

# Emergent Phenomena in Multilayer Graphene with Planar and Vertical Architectures

By

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**Angle-resolved transport non-reciprocity and spontaneous symmetry breaking in twisted trilayer graphene**

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# Contents

|          |                                                                             |           |
|----------|-----------------------------------------------------------------------------|-----------|
| <b>1</b> | <b>Introduction</b>                                                         | <b>1</b>  |
| 1.1      | General concept drawing . . . . .                                           | 1         |
| 1.2      | Two-dimensional materials and van der Waals heterostructures . . . . .      | 3         |
| 1.3      | Moiré in graphene multilayer . . . . .                                      | 5         |
| 1.4      | Quantum Hall bilayer . . . . .                                              | 6         |
| 1.5      | Outline . . . . .                                                           | 8         |
| <b>2</b> | <b>Angle resolved transport in twisted trilayer graphene</b>                | <b>9</b>  |
| 2.1      | Superconductivity in twisted multilayer graphene . . . . .                  | 9         |
| 2.2      | Sunflower geometry and angle resolved transport . . . . .                   | 13        |
| 2.3      | Annex: Full power of the sunflower geometry . . . . .                       | 15        |
| 2.4      | Anisotropy in the metallic states in MATTG . . . . .                        | 18        |
| 2.5      | Anisotropy in superconducting MATTG . . . . .                               | 23        |
| <b>3</b> | <b>Non-reciprocal angle resolved transport in twisted trilayer graphene</b> | <b>30</b> |
| 3.1      | Non-reciprocity and nonlinearity . . . . .                                  | 30        |
| 3.2      | Non-reciprocal transport with angle dependence . . . . .                    | 32        |
| 3.3      | Angle-resolved non-reciprocity in TTG . . . . .                             | 35        |
| 3.4      | Angle-resolved non-reciprocity in TTG phase diagram . . . . .               | 38        |
| 3.5      | Angle dependence of superconducting diode effect . . . . .                  | 45        |



|          |                                                                                |            |
|----------|--------------------------------------------------------------------------------|------------|
| <b>4</b> | <b>Nonlinear transport in Bernal bilayer graphene with sunflower geometry</b>  | <b>49</b>  |
| 4.1      | Bernal bilayer graphene and nonlinear Hall effect . . . . .                    | 49         |
| 4.2      | Nonlinear transport in theory . . . . .                                        | 53         |
| 4.3      | Extracting nonlinear conductivity tensor in BBG . . . . .                      | 56         |
| 4.4      | Nonlinear Hall effect in BBG $n$ - $D$ phase diagram . . . . .                 | 62         |
| 4.5      | Side aisle: the quantitative reconstruction of Fermi surface contour . . . . . | 65         |
| <b>5</b> | <b>Excitons in two-component fractional quantum Hall effect</b>                | <b>67</b>  |
| 5.1      | Excitonic transport in quantum Hall bilayer . . . . .                          | 67         |
| 5.2      | Two-component fractional quantum Hall effect . . . . .                         | 73         |
| 5.3      | Excitons in $\frac{2}{2}$ CF states . . . . .                                  | 79         |
| 5.4      | Excitons in $\frac{2}{1}$ CF states . . . . .                                  | 81         |
| <b>6</b> | <b>Excitons in decoupled fractional quantum Hall effects</b>                   | <b>86</b>  |
| 6.1      | Layer decoupled FQHE . . . . .                                                 | 86         |
| 6.2      | Excitons in $\frac{2}{0}$ CF states with thermal excitation . . . . .          | 88         |
| 6.3      | Excitons in $\frac{2}{0}$ CF states with electrostatic doping . . . . .        | 92         |
| 6.4      | Excitons at even denominator states . . . . .                                  | 94         |
| <b>7</b> | <b>Epilogue</b>                                                                | <b>96</b>  |
| 7.1      | Topping out . . . . .                                                          | 96         |
| 7.2      | Scene from the roof . . . . .                                                  | 98         |
|          | <b>Bibliography</b>                                                            | <b>101</b> |
|          | <b>Appendices</b>                                                              | <b>117</b> |
| <b>A</b> | <b>Experiment scheme for angle resolved transport measurement</b>              | <b>118</b> |
| <b>B</b> | <b>Python code for full extraction of linear conductivity matrix</b>           | <b>122</b> |
| <b>C</b> | <b>Conductivity-resistance conversion in sunflower geometry</b>                | <b>128</b> |



# List of Figures

|      |                                                                              |    |
|------|------------------------------------------------------------------------------|----|
| 1.1  | Family of 2D materials. . . . .                                              | 3  |
| 1.2  | Typical stacking structure for electrical transport studies. . . . .         | 4  |
| 1.3  | Moiré pattern induced from twisting graphene layers. . . . .                 | 6  |
| 1.4  | Calculated band structures of twisted multilayer graphene. . . . .           | 6  |
| 1.5  | Schematic of quantum Hall bilayer structure. . . . .                         | 7  |
| 2.1  | Brillouin zone of graphene and twisted graphene. . . . .                     | 10 |
| 2.2  | Transport features of MATTG. . . . .                                         | 11 |
| 2.3  | Anisotropic SC in MATBG. . . . .                                             | 12 |
| 2.4  | Angle resolved transport and sunflower geometry . . . . .                    | 14 |
| 2.5  | The “sunflower” model beyond angular fit. . . . .                            | 16 |
| 2.6  | Transport anisotropy in the metallic state of MATTG. . . . .                 | 18 |
| 2.7  | Density and temperature dependence of $\Delta R/R_0$ . . . . .               | 19 |
| 2.8  | Twist angled dependence of transport anisotropy. . . . .                     | 20 |
| 2.9  | Temperature induced hysteresis in the metallic state. . . . .                | 21 |
| 2.10 | Anisotropy of strange metallicity. . . . .                                   | 22 |
| 2.11 | Anisotropy of $I_c$ . . . . .                                                | 24 |
| 2.12 | Anisotropy of $T_{onset}$ . . . . .                                          | 25 |
| 2.13 | Anisotropy of SC and metallic phase. . . . .                                 | 26 |
| 2.14 | Evolution of anisotropy between metallic and superconducting phases. . . . . | 27 |
| 2.15 | Temperature induced hysteresis in the superconducting phase. . . . .         | 29 |

|      |                                                                                                    |    |
|------|----------------------------------------------------------------------------------------------------|----|
| 3.1  | Second harmonic response as a probe for nonreciprocity. . . . .                                    | 32 |
| 3.2  | Symmetry analysis of the angle resolved transport response. . . . .                                | 33 |
| 3.3  | Angle dependence of second harmonic response in TTG. . . . .                                       | 36 |
| 3.4  | Angle dependence of transport non-reciprocity and second harmonic re-<br>sponse. . . . .           | 38 |
| 3.5  | Low temperature phase diagram of the TTG sample. . . . .                                           | 39 |
| 3.6  | Magnetic field induced hysteresis loops. . . . .                                                   | 40 |
| 3.7  | Ubiquity of non-reciprocity across the moiré band. . . . .                                         | 41 |
| 3.8  | Temperature and current induced hysteresis in angle resolved transport<br>non-reciprocity. . . . . | 42 |
| 3.9  | Density induced rotation of mirror axis. . . . .                                                   | 43 |
| 3.10 | Temperature dependence of orbital ferromagnetism and non-reciprocity. . . . .                      | 43 |
| 3.11 | Schematics of valley polarization and momentum polarization. . . . .                               | 44 |
| 3.12 | Zero-field superconducting diode effect. . . . .                                                   | 45 |
| 3.13 | Angle dependence of superconducting diode effect in TTG. . . . .                                   | 46 |
| 3.14 | Angle dependence of superconducting diode effect in MATTG. . . . .                                 | 47 |
| 4.1  | $\beta$ as a function of $n$ and $D$ in BBG. . . . .                                               | 50 |
| 4.2  | Transitions as a function of $B$ in BBG. . . . .                                                   | 51 |
| 4.3  | Nonlinear transverse signal vs. nonlinear Hall effect. . . . .                                     | 52 |
| 4.4  | Basis functions of nonlinear electrical potential. . . . .                                         | 55 |
| 4.5  | Quadratic regime of BBG. . . . .                                                                   | 57 |
| 4.6  | Extracting nonlinear conductivity tensor in BBG. . . . .                                           | 58 |
| 4.7  | Nonlinear signal decomposition into three bases. . . . .                                           | 59 |
| 4.8  | Nonlinear response decomposition by dissipation. . . . .                                           | 61 |
| 4.9  | Nonlinear conductivity in BBG $n - D$ phase diagram. . . . .                                       | 64 |
| 4.10 | Quantitative reconstruction of Fermi surface contour. . . . .                                      | 66 |
| 5.1  | Schematic for interlayer excitons at $\nu_{\text{total}} = 1$ . . . . .                            | 68 |

|      |                                                                                |     |
|------|--------------------------------------------------------------------------------|-----|
| 5.2  | BEC superfluid signatures at $\nu_{\text{total}} = 1$ .                        | 69  |
| 5.3  | Double Corbino graphene/hBN quantum Hall bilayer.                              | 70  |
| 5.4  | Measurement configurations in double Corbino geometry.                         | 71  |
| 5.5  | Composite fermion and Jain sequences.                                          | 74  |
| 5.6  | Two-component FQHE phase diagram.                                              | 75  |
| 5.7  | PF conductance in $\nu_{\text{total}}-\Delta\nu$ phase diagram at $B = 12$ T   | 77  |
| 5.8  | ${}^2_2\text{CF}$ FQHE sequence with interlayer excitons.                      | 78  |
| 5.9  | ${}^2_2\text{CF}$ excitons in $\nu_{\text{total}}-\nu_1$ plane.                | 79  |
| 5.10 | Depairing of ${}^2_2\text{CF}$ excitons via temperature and bias               | 80  |
| 5.11 | ${}^2_1\text{CF}$ FQHE sequence with interlayer excitons.                      | 82  |
| 5.12 | Charge gaps at ${}^2_0\text{CF}$ and ${}^2_1\text{CF}$ sequences               | 83  |
| 5.13 | Depairing of ${}^2_1\text{CF}$ excitons via temperature.                       | 83  |
| 5.14 | Excitonic pairing between quasiparticle/quasihole at ${}^2_1\text{CF}$ states. | 84  |
| 6.1  | Transport signatures along $\nu_1 = 1/3$ .                                     | 87  |
| 6.2  | Phase diagram at $\nu_{\text{total}} = 1$ .                                    | 89  |
| 6.3  | $B$ -dependence of the ${}^2_0\text{CF}$ states.                               | 90  |
| 6.4  | $T$ -dependence of the $\nu_1 = \nu_2 = 1/3$ state.                            | 91  |
| 6.5  | Electrostatic doping induced excitons.                                         | 92  |
| 6.6  | Layer asymmetric vs layer symmetric doping.                                    | 93  |
| 6.7  | $G_{\text{PF}}$ around an even denominator state.                              | 94  |
| 6.8  | Excitons at an even denominator state.                                         | 95  |
| A.1  | Angle dependence measurement configurations.                                   | 119 |
| A.2  | More angle dependence measurement configurations.                              | 120 |
| C.1  | Simulated angle dependence.                                                    | 129 |
| C.2  | Anisotropy dependence of $\Delta R/R_0$ and $G$ .                              | 130 |
| C.3  | Simulated angle dependence with different configurations.                      | 132 |

|     |                                                          |     |
|-----|----------------------------------------------------------|-----|
| C.4 | Anisotropy dependence of $G^{III}$ and $g$ .             | 133 |
| C.5 | Anisotropy dependence of $\xi$ and $R_{\max}/R_{\min}$ . | 134 |

# CHAPTER 1

## Introduction

### 1.1 General concept drawing

Ever since the enlightening debut of P. W. Anderson’s “More is different” in 1972, condensed matter physics has been uniquely distinguished for its approach to understanding complex systems [1]. Embracing a more holistic, top-down view of nature’s complexity, condensed matter physics is deeply grounded in the idea that emergent phenomena arising at macroscopic scales cannot be fully understood by merely reducing the system to its microscopic components. This antireductionist perspective has shaped condensed matter physics into a playground for many-body systems and has led condensed matter physicists to search for novel, often surprising, collective behaviors [2–10].

Central to this concept of antireductionism is the role of correlation. Correlation arises from the interactions between charges, spins, and other degrees of freedom. As the interaction gets stronger, the particles correlate more strongly, yielding a system that is less reducible and thus more intricate. Superconductivity (SC) and quantum Hall effect (QHE) are great examples of how strengthening correlations can completely change the complexity of the systems [11–14]: While the BCS model and Landau quantization nicely explains the conventional SC and integer QHE with weak electron-electron interactions, full

descriptions of unconventional, high critical temperature ( $T_c$ ) SC or QHE with fractional fillings that consider strong correlations are still largely elusive, just like many other strongly correlated states [15–20].

From an experimentalist’s perspective, characterizing strongly correlated states is often a challenge. Sometimes they manifest only through specific symmetry-breaking orders, while at other times their very existence results from a delicate balance between competing interactions. In both cases, the ability to create tuning knobs—whether controlling symmetries or interactions—is quintessential to the experimental study of correlations. A compelling example is ferromagnetism, which arises from correlated spin alignment breaking the time-reversal symmetry. In many systems, the tendency toward ferromagnetic order exists as a latent instability, hidden by competing orders that preserves time-reversal [21–24]. A small perturbation, such as a magnetic field, can be introduced to break the time-reversal symmetry, tip the competition balance, and stabilize the ferromagnetic phase, making it experimentally visible. In this sense, the symmetry of an order serves not only as a classification of known phases but also as a viewing window and a tuning knob, helping to reveal hidden correlations and explore nearby quantum phases. The ability to observe and manipulate such parameters is therefore essential to any experimental effort to map and understand the complex landscape of strongly correlated matter.

Low-dimensional materials provide an ideal platform for studying exotic strongly correlated states. Due to the reduced dimensionality, Coulomb screening is reduced, interactions and correlation therefore become prominent, leading to new and exotic phases of matter that are often not observed in bulk materials. The confinement of electrons to a limited space also increases the sensitivity of these materials to external factors like electric fields, magnetic fields, and strain, which can be finely tuned to manipulate the system’s behavior. As a result, these systems not only offer rich playgrounds for investigating fundamental aspects of strongly correlated physics but also enable experimental control over the symmetries and interactions that drive emergent behaviors, making them an



invaluable tool for both basic research and potential technological applications.

## 1.2 Two-dimensional materials and van der Waals heterostructures

Graphene, an atomically thin layer of carbon atoms arranged in a honeycomb lattice, is certainly the best example of a 2D material [26, 27]. Its exceptional electronic, mechanical, and thermal properties sparked a wave of interest in two-dimensional (2D) materials, a class of systems that are only a few atoms thick yet can exhibit rich and often unexpected physical phenomena [28–32]. Beyond graphene, the family of 2D materials has rapidly expanded to include hexagonal boron nitride (hBN), transition metal dichalcogenides (TMDs), and various other compounds. This large family of materials exhibits various electronic properties: semi-metal, insulator, semiconductors, and so on, as summarized in

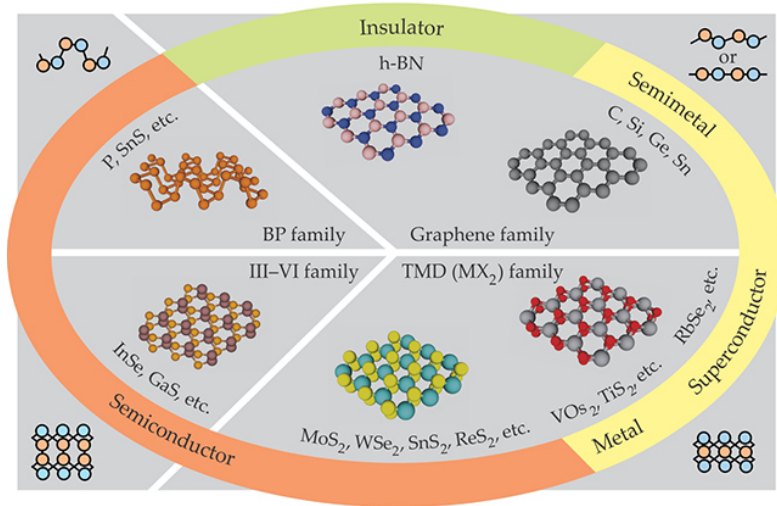


Figure 1.1: **Family of 2D materials.**

The family of 2D materials has expanded to include a large variety of electronic properties. Reproduced from Ref. [25]

Fig. 1.1.

A defining characteristic of many of these materials is their layered crystal structure, in which strong in-plane covalent bonds are held together by relatively weak van der Waals (vdW) forces between the layers. This weak interlayer coupling not only enables the mechanical exfoliation of individual atomically thin flakes but also opens the possibility of restacking them in arbitrary configurations. What was once considered a magical process of thinning becomes, in my hindsight, the foundation of engineering new materials by stacking exfoliated layers in chosen sequences and orientations [33–36]. The advancement of this stacking technique has led to the development of vdW heterostructures, in which different 2D materials are stacked on top of each other without the constraint of lattice matching. The ability to assemble vdW heterostructures has two important implications. First, it enables the fabrication of exceptionally clean devices, with atomically sharp and self-cleaned interfaces [33]. In the context of electric transport, this means the isolation of the sample of interest, free from substrate disorder or uncontrolled electrostatics. A typical

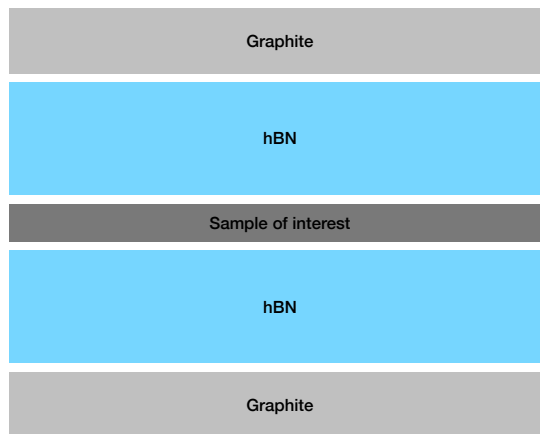


Figure 1.2: **Typical stacking structure for electrical transport studies.**

A illustration of the stacking structure of a typical vdW heterostructure for electrical transport studies. With the dual gate structure, the sample of interest is isolated from the messy, less-controlled environment.

configuration is shown in Fig. 1.2: Top and bottom graphite gates offer metallic screening from the environmental electro-magnetic noises and hBN spacers provide atomically flat and inert dielectric, reducing trapped charges or surface roughness from the Si/SiO<sub>2</sub> substrate, together creating a pristine, tunable environment ideally suited for studying the intrinsic electronic behavior of the sample. Second, it offers unprecedented artificial material design flexibility-the sample of interest itself can also be made of multiple layers of same or different 2D materials. With careful choices made when stacking these materials together, one can create new in-plane modulation, like a moiré length scale in the case of twisted bilayer graphene; or introduce out-of-plane interactions, like the interlayer excitonic pairing in the case of quantum Hall bilayer [37–55].

### 1.3 Moiré in graphene multilayer

Twisted multilayer graphene stands out as the quintessential example of how vdW stacking can generate totally new physics with a few simple ingredients. By introducing a small relative twist angle between adjacent graphene layers, one creates long-range moiré patterns that reshape the low-energy electronic landscape (see Fig. 1.3) [37, 38, 56–58]. For bilayer graphene, when twisted to the so-called the “magic angle,” the energy band structure becomes flat, where kinetic energy is quenched and electronic interactions dominate, leading to a surprising collection of phases, including superconductivity, correlated insulators, and orbital ferromagnetism [39–43, 59–69].

The idea smoothly extends to twisted multilayer systems [70]. In fact, for the case of alternately twisted trilayer graphene, its Hamiltonian can be effectively decomposed into that of a twisted bilayer graphene and a monolayer graphene, as illustrated in their band structures (Fig. 1.4): At its own magic-angle, TTG features a flat band coexisting with a Dirac cone at the  $K$  point. As a result, magic-angle twisted trilayer graphene (MATTG) also hosts a diverse set of quantum phases, just like the bilayer case [44–47, 71–77].

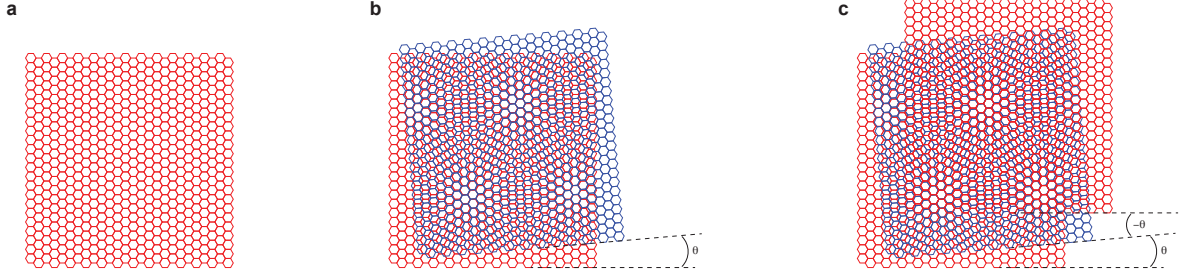


Figure 1.3: **Moiré pattern induced from twisting graphene layers.**

(a) An illustration of a single layer graphene, (b) twisted bilayer graphene, and (c) alternating twisted trilayer graphene. A moiré length scale is visibly induced by a simple twist.

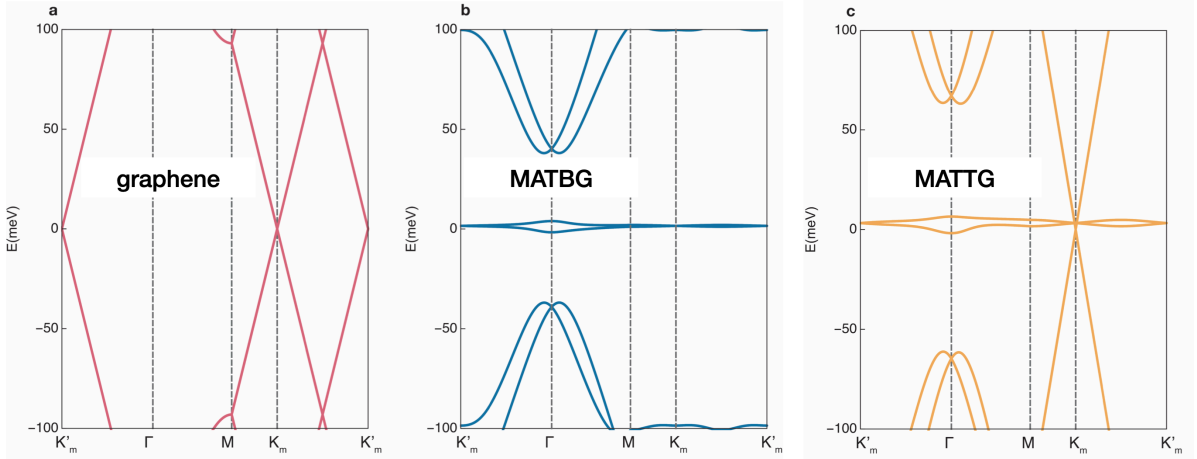


Figure 1.4: **Calculated band structures of twisted multilayer graphene.**

(a) Calculated band structure of monolayer graphene, (b) magic-angle twisted bilayer graphene, and (c) magic-angle twisted trilayer graphene. Adapted from Ref. [78]

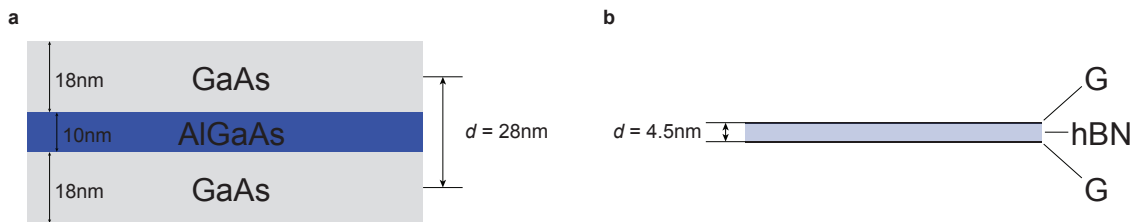
## 1.4 Quantum Hall bilayer

While twistrionics represents a new band engineering method made possible by vdW heterostructures, quantum Hall bilayers have a longer history, rooted in earlier studies of

two-dimensional electron systems [79]. Studies of double-layer two-dimensional electron gas (2DEG) systems in the quantum Hall regime date back several decades, particularly in GaAs/AlGaAs-based heterostructures [79–96]. However, the advent of vdW assembly has revitalized this platform, enabling new levels of cleanliness, tunability, and layer-by-layer control—especially using graphene/hBN-based systems [48–55, 97–101].

In quantum Hall bilayers, two closely spaced 2DEG systems are subjected to a strong perpendicular magnetic field. When the interlayer spacing distance  $d$  is comparable with the magnetic length  $l_B = \sqrt{\hbar/(eB)}$ , where  $\hbar$  is the reduced Planck constant,  $e$  is elementary charge and  $B$  is the perpendicular magnetic field, Coulomb interactions between electrons in different layers become significant. This opens the door to a rich variety of interlayer-correlated phases. One of the most prominent phase in this system is the interlayer exciton condensate at total filling factor  $\nu_{\text{total}} = 1$ , where an electron in one layer binds with a hole in the other to form a neutral bosonic object—interlayer exciton. These excitons spontaneously condense into a phase-coherent superfluid, exhibiting phenomena such as dissipationless counterflow transport and perfect Coulomb drag through the bulk of the sample [83, 89, 93, 102–105].

The dimensionless parameter  $d/l_B$  describes the competition between inter- and intra-layer interactions, which in turn, dictates the phase transitions between interlayer decoupled states to interlayer coherent states. Thus naturally, it is beneficial to create access to a



**Figure 1.5: Schematic of quantum Hall bilayer structure.**

Structure of a typical (a) GaAs/AlGaAs-based quantum Hall bilayer sample, based on values reported in Ref. [93]. (b) Graphene/hBN-based quantum Hall bilayer sample. The interlayer distance  $d$  is clearly reduced. Thickness plotted to scale.

large range of values for  $d/l_B$  experimentally. With this vision in mind, graphene/hBN-based quantum Hall bilayers, in particular, offers as a groundbreaking technical upgrade to the thin film based heterostructures. The advantage is clear: vdW heterostructures enable unprecedented control of thickness. For the length scale that matters in this problem, graphene has essentially no thickness; the center-to-center distance between the two conductive layers solely depends on the thickness of the spacer between the layers. The dielectric spacer, hBN, can also be thinned to few atomic layers via mechanical exfoliation. Thus the interlayer distance  $d$ , can be reduced by an order of magnitude as demonstrated in Fig. 1.5, significantly increasing the accessible  $d/l_B$  range.

## 1.5 Outline

In this thesis, I summarize my work over the past six years, studying the intricacies of correlated electronic systems, focusing on how utilizing planar and vertical device architectures to realize the new functions and thus probing the special behaviors of these interesting states in graphen multilayer systems.

The following chapters are broadly divided into two parts. In Chapters 2 to 4, we investigate rotational symmetry breaking in graphene multilayers through angle-resolved transport measurements. We then turn our focus to quantum Hall bilayer systems in Chapters 5 to 6, exploring interlayer excitons emerging in the fractional quantum Hall regime. Each chapter begins with a brief overview of the relevant background, followed by a description of the experimental methods specific to that study. We then present the core experimental results and conclude with an analysis and interpretation of the data.

Together, these studies aim to showcase how electronic correlation can manifest as symmetry breaking orders and competition between interactions, giving rise to novel quantum phases in low-dimensional systems.

# CHAPTER 2

## Angle resolved transport in twisted trilayer graphene

### 2.1 Superconductivity in twisted multilayer graphene

As mentioned in the introduction, magic angle twisted bilayer graphene (MATBG) and MATTG share lots of similarities. From band theory, they both host low energy flat bands with a four-fold degeneracy, accounting for the two spins (denoted as  $\uparrow$  and  $\downarrow$ ) and two valleys in the mini Brillouin zone coming from the moiré superlattice (denoted as  $K$  and  $K'$ ) [39, 40, 45, 70]. These degrees of freedom, together referred to as isospin flavors, give rise to a natural choice of unit for the charge carrier density, moiré filling  $\nu$ , where  $\nu = x$  means there are  $x$  electrons (or holes if  $x < 0$ ) per moiré cell. With this notation, a fully filled moiré band would correspond to  $\nu = \pm 4$  and within this range is where we will be focusing on. Phenomenologically, they both host correlated insulators at integer moiré fillings, understood as a spontaneous polarization of the isospin flavors. When the moiré band is filled to a integer value, the carriers spontaneously polarize into one isospin flavor, resulting in the opening a pseudo-gap and a resistive peak. And the system loses an isospin degeneracy from that on, supported by the hierarchy of the Landau levels originated from

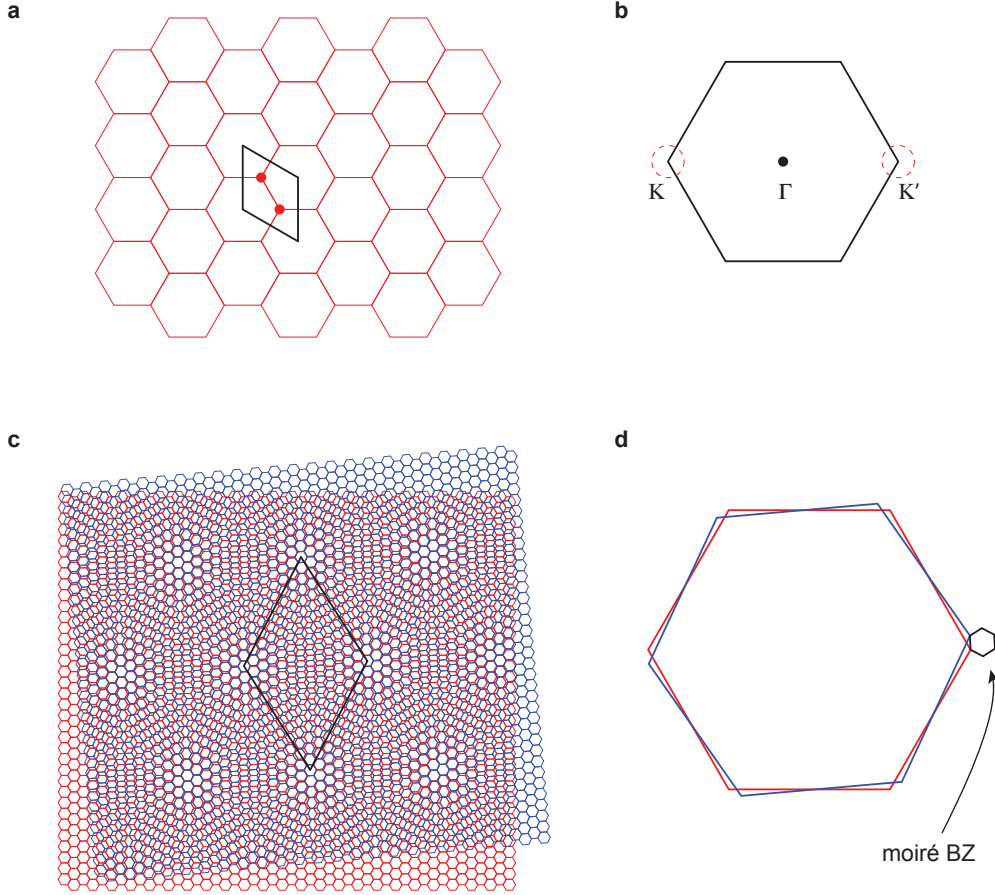


Figure 2.1: **Brillouin zone of graphene and twisted graphene.**

(a) An illustration of crystal lattice of monolayer graphene in real space, the black diamond marks a unit cell with two atoms inside, denoted by red dots. (b) Reciprocal lattice of monolayer graphene in Brillouin zone, with two inequivalent valleys,  $K$  and  $K'$ , where the famous Dirac cones reside. (c) An illustration of the moiré lattice of TBG in real space, the black diamond marks a moiré unit cell. (d) In reciprocal space, mini Moiré Brillouin zones are formed at the original graphene valleys.

each integer filling [77]. Pomeranchuk effect at  $\nu = -1$  and orbital ferromagnetism are also among the key features of these moiré systems. But most importantly, they also both host SC at around half filling  $\nu = \pm 2$  (See Fig. 2.2a-b). [40–42, 45, 47, 63, 68, 72, 106].

One particular motivation of studying moiré systems is to understand the mechanism of its SC. How does SC emerges from a simple twist, and whether it is conventional are still debatable topics. A hand wavy argument starts from the criteria of magic-angle. Take TBG for example, at magic angle, the interlayer and intralayer hopping rates become



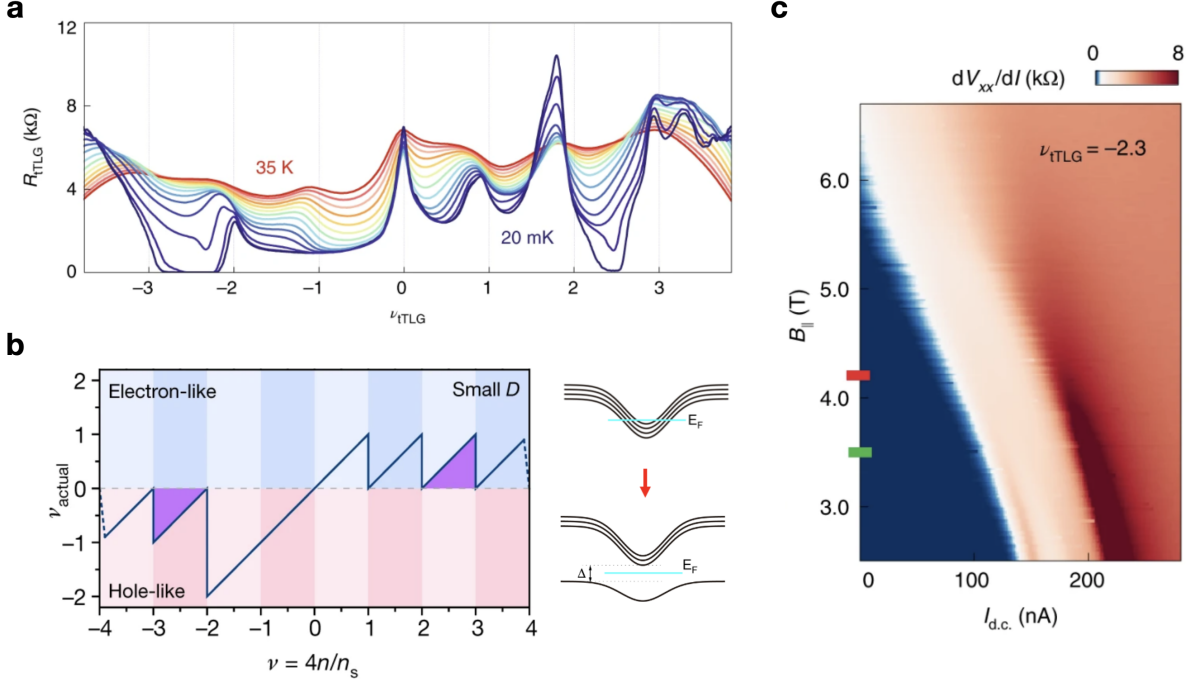


Figure 2.2: **Transport features of MATTG.**

(a) Longitudinal resistance  $R_{xx}$  as a function of moiré filling  $\nu$ , measured at different temperatures. Key features include correlated insulators appearing at integer moiré fillings, SC at near  $\nu = \pm 2$ , and Pomeranchuk effect at  $\nu = -1$ . (b) An illustration of isospin polarization at integer moiré fillings. (c)  $R_{xx}$  as a function of d.c. current bias  $I_{\text{d.c.}}$  and in-plane magnetic field  $B_{||}$ . The red and green ticks mark the Pauli limit determined from  $T_c$  at  $B = 0$ , which are defined as 50% and 10% of the normal-state resistance, respectively. Adapted from Ref. [45, 72]

comparable. Thus the bands from both layers hybridize, creating the flat band which quenches kinetic energy. As a result, electron-electron interaction dominates and promotes different kinds of correlation-driven phase, including SC. This argument, despite being very hand wavy, is consistent with experimental observations of the rich low temperature phase diagram consisting of so many correlation driven states. However, it also begs the question by asserting that the SC in twisted multilayer graphene is correlation driven and thus *unconventional*.

One evidence supporting this assertion is the observation of Pauli limit violation in MATTG (See Fig. 2.2c) [46, 72]. Conventional BCS model for SC assumes a spin singlet structure for the Cooper pairs. Using the critical temperature  $T_c$  of the superconductor at

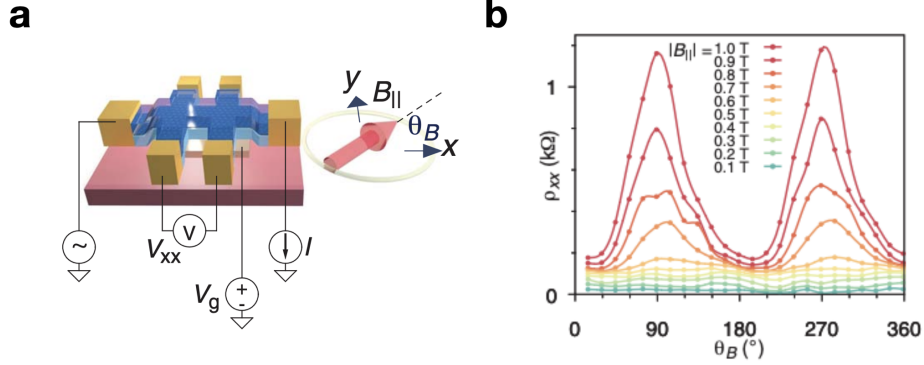


Figure 2.3: **Anisotropic SC in MATBG.**

(a) Schematic of the experimental setup. The MATBG sample is patterned into a Hall bar geometry. A vector magnet is used to rotate the in-plane magnetic field. (b) Longitudinal resistivity  $\rho_{xx}$  as a function of in-plane magnetic field direction  $\theta_B$ , measured at different in-plane magnetic field strength  $|B_{||}|$ . SC is suppressed more easily along  $\theta_B \approx 90^\circ$  and  $270^\circ$ , demonstrating its anisotropic nature. Adapted from Ref. [107]

$B = 0$ , one can calculate a critical in-plane magnetic field, the Pauli limit, at which a spin singlet pair would break, given by:

$$B_{||}^{Pauli} = 1.86(\text{TK}^{-1}) \times T_c, \quad (2.1)$$

assuming  $g$  – factor = 2 [108, 109]. The violation of this Pauli limit points towards the possibility of a spin triplet structure for the Cooper pairs in MATBG samples. This is a very encouraging evidence, though it does not rule out more exotic singlet structures like for example a precessing singlet pair resembling a canted anti-ferromagnetism.

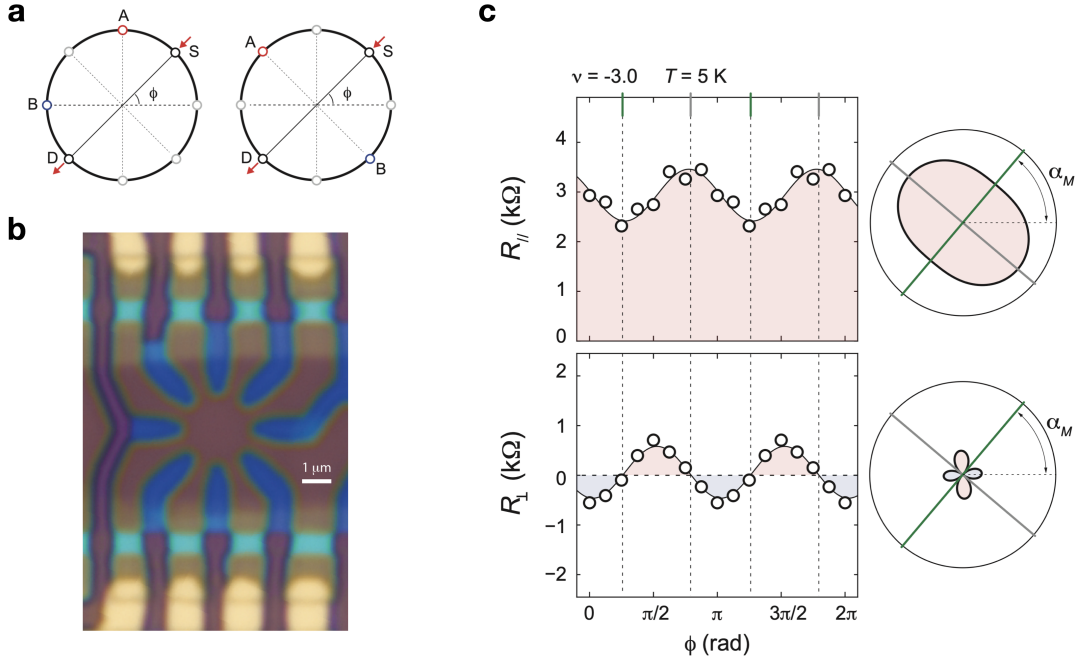
Another way to support the assertion is to study the pairing symmetry of the SC. Cooper pairing in conventional BCS superconductors is  $s$ -wave, isotropic along all directions without any node. By either examining the pairing strength along different directions or searching for the nodes in the superconducting gap, one may be able to make some conclusions about the nature of this superconductor. One exemplary work adapting the former idea is Ref. [107]. A vector magnet is used to suppress SC along different directions, yielding an observation of anisotropic SC as shown in Fig. 2.3. Assuming an

isotropic high temperature metallic state, this could be a very conclusive observation, claiming a non-*s*-wave, nematic pairing symmetry for the superconducting state in MATBG. However, the experimental setup does not generally allow a straightforward extraction of the anisotropy of the metallic state, thus leaving the question unsolved: is the anisotropy of the SC intrinsic to the pairing order, e.g. nematic? Or is it inherited from the parent metallic state? An example of the latter case would be a superconductor with an intrinsically isotropic *s*-wave pairing, formed in a strained, anisotropic lattice, thus also behaves anisotropically. The idea of search for nodes in the superconducting gap has also been experimentally explored, for example using STM methods [71]. The difficulty here is that the distinction between a U-shaped and V-shape superconducting gap is smeared at finite temperatures; instrumental resolution also makes the distinction quantitative rather than qualitative.

The pairing order question and the observed anisotropy in graphene moiré systems motivated me to launch this project that spanned nearly half of my Ph.D. study. To systematically study the evolution of anisotropy across the low temperature phase diagram in these systems, we employ a “sunflower” geometry—a radial multi-terminal device structure that enables angle-resolved transport measurements with minimal geometric bias. In the following section, I will discuss how to use “sunflower” geometry to extract out information regarding rotational symmetry that are generally hidden to Hall bar geometries.

## 2.2 Sunflower geometry and angle resolved transport

Nematicity describes the Coulomb driven, spontaneous breaking of rotational symmetry. When facing a system with possible nematic, or more generally anisotropic, order, it is most natural to probe the system using a geometry that allows angle resolve transport measurement. In the case of high  $T_c$  cuprate, this geometry can be a collection of multiple Hall bars, patterned along different directions [110]. By performing measurement over



**Figure 2.4: Angle resolved transport and sunflower geometry.**

(a) Schematic of longitudinal  $V_{\parallel}$  (left) and transverse  $V_{\perp}$  (right) measurement with a sunflower geometry. The large circle represents the disk shaped sample, the open dots represents the contacts on the circumference. (b) Optical image of a MATTG sample patterned into sunflower geometry. The diameter of the disk is about  $2 \mu\text{m}$ . (c) Example of angle resolved transport measurement from a MATTG sunflower sample at  $\nu = -3$  and  $T = 5$  K. Anisotropy is clearly present at this metallic state. Adapted from Ref. [77]

each Hall bar, Wu et al. were able to plot longitudinal and transverse resistance as a function of the azimuth direction of the current flow,  $\phi$ . Thus giving them unambiguous evidence for anisotropy in the metallic state. However this is a rather luxury design as it requires a very large sample area ( $> 1 \text{ mm}^2$  in Ref. [110]), which is generally not viable for high quality, exfoliated, 2D samples.

