Magnetotransport Studies of GaAs/AlGaAs Heterostructures, Epitaxial Graphene, and CVD Graphene, and Growth of CVD Graphene

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Magnetotransport Studies of GaAs/AlGaAs Heterostructures, Epitaxial Graphene, and CVD Graphene, and Growth of CVD Graphene

by

Tharanga Ranjan Nanayakkara

Under the Direction of Ramesh G. Mani, PhD

A Dissertation submitted in Partial Fulfillment of the Requirements for the Degree of Doctor of Philosophy in the College of Arts and Sciences Georgia State University 2022
The 2D confinement of electrons modifies the physical properties of the electronic system, and this has significant implications for electrons' interactions and kinetic behavior. A 2D electron system (2DES) exposed to a magnetic field at low temperatures provides an excellent platform for mesmerizing physical phenomena such as weak localization and weak anti-localization, giant negative/positive magnetoresistance, Shubnikov-de Haas oscillations (SdHOs), and quantum Hall effects. The photoexcitation of a 2DES has also introduced new phenomena such as radiation-induced magnetoresistance oscillations and zero resistance states. Exploring and understanding novel physical phenomena in 2DES would benefit future advanced semiconductor technology.

This dissertation is based on magnetotransport studies of the 2DES in GaAs/AlGaAs and graphene, and investigation of chemical vapor deposition (CVD) techniques for graphene growth. The effect of microwave (MW) radiation on electron temperature was studied by investigating the photo-excited transport at zero magnetic field and in the SdHOs regime where the cyclotron frequency $\omega_c$ and the MW angular frequency $\omega$ satisfy $2.3 < \omega_c/\omega \leq 5.2$. The results show small discernible electron heating under modest MW radiation, in agreement with theoretical predictions. Additionally, the activation energy at the odd integer filling factors in GaAs/ AlGaAs 2DES was examined using a novel technique of microwave induced heating instead of conventional temperature dependent measurements. We also performed magnetotransport measurements on epitaxial graphene samples, which show large longitudinal resistance values, and examined the Hall resistance values that shift away from the quantized values with increments of temperature. This dissertation also discusses the research efforts on growth and techniques of low pressure CVD graphene. A dual treatment technique was utilized
to lower the nucleation density of single crystal graphene grown on copper. Additionally, we
discuss an impurity-assisted growth mechanism which governs the growth of single-crystal
graphene via isotropic diffusion, producing two-fold, four-fold, and six-fold symmetries in the
resulting flakes. We also examine electrical transport measurements across a graphene p-n
junction formed in a single gated field effect transistor made from CVD graphene.

INDEX WORDS: Magnetoresistance, Two dimensional electron systems, Microwave induced
magnetoresistance oscillations, Shubnikov de Haas oscillations, Single crystal graphene, Low-
pressure chemical vapor deposition, Nucleation density, Surface oxidation
Magnetotransport Studies of GaAs/AlGaAs Heterostructures, Epitaxial Graphene, and CVD Graphene, and Growth of CVD Graphene

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August 2022
DEDICATION

To my loving family and friends.
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LIST OF ABBREVIATIONS

2D: Two dimensional
2DES: Two dimensional electron system
SdHOs: Shubnikov de Haas oscillations
QHE: Quantum Hall effect
MOSFET: Metal-oxide-semiconductor field-effect transistor
IQHE: Integer quantum Hall effect
FQHE: Fractional quantum Hall effect
MW: Microwave
ZRS: Zero resistance state
MIMOs: Microwave induced magnetoresistance oscillations
DC: Direct current
EG: Epitaxial graphene
VTI: Variable temperature insert
OVC: Outer vacuum chamber
GHS: Gas handling system
LPCVD: Low pressure chemical vapor deposition
CVD: Chemical vapor deposition
PDMS: Polydimethylsiloxane
AFM: Atomic force microscope
PMMA: Poly methyl methacrylate
1. FUNDAMENTALS OF TWO-DIMENSIONAL ELECTRON SYSTEMS

New physical properties needed for novel applications can be realized by reducing the effective dimensionality of electronic systems. Presently, scientists can produce two dimensional (2D), one dimensional, and even zero dimensional systems for electrons by confining the spatial dimensions. Among them, 2D electron systems (2DES) are more popular since they are realized in the field effect transistors, building blocks for electronic devices. This chapter mainly focuses on the fundamental properties of 2DES.

1.1 Introduction

The secret of the visibility of amazing electron properties in reduced dimensions is the electron confinement in a region comparable to the de Broglie wavelength. Let us consider confined carriers in a quantum well with infinite depth, and finite width $a$. From the basic quantum mechanics concepts, the quantized energy of carriers can be described as,

$$E_M = \frac{\pi^2 \hbar^2 M^2}{2m^*a^2}$$  \hspace{1cm} (1.1)

where $m^*$ is the effective mass of the carriers, and $M$ is an integer which is the quantum number of a given energy state. However, there is no infinite quantum well in finite sized quantized systems. Therefore, the quantized energy in a finite potential well can be expressed as [1],

$$E_M \sim \frac{\hbar^2}{m^*a^2}$$  \hspace{1cm} (1.2)

If the carriers are confined in $z$-direction only, they can move freely in the $xy$-plane. In that case, the total energy of a finite sized quantized system can be written as,
\[ E = E_M + \frac{(p_x^2 + p_y^2)}{2m^*} \]  

where \( p_x \) and \( p_y \) are the momenta along the \( x \) and \( y \) directions, respectively. Restricting carriers' motion in a quantum region doesn’t alone provide the opportunity to observe the quantum effects due to size quantization. Due to quantum size effects, a large enough energy level separation is another requirement to observe energy quantization. Furthermore, the thermal energy of the carriers must be less than the separation in two consecutive energy levels.

\[ k_B T \ll E_{M+1} - E_M \]  

Carriers in a real system always experience scattering from impurities, defects, phonons, etc. The single particle lifetime \( \tau_s \), is utilized to characterize the scattering probability of a given quantum system. Note that \( \tau_s \) gives the average lifetime of carriers in a quantum state. The Heisenberg uncertainty principle states that \( \Delta t \cdot \Delta E \sim \hbar \). Thus, the uncertainty of energy in a given state \( \Delta E \sim \frac{\hbar}{\tau_s} \) for a finite \( \tau_s \)[1]. Furthermore, the energy gap between consecutive levels in quantized system satisfies,

\[ \frac{\hbar}{\tau_s} \ll E_{M+1} - E_M \]  

In addition, carrier mobility is proportional to the transport lifetime since \( \mu = e\tau/m^* \). Therefore, the system should have high carrier mobility, high surface quality, thin layers, lower temperature, and low enough carrier concentration to demonstrate the quantum size effects.

1.2 Semiconductor Heterointerfaces

The 2DES in heterostructures provides an excellent platform to examine the single particle and many particle properties of nanoscale systems. Heterointerfaces are formed by contacting the semiconductor materials with different band gaps.
Figure 1.1 A schematic diagram of a typical energy band diagram of a heterointerface at n-type and p-type semiconductors. There is a bandgap offset at the interface due to the different bandgaps in the two materials. Here, $\Delta E_c$ is the difference of the electron affinities ($\chi$) of two materials and it is $\Delta E_c = \chi_2 - \chi_1$. Where $\Delta E_{g1}$ and $\Delta E_{g2}$ are band gap between conduction and valence bands of pure materials. Here, $\Delta E_v = (E_{g1} - E_{g2}) - \Delta E_c$.

Figure 1.1 shows a typical energy band diagram of a heterointerface that is formed in between n-type and p-type semiconductors. The 2DES is formed at the heterointerface, and it could have several types of scatterers, such as ionized impurities, phonons, roughness, etc. These scatterers are major issues to approach a high quality 2DES due to the interactions with charge carriers. However, scattering in heterostructures can be controlled by utilizing two semiconductors with a very smooth interface, such as GaAs/AlGaAs heterostructures. In bulk semiconductors of temperature $T$, the carrier mobility ($\mu$) depends on the impurity scattering in the order of $\mu \sim T^2$ and acoustic phonon scattering in the order of $\mu \sim T^{-3/2}$, while impurity scattering and acoustic phonon scattering in 2DES have mobility $\mu$ dependencies proportional to $T^{3/2}$ and $T^{-1}$, respectively. Hence, phonon scattering can be reduced by placing heterostructures at low temperatures. However, impurity scattering remains at low temperatures. To minimize the ionized impurity level, one can try reducing the doping level, but it will also reduce the
concentration of electrons. Therefore, the modulation doping technique has been proposed to tackle this situation.

![Diagram](image)

*Figure 1.2 A sketch of the conduction band structure of a modulation doped heterojunction representing the confined state at the interfaces, spacer (d), and the Fermi energy (E_F).*

Generally, in modulation doped heterojunctions, the wide band-gap material is doped while the narrow band-gap material is un-doped. Some carriers from the wide band-gap semiconductor side migrate to the narrow band-gap semiconductor side while creating an electron layer near the interface to balance the chemical potentials in both semiconductors. Now the ionized impurities and electron layer are farther apart from each other. This separation promotes high carrier mobility in the 2DES due to less interaction between ionized impurity and electrons. The energy band diagram of modulation doped heterostructure is depicted in Figure 1.2.

### 1.3 GaAs/AlGaAs Heterostructures

Gallium (Ga) and Aluminum (Al) are elements from column III, while Arsenide (As) is an element from column V in the periodic table. III-V semiconductor compounds spatially
distribute as zinc-blende crystal structures. GaAs and AlGaAs have a very sharp interface due to their similar lattice constants, which minimizes the lattice errors due to crystal strain. GaAs has a band-gap 1.42 eV while AlAs has a larger band-gap 2.16 eV. The band-gap of Al$_{1-x}$Ga$_x$As is between 1.42 eV and 2.16 eV and it is determined by concentration $x$. The Fermi energy in the AlGaAs side is higher than the GaAs side initially. When the heterojunction approaches the equilibrium, Fermi energies on both sides are aligned at the same level by migrating electrons from AlGaAs side to GaAs side. The electrostatic potential will bend the energy bands, as illustrated in Figure 1.2. At equilibrium state, the Fermi energy is a constant everywhere in the heterostructure, and at the interface, the conduction band creates a triangular quantum well on the GaAs side. Since electrons movements are confined to a 2D plane in the triangular quantum well, it is known as 2DES.

1.4 2DES in a magnetic field

An externally applied magnetic field can be resolved into two components with respect to the plane of 2DES. Those two components are labeled in-plane magnetic field parallel with 2DES and out of plane magnetic field, perpendicular to the 2DES. The out of plane magnetic field component changes the energy spectrum of 2DES, while the parallel field does not change the energy spectrum qualitatively. However, the parallel field component changes both effective mass for the motion perpendicular to the direction of the applied magnetic field and the energy of the size quantization [1]. If we consider a 2DES in a perpendicular magnetic field with no parallel magnetic field, the electron motion along the $z$-axis has no influence from the magnetic field. Therefore, the size quantized energy level has no change.

The motion of spinless, non-interacting, massive electrons in $xy$-plane can be described by
\[-\frac{\hbar^2}{2m^*}\left(-i\frac{\partial}{\partial x} - \frac{eB}{\hbar}\right)^2 - \frac{\partial^2}{\partial y^2}\right] \psi(x, y) = E_\perp \psi(x, y) \quad (1.6)\]

where $B$ is the magnetic field along the z-direction and the vector potentials $(A)$ in x and y directions are $A_x = -yB$ and $A_y = 0$, respectively.

If the wave function is $\psi(x, y) = e^{ipx/\hbar}\chi(y)$ and substituting in Equation (1.6) will result the harmonic oscillator equation as described in Equation (1.7),

\[-\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial x^2} + \frac{m^*\omega_c^2(y - y_0)^2}{2} \chi = E_\perp \chi \quad (1.7)\]

where $\omega_c$ is the cyclotron frequency and $y_0$ is the oscillator center point given by,

\[y_0 = -\left(\frac{1}{eB}\right)p_x \quad (1.8)\]

The magnetic length is $l_0 = \sqrt{\hbar/eB}$. Substituting $l_0$ in Equation (1.8), for the ground state of the Landau oscillator, $y_0$ can be derived as

\[y_0 = -l_0^2 \left(\frac{p_x}{\hbar^2}\right) \quad (1.9)\]

The total energy can be written as

\[E = E_M + \hbar\omega_c(N + \frac{1}{2}) \quad (1.10)\]

where $N = 0, 1, 2, \ldots$ for $\hbar\omega_c(N + 1/2)$ are the energy eigenvalues $E_\perp$ of Equation (1.6), and these are known as Landau levels (LL).

If the electron spin is considered, each LL is split into two energy levels separated by Zeeman energy,

\[E = E_M + \hbar\omega_c(N + \frac{1}{2}) \pm \frac{1}{2} \mu_B gB \quad (1.11)\]
where $\mu_B$ is the Bohr magneton, and the $g$ is the electron $g$-factor. Here in Equation (1.12) the negative and positive signs correspond to the spin up and spin down states of the electrons. The gap between any consecutive Landau levels is equal to cyclotron energy, $\hbar \omega_c$ which can be further split as Zeeman energy $\mu_B g B$ due to the spin nature of the electrons.

Generally, the Fermi energy at low temperatures is given by

$$E_F = \frac{\hbar^2 k_F^2}{2m^*}$$  \hspace{1cm} (1.12)

It is lower than the energy separation in size quantized energy levels. Therefore, the lowest size quantized energy level is occupied, and Equation (1.12) can be written as,

$$E = E_1 + \hbar \omega_c (N + 1/2) \pm \frac{1}{2} \mu_B g B$$  \hspace{1cm} (1.13)

where $E_1$ is the lowest sub band energy in size quantized energy levels.

The momentum component $p_x$ and the discrete LL index $N$ describe the motion in the 2D plane ($xy$-plane) under the non-zero magnetic field along the $z$-direction. The Landau energy levels are degenerate over the momentum $p_x$ or the oscillator position $y_0$ since the energy depends only on the discrete LL index and the spin direction. It can be shown that the degeneracy of LL is $L_x L_y \times \frac{1}{2 \pi l_0^2}$, where $L_x$ and $L_y$ are the dimensions of the sample along the x and y directions, respectively. So, the LL degeneracy per unit area is equal to $(2 \pi l_0^2)^{-1}$. Note that $L_x L_y$ is the area of the sample. Hence, the density of states for a given LL is equal to $n_{DS} = eB/\hbar$. For $l_0 = \sqrt{\hbar/|eB|}$, the number of LL filled with carriers is defined as the filling factor $\nu$,

$$\nu = \frac{n}{n_{DS}}$$  \hspace{1cm} (1.14)

where, $n$ is the charge carrier density. Since $n_{DS} \propto B$, $\nu \propto B^{-1}$. 
1.5 Classical transport: Drude conductivity

The classical model for the transport properties of electrons in metals was proposed by Paul Drude in the early 1900s [2]. The Drude model for electrical conduction was developed from the kinetic theory of gases with some assumptions as described below. It was assumed that there are no electron-electron interactions between collisions which is known as the independent electron approximation, and there are no electron-ion interactions known as the free electron approximation. Therefore, electrons have linear motion as far as long as no external magnetic or electric field is present. It also assumed that the collisions between ion and electrons are instantaneous, uncorrelated events that will only influence the sudden change in the electron velocity. The collision probability of an electron in a time interval $dt$ is $dt/\tau$, where $\tau$ is mean free time between collisions and it is independent of the position and momentum of the electron. The distance traveled by the electrons without collisions is known as the mean free path. The electrons reach thermal equilibrium with their surroundings as a result of collisions.

The resistivity $\rho$, electric field $E$, and the current density $j$, are related by $E = \rho j$. The current density can be written as $j = -nev_{avg}$, where, $n$ is the electron density and $v_{avg}$ is the average velocity of electrons. The $v_{avg}$ is given by $v_{avg} = -eE\tau/m$. Here, the negative sign indicates the electron movement opposite to the applied electric field. If the conductivity is $\sigma_0$, then $j = \sigma_0 E$. The $\sigma_0$ can be written as $\sigma_0 = ne^2\tau/m$.

1.5.1 The motion of an electron in an electric field and a magnetic field

The classical view of the electron motion in an electric field and a magnetic field can be described by
\[ m\ddot{v} + \frac{m\vec{v}}{\tau} = -e(\vec{E} + \vec{v} \times \vec{B}) \]  

(1.15)

The term \( \frac{m\vec{v}}{\tau} \) is related to the electron scattering due to the disorders. For the sake of discussion, let’s consider \( \vec{E} = E\hat{x} \) and \( \vec{B} = B_2 \). At the steady state of the electron,

\[ \frac{m\vec{v}}{\tau} = -e(\vec{E} + \vec{v} \times \vec{B}) \]  

(1.16)

The matrix in 3D from Equation (1.17) can be written as

\[
\begin{pmatrix}
\frac{m}{\tau} & eB & 0 \\
-eB & \frac{m}{\tau} & 0 \\
0 & 0 & \frac{m}{\tau}
\end{pmatrix}
\begin{pmatrix}
\nu_x \\
\nu_y \\
\nu_z
\end{pmatrix}
= -e
\begin{pmatrix}
E \\
0 \\
0
\end{pmatrix}
\]  

(1.17)

The conductivity \( \sigma \) is given by

\[ \sigma = ne^2A^{-1} \]  

(1.18)

where

\[ A = \begin{pmatrix}
\frac{m}{\tau} & eB & 0 \\
-eB & \frac{m}{\tau} & 0 \\
0 & 0 & \frac{m}{\tau}
\end{pmatrix} \]  

(1.19)

Finally, it can be derived that

\[ \sigma = \frac{\sigma_0}{1 + \omega_c^2\tau^2}
\begin{pmatrix}
1 & -\omega_c\tau & 0 \\
\omega_c\tau & 1 & 0 \\
0 & 0 & (1 + \omega_c^2\tau^2)
\end{pmatrix}
\]  

(1.20)

where, \( \omega_c \) is the cyclotron frequency.

