a hundred nanometers). Our scanning-gate setup was designed to obtain resolutions at the lower end of these numbers. Indeed, in Fig. 5.2 we can distinguish Coulomb rings which are separated by about 20 nm in space. Hence, we could conclude that our instruments meets our design values and that we could directly deduce local properties from scanning-gate images.

This chapter will argue that the interpretation of scanning-gate images is a bit more involved and not that intuitive under certain circumstances. The first section will present the main complication: An apparent shift of features in scanning-gate images as a function of gate voltages which cannot be a real shift of the physical system. Additionally, we observe more than one set of Coulomb rings of the quantum dot. In the second section, different possible explanations for the problem will be discussed. The electrostatic influences of gate electrodes will be analyzed qualitatively in the third section. The analysis will then be compared to a scanning-gate measurement. In the final section, the results are compared to previous SGM-experiments on quantum-dot structures and discussed.
Chapter 6. Relevance of Electrostatics for SGM

Figure 6.1: (a) to (d) Four scanning-gate images taken in the hole regime for \( V_{BG} = 12 \text{ V}, \ V_{tip} = 2 \text{ V}, \ V_{bias} = 300 \mu \text{V}, \) and \( T \approx 2 \text{ K}. \) The left and right side gates were changed. This results in an apparent shift of the centers of the Coulomb rings. These are denoted by red crosses; in each image the centers of the previous images are also denoted. The overall shift from image (a) to (d) is \( \Delta = 110 \text{ nm}. \) (e) to (h) A similar series of measurements as in (a) to (d) for \( V_{BG} = 16 \text{ V}. \) Two sets of Coulomb rings are visible in this regime which sense the same quantum dot. By changing side-gate voltages, the centers of the two sets shift in opposite directions by in total \( \Delta_1 = 430 \text{ nm} \) for the lower and \( \Delta_{II} \geq 450 \text{ nm} \) for the upper set. The center of the Coulomb rings in (h) for set II cannot be determined. Hence, just a lower bound for the total shift can be given.

6.1 Unexpected Features of Coulomb Rings

Previous SGM experiments demonstrated that the interpretation of SGM images of quantum dots is not as straightforward as one might expect. For example in Ref. 70, the authors observed two sets of Coulomb rings. This was interpreted as a signature of a double tip. In Refs. 34 and 69 the tip-induced potential was found to consist of two contributions, namely one depending on the tip voltages and a second independent of the applied tip voltage. Here, we will present evidence that mutual screening effects of the gates and the tip can lead to the observation of more than one set of Coulomb rings. Furthermore, their centers show an apparent shift when gate voltages are changed.

We will first study further SGM images of the device introduced in the previous chapter. A closer look at a series of images where a specific gate is changed shows a shift of the centers of Coulomb rings in real space. Fig. 6.1 presents two series of
6.1. Unexpected Features of Coulomb Rings

Figure 6.2: Centers of Coulomb rings of the quantum dot and of the resonances of the localized states in the constrictions plotted in one graph and labeled according to electron or hole regime. The position (0,0) corresponds to the center of the scan range. The lithographic outline of the quantum dot and the adjacent gates is shown in dashed, black lines. Its position was shifted such that as many centers as possible lie within the quantum-dot boundary. The abrupt change in position between electron and hole states is discussed in the main text.
such measurements: (a) to (d) were taken at $V_{BG} = 12\,\text{V}$, (e) to (h) at $V_{BG} = 16\,\text{V}$. In both series, the left and right side-gate voltages were changed as denoted in the images. The eye-catching difference between the two series is that in (e) to (h) two sets of Coulomb rings are visible, whereas there is only one for (a) to (d). The origin of the two sets will be discussed below. The centers of the Coulomb rings are marked with yellow crosses (encircled for the upper set of Coulomb rings in (e) to (h)). We see that there is a shift of the centers of the Coulomb rings in real space which can be greater than 450 nm. This is more than the extent of the quantum dot in that particular direction.

For a further analysis, we determined the centers of Coulomb rings and resonances of localized states A and B for all scanning-gate images taken with this sample (if a determination was possible) in the same way as in Fig. 6.1. The locations of all the centers are plotted in Fig. 6.2 with their corresponding error bars. We distinguish both the different resonances (see chapter 5) and the electron or hole regime by using different symbols. The outline of the lithographic quantum dot is shown as a black, dotted line in order to depict a reference for the geometry of the structure. Some Coulomb rings, like those presented in Fig. 6.1(e) to (h), are strongly distorted such that their contours could not be fitted with a circle or an ellipse. Therefore, the position of the centers was estimated by eye. The error bars are given by the radius of the innermost Coulomb ring of a given set. Because of the strong suppression of the current in the transport gap (see Fig. 5.1(c)), no SGM images could be taken for $20\,\text{V} \leq V_{BG} \leq 35\,\text{V}$. The regime for lower back-gate voltages is classified as the hole regime, the one with higher back-gate voltages as the electron regime.

First of all, it seems surprising that there is a clear separation between centers in the electron and in the hole regime (open and filled symbols in Fig. 6.2). It might be that we would have observed a continuous transition of the centers if we could have taken SGM images at all back-gate voltages. However, it is more likely that the electron states were probed with a different set of Coulomb rings (the upper ones in Fig. 6.1(e) to (h)) than the hole states. During the measurements, it was not obvious that this happened because of the large transport gap and the possible disappearance of one set of Coulomb rings (as discussed below).

We know that we can shift the center of mass of a wave function in real space by applying different side-gate voltages. The shifts observed in Fig. 6.2, on the contrary, cannot be accounted for by a shift in real space alone because they are

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1We neglect the modulations of the amplitude of the Coulomb rings which are induced by the localized states in the constriction as discussed in the previous chapter. Here, we only focus on the Coulomb rings sensed by the quantum dot.

2If there were more than one set of Coulomb rings observable, the centers of the lower-left set (as in Fig. 6.1(e) to (h)) were determined since this is the set all previous measurements focused on.
6.2. Explanations for the Shift of Coulomb Rings

much larger than the structure itself – even if we just consider the centers in the hole regime.

In summary, we observe two effects, namely the appearance of more than one set of Coulomb rings and apparent shifts of their centers larger than the extent of our device. For a correct interpretation of scanning-gate images, especially when we want to extract length scales, it is therefore crucial to understand where these two effects come from and how we can possibly avoid them in future experiments.

