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UNIVERSITY OF CALIFORNIA
RIVERSIDE

Transport Properties of Few-Layer Graphene on Boron Nitride

A Dissertation submitted in partial satisfaction
of the requirements for the degree of

Doctor of Philosophy

in

Physics

by

Bin Cheng

March 2015

Dissertation Committee:

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Acknowledgments

I am grateful to my advisor, without whose help, I would not have been here.

To my parents

ABSTRACT OF THE DISSERTATION

Transport Properties of Few-Layer Graphene on Boron Nitride

by

Bin Cheng

Doctor of Philosophy, Graduate Program in Physics
University of California, Riverside, March 2015
Dr. Marc Bockrath, Chairperson

The electronic transport properties of graphene on hexagonal boron nitride (hBN) have been studied intensively in the past few years, and rich phenomena such as the fractal Hofstadters butterfly energy spectrum and Landau level crossing have been observed. This thesis reports our transport measurements on few-layer graphene on hBN. First, I present our bilayer graphene devices in which the bilayers are aligned to the hBN lattice, creating a superlattice potential due to the lattice and angular mismatch. The energy band structure is strongly altered at the edges of the reduced Brillouin zone, and can be tuned by a perpendicular electric field. As a strong magnetic field turned on, the Hofstadters butterfly spectrum emerges which can also be tuned by electric field. The valley symmetry breaks due to the asymmetric coupling to the aligned hBN, while the valley symmetry is dramatically restored at a critical electric field, with the electron and hole doped regimes showing the opposite critical electric field. I then show our data from twisted trilayer graphene samples with a large twist angle. The Landau levels of massive fermions from the bilayer graphene cross with those of the massless fermions from the monolayer graphene, producing a unique Landau level crossing spectrum.

Contents

List of Figures	viii
1 Introduction	1
1.1 Electronic Properties of Monolayer Graphene	4
1.2 Electronic Properties of Bilayer Graphene	10
1.3 Quantum Hall Effect in Graphene	12
1.4 High Quality of Graphene on Boron Nitride	15
2 Layer Stacking Transfer and Edge Contact Fabrication	17
2.1 Layer Stacking Transfer by Elvacite	17
2.2 Layer Stacking Transfer by Wet Etching	19
2.3 Edge Contact Fabrication	22
3 Bilayer Graphene/Boron Nitride Superlattices	27
3.1 Basic Properties of the Hofstadter's Butterfly in Graphene/hBN Superlattices	27
3.2 Raman Spectroscopy of Aligned Graphene with Boron Nitride	29
3.3 Displacement Field Dependence of Second and Third Generations of Satellite Points in Bilayer Graphene Superlattice	32
3.4 Tunable Hofstadter's Butterfly in Bilayer Graphene Superlattice	36
3.5 Twisted Bilayer Graphene with the Bottom Layer Graphene Aligned to the hBN Lattice	42
4 Landau Level Crossing and Screening in Twisted Trilayer Graphene	51
4.1 Introduction to Twisted Bilayer and Twisted Trilayer Graphene	51
4.2 Tunable Filling Ratio between Monolayer and Bilayer graphene	54
4.3 Landau Level Crossing at $B = 8$ T	59
5 Conclusions	70
Bibliography	72

List of Figures

1.1	Graphene Lattice Structure	5
1.2	Graphene Reciprocal Lattice	6
1.3	Graphene Band Structure	7
1.4	Monolayer Graphene Low Energy Band Structure	8
1.5	Bilayer Graphene Lattice	10
1.6	Bilayer Graphene Band Structure	11
1.7	Monolayer Graphene Landau Levels	13
1.8	Bilayer Graphene Landau Levels	14
1.9	hexagonal boron nitride lattice and Graphene Lattice	15
2.1	SEM Image Graphene on Boron Nitride	18
2.2	Layer Stacking Transfer	19
2.3	HBN/G/hBN and Twisted Trilayer	20
2.4	Process of Wet Etch Transfer	21
2.5	BN on Wafer	22
2.6	Etched hBN on Elvacite	22
2.7	Schematic Diagram of Edge Contact	23
2.8	Edge Contact Fabrication Process	23
2.9	Edge Contact Fabrication 2	24
2.10	Optical Image of Hall Bar Geometry	26
3.1	Raman of Aligned Monolayer and Bilayer Graphene on hBN	30
3.2	Statistics of 2D Peaks	32
3.3	Linear Fit of 2D Peak Width vs Superlattice Wavelength	33
3.4	Bilayer Superlattice Transport Measurement at $B = 0$ T	34
3.5	Displacement Field Dependence of Satellite Point	37
3.6	Displacement Field Dependence of Satellite Points in Device 2	38
3.7	Displacement Field Dependence of Satellite Points in device 3	39
3.8	Electric Tunable Hofstadter's Butterfly	40
3.9	Landau Level Transition near the Charge Neutrality Point	46
3.10	Landau Level Splitting at Positive and Negative Electric Field	47
3.11	Electron-Hole Asymmetry at Bilayer Graphene Superlattice	48
3.12	Large Angle Twisted Bilayer Graphene Raman Signal	48
3.13	Twisted Bilayer Graphene with Bottom Layer Graphene Superlattice at $B = 0$ T	49

3.14 Twisted Bilayer Graphene with Bottom Layer Graphene Superlattice at Non-zero B	50
4.1 Energy Band Structure of Twisted Bilayer and Trilayer Graphene	52
4.2 Longitudinal Resistance of Twisted Trilayer Graphene at Zero Magnetic Field	57
4.3 Landau Level Crossing at $E = 0$ mV/nm in Twisted Trilayer Graphene	62
4.4 Landau Level crossing at twisted trilayer graphene with different filling rate	63
4.5 Landau Level Interval Change in Twisted Trilayer Graphene	64
4.6 Landau Level Crossing with Broadened Energy Levels	65
4.7 Landau Level Crossing at $B = 8$ T in Twisted Trilayer Graphene	66
4.8 Landau Level Crossing at $B = 8$ T in Twisted Trilayer Graphene: Large Carrier Density and Small Electric Field	67
4.9 Landau Level Crossing at $B = 8$ T in Twisted Trilayer Graphene: Near Dirac Point	68
4.10 Landau Level Crossing at $B = 8$ T in Twisted Trilayer Graphene: Zero Landau Level of Monolayer Graphene Crossing	69

Chapter 1

Introduction

Graphene, a one-atom-thick honeycomb lattice of carbon atoms, is the first isolated structural 2-D material. The electronic transport properties and thermal transport properties of graphene are both the best among the any other materials so far. On the other hand, graphene has excellent mechanical properties to permit very high mechanical stress, while in some sense graphene is also the softest material, because it is atomically thin and therefore highly flexible. At the present time graphene shows various application prospects in industry, which motivates people to perform further research on this material.

The first single-layer graphene was achieved by Andre Geim and Konstantin Novoselov[1] by mechanical exfoliation. Then further transport measurements were done for both monolayer graphene [2][3] and bilayer graphene[4]. Since the charge impurities in the SiO₂ substrate lead to a very low mobility of graphene, people then tried to make suspended graphene samples to improve mobility[5][6][7], where the mobility can reach as high as 200,000 cm² V⁻¹ s⁻¹. Suspended graphene is also very useful as a mechanical resonator[8][9][10]. However, the suspended graphene devices are still too

fragile. Although the resist residue from the fabrication process can be removed by current annealing, the current annealing procedure is risky. The above shortcoming pushed people to find a better substrate to support graphene and while maintaining the high mobility. Hexagonal boron nitride is the best substrate so far that satisfies this requirement.

In this thesis I report transport measurements of the heterostructures of few-layer graphene on boron nitride. In chapter 1, I talk about some basic concepts of graphene electronic properties, including the energy band structure, the quantum Hall effect and Landau levels, the graphene/hBN heterostructures.

In chapter 2, I introduce the fabrication process to make the graphene/hBN heterostructures, including several methods to transfer graphene onto boron nitride, and the edge contact fabrication to make the electrodes. The Elvacite dry transfer method allows us to stack the graphene or boron nitride layer by layer, to get hBN/graphene/hBN sandwich structures to get very high mobility devices, as well as hBN/Bilayer graphene/hBN structures with dual gates. In this way we can also make twisted multilayer graphene with various twist angles. Furthermore I show another wet transfer method by buffered oxide etch (BOE) etching, which we can transfer the flakes from the SiO_2/Si wafer to the target substrate. This wet transfer method is useful when we need to pattern the flakes before transferring, for example, to make the hBN/graphene/hBN/graphene/hBN heterostructures to measure Coulomb drag.

In chapter 3, I report an electric field tunable Hofstadter's butterfly spectrum in the bilayer graphene/hBN superlattice sample. Since hexagonal boron nitride has the same honeycomb lattice as graphene, except the lattice constant has a 1.8% mismatch, when the bilayer graphene is aligned with the boron nitride, the lattice mismatch leads to a long-wavelength pattern named moiré superlattice. For new superlattice the wave-

length (the lattice constant of the superlattice) can be as large as 14 nm, and for one superlattice unit cell one can get one flux quantum, so that the Hofstadter's butterfly can be realized with laboratory attainable magnetic fields [39][37][38]. We also discuss another interesting experiment based on the bilayer graphene superlattice with dual gates, in which the interlayer electric field is another tunable degree of freedom. The larger unit cell in the superlattice means a much smaller reduced Brillouin zone, and the initial low energy band structure will be folded in to the reduced Brillouin zone, and form secondary and even tertiary gaps or minimum density of states points. The resistance at those gaps or minimum density of states can be tuned by the electric field in a way different from the primary charge neutrality point (CNP) of bilayer graphene, where the gap of primary CNP is proportional to the applied perpendicular electric field and the gaps of the satellites point can be closed by the electric field. When the device is placed in the perpendicular magnetic field, each satellite point has its own series of Landau levels. Those series of LLs interfere each other when they cross, and the induced LL splitting happens at even zero electric field. The zero- E -field valley degeneracy lifting is because the moiré pattern is only formed at the bottom layer of bilayer graphene and the layer symmetry is broken, and the valley symmetry is affected in a similar way. In our measurement, the electron-hole asymmetry appears when we fix the magnetic field and vary both the Fermi level and electric field, and we believe that asymmetry comes from the interaction of the carbon atoms and the aligned boron and nitrogen atoms.

Chapter 4 shows data on the twisted trilayer graphene multilayers, in which we put one layer of monolayer graphene on a bilayer graphene layer with a large twist angle. The two groups of Landau levels from each layer are weakly coupled, and we can see the LLs crossing whether we sweep both the gate and magnetic field or we sweep the two gates at a fixed magnetic field. Because the monolayer graphene has parabolic level

spectra while the bilayer graphene has nearly linear LL energy spectrum, we observe the two sets of LLs with different level spectra cross. Because the discrete energy levels of LLs, and the different energy gap between the LLs of bilayer graphene and LLs of monolayer graphene, so when the Fermi level tuned to reach one of the LLs of monolayer, the sudden charging behavior will change the electric field in between the mono- and bilayer graphene, and shift the energy of LLs. We will compare the LL crossing behavior of the twisted trilayer graphene and natural ABA trilayer, and highlight the similarities and differences.

1.1 Electronic Properties of Monolayer Graphene

Graphene has a honeycomb lattice (fig. 1.1) with two inequivalent sub-lattices labeled A and B. One atom from sub lattice is surrounded by three atoms from the sub-lattice B, and vice versa. The graphene plane is formed by the sp^2 hybridization of the carbon atoms, where in each atom three electrons form σ bonds with the electrons from the nearest three atoms of the other sub-lattice. The angle between each σ bond is 120° . The fourth electron in the atom will form the π bonds, and the π orbitals are perpendicular to the plane. One unit cell of graphene lattice includes one atom from sub-lattice A and one atom from sub-lattice B. Because the on-site energy is the same for sub lattice A and B, so the electrons in the two sub-lattices are energy degenerate. This degeneracy is the origin of the pseudospin degeneracy of graphene. Use a tight-binding model, we calculate the band structure of graphene.

The energy of the bands coming from the σ orbitals is far away from the Fermi energy, and will not contribute to the transport properties. Here we only consider the electrons from the π orbitals, and the overlap of the π electron wave functions from the

nearest carbon atoms is the hopping energy. Assuming the on-site energy is 0, and the hopping energy is t , which is about 2.8 eV. The lattice as shown in figure we have lattice vector (fig. 1.1)

$$\begin{aligned}\vec{a}_1 &= a\left(\frac{\sqrt{3}}{2}\vec{i} + \frac{1}{2}\vec{j}\right), \\ \vec{a}_2 &= a\left(\frac{\sqrt{3}}{2}\vec{i} - \frac{1}{2}\vec{j}\right)\end{aligned}$$

where $\vec{i}$ and $\vec{j}$ are the two orthogonal vector basis of the lattice plane, and a is the lattice constant about 0.246 nm. The reciprocal lattice of graphene is determined by $\vec{a}_i \cdot \vec{b}_j = \delta_{ij}$, then we get $\vec{b}_1 = \frac{4\pi}{\sqrt{3}a}\left(\frac{1}{2}\vec{i} + \frac{\sqrt{3}}{2}\vec{j}\right)$, $\vec{b}_2 = \frac{4\pi}{\sqrt{3}a}\left(\frac{1}{2}\vec{i} - \frac{\sqrt{3}}{2}\vec{j}\right)$ (fig. 1.2). So the first Brillouin zone is hexagonal, with six corners.

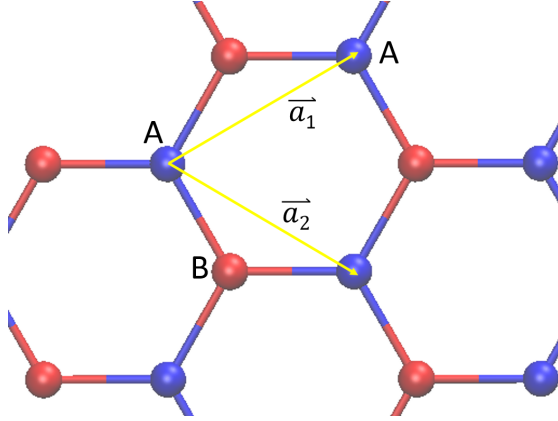


Figure 1.1:

Assuming the hopping energy between the nearest carbon atoms is t , and ignore the next-nearest hopping and so on, we can have the tight-binding Hamiltonian be written as

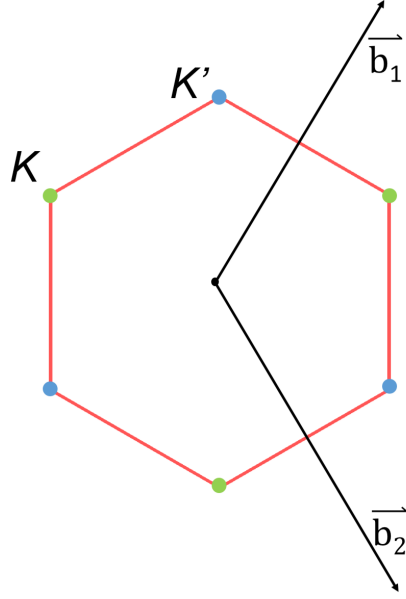


Figure 1.2:

$$H = t \sum_{\langle ij \rangle} (\hat{a}_i^\dagger \hat{b}_j + H.C.)$$

where the sum $\langle ij \rangle$ means we only consider the nearest-neighbor hopping, and ignore the rest. The eigenstate of this wave function in the periodic lattice is the Bloch wave function, which has the form of

$$\Psi_i = \phi_i(r) e^{-i\vec{k} \cdot (\vec{R}_i + \vec{r})}$$

In the tight-binding model, the off-diagonal components of the Hamiltonian matrix are

$$\begin{aligned} H_{off-diagonal} &= t \sum_{\langle j \rangle} e^{-i\vec{k} \cdot \vec{\delta}_j} \\ &= t(e^{-i\vec{k} \cdot \vec{\delta}_1} + e^{-i\vec{k} \cdot \vec{\delta}_2} + e^{-i\vec{k} \cdot \vec{\delta}_3}). \end{aligned}$$

Assuming the on-site energy is zero, then we get the 2×2 Hamiltonian as

$$H = \begin{pmatrix} 0 & t(e^{i\vec{k}\cdot\vec{\delta}_1} + e^{i\vec{k}\cdot\vec{\delta}_2} + e^{i\vec{k}\cdot\vec{\delta}_3}) \\ t(e^{-i\vec{k}\cdot\vec{\delta}_1} + e^{-i\vec{k}\cdot\vec{\delta}_2} + e^{-i\vec{k}\cdot\vec{\delta}_3}) & 0 \end{pmatrix}$$

by diagonalizing the matrix we can get the energy

$$E(\vec{k}) = \pm t \sqrt{1 + 4 \cos\left(\frac{\sqrt{3}k_x a}{2}\right) \cos\left(\frac{k_y a}{2}\right) + 4 \cos^2\left(\frac{k_y a}{2}\right)}$$

the energy band structure is shown in figure 1.3.

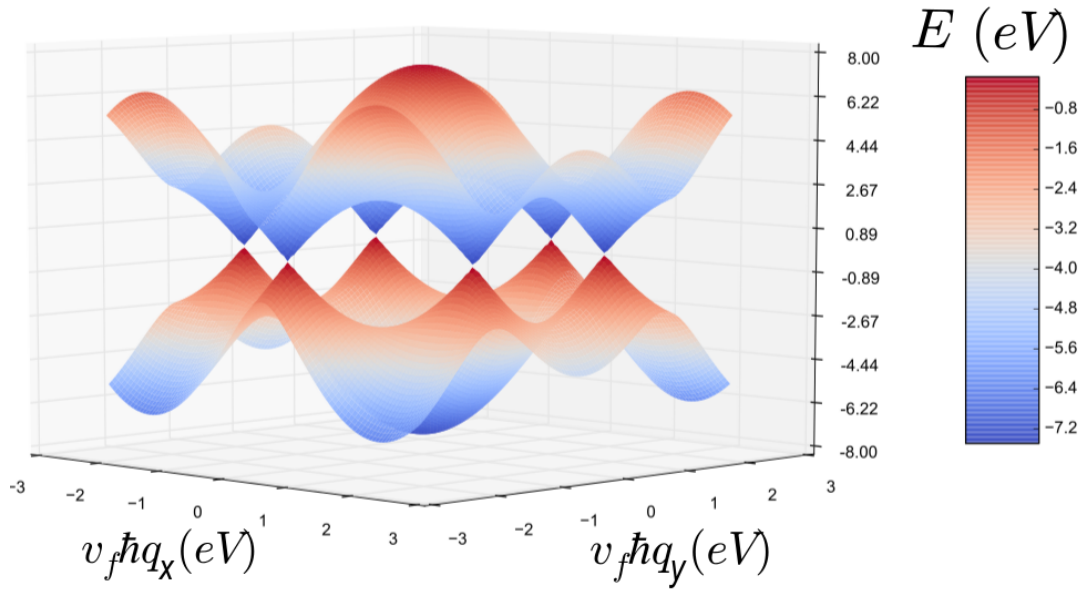


Figure 1.3:

From the figure we can see six isolated points where the valence band and the conduction band touch. Those points are the Dirac points, which are located at the

six corners of the first Brillouin zone. We can expand the energy close the those Dirac points, and rewrite the wave vector as $\vec{k} = \vec{K} + \vec{q}$, so

$$E = \pm t \frac{\sqrt{3}a}{2} |\vec{q}| \quad (1.1)$$

We draw the low energy band structure of monolayer in figure 1.4. Due to the formula $v_f = \frac{dE}{dp} = \frac{dE}{d(\hbar k)}$, we get the Fermi velocity $v_f = t \frac{\sqrt{3}a}{2\hbar}$, which is about $10^6 m/s$. Then the linear energy dispersion can be written as

$$E = \hbar v_f q$$

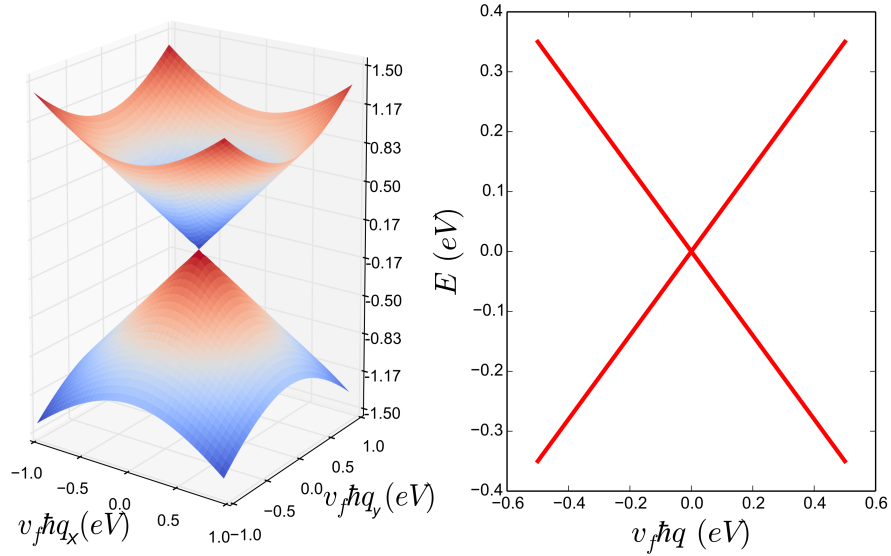


Figure 1.4: Low energy band structure of monolayer graphene. The Dirac cone of the energy dispersion, the conducting band and valence band have both linear energy dispersion relation and touch each other at the Dirac point.

Because each carbon atom supplies just one electron to the π band, and in the undoped case the filling level is just half of the energy band, so we have the Fermi energy at the Dirac point without any doping. In the transport measurement, we dope

the graphene by electronic gating, which can only tune the Fermi energy to as high as 0.3 eV due to the limitations of dielectric breakdown. The energy dispersion relation remains linear even for energies as large as 1 eV[11][12].

Now we consider the density of states in the graphene. For the wave vector $\vec{q}$, the energy in the conduction band is $\hbar v_f |\vec{q}|$. Because of the rotation symmetry, all wave vectors $\vec{q}$ with the same amplitude q are energy degenerate. The number of states in k -space between q to $q + \Delta q$ is

$$N(E) = \frac{\pi q^2}{(2\pi/L)^2} = \frac{L^2}{\pi} \left(\frac{E}{\hbar v_f} \right)^2 \quad (1.2)$$

so the number of the k -space points per unit real-space area is $n(E) = \frac{N(E)}{L^2}$,

so the density of states is

$$D(E) = g_s g_\nu \frac{d(n(E))}{dE} = \frac{2E}{\pi \hbar^2 v_f^2} \quad (1.3)$$

in the valence band, the energy is negative, so we have the density of states as

$$D(E) = g_s g_\nu \frac{d(n(E))}{dE} = \frac{2|E|}{\pi \hbar^2 v_f^2} \quad (1.4)$$

where the g_s is the degeneracy of spin and g_ν is the degeneracy of valley. In graphene both the spin and valley degeneracy are 2. The density of states shows linear dependence on the energy or wave vector. According to the vanishing DOS at the Dirac point, the conductance in the Dirac point should decrease to zero. However in experiment the minimum conductance is on the order of $4\frac{e^2}{h}$ at the Dirac point. One reason is the charge puddles in the graphene sample [13][14][15]. Besides the charge puddle, other effects also lead to the non-zero minimum of the conductance [18][19][20].