Since there is no force due to \( E\hat{x} \) and \( B_2 \) the conductivity term in the z-direction is written as \( \sigma_{zz} = \sigma_0 \). The conductivity term along the y-direction is changed due to the \( B_2 \) component and it is given by,

\[ \sigma_{xy} = -\sigma_{yx} = \frac{\omega_c\tau \sigma_0}{(1 + \omega_c^2\tau^2)} \]  

(1.21)
The presence of a voltage across the sample is known as Hall voltage which is perpendicular to both $B_z$ and to the current flow through the sample. It was discovered by E.H. Hall in 1879. The negative sign in $\sigma_{xy} = -\sigma_{yx}$ reflects that the sign of the Hall voltage which can be changed by changing the direction of the magnetic field or electric field.

1.6 Quantum transport

The wave nature of the electrons plays a crucial role in separating quantum transport from classical transport. In quantum transport, the phase factor is an additional parameter to describe the electron behavior. From the quantum perspective, the scattering process affects the phase factor in the electron wave function. The scattering process can be divided as large angle or backward and small angle or forward scatterings [3]. The large angle scattering events are responsible for electrical resistance, and they are related to the electron mobility ($\mu$) and the transport lifetime ($\tau_t$). The transport lifetime is derived from the solution of the Boltzmann equation in the relaxation time approximation [4]. The dc conductivity ($\sigma_{DC}$) is related to the $\tau_t$ through

$$\sigma_{DC} = N_s e \mu = N_s e^2 \tau_t / m^*. \quad (1.22)$$

Here, $N_s$ is the 2D sheet carrier density.

The small angle or forward scattering is related to the single particle lifetime ($\tau_s$). This $\tau_s$ is inversely proportional to the half-width ($\Gamma$) of the Landau level broadening through

$$\Gamma = \hbar / \tau_s. \quad (1.23)$$

The transport and single-particle lifetimes can be determined from

$$\tau_t^{-1} = \int dk' P(k, k')(1 - \cos \theta) \quad (1.24)$$
\[ \tau_s^{-1} = \int dk' P(k, k') \]  

(1.25)

where \( \theta \) is the scattering angle and \( P(k, k') \) is the probability for scattering from state \( k \) to \( k' \).

The carriers in 2DES of GaAs/AlGaAs heterostructures long-range scattering are due to ionized impurities; in other words, small angle scatterings are dominant. Thus \( \tau_s < \tau_t \).

Systems like metal-oxide-semiconductors have isotropic scattering, which means \( \tau_s = \tau_t \) [3].

### 1.7 Landau level quantization

The energy spectrum of electrons is quantized into discrete energy levels due to the magnetic field, as described in Figure 1.3. These discrete energy levels are known as Landau levels, and they are in equidistance in energy \( E_n \) and equally degenerate to \( n_{LL} \). Thus,

\[ E_n = \hbar \omega_c (1 + 1/2) \]  

(1.26)

\[ n_{LL} = 2eB/\hbar \]  

(1.27)

An additional two appear in Equation (1.28) due to spin degeneracy at the Landau level. It’s clear that while the Landau level energy depends on the material properties, Landau level degeneracy depends on the magnetic field and the \( e/\hbar \) not on the material properties. Further increases of the magnetic field can resolve the Landau levels into two sub-levels according to the spin orientation of carriers. The energy gap between consecutive sub-levels is labeled as \( \Delta_s \) and,

\[ \Delta_s = g \mu_B B \]  

(1.28)
Figure 1.3 Landau level spectrum of a spin degenerate ideal 2DES at various magnetic fields. When there is no magnetic field, the density of states is a constant value in the energy frame. Landau levels are formed in a finite magnetic field. The energy gap between two consecutive Landau levels is equal to $\hbar \omega_c$, where $\hbar$ is the reduced Planck constant and $\omega_c$ is the cyclotron frequency. Further increases of the magnetic field can resolve the Landau levels into two according to the spin orientation of carriers. The energy gap between consecutive sub-levels is labeled as $\Delta_s$. 
1.8 Shubnikov-de Haas oscillations

Figure 1.4 Spin split Shubnikov de Haas oscillation in a GaAs/AlGaAs Heterojunction at 50 mK. Y-axis shows the longitudinal resistance as a function of the magnetic field.

At low enough temperature with sufficient magnetic field, the LL energy spectrum of 2DES shows energy gaps, and the density of states becomes a discrete function of $1/B$. However, in lower magnetic field states, the Landau levels are not separated completely from each other, and the density of states becomes a continuous periodic function of $1/B$. In other words, when the magnetic field changes, the partially separated Landau levels pass through the Fermi level. This is observed as oscillations in longitudinal resistance, and these oscillations are known as Shubnikov-de Haas Oscillations. Also, these oscillations are periodic with the inverse magnetic field. The oscillatory part of the magnetoresistance can be expressed as
\[ \Delta R_{xx} = R_0 \frac{\chi_T}{\sinh(\chi_T)} e^{(-\pi/\omega_c \tau)} \cos \left( 2\pi \frac{E_F - E_M}{\hbar \omega_c} - \phi \right) \] (1.29)

where \( R_0 \) is the zero-field resistance and \( E_M \) is the energy of the \( M \)th sub-band. The temperature dependence of the SdHOs is described by the factor of \( \chi_T/\sinh(\chi_T) \), where, \( \chi_T = \frac{2\pi^2 k_B T}{\hbar \omega_c} \).

From an experimental perspective, \( \Delta R_{xx} \) can be considered an exponential damped cosine function [4, 5]:

\[ \Delta R_{xx} = A e^{-\alpha/\beta} \cos(2\pi F/\beta) \] (1.30)

where, \( A \) is the amplitude of the oscillation, \( \alpha \) is the damping factor, \( F \) is the frequency of the SdHOs that is related to the carrier density \( n \) of the system.

![Figure 1.5 The periodicity of SdHOs in 2DES at GaAs/AlGaAs as a function of the inverse magnetic field. F is the frequency of SdHOs. Here the y-axis is longitudinal resistance, and the x-axis is the inverse magnetic field.](image)

By examining the properties of SdHOs at low temperatures, we can extract the information of the 2DES, such as effective mass, scattering time, carrier density. Interestingly,
the SdHO amplitude depends on the temperature, so it can be used to probe the carrier heating of the 2DES under particular circumstances.

1.9 Quantum Hall effects

1.9.1 Integer Quantum Hall Effect

In 1985, Klaus von Klitzing received the Nobel Prize in Physics for discovering the quantum Hall effect, which was later popularized as the integer quantum Hall effect (IQHE) [6]. The classical Hall resistance results from charge accumulation produced by the magnetic field in the electronic system due to the Lorentz force. However, at the high end of the magnetic spectrum at low temperatures, Hall resistance shows plateaus which were not predicted in the Drude model. Interestingly, the resistance at the plateau only depends on the Planck’s constant and the elementary charge of the carrier but not on the Hall bar geometry.
In 1975, Tsuneya Ando, Yukio Matsumoto and Yasutada Uemura predicted that the integer quantization in Hall conductivity for the first time [8]. Then, Jun-ichi Wakabayashi and Shinji Kawaji observed plateaus in the Hall effect experimentally in 1978 [9]. In 1980, Klaus von Klitzing discovered that the Hall resistance was exactly quantized on the Hall plateaus from high magnetic field measurements on silicon-based MOSFET. That experiment has been carried out by measuring both transverse and longitudinal voltages as a function of the gate voltage while the magnetic field was constant at 18 Tesla, as shown in Figure 1.6 [7]. The Landau level in the system should be well separated at 18 T. The concentration of charge carriers could
change by sweeping the gate voltage, and the Fermi energy is a function of carrier concentration in that study. Therefore, the Fermi energy can be modified by changing the gate voltage. A similar situation can be approached by changing the magnetic field as well.

If there is no external magnetic field in the system, the density of the system is a constant as a function of energy. If there is a nonzero magnetic field along the z-direction, the Landau levels are considered to be delta functions with zero width in an ideal case. However, in a practical situation, these delta functions will be broadened due to the scattering of carriers. Thus, a small magnetic field will not be sufficient to completely separate the Landau levels from each other, which means that the Landau level separation is narrower than the width of a Landau level. As the magnetic field increases, the Landau level separation will increase, and overlap between Landau levels will decrease. Landau levels move relative to the Fermi level when the magnetic field varies.

In classical, 2DES with a strong magnetic field, $\omega_c \tau \gg 1$,

$$\sigma_{xx} = \frac{e^2 n}{m^* \omega_c^2 \tau}$$  \hspace{1cm} (1.31)

and,

$$\sigma_{xy} = \frac{e^2 n}{m^* \omega_c^2}$$  \hspace{1cm} (1.32)

where, $\sigma_{xx}$ and $\sigma_{xy}$ are longitudinal and Hall conductivities, respectively. When the Fermi energy lies in between the N and N+1 Landau levels, which are the spin up and spin down states of the electron in N$^{th}$ Landau level, then the Hall conductivity becomes

$$\sigma_{xy} = \left(\frac{e^2}{\hbar}\right) \nu_N$$  \hspace{1cm} (1.33)
Here $\nu_N$ is the filling factor, where $\nu_N = n / n_{DS}$ and $n_{DS} = eB / h$. When longitudinal resistance reaches zero, the Fermi level lies in the gap of two consecutive Landau levels. Interestingly, Hall conductivity depends only on the integer multiples of $e^2 / h$. The reciprocal of this fundamental ratio is known as the von Klitzing constant, and it is approximately 25.812 kΩ.

### 1.9.2 Fractional Quantum Hall Effect

![Fractional Quantum Hall Effect](image)

**Figure 1.7** The first experimental observation of fractional quantum Hall effects. Longitudinal and Hall resistivity as a function of the magnetic field in GaAs/AlGaAs sample with carrier concentration $1.23 \times 10^{11}$ cm$^{-2}$. The Landau level filling factor was defined as $\nu = nh / eB$ and it is $1/3$. Reproduced from Tsui et al. (1982)[10].

As the external perpendicular magnetic field increases, the Landau levels move to higher energies with respect to the Fermi energy level. The quality specimen can achieve $\nu < 1$ at a low
enough temperature with a high enough magnetic field. As the magnetic field goes higher, the magnetic length or the characteristic length of the carriers becomes small. Magnetic length is a parameter of the electron-electron scatterings in the specimen.

The Fractional Quantum Hall effect (FQHE) was discovered by Tsui, Stomer, and Gossard [10]. A simple understanding of the phenomena, seen in Figure 1.8, appears in the report of Mani et al., where they assert that FQHE’s constitute a fractal [11]. As described in Ref. [11], the main sequence of FQHE where the filling factor \( \nu < 1 \) can be reconstructed using IQHE where filling factor \( \nu > 1 \). This study predicts the possible fractional quantum Hall states, some of which have not been observed in experimentally to date.

![Figure 1.8 Longitudinal resistance \( R_{xx} \) and Hall resistance \( R_{xy} \) in GaAs/AlGaAs heterojunction as a function of magnetic field at 25 mK. Left and right ordinates have been scaled to the multiplier of von Klitzing \( R_K \) constant. Reproduced from Mani et al. (1996) [11].](image-url)
2 ELECTRICAL TRANSPORT IN 2DES UNDER MICROWAVE IRRADIATION

The transverse Hall component of electrical resistance in 2DES is quantized in the presence of a magnetic field at low temperatures, and it is known as the quantum Hall effect. The discoveries related to quantum Hall effects were the subject of two Nobel Prizes in history [7, 10]. The discovery of the zero-resistance state (ZRS) in GaAs/AlGaAs 2DES under the microwaves stimulated this field again. A large number of research activities have been reported on the properties of 2DES irradiated by microwaves.

This chapter summarizes the important theoretical and experimental findings on microwave induced magnetoresistance oscillations (MIMOs) that form the background information required to understand this phenomenon.

2.1 Introduction

In 2002, Mani and coworkers reported the first experimental evidence for radiation induced ZRS in 2DES at the GaAs/AlGaAs interface [9]. This experimental observation inspired physicists to explore the unknown nature of the photo-excited 2DES. In general, if 2DES heats up, then its electrical resistance should increase, which contradicts the experimental observation, and it became a very controversial topic in low temperature physics. Later on, some theoretical explanations were developed to explain the experimental observations of ZRS [12-14].

In addition to using microwaves in conventional experiments, they can be utilized to study the temperature dependency of quantum systems, a novel experimental technique recently reported. In 2021, Mani and coworkers reported that the microwave-induced heating technique
was applied to control the temperature of the quantum systems precisely and rapidly to examine physical phenomena in the FQHE regime [15].

2.2 Microwave induced zero resistance states

Figure 2.1 The first experimental observation of radiation induced zero resistance states on high-mobility GaAs/AlGaAs heterojunction under excitation at 103.5 GHz. The insert describes the appearance of zero resistance states in the lower magnetic field side under photoexcitation while quantum Hall phenomena occur at the high magnetic field side. Reproduced from Mani et al. (2002) [16].

Figure 2.1 shows the longitudinal resistance of high mobility 2DES as a function of magnetic field up to 10 Tesla under the microwave photo-excitation at 103.5 GHz. The well-known quantum Hall phenomena are visible in the high magnetic field side with expected plateaus in the Hall resistance. The microwave induced magnetoresistance oscillations appear below the quantum Hall effects in the low magnetic field regime. Unexpectedly, the microwave induced magnetoresistance oscillations minima approach zero resistance as shown in the insert of Figure 2.1. Note that the corresponding Hall resistances to these zero resistance states don’t show
the plateaus as shown in Figure 2.2. That contrasts with the typical quantum Hall effect since quantum Hall effects show plateaus in Hall voltage where the longitudinal resistance approaches zero values.

![Figure 2.2 Rxx and Rxy as a function of the magnetic field with and without microwaves on the 2DES. The Hall resistance doesn’t show the plateau corresponding to the zero resistance states. Reproduced from Mani et al. (2002)[16].](image)

This experimental study discovered that these characteristic magnetic fields of microwave induced zero resistance state (ZRS) could be described as Equation (2.1).

\[ B = \left[ \frac{4}{4j + 1} \right] B_f \]  \hspace{1cm} (2.1)

where \( j = 1,2,3, \ldots \), and

\[ B_f = \frac{2\pi f m^*}{e}. \]  \hspace{1cm} (2.2)
2.3 Microwave induced magnetoresistance oscillations (MIMOs)

The MIMOs are primarily observable in the region of approximately $2\pi f > \omega_c$, under moderate microwave intensities. As described earlier, the magnetic fields associated with MIMOs appear to scale with the incident microwave frequency $f$, effective mass $m^*$ and electron charge $e$. The MIMOs generated from lower microwave frequencies can be observed typically below and not overlapping with the Shubnikov-de Haas Oscillations (SdHOs) as described in Figure 2.3 (a). However, high microwave frequencies generate MIMOs in the high magnetic field side, and they overlap with SdHOs, as shown in Figure 2.3 (b) [16-19].

![Figure 2.3](image_url)
microwave frequency range. (b) depicts the MIMOs under high microwave frequencies. Reproduced from Mani et al. (2002) [16].

Figure 2.4 (a) Microwave power (b) DC current and (c) temperature dependence of MIMOs in high mobility 2DES in GaAs/AlGaAs. Reproduced from Mani et al. (2002) [16].

As shown in Figure 2.4(a), MIMOs grow with an increase of the power of the incident microwaves, and the oscillations minima approach zero eventually. Later studies show that the MIMOs amplitude has a nonlinear dependence with the microwave power, i.e., MIMOs amplitude $\propto \sqrt{P}$, where $P$ is the microwave power [20, 21]. In contrast, the linear behavior of MIMOs amplitude with the incident microwave power had also been reported [12, 22].
Ref.[16] experimentally showed that MIMOs do not become sensitive to the DC current through the device, as illustrated in Figure 2.4(b). The MIMOs amplitudes decay with increases of the temperature of the specimen as shown in Figure 2.4(c) [16].

The MIMOs with ZRS generated from high frequencies can overlap with the SdHOs [16-19] and quantum Hall effect regime [23]. The Hall resistance plateaus appear in quantum Hall effects when the longitudinal resistance values reach zero. Interestingly, Hall plateaus disappear when microwave induced ZRS under high microwave frequencies overlap with the quantum Hall region. This may be the result of microwave irradiation replacing the integer quantum Hall effect with the traditional classical Hall effect. However, more studies are needed to understand this phenomenon.

2.4 The theoretical approach to Microwave induced magnetoresistance oscillations

The MIMOs have become a central focus of numerous theoretical and experimental studies after the unexpected observation of ZRS in 2DES. It is by now well known that the MIMOs are 1/4-cycle phase-shifted with 1/B periodicity, and oscillatory minima follow Equation (2.1). To date, many theoretical approaches [13, 14, 24-35] have been suggested to understand the physics of MIMOs and microwave induced ZRS. Here we consider only a few theoretical studies that are most cited in the past.
2.4.1 Displacement model

![Figure 2.5 Schematic diagram of the displacement mechanism for MIMOs. (a) the shift of the guiding center of a cyclotron orbit due to quasi-elastic scattering off disorder. (b) cartoon picture of the correlation between the direction of the cyclotron-orbit shifts $\Delta X$ in the process of photon absorption and the sign of the detuning second harmonic of the cyclotron resonance. The yellow strips show the tilted Landau levels due to DC bias. Reproduced from Dmitriev et al. (2002) [36].](image)

Hall voltage develops as a result of deflection of a current through the specimen due to a perpendicular magnetic field. The DC bias across the sample will result in a finite tilt in the Landau levels, as shown in Figure 2.5(b). At small magnetic field values, Landau levels are highly overlapping. Thus the oscillatory nature can be considered in the density of states $\nu(\epsilon)$. When the ratio of incident microwave energy to cyclotron energy is not an integer, the photoexcited electron must have a spatial displacement ($\Delta X$) to satisfy the energy conservation.

