# **Towards quantized conductance in graphene bilayer**



**Master Thesis** 

by

## Maggitti Angelo

Supervisors:

Stijn Goossens(TU Delft, Netherlands)

Prof. Dr. Lieven .M.K.Vandersypen(TU Delft, Netherlands)

Prof. Dr. Martino Poggio (University of Basel, Switzerland)

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#### Abstract

In this Master thesis, split-gate quantum point contacts with 20 nm and 50 nm spacing were fabricated on backgated graphene, both with and without a topgate that covers the entire flake. Whereas significant backgate leakage occurred at voltages of  $\pm 4$  V, negligible topgate leakage was observed up to  $\pm 8.25$  V. The Dirac point was evident as a peak in resistance as a function of backgate voltage. The position of the Dirac point was found to be shifted by chemical doping.

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## **1** Introduction

So far, the unique element carbon (C) has mainly drawn attention in the chemistry community, especially in organic chemistry. Its capability to form covalent bonds, which can lead and effectively leads to big networks, underlines its importance and is the main reason for the existence of cells and life in general[1,2]. Moreover, it is known that carbon forms different allotropes, among them, diamond and graphite have been known since ancient times and drew attention in the physics community as well[1]. Approximately 20 years ago, two further carbon allotropes, the fullerenes and nanotubes were discovered and attracted a lot of attention in the chemistry community as well as in the physics community[3,4,5,6].

In 2004, A. Geim and K. Novoselov from Manchester University discovered graphene, which is a two dimensional allotrope, by applying the technique of micromechanical cleavage[1]. Furthermore, the discovery of graphene opened a new field in the research of physics, not only in fundamental physical research, but also in applied physical research. In the field of fundamental physical research, graphene represented a bridge between condensed matter physics and quantum electrodynamics(QED). Due to the relativistic nature of its massless electrons, it is a unique experimental model to study fundamental aspects of QED[1,7]. On the other hand, if we consider the field of applied physical research, graphene has not only paved the way for future carbon based electronics, because the charge carrier density can be tuned continuously from electrons to holes and vice versa as well as its high mobility[1,7], but also its potential usage as a nanomechanical resonator due to its amazing mechanical stability and high resonant frequency[8].

Towards the usage of bilayer graphene in carbon based electronics, it is important to sufficiently supress the conductivity. With the zero bandgap, this requirement is not full filled. A topgate over bilayer graphene could be a solution to this problem[10]. Split-gates as a QPC are used to getting new insights in the quantum transport of graphene bilayer.

The aim of this master thesis is to measure quantized conductance in bilayer graphene at low temperature, using a split-gate quantum point contact (QPC) as it has not been done and observed up to now. On the way to reach this goal, QPC's as a split-gate geometry were fabricated with a spacing of 20 nm and 50 nm. This master thesis has been carried out in the Quantum Transport group within the faculty of applied sciences at TU Delft under the supervision of Stijn Goossens, Prof. Dr. L. M. K. Vandersypen and Prof. Dr. Martino Poggio.

## Outline

This master thesis is organized as fallows:

## Theory and design of the experiment

In the first chapter, general aspects about graphene, the structure of bilayer graphene and electronic properties of graphene and its bilayer are presented. The subsequent chapter covers calculations that are relevant for the experiment and its design. Further, QPC's as well as the quantum hall effect in general and for bilayer graphene are introduced and related to each other. Finally, the idea of the sample design is developed and explained in detail.

### Fabrication and graphene search

Within this section, the fabrication process as well as the way, how we get and characterize the graphene flakes are demonstrated. The fabrication process is kept general on one hand and on the other hand, details on fabricated samples will be provided.

## Experimental results and discussion

Characterisation of a device is described and discussed. Further, transport measurements are shown and analyzed in detail.

## **Conclusion and Outlook**

Results, which have been obtained, will be summarized and pointed out. Ideas on further experiments, which can be performed, are expressed and explained.

## 2 Theory and design of the experiment

## 2.1 Graphene and its electronic properties

Graphene is a single sheet of carbon atoms that is tightly packed into a two-dimensional (2D) honeycomb lattice and is a basic building block for graphitic materials of all other dimensionalities like 0D buckyballs( $C_{60}$ ), 1D nanotubes and 3D graphite[1,7]. It can be wrapped into these graphitic materials, as shown in Figure 1.



**Figure 1:** Graphene as a basic building block for the three different graphitic material. At the top of the picture, you can see a single graphene sheet. As you go from left to right you can see the buckyball, nanotube and 3D graphite. It is taken from Ref[7].

Graphene's unit cell contains two atoms A and B, as it is shown in Figure 2a, which define complementary sublattices[1,7]. Its lattice vectors can be written as  $a_1 = a_0\sqrt{3}(\frac{1}{2}, \frac{\sqrt{3}}{2})$  and  $a_2 = a_0\sqrt{3}(\frac{-1}{2}, \frac{\sqrt{3}}{2})$ , where  $a_0=1.42$  Å denotes the bondlength of nearest neighbour C atoms, so the length of the lattice vectors is given as  $a:=|a_{1,2}|=\sqrt{3}a_0$  [9]. a is also called the lattice constant. Every carbon atom has four valence electrons. In graphene, three of them are located in hybridized  $sp^2$  molecular orbitals, whereas the fourth one is placed in a  $p_z$  atomic orbital. The two orbitals are perpendicular to each other, where  $sp^2$  lies in the bonding plane.  $sp^2$ -orbitals of neighbouring carbon atoms in graphene overlap and thereby form strong in plane  $\sigma$ -bonds, whereas their  $p_z$ -orbitals overlap as well and form  $\pi$ -(bonding) and  $\pi^*$ -(antibonding) molecular orbitals(Figure 2c). As the energy bands of  $sp^2$ -orbitals are far away from the Fermi energy, they do not contribute to the electrical properties of graphene. It is different with the  $\pi$ -orbitals. Due to the band structure, electrons are delocalized over the entire lattice and therefore, determine the electronic properties of graphene at the Fermi energy. To add, the  $\pi^*$ -band represents the conduction band, which is higher in energy, compared to  $\pi$ , which represents the valence band and is lower in energy.



