

UNIAXIAL HETEROSTRAIN IN TWISTED BILAYER
GRAPHENE ABOVE THE MAGIC ANGLE

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Abstract

Twisted bilayer graphene is an experimentally rich platform, hosting a variety of exotic states of matter with an unprecedented degree of tunability. I present measurements of a twisted bilayer graphene device twisted to 1.4 degrees, well above the magic angle at which correlations dominate. Despite the lack of correlated states, the device shows several unexpected behaviors in magnetotransport: regions in density of large quadratic magnetoresistance concurrent with an apparently non-Diophantine Landau fan diagram. I show that uniaxial heterostrain of only a few tenths of a percent neatly explains these unusual magnetotransport measurements. It is presently difficult to control such small amounts of strain in experiments. If we can control it, then heterostrain becomes a powerful tuning knob to control the band structure of twisted bilayer graphene, much like twist angle.

Acknowledgments

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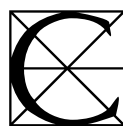
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Chapter 1

Introduction



CONSIDER graphene: a single layer of graphite. It is a planar, honeycomb array of carbon atoms, each atom tightly bound to three neighboring atoms. What is its electrical resistivity, ρ ? How does its resistivity vary with temperature and other relevant details?

You might not expect these to be difficult questions to answer. After all, topics at the frontier of “fundamental” physics—dark energy, quantum gravity, those sorts of things—are irrelevant. We only need to concern ourselves with electrons and carbon nuclei. We can ignore gravity and nuclear forces, leaving only electromagnetism. Surely then, it should be straightforward to compute ρ ?

Well, no. Not only are the equations for electrons in graphene impossible to solve analytically, but they are impossible to compute using a classical computer smaller than the universe. This is a quantum many-body problem: the computational complexity grows exponentially with the number of particles. Building up from the standard model of particle physics is not the way.¹

This wouldn’t be such a bother if graphene were useless and boring. If we could approximate ρ reasonably well using simple models, or if it always behaved as a simple metal, like copper, then it would not warrant the scale and intensity of research that it receives.

¹See P. W. Anderson’s famous “More is Different” article [1] for a discussion of reductionism in condensed matter physics. Although the reductionist viewpoint may be accurate in principle, it is often practically useless.

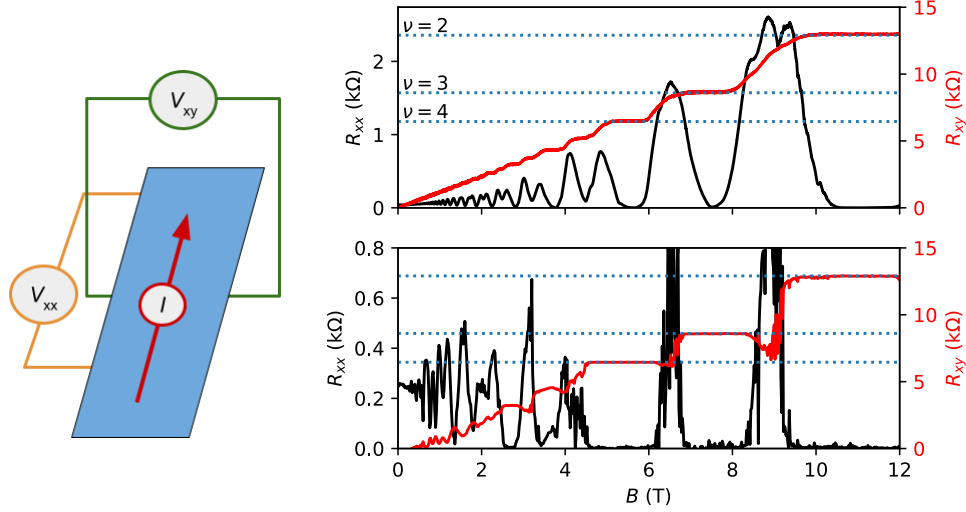


Figure 1.1: **The quantum Hall effect.** **Left:** Hall measurement schematic. The sample is the blue parallelogram, through which flows a current I . The voltage drop along the direction of current (orange) gives the longitudinal resistance $R_{xx} = V_{xx}/I$. In a magnetic field, there will also be a voltage drop transverse to the current (green), giving the transverse (also called Hall) resistance $R_{xy} = V_{xy}/I$. **Right:** Longitudinal and Hall resistances for an InAs quantum well (**top**, 1.5 K) and twisted bilayer graphene (**bottom**, 26 mK) as a function of out-of-plane magnetic field strength B .

Fortunately (or unfortunately, depending on your perspective), graphene and graphene-based materials are not boring. They are instead an excellent platform for studying many of the most tantalizing phenomena in physics: exotic states of matter like superconductivity, among others.

Contrary to what you might expect, in these exotic states, properties like ρ are sometimes simple to describe. For instance, the resistivities of mercury and lead at room temperature differ by a factor of five. When cooled into their superconducting states, however, their resistivities are both equal to precisely zero. Much simpler.

The quantum Hall effect, which concerns both longitudinal and transverse resistances as a function of magnetic field (Fig. 1.1) also has a simple description. The longitudinal resistance is zero, as in a superconductor. However, the transverse resistance is $h/\nu e^2$, where $h/e^2 \approx 26 \text{ k}\Omega$ is a physical constant and ν is either an integer (the integer quantum Hall effect) or, in extremely clean systems, a rational number

like $1/3$ or $2/5$ (the fractional quantum Hall effect).

As with superconductivity, these numbers do not depend on material parameters. The right panels of Fig. 1.1 show measurements of two completely different materials at different temperatures. The two measurements look distinct, except in certain regimes where the longitudinal resistances (black lines) are zero and the transverse resistances (red lines) plateau at particular absolute values given by the formula in the previous paragraph. Above 10 Tesla, for instance, both systems have precisely zero longitudinal resistance and precisely $h/2e^2 = 12906 \, \Omega$. These values are so precise and repeatable that for metrological purposes the quantum Hall effect defines the resistance unit, the ohm.

Although the properties of materials in these exotic states can sometimes be described easily, many of them remain difficult to understand and predict. And not for a lack of desire and effort, either. If we understood superconductivity more thoroughly, for example, we might be able to predict materials that superconduct at ambient temperature and pressure. The technological repercussions would be massive: think efficient power grids, cooler (and thus more powerful) computers, levitating trains, those sorts of things.

Therefore, we seek to study materials that 1. are practical to study both experimentally and theoretically, and 2. display exotic many-body physics that we do not understand. Graphene excels at both.²

In this work, I study a material called twisted bilayer graphene (TBG) consisting of two layers of graphene, one placed directly on top of the other with a small relative twist. The relative twist gives rise to a large moiré pattern as the carbon atoms go into and out of registry with each other (Fig. 1.2).

When the twist angle is within a tenth of a degree of the “magic” angle of 1.1° , TBG can display superconductivity, ferromagnetism, correlated (“Mott”) insulating states, fractional, fractal, and anomalous quantum Hall states, and more. Some of these states are reasonably well understood, but others are not.

²Actually, a single layer of graphene quite stubbornly refuses to display many-body states, with very few exceptions. Most of the many-body experimental results in graphene occur in few-layer structures like bilayer graphene, twisted bilayer graphene, rhombohedral trilayer graphene, and so on.

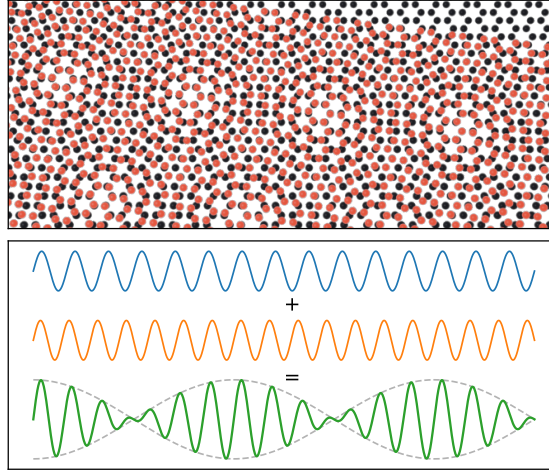


Figure 1.2: **Moiré patterns.** **Top:** Two honeycomb lattices, one rotated by 8° . **Bottom:** Two sin-waves with frequencies f_1 and f_2 add to give a “beating” pattern with frequency $|f_1 - f_2|$. If the x -axis were time, this could describe a poorly-tuned guitar played on top of a tuner. Moiré patterns are the 2D spatial analogue of this beating pattern.

Unfortunately, there is an element of randomness to each sample. I may try to make five samples with identical twist angles, but when I measure them, the twist angles might all be different. Worse still, samples with the same measured twist angle might show distinct exotic behavior. This is a problem. To use TBG to study delicate, exotic physics, I need to carefully control each aspect of the material. This randomness precludes such careful control.

Such was the state of the world when I fabricated one particular TBG device. The device was twisted far enough away from the magic angle that it (mostly) did not show the exotic physics for which TBG is famous. However, it still showed striking and puzzling behavior when immersed in a magnetic field. I was able to demonstrate some of the puzzling behavior using a simple model, and one of the key ingredients to the model was anisotropy—I suspected this anisotropy was caused by strain in the underlying graphene.

Working closely with two excellent theorists Oskar Vafek and Xiaoyu Wang, we were able to show that indeed strain was an excellent explanation for the behavior. Strikingly, only miniscule amounts of strain, a few tenths of a percent, were required to

have major impacts on the electronic properties of the material. This is an unintuitive result. Most of the time we think of 0.1% strain as doing 0.1% of something. Early reports on TBG did not even consider strain. However, in the case of twisted moiré materials like TBG, the effects of *hetero*-strain, strain that differs between the two layers, is magnified by a factor of $1/\theta$, where θ is the twist angle in radians.

We found that uniaxial heterostrain—shrinking or stretching in one direction—has major qualitative effects on the properties of TBG. It dramatically distorts the shape of the moiré lattice, breaking its perfect triangular symmetry. In response, the electronic properties dramatically change. Away from the magic angle, these effects are possible to calculate with textbook methods. At magic angle, where exotic, poorly-understood physics dominates, it is not so easy.

So, to controllably study exotic physics using TBG, the surprisingly large effects of heterostrain must be considered. This may be considered a pessimistic result, because such small amounts of strain are hard to control in experiment. On the other hand, if we can control it, heterostrain is a powerful tuning knob, yielding dramatic electronic effects from small distortions to one layer.

In reaching these conclusions, we will encounter a number of aesthetic and satisfying ideas in all of physics, including moiré patterns, Hofstadter’s butterfly, superconductivity, the fractional quantum Hall effect, and more. I hope that I can convey to you even a small part of the joy that I experienced while researching this fascinating material.

1.1 The quantum Hall effect

Consider a gas of electrons, free to move in two dimensions but tightly confined in the third. Classically, most interesting properties of the system require three spatial degrees of freedom to describe. No matter how tight the confining potential is, there can still be miniscule motion in the third direction. In quantum mechanics, this is not the case. The electronic wavefunction separates into

$$\Psi(x, y, z) = \psi(x, y)Z_n(z), \quad (1.1)$$

for $Z_n(z)$ solving the time-independent Schrödinger equation

$$\left[-\frac{\hbar^2}{2m_z} \frac{\partial^2}{\partial z^2} + V(z) \right] Z_n(z) = \epsilon_n Z_n(z) \quad (1.2)$$

with effective mass m_z and confining potential $V(z)$. The confining potential must be sufficiently tight such that the spacing between its ground state energy ϵ_0 and its first excited state energy ϵ_1 is larger enough than $k_B T$ for there to be negligible population of the first excited state. We say that the third dimension is “frozen out,” leaving us with a two-dimensional electron gas. This “2DEG” can host exotic states of matter that are inaccessible in three dimensional systems.

The simplest realization of a 2DEG in solid-state materials³ is at the inversion layer that can form between a semiconductor and an insulator, as in gated Si/SiO₂ [3]. This is the system out of which we construct the MOSFETs that power our computers and thus all of modern society. Far more importantly, it is the system in which the quantum Hall effect was first observed.

It is also possible to create a 2DEG in a structure known as a “quantum well,” by sandwiching two semiconductors with different band gaps, as with GaAs (1.5 eV) and AlAs (2.2 eV). Such systems can be grown with extremely low disorder, leading to carrier mobilities in excess of 10^7 cm²/Vs [4].

The Hamiltonian for electrons in the 2DEG in the presence of a magnetic field is given by

$$\hat{H} = \frac{1}{2m^*} \left(\vec{p} + e\vec{A} \right)^2 \quad (1.3)$$

for 2D effective mass m^* ($\sim 0.067 m_e$ for GaAs [4], m_e is the free electron mass) and positive electric charge e . The quantum Hall effect occurs when the magnetic field is oriented normal to the 2DEG: $\nabla \times \vec{A} = B\hat{z}$. We have freedom to choose a gauge with which to perform our calculation. There are two natural choices: the Landau gauge $\vec{A} = B(0, x, 0)$ and the symmetric gauge $\vec{A} = (B/2)(-y, x, 0)$. I will proceed with the

³Curiously, one of the first-discovered 2DEGs is at the surface of liquid helium [2].

former. The Hamiltonian is then given by

$$\hat{H} = \frac{\hat{p}_x^2}{2m^*} + \frac{1}{2m^*} (\hat{p}_y + eB\hat{x})^2 = \frac{\hat{p}_x^2}{2m^*} + \frac{1}{2}m^*\omega_c^2 \left(\hat{x} - \frac{\hbar k_y}{m^*\omega_c} \right)^2 \quad (1.4)$$

where $\omega_c = eB/m^*$ is the cyclotron frequency. In the last step, I replaced $\hat{p}_y$ with its eigenvalue $\hbar k_y$, because of the lack of $\hat{y}$ -dependence in the Hamiltonian.

This is precisely the equation for the quantum harmonic oscillator, except shifted in x . This shift does not affect the energies, leaving us with

$$E_n = \hbar\omega_c \left(n + \frac{1}{2} \right) \quad (1.5)$$

for $n \geq 0$. The wavefunctions at a given n are called the n -th Landau level (LL). Each LL is macroscopically degenerate with degeneracy Φ/Φ_0 , neglecting spin degeneracy, where $\Phi = BA$ is the magnetic flux through the entire sample and $\Phi_0 = h/e$ is the magnetic flux quantum.

When the Fermi level of the 2DEG is tuned into a gap between Landau levels,⁴ the Hall resistance takes on quantized values

$$R_H = \frac{V_H}{I} = \frac{h}{\nu e^2} \quad (1.6)$$

for integer ν . It is worth remembering the “von Klitzing” constant $R_K = h/e^2 = 25812.8 \dots \Omega$. At the same time as the Hall resistance is quantized, the longitudinal resistance becomes precisely zero. See Ref. [7] for a concise and beautiful argument for this behavior based on gauge invariance and the existence of a mobility gap between Landau levels.

With all this in mind, imagine setting the magnetic field to some nonzero value. As you then tune density, you will see evenly spaced features associated with tuning through Landau levels, evenly spaced because each LL has the same degeneracy. Within each LL, transport will be dissipative, but within gaps transport is dominated

⁴To actually tune the Fermi energy to within the gap requires there to be localized states within the gap. This is a curious thing: to see the quantum Hall effect, you need at least some disorder! See Refs. [5–7].

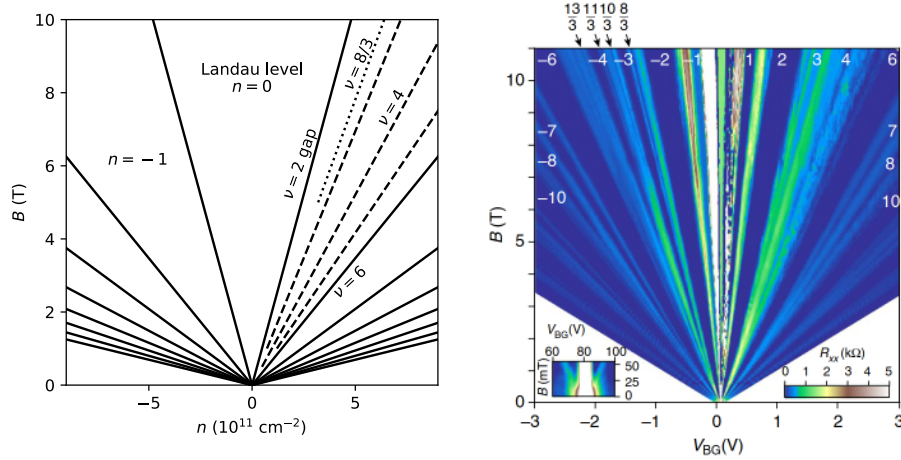


Figure 1.3: **Landau fan diagrams. Left:** Schematic, fictional Landau fan diagram. The $n = 0$ and -1 Landau levels are labeled. Gaps are labeled by ν . The main progression is the same as in graphene: $\nu = 4(i + 1/2)$ for integers i (solid lines). Above a few Tesla, the $n = 1$ LL degeneracy has broken, and we see $\nu = 3, 4$, and 5 gaps (dashed lines). Above 5 T, a fraction appears at $8/3$ (dotted line). **Right:** Landau fan diagram in graphene from Ref. [8].

by the quantum Hall effect. The spacing of these Landau levels increases linearly in field, leading to a fan diagram if resistivity is plotted against both field and density.⁵ If there is some underlying degeneracy in the 2DEG, for example from spin, then this shows up as missing lines in the fan. Instead of every integer ν , we may see only even integers, for instance. Thus, the quantum Hall effect surfaces underlying details that may be otherwise hard to ascertain. Fig. 1.3 demonstrates this type of measurement for graphene.

In some extremely clean 2DEGs, the Landau levels can further separate into rational fractional states $\nu = p/q$ for coprime p and q : the fractional quantum Hall effect (FQHE). Typically, only odd-denominator fractions are seen, such as $1/3$ or $2/5$. These are relatively well-understood in terms of Laughlin wavefunctions and composite fermions [9]. Even-denominator fractions have been observed going back to 1987 in GaAs [10] and more recently in graphene [11]. The FQHE is fundamentally a many-body effect and some of its details, notably the nature of the $\nu = 5/2$ state,

⁵It is useful to remember the slope of the lines $e/h \approx 2.4 \times 10^{10} \text{ cm}^{-2}/\text{T}$.

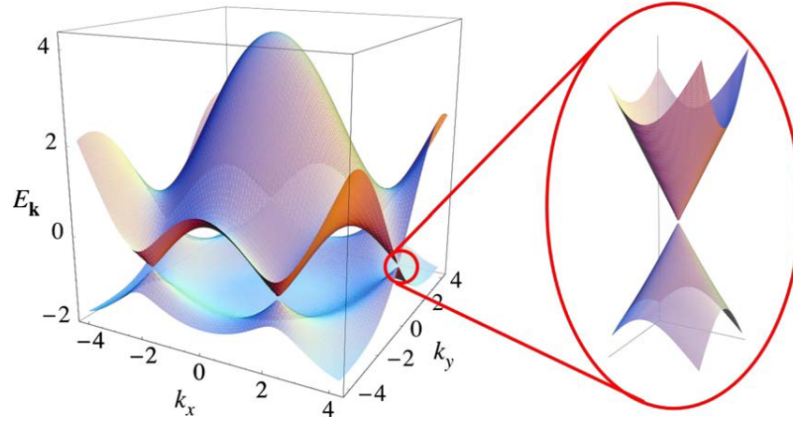


Figure 1.4: **Band structure of graphene.** From Ref. [12]. Energy is in units of 2.7 eV, wavenumbers are in units of $1/a$.

remain contentious to this day.

1.2 Graphene and graphene moirés

Graphene is a single layer of graphite. It is a honeycomb lattice of carbon atoms, each with three neighbors. Three of its four valence electrons form strong sigma bonds with these neighbors, and the fourth is free to roam about the lattice. Each unit cell contains two carbon atoms, leading to a factor of four degeneracy of its bands (two from sublattice (or “valley” in k -space) and two from spin).

Although the basic electronic properties of graphene were known since antiquity,⁶ it was not until 2004 that a technique was devised to isolate large area monolayer flakes for detailed study [15]. The graphene flakes isolated in this way had remarkable properties [12, 16]. They were strong, “the strongest material ever measured” [17], which meant they could be transferred from one substrate to another without tearing, allowing researchers to stack them like Lego blocks into “designer heterostructures” with other 2D van der Waals (vdW) materials [18]. The graphene flakes had extremely low intrinsic defect densities and exceptionally high electronic mobility, especially

⁶Or at least since 1947 [13]. See [14] for a history of graphene from 1859 until shortly after 2004.

when sandwiched between insulating hexagonal boron nitride (hBN) layers [19]. This allowed for the measurement of delicate quantum states that only show up in the cleanest electronic systems [8, 11, 20, 21].

Near charge neutrality (Fig. 1.4 right panel), graphene's electrons have linear dispersion: they are analogous to massless Dirac Fermions, with a Fermi velocity in the ballpark of $v_F \sim 10^6$ m/s. Graphene's Hamiltonian is concisely written as

$$\hat{H} = \hbar v_F \vec{\sigma} \cdot \vec{k} \quad (1.7)$$

where Pauli matrices $\vec{\sigma}$ act on sublattice, rather than the usual spin. Owing to its linear dispersion, in a magnetic field, its Landau levels have energies not equally spaced, but instead given by [12]

$$E_n = \text{sgn}(n) \hbar \omega_c \sqrt{n}, \quad (1.8)$$

where $\omega_c = \sqrt{2} v_F / l_B$. l_B , the magnetic length, equals $\sqrt{\hbar / eB}$.⁷ It has a Landau level at zero energy, so, taking into account its fourfold degeneracy, we see gaps with $\nu = \pm 2, \pm 6, \pm 10, \dots$ in transport measurements. Above a few Tesla, Coulomb interactions lead to complete breaking of LL degeneracy and all integers are observed [23]. Curiously, the half-filled zeroth LL (CNP) becomes strongly insulating in field (See [24] and references within [23]).

Graphene superlattices and Hofstadter's butterfly

Because hBN and graphene are both honeycomb lattices whose lattice constants differ by only around 2%, a moiré superlattice appears when they are stacked one on top of the other and perfectly aligned. This superlattice's wavelength λ reaches up to 14 nm, significantly larger than the underlying graphene and hBN lattices. It is given

⁷Note that the spacing of Landau levels in graphene is large compared to in most materials. It is possible, in the worlds most powerful static magnetic field (45 T), to observe the quantum Hall effect in graphene even at room temperature [22]!

by [25]

$$\lambda = \frac{(1 + \delta)a}{\sqrt{2(1 + \delta)(1 - \cos \theta) + \delta^2}}, \quad (1.9)$$

where δ is the lattice mismatch, $a \approx 0.246$ nm is the lattice constant of graphene, and θ is the twist angle between the two layers.

This hBN superlattice allowed for the first complete mapping of Hofstadter's butterfly [26, 27]. Hofstadter's butterfly is a rather odd phenomenon in physics. It is the extension of the quantum Hall effect, which concerns free electrons, to the case of Bloch electrons. The resulting fractal spectrum is both physically and mathematically interesting.

I give a more complete derivation later on in Chapter 4 where it becomes necessary. The main results are (see Chapter 4, Figs. 4.3 and 4.4)

1. When the magnetic flux through a unit cell per flux quantum $\Phi/\Phi_0 = eBA/h$ (A here is a unit cell, not the whole sample) is a rational number p/q , the Bloch band splits into q magnetosubbands. The plot of the spectrum has a fractal butterfly-like appearance [28]. Alternatively, each Landau level splits into p sublevels [29].
2. In density vs field, the gaps follow the Diophantine relation $n = t\Phi/\Phi_0 + s$ for integers s and t [30]. This can be seen as an extension of the integer quantum Hall effect, wherein $s = 0$ and $t = \nu$. The Hall conductivity within the gaps is given by te^2/h [31].

To see the butterfly unambiguously requires the ability to tune both density through the Bloch band and magnetic field such that $\Phi/\Phi_0 \sim 1$. For ordinary crystals, this condition is impossible to achieve with magnetic fields available in laboratories,⁸ but possible given the large unit cell of graphene superlattices [26, 27].

Twisted bilayer graphene

In twisted bilayer graphene (TBG), a slight interlayer rotation creates a similar-scale moiré pattern (Fig. 1.5, top-left panel). In k -space, the Dirac cones from each layer

⁸If $A = (1 \text{ nm})^2$, then $\Phi/\Phi_0 = 1$ at over 4000 T.

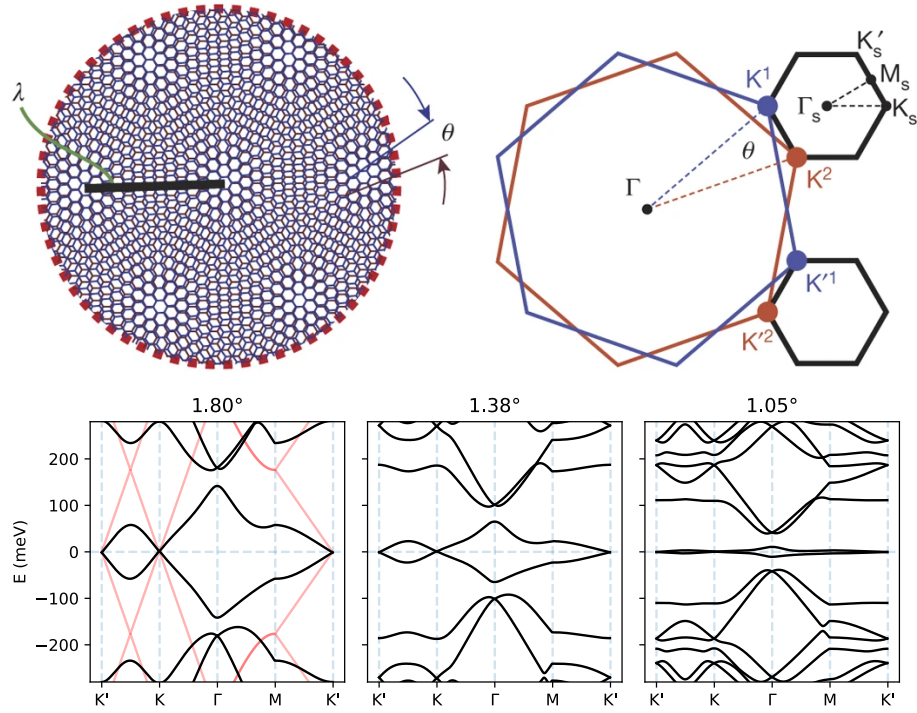


Figure 1.5: **Twisted bilayer graphene.** Top row from Ref. [32]. **Top-left:** Real-space moiré for twisted bilayer graphene. **Top-right:** The moiré Brillouin zone is formed at the corner of the larger graphene Brillouin zones. **Bottom row:** Computed band structures for the Bistritzer-MacDonald model described in Chapter 5 without heterostrain, only plotting one valley. The red curves in the left panel have zero interlayer tunneling.

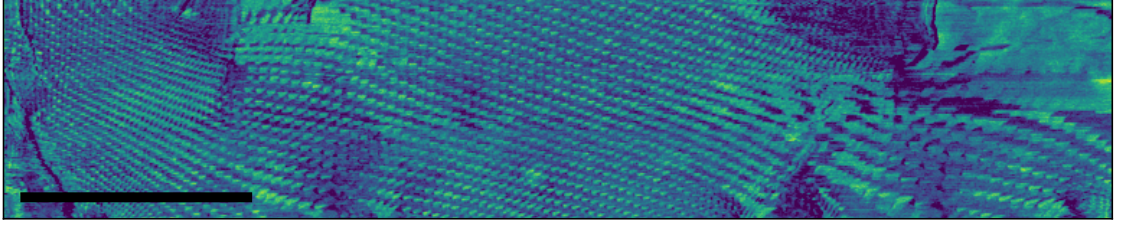


Figure 1.6: **Inhomogeneous TBG.** Torsion force microscopy image [52] of a twisted bilayer graphene sample with a low and inhomogeneous twist angle of 0.1 to 0.3° . Scale bar $1\ \mu\text{m}$.

are extremely close to each other for small twist angles (top-right panel). Interlayer tunneling causes these two Dirac cones to hybridize (bottom panels).⁹ Unlike with graphene-hBN moirés, in TBG there is a gap between lowest and neighboring moiré subbands, leading to insulating behavior at full filling of the moiré bands [34].

As the twist angle approaches the magic angle of 1.1° the isolated moiré bands become flat [35, 36], and strong correlations lead to fascinating insulating [32, 37–48], (potentially unconventional) superconducting [37–39, 41–44, 46–49], strange metal [38, 44, 45, 48], and magnetic [40, 50] states. There is a tangible hope that the presence of so many exotic states in a novel and easily tuned system¹⁰ will lead to real progress in many-body physics. Of particular interest are the phenomenological similarities between TBG’s superconducting phase diagram and that of the high- T_C superconductors [51].

TBG was originally fabricated by tearing one sheet of graphene in half, yielding two graphene flakes with known relative orientation [32, 34, 37]. This tear step was

⁹It is a curious thing that band gaps open when hybridization is added. Why don’t they close? This question is fundamental to solid-state physics, but it is surprisingly complicated to answer. The problem can be stated as such: how do the eigenvalues of an $n \times n$ matrix behave under small perturbations? The answer is that for Hermitian matrices, nearby eigenvalues tend to repel each other, but for general matrices they can attract or repel [33].

¹⁰Compare to the cuprates. In TBG, we can tune between correlated insulator, superconductor, and so on by tuning density with a gate. To completely populate or depopulate the bands near magic angle requires a shockingly small absolute density on the order of one electron per ten thousand carbon atoms. In cuprates, the equivalent process requires growing and measuring multiple samples with different doping levels: the density cannot be tuned so easily *in situ*.

unreliable, and also thought to impart significant strain and inhomogeneity in the resulting devices. Measurements at multiple locations on given sample would often find dramatically different twist angles. Local probe measurements corroborated these inhomogeneity measurements in two ways (Refs. [53–56] and Fig. 1.6): 1. the twist angle varied on the order of 0.1 to 0.2° over a few hundred nanometers, and 2. there was both uniaxial and biaxial heterostrain (heterostrain is the difference of strain between the two layers) of about 0.1% to 0.5% , also varying on a scale of a few hundred nanometers.

This inhomogeneity and strain is expected to some extent in TBG, because TBG is an energetically unstable configuration. Bilayer graphene wants to relax to Bernal stacking, or at least close to it [57,58]. Early works would even purposefully twist their graphene 0.1 or 0.2° too far, expecting it to relax partway (Ref. [59], for instance). There is no expectation for this relaxation to be uniform and homogeneous. One could imagine that bubbles of trapped contaminants, for instance, could pin the layers locally while allowing distant regions to relax [56]. Later works would employ some variant of “cut-and-stack,” in which the large graphene starting flake is cut in half either with an AFM or with a laser prior to stacking. This seems to reduce the inhomogeneity and strain, but may not entirely remove it [54].

All of this fuss about a little inhomogeneity and strain may seem uninteresting and unimportant. 0.1% strain is a very small amount, after all. Graphene can survive strains near 25% without tearing, and typical studies on the effects of strain in graphene involve strains of over 10% [60,61]. The important conclusion in my dissertation is that 0.1% heterostrain dramatically affects the moiré pattern and leads to large qualitative effects, clearly visible in transport. Some of these effects have been described before in theory [62,63], however, along with expanding the theoretical predictions, I go one step further and provide experimental verification. Miniscule amounts of strain cannot be neglected when studying twisted bilayer graphene.

1.3 Overview

This dissertation is organized mostly chronologically. Throughout, I try to explain my thought processes and hypotheses about the measurements, even when they were wrong. Science is an iterative process of refining one’s beliefs in response to evidence, and I believe it would be a mistake to simply state my final conclusions without including intermediate, wrong, hypotheses.

Rather than bog down this chapter, I introduce general physics calculations as they are needed throughout the text.

In Chapter 2, I describe the process I used to fabricate twisted bilayer graphene devices and characterize their basic properties. I present several examples of failed attempts to reproduce ferromagnetic behavior in TBG [40, 50] by aligning hBN to one layer of graphene. This chapter can be skipped by the uninterested reader.

In Chapter 3, I describe unusual magnetotransport measurements of one TBG device. Most notably, I see large positive magnetotransport in a range of densities on either side of charge neutrality, and a splitting, bending pattern of lines in the Landau fan diagram. I next describe a few (wrong) hypotheses that I tried to use to explain these features. In Chapter 4, I present a toy model based on Hofstadter’s butterfly that reproduces the unusual Landau fan diagram. In fact, splitting, bending Landau fan diagrams are actually a generic feature of Hofstadter’s butterfly models with anisotropy and some form of energetic butterfly doubling, such as from a Zeeman effect. Chapters 3 and 4 form the basis of my first publication in Ref. [64].

In Chapter 5, I present work done in collaboration with Oskar Vafek and Xiaoyu Wang to move towards a realistic model of transport in strained TBG. I first demonstrate the unexpectedly large effects of heterostrain on twisted moiré patterns: the effects of heterostrain are “magnified” by a factor of $1/\theta$. I then describe the continuum model of TBG, and show the dramatic effects of small amounts of uniaxial heterostrain on TBG’s energetics. I feed these results into a Boltzmann transport model and find that the results are in surprisingly good agreement with my experimental data at low magnetic fields: not only the large magnetoresistance but also a few more subtle details that I had failed to notice previously. This chapter forms the

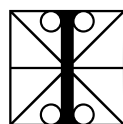
basis of my second publication in Ref. [65].

In Chapter 6, I compare measurements at higher magnetic field values to calculations of Hofstadter's butterfly in strained TBG, again in collaboration with Oskar Vafek and Xiaoyu Wang. And again, the experiment is in good agreement with the theory. Chapters 5 and 6 satisfactorily explain the unusual measurements in Chapter 3 as effects of a small amount of uniaxial heterostrain.

Finally, in Chapter 7, I conclude and discuss prospects for the future.

Chapter 2

Sample fabrication and basic characterization



It is worth describing the sample fabrication and characterization processes before proceeding. First, I thin down bulk crystals of graphite and hBN and deposit them on SiO_2 wafers in a process known as exfoliation. I then search under a microscope for suitable flakes, use a polymer stamp assembly to tear-and-stack the flakes on top of each other, and transfer the stack onto a final substrate. I next use standard nanolithography techniques to add metal gates, etch the stack into a Hall bar geometry, and deposit metal contacts. I screen the resulting devices at room temperature and characterize their basic electronic properties in a cryostat. This chapter describes the process in more detail. Uninterested readers may skip to the next chapter.

Throughout my PhD, I attempted this entire process well over a hundred times. Of these attempts, only a few dozen devices survived to the end of the process and warranted further study. Such is life.

2.1 Exfoliation, searching, stacking, and fab

Exfoliation is the process of thinning out bulk crystals of highly oriented pyrolytic graphite and bulk crystals of hBN by repeatedly peeling them apart with (usually

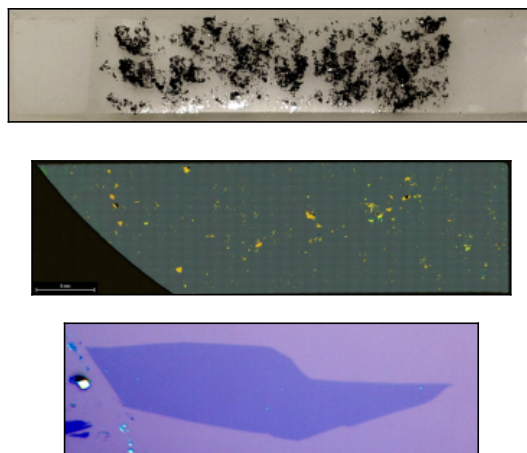


Figure 2.1: **Exfoliation and searching.** **Top:** A tape, several centimeters in length, populated with graphite. **Middle:** A composite image of an SiO_2 wafer populated with graphite. Thick “bulk” flakes at least 100 nm thick, are clearly distinguished by their yellow color. **Bottom:** A graphene flake, several hundred microns wide and several tens of microns tall. The pale blue dots on the flake are tape residue.

Scotch MagicTM-brand) tape [15] (Fig. 2.1 top panel). This exposes atomically-flat and clean facets of the crystal, facets that hopefully have few accumulated adsorbates. After preparing the tape, I press it onto a cleaved wafer of SiO_2 grown on degenerately doped Si. The thickness of the SiO_2 has to be carefully chosen such that any monolayer graphene is easily seen under an optical microscope [66]. To encourage the flakes to adhere to the SiO_2 , I heat the SiO_2 on a hot plate for a few minutes¹ before peeling the tape off slowly (Fig. 2.1 middle panel).

The exfoliation process is inconsistent in terms of flake yield: one chip might have several good flakes while the next might have none. Part of the inconsistency comes from the starting crystal and exfoliation process, however part is simply random variation from small number statistics. Different researchers follow different exfoliation methods. Some use heavy pressure when pressing the tape to the SiO_2 , some use light (I use light). Some swear on oxygen-plasma-cleaned SiO_2 , and some anneal

¹At any temperature between 80 and 110°C, for any amount of time between a few minutes and a few hours. Higher temperatures and longer time yields more flakes, but also significantly more tape residue in between the flakes.

their SiO_2 in forming gas (I do neither). Quite a few groups insist on peeling the tape off of the SiO_2 slowly and at a slight angle (I do not do this). Other groups create a dense “mother” tape and sparse “daughter” tapes from the mother (I do this). It is possible and indeed likely that some of these techniques improve the reliability of the exfoliation process somewhat, however I do not know of convincing (that is: blind, randomized, and/or controlled) evidence for it.

After exfoliating comes the time-consuming, often-frustrating process of searching for flakes of appropriate thickness and quality under a microscope. Most of the exfoliated flakes are thick, bulk flakes, and are easily distinguished by their clear dark outlines and color. Occasionally,² there is a thin, clean flake. Monolayer and bilayer graphene are distinguishable by contrast and color (see Fig. 2.1 bottom panel for an example). Usable hBN thicknesses are distinguishable by color.

After amassing the flakes, the next step is to stack them in the desired order using standard dry transfer techniques [67]. For this, I use a polymer “stamp”: a sticky polymer stretched across a soft polymer and affixed to a glass slide with double sided tape. The sticky polymer I use is typically Poly(Bisphenol A carbonate),³ hereafter PC. The soft polymer is PDMS in the form of Gel-Pak DGL-17-X8. I assemble the stack of flakes from top to bottom. The top layer is either hBN or a graphite top gate. I do the initial pick-up around 80°C to encourage the flake to adhere to the polymer. Subsequent pick-ups can be performed at lower temperatures, because the flakes stick to each other incredibly well because of van der Waals forces between their pristine and flat surfaces.

The “tear-and-stack” method is an ingenious way of creating a precise relative rotation between two graphene layers. If I were to randomly align two graphene layers, then I would have less than a 1% chance of being within 0.1° of the magic angle. Sometimes, graphene flakes have obvious crystalline edges with corners at a multiple of 30°. It is possible, although difficult, to optically align these edges to

²My hit rate for good (hundreds of square microns of area with no tape residue or visible defects) monolayer and bilayer graphene sheets tends to be around one per five square centimeters, however there is significant variance. It typically takes me a few minutes to search one square centimeter, so the total time to find a good monolayer of graphene varies from fifteen minutes to one hour.

³In the form of Gel-Pak DGL-17-X8, double stacked.

within several tenths of a degree. Much more convenient is to start with one flake of graphene, cut it in half, pick up one half, twist it, and then pick up the second half [68].

For the devices in this dissertation, I use the edge of an hBN flake to tear the graphene in half. This step is unreliable (Fig. A.1). Sometimes the graphene does not tear, but instead pulls itself into a string. Sometimes it pulls the hBN down off of the stamp. The process also likely imparts significant heterostrain in the graphene layers. Much more reliable (and potentially less straining) is the “cut-and-stack” process, wherein the starting graphene flake is cut with a laser or an AFM tip prior to stacking. Some groups include an insulating monolayer WSe₂ flake to stabilize the twist angle [49].

At every step in the stacking process, various substances can become trapped between the heterostructure’s layers. Even helium can be trapped due to graphene’s impermeability to gas [69]. Typically however, trapped hydrocarbons coalesce into isolated bubbles due to the extreme strength of the van der Waals interaction between layers, leaving atomically-perfect interfaces where the bubbles are not [70]. For twisted bilayer graphene, I do not typically thermally anneal the final stack out of fear of the twist angle relaxing to 0°. So, I have to hope for a relatively low density of bubbles, and route my final Hall bar through bubble-free regions.

After assembling the structure on the stamp, I melt it down onto the desired final substrate, typically SiO₂ with pre-patterned metal alignment marks, at roughly 160°C. During this process, it is often possible to see bubbles between the layers being pushed or squeezed out of the heterostructure.

After preparing the stack, I use standard lithography techniques to add a gold top gate, etch the stack into a Hall bar shape, and deposit one dimensional metallic edge contacts [67]. I then glue and wire bond the sample to a ceramic chip carrier that interfaces with our cryostats.

I spent several months attempting to replicate my group’s ferromagnetic device [40]: twist angle 1.2°, with hBN aligned to one layer of graphene. I initially attempted 25 stacks, ultimately yielding 8 fabricated devices. The low yield was mostly due to the unreliability of the tear step. The next step was to screen the devices and look

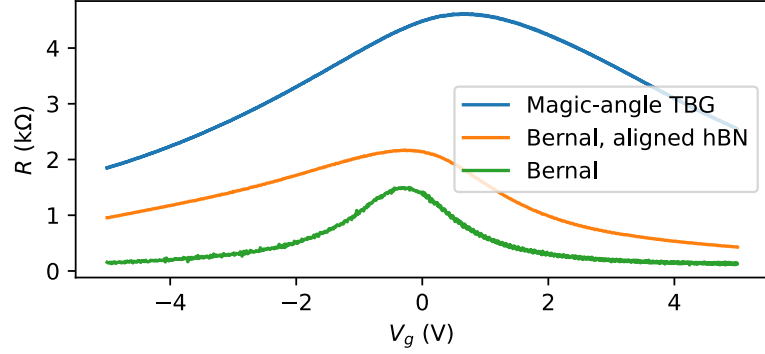


Figure 2.2: **Room-temperature behavior of bilayer graphene devices.** Magic-angle TBG has higher resistivity on the positive gate (electron) side of CNP compared to the negative (hole) side. Bernal bilayer graphene with aligned hBN has the opposite behavior, and Bernal bilayer without aligned hBN is symmetric. The gate capacitances of the three devices in this figure are not identical, so a given gate voltage does not correspond to the same density in the three cases.

for signatures of interesting physics in their transport.

2.2 Device screening and basic characterization

For each fabricated device, I first check contact resistances and gate effect at room temperature. Good contacts to twisted bilayer graphene typically have a resistance of 5 - 10 kΩ. I measure this by sourcing 1 mV DC to each contact in turn, leaving the rest of the contacts grounded. I also check the two-point resistance for each pair of contacts by floating all other contacts. Some information about the twist angle and alignment of the graphene and hBN layers can be ascertained with a quick gate sweep, as shown in Fig. 2.2, although I avoid pushing the gates past a few volts out of fear of electrochemistry involving water in the air. Assuming the contacts work and the gate has an effect at room temperature, I cool the device to 1.7 K in our variable temperature insert (VTI) cryostat for further characterization.

The first measurement in the VTI is always to repeat the contact resistance and gate effect checks. I then carefully measure gate current as I sweep the gate voltages (with everything else grounded) to check for leakage. hBN can typically handle at least

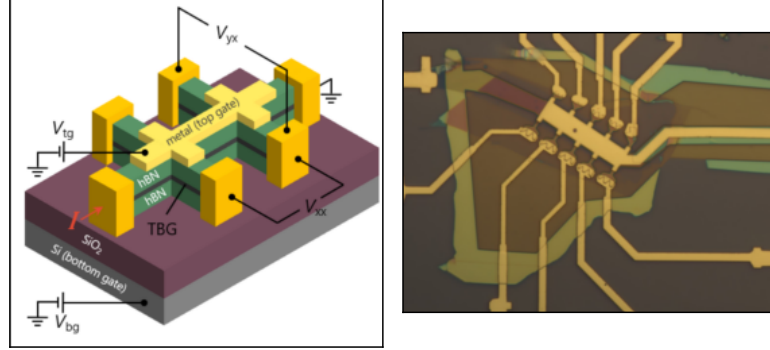


Figure 2.3: **Transport measurement schematic.** **Left:** Schematic of a typical Hall bar with a metal top gate. Some devices are significantly longer, with many voltage probes along their length. Some devices have a graphite top gate and/or a graphite back gate in addition to the Si back gate. Figure adapted from Refs. [71] and [72]. **Right:** An example microscope image of a completed TBG device. The Hall bar is $4\ \mu\text{m}$ wide and $20\ \mu\text{m}$ long. This device has a graphite back gate and a gold top gate.

$0.5\ \text{V/nm}$ of electric field before it starts to break down, however there is significant variation from sample to sample. In thin hBN, shortly before complete dielectric breakdown, an exponential (in gate voltage) tunneling current flows [73]. Back off the gate voltage immediately upon seeing this current well before it exceeds a few hundred pA!

If I know the thickness of one of the hBN layers, usually from an AFM measurement, then I have a reasonably accurate way to determine the scaling between applied gate voltage and charge carrier density, from

$$n = \frac{\epsilon_r \epsilon_0}{de} V \quad (2.1)$$

where $\epsilon_r \sim 3$ is the permittivity of hBN, d is the hBN thickness, and e is the electric charge. With top and bottom gates, this becomes

$$n = \frac{\epsilon_r \epsilon_0}{e} \left(\frac{V_{TG}}{d_{TG}} + \frac{V_{BG}}{d_{BG}} \right) \quad (2.2)$$

and the associated electric displacement field (per ϵ_0 , assuming perfect screening by the graphene⁴) is

$$D = \frac{\epsilon_r}{2} \left(\frac{V_{TG}}{d_{TG}} - \frac{V_{BG}}{d_{BG}} \right). \quad (2.3)$$

With two gates, I can independently tune density and displacement field [74]. I am interested in the resistivity of the sample as a function of these tuning knobs, among others. Despite it being relatively straightforward to measure resistance, source current or voltage and measure voltage or current, some care is still necessary to get good measurements.

For one, I use a four-terminal geometry, which means that I source and drain current in separate contacts from where I measure voltages to avoid picking up the contact resistances. The actual amount of excitation needs to be simultaneously large enough to measure with high signal to noise ratio while being small enough to not dissipate enough power to heat the sample or otherwise disturb its electronic structure. I use a lock-in amplifier with a large bias resistor (typically 100 M Ω or 1 G Ω) to source current, and drain it via an current pre-amplifier.⁵

Two example gate maps are shown in Fig. 2.4. The top left panel is a sample that has alignment to hBN. This is evident from the faint superlattice peaks on either side of charge neutrality (note similarity to Ref. [27]). I suspect that this sample has relaxed to Bernal stacking, because the resistivity of the charge neutrality point increases dramatically with applied displacement field, as in Ref. [74]. The top right and bottom panels show a magic-angle twisted bilayer graphene sample. It has large band-insulator superlattice peaks at [32, 34]

$$n_s = \frac{4}{A} \approx \frac{8\theta^2}{\sqrt{3}a^2} \quad (2.4)$$

where A is the moiré unit cell area and $a = 0.246$ nm is the lattice constant of

⁴Almost certainly a wrong assumption, most of the time, because the graphene's DOS is typically quite low.

⁵At 1.7 K, I typically use a 10 nA current source at 17.76 Hz, and at millikelvin temperatures I use 1 nA at closer to 1 Hz. I amplify voltages with NF corporation LI-75A voltage preamplifiers. I use SR830 and SR860 lock-in amplifiers in some measurements, and a more convenient multiterminal lock-in amplifier [75] in others. I typically use an Ithaco 1211 current pre-amplifier to drain current.