As depicted in Figure 2.5(b), if the excited photon energy is slightly lower than a Landau level, the $\Delta X$ occurs towards the lower energy, increasing the longitudinal resistance. Conversely, if the excited electron energy is slightly above the Landau level, the $\Delta X$ occurs towards the high energy side, which makes longitudinal resistance drops. The origin of this spatial displacement is the short-range scattering due to impurities in the system.
2.4.2 Inelastic model

Figure 2.6 (a) Absorption and emission of microwave photons in the framework of an oscillating density of states. (b) The microwave moderated density of states $v(\epsilon)$ and the resulting oscillatory distribution function $f_\epsilon$ for $k_B T = \hbar \omega_c$, compared with thermal distribution function $f_\epsilon^T$. Reproduced from Dmitriev et al. (2002) [36].

Inelastic theory for the MIMOs is based on the change in the electron distribution function induced by microwaves [12, 37]. This model assumes that the microwave excitation rate and relaxation rate are of the same order. Therefore, there is no significant redistribution of electrons across the density of states at the steady state. The density of states shows oscillatory behavior with the inverse magnetic field. Thus the correction to the electron distribution function has an oscillatory nature too. This phenomenon is reflected in DC conductivity as oscillations in the frame of $\omega / \omega_c$. Further, this is the result of inelastic scattering of the 2DES, mainly electron-electron scattering and, secondly, impurity scattering. Therefore, Inelastic theory predicts that MIMOs are strongly temperature dependent as the effect increases as $T^{-1}$ for $k_B T \ll \hbar \omega$ and $T^{-2}$ for $k_B T \gg \hbar \omega$ [12, 37].
2.4.3 Radiation driven electron orbit model

Figure 2.7 Schematic of electron transport in 2DES with and without microwaves as suggested in radiation driven electron orbit model. (a) No microwave irradiation, electrons jump between fixed position orbits due to scattering. Under microwaves, these orbits are not fixed but oscillate. (b) shows how the orbit moves back while electrons jump forward. (c) the orbit moves forward. (d) the orbit moves forward, but the electron jump is less than the orbital’s amplitude. Since the final state is occupied, the electron movement between orbits is impossible, which explains the microwave induced ZRS. Reproduced from J. Inarrea et al. (2005) [33].

The radiation driven orbital model is developed on an exact solution for the harmonic oscillator wave function interacting with microwaves, and a perturbation treatment for elastic scattering from randomly distributed charge impurities [33, 38, 39]. This theory assumes that when no radiation interacts with the 2DES, the electron orbits are fixed, and electrons jump between the orbits due to randomly distributed charge impurity scatterings. The 2DES in a perpendicular magnetic field and a DC electric field that makes the electric transport along the specimen are considered in this model. Also, electrons interact with the AC electric field \( E_{MW} \) which is from the linearly polarized microwaves. The linearly polarized microwaves can be characterized using polarization angle \( \alpha \),
\[
\tan \alpha = \frac{E_y}{E_x}
\]  
(2.3)

where \(E_y\) and \(E_x\) are microwave amplitudes along \(x\) and \(y\) directions. In such a scenario, the average distance advanced by the electron \(\Delta X^{MW}\) at each scattering event can be written as,

\[
\Delta X^{MW} = \Delta X^0 + A \cos(\omega \tau)
\]  
(2.4)

where \(\Delta X^0\) is the average distance advanced by an electron with no incident microwaves and \(A \cos(\omega t)\) is the distance advanced upon irradiation by microwaves. In addition, \(\omega\) and \(\tau\) are the microwave angular frequency and impurity scattering time, respectively. The amplitude \(A\) of the average distance upon microwaves irradiation is given by

\[
A = \frac{eE_0}{m^* \sqrt{\frac{\omega^2(\omega_c^2 - \omega^2)^2}{\omega^2 \cos^2 \alpha + \omega^2 \sin^2 \alpha} + \gamma^4}}
\]  
(2.5)

where \(E_0\) is the microwave field intensity and \(\gamma\) is a sample dependent parameter. If \(\gamma > \alpha\), the amplitude \(A\) becomes independent of polarization angle \(\alpha\). On the other hand, if \(\gamma < \alpha\), then \(A\) depends on the polarization angle \(\alpha\) [33].

### 2.4.4 Non-parabolicity model

This model is based on the classical view of the magnetotransport due to the perpendicular magnetic field to the 2D plane of the 2DES under strong microwave irradiation [27]. According to this model, the magnetic spectrum of microwave irradiated 2DES shows a weak non-parabolicity near to the cyclotron resonance, leading to a slight change in the effective mass of the carriers. As a result, diagonal resistance shows a change but not the corresponding Hall resistance. Non-parabolicity theory also predicts that MIMOs only happen under linear polarized
microwaves but not circularly polarized microwaves, which contradicts experimental findings [40].
3 MICROWAVE INDUCED ELECTRON HEATING IN THE GaAs/AlGaAs 2DES IN THE REGION OF SdHOs AND NULL MAGNETIC FIELD REGIONS

This chapter will discuss my experimental work to examine the qualitative behavior of microwave induced electron heating in GaAs/AlGaAs heterostructures at cryogenic temperatures [41-45]. The effect of microwave irradiation on both the null field longitudinal magnetoresistance and the amplitude of Shubnikov-de Haas Oscillations (SdHOs) at liquid helium temperatures in GaAs/AlGaAs Hall bar devices has been investigated. The various microwave frequencies with peak source power $0 \leq P_{\text{peak}} < 4$ mW are used to illuminate the high mobility 2DES. The magnetotransport measurements were taken as a function of microwave power and lattice temperature. The results of this experimental study were compared with the theoretical predictions [46-48], and I comment upon the relative role of electron heating in the microwave irradiated high mobility 2DES. This work has been published in the journal Physical Review B [41].

This experimental work indicates that microwave irradiation on 2DES in GaAs/AlGaAs produces a small discernible increase in electron temperatures at zero magnetic fields and finite magnetic fields. The heating effect appears minor at the examined finite magnetic field region comparison to zero magnetic field, which agrees qualitatively with theoretical predictions [48].

3.1 Introduction

A topic of experimental interest is the comprehensive study of electron heating, as the theory has expected the possibility of variable microwave-induced electron heating in the 2DES in the high filling-factors that occur at low-magnetic-field limit [48, 49]. Generally, electrons in the photo-excited 2DES are warmer than the lattice due to energy absorption from the radiation under microwave irradiation. The theory has investigated this electron heating induced by
microwave photo-excitation using a balance-equation concept that considers the photon-assisted electron excitation along with radiation-induced modification of the electron distribution of high-mobility 2DES. The electron temperature is determined by balancing the energy absorption from the microwaves and the energy dissipation to the lattice over electron-phonon interactions. The results extracted from the balance equation approach show that electron temperature depends on the magnetic field, the microwave power, and frequency. Further, the theory suggested that for the $\omega c / \omega \geq 1$ region, both electron temperature ($T_e$) and radiation absorption rate ($S_p$) show relatively monotonic behavior, while in region $\omega c / \omega \leq 1$, both $T_e$ and $S_p$ exhibit oscillatory nature as shown in Figure 3.1 [48].

Figure 3.1 The longitudinal resistivity $R_{xx}$, electron temperature $T_e$, and energy absorption rate $S_p$ of a GaAs-based 2DES under 100-GHz linearly $x$-polarized microwaves, as a function of the ratio of cyclotron resonance frequency to microwave angular frequency. The lattice temperature is $T=1$ K. Reproduced from X.L. Lei et al. (2005)[48].
Previous experimental studies showed that the amplitude of the SdHOs is sensitive to the electron temperature. The decay and vanishing of the SdHOs under the microwaves in the region of the radiation-induced ZRS in GaAs/AlGaAs devices were examined previously, and it was discovered that the SdHOs amplitude vanished in proportion to the background resistance at the centers of the radiation-induced ZRS [19]. The influence of the incident microwaves on the amplitude of the SdHOs over the region of $2\omega \leq \omega_c \leq 3.5\omega$ was discussed in Ref. [5]. Experimentally it shows that the higher microwave frequencies ($> 30$ GHz) produce non-monotonic damping of the SdHOs, and it is understood as the formation of a non-equilibrium electron distribution function by microwave induced intra-Landau level transitions [18]. Microwave high frequencies ($150 \leq f \leq 400$ GHz) induced distinct nodes in the SdHOs that were reported in Ref. [17].

### 3.2 Experiment and Results: Influence of microwave radiation on SdHOs and resistance at zero magnetic field

This section describes the experimental procedure and results of the work that has been done as a part of this dissertation to examine the microwave induced electron heating on 2DES in GaAs/AlGaAs.

#### 3.2.1 Experimental Section

Lock-in-based electrical measurements were performed on photolithographically fabricated Hall bar devices from molecular beam epitaxy grown high mobility GaAs / AlGaAs heterojunctions. The Hall bar device was located at the bottom end of a long cylindrical waveguide sample holder that is placed into a variable temperature insert (VTI) within the bore of the superconducting magnet. Low temperatures in device space were realized by pumping on
and decreasing the vapor pressure of the liquid helium within the VTI insert. The device was submerged in liquid helium for all the reported measurements in this section. The Hall bar device inside the VTI was pre-illuminated with red light to approach a high mobility state. The results reported here were generated from the device with $2.4 \times 10^{11}$ cm$^{-2}$ electron density ($n_e$) and $8.6 \times 10^6$ cm$^2$/Vs mobility ($\mu_e$). The microwaves were generated with a commercially available microwave synthesizer. The Hall bar was irradiated with linearly polarized microwaves for the photo-excited transport measurements. The direction of the microwave polarization was aligned with the long direction of the Hall bar. The longitudinal resistance ($R_{xx}$) was examined at $T = 1.47, 1.53, 1.6, 1.75,$ and $1.92$ K for several microwave source powers within $0 < P_{peak} < 4$ mW.

### 3.2.2 Results and Discussion

![Figure 3.2](image.png)

*Figure 3.2 The longitudinal resistance $R_{xx}$ as a function of magnetic field $B$, at 1.7 K for microwave frequency 48.5 GHz with various powers. Here, $R_{xx}$ also exhibits radiation-induced magnetoresistance oscillations and SdHOs. The observed SdHOs have been numerically fit over*
the range of $B$ as shown by the dashed line, that corresponds to $2.3 \leq \frac{\omega_c}{\omega} \leq 5.2$, where $\omega_c$ is the cyclotron frequency and $\omega = 2\pi f$, with $f = 48.5$ GHz. The inset shows the $R_{xx}$ over the range $-0.022 \leq B \leq 0.022$ T, and it indicates that the $R_{xx}$ is upshifted with increasing microwave power.

As shown in Figure 3.2, the MIMOs were observable for $B \leq 0.2$T at 1.7 K. The linear increment of incident microwave power resulted in the nonlinear growth of MIMOs amplitude as expected [5, 21]. The SdHOs were observable under both microwave irradiated and dark situations above 0.2 Tesla roughly. Here, we focused on two noticeable features in the $R_{xx}$ vs. $B$ traces with the parametric variation of microwave source power. These features are (1) the $R_{xx}$ at zero magnetic field is shifted to larger resistance values with increases of the incident microwave power as shown in the insert of Figure 3.2, and 2) the amplitude of SdHOs gets weaker with the increase of incident microwave power at finite magnetic fields. Interestingly, exactly similar features were observed in the $R_{xx}$ vs $B$ traces when lattice temperature increases even without microwaves on the device. Thus, it is reasonable to conclude that these microwave-induced discrepancies are consequences of a heating effect from the incident microwaves on the Hall bar device. The aim here was to find the source of electron heating for these two scenarios and to determine how the electron temperature changes with microwave irradiation.
Figure 3.3 The upshifting of $R_{xx}$ at zero magnetic field to higher resistance. (a) The $R_{xx}$ at $B=0$ T is plotted as a function of microwave source power, $0 \leq P < 4 \text{ mW}$. The pink circles are data and dashed line is for eye guidance. (b) The $R_{xx}$ at $B=0$ T is plotted as a function of lattice temperature. The blue circles are data and red solid line is the linear fit for $R_{xx}$ vs $T_L$.

As the first case, we considered the upshifting of $R_{xx}$ to higher resistance with the increase of incident microwave power as shown in Figure 3.3(a). Since the increase of $R_{xx}$ at zero magnetic field appeared as a result of microwave irradiation, which could also reasonably produce electron heating, a correlation between the zero magnetic field $R_{xx}$ and the lattice temperature has been established in this study. Therefore, the $R_{xx}$ resistance at zero magnetic field was measured as a function of the lattice temperature ($T_L$), under dark conditions, i.e., without microwave radiation on the Hall bar as depicted in Figure 3.3(b). It was noticed that the temperature interval of interest for comparing with the upshift in $R_{xx}$ at $B = 0$ T observed under microwave excitation turned out to be only $1.47 \leq T \leq 1.92$ K, over a wide temperature interval in this study. Figure 3.3(b) exhibits that the $R_{xx}$ at $B = 0$ T varies approximately linearly with lattice temperature, as shown by the red solid line in Figure 3.3(b), which is a least-squares
The linear relation between the longitudinal resistance and the lattice temperature is given via the parametric equation (see Equation (3.1)).

\[ R_{xx}[\Omega] = 1.33[\Omega/K]T_L[K] + 3.87[\Omega] \]  

(3.1)

Equation (3.1) can be inverted to obtain temperature values corresponding to the \( R_{xx} \) at \( B = 0 \) T, such as \( T = T(R_{xx}) \) as described in eq. (3.2).

\[ T[K] = \frac{(R_{xx}[\Omega/K] - 3.87[\Omega])}{1.33[\Omega/K]} \]  

(3.2)

Such inversion can serve as a temperature gauge to extract the carrier temperature in the 2DES, even in the presence of microwave radiation. The 2DES in the device can potentially be disengaged from the lattice/bath under the microwave photoexcitation. Therefore, the longitudinal resistance could serve as a gauge of the electron temperature, \( T_e \), not the lattice temperature, \( T_L \) of the Hall bar device.

Figure 3.4 The correlation of \( R_{xx} \) at \( B=0 \) T and the electron temperature of 2DES at lattice temperature 1.7 K.
Then, $R_{xx}$ vs $B$ measurements were collected at various microwave source powers ($P$) at a lattice temperature of 1.7 K. The left ordinate of Figure 3.4 summarizes the results at zero magnetic field by plotting the $R_{xx}(B = 0 \text{T})$ vs. $P$ (the abscissa). The corresponding electron temperatures determined using Equation (3.2) are shown in the right ordinate of Figure 3.4. The results here at zero magnetic field suggest that $\Delta T_e / \Delta P \approx 0.2 \text{ K/mW}$.

Figure 3.5 $R_{xx}$ as a function of inverse magnetic field. The black curve is the $R_{xx}$ without microwaves and red curve with microwaves. Now the SdHOs appears toward the origin of the plot as shown in orange dashed lines. The insert shows a close view of the SdHOs in the range of interest.

For the second part of this study, the interaction of microwave photo-excitation on the SdHOs was examined over the range of $2.3 < \omega / \omega_c \leq 5.2$ as indicated in Figure 3.2. A monotonic background term was subtracted from the raw longitudinal magnetoresistance data (see insert of Figure 3.5) to separate the oscillatory term, $\Delta R_{xx}$. This oscillatory $\Delta R_{xx}$ was plotted
as a function of the inverse magnetic field for various microwave source powers as shown in Figure 3.6 (a) for the lattice temperature, $T_L = 1.7$ K.

Figure 3.6 The oscillatory part of background subtracted SdHOs. (a) SdHOs are observable in $\Delta R_{xx}$ which are periodic in inverse magnetic field. $\Delta R_{xx}$ vs $B^{-1}$ has been plotted over the span $1.7 \leq B^{-1} \leq 3.75$ T$^{-1}$ at various microwave powers. (b) The red circles show the $\Delta R_{xx}$ data while the black line represents numerical fits to $\Delta R_{xx} = - Ae^{\alpha/B} \cos(2\pi F/B)$. The electron temperatures at each microwave power and each lattice temperature were extracted by numerical fitting.

The amplitude of the SdHOs decays exponentially with the increase of microwave source power as shown in Figure 3.6(a). To extract the amplitude of the SdHOs, a standard nonlinear least-square fit was applied on $\Delta R_{xx}$ data with an exponentially damped sinusoidal function described in Equation (3.3)

$$\Delta R_{xx} = - Ae^{\alpha/B} \cos(2\pi F/B) \quad (3.3)$$

where $A$ is the amplitude and $F$ is the SdHOs frequency [4, 23]. The parameter $F$ was fixed to a constant value in the numerical fittings because the $F$ is unresponsive to the incident microwaves at a constant lattice temperature.

