

Probing electronic correlation and topology in twisted multilayer graphene

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Abstract

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Physics

Moiré quantum materials, stacked 2D atomic layers with a small twist angle or a slight lattice mismatch, have become emerging platforms to study the physics of correlation, topology, and the interplay between the two. A plethora of novel states, including unconventional superconductivity, generalized Wigner crystal, orbital magnetism and its associated quantum anomalous Hall effect (QAHE), have been observed within their characteristic flat bands. Moiré quantum materials provide unprecedented opportunities in elucidating the nature of these quantum states of matter because of their high degree of tunability, such as doping, displacement field, pressure, and Coulomb screening.

This dissertation presents transport and optical studies on the emergent correlated and topological states in representative twisted multilayer graphene systems. First, we identify correlated metallic states with isospin symmetry breaking in the phase diagram of twisted double bilayer graphene (tDBG). We employ pressure as a dynamic tuning knob to control the electronic correlation in tDBG. Magneto transport further reveals symmetry-broken Chern insulators at high magnetic field and anomalous Hall effect near zero field. In the second part, we study the correlated phase diagram of twisted monolayer-bilayer graphene (tMBG). We reveal abundant orbital magnetic states across a wide range of twist angle, some with exotic isospin polarized ground states. The high field Landau fan exhibits a variety of closely competing Chern insulators and hysteretic first-order phase transitions in between. We discuss these results and their implication regarding the physics of isospin symmetry, orbital magnetism and Hofstadter butterfly spectrum in graphene moiré

superlattice. Finally, we propose a new optical probe for twisted graphene by using the Rydberg excitons in an adjacent monolayer WSe₂. We demonstrate a new set of moiré Rydberg excitons in the heterostructure of WSe₂/twisted bilayer graphene, whose doping tunable moiré potential unravels the underlying flat bands in twisted bilayer graphene.

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ABBERRIVATION

2D - Two-dimensional

tBLG – twisted bilayer graphene

MATBG - magic-angle twisted bilayer graphene

h-BN - Hexagonal boron nitride

tDBG - twisted double bilayer graphene

tMBG - twisted monolayer bilayer graphene

SBCI - symmetry-broken Chern insulator

CI - Chern insulator

AHE - anomalous Hall effect

QAHE - quantum anomalous Hall effect

MOSTFET - metal–oxide–semiconductor field-effect transistor

LED - light-emitting diode

IQHE - integer quantum Hall effect

FQHE - fractional quantum Hall effect

TKNN - Thouless, Kohmoto, Nightingale, and den Nijs

LDOS - local density of states

IVC - intervalley coherent

VP - valley polarized

SOC - spin orbit coupling

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DEDICATION

To my family

Chapter 1

Introduction

1.1 Motivation and outline

While typing down these words sitting comfortably in front of the computer, it's hard for me not to appreciate how much the field of condensed matter physics has contributed to our modern life. From the Silicon MOSFET (metal–oxide–semiconductor field-effect transistor) inside the CPU to the LED (light-emitting diode) on the screen, the advances in the semiconductor industry would not be possible without the development of condensed matter physics.

Condensed matter physics is the field of physics that seeks to understand the physical properties of matters in a variety of phases, from metal, semiconductor, insulator all the way to magnet and superconductor. Microscopically, the subject under study is a large ensemble of periodically arranged atoms, whose number can easily exceed Avogadro's constant ($N_A = 6.02 \times 10^{23} \text{ mol}^{-1}$). Because of it, the goal of condensed matter physics is not to find an exact Hamiltonian that describes the individual particles, but to find an effective model that can account for its collective behavior. This methodology requires physicists to make smart and reasonable assumptions and approximations of the related and dominant interactions in between the particles. One of the most successful approximations is the independent electron approximation, which is that the interaction between the electron and electron is to be ignored (or treated as a small perturbation as in Fermi liquid theory). Built upon it, the band theory of an ideal Fermi gas in solids can readily explain a significant part of observed phases and lay the foundation for the modern semiconductor technology.

Despite its success in explaining most of the metals and insulators, the failure of the independent electron approximation becomes apparent when considering magnetism, superconductivity, and many others that have been loosely defined as strongly correlated phases. The electron-electron interactions, including the Coulomb interaction U and the exchange interaction J , play a decisive role in these phases of matter. These terms cannot be ignored, or in some cases cannot be taken as a weak perturbation, when constructing the corresponding models. The study of strongly correlated materials, for example, the investigation of superconductivity pairing mechanism and the hunt for new ground states in frustrated magnets, is an active research field and composed a major part of contemporary condensed matter physics.

Another major part of condensed matter physics today – topological physics, adds on an important perspective to the band theory, focusing on its global characteristics. Before the discovery of the integer quantum Hall effect (IQHE) in 1980 [1], Landau symmetry breaking theory is very successful in describing the continuous phase transitions by using a locally defined order parameter ϕ . However, the local order parameter alone is not sufficient to explain the redundant symmetry of states with different quantized Hall conductivity ($\sigma_{xy} = Ce^2/h$, where e is the electron charge, h is Plank constant) in IQHE. Shortly after, Thouless, Kohmoto, Nightingale, and den Nijs (TKNN) [2] revealed that the quantized Hall conductivity Ce^2/h is independent of the energy bands details, but only dependent on the global properties of its Bloch wavefunctions:

$$\sigma_H = \frac{ie^2}{2\pi h} \sum_n \int d^2k \int d^2r \left(\frac{\partial u_n^*}{\partial k_x} \frac{\partial u_n}{\partial k_y} - \frac{\partial u_n^*}{\partial k_y} \frac{\partial u_n}{\partial k_x} \right) \quad (1.1)$$

which always integrates into an integer. The TKNN term is soon realized to be the same as an integral over the 'Berry curvature' on the first Brillouin zone, independently defined by Michael Berry [3]. The integration gives a new physical quantity — the Chern number C — a topological invariant of the material's band structure. The Chern number characterizes a time-reversal-odd topological state, which can only occur when time reversal symmetry is broken either by an external magnetic field or by spontaneous magnetic order. More recently, a time-reversal-invariant topological order—topological insulator, characterized by the $\mathbb{Z}_2$ invariant, also has been identified [4; 5; 6] and experimentally realized [7].

This concludes my brief overview of the condensed matter physics relevant to this thesis. The curiosity and the desire to further understand the strong electron-electron interaction, the band topology, and the interplay between the two, bring me to the more specific and realistic questions: In what material system

can we explore these physics and how can we do it?

To understand how the electron-electron interaction U drives the system away from an ideal Fermi gas (or Fermi liquid), one would desire a material platform where the electron-electron interaction U dominates over the bandwidth W (in other words, electron's kinetic energy). This flat band system, previously, has been realized mainly in the high field Landau levels of 2D electron gas, where the strong electron-electron interactions indeed set in at fractional fillings of these flat bands and drives the system into the fractional quantum Hall states (FQH) [8; 9]. Recently, it has been proposed [10] and experimentally realized in the moiré superlattice of two rotated monolayer graphene near a specific small angle 1.1° [11; 12], the magic-angle twisted bilayer graphene (MATBG). MATBG exhibits extremely flat bands near the Fermi energy in which the strong electronic interaction easily dominates. As a result, a phase diagram composed of both the correlated insulator and the unconventional superconductivity, is observed in the MATBG [11; 12]. Its resemblance to the high-temperature superconductors quickly attracts a tremendous amount of interests into this new type of flat band system.

With a small rotational misalignment, the Dirac points of each monolayer graphene are only slightly offset in the reciprocal space. This allows for strong interlayer coupling that renormalizes the original dispersive band structure, forming almost dispersion-less flat bands. The flat bands setting holds for a variety of graphene superlattice, including but not limited to, MATBG [13; 14; 15; 11; 12], twisted double bilayer graphene [16; 17; 18; 19], twisted monolayer bilayer graphene [20; 21; 22], magic-angle twisted trilayer graphene [23; 24] and magic-angle twisted multilayer graphene family [25; 26]... Moreover, even without a small rotational misalignment, the flat bands can also be engineered in the superlattice of materials with a small lattice constant mismatch: for example, the ABC rhombohedral trilayer graphene aligned with h-BN [27; 28]. The investigation of the flat bands in the moiré superlattice blew and bloomed in the past few years [29; 30]. Beyond graphene, the flat bands are ubiquitous [31] in the moiré superlattice of transition metal dichalcogenides [32; 33; 34], 2d topological insulators [35] and 2d magnets [36; 37; 38].

In this thesis, I will present my Ph.D. research, focusing on transport studies of the electronic correlation and topology in the flat bands of twisted multilayer graphene — in particular, twisted double bilayer graphene (tDBG) and twisted monolayer bilayer graphene (tMBG).

In the remaining part of this chapter, I'll first present the basics of monolayer and bilayer graphene.

I'll then give an overview of the Bistritzer-MacDonald model and practice calculation of the flat bands in MATBG. Finally, I will give an overview of the strongly correlated states and topological states that have been observed in MATBG, with an emphasis on isospin symmetry, orbital magnetism, and Hofstadter physics.

Chapter 2 and 3 will focus on the transport studies of strongly correlated states and topological states in twisted double bilayer graphene (tDBG) and twisted monolayer bilayer graphene (tMBG) respectively. In chapter 4, I will present a proposal and preliminary experiments for optical sensing of twisted graphene using the Rydberg exciton states.

1.2 Band structure of monolayer graphene and MATBG

Graphene is a single layer of carbon atoms sitting in a hexagonal lattice. It is a basic building block for graphitic materials of all other dimensionalities, like graphite, carbon nanotube and Fullerenes. Graphene is the very first example of true 2D atomically thin crystal. Andre Geim and Konstantin Novoselov at the University of Manchester first isolated and characterized graphene in 2004 [39], ended a long debate of the existence of single atom thick two-dimensional crystals. Their works on graphene are quickly recognized by the 2010 Nobel Prize in Physics, “for groundbreaking experiments regarding the two-dimensional material graphene”.

1.2.1 Monolayer graphene

Figure 1.1a shows the hexagonal lattice of a monolayer graphene. Two sets of triangular sublattices A and B, with a lattice constant of $a = 2.46 \text{ \AA}$, are represented in red and blue color respectively. The translation vector can be written as:

$$\mathbf{R} = n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2, n_1, n_2 \in \mathbb{Z} \quad (1.2)$$

where the primitive unit vector is defined as:

$$\mathbf{a}_1 = a \left(\frac{1}{2}, \frac{\sqrt{3}}{2} \right), \mathbf{a}_2 = a \left(-\frac{1}{2}, \frac{\sqrt{3}}{2} \right) \quad (1.3)$$

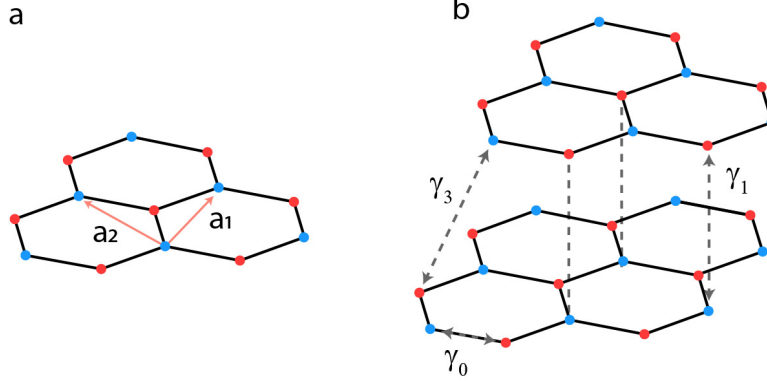


Figure 1.1: Schematic of graphene lattice structure. **a**, Monolayer graphene composed of carbon atoms sitting in a hexagonal lattice with a lattice constant of $2.46 \text{ \AA}$. The A and B sublattice are represented by red and blue color respectively. $\mathbf{a}_1$ and $\mathbf{a}_2$ are the primitive unit vectors. **b**, Lattice structure of Bernal stacked bilayer graphene. The A sublattice of the top graphene layer vertically overlaps with the B sublattice of the bottom layer. The main nearest hopping terms are labelled. γ_0 represent intralayer hopping between adjacent carbon atoms. $\gamma_1 \approx 0.4 \text{ eV}$ represents hopping between top layer A lattice atoms and the bottom layer B lattice atoms. Finally $\gamma_3 \approx 0.3 \text{ eV}$ (not considered in the minimal model discussed in the maintext) represents hopping between top layer B lattice atoms and the bottom layer A lattice atoms.

The electrons in an isolated carbon atom are arranged in the $1s^2 2s^2 2p^2$ configuration. In monolayer graphene, three out of the four outer electrons rearrange into the sp_2 hybridized orbitals, leaving an unpaired electron in the higher energy p_z orbital. The electronic properties of monolayer graphene thus can be modeled accurately by a single orbital tight-binding Hamiltonian. In the nearest-neighbor approximation, we consider only the on-site energy ϵ_{p_z} and the nearest-neighbor hoppings γ_0 . In the second quantization formalism, we write down the tight-binding Hamiltonian:

$$\begin{aligned}
 H = & -\gamma_0 \sum_{\mathbf{R}} c_A^\dagger(\mathbf{R}) (c_B(\mathbf{R}) + c_B(\mathbf{R} + \mathbf{a}_1) + c_B(\mathbf{R} + \mathbf{a}_2)) + h.c. \\
 & + \epsilon_{p_z} \sum_{\mathbf{R}} (c_A^\dagger(\mathbf{R}) c_A(\mathbf{R}) + c_B^\dagger(\mathbf{R}) c_B(\mathbf{R})),
 \end{aligned} \tag{1.4}$$

$c_\alpha^\dagger(\mathbf{R})$ and $c_\alpha(\mathbf{R})$ creates (annihilates) an electron on sublattice α at the site of R . We note that the on-site energy terms $\sum c_\alpha^\dagger(\mathbf{R}) c_\alpha(\mathbf{R})$ sum into a constant thus can be safely offset to zero for convenience. We can rewrite the tight-binding Hamiltonian into momentum space, with:

$$c_A(\mathbf{R}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{R}} c_A(\mathbf{k}), \quad c_B(\mathbf{R}) = \sum_{\mathbf{k}} e^{i\mathbf{k}\mathbf{R}} c_B(\mathbf{k}) \tag{1.5}$$

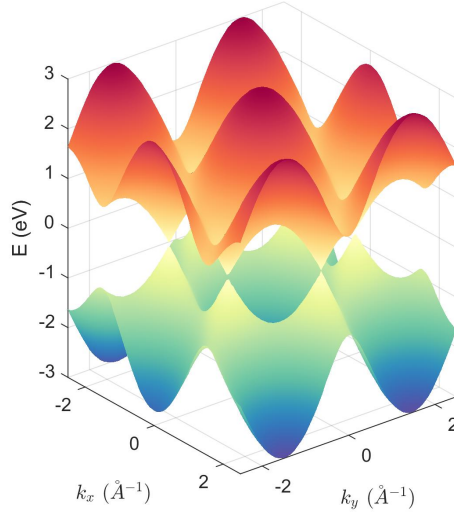


Figure 1.2: Monolayer graphene band structure. Single orbital (p_z) tight-binding calculation of monolayer graphene band structure. We use $\gamma_0 = 2.97$ eV in the calculation. The conduction and valence bands touch at the corners of the first Brillouin zone.

The Hamiltonian in the basis of $|\psi\rangle = [|\psi_A\rangle \ |\psi_B\rangle]$ can be compactly written into a matrix form,

$$\begin{bmatrix} 0 & -\gamma_0 f(\mathbf{k}) \\ -\gamma_0 f^*(\mathbf{k}) & 0 \end{bmatrix} \quad (1.6)$$

with

$$f(\mathbf{k}) = \sum_{i=1}^3 e^{i\mathbf{k}\mathbf{d}_i}, \quad (1.7)$$

in which $\mathbf{d}_1 = (\mathbf{a}_1 + \mathbf{a}_2)/3$, $\mathbf{d}_2 = (-2\mathbf{a}_1 + \mathbf{a}_2)/3$, $\mathbf{d}_3 = (\mathbf{a}_1 - 2\mathbf{a}_2)/3$ are the vectors pointing at the nearest neighboring atomic sites.

We can diagonalize the above matrix and get the eigenvalues of $H(\mathbf{k})$:

$$E_{\pm}(\mathbf{k}) = \pm \gamma_0 \sqrt{4 \cos \frac{\sqrt{3}}{2} dk_x \cos \frac{3}{2} dk_y + s \cos \sqrt{3} dk_x + 3} \quad (1.8)$$

Figure 1.2 shows the calculated energy spectrum. The conduction and valence bands touch at the corners

of the first Brillouin zone, K and K' points:

$$K = \frac{4\pi}{3a}(1, 0), K' = \frac{4\pi}{3a}(-1, 0) \quad (1.9)$$

The low energy excitation of the monolayer graphene, is dictated by the unique massless dispersion near the K and K' points, or to say, Dirac points. We can Taylor expand the Hamiltonian at the Dirac points to a leading term of $\mathbf{q}$, where $\mathbf{k} = \pm K + \mathbf{q}$:

$$H^{\pm K}(\mathbf{q}) = \hbar v_F \begin{bmatrix} 0 & \pm q_x - iq_y \\ \pm q_x + iq_y & 0 \end{bmatrix} = \hbar v_F \mathbf{q}(\pm \sigma_x, \sigma_y) \quad (1.10)$$

where the Fermi velocity, v_F , is defined as $v_F = \frac{3\gamma_0 d}{2\hbar}$, $\hbar$ is the reduced Planck constant, $d = 1.42 \text{ \AA}$ is the distance between adjacent atoms, σ_x and σ_y are Pauli matrices. The spinor structure of the eigenvectors shows an additional pseudospin related to the A and B sublattice. This pseudospin is fully decoupled with the intrinsic electron spin sector. Therefore the monolayer graphene has four flavors (isospin) of massless Dirac quasi-particles in its low energy spectrum, which we usually refer as valley and spin.

1.2.2 Bernal stacked bilayer graphene

In Bernal stacked bilayer graphene, the A sublattice of the top graphene layer overlaps with the B sublattice of the bottom layer in the z direction, with a layer separation of $3.35 \text{ \AA}$ (Fig. 1.1b). So the unit cell of a bilayer is composed of four atoms, two from each layer, which we label as A1, B1, A2 and B2 corresponding to their layer and sublattice. In addition to the nearest hopping term γ_0 between A1 and B1 (as well as A2 and B2), a minimal model of the tight-binding calculation also includes the interlayer hopping term γ_1 , between A1 and B2. We note that a higher order hopping term, γ_3 between B1 and A2, which leads to trigonal warping of the low energy dispersion, is not discussed here. The layer asymmetry can be further controlled by an out-of-plane electric field, that adds a potential difference Δ in the diagonal terms of the Hamiltonian. We write down the full Hamiltonian for this minimal model with a basis in

$$|\psi\rangle = [|\psi_{1,A}\rangle \ |\psi_{1,B}\rangle \ |\psi_{2,A}\rangle \ |\psi_{2,B}\rangle]:$$

$$H(\mathbf{k}) = \begin{bmatrix} \frac{\Delta}{2} & -\gamma_0 f(\mathbf{k}) & 0 & \gamma_1 \\ -\gamma_0 f^*(\mathbf{k}) & \frac{\Delta}{2} & 0 & 0 \\ 0 & 0 & -\frac{\Delta}{2} & -\gamma_0 f(\mathbf{k}) \\ \gamma_1 & 0 & -\gamma_0 f^*(\mathbf{k}) & -\frac{\Delta}{2} \end{bmatrix} \quad (1.11)$$

At $\Delta = 0$, the γ_1 term lifts the degeneracy of $|\psi_{1,A}\rangle$ and $|\psi_{2,B}\rangle$, pushing them to higher energy. This leaves $|\psi_{1,B}\rangle$ and $|\psi_{2,A}\rangle$ degenerate and forms a parabolic bands touching at the corners of the first Brillouin zone. This degeneracy can be further lifted by applying an electric field. As a result, a band gap opens in the low energy spectrum,

$$E(\mathbf{q}) = (v_F \mathbf{q})^2 + \frac{\gamma_1^2}{2} + \frac{\Delta^2}{4} \pm \sqrt{(v_F \mathbf{q})^2 (\gamma_1^2 + \Delta^2) + \frac{\gamma_1^4}{4}} \quad (1.12)$$

so that the system becomes a band gap tunable semiconductor. In the experimental feasible regime, where $E, \Delta \ll \gamma_1$, we can further simplify the description of low energy physics to a two bands model [40]. Hamiltonian after Taylor expansion near the K and K' , in the basis of $|\psi\rangle = [|\psi_{1,B}\rangle \ |\psi_{2,A}\rangle]$ near K and inversibly in $|\psi\rangle = [|\psi_{2,A}\rangle \ |\psi_{1,B}\rangle]$ near K' , can be written as:

$$H^{\pm K}(\mathbf{q}) = -\frac{1}{2m^*} \begin{pmatrix} 0 & (q_x - iq_y)^2 \\ (q_x + iq_y)^2 & 0 \end{pmatrix} \pm \frac{\Delta}{2} \begin{pmatrix} 1 + \frac{q^2}{m^* \gamma_1} & 0 \\ 0 & -1 + \frac{q^2}{m^* \gamma_1} \end{pmatrix} \quad (1.13)$$

where $m^* = \frac{\gamma_1}{2v_F^2}$ is the effective mass. We can solve the low energy spectrum to be:

$$E(\mathbf{q}) = \pm \sqrt{\left(\frac{q^2}{2m^*}\right) + \left(\frac{\Delta}{2}\right)^2} \quad (1.14)$$

Within this two bands model, Bernal stacked bilayer graphene also have an additional pseudospin, related to the A2 and B1 sublattice in the two different layers. Together with the actual electron spin sector, we say that the low energy quasi-particles in Bernal stacked bilayer graphene also have four flavors (isospin).

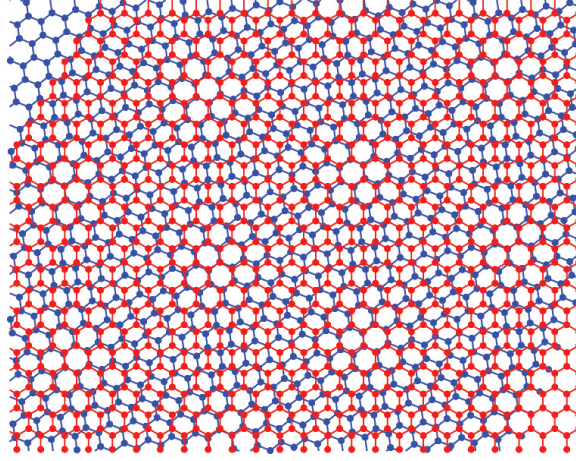


Figure 1.3: Schematic of twisted bilayer graphene lattice structure. Top-view of the moiré pattern in twisted bilayer graphene. Red and blue hexagonal lattice represent the top and bottom monolayer graphene respectively. The new periodicity of the moiré superlattice is $\lambda = a/[2 \sin \theta/2]$. The schematic shows a tBLG with $\theta = 8^\circ$.

1.2.3 Magic-angle bilayer graphene

From the previous section, we've learnt that the low energy quasi-particles in monolayer graphene and Bernal stacked bilayer graphene are featured by linear dispersion and parabolic dispersion respectively. The interlayer hopping γ_1 , despite being only about 1/8 of γ_0 , strongly affects the electronic properties. The interlayer coupling mainly comes from the σ type bonding of the p_z orbitals at A1 and B2 sublattice. Therefore, the coupling strength should be sensitively dependent on the interlayer configuration, including both the layer separation and the lateral displacement of the p_z orbitals caused by a rotational misalignment.

In this section, we discuss the electronic band structure of bilayer graphene with a small rotational misalignment. The control over an arbitrary alignment of the crystalline axis is granted by the weak Van der Waals force in between layers. Figure 1.3 shows the moiré superlattice of two monolayer graphene with a twist angle $\theta = 8^\circ$ (picked for better visualization). The beating of the two sets of hexagonal unit cell with periodicity a give rise to a superlattice with a much larger periodicity $\lambda = a/[2 \sin \theta/2]$. The local atomic registry varies from AA stacking (A1 overlaps with A2, also B1 overlaps with B2) to AB (A1 overlaps with B2 only) and BA (B1 overlaps with A2 only) stacking periodically. The local interlayer hopping, being the strongest at the AA site and relatively weak at AB/BA site, therefore also varies periodically with the same structural periodicity.

Our general goal is to solve the band structure of tBLG given by an Hamiltonian in the following form:

$$H_{TBG} = H_1 + H_2 + H_{\perp} \quad (1.15)$$

where H_1 and H_2 is the Hamiltonian of the top and bottom graphene layer, $H_{\perp}$ is the periodic coupling between the two layers. We directly derive the first two terms following the discussion of monolayer graphene. For simplicity, we assume the top and bottom layer is rotated by $\pm\theta/2$ respectively, the Talyor expansion of the Hamiltonian near valley K is written:

$$\begin{aligned} H_1^{\mathbf{K}}(\mathbf{q}) &= \hbar v_F |\mathbf{q}| \begin{bmatrix} 0 & e^{i(\theta_q - \theta/2)} \\ e^{-i(\theta_q - \theta/2)} & 0 \end{bmatrix}, \\ H_2^{\mathbf{K}}(\mathbf{q}) &= \hbar v_F |\mathbf{q}| \begin{bmatrix} 0 & e^{i(\theta_q + \theta/2)} \\ e^{-i(\theta_q + \theta/2)} & 0 \end{bmatrix} \end{aligned} \quad (1.16)$$

We note that H_1 and H_2 are written in the basis of A/B sublattice of each layer respectively. The Talyor expansion of the Hamiltonian near valley K' is its conjugate.

The real challenge lies in the calculation of $H_{\perp}$. The tight-binding model has limited capability to solve this term. The first issue we will meet is the commensuartion problem of the moiré superlattice. In an incommensurate structure, the transnational vector that relates two identical local atomic registry is infinitely long. Therefore its unit cell, in an exact sense, is infinitely large which forbids us writing down the full Hamiltonian. Rarely at some specific angles, the moiré superlattice becomes commensurate where a tight-binding model is in principle feasible [41; 42].

However as the twist angle approaching zero, to the regime that we are particularly interested in, the number of atoms in one unit cell quickly reach 10^4 below 1° , which is computationally demanding to solve. Therefore, we present solutions following a continuum model mainly developed by Ref. [10]. Without seeking for the atomic orbital details of the tBLG wavefunction, we use a generalized umklapp scattering condition to derive the interlayer hopping term.

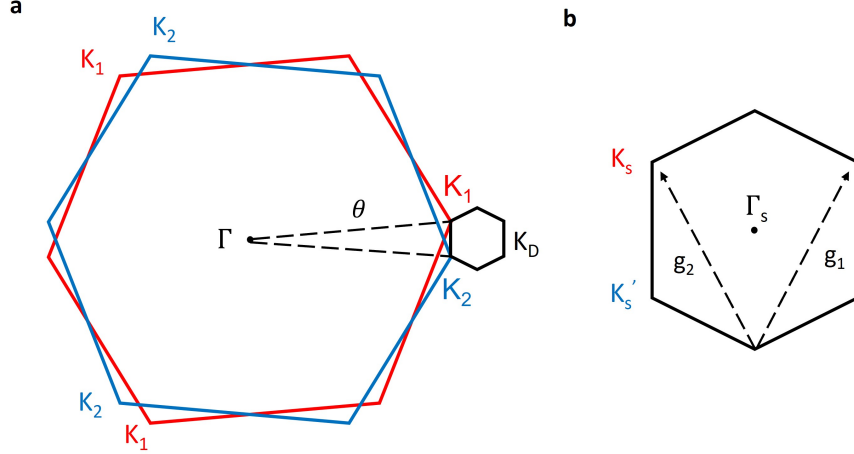


Figure 1.4: tBLG momentum space and the moiré Brillouin zone. **a**, The rotated first Brillouin zones of the two monolayer graphene. In convenience, we assume the top (bottom) layer rotates $\theta/2$ ($-\theta/2$) anticlockwise. The displacement of K_1 and K_2 is defined as $\mathbf{K}_D = \mathbf{K}_2 - \mathbf{K}_1 = \frac{4\pi\theta}{3a}(0, -1)$. **b**, The moiré Brillouin zone emerges at the corners of the original first Brillouin zone, with K_1 (K_2) of the top (bottom) layer being the new valleys of the moiré Brillouin zone K_s (K'_s). $\mathbf{g}_1$ and $\mathbf{g}_2$ are the two reciprocal lattice vectors of the moiré Brillouin zone. In our definition of the graphene lattice unit cell, we have $\mathbf{g}_1 = \frac{2\sqrt{3}\pi\theta}{3a}(1, \sqrt{3})$ and $\mathbf{g}_2 = \frac{2\sqrt{3}\pi\theta}{3a}(-1, \sqrt{3})$.

We've all learnt from Bloch band theory that

$$\psi(\mathbf{r}) = e^{i\mathbf{k}\mathbf{r}}u(\mathbf{r}) \quad (1.17)$$

where $u(\mathbf{r})$ is a periodic function with periodicity same as the crystal lattice. In reciprocal space, this is equivalent to saying that

$$\psi(\mathbf{k}) = \psi(\mathbf{k} + \mathbf{K}) \quad (1.18)$$

in which $\mathbf{K}$ is the reciprocal lattice vector. Therefore, any electronic state with momentum $\mathbf{k}$ can be represented by $\mathbf{k} + m\mathbf{K}$, $m \in \mathbb{Z}$ that falls into the first Brillouin zone. This allows scattering process between states with momentum $\mathbf{k}_1$ and $\mathbf{k}_2 = \mathbf{k}_1 + m\mathbf{K}$, whenever there is a periodic potential providing the reciprocal lattice vector $\mathbf{K}$. This is exactly the case in the tBLG, where the new periodicity is provided by the spatially varying interlayer hopping, with reciprocal lattice vector $\mathbf{g}_1$ and $\mathbf{g}_2$ (Figure 1.4). Only the electronic states that satisfy this generalized umklapp condition can be coupled.

We write down the matrix element of $H_{\perp}$ in momentum space:

$$T_{\alpha\beta}^{12}(\mathbf{q}, \mathbf{q}') = \langle \psi_{\alpha}^1(\mathbf{q}) | H_{\perp} | \psi_{\beta}^2(\mathbf{q}') \rangle \quad (1.19)$$

where 1 and 2 denote the top and bottom layer, α and β denote the A/B sublattice. This term couples the $|\psi_{\alpha}(\mathbf{q})\rangle$ in the top layer to the $|\psi_{\beta}(\mathbf{q}')\rangle$ in the bottom layer. The generalized umklapp condition dicatates that this term is only finite whenever $\mathbf{q} = \mathbf{q}' + m\mathbf{g}_1 + n\mathbf{g}_2, m, n \in \mathbb{Z}$:

$$T_{\alpha\beta}^{12}(\mathbf{q}, \mathbf{q}') = \sum_{\mathbf{g}_{mn}} T_{\alpha\beta}^{12}(\mathbf{g}_{mn}) \delta(\mathbf{q} - \mathbf{q}' + \mathbf{g}_{mn}) \quad (1.20)$$

where $\mathbf{g}_{mn} = m\mathbf{g}_1 + n\mathbf{g}_2, m, n \in \mathbb{Z}$. This formula contains an infinite series of terms which still seems troublesome in computation. However, the beauty of physics comes into help when we further evaluate these terms. Since $T_{\alpha\beta}^{12}(\mathbf{q})$ is the Fourier transform of $T_{\alpha\beta}^{12}(\mathbf{r})$:

$$T_{\alpha\beta}^{12}(\mathbf{q}) = \int d^2r e^{-i\mathbf{q}\mathbf{r}} T_{\alpha\beta}^{12}(\mathbf{r}) \quad (1.21)$$

where $T_{\alpha\beta}^{12}(\mathbf{r})$ describes interlayer hopping between the lattice sites with an in-plane distance r . The formalism of $T_{\alpha\beta}^{12}(\mathbf{r})$, regardless of its details, always depends on the three dimensional separation $\sqrt{\mathbf{r}^2 + d_{\perp}^2}$. Considering that interlayer hopping is dominated by nearest neighbors where $|r| \leq d_{\perp}$, the term $T_{\alpha\beta}^{12}(\mathbf{r})$ should be barely dependent on r . For this reason, $T_{\alpha\beta}^{12}(\mathbf{q})$, as the Fourier transform of a flat $T_{\alpha\beta}^{12}(\mathbf{r})$ function, should strongly peak at zero and rapidly decays for $\mathbf{q}d_{\perp} > 1$. We are therefore safe to consider only the very few first terms with $\mathbf{g}_{mn} = \mathbf{0}, \mathbf{g}_1, \mathbf{g}_2$.

We leave out the lengthy geometric consideration and mathematics, and directly give the dominate interlayer hopping matrices:

$$T^{12}(\mathbf{g}_{00}) = \omega \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix}, T^{12}(\mathbf{g}_{10}) = \omega \begin{bmatrix} e^{-i\phi} & 1 \\ e^{i\phi} & e^{-i\phi} \end{bmatrix}, T^{12}(\mathbf{g}_{01}) = \omega \begin{bmatrix} e^{i\phi} & 1 \\ e^{-i\phi} & e^{i\phi} \end{bmatrix} \quad (1.22)$$

where $\phi = \frac{2\pi i}{3}$, ω is the interlayer hopping strength. We note that the phase of these interlayer hopping terms varies between literature as it depends on the choice of graphene lattice unit cell and the origin of the

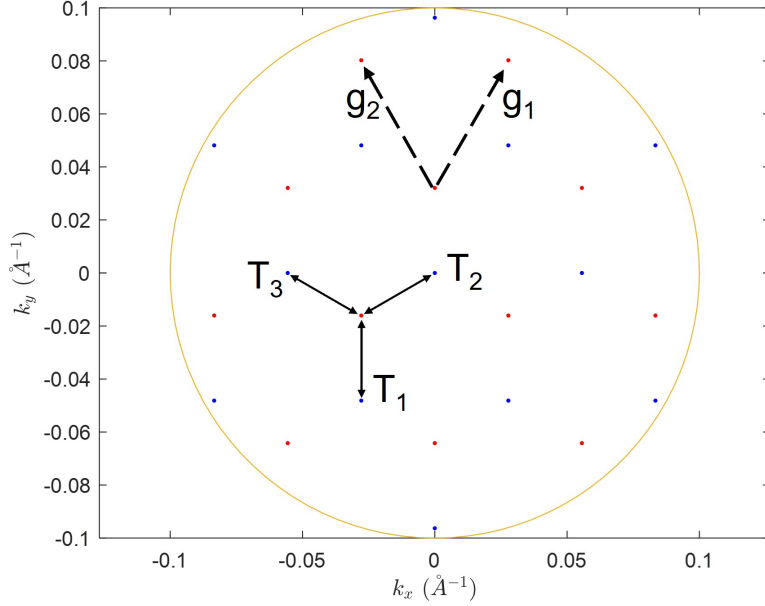


Figure 1.5: A generalized umklapp condition for tBLG momentum space coupling. The momentum space representation of the couplings between states of different q . For any states with momentum q and q' (within the same layer), the periodic interlayer hopping enable coupling only when their momentum satisfy that $\mathbf{q}' - \mathbf{q} = m\mathbf{g}_1 + n\mathbf{g}_2, m, n \in \mathbb{Z}$ (represents by blue dots). Additionally, when we consider states in opposite layers, we need to add the offset between K_1 and K_2 into consideration when we describe them using the same momentum coordinates. The corresponding selection rule reads that $\mathbf{q}' - \mathbf{q} = \mathbf{K}_1 - \mathbf{K}_2 + m\mathbf{g}_1 + n\mathbf{g}_2, m, n \in \mathbb{Z}$ (represents by red dots). $T_{\alpha\beta}^{12}(\mathbf{q}, \mathbf{q}')$ quickly decays for large $\mathbf{q}' - \mathbf{q}$, therefore the interlay hopping are dominated by adjacent dots represented in this momentum space graph. The coupling matrices between them are written as T_1, T_2 and T_3 . The momentum cutoff of the map is $|\mathbf{q}| < 0.1 \text{ \AA}^{-1}$.

moire unit cell. We consider the $\theta = 0$ case, where the tBLG restores to the Bernal stacked bilayer graphene case. $T_{\alpha\beta}^{12}(\mathbf{q})$ has only one nonzero term $T_{AB}^{12}(\mathbf{q}) = 3\omega$, which is equivalent to γ_1 in the previous discussion. Therefore, we get an estimation of the interlayer hopping term $\omega \approx 110 \text{ meV}$.

Now that we have all the terms in the H_{TBG} ready, we will need to find a proper basis to put them together. We consider the spinor state with usual A/B sublattice in layer 1, $|\psi^1(\mathbf{q})\rangle$. It is directly coupled to three spinor states in layer 2 with different momentum, $|\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{00})\rangle$, $|\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{10})\rangle$ and $|\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{01})\rangle$. Each of those states, likewise, are coupled to three states back in layer 1 with momentum adjacent to it. Figure 1.5 shows the map of the coupled momentum states. In summary, the generalized umklapp condition dictates that only momentum states with $\mathbf{q} - \mathbf{q}'$ marching the marked points on the k-space map can be coupled together. Since we only care about the low energy bands near the charge

neutral, we can limit our calculation to the states within a reasonable momentum cutoff, usually around $|\mathbf{g}_{mn}| < 10|\mathbf{K}_D|$.

Figure 1.6 shows the calculation results at a few different twist angles near zero, with a reasonably large momentum cutoff. At all small twist angle, we see strong band structure re-normalization compared with the two simply overlapped Dirac cones. The band structure at the twist angle $\theta = 1.08^\circ$ are featured by a pair of extremely flat bands, additionally with a small band gap to all the other remote bands.

In the original Bistritzer-MacDonald model, the carbon atoms are considered in a rigid hexagonal lattice. The interlayer hopping strength ω is taken to be a uniform function at AA, AB and BA sites. In reality, AA site has σ type bonding of the p_z orbitals at every atomic site while AB/BA site only have half of the amount. As a result, the interlayer separation at AA site is pushed further away compared with the AB/BA site to minimize the electronic wavefunction overlap (due to Pauli exclusion). The ω should be smaller at AA site compared with AB/BA site. The out-of-plane corrugation further induces in-plane atomic displacement: the energetically unfavored AA sites shrink in size while the AB/BA sites expand. This lattice relaxation is captured into the interlayer hopping matrix T , by replacing the diagonal terms with a smaller ω' :

$$T^{12}(\mathbf{g}_{00}) = \begin{bmatrix} \omega' & \omega \\ \omega & \omega' \end{bmatrix}, T^{12}(\mathbf{g}_{10}) = \begin{bmatrix} \omega' e^{-i\phi} & \omega \\ \omega e^{i\phi} & \omega' e^{-i\phi} \end{bmatrix}, T^{12}(\mathbf{g}_{01}) = \begin{bmatrix} \omega' e^{i\phi} & \omega \\ \omega e^{-i\phi} & \omega' e^{i\phi} \end{bmatrix} \quad (1.23)$$

The large moiré unit cell at extremely small twist angle is easily susceptible to lattice reconstruction while it becomes more rigid with increasing twist angle (decreasing unit cell size). Therefore, the lattice relaxation should also be twist angle dependent. At the experimental relevant $\theta \approx 1^\circ$, the usual parameter used in calculation is approximately $\omega' \approx 0.8\omega$. Figure 1.6d shows the band structure of a $\theta = 1.08^\circ$ tBLG, considering the lattice relaxation.