**Figure 2:** Graphene structure and its unit cell with the symmetry points and orbitals. In the top row, the honeycomb structure and its unit cell, described by a dashed line, are given. Next to it, the first Brillouin zone with its real lattice  $a_{1,2}$  and the reciprocal lattice vectors  $b_{1,2}$  are given. In the bottom row, the orbitals and the symmetry points are presented. K and K' denote the CNP and  $\Gamma$  is the center of the first Brillouin zone. It is taken from www.wikipedia.org.

The band structure of an infinite extended graphene sheet can be calculated in the tight-binding approximation. Within tight-binding approximation, two assumptions are made:

- atomic wavefunctions  $\psi_i$  are localized at each atomic site, where electrons, which belong to atoms of different sublattices, are described by independent wavefunctions that obey Bloch's theorem.
- only nearest neighbour interactions are taken into account

In case of single layer graphene, only in plane nearest neighbour interaction are considered, whereas in bilayer graphene nearest neighbour interaction between different sublattices are only considered for atoms that are placed on top of each other[10,11,12]. The arrangement and ordering of the atoms in bilayer graphene is given as  $A_1, B_2, A_2, B_1$  as it is shown in Figure 3b. This arrangement and ordering is called Bernal stacking[10,11,12,13].

The valence and conduction band touch each other at six symmetry points and out of these six symmetry points, only two of them are distinguishable and labled as K and K'. K and K' represent the charge neutrality point(CNP). They are called Dirac points as well and can be seen in Figure 2e. At electrostatic equilibrium, the Fermi level is lying exactly in these points. The valence band is filled, whereas the conduction band is empty. Around the K(K') point, the energy dispersion relation is linear in single layer graphene and given by  $E=\pm\hbar kv_F$ . It can be explained as fallows:

The hamiltonian, which describes the electronic properties of single layer graphene near the Fermi level and around K(K') is given as

$$H = \begin{pmatrix} \triangle & \hbar v_F(k_x - ik_y) \\ \hbar v_F(k_x + ik_y) & -\triangle \end{pmatrix} [10, 11]$$

where  $\triangle$  is the gap that can be induced. In case that  $\triangle = 0$ , the hamiltonian reduces to

$$H' = \begin{pmatrix} 0 & \hbar v_F(k_x - ik_y) \\ \hbar v_F(k_x + ik_y) & 0 \end{pmatrix}.$$

If we diagonalize the hamiltonian H', we get the energy dispersion relation  $E=\pm\hbar kv_F$ , which is linear. If the gap  $\triangle$  is present, meaning that  $\triangle \neq 0$ , then due to breaking the inversion symmetry, the energy dispersion relation is expressed as  $E(k) = \sqrt{\Delta^2 + (k\hbar v_F)^2}$ .

In case of bilayer graphene the hamiltonian looks similiar to the single layer case, but the momentum operator is quadratic within the hamoltonian. The hamiltonian has the form

$$H'' = \begin{pmatrix} \triangle & \frac{-\hbar^2 (k_x - ik_y)^2}{2m} \\ \frac{-\hbar^2 (k_x + ik_y)^2}{2m} & -\triangle \end{pmatrix}$$

The structure of H" is a consequence of the Bernal stacking[10,11]. Bernal stacking in bilayer graphene leads to a unit cell of 4 atoms. This means that we would have four electronic bands. At low energy, only two bands are relevant[10,11]. As we diagonalize H" and set  $\triangle = 0$ , we get the parabolic energy dispersion relation  $E=\pm\frac{1}{2m}(\hbar k)^2$  as it is shown in Figure 3b. Furthermore, if we apply an electric field, a band gap can be opened, as it is shown in Figure 3c. As a consequence, an insulating state has been induced as the fermi level is lying exactly between the valence and conduction band[10]. By sweeping the voltage from positive to negative, electrons or holes are induced as the fermi level is either shifted towards the conduction band or valence band. In single layer graphene, no insulating state can be realized because no gap can be induced by applying an electric field. This is due to its band structure, as it is demonstrated in Figure 3a. If we use hexagonal boron nitride as a substrate for single layer graphene, a gap can be opened[13]. a to c in Figure 3 are taken from Ref[10] and c to e are taken from www.wikipedia.org.



Figure 3: Bandstructure of single and bilayer graphene. (a) and (b) show bandstructure of single and bilayer graphene. (c) Gap opening in bilayer graphene. (d) Touching valence and conduction band at hexagonal plane. (e) Biased Bernal stacking for bilayer graphene.