1.2 Electronic Properties of Bilayer Graphene

In Bernal stacked bilayer graphene lattice, the A atom in the top layer graphene correspond to a B atom in the bottom layer, as shown in the figure 1.5. Here we only consider the layer coupling γ_1 , which is about 0.4eV, and is much smaller than the hopping amplitude between the near-neighbor atoms in the same layer. We ignore the other interlayer hopping parameters γ_2 , γ_3 . Using the tight-binding model we get the low energy dispersion

$$E^2 = \frac{\Delta^2}{4} + v_f^2 \hbar^2 q^2 - \frac{\gamma_1}{2} \pm \sqrt{\frac{\gamma_1^2}{4} + (v_f \hbar q)^2 (\gamma_1^2 + \Delta^4)} \quad (1.5)$$

where Δ is the energy difference between the two layers caused by the vertical electric field[16][17].

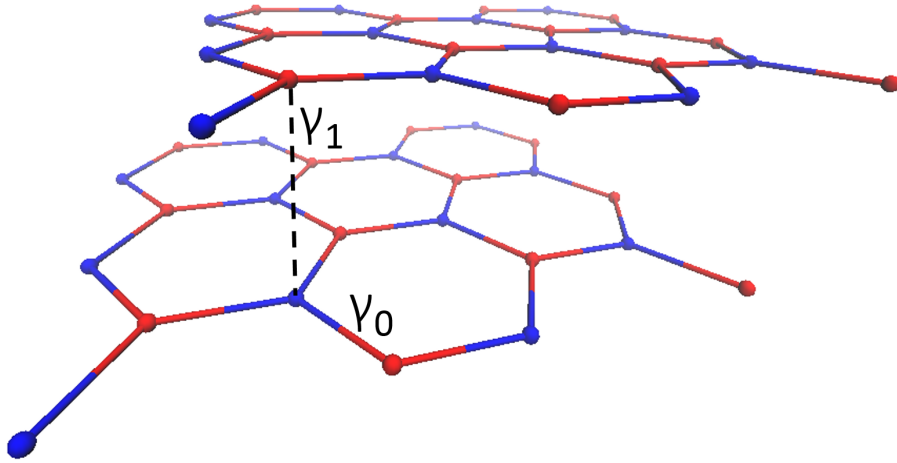


Figure 1.5: Bernal graphene lattice structure, an atom in the top layer corresponds to a B atom in the bottom layer, with the interlayer hopping energy γ_1 . γ_0 is the hopping energy between the A atom and B atom in the same layer.

In figure 1.6, we draw the low energy band structure of bilayer graphene, with both zero layer bias ($\Delta = 0$) and non-zero layer bias ($\Delta \neq 0$). When $\Delta = 0$, the conduction band and valence band are both parabolic, and touch each other at zero

energy, so there is no gap. When the vertical electric field is applied, the two layers have different energy, leading a gap between the conduction band and valence band. In the transport measurement, the layer bias can be added by the dual-gate geometry[43][47].

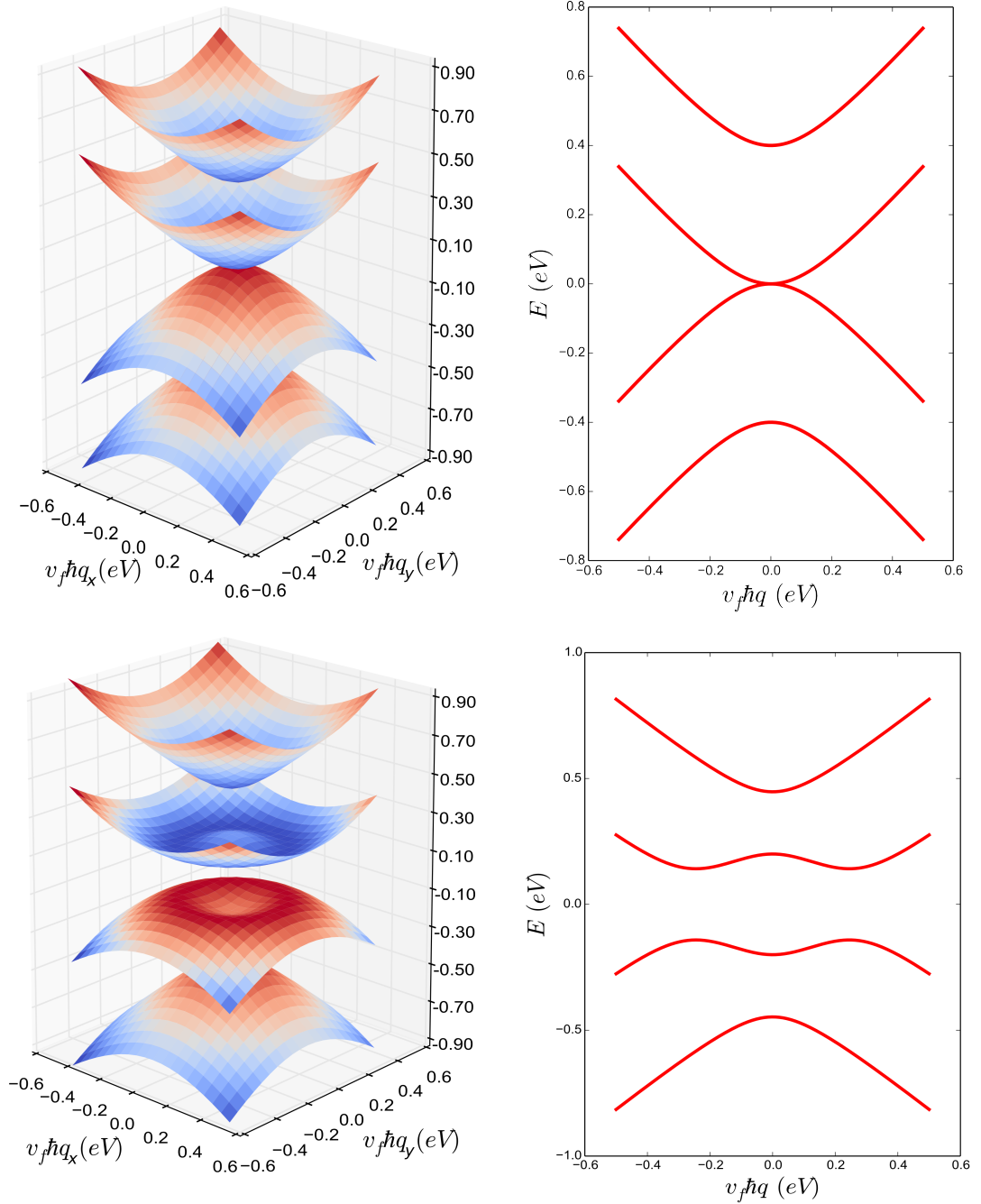


Figure 1.6: Low energy band structure of bilayer graphene, (top) the interlayer bias is zero, and there is no internal vertical electric field. The conduction band and valence band are both parabolic and touch each, so no gap show up. (bottom) Assuming the interlayer bias $\Delta \neq 0$, and a electric induced gap appears.

1.3 Quantum Hall Effect in Graphene

When a graphene sample is put in strong magnetic field, discrete Landau levels form, and the quantum Hall effect (QHE) is observed in the transport measurement. In monolayer graphene, the LLs have energy

$$E_N = \pm \sqrt{2\hbar v_f^2 |N| eB}, \quad |N| = 0, 1, 2, \dots \quad (1.6)$$

In the figure 1.7, we have the Landau levels vs the energy. The red solid lines (peaks) are the Landau levels, and the intervals between the neighboring LLs equals $\sqrt{N+1} - \sqrt{N}$, which becomes smaller as the $|N|$ increases. The number of states in each level is the same. The blue dashed line is the density of states in the graphene without any magnetic field, which is linear.

The Hall conductance in the monolayer graphene is

$$\sigma_{xy} = g_\nu g_s \left(N + \frac{1}{2}\right) \frac{e^2}{h}, \quad |N| = 0, 1, 2, \dots \quad (1.7)$$

where the g_ν and g_s are the valley and spin degeneracy and both equals to 2.

While in the bilayer graphene, the LLs has energy

$$E_N = \pm \hbar \omega_C \sqrt{|N|(|N| - 1)}, \quad |N| = 0, 1, 2, \dots \quad (1.8)$$

as shown in the figure 1.8. Here $\omega_C = \frac{eB}{m^*}$, and m^* is about $0.03m_e$.

The interval between each level is constant, and the Hall conductance is

$$\sigma_{xy} = g_\nu g_s N \frac{e^2}{h}, \quad |N| = 0, 1, 2, \dots \quad (1.9)$$

If both the spin and valley symmetry is conserved, the degeneracy of each LL is 4. As the magnetic field increases, both monolayer graphene and bilayer graphene will

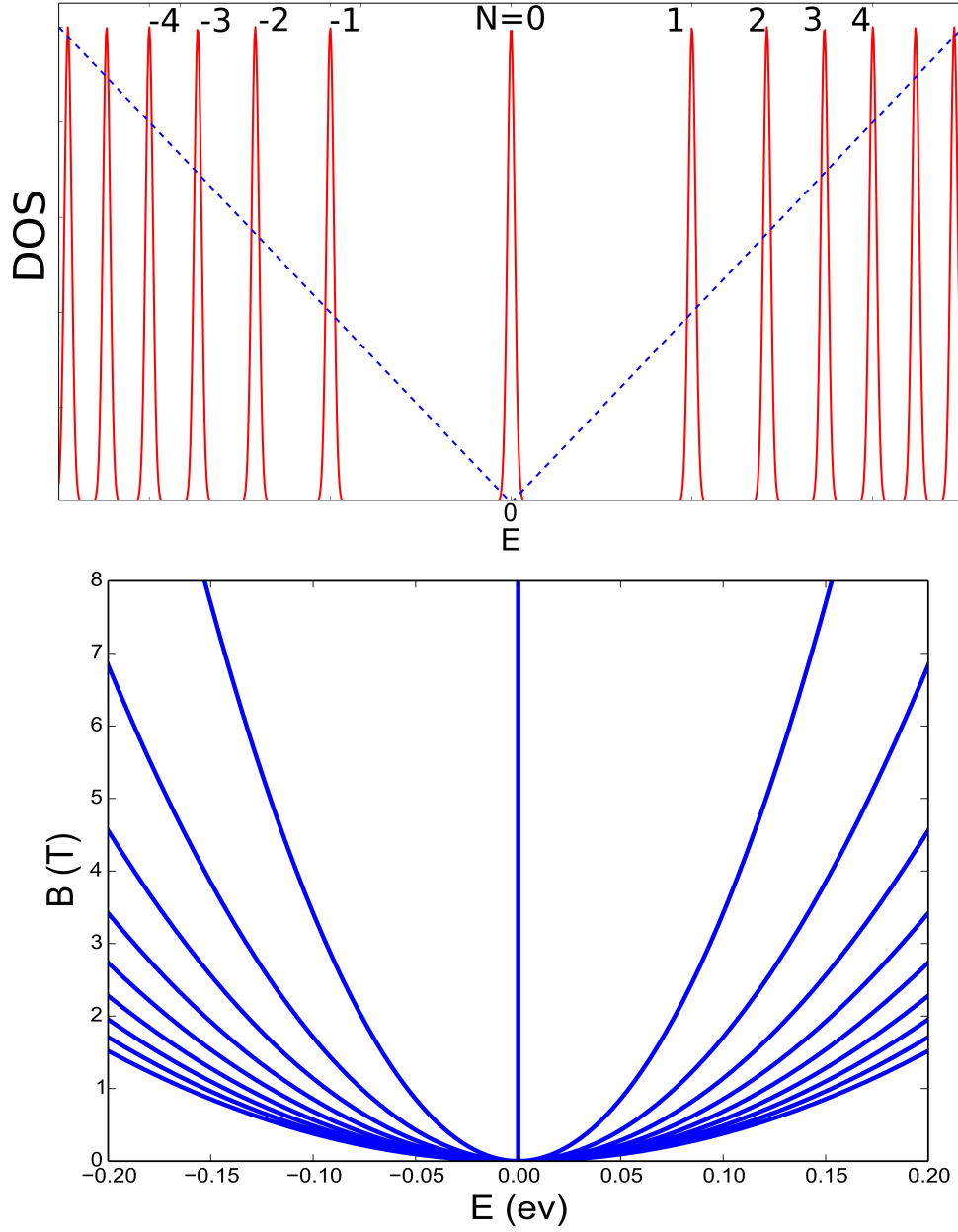


Figure 1.7: The Landau levels in the monolayer graphene. Red solid line: Landau levels vs. energy. y axis is density of states. Blue dashed line: density of states in monolayer graphene without magnetic field.

exhibit Landau level splitting.. In monolayer graphene, the Zeeman energy also increase and Hall conductance σ_{xy} shows new plateaus, while the strong Coulomb interaction will lift the 4-fold degeneracy[21][22][23][24][25]. In the bilayer graphene, when the magnetic field is large enough, the degeneracy of 4 will be lifted[26]. On the other hand, a vertical

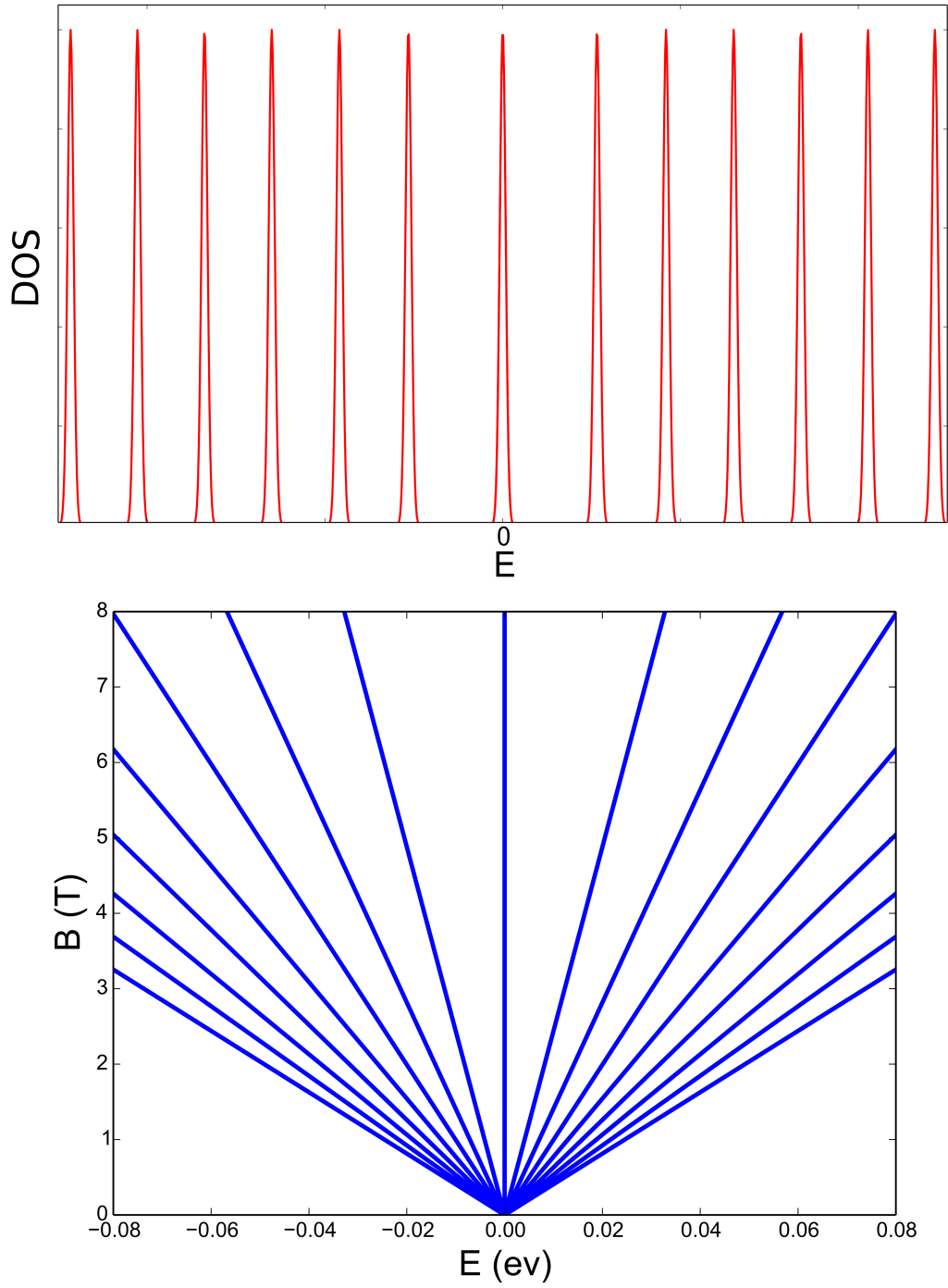


Figure 1.8: The Landau levels in the bilayer graphene. Red solid line: Landau Levels vs. energy. y axis is density of states.

electric field in bilayer graphene breaks the inversion symmetry and then the valley degeneracy, so the LL splitting can happen at lower magnetic field if non-zero E field is applied. Furthermore when the magnetic field is as high as 30 T, then electron-electron interaction induced fractional quantum Hall effect (FQHE) starts to show up in both monolayer graphene [27][28][29][30] and bilayer graphene[43][31].

1.4 High Quality of Graphene on Boron Nitride

Hexagonal boron nitride lattice is planar honeycomb lattice, the same as graphene lattice except with about a 1.8% lattice constant mismatch. (fig. 1.9).

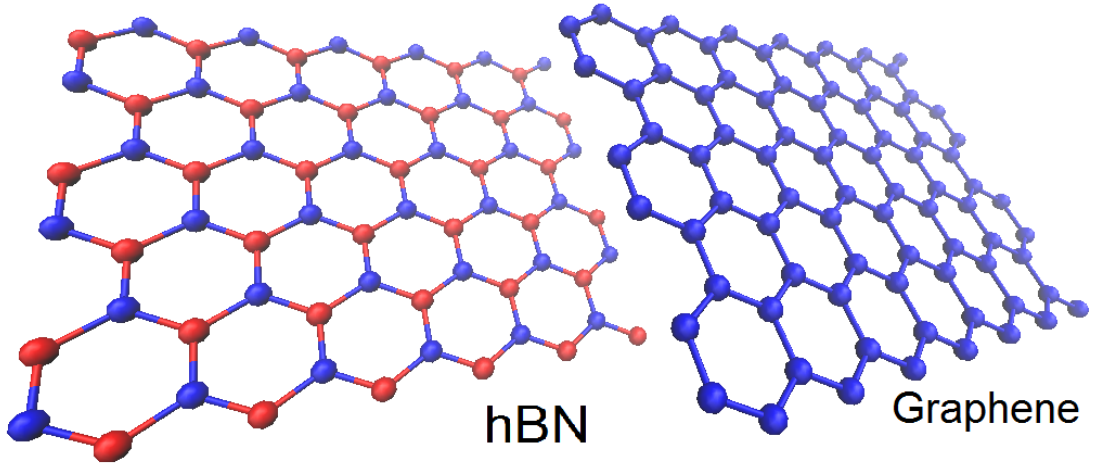


Figure 1.9: The honeycomb lattice structure of hexagonal boron nitride and graphene. They have the same lattice except with a 1.8% lattice constant mismatch.

From the year of 2010 when the first paper of transferred graphene on hexagonal boron nitride came out[34], hBN has been attracting the attention of people, and inaugurated a new era of graphene research.

First of all, graphene on hBN can show a very high mobility. In table 1.1, because of the planar lattice structure the hBN is atomically flat, the transferred graphene

has much smaller surface roughness than on other substrates. Such decreased surface roughness will decrease the synthetic fields under strain in graphene. Also the planar lattice means unlike SiO_2 there is no broken chemical bonds out of the plane, and the charge impurities that can scatter the electrons become much less. Also we notice that the large on-site energy difference of boron atom and nitrogen atom create a large band gap, so hBN is an insulating layer which does not contribute to the transport measurement. Besides acting as a good substrate, we also use BN as a insulating layer for top gate. In table 1.1 we can see the dielectric breakdown field is much larger than SiO_2 , allowing us to dope the graphene to a much higher carrier density.

Table 1.1: hBN and SiO_2

Material	Structure	Dielectric Constant	Band Gap	Breakdown field
hBN	Layered	4	5.5 eV	1.2 V nm^{-1}
SiO_2	Amorphous	3.9	8.9 eV	0.56 V nm^{-1}

Further, when the graphene is aligned with boron nitride, a superlattice (with lattice constant as long as 14 nm) develops. The large unit cell of the superlattice compared to the lattice scale leads to a much smaller reduced Brillouin zone, in which we can see new generations of Dirac points. When the carrier density reaches the secondary or tertiary Dirac points, new groups of Landau levels appear. Hofstadter's butterfly pattern can also be seen at aligned bilayer graphene/hBN heterostructures.

Chapter 2

Layer Stacking Transfer and Edge Contact Fabrication

2.1 Layer Stacking Transfer by Elvacite

Elvacite acrylic resins are polymers or copolymers of methyl methacrylate or other acrylic monomers for use in solvent-applied lacquers, inks, adhesives and specialty coatings. Here we use Elvacite as the support substrate to perform the dry transfer.

First we dissolve Elvacite in the pure methyl isobutyl ketone (MIBK) in a glass bottle, in the ratio that 8 grams Elvacite per 50 ml MIBK. Then we put the bottle on hotplate at 80 °C for at least 5 hours. When all of the powder dissolved we get the Elvacite solution. Spin coating the solution on a transparent Scotch tape/glass slide, in a speed of 2000 rpm for 20 seconds, and a ramp rate of 500 rpm/s. After a few minutes we repeat the spin coating process before putting the glass slide on the 120 °C hotplate for 5 minutes.

After spin coating, we mechanically exfoliate graphene onto the Elvacite/tape/glass slide, then use optical microscope to find good monolayer or bilayer graphene flake. Then

the Elvacite/tape is cut into small pieces which makes the flakes easier to transfer. In the meantime, the boron nitride is exfoliated on the SiO_2/Si wafer, then the wafer is annealed in the furnace by 500°C O_2 gas. We use the optical microscope to find flat and large boron nitride flake as well. Then the graphene on the Elvacite/tape/glass is aligned with boron nitride on the wafer fixed on the transfer stage by the optical microscope. The stage is lowered to let the Elvacite contact the wafer, and a heater is turned on to melt the Elvacite (the temperature is about 80°C in our setup). After a few minutes, if the Elvacite becomes soft and the graphene completely touches the boron nitride, we stop the heater and remove the glass slide and tape, then the Elvacite layer with the graphene is on the wafer (Figure 2.2). In the following step, we put the wafer into the acetone until the Elvacite is dissolved, then use the furnace again to anneal the graphene/hBN in 300°C O_2 for 4 hours. Finally we get very clean graphene on boron nitride (Figure 2.1).

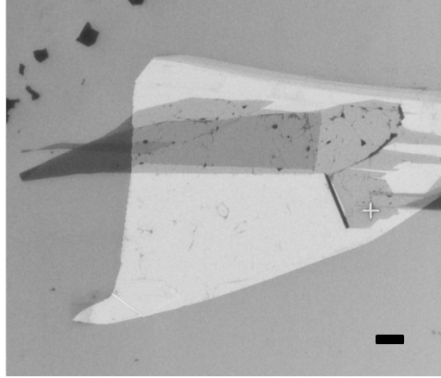


Figure 2.1: SEM image of Graphene on boron nitride. We can see monolayer and bilayer graphene in the sample. The scale bar is $5\mu\text{m}$.

