Quantum Hall Effect and Electromechanics in Graphene

A Thesis

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by

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DECLARATION

This thesis is a presentation of my original research work. Wherever contributions of others are involved, every effort is made to indicate this clearly, with due reference to the literature, and acknowledgement of collaborative research and discussions.

The work was done under the guidance of Professor Mandar M. Deshmukh at the Tata Institute of Fundamental Research, Mumbai.

Vibhor Singh

[Candidate’s name and signature]

In my capacity as supervisor of the candidate’s thesis, I certify that the above statements are true to the best of my knowledge.

Professor Mandar M. Deshmukh

[Guide’s name and signature]

Date:
To My Parents
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Synopsis

Abstract

This synopsis presents a summary of the experiments in the quantum Hall regime, studying graphene’s electron transport in a field effect transistor (FET) geometry and electromechanical properties in a resonator geometry. Recently, the quantum Hall effect (QHE) has been observed in graphene [1, 2] and studied extensively [3]. We studied the breakdown of the quantum Hall state in graphene with two-fold motivation. Firstly, in graphene the cyclotron gaps are unequally spaced and are much larger than that of a 2-dimensional electron gas (2DEG) at a given magnetic field, which make it possible to observe the QHE even at room temperature [4]. Therefore, QHE in graphene could possibly have an entirely different mechanism for breakdown from its counterparts in 2DEG. Secondly, understanding the breakdown mechanisms can also be useful for room temperature metrological resistance standards. In this study, we measured critical currents for dissipationless transport in the vicinity of integer filling factors ($\nu$). It shows a correlation with the cyclotron gaps. It further sheds light on the breakdown mechanism, which can be understood in terms of inter Landau level (LL) scattering resulting from mixing of wave functions of different LLs. We further studied the effect of transverse electric field and observed an invariant point in the measured transverse conductance between the $\nu = 2$ to $\nu = 6$ plateau transition for different bias currents. We explain this invariant point based on a current injection model [5].

In addition to graphene’s electronic properties, it also has remarkable mechanical properties including a high Young’s modulus of rigidity of $\sim 1$ TPa [6, 7, 8, 9, 10]. The large surface-to-mass ratio of graphene offers a distinct advantage over other
nanostructures for applications like ultra low mass sensing, charge sensing and for the study of coupling between charge transport and mechanical motion. In order to better understand the potential of graphene based electrically actuated and detected resonators [9] and the challenges in realizing strain-engineered graphene devices [11, 12], we experimentally measured the coefficient of thermal expansion of graphene ($\alpha_{\text{graphene}}(T)$) as a function of temperature. Our measurements find $\alpha_{\text{graphene}}(T)$ is negative for $30 \text{ K} < T < 300 \text{ K}$ and larger in magnitude than its theoretically predicted value [13]. We also probed the dispersion, or the tunability, of mechanical modes using the DC gate voltage at low temperatures and find that the thermal expansion of graphene, built-in tension and added mass play an important role in changing the extent of tunability of the resonators [14] and the resonant frequency.

As mentioned earlier, graphene nanoelectromechanical systems (NEMS) offer a platform to study the coupling in charge and mechanical degrees of freedom. We studied the electromechanics of graphene resonator in ultra clean devices in the quantum Hall regime at low temperature. We measured the two probe resistance of these devices while mechanically perturbing it at different values of the magnetic field. The system shows change in resistance upon mechanical perturbation. These results can be explained by the rectification of carrier density with mechanical vibrations using a simple model of parallel plate capacitor. Further, we have looked at the complementary question: how does the change in electronic properties with magnetic field affect the mechanical motion. We measured the resonant frequency and quality factor of these resonators and find shifts in resonant frequency and enhancement in the damping with magnetic field. We qualitatively argue that these observations have their origin in nonlinear damping [15] and magnetization which could be related to the electronic properties of the system [16].

In graphene FET with local top gates, independent control over the local charge type and electric field adds a new dimension to study electron transport [17]. Effects like Klein tunneling [18, 19], Andreev reflection [20], collapse of Landau levels [21], Veselago lens [22] and collimation of electrons [23] has been observed. The top gate geometry has also been utilized in controlling the edge channels in the quantum Hall limit. With the control over local carrier density and global carrier density $p-n$ junctions [19, 24] and $p-n-p$ junctions [25] have been studied which show integer and fractional quantized plateaus in the conductance. We studied the effect of local mod-
ulation of charge density and carrier type in graphene FET in dual top gate geometry. The top gates allow us to control the charge density and type independently at two localized regions in graphene leading to the formation of multiple \( p-n \) junctions. By performing electron transport measurements in the quantum Hall regime, we observed various integer and fractionally quantized conductance plateaus. These results can be explained by mixing and partitioning of the edge channels at the junctions. Our analysis on these two probe devices indicates that device aspect ratio and disorder play an important role in determining the quantization of the conductance plateaus.

In this dissertation, we have investigated

- Non-equilibrium breakdown of quantum-Hall state in graphene [26]
- Thermal expansion of graphene and modal dispersion at low temperature using graphene NEMS resonators [27]
- Graphene electromechanics in the quantum Hall regime [28]
- Dual top gated graphene transistor in the quantum Hall regime [29]

We will begin by providing an introduction to the thesis in Chapter 1. A general description of graphene, its electrical and mechanical properties and various other concepts related to the experiment will be given in Chapter 2. The fabrication details like mechanical exfoliation, electron beam lithography techniques and wet chemical etching will be presented in Chapter 3. This will also include electrical measurement schemes like two source heterodyne mixing technique and frequency modulation technique. Chapter 4 will describe the breakdown of quantum Hall state in graphene. Chapter 5 will describe the measurements on graphene NEMS covering the measurement of coefficient of thermal expansion and modal dispersion at low temperatures. Electromechanics of graphene NEMS in quantum Hall regime will be described in Chapter 6. Experiments on dual top gated graphene transistor in the quantum Hall limit will be given in Chapter 7. Chapter 8 will present the summary and outlook of the work.
Introduction

Graphene is one atom thick two-dimensional sheet of carbon atoms arranged in a honeycomb lattice [3]. It has shown exciting electronic properties and perhaps is the most studied material in condensed matter physics in recent years. In this structure, \( sp^2 \) hybridization of carbon atoms gives rise to 3 in-plane \( \sigma \)-bonds with the \( \pi \)-orbital pointing out of the plane. While \( \pi \)-orbital give rise to exciting electronic properties, the in-plane \( \sigma \) bonds provide extraordinary stiffness to graphene. Its in-plane Young’s modulus of rigidity is \( \sim 1 \) TPa while being the thinnest known material, which makes it a very promising material for nanomechanical resonators.

To understand the basic electronic properties of graphene, we start with its band-structure. As shown in Fig. 1(a), the honeycomb lattice of graphene can be represented as a Bravais lattice with a two carbon atom basis on triangular lattice. Within tight binding calculations, the band structure of graphene close to Fermi energy is linear \( (E = \pm c \hbar |k|, \text{where } c \approx 10^6 \text{ ms}^{-1} \text{ is the Fermi velocity}) \). Fig. 1(b) shows the band structure of graphene close to Fermi level. Conduction band and valence band meet at two distinct points (Fermi points, also K and K’ point) in reciprocal space forming two distinct cones close to Fermi level, commonly referred to as Dirac cones. For charge neutral graphene the Fermi level lies exactly at the meeting points of conduction and valence bands. It has zero band gap with vanishing density of states at the Fermi point. Therefore it is possible to tune the Fermi level (or carrier density) of graphene continuously from valence band to conduction band by electrical or chemical doping. The electrical doping is achieved by making graphene devices in FET geometry, where by applying a gate voltage charges can be induced on the graphene sheet.

The most common technique to make graphene is the mechanical exfoliation [30], which essentially is pulling single layer graphene sheets from bulk graphite using scotch-tape. It is then transferred to a degenerately doped Si substrate coated with 300 nm of SiO\(_2\), which gives the best contrast to see monolayer graphene in the visible wavelength range [31] and it becomes possible to see atomically thin sheets using simple tools like optical microscope.
Figure 1: a) Lattice structure of graphene showing the unit cell with two carbon atom basis. b) Band structure of graphene near the Fermi level showing linearly dispersing conduction and valance band meeting at a point (also known as Dirac point) at Fermi level .

Non-equilibrium breakdown of quantum-Hall state in graphene

The quantum Hall effect (QHE) has been studied extensively in two dimensional (2D) systems [32] and its equilibrium electron-transport properties are understood to a large extent. QHE is observed in 2D electronic systems subjected to high magnetic fields perpendicular to its plane at low temperatures. In presence of magnetic field, the otherwise continuous density of states spectrum splits into discrete energy levels called Landau levels (LLs). When Fermi energy lies between two LLs, the longitudinal resistance \( R_{xx} \) vanishes leading to a dissipationless transport and transverse resistance \( R_{xy} \) becomes quantized in units of fundamental constants, given by \( \frac{h}{ie^2} \) ( where \( i \) is an integer). This effect is called integer quantum Hall effect (IQHE). After its observation in graphene [1, 2], it has been studied extensively [3]. In a magnetic field perpendicular to its plane, the energy spectrum of graphene splits into unequally spaced LLs and is given by \( E_n = sgn(n)\sqrt{(2\hbar c^2 e B |n|)} \), where \( n \) is the LL index \( (n = 0,1,2...) \). As mentioned earlier, when the Fermi level lies between two LLs, dissipationless transport occurs in the system \( R_{xx} = 0 \) and \( R_{xy} \) gets quantized to \( \frac{h}{ie^2} \), where \( \nu \) is the integer filling factor and related to LL index by \( \nu = sgn(n)(4|n|+2) \) [1, 2].
Figure 2: Plot of the longitudinal resistance ($R_{xx}$) and transverse resistance ($R_{xy}$) for a monolayer graphene device at $T = 300$ mK and $B = 9$ T. The inset shows an optical microscope image. The scale bar corresponds to 6 $\mu$m. Probes $S$ and $D$ were used to current bias the device. By using a lock-in technique, probe pairs $V_1 - V_2$ and $V_1 - V_3$ were used to measure $R_{xx}$ and $R_{xy}$ respectively.

We probed the non-equilibrium breakdown of the quantum Hall state in monolayer graphene by injecting a high current density ($\sim 1$ A/m). In particular, we measured the injected critical current which leads to the breakdown of the dissipationless transport ($R_{xx} \neq 0$) for different integer filling factors ($\nu = sgn(n)(4|n| + 2)$). To study this, we fabricated monolayer graphene devices in Hall bar geometry following the standard clean-room techniques of electron beam lithography. After fabrication, we cooled down these devices using a He-3 insert down to 300 mK in a cryostat equipped with 10 Tesla magnetic field. Inset of Fig. 2 shows optical microscope image of the device fabricated following the electron beam lithography processes in Hall bar geometry. Fig. 2 shows the measurement of $R_{xx}$ and $R_{xy}$ with the variation of gate voltage at $T = 300$ mK and $B = 9$ Tesla. Vanishing $R_{xx}$ and corresponding quantized plateaus in $R_{xy}$ for different integer filling factors ($\nu = 6, 2, -2, -6, -10$), which are unique to monolayer graphene, can be clearly seen.

To probe the breakdown of quantum Hall state, we biased the source-drain probes of our device with DC current ($I_{DC}^{SD}$) along with a small AC current (50 nA) in the minima of $R_{xx}$ corresponding to filling factors $\nu = \pm 2, \pm 6$ and $-10$ at fixed magnetic field. We measured $R_{xx}$ while keeping AC current fixed and varying $I_{DC}^{SD}$.
Figure 3: Plot of critical current ($I_{SD}^{crit}$) for different filling factors at $T = 300$ mK and $B = 9$ T. The inset shows the plot of Hall voltage developed at breakdown ($V_{Hall}$) and the cyclotron gaps ($\Delta E_\nu$) plotted on the right axis as a function of $\nu$.

as a function of $V_g$ in the vicinity of integer $\nu$. In order to interpret the breakdown from the measured experimental data we define a critical current ($I_{SD}^{crit}$) as the linearly extrapolated value of $I_{SD}^{DC}$ at zero dissipation [33]. Fig. 3 shows the measured $I_{SD}^{crit}$ for different filling factors indicating that $I_{SD}^{crit}$ decreases with $|\nu|$. The inset shows the plot of two relevant quantities, Hall voltage, $V_{Hall} = I_{SD}^{crit} \times \frac{h}{e^2}$, and $\Delta E_\nu$ as a function of $\nu$. There is a correlation between $V_{Hall}$ and $\Delta E_\nu$, which can be explained by considering inter-LL scattering. The origin of the inter-LL scattering is likely to be the strong local electric field that mixes the electron and hole wavefunctions [5, 34, 35] providing a finite rate for inelastic transitions. The presence of a charge inhomogeneity [36] leads to a strong local electric field and thus can reduce the threshold for the breakdown due to inter-LL scattering. We also looked at the plateau to plateau transition in transverse conductance ($\sigma_{xy}$). We measured $\sigma_{xy}$ for $\nu = 2$ to $\nu = 6$ transition for different bias currents and found an invariant point in transverse conductance at $\nu = 4$ for different bias currents. This is also accompanied with a small suppression in $R_{xx}$. We speculate the current invariant point at $\nu = 4$ and suppression of $R_{xx}$ at the same time as a precursor of Zeeman splitting. To understand our data quantitatively, we carried out calculations based on the injection model of QHE in graphene [5].

To summarize, we find that the dissipationless QH state can be suppressed due to a high current density, and the corresponding critical current decreases with $|\nu|$. We also see a current invariant point in the plateau to plateau transition and suppression
in longitudinal resistance at higher currents, which can possibly be a sign of lifting of spin-degeneracy.

**Thermal expansion of graphene and modal dispersion at low temperature using graphene NEMS resonators**

NEMS (nanoelectromechanical systems) devices using nanostructures like carbon nanotubes \[37, 38, 39, 40, 41, 42, 43\], nanowires \[44, 45\] and bulk micromachined structures \[46, 47, 48\] offer promise of new applications and allow us to probe fundamental properties at the nanoscale. NEMS \[49\] based devices are ideal platforms to harness the unique mechanical properties of graphene. Electromechanical measurements with graphene resonators \[8, 9\] suggest that with the improvement of quality factor \((Q)\), graphene based NEMS devices have the potential to be very sensitive detectors of mass and charge. Additionally, the sensitivity of graphene to chemical specific processes \[50\] offers the possibility of integrated mass and chemical detection. We used suspended graphene electromechanical resonators to study the variation of resonant frequency as a function of temperature. Measuring the change in frequency resulting from a change in tension, from 300 K to 30 K, allows us to extract information about the thermal expansion of monolayer graphene as a function of temperature. We also studied the dispersion, the variation of resonant frequency with DC gate voltage, of the electromechanical modes and found considerable tunability of resonant frequency. We quantitatively explained the tunability of resonant frequency with gate voltage.

**Device fabrication and measurement scheme**

In the first step of fabrication, on-substrate devices as described before were made following the standard electron beam lithography procedures. To release graphene, we use wet chemical etching to remove the part of SiO\(_2\) substrate. It is then followed by critical point drying to prevent collapse of the device due to surface tension.
scanning electron microscope (SEM) image of a suspended graphene device is shown in Fig. 4(a).

The electrical actuation and detection is done by using the suspended graphene device as heterodyne-mixer [9, 37, 42] as shown Fig. 4(a) superimposed on the SEM image of the device. The essence of this technique is that the radio frequency (RF) signal of frequency $\omega$ at back gate, modulates the conductance ($G$) of the graphene sheet at $\omega$. The other RF signal of frequency $\omega + \Delta \omega$ at source end leads to a mixing of these two signals. The downmixed signal, also known as mixing current, ($I_{\text{mix}}(\Delta \omega)$) at frequency $\Delta \omega$ can be measured at the drain end. As one sweeps the driving RF signal across the resonant frequency of the resonator, the changes in $G(\omega)$ becomes larger and $I_{\text{mix}}$ shows a change over the background current. Fig. 4(b) shows such a
measurement of mixing current with driving frequency of RF signal applied at gate. The sharp change in $I_{\text{mix}}$ at 64.3 MHz signifies the mechanical resonance of the device.

Modal dispersion of graphene NEMS resonators and probing thermal expansion of suspended graphene

The resonant modes ($f_0$) in these resonators can be described by the modal of a 2-dimensional sheet of length ($L$) under tension ($T$) clamped at the two opposite ends i.e., $f_0 = (1/2L)\sqrt{T/\mu}$, where $\mu$ is the mass per unit length. Due to the electrostatic attraction between the flake and the back gate, tension in these devices becomes a function of the DC gate voltage ($V_{g}^{DC}$). Therefore, we can write the resonant frequency ($f_0$) as,

$$f_0(V_{g}^{DC}) = \frac{1}{2L} \sqrt{\frac{\Gamma_0(T), V_{g}^{DC}}{\rho tw}}.$$  \hspace{1cm} (1)

where $L$ is the length of the membrane, $w$ is the width, $t$ is the thickness, $\rho$ is the mass density, $\Gamma_0(T)$ is the in-built tension and $\Gamma$ is the tension at a given temperature $T$ and $V_{g}^{DC}$. By taking into account the electrostatic interaction due to $V_{g}^{DC}$, we can completely explain the change of resonant frequency with gate voltage. This further allows us to estimate the $\Gamma_0$ and the modal mass of the flake independently (the presence of residues on graphene flake during fabrication can lead to a different mass than that of pristine graphene). In our analysis we also incorporate the terms accounting for the softening of spring constant due to the electric field gradient. These terms compete with gate voltage induced tension in the flake and becomes more important at low temperature or if the added mass is large. With this model, we have been able to explain modal dispersion completely at all temperatures.

The knowledge of $\Gamma_0$ can be very useful in probing the thermal expansion of graphene. We next consider how the resonant frequency ($f_0$) evolves as a function of the temperature. Fig. 5(a) shows an evolution of a mode as a function of temperature at $V_{g}^{DC} = 15$ V. The resonant frequency increases as the device is cooled from room temperature. The frequency shift can be understood by taking into account the contribution of various strains as the device is cooled below room temperature. Three main contributions to the strain in graphene arise from the expansion/contraction of
Figure 5: a) Plot showing the evolution of the resonant frequency of a mode as a function of temperature for $V_{g}^{DC} = 15 \text{ V}$. Inset shows the schematic of all the strains external to the suspended graphene membrane as the device is cooled below 300 K. b) The plot of expansion coefficient of graphene as a function of temperature. Data from two different devices together with theoretical prediction of N. Mounet et al. [13]. The shaded area represents the errors estimated from the uncertainty of the length of the flake, width of the electrode and Young’s modulus of graphene.
the gold electrodes, graphene and the substrate. At any temperature the net strain in the graphene can be written as the algebraic sum of all these strains. Therefore by taking into account the expansion coefficient of gold and substrate, from the measurement of frequency shift with temperature, we calculated $\alpha_{\text{graphene}}$ as a function of temperature. Fig. 5(b) shows the result of calculating $\alpha_{\text{graphene}}$ for two devices using this analysis and comparison with the theoretical calculation for $\alpha_{\text{graphene}}$ by N. Mounet et al. [13]. We find that $\alpha_{\text{graphene}}$ is negative and its magnitude decreases with temperature for $T < 300$ K and its value at room temperature is $\sim -7 \times 10^{-6}$ K$^{-1}$.

Thus, we explained the modal dispersion of these resonators at all temperatures and using such an analysis we further probed the thermal expansion of suspended graphene. The knowledge of $\alpha_{\text{graphene}}$ is essential for the fabrication of the devices intended for strain engineering applications [11, 12].

Graphene electromechanics in the quantum Hall regime

Recent experiments probing the coupling between charge transport and mechanical motion in NEMS [39, 40] have shown that they do influence each other. The electronic transport can give rise to damping in the mechanical motion and resonant frequency to shift. On the other hand, the DC transport can also lead to mechanical oscillations in the system. Motivated by these, we looked at the electromechanics of graphene resonators in quantum Hall regime in very clean samples. We measured the two probe resistance of these devices while mechanically perturbing it for different values of the magnetic field. Further, we looked at how the change in electronic properties with magnetic field affect the mechanical motion. We measured the resonant frequency and quality factor of these resonators with magnetic field.

The fabrication process of these devices is same as described in the earlier experiment probing the thermal expansion of graphene. In order to achieve better charge carrier mobility, we remove any residue adhered to the flake during fabrication by injecting a large current density ($\sim 10^3$ A/m) at low temperature [51, 52, 53]. The large heat dissipation in the flake leads to the evaporation of the residue from the flake and hence improves the mobility. Fig. 6(a) shows the typical measurement of the resistance with back gate voltage of such a device at $T = 5$ K after cleaning it.
The sharp peak in resistance at zero gate voltage and large mobility of charge carrier which exceeds 150,000 cm²V⁻¹s⁻¹, are measures of the high quality of the sample. For the mechanical actuation and detection of the resonator, we have used previously described two-source heterodyne mixing technique and frequency modulation (FM) technique [54]. In both the techniques, the detection of the mechanical motion relies on the finite transconductance \( \frac{dG}{dV_{DC}} \). Both the techniques have characteristic equations which can be used for the estimation of the resonator parameter like amplitude of vibration \( (a) \), resonant frequency \( (f_0) \) and quality factor \( (Q) \). Fig. 6(c) shows a measurement of the mechanical mode of the device using two source heterodyne mixing technique. The resonant frequency of this mode is very close to 3 MHz, which can be seen from the sharp change in the mixing current.

**Graphene electromechanics affecting QHE**

In order to probe the change in electrical properties with mechanical motion, we record the two probe resistance of the device, while an RF signal at the gate sets the
driving force on the flake. A resistance measurement can now be done continuously, while sweeping the driving frequency of the RF source around resonant frequency (which can be first measured separately by heterodyne mixing technique or by FM technique) at the gate for various values of the magnetic field. We define a quantity “change in resistance” by subtracting the resistance value of the device away from the resonance frequency for each magnetic field ($\Delta R = R - R_0$). Fig. 6(d) shows the $\Delta R$ as a function of driving frequency and magnetic field. Following observations can be made a) $\Delta R$ is zero in the plateau region, b) $\Delta R$ can be of both signs, it is negative on one side of the plateau and positive on the other side, c) for low values of magnetic field, $\Delta R$ oscillates with field. To understand this, we model this geometry like an infinite parallel plate capacitor, in which one plate (the substrate) remains fixed and the other plate (graphene flake) moves with large amplitude at resonant frequency. A constant potential difference between the two plates leads to charge density ($n$) oscillations upon mechanical vibrations. The nonlinear dependence of $n$ on distance (between the flake and gate plate), leads to the rectification of $n$ over multiple oscillations, which modifies the DC resistance. Based on this picture, one can derive the expression for $\Delta R$ as, $\Delta R = \frac{1}{2} \alpha(B)V_{DC}^2 \left( \frac{a}{z_0} \right)^2 + \frac{1}{4} \beta(B)(V_{DC}^2)^2 \left( \frac{a}{z_0} \right)^2$, where $\alpha(B) = \frac{dR}{dV_{DC}^2}$, $\beta(B) = \frac{d^2R}{dV_{DC}^2}$, $a$ is amplitude of vibration and $z_0$ is the gap between the flake and the substrate. Qualitatively, it is easy to see that above expression explains the $\Delta R$ data quite well. The second term involving the second derivative (curvature term) dominates in the above expression. At the beginning of the quantum Hall plateau in $R$, the curvature term will be negative giving rise to negative resistance change at the resonant frequency. On the other hand, after the plateau, the curvature term gives a positive sign, and hence giving a positive resistance change. However, this does not explain all the observations made in the experiment. We also see signs of interference between the terms setting the potential of the flake i.e. mechanical motion and RF driving signal. Since the rectification signal dominates, it is difficult to differentiate among the other contributions like, changes due to strain at the deformation of the flake and resonant transmission of electrons between the edge channels [55].
QHE affecting graphene electromechanics

In Fig. 7(a), dispersion of two modes with magnetic field is shown. It is quite evident from the colorscale plot that the two modes disperse differently with magnetic field. In Fig. 7(b), the non-monotonic resonant frequency shift \( f_0(B) - f_0(0) \) with magnetic field can be seen for two modes (extracting from the measurement shown in Fig. 7(a) by fitting to the characteristic equation). For the upper mode, the frequency shift increases with magnetic field until the beginning of the \( \nu = 2 \) plateau. At \( \nu = 2 \) plateau, the detection scheme fails \( \frac{dG}{dV_{DC}} \approx 0 \) and the estimation of \( f_0 \) and \( Q \) become difficult. After the \( \nu = 2 \) plateau, as \( B \) is increased further, the frequency shift starts dropping slowly. Similar frequency shift can be seen for lower frequency modes, though they are much small compared to the one for higher frequency mode. The \( Q \) reflect similar trend for the two modes, for the high frequency mode the frequency shift is large and it shows larger change in \( Q \), whereas for the lower frequency mode, the frequency shift is small and it shows a smaller change or no change in the \( Q \).

In order to understand this behavior, we note that frequency shifts due to linear damping (velocity term in the force equation) are nominally small \( -\frac{f}{8Q^2} \approx -10Hz \).
However, we see much larger and positive frequency shift. It strongly suggests that there are other damping mechanisms which come into play as we increase magnetic field. Recently, it has been shown that for graphene and carbon nanotube, nonlinear damping mechanisms can lead to larger frequency shift with driving amplitude [15]. We also observed such nonlinear damping in our devices. In order to explain the frequency shift with magnetic field, we note that as we increase the magnetic field, the device resistance monotonically increases (from $\approx 1 \, \text{K}\Omega$ to $\approx 18 \, \text{K}\Omega$). This can lead to the increased driving force and hence larger amplitude of vibration. Therefore, amplitude dependent nonlinear damping mechanism can cause the frequency to shift. This explains the frequency shift and larger damping (drop in the $Q$) at low magnetic fields ($B < 2 \, \text{T}$). However, at larger magnetic field ($B > 6 \, \text{T}$), we observe that frequency shift starts to drop whereas damping remains more or less constant. To understand this, we calculate the magnetization of graphene in quantum Hall regime. The parallel component of the magnetization contributes to the total energy of resonator [16] and therefore it can modify the spring constant of the resonator resulting in frequency shifts observed at higher fields.

**Dual top gated graphene transistor in the quantum Hall regime**

In the quantum Hall regime, the electron transport mainly occurs along the physical edge of the sample through the edge channels and the bulk current remains nominally small. With the help of the top gates, these edge channels can be reflected and mixed. Since graphene has zero band gap, therefore it is also possible to form $p-n$ junctions at the interfaces of the top gates. Such $p-n$ [19, 24] and $p-n-p$ junctions [25] show integer and fractionally quantized conductance plateaus. In this experiment, we studied electron transport in a graphene multiple lateral heterojunction device with charge density distribution of the type $q-q_1-q-q_2-q$ with independent and complete control over both the charge carrier type and density in the three different regions. This is achieved by using a global back gate (BG) to fix the overall carrier type and density and local top gates (TG$_1$, TG$_2$) to set the carrier type and density only below their overlap region with the graphene flake. By controlling the density under the two
top gates, various quantized conductance plateaus can be observed in the quantum Hall regime.

To study these effects, the device fabrication process is similar to the one for on-substrate Hall bar device. Here, in addition, we also coated the graphene with dielectric and then, fabricated top gates over it. Fig. 8(a) shows a schematic of the device where, \( V_{bg} (V_{tg1}, V_{tg2}) \) is the back (top) gate voltage and \( I_{ac} \) (∼50 nA) is used to measure the two probe resistance of the device by the lock-in technique. At low temperature, the basic characterization of the device starts with the measurement of charge carrier mobility, which we measured to be \( \sim 4800 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \). In presence of magnetic field perpendicular to the graphene’s plane, the measured resistance shows different plateaus corresponding to monolayer graphene. Also with top gate, we observe various fractionally quantized plateaus.

After the basic characterization, we move to the central experiment, which is the interaction of the edge channels induced by the two top gates. Fig. 8(b) shows the conductance \( (G) \) of the device (in units of \( (G_0 = e^2/h) \)) as a function of \( V_{tg1} \) and \( V_{tg2} \) with the back gate \( (V_{bg} = 31.1 \text{ V}) \) fixing the overall flake at the \( \nu = 2 \) plateau at \( B = 6 \text{ T} \). We observe many fractionally quantized conductance plateaus arising
Figure 9: (a) Schematic of the model employed to calculate the conductance ($G$). It shows the various edge channels and the possible types of equilibration at the junctions. (b) Colorscale plot showing calculated $G$ as a function of the two top gates while $V_{bg}$ set at $\nu = 2$ by taking into account the aspect ratio of the locally gated regions. The dotted line indicates the region in filling factor space probed in the experiment. (c) Line plot showing the slice of colorscale plot in (b) along the marked line.

due to the interactions between the edge channels induced below the two top gates mediated via the intermediate graphene lead. Fig. 8(c) shows line plots for slices of data shown in Fig. 8(b) with one of the top gates being varied continuously, while the other top gate and back gate are set at $\nu = 2$ plateau.