## 2.2 Quantum point contact

QPC as a split-gate is characterized by its lateral confinement in the y-direction (transverse direction) and a narrow channel in the x-direction (longitudinal direction). The lateral confinement in the transverse direction represents a barrier to the motion of electrons. Electrons within the constriction form standing waves. These standing waves represent modes, which are occupied by electrons or holes respectively in graphene. These modes form 1 dimensional subbands. Not all the modes can propagate through the constriction. Propagation occurs if the standing waves constructively interfere. This means that the width W of the constriction obeys

$$W = \frac{N\lambda_F}{2} \Longrightarrow k_F = \frac{N\pi}{W}$$
(1)

where N is an integer and describes the number of propagating modes. We can conclude that these N modes, which describe electronic states of electrons or holes in graphene respectively, are quantized. Therefore, the conductance that is carried by these modes is quantized as well. Each individual mode contributes  $G_Q = \frac{2e^2}{h}$  equally to the total conductance with  $G_Q$ , being the conductance quantum. The total quantized conductance in graphene is expressed as

$$G = Ng \frac{2e^2}{h}$$

$$= N4 \frac{2e^2}{h}$$
(2)

with g = 4, including the vally and spin degeneracy in graphene. This equal contribution of  $G_Q$  by each mode is explained by the property that the product of group velocity  $v_n(k) = \frac{1}{\hbar} \frac{dE_n(k)}{dk}$  and one dimensional density of states  $\rho_{1D}$  is mode independent, despite that  $v_n$  and  $\rho_{1D}$  differ from one mode to the other. An additional reason to add is that  $v_n$  and  $\rho_{1D}$  vanish in the narrow channel as the Fermi level lies exactly at the one dimensional subband.

However, a question that arises is how can we see quantized conductance of the QPC in graphene? This question can be answered by the arguement of subband depopulation. Depopulation of subbands can be done either by applying a magnetic field **B** or at magnetic field **B**=0. The case with zero magnetic field will be explained here and for the applied magnetic field, explainations will be provided in the section of integer quantum hall effect.

#### Shifting the Fermi level with the back gate (B=0)

As we strongly decreasy the voltage on the split-gates, which means that we set the voltage strongly negative, the channel is pushed by the fact that the width of the constriction narrows down, while the depletion region extends. By this, less modes are present. This means that less subbands are visible and occupied. As a matter of fact, we have depopulated the subbands.

## 2.3 Integer Quantum Hall Effect

#### 2.3.1 General

The Integer Quantum Hall Effect is a result of electrons wave nature. In the classical Hall Effect, electrons perform skipping orbits due to an applied perpendicular magnetic field to the conductor. These skipping orbits are stable and remain localized in the center of the conductor because of a constant Lorentz force. In the quantum mechanical case, these electrons are described by their individual electronic states, which are denoted as edge states. As they can be resolved at higher magnetic fields, they refer or correspond to Landau levels. Electrons that get released from a current source contact(=reservoir1)(Figure 5) are generally filled up to its chemical potential, which is bellow the Fermi level. These edge states are one dimensional, characterised by the quantum numbers  $\{n,k_y\}$  and propagate along a specific edge of the confining channel at one side and in a specific direction, whereas electrons from the opposite side are filled up to the chemical potential of the drain contact(=reservoir4)(Figure 5) and propagate in the opposite direction. The different propagation directions are governed by the Pauli exclusion principle. The resulting net current  $I_{net}$  and the Hall resistance that can be determined show quantization in units of  $\frac{e^2}{h}$  [15,16].

### 2.3.2 Integer Quantum Hall Effect in graphene bilayer

Charge carriers in graphene are massless Dirac fermions and obey the relativistic Dirac equation from QED[1,7,18]. Electrons and their counter parts holes are described by the same spinor wave function, but with two different components[1,7,18]. The spinor wave function possesses a spin index. In graphene, this index indicates the sublattice, rather than the real spin of the electrons. That's why the spin index is referred to as the pseudospin[1]. To these massless Dirac fermions for single layer graphene and massive Dirac fermions for bilayer graphene, a Berry's phase is mapped. In case of single layer graphene, Berry's phase is  $\pi$ , while in bilayer graphene, Berry's phase is equal to  $2\pi$ [18]. Berry's phase of  $2\pi$  for bilayer graphene arises, because graphene's charge carriers move on cyclotron orbits as a magnetic field is applied. Cyclotron orbits are closed paths. As electrons move once along the closed cyclotron orbits, they pick up a phase due to the orientation of the pseudospin. Nevertheless, within the standard theory, each filled single-degenerate Landau level contributes one conductance quantum  $\frac{e^2}{h}$  towards the hall conductivity[18]. In bilayer graphene, it was found that the plateau at zero hall conductivity undergoes a step size of 2(Figure 4b). This step is explained by the coupling between two graphene layers[18]. This coupling leads to a transformation of massive Dirac fermions into chiral quasiparticles(holes)[18].



**Figure 4:** Quantum hall in single and bilayer graphene. (a) Conventional IQHE with equidistant step due to edge states, propagating in opposite directions. (b)anomolous QHE in bilayer graphene due to intralayer coupling at zero hall conductivity, leading to formation of quasiparticles. (c) anomolous QHE in single layer graphene due to interlayer coupling at zero hall conductivity, leading to formation of quasiparticles.

#### 2.3.3 Integer Quantum Hall Effect and quantum point contact

To see magnetoconductance with the split-gates, meaning that the conductance through the split-gates is described in presence of high magnetic field and still quantized, two approaches can be applied. On one hand, we consider the split-gates individually and show depopulation of the subbands within the narrow channel, formed by the split-gates. On the other hand, we use the split-gates in combination with the hall bar device to show suppression of the magnetoconductance, but the hall conductance remains quantized and unchanged in its mathematical expression, if the contacts are ideal and placed farther appart. Only the suppression of magnetoconductance will be explained in detail.