Using the dry transfer method we can transfer any flake of graphene or boron nitride layer by layer. For example, after we get the graphene/hBN heterostructure, we can transfer another layer of hBN on top to make a BN/Graphene/BN sandwich, then

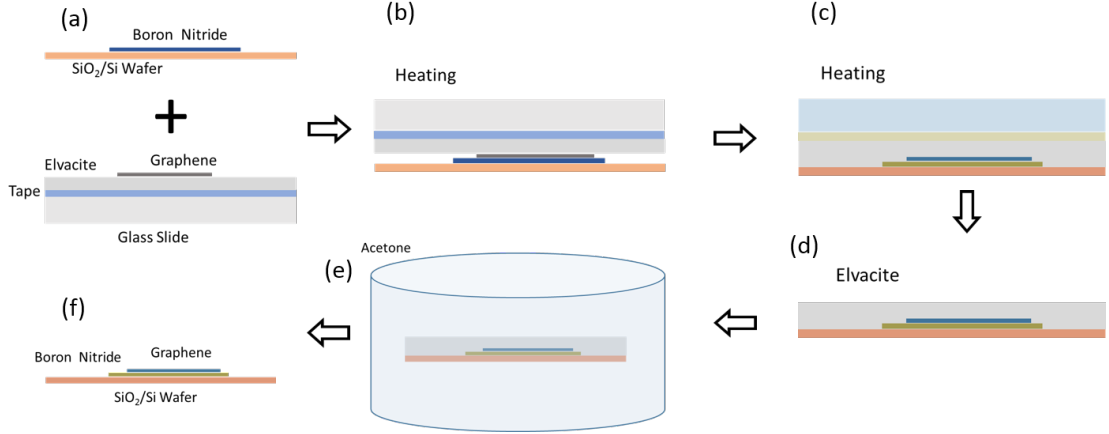


Figure 2.2: Process of layer stacking transfer. (a) a hBN flake is mechanically exfoliated on the wafer, and high temperature O₂ gas is used to get rid of the tape residue. On the other hand, a graphene flake is exfoliated on the Elvacite resist on transparent Scotch tape/glass slide. (b) The hBN flake touches the target graphene after we use optical microscope to align them and lower the transfer stage to get them touch each other. (c) The heater is turned on to keep a temperature at about 80 °C O₂. After a few minutes, the Elvacite layer melts. (d) Peeling off the tape/glass slide. (e) Putting the Elvacite/Graphene/hBN into acetone, to remove the Elvacite. (f) Finally we transfer graphene onto a boron nitride flake.

in the fabrication process the graphene is protected by the hBN from the resist, and can keep the mobility as high as $10^5 \text{cm}^2 \text{V}^{-1} \text{s}^{-1}$. Also we can transfer another layer of graphene onto graphene/hBN sample to make a twisted bilayer device, or transfer one monolayer graphene onto a Bernal bilayer graphene to make twisted trilayer graphene (Figure 2.3). The key point of the layer stacking technology is that both graphene and boron nitride is very stable during high temperature O₂ gas annealing.

2.2 Layer Stacking Transfer by Wet Etching

Sometimes we need to make special pattern of graphene or boron nitride before we transfer it onto the target substrate, which means that we need use electron beam lithography (EBL) to open a poly methyl methacrylate (PMMA) window, then plasma etching to make the pattern. In this sense, we need to develop a new method that can

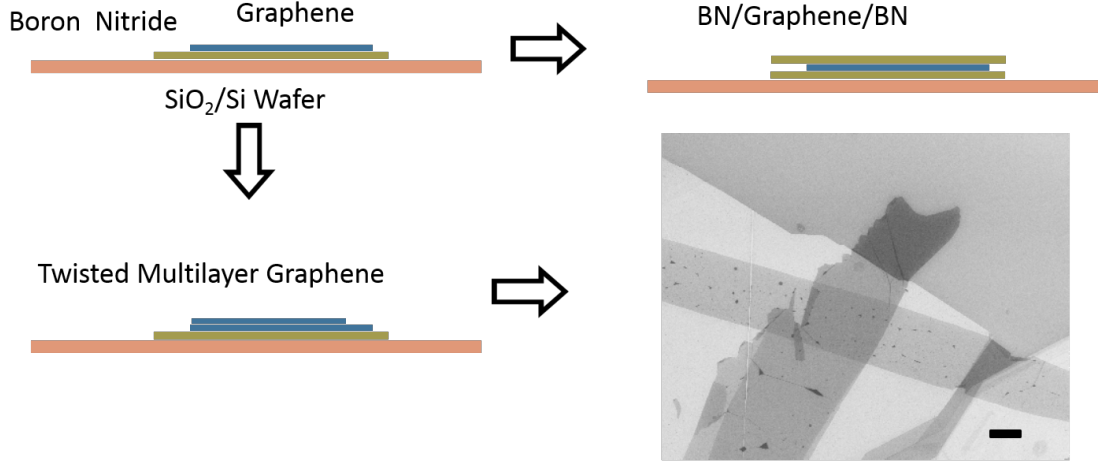


Figure 2.3: By stacking the graphene or boron nitride layer by layer we can make hBN/Graphene/hBN sandwich heterostructure, as well as twisted trilayer graphene. The scale bar is 5 μ m.

transfer the patterned graphene or boron nitride flake from the SiO₂/Si wafer. The BOE etching transfer method is the one meet our demand.

Firstly we prepare the graphene or boron nitride flake on SiO₂ chip, then use EBL and plasma etching to make the sample to the pattern we need. For instance, we make a series of holes on boron nitride flake (figure 2.5), then transfer it to a graphene/hBN sample. After we deposit the metal on the holes, we can make a point contact device. Another example is Coulomb drag device, where we need to etch the graphene/hBN bilayer into a long and narrow stripe before we transfer it onto another graphene stripe on boron nitride.

We also use Elvacite resist to do the BOE etching and transferring (figure 2.4). The BOE removes the SiO₂ while without damaging either hBN or graphene. After several times DI water rinsing, we can get rid of almost all the BOE residue, with a hBN or graphene/hBN bilayer flake on Elvacite layer exposed in air which is ready to transfer (figure 2.6). Because this transfer method is a wet transfer method, there might

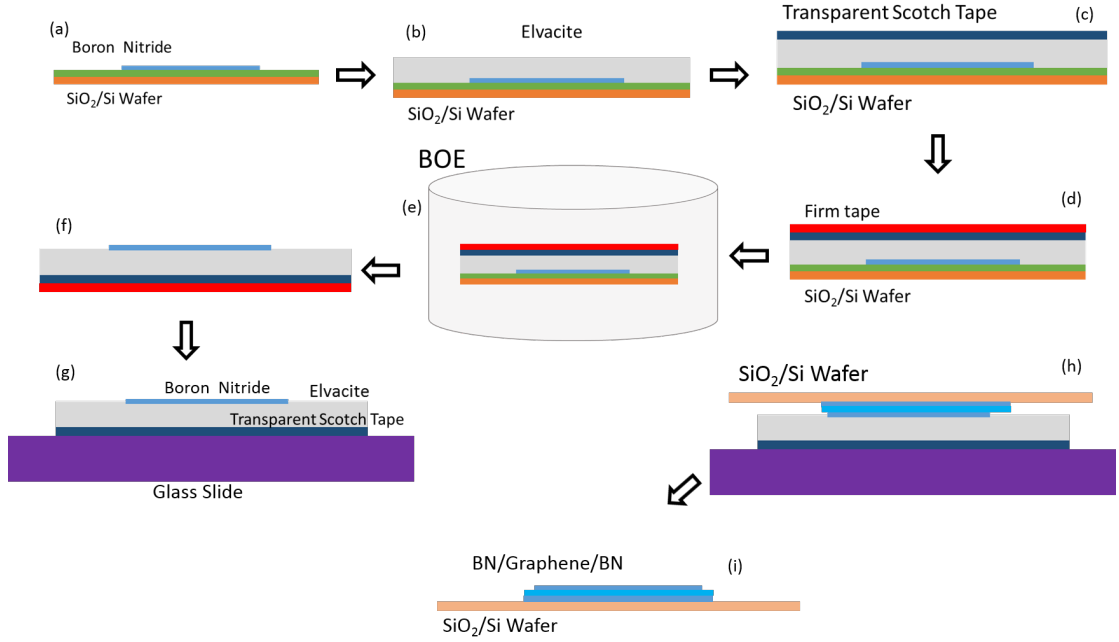


Figure 2.4: The process of BOE etch transfer. (a) preparing a thin layer of boron nitride on wafer. The boron nitride can be made into some special pattern by EBL and plasma Etching (SF_6 gas in ICP), for some special project. For example, we can make holes on hBN to make point contact, or etch the graphene/hBN bilayer into a stripe, to make a coulomb drag device. (b) Spin coating a layer of Elvacite on. The Elvacite resist has a very low glass transition temperature, which can make it a good substrate to do the transfer. (c) Putting a transparent Scotch tape on top. The transparent Scotch tape is used to hold the Elvacite layer after the BOE etch, since the Elvacite layer does not have a good mechanical properties as PMMA layer. (d) Putting another layer of firm tape on. This firm tape is used to avoid the ripple or folding when we are doing the BOE etch. (e) Putting the whole thing into the BOE solution. It might need more than one day to complete the etching process, in order to get rid of all of the SiO_2 . (f) Then the hBN will expose on top of the Elvacite membrane. Using the DI water to clean the sample. (g) Removing the firm tape which is not transparent. Then we put the hBN/Elvacite/transparent scotch tape on on glass slide, which is a supporting substrate for transfer. (h) Finally we transfer the etched hBN onto a Graphene/hBN heterostructure to make a special sandwich structure.

be more bubbles or ripples than the dry transfer. However, if we can avoid those bubbles and shape our sample only at the clean and flat area, we can fabricate a high mobility device. This transfer method is used to make a Coulomb drag device.

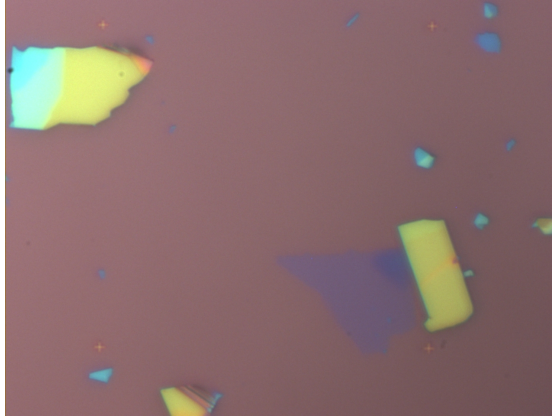


Figure 2.5: Preparation of thin layer boron nitride on chip. We use EBL to pattern this boron nitride, for example, into the long and narrow stripe.

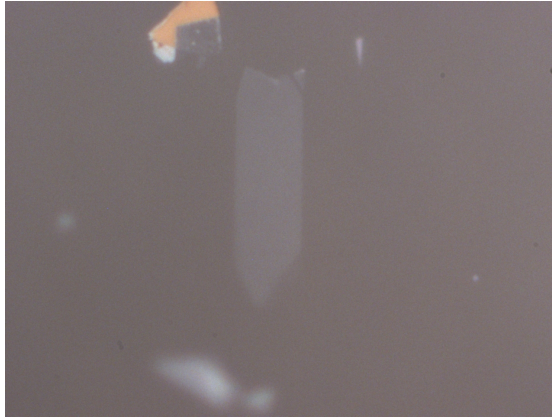


Figure 2.6: Etched boron nitride on the Elvacite Layer.

2.3 Edge Contact Fabrication

The edge contact fabrication method was developed by the Columbia group[35] (figure 2.7), which makes only a one-dimensional carbon chain edge contact between metallic electrodes and the graphene.

After several steps of dry transfer, we prepare a hBN/Graphene/hBN sandwich sample on chip. In figure 2.8 (a) we can see that the encapsulated graphene is protected by the boron nitride to avoid any PMMA residue, which can keep the graphene a very

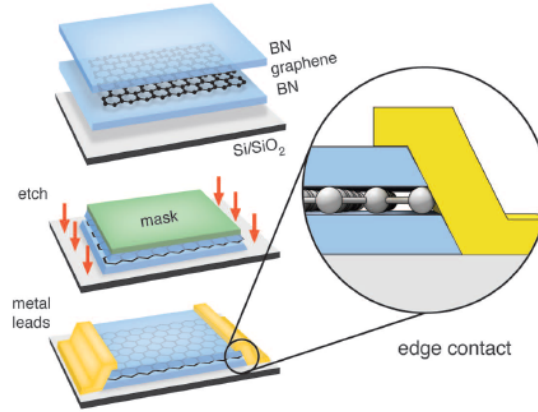


Figure 2.7: One-dimensional edge contact for graphene sample. This figure is from [35].

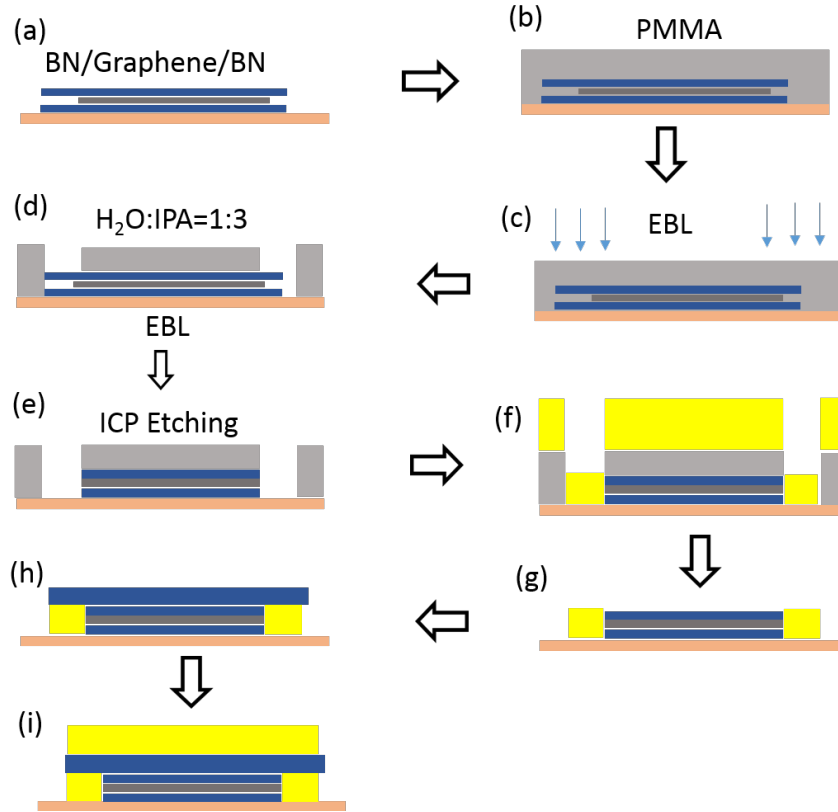


Figure 2.8: Edge Contact Fabrication Process. (a) Preparing the hBN/Graphene/hBN sandwich sample. (b) Spin coating two layers of PMMA. (c)(d) Use EBL system to open a window of PMMA layer. (e) Using SF_6 gas in the ICP system to etch the exposed part of the sample, then use O_2 to flush the sample to make a good electrode contact. (f) Deposit Cr/Au by the E-beam Evaporator. (g) Putting the sample in the acetone to liftoff. (h) Transferring another layer of hBN on top as a insulating layer for top gate. (i) We repeat the process (c)-(g) to make a top gate.

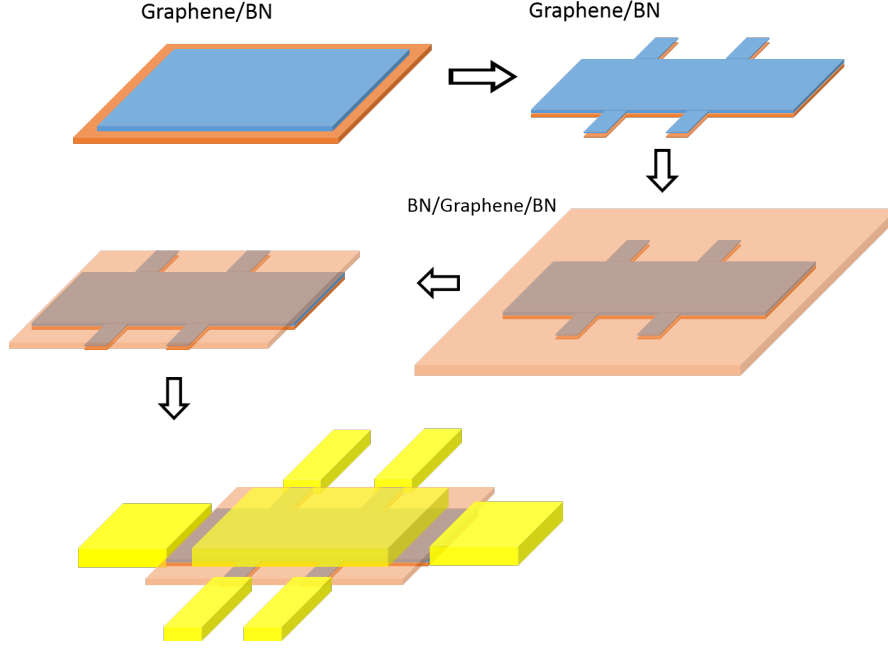


Figure 2.9: Edge contact fabrication. (a) Preparing graphene on hBN. (b) Etching the graphene into the Hall bar geometry. (c) We transfer another layer of hBN on top to cover the whole sample, then (d) etch the top hBN, to let the Hall bar edge exposed. (e) Then we make the edge contact and the top gate the top gate by fabrication process. The top gate only covers the central part, which is enough to do the four terminal measurement.

high quality. (b) Spin coating PMMA resist on the wafer using speed 4000 rpm and ramp rate 1000 rpm/s, then use hotplate to bake the sample for 10 minutes at 180 °C O₂. Typically the longer baking time, the better structure we can get, because of the less mechanical tension on PMMA. But when we doing the EBL in the graphene/hBN heterostructure sample, if the exposed point is exactly inside the hBN flake, it is common to get cracks after we develop by MIBK:IPA=3:1 developer. To avoid those cracks, I use cold Water:IPA=1:3 solution as the developer instead of MIBK/IPA. The temperature of the cold developer is lower than 0 °C O₂, and the developing time is about 10 minutes, depending on the size of the pattern, after that we use IPA to rinse the sample. The cold developing method is useful for very fine structures, and top gate. After cold developing, we make a PMMA window, and then use SF₆ plasma etching to etch the

hBN and graphene exposed in the window. Then we use O_2 plasma to flush the sample to oxidize the one-dimensional carbon chain at the edge, which is important to make a good electrical contact. 10 nm/50 nm Cr/Au is deposited on the chip, then we put the sample in acetone overnight on the hotplate at a temperature at 65 °C for lift-off, and then we get the edge contact device ready for the transport measurement. I also tried Cr/Pd/Au as the Columbia group did, but it did not yield working electrical contacts.

We can also transfer another layer of boron nitride as the top gate insulating layer, then repeat the fabrication process to make the top gate.

The thickness for the hBN insulating layer cannot be too thin, to avoid the tunneling current. Usually if the hBN is too thick, it is not flat after we transfer it on to the sample, because thick hBN is too hard to bend when it's put onto a step (the step could be the edge of the hBN/graphene/hBN sandwich, or the electrode). Besides the the boron nitride layer to insulate the top gate, we also have another layer of hBN on top of graphene coming from the hBN/graphene/hBN sandwich structure, which make the effective distance of the graphene/top-gate capacitor the sum of the two hBN layers. But the hBN to insulate the top gate and edge contact electrode is only the top layer, which means the carrier density cannot be tuned to a very high level before we get the tunneling current leakage. In order to get high carrier density in this kind of device, we need the top layer hBN of the sandwich structure is much thinner than the insulating hBN layer. However as we know, the thick layer of hBN not be flat after transferring, then the local capacitance between graphene and top gate are not uniform, leading to a PN junction structure. If this happens, the mobility will be lowered and we cannot get homogeneous space distributed gap in the bilayer graphene.

To avoid such situations, we can make a small change in the fabrication process (fig. 2.9). Firstly, we transfer a layer of graphene onto a hBN on wafer, then use the

standard process shown in (fig. 2.8) to make a hall bar geometry. After the ICP etching, we put the sample in the acetone to remove the PMMA. Then we put the sample in furnace and use 300°C O_2 to remove the PMMA residue. Then we transfer another layer of hBN on the sample as the insulating layer (fig. 2.10). Etching the sample to let the edge of the Hall bars and two terminals exposed in air, in order to make the edge contact. On the other hand, we can deposit the metal as the top gate, just covering the center part, which is enough for the four terminal measurement. This geometry is the same as the paper [36].

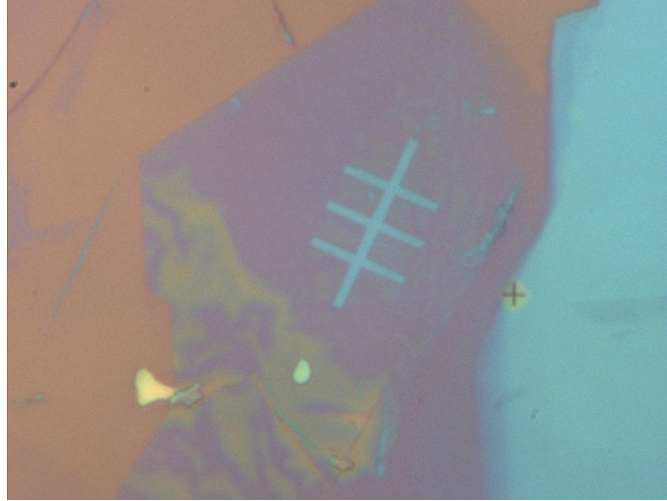


Figure 2.10: Hall bar geometry graphene/hBN sample get another thin layer of hBN on top.

Chapter 3

Bilayer Graphene/Boron Nitride Superlattices

3.1 Basic Properties of the Hofstadter's Butterfly in Graphene/hBN Superlattices

Hexagonal boron nitride/graphene (hBN/G) heterostructures[34] have recently emerged as a model system for experimentally investigating the properties of electrons in a combined periodic potential and magnetic field. The hBN has the same honeycomb lattice as graphene, but with a 1.8% lattice constant mismatch. When graphene is placed on hBN, the lattice and orientation mismatch creates a periodic moiré pattern[37][38][39], and a lattice-commensurate phase with periodic domains reported for form at small mismatch angles when the moiré period is $>10\text{nm}$ [40]. At special points at the edges of the superlattice Brillouin (sBZ) zone, the superlattice potential induces satellite points and is predicted to open small gaps for both electron and hole states if inversion symmetry is broken. Under a magnetic field B , commensuration effects between the lattice period

and the magnetic length lead to predictions of a fractal energy spectrum known as Hofstadter's butterfly [41]. Recently such a spectrum appropriate for graphene Dirac band structure [39] has been observed, as well as in bilayer graphene [37], and accompanied by the opening of a gap at the charge neutrality point, opening the door to the exploration of this system. Compared to previous efforts on semiconductor heterojunction based two-dimensional electron gas, the he graphene-hBN system is readily tunable, enabling the quantum Hall effect to be explored more fully.