To extract the electron temperature from the SdHOs, the parameter $\alpha$ in Equation (3.3) has been expanded as in Equation (3.4).
\[ \alpha = -\lambda (T_L + T_D + \Delta T_e) \left( \frac{m^*}{m_e} \right) \]  
\hspace{1cm} (3.4) 

where

\[ \lambda = \frac{2\pi^2 k_B m_e}{e} \]  
\hspace{1cm} (3.5) 

and \( m^*/m_e \) is effective electron mass ratio, \( T_L \) is the lattice temperature, \( T_D \) is the Dingle temperature and \( \Delta T_e \) is the electron temperature increment with respect to the lattice. In this study \( T_L \gg T_D \), so the fitting function becomes

\[ \Delta R_{xx} = -A e^{-\lambda(T_L+\Delta T_e)(m^*/m_e)/B} \cos \left( \frac{2\pi F}{B} \right). \]  
\hspace{1cm} (3.6) 

We utilized the Equation (3.6) to numerically fit the data in the SdHOs range and extracted the electron temperatures under various microwave powers. As an example, Figure 3.6(b) shows the fit of the \( \Delta R_{xx} \) data at 1.7 K for the microwave power \( P_{peak} = 1 \text{ mW} \).

![Figure 3.7](image)

**Figure 3.7** The increase of electron temperature above the lattice temperature due to microwave photoexcitation, as a function of the microwave source power, both at zero magnetic field and in the SdHOs region, for an incident microwave frequency of 48.5 GHz.
We compare the elevated electron temperature, $\Delta T_e = T_L - T_e$, at zero magnetic field extracted from the zero-field $\Delta R_{xx}$ and, at finite magnetic fields extracted from the SdHOs for 48.5 GHz in Figure 3.7. The $\Delta T_e$ increased with the incident microwave power, both in the null magnetic field and small finite magnetic fields. However, the extracted $\Delta T_e$ over the SdHOs region is smaller than the $\Delta T_e$ at zero magnetic field under microwaves.

![Graph showing electron temperature as a function of microwave power.](image)

**Figure 3.8 (a)-(e):** The extracted electron temperature $T_e$ is plotted as a function of the microwave power, $P$, in the vicinity of $B = 0$ T (square symbols) and in the region of SdHOs, i.e., $2.3 < \omega_c / \omega \leq 5.2$ (disk symbols) for different lattice temperatures, $T_L$. This figure exhibits electron heating under microwave radiation in both regions. The heating at the zero magnetic field appears more noticeable than SdHOs region, in agreement with the theoretical prediction. Reproduced from T.R. Nanayakkara et al. (2018) [41].
Figure 3.8 compares electron temperature, $T_e = T_L + \Delta T_e$ as a function of microwave power at zero-magnetic field with the electron temperature at finite magnetic fields. The electron temperatures increased with the incident microwave source powers in both examined regions, zero magnetic field and finite magnetic field. Nevertheless, the increase in the electron temperature at zero magnetic field under microwave photo-excitation is roughly six times higher than the extracted electron temperature increase over the SdHOs regime. Figure 3.8 suggests that $\Delta T_e/\Delta P \approx 0.015$ K/mW at finite magnetic fields and $\Delta T_e/\Delta P \approx 0.1$ K/mW at zero magnetic field. The highly mobile electrons in the 2D system can absorb energy from the stationary microwave radiation and heat the system [48]. The absorbed energy from the microwaves is dissipated to the lattice by electron-phonon interaction through bulk transverse acoustic, longitudinal acoustic, and longitudinal optical phonons. The electronic energy absorption rate is strongly magnetic field dependent. Also, it shows oscillatory behavior with the periodicity in the inverse magnetic field, reflecting the periodicity of both MIMOs and SdHOs. In addition, theory suggests that the electron temperature, $T_e$, reflects the characteristics shown in the energy absorption rate [48]. Theoretical investigations with typical experimental parameters predict that at lower magnetic fields, i.e., $\omega_c/\omega \leq 1.4$, energy absorption happens via inter Landau level transitions, and it leads to a significantly increased $T_e$, with $T_e = 10$ K. When the magnetic field increases, the inter Landau level transitions weaken and the absorbed energy decreases significantly. As a consequence of this, the electron temperature in that region is only slightly above the lattice temperature.

3.3 Summary

We examined the microwave induced electron heating on the $R_{xx}$ in the GaAs/AlGaAs 2DES both at zero magnetic field, and at finite magnetic fields in the regime of weak SdHOs. The
present study shows that microwaves induce a small discernable increase in the electron
temperature both at zero magnetic field and at finite magnetic fields in the GaAs/AlGaAs 2DES.
The extracted electron temperature over the examined SdHOs region appears smaller in
comparison to the electron temperature at zero magnetic field.
4 MEASUREMENT OF THE SPIN GAP ENERGY AT ODD INTEGER-HIGH FILLING FACTORS USING MICROWAVE PHOTOEXCITATION

This chapter discusses examining the activation energy at high filling factors of 2DES in GaAs/AlGaAs heterostructure using a modern experimental technique of microwave induced localized heating on 2DES [15].

4.1 Introduction

The high quality 2DES in GaAs/ AlGaAs subjected to perpendicular variable magnetic field at low temperatures exhibits many physical phenomena. Among them, SdHOs in longitudinal resistance has long been famous for studying the fundamental nature of 2DES [50-53].

![Figure 4.1 Spin split SdHOs in longitudinal resistance, $R_{xx}$. (a) $R_{xx}$ is plotted as a function of magnetic field ($B$) at 120 mK. For example, the blue and red vertical arrows show the resistance peaks related to spin down and spin up states of electrons, respectively. (b) $R_{xx}$ vs filling factor ($\nu$) for the magnetic field covered in the blue shaded area of the panel (a), where $\nu \propto 1/B$. The green solid vertical lines show the integer filling factors corresponding to resistance minima, while dotted blue vertical lines represent the maxima, in panel (b). The odd $\nu$ correspond to resistance minima at spin splitting while even $\nu$ correspond to cyclotron minima.](image-url)
SdHO’s are attributed to Landau levels passing through the Fermi level of the system when the magnetic field is changed. This phenomenon is seen as oscillations in longitudinal resistance. At the low magnetic field side at high filling factors \((\nu)\), the longitudinal magnetoresistance peak corresponds to unresolved two different spin orientations at the same Landau level. This peak happens at odd integer filling factors. In an ideal case, the energy gap between consecutive Landau levels is given by, \(E = \hbar \omega_c\). Here, \(\hbar\) is the reduced Planck’s constant and \(\omega_c\) is the cyclotron frequency. As the magnetic field increases, the filling factor value decreases, and the single peaks at odd filling factors start splitting as shown in Figure 4.1(a). These split peaks correspond to two resolved spin states at the same Landau level and appear at half integer filling factors as shown in Figure 4.1(b). At low filling factors, the spin gap is larger than the disordered broadening in energy levels, which makes less overlapping between energy levels. But in high filling factors, overlapping of energy levels is dominant due to the lesser gap between the energy levels in the 2DES since the gap is proportional to the perpendicular magnetic field.

In an ideal case, the energy gap between two resolved spin states at the same Landau level is given by spin gap or Zeeman splitting which is given in Equation (4.1),

\[
\Delta_s = g_0 \mu_B B
\]  

where, \(g_0 = \text{bare g-factor}\), \(\mu_B = \text{Bohr magneton}\), and \(B = \text{total magnetic field}\). However, in reality, this g-factor can be different and enhance the spin gap \(\Delta_s (= g^* \mu_B B)\) as described in Equation (4.2) [50],

\[
\Delta_s = g^* \mu_B B = g_0 \mu_B B + E_{ex}
\]
where $g^*$ is the enhanced or effective $g$ factor and $E_{ex}$ is the exchange energy. The enhancement of the bare $g$-factor can be explained in terms of exchange interactions [54]. The exchange energy is given by [55],

$$E_{ex} = E_{ex}^0 (n_\uparrow - n_\downarrow)$$  \hspace{1cm} (4.3)

where $E_{ex}^0$ is a constant and $(n_\uparrow - n_\downarrow)$ is the net spin polarization with spin sub-band populations with $n_\uparrow$ and $n_\downarrow$ of a given a Landau level.

The cyclotron energy is determined by the perpendicular component of the magnetic field, while spin splitting is determined by the total magnetic field. The activation energies (spin gap energy values) at spin splitting in various filling factors can be extracted from the exponential temperature dependence of spin split minima at longitudinal resistance as described in Equation (4.4).

$$R_{xx,minima} = A \exp \left( \frac{-\Delta_s}{2k_BT} \right)$$  \hspace{1cm} (4.4)

In conventional temperature dependence measurements in spin split SdHOs, the temperature in 2DES was controlled by controlling the temperature in sample space which is a large area compared to the sample size. In this study, we utilized a novel technique reported by Ref. [15] to extract the activation energies at high filling factors in 2DES at GaAs/AlGaAs heterostructures. In this novel technique, a microwave induced localized heating mechanism was applied to heat the 2DES systematically.

### 4.2 Experiment

The magnetotransport measurements were performed on a 2DES formed at the interface of photolithographically fabricated GaAs/AlGaAs Hall bar heterostructures in a cryo-free dilution refrigerator. The temperature of the Hall bar device was initially set at 1.5 K, before exposure to a red LED, and the electron density of the 2DES was $n_0(1.5K) = 1.2 \times 10^{11} \text{ cm}^{-2}$
and electron mobility was $\mu(1.5K) = 6.6 \times 10^6 \text{cm}^2/\text{Vs}$. Then we illuminated the device with a red LED to realize $n_0(1.5K) = 2 \times 10^{11} \text{cm}^{-2}$ and $\mu(1.5K) = 1.4 \times 10^7 \text{cm}^2/\text{Vs}$. The contacts on the device were formed by depositing and alloying Au-Ge/Ni. The length to width ratio in the measured channel in the device was $L/W = 1$, where $L = W = 200 \mu\text{m}$. The device was mounted on a nonmagnetic chip carrier in a dilution refrigerator with wirings, as shown in Figure A.0.6. The Hall bar device could be tilted in-situ. Microwaves were generated in the synthesizer outside the dilution fridge and propagated all the way to the sample using a semi-rigid coaxial cable. That coaxial cable was terminated with a magnetic dipole wire-loop around the device. The microwave frequency and the power were controlled from the source. In this experiment, the microwave power dependent measurements were performed at the dilution fridge's base temperature ($\approx 50 \text{ mK}$). The temperature dependent measurements were taken at various temperatures without microwave illumination. We used a 20 GHz microwave frequency to explore the practical microwave power values on the Hall bar.

4.3 Results and Discussion

In this experiment, at the high magnetic field side where $B > 0.35 \text{ T}$, the SdHOs are spin split and the resistance maxima are spaced about half filling factors. At lower magnetic field strength, $B < 0.35 \text{ Tesla}$, the resistance maxima in SdHOs do not have spin splitting, so they are spaced at odd integer filling factors, as shown in Figure 4.1 (b). In the intermediate region where spin splitting is partially resolved, the minima points in SdHOs are highly sensitive to temperature variations. Also, the spin split minima are shallower than the cyclotron minima which reflects $\hbar \omega_c > \Delta_s$ in that intermediate regime. We observed a small asymmetry in spin split SdHOs at the zero tilt condition. This could be a result of an asymmetric density of states.
due to small amount of attractive or repulsive scattering centers or a spin dependent scattering [56].

Figure 4.2 The longitudinal resistance as a function of magnetic field (a) at various temperatures with no microwaves; (b) at various microwave powers of 20 GHz with a constant lattice temperature, 50 mK. The independent measurements related to temperature increases and the microwave power increases indicate similar changes in $R_{xx}$ at zero tilt angle.

Figure 4.2 shows the magnetotransport properties of a GaAs/AlGaAs Hall bar heterostructure. Here the tilt angle is zero since the total magnetic field $B$ was oriented
perpendicular to the plane of the 2DES, which is parallel to the current through the sample. Panel (a) shows the longitudinal resistance, $R_{xx}$, as a function of a magnetic field at various temperatures without microwave illumination. At each temperature, the magnetic field range was $0 \leq B \leq 1.25$ Tesla. The $R_{xx}$ variation under the constant frequency of 20 GHz microwaves at 50 mK is shown in panel (b); different traces correspond to different power values of microwaves in the region of $0 \leq B \leq 1.25$ Tesla.

As a novel experimental approach to examine the activation energy at high filling factors, we utilized a microwave induced localized heating technique on 2DES in GaAs/AlGaAs [15]. The idea is to find the best match temperature curves with microwave irradiated curves for further analysis.
Figure 4.3 illustrates the matching of $R_{xx}$ vs $B$ at elevated temperature and $R_{xx}$ vs $B$ under microwaves at the lowest temperature, 50 mK. This significant feature could be used to correlate
the temperature and microwave power and extract the possible heating on the 2DES under microwave conditions. In this experiment, we obtained microwave irradiated traces with different powers that could match the dark traces at each elevated temperature. For example, the $R_{xx} \text{ vs } B$ at 240 mK matches with the trace under -12 dBm microwaves at 50 mK, as shown in Figure 4.3(a). We correlated the temperature and the microwave power from this curve, as shown in Figure 4.3(b). Here, the temperature appears to increase non-linearly with increasing microwave power. For the sake of fitting, we convert that into a log-log plot of the temperature vs. microwave power, as shown in Figure 4.3(c). From linear fitting, we derive a relation $T(mK) \propto (P(mW))^{0.38}$, which agrees with previously reported exponent values in the FQHE regime [15].

Then we tilted the sample to 66.14° and repeated the experiment as previously at zero angle. The results at that angle are summarized in Figure 4.4 by following the same analyzing technique on the collected data.
Figure 4.4 The longitudinal resistance as a function of magnetic field (a) at various temperatures with no microwaves; (b) at various microwave powers of 20 GHz with a constant lattice temperature, 50 mK. The independent measurements related to temperature increases and to microwave power increases indicate similar changes in $R_{xx}$ at 66.14° tilt angle.

Figure 4.4 (a) and (b) show the $R_{xx}$ vs $B$ at different temperatures and under various microwave powers of 20 GHz at 50 mK, respectively. Since the sample is tilted to 66.14°, the total magnetic field can be resolved to two components, which are the out-of-plane and in-plane magnetic field components. The out-of-plane component is perpendicular to the plane of 2DES and the other component parallel to the plane of 2DES. Note that the perpendicular magnetic
field is now not equal to the total magnetic field. Therefore, we have to provide a high total magnetic field to observe the perpendicular magnetic field-dependent features in $R_{xx}$. The magnetic field range was $0 \leq B \leq 3.1$ Tesla for each temperature and microwave power case when the sample was tilted.

Figure 4.5 Matching of power- and temperature- graphs of $R_{xx}$ vs $B$ and the extraction of the electron temperature at a given microwave power at the sample tilt angle $\theta=66.14^\circ$. For example, panel (a) shows $R_{xx}$ vs. $B$ plot obtained at 20 GHz with -6 dBm, coinciding with $R_{xx}$ vs. $B$ plot obtained at 360 mK without microwaves (dark). The results suggest an excellent match between the power and temperature plots over the examined magnetic fields $0 < B < 4$ T. (b) The electron temperature ($T$) as a function of microwave power ($P$), which was extracted by

\[ T \propto P^{0.36} \]

\[ \log T(\text{mK}) = 2.778 + 0.362 \log P(\text{mW}) \]
matching up microwave power curves with temperature curves, as shown in the (a). (c) A suggested simple calibration curve to extract the electron temperature for given microwave power as $T \propto P^{0.36}$.

Figure 4.5(a) depicts the matching of $R_{xx}$ vs $B$ at elevated temperature and $R_{xx}$ vs $B$ under microwaves at the lowest temperature, 50 mK at the tilt angle of 66.1°. Similar to the zero-tilt case, we used this significant feature to correlate the temperature and microwave power and to extract the possible heating on the 2DES under microwave conditions. We correlated the temperature and the microwave power from this curve matching, as shown in Figure 4.5(b). We utilized a log-log plot of the temperature vs. microwave power, and linear fitting suggests that $T(mK) \propto (P(mW))^{0.36}$, as shown in Figure 4.5(c).

The spin split minima appear at odd filling factors. We examine the temperature dependence of $R_{xx}$ at minima points to determine the energy gaps at odd filling factors. We selected four odd filling factors between 5 and 11 for further analysis. Our idea here is to make Arrhenius plots for chosen filling factors and extract the activation gap under the microwave illumination conditions.
Figure 4.6 Extraction of the activation energy at various filling factors. The panels (a) and (c) display the $R_{xx}$ at filling factors 5, 7, 9, and 11, as a function of microwave power at tilt angles $\theta=0^\circ$ and $\theta=66.14^\circ$, respectively. In these panels, the abscissa $P$ of the left panels have been converted to the inverse temperature $T^{-1}$ using the calibration curves for tilt angle $\theta=0^\circ$ and $\theta=66.14^\circ$. These panels also exhibit an activation fit at each filling factor, represented by the dotted black line for each curve.

The $R_{xx}$ minima values at selected odd filling factors were extracted from the spin split minima under different microwave powers at tilt angles, zero and 66.14°. The results are shown in
Figure 4.6 (a) and (c), respectively. The SdHOs were suppressed with an increase of microwave power, and the resistance at spin split minima with odd filling factors was increased. The logarithm of $R_{xx}$ minima is plotted as a function of inverse temperature, known as the Arrhenius plot, after converting the microwave power values to temperatures using calibration curves at angles zero and 66.14°. The results are summarized in Figure 4.6(b) and (d) for angles zero and 66.14°, respectively. The slope of the Arrhenius plot is related to the effective g-factor ($g^*$), as shown in Equation (4.4).

$$\ln(R_{xx,\text{minima}}) = -\frac{\Delta_s}{2k_B T} + \ln(A)$$ \hspace{1cm} (4.4)

where, $\Delta_s = g^* \mu_B B$.

Usually, the magnitude of exchange energy scales with the interaction energies of the electrons [55]. The electrons’ average separation can be calculated using the Coulomb energy,

$$E_{\text{Coulomb}} = \frac{e^2}{4\pi \varepsilon_0 \varepsilon_r l_B}.$$ Here, $l_B$ is the magnetic length which is $l_B = \frac{\hbar}{eB}$. Therefore, it is expected that $\Delta_s \propto \sqrt{B}$.