# Suppression of magnetoconductance in hall bar device in combination with split-gates for ideal contacts

Split-gates in combination with a hall bar device can be used to pinch off edge channels. At high magnetic field, a large fraction of edge channels is backscattered and remains in thermal equilibrium with the source contact in the hall bar device. Only a small fraction is transmitted through the split-gates and contribute to the magnetoconductance, which is still quantized. Another idea of pinching off the channel at high magnetic field is to depopulate the channels(=subbands) that are occupied by electrons. This is done by strongly decreasing the gate voltage on the split-gate.

Figure 5 shows a schematic drawing of our hall bar device, including the split-gate next to it. It shows

two edge channels, one of the two edge channels is fully backscattered, while the other one transmits through the split-gate. Our calculations refer to Figure 5. We assume of having ideal contacts that are placed farther apart. Further, we consider P' to be the total number of given channels and Q' the number of transmitted channels with Q' < P'. The source is set to the chemical potential  $\mu_L = eV_1$  and drain to the chemical potential  $\mu_R = 0$ .

The net current  $I_1$ , which flows from contact 1 to 4 is given as

$$I_{1} = \frac{2eQ'(\mu_{L} - \mu_{R})}{h} = \frac{2e^{2}Q'V_{1}}{h}$$
(3)

We can define a fraction  $m = \frac{(P'-Q')}{P'}$  that describes the ratio between the number of backscattered edge channels(P'-Q') and the total number of given edge channels P'. Equation (3) can be rewritten as

$$I_1 = \frac{2e^2 P' V_1(1-m)}{h}$$
(4)

In order to be able to calculate the hall resistance out of the hall conductance, we need to know the chemical potentials of the contacts 2,3,5 and 6. If we look at contact 2, we can say that it sees channels, originating from the source. Thus, edge channels that arrive at contact 2 equilibrate to  $\mu_L$  with  $\mu_2 = \mu_L = eV_1$ . Instead, contact 5 sees only edge channels that arrive from contact 4. Therefore,  $\mu_5 = \mu_R = 0$ . With contact 6, we have two contributions that need to be taken into account. On one hand, we have the transmitted channels that are originated from contact 4 and on the other hand, the backscattered channels, originated from contact 1. Then,  $\mu_6$  can be expressed as

$$\mu_{6} = \frac{(P' - Q')\mu_{L} + Q'\mu_{R}}{P'}$$

$$= eV_{1}m$$
(5)

If we consider contact 3, we have two contributions as well. From contact 4, we have edge channels that are fully backscattered with a chemical potential  $(P' - Q')\mu_R$ , whereas from contact 1, transmitted edge channels appear with a chemical potential  $Q'\mu_L$ . Now, we are able to express the chemical potential for contact 3 as

$$\mu_{3} = \frac{Q'\mu_{L} + (P' - Q')\mu_{R}}{P'}$$

$$= eV_{1}(1 - m)$$
(6)

In our last step, the hall resistance and hall conductance will be determined. If we take equations (4,5) and the expression for  $\mu_2$ , the hall resistance and the hall conductance are given as

$$R_{H} = \frac{V_{26}}{I_{1}} = \frac{\frac{\mu_{2} - \mu_{6}}{e}}{\frac{2e^{2}P'V_{1}(1-m)}{h}}$$

$$= \frac{h}{2e^{2}P'}$$
(7)

$$G_{Hall} = R_H^{-1}$$

$$= \frac{2e^2 P'}{h}$$
(8)

Equation (8) proves that the hall conductance remains quantized and unchanged, as backscattering occurs.



**Figure 5:** Hall bar structure with split-gate. Split-gate is used to backscatter one channel, while the other is transmitted. Contacts are chosen to be ideal with respect to the calculations. No disorder is present. It is taken from Ref[17].

## 2.4 Calculations

Some calculations have been performed in order to estimate the range of charge carrier densities that can be reached in graphene bilayer as well as to determine the Fermi wavevector  $k_F$  as a function of the backgate ( $V_b$ ) and topgate ( $V_t$ ) voltages. As the topgate and backgate voltages are experimentally limited in range, it is important to estimate the charge carrier densities. Our Fermi wavevector is given as[16]

$$k_F = \sqrt{\frac{4\pi |n|}{g}} \tag{9}$$

with g describing the degeneracy(=spin+valley) and n refers to the charge carrier density.

For single and bilayer graphene, the degeneracy is 4. The spin degeneracy and the valley degeneracy are both equal to 2. For the spin, the degeneracy is explained as follows: Atoms A and B (A<sub>1</sub>,B<sub>1</sub>;A<sub>2</sub>,B<sub>2</sub>) in single layer (bilayer) graphene, which define a sublattice individually, have a  $p_z$ -orbital that is filled with a single, unpaired electron individually. According to the Pauli exclusion principle, only one electron with the opposite spin can be added to the individual  $p_z$ -orbital, without changing its energy. Because of translational symmetry, only sublattices of one layer have to be taken into account in case of bilayer graphene. Thus, two electrons can be added into the  $p_z$ -orbitals. As a consequence,  $k_F$  from equation (9) can be written as

$$k_F = \sqrt{\pi |n|}$$
 for single and bilayer graphene (10)