To overcome this issue, we develop the so called “sunflower” geometry. Essentially, the sample is patterned into a disk, with multiple (in our case, eight) electrical contacts evenly spaced on the circumference, as shown in Fig. 2.4. The sunflower geometry allows current to flow along different directions, thus enabling a direct observation of anisotropy (for a more detailed description see Appendix A). Take Fig. 2.4c for example, in the metallic phase, longitudinal resistance  $R_{\parallel}$  and transverse resistance  $R_{\perp}$  are measured along

different directions then fitted to:

$$R_{\parallel}(\phi) = R_0 - \Delta R \cos(2(\phi - \alpha_M)) \quad (2.2)$$

$$R_{\perp}(\phi) = R_H + \Delta R \sin(2(\phi - \alpha_M)). \quad (2.3)$$

Here,  $\Delta R$  represents the oscillation amplitude of the resistance in both channels and  $R_0$  is the average value of  $R_{\parallel}(\phi)$ . Their ratio,  $\Delta R/R_0 = 0.17$ , describes the strength of anisotropy.  $\alpha_M = 45^\circ$  is the direction associated with the highest conductivity (or lowest resistivity) in the metallic phase.  $R_H \approx 0$  is the average value of  $R_{\perp}(\phi)$ , indicating the absent of Hall effect.

## 2.3 Annex: Full power of the sunflower geometry

There is a small caveat presenting in Fig. 2.4: There are eight contacts along the circumference of the disk, indicating 8 angular entries from each channel with a total of 16 points. But clearly there are 32 total points shown in panel c. This discrepancy comes from the fact that we can actually use more measurement configurations than just the most intuitive ones as shown in Fig. 2.4a.

In principle, the conductivity matrix:

$$\begin{bmatrix} \sigma_{xx} & \sigma_H \\ -\sigma_H & \sigma_{yy} \end{bmatrix}$$

contains three free parameters:  $\sigma_{xx}$  and  $\sigma_{yy}$  describes the conductivity along two orthogonal principle axes, referred to as  $x$  and  $y$  directions, and  $\sigma_H$  is the Hall conductance. Since the lab frame, arbitrarily chosen by the patterning of the sunflower geometry, does not necessarily align with the principle axes  $x$  and  $y$  of the conductivity matrix, it is necessary to introduce another parameter  $\alpha_M$ , which describes the direction of principle axes in the lab frame. Without loss of generality, we choose the principle axes with higher conductivity

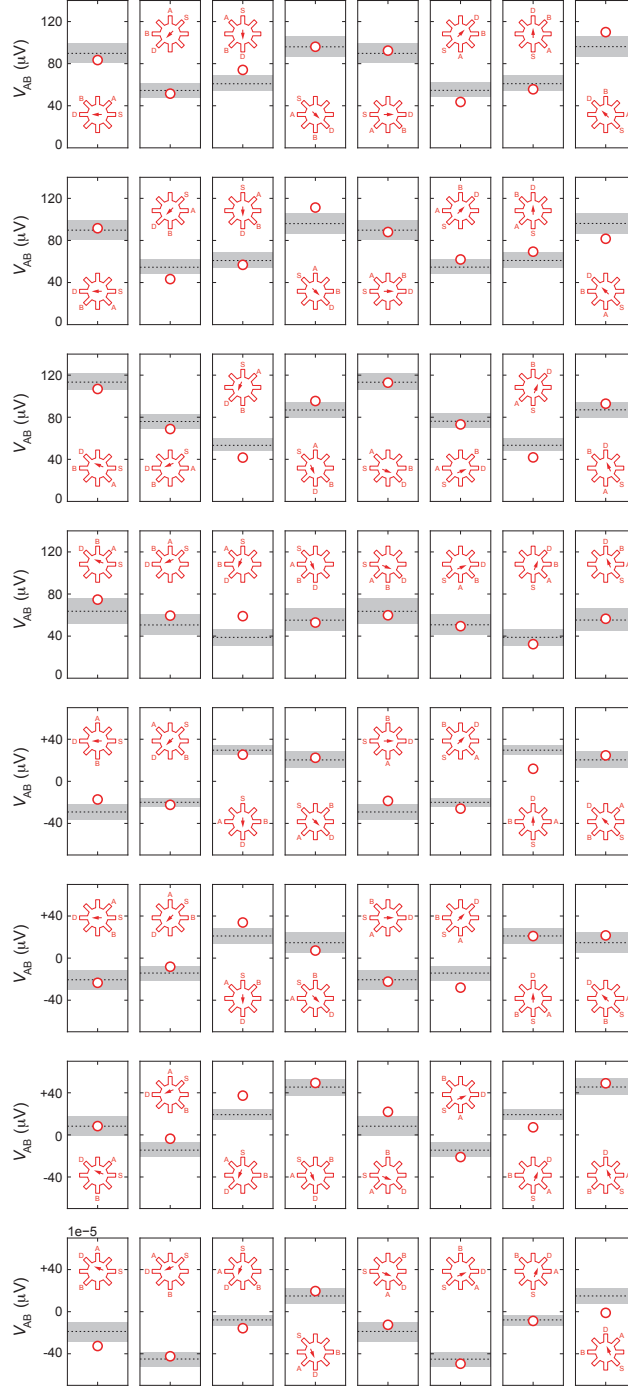


Figure 2.5: **The “sunflower” model beyond angular fit.**

An example of 8 different measurement configuration, with each 8 angular entries, in total of 64 data point, denoted as open circles, compared against their expected values extracted from a single conductivity matrix, marked as black dashed lines. The gray stripes mark the expected measurement uncertainty from the finite width of the contacts. Adapted from Ref. [76]

as the direction of  $\alpha_M$ . Note that these four parameters together describe the system equivalently as the previously mentioned parameters ( $\Delta R$ ,  $R_0$ ,  $R_H$ , and  $\alpha_M$ ) in Eqn. 2.2 and 2.3, related by:

$$R_0 \sim \frac{\frac{1}{2}(\sigma_{xx} + \sigma_{yy})}{\sigma_{xx}\sigma_{yy} + \sigma_H^2} \quad (2.4)$$

$$R_H \sim \frac{\sigma_H}{\sigma_{xx}\sigma_{yy} + \sigma_H^2} \quad (2.5)$$

$$\Delta R/R_0 \sim \frac{\sigma_{xx} - \sigma_{yy}}{\sigma_{xx} + \sigma_{yy}} \quad (2.6)$$

and  $\alpha_M$  is shared in both sets of parameters. Notice that I related the parameters with “ $\sim$ ” signs. This is because there are also subtleties from the geometry shapes of the sample and measurement configurations of our  $R_{\parallel}$  and  $R_{\perp}$ . One can handle this by solving Ohm’s law:

$$\mathbf{j} = \sigma \cdot \mathbf{E} = -\sigma \cdot \nabla V, \quad (2.7)$$

where  $\mathbf{j}$  is the current density,  $\sigma$  is the conductivity matrix,  $\mathbf{E}$  is the electric field, and  $V$  is the electrical potential on the disk, with a boundary condition that assumes a uniform and local conductivity matrix within the disk and no conduction outside the disk. With some serious theoretical effort, one can calculate the expected voltage measurement, with arbitrarily chosen source drain and voltage leads locations on the circumference of the disk with any arbitrary conductivity matrix [111]. Thus reversely, one can perform multiple four-terminal measurements, with arbitrarily chosen contacts, to extract out the unknown conductivity matrix (for details see Appendix A - B) [73, 76]. Having four free parameters means minimally we need four perfect four-terminal measurement to perform the matrix extraction. In reality, we generally perform around 50 measurements to eliminate the influence from measurement noise, as shown in Fig. 2.5.

Given this power, we can also calibrate the geometric factors between different measurement configurations. For example, the first and second row in Fig. 2.5 are geometrically equivalent. The angle dependence of these two rows are expected to have the exact same

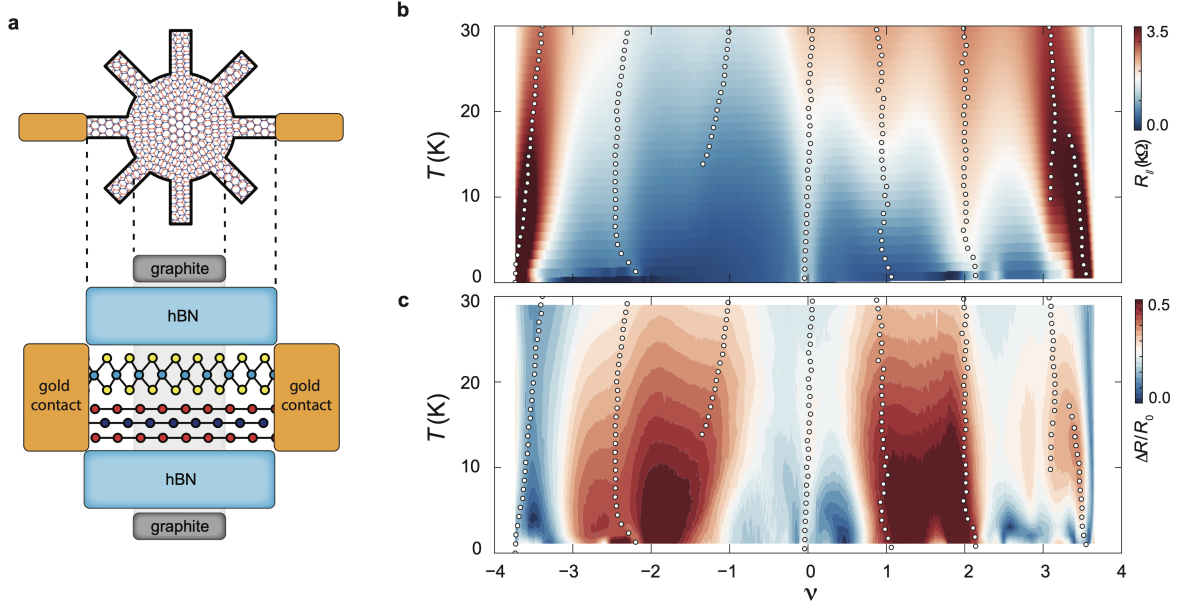


Figure 2.6: **Transport anisotropy in the metallic state of MATTG.**

(a) A schematic of the sample structure. The sunflower geometry allows the angle resolved transport measurement. (b)  $R_{\parallel}$  as a function  $\nu$  and  $T$ . Black open dots denote local peaks of resistance. (c)  $\Delta R/R_0$  as a function  $\nu$  and  $T$ . Adapted from Ref. [77].

result, in both amplitude and phase. To the contrary, the first and the third row are geometrically inequivalent. The angle dependence of these two rows has a phase shift of  $22.5^\circ$ , and a scaling in amplitude that's dependent on the anisotropy strength. We show some examples of this calibration in Appendix C. This geometric calibration frees us from the constrain of only 16 data entries, increasing the credibility of the anisotropy behaviors observed.

Beyond addressing the caveat discussed here, this brief annex eventually feeds into the broader framework for extracting the full conductivity tensor to second order in current, as we will develop further in Chapter 4.

## 2.4 Anisotropy in the metallic states in MATTG

Now we focus on our experiment of angle resolved transport over MATTG. The sample structure is shown in Fig. 2.6a. A MATTG sample is dually encapsulated in graphite gates



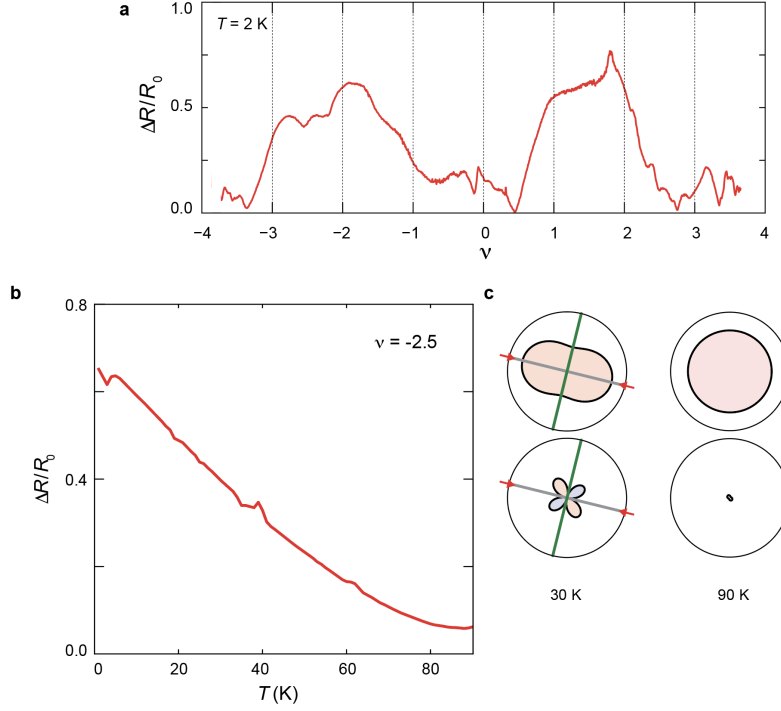


Figure 2.7: **Density and temperature dependence of  $\Delta R/R_0$ .**

(a)  $\Delta R/R_0$  as a function  $\nu$ , measured at  $T = 2$  K. (b)  $\Delta R/R_0$  as a function of  $T$ , measured at  $\nu = -2.5$ . (c) Angle resolved transport measurement taken at  $\nu = -2.5$ ,  $R_{\parallel}$  on top and  $R_{\perp}$  on bottom. Comparison between  $T = 30$  K (left) and  $T = 90$  K (right) illustrates the weakening of anisotropy at high temperatures. Adapted from Ref. [77]

and hBN dielectric spacers. A bilayer  $\text{WSe}_2$  is placed next to the MATTG to introduce spin orbital coupling (SOC) via proximity effect. The heterostructure is made using the standard cut-and-stack method with a poly(bisphenol A carbonate)/polydimethylsiloxane (PC/PDMS) on a glass slide and subsequently released on to a  $\text{Si}/\text{SiO}_2$  substrate. The sample is patterned into the sunflower geometry with electron-beam lithography and reactive ion etching. The electrical contacts are evaporated using  $\text{Cr}/\text{Au}$  with thickness 2nm/100nm. The overall fabrication procedure is within the community standard. Ref. [112] provides a detailed summary of the procedures.

Fig. 2.6b shows  $R_{\parallel}$  as a function of  $\nu$  and  $T$  in a TTG sample with twist angle of  $\theta = 1.55^\circ$  (close to the expected magic angle of TTG). Prominent features includes: correlated insulators at integer fillings, indicated by the resistive peaks, marked with

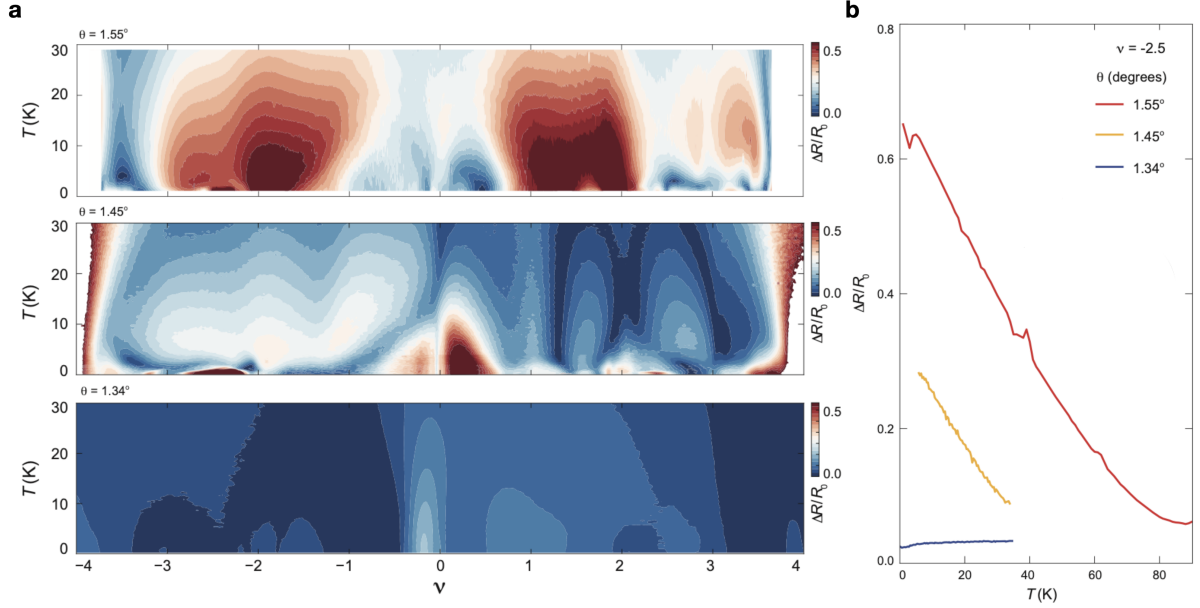


Figure 2.8: **Twist angled dependence of transport anisotropy.**

(a)  $\Delta R/R_0$  as a function  $\nu$  and  $T$ , measured across three samples with twist angle  $\theta = 1.55^\circ$  (top),  $\theta = 1.45^\circ$  (middle) and  $\theta = 1.34^\circ$  (bottom). Anisotropy is generally weaker as twist angle deviates from the magic angle. (b)  $\Delta R/R_0$  as a function of  $T$ , measured at  $\nu = -2.5$  across three samples. Adapted from Ref. [77]

black open dots. Pomeranchuk effect at  $\nu = -1$ , indicated by the fact that resistive peak only emerges at higher temperatures ( $T > 10$  K). SC near  $\nu = \pm 2$ , evidenced by the zero resistance domes (dark blue), which we will touch upon later. We also perform angle resolved transport measurement at every point in the  $\nu - T$  phase diagram, thus extracting the anisotropy strength map,  $\Delta R/R_0$  as a function of  $\nu$  and  $T$ . Notably, the most pronounced anisotropy appears (dark red regions in Fig. 2.6c) near half-filling on both the electron-doped and hole-doped side of the moiré band at relatively low temperatures, coinciding with a three-fold isospin degenerate order. On the other hand, the anisotropy is weak at charge neutrality point (CNP) and moiré band edge  $\nu = \pm 4$ , where the density of states is low and electron-electron interaction is weak. This observation positively correlates anisotropy in MATTG with and electron-electron interaction strength. Linecuts at  $T = 2$  K and  $\nu = -2.5$  clearly demonstrate this observation (Fig. 2.7). We further demonstrate the positive correlation between electron-electron interaction and anisotropy

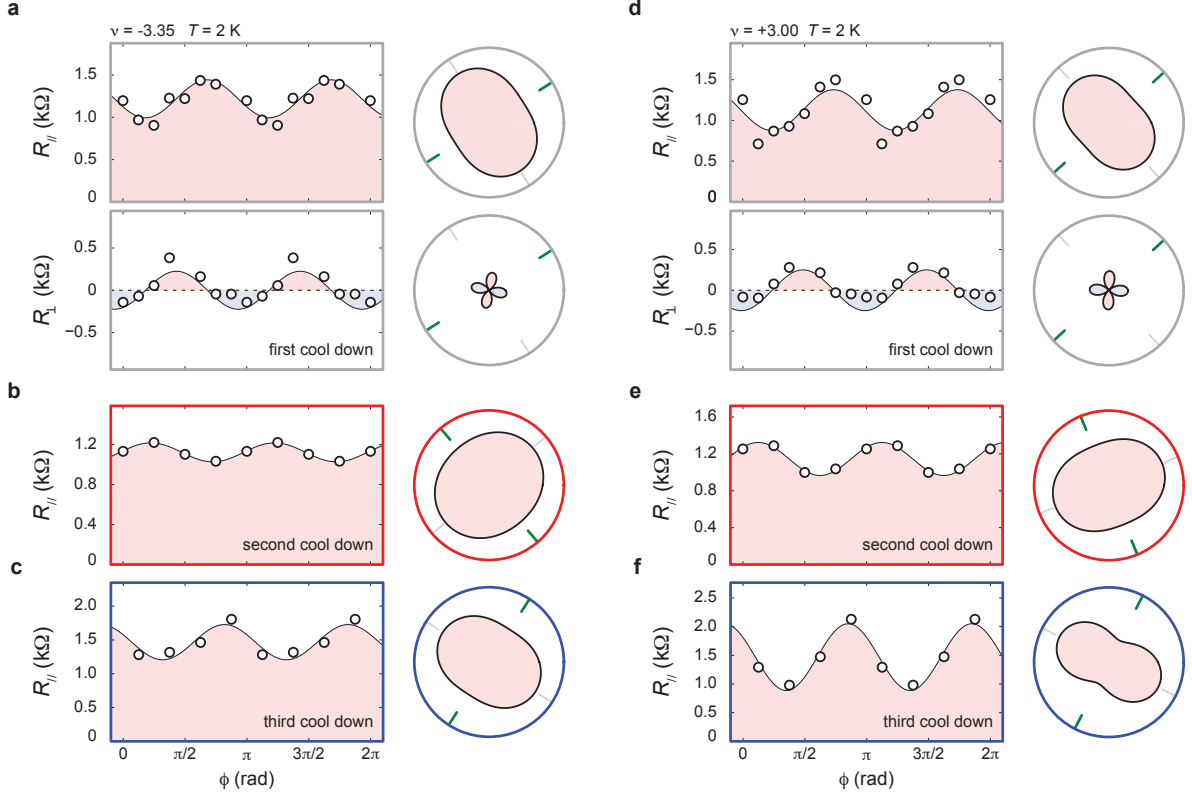


Figure 2.9: **Temperature induced hysteresis in the metallic state.**

The angular dependence of transport responses measured at (a-c)  $\nu = -3.35$  and (d-f)  $\nu = +3.00$  after multiple thermal cycles. Reproduced from Ref. [77]

by contrasting the anisotropic behaviors across multiple TTG samples with various twist angles. As shown in Fig. 2.8, anisotropy is generally weaker as twist angle deviates from the magic angle. All together, the density, temperature, as well as the twist angle dependence suggests that the anisotropy in TTG has a Coulomb-driven origin, thus possibly nematic.

Another evidence supporting the Coulomb-driven picture is the observation of temperature-induced hysteresis of anisotropy. If the rotational symmetry is spontaneously broken, then after restoring and breaking the symmetry again, the anisotropic order could pick up another direction as its principle axis. This observation is demonstrated in Fig. 2.9; the angle resolved transport measurement at each thermal cycle identifies the principle axis  $\alpha_M$ , which is clearly rotating across the thermal cycles. Note that the anisotropy strength  $\Delta R/R_0$  also exhibits hysteretic behavior across the thermal cycles. This could be caused by the formation of domains with different anisotropic orders, but the excellent

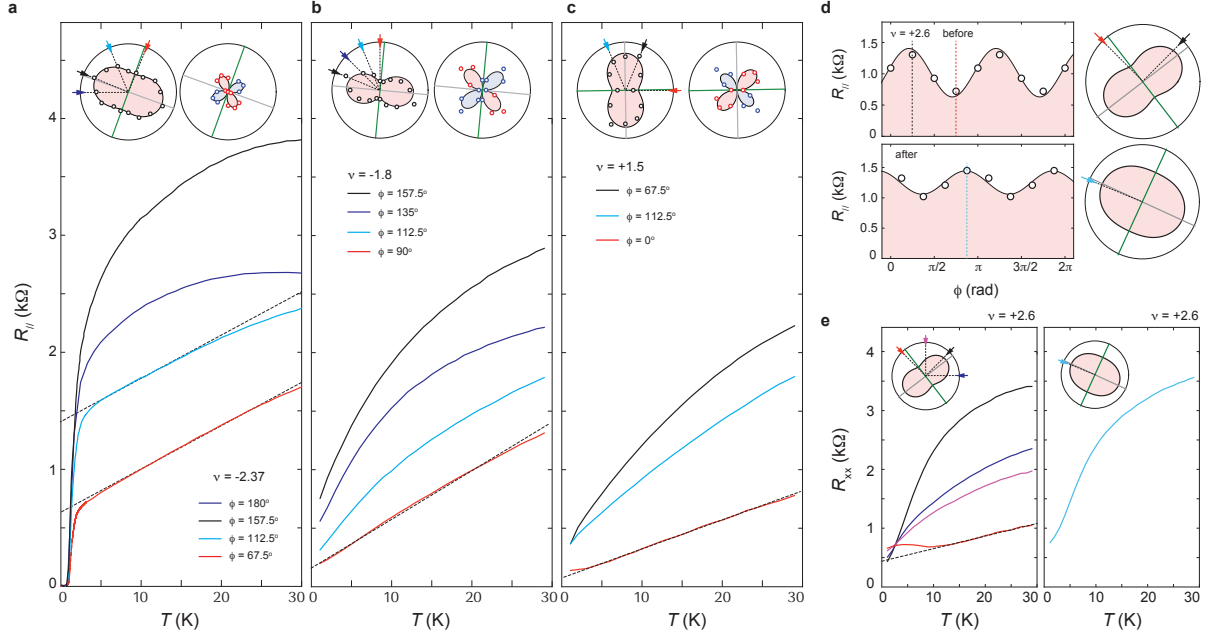


Figure 2.10: **Anisotropy of strange metallicity.**

(a-c)  $R_{\parallel}$  as a function of  $T$  measured along different azimuth direction  $\phi$  at (a)  $\nu = -2.37$ , (b)  $\nu = -1.8$ , and (c)  $\nu = +1.5$ . Insets: angle resolved transport in polar-coordinate plots, measured at  $T = 5$  K. Green and gray solid lines denote the principal axes corresponding to highest and lowest conductivity. Red arrows mark the directions of maximum conductivity, which also exhibits the linear-in- $T$  behavior (red solid curves). (d) The angular dependence of transport responses measured at  $\nu = 2.6$  before (top) and after (bottom) temperature-induced hysteresis. (e)  $R_{\parallel}$  as a function of  $T$  measured at  $\nu = 2.6$  before (left) and after (right) temperature-driven hysteresis. Reproduced from Ref. [77]

fit of Eqn. 2.2 and 2.3 suggests that the sample is still generally uniform and the domains are likely much smaller compared to the length scale of the sample.

Strange metallicity is also often associated with nematicity. In many high-temperature SC systems, the resistance of the metallic state above the superconducting transition temperature increases linearly with temperature [113, 114]. This behavior contradicts the quadratic relation expected from the Fermi liquid model thus referred to as strange metallicity. The exact relation between this phenomenon and the nematic SC in the high- $T_c$  systems is still an unsolved problem, but their often juxtaposition in many cuprate phase diagrams suggests a potential quantum critical connection [115–119]. While linear-in- $T$  resistivity has been observed in twisted graphene systems, our sunflower geometry has allowed us to further investigate the interplay between the anisotropy and strange

metallicity [120, 121]. As shown in Fig. 2.10, the linear-in-T resistivity is observed across the moiré band, but only along the directions with high conductivity at each density. Notably, when the anisotropy experience hysteretic transition through thermal cycles, the linear-in-T behavior also hysteretically changes direction. A possible explanation to this anisotropic linear-in-T behavior is the directional dependent coexistence of a linear-in-T contribution and other nonlinear contributions. Along the high resistivity direction, the linear-in-T contribution is masked by the nonlinear contribution, while the linear-in-T contribution is revealed in the low resistivity direction [77].

All together, our findings in the metallic regime provides strong supporting evidence for a generally nematic low-temperature phase diagram in MATTG, accompanied by an angle-dependent manifestation of strange metallicity.

## 2.5 Anisotropy in superconducting MATTG

Now we discuss the anisotropic nature of SC in MATTG and focus on the behaviors at  $\nu = -2.5$ , where SC is the strongest for this particular sample. Due to the zero resistance feature of the SC phase, the angle dependence of  $R_{\parallel}$  and  $R_{\perp}$  is not a viable probe for studying the anisotropy when sample is superconducting. Here, we show two independent methods to characterize the anisotropy of the SC in MATTG.

First, we use the magnitude of critical supercurrent,  $I_c$ , as the indicator of the low temperature stability of SC along different directions against current induced transitions. As shown in Fig. 2.11a,  $I_c$ , corresponding to the peaks in  $I - V$  curves, is different when current flows along different directions. Fig. 2.11b shows the angle dependence of  $I_c$ , measured at  $T = 20$  mK, well below the superconducting transition temperature, which is then fitted with this phenomenological form, similar to Eqn. 2.2:

$$I_c(\phi) = I_{ave} + \Delta I \cos(2(\phi - \alpha_I)), \quad (2.8)$$

where  $\Delta I$  is the oscillation amplitude of  $I_c$  and  $I_{ave}$  is the averaged  $I_c$  over all directions. Most importantly,  $\alpha_I$  denotes the direction of maximum  $I_c$ , representing the direction of strongest SC pairing. Clearly from Fig. 2.11b, the SC is anisotropic.

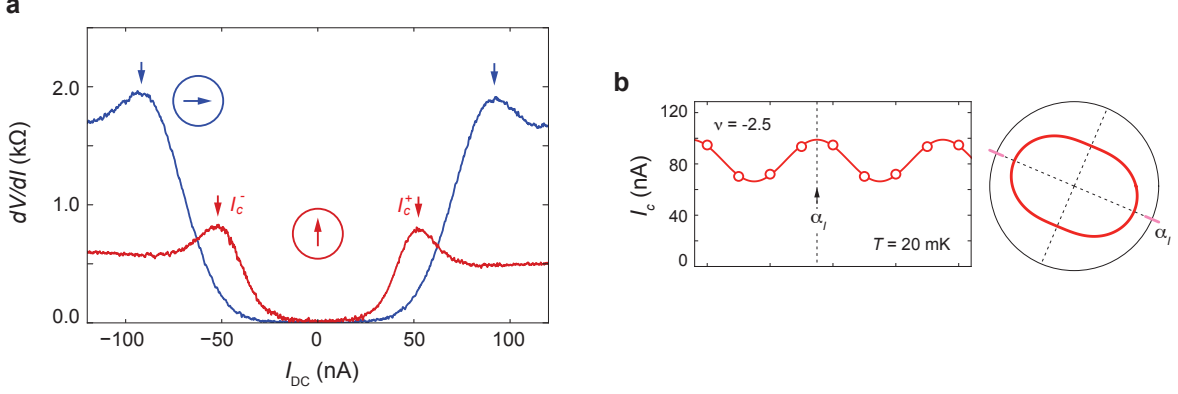


Figure 2.11: **Anisotropy of  $I_c$ .**

(a) Differential resistance  $dV/dI$  as a function of d.c. current bias  $I_{DC}$ , measured at  $\nu = -2.5$ ,  $T = 20$  mK, along different current flow directions. Critical supercurrent  $I_c$  is marked with vertical arrows. (b) Angle dependence of  $I_c$ . Adapted from Ref. [77]

Second, we use the onset temperature of SC pairing,  $T_{onset}$ , as the indicator of the superconducting pairing strength along different directions against temperature induced transitions. As shown in Fig. 2.12a, the solid black line shows the  $R - T$  curve depicting the metal-superconductor transition as  $T$  decreases, evidenced by the diminishing  $R_{\parallel}$  at low temperatures. The black dashed lines is the  $R - T$  curve of the metallic phase, obtained by suppressing the SC with a large current,  $I_{d.c.} = 170$  nA.  $T_{onset}$ , corresponding to the bifurcation temperature between these two  $R - T$  curves, is the indication of the onset of Cooper pairing. It is operationally defined as the temperature at which  $R_{\parallel}$  equals 90% of the metallic phase resistance. Fig. 2.12b shows the angle dependence of  $T_{onset}$ , which is then fitted with this similar phenomenological form:

$$T_{onset}(\phi) = T_{ave} + \Delta T \cos(2(\phi - \alpha_T)), \quad (2.9)$$

where  $\Delta T$  is the oscillation amplitude of  $T_{onset}$  and  $T_{ave}$  is the averaged  $T_{onset}$  over all directions. Most importantly,  $\alpha_T$  denotes the direction of maximum  $T_{onset}$ , representing

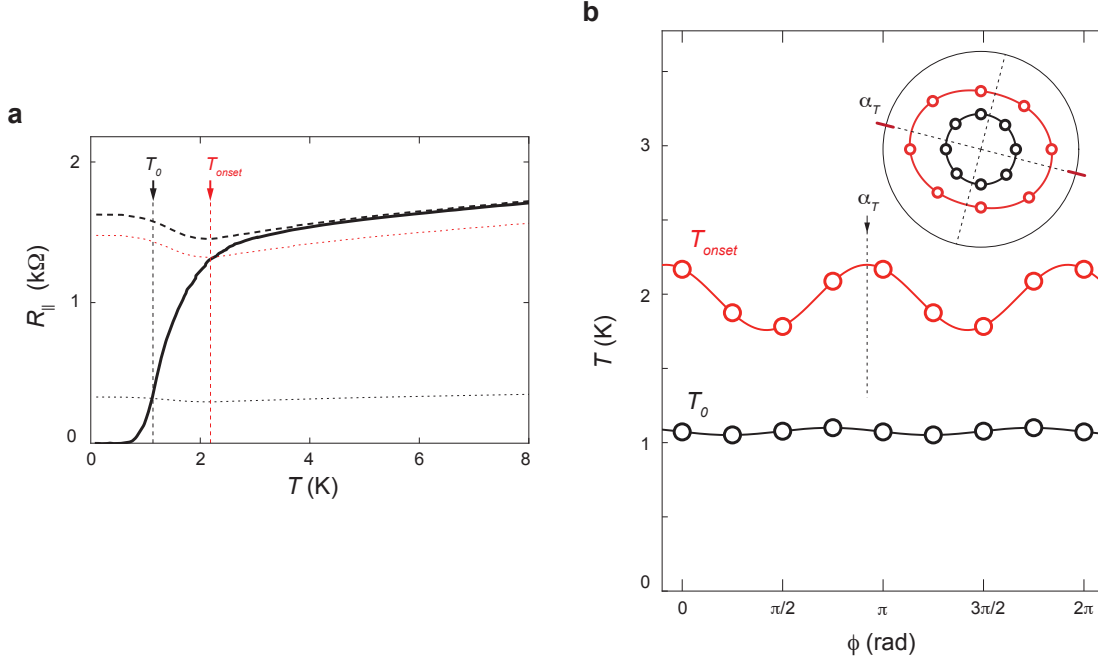


Figure 2.12: **Anisotropy of  $T_{onset}$ .**

(a)  $R_{\parallel}$  as a function of temperature (black solid line), measured at  $\nu = -2.5$  with a small a.c. current  $I_{AC} = 5$  nA. Black dashed line represents the metallic resistance as a function of temperature, measured at  $\nu = -2.5$  with a large d.c. current  $I_{DC} = 170$  nA suppressing SC.  $T_{onset}$  and  $T_0$  are marked with red and black arrows, representing the temperatures at which  $R_{\parallel}$  equals 90% (red dotted line) and 10% (black dotted line) of the metallic resistance. (b) Angle dependence of  $T_{onset}$  (red) and  $T_0$  (black). Adapted from Ref. [77]

the direction of strongest SC pairing. Clearly from Fig. 2.12b, the SC is anisotropic. And most remarkably,  $\alpha_T$  and  $\alpha_I$  aligns closely with each other. This agreement not only checks the consistency between our two methods characterizing the SC anisotropy, but also reveals a connection between the onset SC fluctuation and critical supercurrent.

Another key temperature,  $T_0$ , representing the temperature at which  $R_{\parallel}$  reaching zero is conveniently defined as the temperature when  $R_{\parallel}$  equals 10% of the metallic resistance.  $T_0$  marks the temperature of the SC reaching global coherence, thus is expected and indeed observed to be isotropic, as shown in Fig. 2.12, further demonstrating the observed anisotropy of  $T_{onset}$  is not due to uneven domains of SC in the sample.

We now compare the anisotropic behavior of the SC phase against that of the metallic phase at higher temperature at  $T = 4$  K. Notably,  $\alpha_I$  and  $\alpha_T$  is roughly orthogonal to  $\alpha_M$ .

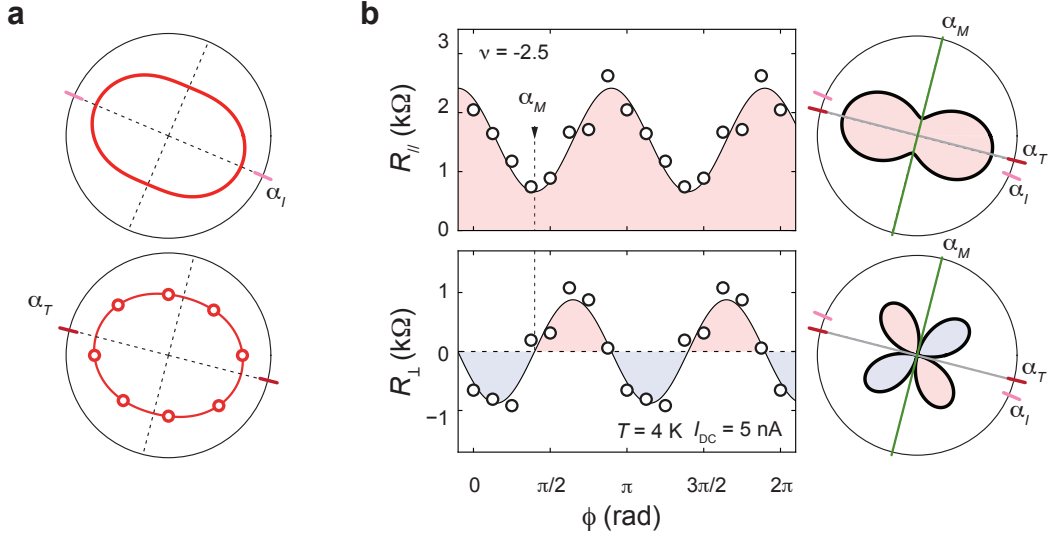


Figure 2.13: **Anisotropy of SC and metallic phase.**

(a) Angle dependence of  $I_c$  (top) and  $T_{onset}$  (bottom) plotted in polar-coordinate. Same data as in Fig. 2.11 and 2.12. (b) Angle resolved transport of the metallic phase,  $R_{||}$  and  $R_{\perp}$  measured at  $\nu = -2.5$  and  $T = 4$  K.  $\alpha_I$  and  $\alpha_T$  are about  $90^\circ$  away from  $\alpha_M$ , as marked in the polar-coordinate plots. Adapted from Ref. [77]

That is, the direction of stronger SC, evidenced by largest  $I_c$  and  $T_{onset}$  is  $90^\circ$  away from the direction of highest conductivity of the high temperature metallic phase, evidenced by the high  $R_{||}$  from the angle resolved measurement. We can extend the study on this surprising observation further by investigating the evolution of anisotropy strength and  $\Delta R/R_0$  and the direction of principle axis, generally  $\alpha$  in the fluctuation regime,  $T_0 < T < T_{onset}$ . As shown in Fig. 2.14b, it makes sense that  $\Delta R/R_0$  decreases shortly below the largest  $T_{onset}$ : The direction of large  $T_{onset}$  is the more resistive direction, as it start gaining conductance through Cooper pairing, the overall anisotropy strength decreases. In the meanwhile, because the stronger direction of SC is  $90^\circ$  away from the principle axis of the metallic phase, as the SC gradually dominates the overall conductance of the sample in the fluctuation regime, the direction of largest conductance naturally rotates from  $\alpha_M$  towards the direction of  $\alpha_I$  and  $\alpha_T$  (Fig. 2.14c). In particular, a linear extrapolation of  $\alpha$  in the fluctuation regime points towards  $\alpha_I$  measured at base temperature.



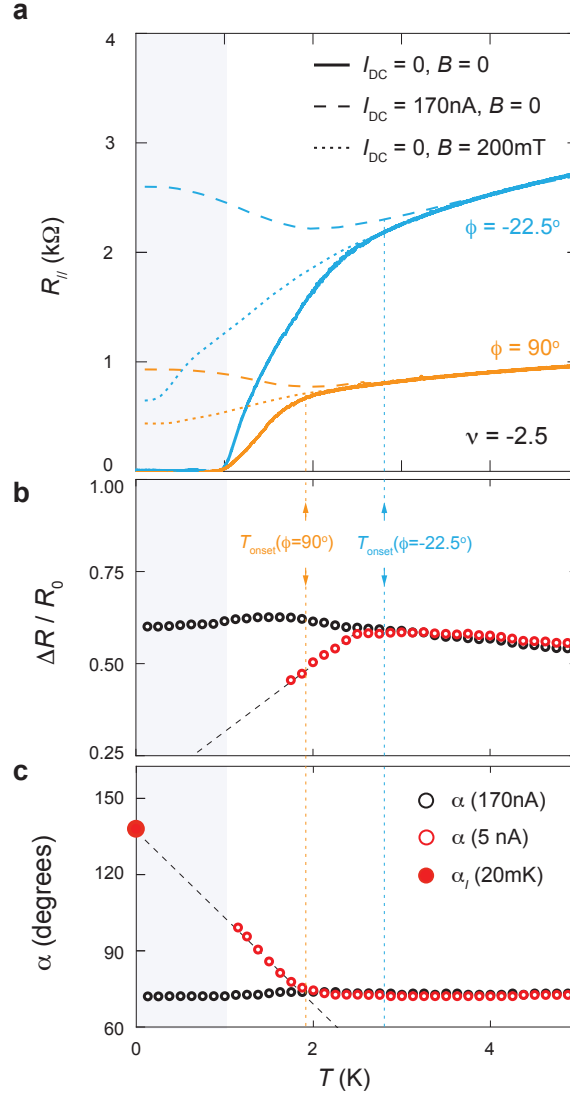


Figure 2.14: **Evolution of anisotropy between metallic and superconducting phases.**

(a)  $R_{\parallel}$  as a function of  $T$ , measured for different current flow directions (solid lines). For each direction, the temperature dependence of the metallic resistance is extracted by suppressing SC using either a small magnetic field  $B = 200 \text{ mT}$  (dotted lines) or a large d.c. current  $I_{DC} = 170 \text{ nA}$  (dashed lines). Cyan denotes the direction near strong SC and small conductance in high temperature metallic phase,  $\phi = -22.5^\circ$ ; orange denotes the direction near weak SC and large conductance in high temperature metallic phase,  $\phi = 90^\circ$  (b) Anisotropy strength  $\Delta R/R_0$  and (c) direction of principle axis  $\alpha$  as a function of temperature, extracted from the angle-resolved transport measurements. Red circles denote the extracted values from the measurements with small a.c. current  $I_{AC} = 5 \text{ nA}$  and  $B = 0$ ; Black circles denote the behaviors from the metallic state, with SC suppressed with a large d.c. current  $I_{DC} = 170 \text{ nA}$ . All data measured  $\nu = -2.5$ . Adapted from Ref. [77]

Similar to the metallic state, the anisotropic behavior in the superconducting phase also exhibits temperature induced hysteresis. As illustrated in Fig. 2.15a-b, both SC and metallic angle resolved transport shows clear rotation of the principle axis after a thermal cycle up to  $T = 300$  K. Despite the hysteretic rotation across the thermal cycle, the orthogonality between  $\alpha_I$  and  $\alpha_M$  is held during both cool downs. In fact, as shown in Fig. 2.15c-d, the principle axis  $\alpha$  in the fluctuation regime rotates towards the preferred direction of SC,  $\alpha_I$ , in both cool downs, demonstrating the reproducibility of the phenomena.

Our findings on the anisotropic behaviors of the superconducting phase, the high temperature metallic states, as well as the evolution in the fluctuations regime in between cast strong constraint for understanding the nature of the SC in MATTG. It is important to note that, the most vital constraint for the arguments below is the orthogonality between  $\alpha_I$  and  $\alpha_M$ , which cannot be obtained by examining the anisotropy of the superconducting state alone. This nematic metallic phase suggests an effective mass anisotropic Fermi surface. Within a BCS model, mass anisotropy alone is insufficient to produce an anisotropic gap. However, an isotropic gap with mass anisotropy will predict a critical supercurrent that is largest in the direction with largest conductivity in the metallic phase, contradicting the observation of  $\alpha_I$  being orthogonal to  $\alpha_M$ . This contradiction also holds when considering vortices as the mechanism for the critical current. In fact, with a more detailed analysis, the observed orthogonality between  $\alpha_I$  and  $\alpha_M$  can only be accounted for by assuming a  $p$ -wave or a mixture of  $s$  and  $d$  wave pairing symmetry [73]. All together, our data rule out a simple  $s$ -wave pairing order mechanism and strongly suggest that the SC pairing order in MATTG is intrinsically anisotropic and thus *unconventional*.