To explain the observed data we employ a simple model as shown in Fig. 9(a). Here the red/green channels represent electrons/holes. This shows just one of the various possible configurations of the regions [56]. This model calculates the effective filling factors based on the current conservation at each of the junctions with the reflection coefficients of the edge channel currents determined by the type of charge and also the number of channels present. As an example, let $\nu_1$ and $\nu_2$ be the number of edge channels on either side of the junction with $\nu_2$ being below a top gate. Then, one of the possibilities is, when both $\nu_1$ and $\nu_2$ are of the same carrier type but $|\nu_2| < |\nu_1|$ in which case only the channels present in $\nu_2$ are transmitted giving a reflection coefficient $r_1 = 1 - \frac{\nu_2}{\nu_1}$ into region 1. Similarly other cases for different configuration (as shown in Fig. 9(a)) of filling factors can be solved. Applying current
conservation at each interface gives an effective filling factor \( \nu_{\text{eff}} = \frac{\nu_1 \nu_2}{\nu_1 - \nu_2 + 3 \nu_1 \nu_2} \). To further improve upon this, we took into account the aspect ratio (ratio of device width to the length) and incorporated it for the calculation of the effective filling factors \[57\]. Conductance calculated following this procedure is plotted in Fig. 9(b). Comparing Fig. 8(b) with Fig. 9(b), we see a reasonable match between them.

This calculation is however still unable to explain the diagonal asymmetry and the peaks and valleys in Fig. 8(c) deviating from flat plateaus in conductance as expected from Fig. 9(b). There could be various possibilities like aspect ratio of device \[57, 58\] and inhomogeneities present under the locally top gated region. The distribution of the impurity below the two top gates can lead to differences in their effect on the conductance quantization, including the oscillations seen in the modulation due to \( TG_2 \) which is absent in the modulation due to \( TG_1 \) \[55\]. This shows that there is an asymmetry in the properties of the regions below the two top gates leading to the asymmetry seen in Fig. 8(b).

We studied the effect of two independently locally gated regions on the conductance of graphene in quantum Hall regime and found various conductance plateaus and also the critical role that impurity plays. We explained these results with a model based on the equilibration of the channels at multiple junctions. This will help in developing a better understanding of lateral heterostructures for applications like metrology \[59\].
Bibliography


[28] Singh *et al.*, V. Graphene electromechanics in quantum hall regime. (*Manuscript under preparation*).


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Chapter 1

Introduction

In the quest for studying nanostructures several breakthroughs have been made in the field of materials science. The nobel prize winning discovery in 1996 of a new form of carbon, C60, in the shape of a soccer ball, besides already known allotropes like diamond and graphite, has let to a renewed focus on carbon based nanostructures. Carbon with an atomic number of 6 has a electronic configuration of 1s\(^2\) 2s\(^2\) 2p\(^2\). The the outer shell of 2s\(^2\) 2p\(^2\) is very adaptable and can hybridize to form molecular orbitals (starting with half filled 2s\(^1\) 2p\(^3\)) that have the character of both s and p orbital. A hybridization of sp\(^3\) nature leads to four orbital with tetrahedral symmetry seen in diamond, sp\(^2\) hybridization leads to a hexagonal symmetry seen in graphite, graphene, carbon nanotubes and C60, and sp hybridization leads to organic molecules like acetylene. The malleable nature of carbon’s molecular orbital can be seen in the world around us dominated by carbon – key ingredient of the living world. As a result of the discovery of C60 [1], there is a push for development of electronics based on carbon. Ijima discovered in 1991 another allotrope of carbon called carbon nanotubes [2]. Carbon nanotubes (CNT) with single walls consist of a single atom thick sheet of graphite (also called graphene) rolled into a seamless cylinder. Depending on the diameter and the axis of rolling these carbon nanotubes were either metallic, or semiconducting. As a natural sequence of scientific evolution researchers worked in several teams all over the world to isolate a single layer of graphene. Andre Geim and Konstantin Novolselov came up with an ingenious method after years of effort [3] to isolate monolayer graphene flakes and demonstrated field effect transistor using
Chapter 1. Introduction

graphene [4]. This simple idea behind the discovery has led to a new field that is growing very rapidly exploring amazing electrical, mechanical, thermal and optical properties of graphene, perhaps making it the most studied material in condensed matter physics in last few years. It is important to understand the underlying science behind the material to appreciate the impact of this discovery.

It is interesting to note that the basic structure that gives rise to graphite, carbon nanotubes and C60 is graphene with sp² hybridized molecular orbital, however, chronologically it was the last to be isolated. The sp² bonds between the nearest neighbor atoms have a strong wavefunction overlap and give rise to a very strong covalent bond. However, it is the half-filled shell of unhybridized pz gives the state its unique electrical properties due to the overlap with nearest neighbors to form π orbital. These encompass properties important from the technological application point of view like high mobility and faster response of electrons but also important from the fundamental physics point of view by providing a playground to study 2+1 dimensional quantum electrodynamics [5].

In addition to the interesting nature of electrons, graphene is electronically an exciting material because it is strictly 2 dimensional as a result some very unique experiments like the quantum Hall effect (QHE), which requires a 2 dimensional electron gas, can be seen in graphene. On the other hand, strong covalent bonds due to a large overlap between the nearest neighbor carbon atoms that are sp² hybridized, give rise to very large in-plane Young’s modulus (~1 TPa) [6]. In addition to this, graphene can sustain 20% strain before undergoing permanent deformation [6]. A large field has developed over the last several decades studying nanoelectromechanical systems (NEMS) [7, 8]. High Young’s modulus, large surface to volume ratio and sensitivity towards chemical specific processes [9] of graphene offer a distinct advantage over other nanostructures for such applications.

This thesis presents results probing the fundamental physics in graphene as well as results pertaining to the technological aspects of graphene resonators. Later in this thesis, we also presents results merging these two aspects, specifically QHE and the mechanical motion. Such results are helpful in our understanding of quantum Hall state against the mechanical perturbation and further allow us to manipulate the electromechanics of graphene resonators with the help of external parameter like
magnetic field.

Integer quantum Hall effect (IQHE) was first observed by Klaus von Klitzing in the seminal experiment [10] in 1980. Soon after, improvements in electron mobility lead to the discovery of fractional quantum Hall effect in 1982 [11], which arises from the electron-electron interaction (this interaction is not essential for IQHE). It is quite interesting to see that being such an old area of research, it still brings surprises. In 2005, relativistic-like or anomalous quantum Hall effect was observed in graphene [12, 13] and studied extensively [14]. QHE has been studied extensively in 2D systems [15, 16, 17] and its equilibrium electron-transport properties are understood to a large extent. The breakdown of the QHE under non-equilibrium conditions due to a high current density has been studied to understand its microscopic origin [18, 19]. There has been a considerable debate in the literature regarding the details of the mechanism of QHE breakdown. The proposed mechanisms include electron heating [20], electron-phonon scattering [21, 22], inter and intra Landau level (LL) scattering [23, 24], percolation of incompressible regions [25] and the existence of compressible regions in the bulk [26]. The unique band structure of graphene near the Fermi energy ($E = \pm c\hbar|k|$, where $c \approx 10^6$ ms$^{-1}$ is the Fermi velocity) gives rise to a ‘relativistic’ QHE. The energy scale set by the cyclotron gap in graphene is much larger than that of 2 dimensional electron gas realized in semiconductor heterostructures at same magnetic field. In presence of transverse electric field, mixing of wavefunctions could cause inter Landau level scattering leading to the breakdown of QHE. Chapter 4 experimentally probes the breakdown of QHE in graphene by injecting a high current density.

In addition to the electronic properties, the remarkable mechanical properties of graphene include a high in-plane Young’s modulus of $\sim$1 TPa probed using nanoindentation of suspended graphene [6], force extension measurements [27], and electromechanical resonators [28, 29, 30]. NEMS devices using nanostructures like carbon nanotubes [31, 32, 33, 34, 35, 36, 37], nanowires [38, 39] and bulk micromachined structures [40, 41, 42] offer promise of new applications and allow us to probe fundamental properties at the nanoscale. NEMS [7, 8] based devices are ideal platforms to harness the unique mechanical properties of graphene. Electromechanical measurements with graphene resonators [28, 29] suggest that with improvement of quality factor ($Q$), graphene based NEMS devices have the potential to be very sensitive de-
Chapter 1. Introduction

detectors of mass and charge. In order to better understand the potential of graphene based electrically actuated and detected resonators and the challenges in realizing strain-engineered graphene devices [43, 44], we experimentally measure the coefficient of thermal expansion of graphene ($\alpha_{\text{graphene}}(T)$) as a function of temperature. Chapter 5 covers the study of graphene NEMS in doubly clamped geometry at low temperatures providing details about the tunability of the resonant frequency and describes the measurement of thermal expansion coefficient of suspended graphene.

NEMS have emerged as an active field for studying mechanical oscillations of nanoscale resonators and thereby provide a good platform for sensing mass [35, 36, 42], charge [45], magnetic flux and magnetic moments [46, 47, 48, 49]. Actuation and detection of these systems can be done using electrical signals; this makes them suitable for probing the coupling of electrical and mechanical properties [33, 34, 50]. Graphene in doubly clamped suspended geometry devices not only form an active part of the NEMS but also provide a quantum Hall system in presence of magnetic field. This enables us to conduct experiments to answer the following two questions - how is the quantum Hall state modified due to mechanical vibrations, and how does the quantum Hall (QH) state affect the electromechanics of the resonator? Chapter 6 tries to answer some of these questions.

The advantage of controlling the charge type and electric field locally adds a new dimension to study electron transport in graphene [51] to see effects like Klein tunneling [52, 53], Andreev reflection [54], collapse of Landau levels [55], Veselago lens [56] and collimation of electrons with top gates [57]. The top gate geometry has been utilized in controlling the edge channels in the quantum Hall regime and with control over local and global carrier density. Such $p-n$ [53, 58] and $p-n-p$ junctions [59] show integer and fractional quantized conductance plateaus. These integer and fractional quantized plateaus have been explained with the reflection and mixing of the edge channels leading to the partition of the current [60]. In Chapter 7, we study electron transport in a graphene multiple lateral heterojunction device with charge density distribution of the type $q_{1}-q_{2}-q$ with independent and complete control over both the charge carrier type and density in the three different regions.

We start by providing necessary theoretical background in Chapter 2. Various steps in device fabrication and various measurement schemes are explained in Chapter
3. In Chapter 8, we provide summary and the outlook of the future work.
Chapter 2

Theoretical background

In this chapter, we will describe the basic ideas which will help in understanding the experiments presented in the following chapters. We will start with the band-structure calculations of graphene, which helps in understanding the electronic properties. Thereafter, we discuss anomalous quantum Hall effect in graphene contrasting it with the integer quantum Hall effect in two dimensional electron gas. We discuss the concepts related to nanoelectromechanical systems next, covering the concepts related to Duffing nonlinearity and nonlinear damping.

2.1 Band structure of graphene

Graphene is one atom thick two-dimensional sheet of carbon atoms arranged in a honeycomb lattice \[14, 61\]. In this structure, \(sp^2\) hybridization of carbon atoms gives rise to 3 in-plane \(\sigma\)-bonds with the \(p_z\)-orbital pointing out of the plane. While the \(p_z\) orbital, which form \(\pi\) bonds, give rise to exciting electronic properties.

The honeycomb lattice of graphene is not a Bravais lattice since the two neighboring sites are inequivalent. For example, as shown in Figure 2.1(a), there is one lattice point on the right side of the point A, however for point B there is no lattice point on the right. The crystal structure of graphene, however, can be thought of as made from two carbon atom basis on triangular Bravais lattice. The dotted rhombus
2.1. Band structure of graphene

in Figure 2.1(a) indicates the unit cell with two carbon atoms in it. To describe the triangular lattice, we can choose the primitive vectors as indicated in Figure 2.1(a) and given by

\[ \vec{a}_1 = \left( \frac{\sqrt{3}}{2}a, a \right), \quad \text{and} \quad \vec{a}_2 = \left( \frac{\sqrt{3}}{2}a, -a \right), \]  

(2.1)

where \( a = |\vec{a}_1| = |\vec{a}_2| = 1.42 \times \sqrt{3} = 2.46\text{Å} \) and \( a \) denotes the lattice constant. Following the orthogonality relations, \( \vec{a}_i . \vec{b}_j = 2\pi\delta_{ij} \), we can write down the primitive vectors to describe the reciprocal lattice as,

\[ \vec{b}_1 = \left( \frac{2\pi}{\sqrt{3}a}, \frac{2\pi}{a} \right), \quad \text{and} \quad \vec{b}_1 = \left( \frac{2\pi}{\sqrt{3}a}, -\frac{2\pi}{a} \right). \]  

(2.2)

Figure 2.1(b) shows the reciprocal space lattice indicating the various symmetry points in the Brillouin zone (shaded region). The band structure of graphene provides very useful insights in understanding the basic electronic properties. The band structure of graphene was calculated in the seminal work by Wallace [62] while studying the band structure of graphite using tight binding model. The band structure of graphene not only forms the basis for the understanding of graphite (3D), but also other carbon allotropes like carbon nanotube (1D) and fullerene (0D).

The tight binding method for crystals is very similar to the linear combination of the atomic orbital (LCAO) for molecules used in chemistry and we will be following the treatment given in Ref[61]. To express the eigenfunctions in solid \( \Psi_j(\vec{k}, \vec{r}) \) linear combinations of Bloch functions \( \Phi_j(\vec{k}, \vec{r}) \) are used as follows:

\[ \Psi_j(\vec{k}, \vec{r}) = \sum_{j'=1}^{n} C_{jj'}(\vec{k})\Phi_{j'}(\vec{k}, \vec{r}), \]  

(2.3)

where the index runs over the number of Bloch wavefunctions and \( C_{jj'} \) are the unknown coefficient which need to be determined. The Bloch wavefunctions are constituted as a linear sum over the lattice vectors \( \vec{R} \) of atomic orbital in the unit cell weighted by a phase factor \( e^{i\vec{k}.\vec{R}} \) as written below:
Figure 2.1: a) Lattice structure of graphene showing the unit cell with two carbon atom basis. Unit vectors $\vec{a}_1$ and $\vec{a}_2$ can be used to describe the crystal structure of graphene on a triangular lattice with two carbon atom basis. b) Brillouin zone of graphene indicating the primitive vectors for reciprocal lattice ($\vec{b}_1$ and $\vec{b}_2$). Symmetry points of the reciprocal space $\Gamma$, $K$ and $M$ are also shown.
2.1. Band structure of graphene

$$\Phi_j(\vec{k}, \vec{r}) = \frac{1}{\sqrt{N}} \sum_{\vec{R}} e^{i\vec{k} \cdot \vec{R}} \phi_j(\vec{r} - \vec{R}), \quad (2.4)$$

where $\phi_j$ denotes the atomic wavefunction in state $j$. The weighted sum with the phase factors is taken to satisfy Bloch’s theorem due to the translation symmetry of the lattice.

Once the wavefunctions of the solid $\Psi_j(\vec{k}, \vec{r})$ are written, $j$-th state energy eigenvalue can be written as,

$$E_j(\vec{k}) = \frac{\int \Psi_j^* H \Psi_j d\vec{r}}{\int \Psi_j^* \Psi_j d\vec{r}}. \quad (2.5)$$

Using Equation (2.3) and (2.5), we can write,

$$E_i(\vec{k}) = \frac{\sum_{j,j'=1}^N H_{ij}(\vec{k}) C_{ij}' C_{ij'}}{\sum_{j,j'=1}^N S_{jj'}(\vec{k}) C_{ij}' C_{ij'}}, \quad (2.6)$$

where $H_{jj}(\vec{k}) \equiv \langle \Phi_j | H | \Phi_{j'} \rangle$ and $S_{jj'}(\vec{k}) \equiv \langle \Phi_j | \Phi_{j'} \rangle$ are called transfer and overlap matrix, respectively. In order to calculate the energy eigenvalue the coefficient $C_{ij}$ needs to be optimized so that $E_i(\vec{k})$ can be minimized, which leads to,

$$\sum_{j'=1}^N H_{jj'}(\vec{k}) C_{ij'} = E_i(\vec{k}) \sum_{j'=1}^N S_{jj'}(\vec{k}) C_{ij'}. \quad (2.7)$$

For a non-trivial solution of $C_{ij}$ (and hence wavefunctions), $Det[H - ES]$ must vanish. This is called secular equation and solution of this gives the energy dispersion in reciprocal space.

This so far is the general prescription for the tight binding calculation of the band structure. Now, we will apply this to the case of graphene. As discussed before, $sp^2$ hybridization in graphene leads to the formation of $3$-$\sigma$ bonds while the $p_z$ orbital, which is perpendicular to the plane of graphene, makes $\pi$ bonds. Here we consider the $\pi$ energy bands only as these determine the electronic properties. The Bloch wavefunctions consisting of the site A and B in the unit cell as shown in Figure 2.1(a)
Chapter 2. Theoretical background

can be written as,

$$\Phi_{2p_z}(\vec{r}) = \frac{1}{\sqrt{N}} \sum_{\vec{R}_\alpha} e^{i\vec{k} \cdot \vec{R}_\alpha} \phi_{2p_z}(\vec{r} - \vec{R}_\alpha),$$ (2.8)

where $\alpha$ represents sites $A$ and $B$ and summation is taken over atom site coordinate $\vec{R}_\alpha$ for $A$ or $B$. Since we will be dealing with $2p_z$ orbital only, we can discard this index. The overlap matrix can be calculated as,

$$H_{AA}(\vec{r}) = \frac{1}{N} \sum_{\vec{R}, \vec{R}'} e^{i\vec{k} \cdot (\vec{R} - \vec{R}')} \langle \phi_A(\vec{r} - \vec{R}) | H | \phi_A(\vec{r} - \vec{R}) \rangle$$

$$= \frac{1}{N} \sum_{\vec{R} = \vec{R}'} \epsilon_{2p} + \frac{1}{N} \sum_{\vec{R} \neq \vec{R}'} e^{i\vec{k} \cdot (\vec{R} - \vec{R}') \langle \phi_A(\vec{r} - \vec{R}) | H | \phi_A(\vec{r} - \vec{R}) \rangle}. \quad (2.9)$$

The largest contribution in the above equation comes from the first term which is equal to $\epsilon_{2p}$, which is the energy of the $2p$ level. The second term in the above equation can be neglected for simplicity as the overlap between the wavefunction at $\vec{R}$ and $\vec{R} + \vec{a}_i$ contributes by a small amount. Similarly, the other component for site $B$, $H_{BB}$, gives $\epsilon_{2p}$. To calculate the off-diagonal terms of transfer matrix $H_{AB}$, we have to consider three nearest neighbor $B$ atoms relative to an $A$ atom. Lets denote them by $\vec{R}_1$, $\vec{R}_2$ and $\vec{R}_3$. Following the reasons given before we will consider only the first nearest neighbors, therefore,

$$H_{AB} = \frac{1}{N} \sum_{\vec{R}, \vec{R}_i} e^{i\vec{k} \cdot \vec{R}_i} \langle \phi_A(\vec{r} - \vec{R}) | H | \phi_B(\vec{r} - \vec{R} - \vec{R}_i) \rangle$$

$$= t \left( e^{i\vec{k} \cdot \vec{R}_1} + e^{i\vec{k} \cdot \vec{R}_2} + e^{i\vec{k} \cdot \vec{R}_3} \right)$$ (2.10)

where $t = \sum_{\vec{R}_i} \langle \phi_A(\vec{r} - \vec{R}) | H | \phi_B(\vec{r} - \vec{R} - \vec{R}_i) \rangle$. By taking $\vec{R}_1 = \frac{a}{\sqrt{3}} (1, 0)$, $\vec{R}_2 = \frac{a}{\sqrt{3}} \left( \frac{1}{2}, \frac{\sqrt{3}}{2} \right)$ and $\vec{R}_3 = \frac{a}{\sqrt{3}} \left( \frac{1}{2}, -\frac{\sqrt{3}}{2} \right)$, we can simplify $H_{AB}$ as,
2.1. Band structure of graphene

\[ H_{AB} = t \left( e^{ik_x a/\sqrt{3}} + 2 e^{-ik_x a/2 \sqrt{3}} \cos \left( \frac{k_y a}{2} \right) \right), \]
\[ \equiv tf(k). \]  \hspace{1cm} (2.11)

Since Hamiltonian forms a Hermitian matrix, we can write \( H_{BA} = H_{AB}^* \). Therefore 2×2 transfer matrix can be written as,

\[ H = \begin{pmatrix} \epsilon_{2p} & tf(k) \\ tf(k)^* & \epsilon_{2p} \end{pmatrix}. \]  \hspace{1cm} (2.12)

Following along the same line we can also calculates the components of overlap matrix \( S \), given by,

\[ S = \begin{pmatrix} 1 & sf(k) \\ sf(k)^* & 1 \end{pmatrix}, \]  \hspace{1cm} (2.13)

where \( s = \sum_{\vec{R}_i} \langle \phi_A(\vec{r} - \vec{R}) | H | \phi_B(\vec{r} - \vec{R} - \vec{R}_i) \rangle. \)

As described earlier, solving \( \det[H - ES] = 0 \) gives the energy dispersion relation. Therefore using Equation (2.12) and (2.13), we can write

\[ E = \frac{\epsilon_{2p} \pm tw(k_x, k_y)}{1 \pm sw(k_x, k_y)}, \]  \hspace{1cm} (2.14)

where \( w(k_x, k_y) = \sqrt{|f(k)|^2} = \sqrt{1 + 4 \cos \frac{\sqrt{3} k_x a}{2} \cos \frac{k_y a}{2} + 4 \cos^2 \frac{k_y a}{2}}. \)

Equation (2.14) gives the energy dispersion relation for graphene with \( t \) and \( s \) being the only unknown parameters. First principle calculations \cite{63} suggest \( t = -3.033 \) eV and \( s = 0.129 \) eV. Figure 2.2(a) shows the energy dispersion in the first Brillouin zone. The conduction and valence band meet at 6 points (\( K \)-points) out of which only 2 are distinct from the symmetry of the Brillouin zone. Since each band has \( 2N \)-states (including spin) and each carbon atom contributes one electron, therefore the valence band gets completely filled and Fermi level lies exactly at six meeting points of conduction and valence band. Figure 2.2(b) shows a zoomed-in view of the dispersion relation at the \( K \)-point where linearly dispersing conduction and valence
Figure 2.2: a) Energy dispersion plotted within first Brillouin zone. The conduction and valence band meets at six different points (K-point). b) Zoomed-in view of energy dispersion relation at one of the K-point, showing the linear dispersion of energy with $|\vec{q}|$ (also commonly referred as Dirac cone)
2.2 Density of states of graphene

The density of states (DOS) for graphene was first derived by Wallace \cite{62}. Close to the Dirac point the DOS per unit area per unit energy for graphene is given by \cite{14},
Chapter 2. Theoretical background

\[ \rho(E) = \frac{2}{\pi} \frac{|E|}{\hbar^2 v_f^2}, \]

(2.18)

which indicates that right at the Dirac point the DOS vanishes. Therefore, owing to zero band gap (Equation (2.17)) and vanishing DOS at Dirac point, graphene can be called a zero-gap semiconductor, or a zero-overlap semimetal.

2.3 Graphene field effect transistor

Vanishing density of states at the Fermi point and zero band gap in graphene makes it possible to tune the Fermi level (or carrier density) continuously from valence band to conduction band by electrical means. The electrical doping is achieved by making graphene devices in a field effect transistor (FET) geometry. Graphene flake and gate electrode form a parallel plate capacitor with a dielectric in between as shown in Figure 2.3(a). Source-drain contacts are used to measure the resistance/conductance of the graphene flake. By applying a gate voltage, charges can be induced on the graphene sheet leading to the electrical doping of the flake. Figure 2.3(b) shows a typical measurement of resistance of the graphene device with gate voltage. A positive/negative gate voltage induced electrons/holes on the flake and hence pushes the Fermi level deeper into the conduction/valence band. DOS increases as Fermi level moves away from Dirac point which reflects in a decrease in resistance. To understand the charge transport in graphene Drude model can be a good starting point, however to understand it better one has to consider the effect of short range scatterers [64, 65, 66], presence of charges impurities [64, 65, 67] and ripples on the surface of graphene [68].

Following the semi-classical Drude model, for both types of charge carrier, the conductivity of graphene \( \sigma \) can be related to the number of charge carriers per unit area \( n \), by \( \sigma = ne\mu \), where \( e \) is the electronic charge and \( \mu \) is charge carrier mobility. The charge carrier density \( n \) on the graphene is related to the gate voltage via the capacitance per unit area \( C_g' \) as \( n = \frac{1}{e} C_g' V_g \), where \( V_g \) is the applied gate voltage and \( C_g' \) is the capacitance per unit area between graphene flake and the gate electrode. Therefore following a linear response of conductivity with carrier density, the field
2.3. Graphene field effect transistor

Figure 2.3: a) Schematic diagram of a graphene field effect transistor along with the measurement scheme. b) A typical measurement of resistance of graphene flake with gate voltage in FET geometry. The insets show the position of Fermi level for two types of induced carriers.
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effect mobility \( \mu \) for a graphene device of length \( L \) and width \( W \) can be written as,

\[
\mu = \frac{1}{C_g' L} \frac{dG}{dV_g}.
\] (2.19)

For a 300 nm SiO\(_2\) coated wafer, which are predominantly used for graphene device fabrication, using \( C_g' = \varepsilon_0 \varepsilon_r d \) with \( d = 300 \) nm, we get \( n[\text{cm}^{-2}] = 7.2 \times 10^{10} V_g[\text{V}] \). Equation (2.19) can be simplified to give \( \mu[\text{cm}^2 \text{V}^{-1} \text{s}^{-1}] \approx 86 \times \frac{L}{W} \times \frac{dG}{dV_g} [\mu \text{S/ V}] \). It also useful to relate the change in the position of the Fermi level \( E_f \), relative to the charge neutral graphene, with gate voltage \( V_g \) in such devices. Using Equation (2.18) and taking \( v_f = 1 \times 10^6 \text{ ms}^{-1} \) at \( T = 0 \) K, we get \( E_f[\text{meV}] \approx 31.3 \times \sqrt{V_g[\text{V}]} \).

All the description presented above is valid for undoped graphene, where resistance maxima occurs at zero gate voltage. However, during the device fabrication, most of the times, resist residue or other kinds of dopants cause the charge neutral point to occur at a nonzero gate voltage.

2.4 Quantum Hall effect

Quantum Hall effect (QHE) is a quantum mechanical phenomena that occurs in two dimensional electronic systems subjected to magnetic field and is typically observable at low temperatures. In presence of magnetic field the current density \( \vec{J} \) no longer remains parallel to the applied electric field \( \vec{E} \), \( \vec{J} = \sigma \vec{E} \), and hence conductance of the sample is described by 2 \( \times \) 2 conductivity tensor with two independent components namely, longitudinal conductivity \( \sigma_{xx} \) and transverse conductivity \( \sigma_{xy} \). The localization-delocalization of electrons (for integer QHE) and electron-electron interactions (for fractional QHE) lead to versatile phenomena like dissipationless transport across the system and at the same time transverse conductance becomes quantized in units of \( e^2/h \) irrespective of the material properties and sample geometry. A complete description of quantum Hall phenomena is out of the scope of this thesis. Here we try to present essential concepts for the case of integer quantum Hall effect. A complete and detailed description of quantum Hall phenomena is available in literature [15, 16, 17].
2.4. Quantum Hall effect

To illustrate the idea of quantum Hall effect, we will consider spinless free electrons in two dimensions and in the presence of magnetic field perpendicular to its plane. In the presence of magnetic field, Schrodinger’s equation can be written as,

\[
\frac{(\vec{p} + e\vec{A})^2}{2m} \psi = E \psi, \tag{2.20}
\]

where \( \vec{p} \) and \( \vec{A} \) are the momentum operator and vector potential, respectively. To solve it further, we need to pick up a particular gauge to relate the potentials to the fields. The most convenient gauge for this problem is the Landau gauge where \( \vec{A} \) is taken as \((0, Bx)\). Therefore above equation can be written as,

\[
\frac{1}{2m} \left( -\hbar^2 \frac{\partial^2}{\partial x^2} + \left( -i\hbar \frac{\partial}{\partial y} + eBx \right)^2 \right) \psi = E \psi. \tag{2.21}
\]

By applying the method of separation of variables and demanding a solution of the form \( \psi \propto e^{iky} \phi(x) \), we get

\[
\left( -\frac{\hbar^2}{2m} \frac{d^2}{dx^2} + \frac{1}{2} m\omega_c^2 \left( x + \frac{\hbar k}{eB} \right)^2 \right) \phi(x) = E \phi(x), \tag{2.22}
\]

where \( \omega_c = \frac{eB}{m} \). This equation is same as that for a one-dimensional simple harmonic oscillator (SHO) with shifted center of potential energy at \( x_k = -\frac{\hbar k}{eB} \equiv -l_B^2 k \). Here, the length scale \( l_B = \sqrt{\frac{\hbar}{eB}} \) is called magnetic length. Therefore, following the solution of a SHO, we can write the energy eigenvalue and wavefunctions as

\[
E_{n,k} = (n + \frac{1}{2})\hbar \omega_c, \tag{2.23}
\]

\[
\psi_{n,k}(x, y) \propto H_n \left( \frac{x - x_k}{l_B} \right) e^{iky} \left( -\frac{(x - x_k)^2}{2l_B^2} \right) e^{iky}, \tag{2.24}
\]

where \( n = 0, 1, 2..., \) and the \( H_n \) are Hermite polynomials. Several key points can be noted in the solution given by Equation (2.23) and (2.24). First, we note that energy
eigenvalues depend only on $n$, called Landau level index, and not on wavevector $k$. For a given Landau level (LL), various states with different $k$ will have same energy making the LL highly degenerate. Therefore a continuum density of states spectrum (2D case) splits into $\delta$-function like highly degenerate Landau levels. The position of these levels is given by Equation 2.23. Second, the wavefunctions have plane wave like delocalized solution in $y$-direction and harmonic oscillator like localized solution in $x$-direction. Such a preference in direction is a consequence of Landau gauge chosen to relate the potentials to the fields. By taking a different gauge, a different set of wave functions can be obtained. However, high degeneracy of Landau levels allows us to obtain these wavefunctions from the linear combinations of the wavefunctions given by Equation 2.24, therefore forming a complete set to describe the problem. Another remarkable point is that by taking the linear combination of the wave functions given by Equation 2.24, it is possible to construct the states which are completely localized in both directions with the same energy. Such results can also be directly obtained by choosing a symmetric gauge $\vec{B} = \frac{1}{2} \vec{A} \times \vec{r}$. The importance of these localized states for the observation of QHE will become later in this section.