In the next step, we are going to calculate the average charge carrier density n' as a function of backgate and topgate voltage for graphene bilayer. We use a parallel capacitor model, where the space within graphene bilayer is treated as a dielectric and use the Gaussian divergence theorem to calculate the individual charge carrier densities and choose the positive z-axis perpendicular to the graphene layers. The displacement field of the graphene innerspace has the component  $D_b=(0,0,D_b)$  with respect to the bottom layer and  $D_t=(0,0,D_t)$  with respect to the top layer. For the calculations here, we choose the applied backgate voltage as positive and the applied topgate voltage as negative. In that case, the electric field is orientated towards the positive z-axis.

$$div(D_b) = div(-\varepsilon_b \varepsilon_0 E_b)$$

$$= \frac{ne}{\varepsilon_0} \Longrightarrow n(V_b) = \frac{-\varepsilon_b \varepsilon_0 (V_b - V_b^\circ)}{ed_b}$$
(11)

$$div(D_t) = div(-\varepsilon_t \varepsilon_0 E_t)$$

$$= \frac{ne}{\varepsilon_0} \Longrightarrow n(V_t) = \frac{-\varepsilon_t \varepsilon_0 (V_t - V_t^\circ)}{ed_t}$$
(12)

with  $V_b^{\circ}, V_t^{\circ}$  the individual offset voltages of backgate and topgate.  $d_b$  and  $d_t$  are the individual thicknesses of the dielectric. These offsets are determined experimentally by measuring the resistance, while a backgate and topgate sweep are performed.

The average charge carrier density n' is determined as

$$n' = \frac{1}{2}(n(V_b) + n(V_t))$$

$$= \frac{1}{2}\left(\frac{-\varepsilon_b\varepsilon_0(V_b - V_b^\circ)}{ed_b} + \frac{-\varepsilon_t\varepsilon_0(V_t - V_t^\circ)}{ed_t}\right)$$
(13)

In Figure 6, the plot of the average density n' is shown.



**Figure 6:** Average charge carrier density n' as a function of backgate and topgate.  $V_b$  denotes the backgate and  $V_t$  the topgate. The individual offset voltages have been set to zero.  $\varepsilon_b = \varepsilon_t = \varepsilon$  as the dielectric is  $SiO_2$ . The chosen range for backgate and topgate are -100  $V \le V_b \le 100$  V and -40  $V \le V_t \le 40$  V and agree with the possible voltages that can be applied with the given experimental set up. The yellow to red region corresponds to the inducement of holes, whereas the bright blue to dark blue region corresponds to the inducement of electrons. The point of zero density with  $V_t = V_b = 0$  is the charge neutrality point (CNP).

## 2.5 Sample design



**Figure 7:** Device Design of AM1. It consits of 6 devices. (a) topgate, (b) global topgate, (c) split-gate with 20 nm gap size + global topgate, (d) split-gate with 50 nm gap size + the global topgate, (e) split-gate with 20 nm gap size, (f) split-gate with 50 nm gap size, (g) hall bar device, (h) graphene flake of 5  $\mu$ m width and 25  $\mu$ m length and (a') leads for 4-terminal measurement.



**Figure 8:** Schematic sideview of split-gates with 20 nm as well as 50 nm gap size. The graphene flake is shown as two parallel linse. Split-gates are composed of a 20 nm thick  $SiO_2$  layer, fallowed by two metal layers. The first layer is 5 nm of Cr and the second layer is 60 nm of Au.



**Figure 9:** Schematic sideview of split-gates with global topgate and 20 nm as well as 50 nm gap size. The graphene flake is shown as two parallel linse. Split-gates are composed of a 20 nm thick  $SiO_2$  layer, fallowed by two metal layers. The first layer is 5 nm of Cr and the second layer is 60 nm of Au. The global topgate is composed of a 50 nm thick  $SiO_2$  layer, fallowed by two metal layers. The first layer is 10 nm of Ti and the second layer is 50 nm of Au.

The design for our device, with which we have the intention to perform quantized conductance measurements, is built in such a way that we can first check if our graphene flake is a bilayer. To do this, we designed a topgate with a width of  $1\mu m$ . Together with the backgate, we apply an electric field to open a band gap. Then, we push the Fermi level into the gap with a suitable choice of gate voltages. As a consequence, the graphene flake is in a insulating state and conductance is suppressed. This property is unique for graphene bilayer. Furthermore, the two split-gate devices with 20 nm and 50 nm spacing were intended to be used for the quantized conductance measurements. A global topgate was designed to provide a ballistic regime by locally tuning the charge carrier density as well as keeping the charge carrier density constant. The width of the contacts, which are placed aside the split-gates, are chosen to be 300 nm with a spacing of 200 nm each to the slightly wider contacts. Moreover, two split-gates with a spacing of 20 nm and 50 nm are put next to each other, but excluding a topgate. One reason is to compare the effect of having a topgate or not. Although, it is worth and promising to be able to compare the influence of a topgate, but the core reason for these single split-gates was to see and measure quantized conductance. We expect that the split-gate device, excluding a global topgate, will rather show quantized conductance, because the meanfree path is longer than in the topgated device. First of all, in order to do that, we had to choose a proper dimension for the split-gate spacing. We have chosen the spacing dimensions to provide

the regime for quantized conductance. If we had chosen a spacing, which was to small, we would have been either in the range of the Fermi wavelength  $\lambda_F$  or below. In that case, we would not see quantized conductance. By this fact, we are lead to choose the spacing of the split-gates beyond the Fermi wavelength  $\lambda_F$ . With 50 nm spacing we are clearly above the Fermi wavelength  $\lambda_F$ . Moreover, to see quantized conductance we need to pinch off the channel. This can be done either with or without a magnetic field. In case of the magnetic field, we designed a hall bar device. The hall bar device in combination with the split-gate, which is placed next to it, pinch off the channel can be realized by backscattering as it was explained in chapter 2.3. Main usage of the hall bar device was to measure the quantum hall effect in order to have a further proof for graphene bilayer.