As in the simplest case, we consider only the central four momentum states. we write down its Hamiltonian in the basis of $(|\psi^1(\mathbf{q})\rangle, |\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{00})\rangle, |\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{10})\rangle, |\psi^2(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{01})\rangle)^T$:

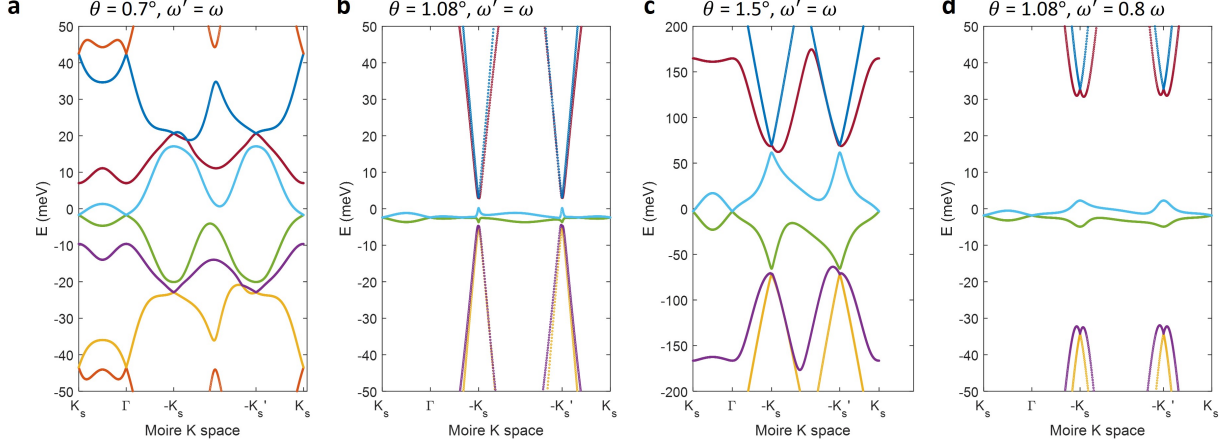


Figure 1.6: tBLG band structure calculated following the continuum model. **a-c**, Calculated band structures of tBLG with a twist angle of **a** $\theta = 0.7^\circ$, **b** $\theta = 1.08^\circ$ and **c** $\theta = 1.5^\circ$, without lattice relaxation. **d**, Calculated band structures of tBLG with a twist angle of $\theta = 1.08^\circ$, considering the lattice relaxation. The high symmetry points are defined in Fig.1.4. Note that the Energy scale in three panels are different. The band structure near the Magic-angle are featured by a pair of extremely flat bands, isolated from other remote bands.

$$H_{BLG}^{\mathbf{K}}(\mathbf{q}) = \begin{bmatrix} H_1^{\mathbf{K}}(\mathbf{q}) & T^{12}(\mathbf{g}_{00}) & T^{12}(\mathbf{g}_{10}) & T^{12}(\mathbf{g}_{01}) \\ T^{12*}(\mathbf{g}_{00}) & H_2^{\mathbf{K}}(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{00}) & 0 & 0 \\ T^{12*}(\mathbf{g}_{10}) & 0 & H_2^{\mathbf{K}}(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{10}) & 0 \\ T^{12*}(\mathbf{g}_{01}) & 0 & 0 & H_2^{\mathbf{K}}(\mathbf{q} + \mathbf{K}_D + \mathbf{g}_{01}) \end{bmatrix} \quad (1.24)$$

Ref. [10] gives important insights by solving the Hamiltonian 1.24 analytically. By solving the Hamiltonian to the first order of momentum $\mathbf{q}$ near the moiré Brillouin zone corners, one can find that the low energy dispersion in tBLG is almost the same as in monolayer graphene, but with a renormalized Fermi velocity,

$$v_F/v_{F0} = \frac{1 - 3\alpha^2}{1 + 6\alpha^2} \quad (1.25)$$

where $\alpha = \frac{\omega}{\hbar v_{F0} |K| \theta}$, is the single dimensionless parameter in the tBLG system. When $\alpha^2 = 1/3$, the renormalized Fermi velocity is expected to be zero in the leading order of q . The lowest pair of moiré bands are expected to be completely flat. The corresponding twist angle is calculated to be at $\theta = 1.1^\circ$, which is named the first magic-angle in tBLG. This finding is later reaffirmed by calculation using a tight-binding

model in a commensurate moiré superlattice with $\theta = 1.08^\circ$ [42], and eventually confirmed experimentally in recent years [43; 44; 45].

1.3 Strong electronic correlation and topology in MATBG

Indeed, the flat bands in MATBG nurture many strongly correlated and topological states, including unconventional superconductivity [12; 46; 47; 48; 49], correlated insulator [11; 47], linear-T resistivity [50; 51; 52], orbital magnetism [53; 54; 55], nematicity [56], Chern insulators [57; 58; 59; 60; 61; 62], symmetry-broken Chern insulators [59; 63] and fractional Chern insulators [63], just to name a few. Most of those exotic phase of matters have been identified in other systems before, nevertheless, MATBG has great advantages compared with traditional quantum materials thanks to its simple chemical composition, extraordinary designer integration ability, high accessibility and numerous degree of freedom tunabilities. The main experimental effort, in the past few years, has been mainly focusing on building a full twist angle survey and observing new correlated and topological phases. More sophisticatedly designed experiments are yet to be done to look into the underlying physics.

1.3.1 isospin symmetry

In both monolayer and Bernal stacked bilayer graphene, the low energy quasi-particles have an extra pseudospin in addition to the real electron spin. This extra degeneracy can be understand as the two valleys K and K' in the momentum space. Altogether, the spin and the valley constitute four degenerate copies of the low energy electronic bands, or in another words, four isospin.

The small-angle tBLG, including MATBG, essentially has the same isospin degeneracy as Bernal stacked bilayer graphene, only that the latter being an extreme case with $\theta = 0^\circ$. There are four copies of each band calculated in Fig. 1.6, that we can label by their isospin: $|K, \uparrow\rangle, |K', \uparrow\rangle, |K, \downarrow\rangle, |K', \downarrow\rangle$. Early local spectroscopy measurement on tBLG shows two Van Hove singularity peaks [13], a first hint of flat electronic bands. Later transport measurements identify isolated flat band with a robust band insulator state at the full filling of the lowest flat bands [14; 15]. The band insulator corresponds to having four electron (holes) in each moiré unit cell, consistent with the isospin degeneracy of the flat bands. The filling factor ν is defined as the number of particles in each moiré unit cell.

The first experimental evidence of strong correlation is the discovery of the insulator states at half filling of the lowest flat bands, or to say at $\nu = \pm 2$ [11]. At both electron and hole doping side of the $\nu = -2$ correlated insulator, superconductivity domes are found in devices with a twist angle very close to the magic-angle [12]. Measurement in samples with improved quality further shows that the correlated insulators (or semi-metals) are ubiquitous at all integer filling ν within the lowest flat bands, and more superconductivity domes can be found in between them [47].

The electron-electron interaction is enhanced at each integer ν . It drives the system into strongly correlated phases. However, at non-integer ν , the observation of SdH oscillation shows that the system maintains Fermi liquid behavior. More interestingly, the magneto-transport measurement always shows an outgoing Landau fan with electron type carrier in between $0 < \nu < 4$ and hole type carrier in between $-4 < \nu < 0$. Before reaching the Van Hove singularities near the band edge, the Hall carrier density always increases until the doping level reaches an integer ν , where it will suddenly reset to zero and resume to increase upon doping over the integer ν . This peculiar behavior of the charge doping is summarized into a cascade behavior of band filling in MATBG [64; 45; 59]: We consider four empty degenerate flat bands. At non-integer ν , the doped carriers are distributed evenly in the empty degenerate bands. However when the doping level approaches integer ν , the enhanced Coulomb interaction lifts the band degeneracy and depletes the carriers in the higher energy bands into the lowest energy bands, breaking certain isospin symmetry. This process repeats from integer ν to the next integer $\nu + 1$ until we fill all the flat bands. Moreover, the doping has to go through the Van Hove singularity of the fourth band and reverse carrier type to reach $\nu = \pm 4$, consistent with transport measurements.

One important open question is the exact isospin symmetry of the correlated states at each integer filling ν . There has been many theoretical attempts using Hartree-Fock calculations to include the Coulomb interaction in various theoretical limits [65; 66; 67], trying to capture the isospin symmetry breaking. Many plausible ground states are proposed, however their energy appears too close to each other and the exact values heavily dependent on experimental details, such as dielectric screening and heterostrain. Limited by spin and valley sensitive techniques, conclusive experimental evidence for those exotic isospin polarized state is yet to be seen. Fortunately, we can take a peek into the valley degree of freedom by the observed orbital magnetic states, thanks to the topological properties of the flat bands.

1.3.2 Orbital magnetization

Ferromagnetism is a phase of matter where time-reversal symmetry is spontaneously lifted at zero magnetic field. The local strength of the ferromagnetism can be characterized by its magnetization M . Microscopically, magnetization can be attributed to either spin magnetic moment or orbital magnetic moment. Most of the ferromagnets discovered to date, are driven by exchange interaction, where parallel aligned spins allow electron wavefunction to be further separated to decrease the total electrostatic energy. Therefore, the spin magnetic moment usually dominates over the orbital magnetic moment, which is a secondary contribution arising from the spin-orbit coupling in these high atomic number elements.

The ferromagnetism discovered in MATBG [53; 54] is unique considering its vanishingly small spin-orbit coupling and the lack of spin anisotropy in such a pure carbon two dimensional system. While a usual spin driven mechanism is highly unlikely in the graphene moiré system, a new type of orbital magnetism, where time-reversal symmetry is lifted by spontaneous orbital magnetic moments, is proposed and gaining support from more and more experimental evidence [68; 69; 70; 71].

The magnetization in graphene moiré is dominated by its orbital component [68; 71]. One unique and delighting property of the orbital magnetism is a new pure electrical switching mechanism of the magnetic states [72]. In general, the orbital magnetization in solids is composed of two parts [73]:

$$M(\mu) = \sum_n \int \frac{dk}{(2\pi)^2} \left[m_n(\mathbf{k}) - \frac{e}{\hbar} (\epsilon_n(k) - \mu) \Omega_n(\mathbf{k}) \right] f(\mu - \epsilon_n(\mathbf{k})) \quad (1.26)$$

where the first term $m_n(\mathbf{k})$ is the self rotation of the wave packet, corresponding to the cyclotron motion of the bulk electrons. The second part describes the center of mass velocity of the wave packet, which is equivalent to the magnetization induced by the edge current. While the first term is insensitive to the chemical potential, the second term contributes a drastic change in the total magnetization when doping the system across a topological gap. At the slightly electron doping side, the orbital magnetization change by an amount:

$$\Delta M = \frac{Ce\Delta}{2\pi\hbar} \quad (1.27)$$

compared with the slightly hole doping side. Since the orbital magnetization dominates over spin magnetization in the graphene moiré, its change ΔM across an Chern gap is considerably large so that the total

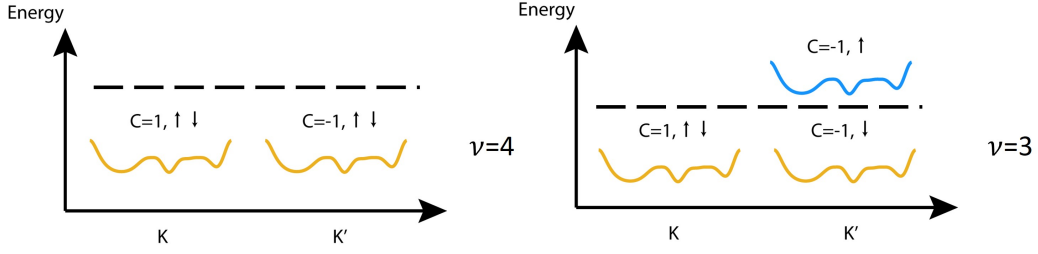


Figure 1.7: Isospin polarization of an orbital magnetic state at $\nu = 3$. Schematic of the band filling at $\nu = 4$ and $\nu = 3$. We consider only the four-fold (spin and valley) degenerate conduction bands, each with a valley contrasting valley Chern number $C_v = \pm 1$. All four bands are occupied at $\nu = 4$. The strong Coulomb interaction at $\nu = 3$ lifts the band degeneracy. As a result, only the $|K', \uparrow\rangle$ band is left empty while the rest are filled. The total Chern number of all filled bands add up to be finite $C = 1$. The time-reversal symmetry is lifted by the spontaneous orbital magnetic moment, forming the orbital magnetism at $\nu = 3$.

magnetization can reverse sign on the two sides of the gap. This is indeed observed in numerous cases to date [69; 71; 74], which further strength the orbital origin of the magnetic states.

Ref. [54] reports a quantized anomalous Hall effect (QAHE) with Chern number $C = 1$ associated with the underlying orbital magnetism at $\nu = 3$ in a h-BN aligned MATBG sample. This measurement, together with reports of QAHE in ABC trilayer aligned with hBN [75] and in tMBG [69], show a ubiquitous topological nature of the observed magnetic states in graphene moiré superlattice. The topological properties of the orbital magnetism can be understand by considering the isospin polarization of the occupied flat bands.

The combined C_2T symmetry protects the Dirac points crossing in MATBG. In h-BN aligned cases, h-BN imprints a stagger potential to the graphene A and B sublattice and breaks the C_2 symmetry. This adds a mass term to the original Dirac Hamiltonian and opens a band gap at the corners of the moiré Brillouin zone K_s and K'_s . The flat bands now can be described as four-fold (spin and valley) degenerate conduction and valence bands, each with a valley contrasting Chern number $C_v = \pm 1$. The simplest isospin polarized state that can explain the observed $C = 1, \nu = 3$ orbital magnetic state is a fully spin and valley polarized state: three out of four conduction bands are fully filled, while only one conduction band with $C_v = -1$ is left empty (Figure 1.7).

More recently, AHE has also been observed in tBLG without h-BN alignment at $\nu = 1$ [55] and $\nu = 2$ [74]. A partially intervalley coherent (P-IVC) state and a partially valley polarized (PVP) state are proposed to be plausible ground state candidates of the observed orbital magnetism respectively. While

their exact isospin polarization remain an open question, these observation reveal possibilities of more complicated orbital magnetism, both in MATBG and other graphene moiré superlattice.

1.3.3 Hofstadter physics

Electrons in solids are subject to the periodic crystal potential $U(\mathbf{r})$ and their wavefunction is governed by the Bloch theory. On the other hand, two dimensional free electron gas forms discretely quantized energy spectrum in a uniform magnetic field described by Landau levels, where electrons motion follow a circular orbital with a length scale $l_B = \sqrt{\frac{\hbar}{eB}}$. In solids, the electron's motion in a uniform magnetic field under a periodic lattice potential can be described by the Hamiltonian:

$$H = \frac{(\mathbf{p} - e\mathbf{A}/c)^2}{2m} + U(\mathbf{r}) \quad (1.28)$$

Philip G. Harper first writes down the solution to the above Hamiltonian in a square lattice with lattice constant a [76]:

$$2\phi_l \cos(2\pi lb - \kappa) + \phi_{l+1} + \phi_{l-1} = E\phi_l \quad (1.29)$$

where $b = \frac{Ba^2}{\phi_0}$ is the magnetic flux through single unit cell, and l is the band index. A full energy spectrum is not given until 20 years later by Douglas Hofstadter [77](Figure 1.8a), mostly due to the commensuration issue between the two length scale l_B and a . The energy spectrum plotted as a function of energy and flux ratio, despite its beautiful fractal structure, is hard to interpret. Instead of focusing on where the energy levels are, Gregory Wannier gives a new perspective by focusing on the open white space in the Hofstadter spectrum where a gap exists. By taking account of the density of states, the energy scale can be convert into carrier density n or filling factor n/n_0 after normalization [78]. Figure 1.8b shows the converted Wannier diagram, where the gap states are represented by the straight lines. Each of the lines can be described a Diophantine equation:

$$\frac{n}{n_0} = t \frac{\phi}{\phi_0} + s, \quad t, s \in \mathbb{Z} \quad (1.30)$$

where t and s are two integers corresponding to the slope and horizontal intersection respectively. These two integers turn out to have important physical meaning. The horizontal intersection s is the band filling factor, or to say, number of electrons per unit cell. The slope t is the total Chern number C of the filled bands, as

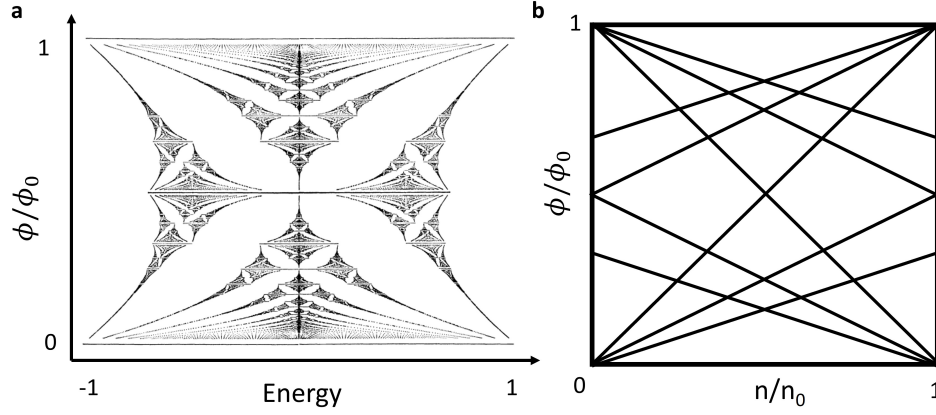


Figure 1.8: Hofstadter butterfly spectrum and the Wannier diagram. **a**, The energy spectrum of electron in a uniform magnetic field under periodic square lattice potential. The beautiful fractal energy spectrum is now known as Hofstadter butterfly. **b**, The Wannier diagram of the same Hofstadter butterfly spectrum. The horizontal axis is normalized carrier density. Each line in the diagram represent a gap state, that can be described by Diophantine equation.

derived from the Streda formula: $\sigma_{xy} = ec \frac{\partial n(E)}{\partial B} = t \frac{e^2}{h}$ [79].

The experimental realization of Hofstadter butterfly is hard because of the length scale l_B would require magnetic field as large as 1000 T to reach lattice constant scale of 1 nm. The only alternative scheme is to engineer artificial lattice with much larger periodicity so that the required magnetic field is achievable.

The moiré superlattice turns out to be a perfect playground providing a length scale on the order of 10 nm. The first Hofstadter spectrum is observed in the moiré superlattice of graphene (monolayer/bilayer) and h-BN [80; 81; 82], where additional integer quantum Hall states traces to finite doping away from Charge neutrality are observed. The gapped state with integer t and s in the Hofstadter spectrum is a simple result of the dual periodicity in a single particle picture. Moreover, the Coulomb interaction is expected to play a significant role in the energy spectrum considering its flatness at certain flux ratio. Indeed, with improved sample quality and by using a non-perturbative capacitance technique, gapped states with fractional Chern number t and fractional band filling factor s have been observed [83]. The observed gapped states can be categorized by their t and s :

1. Gapped states with integer t and $s = 0$ are integer quantum Hall states (IQH);
2. Gapped states with fractional t and $s = 0$ are fractional quantum Hall states (FQH), the FQH is a combined result of topology and strong Coulomb interaction;

3. Gapped states with integer t and integer nonzero s are Chern insulators (CI);
4. Gapped states with integer t and fractional s are symmetry-broken Chern insulators (SBCI), the fractional s requires translational symmetry breaking;
5. Gapped states with fractional t and fractional s are fractional Chern insulators (FCI), the FCI is a combined result of topology, strong interaction and Hofstadter spectrum;

In MATBG, the zero field bands are already flat enough to form a variety of strongly correlated states at integer fillings. The magnetic field further flattens these bands and give birth to a rich set of Hofstadter states [12; 46; 47; 57; 58; 59; 60; 61; 62]. The main gapped states can reach to a quantized Hall plateau with only 1 or 2 T, which is one order lower compared with 15 T that is needed in graphene h-BN moiré superlattice. The main gapped states can maintain quantized ρ_{xy} up to around 4K. These observations point out the importance of Coulomb interaction in stabilizing the Hofstadter states. While in graphene/h-BN moiré, additional energy cost is required to lift the isospin degeneracy to have Chern insulators with s that $\text{mod}(s, 4) = 1, 2, 3$, it is already isospin polarized one way or another in MATBG at zero field. Another important observation is that the sequence of the main Chern insulators follows a cascade behavior with their corresponding (t, s) being $(-1, -3)$, $(-2, -2)$, $(-3, -1)$, $(-4, 0)$, $(4, 0)$, $(3, 1)$, $(2, 2)$, $(1, 3)$ from $\nu = -4$ to $\nu = 4$ [57; 58; 59; 60; 61; 62]. The broken time-reversal symmetry under high magnetic field, thus the broken combined C_2T symmetry, no longer protects the Dirac point crossing. A band gap opens at the moiré Brillouin zone corner K_s and K'_s , very much like the case with broken C_2 symmetry by h-BN alignment at zero field. A significant difference is the energy ordering of the eight total bands now heavily depends on their orbital Zeeman energy. As a result, the four bands with $C = 1$ have lower energy compared with the $C = -1$ bands. The high field cascade of Chern insulators is then a simple reflection of sequential filling of the $C = -1$ bands at $\nu < 0$ and $C = 1$ bands at $\nu > 0$. The total Chern number change by $+8$ as a result of filling the zero-energy Landau levels.

The Chern insulators are stabilized by the strong Coulomb interaction to a very small field, and in the case of Ref. [54], directly connected to a QAHE at zero field. Similarly, the strong Coulomb interaction also stabilizes the symmetry-broken Chern insulators [59] and fractional Chern insulators [63] to a very small magnetic field. To date, the hunt for a zero field fractional Chern insulator remains an active direction. A promising route is to look into moiré systems with a smaller superlattice periodicity where the electron-

electron Coulomb interaction can be enhanced by decreasing particle separation. This can be achieved either in magic-angle twisted multilayer graphene [23; 24; 26] or tBLG at larger twist angle under pressure [46].

1.3.4 Sample fabrication

MATBG provides an unprecedented material platform to study the exotic strongly correlated phases born out from its flat moiré bands [10]. The theoretical works have provided a clear target at the twist angle of 1.1° , yet its experimental implementation is not ready until years after due to difficulties in sample fabrication.

The full stack of a MATBG device is composed of a heterostructure of two rotated monolayer graphene layers, gate dielectric layers and the metallic gates. While different choices of the latter two components exist in literature, in the most ideal case, we use hexagonal boron nitride (h-BN) as the dielectric and few-layer graphite as local top and bottom gate in our sample fabrication. The h-BN is the most commonly used gate dielectric used in 2d material field effect transistor devices due to its atomically flat surface and huge electronic bandgap [84]. Graphite local gates, being atomically flat and mostly defect free, can further act as screening layer for remote charge puddles, defects in the SiO_2 substrate or on top of the sample. Heterostructure of the full MATBG device, being few-layer graphite/h-BN/two rotated monolayer graphene layers/h-BN/few-layer graphite, can be stacked following a polymer-free layer assembly method developed by Ref. [85].

The tBLG bandwidth is calculated to be highly sensitive to twist angle near the magic-angle. Rudimentary rotational alignment of the two monolayer graphene can be achieved by using the straight edges of the exfoliated graphene flakes, which are most likely to be along the zigzag or armchair directions. In alignment procedure of materials without inversion symmetry, nonlinear optical properties such as second-harmonic generation are used to determine the crystal axis, to a precision of roughly 1° . Both ways are not applicable in the fabrication of MATBG. A clever tear-and-stack method [86; 14] comes into rescue for a reliable control over twist angle to roughly 0.1° precision. Instead of trying to figure out the crystal axis of two different exfoliated monolayer, we use the two halves of a single graphene flake and control their relative rotational alignment. This method involves first tearing the graphene flake into halves by using the edge of the h-BN and then stacking them together after rotation, therefore the name tear-and-stack.

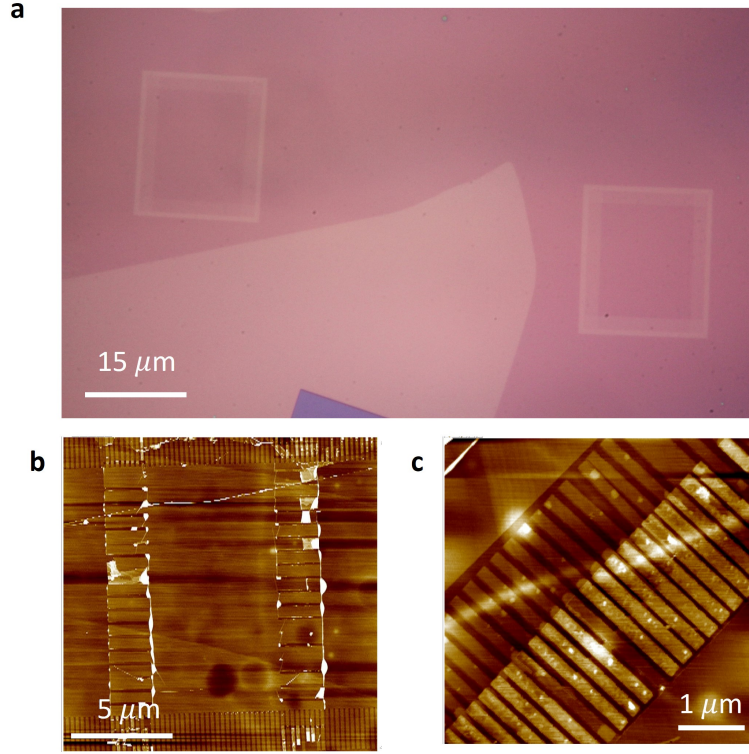


Figure 1.9: Cut-and-stack method in graphene superlattice fabrication. **a**, Optical image of an exfoliated bilayer graphene flake. The bilayer flake is cut into desired rectangular pieces with their edges rotated by the target twist angle. We additionally cut the finger structure on its edge to help stabilize the twist angle in the fabrication process. **b-c**, AFM images of stacked double bilayer graphene on a polymer handle. Panel **c** zooms in at the edge and shows that the finger structure of the two bilayer tend to lock with each other.

More recently, an improved cut-and-stack method is developed [27; 87] to mitigate the unintentional strain induced by the graphene tearing process. A graphene flake is cut into two or more parts by AFM [88] before assembly. Figure 1.9a shows the optical image of a bilayer graphene after cut. We cut the graphene flake into the desired rectangular shape, with their edges rotated by the target twist angle. In addition, we design and cut the finger structure at the edges of the two flakes. The fingers tend to lock between the layers, pose restriction for rotational relaxation therefore stabilize the twist angle (Figure 1.9b-c). This new fabrication method turns out to be useful and is what made possible of the following measurements presented in this thesis. Yet to date, macroscopic measurements still suffer severely from various disorders in the moiré quantum materials. Future experiments on twist-angle-disorder free samples would further enlight the underlying delicate physics.

Chapter 2

Correlated and topological states in twisted double bilayer graphene

Twisted double bilayer graphene (Figure 2.1) — comprising two sheets of Bernal-stacked bilayer graphene rotated by an angle θ — provides a model platform for investigating the properties of graphene moiré beyond MATBG. In this chapter, we explore the strongly correlated states and topological states in twisted double bilayer graphene (tDBG) with electrical transport measurements.

Prior to our study, a number of works of tDBG have identified a robust correlated insulating state at half filling of the conduction band stabilized over a finite range of D , as well as additional CI states at one- and three-quarters fillings which emerge in a magnetic field [16; 17; 18; 19]. Abrupt drops in the resistivity of the neighboring metallic states are observed as the temperature is lowered, saturating to small values at low temperature [16; 17; 18]. In addition, these states exhibit highly nonlinear I - V characteristics [17]. Together, this raises the possibility that these transport features arise as a consequence of superconductivity [16; 17; 89]. More recently, correlated states at charge neutrality [90] and the valence bands [91; 92] have been reported as well, the role of correlations in all these states remains unclear. Overall, transport in tDBG is complicated as it is determined by a number of factors which appear to be of similar magnitude: width of the flat bands, energy gaps isolating these bands, and strength of Coulomb interactions. To date, a complete understanding of the correlated phase diagram of tDBG has not been achieved.

In this chapter, we first examine the fundamental role of spontaneous symmetry breaking in tDBG's

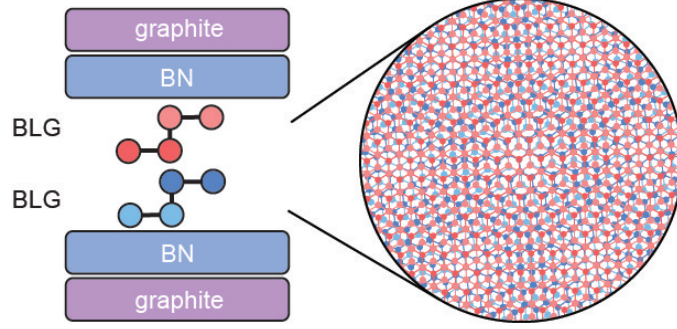


Figure 2.1: Schematic of a tDBG device encapsulated in BN with graphite gates. The dual-gated geometry of the devices allows independent tuning of n and D . We calculate $n = (V_t C_t + V_b C_b)/e$, and $D = (V_t C_t - V_b C_b)/2\epsilon_0$, where C_t and C_b are the top and bottom gate capacitances, V_t and V_b are the top and bottom gate voltages, e is the electron charge, and ϵ_0 is the vacuum permittivity. We note that our definition of displacement field D follows the convention in the field whereby it has the same units as electric field.

phase diagram by investigating its temperature dependent transport properties. We then demonstrate the pressure tuning effect on the correlated phase diagram of tDBG. Finally, we study and compare tDBG with twist angle near both 0° and 60° . We identify symmetry-broken Chern insulators and anomalous Hall effect in both AB-AB and AB-BA samples.

2.1 Symmetry breaking in tDBG

In this section, we investigate temperature-dependent electrical transport of dual-gated tDBG. We have studied five devices, with θ ranging from 1.17° to 1.53° near zero degree twist, and observed qualitatively similar behavior in all devices. We will focus our attention to a $\theta = 1.30^\circ$ device for simplicity of discussion, where the correlated states are observed to be the strongest. We present a detailed survey of the twist angle dependence of the correlated phase diagram in Appendix A.1.

We first identify transport features corresponding to the single-particle band structure of tDBG by measuring transport at high temperature, such that correlated states are suppressed. Figure 2.2b shows the device resistivity, ρ , as a function of charge density, n , and D at temperature $T = 30$ K. The top axis denotes the filling factor of the bands, ν . For small $|D| \lesssim 0.3$ V/nm, the device exhibits only weak resistivity modulations as n and D are tuned. Most notably, a broad cross-like feature is observed for hole-type doping ($\nu < 0$). For larger $|D|$, we observe a substantial difference in ρ between electron- and hole-type doping.

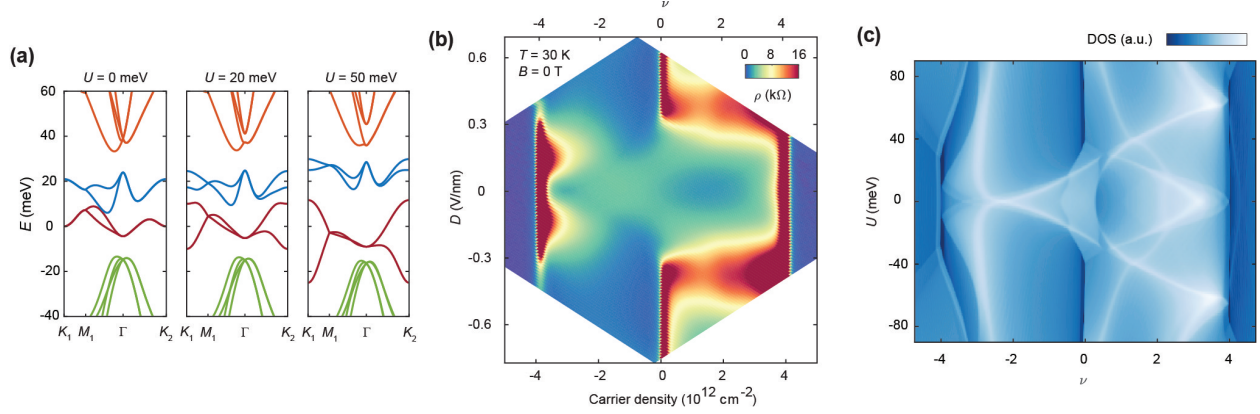


Figure 2.2: Tunable band structure and transport in a 1.30° tDBG device. **a**, Calculated band structure of tDBG with $\theta = 1.30^\circ$ at various values of interlayer potential, U . **b**, Resistivity of a tDBG device with $\theta = 1.30^\circ$ at $T = 30$ K. The corresponding band filling factor ν is shown on the top axis. **c**, Calculated density of states as a function of ν and U , plotted on a log color scale.

We compare our observations to the single-particle band structure of tDBG, calculated following the procedure developed in Ref. [89] with $\theta = 1.30^\circ$. Figure 2.2a shows the band structure for various values of the interlayer potential, U , and Fig. 2.2c plots the corresponding density of states as a function of ν and U . The model predicts a semimetal-to-semiconductor transition at the CNP ($\nu = 0$) with increasing $|D|$, as well as band gaps at full filling of the valence and conduction bands ($\nu = \pm 4$, where the factor of 4 accounts for the spin and valley degeneracy of the bands) which diminish with increasing $|D|$. These features correspond well with insulating states observed over similar ranges of $|D|$ in our measurement (Fig. 2.2b). Additionally, the model predicts a bifurcation of the van Hove singularity in the valence band with increasing $|D|$, forming a cross-like feature in the density of states (white contours in Fig. 2.2c for $\nu < 0$) reminiscent of our observed cross-like transport feature in Fig. 2.2b. Finally, the model predicts that the conduction band becomes flatter with increasing $|D|$, whereas the valence band becomes more dispersive (Fig. 2.2a). When the two bands are isolated by a gap at the CNP (*i.e.* at large $|D|$), differences in mobility arising from the relative flatness of each band are likely responsible for the large discrepancy in ρ observed between electron- and hole-type doping. Thermal activation measurements at $\nu = 0$ confirm that a gap emerges for $|D| \gtrsim 0.25$ V/nm and grows monotonically upon further increasing $|D|$ (Fig. 2.3c).

We now turn our attention to transport at our base temperature of $T = 20$ mK, shown in Fig. 2.3a. Most of the transport features described above persist to base temperature, however we observe the additional emergence of new features in the conduction band at large $|D|$ which are not anticipated within the single-

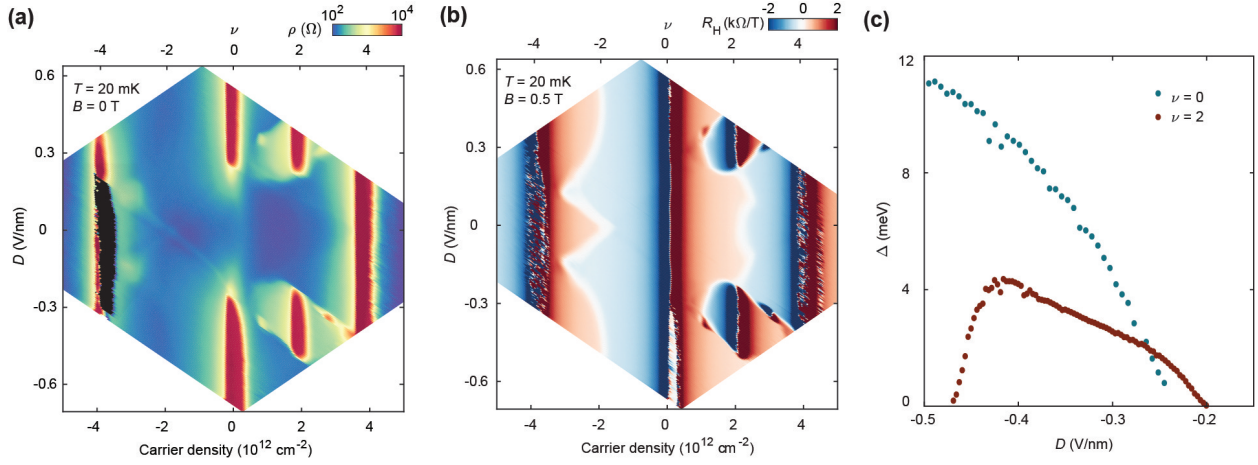


Figure 2.3: Correlated states in a 1.30° tDBG device. **a**, Resistivity at $T = 20$ mK. The region colored in black corresponds to artificially negative resistance in the contacts used for this measurement. **b**, Hall coefficient, R_H , antisymmetrized at $|B| = 0.5$ T. **c**, Energy gaps of the CNP ($\nu = 0$) and the CI state at $\nu = 2$ as a function of D measured by thermal activation.

particle model. A well developed insulating state emerges at half filling of the conduction band ($\nu = 2$) over a finite range of $|D|$, as well as incipient insulating states at $\nu = 1$ and 3. We additionally observe tilted halo-like features surrounding these states characterized by a weak enhancement of ρ . These states arise as a consequence of correlations, as previously identified in prior studies of tDBG [16; 17; 18; 19]. Thermal activation measurements of the energy gap of the $\nu = 2$ CI state, as shown in Figure 2.3c), mark the approximate range of $|D|$ over which correlated states are observed. In particular, CI states emerge when $|D|$ is approximately large enough to gap the bands at $\nu = 0$. The suppression of the CI gap at even larger $|D| \approx 0.47$ V/nm is consistent with prior reports, in which this was argued to be tied to the gap closing at $\nu = 4$ [16; 17; 18; 19]. However, we are not able to directly confirm this in our device owing to limitations in the accessible range of gate voltage.