6.2 Explanations for the Shift of Coulomb Rings

There are three possible explanations for the observed shifts of Coulomb-ring centers, namely (i) a drift of the sample holder with respect to the tip over time, (ii) bending of the tip because of electrostatic forces between the tip and the surrounding, and (iii) a change in the electrostatic fields and potentials shifting the electrochemical potential of the quantum dot.

In order to make sure that the quantum-dot structure is always at the same position, topographic scans of the sample were taken in certain intervals. A drift of the sample holder was not detectable and can therefore be excluded.

We give a rough estimate of the ratio of the electric force acting on the tip and the mechanical shear force necessary to lead to a deflection of the tip,

$$\frac{F_{\text{el}}}{F_{\text{mech}}} \approx \frac{d\left(\frac{1}{2}C(z)U^2\right)/dz}{G_{\text{Pt}}\gamma A} \bigg|_{z=100\,\text{nm}} \sim 10^{-3}.$$ 

We approximated the tip-sample capacitance $C(x)$ with a plate capacitor with area $A$ which will lead to a drastic overestimate of the energy and hence of the electric force. The tip-sample separation was set to $z = 100\,\text{nm}$ and the tip deflection to $400\,\text{nm}$ (about the largest shift observed in our measurements) for a rather long tip of $500\,\mu\text{m}$. This yields a deflection angle of $\gamma = 0.4\,\mu\text{m}/500\,\mu\text{m}$. The shear modulus for platinum which is a good approximation for our PtIr-tip is $G_{\text{Pt}} = 6 \times 10^{10}\,\text{Pa}$, and we assumed a rather large tip-sample voltage of $U = 10\,\text{V}$. The small ratio of $10^{-3}$ which is an upper bound clearly indicates that electrostatic forces cannot lead to a significant bending of the tip.

Hence, we are left with the third explanation, namely that the shift of Coulomb rings is due to a change in the electrostatic potentials when tuning gates. In fact, when we have a closer look at Fig. 6.1, we see that strong voltage differences between gates (especially in (c) and (h)) lead to a significant distortion of the Coulomb rings. This distortion cannot be explained with a drift of the sample holder or bending of the tip. It rather corroborates the importance of electrostatics for the interpretation of SGM images. The shifts will be discussed in more detail.
Figure 6.3: (a) Arrangement of the metallic electrodes (grey) and the quantum dot (black, dashed lines) as imaged in Fig. 5.1(b). (b) Qualitative behavior of the tip-dot capacitance $C_{\text{tip}}$ and the gate-dot capacitance $C_{\text{PG}}$ along the arrow in (a). For $x_{\text{tip}} \to \pm \infty$, $C_{\text{tip}}$ will go to zero, whereas $C_{\text{PG}}$ will approach a maximal value. $C_{\text{tip}}$ has a maximal value close to the dot. Due to screening by the tip, $C_{\text{PG}}$ is minimal somewhere between dot and gate position.

in the next section because they are intricately entangled with the appearance of multiple sets of Coulomb rings.

6.3 The Origin of More Than One Set of Coulomb Rings

As mentioned above, the appearance of two sets of Coulomb rings was previously assigned to a double tip \[70]. Double-tip behavior can show up in topographic images as double features since a topographic feature, e.g., an edge of a metallic contact, is imaged by each tip individually. The excellent quality of the topographic images (cf. Fig. 5.1(b)) indicates that we use a sharp, single tip.

In Fig. 6.3(a), we present the outline of the sample again: The quantum dot in black, dashed line and the metallic electrodes in grey. For the qualitative arguments in the following, we use a simplified geometry where we just consider the plunger gate in addition to the tip. According to Eq. (3.1), the electrochemical potential of the quantum dot along the arrow in Fig. 6.3(a) is then described by

$$\mu_N(x_{\text{tip}}) = \frac{e^2}{C_{\Sigma}(x_{\text{tip}})} \left( N - \frac{1}{2} \right) + e \frac{C_{\text{tip}}(x_{\text{tip}})}{C_{\Sigma}(x_{\text{tip}})} V_{\text{tip}} + e \frac{C_{\text{PG}}(x_{\text{tip}})}{C_{\Sigma}(x_{\text{PG}})} V_{\text{PG}} + \text{const.} \quad (6.1)$$

\[\text{Double dots also yield two sets of rings in SGM but they intersect at the triple points. This is not observed in our case – apart from the fact that we do not have a double-dot device anyway.}\]
in the constant-interaction model, where the tip-dot capacitance $C_{\text{tip}}(x_{\text{tip}})$, the gate-dot capacitance $C_{\text{PG}}(x_{\text{tip}})$, and the total capacitance

$$C_{\Sigma}(x_{\text{tip}}) = C_0 - C_{\text{tip}}(x_{\text{tip}}) - C_{\text{PG}}(x_{\text{tip}})$$

depend on the tip position $x_{\text{tip}}$. A constant background capacitance $C_0$ was incorporated into the total capacitance $C_{\Sigma}$. We neglect the single-particle energy $\epsilon_N$ since the quantum dot under investigation did not show any signatures of excited states.

We qualitatively plot $C_{\text{tip}}$ and $C_{\text{PG}}$ as a function of $x_{\text{tip}}$ in Fig. 6.3(b). For $x_{\text{tip}} \to \pm \infty$, $C_{\text{tip}} = 0$ and $C_{\text{PG}} = \text{const}$. $C_{\text{tip}}$ has a maximum somewhere at the dot position $x_{\text{dot}}$, whereas $C_{\text{PG}}$ has a minimum when the tip is located somewhere between the dot position $x_{\text{dot}}$ and the gate $x_{\text{PG}}$ due to screening by the tip.

Coulomb rings are centered at the extrema of $\mu_N(x_{\text{tip}})$ in an SGM image. We calculate the derivative $\partial \mu_N/\partial x_{\text{tip}}$ of Eq. (6.1) and set it to zero. The total capacitance $C_{\Sigma}$ depends only weakly on $x_{\text{tip}}$ because its value is mainly determined by the constant background capacitance $C_0$. Experimental findings [70] and our numerical model below confirm this approximation. Using $C_{\Sigma}' \approx 0$, we then find

$$0 = C_{\text{tip}}'(x_{\text{tip}}) V_{\text{tip}} + C_{\text{PG}}'(x_{\text{tip}}) V_{\text{PG}}.$$ 

This is an implicit equation for the value of $x_{\text{tip}}$ where the Coulomb rings are centered. The interpretation of this expression is straightforward: If the extrema of the two individual capacitances are sufficiently sharp, the equation can be fulfilled at two points for certain gate voltages and two sets of Coulomb rings will be observed. Their centers can be shifted by applying different voltages to the tip and the gate. In the last chapter, we observed an offset of 240 nm between the SGM image, see Fig. 5.2, and the topographic image. This offset can be explained with the above expression. However, also a difference in the electronic and the topographic tip is conceivable.