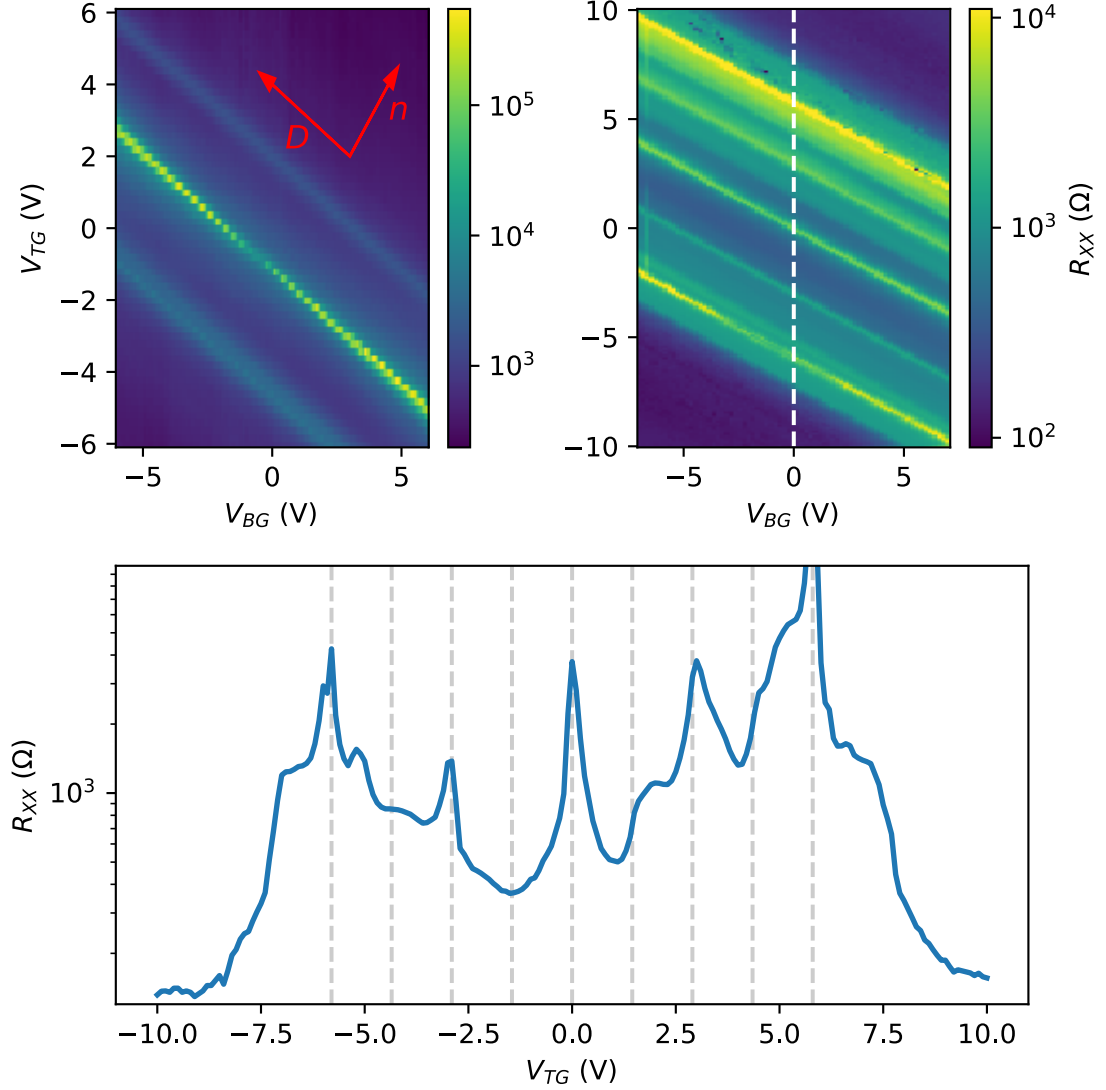


Figure 2.4: **Typical gate maps for twisted graphene devices at 1.7 K.** Discussed in the main text. **Top left:** A 2D gate map of a sample with aligned hBN but no graphene twist. The red arrows show the approximate directions of increasing density and displacement field. **Top right:** Magic-angle twisted bilayer graphene, same device as shown in Fig. 2.5. **Bottom:** Line cut through the gate map from the top right panel. The vertical dashed lines are integer n/n_s up to ± 4 .

graphene.⁶ This sample, being close to magic angle, also shows correlated insulator peaks at various integer filling factors within the flat moiré bands.

I also apply a weak out-of-plane magnetic field to the sample in order to measure the classical Hall effect and back out a second and more accurate measurement of the gate voltage vs density plot according to

$$\frac{dR_{xy}}{dB} = \frac{1}{ne}. \quad (2.5)$$

At higher fields, Hofstadter’s butterfly gives the another measurement of n_s , as described in Ref. [32]. The intersection points of Landau levels emanating from CNP and full-filling are evenly spaced as a function of $1/B$. This spacing (times h/e) gives the most accurate measurement of the superlattice area A .

Annoyingly, many of the devices that I made had relaxed to Bernal stacking at some point before measurement. This was particularly frustrating in the few cases where I managed to properly align the TBG to the hBN (as in the left panel of Fig. 2.4). I managed to hit magic angle twisted bilayer graphene in several devices and aligned hBN in several devices, however I never fabricated a magic-angle sample with aligned hBN.

Most of the devices that I measured had a significant amount of disorder. It manifested in several ways:

- Calculations of twist angle between adjacent contact pairs (typically separated by one to three microns) would disagree by at least 0.1° .
- Behavior from one contact pair to the next, and even between contact pairs on opposite sides of the Hall bar, would be dramatically different, even if their calculated twist angle was similar. For instance, one contact pair would superconduct, but its neighbors would not.
- There would be signatures of superconductivity, however the resistance would not go to zero, indicating ”puddles” of superconductivity. dV/dI curves would

⁶Note that this equation does not account for strain in the graphene layers. This is included in Chapter 5.

appear to have multiple critical currents (Fig. 2.5 top right).

- Features that I would expect to be sharply-defined would instead be blurred or doubled up, especially in field.

In 2019, most TBG groups were still using the rather crude tear-and-stack method, and tended to have similar amounts of disorder. There were results showing beautiful and complicated phase diagrams in potentially lower-disorder samples, such as in Ref. [59], in which the authors attempted to clean and relax their TBG using an AFM. However, it was still practically difficult to reliably realize such clean samples.

For the devices that show promising transport at 1.7 K, I cool them to millikelvin temperatures in a dilution refrigerator and measured them more carefully. The magic-angle device from Fig. 2.4 is shown again in Fig. 2.5. As is typical, the correlated insulating states at half filling of electrons and holes are sharply defined, and signatures of superconductivity appear immediately next to them. Quantum oscillations become strongly pronounced, and the associated quantum Hall plateaus appear. At such low temperatures, contact resistances tend to become extremely high, requiring careful tuning of the Si back gate to yield useful measurements. Some devices, such as that shown in Fig. 2.5, become too resistive to measure, severely limiting their usefulness.

I summarize the stacking and fabrication process of one particular device in Fig. 2.6. In order: I first found and picked up the top layer of hBN (20 nm thick, **a** and **b**), then found and performed tear-and-stack with a flake of monolayer graphene (**c** and **d**, at this step I also attempted to align a straight edge of the graphene with an edge of the top hBN, see Fig. A.2), then found and picked up the bottom layer of hBN (13 nm thick, **e** and **f**), and then the bottom graphite gate (**g** and **h**), then melted down the stack (**i**), deposited a Ti/Au top gate (**j**), etched the Hall bar (**k**), and deposited Cr/Au edge contacts (**l**). The electrical measurements of this device will occupy the majority of this dissertation.

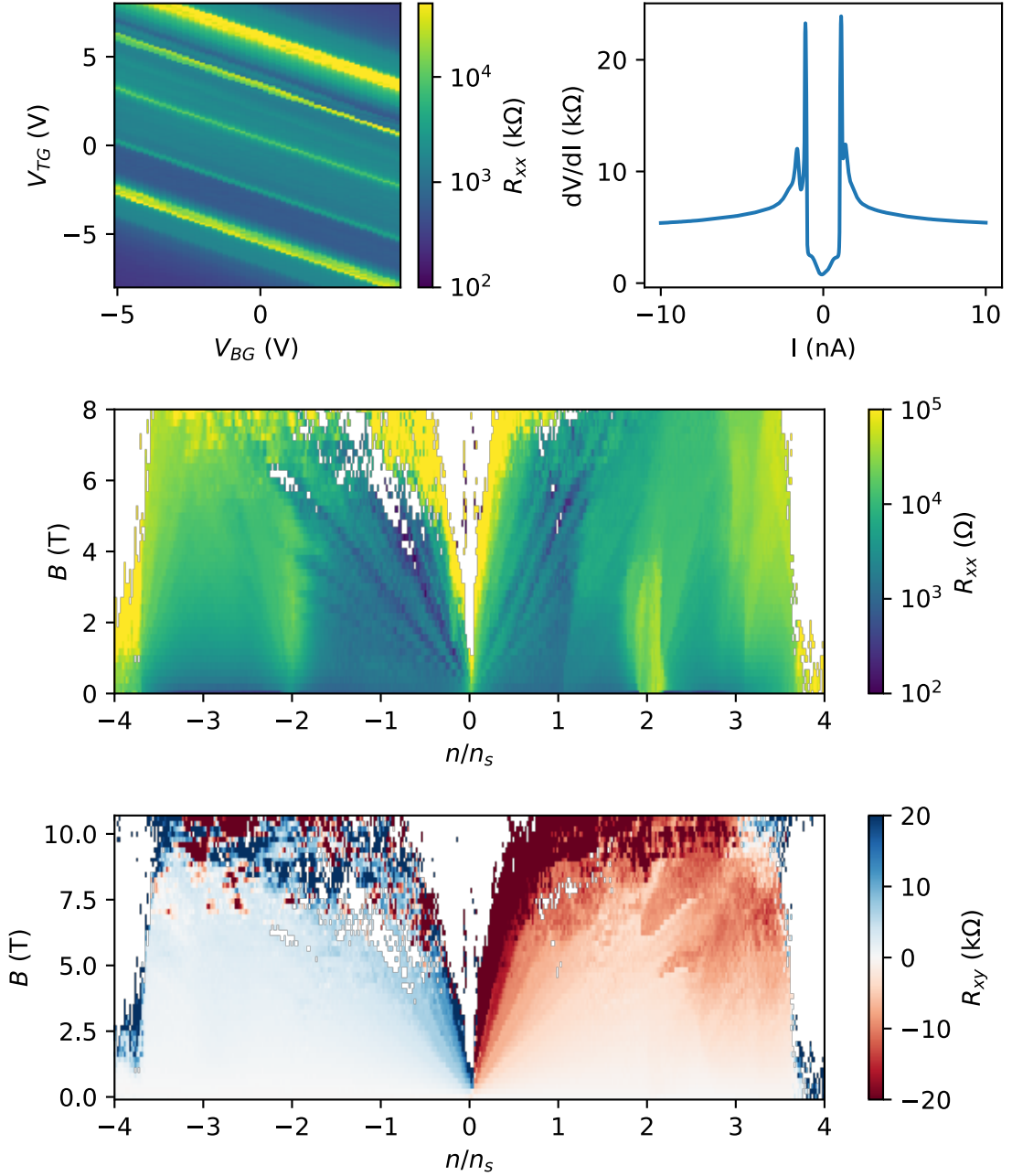


Figure 2.5: **Transport data for a typical disordered magic-angle device.** **Top left:** The same gate map as in Fig. 2.4’s top right panel, except at 30 mK. At this low temperature, the correlated insulating peaks become significantly sharper in gate. **Top right:** dV/dI curve at a point of low resistance adjacent to a correlated insulating peak, showing signatures of superconductivity. **Middle:** R_{xx} vs filling and field. Features emanate from 0 field and n/n_s -4, -2, 0, 1, 2, 3, 4. **Bottom:** R_{xy} vs filling and field. Near CNP, quantum Hall plateaus are visible.

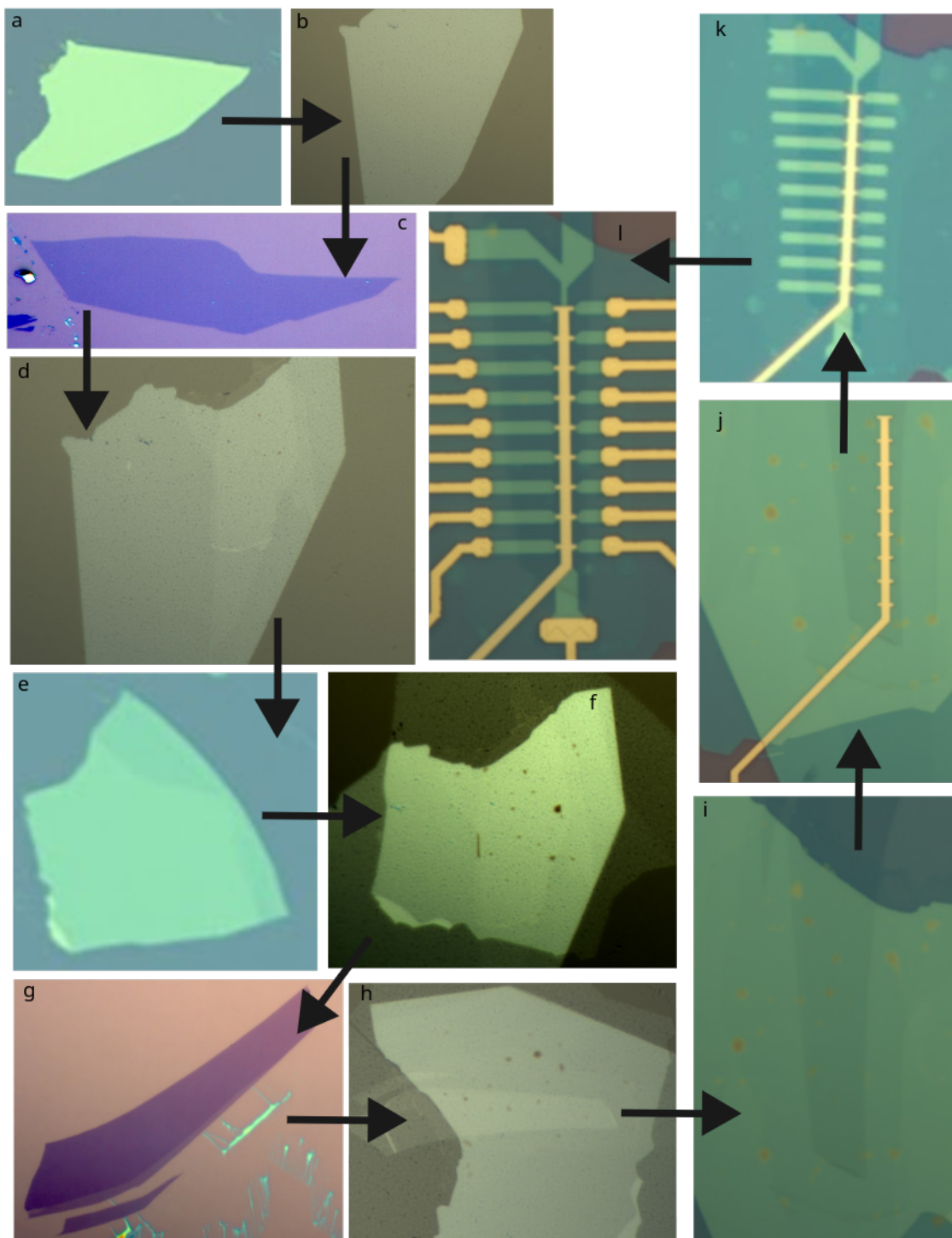
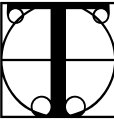


Figure 2.6: Fabrication steps for my best device. Described in the text.

Chapter 3

Unusual magnetotransport measurements

O summarize the end of Chapter 2: in an attempt to replicate ferromagnetism in magic-angle TBG [40, 50], I used the “tear-and-stack” dry transfer method [34] to create a TBG device with a target twist angle of 1.2° and with an optically-aligned hBN/graphene edge. The device, shown in Fig. 3.1 (left), had a gold top gate and thin graphite back gate.¹ Its top and bottom hBN were 20 and 13 nm thick respectively.² It had 9 voltage probes on each side, labelled 1-9 on the left and 11-19 on the right. The channel width was $1\text{ }\mu\text{m}$, and longitudinal voltage probes were separated by 3 squares.³

The device did not show ferromagnetism in transport. It had no signatures of aligned hBN, and its twist angle was 1.4° , well *above* the target angle, and far enough

¹The thin graphite back gate was a poor choice, as it itself is a high quality 2D material whose density of states, and hence ability to function as a gate, can vary with voltage and magnetic field [76]. I was not aware of this effect when I fabricated the device.

²The thinness of the hBN may also be problematic. It is comparable to the wavelength of the moiré pattern, potentially leading to screening effects [39, 47].

³In 2D, the conversion between resistance and resistivity is a geometric factor, because the two quantities have identical units: ohms. For a simple Hall bar, the geometric factor is the distance between longitudinal probes divided by the width of the channel, commonly referred to as the number of squares.

above the magic angle that no strong correlations should have existed. When immersed in an out-of-plane magnetic field, however, the device displayed large magnetoresistance within a range of densities on either side of charge neutrality along with quantum oscillations that defied the usual Diophantine description. The device turned out to be an enigma. Every time I looked more closely at its measurements or tried some new analysis, I noticed something new and unexpected.

I measured it three separate times in our VTI (base temperature 1.7 K, 14 T max field),⁴ twice in our dilution refrigerator (base temperature 27 mK, 14 T max field), and once at MagLab (base temperature 50 mK, 28 T max field, Chapter 6), all the while trying to piece together some coherent picture of its underlying physics. In this chapter, I present the measurements in approximately chronological order, along with various hypotheses that I (and others) came up with to try to explain them.

3.1 Initial unusual measurements

I show the device's resistivity as a function of top gate voltage in Fig. 3.1 (middle). The vertical dashed line at 0 gate is charge neutrality,⁵ and the dash-dotted lines at ± 5.5 V are full filling/emptying of the moiré bands.⁷ For the 20 nm hBN thickness, this corresponds to $1.4 \pm 0.1^\circ$ twist angle (from Eqs. 2.1 and 2.4). Aside from a few

⁴For measurements in the VTI, I used our multi-terminal lock-in amplifier [75] to simultaneously measure every one of the 16 longitudinal and 9 Hall contact pairs in the device. These sets of plots were useful for characterizing variation across the length of the device, for instance leading to Fig. 3.1's right panel. Where such plots provide useful additional information, I have included the details from all contact pairs in Appendix A.

⁵Often charge neutrality is not at exactly 0 V applied, but is shifted by several volts, likely due to trapped charged impurities. We know that it is still charge neutrality because it has a characteristic shape (linear conductivity in gate [19, 77]), and a maximum resistivity on the order of a few k Ω .⁶

⁶The theoretical maximum DC resistivity of graphene is $\pi h/4e^2 \approx 20$ k Ω [78–80], however measurements tend to find anywhere between 2 and 6 k Ω [19, 77, 81]. At low energies, twisted bilayer graphene above the magic angle should appear as two monolayers in parallel: half the resistivity. hBN alignment typically opens a gap at charge neutrality by breaking sublattice symmetry, resulting in a much higher maximum resistivity [26, 82, 83].

⁷With TBG it is relatively straightforward to orient ourselves within the bands by looking for CNP plus insulating full-filling/emptying peaks in resistivity. For magic-angle samples, it is possible to confuse the correlated insulating states at half filling with these full-filling states [32]. Be careful!

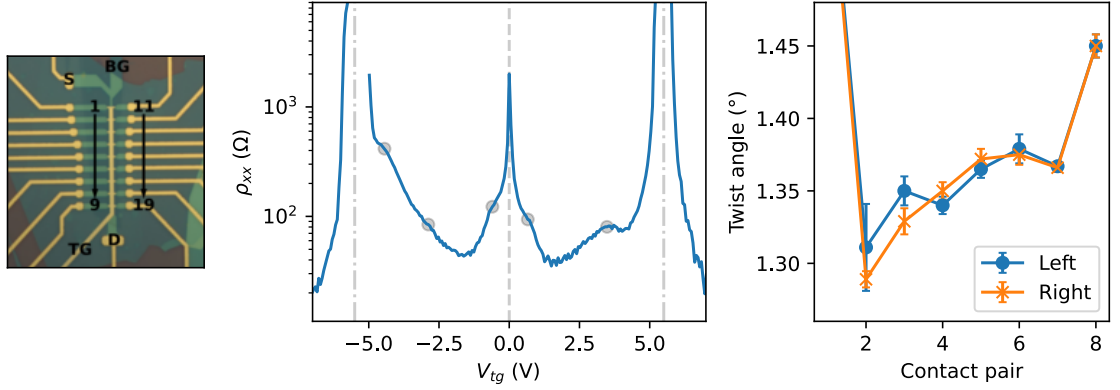


Figure 3.1: **Device overview.** **Left:** Naming scheme for the many contacts on the device. S and D were used to source and drain current. BG and TG were graphite back and gold top gates. 1-9 and 11-19 were voltage probes. **Middle:** Top gate sweep for contact pair 16-17, described in the text. **Right:** Twist angle variation along the length of the device.

shoulders and small bumps in the data (grey circles),⁸ there are no strong correlation features between CNP and full-filling, as expected for a device twisted so far above the magic angle.

3.1.1 Large, quadratic magnetoresistance

I show a 2D gate map at ± 0.5 T in Fig. 3.2. A large QAH response associated with ferromagnetism would appear as a strong feature in the difference of the two measurements. In my measurements there is no such feature. Instead, in the longitudinal resistance, a region of large, positive magnetoresistance appears in the vicinity of half filling on both sides of CNP. Even for a field of 0.5 T, the magnetoresistance ratio, defined as

$$\text{MR} = \frac{\rho(B) - \rho(0)}{\rho(0)}, \quad (3.1)$$

⁸These small bumps and shoulders would later turn out to be a crucial piece of the puzzle: they are the van Hove points that I analyze in Chapter 5. At the time, I assumed they were either extremely weak correlated insulating states, or else somehow a product of inhomogeneity.

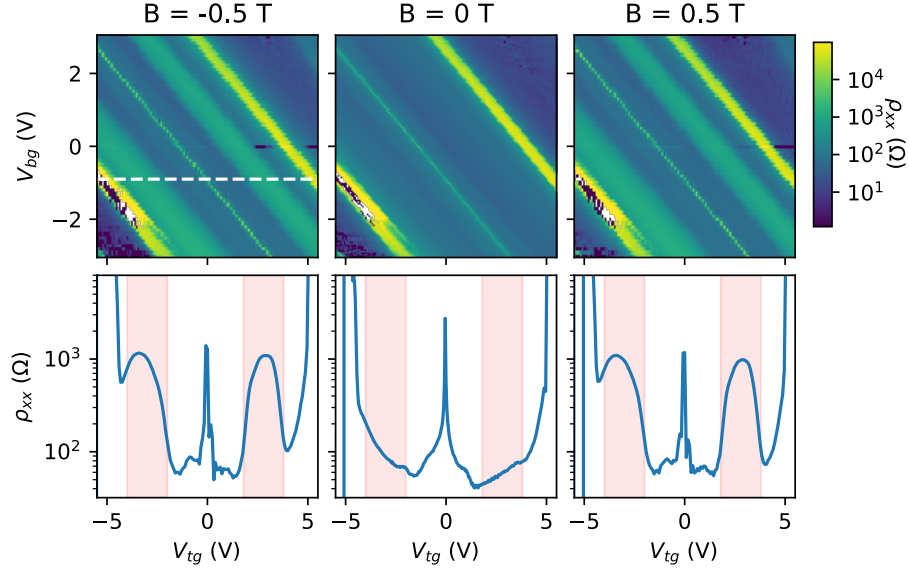


Figure 3.2: **2D gate maps in low fields.** The dashed white line indicates the location of line cuts in the bottom panels. The pink vertical sections indicate approximate locations of magnetoresistance regions.

reaches over 80. The magnetoresistance continues to grow roughly quadratically out to about 5 T, where it eventually saturates at 300 (Fig. 3.3, bottom panel). The shape of the magnetoresistance as a function of density is also curious, being relatively flat-topped, rather than sharply peaked (Fig. 3.3, top panel). It is not clear whether the magnetoresistance saturates because of the onset of quantum oscillations or for some other reason.

3.1.2 Splitting, bending Landau levels

At stronger magnetic fields, a complicated set of quantum oscillations appear, originating from CNP as well as full filling and emptying of the moiré bands (Fig. 3.4). The pattern of Landau level degeneracies varies from one contact pair to the next (Fig. A.6), but in most of them, near CNP, $\nu = \pm 4, \pm 12, \pm 20, \dots$ are the most prominent. This is the expected sequence for twisted bilayer graphene at high twist angles [34, 84]: exactly double that observed in monolayer graphene. The pattern

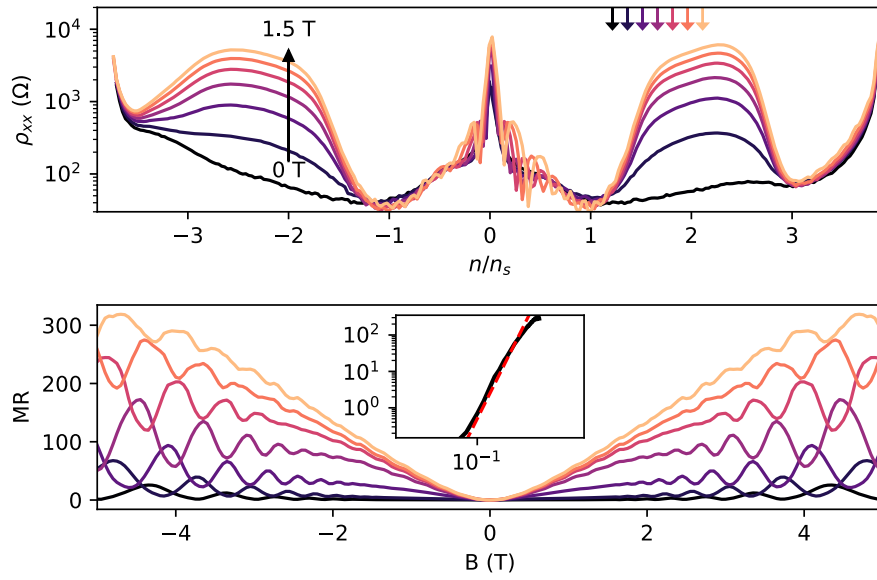


Figure 3.3: **Magnetoresistance characteristics.** See Figs. A.4 and A.5 for the fans that these line cuts are taken from. **Top:** Longitudinal resistivity as a function of density for contacts 16-17 at evenly spaced magnetic fields between 0 and 1.5 T. **Bottom:** Magnetoresistance vs field at the densities indicated with arrows in the top panel. **Inset:** Same axes, log-log plot of the largest-magnetoresistance trace. The dotted red line is a quadratic overlaid by eye.

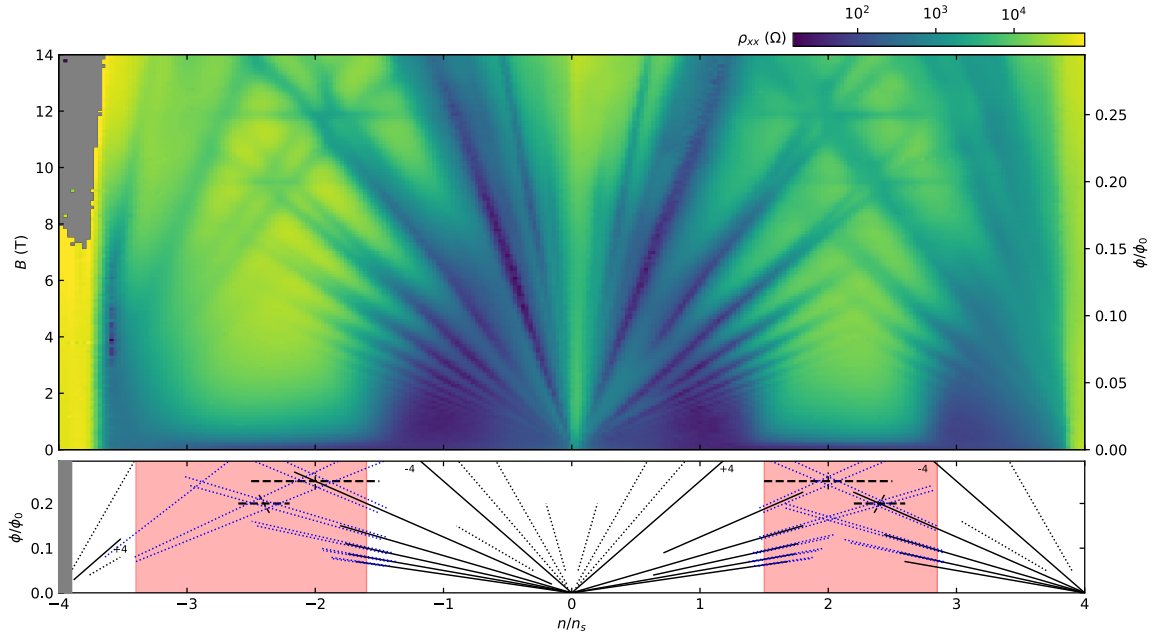


Figure 3.4: **Landau fan at 1.7 K.** See Fig. A.6 for all fans. **Top:** Longitudinal resistivity as a function of density and field for contacts 16-17. Grey indicates lost signal. **Bottom:** Schematic for the top panel. Pink shaded regions are magnetoresistance regions. Solid lines are ν multiples of 4 LLs, grey dotted lines are not. Blue dotted lines are split LLs with non-integer slopes. Dashed crosses indicate visible crosses at intersections of LLs.

is not electron-hole symmetric. For instance, $\nu = -8$ is visible, although faint, at far lower field/fillings than $\nu = +8$. Similarly, $\nu = -16$ appears prominently, while $\nu = 16$ does not. Such inconsistent behavior between electrons and holes as well as between devices, or different contact pairs on the same device, is typical of TBG devices from the literature.

At higher doping, $\nu = \pm 8, \pm 16, \pm 24, \dots$ appear prominently. The zeroth LL degeneracy breaks into $\nu = \pm 1, \pm 2, \pm 3$ above roughly 4 T.

When these quantum oscillations enter the regions of strong magnetoresistance, the negative-slope lines of minimum-resistivity from CNP appear to split into $\nu \rightarrow \nu \pm 0.4$ (blue dotted lines in Fig. 3.4). So, for example, $\nu = -8$ splits into $\nu = -7.6$ and $\nu = -8.4$. Positive-slope lines split by a smaller amount, roughly 0.3. From $n/n_s = -4$, there are $\nu = 4.5, 7.4$, and 8.6 , whereas from $n/n_s = 4$ the splitting is less. Some of the split lines appear to bend when approaching lines travelling the other direction. Where prominent pairs of split lines intersect, there are crosses (vertical or near-vertical lines meeting horizontal lines) of diminished resistivity.

3.2 Plausible explanations

This magnitude of magnetoresistance is rare in materials [85]. Most metals' magnetoresistances max out at a few percent; here I see tens of thousands of percent. Large magnetoresistance can have many causes. For example:

- In structures consisting of alternating magnetic and nonmagnetic materials, one can observe a few tens of percents of negative “giant magnetoresistance” [86–88].
- In certain magnetic perovskites one can observe a few orders of magnitude of negative “colossal magnetoresistance” near ferromagnetic to paramagnetic phase transitions [89–91].
- In semiconductor-metal hybrid structures, in which the current can flow through the metal at low field, but at high field it is redirected to flow through the semiconductor, positive “extraordinary magnetoresistance” can be made even larger [92, 93].

None of these explanations apply to a Hall bar of twisted bilayer graphene with no signs of magnetism. A few other explanations are more likely.

3.2.1 Compensated semimetal?

Large, quadratic, positive magnetoresistance can occur in compensated semimetals, in which multiple Fermi pockets and thus types of charge carriers coexist (see [85], chapter 12).

There is a simple and intuitive explanation for this behavior. Consider a material with only one type of charge carrier. In a magnetic field, the Lorentz force leads to an electric field perpendicular to the flow of current. In equilibrium, the force from this electric field cancels out the Lorentz force from the magnetic field, so the charge carrier trajectories are straight lines.⁹ With two carrier types, the single transverse electric field cannot cancel the transverse force on both types simultaneously. The carrier trajectories will take longer, more circular paths through the material, leading to a positive magnetoresistance. If the carriers have opposite sign and equal density, then there will be precisely zero transverse electric field, maximizing the effect.

In a simple Drude model, the conductivity tensor can be written as [94]

$$\hat{\sigma} = e \left[\frac{n\mu}{1 + i\mu B} + \frac{p\mu'}{1 - i\mu' B} \right], \quad (3.2)$$

with its inverse, the resistivity tensor,

$$\hat{\rho} = \frac{1 + \mu\mu' B^2 + i(\mu - \mu')B}{e(n\mu + p\mu' + i(p - n)\mu\mu' B)} \quad (3.3)$$

where n , μ and p , μ' are the densities and mobilities of electrons and holes, respectively. The real (imaginary) parts of $\hat{\sigma}$ and $\hat{\rho}$ are the longitudinal (transverse) conductivity and resistivity. If $p = n$, then the longitudinal resistivity grows as B^2

⁹In reality, because the drift velocity is only an average, there is still a slight deviation from straight lines and thus a small magnetoresistance, typically saturating at low field.

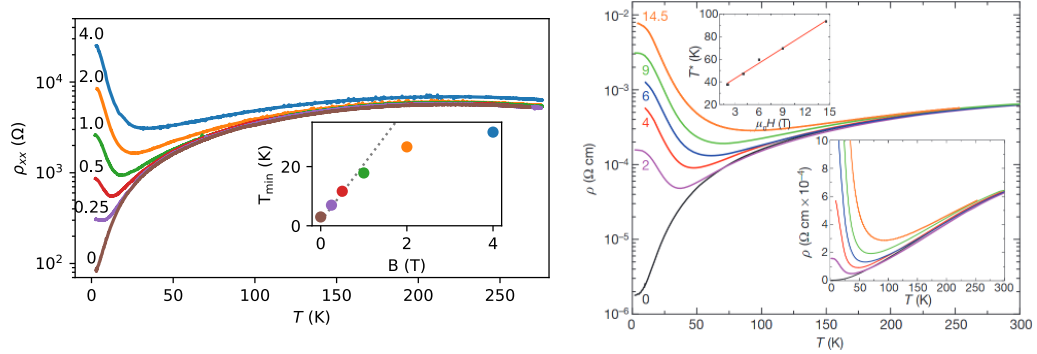


Figure 3.5: **Temperature dependence of magnetoresistance. Left:** Resistivity vs temperature at indicated fields, in Tesla, for contact pair 16-17 at $n/n_s = -2.8$. See Fig. A.8 for all contact pairs. **Inset:** Temperature of the minima of the curves vs field. The dashed line is an approximate fit to the lowest three fields. **Right:** Fig. 2 from Ref. [94]. The top inset is the same as the inset in the left panel.

without saturating. This behavior is claimed to account for magnetoresistance in several materials, notably WTe_2 [94,95]¹⁰ and Cd_3As_2 [96], but also in elemental metals such as Bi, Sb, W, and Mo [85].

So, I hypothesize that in those regions with large magnetoresistance, my device features multiple carrier types. This hypothesis makes several predictions for transport that I describe next. Ultimately, the hypothesis will be shown inadequate.

Temperature dependence of magnetoresistance

Ref. [94] Fig. 2 (reproduced in Fig. 3.5, right panel) shows resistivity vs temperature at several fixed magnetic fields in WTe_2 . I performed the same measurement and found qualitatively similar behavior (Fig. 3.5, left panel): as temperature increases, the resistivity in field drops to a minimum before slowly increasing again. Curiously, a 20% magnetoresistance ratio at 4 T persists all the way up to 275 K.¹¹

¹⁰There is conflicting evidence on whether or not the magnetoresistance in WTe_2 originates from equal electrons and holes, or from some other mechanism. See Ref. [95] and references therein for an overview.

¹¹In other contact pairs the room temperature magnetoresistance is much higher, reaching above 250% at 14 T (Figs. A.9 and A.10). It is very high at charge neutrality in one contact pair, but highest at full filling in most others. The entire moiré bandwidth is roughly one hundred meV at these twist angles, somewhat larger than room temperature. Even contact pairs with no appreciable

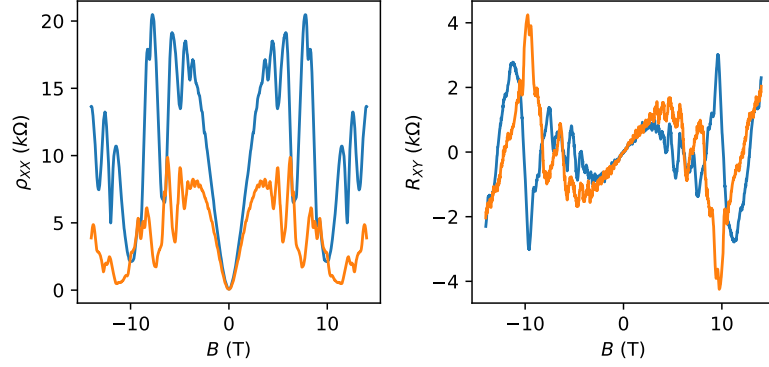


Figure 3.6: **Nonlinear Hall slope.** Longitudinal resistivity (left, contact pair 16-17 in blue and 6-7 in orange) and Hall resistance (right, contact pair 6-16 in blue and 7-17 in orange) vs field at 1.7 K at $n/n_s = 2.5$. See Figs. A.11 and A.12 for all contact pairs. The left panel is symmetrized, and the right panel is antisymmetrized. Note the extent to which the neighboring contact pairs show quantitatively different behavior.

For comparison with Ref. [94], I also show the temperature at which the resistivity is minimized (Fig. 3.5, left panel inset). Ref. [94] found a roughly linear relationship all the way out to 14.5 T with a slope of approximately 4.4 K/T (right panel upper inset). My results are clearly nonlinear above 1 T, with a slope of roughly 20 K/T below 1 T. Potentially, the nonlinearity in my measurements is caused by saturating magnetoresistance.

The similarity between the two systems encouraged me to look for additional evidence for two carrier types.

Nonlinear Hall resistance

Ref. [95] Fig. 2b shows the Hall resistance vs field at various densities in WTe_2 . For densities where they observe large magnetoresistance, the Hall slope is nonlinear. From Eq. 3.3, at low fields, the slope is dominated by whichever carrier has the highest mobility. At high fields, it is dominated by the sum of the two densities.

I had already noticed significant nonlinearity in my own measurements of the Hall magnetoresistance at 1.7 K had high magnetoresistance at room temperature.

resistance (Fig. 3.6 right panel) after antisymmetrizing.¹² Unfortunately, despite my best efforts, I could not find a set of parameters $\{n, p, \mu, \mu'\}$ that gave an acceptable fit to both longitudinal resistivity and Hall resistance simultaneously for any adjacent contact pairs, as was performed in Ref. [95]. I assumed that this was caused by inhomogeneity in the device: contact pairs that should behave similarly, such as the two curves in each panel of Fig. 3.6, were actually remarkably different.

The clear nonlinearity in my Hall measurements was promising evidence for my two-carrier hypothesis, despite numerically not working out.

Magnetoresistance exponent

Ref. [95] fit their low-field magnetoresistance to an explicit power law of the form

$$\rho_{xx}(B) = a + b|B|^c \quad (3.4)$$

for fit parameters a , b , and c . They (along with Refs. [97, 98]) found that the power law was not quadratic, but instead typically varied between 1.6 and 2.0 depending on density and temperature. They did not have a theoretical expectation of nonanalytic behavior.

I found similar behavior in my sample. In all contact pairs with significant magnetoresistance, within the magnetoresistance regions the best-fit exponent varied between 1.0 and 1.6 (Fig. 3.7). This was yet more weak evidence for a compensated semimetal hypothesis.

¹²There was an unusual amount of mixing of XX into XY and vice versa. At the time, I didn't think much of it. Most devices typically have a small amount (a few percent at most) of mixing from lithographic imprecision. More mixing can occur if inhomogeneity causes current to serpentine through the device rather than follow the Hall bar directly, which is what I assumed I was dealing with here. The sheer amount of it in every contact pair with large magnetoresistance (Hall resistances with *larger* symmetric components than their neighboring longitudinal resistances) should have tipped me off that something funny was going on. This mixing would later turn out to be a critical clue, covered in Chapter 5.

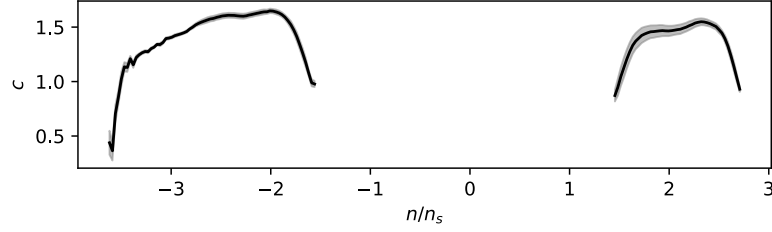


Figure 3.7: **Magnetoresistance exponent.** Best-fit exponent for magnetic fields between 0.4 and 1.5 T for contact pair 16-17. See Fig. A.13 for all contact pairs. The grey outline is one-sigma deviation using the least squares error.

3.2.2 Partial band polarization?

I had suggestive evidence for compensated-semimetal-like behavior in my sample. However, this was not predicted by any band structure calculations. I reasoned that maybe I was close enough to magic angle for correlations to have a weak restructuring effect on the bands, potentially causing compensated semimetallic behavior.

It was noticed early on¹³ that in magic-angle TBG samples, Landau levels emit in only one direction from each integer filling factor: towards higher absolute density relative to charge neutrality [37, 42, 59, 99]. This effect coincides with a “reset” in the Hall resistance back to zero. The mechanism was eventually worked out to be a particular set of sharp phase transitions happening at each integer filling factor [43]. The bands initially populate with carriers equally, but at integer filling factors one band consumes all carriers from the other bands, filling itself entirely and emptying the others. This process repeats at each integer filling factor, and neatly explains outward-projecting Landau levels.

My device was not precisely at the magic angle, so I reasoned that this band polarization may be occurring, but only partially. In TBG devices from the literature (Ref. [37] for instance), the Hall number does not always reset entirely to 0 at all filling fractions, so this seemed reasonable.

A schematic of this idea is shown schematically in Fig. 3.8. There are four bands, but I will assume that they operate in pairs, for simplicity.

¹³Including in my own samples, see Fig. 2.5 for an example.

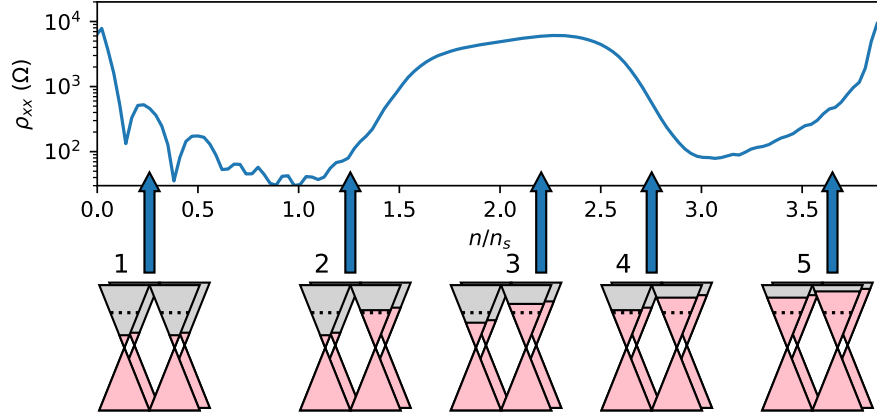


Figure 3.8: **Partially polarized bands schematic.** The top panel is the 1.5 T curve from Fig. 3.3. The schematic beneath it is described in the text.

1. At low densities, both sets of bands begin to fill as normal, as indicated by the partly shaded pink regions.
2. At the density at which the magnetoresistance begins to turn on, one set of bands steals carriers from the other, but it does not polarize completely. Perhaps its energy crosses a van Hove singularity (horizontal dotted line) and it becomes hole-like. If not that, then at least it has a different density than the other set of bands.
3. In the magnetoresistance region, there are two carrier types, leading to large magnetoresistance according to the compensated semimetal model. The Landau levels should split because there are now two distinct Fermi surfaces, and the amount of splitting should be a function of the amount of band polarization.
4. When the magnetoresistance region begins to turn off, the second set of bands has reached a similar filling as the first set.
5. Things now proceed as normal to the band edge. There is only one type of carrier, so there is no large magnetoresistance.

This model made one main prediction outside of those implied by the two-carrier

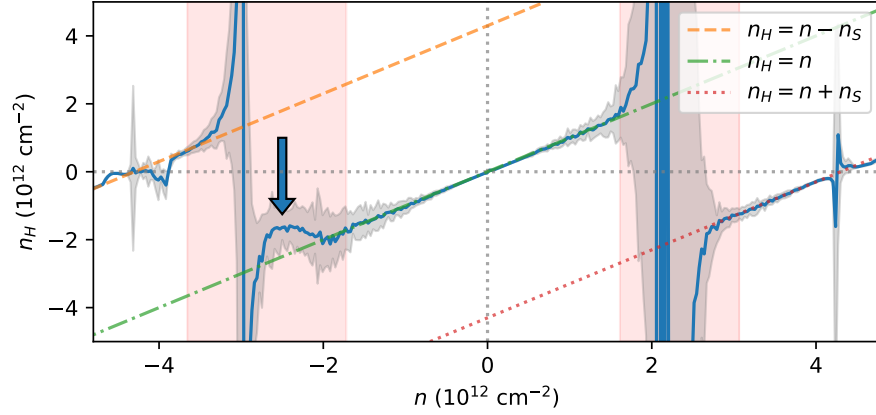


Figure 3.9: **Hall number vs density.** Contact pair 6-16. All contact pairs are shown in Fig. A.14. The grey region surrounding the blue line is ± 1 standard deviation away from the slope. Pink shaded regions are approximate locations of the magnetoresistance regions.

predictions: there should have been a partial Hall number reset at densities where the magnetoresistance appears and disappears. This prediction was easy to check.

The Hall number vs density is shown in Fig. 3.9. There was indeed what I would call a partial reset in the Hall number upon entering the magnetoresistance region in one location, at least on one side (indicated with an arrow). It was visible in most contact pairs with magnetoresistance, although it was not visible on the other side of CNP. Near the center of the magnetoresistance regions there was a sign-change singularity in the Hall number, which I assumed was associated with a van Hove singularity and a Lifshitz transition: the Fermi surface switched from electron-like to hole-like or vice versa.¹⁴

3.3 Dilution refrigerator measurements

The remaining piece of evidence for a compensated semimetal interpretation of my results was the presence of two distinct frequencies in the quantum oscillations as a

¹⁴In Chapter 5, I prove the preceeding paragraph basically entirely wrong. I do not think the bump indicated with an arrow is a partial Hall reset, and the sign-change is not associated with a van Hove singularity.