## **3** Fabrication and graphene search

## **3.1 Graphene search**

#### 3.1.1 How to get graphene flakes

Comercially available graphite flakes are taken as a source. One graphite flake is placed onto a scotch tape with the dimension of the Si chip. The length of the scotch tape is chosen in such a way that it can be partly folded at the two ends. Parts of the scotch tape are glued together and slowly pealed off again. Thereby, a graphite flake has replicated itself. We apply this procedure, which is called mechanical cleavage, until a nice coverage of graphite has been reached on the scotch tape.

In the next step, we put our scotch tape with graphite on top of a Si wafer, where we apply mechanical stress by rubing the scotch tape on the surface of the Si wafer. As a matter of fact, graphite sticks better to the surface of the Si wafer due to the applied mechanical stress. In advance, the surface was treated with HMDS(Hexamethyldisilazan) that makes the surface more hydrophobic and therefore, it increases the probability to get a larger amount of graphene sheets. The chemical structure of HMDS is shown in Figure 10. A further reason to treat the surface with HMDS was its property to increase the mobility in graphene.



**Figure 10:** Chemical structure of HMDS. The NH group is the reactive center. When  $Si(OH)_4$  together with water reacts with the NH group of HMDS, Si-O-Si bridges are formed on the Si wafer surface. As a consequence, the surface gets hydrophobic. It was taken from www.wikipedia.org.

Finally, the scotch tape is released very gently from the piece of Si. The whole process is carried out in the clean room, because it is known that graphene's sticking property is very sensitive to humidity and temperature.

#### 3.1.2 Graphene search and its characterisation

If we consider the dimension of a graphene flake, which is in the range of microns, microscopic techniques have to be used to look for graphene flakes. To make the search of graphene flakes easier, a marker pattern was written on the Si wafer, before graphene flakes were put on top of the Si wafer. In this work, we used the light microscop Olympus to look for graphene. To add, a SEM cannot be used as a tool to look for graphene, because dirt is deposited on the graphene flake. As a consequence, it would be damaged.

However, in order to characterise the required graphene flake, three methods can be used. One method is Raman Spectroscopy, which is very accurate to distinguish single layer from bilayer as mentioned in Ref[19], but has not been used in this work. A further method is the suppression of conductance, which can be used specifically for graphene bilayer. In this method, we make use of the fact that graphene bilayer has a band gap, when an electric field is applied. This has been used for one device. As a last method, which has been used in this work, is the contrast method. Within this method, we take a picture of our graphene flake with the light microscop Olympus at a 100 times magnification. The picture is imported in the photo shop software to determine the intensities that are needed to calculate the contrast as it is defined in Ref[21]. For bilayer graphene, the expected contrast should be around 0.08. The value is taken from previous data. If graphene is chemically doped, the contrast can be less than 0.08[19]. This is especially true for graphene on a Si/SiO<sub>2</sub> substrate. To overcome this problem, we planed to use pure Si wafers without a SiO<sub>2</sub> layer and deposit a layer of HfO<sub>2</sub>(Hafinium oxide) because it increases the contrast of graphene as it is reported in Ref[22]. In order to use the properties of HfO<sub>2</sub>, we first have to figure out what kind of thickness we need to deposit on the clean Si wafer. This question is answered by plotting the contrast as a function of its dielectric thickness at a certain wavelength  $\lambda$ . The plot of the optimum contrast and determination of the right thickness is going to be part of Maake's Master Thesis.

## 3.2 Fabrication

In the present section I am going to explain, how the samples are cleaned. Furthermore, I will briefly present the process to generate a device.

## 3.2.1 Sample cleaning

Two by two cm Si wafers with an intrinsic  $SiO_2$  layer are sonicated for two minutes in acetone and two minutes in water. Then, the wafers are blown dry with nitrogen. In addition, another beaker is filled with a "piranha" solution. This solution is 3:1 mixture of  $H_2SO_4$ :HOOH sulfuric acid and hydrogenperoxyd. It

is important to remember that the beaker is first filled with sulfuric acid, before hydrogenperoxyd is added. This is explained by the fact that sulfuric acid is a weaker acid compared to hydrogenperoxyd. However, the Si wafers are inserted into the beaker with piranha solution and left there until no chemical reaction is visible. As a last step, the sample holder with the Si wafers are droped into a beaker with water and left there for a few minutes, before the samples are blown dry with nitrogen.

#### **3.2.2** Device fabrication



**Figure 11:** The process flow in lithography as we start from left with e-beam exposure towards the right, where we end with the lift off. It was taken from Ref[20].