In the following, we want to extend this qualitative model to a more realistic one in two dimensions based on the real quantum-dot structure. The details of the model can be found in appendix [C]. The tip capacitance $C_{\text{tip}}$ is assumed to have a Lorentzian shape with an amplitude of 2.4 aF and a width of 600 nm deduced from experimental values. We consider three gates corresponding to the left, plunger, and right gate of the real device; Eq. (6.1) is extended accordingly. Their capacitances approach a constant value of 4 aF if the tip is placed far away but are screened by the tip for tip positions in the dot-gate region. The precise function is discussed in the appendix. The total capacitance $C_{\Sigma}$ also includes a constant background capacitance of 46 aF. These numbers are taken from experimental data as discussed in appendix [C].
Figure 6.4: Numerical simulations of the electrochemical potential of a quantum dot for a fixed number of charge carriers $N = 1$ (upper row) and the resulting Coulomb resonances as imaged by SGM (lower row). The quantum dot is located at the red circle; the red parabolas denote the rims along which the screening of the three gates by the tip is strongest, see appendix C. All images have an area of $2 \times 2 \mu m^2$. The gate voltages of the three gates are changed from (a) to (c) as denoted in the images. The energy range in the upper row comprises about 100 meV (brighter colors indicating higher energies) resulting in about 30 Coulomb rings in the lower row. The images clearly reflect the formation of two and more sets of Coulomb rings and how their centers can be shifted by applying different gate voltages.

In Fig. 6.4 we show the results of a numerical evaluation of the model for three different gate-voltage settings. In the upper row, the electrochemical potential $\mu_N$ of the quantum dot is plotted in color code for a fixed number of charge carriers. In the lower row, the contour lines of the electrochemical potential is plotted for varying $N$ but the same gate-voltage settings. This is in analogy to the SGM measurement. Our model clearly resembles the measurements presented in Fig. 6.1(e) to (h): We observe at least two sets of Coulomb rings, and their centers can be shifted in real space by applying different gate voltages. If we apply a higher voltage to a particular gate, the corresponding peak in $\mu_N$ induced by that gate becomes more pronounced and more Coulomb rings appear. This is also observed in both the measurements (e. g. in Fig. 6.1(e) around the left gate) and
Figure 6.5: Overlays of the gate-electrode arrangement with SGM images. (a) The total area is $7 \times 7 \mu m^2$, and the scanning parameters are $V_{BG} = 25 V$, $V_{tip} = -1 V$, $\Delta z = 200 n m$, and $V_{bias} = 500 \mu V$. All other gates are grounded. (b) shows a zoom into the central area of (a) (area $3 \times 3 \mu m^2$) for a different scanning-gate regime ($V_{PG} = -11.89 V$, other parameters as in (a)). A clear spatial correlation between gate electrodes and Coulomb rings is visible in both images; especially (b) demonstrates how Coulomb rings of the tip are “squeezed” into the electrode-free area.

We see that our model can explain our experimental findings; however it does not exclude the possibility of the existence of a double tip. If our argumentation is relevant for our measurements, then we should see a spatial correlation between the gate-electrode arrangement and the appearance of Coulomb rings. In the case that the different sets of Coulomb rings are created by a double or multiple tip, such a correlation would be accidental and is therefore very unlikely. For this reason, we present overlays of the arrangement of the gate electrodes with scanning-gate images in Fig. 6.5(a) and (b): It can be seen that the pattern of the Coulomb rings correlates with the geometry of the device. Furthermore the observed Coulomb rings are centered on lines connecting the quantum dot to certain gate electrodes. This is strong evidence that our explanation for the formation of multiple sets of Coulomb rings is relevant for our sample.
6.4 Previous SGM-Experiments on Quantum Dots, Discussion

To our knowledge, a strong shift of the centers of Coulomb rings as observed here has not been reported in the literature yet. If more than one set of rings appeared, this was attributed to a double tip [70]. The question remains whether previous results have to be re-interpreted in the light of our findings or whether our findings were irrelevant for samples previously investigated. In Ref. 80, the authors describe SGM of carbon nanotubes on a back-gated Si-substrate. In fact, the observed Coulomb rings are strongly distorted and resemble the symmetry of the device. This indicates that the gate-electrode arrangement also plays a significant role. The authors realized that but did not perform any further systematic studies.

Most SGM-experiments have been performed on GaAs quantum dots [34, 69, 70]. The interpretation given there for two sets of Coulomb rings, namely a double tip, seems reasonable since no obvious spatial correlation between SGM images and the topography is seen. The authors do not report any significant shifts, either. In their structures, fabricated using AFM-oxidation techniques, gate voltages do not exceed a few hundred mV’s – values which can be more than a factor of ten higher in graphene nanostructures. Consequently, any shifts observed in GaAs will not be as pronounced as in graphene. Moreover, in case of GaAs-dots, the in-plane gates are formed by the 2DEG. Metallic electrodes contacting the 2DEG are tens of micrometers away from the quantum dot. The gallium capping layer with a dielectric constant $\varepsilon_{\text{Ga}} \approx 12$ may reduce the relevance of mutual screening effects between the tip and the in-plane gates.

SGM was also performed on a superconducting SET [35]. This structure is interesting to compare to graphene quantum dots because in both cases the sample is fabricated on a SiO$_2$-substrate and the metallic gates are very close to the dot and the SET. The SGM images of the SET show a pronounced distortion along the axis connecting the source and drain contacts. This may indicate that mutual screening effects of the tip and the gate electrodes is important for this sample as well. The authors, on the other hand, ascribe the distortion to an asymmetric tip shape. However, the main findings reported in the paper are independent of the correct interpretation of the distortion.

A fully tunable graphene quantum dot consists of at least five electrodes – source and drain, two gates for tuning the tunnel barriers, and a plunger gate – plus a back gate. The effects described in this chapter are intrinsic to a graphene sample and cannot be avoided using more advanced technology. Therefore, it is important to take these effects into account whenever local properties shall be deduced from a SGM image. In principle, it is possible to measure the influence of each individual gate as it is described in Ref. 69 for the tip. Since the contributions
to the electrochemical potential are additive, each contribution can be subtracted once determined by measurement. However, this is a very tedious task. Additional sets of Coulomb rings and strong shifts of their centers are therefore a phenomenon which probably has to be dealt with again in future SGM on graphene devices.
Chapter 6. Relevance of Electrostatics for SGM
Chapter 7

Conclusions

7.1 Summary

Graphene was discovered in 2004; research on graphene nanostructures like nanoribbons and in particular quantum dots started about two years later. Although quantum-dot physics is an interesting research topic in its own right, research on quantum dots roughly followed the path pointed out by D. Loss and D. DiVincenzo in 1998 [23]. This is also true for the research on graphene quantum dots. This thesis started around the time of the first successful demonstration of Coulomb blockade in graphene quantum dots.