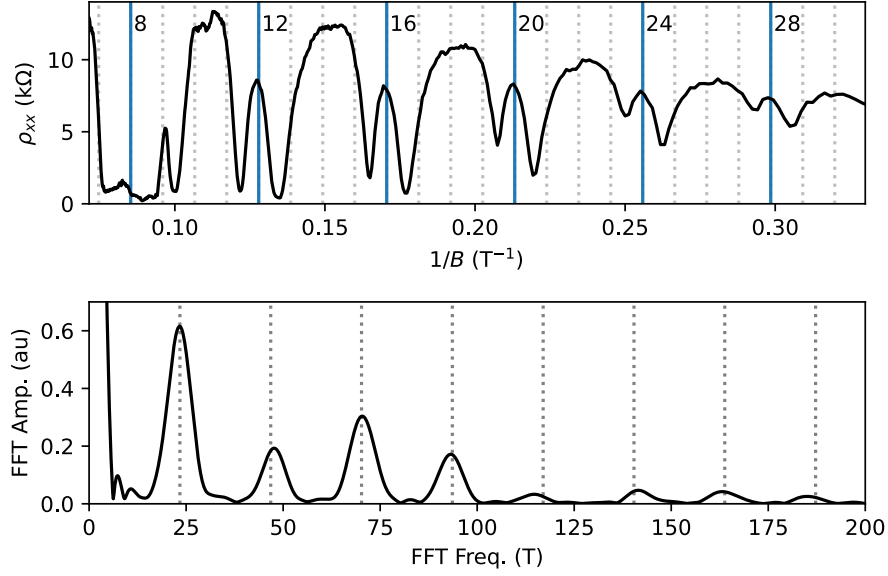


Figure 3.10: **There is only one SdH frequency.** Contact pair 16-17, $n/n_s = -2.07$. **Top:** Split LLs as a function of $1/B$. Every 4th LL is indicated with a blue vertical line. The split LLs clearly have the same frequency in $1/B$, but shifted phases. **Bottom:** Fourier transform of the above, with vertical dotted lines every 23.4 T.

function of $1/B$, like those measured in WTe_2 [95]. I reasoned that because there was only partial polarization of the bands, the two densities would likely be similar and thus the two frequencies would be similar. In addition, it was possible that, for whatever reason, the two frequencies might have distinct mobilities, leading one to dominate at 1.7 K. So, I suspected that I would need lower temperatures to observe the two peaks distinctly. While I waited for dil fridge time, I prepared a draft of a manuscript describing the data and my partial polarization plus two-carrier hypotheses. Eventually, the dil fridge opened up, and I cooled the device to roughly 26 mK¹⁵ and started collecting data.

There is only one frequency (plus harmonics) of SdH oscillations at all densities where there is magnetoresistance. An example is shown in Fig. 3.10, where the frequency is roughly 23.4 T. $4e/h \times (23.4 \text{ T}) \approx 2.3 \times 10^{12} \text{ cm}^{-2}$. This frequency

¹⁵At such low temperatures the contacts tend to become highly resistive. I applied 20 V to the Si gate to help alleviate this issue.

agrees with the extrapolated Hall density, meaning that if there were two carriers, one of them must have an extremely low density, low enough that it would not give large magnetoresistance according to Eq. 3.3. So, the entire compensated semimetal hypothesis is wrong.

In hindsight this should have been obvious. I had previously described the Landau levels as splitting into $\nu \rightarrow \nu \pm 0.4$, implying that the split Landau level gaps project backwards to $B = 0$ and $n = 0$, which they mostly do. With polarized bands, the Landau level gaps from the polarized bands should project back to some density other than $n = 0$. This would be the case even for partially polarized bands, and my 1.7 K data was perfectly sufficient to show it.

There was only one thing to do, and that was to keep taking data and trying to figure out what was going on.

3.3.1 Surprise superconductivity

Upon cooling the sample to 26 mK, two longitudinal contact pairs around 1.34° twist angle¹⁶ displayed superconductivity when hole doped (Fig. 3.11, top panel). The superconductivity was extremely weak, with a critical temperature of roughly 150 mK, a critical field of only 3 mT, and a critical current of 12 nA (Fig. 3.11, bottom panels). The two contact pairs with superconductivity had relatively weak magnetoresistance. One had essentially none, and the other reached a magnetoresistance ratio of about 15—high for normal materials, low for this device.

I measured the differential resistance dV/dI as a function of applied DC current and magnetic field. This revealed phase-coherent transport in the form of a Fraunhofer-like pattern with a period of roughly 3 mT, indicating a normal area of roughly $h/2e(3 \text{ mT}) \approx 0.7 \mu\text{m}^2$.

We did not purposefully fabricate any Josephson junctions within our device, so such behavior should not be expected. However, the presence of Fraunhofer patterns in TBG is well-known, and likely arises from accidental Josephson junctions created

¹⁶Pair 13-14, 1.33° , dropped below my measurement floor of roughly 0.5Ω . Pair 3-4, 1.35° , looked qualitatively similar, but bottomed out at a few ohms, presumably due to a small normal region in series with the superconductor.

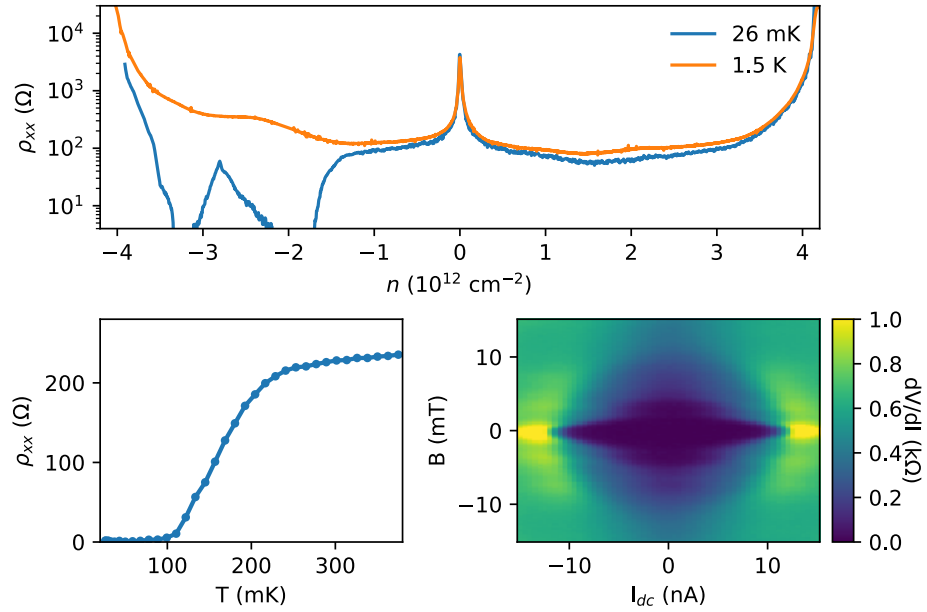


Figure 3.11: **Unexpected superconductivity.** Contact pair 13-14, twist angle 1.33° . **Top:** Comparison of gate sweep in VTI at 1.5 K and dil fridge at 26 mK. **Bottom left:** Temperature dependence of superconductivity at $n = -3.2 \times 10^{12} \text{ cm}^{-2}$. **Bottom right:** Differential resistance as a function of DC bias current and field demonstrating weak Fraunhofer-like behavior at $n = -3.2 \times 10^{12} \text{ cm}^{-2}$. I removed a small flux jump around $B = -7 \text{ mT}$ in preparing this figure.

by moiré inhomogeneity [37, 42]. Indeed, such patterns are one of the strongest pieces of evidence for a true superconducting state in TBG, as opposed to some other state with very low resistivity.

To our knowledge, superconductivity had not been observed at such a high angle previously. It had been seen at angles as low as 0.93° [100], or 0.79° with an insulating WSe₂ layer between hBN and TBG [49], but it had not been seen above 1.2° , as far as I was aware. Consistent with previous reports claiming strange metal behavior [38, 44, 45], I observed linear-in- T resistivity between 20 and 90 K above the superconducting domes.

Curiously, the SC became significantly weaker after the magnetic field was ramped to 14 T and back to 0. The minimum resistivity became $10\ \Omega$, where before it was below my measurement floor of $0.5\ \Omega$. This happened on both cooldowns of the sample in the dil fridge: upon first cooling it the resistivity was indistinguishable from zero, but after ramping the field, it became finite but small. It could not be explained with a small trapped flux in the superconducting magnet.

3.3.2 Magnetotransport

The sample was cold in the dil fridge in mid-March, 2020. I had previously taken a cold Landau fan diagram (the dil-fridge equivalent of Fig. 3.4), however it was at relatively low resolution in order to save time and prepare for the next user of the fridge. Greater world events occurred, however, that meant that for at least a few months we would not be able to prepare and swap out samples. So, I had extra time to measure. I decided to use the time to take high resolution gate maps, as well as a high resolution Landau fan diagram.

Gate maps in field

Gate maps at fixed fields are shown in Fig. 3.12. There is no apparent effect of displacement field on transport at 0 T. At 3 T, negative displacement field enhances the peak value of the magnetoresistance and widens the density extent of the magnetoresistance region. The resistivity at the charge neutrality point increases with

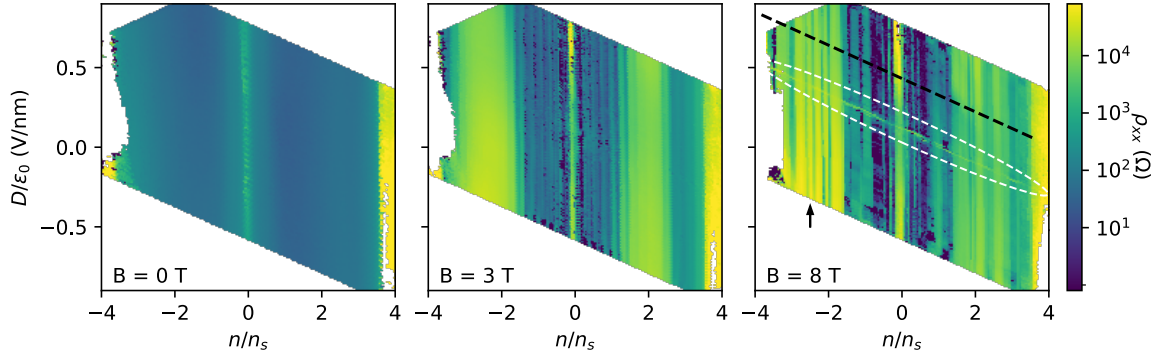


Figure 3.12: **26 mK gate maps in field.** Gate maps at 0 field, at 3 T, and at 8 T for contact pair 16-17. The black dashed line is the cut used for the high-resolution Landau fan diagram, while the white ellipse indicates the loss of signal when the back gate is fixed at 0 V. The arrow indicates one pair of split LLs. Additional contact pairs in Fig. A.15.

increasing magnitude of displacement field, regardless of the polarity of this field. At 8 T, the split LLs are visible as vertical lines at constant density.

In other contact pairs (Fig. A.15) there were oscillations as I tuned the displacement field at certain densities. I did not have a satisfactory explanation for this observation, however I noted that they appeared to be more closely related to the value of the back gate voltage rather than the displacement field. Just as the back gate gated the moiré channel, the moiré channel gated the back gate [76]. At high magnetic field, carrier density in the back gate might have been substantially and non-monotonically modulated with back gate voltage. One way this could affect electronic properties of the moire channel is through changes in screening, given that the lower hBN layer was only about 13 nm thick, comparable with the moiré wavelength.¹⁷

Landau fan

The Si back gate was screened by the graphite back gate which extended only over the main conduction channel, but was not screened near the contacts. This allowed me to set the carrier density near contacts to be high to encourage contact transparency.

¹⁷In Chapter 6 I return to this observation, as it becomes necessary to describe transport at high fields.

Unfortunately, when the graphite back gate was set near 0 V, I found that the contact resistances dramatically increased in a magnetic field, leading to a loss of signal at lower temperatures, regardless of the Si back gate voltage. Thus, for the following high-resolution Landau fan diagram, I fixed the back gate at 1.5 V and swept the top gate. This meant that the measurement was not performed at a constant displacement field. Instead, it followed the black dashed line in Fig. 3.12.

As I tune both field and density, a complicated series of quantum oscillations originates at the charge neutrality point and $B = 0$ and propagates outwards (Fig. 3.13). Near charge neutrality, the oscillations look similar to those of ordinary magic-angle TBG devices [37, 39, 42], with filling fractions $\nu = \pm 4, \pm 8, \pm 12, \dots$ being the most prominent: a different sequence than at 1.7 K (Fig. 3.4). To within experimental precision, these have zero longitudinal resistance and quantized Hall resistance. As I tune the density into the regions with large magnetoresistance, gaps with $\nu = \pm 6, \pm 10, \dots$ disappear. Each fourfold degenerate Landau level gap appears to split into a pair with slopes roughly corresponding to $\nu = \pm 8 \pm 0.5, \pm 12 \pm 0.5$, and so on (14 T is not enough to allow tracking the $\nu = \pm 4$ levels into the magnetoresistance regions, see Chapter 6 for higher-field data). These split lines do not have zero longitudinal resistance, reaching a minimum of a few hundred ohms. Nor do they follow exactly straight lines. Instead, they bend when approaching other levels. For lack of a better term, I will continue to refer to them as Landau level gaps.

Landau levels also propagate inward from full filling/emptying of the isolated moiré bands toward lower electron/hole filling, respectively, and these split and bend similarly to those originating from charge neutrality. We can determine Φ/Φ_0 by considering the points where these levels cross those originating at charge neutrality. For instance, the line with $\nu = +8$ originating at $n/n_s = -4$ must intersect the line with $\nu = -12$ originating at charge neutrality at $\Phi/\Phi_0 = 1/5$. In the following discussion, I refer to fields by their values of Φ/Φ_0 .

The phenomenology near the intersection of split Landau level gaps travelling in opposite directions follows a consistent pattern throughout the fan diagram. The example of the +8 and -12 lines from the previous paragraph is shown in Fig. 3.14, left panel. As mentioned above, each Landau level gap splits into a lower and upper

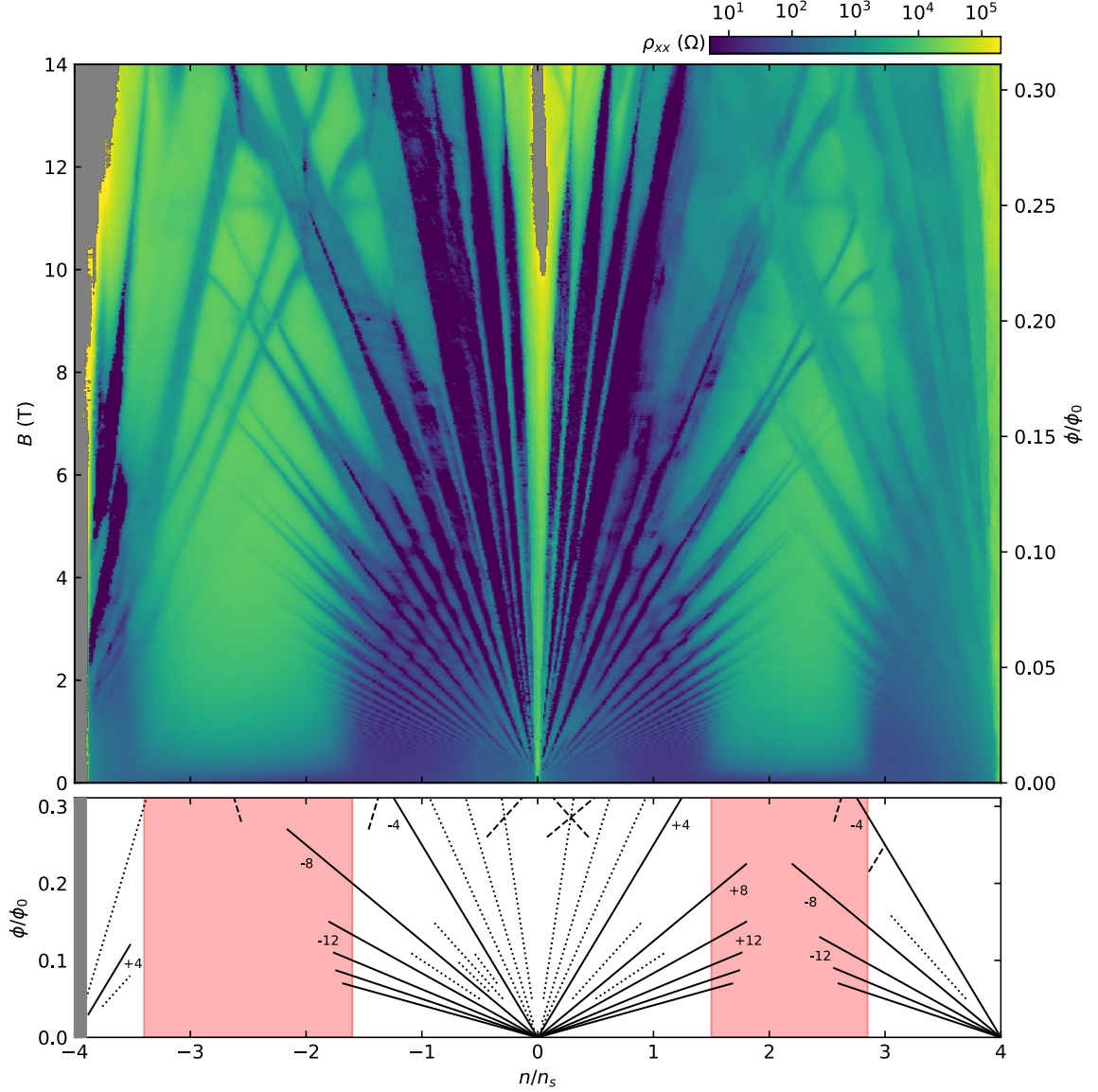


Figure 3.13: **Unusual Landau fan diagram.** Other contact pairs in Figs. A.16 and A.17. **Top:** Landau fan diagram taken at 26 mK for contact pair 16-17, the same as in 3.4. **Bottom:** Schematic fan diagram corresponding to top. Red shaded regions are regions with large magnetoresistance at low field. Solid (dotted) lines are symmetry-preserving (-broken) LLs coming from either charge neutrality or a band edge. Dashed lines are resistance minima corresponding to non zero s and t .

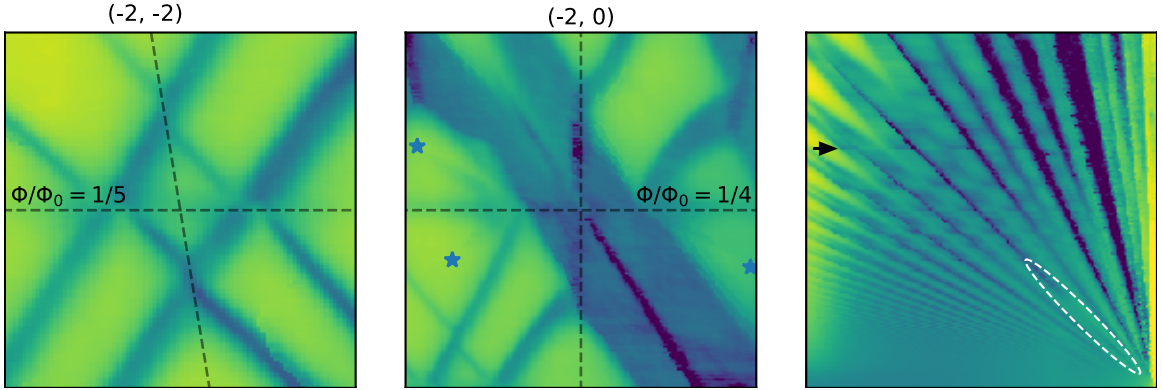


Figure 3.14: **Fine detail within the fan diagram.** **Left:** Detail of the crossing of split LL gaps -12 from charge neutrality ($s, t = 0, -12$) and $+8$ from $n/n_s = -4$ ($s, t = -4, 8$), and **(Middle)** -8 from charge neutrality and $+8$ from $n/n_s = -4$. The horizontal lines are at the indicated Φ/Φ_0 , and the lines with steep slopes are the average (s, t) of the crossing LL gaps. For the case of panel A, this is the average of $(0, -12)$ and $(-4, 8)$ which is $(-2, -2)$ as indicated. **Right:** Magnetotransport for contact pair 7-8 at low field and low density. The dashed circle indicates a disappearing-then-reappearing LL gap, and the horizontal arrow indicates a horizontal line of low resistance.

level. When a lower (upper) level overlaps with a lower (upper) level moving in the other direction, it changes direction to follow a line originating from half filling ($s = \pm 2$, steeply sloped dashed lines in Fig. 3.14) with slope equal to the average of the two intersecting levels, which is -2 in this case. Within the overlap, the resistivity minima tends to be deeper. Two crossings of a lower with an upper line occur at the same field that the non-split Landau level gaps would have intersected (horizontal dashed lines in Fig. 3.14), which is $1/5$ for this example. There is no drop in resistivity where these two intersect. Instead, they appear to displace horizontally by the width of the level that they are crossing before resuming their previous slope. The result of these changes in direction is that in between the overlaps the split LL gaps are shifted slightly toward each other.

The overlap of the split LL gaps around $+8$ and -8 originating from $n/n_s = -4$ and charge neutrality, respectively, shows the same phenomena (Fig. 3.14, middle panel). In this case, the intersection is at $\Phi/\Phi_0 = 1/4$, and the average slope is 0 , so we see a vertical line of low resistivity. In addition, there are faint additional levels

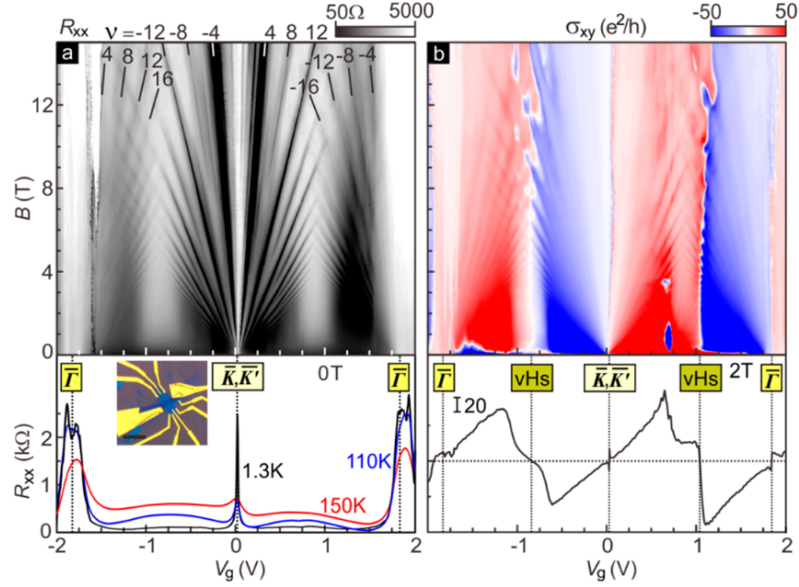


Figure 3.15: **Identical phenomenology from the literature.** Figure 1 from Ref. [101]. Panel a is longitudinal resistivity R_{XX} , and panel b is Hall conductivity σ_{XY} .

emanating outwards from the two intersections of lower with upper lines.

At low density and fields, some of the Landau level gaps disappear and then reappear as the field is increased (Fig. 3.14, right panel). Also, unusual horizontal lines (constant field) appear in within many of the Landau levels. These lines appear to take steps upon crossing Landau levels. These are seen to some extent near full-filling and emptying ($n/n_s = \pm 4$).

3.4 Additional devices, mine and others'

Scanning the literature, I found several published measurements that looked at least a little bit like what I saw in my device. The most strikingly similar was in Ref. [101], which studied a 2° TBG device. I reproduce their main figure in Fig. 3.15. They clearly see large magnetoresistance regions and split Landau level gaps (top of panels a and b), although they do not mention these features in the text. Instead, they focus on the van Hove singularity that they associate with the sign change in Hall number

(lower panel b).¹⁸

I found similar-looking behavior in Ref. [42] Fig S5 panels A and G, however this device was close enough to the magic angle (1.27°) that it had weak Landau levels projecting from half-filling on both sides. The magnetoresistance in that regime may have been caused by correlations, rather than whatever caused it in my sample.

After we shared our measurements with other researchers, Matt Yankowitz contacted us to tell us that he had measured similar behavior in two of his devices, both also above the magic angle. As he was hunting for magic-angle samples at the time, he had not published them anywhere. He shared the measurements with us, and I reproduce them in Figs. A.18 (D34, $\theta = 1.59^\circ$) and A.19 (D25, $\theta = 1.52^\circ$). D34 clearly showed regions of quadratic magnetoresistance and split Landau levels, however the features were not as crisp as in my device, probably because of inhomogeneity within the device. D25 had smaller regions of quadratic magnetoresistance, however it did not show clear split Landau levels.

I also attempted to reproduce the strange magnetotransport in additional devices of my own, by simply fabricating twisted bilayer graphene devices around 1.5° target angle. It turned out that the “unusual” magnetotransport was actually fairly ordinary. Most TBG devices that I made showed some aspect of the “unusual” behavior! I will highlight one such device here, a device that I named French, after the chess opening.

French had six voltage probes on each side of a long Hall bar with a graphite back gate and a gold top gate. Unfortunately, several of the voltage probes did not work,¹⁹ so I only effectively had seven longitudinal and three Hall contact pairs. I will show results from only one pair here, but the others are qualitatively similar.

French’s gate maps (Fig. 3.16) looked similar to my good 1.4° device’s without an applied magnetic field: a charge neutrality point whose resistivity does not significantly change with displacement field, insulating superlattice peaks on either side,

¹⁸Several chapters from now, I will show that this sign change is surprisingly not associated with a van Hove point!

¹⁹For some reason, on the final metal deposition step several of the metal traces out to bond pads lifted off.

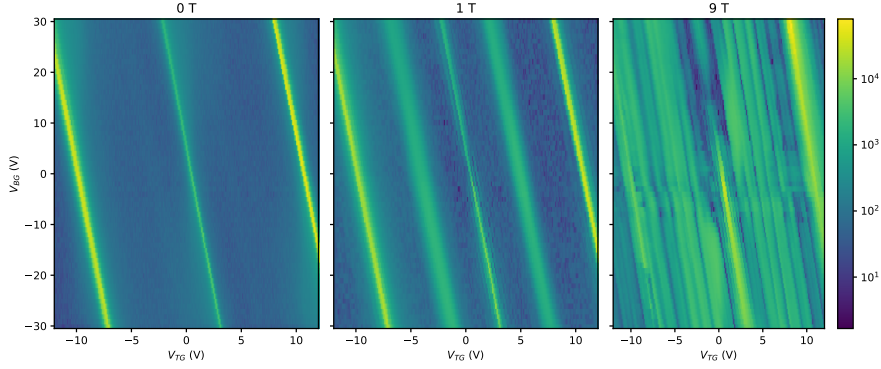


Figure 3.16: **French gate maps.** Gate maps taken at the indicated fields at 1.7 K.

and nothing much in between.²⁰ Upon applying a weak magnetic field, magnetoresistance regions appear. Note similarity with Figs. 3.2 and 3.12. Only at high fields, here 9 T, did displacement-field dependence appear, again like in my previous device. From the full-filling density, I backed out a twist angle of roughly 1.7° .

I cooled the device to dil-fridge temperatures and took a fan, shown in Fig. 3.17. Clearly visible are magnetoresistance regions with MR ratio reaching 80-100 before saturating around 2 T. Unlike in the previous device, there was a broader MR region that appeared around 0.2 T and disappeared by 0.5 T. A similar effect was visible in Matt's D34. The progression of prominent Landau level gaps originating at CNP was, on the hole side, $\nu = -4, -12, -20, -28, \dots$, and on the electron side, $\nu = 4, 8, 12, 16, 24, \dots$. From both full-emptying and full-filling, I saw all ν multiples of 4, however the longitudinal resistance did not go to 0 and the Hall slope was not quantized.

On the hole side, where I expected to see $\nu = -8, -16, \dots$, I instead saw only split LLs. Where they met split LLs originating at full-emptying, they formed an intricate pattern of bends: the same as in my previous device, only more intricate because there were more overlapping split LLs. As with my previous device, the electron side appeared more blurry than the hole side. I happened to measure French in a system with an in-plane magnet. Out to 4 T in-plane, there was no change in

²⁰If you look closely, there are faint features in between that I would later ascribe to van Hove singularities.

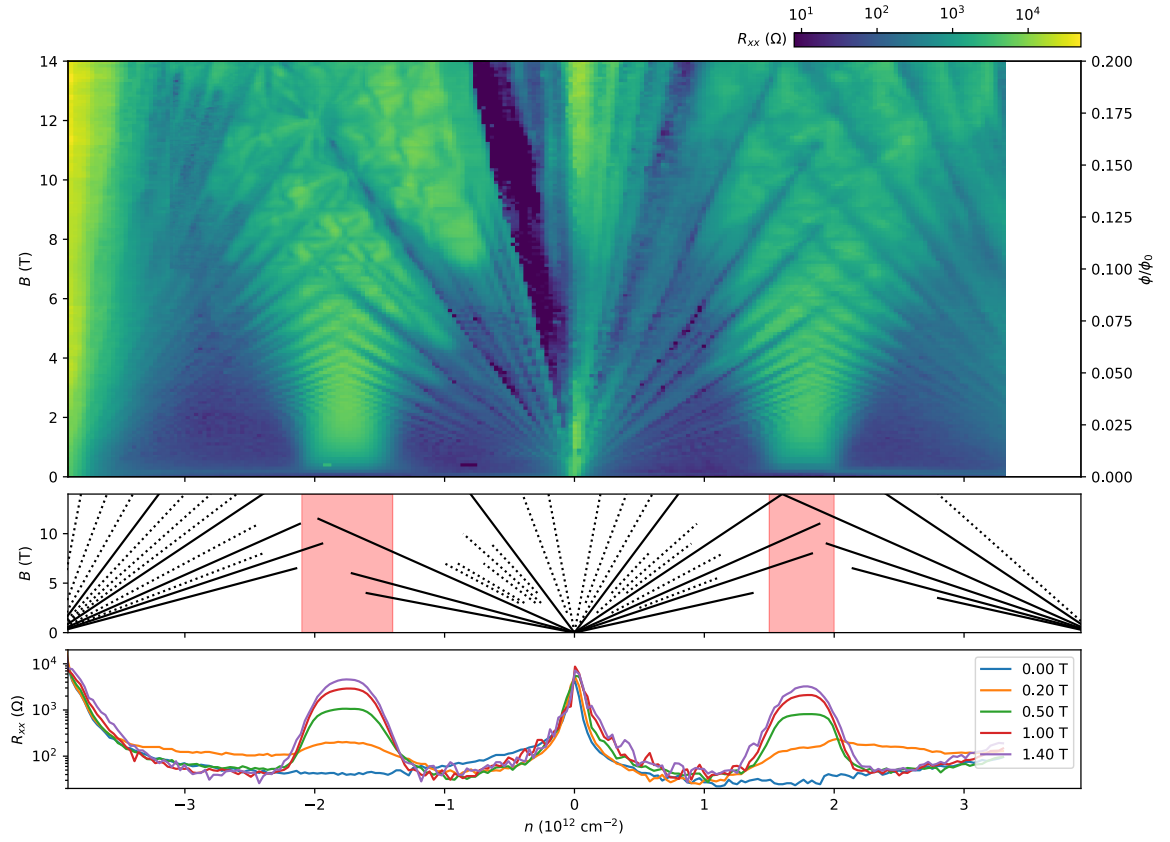


Figure 3.17: **French fan.** **Top:** Longitudinal resistivity vs field and density at 27 mK. **Middle:** Schematic diagram. **Bottom:** Line cuts from the top panel at the indicated fields.

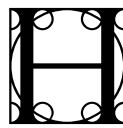
the longitudinal resistivity (Fig. A.20).

The ease with which I and others replicated the unusual magnetotransport led me to believe that it is not at all unusual. What was unusual was the quality of my nice device: the others all showed blurrier, less-crisply-defined features. This was probably a consequence of relative homogeneity in my device compared to others.

The sharpness of the features, along with the generic behavior in other devices, led me to suspect that the problem could be solved relatively simply. Because it appeared in devices up to 2° twist angle, I expected that I could solve it in a single-particle framework. Because it involved the overlap of Landau levels from opposite sides of a Bloch band, I expected that Hofstadter's butterfly, the general single-particle theory of Bloch electrons in a magnetic field, would play a role. By pure chance, I had learned how to compute Hofstadter's butterfly in a square lattice in Steve Kivelson's condensed matter theory class only a few months prior. So, I started there.

Chapter 4

Toy model based on Hofstadter’s butterfly

 OFSTADTER’S butterfly is the extension of the quantum Hall effect to the case of Bloch electrons. I guessed that a modified Hofstadter’s butterfly might capture the intricate behavior at intersections of split LL gaps shown in Fig. 3.14. All realistic Hofstadter’s butterfly models share certain traits: notably a fractal, butterfly-like spectrum, and Diophantine behavior of the gaps when plotted against density and field. I hoped that if I could find the intricate LL intersection behavior in a theoretically and computationally simple model then these shared traits would mean that it would apply equally well to twisted bilayer graphene.

I spent a few months attempting to modify the butterfly in physically realistic ways that might replicate the behavior.¹ On one attempt, I considered the effect of two commensurate moiré lattices, perhaps one from TBG and one from hBN alignment. The effect on the spectrum was to double up the butterfly: two (transformed) copies of it overlapped. By sheer chance, I had a bug in my implementation of the two-butterfly model—misplaced parentheses—leading to an extra factor of two in a hopping term in one direction but not in the other direction. To my surprise, I saw, in one corner of the resulting simulation, split Landau levels with exactly the same overlapping behavior as in my data.

¹This was during the early days of COVID lockdown, so I didn’t have anything better to do.

I soon isolated two ingredients that were necessary for this behavior: anisotropy, and butterfly doubling. I was briefly excited that I had found new evidence for nematic order and unit cell doubling, the former yielding anisotropy and the latter yielding butterfly doubling. In Chapters 5 and 6 I show that uniaxial heterostrain and Zeeman splitting are better candidates for the two added ingredients.

In this chapter, I first give a basic overview of Hofstadter's butterfly. Then, I show how the two added ingredients combine to give rise to precisely the splitting, bending, and intersecting behavior that I see in my measurements.

4.1 Butterfly basics

Start with a single-particle tight-binding model on a square lattice with lattice constant a , following Ref. [28]. Let $x = ma$ and $y = na$. The basis states are then $|m, n\rangle$ for $m, n \in \mathbb{N}$. Next, assume the simple Hamiltonian

$$\hat{H} = \sum_{m,n} |m+1, n\rangle \langle m, n| + |m, n+1\rangle \langle m, n| + \text{h.c.} \quad (4.1)$$

Use the Landau gauge $\vec{A} = Bx\hat{y}$. The Peierl's substitution [102], whereby the translation operators pick up a phase of

$$\theta = \frac{q}{\hbar} \int \vec{A} \cdot d\vec{r}, \quad (4.2)$$

does not affect hopping terms in x . Hopping terms in y , however, pick up a phase of $eBxa/\hbar$. Let $\alpha = \Phi/\Phi_0 = eBa^2/h$, the ratio of the magnetic flux per unit cell to the magnetic flux quantum. With this addition, the Hamiltonian becomes

$$\hat{H} = \sum_{m,n} |m+1, n\rangle \langle m, n| + e^{2\pi i \alpha m} |m, n+1\rangle \langle m, n| + \text{h.c.} \quad (4.3)$$

None of the hopping terms in the Hamiltonian depend on n , so we take as an ansatz plane wave behavior in the y direction, so $|m, n\rangle \rightarrow e^{-i\alpha n} |m\rangle$. After expanding

the Hermitian conjugate, we find

$$\hat{H} = \sum_m |m+1\rangle\langle m| + |m-1\rangle\langle m| + 2 \cos(2\pi\alpha m + \nu) |m\rangle\langle m|. \quad (4.4)$$

Plug it into the time-independent Schrödinger equation $\hat{H}\Psi = E\Psi$, and we can write it as a simple difference equation

$$g_{m+1} + g_{m-1} + 2 \cos(2\pi m\alpha + \nu)g_m = E g_m, \quad (4.5)$$

where the full wave function is

$$\Psi(x, y) \propto \sum_{m,n} g_m e^{-i\nu n} \Phi(x - ma, y - na) \quad (4.6)$$

for a single atomic orbital Φ . Equation (4.5) is known as Harper's equation or the Almost Mathieu operator, depending on who you ask. There are multiple ways of computing the spectrum of this Hamiltonian.

Hofstadter's computation method

Following Hofstadter's original work, [28], we can rewrite equation (4.5) as

$$\begin{pmatrix} g_{m+1} \\ g_m \end{pmatrix} = \begin{pmatrix} E - 2 \cos(2\pi m\alpha + \nu) & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} g_m \\ g_{m-1} \end{pmatrix}. \quad (4.7)$$

We call this 2×2 matrix $A(m)$. In order to be physically realistic, g must remain bounded for all m . If A is periodic in m with period q , then there must be an integer p such that

$$2\pi\alpha(m + q) + \nu = 2\pi\alpha m + \nu + 2\pi p, \quad (4.8)$$

which implies $\alpha = p/q$, and α is rational. We will proceed with this rationality as an assumption. We call the product of q successive A matrices Q , and it is a function of E and ν . For g to remain bounded, the eigenvalues of Q must have a magnitude of at most 1. A necessary condition for this is that the trace of $Q(E, \nu)$ is less than or

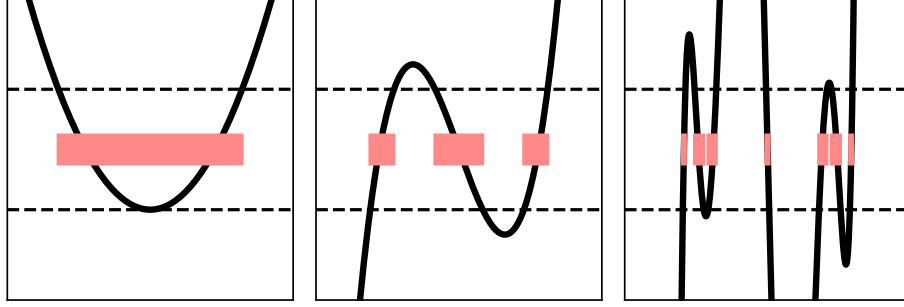


Figure 4.1: **Hofstadter's calculation examples.** $\text{Tr}(E)$ vs E for $\alpha = p/q = 1/2$ (**left**), $1/3$ (**middle**), $4/7$ (**right**). Dashed horizontal lines are ± 4 , and the thick red horizontal lines mark where the trace is within the dashed lines. Note that for $\alpha = 1/2$, the trace is equal to -4 at $E = 0$, so the two subbands kiss in the middle.

equal to 2 in magnitude. Hofstadter then argues that we can eliminate the factor of ν by restricting the trace to 4, yielding the final inequality

$$|\text{Tr}Q(E)| \leq 4. \quad (4.9)$$

This is now straightforward to solve: let $\alpha = p/q$, compute $Q(E)$ and its trace. Where the trace is less than 4 in magnitude, those are the allowed energies.

Some examples are shown in Fig. 4.1. The trace of Q is a degree- q polynomial, so we expect it to have q roots and thus approximately q regions where its magnitude is under 4. Put succinctly, **when the magnetic flux is a rational number p/q , the Bloch band splits into q subbands**. This statement is both alarming and intriguing. Small variations in α can lead to arbitrarily large changes in q , which seems at first glance to render the entire calculation unphysical.

As q grows, the width of the subbands become thinner and thinner. Irrational numbers can be (very) loosely thought of as rational numbers with infinitely large numerator and denominator, so we might guess that the spectrum for irrational α will consist of infinitely-many subbands with 0 length, perhaps somehow equivalent to a Cantor set. This guess turns out to be true, but it was not easy to prove [103].

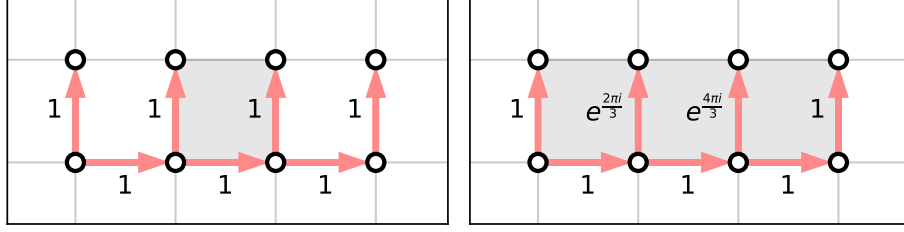


Figure 4.2: **Magnetic BZ. Left:** the unit cell is shaded and some hopping terms are shown for $\alpha = 0$. **Right:** $\alpha = 1/3$, each hopping term in y picks up a phase of $\exp(2\pi i/3)$ compared to its neighbor on the left, and so the magnetic unit cell is tripled in size in x but not in y .

Magnetic BZ, Bloch's theorem

Returning to equation (4.3): if α is rational, then the matrix elements of the Hamiltonian repeat after q steps. This motivates us to define a larger magnetic unit cell, the same size in y but q times larger in x , in which we can apply Bloch's theorem, as demonstrated in Fig. 4.2. We then solve the problem in reciprocal space in the magnetic Brillouin zone $0 \leq k_x < 2\pi/aq$, $0 \leq k_y < 2\pi/a$.

To solve this easily, we set q to some large prime number so that $k_x \approx 0$, and then sweep p from 1 to $q - 1$, solving the associated eigenvalue equation for each value of p/q . The Hamiltonian for each p/q is a $q \times q$ matrix with the form

$$\hat{H} = \begin{pmatrix} V_0 & 1 & 0 & 0 & \cdots & 0 & 1 \\ 1 & V_1 & 1 & 0 & \cdots & 0 & 0 \\ 0 & 1 & V_2 & 1 & \cdots & 0 & 0 \\ 0 & 0 & 1 & V_3 & \cdots & 0 & 0 \\ \vdots & \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \cdots & V_{q-2} & 1 \\ 1 & 0 & 0 & 0 & \cdots & 1 & V_{q-1} \end{pmatrix} \quad (4.10)$$

for $V_m = 2 \cos(2\pi mp/q + k_y)$. This matrix can be numerically solved by a computer in $\mathcal{O}(q^3)$ time, and the resulting plot is shown in Fig. 4.3. A few notes on symmetries:

- We only show the spectrum up to $\alpha = 1$. The spectrum repeats indefinitely for

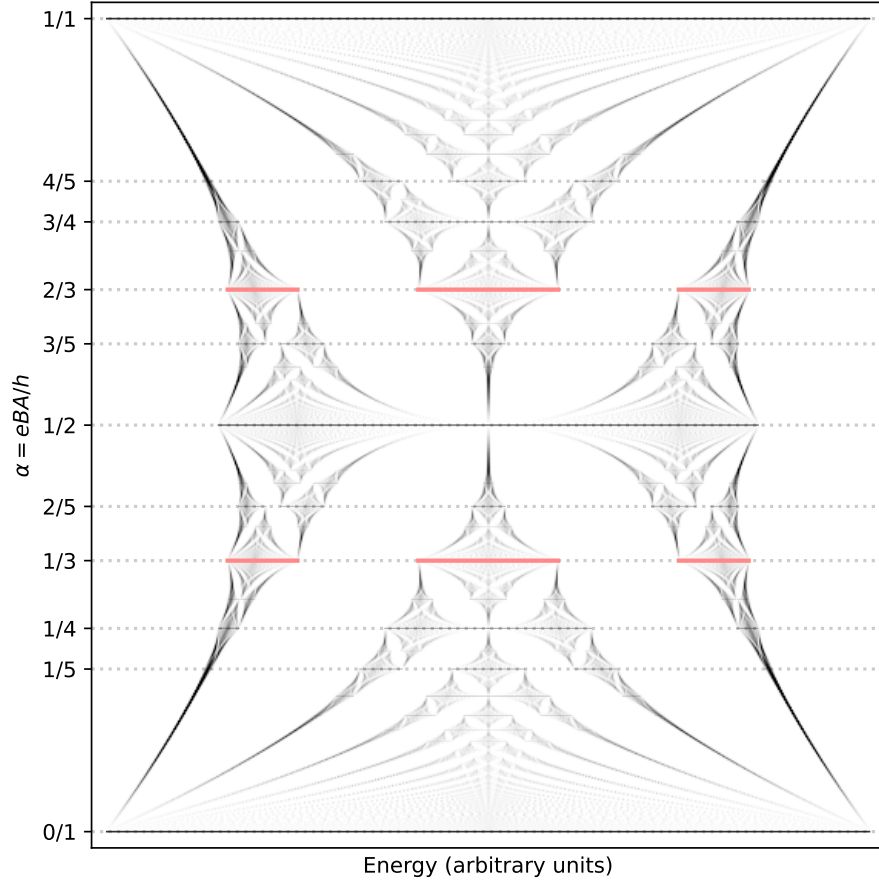


Figure 4.3: **Hofstadter's butterfly.** Greyscale image of the density of states for $q = 997$, with solutions for $q < 30$ using Hofstadter's original computation method overlaid. $\alpha = 1/3$ and $2/3$ are highlighted in red to make clear that there are 3 subbands when $q = 3$.

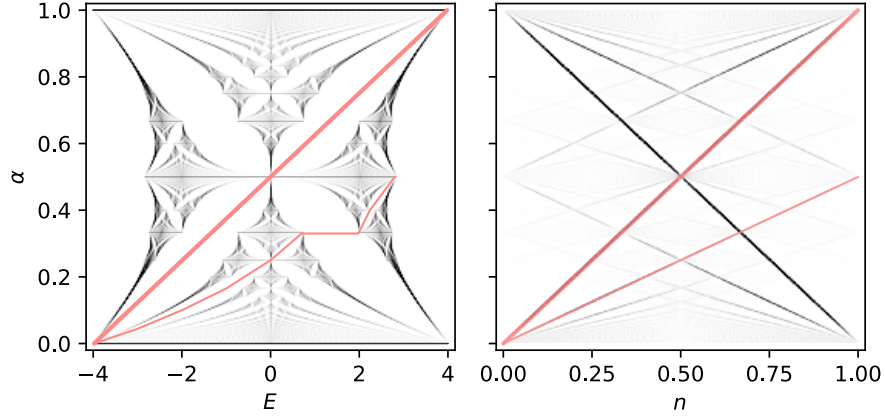


Figure 4.4: **Wannier plot. Left:** The butterfly from Fig. 4.3, with two large gaps indicated. **Right:** The inverse density of states $d\mu/dn$, with the same two gaps highlighted. I apply a fine Gaussian filter ($\sigma \approx 1/q$) to the density of states. Without this filter, the gaps are so fine that there are aliasing artifacts when the image is downsampled.

higher α .

- The direction of the field does not matter; the spectra are symmetric about the line $\alpha = 0$.
- The lines $E = 0$ and $\alpha = 1/2$ are axes of reflection.

Wannier plots

At $\alpha = p/q$, the Bloch band splits into q subbands. Each of these subbands has the same integrated density of states as the others (Ref. [104], but you might guess this based on the intuition that the magnetosubbands are an extension of Landau levels, which themselves have equal degeneracies), so if the total number of states in the band is equal to 1, then each subband has $1/q$ states.

Curiously, the gaps between states have a distinctly continuous appearance. If we follow these gaps along and keep track of the number of states with lower energy, then we recover behavior that does not appear to depend on the rationality of the magnetic field. Instead, the gaps between subbands follow straight lines in density/field space

(Fig. 4.4). This was first pointed out by Gregory Wannier in 1978 [30]. This gives us the Diophantine relation for gaps between magnetosubbands

$$n = t\alpha + s \quad (4.11)$$

for integers s and t . Because I control density with coplanar gates, and not energy, this Wannier plot (along with the quantized Hall resistance, covered shortly) is the fundamental prediction of the butterfly in transport measurements. It is a beautiful thing that such a complicated fractal spectrum yields such a simple physical description: each gap is labelled by only two integers.

Other simple lattices

The triangular lattice is distinct from the square lattice in that each lattice point has six neighbors, rather than four. The calculation of the spectrum in a magnetic field follows the same course. We end up with

$$\begin{aligned} \hat{H} = & 2 \sum_m [\cos(2\pi\alpha m) |m\rangle \langle m| \\ & + \cos(\pi\alpha(m + 1/2)) |m + 1\rangle \langle m| \\ & + \cos(\pi\alpha(m - 1/2)) |m - 1\rangle \langle m|]. \end{aligned}$$

The honeycomb lattice can be thought of as either two overlaid triangular sublattices, or as a single triangular lattice with two atoms in its unit cell. Each site is adjacent to three sites from the opposite sublattice. We find

$$\begin{aligned} \hat{H} = & \sum_m [\exp(2\pi i\alpha m/3) |m, B\rangle \langle m, A| \\ & + \exp(-\pi i\alpha(m + 1/2)/3) |m + 1, B\rangle \langle m, A| \\ & + \exp(-\pi i\alpha(m - 1/2)/3) |m - 1, B\rangle \langle m, A|] + \text{h.c.}, \end{aligned}$$

where A and B index the sublattice. The Hamiltonian has now doubled in size, but otherwise it is easy to compute.