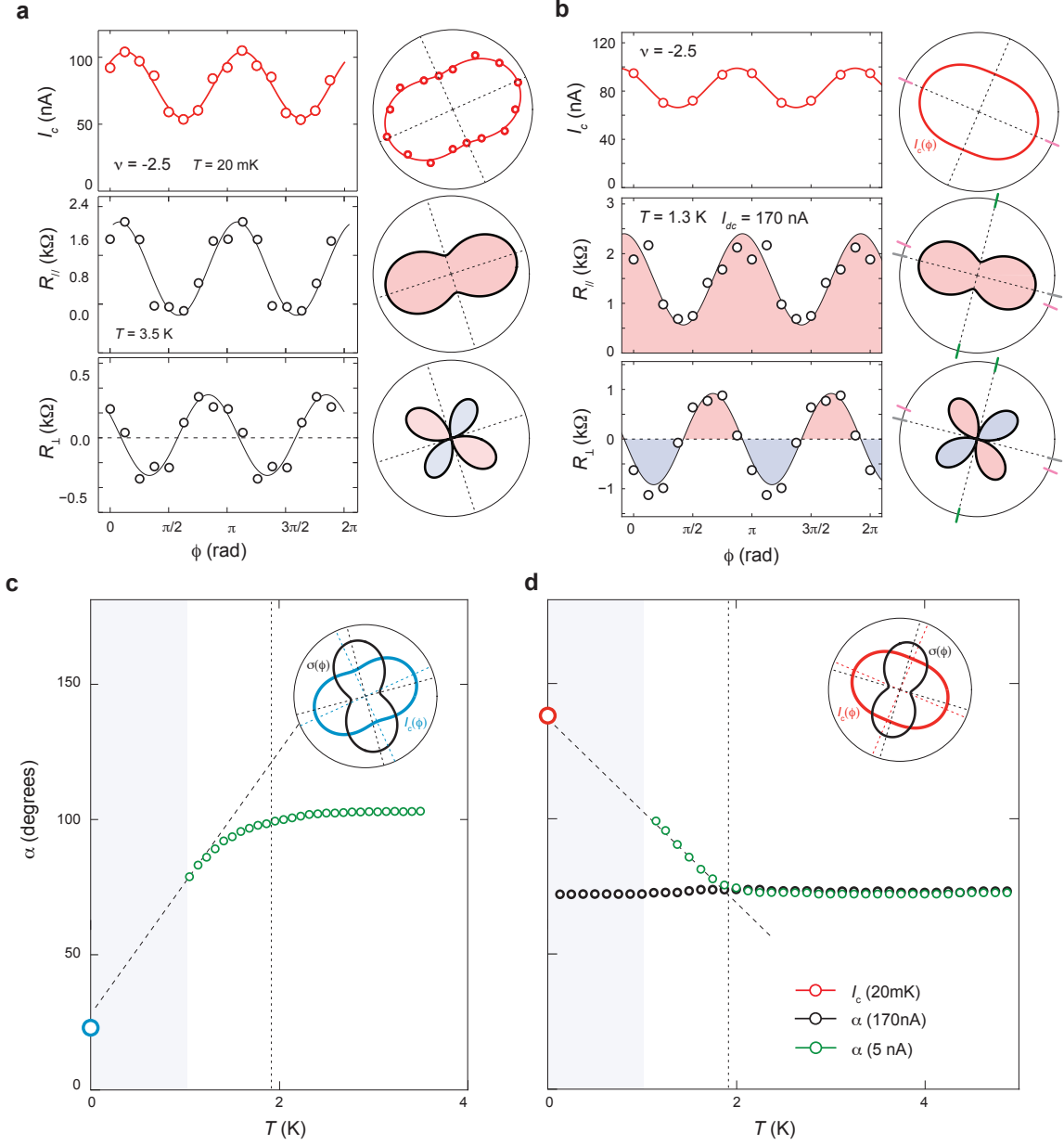


Figure 2.15: **Temperature induced hysteresis in the superconducting phase.**

(a-b) Angle dependence of  $I_c$  (top) measured at  $T = 20$  mK,  $R_{||}$  (middle) and  $R_{\perp}$  (bottom) measured at  $T = 3.5$  K, during the (a) first and (b) second cool down. (c-d) Principal axis orientation  $\alpha$  as a function of  $T$ , measured during (c) first and (d) second cool down. Green open circles denote  $\alpha$  extracted from measurement with small a.c. current  $I_{AC} = 5$  nA. Black open circles denote  $\alpha$  extracted from measurement with SC suppressed with large d.c. current  $I_{DC} = 170$  nA. Blue and orange open circles denote  $\alpha_I$  from the first and second cool down. All data measured at  $\nu = -2.5$ . Adapted from Ref. [77]

# CHAPTER 3

## Non-reciprocal angle resolved transport in twisted trilayer graphene

### 3.1 Non-reciprocity and nonlinearity

In systems that break inversion symmetry, non-reciprocal transport behavior can emerge. A well-known example of this symmetry–phenomenon relationship is the diode: by placing two semiconductors with oppositely charged dopants, a  $p$ – $n$  junction is formed that allows current to flow preferentially in one direction. While perhaps less obvious, a small twist between layers in multilayer graphene also subtly breaks inversion symmetry. As a result, non-reciprocal transport effects are also expected to arise in such moiré systems. In the metallic phase, non-reciprocity describes the resistance difference when sample is biased with opposite d.c. current and is intimately linked to the system’s nonlinear electronic response. For a general  $I - V$  relation:

$$V = a_1 I + a_2 I^2 + a_3 I^3 + \dots, \quad (3.1)$$

where the first term describes the standard Ohm's law and higher order terms contribute to the nonlinear response, all the even-powered terms contribute to the overall non-reciprocal response. This can be understood from the definition of resistance non-reciprocity, defined as:

$$\eta_R(I) = \frac{V(I)}{I} - \frac{V(-I)}{-I} = 2a_2I + 2a_4I^3 + \dots \quad (3.2)$$

If the second order term is the leading nonlinear term in Eqn. 3.1,  $\eta_R(I)$  is then governed only by  $a_2$ . In this case,  $a_2$  can be obtained by measuring the second-harmonic response of an a.c. current. This is most easily understood using basic trigonometric identities. When applying an a.c. current  $I(t) = I_0 \cos(\omega t)$ , the second-order voltage response takes the form:

$$a_2 I_0^2 \cos^2(\omega t) = \frac{1}{2} a_2 I_0^2 [1 + \cos(2\omega t)], \quad (3.3)$$

where  $I_0$  is the current amplitude,  $\omega$  is the angular frequency, and  $t$  is time. The term oscillating at  $2\omega$  constitutes the measurable second-harmonic voltage, from which  $a_2$  can be determined. In this way, second-harmonic transport provides a direct probe of nonreciprocity. Fig. 3.1 demonstrates of this relation experimentally.

So far, we have introduced the concept of non-reciprocity and outlined how second-harmonic transport measurements can be an important experimental probe. These effects fundamentally rely on the breaking of inversion symmetry. In the context of 2D, inversion symmetry and two-fold rotational symmetry  $C_2$  are mathematically equivalent. As a result, non-reciprocity is naturally rotational symmetry breaking and a suitable subject of study using angle resolved transport. Next, we examine the expected angular dependence of non-reciprocal transport and our phenomenological functional description.

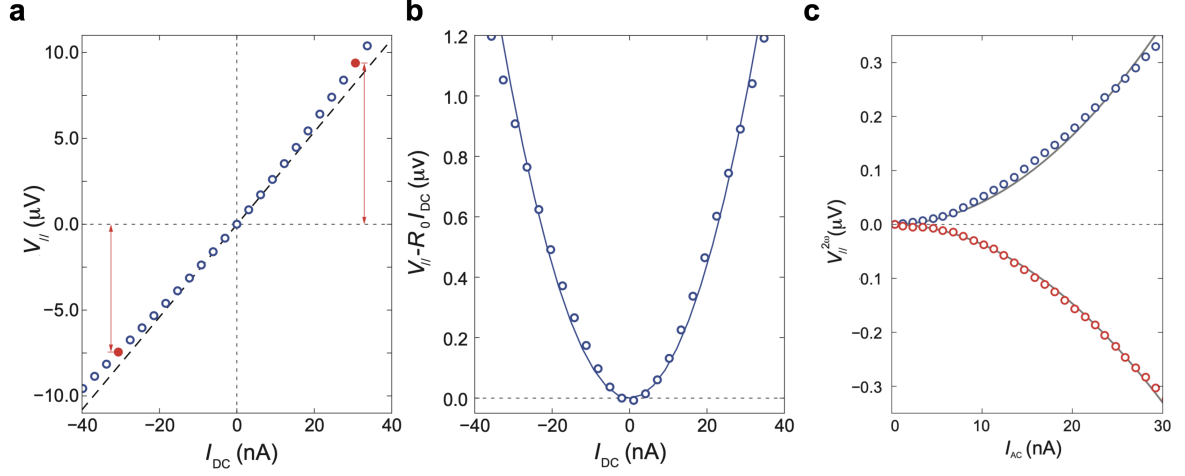


Figure 3.1: **Second harmonic response as a probe for nonreciprocity.**

(a) An example of  $I - V$  characteristics (blue open dots) measured with a d.c. bias. The black dashed line denotes the linear component of the response (taking the slope at  $I_{DC} = 0$ ). The asymmetric deviation from the linear expectation, marked by the red arrows, depicts the voltage non-reciprocity. (b) The nonlinear component of the voltage response (blue open dots), extracted by subtracting the linear component from the data, follows a quadratic fit (blue solid line), suggesting that the second order term in Eqn. 3.1 is the leading nonlinear term. (c) Second harmonic responses (blue and red open dots) measured as a function of a.c. current amplitude, measured along forward (blue) and reverse (red) current directions. As expected, they also follow quadratic fits (solid lines) and have opposite magnitude when the measurement configuration is reversed. All data measured on a TTG sample at  $\nu = 0.2$ ,  $T = 20$  mK,  $B = 0$ . Adapted from Ref. [76]

## 3.2 Non-reciprocal transport with angle dependence

While the linear transport, governed by the  $2 \times 2$  conductivity matrix, has been thoroughly studied across various sample geometries, a complete framework for nonlinear transport remains elusive at the time of our project [111, 122, 123]. To develop an intuition for angle-resolved non-reciprocal transport, at least on the phenomenology level, we begin with a symmetry-based analysis of angle-dependent responses in the linear regime. As demonstrated extensively in the previous chapter, the linear longitudinal  $R_{\parallel}$  and transverse  $R_{\perp}$  responses are expected to follow cosine-like oscillations with 2-fold symmetry (period of  $\pi$ ) with the same oscillation amplitude. They also share a phase shift of  $\pi/4$ , as indicated by the fitting form Eqn. 2.2 -2.3. This behavior can be understood

by assuming two orthogonal mirror axes in the Fermi surface, e.g., the principle axes of the conductivity matrix, around which  $R_{\parallel}$  and  $R_{\perp}$  are expected to be symmetric and antisymmetric, respectively.

Extending this analysis to the nonlinear, non-reciprocal regime, where  $C_2$  symmetry is broken, one would expect the Fermi surface to have odd number of mirror axes. For

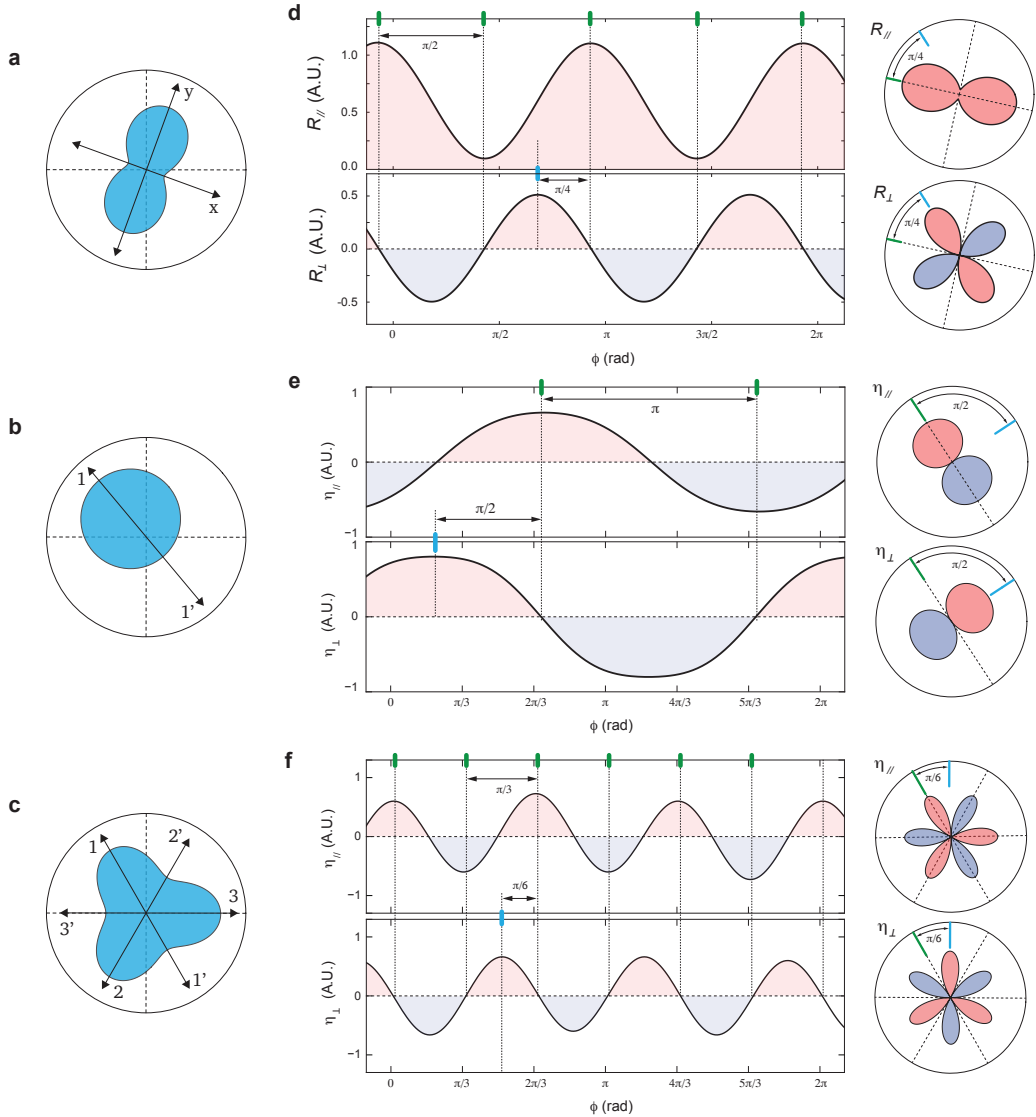


Figure 3.2: **Symmetry analysis of the angle resolved transport response.**

(a-c) Schematics of the Fermi surface contour with different angular symmetries. The black arrows mark the azimuth direction of the mirror axes. (d-f) Expected angle resolved transport responses in the (d) linear regime and non-reciprocal regime with (e) 1 mirror axis and (f) 3 mirror axes. Adapted from Ref. [76]

example, in the case where the system retains only 1 mirror axis, we expect an angle resolved transport non-reciprocity to follow a cosine-like modulation with 1-fold symmetry with a period of  $2\pi$ . Analogous to the linear case, the longitudinal  $\eta_{\parallel}$  and transverse  $\eta_{\perp}$  non-reciprocity are expected to be symmetric and antisymmetric around that mirror axis, respectively, resulting in a relative phase shift of  $\pi/2$ . We can further generalize this argument to a system with three mirror axes (as shown in Fig. 3.2) and potentially even more axes. Depending on the number of mirror axes, angle resolved transport non-reciprocity in the longitudinal and transverse channel follow this form:

$$\eta_{\parallel} = \eta_N \cos(N(\phi - \beta_N)) \quad (3.4)$$

$$\eta_{\perp} = \eta_N \sin(N(\phi - \beta_N)), \quad (3.5)$$

where  $N$  is the number of mirror axes and must be odd,  $\eta_N$  is the oscillation amplitude,  $\beta_N$  is the direction of the mirror axes. Notably, there are no analogous terms to the  $R_0$  and  $R_H$  offsets from the linear case, as the broken  $C_2$  symmetry does not permit an angularly constant contribution (i.e., a  $C_0$  component). Moreover, in the absence of additional constraints, the underlying electronic order can have coexisting or competing contributions, e.g., one with one mirror axis, another with three mirror axes. Therefore, the overall angle resolved transport non-reciprocity is really governed by this general form:

$$\eta_{\parallel} = \sum_N \eta_N \cos(N(\phi - \beta_N)) \quad (3.6)$$

$$\eta_{\perp} = \sum_N \eta_N \sin(N(\phi - \beta_N)), \quad (3.7)$$

where  $N$  must be odd integers. This functional form naturally captures both the rotational symmetry and the phase relationship between longitudinal and transverse non-reciprocity, while also accommodating possible contributions with different angular symmetries.



### 3.3 Angle-resolved non-reciprocity in TTG

Having established a phenomenological framework for angle-dependent non-reciprocity based on symmetry considerations, we now turn to experimental measurements of the longitudinal and transverse non-reciprocal signals in twisted trilayer graphene. In this project, we focus our measurement on a TTG sample with twist angle  $\theta = 1.45^\circ$ , the same sample used for the data shown in Fig. 3.1. The sample structure is essentially the same as shown in Fig. 2.6a, dually graphite gated and patterned into the sunflower geometry. In Fig. 3.3, we show examples of the angle resolved non-reciprocal transport probed using second harmonic measurement, examining our phenomenological framework.

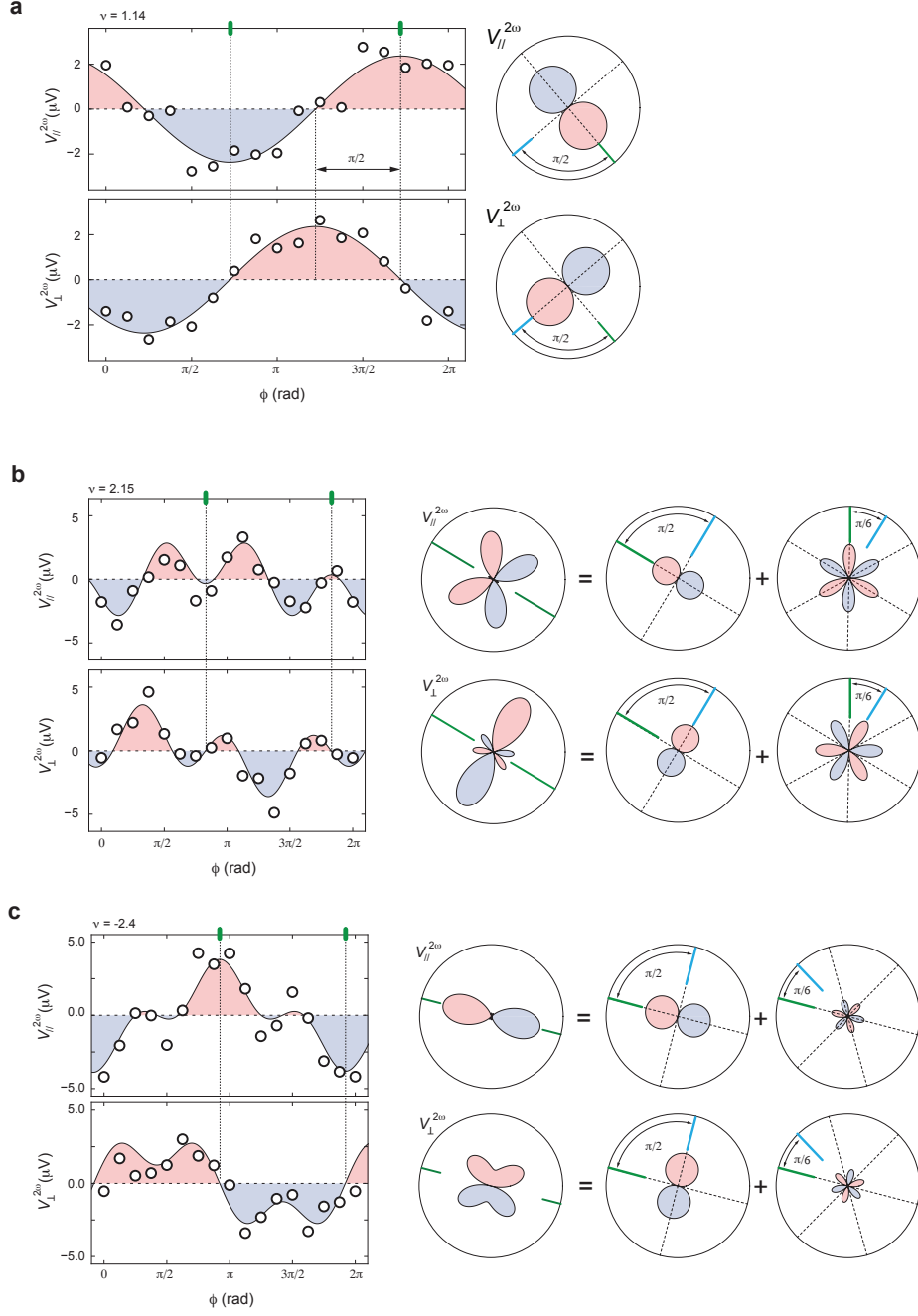
At  $\nu = 1.14$ , the sample exhibits a non-reciprocal angle dependence with a period of  $2\pi$ , consistent with the general functional form, Eqn. 3.6 - 3.7, with only  $N = 1$  contribution, indicating the underlying electronic order has one mirror axis pointing towards  $\phi \sim 315^\circ$  (see Fig. 3.3a). Moving forward, the response at  $\nu = 2.15$  and  $\nu = -2.40$  (shown in Fig. 3.3b-c) exhibit much complicated angle dependence, with contributions from both  $N = 1$  and  $N = 3$  terms, which is clear from the Fourier decomposition shown on the right. Although the general form, Eqn. 3.6 - 3.7 allows any odd integer values of  $N$  to contribute, with a large number of data set, we consistently observe only  $N = 1$  and  $N = 3$  components. Interestingly, it is also generally the case that the axis of the  $N = 1$  term aligns or anti-aligns with one of the mirror axes of the  $N = 3$  term. Thus, the general form of angle resolved non-reciprocal transport is empirically reduced to:

$$\eta_{\parallel} = \eta_1 \cos(\phi - \beta) + \eta_3 \cos(3(\phi - \beta)) \quad (3.8)$$

$$\eta_{\perp} = \eta_1 \sin(\phi - \beta) + \eta_3 \sin(3(\phi - \beta)), \quad (3.9)$$

where  $\beta$  is the direction of maximum one-fold contribution in the longitudinal channel,  $\eta_1$  and  $\eta_3$  are oscillation amplitude from the one-fold and three-fold contributions, respectively.

Notice how we have been using the general form of resistance non-reciprocity  $\eta_R$  to



**Figure 3.3: Angle dependence of second harmonic response in TTG.**

Angular dependence of  $V_{||}^{2\omega}$  (top) and  $V_{\perp}^{2\omega}$  (bottom) measured at (a)  $\nu = 1.14$ , (b)  $\nu = 2.15$ , and (c)  $\nu = -2.40$ , plotted in Cartesian-coordinates (left) and polar-coordinates (right). For (b-c), the angle dependence is further decomposed into one-fold and three-fold contributions, plotted in polar-coordinates. All data measured at  $T = 20$  mK and  $B = 0$ . Adapted from Ref. [76]

describe the angle dependence data of second harmonic voltage response. As discussed in section 3.1, when the second order term is the dominating nonlinear term in the  $I - V$  characteristic (evidenced by Fig. 3.1), transport non-reciprocity is related to second harmonic response by:

$$V^{2\omega} = \frac{1}{4}\eta_R I_0, \quad (3.10)$$

which holds true regardless of the direction of current flow. Therefore, transport non-reciprocity and second harmonic response should exhibit the same angle dependence. As demonstrated in Fig. 3.4, it is indeed the case: resistance non-reciprocity and second harmonic voltage response follow nearly identical angular dependence form, with a matching amplitude. As such, we will take  $\eta_R$  and  $V^{2\omega}$  as equivalent description of nonreciprocity. And when treating the angle dependence of second harmonic voltage responses, we simply substitute  $\eta$  with  $V$  in Eqn. 3.8 - 3.9:

$$V_{\parallel}^{2\omega} = V_1 \cos(\phi - \beta) + V_3 \cos(3(\phi - \beta)) \quad (3.11)$$

$$V_{\perp}^{2\omega} = V_1 \sin(\phi - \beta) + V_3 \sin(3(\phi - \beta)), \quad (3.12)$$

where  $\beta$  is the direction of maximum one-fold contribution in the longitudinal channel,  $V_1$  and  $V_3$  are oscillation amplitude from the one-fold and three-fold contributions, respectively.

By now, we have experimentally validated our reduced framework for TTG: using just three parameters,  $\beta$ ,  $\eta_1$ , and  $\eta_3$  (or equivalently  $V_1$  and  $V_3$ ), we are able to phenomenologically describe the transport non-reciprocity in a TTG sample. Remarkably, as we will show in the next section, this non-reciprocity is ubiquitous across the low temperature phase diagram of TTG systems. We now proceed to study its origin by examining its evolution across the phase diagram.

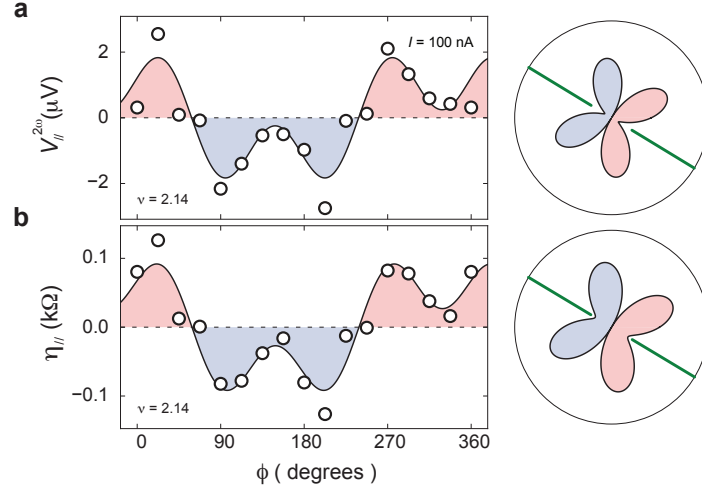


Figure 3.4: **Angle dependence of transport non-reciprocity and second harmonic response.**

Angular dependence of (a)  $V_{\parallel}^{2\omega}$  and (b)  $\eta_{\parallel}$  measured at  $\nu = 2.14$  plotted in Cartesian-coordinates (left) and polar-coordinates (right). The green lines highlight the direction of the mirror axes. The matching angle dependence form and amplitude illustrate the equivalence between  $\eta_R$  and  $V^{2\omega}$ .  $V_{\parallel}^{2\omega}$  data are measured with a.c. current  $I_{AC} = 100$  nA;  $\eta_{\parallel}$  data are measured with d.c. current  $I_{DC} = 100$  nA. All measurements are performed at  $T = 20$  mK and  $B = 0$ . Adapted from Ref. [76]

### 3.4 Angle-resolved non-reciprocity in TTG phase diagram

First, we highlight a few key features in the low temperature phase diagram of the TTG sample of interest. Shown in Fig. 3.5, this TTG sample exhibits two superconducting domes near  $\nu = \pm 2$ , isospin polarization induced insulator at integer fillings, as well as orbital ferromagnetism at  $\nu = 1$ ,  $\nu = 2$ , and  $\nu = 3$ , evidenced by the magnetic field and current induced hysteretic loops at these fillings (see Fig. 3.5c-d and Fig. 3.6).

Next, we demonstrate the ubiquity of non-reciprocity across the moiré band at low temperature. As shown in Fig. 3.7, prominent  $V_{\perp}^{2\omega}$  is observed at each filling, spanning the entire moiré band, at base temperature. The quick suppression of  $V_{\perp}^{2\omega}$  at around  $T = 5$  K suggests the non-reciprocity in TTG is likely due to a spontaneous symmetry breaking order. This claim is further supported by the observation of temperature and current

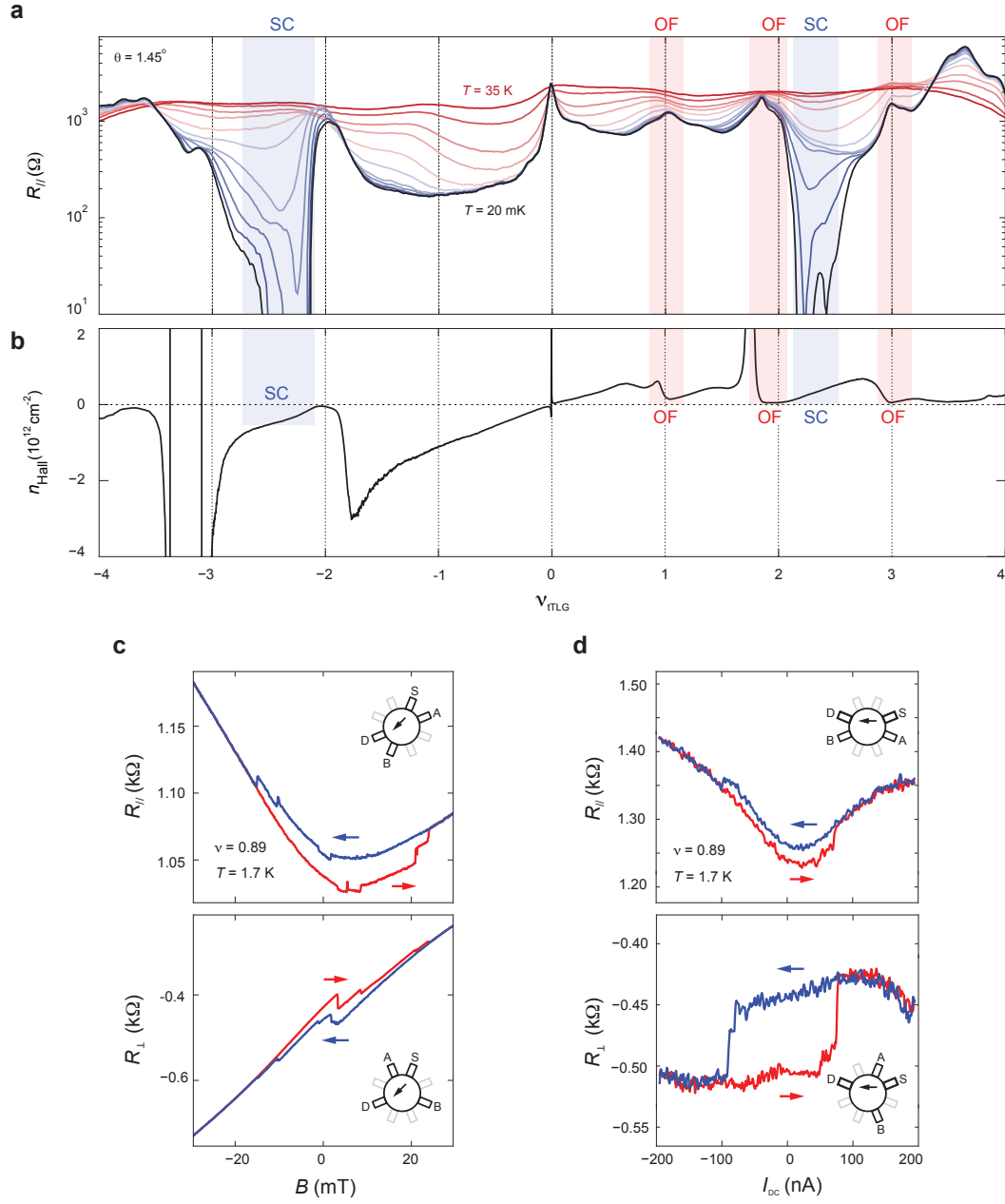
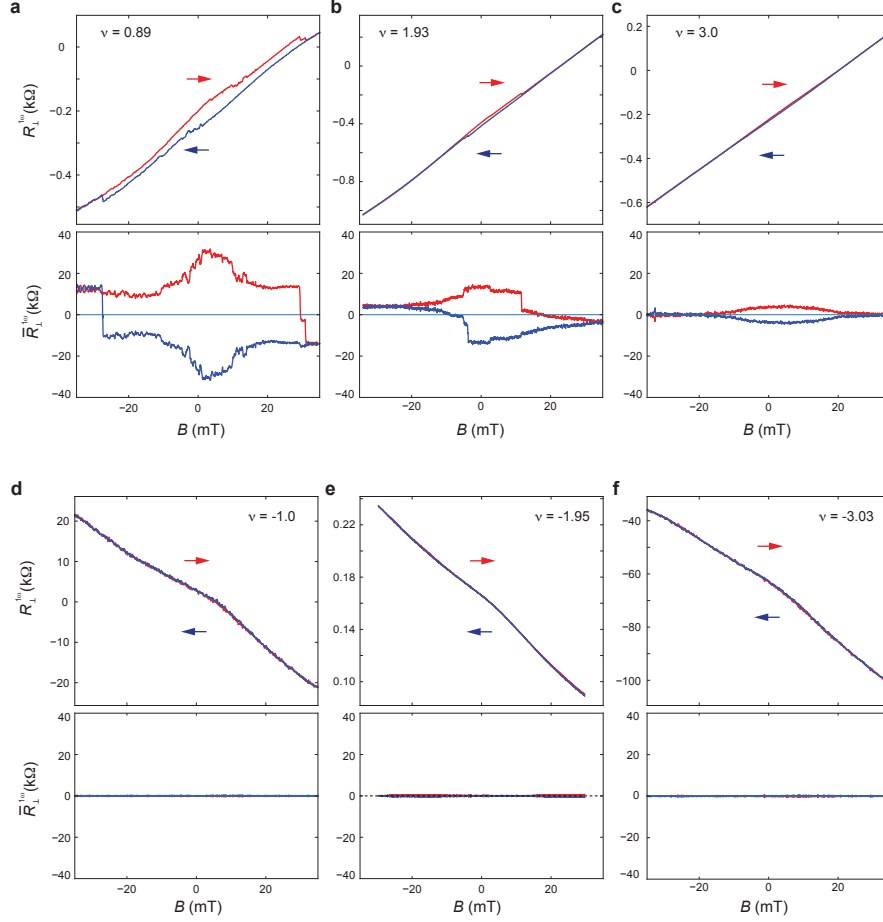


Figure 3.5: **Low temperature phase diagram of the TTG sample.**

(a) Linear longitudinal resistance  $R_{||}$  and (b) Hall density  $n_{\text{Hall}}$  as a function of moiré band filling  $\nu$ , measured at  $T = 20$  mK and  $I_{AC} = 5$  nA. The density regimes of the SC and orbital ferromagnetic order are marked with blue and red shades, respectively. (c) Magnetic field and (d) current induced hysteresis loops of Linear longitudinal resistance  $R_{||}$  (top) and linear transverse resistance  $R_{\perp}$  (bottom), measured at  $\nu = 0.89$  and  $T = 1.7$  K, evidencing the orbital ferromagnetism near  $\nu = 1$ . Adapted from Ref. [76]



**Figure 3.6: Magnetic field induced hysteresis loops.**

Linear transverse resistance  $R_{\perp}^{1\omega}$  (top) and renormalized linear transverse resistance  $\bar{R}_{\perp}^{1\omega}$  (bottom) as a function of perpendicular magnetic field, measured at (a)  $\nu = 0.89$ , (b)  $\nu = 1.93$ , (c)  $\nu = 3.0$ , (d)  $\nu = -1.0$ , (e)  $\nu = -1.95$ , and (f)  $\nu = -3.03$ .  $\bar{R}_{\perp}^{1\omega}$  is obtained by subtracting the common  $B$ -dependent background from  $R_{\perp}^{1\omega}$  from both sweeps. All measurements performed at  $T = 20$  mK and  $I_{AC} = 100$  nA. Adapted from Ref. [76]

induced hysteresis in angle resolved transport non-reciprocity, as shown in Fig. 3.8. In both cases, the angle dependence of transport non-reciprocity exhibits a rotation of the mirror axis  $\beta$ , as well as changes in the amplitudes  $V_1$  and  $V_3$ .

Furthermore,  $\beta$  as a function of  $\nu$  also demonstrates a new interesting cascade phenomenon. Fig. 3.9a plots  $\beta$  in the carrier density regime  $1.2 < \nu < 2.9$ . Remarkably,  $\beta$  preferentially plateaus at steps which are  $60^\circ$  apart, defining six characteristic directions, hinting at a possible connection to the crystallographic axes of the moiré lattice of the TTG sample. This observation further highlights the previous claim of non-reciprocity in

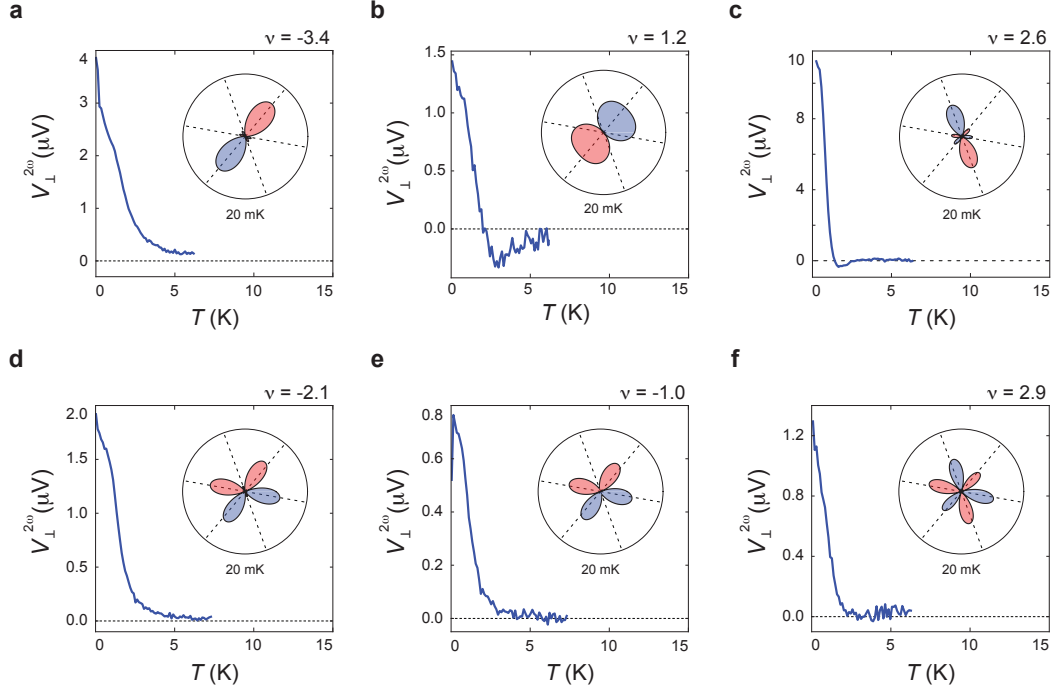
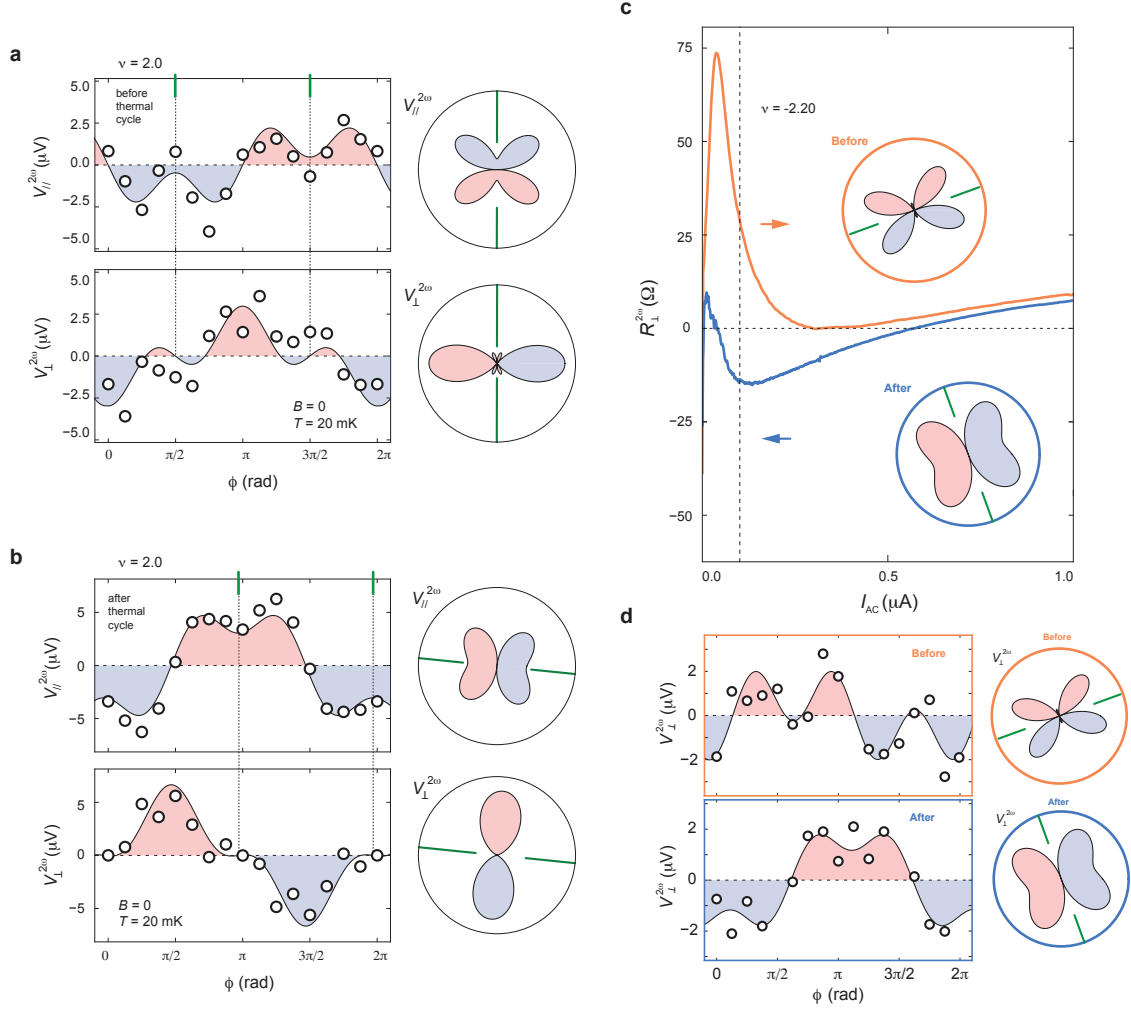


Figure 3.7: **Ubiquity of non-reciprocity across the moiré band.**

(a-f)  $V_{\perp}^{2\omega}$  as a function of  $T$ , measured at (a)  $\nu = -3.4$ , (b)  $\nu = 1.2$ , (c)  $\nu = 2.6$ , (d)  $\nu = -2.1$ , (e)  $\nu = -1.0$ , and (f)  $\nu = 2.9$ . Inset, the angle dependence of  $V_{\perp}^{2\omega}$  measured at  $T = 20$  mK at each density. All data measured with  $I_{AC} = 100$  nA and  $B = 0$ . Adapted from Ref. [76]

TTG being spontaneous, as uni-axial strain alone cannot produce such nicely plateaued rotation of the mirror axis that spanning the entire  $360^\circ$  of the plane.

To further pin down the microscopic mechanism behind non-reciprocity in TTG, we revisit the observed phenomena from a symmetry perspective. While non-reciprocity guarantees the breaking of  $C_2$  symmetry, time-reversal symmetry  $T$  is not necessarily broken. A combination of broken  $C_2$  and preserved  $T$  symmetry can be realized by scattering mechanisms like skew scattering or Berry curvature dipole induced nonlinear Hall effect [124–128]. However, this scenario is unlikely for two reasons: First, the temperature and current induced hysteretic rotation and the density induced evolution of  $\beta$  cannot be explained by skew scattering or Berry curvature dipole. Second, the absence of non-reciprocity for  $T > 5$  K is much lower than the onset temperature of scattering, which is usually around hundreds of Kelvins [125–128]. On the other hand,  $C_2$  and  $T$  can be simultaneously broken by the presence momentum polarization, where charge carriers



**Figure 3.8: Temperature and current induced hysteresis in angle resolved transport non-reciprocity.**

(a-b) Angle dependence of  $V_{\parallel}^{2\omega}$  (top) and  $V_{\perp}^{2\omega}$  (bottom) measured at  $\nu = 2.0$  (a) before and (b) after a thermal cycle up to  $T = 30$  K. (c)  $R_{\perp}^{2\omega} = V_{\perp}^{2\omega}/I$ , as a function of a.c. current amplitude  $I_{AC}$ , measured at  $\nu = -2.20$  A hysteresis loop emerges as current amplitude is swept up and down. (d) Angle dependence of  $V_{\perp}^{2\omega}$  measured at  $\nu = -2.20$  before (top) and after (bottom) the current sweep. All angle dependence data measured at  $I_{AC} = 100$  nA,  $T = 20$  mK, and  $B = 0$ . Adapted from Ref. [76]

occupy the corners of the Fermi surface in an imbalanced manner, under the influence of trigonal warping [25, 26, 33, 47, 70, 129, 130].

