Transport Experiments on Suspended Graphene Devices

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Abstract

In this thesis, sophisticated conductance and noise measurements were employed for studying electron transport through suspended graphene devices in order to understand the fundamental properties of graphene. The experiments were conducted at low temperatures down to $T = 10$ mK, and at high magnetic fields up to $B = 9$ T on suspended graphene devices. In these devices, graphene is connected only to the metallic contacts leaving the graphene flake intact of outside disturbances, and close to ideal theoretical behavior. The work was divided into two segments: quantum transport studies in the zero magnetic field using rectangular bi- and monolayer graphene devices, and magnetotransport measurements at high magnetic fields on Corbino ring devices.

In the case of the rectangular graphene devices, a model for contact doping in monolayer graphene by the metal leads was developed first. This facilitated understanding of the transport through the whole device and served as a basis for understanding the origin of the observed Fabry-Pérot resonances. The resonances were used to demonstrate the phase-coherent transport and long mean free path in the devices. Two sets of noise measurements were performed on these devices. First, low frequency 1/f noise measurements on suspended bilayer graphene (BLG) devices revealed extremely low flicker noise levels that was contributed to the substrate-free form of the devices and the effective screening of fluctuations in BLG. The low intrinsic noise level was exploited in a gas sensing application, where adsorbed gases were detected through the extra noise caused by molecules that had landed on the device. In the later set of noise measurements at higher frequencies, $f = 600 - 900$ MHz, noise thermometry was employed for characterization of the electron-phonon coupling in bi- and monolayer graphene.

Finally, suspended graphene Corbino devices were developed for studying integer and fractional quantum Hall effect (IQHE and FQHE). The observed FQHE was explained with the established theory of composite fermions. Based on the measurements, it was concluded that the composite fermions in graphene are Dirac particles with cyclotron mass around one electron rest mass. At very high fields and low charge carrier densities, evidence of Wigner crystallization was obtained. Additionally, the breakdown of quantum Hall effect was studied at the filling factor $\nu = 0$ in the middle of the lowest Landau level. Zener tunneling between Landau sublevels was found to facilitate the breakdown at fields below 7 T, while a more standard behavior due to bootstrapped electron heating was observed at higher fields.

Keywords graphene, electron transport, Fabry-Pérot, shot noise, electron-phonon coupling, 1/f noise, quantum Hall effect, fractional quantum Hall effect, composite fermions, Wigner crystal, Zener tunneling
The result presented in this thesis are part of the work carried out during my doctoral studies starting from 2014. The work was carried out in the Nano group of the Low Temperature Laboratory, Department of Applied Physics at Aalto University.

Originally, I came to Nano group of the Low Temperature Laboratory (LTL) of Helsinki University of Technology (TKK in Finnish) as a master student/summer trainee for the summer of 2010 to study suspended graphene mechanical resonators. Since that time many things have changed: the laboratory has changed its name twice, the university has changed its name, and my main research direction has changed many times from mechanical resonators to zero magnetic field electron transport measurements, and from there to quantum Hall experiments. The two later subjects fall under general category of transport experiments, which subsequently is the title of this thesis. Interestingly, I have not published a single paper till this date on the original subject. However, I believe that exploring multiple rather different subjects has ultimately been more interesting than it would have been to stick with a single subject. I have to acknowledge that I have been lucky in a sense that many experiments during my doctorate studies worked without big issues and publishable results were obtained.

This work was not conducted alone. Foremost, I would like to thank my supervisor professor Pertti Hakonen for his guidance, general freedom to choose my working practices, and abundant supply of new ideas that have facilitated the transitions to new research directions. It has been a pleasure working with you. I would also like to thank all the collaborators and co-authors for their contributions and the tremendous support. I especially acknowledge Ari Harju, Manohar Kumar, Sorin Paraoanu, Bernard Plaçais, Edouard Sonin, Andreas Uppstu, and Pauli Virtanen.

I would also like to thank all the support personal in the lab. Without our secretaries and the work shop guys everyday work would have been much more difficult. Staff scientist Alexander Savin deserves special thanks, with his experience and can-do attitude many problems were solved.

In addition to the research work, I had the pleasure to function as a lab
presenter for the Low Temperature Laboratory during 2012-2016. This task involved presenting the laboratory, research done in LTL, and (low temperature) physics in general to the public. Visitors included a wide variety of groups from kindergarten children to university student guilds, as well as visiting researchers. During these visits I had the pleasure to work with several copresenters from helium research groups Petri Heikkinen, Niklas Hietala, and Juho Rysti as well as various people from the Brain Research Unit. This collaboration taught me a lot about subjects I would not have necessarily become so familiar with otherwise. It also challenged us to come up with ways to illustrate very complicated physics in a relatively short time to people from various backgrounds without losing their interest. Luckily, we had liquid nitrogen on our side.

I also thank the current members of Nano group for support Teemu Elo, Mohammad Tasnimul Haque, Jukka-Pekka Kaikkonen, Juuso Manninen, Jayanta Sarkar, Abhilash Thanniyl Sebastian, Xuefeng Song, Zhenbing Tan, Matti Tomi, Marco Will, Liu Ying, and Vladislav Zavyalov. I also had a pleasure working with and/or along with many people formerly affiliated with Nano group Daniel Cox, Aurélien Fay, Akira Hida, Pasi Häkkinen, Dmitry Lyashenko, Pasi Lähteenmäki, Mika Oksanen, Antti Puska, and Maciej Weisner. In addition I appreciate professional encounters relevant to this work with the following people Matthias Brandt, Sergey Danilin, Erno Damskägg, and Jaakko Sulkko. In addition interesting discussion were had with Ville Kauppila, Karthikeyan Sampath Kumar, Jere Mäkinen, Tapio Riekki, Jorge Tiago Santos, and Antti Vepäläinen.

In general, I thank everybody in the lab for pleasant and free atmosphere. I also appreciated all the activities outside work, such as summer barbecues. Special thanks for Sergey Danilin for all the sports activities that we did.

Lastly, I would like to thank my father Aki Laitinen who has always supported me with my way of doing things. My sister Annamari Laitinen also deserves thanks.

Otaniemi, Espoo, May 7, 2018,

Antti Laitinen
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This thesis consists of an overview and of the following publications which are referred to in the text by their Roman numerals.


List of Publications


Author’s Contribution

Publication I: “Electron-Phonon Coupling in Suspended Graphene: Supercollisions by Ripples”

The author conducted the measurements, analyzed the results, and wrote the paper with the other authors.

Publication II: “Coupling between electrons and optical phonons in suspended bilayer graphene”

The author fabricated the LOR-based samples, conducted the measurements, analyzed the results, and wrote the paper with the other authors.

Publication III: “Contact doping, Klein tunneling, and asymmetry of shot noise in suspended graphene”

The author conducted the measurements, analyzed the results, and wrote the paper with the other authors.

Publication IV: “Single-mode and multimode Fabry-Pérot interference in suspended graphene”

The author conducted the measurements with M.O., and analyzed the data with the other authors.

Publication V: “Ultra low $1/f$ noise in suspended bilayer graphene”

The author fabricated the LOR-based samples, conducted the experiments and
analyzed the data with M.K., and participated in writing the manuscript.

**Publication VI: “Unconventional fractional quantum Hall states and Wigner crystallization in suspended Corbino graphene”**

The author developed the technique for fabricating the samples, conducted the experiments and analyzed the data with M.K., and wrote the manuscript with the other authors.

**Publication VII: “Weak antilocalization of composite fermions in graphene”**

The author developed the technique for fabricating the samples, conducted the experiments and analyzed the data with M.K., and wrote the manuscript with the other authors.

**Publication VIII: “Gyrotropic Zener tunneling and nonlinear IV curves in the zero-energy Landau level of graphene in a strong magnetic field”**

The author developed the technique for fabricating the samples, conducted the experiments and analyzed the data, and wrote the manuscript with the other authors.

**Publication IX: “Breakdown of zero-energy quantum Hall state in graphene in the light of current fluctuations and shot noise”**

The author developed the technique for fabricating the samples, conducted the experiments and analyzed the data, and wrote the manuscript with the other authors.
Graphene, the hexagonal 2D allotrope of carbon, is the material of superlatives. Altogether, it one of the strongest, thinnest, lightest, and most conducting material even at zero charge density. These and many other extreme qualities [1] make graphene very promisingly advantageous material in the development of future technologies. Consequently, since the groundbreaking work of Novoselov and Geim in 2004 [2], a completely new field of research - 2D Materials and their heterostructures - has spawned. Although today’s commercial graphene applications are mostly limited to composite materials where graphene is just a reinforcing part, such as graphene-reinforced concrete [3], in the future graphene could be utilized in flexible/printable electronics [4], or even in quantum information processing [5].

In addition to the technological potential, graphene is of great interest for fundamental physics research since it hosts many peculiar physical phenomena that cannot be realized in conventional materials such as metals and semiconductors. The charge carriers in graphene have linear dispersion relation and they behave as massless Dirac fermions. Moreover, the charge carrier polarity (electrons/holes) can be tuned, for example by electrostatic gating. Due to these properties, researchers have been able to observe perfect transmission of injected charge carriers through potential barriers (Klein tunneling [6, 7]) and negative index of refraction (Veselago lensing [8]). Moreover, when a graphene flake is put in a magnetic field, the onset of the anomalous half-integer quantum Hall effect (QHE) is observed at very low fields below 5 mT in cleanest samples [9]. At high magnetic fields, the fractional quantum Hall effect (FQHE) [10], and Wigner crystallization can be observed, see Sec. 7.2 and Ref. [11].

While some phenomena characteristic to graphene are robust against disorder in the samples, many interesting physics, such as FQHE, are rather delicate and put strict requirements on the sample quality. In fact, improvement of the sample quality has been pivotal in graphene research during its brief history. Soon after the discovery of graphene, the limitations of the used standard nanofabrication substrate, silicon dioxide $SiO_2$, became evident: it was too rough and dopants were often present. Later, two major solutions to this problem have been developed: getting rid of the substrate (suspended graphene [12]) and
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encapsulation between hexagonal boron nitride (hBN) flakes [13]. While the first one remains the cleanest option, reaching closest to ideal graphene, the former option seems to be the direction where the field of graphene research is heading today, mainly because of its robustness and technological applicability. Suspended graphene remains as the more delicate option and not suitable for the majority of applications, but it still provides a good platform for the studies of innate properties of graphene.

In this work, suspended graphene devices were utilized for the study of fundamental properties of graphene at low temperatures down to \( T = 10 \text{ mK} \) and at high magnetic fields up to \( B = 9 \text{ T} \). The experiments were conducted using sophisticated low and high frequency noise measurement techniques as well as delicate conductance measurements. Thus, the design of the experimental installation incorporated careful filtering/blocking of unwanted noise as well as sufficient thermal anchoring of the measured devices in order not to destroy the elusive quantum effects.

The devices measured were state of the art suspended graphene samples. Two different geometries were utilized: the two-terminal rectangular bridge geometry, and the suspended Corbino geometry. Two different fabrication methods for the rectangular samples were used, but the results were similar with both of them. The suspended graphene Corbino devices were developed in this work, which is also the first report of such devices. They were specifically designed for high field magnetotransport measurements for probing the quantum Hall effects in graphene. The five first publications I-V present the work regarding the rectangular samples. The later four publications VI-IX deal with experiments where the Cobino devices were investigated.

In the zero magnetic field and the lowest temperatures down to \( T = 10 \text{ mK} \), rectangular suspended graphene devices were used to study the doping of graphene by the top contacts and Klein tunneling through the subsequently formed \( pn \) junctions at the edges. Additionally, reflection from the edge junctions and inevitable interference with other charge carriers led to Fabry-Pérot interference. These interference patterns were used to demonstrate the phase coherent transport and long mean free path in the measured devices.

The low frequency noise in suspended graphene was investigated in order to make sure that the upconverted \( 1/f \) noise would not be a problem with the shot noise measurements at microwave frequencies. The noise levels turned out to be extremely low, especially in bilayer graphene that was utilized for a gas sensing application based on added noise produced by adsorbed gases.

At higher frequencies, microwave shot noise measurements were used to determine the temperature of the electron system in graphene as a function of applied bias voltage. Since the biasing caused Joule heating that was carried to the thermal heat sinks through the phonon system, these measurements enabled the determination of electron-phonon coupling in mono and bilayer graphene. The particular physical processes were identified by comparing the obtained data to the theoretical models formulated in collaboration with
theoreticians. While bilayer graphene exhibited quite ordinary coupling to optical phonons, the electron-phonon coupling in monolayer graphene was found to be more complicated "supercollision" process involving an acoustic phonon and two flexural phonons.

In addition to rectangular graphene devices also measured by many groups, the unique suspended Corbino ring devices were studied at high magnetic fields up to $B = 9$ T. The Corbino geometry turned out to be great for observation of FQHE that was found to follow well the framework of the composite fermion theory developed by Jain [14]. The experimental setting allowed probing of the emergent composite particles, the composite fermions of graphene that were found to behave as Dirac particles with cyclotron mass close to one electron rest mass. Moreover, at the highest field available the conductance measurements exhibited evidence of the Wigner crystal phase that had not been reported in graphene before. In addition to these delicate phases, breaking of quantum Hall state at the filling factor $\nu = 0$ in the middle of the lowest Landau level was also investigated by measuring current-voltage characteristics and noise. The IV-characteristics revealed Zener tunneling between Landau sublevels, which was found to act as a precursor for the break down of QHE. At very high fields and high break down voltages, the dielectric breakdown displayed more conventional bootstrap-type electron heating that dominated the Zener tunneling mechanism.

This thesis is organized as follows. First, a brief introduction to graphene fundamentals regarding the subjects of this thesis is given. Second, the fabricated samples, experimental setup, measurement methods are reviewed. Third, the conducted transport experiments regarding Fabry-Pérot resonances and Klein tunneling in rectangular suspended graphene devices are covered. The next two chapters review the noise measurements on the rectangular samples. First, the $1/f$ noise (low frequency noise) studies with bilayer graphene are presented, followed by the electron-phonon coupling experiments (high frequency shot noise measurements). The final two chapters cover the experiments with suspended graphene Corbino samples in high magnetic fields, namely the observation of QHE and Wigner crystallization, as well as breaking of the quantum Hall state $\nu = 0$. The details about device fabrication and selected experimental techniques are given in the appendixes.
Introduction
2. Fundamentals of graphene

Carbon is the element with the atomic number \( Z = 6 \), meaning it has half-filled valence shell with four valence electrons on it. This makes carbon very versatile, it can form up to four bonds to different directions depending on the other atoms it is bonded to. Consequently, carbon often serves as the backbone for complex molecules, such as hydrocarbons important for modern society, and complex biomolecules on which life is based on. On the other hand, carbon is found in small inorganic molecules, such as carbon dioxide. Moreover, carbon is also capable of forming multiple different structures without other elements, these are known as allotropes of carbon. The most well-known allotropes are probably diamond and graphite.

An isolated single layer of graphite is called graphene. It is the two-dimensional allotrope of pure carbon where carbon atoms are arranged in hexagonal grid resembling chicken wire. The carbon atoms in graphene’s honeycomb lattice are \( sp^2 \) hybridized so that the an atom is connected to its three neighbors with
strong $\sigma$ bonds oriented in plane of the graphene sheet. This is what makes graphene incredibly strong and tough [16, 17]. The remaining $\pi$ electrons are free to move in the plane making graphene an excellent electric and thermal conductor.

As seen in Fig. 2.1, other $sp^2$ hybridized carbon allotropes can be derived from graphene by cutting and curling it. Fullerenes are just graphene cut and curled in to a spherical shape, and (single walled) carbon nanotubes, CNTs, are rolled up graphene sheets. When one stacks two monolayer graphene (MLG) sheets on top of each other bilayer graphene (BLG) is formed, and with three layers trilayer graphene (TLG). With more layers the structure is usually called few layer graphene (FLG), or if the number of layers is high ($N > 10−15$) the stack is called graphite. Although all these structures are derived form graphene, they have very different properties in many respects. Rest of this thesis will focus on mono and bilayer graphene that will be reviewed in more detail.

## 2.1 Electron transport in graphene

The structure of monolayer graphene is depicted in Fig. 2.2. Each atom in the lattice is connected to its three neighbors with three single covalent $\sigma$ bonds repelled $120^\circ$ apart in the plane of graphene sheet creating the hexagonal grid. The lattice can be divided into sublattices A and B in a way that every atom is surrounded by atoms from the opposite sublattice. The lattice constant of graphene is $a = 142$ pm, which is also the separation of the two sublattices. The lattice vectors $\mathbf{a}_1, \mathbf{a}_2$ define translations within the sublattices.

By making the standard transformation to the reciprocal lattice, the consequence of the two sublattices in real space becomes evident: two equivalent valleys $K, K'$ emerge at the edge of the Brillouin zone. These points are of special interest since there the valence and conduction bands touch enabling electronic conduction.

![Graphene lattice](image)

**Figure 2.2.** a) Graphene lattice with lattice constant $a$ and lattice vectors $\mathbf{a}_1 = a \left( \frac{3}{2}, \frac{\sqrt{3}}{2} \right)$ and $\mathbf{a}_2 = a \left( \frac{3}{2}, -\frac{\sqrt{3}}{2} \right)$ b) The inverse lattice with the inverse lattice vectors $\mathbf{b}_1 = \frac{2\pi}{a} \left( \frac{1}{3}, \frac{1}{\sqrt{3}} \right)$ and $\mathbf{b}_2 = \frac{2\pi}{a} \left( \frac{1}{3}, -\frac{1}{\sqrt{3}} \right)$ c) The low energy approximation of the graphene band structure near the $K$-point where the valence and conduction bands touch.
By following Ref. [18], the electron transport characteristics can be derived from the graphene structure by considering hopping between the graphene sublattices. One can write down the tight binding Hamiltonian for monolayer graphene as:

\[ H = -t \sum_{<i,j>_{\sigma}} (a_{\sigma,i}^\dagger b_{\sigma,j} + h.c.) - t' \sum_{<i,j>_{\sigma}} (a_{\sigma,i}^\dagger a_{\sigma,j} + b_{\sigma,i}^\dagger b_{\sigma,j} + h.c.), \] (2.1)

where \( a_{\sigma,i}, a_{\sigma,i}^\dagger \) are annihilation and creation operator for electrons with spin \( \sigma \) in sublattice A, \( b_{\sigma,i}, b_{\sigma,i}^\dagger \) the corresponding operators for sublattice B, and the sums with the indices \( i \) and \( j \) run over all lattice sites. \( t \approx 2.8 \text{ eV} \) and \( t' \approx 0.2t \) represent hopping to the nearest and the next-nearest neighbors, respectively.

One can calculate the band structure of this tight binding Hamiltonian in the limit of nearest neighbor hopping by linearizing it with suitable operator replacements and then finding the eigenvalues, as demonstrated in Refs. [18, 19]. The eigenvalues are:

\[ E(k) = \pm t \sqrt{3 + 2 \cos(\sqrt{3} k_y a) + 4 \cos \left( \frac{\sqrt{3}}{2} k_y a \right) \cos \left( \frac{3}{2} k_x a \right)}, \] (2.2)

where \( k = (k_x, k_y) \) is the 2D wave vector.

One can easily verify that \( E(k) = 0 \) at the Dirac points: \( K, K' = \left( \frac{2n}{3\sqrt{3}a}, \pm \frac{2n}{3\sqrt{3}a} \right) \). Consequently, the expression in Eq. 2.2 can be linearized in the vicinity of the Dirac points by considering new wave vectors of form \( q = k - K \). The original Hamiltonian can also be linearized around the Dirac point and expressed in terms of the new wave vectors \( q \), as given in [18]:

\[ \hat{H}_{K,K'}(q) = \hbar v_F \begin{pmatrix} 0 & q_x \mp i q_y \\ q_x \pm i q_y & 0 \end{pmatrix}, \] (2.3)

where \( v_F = \frac{3ta}{2} \approx 10^6 \text{ m/s} \) is the Fermi velocity near the Dirac point, \( \hbar \) is the reduced Planck constant, and the optional signs correspond to different Dirac points \( K \) and \( K' \).