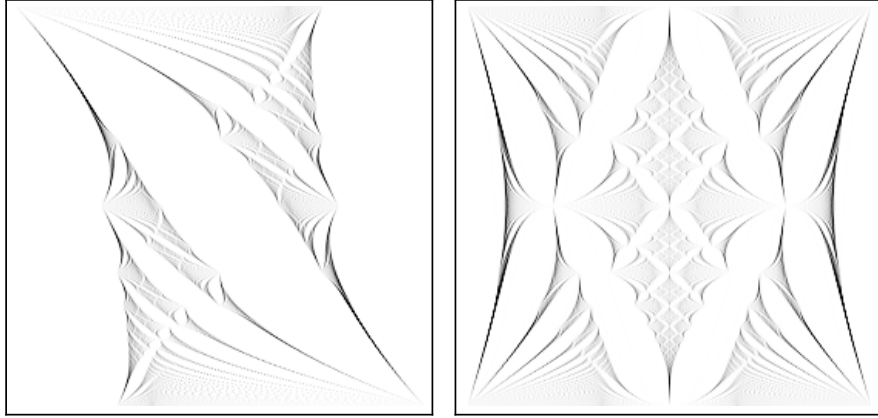


Figure 4.5: **Butterflies.** Triangular lattice (**left**) and honeycomb lattice (**right**).

The solutions for the triangular lattice and honeycomb lattice have the same butterfly-like appearance (Fig. 4.5), but the details of each are different. The triangular lattice looks like it has only one wing, and the spectrum is not symmetric about $1/2$ field. The honeycomb lattice looks like two butterflies side-by-side, with a zeroth Landau level projecting upwards from half-filling and 0 field. The Wannier plots for the two lattices again have Diophantine gaps, however the presence of specific gaps and the relative sizes of the gaps are distinct from the square lattice and each other.

Connection to the IQHE

The gaps in the butterfly spectra follow straight lines given by the Diophantine equation $n = t\alpha + s$ for integers t and s . The number t is, somewhat surprisingly, proportional to the Hall conductivity of the system when the Fermi energy is tuned into the respective gap:

$$\sigma_H = \frac{te^2}{h}. \quad (4.12)$$

This fact, along with an algorithm for computing the numbers t and s for a given gap at a given field,² was derived in a now-famous paper by Thouless, Kohmoto, Nightingale, and den Nijs, typically referred to as TKNN [31].

²Along with a very important lemma relating the Hall conductivity to a particular integral around the Brillouin zone.

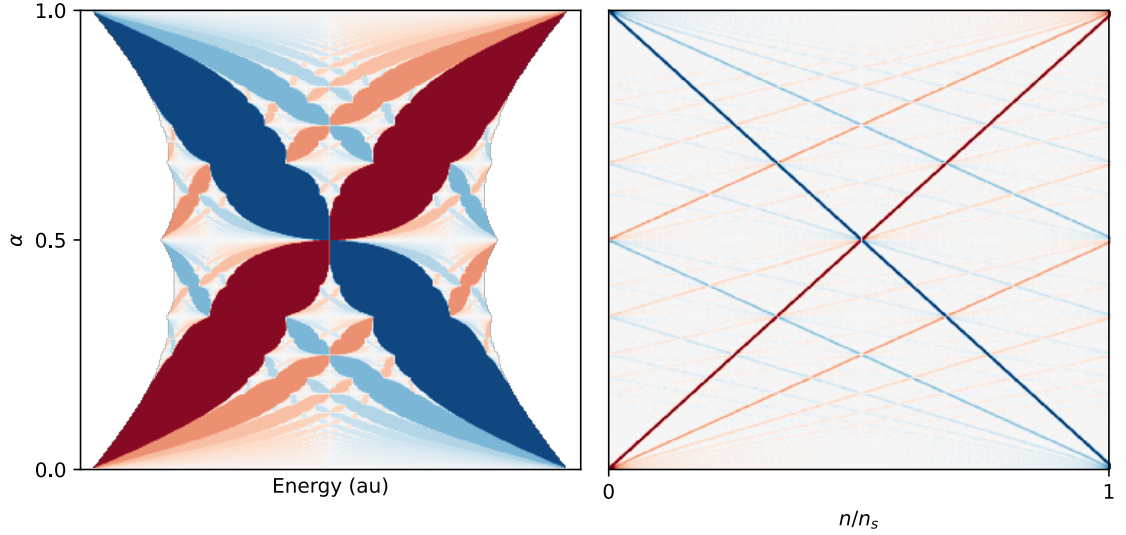


Figure 4.6: **Hall resistance of butterfly gaps.** Spectrum (**left**) and associated Wannier plot (**right**), with t in each gap labelled by color, red positive, blue negative.

The algorithm for computing s and t is simple. For $\alpha = p/q$, there are q subbands with $q - 1$ gaps between them. For the r -th gap, solve the diophantine equation

$$r = s_r q + t_r p \quad (4.13)$$

for integer r with $r \leq 1 < q$ and $|t_r| \leq q/2$. For instance, at a field of $p/q = 2/5$ and $r = 1$, the equation $1 = 5s + 2t$ has solution $s = 1$ and $t = -2$. So, the Hall conductivity within that gap is $-2e^2/h$. Computing these gaps over the entire butterfly yields Fig. 4.6.

4.2 Two ingredients added

I was able to reproduce the splitting and bending behavior of my Landau fan diagram with a surprisingly simple extension to the Hofstadter's butterfly model described in the previous section.

Rather than attempting to modify the standard BM continuum model of twisted bilayer graphene [105, 106], I made my calculation in a simple triangular lattice.

Therefore, the exact details of the calculation did not match the details in the TBG device. For instance, degeneracies from spins and valleys were not captured. Instead, I showed that these splitting and bending features are generic properties of Hofstadter-like models. I separately verified that the same behavior arised in square lattices, honeycomb lattices, and eventually the BM continuum model (Chapter 6).

Start with Eq. 4.1, written more concisely as

$$H = \sum_{\langle i,j \rangle} a_{ij} |i\rangle \langle j| + \text{h.c.} \quad (4.14)$$

where $i, j \in \mathbb{Z}$ index lattice sites. Each unit cell contains three bonds with associated hopping amplitudes a_1 , a_2 , and a_3 . Augment the Hamiltonian by allowing the hopping amplitudes to be different along the three symmetric directions (a_1 , a_2 , and a_3 not all equal), and add a second Fermion species with a tunable energy splitting V , yielding

$$H = \sum_{\alpha \in \{A,B\}} \sum_{\langle i,j \rangle} a_{ij} |i\alpha\rangle \langle j\alpha| + V \sum_i (|iA\rangle \langle iA| - |iB\rangle \langle iB|) + \text{h.c.} \quad (4.15)$$

In the following text and figures, I set $a_2 = a_3 = 1$. The specific values of a_1 and V that I show are not motivated by any microscopic details of the system, but are instead chosen for their phenomenological match to the data. Fig. 4.7 shows spectra and the corresponding inverse density of states for several values of a_1 and V .

The spectrum for $a_1 = a_2 = a_3 = 1$ is identical to the classic isotropic butterfly, and the corresponding inverse density of states demonstrates clear Diophantine behavior (leftmost panels). However, as there are two fermion species, each state is doubly degenerate and the gaps follow even-integer slopes only.

Anisotropy has previously been shown to smear out the energy levels and partially close the gaps in the spectrum [31, 107–110], which I reproduce by tuning a_1 away from a_2 and a_3 (second panels from the left). Note that although the gaps partially close, they do not entirely close.³

³For a square lattice with two hopping terms a_1 and a_2 , the size of the r -th gap goes as

$$\Delta E_r \sim \left| \frac{a_1}{a_2} \right|^{|s_r|}, \quad (4.16)$$

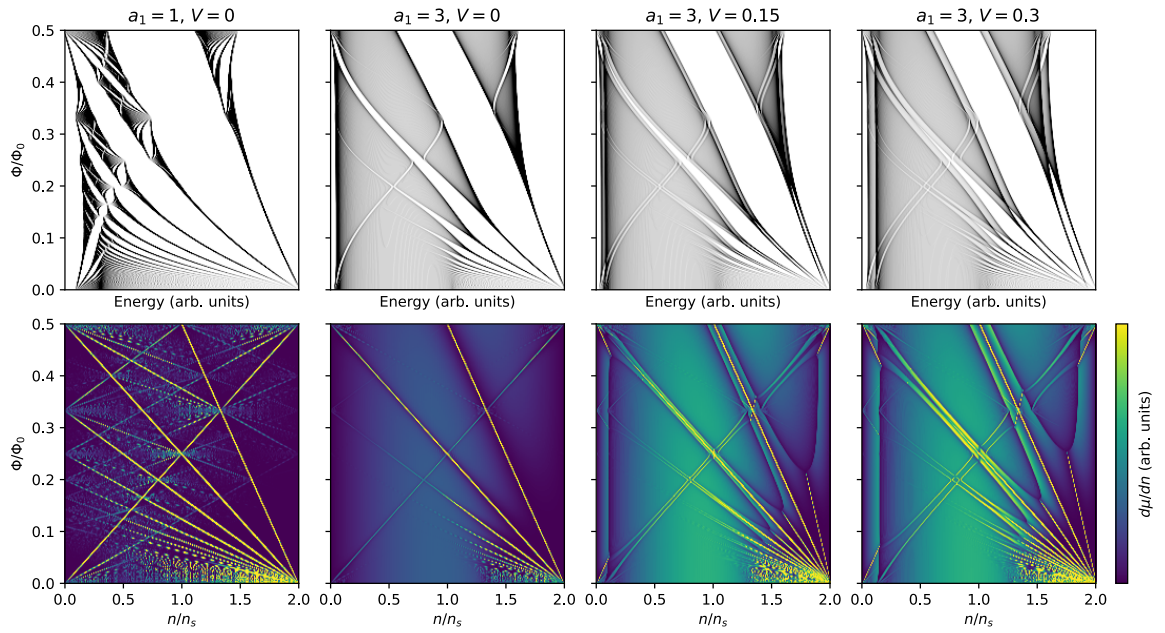


Figure 4.7: **Model behavior overview.** Described in detail in the text. Spectra (**top row**) and inverse density of states (**bottom row**) for the indicated model parameters, for $q = 1999$. n/n_s runs from 0 to 2 to account for the two fermion species.

Upon introducing a small amount of V , a second butterfly pattern appears (second panels from the right, and rightmost panels). At low fields and low densities the two butterflies are almost parallel and seldom overlap, and every integer filling of Landau levels gives a ground state with a gap for excitations. However, at higher fields and energies, the anisotropy-broadened butterflies overlap, and odd-integer Landau level fillings have no gap to excitations.

The even-integer Landau level gaps appear to split and bend in the same way as the measured Landau level gaps in my device (Fig. 4.8). This complicated intersection behavior from my model is not difficult to understand, however it is somewhat intricate. Begin by noting that many of the gaps in Hofstadter's butterfly take horizontal steps—a discontinuity in energy at a given magnetic field—when they cross gaps moving the other direction (Fig. 4.9, left panels). When the second butterfly appears out of the first ($V \neq 0$), it is shifted horizontally in energy. There are then then effectively two Xs next to each other. When two upper or two lower split LL gaps overlap, the system is simultaneously in the gap of both butterflies, and so the gap follows the average (s, t) of the two intersections. When an upper meets a lower split LL gap, it is always the intersection of LL gaps within a single butterfly. Therefore, it happens at the appropriate field for that intersection. As there is a background DOS from the other butterfly, there is a horizontal step in energy, that is present but not visible in Wannier plots of undoubled lattices.

Note that this detailed behavior at the intersection of Landau level gaps does not depend on the underlying lattice. Any doubled butterfly with anisotropy will show the same behavior.

Behavior of the split LL gaps for an array of a_1 and V are shown in Fig. A.21. Of note, increasing V with no anisotropy does not give split, bending LLs. Instead, it retains perfect Diophantine behavior, but changes the particular values of s and t that are visible. Both ingredients are necessary to see what I see in my measurements. The shape of the split LL gaps is in rough agreement with my experiment for the range of parameters $1.5 < a_1 < 4$ and $0.1 < V < 0.4$.

I left the nature of the energy splitting term V general. One would expect that where r and s_r are defined as in Eq. 4.13 [31].

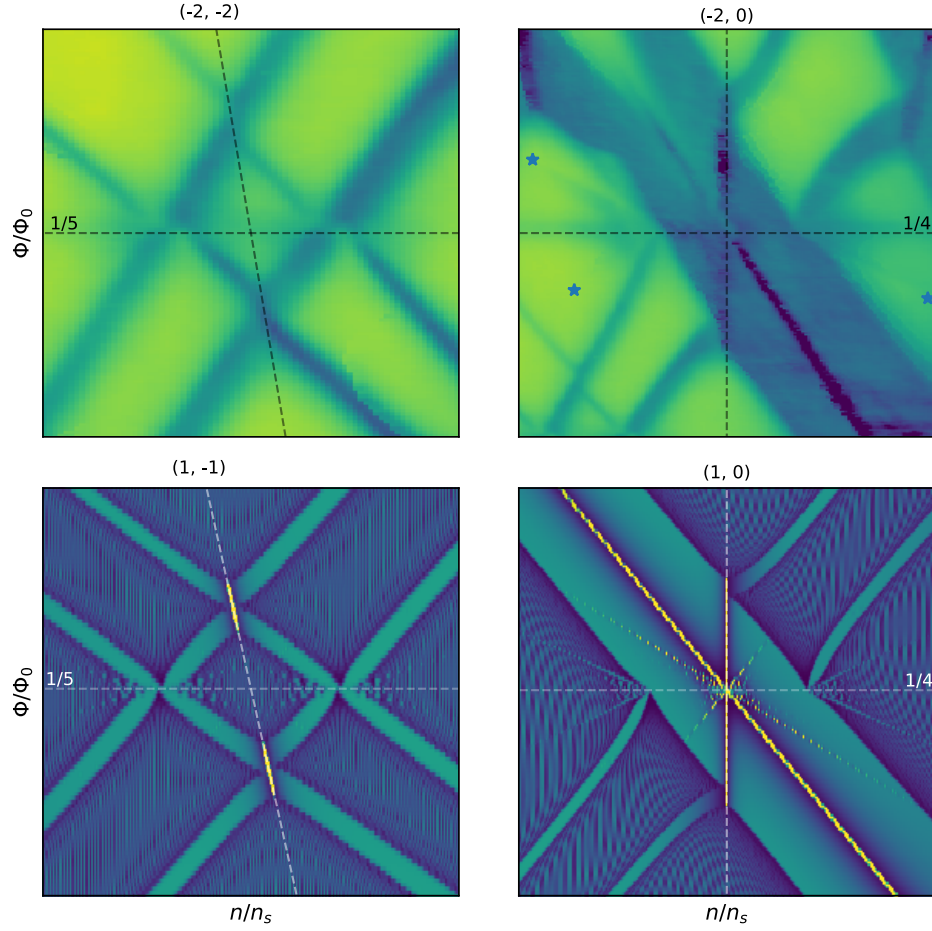


Figure 4.8: **Split Landau level overlap behavior in experiment and computation.** (**Top left:**) detail of the crossing of split LLs -12 from charge neutrality ($s, t = 0, -12$) and $+8$ from $n/n_s = -4$ ($s, t = -4, 8$), and (**top right**) -8 from charge neutrality and $+8$ from $n/n_s = -4$. The horizontal lines are at the indicated Φ/Φ_0 , and the lines with steep slopes are the average (s, t) of the crossing LLs. For the case of the top left panel, this is the average of $(0, -12)$ and $(-4, 8)$ which is $(-2, -2)$ as indicated. (**Bottom left:**) computed inverse density of states for $q = 2999$, $a_1 = 3$, $a_2 = a_3 = 1$, and $V = 0.3$ near the crossing of the split levels $s, t = 2, -6$ and $0, 4$, and (**bottom right**) $s, t = 0, 4$ and $2, -4$. Note that if I were to add in an additional factor of two to s and t , to account for an additional degeneracy, then the values of s and t in the bottom two panels would match those in the top two panels after shifting the zero-point of the density. The color scale for the bottom panels is as in Fig. 4.7. Stars in the top right panel indicate the ends of the faint “extra” LLs originating from the intersections of lower (upper) with upper (lower) split LLs. These features are clearly reproduced in computation in the bottom right panel. There are moiré patterns visible in the bottom panels where the Landau levels overlap, which is nice.

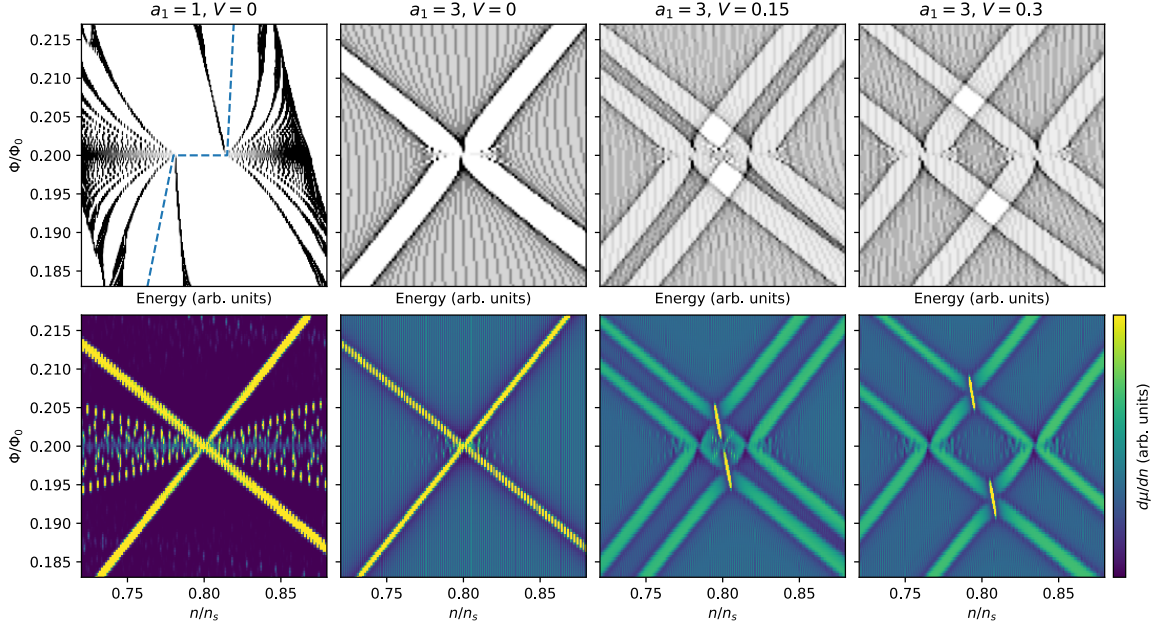


Figure 4.9: **Split LL detail in the model.** Spectra (**top rows**) and associated inverse density of states (**bottom rows**) for $q = 2999$, zoomed in to the intersection of LLs $s, t = 4, -6$. The gap is indicated with the dashed line.

the ordinary spin Zeeman effect would give rise to such a splitting term as a linear function of B , $V = g\mu_B B$, for Landé factor $g \approx 2$ and Bohr magneton μ_B . I found that in the model, $V = \Phi/\Phi_0 \approx 0.02B/\text{Tesla}$ gives resulting behavior that is roughly equivalent to the fit results (Fig. A.22). The bandwidth of TBG at 1.4° is roughly 100 meV, and the bandwidth of the model for $a_1 = 3$ and $V = 0$ is roughly 16 (arb. units). $2\mu_B(16)/(100 \text{ meV}) \approx 0.02\text{T}^{-1}$. That such a rough calculation gives approximate agreement is strong evidence that spin Zeeman is the correct physical description of the system.⁴

There is one notable but subtle difference between constant- V and Zeeman-like models: At low fields near charge neutrality and full filling, the constant- V Landau levels from the two butterflies cross each other, whereas in the Zeeman-like model the two butterflies are not split at low field and thus cannot cross at low field. These crossings lead in the constant V model to the disappearance and reappearance of

⁴Refs. [111, 112] describe a *valley*-Zeeman effect with $g \sim 2$ at $3/4$ filling in TBG with aligned hBN, however I do not believe that this is relevant to my device.

gaps along the density/field trajectory of a given Landau level, as seen in Fig. 3.14 (right panel). This is evidence for a constant- V model, so I chose to leave the model agnostic to the origin of the V term. Later, I learned that spin Zeeman was indeed the most reasonable term, and the LLs crossings at low density have another root cause (Chapter 6).

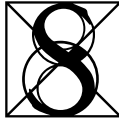
Ultimately, I published the measurement and the toy model in Ref. [64]. The paper left two major questions unanswered:

- What caused the large magnetoresistance? I noticed that the large magnetoresistance appeared to correlate with regions of density/field where the two butterflies overlapped, however it was not clear why this should lead to magnetoresistance. As noted previously, I found the compensated semimetal picture unlikely. Perhaps it was related to proximity to the van Hove singularity near half filling⁵, as described in Ref. [68]?
- Assuming there really was anisotropy in the sample, what causes it? It was known (in theory [63, 113–117] and experiment [53, 118–120]) that TBG and twisted double bilayer graphene near the magic angle displayed signatures of nematic states. One would expect that nematic order would accompany an anisotropic effective Hamiltonian. I did not believe that this explanation was relevant, simply because I saw the unusual magnetotransport at large enough angles such that the electron interactions were too weak for nematic order to exist. Instead, I believed that uniaxial strain, either in one layer or both, was a more likely explanation.

⁵I later discovered that the sign-change singularity in the Hall slope near half-filling of the moiré was *not* associated with a van Hove singularity, a surprise to us. See Chapter 5.

Chapter 5

More than a toy model: low field



SEVERAL months after publishing my unusual magnetotransport measurements alongside my toy model, theorists Oskar Vafek and Xiaoyu Wang contacted us with wonderful news. They had augmented the standard Bistritzer-MacDonald (BM) model to include heterostrain, and they found features within the resulting band structure that they believed explained the wide density-ranges of quadratic magnetoresistance within my measurements. Over the next months, I worked closely with them to compare my experimental results with their model. We found surprisingly excellent agreement in many details, and published the results in Ref. [65].

While we found the effects of homostrain—equal strain between the two layers—to be small, the effects of heterostrain on twisted moiré patterns are “magnified” by a factor of $1/\theta$ [121, 122]. This means that even miniscule heterostrains of 0.1 to 0.5%, amounts typically observed in twisted bilayer graphene devices with scanning measurements [53–56], distort the moiré pattern by 5 to 25%.

In particular, uniaxial heterostrain distorts the moiré pattern by breaking C_{3z} symmetry: the lengths of the triangular unit cell vectors become unequal. This is known to modify the bands [62] and interacting phase diagram [63] of magic-angle TBG. Above the magic angle, uniaxial heterostrain breaks the degeneracies of the three van Hove points. At each of these van Hove points, the Fermi surface topology changes. Notably, between two of the van Hove points, the theory predicts open

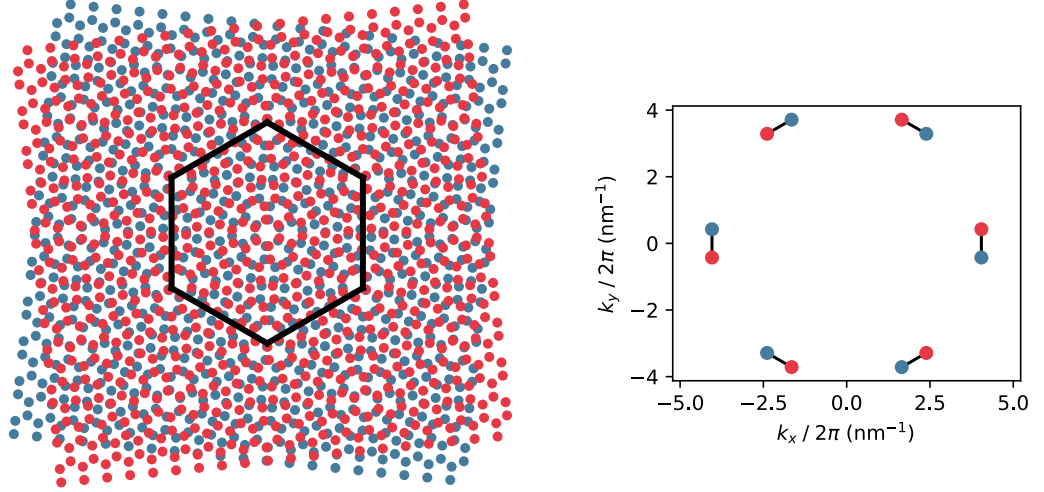


Figure 5.1: **Basic idea of moiré patterns.** **Left:** real-space moiré caused by 12° interlayer twist. **Right:** k -space description of the same. The black lines between the two lattice's wavevectors are the moiré wavevectors.

Fermi surface contours—a known source of quadratic magnetoresistance!

We carefully compared experimental results to a semiclassical Boltzmann transport calculation, and found that in addition to the two van Hove points bounding the quadratic magnetoresistance regions, we had strong evidence for the third van Hove point closer to CNP.

Thus, we were able to answer the open questions from my first publication—the origins of the anisotropy and magnetoresistance—with uniaxial heterostrain.

5.1 Effects of strain on twisted moiré patterns

Assume homogeneous lattices without relaxation. Twisted bilayer graphene's moiré pattern arises from the following parameters: global rotation angle (θ_g), interlayer twist angle (θ), and strain tensors (biaxial amount ϵ_{bi} , uniaxial amount ϵ_{uni} , and uniaxial direction ϕ for both homo and heterostrain). I will describe the effects of each of these parameters in turn.

To compute the properties of moiré patterns, start with two copies of the same lattice. Apply the appropriate transformations based on the above parameters, which will distort the two lattices. In Fourier space, they will now be slightly offset from each other. The differences between the corresponding wavevectors of the two lattices are the moiré pattern's wavevectors (Fig. 5.1, right panel).

Interlayer twist angle in real space maps to a twist in k space. The relevant transformation is

$$\begin{aligned} L_1 &= R(\theta/2) \\ L_2 &= R(-\theta/2), \end{aligned} \tag{5.1}$$

where L_1 and L_2 are the two layers and $R(\theta)$ is the standard 2D rotation matrix. An example is shown in Fig. 5.1. The unit cell area is proportional to θ^{-2} (Eq. 2.4). From now I will consider the effect of adding transformations on top of the interlayer twist angle.

Global angle θ_g appears as

$$\begin{aligned} L_1 &= R(\theta/2)R(\theta_g) \\ L_2 &= R(-\theta/2)R(\theta_g). \end{aligned} \tag{5.2}$$

It directly sets the overall angle of the moiré pattern in real-space. This is more interesting than it may seem, because it means that measurements of the moiré pattern's rotational angle give information about the underlying lattices' absolute angles.

The homostrain tensor applies equally to both layers as

$$\begin{aligned} L_1 &= R(\theta/2)E(\epsilon_{bi}, \epsilon_{uni}, \phi) \\ L_2 &= R(-\theta/2)E(\epsilon_{bi}, \epsilon_{uni}, \phi) \end{aligned} \tag{5.3}$$

for

$$E(\epsilon_{bi}, \epsilon_{uni}, \phi) = R^T(\phi) \begin{pmatrix} \frac{1}{1+\epsilon_{uni}} & 0 \\ 0 & \frac{1}{1-\nu\epsilon_{uni}} \end{pmatrix} R(\phi) \begin{pmatrix} \frac{1}{1+\epsilon_{bi}} & 0 \\ 0 & \frac{1}{1+\epsilon_{bi}} \end{pmatrix}, \tag{5.4}$$

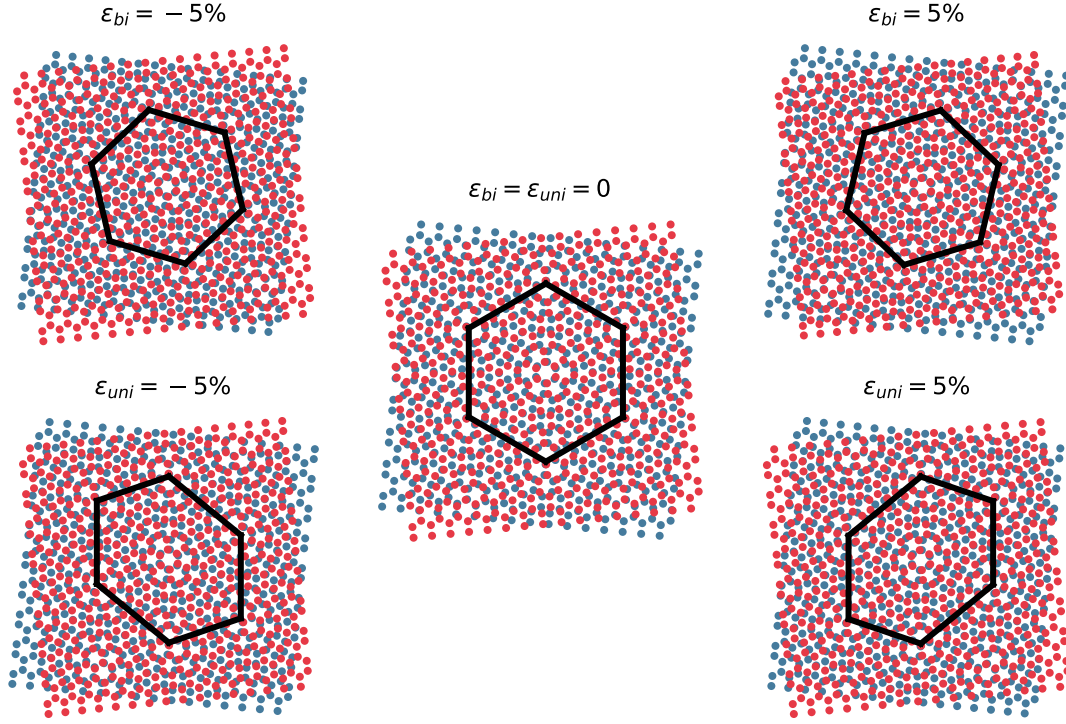


Figure 5.2: **Heterostrain's effect on twisted moiré patterns.** Indicated amounts of heterostrain for 12° twist angle, $\phi = 0$. Biaxial heterostrain rotates the moiré, while uniaxial heterostrain shears it.

where $\nu \approx 0.16$ is the Poisson ratio [53]. Biaxial homostrain modifies the unit cell area like $A \rightarrow A(1 + \epsilon_{bi})^2$, without distorting or rotating it. Uniaxial homostrain squeezes it in the direction set by ϕ and stretches it in the perpendicular direction, changing the area like $A \rightarrow A(1 + \epsilon_{uni})(1 - \nu\epsilon_{uni})$. Homostrains of even few percent are typically ignored, because they only affect the moiré pattern by a few percent. This is not the case for heterostrains.

The heterostrain tensor applies opposite in both layers:

$$\begin{aligned} L_1 &= R(\theta/2)E(\epsilon_{bi}/2, \epsilon_{uni}/2, \phi) \\ L_2 &= R(-\theta/2)E(-\epsilon_{bi}/2, -\epsilon_{uni}/2, \phi). \end{aligned} \tag{5.5}$$

Its effect on the moiré is more intricate than the effect of the homostrain tensor. See

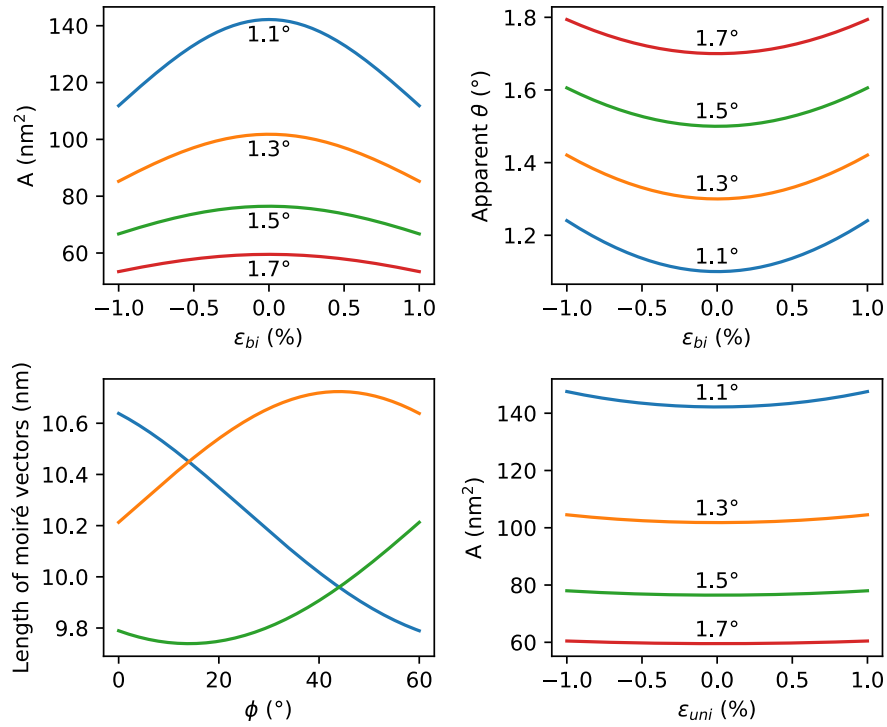


Figure 5.3: **Heterostrain's effect on moiré pattern parameters.** Unit cell areas (**top left and bottom right**) vs strain amounts at various twist angles. Uniaxial strain direction has no impact on the area. **Top right:** apparent θ if we apply the formula with no strain to a strained moiré. **Bottom left:** lengths of moiré unit cell vectors as a function of ϕ for $\theta = 1.38^\circ$ and $\epsilon_{uni} = 0.2\%$.

Fig. 5.2 for an example.

Biaxial heterostrain displaces the lattice vectors of one layer towards or away from the origin in k space. Its effect is perpendicular to the wavevectors from twist angle, so it rotates the moiré, like

$$\theta_g \rightarrow \theta_g + \frac{\epsilon_{bi}}{\theta}. \quad (5.6)$$

The twist angle is a small number, so even small amounts of biaxial heterostrain have a magnified effect on the moiré. For intuition, $1\%/1^\circ \approx 0.6 \approx 33^\circ$, so a 0.3% biaxial heterostrain rotates a 1° moiré by almost 10° . Biaxial heterostrain changes the area by a factor of order ϵ_{bi}^2/θ^2 . So, biaxial heterostrain presents as a twist angle and a global angle. Because of this, it cannot be inferred independently with knowledge of only the moiré pattern's lattice vectors.

In transport, we infer the twist angle from the unit cell area assuming 0 strain, however, there is typically biaxial heterostrain of 0.1 to 0.5% in measurements that can resolve individual atoms [53–56]. So, the twist angle figures that we report should have uncertainties of at least 0.05° for samples at and above magic angle, even though we can measure the moiré unit cell area with significantly less fractional uncertainty than that (Fig. 5.3, top row).

Uniaxial heterostrain distorts the shape of the moiré, but not in the same way as uniaxial homostrain does. Instead, it has a shear effect (Fig. 5.2 bottom panels). This effect dramatically changes the lengths of the moiré unit cell vectors, again of order ϵ_{uni}/θ (Fig. 5.3 bottom left panel). This means that scanning measurements that can resolve the moiré unit cell structure will observe three different moiré wavelengths, and thus cannot easily back out a twist angle according to Eq. 1.9.

Surprisingly, uniaxial heterostrain does not affect the area of the moiré unit cell to first order. I say surprisingly, because it does affect the lengths of the unit cell vectors. This is possible because it is a shear transformation.¹ Its effect on the area is only caused by the fact that the Poisson ratio leads to a small biaxial heterostrain, so the area changes like $\nu^2 \epsilon_{uni}^2/\theta^2$.

¹Intuitively, a square and a parallelogram with the same base and height have the same areas, despite the parallelogram having arbitrarily longer side lengths.

5.2 Energetics

5.2.1 The BM model with heterostrain

The energetics of twisted bilayer graphene is relatively straightforward to compute within a continuum model [35]. I will describe the model with uniaxial heterostrain, as parameterized in the previous section. This was initially worked out by Xiaoyu Wang and Oskar Vafek in [65].

We choose reciprocal lattice vectors for unstrained graphene layers

$$\mathcal{G}_1 = \frac{4\pi}{\sqrt{3}a}(0, -1) \quad \mathcal{G}_2 = \frac{4\pi}{\sqrt{3}a}\left(\frac{\sqrt{3}}{2}, \frac{1}{2}\right). \quad (5.7)$$

Then, the moiré reciprocal lattice vectors are $\mathbf{g}_i = \mathcal{E}^T \mathcal{G}_i$ for $\mathcal{E}_t = -\mathcal{E}_b \equiv \mathcal{E}/2$, where $\mathcal{E} \equiv \mathcal{T}(\theta) + \mathcal{S}(\epsilon, \phi)$, where

$$\mathcal{T}(\theta) = \begin{pmatrix} 0 & -\theta \\ \theta & 0 \end{pmatrix}, \quad \mathcal{S}(\epsilon, \phi) = R_\phi^T \begin{pmatrix} -\epsilon & 0 \\ 0 & \nu\epsilon \end{pmatrix} R_\phi. \quad (5.8)$$

The Hamiltonian for a given valley (omitting spin) is

$$\hat{H}_\eta = \left(\sum_{l=t,b} \hat{H}_{\eta,l}^{\text{intra}} \right) + \hat{H}_\eta^{\text{inter}} \quad (5.9)$$

where $\eta = \pm 1$ labels $\mathbf{K}$ and $\mathbf{K}'$ valleys and $l = t, b$ is the layer. The intra layer Hamiltonian is given by

$$\hat{H}_{\eta,l}^{\text{intra}} = \int d^2\mathbf{r} \psi_{\eta,l}^\dagger(\mathbf{r}) \left[\alpha(\text{tr}[\mathcal{E}_l]\mathbf{I}) - \frac{\hbar v_F}{a} [(-i\nabla - \mathbf{A}_{\eta,l}) \cdot (\eta\sigma_x, \sigma_y)] \right] \psi_{\eta,l}(\mathbf{r}). \quad (5.10)$$

The first term in brackets is known as the deformation potential. Values of α ($\alpha = -dW/d\epsilon$, the change in work function w.r.t strain) in the literature range from -4.1 eV to 30 eV [123–126], depending on how it is measured. We use -4.1 eV, the result

from [126].² $\mathbf{A}_{\eta,l}$ is the pseudovector potential,³ given by [127, 128]

$$\mathbf{A}_{\eta,l} = \frac{\sqrt{3}\beta\eta}{2a}(\epsilon_{l,xx} - \epsilon_{l,yy}, -2\epsilon_{l,xy}) \quad (5.11)$$

where $\beta \approx 3.14$ [62, 128]⁴ and the derivatives of ϵ are

$$\begin{aligned} \epsilon_{xx} &= -\epsilon \cos(\phi)^2 + \nu \epsilon \sin(\phi)^2 \\ \epsilon_{yy} &= \nu \epsilon \cos(\phi)^2 - \epsilon \sin(\phi)^2 \\ \epsilon_{xy} &= (1 + \nu) \epsilon \cos(\phi) \sin(\phi). \end{aligned} \quad (5.12)$$

Note that in the absence of strain, the intra layer Hamiltonian simplifies to the ordinary single layer graphene Hamiltonian of Eq. 1.7. From here, we set $\hbar v_F/a = 2680$ meV based on monolayer graphene's Fermi velocity.

The form of the interlayer Hamiltonian is unchanged from the ordinary BM model:

$$\hat{H}_{\eta}^{\text{inter}} = \int d^2\mathbf{r} \psi_{\eta,t}^{\dagger}(\mathbf{r}) \left(\sum_{j=1,2,3} T_{\eta,j} e^{-i\eta\mathbf{q}_j \cdot \mathbf{r}} \right) \psi_{\eta,b}(\mathbf{r}) + \text{h.c.}, \quad (5.13)$$

where the T matrices can be concisely written as

$$T_{\eta,j} = w_0 \mathbf{I} + w_1 \left(\cos \frac{2\pi(j-1)}{3} \boldsymbol{\sigma}_x + \eta \sin \frac{2\pi(j-1)}{3} \boldsymbol{\sigma}_y \right). \quad (5.14)$$

We take $w_0 = 88$ meV and $w_1 = 110$ meV to account for relaxation: AB stacking is a lower-energy configuration than AA.

To numerically compute the BM model, the matrix elements are Fourier transformed to k -space, and we fold in only a few nearest neighbors. Their contribution rapidly dies off because their distance from the Dirac points is large in the intralayer term.

²This choice of deformation potential value does not make a qualitative difference to the resulting calculations, however it does affect the quantitative results. Notably, it affects the amount of energetic splitting of the Dirac points.

³So, $\mathbf{B}_{\text{eff}} = \nabla \times \mathbf{A}$. This pseudomagnetic field can be up to 300 T in highly strained graphene [61].

⁴ β has no relation to π , but is instead the ratio of the tight-binding decay length of the transfer integral, roughly $0.184a$, and the nearest-neighbor bond length $a/\sqrt{3}$ [128].

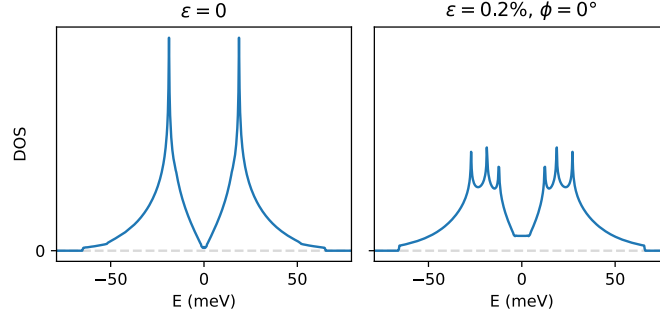


Figure 5.4: Uniaxial strain separates the van Hove points.

5.2.2 Effects of heterostrain on TBG energetics

The isolated moiré bands in TBG have six critical points: two Dirac points at K and K' , one band edge at the Γ point, and three van Hove points. That there are three van Hove points can be shown within Morse theory, a subfield of topology, wherein

$$\sum_i (-1)^{\gamma_i} = \chi, \quad (5.15)$$

where i indexes critical points, γ_i is the index⁵ of the i -th critical point, and χ is the Euler characteristic of the manifold, which is 0 for a torus. In order for a van Hove point to appear or disappear in response to a continuous deformation, it must do so by colliding with a band edge or Dirac point.

Fig. 5.4 shows the density of states of the moiré bands of $\theta = 1.4^\circ$ with no (left panel) and with 0.2% (right panel) uniaxial heterostrain. Two effects are evident: 1. the peak corresponding to the energetically degenerate van Hove points splits into three peaks, and 2. the Dirac points also energetically split, so the DOS does not go to 0 at 0 energy.

The full behavior of the DOS as a function of strain amount and angle is shown in Fig. 5.5. When a small amount of uniaxial heterostrain is present, rotating ϕ sinusoidally winds the three van Hove points around each other in energy.⁶ In density,

⁵The index of a critical point is the number of directions in which the function decreases from the point: 0 for minima, 1 for saddle points, and 2 for maxima.

⁶Note similarity with Fig. 5.3's bottom-left panel.

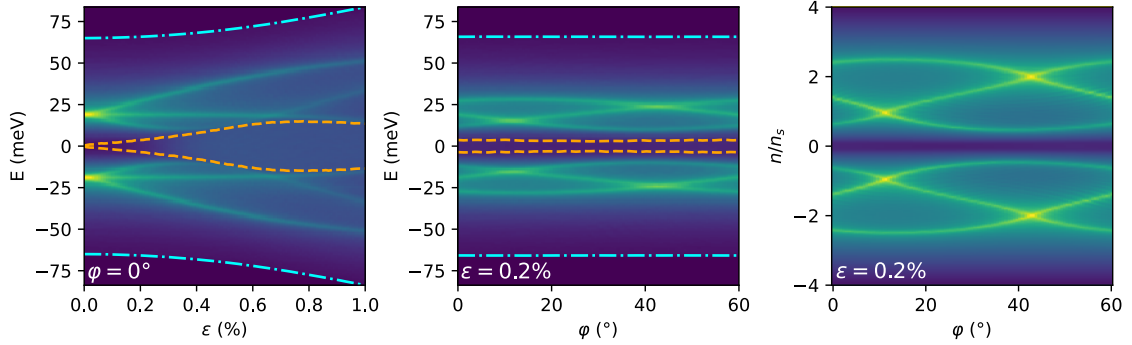


Figure 5.5: **Effect of uniaxial heterostrain on the van Hove points.** Color indicates density of states. Blue dash-dotted lines are isolated band edges, and orange dashed lines are the other band edges. **Left:** As strain is increased from zero, the three van Hove points energetically separate. **Middle:** For a fixed, small strain, varying ϕ causes the three van Hove points to sinusoidally wind around each other. **Right:** same as the middle panel, plotted as a function of density instead of energy.

their paths are not sinusoidal. At $\phi = 12^\circ$, the innermost van Hove points overlap, and at $\phi = 42^\circ$, the outermost two points overlap. At strain amounts above roughly 0.4%, the innermost van Hove point has collided with the Dirac point. This collision can lead to a type-II Dirac crossing, where the Dirac cone is tilted onto its side (Fig. A.23). The Fermi surface at a regular type-I Dirac crossing becomes a point. At a type-II crossing, a hole-like pocket touches an electron-like pocket. Such crossing are conjectured to exist in, for example, WTe_2 [129]. They can be directly observed with techniques such as ARPES,⁷ but they also have signatures in transport [131].

From here on, I refer to the van Hove points by their order: the first van Hove point is that that is energetically nearest to CNP, then the second, then the third is energetically nearest to the Γ point. Fig. 5.6 helps us understand the structure of the Fermi surface as energy is tuned through the van Hove points for a modest amount of uniaxial heterostrain. At energies below the first van Hove point, there are two Fermi surfaces of different size, one around each Dirac point, potentially of opposite carrier sign. Between the first and second van Hove points, there is one Fermi surface surrounding both Dirac points. Between the second and third van Hove points, the

⁷For example, [130], which found type-II crossings in LaAlGe .

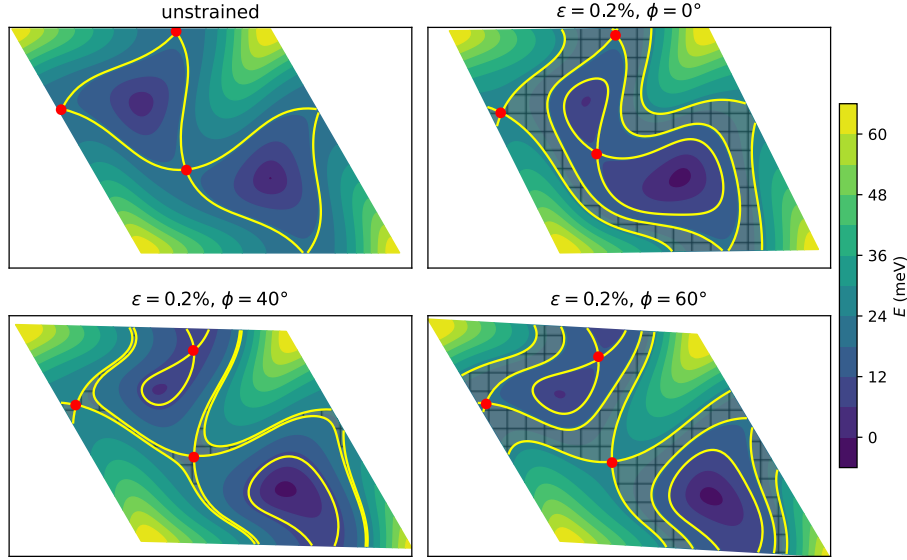


Figure 5.6: **Effect of uniaxial heterostrain on the Fermi surface topology.** Isolated moiré bands within the Brillouin zone for the indicated strain and strain angle. Color indicates energy. Red dots indicate van Hove points, with associated Fermi surface contours in yellow. Hatched regions feature open Fermi surfaces contours.