This picture can be further examined by comparing non-reciprocity and orbital ferromagnetism, which-in the case of TTG-entails both sublattice polarization and valley



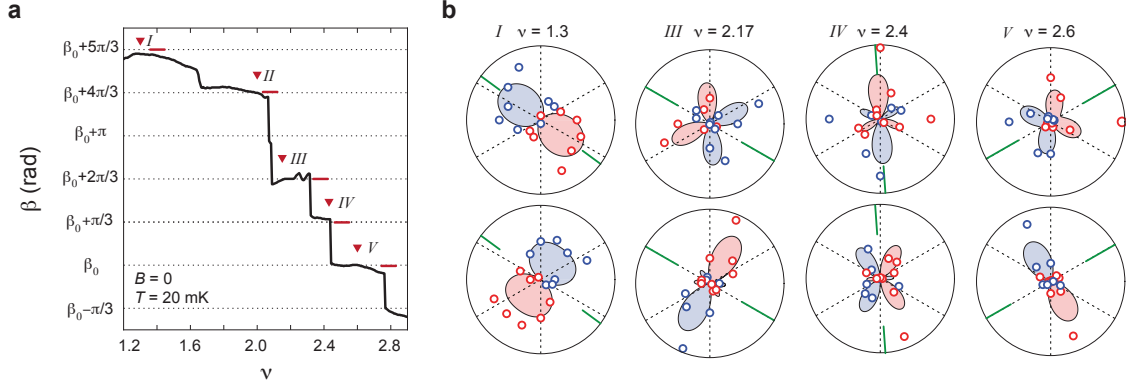


Figure 3.9: **Density induced rotation of mirror axis.**

(a)  $\beta$  as a function of  $\nu$ , extracted from angle resolved non-reciprocal transport data at each density, exhibiting 5 prominent plateaus, labeled as I to V. (b) Angle dependence of  $V_{\parallel}^{2\omega}$  (top) and  $V_{\perp}^{2\omega}$  (bottom) measured at each plateau, plotted in polar-coordinates. Data for plateau II is shown in Fig. 3.8a. All measurements performed at  $T = 20$  mK,  $I_{AC} = 100$  nA, and  $B = 0$ . Adapted from Ref. [76]

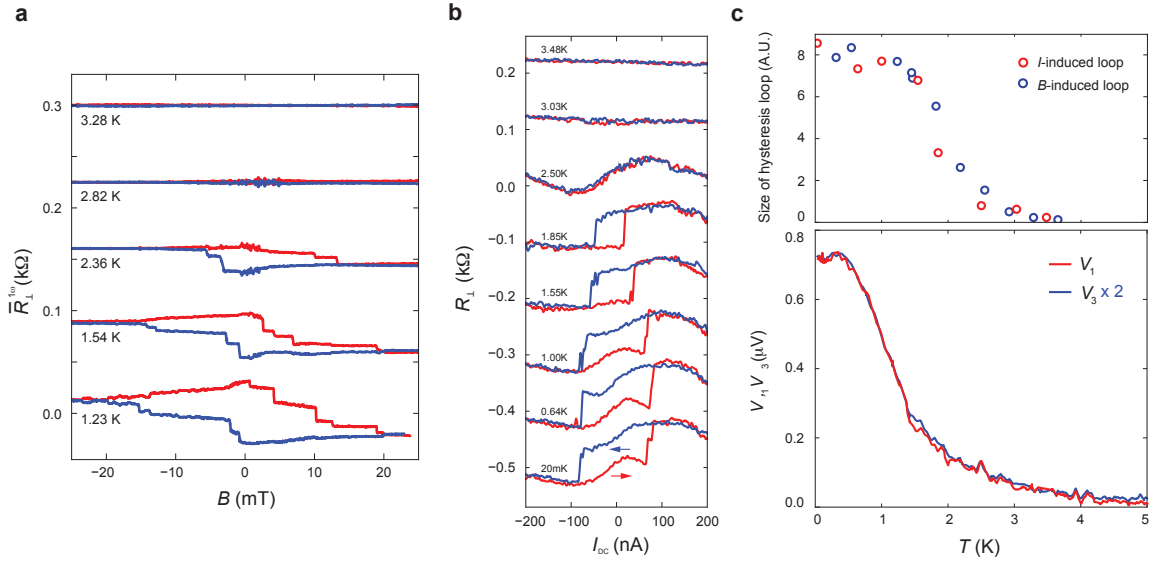


Figure 3.10: **Temperature dependence of orbital ferromagnetism and non-reciprocity.**

(a) Magnetic field and (b) current induced hysteresis loop of  $V_{\perp}^{2\omega}$  measured at different temperatures. (c) The area of hysteresis loops (top) and oscillation amplitudes of the angle dependence of nonreciprocity (bottom) as a function of temperature. All measurements performed at  $\nu = 0.9$ . Adapted from Ref. [76]

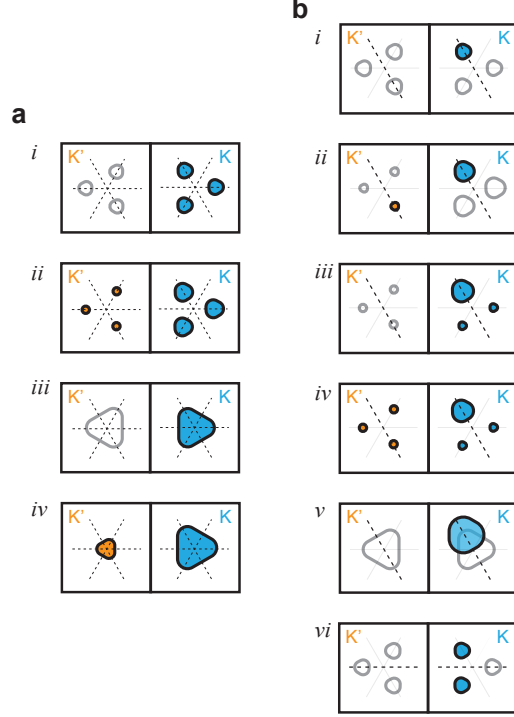


Figure 3.11: **Schematics of valley polarization and momentum polarization.**

Schematics of Fermi pocket occupation in the case of (a) valley polarization and (b) momentum polarization. Adapted from Ref. [76]

polarization. As shown in Fig. 3.10, the strength of orbital ferromagnetic order, gauged by the area of the hysteresis loop, sharply onsets with decreasing temperature. This behavior tracks the temperature dependence of  $V_1$  and  $V_3$ , the oscillation amplitude of the angle resolved transport non-reciprocity, underscoring the relationship between non-reciprocity, valley polarization, as well as orbital ferromagnetism in TTG. It also worth noting that, beyond the already discussed  $C_2$  and  $T$  symmetries, orbital ferromagnetism also requires the breaking of their product  $C_2T$ . We argue a broken  $C_2T$  is not necessary for non-reciprocity, as evidenced by it's universal appearance in the moiré band, even in the absence of orbital ferromagnetism.

We conclude our symmetry analysis by discussing the details of the angular symmetry of possible electronic orders. In the case of valley polarization or partial polarization, the Fermi surface contour would respect a  $C_3$  symmetry, as illustrated in Fig. 3.11a. The

observation of nonzero  $V_1$  suggests that the underlying electronic order must further break  $C_3$ , indicating the possibility of exchange-driven momentum polarization. Such instability in the momentum space breaks  $C_3$  symmetry by accumulating charge carriers into subset of Fermi pockets, as illustrated in Fig. 3.11b [131–135]. All together, our observation on angle resolved transport non-reciprocity in TTG points towards a *spontaneous momentum-space instability*, breaking  $C_2$  and  $T$  symmetry simultaneously.

### 3.5 Angle dependence of superconducting diode effect

At last, we report our observation on the angle dependence of the superconducting diode effect in TTG samples. Zero-field superconducting diode effect, manifested as an asymmetric  $I - V$  curve at zero magnetic field, where the superconducting critical current is different along opposite directions of d.c. current flow, was first observed in a small angle TTG sample (see Fig. 3.12) [136]. The observation of transport non-reciprocity in

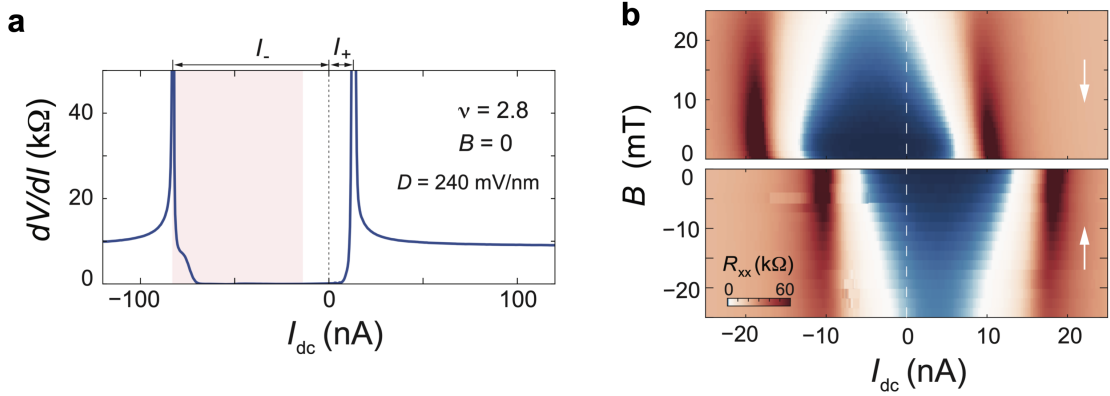


Figure 3.12: **Zero-field superconducting diode effect.**

(a) Differential resistance  $dV/dI$  as a function of d.c. current bias  $I_{dc}$  of the superconducting phase measured at  $B = 0$ .  $I_+$  and  $I_-$  marks the critical supercurrent with positive and negative d.c. current bias, respectively. The strong asymmetry of this  $I - V$  characteristics demonstrates the diodic behavior of the SC. (b)  $dV/dI$  as a function of  $I_{dc}$  and perpendicular magnetic field  $B$ . The arrows indicate the direction of magnetic field sweeping. By the sweeping the magnetic field from opposite directions, the superconducting diode direction can be controlled. The measurements were done in a TTG sample with twist angle  $\theta = 1.25^\circ$ . Adapted from Ref. [136]

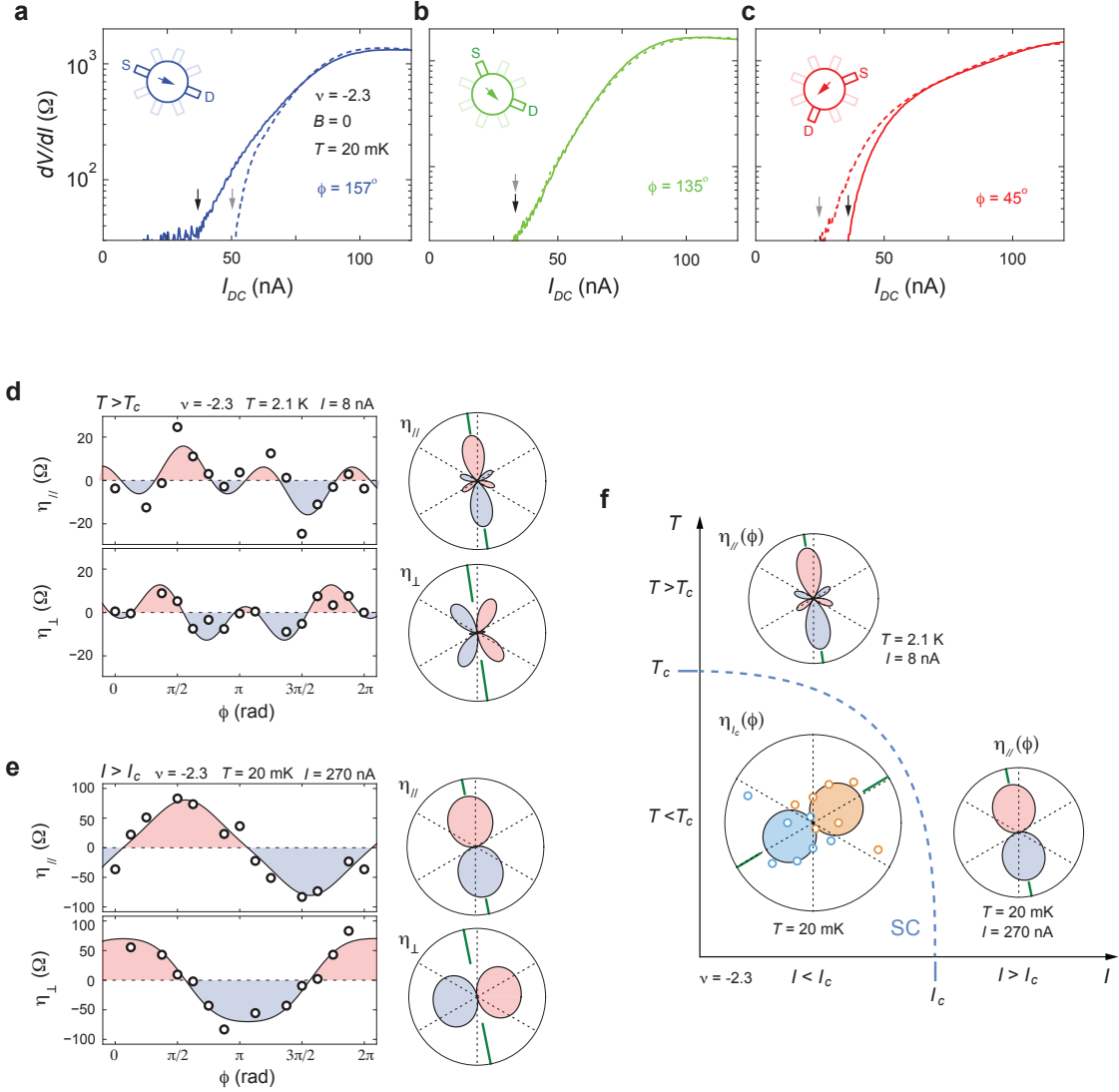


Figure 3.13: **Angle dependence of superconducting diode effect in TTG.**

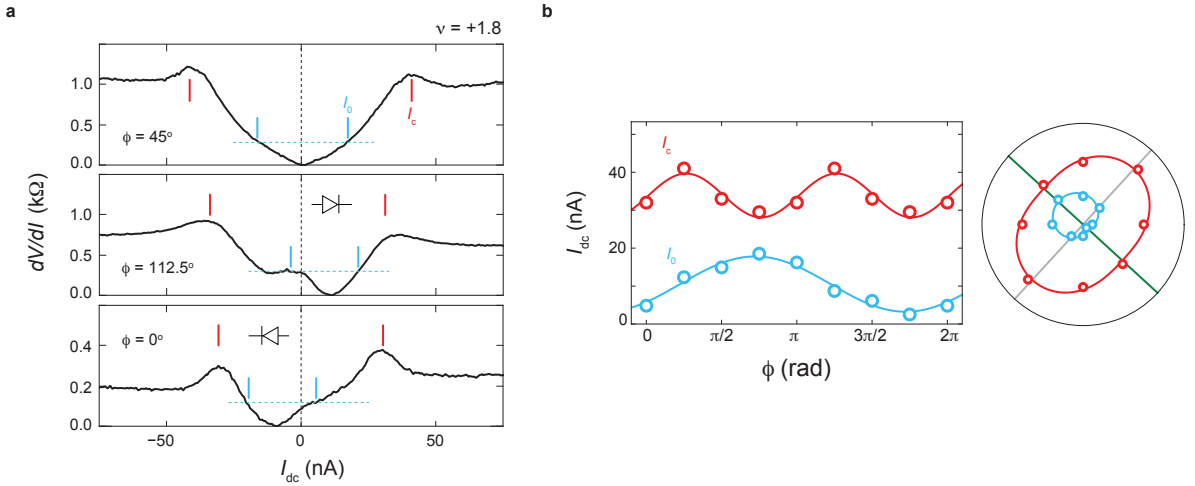
(a-c)  $I - V$  characteristics of the superconducting phase with current flowing along (a)  $\phi = 157^\circ$ , (b)  $135^\circ$ , and (c)  $45^\circ$ . Solid and dashed lines denote forward and reverse DC current biasing, respectively. Black and gray vertical arrows mark  $I_c$  in the forward and reverse direction, where  $dV/dI$  becomes larger than the noise floor. Defined in Eqn. 3.13,  $\eta_I$  is negative, zero, and positive from (a) to (c). Measurements are performed at  $B = 0$  and  $T = 20$  mK. (d-e) Angle dependence of resistance non-reciprocity in the longitudinal  $\eta_{||}$  (top) and transverse  $\eta_{\perp}$  (bottom) channels, measured in the metallic state at (d)  $T > T_c$  and (e)  $I > I_c$ . (f) Schematic  $I - T$  phase diagram of the superconducting phase. The blue dashed line marks the SC-normal state transition. All data measured at  $\nu = -2.3$ , the optimal doping of SC. Adapted from Ref. [76]

SC at zero magnetic field suggests a coexistence of inversion symmetry breaking order and the SC, hinting the possibility of an exotic, finite-momentum, Cooper pairing [137–141]. The non-reciprocity in the superconducting regime along each direction  $\phi$  is defined as:

$$\eta_{Ic}(\phi) = I_c(\phi) - I_c(\phi + \pi), \quad (3.13)$$

equivalent to the difference of supercurrents between forward and reverse bias in each direction [131, 136]. To minimize the potential influence of metallic state non-reciprocity at large current, we operationally define  $I_c$  as the supercurrent at which the differential resistance is above the noise floor (see Fig. 3.13).

As shown in Fig. 3.13, the superconducting diode effect exhibits a dominating one-fold symmetric contribution, similar to the high temperature or large current metallic state non-reciprocity angle dependence, suggesting the possibility of SC inheriting the  $C_2$ ,  $C_3$  and  $T$  breaking order from the metallic state.



**Figure 3.14: Angle dependence of superconducting diode effect in MATTG.**

(a)  $I - V$  characteristics of the superconducting phase with current flowing along  $\phi = 45^\circ$  (top),  $112.5^\circ$  (middle), and  $0^\circ$  (bottom), measured from a MATTG sample with  $\theta = 1.55^\circ$ . Red vertical lines mark the coherent critical supercurrent  $I_c$ , defined as the current with maximum  $dV/dI$ . Blue vertical lines mark the zero resistance critical supercurrent  $I_0$ , defined as the current when  $dV/dI$  reaches  $100\Omega$ . (b) Angle dependence of  $I_c$  and  $I_0$ . Measurements are performed at  $\nu = 1.8$ ,  $B = 0$  and  $T = 20$  mK. Reproduced from Ref. [77]

On a separate note, in a MATTG sample with  $\theta = 1.55^\circ$ , the mirror axis of the superconducting diode effect on the electron-doped side ( $\nu = +1.8$ ), extracted from the angle dependence of the onset current  $I_0$ , aligns with the direction of minimum coherent supercurrent  $I_c$ , as shown in Fig. 3.14. This observation suggests a potential underlying connection between non-reciprocity and the nematic order of superconductivity, an intriguing possibility that warrants further investigation.

# CHAPTER 4

## Nonlinear transport in Bernal bilayer graphene with sunflower geometry

### 4.1 Bernal bilayer graphene and nonlinear Hall effect

In the previous chapter, we demonstrated a phenomenological interpretation of the observed non-reciprocity in TTG, probed using angle resolved second harmonic transport with simple angular symmetry analysis. Encouraged by the results but constrained by the complex model for TTG, we also measured the angle resolved transport non-reciprocity in a Bernal bilayer graphene (BBG) sample [135]. Similar to the TTG case, we found significant second harmonic signals that follow the reduced phenomenological form Eqn. 3.8 - 3.9. Upon extracting  $V_1$ ,  $V_3$  and  $\beta$ , we observe mirror axis rotation plateauing at  $60^\circ$  steps as functions of carrier density  $n$ , displacement field  $D$ , as well as perpendicular magnetic field  $B$ . At these transitions, we also observe a systematic evolution of  $V_1$  upon the rotational transition of  $\beta$  (see Fig. 4.1 and 4.2). These findings point towards a spontaneous breaking of  $C_2$  and  $T$  symmetry in BBG, in accordance with various possible scenarios [132–134]. While our approach is effective in identifying broken symmetries, the lack of a thorough understanding of the nonlinear transport leaves open questions about

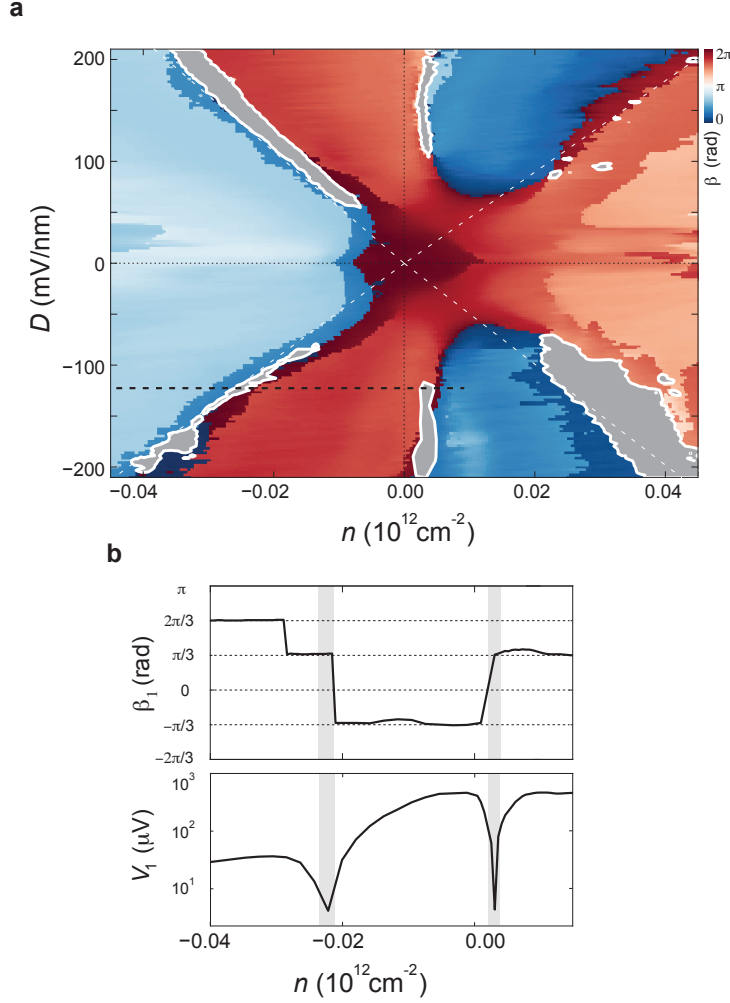


Figure 4.1:  $\beta$  as a function of  $n$  and  $D$  in BBG.

(a)  $\beta$  as a function of carrier density  $n$  and electric field  $D$ . Gray color indicates regimes with vanishing  $V_1$ . (b)  $\beta$  (top) and  $V_1$  (bottom) as a function of  $n$  measured at  $D = -150 \text{ mV/nm}$ , marked as black dash line in panel (c).  $V_1$  vanishes at the rotational transition of  $\beta$ . Adapted from Ref. [135]

the underlying microscopic mechanisms.

Beyond understanding what we have observed in TTG, another motivation of studying nonlinear transport lies in the concept of nonlinear Hall effect. In the linear case, Hall effect is a non-dissipative transport phenomenon in which the off-diagonal term,  $\sigma_H$ , in the  $2 \times 2$  conductivity matrix (in 2D) generates a transverse current perpendicular to the applied electric field. Similarly, nonlinear Hall effect describes the non-dissipative



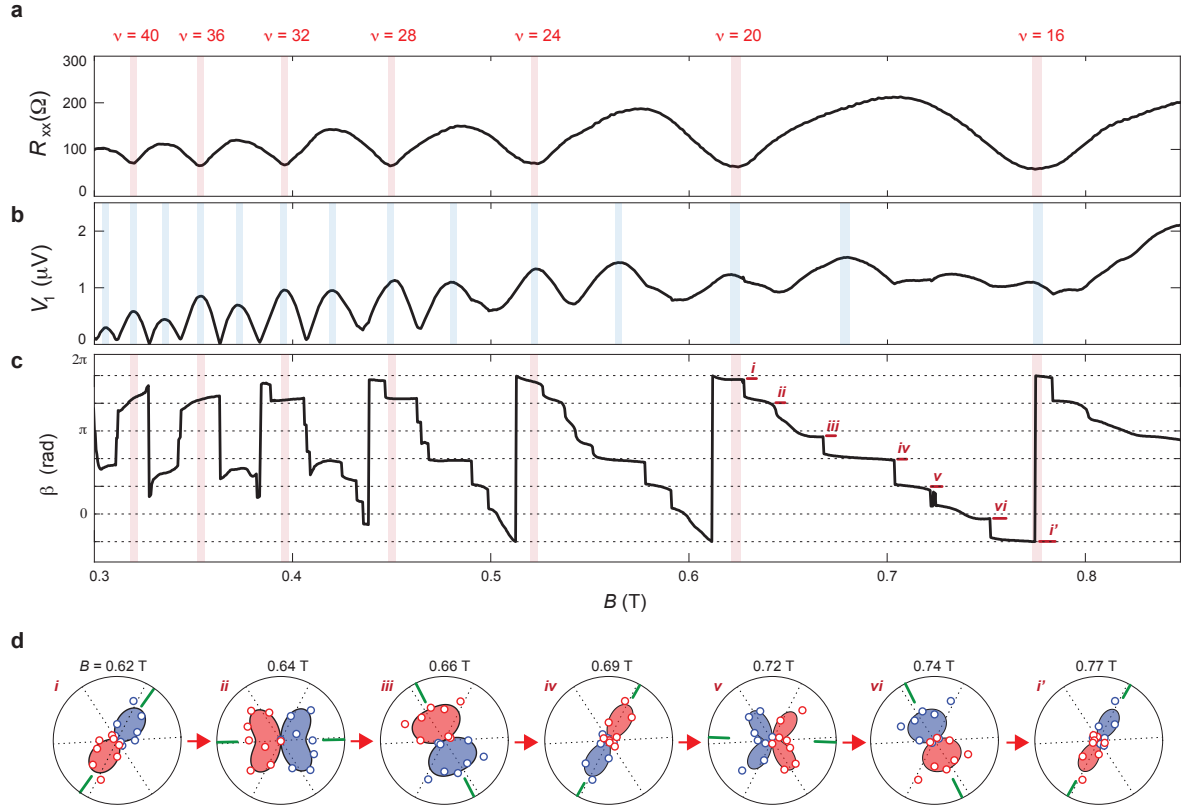


Figure 4.2:  $\beta$  as a function of  $B$  in BBG.

(a) Longitudinal resistance  $R_{\parallel}$ , (b)  $V_1$ , and (c)  $\beta$  as a function of an out-of-plane magnetic field  $B$  measured at  $n = -0.3 \times 10^{12} \text{ cm}^{-2}$  and  $D = 0$ . Red vertical stripes mark minima in  $R_{\parallel}$ . Blue vertical stripes mark maxima in  $V_1$ . (d) The angular dependence of  $V_{\parallel}^{2\omega}$  plotted polar-coordinated plots at plateaus in (c), marked with panel number  $i$  through  $iv$ . Interestingly,  $V_1$  exhibits modulations (evidenced by maxima) at twice the rate of  $R_{xx}$  (evidenced by minima) in  $B$ .  $\beta$  exhibits transitions (evidenced by steps) at much higher rate than  $R_{xx}$  in  $B$ . Adapted from Ref. [135]

transport phenomenon at the second order of the applied electric field, governed by the rank 3 nonlinear conductivity tensor,  $\tilde{\sigma}_{\alpha\mu\nu}$ . It challenges our understanding of the microscopic mechanism on dissipationless transport as it can, under the influence of Berry curvature dipole, preserve  $T$  symmetry, which is strictly required to be broken in the linear case [124, 142].

Despite the promising scientific prospect, a systematic identification of nonlinear Hall effect is still generally elusive before this project. Many existing studies take a nonzero second harmonic transverse signal  $V_{\perp}^{2\omega}$ , sometimes accompanied by a vanishing second

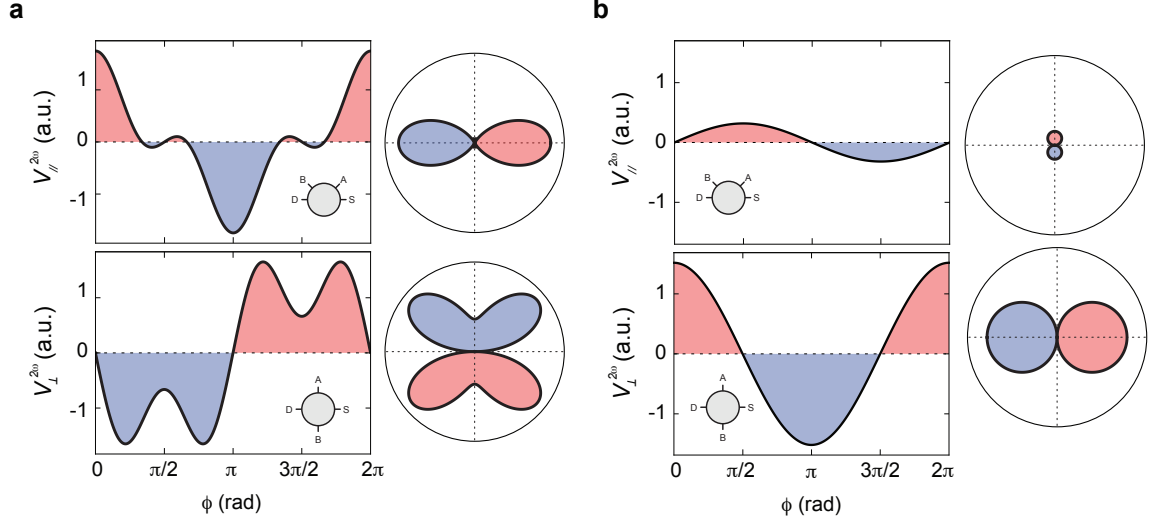


Figure 4.3: **Nonlinear transverse signal vs. nonlinear Hall effect.**

(a-b) Expected angle resolved nonlinear transport signal in the longitudinal (top) and transverse (bottom) channels. Plotted for the scenario with (a)  $\tilde{\sigma}_{xxx}$  as the only nonzero tensor component and (b) nonlinear Hall component as the only nonzero tensor component. Adapted from Ref. [142]

harmonic longitudinal voltage  $V_{||}^{2\omega}$ , as the signature of nonlinear Hall effect [125–127, 143, 144]. However, as we show below, these two behaviors alone, while providing evidence of a nonzero nonlinear conductance, are incomplete arguments for demonstrating nonlinear Hall effect.

In Fig. 4.3a, we show the expected angle resolved nonlinear transport response for a second order conductivity tensor with  $\tilde{\sigma}_{xxx}$  as the only nonzero tensor component. In this scenario, the nonlinear conductivity tensor has only symmetric components, generating only dissipative signals [142]. However, as clearly shown in the figure, sizable  $V_{\perp}^{2\omega}$  and vanishing  $V_{||}^{2\omega}$  can be observed, depending on the azimuth direction of the measurement. Conversely, Fig. 4.3b illustrates the possibilities of observing a vanishing  $V_{\perp}^{2\omega}$  signal, along the nodal directions of  $V_{\perp}^{2\omega}$  oscillation (e.g.,  $\phi = \pi/2$  and  $3\pi/2$  in the example), even when nonlinear Hall conductivity is the only nonzero component in the tensor.

This intriguing observation can be understood with a comparison to the linear transport

with orthorhombic anisotropy. As illustrated previously in Fig. 2.4c, anisotropy can generate linear transverse signals, while the linear conductivity matrix has no Hall component. If the sample is probed along a random azimuth direction, a nonzero  $V_{\perp}$  signal can appear. This feature is generally not an experimental issue as the anisotropy induced nonzero  $V_{\perp}$  signal, respecting  $C_2$  symmetry, can be removed upon geometric anti-symmetrization, which is a common practice for reducing mixing of the longitudinal signals due to misaligned contacts. Since nonlinear transport breaks  $C_2$  symmetry, it guarantees anisotropy. Thus, in a hand wavy manner, it is expected that a nonzero  $V_{\perp}^{2\omega}$  would show up with any general nonlinear conductivity tensor. But this anti-symmetrization method, deeply rooted in the principles from Onsager reciprocal relations, is unfortunately no longer applicable due to the non-reciprocal nature of nonlinear transport [145, 146].

## 4.2 Nonlinear transport in theory

Motivated by these challenges, we now construct a more complete theoretical framework of nonlinear electronic transport in the sunflower geometry. Similar to what we have discussed in section 2.3, we start with Ohm's law with quadratic corrections, using Einstein summation notation:

$$\mathbf{j}_{\alpha} = \sigma_{\alpha\mu} E_{\mu} + \tilde{\sigma}_{\alpha\mu\nu} E_{\mu} E_{\nu}, \quad (4.1)$$

where  $\alpha, \mu, \nu$  are spatial indices,  $E$  is the electric field,  $\sigma$  and  $\tilde{\sigma}$  are linear and nonlinear conductivity matrix and tensor, respectively. In a two dimensional system, in DC limit,  $\sigma$  and  $\tilde{\sigma}$  generally have 4 and 6 independent components, respectively. As we can show later, while the two “purely” symmetric components of  $\tilde{\sigma}$ , e.g.  $\tilde{\sigma}_{xxx}$  and  $\tilde{\sigma}_{yyy}$  contain information about nonlinear dissipative transport only, the other four components, with two of the spatial indices unequal, correspond to an intricate intertwinement between dissipative and non-dissipative nonlinear transport signals.

Now rewriting Eqn. 4.1 in complex notations, we obtain:

$$j_x + ij_y = \Xi_+^{(1)}(E_x + iE_y) + \Xi_-^{(1)}(E_x - iE_y) + \Xi_-^{(2)}(E_x - iE_y)^2 + \Xi_+^{(2)}(E_x + iE_y)^2 + \Xi_0^{(2)}(E_x^2 + E_y^2), \quad (4.2)$$

where parameters  $\Xi_b^{(a)}$  each is a complex number, thus described by a magnitude and a phase in the complex plane. In the linear case,  $\Xi_{+,-}^{(1)}$  together represent the general linear conductivity matrix. In other words, the two complex parameters  $\Xi_{+,-}^{(1)}$  describe the linear transport behavior equivalently as the 4 real parameters  $\sigma_{xx}$ ,  $\sigma_{yy}$ ,  $\sigma_H$ , and  $\alpha$ . Similarly, in the nonlinear case, three complex parameters  $\Xi_{+,0,-}^{(2)}$  represent the general nonlinear conductivity tensor equivalently as the 6 real parameters  $\tilde{\sigma}_{\alpha\mu\nu}$ , but in a more complicated fashion [142]:

$$\tilde{\sigma}_{xxx} = \text{Re}(\Xi_-^{(2)}) + \text{Re}(\Xi_+^{(2)}) + \text{Re}(\Xi_0^{(2)}) \quad (4.3)$$

$$\tilde{\sigma}_{xxy} = \text{Im}(\Xi_-^{(2)}) - \text{Im}(\Xi_+^{(2)}) \quad (4.4)$$

$$\tilde{\sigma}_{xyy} = -\text{Re}(\Xi_-^{(2)}) - \text{Re}(\Xi_+^{(2)}) + \text{Re}(\Xi_0^{(2)}) \quad (4.5)$$

$$\tilde{\sigma}_{yxx} = \text{Im}(\Xi_-^{(2)}) + \text{Im}(\Xi_+^{(2)}) + \text{Im}(\Xi_0^{(2)}) \quad (4.6)$$

$$\tilde{\sigma}_{yyx} = -\text{Re}(\Xi_-^{(2)}) + \text{Re}(\Xi_+^{(2)}) \quad (4.7)$$

$$\tilde{\sigma}_{yyy} = -\text{Im}(\Xi_-^{(2)}) - \text{Im}(\Xi_+^{(2)}) + \text{Im}(\Xi_0^{(2)}). \quad (4.8)$$

With such rewriting, we now convert the nonlinear conductivity from the Cartesian description into three bases:  $(E_x - iE_y)^2$ ,  $(E_x + iE_y)^2$ , and  $(E_x^2 + E_y^2)$ , each associated with an amplitude and a phase, represented by a complex coefficient  $\Xi^{(2)}$ .

Again with some very serious theoretical effort, one can analytically solve for the expected potential distribution over the disk, with arbitrarily chosen source-drain locations on the circumference, from each basis with an arbitrary complex coefficient [142]. It is worthwhile to note that we also assume similar boundary conditions: uniform and

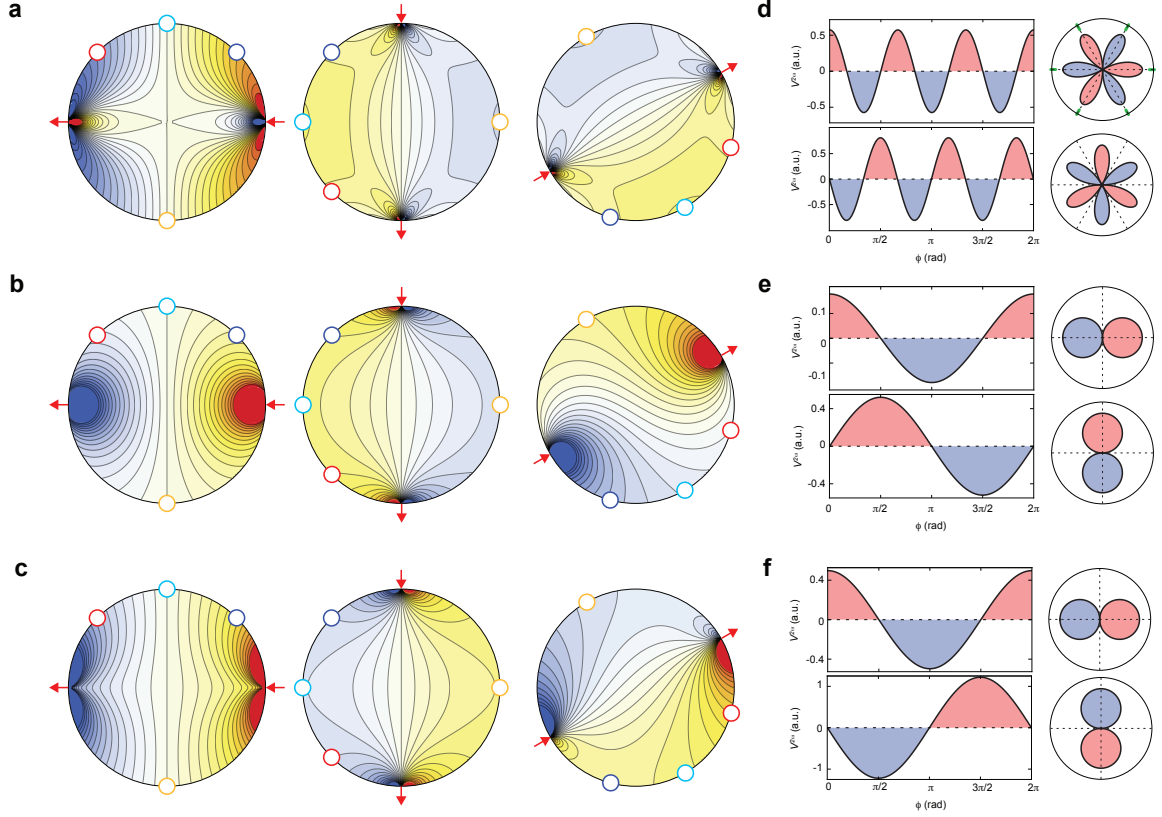


Figure 4.4: **Basis functions of nonlinear electrical potential.**

(a-c) Expected nonlinear electrical potential from the (a)  $(E_x - iE_y)^2$ , (b)  $(E_x + iE_y)^2$ , and (c)  $(E_x^2 + E_y^2)$  basis, with current source located at  $\phi = 0$  (left),  $\phi = 90^\circ$  (middle), and  $\phi = 210^\circ$  (right). Red arrows denote the location of current source and drain. Blue-red and cyan-orange open dot pairs denote the contact location for longitudinal and transverse channels, respectively. (d-f) Expected angle dependence of the nonlinear voltage response  $V_{\parallel}^{2\omega}$  (top) and  $V_{\perp}^{2\omega}$  (bottom) of each basis, illustrating different rotational symmetries and chiralities. Adapted from Ref. [142]

local conductivity matrix and tensor within the disk and no conduction outside the disk. Avoiding severe nonlinearities appearing in the nonlinear Ohm's law, we constrain ourselves at the perturbative second-order regime, where the only nonlinear contribution is the second order term and it is small compared to the linear response. For simplicity, we have also assumed an isotropic linear conductivity matrix without Hall coefficient, e.g.,  $\sigma_{xx} = \sigma_{yy}$  and  $\sigma_H = 0$ , while allowing  $\tilde{\sigma}$  to be completely general. Logically equivalent derivation can be done analytically for anisotropic linear conductivity matrix too, as demonstrated in the supplementary of Ref. [142].

Conveniently, the total nonlinear potential found to be the linear combination of the three contributions:

$$\Phi^{(2)} = \Phi^{(2-)} + \Phi^{(2+)} + \Phi^{(20)}, \quad (4.9)$$

where  $\Phi^{(2-)}$ ,  $\Phi^{(2+)}$ , and  $\Phi^{(20)}$  denotes the potential yielded from  $(E_x - iE_y)^2$ ,  $(E_x + iE_y)^2$ , and  $(E_x^2 + E_y^2)$  contributions, linearly proportional to the three complex coefficients  $\Xi_-^{(2)}$ ,  $\Xi_+^{(2)}$ , and  $\Xi_0^{(2)}$ , respectively. Remarkably, the individual potential contributions behave distinctively regarding rotational symmetry and chirality:  $\Phi^{(2-)}$  respects  $C_3$ , whereas  $\Phi^{(2+)}$  and  $\Phi^{(20)}$  preserves  $C_1$ . As illustrated by the yielded angle dependence of the longitudinal and transverse responses,  $\Phi^{(2+)}$  and  $\Phi^{(20)}$  can be further distinguished by the opposite phase shift between the two channels.

Having solved the expected nonlinear potential for arbitrary  $\Xi^{(2)}$ , we are now equipped with the ability to predict the second harmonic voltage response for any given measurement configuration by simply computing  $\Phi^{(2)}(\mathbf{r}_\mathbf{A}) - \Phi^{(2)}(\mathbf{r}_\mathbf{B})$ , where  $\mathbf{r}_\mathbf{A}$  and  $\mathbf{r}_\mathbf{B}$  denote the locations of the voltage contacts. In a reverse logic, we can perform multiple four-terminal measurements, with arbitrarily chosen contacts, to extract out the complex parameters,  $\Xi^{(2)}$ . As such, along with section 2.3, we have established a complete framework for extracting the conductivity tensor up to second order in current.

### 4.3 Extracting nonlinear conductivity tensor in BBG

In this section, we demonstrate the experimental extraction of nonlinear conductivity tensor in a BBG sample. The choice of BBG is made with two considerations: First, to test out a new experimental method, we generally prefer the system to be simple and clean. As the natural, stable crystalline structure of bilayer graphene, BBG has a much simpler band structure and is usually more uniform than twisted graphene systems [43, 62, 147–150]. Second, having second harmonic response in mind, we need a system that would potentially break  $C_2$  symmetry. As we demonstrated in section 4.1, second

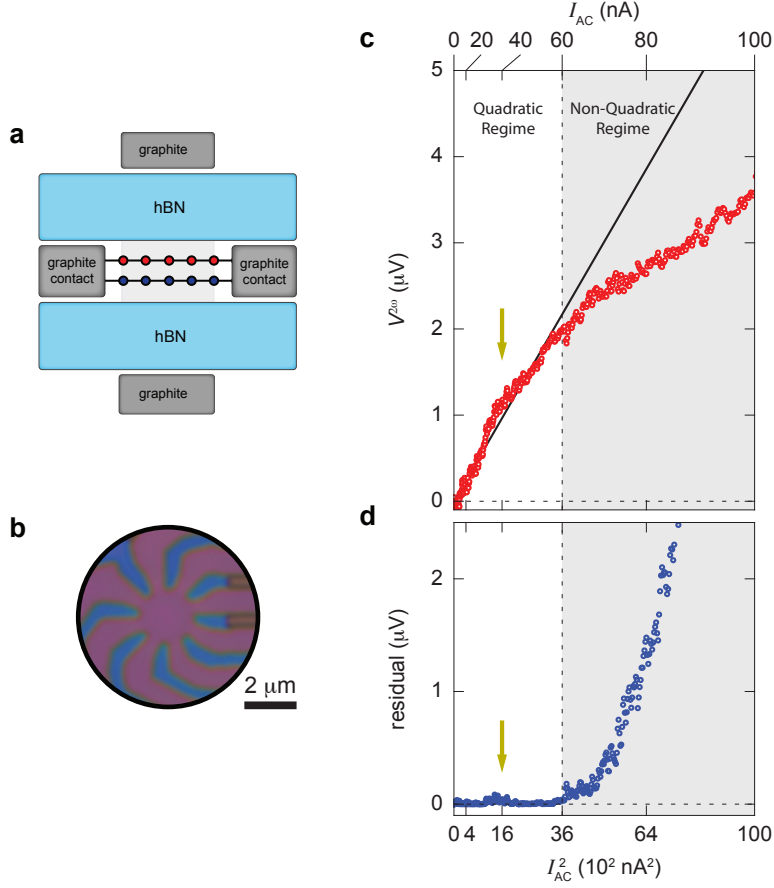


Figure 4.5: **Quadratic regime of BBG.**

(a) Schematic of the BBG sample structure. (b) An optical image of the sample (c)  $V^{2\omega}$  and (d) its residual from a quadratic fit ( $f = aI^2$ ), as a function of the square of AC current amplitude  $I^2$ . Deviation from the quadratic fit, evidenced by the residual onset at  $I = 60$  nA operationally makes the cutoff between quadratic and non-quadratic regime. Measurement performed at  $T = 20$  mK and  $B = 0$ . Adapted from Ref. [142]

harmonic response has been observed in BBG. Inversion symmetry in BBG can be broken by applying a displacement field.