The confinement of charge carriers leads to excited states with a certain energy spacing due to their wave nature. These excited states were demonstrated unambiguously for graphene quantum dots, chapter 4, which was the next obvious step after the demonstration of Coulomb blockade. The identification of spin states turned out to be infeasible for this particular sample but a transition from confinement-dominated to magnetic-field dominated behavior was observed. An analytic description reproducing the aspects of this transition qualitatively could be derived using the infinite-mass boundary condition in section 2.3.

A second graphene quantum dot was investigated locally in chapter 5 using SGM. The images proved to be of excellent quality showing more than 40 Coulomb rings in one scanning-gate image. Employing the scanning-gate technique, we verified directly the existence of one localized state per constriction at a particular Fermi energy. The localized states showed quantum-dot behavior depending on the transparency of their tunnel barriers. From their charging energy, we extracted a radius of about $10 - 13$ nm.

Scanning-gate images pretend to give a very intuitive picture of where localized states sit, namely at the center of the Coulomb rings. In graphene nanostructures, where electrodes are very close to the quantum dot, mutual screening effects of the
Chapter 7. Conclusions

Tip and the gate electrodes may give rise to spurious additional Coulomb rings. Indications for these effects are presented in chapter 6. Moreover, by tuning the gates, Coulomb rings can be strongly distorted and shifted. Both effects must be carefully taken into account when scanning-gate data of quantum dots are interpreted.

As pointed out before, the combination of scanning-gate imaging with scanning-tunneling microscopy would be a particularly powerful tool for graphene research. The STM-feedback comprises just one feedback loop, namely the $z$-controller, whereas in AFM-mode a PLL and the $z$-controller regulate the driving frequency of the tuning fork and the $z$-deflection of the piezo simultaneously. The derivation of the corresponding feedback formulas for an STM is described in section 3.3. Preliminary current-distance curves and response functions are in good agreement with the theoretical considerations.

Every experimental work is accompanied with – in the best case minor – repair work. Apart from standard repairs as soldering wires, exchanging tuning-fork sensors etc., a severe contamination of the mixing chamber and the silver heat exchangers with oil accompanied with a leak at the heat exchangers occurred at the dilution refrigerator. The repair at the manufacturer OXFORD INSTRUMENTS and amendments on the setup drag on over seven months. Some of the cryostat tests and the operating mode of a newly implemented pneumatic security valve are described in appendix D.

Potential Technical Improvements

Albeit the setup has been optimized for SGM, there are always possibilities for improvements. At the end, the quality of scanning-gate data is given by the quality of the tip\footnote{We neglect the sample here since it is not part of the setup.}. A technical change yielding better tips is therefore desirable. This can be achieved by either reducing the amount of topographic scans with the danger of tip damages, of picking up dirt etc. before performing SGM measurements or by improving the sensor design directly.

Lateral coarse positioning of the sample with respect to the tip is achieved with home-built slip-stick motors. The motion in $X$- and $Y$-direction is not decoupled from each other and can lead to unwanted sideward motions. Additionally, there is no positioning sensor for the $X$-$Y$-motor. Step motors decoupling $X$- and $Y$-direction and equipped with a positioning sensor are therefore highly desirable for fast and precise positioning. Slip-stick motors meeting these requirements are available from ATTOCUBE SYSTEMS. Their implementation should be possible with moderate modifications to the setup.

Regarding the sensor, in particular the tip, the un reproducible etching process
yields tips with diameters of around 30 nm in the best case, but probably an order
of magnitude higher in most cases. Focused-ion beam can be used to sharpen
PtIr-tips to diameters below 10 nm [87]. Alternatively, one could think of growing
a metallic nanowire on top of an existing PtIr-tip or directly on the upper prong
of the tuning fork. A completely different sensor design might also be worth to be
considered: Piezoresistive sensors have the advantage to be conceptually simpler
than tuning-fork sensor. Moreover, the use of silicon-fabrication technology can in
principle lead to mass production of sensors and reproducible tips with diameters
of around 10 nm [88].

In the case of using the setup as a scanning-tunneling microscope, the noise
of the scan piezo will probably be the limiting factor for lateral resolution (in
combination with mechanical vibrations coupling into the setup). Hence, a smaller
scan piezo would help at the expense of a smaller scan range. At least, the noise
of the NANONIS hardware can be further reduced by replacing the existing voltage
dividers (3.5 : 1) at the $X$, $Y$, and $Z$-output with dividers with a higher division
rate.

7.2 Outlook

The as-is state of the dilution refrigerator AFM turned out to be highly suitable
for local-probe experiments on graphene nanostructures. Spatial resolutions of
significantly below 20 nm demonstrate that impressively. In fact, due to the tech-
nical problems with the cryostat mentioned above (see also appendix D), the setup
could not exhaust its potential since the base temperature could not be kept stable
below 100 mK. Nevertheless, the scanning-gate images of the graphene quantum
dot are of excellent quality, much better than of any other material system. The
question one should keep in the back of one’s mind is whether this happened “just
by chance” or whether it is something “intrinsic” to graphene that makes graphene
particularly suited for SGM. Only further scanning-gate measurements can answer
this question.

There are at least two obvious follow-up experiments:

- So far, it turned out to be impossible to image the discrete energy spectrum
  of confinement-induced effects using SGM. Very small quantum dots and
  consequently very sharp tips are necessary for this purpose. Puddles, which
  show quantum-dot behavior under suitable conditions, might be interesting
  objects to study for that reason. Our measurements showed that we achieved
  an energy resolution of $\sim 3.5 \text{meV}/20 \text{nm}$ (charging energy over minimal
  Coulomb ring spacing), cf. Fig. 5.2. This can easily be increased further by
  reducing the tip-sample separation to values of $\sim 10 \text{nm}$ and going to lower
temperatures yielding sharper Coulomb rings. Nevertheless, the possibility of wave-function mapping seems to be unlikely with the present sensors.