Fermi surfaces become open. Finally, above the third van Hove point, the Fermi surface closes once again around the Γ point. These properties hold as long as the strain amount is small enough that none of the van Hove points have contacted a band edge.

5.3 Boltzmann transport

Ultimately, we would like to use the band structure to compute transport properties. This is challenging to do in the general case, because we do not know the dominant scattering mechanisms in TBG. So, we choose a simple semiclassical model to start with: Boltzmann transport in the relaxation-time approximation. As with the strained BM calculation, this calculation was first worked out for the case of strained TBG by Oskar Vafek and Xiaoyu Wang.

We wish to find the non-equilibrium distribution n of electrons in $\mathbf{k}$ and $\mathbf{r}$ -space when subjected to an external Lorentz force. The force must be small enough that

it does not excite electrons to the neighboring bands, and the wave packets need to satisfy the uncertainty principle.

Liouville's theorem states that the total derivative of the distribution function with time is zero. Loosely,

$$\frac{dn}{dt} = \frac{\partial n}{\partial \mathbf{r}} \cdot \frac{d\mathbf{r}}{dt} + \frac{\partial n}{\partial \mathbf{k}} \cdot \frac{d\mathbf{k}}{dt} + \frac{\partial n}{\partial t} = 0. \quad (5.16)$$

$d\mathbf{r}/dt$ is the velocity, and $d\mathbf{k}/dt$ is the force $\mathbf{F}$. We assume the distribution is uniform in space, so $\partial n/\partial \mathbf{r} = 0$. Collisions do change the distribution function, which we incorporate in the “relaxation time” approximation,

$$\frac{dn}{dt} = \frac{\partial n}{\partial \mathbf{k}} \cdot \mathbf{F} + \frac{\partial n}{\partial t} = \left(\frac{\partial n}{\partial t} \right)_{\text{coll}} = \frac{n_0 - n}{\tau}, \quad (5.17)$$

where n_0 is the equilibrium Fermi-Dirac distribution and τ is the relaxation time. This is the starting point of the calculation that follows.

First, some bookkeeping. The resistivity tensor is defined as

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix} = \begin{pmatrix} \rho_{xx} & \rho_{xy} \\ \rho_{yx} & \rho_{yy} \end{pmatrix} \begin{pmatrix} j_x \\ j_y \end{pmatrix}, \quad (5.18)$$

where $\mathbf{E} = (E_x, E_y)$ and $\mathbf{j} = (j_x, j_y)$ are electric field and current vectors. Uniaxial heterostrain breaks the point group symmetry down to C_{2z} . As a consequence of this, ρ_{xx} may not equal ρ_{yy} , and ρ_{xy} may not equal $-\rho_{yx}$. However, it is always possible to rotate the axes of measurement like

$$\rho' = R_\theta^T \rho R_\theta, \quad R_\theta = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix}, \quad (5.19)$$

such that the off-diagonal terms are opposite of each other, leaving different on-diagonal terms. We call this special choice the “principle axis of transport” and parameterize it as

$$\rho_{\text{principal}} = \frac{1}{2}(\rho_1 + \rho_2)\mathbf{I} + \frac{1}{2}(\rho_1 - \rho_2)\boldsymbol{\sigma}_z + \rho_H i\boldsymbol{\sigma}_y, \quad (5.20)$$

where $\rho_{1,2}$ are resistivities along the principal transport directions $\hat{e}_{1,2}$. We set $\rho_1 < \rho_2$ to enforce one solution with $\theta < 180^\circ$.

5.3.1 Transport calculation

Within the relaxation time approximation, the Boltzmann equation for a given energy band becomes

$$\frac{\partial n_{\mathbf{k}}}{\partial t} + (q\mathbf{E} + q\mathbf{v}_{\mathbf{k}} \times \mathbf{B}) \cdot \frac{\partial n_{\mathbf{k}}}{\partial \mathbf{k}} = -\frac{n_{\mathbf{k}} - n_{0,\mathbf{k}}}{\tau}, \quad (5.21)$$

where $q\mathbf{E} + q\mathbf{v}_{\mathbf{k}} \times \mathbf{B}$ is the total force on the Bloch electrons, with $\mathbf{v}_{\mathbf{k}} \equiv \nabla_{\mathbf{k}} \varepsilon_{\mathbf{k}}$ and charge q ; $n_{0,\mathbf{k}}$ is the equilibrium Fermi-Dirac distribution and $n_{\mathbf{k}}$ is the desired non-equilibrium distribution function.

We consider a stationary solution to the Boltzmann equation by parameterizing the distribution function as:

$$n_{\mathbf{k}} = n_{0,\mathbf{k}} + n_{1,\mathbf{k}}. \quad (5.22)$$

As a result, the Boltzmann equation for the deviation of the distribution function from equilibrium is:

$$(q\mathbf{E} \cdot \mathbf{v}_{\mathbf{k}}) \frac{\partial n_{0,\mathbf{k}}}{\partial \varepsilon_{\mathbf{k}}} + (q\mathbf{v}_{\mathbf{k}} \times \mathbf{B}) \cdot \frac{\partial n_{1,\mathbf{k}}}{\partial \mathbf{k}} = -\frac{n_{1,\mathbf{k}}}{\tau}. \quad (5.23)$$

Note that the magnetic field only couples to n_1 since $(q\mathbf{v}_{\mathbf{k}} \times \mathbf{B}) \cdot \nabla_{\mathbf{k}} n_{0,\mathbf{k}} = (q\mathbf{v}_{\mathbf{k}} \times \mathbf{B}) \cdot \mathbf{v}_{\mathbf{k}} \partial_{\varepsilon_{\mathbf{k}}} n_{0,\mathbf{k}} = 0$.

To solve the above partial differential equation (PDE), we seek a family of curves covering the $\mathbf{k}$ -space which we parameterize as $\mathbf{k}(s)$ with $s \in [0, s_0)$, such that along these curves the PDE becomes an ordinary differential equation (ODE). If a curve $\mathbf{k}(s)$ satisfies

$$\frac{d\mathbf{k}(s)}{ds} = q\mathbf{v}(s) \times \mathbf{B}, \quad (5.24)$$

then $n_{1,\mathbf{k}(s)} \equiv n_1(s)$ satisfies

$$(q\mathbf{E} \cdot \mathbf{v}_{\mathbf{k}}) \frac{\partial n_{0,\mathbf{k}}}{\partial \varepsilon_{\mathbf{k}}} \Big|_{\mathbf{k}=\mathbf{k}(s)} + \frac{dn_1(s)}{ds} = -\frac{n_1(s)}{\tau}. \quad (5.25)$$

Because

$$\frac{d\varepsilon(s)}{ds} = \mathbf{v}(s) \cdot \frac{d\mathbf{k}(s)}{ds} = 0, \quad (5.26)$$

the curve $\mathbf{k}(s)$ must coincide with the contour of constant energy. Thus, the Boltzmann equation becomes:

$$[q\mathbf{E} \cdot \mathbf{v}(s)] \frac{\partial n_0(s)}{\partial \varepsilon(s)} + \frac{dn_1(s)}{ds} = -\frac{n_1(s)}{\tau}. \quad (5.27)$$

The ODE is readily solved with:

$$n_1(s) = \chi_0 e^{-s/\tau} - e^{-s/\tau} \int_0^s ds' e^{s'/\tau} [q\mathbf{E} \cdot \mathbf{v}(s')] \frac{\partial n_0(s')}{\partial \varepsilon(s')}. \quad (5.28)$$

where χ_0 is a constant determined by the following argument. Since $\mathbf{k}(s)$ describes a constant energy contour in a two-dimensional Brillouin zone, it is either a closed contour, or several open contours that terminate on boundaries of the Brillouin zone such that they form a closed loop on a torus. In either case, $\mathbf{k}(s)$ is periodic under $s \rightarrow s + s_0$ modulo a moiré reciprocal lattice vector, where s_0 is the periodicity. The periodicity condition $n_1(s_0) = n_1(0)$ leads to

$$\chi_0 = \frac{1}{1 - e^{s_0/\tau}} \int_0^{s_0} ds' e^{s'/\tau} (q\mathbf{E} \cdot \mathbf{v}(s')) \frac{\partial n_0(s')}{\partial \varepsilon(s')}, \quad (5.29)$$

which determines the desired $n_1(s)$.

In the low-temperature limit, the steady state current from a given energy band is calculated as:

$$\begin{aligned} j^\mu &= q \int \frac{d^2\mathbf{k}}{(2\pi)^2} v_{\mathbf{k}}^\mu n_{1,\mathbf{k}} \\ &= \frac{q^2 B}{(2\pi)^2} \int d\varepsilon \int_0^{s_0} ds v^\mu(s) n_1(s) \\ &= \frac{q^3 B}{(2\pi)} \frac{\tau}{\omega_c} \sum_{n=-\infty}^{\infty} \frac{v_n^\mu v_{-n}^\nu}{1 + in\omega_c\tau} E^\nu, \end{aligned} \quad (5.30)$$

where $(\mu, \nu) = x, y$, and we have defined the cyclotron frequency as:

$$\omega_c \equiv 2\pi/s_0. \quad (5.31)$$

We have also made use of the periodicity of velocity under $s \rightarrow s + s_0$ to write it in terms of Fourier series, $\mathbf{v}(s) = \sum_{n=-\infty}^{\infty} \mathbf{v}_n e^{-in\omega_c s}$.

To show that the second line of Eq. (5.30) holds, note that at every $\mathbf{k}$ we can define a local coordinate system $(\hat{e}_v, \hat{e}_s)$ such that $\mathbf{v} \equiv v\hat{e}_v$ where $v \geq 0$, and $\hat{e}_s = \hat{e}_v \times \hat{z}$. The infinitesimal wavevector can be equivalently written as:

$$d\mathbf{k} = dk_x \hat{e}_x + dk_y \hat{e}_y = dk_s \hat{e}_s + dk_v \hat{e}_v.$$

Eq. (5.24) can then be written as $d\mathbf{k}/ds = qvB\hat{e}_s$, or equivalently $dk_s = qvBds$. As a result,

$$\int dk_x dk_y = \int dk_s dk_v = qB \int d\varepsilon ds.$$

The conductivity tensor is therefore given by the following expression:

$$\sigma^{\mu\nu} = \frac{q^3 B}{2\pi} \frac{\tau}{\omega_c} \sum_{n=-\infty}^{\infty} \frac{v_n^{(\mu)} v_{-n}^{(\nu)}}{1 + in\omega_c \tau}. \quad (5.32)$$

Eq. (5.32) gives the magnetoconductivity for a given FS contour. In the case of multiple FS contours and multiple bands – associated for example with spin and valley degeneracy in TBG – conductivities from different FS contours and bands add. Finally, the MR tensor is obtained by inverting the conductivity tensor, i.e., $\rho = \left(\sum_{n,i} \sigma_{n,i} \right)^{-1}$, where n, i are band and contour labels respectively for a given energy level.

5.3.2 Intuition and example

Fig. 5.7 shows the x -component of the Fermi velocity for three Fermi surface contours. For the closed Fermi surface contours, the averaged velocity, $\mathbf{v}_{n=0} \equiv \frac{1}{s_0} \int_0^{s_0} ds \mathbf{v}(s)$, is zero. On the other hand, for a typical open Fermi surface contour, it is finite, and as

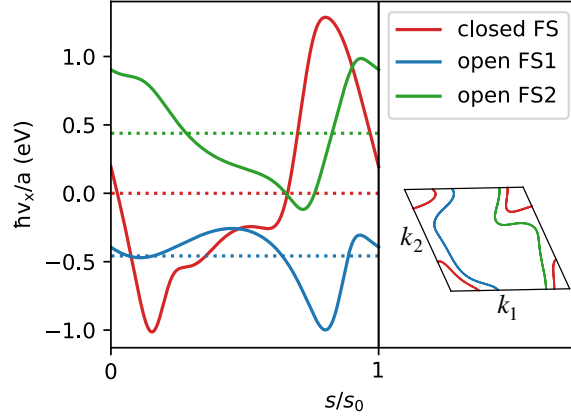


Figure 5.7: **Fermi velocity of closed and open Fermi surfaces.** Horizontal dotted lines are the average velocity for the associated contour.

a result the electron traversing the open Fermi surface contour in the presence of a magnetic field has a finite drift velocity in a particular direction.

To better understand Eq. (5.32) consider an example of a parabolic dispersion with $\varepsilon_{\mathbf{k}} = \frac{1}{2m_0}(k_x^2 + k_y^2)$, where m_0 is the bare electron mass. At a fixed energy μ the contour is a circle parameterized as: $(k_x, k_y) = \sqrt{2m_0\mu}(\cos\theta, \sin\theta)$, $\theta \in [0, 2\pi)$. Using the method of characteristics, we get: $\frac{d\theta}{ds} = -\frac{qB}{m_0}$, or $\theta = \theta_0 - \omega_0 s$, where $\omega_0 \equiv \frac{qB}{m_0}$ is the cyclotron frequency of bare electrons. This leads to the periodicity in s to be $s_0 = 2\pi/\omega_0$, where we have chosen the clockwise trajectory such that $s_0 > 0$. The Fourier series of the velocity along the constant energy contour is given by: $v_x(s) = \sqrt{\frac{\mu}{2m}}(e^{-i\omega_0 s} + e^{i\omega_0 s})$, and $v_y(s) = \sqrt{\frac{\mu}{2m}}\frac{1}{i}(e^{-i\omega_0 s} - e^{i\omega_0 s})$. Substituting into Eq. (5.32), we obtain the conductivity tensor:

$$\sigma = q^2 \tau \frac{\mu}{2\pi} \frac{1}{1 + \omega_0^2 \tau^2} \begin{pmatrix} 1 & -\omega_0 \tau \\ \omega_0 \tau & 1 \end{pmatrix}. \quad (5.33)$$

Note that the total number density of filled electrons is given by $n = \int \frac{d^2\mathbf{k}}{(2\pi)^2} \Theta(\mu - \varepsilon_{\mathbf{k}}) = \frac{m_0 \mu}{2\pi}$. We therefore reproduce the well known magnetoconductivity tensor:

$$\sigma = \frac{n q^2 \tau}{m_0} \frac{1}{1 + \omega_0^2 \tau^2} \begin{pmatrix} 1 & -\omega_0 \tau \\ \omega_0 \tau & 1 \end{pmatrix}. \quad (5.34)$$

In this simple example of a closed FS, the longitudinal resistivity is given by $\frac{m_0}{nq^2\tau}$, independent of the magnetic field. The average of the velocity field, $\mathbf{v}_{n=0} \equiv \frac{1}{s_0} \int_0^{s_0} ds \mathbf{v}(s)$, vanishes. However, for an open FS generally $\mathbf{v}_{n=0} \neq \mathbf{0}$, i.e. electrons have a finite drift velocity when a magnetic field causes them to traverse the contour. The impact of such a finite drift velocity on the magnetotransport can be qualitatively understood using the following example: in the expression for the conductivity tensor (Eq. (5.32)), we consider $v_{n=0}^x \neq 0$ but $v_{n=0}^y = 0$. This corresponds to an open FS with a drift velocity along the x direction. In the high-field limit ($\omega_c\tau \propto B \gg 1$), we truncate the Fourier series at the leading order, yielding

$$\sigma_{\text{open FS}} \approx \frac{q^3 B}{2\pi} \frac{\tau}{\omega_c} \begin{pmatrix} (v_0^x)^2 & -\frac{2\text{Im}(v_{-1}^x v_1^y)}{\omega_c \tau} \\ \frac{2\text{Im}(v_{-1}^x v_1^y)}{\omega_c \tau} & \frac{|v_1^y|^2}{\omega_c^2 \tau^2} \end{pmatrix}, \quad (5.35)$$

where we made use of the equality: $\mathbf{v}_{-n} = \mathbf{v}_n^*$. Inverting the matrix, we obtain the MR tensor:

$$\begin{aligned} \rho_{\text{open FS}} &\approx \frac{(2\pi)\omega_c}{q^3 B \tau} \frac{1}{4\text{Im}(v_{-1}^x v_1^y)^2 + (v_0^x)^2 |v_1^y|^2} \\ &\times \begin{pmatrix} |v_1^y|^2 & 2\text{Im}(v_{-1}^x v_1^y) \omega_c \tau \\ -2\text{Im}(v_{-1}^x v_1^y) \omega_c \tau & (v_0^x)^2 (\omega_c \tau)^2 \end{pmatrix}. \end{aligned} \quad (5.36)$$

We see that for an open FS, the longitudinal MR has non-saturating B^2 behavior along the axis with a zero drift velocity ($\hat{y}$ in the above example), and saturating behavior (constant in B in this simple model) along the other axis.

5.4 Transport results and comparison to experiment

Individually, the three aspects of the model are not complicated: we create a moiré lattice based on a twist angle and strain, then we solve the BM Hamiltonian to find the isolated moiré bands, and then we compute transport properties based on the Boltzmann equation in the relaxation time approximation.

Parameter	Symbol	Typical value(s)
Twist angle	θ	0 to 15°
Global angle	θ_g	0 to 60°
Uniaxial homostrain amount	$\epsilon_{\text{uni,homo}}$	0 to 1%
Uniaxial homostrain direction	φ_{homo}	0 to 60°
Biaxial homostrain amount	$\epsilon_{\text{bi,homo}}$	-1% to 1%
Uniaxial heterostrain amount	ϵ_{uni}	0 to 1%
Uniaxial heterostrain direction	φ	0 to 60°
Biaxial heterostrain amount	ϵ_{bi}	-1% to 1%
Poisson ratio	ν	0.16
Fermi velocity	v_F	10^6 m/s
Deformation potential	α	-4.1 to 30 eV
Pseudovector potential	β	3.14
AA hopping energy	w_0	88 meV
AB hopping energy	w_1	110 meV
Relaxation time	τ	Picoseconds

Table 5.1: Overview of model parameters.

Each step of this process brings with it a set of model parameters (described in Table 5.1). Some of these parameters are known well from first principles or experiments, such as the Poisson ratio and the pseudovector potential strength. Others vary from sample to sample, such as twist angle and strain. Our dream would be to constrain the model parameters based on experimental measurements, so that, using transport, we could precisely define twist angle and strain within the device.

In practice, this dream is not entirely feasible. For one, the models we are using are too simple. The BM model that we build on, for instance, has electron-hole symmetry, and yet the transport measurements are clearly electron-hole asymmetric. More sophisticated non-interacting model calculations [132, 133] as well as interaction renormalizations [134] would likely be necessary to properly account for such details. For another, the measurements themselves are subject to inhomogeneity, which will be hopelessly complicated to model generally. As an example, it is theoretically possible to back out the direction of strain from the amount of mixing of XX into XY in the open-FS regions. Unfortunately, while neighboring longitudinal and Hall pairs show similar angles, they are different enough to preclude any certainty in the

results (Fig. A.24).

So, we will not attempt the full dream. Instead, we will find the general phenomenological predictions of the model. We will then reanalyze the data from our devices and decide if these predictions are upheld.

We present calculations for $\theta = 1.38^\circ$, $\epsilon = 0.2\%$, and $\varphi = 0^\circ$, parameters which are not measured in the experiment but are chosen to yield reasonable quantitative agreement between the theoretical and experimental results with respect to the filling range of open FSs, and to the frequencies of magnetoresistance oscillations to be presented later. The general phenomena of open FSs and quadratic MR hold for a broad range of heterostrain parameters ϵ and φ . We do not perform fine-tuning of these input parameters for two reasons: 1. We do not expect our strained BM model in Eq. (5.9) to yield precise quantitative agreement with experiment. 2. Increasing ϵ broadens the filling range that displays open FSs, but for a given ϵ , varying φ strongly tunes this range (see Fig. 5.5), so there is some flexibility in assigning the two parameters to match the experimental measurements. This might be overcome by also seeking to match the position of the lowest van Hove singularity and the evolution of sizes of the two Fermi pockets near the CNP, but the simplifications in the BM model noted in (1) above caution us against drawing strong quantitative conclusions from the comparison.

Magnetotransport for unstrained TBG at 1.38° is shown in Fig. 5.8. First, the unstrained model has C_{3z} symmetry, so $\rho_{xx} = \rho_{yy}$. Second, cusp-like features develop at the triply-degenerate van Hove point at filling fractions $\nu \approx \pm 1.4$, and are attributed to the non-analyticities in the density of states at the van Hove singularities. Third, unstrained TBG has saturating MR across all filling range, as illustrated in the inset. Finally, the Hall number changes sign precisely at the density associated with the van Hove point.

5.4.1 Quadratic magnetoresistance

In Fig. 5.9, we show the computed MR for strained TBG along the principal transport axes (a) and the Hall number (b). For comparison, we plot the experimentally

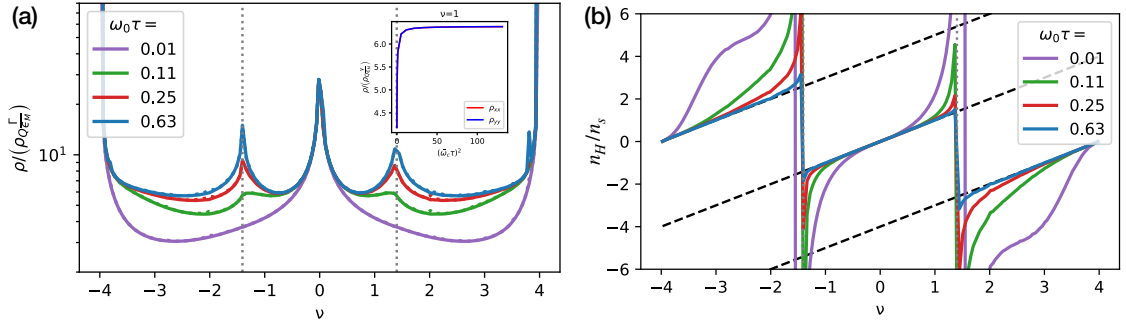


Figure 5.8: **Magnetotransport properties of unstrained TBG.** Theoretical calculations of transport properties as a function of magnetic field strengths $\omega_0\tau$ for $\theta = 1.38^\circ$, $\epsilon = 0\%$ and $\varphi = 0^\circ$. The cyclotron frequency ω_c defined in Eq. (5.31) is filling-dependent, hence our choice to use the bare cyclotron frequency $\omega_0\tau = eB\tau/m_0$. The vertical dashed lines mark the calculated van Hove points. (a) Longitudinal MR ρ_{xx} (dashed) and ρ_{yy} (solid) in units of $\rho_Q \frac{\Gamma}{\epsilon_M}$, where $\rho_Q \equiv h/e^2$ is the quantum of resistance, $\Gamma \equiv \hbar/\tau$ is the transport decay rate, and $\epsilon_M \equiv \hbar v_F |\mathbf{K}| \theta$ is the characteristic energy scale for moiré electrons. For a transport rate $\Gamma = 0.1 \text{ meV}$, $\rho_Q \frac{\Gamma}{\epsilon_M} \approx 9.6 \Omega$, and $\omega_0\tau \approx 0.13$ is equivalent to a magnetic field strength $B \approx 0.11 \text{ T}$. The inset shows a saturating MR as magnetic field is increased. (b) Hall number $n_H \equiv B/e\rho_H$.

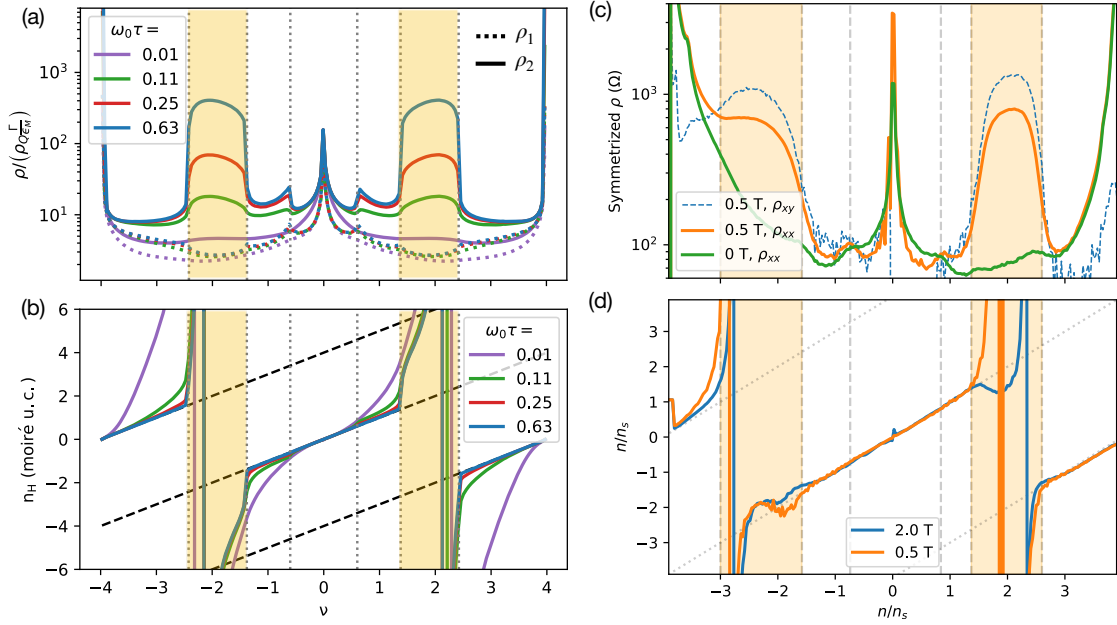


Figure 5.9: **Magnetotransport properties of strained TBG.** (a-b) Theoretical calculations of transport properties as a function of magnetic field strengths $\omega_0\tau$ for $\theta = 1.38^\circ$, $\epsilon = 0.2\%$ and $\varphi = 0^\circ$. The vertical dashed lines mark the calculated van Hove points, with yellow regions indicating open FSs. (a) Longitudinal MR along the principal axes $\hat{e}_1$ (dashed) and $\hat{e}_2$ (solid). (b) Hall number. (c-d) Experimental measurements of longitudinal MR (contact pair 14-15) and transverse MR (contact pair 15 - 5), at 1.6 K. Vertical dashed lines mark the densities that we ascribe to van Hove points based on the cusp near $\nu \sim 0.8$ and the onset of quadratic MR (shaded yellow). Finite-field resistivities in panel (c) are symmetrized: $\rho = (\rho(B) + \rho(-B))/2$. Panel (d) is calculated from the antisymmetrized transverse resistivity. In the experimental figures, n/n_s labels the electron filling fractions per moiré unit cell.

measured longitudinal and transverse resistivities (c) and Hall number (d).

In the filling ranges with open FSs, the calculated $\rho_2(B)$ exhibits quadratic non-saturating MR, whereas $\rho_1(B)$ saturates. The filling range for which quadratic MR occurs is bounded by the two outermost van Hove points of the zero-field strained band structure. In experiment, we observe quadratic MR in longitudinal resistivity within a similar range of fillings. More strikingly, we observe quadratic MR in the transverse resistivity as well. In some cases, with increasing field the symmetric part of the transverse resistivity becomes larger than that of the longitudinal resistivity. This degree of mixing can be attributed to the misalignment between the strain-induced but filling-dependent principal axis of transport and the direction of current flow in the Hall bar geometry.

At the first van Hove point ($\nu \approx \pm 0.6$), the non-analyticity in the density of states leads to a cusp in the first derivative of the zero-field resistivity with respect to filling. As shown in Fig. 5.9(a), at $B \neq 0$ the longitudinal resistance as a function of filling develops a cusp at the first van Hove point. The cusp becomes more pronounced with increasing B . Experimentally as shown in Fig. 5.9(c), there is a cusp-like feature developing at $|\nu| \sim 0.5 - 0.8$ depending on the contact pair within the device used, consistent with theoretical predictions. In many contact pairs, this feature presents as a shoulder at $B = 0$, only developing into a cusp at $B \sim 0.1$ T (see Fig. A.25).

As depicted in Fig. 5.9(b), the calculated filling dependence of the Hall number shows two singular sign changes near $\nu \approx \pm 2$, each falling within inside an open FS region. The filling at which each sign-changing singularity occurs is B -independent, and is not directly associated with any van Hove point. Moreover, the filling dependence of the Hall number n_H tracks the filling fraction in a broad filling range near the CNP, with the filling range being extended upon increasing B . In Fig. 5.9(d), we observe the same general shape of the Hall number.

Within the open FS filling range, however, the measured Hall number qualitatively deviates from the theoretical curves, as previously noted in Chapter 3, Fig. 3.9. We tentatively attribute this to a small constant offset in the magnetic field of order 10-20 mT, an amount typical for trapped flux in a NbTi/Nb₃Sn superconducting magnet like ours. To explicate: in the open FS regime we expect (and observe)

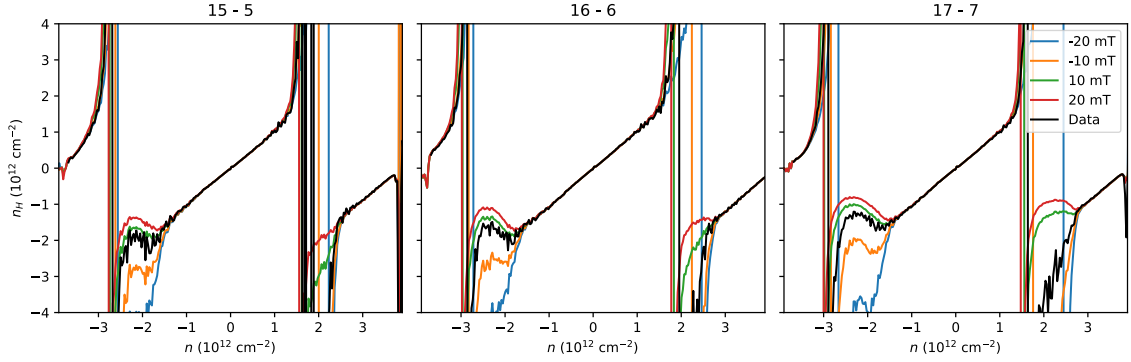


Figure 5.10: **Potential error from improper antisymmetrization:** Filling dependent Hall number corrected for antisymmetrization error of the indicated contact pairs at $B = 0.5\text{T}$. All other transverse contact pairs do not have a large degree of mixing and also do not have these features (Fig. A.25).

a large quadratic symmetric component of the transverse resistivity together with a vanishing antisymmetric component. Hence quantification of the antisymmetric component involves subtracting two large numbers. An offset of only a few mT in assigned field will lead to a small part of the symmetric component mixing into the antisymmetric component, as in

$$\tilde{\rho}_H(B) = \frac{\rho_{yx}(B + \delta B) - \rho_{yx}(-B + \delta B)}{2}, \quad (5.37)$$

where δB is a systematic error. Due to the misalignment of transport principal axis with the Hall bar geometry, longitudinal MR also contributes to $\rho_{yx}(B)$. In the filling range with open Fermi surfaces, the longitudinal resistance exhibits non-saturating quadratic MR, and mixes into the Hall component which is odd in B . As a result, one expects the improper antisymmetrization error to be largest in this filling range.

We investigate this possibility by first fitting a polynomial to the low-field transverse resistivity. This allows us to interpolate the data and add small constant offsets prior to antisymmetrization. Accounting for an offset of roughly 20 mT largely removes the bumps from the data (Fig. 5.10). This offset is larger than what we would expect from trapped flux in a superconducting magnet, however we do not expect the procedure to be accurate to such a fine degree, simply because we do not have fine

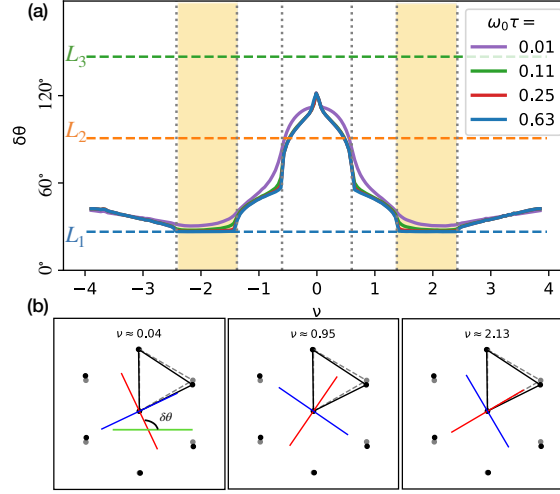


Figure 5.11: **Rotation of the transport principle axis.** (a) Rotation of the transport principal axis $\hat{e}_1$ with respect to the global coordinate system for strained BM with $\epsilon = 0.2\%$ and $\varphi = 0^\circ$. The three horizontal dashed lines are the bond directions. In the open FS region, the saturating MR axis is locked to the shortest bond ($\mathbf{L}_1$) direction. However, it rapidly rotates in the closed FS region upon approaching the CNP. (b) Principal transport axes $\hat{e}_1$ (red) and $\hat{e}_2$ (blue) for a few filling fractions. Near the CNP, $\hat{e}_1$ is perpendicular to the shortest moiré bond direction. In the open FS filling range (e.g. $\nu \approx 2.13$) it is rotated to be parallel to the shortest bond direction.

enough resolution in field to get an accurate polynomial fit.

5.4.2 Rotation of the principle axis of transport

Our calculation finds a dramatic rotation of the principal axis with filling, as illustrated in Fig. 5.11. In the filling range with open FSs, the principal axis with saturating MR ($\hat{e}_1$) is aligned with direction of the shortest moiré triangular bond, suggesting that the electrons are hopping more efficiently along the shortest bond, which leads to a larger conductivity and therefore a smaller resistivity. Remarkably, when filling is changed from the second van Hove point ($\nu \approx \pm 1.3$) to the vicinity of the CNP, $\hat{e}_1$ rotates by about 90° . This is likely associated with the larger Fermi pocket encircling a Dirac point being elongated in a direction nearly perpendicular to the open FS contours. Such rotation of the principal transport axis with filling has

been attributed to interaction-induced nematicity [135], but we can now see that in some contexts it can occur purely due to strain-induced bandstructure effects.

Such a filling-dependent rotation of the principal transport axis was not possible to observe using the Hall bar geometry, where only ρ_{xx} and ρ_{yx} are measured but not ρ_{yy} . Additional transport measurements are needed, where the filling-dependence of the entire resistivity tensor can be mapped out.⁸

5.4.3 Two FS to one FS transition at first van Hove point

Since this theory predicts a van Hove point between the CNP and the filling range with open FSs, a direct experimental signature of this van Hove point is desired. In Fig. 5.12 we reanalyze quantum oscillation measurements of the TBG device discussed in Ref. [64].

The effective cyclotron mass m^* is light in the filling range with two small closed Fermi pockets, and dramatically heavier in the filling range with only one closed pocket (Fig. A.26). The large difference in masses on either side of the innermost van Hove singularity can account for the substantially lower-field onset of quantum oscillations close to the CNP than away from it, as shown in Fig. 5.12(a).

Fig. 5.12(b) is a Fourier transform of the quantum oscillation data with respect to $1/B$. In the filling range of $-0.7 \leq \nu \leq 0.8$ three distinct frequencies $f_{i=1,2,3}$ are clearly observed in the data, with f_1 and f_2 corresponding to two small Fermi pockets, and $f_3 = f_1 + f_2$ to the breakdown orbit: by 1 T the inverse magnetic length is comparable to the momentum space distance between the two small Fermi pockets [137]. Each edge of this filling range is marked by two features: 1. f_1 and f_2 disappear from the Fourier transform, leaving only f_3 at higher electron or hole filling. 2. A cusp-like feature occurs in longitudinal MR (Fig. 5.9(a) and (c)). These both are predicted by our model as features of a Lifshitz transition on either side of the CNP, associated with crossing the lowest-energy van Hove points. This detailed

⁸I attempted to fabricate “clock” devices [136], with many voltage probes arranged symmetrically around a small $\sim 1 \mu\text{m}^2$ circle of TBG. While I made a few such devices that showed unusual magnetotransport (quadratic MR regions, splitting/bending LLs), they were too inhomogeneous to draw any conclusions from.

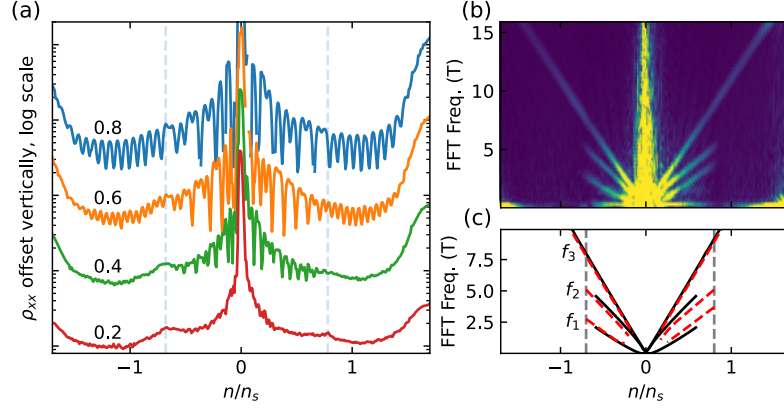


Figure 5.12: **Evidence for two-FS to one-FS transition.** (a) Line cuts of MR near the CNP taken at 26 mK in contact pair 4 - 5 at the indicated field strengths, in Tesla. Vertical dashed lines indicate our estimated location of the lowest-energy van Hove points, based on the cusps in resistivity at low field. Within the region bounded by these points, the quantum oscillations show up before 0.4 T, and their relative strengths do not follow a simple pattern. Outside of this region, the quantum oscillations onset at higher field, and every multiple of 4 quantum Hall filling fraction is observed relatively equally. (b) Fourier transform of the quantum oscillation data with respect to $1/B$. It reveals a transition from two pockets to one pocket at the lowest-energy van Hove points. (c) Schematic description of the frequencies observed in panel (b). Red dashed lines are frequencies from the experimental data. Solid black lines are predictions from the theory for $\epsilon = 0.2\%$ and $\varphi = 0^\circ$. The two frequencies f_1 and f_2 sum to the one-pocket frequency f_3 that extends beyond the first van Hove point. They additionally account for the nontrivial relative strengths of the quantum oscillations within the bounds of the first van Hove points. As with other details of this work, the theory predicts electron-hole symmetry, while some asymmetry is observed in experiment.

match unambiguously demonstrates the existence of a third van Hove singularity to each side of the CNP, at electron or hole filling range (respectively) lower than the onset of B^2 MR. The Hall number does not show a sign-changing singularity at this van Hove point, as illustrated in Fig. 5.9(b) and (d).

The frequencies $f_{1,2}$ are a strong constraint on the amount of heterostrain in the TBG sample. Specifically, as illustrated in Fig. 5.12(c), the frequency f_2 is roughly two times f_1 , showing that the two small Fermi pockets have an area ratio $\sim 2 : 1$. Theoretically as illustrated by the solid black lines in Fig. 5.12(c), for a heterostrain strength $\epsilon = 0.2\%$ and $\varphi = 0^\circ$, the areas $A_{i=1,2}$ of the two small pockets, when converted to frequency via $f_i^{-1} \equiv (\Delta \frac{1}{B})_i = \frac{2\pi e}{\hbar A_i}$, are in good agreement with experiment. Furthermore, also note that the frequencies $f_{1,2}$ extrapolate to 0 at $\nu \approx \pm 0.04$, showing that the two Dirac points are shifted to finite (opposite) filling fractions by heterostrain.

We observe behavior qualitatively similar in all respects to that in Fig. 5.12(a) in all 3 longitudinal contact pairs for which we have dilution-fridge measurements, however the other contact pairs do not show the effect as cleanly (Fig. A.27).

5.5 Summary and outlook

In summary, we have shown that due to the large size of the moiré unit cell at small twist angles, even a small amount of uniaxial heterostrain on the microscopic scale can lead to dramatic changes in the narrow bands of twisted bilayer graphene. A key feature of the strained bandstructure is the splitting of the respective energetic degeneracies of the two Dirac points and the three van Hove points. The splitting of the two Dirac points leads to a semimetallic state with two small Fermi pockets at the CNP. On the other hand, the two outermost van Hove points bound a broad filling range near $\nu = \pm 2$ where the constant energy contours become open. Interestingly, the elongation of the larger Fermi pocket near the CNP is perpendicular to that of the open FSs, the latter being perpendicular to the direction of the shortest moiré triangular bond.

We have analyzed the resulting magnetotransport in strained TBG in the framework of the Boltzmann equation using the method of characteristics, treating the magnetic field non-perturbatively. We showed that a non-saturating quadratic longitudinal magnetoresistance in a broad filling range near $\nu = \pm 2$ naturally arises due to the heterostrain-induced open Fermi surfaces, therefore explaining in remarkable detail experimental results in off-magic-angle devices with lattice anisotropy. We have also shown that the sign-changing singularities in the Hall number occur in the open FS filling range and are not directly associated with any van Hove singularity as commonly assumed, e.g., in Ref. [138]. Furthermore, our theory reveals a dramatic rotation of the transport principal axis as the filling is tuned from the charge neutrality point to the filling range of open Fermi surfaces, without invoking interaction-induced electronic nematicity.

Given the importance of energy-shifted van Hove points in the transport properties of TBG devices, we have analyzed previous quantum oscillation data, which has revealed a Lifshitz transition from two pockets to one pocket at a filling fraction where the innermost van Hove singularity is predicted to occur based on theoretical calculations, offering strong evidence of heterostrain effects on these devices. We have further proposed several additional signatures to look for in future experiments. These include a significant difference in cyclotron mass on either side of the innermost van Hove singularity (probed qualitatively but not quantitatively in the extant experiment) and a principal transport axis with saturating magnetoresistance in the open Fermi surface filling range.

Chapter 6

More than a toy model: high field

HAVING shown that the low-field magnetotransport is well-captured by a strained BM model calculation in Chapter 5, I now return to the question of the split Landau levels that I previously phenomenologically modeled in Chapter 4. To recap: I found that an energetically-doubled Hofstadter’s butterfly toy model with anisotropy accurately captured the behavior of my high-field data. I suggested that strain and Zeeman splitting could account for the two added ingredients, but I could not say for sure without a more accurate model of strained TBG.

Bistrizer and MacDonald extended their continuum model of TBG into the Hofstadter’s butterfly regime in Ref. [105]. While their method works well, it is computationally expensive. Oskar Vafeek and Xiaoyu Wang invented a method of computing TBG’s butterfly that is significantly computationally cheaper [139]. They were kind enough to share their code with me so that I can compare my experiment to theory. Conveniently, the exact same modifications to the BM model to account for strain, described in Chapter 5, can be included just as easily in this model.

In this chapter, I first show that the question of four-vs-eightfold-degenerate Landau levels in twisted bilayer graphene can likely be neatly resolved by strain, as predicted in Refs. [62, 140]. I present measurements taken up to 28 T at the National High Magnetic Field Lab. At such fields, exotic many-body states are to be expected in clean samples, even samples twisted away from the magic angle. I describe the various interesting states that I see, making reference to the strained BM calculations

as it is useful, however I do not go into a thorough explanation of them. This dissertation needs to end at some point, after all. I close with the claim that the same 0.2% uniaxial heterostrain from Chapter 5 plus Zeeman splitting makes a good match to my high-field experimental results.

Using Oskar and Xiaoyu’s strained BM butterfly code

The code takes as input magnetic field as p and q and produces a spectrum for that field in $\mathcal{O}((q/p)^3)$ time. On my personal computer, I can run up to $q/p = 40$ before it becomes prohibitively slow, producing a low-field cutoff of roughly 1 T for my 1.38° sample. To compute density of states $\rho(E)$ and carrier density $n(E)$, I follow Ref. [141] by introducing a smoothing parameter γ (in meV) and writing

$$\rho(E) = \sum_i \frac{1}{\pi} \frac{\gamma}{(E - E_i)^2 + \gamma^2}, \quad (6.1)$$

$$n(E) = \sum_i \frac{1}{\pi} \arctan \left(\frac{E - E_i}{\gamma} \right). \quad (6.2)$$

The spectra are not evenly spaced in field, leading to visible imperfections of the computed butterflies, especially near simple fractions.

The spectra for $\theta = 1.38^\circ$ are shown in Fig. 6.1 with and without both 0.2% uniaxial heterostrain and Zeeman splitting $V = g\mu_B B$. Without strain, the spectrum is similar, although not identical, to the honeycomb spectrum in Fig. 4.5. With strain, the butterfly separates into two butterflies. Do note that there is not a gap in the strained butterflies (right panels of Fig. 6.1) at zero field: the Landau levels moving towards CNP from the two separated butterflies cross below my low-field cutoff.

6.1 Landau level degeneracies in strained TBG

The first studies on TBG were on relatively high-angle samples (1.8° and above), in which they observed a primary Landau level sequence near CNP of $\nu = \pm 4, \pm 12, \pm 20, \dots$ [34], each Landau level eightfold degenerate. This sequence is precisely double that

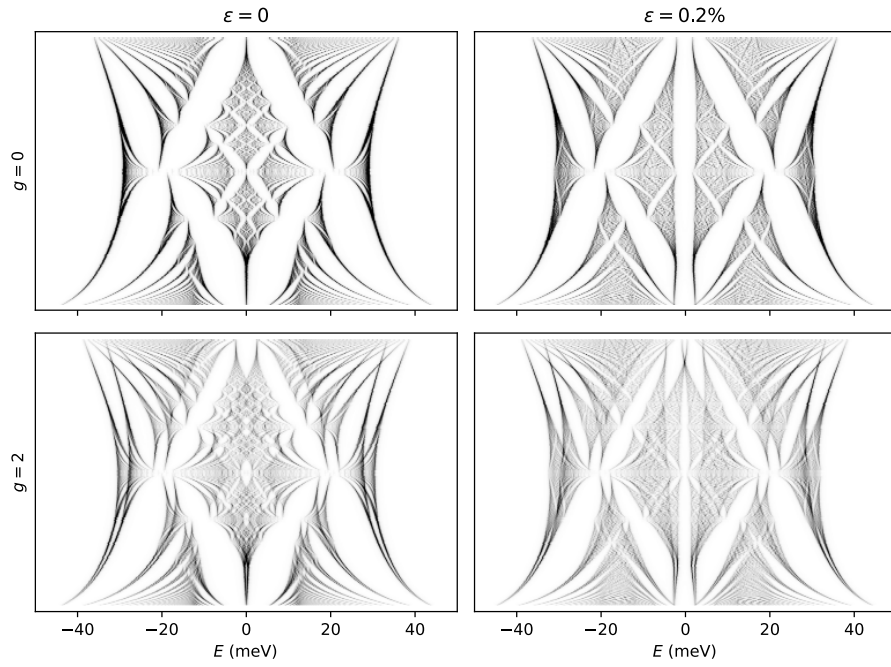


Figure 6.1: **Spectra of BM model with and without strain and Zeeman splitting.** All spectra computed up to $q = 44$ and plotted for $\theta = 1.38^\circ$, $\varphi = 0$, $\gamma = 0.1$ meV. The y axis is field and ranges from 0 to 1.

observed in monolayer graphene, which makes sense under the intuition that high twist angle TBG is effectively described as two decoupled monolayers. Indeed, the unstrained BM model predicts precisely this LL sequence at all twist angles at and above the magic angle.

TBG devices closer to the magic-angle tend to show a different sequence of fourfold-degenerate Landau levels: $\nu = \pm 4, \pm 8, \pm 12, \dots$ (Refs. [37, 42, 46], for example). I would like to note, however, that on closer inspection, the case for dominantly-fourfold-degenerate LLs is not always so clear. For instance, in Ref. [46] Figures 1A and S1A, $\nu = -12$ and -20 clearly persist to significantly lower fields than $\nu = -8$ and -16 , implying that the eightfold degeneracy is still slightly preferred.