Third, the wavefunctions in $x$-direction are centered at $x_k = \frac{l_B^2}{2} k$. If the y dimension of the system is confined between 0 and $W$, it sets the bound for $k$ to be within 0 and $\frac{W}{l_B}$. On the other hand, the periodic boundary condition $\psi(x, y) = \psi(x, y + L)$ leads to the allowed values for the wavevector $k$, given by $k = \frac{2\pi p}{L}$, where $p$ is an integer. Together these boundary conditions imply that total number of states for a LL is given by $\frac{LW}{2\pi l_B^2}$, which is the degeneracy $n_B$ of each Landau level. Therefore, the number of states per unit area in any LL is $n_B = \frac{1}{2\pi l_B^2} = \frac{eB}{\hbar}$. Also the gap between the LLs, cyclotron gap, $\hbar\omega_c = \frac{eB}{m}$ increase as we increase the magnetic field causing the LLs to move further apart from each other. At the same time degeneracy of the LLs $\frac{eB}{\hbar}$ increases too, creating more states within each Landau level. For a fixed number of total carriers when magnetic field is increased, these two competing effects lead to the oscillations in the chemical potential $\mu$.

In this idealized model, therefore, a continuous DOS spectrum transformed into discrete, highly degenerate, uniformly spaced Landau levels as depicted in schematic diagram in Figure 2.4(a) and (b). When the Fermi energy lies between the gap of two LLs there is no scattering leading to vanishing $\rho_{xx}$. So far, we have completely ignored the role of disorder and impurities. However disorder plays a very important
2.4. Quantum Hall effect

Figure 2.4: a) Density of states (DOS) for a 2-dimensional free electron gas in zero magnetic field. b) DOS plotted in presence of magnetic field. A continuous DOS spectrum splits into sharp discrete levels. c) Disorder in the system leads to the broadening of the otherwise sharp Landau levels by creating localized states around the extended state band.
role in the observation of quantum Hall effect. To illustrate this, we revisit the model without disorder presented above. At low temperatures, the charge carrier density \( n \) in the system can be written as \( n = \int_{-\infty}^{\epsilon_f} \rho(\epsilon, B) d\epsilon \), where \( \rho(\epsilon, B) \) is the DOS. Since the change in electron density can be related to the change in Fermi energy by \( \delta n = \rho(\epsilon_f, B) \delta \epsilon_f \). As we add or remove the charge carriers in the system, Fermi level moves faster close to the energies with low DOS and slower in the regions of large density of states. Therefore large DOS regions have a tendency to pin the Fermi level as we add or remove charge carriers. In our idealized model, Fermi level will get pinned to the position of the Landau level until it is completely filled. Once completely filled, Fermi level will jump to the next LL without staying in the gap since there is zero DOS. Therefore Fermi level would never be located between two Landau levels. This would not be much of a problem for a narrow conductor where edge states at the sample boundary provide enough DOS to stabilize the Fermi level in the gap. However in wide samples, the fraction of edge states to the bulk states could be very small and vanishing \( \rho_{xx} \) should not be seen as Fermi level will never be stabilized in the gap.

Disorder and impurities in the sample provide localized states between the LLs in order to stabilize the Fermi level while moving from one LL to the next. The effect of impurities on the energy spectrum and wavefunctions can be seen by considering a simplified model of a \( \delta \)-function like impurity potential. Consider a single impurity potential given by \( V = \lambda \delta(x-x_0) \delta(y-y_0) \), where \( \lambda \) is the strength of the potential and it is located at \((x_0, y_0)\). For weak impurity potential, we can treat it as a perturbation term in the free electron Hamiltonian in magnetic field used in Equation (2.21) and we can determine the perturbed energy levels and wave functions. The results are that: 1) Each perturbed energy level lies between two unperturbed levels except for lowest LL and there is one state completely localized for each LL. 2) Energy of these perturbed levels lies away from \( n\hbar\omega_c \), roughly by \( \lambda \) and these states are centered at the position of impurity \((x_0, y_0)\). 3) If the degeneracy of LL is \( n_B \), there are \( n_B - 1 \) extended states in each LL. Therefore, in the limit of large impurities, the sharp Landau levels evolve into broader LL. This is schematically shown in Figure 2.4(c). There are two kinds of states in these broad LLs. At the core of LLs, there are extended states, which carries current. These cores are surrounded by localized states, which do not carry any current. Thus, impurities reduced the number of
current carrying states, but current remains same as long as all the extended states are occupied. This is the reason for highly flat plateaus in \( R_{xy} \).

## 2.5 Electron transport in quantum Hall limit

Most of the measurements in quantum Hall limit are performed in a Hall bar geometry which allows a direct measurement of longitudinal resistivity \( \rho_{xx} \) and transverse resistivity \( \rho_{xy} \). A schematic diagram of Hall bar geometry is shown in Figure 2.5(a). In this section, we give a brief overview of the characteristics of the electron transport in Hall bar geometry devices. We will keep our discussion within the terms of single electron states described in the previous section. In previous discussion, we have completely ignored the confining potential due to finite sample size except for deriving the degeneracy of the Landau levels. This description is valid for the Landau levels in the bulk of the sample. However, as one moves closer to the physical edges of the sample confining potential leads to the divergence in the otherwise flat Landau level energies as depicted in Figure 2.5(b). Inhomogeneities in the sample bulk leads to small modulation in the bulk LLs energy. When Fermi level lies between two LLs, states only at the sample boundaries take part in electron transport. Under small bias \( (\delta \mu) \), these edge states participate in electron transport whereas electron transport through the sample bulk remains largely suppressed. The states at the two edges of the sample move with opposite momentum with group velocity \( \frac{1}{\hbar} \frac{dE_n(k)}{dk} \). Since the states with opposite momentum get spatially separated, electron can move along the edges without momentum relaxation. This leads to the vanishing longitudinal resistance. The source and the drain contacts equilibrate the edge channels to the chemical potentials of the two contacts \( \mu_1 \) and \( \mu_2 \) (as depicted in Figure 2.5(a)). Therefore measurement of transverse resistance will be given by \( R_H = \frac{1}{e} \frac{\mu_1 - \mu_2}{T} \), where \( I \) is the injected current. Following the Landaurer-Buttiker (LB) formalism for ballistic transport [69], the current in the sample can be related to the chemical potential difference as \( I = \frac{2e}{\hbar} (\mu_1 - \mu_2) M \), where \( M \) is the number of edge channels lying below the Fermi level. Therefore,

\[
R_H = \frac{\hbar}{2e^2 M} \tag{2.25}
\]
Chapter 2. Theoretical background

Figure 2.5: a) Schematic diagram of Hall bar geometry. Chemical potential imbalance ($\delta \mu = \mu_1 - \mu_2$) leads the electrons to flow between source and drain allowing direct measurement of $R_{xx}$ and $R_{xy}$. b) Landau level spectrum plotted along the width of the sample. Landau level energies diverge at the sample boundaries. Electronic states with forward and backward momentum get spatially separated.
2.6 Quantum Hall effect in graphene

Many of the concepts from previous discussions get carried through to describe the quantum Hall effect (QHE) in graphene. However, linear dispersion and vanishing density of states at the Dirac point add a different flavor to it (sometimes also referred as anomalous QHE or relativistic-like QHE). The quantized tight binding Hamiltonian of graphene in zero magnetic field is given by \[\hat{H} = v_F \sigma \cdot \vec{p},\] where \(\sigma\) is \(2 \times 2\) Pauli matrix. In the presence of magnetic field, the Hamiltonian can be obtained by replacing \(\vec{p}\) by \(\vec{p} + e\vec{A}\). Therefore 2-dimensional Dirac equation in magnetic field can be written as,

\[
v_F \vec{\sigma}. \left( -i\hbar \vec{\nabla} + e\vec{A} \right) \psi = E \psi. \tag{2.26}
\]

By taking Landau gauge \(\vec{A} = (-By, 0)\) and demanding the solution of the form \(\psi = e^{ikx} \phi(y)\), above equation can be simplified as,

\[
\hbar v_F \begin{pmatrix} 0 & \partial_y - k + \frac{Bey}{\hbar} \\ -\partial_y - k + \frac{Bey}{\hbar} & 0 \end{pmatrix} \phi(y) = E \phi(y). \tag{2.27}
\]

As was done in the previous section, we can shift the origin in y-direction to \(\xi = \frac{y}{l_B} - l_Bk\), where \(l_B\) is the magnetic length. By introducing 1-dimensional harmonic oscillator operators \(O = \frac{1}{\sqrt{2}} (\xi + \partial_\xi)\) and \(O^\dagger = \frac{1}{\sqrt{2}} (\xi - \partial_\xi)\), above equation can be further simplified to,

\[
(O\sigma^+ + O^\dagger \sigma^-) \phi(y) = \frac{2E}{\omega_c} \phi(y), \tag{2.28}
\]

where \(\sigma^\pm (= \sigma_x \pm i\sigma_y)\) are raising and lowering operators and \(\omega_c\) is the cyclotron frequency given by \(\sqrt{\frac{2v_F}{\hbar}}\). The eigenvalues and eigenfunctions of the above equation were first obtained by McClure \[70\] while studying the diamagnetism of graphite.

gives the quantized transverse resistance observed in quantum Hall limit.
Chapter 2. Theoretical background

The solutions are given by,

$$E_{\pm}(n) = \pm \sqrt{2} \frac{\hbar v_F}{l_B} \sqrt{n} = \pm \sqrt{2eB\hbar v_Fn},$$  \hspace{1cm} (2.29)

and

$$\phi_{n, \pm}(\xi) = \begin{pmatrix} \psi_{n-1}(\xi) \\ \psi_n(\xi) \end{pmatrix},$$  \hspace{1cm} (2.30)

where $n = 0, 1, 2...$ and $\psi_n(\xi)$ are the solution of the 1-dimensional harmonic oscillator $\psi_n(\xi) = \frac{1}{\sqrt{2^{n}n!}} e^{-\frac{\xi^2}{2}} H_n(\xi)$ and $H_n(\xi)$ are Hermite polynomials. The positive sign is taken to describe the electrons in the conduction band and negative sign is taken to describe the holes in the valence band.

Several key points can be noted in the solution given by the Equation (2.29). We note that the linear density of states spectrum splits into non-uniformly spaced LLs since the position of the LLs is proportional to $\sqrt{n}$ (unlike the case of 2-dimensional free electron gas). As one moves to higher index $(n)$ levels, LLs starts coming closer to each other (for 2-D free electron gas, this gap remains constant $\hbar\omega_c$). Apart from 2-fold spin degeneracy ($g_s$), distinct wavefunction with same energies at $K'$-point give an additional 2-fold degeneracy (also called valley degeneracy $g_v$). The 4-fold degeneracy ($g_s \times g_v$) provide four independent channels for electron transport. Following Equation (2.25), one would expect that transverse resistance of graphene in quantum Hall limit will follow the sequence $\frac{h}{e^2 \nu}$, where $\nu = \pm 4, \pm 8, \pm 12...$. However, the interesting scenario arises due to the $n = 0$ LL, which is equally shared by electrons and holes between $K$ and $K'$-points. States below $\mu < 0$ are occupied by the holes whereas states above $\mu > 0$ are occupied by the electrons. Such a shared filling of zeroth LL shifts the overall quantized sequence to $\pm 2, \pm 6, \pm 10...$ without a plateau at zero filling. Stronger statement based on the Laughlin’s gauge invariance argument can also be made where it can be shown [14, 71] that due to the presence of zero mode shared between two Dirac points, there are exactly $(4n + 2)$ occupied states get transferred from one edge to the other when Fermi level cross the $n = 0$ LL. Therefore, for the case of monolayer graphene,

$$R_{xy} = \pm \frac{1}{4n + 2} \frac{\hbar}{e^2}. \hspace{1cm} (2.31)$$
2.7 Dynamics of nanoelectromechanical systems and simple harmonic oscillator

In this section, we will provide the theoretical framework to describe the graphene nanoelectromechanical systems (NEMS) \[7, 8\]. Though graphene is atomically thin, framework of continuum mechanics describes graphene NEMS quite well. We will start with Euler-Bernoulli equation commonly used to describe the NEMS \[72, 73\].

Let us consider a thin elastic beam, which is clamped at the two ends. For a transverse displacement \(Z(x,t)\) from equilibrium,

\[
\rho S \frac{\partial^2 Z}{\partial t^2} = -EI \frac{\partial^2 Z}{\partial x^4} + T \frac{\partial^2 Z}{\partial x^2}, \tag{2.32}
\]

where \(x\) is the coordinate along the length of the beam, \(\rho\) is mass density, \(S\) is cross-sectional area of the beam, \(E\) is Young’s modulus, \(I\) the moment of inertia, \(T\) is the total tension. For small displacements, the total length \(L\) can be written as

\[
L + \Delta L = \int_0^L dx \sqrt{1 + \left(\frac{dZ}{dx}\right)^2}, \tag{2.33}
\]

\[
\simeq L + \frac{1}{2} \int_0^L dx \left(\frac{dZ}{dx}\right)^2. \tag{2.34}
\]

Therefore, an extension \(\Delta L\) in the length of the beam induces additional tension, \(\Delta T\), (apart from any in-built tension \(T_0\)). Using Hooke’s law we can write,

\[
\Delta T = \frac{ES}{2L} \int_0^L dx \left(\frac{dZ}{dx}\right)^2. \tag{2.35}
\]

Using Equation (2.32) and (2.35), we get

\[
\rho S \frac{\partial^2 Z}{\partial t^2} = -EI \frac{\partial^2 Z}{\partial x^4} + \left[ T_0 + \frac{ES}{2L} \int_0^L dx \left(\frac{dZ}{dx}\right)^2 \right] \frac{\partial^2 Z}{\partial x^2}. \tag{2.36}
\]
Chapter 2. Theoretical background

This equation can be solved exactly in two limiting cases. In the limit, when flexural rigidity dominates over the in-built tension \( T_0 \), Equation (2.36) simply reduces to

\[
\rho S \frac{\partial^2 Z}{\partial t^2} = -EI \frac{\partial^2 Z}{\partial x^4}.
\]  (2.37)

The eigenfrequencies are given by [72],

\[
\omega_n = (k_n)^2 \sqrt{\frac{EI}{\rho S}},
\]  (2.38)

and the spatial amplitude profile \( \phi(x) \) for the eigenmodes is given by,

\[
\phi_n(x) \propto \frac{1}{a_n} \{(\sin k_n L - \sinh k_n L)(\cos k_n x - \cosh k_n x) \}
\]

\[
- (\cos k_n L - \cosh k_n L)(\sin k_n x - \sinh k_n x),
\]  (2.39)

\( k_n \)'s are the solutions of the transcendental equation \( \cos k_n L \cosh k_n L = 1 \). First few solutions are given by \( \{k_n L\} = \{4.73, 7.85, 11.0...\} \).

In the limit, when equilibrium tension dominates over the flexural rigidity, Equation (2.36) becomes,

\[
\rho S \frac{\partial^2 Z}{\partial t^2} = T \frac{\partial^2 Z}{\partial x^2}.
\]  (2.41)

The eigenfrequencies are given by,

\[
\omega_n = \frac{n\pi}{L} \sqrt{\frac{T}{\rho S}},
\]  (2.42)

and the spatial amplitude profile \( \phi(x) \) for the eigenmodes is given by,

\[
\phi_n(x) \propto \sin \left( \frac{n\pi x}{L} \right),
\]  (2.43)

where \( n = 1, 2, 3, ... \).

Typically, in real systems both flexural rigidity and tension play an impotent role and both terms on the r.h.s. of Equation (2.36) have to be considered. In such cases
2.7. Dynamics of nanoelectromechanical systems and simple harmonic oscillator

Equation (2.36) has to be solved self-consistently. The second order corrections to the fundamental modes solved above in the two limiting cases are given by [74],

$$\omega = \frac{22.4}{L^2} \sqrt{\frac{EI}{\rho S}} + 0.28 T \sqrt{\frac{1}{\rho SEI}}, \text{ when } \frac{EI}{L^2} \gg T \quad (2.44)$$

$$\omega = \frac{\pi}{L} \sqrt{\frac{T}{\rho S}} + \frac{2\pi}{L^2} \sqrt{\frac{EI}{\rho S}}, \text{ when } \frac{EI}{L^2} \ll T. \quad (2.45)$$

Another very useful approximation of Equation (2.36) is the simple harmonic oscillator, where continuous degree of freedom $Z$ gets mapped to the discrete single degree of freedom [73, 75, 76]

$$m \frac{d^2 z}{dt^2} + \Gamma \frac{dz}{dt} + m\omega^2_0 z + \alpha z^3 + \eta z^2 \frac{dz}{dt} = F, \quad (2.46)$$

where $m$ is the effective modal mass, $\Gamma$ is the linear damping rate, $\alpha$ is Duffing nonlinearity coefficient, $\eta$ is the coefficient of nonlinear damping and $F$ is the time dependent driving force. The modal mass of the resonator is different from the actual mass of the resonator and depends upon the modal shape of the resonant mode. $\Gamma$ is taken to describe the losses in the resonator. Cubic nonlinearity in the spring constant can either arise from the anharmonicity of the potential energy (electrical actuation schemes are often used for the NEMS, where nonlinear dependence of capacitance on the gap with another electrode can give rise to Duffing terms) or from the geometric elongation in the limit of amplitude being larger than the thickness of the beam [75]. Since graphene is just one atom thick, even small amplitude like few nanometer can make the Duffing term appreciable. The term $\eta z^2 \frac{dz}{dt}$ is included to describe the amplitude dependent damping. The microscopic origin of this term is not entirely understood.

The solution of Equation (2.46) for a Harmonic driving $F = F_0 \cos \omega t$ force can be obtained using secular perturbation theory [75, 77] and is given by $z(t) = z_0 \cos(\omega t + \phi)$, where
Chapter 2. Theoretical background

\[ z_0^2 = \frac{\left( \frac{F_0}{2m\omega_0} \right)^2}{\left( \frac{\omega - \omega_0}{\omega_0} - \frac{3\alpha z_0^2}{8m\omega_0} \right)^2 + \left( \frac{\Gamma}{2m\omega_0} + \frac{\eta z_0^2}{8m\omega_0} \right)^2} \]  

(2.47)

and

\[ \tan \phi = \frac{\frac{\Gamma}{2} + \frac{\eta z_0^2}{8}}{m\omega - m\omega_0 - \frac{3\alpha z_0^2}{8\omega_0}}. \]  

(2.48)

Several aspects about these solution can be noted immediately:

1) In the limit of the \( \alpha, \eta \to 0 \), we recover the solution of a driven damped harmonic oscillator. By using \( Q = \frac{m\omega_0}{\Gamma} \) we get,

\[ z_0 = \frac{1}{2m\omega_0} \frac{F_0}{\sqrt{(\omega - \omega_0)^2 + \left( \frac{\omega_0}{2Q} \right)^2}}, \text{ and } \tan \phi = \frac{\omega_0/2Q}{\omega - \omega_0}. \]  

(2.49)

Using the complex notations, the time dependent solution \( z(t) \) can be put in an elegant form as given below,

\[ z(t) = \frac{F_0}{2m\omega_0} \frac{e^{i\omega t}}{\omega - \omega_0 + i\frac{\omega_0}{2Q}}. \]  

(2.50)

The exact solution of Equation (2.46) in the limit \( \alpha, \eta \to 0 \) is given by,

\[ z(t) = \frac{F_0}{m} \frac{e^{i\omega t}}{\omega^2 - \omega_0^2 + i\frac{\omega_0}{Q}} \]  

(2.51)

which in the limit \( \omega \to \omega_0 \) reduces to Equation (2.50).

2) The term \( \alpha \) gives multiple real solutions in the frequency-response function making it bistable. Depending on the sweep direction of \( \omega \), amplitude \( z_0 \) can have different values. This means that at these bistable points, amplitude changes from one solution to the other solution curve abruptly with an infinitesimal increase in frequency; leading to the slope and curvature of driving frequency with respect to amplitude goes to zero. Therefore, in the limit of \( \eta \to 0 \), the condition for bi-stability
2.8. Summary

in the frequency response can be recovered by letting \( \frac{d\omega}{dz} = 0 \) and \( \frac{d\omega^2}{(dz)^2} = 0 \), given by

\[
F_0 = \left( \frac{4}{3} \right)^{5/4} \omega_0^3 \sqrt{\frac{m^3}{\alpha}}.
\]

3) The effect of nonlinear damping term \( \eta \) is to reduce the magnitude of the response when amplitude is large. It can be viewed with an effective damping rate set as \( \Gamma + \frac{1}{4} \eta z_0^2 \). In Figure 2.6(a) and (b), we have plotted the responsivity (normalized amplitude with driving force \( \frac{z}{F_0} \cdot \frac{m\omega_0^2}{\sqrt{Q}} \)) and phase as a function of normalized frequency \( \Omega \equiv \frac{\omega - \omega_0}{\omega_0} \) for different driving amplitudes while keeping \( \eta = 0 \) for the typical parameters of graphene resonators. With increased driving in the absence of \( \eta \), maximum responsivity remains the same. However response becomes bistable due to Duffing term \( \alpha \) and a hysteresis in the up and down sweep can be observed. Figure 2.6(c) and (d) are the plots of responsivity and phase with the same parameters as used in (a). The effect of \( \eta \) can be clearly seen on reduced responsivity as driving is increased. Another effect of the damping term can be seen as to reduce the hysteresis between the up and down sweep.

2.8 Summary

In this chapter we have described ideas essential to understand the electronic properties of graphene, like band structure calculations using tight-binding method, form of the density of states near Dirac point and working of the graphene field effect transistor. The core ideas of this thesis, quantum Hall effect and electromechanics of NEMS have been presented. A simplified quantum-mechanical picture of QHE is presented later. Afterwards, we discussed modal analysis of NEMS using Euler-Bernoulli equation and results from its approximations to a Duffing oscillator with nonlinear damping.
Figure 2.6: a, b) Plot of responsivity and phase respectively as a function of normalized frequency $\Omega \equiv \frac{\omega - \omega_0}{\omega_0}$ for a Duffing oscillator without nonlinear damping with increasing drive. c, d) Responsivity and phase plotted as a function of $\Omega$ for a Duffing oscillator in presence of nonlinear damping with increasing drive. At larger drives bistability in the response can be clearly seen. In the presence of nonlinear damping, responsivity reduces with drive. Also it reduces the hysteresis between the up and down sweep directions. For these plots typical parameters for graphene resonators are used: $\omega_0 = 2\pi \times 100$ MHz, $m = 3 \times 10^{-18}$ Kg, $Q = 1800$, $\alpha = 2.4 \times 10^{14}$ kgm$^{-2}$s$^{-2}$, $\eta = 10^5$ kgm$^{-2}$s$^{-1}$ and $F_0 = 7.5 \times 10^{-13}$ N. Red and blue curves represent up and down frequency sweeps, respectively.
Chapter 3

Device fabrication and measurement schemes

In this chapter, we will first describe the mechanical exfoliation technique for making graphene flakes, which is the primary step for making all the devices discussed in this thesis. After that, various lithography procedures are described to pattern the electrical contacts and graphene flake to make devices in Hall bar geometry. Next, we discuss fabrication of the suspended graphene devices for resonator and transport studies based on the two key processes—chemical wet etching of SiO$_2$ and critical point drying. We then describe various measurement schemes used in this work like schemes for non-equilibrium transport measurement on Hall bar device, actuation and detection schemes on suspended graphene devices for resonator measurements. Any adhered impurities on suspended graphene devices can be removed to improve the device quality by passing a large current between through it. We also discuss this technique, commonly known as current annealing.

3.1 Making graphene

In this section, we discuss the mechanical exfoliation technique for making graphene, also known as the ‘scotch-tape’ technique [4]. In this technique graphene sheets are literally pulled out from the graphite using a scotch-tape and then transferred
Chapter 3. Device fabrication and measurement schemes

to 300 nm SiO$_2$ coated silicon wafers. The process starts with the cleaning of the wafers to remove any residue adhered previously on substrate and to make the surface hydrophobic to enhance the transfer of hydrophobic graphene onto it. During the course of this work, we tried various processes for the cleaning of SiO$_2$ wafers like, by buffered hydrofluoric acid (BHF), by piranha solution (3 parts H$_2$SO$_4$ + 1 part H$_2$O$_2$) and by reactive ion etching the few nanometers of the top layer using O$_2$ plasma. From the yield of the thin flakes exfoliated using these three cleaning techniques, it is difficult to pick one method over the other. However, the relative ease of the RF plasma etching over the other two gives an upper hand for cleaning the wafer and modifying surface chemistry. These cleaned wafers are then used to transfer the graphene flakes onto them. The exfoliation of graphene flakes from graphite begins by repeatedly sandwiching the thin graphite crystal between the scotch tape till it becomes translucent. Snap shots of scotch-tape in these process are shown in Figure 3.1(a) and (b). Dabbing such tape on the SiO$_2$ wafer and peeling it off leads to the deposition of an assortment of flakes of different thicknesses on the surface. Examining the surface with a simple optical microscope allows one to sift through the debris and locate the very thin fakes of graphene. Figure 3.1(c) shows the optical microscope image with 20 X magnification from such a wafer. Figure 3.1(d) shows an optical microscope image of such a deposition with 50 X magnification. One can clearly see that there are several flakes with differing colors. The thickest flake at the bottom of the image has silver color, like a typical metal, and is very thick (> 500 nm), whereas ones that have a dark blue color are < 50 nm thick and the triangular flake (inside dotted area) that is barely visible is monolayer graphene.

We would like to emphasize the importance of thickness of SiO$_2$ in the wafers used for deposition. 300 nm thickness of SiO$_2$ gives the best contrast to see a monolayer flake for the light in visible range [78]. It sets up the right path difference between the light on and off graphene to produce maximum contrast. Though optical imaging quickly helps in sorting out thin flakes down to one layer, to confirm the thickness of any flake additional tests has to be performed. Raman spectroscopy and unique quantization of quantum Hall plateaus have evolved as “The” test for differentiating between mono-, bi- and tri- layer graphene flakes. For the work described in this thesis, our Hall measurements confirm the thickness of the flake conclusively. Since quantum Hall measurements are performed at low temperature and in large magnetic
3.1. Making graphene

Figure 3.1: a) A small and thin graphite crystal placed on scotch-tape. b) A translucent tape after repeatedly sandwiching and peeling off graphite between the tape. c) Optical microscope image of a deposition from such a tape on a 300 nm SiO$_2$ coated Si-wafer. The scale bar indicates 25 $\mu$m. d) A zoomed-in view: Flakes of varying colors can be seen reflecting the fact of varied thickness. Triangular portion of the flake at the center of the dotted region is a monolayer flake. The scale bar indicates 10 $\mu$m.
field, it may sound like a difficult means to find out the thickness of the flake used for device fabrication. At this point we would like to point that with experience one can identify a monolayer flake by looking at the optical images and also differentiate between a monolayer and bilayer graphene flake.

Once desired flakes are located in the optical microscope, their $X$-$Y$ coordinates relative to the corners of the wafer are noted down using the vernier scales attached the optical microscope stage. These relative positions are then used for the coarse alignment while exposing the markers (used for fine alignment while writing electrodes) using electron beam lithography.