Corresponding measurements of the Hall coefficient, $R_H = (R_{xy}[B] - R_{xy}[-B])/(2B)$, provide additional insight into the nature of these correlated states (Fig. 2.3b). The small applied $|B| = 0.5$ T does not substantially alter the transport, and can therefore be directly compared with Fig. 2.3a. We observe rapid sign changes in R_H as a function of carrier density at nearly all of the insulating states ($\nu = 0, 2, 3$, and ± 4), as well as a weaker modulation near the incipient CI state at $\nu = 1$, owing to the sign change of the carrier mass on opposite sides of the band gaps. R_H also changes sign at partial band filling in regions of the phase diagram that are well described by single-particle considerations, consistent with the required

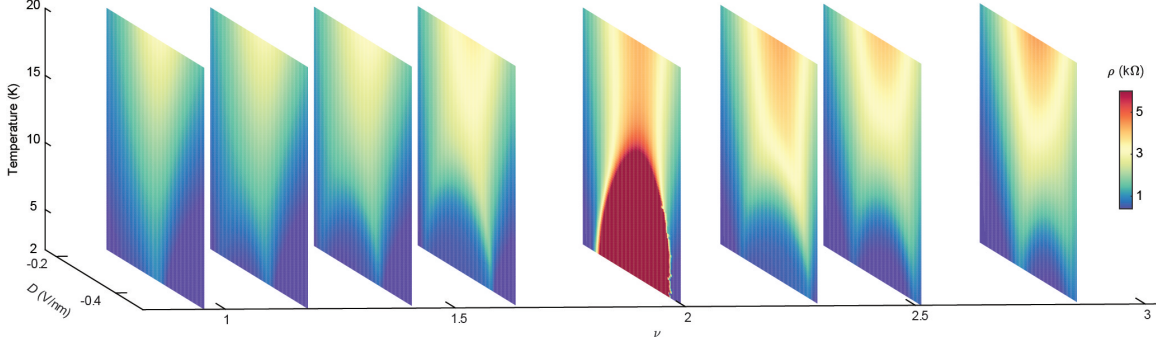


Figure 2.4: Temperature-dependent transport in tDBG. Resistivity as a function of temperature and displacement field at a few select values of ν .

reversal of the carrier sign as the band is tuned from empty to filled. We observe additional sign changes in R_H corresponding nearly exactly to the boundary of the halo observed in measurements of ρ in Fig. 2.3a. Furthermore, replica halo features are also apparent in R_H surrounding $\nu = 1$ and 3. As we will discuss in detail later, these sign reversals arise as a consequence of correlations restructuring the bands.

We investigate the properties of the metallic states by measuring the temperature-dependence of ρ . Figure 2.4 shows $\rho(T)$ as a function of D for a few select values of ν . The development of the CI state at $\nu = 2$ results in an arch-like feature centered at optimal D , with an associated metal-insulator transition at $T \approx 15$ K. The arch-like feature persists for $\nu \neq 2$, however in contrast ρ continues to decrease monotonically as the temperature is lowered. The maximum onset temperature of the arch decreases as ν is detuned further from 2, with the arms of the arches at base temperature corresponding to the boundaries of the primary halo in Figs. 2.3a. Figure 2.5a shows $\rho(T)$ at four values of (ν, D) within the halo, as denoted in the figure inset. The orange curve exhibits a metal-insulator transition associated with the development of the CI state at $\nu = 2$. Away from $\nu = 2$, $\rho(T)$ scales linearly with T at high temperatures, but further exhibits an abrupt drop at a temperature corresponding to the formation of the arch in Fig. 2.4, before eventually beginning to saturate to a finite residual resistivity at low T . The dark green curve, acquired precisely on the halo boundary, exhibits the most rapid drop in $\rho(T)$. In contrast, transport outside the halo region does not manifest such resistivity drops, instead always exhibiting linear-in- T resistivity until eventually saturating to a finite residual resistivity at low T (Fig. 2.5b). Such behavior is commonly observed in graphene [93; 84], however here we observe a very large slope of a few hundred ohms/Kelvin comparable to prior observations in tBLG [50; 51].

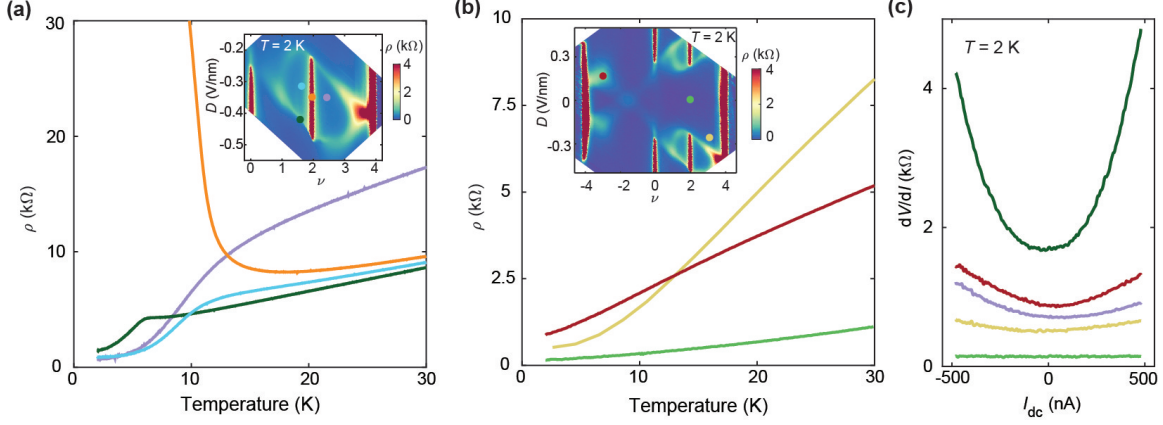


Figure 2.5: Rapid resistance drop and non-linear IV curves. **a-b**, $\rho(T)$ inside and outside the halo region, respectively, at select values of (ν, D) denoted by the color-coded circle markers in the insets. (Insets) Resistivity at $T = 2$ K. **c**, Differential resistance, dV/dI versus applied dc current bias, I_{dc} , at $T = 2$ K for a representative subset of the curves in **a-b**, following the same color-coding scheme.

The abrupt drops in $\rho(T)$ in curves acquired within the halo region resemble the onset of superconductivity. However, despite heavy electronic filtering, we observe a finite residual resistivity at base temperature of a few hundred ohms (Figs. 2.3a) in all devices. As an additional check, we measure the differential resistance, dV/dI , as a function of applied dc current bias, I_{dc} at $T = 2$ K (Fig. 2.5c). The curves are acquired at the same (ν, D) as in Figs. 2.5a-b. Inside the halo (dark green and purple curves), we observe nonlinear transport that is not generally anticipated for a typical metal. Although superconducting states also exhibit nonlinear $I-V$, we do not observe signatures of a well defined critical current in our measurements, nor do we observe saturation to a “normal state” value of dV/dI up to $I_{dc} = 500$ nA. Furthermore, nonlinear transport is not limited solely to curves acquired within the halo, but is also observed outside the halo in cases where the corresponding $\rho(T)$ does not exhibit an abrupt drop (yellow and red curves). Overall, nonlinear transport in our devices is generically observed in states close to the van Hove singularities (Fig. 2.2c), and therefore does not appear to be a signature of superconductivity.

Similar non-linear transport properties have recently been observed in the normal states of MATBG [94] and tBLG away from the magic angle [95]. In the graphene superlattice with a narrow bandwidth and a

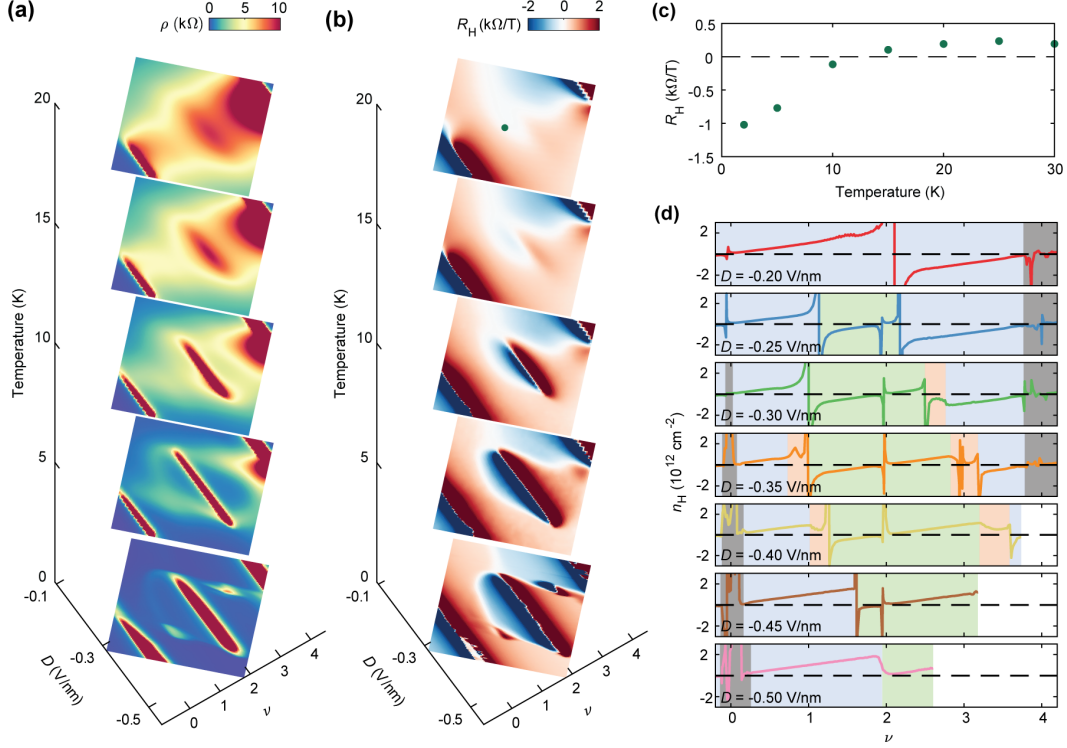


Figure 2.6: Evidence for spontaneous symmetry breaking inside the halo region. **a**, Resistivity acquired at $T = 20$ K, 15 K, 10 K, 5 K and 20 mK, respectively (top to bottom). **b**, Corresponding maps of the Hall coefficient, R_H . **c**, Hall coefficient, R_H , as a function of T acquired at slight underdoping of the CI state at $\nu = 2$ (corresponding to the green circle marker in **b**). **d**, Hall density, n_H , as a function of ν at various D . Color shading denotes regions of the phase diagram with different inferred degeneracy, where blue corresponds to four-fold degeneracy, green corresponds to two-fold degeneracy, and orange has no remaining degeneracy. Gray-shaded regions denote insulating states at $\nu = 0$ and 4.

small Fermi velocity v_F , a very small electric field E can drive the Fermi sea out-of-equilibrium (with drift velocity $v_d \geq v_F$). The dV/dI as function of I_{dc} therefore features an U shape turning up in the small I_{dc} limit, and reach to a pronounced quasi-particle peak at $v_d = v_F$, marking the electron-hole pairs generated by the Schwinger-like mechanism [96; 97]. Our observation of the pronounced non-linear transport near the van Hove singularities is in general consistent with this mechanism.

Figures 2.6 a-b show maps of ρ and R_H , respectively, surrounding the halo region from $T = 20$ mK to 20 K acquired in ~ 5 K steps. The CI state and the surrounding resistive halo smear together as the temperature is raised (Fig. 2.6a), eventually forming a highly resistive stripe spanning the entire conduction band. In the corresponding R_H (Fig. 2.6b), the $R_H = 0$ contour shrinks towards $\nu = 2$ as the temperature is raised. This is particularly evident for $\nu < 2$, as the corresponding contour for $\nu > 2$ is complicated by

thermal activation to the higher-energy remote band [51]. Within much of the halo region, we find that the sign of R_H reverses as the temperature is lowered (Fig. 2.6c, corresponding to the green circle marker in Fig. 2.6b).

Given the apparent association of the halo features in ρ with the sign changes in n_H , we interpret the halo regions as footprints of spontaneous symmetry breaking within the phase diagram of tDBG. Figure 2.6d shows the corresponding evolution of the Hall density, $n_H = e/R_H$, for select values of D at $T = 20$ mK. While n_H exhibits a single sign change near $\nu = 2$ at small D , the emergence of the CI states results in additional sign changes and abrupt resets of n_H at integer ν . The sign of n_H follows single-particle expectations within the blue-shaded regions of Fig. 2.6d. We therefore infer that these regions correspond to metallic states with full spin and valley degeneracy. The green shading corresponds to the primary halo region observed in Figs. 2.6a-b, and is bounded by additional sign changes in n_H . This denotes regions of the phase diagram associated with the formation of the CI state at $\nu = 2$, in which we presume that a single degeneracy is lifted. The orange shading corresponds to the replica halos in Figs. 2.6a-b, and is bounded by additional abrupt resets in n_H . This marks regions of the phase diagram which we presume to have no remaining degeneracies owing to the additional development of the CI states at $\nu = 1$ and 3.

Collectively, our results suggest that correlations dramatically restructure the Fermi surface as a function of n , D , and T . In particular, correlated states emerge as T is lowered, and appear to be strongest at optimal values of $\nu \approx 2$ and $D \approx 0.35$ V/nm at this twist angle. At base temperature, detuning n and D from these values further modifies the band structure and results in the formation of additional correlated ground states. Within the halo region, abrupt drops in $\rho(T)$ arise concomitantly with sign changes in R_H , and therefore appear to be directly associated with the development of symmetry-broken states within the isolated flat band.

Although we are not able to unambiguously determine the ground state ordering of the CI states or the neighboring symmetry-broken metallic states, the CI states always become more resistive with in-plane field suggestive of spin-polarized ordering. Our measurements of the energy gap of the $\nu = 2$ CI state do not scale trivially with in-plane magnetic field (Figure 2.7), possibly due to a unique in-plane magnetic field orbital contribution in tDBG [89] which arises in addition to the usual Zeeman energy. The precise mechanism driving the abrupt drop in $\rho(T)$ within the halo region also remains an open question, however qualitatively

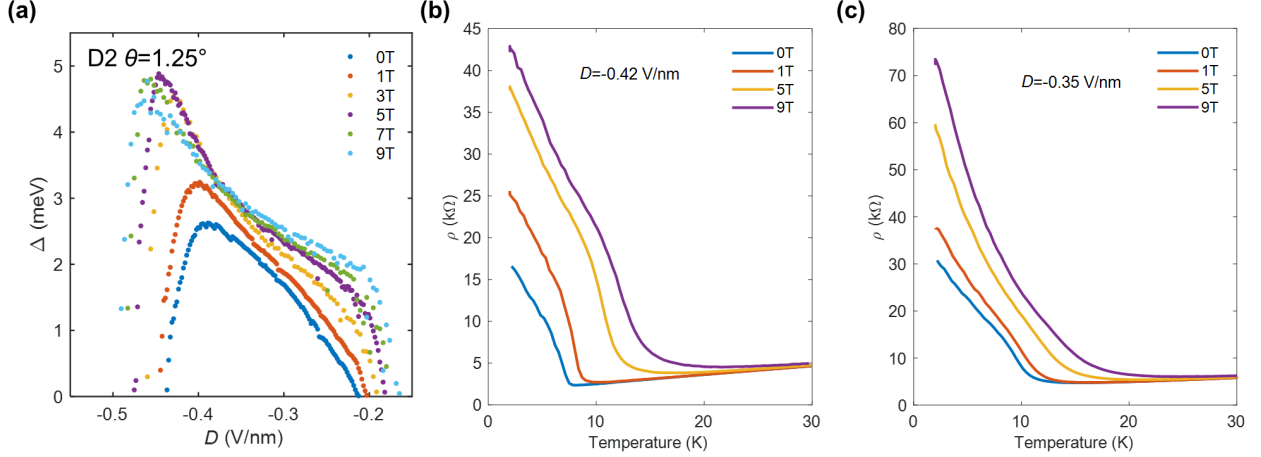


Figure 2.7: Energy gaps of the $\nu = 2$ CI state with $B_{||}$ in a $\theta = 1.25^\circ$ tDBG. **a, Gaps measured by thermal activation, with examples of corresponding $\rho(T)$ in **b**, at $D = -0.42$ V/nm and **c**, at $D = -0.35$ V/nm. Kinks develop in $\rho(T)$ for larger $|D|$, indicating a departure from simple activated transport.**

similar transport has previously been observed in certain bulk crystals owing to reduced scattering associated with the emergence of various types of magnetic ordering [98], including in MnSi [99] and BaFe₂As₂ [100]. The abrupt reduction in ρ may also be associated with reduced inelastic scattering in the symmetry-broken band or from variations in the strength of electron-phonon coupling. Further theoretical analysis is necessary to provide a detailed quantitative understanding of these observations.

2.2 Pressure tuning of the electronic correlation in tDBG

We have thoroughly discussed the symmetry-broken states in a $\theta = 1.30^\circ$ tDBG sample in the previous section. The size of the halo-like symmetry-broken states in the $\nu - D$ phase diagram reflects the underlying electronic correlation strength, which depends sensitively on the twist angle (see in Appendix A.1). In addition to the rotational alignment of the crystal axis, the interlayer coupling strength of a graphene superlattice is also very sensitive to the interlayer distance $d_{\perp}$. Thanks to the out-of-plane weak binding of the Van der Waals materials, hydrostatic pressure can easily compress the interlayer distance by 5% with less than 2.5 GPa [101]. Previous works in tBLG have predicted pressure-dependent magic-angle condition for maximized correlation strength[102; 103]. This is immediately confirmed in experiments where superconductivity is induced in a $\theta = 1.27^\circ$ tBLG under pressure[46].

Similar effect has been theoretically predicted in tDBG[104], yet waiting to be explored experimentally.

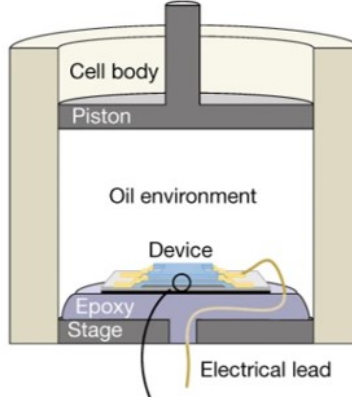


Figure 2.8: Schematic of the high-pressure experimental setup. The tDBG device is mounted inside a piston-cylinder pressure cell. Hydrostatic pressure is applied onto the sample through oil as pressure media. We measure the applied pressure strength by measuring ruby fluorescence. The yellow line embedded in epoxy represents the electrical feedthrough.

In this section, we will study the pressure tuning effects of electronic correlation strength in tDBG. We apply hydrostatic pressure up to 2.5 GPa using a piston-cylinder pressure cell. We observe pressure induced modification of the tDBG's single particle band structure. In addition, we observe monotonic suppression of the correlation strength with increasing pressure in a $\theta = 1.17^\circ$ and $\theta = 1.25^\circ$ tDBG device. We briefly discuss how the interlayer coupling strength affects the strongly correlated states in twisted multilayer graphene.

Figure 2.8 shows the schematic of the high-pressure setup. We fabricate tDBG devices following the standard procedure, except that we use a pre-diced 285nm SiO_2/Si wafer for the precise control of the sample size. The sample is carefully mounted into a piston-cylinder pressure cell, which is then inserted into a variable temperature insert (VTI) fridge.

We have studied two tDBG samples at different twist angles. Figure 2.9a shows the resistivity ρ of a $\theta = 1.17^\circ$ sample at ambient pressure. We observe displacement field D tuned band insulators at $\nu = 0, \pm 4$, and the cross-like van Hove singularities feature in the flat valence bands, consistent with our discussion in the previous section. In the flat conduction bands, we also observe the correlation driven gaped states at $\nu = 2$ and its surrounding halo-like symmetry-broken metallic states. However, compared with the $\theta = 1.30^\circ$ tDBG sample, we see that the $\nu = 2$ correlated insulator only extends to a small range in between $0.35 \text{ V/nm} < |D| < 0.4 \text{ V/nm}$. Moreover, the halo-like symmetry-broken metallic states shrink in the

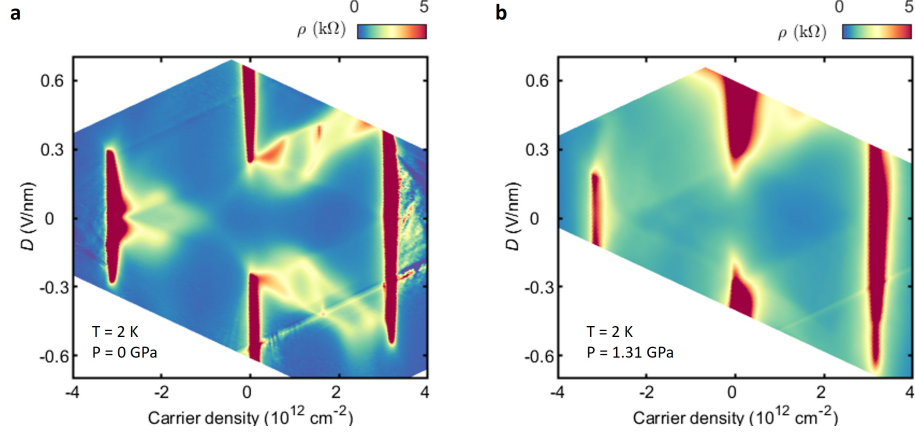


Figure 2.9: Pressure tuned transport in a $\theta = 1.17^\circ$ tDBG device. a-b, Resistivity ρ of a $\theta = 1.17^\circ$ tDBG device at, a, 0 GPa and b, 1.31 GPa. Data is acquired at $T = 2$ K.

$\nu - D$ phase diagram. These features suggest that the electronic correlation is weaker in the $\theta = 1.17^\circ$ sample compared to the sample at $\theta = 1.30^\circ$. Figure 2.9b shows the resistivity ρ of the same $\theta = 1.17^\circ$ sample after applying a hydrostatic pressure of 1.31 GPa. We note that the range of the accessible $\nu - D$ phase diagram changes slightly as a result of the pressure modified dielectric constant of the encapsulating h-BN [101]. Both the $\nu = 2$ correlated insulator and its surrounding symmetry-broken states are missing in the resistivity measurement after pressure. Their disappearance suggests that the electronic correlation strength is strongly suppressed with increasing pressure at this particular angle. Additionally, We see that the cross-like van Hove singularities feature is also suppressed in the valence bands. Instead, the sample exhibits a diamond like feature after pressure, symmetric in the conduction and valence bands, at low displacement field D . Although the band insulators at $\nu = 0, \pm 4$ remain robust after pressure, their displacement field D range changes. Altogether, these results show that pressure can strongly modify the tDBG's band structure.

Figure 2.10a-c show the resistivity ρ map of a second $\theta = 1.25^\circ$ tDBG device at ambient pressure, $P = 1.17$ GPa and $P = 2.41$ GPa respectively. At this twist angle, the electronic correlation strength at ambient pressure is strong and near optimal, suggested by the clear $\nu = 2$ over a wide range of D and the well-developed halo-like symmetry-broken states. At $P = 1.17$ GPa, the $\nu = 2$ correlated insulator vanishes entirely while a resistive hump in a similar halo-like shape persists. It occupies a shrunk phase space compared with the symmetry-broken metallic states at ambient pressure. The halo-like resistive hump is eventually suppressed at $P = 2.41$ GPa. We can gain more insight of the shrunk halo-like phase space

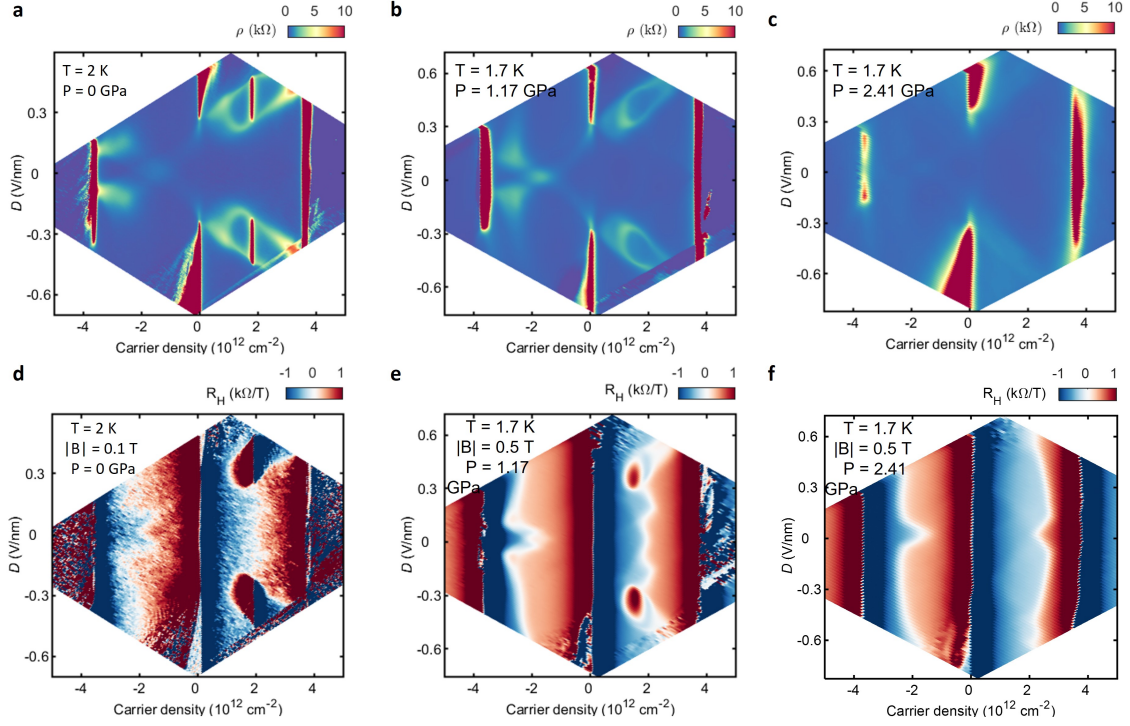


Figure 2.10: Pressure tuned transport in a $\theta = 1.25^\circ$ tDBG device. **a-c**, Resistivity ρ of a $\theta = 1.25^\circ$ tDBG device at, **a** 0 GPa, **b** 1.31 GPa, and **c** 2.41 GPa. All data is acquired at zero magnetic field. **d-f**, Corresponding Hall coefficient R_H in the same device at, **d** 0 GPa, **e** 1.31 GPa, and **f** 2.41 GPa.

by looking into the field anti-symmetrized Hall coefficient R_H (Figure 2.10d-f). Similar to our discussion in the previous section, we see that the phase space of the ρ resistive hump in the flat conduction bands overlaps well with the additional carrier type reversal in R_H map. This indicates the shrunk halo-like phase at $P = 1.17$ GPa to be the same symmetry-broken metallic states as at ambient pressure, but only with a reduced electronic correlation strength. This assignment is further confirmed by performing temperature dependent transport measurements inside the shrunk halo-like phase (see Figure 2.11). We observe similar rapid resistance drop at $P = 1.17$ GPa with a noticeably reduced critical temperature. At $P = 2.41$ GPa, we observe neither additional carrier type reversal in R_H map nor rapid resistance drop in temperature dependent transport, indicating a full suppression of the symmetry-broken metallic states.

With increasing pressure, we observe a gradual and consistent trend of the correlated states' suppression in the $\theta = 1.25^\circ$ tDBG sample. In addition, the underlying single particle band structure is also strongly modified by pressure, as can be seen from the change of the cross-like van Hove singularities feature as well as the $\nu = 4$ band insulator. At $P = 2.41$ GPa, the van Hove singularities at the flat conduction bands and

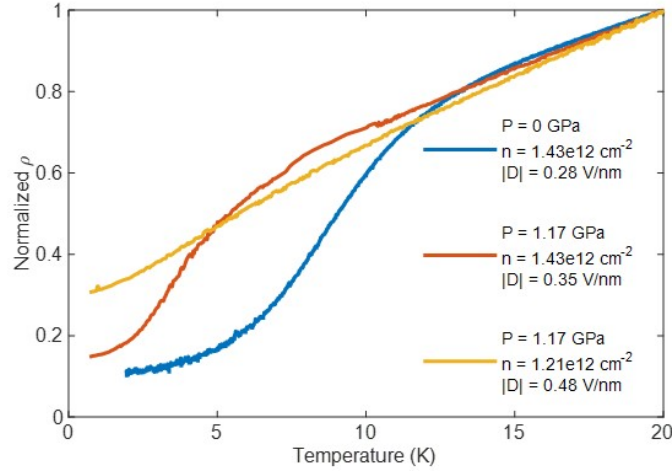


Figure 2.11: Temperature dependent transport tuned by pressure. Normalized resistivity ρ as a function of temperature T in a $\theta = 1.25^\circ$ tDBG device, measured at different pressure. The exact ν and D where the data is taken are labelled by the legend. The resistivity ρ is normalized respect to its value at $T = 20$ K.

valence bands exhibit strong particle-hole symmetry, in contrast to the apparent particle-hole asymmetry at low pressure (Figure 2.11f). At $D = 0\text{V}/nm$, the $\nu = 4$ band insulator transitions to a metallic ground states at $P = 2.41$ GPa.

In summary, we demonstrate in both tDBG samples, with a twist angle smaller or near the optimal angle at ambient pressure, that pressure can strongly modify the band structure and suppress the electronic correlation in the conduction bands. The reduction of electronic correlation, in turn, serves as evidence of increased conduction bands' bandwidth. Our experimental results align well with previous theoretical works [104], where the conduction bands' bandwidths are predicted to increase with pressure at twist angle smaller than the optimal angle. Most importantly, these results inspire further experiments focusing on tDBG with a twist angle larger than the ambient pressure optimal angle, where non-monotonic change of bands' bandwidths are predicted and enhancement of electronic interaction is expected.

2.3 Symmetry-broken Chern insulators and anomalous Hall effect

In $(M+N)$ twisted multilayer graphene, comprising rotated sheets of M- and N-layer chirally stacked graphene, an additional degree of freedom emerges in which the two multilayer graphene sheets can be stacked with either the same or opposite chirality. The band structure and topology of twisted multilayer

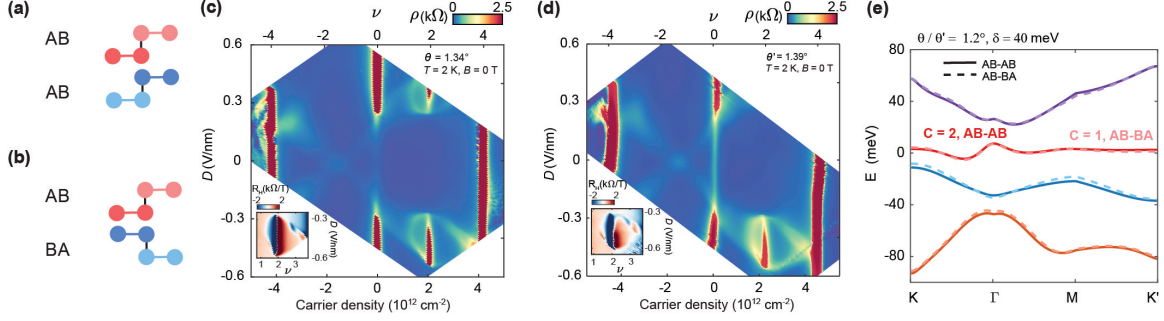


Figure 2.12: Twisted bilayer graphene in two stacking chiralities. **a-b**, Side-view lattice schematics of AB-AB and AB-BA tDBG. AB-AB (AB-BA) tDBG is composed of two Bernal stacked graphene bilayers with the same (opposite) stacking chirality. **c-d**, Resistivity ρ_{xx} maps of $\theta = 1.34^\circ$ (AB-AB) and $\theta' = 1.39^\circ$ (AB-BA) tDBG samples, respectively. Measurements are taken at $T = 2 \text{ K}$ with zero magnetic field. Insets shows the zoomed-in Hall coefficient R_H at $|B| = 0.5 \text{ T}$, focusing on the correlated phases.

graphene are predicted to depend sensitively on the chirality of the stacking [105; 89; 17; 106]; however, this has yet to be explored experimentally.

Twisted double bilayer graphene (tDBG), consisting of two sheets of Bernal-stacked bilayer graphene, can be constructed either by assembling the two bilayers with the same (AB-AB) or opposite (AB-BA) stacking chirality (Figure 2.12a-b). Whereas tDBG samples with AB-AB stacking have been studied extensively over the past few years [17; 19; 18; 16; 107; 108; 109; 110; 111; 91; 112], our study focuses on a new class of tDBG samples with AB-BA stacking constructed by rotating the two constituent bilayer graphene sheets by a small twist angle away from 60° . We denote the effective twist angle of the AB-BA tDBG by $\theta' = 60^\circ - \theta$, where θ is the actual rotated angle between the two halves of the same bilayer graphene flake. Although the band dispersions for AB-AB and AB-BA are nearly indistinguishable (Figure 2.12e), our calculations show a different distribution of the Berry curvature within the moiré Brillouin zone and correspondingly different Chern numbers of the lowest conduction band.

We perform electrical transport measurements on six AB-BA tDBG samples in which θ' varies from 1.06° to 1.39° , and compare them with AB-AB samples over a similar range of twist angles, denoted by θ . Figures 2.12c-d compare measurements of the resistivity, ρ_{xx} , of AB-AB and AB-BA tDBG samples as a function of carrier density, n , and displacement field, D . The filling factor of the moiré minibands, ν , is shown on the top axis, where $\nu = \pm 4$ corresponds to full filling of the lowest moiré conduction and valence bands (see Methods). The samples have similar twist angles of $\theta = 1.34^\circ$ (device S1) and

$\theta' = 1.39^\circ$ (device O1), respectively. The phase diagram of AB-AB tDBG has already been detailed extensively elsewhere [17; 19; 18; 16; 107]. In short, we observe gaps at $\nu = 0$ and ± 4 tuned by D , a cross-like resistive feature for $\nu < 0$ reflecting van Hove singularities in the valence bands, and a halo-like feature surrounding the insulating state at $\nu = +2$, marking the emergence of symmetry-broken correlated metallic and insulating states. Measurement of the Hall coefficient, $R_H = (R_{xy}[B] - R_{xy}[-B])/(2B)$, further reveal symmetry-broken states at $\nu = +3$ bounded by the tiny halo-like sign-reversal in R_H . The primary transport features observed in AB-BA samples are very similar at zero magnetic field and also in the Hall coefficient measurement. Their similarities are shared across a large range of twist angle, consistent with the nearly identical band dispersions in calculations (Figure 2.12e).

Our primary finding in the AB-BA tDBG samples is the symmetry-broken Chern insulators that emerges at high magnetic field. Figure 2.13a-b shows the evolution of ρ_{xx} and ρ_{xy} for AB-BA device O1 in a large magnetic field up to 12 T. We observe a variety of gapped states that evolve with a linear trajectory as described by the Streda formula, $C = \frac{h}{e} \frac{\partial n}{\partial B}$ [79]. These states are characterized by strongly suppressed ρ_{xx} and (nearly) quantized ρ_{xy} , with linear trajectories captured by the Diophantine equation $\nu = Cn_\phi + s$ [78], where n_ϕ is the number of magnetic flux quanta per moiré cell, the integer C is the Chern number, and the s is the band filling index, giving the number of electrons per moiré unit cell at $B = 0$. Figure 2.13c shows a schematic denoting the most robust gapped states observed. States with $C \neq 0, s = 0$, equivalent to integer quantum Hall states (IQH), are indicated in purple, while states with $C \neq 0$ and s a nonzero integer, or Chern insulators (CI), are indicated in orange, red, or blue depending on the value of s . Topologically trivial ($C = 0$) insulating states are indicated in black.

Both IQH and CI can be described by the Hofstadter butterfly energy spectrum [77; 78; 80; 81; 82] without taking Coulomb interaction into consideration. In addition to the above states with integer s , we observe a robust gapped state with $C = 1$ and but a fractional band filling index $s = 7/2$ above 6 T with quantized $\rho_{xy} = h/e^2$, indicated by the green dashed line. Its thermal activation gap is measured to be 0.5 meV at $B = 8$ T, about one sixth of the nearby $C = 2, s = 3$ Chern insulator state (Figure 2.15d insets). The Landau Fans taken at other D show that the gapped state persists over a relatively wide range of D . The same $C = 1, s = 7/2$ state is also observed in two other AB-BA tDBG at $\theta' = 1.33^\circ$ and $\theta' = 1.38^\circ$. While more exotic scenario involving fractionalized charge remains possible [113], the states with $C \in \mathbb{Z}$

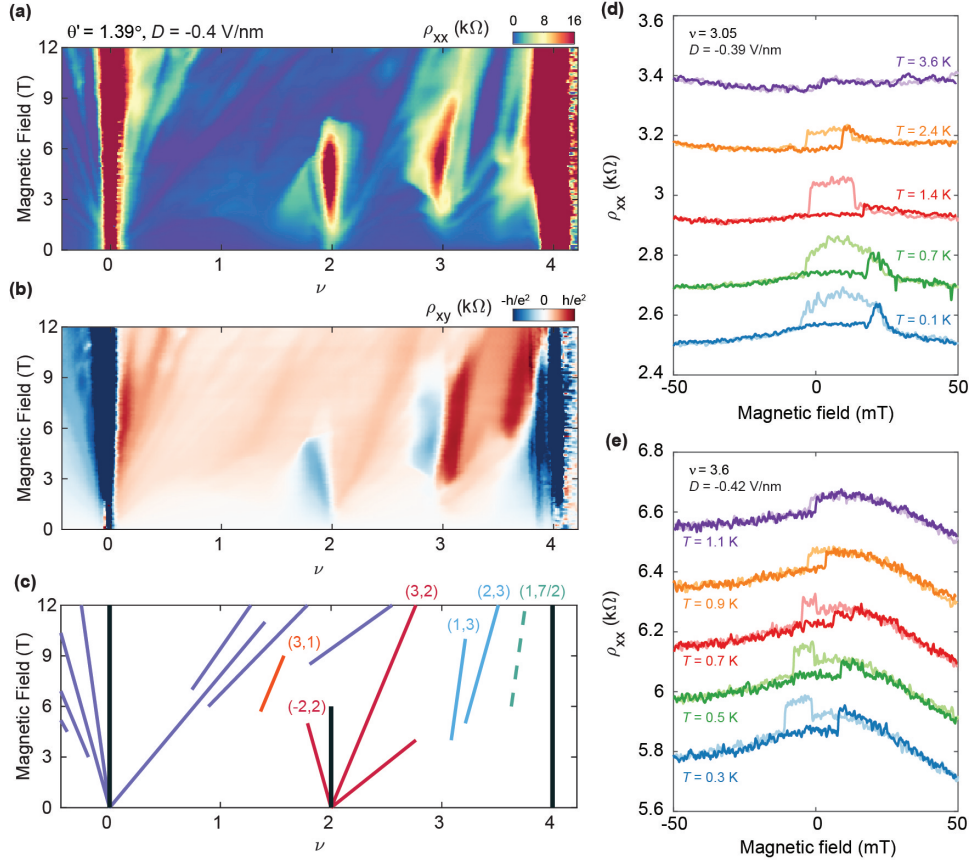


Figure 2.13: $C = 1, s = 7/2$ symmetry-broken Chern insulator and anomalous Hall effect. **a-b**, Landau fan diagram of the longitudinal (ρ_{xx}) and Hall (ρ_{xy}) resistivity at $D = -0.4$ V/nm in a $\theta' = 1.39^\circ$ AB-BA tDBG. Data is taken at 100 mK. **c**, Schematics of all observed gapped states in panel **a-b**. Several main gapped states are labelled by their respective (C, s) values. Colors differentiate different values of s . Black vertical lines (with $C = 0$) denote topologically trivial insulating states. The green dashed line denotes the $(1, 7/2)$ symmetry-broken Chern insulator. **d-e**, ρ_{xx} measured as magnetic field is swept back and forth at **d**: $\nu = 3.05$, $D = -0.39$ V/nm and **e**: $\nu = 3.6$, $D = -0.42$ V/nm in the same $\theta' = 1.39^\circ$ AB-BA sample. The data taken at different temperatures are offset vertically for clarity.

but $s \notin \mathbb{Z}$ are generally known as symmetry-broken Chern insulators (SBCI) [83; 59; 113]. In addition to the fully lifted spin and valley symmetry at high magnetic field, the strong electronic interaction further breaks the moiré translational symmetry, doubling the moiré unit cell. The SBCI state thus can be understood as a gapped state with seven electrons bounded to a doubled moiré unit cell.