In my own measurements, I see the same pattern, but again it is not always so clear. In my 1.7° sample and Matt Yankowitz' 1.59° sample, I see eightfold-degeneracy on the hole side but fourfold-degeneracy on the electron side (Figs. 3.17 and A.18). I see the fourfold-degenerate sequence clearly in Matt Yankowitz' 1.52° device in Fig. A.19, however in that sample, there appear to be many LL crossings: the gaps disappear and then later reappear (as in the right panel of Fig. 3.14). In my 1.4° device (Chapter 3), I see mostly eightfold-degenerate LLs at 1.7 K and fourfold-degenerate LLs at 26 mK, but the pattern is not totally consistent.

There was initially significant theoretical confusion about the origin of this change in Landau level degeneracy near the magic angle, with some authors suggesting finely-tuned modifications due to interactions (for instance, Ref. [141]). Refs. [62, 140] suggested that the lack of threefold rotational symmetry induced by uniaxial strain could explain the fourfold-degenerate Landau levels, however neither computed the actual spectra for a realistic model of TBG with strain. Refs. [142, 143] find in STM measurements that the Landau spectrum near CNP can be explained by a small shift in the energies of the two Dirac cones: consistent with strain.

The Wannier plots corresponding to the spectra in Fig. 6.1 are shown in Fig. 6.2. With neither strain nor Zeeman (top left panel), the Landau level spectrum emitting from CNP is eightfold degenerate. Adding a small amount of uniaxial heterostrain (top right) breaks this down to fourfold. Adding Zeeman splitting in both cases yields small gaps at half the degeneracy that close up both as they approach CNP and as

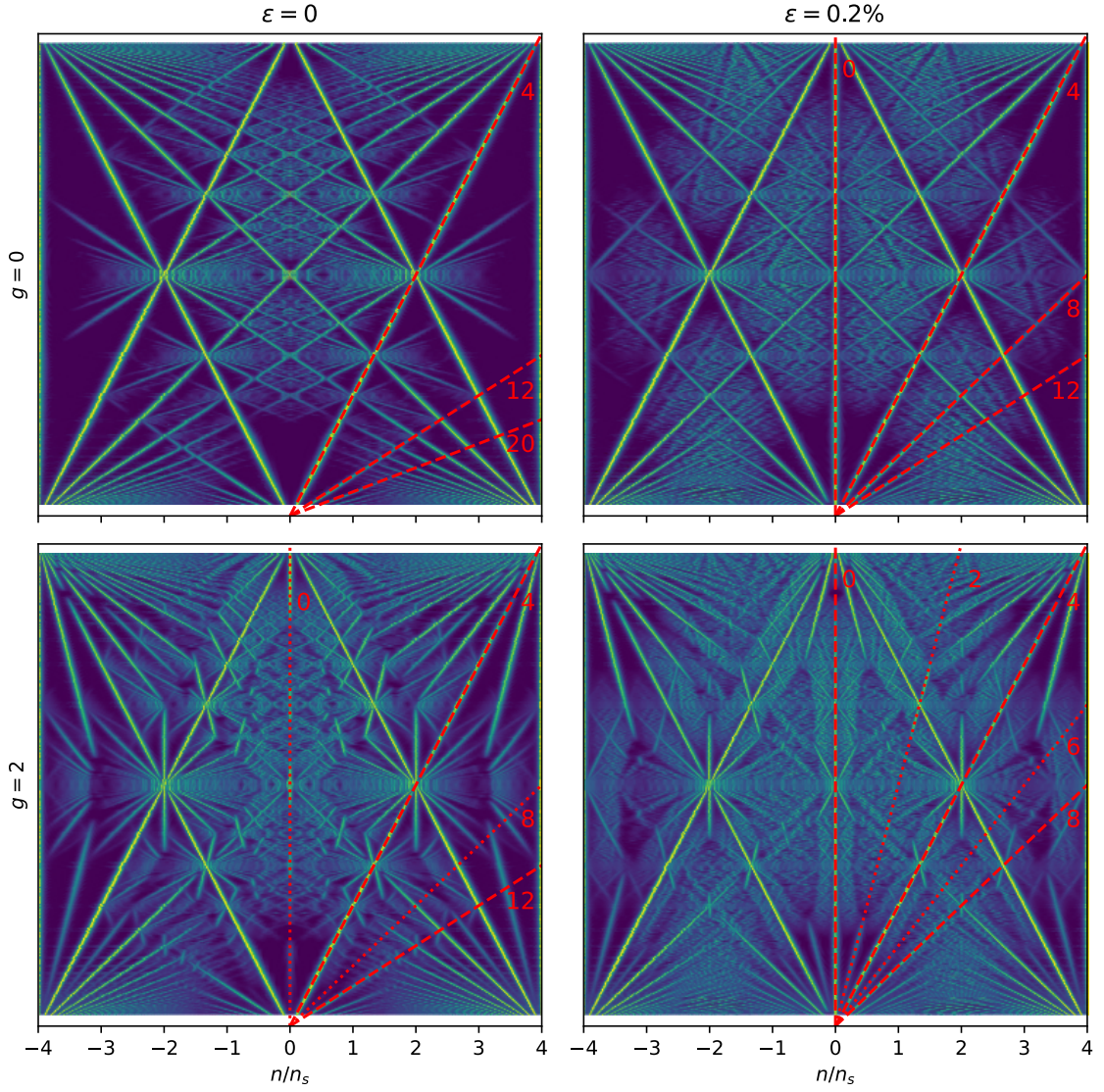


Figure 6.2: **Wannier plots of BM model with and without strain and Zeeman splitting.** Color is inverse density of states. These are the same spectra as in Fig. 6.1 with $\theta = 1.38^\circ$. I have included labelled dashed red lines along the major LL gaps originating at CNP. The dotted red lines are small gaps opened by Zeeman splitting.

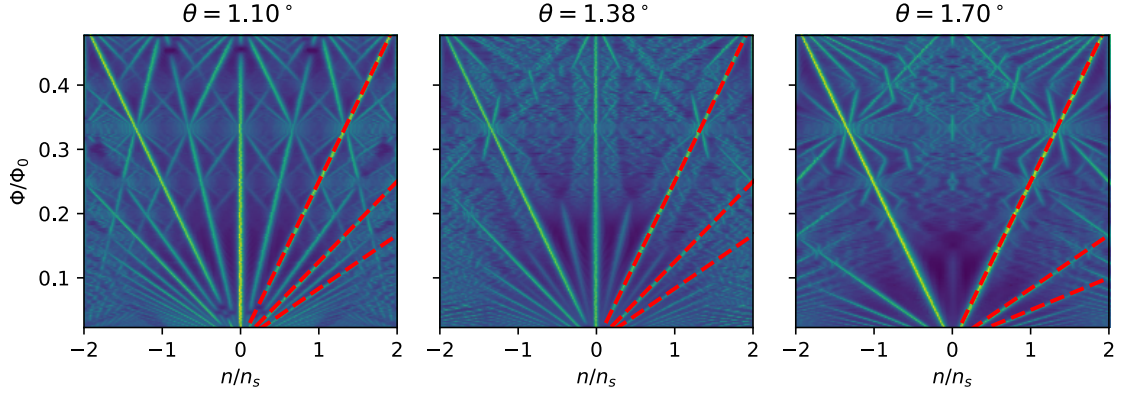


Figure 6.3: **Landau level degeneracy at a few twist angles.** In all cases, I take q up to 44 and use $\epsilon = 0.2\%$ and $\varphi = 0$. I take $\gamma = 0.1, 0.2, 0.3$ for the three respective plots, to account for the increasing bandwidth. Color is inverse density of states. Colors across the three plots do not have the same values. Red dashed lines are $\nu = 4, 8, 12$ for the left two plots and $\nu = 4, 12, 20$ for the rightmost plot.

the two larger neighboring LLs overlap.

Fig. 6.3 shows Wannier plots for 1.10° , 1.38° , and 1.70° , in all three cases with 0.2% uniaxial heterostrain. In the first two cases, the dominant LL spectrum is $\nu = \pm 4, \pm 8, \pm 12, \dots$ (dashed red lines). In the third plot, the dominant LL spectrum is $\nu = \pm 4, \pm 12, \pm 20, \dots$ (dashed red lines).

This behavior makes intuitive sense. See Fig. 6.4. First, recall that the effects of strain are magnified by twist angle: the various effects go as powers of ϵ/θ . So, the moiré lattice distortion from strain, and hence magnitude of C_{3z} symmetry breaking, is relatively lower at higher twist angles (Fig. 5.3). Additionally, the bandwidth (and thus maximal gap sizes) are dramatically higher at higher twist angles compared to the Dirac point energetic splitting. The Fermi velocity and thus energetic separation of Landau levels (from Eq. 1.8) is also higher at higher twist angles [35]. The numerics work out so that strains of a few tenths of a percent, like those seen in typical TBG samples, lead to noticable differences in Landau level sequences above and below roughly 1.5° (Fig. 6.4, right panel). At higher strain amounts, the fourfold degeneracy can be visible at even higher twist angles, and at lower twist angles, less strain is required to see the fourfold degeneracy. At 1.4° , for instance, less than 0.05%

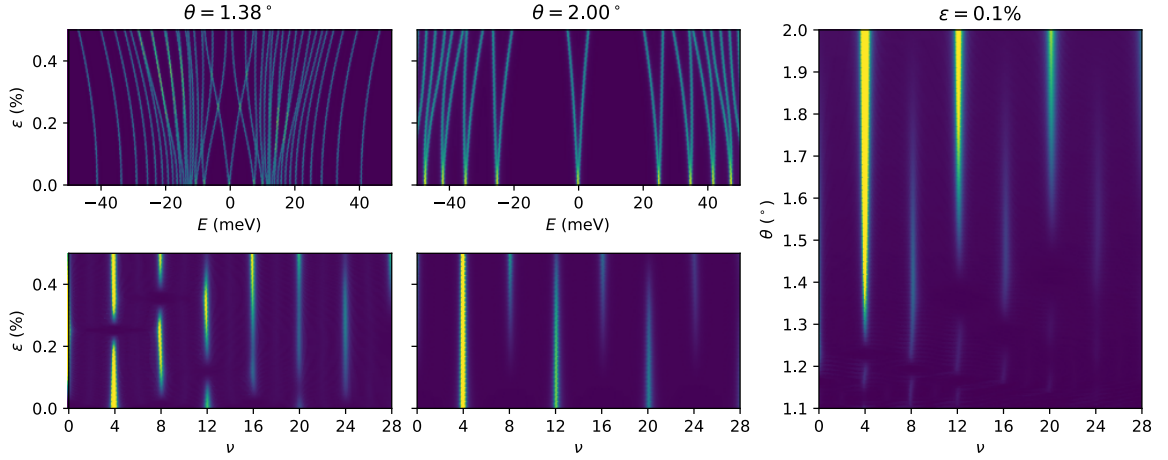


Figure 6.4: **Landau level degeneracies with strain.** The top two panels are DOS, and the lower three are inverse density of states. I show two twist angles: 1.38° (leftmost panels, $p/q = 1/20$) and 2.00° (center panels, $p/q = 1/35$). I chose p/q for each so that the real magnetic field is the same in both cases, near 2.5 T, however this choice does not matter much. The top row is the spectrum as uniaxial heterostrain is turned on from 0, where both twist angles are eightfold degenerate, up to 0.5%. The lower row shows inverse density of states as a function of strain and Landau level filling factor ν . The plot on the right is at fixed strain and $p/q = 1/30$. As twist angle increases, the relative strength of the fourfold-degenerate LLs decreases compared to the eightfold LLs. Note that in all plots, LL gaps may disappear and reappear as they cross each other.

uniaxial heterostrain is required for fourfold degeneracy.

Note that the quantitative numerics should not be taken completely seriously, for the same reasons as described in Chapter 5: the model is too simple. Additionally, the amount of energetic splitting of the Dirac points is affected by the deformation potential, which has a wide range of measured values in the literature. Still, the rapid increase in Fermi velocity with twist angle above the magic angle [35] implies that the same general trend should occur regardless.

6.2 Maglab measurements

The evident homogeneity of the device near contact pairs 6, 7, and 8, as implied by its crisp and finely-detailed Landau fan measurements, encouraged us to perform additional measurements at MagLab. We measured in SCM-32T: an all-superconducting system composed of an Oxford outer magnet and a homemade high- T_C superconducting inner magnet with a maximum combined field of 32 T. We were one of the early users of the system, and so we were only allowed to run the field to 28 T.¹ The nominal base temperature of the system was 50 mK, however it did not feature low-pass filtering on the measurement lines. This led to significant noise within the system and ultimately blurrier measurements. Humerously, we could see a dramatic increase in the cleanliness of our measurements every day at 8 PM, when the local radio station turned down their output power to comply with FCC regulations.

We first took a Landau fan diagram of multiple contact pairs,² some longitudinal and some Hall. We found that there was significant noise when we had many instruments connected to the sample. This noise fortunately did not show up in all contact pairs at all gate voltages and fields. It did show up at all fields in the current measurement (Fig. A.28), but crucially not in the current itself (the voltage does not show the same features). Measurements of all longitudinal pairs are shown in Fig. A.29, and all Hall pairs are shown in Fig. A.30.

¹Additionally, unlike in their other superconducting magnet systems, we could not run the field when we were not physically present. This made the trip very significantly more boring than we expected, because the measurement itself did not require much of our attention.

²The probe only had 16 DC wires, so I had to choose a small subset of contact pairs to measure.

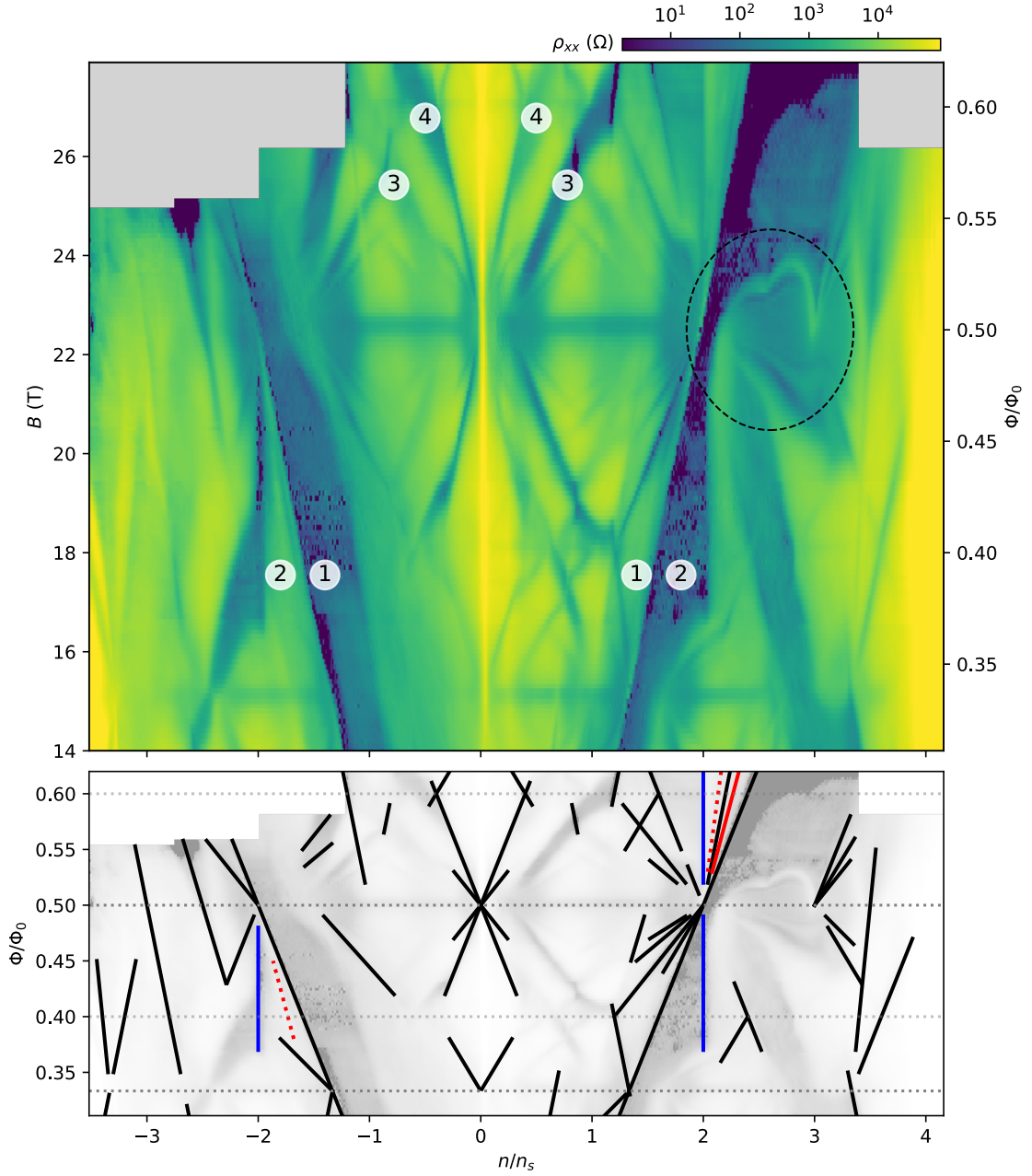


Figure 6.5: **Maglab data: longitudinal.** Described in the text. **Top:** Resistivity vs field and density for contact pair 7 - 8. Circled numbers are described in the text. **Bottom:** Schematic showing some of the Streda lines in the top panel, with a few (s, t) pairs labelled. Blue lines have 0 Chern number. See the text for the meaning of the red lines. Horizontal dotted lines are at $\Phi/\Phi_0 = 1/3, 2/5, 1/2, \text{ and } 3/5$.

The noise was greatly reduced when we only measured one contact pair. So, we redid the measurements for the single longitudinal contact pair that showed the sharpest features, which turned out to be 7 - 8. The results are shown in Fig. 6.5. We started from 14 T and worked to higher fields over several days. Near the end of the measurement, the device's gate began to leak. We had to back off the gate range towards the end, hence the missing regions at high field. This ultimately spelled the demise of my wonderful TBG device. Despite these troubles, we managed to capture a complex and beautiful set of measurements.

Brown-Zak oscillations

At simple rational magnetic fields $\Phi/\Phi_0 = p/q$ the system is described by Bloch bands, and the magnetic field experienced by the Bloch electrons is zero. The resulting oscillations in magnetoresistance can persist above room temperature and are referred to as Brown-Zak oscillations [144, 145]. I clearly observed Brown-Zak oscillations at $\Phi/\Phi_0 = 1/4$ and $1/5$ in Figs. 3.4 and 3.13. In the high-field measurements, I again observed them at $\Phi/\Phi_0 = 1/3, 2/5, 1/2$, and $3/5$.

Near $1/2$ and $1/3$ flux in graphene/hBN superlattices, Ref. [145] (Fig. 4) observes features similar to those in the dashed circle in Fig. 6.5. In my measurement, the Landau minifan emitting from $n/n_s = 2$ and $\Phi/\Phi_0 = 1/2$, on the hole side of $\nu = 4$, bends towards horizontal. Such bending cannot be explained in a single-particle, rigid-band Hofstadter picture, even with strain. Ref. [145] does not give an explanation for this behavior. It is possible that the behavior is caused by spontaneous miniband polarization, as seen at low field in magic-angle samples (the cascade of Dirac revivals). For example, Ref. [138] (Fig. 1d) shows similar behavior of Landau levels upon crossing integer moiré filling in twisted trilayer graphene.

Asymmetry and potential spin-selective transport

There is a very curious asymmetry to the measurements. The fan diagram has some degree of electron-hole symmetry: most features at a given density and field are qualitatively duplicated at opposite density. Several examples are noted in numbered

circles in the top panel of Fig. 6.5. There is asymmetry to the quantitative resistivities of these mirrored states, however. If you squint at the figure, it looks unbalanced to the right.

For instance, the circled numbers 1 and 2 denote regions on either side of $\nu = \pm 4$. Consider the pair of regions on the hole side, the 1 and 2 near $n/n_s = -1.5$. The region around 1 is clearly an order of magnitude lower resistivity than the region around 2. Curiously, the opposite behavior is the case on the electron side of CNP, near $n/n_s = 1.5$. Points 3 and 4 denote a split LL originating from CNP at half flux. Like with points 1 and 2, for each pair on each side of CNP, the region on the electron side has significantly lower resistivity than the corresponding region on the hole side.

There is actually a simple phenomenological correspondence between this behavior and the BM Hofstadter's butterfly model with Zeeman splitting, as illustrated in Fig. 6.6. The numbers one to four in Fig. 6.5 correspond to regions of the butterfly with only one spin species at the Fermi level. In all cases throughout the butterfly where the correspondence between the measured data and the computed spectrum is clear, any time the Fermi level is within a blue Landau level, the resulting resistivity is lower than both the neighboring red regions and the red region at opposite energy.

Spin-selective transport has been demonstrated in giant magnetoresistance structures [87] as well as WSe₂ bilayers under high fields [146]. I do not have a satisfying explanation for this behavior in my sample, however it appears in multiple measurements at lower field.

Similar behavior appears in my Landau fan diagrams taken at Stanford. See Figs. 3.13, 3.14 (right panel), and A.16. In all cases, there are horizontal lines of decreased resistivity within Landau levels that switch sign upon crossing LL gaps. See Fig. A.31 for line cuts at constant field. The horizontal lines are approximately evenly spaced in $1/B$ with a spacing of 0.05 T^{-1} , corresponding to a density of roughly $5 \times 10^{11} \text{ cm}^{-2}$ with no factors for degeneracy included.

The behavior is reversible with applied back gate voltage. In Chapter 3, I noted that in Fig. A.15's right panels ($B = 8 \text{ T}$), there are oscillations as displacement field is tuned, and that these oscillations are closely related to the thin back gate voltage. These oscillations are precisely those described in the previous paragraph.

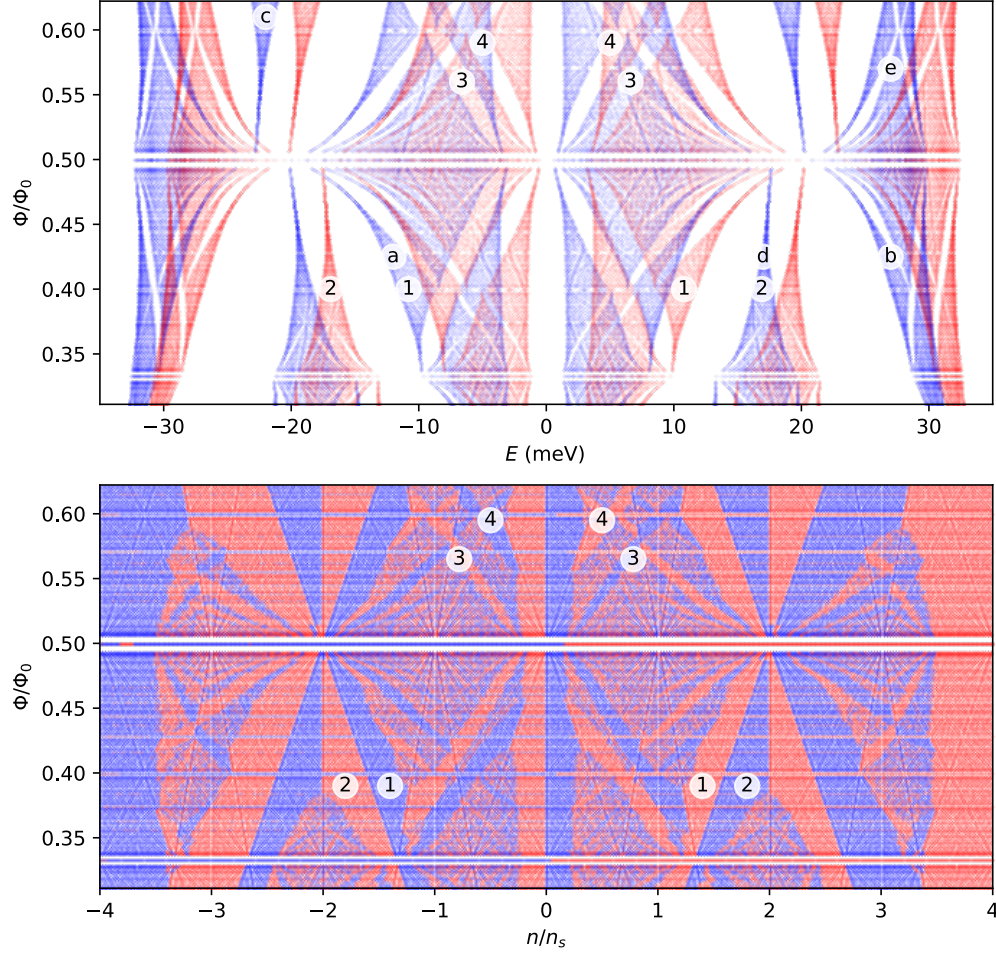


Figure 6.6: **Spin-dependent transport?** **Top:** Spectrum for strained BM model between 14 and 28 T with $\theta = 1.38^\circ$ and $\epsilon = 0.2\%$ up to $q = 79$. Blue points are the spin-aligned states and red are antialigned. The numbered circles indicate the same regions labelled in Fig. 6.5. Letters indicate regions labelled in Fig. 6.7. **Bottom:** Same data, plotted vs density.

I would hazard a guess that I have quantum oscillations in my few-layer graphite gate (perhaps with density set by the silicon gate's 20 V), and these oscillations are influencing my device's transport, somehow in a spin-dependent manner.

Vertical lines

There are strong vertical lines of minimal resistivity at $n/n_s = \pm 2$ emanating from half flux, indicated in the bottom panel with blue lines. This is the location where $\nu = \pm 4$ from $n/n_s = 0$ and ± 4 intersect. These lines typically have longitudinal resistivities of a few hundred Ω , but the resistivity drops to zero in some places along the lines. They have zero Hall resistance. I expect that the absence of both longitudinal and Hall resistance is not of superconducting origin, but instead enabled by inhomogeneity.

These lines are generically expected in doubled (because of Zeeman splitting or otherwise) Hofstadter's butterflies where two equal and opposite Středa lines meet. For instance, in Fig. 4.8, right panels, a vertical line is visible where $\nu = \pm 8$ intersect. The explanation for these features is the same as in Fig. 4.9.

Potential fractional Středa lines

The red lines in the lower panel of Fig. 6.5 have either fractional slope or fractional intercept. It is difficult to precisely state which, because the lines are broadened by disorder, and quantum capacitance means that small offsets in both slope and intercept are to be expected [147].

I assign $(1/2, 3)$ to the solid red line, and I am reasonably confident that it is the correct assignment. To fit it, I placed points along the minimum by eye, then did a least squares fit. The result was nearly precisely $(1/2, 3)$. The most likely possible alternative is $(2/3, 8/3)$.

The two dotted red lines in the schematic are $(4/3, 4/3)$ (visible in two contact pairs) and $(-2/3, -8/3)$ (visible in four contact pairs). For these, the same procedure of placing points by eye and doing least squares cannot distinguish between the $1/3$ fractional denominators and the nearby $(3/2, 1)$ and $(-1/2, -3)$ lines. There may

be hints of a $(-4/3, -4/3)$ line in a few contact pairs, but it is so faint and hard to distinguish from $(-2, 0)$ that I cannot say for sure.

These lines all appear to be associated with $\Phi/\Phi_0 = 1/2$, which would classify them as either symmetry-broken Chern insulating states (fractional s , $t \in \mathbb{Z}$) or fractional Chern insulating states (fractional s , fractional t). They are only seen in high-resistivity regions of the fan with regards to the asymmetry previously mentioned, although this may be a coincidence.

Possible fractional Chern insulating Hall plateaus

Only one Hall contact pair out of three (7 - 17) shows quantized R_{xy} values, even for integer ν . Fig. 6.7 shows data from this contact pair, plotted in multiple ways. Note that the measurements from 14 to 17 T were taken the day before the measurements taken above 17 T. There is a sharp cutoff in many of the features to follow at 17 T: this is not a physically-interesting feature, but instead another manifestation of our struggle with noise at MagLab.

The top panel shows every line cut at constant field plotted with transparency. This style of plot³ makes Hall plateaus stand out, because they overlap and thus appear as dark horizontal lines. Horizontal lines are apparent at integer filling factors -8, -4, -2, 2, 4, and 8. Extremely clear horizontal lines also appear at $\pm 8/3$. Fascinatingly, there is more $-8/3$ than any other ν . Weak lines appear at $-8/5$ near $n/n_s = 3$ and at $8/5$ near $n/n_s = 3.5$.

The bottom left panel shows the n and B distribution of the $+8/3$ states (a, b, c) in blue and $-8/3$ states (d, e) in red. The solid black lines are $\nu = \pm 4$. The dashed line passing just to the left of region c is $(-2/3, -8/3)$, and the dashed line just to the left of region d is $(2/3, 8/3)$. These are the same lines that we see as likely minima in ρ_{xx} . Region b is just to the right of $(11/3, -8/3)$, and region e surrounds $(5/3, 8/3)$. These are all marked with dotted lines. Regions a and e are broad enough to span multiple possible s values. I hesitate to confidently assert that these are fractional Chern insulating states, because they do not clearly follow straight lines in field and density.

³I've heard it named "smoke plot," "waterfall plot," and "ghost plot."

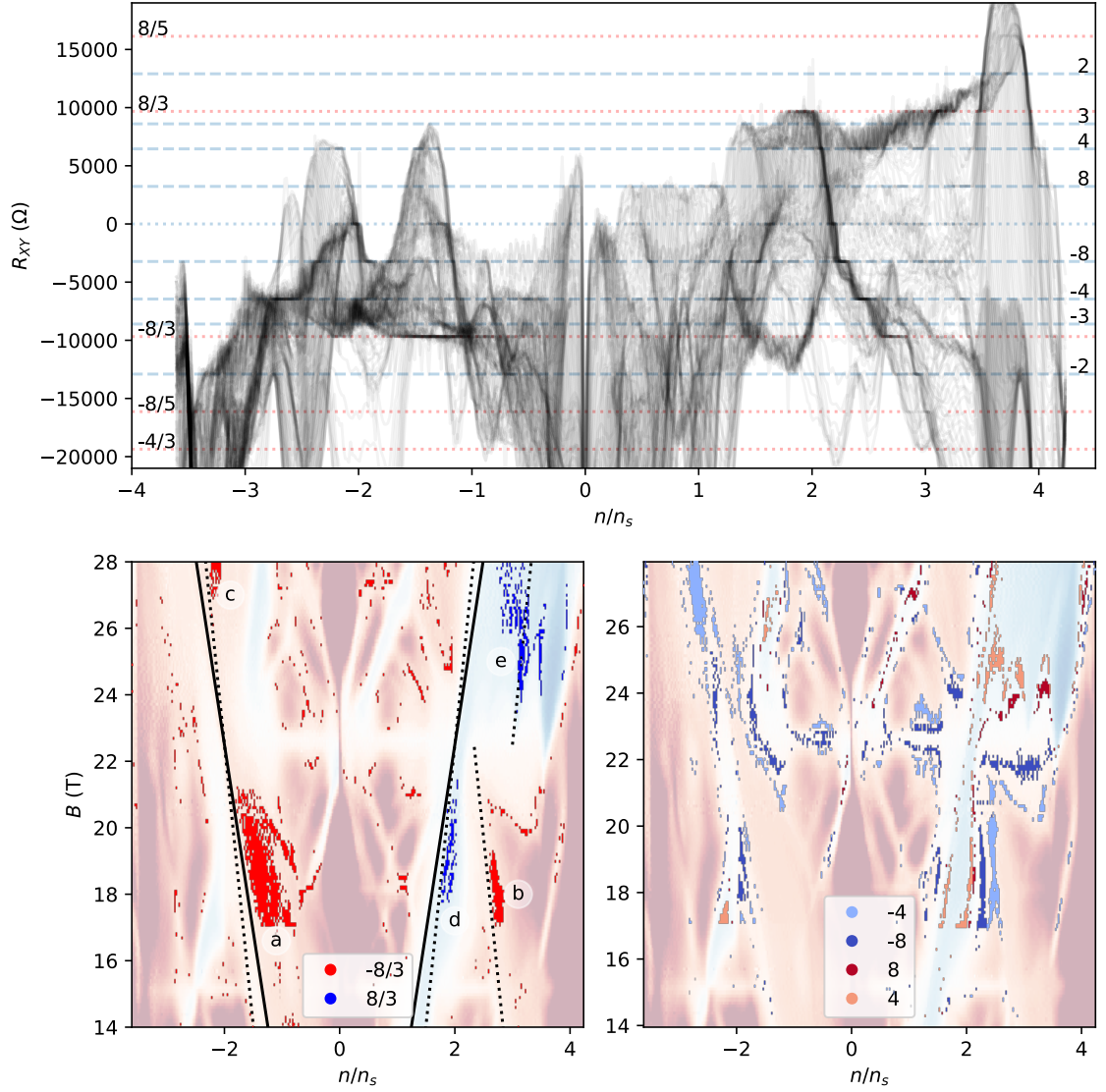


Figure 6.7: **Fractional Chern insulating Hall effect.** Contact pair 7 - 17, described in the text. **Top:** Line cuts of R_{xy} for all fields, plotted with transparency. Horizontal dashed blue lines are indicated integer ν . Horizontal dotted red lines are indicated fractional ν . **Bottom left:** Hall resistance, plotted with transparency. In blue and red are points with Hall resistance within $50 \, \Omega$ of $\nu = +8/3$ and $-8/3$ respectively. Letters and lines are described in the text. **Bottom right:** Same as bottom left, $\nu = \pm 4$ and ± 8 .

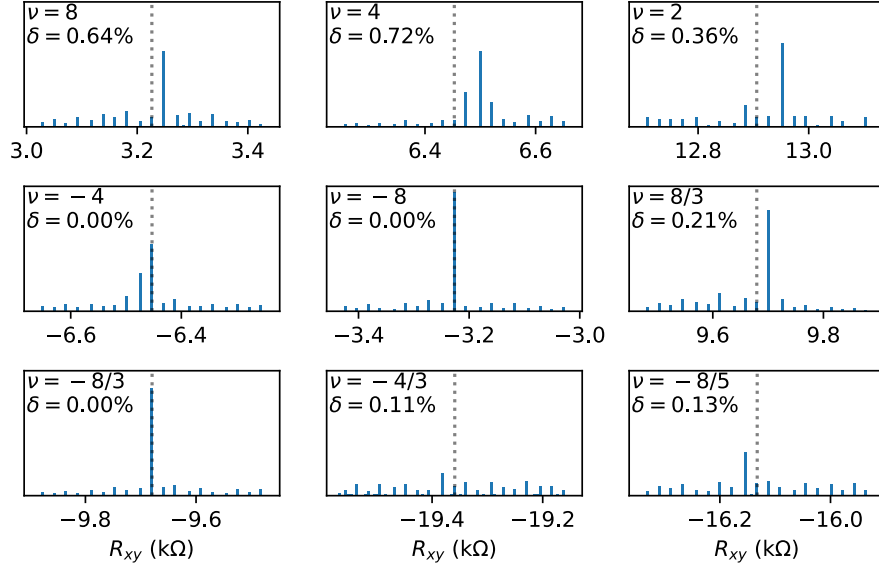


Figure 6.8: **How well quantized is the FCI?** Described in the text.

The bottom right panel shows $\nu = \pm 4, \pm 8$ states. There are a few things to note. First, as with the FCI states, even the integer states do not always follow the expected straight lines. In particular, near the bending feature at half flux and $n/n_s \approx 3$, all of these integers visibly bend. In fact, the $-8/3$ FCI state (bottom left panel) bends here as well!

To hammer the asymmetry point home, regions a to e are indicated in Fig. 6.6. Note that b and e are hard to accurately place within a band; the locations in Fig. 6.6 are my best guess. These FCI states are entirely located within “blue” bands. This asymmetry point may be a coincidence.

When I took this measurement, I committed a cardinal sin of experimental physics. Because of the noise in the measurements, I did not carefully analyze the Hall data until I had returned from MagLab. I assumed they were uninteresting, ruined by the noise. I did not, therefore, notice the fractional quantized Hall plateaus until it was too late to investigate them further! Had I noticed at MagLab, I would have more carefully measured the precise value of R_{xy} within the $\pm 8/3$ regions.

Alas, the best I can do is summarized in Fig. 6.8. Each panel shows a histogram

of R_{xy} in close proximity of the specified ν . It's not precisely R_{xy} , though. Instead, it is $V_{xy}/(1 \text{ nA})/g$ for some gain factor g , typically a few percent away from 1. I use 1 nA for current instead of the measured value because the measured value has massive noise, as mentioned earlier. This correction factor g is not as arbitrary as it might at first seem. Voltage amplifiers are almost never precisely the gain that they claim, but almost always a few percent off.

Fortunately, within the same measurement we have access to one of the best resistance standards: the integer quantum Hall effect. In this case, I tweaked g until the $\nu = -8$ peak, the strongest IQH peak, fell directly on the expected resistance of $-h/8e^2$. In each panel, I plot a ν value. The spacing of the data points is set by the resolution of the lock-in amplifier. I compute a metric of quantization as

$$\delta = \frac{\text{argmax}_R - R^*}{R^*} \quad (6.3)$$

for $R^* = h/\nu e^2$, the expected resistance. This metric is not perfect. It only considers the highest point within the window and not any shoulders, for instance. Still, it should do a reasonable job, and I am not interested in perfection here. Within this scheme, the $-8/3$ plateau is perfectly quantized to the experimental precision, better than one part in a thousand.

The other peaks are not as well quantized. The $8/3$ peak is roughly two parts in a thousand, but $\nu = 4$ is almost a percent off. Terrible! It is possible to adjust g to minimize the sum of δ s, and it may even be physically meaningful to do so,⁴ but I do not perform such optimization here. The proper thing to do was to more carefully measure these plateaus when I was at MagLab.

Note that R_{xx} does not identically go to zero where the FCI states exist in all four adjacent longitudinal contact pairs, but only in some (Fig. A.29). In others, it is in the 10 to 100 Ω range. From

$$\sigma_{xy} = \frac{R_{xy}}{R_{xy}^2 + \rho_{xx}^2}, \quad (6.4)$$

if it is σ_{xy} that is quantized, then to find R_{xy} quantized to within 10 Ω for $\nu = 8/3$,

⁴For instance, if the SR550 preamplifiers that I used at MagLab do not have equal gains for opposite polarities then we shouldn't expect the fit for $\nu = 8$ to match $\nu = -8$.

$|\rho_{xx}|$ must be less than roughly $300 \, \Omega$. This is a large budget, and may help explain the lack of straight lines of quantized Hall effect: it is possible that the system is in a fractional gap of one set of bands, but also within another band that has very low resistivity.

Still, this is likely strong enough evidence to claim fractional Chern insulating Hall plateaus in twisted bilayer graphene. Fractional quantum Hall states, that is $s = 0$, t fractional, have been observed in graphene samples with no superlattice ([11], for example) and with a superlattice [147,148]. Similarly, symmetry-broken quantum Hall states with integer t but fractional s have also been observed in graphene superlattices [147,148] and TBG [99,149].

Fractional Chern insulating behavior ($s \neq 0$, t fractional) has been reported in graphene superlattices in capacitance measurements [147] above 25 T, magic-angle TBG from local compressibility measurements [150] above 5 T, and in dual-gated bilayer graphene/hBN superlattices [151] above 14 T as minima in R_{xx} .

6.3 Theory and experiment comparison

In Chapter 5, we used moiré parameters $\theta = 1.38^\circ$, $\epsilon = 0.2\%$, and $\varphi = 0$ to model my device. We settled on these parameters from the locations of the van Hove points in density—inferred from the onset of MR and subtle cusps in longitudinal resistivity at low field—and the ratio of Fermi surface areas from low-field quantum oscillations between CNP and the first van Hove point. All of these features are present in fields below 1 T. Fig. 6.9 shows calculated inverse density of states (top panel) and, for comparison, measured resistivity (bottom panel) from 1 to 28 T.

As I described previously, the strained and Zeeman-split calculation does an excellent job of matching the sequence of quantum oscillations emitting from charge neutrality: the main sequence is $\nu = \pm 4, \pm 8, \pm 12, \dots$. This sequence splits and bends at very nearly the same density/field points in both experiment and theory, for instance, the $\nu = 8$ gap closes up just before $n/n_s = 2$ and $B = 11$ T. In between, $\nu = \pm 2, \pm 6, \pm 10$ appear at higher fields than do the main sequence. They disappear at very similar points in both field and density.

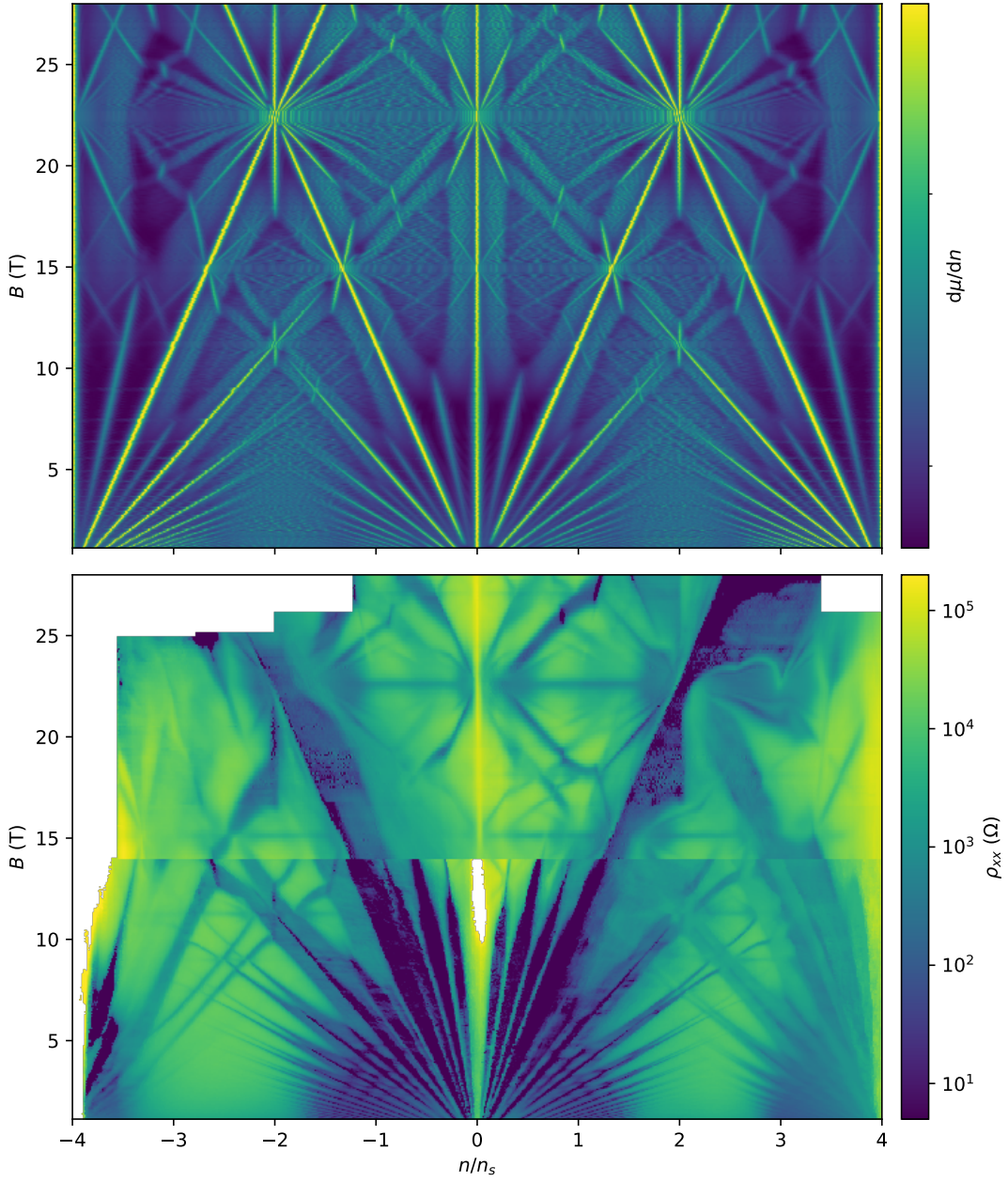


Figure 6.9: **Comparison between theory and experiment.** **Top:** Computed inverse density of states for $\theta = 1.38^\circ$, $\epsilon = 0.2\%$, $\varphi = 0$, $g = 2$, and $\gamma = 0.1$ meV. **Bottom:** Figs. 3.13 and 6.5 overlaid.

Landau level gaps projecting from $B = 0$ and $n/n_s = 4$ are actually quite well fit by the computation, in sequence and in shape. In experiment, there doesn't seem to be a fully-developed gap, however: ρ_{xx} does not go to 0 even in the $\nu = -4$ gap.

Where the calculation has sufficient resolution, the behavior of split lines at intersections is identical between theory and experiment. As I showed in Chapter 4, this behavior is generic in any anisotropic, doubled butterfly.

There are, of course, differences between computation and experiment. Some, like the absence of odd-integer quantum Hall states, are to be expected from the symmetries of the model. Additionally, the inverse density of states will not capture features such as Brown-Zak oscillations (horizontal lines of low resistivity at simple magnetic flux fractions). The model also does not capture the delicate horizontal lines (constant field) within Landau levels near CNP (Fig. 3.14, right panel), discussed in the previous section.

Most obviously, of course, is the electron-hole asymmetry. I mentioned that the model is a very good fit on the electron side, but it is clearly inadequate at capturing the quantum oscillations emitting from $n/n_s = -4$. To some extent this is expected—the BM model from Chapter 5 has electron-hole symmetry—and a more sophisticated model (such as Refs. [132, 133]) may resolve the disagreement.

Overall, the agreement between theory and experiment, without any fine tuning beyond what was performed in Chapter 5, is strong, and in my opinion conclusive, evidence in favor of the hypothesis that strain and Zeeman splitting explain my results.

Chapter 7

Concluding remarks



WHAT started as an attempt to understand the unusual magnetotransport properties of a particular device ended up as a thorough investigation of the role that uniaxial heterostrain plays in TBG above the magic angle. I found that even miniscule amounts of uniaxial heterostrain have dramatic effects on the energetics and transport of TBG. This presents a major problem for studies of TBG near magic angle: it is not straightforward to back out the strain from transport when correlated effects dominate, and as of now we do not have an effective way of controlling the strain. The problem is not unique to TBG, either. Any twisted homobilayer moiré system will suffer from the same underlying issue that the effects of small amounts of heterostrain are magnified by the small twist angle.

There are a few plausible ways to mitigate the problem. The first is to study related moiré systems that are intrinsically predisposed to less strain. If you assume that heterostrain comes hand in hand with twist angle inhomogeneity, then systems with less twist angle inhomogeneity should have less heterostrain. TBG, for example, is rotationally metastable: it prefers to relax down to Bernal stacking [57, 58]. At higher twist angles, this desire to relax becomes energetically less favorable. So we would expect that twisted graphene systems at higher twist angles may have less heterostrain.

Mirror-symmetric twisted trilayer graphene has a magic angle of $\sqrt{2}$ times twisted bilayer graphene's magic angle, roughly 1.6° [138]. One might suspect that adding an

additional layer ought to only make the situation worse: now there are two possible moirés to heterostrain. However, theory predicts that this system energetically prefers A-twist-A stacking, where the bottom and top layers perfectly align. There is some evidence for this claim in transport measurements [138] and STM measurements [152]. The system is still not strain-free, featuring distortion in stripes attributed to maintaining the A-twist-A stacking across domains.

The goal of zero heterostrain may be too much to ask for. Instead, we may wish to either measure the heterostrain within either partial or completed devices, or, even better, control the heterostrain *in situ*.