### 3.2 Patterning contacts using electron beam lithography

The procedure of patterning metal electrodes to contact the graphene flakes involves several steps like spinning the resist, designing the electrodes using computer aided design, exposing the design using electron beam lithography machine, development of the exposed chip, metal deposition and liftoff (process to remove unwanted metal). First, the wafer is spin-coated with two layers of electron beam resist. First layer is coated by EL-9 resist (lower molecular weight resist with a formulation of copolymerized methyl-methacrylate (MAA) in ethyle lactate (EL) solvent), giving a thickness of $\sim$ 310 nm. For the second layer, PMMA-950 (higher molecular weight resist with a formulation of poly methyl-methacrylate (PMMA) dissolved in chlorobenzene) is used making the total resist thickness of $\sim$ 400 nm. Post exposure, the use of bi-layer resist with different molecular weights helps in forming an undercut after the development. For the fabrication of graphene devices such a high thickness of total resist is not essential requirement but larger resist thickness and an undercut in resist coating results in clean liftoff (process of removal of excess metals in the final step of fabrication).

In order to pattern electrodes, the resist coated wafer is exposed with an electron beam using electron beam lithography machine. We use Raith e-Line for all the fabrication discussed in this thesis. The electrode pattern is designed using computer
3.2. Patterning contacts using electron beam lithography

Figure 3.2: Various steps in the lithography process showing the exposure with electron beam, development, metal deposition and lift-off. Post development, bilayer resist coating helps in forming an undercut (inverted ‘V’-shape developed cross-section). This prevents a continuous metal deposition over the resist helping in a cleaner lift-off.
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aided design (CAD). During the exposure, in selected areas as per design, electrons deposit energy in the resist breaking the long polymer chains into smaller chains. In the next step, wafer is placed in developer solution for a specified time. The solubility of resist in the developer solution depends on the polymer chain length, therefore the exposed area of the resist dissolves much faster than the unexposed area of the resist. This results in the patterning of design into polymer resist. Next step is metal deposition to make electrical contacts. We have used 10-15 nm of chromium (Cr) and 60-80 nm of gold (Au) for making ohmic contacts in most of the devices studied. Metal can be deposited by sputtering or by evaporation. Both techniques give roughly the same contact resistance. However, evaporation is preferred since it gives sharper electrodes (line of sight deposition) compared to electrodes formed by the sputtering process (isotropic deposition) and also it is easy to use. After metal deposition, wafer is placed in acetone which removes the unexposed resist and metal deposited on top of it. At the end of it, we get only the desired metallic pattern as per the design. This sequence of steps is illustrated in Figure 3.2.

3.3 Devices in Hall bar geometry

As there is no control over the shape of graphene flakes in mechanical exfoliation technique, flakes prepared by this method have all possible irregular shapes. In order to make devices in well defined Hall bar geometry, unwanted portions of graphene can be removed by RF O\textsubscript{2} plasma technique. After patterning the metal electrodes, wafer is again spin coated with two layers of resist and unwanted parts of graphene are exposed using electrode beam lithography. After development, wafer is placed in O\textsubscript{2} plasma of pre-calibrated power and for a pre-calibrated time, leading to the removal of unwanted sections of graphene. We use 25 W of RF power for 30 sec in reactive ion etcher (Sentech ICP-RIE Plasma Etcher SI 500). The portion of graphene masked by the resist remain intact in this process and we get graphene device in desired shape after the removal of mask resist by acetone. Figure 3.3 shows pre and post etching optical microscope images from one such device.
3.3. Devices in Hall bar geometry

Figure 3.3: a) Optical micrograph of a device fabricated on an irregular flake. b) Image of the same device after removal of the unwanted portions of graphene by O$_2$ plasma making it in a regular Hall bar geometry. The scale bar corresponds to 3 µm in both the images.
3.4 Fabrication of suspended graphene devices

To make suspended graphene devices, one can use mechanical exfoliation technique directly on substrates with predefined trenches [28]. This method, however, is limited by the very nature of stochastic yield and poor electrical contacts. The other method is releasing the graphene flake from substrate by partial removal of SiO$_2$ [79 80]. The primary step to make suspended graphene devices by this method is to make on-substrate device. And then SiO$_2$ is etched in controlled manner using buffered hydrogen fluoride (BHF) to release the graphene from under the substrate. BHF etches the silicon dioxide isotropically without doing any harm to graphene or metal electrodes (Cr/Au) and even the presence of graphene flake does not affect the isotropic etch. It also etches the SiO$_2$ under the portions of gold electrodes overlapping with graphene and releases this portion of gold electrode from the substrate.

Before the etching with BHF, the wafer is first masked by resist and a window around the graphene flake (approximately 30 $\mu$m $\times$ 30 $\mu$m) is opened using electron beam lithography. This way only area near the graphene flake comes in contact with BHF and rest of wafer containing the wider electrodes and pads gets protected. The wafer is then placed in 1:5 diluted solution of BHF (Aldrich AF-875-125 Ammonium Fluoride etching mixture) for approximately 5 min 30 sec that results in a 170 nm deep trench in SiO$_2$. Dilute BHF is preferred to gain better control over the etch rate and uniformity. The device is then rinsed in deionized (DI) water to stop the etching. While moving from BHF solution to the DI water solution, wafer should always remain inside liquid, otherwise surface tension, during the drying process, between graphene and substrate can easily make the flake collapse on the substrate. To ensure this, we have used a teflon made spatula with a small pit in the center to place the chip in it. This way during the transfer of chip from one liquid to another, the suspended graphene device does not collapse. From DI water, chip is then moved to the container of acetone to remove the resist coating which is used for masking and then finally in isopropyl alcohol (IPA) for $\sim$ 2 minutes. In order to dry the chip for final use, a critical point dryer (CPD) is used. In this process, IPA is gradually replaced by liquid CO$_2$ under high pressure. Once all IPA is drained out and replaced by liquid CO$_2$, the fluid is moved to gas phase around the critical point (for CO$_2$, $T_c = 31.1^\circ$C and $P_c = 73.8$ Bar). By moving around the critical point, the CO$_2$ fluid
3.5. Quantum Hall measurement scheme

goes from liquid phase to gas phase without cutting the phase boundary in the phase space \((P-T)\) diagram at constant volume) implying no meniscus formation. Therefore, while drying the graphene flake does not experience any force due to surface tension.

Figure 3.4(a) shows the tilted scanning electron micrograph of a suspended graphene device with multiple leads connected to it. The suspended flake can be clearly seen held by the gold electrodes approximately 170 nm above the substrate. The boundaries of the resist mask window opened for BHF etch can be seen as well. As mentioned earlier BHF also etches the SiO\(_2\) under the portion of gold electrodes overlapping with graphene. To confirm this, we took a suspended graphene device, and mildly sonicated it to remove the graphene flake. Figure 3.4(b) shows a tilted scanning electron microscope image of such a three terminal device after sonication. The dashed outline shows the position of the suspended graphene flake before its removal by sonication. It can be seen from the image that during sonication, part of the gold electrodes covering the graphene flake before sonication has also been removed, revealing the etched SiO\(_2\) under the electrode covering graphene flake. This clearly shows that BHF etching removes the SiO\(_2\) uniformly under the graphene flake \([29]\). However, for metal electrodes that do not cover graphene a thin wall of SiO\(_2\) supports the electrodes. We would also like to point out some design rules while making the suspended graphene using this method. Since SiO\(_2\) gets etched under the gold electrode overlapping with graphene making them suspended, therefore, in devices where graphene flake is wider than \(\approx 8\) \(\mu\)m the yield of survival is very low (even with CPD) as the gold electrodes which hold the graphene above substrate collapse themselves. Another point is related to the CPD. In the last step of CPD, a fast vent of CO\(_2\) can lead to local cooling on the chip forming very small droplets of CO\(_2\) and this could lead to the collapse of the graphene flake.

3.5 Quantum Hall measurement scheme

Once devices are fabricated, we can perform electrical transport measurements on them. Figure 3.5 shows the schematic circuit diagram for the measurement of longitudinal and transverse resistance. One probe is used as source to inject constant current \((I_{SD} = 50\) nA, typically\). Any of the central two electrodes on one side can be
Figure 3.4:  a) Tilted scanning electron microscope (SEM) image of a suspended graphene flake connected with multiple electrodes held $\sim$170 nm above the substrate. The scale bar corresponds to 2 $\mu$m. b) Tilted SEM image of a suspended graphene device after removal of the graphene flake. The dashed outline shows the original position of the graphene flake. The scale bar corresponds to 1 $\mu$m.
3.5. Quantum Hall measurement scheme

![Circuit schematic for the measurement of longitudinal and transverse resistance for a device in Hall bar geometry.](image)

Figure 3.5: Circuit schematic for the measurement of longitudinal and transverse resistance for a device in Hall bar geometry.

used for the measurement of the longitudinal resistance \( R_{xx} = V_{xx}/I_{SD} \). Similarly, any central pair of probes on the opposite side can be used to measure the transverse resistance \( R_{xy} = V_{xy}/I_{SD} \).

In order to probe the non-equilibrium transport, a DC current along with the AC current can be injected in the device. While the DC current sets the non-equilibrium transport in the system, the AC current, on top of it, can be used to measure \( R_{xx} \) and \( R_{xy} \) using a lock-in technique. We have used a home-made voltage to current convertor, voltage adder (to add DC and AC signals) and SRS 830 lock-in for the measurements. The large dynamic range of the lock-in allows us measure AC component in the presence of a DC signal as well. This measurement scheme will be used in the experiment described in Chapter 4.
Chapter 3. Device fabrication and measurement schemes

3.6 Actuation and detection schemes for graphene nanoelectromechanical system

There are several methods for the actuation and detection of mechanical motion in nanoelectromechanical systems (NEMS) like magnetomotive [81], optical [82], piezoelectrical [83] etc. The choice of one particular technique depends on the system properties. Optical actuation and detection method for mechanical resonance has been used for graphene NEMS [28, 84]. However, such a technique is not easy to implement at low temperatures. In graphene, the tunability of conductance with gate voltage allows us to use electrical signals for actuation and detection. In the next few sections, we will describe the two variants of mixing technique used in this work, namely two source heterodyne mixing technique and frequency modulation (FM) technique. The mixing techniques rely on the finite transconductance ($\frac{dG}{dV_g}$) and the electrostatic interaction between resonator and gate electrode. To illustrate this, we can model our system as an infinite parallel plate capacitor, where one plate is fixed and other plate is attached to a spring as shown in Figure 3.6(a). When we apply a DC signal along with an AC signal between the plates, the electrostatic force can be written as,

$$F_{el} = -\frac{1}{2} C'_g (V_{dc}^g + \tilde{V}_g(t))^2$$  \hspace{1cm} (3.1)

where $C'_g = \frac{dC_g}{dz}$ is the derivative of capacitance with respect to the gap between the plates $(z)$, $\tilde{V}_g(t) = V_{ac}^g \sin \omega t$ and negative sign indicates the attractive nature of the force. Upon expanding, we get $F_{el} = \frac{1}{2} C'_g \left( (V_{dc}^g)^2 + 2V_{dc}^g V_{ac}^g \sin \omega t + (V_{ac}^g)^2 \sin^2 \omega t \right)$. The first component of force in this expression is the pure electrostatic force and sets the overall tension in the spring, the second component $C'_g V_{dc}^g V_{ac}^g \sin \omega t$ drives the plate at angular frequency $\omega$. The third component drive the system at frequency $2\omega$ and in the limit $V_{dc}^g \gg V_{ac}^g$ can be neglected. Therefore the driving force can be written as,

$$F_{drive} \simeq C'_g V_{dc}^g V_{ac}^g \sin \omega t.$$  \hspace{1cm} (3.2)
3.6. Actuation and detection schemes for graphene nanoelectromechanical system

Close to the natural frequency of the system, $F_{\text{drive}}$ leads to the large amplitude vibration in the system. Finite transconductance of the system in turn makes the conductance ($G$) to oscillate at frequency $\omega$. For typical device dimensions, these conductance oscillations occur at large frequency (10-100 MHz) and a direct readout of these high frequency signal is difficult due to large parasitic capacitance arising from the cables used in the low temperature measurement setup and from the contact pads fabricated on the wafer for making electrical connections.

To estimate the cut-off frequency, the device can be treated as low pass filter formed by the device resistance ($R_d \sim 10$ KΩ) and parasitic capacitance between the contact pads (source and drain electrodes) and gate electrode ($C_{\text{pads}} \sim 6$ pF). This gives a cutoff frequency $\frac{1}{2\pi R_d C_{\text{pads}}} \sim 2$ MHz, which is smaller than the typical resonance frequency of the devices. To circumvent this problem, the high frequency signal can be mixed down with another RF signal to make the measurement of readout signal easier. However, this compromises the rate at which measurement can be done. This will become clear in next sections.

3.6.1 Two source heterodyne mixing technique

In two source heterodyne mixing technique, a radio frequency (RF) signal $\tilde{V}_g(\omega)$ and a DC voltage $V_{gd}^{\text{dc}}$ are applied at the back gate terminal using a bias-tee. Another RF signal $\tilde{V}_{sd}(\omega + \Delta \omega)$ of slightly detuned frequency is applied to the source electrode. A detailed schematic of circuit is shown in Figure 3.6(b). At the source and gate terminals, we also add 50 Ω resistance in parallel with device for impedance matching with RF source. At the same time, a capacitor (10 nF) in series makes this path open for low frequency signals. Therefore, both DC transport and resonator based measurement can be done without modifying the circuit. While $V_{gd}^{\text{dc}}$ alters the overall tension and carrier density in the membrane, RF signal applied at the gate drives the flake with a force given by Equation (3.2) leading to the modulation in the gap between graphene and substrate at frequency $\omega$. This in turn causes modulation in conductance at frequency $\omega$. To mix down these conductance modulation, another RF signal of frequency $\omega + \Delta \omega$ is applied at the source terminal. The current through the device is given by $I = G(\omega)\tilde{V}_{sd}(\omega + \Delta \omega)$. Sinusoidal form of both quantities $i.e. G(\omega)$ and $\tilde{V}_{sd}(\omega + \Delta \omega)$ leads to frequency mixing. The product of these two
Figure 3.6: a) A toy model of parallel plate capacitor to understand the driving of resonator by AC signal. b) Circuit diagram for the two source heterodyne mixing technique (figure taken from Ref[85]). c) Schematic of the circuit diagram for frequency modulation technique.
3.6. Actuation and detection schemes for graphene nanoelectromechanical system

quantities produces current with two frequency components $2\omega + \Delta \omega$ and $\Delta \omega$. The higher frequency signal can be filtered out with the help of a low pass filter and low frequency component in current can be measured easily. At the mechanical resonance of the graphene membrane, modulation in $G(\omega)$ becomes largest and it reflects in the measurement of current at frequency $\Delta \omega$, also called mixing current ($I_{\text{mix}}(\Delta \omega)$). For the measurements described in this thesis we have typically used $\Delta \omega = 2\pi \times 13$ KHz.

3.6.2 Frequency modulation technique

Another variant of the mixing technique is the frequency modulation (FM) technique [87]. Figure 3.6(c) shows the measurement schematic for this technique. Here, only one frequency modulated RF signal is applied at the source end. A 50 $\Omega$ resistance in series with a capacitor ($\approx 10$ nF) is terminated parallel to the device due to the same reason as described in the previous section. The FM signal ($V_{sd}^{FM}(t)$) encodes the actuation and detection signal in itself and is given by,

$$V_{sd}^{FM}(t) = V_0 \sin \Omega(t), \quad \text{where} \quad \Omega(t) = \omega t + \frac{\omega_{\Delta}}{\omega_r} \sin \omega_r t, \quad (3.3)$$

$\omega$, $\omega_{\Delta}$ and $\omega_r$ are commonly referred to as carrier frequency, frequency deviation and modulation rate, respectively. From the form of Equation (3.3), it is clear that frequency of the RF signal is modulated in a range $\omega - \omega_{\Delta}$ to $\omega + \omega_{\Delta}$ at a rate of $\omega_r$. On time scales shorter than that of resonator time scale ($Q/\omega_0$), the resonator see a harmonic drive modulated by a slowly varying phase at frequency $\omega_r$. This in turn leads to the amplitude response to vary at frequency $\omega_r$. Therefore, measurement of the current component at frequency $\omega_r$ provides the information of the amplitude of the resonator. As shown in detailed calculations in next section, in fact, current component at frequency $\omega_r$ is proportional to the derivative of the real part of the amplitude [87].
Chapter 3. Device fabrication and measurement schemes

3.6.3 Characteristic equation for mixing current

After getting the basic working idea of the two techniques, in this section we derive the expression for the mixing current for these two techniques. We start with a generic treatment for current [87] and equation of mixing current for two cases, namely, two source heterodyne mixing technique and FM technique is recovered from it as special cases.

The current through the device \( I(V(t), z, V_g) \) at the drain end depends on the voltage applied at the source end \( V(t) \), total gate voltage \( V_g = \tilde{V}_g(t) + V_{g}^{dc} \) and the position of the flake \( z = z_0 + \delta z(t) \), where \( z_0 \) is the equilibrium position of the flake. Since we are interested in the current close to resonance frequency, we can do a Taylor series expansion around \( V_{sd} = 0, z = z_0 \) and \( V_g = V_g^{dc} \) retaining terms up to second order and we get,

\[
I(V(t), z_0 + \delta z(t), V_g) = I(0, z_0, V_g^{dc}) + \frac{\partial I}{\partial V_{sd}} V(t) + \frac{\partial I}{\partial z} \delta z(t) + \frac{\partial I}{\partial V_{g}^{dc}} \tilde{V}_g(t) + \frac{1}{2} \frac{\partial^2 I}{\partial V_{sd}^2} (V(t))^2 + \frac{1}{2} \frac{\partial^2 I}{\partial z^2} (\delta z(t))^2 + \frac{1}{4} \frac{\partial^2 I}{(\partial V_{g}^{dc})^2} (\tilde{V}_g(t))^2 \\
+ \frac{\partial^2 I}{\partial V_{sd} \partial z} V(t) \delta z(t) + \frac{\partial^2 I}{\partial z \partial V_g} \delta z(t) \tilde{V}_g(t) \\
+ \frac{\partial^2 I}{\partial V_{g}^{dc} \partial V_{sd}} \tilde{V}_g(t) V(t) + H.O. \tag{3.4}
\]

In the above equation all the derivatives are calculated at \( V_{sd} = 0, z = z_0, V_g = V_g^{dc} \). Now, we analyze each term appearing in Equation (3.4) to find the terms that give low frequency mixing current signal. The first term is zero since no current can flow in the absence of source drain bias \( (V_{sd} = 0) \). The second term is direct product of DC conductivity with RF signal and gives only high frequency component. The third and sixth terms are again zero since no displacement can cause the current to flow. The fourth term gives only high frequency current for two source technique and is essentially zero for FM technique. The fifth term involves \( (V(t))^2 \), which upon expansion gives a DC current and current with higher harmonics of frequency for both types of signal used in two source and FM techniques. Therefore this term can
be neglected too. Similarly, seventh term gives a DC signal and higher harmonics components in current for two source technique and zero for FM technique. Also ninth term gives higher harmonics components in current for two source technique and zero for FM technique. Therefore,

\[ I \approx \frac{\partial^2 I}{\partial V_{sd} \partial z} V(t) \delta z(t) + \frac{\partial^2 I}{\partial V_{dc} \partial V_{sd}} \bar{V}_g(t) V(t) \]

\[ = \left( \frac{\partial G}{\partial z} \delta z(t) + \frac{\partial G}{\partial V_{dc}} \bar{V}_g(t) \right) V(t). \quad (3.5) \]

The total charge on the resonator is given by \( q = C_g V_{dc}^g \). Using \( q \) at \( V_{sd} = 0 \), \( z = z_0 \), \( V_g = V_{dc}^g \), expression of current can be further simplified to,

\[ I = \left( \frac{\partial G}{\partial q} \frac{\partial q}{\partial z} \delta z(t) + \frac{\partial G}{\partial V_{dc}} \bar{V}_g(t) \right) V(t) \]

\[ = \frac{\partial G}{\partial V_{dc}} \left( C_g \frac{V_{dc}^g}{C_g} \delta z(t) + \bar{V}_g(t) \right) V(t) \quad (3.6) \]

The above equation contains the low frequency component in current arising from the mixing of the electrical signals to the mechanical motion. Now, we will take particular cases of two source technique and FM technique to derive the explicit formulae of mixing current.

**(a) Two source heterodyne mixing technique:**

In the heterodyne mixing technique, we apply \( V(t) = V_{sd}^{ac} \sin(\omega + \Delta \omega) t \) at the source contact and \( \bar{V}_g(t) = V_{dc}^{ac} \sin \omega t \) at the gate contact. The only remaining term in Equation (3.6) is the time dependence of the amplitude \( \delta z(t) \). The resonator can be approximated to a driven simple harmonic oscillator with single degree of freedom, solved in section 2.7. Therefore, using the real part of Equation (2.51) we can write,
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\[ I = \frac{dG}{dV_{dc}} \left( C_g' V_{dc} z_0 \frac{\sin(\omega t + \phi)}{Q} \left( 1 - \left( \frac{\omega}{\omega_0} \right)^2 \right)^2 + \left( \frac{\omega}{\omega_0} \right)^2 \right) \frac{V_{ac}^{ac} \sin \omega t}{V_{sd}^{ac} \sin(\omega + \Delta \omega) t} \]

(3.7)

where, \( z_0 \) is the maximum amplitude of vibration, \( Q \) is the quality factor of the resonator and \( \phi \) is given by, \( \phi = \arctan \left( \frac{\omega_0/Q}{\omega^2 - \omega_0^2} \right) \). Since we are interested in the current at frequency \( \Delta \omega \), upon expanding Equation (3.7) and collecting terms with \( \Delta \omega \) dependence we get,

\[ I_{mix}(\Delta \omega) = \frac{1}{2} \frac{dG}{dV_{dc}} \left( C_g' V_{dc} z_0 \frac{\cos \left( \arctan \left( \frac{\omega_0/Q}{\omega^2 - \omega_0^2} \right) \right)}{Q} \left( 1 - \left( \frac{\omega}{\omega_0} \right)^2 \right)^2 + \left( \frac{\omega}{\omega_0} \right)^2 \right) \frac{V_{ac}^{ac}}{V_{sd}^{ac}}. \]

At this point, we would like to note that in the derivation of above equation, we have added two currents (see Equation (3.6)) with different physical origin. The first term inside the bracket is due to the change in potential of the graphene flake due to mechanical motion and is significant only at the resonance frequency. The second term inside the bracket is due to the change in potential of the flake induced by RF signal applied at back gate. However, if we look at the time dependence of these two terms, we find that first term, due to mechanical motion, remains in phase with the mechanical motion of graphene flake while second term remains in phase with the voltages applied at source and gate electrode. And this phase difference between the two terms depends on the contact resistance and capacitance of the system. Therefore, while calculating the total mixing current, one should add both the terms with a device dependent phase factor (\( \Phi \)). Depending upon this phase difference between the two currents, the shape of the resonance curve can be anything from a peak to a dip or something in between like a heart beat shape [86, 88]. Taking this into account, the final expression of mixing current can be written as,
3.6. Actuation and detection schemes for graphene nanoelectromechanical system

\[ I_{\text{mix}}(\Delta \omega) = \frac{1}{2} \frac{dG}{dV_{\text{sd}}^\text{dc}} \left( C'_g \frac{V^\text{dc}_{\text{g}} z_0}{C_g Q} \cos \left( \Phi + \arctan \left( \frac{\omega_0/Q}{\omega^2 - \omega_0^2} \right) \right) \right) \left( 1 - \left( \frac{\omega}{\omega_0} \right)^2 \right)^2 + \left( \frac{\omega/\omega_0}{Q} \right)^2 + V_{\text{sd}}^\text{ac} \right) V_{\text{sd}}^\text{ac}. \]  

(3.8)

Above equation can be used to fit the experimental data for the extraction of resonant frequency \( f_0 \) and quality factor \( Q \).

(b) Frequency modulation (FM) technique:

In FM technique, we apply a frequency modulated RF signal at the source end, given by the Equation (3.3). Since no RF signal is applied at the back gate, therefore the second term inside the bracket in Equation (3.6) does not appear. Hence we get,

\[ I = \frac{\partial G}{\partial V_{\text{dc}}^\text{g}} \left( C'_g \frac{V^\text{dc}_{\text{g}}}{C_g} \right) \delta z(t) V(t). \]  

(3.9)

Before we start calculating low frequency components in the above equation, the FM signal can be decomposed in to its spectral components to gain better insight.

\[ V_{\text{sd}}^{\text{FM}}(t) = V_0 \sin \left( \omega t + \frac{\omega_\Delta}{\omega_r} \sin \omega_r t \right) \]
\[ = V_0 \left( \sin \omega t \cos \left( \frac{\omega_\Delta}{\omega_r} \sin \omega_r t \right) + \cos \omega t \sin \left( \frac{\omega_\Delta}{\omega_r} \sin \omega_r t \right) \right). \]  

(3.10)

Using the Jacobi-Anger expansion given by,
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\[
\sin(z \sin \theta) = 2 \sum_{n=1}^{\infty} J_{2n-1}(z) \sin((2n-1)\theta)
\]

\[
\cos(z \sin \theta) = J_0(z) + 2 \sum_{n=1}^{\infty} J_{2n}(z) \cos(2n\theta)
\]  

(3.11)

the FM signal can be written as,

\[
V_{sd}^{FM}(t) = V_0 \sin \omega t \left( J_0(\omega \Delta \omega_r) + 2 \sum_{n=1}^{\infty} J_{2n}(\omega \Delta \omega_r) \cos\{2n\omega_r t\} \right) \\
+ V_0 \cos \omega t \left( 2 \sum_{n=1}^{\infty} J_{2n-1}(\omega \Delta \omega_r) \sin\{(2n-1)\omega_r t\} \right) \\
= J_0(\omega \Delta \omega_r) V_0 \sin \omega t + 2V_0 \sum_{n=even}^{\infty} J_n(\omega \Delta \omega_r) \sin \omega t \cos n\omega_r t \\
+ 2V_0 \sum_{n=odd}^{\infty} J_n(\omega \Delta \omega_r) \cos \omega t \sin n\omega_r t \\
= J_0(\omega \Delta \omega_r) V_0 \sin \omega t + V_0 \sum_{n=even}^{\infty} J_n(\omega \Delta \omega_r) (\sin(\omega + n\omega_r)t + \sin(\omega - n\omega_r)t) \\
+ V_0 \sum_{n=odd}^{\infty} J_n(\omega \Delta \omega_r) (\sin(\omega + n\omega_r)t - \sin(\omega - n\omega_r)t) \\
= J_0(\omega \Delta \omega_r) V_0 \sin \omega t + V_0 \sum_{n=1}^{\infty} J_n(\omega \Delta \omega_r) (\sin(\omega + n\omega_r)t + (-1)^n \sin(\omega - n\omega_r)t) .
\]  

(3.12)

Upon decomposing the FM signal into its spectral components, we can see that for \(V^2(t)\) no term proportional to \(\cos(\omega_r t)\) or \(\sin(\omega_r t)\) arises. This justifies our reasoning for ignoring fifth term in Equation (3.4). In order to further simplify the expression for the calculation of the current, we expand the argument of FM signal in time scales shorter than \(Q/\omega_0\).
3.6. Actuation and detection schemes for graphene nanoelectromechanical system

\[ \Omega(t + \Delta t) = \Omega(t) + \Delta t \frac{\partial \Omega}{\partial t} + H.O. \]  

(3.13)

where, \( \frac{\partial \Omega}{\partial t} = \omega + \omega_\Delta \cos \omega_r t \equiv \omega_i \). In the above expansion, all the higher order terms can be neglected for \( \omega \gg \omega_\Delta \) and \( \omega_r \ll \omega_0/Q \), which is easy to satisfy in experiments. Within these constraints, we can say that for time scales shorter than the resonator time \( Q/\omega_0 \), the resonator sees a harmonic force modulated by slow varying phase given by \( \omega + \omega_\Delta \cos \omega_r t \).