If one transforms the wave vectors into operators, e.g. \( q_x \to -i \frac{\partial}{\partial x} \), and includes a spatially variable potential \( V(\mathbf{r}) \), one can write down the equivalent of the Schrödinger equation for massless Dirac particles: the Dirac equation. At the \( K \) point it takes the form:

\[ [-i\hbar v_F \sigma + V(\mathbf{r})] \cdot \nabla \Psi(\mathbf{r}) = E \Psi(\mathbf{r}), \] (2.4)

where \( \sigma \) is the Pauli spin matrix where only \( x \) and \( y \) components are included. At the other Dirac point, the Hamiltonian is \( \hat{H}_{K'} = \hat{H}_{K}^T \).

The solutions for Eq. 2.4, neglecting the potential \( V(\mathbf{r}) \), at \( K \) and \( K' \) are given in momentum space in Ref. [18] as:

\[ \Psi^{(K)}(q) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\theta_q/2} \\ \pm e^{i\theta_q/2} \end{pmatrix}, \quad \Psi^{(K')}(q) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{i\theta_q/2} \\ \pm e^{-i\theta_q/2} \end{pmatrix}, \] (2.5)
where ± marks conduction and valence bands, respectively, and $\theta_q$ is the angle of the wave vector defined as:

$$\theta_q = \arctan \left( \frac{q_x}{q_y} \right),$$

(2.6)

where $q_x$ and $q_y$ are the wave vector components around the given Dirac point. The two-component nature of the graphene wave function stems from the division of the lattice into the two sublattices. In fact, the two components reflect the distribution of electrons on the sublattices, e.g. with a spinor $\begin{pmatrix} 1 \\ 0 \end{pmatrix}$ the wave function reside solely on the sublattice A. This quality of the wave function is called the pseudospin. The name comes from the binary nature of the spinor wave functions and the fact that the pseudospin acts like a spin in some respects [20].

Additionally, the eigenvectors in Eq. 2.5 reflect the Dirac nature of electrons in graphene. If one rotates the wave vector $\mathbf{q}$ around $2\pi$, i.e. the electron makes a loop, the wave function acquire an additional phase factor of $\exp(i\pi)$. This is a consequence of the Berry phase being $\pi$ in graphene [21]. This a non-trivial property of the wave function that is connected to the electron transport in many ways, such as interference phenomena, the half integer quantum Hall effect [22], and weak antilocalization [23].

The corresponding eigenvalues of the Hamiltonian of Eq. 2.3 are:

$$E(\mathbf{q}) = \pm \hbar v_F |\mathbf{q}|,$$

(2.7)

where ± corresponds to electrons and holes, see also illustration in Fig. 2.2. This solution has many implications. For example, the linear dispersion is a signature of massless Dirac particles, contrary to the fundamentally different quadratic form $E(k) = \frac{\hbar^2 k^2}{2m^*}$ typically found in semiconductors. Typically the linear dispersion relation is associated with massless particles moving at the speed of light, i.e. photons. Indeed, in some respects the relativistic electron system in graphene is analogous to photons, with the Fermi velocity $v_F$ substituting the speed of light $c$.

### 2.1.1 Bilayer graphene

In bilayer graphene, two graphene layers lay on top of each other bound together by relatively weak $\pi$ bonds. In fact, the weakness of these bonds allows isolation of single layer graphene by mechanical exfoliation from graphite in multiple ways [24]. Additionally, the orientation of the two layers in bilayer graphene is not strictly fixed. Most commonly bilayer graphene is found in the thermodynamically stable AB-stacked form (Bernal-stacking), where the upper layer atoms are positioned in the middle of the lower layer hexagons, as seen Fig. 2.3a. The layers can also be AA-stacked where the atoms in different layers are aligned, or the layers can be rotated with respect to each other (twisted bilayer graphene). The offsets and rotations can have complex effects on the
transport properties of bilayer graphene. As presented in Ref. [25] and Fig. 2.3b, the AA-stacked bilayer graphene should have a small gap. With twisted BLG, superconducting transition has been observed [26]. However, since the rotated and AA-stacked bilayer graphene both are less likely to appear naturally, see Ref. [27], they are not considered in this work.

\[ H_K(q) = \frac{\hbar^2}{2m^*} \begin{pmatrix} 0 & (\hat{q}_x - i\hat{q}_y)^2 \\ (\hat{q}_x + i\hat{q}_y)^2 & 0 \end{pmatrix}, \] (2.8)

where \( m^* \approx 0.03m_e \) is the effective electron mass (\( m_e \) is the electron rest mass), and \( q \) is the wave vector in the vicinity of the Dirac points \( K, K' \).

The corresponding eigenvalues of Eq. 2.8, i.e. dispersion relation for the bilayer graphene, are:

\[ E_{e,h} = \pm \frac{\hbar^2 q^2}{2m^*}, \] (2.9)

where the \( \pm \) denotes electrons and holes, respectively. Essentially, bilayer graphene is a zero band gap semiconductor where the valence and conduction band touch at \( q = 0 \).

The eigenstates of the Hamiltonian in Eq. 2.8 around \( K \) point are:

\[ \Psi_{\pm}^{(K)}(q) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\theta q} \\ \pm e^{i\theta q} \end{pmatrix}, \] (2.10)

where \( \pm \) denotes electrons and holes.

It has been shown that the electrons in bilayer graphene are massive Dirac particles with Berry’s phase \( 2\pi \) [29], thus, bilayer graphene is more than a regular semiconductor. The charge carriers in bilayer graphene behave as chiral massive Dirac particles, consequently, bilayer graphene exhibits many nontrivial...
transport properties similar to the ones seen in monolayer graphene, such as chiral tunneling through \textit{pn} barriers [6]. On the other hand, for example instead of weak antilocalization, more conventional weak localization is expected [30].

2.1.2 \textit{pn} junctions and Klein tunneling

A \textit{pn} junction is an interface between two regions with opposite types of doping, electrons and holes. In conventional semiconductor devices, the doping in the different regions is usually achieved by implanting ions, such as boron (donor) or phosphorous (acceptor), into neutral bulk semiconductor, such as silicon. Consequently, the characteristics of the junction are mostly fixed during fabrication.

In case of graphene, doping can be tuned to both electrons and holes, and thus \textit{pn} junctions can be formed into otherwise neutral graphene simply by gating. This makes graphene \textit{pn} junctions tunable and potentially dynamic. Additionally, graphene can also be doped by dopants that are brought in contact with it. In dirty on-substrate samples, the whole graphene sheet is typically riddled with \textit{pn} junction puddles that are formed because of doping residues left by the fabrication process [31]. Moreover, even in the cleanest samples the metallic contacts cause doping due the fact that work functions are different in neutral graphene and most metals. Thus, \textit{pn} junctions are usually inherently present in graphene devices.

\textbf{Figure 2.4.} Klein tunneling through graphene \textit{pn} junctions (picture adapted from Ref. [6]): a) Dirac cones at three different regions in an \textit{n}p\textit{n} doping structure, the dashed line denotes the Fermi level. b) Structure of the potential step in the panel a, \textit{q} denotes the wave vector of incoming electrons. c) Transmission through 100 nm wide barrier presented as a function of the incident angle of the wave vector \textit{q} for two different charge densities in the \textit{p} region \(10^{12}\ \text{cm}^{-2}\) and \(3 \times 10^{12}\ \text{cm}^{-2}\), the density in the \textit{n}-regions is \(0.5 \times 10^{12}\ \text{cm}^{-2}\) in both cases.

Transmission through graphene \textit{pn} junctions can be calculated by considering
the electron wave functions in monolayer graphene presented earlier. In the situation presented in Fig. 2.4b where there is a barrier $V_0$ in region II with opposite doping than in the bordering regions (sections I and III), the wave functions can be written as:

\[
\begin{aligned}
\psi_I(x, y) &= \frac{1}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} e^{i\theta} \right) e^{iq_x x + q_y y} + \frac{r}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} e^{i(\pi - \theta)} \right) e^{-iq_x x + q_y y}, \quad x < 0 \\
\psi_{II}(x, y) &= \frac{a}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} e^{i\theta} \right) e^{iq'_x x + q'_y y} + \frac{b}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} e^{i(\pi - \theta)} \right) e^{-iq'_x x + q'_y y}, \quad 0 < x < D \\
\psi_{III}(x, y) &= \frac{t}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} e^{i\theta} \right) e^{iq_x x + q_y y}, \quad x > D,
\end{aligned}
\] (2.11)

where an additional trivial phase factor $\exp(i\theta/2)$ has been introduced, this gauge transformation simplifies the equations slightly. The sign factors are $s = \text{sgn}(E)$ and $s' = \text{sgn}(E - V_0)$. The factors $a, t$ denote the amplitudes of the transmitted wavefunctions in sections II and III, whereas the factors $b, r$ denote the amplitudes of the reflected wavefunctions in sections I and II. The angle $\phi = \arctan(q_y/q_x)$ is the incidence angle of electrons in section I, and the other angle $\theta = \arctan(q'_y/q'_x)$ is the angle of refraction in section II. Note that if the charge densities in sections I and II are not the same, then $\phi \neq \theta$. In fact, by tuning the charge densities in the proper way, even negative angles of refraction can be achieved. This effect has been utilized in electron optics experiments to realize Veselago lensing in graphene [8].

The transmission coefficients as a function of incident angle $\phi$ can be calculated from Eqs. 2.11 by requiring the wave functions to be continuous at the border of different segments [6]. The transmission through the barrier in the high barrier limit $V_0 >> E$ is:

\[
T(\phi) = \frac{\cos^2 \phi}{1 - \cos^2(q_x D) \sin^2 \phi}.
\] (2.12)

The transmission as a function of the incident angle is presented in Fig. 2.4. At $\phi = 0$, transmission is perfect. Even more peculiar is that this is strictly true only when the barrier is infinitely high. This is totally different from conventional systems where the transmission will decay exponentially inside the barrier, ever faster when the barrier height and width are increased [32]. In the case of graphene, the perfect transmission is facilitated by turning an incident electron coming from region I into a hole inside the barrier and then back into an electron in region III. This behavior is more commonly known as Klein tunneling. Note that the momentum of the hole inside the barrier points to the opposite direction $w.r.t.$ the incident electron wave vector, however, as shown in Ref. [18], the group velocity always remains in the same direction and current is conserved. In other words, the direction of the moving holes is also in line with the conservation of charge.

### 2.1.3 Weak localization and antilocalization

Another way to demonstrate the Dirac nature of charge carriers in graphene is to look at interference effects, such as weak localization. Weak localization is an interference effect caused by weak disorder in the electronic system [33,
34]. When an electron trajectory makes a loop due to disorder scattering, as illustrated in Fig. 2.5, the electron wave function acquires a phase. Depending on this phase, the interference of the electron wave function with itself can be constructive or destructive. Experimentally, this interference effect can be seen for example in magnetoconductance measurements around zero magnetic field, where constructive interference leads to a resistance peak at exactly zero magnetic field. Correspondingly, decreased resistance (a dip) is observed with destructive interference. However, even a small magnetic field of the order of 10 mT destroys these interference effects due to breaking of time reversal symmetry in the system.

![Electrons trajectories due to disorder leading to quantum interference effects: weak localization and weak antilocalization](image)

**Figure 2.5.** Electrons trajectories due to disorder leading to quantum interference effects: weak localization and weak antilocalization, picture adapted from Ref. [35]

As discussed earlier, in monolayer graphene the phase acquired when rotating the wave vector once around, \( i.e. \theta_q \to \theta_q + 2\pi \), results in the phase factor \( \exp(i\pi) \) in the wave function. This signifies destructive interference in case of trajectories described above, \( i.e. \) weak antilocalization is expected to occur [30]. This happens also in other materials housing Dirac particles, such as topological insulators (TI), see Ref. [34]. In case of bilayer graphene, the phase acquired while \( \theta_q \to \theta_q + 2\pi \) is \( \exp(i2\pi) = 1 \), and more regular weak localization will occur instead.

### 2.2 Noise

Noise is an inevitable phenomenon in almost all electronic systems. Typically, noise is a nuisance that one tries to eliminate, or at least suppress. One example of this is the reduction of thermal noise by cooling down the investigated device and/or the measurement system. However, in some cases useful information can be extracted with careful analysis. In the following sections different noise types relevant to this work are reviewed.
2.2.1 Switching noise

Switching noise, also known as random telegraph noise or burst noise, is a type of noise where the measured signal jumps between two levels at random times with some characteristic switching rate $1/\tau$. This type of noise is typical for systems that are unstable in one way or another, e.g., a broken cable. In Fig. 2.6a there is a schematic of switching noise pulses with a duration $\tau_s$ separated by the characteristic time $\tau_0$.

The spectrum for this type of noise in case of a single time scale $\tau$ dominates current switching between 0 and $I_0$ is given in Ref. [36] as:

$$S_I(\omega) = \frac{4\tau}{1+\omega^2\tau^2} \langle I \rangle^2,$$

where $\langle I \rangle$ is the average current through the conductor, and $\omega$ is the frequency.

Lorentzian spectra of Eq. 2.13 for different $\tau$ are presented in Fig. 2.6b. Clearly, the spectrum is flat at low frequencies and fall as $1/\omega^2$ above the cutoff frequency. The cutoff is approximately $\omega_c = \frac{1}{\tau}$.

2.2.2 $1/f$ noise

In a real system, there typically are many fluctuators that have different time scales $\tau$, and the whole noise spectrum at low frequencies is a sum of the individual Lorentzian spectra. One can obtain the spectrum by integrating Eq. 2.13 over all relaxation times $\tau$ assuming distribution of form $1/\tau$, as has been schematically illustrated in Fig. 2.7 (note the loglog-scale in the figure). It can be seen that an uniform collection of fluctuator spectra will indeed lead to $1/f$ spectrum since every fluctuator will have their main contribution to the spectrum near the corner frequency. At frequencies lower that the corner frequency, the amplitude of the fluctuator is too low to contribute significantly, and at frequencies above the corner frequency the noise amplitude fall as $1/f^2$, which is faster than the total noise that decays as $1/f$.

However, the integration over the fluctuators requires some assumptions, in particular on the distribution of fluctuator time scales and amplitudes, and
hence different models have been developed. Perhaps the most successful has been the so-called McWhorter’s model [37]. This model was originally invented to describe 1/f noise in germanium field effect transistors (FET), where the oxide layer separating the conduction channel and the gate houses charge traps that fluctuate with different time constants $\tau$. By assuming a $1/\tau$ distribution and thermally activated trap occupancy $\tau \propto \exp(E/k_B T)$, where $E$ is the activation energy, McWhorter formulated the model that yields 1/f spectrum, which was observed in the experiments.

![Figure 2.7](image_url)

**Figure 2.7.** Conceptual drawing of generation of 1/f noise spectrum, where spectra of individual fluctuators from Eq. 2.13 with different $\tau$ (blue curves) have been plotted in the same picture on a loglog-scale. The 1/f spectrum (red dashed line) is formed as the edge of the area that this ensemble of fluctuator spectra takes.

Instead of fluctuations in the total number of charge carriers, the low frequency noise can also be dominated by electron mobility fluctuations [38]. This mechanism is based on fluctuation of the impurity scattering cross-sections of the scattering centers. The charge carriers feel these fluctuations, but the charge carrier density itself does not fluctuate. Instead, the fluctuations are seen in the measured quantity (voltage, current, or resistance) through electron mobility.

In addition to McWhorter’s model and mobility fluctuations that give physical explanation of the origin of 1/f noise, the 1/f noise is usually compared to Hooge’s empirical formula [39] that is usually used to quantitatively characterize the amount of noise:

$$S_I = A_n \frac{I^\gamma}{f^\beta},$$  \hspace{1cm} (2.14)

where typically $\gamma \approx 2$ and $\beta \approx 1$. The parameter $A_n$ is the noise amplitude that can be used to compare the amount of 1/f noise in different materials.

While Hooge’s empirical relation and total carrier number fluctuations have been successful in describing 1/f noise in many semiconductor materials, there are situations where they do not work properly. One obvious one of these situations is ballistic transport with perfect transmission without scattering, where one could expect no noise from the conducting channel itself. However, typically some contact noise is always present since contacts can be dirty and house charge traps. In experiments this contact noise should be easy to distinguish
from the noise originating from the conduction channel, since it should not depend on charge carrier density that may not be changed by the gate due to the doping and consequent Fermi level pinning by the contact, see Sec. 4.1 for further details.

2.2.3 Shot noise

Shot noise is a type of noise generated by discrete nature of charge carriers. The spectrum of shot noise has been calculated in Ref. [40] by considering the average current in a system given by the ratio of the elementary charge transported through $e$, and the average time between carrier arrivals $\tau$: $\langle I \rangle = \frac{e}{\tau}$.

It can be shown that the noise spectral density of the shot noise in this current is:

$$ S_I = 2Fe \langle I \rangle. \quad (2.15) $$

Note also that $S_I$ doesn’t depend on frequency, i.e. the spectrum is flat. The prefactor $F$ is the so called Fano factor defined in Ref. [41] as the ratio between shot noise $S_I$ and fully random Poissonian noise $S_p$ as $F = S_I/S_p$. Thus, it takes into account correlations between electrons.

Generally, three regimes different from $F = 1$ can be identified: super-Poissonian $F > 1$, sub-Poissonian noise $F < 1$, and noiseless transport regime $F = 0$. The super-Poissonian noise essentially implies electron bunching, seen for example in avalanche diodes. In the sub-Poissonian regime, the noise is reduced due to correlations between electrons. This happens in many systems at sufficiently low length scales. If the length of the system is made short enough to reach the ballistic transport regime, the shot noise goes to zero, because without scattering/tunneling $F = 0$.

The Fano factor in monolayer graphene can be calculated using the Landauer-Büttiker formalism [41], where one considers the electrical transport through the 2D graphene sheet as a sum of 1D transmission channels. This problem was solved in Ref. [42] for rectangular graphene sheet with length $L$ and width $W$. In the limit of wide and large graphene sheet, the 1D transmission coefficients are:

$$ T_n = \left| \frac{q_n}{q_n \cos(q_nL) + i(\mu/\hbar v_F)\sin(q_nL)} \right|^2, \quad (2.16) $$

where the longitudinal wave vectors (including the chemical potential shift $\mu$ due to carrier doping, and $\mu = 0$ contribution $q_n^0$) are $q_n = \sqrt{(\mu/\hbar v_F)^2 - q_n^2}$, and $n$ is an index running through all the $N$ channels.

Using these transmission coefficients, one can calculate the conductance and the Fano factor using the standard Landauer-Büttiker formulas:

$$ G = \frac{4e^2}{h} \sum_{n=0}^{N-1} T_n, \quad (2.17) $$
Note that the conductance per channel equals the conductance quantum $G_0 = \frac{4e^2}{h}$, and for ballistic conduction one would have $T = 1$, which yields exactly $F = 0$, as discussed earlier.

As shown in Ref. [42], when plugging in the transmission coefficients into the equations of $F$ and $G$, pseudodiffusive transport in graphene is obtained with the conductivity $\sigma = \frac{4e^2}{\pi h}$ and the Fano factor $F = 1/3$ in the limit of wide graphene sheet $W/L \rightarrow \infty$ at the Dirac point. The term “pseudodiffusive” comes from the fact that in the obtained value for the Fano factor, $F = 1/3$ at the Dirac point, is the same as that measured in the diffusive transport regime in sufficiently short metallic samples at low temperatures [43]. In these samples, the Fano factor actually depends on the relaxation length scales $w, r, t$. the sample length, see Fig. 2.8.