I spent significant time with others in my group trying to make the former a possibility, using AFM-based techniques similar to those in Ref. [55]. This ended up being extremely difficult, for various reasons. Although we eventually succeeded in establishing a reliable protocol for measuring moirés and analyzing the results, we have not managed to measure moirés that are buried under hBN and graphite gates.

An even more exciting dream would be to control the strain in transport devices. Some work has been done in this area. In Ref. [153], researchers deposited a stressed, thin film on the top layer of graphene. Some of the stress was transmitted to the graphene, as evidenced by Raman spectroscopy. While this method is tantalizing, it is not trivial to extend it to fully encapsulated and doubly-gated samples, because the encapsulation will shield the graphene from the straining film. Another try is described in Ref. [154], in which the authors bent a long strip of monolayer graphene by pushing on one end with an AFM tip. Again, it would be difficult to extend this scheme to fully-encapsulated transport devices.

In a way, this dissertation presents a pessimistic result. Heterostrain in moiré materials can be seen as an intrinsic problem, reducing reliability and repeatability of the measurements and confounding theoretical descriptions of the system. Unless it can be controlled for in some way, it will permanently render transport measurements on moiré systems suspect. If it *can* be measured or controlled for, then heterostrain in moiré materials becomes an incredibly sensitive tuning knob, the so-called “strain-tronics.” We will have to wait and see.

Appendix A

Additional measurements

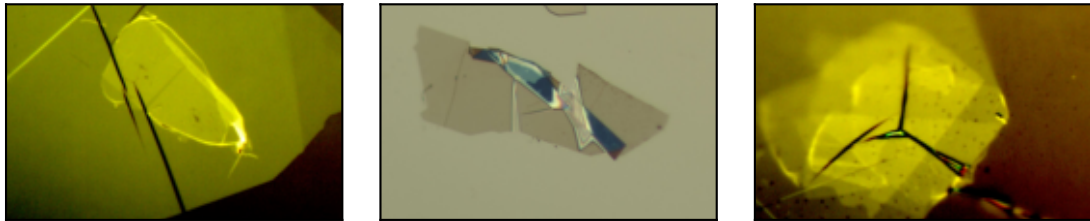


Figure A.1: **A few examples of failed stacks.** These are optical images of stacks on the stamp. **Left:** Graphene is the darker grey in the center on top of the lighter grey hBN. The graphene did not tear cleanly at the edge of the hbn. instead, it wrinkled over itself on two edges, and tore down the middle. A fold has also appeared in the hBN. **Middle:** After picking up top hBN, then twisted bilayer graphene, the bottom hBN failed to pick up neatly and caused the top hBN to tear and detach from the stamp. **Right:** A fold appeared in the hBN, passing directly through the twisted bilayer graphene.

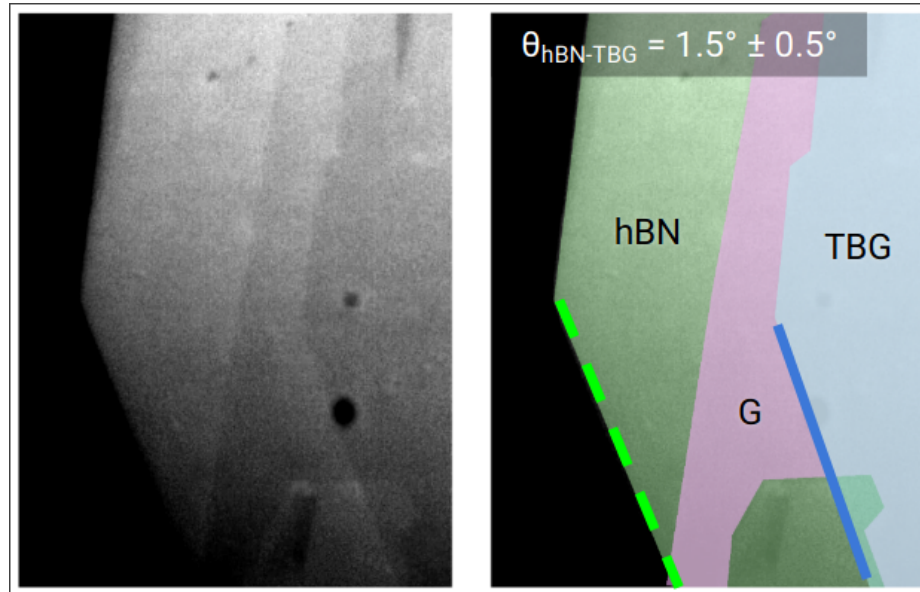


Figure A.2: **Optical evidence of hBN alignment.** The green dashed line in the right panel is an edge of the hBN, while the blue line is the edge of a layer of graphene.

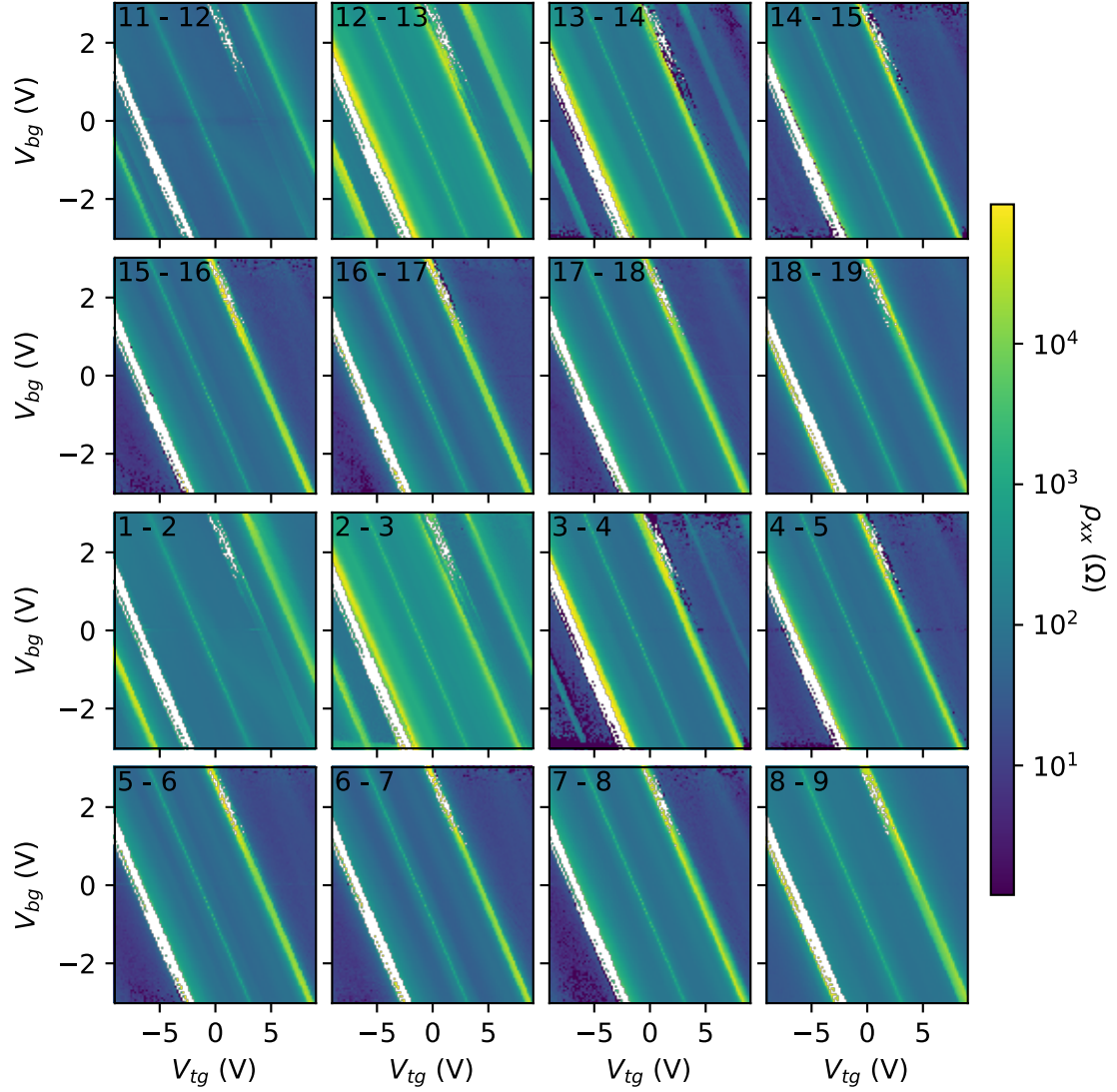


Figure A.3: **2D gate maps at 0 field and 1.7 K.** White regions indicate that the lock-in signal was lost, usually because the device became so resistive that the current dropped. Note that contacts 1 to 4 and 11-14 have an extra insulating peak at high densities. I assume this is a domain of lower twist angle that cuts through the device.

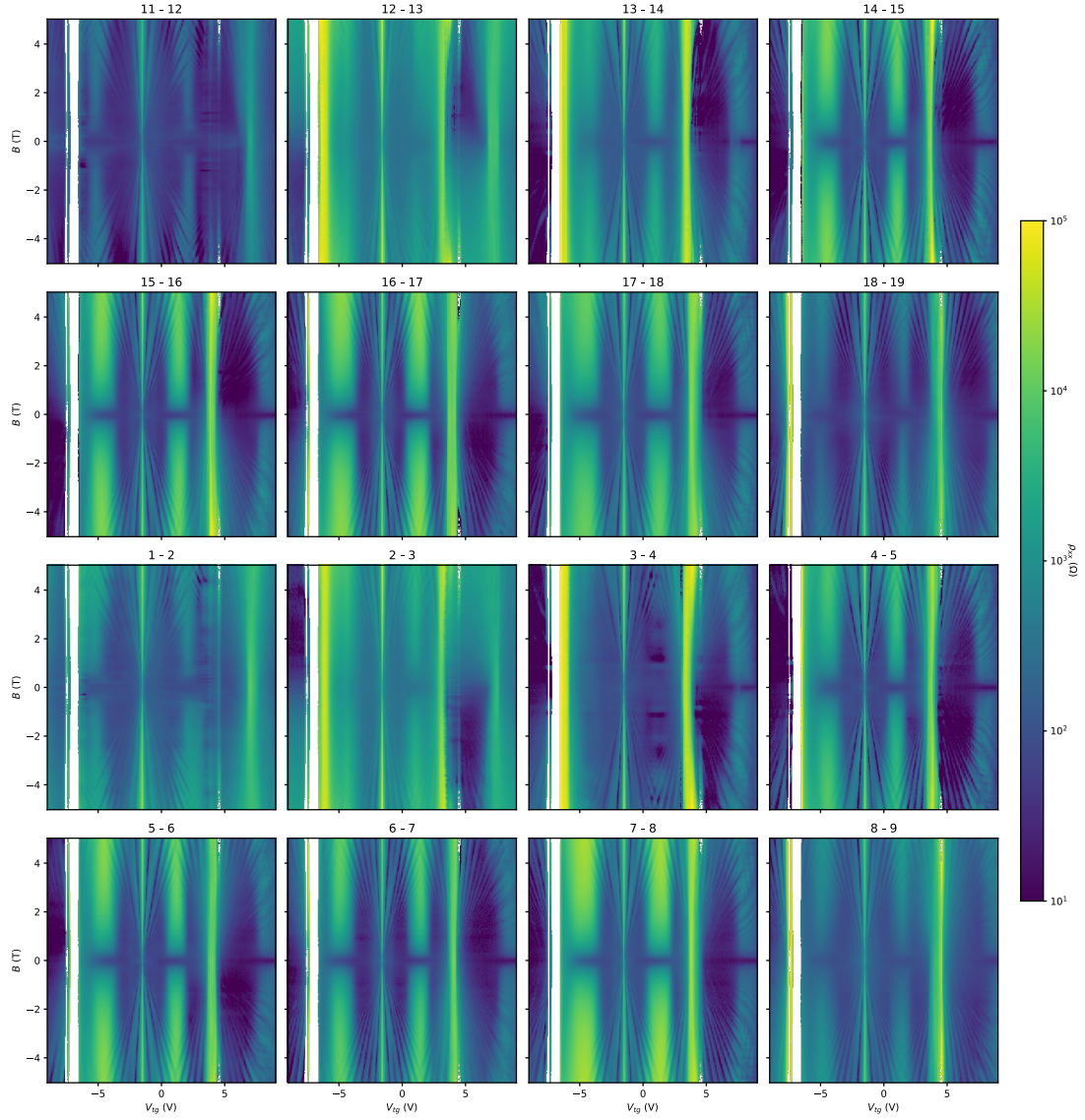


Figure A.4: Landau fan out to 5 T at 1.7 K. Longitudinal.

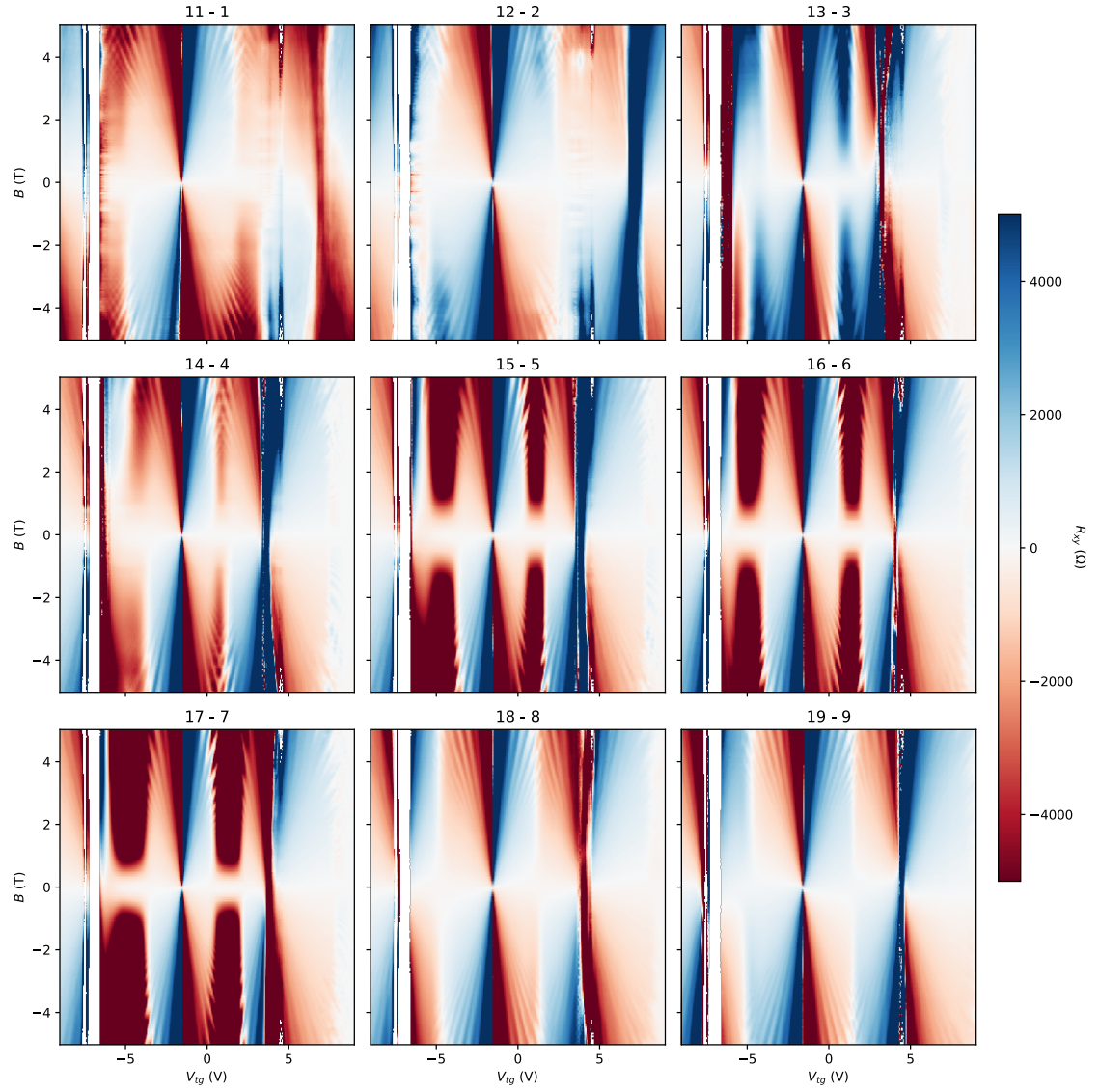


Figure A.5: Landau fan out to 5 T at 1.7 K. Hall.

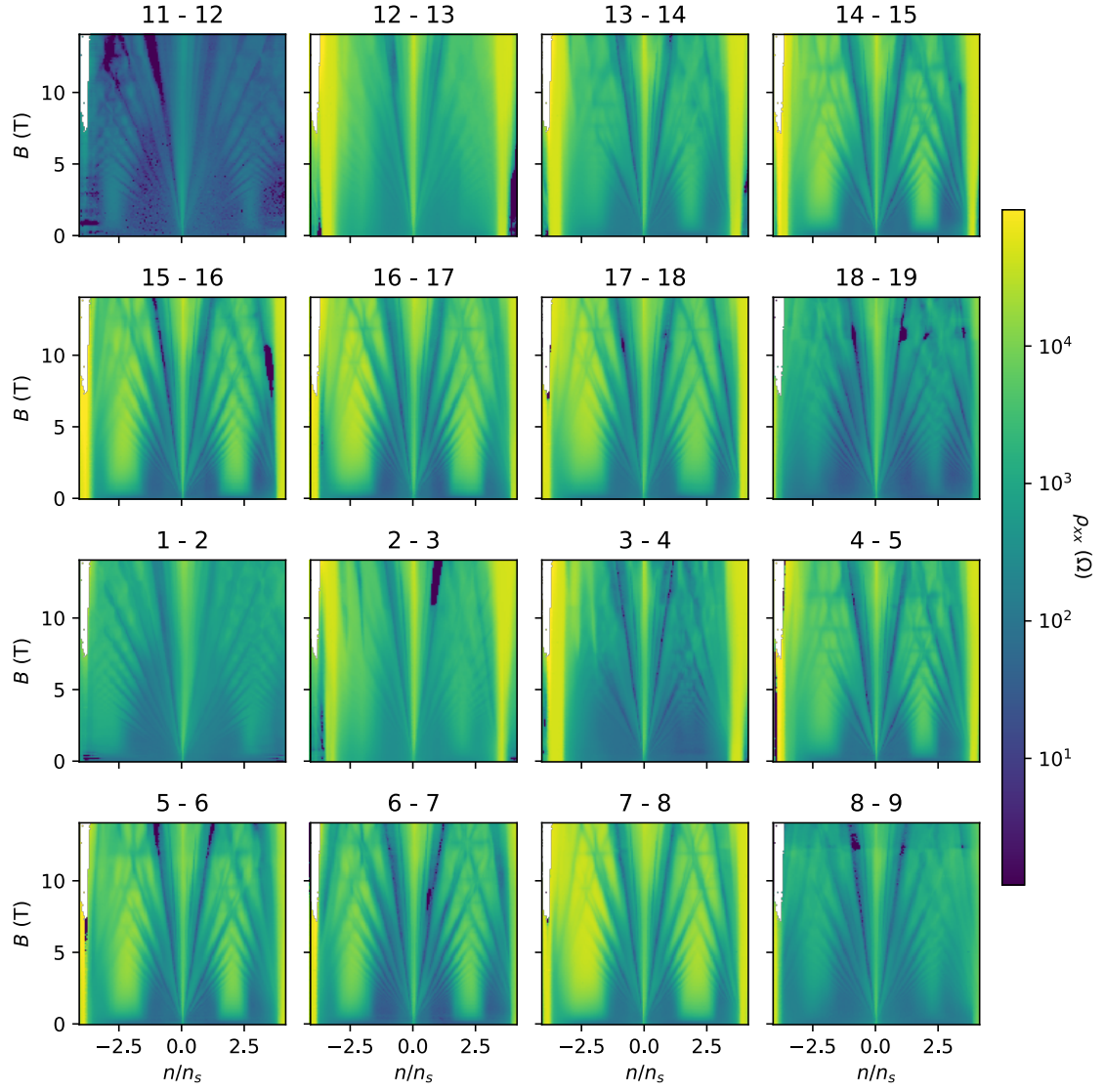


Figure A.6: **Landau fan out to 14 T at 1.7 K. Longitudinal.** Note that contact pairs near 3 - 4 have features at 8 T projecting back to half filling of holes. These are precisely the contact pairs where I observe superconductivity at half-filling of holes.

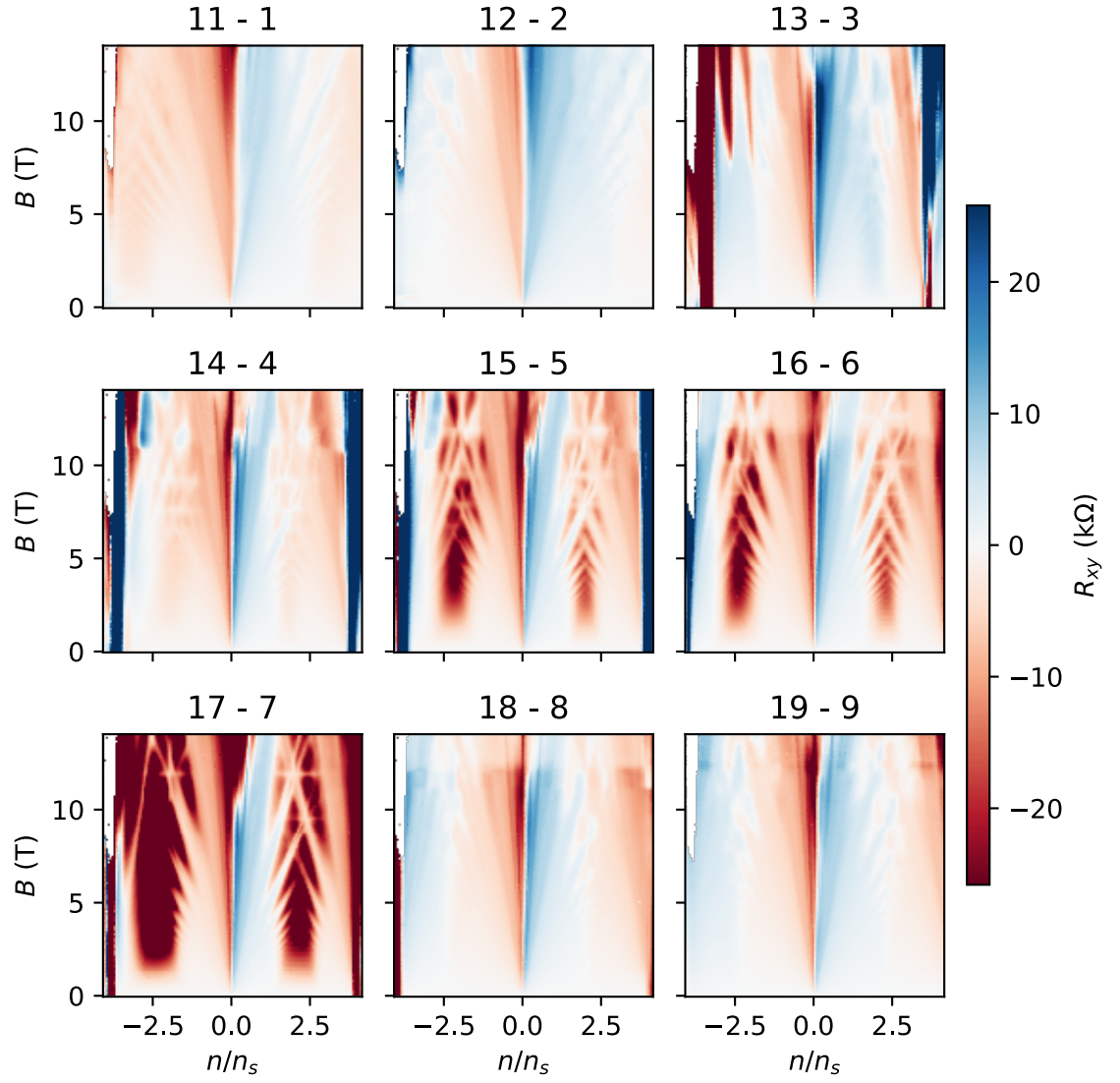


Figure A.7: **Landau fan out to 14 T at 1.7 K. Hall.** Note the significant mixing of XX into XY in contact pairs 15 - 5 to 17 - 7.

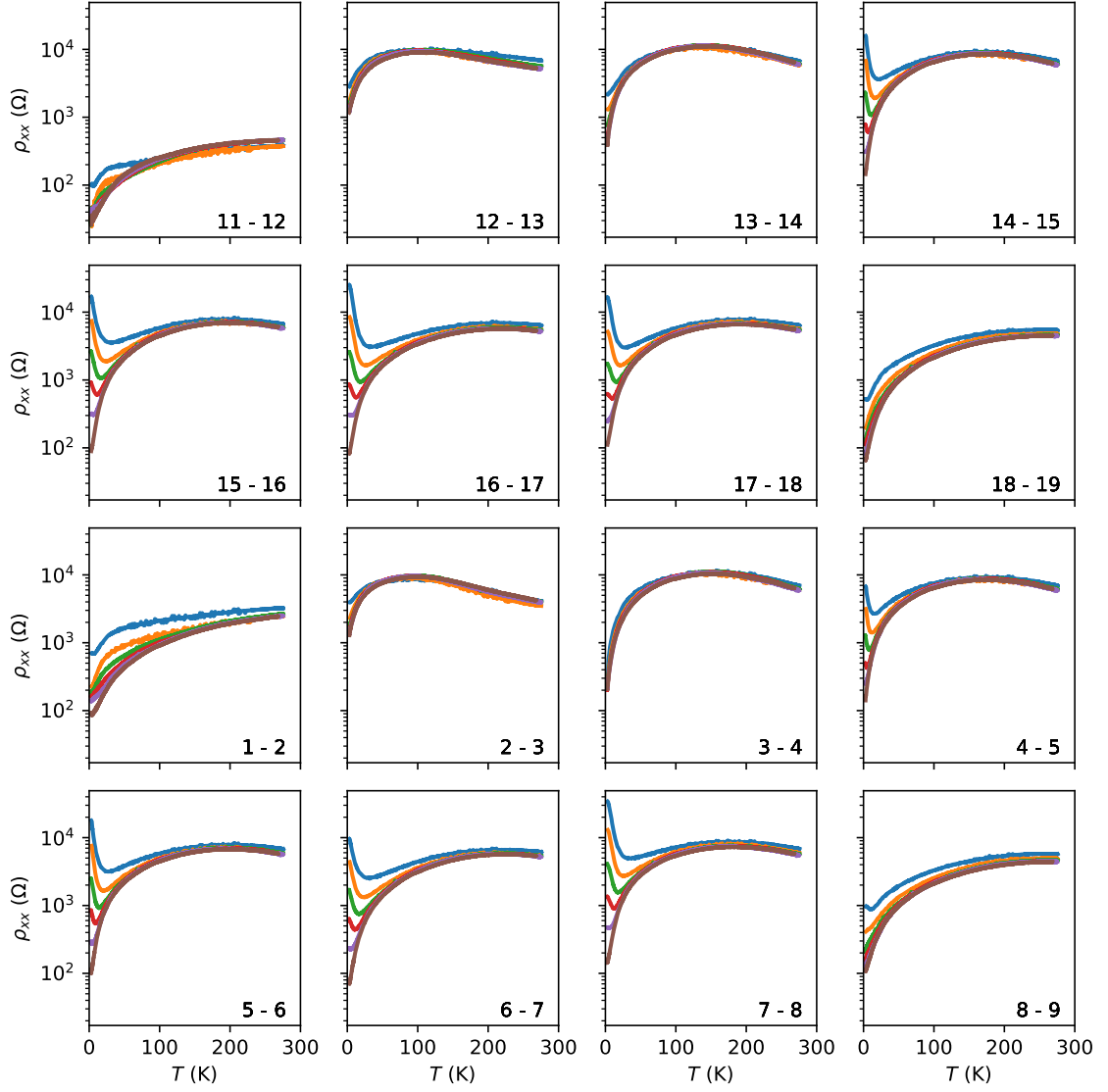


Figure A.8: Temperature dependence of MR at fixed fields.

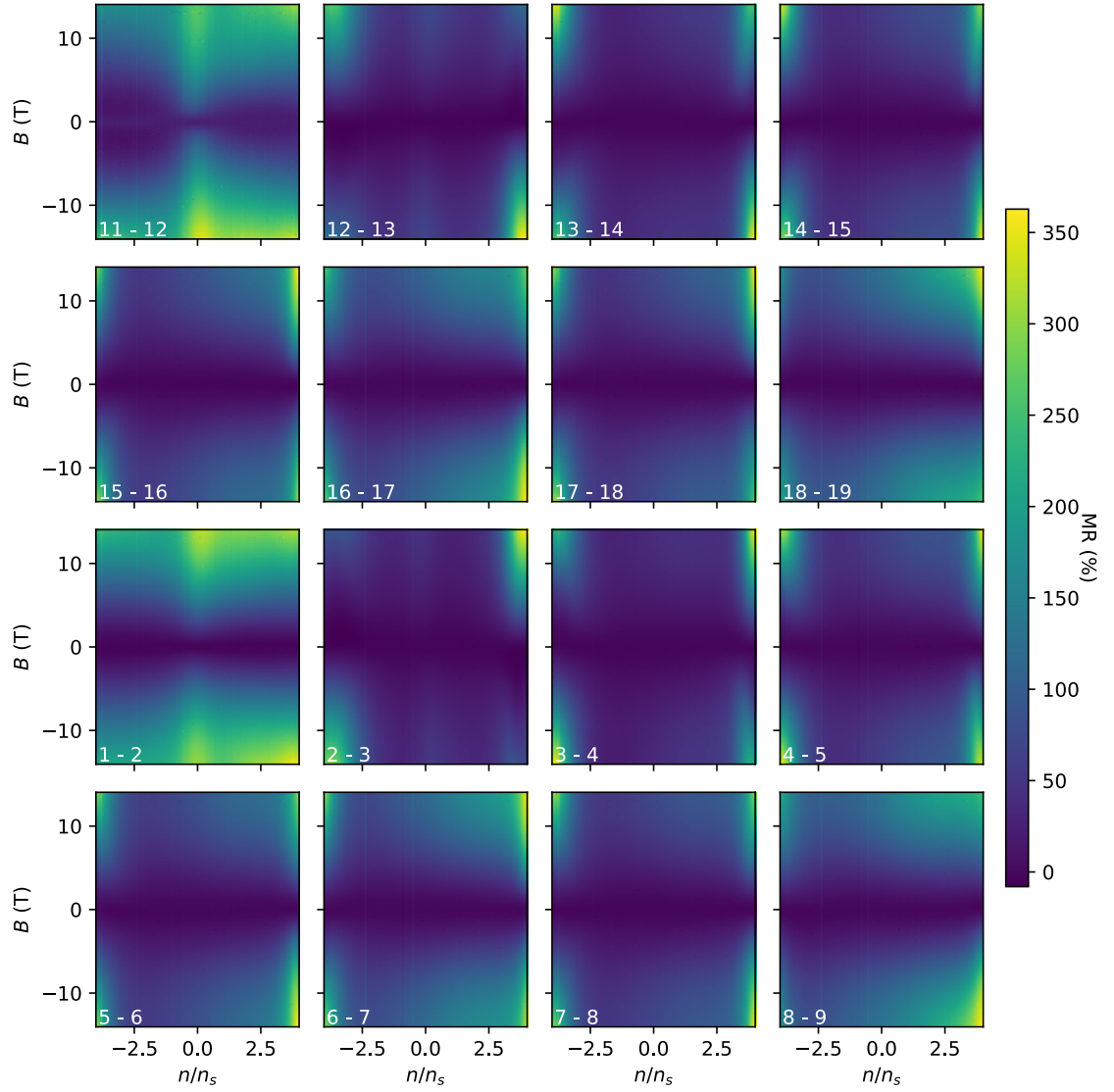


Figure A.9: Longitudinal MR vs field and density at room temperature.

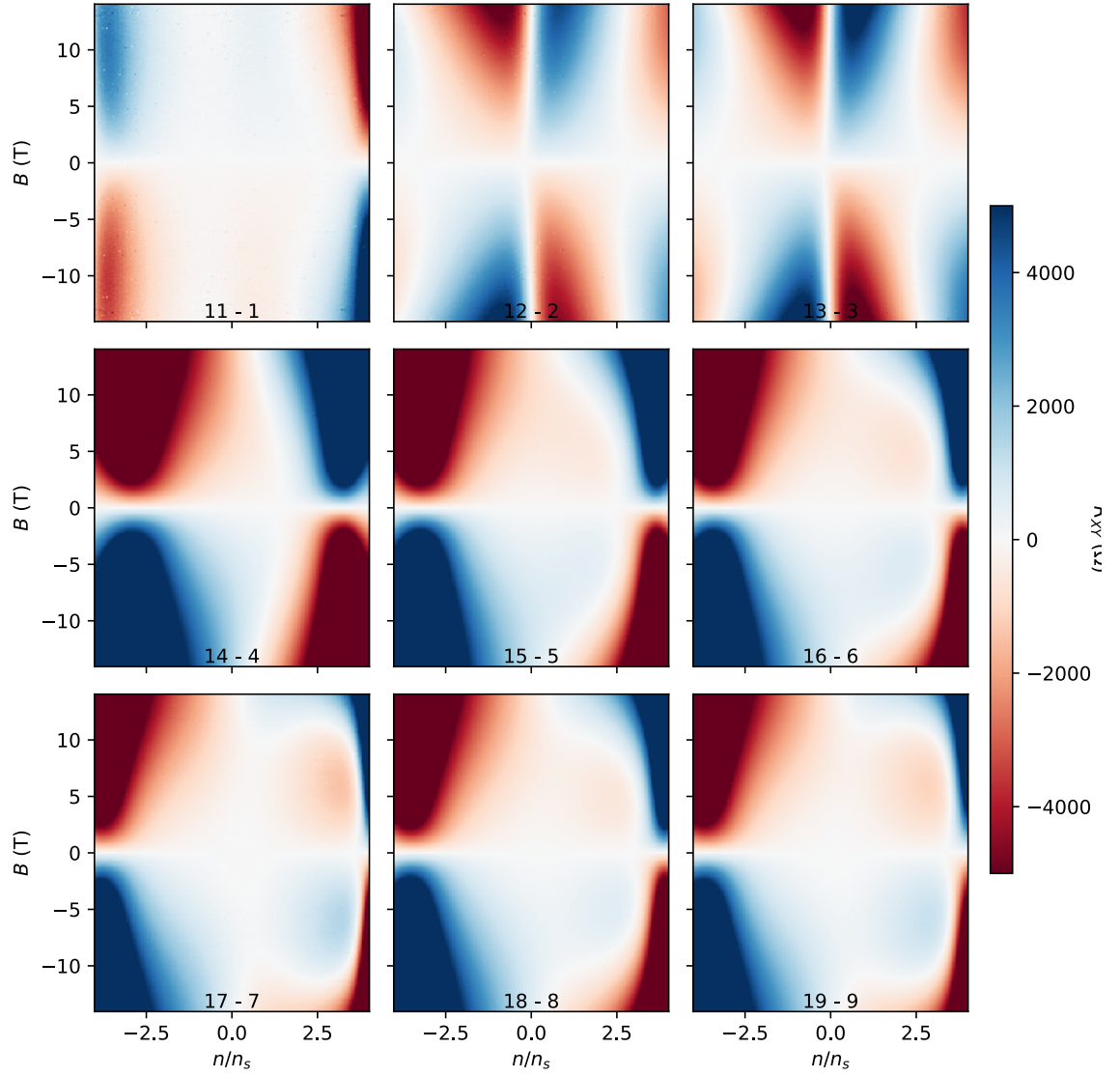


Figure A.10: Transverse resistance vs field and density at room temperature, antisymmetrized.

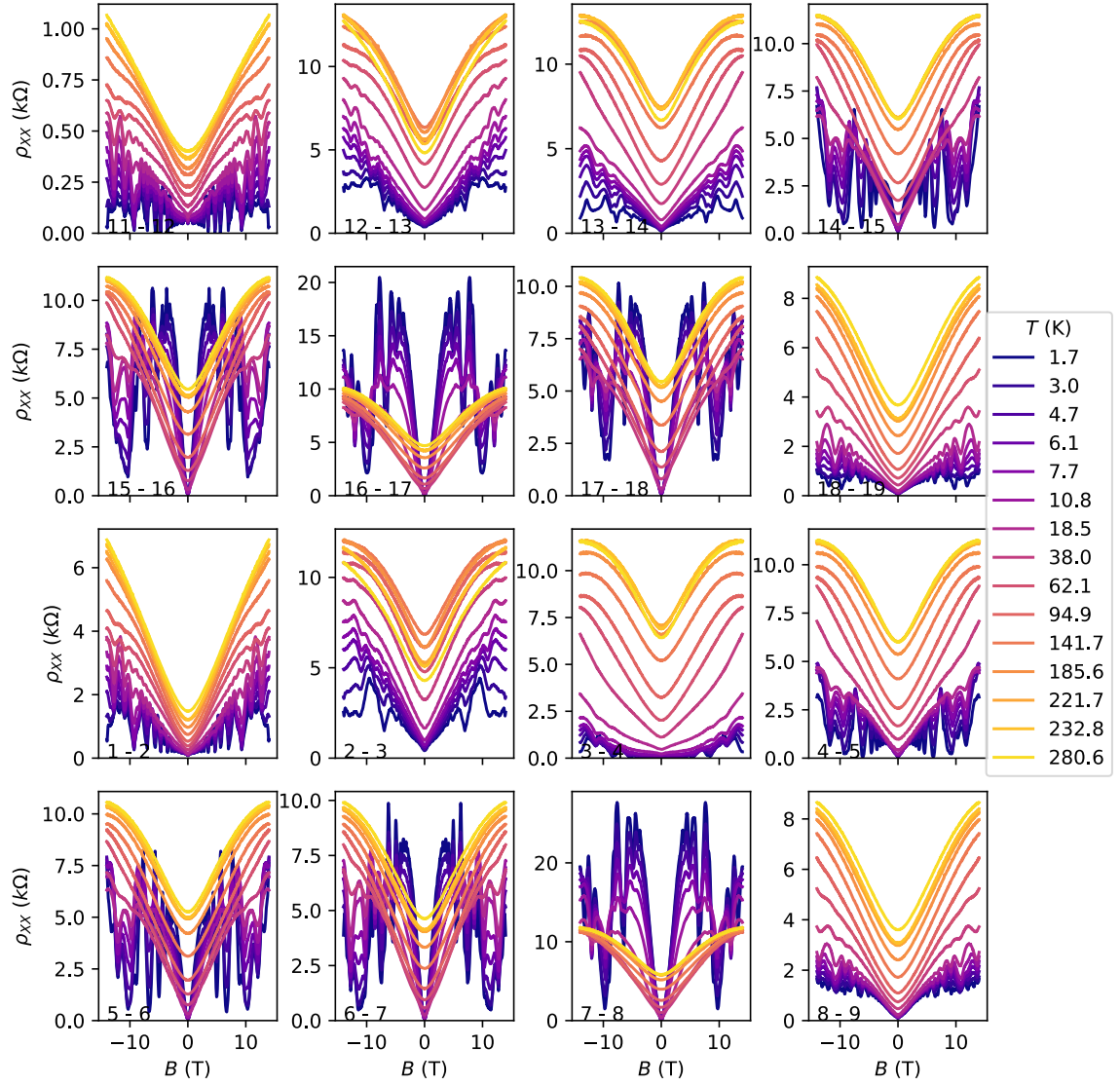


Figure A.11: Longitudinal resistivity vs field and density at various temperatures, symmetrized.

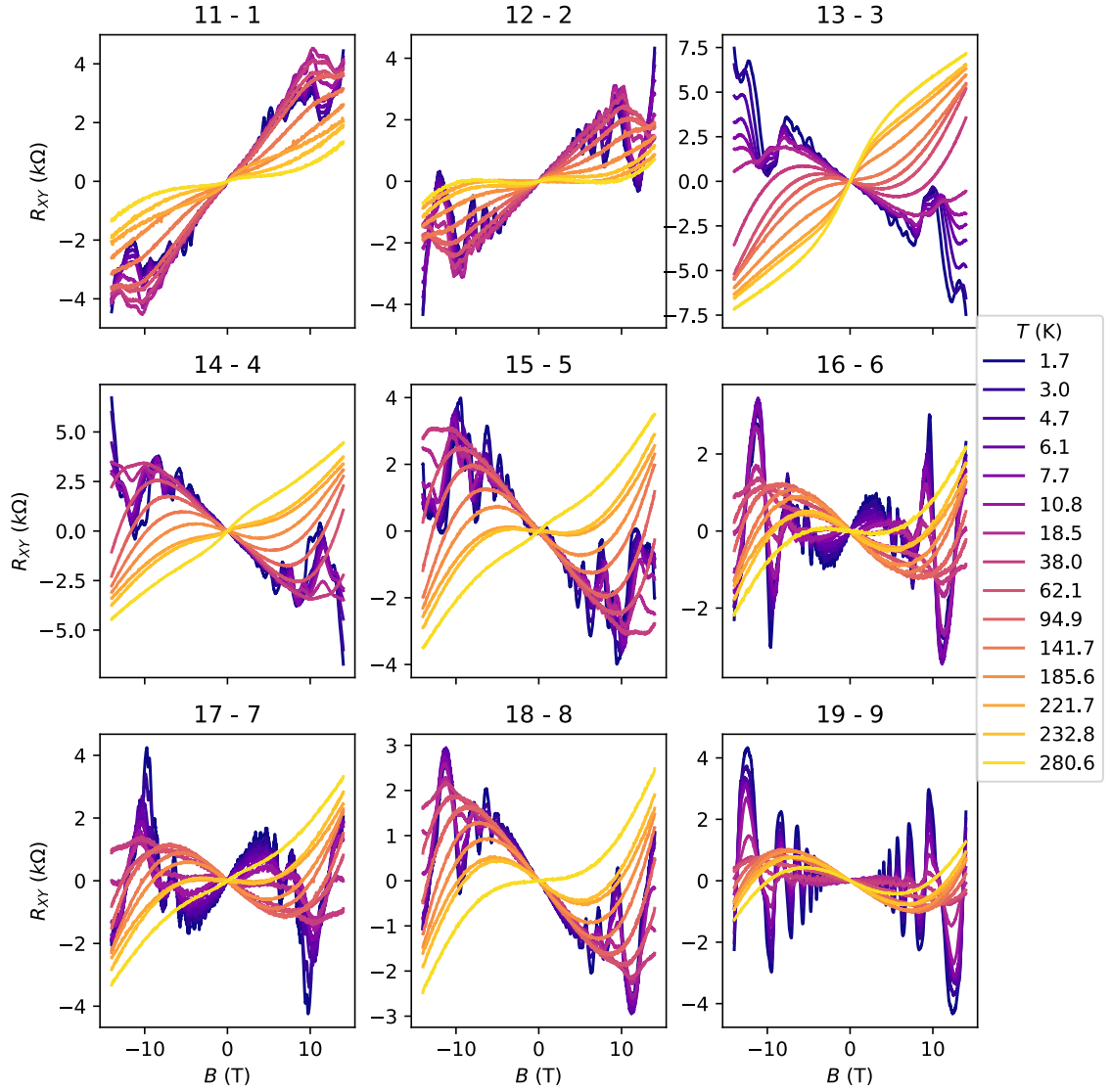


Figure A.12: Transverse resistance vs field and density at various temperatures, antisymmetrized.

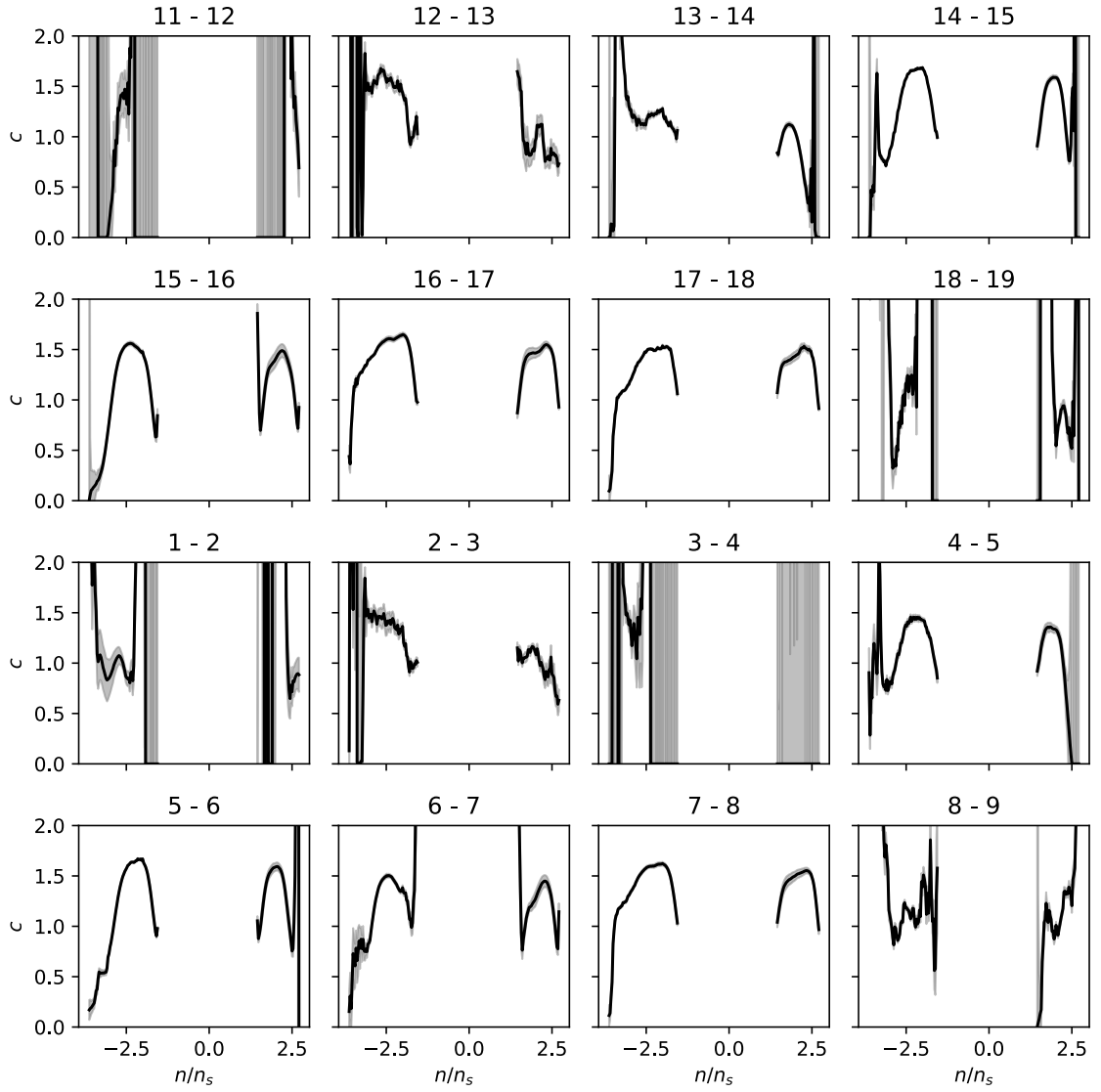


Figure A.13: Magnetoresistance exponents for all contact pairs at 1.7 K.