Unlike monolayer graphene, Bernal stacked bilayer graphene consists of two layers, whose inversion symmetry can also be broken by a perpendicular electric field, and the E -field induced gaps will open at the charge neutrality points (CNP). In the case that the bilayer graphene and BN are aligned, the inversion symmetry is already broken at zero electric field due to a asymmetric coupling to the hBN, so a gap is predicted at zero E field in the bilayer graphene superlattice[42]. In our experiment of encapsulated bilayer graphene superlattice with dual gates which both the density and electric field, we did not see any gap at zero electric field, the possible reason for the absent of the gap at CNP is that the encapsulated geometry changes the dielectric environment of bilayer graphene.

On the other hand, when the magnetic field is turned on, the LLs have filling factor of $4Ne^2/h$ with N integer. The degeneracy of 4 comes from the two spin and two valley degeneracies. When the magnetic field strong enough then the spin degeneracy will be lifted because of the Zeeman splitting. Meanwhile if we have some nonzero electric field the valley degeneracy will be lifted, and complex transitions occure as a result [43]. However in the case of bilayer graphene/hBN superlattices, the Landau levels of the CNP and the satellite point (sP) cross, the interference pattern could change if we tune the electric field, which means that the butterfly pattern can be tuned by the

perpendicular electric field. At fixed magnetic field when we tune the electric field and carrier density at the same time the Landau level splittings happen and shows electron-hole asymmetric behavior which is not be observed before.

3.2 Raman Spectroscopy of Aligned Graphene with Boron Nitride

In this section I introduce a new method based on Raman Spectroscopy to check the alignment angle between the bilayer graphene and hBN. The Raman system in our measurement has laser wavelength 532 nm. Because of the slight mismatch of the lattice, when graphene is exactly aligned with hBN, the wavelength of the moiré pattern is about 14 nm. If the twist angle increases, the wavelength of superlattice decreases, following the formula below

$$\lambda = \frac{1.018a}{\sqrt{2.306(1 - \cos(\theta)) + 0.018^2}} \quad (3.1)$$

and

$$E_{sP} = \frac{2\pi\hbar v_F}{\sqrt{3}\lambda} \quad (3.2)$$

where a is the lattice constant of graphene. This means that the wavelength of the superlattice, the alignment angle and the E_{sP} are related. By Raman spectroscopy we can check the alignment of the graphene and hBN [44]. Here we develop a new method to check the alignment of bilayer graphene and hBN (figure 3.1). We transfer graphene with monolayer and bilayer graphene regions on a hBN, and get the Raman spectrum of the monolayer graphene, so we can get the twist angle (or superlattice wavelength) by the above formula. On the other hand we can get the Raman spectrum of the bilayer graphene on hBN, and combine the Raman spectra of monolayer graphene and bilayer

graphene we obtain the Raman spectroscopy of the bilayer graphene superlattice (bGS).

First we focus on the G peak. We can see another much weaker peak named R' here, which does not show up in the unaligned graphene/hBN. For the 2D peak of monolayer graphene superlattice (mGS) the full width half maximum (FWHM) is much wider than the unaligned sample. A example is shown in the figure 3.1, here we have a sample with wavelength 10.5 nm, which means the twist angle is 0.9° . Then after the Lorentzian fitting for the 2D peak of bGS, we get the widths of the four sub peaks are P_{22} : 21.3 cm^{-1} , P_{21} : 27.1 cm^{-1} , P_{12} : 24.6 cm^{-1} , P_{11} : 22.5 cm^{-1} .

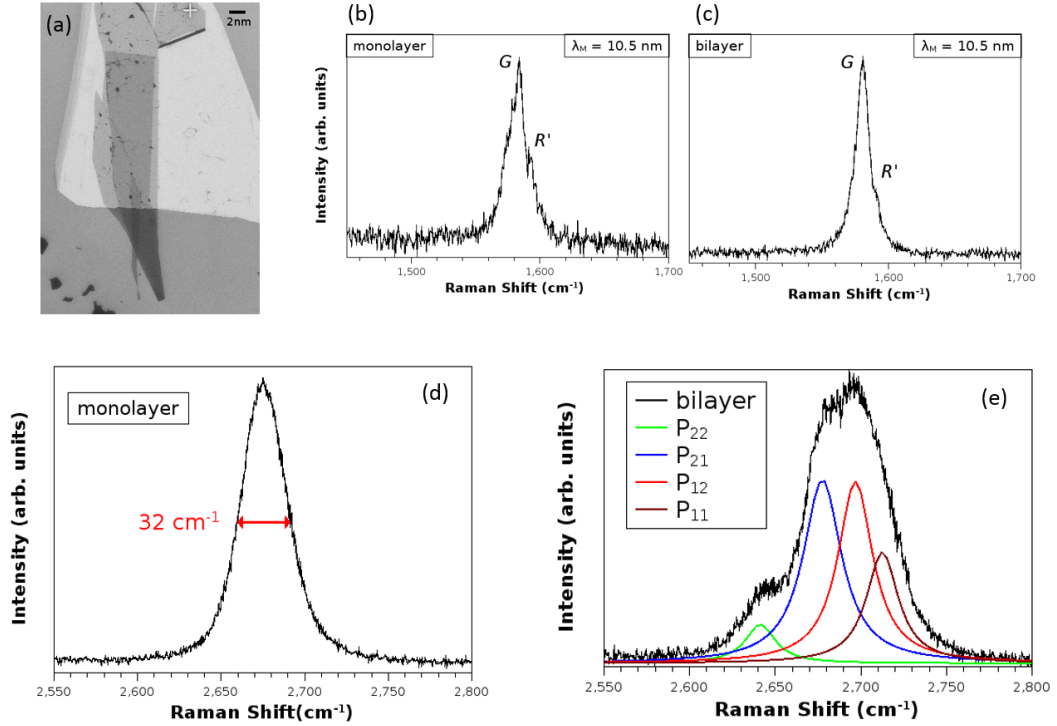


Figure 3.1: The Raman signal of monolayer graphene superlattice (mGS) and bilayer graphene superlattice (bGS). (a) Transferred graphene on hBN, and graphene is mGS and partial bGS. (b) (c) G peak of the mGS and bGS, we get R' features. (d) FWHM of 2D peak of mGS of the sample (a). (e) 2D peak of bGS. Lorentzian fit is used for the four sub peaks.

By measuring hundreds of graphene samples with partial monolayer and partial bilayer on hBN, we select more than ten with alignment angle smaller than 2° , where

we get the FWHM of the monolayer part. Since the relation between the FWHM of mGS and the twist angle is known[44] as below

$$FWHM(2D)[\text{cm}^{-1}] = 5 + 2.6\lambda[\text{nm}] \quad (3.3)$$

Then we can calculate the superlattice wavelength for each sample. Since the 2D peak Raman signal of the bilayer graphene part is also measured, we can get the statistics of Raman spectroscopy of bGS (figure 3.2). In the figures 3.2, we find that the P_{12} is almost flat when the twist angle changes, and the widths of the rest three peaks show linear dependence of the align angle. Then we do the linear fit to get the slopes (figure 3.3). As shown in the figure, P_{21} has the largest slope while peak P_{12} has the smallest slope. This means that we can use Lorentzian fitting width of P_{21} as a criterion to check whether the bilayer graphene is aligned with hBN.

The 2D peak of the graphene Raman comes from the double resonant process is related to two phonons of optical branch with opposite momentum [45]. The broadening of the mGS 2D peak only happens when the twist angle is small than 2.5° [44], because of the spatial periodic strain distribution. For bilayer graphene, the 2D peak has four components, because in both valley K and K' the energy band has two sub bands, and the double resonant process can have four possible pathways. Since the spatial periodic strain distribution broadens the width of the 2D peak of monolayer graphene when it is aligned with hBN, then it appears to act similarly to bilayer graphene aligned with hBN. So we can see similar relations between the widths of 2D peak components and the twist angle, except P_{12} in which the linear fit slope is much smaller than the other three components.

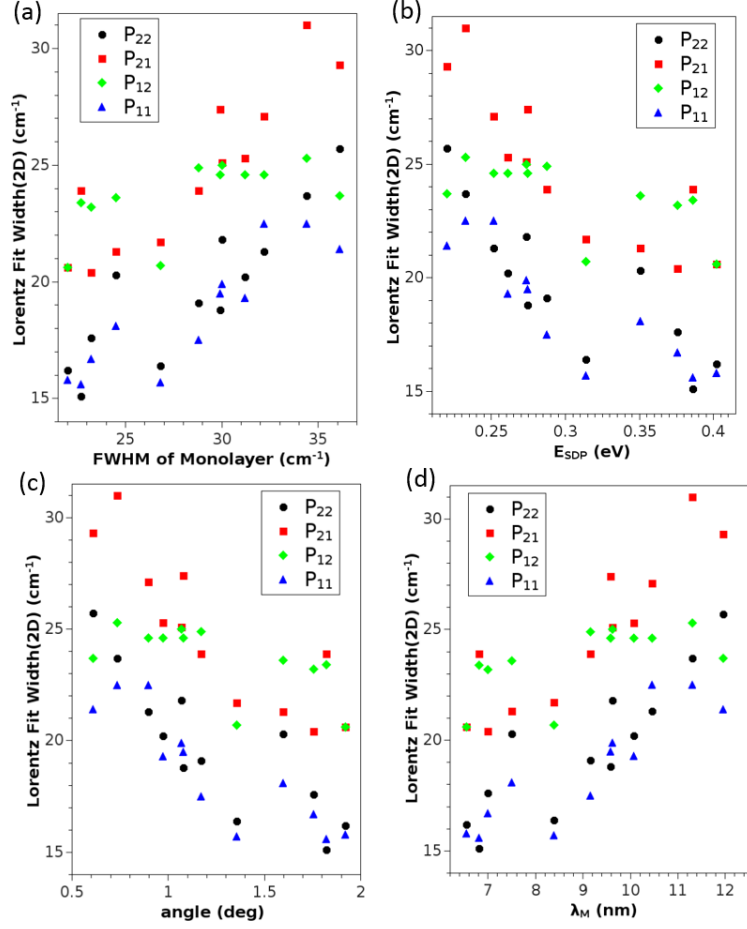


Figure 3.2: (a) Lorentzian fit width (LFW) of the four components of bGS 2D peak depends on the FWHM of mGS 2D peak. (b)(c)(d) The relations between LFW of bGS and the energy of satellite points, angle and superlattice wavelength.

3.3 Displacement Field Dependence of Second and Third Generations of Satellite Points in Bilayer Graphene Superlattice

When the bilayer graphene is aligned with a boron nitride layer, only one layer is in contact with the hBN and forms a moiré pattern, and Hofstadter's butterfly pattern shows up when the magnetic field is applied[37]. Because the moiré pattern only forms at one side of the bilayer graphene, the layer symmetry is broken even without electric field. The inversion symmetry breaking is supposed to open a gap at zero electric field in the

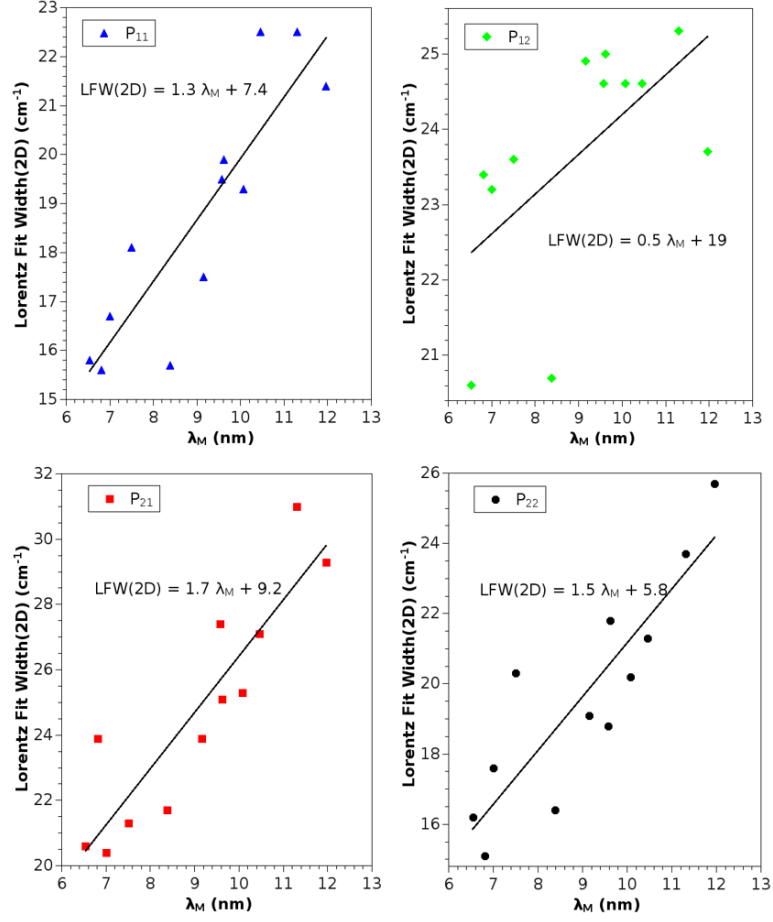


Figure 3.3: linear fit of LWF widths of the four components of 2D peak in bilayer graphene superlattice vs the wavelength of superlattice. The slope is around 1.5 except the P_{21} components.

bilayer graphene superlattice [42], just as the case of monolayer graphene superlattice [38]. However in our measurement we do not see any gap in the zero electric field (figure 3.4). The reason is probably that our bilayer graphene sample is encapsulated hBN, so even if the twist angle between the graphene and boron nitride is smaller than 1° the graphene is still in a incommensurate state, and the gap is absent[46].

By tuning the two gates we can control both the carrier density and displacement field (electric field) [43] [47] as the formula

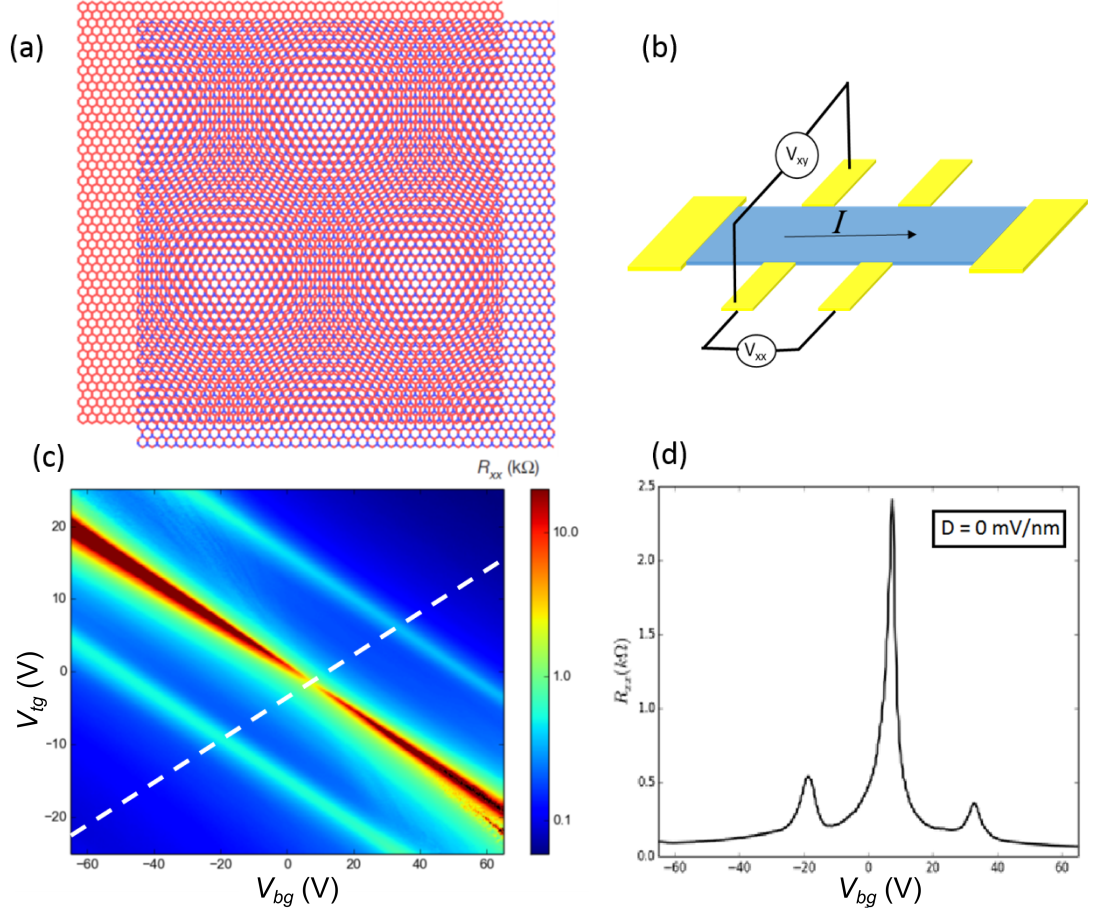


Figure 3.4: (a) Schematic of moiré superlattice from stacked hBN and graphene. (b) schematic diagram of Hall bar device. Current I goes through the whole device while the longitudinal voltage drop V_{xx} and transverse voltage drop V_{xy} are measured by lock-in system. (c) R_{xx} vs dual gates. Charge neutrality point opens a gap when interlayer electric field is turned on. (d) line trace of R_{xx} at zero displacement field.

$$n = (C_{topgate}V_{topgate} + C_{backgate}V_{backgate})/e,$$

$$D = (C_{backgate}V_{backgate} - C_{topgate}V_{topgate})/2e.$$

The positive displacement field means the direction is from back gate to top gate. Then we transform the x and y axis from the back gate and top gate to carrier density and displacement field (figure 3.5). The position of three Dirac Points (three perpendicular bright lines) only depends on carrier density, and are independent on the displacement field. At the CNP point, the total carrier density remains zero, and the

displacement field only opens a gap. The two satellite point are at the corners of the reduced Brillouin zone, and the carrier density required for the Fermi level to reach this level is also fixed, which we label as n_0 . The resistance of the sP is slightly dependent on the displacement field, and the electron and hole sides show opposite dependence. The possible reason for this D -field dependence is that the displacement field will push the wave function of electron or hole to prefer one layer of the bilayer graphene, and the layer symmetry is broken because only the bottom layer forms the moiré superlattice, so the resistance of two layers is different. If this is the case, we suppose to see a larger resistance at the lower layer of bilayer graphene since the aligned boron nitride lattice supply stronger scattering. In this sense in the electron side when the D field is upward (positive in our figures), then the electrons will prefer to occupy the lower layer, and the resistance should be larger than the case that the displacement field goes down. On the other hand in the hole side, the resistance is supposed to be larger when the displacement field points down. However our measurement has a opposite result (figure 3.5 right) to the discussion above, which means the layer polarization of charge is not the reason. Another possibility is charge puddles in the graphene sheet. Fig. 3.4 shows some charge puddle induced bright lines go through the CNP, and cross the sP. However, the line trace of the sP shows that even before these bright lines crossing the sP, the resistance already starts to change near the zero displacement field. So the charge puddle is also not the reason of the D -field dependent resistance in the sP.

In another sample that the twist angle is smaller than 0.5° , besides two satellite point, we can also see third generation of satellite point (tP). Device 2 has a twist angle of 0.7° (figure 3.6), and a the hole side we see a tertiary Dirac point, and when we look at the displacement field dependence of this tP, the linear slope is different from the sP of the hole side. It means that the displacement induced occupation imbalance between

the two layers is not the reason of the displacement field dependence of R_{xx} at the sP and tP. Device 3 has a twist angle about 0.6° (figure 3.7), and the D field dependence of sP and hole side tP is the same. We also see the electron side tP, and R_{xx} has a maximum at about -100 mV/nm, and the resistance is much smaller than other Dirac Points. To explain such displacement field dependence of R_{xx} we need to consider how the band structure changes with the perpendicular electric field between the two layers of bilayer graphene superlattice. For the hole side, the resistance increases at sP when we increase the displacement field, and the resistance of the third generation of satellite points (tP) increases when we decrease the displacement field. At the electron side, the satellite point opens a gap or the density of states there decrease as the D field decreasing, and the tP might do not open a gap, but it has a density of state minimum.

3.4 Tunable Hofstadter's Butterfly in Bilayer Graphene Superlattice

When the perpendicular magnetic field is turned on, the electrons in the bilayer graphene form Landau levels, and each level has[33] an additional degeneracy of 4, including 2 from the spin, and 2 from the valley. When the magnetic field is strong enough, the electric field induced valley degeneracy splitting and magnetic field induced spin degeneracy splitting compete with each other, and we can find the electric field tunable Landau level transition at fixed magnetic field (figure 3.9 (b)), as first reported by Columbia group [43]. This appearance of the LLs transition means that our device has a very high transport mobility.