As before, the sample is dually gated and patterned into a sunflower geometry (see Fig. 4.5a-b). Firstly, the validity of our assumptions is examined. In Fig. 4.5c-d,  $V^{2\omega}$  is quadratic to a.c. current amplitude, for  $I_{AC} < 60$  nA; thus we limit our measurement at  $I_{AC} = 40$  nA. In Fig. 4.2a-b, amplitude of second harmonic response, gauged by  $V_1$ ,

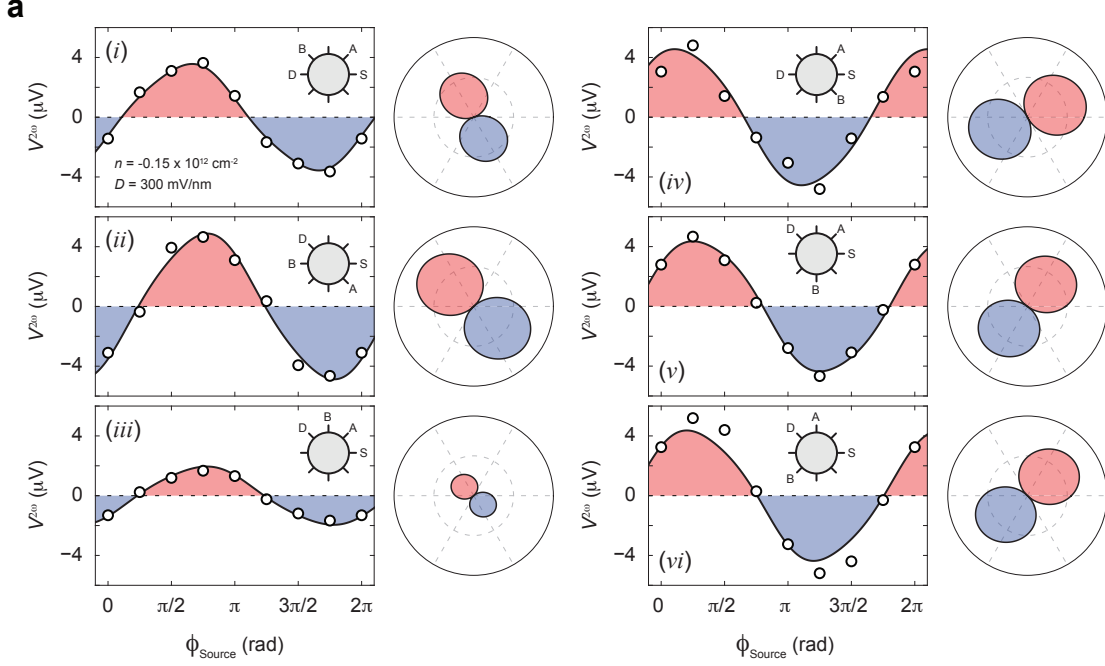


Figure 4.6: **Extracting nonlinear conductivity tensor in BBG.**

(a) Second harmonic voltage measurement (black open dots) as a function of current source location, measured with 6 different configurations, with 8 current source location for each, yield a total of 48 data points for the same state. Insets show the schematic of the configurations for source located at  $\phi = 0^\circ$ . By rotating all four current and voltage contacts by 1 lead at a time, we measure all the other angular entries. Black lines denote the theoretically expected signal for each configuration, all predicted from the same nonlinear conductivity tensor. The remarkable agreement between 48 independent measurements and their theoretical fit demonstrates the extraction of nonlinear conductivity tensor, as well as the uniformity of the sample and its conductivity in general. Measurement performed at  $n = -0.15 \times 10^{12} \text{ cm}^{-2}$ ,  $D = 300 \text{ mV/nm}$ ,  $I_{AC} = 40 \text{ nA}$ ,  $T = 20 \text{ mK}$  and  $B = 0$ . Adapted from Ref. [142]

is always orders of magnitude smaller than the linear response  $R_{xx}$ ; hence the nonlinear contribution is always perturbative. The lack of anisotropy and absence of Hall component in the linear conductivity matrix can be confirmed using the model described in section 2.3.

Now, we extract the nonlinear conductivity tensor by measuring 6 different configurations, 8 angles each, and then fit all 48 data against the predicted second harmonic voltage response for each configuration, specified by the same set of  $\Xi^{(2)}$  (see Fig. 4.6). The remarkable agreement between the experiment and the theoretical fit demonstrates



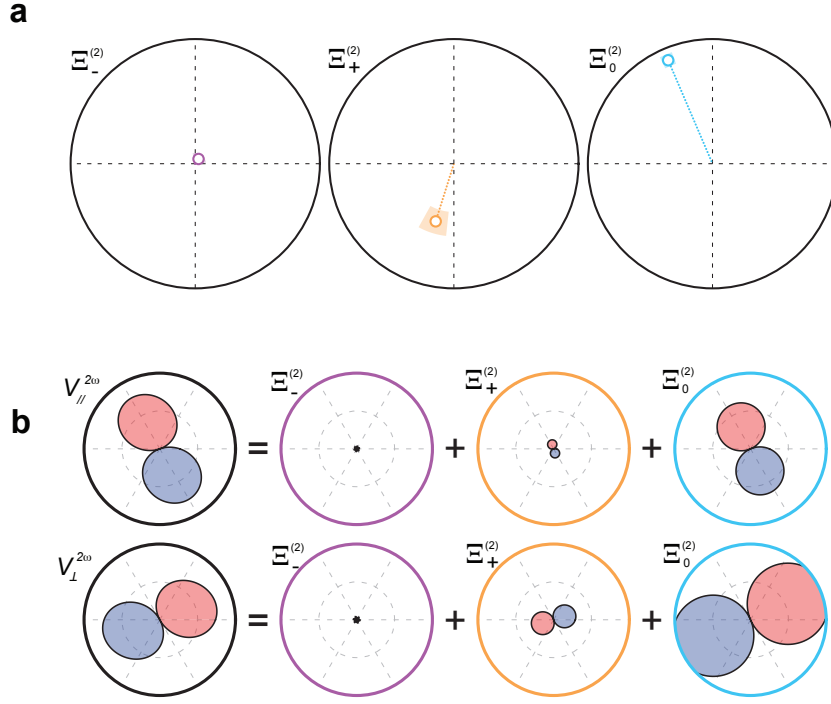


Figure 4.7: **Nonlinear signal decomposition into three bases.**

(a) The complex parameters,  $\Xi^{(2)}$  from the extraction in Fig. 4.6, plotted in the complex plane, where horizontal axis is real and vertical axis is imaginary. (b) The angle dependence of  $V_{||}^{2\omega}$  and  $V_{\perp}^{2\omega}$  at the same state, decomposed into three contributions, each from one basis. Adapted from Ref. [142]

the successful extraction of the nonlinear conductivity tensor. Note that it is not necessary to perform measurement in an angle complete manner, e.g. always measuring all 8 angles for each configuration. But it is generally helpful in terms of avoiding systematic errors by using contacts evenly.

To illustrate the fitting result, we further decompose our measured responses back to the contributions from three bases. As shown in Fig. 4.7a, the coefficient for the three fold term,  $\Xi_{-}^{(2)}$  is vanishingly small.  $\Xi_{+}^{(2)}$  is about half in magnitude and mirror reflected from  $\Xi_{0}^{(2)}$  against the real axis in orientation, yielding signals that are basically aligned in  $V_{||}^{2\omega}$  and anti-aligned in  $V_{\perp}^{2\omega}$ . Together, these three contributions surprisingly give rise to an overall measurement result that is one fold symmetric, and has roughly similar amplitude in  $V_{||}^{2\omega}$  and  $V_{\perp}^{2\omega}$ , in accordance to our reduced phenomenological form Eqn. 3.8 - 3.9. Here

it is worth mentioning that, albeit the amazing agreement between the extraction in this example and our reduced phenomenological form, our new complete framework allows for the true general  $\tilde{\sigma}$ , where the three fold term and the two one fold term are not necessarily aligned or anti-aligned with each other.

The power of extracting the complete nonlinear conductivity tensor also enables us to distinguish the contributions of nonlinear signal based on their dissipation. By demanding a total current density  $\mathbf{j}$  perpendicular to  $\mathbf{E}$  for any in-plane direction of  $\mathbf{E}$ , we find the general form of non-dissipative current:

$$\mathbf{j}_H = \sigma_H \mathbf{E} \times \hat{\mathbf{z}} + \hat{\mathbf{z}} \times \mathbf{E}(\mathcal{C} \cdot \mathbf{E}), \quad (4.10)$$

where the first term is the linear Hall current, determined by  $\sigma_H$ , and the second term is the nonlinear Hall current, governed by a real vector  $\mathcal{C}$ , which can be related back to the complex parameters by [142]:

$$i\mathcal{C}_x + \mathcal{C}_y = \Xi_+^{(2)} - \Xi_0^{(2)*}. \quad (4.11)$$

Therefore, the nonlinear Hall current  $\mathbf{j}_H^{(2)} = \hat{\mathbf{z}} \times \mathbf{E}(\mathcal{C} \cdot \mathbf{E})$ , having no contribution from  $\Xi_-^{(2)}$ , prohibits all rotational symmetry but  $C_1$ . In the same coordinate-free form, the dissipative part of the nonlinear current is given by:

$$\mathbf{j}_{\parallel}^{(2)} = \mathbf{E}(\mathcal{B} \cdot \mathbf{E}) \quad (4.12)$$

$$\mathbf{j}_{\times}^{(2)} = \mathcal{A}((\mathcal{A} \cdot \mathbf{E})^2 - (\mathcal{A} \times \mathbf{E})^2) + 2\mathcal{A} \times (\mathcal{A} \times \mathbf{E})(\mathcal{A} \cdot \mathbf{E}), \quad (4.13)$$

where  $\mathbf{j}_{\parallel}^{(2)}$  and  $\mathbf{j}_{\times}^{(2)}$  describe the one-fold and three-fold symmetric contribution to the dissipative current, respectively. To be complete, real vectors  $\mathcal{A}$  and  $\mathcal{B}$  are related to the

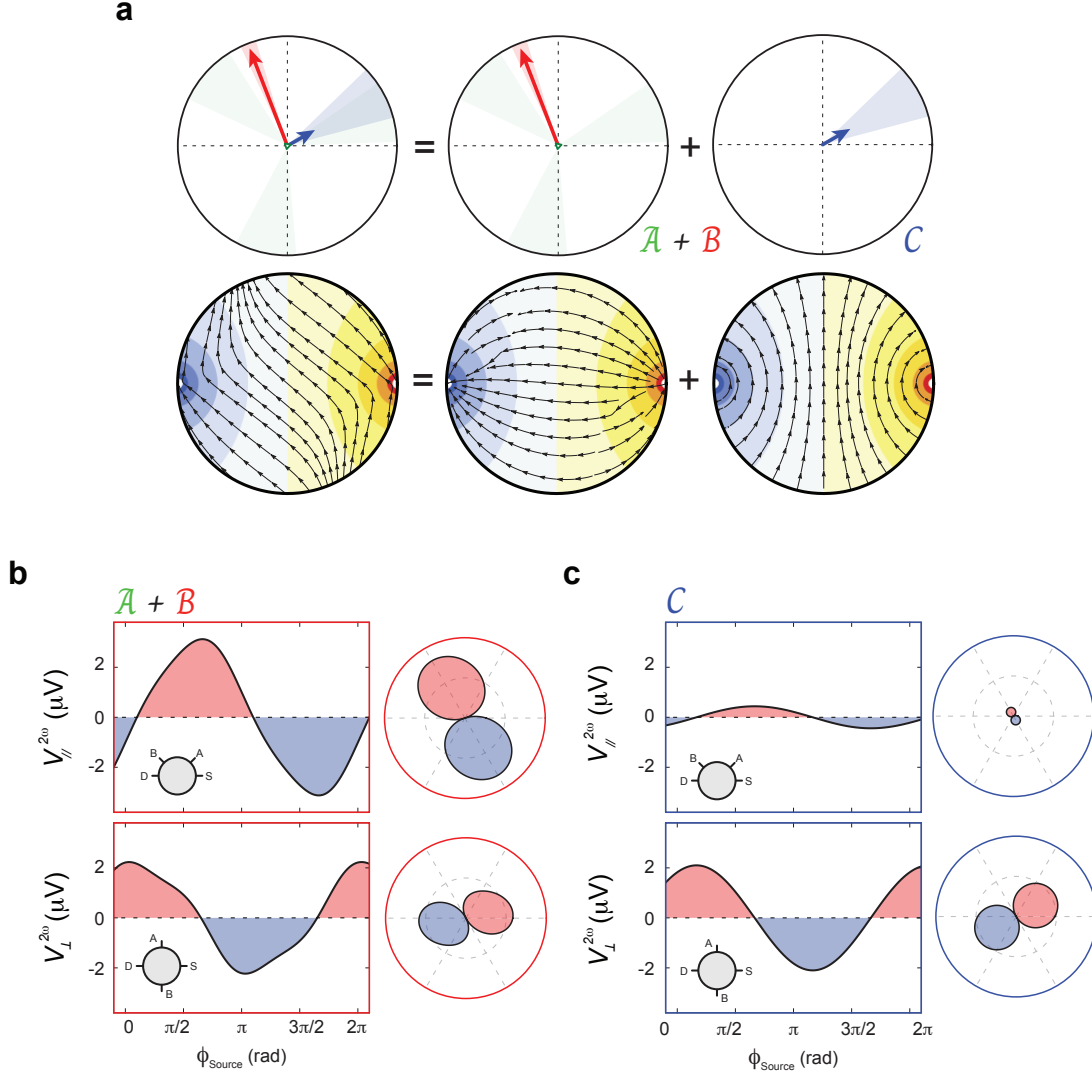


Figure 4.8: **Nonlinear response decomposition by dissipation.**

(a) Decomposition of (top)  $\mathcal{A}$ ,  $\mathcal{B}$ ,  $\mathcal{C}$  vectors representation for the nonlinear conductivity tensor from Fig. 4.6 and (bottom) calculated nonlinear current distribution (black arrows) across the sample by dissipation. Calculation assuming current source-drain located at  $\phi = 0^\circ$  and  $\phi = 180^\circ$ , respectively. Vectors are color coded: green for  $\mathcal{A}$ , red for  $\mathcal{B}$ , and blue for  $\mathcal{C}$ , with the same color shading presenting the uncertainty in orientation. Color map denotes the electrical potential in the sample. Clearly, the nonlinear current in right most diagram, representing the contribution from  $\mathcal{C}$  term, is always parallel to the equal potential line, in other words, perpendicular to the electric field in the sample, thus non-dissipative. (b-c) The angle dependence of  $V_{\parallel}^{2\omega}$  and  $V_{\perp}^{2\omega}$  at the same state but plotted separately for (b) the dissipative contributions and (c) the non-dissipative contribution. Adapted from Ref. [142]

complex parameters by [142]:

$$\mathcal{A}_x + i\mathcal{A}_y = \sqrt[3]{\Xi_-^{(2)}} \quad (4.14)$$

$$\mathcal{B}_x + i\mathcal{B}_y = \Xi_+^{(2)*} + \Xi_0^{(2)}. \quad (4.15)$$

This analysis is essentially another change of basis. The real vectors,  $\mathcal{A}$ ,  $\mathcal{B}$ , and  $\mathcal{C}$  together describe the nonlinear conductivity tensor equivalently as the complex parameters  $\Xi_{+,0,-}^{(2)}$ , or the Cartesian coordinated  $\tilde{\sigma}_{\alpha\mu\nu}$ . The mathematical relations between the three equivalent descriptions are summarized in Appendix D. Importantly, this coordinate free representation offers a straightforward dissociation between the dissipative contributions and non-dissipative contribution of the nonlinear current, as demonstrated in Fig. 4.8a. With the same logic, we can further decompose the nonlinear voltage signals by their dissipation. As shown in Fig. 4.8b-c, the total nonlinear voltage signal contains a large portion of dissipative contribution, with non-trivial functional form and significant magnitude in the transverse channel. Contrasting Fig. 4.8c and Fig. 4.3b, the decomposed non-dissipative signal follows the exact form of the expected response of a nonlinear tensor with only nonzero Hall conductivity. This agreement highlights the *first unambiguous observation* of the nonlinear Hall effect.

## 4.4 Nonlinear Hall effect in BBG $n$ - $D$ phase diagram

To further investigate the origin of this nonlinear Hall effect in BBG, we examine its behavior across the  $n - D$  phase diagram. Fig. 4.9a plots  $\mathcal{A}$ ,  $\mathcal{B}$ , and  $\mathcal{C}$  at three locations in the phase diagram. Upon the reversal of displacement field  $D$ , the directions of  $\mathcal{B}$  and  $\mathcal{C}$  (red and blue vectors) do not rotate. If Berry curvature dipole alone is the origin for the nonlinear response, reversing  $D$  is expected to cause the rotation of  $\mathcal{C}$ , whereas if the underlying mechanism is Berry curvature dipole with quantum geometric or second-order Drude contributions, reversing  $D$  would change the orientation of  $\mathcal{A}$  and  $\mathcal{B}$  [124, 142]. The

invariance of the directions of  $\mathcal{B}$  and  $\mathcal{C}$  contradicts the expectation from a Berry curvature dipole induced nonlinear conductance. Moreover, quantitatively, our observed nonlinear Hall conductivity is several orders of magnitude ( $\sim 10^5$ ) higher than the typical nonlinear Hall conductivity associated with Berry curvature dipole, further refuting Berry curvature dipole as the origin of this nonlinear Hall effect.

Along the same vein, our observed nonlinear Hall conductivity is also two orders of magnitude higher than the typical strength induced by skew scattering. The vanishing  $\mathcal{A}$  further contradicts the prediction from skew scattering or side jumps, which by generally preserving all rotational symmetry, is expected to induce a large 3-fold response, respecting highest rotational symmetry allowed for second harmonic signals.

On the other hand, both  $\mathcal{B}$  and  $\mathcal{C}$  exhibits large rotation, with both of their  $x$ - and  $y$ -components flipping signs, as functions of charge carrier density  $n$ . In fact, such rotation can be demonstrated (in Fig. 4.9b) across the PIP<sub>2</sub> to Sym<sub>4</sub> transitions at various  $D$ , where PIP<sub>2</sub> denotes a region in the BBG  $n - D$  phase diagram with partial isospin polarization consisting of two dominantly filled large Fermi pockets and two smaller Fermi pockets, and Sym<sub>4</sub> denotes a region with degenerate four isospin flavors [132, 151]. This rotation upon isospin order PIP<sub>2</sub>-Sym<sub>4</sub> transitions can be qualitatively accounted for within the second-order Drude contribution by assuming a weak spin-orbit coupling and a weak order parameter that breaks time reversal and rotational symmetries [142]. Such mechanism also correctly reproduces the relative magnitude hierarchy and the orientation between  $\mathcal{A}$ ,  $\mathcal{B}$  and  $\mathcal{C}$ , while persevering an essentially isotropic linear conductivity. However, this mechanism also does not produce the correct absolute magnitudes of the vectors, falling short by a factor of  $\sim 10^4$  than our observation.

To sum up, by systematically analyzing the nonlinear transport behavior, we constructed a complete methodology for *extracting the second order nonlinear conductivity tensor*. In a BBG sample, we demonstrate our method, *observing large nonlinear Hall conductivity* across the  $n - D$  phase diagram. We further demonstrated that the quali-

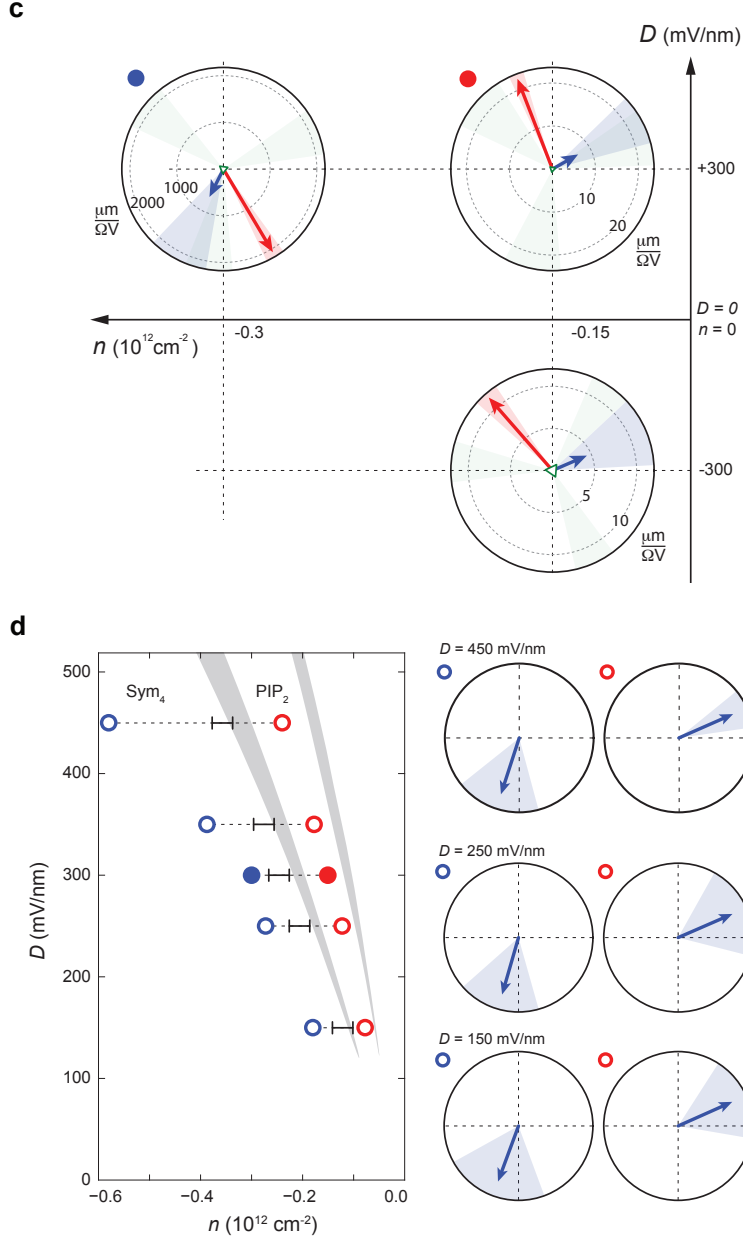


Figure 4.9: **Nonlinear conductivity in BBG  $n - D$  phase diagram.**

(a)  $\mathcal{A}$ ,  $\mathcal{B}$ ,  $\mathcal{C}$  representation of the extracted nonlinear conductivity tensor at (left)  $n = -0.3 \times 10^{12} \text{ cm}^{-2}$ ,  $D = 300 \text{ mV/nm}$ , (right top)  $n = -0.15 \times 10^{12} \text{ cm}^{-2}$ ,  $D = 300 \text{ mV/nm}$ , and (bottom)  $n = -0.15 \times 10^{12} \text{ cm}^{-2}$ ,  $D = -300 \text{ mV/nm}$ . Notice the invariant in vector directions upon reversing  $D$  and the dramatic rotation with change  $n$ . (b)  $\mathcal{C}$  extracted across the  $\text{PIP}_2$ - $\text{Sym}_4$  transitions at various  $D$ . The rotation of  $\mathcal{C}$  is general to the isospin transition. All extraction done with measurements performed at  $I_{AC} = 40 \text{ nA}$ ,  $T = 20 \text{ mK}$  and  $B = 0$ . Adapted from Ref. [142]

tative behavior for the observed nonlinear Hall conductivity in BBG can be explained by second-order Drude contribution with some assumptions, yet no existing theoretical framework, to the best of our knowledge, can reproduce the observed nonlinear effect quantitatively.

## 4.5 Side aisle: the quantitative reconstruction of Fermi surface contour

Having the ability of extracting the nonlinear conductivity tensor, we are also granted the power of quantitatively reconstructing the Fermi surface contours. Quantum oscillation data can reveal the number of Fermi pockets at a particular state. In Fig. 4.10b, a sharp peak at  $1/4$  of the renormalized magnetic field frequency suggests a (roughly) 4-fold generate Fermi surface. Indeed, BBG at this particular  $n, D = -0.3 \times 10^{12} \text{ cm}^{-2}, 300 \text{ mV/nm}$  is expected to be at its  $\text{Sym}_4$  phase [151]. A generic, unperturbed 4-fold generate Fermi surface is shown in Fig. 4.10c as the black dashed lines, with each square representing one isospin flavor.

We then perturbatively distort these generic contours, and use second order Boltzmann transport theory to calculate the expected nonlinear conductivity tensor. With some fine-tuning and iterations, we are able to find the correct distortion that would generate the same nonlinear conductivity tensor from our experimental extraction. In other words, knowing the number of Fermi pockets from quantum oscillation, and the extracted nonlinear conductivity tensor, we can reconstruct the distortion on Fermi surface contours quantitatively using second order Boltzmann transport theory.

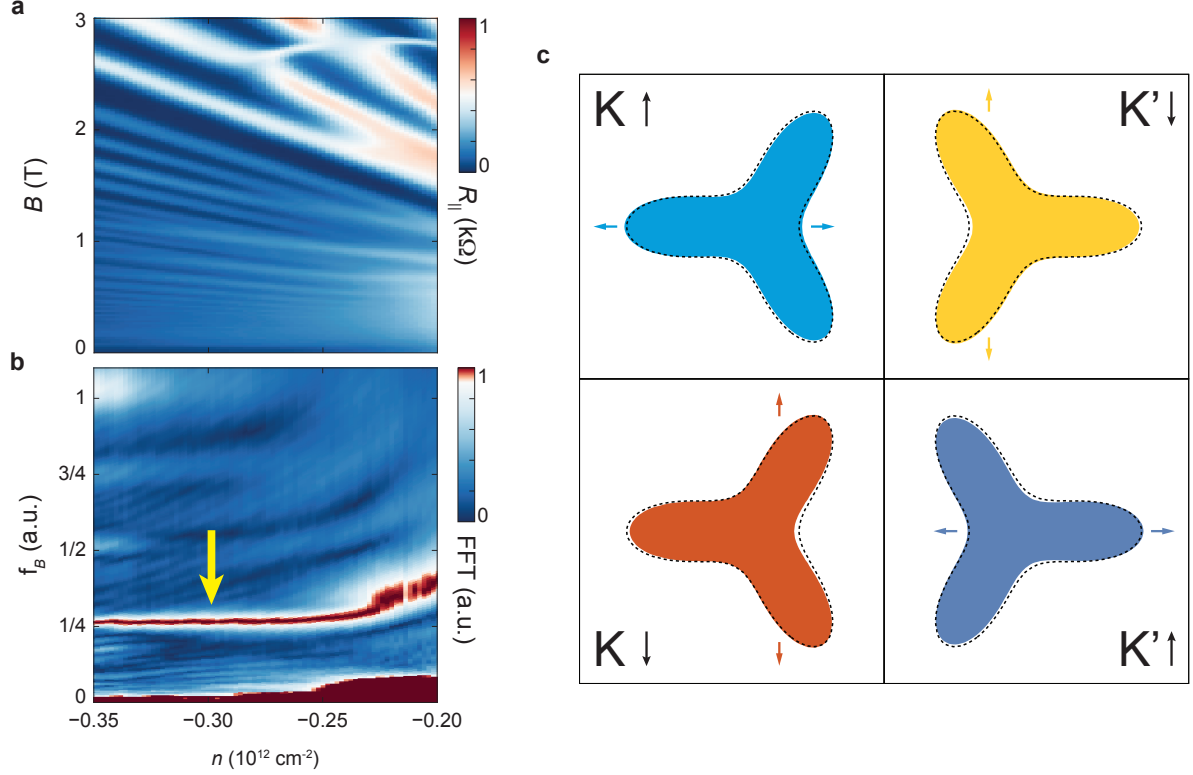


Figure 4.10: **Quantitative reconstruction of Fermi surface contour.**

(a) Linear longitudinal resistance  $R_{\parallel}$  as a function of density and magnetic field, measured at  $D = 300$  mV/nm. (b) Fast Fourier transform of panel (a). The peak near  $1/4$  at  $n = -0.3 \times 10^{12} \text{ cm}^{-2}$  (marked by yellow arrow) indicates four nearly degenerate Fermi pockets, expected for a  $\text{Sym}_4$  order. (c) The quantitatively reconstructed Fermi surface contours for different isospin flavors in the  $\text{Sym}_4$  phase. The black dashed contours represent the unperturbed Fermi surface contours. Interestingly, pockets are distorted along the same direction for same spins and oppositely for the same valleys. Grouping the isospin flavors into two antiferromagnetic pairs ( $K \uparrow, K' \downarrow$  vs  $K \downarrow, K' \uparrow$ ), the Fermi surface contours corresponding to one of the pairs (demonstrated as  $K \uparrow, K' \downarrow$  here) is subtly larger than the contours of the other. Adapted from Ref. [142]



# CHAPTER 5

## Excitons in two-component fractional quantum Hall effect

### 5.1 Excitonic transport in quantum Hall bilayer

So far, we have explored the rich physics associated with rotational symmetry breaking in two-dimensional electron systems. Naturally, while such in-plane symmetry breaking orders are well-suited to be probed using planar device architectures, such as the sunflower geometry discussed earlier, phenomena governed by interlayer interactions, which extend beyond the plane, call for vertically structured devices. As mentioned in the introduction, quantum Hall bilayers provide a versatile platform for engineering strong interlayer Coulomb coupling, enabling the realization of interlayer excitons: Coulomb driven bound states formed between electrons and holes residing in different layers with the presence of perpendicular magnetic field.

In a single 2DEG system under a strong perpendicular magnetic field, kinetic energy of 2D electrons is quantized into discrete Landau levels (LLs), each with a degeneracy  $n_0 = eB/h$  per unit area, where  $e$  is the elementary charge and  $h$  is the Planck constant. When the carrier density  $n$  satisfies  $\nu = n/n_0 \in \mathbb{Z}$ , the system forms an incompressible

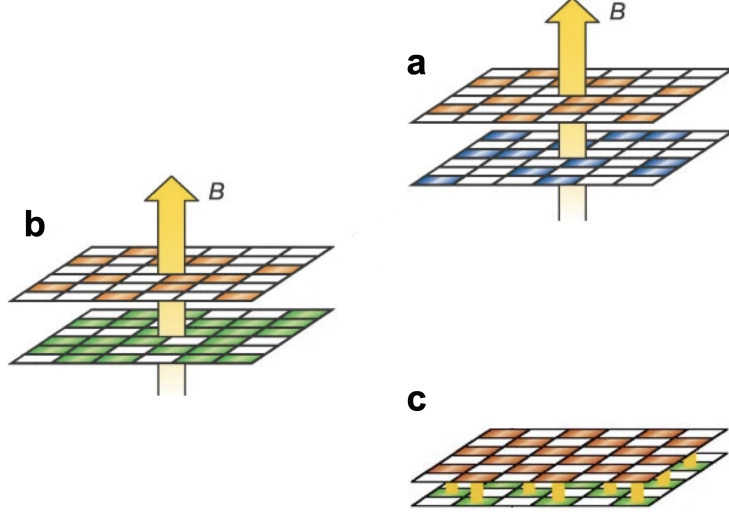


Figure 5.1: **Schematic for interlayer excitons at  $\nu_{\text{total}} = 1$ .**

(a) Schematic of a two parallel layers of 2DEG in magnetic field. Degeneracy of LL is denoted as checkerboard sites here. Sites occupied with electrons are colored as orange and blue. (b) Applying particle-hole transformation to the bottom electron layer emphasizes the unoccupied sites, or in other words, the holes (green) in that layer. This transformation changes the sign of Coulomb interactions between the layers. (c) Interlayer coherent, excitonic BEC superfluid occurs when electrons in the top layer bind with the holes in the bottom layer, happening when  $\nu_{\text{total}} = 1$ . Magnetic field not shown in this panel for visual clarity. It is important to note that, these checkerboards only represent the degeneracy of the LL, not spatial relation of the particles within each layer and does not imply crystalline structure. Adapted from Ref. [89]

quantum Hall state characterized by quantized edge/Hall conductance and charge gap in bulk. In bilayer systems, each layer hosts its own set of LLs and corresponding filling factors  $\nu_1$  and  $\nu_2$ . Strong interlayer Coulomb interactions can correlate electrons across layers, driving the system into a collective state when the total filling factor  $\nu_{\text{total}} = \nu_1 + \nu_2$  becomes an integer. In particular, at  $\nu_{\text{total}} = 1$ , it is demonstrated that the system can develop spontaneous interlayer phase coherence, forming interlayer excitons in which each electron in one layer is bound to a hole in the other (see Fig. 5.1) [89]. Unlike their optically generated, short-lived counterparts, these magnetically stabilized excitons emerge

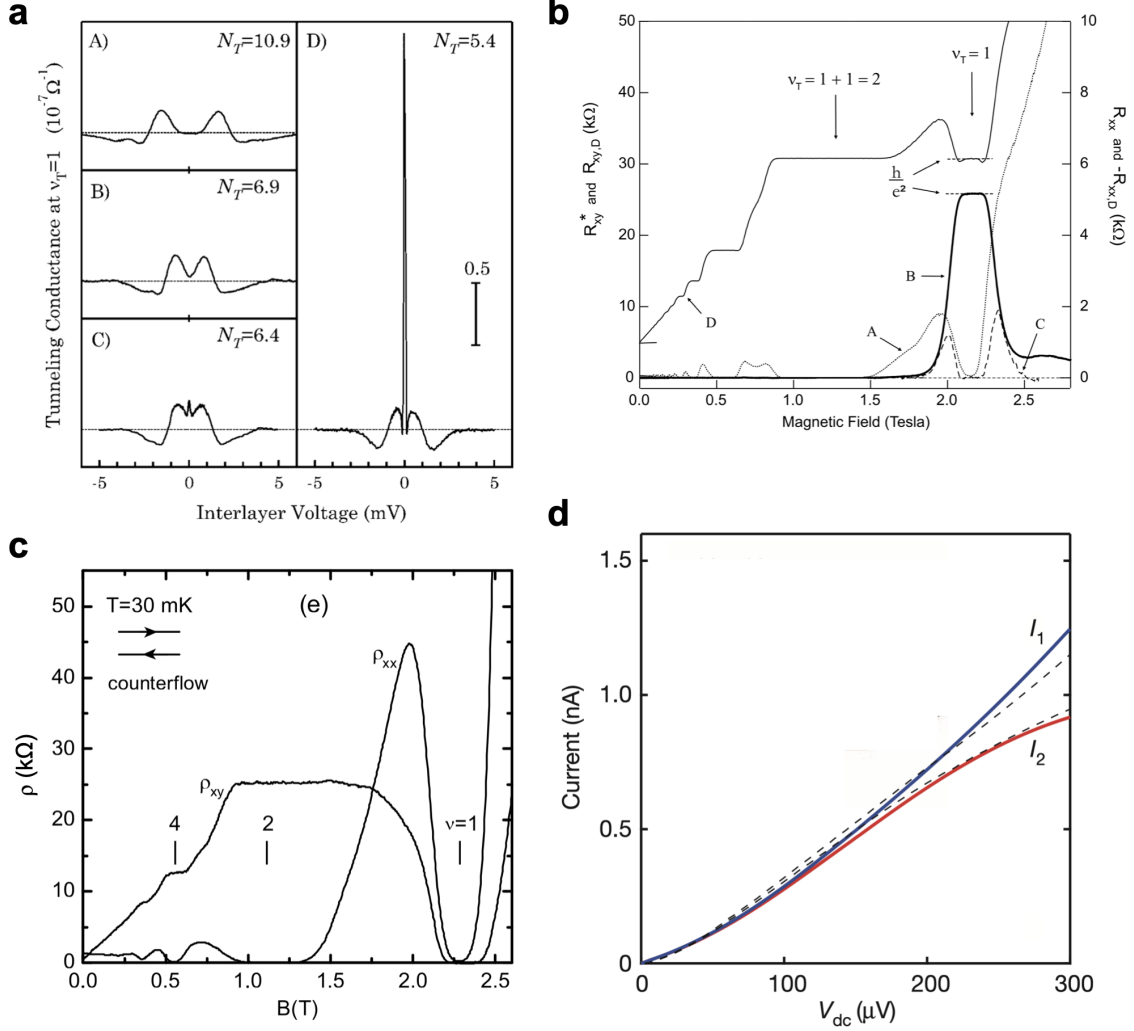


Figure 5.2: **BEC superfluid signatures at  $\nu_{\text{total}} = 1$ .**

(a) Tunneling conductance across the two layers measured at different  $d/l_B$ , at  $\nu_{\text{total}} = 1$ . In the small  $d/l_B$  regime (right), where the interlayer couple is strong compared to the intralayer interactions, a sharp peak in the tunneling conductance at zero bias suggest the presences of interlayer coherence, a signature of excitonic BEC superfluidity. (b) Focusing on trace B (thick solid line), Hall drag resistance quantizes into  $h/e^2$  at  $\nu_{\text{total}} = 1$ . (c) Counterflow resistivity as a function of  $B$ . At the  $\nu_{\text{total}} = 1$ , both longitudinal and transverse counterflow resistivity drop to zero, demonstrating the dissipationlessness. (d) Perfect Coulomb drag demonstrated by the matching drive and drag current ( $I_1$  and  $I_2$ ) at low d.c. bias voltage. The bifurcation of  $I_1$  and  $I_2$  at larger d.c. bias suggests the breakdown of the interlayer coherent state, analogous to the critical supercurrent behavior in a SC. Adapted from Ref. [84, 85, 88, 93]

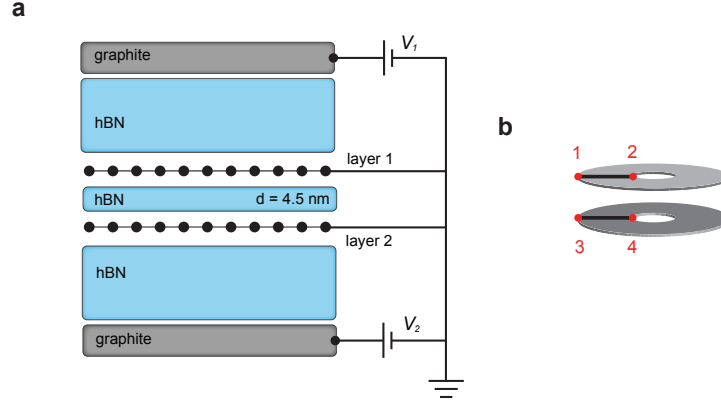


Figure 5.3: **Double Corbino graphene/hBN quantum Hall bilayer.**

(a) Schematic of the sample structure used in our experiment. A graphene/hBN quantum Hall bilayer is encapsulated two top and bottom graphite gate, allowing independent control of carrier density in each layer. (b) The device is fabricated into the edgeless double Corbino geometry, sensitive to only the bulk properties of the sample. Separate electrical contacts to each graphene layer allow layer sensitive transport measurement. Outer and inner contacts on each layer are labeled as 1 to 4. Adapted from Ref. [152, 153]

as true ground states of the system [154–156]. As a result, they are inherently stable and can directly participate in d.c. transport. Even more intriguingly, since these excitons are bosonic pairs, they undergo Bose-Einstein condensation into a coherent superfluid ground state, giving rise to distinctive transport phenomena such as zero-bias interlayer tunneling, quantized Hall drag, dissipationless counterflow, and perfect Coulomb drag (see Fig. 5.2) [54, 83–85, 87–89, 93, 101]. In the following, I will review key experimental signatures of excitonic transport in our setup, focusing on the well studied state at  $\nu_{\text{total}} = 1$ .

The device structure is shown in Fig. 5.3. Using the general stacking design for electronic transport studies in Fig. 1.2, the graphene/hBN quantum Hall bilayer is sandwiched by top and bottom graphite gates, enabling independent electrostatic doping for each graphene layer. Electrical contacts are made to top and bottom graphene layers separately, allowing layer independent voltage bias. Owing to the topological nature of the QHE, it is useful to distinguish between edge and bulk transport. In our case, the Corbino geometry naturally isolates the bulk response, as it does not couple to edge modes. However, it also

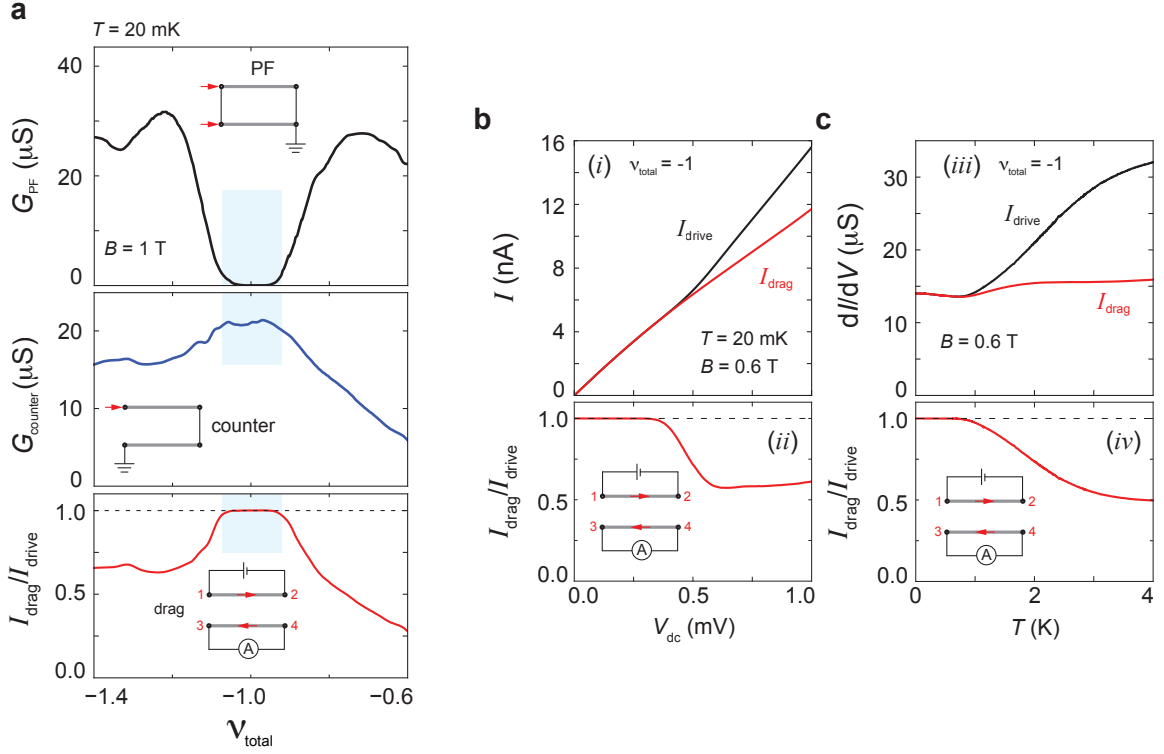


Figure 5.4: **Measurement configurations in double Corbino geometry.**

(a) Three measurement configurations enabled by the sample structure. In the pretense of robust charge gap with charge neutral excitonic flow, as demonstrated, we observe (top) zero conductance in parallel flow (PF) geometry,  $G_{\text{PF}} = 0$ , (middle) finite counterflow (counter) conductance  $G_{\text{counter}} > 0$  and (bottom) matching drive and drag current the Coulomb drag (drag) setup,  $I_{\text{drag}}/I_{\text{drive}} = 1$ . (b-c) With finite binding energy, excitons can be depaired when given large kinetic energy via (b) large d.c. bias, or (c) high temperature, as indicated by the bifurcation of  $I_{\text{drag}}$  and  $I_{\text{drive}}$  and thus the deviation of their ratio from 1. Adapted from Ref. [152]

introduces finite contact resistance by limiting us at 2-terminal measurements.