Figure 4.7 (a) and (b) show the spin energy gap as a function of magnetic field at zero and 66.14° angles, respectively. The results here also indicate that a linear increase of the energy gap is due to exchange energy. However, this linear behavior contradicts the idea that $\Delta_s \propto \sqrt{B}$.

The expected $\Delta_s \propto \sqrt{B}$ may not be true when $E_{\text{Coulomb}} > \hbar \omega_c$. At high filling factors the correlation length would be determined by the Fermi-wave vector ($k_F$) rather than the magnetic length, where, $k_F \propto$ carrier density ($N_s$) [46]. Further, at high odd filling factors only $N_s/\nu$ of the electron population can contribute the exchange energy, $E_{ex}$, since the rest occupy full Landau levels. So, $E_{ex} \sim \left(\frac{e^2}{4\pi \varepsilon_0 \varepsilon_r}k_F/\nu\right)$ [50]. Simply, $E_{ex} \propto B$ for the magnetic field range examined here.
Figure 4.7(a) and (b) Spin energy gap as a function of magnetic field at zero tilt angle and 66.14°, respectively. Dark symbols represent the spin gap while solid lines show the linear fits which follow the given equations in the panels.

We extract the effective g-factors from the gradient of the linear fits in Figure 4.7. Here, we assume that the effective g-factor is the same at all fields in both zero and 66.14° tilted cases to describe the spin gap as $\Delta_s = g^* \mu_B B$. The extracted effective g factors are 6.47 and 2.79 at zero and 66.14° angles, respectively. The intercepts of the graphs give an idea about the thermal broadening of the Landau levels.

4.4 Summary

We measured the spin gap energy at odd integer-high filling factors from 5 to 11 of 2DES in GaAs/AlGaAs heterostructure using microwave photoexcitation induced heating techniques proposed by Mani and coworkers [15]. Compared with conventional temperature dependent measurements, this microwave induced heating technique provides precisely controlled localized heating in the specimen, which is the main advantage of this experimental technique.
We observed that the spin gap energy at odd filling factors were enhanced by the exchange interaction of the system and $E_{ex} \propto B$. The resulting $g^*$ values are higher than the expected bare $g$ value (-0.44) in the GaAs band edge [57].
5 MAGNETOTRANSPORT MEASUREMENTS IN EPITAXIAL GRAPHENE

This chapter will discuss the magnetotransport properties in n-type and p-type epitaxial graphene (EG) samples. Interestingly, we observed broad plateaus develop at $v = 2$ quantum state, which appeared at significantly low magnetic fields in EG samples. However, the resistance values in these plateaus shift away from the quantized value with an increasing magnetic field. This is a rare scenario in EG devices, and it may suggest the need for a linear correction to the Hall resistance.

5.1 Introduction

The quantum Hall effect (QHE) shows well-defined constant values in Hall resistance in for a wide range of magnetic fields or charge carrier densities. These constant Hall resistance values, $R_H$, (at the Hall plateaus) are related with the fundamental physical constants as described as $R_H = h/ve^2$, where $h$ is Planck’s constant, $e$ is the elementary charge and $v$ is an integer for IQHE. Also, these quantized resistance values reproduce very accurately in many quantum 2-DES systems. Due to these remarkable features in IQHE, it is being used in metrology applications as a resistance standard to calibrate resistance values. Further, the ratio of $h/e^2$ is known as the von-Klitzing constant, and it is denoted as $R_K (= 25812.807 \, \Omega)$ [6].

The 2DES in high-quality GaAs/AlGaAs heterostructures exhibit the QHE phenomena; thus, it has been used in metrology applications. However, graphene is replacing the conventional material in metrology applications due to its remarkable properties and the relatively low cost of the QHE devices [58-61].

When SiC substrate is annealed at high temperature (> 1000°C) in an ultra-high vacuum or inert gas environment, Si atoms sublimate while C atoms remain on the SiC surface. The remaining C atoms rearrange to form graphene layers [62]. Interestingly, this EG can be used in
device fabrication without transferring to an insulating substrate. As a unique property, graphene has a linear conical band structure near the Dirac points, allowing the carriers (either electron or holes) to behave as relativistic Dirac particles. When graphene exhibits the anomalous quantized Hall effect at a high magnetic field, these carriers occupy Landau energies that scale as the $\sqrt{B}$, where $B$ is magnetic field. The energy gap between Landau level index $n = 0$ and $n = \pm 1$ is relatively large compared to the energy gaps in other Landau levels and the energy gaps in semiconductor heterostructures’ Landau levels. Therefore, it allows the robust quantized Hall resistance plateau with resistance $h/2e^2$ to correspond to filling factor $v = 4(n + 1/2) = 2$, where $n = 0$ is the lowest Landau level index for single layer graphene [59, 63]. Also, these quantized features in graphene devices can be sustained at higher temperatures and lower magnetic fields than is the case for conventional GaAs/AlGaAs devices. Therefore, EG on SiC is particularly well suited to fabricate quantized Hall resistance devices due to EG’s unique 2D magnetotransport characteristics.
Figure 5.1 The quantization of Hall resistance in the 2DES in (a) EG and (b) GaAs/AlGaAs devices. (a) In an EG sample at a charge carrier density of about \( n \approx 1.5 \times 10^{11} \, \text{cm}^{-2} \) the plateau with filling factor \( i=2 \) becomes visible at relatively low magnetic flux densities of \( B \gtrsim 3 \, \text{T} \) once the longitudinal resistivity disappears. (b) For GaAs/GaAlAs devices, a series of Hall resistance plateaus appear within the typical range of magnetic flux densities (0 T \( \leq B \leq 15 \, \text{T} \)). The \( i = 2 \) plateau starts around \( B = 10 \, \text{T} \) for typically chosen carrier concentrations slightly below \( 5 \times 10^{11} \, \text{cm}^{-2} \). After M. Kruskopf and et al. (2018) [61].

Figure 5.1 compares the resistance quantization in the 2DEG of EG and GaAs/GaAlAs. Generally, EG samples exhibit the \( v = 2 \) plateau at relatively low magnetic fields values compared with the conventional GaAs/AlGaAs. Also, the quantized resistance values appear over a more comprehensive range of magnetic fields compared to the range for GaAs/AlGaAs. The quantized Hall resistance at \( v = 2 \) plateaus in EG usually can extend from low magnetic field values about 2 Tesla to very high values above 15 Tesla with suitable control of the charge carrier concentration [61]. Further, this plateau-broadening effect is realized due to the charge transfer at the substrate/graphene interface [64, 65]. The longitudinal resistance values in GaAs/AlGaAs devices are generally larger than the resistance in graphene devices [61].
In an ideal situation, if the magnetic field is at the integer filling factor $v$, then the value of quantized resistance is exactly $h/ve^2$. But in reality, the Hall resistance can deviate from the expected quantized value due to unwanted dissipation in the device because $R_{xx}$ is not precisely zero. Also, the contact resistance to the 2DES should be less to avoid excessive dissipation and observe the standard quantized resistance [58].

5.2 Experimental section

The digital multi-meter based DC-electrical measurements were performed on EG Hall bar devices. The Hall bar devices were wired into chip carriers loaded into the puck in the bore of the superconducting magnet of the Triton cryo-free dilution refrigerator system. The sample could be tilted in-situ. The sample space temperature could be controlled accurately from the refrigerator. Here, we measured n-type and p-type EG Hall bar devices in a wide temperature range of 30 mK to 28 K. All the samples have a length-to-width ratio equal to 1.
5.3 Results and Discussion

5.3.1 Temperature dependency of magnetotransport measurements in EG

Figure 5.2 Temperature dependency of magnetotransport properties in EG, within $30 \text{ mK} \leq T \leq 28 \text{ K}$. As temperature increases, $R_{xx}$ values increase at high magnetic field side, Hall resistances shift away to lower resistance from the quantized value. Panels (a) and (b) show the results from sample d13, while panels (c) and (d) exhibit the results from sample d14; both are p-type samples. Panels (a) and (c) show the $R_{xx}$ variation with the temperature, while (b) and (d) exhibit the Hall resistance variation with the temperature.

Figure 5.2 illustrates the temperature dependency of magnetotransport characteristics of p-type EG Hall bars within $30 \text{ mK} \leq T \leq 28 \text{ K}$ in a dilution refrigerator. Here, the magnetic field $B$ is oriented perpendicular to the current and the plane of the devices. Hence, the tilt angle is zero. The $R_{xx}$ at the high magnetic field side increases with temperature increases, as shown in Figure 5.2 (a) and (c) in both samples. Interestingly, the Hall resistances at high
magnetic field sides shift away from the quantized value with the magnetic field increases. The weak localization-like peaks in both samples are very narrow within the examined temperatures, and the peak values drop in resistance with rising temperature.

Figure 5.3 Deviation of Hall resistance from the quantized value in p-type EG with temperature increment. (a) The quantized Hall resistance of sample d13 deviates from the standard value $h/2e^2$ with an increase of the temperature. (b) The $R_{xx}$ variation (left ordinate) and the Hall resistance variation (right ordinate) of sample d13 at 5 Tesla as a function of temperature. (a) The quantized Hall resistance of sample d14 deviates from the standard value $h/2e^2$ with the temperature increase. (d) The $R_{xx}$ (left ordinate) and the Hall resistance variation (right ordinate) of sample d14 at 5 Tesla as a function of temperature.
ordinate) of sample d14 at 5 Tesla as a function of temperature. In panels (a) and (c), the pink dash lines represent the $h/2e^2$ value.

Figure 5.3 describes the deviation of Hall resistance from the expected quantized value $h/2e^2$ in p-type EG with an increase of temperature. Figure 5.3 (a) and (c) show the Hall resistance as a function of temperature in both samples. The pink dash lines represent the expected quantized value of the $h/2e^2$. Figure 5.3 (b) and (d) depict the longitudinal resistance at 5 Tesla (left ordinate) and Hall resistance (right ordinate) as a function of temperature, in samples d13 and d14, respectively. The sample d13 shows the development of plateau at ~1.2 T at 30 mK, as in Figure 5.3 (a). However, the quantized resistance of the plateau starts decreasing at ~2.2 T when the magnetic field increases, while $R_{xx}$ increases. The sample d14 developed the plateau at ~1.6 T and saturated the entire magnetic field at 30 mK, as in Figure 5.3 (b). However, the Hall resistance of that plateau deviated from the quantized value at high temperatures when magnetic field increases. The Hall resistance at a given magnetic field shifts away exponentially from quantized values with an increase of temperature, as shown in Figure 5.3 (b) and (d) for both samples, d13 and d14.
Figure 5.4 Temperature dependency of magnetotransport properties in n-type EG Hall bar device, within $30 \text{ mK} \leq T \leq 28 \text{ K}$. In Sample: d23, as temperature increases, $R_{xx}$ increases at high magnetic field side as shown in panel (a), while Hall resistances shift away from the quantized value as in panel (b). (c) The $R_{xx}$ (left ordinate) and the Hall resistance (right ordinate) variations of sample d23 at 5 Tesla as a function of temperature. (d) The quantized Hall resistance of sample d23 deviates from the quantized value with temperature increases.

Figure 5.4 (a) shows that the $R_{xx}$ at the high magnetic field side goes up with the temperature while Hall resistance shifts away from the quantized value as shown in Figure 5.4(b) in this n-type EG sample. The weak localization-like peak in this sample is also very narrow in examined temperatures while it drops down with rising temperature. Figure 5.4(d) shows the Hall resistance value quantized to lower than the expected value $\frac{h}{2e^2}$ in this graphene sample, and the resistance shifts away from the quantized value with increasing temperature.
Interestingly, the Hall plateau for sample d23 also developed at a small magnetic field value ~0.7 Tesla at 30 mK.

Remarkably, all these graphene samples showed the development of Hall plateaus at significantly small magnetic fields even at high temperatures as described above.

5.3.2 Angle dependency of magnetotransport measurements in EG

We performed angle dependency magnetotransport measurements at 30 mK and 28 K to further analyze these EG samples. Interestingly, these measurements reveal the 2D nature in the charge carrier systems in these EG samples even at 28 K, which reflects the quality of these samples.

Figure 5.5 Angle dependency of magnetotransport measurements of p-type EG Hall bar devices, within the tilt angles $0 \leq \theta < 90^\circ$. Various colors represent different angles. Panels (a) and (c) show the $R_{xx}$ as a function of the total magnetic field at various tilt angles. Panels (b) and (d) exhibit the Hall resistance vs total magnetic field at different tilted angles for samples d13 and d14, respectively.
Figure 5.5 (a) and (c) show the angle dependency of the longitudinal resistances $R_{xx}$ of samples d13 and d14 as a function of total magnetic field at 30 mK. Figure 5.5 (b) and (d) show the Hall resistances $R_{xy}$ of samples d13 and d14 as a function of total magnetic field at various angles at 30 mK. Note that all four panels in Figure 5.5 are plotted as a function of total magnetic field.

$\text{Figure 5.6 Angle dependency of magnetotransport measurements of p-type EG Hall bar devices based on EG, within the tilt angles } 0 \leq \theta < 90^\circ. \text{ Panels (a) and (c) show the } R_{xx} \text{ as a function of the perpendicular magnetic field } (B \cos \theta) \text{ at various tilt angles. Panels (b) and (d) exhibit the Hall resistance vs perpendicular magnetic field } (B \cos \theta) \text{ at different tilted angles for samples d13 and d14, respectively.}$

Figure 5.6 shows the magnetotransport measurement of samples d13 and d14 as a function of perpendicular magnetic field, $B \cos \theta$, where $\theta$ is tilt angle. The $R_{xx}$ and $R_{xy}$ measurements at different tilt angles in both samples, d13 and d14, collapse on to curves at zero
tilt angle when x-axes are scaled to $B \cos \theta$. This is a well-known feature for a 2D carrier systems [66]. Hence, it confirmed that the measurements in these samples at 30 mK are from their 2D carrier systems.

Figure 5.7 Angle dependency of magnetotransport measurements of p-type EG, within the tilt angles $0 \leq \theta < 90^\circ$ at 28 K. (a) and (c) show the diagonal resistance as a function of the total magnetic field at various tilt angles. (b) and (d) exhibits the Hall resistance as a function of the total magnetic field at different tilt angles for samples d13 and d14, respectively.

The angle dependency of the longitudinal resistances $R_{xx}$ of samples d13 and d14 as a function of total magnetic field at 28 K are shown in Figure 5.7 (a) and (c). Figure 5.7 (b) and (d) show the Hall resistances $R_{xy}$ of samples d13 and d14 as a function of total magnetic field at various angles at 28 K. All four panels in Figure 5.7 are plotted as a function of total magnetic field.
Figure 5.8 Angle dependency of magnetotransport measurements of p-type EG Hall bar devices, within the tilt angles $0 \leq \theta < 90^\circ$ at 28 K. Panels (a) and (c) show the $R_{xx}$ as a function of the perpendicular magnetic field ($B \cos \theta$) at various tilt angles. Panels (b) and (d) exhibit the Hall resistance vs perpendicular magnetic field ($B \cos \theta$) at different tilt angles for samples d13 and d14, respectively.

Figure 5.8 depicts the $R_{xx}$ and $R_{xy}$ measurements of samples d13 and d14 as a function of $B \cos \theta$. The $R_{xx}$ and $R_{xy}$ measurements at different tilt angles in both samples collapse onto curves at zero tilt angle when the x-axes are scaled to $B \cos \theta$. Thus, it confirms the 2D nature of these systems even at 28 K.
Figure 5.9 Angle dependency of magnetotransport measurements on n-type EG, within the tilt angles $0 \leq \theta < 90^\circ$ at 30 mK in panel (a) and (b) while (c) and (d) show similar measurements at 28 K. Different color lines represent various angles.

The angle dependency of the $R_{xx}$ of sample d23 as a function of total magnetic field at 30 mK and 28 K is shown in Figure 5.9 (a) and (c). Figure 5.9 (b) and (d) depict the $R_{xy}$ of sample d23 as a function of total magnetic field at various angles at 30 mK and 28 K.
Figure 5.10 shows the $R_{xx}$ and $R_{xy}$ measurements of sample d23 as a function of $B \cos \theta$. The $R_{xx}$ and $R_{xy}$ measurements at different tilt angles collapse on to curves at zero tilt angle when the x-axes are scaled to $B \cos \theta$. This confirms that the measurements at 30 mK and 28 K are from the 2D carrier system in d23. Interestingly, our tilted field measurements on both p-type and n-type samples reveal that the 2D nature in samples are protected even at high temperatures.

Remarkably, all these graphene samples show that the Hall plateaus developed at significantly smaller magnetic fields even at high temperatures compared to previous studies in IQHE at $v = 2$ quantum states in graphene [61, 67-69]. Interestingly, the $v = 2$ plateaus in these EG samples appeared through the entire range of the high magnetic field side. Also, they did not
show developments of plateaus at other high filling factors. This could be a result of the charge carrier transfer from the surface-donor states of substrate SiC and EG. A possible transferring mechanism is based on magnetic field dependent pinning of the filling factors determined by the dominance of the quantum capacitance over classical capacitance [70]. Thus, the Landau levels remain completely filled over a wide range of magnetic field and show broad plateaus in the Hall resistance.

Also, we observed a rare scenario in EG devices, which is the quantized Hall resistance at \( \nu = 2 \) shifts away from the quantized values when the magnetic field increases. This does not usually happen in these types of EG devices. This may suggest implementing a linear correction to \( R_{xy} \) as described as shown in Figure 5.11.