Moreover, we observe an anomalous Hall effect (AHE) in the same AB-BA device O1 at low magnetic field. Figure 2.13d shows measurements of a nominally ρ_{xx} at $\nu = 3.05$, $D = -0.39$ V/nm, as magnetic field is swept back and forth at selective temperature. The ρ_{xx} shows largely AHE behavior as purely in ρ_{xy} likely due to the moiré disorder-induced domain structure, which appears to change in between different thermal cycles. Nevertheless, we observe an AHE hysteresis loop develops below 3 K and acquires multiple Barkhausen jumps at lower temperature, indicating a complicated magnetic domains structure [68]. The reason for the observed small AHE amplitude, compared to a quantized value of h/e^2 , is two fold. The first being self-explanatory in the disordered domain structure, the second reason is that its parent symmetry-broken ground state at $\nu = 3$ remains metallic to the instrument's base temperature below 50 mK. As with the SBCI state, we also observe AHE in two other AB-BA tDBG at $\theta' = 1.33^\circ$ and $\theta' = 1.38^\circ$.

In device O1, we additionally observe the AHE at some filling factors far away from integers, as illustrated in Figure 2.13e for $\nu = 3.6$, $D = -0.42$ V/nm, but with a lower critical temperature around 1.2 K. To examine the doping dependence of the AHE, we map the difference in ρ between the two field sweep directions, $\rho = \rho^{B\downarrow} - \rho^{B\uparrow}$, with $D = -0.42$ V/nm (see Figs. 2.14c). We observe two distinct pockets of clear AHE, one around $\nu = +3$ and one around $\nu = +3.5$ with different optimal D . Although the two pockets appear to merge and extend over a wide range of ν for some specific values of D , their typical separation in both doping and displacement field suggests that they might associate with distinct correlated ground states. At zero magnetic field, correlated states at fractional filling have been observed previously in many other graphene moiré, including twisted monolayer-bilayer graphene [113] (tMBG) and twisted bilayer graphene (tBLG) aligned with h-BN [63]. These states may be associated with a spontaneous charge density wave (CDW) formation that expands the effective unit cell, or potentially with a moiré lattice-driven charge fractionalization [114; 115; 116; 117; 118]. To explore the possibility of a spontaneous CDW formation, we performed a modified continuum model calculation generalized to include a CDW order (Appendix B.1). This model predicts that the lowest moiré conduction band splits into two gapped subbands upon inclusion

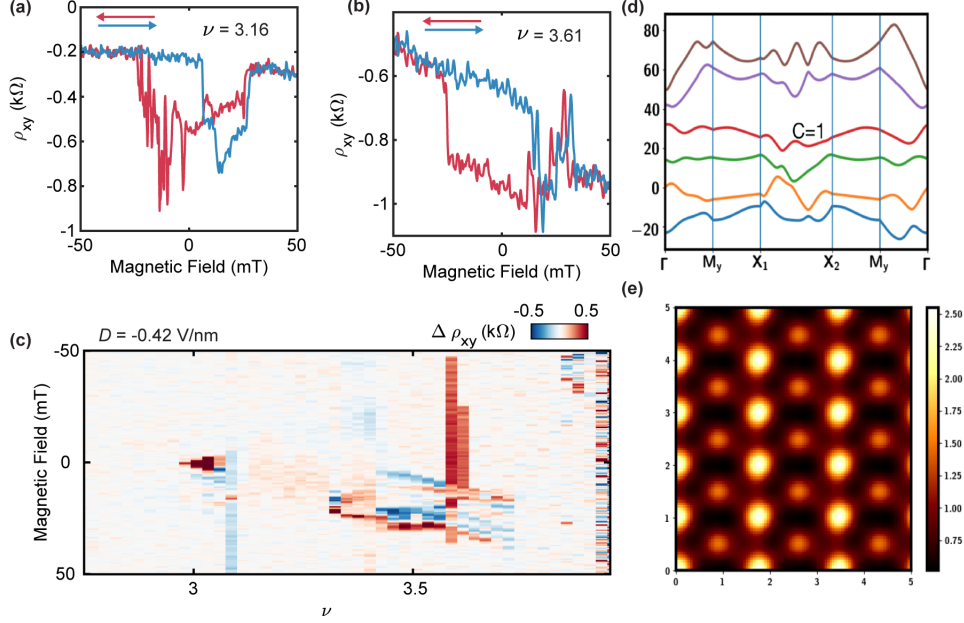


Figure 2.14: Anomalous Hall effect (AHE) near integer filling $\nu = 3$ and fractional filling $\nu = 7/2$. **a-b**, ρ_{xy} as magnetic field is swept back and forth at $\nu = 3.16$, $D = -0.44$ V/nm, exhibiting anomalous Hall effect. **b**, ρ_{xy} as magnetic field is swept back and forth at $\nu = 3.61$, $D = -0.42$ V/nm, showing AHE near $\nu = 7/2$. **c**, $\Delta\rho_{xy}$ as a function of magnetic field measured at different filling factor at fixed $D = -0.42$ V/nm, $T = 100$ mK. We define $\Delta\rho_{xy} = \rho_{xy}^{B\downarrow} - \rho_{xy}^{B\uparrow}$ to visualize the AHE. The AHE near $\nu = 3$ and $\nu = 7/2$ are isolated, implying their independent correlated ground states. **d**, Calculated band structure reconstructed with $\phi = 16$ meV charge density wave (CDW) order. The red bands are the two splitted subbands from the conduction flat band with a CDW gap $\Delta = 2$ meV. Chern number are labelled to each conduction subband. **e**, Calculated charge density profile after introducing the CDW order with $\phi = 16$ meV. A gauge is chosen to pin the maximum of the CDW order to AA site. a_M is the moiré lattice constant.

of the density wave order (Fig. 2.14d), with a corresponding electron density profile shown in Fig. 2.14e. After considering remote band mixing, we calculate a valley Chern number of $C_v = 1$ for the higher-energy subband, consistent with our observation of a topological correlated state near $\nu = 7/2$.

We observe both $C = 1$, $s = 7/2$ SBCI and AHE near $\nu = 3$ in all AB-BA samples with $1.33^\circ < \theta' < 1.39^\circ$ (three out of three). It turns out these correlated and topological states are also ubiquitous in AB-AB samples. Figure 2.15a-b shows the ρ_{xx} and ρ_{xy} at high magnetic field measured in a second AB-AB sample at $\theta = 1.34^\circ$. The schematic of the main gapped states is plotted in Figure 2.15c, following the convention in Figure 2.13. In addition to the IQHs and CIs, we observe a $C = 1$, $s = 7/2$ SBCI above 5 T, similar to the AB-BA samples. Moreover, a second $C = 1$, $s = 11/3$ SBCI emerges above 9 T. Figure 2.15d shows a line cut of ρ_{xx} and ρ_{xy} at $B = 10T$, where the two SBCI states are emphasised by the green shades and the

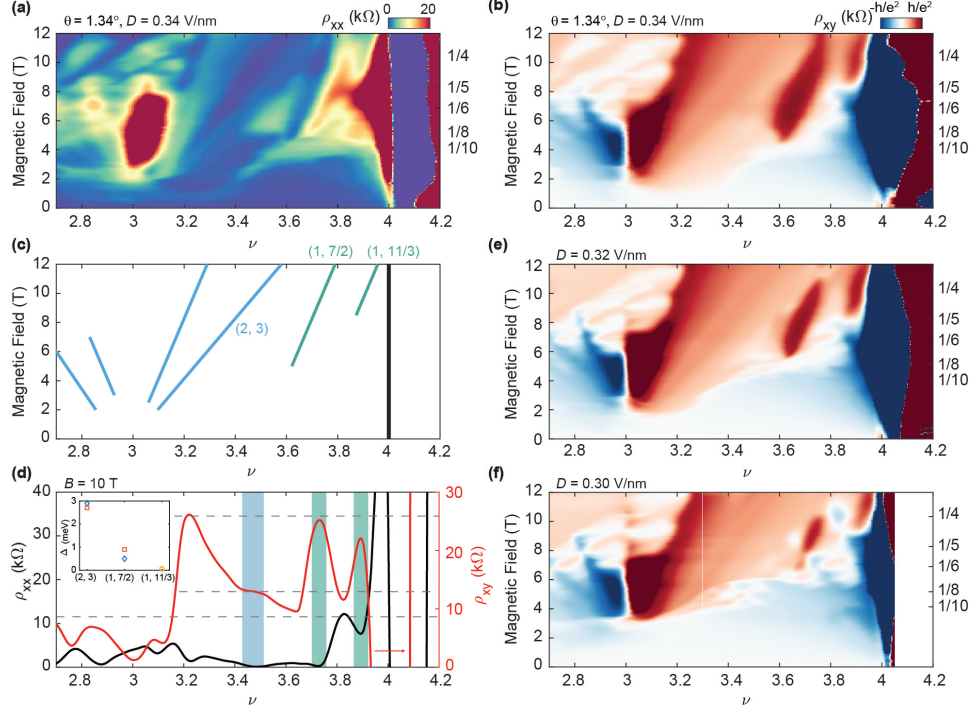


Figure 2.15: $C = 1, s = 11/3$ symmetry-broken Chern insulator. **a-b**, Landau fan diagram of the longitudinal (ρ_{xx}) and Hall (ρ_{xy}) resistivity at $D = 0.34$ V/nm in a $\theta = 1.34^\circ$ AB-AB tDBG. Data is taken at 100 mK. **c**, Schematics of main observed gapped states in panel **a-b**, using the same color coding as in Fig. 2.13. The green dashed lines denote the $(1, 7/2)$ and the $(1, 11/3)$ SBCIs. **d**, Linecut of ρ_{xx} and ρ_{xy} in panel **a-b** at $B = 10$ T. The SBCIs are emphasised by the green shades. The nearby quantized $(2, 3)$ Chern insulator is marked by the blue shade. The inset shows the thermal activation gap of $(2, 3)$, $(1, 7/2)$ and $(1, 11/3)$ states. The blue diamonds denote data from the $\theta' = 1.39^\circ$ AB-BA sample at $D = -0.4$ V/nm with $B = 8$ T. The orange square denote data from the $\theta = 1.34^\circ$ AB-AB sample at $D = 0.4$ V/nm with $B = 9$ T. The yellow circle denotes data from the same $\theta = 1.34^\circ$ AB-AB sample at $D = 0.34$ V/nm with $B = 9$ T. **e-f**, Hall resistivity ρ_{xy} at **e**: $D = 0.32$ V/nm and **f**: $D = 0.30$ V/nm in the same $\theta = 1.34^\circ$ AB-AB sample. All Data is acquired at 100 mK.

quantized $C = 2, s = 3$ Chern insulator is marked by the blue shade. The ρ_{xy} at the $C = 1, s = 11/3$ only reach 80% quantization at base temperature, consistent with its small thermal activation gap about 0.06 meV (at $B = 9$ T, Figure 2.15d inset) and its onset with higher magnetic field. The fractional band filling index $s = 11/3$ can be understood as eleven electrons bound with a tripled moiré unit cell. The two SBCI states, albeit their proximity in the phase diagram, have distinct translational symmetry in the basis of a doubled and tripled moiré unit cell respectively.

Measurements at other D show that the $C = 1, s = 7/2$ SBCI does not monotonically strengthen with increasing field, but only appears within a finite range of magnetic field dependent on D (Figure 2.15e-

Twist angle	Sample type	$\nu = 2$	$\nu = 3$	AHE	SBCI
1.06	AB-BA	None	None	/	/
1.17	AB-BA	halo only	None	/	No
1.17	AB-AB	CI with halo	None	/	/
1.25	AB-AB	CI with halo	None	/	/
1.28	AB-BA	CI with halo	None	/	No
1.30	AB-AB	CI with halo	CI with halo	No AHE	No
1.33	AB-BA	CI with halo	Resistive state with halo	AHE	(1, 7/2)
1.34	AB-AB	CI with halo	Halo only	/	/
1.34	AB-AB	CI with halo	Halo only	AHE	(1, 7/2) and (1, 11/3)
1.38 (h-BN aligned)	AB-BA	CI with halo	Halo only	AHE	(1, 7/2)
1.39	AB-BA	CI with halo	Halo only	AHE	(1, 7/2)
1.41	AB-AB	Resistive state with halo	None	No AHE	No
1.53	AB-AB	None	None	/	/

Table 2.1: Correlated and topological states in tDBG. Summary of the properties of the correlated states and topological states in 13 tDBG samples, some of the AB-AB samples are reported in our previous work [].

f). This indicates the underlying Coulomb interaction is D tunable and likely magnetic flux ratio ϕ/ϕ_0 dependent (ϕ is the magnetic flux through one moiré unit cell, $\phi_0 = h/e$ is one magnetic quantum flux), as labelled on the right y axis. At $D = 0.30$ V/nm away from optimal condition, the Coulomb interaction becomes comparatively weak and only briefly opens a $C = 1, s = 7/2$ gapped state right below $\phi/\phi_0 = 1/4$. A similar observation of SBCI states in tBLG also appears to be closely related to flux ratio $\phi/\phi_0 = 1/4$ [59] while SBCI states in graphene moiré are mainly observed near $\phi/\phi_0 = 1/2$ at higher magnetic field [83]. The relation of the SBCI, the flux ratio and the underlying Coulomb interaction requires further theoretical exploration.

Table 2.1 summarizes the correlated and topological states in 13 devices of both AB-AB and AB-BA tDBG, including those reported in previous sections, in the order of ascending twist angle. The twist angle of each device is determined by collaborating Hall effect and Landau fan fitting, with an twist angle disorder around 0.02° in between contact pairs. The cell is left blank when no measurement is taken at the particular phase. In general, the appearance of the correlated and topological states is seemingly dependent on twist angle solely, regardless of the sample stacking chirality. The $\nu = 3$ correlated insulator ground state appears only within a tight range of $1.30^\circ \leq \theta(\theta') \leq 1.34^\circ$ [107; 119], and transitions to a metallic ground state at larger twist angle $1.34^\circ \leq \theta(\theta') \leq 1.39^\circ$. There are many potential correlated ground states that could occur in tDBG at $\nu = +3$ [89], including gapped states with full spin and valley polarization [54], intervalley coherent (IVC) states with various order parameters [120], density wave states, and many other gapless

isospin polarized state [89]. Although it is difficult to differentiate between these possibilities in transport experiments unambiguously, the observed AHE implies spontaneous time-reversal symmetry (TRS) breaking occurs near $\nu = +3$. These observations should help elucidate the nature of correlated states at $\nu = 3$.

Specifically, the AHE near $\nu = 3$ and the $C = 1, s = 7/2$ SBCI is observed within $1.33^\circ \leq \theta(\theta') \leq 1.39^\circ$. Their exact twist angle range is tightly bounded by their absence in a $\theta = 1.30^\circ$ and a $\theta = 1.41^\circ$ tDBG sample. We note that while the AHE near $\nu = 3$ shows strong domain effect with measurement results varies between different contact pairs, the SBCI appears robust across all contact pairs in each sample. The sole observation of $C = 1, s = 11/3$ in single device is likely restricted by a tighter twist angle range, considering that the state is only weakly gapped and a large Coulomb interaction is needed. Both $C = 1, s = 7/2$ and $C = 1, s = 11/3$ symmetry-broken Chern insulators locate right below the conduction band edge at $\nu = +4$. Meanwhile at the same phase space, the zero field phase diagram also appears much more crowded as compared to other doping. These observation point to a stronger Coulomb interaction at the band edges, likely enhanced by the Van Hove singularities. Our experiments are based on one caveat that we assume the Bernal bilayer graphene used in fabrication to be chirally stacked without domain walls. The hall bar devices under study are usually $1\ \mu m$ by $2\ \mu m$ to avoid transport through the domain boundaries. We label the devices by their nominal stacking chirality while we point out that actual stacking order can be mixed or reversed in some devices. Our results provide statistics but make no assertion on physics related to stacking chirality.

Finally, we study an ($\theta' = 1.38^\circ$) AB-BA sample, which is also closely aligned to the top encapsulating boron nitride (hBN) crystal. Measurements of the Landau fan diagram at $D = 0$ V/nm confirm the existence of a second moiré potential arising from a twist angle of 0.55° between the uppermost graphene and hBN. This aligned hBN layer breaks the equivalence of the band structure for opposite orientations of D . As a result, the correlated states depend on the sign of D (Fig. 2.16a), unlike in the hBN-misaligned samples. For positive D , electrons are pushed away from the graphene layer at the aligned hBN interface (Fig. 2.16b) while for negative D they are pushed towards it (Fig. 2.16c). For positive D , the correlated insulating state at $\nu = 2$ resembles that in hBN-misaligned samples, while for negative D the correlated states are more complicated and exhibit new splitting features. The right-most panel of Fig. 2.16a shows the temperature dependence of ρ_{xx} as a function of D at $\nu = 2$. For $D < 0$ the correlated insulating state at $\nu = 2$ appears

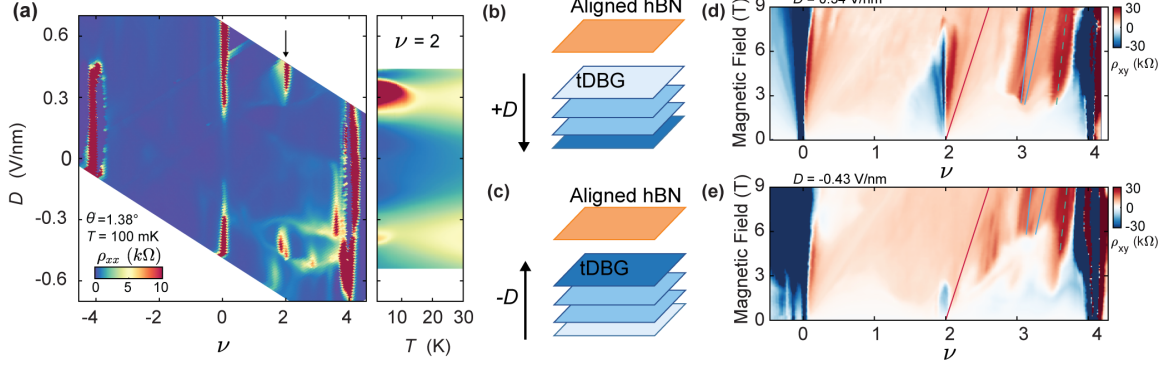


Figure 2.16: Robust SBCI with additional top hBN alignment. **a**, Resistivity (ρ_{xx}) map of a $\theta' = 1.38^\circ$ AB-BA tDBG device aligned with top hBN at a twist angle of 0.55° . The right panel shows ρ_{xx} as function of temperature and D at $\nu = 2$. **b-c**, Schematics of the layer-resolved electron density upon a positive and negative D , respectively. The electron density is schematically indicated by intensity of blue coloring. **d-e**, Hall resistivity, ρ_{xy} , measured at $D = 0.34$ V/nm (top) and $D = -0.43$ V/nm (bottom), respectively. Schematics of observed gapped states are overlaid, using the same color coding as in Fig. 2.13.

weakened, witnessed by the suppression of ρ_{xx} . Despite this, we still observe a robust $C = 1, s = 7/2$ SBCI state upon applying a magnetic field, albeit with a larger onset field than for $D > 0$ (green dashed lines in Figs. 2.16d-e). These observations underline the robustness of the SBCI at high magnetic field, as the SBCI state survives even upon weakening the correlation strength by a second set of moiré.

In conclusion, we have studied transport properties in AB-BA twisted double bilayer for the first time. We observe a ubiquitous $C = 1, s = 7/2$ symmetry-broken Chern insulator and anomalous Hall effect near $\nu = 3$ in twisted double bilayer, regardless of the stacking order. At high magnetic field, the Coulomb interaction is enhanced near flux ratio $\phi/\phi_0 = 1/4$. Our findings inspire measurements at higher magnetic field (flux ratio) where the enhanced Coulomb interaction can likely drive new gapped states, including other Symmetry-broken Chern insulator and possibly fractional Chern insulators. Our results reveal a clear trend of how the various correlated and topological states, and their underlying Coulomb interaction, evolve as a function of twist angle. This result provide a guideline to engineering the topology and the electronic interaction in twisted double bilayer graphene by pressure and strain.

Chapter 3

Correlated and topological states in twisted monolayer-bilayer graphene

In moiré heterostructures composed entirely of graphene, the correlated phase diagram depends sensitively on the number of graphene sheets. We have explored the electron correlation and topology in twisted bilayer graphene (tBLG) and twisted double bilayer graphene (tDBG) in the previous chapters. In this chapter, we focus our attention on twisted monolayer-bilayer graphene (tMBG) — heterostructures comprising rotated sheets of monolayer and Bernal-stacked bilayer graphene.

In the first part, we discuss the structure of the tMBG's correlated phase diagram. Its asymmetry under a positive or negative D reflects its low crystal symmetry, breaking both two-fold rotation symmetry, C_2 , and mirror reflection symmetry, M_y (Fig. 3.1a). In the second part, we show abundant orbital magnetism within the correlated phase diagram of tMBG. The behavior of some orbital magnetic states at zero field appears to be inconsistent with a spin and valley polarized state. Alternatively, we propose an intervalley coherent (IVC) state with an order parameter that breaks time reversal symmetry to be one plausible candidate ground state. In the last part, we show closely competing Chern insulators at high magnetic field in tMBG. We observe hysteresis while sweeping B back and forth across the boundary of different isospin flavor states on the negative D side, indicating the phase transition to be of first-order.

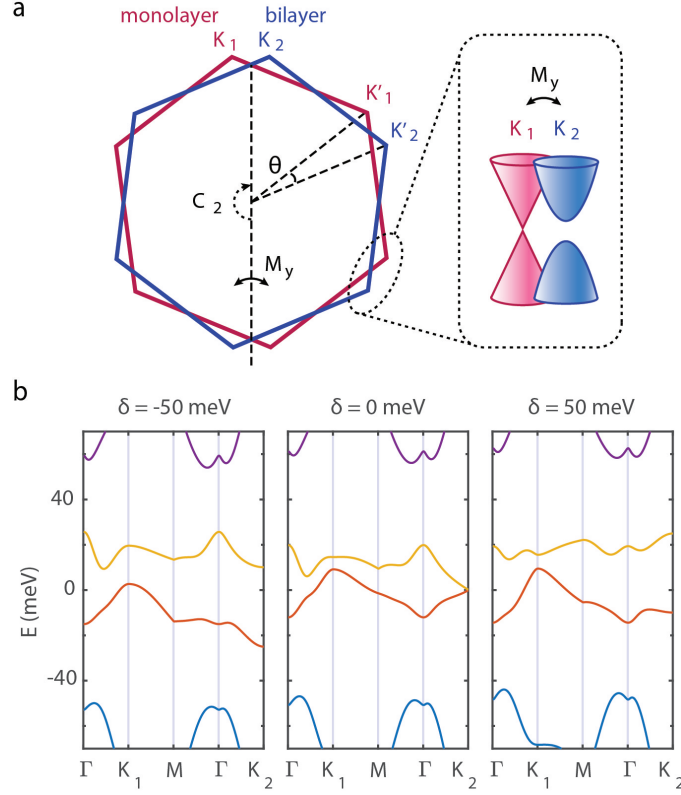


Figure 3.1: Band structure of tMBG. **a**, First Brillouin zones of monolayer (red) and bilayer (blue) graphene, rotated by a twist angle θ . Both C_2 rotation symmetry and M_y mirror symmetry are broken in tMBG. **b**, Calculated band structure of tMBG with $\theta = 1.08^\circ$ at various values of interlayer potential, δ .

3.1 Electrically tunable correlated states in tMBG

We begin with a discussion of the band structure and crystal symmetry of tMBG. Figure 3.1b shows the continuum model calculation of the band structure of tMBG at a twist angle of $\theta = 1.08^\circ$ as a function of the interlayer potential difference, δ . In contrast to tBLG and tDBG, because of the low symmetry the band structure of tMBG differs depending on the direction of D (i.e. the sign of δ). Additionally, calculations of the layer-resolved local density of states (LDOS) reveal that it is favorable for charge to be arranged highly asymmetrically amongst the three constituent graphene layers. Panels **a** and **b** of Fig. 3.2 show the LDOS of tMBG calculated at full filling of the conduction band for opposite signs of D . In both cases, the moiré potential localizes the LDOS on a triangular lattice of ABB-stacked sites (analogous to AA-stacked sites in tBLG, in which the carbon atoms sit directly atop one another). For $D < 0$ ($\delta < 0$), when the field points

from the bilayer towards the monolayer graphene, the LDOS on the outer sheet of the bilayer graphene is suppressed, and the LDOS on the two graphene layers at the rotated interface resembles that of tBLG (panel **c**). On the other hand, for $D > 0$ ($\delta > 0$), the LDOS on the outer graphene layer is instead enhanced. In this case the LDOS resembles that of tDBG (panel **d**), where the fourth (uppermost) graphene layer has a small LDOS. We would therefore anticipate that tMBG will act similarly to either tBLG or tDBG depending on the sign of D , assuming the additional or absent layer of lightly doped graphene plays a limited role.

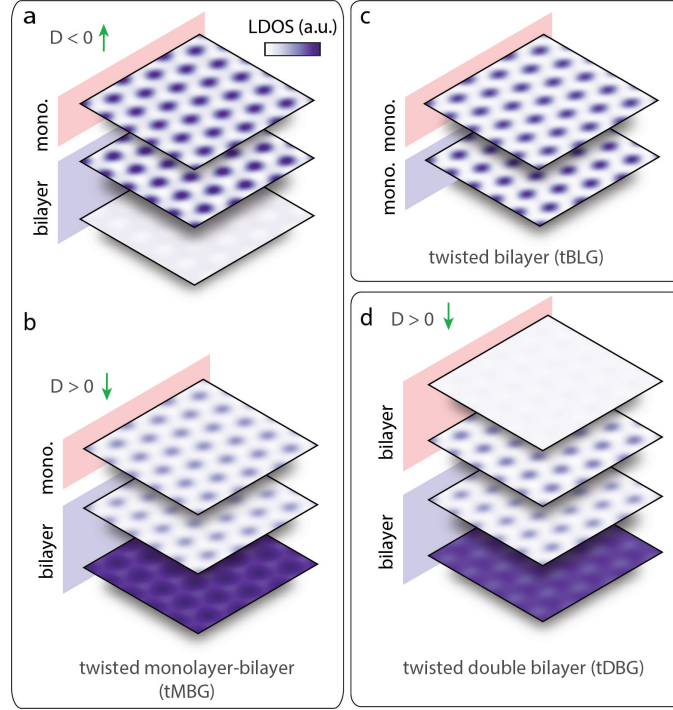


Figure 3.2: Layer-resolved local density of states in tMBG. Corresponding calculated local density of states for each layer of tMBG at full filling of the conduction band with (a) $D < 0$ and (b) $D > 0$. Green arrows indicate the direction of the electric field. They are compared with the LDOS calculated for (c) tBLG and (d) tDBG at the same twist angle. The rotated interface is between layers with different color projections (red or blue).

This picture is borne out well by the experiments. Figure 3.3a shows the resistivity, ρ , of a tMBG device with $\theta = 1.08^\circ$ as a function of carrier density, n , and D at temperature $T = 0.3$ K. The top axis indicates the filling factor of the bands, ν . Qualitatively, many of the observed features can be associated with the band structure calculated in Fig. 3.1b. In particular, at $\nu = 0$ the device behaves as a semi-metal for small $|D|$, but becomes semiconducting at larger $|D|$. Insulating features are observed over a wide range of $|D|$ at full filling of the conduction or valence band ($\nu = \pm 4$). Additional resistance peaks disperse as a function

of D within the flat bands, and likely correspond to van Hove singularities (vHs) as in tDBG [107].

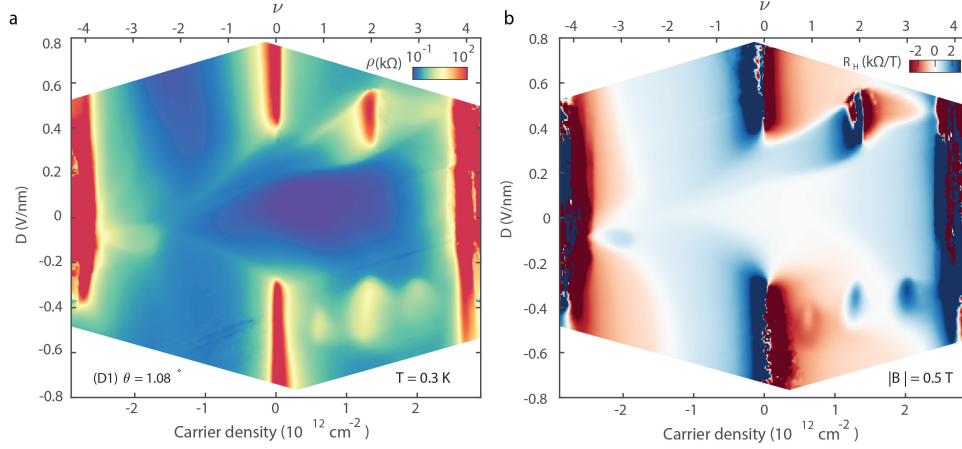


Figure 3.3: Transport in a $\theta = 1.08^\circ$ tMBG device. **a**, Resistivity of a $\theta = 1.08^\circ$ tMBG device at $T = 0.3$ K. The corresponding band filling factor ν is shown on the top axis. **b**, Corresponding Hall coefficient, R_H , antisymmetrized at $|B| = 0.5$ T.

In the conduction band ($0 < \nu < +4$), additional resistive peaks are observed over a finite range of D that appear to have no counterparts in the single-particle model. These resistive peaks are observed at integer ν , each within a different finite range of D . Notably, these features are inequivalent for opposite signs of D , in contrast with tBLG and tDBG where the correlated states are approximately symmetric with D . Measurements of the antisymmetrized Hall coefficient, $R_H = (R_{xy}[B] - R_{xy}[-B])/(2B)$, provide additional insight into their nature (Fig. 3.3b). For $D > 0$, there is a highly resistive state at half filling surrounded by a stretched halo of somewhat increased resistance. As in tDBG, R_H changes sign within this extended halo region, suggestive of a spontaneously broken symmetry within the bands [107]. In contrast, for $D < 0$ there are three weakly resistive features at each integer ν , and abrupt changes in R_H occur only near these features.

Figure 3.4a shows the temperature dependence of ρ in the conduction band at $D = +0.5$ V/nm, in which we expect the LDOS distribution to resemble that of tDBG. We observe clear insulating behavior at half filling ($\nu = 2$), with energy gaps that appear to grow upon applying an in-plane magnetic field indicative of spin-polarized ordering. Slightly away from half filling, we observe abrupt drops in ρ as the temperature is lowered (blue curve in the inset of Fig. 3.4a). These abrupt drops arise within regions of the phase diagram in which the magnitude or sign of the Hall density, $n_H \equiv 1/eR_H$, departs from single-particle

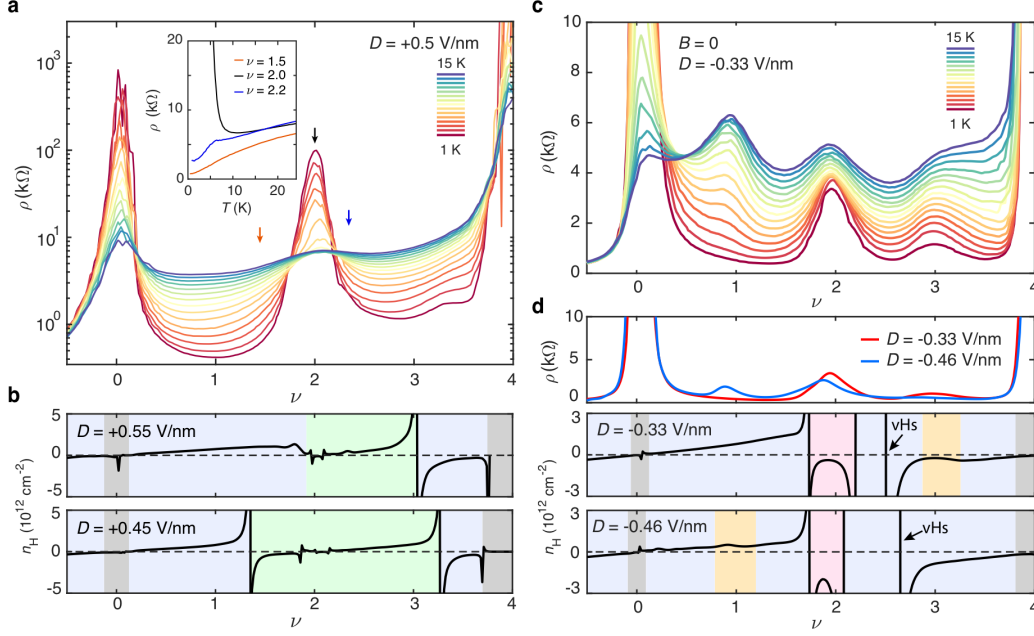


Figure 3.4: Transport characteristics of correlated states in tMBG. **a**, Temperature dependence of ρ at $D = +0.5$ V/nm. (Inset) $\rho(T)$ acquired at the filling factors denoted by the colored arrows in the main panel. **b**, Hall density, n_H , as a function of ν at various D . Color shading denotes regions of the phase diagram with different inferred degeneracy, where blue corresponds to four-fold degeneracy and green corresponds to two-fold degeneracy. Gray-shaded regions denote insulating states at $\nu = 0$ and 4. **c**, Temperature dependence of ρ at $D = -0.33$ V/nm. **d**, (Top panel) ρ vs. ν at selected D and $T = 0.3$ K. (Bottom panels) Corresponding n_H . Color shading is as in **b**, but with pink denoting regions with correlation-driven sign changes in n_H , and gold denoting regions with deviations from the anticipated gate-induced density n . An additional sign change related to the single-particle vHs is denoted by an arrow.

expectations (green shaded regions in Fig. 3.4b). This behavior has been previously observed in comparable regions of the tDBG phase diagram, and understood to likely arise as a consequence of the development of symmetry-broken correlated metallic states within the flat conduction band [107]. Overall, our observations correspond closely with the behavior of the CI and metallic states in tDBG [16; 17; 50; 19; 107], suggesting that at this twist angle the phase diagram of tMBG is similar to that of tDBG, with small differences arising owing to the absence of the weakly charged fourth graphene sheet (see Fig. 3.2d).

Figure 3.4c shows similar temperature dependence of ρ at $D = -0.33$ V/nm, in which we expect the LDOS distribution to resemble that of tBLG. The states at $\nu = 1, 2, 3$ all exhibit metallic temperature dependence, defined by a reduction in ρ as T is lowered, in direct contrast to the behavior of the CI state for $D > 0$. Notably, the feature at $\nu = 1$ is apparent at high temperature ($T = 15$ K in Fig. 3.4c),

but becomes suppressed at low temperature. Qualitatively similar behavior has been observed previously in tBLG, most clearly in devices with twist angles slightly detuned from the “magic angle” [46] and in devices with strong electrostatic screening from a nearby metal gate [121]. This suggests that our devices are marginally correlated compared to MATBG, potentially owing to a combination of larger bandwidth and electrostatic screening from the addition of the weakly charged third graphene sheet. However, unlike in tBLG, these states can be tuned with D . For example, Fig. 3.4 d shows ρ for two values of D , in which the state at $\nu = 1$ is apparent at $D = -0.46$ V/nm but not at $D = -0.33$ V/nm. These correlated metallic states are also associated with anomalies in n_H (Fig. 3.4 d); either a sign change (pink shading) or deviations from a linear gate-induced charge density, n (gold shading). An additional sign change in n_H is observed for $2 < \nu < 3$ that matches the vHs in the single-particle band structure, suggesting that correlations do not greatly modify the conduction band Fermi surface at filling factors away from the integers.

These results demonstrate that the low symmetry of tMBG enables a wide array of highly tunable correlated states that differ on changing the sign of D . Moreover, these correlated states vary significantly as a function of the twist angle. We present a detailed survey of the correlated (and topological) states over a wide range of twist angle in the Appen. B.1.

3.2 Orbital magnetism in tMBG

Similar to the case of MATBG and tDBG, the tight-binding calculation shows that the spin and valley degenerate flat bands in tMBG also carry non-zero Chern numbers [122; 123; 124]. In this section, we discuss the abundant orbital magnetic states in tMBG that arise from this non-trivial band topology. In the simplest case, an orbital magnetic state in graphene moiré is expected to arise upon isospin polarization into a single spin and valley polarized band precisely at odd integer ν . We show observation of magnetic states both within and beyond the scope of the spin and valley polarized ground state.

3.2.1 Electrically tunable orbital magnetism in a $\theta = 0.89^\circ$ tMBG device

We first focus on orbital magnetism observed in a $\theta = 0.89^\circ$ tMBG device. Figure 3.5 shows the resistivity of a $\theta = 0.89^\circ$ tMBG device at $T = 50$ mK and $B = 0$ T. The overall behavior is similar to the $\theta = 1.08^\circ$ device discussed in previous section, with a few key exceptions. First, the insulating behavior

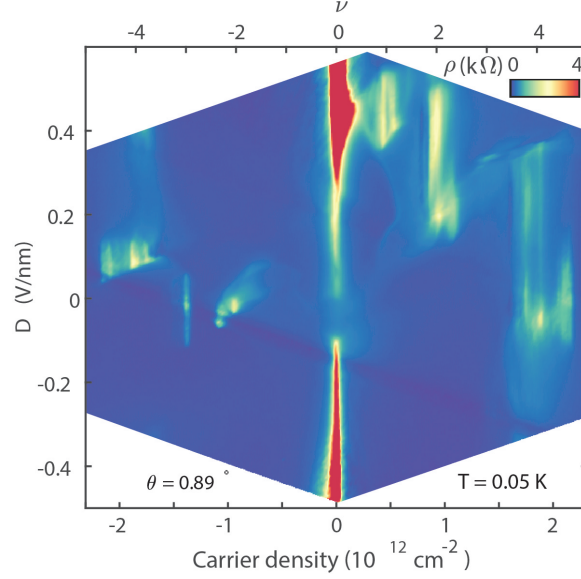


Figure 3.5: Transport in a $\theta = 0.89^\circ$ tMBG device. Resistivity of a $\theta = 0.89^\circ$ tMBG device at $T = 50$ mK. The corresponding band filling factor ν is shown on the top axis.

is weak or absent at $\nu = \pm 4$, indicating the energy gaps to the remote bands are small or zero at this twist angle. Second, incipient CI states can be seen at $\nu = -2$ and -3 for $D \approx 0$. Third, an additional resistive state emerges at $\nu = 1$ over a small range of $D > 0$.