As illustrated in Fig. 5.4a, there are three distinct transport configurations that we use to identify the coexistence of charge gap and excitonic neutral mode. In the parallel flow (PF) configuration, a charge gap is identified as an insulating feature in zero bulk conductance,  $G_{\text{PF}} = 0$ , due to the lack of free charge carriers in both layers. Enabled by the exciton flow in the system, equal and opposite current can flow across the layers in the counterflow (counter) configuration, giving rise to a nonzero conductance in the counterflow geometry,  $G_{\text{counter}} > 0$ . It is worth noting that, as demonstrated here, because

exciton are overall charge neutral objects, such exciton flow can coexist with charge gap indicated by  $G_{\text{PF}} = 0$ . In fact, as demonstrated in Fig. 5.2c, since a BEC excitonic superfluid is expected at  $\nu_{\text{total}} = 1$ , counterflow is expected to be dissipationless. However the contact resistance, introduced by the 2-terminal nature of Corbino geometry, prohibits us from observing a diverging  $G_{\text{counter}}$ . Most interestingly, if the two layers are separated into two circuits, we can send a current,  $I_{\text{drive}}$ , through one layer and measure the induced Coulomb drag current,  $I_{\text{drag}}$ , on the other layer. With the presence of exciton flow and absence of free charge carriers, the currents in two layers perfectly match each other in opposite directions, giving rise to a drag-to-drive current ratio  $I_{\text{drag}}/I_{\text{drive}} = 1$ , or the so-called “perfect drag.”

Furthermore, since excitons are bounded pairs, to an extent just like Cooper pairs in superconductors, their binding can be broken with high temperature or large bias, similar to the critical temperature and critical supercurrent for Cooper pairs. In Fig. 5.4b-c, we demonstrate the depairing of perfect drag using both mechanisms. Notice our measurement is done at  $\nu_{\text{total}} = -1$ , the electron-hole conjugate state of the generally discussed  $\nu_{\text{total}} = 1$  phase. Related by electron-hole transformation, they are equivalently described by the same wavefunction [157]. This choice is made by the fact that electrical contacts perform more consistently on the hole filling side. In the following, while we generally carry out discussions assuming electrons as the charge carriers, while our measurements are done mainly in hole conjugate states, with essentially identical physical pictures guaranteed by the electron-hole symmetry of the system [99].

The  $\nu_{\text{total}} = 1$  state demonstrates a wealth of interesting transport behaviors, including clear evidence of interlayer coherence and excitonic condensation, as seen both in GaAs quantum Hall bilayers and in our own graphene/hBN sample. While much remains to be understood in this regime, it is natural to ask what new phenomena may arise in the fractional quantum Hall regime of bilayers. Could similar pairing mechanisms appear at fractional fillings? In particular, the outstanding similarity between the proposed

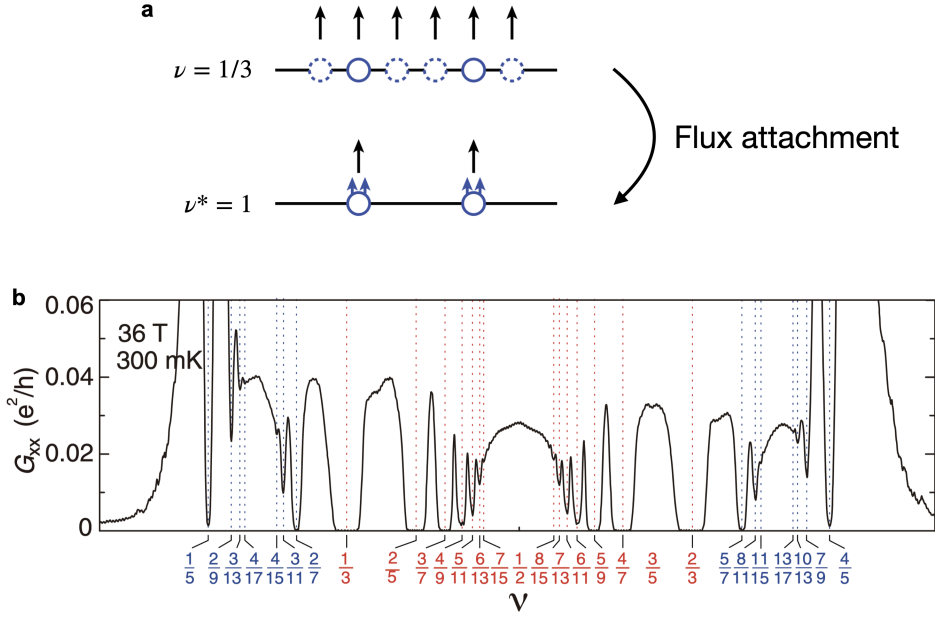
states at  $\nu_{\text{total}} = 1$  and  $\nu_{\text{total}} = 1/3$ , described by the analogous (111) and (333) Halperin wavefunctions, suggests that excitonic physics may also play a role in two-component FQHE systems [157]. To explore this, we now shift our focus to fractional quantum Hall bilayer system, beginning with a brief overview of the composite fermion picture, its extension to two-component systems, and the resulting phase diagram.

## 5.2 Two-component fractional quantum Hall effect

Again, we start with a single layer of 2DEG, subject to a even stronger perpendicular magnetic field. As kinetic energy is further quenched with higher  $B$ , stronger electron-electron interaction causes more gaps to open within a LL at fractional filling factors, yielding incompressible quantum Hall states at these fractional fillings. This phenomenon is referred to as the fractional quantum Hall effect (FQHE). Similar to the integer case, these FQH states manifests themselves as bulk insulators with conducting edge modes (see Fig. 5.5b). Remarkably, the fractions at which FQHE would emerge can be nicely summarized using the composite fermion model, where an even number of magnetic flux quanta are attached to each electron, together viewed as the so-called composite fermions (CFs) [158]. Upon this operation of flux attachment, the strongly interacting electrons are converted into weakly coupled CFs. The magnetic field  $B$  is also effectively reduced for these CFs, given by:

$${}^aB^* = B - an\phi_0, \quad (5.1)$$

where  $a$  is the flux attachment number,  $\phi_0$  is the magnetic flux quantum,  ${}^aB^*$  is the effective magnetic field for CFs with  $a$  flux attached. Reducing the effective magnetic field, the number of flux attachment essentially describes the interaction strength in the system. CFs under  ${}^aB^*$  also form Landau-like levels ( $\Lambda$ -levels), corresponding to a series of effective QHE of CFs, when the effective CF filling factor,  $\nu^*$ , is integer. And  ${}^a\nu^*$  is





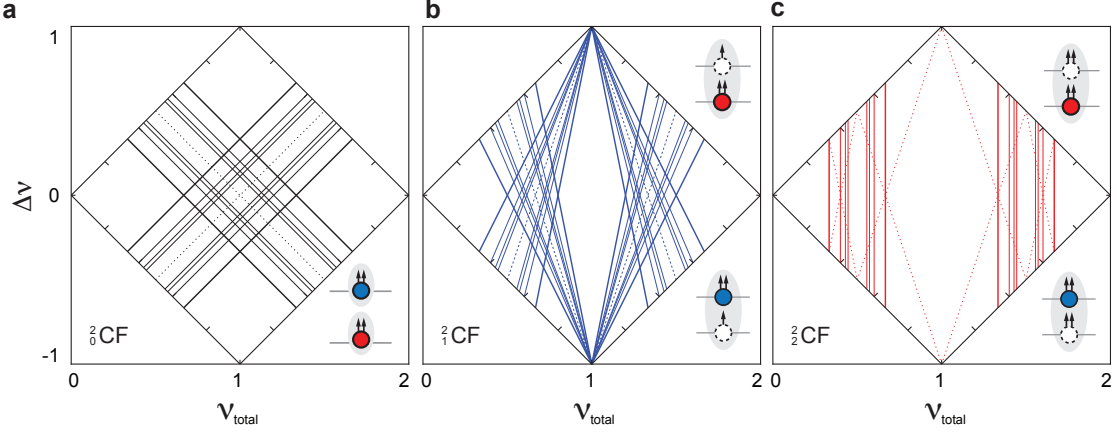


Figure 5.6: **Two-component FQHE phase diagram.**

Schematic phase diagrams of FQHE associated with the (a)  $\frac{2}{0}$ CF, (b)  $\frac{2}{1}$ CF, and (c)  $\frac{2}{2}$ CF construction. The diamond denotes the boundaries of  $0 \leq \nu_i \leq 1$  for each layer. Solid lines denotes the location of the expected FQH states in the  $\nu_{\text{total}} - \Delta\nu$  phase diagram. Adapted from Ref. [152]

we denote the CF constructed with  $a$  intralayer and  $b$  interlayer flux attachments as  $\frac{a}{b}$ CF.

The effective CF filling factor for each layer is given by:

$$\frac{a}{b}\nu_1^* = \frac{\nu_1}{1 - a\nu_1 - b\nu_2} \quad (5.3)$$

$$\frac{a}{b}\nu_2^* = \frac{\nu_2}{1 - a\nu_2 - b\nu_1}, \quad (5.4)$$

where  $\frac{a}{b}\nu_i^*$  is the effective filling of  $\frac{a}{b}$ CFs on layer  $i$ . Similarly, the flux attachment number  $a$  and  $b$  describes the intra- and inter-layer interaction strength in the system, and two-component Jain states arise as a sequence when  $\frac{a}{b}\nu_i^* \in \mathbb{Z}$  [99, 100]. Note that the intralayer flux attachment number  $a$  is still required to be even, in order to keep the composite particle fermionic upon exchange. The interlayer flux attachment number  $b$  can be any non-negative integer, odd or even, that is smaller than or equal to  $a$ . The disregarding towards evenness is due to the fact that particles from opposite layers are distinguishable from each other. Thus  $b$  does not alter the exchange statistics. And the requirement  $b \leq a$  arises from the stability requirement of the plasma analogy as well as

the positive-definiteness of the  $\mathbf{K}$ -matrix [157, 163, 164].

With two kinds of flux attachment joining the picture, even the simplest two-component fractional quantum Hall constructions can give rise to a very rich phase diagram. We now limit ourselves with  $a = 2$ , the smallest possible nonzero intralayer flux attachment number, and examine the appearance of states across the phase diagram of  $\nu_{\text{total}} - \Delta\nu$ , where  $\nu_{\text{total}} = \nu_1 + \nu_2$  is the total filling of both layers as before, and  $\Delta\nu = \nu_1 - \nu_2$  is the layer imbalance of fillings.

First, somewhat trivially shown in Fig. 5.6a, without interlayer flux attachment, the two layer are essentially decoupled. In this scenario, the electrons in each layer does not “feel” their neighbors in the other layer. Two sets of independent FQHE appear the phase diagram, which are expected by plugging  $b = 0$  into Eqn. 5.3 - 5.4, re-obtaining the single layer effective filling Eqn. 5.2 for each layer. Next, if the interlayer coupling is somewhat strong, e.g.,  $b = 1$ , Eqn. 5.3 - 5.4, predicts a different Jain sequence, as shown in Fig. 5.6b. Due to the two-component nature, the location at which the states appear in the phase diagram now simultaneously depends on the filling of both layers. Lastly, Fig. 5.6c shows the expected trajectories of the Jain sequence when the interlayer coupling is comparably strong to intralayer interactions,  $a = b = 2$  in our case. One important feature of these states is the insensitivity to  $\Delta\nu$ , which is, as we are going to explore in the next section, closely related to the interlayer coherent nature of these states.

To identify these theoretical expected states in our system, we now turn to the phase diagram obtained from our measurements. By locating charge gaps as insulating features in the  $\nu_{\text{total}} - \Delta\nu$  plane and comparing them against the predicted trajectories in Fig. 5.6, we are able to assign the observed states to their corresponding CF sequences as shown in Fig. 5.7a. In particular, at  $B = 12$  T, we observe two-component FQHE sequences from all three CF constructions, among whom the competition is delicately biased by the screening effects. At the center of the LL, the high density of state yields a stronger intralayer coupling, thus promoting the  ${}^2_0\text{CF}$  construction. Conversely, at the edge of the

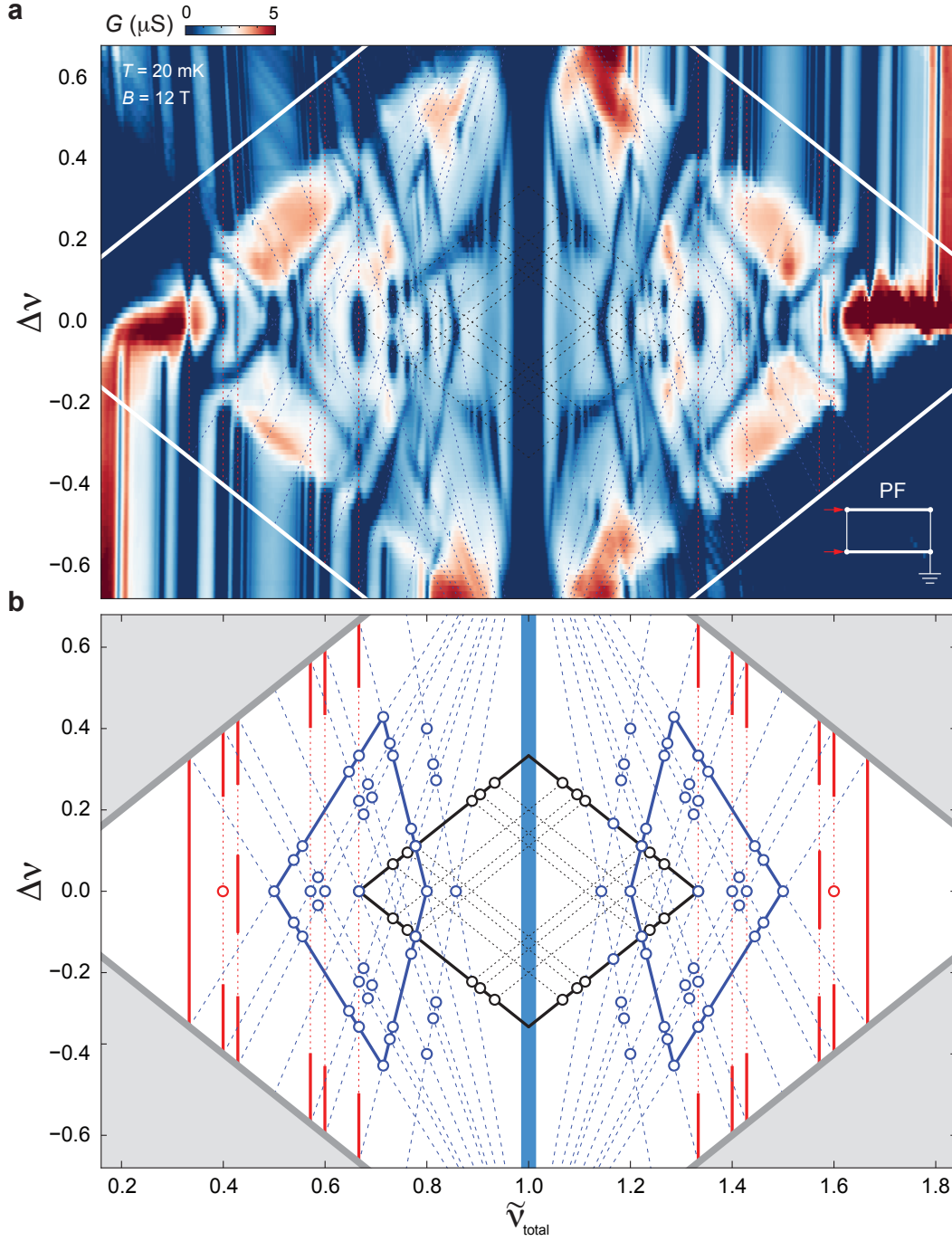


Figure 5.7: **PF conductance in  $\nu_{\text{total}}-\Delta\nu$  phase diagram at  $B = 12$  T**

(a)  $G_{\text{PF}}$  as a function of  $\nu_{\text{total}}$  and  $\Delta\nu$  measured at  $B = 12$  T,  $T = 20$  mK. (b) Schematic phase diagram showing the FQH states associated with  $0^2\text{CF}$  (black),  $1^2\text{CF}$  (blue), and  $2^2\text{CF}$  (red) constructions. Adapted from Ref. [152]

LL, intralayer interaction is relatively weaker, manifested as  $\frac{2}{2}$ CF states. Somewhere in between, we observe the FQH sequence corresponding to the  $\frac{2}{1}$ CFs (see Fig. 5.7b). As mention earlier, due to electrical contact constraints, the measurement is carried out on the hole doping side, meaning  $-1 \leq \nu_1 \leq 0$  and  $-1 \leq \nu_2 \leq 0$ , and we plot our data in the electron-hole transformed axis. The transformation is two-fold: we first mirror  $\nu_1$  and  $\nu_2$  around 0, using the general electron-hole symmetry; then we mirror  $\nu_1$  and  $\nu_2$  around  $1/2$ , taking advantage of the electron-hole symmetry within the first LL e.g.,  $\nu = 2/3$  state is the hole conjugate version of  $\nu = 1/3$  for electrons. Together, we have:  $\tilde{\nu}_1 = \nu_1 + 1$  and  $\tilde{\nu}_2 = \nu_2 + 1$  or equivalently  $\tilde{\nu}_{\text{total}} = \nu_1 + \nu_2 + 2$  and  $\Delta\tilde{\nu} = \nu_1 - \nu_2 = \Delta\nu$  as our electron-hole transformed axes.

Having this clear mapping of the phase diagram, we now examine the excitonic behaviors at the strong interlayer coupling regime, the sequences corresponding to  $\frac{2}{2}$ CF constructions, where the excitonic pairing might be favored by the interlayer coupled nature of the states.

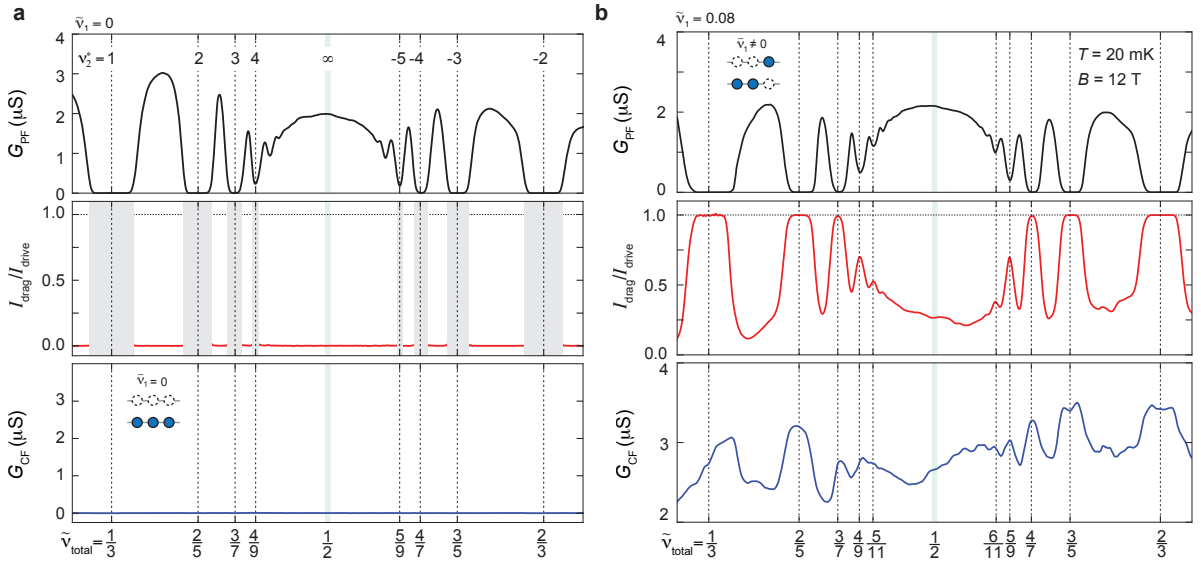


Figure 5.8:  $\frac{2}{2}$ CF FQHE sequence with interlayer excitons.

(a-b)  $G_{\text{PF}}$  (top),  $I_{\text{drag}}/I_{\text{drive}}$  (middle), and  $G_{\text{CF}}$  (bottom panel) as a function of  $\tilde{\nu}_{\text{total}}$  measured in (a) single-layer regime with  $\tilde{\nu}_1 = 0$  and (b) the two-component regime with  $\tilde{\nu}_1 = 0.08$ . In panel (a), drag ratio is ill-defined at  $\tilde{\nu}_0 \nu_2^* = N \in \mathbb{Z}$  where both  $I_{\text{drive}}$  and  $I_{\text{drag}}$  are zero (masked by gray-shaded stripes). Measurements performed at  $B = 12$  T and  $T = 20$  mK. Adapted from Ref. [152]

### 5.3 Excitons in $\frac{2}{2}\text{CF}$ states

As shown in Fig. 5.8a,  $G_{\text{PF}}$  as a function of  $\nu_{\text{total}}$  at  $\nu_1 = 0$  exhibits the classic Jain sequence behavior, with insulating features appearing at  $\frac{2}{0}\nu_2^* = N \in \mathbb{Z}$ . Due to the fact that one two layer is empty, and thus no particle to be interacting with, all three CF constructions predicts the same state sequence, disregarding their difference in  $b$ . Similarly, the empty layer 1 also prohibits any exciton pairing, evidenced by the vanishing  $G_{\text{counter}}$  and ill-defined  $I_{\text{drag}}/I_{\text{drive}}$ . From this “vacuum” FQHE sequence, excitons can be introduced by filling  $\nu_1$  to be nonzero. As shown in Fig. 5.8b,  $G_{\text{PF}}$  as a function of  $\nu_{\text{total}}$  at  $\nu_1 = 0.08$  exhibits the same Jain sequence behavior, accompanied by highly conductive  $G_{\text{counter}}$  and perfect drag  $I_{\text{drag}}/I_{\text{drive}} = 1$  at each state, demonstrating the presence of excitonic pairings.

By examining the evolution of these transport behaviors with varying  $\nu_1$ , we further

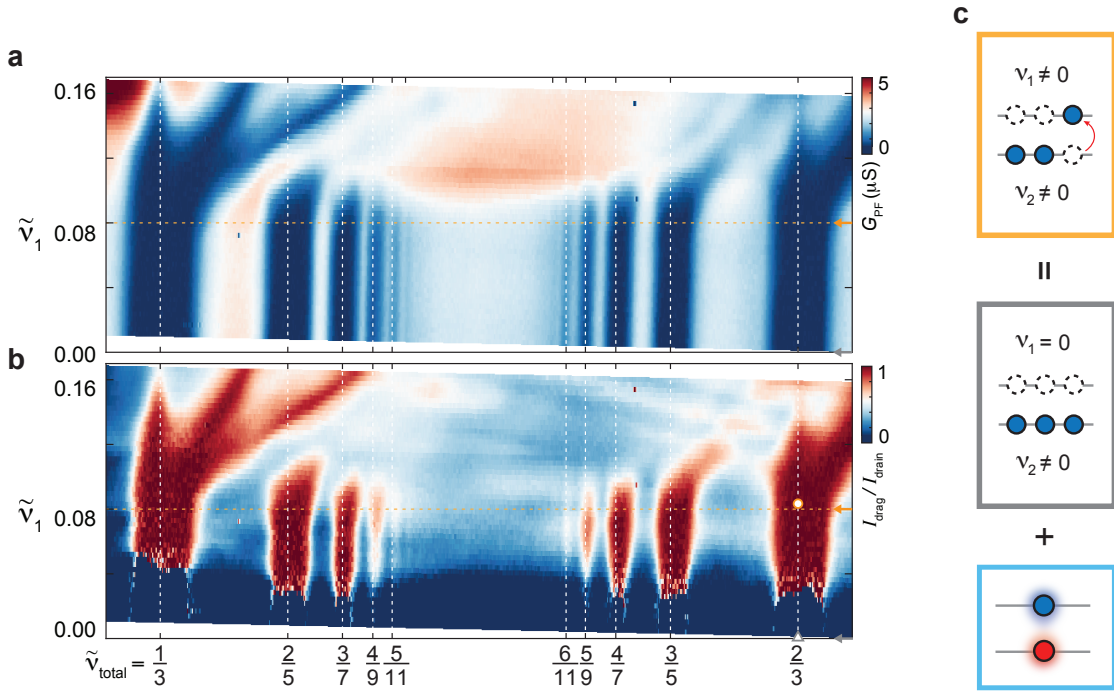


Figure 5.9:  $\frac{2}{2}\text{CF}$  excitons in  $\nu_{\text{total}}-\nu_1$  plane.

(a)  $G_{\text{PF}}$  and (b) drag ratio as a function of  $\tilde{\nu}_{\text{total}}$  and  $\tilde{\nu}_1$ . Gray and orange arrows marks where the measurements in Fig. 5.8a and b are made. (c) Schematic diagram illustrating interlayer exciton doping via varying  $\Delta\nu$ . Measurements performed at  $B = 12$  T and  $T = 20$  mK. Adapted from Ref. [152]

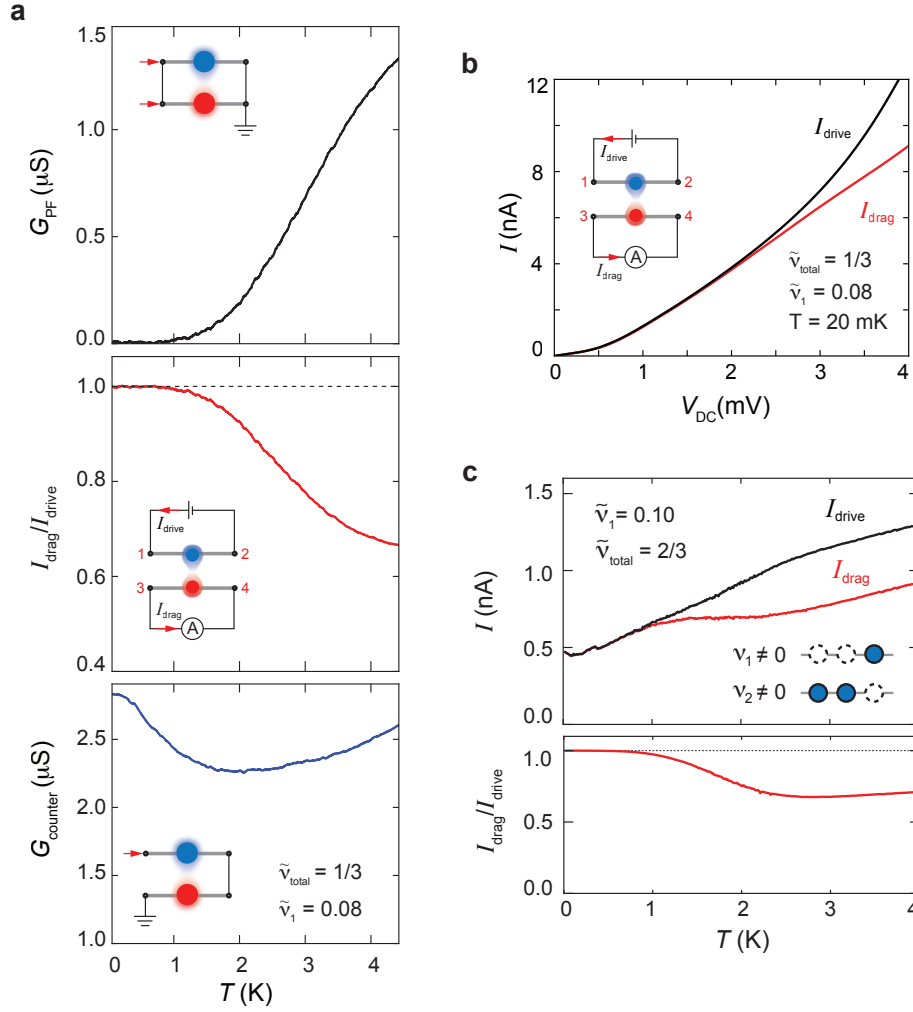


Figure 5.10: **Depairing of  $2/2$  CF excitons via temperature and bias**

(a)  $G_{PF}$  (top),  $I_{drag}/I_{drive}$  (middle), and  $G_{counter}$  (bottom panel) as a function of temperature. (b)  $I_{drag}$  (red) and  $I_{drive}$  (black) as a function of d.c. bias. Measured at  $\tilde{\nu}_{total} = 1/3$  and  $\tilde{\nu}_1 = 0.08$ . The bifurcation between  $I_{drag}$  and  $I_{drive}$  and thus the deviation of  $I_{drag}/I_{drive}$  from 1 demonstrates the depairing of excitons at this state with high temperature and large d.c. bias. (c)  $I_{drag}$  (red) and  $I_{drive}$  (black) (top) and  $I_{drag}/I_{drive}$  (bottom) drag ratio as a function of temperature, measured at  $\tilde{\nu}_{total} = 2/3$  and  $\tilde{\nu}_1 = 0.1$ . Similarly, excitons here can be broken with temperature. Measurements performed at  $B = 12$  T. Adapted from Ref. [152]

explore the nature of the excitonic pairing at these states. Fig. 5.9a-b shows  $G_{PF}$  and  $I_{drag}/I_{drive}$  as a function of  $\nu_{total}$  and  $\nu_1$ . Notably, the charge gap of the FQH states remain robust against the introduction of excitons, and the excitons extends over a large range of layer imbalance. The first observation is consistent with the charge neutral nature of excitons and the mechanism of this exciton doping is illustrated in Fig. 5.9c.

The second observation reveals the similarity between these excitons in the  $\frac{2}{2}\text{CF}$  FQH states and those at  $\nu_{\text{total}} = 1$ . Focusing particularly on the  $\nu_{\text{total}} = 1/3$  state described by the (333) Halperin wavefunction. The doping dependence of the state satisfy  $\frac{2}{2}\nu_{\text{total}}^* = \frac{2}{2}\nu_1^* + \frac{2}{2}\nu_2^* = 1$ , resembling that of the (111) state, where  $\nu_{\text{total}} = \nu_1 + \nu_2 = 1$ , only substituting electron fillings with  $\frac{2}{2}\text{CF}$  effective fillings. Such requirement, illustrated as vertical trajectories Fig. 5.6c, suggests that the excitons are paired across partially filled  $\Lambda$ -levels of the  $\frac{2}{2}\text{CF}$ s, fractionally analogous to the  $\nu_{\text{total}} = 1$  case. Furthermore, just like the  $\nu_{\text{total}} = 1$  case, we can demonstrate the depairing of these excitonic parings using d.c. bias and temperature, as shown in Fig. 5.10a-b.

Given the observational and theoretical high resemblance to the superfluidity at  $\nu_{\text{total}} = 1$ , the ground state of the excitonic pairings at this  $\nu_{\text{total}} = 1/3$  state is likely a *condensate*, defined by a low temperature Berezinskii-Kosterlitz-Thouless transition [95, 101, 165–170]. Besides  $\nu_{\text{total}} = 1/3$ , other  $\frac{2}{2}\text{CF}$  state also exhibit similar behaviors, as demonstrated in Fig. 5.8, 5.9, and 5.10, suggesting a potentially similar excitonic nature.

## 5.4 Excitons in $\frac{2}{1}\text{CF}$ states

Next, we examined the transport behaviors at FQH states corresponding to the  $\frac{2}{1}\text{CF}$  construction. Although weaker than the already discussed  $\frac{2}{2}\text{CF}$  states,  $\frac{2}{1}\text{CF}$  sequences also require interlayer coupling to emerge, thus can potentially host excitons. We fix the effective density of one layer always at  $\frac{2}{1}\nu_1^* = 1$ , and sweep the density of the other layer. Because layer 1 is always in the  $\frac{2}{1}\nu_1^* = 1$  FQH state, thus the overall  $G_{\text{PF}}$  is governed by the conductance of layer 2. As shown in Fig. 5.11b,  $G_{\text{PF}}$  again, exhibits the classic Jain sequence behavior, with insulating features appearing at  $\frac{2}{1}\nu_2^* = N \in \mathbb{Z}$ , confirming that the  $\frac{2}{1}\text{CF}$  construction is responsible for the observed states. We can further demonstrate this by measuring the activation behavior and extract the value of charge gap  $\Delta$  at each state along this sequence. As shown in Fig. 5.12 the hierarchical behavior of  $\Delta$ , typical to FQHE sequences, provides strong evidence that these observed states indeed came the

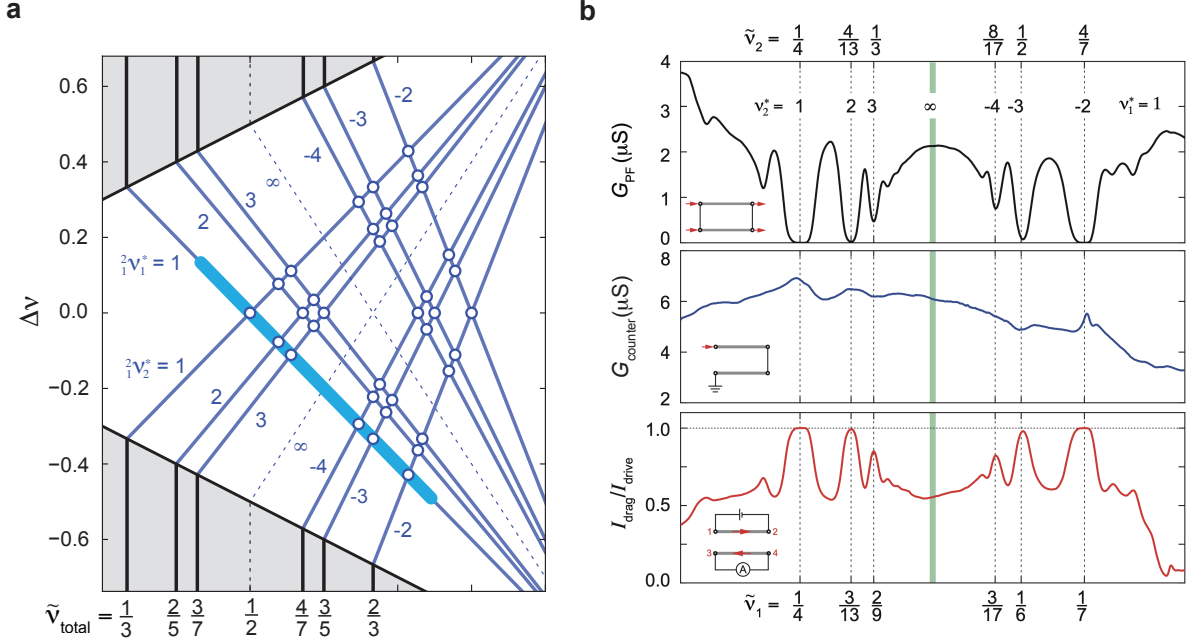


Figure 5.11:  **${}^2_1\text{CF}$  FQHE sequence with interlayer excitons.**

(a) Schematic phase diagram of  ${}^2_1\text{CF}$  states. Solid lines denote  ${}^2_1\text{CF}$  FQHE trajectories in each layer with  ${}^2_1\nu_i^* = N \in \mathbb{Z}$ . Open circles mark the intersection of these trajectories, denoting where both layer are in some  ${}^2_1\text{CF}$  FQH states. (b)  $G_{\text{PF}}$  (top),  $G_{\text{counter}}$  (middle), and  $I_{\text{drag}}/I_{\text{drive}}$  (bottom), as a function of  ${}^2_1\tilde{\nu}_2^*$  measured while fixing  ${}^2_1\tilde{\nu}_1^* = 1$ , labeled as the thick blue line in panel a. Measurements performed at  $B = 12$  T and  $T = 20$  mK. Adapted from Ref. [152]

same,  ${}^2_1\text{CF}$  construction.

Strikingly, whenever both layers are simultaneously at their  ${}^2_1\text{CF}$  FQH states, we observed excitonic pairing, evidenced by the nonzero  $G_{\text{counter}}$  and the perfect drag features as shown in Fig. 5.11c-d. The depairing of these excitons is also demonstrated Fig. 5.13a. This observed excitonic pairing is verified to be general to all  ${}^2_1\text{CF}$  FQH states by repeating the measurement at  ${}^2_1\nu_1^*$  equal to other integers with example of  ${}^2_1\nu_1^* = 2$  show in Fig. 5.13b. Although the states generally less well developed, due to the smaller gap size at  ${}^2_1\nu_1^* = 2$ , the excitonic features, such as perfect drag, still persist at  ${}^2_1\nu_2^* = 1$  and  ${}^2_1\nu_2^* = -2$ . As such, our findings suggest a general excitonic pairing order for  ${}^2_1\text{CF}$  FQHE, e.g., when  ${}^2_1\nu_1^*$  and  ${}^2_1\nu_2^*$  are both integers.

Another important subtext from the activation behaviors measurement lies in stark



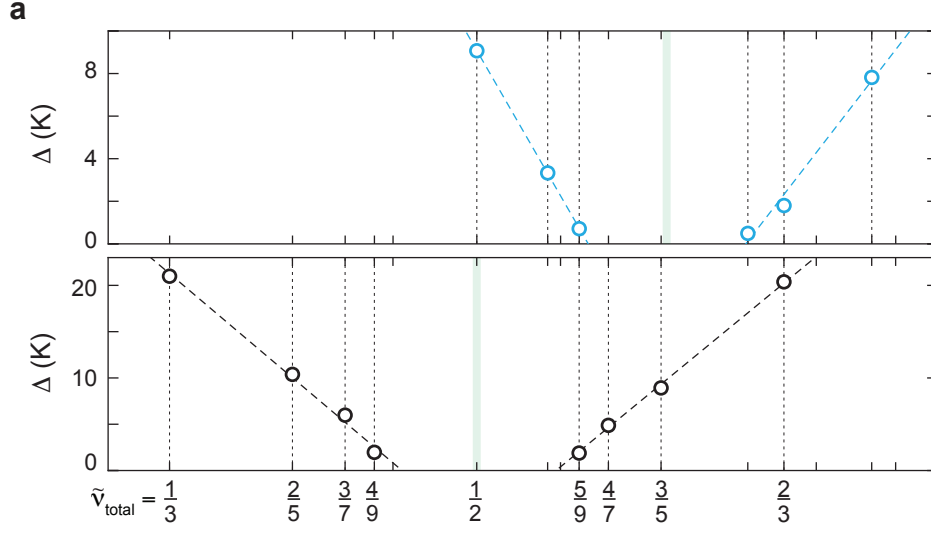


Figure 5.12: **Charge gaps at  ${}^2_0\text{CF}$  and  ${}^2_1\text{CF}$  sequences**

(a-b) Charge gap  $\Delta$ , measured as a function of filling for (a)  ${}^2_0\text{CF}$  and (b)  ${}^2_1\text{CF}$  sequences, along the same trace as in Fig. 5.8a and Fig. 5.11b, respectively. Measurements performed at  $B = 12$  T and  $T = 20$  mK. Adapted from Ref. [152]

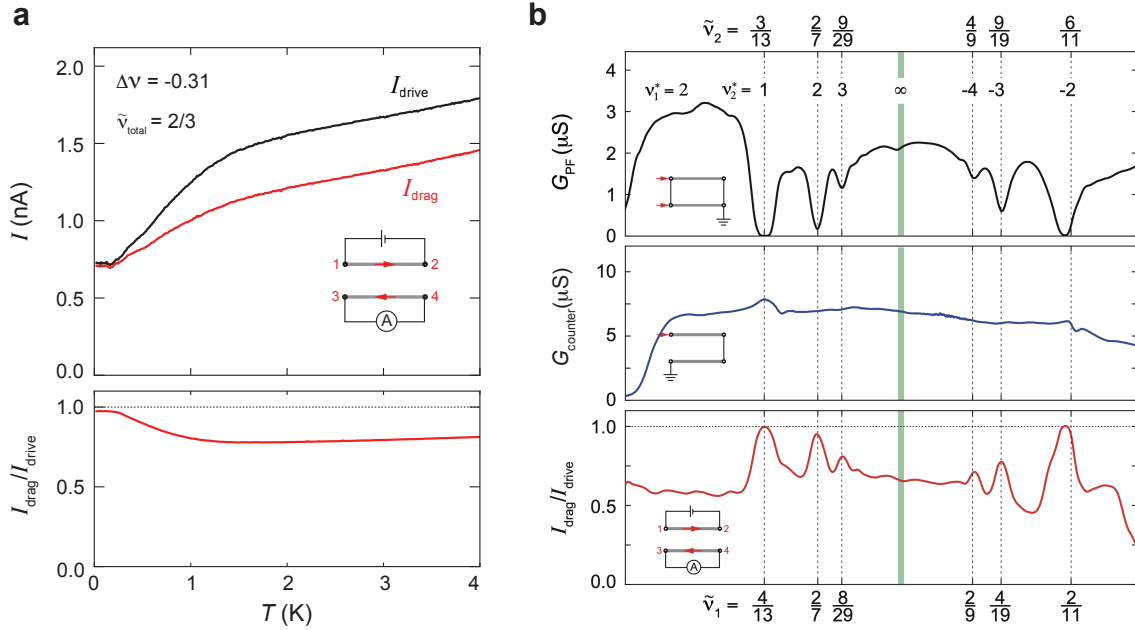


Figure 5.13: **Depairing of  ${}^2_1\text{CF}$  excitons via temperature.**

(a)  $I_{\text{drag}}$  (red) and  $I_{\text{drive}}$  (black) (top) and  $I_{\text{drag}}/I_{\text{drive}}$  (bottom) drag ratio as a function of temperature, measured at  ${}^2_1\tilde{\nu}_1^* = 1$  and  ${}^2_1\tilde{\nu}_2^* = -3$ . Demonstrating the breakdown of the excitons. (b)  $G_{\text{PF}}$  (top),  $G_{\text{counter}}$  (middle), and  $I_{\text{drag}}/I_{\text{drive}}$  (bottom), as a function of  ${}^2_1\tilde{\nu}_2^*$  measured while fixing  ${}^2_1\tilde{\nu}_1^* = 2$ . Measurements performed at  $B = 12$  T. Adapted from Ref. [152]

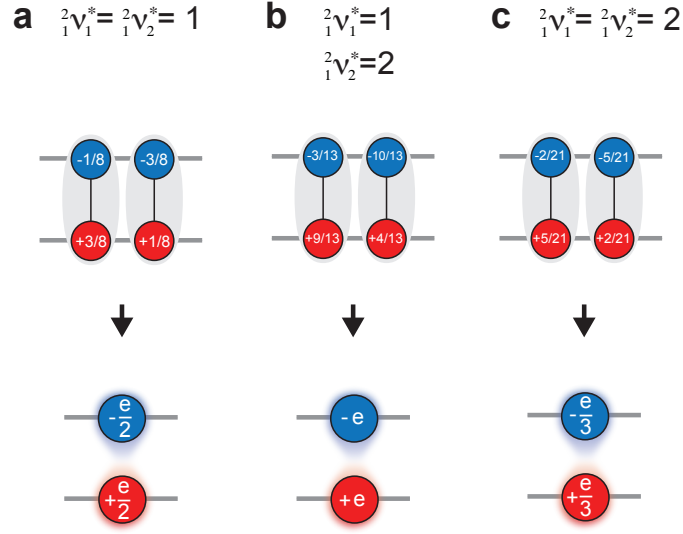


Figure 5.14: **Excitonic pairing between quasiparticle/quasihole at  ${}^2_1\text{CF}$  states.** Schematic of excitonic pairing between quasiparticle and quasihole excitations at  ${}^2_1\text{CF}$  states for (a)  ${}^2_1\nu_1^* = {}^2_1\nu_2^* = 1$ , (b)  ${}^2_1\nu_1^* = 1$  and  ${}^2_1\nu_2^* = 2$ , and (c)  ${}^2_1\nu_1^* = {}^2_1\nu_2^* = 2$ , from  $\mathbf{K}$ -matrix analysis, obeying fermionic, bosonic, and anyonic statistics, respectively. Adapted from Ref. [152]

contrast between the absolute sizes of the charge gaps, on the order of 10 K, and the base temperature of our measurement setup. The excitonic transport signature are measured at  $T = 20$  mK, orders of magnitude lower than the gaps. This means that the excitons that we observe are either the ground state or extremely low energy excitations. Moreover, these robust charge gaps formed with the  ${}^2_1\text{CF}$  FQH states means that there is no free charge left to form excitons, and likely it is the quasiparticle and quasiholes that are responsible for the excitonic pairings.