![Figure 5.11 Correction of quantized Hall resistance in EG samples with comparatively large longitudinal resistance. \( R_{xy} \) plotted as a function of \( R_{xx} \) in two different p-type and a n-type samples at various temperatures as shown in panels (a), (b), and (c), respectively.](image)

Generally, the Hall resistance deviates from the quantized value with increasing temperature. The remarkable thing about these EG samples is that the \( R_{xx} \) is very large comparable to \( R_{xy} \). Under these rare conditions, there is a correction to \( R_{xy} \) that is linearly proportional to \( R_{xx} \) as shown in Figure 5.11. This was known from the GaAs system, but their
corrections are small due to their small $R_{xx}$ and it is difficult to observe. Here large $R_{xx}$ values make that difference, and the linear correction is visible. This could be the reason for the Hall resistance deviation from the standard quantized value at $\nu = 2$.

5.4 Summary

We performed magnetotransport measurements in both p-type and n-type EG samples in a dilution refrigerator at temperatures from 30 mK to 28 K. We observed a rare scenario in our measurements, the quantized Hall resistance at $\nu = 2$ shifts away from the standard quantized values when the magnetic field increases. This is unusual in EG devices. This deviation of Hall resistance suggests a linear correction to Hall resistance in these EG samples. Remarkably, $\nu = 2$ Hall plateaus are developed at very low magnetic fields in these p-type and n-type EG samples. Although the magnetotransport measurements show that the resistance values at $\nu = 2$ are not well-quantized at any magnetic field level, it is interesting that the $\nu = 2$ plateaus in these EG samples are spreading through entire range of the high magnetic field side and they do not show development of any other high filling plateaus. This could be a result of the carrier transferring from the substrate to EG and it leads to a robust Hall plateau at $\nu = 2$. 
LIMITING NUCLEATION DENSITY IN CVD GROWTH GRAPHENE USING A DUAL TREATMENT TECHNIQUE

This chapter will discuss the growth of single-crystal graphene on polycrystalline copper foils using low pressure chemical vapor deposition (LPCVD) techniques. Here, we will discuss a dual treatment technique, which consists of two main steps (using a copper enclosure and exposing the copper surface to oxygen before introducing CH₄ and H₂/Ar gases) to limit the nucleation density.

6.1 Introduction

Graphene [71], a single layer of carbon atoms arranged in a hexagonal lattice, has attracted significant interest due to a host of intriguing electronic properties [72, 73] and its promising expectations as a key material for future electronics applications [74-80]. Several methods of graphene growth have been employed, including exfoliation from graphite [81, 82], epitaxial growth on SiC [83-86], mechanical cleavage from bulk graphite [87], chemical vapor deposition (CVD) [88-94] and the reduction of graphite oxide [95, 96]. Among these methods, the CVD technique has attracted great interest in academia and industry since it is a promising method for producing comparatively large scale, high quality graphene for a reasonably low cost.

The transition metal supported CVD technique utilizing the thin films/foils of copper [88, 89], nickel [97], ruthenium [98, 99], platinum [100], and cobalt [101] have been examined previously. In these methods, thin films/foils assist in decomposing the carbon groups from the precursor and in the nucleation of the graphene crystal. The graphene synthesis process on the metal catalyst is apparently affected by factors such as the limit of the carbon solubility in the metal, crystallographic parameter of the metal, and thermo-dynamic variables such as growth temperature and pressure [102]. The CVD growth of graphene on copper is attractive since
copper is known as a very low carbon solubility catalyst, and the growth process is restricted to
the surface of the catalyst [88, 103-105]. The graphene growth process on a copper surface is
known to be self-limiting, producing a single layer of graphene since carbon precursor
decomposition is not favorable on graphene [106-108].

In contrast to the self-limiting growth on copper, the few layer growth of graphene on
copper had been reported by showing that the self-limit effect is broken [109-112] sometimes by
specific conditions, such as higher methane concentration [102, 110] and impurity enhanced
growth [113]. In addition, the graphene grown on copper is polycrystalline on a micrometer
scale. This polycrystalline nature is a consequence of the growth process. The graphene growth
process in CVD consists of formation of stable nuclei, those nuclei grow and merge with each
other at the grain boundaries, resulting in the polycrystalline nature of CVD graphene grown on
copper. The polycrystalline morphology degrades the quality of the graphene which is
problematic for graphene applications. Thus the development of growth techniques to reduce the
density and orientation of two dimensional grain boundaries is important for the production of
high quality single crystal graphene.

There has been a significant amount of research conducted on developing controlled
growth of high quality graphene, using techniques such as substrate electro-polishing [91, 108,
114-116], the two step CVD process [117], seeded growth CVD technique [118, 119], oxygen
assisted CVD [120], plasma assisted CVD technique [121-123], and proton assisted CVD growth
method [124]. To tailor the grain boundaries, it is essential to obtain control over the location of
the nucleation sites and nuclei shapes concerning the underlying crystal symmetry of the copper
substrate. Previous works have reported a clear correlation between the shapes of the graphene
nuclei and the crystal orientation of the underlying copper substrate [106, 125, 126]. For
example, the six-fold symmetry of the Cu(111) surface leads to the formation of nuclei shapes with six lobes, while four-fold symmetry of the Cu(100) surface produces a four-lobes nuclei shape[126]. In addition, the shape and the size of the graphene nuclei are also influenced by the growth parameter such as pressure [112, 125, 127-129], temperature [117, 129], cooling rate [130], precursors composition and flux [111, 131]; these factors have been investigated both experimentally and theoretically.

6.2 Experimental section

Figure 6.1 Preparation of copper pocket and the CVD growth profile of graphene. (a) The cleaned and dried copper foil rinse after acetone, methanol, deionized water, and nitrogen gas. (b) The copper enclosure from the pre-oxidized copper foil on a hot plate for 15 minutes under ambient conditions. (c) This panel depicts the graphene growth profile used in this work.

Commercially available copper foil (25 μm thick) was cleaned using acetone, methanol, and deionized water, respectively. Both sides of the dried copper foil were pre-oxidized [90, 132, 133] on a hot plate for 15 minutes under ambient conditions. Then a nearly airtight copper pocket was made for the LPCVD, as in Figure 6.1. The copper pocket was loaded in a 1-inch quartz tube mounted inside a tube furnace. The CVD chamber, including the gas transferring
tubes were pumped from ambient pressure to ~40 mTorr to evacuate the entire system. Then the copper pocket was heated to ~1000°C without supplying any gas. When growth chamber reached the ~1000°C, oxygen gas was introduced to the chamber for 10 minutes. Later, CH₄ and Ar/H₂ gas mixture was introduced with desired flow rates to the growth chamber simultaneously for the growth of graphene. After 45 minutes, the growth chamber was cooled down by simply shutting off the heating coils and later opening the furnace. The graphene growth profile is shown in Figure 6.1(c).

### 6.3 Results and Discussion

![Comparison of nucleation density](image-url)

*Figure 6.2 Comparison of the nucleation density of LPCVD growth graphene in the outer (a) and inner (b) surfaces of the copper enclosure. Note that the nucleation density in the outer surface of the copper enclosure is significantly higher than in the inner surface of the enclosure.*

As the initial technique to limit the nucleation density of graphene, an oxygen assisted pre-anneal technique was applied on the copper foil under ambient conditions. Then the second technique exposed the copper pocket to oxygen gas which also reduced the nucleation density. The outer surface of the copper pocket showed a comparatively very high nucleation density with relatively small graphene flakes, as shown in Figure 6.2 (a). The low nucleation density on
the inner wall of the pocket was attributed to the redeposition of copper in the confined volume, which could help the inner copper surface keep on smoother with less defect sites throughout the growth process [134]. In CVD growth on the copper surface process, CH₄ dehydrogenates and produces hydrocarbons. The preadsorbed oxygen on the copper surface can enhance the dissociation of hydrocarbons. Thus, nucleation density can be controlled on an oxygen-rich copper surface [112, 119, 132].

![Image of graphene flakes](image)

Figure 6.3 The optical microscope images of single crystal graphene with hexagonal star shape on the oxidized copper surface. The areas of the bright contrast are graphene, and the dark reddish areas are the exposed regions without graphene.

The majority of observed graphene flakes here are hexagonal star shapes of the same size, as shown in Figure 6.3. These single crystal graphene flakes were transferred on to the target metal contacts on Si/SiO₂ using the Polydimethylsiloxane (PDMS) assisted hybrid transferring techniques.
Figure 6.4 Schematic diagram of PDMS assisted hybrid transferring process of CVD growth single crystal graphene. (a) PDMS is stuck on the glass slide and then the CVD growth graphene flake is gently pressed on the copper to the PDMS. (b) Using oxygen plasma, etch away the excess graphene on the other side of the copper surface. (c) After plasma etching, the glass slide with PDMS/graphene/copper floats on FeCl$_3$ liquid. Then FeCl$_3$ reacts with copper foil and etches away the copper. Graphene flakes will stick onto PDMS. (d) After completely dissolving the copper, the graphene/PDMS/glass slide is cleaned and transferred onto a target substrate using a dry transfer setup which is not shown in here. The home-built dry transfer setup is done with a micro-stage at the transferring step. (e) The final transferred graphene flake on the metal contacts.

Figure 6.4 depicts the steps of PDMS assisted hybrid transfer technique utilized here to transfer single crystal graphene flakes onto a target substrate with contact patterns.
Figure 6.5 Device fabrication from single crystal graphene grown by LPCVD. (a) Single crystal graphene (bright area) on an oxidized copper surface (reddish area). (b) Single crystal graphene on PDMS at the intermediate step of transferring graphene flake to target contact pattern. (c) Successfully transferred single crystal graphene domain on target contact pattern on Si/SiO\textsubscript{2} substrate. (d) Close view of the single crystal graphene on the Au/Cr contacts.

Figure 6.5 shows the single crystal graphene domain on an oxidized copper surface. This graphene flake was transferred onto a target contact pattern. Figure 6.5 (b) shows the graphene domain is on the Polydimethylsiloxane (PDMS) at the intermediate step of the transferring process. The transferred single crystal graphene onto the target contacts pattern on the Si/SiO\textsubscript{2} substrate is shown in Figure 6.5(c). Figure 6.5(d) shows the close view of the graphene flake on the contacts. It’s worth noticing that we can transfer complete graphene flakes onto the target metal contacts with a highly successful rate using this transferring technique.

6.4 Summary

We used dual treatment techniques, which are (i) surface peroxidation and (ii) copper enclosure, to control the nucleation density in graphene growth on a polycrystalline copper surface using LPCVD. By analyzing optical images, we confirmed that the nucleation density was limited to the 4 mm\textsuperscript{2} on the polycrystalline copper.
7 STRAIN RELAXATION IN VARIOUS SHAPES OF SINGLE CRYSTAL GRAPHENE GROWN BY CVD TECHNIQUE

This chapter will present the growth of single-crystal graphene on polycrystalline copper foils using low pressure chemical vapor deposition (CVD) techniques studied as part of this dissertation work [135, 136]. I show that an impurity assisted growth mechanism governs the growth of single crystal graphene through isotropic diffusion, creating two-fold, four-fold, and six-fold symmetries in the graphene crystals. However, single crystal graphene grown through anisotropic diffusion does not show the signs of an impurity assisted growth mechanism. In conclusion, strain relaxation in two-fold and four-fold symmetric graphene structures through isotropic diffusion is more complex than the six-fold structures created via isotropic diffusion, which shows multiple orientations in low symmetry structures.

7.1 Introduction

Previous studies show that the strain variations in graphene open opportunities to adapt the transport properties of graphene based electronic devices [137-139]. CVD growth graphene is strained most of the time because of the mismatch between the thermal expansion coefficients of graphene and the substrate. That mismatch in coefficient results in a different thermal expansion in graphene and substrate as the system cools down to room temperature after the growth process [140, 141]. The strain may not be released entirely even after transferring the CVD graphene from the growth substrate [142]. Also, if a supporting substrate is involved in transferring process, that can add additional strain due to deformation of the supporting substrate [143]. The nature of these strains and their relaxations in CVD growth graphene is not fully understood as a factor of the shape and the size of the graphene flakes. Our study focuses on the
strain relaxation in different shapes of single crystal graphene grown by LPCVD on copper substrate.

7.2 Experimental Section

![Figure 7.1](image)

Figure 7.1. Preparation of a copper pocket and the CVD growth profile of graphene. (a) shows the cleaned and dried copper foil after the rinse from acetone, methanol, and deionized water. (b) The pre-oxidized copper foil was placed on a hot plate for 15 minutes under ambient conditions. (c) The enclosure from oxidized copper foil was placed in a home-built CVD system for graphene growth. (d) This panel depicts the graphene growth profile used in this work. Reproduced from T.R. Nanayakkara et al. (2020) [135].

Commercially available copper foil (25 μm thick) was utilized in the growth process. It was rinsed with acetone, methanol, and deionized water, respectively. Both sides of the dried copper foil were pre-oxidized [90, 132, 133] on a hot plate for 15 minutes under ambient conditions, and then a copper enclosure was made for the LPCVD, as shown in Figure 7.1. The copper enclosure was kept in a 1-inch quartz tube mounted inside a tube furnace. The CVD chamber, including the gas transferring tubes, were pumped from ambient pressure to ~40 mTorr to evacuate the entire system. Next, a H₂/Ar gas mixture was introduced to the CVD chamber while pumping. Here, the stabilized pressure inside the chamber was ~340 mTorr. Subsequently,
the copper pocket was heated to ~1000°C. At this stage, the valve for the H₂/Ar gas mixture was closed and the copper pocket was annealed at low pressure for ~60 minutes. Then H₂/Ar and CH₄ gaseous species were introduced into the CVD chamber simultaneously, with desired flow rates for graphene growth. After growth, the system was cooled to room temperature by simply shutting off the heating coils and opening the furnace. The graphene growth profile is shown in Figure 7.1(d).

A Park XE7 atomic force microscope (AFM) and an optical microscope served to examine the diverse growth morphologies of the graphene. AFM and optical images were obtained under ambient conditions, roughly two days after the growth, and the samples were stored under ambient conditions. Here, the root mean squared smoothness was determined by analyzing AFM images to compare the surface smoothness of the graphene and the exposed adjacent copper surface with no graphene. The parameter λ, which is the average distance between two adjacent steps, is defined as λ = d/(n – 1), where d is the distance between the n number of steps. ΔSq gives the relative root mean squared smoothness, defined by Equation (7.1).

\[ \Delta S_q = \left( \frac{(S_q)_{\text{outside the flake}} - (S_q)_{\text{inside the flake}}}{(S_q)_{\text{outside the flake}}} \right) \times 100\% \] (7.1)

Sq measurements were taken from the roughness analysis by considering the same surface areas both inside and outside the graphene domain, utilizing XEI software developed by Park Systems.

7.3 Results and Discussion

In this study, an oxygen assisted pre-anneal technique was applied on the copper foil under ambient conditions to suppress the nucleation density of graphene. A copper pocket was
used to realize these results, and all the shapes described here are observed on the inner surface. The outer surface of the copper pocket had a comparatively very high nucleation density with relatively tiny graphene domains. The H$_2$/Ar gas flow was not supplied while annealing to maintain the low pressure condition during the annealing. Perhaps, low pressure annealing enhances the possible surface reconstructions of copper surface to get more smoothness control of the carbon solubility during the growth process, as it leads to the creation of graphene islands on the melt copper surface with different shapes as well as low nucleation density.

![Figure 7.2](image)

**Figure 7.2.** Density distribution of various shapes of single crystal graphene domains in each zone of the copper enclosure inner surface. In the histograms, the Y-axis represents the nucleation density for isotropic growth crystals with 6-lobes, 4-lobes, 2-lobes, and anisotropic growth crystals with 6-lobes.

Figure 7.2 shows the distribution of graphene flakes on the inner surface of the copper enclosure. It suggests a variation in the density of graphene domains along the direction of gas flow within the CVD chamber with greater density upstream compared to the downstream regions.
Figure 7.3 Schematic illustration of the possible growth mechanism of single crystal graphene on the inner wall of the copper pocket utilized in this study (not to scale). (a) The oxidized copper foil is folded into a pocket. (b) Cross section of the copper pocket. The red outline represents the oxide layer in both the outer and inner walls of the pocket. Black dots represent the impurity particles, while the yellow intermediate layer shows the non-oxidized copper region. (c) This panel depicts a close view of the circled region in panel (b). (d) The oxide layer is released from the surface more effectively around the impurity particle by promoting copper sublimation at low pressure. The origin of nucleation is possible on the impurity particles. (e) Perspective view of a single crystal graphene flake. Reproduced from T.R. Nanayakkara et al. (2020) [135].

Figure 7.3 suggests a possible impurity assisted growth mechanism of single crystal graphene. The oxidation under ambient conditions can introduce impurities onto the copper surface, which can act as seeds for nucleation [119]. Here, the nucleation could be started by an impurity particle on the copper surface. However, the impurity particles that appear in this study are larger than previously reported impurities on the copper surface [119]. During the anneal and growth process, the copper oxide is perhaps detached from the surface easily around the impurity particle since copper sublimation increases at lower pressure [144]. The gas precursors could enter the copper pocket through gaps of the pocket and possibly by diffusion through the walls of the pocket. Then, the nucleation could initiate on the impurity particle [119] and continue the
growth of graphene on fresh copper below the neighboring surface, as illustrated in Figure 7.3(d). Thus, it seems that the graphene grows below the adjacent exposed surface. The AFM topographic study shows that graphene flakes observed here are sunk below the neighboring surface. Further analysis confirmed that observable flakes consist of graphene, and there is no graphene outside the flakes.