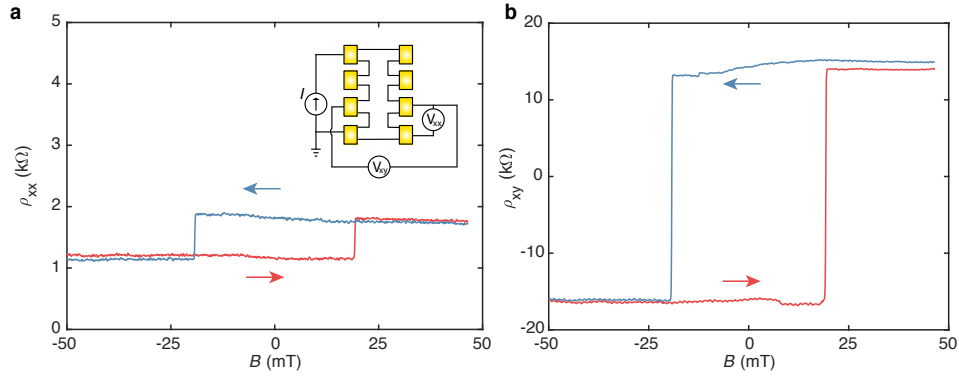


Figure 3.6: AHE in the $\theta = 0.89^\circ$ tMBG device. ρ_{xx} and ρ_{yx} raw data acquired as $B_\perp$ is swept back and forth in the $\theta = 0.89^\circ$ tMBG device, at $\nu = 0.94$ and $D = +0.415$ V/nm. The inset shows the measurement setup. We observe a robust AHE at this state as shown by the hysteresis loops.

This state exhibits a robust AHE with large Hall angle $\rho_{yx}/\rho_{xx} \approx 10$ at $B = 0$ (Fig. 3.6). A single loop with a coercive field of $B_\perp \approx 20$ mT is formed as the field is swept back and forth. ρ_{xx} exhibits a single loop alike ρ_{yx} but with a smaller loop size, due to mixing effect. This results from the imperfect alignment

of the longitudinal contacts. It can be a particularly strong effect when ρ_{xx} is relatively small whereas ρ_{yx} is sizeable. Therefore, we field symmetrize ρ_{xx} and antisymmetrize ρ_{yx} to eliminate the mixing (Fig. 3.7 a). The temperature dependence of the field antisymmetrized ρ_{yx} is shown in the bottom panel of Fig. 3.7a. The coercive field and magnitude of the AHE loop are suppressed upon raising the temperature, and vanish above $T \approx 2.5$ K. The sharpness of the magnetization flip implies that the easy axis is perpendicular to the sample, and that a single magnetic domain is dominant within the measured region of the sample.

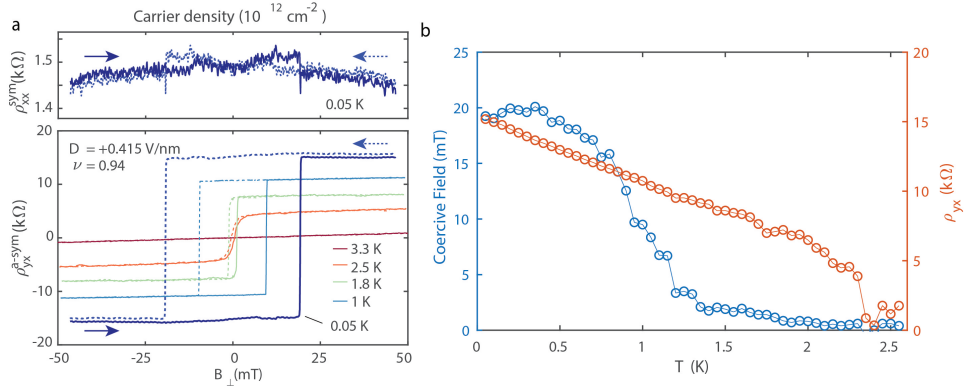


Figure 3.7: Temperature dependence of AHE in a device with $\theta = 0.89^\circ$. **a**, Symmetrized ρ_{xx} and antisymmetrized ρ_{yx} acquired as $B_\perp$ is swept back and forth at $\nu = 0.94$ and $D = +0.415$ V/nm with $T = 0.05$ K (blue curves). ρ_{yx} is additionally shown at different temperatures. Arrows indicate the direction of the magnetic field sweep, corresponding to solid and dashed curves. **b**, Temperature dependence of the AHE resistivity and coercive field. Measurements taken at the exact state shown in **a** at different temperature.

Figure 3.8a shows a map of ρ_{yx} antisymmetrized at $|B| = 50$ mT. The regime of ferromagnetic order as a function of ν and D is indicated by the dark red at $\nu \approx 1$. The ferromagnetism occurs over an extended range of D , and persists into the metallic states slightly outside of the $\nu = 1$ gap. We do not observe any signs of ferromagnetic ordering at any other filling factors in this device. Despite the fact that ρ_{yx} is not quantized to an integer fraction of h/e^2 , and ρ_{xx} does not vanish, the large Hall angle indicates transport through edge states. Nonlocal resistance measurements provide further evidence supporting chiral edge conduction in our sample (see Fig. 3.8b) [125; 53]. It seems likely that a fully developed Chern insulator state could be realized in a sample with lower disorder or a larger energy gap.

In addition to switching the magnetic ordering with $B_\perp$, we are able to control the magnetic state purely with charge doping at $B = 0$. Figure 3.9a shows ρ_{yx} as the doping is swept back and forth, exhibiting opposite signs of the magnetic ordering for each sweep direction. This doping-induced switching effect can be

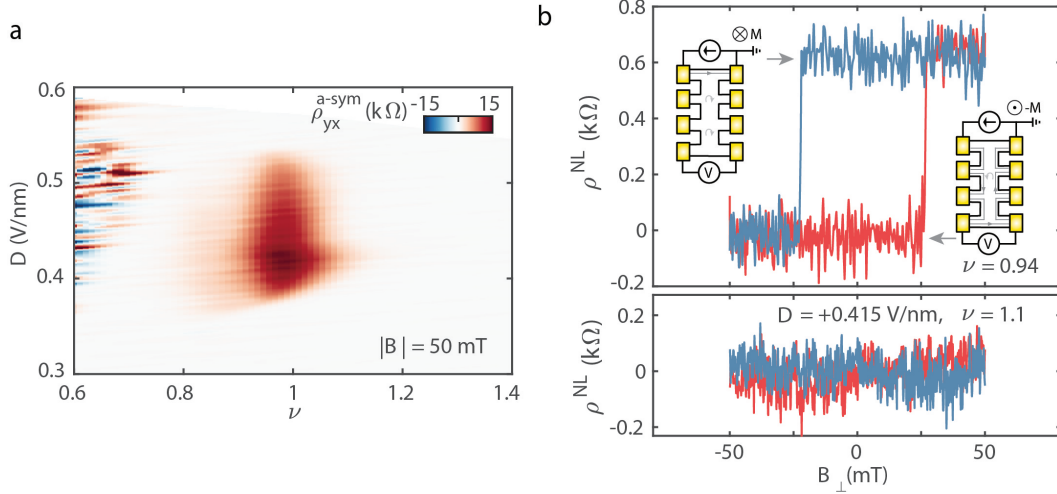


Figure 3.8: Field antisymmetrized ρ_{yx} map and the non-local transport. **a**, ρ_{yx} , antisymmetrized at $|B| = 50$ mT. Rapidly oscillating red and blue points near $\nu \approx 0.6$ are related to the nearby insulating charge neutrality point rather than magnetic order. **b**, Non-local resistance, ρ^{NL} , acquired at $D = +0.415$ V/nm and $\nu = 0.94$ (top) and $\nu = 1.1$ (bottom). Schematics illustrate the contact configuration used for the measurement, as well as the corresponding chiral edge modes equilibrated to the source electrode. Outside the magnetic regime (bottom panel), vanishingly small ρ^{NL} is observed. In contrast, substantial ρ^{NL} is observed for a single sign of the magnetization. This has previously been understood to result from differences in equilibration between the nonlocal voltage probes depending on the chirality of the edge modes [125; 53], indicating the existent of these topological edge modes in our device.

robustly observed within a few mT range near zero field (Fig. 3.9d). In contrast, we do not observe switching upon sweeping D back and forth (Figure 3.9b). The exact mechanism responsible for these unusual switching dynamics — including whether it is of fundamental origin or related to sample inhomogeneity — remains an open question for future work. We should note here that a concurrent work shows tMBG device, at a similar twist angle, with similar weakly-gapped states at $\nu = \pm 4$ but no correlated (nor topological) states [21]. Further experiments at nearby twist angle are needed to identify the hidden parameter of the reported two devices.

3.2.2 Isospin flavor of the orbital magnetism

We have discussed orbital magnetism in a $\theta = 0.89^\circ$ tMBG sample in the previous part. Concurrently, Ref. [21] reports orbital magnetism in $\theta = 1.25^\circ$ tMBG samples with nearly quantized AHE at $\nu = 1, 3$, both with a Chern number $C = 2$ judging from their Hall resistance. In both case of AHE (QAHE) reported in tMBG by then [21; 20], we see that quantization is lost upon doping away from integer filling with

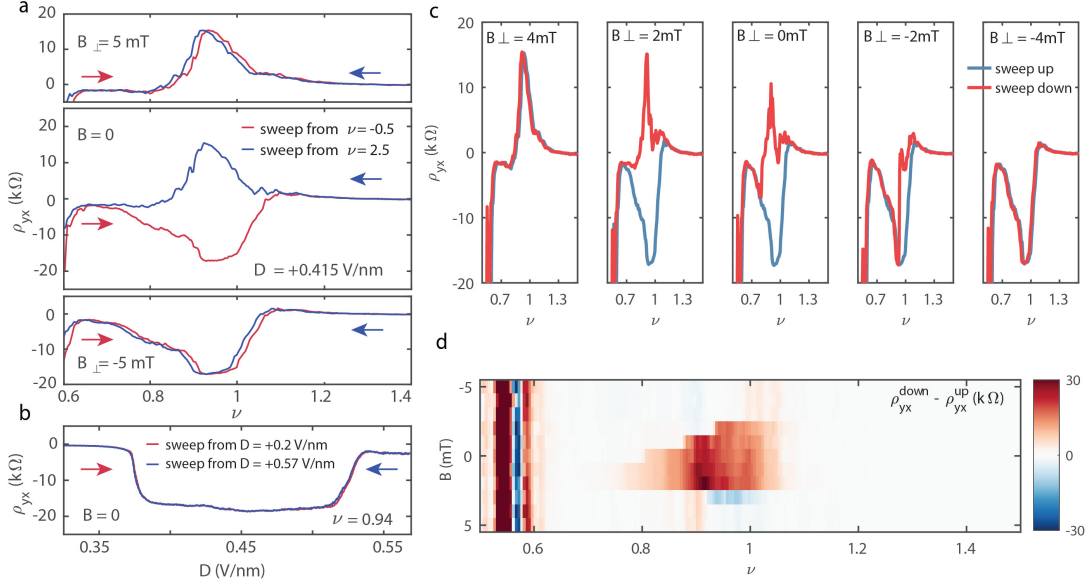


Figure 3.9: Doping-induced switching of the magnetic order in tMBG. **a**, ρ_{yx} as ν is swept back and forth from -0.5 to $+2.5$ at fixed D in the $\theta = 0.89^\circ$ device. The data is acquired at $B_{\perp} = 5$ mT, 0, and -5 mT (top to bottom). $B = 0$ corresponds to zero current in the superconducting magnet coil, with the offset due to trapped flux being less than ~ 1 mT. **b**, ρ_{yx} as D is swept back and forth from $+0.2$ V/nm to $+0.57$ V/nm whereas ν remains fixed at $B = 0$. **c**, ρ_{yx} as ν is swept back and forth from -0.5 to $+2.5$ at $D = +0.415$ V/nm, with $B_{\perp} = +4$ mT, $+2$ mT, 0 mT, -2 mT, -4 mT. Doping-induced switching of the magnetic order is only observed within a small range of $B_{\perp}$ from $+2$ mT to -2 mT. At larger $B_{\perp}$, the magnetic state is aligned with the applied field regardless of the sweep direction. The measurement is performed subsequent to thermally-cycling the device to $T = 4$ K — above the Curie temperature — and then back to base temperature. **d**, The difference between ρ_{yx} sweeping up and down as a function of $B_{\perp}$. The red region denotes the regime in which the magnetization can be switched with doping.

electrons or holes. However a weak AHE can still persist owing to the large Berry curvature at the band edge. Upon applying a magnetic field, the trajectory of the state drifts in the $n - B$ phase space as described by the Štředa formula [126], $C = (h/e)(\partial n / \partial B)$, with slope equal to the Chern number, C , of the state (h is Planck's constant, e is the charge of the electron, and n is the charge carrier density) [21]. In a model of the isolated conduction bands of tMBG as four overlapping flat bands with varying spin and valley order, the two reports of orbital magnetic states can be well described by a simplest ground state with spontaneous polarization of carriers into a single symmetry-broken band with non-zero Chern number — a spin and valley polarized state. We should cautiously note here that we always limit our discussion to a simplest scenarios possible, as more exotic ground states with mixed spin and valley polarization may also be possible.

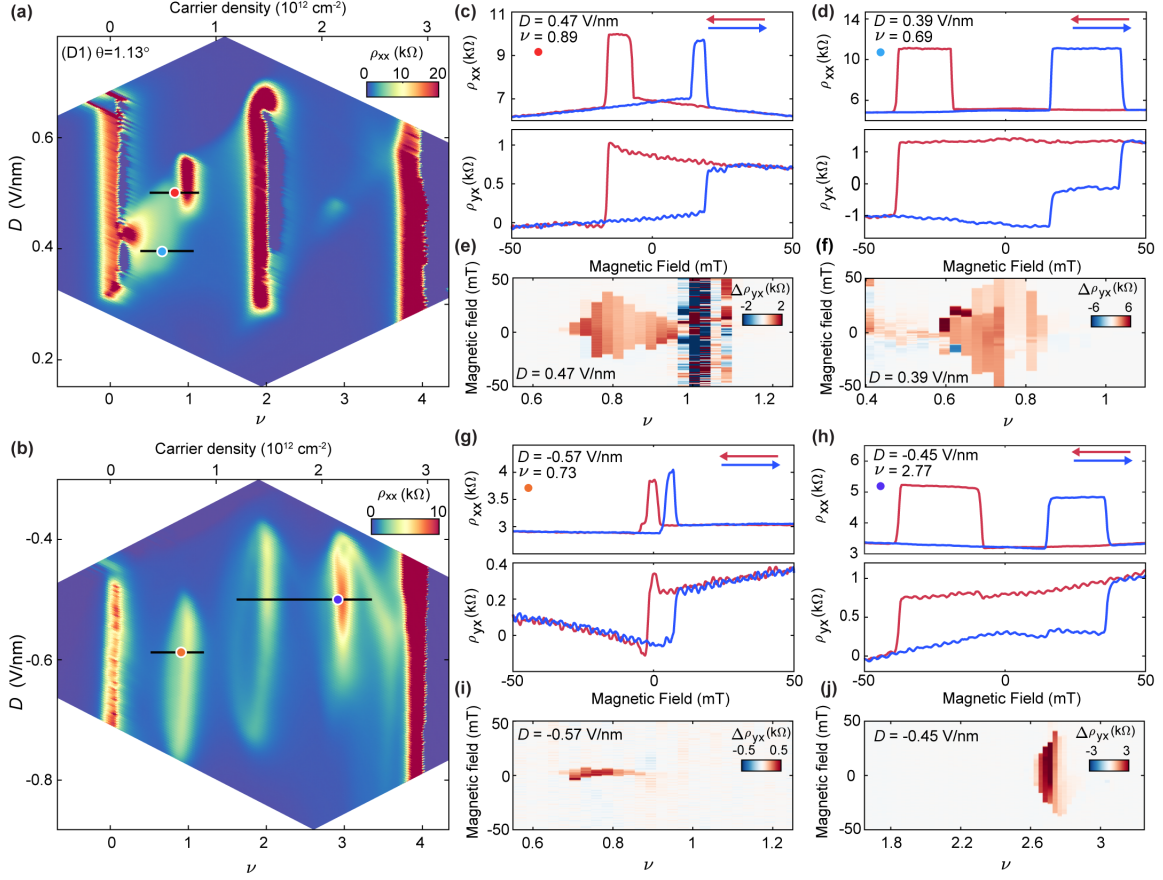


Figure 3.10: AHE in metallic states of tMBG. **a-b**, Longitudinal resistivity, ρ_{xx} , of a $\theta = 1.13^\circ$ sample for $D > 0$ (**a**) and $D < 0$ (**b**). ρ_{xx} is symmetrized at $|B| = 0.5$ T in order to suppress any magnetic hysteresis effects. **c-d**, ρ_{xx} (top) and ρ_{yx} (bottom) acquired as B is swept back and forth at ν and D indicated by the labels, and by the red and blue markers in **a**, respectively. **e-f**, Hysteresis loop height, $\Delta\rho_{yx}$, as a function of doping at $D = 0.47$ V/nm (**e**) and $D = 0.39$ V/nm (**f**), as indicated by the black lines in **a**. Rapidly oscillating red and blue points near $\nu = 1$ in **e** arise due to the correlated trivial insulating state rather than magnetic ordering. **g-h**, ρ_{xx} (top) and ρ_{yx} (bottom) acquired as B is swept back and forth at ν and D indicated by the labels, and by the orange and purple markers in **b**, respectively. **i-j**, $\Delta\rho_{yx}$ as a function of doping at $D = -0.57$ V/nm (**i**) and $D = -0.45$ V/nm (**j**), as indicated by the black lines in **b**. We note that small oscillation features in ρ_{yx} curves are noise, arising due to the low excitation current (1 nA) used in the measurements. ρ_{yx} is not antisymmetrized, and is offset from zero as a result of mixing with ρ_{xx} . All data are acquired at $T = 0.3$ K.

In this section, we discuss orbital magnetic states that cannot be understood as a spin and valley polarized state. We report this new type of orbital magnetic states from measurements of tMBG samples over a tight range of twist angles, $1.13^\circ \leq \theta \leq 1.19^\circ$. We primarily focus our attention on two devices with twist angles of $\theta = 1.13^\circ$ and 1.19° . Figures 3.10a-b show maps of the longitudinal resistivity, ρ_{xx} , in the $\theta = 1.13^\circ$

sample for $D > 0$ and $D < 0$, respectively. The maps are primarily confined to the flat conduction band ($0 \leq \nu \leq 4$) and large $|D|$, for which correlated states are observed at low temperature. We observe robust insulating states at $\nu = 0$ and 4 for both signs of D , as anticipated from calculations of the single-particle band structure of tMBG. We additionally see well-developed correlated insulating states at $\nu = 1$ and 2 for $D > 0$ (Fig. 3.10a), as well as correlated metallic states at $\nu = 1, 2$, and 3 for $D < 0$ (Fig. 3.10b).

This correlated phase diagram connects smoothly to prior measurements of devices with slightly different twist angles. For $D > 0$, devices over a wide range of twist angles manifest a robust correlated insulating state at $\nu = 2$ [20; 21; 22]. However, the states at $\nu = 1$ and 3 are absent in devices with slightly smaller twist angles (although appear to re-emerge in a device with even smaller twist angle, $\theta = 0.89^\circ$, subsequent to an anticipated topological transition in the band from $C_v = 2$ to 1 [127]). In contrast, both of these states are seen in devices with slightly larger twist angles [21; 22], before eventually disappearing again in devices with $\theta \gtrsim 1.4^\circ$ [20; 21; 22]. The correlated states at $D < 0$ are less sensitive to twist angle, with resistive bumps observed at all integer ν in devices with $1.05^\circ \lesssim \theta \lesssim 1.4^\circ$ [20; 21; 22]. The absence of robust correlated insulating states is likely a consequence of the larger bandwidth compared to the $D > 0$ bands [128; 127; 129].

In all three of our devices measured with the twist angle range $1.13^\circ \leq \theta \leq 1.19^\circ$, we observe an anomalous Hall effect (AHE) within the halo-like phase space associated with the symmetry-broken state at $\nu = 1$ for $D > 0$. We see hysteretic behavior in both ρ_{xx} and ρ_{yx} as B is swept back and forth at fixed ν and D (Figs. 3.10c-d). Figures 3.10e-f show maps of the AHE versus ν at two different fixed $D > 0$, acquired by taking the difference of ρ_{yx} between the two field sweeping directions, $\Delta\rho_{yx} = \rho_{yx}^{B\uparrow} - \rho_{yx}^{B\downarrow}$. Precisely at $\nu = 1$ in the map acquired at $D = 0.47$ V/nm (Fig. 3.10e), we measure a large ρ_{xx} of tens of kilohms and a rapidly oscillating $\Delta\rho_{yx}$ consistent with the behavior of a trivial insulating state, rather than a quantum anomalous Hall (QAH) state. This contrasts prior reports in devices with $\theta = 1.25^\circ$, in which a nearly quantized AHE in ρ_{yx} and a small ρ_{xx} are observed precisely at $\nu = 1$ [21]. For $D < 0$, our measurements additionally reveal large pockets of AHE near $\nu = 1$ and 3 (Figs. 3.10g-j), which have previously not been reported.

Our results in devices with slightly smaller twist angles exhibit neither signatures of a QAH state precisely at odd integer ν , nor the anticipated finite sloping of these states in a weak magnetic field. Figure 3.11a

shows the low-field Landau fan diagram at $D > 0$ in the $\theta = 1.13^\circ$ sample, along with a cut of ρ_{xx} versus B taken precisely at $\nu = 1$ in the panel to the right. We find that the insulating state at $\nu = 1$ projects vertically and is suppressed with small B , before eventually re-emerging at larger B . Although in principle a disordered network of spin and valley polarized states with nearly equal mixture of valley K and K' domains may localize to form a trivial insulator, the application of a weak B should rapidly align the domains and form a Chern insulator state. Our observations are inconsistent with this scenario, suggesting that the $B = 0$ ground state is not a spin and valley polarized state.

Figure 3.11b shows a similar measurement for $D < 0$ in the $\theta = 1.19^\circ$ sample. This device is notable in that it exhibits the only known instance of insulating temperature dependence in a $D < 0$ correlated state, in this case arising at $\nu = 1$. Similar to the case of the $\nu = 1$ state in the $\theta = 1.13^\circ$ sample at $D > 0$, we find that ρ_{xx} projects vertically, first becoming less resistive with B before eventually growing at high field. Both of these insulating states at $B = 0$ are accompanied by an AHE in a small region of $\nu < 1$, approximately corresponding to the regions of sharply enhanced resistivity in Figs. 3.11a-b. This is consistent with our understanding that the AHE is associated with the symmetry-broken state at $\nu = 1$, which persists well away from integer filling. Similar to the case of the $\nu = 1$ state for $D > 0$, we find that the AHE is strongest for $\nu \sim 0.9$ but vanishes at $\nu = 1$, where a trivial insulating state emerges instead. Similarly, this state is not consistent with a spin and valley polarized ground state.

A number of correlated ground states have previously been proposed for twisted double bilayer graphene (tDBG) [89], and are candidate ground states for tMBG as well. Among those that result in an insulating state at odd integer ν , none naturally explain our observation of an AHE upon doping, as this requires a highly unusual combination of broken time reversal symmetry (TRS), finite Berry curvature, but a net $C = 0$. In order to gain more insight, we start with a fully spin- and valley-polarized ground state at $\nu = 1$ and determine its stability by calculating the dispersion of the collective spin and valley wave excitation (see Appendix C.1 for full details). Negative energy of this excitation is a signature of the instability of the spin-valley polarized state. Figure 3.12a shows the calculated valley magnon energy for various values of the dielectric constant, ϵ , which acts as an effective tuning parameter for the ratio U/W , where U is the Coulomb interaction strength. We define a valley magnon as a spin-singlet valley-flip exciton (i.e. an exciton comprising two particles from opposite valleys). Condensation into an IVC order is favored when

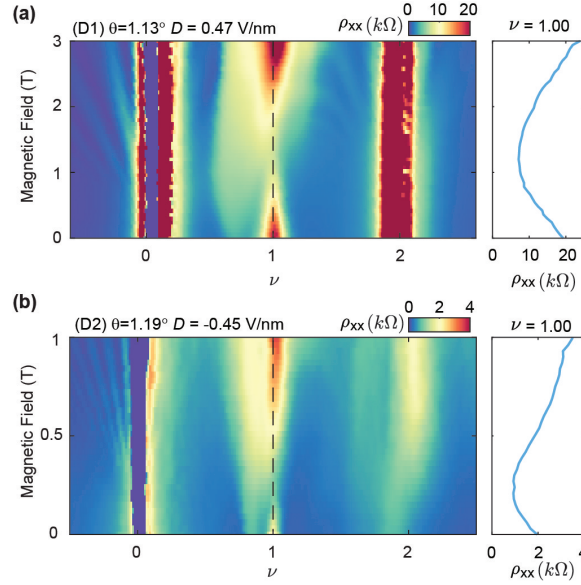


Figure 3.11: Magnetic field dependence of the $\nu = 1$ state. **a-b**, ρ_{xx} as a function of doping and B at $T = 0.1$ K for $D > 0$ in the $\theta = 1.13^\circ$ sample **(a)** and at $T = 0.05$ K for $D < 0$ in the $\theta = 1.19^\circ$ sample **(b)**. Cuts of $\rho_{xx}(B)$ are shown at $\nu = 1$ in the panels to the right of each map, at positions indicated by the black dashed lines in the main panels. Note that ν is the fast sweeping axis in these measurements, and as a consequence the measured ρ_{xx} is substantially smaller than its true value for the insulating states.

the valley magnon energy becomes negative at any point within the first Brillouin zone. We find that the valley polarized state is stable at small ϵ (i.e. large U/W), however there is a first-order phase transition into an IVC order above a modest critical ϵ (i.e. intermediate U/W). Figure 3.12b shows schematic representations of the VP and IVC states on the Bloch sphere.

Generically, the IVC state will retain TRS since it is a superposition of states at valleys K and K', trivializing the overall Chern number. However, our analysis of the IVC order parameter reveals that it prefers to carry finite angular momentum owing to the non-zero C_v of the constituent bands, breaking TRS (Fig. 3.12a inset). The IVC order additionally prefers to carry a non-zero crystal momentum $\mathbf{Q}$, because the valley magnon dispersion has minimum at $\mathbf{Q} \neq 0$ (Fig. 3.12a). We therefore denote this as a “Q-IVC” state, in order to distinguish from previously considered IVC orders with $\mathbf{Q} = 0$. In combination with the large Berry curvature anticipated at the band edge, this Q-IVC order is expected to result in a trivial insulating state at integer ν when gapped, but an AHE upon weakly doping the band into a metallic state. Although a full theoretical analysis is yet to be developed, we find the Q-IVC state to be a plausible ground state order that is consistent with all of our observations near $\nu = 1$ for both signs of D . Furthermore, the potential

Q-IVC state at odd integer ν appears to cede to a spin and valley polarized state for $D > 0$ in devices with slightly larger twist angles [21], whereas flavor-unpolarized states are observed in devices with slightly smaller twist angles [20]. This suggests that the ground state order may be controlled sensitively by U/W , in which sequential transitions from VP to Q-IVC to flavor-unpolarized states are driven by an increasing W with reducing θ .

Alternatively, a more subtle origin of the behavior of the states at $\nu = 1$ is that the ground state order changes upon doping. In this scenario, the ground state at and very near $\nu = 1$ is an IVC that preserves TRS. Such an IVC state also trivializes the Chern number, resulting in a trivial insulating state when gapped. A phase transition to a fully flavor-polarized state arises upon doping away from $\nu = 1$, leading to an AHE in the metallic states owing to the inherent TRS breaking and finite C . Theoretically, this phase transition is expected to be first order. Since we do not observe any obvious signatures of hysteresis with doping, our results are naively inconsistent with this scenario. However, it remains possible that this phase transition is smeared by various forms of disorder in the sample, including twist angle disorder. For this reason, we are not able to unambiguously distinguish between this scenario and the case of the TRS-breaking Q-IVC order detailed above. Although we believe it to be unlikely to achieve a first-order phase transition between a IVC and VP state with a small amount of doping, a more detailed theoretical analysis is necessary to assess its feasibility. Another possibility is that interaction-induced mixing with remote bands renormalizes the valley Chern number to 0. In this case, remnant Berry curvate at the band edge could give rise to an AHE neighboring the trivial correlated insulator state, even in a spin and valley polarized state. However, we believe this to be highly unlikely owing both to the emergence of correlated Chern insulating states at small B (discussed in the next section), and due to the absence of such a renormalization in our band structure calculations. Finally, we note that we have not performed an exhaustive search for all possible ground states at $\nu = 1$. Although there may be other states that are consistent with trivial insulating behavior at integer ν and an AHE upon doping, they are likely to be more exotic than the various IVC orders considered here.

Determining the ground state order for the correlated metallic states at $D < 0$ is more challenging, since transport measurements cannot directly probe the topology of the state in absence of a gap. Since we do not observe any insulating behavior for the $D < 0$ correlated states in device D1, the AHE we observe at $\nu = 1$ and 3 are consistent with either an ungapped valley polarized state or an ungapped IVC state with

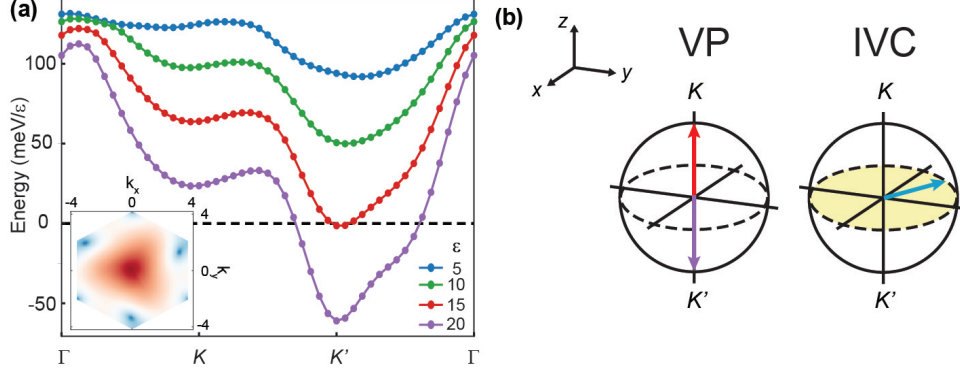


Figure 3.12: The ground state ordering of the $\nu = 1$ state. **a**, Calculated energy of valley magnon formation in tMBG with $\theta = 1.16^\circ$ at selected values of ϵ . The inset shows the profile of the intervalley exciton, $F(\mathbf{k})$, as a function of crystal momentum in the first Brillouin zone, with the magnitude represented by a log color scale. $F(k) \neq F(-k)$, indicating that the Q-IVC state breaks TRS. $k_{x,y}$ have units $1/a_M$, where a_M is the moiré lattice constant. **B**, Bloch sphere representation of the VP and IVC states. VP states point towards the north (south) pole for K (K') polarization, as indicated by the red (purple) arrows. IVC states (blue arrow) point along any direction in the x - y plane (shaded).

finite angular momentum. The combination of larger W and a smaller predicted $C_v = 1$ naively favors IVC ordering even more strongly for the $D < 0$ band compared with the $D > 0$ band, which is calculated to have $C_v = 2$. However, future work will be necessary to unambiguously identify these ground state orders.

3.3 Hofstadter physics and Chern insulators in tMBG

3.3.1 Closely competing Chern insulators at $D > 0$

The application of a large magnetic field serves to further modify the competition between different correlated ground states, both by tuning the orbital and spin Zeeman energies, and by transforming the low energy bands into a recursive series of Hofstadter minibands. In this section, we discuss the Hofstadter physics in tMBG.

Figure 3.13 shows Landau fan diagrams in the $\theta = 1.13^\circ$ sample for both ρ_{xx} and ρ_{xy} at different values of $D > 0$ (leftmost and middle columns, respectively). The schematics (rightmost column) denote the well-developed gapped states observed in each map. All are anticipated within a Hofstadter butterfly picture, in which gapped states follow trajectories described by the Diophantine equation, $\nu = tn_\phi + s$, where $t, s \in \mathbb{Z}$, and $n_\phi = \Phi/\Phi_0$ is the normalized magnetic flux. We refer to gapped states using the notation

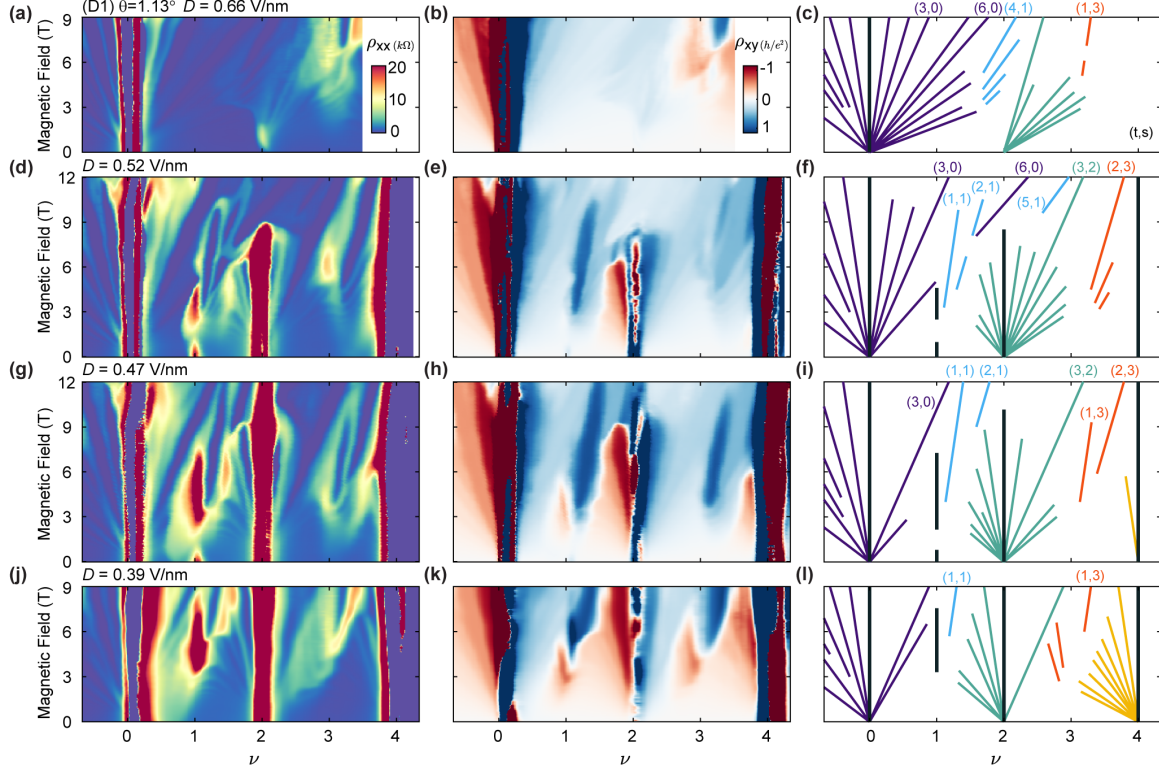


Figure 3.13: Landau fan diagrams and spontaneous flavor polarization at high field for $D > 0$. Landau fan diagrams in the $\theta = 1.13^\circ$ sample at **a-c**, $D = 0.66$ V/nm, **d-f**, $D = 0.52$ V/nm, **g-i**, $D = 0.47$ V/nm, and **j-l**, $D = 0.39$ V/nm, all acquired below $T = 0.3$ K. The leftmost column shows ρ_{xx} , the central column shows ρ_{xy} , and the rightmost column schematically denotes the strongest observed gapped states. In the schematic, purple lines correspond to states tracing to $\nu = 0$, blue to $\nu = 1$, green to $\nu = 2$, orange to $\nu = 3$, and yellow to $\nu = 4$. The vertical black lines denote topologically trivial insulating states. Selected states are labeled by their respective (t, s) indices.

(t, s) , where t corresponds to the Chern number of the state and s corresponds to the number of electrons bound to each moiré unit cell. By convention, $(|t| > 0, s = 0)$ states are referred to as “integer quantum Hall states,” whereas $(|t| > 0, s > 0)$ are “Chern insulators” [83]. States with different s are distinguished by color in the schematics, whereas trivial insulating states ($t = 0$) are depicted in black irrespective of their corresponding s .

We observe numerous similarities between the gapped states at filling factors $0 \leq \nu < 2$ and $2 \leq \nu \leq 4$. Although far from exact, there is an approximate mapping of our observed states between ν and $\nu + 2$, especially for maps in which we observe a robust correlated insulating state at $\nu = 2$ (i.e. Figs. 3.13d-l). The state at $\nu = 2$ is thought to be spin-polarized at $B = 0$ [20], suggesting that interactions split the four-fold degenerate conduction band into two sets of spin-polarized but valley-unpolarized bands over a wide

range of $D > 0$. The remaining valley degeneracy can also be spontaneously broken, however this often requires the assistance of a finite B depending on ν and D for our studied range of twist angles. Symmetry-broken states only persist to $B = 0$ around $\nu = 1$ for a small range of D , and we do not observe $B = 0$ symmetry breaking at $\nu = 3$ within our studied range of θ . However, we find that numerous Chern insulator states emerge spontaneously at finite B over a wide range of D with both $s = 1$ and 3 . These states are expected to be especially strong given the finite $C_v = 2$ of the band, consistent with our observations and very likely indicative of a full flavor polarization of the $\nu = 1$ and 3 states at high field. In this context, the reentrant insulating behavior observed over a small range of D at $\nu = 1$ (Figs. 3.13d,g), along with the strong $(t > 0, 1)$ Chern insulators at high field, is consistent with a phase transition from a Q-IVC state at low field to a valley polarized state at high field.

More generally, we find that the states we observe in Fig. 3.13 do not follow a simple progression with ν and B , reflecting the rich competition between the gapped single-particle Hofstadter subbands and the tendency towards spontaneous flavor polarization into a subset of these bands. This is similar to observations in tBLG [57; 58; 59; 60; 61; 62], however here we observe a more complicated sequence of symmetry breaking in which various flavor-polarized and unpolarized ground states closely compete. For example, in Figs. 3.13d-f, the main-sequence quantum Hall state emanating from the charge neutrality point, $(6, 0)$, clearly intercedes both the $(2, 1)$ and $(0, 2)$ states and closes those gaps at high field. In particular, the $(2, 1)$ gap closes and then reopens at higher field following this interruption. Additionally, a $(5, 1)$ state is observed at filling factors $\nu > 2$ for $B \gtrsim 9$ T. Tuning D further tips the balance in the competition between these states, highlighting their near degeneracy.

3.3.2 First order phase transitions at $D < 0$

For $D < 0$, we observe different manifestations of field-driven competitions between correlated ground states. The correlated metallic states observed for $D < 0$ have little to no dependence on magnetic field applied in-plane, $B_{||}$ (shown up to 9 T in Fig. 3.14a). However, applying a perpendicular field, $B_{\perp}$, causes a sharp transition to an insulating behavior at $\nu = 2$ and 3 above a critical field value (Fig. 3.14b). Signs of an incipient resistive state can also be seen over a finite range of $B_{\perp}$ at $\nu = 1$. Quantum oscillations associated with the states at $\nu = 2$ and 3 emerge only above a finite $B_{\perp}$, with apparent two-fold degeneracy

neighboring $\nu = 2$ and no remaining degeneracies neighboring $\nu = 3$ (Fig. 3.14b, lower panel). This too is consistent with prior studies of tBLG [46; 87; 121], in which recent measurements have indicated that a Dirac-like dispersion is revived at each quarter band filling owing to a cascade of spontaneously broken symmetries [45; 64].