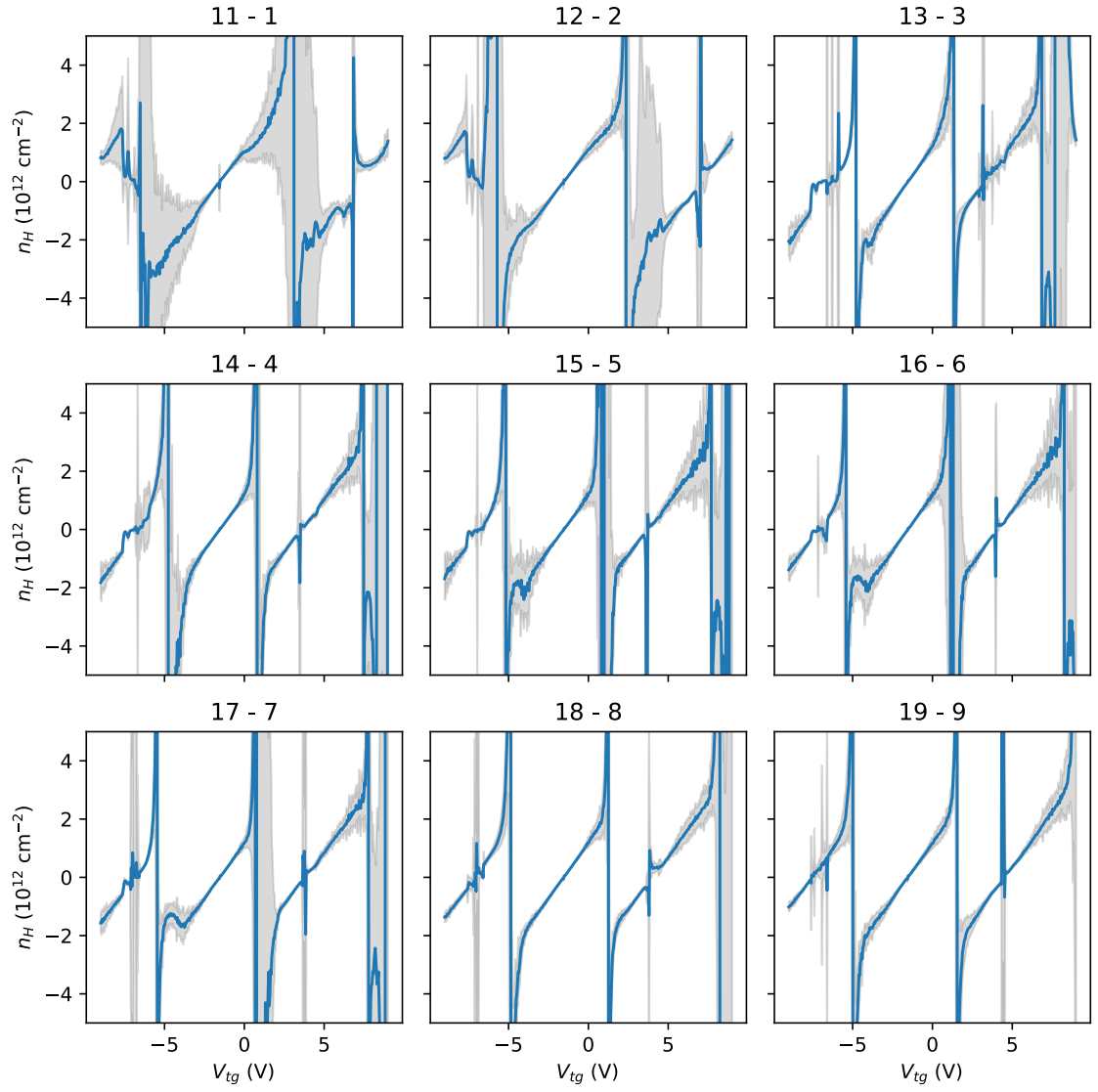


Figure A.14: Hall number for all Hall contact pairs at 1.7 K.

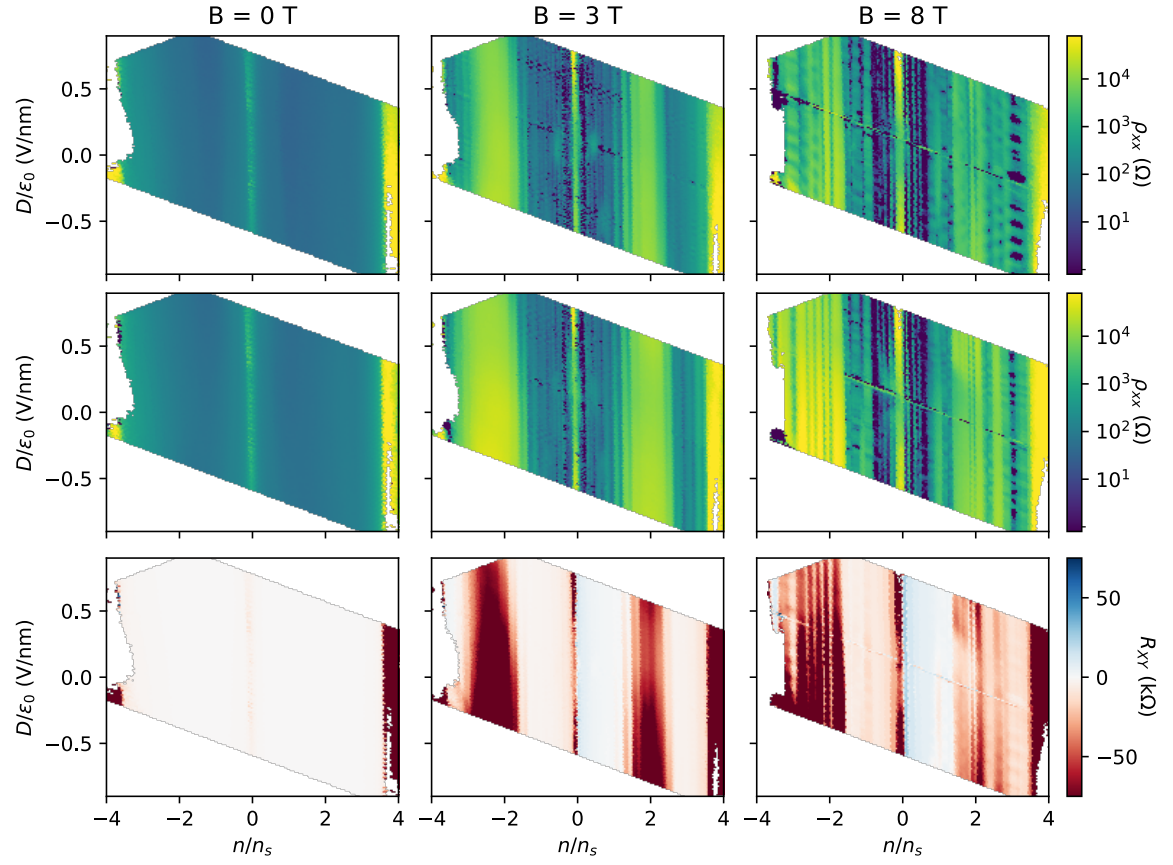


Figure A.15: **26 mK gate maps in field.** Contact pair 7-8 (top row), 4-5 (middle row), and 16-6 (bottom row).

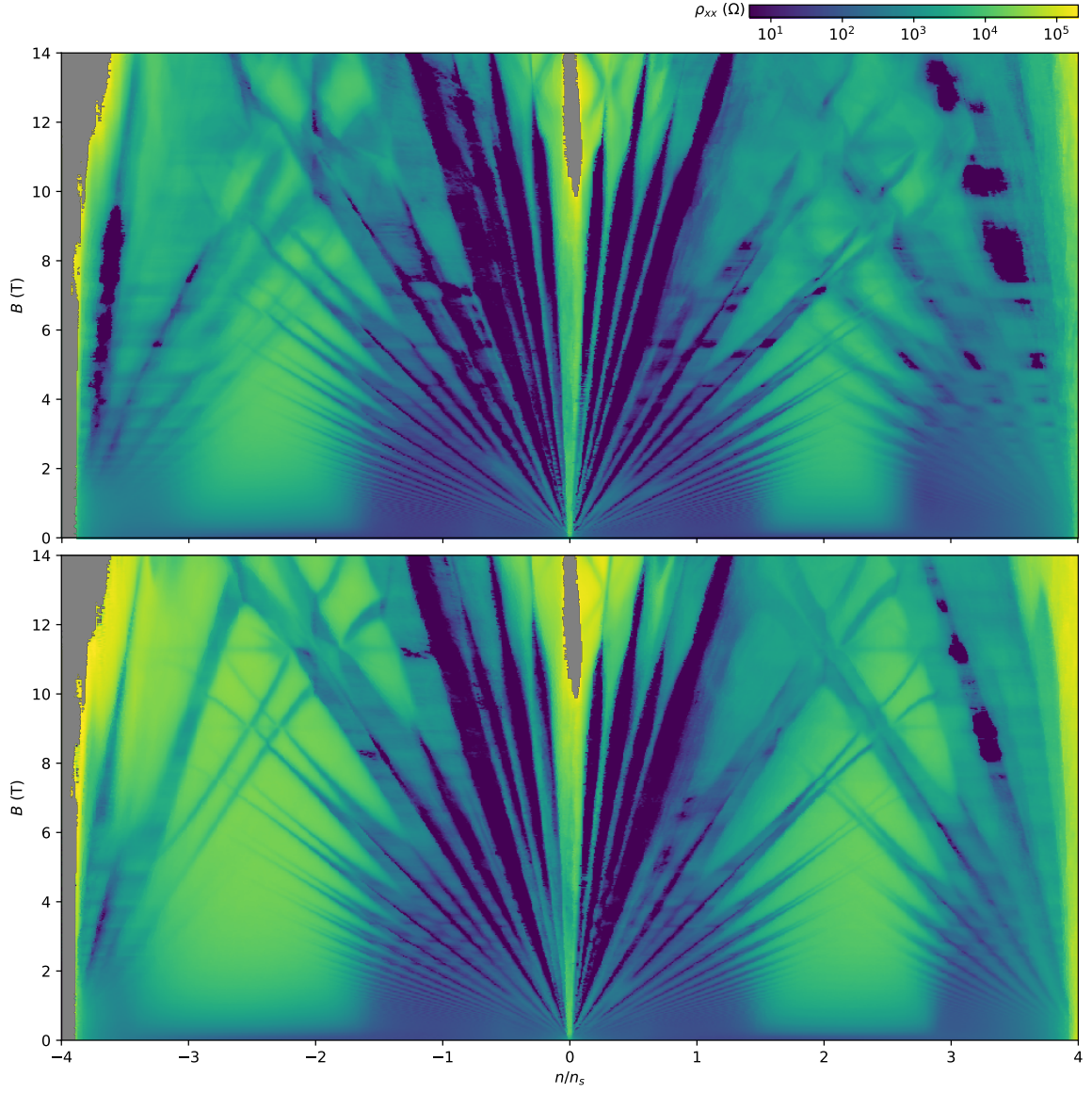


Figure A.16: Dil-fridge fan diagram for other contact pairs. Contact pair 4-5 (top), 7-8 (bottom).

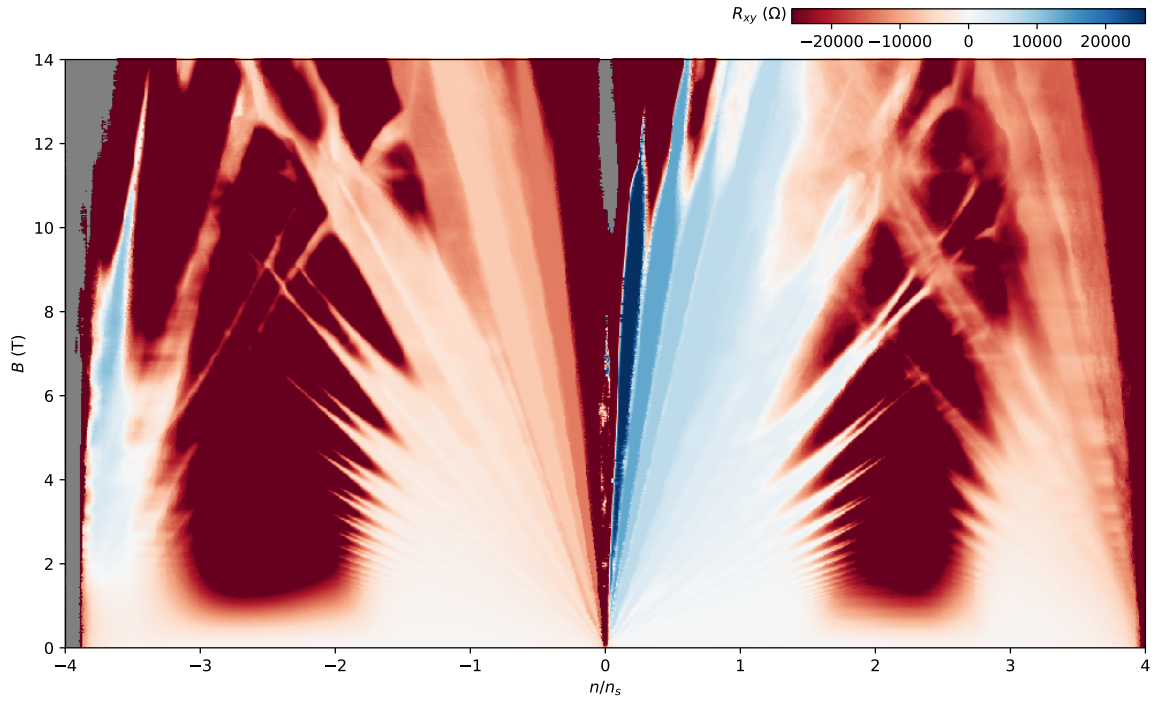


Figure A.17: **Dil-fridge fan diagram for Hall pair.** Awkwardly, I don't know which contact pair this is. Fortunately, I have a decent excuse: because of COVID lockdown, I was not allowed to physically go in to lab, and my notes were unclear. I asked the next user to photograph the wiring before disconnecting the device, however this particular contact pair is unclear in the photograph.

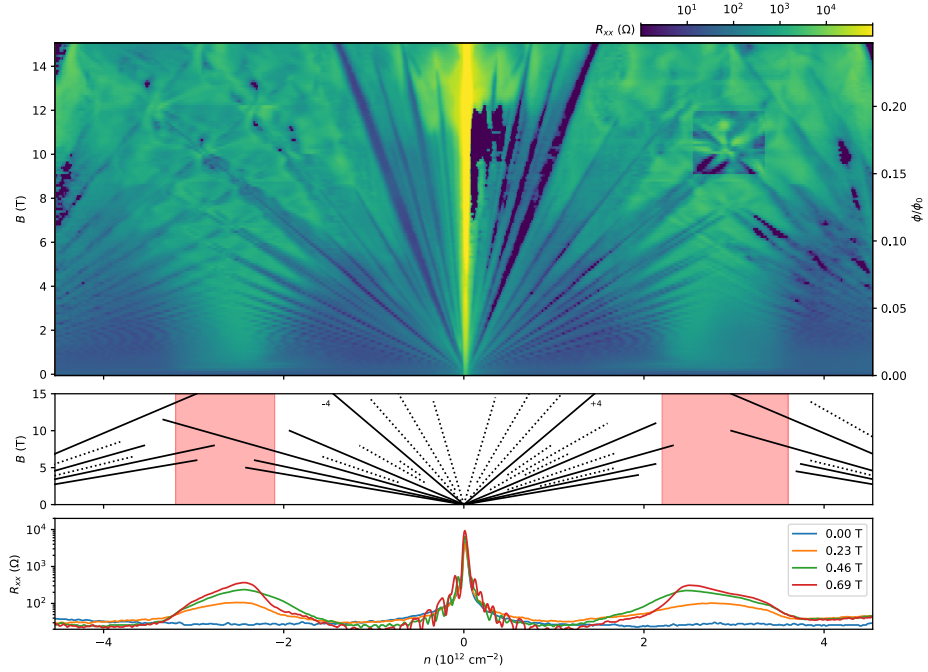


Figure A.18: **Replication of split LLs in a second device.** **Top:** Longitudinal resistance vs density and perpendicular magnetic field for device D34 ($\theta = 1.59^\circ$). The window with increased contrast (color bar does not apply) highlights the intersection of $\nu = 12$ from charge neutrality and $\nu = -12$ from full filling. There is a faint vertical line, corresponding to the average of the slopes of the two Landau levels. **Middle:** Schematic fan diagram. **Bottom:** Line cuts at the indicated fields

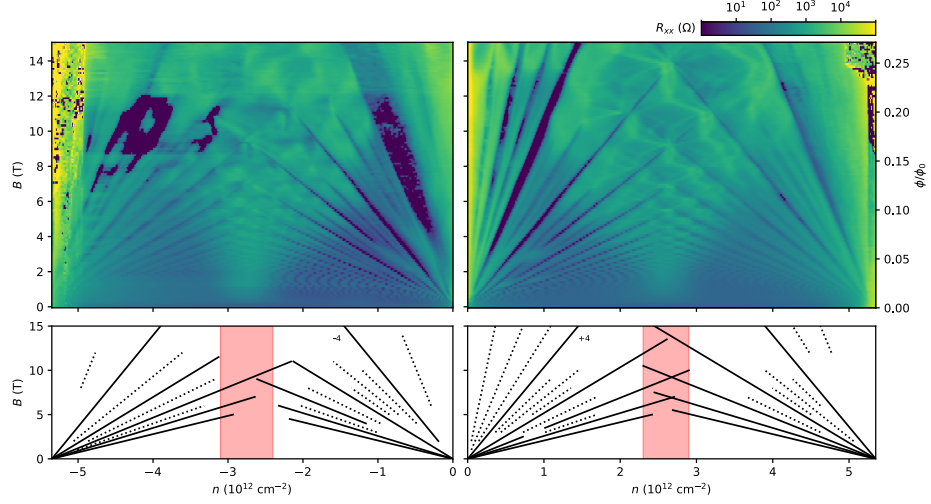


Figure A.19: **Lack of split LLs in a third device similarly far from magic angle.** Longitudinal resistance vs density and perpendicular magnetic field for device D25 ($\theta = 1.52^\circ$) for holes (**top left**) and electrons (**top right**). The measurements for electrons and holes are taken in different contact pairs and at different Si gate voltages. **Bottom panels:** Schematic fan diagrams.

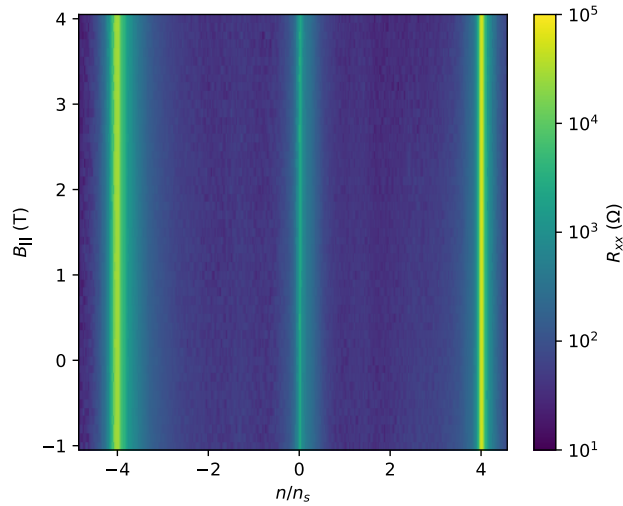


Figure A.20: **French parallel field map.** Longitudinal resistivity vs parallel field and density at 1.7 K. From the Hall slope, I calculated the sample angle to be roughly 0.7° .

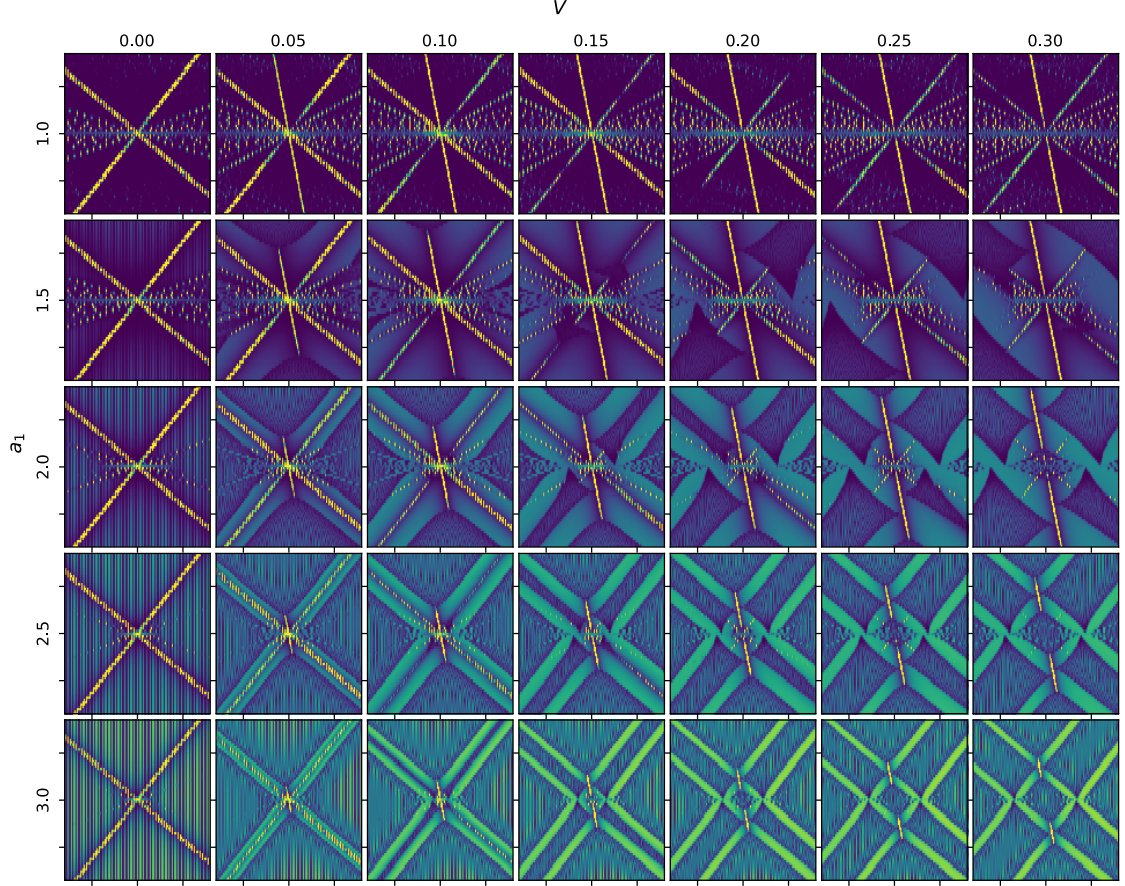


Figure A.21: **Toy model behavior as a function of a_1 and V .** Each plot shows the same region of (n, B) as in Fig. 4.9 (bottom left) of the main text, taken at $q = 1999$ and $a_2 = a_3 = 1$. The combination of experimentally observed qualitative phenomenology is reproduced by the model not just at a fine-tuned value of the parameters a_1 and V but rather for ranges of parameters a_1 and V such that $2.5 < a_y < 4$ and $0.1 < V < 0.4$. The ranges are comparable for different (n, B) . For parameter values that show clear split LLs, increasing V increases the distance between the split LLs (which goes to zero when $V = 0$), and increasing a_1 decreases their width. Increasing a_1 beyond the stated range suppresses the discontinuities at LL crossings, as a direct consequence of the decreasing width of the LLs: compare $(a_y, V) = (2.5, 0.3)$ to $(3, 0.3)$.

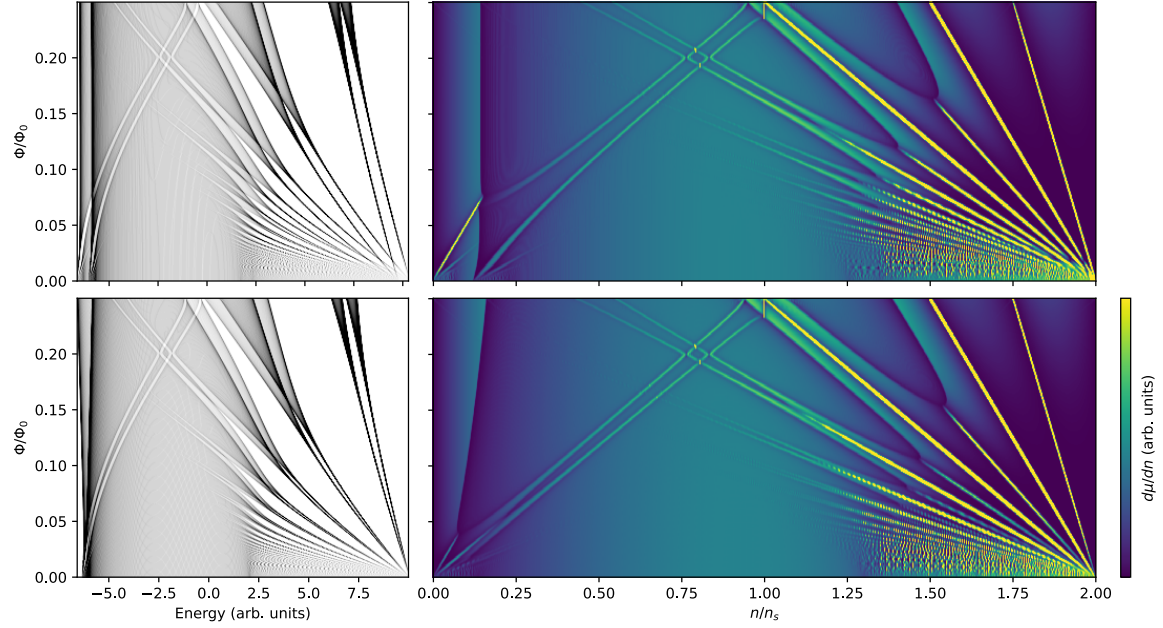


Figure A.22: **Comparison of Zeeman-like splitting to fixed splitting.** Energy spectra ($q = 1499$) and inverse density of states for $a_1 = 2$, and (top row) $V = 0.2$, (bottom row) $V = \Phi/\Phi_0$.

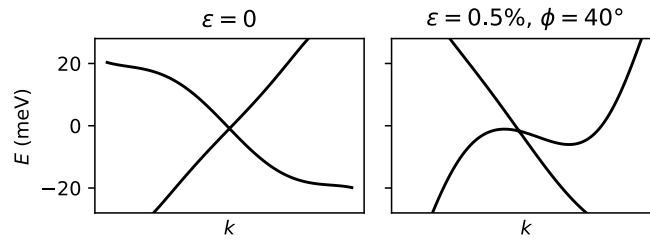


Figure A.23: **Types of Dirac crossings.** **Left:** Type-I Dirac crossing at a K point with no uniaxial heterostrain. **Right:** Type-II Dirac crossing near, but not exactly at a K point for $\epsilon = 0.5\%$ and $\phi = 40^\circ$.

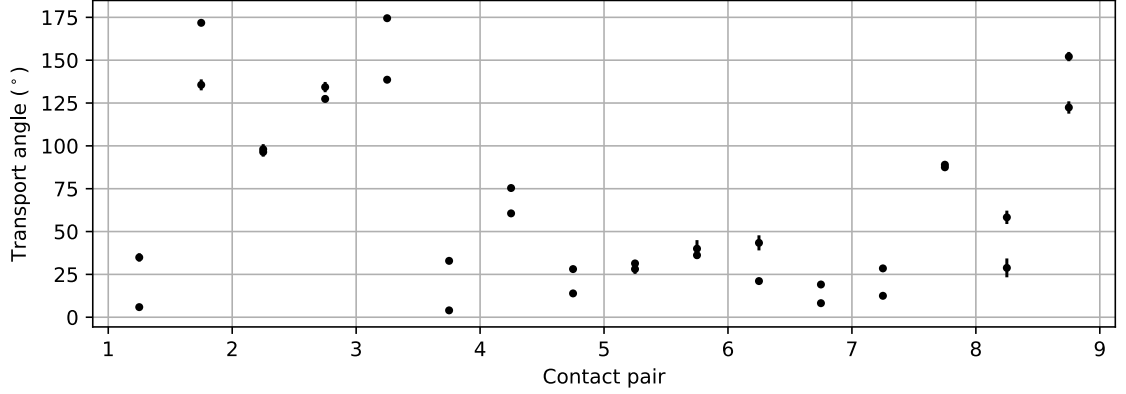


Figure A.24: **Extraction of principle axis in MR regions.** Within the open-FS regions (large MR) and at fields such that the quadratic MR dominates but has not yet saturated, the principle axis $\theta = \arctan(-\rho_{xx}/\rho_{xy})$. Here I plot every contact pair's extracted θ along the length of the device for every pair of neighboring XX and XY. Only contacts between 4 and 8 have significant MR.

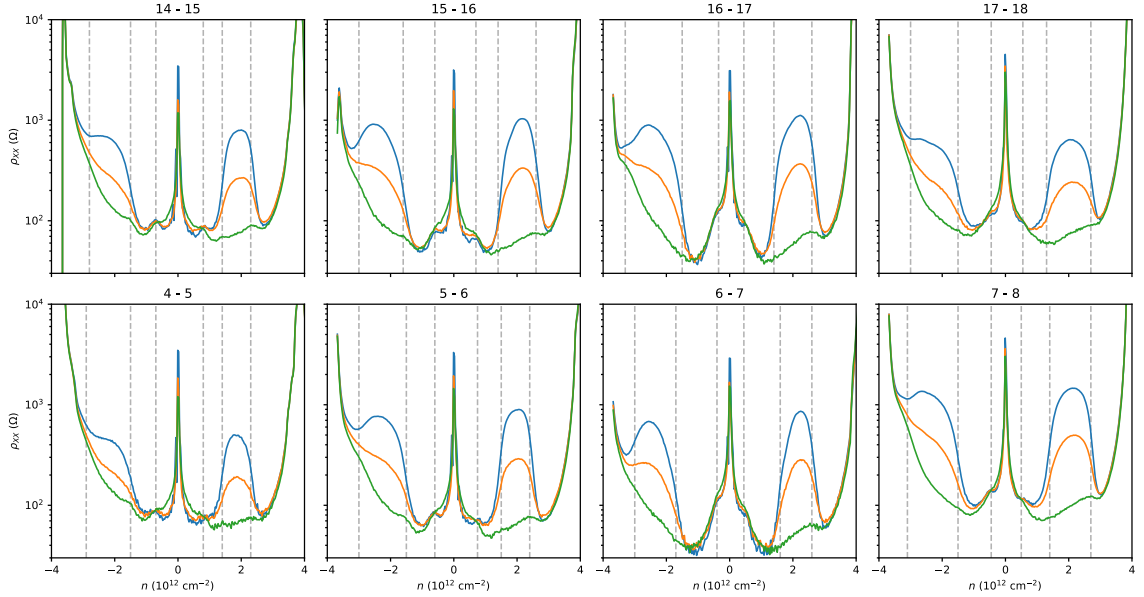


Figure A.25: All longitudinal contact pairs with quadratic MR at $B = 0$ (green), 0.25 T (orange), and 0.5 T (blue), symmetrized. Data taken at 1.6 K. Every contact pair with quadratic magnetoresistance has a well-developed shoulder or cusp near $n/n_s \approx \pm 0.5$ that we associate with the lowest-energy van Hove point. The additional vertical lines are by-eye guesses for the location of the other van Hove points.

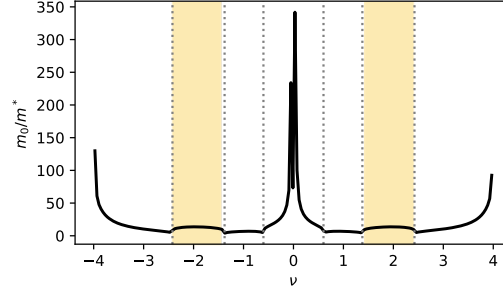


Figure A.26: Filling dependence of the averaged inverse cyclotron mass $1/m^*$, extracted from the averaged cyclotron frequency $\bar{\omega}_c = \frac{eB}{m^*}$. Here $\bar{\omega}_c = \frac{1}{N} \sum_{n,i} \omega_{c,n,i}$, where n and i label the FS (i) coming from a given band (n), in units of the bare electron mass. $1/m^*$ is larger near the charge neutrality and band edges, explaining the earlier onset of quantum oscillations in these filling regions.

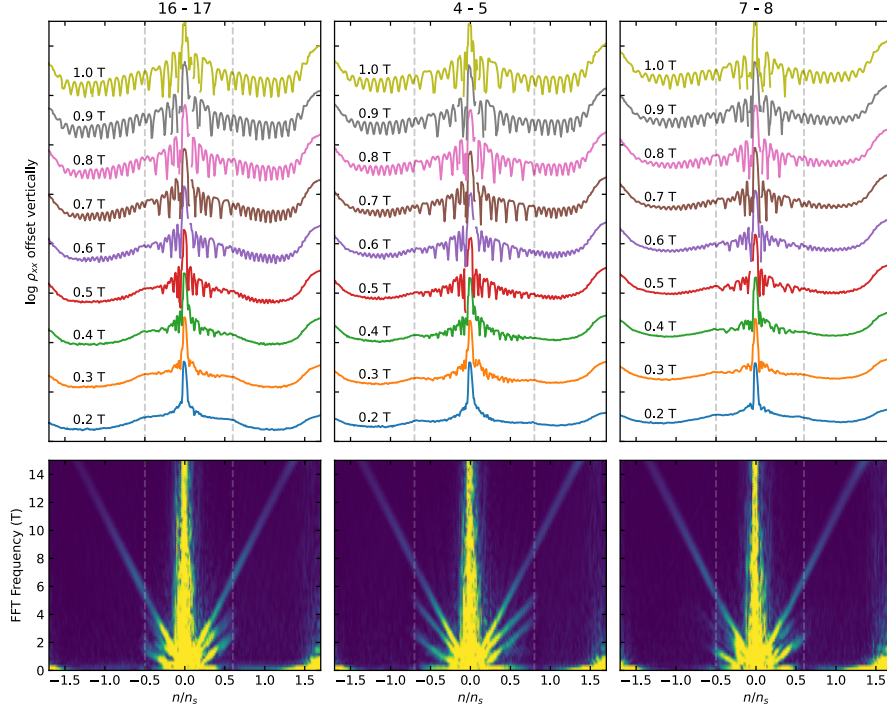


Figure A.27: Quantum oscillations near CNP for every longitudinal contact pair for which we have dilution refrigerator data ($T = 26$ mK). Vertical dashed lines are our estimate of the low-energy van Hove point based on cusps in resistivity at 0.2 T. Contact pair 4 - 5 is the pair shown in the main text.

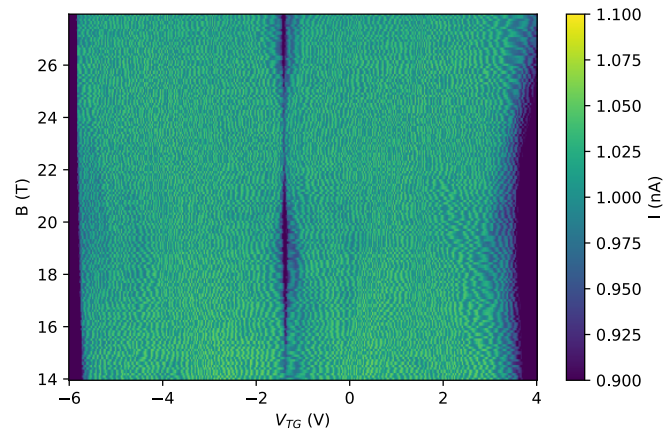


Figure A.28: Current noise in MagLab measurements.

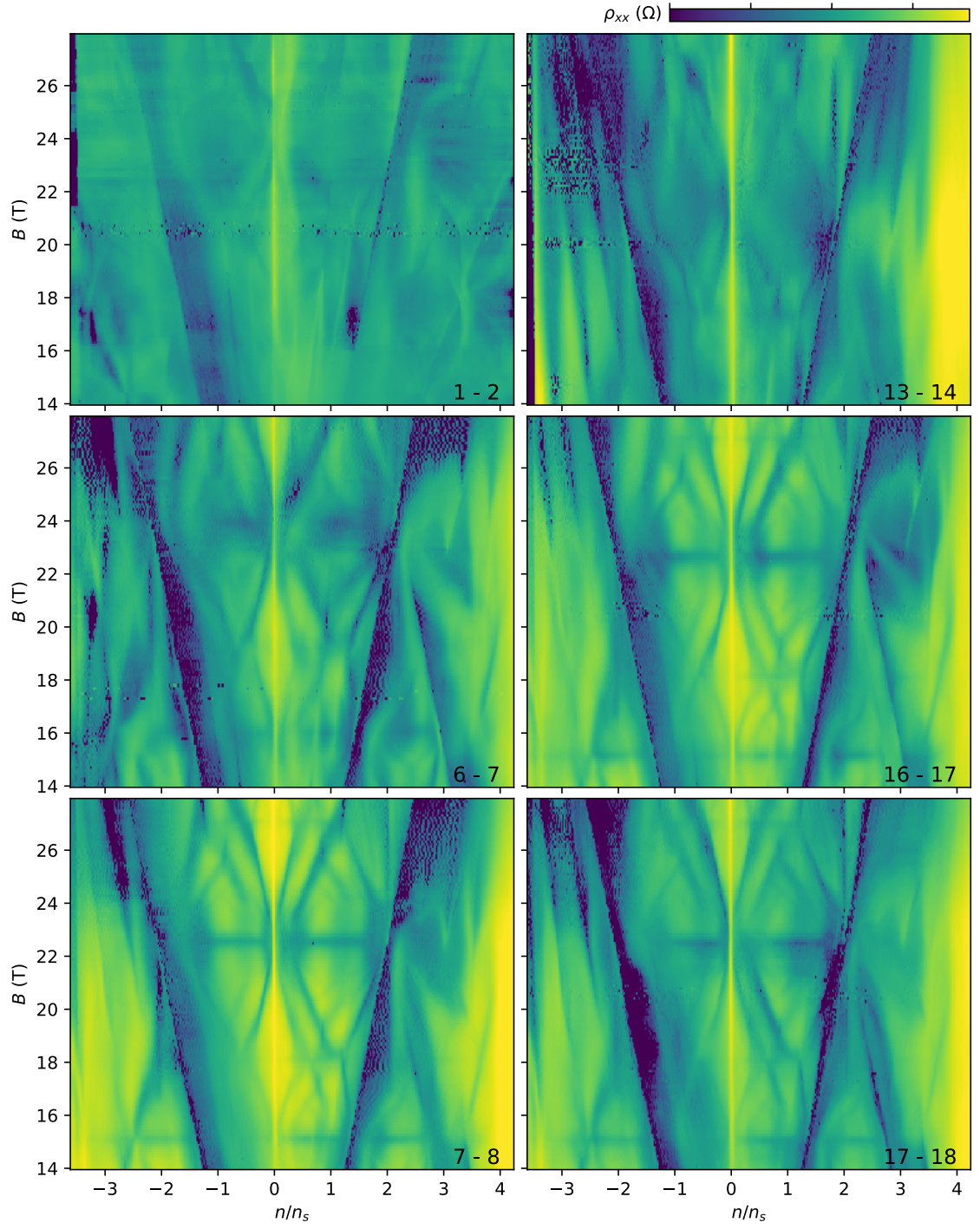


Figure A.29: All longitudinal resistivities at MagLab.

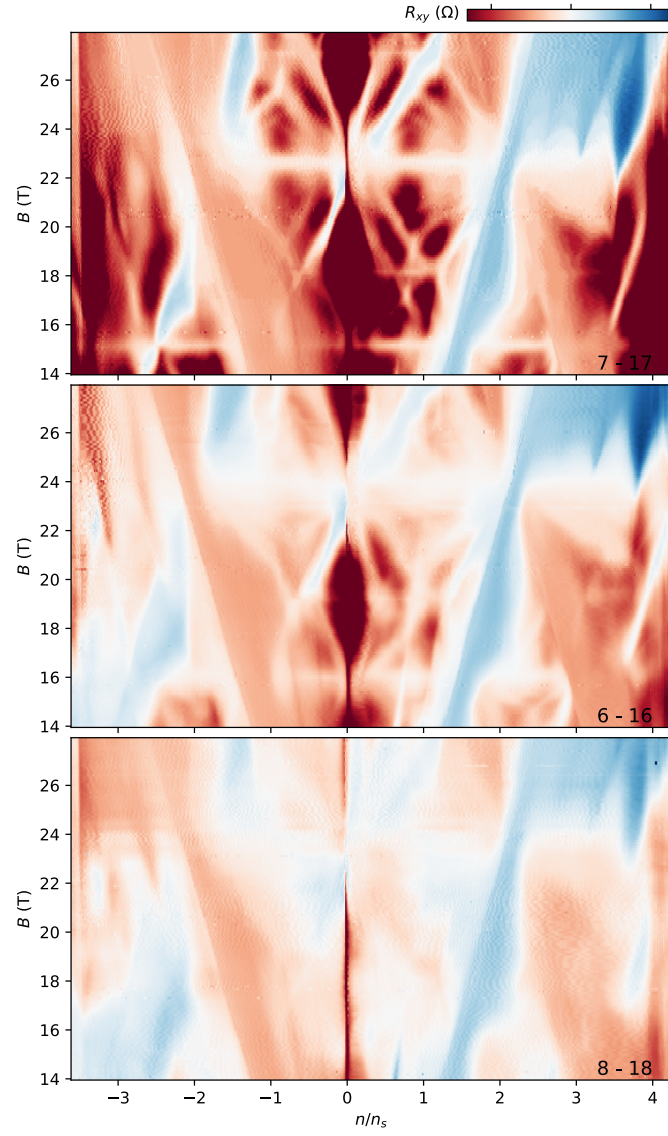


Figure A.30: All Hall resistances at MagLab.

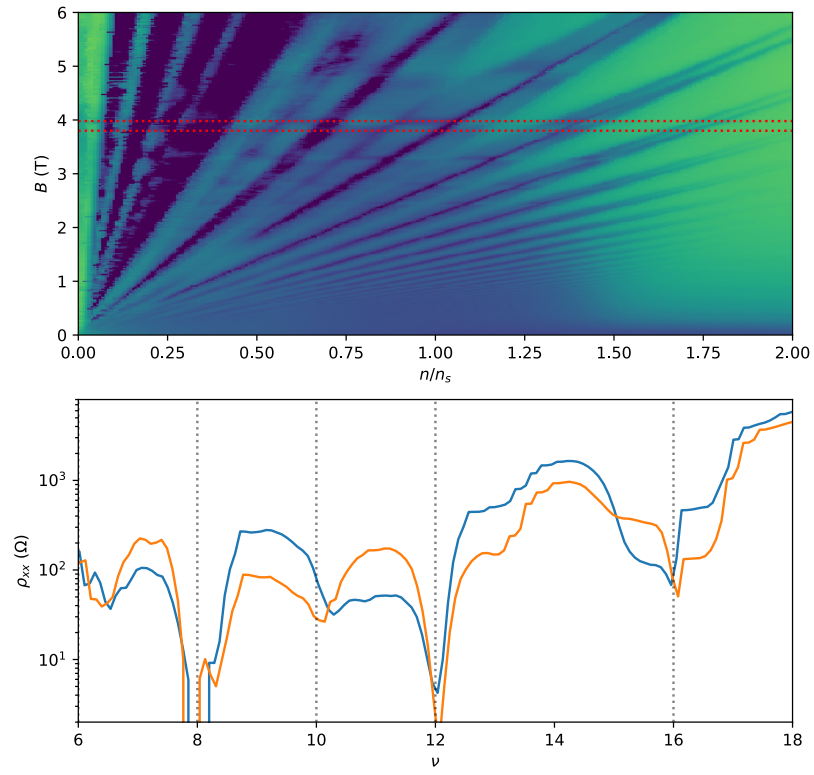


Figure A.31: **More spin-dependent transport?** Line cuts at indicated fields for contact pair 7 - 8.

Appendix B

Other lab tasks

Cryostat

I inherited ownership of our variable temperature insert (VTI). For many years, we used a repurposed Oxford dewar with a liquid nitrogen shield and an ICEoxford VTI and probe. This system worked reasonably well, however it had a critical flaw: it consumed liquid helium at a rate of roughly 10 L / day depending on magnet usage. Increasing helium prices over time led to this consumption costing us in the realm of \$100,000 per year simply to keep cold.

To save money, we ordered a recondensing liquid helium dewar and VTI from Cryo Industries of America. We chose to reuse our preexisting Oxford 14 T superconducting magnet of NbTi/Nb₃Sn rather than purchase a new magnet. In principle, the magnet can be run at 16 T if the helium is pumped to bring its temperature to the lambda point, however we do not presently support this. The choice to reuse our own magnet was primarily motivated by saving both money and time, however it ended up causing issues. First, it was unexpectedly difficult to remove the magnet from the dewar that it previously inhabited. More importantly, it was a different form factor from what Cryo Industries was accustomed to, and they mistakenly made a dewar for it that was much too small. These issues meant that it took significantly longer and more effort to order the recondensing dewar than we originally expected.

This VTI is essentially a continuous-flow cryostat that resides within a larger static dewar that is itself cooled by a Gifford-McMahon fridge (SHI cryogenics RDK-415D2

cold head and F70L compressor). In normal operation, the dewar is “subcooled,” that is, the dewar pressure is below an atmosphere and the helium temperature is below its normal 4.2 K boiling point. This means that all of the fittings on the dewar need to be leak-tight, otherwise air will be drawn into and freeze inside the dewar over time and compromise its performance.

The VTI itself is constructed as a double-walled tube; the inner tube is the sample chamber, and it is surrounded by a vacuum jacket that insulates the inner tube from the outer tube. The lower end of this tube goes inside the bore of the magnet, which is submerged in the main liquid helium bath, and it is closed except for a narrow capillary tube that connects the inner sample chamber to the main bath. This capillary is thus the connection by which liquid helium enters the sample chamber. The flow rate of helium is controlled by a needle valve on this capillary. The temperature of the sample chamber is controlled by heating the helium that enters it. This helium vapor flows upward, exchanging heat with the sample, and then is pumped out at the top of the chamber. Keeping the chamber pressure low with a pump allows the helium and sample temperature to be as low as 1.3 K.

The vacuum jacket blocks conduction of heat to allow the sample chamber to be either colder or warmer than the liquid helium of the cryostat main bath. The space constraints inside the 52 mm diameter superconducting magnet bore requires this jacket to be very thin, so it cannot act as an effective barrier against thermal conduction unless it achieves and retains very low pressure. It must therefore be completely leak-tight; if a leak slightly elevates the vacuum jacket pressure, the thermal coupling between the sample chamber and main bath will increase. When the system was first shipped to us, some of the bolts to an indium seal on the vacuum jacket had come loose, allowing helium into the jacket and preventing us from condensing helium. When the vacuum jacket is working well, it is possible to keep the sample chamber temperature as high as 320 K and simultaneously ramp the magnet to 14 T.

Up-to-date instructions for the usage of the VTI are available on Google Drive.

Measurement code

Measurement code needs to accomplish several things: 1. set outputs to desired values, 2. read inputs, 3. plot the two against each other, and 4. save the results to disk. I was overall unsatisfied with existing projects that accomplish these tasks, so I built my own. I based it on Python, matplotlib, and Jupyter notebooks, because it is the standard for such things in the lab. I used QCoDeS instrument drivers [155], which have several convenient features. Aside from having a wide range of already-written instrument drivers, they also allow for setting software limits and ramp rates. However, I was not happy with the QCoDeS data save format. I found it overly complex, the saved data were large in filesize, difficult to inspect, and did not play nicely with DropBox, they had no facilities for live plotting, and other reasons.

So, I wrote my own.¹ What I wrote is more restrictive than what is included with QCoDeS, however it suffices for basically all of the measurements that we do in our lab. It allows for 0D (measure), 1D (sweep), and 2D (megasweep) scans, with adjustable delays in between setting and reading the measurements. It allows for live plotting, and it saves results to CSV files as the data are taken, so that a power outage will not lead to lost data.

I was never entirely satisfied with this project. There were always more features that I would have liked to add, such as an editable queue of measurements and better 2D live plotting. Plus, it is a real pain to install on new computers, because Python is fundamentally bad at packaging, especially on Windows. Some day I would prefer to redo this project in a compiled language, so I can distribute a single binary.

¹<https://github.com/spxtr/measurement>

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