Therefore, at any instant \( t + \Delta t \), the current can be written as,

\[ I = \frac{\partial G}{\partial V_{dc}} \left( C'_g \frac{V_{dc}}{C_g} \right) \delta z(t + \Delta t) V_0 \sin(\Omega(t) + \omega_i \Delta t). \]  

(3.14)

Using the complex notation of the amplitude given by Equation (2.51), the displacement of the resonator at \( t + \Delta t \) can be written as,

\[ \delta z(t + \Delta t) = \text{Re}[\delta z^*(\omega)] \sin(\Omega(t) + \omega_i \Delta t) + \text{Im}[\delta z^*(\omega)] \cos(\Omega(t) + \omega_i \Delta t). \]  

(3.15)

Using Equation (3.14) and (3.15), we can write

\[ I = \frac{dG}{dV_{dc}} \left( C'_g \frac{V_{dc}}{C_g} \right) V_0 (\text{Re}[\delta z^*(\omega)] \sin^2(\Omega(t) + \omega_i \Delta t) \\
+ \text{Im}[\delta z^*(\omega)] \sin(\Omega(t) + \omega_i \Delta t) \cos(\Omega(t) + \omega_i \Delta t)). \]

The low frequency component (\( \omega_r \) or the slowly modulating phase) in the above equation arise from the first term inside the bracket (\( \sin^2 \theta = \frac{1}{2} (1 - \cos 2\theta) \)). Remaining terms gives high frequency components only. Therefore the mixing current
Chapter 3. Device fabrication and measurement schemes

at frequency $\omega_r$ can be written as,

$$I_{mix}(\omega_r) = \frac{1}{2} \frac{dG}{dV_g} \left( C_g \frac{V_{dc}}{C_g} \right) V_0 \text{Re}[\delta z^*(\omega)]. \quad (3.16)$$

Upon performing a Taylor series expansion of $\text{Re}[z^*(\omega_i)]$ for $\omega_\triangle \ll \omega$, we get

$$\text{Re}[\delta z^*(\omega_i)] = \text{Re}[\delta z^*(\omega)] + \left( \frac{\text{Re}[\delta z^*]}{\partial \omega_i} \right)_{\omega_i=\omega} \omega_\triangle \cos \omega_r t + \text{H.O.} \quad (3.17)$$

Here we observe that we get only terms with $\cos \omega_r t$, no term with $\sin \omega_r t$ or any other additional background terms. Therefore, in the detection of mixing current in FM technique imaginary component of the current and mixing current away from the resonance, both remain zero. Combining Equation (3.16) and (3.18), we can write

$$I_{mix}(\omega_r) = \frac{1}{2} \frac{dG}{dV_g} \left( C_g \frac{V_{dc}}{C_g} \right) V_0 \omega_\triangle \left( \frac{\text{Re}[\delta z^*]}{\partial \omega_i} \right)_{\omega_i=\omega} \quad (3.18)$$

Using the complex notation of amplitude (Equation (2.51))

$$z^*[\omega_i] = z_0 \frac{\omega_0^2}{Q} \frac{1}{\omega_0^2 - \omega_i^2 + i \left( \frac{\omega_0 \omega_i}{Q} \right)}$$

we can write,

$$\left( \frac{\text{Re}[\delta z^*]}{\partial \omega_i} \right)_{\omega_i=\omega} = \left( \frac{2z_0 \omega_0^2 \omega}{Q} \right) \left( \frac{(\omega^2 - \omega_0^2 + \frac{\omega_0^2}{Q})(\omega^2 - \omega_0^2 - \frac{\omega_0^2}{Q})}{((\omega_0^2 - \omega^2)^2 + (\frac{\omega_0 \omega_i}{Q})^2)^2} \right). \quad (3.19)$$
3.7 Current annealing to improve charge carrier mobility

Therefore a final expression for the mixing current can be written as,

\[ I_{mix}(\omega_r) = B\omega \left( \frac{(\omega^2 - \omega_0^2 + \frac{\omega_0^2}{Q})(\omega^2 - \omega_0^2 - \frac{\omega_0^2}{Q})}{(\omega_0^2 - \omega^2)^2 + \left(\frac{\omega_0}{Q}\right)^2} \right)^2 \]  \hspace{1cm} (3.20)

where \( B = \frac{dG}{dV_{dc}} \left( \frac{C_g'}{C_g} \right) (V_{dc}V_0) \left( \frac{z_0\omega_0^2}{Q} \right) \omega_\Delta. \)

This completes the discussion about the heterodyne mixing techniques used for the actuation and detection of mechanical motion of the NEMS.

3.7 Current annealing to improve charge carrier mobility

We now discuss how these suspended graphene devices can be cleaned to achieve very high mobility. The process is very simple and involves heating of graphene by injecting a large current density (\( \approx 1\text{mA/\mu m} \)) [79, 80, 89]. Joule heating due to large dissipation locally increase the temperature of graphene, leading to the evaporation of any adhered impurities on graphene flake [89]. The removal of impurities also changes the doping concentration and hence the resistance of the device changes in time while injecting the large current.

Figure 3.7(a) shows the variation of zero bias conductance of graphene device with gate voltage right after fabrication (top curve). The device after fabrication has no clear Dirac peak (or charge neutrality point) and poor field effect mobility. To perform the current annealing, we slowly ramp the source-drain voltage up in steps of 50-100 mV and observe the current for 2-3 minutes. If no change in current is observed with time, the applied voltage is increased to the next step. However, if any change in current (\( \sim 1\mu A/2 \text{ sec} \)) is observed, we wait till it stabilizes (3-5 minutes). For a faster response (\( > 1\mu A/2 \text{ sec} \)) in current change, we reduce the source drain voltage. We use the same process for both polarities of source-drain
Figure 3.7: a) Plot of resistance with gate voltage during different stages of current annealing of the graphene device by sending bipolar current. Initially the graphene device does not show the Dirac peak and has overall high resistance. Gradual improvement in the sample quality can be seen with additional annealing steps. b) Plot of current and voltage as the suspended graphene device is biased. The abrupt jumps at the extreme voltage bias indicates the resistance change in time while injecting the large current through the device. With removal of impurity doping concentration changes leading to a change in resistance and hence a jump in I-V curve.
voltage in tandem. To optimize the cleaning process the gate response is checked after every cleaning step. The devices are annealed at cryogenic temperatures in cryogenic vacuum, though low temperature is not essential for this process since we are going to heat the device anyway, but a cryogenic vacuum certainly helps in faster removal of evaporated contamination. Figure 3.7(a) shows the result of various repetitions of current annealing procedure on the same device and gradually one observes a sharp Dirac peak as a function of gate voltage. I-V plots at large are shown in Figure 3.7(b). The sudden drops in the current are due to drop in current after some time. At the end of the last annealing step the mobility of the suspended is in the excess of 150,000 cm²V⁻¹s⁻¹ shown in Figure 3.7(a).

3.8 Summary

In this chapter, we described the key processes involved in device fabrication in a comprehensive manner including mechanical exfoliation, electron beam lithography, wet chemical etching and critical point drying. We also described the mixing technique for the actuation and detection of the mechanical response in suspended graphene devices and derived the expression for the mixing current. We also provide details of the current annealing process which is used to improve the charge carrier mobilities in suspended graphene devices.
Chapter 4

Non-equilibrium breakdown of quantum Hall state in graphene

In this chapter, we describe the measurements on the non-equilibrium breakdown of the quantum Hall effect (QHE). We begin by providing details about the device characterization for such a study. Afterwards we provide experimental results of breakdown of the QHE by injecting a high current density ($\sim 1\text{A/m}$). Measurements of the critical current for breakdown of the QHE indicate towards the inter Landau level scattering mechanism to be the dominant one. We also observe current invariant point in transverse conductance between the plateau to plateau transition and provide a quantitative analysis based on the current injection model [90]. The results presented here have been published in Ref[91].

4.1 Quantum Hall state under non-equilibrium

As described in Chapter 2, in quantum Hall limit, when Fermi energy lies between two Landau levels, the longitudinal resistance ($R_{xx}$) vanishes leading to dissipationless transport and transverse resistance ($R_{xy}$) gets quantized to $\frac{\hbar}{(4|n|+2)e^2}$, where $n$ is the Landau level index. However, the description of zero resistance is valid only at absolute zero temperature and zero bias condition. At finite temperature the resistance remains exponentially small. The quantum Hall state can also be driven out of
the equilibrium, by applying a finite bias \( i.e. \) injecting finite current leading to the breakdown of the dissipationless transport. The motivation for exploring the breakdown of QHE in graphene is twofold – first, the QHE in graphene is very different from the QHE in a 2 dimensional electron gas (2DEG). The LL energy spectrum of 2DEG is equispaced unlike that in graphene. The energy scale set by the cyclotron gap \((\Delta E_{\nu})\) in graphene at \( B = 10 \) T, is much higher \((\sim 1300 \) K\) than its value for a 2DEG \((\sim 20 \) K\) at the same magnetic field \([14]\). The mechanism of breakdown in graphene could be the inter LL scattering due to wavefunction-mixing or possibly entirely different if the lengthscale for variation of the local electric field due to defects is comparable to the magnetic length. In such a situation, a “collapse” of the LL is possible before a longer lengthscale breakdown of QHE \([92]\). Second, graphene shows room temperature QHE \([93]\) at high magnetic field, therefore understanding the breakdown mechanism can also be useful for room temperature metrological resistance standards \([94]\). With these motivations in mind we have probed the breakdown of QHE near the filling factors \( (\text{sgn}(n)(4|n| + 2)) \) \([12, 13]\), \( \nu = -10, -6, -2, 2, 6 \).

### 4.2 Device characterization and measurement scheme

The presence of back gate in our devices allows us to change the Fermi level by tuning the carrier density in the flake. The basic characterization of the device start with the measurement of 4-probe resistance as a function of gate voltage which is proportional to the density of the induced charge carriers. The inset of Figure 4.1(a) shows an optical image of a Hall bar device with labeled electrodes. Figure 4.1(a) shows the resistance of the device with gate voltage. The maxima in resistance occurs at \( V_g \sim 13 \) V indicating the position of Dirac peak. The unintentional doping during the fabrication can move the Dirac point away from zero gate voltage. From the estimate of half width of the Dirac peak, this corresponds to a charge inhomogeneity of \( 6 \times 10^{-11} \text{cm}^{-2} \) \([95]\). On both sides of the Dirac point, we change the gate voltage, resistance decreases due to increased number of carriers. Within the linear response regime, following Equation (2.19), we can estimate the field effect mobility of the charge carriers, which comes out to be \( \sim 11000 \) \( \text{cm}^2/(\text{Vs})^{-1} \) for both types of carriers at 300 mK. Using the semi-classical relation for mean free path \( l \) \([95]\), the measured mobility gives \( l \sim 70 \) nm for carrier density \( 3 \times 10^{-11} \) \( \text{cm}^{-2} \).
Figure 4.1: a) Measurement of 4-probe resistance with gate voltage at $T = 300$ mK. Inset shows the optical microscope image of a Hall bar device with labeled electrode. The scale bar corresponds to 6 µm. Probe-1 and probe-4 are used for current biasing while probe-2 and probe-3 are used for voltage measurement. b) Resistance measured in different probe configurations to check the inhomogeneity in the sample at 1.5 K.
4.2. Device characterization and measurement scheme

For the measurement of 4-probe resistance, voltage probes are assumed to be non-invasive and sample to be homogeneous. However, the presence of metal electrodes and unintentional doping during fabrication can affect the measurement of resistance\[96\]. To check this, we performed measurement of resistance with different current leads and voltage leads configurations. We define $R_{mn-pq}$ as $\frac{V_{mn}}{I_{pq}}$; voltage is measured between probes $m$ and $n$ while current is sent from probe $p$ to $q$. Figure 4.1(b) shows the measurement of resistance in four different configuration at 1.7 K. In all configuration of measurements, the Dirac peak occurs almost at the same gate voltage. It is quite evident from this measurement that unintentional dopants are quite uniformly distributed over a length scale set by the gap between the electrodes.

After the basic characterization of device at low temperatures, we perform measurements in magnetic field applied perpendicular to the plane of graphene. Using probe-2 and probe-3, we measured longitudinal resistance ($R_{xx}$), while probe-2 and probe-6 can be used for the measurement of transverse resistance ($R_{xy}$). In Figure 4.2(a), we plot $R_{xx}$ and $R_{xy}$ with gate voltage at $T = 300$ mK and $B = 9$ T. At a fixed magnetic field, by changing the gate voltage the Fermi energy can be moved across the Landau levels without modifying the energy spectrum. As gate voltage is swept vanishing $R_{xx}$ and quantized plateaus in $R_{xy}$ can be seen clearly. These filling factors correspond to monolayer graphene ($\nu = \pm 2, \nu = \pm 6$). In Figure 4.2(b), we plot the evolution of $R_{xy}$ as a function of $V_g$ and magnetic field $B$. As carrier density and magnetic field are changed, the integer filling factors evolve and it can be seen from the fan diagram shown in Figure 4.2(b). The plateaus in $R_{xy}$ corresponding to $\nu = \pm 2, \pm 6$ and $-10$ are clearly seen.

Although the device shows nicely developed quantum Hall plateaus and vanishing resistance, more information about the contacts can be gained from the two probe resistance measurement in quantum Hall limit. In Figure 4.3, we show two probe resistance measurement between three different pairs of voltage probes at $T = 1.7$ K and $B = 8$ T from a different device. Here we can see that quantum Hall plateaus are shifted from the exact quantized values expected. Such a shift in resistance arises from two sources. First, in a two probe resistance measurement contact resistance and line resistance are inherently present. Secondly, due to finite temperature, a finite microscopic longitudinal resistivity ($\rho_{xx}$) can modify the resistance measured using two probe method from the exact quantization [97]. Such an effect strongly depends
Chapter 4. Non-equilibrium breakdown of quantum Hall state in graphene

Figure 4.2: a) Plot of the longitudinal resistance ($R_{xx}$) and transverse resistance ($R_{xy}$) for a monolayer graphene device at 300 mK and 9 T clearly indicating well resolved Landau levels and well developed quantum Hall plateaus. b) Colorscale plot of $R_{xy}$ to show the plateaus of varying magnitude clearly as a function of magnetic field at 300 mK.
4.3. Experimental results and discussion

upon the aspect ratio between the probes used for two probe measurement. This can be seen clearly in the stark difference in resistance behavior close to Dirac point between $R_{23}$ and $R_{26}$. For the measurement of $R_{26}$, the large width of the sample over the gap between probes leads to a smaller contribution of $\rho_{xx}$ to the total resistance and hence, a dip in resistance at the Dirac point. The difference in resistance value at the plateau, therefore, can be used as an upper bound for the contact resistance in quantum Hall limit. This we find to be smaller than 700 $\Omega$ for each probe in quantum Hall limit.

To probe the breakdown of QHE, we biased the source-drain probes (probe-1 and probe-4) of our device with DC current ($I_{SD}^{DC}$) along with a small AC current (50 nA) in the minima of $R_{xx}$ corresponding to filling factors $\nu = \pm 2, \pm 6$ and $-10$ at fixed magnetic field. The AC current remains fixed and $I_{SD}^{DC}$ is then varied as a function of $V_g$ in the vicinity of integer $\nu$. The AC signals between the voltage probes-2 and probe-3 and the voltage probe-2 and probe-6 were monitored with a lock-in amplifier to record the values of $R_{xx}$ and $R_{xy}$, respectively. Filling factor $\nu = 10$ is not studied as the gate voltage required to study it can lead to a dielectric breakdown of the gate-dielectric SiO$_2$.

4.3 Experimental results and discussion

Figure 4.4 shows the evolution of the $R_{xx}$ minima as function of $I_{SD}^{DC}$ for different filling factors. The line plots show slices of the data in equilibrium and non-equilibrium biasing conditions. In order to interpret the breakdown from the measured experimental data we define a critical current ($I_{SD}^{crit}$) as the linearly extrapolated value of $I_{SD}^{DC}$ at zero dissipation [98].

We point out the qualitative features of our data – first, with the increase of $I_{SD}^{DC}$, the width of the dissipationless region reduces (represented by white dotted lines overlayed on the colorplots), eventually leading to the breakdown. Second, the critical current is $\nu$ dependent. Third, on either side of the integer filling factor the boundary of dissipation evolves asymmetrically. In Figure 4.4a, b, c, d and e for $\nu = 6, 2, -2, -6$ and $-10$ respectively we see a non-linear evolution of the boundary of dissipation as a function of $I_{SD}^{DC}$ and $V_g$. Before discussing details of the data, we address the
Figure 4.3: Plot of two probe resistance measured for different probe combinations at 1.7 K and 8 T.
Figure 4.4: Critical current measurements in the vicinity of integer filling factors at 9 T and 300 mK. a, b, c, d and e show the colorscale plot of $R_{xx}$ as a function of $I_{SD}^{DC}$ and $V_g$ near filling factors 6, 2, -2, -6, and -10 respectively. The white dotted lines on the colorscale plot mark the dissipationless region. The line plots in f, g, h, i and j show slices along the current axis at the gate voltages shown in the figure (top) and two slices (bottom) each for the equilibrium (labeled with solid circles) and non-equilibrium (solid line) biasing conditions. The position of these slices is marked in adjoining colorscale plots (marked a, c, e, g and i ).
possible concerns about local heating of the sample that could occur in these studies. We have done control experiments to confirm that heating is not responsible for the key experimental observations; by comparing change of resistance with temperature and due to current. We injected a large DC current through the voltage probes-2 and probe-3 of the Hall bar geometry device to intentionally heat it locally while we simultaneously measured two probe resistance-gate voltage characteristic of the opposite two probe-5 and probe-6 using a lock-in technique. Then, we observed the evolution of two probe resistance-gate voltage characteristic of the same pair (probe-5 and probe-6) with temperature while the injected DC current was set to zero. By comparing these two data sets (change of resistance with temperature and due to current), we could estimate that the temperature of the device does not increase beyond \( \approx 3 \) K while injecting currents as high as \( 10 \) \( \mu \)A. We have also seen an overall asymmetry in the evolution of \( R_{xx} \) with the sign of injected current close to integer filling factor. Pure thermal effects cannot explain this asymmetry. Additionally, cyclotron gaps in graphene are large and thermal effects cannot completely suppress the electric field induced effect. Also, superior thermal conductivity of graphene \[99\] is likely to suppress any local thermal hot-spot formation.

In order to understand the mechanism of the breakdown we examine the dependence of \( I_{SD}^{SD} \) on \( \nu \). Figure 4.5(a) shows the plot of \( I_{SD}^{SD} \) for various filling factors \( \nu \) indicating that \( I_{SD}^{SD} \) decreases with \( |\nu| \). As we probe the higher \( \nu \), the cyclotron gaps \( (\Delta E_\nu) \) reduce and excitations across the Landau level can be caused at lower energy. This explains qualitatively the dependence of critical current on \( \nu \). Figure 4.5(b) shows the plot of two relevant quantities, Hall voltage, \( V_{Hall} = I_{SD}^{SD} \times \frac{\hbar}{\nu e^2} \), and \( \Delta E_\nu \) as a function of \( \nu \). There is a correlation between \( V_{Hall} \) and \( \Delta E_\nu \), which can be explained by considering inter LL scattering. The origin of the inter LL scattering is likely to be the strong local electric field that mixes the electron and hole wavefunctions \[90, 92, 100\] providing a finite rate for inelastic transitions. The energy for the transitions is provided by the transverse electric field parallel to the electron trajectory. This results in inelastic scattering between LLs leading to a breakdown of the dissipationless QH state \[23\]. Similar inter LL scattering mechanisms have been used to explain the breakdown of the QHE in a 2DEG, in samples of smaller widths \( (< 10 \) \( \mu \)m) and moderate mobility \[18, 23, 98\]. The electric field for inter LL \( (E_{LL}) \) scattering can be estimated to be that field where the quasiparticle can
4.3. Experimental results and discussion

Figure 4.5: a) Plot of critical current ($I_{SD}^{crit}$) for different filling factors at $T = 300$ mK and $B = 9$ T. b) Plot of Hall voltage developed at breakdown ($V_{Hall}$, labeled with circles) and the cyclotron gaps ($\Delta E_{\nu}$) (solid triangles) plotted on right axis as a function of $\nu$. 
pick up an energy corresponding to the LL separation within few cyclotron radii \( r_c \) i.e. \( eE_{LL} \sim \Delta E_\nu / r_c \approx 10^6 \text{ V/m} \). This is much higher than the experimentally observed electric fields. However, Martin et al. [101] found a much shorter lengthscale associated with the charge inhomogeneity (~150 nm). The presence of a charge inhomogeneity [101] leads to a strong local electric field and thus can reduce the threshold for the breakdown due to inter LL scattering.

For \( \nu = \pm 2 \), \( V_{Hall} \) matches quite well with \( \Delta E_\nu=\pm 2 \), which indicates that the \( n = 0 \) LL width is small. However for \( \nu = \pm 6, -10 \), \( V_{Hall} \) is smaller than the corresponding cyclotron gap. This deviation for \( \nu = \pm 6, -10 \) can be explained by considering disorder-induced broadening of \( n = \pm 1, -2 \) LLs. The difference between \( \Delta E_\nu \) and \( eV_{Hall} \) is approximately \( \sim 35 \text{ meV} (\sim 415 \text{ K}) \). These observations are consistent with the experiments measuring quantum Hall activation gap [102], which have also revealed similar width of these LLs in samples of similar mobilities. Additionally, the difference between \( V_{Hall} \) and \( \Delta E_\nu \) can also be attributed to inhomogeneous charge distribution. Considering inhomogeneous charge distribution, the critical current is predicted to be filling factor and length scale dependent [92]. However, the \( n = 0 \) level remains protected from the local electric field fluctuations [92, 103]. The correlation between \( V_{Hall} \) and \( \Delta E_\nu \) shows consistency with the breakdown mechanism based on this picture too. In addition, experimental finding that there is a non-linear evolution of dissipation boundary can possibly be attributed to Hall field induced broadening of the extended state band.

### 4.4 Effect on plateau to plateau transition

To further explore the effect of transverse electric field, due to the high current density, we look at the plateau to plateau transition in transverse conductance \( (\sigma_{xy}) \). Figure 4.6 shows the \( \nu = 2 \) to \( \nu = 6 \) plateau transition at \( T = 300 \text{ mK} \) and \( B = 10 \text{ T} \) for different values of current. The lowest curve (in red-circle) is the measured transverse conductance from \( \nu = 2 \) to \( \nu = 6 \) plateau at a bias current of 0.75 \( \mu \text{A} \). Consecutive curves are measured similarly with an increment of 1.5 \( \mu \text{A} \) for each. As we increase the injected current, the transition width starts to increase as well. Interestingly, in the transition region, all the curves intersect at the filling factor 4 and \( R_{xx} \) shows a
4.4. Effect on plateau to plateau transition

small suppression in peak resistance around the same gate voltage ($R_{xx}$ is shown in top-left inset).

Such an invariant point indicates that as we increase the current, the center of the electric field induced broadened extended state band does not move with the current. For $\nu = 2$ to $\nu = 6$ plateau transition, the Fermi level crosses the four fold degenerate $n = 1$ LL. It has been shown that at very high magnetic field, spin degeneracy can be lifted [104] giving rise to an additional plateau at $\nu = 4$. We speculate the current invariant point at $\nu = 4$ and suppression of $R_{xx}$ at the same time as a precursor of Zeeman splitting. We performed numerical calculations to understand our data quantitatively. These calculations were done based on current injection model described briefly in the next section.

4.4.1 Current injection model

The current injection model of QHE in graphene is described in greater details in Ref[90]. We present this model in brief with key steps here. The model calculates the potential distribution of the graphene flake in quantum Hall limit when source electrode is biased with $V_{SD}$. Following the calculation of potential distribution, spatial dependence of electric field can be calculated. This solution of electric field has a singularity at the source contact. Solution of Dirac equation in crossed electric and magnetic field [92, 100] can be used to calculate the local density of states. These solution in crossed electric and magnetic field can be obtained by performing a Lorentz boost in frame a reference where electron see only magnetic field. This leads to the modification of energy spectrum and wavefunctions by a group velocity ($\frac{E}{B}$) term. Once wave functions are found, local density of states can be obtained. The effect of transverse electric field on the local density of states is to cause spin splitting in otherwise spin degenerate Landau levels. For the calculation of current, in this model, it has been assumed that the fate of electron is decided by the density of states at the injection point near the source contact as later in the sample the transport is ballistic. Since there is a singularity in the electric field solutions at the contacts, one can choose a location slightly inside the contacts where injection of electrons takes place. This position is the only free parameter in the model and can be thought of as the position where hot spot forms. Once local density of states are calculated at the
Figure 4.6: a) Plot of $\sigma_{xy}$ as a function of $V_g$ for $\nu = 2$ to $\nu = 6$ plateau transition at $T = 300$ mK and $B = 10$ T for different values of currents starting from $0.75 \mu A$ with an increment of $1.5 \mu A$. The invariant point at $\nu = 4$ is clearly seen. The top-left inset shows the plot of $R_{xx}$ as a function of $V_g$ for the same transition at the same values of current as indicated in the main plot. b) Plot of calculated $\sigma_{xy}$ as a function of $V_g$ using the current injection model for the same values of current as shown in Figure (a). The inset shows the shape of calculated density of states in presence of transverse electric field for $n = 1$ Landau level. Spin splitting of otherwise degenerate levels can be seen.
hot spot, one can write the current expression as the product of the group velocity and the local density of states convoluted with the temperature dependent Fermi-Dirac distribution. Inset of Figure 4.6(b) shows the local density of states calculated for $n = 1$ Landau level with electric field induced spin split levels calculated by taking hot spot position 40 nm inside the sample. As current in the sample is increased, the larger electric field tends to split degenerate levels further apart keeping the center of the level intact. This leads to an invariant point with bias current. Figure 4.6(b) shows the conductance curves calculated for different values of the injected current. With this model, we could accurately describe the position of the current invariant point but it fails to explain the width of the transition region. One possible reason for the failure of this model could be the assumption that all states are extended. Further detailed analysis is needed to take into account the effect of disorder.

4.5 Summary

To summarize this chapter, we have studied the non-equilibrium breakdown of the quantum Hall state in graphene. We explain the device characterization methods with and without magnetic field. We find that the dissipationless QH state can be suppressed due to a high current density, and the corresponding critical current decreases with $|\nu|$. The correlation between $V_{\text{Hall}}$ and $\Delta E_{\nu}$ is consistent with the disorder-induced broadening of LLs and inhomogeneous charge distribution. The value of $V_{\text{Hall}}$ at breakdown gives an idea about the activation energy. We also see a current invariant point in the plateau to plateau transition and suppression in longitudinal resistance at higher current, which can possibly be a sign of spin-degeneracy breakdown and present a quantitative analysis based on current injection model to explain these observations.
Chapter 5

Probing thermal expansion of graphene and modal dispersion at low-temperature using graphene NEMS resonators

The aim of this chapter is to describe the study of graphene as an active part of the nanoelectromechanical system, as well as to show that such measurements can be very well directly applied to measure the physical properties of graphene. We study the dispersion, the variation of resonant frequency with DC gate voltage, of the electromechanical modes and find considerable tunability of resonant frequency. We provide a quantitative analysis for the frequency tunability. Measurement of the change in frequency resulting from a change in tension, from 300 K to 30 K, allows us to extract information about the thermal expansion of monolayer graphene as a function of temperature. We find that thermal expansion of graphene is negative for all temperatures between 300 K and 30 K. At the end of this chapter, we discuss the nature of multiple modes and dissipation in graphene resonator. The results presented here have been published in Ref[105].
5.1 Suspended graphene device as a mixer

As described in Chapter 3, due to the tunability of conductance with gate voltage, the electrical actuation and detection can be done by using the suspended graphene device as heterodyne-mixer [29, 31, 36]. For the sake of completeness we describe the scheme for electrical actuation and detection in Figure 5.1(a) superimposed on the SEM image of a multi-terminal device. A radio frequency (RF) signal $V_g(\omega)$ and a DC voltage $V_g^{DC}$ are applied at the gate terminal using a bias-tee. Another RF signal $V_{SD}(\omega + \Delta \omega)$ is applied to the source electrode. The amplitude of the current through the graphene membrane at the difference frequency ($\Delta \omega$), also called the mixing current $I_{\text{mix}}(\Delta \omega)$ is recorded for the measurement of resonant mode. Figure 5.1(b) shows such a measurement from a device (D2) at $V_g^{DC} = 10$ V at room temperature. Solid line is the fit to the experimental data using Equation (3.8), which gives resonant frequency ($f_0$) and quality factor ($Q$) to be 70 MHz and 80, respectively.

At the electromechanical resonance of the membrane, the first term inside the bracket in Equation (3.8) contributes significantly and the second term which does not depend on the mechanical motion of the graphene membrane provides a smooth background.

As described in the Chapter 3, a DC voltage at the gate electrode exerts an electrostatic force on the membrane inducing tension in it (see Equation (3.1)). This leads to the tunability of the resonant frequency with gate voltage and will be referred as modal dispersion in the forthcoming text. Figure 5.1(c) shows the colorscale plot of the measurement of mixing current for a device (D2) with gate voltage at room temperature. Here, two mechanical modes can be seen. The resonant frequency of these two modes increase as $|V_g^{DC}|$ is increased. This we will refer as positive dispersion. The positive dispersion of the resonant modes is easy to understand from the tension induced in the graphene membrane due to the electrostatic attraction from the back-gate as gate voltage is changed on either side of the $V_g^{DC} = 0$ V. Also, the amplitude of the mixing current at resonance is very poor in the vicinity of $V_g^{DC} = 0$ V as the actuation force is directly proportional to the $V_g^{DC}$ (see Equation (3.2)). At zero gate voltage the resonant frequency of fundamental mode of graphene is $\sim 63$ MHz. The energy contribution of flexural rigidity of graphene to the elastic energy of the system is small and pure flexural modes are expected to be at lower frequency for typical dimensions of our devices. The presence of high frequency modes
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Figure 5.1: a) Tilted angle scanning electron microscope (SEM) image of a suspended monolayer graphene device and the electrical circuit for actuation and detection of the mechanical motion of the graphene membrane. The scale bar indicates a length of 2 µm. b) A plot of the mixing current $I_{mix}(\Delta \omega)$ as a function of frequency $f (= \frac{\omega}{2\pi})$ at 300 K with the DC gate voltage $V_g^{DC} = 10$ V for device (D2). The sharp feature in the mixing current corresponds to the mechanical resonance. The solid curve is the fit to the experimental data. c) Colorscale plot of $I_{mix}(\Delta \omega)$ as a function of frequency $f$ and the DC gate voltage for device (D2) at 300 K. Two positively dispersing modes of the device are seen.
5.1. Suspended graphene device as a mixer

indicates that contribution of in-built tension to the elastic energy dominates over the flexural rigidity contribution. The in-built tensile stress at 300 K can arise during the fabrication process. To better understand the modal dispersion quantitatively, we model the graphene membrane within the elastic continuum regime and in a limit where tension in the membrane dominates over the flexural rigidity [6, 28, 106].