$\mathbf{K}$ -matrix formalism can be utilized to extract information about the quasiparticle/quasihole excitations at these  ${}^2_1\text{CF}$  states. For example, as illustrated in Fig. 5.14a, at  ${}^2_1\nu_1^* = {}^2_1\nu_2^* = 1$ , the system host quasiparticles with a  $-3e/8$  charge on one layer and  $+e/8$  charge on the other layer and quasiholes with a  $+3e/8$  charge and  $-e/8$  charge across the layers. Notably, an excitonic pairing between these quasiparticles and quasiholes does not yield composite bosons, as what excitons are traditionally. In fact, the pairing at  ${}^2_1\nu_1^* = {}^2_1\nu_2^* = 1$  gives rise to a new type of exciton obeying fermionic statistics. In a

similar fashion, the charge distribution of quasiparticle/quasihole across the layer can be calculated at any other  ${}^2_1\text{CF}$  states. In particular, when  ${}^2_1\nu_1^* = 1$  and  ${}^2_1\nu_2^* = 2$ , quasiparticles are determined to have  $+9e/13$  and  $-3e/13$  across the layers, while quasiholes have  $+4e/13$  and  $-10e/13$  across the layers. The pairing at this state thus resulting in excitons with a net charge of  $\pm e$  on each layer, obeying bosonic statistics. Worth to note that, the breakdown of such exciton would result in the fractionally charged quasiparticles and quasiholes, as oppose to an unbounded electron-hole pair. Remarkably, at  ${}^2_1\nu_1^* = {}^2_1\nu_2^* = 2$ ,  $\mathbf{K}$ -matrix analysis predicts quasiparticles and quasiholes with  $\pm 2e/21$  and  $\mp 5e/21$  charge across the layers, together giving rise to excitons with  $\pm e/3$  charge across the layers, following anyonic statistics with a statistical phase of  $\theta = 4\pi/3$  upon exchange.

Together, in the two-component fractional quantum Hall effect with interlayer coupling, we observe two types of excitons. The first, manifested at the  ${}^2_2\text{CF}$  FQH sequences, behaves as the fractional analogue of the *exciton condensate* at total filling of one. Whereas the second, appearing at the  ${}^2_1\text{CF}$  FQH sequences, is predicted to follow *fermionic*, *bosonic*, and *anyonic* statistics.

# CHAPTER 6

## Excitons in decoupled fractional quantum Hall effects

### 6.1 Layer decoupled FQHE

In the previous chapter, we extensively explored excitonic behaviors arising in two component FQHE, specifically those described by the  ${}^2_1\text{CF}$  and  ${}^2_2\text{CF}$  constructions. Owing to the inherent interlayer coupling present in these states, it is not unfathomable that they would host interlayer excitonic neutral modes. However, a more subtle scenario emerges when considering fractional quantum Hall states without interlayer flux attachment, where excitonic behavior cannot be trivially presumed. In this section, we will delve into the excitonic phenomena within layer-decoupled fractional quantum Hall effects. We will begin by examining a peculiar observational conundrum.

Similar to our approach for tackling  ${}^2_1\text{CF}$  states, we hold one layer at a constant  ${}^2_0\text{CF}$  effective filling factor  ${}^2_0\nu_1^* = 1$ , and measure the transport behavior as a function of the effective filling in the other layer. When two layers are decoupled from each other, we would expect  $G_{\text{PF}}$  to exhibit the classic Jain sequence behavior, depicting the charge gaps developed at each states at  ${}^2_0\nu_2^* = N \in \mathbb{Z}$ , as well as vanishing  $G_{\text{counter}}$  and

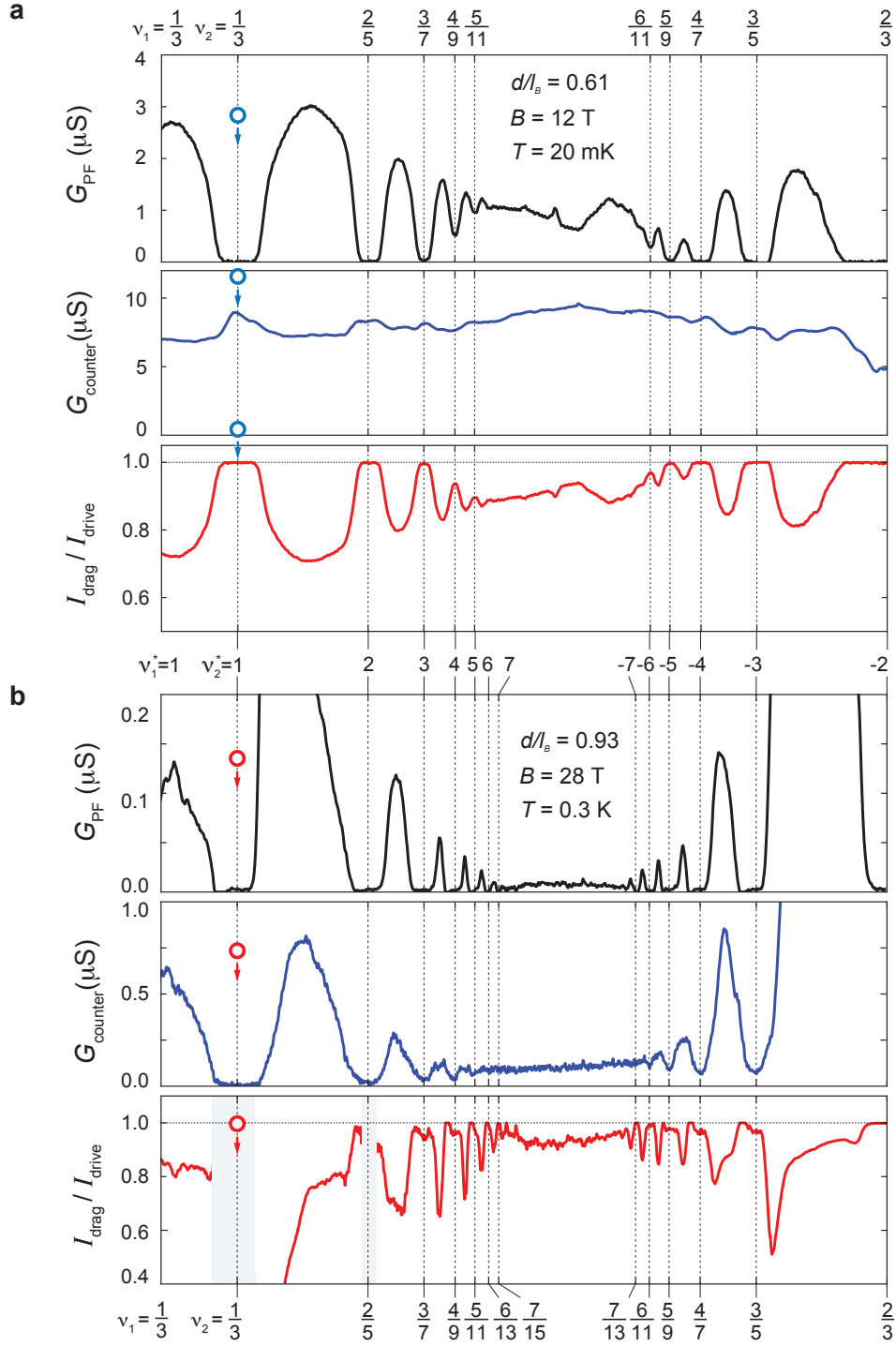


Figure 6.1: **Transport signatures along  $\nu_1 = 1/3$ .**

(a-b)  $G_{\text{PF}}$  (top),  $G_{\text{counter}}$  (middle) and  $I_{\text{drag}}/I_{\text{drive}}$  (bottom) measured as a function of  $\nu_2$ , along  $\nu_1 = 1/3$  at (a)  $B = 12 \text{ T}$  and (b)  $B = 28 \text{ T}$ . The red and blue circles highlights the change in excitonic signatures at  $\nu_1 = \nu_2 = 1/3$ . (a) is measured at  $T = 20 \text{ mK}$ , (b) is measured at  $T = 300 \text{ mK}$ . Adapted from Ref. [153]

ill-defined  $I_{\text{drag}}/I_{\text{drive}}$ , resulting from the highly layer-localized Laughlin wavefunctions and consequent absence of interlayer excitonic pairing, similar to the case shown in Fig. 5.11a. However, to our surprises, as clearly demonstrated in Fig. 6.1a, we observe signature excitonic transport behaviors, including finite  $G_{\text{counter}} > 0$  and perfect drag  $I_{\text{drag}}/I_{\text{drive}} = 1$ , as if the two layers are coupled. This unusual observation poses important questions regarding the nature of these excitons, as we will investigate in the next section.

## 6.2 Excitons in ${}^2_0\text{CF}$ states with thermal excitation

For our graphene/hBN quantum Hall bilayer with  $d = 4.5$  nm, a magnetic field of 12 T yields a  $d/l_B = 0.61$ . This is certainly on the smaller side for the FQHE to be layer-dissociated, as observed along  $\nu_{\text{total}} = 1$  in Ref. [171] (see Fig. 6.2). While our identification of the  ${}^2_0\text{CF}$  states are evidenced by the remarkable alignment with the expected location of these state in the  $\nu_{\text{total}}-\Delta\nu$  plane from the model (see Fig. 5.6a and 5.7), their presence at such a small  $d/l_B$  is somewhat unsettling and requires further justification than the screening reasoning alone. An ease check is to directly compare the behaviors at a larger  $d/l_B$ . As shown in Fig. 6.1, one key observation is highlighted by the contrast between the  $B = 12$  T measurement and the  $B = 28$  T measurement: Upon increasing  $d/l_B$ , while the sequence of the insulating features becomes more robust, many excitonic features, for example at  $\nu_1 = \nu_2 = 1/3$ , disappear.  $G_{\text{counter}}$  vanishes at  $d/l_B = 0.93$ , and  $I_{\text{drag}}/I_{\text{drive}}$  becomes ill-defined.

This comparison can be explained by two possible mechanisms: One, as we increase  $d/l_B$ , a transition from interlayer coupled states to interlayer decoupled states happened, and our identification of the CF constructions at  $B = 12$  T was hindered by some competing states at the same locations. Two, thermal excitation is responsible for generating excitons at  $\nu_1 = \nu_2 = 1/3$  at  $B = 12$  T. Since the gap size of LLs and  $\Lambda$ -levels are positively related to  $B$ , this scenario points towards an interesting case where the charge gaps between the Laughlin states can be overcome by the quasiparticle/quasiholes at the measurement

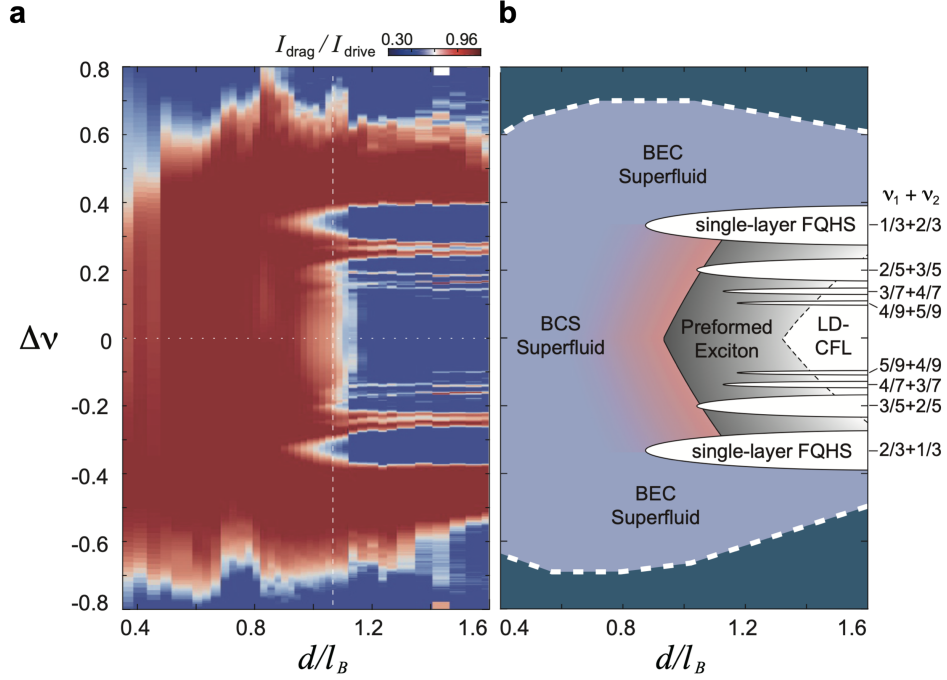


Figure 6.2: **Phase diagram at  $\nu_{\text{total}} = 1$ .**

(a)  $I_{\text{drag}}/I_{\text{drive}}$  and (b) schematics of states in the  $d/l_B$ - $\Delta\nu$  phase diagram. Clear change in  $I_{\text{drag}}/I_{\text{drive}}$  highlights the change in interlayer coupling. In this work,  $d/l_B = 0.6$  would correspond to layer-coupled states,  $d/l_B > 1$  would correspond to a layer-decoupled states. However note that at  $\nu_{\text{total}} = 1$ , the layer-coupled state is a much more robust, two-component QHE with integer filling. Adapted from Ref. [171]

temperature at  $B = 12$  T whereas the charge gaps at  $B = 28$  T become too large for temperature induced excitation. While both mechanisms phenomenologically resolve the conundrum, we claim our observation is most possibly due to the second for the following reasons.

First, it is unlikely that for each expected Jain state of  ${}^2_0\text{CF}$  construction, there is competing state at  $B = 12$  T, resulting in an nearly exact replica of the observed Jain sequence, with the correct hierarchy of the charge gaps, which are indicated by the widths of the insulating features in Fig. 6.1a. The seemingly small  $d/l_B = 0.61$  compared to the literature can be explained by the competition at  $\nu_{\text{total}} = 1$ , which is absent at all states that we show in Fig. 6.1a, with the exception of  $\nu_2 = 2/3$ .

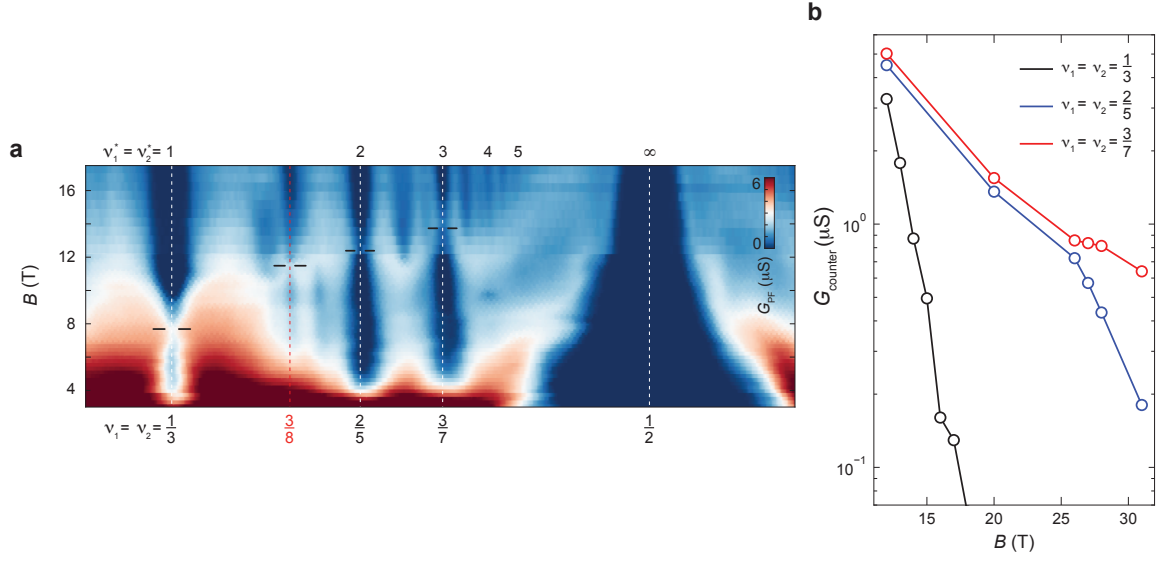


Figure 6.3:  **$B$ -dependence of the  ${}^2_0\text{CF}$  states.**

(a)  $G_{\text{PF}}$  as a function of  $B$  and filling, along  $\nu_1 = \nu_2$ . Transitions along  $B$  are marked by black dashes at each filling. (b)  $G_{\text{counter}}$  as a function of  $B$ , measured at  $\nu_1 = \nu_2 = 1/3$  (black),  $\nu_1 = \nu_2 = 2/5$  (blue),  $\nu_1 = \nu_2 = 3/7$  (red). All data measured at  $T = 300$  mK. Adapted from Ref. [153]

Second, As we demonstrate in Fig. 6.3a, the pinches in the insulating features, marked by the black dashes, indicate transitions between competing states as a function of  $B$ . At  $\nu_1 = \nu_2 = 1/3$ , no similar feature is observed beyond  $B = 8$  T, indicating that it is likely the same state which persists from 8 T to 28 T. Since at  $B = 28$  T, we can confirm layer decoupling by the absence of excitonic transport features, this lack of transitioning evidences that it is the same layer-decoupled state that we observe at  $B = 12$  T.

Third, comparing the transport behavior at different states at  $B = 28$  T, we observe an interesting evolution: As  $\nu_2^*$  increases, the charge gaps become less robust, evidenced by the shrinking width of  $G_{\text{PF}} = 0$  features, while the excitonic features become more robust, evidenced by the perfect drag at higher  $\nu_2^*$ . This means that at  $B = 28$  T, the larger charge gaps, at for example  $\nu_2 = 1/3$  and  $\nu_2 = 2/5$ , are too large to be overcome, whereas the gaps at weaker states like  $\nu_2 = 5/11$  and  $\nu_2 = 6/13$  are still small enough for  $T = 300$  mK.

Along the same vein, we show  $G_{\text{counter}}$  as a function of  $B$  at three different  ${}^2_0\text{CF}$  states



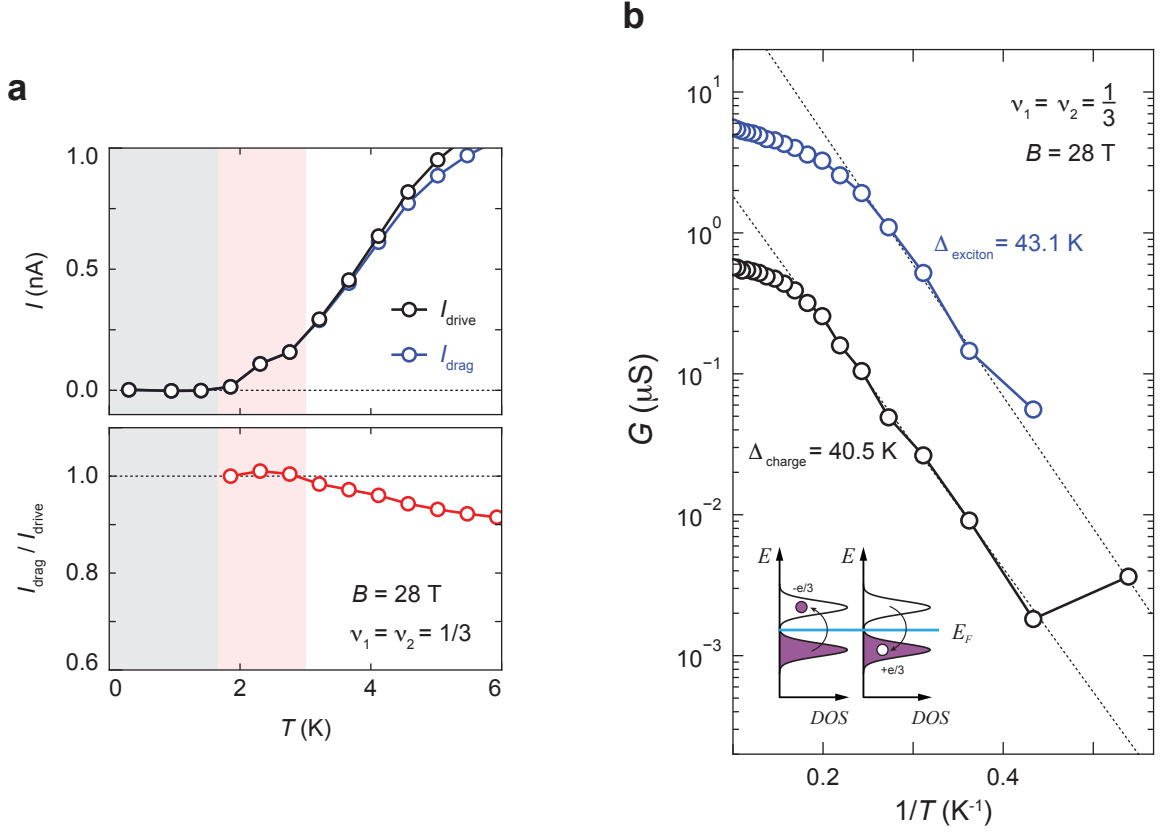


Figure 6.4:  **$T$ -dependence of the  $\nu_1 = \nu_2 = 1/3$  state.**

(a)  $I_{\text{drive}}$  and  $I_{\text{drag}}$  (top) and their ratio (bottom) as a function of  $T$  at  $\nu_1 = \nu_2 = 1/3$  at  $B = 28$  T. (b) Arrhenius plot of  $G_{\text{PF}}$  (black) and  $G_{\text{counter}}$  (blue), showing the similar energy gaps between charge and excitonic pairing, offset is made for clarity. Inset: a schematic for thermally generating excitons. Adapted from Ref. [153]

in Fig. 6.3b. For  $\nu_1 = \nu_2 = 1/3$ , which has the largest gap among the three,  $G_{\text{counter}}$  is suppressed at a relatively smaller magnetic field  $B \approx 15$  T. For the intermediate  $\nu_1 = \nu_2 = 2/5$  state, the  $G_{\text{counter}}$  vanishes at  $B \approx 30$  T. And for the smallest gap at  $\nu_1 = \nu_2 = 3/7$ ,  $G_{\text{counter}}$  persists beyond the limit of our magnet at  $B = 31$  T.

Lastly, if the excitons are truly due to thermal excitations, they should re-emerge at  $B = 28$  T when temperature is increased. Indeed as shown in Fig. 6.4a, we observe two transitions as we increase the temperature at  $B = 28$  T. At low temperature, there is no current in either the drag or drive layer, indicating robust charge gaps in each layer. At intermediate temperature, both  $I_{\text{drag}}$  and  $I_{\text{drive}}$  onsets, give rise to a drag ratio  $I_{\text{drag}}/I_{\text{drive}} = 1$ . Then at high temperature,  $I_{\text{drag}}$  and  $I_{\text{drive}}$  start to bifurcate, reducing

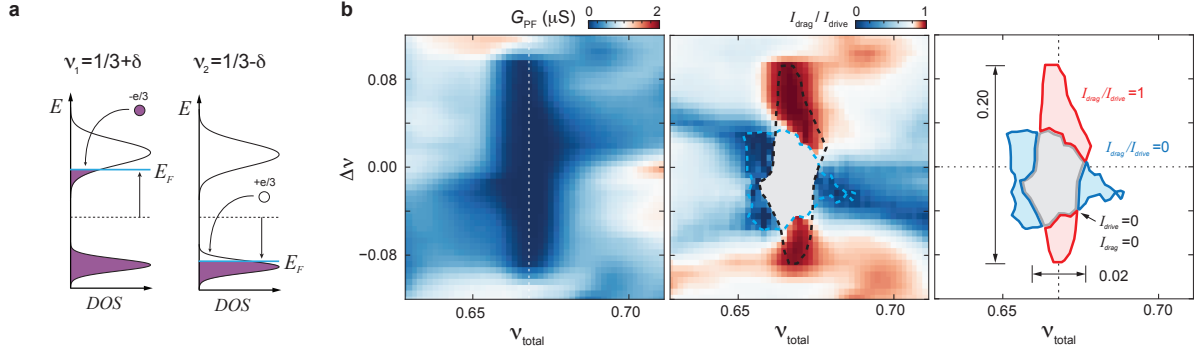


Figure 6.5: **Electrostatic doping induced excitons.**

(a) Schematic of generating excitons using layer asymmetric doping. (b)  $G_{PF}$  (left),  $I_{drag}/I_{drive}$  (middle), and a schematic (right) around  $\nu_1 = \nu_2 = 1/3$ . Grey marks the layer-decoupled region with charge gap and no exciton. Red in marks the region with perfect drag and charge gap, indicating exciton formation. Blue marks the region with charge gap closing without exciton. All data measured at  $T = 300$  mK. Adapted from Ref. [153]

$I_{drag}/I_{drive}$  from 1, indicating the depairing of the excitons. Further extracting the gaps from the activation behavior, as seen in Fig. 6.4b, we see that the charge gap and the exciton gap are essentially the same at  $\nu_1 = \nu_2 = 1/3$  at  $B = 28$  T, suggesting that quasiparticle/quasihole and excitonic neutral modes are generated by the same thermal process.

Together, our experiment evidences the second hypothesis that the observed excitonic transport behaviors arise from pairings between thermally generated quasiparticles and quasiholes. As such, these excitons, composed of fractionally charged constituents, are composite, *charge neutral anyons*.

### 6.3 Excitons in ${}^2_0\text{CF}$ states with electrostatic doping

Naturally we would like to ask next is, can we create excitons with other mechanisms in these layer decouple states? If populating quasiparticles and quasiholes via thermal excitation can induce excitonic pairing, similar neutral modes should arise by directly doping equal amount of quasiparticles and quasiholes across the layer, as illustrated in Fig. 6.5a. Indeed as shown in Fig. 6.5b, moving away from  $\nu_1 = \nu_2 = 1/3$  along  $\Delta\nu$ ,

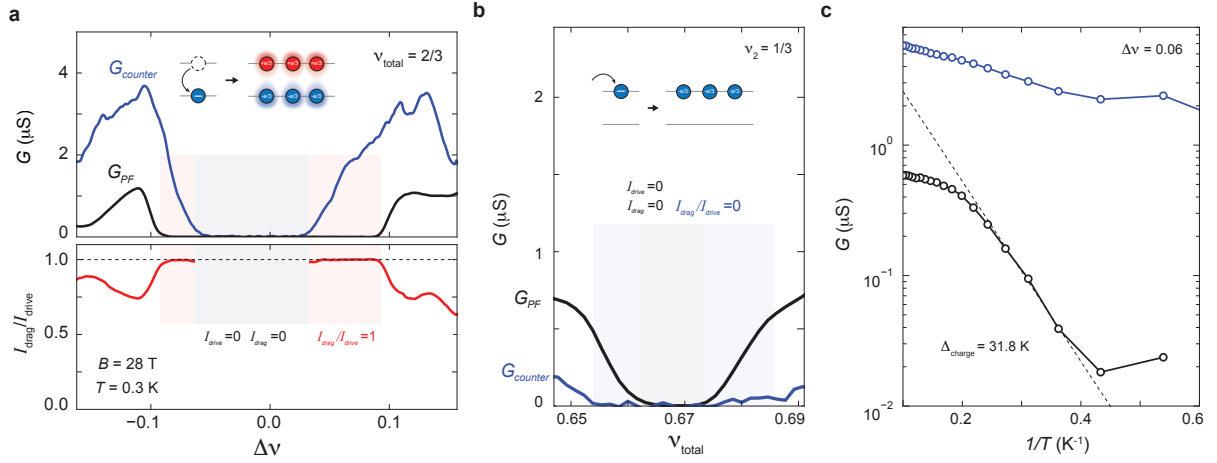


Figure 6.6: **Layer asymmetric vs layer symmetric doping.**

(a)  $G_{\text{PF}}$  (top black),  $G_{\text{counter}}$  (top blue), and  $I_{\text{drag}}/I_{\text{drive}}$  (bottom) as a function of  $\Delta\nu$ . (b)  $G_{\text{PF}}$  and  $G_{\text{counter}}$  as a function of  $\nu_{\text{total}}$ . Gray shades mark the decoupled region with charge gap and no exciton, red shades mark the regions with exciton doping, and blue shade mark the regions with gap closing. (c) Arrhenius plot for  $G_{\text{PF}}$  and  $G_{\text{counter}}$  measured at  $\nu_1 = 1/3 + 0.03$ ,  $\nu_2 = 1/3 - 0.03$ , highlighting the large charge gap with low temperature excitonic feature. All data measured at  $B = 31$  T and  $T = 300$  mK. Adapted from Ref. [153]

we recover the neutral mode while maintaining a robust charge gap, evidenced by the vertically elongated  $G_{\text{PF}} = 0$  feature and the perfect drag above and below the  $I_{\text{drag}}/I_{\text{drive}}$  ill-defined region at  $\nu_1 = \nu_2 = 1/3$ . On the other hand, moving along  $\nu_{\text{total}}$  would not generate excitonic pairing, as evidenced by the absence of Coulomb drag signal on the left and right side of  $\nu_1 = \nu_2 = 1/3$ , as well as the breaking of the FQH state, evidenced by the nonzero  $G_{\text{PF}}$ . This is reasonable as moving in  $\Delta\nu$  direction, we are essentially doping the two layers with opposite amount of charge, essentially creating the same physical picture as the inset of Fig. 6.4b. But moving in the  $\nu_{\text{total}}$  direction, we are doping two layer with same amount of charge, yielding mismatched quasiparticle/quasihole populations and these unpaired charges eventually cause the gap to close. Fig. 6.6a-b shows the linecuts along each direction, demonstrating the picture with better clarity. Fig. 6.6c shows the temperature dependence of  $G_{\text{PF}}$  and  $G_{\text{counter}}$  at  $\nu_1 = 1/3 + 0.03$  and  $\nu_2 = 1/3 - 0.03$ , demonstrating the charge gap at low temperature as well as the unique electrostatic doping of anyonic excitons.

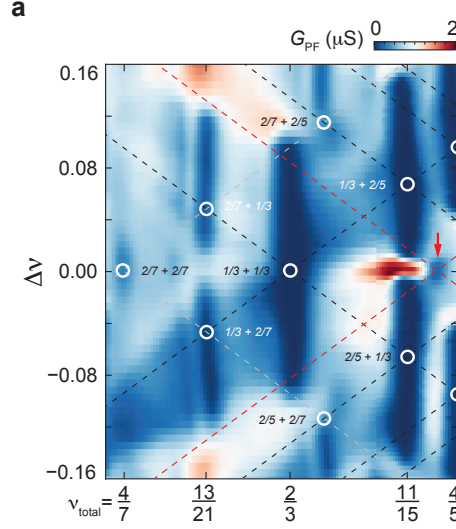


Figure 6.7:  $G_{\text{PF}}$  around an even denominator state.

(a)  $G_{\text{PF}}$  as a function of  $\nu_{\text{total}}$  and  $\Delta\nu$ , Standard Jain sequence states are marked by black dashed lines and white circles. The even denominator state at  $\nu_1 = \nu_2 = 3/8$  is marked by red dashed lines and a red arrow. Measured at  $B = 31\text{T}$  and  $T = 300\text{ mK}$ . Adapted from Ref. [153]

## 6.4 Excitons at even denominator states

As already shown in Fig. 6.3, we observe an even denominator state at  $\nu_1 = \nu_2 = 3/8$ . We assign this state to a layer decoupled CF construction for the following reasons. Examining the surroundings of this state in the  $\nu_{\text{total}}-\Delta\nu$  phase diagram, we observe well organized other layer independent states following the expected Jain sequence (see Fig. 6.7. Second, the pinching in Fig. 6.3 demonstrates a transition at  $B \approx 12\text{ T}$ , naturally describing decoupling between the layers, assigning the high- $B$  state to the single component Jain sequence.

Remarkably, this layer-decoupled, even denominator state also exhibits clear excitonic behavior, with a sizable  $G_{\text{counter}}$  and a  $I_{\text{drag}}/I_{\text{drive}}$  that is nearly 1 (see Fig. 6.8). Previous numerical simulations have identified a Bonderson-Slingerland-like wavefunction as the ground state at  $\nu = 3/8$ , within a single 2D confinement [172–175]. These results highlight a scenario where the CFs undergo Cooper pair instability and condense into a  $p$ -wave paired state. That is to say, the bulk quasiparticle/quasihole excitations at  $\nu = 3/8$

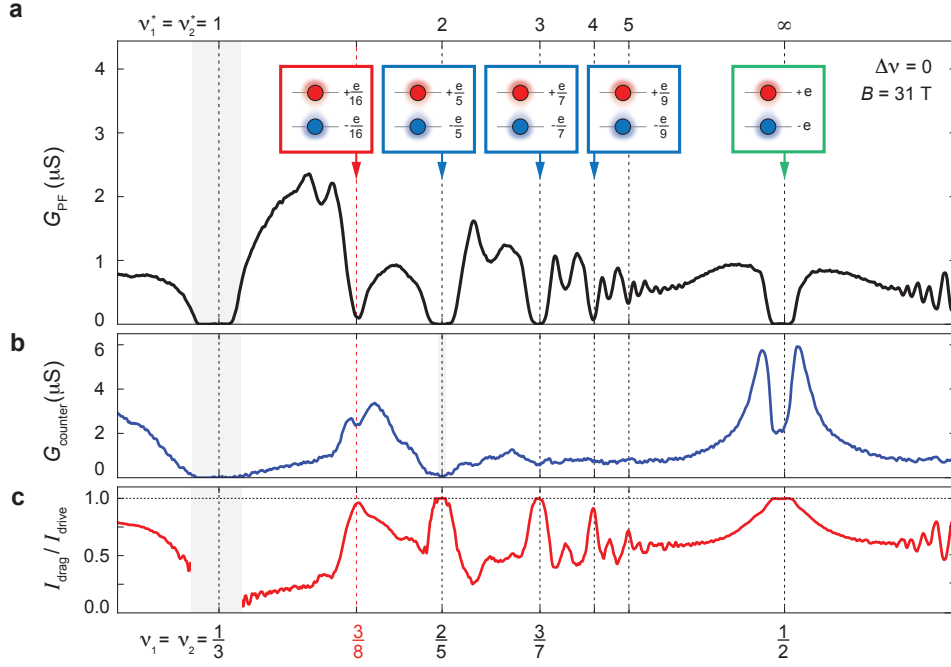


Figure 6.8: **Excitons at an even denominator state.**

(a)  $G_{PF}$ , (b)  $G_{counter}$ , and (c)  $I_{drag}/I_{drive}$  as a function of filling, along  $\nu_1 = \nu_2$ . Inset: constituents of different types of anyonic excitons, including bosonic (green), anyonic (blue), non-Abelian (red). Measured at  $B = 31\text{ T}$  and  $T = 300\text{ mK}$ . Adapted from Ref. [153]

likely obey non-Abelian statistics, which arises from the non-trivial fusion rules between the distinct topological sectors. Following this argument, we expect the excitons paired between these quasiparticles and quasiholes also obey *non-Abelian anyonic* statistics, with an unusual overall charge neutrality.

# CHAPTER 7

## Epilogue

### 7.1 Topping out

This thesis has explored a broad spectrum of quantum phenomena in low-dimensional, strongly correlated electron systems, using transport experiments as a unifying investigative tool.

In twisted trilayer graphene samples, we employed angle-resolved transport measurements to uncover subtle symmetry breaking behaviors, ranging from nematic metallic phase and superconductivity to non-reciprocal transport arising from inversion symmetry breaking. These results highlight the moiré platform’s tunability to interaction-driven anisotropies, and the utility of transport measurements in resolving them.

Beyond those moiré-specific phases, this planar, rotationally symmetric, sunflower device architecture has also demonstrated how novel experiment design can offer unparalleled sensitivity to the symmetry breaking orders themselves. We showcase this empowerment with a complete framework for extracting conductivity tensors up to second order in current and the first unambiguous experimental observation of nonlinear Hall effect in a Bernal bilayer graphene. While developed in the context of low dimensional electronics,

its utility extends far beyond a single set of material systems. By providing a general platform for probing the conductivity tensors, the sunflower architecture offers a powerful and flexible tool for exploring correlated states across a wide range of systems—opening new pathways for experimental discovery well beyond the scope of this work.

Equally central to this work are the quantum Hall bilayer experiments, where the competition between intra- and inter-layer coupling unlocks a wide variety of exotic collective states. Enabled by the careful layer-sensitivity transport design, we observe excitonic behaviors from the interlayer correlated regime to the interlayer decoupled limit, reflecting the richness of interaction-driven physics in multi-component quantum Hall systems. These experiments underscore how vertically layered architectures can not only host entirely new categories of correlated quantum matter, but also allow the direct experimental observation over them.

Across the systems studied in this thesis, a recurring theme is the emergence of complex collective behavior from the interplay of symmetry and interaction. In strongly correlated materials, microscopic details often give way to macroscopic orders that reflect not the constituents themselves, but the constraints imposed by symmetry and the organizing power of many-body interactions. These emergent phenomena, whether they exhibit superconductivity or quantum Hall effect, are not hardwired into the lattice structure, but arise dynamically as the system seeks a new ground state under competing forces.

Symmetry plays a dual role in this landscape. It shapes not only how these orders manifest themselves, but also fundamentally governs what kinds of orders are allowed to exist. Spontaneous symmetry breaking therefore signals a reorganization of the system’s internal logic. In this way, a straightforward combination between different symmetry preserving or breaking orders can easily yield a remarkably rich phase diagram, where delicate competitions between interactions give rise to entirely new forms of order. Understanding this interplay between symmetry and interaction remains one of the central challenges—and greatest fascinations—of modern condensed matter physics.

## 7.2 Scene from the roof

Echoed by Albert Einstein, Karl Popper’s principle of falsifiability stands as the cornerstone of modern science [176, 177]. All theories, disregarding their elegance or predictive ability, are only provisionally true in an absolute sense: A theory may withstand the tests imposed by successive experiments, but with each attempt, we push it to the edge of its applicability, seeking not confirmation, but contradiction. It is this spirit of critical testing that propels scientific progress forward.

In this light, experiment is not a process with a final destination, but an open-ended endeavor—one that must be repeated, refined, and re-imagined as our questions deepen and our tools improve. Adapting from Camus’ image of Sisyphus, I view the work of being an experimentalist as forever pushing the stone uphill, not in futility, but in resilience. Each iteration builds toward something greater: a clearer understanding, a tighter constraint, a newly exposed layer of physical law. The work is never finished, and that is precisely what makes it meaningful and truly heroic.

Several directions naturally arise from the results presented in this thesis. On the nematic orders in graphene moiré systems, a direct step forward is to control the strength of the interaction via Coulomb screening, examining how anisotropy evolves regarding the change. In the context of 2D materials, such experimental knob has been demonstrated in Ref. [68, 72]. By tuning the band gap of a closely placed BBG layer, one can realize an *in situ* control over the Coulomb screening effect, thus actively modifying the interaction strength, further evidencing for or against the picture of Coulomb interaction as the underlying mechanism for anisotropy. If each sunflower contact can be made into a tunneling junction, a phase-sensitive measurement for quantitatively probing the superconductivity pairing order is also achievable [178, 179]. One could imagine a sunflower device in which each lead splitting into two: one forming an Ohmic contact with gold to probe the principle axis of the nematicity with angle resolved transport, and the other forming a superconducting-insulating-superconducting junction using a thin hBN tunneling barrier



between the exposed graphene edge and a superconducting aluminum electrode. Since the sunflower design and the general measurement scheme is readily available for adaptation, it is also intuitive to examine the rotational symmetry of other systems, including but not limited to the highly celebrated rhombohedral multilayer graphene [180–182], newly realized bulk superconductors like nickelates [183], or semiconducting heterostructures if Ohmic contacts can be achieved [184–187]. The rotational symmetry of a large variety of samples and states is yet to be examined.

On the other hand, the double Corbino geometry only probes the bulk of quantum Hall bilayer samples. The edge mode and its anisotropy could be examined with a double sunflower design. Naturally four-terminal, the sunflower design will also enable the direct observation of a diverging counterflow conductance if present, further examining the condensate nature of the fractional excitons [85]. Combining the sunflower geometry with a center Corbino contact, the anisotropy of the excitons residing in the bulk can also be probed by checking the angle dependence of different instabilities, such as critical temperatures and depairing d.c. voltages. As such, the pairing symmetry of the fractional excitons may be revealed with some modeling. Further thinning the hBN spacer, quasiparticle tunneling across the layers can also be examined, in the hope of demonstrating a resonant tunneling and therefore interlayer coherence [84]. By changing each monolayer graphene into a Bernal bilayer graphene, one can also introduce single-layer even denominator states, further promoting the pairing between exotic quasiparticles and quasiholes [188, 189]. An excitonic Josephson junction is also within the theoretical prediction, requiring only a split top gate [190].

But ultimately, the future of these research projects lies not only in refining existing techniques or revisiting known phenomena, but in continuing to ask how symmetry, interaction, and emergence shape the quantum world. These three pillars frame our current understanding of correlated matter, and they will undoubtedly remain central as we probe new materials, uncover novel phases, and strive to connect abstract theoretical

frameworks with concrete experimental signatures. The questions deepen, the terrain expands—but the guiding ideas endure.

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# Appendices

# Appendix A

## Experiment scheme for angle resolved transport measurement

Here I give an overview on the scheme for angle resolved transport measurement using the sunflower geometry. The setup for measuring  $V_{\parallel}$  and  $V_{\perp}$ , labeled as  $I$  and  $II$ , are shown in Fig. A.1a. With the naive assumption of current always flows from source to drain, we can define the current flow direction  $\phi$  by the angle of the segment connecting source and drain. As such, by rotating all contact by 1 sample lead, we can increase  $\phi$  by  $45^\circ$ , yielding us a total of 8 angular entries for each channel.

Now to get a bit more general, we can think of a few different measurement configurations that can also be considered “longitudinal” or “transverse” per se. As shown in Fig. A.2, each of the configurations on the top row have segments connecting current source and drain parallel to the segments that connect the voltage leads. Similarly in the bottom row, the segments are perpendicular to each other. Each of these configurations would also give rise to 8 angular entries using the same logic. Next we discuss how are they comparable to each other.

Clearly, the setup  $I'$  here is just the mirror image of the setup for  $I$ , and we would

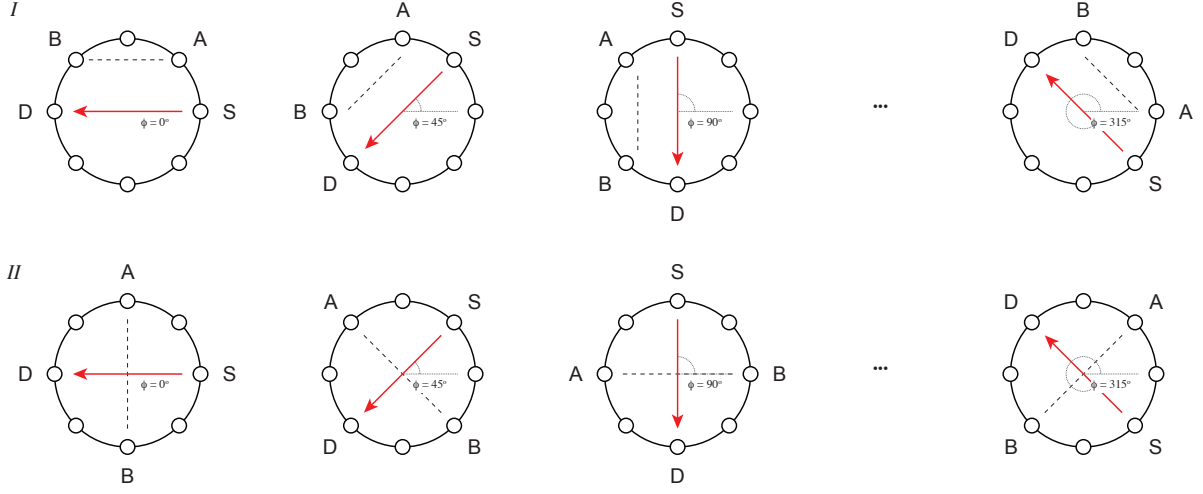


Figure A.1: **Angle dependence measurement configurations.**

Measurement configurations *I* (top) and *II* (bottom) for angle dependence transport. S,D,A,B labels the leads used for current source and drain as well as voltage contacts. Red arrows, connecting source to drain, denote the direction of the current flow. Black dashed lines connect the voltage contacts. The relative orientation between red and black lines illustrates that *I* is a sunflower analogue of the longitudinal measurement in Hall bar geometry and *II* is a sunflower analogue of the transverse measurement. Gray dotted lines mark the direction of  $0^\circ$  in the polar coordinate and the gray dotted arcs mark the azimuth angle of the current flow  $\phi$ . From left to right in each row, all four contacts are rotated by 1 lead at a time, yielding an angular separation of  $45^\circ$  in  $\phi$ .

expect the angle dependence of *I* and *I'* to behave roughly the same, given that sample is uniform. As such, one can take the average of *I* and *I'* to reduce both the noise in measurement and the non-uniformity of contacts. Comparing *III* and *I*, there are two observations. First, the segment connecting current source and drain in *III* is  $22.5^\circ$  apart from *I*. Second, the length of the segment that connects the voltage leads in *III* is longer than that in *I*. As such, the angle dependence of *III* gives rise to the points that are  $22.5^\circ$  away from the points in *I* and requires a small geometric scaling  $g < 1$ . Similar to *I'*, there is also a mirror version of *III*, denoted *III'*, which can help with the measurement noise and the contact non-uniformity.