- We could image one localized puddle in constrictions. The precise location, namely whether it sits closer to the edge or more in the bulk, could not be determined. The knowledge of this would provide new insights into the localization mechanism and hence into the appearance of a transport gap in graphene nanoribbons. A scanning-gate measurement on graphene nanoribbons might shed new light on this problem.

Albeit not motivated by the experiments presented in this thesis, there are prospective experiments which seem to be a logical continuation of previous work:

- SGM was recently successfully applied to study GaAs-double dots [89]. On the other hand, graphene double dots were also fabricated recently [79, 80]. It is therefore desirable to look at the questions raised in Ref. 89 from a different perspective, namely using graphene as a host material.

- Graphene is most famous for its relativistic charge carriers giving rise to the so-called unconventional quantum Hall effect and Berry’s phase of $\pi$. What happens in the quantum Hall regime of graphene when the tip is used as a scatterer? Is it possible to image edge-channel transport?

All these prospective experiments can be carried out with the setup as it is. The use of new scanning techniques like scanning-capacitance or – more demanding – scanning-tunneling spectroscopy does not require any changes at the setup but they would open up many new possibilities for research on graphene nanostructures.

A scanning-probe setup like the one described here consists of three main components: The sample, the cryostat, and the AFM. Compared to a conventional transport setup, the enhanced complexity (the AFM) increases the challenges of the experiment significantly. On the other hand, the potential our setup proved to have is so big that it is worth to undertake these challenges and to pursue the direction of this thesis.
Appendix A

Derivation of the Energy Spectrum

We want to solve the Dirac equation $H \psi(r, \phi) = E \psi(r, \phi)$ with the Hamiltonian given by Eq. (2.4). Since the operator for the total angular momentum, $J_z = l_z + \frac{1}{2} \sigma_z$ commutes with $H$, $[H, J_z] = 0$, we can construct simultaneous eigenspinors for $H$ and $J_z$ ($m$ being an integer),

$$\psi(r, \phi) = e^{im\phi} \left( \begin{array}{c} \chi_1(r) \\ e^{i\phi} \chi_2(r) \end{array} \right).$$

Plugging this expression into the Dirac equation and decoupling the system of differential equations, we arrive at a second-order differential equation for, e.g., $\chi_1(r)$ which depends only on $r$,

$$\left[ \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} - \frac{m+1}{l_B^2} - \frac{m^2}{r^2} - \frac{r^2}{4l_B^4} + k^2 \right] \chi_1(r) = 0.$$

The energy $E$ is related to the wave vector $k$ according to $E = \hbar c \sqrt{k}$. We have introduced the magnetic length $l_B = \sqrt{\hbar/(eB)}$. In order to solve this differential equation, we make the ansatz $\chi_1(r) = r^m \exp\left[-r^2/4l_B^2\right] \xi(r^2)$. This yields the associated Laguerre differential equation

$$\left[ \tilde{r} \frac{\partial^2}{\partial \tilde{r}^2} + \left(m + 1 - \frac{\tilde{r}}{2l_B^2}\right) \frac{\partial}{\partial \tilde{r}} + \frac{k^2 l_B^2}{4l_B^4} - 2(m + 1) \right] \xi(\tilde{r}) = 0 \quad (A.1)$$

with $\tilde{r} := r^2$. The solution is $\xi(\tilde{r}) = c L\left(\frac{k^2 l_B^2}{4l_B^4} - (m + 1), m, \frac{\tilde{r}}{2l_B^2}\right)$, where $L(a, b, x)$ is the generalized Laguerre polynomial and $c$ is a normalization constant. The second linearly independent solution of Eq. (A.1), the confluent hypergeometric function of the second kind, does not appear in the wave function $\xi$ because it
is not analytic at the origin. With the final result for $\xi$ and hence for $\psi_1$, $\psi_2$ can be derived directly from Eq. (2.4). The wave functions finally read

$$
\psi_1(r, \phi) = c e^{im\phi} r^m e^{-r^2/4l_B^2} L \left( \frac{k^2 l_B^2}{2} - (m + 1), m, r^2/2l_B^2 \right),
$$

$$
\psi_2(r, \phi) = c i e^{i(m+1)\phi} r^m e^{-r^2/4l_B^2} \frac{r/l_B}{kl_B} \times \left[ L \left( \frac{k^2 l_B^2}{2} - (m + 2), m + 1, r^2/2l_B^2 \right) + L \left( \frac{k^2 l_B^2}{2} - (m + 1), m, r^2/2l_B^2 \right) \right].
$$

The wave function combined with Eq. (2.5) yields the energy spectrum of a graphene quantum dot in a magnetic field, Eq. (2.6).

It is instructive to look at the limits $B \to 0$ and $R/l_B \to \infty$ of the energy spectrum, Eq. (2.6). For the first case, we can exploit that Bessel functions of the first kind are limits of the generalized Laguerre polynomial \[^90\],

$$
\lim_{a \to \infty} \left[ \frac{1}{a^b} L \left( a, b, \frac{x}{a} \right) \right] = x^{-b/2} J_b \left( 2\sqrt{x} \right),
$$

because $l_B \to \infty$ in this case. The spectrum then simplifies considerably and is given by Eq. (2.7).

Landau levels should be retrieved from Eq. (2.6) if the confinement is lifted. Mathematically, this is achieved for $R/l_B \to \infty$. We express the generalized Laguerre polynomial in terms of the confluent hypergeometric function of the first kind $M(\alpha, \beta, \gamma) \[^90\],

$$
L(a, b, x) = \binom{a + b}{a} M(-a, b + 1, x).
$$

For $R/l_B \to \infty$, a power expansion of $M$ is possible and yields to first order \[^90\]

$$
M(\alpha, \beta, \gamma) = \frac{\Gamma(\beta)}{\Gamma(\alpha)} e^\gamma \gamma^{\alpha-\beta} \left( 1 + O \left( |\gamma|^{-1} \right) \right).
$$

Rewriting the binomial coefficients with Gamma functions $\Gamma(x)$ and using one of their defining relations, $\Gamma(x + 1) = x \Gamma(x)$, algebraic manipulations of Eq. (2.6) give the expected result, namely the Landau levels of Eq. (2.8).
Appendix B

Derivation of the STM Feedback Formula

The IV-converter and the PIC have a unique frequency response which is low-pass behavior for the former and the Fourier-transform of the expression $\text{PIC}(V_{in}) = PV_{in}(t) + I \int_0^t V_{in}(\tau)d\tau$ for the latter. Hence, we get

$$\text{IV}(f) = \frac{1}{1 + i \frac{f}{f_{IV}}},$$

$$\text{PIC}(f) = P \left( 1 - i \frac{I}{P\omega} \right) = P \left( 1 - i \frac{1}{\omega\tau_{PI}} \right) = P \left( 1 - i \frac{f_{PI}}{f} \right).$$

The bandwidth of the IV-converter is $f_{IV}$. The PIC is tuned by the proportional gain $P$ and the integral gain $I$. The time constant $\tau_{PI} = P/I$ is the ratio of these two numbers and is inversely proportional to the bandwidth of the PIC $f_{PI} = 1/(2\pi\tau_{PI})$. These parameters can be adjusted manually in the NANONIS software.