5.1.1 Putting electrostatics in graphene resonators

Following Equation (2.42), the resonant frequency \( f_0 \) of the graphene membrane under in-built tension clamped at two opposite sides can be written as,

\[
f_0(V_{g}^{DC}) = \frac{1}{2L} \sqrt{\frac{\Gamma(\Gamma_0(T), V_{g}^{DC})}{\rho tw}}, \tag{5.1}
\]

where \( L \) is the length of the membrane, \( w \) is the width, \( t \) is the thickness, \( \rho \) is the mass density, \( \Gamma_0(T) \) is the in-built tension and \( \Gamma \) is the tension at a given temperature \( T \) and \( V_{g}^{DC} \). Our next goal is to derive the functional form of \( \Gamma(\Gamma_0(T), V_{g}^{DC}) \) in terms of the device parameters by taking into account the electrostatic and elastic energies. Here, we note that from the resonant frequency at zero gate voltage, given by \( f_0(0) = \frac{1}{2L} \sqrt{\frac{\Gamma_0(T)}{\rho tw}} \), an independent estimation of \( \Gamma_0(T) \) and \( \rho \) is not possible. As the mass density of graphene could be different from that of pristine graphene due to the presence of residue adhered during the fabrication processes. However, resonant frequency dispersion with gate voltage \( f_0(V_{g}^{DC}) \) should allow us to estimate \( \rho \) and \( \Gamma_0(T) \) simultaneously.

After neglecting the flexural rigidity term in Euler-Bernoulli equation (see Equation (2.32) describing the vibration of a membrane is

\[
\rho t \frac{\partial^2 z}{\partial \tau^2} = \frac{\Gamma}{w} \frac{\partial^2 z}{\partial x^2} + F(x), \tag{5.2}
\]

where \( F(x) \) is the externally applied force per unit area, \( \Gamma \) is the tension, \( \rho \) is the mass density of graphene, \( w \) is the width of the flake, \( t \) is the thickness and \( \tau \) is the time. A graphene sheet is held between two electrodes distance \( D \) apart. Distance
measured along the length of the sheet from any one end is denoted by $x$ and $z(x)$ is the distance by which it is pulled towards the substrate due to an applied gate voltage as per the axis shown in Figure 5.1(a). The capacitive force per unit area at a point on the membrane is assumed to have the same form as that for an infinite parallel plate capacitor, depending on the height of the point above the substrate.

$$F(x) = \frac{\varepsilon_0 V^2}{2[R - z(x)]^2} \simeq \frac{\varepsilon_0}{2R^2} V^2 + \frac{\varepsilon_0}{R^3} V^2 z(x), \quad (5.3)$$

where $R$ is the height above which the sheet is suspended and $V$ is the potential difference between two plates of the capacitor. At an applied DC gate voltage $V_g^{DC}$, the bending of the sheet, assuming $z(x) \ll R$ can be expressed as

$$z(x) = \frac{\varepsilon_0 w V_g^{DC^2}}{4R^2 \Gamma} (Dx - x^2).$$

The actual length of the stretched graphene sheet is

$$\int_0^D \sqrt{1 + \left(\frac{\partial z}{\partial x}\right)^2} dx \simeq D + \xi \frac{(V_g^{DC})^4}{\Gamma^2}, \quad \text{where, } \xi = \frac{\varepsilon_0 w^2 D^3}{96 R^4}.$$ 

Let us denote by $L_0$ the length the sheet would have taken if tension was zero. From the stress-strain relationship, we can write

$$\frac{\Gamma}{wt} = \frac{E_{\text{graphene}}}{L_0} \left[D + \xi \frac{(V_g^{DC})^4}{\Gamma^2} - L_0\right]. \quad (5.4)$$

The cubic equation in tension written above can be solved for the tension to get

$$\Gamma = \frac{\Gamma_0}{3} + \frac{\sqrt{2} \Gamma_0}{3b} + \frac{b}{3\sqrt{2}}, \quad (5.5)$$

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5.2 Modal dispersion at low temperature

where,

\[ a = \frac{\varepsilon_0^2 w^2 D^3 (V_g^{DC})^4}{96 R^4}, \]

\[ b = 3 \sqrt{3} \sqrt{27 \sigma^2 + 4a\Gamma_0 + 27a + 2\Gamma_0^3}, \]

and

\[ \Gamma_0 = E_{\text{graphene}} wt \left( \frac{D - L_0}{L_0} \right). \]

Therefore, Equation (5.1) combined with Equation (5.5) can completely describe the dispersion of resonant modes at room temperature. Fitting based on these two equations, we estimated the mass density \( \rho = 7.4 \rho_{\text{graphene}} \) and the in-built tension \( \Gamma_0(300K) \) to be 68.6 nN for the fundamental mode, where \( \rho_{\text{graphene}} \) is the mass density of pristine graphene for the device data shown in Figure 5.1(c). This extra mass and in-built tension can be attributed to the resist residue that can get deposited on the graphene membrane during the fabrication process [29]. We note that the dispersion of the two modes shown in the Figure 5.1(c) is different and our modal calculations give \( \rho = 7.3 \rho_{\text{graphene}} \) and \( \Gamma_0(300K) = 107.6 \) nN for the upper mode. However, the simple model of a rectangular membrane under tension predicts higher order resonant modes at much higher frequencies – this is a limitation of the simple assumptions we have made in our calculations. The presence of other resonant modes at small intervals of frequency \( f \) seen in Figure 5.1(c) indicates a deviation from this simplified picture of membrane under tension due to either edge-modes [30], or due to the rippling/curling of the membrane [84, 107]. In the later part of this chapter, we discuss in detail the reasons for positive or negatively dispersing modes and temperature induced change in dispersion.

5.2 Modal dispersion at low temperature

At room temperature, most of the devices show positive dispersion with gate voltage (show an increase in the resonant frequency with \( |V_g^{DC}| \)). As we cool down, in all the devices studied, resonant frequency increases and its tunability on gate voltage reduces. Figure 5.2(a) and (b) shows the resonant frequency dispersion with gate voltage at various temperatures for one of our devices (D3). Cooling from room tem-
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Temperature to 6 K, resonant frequency increases from ~ 60 MHz to ~ 105 MHz. At room temperature the frequency can be tuned by 40 MHz by applying 20 V gate voltage. However, at 6 K, tunability is less than 0.5 MHz for the same range of gate voltage. Also, at low temperatures, in the vicinity of the zero gate voltage, we see negative dispersion (resonant frequency decreases with the increase in $|V_{g}^{DC}|$). Figure 5.2(b) shows the negative dispersion before a positive upturn at low temperature.

The observed modal dispersion behavior at all temperature can be qualitatively understood from the contraction of the suspended gold electrodes with temperature, which leads to increased tension in the membrane. This increase in in-built tension is accompanied by the reduction in tunability with $V_{g}^{DC}$. However to explain the negative dispersion, we look back at approximations made while deriving Equation (5.5). While calculating the electrostatic force per unit area on the graphene sheet we have ignored the higher order term and has kept only the leading order term. As an improvement over this approximation, we can include the first order term in the equation of small vibrations for the graphene flake about the equilibrium position. Therefore, from Equation (5.2) and (5.3), we can write down the equation for the small oscillation of the membrane about the equilibrium configuration

$$\rho t \frac{\partial^2 \delta z}{\partial \tau^2} = \frac{\Gamma w}{\partial x^2} + \frac{\varepsilon_0 (V_{g}^{DC})^2}{R^3} \delta z,$$

(5.6)

where the small displacement $\delta z$ is a function of both $x$ and time $\tau$. The fundamental resonant frequency is given by

$$f_0 = \frac{1}{2\pi} \sqrt{\frac{\pi^2 \Gamma}{D^2 \rho tw} - \frac{\varepsilon_0 (V_{g}^{DC})^2}{\rho t R^3}},$$

(5.7)

where $\Gamma$ is given by Equation (5.5).

In order to gain more insight into Equation (5.7), we perform a Taylor series expansion of the Equation (5.5), which gives

$$\Gamma = E_{graphene} wt \left( \frac{D - L_0}{L_0} \right) + \frac{\varepsilon_0^2 D^3 w L_0}{96 E_{graphene} t R^4 (D - L_0)^2} (V_{g}^{DC})^4 + H.O.$$
This leads to

$$f_0 \simeq \frac{1}{2\pi} \sqrt{\frac{\pi^2 E_{\text{graphene}}(D - L_0)}{D^2 \rho L_0} - \frac{\varepsilon_0 (V_{g}^{\text{DC}})^2}{\rho t R^3}} + \frac{\pi^2 \varepsilon_0^2 D L_0}{96 \rho E_{\text{graphene}} t^2 R^4 (D - L_0)^2} (V_{g}^{\text{DC}})^4. \quad (5.9)$$

Qualitatively, this behavior can be understood from Equation (5.9). The first term inside the square-root on the r.h.s of the Equation (5.9) comes from the in-built tension. As the temperature is lowered, \((D - L_0)\) increases since contraction of gold electrodes outweighs the thermal expansion of graphene and resonant frequency at \(V_{g}^{\text{DC}} = 0\) goes up. Also, the coefficient of the \((V_{g}^{\text{DC}})^4\) inside the square root of Equation (5.9) decreases and fails to suppress the negative \((V_{g}^{\text{DC}})^2\) term, resulting in a prominent negative dispersion at low temperatures.

At room temperature, Equation (5.7) describes the resonance frequency dispersion with \(V_{g}^{\text{DC}}\) accurately as shown in 5.2(c). However, it fails to describe the resonant frequency dispersion with \(V_{g}^{\text{DC}}\) at lower temperatures quantitatively. To describe it quantitatively as well, we introduce one parameter \(\lambda\) while estimating the tension (see Equation (5.4)) in the flake, \(\Gamma = \lambda E_{\text{graphene}} wt \frac{(L - L_0)}{L_0}\). At room temperature \(\lambda = 1\), however to fit the dispersion data at 6 K, \(\lambda = 0.05\) is the optimum value. Figure 5.2(d) shows the fitted curve obtained using fitting parameter \(\lambda\) at low temperatures.

There can be many microscopic contributions towards this parameter that are not appropriately accounted for within our model. One of them is the estimate of the capacitance. We have used a simple parallel plate capacitance model for the calculation of the capacitance between the graphene flake and the substrate; this is a very good approximation in the neighborhood of \(V_{g}^{\text{DC}} = 0\) V. However, at higher gate voltages, the electrostatic attraction causes the graphene flake to flex down (this can be of the order of \(\sim 10\) nm) and comes closer to the substrate, which can lead to change in capacitance and its derivative. Second contribution can arise from the fact that we have used the room temperature value of the Young’s modulus (1 TPa) for describing the low temperature data as well, a change in the Young’s modulus can also affect the resonance frequency dispersion with gate voltage. Theoretical calculations do suggest that the assumption of constant Young’s modulus over this temperature range is reasonable [108]. Thirdly, the thermal response of impurities on the flake cannot be ignored while explaining the low temperature data. Residue on the flake
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Figure 5.2: a) Measured dispersion of an electromechanical mode as function of temperature for device (D3). b) Closeup of the experimentally measured low-temperature data showing the non-monotonic dispersion. c) Modeling of the dispersion, incorporating thermal expansion of graphene, at various temperatures by varying temperature and the parameter $\lambda$. d) Closeup of the calculated low-temperature data showing the non-monotonic dispersion similar to the experimentally measured dispersion.
5.2. Modal dispersion at low temperature

can modify the strain estimation \( \frac{L-L_0}{L_0} \) at low temperatures as the system acts like a composite polymer-graphene system. We have tried to capture all of these effects in the form of a parameter \( \lambda \) as an independent measurement is not possible from the current experiment.

Now we present a phenomenological model to describe the modal dispersion at low temperature. The two limiting cases of modal dispersion – pure positively and negatively dispersing modes can be respectively understood by considering the limits where the tension (or in the case of flexural modes mechanical rigidity) dominates with increasing \( V_g^{DC} \) and the case when capacitive pulling leading to the softening of the spring constant dominates \([39, 86, 109]\). A simple model \([39]\) to understand the intermediate regime where these two interactions compete is to model the resonator with an intrinsic spring constant \( K_i = k + \alpha (V_g^{DC})^2 + \beta (V_g^{DC})^4 + H.O.(V_g^{DC}) \) with \( \alpha \) and \( \beta \) as constants of the system (when \( \beta > 0 \), this is consistent with the positive dispersion of modes with an increased \( V_g^{DC} \)). A second contribution due to the electrostatic interaction (capacitive coupling), softens the intrinsic spring constant \( K_i \) so that the effective spring constant

\[
K_{eff} = K_i - \frac{1}{2} (V_g^{DC})^2 \frac{d^2 C_g}{dz^2} \tag{5.10}
\]

with \( C_g \) being the capacitive coupling of the membrane with gate electrode. As \( V_g^{DC} \) is varied the \( K_{eff} \) varies and the modal dispersion changes from a negatively dispersing mode to a positively dispersing mode at large \( V_g^{DC} \) – the value of \( V_g^{DC} \) at which the crossover happens is a measure of the relative contribution of capacitive and elastic energies. This phenomenological model describes the generalized modal dispersion. In Equation \([5.9]\), an attempt has been made to connect these parameters to the material properties. \( \alpha \) and \( \beta \) depend on temperature through the coefficient of thermal expansion, and also on the elastic constant of graphene.

Results of our calculation shown in Figure \([5.2c]\) clearly show that we can successfully model the temperature evolution of dispersion, using only a single fit parameter in our model. It is critical to understand the origin of observed dispersion of resonant frequency as a function of temperature due to the desirable property of resonator – its tunability. Additionally, if the loss mechanisms are frequency independent such tunability can increase the \( Q \) of the system \([86]\).
5.3 Probing thermal expansion of graphene

We now consider how the resonant frequency \( f_0 \) evolves as a function of the temperature. Figure 5.3(a) shows the result of an evolution of a mode as a function of temperature at \( V_{g}^{DC} = 15 \) V. Data acquisition was done during a single sweep over twelve hours to allow the resonator to equilibrate and the window of acquisition window automatically adjusted to follow the resonance. The resonant frequency increases as the device is cooled from room temperature. This increase has been seen in all the devices we have studied. Further the width of the resonance feature in mixing current reduces as we cool down the resonator. This reduction is related to the dissipation in the resonator and will be discussed in detail later in this chapter.

The degree of frequency shift varies from one device to another depending on the device geometry. The origin of this frequency shift with temperature is the increase in tension in graphene due to the expansion/contraction of substrate, gold electrodes and graphene. The frequency shift can be understood by taking into account the contribution of various strains as the device is cooled below 300 K. The three main contributions are – first, the thermal strain in unconstrained graphene, \( \epsilon_{\text{graphene}}(T) = \int_{T}^{300} \alpha_{\text{graphene}}(t) dt \), due to the coefficient of thermal expansion of graphene \( \alpha_{\text{graphene}}(T) \), second, the thermal strain due to the gold electrodes, \( \epsilon_{\text{gold}}(T) = \int_{T}^{300} \alpha_{\text{gold}}(t) dt \), and last the contribution of the strain induced by the substrate \( \epsilon_{\text{substrate}}(T) \); here \( \alpha_{\text{gold}}(T) \) is the coefficient of thermal expansion for gold. We will be measuring the strain relative to the strain at room temperature (300 K). The strain in gold electrodes plays an important role due to the geometry of the device. The under-etch that releases the graphene membrane also etches under the graphene covered by the electrodes – resulting in the graphene membrane being suspended off the gold electrodes as described in Chapter 3. The geometry of the resulting device is shown in the inset to Figure 5.3(a).

As described in previous sections, the frequency dispersion with gate voltage allows us to estimate \( \Gamma_0(T) \) and \( \rho \) of the graphene flake independently. And with one fitting parameter \( (\lambda) \), we can completely describe the frequency dispersion with gate voltage at all temperatures. However, the resonance frequency at zero gate voltage \( (f_0(0)) \) solely depends on the \( \Gamma_0(T) \) \( (f_0(0) = \frac{1}{2\pi} \sqrt{\frac{\Gamma_0}{\rho wt}}) \). Therefore, using \( \rho \) calculated from the room temperature frequency-dispersion curve, we can estimate the \( \Gamma_0(T) \) and hence
5.3. Probing thermal expansion of graphene

Figure 5.3: a) Plot showing the evolution of the resonant frequency of a mode for device (D2) as a function of temperature for $V_{g}^{DC} = 15$ V. Inset shows the schematic of all the strains external to the suspended graphene membrane as the device is cooled below 300 K. b) The plot of expansion coefficient of graphene as a function of temperature. Data from two different devices together with theoretical prediction of N. Mounet et al. [110]. The shaded area represents the errors estimated from the uncertainty of the length of the flake, width of the electrode and Young’s modulus of graphene.
net strain at any given temperature. Therefore, measurement of resonant frequency variation with temperature directly allows us to probe the in-built tension and hence thermal strain in the system.

Now, we consider the various contributions to the net strain in the flake. First, we consider the contribution from substrate. Different coefficients of thermal expansion of Si and SiO$_2$ also contribute to the net strain in the flake. At any temperature, substrate’s contribution to the net strain between the electrode can be calculated using generalized Stoney’s formula \[112\] and is given by,

\[
\epsilon_{\text{substrate}}(T) = -\frac{d_{\text{SiO}_2} E_{\text{SiO}_2}(1 - \nu_{\text{Si}})}{d_{\text{Si}} E_{\text{Si}}(1 - \nu_{\text{SiO}_2})} \int_{T}^{300} (\alpha_{\text{Si}}(t) - \alpha_{\text{SiO}_2}(t)) \, dt, \tag{5.11}
\]

where $E_{\text{Si}}$ and $E_{\text{SiO}_2}$ are the Young’s modulii, $d_{\text{Si}}$ and $d_{\text{SiO}_2}$ are the thicknesses, $\nu_{\text{Si}}$ and $\nu_{\text{SiO}_2}$ are the Poisson’s ratio, and $\alpha_{\text{Si}}(T)$ \[113\] and $\alpha_{\text{SiO}_2}(T)$ \[114\] are the coefficient of thermal expansion of silicon and silicon oxide respectively. We can now write an effective coefficient of thermal expansion for trench as $\alpha_{\text{trench}} = \frac{d\epsilon_{\text{substrate}}(T)}{dT}$. The value $\alpha_{\text{trench}}$ at room temperature can be estimated by taking, $d_{\text{SiO}_2} = 300 \text{ nm}$, $d_{\text{Si}} = 300 \mu\text{m}$, $E_{\text{Si}} = 160 \text{ GPa}$, $E_{\text{SiO}_2} = 70 \text{ GPa}$, $\nu_{\text{Si}} = 0.22$ and $\nu_{\text{SiO}_2} = 0.16$. By using the thermal expansion coefficient for Si and SiO$_2$ as a function of temperature \[113\] \[114\], we estimate, $\alpha_{\text{trench}}(300) = -8.1 \times 10^{-10} \text{ K}^{-1}$ and $\alpha_{\text{trench}}(30) = -2.0 \times 10^{-10} \text{ K}^{-1}$. Therefore, substrate’s contribution to the net strain in the flake is very small and can be neglected.

Next, we consider the contribution coming from the suspended gold electrodes. At the interface of gold electrodes supporting the graphene membrane, the net force must balance to zero; however, the stresses are different considering the cross-sectional area of gold electrodes ($\sim 500 \text{ nm} \times 60 \text{ nm}$) and graphene ($\sim 500 \text{ nm} \times 0.3 \text{ nm}$). The large difference in the cross-sectional area implies that the effective stiffness of gold electrodes is large compared to the stiffness of graphene. As result, to a very good approximation, the total elastic strain at a given temperature in graphene that is confined by “rigid” gold electrodes is $\epsilon_{\text{graphene clamped}} = \epsilon_{\text{graphene}}(T) + \epsilon_{\text{substrate}}(T) - \epsilon_{\text{gold}}(T) \frac{w_{\text{electrode}}}{L}$, where $w_{\text{electrode}}$ is the average of the width of gold electrodes holding the suspended flake. The change in tension in the membrane can be written as a
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function of temperature as \( \Delta \Gamma_0(T) = w t \epsilon_{\text{grapheneclamped}}(T) E_{\text{graphene}} \), where \( E_{\text{graphene}} \) is Young’s modulus of graphene.

By considering the expansion/contraction of the graphene flake itself and assuming that Young’s modulus of graphene does not vary significantly over the temperature range [108], we can write the net built-in tension in the flake as,

\[ \Gamma_0(T) = \Gamma_0(300) + \frac{E_{\text{graphene}} w t}{L} \left( L \int_{300}^{T} \alpha_{\text{graphene}}(T) \, dT - w_{\text{electrode}} \int_{300}^{T} \alpha_{\text{gold}}(T) \, dT \right) . \]  

\[ (5.12) \]

Using Equation (5.1) and assuming a uniform contraction/expansion of gold electrodes, we can write,

\[ \alpha_{\text{graphene}} = -\frac{w_{\text{electrode}}}{L} \alpha_{\text{gold}}(T) - \frac{4L^2 \rho}{E_{\text{graphene}}} 2f_0(0) \frac{df_0(0)}{dT} . \]  

\[ (5.13) \]

Figure 5.3(b) shows the result of calculating \( \alpha_{\text{graphene}} \) for two devices using this analysis and comparison with the theoretical calculation for \( \alpha_{\text{graphene}} \) by N. Mounet et al. [110]. We find that \( \alpha_{\text{graphene}} \) is negative and its magnitude decreases with temperature for \( T < 300 \) K. At room temperature, \( \alpha_{\text{graphene}} \sim -7 \times 10^{-6} \) K\(^{-1}\), which is similar to the previously reported values measured by others [29, 107]. At 30 K, \( \alpha_{\text{graphene}} \sim -1 \times 10^{-6} \) K\(^{-1}\). The deviation of \( \alpha_{\text{graphene}} \) from the theoretically predicted values can possibly be due to the presence of the impurities on graphene membrane. The knowledge of \( \alpha_{\text{graphene}} \) is essential for the fabrication of the devices intended for strain engineering applications [43, 44]. Strain engineering of graphene devices at low-temperatures using this picture can improve device performance, for example by enabling temperature compensation [115]. A simple design rule for width of electrodes to achieve temperature compensation in the vicinity of a temperature \( T_0 \) is to ensure that the \( w_{\text{electrode}} = L \times \left| \frac{\alpha_{\text{graphene}}(T_0)}{\alpha_{\text{gold}}(T_0)} \right| \) (using Equation (5.13)). Additionally, these measurements indicate that using NEMS resonators for measuring internal stress of nanowires of metal and phase change materials as a function of temperature, using

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phase locked loop (PLL) technique [42], can provide useful information about stresses and strains in individual nanostructures.

Here, it is important to point out that, residue on the graphene sheet can also affect the calculations of the coefficient of thermal expansion. This can possibly be one reason for its deviation from the theoretically predicted values. In our calculation, we have assumed that presence of residue does not affect the expansion of graphene, which may not be the complete description of the experimental system. However, these values of $\alpha_{\text{graphene}}$ still remain valuable for device engineering as the complete elimination of the resist residue from the device is difficult.

5.4 Multiple modes

Usually graphene resonators fabricated in a geometry with two opposite side clamped show closely spaced multiple modes. In order to use graphene electromechanical resonators for applications like mass and charge sensing it is important to understand the microscopic origin of these modes and to test the suitability of continuum models [28, 29, 106] beyond basic properties such as dispersion.

Soft degrees of freedom arising from the free edges along the length of the flake can give rise to the additional modes without the predictability over their resonant frequency. Experiments done on the graphene resonator clamped along all four edges show better predictability over their resonant frequency [116]. Scanned probe measurements to image modes [30] indicate that the origin of these modes is complex – for instance some of the modes are associated with the edges of the graphene membrane. The reason this is critical is that the notion of effective mass of mechanical modes [8, 73, 76] is associated with the spatial amplitude distribution of the mode. For example, for flexural oscillations, the effective mass $m_{eff}$ is mode shape dependent. However, for the case of a string under tension, $m_{eff}$ is independent of the mode shape [76]. We try to understand the nature of modes in graphene resonators by studying devices with multiple resonances in the measurement frequency range. Figure 5.4(a) shows two higher order modes along with fundamental mode for a graphene resonator at 7 K. The dispersions of these three modes are different. Though it is difficult to determine the exact nature of these modes using mixing current technique, a semi-
Figure 5.4: 
a) Colorscale plot of $I_{mix}(\Delta \omega)$ showing three negatively dispersing modes with varying slopes. b) Fitted data for varying effective mass for the three modes. The inset shows two possible modes for a uniform rectangular membrane under tension. The effective mass is expected to be independent of the mode – an observation that is not experimentally seen within the measured frequency range.
quantitative explanation can be provided for this. At lower temperatures, negative dispersion can be understood in terms of spring constant softening as discussed earlier. Following Equation (5.10), the effective spring constant $K_{\text{eff}}$ can be used to calculate the modal dispersion with gate voltage ($V_g^{DC}$),

$$f^2 = f_0^2 - \frac{1}{8\pi^2 m_{\text{eff}}} (V_g^{DC})^2 \frac{d^2 C_g}{dz^2},$$  

(5.14)

where $f_0 = \frac{1}{2\pi} \sqrt{\frac{K_i}{m_{\text{eff}}}}$ and $m_{\text{eff}}$ is the effective mass for a given mode.

Using Equation (5.14), we can describe the higher modes also by using $f_0$ and modal mass ($m_{\text{eff}}$) as the fitting parameters. For a sheet under tension with uniform loading, the effective modal mass ($m_{\text{eff}}$) for different modes does not change, unlike the case of flexural modes, and is equal to $m_{\text{eff}} = 0.785 \rho L w t$ [76]. However, our fitting scheme gives different effective masses for different modes, which is expected as these are not the higher order harmonics of the fundamental mode (in the model described above, the higher order modes will be integral multiples of the fundamental mode). This suggests that the simple picture of rectangular membrane under tension within the continuum description does not work well to describe the higher modes of the system. This could be due to four main reasons: a) non-uniform loading of the membrane (due to resist residue) [29] can modify the $m_{\text{eff}}$ for different modes, b) due to the presence of edge dependent modes [30], c) due to the curvature of membrane [84] or rippling of graphene [107] (as seen in Figure 5.1a) the effective stiffness of the modes could be a very complex quantity with a tensor nature and could be significantly different from the ideal value of $\sim 1$ TPa as observed for the case of rippled carbon nanotubes [117, 118, 119] and d) the capacitance to gate ($C_g$) for the graphene membrane is likely to be mode dependent. Further experiments with pristine unrippled graphene resonators are needed to clarify our understanding of the microscopic origin of modes. The presence of multiple closely spaced modes can be potentially useful for mass spectrometry as the position of the added mass can be extracted accurately [120].
5.5 Dissipation in graphene NEMS

Quality factor in graphene devices fabricated by methods presented are is low when compared to other nanostructures. Therefore, an understanding of various dissipation channels in these devices is highly desirable. Understanding dissipation in NEMS in itself is a very broad subject [12] and a complete discussion is out of the scope of this work. Here, we present our experimental findings on dissipation in suspended graphene devices without giving any insights on its microscopic origins. Figure 5.5(a) shows the measurement of mixing current at 7 K for a suspended graphene device (D1) while sweeping $V_{g}^{DC}$. A negative dispersing resonant mode can be clearly seen. As explained earlier, such a negative dispersion is well understood in terms of mode-softening due to the capacitive contribution to the energy of resonator [39, 109]. Also seen in Figure 5.5(a) is the information regarding the amplitude of the mixing current. The overall amplitude of the mixing current at resonance scales with $V_{g}^{DC}$ (first term in Equation (3.8)) and as a result the amplitude of the mixing current is very small near $V_{g}^{DC} = 0$ V. However, at $V_{g}^{DC} = 13$ V, the amplitude of the mixing changes abruptly due to the Dirac peak ($V_D$). At this point, the prefactor $\frac{dG}{dV_{g}^{DC}}$ appearing in Equation (3.8) becomes zero. Figure 5.5(b) shows the change of conductance with gate voltage for device D1 at 7 K. The position of Dirac point at 13 V can be seen clearly. The Dirac peak for our device is shifted ($V_D = 13$ V) from the expected position (0 V) due to unintentional doping during the fabrication process. A shift in the Dirac peak away from $V_{g}^{DC} = 0$ V is desirable in our experiments because the actuation efficiency is feeble at $V_{g}^{DC} = 0$ V and that would make the observation of physics near the Dirac point inaccessible for electromechanical measurements. Figure 5.5(c) shows the quality factor ($Q$) dependance on $V_{g}^{DC}$ calculated from the fits to the data shown in Figure 5.5(a) using Equation (3.8). The quality factor of the device is largest around $V_{g}^{DC} = 0$ V and decreases as the gate voltage is swept away from zero; a similar trend as followed by the $f_0$. As negative dispersion, caused by softening of spring constant, is accompanied by the movement of graphene towards the gate electrode, it leads to an increase in the modulated capacitance and results in larger dissipative current [109]. In the neighborhood of $V_{g}^{DC} = V_D$, $Q$ is smaller than the value expected from the trend from higher $V_{g}^{DC}$ side. One possible reason for this behavior might be that in the vicinity of the Dirac point, large device resistance leads to increased ohmic losses [86]. At Dirac point the charge inhomogeneity in sample
Chapter 5. Thermal expansion and modal dispersion of graphene NEMS

is largest \[101, 122\]. An additional mechanism that can result in dissipation is the mechanical deformation modifying the distribution of charge puddles of electrons and holes in the graphene sheet. These phenomena warrant further detailed investigation.