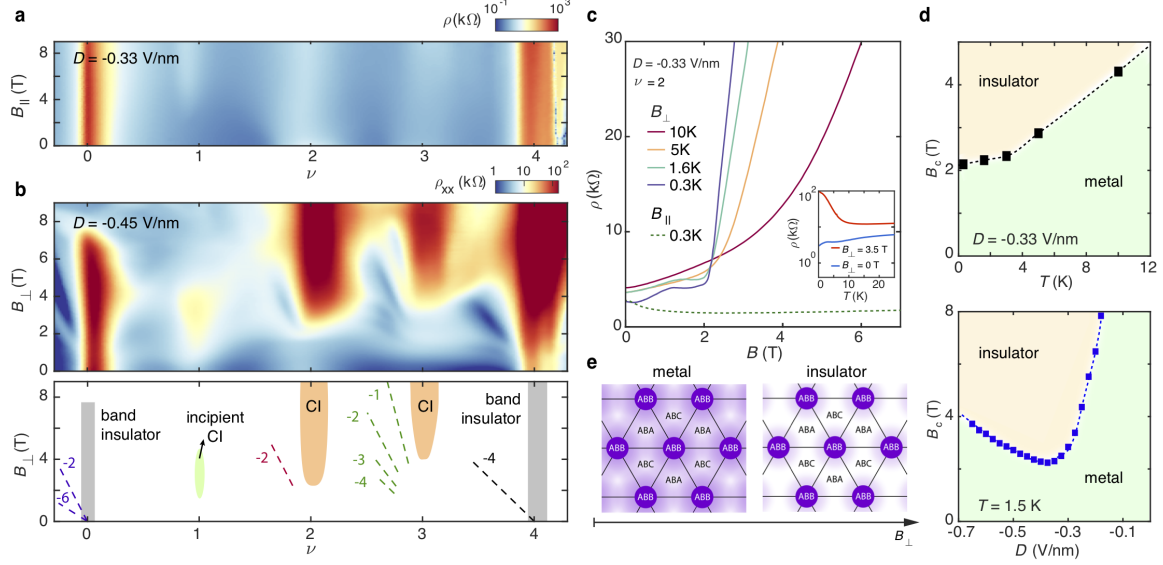


Figure 3.14: Magneto transport for $D < 0$ in the $\theta = 1.08^\circ$ tMBG. **a**, Device resistivity up to full filling of the moiré unit cell with $D = -0.33$ V/nm as a function of $B_{||}$ at $T = 300$ mK. **b**, Landau fan diagram with $D = -0.45$ V/nm. The bottom panel schematically denotes the observed quantum oscillations, as well as band insulator states (gray), CI states (orange), and an incipient CI state (green). CI states at $\nu = 2, 3$ and associated quantum oscillations only emerge for finite $B_{\perp}$. **c** $\rho(B_{\perp})$ at various T (solid curves) and $\rho(B_{||})$ at $T = 300$ mK. (Inset) $\rho(T)$ at $\nu = 2$ for $B_{\perp} = 0$ and 3.5 T. **d**, Critical magnetic field for the onset of insulating behavior, B_c , versus T (top) and D (bottom). Dashed lines are guides to the eye. Regions colored green correspond to metallic temperature dependence, whereas beige regions correspond to insulating behavior. **e**, Cartoon schematics of the inferred LDOS (light pink) on the moiré lattice (ABB sites denoted by dark purple dots) as a function of $B_{\perp}$.

Figure 3.14c shows $\rho(B_{\perp})$ at $\nu = 2$ in the $\theta = 1.08^\circ$ sample for various temperatures (solid curves), as well as $\rho(B_{||})$ at $T = 0.3$ K (dashed curve). While ρ is nearly insensitive to $B_{||}$, we observe an abrupt transition to insulating behavior above a critical perpendicular magnetic field, B_c , at each temperature, defined as the crossover point between separate linear fits to $\rho(B_{||})$ at low and high $B_{\perp}$. The inset of Figure 3.14c compares $\rho(T)$ at $B_{\perp} = 0$ and 3.5 T for $\nu = 2$, showing the metallic and insulating behavior, respectively (see Extended Data Fig. 7 for $\nu = 1, 3$). We find that B_c first grows weakly with increasing temperature, then increases rapidly at higher temperature (Fig. 3.14d). B_c can also be tuned with D , as

shown for $T = 1.5$ K in (Fig. 3.14d). It is peculiar that insulating behavior onsets with $B_{\perp}$ but has almost no dependence on $B_{\parallel}$. In the simple approximation of a perfectly 2D sample, $B_{\perp}$ couples to both the spin and orbital Zeeman energies whereas $B_{\parallel}$ only couples to the spin Zeeman energy. Consequentially, spin-polarized (unpolarized) states are typically strengthened (suppressed) by a magnetic field pointing in either orientation. The correlated states for $D < 0$ do not appear to have a natural interpretation in this context.

Insulating states emerging with $B_{\perp}$ have been previously observed in bulk correlated systems including Bechgaard salts [130] and high-mobility semiconductors such as $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$ [131; 132]. These insulating states were thought to be driven by orbital confinement effects, associated with spin-density waves in the former and Wigner crystal formation in the latter. The overall similarity of our $B_c(T)$ to those results suggests that related orbital confinement effects may be at play in tMBG. Phenomenologically similar behavior has also been observed in the $\nu = 0$ quantum Hall states of monolayer and bilayer graphene. The ground state is thought to be a spin antiferromagnet in the absence of explicit sublattice symmetry breaking [133], and originates from spontaneous valley anisotropy driven by orbital effects. Although these states should be stabilized at any finite $B_{\perp}$, disorder can result in an artificial high-field transition between a metallic and insulating state at $\nu = 0$ in graphene [134]. However, in disordered graphene the low-field resistance is independent of temperature, in sharp contrast to our observations of metallic temperature dependence. Therefore, it is more likely that the correlated states we observe here require a finite $B_{\perp}$ in order to undergo an orbitally-driven transition from a metallic to an insulating ground state. Future experiments in a tilted magnetic field should help to further elucidate the spin and valley ordering of these states.

The cartoon schematic in Fig. 3.14e qualitatively illustrates the presumed atomic-scale electronic structure of tMBG in the metallic and field-assisted insulating phases. The dark purple sites correspond to regions of ABB stacking, and the lighter pink halo represents the associated LDOS. $B_{\perp}$ is expected to orbitally confine the LDOS more tightly around the ABB sites, resulting in an enhancement in the effective interaction strength. In contrast, the LDOS does not depend strongly on $B_{\parallel}$ as orbital effects are anticipated to be weak owing to the atomically-thin nature of tMBG, and therefore the interaction strength is also not substantially modified. Although our results at $D < 0$ appear to share a number of qualitative similarities with weakly correlated tBLG, one important difference is that superconductivity appears to be absent in our device, whereas it remains a robust ground state even in screened tBLG devices [121; 87; 17]. Differences in

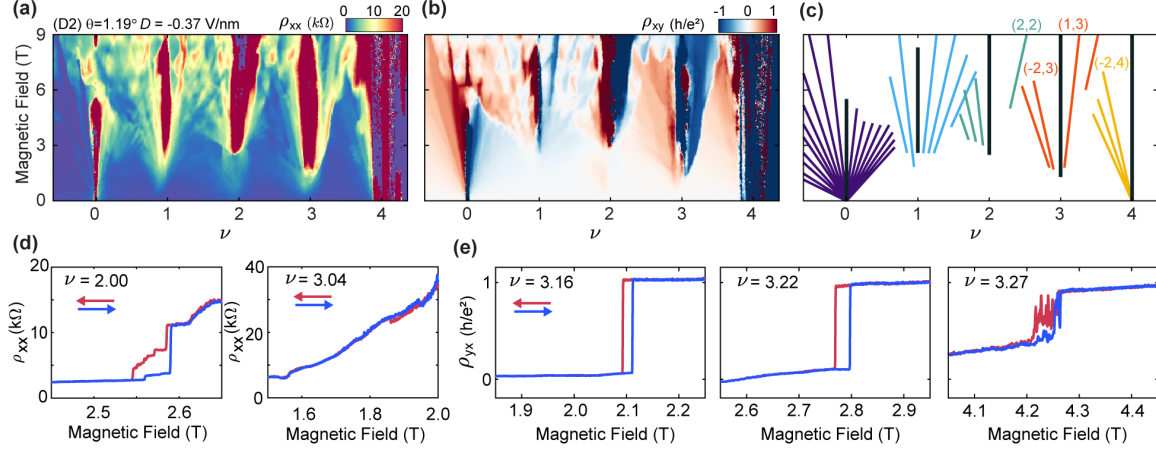


Figure 3.15: First-order orbital phase transitions at high field for $D < 0$. **a-b**, Landau fan diagram of ρ_{xx} (**a**) and ρ_{xy} (**b**) for the $\theta = 1.19^\circ$ sample at $D = -0.37$ V/nm, acquired at $T = 0.1$ K. **c**, Schematic representation of the observed states, following the convention established in Fig. 3.13. A subset of the most robust gapped states we observe are labeled by their respective (t, s) indices. States at all integer t are observed for $s = 0$ and 1 , indicating full degeneracy lifting. **d**, ρ_{xx} acquired as B is swept back and forth at $\nu = 2.00$ (left) and $\nu = 3.04$ (right). **e**, ρ_{yx} acquired as B is swept back and forth at $\nu = 3.16$ (left), $\nu = 3.22$ (middle), and $\nu = 3.27$ (right). Data in **d-e** are acquired at $T = 0.1$ K.

bandwidth may be a contributing factor, however it is also possible that the low symmetry of tMBG plays a role.

Figures 3.15a-b show Landau fan diagrams for ρ_{xx} and ρ_{xy} at $D = -0.37$ V/nm in another sample at $\theta = 1.19^\circ$, with an associated schematic of the observed gapped states shown in Fig. 3.15c. Consistent with prior discussion, we see the emergence of correlated insulating states at all integer ν at finite B . However, the improved quality of this device reveals a number of previously obscured features of these states. We observe abrupt transitions from metallic to insulating states at $\nu = 2$ and 3 above a critical magnetic field, B_c , as shown in Fig. 3.15d. We additionally observe hysteresis at high field upon sweeping B back and forth across this phase transition at $\nu = 2$ (as well as very weak hysteresis signatures at $\nu = 3.04$), indicative of a first-order phase transition. The hysteresis additionally extends to band fillings well away from integer ν . For example, Fig. 3.15e shows ρ_{yx} at various $\nu > 3$, in which we see a hysteretic transition to a state that is nearly quantized to h/e^2 above B_c . Additional measurements in the $\theta = 1.13^\circ$ sample show a $(1, 3)$ Chern insulator that persists nearly to $B = 0$, suggesting that the high-field state is likely fully flavor polarized owing to the anticipated $C_v = 1$ of the band (Figure 3.16). The hysteresis we observe in Fig. 3.15e persists to values of ν for which the gapped states at $B < B_c$ carry $s = 4$ (see Fig. 3.15c), indicating a first-

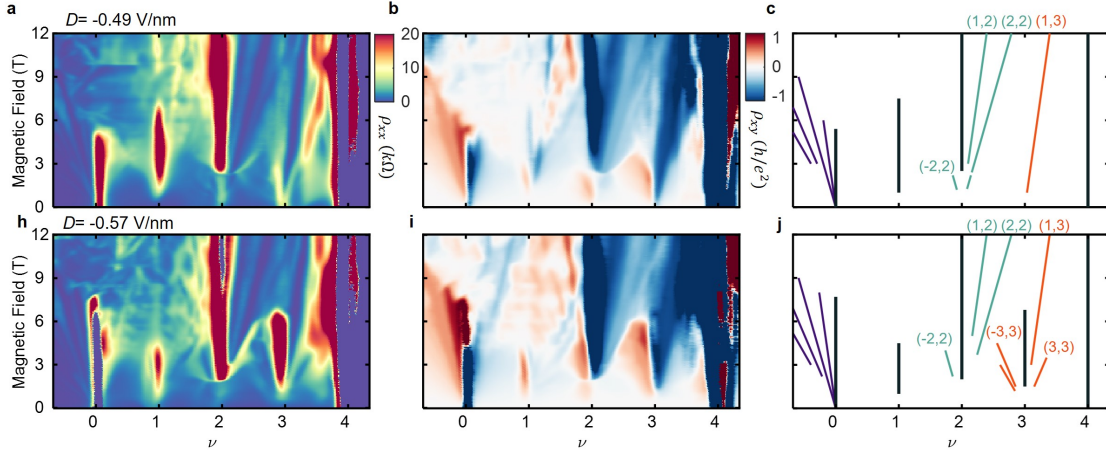


Figure 3.16: Landau fan diagrams for $D < 0$ in the $\theta = 1.13^\circ$ sample. **a-c**, Landau fan diagrams at $D = -0.49$ V/nm (**e-g**) and $D = -0.57$ V/nm (**h-j**), acquired at $T = 300$ mK. The leftmost column shows ρ_{xx} , the central column shows ρ_{xy} , and the rightmost column schematically denotes the observed states. In **e-g**, we observe a very strong $(1, 3)$ state that emerges and becomes quantized at very small B . This likely reflects the anticipated $C_v = 1$ of the band at $D < 0$, however we are unable to determine this unambiguously in the absence of a QAH state at $B = 0$. We additionally observe a clear Landau fan corresponding to the correlated state at $\nu = 2$ at very low fields in (**e-g**). These gapped states close at the phase transition ($B \sim 2.8$ T) and immediately reopen at higher field along with a robust trivial insulating state, $(0, 2)$. Although this implies a first-order phase transition between two different correlated states, we are not able to unambiguously identify the ground state order of either from our measurements.

order topological phase transition between the flavor-unpolarized Fermi surface near full band filling and the (presumably) fully flavor-polarized Fermi surface associated with $\nu = 3$.

Overall, these results reveal the richness of the correlated phase diagram of tMBG, which can be tuned sensitively with the combination of ν , D , B , and θ . Magnetic field further tunes the close competition between flavor-unpolarized ground states and numerous flavor-polarized states, resulting in abrupt and occasionally hysteretic topological phase transitions at high field.

Chapter 4

Optical sensing of twisted graphene superlattices

We have explored the strongly correlated states and topological states in tDBG and tMBG in the previous two chapters. We mainly employ electrical transport measurements in these studies to probe the macroscopically averaged resistivity (or Hall resistivity). Twist angle disorder remains one of the main obstacles in moiré quantum materials. Therefore many fragile states (like fractional Chern insulators [63]) tend to smear out in such macroscopic measurements.

In this chapter, we propose and show preliminary experiments on a new optical probe of twisted graphene by using the Rydberg excitons in monolayer transition metal dichalcogenides MX_2 . We first introduce the fundamentals of the monolayer semiconductor MX_2 , which itself composes an important part of 2D materials field. We then investigate the valley phonons in monolayer WSe_2 in depth, which leads to a number of exciton complexes in its photoluminescence spectrum. With these background knowledge, we focus our attention on the Rydberg exciton states. Their large Bohr radius makes them ideal for sensing dielectric environment of adjacent layers. We investigate this local probe in the heterostructure of WSe_2/tBLG .

4.1 Fundamentals of monolayer transition metal dichalcogenides

In this section, we briefly overview the electronic band structure and the important physics of the group-6 monolayer transition metal dichalcogenides (MX_2). We are mainly interested in the semiconducting TMD monolayers in the 2H phase, including WSe_2 , WS_2 , MoSe_2 , MoS_2 , and MoTe_2 . The monolayer MX_2 is composed of three atomic sublayers, with X atoms in the top/bottom layers and M atoms in the middle layer each forming a triangular lattice (Figure 4.1). The two sets of triangular lattice together form a hexagonal lattice like the A/B sublattice in graphene. Despite their similarities, the monolayer TMD does not have inversion symmetry, breaking the A/B sublattice symmetry. This adds an A/B site-dependent on-site energy term in its tight-binding Hamiltonian, which opens a band gap at the two Brillouin zone corners K and K' :

$$H(\mathbf{q}) = at(\tau \mathbf{q}_x \sigma_x + \mathbf{q}_y \sigma_y) + \frac{\Delta}{2} \sigma_z + \dots \quad (4.1)$$

Here, a is the lattice constant of MX_2 , t is the hopping integral of adjacent atoms, $\tau = \pm 1$ is the valley index, σ_x , σ_y , and σ_z are the Pauli matrix for valley, and finally Δ is the band gap at the K and K' .

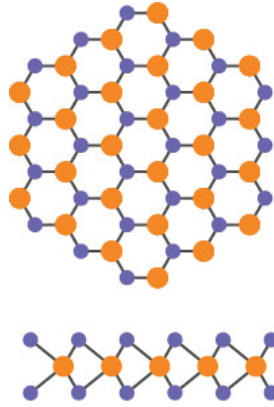


Figure 4.1: Schematic of monolayer MX_2 in 2H phase. Top and side view of the monolayer MX_2 lattice structure. The bigger yellow circles represent transition metal atoms while the smaller purple circles represent chalcogen atoms.

The bulk forms of TMDs have been intensively investigated back in the 1960s, yet the observation of an indirect-to-direct band gap transition when thinning them down to a monolayer crystal revives the field in 2010 [135]. The direct band gap at K and K' allows optical accessibility to study the above simple but intriguing Hamiltonian. At the new band edges in monolayers, the conduction bands are mainly composed

of the d_{z^2} orbitals of the M atoms, and the valence bands are mainly composed of the $d_{x^2-y^2} + d_{xy}$ orbitals of the same M atoms [136; 137]. Therefore we have to consider the large spin-orbit coupling (SOC) of the M atoms in the low energy effective Hamiltonian, especially in the valence band edges:

$$H(\mathbf{q}) = at(\tau \mathbf{q}_x \sigma_x + \mathbf{q}_y \sigma_y) + \frac{\Delta}{2} \sigma_z + \lambda \tau \frac{\sigma_z - 1}{2} s_z \quad (4.2)$$

where 2λ is the valence band edges spin splitting induced by SOC, and s_z is the Pauli matrix for spin. The spin splitting in the valence bands is around 200 to 400 meV depending on the exact chemical compounds. As a result, the charges in the top most valence band edges have opposite spin at the K and K' valley, exhibiting a unique spin-valley locking effect. Similar SOC induced spin-valley locking effect holds for the conduction bands, but with a much smaller spin splitting around 20 to 30 meV. The important valley physics in monolayer TMD can be readily derives from this simple Hamiltonian:

1. The large Berry curvature at the band edges contributes to a significant anomalous Hall conductivity [138]. Most importantly, due to the opposite Berry curvatures in the K and K' valley, a charge-neutral spin-valley current will be generated perpendicular to the applied electric field, and valley polarized charges can be detected on the sample edges [136]. The unique valley Hall effect has been observed in experiments [139].
2. The optical interband transitions at the two valleys are uniquely coupled to σ^+ and σ^- circularly polarized light [140; 136; 141; 142; 143; 144]. Moreover, a linear polarized light can generate exciton residing in the two valleys coherently [145].
3. The orbital magnetic moment $m(\mathbf{k})$, generated by the Berry curvature at the band edges, contributes significantly to the exciton's Zeeman energy in additional to the atomic orbital contribution and spin contribution [146; 147; 148; 149].

In the monolayer TMD semiconductors, an photon-excited electron in the conduction band and the hole left behind in the valence band are strongly coupled together to form a new quasiparticle — exciton. Thanks to the reduced Coulomb screening in two dimension, the electron and hole are tightly bound together with its binding energy reaching 0.5 eV [150; 151; 152], at least one order larger than the traditional quantum wells systems, despite the Wannier-Mott nature of the exciton. The strong Coulomb interaction enables numerous

type of excitonic species:

1. A spin conserving interband transition from the spin splitted top and bottom valence band edges form two bright exciton species, which we name A and B excitons respectively [150; 151]. Interestingly in Tungsten based MX_2 , the bright A exciton itself is not the ground state but a spin-forbidden dark exciton about 40 meV below it [153; 154; 155; 156; 157; 158]. The bright A exciton can scatter with a Γ_5 zone-center phonon and relax into the dark exciton state. The non-radioactive dark exciton has a huge population at low temperature and further allows formation of other excitonic species.
2. In slightly electron and hole doped MX_2 , the bright A exciton (also the dark exciton in WX_2) can additionally bounds with a free charge in the Fermi sea. The three-particle quasiparticle is known as a trion state and behaves as a fermionic particle.
3. The strong Coulomb interaction further allows coupling between exciton and exciton, or exciton and trion, to form four-particle or five-particle bound states [159; 160; 161; 162; 163].
4. In general, the free exciton can potentially be trapped by a deep localized potential and form a localized exciton. This is considered to be ubiquitous in the early generation of TMD monolayer samples due to low crystal quality and contamination from sample fabrication. With improved crystal quality [164] and sample fabrication technique [85], intrinsic localized excitons that bound to charged point defects are observed [165]. These circularly polarized, long-living excitonic states are assigned to be donor-bound dark trion and its phonon replicas.
5. The tightly bound electron-hole pair can be think as an analogue to a Hydrogen atom, only except that now the negatively charged particle has a similar effective mass as the positively charged particle in the exciton. Therefore, the relative motion of the electron-hole pair gives a very similar energy spectrum as the Hydrogen atom, governed by a main quantum number n . These exciton excited states are known as Rydberg exciton states. They are particularly strong in Frankel exciton, for example the $n = 25$ Rydberg exciton observed in Cu_2O [166]. Rydberg states have also been observed in monolayer TMDs [150], particularly with an enhanced contrast at high magnetic field [167; 168]. The energy spectrum of Rydberg states in two dimension deviates from a simple $1/n^2$ relation as a result of non-uniform dielectric environment, which can be more precisely captured by a Rytova-Kelysh potential [169; 170].

The valley contrasting optical properties and the large family of excitonic species are both of fundamental science interests and numerous application interests. Moreover, the tightly bound excitons have strong interactions with other type of quasiparticles, including polarons in a heavily doped Fermi sea and phonons of various type. For example, the nature of the interacting exciton-polaron at heavily doped regime remains an open question.

In the next part, I will present a work exploring the interaction between exciton and valley phonons — phonons with momentum vectors pointing to the corners of the hexagonal Brillouin zone — and the resulting exciton complexes in the monolayer semiconductor WSe₂.

4.2 Valley phonons and exciton complexes in a monolayer semiconductor

Electron-phonon interaction is a ubiquitous process in solids. The emergent valley-contrasting properties not only impact quasiparticles, but also are expected to give rise to new physics involving zone edge phonons, or valley phonons, which are collective lattice oscillations at the corners of hexagonal Brillouin zone ($\pm K$ points). These phonons have been predicted to play an important role in spin and valley pseudospin relaxation through phonon-assisted intervalley scattering[171; 172]. Additionally, valley phonons can possess chirality with intrinsic pseudo-angular momentum[173], which has recently attracted wide attention[174; 175; 176; 177]. Such chiral phonons have nontrivial Berry curvature and are predicted to give rise to valley phonon Hall effect, a counterpart of valley Hall effect of electrons in 2D semiconductors[178; 139; 179]. Despite the importance of valley phonons, experimental progress in understanding their properties has been limited, since it is challenging to probe phonons with large momentum vectors.

Here, we identify the signatures of multiple valley phonons in monolayer WSe₂. The monolayer WSe₂ hosts stable and long-lived dark exciton and trion as ground states[153; 154; 155; 156; 157; 158]. The resulting accumulated exciton and trion populations are highly desirable for studying their interactions with phonons. We found that three valley phonons facilitate efficient spin-conserving intervalley scattering, which results in a series of dark exciton and dark trion phonon-replicas in the low temperature photoluminescence (PL) spectrum. The sign and magnitude of Landé effective g-factors of the various replicas, together with their PL helicity under optical pumping, reveals that the spin-preserving intervalley scattering

of the electron is more efficient than its intravalley spin-flip during the dark exciton/trion formation process. This results in a surprising finding: the single electron in both the positive dark trion and intervalley exciton resides in the valley opposite to that which the optical pump is coupled to. Moreover, the identified intervalley exciton resonance enables us to infer a short-range electron-hole exchange interaction of 10 meV by extracting the energy splitting between intervalley and dark (intravalley) excitons.

The samples are exfoliated monolayer WSe₂ encapsulated between thin flakes of hexagonal boron nitride (hBN). Few-layered graphene serves as a local bottom gate for electrostatic control of the monolayer carrier density. Figure 4.2 shows the PL intensity plot as a function of gate voltage (V) and photon energy, at a temperature of 1.6 K. The laser energy is 1.775 eV with right circularly polarized (σ^+) excitation and unpolarized detection. Several previously identified excitonic states are indicated in the figure, including the neutral bright exciton, bright trions[145; 151; 180; 181; 135], the intravalley spin-forbidden dark exciton[153; 154; 155; 156; 182], and dark trions[154; 183]. The recently identified zone-center $\Gamma_5 -$ or $E'' -$ phonon replicas below both neutral[176; 177] and charged dark excitons[176] are also resolved, as indicated by black arrows.

Our focus is to understand the valley phonon origin of several previously unidentified PL peaks. In particular, we focus on the triplet and quadruplet PL peaks outlined by dashed black boxes, and those pointed at by the white arrows in Fig. 4.2. We have measured multiple samples that exhibit similar spectrum, and the nearly identical power dependence of the peaks of interest across several samples rules out the possibility of them arising due to defect states. As we explain below, all these peaks arise from the coupling of optically dark states to the bright ones via emitting valley phonons, which results in a series Stokes-shifted valley phonon-replicas of the dark exciton and dark trions in the PL spectrum.

We first consider the quadruplet PL peaks in the hole doping regime. Fig. 4.3a shows the circular polarization resolved PL with σ^+ polarized excitation. The photon energy is relative to the positive dark trion (D^+), and the intensity of the quadruplets is multiplied by a factor of 4 to emphasize these weak spectral features. We ascribe the four peaks to bright replicas of the dark trion, mediated by interactions with phonons. The quadruplets are labeled as $D_{K_3}^+$, $D_{\Gamma_5}^+$, $D_{K_1}^+$, $D_{K_2}^+$, which correspond to their origin from coupling of D^+ with valley phonons K_3 , K_1 , and K_2 , and the zone-center phonon Γ_5 . This nomenclature has its origin in the Koster notation of the K-point irreducible representations of the C_{3h} point double-group

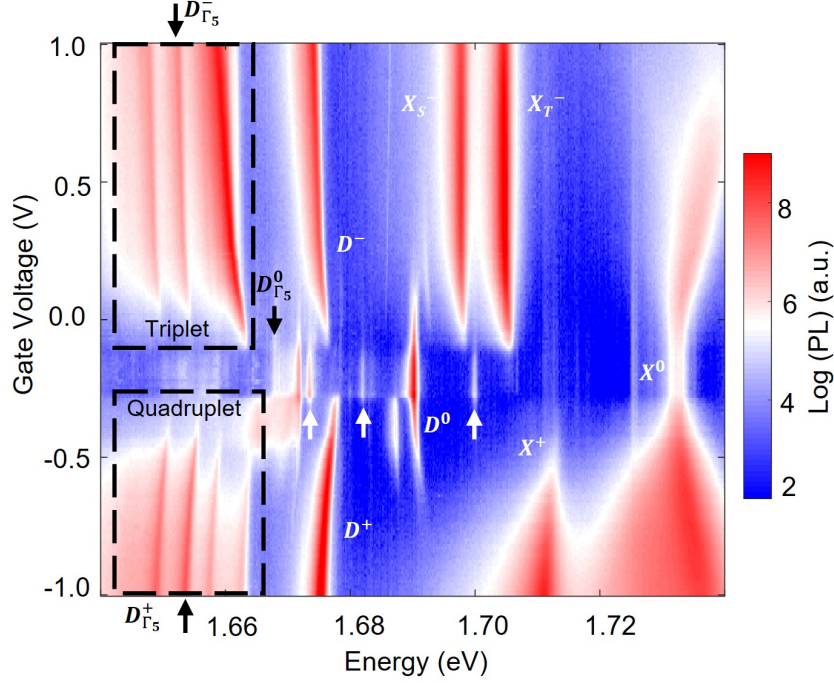


Figure 4.2: Gate dependent photoluminescence. Photoluminescence (PL) as a function of back gate voltage and photon energy. Excitonic states which have been reported in the literature are identified and marked. X^0 : neutral exciton; X^+ : positive trion; X_S^- and X_T^- : intravalley and intervalley trion; D^0 : spin forbidden neutral dark exciton; D^+ : positive dark trion; D^- negative dark trion. The unidentified triplet PL peaks at electron doping, quadruplet PL peaks at hole doping, and three states at the neutral regime, pointed to by the white arrows, are the focus of this work.

corresponding to the symmetry of the monolayer semiconductors.

The valley phonon replicas $D_{K_3}^+$, $D_{K_1}^+$, $D_{K_2}^+$ are 26, 18, and 13 meV below D^+ , respectively. The association with zone-edge phonons K_3 , K_1 , and K_2 is two-fold. The first reason is that the energy differences between D^+ and its replicas match the energies of the associated phonon modes (Figure E.1b). The second reason is rooted in the selection rules of electron-phonon coupling[171]; intervalley transitions of electrons and holes are mediated only by specific phonons (see discussions in Appendix E.1). Finally, we note that $D_{\Gamma_5}^+$ appears 22 meV below D^+ , which is consistent with the recently reported Γ_5 phonon replica of D^+ [176].

To further examine these assignments, we performed magneto-PL to extract the effective Landé g factors of the various states, which are then used to identify the spin and valley indices of the constituent electrons and holes in the excitonic states[146; 147; 184; 148; 149]. The extracted g factors of the states of interest are

listed in Table 4.1. In summary, for intravalley electron-hole recombination, the magnitude of the g-factor is about 4 for parallel electron and hole spins[146; 147; 148; 149], and about 9 when the two spins are anti-parallel[176; 177; 157]. Meanwhile, if the recombination involves electron and hole from opposite valleys but with parallel spins, a value about 13 is expected.

Figure 4.3b shows the circular polarization resolved PL intensity as a function of out-of-plane magnetic field. The energy scale is relative to the position of D^+ at zero applied magnetic field. Both the cross pattern of D^+ in Fig. 4.3b and its unpolarized light emission (see Fig. 4.3a) result from the underlying out-of-plane dipole orientation[176], and are hallmarks of direct intravalley recombination of dark trions. Following the convention of valley Zeeman splitting as $\Delta = E(\sigma^+) - E(\sigma^-)$, where $E(\sigma^+)$ and $E(\sigma^-)$ are the peak energies of the σ^+ and σ^- polarized PL components, we obtained $g(D^+) = -8.6$. The obtained g-factor of D^+ is therefore consistent with expectations for the direct recombination through the intravalley spin-flip transition.

The Zeeman shifts of the quadruplet PL peaks underpin their origin as phonon replicas of the dark positive trion D^+ . The extracted g-factors of -13.4, 12.2, and -13.0 for $D_{K_3}^+$, $D_{K_1}^+$, $D_{K_2}^+$, respectively, are consistent with intervalley recombination of electron and hole. Note that the g factor sign of $D_{K_1}^+$ is opposite to others, which will be discussed later. While the intervalley recombination is naturally forbidden for delocalized exciton complexes, because of the large momentum mismatch, emission of a valley phonon can supply the required momentum, resulting in phonon-assisted luminescence of the otherwise dark states. Combined with the concurrence of similar valley phonon energies and energy difference between the peaks and D^+ , we conclude that $D_{K_3}^+$, $D_{K_1}^+$, $D_{K_2}^+$ are K_3 , K_1 , and K_2 valley phonon replicas of D^+ .

The polarization of the $D_{\Gamma_5}^+$ peak highlights the importance of valley phonons in the formation process of D^+ . From Fig. 4.3a, we observe that D_{Γ}^+ is cross-circularly polarized. The valley optical selection rules dictate that σ^+ excitation creates an electron and hole in the $+K$ valley, while σ^- polarized emission can only happen through spin-conserved electron-hole recombination in the $-K$ valley. In addition, $g(D_{\Gamma}^+) = -9.7$ indicates the intravalley electron-hole recombination nature of the peak. The cross-polarized emission of D_{Γ}^+ therefore leads to the surprising conclusion that σ^+ excitation results in D^+ with the single electron located in the K valley: i.e. $D^+(-K)$.

The cross polarization of D^+ can be understood by considering the impact of phonons on the electron

relaxation pathways following photoexcitation. The D^+ population is created by optical pumping of the spin-conserved interband transition (Fig. 4.3c), followed by the relaxation of the electron from the higher energy spin-valley locked sub-band to the lower energy one. The latter step requires either a spin-flip or a valley-flip. From symmetry analysis, the Γ_5 phonon can lead to the intravalley spin-flip relaxation of electron from upper to lower conduction band[171], but it cannot cause intervalley scattering. On the other hand, spin-conserving intervalley scattering of the electron with K_3 phonon is a symmetry-allowed zeroth-order channel[172]. Fig. 4.3c illustrates the intervalley electron-phonon relaxation process that leads to the formation of $D^+(-K)$. First, σ^+ excitation creates electron in the spin up conduction band in the K valley. Assisted by the K_3 valley phonon (Fig. 4.3d), this spin up electron is then scattered into the spin up band in the $-K$ valley, forming $D^+(-K)$ with two holes separately located at the top of $\pm K$ valleys. $D^+(-K)$ then couples to σ^- polarized photon by emitting Γ_5 phonon, as shown in Fig. 4.3e. Evidently, the observed cross-polarized $D_{\Gamma_5}^+$ emission implies that the valley-flip rate exceeds the spin-flip one in the relaxation of electron.

Having established that σ^+ polarized excitation results in $D^+(-K)$, the understanding of both $D_{K_3}^+$ and $D_{K_2}^+$ is straightforward. As indicated in Fig. 4.3f, the spin up electron in the lower $-K$ sub-band is virtually scattered to the higher $+K$ sub-band by emitting either a K_3 or K_2 valley phonon. The spin-conserving intervalley scatter then allows for recombination with the hole in the $+K$ valley, emitting σ^+ polarized photon with energy either 26 meV K_3 or 13 meV K_2 below D^+ . The g-factors of $D_{K_3}^+$ and $D_{K_2}^+$ are nearly equal, about -13.4 and -13.0, respectively, and correspond to expected values for the intervalley spin-conserving electron-hole recombination.

Moreover, the measured amplitude of $D_{K_3}^+$, which is several times stronger than that of $D_{K_2}^+$, is also consistent with group-theory selection rules. In particular, the selection rules dictate that, in pristine monolayer WSe₂, only the K_3 phonon mode can induce intervalley electron transitions between conduction band edges at the high symmetry K and $-K$ points⁷. Meanwhile, intervalley electron transitions that are mediated by other K -point phonon modes are higher-order processes with correspondingly smaller amplitudes that involve electron states in the neighborhood of $\pm K$. Such relatively weak processes can be amplified by several possible sources such as localization next to defects, breaking WSe₂ mirror inversion symmetry by hBN encapsulation, or WSe₂/hBN moire superlattice providing in plane momentum.

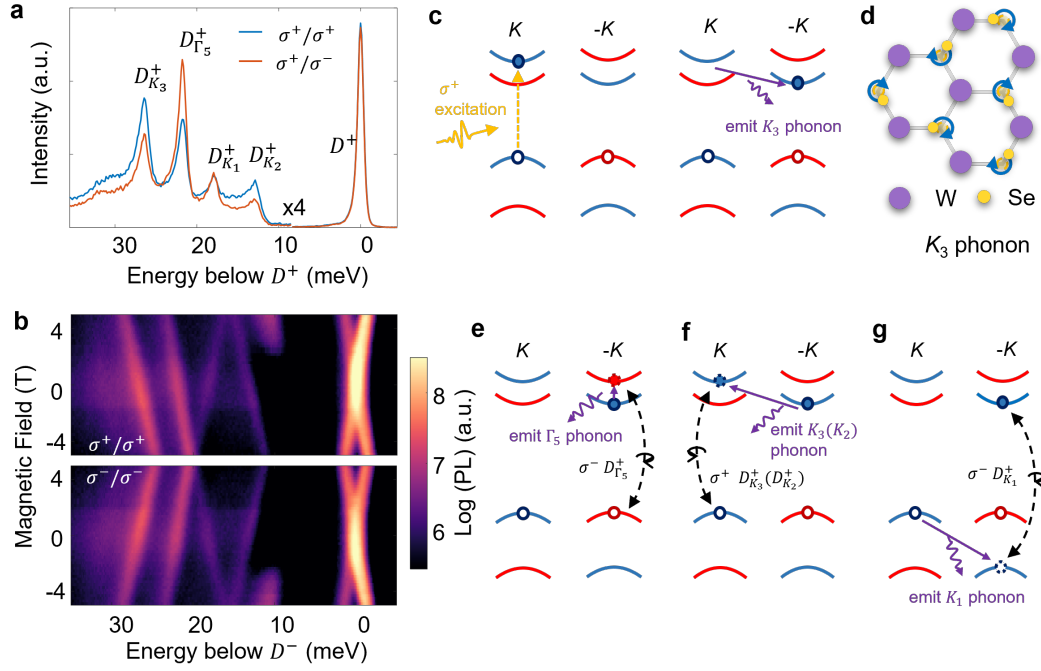


Figure 4.3: Valley phonon replicas of positively charged dark trion. **a**, Circular polarization resolved PL of the quadruplet PL peaks on the hole doping side. The photon energy of the spectrum is offset with respect to the positively charged dark trion (D^+). The intensity of the quadruplets is multiplied by a factor 4 to emphasize the weak spectrum features. **b**, Circular polarization resolved magneto PL of the quadruplets with σ^+/σ^+ (top) and σ^-/σ^- (bottom) excitation/detection. **c**, Schematic of D^+ formation process under σ^+ excitation. Blue and red represent bands with electron having spin up and down, respectively. Filled and unfilled circles represent electron and missing electron (hole) in the conduction and valence band, respectively. Orange dashed line indicates photo excitation. Purple arrow denotes the emission of valley phonon. **d**, Illustration of vibrational normal mode of K_3 phonon, wherein Se atoms orbit around their equilibrium positions. See Supplementary Figure 6 for other valley phonons. **e-g**, Schematic of phonon-assisted emission process of $D_{\Gamma_5}^+$, $D_{K_3}^+$ ($D_{K_2}^+$), and $D_{K_1}^+$ phonon replicas, respectively. Dashed circle: virtual state. As indicated by the purple arrow, the optically dark state couples to the bright ones by emitting a valley phonon, resulting in a Stokes shift of the dark trion emission by the recombination of an electron-hole pair (black dashed line). See text for details.

	bright state	dark state	k_2 replica	k_1 replica	γ_5 replica	k_3 replica
Neutral regime: I^0	-	-12.5	-	12.0	-	-12.6
Neutral regime: D^0	-4.7	-9.1	-	-	-9.8	-
Hole doped: D^+	-2	-8.6	-13.0	12.2	-9.7	-13.4
Electron doped: D^-	X_T^- : -5.2	-9.5	-13.6	12.2	-9.9	-12.5
	X_S^- : -3.4					

Table 4.1: Effective Landé g-factors of WSe₂ excitonic states and their phonon replicas.