The tunneling current $I_{tc}$ is a function of two externally controlled parameters, namely the tip-sample separation $z$ and the tip-sample voltage $V_{bias}$. In linear response, we expand the current up to first order for small deviations $\Delta z = z - z_0$ and $\Delta V = V_{bias} - V_0$:

$$I_{tc}(z_0 + \Delta z, V_0 + \Delta V) \approx I(z_0, V_0) + \frac{\partial}{\partial z} I(z, V_0) \bigg|_{z=z_0} \Delta z + \frac{\partial}{\partial V} I(z_0, V) \bigg|_{V=V_0} \Delta V. \quad (B.1)$$

A change in $\Delta z$ is due to a change in the high voltage applied to the scan piezo, $\Delta z = \mu \Delta V_{HV}$ where $\mu$ is the proportionality coefficient of the piezo and $\Delta V_{HV}$ is basically the output of the PIC. The input for the PIC is the error signal
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\[ e = I_{\text{set}} - IV(f)I_{tc} \] which is the difference between the set point of the tunneling current and the actual current. Combining all expressions, we retrieve

\[
\Delta z = \mu \cdot \Delta V_{\text{HV}} = \mu \cdot \text{PIC}(f) \cdot e = \mu \cdot \text{PIC}(f) \cdot (I_{\text{set}} - IV(f) \cdot I_{tc})
\]
\[
= \mu \cdot \text{PIC}(f) \cdot (I_{\text{set}} - IV(f) \cdot I_{tc}(z_0, V_0) - IV(f) \cdot \beta \Delta z - IV(f) \cdot \alpha \Delta V)
\]

\[ \Rightarrow \Delta z = \frac{\mu \cdot \text{PIC}(f)}{1 + \mu \beta \cdot IV(f) \cdot \text{PIC}(f)} (I_{\text{set}} - IV(f) \cdot I_{tc}(z_0, V_0) - \alpha \cdot IV(f) \cdot \Delta V). \]

We can measure the response of the system for example by applying an AC tip-sample voltage. The change in tip-sample separation is then effectively given by

\[
\Delta z = -Z_{\text{STM}}(f) \cdot \frac{\alpha}{\beta} \Delta V
\]
with the response function of our STM-feedback

\[
Z_{\text{STM}}(f) = \frac{\mu \beta \cdot \text{PIC}(f) \cdot IV(f)}{1 + \mu \beta \cdot \text{PIC}(f) \cdot IV(f)}.
\]

We want our STM-feedback to show low-pass behavior to maximize its bandwidth. Therefore, we match the bandwidth of the PIC and the IV-converter, \( f_{\text{PI}} = f_{\text{IV}} \). The response function then simplifies significantly to the expression given in Eq. (3.2) together with the corresponding bandwidth.

\[1^\text{This is not exactly true. Because of the IV-converter, the error signal is in fact a voltage. Since the voltage is converted into a current in the displays of the \text{Nanonis} software, we also stick to this convention.}\]
Appendix C

Electrostatic Model of a Quantum Dot and a Tip

In the following, we will describe the electrostatic model which is used to obtain the numerical results presented in chapter 6. The tip capacitance is given by a Lorentzian function

\[ C_{\text{tip}} (r_{\text{tip}}) = -2.4 \ \text{aF} \times \frac{(600 \ \text{nm})^2}{(r_{\text{tip}} - r_0)^2 + (600 \ \text{nm})^2}, \]

where the amplitude and the width is deduced from the measurement shown in Fig. C.1. We use an offset of \( r_0 = -(300 \ \text{nm}, 300 \ \text{nm}) \) in the simulation; this is the position of the quantum dot in the experimental scan frame. The left-gate capacitance looks

\[ C_{\text{LG}} (r_{\text{tip}}) = -4 \ \text{aF} - 2.4 \ \text{aF} \times \exp \left( \frac{(x_{\text{tip}} - x_{0,\text{LG}})^2}{(400 \ \text{nm})^2} \right) \times \frac{(500 \ \text{nm})^2}{((y_{\text{tip}} - y_{0,\text{LG}}) - (x_{\text{tip}} - x_{0,\text{LG}})^2 / 700)^2 + (500 \ \text{nm})^2}. \]

The first part denotes the constant capacitance for \( |r_{\text{tip}}| \to \infty \). The second part is the phenomenological screening term which has an amplitude of 2.4 aF, a lateral decay of the screening effect given by the exponential term with a decay constant of 400 nm, and a Lorentz-like screening term where the rim of the Lorentzian proceeds along a parabola with a coefficient of 1/700. The parameters \( x_{0,\text{LG}} \) and \( y_{0,\text{LG}} \) are used to position the gate in space. The plunger and the right gate are defined correspondingly with different parameters to position them. They are turned by 45° and 90°, respectively, in order to mimic the real device geometry. The total capacitance is then \( C_{\Sigma} (r_{\text{tip}}) = 46 \ \text{aF} - C_{\text{tip}} (r_{\text{tip}}) - C_{\text{LG}} (r_{\text{tip}}) - C_{\text{PG}} (r_{\text{tip}}) - C_{\text{RG}} (r_{\text{tip}}). \)
Appendix C. Electrostatic Model of a Quantum Dot and a Tip

Figure C.1: (a) Sweep of the tip voltage $V_{\text{tip}}$ as a function of tip position $x_{\text{tip}}$ along a line through the quantum dot. From the spacing of the Coulomb resonances along the $V_{\text{tip}}$-axis, the tip capacitance $C_{\text{tip}} = e/\Delta V_{\text{tip}}$ as a function of tip position can be deduced, where $\Delta V_{\text{tip}}$ is the (average) spacing of two Coulomb resonances. In graph (b), the tip capacitance, as measured in (a), is plotted over tip position (blue markers). The line is a fit using a Lorentzian function giving $C_{\text{tip}}(x_{\text{tip}}) = 2.3\, \text{aF} \times (613\, \text{nm})^2/((x_{\text{tip}} - 651\, \text{nm})^2 + (613\, \text{nm})^2)$.