Figure 7.4 The AFM topographical images of graphene flakes on the copper surface demonstrate six-fold symmetry. The bright surface is the exposed region with no graphene, and the region with dark contrast is graphene. (a) shows the six lobe-star like polygon shape graphene flake with isotropic diffusion along the vertices. According to the AFM height profile in panel (b) which is along the green line in panel (a), this graphene flake appears below the adjacent copper surface. (e) and (f) depict the height profiles on the nanosteps at the center and dendritic regions of the flake in panel (a), respectively. Panel (c) shows the six-fold symmetric microscale flower shape of graphene, suggesting an isotropic diffusion along the vertices. Panel (d) reveals that the flake lies below the neighboring copper surface. An impurity particle appeared at the geometric center of this flake, as shown in panel (g), which also shows the height profile of nanosteps at the center of the crystal. The height profile in the wide dendritic region is shown in panel (h). Reproduced from T.R. Nanayakkara et al. (2020) [135].

Figure 7.4 illustrates topographic AFM images of single crystal graphene with six-fold symmetry, which are grown by LPCVD techniques. Commonly, the six-fold symmetry of this type of crystal is governed by the underlying crystal orientation of the copper surface, i.e.,
typically reflects the six-fold symmetry of the Cu(111) facet [106, 126, 145]. Due to its six-fold symmetry, the nucleation starting point possibly would be the geometric center of the crystal. A nanoparticle in the geometric center of the graphene flake possibly served as the nucleation center for graphene growth. It suggests an impurity assisted growth mechanism here. The distances between periodic nanoscale steps, $\lambda$, at random locations inside the graphene flake are roughly the same. This characteristic suggests the flake consists of monolayer graphene since the $\lambda$ is supposed to vary with the number of layers [146].

Figure 7.5 illustrates AFM topographical images of graphene flakes with two-fold and four-fold symmetry. Figure 7.5(a) shows the two-fold symmetric “hourglass” shape of single...
crystal graphene. The terrace steps on this crystal are very dense near the boundaries of two receptacles and display lower density near the neck region of the flake, as described in Figure 7.5(e) and Figure 7.5(f), respectively. An impurity particle is localized at this graphene crystal's geometric center indicating an impurity assisted growth mechanism as depicted in Figure 7.5(e).

The hour-glass shaped graphene crystals are rarely reported in CVD graphene since they grow on a high index facet of copper [126]. Figure 7.5(c) shows a four-fold symmetric graphene crystal. The four-fold symmetric graphene flakes have been reported on Cu(100) surface previously [126]. As shown in Figure 7.5(g), an impurity particle appears near to the geometric center of this graphene domain. A wider crest appears with less dense steps near the middle of the graphene flake, while denser steps exist with slender crests close to the boundaries of the lobes, as shown in Figure 7.5(h).

Figure 7.6 The AFM topographical images of the LPCVD growth single crystal graphene on the copper surface. Single crystal graphene flakes grown via anisotropic diffusion are shown in (a) and (c). These graphene domains lie below the neighboring copper surface, as shown in height profiles in panels (b) and (d). Panel (e) depicts the height profile end-to-end along the green line in the inset, which sits in the solid yellow square region in the graphene domain in panel (a). (f) shows the height profile on the graphene surface in the area covered within the dashed white square in panel (a). (g) and (h) represent that step edges in the solid yellow square and dashed
white square in graphene crystal in panel (c), respectively. Reproduced from T.R. Nanayakkara et al. (2020) [135].

Figure 7.6 shows six-lobe graphene flakes growth via very anisotropic diffusion with two-fold symmetry. This type of shape possibly grows on the Cu(310) facet [126]. There is no sign of impurity particles at the geometrical center of the graphene domain, which indicates that the impurity assisted growth mechanism may not be valid here. The step driven competitive growth mechanism [147] can be suggested as a possible growth mechanism for these anisotropic flakes.

7.4 Summary

This study investigated the growth and strain relaxation in various shapes of single crystal CVD graphene grown on the inner wall of a copper pocket. The AFM topographical analysis revealed that the single crystal graphene domains appeared below the neighboring copper surface. An impurity assisted growth mechanism leads to the growth of single crystal graphene via isotropic diffusion, forming two-fold, four-fold, and six-fold symmetries in the resulting crystals. In addition, single crystal graphene regions formed via anisotropic diffusion also appear here, but they do not show evidence of an impurity assisted growth mechanism. Nevertheless, the density and size of the steps are different in each graphene flake, like a fingerprint for each flake. Our study also explains that strain relaxation in two-fold and four-fold symmetry graphene crystals via isotropic diffusion is more complicated than that for the six-fold symmetric crystals via isotropic diffusion, which reflects the various step orientations in two-fold and four-fold symmetric flakes. The periodic nanoscale steps observed in graphene crystals are due to possible strain relaxation during the CVD cooling process occurring due to the different thermal expansion coefficients of copper and graphene.
We study the transport properties of a graphene p-n junction which is formed in a single gated graphene field effect transistor. Here, an electrical stressing-voltage technique was utilized to create the p-n junction by modifying the electrostatic potential in the SiO₂ substrate. We examine the transport characteristics of the Dirac points localized in the perturbed and unperturbed regions in the graphene channel.

8.1 Introduction

Graphene [81, 148] denotes a single layer of carbon atoms arranged in a honeycomb structure. This material has attracted considerable theoretical and experimental interest due to its unusual characteristics [72, 77, 80, 149, 150]. The Fermi level of the pristine graphene lies precisely at the Dirac or charge neutrality point. A feature of graphene is that the Fermi level can be swept across the Dirac point into a conduction or valence band by applying an external gate voltage. For pristine graphene, one expects a V-shaped conductance vs. gate voltage relation. The shape of the characteristic curve, the value, and the location of the charge neutrality point provide useful information regarding the carrier mobility, the asymmetry between electron and hole conduction, the doping level, etc., in non-ideal graphene [81, 149, 151]. The V-shaped conductivity vs. gate voltage curve, implying a single Dirac point, has been widely observed in graphene-based experiments. However, unusual W-shaped conductivity vs. gate voltage curves, which imply double neutrality points, are rarely observed in single gated graphene field effect transistors.
Double neutrality points in a single device are the signature of the existence of p-n junctions in graphene. Such p-n junctions can be realized by using multiple gates in a single device to tune the channel to n-p-n, p-n-p, n-n'-n, and p-p'-p type junctions. The device then shows the double neutrality points for a specific range of parameters [152-154]. Double neutrality points can also be exhibited in p-n junctions created by separate chemical doping [155-157]. Here, the misalignment of Fermi energies between in-homogeneously doped local zones in graphene leads to double neutrality points. The invasive nature of contacts also leads to the double neutrality points in graphene transport measurements [158, 159]. These multiple gating methods require additional or special sample preparation steps, which might increase fabrication complexity for real-world applications. Chiu et al. reported a novel approach to creating p-n junctions in single layer graphene on SiO₂ substrate by changing the local electrostatic potential without multiple gates [160]. This technique has the advantage of electronic modification occurring in the substrate but not in the graphene film. Yu et al. also reported p-n junction formation in graphene via this electrical stressing-voltage method. They reported that mobility improvement is realized by reducing the impurity induced scattering in graphene as a result of low-level voltage stressing [161]. Therefore, this technique has been applied to improve the quality of graphene-based devices [93, 162, 163].

8.2 Experimental Section
Figure 8.1 (a) An optical image of single layer graphene on top of the SiO$_2$ substrate. The white dotted line shows the boundary between SiO$_2$ and graphene. (b) AFM topographical image of the graphene on top of the SiO$_2$. The insert shows the height profile along the red line. (c) A schematic diagram of a Hall bar device. Here $V_{xx1}$ and $V_{xx2}$ are the longitudinal voltages, and $V_{xy1}$ and $V_{xy2}$ are the Hall voltages examined in this study. S and D represent the source and drain contacts. (d) The Dirac point shifting and the formation of the pn-junction after the vacuum and current anneal. (e) Tuning the Fermi level in a graphene device by sweeping the back gate voltage can allow for the emergence of a p-n junction. Here, the two neutrality cones characterize two regions in graphene, and the yellow plane represents the Fermi level of the system.

CVD graphene [108, 116, 135, 164] was fabricated into millimeter-scale devices using wet transfer techniques and photolithography, and four-terminal DC electrical measurements were carried out in a vacuum. The color contrast of graphene on 300 nm thick SiO$_2$ on p-Si confirmed monolayers, which was consistent with the height profile (~1 nm) observed in AFM measurements as shown in Figure 8.1(a) and (b), respectively. A sketch of the fabricated Hall bar device with the pin configuration is shown in Figure 8.1(c), and the measurements are as labeled in this figure. Here, the p-Si-substrate layer served as a back-gate to control carrier density in the
graphene layer, and the back gate voltage sweep range was limited to \(-50 \leq V_G \leq +50\) V. Under ambient conditions, the gate voltage dependence of the diagonal resistance, \(R_{xx}\), on a freshly prepared graphene Hall bar sample at the room temperature exhibited strong hysteretic behavior. The observed behavior of the sample is attributed to dipolar adsorbates such as water, Oxygen, and poly methyl methacrylate (PMMA), which act as charge carrier traps [93]. The charge carrier trapping by water is a dynamic process that depends on the sweep conditions of the back gate voltage [151] consistent with our measurements.

We stored the freshly prepared sample under medium vacuum conditions at room temperature overnight to remove such trapped charge carriers. Such storage reduced the hysteretic behavior significantly, and the charge neutrality point fell within the swept \(V_G\) span. We applied electrical stressing voltages to the samples under the vacuum to introduce the second charge neutrality point as described in Figure 8.1(d). The curve ① in panel Figure 8.1(d) shows the \(R_{xx1}\) as a function of gate voltage at the ambient conditions before any treatment. After vacuuming the sample space, the first neutrality point appeared around \(+33\) V at the gate voltage as shown in the curve ② in panel Figure 8.1(d). The curve ③ depicts the \(R_{xx1}\) with gate voltage after anneal with 1 mA direct current through the sample for 30 min at room temperature in the vacuum. The first charge neutrality point was shifted to \(+23\) V, and there was a sign of the second neutrality point at the high end of the gate voltage. Then the sample was annealed with 1 mA for 60 min with back gate voltage \(-40\) V. The first neutrality point was shifted close to \(+8\) V, and the second one appeared around \(+45\) V in gate voltage as shown in curve ④ in panel Figure 8.1(d). Then, the sample was cooled down to \(~15\) K with the gate voltage value \(-30\) V and without an annealing current. The formation of double neutrality points in a p-n junction device and electrical data illustrating double neutrality points are shown in curve ⑤ in Figure 8.1(d).
Here, we present the results with double neutrality points at the back gate voltages close to -4 V and +17 V, see Figure 8.2, which are labelled as $V_{D1}$ and $V_{D2}$ respectively. Here, $V_{D2}$ is the second neutrality point that appeared after the electrical stress. We confirmed that the double neutrality point appeared only in the longitudinal voltage measurements from $V_{xx1}$ and $V_{xx2}$ as shown in Figure 8.1(c), as diagonal resistance measurements across other contacts further down the channel showed only one neutrality point within the sweep range of $V_G$. We noticed that the second charge neutrality point disappeared from the swept $V_G$ range when the device was exposed to ambient conditions for a couple of hours, but it could be reestablished easily by reapplying the electrical stressing voltages in vacuum condition. These repeatable results confirmed that the applied electrical stressing voltage did not irreversibly damage the device.

8.3 Results and Discussion

The double neutrality points observed in the $R_{xx}$ vs. the gate voltage traces are a signature of the formation of a p-n junction in the graphene channel. The schematic in Figure 8.1(e) depicts the p-n junction formation in the graphene channel as a function of the gate voltage. Two cones represent the graphene dispersion in two regions, and the yellow plane represents the Fermi level of the system. When the gate voltage is equal to -50 V, the Fermi level lies below both neutrality points as the entire channel is p-type, as in Figure 8.2(a). When the gate voltage is swept from -50 V towards zero, the Fermi level is shifted up and crosses the first neutrality point at $V_G \approx -4$ V, as $R_{xx1}$ (see Figure 8.1(e)) increases and crosses its first maximum with the gate voltage increment. A further increase in $V_G$ progressively moves up the Fermi level, and when $V_G \approx +17$V the Fermi level crosses the second neutrality point and this reflected as the second peak in $R_{xx1}$, see Figure 8.1(e). When the gate voltage is $-4 < V_G < +17$ V, the sample includes a p-n junction. For $V_G > 17$V, the entire channel is n-type as the Fermi level lies
in the conduction band throughout the channel. Figure 8.1(e) shows the $R_{xx}$ vs. $V_G$ at zero magnetic field ($B=0$); this plot shows maxima at $V_G \approx -4V$ and $V_G \approx +17V$. The application of a small perpendicular magnetic field, within the range $-0.2 < B < +0.2$ T produced a small shift in the peak locations $\Delta \nu$, where $\Delta \nu$ is the difference of the back gate voltage at peak resistances. This $\Delta \nu$ is associated with a trapped charge concentration $n_t = \alpha \Delta \nu$ with $\alpha = 7.2 \times 10^{10}$ cm$^{-2}$V$^{-1}$.

To extract further information, we applied Equation (8.1), the extended fitting equation for double neutrality points [165], which assumes the existence of multiple parallel channels contributing to the current, with each channel characterized by its own value of the neutrality point.

$$R_{xx} = \frac{(1 - \lambda)}{e\mu_1 \sqrt{n^2_{01} + n^2_1}} + \frac{\lambda}{e\mu_2 \sqrt{n^2_{02} + n^2_2}} + r_s$$

(8.1)

Here, $(\lambda = 0.63)$ is a parameter related to a geometrical factor in the graphene channel, and that depends on the physical locations of the neutrality points. Also, $r_s$ is the resistance due to short range scatters, assumed to be gate independent [165]. We found that the $r_s$ can be neglected from the fittings of our results. The $\mu_1$ and $\mu_2$ parameters are the charge carrier mobilities and $n_1$ and $n_2$ are residual charge concentrations in the unperturbed- and perturbed- regions, respectively, in the graphene channel. The electrostatic potentials determine the carrier densities in these two regions. Data fits indicated that the mobilities are $6148$ cm$^2$/V s and $1388$ cm$^2$/V s in unperturbed and perturbed areas of the graphene, respectively. Also, the extracted values for the residual carrier concentrations are $1.59 \times 10^{11}$ cm$^{-2}$ and $1.11 \times 10^{12}$ cm$^{-2}$ in unperturbed and perturbed regions of the graphene, respectively. As the carrier density increases, the mobility decreases for single layer graphene [166], so the high density of residual carriers in the perturbed region could be responsible for corresponding lower mobility.
Figure 8.2 The longitudinal resistance ($R_{xx}$) and Hall resistances ($R_{xy}$) color maps for a Hall bar sample with double neutrality points, as a function of back gate voltage ($V_G$) and magnetic field ($B$). (a) shows the $R_{xx1}(B,V_G)$. The $R_{xx1}(B,V_G)$ plot exhibits maxima near two distinct charge neutrality points; see also the curve $\mathbb{5}$ in Figure 8.1(d). (b) shows the $R_{xy1}(B,V_G)$ from the unperturbed region (see text) of the graphene channel. (c) shows the $R_{xy2}(B,V_G)$ from the perturbed region (see text) of the graphene channel.

Figure 8.2 shows the color maps of the transport characteristics of the graphene Hall bar sample in a perpendicular magnetic field. Figure 8.2(a) shows the longitudinal resistance ($R_{xx1}$) as a function of magnetic field and gate voltage. The $R_{xx1}$ color map shows maxima at the charge neutrality points. An asymmetry between hole side and electron side of the characteristic curve is observable relative to the first charge neutrality point. The second charge neutrality point in the positive side of the gate voltage may lead to this $R_{xx1}$ asymmetry around the original neutral point. Also, there was no significant difference in longitudinal voltage measurements $V_{xx1}$ and $V_{xx2}$ (see Figure 8.2(c)) from two parallel sides of the Hall bar. Figure 8.2(b) represents the Hall resistance $R_{xy1}$ in the unperturbed region of the graphene channel, which is physically away from the second charge neutrality point. Figure 8.2(c) depicts the Hall resistance $R_{xy2}$ in the perturbed area of the graphene sample, which is physically close to the second neutrality point.
Figure 8.3 The separation between symmetric and asymmetric contributions allows the pure Hall voltage to be extracted from the graphene pn junction. (a) shows the symmetric parts of the longitudinal resistance ($R_{xx1}$) while (d) presents the asymmetric components of the $R_{xx1}$, at different values of the perpendicular magnetic field. (b) and (e) show the symmetric and asymmetric components of Hall resistance of the unperturbed region of the graphene Hall bar device ($R_{xy1}$), respectively. (c) and (f) show the symmetric and asymmetric contributions of the Hall resistance of perturbed area of the graphene Hall bar device ($R_{xy2}$), respectively.

The separation between symmetric and antisymmetric contributions allows the extraction of pure voltages that originate in graphene pn-junction. Figure 8.3 depicts the separation between symmetric and asymmetric contributions of the voltages originated in the graphene pn-junction. The symmetric part of the longitudinal resistance ($R_{xx1}$) did not change due to the normal magnetic field while the asymmetric component of the $R_{xx1}$ changed, as shown in Figure 8.3 (a) and (d), respectively. Panels (b) and (e) show the symmetric and antisymmetric components of Hall resistance from the unperturbed region in the graphene Hall bar device ($R_{xy1}$), respectively. Panels (c) and (f) show the symmetric and antisymmetric contributions of the Hall resistance of perturbed area in the graphene Hall bar device ($R_{xy2}$), respectively. Panels (c) and (f) depict the
symmetric and antisymmetric components of the Hall voltage from the perturbed region of the graphene channel.