Besides the Landau levels of the normal bilayer graphene, the bilayer graphene

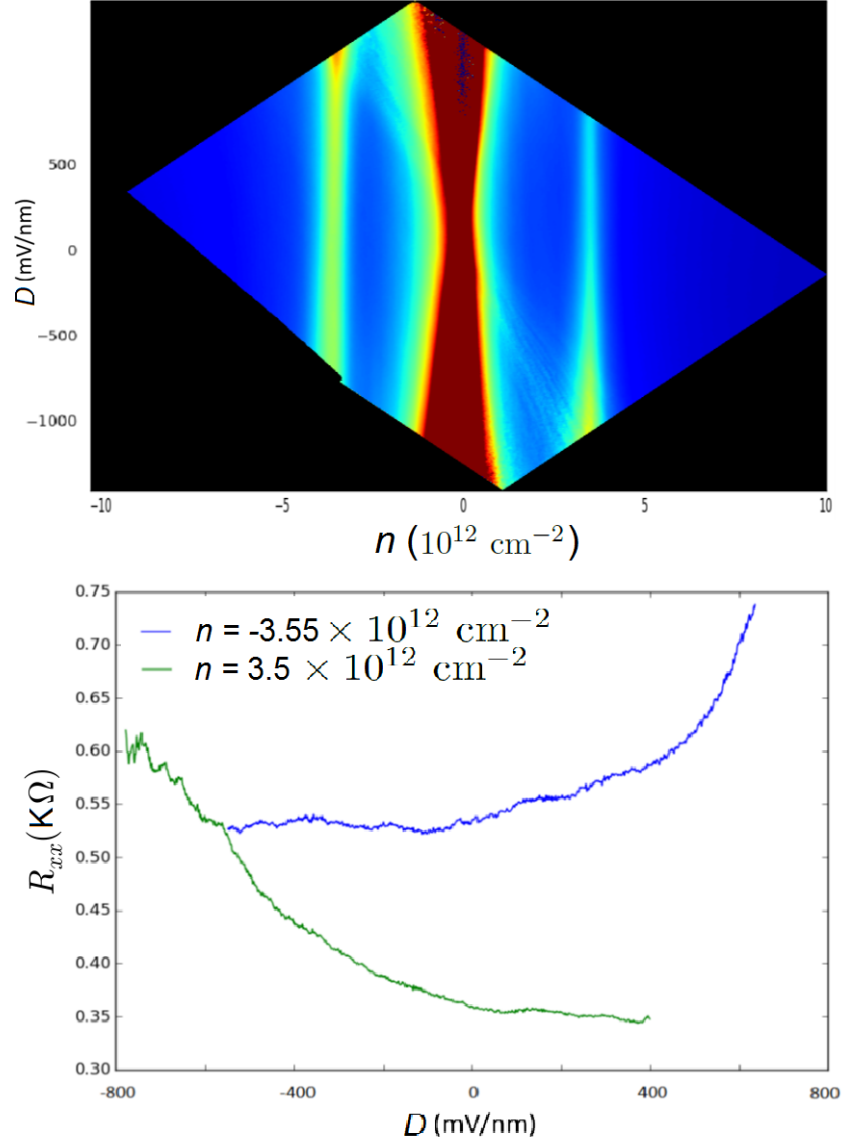


Figure 3.5: (up) R_{xx} pattern, x is carrier density, and y is displacement field. (down) The resistance vs displacement field at sP. The electron side: sP is at $n = 3.5 \times 10^{12} \text{ cm}^{-2}$, as D field increases the R_{xx} decrease. The hole side: sP is at $n = -3.55 \times 10^{12} \text{ cm}^{-2}$, the R_{xx} increase with increasing D field.

superlattice has other groups of LLs from the new generation of Dirac points, and form the Hofstadter's butterfly pattern[41]. Here we choose the white lines in figure 3.8 (a) to sweep along with the magnetic field. One line corresponds to the zero displacement field, the other line is fixed at 230 mV/nm upward displacement field. In figure 3.8 (c)(d) the Hofstadter's butterfly at the two different D fields. (c) When the D field

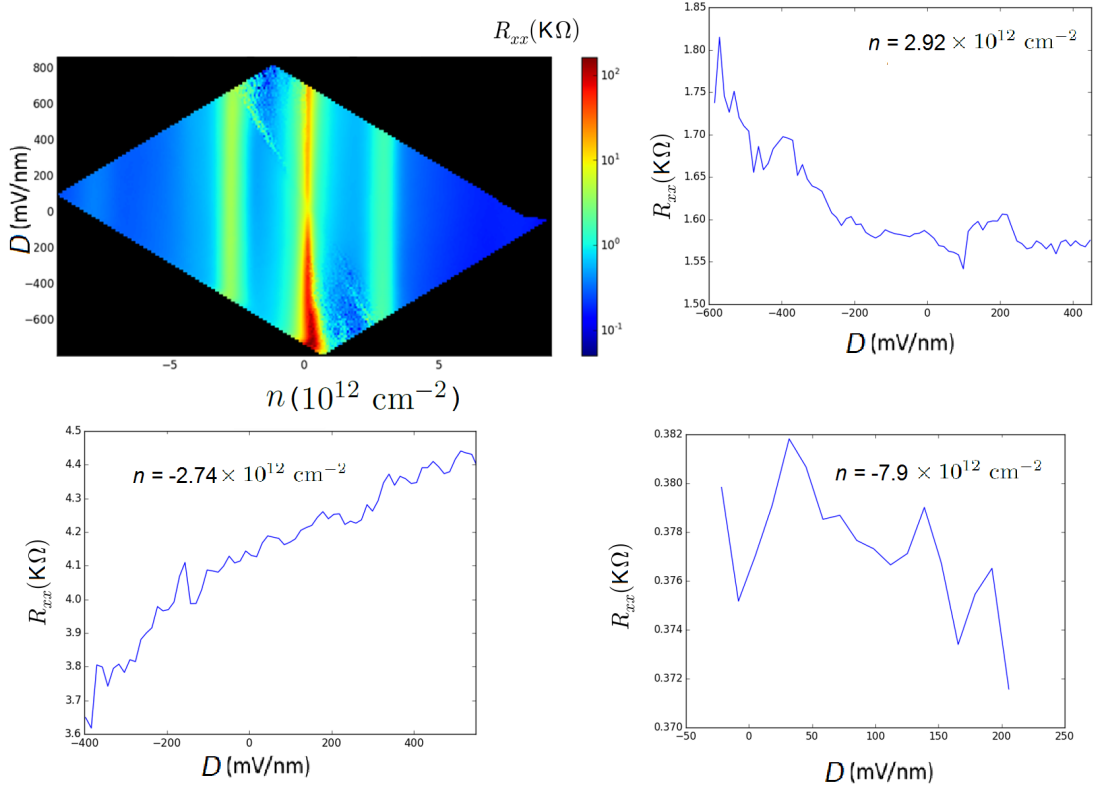


Figure 3.6: R_{xx} at $B = 0$ T for the sample 2. D field dependence of R_{xx} at the sPs is the same as sample 1. At the hole side we also see a tertiary Dirac point (tP) at $n = -7.9 \times 10^{12} \text{ cm}^{-2}$, and the D field dependence of this tP at the hole side is opposite compared with that of the sP at the hole side.

is zero we can see that at the electron side the Landau levels split when the LLs from the sP cross with those from charge neutrality point (CNP), the LLs from CNP split. At the hole side, when the two groups of LLs cross, at low density we did not see any splitting, because we find that after the crossing, the LLs from CNP just moves down and continue with the higher LLs, and the filling factor changes 4. This 2π berry phase change is different from the case of the monolayer graphene superlattice which the berry phase shift is π [50] [51]. At high carrier density, we see the LLs splitting just the same as electron side. In the figure 3.8 (d) we have the Hofstadter's butterfly pattern at upward displacement field at 230 mV/nm. At the electron side, when the two groups of LLs cross, the splitting disappear. The upward displacement field suppress the valley

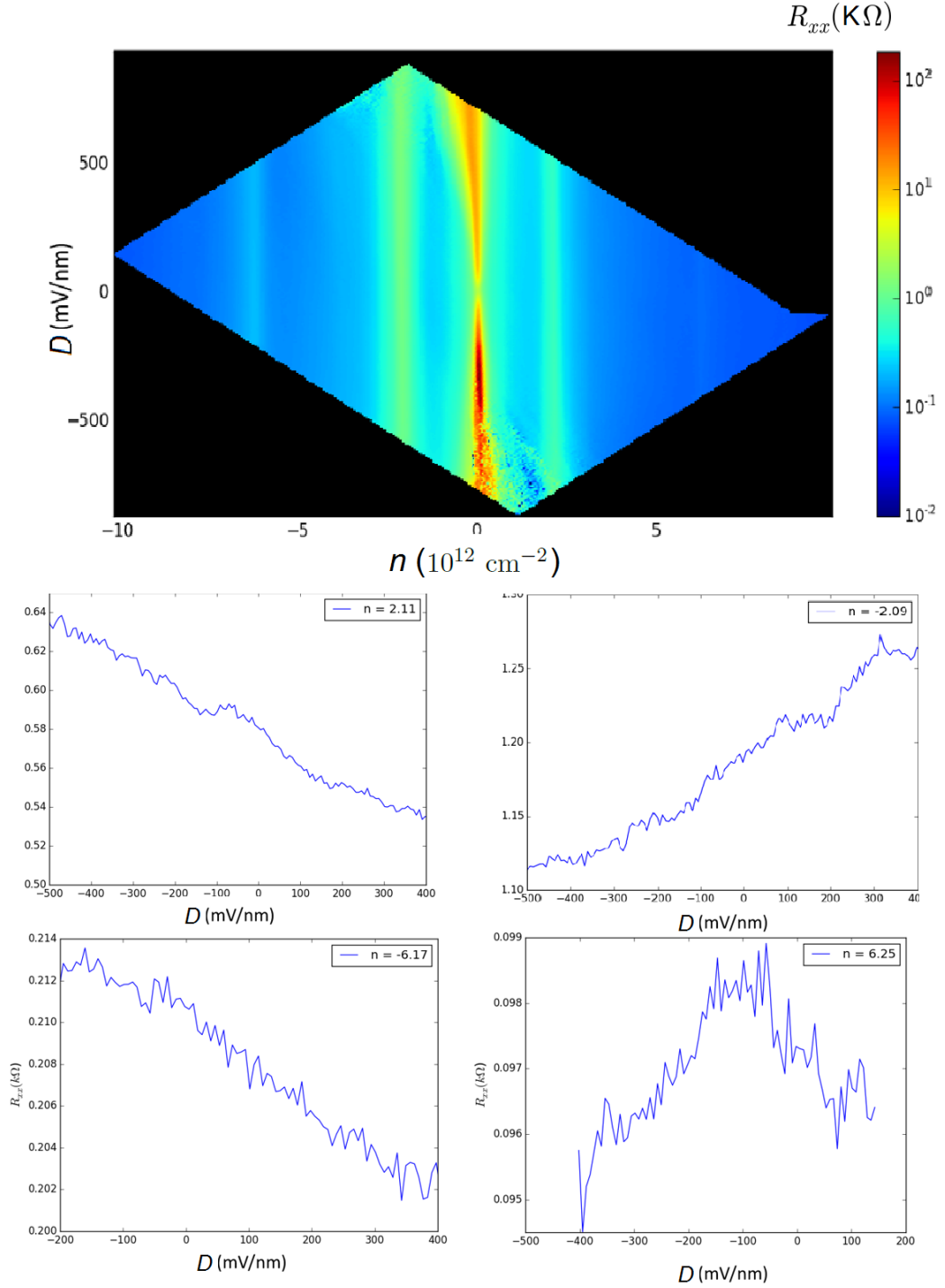


Figure 3.7: R_{xx} at zero B field for device 3. We see five Dirac points here, and we repeat the same D field dependence at the sP and the hole side tP. For the electron side tP, when we increase the D field, the R_{xx} increases first and then decreases.

symmetry breaking and we only see a LLs with degeneracy of 4. At the hole side, as long as the LLs cross, the LLs from the CNP split, and the zag band shows much more

features between the two LLs. However, the crossing just shifts the LLs as filling factor of 4, which is the same as the $D = 0$. It means that the displacement field won't change the Berry phase of the sP.

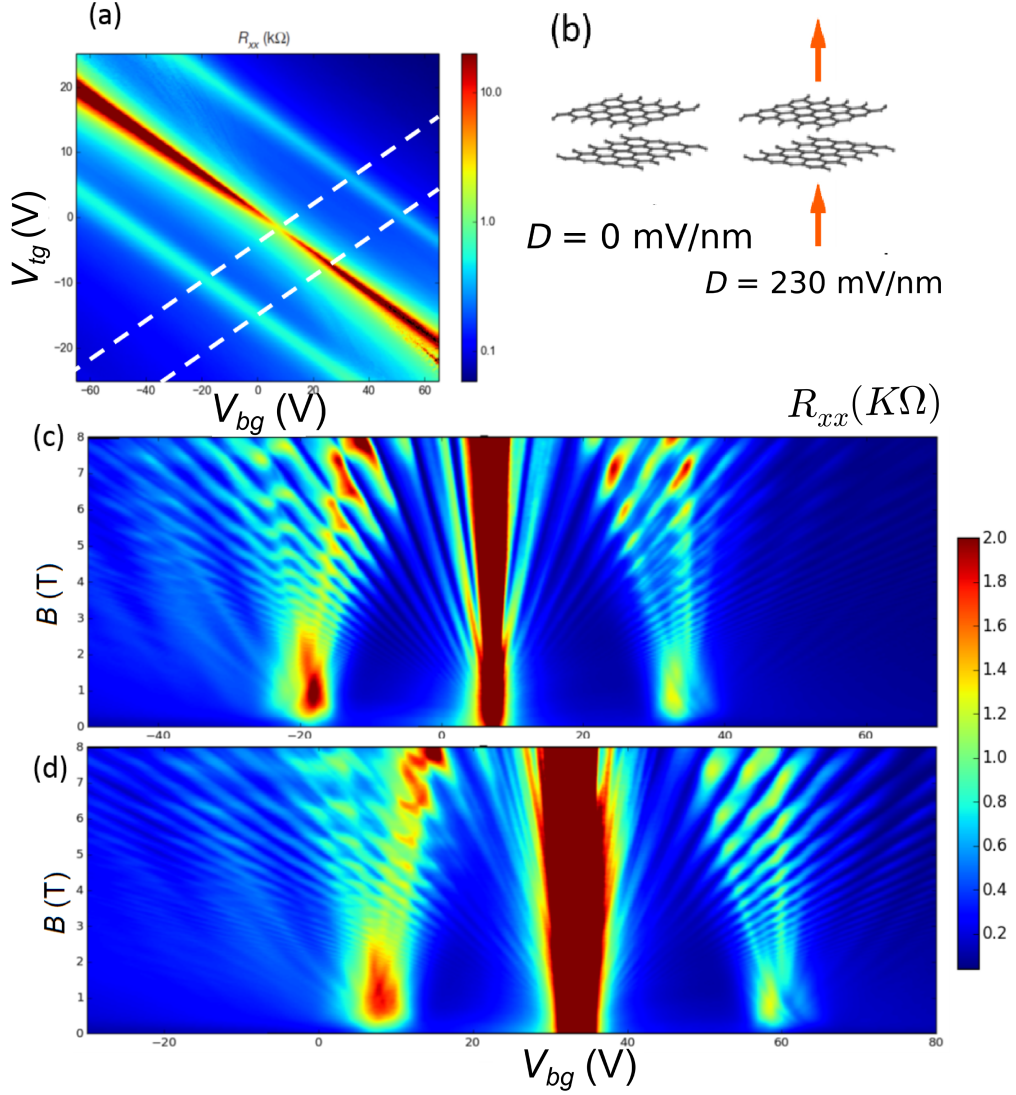


Figure 3.8: (a) Two lines for the butterfly pattern of (c) and (d). (c) Zero displacement field, in the electron side landau levels (LLs) from the CNP split when they cross the LLs from the sP. On the hole side the splittings only appear at large density. (d) 230 mV/nm upward displacement field. The splitting at the electron side disappear.

This electric field tunable Hofstadter's butterfly pattern is very interesting, since it looks like the valley degeneracy is already lifted at zero electric field, which is

different from the normal bilayer graphene [52], where the valley degeneracy is only lifted by a finite electric field. Additionally, when the electric field is tuned to some non-zero value, the Landau level splitting disappears. This behavior encouraged us to sweep the density and electric field at a fixed high magnetic field. In the figure 3.9(a), we measure R_{xx} vs dual gates at a constant magnetic field $B = 8$ T. Besides the displacement field tunable Landau level transitions near the CNP, we also see Landau level splitting at the right-up corner and left-down corner. Then we transform the x and y axis from the two gates to the density and displacement field, obtaining the figure 3.10(a). The bright line in the center corresponds to zero total density, and an electric field induced gap in the normal bilayer graphene. When we increase the carrier density, we can see other perpendicular lines which are the Landau levels, and the dark lines between the Landau levels are the gaps. In the figure 3.10(b) we get the line cut of the R_{xx} at a fixed displacement field 320 mV/nm. On the electron side, the degeneracy of LLs is 4, and the position of the gaps are just at $4 \times N$, where N is positive integer. This means both the spin and valley degeneracy are conserved, and the electric field suppresses those degeneracy lifting. In the hole side, we can see much more dips, and the number of the dips just doubles that of the non-split LLs, but the position of the gaps is not $4 \times N$ or $2 \times N$. In the figure 3.10(c), we have the line trace of R_{xx} at a displacement field -320 mV/nm. At the hole side, the spin and valley degeneracy are conserved, and the position of the dips is just $-4 \times N$, which means on the hole side the negative electric field will suppress the spin and valley symmetry breaking, and the degeneracy of the LLs is 4. On the electron side, the valley degeneracy is lifted, and we can see the dips at the position of $2 \times N$.

The evolution of the LLs with the varying electric field is not the same as the LLs transition, where we can see one LL becomes two or two levels become one (figure 3.9 (b)). Instead the pattern here is that one level with degeneracy of 4 disappears gradually, and another new LL show up when we increase the D field in one direction (figure 3.5). And we find that the electron side and hole side show opposite displacement field dependence. This e-h asymmetry only shows up in the bilayer superlattice, where the valley symmetry conserved line is tilted. While in a normal bilayer graphene, such line is just the zero displacement line[52]. I hypothesize that the tilted angle for that line might depend on the alignment angle, because in our measurement we have some devices where the bilayer graphene is exactly aligned with hBN, the valley symmetry conserved line seems to have a too large tilt angle and cannot be seen. In other two device whose angles are around 1° the splitting is very clear. But we do not yet have enough devices to verify this hypothesis.

3.5 Twisted Bilayer Graphene with the Bottom Layer Graphene Aligned to the hBN Lattice

Hre we discuss bilayer graphene where the bottom layer is aligned to the hBN, while top top graphene layer is placed with a twist angle. The difference between this and the previous system is clear, because the interlayer hopping energy in the natural bilayer is about 0.4 eV [32], which is much larger than the twisted bilayer graphene interlayer coupling[48]. In this sense the bilayer graphene superlattice is considered as one conducting layer, but twisted bilayer graphene is weakly coupled two layers [49]. In order to find out the difference between the two systems, we made a twisted bilayer

graphene and the bottom layer graphene is aligned with boron nitride. The alignment was checked by the Raman, and the twist angle is calculated to be about 1.1° .

Since when the Raman of twisted bilayer graphene is similar as that of natural bilayer, I use the Raman measurement[53] to exclude the superlattice created by the two monolayer graphene. In the latter case we can get another Hofstadter's butterfly pattern [54] [55] and van Hove singularity [56][57]. The Raman signal of our sample is shown in figure 3.12, which means the twist angle is large.

At zero magnetic field, we sweep two gates at about 1.5 K (fig. 3.13 (a)), we can see three Dirac points. The CNP corresponds to the total density

$$n_{tot} = n_{bottom} + n_{top} = 0 \quad (3.4)$$

While the satellite means that

$$n_{bottom} = n_0 \quad (3.5)$$

where n_0 is the density of the sP in the bottom layer graphene superlattice. The slope $\frac{dV_{bg}}{dV_{tg}}$ at the line of CNP and the line of sP are different, and we can calculate the screening of the graphene by the slope difference. Assuming C_{1g} , C_{2g} , C_{1t} and C_{2t} are the capacitance between the graphene and gate, where we use the following label: bottom layer graphene (label 1), the top layer graphene (label 2), and the back gate (label g), top gate (label t). Then we have equation

$$n_{tot} = n_b + n_t = C_{1g}V_g + C_{1t}V_t + C_{2g}V_g + C_{2t}V_t \quad (3.6)$$

$$= (C_{1g} + C_{2g})V_g + (C_{1t} + C_{2t})V_t \quad (3.7)$$

Assuming the screening factor is $1 - \chi$, then we have $C_{2g} = \chi C_{1g}$, $C_{1t} = \chi C_{2t}$,

so

$$n_{tot} = (1 + \chi)C_{1g}V_g + (1 + \frac{1}{\chi})C_{1t}V_t \quad (3.8)$$

$$n_b = C_{1g}V_g + C_{1t}V_t \quad (3.9)$$

At the CNP, we have $n_{tot} = 0$, so the slope of charge neutrality line S_{CNP} is

$$(1 + \chi)C_{1g}V_g + (1 + \frac{1}{\chi})C_{1t}V_t = 0 \quad (3.10)$$

$$S_{CNP} = -\frac{1 + \frac{1}{\chi}}{1 + \chi} \frac{C_{1t}}{C_{1g}} = -\frac{1}{\chi} \frac{C_{1t}}{C_{1g}} \quad (3.11)$$

At the sP, we have $n_b = n_0$, then the slope dV_{bg}/dV_{tg} for the line of the sP S_{sP}

is

$$S_{CNP} = -\frac{C_{1t}}{C_{1g}} \quad (3.12)$$

Combining the equations above we get

$$\chi = \frac{S_{sP}}{S_{CNP}} \quad (3.13)$$

In our device $S_{sP} = 2.133$ and $S_{CNP} = 3.689$, so the screening $1 - \chi$ is about 40%.

The displacement field dependence of the sP shows a different result compared to the bilayer graphene superlattice. We find that at the hole side sP (fig. 3.13 (b)), when we increase D field, R_{xx} decreases. The reason is that the positive electric points upward, and the hole carriers are "pushed" to the top, and the scattering from the

bottom aligned hBN is suppressed, so the resistance is smaller. On the other hand, if the D field is negative, and the holes are "pushed" to the bottom aligned hBN, then the scattering is stronger, and the resistance is larger. But for the electron side sP (fig. 3.13 (c)), the D field dependence is not monotonic, it has a minimum, but the overall tendency is that as the displacement field increase the resistance increase, which can be explained by the above discussion.

Now we turn on the magnetic field. Since the top layer graphene is not aligned, all the Landau levels are supposed to come out from the CNP. While bottom layer is a graphene superlattice, and Hofstadter's butterfly pattern will show up. The weak coupling between the two layers leading to weak interaction between the LLs from the separate two layers, and the total pattern is just the sum of normal graphene Landau Levels and Hofstadter's butterfly (fig. 3.14 (up)). We fix the displacement field at zero, which guarantees that the carrier densities in the two layers are the same. Using SdH oscillation we calculate the density vs the gate voltage, and the interlayer displacement field. Then we transform x and y axis from two gates to carrier density and displacement field at $B = 8$ T (fig. 3.14 (down)). Before the total carrier density is small, we can see the Landau level crossing, and each group come from each graphene layer. The data is the same as the paper[49]. As the carrier density increases, the LLs from the sP interfere those from CNP, forming patterns like ripple, which is never reported before.

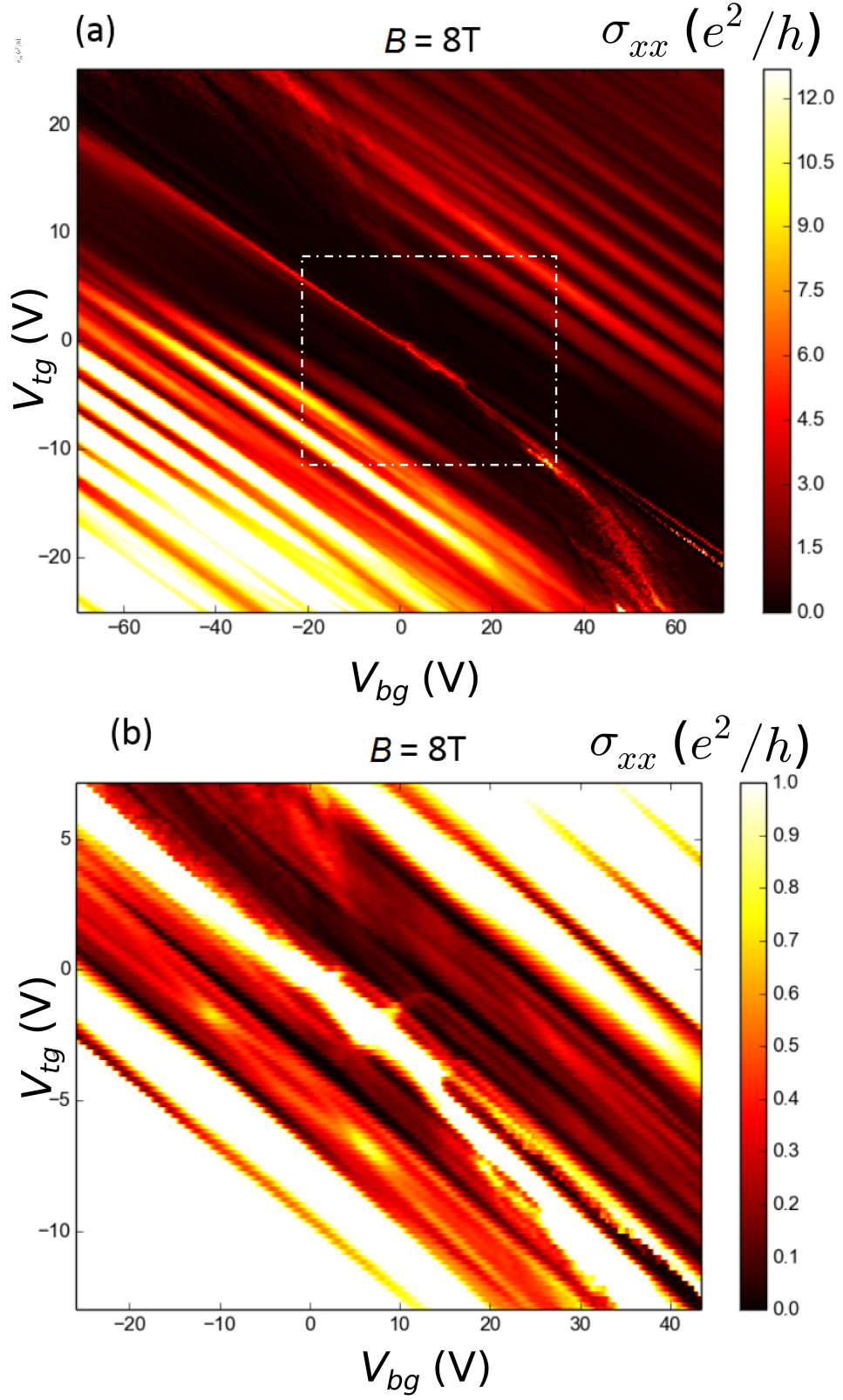


Figure 3.9: (a) The dual gate sweep at $B = 8$ T. Near the sP, we can see the Landau level splittings. (b) Close to the CNP, electric field dependent LLs transitions happens because of degeneracy lifting of the spins and valleys.