The constant background capacitance accounts for the measured charging energy of the order of 3 meV which is observed even in the absence of the tip. The electrochemical potential is then given by Eq. (6.1) with the extension to include the three gates.

With these expressions at hand, it is now possible to calculate the electrostatic influence in our sample numerically and compare them to our experimental findings. In Fig. C.2, we present the left-gate capacitance $C_{\text{LG}}$ in (a), the resulting left-gate lever arm $\alpha_{\text{LG}}$ in (b), the total capacitance $C_{\Sigma}$ in (c), and the corresponding charging energy $\Delta E_{\text{C}}$ in (d). All quantities are plotted for tip positions within an area of $2 \times 2\, \mu\text{m}^2$. The calculated values are in good agreement with experimental findings; in particular the spatial fluctuations of the charging energy of about 10% of its absolute value are confirmed by earlier experiments [70]. This also justifies the approximation $\nabla_{\text{r}_{\text{tip}}} C_{\Sigma}(\text{r}_{\text{tip}}) \approx 0$ in chapter 6 for the qualitative understanding. Calculations of the electrochemical potential for a fixed number of charge carriers $N$ and corresponding Coulomb rings are shown in chapter 6. The outline of the geometry is also depicted there.
Figure C.2: (a) Left-gate capacitance $C_{LG}$ as a function of tip position as it follows from the above expression. The resulting lever arm $\alpha_{LG}$ is presented in (b). It has a maximum value of below 8%. (c) Total capacitance $C_{\Sigma}$ as a function of tip position. As expected, the spatial fluctuations are small compared to the absolute value of the capacitance, namely about 10%. The corresponding charging energy $\Delta E_C = e^2/C_{\Sigma}$ is shown in (d). Again, the fluctuations are about 10% of the absolute value. These values are in good agreement with experimental findings [70]. All images were calculated for an area of $2 \times 2 \mu m^2$. 
The Script for the Numerical Evaluation

This is a quote from the main part of the MATHEMATICA-script used to create the numerical results presented here and in chapter 6. All capacitances are given in aF, all length scales in nm, all voltages in mV, and all energies in meV.

\[ \sigma = 400; \]
\[ \Gamma = 500; \]
\[ \text{Ctip}[x_, y_] := -2.4 \times \frac{600^2}{(x+300)^2 + (y+300)^2 + 600^2}; \]
\[ \text{Crg}[x_, y_] := - \left( 4 - \text{Exp}[-(y + 100)^2/\sigma^2] \times 2.4 \times \frac{r^2}{((y+100)^2+200r+200r^2+\Gamma^2)} \right); \]
\[ \text{Clg}[x_, y_] := - \left( 4 - \text{Exp}[-(x + 100)^2/\sigma^2] \times 2.4 \times \frac{r^2}{((y-200) - (x+100)^2+200r+200r^2+\Gamma^2)} \right); \]
\[ \text{Cpg}[x_, y_] := - \left( 4 - \text{Exp}[-(y - x)^2/\sigma^2] \times 2.4 \right); \]
\[ \text{Which}[y > 0\&\& x > 0, \Gamma^2 / ((\text{Sqrt}[y^2 + x^2] \times \text{Cos}[\text{ArcTan}[y/x] - \pi/4] \times -\text{Sqrt}[y^2 + x^2] \times \text{Sin}[\text{ArcTan}[y/x] - \pi/4] / 2/700)^2 + \Gamma^2), \]
\[ y > 0\&\& x < 0, \Gamma^2 / ((\text{Sqrt}[y^2 + x^2] \times \text{Cos}[\text{ArcTan}[y/x] - \pi/4 + \pi] \times -\text{Sqrt}[y^2 + x^2] \times \text{Sin}[\text{ArcTan}[y/x] - \pi/4 + \pi] / 2/700)^2 + \Gamma^2), \]
\[ y < 0\&\& x < 0, \Gamma^2 / ((\text{Sqrt}[y^2 + x^2] \times \text{Cos}[\text{ArcTan}[y/x] - \pi/4 + \pi] \times -\text{Sqrt}[y^2 + x^2] \times \text{Sin}[\text{ArcTan}[y/x] - \pi/4 + \pi] / 2/700)^2 + \Gamma^2), \]
\[ y < 0\&\& x > 0, \Gamma^2 / ((\text{Sqrt}[y^2 + x^2] \times \text{Cos}[\text{ArcTan}[y/x] - \pi/4] \times -\text{Abs}[\text{Sin}[\text{ArcTan}[y/x] - \pi/4]] / 2/700)^2 + \Gamma^2)); \]
\[ \text{CΣ}[x_, y_] := 46 - \text{Ctip}[x, y] - \text{Clg}[x, y] - \text{Crg}[x, y] - \text{Cpg}[x, y]; \]
\[ \mu[x_, y_, n_, \text{Vtip}_-, \text{Vlg}_-, \text{Vrg}_-, \text{Vpg}_-] := \frac{160}{\text{CΣ}[x, y]}(n - 1/2) + \frac{\text{Ctip}[x, y]}{\text{CΣ}[x, y]} \times \text{Vtip} + \frac{\text{Clg}[x, y]}{\text{CΣ}[x, y]} \times \text{Vlg} + \frac{\text{Crg}[x, y]}{\text{CΣ}[x, y]} \times \text{Vrg} + \frac{\text{Cpg}[x, y]}{\text{CΣ}[x, y]} \times \text{Vpg}; \]

The graphs were created using the ContourPlot- and the Plot3D-command in MATHEMATICA, e. g. for creating the Coulomb rings.

\[ \text{Show}[\text{Table}[\text{ContourPlot}[\mu[x, y, n, 2000, 2000, 0, 0] == -150, \{x, -1000, 1000\}, \{y, -1000, 1000\}, \text{PlotPoints} \to 30, \text{ContourStyle} \to \{\text{Thick, Blue}\}, \text{Frame} \to \text{True, FrameTicks} \to \text{None}, \{n, -18, 21, 1\}]]. \]
Appendix D

Maintenance of the Dilution Refrigerator

Diagnosis and Repair

During a standard warm-up procedure, a leak at one of the silver heat exchangers occurred at system II (a Kelvinox 100 dilution refrigerator manufactured by Oxford Instruments). The leak was accompanied by a loss of about one third of the $^3$He-$^4$He-mixture (corresponding to about 200 mbar dump pressure). The insert had to be sent to the manufacturer for repair. There, it turned out that the mixing chamber and the silver heat exchanger were heavily contaminated with oil. Other parts of the circulation line like the outlets of the nitrogen coldtraps were also oily.