Figure 8.4 (a) The Hall voltage in the perturbed region \( (V_{xy}) \) is plotted vs \( B \) with the back gate voltage \( (V_G) \) as the varying parameter. The red traces display a positive slope that increases with \( V_G \), while the blue lines exhibit a negative slope that decreases with \( V_G \). (b) Typical non-monotonic variation in \( \frac{dR_{xy}}{dB} \) vs \( V_G \). The blue spheres show the extracted \( \frac{dR_{xy}}{dB} \) values from the data in the unperturbed region of the graphene channel. The red spheres represent the extracted \( \frac{dR_{xy}}{dB} \) values from the data in the perturbed area of the graphene channel. The derivative \( \frac{dR_{xy}}{dB} \) represents the slope of \( V_{xy} \) vs \( B \) graphs.

Figure 8.4 presents the non-monotonic behavior in the slope of Hall voltages across the perturbed and unperturbed region of the graphene channel. Figure 8.4(a) shows the Hall voltage with positive and negative slopes in the perturbed area of the graphene channel for selected back gate voltage values. Figure 8.4(b) summarizes the slope of Hall resistances \( (dR_{xy}/dB) \) as a function of gate voltage. Blue spheres show the Hall slope variation across the unperturbed region of the graphene channel. From \(-50 \leq V_G \leq +50 \) V, the slope remains positive and
increase in magnitude, then it starts decreasing from positive values to negative values within the gate voltage range \(-5 \leq V_G \leq 0\) V. From \(0 \leq V_G \leq +50\) V, the Hall slope remains negative but decrease in magnitude with increasing gate voltage. In Figure 8.4(b), the red spheres show the Hall slope in the perturbed region of the graphene, which stays positive over the span \(-50 \leq V_G \leq +17\) V, with a small ripple within the span \(-5 \leq V_G \leq 0\) V. From \(+12 \leq V_G \leq +25\) V, slope changes from positive to negative. Above the gate voltage \(+25\) V, the slope remains negative but decreases in magnitude. Here, the crossover band for the unperturbed region is narrower than the band for the perturbed area.

![Figure 8.5](image)

**Figure 8.5** Characteristic curves of the current related sensitivity \(S_I\) of the Hall bar device. (a) The \(S_I\) of the Hall bar device as a function of the gate voltage at a constant magnetic field. Green circles are data, and the solid red curve is the numerical fit. (b) This panel shows that the maximum current sensitivity \(S_{I\text{max}}\) is inversely proportional to residual carrier concentration \(n_0\) in the graphene channel. Blue circles are data, and the dashed red line guides the eye.

The current related Hall sensitivity \((S_I)\) of the device as a function of gate voltage and the magnetic field is shown in Figure 8.5. The current related sensitivity is defined as \(S_{I} = \)
\[ V_{xy}/IB = \rho_{xy}/B \] [167]. Both types of carriers play an important role near the charge neutrality points of graphene, not as in unipolar type conventional semiconductors. Thus, we utilized the ambipolar transport model to describe the experimental data [168]. The current related sensitivity can be written as Equation (8.2).

\[
S_I = \frac{r_H (n_h \mu_h^2 - n_e \mu_e^2)}{e (\mu_h n_h + \mu_e n_e)^2}
\] (8.2)

where \( e \) is the electron charge, \( r_H \) is the Hall coefficient, \( n_h \) and \( \mu_h \) are density and mobility of hole carriers, and \( n_e \) and \( \mu_e \) are density and mobility of electrons. By assuming \( \mu_h = \mu_e \) close to the neutrality point, we can derive Equation (8.3).

\[
S_I = \frac{-r_H}{e} \frac{n_{[V_G]}}{(n_{[V_G]}^2 + n_0^2)}
\] (8.3)

where, \( n_{[V_G]} = \alpha (V_{neutrality} - V_G) \) and \( \alpha = 7.2 \times 10^{10} \text{ cm}^{-2} \text{V}^{-1} \). We utilized Equation (8.3) to fit the experimental data from the unperturbed region in graphene, as shown in Figure 8.5(a). We extract the residual carrier concentration \( n_0 \) from the fits, and \( n_0 \) is inversely proportional to maximum current sensitivity as described in Figure 8.5(b). Here, the maximum current sensitivity is equal to 2635 V/AT, which is higher than conventional Hall sensors build from GaAs/AlGaAs (~1500 V/AT), GaAs (~200 V/AT), and InAs (~500 V/AT) [168, 169].

### 8.4 Summary

We utilized the electrical stressing-voltage method to create p-n junctions in graphene Hall bar devices and examined their transport characteristics. The Hall voltage measurements across the electrical stress-induced neutrality point show magnetic field dependence not observed in the unstressed device. Further, it appears that the extracted mobility for the unperturbed graphene region is higher than the mobility in the perturbed region. We think that a high density
of residual carriers in the perturbed area leads to corresponding lower mobility in that graphene region due to impurity scattering and sample imperfection. The Hall bar device appears highly sensitive to the external magnetic field when it operates as a p-n junction. These results help to understand better the behavior of double neutrality points in graphene field effect transistors, particularly the role of charge carrier characteristics for real-world applications based on graphene p-n junctions.
9 SUMMARY

This dissertation is based on magnetotransport studies of the 2D electron systems (2DES) in GaAs/AlGaAs and graphene, and an investigation of chemical vapor deposition (CVD) techniques for graphene growth. The effect of microwave (MW) radiation on electron temperature was examined by investigating the photo-excited transport at zero magnetic field and in the Shubnikov-de Haas oscillations (SdHOs) regime where the cyclotron frequency $\omega_c$ and the MW angular frequency $\omega$ satisfy $2.3 < \omega_c/\omega \leq 5.2$. The results show small discernible electron heating under modest MW radiation, in agreement with theoretical predictions. The heating effect appears greater at zero field in comparison to the examined SdHOs regime, in line with theoretical predictions. However, the increase in the electron temperature in the zero-field limit is smaller than theoretical predictions mostly because theory assumes no microwave attenuation between source and sample while, in our experiment, the attenuation appears substantial. In future studies, developing an experimental technique to probe the electron temperature in microwave induced magnetoresistance oscillation would be beneficial for understanding the complete picture of electron heating.

Additionally, the activation energy at the odd integer filling factors in GaAs/AlGaAs 2DES was examined using a novel microwave-induced heating technique instead of conventional temperature-dependent measurements. The main advantage of this microwave induced heating technique is controllability with great precision and localized heating on the specimen. We examined four odd filling factors between 5 and 11 by making Arrhenius plots for chosen filling factors and extracting the activation gap under the microwave condition. The results here indicate a linear increase of spin energy gap vs magnetic field. The extracted effective g factors are 6.47 and 2.79 at zero and 66.14 angles, respectively. It is interesting
further examine these odd high filling factors’ activation energies and enhance g-factors as a function of tilt angle in future studies.

We also performed magnetotransport measurements on epitaxial graphene samples, which show large longitudinal resistance values, and examined the Hall resistance values at filling factor two shift away from the quantized values with increments of the magnetic field. This Hall resistance deviation could be realized as a correction to $R_{xy}$ that is linearly proportional to $R_{xx}$. It is very rare incident in epitaxial graphene. Also, examining the spin resonance of graphene under microwave conditions as a future project would be an exciting research work.

This dissertation also focused the research efforts on the growth and techniques of low-pressure CVD graphene. A dual treatment technique was utilized to lower the nucleation density of single crystal graphene grown on copper. Additionally, we discuss an impurity-assisted growth mechanism which governs the growth of single-crystal graphene via isotropic diffusion, producing two-fold, four-fold, and six-fold symmetries in the resulting flakes. In addition, single crystal graphene formed via anisotropic diffusion also appears here, but they do not show the sign of an impurity assisted growth mechanism. Nevertheless, the density and size of the steps are different in each graphene flake, like a fingerprint for each flake. Our study also explains that strain relaxation in two-fold and four-fold symmetry graphene crystals via isotropic diffusion is more complicated than the six-fold symmetric crystals via isotropic diffusion, which reflects in various steps orientations in two-fold and four-fold symmetric flakes. The periodic nanoscale steps observed in graphene crystals are due to possible strain relaxation during the CVD cooling process occurring due to the different thermal expansion coefficients of copper and graphene.
We also examined electrical transport measurements across a graphene p-n junction formed in a single gated field-effect transistor from CVD graphene. Here, an electrical stressing-voltage technique was utilized to create the p-n junction by modifying the electrostatic potential in the SiO$_2$ substrate. The advantage of this technique is electronic modification occurs in the substrate but not in the graphene film. The maximum current sensitivity of this device is equal to 2635 V/AT, which is higher than conventional Hall sensors build from GaAs/AlGaAs, GaAs, and InAs.

Overall, this research work examined the topics in magnetotransport measurements of 2DES in graphene and AlGaAs/ GaAs heterostructures and CVD techniques for graphene. Such a study could benefit many areas in condensed matter physics, such as nanotechnology in general, Graphene field-effect transistors, the design of fast semiconductor devices, microwave sensors and sources, high-speed communication devices, and understanding the fundamentals of physics in the 2DES.
REFERENCES

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APPENDICES

Appendix A: EXPERIMENTAL APPARATUS

Temperature is an exhaustive thermodynamic factor of the system that is correlated with the system's energy. Temperature can be considered as the average energy of a system that can include vibrational, rotational, translational energy of the system. If we can cool down a system to the absolute zero temperature (0 K), all these energy terms vanish except the quantum mechanical zero-point energy.

For our experiments, low temperature conditions are necessary to minimize the thermal noise as much as possible. To achieve low temperatures, we use various types of dilution refrigerators (as described below), which can maintain low temperatures in stable values.

Appendix A.1: Dilution refrigerators and superconducting magnet

Appendix A.1.1: Dilution refrigerators with wet cryogenic system

I used an Oxford instrument liquid $^4$He cryostat with superconducting solenoid magnet (see Figure A.1), which can provide a maximum magnetic field of 14 T at 4.2 K. Since liquid $^4$He is utilized in the system, it is considered as a wet cryogenic system. The temperature of the system can be varied using the variable temperature insert (VTI) from 1.5 K to 400 K. It is essential to measure the temperature at sample space accurately and control that temperature during the measurements. We use various type of temperature sensors such as a silicon diode with a Lake Shore DRC 93 temperature controller for temperatures above 8 K, Allen Bradley resistors for temperatures below 5 K (calibrated by using vapor pressure thermometry), Cernox resistors above 2 K (calibrated by Lake Shore Cryotronics), and RuO sensors for below 2 K (calibrated by using vapor pressure thermometry). The high temperatures are controlled by using
heaters in the dilution refrigerator while temperatures below 4.2 K are controlled by changing the $^4$He vapor pressure of the system.

![Diagram of liquid $^4$He cryostat](image)

*Figure A.1 Sketch diagram of the liquid $^4$He cryostat.*

The $^3$He/$^4$He dilution refrigerator from Leiden Cryogenics was also utilized in our measurements (see Figure A.2). The difference of this system is that it uses a mixture of liquid $^3$He/$^4$He to approach the low temperatures, where $^3$He and $^4$He are two isotopes of Helium. The preliminary parts of this cryogenic system are a gas handling system (GHS) and an insert, which are connected using stainless-steel hoses and cables for the gauges and valves of the insert. The $^3$He/$^4$He mixture is circulated by a turbo molecular pump with a dry rotary pump as a backing pump so there is no oil contamination in the refrigerator. A sorb pump is utilized to pump out the inner vacuum chamber (IVC).
Figure A.2 (a) User interface of gas handling system (GHS) of Leiden cryogenic system. (b) The cryostat of the system with an insert connected to the GHS via stainless-steel hoses and cables.

Appendix A.1.2: Dilution refrigerators with the dry cryogenic system

The Triton dilution refrigerator system from the Oxford instrument is known as the dry cryogenic system since it doesn’t use liquid Helium. It has six preliminary parts, including a 12 Tesla superconducting solenoid magnet, as shown in Figure A.3. The cold part of the system is inside the cryostat. Inside the cryostat is an ultrahigh vacuum to protect the cold parts of the system, and it’s known as outer vacuum chamber (OVC). Also, inner cold parts are covered from radiation shields to prevent unwanted heating. Here the gas mixture is circulated in two main loops at the cooling process, and they are referred to as the Pre-cool loop and the Dilution Unit. A circulating gas mixture in the pre-cool loop helps cool down the system to 10 K or lower. Then a precool loop is evacuated using the turbopump. The gas mixture is circulated in the dilution system to reach the base temperature below 1 K. At this stage, condensing the helium gas mixture into a liquid requires high pressure and low enough temperature. First, the mixture is
compressed using the $^3$He compressor. Then the mixture flows through a series of heat exchangers located in the cryostat to cool the mixture to 4 K. In a traditional dilution system helium gas is condensed by a separate pump at the liquid $^4$He stage, also known as 1K pot. This is not available in this system. Here, the mixture is condensed using a Joule-Thompson stage.

![Figure A.3](image_url) The preliminary parts of the Triton cryo-free dilution refrigerator. (a) cryostat with the pulse-tube cooler connected with the insert. (b) gash handling system and control rack (c) magnet power supply (d) the pump station (e) pulse tube compressor (f) a liquid nitrogen drawer.

**Appendix A.2: low-noise electrical measurements lock-in techniques**

Phase sensitive electrical signal detection is an advanced technique to detect small electrical signals with a noise background. Using lock-in amplifiers in phase sensitive detection of electrical signals allows achieving high signal to noise ratios. Let’s consider the typical
experiment set up with a known frequency of supplied voltage/ current and the same frequency
used as a reference signal in lock-in amplifiers to detect the electrical signal. The reference signal
can be written as,

\[ V_{\text{ref}} = V_r \sin(\omega_r t + \varphi_r), \quad (A.1) \]

where \( V_r, \omega_r \) and \( \varphi_r \) are amplitude, frequency and the phase of the reference signal,
respectively. The measured electrical signal can be expressed in a similar form,

\[ V_s = V_s \sin(\omega_s t + \varphi_s), \quad (A.2) \]

where \( V_s, \omega_s \) and \( \varphi_s \) are amplitude, frequency and the phase of the measured signal,
respectively. The lock-in amplifier amplifies the measured signal and then multiplies that signal
with the reference signal using phase sensitive detector. The phase sensitive detector signal,

\[ V_{\text{PSD}} = \frac{1}{2} V_s V_r \left( \cos[(\omega_s - \omega_r) t + (\varphi_s - \varphi_r)] - \cos[(\omega_s + \omega_r) t + (\varphi_s + \varphi_r)] \right) \quad (A.3) \]

At typical measurements, \( \omega_s = \omega_r = \omega_0 \), then Eq. (A.3) can be simplified as,

\[ V_{\text{PSD}} = \frac{1}{2} V_s V_r \left( \cos[(\varphi_s - \varphi_r)] - \cos[2\omega_0 t + \varphi_s + \varphi_r] \right) \quad (A.4) \]

As the final step, the output of the phase sensitive detector flows through the low pass filter to
remove the higher frequencies and harmonics of the reference signal.

Low frequency lock-in techniques have been utilized in our experimental setup to
measure the electrical signals from the samples, as shown in Figure A.4.
Figure A.4 schematic diagram of electrical connections for typical measurements. lock-in amplifier 1 measure the Hall voltage while supplying AC voltage with specific frequency to the sample through the variable resistors. The same signal uses as the input reference for lock-in amplifier 2, which measures the longitudinal voltage of the sample. The purpose of having the variable resistors is that set the known current through the sample. Note: The display of lock-in amplifiers don’t show the real values in this diagram.

The oscillator output of a lock-in 1 amplifier can be used as the constant voltage source. The transistor-transistor-logic (TTL) output of the same lock-in can be utilized as the reference signal for the other lock-in amplifiers.
Appendix B: DESIGNING AND MACHINING ROTATABLE SAMPLE PLATFORM

As a subproject of this dissertation work, I designed and machined parts for a sample rotation platform based on a worm-gear mechanism driven by an external stepper motor to perform rotations. It was assembled on the puck that is located inside the bore of a solenoid superconducting magnet in our Triton cryofree dilution refrigerator. The stepper motor is at room temperature, outside the cryostat, and it is connected with the worm gear system at base temperature through a vacuum feedthrough and stainless-steel capillary tube. Having such a rotatable system allows us to study the angle-dependent properties of quantum transport in semiconductor nanodevices at milli-Kelvin temperatures in total magnetic fields up to 12 T. This system provides continuous in situ rotation of a device through roughly 100° to perpendicular to the device plane. It also has an integrated angle sensor to measure the angle. The sample holder consists of two buckets of copper and polyetherimide, as described in Figure A.5.

Figure A.6 shows the copper bucket assembled in the sample puck.
Figure A.5 Technical drawing of the copper bucket and polyetherimide bucket in the rotatable sample holder.

**DESCRIPTION** Rotatable sample holder. This drawing belongs to Nanoscience, Low Temperature and High Magnetic Field Laboratory, PI Prof. Naval G. Muni

**MATERIAL** Copper and Polyetherimide

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Figure A.6 Sample rotation platform based on a worm-gear mechanism. (a) shows that the shiny copper bucket is assembled to the worm gear system at the center of the puck. (b) depicts the complete rotational system with a sample that is connected with flexible wire strips for the measurements. There is a semi-rigid coax loop around the sample and an aluminum cone above the sample to propagate the microwaves to the sample space.
Appendix C: GRAPHENE SAMPLE PREPARATION TECHNIQUE

Appendix C.1: PDMS assisted hybrid transferring technique – CVD single crystal graphene

Figure A.7 Transferred single crystal graphene using the hybrid transferring technique. (a) single graphene flake on the PDMS at the intermediate step of the transferring. (b) transferred single crystal graphene on target Au/Cr contact on Si substrate. (c) and (d) show the transferred graphene flakes on to bare Si surface. Note that we can transfer complete graphene flakes with a highly successive rate using this technique.
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