We have used e-beam lithography to define our contacts and gates in a resist layer. The process is shown in Figure 11 and consists of the following steps:

- Resist spinning
- E-beam exposure and development
- Metal and oxide deposition
- Lift off
- **Resist spinning:** A double layer of PMMA has been used as a resist. The double layer improves the lift off process. As the bottom layer, PMMA 495K a4 was used. It was spun with 6000 rpm and baked for 10 minutes at 175 °C. For the top layer, PMMA 950K a3 was used. It was spun with 5000 rpm and baked for 10 minutes at 175 °C.
- E-beam exposure and development: First of all, the structure, which is going to be written with e-beam, has to be designed. This is done by using the CAD program. Moreover, Dose-Tests need to be performed in order to figure out, which dose is most suitable for our structure. If we use the wrong dose, the structure will be over exposed. This means that the structure will not be properly written. Further details on these Dose-Tests are given in the appendix. The designed structure is

written with an e-beam pattern generator (EBPG) into the resist. In case of the contacts, a dose of  $1300 \frac{\mu C}{cm^2}$ . Thereby, the covalent bonds in the polymer are broken. Thus, the resist becomes soluble in a developer. We have used MIBK(=methyl isobutyl ketone):IPA(=Isopropylalcohol) 1:3 as a developer with a development time of 120 seconds and rinsed it subsequently in IPA. After that, it was blown dry with nitrogen.

- Metal and oxide deposition: Metal and oxide deposition are done by electron beam evaporation in a vacuum system, called AJA with a constant presure of ~ 3 ·10 <sup>-8</sup> mbar. The deposited metal were Cr, Ti and Au. The amount of metal and oxide, which were deposited, are listed in Figure 8 and 9. Only approximate values for the metal and oxide deposition can be given, because the individual Z-ratio of the metals and oxide were not accurate. An explaination could be that the calibration was not perfect or some problems with the individual crystals might have occurred.
- Lift off: The final step in the fabrication process is lift off. Within this step, the remaining resist is dissolved by immersing the sample in hot acetone (53 °C) for 4 hours in total. We did it first for two hours, but it did not come off properly when we rinsed it in cold acetone. Therefore, we had to put the sample in hot acetone (53 °C) again for another two hours.

## **4** Experimental results and discussion

## 4.1 Device characterization

In order to start with the measurements, backgate and topgate voltages need to be checked. This is done by a leak-test.

### 4.1.1 Leak-Tests

For our device, a leak-test for the backgate and topgate has been done at 4 K. The backgate leaked at  $\pm 4$  V (Figure 12a), whereas the topgate did not leak up to  $\pm 8.25$  V(Figure 12b). The topgate, which has been tested, is denoted with a in Figure 7. As the backgate has already leaked at low voltages, we have tried to figure out, what caused the leakage. Three reasons came up to our mind. Either a contact was broken or slightly damaged, a deffect in the  $SiO_2$  layer was present or due to the bonding with a higher force, the  $SiO_2$  layer was damaged. With an optical microscope, we checked the contacts and the bonding pads. No broken or damaged contacts were found. It must have been a defect in the  $SiO_2$  layer. If the defect was caused by bonding or during evaporation, could not be determined.



Figure 12: Leaktest of backgate and topgate. **a** Leaktest of backgate with leakage at  $\pm 4$  V. **b** Leaktest of topgate with no leakage upto  $\pm 8.25$  V.

#### 4.1.2 Accessing the Dirac Point

As the backgate was leaking, reproducement of the Dirac point could only be done by sweeping the topgate and keeping the backgate at fixed voltages. Three topgate sweeps have been performed for backgate voltages -4 V, 0 V, 4 V and thereby, the current was measured. With ohms law, the current and topgate voltages were converted into resistance. Dirac Point, where the resistance was plotted against the topgate voltages, was reproduced successfully (Figure 13). If we look at the resistances in Figure 13, we can see that for  $V_b(backgate) = 0$  V the resistance is higher compared to  $V_b(backgate) = 4$  V. This could suggest that our graphene flake is not a bilayer, because as we open the gap in graphene bilayer, less charge carriers are present. According to that, the Resistance at  $V_b(backgate) = 4$  V should be higher. As we have chemical doping in graphene, some charges could be trapped in the gap and this would lead to a lower resistance. These trapped charges could have already been present in graphene as a fact of either processing or an intrinsic property. Disorder leads or can lead to a local accumulation of charges, which decreases the resistance. To sum up, we can not exclude that we had a bilayer or not. It remains an open question, but as the backgate was leaking, we decided that there is no reason to carry on with this sample.

## 4.2 Quantized Conductance Measurements: Strategy and Approach

Quantized Conductance can be measured as long as the regime within the channel, which is formed by the split-gate, is ballistic. To give evidence for this ballistic regime, we choose to put a global topgate over the split-glates with 20 nm and 50 nm spacing. With this global topgate, we want to tune as well as



Figure 13: Reproducement of Dirac point. Three topgate sweeps were performed at  $V_b(backgate) = 0$  V(black curve),  $V_b(backgate) = 4$  V (red curve) and  $V_b(backgate) = -4$  V (blue curve). Dirac peak is shifted, which indicates the presence of chemical doping in graphene.

to keep the charge carrier density constant. This leads to an elongation of the mean free path. Further, quantized conductance manifests itself in depopulation of subbands. In case of zero magnetic field, subband depopulation will be visible by keeping the charge carrier density n constant with the global topgate and push the Fermi level downwards with the backgate. The steps in the resistance should correspond to multiple values of n. As we apply a constant magnetic field and decrease the split-gate voltage, we expect the longitudinal resistance  $R_{xx}$  to increase in steps. The steps in resistance tells us, how many subbands have been depopulated. Instead of keeping the magnetic field constant, we can change the filling factor and leave the split-gate at a constant voltage. Then, while we measure the longitudinal resistance  $R_{xx}$  as a function of varying magnetic field,  $R_{xx}$  demonstrates finite oscillations. The maxima of these oscillations increase with increasing field. The individual maxima of  $R_{xx}$  revise the information about the number of edge states, which were transmitted through the split-gates.