If electron heating occurs, one enters into the so called hot electron regime, where the Fano factor becomes modified [44]. In this case, the value of $F = \frac{\sqrt{3}}{4}$ is expected, see Fig. 2.8. This happens in samples where the electron-electron scattering length is shorter than the sample length. In even longer samples, the electron phonon coupling reduces the Fano factor towards zero. Experimentally, all these regimes can in principle be seen in the same sample of fixed length $L$ by changing the scattering lengths by biasing.

**Figure 2.8.** Fano factor in different transport regimes in short metallic samples (picture taken from Ref. [43]). In sufficiently short samples $F \rightarrow 0$ due to ballistic transport, and in longer samples that are still shorter than the electron-electron scattering length $L_{e-e}$ one has $F = 1/3$. The length scale $L_\phi$ denotes the phase coherence length. If the sample length further increased from $L_{e-e}$, the hot electron regime with $F = \frac{\sqrt{3}}{4}$ is reached. Increasing the sample length above the electron phonon scattering length $L_{e-ph}$ will result in decreasing Fano factor, $F \rightarrow 0$.

**Corbino geometry**

Similarly to the case of rectangular graphene sheets, the Fano factor and conductivity can be calculated for Corbino ring samples where the graphene is ring shaped and contacted from the inner and outer edges, see Fig. 2.10 for details.
This has been done in Ref. [45], and the conductance and the Fano factor take the forms:

\[ G = \frac{s e^2}{\hbar} \sum_{n = \frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \ldots} T_n, \quad F = \frac{\sum_{n = \frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \ldots} T_n(1 - T_n)}{\sum_{n = \frac{1}{2}, \frac{3}{2}, \frac{5}{2}, \ldots} T_n}, \] (2.19)

where \( s = 8 \) and \( T_n \) is the transmission coefficient of the \( n \)th channel. Transmission coefficients are given by

\[ T_n = \frac{16}{\pi^2 q^2 R_1 R_2} \left( D + n \right)^2 + \left( D - n \right)^2, \] (2.20)

with

\[ D_n = \Im \{ H^{(1)}_{\nu}(i n) - \frac{1}{2} (q r_1) H^{(2)}_{\nu}(n) \pm H^{(1)}_{\nu}(i n) + \frac{1}{2} (q r_1) \}, \]

where \( H^{(K)}_{\nu}(x) \) is the \( K \)th Hankel function, \( r_i \) and \( r_o \) are the inner and outer radii of the Corbino ring, and \( q \) denotes the wave vector.

In the case of Corbino geometry, the Fano factor and conductance can be reduced to the limits of narrow and wide ring, but neither is relevant for the experiments conducted in this work. However it is useful to determine the conductivity of the device with certain ratio of radii, as shown in Ref. [45]:

\[ \sigma = G \frac{2\pi}{\log \left( \frac{r_o}{r_i} \right)}, \] (2.21)

where \( G \) is the conductance of the device.

### 2.3 Quantum Hall effect in graphene

Perpendicular magnetic field \( B \) can be taken into account in the Dirac equation by replacing the momentum operators \(-i\hbar \nabla → -i\hbar \nabla - eA\), where \( A = (0, Bx, 0) \) is the vector potential in the Landau gauge. By making this substitution and taking the operator choice given in Ref. [18], the Dirac equation reduces to the form:

\[ \hbar \omega_c \begin{pmatrix} 0 & \hat{b}^\dagger \\ \hat{b} & 0 \end{pmatrix} \psi = E \psi, \] (2.22)

where \( \hbar \omega_c \) is the cyclotron energy quantum, and \( \hat{b}, \hat{b}^\dagger \) are the new normalized creation and annihilation operators with \([ \hat{b}, \hat{b}^\dagger ] = 1\).

The eigenvalues of the Hamiltonian of Eq. 2.22 are the Landau levels in graphene:

\[ E_n = \pm \sqrt{2 \hbar c \mu^2 |n| B \sqrt{N}} = \pm \epsilon_n \sqrt{N} \] (2.23)

where \( n \) is the charge carrier density, \( N \) is the Landau level index, and the \( \pm \) denotes electrons and holes, respectively.

This energy spectrum differs from the conventional type \( E_n = \frac{\hbar e B}{m^*} \left( N + \frac{1}{2} \right) \) found for example in GaAs two-dimensional electron gas (2DEG). In graphene,
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Figure 2.9. a) Landau levels in graphene, the cones denote graphene dispersion relation around $K$-point in the zero magnetic field and the rings mark the Landau levels forming when $B \neq 0$, image taken from Ref. [46]. b) Resistivity quantization in monolayer graphene as a function of charge carrier density $n$ (image from Ref. [47]), where the green line marks the diagonal resistivity $\rho_{xx}$, and the red line denotes the Hall conductivity $\sigma_{xy}$ in the units of conductance quantum $4e^2/h$. The inset displays the Hall conductivity in case of bilayer graphene.

the spectrum is not evenly spaced, and the lowest Landau level resides at zero energy. As discussed in Sec. 2.1, these differences stem from the fact that the charge carriers in graphene are massless Dirac particles.

**Degeneracy of the quantum Hall states and the conductivity matrix**

At high magnetic fields, the charge carriers undergo cyclotron motion due to the Lorentz force. The radius of the guided center motion of an electron in such state is called the magnetic length $\ell_B$. It can be estimated using the Heisenberg uncertainty principle as the radius of the smallest allowed round orbit, as demonstrated in [48]:

$$\ell_B = \sqrt{\frac{\hbar}{eB}}.$$  \hspace{1cm} (2.24)

Thus, the electron will occupy an area of $A_0 = 2\pi \ell_B^2$. In fact, this area $A_0 = 2\pi \ell_B^2$ is the area penetrated by a single flux quantum $\Phi_0 = h/e$ of magnetic flux, which means that the number of states allowed per unit area can be written as:

$$n_L = \frac{1}{2\pi \ell_B^2}.$$  \hspace{1cm} (2.25)

The degeneracy of the quantum Hall states is given as the ratio of number of electrons $N_e$ to the total number of states $N_L$: $\nu = N_e/N_L$, where $\nu$ is the filling factor. Given the charge carrier density $n$, one can write down the commonly used expression:

$$\nu = \frac{n}{n_L} = \frac{nh}{eB}.$$  \hspace{1cm} (2.26)
The formation of the Landau levels will also result in quantization of certain components of conductivity (or resistivity) to specific values, as first reported by von Klitzing in Ref. [49]. Since the current and electric field vectors are not generally aligned in magnetic field, the conductivity $\sigma$ in the Ohm’s law $J = \sigma E$, where $J = (J_x, J_y)$ and $E = (E_x, E_y)$, is actually a tensor:

$$\sigma = \begin{pmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{pmatrix}, \quad (2.27)$$

where the Hall conductivity components $\sigma_{xy}, \sigma_{yx}$ are the ones displaying quantization, i.e. $\sigma_{xy} = \nu e^2/h$. On the other hand, the diagonal components $\sigma_{xx}, \sigma_{yy} \to 0$ as the Hall current $J_y \to 0$, see chapter 10.7 in Ref. [50] for details.

In graphene with the half integer quantum Hall effect, the edge states have conductivity:

$$\sigma_{xy} = 4 \left( N + \frac{1}{2} \right) \frac{e^2}{h}. \quad (2.28)$$

This quantization is specific for monolayer graphene and different from the conventional 2DEG form $4N \frac{e^2}{h}$. It has been experimentally confirmed with graphene Hall bar device, see Fig. 2.9b and Ref. [47]. Similarly, the conventional sequence has been confirmed for the bilayer graphene, apart from the missing $N = 0$ state at the Dirac point [51].

**Corbino geometry**

In the case of a specific geometry, the measured conductance $G$ can be related to the conductivity components given by the conductivity matrix. Here, the Corbino geometry is considered since the magnetoconductance measurements in this work were carried out on Corbino devices.

In the Corbino geometry, one can only apply a radial electric field $E_r$ and measure the produced radial current density $J_r$, as illustrated in Fig. 2.10a. By using Ohm’s law $J = \sigma E$, one can write down the currents in the radial and azimuthal directions in the narrow ring limit $r_o \sim r_i$, where one can consider $E_r$ to be uniform and $E_\theta = 0$. Thus, one can substitute $E_x \to E_r$ and $E_y \to E_\theta$, and obtain the following relations:

$$J_r = \sigma_{xx} E_r + \sigma_{xy} E_\theta = \sigma_{xx} E_r \quad (2.29)$$

$$J_\theta = \sigma_{yx} E_r + \sigma_{yy} E_\theta = \sigma_{xy} E_r, \quad (2.30)$$

where the second equalities were obtained by using $E_\theta = 0$, since no azimuthal electric field exists.
With the above expressions one can conclude that:

\[
\sigma_{xx} = \frac{J_r}{E_r} = \frac{I_r \log(r_0/r_i)}{V_r 2\pi} = G \frac{\log(r_0/r_i)}{2\pi},
\]

(2.31)

where \(I_r\) is the tunneling current through the device, \(V_r\) the applied voltage, and the second equality is obtained using the conformal mapping presented in Ref. [45].

Thus, the measurements in the Corbino geometry probe the diagonal conductivity \(\sigma_{xx}\) that in this case describes the tunneling transport through the ring, and not the edge state conductance quantization. Direct measurement of \(\sigma_{xy}\) is not possible since it is a property of the edge states that are localized at the edges of the Corbino ring. These counter rotating edge states carry current \(J_0 = \sigma_{xy} E_r\), where \(\sigma_{xy}\) is quantized in the regular manner. The schematic view of the edge states is presented in Fig. 2.10b.

### 2.4 Electron-electron interaction effects in the quantum Hall regime

Regardless of the geometry used in measurements, the quantum Hall effect is inherently a single particle phenomenon, i.e. all possible electron-electron interactions are neglected. Similarly, spin and valley symmetries are assumed to hold.

While this is the case at low magnetic fields and in impure samples, additional states corresponding to the breaking of these symmetries are developed inside Landau levels at large fields in high mobility samples. The valley and spin breaking leading to the so called "broken symmetry" states \(\nu = 0, \pm 1, \pm 3, \pm 5, \ldots\) have been observed in monolayer graphene [52]. These states can have complicated ordering related to the symmetry being broken. For example the \(\nu = 0\)
state in monolayer graphene is believed to have canted antiferromagnetic (CAF) spin ordering [53].

At even higher magnetic fields and lower disorder, electron-electron interactions (Coulomb energy) starts to dominate over the kinetic energy of electrons, and correlated states emerge [54]. One such phenomenon is the fractional quantum Hall effect (FQHE) where liquid-like phase is formed due to suitable fluctuations [55, 56]. Formally, this phenomenon is explained by emergence of new particles "composite fermions", formulated by Jain [14]. This approach has been very successful in explaining most of the experimental data in various semiconducting systems, and graphene, for details see chapter 7.

At very low charge densities corresponding to filling factors $\nu \lesssim 0.2$ the fluctuations leading to FQHE may be overpowered by a tendency to solid ordering, i.e. formation of Wigner crystal [56]. This competition between the liquid (FQHE) and solid (Wigner) phases has been probed extensively in GaAs-systems [57], while definitive proof of the Wigner phase has been lacking in graphene. Further discussion in the context of suspended graphene Corbino samples is presented in chapter 7.
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3. Experimental methods

This work was experimental research that involved fabrication graphene nano-electronic devices, precise control of experimental conditions, and delicate measurements probing quantum transport properties in the fabricated samples. In this section, the experimental details regarding this work are reviewed. The first part discusses different types of devices used in the experiments, as well as their basic properties. In the second part, the experimental setup and the measurement schemes are discussed.

3.1 Suspended graphene samples

3.1.1 Doubly-clamped bridge geometry

The most employed geometry for suspended graphene samples has been the doubly-clamped bridge where the graphene flake is contacted from the top by metallic leads that support and border the rectangular suspended part. Devices of the order of 1 μm in length can be made routinely. More complicated geometries have also been used, such as the Hall bar geometry [58]. However, they all suffer from the fact that current density in the graphene sheet is not uniform when biased. This reduces the effectiveness of current annealing, i.e. cleaning of graphene by passing a current of the order of 1 mA per 1 μm of sample width through it, since parts with lower current density are not heated sufficiently during the process.

Many methods have been invented to fabricate rectangular two-terminal suspended graphene devices: etching part of the SiO$_2$ substrate away [59], direct exfoliation on ready-made leads [60], dry stamping on ready-made leads [61], stamp transfer with PMMA support [62], wafer-scale wet-transfer on a hole using a PMMA membrane [63], selective removal of PMMA support [64], and selective removal of supporting lift-off-resist (LOR) [58]. The two most popular ones with high quality samples were used in this work: suspension by hydrofluoric acid etching (the HF method), and suspension by removing lift-off resist.
under graphene using electron beam (the LOR method).

The HF method

The HF etched samples were fabricated in a manner described in Ref. [65]. First, the graphene flakes that were mechanically exfoliated on a highly doped silicon substrate with 300 nm of thermally grown silicon dioxide on the top. In addition to serving as the dielectric between the $Si^{++}$ back gate and graphene, the oxide also facilitated the optical interference that rendered graphene visible on $SiO_2$ [66]. Then, the current leads were deposited using electron beam lithography and metal evaporation in ultra-high vacuum. During this step the top contacts made of triple layer metal Au/Cr/Au (7/5/50 nm) were deposited. The micron-scale contacts were connected to the on-substrate bonding pads deposited along the fine contacts. Finally, the graphene flake was released from the substrate by etching $\sim 150$ nm of the $SiO_2$ away by immersing the sample in buffered hydrofluoric acid for $\sim 1$ min. The etching was stopped by immersing the sample in water and subsequently taking it into isopropanol bath. In order to minimize the surface tension caused by the passing liquid surface, the isopropanol bath was heated to 50 °C as the chip was lifted from the liquid. Alternatively, a critical point dryer could have been used, however, this method was found lead to dirtier end product.

![Figure 3.1.](image.png)

**Figure 3.1.** a) Schematic drawing of a sample fabricated using the HF method. the black part hanging in the middle denotes graphene, yellow parts are the metal leads, the blue bottom parts denote the substrate, and the black lines are wire bonds connecting the bonding pads to the measurement lines. b) SEM image of a graphene sample suspended using HF-etching, the graphene sheet has been colored green, the Au/Cr/Au contacts appear as yellow, and the substrate at the background is gray. The sharp-edged scattered objects are ammonia crystals used as the buffering agent in the hydrofluoric acid etching.

Fig. 3.1a displays a schematic view of a sample fabricated with the HF method. Note that since the hydrofluoric acid is an isotropic etchant, the $SiO_2$ is slightly etched under the metal contacts at the edges. However, the $SiO_2$ under graphene is etched the full 150 nm since the acid can wet the interface between the graphene flake and the $SiO_2$ surface, and etching underneath starts almost immediately after immersing the chip into the etchant. Fig. 3.1b displays a scanning electron microscope (SEM) image of a sample fabricated using the HF method.

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After the fabrication, the sample was mounted in a sample box with microstrip lines that could be connected to the DC and RF ports in the measurement setup with standard electrical connectors. The sample was wire bonded to the striplines using 25 μm aluminum wire. The final step of sample preparation, the current annealing, was performed at low temperatures $T \leq 4$ K. In this step, a large current was passed through the device in order to heat up the graphene and to subsequently evaporate adsorbates and fabrication residues off; for further details of the procedure see Appendix A.1.3. This procedure resulted in very clean graphene with residual charge density of $n_{impurity} \approx 6 \times 10^9$ cm$^{-2}$, which is close to the best reported values, see Ref. [67].

**The LOR method**

The LOR-supported samples were fabricated following a recipe adapted from Ref. [58]. For the exact fabrication procedure used here, see Appendix A.1.1. The major differences compared to the HF method is that the metal structures were made of Cr/Au (5/70 nm) and supported by $\sim 500$ nm of lift-off-resist instead of SiO$_2$ that remained below serving as part of the gate dielectric. Consequently, the distance between the graphene flake and the back gate was approximately 800 nm ($\sim 500$ nm vacuum gap and 280 nm of SiO$_2$). However, note that due to the softness of the LOR under the bonding pads, the wire bonding pushed pads to the SiO$_2$ surface, as seen in the schematic in Fig. 3.2a. This happened relatively far away form the graphene junction and didn’t thus have any effect on it. Fig. 3.2b displays an SEM image of a sample fabricated using the LOR method.

As with the HF samples, the final stage of LOR-supported sample preparation was the low temperature current annealing. After annealing, the sample quality ended up being very similar to the case of the HF samples. All in all, the end result with both fabrication methods was very similar. However, the LOR technique has two advantages over the HF method. First, the geometry can be adjusted more easily, for example graphene-gate distance can be controlled and
layered structures, such as local gates, are easier to make. Second, the yield of high quality samples is higher using the LOR method [58].

### 3.1.2 Suspended graphene Corbino devices

In addition to the regular rectangular devices, samples with suspended Corbino disk geometry were developed. In the Corbino geometry, the graphene sheet was ring shaped (shape defined by the metal contacts), and contacted with metal leads at the inner and outer edges in a configuration where the upper contact is air-bridged over the lower one to the center of the device. Strictly in 2D this is impossible, however, the LOR method allows layered designs that can be used to realize the bridge contact needed, see Appendix A.1.2 for details about the fabrication. The sample structure is displayed in a tilted SEM micrograph in Fig. 3.3.

![Figure 3.3. SEM micrograph of a suspended graphene Corbino disk device where the blue-green denotes the graphene membrane, the Cr/Au metal contacts are colored yellow, supporting LOR is dark green, and the Si/SiO₂ substrate appears gray. The scale bar denotes 1 μm.](image)

The LOR support layers were ~500 nm thick for the both metal layers, meaning that the graphene-gate distance is the same as in the case of the rectangular LOR samples (around 800 nm), and that the top contact is another 500 nm above the bottom contact. Both contacts were made of Cr/Au as before, though, thicker metal layer (around 5/100 nm of Cr/Au) was used for the top contact in order to make sure that the metal film remains continuous close to the middle contact area.

As in the case of rectangular samples, the Corbino devices were current annealed at low temperatures in order to clean them. Despite the geometric asymmetry and consequent uneven heating, current annealing was effective and the residual density was reduced to the level of the order of ~10⁹ cm⁻². The high quality of the devices was confirmed by high magnetic field measurements that displayed phenomena distinct to extremely clean samples, foremost a clear fractional quantum Hall effect was observed (for details see Sec. 7).
3.1.3 Charge density modulation using the back gate

The charge carrier density and the carrier type in graphene can be tuned by the gate voltage $V_g$. As charge is accumulated on the surface of the gate electrode, a charge of equal magnitude and opposite polarity is induced in graphene. The resulting charge carrier density is expressed as:

$$n = \frac{C_g(V_g - V_d)}{e}, \quad (3.1)$$

where $V_d$ is the gate voltage at the charge neutrality point (the Dirac point), and $C_g$ is the specific gate capacitance. In the conductance measurements, the Dirac point can be identified as the point of minimum conductivity, i.e. "the Dirac peak" in a resistance vs gate voltage sweep.

The specific capacitance of a back-gated graphene device can be estimated using a parallel plate capacitor model where two capacitors corresponding the air gap and $SiO_2$ dielectric are in series:

$$C_g = \frac{C_{vacuum}C_{SiO_2}}{C_{vacuum} + C_{SiO_2}} = \frac{\varepsilon_{r, SiO_2}\varepsilon_0}{d_{SiO_2} + d_0\varepsilon_{SiO_2}}, \quad (3.2)$$

where the individual components of the specific capacitance are of the form $C_x = \frac{\varepsilon_0\varepsilon_r}{d_r}$ where $x$ denotes either vacuum or $SiO_2$. The vacuum permittivity is $\varepsilon_0 \approx 8.854 \times 10^{-12} \text{ F/m}$, and $\varepsilon_r$ is the effective relative permittivity of the medium in question ($\varepsilon_{r, vacuum} = 1$ and $\varepsilon_{r, SiO_2} = 3.9$). In case of HF-etched samples the parallel plate capacitor model yields $C_{g, HF} = 4.7 \times 10^{-5} \text{ F/m}^2$ and for the LOR samples $C_{g, LOR} = 1.5 \times 10^{-5} \text{ F/m}^2$.