On cooling the graphene electromechanical resonators the resonance frequency as well as quality factor \((Q)\) increase. Figure 5.6 shows the variation of \(Q\) with temperature from a device. It can be seen that \(Q\) can be increased by a factor of four for this device. Similar increase in \(Q\) can also be seen in the Figure 5.3(a), where narrowing of the resonance peak with temperature is very evident. All the devices we measured showed increase in \(Q\) upon cooling. The observation that the increase in resonant frequency is accompanied by an increase in the \(Q\) suggests that the loss mechanism may be frequency independent \[86\]. The inset of the figure shows the plot of \(Q^{-1}\), which is directly proportional to the dissipation in the resonator, with temperature. At lower temperatures, dissipation reduces more slowly than compared to the temperatures greater than 100 K. This seems to suggest that there could be possibly be two channels for the dissipation in these devices. One could be the standard temperature dependent loss mechanism which reduces significantly below \(\approx 100\) K, while the other dissipating channel remain active even at low temperature \[29\].

5.6 Summary

In this chapter we have seen that modal dispersion of graphene resonators is affected by the thermal expansion of graphene reducing the tunability. The continuum description of the mechanics of rectangular graphene membrane is inadequate for explaining the resonances due to the presence of non-uniform impurities and rippling of the membrane. Using these NEMS devices measurement of coefficient of thermal expansion of graphene at low temperatures is possible. Our measurements indicate that \(\alpha_{\text{graphene}}\) is negative for all temperatures between 300 K and 30 K and larger in magnitude than the numbers predicted by theoretical calculations \[110\]. We provided measurements of \(Q\) for graphene resonators, as a function of DC gate voltage and temperature indicate that larger dissipation in the resonator can occur around the Dirac point.
Figure 5.5: a) Colorscale plot of the mixing current as function of frequency $f$ and gate voltage $V_{g}^{DC}$ at 7 K for device (D1). The maxima (blue) and minima (red) of the colorbar correspond to 2.6 nA and 0 nA, respectively. The dot-dash line indicates the position of the Dirac peak for the graphene device. b) The change of conductance as a function of gate voltage indicating the position of Dirac point. c) Plot of the measured quality factor ($Q$) for the data shown in (a). The $Q$ drops as $|V_{g}^{DC}|$ increases and around the Dirac point the Q shows a dip.
Figure 5.6: The plot shows the variation of the quality factor of resonance with temperature. The inset shows the plot of $Q^{-1}$ with temperature.
Chapter 6

Coupling between quantum Hall state and electromechanics in suspended graphene resonator

In this chapter, we describe the coupling between two different aspects of suspended graphene devices. As described in the previous chapters, graphene is a two dimensional electron system, which in presence of magnetic field, shows quantum Hall effect. Quantum Hall effect can be probed with the help of electron transport measurements. At the same time electrical signals can be used to actuate and detect the mechanical motion of suspended graphene devices. Using graphene NEMS [28, 29, 116, 105], we perform electromechanical measurements in quantum Hall regime to probe the coupling between a quantum Hall (QH) system [12, 13] and its mechanical motion. Mechanically perturbing the QH state through resonance modifies the DC resistance of the system and results in a Fano-lineshape due to electronic interference. We provide quantitative analysis for the observed behavior. Magnetization of the system modifies the resonator’s equilibrium position and effective stiffness leading to changes in the resonant frequency. We provide a quantitative model for the calculation of magnetization and observed behavior in mechanical response of the resonator.
6.1 Magneto-transport in high mobility graphene samples

As described in Chapter 3, suspended graphene devices can be cleaned by passing a large current using, the process known as current annealing. Figure 6.1(a) shows the variation in resistance with gate voltage \(V_{DC}^g\) of a device at 5 K after current annealing \([79, 80, 89]\); this cleaning step is possible due to the low contact resistance. The sharp Dirac peak at zero gate voltage and large mobility of charge carriers of \(\approx 150,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}\), are the measures of the high quality of the sample. This is further confirmed in the magneto-transport measurements as shown in Figure 6.1(b) where colorscale plot of two probe resistance is plotted a function of the magnetic field and gate voltage. Various filling factors appeared as dispersing color bands on the fan diagram. Figure 6.1(c) shows the plot of resistance with magnetic field at \(V_{DC}^g = 5 \text{ V}\) from Figure (b) where different quantum Hall plateaus \((\nu = 2, 6)\), unique to the monolayer graphene, are clearly seen.

6.2 Various resonant modes in graphene NEMS

Devices fabricated by this process, described earlier in Chapter 3, show two kinds of resonant modes - resonant modes which arise from the suspended portion of gold electrodes (see Section 3.4) that overlap with the graphene (gold modes) and modes arising from the mechanical motion of the graphene flake being clamped at two opposite edges (graphene modes) \([29, 105]\). At room temperature, the resonant frequency of the gold modes roughly matches with the value calculated by assuming a doubly clamped beam geometry \([29]\). At low temperatures, for typical device dimensions, gold modes have resonant frequency of \(\sim \text{MHz}\) and do not show tunability with the gate voltage (small induced tension due to large bending rigidity). Figure 6.2(a) shows the measurement of mixing current using two source heterodyne mixing technique with gate voltage for a gold mode at 5 K. It can be clearly seen that the tunability of resonant frequency with gate voltage is very poor. Another faint mode \(\approx 3.5 \text{ MHz}\) can be seen too, which possibly is due to the second gold electrode. However, at
6.2. Various resonant modes in graphene NEMS

Figure 6.1: a) Two probe device resistance as a function of the gate voltage. Inset shows the scanning electron microscope image of a device with a scale bar of 1 μm. b) Colorscale plot of resistance with magnetic field and gate voltage at 5 K (Landau level fan diagram). c) Resistance as a function of magnetic field at $V_g^{DC} = 5$ V.
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low temperature graphene modes have larger resonant frequency (≈100 MHz, due to increased tension due to contraction of gold electrodes) and show tunability with gate voltage. Figure 6.2(c) shows the colorscale plot of mixing current with gate voltage using frequency modulation (FM) technique at 5 K for a graphene mode. Therefore, such a characterization of the sample allows us to differentiate between the gold modes and graphene modes.

6.3 Effect of graphene electromechanics on quantum Hall state

In the first part of our experiment we probe the response of the quantum Hall state at resonant actuation. For this, we measure the DC resistance of the device while driving it through resonant frequency, by applying an RF signal at the gate electrode. Figure 6.3(a) shows the measurement of resistance with magnetic field across the resonant mode due to gold electrode (3.05 MHz). The resonant frequency of the device can be measured using mixing technique prior to these measurements. Here, we focus on the response near the low frequency gold mode and discuss in detail the difference in response of the graphene mode later in this chapter. We define change in resistance ($\Delta R$) by subtracting the device resistance measured away from the resonant frequency ($R_0(B)$) from the device resistance ($\Delta R(B) = R(B) - R_0(B)$). Figure 6.3(b) shows $\Delta R$ as a function of driving frequency and magnetic field. Some features can be clearly observed: a) $\Delta R$ is zero in the plateau region, b) $\Delta R$ has different signs across the quantum Hall plateaus (represented by blue and red color across the plateau), and c) $\Delta R$ oscillates with $B$ for low values of magnetic field as shown in Figure 6.3(c).

6.3.1 Theoretical model

To understand this, we model this geometry as a parallel plate capacitor, in which one plate (the substrate) remains fixed and the other plate (graphene flake) moves with large amplitude at the resonant frequency since it is tethered to the gold electrode. The voltage applied at the back gate influences the carrier density on the flake in two ways. Firstly, providing a driving force on the mechanical resonator leads to
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Figure 6.2: a) Colorscale plot of mixing current using two source heterodyne mixing technique with gate voltage for a gold mode. b) Lineplot from (a) at $V_g^{DC} = 5$ V. c) Modal dispersion of a graphene mode with gate voltage at 5 K. The colorscale plot of the mixing current (X-component at the lock-in) measured using frequency modulated (FM) technique. d) Lineplot from (c) at $V_g^{DC} = 5$ V along with the fitted curve with Equation (3.20) of mixing current for FM technique.
Figure 6.3: At 5 K a) Surface plot of resistance measured with magnetic field across the resonant frequency of the gold mode. b) Colorscale plot of the change in resistance ($\Delta R$) with magnetic field and driving frequency. The resonant frequency of the mode is 3.05 MHz. c) Plot of $\Delta R$ with magnetic field for smaller values of magnetic field taken at 3.05 MHz.
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capacitance oscillations due to physical motion. Secondly, due to a direct capacitive coupling of RF voltage, the carrier density on the flake also oscillates. Within the adiabatic approximation, the total number of charge carriers \( (N(t)) \) on the flake at any time can be written as the sum of these two contributions. As the flake vibrates at the resonant frequency with large amplitude, \( N(t) \) oscillates in time. The total time dependent voltage at the gate can be written as:

\[
V_g(t) = V_g^{DC} + (V_{ac}e^{-i\omega t} + c.c.).
\]

From this, we can write the total number of charge carrier on the flake as,

\[
N(t) = \frac{1}{e} C_g(t)V_g(t) = N_0 + \delta N(t), \tag{6.1}
\]

where \( N_0 \) is its value at \( V_{ac} = 0 \). In order to calculate the modification in resistance \( R_{DC} \) upon mechanical vibrations, we expand it in \( V_{ac} \) to second order,

\[
R_{DC} \equiv R_0 + R_{rect} \simeq R_0 + \frac{dR}{dN} \delta N(t) + \frac{1}{2} \frac{d^2 R}{dN^2} (\delta N(t))^2. \tag{6.2}
\]

The over-bar in the above equation indicates the DC part of a time varying quantity. \( \delta N(t) \) can also be expanded to the second order in \( V_{ac} \),

\[
\delta N(t) = \delta N_1(t) + \delta N_2(t), \tag{6.3}
\]

where,

\[
\delta N_1(t) = \frac{1}{e} \{ C_0 \delta V_g + V_0 \delta C \} = \frac{1}{e} \{ C_0 + V_0 \frac{dC}{dz} \frac{dz}{dV_g} \} \delta V_g, \tag{6.4}
\]

\[
\delta N_2(t) = \frac{1}{e} \frac{dC}{dz} \frac{dz}{dV_g} (\delta V_g)^2. \tag{6.5}
\]

In the equation above, \( z \) represents the separation between the graphene flake and the substrate. The displacement \( \xi \) of the graphene flake, under the application of a driving force \( F = F_0 e^{-i\omega t} \), will be \( \xi = \xi_0 e^{-i\omega t} \). The amplitude of oscillation \( \xi_0 \) is related to the force amplitude \( F_0 \) by the susceptibility \( \chi[\omega] \) (see Equation (2.50)).
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\[ \xi_0 = \chi F_0 \]

where,

\[ \chi[\omega] = -\frac{1}{2m\omega_0} \frac{1}{\omega - \omega_0 + \frac{\gamma}{2}}, \quad (6.6) \]

\[ m \] is the mass of the resonator, \( \omega_0 \) its natural angular frequency of resonance and \( \gamma \) is the damping coefficient, related to the quality factor (\( Q \)) of the oscillator as \( \gamma = \omega_0/Q \).

Using Equation (6.6), \( \delta N_1(t) \) and \( \delta N_2(t) \) can be written as,

\[ \delta N_1(t) = [C_0 + V_0 \frac{dC}{dz} \chi[\omega] \frac{dF}{dV} V_{ac} e^{-i\omega t} + c.c. \quad (6.7) \]

\[ \delta N_2(t) = \frac{1}{e} \frac{dC}{dz} (\chi[\omega] \frac{dF}{dV} V_{ac} e^{-i\omega t} + c.c.) (V_{ac} e^{-i\omega t} + c.c.). \quad (6.8) \]

Now we consider the effect of leading order term in \( \delta N \), \( \delta N_1 \); this term has no non-vanishing DC component, and thus only contributes to \( R_{rect} \) at second order. Therefore its contribution will be proportional to \( |\delta N_1[\omega]|^2 \). Following the Equations (6.2), (6.6) and (6.7), we find that,

\[ R_{rect} = \frac{d^2 R}{dV_g^2} (V_{ac})^2 \frac{(\tilde{\omega} + q_x)^2 + q_y^2}{\tilde{\omega}^2 + 1} \quad (6.9) \]

where,

\[ \tilde{\omega} = \frac{\omega - \omega_0}{\gamma/2}, \quad (6.10) \]

\[ q_x = -\frac{1}{C_0} \frac{dC}{dz} (V_g \frac{dC}{dV}) \left( \frac{Q}{m\omega_0^2} \right), \quad (6.11) \]

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and \( q_y = 1 \).

Next we consider the effect of the second order term \( \delta N_2(t) \) in \( \delta N(t) \). Since this term does have a non-vanishing DC component; it will thus contribute to \( R_{\text{rect}} \) at the same order in \( V_{\text{ac}} \) as the \( \delta N_1(t) \). The DC part of \( \delta N_2(t) \) has the form,

\[
\delta N_2(t) = \frac{1}{e} \frac{dC}{dz} (\chi[\omega] + \text{c.c.}) \frac{dF}{dV} |V_{\text{ac}}|^2. \tag{6.12}
\]

As this term is \( \propto \text{Re} \chi[\omega] \), it is proportional to \( \tilde{\omega} \). It will thus modify \( q_x \) as well as \( q_y \). Including this term in the expression of \( \Delta R \), we find that the new Fano parameters \( \tilde{q}_{x,y} \) as,

\[
\tilde{q}_x = q_x + \Delta q_x,
\]

\[
\tilde{q}_x = q_x - 2 \frac{dR}{dV_g} \left( \frac{1}{C_0} \frac{dC}{dz} \right) \frac{dF}{dV} \left( \frac{Q}{m\omega_0^2} \right), \tag{6.13}
\]

\[
\tilde{q}_y = \sqrt{1 - q_x \Delta q_x - (\Delta q_x)^2}.
\]

By including the higher order term, we see that its effect is to modify \( q_x \) and further it forces \( q_y \neq 1 \). In the derivation of Equation (6.9), \( R_{\text{rect}} \) is the difference in resistance from its value when no ac signal is applied at the gate. To get the change in resistance (\( \Delta R \)) as defined in main text, we subtract this contribution. Therefore,

\[
\Delta R = \frac{d^2 R}{dV_g^2} \left( \frac{(\tilde{\omega} + q_x)^2 + q_y^2}{\tilde{\omega}^2 + 1} - 1 \right) (V_{\text{ac}}^2). \tag{6.14}
\]

Equation (6.14) agrees reasonably well with the observed variation of \( \Delta R \) with magnetic field. Across the quantum Hall plateau in \( R \), curvature \( \frac{d^2 R}{dV_g^2} \) has opposite signs giving rise to negative and positive \( \Delta R \) at the resonant frequency. The origin of oscillations in resistance change (\( \Delta R \)) at low magnetic fields lies in Shubnikov-de Haas oscillations in resistance observed at low magnetic fields. Equation (6.14) involves the second derivative of resistance with gate voltage. This second order derivative of resistance with respect to gate voltage can be mapped to the second order derivative with respect to the magnetic field for a constant density of charge
Figure 6.4: a) At $B = 2.4$ T, $\Delta R$ plotted with driving frequency along with the fitted curve using Equation (6.14). The Fano-lineshape is clearly seen. b) $\Delta R$ plotted across the resonant frequency at $B = 8.2$ T. The solid line is the fit to the data giving $q_x = -0.95$ and $q_y = 1.08$. 
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carrier (at fixed gate voltage). Therefore, at low magnetic fields, when resistance oscillates, the second order derivative also changes sign leading to oscillation in $\Delta R$. Figure 6.4(a) shows the line plot of $\Delta R$ from Figure 6.3(b) at $B = 2.4$ T and the fit using Equation (6.14), which gives $q_x = -4.18$. This is close to our estimate of $q_x = -4.80$. During the quantum Hall plateau to plateau transition, the term $\frac{dR/dV_g}{d^2R/dV_g^2}$ can become significant, leading to the comparable corrections in Fano parameters $q_{x,y}$ given by the Equation (6.13). In Figure 6.4(b), we have plotted the change in resistance ($\Delta R$) for $B = 8.2$ T from the same colorscale plot as shown in Figure 6.3(b). The thin red line is the fit to the data using Equation (6.9) giving $\tilde{q}_x = -0.95$ and $\tilde{q}_y = 1.08$, which is consistent with Equation (6.13).

6.3.2 Graphene modes

It is interesting to contrast the above mentioned observations and model with some high frequency graphene modes ($\approx 100$ MHz) where we observe very small change in resistance with mechanical vibrations. However by increasing the amplitude of the driving signal (RF signal applied at the gate), we can see small changes in resistance at the resonant frequency, clearly. Figure 6.5(a) shows the measurement of resistance for a resonant mode ($\approx 139.4$ MHz) at three different RF amplitudes for another device. For the largest RF amplitude (-27.7 dBm), changes in $R$ ($\approx 10$ $\Omega$) can be seen. In Figure 6.5(b), we have shown the colorscale plot of $\Delta R$ with magnetic field for a resonant mode (56.55 MHz) from a device at 5 K. The resonator is driven with higher amplitude RF signal (-26.5 dBm). At certain fields, change in resistance of $\approx 45$ $\Omega$ can be seen. The change in resistance is also abrupt, which could be due to Duffing non-linearity at higher amplitudes.

One reason for such a behavior for graphene modes might be the assumption that electron density in the flake follows the mechanical motion adiabatically at all frequencies; this may not be correct in magnetic field when charge density splits into compressible and incompressible regions. Such a time lag between drive and response would effectively reduce the value of $\Delta R$. The second possibility is that for these high frequency device modes, the spatial amplitude profile can be very complex [30, 116]. For the modes localized at the edges, the “effective density” which gets modulated by the mechanical motion can be small as compared to gold modes where the whole
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Figure 6.5: a) Measurement of resistance for different driving amplitudes for a mode (≈ 139.4 MHz) at $V_{\text{DC}}^g = 5$ V and $B = 6$ T (just after the $\nu = 2$ plateau). b) Colorscale plot of $\Delta R$ with magnetic field for a mode (≈ 56.55 MHz). The overlaid plot is the two probe resistance (right axis) at 5 K.
6.4 Effect of quantum Hall state on graphene electromechanics

The gold modes discussed above modulate a larger fraction of the total carrier density in the flake. However, low frequency, low quality factor and poor dispersion reduce the sensitivity of these modes to the forces on the resonator. Therefore to look at the consequence of the quantum Hall state on the mechanics, namely the resonant frequency and quality factor, we turn our attention to the high frequency graphene modes with...
higher quality factor. At low temperatures, these devices show low tunability of resonant frequency with the gate voltage due to large built-in strain as shown in Figure 6.2(c). Also, with a change in the gate voltage from zero, resonant frequency decreases [29, 105]. Such a change in resonant frequency at low temperature originates from the electrostatic softening of the spring constant [105] as described in Chapter 5. Figure 6.6 shows the measurement of $I_{mix}$ using FM technique for multiple modes as a function of magnetic field at $V_g^{DC} = 5 \text{ V}$. The presence of multiple modes in a very close range of frequency indicates that many of these are possibly edge modes arising due to the soft degrees of freedom along the length of the flake [30, 116] and their dispersion with $B$ is mode dependent. In Figure 6.6, the overlaid line plot shows resistance measured with increasing magnetic field at the same gate voltage. Clearly, we see larger $I_{mix}$ signal, when change in the resistance with magnetic field is large. However, in the plateau region, the resistance does not change with $V_g^{DC}$ ($\frac{dG}{dV_{DCg}} \sim 0$), which makes the detection of resonant mode difficult in these regions. Surprisingly, for certain modes even in the plateau region, we can measure some detectable signal for mixing current (for example compare the modes at 110.5 MHz and 111.2 MHz) and it varies to a different extent across modes. These two observations suggest a mode dependent amplitude profile.

In Figure 6.7(a), we have shown $I_{mix}$ for two modes, using FM technique, with magnetic field. It is clear from the colorscale plot that the two modes disperse differently with magnetic field. In Figure 6.7(b), the non-monotonic resonant frequency shift ($\Delta f = f_0(B) - f_0(0)$) with $B$ can be seen for the two modes. At low magnetic fields, for the upper mode (115.77 MHz) $\Delta f$ increases with $B$ accompanied by a reduction in $Q$ (green color shaded area in Figure 6.7(b) and (c)). At the $\nu = 2$ plateau, $I_{mix}$ signal becomes very small ($\frac{dG}{dV_{DCg}} \sim 0$) and the estimation of $f_0$ and $Q$ becomes difficult. As $B$ is increased further, after the $\nu = 2$ plateau, $\Delta f$ starts dropping slowly without much change in $Q$ (yellow color shaded area in Figure 6.7(b) and (c)). Similar behavior can be seen for the lower frequency mode (115.124 MHz), though the effect is less pronounced (Figure 6.7(d)).
6.4. Effect of quantum Hall state on graphene electromechanics

Figure 6.7: Resonant frequency and quality factor with magnetic field \(B\) at 5 K and \(V_{g}^{DC} = 5\) V: a) Measurement of mixing current for two close-by mechanical modes with \(B\). b) Frequency shift with \(B\) by fitting the data in (a). Different behavior in frequency shift across the \(\nu = 2\) plateau is shown with shaded region. c) and d) show the variation of the quality factor with \(B\) for the two modes shown in (a) at frequency 115.77 MHz and 115.124 MHz, respectively.
6.4.1 Magnetization of quantum Hall state

To understand the dispersion of the modes with magnetic field, we examine various contributions to the total energy of the resonator ($E_{\text{tot}}$), which can be written as a sum of the mechanical energy, electrostatic energy and the contribution arising from the magnetization ($M$) of graphene ($E_{\text{mag}}$). The contribution of $E_{\text{mag}}$ ($= -M.B$) to the total energy of the resonator comes from the magnetization’s implicit dependence on $z$ through the total charge carriers ($N(z)$), where $z$ is the position of the flake from its equilibrium position and increases towards the gate.
6.4. Effect of quantum Hall state on graphene electromechanics

For the estimation of the $E_{mag}$, we first calculate the magnetization of graphene by assuming a model density of states (DOS) with Landau level’s (LL) width being the only free parameter. To calculate magnetization of the graphene in quantum Hall limit, we start with the density of states ($\rho(\epsilon)$) model with normalized Gaussians of finite width $\Gamma$ centered at the positions of Landau level eigenvalues given by $\epsilon_n = \text{sgn}(n)\sqrt{2\hbar c^2 eB|n|}$, where $c$ is the Fermi velocity ($\approx 10^6 \text{ ms}^{-1}$), $B$ is the magnetic field and $n$ is the Landau level index. Therefore, DOS can be written as,

$$\rho[\epsilon, B] = \sum_{n=-\infty}^{\infty} g_s g_v g_{LL} \left( \frac{1}{\sqrt{2\pi\Gamma^2}} \right) e^{-\frac{(\epsilon - \epsilon_n)^2}{2\Gamma^2}}, \quad (6.15)$$

where, $\Gamma$ is taken as the width of the disorder broadened Landau level (LL), $g_s = 2$ is the spin degeneracy, $g_v = 2$ is the valley degeneracy and $g_{LL} = eB/h$ is the degeneracy of the each LL. The total number of charge carriers ($N$) is set by the gate voltage, which is related to the density of states (DOS) through,

$$N = \int_{-\infty}^{\infty} \rho(\epsilon)f(\epsilon, \mu)d\epsilon, \quad (6.16)$$

where, $f(\epsilon, \mu)$ is the Fermi-Dirac distribution function. Now Equation (6.16) can be solved for the chemical potential ($\mu$) for different values of magnetic field. In Figure 6.8(a), colorscale plot shows the DOS spectrum with magnetic field. By solving Equation (6.16) numerically, we can calculate the change in $\mu$ with $B$ for a fixed number of carriers set by the gate voltage. Result of such a calculation is shown in the overlaid plot in Figure 6.8 by taking $\Gamma$ 8 meV for each LL.

In order to calculate the magnetization ($M$), we have mainly followed the general prescription described in Ref[123, 124]. Figure 6.8(b) shows result of such a calculation, where magnetization is plotted with magnetic field for different values of the flake position, where de Haas-van Alphen oscillations in $M$ with $B$ can be clearly seen. Across the $\nu = 2$ filling factor (close to $B = 4.2$ T), $M$ shows a sharp jump. The magnetization of graphene can be thought of as arising from the two parts- bulk and edge contributions. The bulk contribution is diamagnetic in nature and in the high field limit its relative contribution becomes very small, while the edge contribution dominates in the high field limit as a fraction of the total magnetization. In
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Figure 6.8(c), we plot total magnetization with magnetic field along with bulk and edge contributions. For this calculation we have taken $n = 2.2 \times 10^{-15} \text{m}^{-2}$, $T = 1 \text{K}$ and width of each LL ($\Gamma = 8 \text{meV}$). In the proximity of integer $\nu$, such a sharp change in $M$ makes it very sensitive to the position of the flake as shown in Figure 6.8(d). Here we would like to stress that it is the presence of back gate in these devices which makes $E_{mag}$ to contribute to the total energy of the resonator. For a system without gate electrode and with fixed carrier density, $E_{mag}$ will not have any $z$ dependence and hence will not contribute to $E_{tot}$. The total energy of the resonator ($E_{tot}$) can be written as,

$$E_{tot}(z, B) = E_{mech} + E_{elec} + E_{mag},$$

where, $E_{mech} = \frac{1}{2}kz^2$; $E_{elec} = -\frac{1}{2}C_g(z)V^2_g$; and $E_{mag} = -M(z)B$.

Assuming a parallel plate geometry, the total energy of system can be minimized with respect to $z$. By minimizing the $E_{tot}$ with respect to $z$, the equilibrium position of the flake ($z_0$) with varying $B$ can be calculated and it is shown in Figure 6.9(a). It is clear that before the $\nu = 2$ plateau as we increase the field, the flake moves closer to the substrate. Another important effect of $E_{mag}$ on the mechanics of the resonator is to modify the spring constant ($k_{eff} = \frac{d^2E_{tot}}{dz^2}|_{z=z_0}$) which leads to the frequency shift. In Figure 6.9(b), we show the change in resonant frequency calculated from $k_{eff}$ with varying $B$. From this model calculation, we can see that with an increase in $B$ on either side of the $\nu = 2$ plateau, contribution of $E_{mag}$ leads to the softening of $k_{eff}$ and reduction in the resonant frequency. This is consistent with our experimental observation beyond $\nu = 2$, where we observe a decrease in $\Delta f$ (yellow color shaded area in Figure 6.7(b) and Figure 6.9(b)).

However, before the $\nu = 2$ plateau, we observe larger dissipation accompanying the increase in $\Delta f$ (green color shaded area in Figure 6.7(b) and (c)), which indicates that the contribution of $E_{mag}$ to $k_{eff}$ is not enough to fully understand such behavior and there are more subtle effects which arise when the flake moves closer to the substrate. To gain further insight into the device behavior, we also characterize the device modes with different driving amplitudes. In Figure 6.10(a), we have shown the dispersion of the mode. The colorscale plot shows the mixing current measured using FM technique.
Figure 6.9: a) Equilibrium position of the flake ($z_0$) with $B$. Two different regimes across $\nu = 2$ plateau are shaded with colors. b) Change in resonant frequency with $B$ calculated from $k_{eff}$. 
at 5 K. Figure 6.10(b) and (c) show the lineshape plots of mixing current for different amplitudes of frequency modulated RF signal for two gate voltages indicated by the white arrows on the colorscale plot of Figure 6.10(a). At $V_{g}^{DC} = 2$ V, we observe neither any change in resonance frequency ($f_0 \approx 115.95$ MHz) nor any significant change in the quality factor ($Q \approx 1295$) for different driving powers. However, at $V_{g}^{DC} = 5$ V, we can clearly see a shift ($\approx 50$ KHz) in the current minima towards the higher frequency side and also the shape of the curve develops an asymmetry.