The positive g-factor of $D_{K_1}^+$ is a signature of the interaction of the hole and K_1 valley phonon. According to the group-theory selection rules, K_1 is the only phonon mode that enables the spin-conserving intervalley transition between valence-band states at K and $-K$ points (i.e., it is a zeroth-order process)[171]. As shown in Fig. 4.3g, the hole in the higher valence band of K valley is virtually scattered to the lower valence band of the $-K$ valley with the same spin orientation via emission of a K_1 valley phonon, forming an intermediate virtual B trion. The recombination of the electron with this scattered hole in the $-K$ valley results in $D_{K_1}^+$, with energy 18 meV below D^+ . While the initial and final states have the same spin-valley configuration as in $D_{K_3}^+$ and $D_{K_2}^+$, the emission from $D_{K_1}^+$ is σ^- polarized. Therefore, the g-factor of $D_{K_1}^+$ is expected to have similar magnitude, but opposite sign, compared to the g-factors of $D_{K_3}^+$ and $D_{K_2}^+$, as we have observed. Note that although the interaction between the hole and valley phonon K_1 is relatively strong, the coupling of $D^+(-K)$ (or A trion) with the virtual B trion state is 400 meV detuned, which is much larger than in the case of K_3 intervalley electron scattering. Therefore, the PL intensity of $D_{K_1}^+$ is expected to be several times weaker than that of $D_{K_3}^+$, which is in agreement with our observation.

The valley phonon replicas of the negative dark trion D^- can be understood using similar analysis as above. Fig. 4.4a shows the helicity-resolved PL under σ^+ polarized excitation. The lower energy spectral features, denoted as T_1 , $D_{K_3}^-$, and D_{Γ}^- show appreciable co-circular polarization. Fig. 4.4b shows the PL intensity as a function of magnetic field and photon energy, with σ^-/σ^- polarized excitation/detection. The extracted g-factor of $D_{K_3}^-$ is -12.5, indicating intervalley recombination, and the energy is 26 meV below D^- . As such, we can identify that $D_{K_3}^-$ originates from the interaction of $-K$ valley electron with K_3 phonon, as depicted in the inset of Fig. 4.4a. The g-factor of D_{Γ}^- (-9.9) is nearly the same as that of D^- (-9.5), and as expected, the peak appears 22 meV below D^- .

The observation of triplet peak pattern in the PL spectrum under electron doping is slightly different from the quadruplet one observed under hole doping. Since T_1 is more intense than D^- , and its g-factor is about -4.5, it is not likely to be a phonon replica of D^- . Its origin is unknown, but T_1 is near the spectral range of the K_2 and K_1 valley phonon replicas, and thus obscures them entirely. Nevertheless, the large difference in g-factor of T_1 from that of D^- allows us to identify $D_{K_2}^-$ in magneto-PL, since the states shift away from one another. We note that there is a faint line in Fig. 4.4b, appearing on the high energy shoulder of T_1 at high magnetic field. We ascribe this peak to $D_{K_2}^-$ based on its g-factor of about -13.6, and its energy

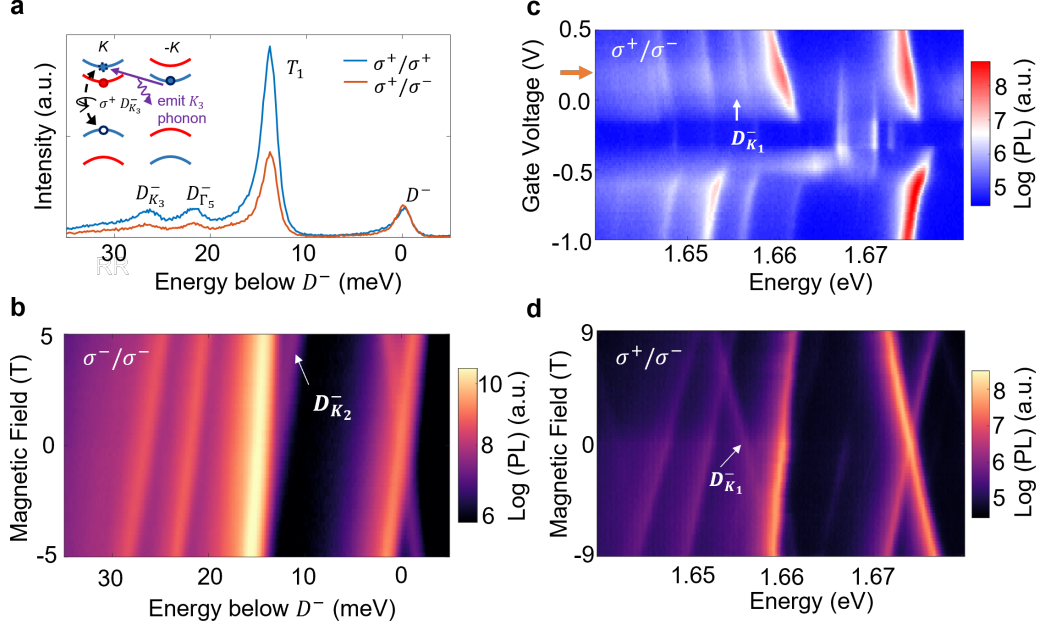


Figure 4.4: Valley phonon replicas of negatively charged dark trion. **a**, Circular polarization resolved PL of the triplet PL peaks under electron-doped conditions. The photon energy of the spectrum is offset with respect to the negatively charged dark trion (D^-). **b**, Magneto PL of the triplets with σ^- excitation and co-polarized detection. The photon energy of the spectrum is offset with respect to D^- at zero field. **c**, Gate dependent PL with σ^+ resonant pumping of the bright exciton and cross polarized (σ^-) detection. The $D_{K_1}^-$ state on the electron doping side, which is 18 meV below D^- , is clearly resolved. **d**, Magneto PL at the gate voltage indicated by the orange arrow in **c**, with σ^+ excitation and σ^- detection.

difference from D^- of 13 meV, both of which are in good agreement with expectations for K_2 phonon replica.

Meanwhile, $D_{K_1}^-$ becomes evident when the excitation laser is resonant with the bright neutral exciton (X^0). Fig. 4.4c presents the PL intensity as a function of gate voltage and photon energy, with σ^+ polarized excitation laser in resonance with X^0 (1.733 eV) and σ^- polarized detection. For $V = 0.2V$, arrow on side of Fig. 4.4c, $D_{K_1}^-$ becomes apparent at 18 meV below D^- . Fig. 4.4d shows magneto-PL with the same cross-polarized polarization (σ^+/σ^-). We find that $D_{K_1}^-$ has a positive g-factor of 12.2, which is identical to that of $D_{K_1}^+$. Therefore, despite the initial appearance, the four phonon replica states observed under hole-doping also appear under electron-doping conditions. Moreover, we observe similar magnitude and sign of g-factors, as well as energy separation from the parent dark trion state.

The valley phonon-assisted momentum relaxation mechanism also produces phonon replicas for neutral

dark excitons. Fig. 4.5a shows the helicity-resolved PL spectra near charge neutrality ($V = 0.2V$). The energy axis is offset from I^0 , which is the sharp line 32 meV below the bright neutral exciton in Fig. 4.2c. We also observe strong direct recombination from the neutral intravalley dark exciton D^0 , at 42 meV below X^0 . We confirm that D^0 has negligible circular polarization, resulting from its out-of-plane dipole orientation, and the extracted g-factor (-9.1) is in good agreement with previously reported values[176; 177; 157]. Magneto-PL measurements with linear excitation (V) and σ^+/σ^- collection are shown in the top/bottom panel of Fig. 4.5b, respectively. The observed PL peak at 22 meV below D^0 , with a g-factor of -9.8 is consistent with the reported Γ_5 phonon replica D_Γ^0 [176; 177]. We further note that D_Γ^0 shows a zero-field splitting of 0.6 meV, which arises from the fine structure of D^0 [172; 157]. We note that there are two replicas of D^0 that are apparent in the magneto-PL (cross-patterns at 3 meV and 13 meV below D^0). The peak position of the latter one matches that of positive dark trion thus is precursor of D^+ , while the former one cannot be unambiguously identified at this time.

The I^0 emission is distinct from that of D^0 and exhibits all the expected behavior of the momentum indirect, or intervalley dark exciton, which has not been previously identified in this system. This emission peak is strongly co-circularly polarized, with near unity polarization. Its g-factor of about 12.5 corresponds to the recombination of electron and hole residing in opposite valleys, implying that I^0 is the direct recombination of intervalley exciton. The formation of I^0 is a consequence of the fast valley-flip of electrons via scattering with K_3 phonon in the dark exciton formation, as illustrated in Fig. 4.5c. This is consistent with formation of $D^+(-K)$ discussed above (Fig. 4.3c). The 10 meV energy splitting between D^0 and I^0 is then a direct measure of the short-range electron-hole exchange interactions[185]. We note that the direct photon emission from the momentum indirect I^0 is weak in intensity, even when compared to the spin-forbidden dark state D^0 . However, weak PL from indirect states is not unprecedented. Similar to the case of indirect band-gap semiconductors such as silicon, recombination of intervalley excitons without phonons can be mediated by localization next to defects[186], which alleviates the need to conserve crystal momentum due to translation symmetry.

The assignment of I^0 is corroborated by the identifications of its K_1 and K_3 phonon replicas, made evident by examining the energy, polarization, and g-factors of the spectral features. The feature indicated as $I_{K_1}^0$ in Fig. 4.5a, is located 18 meV below I^0 , is cross-circularly polarized, and has a positive g-factor

of 12.0. These values are nearly identical to those found for both $D_{K_1}^+$ and $D_{K_1}^-$, and consistent with expectations for the K_1 valley phonon replica of I^0 . The $I_{K_1}^0$ recombination process via intervalley hole scattering is illustrated in Fig. 4.5d. In addition, the peak $I_{K_3}^0$ is about 26 meV below I^0 and has a g-factor of -12.6, which is the same as I^0 and supports its origin as K_3 valley phonon replica of I^0 (Fig. 4.5e).

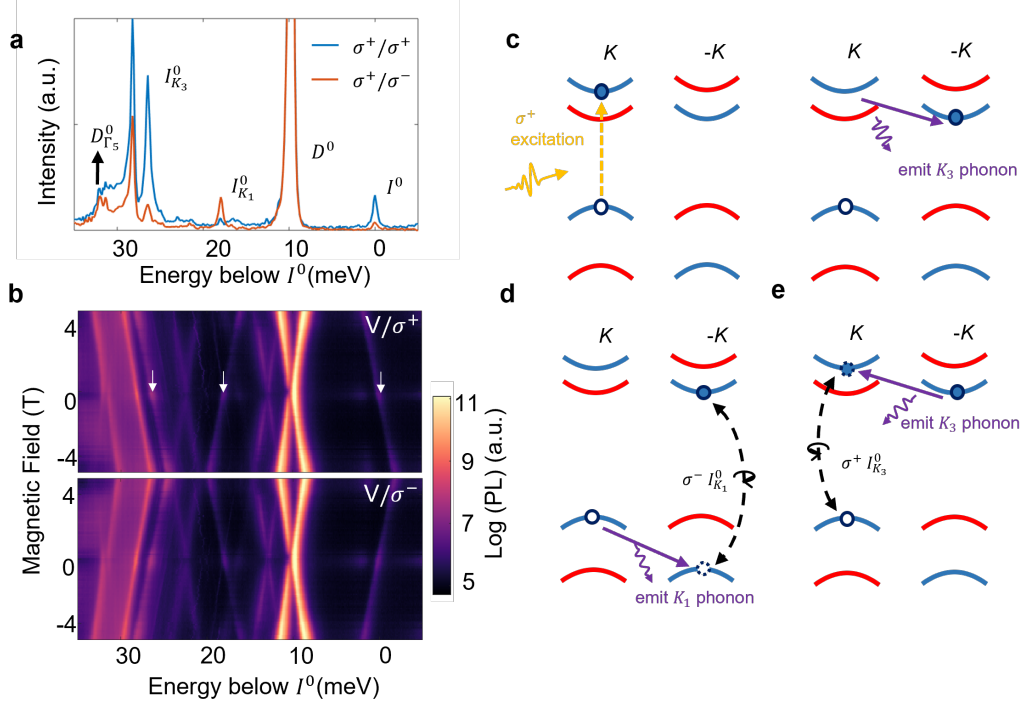


Figure 4.5: Identification of intervalley dark exciton and its valley phonon replicas. **a**, Circular polarization resolved PL in the neutral regime. The photon energy of the spectrum is offset with respect to the intervalley exciton (I^0). **b**, Magneto PL with linearly polarized excitation, σ^+ (up) and σ^- polarized (down) detection. $I_{K_3}^0$, $I_{K_1}^0$ and I^0 are labelled by the white arrows, from left to right. **c**, Schematic of intervalley exciton formation process under σ^+ excitation, with assistance of K_3 phonon. **d-e**, Schematic of light emission process of $I_{K_1}^0$ (left) and $I_{K_3}^0$ (right) phonon replicas.

In conclusion, we unravel the role of valley phonons in exciton and trion formation and their recombination in semiconducting monolayer WSe₂. Our work settles questions of the origin of nearly all of the observed peaks in the complex excitonic spectrum of monolayer WSe₂. Another important result is that the relaxation of optically generated electrons from the upper conduction band to the lower conduction band is dominated by K_3 phonon-assisted spin-conserving intervalley scattering, rather than the Γ_5 phonon-assisted spin-flip intravalley scattering. Such a relaxation pathway gives rise to the unexpected initial state for the positively charged dark trion, and efficient formation of intervalley exciton I^0 . This understanding is im-

portant for correct interpretation of excitonic spectral features, and may allow for new schemes to control the electron/exciton spin-valley state via optical pumping, e.g. coherent control of D^0 and I^0 populations via stimulated Raman adiabatic passage. Our work further motivates detailed studies of the electron-valley phonon coupling matrix elements by the first principle calculations, which should provide insights for theoretical models to gain a complete understanding of the complex monolayer WSe₂ spectrum.

4.3 Optical sensing of twisted bilayer graphene with exciton Rydberg states

In the previous section, we have discussed a variety of excitonic species that are hosted in monolayer MX₂, in particular those in WSe₂ (Figure 4.2). In essence, these tightly bound excitons arise from strongly reduced Coulomb screening in the 2D limit. While the excitons embedded in bulk materials experience a uniform dielectric environment ϵ_{3D} , the excitons in monolayer MX₂ sense both its own dielectric ϵ_{2D} and the dielectric of its adjacent layers $\epsilon_{top/bottom}$. By carefully engineering the dielectric environment, one can tune the exciton binding energy, as well as the electronic bandgap, by hundreds of meV [187].

A parallel yet more exciting idea is to explore this unique sensitivity as an excitonic probe for materials' dielectric properties [34; 188; 189]. By integrating monolayer MX₂ sensor into the target Van der Waals heterostructure, it is convenient to read the charge incompressible (or compressible) states from enhanced (or quenched) exciton Rydberg states amplitude and their energy. Moreover, the exciton Rydberg states are sensitive to charge layer polarization [190] and can provide insight on the dielectric properties at the metal-insulator transition [191].

In this section, we discuss optical sensing measurements of twisted bilayer graphene using the exciton Rydberg states. We observe multiple moiré Rydberg exciton states of the WSe₂ 2s state in the heterostructure of WSe₂/tBLG. Moreover, the moiré Rydberg excitons emerge sequentially with increasing charge carrier density, driven by the doping enhanced moiré potential.

Figure 4.6a shows the schematic of a typical WSe₂/tBLG device, where the monolayer WSe₂ sensor is stacked directly on top of the twisted bilayer graphene. While the induced spin-orbit coupling (SOC) could tip the balance of the closely competing correlated ground states, the electronic band structure of tBLG is only weakly perturbed and the lowest moiré bands remain flat [192; 193; 194]. We first examine the transport properties of these WSe₂/tBLG devices and determine their twist angle. Figure 4.6c shows

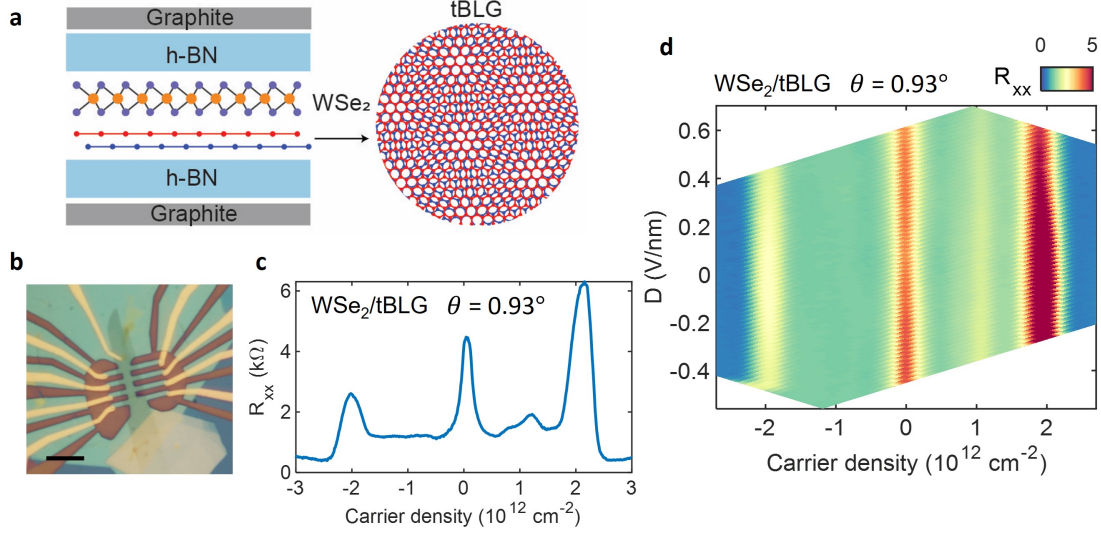


Figure 4.6: Schematic of device geometry and transport characterizations. **a**, Schematic of the device geometry employed in this study. The WSe₂ sensor is stacked directly on top of twisted bilayer graphene. The device has both top and bottom local graphite gates to achieve individual control over displacement field and carrier density. **b**, Optical image of a WSe₂/tBLG $\theta = 0.93^\circ$ device. The scale bar represents 7 μm . **c**, Resistance R_{xx} as a function of carrier density in the $\theta = 0.93^\circ$ device. **d**, Dual gate map of R_{xx} as a function of displacement field D and carrier density n . The transport feature is nearly independent of D . All data are taken at 2 K.

resistance R_{xx} of a WSe₂/tBLG device, measured at $T = 2\text{K}$ (device shown in Fig. 4.6b). The resistance peaks at carrier density $n = \pm 2 \times 10^{12}\text{cm}^{-2}$ mark the $\nu = \pm 4$ band insulators, separating the lowest flat bands from remote bands. Therefore, we determine the twist angle of tBLG to be $\theta = 0.93^\circ$ in this device. The transport properties of WSe₂/tBLG remain mostly independent of the out-of-plane displacement field D (Figure 4.6d), consistent with previous reports [193; 194].

To access the WSe₂ excitonic states, we perform optical reflection measurements on the WSe₂/tBLG devices. Figure 4.7a shows the reflection contrast $dR/R = \frac{R-R_0}{R_0}$ of the monolayer WSe₂ (top) and the WSe₂/tBLG device at $\theta = 0.93^\circ$ (bottom) respectively. In the optical spectrum of monolayer WSe₂, we identify the 1s and 2s excitons at 1.736 eV and 1.866 eV respectively. In the WSe₂/tBLG heterostructure, both the WSe₂ electronic bandgap and the exciton binding energy are reduced by the screening effect of the adjacent graphene, giving rise to an overall red shift of both 1s and 2s exciton energy to 1.709 eV and 1.789 eV. Moreover, the graphene screening effect suppresses the WSe₂ 2s exciton's oscillator strength by an order. Figure 4.7b further shows the gate dependent reflection contrast map of the monolayer WSe₂ (top) and the

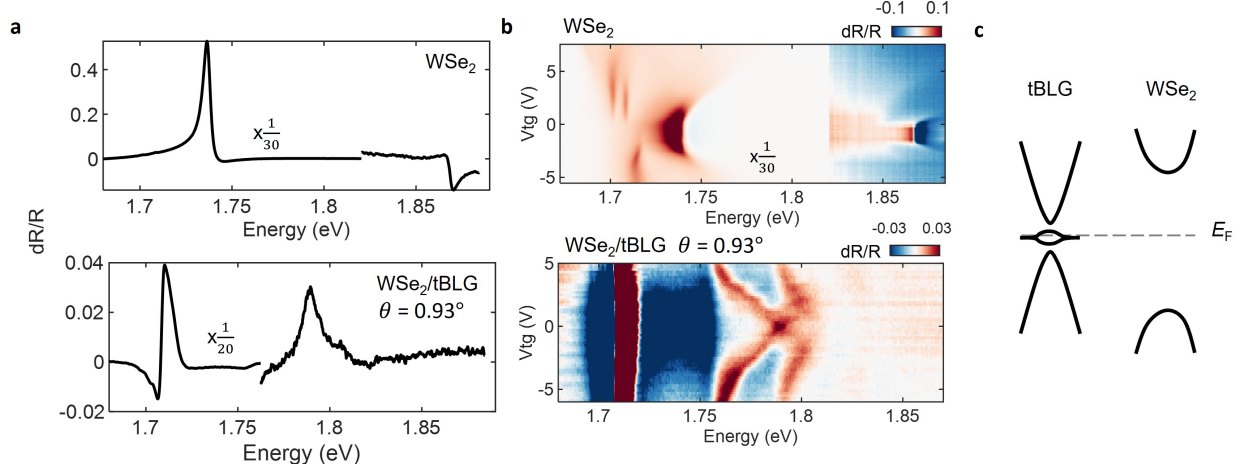


Figure 4.7: Reflection contrast of WSe₂/tBLG and its band alignment. **a**, Reflection contrast of a monolayer WSe₂ (top) and WSe₂/tBLG (bottom) with $\theta = 0.93^\circ$ at zero gate voltage. Data is taken in the same device, at $T = 5$ K. The strong WSe₂ 1s states are converted to a comparable scale with the 2s states in each plot. **b**, Gate dependent reflection contrast map of a monolayer WSe₂ (top) and WSe₂/tBLG (bottom) with $\theta = 0.93^\circ$. **c**, Schematic of the band alignment of WSe₂/tBLG. The Fermi energy can be gate tuned across the lowest flat bands in tBLG, while it always remains in the electronic bandgap of WSe₂.

WSe₂/tBLG device at $\theta = 0.93^\circ$ (bottom). We identify two negatively charged excitons (X_S^- and X_T^-) and a positively charged exciton (X^+) in the electron and hole doped regime separately as previously reported. In stark contrast, we observe a robust 1s exciton, almost independent of charge doping in the optical spectrum of WSe₂/tBLG. It suggests that the carrier density in the WSe₂ layer remains unchanged regardless of the gate voltage. This can be explained by the band alignment of the tBLG and WSe₂ (Figure 4.7c), where the Fermi energy is only slightly gate tuned around the tBLG charge neutrality and always remains within the WSe₂ bandgap.

The WSe₂ 2s Rydberg state, however, shows strong red shift with both increasing electron and hole doping. More interestingly, a new higher energy excitonic state emerges above certain doping threshold, on both the electron and hole doping sides. This new excitonic state then also exhibits a red shift with increasing doping but with a slower rate (Figure 4.8a). We identify the origin of the new excitonic state to be the moiré potential induced exciton band folding. Like the electronic band folding, the exciton bands can also be folded into a moiré Brillouin zone and give rise to new excitonic states at the zone center (Γ point) (Figure 4.8b). Similar moiré exciton states have been reported in the heterostructure of WSe₂/WS₂ [195], where the WSe₂ 1s exciton state (intralayer exciton) is reconstructed to three moiré exciton states. More recently, moiré

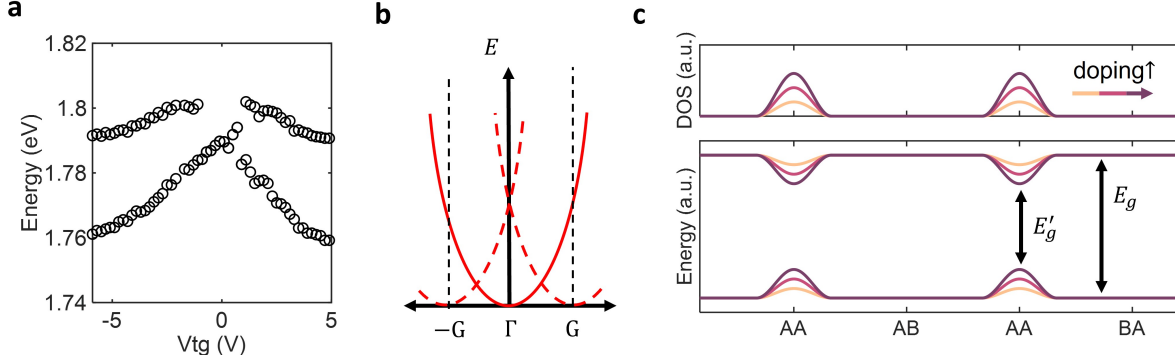


Figure 4.8: Moiré Rydberg exciton states and doping dependent moiré potential. **a**, Extracted energy of the WSe₂/tBLG 2s Rydberg exciton states in the $\theta = 0.93^\circ$ device. **b**, Schematic of exciton band folding. $\mathbf{G}$ is the momentum space unit vector. New excitonic states emerge at Γ as a result of band folding. **c**, Top panel shows the schematic of the density of states (DOS) in tBLG at different doping. Bottom panel shows the WSe₂ electronic bandgap at different doping. The local bandgap E'_g at AA sites are strongly reduced by the screening, creating a doping dependent moiré potential $E_0 = E_g - E'_g$.

Rydberg exciton states are also reported in the heterostructure of WSe₂ and aligned graphene/h-BN [188].

The energy separation between the moiré exciton states are decided by two main factors: the moiré superlattice periodicity λ [188] and the moiré potential E_0 [195]. Figure 4.8a shows the extracted energy of the moiré Rydberg exciton states in the WSe₂/tBLG device at $\theta = 0.93^\circ$, showing an increasing energy separation with doping. Therefore, our observation reveals a doping enhanced moiré potential E_0 , in stark contrast to the previous report [188]. We show that the doping dependent moiré potential E_0 is a unique consequence of the tBLG's flat bands. The top panel in Figure 4.8c shows the local density of states (DOS) at different doping levels. In the flat band regime, the charge are always highly localized at AA atomic sites. The periodic charge distribution in tBLG imprints a periodically screened electronic bandgap in the monolayer WSe₂. The moiré potential E_0 can be understand as the band gap difference between the AB/BA sites E_g and AA sites E'_g . With increasing doping, the E'_g is dynamically suppressed by the increased local DOS at AA sites while the E_g remains almost unchanged. However, in the case of moiré Rydberg excitons in WSe₂ and aligned graphene/h-BN, the electronic bands are much more dispersive and the charge distribution is more uniform. As a result, the moiré potential E_0 in the latter case is not doping dependent.

The identification of the moiré Rydberg excitons are further confirmed by WSe₂/tBLG devices at other twist angles. Figure 4.9a shows the gate dependent reflection contrast map of a WSe₂/tBLG sample where the twist angle of the tBLG is fully relaxed. At extremely small twist angle, tBLG lattice reconstructs into

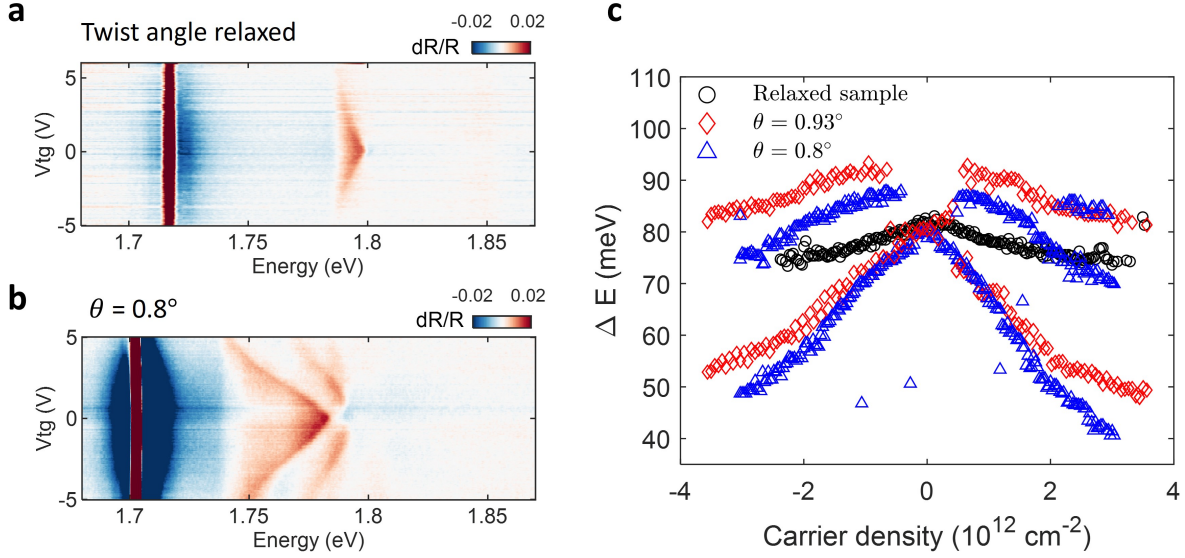


Figure 4.9: Twist angle dependence of moiré Rydberg exciton states. **a**, Gate dependent reflection contrast map of a WSe₂/tBLG sample where the twist angle of the tBLG is fully relaxed. **b**, Gate dependent reflection contrast map of a WSe₂/tBLG sample with $\theta = 0.8^\circ$. **c**, Extracted energy of the moiré Rydberg excitons as a function of carrier density. ΔE represents the energy relative to the WSe₂ 1s exciton energy.

large (several hundreds nm scale) alternating AB and BA triangular domains with vanishingly small AA sites. As a result, we observe single WSe₂ 2s Rydberg exciton lacking a well defined moiré potential E_0 in this regime. Figure 4.9b shows the gate dependent reflection contrast map of a WSe₂/tBLG sample at $\theta = 0.8^\circ$, slightly smaller than the device discussed above. The moiré Brillouin zone has a smaller unit vector G at smaller twist angle, therefore the exciton band folding results in moiré excitons with smaller energy separation in between. Our results, shown in Figure 4.9c, align well with above theoretical predictions. Moreover, we observe a third moiré Rydberg exciton state in the $\theta = 0.8^\circ$ sample above electron density $n = 2 \times 10^{12} \text{ cm}^{-2}$, signifying the enhanced moiré potential E_0 at highly doped regime.

Our results establish a new set of moiré Rydberg excitons in the heterostructure of WSe₂/tBLG, with a unique doping tuned moiré potential. These moiré Rydberg excitons provide a sensitive probe for the underlying flat bands in tBLG, and potentially the strongly correlated states for the tBLG near the magic angle.

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Chapter A

Appendix A

A.1 A survey of tDBG at different twist angle

Figure A.1k-o summarize phase diagrams for our AB-AB twisted double bilayer graphene, focusing on $\nu > 0$ as a function of D . The phase diagrams are shown for $D < 0$, but are approximately equivalent for $D > 0$ within our experimental resolution. The color shading denotes regions of the phase diagram with different inferred degeneracy, where blue corresponds to four-fold isospin degeneracy, green corresponds to two-fold isospin degeneracy, and orange has no remaining degeneracy (consistent with the coloring scheme in Fig. 2.7d). The dark gray lines denote CI states. The light gray coloring corresponds to regions of the phase diagram which are experimentally inaccessible owing to the onset of dielectric breakdown to the gates. The boundaries of the symmetry-broken regions of the phase diagram correspond to regions of the R_H map in which the sign inverts or there is an abrupt change in magnitude. The symmetry-broken region of the phase diagram appears to be largest around $\theta = 1.30^\circ$ degrees, and shrinks for both larger and smaller twist angles. Correlated states at $\nu = 1$ also emerge only near this twist angle. In devices D1 and D5, we observe large regions of metallic symmetry-broken states even in the absence of a well-developed CI state at $\nu = 2$, suggesting that the bands have started to split but are not yet fully gapped.

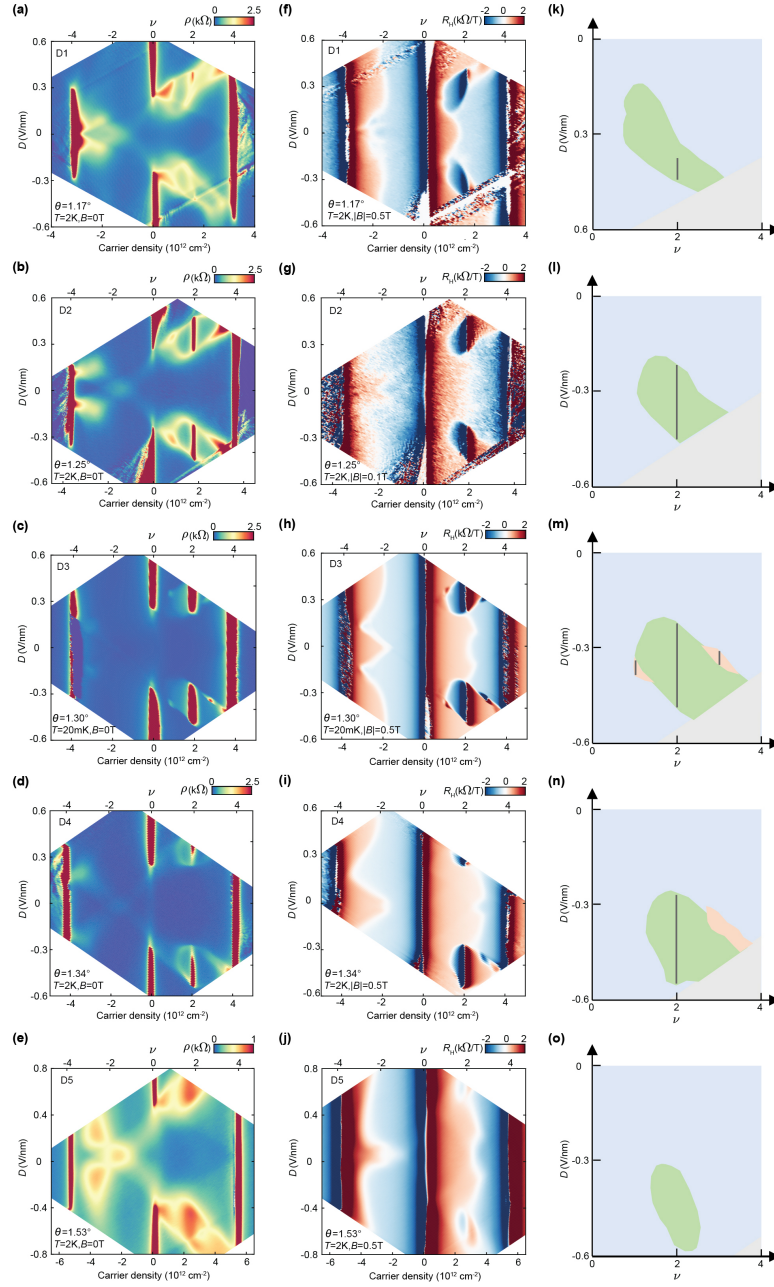


Figure A.1: Transport in AB-AB tDBG at a variety of twist angles. **a-e**, ρ as a function of n and D in five devices acquired at $B = 0$ T. All are acquired at $T = 2$ K, except for **c** which is acquired at $T = 20$ mK. **f-j**, Corresponding R_H for the same devices. The maps in **f**, **h** and **i** are antisymmetrized at $|B| = 0.5$ T, the map in **g** is antisymmetrized at $|B| = 0.1$ T, and the map in **j** is acquired at $B = +0.5$ T but not antisymmetrized. **k-o**, Schematics of the phase diagram for each device. Blue corresponds to four-fold isospin degeneracy, green corresponds to two-fold isospin degeneracy, and orange indicates no remaining degeneracy. The dark gray lines denote CI states. The light gray coloring corresponds to experimentally inaccessible regions of the phase diagram.

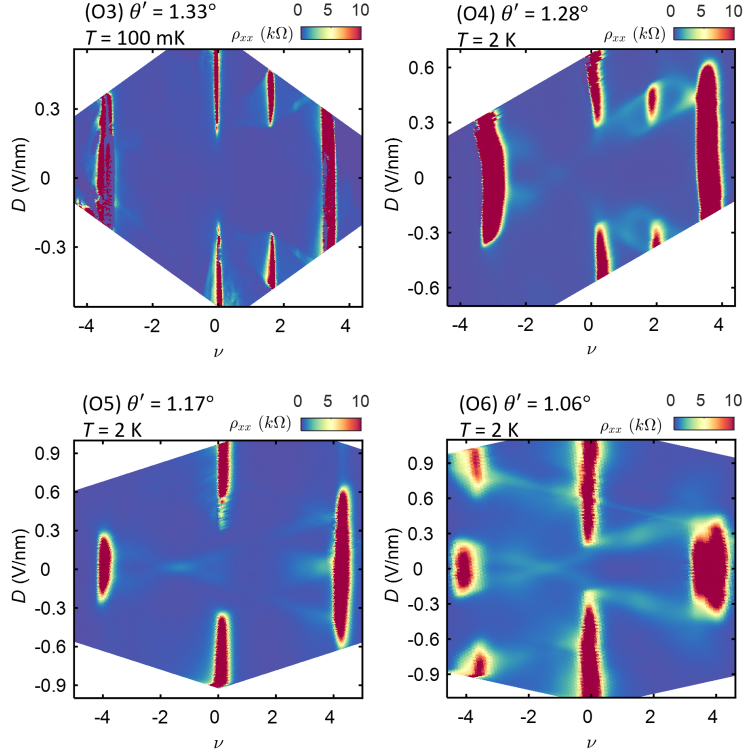


Figure A.2: Low temperature transport in additional AB-BA tDBG devices. **a-e**, ρ as a function of n and D in five devices acquired at $B = 0$ T. All are acquired at $T = 2$ K, except for **c** which is acquired at $T = 20$ mK. The twist angle and measurement temperature are denoted at the top left corner of each map. We observe displacement field-tunable insulating states at $\nu = 0$ and ± 4 in all devices. Correlated insulating states appear clearly in devices O3 and O4.