The exact gate capacitance for a specific sample can also be determined directly from magnetoconductance measurements by identifying the quantum Hall states. As was shown in Sec 2.3, the identification of the states is unique for monolayer graphene due to the anomalous quantum Hall effect, this will fix the $n$ scale through the filling factor $\nu = \frac{\hbar n}{eB}$ as long as the magnetic field $B$ is known, see Fig. 7.1 where the states are identified. These measurements yielded the average specific capacitance around $C_{g, LOR} = 1.3 \times 10^{-5} \text{ F/m}^2$, which is well in line with the value estimated with the parallel plate capacitor model. The small deviation is most likely due to inaccuracy of the LOR thickness estimation.

When the charge carrier density induced by the gate is known, one can also obtain an expression for the chemical potential offset, from Ref. [68]:

$$\mu = \hbar v_F \sqrt{\frac{4\pi n}{g_s g_v}} = \hbar v_F \sqrt{\pi n} = \hbar v_F q_F, \quad (3.3)$$

where $g_s = 2$ and $g_v = 2$ are the spin and valley degeneracies, and $q_F$ is the Fermi wave vector.

Similarly, one can also obtain chemical potential expression of bilayer graphene, from Ref. [68]:

$$\mu = \frac{2\pi \hbar^2 n}{m^* g_s g_v} = \frac{\pi \hbar^2 n}{2m^*} \quad (3.4)$$
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where \( m^* \) is the effective mass in bilayer graphene.

### 3.2 Cryogenic measurement setup

The measurements were conducted in a BlueFors LD-400 dry dilution refrigerator with a base temperature of \( T \approx 10 \) mK at the mixing chamber. The upper parts of the cryostat (stages at 45 K and 4 K) were cooled by a two-stage pulse tube cooler, and the lower parts were cooled by the dilution machine \( T_{\text{still}} \approx 800 \) mK and \( T_{\text{MXC}} \approx 10 \) mK). Furthermore, the temperature of the mixing chamber, on which the devices were mounted, could be controlled accurately using a PID controller. Naturally, the cryostat was enclosed in a vacuum that maintained the ultra-high vacuum conditions necessary for not only for the operation of the cryostat, but also for the experiments due to the sensitive nature of the suspended graphene devices that were essentially all surface. Additionally, the cryostat was fitted with a 9 T superconducting magnet used in the quantum Hall measurements. Fig. 3.4a displays the installation inside a shielded room that is used to reduce electromagnetic interference in the measurements.

![Figure 3.4.](image)

**Figure 3.4.** a) Photo of the cryogenic measurement setup inside the shielded room, the white tube is the vacuum can in which the cryostat and the 9 T superconducting magnet resided. The preamplifiers were positioned on the top plate of the installation. b) Photo of a sample box used in the measurements with a 5 mm X 5 mm silicon chip placed in it. Note the wire bonds that connect the sample bonding pads to the microstrip lines that are in turn soldered to SMA connectors seen on all sides of the box.

The cryostat had 24 DC-lines (12 twisted pairs of phosphor-copper wire) with \( \pi \)- and \( RC \)-filtering (nominal cutoff 20 kHz) at the mixing chamber, four high frequency (RF) input lines, and four high frequency (RF) output lines. The output lines are fitted with circulators for blocking incoming noise, and cryogenic LNAs at the 4 K stage to amplify the signals (shot noise) generated by the samples. Outside the cryostat, the measured signals were amplified further using preamplifiers (high and low frequency alike), note the low frequency amplifiers positioned on top of the cryostat in Fig. 3.4a. The amplified signals were measured using appropriate instruments either inside or outside the
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shielded room and the data were collected by a measurement computer via a GPIB link. In addition to running the measurements, the measurement computer was also used to control the cryostat operation, the temperature and the magnetic field, as well as to log all the conditions, such as temperatures and pressures in various parts of the system.

The fabricated samples (on 5 mm X 5 mm Si chips) were connected to the measurement setup by first placing them in sample boxes, such as the one displayed in Fig. 3.4b, and then connecting the measurement lines to the box connectors. The sample boxes made of copper were clamped to the mixing camber plate for good thermal contact, or hung inside the bore of the 9 T superconducting magnet by copper coaxial cables in the case of high field measurements.

3.2.1 Noise measurement setup

In Fig. 3.5, there is a schematic drawing of the electrical measurement setup inside the cryostat. Note that some details such as filters, attenuators, and RF-input lines have been left out from the image. This setup was used for all the measurements in this work, however, only the parts relevant to the measurement were connected in any given experiment. For example, the diode setup depicted red in the figure was used in the electron-phonon coupling measurements, and during these measurements the correlation setup and the low frequency spectrum analyzer SRS 785 were not connected. In the noise measurements on the $\nu = 0$ quantum Hall state breakdown, the correlation setup was used while the diode setup was not connected. The reason for using the more complicated correlation setup with the latest quantum Hall breakdown measurements was to eliminate uncorrelated and unwanted noise in different channels that could have been detrimental for the noise measurement at extremely small biases and long integration times required in that work. With relatively fast electron-phonon coupling measurements, the simpler single channel diode measurement was sufficient.

Conductance measurements were needed in all experiments, and that part of the setup remained the same and always connected.

Regardless of the scheme used in a noise experiment, the goal of the measurement was to measure the excess shot noise spectral density $S(V) - S(0)$, i.e. the noise that is generated only by the biasing. From the bias dependence of the noise one can extract the Fano factor $F$ by calibrating against a known noise source.

In case of the diode measurement setup, the voltage measured after the Schottky noise diode was directly proportional to the noise power in the band 600-900 MHz limited by the noise measurement setup [69]. During these measurements the noise was calibrated against $Al-AlO_x-Al$ tunnel junction with the Fano factor $F = 1$. The calibration was done by simply comparing the slopes of the measured $S$ vs $I$ plots. This could be done since the same measurement lines
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were used up from the mixing chamber where a microwave switch was used to select either the tunnel junction or the sample as the noise source.

![Diagram of noise measurement setup](image)

**Figure 3.5.** Simplified presentation of the electrical measurement setup including high frequency noise measurements, as well as low frequency noise and conductance measurements. Filters and attenuators have been left out of the picture for clarity. The ring at the bottom represents a two-terminal Corbino sample connected to high and low frequency source-drain lines through bias tees (Mini-Circuits ZX85-12G-S+). The capacitor plate at the bottom denotes the DC back gate. The high frequency lines are connected to the amplifier chains through microwave switches (Dowkey 431-series, where the TTL logic circuits had been removed) and circulators (Pamtec 55387). In addition to the sample, the switches could be connected to calibration sources, such as 50 Ω resistors depicted here. At room temperature, the two optional noise measurement setups are displayed: the diode setup, and the correlation setup. Note that only one channel is needed with the diode setup. The low frequency spectrum analyzer SRS 785 was used to record the low frequency noise after the current amplifier SR570.

In the case of the correlation measurement the situation was similar. However, instead of direct analog measurement of the noise power, the noise in the band 650-900 MHz was first downmixed using ADL-5380 IQ-mixers and a local oscillator at frequency $f = 780$ MHz. The $I$ and $Q$ components were then digitized using a 125 MS/s (mega samples per second) AlazarTech ATS9440 digitizer card. The noise power in each channel, as well as their cross correlation, were calculated from the digitized data in real time using GPU accelerated fast
Fourier transform and recorded as a function of bias voltage. However, since the measured noise originated from a two-terminal sample, the recorded cross correlation of the channels should be viewed as autocorrelation of the excess noise produced by the sample, i.e. \( S(V) - S(0) \). The shot noise in the two ports is correlated due to the current conservation. More details of this measurement and the measurement setup can be found in Refs. [70, 71].

In the case of the quantum Hall breakdown measurements, where the correlation technique was used, the noise was calibrated against the thermal noise from the 50 \( \Omega \) resistors positioned at the mixing chamber (\( T \approx 10 \) mK) and still (\( T \approx 800 \) mK) positions. Since the thermal noise power spectral density from a resistor \( R \) at a temperature \( T \) is given as \( v_n^2 = 4k_B T R \), the correct noise level from a graphene sample could be fixed by this known power spectral density, see Ref. [71] for details. The accuracy of this calibration was tested by calculating the low-bias Fano factor from measured data and comparing it to the prediction of Eq. 2.19. Relatively good agreement between the theory and experiments was obtained in the light of the extreme conditions that the measurements were conducted.

### 3.2.2 Shot noise measurements and noise thermometry

The excess noise spectral density, the additional noise generated due to biasing, is dominated by thermal noise at low bias voltage and low frequency (\( eV, \hbar \omega < k_B T_e \)). The crossover from this regime to the regime where shot noise dominates is described by the Khlus formula [72, 69]:

\[
S(\omega) = \frac{e^2}{2\pi \hbar} \left\{ 2\hbar \omega \coth \left( \frac{\hbar \omega}{2k_B T_e} \right) \sum_n T_n^2 \right\} + \frac{e^2}{2\pi \hbar} \left\{ \left[ (\hbar \omega + eV) \coth \left( \frac{\hbar \omega + eV}{2k_B T_e} \right) + (\hbar \omega - eV) \coth \left( \frac{\hbar \omega - eV}{2k_B T_e} \right) \right] \sum_n T_n (1 - T_n) \right\},
\]

(3.5)

where \( k_B \) is the Boltzmann constant, \( T_e \) the electron temperature, and \( \omega \) is the angular frequency. Since the frequencies relevant in to this work were relatively low, expression in Eq. 3.5 can be simplified by taking the zero-frequency limit \( \omega = 0 \) leading to:

\[
S(V) = \frac{4k_B T_e}{R} \left[ \frac{Fe|V|}{2k_B T_e} \coth \left( \frac{e|V|}{2k_B T_e} \right) + 1 - F \right],
\]

(3.6)

where \( R \) is the sample resistance, and \( F \) is the Fano factor.

The bias voltage dependence of the Fano factor at high bias can be extracted from Eq. 3.6 with approximation: at high bias \( eV >> k_B T \) the hyperbolic cotangent term \( \coth \left( \frac{eV}{2k_B T_e} \right) \approx 1 \). Thus, the non-equilibrium noise spectral density can be approximated to be:

\[
S(V) - S(0) = \frac{2Fe|V|}{R}.
\]

(3.7)
As explained in Sec. 2.2.3, the Fano factor depends on the scattering lengths in the sample. These length scales in turn depend on bias voltage $V$. At low bias, $F = \frac{1}{3}$ for graphene at the Dirac point. However, at moderate bias voltages (so-called “hot electron regime”) the Fano factor for diffusive systems is expected to be $F = \frac{\sqrt{3}}{4}$ due to electron-electron interaction effects, as discussed in Ref. [44]. At higher bias, inelastic processes, namely the electron-phonon collisions, suppress the Fano factor. This is indeed what is observed in the graphene experiments, such as the one presented in Fig. 3.6.

By equating the excess shot noise $S(V) - S(0)$ in Eq. 3.7 to the formula of thermal noise $\frac{4k_BT_e}{R}$, one finds the system electron temperature [74, 75]:

$$T_e = \frac{Fe|V|}{2k_B}. \quad (3.8)$$

This equation enables one to determine the electron temperature $T_e$ of the graphene electron system by measuring the excess high frequency noise as a function of bias voltage $V$. 

**Figure 3.6.** a) Shot noise measured using a Krytar Schottky diode near ($V_g = 0$ V) and far ($V_g = 10$ V) from the Dirac point. b) The Fano factor extracted from the noise curves. The horizontal lines indicate $F = \frac{1}{3}$ and $F = \frac{\sqrt{3}}{4}$ levels. All the data is from a bilayer graphene sample S1 in Ref. [73].
4. Transport through suspended graphene sheet

In this section zero-magnetic-field quantum transport experiments on rectangular suspended graphene samples are discussed. As it is evident that the contacts to the graphene sheet are a crucial and non-trivial part of transport in the devices, they will be reviewed first. Then, transport through $pnp$ structures and Fabry-Pérot interfaces observed in the cavity defined by the contacts and the edges, are presented.

4.1 Contact doping

When two conducting materials are brought in contact, the Fermi levels $E_F$ at the interface are equilibrated so that a steady state without current flow from one material to the other one is reached. However, this leads to charge transfer between the two materials, since generally different materials have different work functions. The direction of the electron movement is from the smaller work function material to the larger work function material. A common example of this is the Schottky diode where a semiconductor is paired with a metal and charge transfer leads to band bending.

In the case of top contacted graphene (or gate-induced doping), the simplest way to model the doping and transport through the formed junctions is to consider rectangular infinitely sharp potential, as was the case with Klein tunneling reviewed in Sec. 2.1.2. While this approach nicely demonstrates the principles of relativistic electron transport through a barrier, it fails to provide any insight into details inherent in real devices, such as the width of the formed junctions. This type of information can be vital when considering for example interference effects, such as Fabry-Pérot, between the contacts.

In this work, contact doping by top contacts in HF-etched suspended graphene devices with a $Si^{++}$ back gate was considered. The structure of the device is presented in Fig. 4.1 where separation to two sections is made. First, graphene under the contact gated by the back gate with 300 nm of $SiO_2$ as dielectric, and mainly doped by the metal contact due to work function difference. Second, the suspended graphene section that is gated by the same back gate but 150 nm of
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the gate dielectric, SiO$_2$, had been etched away.

Figure 4.1. Cross-sectional view of the suspended and supported regions in the HF-etched devices, from the right to left the materials are Si$^{++}$ back gate (grey), SiO$_2$ dielectric (orange), graphene (red), metal-graphene separation layer (yellow), and metal (gray), note also partial vacuum gap in SiO$_2$. The charge densities are marked with $n_*$ for different parts: $n_{gM}$ in the back gate under the contact, $n_M$ in graphene under the contact, $n_c$ in the metal contacts, $n_G$ in suspended graphene, and $n_{gG}$ in back gate under the suspended part. Correspondingly, $d_*$ denotes thickness of the different parts in $z$-direction: $d_G$ is the oxide thickness under contact, $d_c$ is the width of the contact region, and $d_{vac}$ is the vacuum gap. Electrochemical potential structures along the lines M-g and G-g are presented in Fig. 4.2.

Figure 4.2. a) Variation of electrochemical potential along the line M-g, from the metal contact to the back gate at the contact region. b) Potential structure along the line G-g from suspended graphene to the back gate.

The charge transfer in the contact region has two contributions: the contact doping $n_c$, and gating by the back gate $n_{gM}$. Thus, the charge density induced in graphene under the contacts is $n_M = n_{gM} + n_c$. The Fermi level shift corresponding to $n_M$ can be written using Eq. 3.3 as:

$$\Delta E_{FM} = \hbar v_F \cdot \text{sgn}(n_M) \sqrt{\pi |n_M|}. \quad (4.1)$$

The potential shifts corresponding to the individual components, which in turn can be written as: $U_{gM} = e n_{gM} / C_{gM}$ and $U_c = e n_c / C_c$. Where $C_{gM} \approx 1.2 \times 10^{-4}$ F/m$^2$ is the specific gate capacitance of the fully SiO$_2$-filled capacitor, and $C_c \approx 10^{-1}$ F/m$^2$ is the specific contact capacitance between gold and graphene [76, 73].

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In addition to the potential shifts associated with the charge accumulation, one has to also consider the work function difference between the metal $W_M$ and graphene $W_G$ denoted as $\chi$:

$$\chi = W_G + \Delta_c - W_M,$$

(4.2)

where the additional term $\Delta_c$ is a contribution from chemical interactions between graphene and the metal.

In order to fix the potential, one can use three conditions:

$$e\delta V_g = W_g - W_G$$

(4.3)

$$e\delta V_g = \chi + eU_{gM} - eU_c$$

(4.4)

$$\Delta E_{FM} + eU_c = \chi,$$

(4.5)

where the first one equates the work function difference between graphene $W_G$ and the back gate material $W_g$ to the Dirac point shift ($\delta V_g = V_g - V_d$) in otherwise neutral graphene. The second equilibrium condition comes from the fact that the difference between the Fermi levels of the metal and the gate equals to $e\delta V_g$. The third comes from the condition that the Fermi levels in the metal and graphene under metal coincide.

The complete potential distribution under the contact is presented in Fig. 4.2a. Taking the structure and the above conditions into account, one can define the Fermi level shift in graphene under the metal contact as:

$$\Delta E_{FM} = \text{sgn} \left[ \delta V_g + \frac{\chi C_c}{eC_{gM}} \right] \left\{ -\frac{C_c + C_{gM}}{2} \zeta_F^2 + \sqrt{\left( \frac{C_c + C_{gM}}{2} \zeta_F^2 \right)^2 + \zeta_F^2 C_c \chi + \frac{C_{gM}}{C_c} e\delta V_g} \right\},$$

(4.6)

where $\zeta_F = \sqrt{\frac{\pi \hbar v_F}{e}}$ denotes the "Fermi electric flux".

When considering the suspended region, the potential structure is slightly simpler. There, only the work functions of graphene and the gate material needed to be taken into account along with the applied DC gate voltage $V_g$ (potential $U_{gG} = eU_{gG}/C_{gG}$). Similarly to the earlier case of graphene under the contact, one can define the Fermi level shift in the free-standing graphene by following the energy diagram of Fig. 4.2b as:

$$\Delta E_{FG} = \text{sgn} \left[ \delta V_g \right] \left\{ -\frac{C_{gG}}{2} \zeta_F^2 + \sqrt{\left( \frac{C_{gG}}{2} \zeta_F^2 \right)^2 + eC_{gG}\zeta_F^2 |\delta V_g|} \right\},$$

(4.7)

where the specific gate capacitance of the suspended region is $C_{gG} = 4.7 \times 10^{-5}$ F/m$^2$.

Eqs. 4.6 and 4.7 define the Fermi level shift in graphene under the metal contacts and in the freely suspended region, respectively. Apart from the trivial case, there is a potential barrier $eV_0 = \Delta E_{FM} - \Delta E_{FG}$ in the device. However, instead of modeling the interface as a step, as was presented in the toy model in Sec. 2.1.2, a constant slope from one region to the next in the contact region
with the width $d$ was incorporated in the model. The structure of this potential barrier is presented in Fig. 4.3a.

Note that since the Fermi level shifts $\Delta E_{FM}$ and $\Delta E_{FG}$ have rather complex dependence on $V_g$, this model is more than just adding slopped potential to the problem of tunneling through a $pnp$ or $pp'p$ barrier (here $pp'p$ refers to a case where the suspended region is doped with the same type of carriers as the parts under the contacts, but with a different density). This model takes into account the potential shift due to gating and doping under the contacts as well as in the suspended region jointly, making it more realistic description of the situation in the experiments.

### 4.2 Transport through pnp-junctions

Given the above electrochemical-potential analysis one can construct the model presented in Fig. 4.3a. This model consists of Fermi level offsets $\Delta E_{FM}$ and $\Delta E_{FG}$, as well as constant potential slope regions with width $d$ between them. As presented in Sec. 2.1.2, one can include the effect of potential barriers in the Dirac equation Eq. 2.4 as a part of the potential $V(r)$.

The solution of the Dirac equation, now in five separate regions including the sloped ones, can be used to calculate the transmission coefficients $T$ in a similar manner as in the case of $pn$ junctions in Sec. 2.1.2. This rather lengthy calculation is presented in Ref. [77].