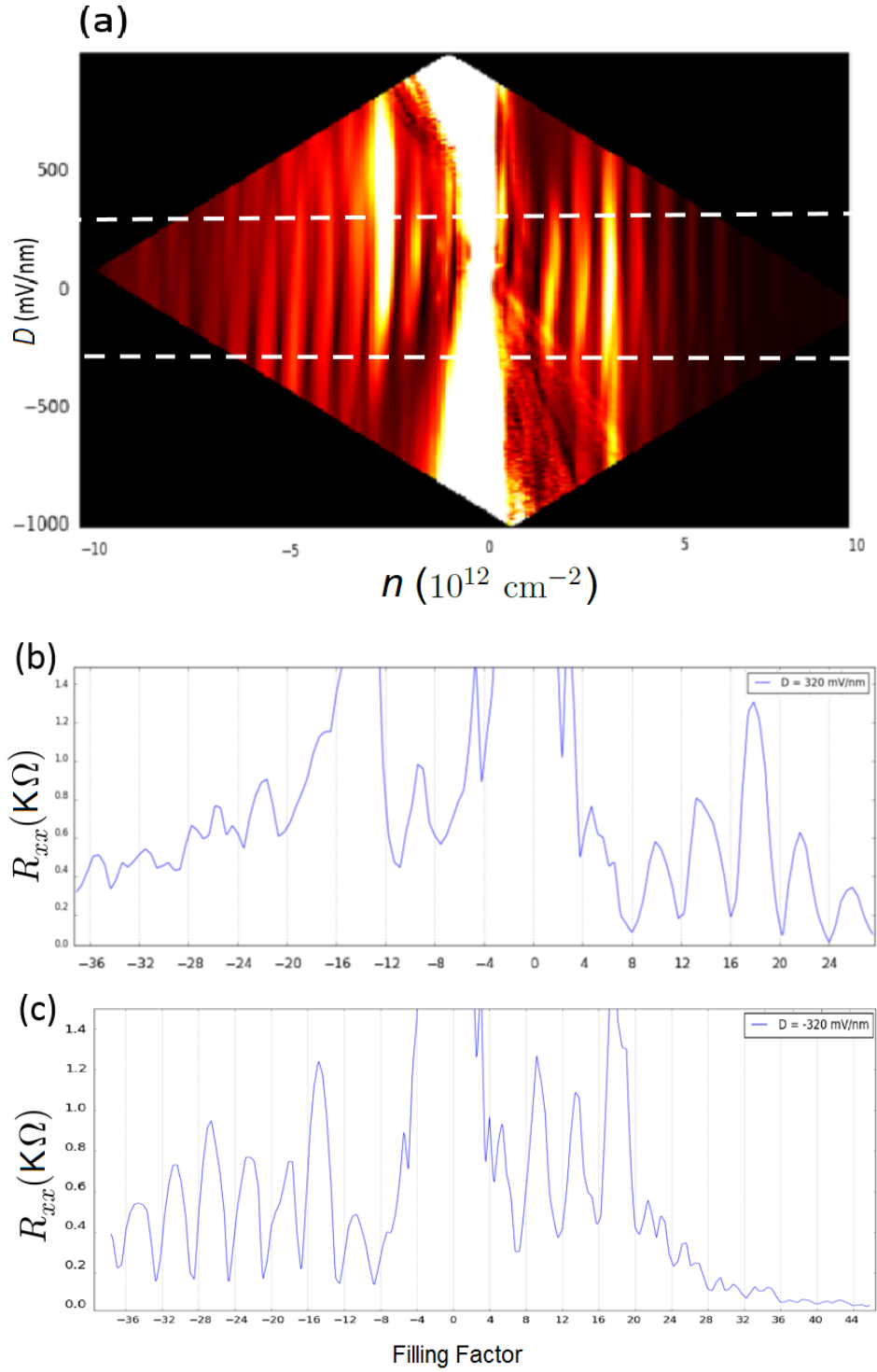


Figure 3.10: (a) Landau levels at $B = 8 \text{ T}$, x axis is carrier density, y axis is displacement field. Two white dashed line is the line trace for (b) and (c). (b) line trace of figure (a) at constant D field 320 mV/nm . The x axis is the LLs filling factor ν , We can see LLs splitting at $\nu < 0$. (c) line cut of R_{xx} at constant D field -320 mV/nm . The LLs splitting happens at $\nu > 0$.

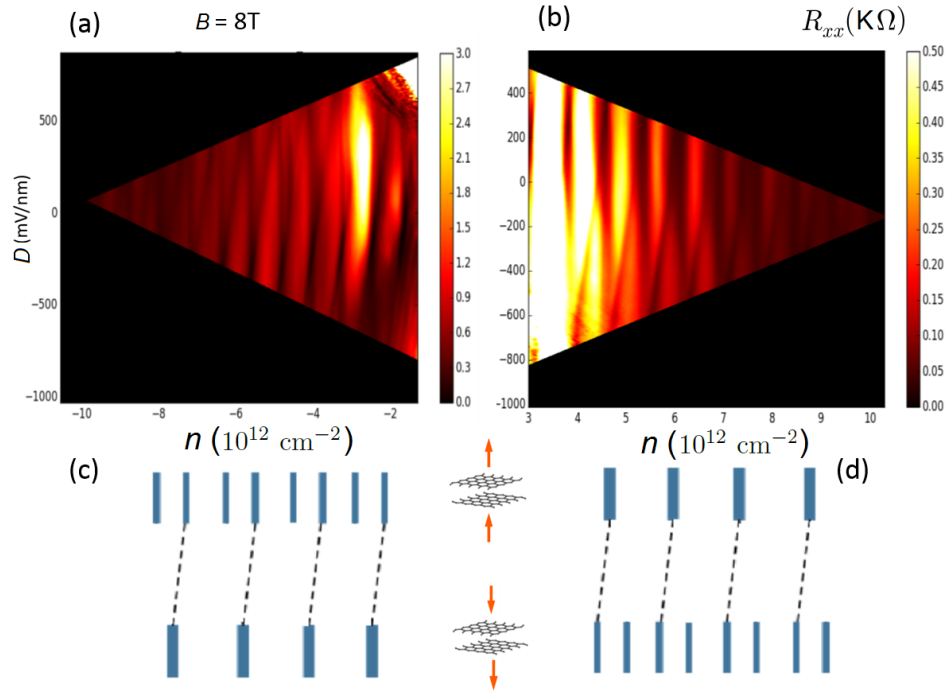


Figure 3.11: (a) Landau level splitting at hole side ($n < 0$). (b) LLs splitting at electron side ($n > 0$). (c)(d) Schematic diagram of the LLs splitting in the hole and electron side. The electron-hole asymmetry shows up.

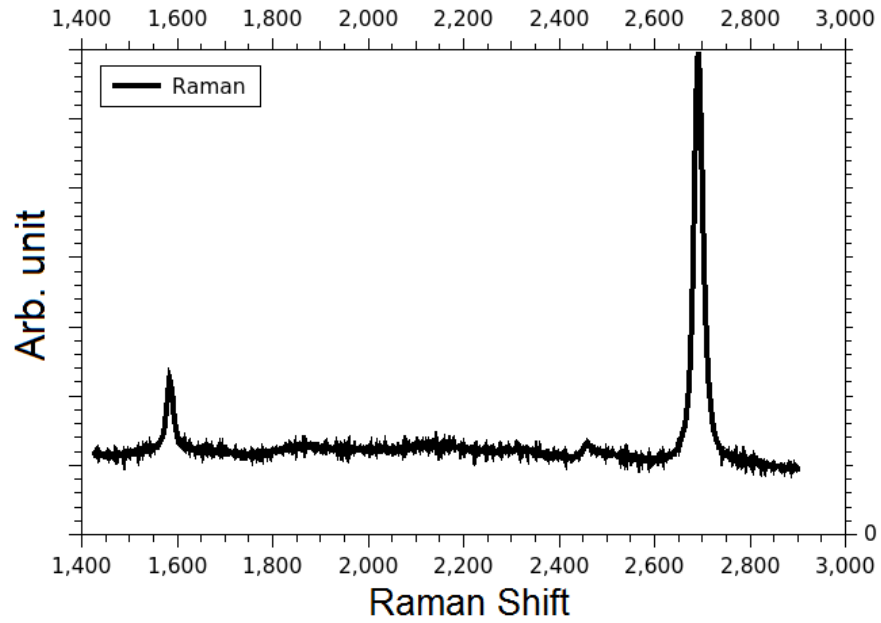


Figure 3.12: Because the 2D peak is much higher than the G peak, this indicates the twist angle is large.

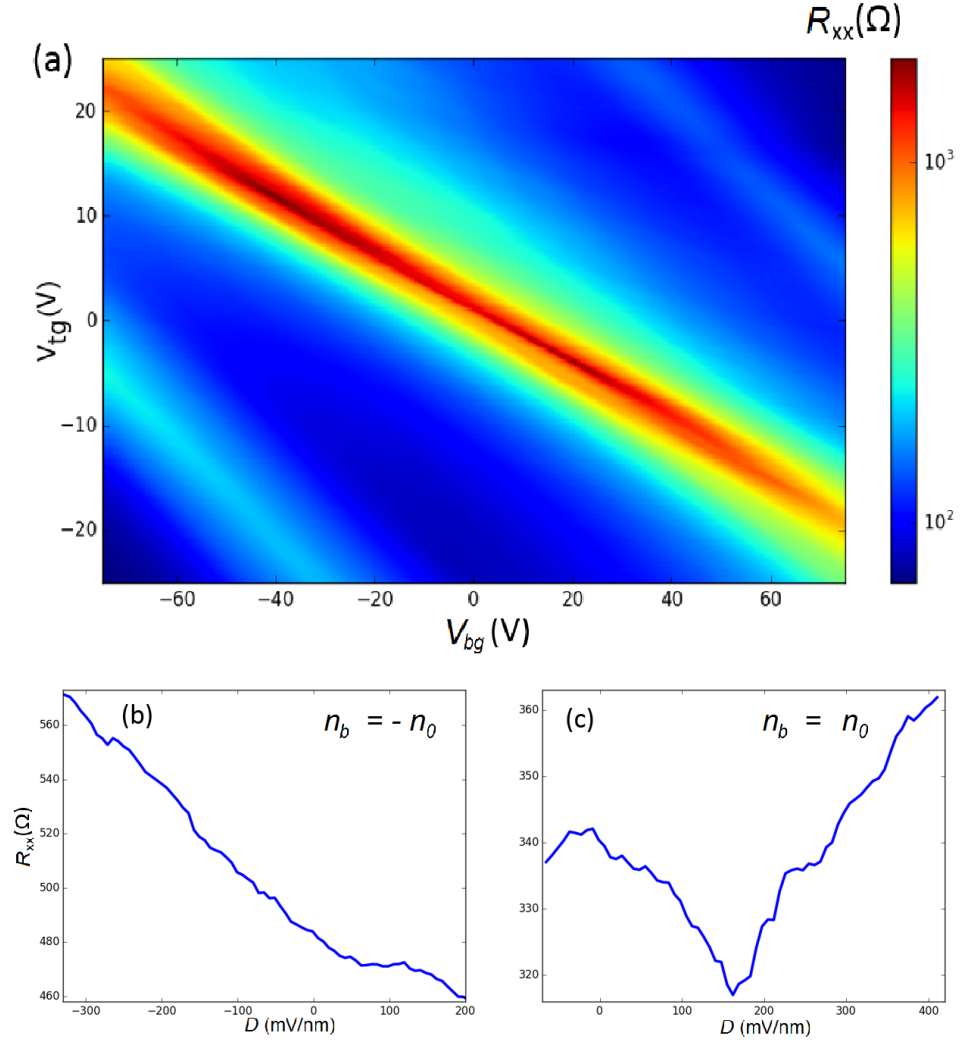


Figure 3.13: (a) R_{xx} of two gates dependence. The bright line in the center is $n_{tot} = 0$, and the other bright line is the sP of the bottom layer superlattice. (b) The displacement field dependence of R_{xx} at the hole side sP $n_b = -n_0$, n_b is the electron density of the bottom layer graphene. (c) The displacement field dependence of R_{xx} at the electron side sP $n_b = n_0$.

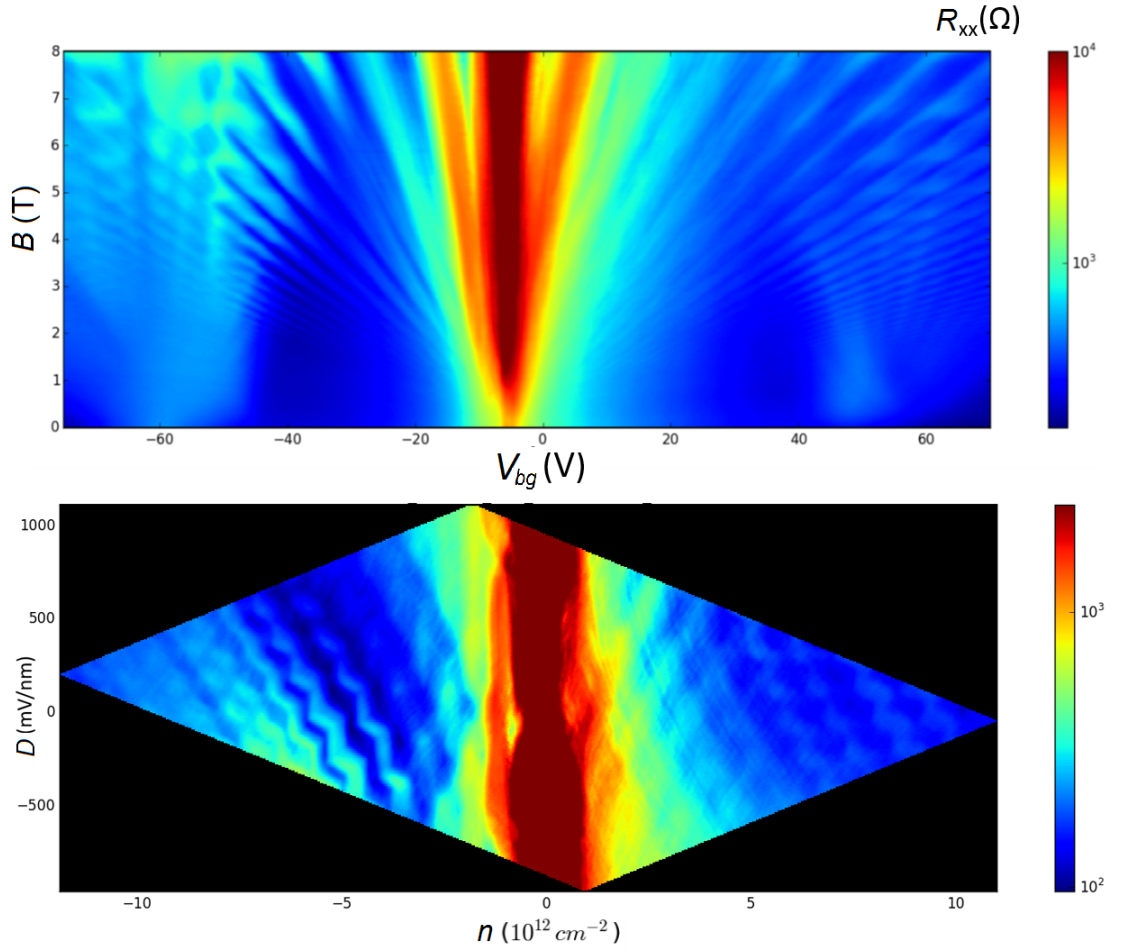


Figure 3.14: (up) The Hofstadter's butterfly pattern of the twisted bilayer graphene with the bottom layer graphene superlattice. (down) The Landau Level crossing at the $B = 6$ T. In the small density regime we have Landau Level crossing and in the large density regime we have the unusual ripple pattern.

Chapter 4

Landau Level Crossing and Screening in Twisted Trilayer Graphene

4.1 Introduction to Twisted Bilayer and Twisted Trilayer Graphene

Twisted multilayer graphene is currently being studied intensively, for both twisted bilayer graphene and twisted trilayer graphene. The most interesting thing in the twisted multilayer graphene is the moiré Superlattice and Hofstadter's butterfly when the twist angle is small[58] [59]. A few year ago, the experiment of twisted bilayer device with large twist angle was reported, in which the two layers is weakly coupled[49]. Recently the twisted trilayer graphene with small twist angle is attracting people's attention[60] [61] [62] [63]. But the transport measurements need a very well designed device, in which the twist angle cannot be larger than 1.5° for the gate tuning. In the meantime

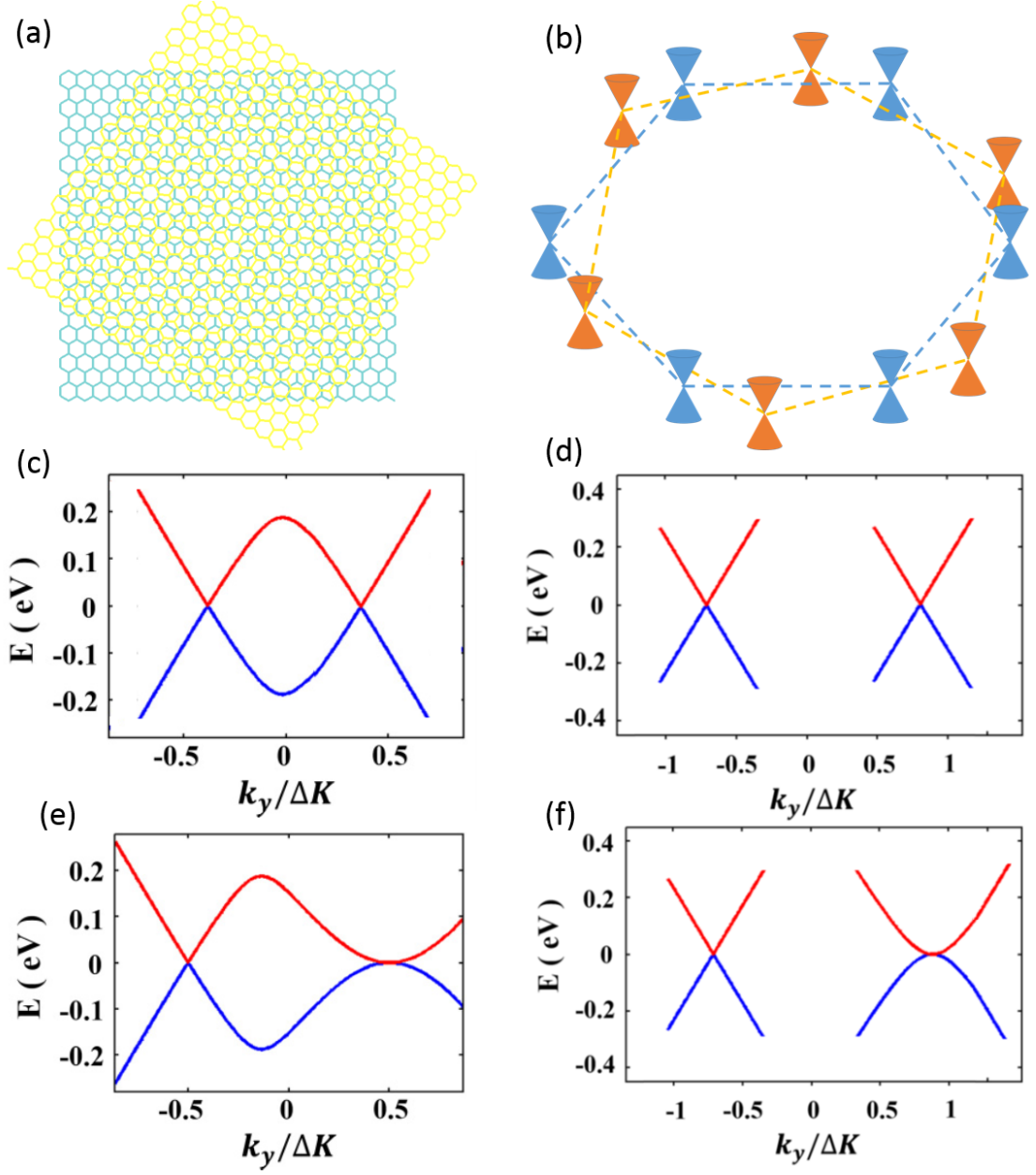


Figure 4.1: (a) Schematic diagram of large angle twisted bilayer or twisted trilayer graphene. (b) Low energy dispersion of twisted bilayer graphene. since the twist angle is large, the Dirac points separate far away. (c)(d) Low energy band structure of twisted bilayer graphene with small and large twisted angle. (e)(f) Low energy band structure of twisted trilayer graphene with small and large twisted angle. The graphs of (c)(e) are adapted from [62].)

if twist angle is smaller than 1° , the sample is unstable and the high temperature O_2 or H_2 gas anneal will make the twist angle zero. So as a first trial, it is reasonable to make a large twisted angle trilayer graphene sample.