Along the same vein, *IV* is somewhat analogous to *II*, with a  $22^\circ$  separation in  $\phi$  and a geometric scaling factor  $g > 1$  due to shorter segment between voltage contacts. It also acquires a small mixing from  $V_{||}$ , making it generally higher than *II*. Interestingly, another

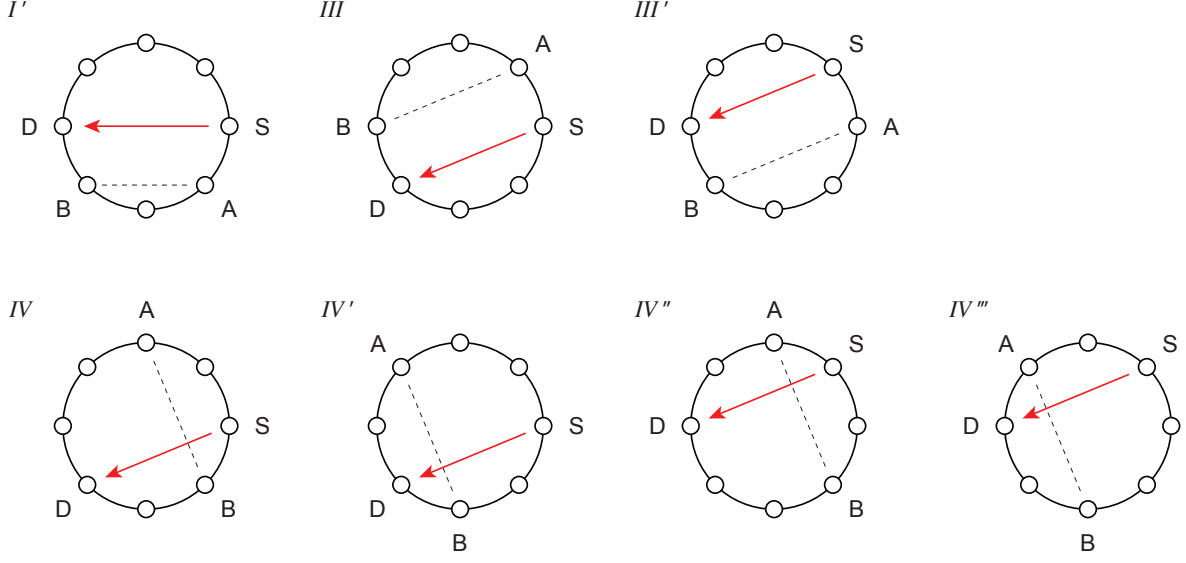


Figure A.2: **More angle dependence measurement configurations.**

Measurement configurations  $I'$ ,  $III$ ,  $III'$ ,  $IV$ ,  $IV'$ ,  $IV''$ , and  $IV'''$ . S,D,A,B labels the leads used for current source and drain as well as voltage contacts. Red arrow denotes the direction of the current flow. The  $III$  and  $IV$  series give rise to  $22.5^\circ$  angular entries.

version of  $IV$ , shown as  $IV'$ , acquires the same  $V_{\parallel}$  mixing with an opposite sign. Thus the average between  $IV$  and  $IV'$  conveniently cancels the mixing contribution. Their mirror configurations,  $IV''$  and  $IV'''$ , sharing the same properties, can also be used to increase the measurement validity.

Altogether, we usually employ three kinds of angle dependence measurement schemes. For the first kind, we measure only  $I$ ,  $I'$ , and  $II$ . This gives us 8 angular entries in  $V_{\parallel}$ , resulted from averaging  $I$  and  $I'$ , and 8 angular entries in  $V_{\perp}$  from configuration  $II$ . The accessible  $\phi$  are  $0^\circ$ ,  $45^\circ$ ,  $90^\circ$ , etc. Second, we measure only the groups of  $III$  and  $IV$ . This gives us 8 angular entries in  $V_{\parallel}$  from averaging  $III$  and  $III'$ , and 8 angular entries in  $V_{\perp}$  from averaging  $IV$ ,  $IV'$ ,  $IV''$  and  $IV'''$ . The accessible  $\phi$  are  $22.5^\circ$ ,  $67.5^\circ$ ,  $112.5^\circ$ , etc. Third, we combine the first and second schemes, obtaining a larger accessible range for  $\phi$ :  $0^\circ$ ,  $22.5^\circ$ ,  $45^\circ$ , etc. If only a rough qualitative comparison between the anisotropy states is needed, the fact that  $III$  and  $IV$  scales oppositely from  $I$  and  $II$ , makes the geometric scaling not as important, since they constrain the fit function oppositely. As a result, operationally, we take the geometric scaling for  $III$  and  $IV$  as 1 if they enters the fit

Eqn. 2.2 - 2.3 together.

In the case where an quantitative comparison is required, an extraction of the matrix is encouraged. In this case, we measure as many voltages as possible with any contacts being the current source and drain, and voltage leads. This yields a data set structured in a  $N \times 6$  matrix, where  $N$  is the number of measurement performed, and the 6 columns each represents: the locations of current source  $S$ , drain  $D$ , voltage lead  $A$ , and  $B$ , the voltage measured  $V_{SDAB}$ , and the current measured  $I_{SD}$ . All of these six columns will serve as the input for the extraction. A python script for the extraction and its description are provided in the next appendix chapter.

## Appendix B

# Python code for full extraction of linear conductivity matrix

As demonstrated in section 2.5, the sunflower geometry allows a new framework for extracting the general linear conductivity matrix. Below I show my code to realizing the extraction in Python.

Firstly, here are the functions I use to calculate the voltage for a given measurement configuration and given conductivity matrix. Based on theory in Ref. [111], I wrote this function to calculate the potential,  $\Phi_{SDA}$ , at point  $A$  on the circumference of the disk with current source and drain located at  $S$  and  $D$ . It takes in total 7 variables, three for the locations, four for the conductivity matrix.

```
def potentialOnDisk(S,D,A,theta0,sigma1,sigma2,sigmaH):  
    # theta0 = alpha, sigma_xx = sigma1, sigma_yy = sigma2, and sigma_Hall = sigmaH  
    ## S,D,A are specified by angles from x-axis  
  
    S=S-theta0  
    D=D-theta0  
    A=A-theta0  
  
    sigmaBar=(sigma1+sigma2)/2
```



```

dsigma=sigma1-sigmaBar

ome1=cmath.sqrt( 1 - (dsigma/sigmaBar) ) - cmath.sqrt( 1 + (dsigma/sigmaBar) )
ome2=cmath.sqrt( 1 - (dsigma/sigmaBar) ) + cmath.sqrt( 1 + (dsigma/sigmaBar) )
Omega=cmath.sqrt(ome1/ome2)

r=1e-6 #radius of sample, will be cancelled
smallx=np.cos(A)*r
smally=np.sin(A)*r
X=smallx / cmath.sqrt( 1 + (dsigma/sigmaBar) )
Y=smally / cmath.sqrt( 1 - (dsigma/sigmaBar) )
Z=X + (1j)*Y

alpha=(r/2)*( 1/(cmath.sqrt(1 + (dsigma/sigmaBar) )) + 1/(cmath.sqrt( 1 - (dsigma/\
sigmaBar) ) ) )
preFactor=(1/cmath.sqrt(sigma1*sigma2)) * (1/math.pi) * (1 / (1+ (sigmaH**2/sigma1/\
sigma2)) )

sum1=0
for i in range (0,100,2):
    term1=abs( 1 + cmath.exp( (-2j)*D ) * (Omega** (4*i+2)) - cmath.exp( (-1j)*D \
) * (Omega** (2*i)) * Z / alpha )
    term2=abs( 1 + cmath.exp( (-2j)*S ) * (Omega** (4*i+2)) - cmath.exp( (-1j)*S \
) * (Omega** (2*i)) * Z / alpha )
    term=cmath.log (term1/term2)
    sum1=sum1+term
R1=preFactor * sum1

sum2=0
for i in range (1,100,2):
    term3=abs( 1 + cmath.exp( (2j)*D ) * (Omega** (4*i+2)) - cmath.exp( (1j)*D ) \
* (Omega** (2*i)) * Z / alpha )
    term4=abs( 1 + cmath.exp( (2j)*S ) * (Omega** (4*i+2)) - cmath.exp( (1j)*S ) \
* (Omega** (2*i)) * Z / alpha )
    term=cmath.log (term3/term4)
    sum2=sum2+term
R2=preFactor * sum2

sum34=0
for i in range (0,100,2):
    term1=1 + cmath.exp( (-2j)*D ) * (Omega** (4*i+2)) - cmath.exp( (-1j)*D ) * (\
Omega** (2*i)) * Z / alpha

```

```

term2=1 + cmath.exp( (-2j)*S ) * (Omega** (4*i+2)) - cmath.exp( (-1j)*S ) * (\
Omega** (2*i)) * Z / alpha
phase1=cmath.phase(term1)
phase2=cmath.phase(term2)
term= (phase1-phase2) * sigmaH/ cmath.sqrt(sigma1*sigma2)
sum34=sum34+term
R34=preFactor*sum34

sum56=0
for i in range (1,100,2):
    term3= 1 + cmath.exp( (2j)*D ) * (Omega** (4*i+2)) - cmath.exp( (1j)*D ) * (\
Omega** (2*i)) * Z / alpha
    term4= 1 + cmath.exp( (2j)*S ) * (Omega** (4*i+2)) - cmath.exp( (1j)*S ) * (\
Omega** (2*i)) * Z / alpha
    phase3=cmath.phase(term3)
    phase4=cmath.phase(term4)
    term= (phase3-phase4) * sigmaH/ cmath.sqrt(sigma1*sigma2)
    sum56=sum56+term
R56=preFactor*sum56

R=R1+R2+R34+R56
return (R)

```

Next it is trivial to write functions that gives an expected voltage by simply take the different between the potential at two locations  $V_{SDAB} = \Phi_{SDA} - \Phi_{SDB}$ . We also handle the error of expectation from contacts have finite width here.

```

## calculates V, for given A-B contacts
def V0nDisk (S, D, A, B, theta0, sigma1, sigma2, sigmaH):
    VA=potentialOnDisk(S,D,A,theta0,sigma1,sigma2,sigmaH)
    VB=potentialOnDisk(S,D,B,theta0,sigma1,sigma2,sigmaH)
    return (VA-VB)

## includes the finite width of contact at source-drain, taken as an arc angle
def FiniteWidthV_SD (S, D, A, B, theta0, sigma1, sigma2, sigmaH):
    Angleshift=4/180*math.pi ## half of the contact width
    Rminus=theRwithB (S+Angleshift, D-Angleshift, A, B, theta0, sigma1, sigma2, sigmaH)
    R0=theRwithB (S, D, A, B, theta0, sigma1, sigma2, sigmaH)
    Rplus=theRwithB (S-Angleshift, D+Angleshift, A, B, theta0, sigma1, sigma2, sigmaH)
    ## We take finite width source drain as three pairs of point like source drains. \

```

```

In principle we can do as fine of a mesh as we wish.
return ((Rminus+R0+Rplus)/3).real

## includes the finite width of contact at A-B, taken as an arc angle
def FiniteWidthV_SDAB(nuvalue, S, D, A, B, theta0, sigma1, sigma2, sigmaH):
    R0=FiniteWidthV_SD(nuvalue, S, D, A, B, theta0, sigma1, sigma2, sigmaH)
    AngleS=4/180*math.pi ## half of the contact width
    Rlist=[]
    R1=FiniteWidthV_SD(nuvalue, S, D, A+AngleS, B+AngleS, theta0, sigma1, sigma2, \
sigmaH)
    R2=FiniteWidthV_SD(nuvalue, S, D, A+AngleS, B-AngleS, theta0, sigma1, sigma2, \
sigmaH)
    R3=FiniteWidthV_SD(nuvalue, S, D, A-AngleS, B+AngleS, theta0, sigma1, sigma2, \
sigmaH)
    R4=FiniteWidthV_SD(nuvalue, S, D, A-AngleS, B-AngleS, theta0, sigma1, sigma2, \
sigmaH)
    Rlist=[R1,R2,R3,R4]
    ## upper and lower bound, and the true expected value of voltage drop
    return (max(Rlist), min(Rlist),R0)

```

Next, for a given set of measurement on the same state, imagine 6 columns of input: locations of source, drains, voltage contact A, voltage contact B, and the measured voltage and current. The functions below generate a column of expected voltage based the first four location columns, for some arbitrary matrix, the divide it with the averaged measured current,  $1/\text{inverse}I$ , converting them into resistances. As such, we obtain an array of *theoretically* predicted  $R_{SDAB}^{\text{the}}$ .

```

def RList(theta0,sigma1,sigma2,sigmaH,AngleSList,AngleDList,AngleAList,AngleBList,\
inverseI):
    ## Outputing three columns, expected resistance and their upper and lower bounds, \
will be used for plotting,

    Vlist=np.array(len(AngleSList))
    Vlistmax=np.array(len(AngleSList))
    Vlistmin=np.array(len(AngleSList))
    for i in range (0,len(AngleSList)):
        a,b,c=FiniteWidthV_SDAB (AngleSList[i], AngleDList[i], AngleAList[i], \
AngleBList[i], theta0,sigma1,sigma2,sigmaH)
        Vlist[i]=c

```

```

        Vlistmax[i]=a
        Vlistmin[i]=b

## if you want to work with voltage, then no need for inverseI
    return (np.array(Vlist)*inverseI, np.array(Vlistmax)*inverseI, np.array(Vlistmin)*\
            inverseI)

def RListforFit(theta0,sigma1,sigma2,sigmaH,AngleSList,AngleDList,AngleAList,AngleBList\
, inverseI):
    ## Outputing only the expected resistance for speed, will be used for fitting

    Vlist=np.array(len(AngleSList))
    for i in range (0,len(AngleSList)):
        a,b,c=FiniteWidthV_SD (AngleSList[i], AngleDList[i], AngleAList[i], AngleBList[\
            i], theta0,sigma1,sigma2,sigmaH)
        Vlist=Vlist+[c]

    return (np.array(Vlist)*inverseI)

```

From the measurement, we also do  $R_{SDAB}^{\exp} = V_{SDAB}^{\exp}/I_{SD}^{\exp}$  and obtain a column of *experimentally* measured resistances. Finally, we achieve the extraction by fitting for a column of  $R_{SDAB}^{\text{the}}$  that matches the  $R_{SDAB}^{\exp}$  the most. Fitting method can vary, I used Nelder-Mead to minimize the sum of squares of the residues,  $\sum (R_{SDAB}^{\text{the}} - R_{SDAB}^{\exp})^2$ .

```

def NMFit(ExpV,AngleSList,AngleDList,AngleAList,AngleBList,inverseI,guessPara,kickL\
=[-1]):
    ## ExpV is the experimental value of resistance
    ## TheV is the theory value of resistance
    ## guessPara is the initial guess of the matrix, in the format of [alpha, (sigma_xx\
+sigma_yy)/2, abs(sigma_xx-sigma_yy)/(sigma_xx+sigma_yy), sigmaH]
    ## kick is the list of indices at which in the column of data that you want to \
remove from the fit, in case there are bad points.

    timerS=time.time()
    initial=[guessPara[0],guessPara[1]*(1+guessPara[2]),guessPara[1]*(1-guessPara[3]),\
guessPara[3]]

    def LossFunction(parameters):
        TheV=RListforFit(theta0,sigma1,sigma2,sigmaH,AngleSList,AngleDList,AngleAList,\

```

```

    AngleBList, inverseI)
    SqDifV=(ExpV-TheV)*(ExpV-TheV)
    if kickL[0]>=0:
        SqDifV=np.delete(SqDifV,kickL)
    return (np.sum(SqDifV))

## if you want to work with voltage, then multiply a large number here to make \
calculation easier
## like np.sum(SqDifV)*100000000

## fitting now... I set bounds for my parameters, this is generally large enough
result=optimize.minimize(LossFunction, initial, method="Nelder-Mead", hess=None, \
hessp=None, bounds=((0,np.pi),(0,0.5),(0,0.99),(-0.5,0.5)), constraints=(), tol=1e\
-8, callback=None, options={'maxiter':1e7,'xatol': 1e-7,'maxfev':1e7,'fatol':1e-7,'\
adaptive':True})

if not result.success: ## in case the fit fails
    print("Success:",result.success)

fittedP=result.x
printout1=fittedP[0]
printout2=(fittedP[1]+fittedP[2])/2
printout3=(fittedP[1]-fittedP[2])/(fittedP[1]+fittedP[2])
printout4=fittedP[3]

## calculate the RRMSE of the fit.
loss=np.sqrt(result.fun*(1e-10)/len(ExpV)) / np.sqrt(np.sum(ExpV*ExpV))

print('fittedParaB= ',printout1,",",printout2,",",printout3,",",printout4,") ## NM\
method, loss=",loss, ", kickL=",kickL ,sep="")
print("#### total time=",time.time()-timerS)

return (result)

```

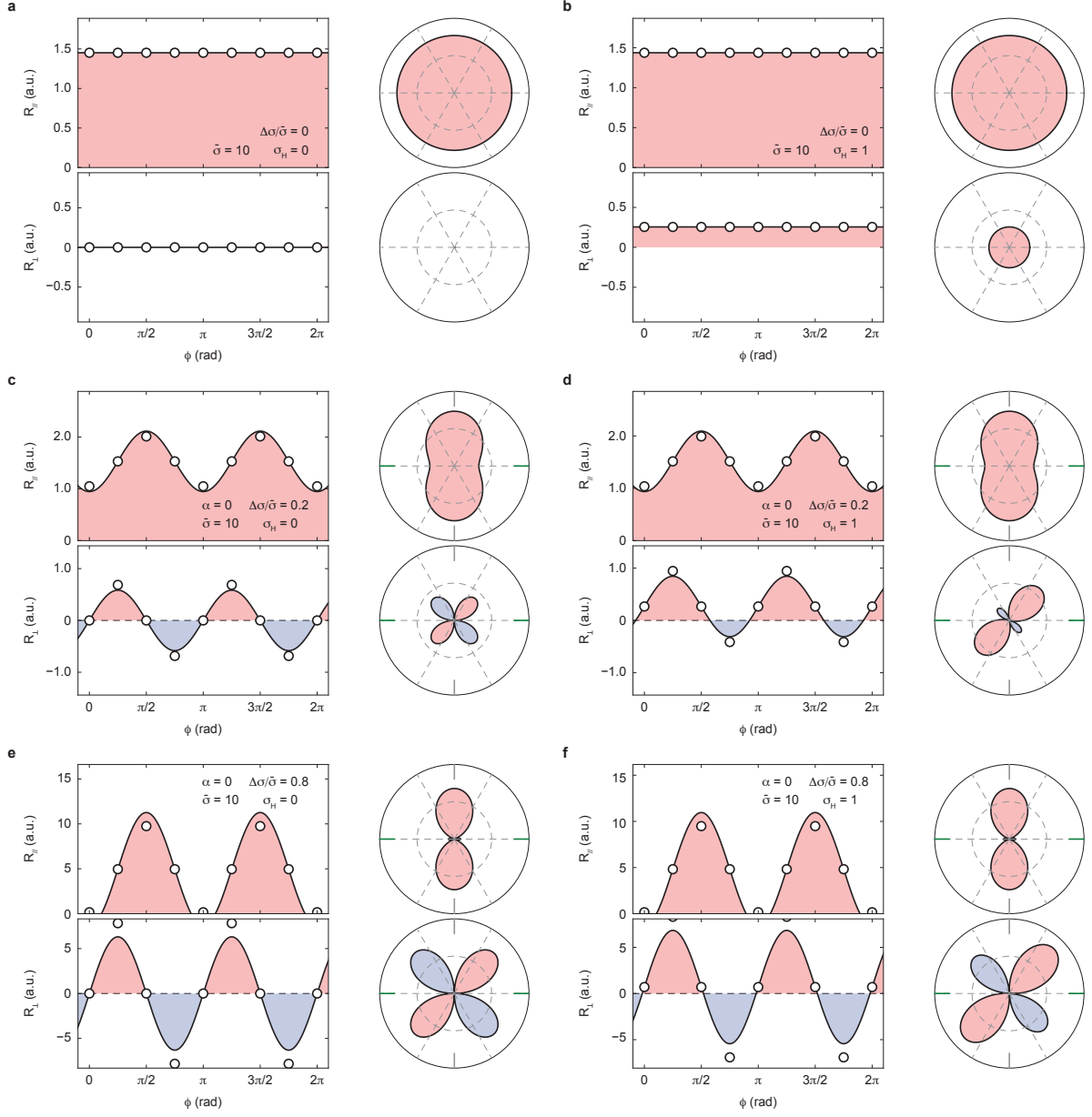
Nelder-Mead algorithm requires an initial guess, which can take a bit of experience. But generally speaking, from the angular dependence fit Eqn. 2.2 - 2.3, taking the initial guess as  $\alpha$ ,  $1/R_0$ ,  $\Delta R/R_0$ , and  $R_H/R_0$  is a good start.

# Appendix C

## Conductivity-resistance conversion in sunflower geometry

As mentioned in Chapter 2 and discussed in Appendix A, different measurement configurations can give rise to different expected angle dependence curves. Fundamentally, due to the point-like contacts and the disk shape of the ideal sunflower geometry, the potential distribution in the sample is expected to be non-uniform. As a result, it is not readily obvious how the structure of the conductivity matrix would translate into the observed variables such as  $\alpha$ ,  $R_0$ ,  $\Delta R/R_0$  and  $R_H$ . In this appendix chapter, we discuss how are the observed resistance behaviors related to the linear conductivity matrix  $\sigma$ .

Using the script from the previous appendix chapter, we are able to calculate the expected voltage measurement result for any arbitrary conductivity matrix with any given measurement configuration. This allows us to simulate the expected angle dependence of the resistance measurements and compare that against the simple phenomenological cosine fit functions Eqn. 2.2 - 2.3. Fig. C.1 shows the simulated data fitted with Eqn. 2.2 - 2.3 assuming different conductivity matrices. In the case of an isotropic conductivity (Fig. C.1a-b), we can show that the  $R_0$  and  $R_H$  accurately describes  $\sigma_{xx} = \sigma_{yy} = \bar{\sigma}$  and  $\sigma_H$  respectively by:



**Figure C.1: Simulated angle dependence.**

Simulated angle dependence of  $R_{\parallel}$  and  $R_{\perp}$  assuming different conductivity matrices:  $\bar{\sigma} = 10$  for all panels; top row is isotropic, middle row has anisotropy  $\Delta\sigma/\bar{\sigma} = 0.2$ , bottom row has strong anisotropy  $\Delta\sigma/\bar{\sigma} = 0.8$ ; left column assumes no Hall conductance, right column has  $\sigma_H = 1$ . For the ones with nonzero anisotropy,  $\alpha = 0$ . As anisotropy strength increases, the angle dependence deviates from cosine functional form. The black open circles denote the simulated data assuming angle-resolved measurement with configuration *I* and *II*. Black curves in each panel denote the simultaneous fit of  $R_{\parallel}$  and  $R_{\perp}$  using Eqn. 2.2 - 2.3.

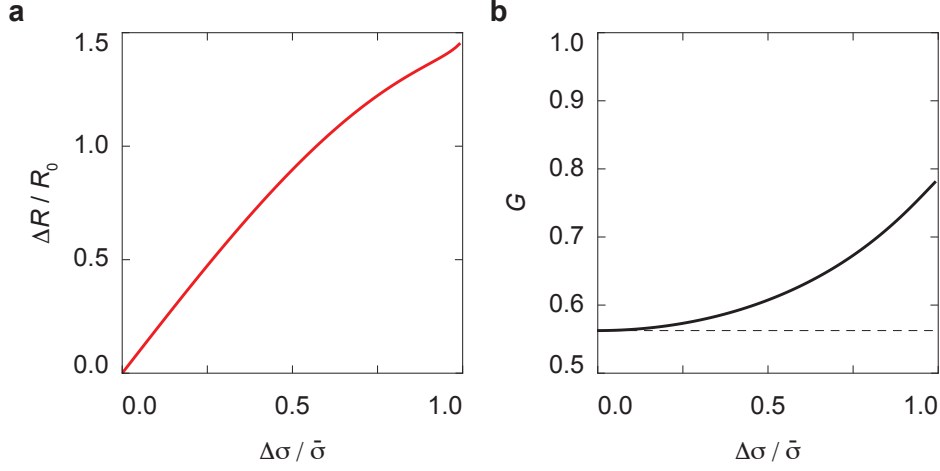


Figure C.2: **Anisotropy dependence of  $\Delta R/R_0$  and  $G$ .**

(a)  $\Delta R/R_0$  and (b)  $G$  as a function of  $\Delta\sigma/\bar{\sigma}$ . Black dashed line denotes  $G(\Delta\sigma/\bar{\sigma} = 0)$ .  $\Delta R/R_0$  and  $R_0$  extracted from simultaneously fitting the angle dependence of  $R_{\parallel}$  and  $R_{\perp}$  using Eqn. 2.2 - 2.3 with configuration I and II.

$$R_0 = G * \bar{\sigma} / (\bar{\sigma}^2 + \sigma_H^2) \quad (\text{C.1})$$

$$R_H = \sigma_H / (\bar{\sigma}^2 + \sigma_H^2), \quad (\text{C.2})$$

where  $G \approx 0.56$  is a dimensionless scalar accounting for the geometric scaling of the  $V_{\parallel}$  measurement setup (configuration I).  $\Delta R/R_0 = 0$  is trivially in accordance with  $\Delta\sigma/\bar{\sigma} = 0$ , where  $\bar{\sigma} = (\sigma_{xx} + \sigma_{yy})/2$  and  $\Delta\sigma = \text{abs}(\sigma_{xx} - \sigma_{yy})/2$ , and  $\alpha$  is ill-defined in these isotropic examples.

When the conductivity is anisotropic, clearly from Fig. C.1c-f, the simulated data deviate from the fit functions Eqn. 2.2 - 2.3, showing how the simple cosine functions are actually not a perfect description of angle dependence of resistance but rather a low-anisotropy approximation: the discrepancy increases as the anisotropy strength increases. Despite the discrepancy, these cosine functions still captures all the information of the conductivity matrix.  $\alpha$ , defined as the direction of minimum  $R_{\parallel}(\phi)$ , is equivalent to the direction of the principle axis with maximum conductance. As illustrated in Fig. C.2a,



$\Delta R/R_0$  describes the strength of anisotropy with a non-trivial monotonic relation to  $\Delta\sigma/\bar{\sigma}$ . The relation in Eqn. C.1 - C.2 are generalized to handle the influence of anisotropy:

$$R_0 = G \times \bar{\sigma} / (\sigma_{xx}\sigma_{yy} + \sigma_H^2) \quad (\text{C.3})$$

$$R_H = \sigma_H / (\sigma_{xx}\sigma_{yy} + \sigma_H^2), \quad (\text{C.4})$$

with the geometric scaling factor  $G$  now dependent on the anisotropy strength  $\Delta\sigma/\bar{\sigma}$ . We calculate this dependency and plot  $G$  as a function of  $\Delta\sigma/\bar{\sigma}$  in Fig. C.2b.

It is also instructive to think of how different measurement configurations can impact the observed resistance for a given conductivity matrix. Fig. C.3 shows the simulated angle dependence of different measurement configurations assuming the same conductivity tensor. Configuration group *IV* exhibits some mixing component from the longitudinal channel, which can be taken care of by averaging *IV* and *IV'* as shown in Fig. C.3d. The angle dependence of each configuration captures the same  $\alpha$  and  $R_H$ . But  $R_0$  and  $\Delta R/R_0$  are influence by the choice configurations. In particular, for the configuration group *III*, there is a separate geometric factor, similar to Eqn. C.3, denoted by  $G^{III}$ . Its relation with  $\Delta\sigma/\bar{\sigma}$  is shown in Fig. C.4a. Taking the best fit of a simple cosine function, we can roughly quantify the difference between oscillation amplitudes,  $\Delta R$ , from different configurations. Fig. C.4b shows  $g$ , the ratio between  $\Delta R$ , from configuration groups *III* and *I*, as well as *IV* and *II*, as a function of anisotropy strength. As mentioned in Appendix A, we have  $g < 1$  for configuration *III* for all anisotropy strength and  $g > 1$  always for configuration *IV*.

With these information, one can fairly accurately calibrate the data from configuration groups *III* and *IV* in order to fit with the data from configuration groups *I* and *II* by:

$$\tilde{R}^{III}(\phi) = (R^{III} - R_0^{III}) \times g^{III} + R_0^I = g^{III} R^{III} + (1 - g^{III} \frac{G^{III}}{G^I}) R_0^I \quad (\text{C.5})$$

$$\tilde{R}^{IV}(\phi) = (R^{IV} - R_H) \times g^{IV} + R_H = g^{IV} R^{IV} + (1 - g^{IV}) R_H, \quad (\text{C.6})$$

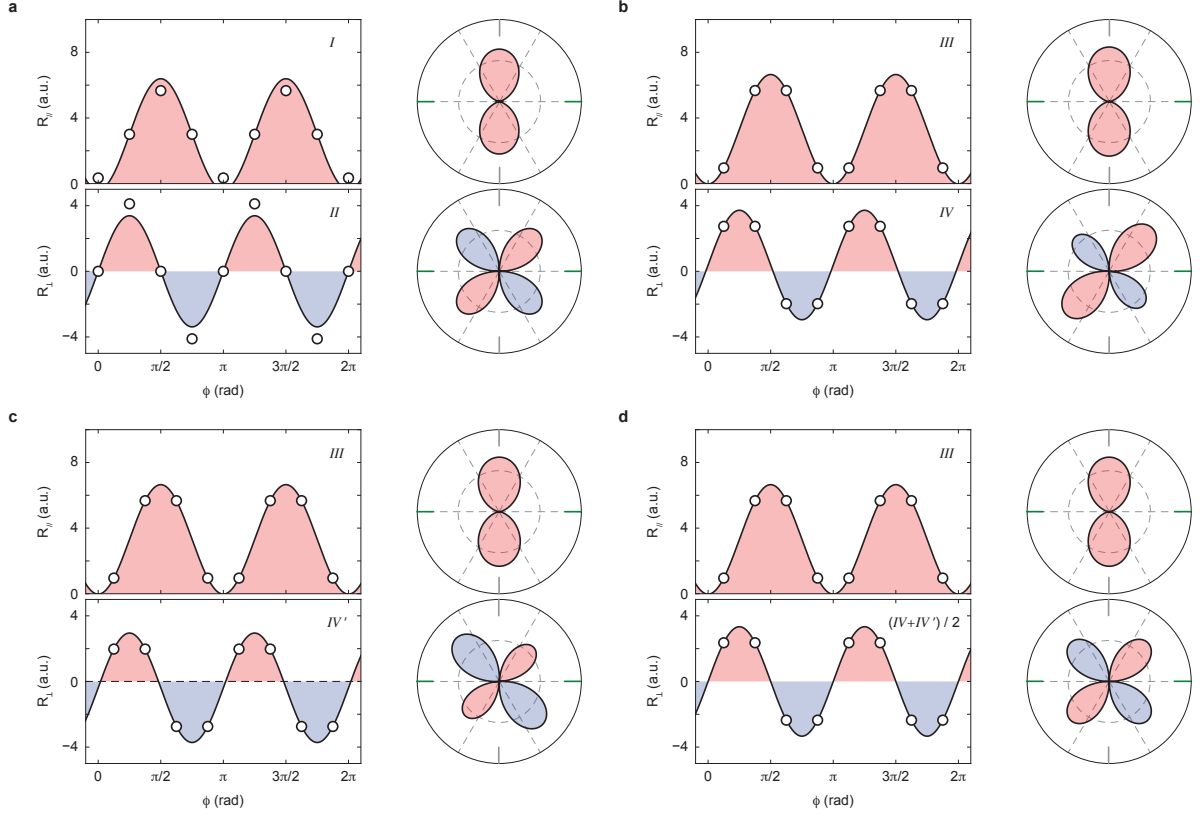


Figure C.3: **Simulated angle dependence with different configurations.**

Simulated angle dependence of  $R_{\parallel}$  and  $R_{\perp}$  assuming the same conductivity matrices:  $\bar{\sigma} = 10$ ,  $\Delta\sigma/\bar{\sigma} = 2/3$ ,  $\alpha = 0$ , and  $\sigma_H = 0$ . The black open circles denote the simulated data assuming measurement configuration (a) I and II, (b) III and IV, (c) III and IV', and (d) III and average between IV and IV'. Black curves in each panel denote the simultaneous fit of  $R_{\parallel}$  and  $R_{\perp}$  using Eqn. 2.2 - 2.3. IV and IV' contains opposite mixing from the longitudinal channel, manifested as a shift in the average of all angle-resolved measurement. Such mixing cancels out by averaging IV and IV', as shown in panel d.

where the superscript denotes the configuration.  $R_0^{III}$  and  $R_0^I$  are the average of  $R^{III}(\phi)$  and  $R^I(\phi)$ , respectively, and related by  $R_0^{III} = R_0^I \times G^{III}/G^I$  through Eqn. C.3.

Notice for any non-vanishing anisotropy,  $(1 - g^{III} \times G^{III}/G^I)/(1 + \Delta R/R_0) < 0.1$ . In the case of weak Hall effect, we also have  $R_H \ll \Delta R$ . Then the calibration is approximated by:

$$\tilde{R}^{III}(\phi) \approx g^{III} R^{III} \quad (\text{C.7})$$

$$\tilde{R}^{IV}(\phi) \approx g^{IV} R^{IV} . \quad (\text{C.8})$$

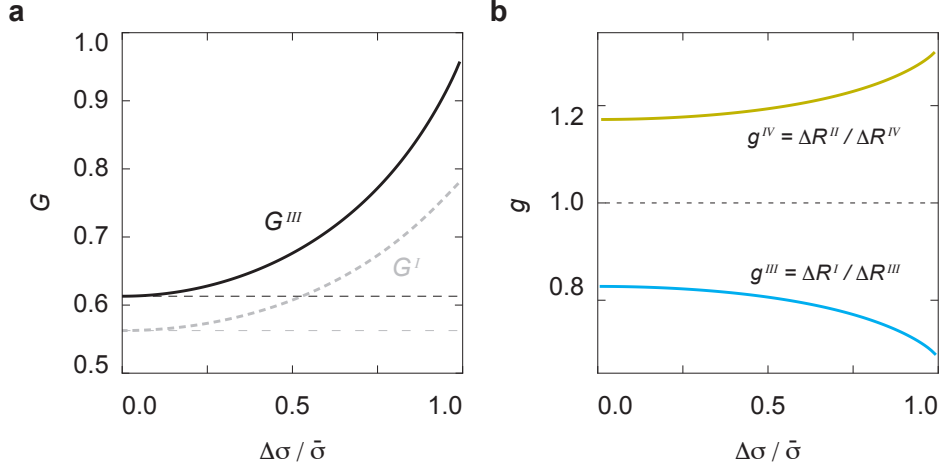


Figure C.4: **Anisotropy dependence of  $G^{III}$  and  $g$ .**

(a)  $G^{III}$  (black curve) as a function of  $\Delta\sigma/\bar{\sigma}$ . Gray curve shows  $G^I$  ( $G$  in Fig. C.2b), illustrates the difference between  $G^I$  and  $G^{III}$ . Black and gray dashed lines denote  $G^{III}(\Delta\sigma/\bar{\sigma} = 0)$  and  $G^I(\Delta\sigma/\bar{\sigma} = 0)$ , respectively. (b)  $g$  as a function of  $\Delta\sigma/\bar{\sigma}$ . Cyan curve denotes  $g^{III} = \Delta R^{III} / \Delta R^I$ ; yellow curve denotes  $g^{IV} = \Delta R^{IV} / \Delta R^{II}$ .  $\Delta R$  and  $R_0$  extracted from simultaneously fitting the angle dependence of  $R_{\parallel}$  and  $R_{\perp}$  using Eqn. 2.2 - 2.3 assuming configurations of either  $I$  and  $II$ , or  $III$  and the average of  $IV$  and  $IV'$ .

One interesting observation is that,  $g^{III}$  and  $g^{IV}$  are related by  $g^{IV} = 2 - g^{III}$ , perfectly mirroring each other around 1. As such, if data from  $III$  and  $IV$  enter the fit together, they constrain the naive cosine functions of Eqn. 2.2 - 2.3 oppositely, allowing us to operationally omit any geometric calibration, as mentioned in Appendix A.

Some other useful relations can be derived using this simulation method as well. Here we list a few that is worth mentioning.  $\xi = (R_{\max} - R_{\min}) / (R_{\max} + R_{\min})$ , is another description of the anisotropy, where  $R_{\max}$  and  $R_{\min}$  are the maximum and minimum values of measured  $R_{\parallel}$ . Without fitting into a continuous (in  $\phi$ ) function,  $R_{\max}$  and  $R_{\min}$  clearly are influenced by  $\alpha$ . For example, if  $\alpha = 0^\circ, 45^\circ, 90^\circ$ , etc., then the measured maximum and minimum values of configuration  $I$  would be the true extrema. But if  $\alpha$  is misaligned from those angles, maximally by  $22.5^\circ$ , then configuration  $I$  would miss the true extrema, resulting in a lower  $\xi$  and an underestimation of  $\Delta\sigma/\bar{\sigma}$ . Similar arguments can be made for  $III$  too, where  $\alpha = 22.5^\circ, 67.5^\circ, 112.5^\circ \dots$  makes the true extrema of the angle dependence aligns with the accessible  $\phi$  and any misalignment from those angles would resulting in a

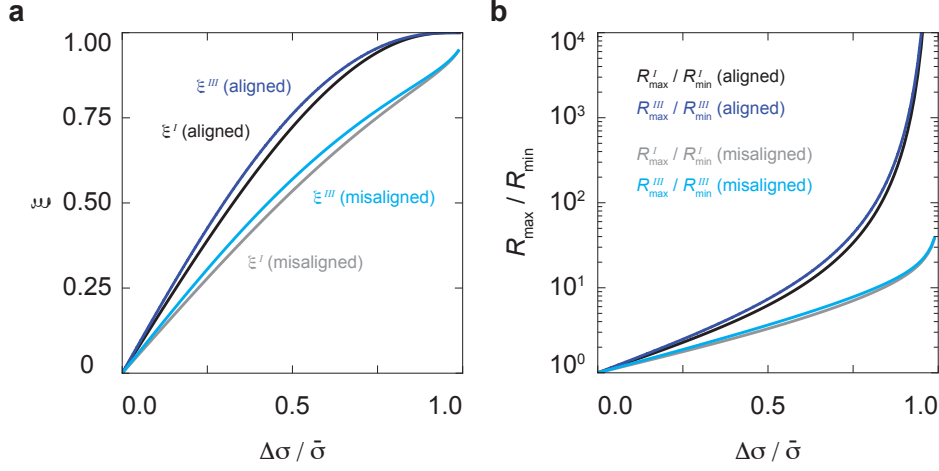


Figure C.5: **Anisotropy dependence of  $\xi$  and  $R_{\max}/R_{\min}$ .**

(a)  $\xi$  and (b)  $R_{\max}/R_{\min}$  as a function of  $\Delta\sigma/\bar{\sigma}$ . Black and blue curves denote the values from configuration *I* and *III*, respectively, with  $\alpha$  aligned with the accessible  $\phi$ , e.g.  $\alpha = 0^\circ$  for *I* and  $\alpha = 22.5^\circ$  for *III*. Gray and cyan curves denote the values from configuration *I* and *III* with  $\alpha$  maximally misaligned with the accessible  $\phi$ , e.g.  $\alpha = 22.5^\circ$  for *I* and  $\alpha = 0^\circ$  for *III*.

lower  $\xi$  and an underestimation of  $\Delta\sigma/\bar{\sigma}$ . Another indicator of anisotropy is  $R_{\max}/R_{\min}$ , requiring minimal knowledge about the state or conductivity matrix, simply comparing the ratio between the most resistive data point and the most conductive data point. Again, this description of anisotropy is also dependent on the alignment of  $\alpha$  with respect to the accessible  $\phi$  of a given configuration in the same fashion. With some simulation, both  $\xi$  and  $R_{\max}/R_{\min}$  can be translated back to  $\Delta\sigma/\bar{\sigma}$ , as plotted in Fig. C.5.

## Appendix D

# Representations of the nonlinear conductivity tensor

Here I summarize the three representations of the nonlinear conductivity tensors:

| Representation                               | Coordinate      | Type             | Description                                                 |
|----------------------------------------------|-----------------|------------------|-------------------------------------------------------------|
| $\tilde{\sigma}_{\alpha\mu\nu}$              | Cartesian       | 6 real scalar    | an intuitive way of writing tensors.                        |
| $\Xi_{+,0,-}^{(2)}$                          | Rotational      | 3 complex values | decomposing the tensor by rotational symmetry and chirality |
| $\mathcal{A}, \mathcal{B}$ and $\mathcal{C}$ | coordinate free | 3 real vectors   | decomposing the tensor by dissipation                       |

And their mutual mathematical relations are derived in Ref. [\[142\]](#):

$$\tilde{\sigma}_{xxx} = \text{Re}(\Xi_-^{(2)}) + \text{Re}(\Xi_+^{(2)}) + \text{Re}(\Xi_0^{(2)}) \quad (\text{D.1})$$

$$\tilde{\sigma}_{xxy} = \text{Im}(\Xi_-^{(2)}) - \text{Im}(\Xi_+^{(2)}) \quad (\text{D.2})$$

$$\tilde{\sigma}_{xyy} = -\text{Re}(\Xi_-^{(2)}) - \text{Re}(\Xi_+^{(2)}) + \text{Re}(\Xi_0^{(2)}) \quad (\text{D.3})$$

$$\tilde{\sigma}_{yxx} = \text{Im}(\Xi_-^{(2)}) + \text{Im}(\Xi_+^{(2)}) + \text{Im}(\Xi_0^{(2)}) \quad (\text{D.4})$$

$$\tilde{\sigma}_{yyx} = -\text{Re}(\Xi_-^{(2)}) + \text{Re}(\Xi_+^{(2)}) \quad (\text{D.5})$$

$$\tilde{\sigma}_{yyy} = -\text{Im}(\Xi_-^{(2)}) - \text{Im}(\Xi_+^{(2)}) + \text{Im}(\Xi_0^{(2)}). \quad (\text{D.6})$$

$$\begin{aligned} \tilde{\sigma}_{xxx} &= \mathcal{A}_x^3 - 3\mathcal{A}_x\mathcal{A}_y^2 + \mathcal{B}_x, \\ \tilde{\sigma}_{xxy} &= 3\mathcal{A}_x^2\mathcal{A}_y - \mathcal{A}_y^3 + \frac{\mathcal{B}_y - \mathcal{C}_x}{2}, \\ \tilde{\sigma}_{xyy} &= -(\mathcal{A}_x^3 - 3\mathcal{A}_x\mathcal{A}_y^2) - \mathcal{C}_y, \\ \tilde{\sigma}_{yxx} &= 3\mathcal{A}_x^2\mathcal{A}_y - \mathcal{A}_y^3 + \mathcal{C}_x, \\ \tilde{\sigma}_{yyx} &= -(\mathcal{A}_x^3 - 3\mathcal{A}_x\mathcal{A}_y^2) + \frac{\mathcal{B}_x + \mathcal{C}_y}{2}, \\ \tilde{\sigma}_{yyy} &= -(3\mathcal{A}_x^2\mathcal{A}_y - \mathcal{A}_y^3) + \mathcal{B}_y. \end{aligned} \quad (\text{D.7})$$

$$\mathcal{A}_x + i\mathcal{A}_y = \sqrt[3]{\Xi_-^{(2)}} \quad (\text{D.8})$$

$$\mathcal{B}_x + i\mathcal{B}_y = \Xi_+^{(2)*} + \Xi_0^{(2)} \quad (\text{D.9})$$

$$i\mathcal{C}_x + \mathcal{C}_y = \Xi_+^{(2)} - \Xi_0^{(2)*}. \quad (\text{D.10})$$