Given the transmission $T$ through a single $pn$ junction governed by Klein tunneling, the total transmission through a $pnp$ barrier can be calculated as an incoherent sum of both contributions [77], which yields the form:

$$T_{tot} = \frac{1}{2T^{-1} - 1},$$

(4.8)
where equal transmission $T$ is assumed on both sides. The incoherent sum refers here to the fact that there is no phase coherent transport through the $pn$ barriers at the edges, and consequently no interference terms (higher order reflections) are included in the summing. Note that this only implies that there is no coherent transport from graphene under the left contact to graphene under the right contact, the suspended region has phase coherence since Fabry-Pérot resonances are observed, as discussed in the next section. Additionally, the incoherent nature of transport under the contact does not imply low transmission, in fact, the transmission through can remain close to $T = 1$, while the phase coherence that is usually more sensitive to disorder effects and noise is expected to be destroyed first.

By using the transmission coefficients and Landauer-Büttiker formulas, one can calculate the total conductivity and Fano factor of the whole structure as:

$$
\sigma = \frac{4e^2W}{\pi h} \int_0^{\min\{k_{FM},k_{FG}\}} dk_y T_{tot}, \quad (4.9)
$$

$$
F = 1 - \frac{1}{\sigma} \int_0^{\min\{k_{FM},k_{FG}\}} dk_y T_{tot}^2, \quad (4.10)
$$

where the Fermi wave vectors in regions G and M, respectively, are $k_{FG} = |\Delta E_{FG}|/\hbar v_F$ and $k_{FM} = |\Delta E_{FM}|/\hbar v_F$, and $W$ is the width of the sample. There are two parameters built into these expressions: the slope of the barrier $a$ and the so called impact parameter $p_0 = \text{sgn}(V_0) \sqrt{\alpha d}$, which are the only fitting parameters for the whole model. $\sigma$ and $F$ depend on $V_g$ in a non-trivial way through the wave vectors. Consequently, the expressions of Eqs. 4.9-4.10 can be fitted to the data, as has been done in Fig. 4.3b for the conductivity. In order to fix all fitting parameters, the Fano data was also compared to the model in Ref. [77].

Four main results were obtained from this analysis. First, the analysis agrees with the idea of Fermi level pinning and subsequent weak gate dependence of carrier density under the metal contacts, and thus it is qualitatively in line with the DFT calculations of Ref. [76]. From the model one gets the contact doping $\chi = -18$ meV (hole doping in the sign convention used here), which is rather small compared to DFT calculations $\chi \approx -0.1 - 0.2$ eV [76, 78] for Au contacts, as well as $\chi = -0.1$ from measurements with Cr/Au contacts in Ref. [79]. However, this difference most likely do to the fact that those works considered Cr/Au or pure Au contacts whereas in this work chromium was the middle layer in the Au/Cr/Au (5/7/50 nm) structure.

Second, according to the model there should be an offset in the charge carrier density in the suspended part even at the $V_g = 0$ V due to the work function difference between graphene and the back gate material $W_g - W_G = e\delta V_g \approx -0.2$ V. Thus, the fact that the Dirac point is not exactly at $V_g = 0$ V seems to reflect the choice of gate material rather than residual doping. In fact, the measurement of this voltage caused by the work function difference is the basis for Kelvin probe microscopy [80].
Third, from the fits one can extract the width of the \textit{pn} junction, \( d \sim 20 \text{ nm} \). This in line with Refs. [81, 82] where the values ranged between \( d = 5 - 40 \text{ nm} \). Thus, the junction width is small compared to the length of the suspended region, typically 1 \( \mu \text{m} \) in length. Consequently, when considering interference effects, such as Fabry-Pérot in the next section, the width of the \textit{pn} interface can be neglected.

Finally, the presented model was extended by adding contact resistance that led to less-than-perfect transmission through the contacts. This was done in order to estimate the contact transparency. By varying this contact reflection and comparing the resulting fits to the data, agreement was found with \( T = 0.95 - 1 \) while values \( T < 0.9 \) resulted in poor correspondence with the measurement. Consequently, it can be concluded that the analysis leading to Eq. 4.8 seems reasonable and one expects coherent transport only in the "cavity" of the suspended region.

### 4.3 Fabry-Pérot resonances

The Fabry-Pérot resonance is a type of interference phenomenon where transmission through a cavity defined by two semi-transparent mirrors gets modulated as a function of the cavity length or the wavelength of the transmitted waves. The modulation is due to alternating constructive and destructive interference between the waves entering the cavity and the ones being reflected back and forth by the semi-transparent mirrors, as illustrated in Fig. 4.4a. The Fabry-Pérot resonance was discovered in optical systems by virtue of access to good mirrors and phase coherent sources. Today, Fabry-Pérot resonances are utilized in various optical instruments due to their strong frequency selectivity.

\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig44.png}
\caption{a) Schematic representation of a Fabry Pérot cavity when the incoming waves are in resonance (top), out of resonance (middle), and anti-resonance (bottom). The size of the arrow on the right side signifies the relative transmission strength. b) Schematic view of the graphene Fabry-Pérot cavity used in the experiments with the correct aspect ratio \( W/L \approx 4 \), the width \( W \) and the length \( L \) of the cavity are marked in the figure. The yellow parts represent the metal contacts, and the hexagonal grid depicts graphene.}
\end{figure}

The Fabry-Pérot resonance can also be observed in an electronic solid state...
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system under two conditions: there are semi-transparent contacts defining the cavity, and the electron transport is phase coherent at the length scale of the cavity. Since clean graphene has extremely long mean free path at low temperatures, lengths exceeding 28 μm have been reported in hBN-encapsulated samples [83], observation of such resonances is feasible in the commonly fabricated μm-scale samples. Additionally, as the contact doping analysis in the previous section suggested, the top contacts in our samples are very transparent while allowing some reflection, i.e. they act as semitransparent mirrors for electrons.

Since the Fabry-Pérot modulates the transmission through the graphene flake $T$, one can expect the same pattern in the measured conductivity $\sigma$ and Fano factor $F$ (or in noise spectral density $S$), see Landauer-Büttiker formulas in Sec. 2.2.3. The theory behind the conductance fluctuation is presented fully in Ref. [84] where each Fabry-Pérot resonance is associated with a particular energy scale which depends on the channel length and the velocity of the charge carriers. While the analysis in Ref. [84] takes into account resonances in the longitudinal direction (between contacts separated by $L$), it was theorized in Ref. [85] that similar resonances should arise also from transversal confinement (between edges separated by $W$). By taking both modes into account, the locations of the Fabry-Pérot resonances can thus be written in terms of energy associated with longitudinal and transverse quantization as:

$$E_{q_L,q_W} = \pm \sqrt{E_L^2(q_L + \delta_L)^2 + E_W^2(q_W + \delta_W)^2},$$

(4.11)

where $E_L = \hbar v_F / 2L$ and $E_W = \hbar v_F / 2W$ are the "particle in the box energies" in the longitudinal and transverse directions, and $q_L/W$ are the corresponding wave vectors. $\delta_{L/W}$ take into account details of the contacts and edges as boundary conditions [84].

Note that observation of the transversal modes not only requires high quality graphene sheet (long mean free path) and high-quality edges, but also aspect ratio that is $W/L \neq 1$, so that they can be distinguished from the longitudinal modes. In the results presented here $W/L \sim 4$, see Fig. 4.4 for illustration of this. Additionally, since the transverse modes were expected to have weak visibility (due to weak coupling to the longitudinal transport that was probed in the experiment), the longitudinal resonances must remain weak as well, so that the transversal resonances would not be completely overwhelmed.

The Fabry-Pérot resonances were searched in the conductivity $\sigma$ and shot noise $S$ of a rectangular suspended graphene device with high aspect ratio $W/L \approx 4$. Essentially, Eq. 4.11 tells us that the resonances should present themselves as diamonds in the two-dimensional mappings $\sigma(\mu, V)$ and $S(\mu, V)$, where $\mu = sqn(n)\hbar v_F \sqrt{\pi|n|}$ is the chemical potential and $eV$ is the energy of associated with biasing. Indeed, these diamonds were visible in the conductance, but due to relatively strong background variation derivatives $d\sigma/d\mu$ and $dS/d\mu$ displayed the oscillations better, and were thus easier to analyze, see Figs. 4.5a and 4.5c.
In order to better understand the resonances, the $d\sigma(\mu,V)/d\mu$ and $dS(\mu,V)/d\mu$ mappings were Fourier transformed to see periodicity in the bias and gate directions. The 2D Fourier transforms presented in Figs. 4.5d and 4.5d reveal two sets of resonances (solid and dashed lines in the figure) that are separated by a factor of 4, as expected for longitudinal and transversal modes based on the aspect ratio of the measured sample. Moreover, the energy scales match to the expected energies $E_{L,W}$, when the Fermi velocity is set to a value $v_F = 2.4 - 2.8 \times 10^6$ m/s. Although this value is well over the usually quoted value $v_F = 1 \times 10^6$ m/s, it is fully in line with the Fermi velocity renormalization previously measured in ultra-clean suspended graphene at very low charge densities [86].

The slight skew in the Fourier transformation figures (also present in the data itself) is attributed to slight asymmetry in the contacts, as explained in Ref. [67]. Furthermore, these conclusions were reinforced by comparing the measured results to DFT simulations, which agreed well with the experiments.

The results have several implications. First, the fact that the Fabry-Pérot resonances are visible confirms that the electron transport is phase coherent in the suspended part of the graphene flake. Since the resonance spacing matches the aspect ratio of the suspended region, the Fabry-Pérot resonances and thus phase coherent transport does not seem to happen through the $pn$
Transport through suspended graphene sheet

junctions. Second, the presence of transverse resonances suggests that the mean free path is quite large $l \gg W \approx 4 \mu m$. Finally, the obtained Fermi velocity $v_F = 2.4 - 2.8 \times 10^6$ m/s at $n \sim 1 - 2 \times 10^{10}$ cm$^{-2}$ concurs the Fermi velocity renormalization which has been observed earlier in cyclotron resonance measurements in Ref. [86].
Transport through suspended graphene sheet
5. 1/f noise in bilayer graphene

5.1 Bilayer graphene low frequency noise characteristics

In this section, the work on 1/f noise in bilayer graphene is presented. The motivation for using bilayer graphene instead of monolayer in this 1/f noise study was two-fold. First, while 1/f noise is nuisance mainly at low frequencies, it can be upconverted at higher frequencies causing phase noise there [87]. If the noise were large, this upconversion could cause problems in the shot noise measurements (electron-phonon coupling experiments), which are presented in the following chapter. Second, at the time of these measurements suspended monolayer graphene 1/f noise had already been investigated [88], but bilayer was not. It was, however, reported that noise in on-substrate bilayer graphene devices was reduced due to effective screening of charged impurities based on bilayer graphene’s double layer structure [89]. This brought up the question if the noise level could be reduced further by eliminating the substrate.

As the review in Ref. [90] shows, the 1/f noise has been studied extensively in graphene, bilayer graphene, and in few layer graphene. The noise was found to follow Hooge’s empirical law, presented in Sec. 2.2.2, except for the charge carrier density dependence of the noise amplitude. Various observations were explained by suggesting contact noise and mobility fluctuations as dominant processes. Additionally, the noise was found to decrease as the number of layers in the graphene stack was increased, at least up to the point where the volume noise takes over at \( n \approx 7 \) [90]. More specifically, the noise reduction in bilayer graphene was explained in Ref. [89] as a result of the efficient screening of external charge impurity fluctuations. In addition to these on-substrate experiments, a reduction of 1/f noise has also been observed in monolayer suspended graphene \( w.r.t. \) devices on \( SiO_2 \) [88]. In this case, the noise reduction was attributed to the absence of substrate and the charge trap fluctuators that come with it.

In this work, the noise characteristics of suspended bilayer graphene were found to follow the above trends. By suspending the bilayer graphene, the
1/f noise in bilayer graphene

noise was lowered by one decade below the previously reported values. This result is summarized in Fig. 5.1a, where the summary figure from Ref. [90] is supplemented with the suspended BLG data of this work. The figure shows the scaled noise $S_f/I^2$ as a function of device area at $f = 10$ Hz. It can be seen that for a given device area the noise in suspended BLG is at least one decade lower compared to the other structures. At the lowest, a noise amplitude of $A_n = f \times S_f/I^2 = 6.2 \times 10^{-10}$ is reached.

More detailed analysis of the suspended BLG noise revealed that the noise followed well generalized Hooge's empirical relation, see Ref. [91]. The spectrum is very clearly $f^\beta$ type with $\beta \approx -1$, and the current dependence $S_f \sim I^2$. However, there is practically no $V_g$ dependence in the noise amplitude $A_n$. This led to an interpretation that contact noise dominates the spectrum. The contact area can only be gated weakly (see discussion with monolayer graphene in Sec. 4.1), and thus the charge carrier density could not be modulated where the noise was produced. This makes sense in the case of current annealed suspended samples, since the disorder density on the graphene flake is extremely low and number of noise-producing fluctuators in the gateable area should be extremely low.

The contact noise most likely originates from charge traps leading to resistance fluctuation under the contacts, as suggested in Ref. [91]. In fact, they can be modeled as thermally activated fluctuators of form $S_f \sim \exp(-\Delta/k_BT)$, where $\Delta = 0.12$ eV is the activation energy extracted from the temperature dependence of the noise in the range of $10 - 100$ K. It is likely that these traps were formed as a byproduct of the current annealing.

Figure 5.1. a) Scaled noise $S_f/I^2$ presented as a function of device area at $f = 10$ Hz. The original picture is taken from Ref. [90] and supplemented with the suspended monolayer result (SLG) from Ref. [88] (pink star), and the BLG results of this work (black starts). The other symbols denote monolayer graphene (SLG FET, filled symbols), bilayer graphene (BLG FET, open symbols), and variable thickness graphene (GTG FET, half-filled symbols). The different shapes of symbols denote different devices. b) Noise spectra for a suspended BLG sample with different bias currents.
5.2 Gas sensing

Since the noise level in the suspended BLG devices was found to be extremely low, and suspended graphene is essentially all surface (part that is exposed to the environment), it intuitively makes a good platform gas sensing. In this work, the gas sensitivity was tested for air and neon gases by introducing small amounts of them, \( P \approx 10^{-4} \) mBar, into the vacuum can of the cryostat and measuring the change in the \( 1/f \) noise. The neon experiments were conducted by introducing the gas at higher temperatures where it was gaseous \( (T \approx 26 \text{ K}) \) and condensing it onto the graphene flake by cooling the cryostat down to \( T \approx 4 \text{ K} \) where the experiments were conducted. The experiments with air were conducted at higher temperatures around \( T \approx 80 \text{ K} \).

A molecule landing on a bilayer graphene flake can be seen as a single fluctuator producing a Lorentzian spectrum in a manner discussed in Sec. 2.2.1 in the context of random telegraph noise. After introducing gas onto the device, this is exactly what is observed in the experiment. Fig. 5.2 presents the noise amplitude \( A_n = f \times S(f)/I^2 \) before the Ne gas deposition, after the deposition, and after re-annealing that was performed in order to clean graphene close to the original state. It can be seen that the spectra of the clean sample before deposition and after annealing are flat, which implies that the spectrum is \( 1/f \) type. With the deposited gas on graphene there is a bulge in the spectrum at \( f \approx 70 \text{ Hz} \). This bulge corresponds to the corner frequency of the Lorentzian fluctuator spectrum discussed in Sec. 2.2.1. Correspondingly, one can estimate the approximate time constant of the fluctuations: \( \tau \sim \frac{1}{f} \approx 14 \text{ ms} \).

![Figure 5.2](image.png)

**Figure 5.2.** The noise amplitude \( A_n \) presented as a function of frequency in clean (blue), neon-contaminated (red), and re-cleaned (yellow) BLG device measured at \( T \approx 4 \text{ K} \).

A rough estimate for the amount of molecules deposited onto graphene was obtained by taking the pressure in the vacuum chamber and assuming uniform Ne adsorption on all surfaces in it. During the procedure, the cryostat was warmed up to \( T \approx 26 \text{ K} \) (neon in gas form at this temperature), and the vacuum
can volume \((V \approx 0.3 \text{ m}^3)\) was filled to a pressure \(P = 10^{-4} \text{ mBar}\) of neon gas. The total number of Ne atoms in the volume can roughly be estimated using the ideal gas law: \(N = PV/k_B T \approx 10^{19}\). Then the cryostat was cooled down to \(T = 4 \text{ K}\) using the pulse tube cooler, hence condensing neon onto the cold surfaces including the bilayer graphene sheet. By taking a rough estimate for the surface area of the condensing parts \((A \approx 10 \text{ m}^2)\), the area of the bilayer graphene sheet \((A_{BG} = 4 \mu \text{m}^2)\), and assuming uniform condensation on them, one can estimate that number of atoms on graphene was \(N \approx 10^6\). This corresponds to mass \(\Delta m \approx 10^{-19} \text{ kg}\) of neon. For comparison, the mass of the bilayer graphene flake was \(6 \times 10^{-18} \text{ kg}\), calculated from the mass density of bilayer graphene \(\rho = 2 \times 7.6 \times 10^{-7} \text{ kg/m}^2\).

The effect of ambient air was also observed at higher temperatures around \(T \approx 80 \text{ K}\) while warming up the cryostat without condensing the object gas onto the device. Air pressure change of \(P = 10^{-4} \text{ mBar}\) and the subsequent noise change \(\Delta S/I = 3 \times 10^{-11} 1/\text{Hz}\) gave an estimate of 0.3 ppm for the gas sensitivity.

In summary, the suspended bilayer graphene devices exhibited extremely low 1/f noise levels down to \(A_n = 6.2 \times 10^{-10}\). This presented an interesting possibility to observe small added noise by adsorbates, which indeed worked and spectrum dominated by Lorentian fluctuators was obtained. While the estimated mass sensitivity in the measurements was rather poor compared to the state of the art CNT devices with yoctogram resolution [92], this technique has the advantage of providing additional information of the adsorbates. In Ref. [90] it was reported that different molecules could be identified based on the corner frequency in the spectra (or bulge in the noise amplitude). Additionally, the fact that the devices in this work were suspended makes resonance measurements possible. In fact, a combination of mechanical resonance measurements with simultaneous low frequency noise determination forms a subject of future work.
6. Electron-phonon coupling

Graphene can be understood to consist of a periodic ionic lattice held together by the $\sigma$ bonds and the electron gas that resides in the lattice potential. In ideal graphene at low energies, the electrons are allowed to move without scattering across the lattice (ballistic transport), however, at high energies the electrons can scatter from lattice vibrations or excite them. This interaction is called the electron-phonon coupling (e-ph coupling).

Knowledge of e-ph coupling is of great importance in many applications since it often governs the energy relaxation in the system. For example, in applications where high power is absorbed in the electron system of graphene, the cooling rate and consequently the operating temperature reached in the device are determined by the e-ph coupling. Similarly, if graphene is used as an absorber in a bolometer device, the device performance (speed and sensitivity) is governed by the heat capacity of graphene and the energy relaxation rate (to phonons).

In this work, the electron-phonon coupling was investigated by heating the electron system with applied bias voltage and measuring the resulting electron temperature using noise thermometry. The measurements were conducted using suspended mono- and bilayer graphene devices [93, 73]. The diode-based shot noise measurement setup, described in Sec. 3.2.1, was used in both of these studies. The measured relaxation rates were analyzed by comparing to the theoretical predictions of the heat flow to the phonon system that allowed to gain insight into the physical processes involved.