Fig. 4.1 (a) shows a schematic diagram for the twisted trilayer graphene with a large angle. Then the low energy dispersion is shown in (b), where the Dirac cones from the two layers are separated far away. As discussed earlier, the band structure of monolayer graphene is linear while bilayer graphene is parabolic. In the case of twisted bilayer and trilayer graphene the small twist angle will form a van Hove singularity (Fig. 4.1(c)(e)), where it is supposed to see the gaps and Hofstadter's butterfly pattern in the transport measurement. If the twist angle is large (fig. 4.1(d)(f)), the Dirac cones from the two layers is weakly coupled, and the scattering between the two layers is negligible. In the actual measurement, the inter-layer tunneling is weak, but not zero, and depends on the temperature[48]. When the magnetic field is turned on, the LLs from the separate two layers have different dual-gate dependence, so they have different slope in the back gate and top gate plane[49]. The reason for different slopes is the screening between the twisted two layers, and then the capacitance of the top layer graphene and bottom layer graphene induced by top gate or back gate is different:

$$n_1 = (C_{1t}V_{tg} + C_{1g}V_g)/e, \quad (4.1)$$

$$n_2 = (C_{2t}V_{tg} + C_{2g}V_{bg})/e. \quad (4.2)$$

We assume the interlayer capacitance of the two layers is C_{int} , so

$$\frac{1}{C_{2g}} = \frac{1}{C_{1g}} + \frac{1}{C_{int}}, \quad (4.3)$$

$$\frac{1}{C_{1t}} = \frac{1}{C_{2g}} + \frac{1}{C_{int}} \quad (4.4)$$

so the slope of the LLs from bilayer graphene S_m and the LLs from monolayer graphene S_b

$$S_m = \frac{V_{tg}}{V_{bg}} = \frac{C_{1g}}{C_{1t}}, \quad (4.5)$$

$$S_b = \frac{V_{tg}}{V_{bg}} = \frac{C_{2g}}{C_{2t}}. \quad (4.6)$$

This simple model of capacitors in series can explain the crossing of the LLs at a fixed magnetic field with dual-gate sweep.

When we have a twisted trilayer graphene with large angle, the two layers also have weak interlayer coupling. So we can use the same two capacitors model as the twisted bilayer graphene, except that one layer is natural bilayer graphene instead of monolayer. Since the low energy band structure for bilayer graphene is parabolic, and at the same energy the density of state is much larger than the monolayer graphene, so the slope of V_{bg}/V_{tg} is larger (with the bilayer on bottom) than the twisted bilayer graphene. When the magnetic field applied, the number of filled LLs at the same Fermi energy is different, so we will see much phenomena that not been observed before.

4.2 Tunable Filling Ratio between Monolayer and Bilayer graphene

As in the figure 4.2, we measure the longitudinal resistance of the twisted trilayer graphene by four terminal measurement setup using a lock-in amplifier. The measurement is set at about 1.5 K, while the magnetic field is applied the temperature might be 0.1 K higher. The bright line is the resistance maximum. Since the twist angle is large in our device and the bilayer and monolayer graphene can be considered as separate layers, so we use a very simple model of parallel conducting plates to simulate the system. Then the total resistance of the parallel plates is

$$R_{total} = \frac{1}{\frac{1}{R_m} + \frac{1}{R_b}} \quad (4.7)$$

where R_{total} is the total resistance of the twisted trilayer graphene, and R_m is the resistance of the monolayer graphene and R_b is the bilayer graphene resistance. As we discussed above, the capacitance of bilayer and monolayer graphene on the back gate and top gate are different, so the Dirac point of bilayer graphene $n_b = 0$ has a different slope V_{bg}/V_{tg} from the Dirac point of monolayer graphene $n_m = 0$. The resistance maximum line corresponds to the total density $n_{total} = n_b + n_m = 0$. When we increase the internal electric field, the total resistance firstly increases and then decrease.

When we sweep the two gates at a fixed high magnetic field, the slopes of V_{bg}/V_{tg} is different for the Landau level from the two layers of the twisted multilayer graphene. In the twisted bilayer graphene, two groups of Landau levels follow straight lines in the plot, and the crossing points of the LLs do not make any shift to the LLs. But in the twisted trilayer case, at the crossing points, the LLs will shift. On the other hand, if we fix the ratio of the two gate V_{bg}/V_{tg} and sweep the gate and magnetic field, then we can see two groups of LLs cross, and LLs from the bilayer graphene is nearly linear and the LLs from the monolayer graphene is parabolic. If we change the ratio of the two gates, the pattern will change because of different filling rate between the two layers. In zero magnetic field, we get the slope for the neutral charge line $n_{bottom} + n_{top} = 0$, which is $S_{n=0} = V_{tg}/V_{bg}$, so the line of zero internal electric field is $S_{D=0} = -S_{n=0}$. Along this zero D -field line we sweep the magnetic field, and get the LL crossing pattern. In figure 4.3 (left), we numerically simulate the LL crossing. The x axis is the energy of band from the Dirac point, and the y axis is the magnetic field, the linear solid lines is the LLs of the bilayer graphene, and the parabolic dashed lines are the LLs of the

monolayer graphene. The formula we use for the LLs of monolayer graphene is:

$$B = \frac{\varepsilon^2 e}{2N\hbar v_f^2} \quad (4.8)$$

And for bilayer:

$$B = \frac{\varepsilon m^*}{\hbar \sqrt{N(N-1)}} \quad (4.9)$$

Where N is the Landau level index and m^* is the effective mass in the bilayer graphene, ε is the band energy, v_f is the Fermi velocity of graphene ($\sim 1 \times 10^6$ m/s). The effective mass is about between $0.03m_e \sim 0.05m_e$, which m_e is the mass of electron. Here we use $m^* = 0.04m_e$ to fit the experimental results.

In figure 4.3 (right) we get the longitudinal resistance R_{xx} of the twisted trilayer graphene, with the internal perpendicular electric field is zero. This experimental data is the same as the numerical result we got at figure 4.3 (left).

When we change the ratio of V_{bg}/V_{tg} , the filling rate of the bilayer graphene and monolayer graphene changes. In the figure 4.4(c), we fix the ratio $\frac{V_{tg}}{V_{bg}} = \frac{1}{2} \times S_{D=0}$, where $\frac{V_{tg}}{V_{bg}} = S_{D=0}$ is the case that the interlayer perpendicular electric field is zero. In this filling rate the bilayer graphene has a much higher density than the monolayer, and the interval for the monolayer graphene LLs is much larger than the case that the carrier density in the monolayer and bilayer graphene is the same (Fig. 4.3 (right)). Because the density in the monolayer and bilayer graphene is different and the difference will increase if we increase the carrier density, so the interlayer electric field is also increasing to support the carrier density difference. In this sense, the interlayer electric field will cause an energy difference in the two layers. In the figure 4.4(d) we tune the ratio of two

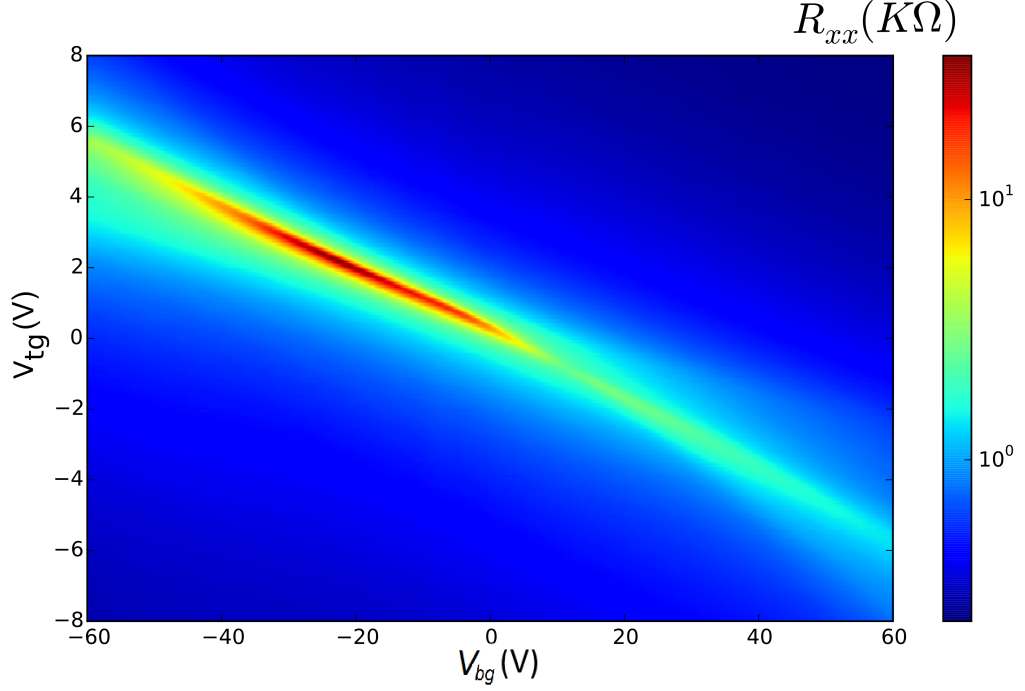


Figure 4.2: Longitudinal resistance of twisted trilayer graphene at zero magnetic field. The resistance is measurement by four terminal setup by low frequency lock-in. The bright line is the resistance maximum line.

gates to satisfy $\frac{V_{tg}}{V_{bg}} = \frac{1}{4} \times S_{D=0}$, so we find that the interval for the LLs from monolayer graphene is even larger, since the density in the monolayer on the top is smaller. In the figure 4.4(a) and (b) we try to get the same LL crossing pattern with the experimental data. Because of the screening, and the screening of bilayer graphene is stronger than that of the monolayer, so the filling rate ratio n_m/n_b is not $\frac{1}{2}$ or $\frac{1}{4}$. We make some correction and find that $n_m/n_b = 0.65$ instead of $\frac{1}{2}$ best fits for the data in figure 4.4(c). Also we use $n_m/n_b = 0.45$ (fig 4.4(b)) instead of $\frac{1}{4}$ to fit the data in figure 4.4(d).

When the LL crossing, the LLs from the monolayer graphene and bilayer graphene have the same energy, and they are degenerate levels. At a fixed magnetic field, as the carrier density increases and reach the crossing point, the interval of σ_{xy} becomes $8\frac{e^2}{h}$. In figure 4.5 (a) we get the σ_{xy} of the device 2, where we can see very

clear plateaus. When the LLs cross, the edge of the plateaus becomes irregular. In figure 4.5 (b) we get several line trace of the σ_{xy} of the magnetic field at $B = 5, 5.5, 6, 6.5, 7, 7.5$ T. We can see the double of the plateau interval when LLs cross. As the magnetic field increases, the LL from the monolayer that crossing with the same LL of the bilayer graphene will change, and we can see the interval-doubled LL will also change. This means the double of the σ_{xy} comes from the crossing of the LLs, where the LLs are energy degenerate and the electrons need fill both layers. Now we consider a simple model, since the coupling between the monolayer graphene and bilayer graphene is weak, so the total conductance can be written as:

$$\sigma_{total} = \sigma_{bilayer} + \sigma_{monolayer}. \quad (4.10)$$

The conductance can be longitudinal component σ_{xx} and transverse component σ_{xy} . Assume the bilayer graphene at N LL, and the $\sigma_{xy} = 4N\frac{e^2}{h}$, while the monolayer graphene has $\sigma_{xy} = 4M\frac{e^2}{h}$, so the total Hall conductance is just the sum of the two weak coupled two parts: $\sigma_{xy} = 4(M+N)\frac{e^2}{h}$. So when the density increase to across LLs from bilayer or monolayer, the “jump” of $\Delta\sigma_{xy} = \Delta(4M\frac{e^2}{h})$ or $\Delta(4N\frac{e^2}{h})$, which is $4\frac{e^2}{h}$. On the other hand if when we increase the carrier density at a fixed magnetic field and the line cut just pass the crossing point of LLs, then the “jump” of Hall conductance is $\Delta\sigma_{xy} = \Delta(4M\frac{e^2}{h}) + \Delta(4M\frac{e^2}{h}) = 8\frac{e^2}{h}$. In the real sample, because of the impurities the energy of LLs is not perfect δ function but is broadened, so the “jump” transition is not exactly from 4 conductance quanta to 8 conductance quanta. In the figure 4.5 (right), we find that the “jump” can be 4, 8 in the unit of quanta conductance, which is consistent with the model. But we also find some “jump” in the value of 10 conductance quanta, which is larger than we predict in the calculation, and after the “jump” the plateau is shorter than normal and is not that flat. As discussed previously, the deviation comes

from the broadening of the LLs, and if the energy width of broadened LL is comparable to the energy interval between LLs, then we will see such change or “jump”. As shown in figure 4.6, when LL crossings happen, if the LLs is not broadened and the peak is very sharp, then one LL from the monolayer graphene has overlap with only one LLs from bilayer graphene. On other hand, if the LLs are broadened, one LL from monolayer graphene has overlap with two LLs from the bilayer, in which case the “jump” of σ_{xy} is larger than $8\frac{e^2}{h}$, and the plateau of LLs just become sloped.

Another interesting thing in the LL crossing experimental data is that after crossing, the LLs shift (Fig. 4.4(c)(d)). This kind of LLs shift does not show up in the twisted bilayer graphene sample. If we look at one LL of bilayer graphene, and increase the magnetic field, then after the crossing with LL from monolayer graphene, the slope dB/dV_{bg} just become much smaller suddenly, which means the sudden decrease of the capacitance of the bilayer graphene. The reason for this capacitance sudden decrease is that at the crossing point, the LL from monolayer and bilayer graphene are degenerate, and as we increase the carrier density in the whole twisted trilayer sample, the filling rate (capacitance) in the bilayer graphene just decreases.

4.3 Landau Level Crossing at $B = 8$ T

Next we fix the magnetic field at 8 T, and sweep two gates to get the longitudinal resistance (Fig. 4.7). The x and y axis are the back gate and top gate separately. The color scale for the R_{xx} has been transformed to logarithmic. Here we get a complex pattern, and I will discuss different regimes in this figure one by one.

First, we consider the lower left part, where we can see clearly that bilayer graphene LL features shift in the plot after crossing with the LLs of the monolayer (Fig. 4.8). In this regime, the carrier density is large and the electric field is near zero. The lower slopes is from the monolayer LLs, since the monolayer graphene is on the top part of twisted trilayer graphene sample, and the top gate will tune its density more. The other group of features with larger slope $|dV_{tg}/dV_{bg}|$ are from the bilayer graphene, since the bilayer graphene is on the bottom part and the back gate will tune its density more. When the crossing happens, the LLs of the bilayer graphene have a half-period shift. This shift is consistent with the phenomena we observed in fig. 4.4(c)(d).

Now we focus on the regime near the global Dirac point (fig. 4.9), where the LLs filling factor index in the monolayer is from -2 to 2 , and the LLs in the bilayer graphene has the filling factor index between -4 to 4 . The LL crossings form a series of hexagonal patterns. The similar hexagonal patterns also find in the regime shown in fig. 4.10, where the lowest LLs from the monolayer across the LLs in the bilayer graphene which have filling factors from 4 to 12 . We now consider the formation of the hexagonal pattern: two groups of parallel lines crossing with each other will form a series of rhombus pattern, however the LLs in the bilayer graphene have a small shift when crossing happens, and this shift will make the rhombus shape to a hexagonal shape. We also note that there are three separate regimes that have those hexagonal patterns. The left two groups of the hexagonal pattern are straightforward to understand, which are just the LLs from the bilayer graphene and monolayer graphene as we just discussed above. But the group of hexagonal pattern on the right side is different, since there is no LL from the monolayer graphene, and all these LLs are from the bilayer graphene. If this hypothesis is correct then the LLs from 0 to 4 intersect with the LLs from 0 to

-4, although they are both from the bilayer graphene. These self-intersecting LLs are unusual, and further work will be required to fully understand this behavior.

As mentioned above, in the regime shown in fig. 4.10 also have a hexagonal LL crossing pattern. Furthermore, unlike the regime that close to the Dirac point, this regime has a much larger internal perpendicular electric field to support the large difference of the carrier density between the bilayer and monolayer graphene. In such large electric fields, the valley splitting energy is larger than the Zeeman energy splitting. So we can see the spacing between the LLs with filling factors of $2N + 1$ and $2N + 2$ in the bilayer graphene is larger than the spacing between $2N$ and $2N + 1$, so the crossing pattern with the LLs in monolayer graphene follows the same behavior.

Also, from fig. 4.7 and fig. 4.9, we find that the LLs with filling factor from -2 to 2 have different slopes with other LLs in the monolayer graphene, and when we zoom in to the first 4 LLs, we find that their slopes have a little deviation already. Those lowest LLs in monolayer graphene have very clear shift when crossing happens, while the high monolayer graphene LLs do not shift at the crossing point. more work will be required in order to fully understand this behavior.

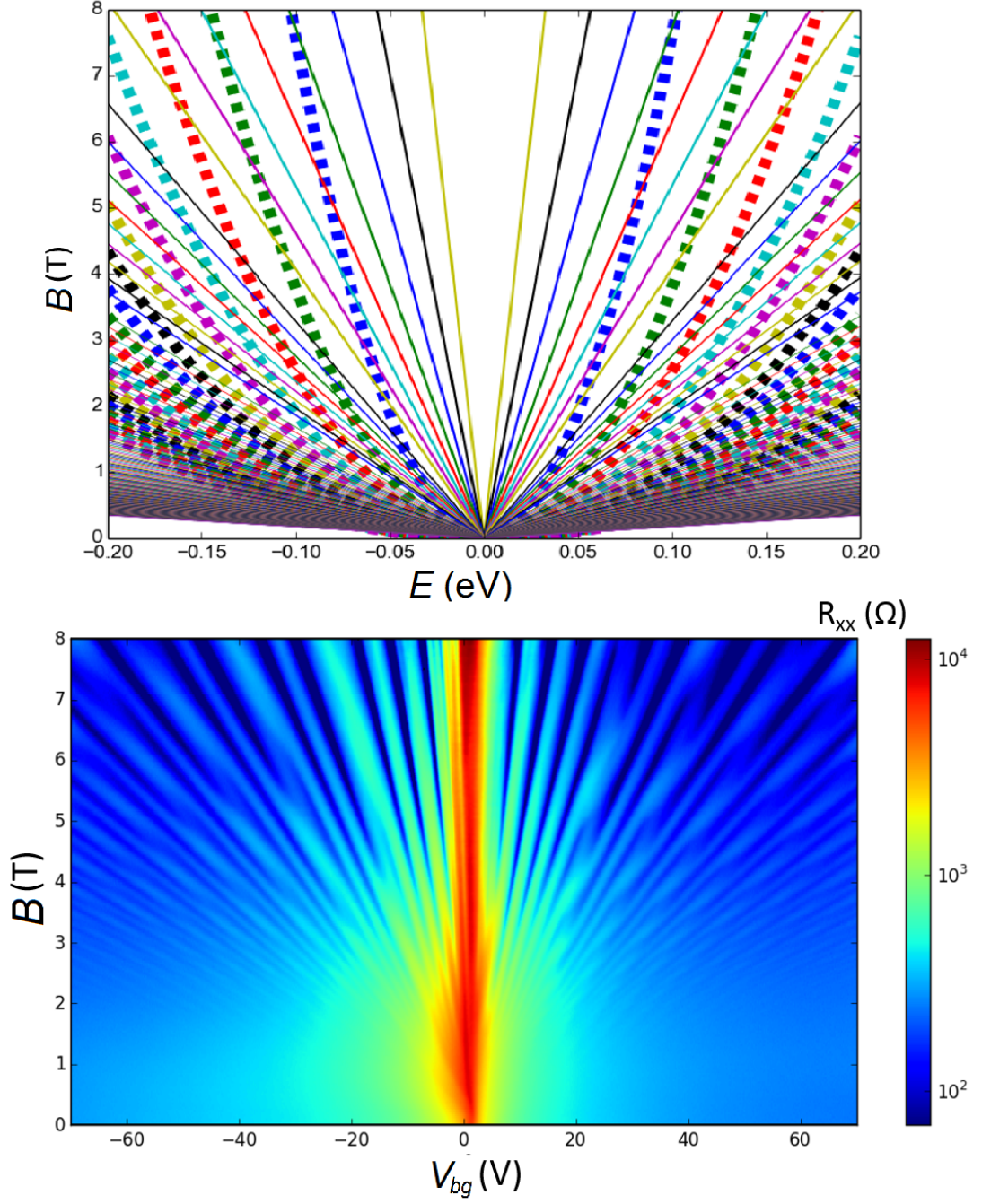


Figure 4.3: Landau level crossing at twisted trilayer graphene. (left) The numerical simulation of the LL crossing of twisted trilayer graphene. The internal electric field is zero. (right) R_{xx} pattern, x axis is back gate and y axis is magnetic field. The lineal lines are the bilayer graphene LLs, and the parabolic lines are the monolayer graphene LLs.

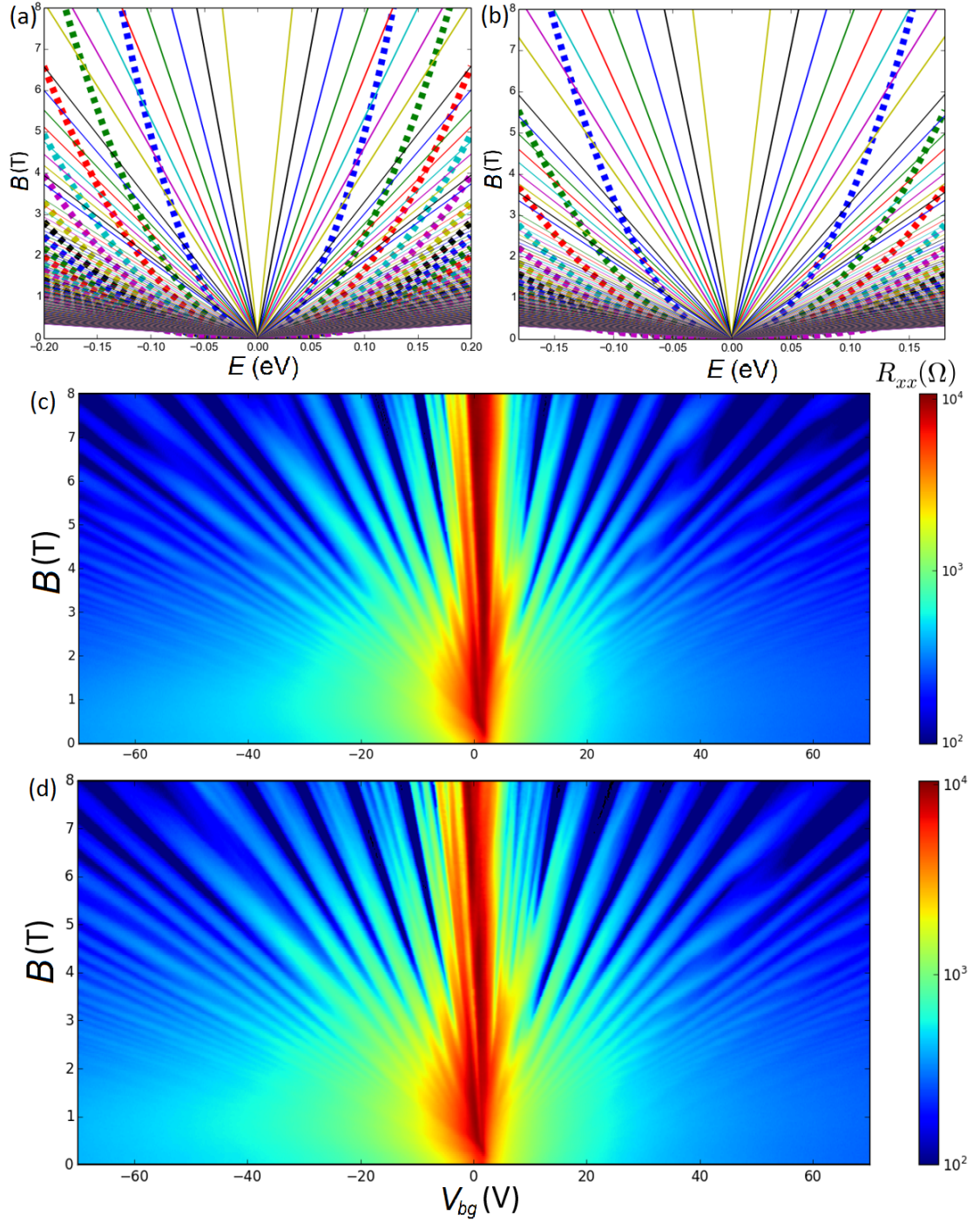


Figure 4.4: Landau Level Crossing at different filling rate for the bilayer and monolayer graphene. (a)(b) The numerical simulation of the LL crossing with the filling rate ratio $rate_{monolayer}/rate_{bilayer} = 0.65$ and 0.45 . (c)(d) R_{xx} pattern when we tune the ratio $\frac{V_{tg}}{V_{bg}} = \frac{1}{2} \times m_2$, and $\frac{1}{4} \times m_2$ where m_2 is the slope that the internal perpendicular electric field is zero. Here x axis is back gate and y axis is magnetic field. The linear lines are the bilayer graphene LLs, and the parabolic lines are the monolayer graphene LLs.