The causal chain leading to the accident was probably the following: During operation of the cryostat, a power failure stopped the $^3$He-pump. The cold insert then cryo-pumped the $^3$He-pump against the normal gas flow. Therefore, oily $^3$He was sucked into the dilution unit. During warm-up, liquid helium could not expand because the frozen oil blocked the condenser. Consequently, the circulation line broke at the weakest part, namely at the silver heat exchangers.

The repair included the following steps: Replacement of the mixing chamber and the silver heat exchangers, flushing of the dilution unit with ethanol and subsequent baking, and a final superfluid leak test at Oxford Instruments and replacement of all vacuum tubes belonging to the circulation line, replacement of the nitrogen and helium coldtraps, and replacement of the $^3$He-pump with a modern Pfeiffer-pump at our setup. Additionally, a pneumatic security valve was built into the circulation line.
The Pneumatic Security Valve

The location of the pneumatic security valve is right at the inlet of the $^3$He-pump. It closes and opens with compressed air. As long as a voltage is applied to the valve, it stays open; otherwise it closes. The voltage source of the valve is connected to the pump power supply. Therefore, the valve closes automatically if the pump stops (e.g., during a power failure) and prevents cryo-pumping of the pump against the normal gas flow. If the power failure is so short (few seconds) that the pump resumes work, the valve also opens automatically again. If for some reason the valve shall be closed while the pump is running, one simply disconnects the plug of the valve at the power supply of the pump.

Determination of System Parameters

Heat Load

After reassembling the cryostat, preliminary tests of the functionality were carried out. All tests presented in the following have been done with a dummy secondary insert instead of the AFM-insert and after refilling 16.7 l of $^4$He (corresponding to 167 mbar in dump pressure). Similar tests will be carried out with the AFM-insert in the future for comparison.

The cooling power $\dot{Q}$ of a dilution refrigerator is given by [91, Eq. (7.38)]

$$\dot{Q}(T_{MC}) = 95 \frac{W_s}{K^2\text{mol}} \dot{n}_3 T_{MC}^2,$$

where $\dot{n}_3$ is the molar flow rate of $^3$He from the concentrated phase into the diluted phase and $T_{MC}$ is the temperature at the phase boundary (i.e., the temperature of the mixing chamber in good approximation). The total heat load is given as the sum of the power $P_{MC}$ of the mixing-chamber heater and the incoming heat flux $P_{hl}$ due to warm $^3$He from the heat exchangers but also cabling, etc. In thermal equilibrium, we therefore have $\dot{Q} = P_{MC} + P_{hl}$. Hence, we can deduce $P_{hl}$ and $\dot{n}_3$ from measuring $T_{MC}$ as a function of $P_{MC}$.

In Fig. D.1 such a measurement is shown for the dummy secondary insert with a minimal heat load. We plot $T_{MC}^2$ and fit the data points with a straight line:

$$T_{MC}^2 = 128.5 \frac{\text{mK}^2}{\mu\text{W}} (P_{MC} + 17.9 \mu\text{W}).$$

The heat load is therefore approximately 18 $\mu$W, and from the prefactor we deduce a $^3$He flow rate of $\dot{n}_3 \approx 8 \times 10^{-5}$ mol/s = $5 \times 10^{19}$ s$^{-1}$.

$^1$This was chosen because roughly 5/6 of the mixture consists of $^4$He and about 200 mbar were lost.
Figure D.1: Measurement of the mixing chamber temperature squared as a function of applied power. The theoretically expected linear behavior is clearly visible and can be fitted accordingly. The resulting heat load is approximately 18 $\mu$W.

Cooling Power

We measure the cooling power by setting $T_{MC}$ to a particular value, namely $T_{MC} = 100$ mK. The cooling power $P_{cool}$ is then the applied power $P_{MC}$ necessary to maintain the temperature $T_{MC}$. We can vary $P_{still}$ to optimize the cooling power. We see from Fig. D.2 that we have a maximum cooling power of $P_{cool} \approx 92 \mu$W at a still power of roughly 12 mW. The name of the cryostat, KELVINOX 100, refers to the fact that this system has a nominal cooling power of at least 100 $\mu$W at 100 mK. The fact that we do not obtain this value indicates that the $^3$He-$^4$He-mixture is still not adjusted precisely.

Single-Shot Measurement

A simple method to quantitatively determine the $^3$He-$^4$He-ratio is a so-called single shot: The running circulation is aborted by closing valve V 13A/V 13B and opening valve V 9. The mixture is then pumped back into the dump and all system parameters (in particular pressures and temperatures) are recorded over time. At the beginning, mainly $^3$He is pumped back due to the different vapor pressures of the two He-isotopes. As long as $^3$He is in the system the temperature stays rather constant or even drops since no warm He-gas has to be condensed-in. When all $^3$He is pumped into the dumps, the temperature of the mixing chamber will rise.
Figure D.2: Measurement of the cooling power as a function of still power at $T_{MC} = 100\, \text{mK}$. The maximal cooling power is therefore about ca. $92\, \mu\text{W}$.

quickly. This is clearly observed in Fig. D.3 after almost 5000 s at the dashed line labeled (2). The dump pressure, i.e. $G2 \approx 130\, \text{mbar}$, is an upper estimate for the $^3\text{He}$-amount.

During the whole process, the evaporation rate and hence the temperature (or the pressure) in the still must be kept constant by adjusting the still heater power. This is shown in the bottom panel of Fig. D.3. After approximately 4500 s (dashed line labeled (1)), pressure $P1$ drops quickly. Increasing the still heater power does not help anymore. This indicates that the liquid He-level drops below the still bottom; a fact which is corroborated by a slower increase of pressure $G2$. For a precisely adjusted $^3\text{He}-^4\text{He}$-mixture, the still would run empty after the $^3\text{He}$ is completely pumped out. To increase the liquid He-level in the still, $^3\text{He}$ (and maybe some more $^4\text{He}$) should be refilled.
Figure D.3: Single-shot measurement with a dilution refrigerator. The pressure $G_2$ at the dashed line labeled (2) indicates the $^3$He-amount in the mixture since at that point in time all $^3$He is pumped out of the system (the temperature of the mixing chamber increases quickly). Before that, the still runs empty (dashed line labeled (1)). This is obvious since the still pressure $P_1$ drops quickly and cannot be kept constant. Note that the pressure $P_1$ is scaled with a factor 100 in order to plot it into the same graph as $G_1$ and $G_2$. 
Bibliography


Publications


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