6.1 Electron-phonon coupling in monolayer graphene

The samples considered here were rectangular HF-etched monolayer graphene devices with Au/Cr/Au contacts (5/7/50 nm). The electron system was heated by Joule heating $P = UI$ and the electron temperature $T_e$ was measured using noise thermometry. Since suspended graphene had no direct contact to the substrate but only to the metallic leads, the electron system could only be thermalized through electronic heat diffusion to the leads or by electron-phonon coupling to the graphene lattice, as illustrated in 6.1a. The graphene lattice in turn was
Electron-phonon coupling

well thermalized to the phonon system of the leads that functioned as heat sinks in this system. The leads remained at low temperatures during the experiment due to their relatively large mass (compared to the graphene), good conductance, and good thermal contact to the cryostat. Thus, in the following analysis the approximation \( T_{\text{sub}} \approx 0 \) has been made. Clearly, this approximation is justified, since the cryostat temperature remained at \( T < 0.5 \) K at all times while the electron temperature reached hundreds of Kelvins.

The phonon system in graphene was also kept relatively cool, \( T_{\text{ph}} < 100 \) K, so that always \( T_e >> T_{\text{ph}} \). This result was obtained by modeling the heat flow in the system using the value for the thermal boundary resistance (Kapitza resistance) between phonon systems in the leads and graphene from Ref. [94]. Essentially, this led to the situation where the power flow from the electron system to the phonon system was approximately equal to the Joule heating, \( P_{e - ph} \approx P_{\text{Joule}} \), at high \( T_e \) where electron-phonon coupling dominates the heat diffusion to the leads. Thus, different e-ph coupling mechanisms could be identified by looking at \( P_{e - ph} \) vs \( T_e \)-plots at high temperatures \( T_e > 200 \) K, where different phonon processes have characteristic cooling rates.

Coupling to acoustic and optical phonons was calculated in Refs. [95, 96]. In the case of acoustic phonons the energy flow per unit area is of form:

\[
Q = \Sigma \left( T_e^\delta - T_{\text{ph}}^\delta \right) \approx \Sigma T_e^\delta, \tag{6.1}
\]

where \( \Sigma \) is the e-ph coupling constant, and \( \delta \) a positive integer characteristic to a particular process. The approximation is obtained by using the fact that \( T_e >> T_{\text{ph}} \) in experiments considered here.

More exotic mechanisms, called "supercollisions", involving scattering via impurities [97] or ripples [98] have also been suggested. These processes may dominate simple acoustic and optical phonon scattering due to large energy...
Electron-phonon coupling

transfer associated with them. However, the energy transfer rate is generally of the form presented in Eq. 6.1.

Regular single phonon processes are restricted inside the Fermi sphere, thus limiting the momentum transfer to $2k_F$. This restriction also defines a characteristic temperature, the Bloch-Grüneisen temperature $T_{BG} = \frac{2\hbar k_F}{k_B}$, above which an arbitrary acoustic phonon with momentum $k \leq 2k_F$ can be excited. At low temperatures $T_{BG} > T_{ph}$, acoustic phonons from certain angles are available for scattering, as illustrated in Fig. 6.2. At high temperatures, full back scattering of electrons by acoustic phonons is possible. Note that the scale of $T_{BG}$, i.e., what high temperature means here, depends on the charge density $n$ since $T_{BG} \propto k_F \propto \sqrt{n}$. Consequently, the Bloch-Grüneisen temperature is only relevant in materials with low $n$, such as graphene, and not for example in metals.

Figure 6.2. Available phonon space at different temperature regimes defined by the Bloch-Grüneisen temperature $T_{BG}$, illustration from Ref. [99]. In the case of panels a and b the temperature is high enough to only scatter electrons inside the Fermi surface. However, in the case high phonon temperature $T_{BG} < T_{ph}$ in panel c scattering outside the Fermi sphere is possible, these processes are called supercollision.

Supercollisions can dominate single phonon scattering at high temperatures, $T_{BG} < T_{ph}$, since they can facilitate larger momentum transfers than $2k_F$ allowed for the acoustic phonons, and thus larger heat flow can be achieved. However, this type of scattering is not possible with a single phonon due to the fact that the scattering electron must end up on the Fermi surface at the end of the process. In samples lying on a substrate this "back kick" is provided by impurities, as reported in Ref. [99].

In suspended graphene, the amount of impurities is extremely low, $n_0 \approx 0.8 \times 10^{10} \frac{1}{cm^2}$ in this set of measurements [93], and thus impurity-assisted supercollisions are rare. Instead, supercollisions aided by flexural phonons can dominate. This was indeed observed in our experiments. The electron-phonon energy flow via the so-called ripple-supercollisions is reviewed in Ref. [98] and the schematic of scattering process is presented in 6.1b. It turned out that, indeed, the power flow obeys a power law, however, when the charge carrier density is tuned, a cross-over from $T_e^3$ to $T_e^5$ is observed. The energy flow from
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electrons to phonons per unit area $\dot{Q}_S$ reads:

$$
\dot{Q}_S|_{T_{ph}=0} \approx - \frac{N}{(2\pi \hbar^2 v_F^2)^2} \frac{g_D^2 k_B^5}{h k_F \ell} q^2\left(\frac{\hbar v_F \sqrt{\pi n}}{k_B T_e}\right),
$$

(6.2)

where $q(z) \approx 9.69 + 1.93z^2$ for $|z| \ll 5$ and $q(z) \approx 2\zeta(3)z^2 \approx 2.40z^2$ for $|z| \gg 5$, $N = 4$ is due to the spin and pseudospin degeneracy in graphene, and $\zeta(x)$ is the Riemann zeta function. The electron-phonon coupling constant for deformation potential is $g_D = D/\sqrt{2\rho s^2}$, where $D$ is the deformation potential and $\rho$ the mass density of the graphene sheet. The quantity $k_F \ell$ denotes the dimensionless mean free path associated with the additional scattering mechanism enabling the supercollisions.

An effective value of $k_F \ell = 3$ was used here. This bias-induced value for $k_F \ell$ is much smaller than expected for short-range scatterers in high-quality graphene, however, the value is justified in the model involving flexural phonons. In this model, the value of $k_F \ell$ is related to the strain in graphene, which was deduced from mechanical resonance measurements, and $R = T_e^{q^2}$-behavior of the measured resistance (see Refs. [100] and [93] for details). Small $k_F \ell \approx 2$ has also been observed in STM studies in Ref. [101]. Thus, it is clear that the low $k_F \ell$ value caused by the presence of flexural phonons is not fully comparable with previous small bias experiments with large values of $k_F \ell$. Additionally, the renormalization of the Fermi velocity by electron-electron interactions [102, 86] was taken into account with dependence on both charge density and electronic temperature $k_F \ell$, as explained in detail in the supplementary material of Ref. [93].

![Figure 6.3. Monolayer graphene power flow: a) $P/T_e^5$ for different gate voltages as a function of $T_e$ b) dependence on the charge carrier density at different temperatures $T_e$. The solid lines mark the theory from Eq. 6.2 in the high charge carrier limit ($|z| \gg 5$) and the dashed lines in the low charge carrier limit ($|z| \ll 5$).](image)

The heat flow $P$ from electrons to phonons scaled by $T_e^5$ is depicted in Fig. 6.3a as a function of $T_e$. It can clearly be seen that the Dirac point data levels off implying $T_e^5$-dependence while at $V_g = -10$ V we observe a distinct slope, which implies weaker behavior corresponding to $\delta = 3$. Reasonable fits to Eq. 6.2 were made, as can be seen in Ref. [93]. In Fig. 6.3b, the measured heat flow is
presented as a function of charge carrier density \( n \) at three different values of \( T_e \). The solid and dashed lines indicate the theoretical fits using Eq. 6.2.

All in all, best fit to the measured heating curves \( P - T_e \) was obtained with the theory based on ripple-assisted supercollisions. Simple single-phonon processes and the impurity assisted supercollisions observed in on-substrate samples produced too low cooling rates and didn’t match with the charge carrier dependence. It can be concluded that ripples inherently present in suspended graphene dominate the heat transport at electron temperatures near the room temperature.

### 6.2 Electron-phonon coupling in bilayer graphene

The electron-phonon coupling in suspended bilayer graphene was studied the same way as in the case of monolayer graphene using the diode measurement scheme. In this series of experiments, however, the suspended bilayer samples were fabricated with two different methods (HF etching and LOR method). The measurement result were found to be equal for both type of devices. The detailed study can be found in Ref. [73]. All in all, this study was identical to the monolayer case except for the results that turned out being different for bilayer graphene.

![Figure 6.4](image.png)

**Figure 6.4.** Theoretical heat flow between the electrons and phonons with different processes in bilayer graphene [73]. Here \( n = 10^{11} \frac{1}{cm^2} \) and \( T_{ph} = 0 \). The labels are as follows: “ac” acoustic phonons, “superc.” supercollisions, “opt.” optical phonons with contribution from different parts of the Brillouin zone (zone edge ZE or zone center ZC).

The different phonon processes in suspended bilayer graphene at relevant parameters, \( n = 10^{11} \frac{1}{cm^2} \) and \( T_{ph} = 0 \), reviewed in Ref. [73] are plotted in Fig. 6.4. It can be seen that simple acoustic phonon scattering and supercollision processes are overshadowed by the optical phonons that start to dominate around 200 K. Below \( T = 200 \) K, electronic heat diffusion is expected to exceed
the efficiency of all electron-phonon coupling processes, similarly to heat flow balance in monolayer graphene.

In the case of optical phonons, there are two different contributions originating from scattering near zone center (ZC) and edge (ZE, inter-valley scattering). The power density to the zone center (ZC) longitudinal and transverse optical modes in BLG, which have energies $\Omega_{ZC} \approx 0.2$ eV, can be estimated as in Ref. [95]:

$$P_{ZC}^{e-\text{op}} = \frac{18\pi^3 A^3 \gamma_0^2}{\pi \hbar v_F^4 \rho} \frac{\gamma_1}{\Omega_{ZC}} \left[ n_v(\Omega_{ZC}) - n_{op}(\Omega_{ZC}) \right] g(\mu, T_e), \quad (6.3)$$

where $A$ is the graphene flake area, $\gamma_0 = 42$ eV/nm [103], $\rho$ is the mass density of bilayer graphene, $v_F \approx 10^6$ m/s, and $n_v(\Omega_{ZC})$ and $n_{op}(\Omega_{ZC})$ are Bose distribution functions evaluated at temperatures $T_e$ and $T_{ph}$. Finally,

$$g(\mu, T_e) = \int_{-\infty}^{\infty} \frac{1}{4} (|x| + |x - 1|) [f(\Omega_{ZC}(x - 1)) - f(\Omega_{ZC}x)] dx, \quad (6.4)$$

describes the dependence on the chemical potential $\mu$. In the derivation it was assumed that the coupling between optical and acoustic phonons is not limiting the energy flow [104, 75], implying that the optical phonon temperature $T_{ph}$ remains constant.

Inter-valley scattering by zone edge (ZE) optical phonons also contributes to the heat current. In MLG, the ZE point optical modes dominate over ZC phonons in resistance [105, 106]. The results of Ref. [105] indicate $\sum \langle M_{ZE,j}^2 \rangle / \sum \langle M_{ZC,j}^2 \rangle = \Omega_{ZC}/\Omega_{ZE} \approx 1.3$ for the ratio of the angle-averaged squared matrix elements, which are relevant for the heat current. The corresponding power density is obtained by substituting $\Omega_{ZC}$ with $\Omega_{ZE}$ in Eqs. 6.3 and 6.4.

The total heat flow by optical phonons is the sum of these two contributions:

$$P_{e-\text{op}} = P_{e-\text{op}}^{(ZE)} + P_{e-\text{op}}^{(ZC)}. \quad (6.5)$$

At temperatures $T = 300 – 1000$ K, the two contributions are of the same order of magnitude, $P_{e-\text{op}}^{(ZE)} = 4 - 1.5 P_{e-\text{op}}^{(ZC)}$. Both of these contributions as well as the electronic conduction were taken into account when fitting the model.
Figure 6.5. a) Heat flow to the phonons for the data measured of a suspended bilayer graphene sample S1 in Ref. [73] (red markers), a model with optical phonons (blue solid line), and combined contribution of electronic conduction and optical phonons (black dashed line) presented as a function of electron temperature $T_e$. The data and models have been scaled by $T_e^3$. b) The heat flow $P$ presented as a function of chemical potential at $T_e = 600$ K. The data and the models are denoted the same way as in the panel a.

correction contribution (WF) fits very well to the measured data. The contribution from electronic conduction was determined by fitting $P = a T_e^2$-curves to the data at low temperatures $T_e < 150$ K and extrapolating them to higher temperatures.

The heat flow was also studied at different gate values (different chemical potentials). This data and the corresponding model are presented in Fig. 6.5b. As gating bilayer graphene is substantially harder compared to MLG, the optical phonon model displays extremely weak gate dependence. However, our data has small dependence on the chemical potential $\mu$ since the electronic heat conduction (WF) is influenced by gating.

In summary, electron-phonon coupling in suspended bilayer graphene seems more ordinary than in the monolayer case. Single optical phonon processes dominate exotic supercollisions at high temperatures, and the electronic conduction dominates all phonon processes at low temperatures. Ripples don’t seem to have a noticeable effect, possibly due to higher rigidity of BLG provided by its double layer structure.
Electron-phonon coupling
7. Transport through Corbino rings at high magnetic fields

Electron transport experiments on two-dimensional electron gas (2DEG) in magnetic fields, i.e., magnetotransport measurements, are typically conducted in specific well-established and symmetric geometries that simplify the analysis of conductivity/resistivity. The three main geometries are Hall bar, van der Pauw, and Corbino, all of which portray the field-induced phenomena slightly differently. Generally, the Hall bar geometry can be considered being the most complicated and the Corbino disk geometry the simplest, while van der Pauw is the intermediate one. This is reflected by the number of contacts to the two-dimensional electron gas, as well as the amount of information that can be extracted from the measurements, namely different components of the conductivity matrix.

However, the nature of measurements in the Corbino geometry differ from the other geometries. In the quantum Hall regime, continuous edge states connect different ports in the Hall bar and van der Pauw geometries. In the Corbino geometry this is not the case, and instead magnetoconductance always probes the bulk. In fact, on a quantum Hall state the current through the disk driven by a bias voltage decreases to a vanishingly small values as the diagonal conductivity approaches zero.

In this work, suspended graphene Corbino disk devices were developed for magnetotransport experiments, and this chapter reviews the experiments conducted on them. The first part presents the experiments dealing with the observation of integer and fractional quantum Hall effect (IQHE/FQHE), as well as the properties of composite fermions, the new particles emerging in a 2DEG undergoing FQHE. Second, experiments dealing with Wigner crystal order are presented.

7.1 Observation of integer and fractional quantum Hall effect

Even though one can only measure the diagonal conductivity $\sigma_{xx}$ in Corbino devices, and the conductance quantization of the counter-rotating edge states is beyond simple measurements, Corbino devices can be used to observe integer
Transport through Corbino rings at high magnetic fields

and fractional quantum Hall states and their properties. As explained in Sec 2.3, the diagonal conductivity $\sigma_{xx} \to 0$ on a quantum Hall state due to vanishing current. These conductance dips happen at certain filling factors, i.e. at certain charge carrier densities $n$ and magnetic fields $B$. This results in a fan-like structure in a $\sigma_{xx}(n,B)$ plot, known as the Landau fan diagram, where the Dirac point is at the epicenter of the fan. Fig. 7.1a displays such a Landau fan diagram for a measured monolayer suspended graphene Corbino sample. It can be seen in the figure that many quantum Hall states are developed already at very low fields down to $B = 100$ mT.

![Image of Landau fan diagram](image)

**Figure 7.1.** a) Logarithm of the conductance $\log_{10}(G)$ through a Corbino device presented as a function of applied back gate voltage $V_g$ and perpendicular magnetic field $B$. The colorbar on the right denotes the conductance scale $\log_{10}(G)$. b) The data in panel a differentiated w.r.t gate voltage $dG/dV_g$ presented as a function of the filling factor $\nu = \frac{en}{\hbar}$. The arrows mark the highest resolved filling factors with both charge carrier types.

In order to identify the states in Fig. 7.1a, the same data was differentiated w.r.t. $V_g$ and plotted as function of the filling factor. The mapping $dG(\nu,B)/dV_g$ is presented in Fig. 7.1b. In this figure, the quantum Hall states present themselves as vertical stripes at certain $\nu$. The highest resolved filling factors at both gate polarities $\nu = \pm 50$ are indicated in the picture. At these low fields $B < 2$ T, the states belong foremost to the main sequence $\nu = 4(n + 1/2)$, except for the broken symmetry states $\nu = 0$ and $\pm 1$ (caused by breaking of the spin and valley symmetries [52]) that are visible from $B \sim 0.5$ T upwards.

As the magnetic field is increased further, more broken symmetry and even fractional quantum Hall states (FQHS) appear. First appearance of the strongest FQHS $\nu = 1/3$ takes place at $B \approx 3$ T. However, the visibility of the FQHE in the conductance was significantly weaker than with the integer states due to smaller gaps associated with them. Another reason might be the fact that the fractional states observed actually originate from the localized states in the bulk. Thus, the relatively good conductivity on top of the fractional states is due to quantum tunneling between impurity islands housing the fractional states. Thus, the conduction on a fraction quantum Hall state can be viewed as a type of percolation through the disk.
In addition to the simple conductance measurements, transconductance measurements similar to Ref. [107] were conducted in order to better resolve the fractional states. With this technique, the sample was biased with constant DC voltage while the back gate was modulated with a low frequency AC signal (typically \( f = 17.77 \) Hz) that modulated the tunneling current. Thus, the device operated essentially as a field effect transistor with transconductance \( g_m \). As described in Ref. [107], this technique modulates the conductance of the quantum dot chain through the impurity islands directly, and can thus be a useful tool to image weak fractional states in particular. A mapping of the transconductance phase \( \arg(g_m(n,B)) \) around \( \nu = 1/2 \) at \( B = 6 \text{–} 8 \) T is presented in Fig. 7.2, and, as with the Landau fan diagram, a fractional state presents itself here as an inclined line with a certain slope. Consequently, states can be identified by determining slopes \( \nu \) of the observed lines.

The transconductance data was further analyzed in a similar manner as in Ref. [107] by looking at correlations in the data. The analysis is based on the fractional states presenting themselves as ridges that stick out from the background. Thus, the elements in the transconductance mapping \( A(n,B) \) can be summed together along inclined lines in a way described in the supplement of Ref. [11], there by obtaining a type of correlation spectrum \( C(\nu) \). This spectrum peaks at the filling factors that have high ridges, \( i.e. \) fractional states. Such correlation plot displaying the states around \( \nu = 1/2 \) is displayed in Fig. 7.2b.
7.1.1 Composite Fermions

The sequence of states observed fits to the well-known Jain sequence from the composite fermion theory [14, 108]:

\[ \nu = \frac{p}{2m p \pm 1}, \quad (7.1) \]

where \(2m = 2, 4, \ldots \) is the number of flux quanta attached to an electron, and \(p = 1, 2, \ldots \) is the filling factor of composite particles.

Essentially, the composite fermion theory reduces the complicated interacting many-body problem of fractional quantum Hall effect back to an effective single particle theory, more specifically to that of the quantum Hall effect of the new particles, the composite fermions. These new particles are constructed by attaching an even number \(2m\) of magnetic flux quanta to an electron. This procedure leads to renormalization of the effective magnetic field felt by the composite fermions. Essentially, each set of composite fermions with specific number of flux quanta \(2m\) has effective zero magnetic field at its center of sequence located at even-denominator fillings, such as \(\nu = 1/2\). The complete theory has been reviewed by many, see for example Refs. [108, 50, 109]. Besides rigorous theory, extensive experimental evidence of composite fermions exists. The results indicate that they indeed are real particles with properties that can differ from bare electrons [110].