## 5 Conclusion and Outlook

Fabrication of QPC's as split-gate with 20 nm and 50 nm gap spacing was demonstrated. One set of QPC's were entirely covered with a global topgate, while the others were lacking of a global topgate.

Split-gate with 50 nm gap spacing is a promising candidate to observe quantized conductance, as we are clearly above the Fermi wavelength 7  $nm \le \lambda_F \le 11$  nm (Calculated for graphene with backgate voltage 100 V and global topgate voltage 40 V). The basement of observing quantized conductance is to show subband depopulation. Subband depopulation can be observed by considering a magnetic field or not. With no magnetic field, depopulation of the subbands is realized and made visible by shifting the Fermi level. Fermi level shifts are done by using a backgate. In connection with subband depopulation, the backgate needs to be decreased in order to observe the decreasing number of populated subbands. As we include the magnetic field, depopulation of subbands can be reached at constant magnetic field as well as at varying magnetic field. At constant magnetic field, depopulation of subbands can be realised by strongly decreasing the split-gate voltage or suppressing magnetoconductance, when split-gates are coupled to the hall bar device. In a calculation, it has been shown that suppression of magnetoconductance does not change the quantization of the hall conductance, if ideal contacts are provided and split-gates are placed farther appart from the hall bar device. Varying the magnetic field and keeping the split-gate voltages constant, depopulation is visible in increasing oscillation maxima of the longitudinal resistance  $R_{xx}$ , which are finite. Leak tests were performed and showed a backgate leakage at 4 V. For the topgate, no leake was visible up to 8.25 V. Dirac point was reproduced, but was shifted due to chemical doping in graphene. Sweeps of the topgate lead to a lower resistance at the Dirac point for  $V_b$ (backgate)= 4 V. Reasons were given, among them disorder, trapped charges or impurities due to fabrication process were discussed. It remained unclear, if a bilayer was present or not.

As soon as quantized conductance is measured in this device, the coming expriment can be to couple the QPC as a split-gate to a quantum dot. In the first step, QPC acts as a tunable tunnel barrier, where selective spins from graphene electrons could be or are chosen to tunnel through the created barrier by the split-gates. The spins are confined in a quantum dot and therefore, usable to form spin qubits. In the second step, the QPC as a tunnel barrier could be modified to a selective spin filter.

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## 7 Appendix

Dose tests were performed by using e-beam lithography in order to determine the appropriate dose for the split-gates and topgates. In the subsequent Figures, SEM pictures of the dose test results are shown for the dose 1000  $\frac{\mu C}{cm^2}$  and 1300  $\frac{\mu C}{cm^2}$ . From Figures 14 to 18 we can deduce that 1300  $\frac{\mu C}{cm^2}$  or even higher



**Figure 14:** Layouts of test designs for different side size spacing. The gap for the channel, formed by the split-gates are set to zero and 20 nm. The left number column denotes the gap size of the split-gates on one hand, and on the other hand the side gap spacing for the composed structure. The gap size for the split gates varies from 10 nm to 50 nm.

is suitable to write and pattern the structure if the side spacing is at least 50 nm. In Figures 17, 18 and 19



**Figure 15:** SEM picture of the patterned test designs. Designs were patterned with a dose of 1300  $\frac{\mu C}{cm^2}$  for different side and gap spacing. For 20 nm to 30 nm side spacing, overexposure occurred. Therefore, PMMA was accumulated in the side space.



**Figure 16:** SEM picture of the patterned test design. Design was patterned with a dose of 1300  $\frac{\mu C}{cm^2}$  with a side spacing of 50 nm and a gap of 20 nm. Structure was well written and no overexposure is visible.

we can see the split gates, which were patterend with a dose of  $1300 \frac{\mu C}{cm^2}$ . With this dose, a gap size of 30 nm can be reached and easily written, without expecting overexposure. A higher dose can be chosen as well and no overexposure should occur. From Figure 20 and 21 we can deduce that a dose of  $1000 \frac{\mu C}{cm^2}$  or smaller is not suitable for the structures.



**Figure 17:** SEM picture of the patterned split-gate design. Design was patterned with a dose of 1300  $\frac{\mu C}{cm^2}$  with a gap of 20 nm. Structure was well written and no overexposure is visible. The measured gap in the SEM picture is higher. This difference might be due to the resolution of the SEM.



**Figure 18:** SEM picture of the patterned split-gate design. Design was patterned with a dose of 1300  $\frac{\mu C}{cm^2}$  with a gap of 30 nm. Structure was well written and no overexposure is visible. The measured gap in the SEM picture is slightly higher. This difference might be due to the resolution of the SEM.



**Figure 19:** SEM picture of the patterned split-gate design. Design was patterned with a dose of 1300  $\frac{\mu C}{cm^2}$  with a gap of 40 nm. Structure was well written and no overexposure is visible. The measured gap in the SEM picture is slightly higher. This difference might be due to the resolution of the SEM or the e-beam itself.



**Figure 20:** SEM picture of the patterned test design. Design was patterned with a dose of  $1000 \frac{\mu C}{cm^2}$  with a gap of 20 nm and a side spacing of 40 nm. Structure is not properly written and a slight overexposure is visible. The measured side spacing in the SEM picture agrees with the wanted size.



**Figure 21:** SEM picture of the patterned test design. Design was patterned with a dose of  $1000 \frac{\mu C}{cm^2}$  with a gap of 20 nm and a side spacing of 50 nm. Structure is not properly written and a slight overexposure is visible. The measured side spacing in the SEM picture agrees with the wanted size.

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