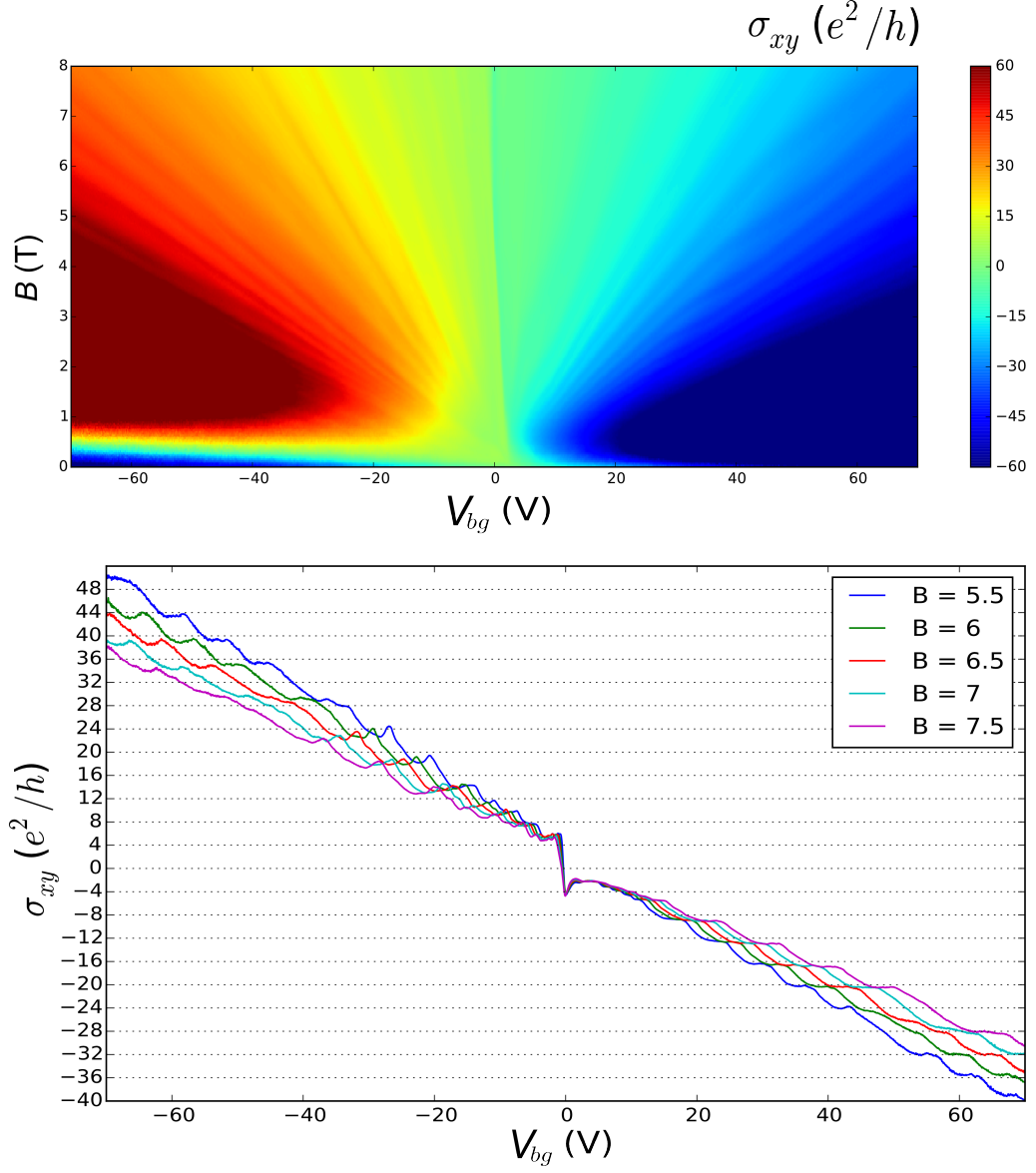


Figure 4.5: (up) σ_{xy} in device 1. We fix the filling rate ratio the same as figure 4.4(d). (down) The line trace of σ_{xy} at different magnetic fields. We can see that the interval of the plateaus doubles when LLs from the bilayer graphene cross those from monolayer graphene.

LLs Crossing with narrow and broad energy levels

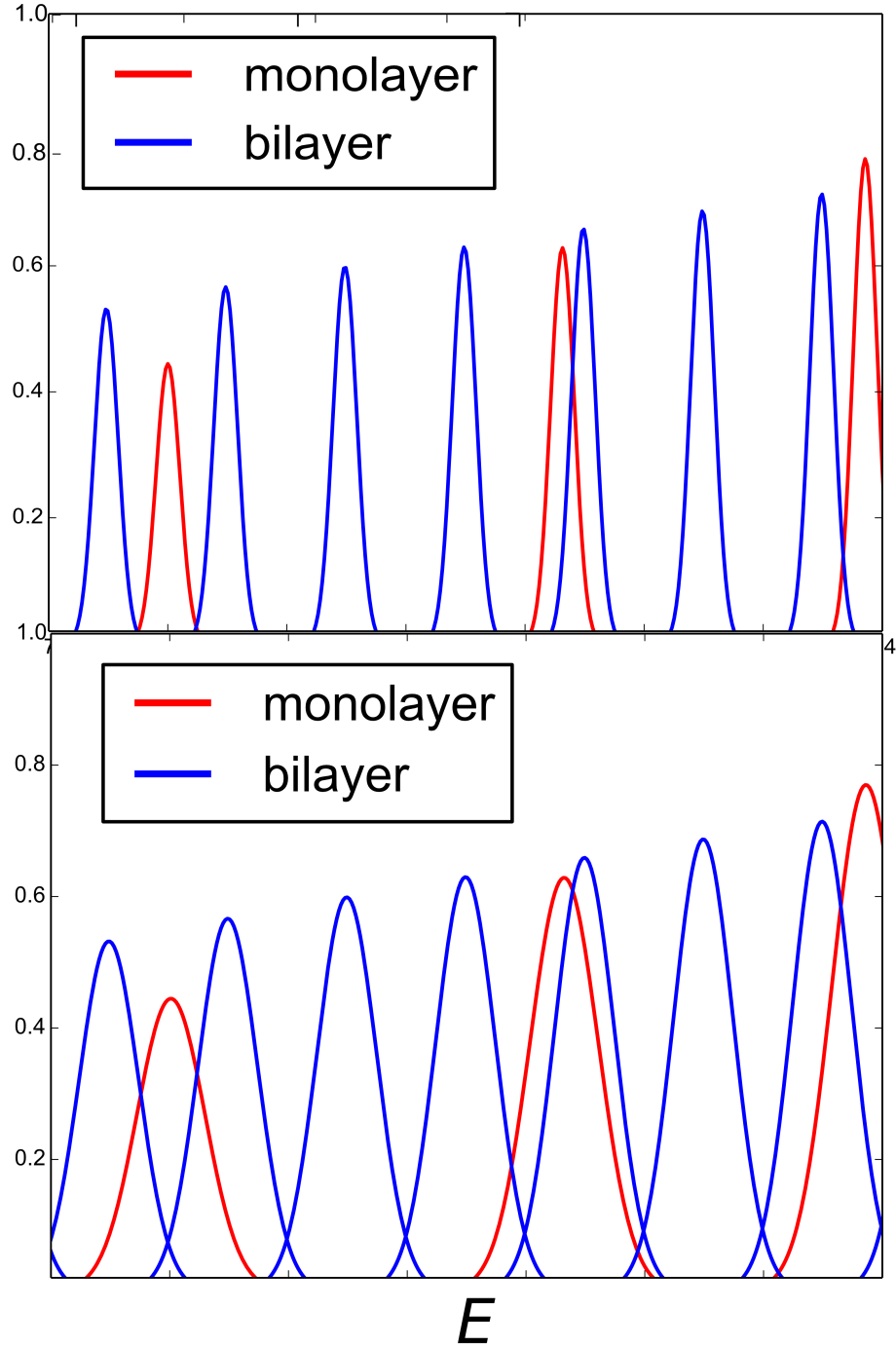


Figure 4.6: LL crossing with narrow energy levels and broadened energy levels. (left) The LLs are not broadened and there is not much overlap between LLs. When LLs from monolayer graphene cross those from the bilayer, the overlap only happens between one energy level from mono- and bilayer graphene. (right) The LLs broaden and the overlap between LLs is much larger. When LL crossing happens, one LL from the monolayer graphene will overlap with two LLs from bilayer graphene.

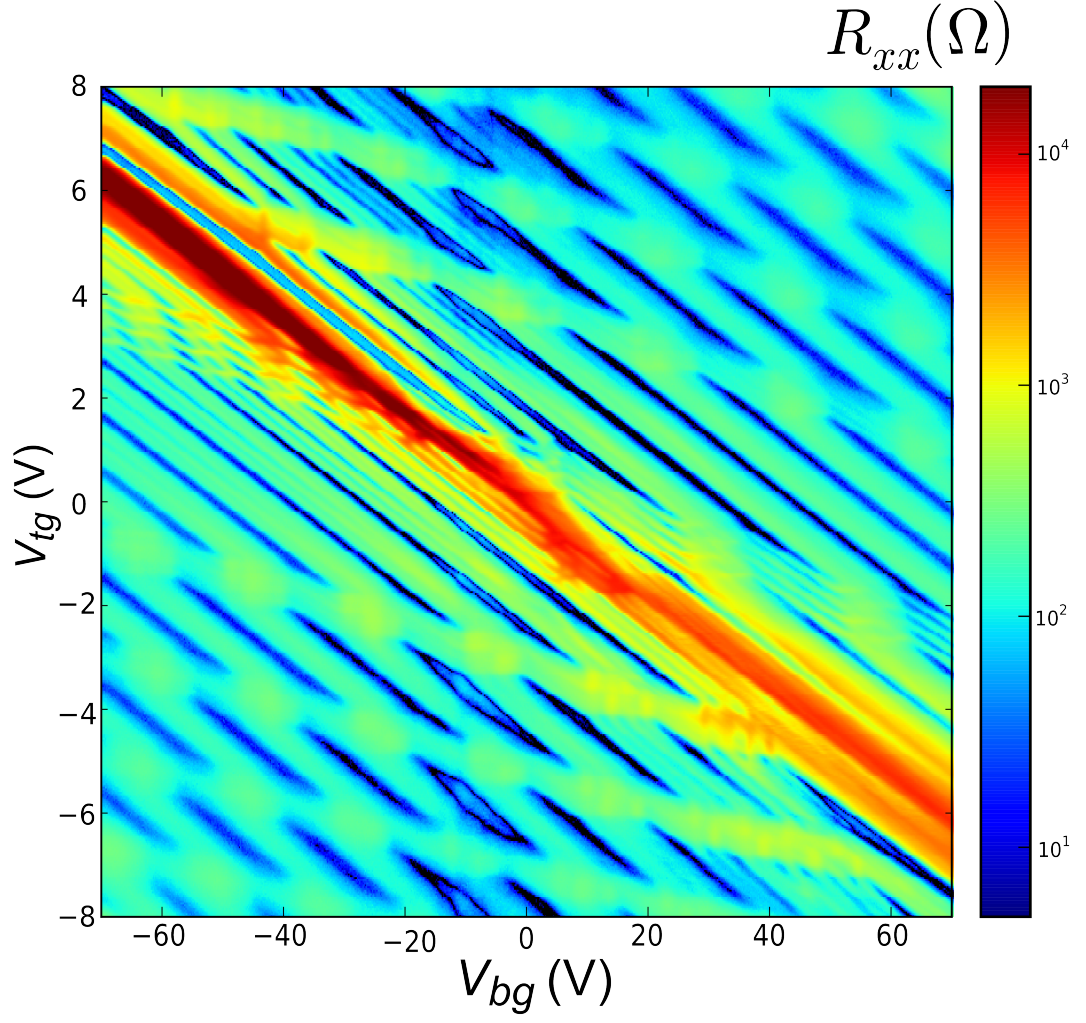


Figure 4.7: Landau Level Crossing at $B = 8$ T in twisted trilayer graphene. LLs from monolayer graphene have a smaller slope of dV_{tg}/dV_{bg} , and LLs from bilayer graphene have larger slope of dV_{tg}/dV_{bg} .

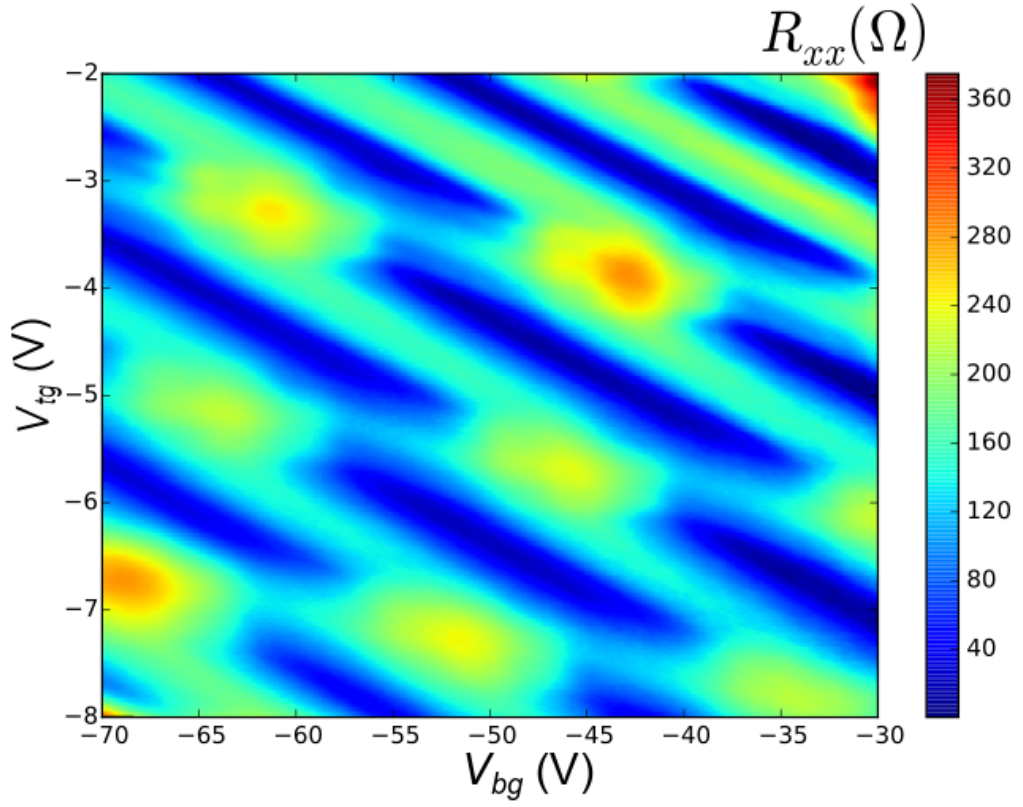


Figure 4.8: Landau Level Crossing at $B = 8$ T in the twisted trilayer graphene in the case of large carrier density and small electric field. The LL crossing lead to the shift of LLs from the bilayer graphene, but no shift of LLs from the monolayer graphene.

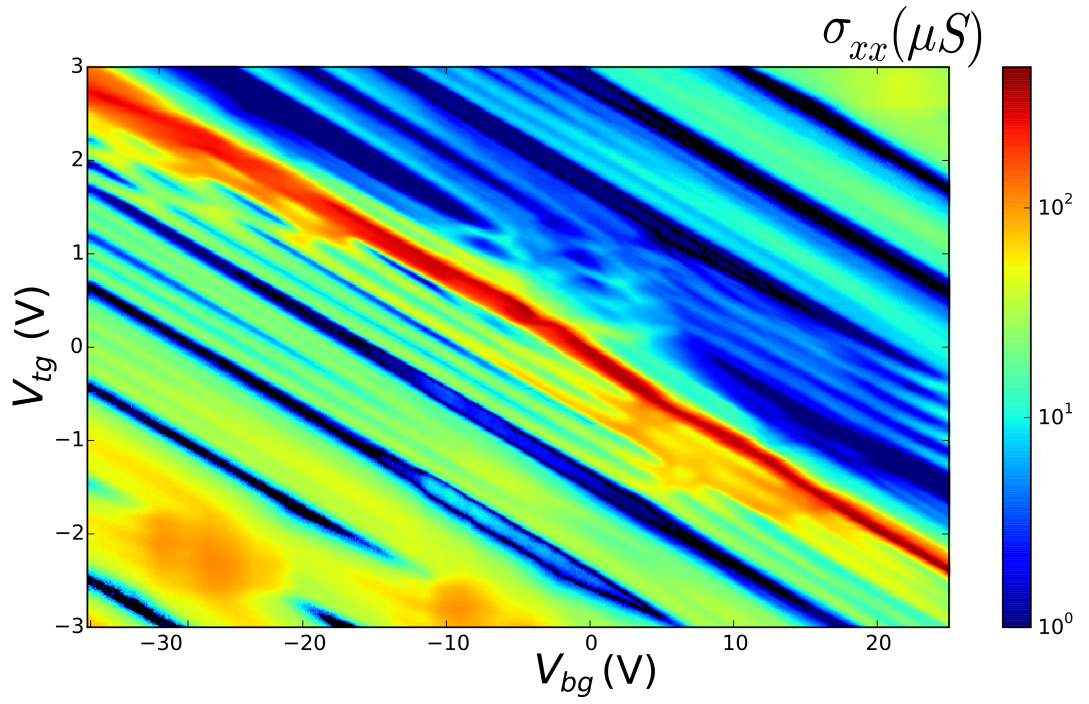


Figure 4.9: Landau Level Crossing at $B = 8$ T in twisted trilayer graphene near the global Dirac point. In both the monolayer and bilayer graphene, the spin and valley degeneracy are broken for the lowest LLs are lifted.

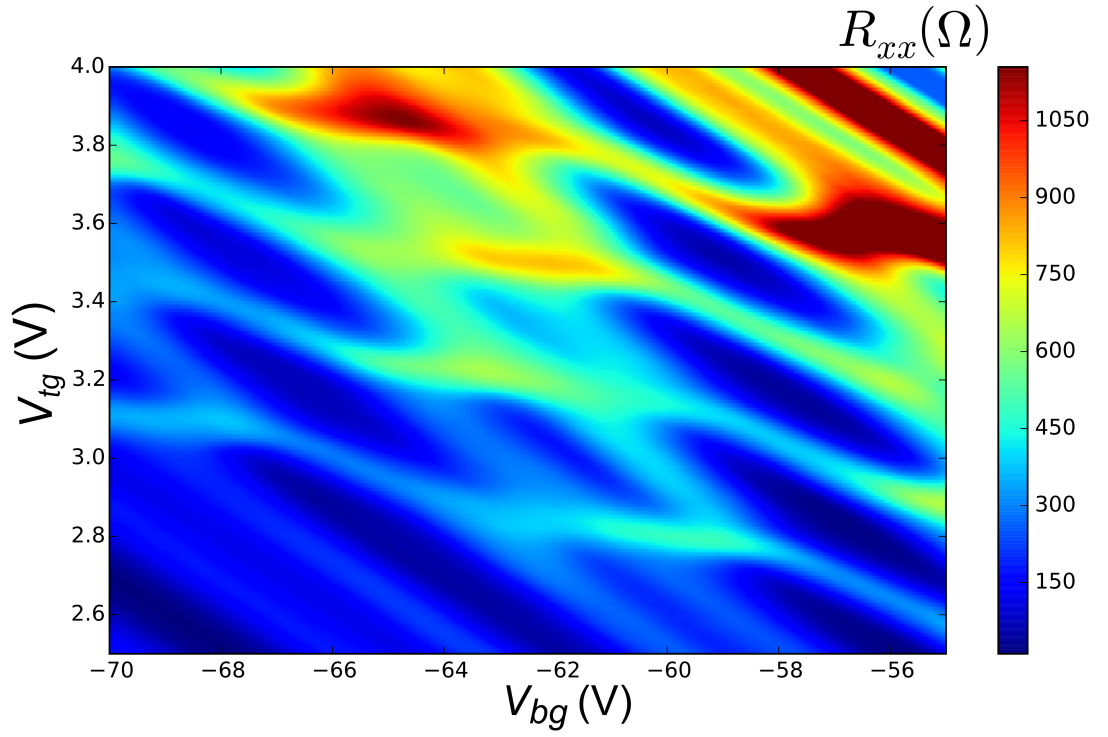


Figure 4.10: Landau level crossing at $B = 8$ T in twisted trilayer graphene when we focus on the regime that the first 4 LLs of Monolayer graphene cross with the LLs with filling factor from 4 to 12 in the bilayer graphene. The spin and valley degeneracy are all lifted, and each energy level only has one spin and one valley.

Chapter 5

Conclusions

In this thesis I reported our transport measurements of few-layer graphene on boron nitride, including bilayer graphene superlattices and twisted trilayer graphene. In the bilayer graphene superlattice samples, we observed secondary and tertiary resistance peaks, and the band structure near the new generation of Dirac points is related to the perpendicular electric field. When the magnetic field turned on, the Hofstadter's butterfly shows electric field dependence as well, and the valley symmetry breaks by the asymmetric coupling and can restore by interlayer electric field. In the twisted trilayer graphene device, we can see controllable Landau level crossing, and the LLs from the bilayer graphene are shifted after the LL crossings. Some new conducting mode also show up in the gaps between the LLs.

Besides the two projects I presented in this thesis, I also worked on a number of other projects, such as graphene/hBN/graphene Coulomb drag, valley current in gapped bilayer graphene, and point contact devices. In the graphene/hBN heterostructure, because of the very high mobility and robustness of device, a lot of experiments can be achieve. For instance, some groups are trying to detect the negative diffraction at the

PN junction, which requires very high mobility and very sharp edges of the junction and can only be achieved recently in graphene/hBN heterostructures.

Many promising directions for future work exist. For example, one question is how to realize the measurement setup to probe the hydrodynamic properties of the electron system in graphene. The key points include how to create shear current and how to measure the current decay on the distance. Another possible thing we can do is coulomb drag between bilayer graphene and monolayer graphene. Another idea is to measure how the valley current goes through a PN junction. Since when the electric field changes its direction at the PN junction, the Berry curvature changes sign at both conducting band and valence band, then it is of interest to determine what will happen when the valley current goes through such PN junction.

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