In order to understand the composite fermions in graphene further, it is helpful to consider the effective field that the composite fermions feel. As given in Ref. [109], the effective field near \(\nu = 1/2\) is:

\[ B_{\text{eff}} = B - B_{1/2} = B - \frac{2hn}{e}, \quad (7.2) \]

where \(n\) is the charge carrier density (of electrons). One can also write Eq. 7.2 in terms of \(n\) and the filling factor \(\nu\):

\[ B_{\text{eff}} = \frac{hn}{ev} - \frac{2hn}{e} = \frac{hn}{ev}(1 - 2\nu). \quad (7.3) \]

The obtained effective field \(B_{\text{eff}}\) can be related to the effective mass of the composite fermions. This can be done by interpreting the quasiparticle pair excitation energy \(\Delta = k_B T_0\), determined from the Arrhenius plots, as the energy difference between Landau levels for the composite fermions:

\[ \hbar \omega_c^* = \Delta = \frac{\hbar e B_{\text{eff}}}{m_{\text{eff}}} \quad \rightarrow \quad m_{\text{eff}} = \frac{\hbar e B_{\text{eff}}}{\Delta}. \quad (7.4) \]

Thus, one can use the \(\sigma_{xx}(T)\) data presented in Fig. 7.3a to determine the effective mass as a function of the effective field. The result is presented in Fig. 7.3b. The effective mass is found to be around \(0.5 - 1\) electron rest mass and to diverge when \(B_{\text{eff}} \rightarrow 0\). These results are well in line with the previous GaAs 2DEG results [109].
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Figure 7.3. a) Temperature dependence of $\sigma_{xx}$ around half filling $\nu = 1/2$ at $B = 5$ T. The fractional states are marked by black arrows. b) Effective mass of composite fermions presented as a function of effective magnetic field extracted from the data in panel a. The markers denote the data points (the fractional states) and the solid lines are drawn in accordance to Eq. 7.4. The data in both panels is from the sample EV3 in Ref. [111].

Half filling temperature dependence

As seen in the above analysis, the $\nu = 1/2$ is a special filling factor even though there are no fractional quantum Hall states there. This is the point where the composite fermions do not feel magnetic field, even though the real external magnetic field can be very large, or at least high enough for composite fermions to form (the FQHE regime). Additionally, it can be seen in Fig. 7.3a that the temperature dependence is very weak there, much weaker than anywhere else. The behavior at $\nu = 1/2$ can be explained by formation of a composite fermion Fermi sea analogous to the situation at the zero magnetic field where the Fermi sea of electrons is formed. When the magnetic field is increased, the particles undergo integer quantum Hall effect. In the case of electrons, the result is the integer quantum Hall effect, and with the composite fermions the fractional quantum Hall effect (the integer quantum Hall effect of composite fermions). This explains the relative strength of temperature dependence around $\nu = 1/2$. The fractional states have gaps across which excitations can be thermally activated, and thus exponential temperature dependence in $\sigma$ is observed. At half filling on the other hand there is no gapped state that would lead to strong temperature dependence, and consequently weaker processes can dominate there.

In the following, the weak logarithmic temperature dependence due to weak localization is considered. As stated in Ref. [34], the weak localization effects lead to temperature dependence of form $\sigma(T) \propto \pm \ln(T)$, where the $+$ and $-$ denote the weak localization and antilocalization, respectively. As discussed in Sec. 2.1.3, the Dirac nature of electrons in graphene leads to the weak antilocalization behavior, which has indeed be seen in the experiments as the particular type logarithmic temperature dependence of $\sigma(T)$ with negative slope [112]. This raises an interesting question: do composite fermions in graphene exhibit weak antilocalization like electrons in real zero magnetic field?

In order to answer this question, the $T$ dependence at $\nu = 1/2$ was investigated
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carefully in several samples. However, now one needs to consider not the measured conductivity of electrons but the resistivity of composite fermions themselves. By following the discussion in Ref. [14], the composite fermion resistivity $\rho_{yy}^{CF}$ can be expressed as:

$$\sigma_{xx} = \frac{\rho_{yy}^{CF}}{4 + \rho_{xx}^{CF} \rho_{yy}^{CF}},$$  \hspace{1cm} (7.5)

which simplifies to $\rho_{xx}^{CF} = 4\sigma_{xx}$ when $\rho_{xx} << 1$ (the experimentally relevant limit). The temperature dependence can now be plotted directly for composite fermions, in order to see the sign of this dependence (the sign of quantum corrections). The $T$-dependence is displayed for two different samples in Fig. 7.4.

![Figure 7.4](image)

**Figure 7.4.** Temperature dependence at half filling $\nu = 1/2$ for two different samples (green and yellow markers). Two y-scales denote the real conductivity $\sigma_{xx}$ and the composite fermion resistivity $\rho_{xx}^{CF}$. The solid lines denote the theoretical predictions in accordance with Eq. 7.6 where $\lambda_p = 0.02$. The data are from samples EV2 and EV3 in Ref. [111]

It can be seen in the figure that the temperature dependence is logarithmic. The following model was fitted to the data successfully:

$$\rho_{xx}^{CF} = \lambda_p \frac{h}{e^2} \log T + \text{const.},$$  \hspace{1cm} (7.6)

where the parameter $\lambda_p = 0.02$ was used in the fits.

The positive sign of $\lambda_p$ is the exact opposite result compared to the GaAs 2DEG measurements in Refs. [113, 114] where weak localization of composite fermions was reported. On the same basis, one can conclude that the temperature dependence in these graphene Corbino samples is due to weak antilocalization. One might say that this is a trivial result since the electrons in graphene display weak antilocalization, and at their simplest composite fermions are just electrons with flux attached to them, so they should also display weak antilocalization. However, the situation is not that simple even in a conventional 2DEG GaAs where the charge carriers in zero magnetic field are not Dirac particles. However, recently it was theoretically suggested that the composite fermions in GaAs are Dirac particles [115]. Thus, the indirect experimental proof presented here about the Dirac nature of graphene composite fermions is far from trivial.
7.1.2 Unconventional fractional quantum Hall states

In addition to the regular Jain sequence, the conductance data in Ref. [11] at high fields $B > 6.5$ T displayed extra peaks in between the expected Jain states, see Fig. 7.5a. These peaks were identified being caused by the so called unconventional fractional quantum Hall states. While the FQHE was earlier considered as the integer quantum Hall effect of composite fermions, the unconventional states can be understood as the fractional quantum Hall effect of composite fermions [116], essentially "the second order fractional quantum Hall effect". These states reside at the filling factors:

$$\nu' = \frac{\nu}{2m \nu \pm 1},$$

(7.7)

where $2m = 2, 4, ...$ is number of flux quanta tied to a composite fermion, and $\nu$ is the filling factor of the fractional state undergoing quantum Hall effect.

Specifically, when one sets $m = 1$ and $\nu = 4/3$ and $5/3$ in Eq. 7.7, one obtains $\nu' = 4/11$ and $4/13$, respectively. Peaks were observed on these filling factors in the experiment.

In order to further characterize these states, their gaps were extracted from the Arrhenius plots (see Fig. 7.5b). It can be observed that the gaps of the unconventional states are rather small compared to the conventional state $\nu = 1/3$. In fact, the ratio of the gaps is $\Delta_{1/3}/\Delta_{4/11} \approx 32$. This is relatively close to the theoretical expectation $\Delta_{1/3}/\Delta_{4/11} \approx 50$ calculated in Ref. [116].

7.2 Wigner crystallization

It was predicted by Eugene Wigner already at 1934 that electrons situated in a uniform neutralizing background will form a crystalline phase under certain
critical density as temperature is lowered sufficiently [117]. This phase has been realized in many systems, such as semiconducting structures and in a layer of electrons accumulated on top of liquid helium, as schematically illustrated in Fig. 7.6. In addition to this spontaneous form of Wigner crystallization, the Wigner crystal can also be induced by a large magnetic field. In this field-induced Wigner crystal the Coulomb interactions that are also responsible for the formation of the fractional quantum Hall liquid favor the solid phase [56]. This phenomenon has been extensively investigated in GaAs 2D electron gas and its heterostructures [57].

Since kinetic and potential energies in graphene scale similarly with \( k_F \) due to linear dispersion relation, spontaneous Wigner crystallization is not possible in it [119]. However, equivalently to GaAs 2DEG, the magnetic-field-induced Wigner phase should form at sufficiently clean samples and high magnetic fields. However, previous measurements have failed to show clear signs of Wigner crystallization even with suspended graphene devices. The reason for this absence of Wigner crystallization in the experiments is unclear, especially since many fractional quantum Hall states have been seen in the same measurements demonstrating excellent sample quality. A possible cause for this result is the usage of too invasive techniques, such as scanning probes [120] that could have disturbed the delicate Wigner crystal order. Moreover, in the transport studies the high quality samples have utilized Hall bar, van der Pauw geometries, or two-terminal bridges that all involve transport along edge states at high magnetic fields. It’s possible that the effect of these edge states might prevent the observation of Wigner crystal ordering, for example certain type of edges have been shown to produce magnetic ordering [121] that could be detrimental for an experiment attempting to probe Wigner crystal order. In this work, suspended graphene Corbino devices were used to directly probe the whole bulk of graphene were the Wigner crystal should form, without complications from edge states or interaction with invasive probes. Three different methods were used: temperature dependence of conductivity, \( IV \) characteristics, and RF absorption spectroscopy.
In conventional 2DEG, the Wigner crystallization has been observed to happen at filling factors slightly below $\nu = 0.2$. Since the only relevant difference between the 2DEG and graphene is the permittivity of the material, one can expect a Wigner crystal to form in graphene around the same filling factors. In Corbino samples, the Wigner crystal should present itself as highly insulating phase below the melting temperature, followed by rapid growth of conductance above it. This is exactly what was observed in the measurements at $\nu = 0.15$ at $B = 9\, T$, as can be seen in Fig. 7.7.

![Figure 7.7. Conductance through the Corbino ring at filling factor $\nu = 0.15$ presented as a function of cryostat temperature $T$ at magnetic field $B = 9\, T$. The solid line is a linear trend line used to approximate the melting temperature of the Wigner crystal, $T_m = 1.4\, K$.](image)

In addition to the conductance measurements, $IV$ characteristics were measured in the Wigner regime. As seen in Fig. 7.8, the current-voltage characteristics in the Wigner state differ significantly from the $IV$ curve measured in the fractional quantum Hall state $\nu = 1/3$ at the same field. In order to further understand the result, the $IV$ curves were fitted with different transport models. It turns out that the fractional state data can be explained well using a simple model of quantum tunneling over a chain of localized states with linear $IV$ characteristics, for further information see Ref. [11].

On the other hand, in the Wigner regime the $IV$ characteristics are best explained by a model of thermally activated depinning of the Wigner crystal. According to Ref. [122], the Wigner crystal can be pinned to impurities in the platform the crystal is formed on. The exact nature of the disorder is not necessary adatoms left from fabrication, but for example corrugations in graphene (dimples) could provide the needed pinning potential as well. The existence of a pinning potential is necessary, since without tying the Wigner crystal down to something, it would slide and the insulating state would be lost. Even with pinning, relatively good conduction should be achievable if one excites electrons sitting in the potential at their characteristic resonance frequency or removes the gap using sufficiently high bias/temperature. For further details see the supplemental material of Ref. [11]. In spite of the exact nature of the pinning potential, the thermally activated depinning of Wigner crystal produces
a current $I_W$ when biased with a voltage $V$:

$$I_W = e f_a \left\{ \exp \left(-\frac{\bar{\Delta} - eV/2N}{k_B T}\right) - \exp \left(-\frac{\bar{\Delta} + eV/2N}{k_B T}\right) \right\},$$  \hspace{1cm} (7.8)

where $f_a$ is the depinning attempt frequency, $\bar{\Delta}$ is the depinning potential, and $N$ denotes the number of crystallites in the percolation path that facilitates conduction. In Fig. 7.8, the fit at $\nu = 0.16$ is obtained using the parameters $f_a = 3.0 \text{ GHz}$, $\bar{\Delta} = 140 \text{ } \mu\text{eV}$, $N = 7$, and $T = 0.2 \text{ K}$. The depinning potential implies that the melting temperature is $T_m \approx 1.4 \text{ K}$.

![Figure 7.8. IV characteristic of a Corbino device at \( \nu = 0.16 \) (blue markers) and \( \nu = 1/3 \) (red markers), \( V_T \) denotes the threshold voltage of the Wigner crystal. The black line denotes the thermal depinning model of Eq. 7.8 and the yellow line denotes linear IV characteristics of quantum tunneling.](image)

In addition to the IV characteristics, the RF absorption spectrum was measured by inputting a high frequency signal to the middle contact of the Corbino device and simultaneously measuring the DC current through it using a voltage bias of 50 $\mu\text{V}$ (see Appendix A.2 for details). The spectra presented in Fig. 7.9a were obtained by sweeping the RF frequency in the range of 0.01-5 GHz while recording DC current $I$ through the device. The idea behind this measurement was that when the fed RF frequency $f$ is around the resonance frequency of the pinning potential, increased conduction is observed. If there is no Wigner crystal no resonance should be observed in the range of investigated frequencies due to lack of rigidity in the electron system, as previously concluded in similar experiments with GaAs [123]. Indeed, the data in Fig. 7.9a displays a clear resonance only around filling factors $\nu = 0.15 - 0.16$ where a peak at 3 GHz is seen. Note that the frequency matches the depinning attempt frequency obtained from the IV measurements.

As mentioned earlier, without pinning the lattice would start sliding and no insulating phase would be observed. However, in real samples there are always imperfections leading to pinning. In graphene, two mechanisms can pin the lattice: impurities and "dimples" (corrugations in graphene). In addition to immobilizing the crystal, these pinning sites also divide the crystal into segments, crystallites. The average size of the crystallites was approximated in
Ref. [124] as:
\[ L = \left( \frac{2\pi c}{neBf_p} \right)^{1/2}, \]  
(7.9)
where \( c = 0.245e^2n^{3/2}/4\pi\epsilon_0\epsilon_r \) is the shear modulus of a 2D solid. Using Eq. 7.9 and the resonance frequency of 3 GHz at \( \nu = 0.15 \), one can estimate the domain size \( L \approx 0.7 \mu m \) in these measurements. This is smaller but comparable to the ring width 1.15 \( \mu m \) of the measured sample.

![Graph](image)

**Figure 7.9.** a) DC current through the Corbino ring as a function of RF input frequency at four different filling factors: \( \nu = 1/3 \) (red), \( \nu = 0.16 \) (green), \( \nu = 0.15 \) (blue) and \( \nu = 0.12 \) (yellow). The RF input power was -68 dBm and the DC bias 50 \( \mu V \) in all measurements. b) DC current through the device at \( \nu = 0.15 \) presented as a function of temperature when RF excitation of -68 dBm at \( f = 3 \) GHz was applied.

The temperature dependence of the pinning resonance was also investigated and the result is presented in Fig. 7.9b. The initial increase of current here is due to thermally assisted activated transport due to RF input. However, when the Wigner crystal melts at \( T_m \approx 1.5 \) K, this contribution is subsided. Thus, the current will actually decrease at higher temperatures. Recently, very similar result was obtained in GaAs [125].

To sum up, the three methods used to investigate the Wigner crystal in the Corbino devices seem to consistently indicate the presence of a Wigner crystal with the melting temperature of \( T_m \approx 1.4 \) K. This is quite a bit larger than values typically measured in GaAs 2D electron gases, in which \( T_m \) around 100-200 mK has been observed [125]. However, since the Coulomb interaction is the only interaction in the system and its energy scales as \( \sim 1/\epsilon_r \), one would expect graphene to have higher \( T_m \). The relative permittivities in the two materials are \( \epsilon_{r, GaAs} \approx 12.9 \) and \( \epsilon_{r, graphene} \approx 3 \). Additionally, the classical theoretical estimate for \( T_m = e^2/\pi(4\pi\epsilon_0\epsilon_r k_B \Gamma) \approx 0.8 \) K for graphene [126]. Thus, the measurement results seem to be approximately in line with the expectation regarding field-induced Wigner crystallization in graphene.
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8. Breaking of quantum Hall effect at the filling factor $\nu = 0$

Breaking of quantum Hall effect was investigated in the case of the $\nu = 0$ quantum Hall state. This regime was chosen because of two reasons. First, the gap of this state is quite large, even at relatively low magnetic fields, making the quantum Hall breakdown studies convenient to conduct. Second, the $\nu = 0$ state resides in the middle of the zero-energy Landau level making it special in many ways. The lowest Landau level in graphene is shared between electrons and holes, and at $\nu = 0$ electrons and holes populate the state equally. On top of that, the $\nu = 0$ state itself is quite exotic, it is currently believed to house a canted antiferromagnetic (CAF) phase [53]. Here this state was investigated by measuring the $IV$ characteristics and noise near the dielectric breakdown, the point where the highly insulating state starts to conduct under applied voltage.

8.1 Zener tunneling

By definition, insulators do not support flow of electric current. However, at sufficiently high critical voltage denoted by $V_{cr}$ any insulator starts to conduct. This phenomenon is known as the dielectric breakdown. Typical mechanisms facilitating the breakdown in many materials include the avalanche breakdown and thermal instabilities. With the avalanche breakdown, a small number of electrons can excite a large number of others through impact ionization that cascades into an avalanche. In thermal instabilities, a small number of current-carrying electrons locally heat up the material thereby exciting more and more charge carriers, which in turn heat the material more until steady conducting state is reached.

In a system with discrete energy levels, an insulating state may under certain circumstances be broken by tilting the energy bands using a bias voltage. This is the case in a reverse-biased Zener diode where the conduction band in the $n$-type semiconductor is aligned with the valence band in the $p$-type semiconductor by applying bias to shift the bands with respect to each other. Above the critical voltage $V_{cr}$, electrons from the $n$-side conduction band can tunnel to the valence band on the $p$-side, while holes move in the opposite way.
As discussed in Sec. 2.3, discrete energy levels called the Landau levels are formed in graphene at high magnetic fields. At sufficiently high fields, Landau sublevels corresponding to broken symmetry states, in which spin and valley degeneracies are lifted, are formed. The states \( \nu = 0 \) and 1 are examples of such states. Tunneling from one level to another can be achieved by tilting the potential with bias voltage in a similar manner as in a Zener diode. This scheme can be achieved in the Corbino geometry by biasing the device in which edge states at the inner and outer edges of the ring are separated by insulating bulk. A schematic view of the two relevant tilted Landau sublevels along the radial direction is presented in Fig. 8.1. Tunneling current starts to flow when the tilting bias equals the gap \( \Delta \) between the levels and an electron can tunnel into the empty upper band and a hole is left in the lower level. Mobile charges are driven to opposite directions by the applied electric field. Altogether, the situation is very similar to the semiconductor Zener diode presented above.

![Figure 8.1. Schematic view of two Landau sublevels in the radial direction of Corbino ring separated by a gap \( \Delta \) and tilted by an applied bias. The horizontal arrow denotes Zener tunneling from the lower Landau sublevel to the higher. Spatially these points are separated by the ring width \( r_o - r_i = d \).](image)

As stated in Ref. [127], the Zener tunneling current between two bands is generally of the form:

\[
I \propto e^{-\frac{V_Z}{V}}, \tag{8.1}
\]

where the parameter \( V_Z \sim \frac{\Delta^2 d}{e \sqrt{E_b a}} \) with its magnitude comparable to the critical voltage of the dielectric breakdown, \( V_Z \sim V_{cr} \). The parameter \( E_b \) denotes the width of the Landau sublevels, \( d \) is the dielectric thickness (the Corbino ring width), and \( a \) corresponds to the lattice constant.