Here, we note that enhancement of linear damping alone cannot explain the observed frequency shifts with magnetic field. The frequency shift due to linear damping ($\Delta f \approx -\frac{f_8}{8Q^2}$) would be very small ($\approx$ -10 Hz, for the two modes), whereas we observe much larger positive $\Delta f$. It strongly suggests that there are other damping mechanisms present in the system. The basic characterization of the device, in zero magnetic field as described above, shows that nonlinear damping mechanism [125] can become important as the flake moves closer to the substrate. To quantitative understand the nonlinear damping mechanism appearing in magnetic field (due to the flake movement closer to the substrate), we numerically solve the equation of motion (EOM) for doubly clamped membranes mapped to the equation of a simple harmonic oscillator with a single degree of freedom [75] (See section 2.7),

$$m\dddot{z} + \Gamma \dot{z} + m\omega_0^2 z + \alpha z^3 + \eta z^2 \dot{z} = F_0 \cos \omega t,$$

(6.18)

where, $m$ is the effective modal mass, $\Gamma$ is the linear damping rate, $\alpha$ is the Duffing parameter, $\eta$ is the coefficient of nonlinear damping and $F_0$ is the amplitude of the driving force. In order to extract various parameters of the EOM, we numerically solve it for the response of the resonator (amplitude $z_0$ and phase $\phi$) using Equation (2.47) and (2.48) by taking effective modal mass $m$ as 0.7 times the actual mass of the graphene sheet and linear damping rate $\Gamma$ set to be a constant to its value calculated at very low powers where nonlinear damping can be ignored. Force amplitude $F_0$ can be calculated using Equation (3.2). After this, we calculate the mixing current measured in FM technique using Equation (3.20) and fit it to the experimental data by treating $\alpha$, $\eta$ and $\omega_0$ as fitting parameters. As this fit is achieved by fitting discrete experimental data to discrete calculated values (As closed form of the amplitude is not available), in order to minimize $\chi^2$, we use Nelder-Mead optimization technique [126].
6.4. Effect of quantum Hall state on graphene electromechanics

Figure 6.10: a) Modal dispersion of a mode with gate voltage at 5 K. The colorscale plot of the mixing current (X-component at the lock-in) measured using frequency modulated (FM) technique. b) and c) are the lineshapes of mixing current for different values of excitation amplitude (labeled in the panel) of the FM signal at $V_g^{DC} = 2$ V and $V_g^{DC} = 5$ V (indicated by the white arrows shown in (a)), respectively.
Chapter 6. Graphene NEMS in quantum Hall regime

For the data shown in Figure 6.9(a) we get typical values of $\alpha = 1.5 \times 10^{13} \text{ kgm}^{-2}\text{s}^{-2}$ and $\eta = 2 \times 10^4 \text{ kgm}^{-2}\text{s}^{-1}$ which are comparable to the previous estimates of these two parameters [125]. For our observations, mechanical response before the $\nu = 2$ plateau, the frequency shift and quality factor decrease can be explained by the movement of the flake closer to the substrate, which leads to the enhanced damping and frequency shift.

6.5 Electromechanics across $\nu = 1$ state

For similar measurements on clean graphene devices at $T = 100 \text{ mK}$, the consequence of the chemical potential jumps in gapped regions of DOS gets enhanced. In Figure 6.11(a), we show the variation in resistance and the resonant frequency of a device with magnetic field. In the plot of resistance as a function of magnetic field, the plateau at $\nu = 2$ and $\nu = 1$ can be clearly seen. The plateau at $\nu = 1$ originates from the interaction induced broken symmetries of zeroth LL. Previous experiments seem to suggest that LL gap for $\nu = 1$ is related to the broken valley degeneracy and not to the spin [127]. In the resonant frequency, we see jumps at certain fields over a monotonic decrease in resonant frequency. The jumps in resonant frequency can be thought of as arising from the magnetization changes as magnetic field sweeps the chemical potential across the gaps induced by the interactions. If a smooth parabolic fitted background of frequency is subtracted jumps in resonant frequency can be accentuated as shown in Figure 6.11(b). The origin of such an overall background of frequency in our samples is not clear to us. However, previous measurements on magnetic two dimensional electron gas [48] has suggested that such a background can arise from the component of the magnetic moments from the paramagnetic ions present in the magnetic 2DEG. An overall decrease in resonant frequency and jump at $\nu = 1$ plateau is beyond our model calculations and need further studies.
6.5. Electromechanics across $\nu = 1$ state

Figure 6.11: a) Measurement of resistance and resonant frequency at $T = 100$ mK and $V_g^{DC} = 3$ V. b) Frequency shift after removing a smooth parabolic background of frequency from the data shown in (a).
Chapter 6. Graphene NEMS in quantum Hall regime

6.6 Effect of chemical potential change due to electrostatics

The change in chemical potential with magnetic field can also affect the mechanical response by an electrostatic force. However, in devices with small dispersion like the ones studied here, this correction is small. Below we discuss how our present model can be modified to incorporate this effect. The electrostatic contribution of the change in chemical potential ($\mu$) is relatively small compared to the effect of the magnetic contribution. To account for this, $E_{\text{elec}}$ can be modified to $\frac{1}{2} C_g (V_g^{DC} - \frac{\mu}{e})^2$. Since, the cyclotron gap is the largest for the $\nu = 2$ gap, the largest change in the chemical potential occurs across it due to changing magnetic field. For the density of states used in the model calculations and the carrier density set by the gate voltage, change in chemical potential across $\nu = 2$ plateau is approximately 50 meV as seen in the overlaid plot in Figure 6.8(a). Below we have plotted the dispersion of the upper mode with gate voltage discussed in Figure 6.9. The colorscale plot of dispersion for this mode is shown in Figure 6.10(a). From the dispersion, we can say that, at $V_g^{DC} = 5V$, effect of chemical potential change on resonant frequency across the $\nu = 2$ plateau would result in frequency shift $\approx 10$ KHz which is smaller than the frequency shift we observed in Figure 6.7. However, the chemical potential jump through the magnetic energy contribution affects the resonant frequency by a larger amount. In Figure 6.12 we have reproduced the results of Figure 6.9(a) and (b), by taking into the correction to the electrostatic energy contribution. It can be seen from these calculations, the correction due to the chemical potential to electrostatic energy results in very small corrections to equilibrium position of the flake and frequency shifts.

6.7 Frequency shift for a hypothetical device with higher tunability

The coupling between the magnetization of the quantum Hall state and mechanics of the resonator can be made large by engineering devices to have small in-built tension at low temperature (with large tunability with gate voltage). Figure 6.12(c)
6.7. Frequency shift for a hypothetical device with higher tunability

Figure 6.12: a) Equilibrium position of the flake ($z_0$) calculated by considering the chemical potential correction to the electrostatic contribution to the total energy (blue line). Plot of $z_0$ without considering this correction is also shown (red square) for comparison (same as Figure 6.9(a)). b) Change in resonant frequency calculated from $k_{eff}$ by considering the chemical potential correction to the electrostatic contribution to the total energy (blue circle). Plot of frequency shift without considering this correction is also shown (red circle) for comparison (same as Figure 6.9(b)). c) A hypothetical dispersion of resonant frequency with gate voltage. A large positive tunability can be seen in devices with low in-built tension, like the one shown here. d) Resonant frequency change as a function of magnetic field calculated using the model.
Chapter 6. Graphene NEMS in quantum Hall regime

shows the calculated dispersion from a hypothetical device by adjusting the spring constant. By following the calculations described above, we calculated the resonance frequency with magnetic field. Figure 6.12(d) shows the frequency shifts as a function of magnetic field for this hypothetical device with large dispersion. Substructures in frequency shift for higher filling factor (towards the low magnetic field) can be seen too. The small frequency shifts we observe are a consequence of the small negative dispersion and high in-built tension. If devices were to be specifically fabricated with less in-built tension then the dispersion, with gate voltage, would be larger; this would result in larger frequency shifts.

6.8 Summary

In this chapter, we have described that with high electronic mobility samples (due to current annealing), a coupling between the quantum Hall state and electromechanics can be observed. Our observation and quantitative analysis of resistance change giving a Fano-lineshape helps in better understanding the effect of electromechanical drive of graphene resonators. Also, the magnetization of graphene couples to the mechanical motion to modify the spring constant. The results can be further enhanced in devices with larger tunability of resonant frequency with gate voltage. We would like to emphasize that the change in the spring constant is a consequence of the coupling between magnetization and the carrier density set by the gate voltage. Therefore such results should be possible to see in other systems where the magnetization can be modified by both electrostatics and magnetic field.
Chapter 7

Dual top gated graphene transistor in the quantum Hall regime

This chapter describes the study of local modulation of charge density and carrier type in graphene field effect transistors using a double top gate geometry and the experiments were performed in collaboration with Ajay K. Bhat and Sunil Patil. The two top gates lead to the formation of multiple $p$-$n$ junctions. First we discuss the details about the device fabrications; this is followed by the electron transport measurements at low temperature and in the presence of magnetic field showing various integer and fractionally quantized conductance plateaus. These results are explained by the mixing of the edge channels and we find that inhomogeneity plays an important role in defining the exact quantization of these plateaus. The results presented here have been published in Ref[128].

7.1 Introduction

We study electron transport in a graphene multiple lateral heterojunction device with charge density distribution of the type $q-q_1-q_2-q$ with independent and complete control over both the charge carrier type and density in the three different regions. This is achieved by using a global back gate (BG) to fix the overall carrier type and density and local top gates (TG$_1$, TG$_2$) to set the carrier type and density only
Chapter 7. Dual top gated graphene transistor in the quantum Hall regime

below their overlap region with the graphene flake. By controlling the density under the two top gates, various conductance plateaus can be observed in quantum Hall regime. We explain these results by mixing and partitioning of the edge channels at the junctions. Our analysis on these two probe devices indicates that aspect ratio and inhomogeneity play an important role in determining the quantization of the conductance plateaus. The issues we explore here are also important in the use of graphene heterojunctions for collimation \[57\] and broad field of metamaterials based on graphene heterostructures \[129\].

7.2 Device fabrication

The device fabrication starts with the mechanical exfoliation of graphene flakes from graphite \[12, 13\] on 300 nm SiO\(_2\) grown on degenerately doped silicon substrates as described in detail in Chapter 3. Using e-beam lithography, source-drain contacts were fabricated by depositing 10 nm/50 nm of Cr/Au by thermal evaporation. For the fabrication of top gates, we first spin coat 30 nm of NFC 1400 (JSR Micro) \[130\] as a buffer layer followed by the 10 nm of HfO\(_2\) using atomic layer deposition to serve as the dielectric. Following this we fabricate two 500 nm wide top gates with a gap of 2 \(\mu\)m. Figure 7.1(a) shows a schematic of the device where, \(V_{bg}(V_{tg1}, V_{tg2})\) is the back (top) gate bias and \(I_{ac} (\sim 50 \text{ nA})\) is used to measure the two probe resistance of the device using the lock-in technique.

7.3 Experimental results and discussion

To understand the effect of local charge density and type modulation, we start by measuring the resistance with voltages applied at back gate and at one of the top gates while the other top gate voltage set to 0 V. Figure 7.1(b) shows the colorscale plot of such a measurement. The global maximum in resistance corresponds to the charge neutrality point due to \(V_{bg}\) variation. Figure 7.1(c) shows line plots from the colorscale plot in Figure 7.1(b) while the top gate is set at 0 V. The Dirac point is shifted to 23 V due to unintentional doping during the fabrication of the devices.
7.3. Experimental results and discussion

Figure 7.1: (a) Schematic diagram of the device. (b) Colorscale plot of two probe resistance as a function of $V_{bg}$ and $V_{tg1}$ at $T=1.7$ K with $V_{tg2}=0$ V. (c) Line plots of slices of data shown in (b) at indicated top gate voltages. (d) Colorscale plot of resistance as a function of $V_{bg}$ and $V_{tg1}$ at $B=6$ T. (e) Line plots of slices of data shown in (d) as indicated by the colored lines.
Chapter 7. Dual top gated graphene transistor in the quantum Hall regime

From the resistance variation with back gate voltage, we measure the carrier mobility to be \( \sim 4800 \, \text{cm}^2\text{V}^{-1}\text{s}^{-1} \). A local maxima in resistance (reflected in the angular band on the colorscale plot) is also observed. The red curve in Figure 7.1(c) is the line plot from the colorscale plot shown in (b) with top gate being set at 1.5 V. A clear local maximum at \( \sim 40 \, \text{V} \) can be seen which is due to the effect of the top gate which is biased at 1.5 V. This can arise from the charge neutrality under the top gate. When the graphene region under the top gate is charge neutral it adds the largest resistance in series with the graphene leads (portion of graphene not overlapping with top gates) giving a local maxima in resistance. Since the total carrier density under the top gate is given by \( n_{tot} = C_{bg}V_{bg} + C_{tg}V_{tg} \), sweeping both gates (back gate and top gate) results in an angular band on the colorscale plot. By following the evolution of two resistance peaks due to the top gate and back gate, relative capacitive coupling (\( \eta \)) of the top gate with respect to the back gate can be calculated [131]. From the data shown in Figure 7.1(b), we calculated \( \eta \) to be \( \sim 7 \). Capacitance calculated by taking into account the dielectric coefficient of HfO\(_2\) and NFC [130] gives an \( \eta \) of 5.8.

Figure 7.1(d) shows the colorscale plot of resistance of the device with voltages applied at back gate and at one of the top gates (while other top gate voltage is set to 0 V) in the presence of a 6 T magnetic field applied perpendicular to the plane of graphene. The first thing to be noted is the formation of the quantum Hall plateaus when the top gate is fixed at 0 V. Figure 7.1(e) shows line plots of Figure 7.1(d) with the green plot showing the plateaus as a function of the back gate. The plateaus corresponding to \( \nu = 2 \) and 6 can be seen. Also seen at \( V_{tg} \neq 0 \, \text{V} \) are the fractionally quantized conductance plateaus which arise due to the equilibration of the edge channels interacting at the junctions [59]. The plateaus occurs at slightly higher values than that expected from the exact quantization. This is possibly due to finite contact resistance, as this is a two probe measurement. Also, finite longitudinal conductivity (\( \sigma_{xx} \neq 0 \)) in rectangular graphene devices can cause deviation from the ideal quantization [59, 97]. The red plot in Figure 7.1(e) shows the plateaus that are not expected for monolayer graphene and arise due to the effect of the top gate.

We next consider the central experiment that studies the interaction of the edge channels induced by two top gates. Figure 7.2(a) shows the conductance(G) of the device (in units of \( G_0 = e^2/h \)) as a function of \( V_{tg1} \) and \( V_{tg2} \) with the back gate \( (V_{bg} = 31.1 \, \text{V}) \) fixing the overall flake at the \( \nu = 2 \) plateau at B=6 T. We observe many
7.3. Experimental results and discussion

Figure 7.2: (a) Measured conductance (in units of $G_0 = e^2/h$) of the device with the voltages applied at two top gates while $V_{bg} = 31.1\ V$ corresponding to the $\nu = 2$ at $T = 1.7\ K$ and $B = 6\ T$. (b) Line plots from (a) at the slices indicated by the colored lines on it to show the observed conductance plateaus.
fractionally quantized conductance plateaus arising due to the interactions between the edge channels induced below the two top gates mediated via the intermediate graphene lead. Figure 7.2(b) shows line plots for slices of data shown in Figure 7.2(a) with one of the top gates being varied continuously, while the other top gate and back gate are set at $\nu=2$ plateau. Another feature of the data shown in Figure 7.2(a) is that there are fluctuations in the conductance plateau values that extend over a range of top gate values; seen as horizontal bands at fixed values of $V_{tg2}$. This observation can be understood as resonant reflection due to impurities embedded underneath the top gate [132] (we discuss the role of impurities later). Varying $V_{tg2}$ modifies the chemical potential and that changes the amplitude of scattering from delocalized to localized states.
7.4 Current equilibration model

To explain the observed data we employ a simple model as shown in Figure 7.3(a). Here the red/green channels represent electrons/holes. This shows just one of the various possible configurations of the regions [60]. We now solve the system of equations framed using current conservation at each of the junctions with the reflection coefficients of the edge channel currents determined by the type of charge and also the number of channels present. Let \( \nu_1 \) and \( \nu_2 \) be the filling factors present under the two top gates with \( \nu \) being the filling factor set by the overall back gate. In all there are three configurations of junctions possible.

- The first possibility is when both \( \nu_1 \) and \( \nu_2 \) are of the same carrier type but \(|\nu_2| < |\nu_1|\) in which case only the channels present in \( \nu_2 \) are transmitted giving a reflection coefficient \( r_1 = 1 - \frac{\nu_2}{\nu_1} \) into region 1.

- The second case is when \( |\nu_1| < |\nu_2| \) leading to circulating channels from among \( \nu_2 \) in region 2. This case is shown in Figure 7.3(a) between \( \nu \) and \( \nu_1 \) where \( r_1 = 1 - \frac{\nu_1}{\nu} \) into the region TG1. In our device such circulating channels can be formed even in the region between TG1 and TG2 for the case when \( |\nu_1| < |\nu| \).

- The remaining possibility is that of \( \nu_1 \) and \( \nu_2 \) being of different charge carrier types, i.e. electrons on one side and holes on the other side. This leads to the complete mixing of electron and hole channels at the junction.

In Figure 7.3(a) this is shown by \( \nu \) and \( \nu_2 \) giving a reflection coefficient of \( r_2 = \frac{\nu_2}{\nu_2 + \nu} \) into region below TG2. Using current conservation at each interface leads to the following set of equations;

\[
\begin{align*}
I_1 &= I + I_4 \text{ and } I_2 = r_1 \times I_1 \\
I_3 &= I_2 + I_7 \text{ and } I_4 = r_1 \times I_3 \\
I_5 &= I_1 - I_2 \text{ and } I_6 = I_5 + I_8 \\
I_7 &= I_6 - I_{10} \text{ and } I_8 = r_2 \times I_9 \\
I_9 &= I_{10} \text{ and } I_{10} = r_2 \times I_6
\end{align*}
\]
where $I_i$ represent the current due to channel $i$ as labeled in Fig 7.3(a). This set of linear equation can now be solved to give a relation between the final output current (for example $I_9$) and input current ($I$). Replacing the ratio of output current and input current by $\nu_{eff}\frac{e^2}{h}$ gives an effective filling factor $\nu_{eff}$. Similarly solving for all the possible configuration of the local filling factors leads to the effective filling factors for a range of filling factors under the two top gates. These results are summarized in the table below.

Table 7.1: Table listing the results of effective filling factor for different configuration of densities under the two top gates.

<table>
<thead>
<tr>
<th>Case</th>
<th>Effective filling factor ($\nu_{eff}$)</th>
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<tbody>
<tr>
<td>$\nu_1 &lt; 0$ and $\nu_2 &lt; 0$</td>
<td>$\frac{</td>
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<tr>
<td>$\nu_1 &lt; 0$ and $\nu_2 \geq 0$</td>
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<td>$\nu_1 \geq \nu$ and $\nu_2 &lt; 0$</td>
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<td>$\nu_1 &gt; \nu$ and $\nu_2 \geq \nu$</td>
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<tr>
<td>$0 &lt; \nu_1 &lt; \nu$ and $\nu_2 \geq \nu$</td>
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<tr>
<td>$\nu_1 \geq \nu$ and $0 &lt; \nu_2 \leq \nu$</td>
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<td>$\nu_1 \leq \nu$ and $0 &lt; \nu_2 \leq \nu$</td>
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<td>$0 &lt; \nu_1 \leq \nu$ and $\nu_2 &lt; 0$</td>
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<td>$0 &lt; \nu_1 \leq \nu$ and $0 &lt; \nu_2 \leq \nu$</td>
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</table>

The effective filling factors calculated above gives the two probe conductance $G$ of the device assuming a vanishing longitudinal microscopic conductivity $\sigma_{xx} = 0$. Under finite $\sigma_{xx}$, two microscopic conductivities $\sigma_{xx}$ and $\sigma_{xy}$ can make the two probe sample conductance $G$ aspect ratio $\frac{W}{L}$ dependent, where $W$ and $L$ are the width and length of the sample, respectively. The graphene leads (area not under the top gates) have almost unit aspect ratio, however, region under the top gates have an aspect ration of almost 3 and under finite $\sigma_{xx}$ it can affect $G$. A measure of the inhomogeneity in charge density can be obtained from the width of the Dirac peak as a function of $V_{tg1}$ and $V_{tg2}$ ($\Delta n = C_{tg} \times \Delta V_{tg}$). For the two top gates, $\Delta n$ turns out to be $7.9 \times 10^{11}/cm^2$ and $5.4 \times 10^{11}/cm^2$. This gives an estimate of the impurity doping in the sample. To take these aspects into account, we have followed the model developed by Abanin [133], where macroscopic conductance $G$ of the sample can be related to the microscopic conductivities and aspect ratio of the device. The
7.4. Current equilibration model

Microscopic conductivities are calculated using a model density of states formed with finite width Landau levels. Following this approach, we first calculate the corrected filling factors for the regions under the two top gates by taking an aspect ratio of 3. Current conservation at each junction, as described before, gives the two terminal conductance \( G = \nu_{\text{eff}} e^2 / h \).

Using the effective filling factors now obtained we calculate the conductance and plot it in Figure 7.3(b). Comparing Figure 7.2(b) with the line plot of Figure 7.3(b) shown in Figure 7.3(c), we see a reasonable agreement between them. Further refinement of the calculation can be done. This calculation is however still unable to explain the diagonal asymmetry and the peaks and valleys in Figure 7.2(a) deviating from flat plateaus in conductance as expected from Figure 7.3(b). There could be three possible mechanisms that can cause such a deviation – firstly, depending on the geometry of the flake this quantization is susceptible when aspect ratio deviates from unity [97, 134]. Secondly, it can arise due to inhomogeneities present under the locally top gated region, which can affect the exact quantization of the plateaus. The distribution of the impurity below the two top gates can lead to differences in their effect on the conductance quantization, including the oscillations seen in the modulation due to \( TG_2 \), which is absent in the modulation due to \( TG_1 \) [132]. This shows that there is an asymmetry in the properties of the regions below the two top gates leading to the asymmetry seen in Figure 7.2(a). Further, the \( \nu = 2 \) plateau has been found to be more susceptible to inhomogeneity [135] under the locally gated region as compared to the other plateaus when the leads are set at the same \( \nu \), which is \( \nu = 2 \). Lastly, we speculate that the mismatch in the conductance plateaus from ideal values is possibly due to unintentional dopants leading to multiple uncontrolled \( p-n \) junctions [136, 137].

Figure 7.4 shows histograms of the conductance plots, both experimental and calculated. It is clear that there is a good correspondence between the two in terms of the overall trend and the expected features. The peaks at the calculated filling factor values can be seen in the histogram from the measured data. The histogram also gives a sense for the mismatch present in the sample shown by the spread of the expected peaks.
Figure 7.4: (a) Histogram of the conductance data plotted in Figure 3(b). (b) Plots a histogram of the experimentally measured conductance shown in Figure 2(a).

7.5 Summary

In summary, we have studied the effect of two independently locally gated regions on the conductance of graphene in quantum Hall regime. We have been able to study the equilibration of the channels at multiple junctions giving rise to fractional quantized conductance plateaus and also the critical role that impurity plays.
Chapter 8

Summary and Outlook

In this thesis, we have described electron transport measurements and electromechanical measurements in devices of two kinds, namely on substrate devices and suspended graphene devices. The electron transport measurements are performed in quantum Hall regime. The electromechanical measurements are performed from room temperature to low temperatures (7 K). Later, we study the coupling between these two aspects of graphene by performing electromechanical measurements in quantum Hall regime on ultra-clean samples. Towards the end, we also probed electron transport in dual top gated graphene transistor in quantum Hall regime. During the course of the work presented in this thesis, a lot of progress have been made in the fast moving area of graphene research. In this chapter, we will summarize the results discussed in previous chapters and present an outlook based on the current experimental and theoretical findings.

In Chapter 4, we described the results on the breakdown of the quantum Hall effect in graphene. By injecting large DC current quantum Hall state can be brought out of the equilibrium. The critical current, defined at the onset of the dissipation, shows correlation with the cyclotron gaps. Our measurements also show that inter Landau level scattering assisted by the charge carrier inhomogeneities is the dominant one to cause the dissipation in the system. Under large transverse electric field, we also observed that transverse conductivity shows a current invariant point at $B = 10$ T, which could be a signature of the lifting of the spin degeneracy due to transverse Hall
Chapter 8. Summary and Outlook

electric field.

The samples studied in Chapter 4 were of moderate mobility ($\sim 10000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$). Recently it has been demonstrated that very large carrier mobility in on-substrate graphene Hall bar devices can be achieved by making these devices on boron-nitride (BN) substrate. With these high mobility samples correlated states arising from the electron-electron interaction have been observed [138]. It would be interesting to perform breakdown experiment on these high mobility graphene devices, where charge carrier inhomogeneity and width of Landau levels would be much smaller. Such a study should be more helpful in pin pointing the breakdown mechanism clearly. Another interesting effect which has been predicted is the collapse of the Landau level structure in the presence of transverse electric field. In crossed electric and magnetic field, the Landau level energy spectrum gets scaled by Lorentz boost factor ($\sqrt{1-\beta^2}$), where $\beta$ is the ratio of group velocity $E/B$ to the Fermi velocity in graphene ($v_F = 10^6 \text{ ms}^{-1}$). In the limit $\beta > 1$, a collapse of Landau level is predicted [92]. Such a collapse of Landau levels has been observed in top gated devices, where large electric field perpendicular to graphene flake can be applied in a local region [55]. Signature of the collapse of Landau levels can be observed in the conductance of graphene through the locally gated graphene region. It would be interesting to perform experiment on these high mobility samples where a crossed electric field can be applied with the help of closely spaced side gates. For the current invariant point observed in transverse conductivity, it would be interesting to perform this experiment at higher magnetic field ($B = 14 \text{ T}$), a clearer splitting could be seen with the injected current (hence, transverse electric field).

In Chapter 5, we studied graphene nanoelectromechanical system (NEMS) at low temperatures. We present a model incorporating the electrostatics in graphene NEMS and explain the modal dispersion of the resonant frequency with gate voltage. We measure the thermal expansion coefficient of suspended graphene. Such measurement of thermal expansion coefficient can be quite useful for the optimal electromechanical properties that are desirable for temperature-compensated sensors [115]. We also observed enhanced dissipation (reduction in $Q$) close to the Dirac peak. The measurements were performed on not so clean devices ($\rho \sim 6\rho_0$, where $\rho_0$ is the mass density of pristine graphene). However, it was shown later (as seen in Chapter 6) that these devices can be cleaned by passing a large current through it and ablating
the residue leading to better electronic performance. It would be interesting to probe the dissipation in the system close to the Dirac point \( (V_D) \) with direct RF-readout technique \[139\]. Heterodyne mixing techniques fail to work close to the Dirac point as for clean devices, \( V_D \) lies close to zero gate voltage. Also a better insight into the thermal expansion coefficient can be gained with these cleaner devices.

In Chapter 6, we studied coupling between quantum Hall state and electromechanics in graphene NEMS. The electromechanics of graphene affects the quantum Hall state by modulating the charge carrier density \( n \) at the rate of resonant frequency. Therefore, a time averaged measurement of resistance changes and becomes significantly observable at the points where the nonlinearity in resistance with respect to \( n \) is large. Further, a direct coupling of the driving voltage to the carrier density leads to the interference resulting in a Fano-lineshape change in resistance across the resonance. On the other hand, we find change in resistance for high frequency graphene mode to be much smaller. This could possibly be due to the time lag between the driving force and the density response or due to the complex spatial amplitude profile. It would be interesting to perform time-resolved measurement in quantum Hall regime to probe the dynamics of compressible and incompressible charge densities. Optical or piezo based methods can be used to avoid a direct coupling of RF-signal to conductance of the flake. Further, by modifying the device geometry, devices with low in-built tension can be studied for the fundamental mode of vibration, which is easier to correlate with the model presented. Another aspect of these measurements is the modification of the electromechanics of graphene due to the presence of quantum Hall state. Due to the presence of back gate in these devices, the magnetization \( M \) of quantum Hall state becomes a function of the gap between the flake and the gate electrode. This leads to a coupling between electromechanics and quantum Hall state through magnetization. The frequency shifts with magnetic field across different Landau levels, gives information of the magnetization revealing the density of states of the system. Across the Landau levels, these frequency shifts also accompanied by the movement of flake closer or away from the gate electrode. It would quite interesting to check whether by applying magnetic field, thermal energy induced motion of resonator can be reduced leading to an effective cooling of the resonator.

In Chapter 7, we have studied the dual top gated graphene transistor in the quantum Hall regime. With a local control over charge carrier density and charge
carrier type, we were able to study the equilibration of the channels at multiple junctions giving rise to fractional quantized conductance plateaus. We present model calculations to understand it and find that impurity plays an important role in the robustness of the quantum Hall plateau in two probe measurement. The top gates used in this study were quite wide (\(~\sim 500\) nm) when compared to the mean free path (\(~\sim 40\) nm) for typical device parameter. With the fabrication of thin gates, signature of Klein tunneling has been observed in graphene \cite{52,53}. Electrons tunneling out of the barrier have collimated momentum. It is an interesting question to pose whether the presence of multiple narrow top gates can lead to a ballistic transport under the top gates. This can be probed by measuring the four probe resistance under the top gates.
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