However, the expression in Eq. 8.1 does not take into account the circular motion that the electrons undergo at high magnetic fields due to the Lorentz force. Thus, instead one needs to consider the center of mass motion of the circulating electrons, which leads to vortex-like equations of motion more commonly associated with vortecies in superfluid helium and type-II superconductors. However, with this gyrotropic motion taken into account, as derived in Ref. [127], the tunneling current takes nearly the same form as before but with a modified exponent:

\[
I \propto e^{-\frac{(V_Z/V)^2}{2}}, \tag{8.2}
\]
where \( V_Z = \frac{eBd}{\sqrt{8\pi\epsilon\Phi_0}} \) is a parameter with magnitude of the order of the critical voltage. \( \epsilon = \epsilon_0\epsilon_r \) is the permittivity in graphene, and \( \Phi_0 = \hbar/e \) is the single electron flux quantum.

In addition to the Zener tunneling models, one needs to consider possible heating effects, such as thermal activation, for the quantum Hall breakdown. However, it became evident already at the early stages of the measurements that the regular activation formula (Arrhenius equation) with the large gap of \( \nu = 0 \) state would predict too fast current growth at the border of the onset compared to the data. Thus, a model taking into account percolation through \( N \) localized states in the bulk was considered:

\[
I = I_0 \left[ e^{-\frac{\Delta V/N}{k_B T}} - e^{-\frac{\Delta V/N}{k_B T}} \right],
\]

(8.3)

where \( \Delta \) is the \( \nu = 0 \) state gap, and \( T \) the average system temperature.

Figure 8.2. a) Tunneling current through a Corbino ring (blue markers) at \( B = 2 \) T presented as a function of the applied bias voltage \( V \). The solid lines denote different models with indicated parameters: gyrotropic Zener tunneling (red), regular Zener (green), and thermal activation percolation model with \( N = 6 \) (black). The inset displays a back and forth sweep (blue markers and red line) under the same conditions as a hysteresis check. b) Parameters \( V_Z \) and \( V_{cr} \) extracted from the fits, such the one in panel a, presented as a function of perpendicular magnetic field \( B \). The inset displays an Arrhenius plot for \( \nu = 0 \) state at \( B = 1.8 \) T, where the red line denotes the model \( \sigma = \sigma_0 \exp\left(-\Delta_0/k_B T\right) \) with \( \Delta_0 = 1.2 \) meV.

Fig. 8.2 displays the tunneling current through the Corbino ring at \( B = 2 \) T, as well as the three models considered above. The gyrotropic Zener model is found to yield the best fit, as well as the most reasonable parameters used in the fitting. While the critical voltage was \( V_{cr} = V(I = 10 \) nA\( ) = 10 \) mV, the parameter \( V_Z = 21.5 \) mV in the fit, which is in the line with the expectation \( V_{cr} \sim V_Z \). With the regular Zener tunneling with \( I \propto \exp(-V/V_Z) \) the fit is worse and the parameter \( V_Z \) is an order of magnitude larger than \( V_{cr} \), which makes this model worse for explaining the data. Moreover, the basic thermal activation model does even worse in terms of fitting. Additionally, the back and forth sweep of the bias voltage didn’t show any signs of hysteresis (see Fig. 8.2a inset) that one would expect if local heating and activation would have been involved.

The gyrotropic Zener tunneling model also fits well to the field dependence of \( V_Z \) at least up to \( B = 7 \) T, as seen in Fig. 8.2a. As predicted by the model, the
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parameter $V_Z$ is found to grow as a function of field as $V_Z \propto B$, as is the case with $V_{cr}$ too. In conclusion, the gyrotropic Zener tunneling is found to explain the observations and facilitate the breakdown of quantum Hall effect at $\nu = 0$, at least in the range $B = 1 - 7$ T.

### 8.2 Noise and breaking of the quantum Hall state

In order to further understand the details regarding the breakdown of the $\nu = 0$ quantum Hall state under bias, noise measurements at low and high frequencies were employed along with the $IV$ measurements. The noise was measured at low frequency $f \leq 10$ Hz and at microwave frequencies ranging over $f = 600 – 900$ MHz. This work extends the analysis of the previous section beyond the gyrotopic Zener tunneling that was found to facilitate the quantum Hall breakdown at small currents and magnetic fields below $B = 7$ T.

Fig. 8.3 displays an $IV$ trace and simultaneously measured low frequency noise $S(I, f = 10$ Hz$)$ at $B = 5.6$ T. The $IV$ trace is accompanied by a theoretical curve representing expected gyrotopic Zener tunneling from Eq. 8.2. It can be seen that while the gyrotopic Zener tunneling model works well at small currents, it fails above $I = 500$ pA where the current grows extremely fast. Interestingly, the noise starts to grow as $S_I \sim I^2$ at the same point, as seen in Fig. 8.3. Similarly, the rapid current growth and the noise increase seem to stop at the same current too, i.e. at $I = 30$ nA.

![IV curve on the $\nu = 0$ state at $B = 5.6$ T](image1)

![Low frequency noise at $f = 10$ Hz measured concurrently with the IV data in panel a. The black dashed line denotes $I^2$ behavior, and the arrows mark the same currents as in panel a.](image2)

**Figure 8.3.** a) $IV$ curve on the $\nu = 0$ state at $B = 5.6$ T: the blue markers are data points and the red line denotes the contribution from the gyrotopic Zener tunneling from Eq. 8.2. The arrows denote the end points of the Zener and avalanche processes. b) Low frequency noise at $f = 10$ Hz measured concurrently with the $IV$ data in panel a. The black dashed line denotes $I^2$ behavior, and the arrows mark the same currents as in panel a.

As already discussed in the previous section, the gyrotopic Zener tunneling seems to facilitate the quantum Hall breakdown at low fields, but there seem to be deviations from the theory at high fields. These deviations and the above noise and $IV$ characteristics can be explained by a transition into an avalanche
type of transport above the Zener tunneling regime that functions as a precursor for it. In this scheme, an electron creates additional current through impact excitation, and consequently the charge is transported in pulses with duration $\tau_s$ separated by $\tau_0$. As discussed in Sec. 2.2.1, this leads to the generation of switching noise that depends on the current as $S_I \propto I^2$, which is exactly what is observed.

As discussed in Refs. [128, 129, 71], the measured switching noise magnitude can be used to estimate the number of electrons in a single pulse $N_{tot}$. This is done by defining the low frequency Fano factor $F_{AV}$ as $S_I = F_{AV} 2e \langle I \rangle$, where $F_{AV} = 2N_{tot}$. From the measurement at $B = 5.6$ T, one can extract $F_{AV} = 10^4$, which indicates significant electron bunching with $N_{tot} = 5 \times 10^3$ electrons in one current pulse. Additionally, one can also estimate the average time between pulses $\tau_0$ by taking the switching noise maximum and comparing it to the theory. In Ref. [71], it was estimated as $\tau_0 = 100$ ns.

When the bias is increased further, the current burst get longer, i.e. $\tau_s$ increases. Eventually, the time scales become nearly equal $\tau_s \sim \tau_0$ (overlapping of pulses) and the transport cannot be considered pulsed any longer. This is the point of transition towards the Ohmic regime seen in the data at $I = 30$ nA, where the noise also decreases momentarily before the contact noise takes over.

![Figure 8.4](image.png)

**Figure 8.4.** Shot noise Fano factor as a function of the tunneling current at $B = 6$ and 8 T. The arrows denote the end of the avalanche region in both cases.

The simultaneous shot noise measurement at microwave frequencies supports the model presented above. Since the Lorentzian spectrum produced by the switching decays as $1/\omega^2$ and has characteristic time scales of the order of 100 ns, the shot noise spectrum should not be affected by the switching noise. Instead the spectrum should be flat, i.e. as the spectrum of shot noise should be. In fact, the shot noise measurement probes the noise inside the current pulses. Fig. 8.4 displays the Fano factor extracted from the shot noise measurements at $B = 6$ and 8 T where the $IV$ characteristics were very similar to the data discussed above. It can be seen that the Fano factor is $F \leq 1$ within the error bars indicating that there is no electron bunching. Instead, we have $F < 1$ in the avalanche regime (below $\sim 10$ nA), which indicates correlation between electrons within the pulses. At the end of the avalanche regime, the noise reaches $F \approx 1$.
Breaking of quantum Hall effect at the filling factor $\nu = 0$

the fully Poissonian value, or slightly above it that might be an indication of multisite generation (electron multiplication at small scale). After the avalanche regime, the Fano factor decreases to value $F \approx 0.5$ for a while, possibly indicating hot-electron transport. Finally, when the bias is increased further inelastic processes, namely the electron-phonon coupling, suppress the noise.

All in all, the avalanche type of transport and the consequent switching noise seem to explain well the part of measured data where the gyrotropic Zener tunneling model fails, i.e. at high magnetic fields. The observations are also in line with previous measurements in GaAs Corbino rings [130] and with larger graphene Hall bar devices grown on SiC [131] where the quantum Hall breakdown was explained using bootstrap-type electron heating (BSEH) models. In the BSEH model, the origin of the initial electron excitation is electron heating that in our case is most likely caused by the gyrotropic Zener tunneling. Thus, it is natural that the BSEH behavior can overpower Zener tunneling only when the gap is large and large bias voltages are used to break it. Naturally, the large bias leads to thermal excitation only at sufficiently high currents, and the BSEH process is seen to be activated only at high bias and at high magnetic fields. In fact, the local temperature and not the input power seems to be the prerequisite that determines the onset point of the BSEH behavior, as discussed in Ref. [71].
A. Appendix

A.1 Graphene sample fabrication

A.1.1 Rectangular samples on LOR

Fabrication using the so called LOR-method followed a procedure adapted from Ref. [58]. This method is based on two different resists with different selectivity: one for the lithography used to deposit the metal contacts (PMMA), and another for supporting the suspended structures (lift-off resist, LOR). The process is illustrated in Fig. 1.1.

**Figure 1.1.** Fabrication of a suspended graphene device on LOR: a) clean SiO$_2$ substrate b) spin LOR onto the substrate c) graphene exfoliation d) deposition of metal contacts e) suspension using e-beam f) bonding.

**Detailed fabrication procedure:**

1. Clean the substrate (280 nm of thermally grown SiO$_2$ on top of highly doped bulk silicon Si++) in hot acetone and an ultrasonic bath.
2. Spin two layers of LOR 3A at 5000 rpm speed for 60 s onto the substrate. Bake 5 min and 12 min at 200 °C after the first and second layer, respectively.

3. Exfoliate graphene from bulk graphite. First, thin graphite flakes by peeling top layers off with a blue wafer tape. Then, press the thinned graphite flakes on the tape against the LOR covered chip for 60 s, after which bake the chip on a hot plate at 100 °C for 2 min with the tape still on top. Let the chip cool down for 1 min after baking. Finally, peel the tape off of the chip.

4. Locate graphene flakes on the chip using an optical microscope. The contrast of graphene flakes against the background is based on an optical interference effect dependent on the dielectric thickness under graphene [66]. The contrast is quite small with the LOR residing between graphene and SiO₂, and consequently flakes might be visible only in certain parts of the chip where the LOR thickness is suitable.

5. Confirm the thickness of the located flakes using a Raman spectrometer operating at 633 nm. The width of the 2D peak tells the number of layers [132]. Typically, full width at half maximum is around 27 cm⁻¹ for a monolayer flake on LOR.

6. Spin PMMA double layer (PMMA 50k A11 / PMMA 950k A3) for the e-beam lithography. Spin at 6000 rpm for 60 s, bake 90 s at 180 °C.

7. Pattern the contacts and the bonding pads using a 20 kV e-beam (JEOL JSM-7100F SEM) with the doses 120 μC/cm² and 130 μC/cm², respectively.

8. Develop in xylene for 4 min, rinse in hexane for 60 s, and blowdry with nitrogen.

9. Deposit 5 nm of Cr (sticking layer) and 70 nm of Au using UHV e-beam metal evaporation with deposition rates 0.5 Å/s and 2.5 Å/s for Cr and Au, respectively.

10. Lift off excess metal in 80 °C xylene bath for 2 h. Assist the liftoff at the end by blowing xylene onto the chip with a syringe, gently. Rinse in hexane for 60 s and blowdry with nitrogen.

11. Expose the LOR under and around graphene to the 20 kV e-beam with a dose of 800 μC/cm².

12. Develop in ethyl lactate for 60 s, rinse in hexane for 60 s.

13. Lift the chip carefully from hexane with the chip edge facing up, it should emerge dry.

14. Mount the chip in a sample box by gluing it to the bottom with electrically insulating varnish.

15. Wire bond the bonding pads to microstrip lines on the box circuit board using bonding force of $F = 1.50 \text{ N}$. 
A.1.2 Suspended graphene Corbino devices

The LOR technique can also be used to fabricate suspended graphene devices in the Corbino geometry, as presented in Fig. 1.2. The first steps in the fabrication follow the same procedure as in the case of rectangular LOR devices until the liftoff, however, only one contact is deposited during the first lithography round. This first contact is the bottom electrode that defines the outer radius $r_o$ of the Corbino ring, see Fig. 1.2c. The subsequent steps are depicted in Fig. 1.2.

After the first metallization, another support layer of LOR (double layer LOR with thickness 500 nm) is spun to support the top electrode that bridges over the bottom electrode without touching it. Then, a hole is made to the top LOR layer by exposing the area of the middle contact to an e-beam with a dose of 800 $\mu$C/cm$^2$, developing in ethyl lactate for 60 s, and rinsing in hexane. For larger middle contacts the dose at the edge of the hole had gradient (five steps of ~40 nm wide each) in order to round the edge and make the metal deposition easier (smoother edge ensured continuity of the metal film). Note that LOR under the graphene flake is not washed away since there is no way for the developer to get in touch with it. Consequently, the top contact could be deposited using the same e-beam lithography and metal deposition processes as with the bottom electrode. However, thicker gold layer of ~100 – 120 nm was used in order to make sure that the film stays continuous at the edge of the hole in LOR (the middle contact to the graphene flake). The liftoff was done the same way as with the bottom layer and with the rectangular samples.

The graphene and the immediate contacts were suspended by exposing the LOR in the area to a 20 kV e-beam the same way as with the rectangular samples. However, slightly higher dose of ~1000 $\mu$C/cm$^2$ was used due to the thicker LOR layer. The development, rinsing, and drying of the sample were done as before. The bonding parameters remained the same as well.
Despite the rather complex fabrication procedure, the sample yield remained rather high. 40-50% of the samples didn’t collapse and showed conduction through the device. It was also found that even with samples that initially had MΩ-level resistances, reasonable sample quality could still be obtained using a low temperature current annealing procedure, see the next section. A bigger problem with the Corbino devices turned out to be the alignment of the middle contact. Small offsets were hard to avoid, and in the smallest samples they made the current annealing impossible due to large geometric asymmetry and consequent asymmetric heating. This ended up being the limiting factor for making smaller samples (with the outer diameter < 2 μm).

The advantage with smaller devices would have been higher tolerance for high gate voltages that in turn would have enabled observation of higher Landau levels at high magnetic fields. However, due to the distant back gate quite large voltages could already be used with more commonly used 3-4 μm devices. The breakdown voltage was tested with a monolayer device with dimensions: the outer diameter $D_o = 2.8 \mu m$ and the inner diameter $D_i = 0.9 \mu m$. The breakdown happened at $V_g = 176 V$ due to the graphene membrane collapsing, the middle contact was pulled down with it, see Fig. 1.3 for before and after images of such an even with a similar sample.

![Figure 1.3. SEM image of the same Corbino device a) before the collapse, b) after the collapse.](image)

### A.1.3 Current annealing

A crucial step in the preparation of suspended graphene samples is the current annealing. By passing a large current, of the order of 1 mA per 1 μm of sample width, graphene is heated to extreme temperatures, close to the point of breaking. The annealing is performed at low temperatures, below $T = 4 K$, so that the current leads act as heat sinks for the heat produced by Joule heating in graphene. Since the graphene flake accounts for most of the resistance in the system (the lead resistance of the order of 10 Ω to the bonding pads, and the graphene resistance of the order of 1 kΩ), rest of the system remains relatively cool. Typically the cryostat warmed up to $T = 200 mK$ from the base temperature of 10 mK when performing the annealing process. However, the main reason for performing the annealing at low temperatures was that cold parts around the hot graphene (the substrate, the contacts, and sample box walls) also functioned
as cryopumps for the material evaporated off of the graphene flake. Thus, as long as the cryostat was kept cold the removed dirt would not return back.

Contrary to the name, "current annealing", the applied voltage and not the current seems to be the relevant parameter in the process. Consequently, annealing was performed in the voltage bias configuration in this work. This was because of two reasons. First, with a constant current source the power dissipated in graphene would increase significantly in the situation where the graphene resistance grows significantly (shifting of the Dirac point as graphene gets cleaner). Thus, it is actually safer to fix the voltage and let the current be determined by the sample resistance. Second, voltage, not current, seems to be the quantity that defines the temperature in graphene. In fact, the electron temperature is directly proportional to the bias voltage, see discussion about noise thermometry in Sec. 3.2.2. Typically, the voltage required to anneal our suspended devices was $\sim 1 \text{ V}$ for the HF-etched samples and $\sim 1.6 \text{ V}$ for the LOR samples.

The annealing progress of an HF-etched sample is presented in Fig. 1.4. As can be seen in the figure, initially at room temperature the sample exhibits weak response to the external doping by the back gate, which implies strong doping caused by adsorbates. Our samples were usually p-doped, which is consistent with PMMA residue that is known to cause $p$ doping in graphene devices [133]. When cooled down to $T = 100 \text{ mK}$, the resistance is slightly lowered possibly due to decreased resistivity in the metal leads, but still the gate modulation remains weak indicating $p$ doping. After annealing, the device exhibits strong gate dependence with charge neutrality point very close to $V_g = 0 \text{ V}$.

Annealing of the Corbino devices followed the same procedure as with the rectangular samples. Typically, the positive annealing voltage was applied to the center contact, while the outer rim was grounded. The annealing voltage
where significant change started to happen was similar to the case of rectangular samples, however, usually higher voltages in the range 1.6 – 2.1 V were needed to fully clean the device. Devices with significant offset in the middle contact centering or very small devices with the outer diameter < 2 μm were typically impossible to anneal properly to the point where FQHE could be observed. Best results were obtained with 3 – 4 μm outer diameter devices.
A.2 Wigner crystal RF absorption spectrum measurement setup

The RF absorption by the electron system in the Corbino device was probed by inputting microwaves to the middle contact and simultaneously measuring the DC conductance through the ring. The sample was voltage biased from the room temperature using an HP 33120A waveform generator as voltage source. The output of the generator was stepped down with a 1:2000 homemade voltage divider resulting in a DC bias of $V = 50 \mu V$. The bias was fed to the center contact of the Corbino ring through a bias tee (mini-circuits ZX85-12G-S+), and the resulting DC current was measured from the outer contact through a similar bias tee. The DC current was amplified by a current preamplifier (Stanford Research SR570, sensitivity set to $10^{10} \text{ V/A}$) at room temperature before measurement with an HP 34410A multimeter.

The high frequency signal fed to the middle contact was produced by an Anritsu MG3692B RF generator. The input line had attenuation of $L = 50 \text{ dB}$ at $f = 1 \text{ GHz}$. This level was based on a simple approximation that attenuation required to reduce noise from a resistor at 300 K temperature to 10 mK:

$$L = 10 \log \left( \frac{300 \text{K}}{0.01 \text{K}} \right) \approx 44 \text{ dB}. \quad (1.1)$$

The DC conductance was recorded as a function of the input frequency in the range of 0.01-5 GHz at a constant power (in the range of -100 to -55 dBm at the sample).

![Simplified measurement setup](image)

**Figure 1.5.** Simplified measurement setup used to investigate RF absorption by Wigner crystal ordering in the graphene Corbino ring (light blue ring) hanging from the gold contacts (yellow parts). The parts below the light blue dashed line were in $T = 10 \text{ mK}$ and the others at room temperature.
References


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"To him who looks upon the world rationally, the world in its turn presents a rational aspect. The relation is mutual."
G. W. F. Hegel