Chapter B

Appendix B

B.1 Band structure calculation of tDBG

We calculate the band structure of the tDBG, in both AB-AB stacking and AB-BA stacking, based on the standard Bistritzer-MacDonald continuum model. The full Hamiltonian for the valley K are:

$$H_{AB-AB} = H_t^{AB} + H_b^{AB} + H_M \quad (\text{B.1})$$

$$H_{AB-BA} = H_t^{AB} + H_b^{BA} + H_M \quad (\text{B.2})$$

where H_t^{AB} and H_b^{BA} , H_b^{AB} are the Hamiltonian of the two layers separately. H_M includes the interlayer moiré tunneling terms. We have

$$H_t^{AB} = \Psi_t^\dagger \begin{pmatrix} \frac{D}{2} & t(k_x - ik_y)e^{i\frac{\theta}{2}} & t_4(k_x - ik_y)e^{i\frac{\theta}{2}} & t_3(k_x + ik_y)e^{-i\frac{\theta}{2}} \\ t(k_x + ik_y)e^{-i\frac{\theta}{2}} & \frac{D}{2} & \gamma_1 & t_4(k_x - ik_y)e^{i\frac{\theta}{2}} \\ t_4(k_x + ik_y)e^{-i\frac{\theta}{2}} & \gamma_1 & \frac{D}{6} & t(k_x - ik_y)e^{i\frac{\theta}{2}} \\ t_3(k_x - ik_y)e^{i\frac{\theta}{2}} & t_4(k_x + ik_y)e^{-i\frac{\theta}{2}} & t(k_x + ik_y)e^{-i\frac{\theta}{2}} & \frac{D}{6} \end{pmatrix} \Psi_t \quad (\text{B.3})$$

and

$$H_b^{AB} = \Psi_b^\dagger \begin{pmatrix} -\frac{D}{6} & t(k_x - ik_y)e^{i\frac{\theta}{2}} & t_4(k_x - ik_y)e^{i\frac{\theta}{2}} & t_3(k_x + ik_y)e^{-i\frac{\theta}{2}} \\ t(k_x + ik_y)e^{-i\frac{\theta}{2}} & -\frac{D}{6} & \gamma_1 & t_4(k_x - ik_y)e^{i\frac{\theta}{2}} \\ t_4(k_x + ik_y)e^{-i\frac{\theta}{2}} & \gamma_1 & -\frac{D}{2} & t(k_x - ik_y)e^{i\frac{\theta}{2}} \\ t_3(k_x - ik_y)e^{i\frac{\theta}{2}} & t_4(k_x + ik_y)e^{-i\frac{\theta}{2}} & t(k_x + ik_y)e^{-i\frac{\theta}{2}} & -\frac{D}{2} \end{pmatrix} \Psi_b \quad (\text{B.4})$$

$$H_b^{BA} = \Psi_b^\dagger \begin{pmatrix} -\frac{D}{6} & t(k_x - ik_y)e^{-i\frac{\theta}{2}} & t_4(k_x + ik_y)e^{i\frac{\theta}{2}} & \gamma_1 \\ t(k_x + ik_y)e^{i\frac{\theta}{2}} & -\frac{D}{6} & t_3(k_x - ik_y)e^{-i\frac{\theta}{2}} & t_4(k_x + ik_y)e^{i\frac{\theta}{2}} \\ t_4(k_x - ik_y)e^{-i\frac{\theta}{2}} & t_3(k_x + ik_y)e^{i\frac{\theta}{2}} & -\frac{D}{2} & t(k_x - ik_y)e^{-i\frac{\theta}{2}} \\ \gamma_1 & t_4(k_x - ik_y)e^{-i\frac{\theta}{2}} & t(k_x + ik_y)e^{i\frac{\theta}{2}} & -\frac{D}{2} \end{pmatrix} \Psi_b \quad (\text{B.5})$$

In the above, we choose basis as $\Psi_{t/b} = (f_{A_{t/b;1}}, f_{A_{t/b;2}}, f_{B_{t/b;1}}, f_{B_{t/b;2}})^T$ and $f_{A_{t/b;1}}, f_{A_{t/b;2}}, f_{B_{t/b;1}}, f_{B_{t/b;2}}$ label the layer-sublattice degree of freedom of the top/bottom bilayer graphene. We use the parameters $t = -3100\frac{\sqrt{3}}{2}$ meV, $t_1 = 283\frac{\sqrt{3}}{2}$ meV, $t_4 = 110\frac{\sqrt{3}}{2}$ meV and $\gamma_1 = 361$ meV, where the momentum k_x, k_y is in the unit of $\frac{1}{a}$, and $a \approx 0.246$ nm is the lattice constant of graphene.

The interlayer moiré tunneling Hamiltonian H_M is

$$H_M = \sum_{j=1,2,3} (f_{A_{t;2}}^\dagger(\mathbf{k}), f_{B_{t;2}}^\dagger(\mathbf{k})) T_j \begin{pmatrix} f_{A_{b;1}}(\mathbf{k} + \mathbf{Q}_j) \\ f_{B_{b;1}}(\mathbf{k} + \mathbf{Q}_j) \end{pmatrix} + h.c. \quad (\text{B.6})$$

where $\mathbf{Q}_1 = 0$, $\mathbf{Q}_2 = \mathbf{G}_1 = (-\frac{2\pi}{\sqrt{3}a_M}, -\frac{2\pi}{a_M})$, and $\mathbf{Q}_3 = \mathbf{G}_2 = (\frac{2\pi}{\sqrt{3}a_M}, -\frac{2\pi}{a_M})$. The moiré lattice constant at twist angle θ is denoted as $a_M = \frac{a}{2\sin\frac{\theta}{2}}$. We have $T_j = t_M \begin{pmatrix} \alpha & e^{-i\frac{2\pi j}{3}} \\ e^{i\frac{2\pi j}{2}} & \alpha \end{pmatrix}$, where t_M is the interlayer tunneling strength and $\alpha \in [0, 1]$ is introduced to incorporate the effects of lattice relaxation. We use $t_M = 110$ meV and $\alpha = 0.8$.

At the limit of $D = 0$, there is a mirror symmetry in AB-BA stacked tDBG $M : k_x \rightarrow k_x, k_y \rightarrow -k_y$, which acts as $M_y K$, where K is the complex conjugate. M_y maps $A_{t;1} \rightarrow A_{b;2}, A_{t;2} \rightarrow A_{b;1}, B_{t;1} \rightarrow B_{t;2}, B_{t;2} \rightarrow B_{b;2}$, where the momentum changes as $k_x \rightarrow -k_x, k_y \rightarrow k_y$. There is no flip of the valley index in this transformation. We note that the realization of the mirror symmetry is different for the AB-AB

stacked tDBG, in which $M : k_x \rightarrow k_x, k_y \rightarrow -k_y$, acting as M_x , with $A_{t;1} \rightarrow B_{b;2}, A_{t;2} \rightarrow B_{b;2}, B_{t;1} \rightarrow A_{b;2}, B_{t;2} \rightarrow A_{t;2}$. Again there is no valley flip in the transformation. Note that there is no complex conjugate compared to the AB-BA stacking. As a result, the constraint of the Chern number C is different. For AB-AB stacking, we have $C(D) = -C(-D)$ within one valley, while we have $C(D) = C(-D)$ within each valley for the AB-BA stacking.

The band dispersions of AB-AB tDBG and AB-BA tDBG are quite similar [105; 89; 104; 196; 197]. We calculate the band structure of tDBG in both stacking chiralities at $\theta(\theta') = 1.2^\circ$ with a finite D , represented by interlayer potential $\delta = 40$ meV. Although the band structures are almost identical (Fig. 2.12e), the Berry curvature distributions are very different. For AB-AB stacked tDBG, the Berry curvature from the K and K' points of the moiré Brillouin zone are the same and sum together to result in $C = 2$ for the lowest conduction band. In contrast, for AB-BA tDBG, K and K' carry opposite Berry curvatures. The net Berry curvature comes primarily from the center region of the moiré Brillouin zone (*i.e.*, the Γ point), resulting in $C = 1$.

B.1.1 Topological charge density wave

We calculate reconstructed band structure and Chern number at $\nu = \frac{1}{2}$ after adding a phenomenological CDW order with 2×1 Moiré unit cell in the AB-BA stacked tDBG.

The CDW is generated by a potential with nesting momentum $\mathbf{Q} = (\frac{2\pi}{\sqrt{3}a_M}, 0)$. The interaction Hamiltonian at this momentum is given as:

$$H_V = V(\mathbf{Q})\rho(\mathbf{Q})\rho(-\mathbf{Q}) \quad (\text{B.7})$$

where $\rho(\mathbf{Q})$ is the density operator projected to the active band and $V(\mathbf{Q})$ is the Fourier-transformed screened Coulomb potential.

The mean field Hamiltonian decoupled from the interaction is:

$$H = H_{\text{nonint}} + (\Phi\rho(\mathbf{Q}) + h.c.) \quad (\text{B.8})$$

where, H_{nonint} is non-interacting Hamiltonian defined in Eq.1, $\Phi = V(\mathbf{Q}) \langle \rho(\mathbf{Q}) \rangle$ is the charge density wave

order parameter. In a more detailed form:

$$H = H_{\text{nonint}} + \sum_{\mathbf{k}} (\Phi \Lambda_{ab}(\mathbf{k}, \mathbf{Q}) + \Phi^* \Lambda_{ab}(\mathbf{k}, -\mathbf{Q})) c_a^\dagger(\mathbf{k} + \mathbf{Q}) c_b(\mathbf{k}) + h.c. \quad (\text{B.9})$$

where $\Lambda_{ab}(\mathbf{k}, \mathbf{q}) = \langle \mu_a(\mathbf{k} + \mathbf{q}) | \mu_b(\mathbf{k}) \rangle$ is a form-factor entering the density operator. $\mu_a(\mathbf{k})$ is the Bloch wavefunction for band a .

Note here Φ can be a complex number, whose phase ϕ determines the phase of the CDW order $\rho(\mathbf{r}) = \rho_0 + A \cos(\pi x + \phi)$, where x is the coordinate along the $\mathbf{a}_1$ direction. We label the AA site (maximum of the LDOS) as $\mathbf{r} = 0$ and choose $\phi = 0$ to pin the CDW with the moiré lattice (see Fig. 3e of the main text).

In the original electron operator,

$$H = H_{\text{nonint}} + \sum_{\mathbf{k}} \sum_{a,b} (\Phi \Lambda_{ab}(\mathbf{k}, \mathbf{Q}) + \Phi^* \Lambda_{ab}(\mathbf{k}, -\mathbf{Q})) \mu_{a;\alpha}(\mathbf{k} + \mathbf{Q}) \mu_{b;\beta}^*(\mathbf{k}) f_\alpha^\dagger(\mathbf{k} + \mathbf{Q}) f_\beta(\mathbf{k}) + h.c. \quad (\text{B.10})$$

where α, β is a combination of the sublattice-layer index and the $m\mathbf{G}_1 + n\mathbf{G}_2$ index. a, b is the band index. We sum a, b with M to be the band index cutoff, *i.e.*, over the conduction band, M bands below and M bands above. If we set $M = 0$, then the CDW only influences the conduction band. If $M \geq 1$, the CDW order also acts on the remote bands and couples the remote band to the conduction band. In practice, the result converges quite rapidly when increasing M . However, we note that $M \geq 1$ and $M = 0$ can give different results. The CDW order also hybridizes the remote band and the conduction band, which can renormalize the Chern number of the upper band split from the remainder of the conduction band.

We consider tDBG in both stacking chiralities at the same condition discussed above. In AB-AB tDBG, it is natural to expect that a CDW with 2×1 unit cell will split the original $C = 2$ conduction band into two $C = 1$ and $C = 1$ subbands. However, for AB-BA stacking, the $C = 1$ conduction band will split into a $C = 0$ and $C = 1$ band: the half around the zone center region carries $C = 1$, whereas the half around the K and K' carries $C = 0$. We calculate band structure in AB-BA tDBG with a CDW order $\Phi = 16$ meV. We find that the Chern numbers of the gapped subbands are sensitive to the remote band renormalization. For $M = 0$, the upper subband has $C = 0$, inconsistent with our experimental observations of a topological state at $\nu = \frac{7}{2}$. After considering $M = 4$ remote bands, the Chern number of the upper subband renormalizes to

$C = 1$, in agreement with our experiment. We note that $M = \infty$ (i.e. the realistic limit) should yield the same result for the conduction band as $M = 4$ in our calculation owing to its rapid convergence.

Chapter C

Appendix C

C.1 A survey of tMBG at different twist angle

Table C.1 summarizes the correlated states observed at all integer ν , and more recently fractional ν in the conduction bands of tMBG for both signs of D in devices over a range of twist angles, $0.89^\circ \leq \theta \leq 1.385^\circ$. The data are compiled mainly from Ref. [20; 21; 22; 120; 113]. The rotational alignment of the tMBG to the top and bottom encapsulating BN is a latent parameter. We have no reliable estimate of these twist angles for any of the device, nor do we see obvious signatures of close rotational alignment, however this may nevertheless play a role in driving variations between tMBG devices with otherwise comparable twist angles.

We now comment on a number of details of the collectively reported tMBG devices. First, we note that in principle it is possible that devices with $\theta \geq 1.25^\circ$ from Refs. [21; 113; 22] also exhibit weak AHE around $\nu = 1$ and 3 for $D < 0$ states, however such measurements are not reported in those manuscripts, so we are unable to make such a determination. We also note that a second device with $\theta = 1.25^\circ$ was also reported in Ref. [21], exhibiting a very similar phase diagram to the first. However, measurements of the AHE are not shown, and we therefore omit it from the table. Finally, we note that devices with larger twist angles up to $\sim 1.6^\circ$ have exhibited signatures of symmetry broken states for $D > 0$ [20; 22], however these states all exhibit metallic temperature dependence and have not been investigated in detail. We also omit these devices from the table.

Although identical sets of measurements have not been performed for all devices, the table appears to reveal a number of trends in the evolution of the correlated states with θ .

For $D > 0$, the $\nu = 2$ state is a robust trivial insulator over a wide range of twist angles, and appears to be spin-polarized from measurements of the gap with in-plane magnetic field, $B_{||}$ [20]. A Chern insulator state at $B = 0$ appears to emerge around $\nu = 3$ only for a small range of twist angles near $1.25^\circ \leq \theta \leq 1.29^\circ$. The AHE at $\nu = 3$ appears to approach a quantization at $h/2e^2$, suggesting the band has $C_v = 2$ at this twist angle. Moreover, a symmetry-broken Chern insulator state (SBCI) with a Chern number $C = 1$ persists down to zero magnetic field at $\nu = 7/2$ in a $\theta = 1.29^\circ$ sample [113]. In addition to the broken spin and valley symmetry at $\nu = 3$, the emergence of SBCI state at fractional filling indicates further translational symmetry breaking that doubles the moiré unit cell. The SBCI state is directly connected to an gapped state at $\nu = 7/2$, which also display a strong AHE effect. These observations point to a topological charge density wave state as the ground state at $\nu = 7/2$ at zero magnetic field. For other twist angles, symmetry-broken states at $\nu = 3$ are only observed at finite B . The $\nu = 1$ state exhibits more complicated behavior. A gapped state is observed in one device with $\theta = 0.89^\circ$ [20], but not in another at 0.90° [21]. In the former, a large AHE is observed, with quantization that appears to approach h/e^2 , suggesting the band has $C_v = 1$ at this twist angle. This behavior is consistent with a theoretically anticipated topological transition with twist angle around $\theta \approx 1.0^\circ$ [127]. The gap closes as the twist angle increases to around 1.05° , before reemerging again in devices with twist angles larger than 1.13° . The unusual behavior surrounding this state has been discussed at length in 3, but in short appears to be consistent with a transition from a Q-IVC to a spin and valley polarized state with increasing twist angle. The absence of a gapped state at slightly smaller twist angles of $\sim 1.05^\circ$ is superficially consistent with this picture, understood collectively as a decrease in U/W as the twist angle grows over this range of angles. Similar to $\nu = 7/2$, a symmetry-broken Chern insulator emanates from $\nu = 3/2$ with Chern number $C = 1$ is observed at high magnetic field in the $\theta = 1.25^\circ$ and 1.29° sample [113]. However the $\nu = 3/2$ SBCI state does not extend all the way to zero magnetic field and no corresponding AHE is reported.

For $D < 0$, the behavior of the correlated states appears to be less sensitive to twist angle. Correlated states with metallic temperature dependence are observed in devices within the approximate range $1.05^\circ \lesssim \theta \lesssim 1.25^\circ$. The lone exception is the $\nu = 1$ state in a $\theta = 1.19^\circ$ sample [120], which becomes weakly

insulating at temperatures below 1 K. In all devices for which there are suitable measurements, the correlated metallic states at $\nu = 1$ and 3 have an associated AHE. Although overlooked initially, we note that a retrospective analysis of the $\theta = 1.05 - 1.08^\circ$ device from Ref. [20] also reveals a similar AHE to the devices we report here. Gradual transitions to insulating behavior are observed upon increasing B at $\nu = 1$, whereas abrupt transitions (accompanied by hysteresis in a $\theta = 1.19^\circ$ sample) are observed at $\nu = 2$ and 3. Previous measurements from the $\theta = 1.08^\circ$ device in Ref. [20] show that these states are nearly insensitive to $B_{||}$, suggesting that all of the phase transitions are driven by orbital effects.

	$D > 0, \nu = 1$	$D > 0, \nu = 2$	$D > 0, \nu = 3$	$D < 0, \nu = 1$	$D < 0, \nu = 2$	$D < 0, \nu = 3$
0.89° (Ref. [20])	Chern insulator (approaching quantization with $C = 1$)	weakly insulating	none	none	none	none
0.90° (Ref. [21])	none	none	none	none	none	none
1.05 – 1.08° (Ref. [20])	none (weak state emerges in large $B_{ }$)	trivial insulator (gap grows with $B_{ }$, suggesting spin-polarized)	none (weak state emerges in large $B_{ }$)	metallic resistive state with AHE (incipient insulator emerges gradually with B)	metallic resistive state (insulator emerges abruptly at finite B)	metallic resistive state with AHE (insulator emerges abruptly at finite B)
1.13° (Ref. [120])	trivial insulator (AHE upon doping)	trivial insulator	none	metallic resistive state with AHE (insulator emerges gradually with B)	metallic resistive state (insulator emerges abruptly at finite B)	metallic resistive state with AHE (insulator emerges abruptly at finite B)
1.14° (Ref. [120])	trivial insulator (AHE upon doping)	trivial insulator	none	metallic resistive state with AHE (insulator emerges gradually with B)	metallic resistive state (insulator emerges abruptly at finite B)	metallic resistive state with AHE (insulator emerges abruptly at finite B)
1.19° (Ref. [120])	trivial insulator (AHE upon doping)	trivial insulator	obscured by poor contacts	trivial insulator (AHE upon doping)	metallic resistive state (insulator emerges abruptly at finite B)	metallic resistive state with AHE (insulator emerges abruptly at finite B)
1.25° (Ref. [21; 113])	Chern insulator (approaching quantization with $C = 2$) $\nu = 3/2$ SBCI with $C = 1$ at high field	trivial insulator	Chern insulator (approaching quantization with $C = 2$)	metallic resistive state	metallic resistive state	metallic resistive state
1.29° (Ref. [113])	Chern insulator with $C = 2$ $\nu = 3/2$ SBCI with $C = 1$ at high field	trivial insulator	Chern insulator with $C = 2$ $\nu = 7/2$ SBCI with $C = 1$ at zero field	none	none	none
1.385° (Ref. [21])	metallic with weak AHE	trivial insulator	symmetry-broken metal	none	none	none

Table C.1: Summary of the properties of the correlated states in selected devices from Refs. [20; 21; 120; 113]. Entries reading “none” indicate that no correlated state is clearly observed or reported.

Chapter D

Appendix D

D.1 Band structure calculation of tMBG and the Q-IVC order

We calculate the band structure of tMBG using the standard continuum model. The Hamiltonian is

$$H = H_{MG} + H_{BG} + H_M \quad (\text{D.1})$$

We have

$$H_{MG} = \sum_{\mathbf{k}} (\tilde{c}_A^\dagger(\mathbf{k}), \tilde{c}_B^\dagger(\mathbf{k})) \begin{pmatrix} -\frac{\delta}{2} & -\frac{\sqrt{3}}{2}t(\tilde{k}_x - i\tilde{k}_y) \\ -\frac{\sqrt{3}}{2}t(\tilde{k}_x + i\tilde{k}_y) & -\frac{\delta}{2} \end{pmatrix} \begin{pmatrix} \tilde{c}_A(\mathbf{k}) \\ \tilde{c}_B(\mathbf{k}) \end{pmatrix} \quad (\text{D.2})$$

where $\tilde{\mathbf{k}} = R(-\theta/2)\mathbf{k}$, with twist angle θ . $R(\varphi)$ is the transformation matrix for anticlockwise rotation with angle φ .

The Hamiltonian for the bilayer graphene is

$$H_{BG} = \sum_{\mathbf{k}} \Psi^\dagger(\mathbf{k}) \begin{pmatrix} 0 & -\frac{\sqrt{3}}{2}t(\tilde{k}_x - i\tilde{k}_y) & -\frac{\sqrt{3}}{2}\gamma_4(\tilde{k}_x - i\tilde{k}_y) & -\frac{\sqrt{3}}{2}\gamma_3(\tilde{k}_x + i\tilde{k}_y) \\ -\frac{\sqrt{3}}{2}t(\tilde{k}_x + i\tilde{k}_y) & 0 & \gamma_1 & -\frac{\sqrt{3}}{2}\gamma_4(\tilde{k}_x - i\tilde{k}_y) \\ -\frac{\sqrt{3}}{2}\gamma_4(\tilde{k}_x + i\tilde{k}_y) & \gamma_1 & \frac{\delta}{2} & -\frac{\sqrt{3}}{2}t(\tilde{k}_x - i\tilde{k}_y) \\ -\frac{\sqrt{3}}{2}\gamma_3(\tilde{k}_x - i\tilde{k}_y) & -\frac{\sqrt{3}}{2}\gamma_4(\tilde{k}_x + i\tilde{k}_y) & -\frac{\sqrt{3}}{2}t(\tilde{k}_x + i\tilde{k}_y) & \frac{\delta}{2} \end{pmatrix} \Psi(\mathbf{k}) \quad (\text{D.3})$$

where $\tilde{\mathbf{k}} = R(\theta/2)\mathbf{k}$ and $\Psi^\dagger(\mathbf{k}) = (c_{A_1}^\dagger(\mathbf{k}), c_{B_1}^\dagger(\mathbf{k}), c_{A_2}^\dagger(\mathbf{k}), c_{B_2}^\dagger(\mathbf{k}))$.

Finally the interlayer moiré tunneling term is

$$H_M = \sum_{\mathbf{k}} \sum_{j=0,1,2} (\tilde{c}_A^\dagger(\mathbf{k}), \tilde{c}_B^\dagger(\mathbf{k})) \begin{pmatrix} \alpha t_M & t_M e^{-i\frac{2\pi}{3}j} \\ t_M e^{i\frac{2\pi}{3}j} & \alpha t_M \end{pmatrix} \begin{pmatrix} c_{A_1}(\mathbf{k} + \mathbf{Q}_j) \\ c_{B_1}(\mathbf{k} + \mathbf{Q}_j) \end{pmatrix} + h.c. \quad (\text{D.4})$$

where $\mathbf{Q}_0 = (0, 0)$, $\mathbf{Q}_1 = \frac{1}{a_M}(-\frac{2\pi}{\sqrt{3}}, -2\pi)$ and $\mathbf{Q}_2 = \frac{1}{a_M}(\frac{2\pi}{\sqrt{3}}, -2\pi)$, with a_M as the moiré lattice constant.

We use parameters $(t, \gamma_1, \gamma_3, \gamma_4) = (-2610, 361, 283, 140)$ meV. For the interlayer tunneling, we use $t_M = 110$ meV and $\alpha = 0.5$. δ is the potential difference, which is tuned by the displacement field, D .

The dispersion at $\delta = -40$ meV and $\delta = 40$ meV for twist angle $\theta = 1.16^\circ$ is shown in Fig. D.1. The valley Chern numbers, C_v , for the conduction band are 1 and 2, respectively. Note that we take a convention in which $\delta > 0$ corresponds to the electric field pointing from the monolayer to the bilayer graphene (similarly, this corresponds to $D > 0$).

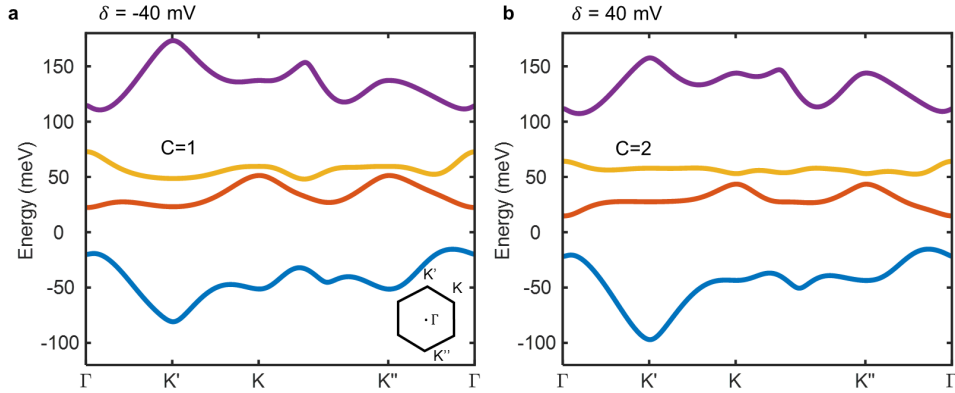


Figure D.1: Band structure of tMBG at twist angle $\theta = 1.16^\circ$. **a**, $\delta = -40$ meV and **b**, $\delta = 40$ meV. The valley Chern number of the lowest moiré conduction band (yellow) is $C_v = 1$ for $\delta = -40$ meV, and $C_v = 2$ for $\delta = 40$ meV. The inset in **a** shows the relevant high symmetry points in the first Brillouin zone.

D.1.1 Calculation of valley wave dispersion

For simplicity, we project the Hamiltonian to include only the conduction band. However, the other remote bands still play a role in renormalizing the dispersion of the active conduction band from the interaction:

$$H_V = \frac{1}{2} \frac{1}{N_s} \sum_{\mathbf{q}} V(\mathbf{q}) \sum_{\mathbf{k}_1} \sum_{\mathbf{k}_2} c_{a;m_1}^\dagger(\mathbf{k}_1 + \mathbf{q}) c_{b;n_1}^\dagger(\mathbf{k}_2 - \mathbf{q}) c_{b;n_2}(\mathbf{k}_2) c_{a;m_2}(\mathbf{k}_1) \lambda_{a;m_1 m_2}(\mathbf{k}_1, \mathbf{q}) \lambda_{b;n_1 n_2}(\mathbf{k}_2, -\mathbf{q}) \quad (\text{D.5})$$

where,

$$V(\mathbf{q}) = \frac{15\text{nm}}{a_M} \frac{1392\text{meV}}{\epsilon} \frac{1}{qa_M} \tanh(qr_0) \quad (\text{D.6})$$

Here r_0 is the screening length from the metallic contact, which we use $r_0 = 5a_M$. In the above $\lambda_{a;mn}(\mathbf{k}, \mathbf{q}) = \langle \mu_{a;m}(\mathbf{k} + \mathbf{q}) | \mu_{b;n}(\mathbf{k}) \rangle$ is the form factor. Here $a, b = +, -$ is the valley index and m, n is the band index.

Let us fully fill all of the bands below the conduction band. Then the kinetic term of the conduction band is renormalized:

$$H_K = H_K^0 + \sum_{\mathbf{k}} \xi_H(\mathbf{k}) c^\dagger(\mathbf{k}) c(\mathbf{k}) + \sum_{\mathbf{k}} \xi_F(\mathbf{k}) c^\dagger(\mathbf{k}) c(\mathbf{k}) \quad (\text{D.7})$$

where c is the operator for the valley $+$ of the conduction band. ϵ_H is from the Hartree term and ϵ_F is from the Fock term.

We have

$$\xi_H(\mathbf{k}) = \sum_{\mathbf{G}_M} V(\mathbf{G}_M) \rho(\mathbf{G}_M) \lambda_+(\mathbf{k}, \mathbf{G}_M) \quad (\text{D.8})$$

where,

$$\rho(\mathbf{G}_M) = 4 \frac{1}{N_s} \sum_{m \in O} \lambda_{a;mm}(\mathbf{k}, \mathbf{G}_M) \quad (\text{D.9})$$

Here the factor 4 comes from spin and valley degeneracy. O is the set of occupied bands.

The Fock term is

$$\xi_F(\mathbf{k}) = -\frac{1}{N_s} \sum_{\mathbf{q}} V(\mathbf{q}) \sum_{m \in O} |\lambda_{a;mc}(\mathbf{k}, \mathbf{q})|^2 \quad (\text{D.10})$$

where $m \in O$ is the index of the occupied band. c is the index of the conduction band.

Next, we calculate the valley wave assuming that the ground state is spin-valley polarized at $\nu = 1$. Without loss of generality, we assume the ground state fully occupies $+, \downarrow$. A collective excitation with momentum $\mathbf{q}$ is generated by a particle-hole boson $b_{ab;\mathbf{q}}(\mathbf{k})^\dagger = c_{a;\uparrow}^\dagger(\mathbf{k} + \mathbf{q})c_{b;\downarrow}(\mathbf{k})$, where $a, b = +, -$ labels the valley. Due to the $SU(2) \times SU(2)$ symmetry, the dispersion of the exciton corresponding to $c_{+;\sigma}^\dagger(\mathbf{k} + \mathbf{q})c_{-;\sigma'}(\mathbf{k})$ does not depend on the spin index $\sigma\sigma'$. In the following we will only consider the spin-singlet valley-flip exciton.

For a fixed momentum $\mathbf{q}$, we can derive Hamiltonian for $b_{ab;\mathbf{q}}(\mathbf{k})$:

$$H_{ab}(\mathbf{q}) = H_{ab}^V(\mathbf{q}) + H_{ab}^K(\mathbf{q}) \quad (\text{D.11})$$

where,

$$\begin{aligned} H_{ab}^V(\mathbf{q}) = & - \sum_{\mathbf{k} \in MBZ} \sum_{\tilde{\mathbf{q}}} V(\tilde{\mathbf{q}}) (\lambda_a(\mathbf{k} + \mathbf{q}, \tilde{\mathbf{q}}) \lambda_b(\mathbf{k} + \tilde{\mathbf{q}}, -\tilde{\mathbf{q}}) b_{ab;\mathbf{q}}^\dagger(\mathbf{k} + \tilde{\mathbf{q}}) b_{ab;\mathbf{q}}(\mathbf{k}) \\ & + \sum_{\tilde{\mathbf{q}}} V(\tilde{\mathbf{q}}) \lambda_b(\mathbf{k}, \tilde{\mathbf{q}}) \lambda_b(\mathbf{k} + \tilde{\mathbf{q}}, -\tilde{\mathbf{q}}) b_{ab;\mathbf{q}}^\dagger(\mathbf{k}) b_{ab;\mathbf{q}}(\mathbf{k}) \end{aligned} \quad (\text{D.12})$$

and

$$H_{ab}^K(\mathbf{q}) = \sum_{\mathbf{k} \in MBZ} (\xi_a(\mathbf{k} + \mathbf{q}) - \xi_b(\mathbf{k})) b_{ab;\mathbf{q}}^\dagger(\mathbf{k}) b_{ab;\mathbf{q}}(\mathbf{k}) \quad (\text{D.13})$$

where ξ_a is the dispersion after including the renormalization from remote bands.

For a fixed momentum $\mathbf{q}$, the ground state of $H_{ab}(\mathbf{q})$ gives the energy of the excitation, $\omega_{ab}(\mathbf{q})$. Here $\omega_{++}(\mathbf{q})$ is the spin wave excitation and $\omega_{+-}(\mathbf{q})$ is the spin-valley wave excitation.

We focus on the intervalley exciton $\omega_{+-}(\mathbf{q})$. As shown in Figure 3.12a, when we increase the dielectric constant ϵ , the minimum energy of the exciton becomes negative at momentum $\mathbf{q} = K'$ in the mini Brillouin Zone (MBZ), suggesting an instability of the polarized ground state towards an intervalley coherent (IVC) state. As the magnon dispersion has its minimum at a non-zero momentum, the resulting IVC order must also carry a non-zero momentum, and is distinct from the IVC previously proposed in tBLG [198]. We denote this as a “Q-IVC” to emphasize the non-zero momentum $\mathbf{Q}$.

D.1.2 Symmetry analysis of Q-IVC order

As discussed above, the valley polarized ground state is unstable because the energy of the valley magnon (or exciton) becomes negative when we increase the dielectric constant ϵ . The condensation of valley magnons will lead to an IVC state. Here we perform a general symmetry analysis of the possible IVC states. Given that the minimum of valley magnon dispersion is generically at non-zero momentum $\mathbf{Q}$, the resulting IVC carries momentum $\mathbf{Q}$.

We consider a general IVC ansatz described by a mean field theory:

$$H_M = - \sum_{\mathbf{k}} [F(\mathbf{k})c_+^\dagger(\mathbf{k} + \frac{1}{2}\mathbf{Q})c_-(\mathbf{k} - \frac{1}{2}\mathbf{Q}) + F^*(\mathbf{k})c_-^\dagger(\mathbf{k} - \frac{1}{2}\mathbf{Q})c_+(\mathbf{k} + \frac{1}{2}\mathbf{Q})] \quad (\text{D.14})$$

where $F(\mathbf{k})$ represents the profile of the intervalley exciton, similar to $\Delta(\mathbf{k})$ in Cooper pairing. Here we assume that the exciton is condensed at momentum $\mathbf{Q}$, which may not be zero.

Using $Tc_+(\mathbf{k})T^{-1} = c_-(-\mathbf{k})$ and $Tc_-(\mathbf{k})T^{-1} = c_+(-\mathbf{k})$, we find:

$$TH_MT^{-1} = - \sum_{\mathbf{k}} [F^*(\mathbf{k})c_-^\dagger(-\mathbf{k} - \frac{1}{2}\mathbf{Q})c_+(-\mathbf{k} + \frac{1}{2}\mathbf{Q}) + F(\mathbf{k})c_+^\dagger(-\mathbf{k} + \frac{1}{2}\mathbf{Q})c_-(-\mathbf{k} - \frac{1}{2}\mathbf{Q})] \quad (\text{D.15})$$

Then the time reversal symmetry $H_M = TH_MT^{-1}$ requires

$$F(\mathbf{k}) = F(-\mathbf{k}) \quad (\text{D.16})$$

For an IVC state in the case of non-zero C_v , $F(\mathbf{k})$ needs to acquire the Berry phase of the two bands with opposite signs of the Chern number. A general argument shows that the complex field $F(\mathbf{k})$ cannot be a constant in momentum space, and should have total vorticity equal to $2C_v$. As a result, one usually finds that $F(\mathbf{k}) \neq F(-\mathbf{k})$. This is reflected in our direct calculation of the intervalley exciton wavefunction on top of the spin-valley polarized ground state (see the inset of Figure 3.12a). As a consequence, the Q-IVC order breaks the time reversal symmetry. Note that there is always a degenerate IVC state with $\tilde{F}(\mathbf{k}) = F(-\mathbf{k})$ as the time reversal partner of the IVC with $F(\mathbf{k})$. These two IVC states are degenerate at zero magnetic field, but split for $|B| > 0$. This degeneracy can also be broken spontaneously at $B = 0$.

Let us also comment on the translation symmetry for the Q-IVC order. If only the IVC corresponding to

one momentum $\mathbf{Q}$ is condensed, the resulting phase still preserves the moiré translation symmetry because we can apply a global phase generated by the valley charge. We call the IVC in this class a uniform Q-IVC. Note that the non-zero $\mathbf{Q}$ can be gauged away by applying a constant gauge field $\mathbf{A}\tau_z$, where τ_z is the Pauli matrix in valley space and $\mathbf{A} = -\mathbf{Q}$. Theoretically the Q-IVC order can hence be treated in the same way as the usual IVC with $\mathbf{Q} = 0$, and a gap will be opened if the Q-IVC order is sufficiently strong at integer filling. Microscopically, however, the momentum $\mathbf{Q}$ is still meaningful when considering the density profile at the atomic scale: $\rho(\mathbf{r}) = \Psi_K^\dagger(\mathbf{r})\psi_K(\mathbf{r}) + \Psi_{K'}^\dagger(\mathbf{r})\psi_{K'}(\mathbf{r}) + (\Psi_K^\dagger(\mathbf{r})\psi_{K'}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}} + h.c.)$, where $\Psi_{K/K'}(\mathbf{r})$ is the electron operator for the valley K and K' respectively. If there is a Q-IVC order, $\langle \psi_K^\dagger(\mathbf{r})\psi_{K'}(\mathbf{r}) \rangle \sim e^{i\mathbf{Q}\cdot\mathbf{r}}$, and as a result the density profile will have a component $\rho(\mathbf{r}) = \rho_0 + A \cos((\mathbf{K} + \mathbf{Q}) \cdot \mathbf{r})$. Such a density profile with momentum $\mathbf{K} + \mathbf{Q}$ can in principle be observed with scanning tunneling microscopy. Note here that $|\mathbf{Q}| \sim \frac{4\pi}{3a_M}$ where a_M is the moiré lattice constant. On the other hand $|\mathbf{K}| = \frac{4\pi}{3a}$ is much larger given that a is the original graphene lattice constant. Therefore the momentum $\mathbf{K} + \mathbf{Q}$ of the density profile is only slightly different from the Kekulé pattern with momentum $\mathbf{K}$.

However, if the IVC with at least two distinct momenta $\mathbf{Q}_1, \mathbf{Q}_2$ condenses, the final state breaks the moiré translation symmetry and there is a charge density wave order (CDW) at momentum $\mathbf{Q}_1 - \mathbf{Q}_2$. If there is a C_3 symmetry, the valley magnon dispersion should have minima at three $\mathbf{Q}$ related by the C_3 rotation. However, when there is strain, C_3 is broken and one momentum will be selected and the final phase is likely to be the uniform Q-IVC order with only one momentum $\mathbf{Q}$.

Chapter E

Appendix E

E.1 Selection rules for intervalley transitions in monolayer WSe₂

The character table of D_{3h} point double-group is used to derive selection rules for phonon-mediated transitions of intravalley excitons, corresponding to the case that both electron and hole reside in the same valley (e.g., both in the K-point valley) [171]. The transformation properties of optically-inactive dark excitons are captured by the irreducible representation (IR) Γ_3 , semi-dark excitons with out-of-plane optical transition dipole by Γ_4 , and bright excitons with in-plane optical transition dipole by Γ_6 [172]. One can readily verify the selection rule,

$$(\Gamma_6^* \times \Gamma_3)^* = (\Gamma_6^* \times \Gamma_4)^* = \Gamma_5 \quad (\text{E.1})$$

implying that transitions between dark and bright excitons due to intravalley spin flip (of the electron component) can be induced by a zone-center phonon that transforms like the IR Γ_5 . This phonon is the homopolar optical mode, whose polarization vector is denoted by in-plane and out-of-phase vibration of the chalcogen atoms, as shown in Fig. E.1a [172]. Note that the spin-flip matrix element due to interaction with a long-wavelength flexural phonon, which transforms as Γ_4 , is nonzero only if the phonon wavevector is finite (i.e., when $q \neq 0$) [171]. On the other hand, the phonon mode Γ_5 is the only one for which the spin-flip matrix element is nonzero exactly at the zone center ($q = 0$).

Using Quantum Espresso, Fig. E.1b shows the calculated phonon spectrum in monolayer WSe₂. Because the concept of acoustic and optical phonon modes near the Γ -point loses its meaning when qa is no

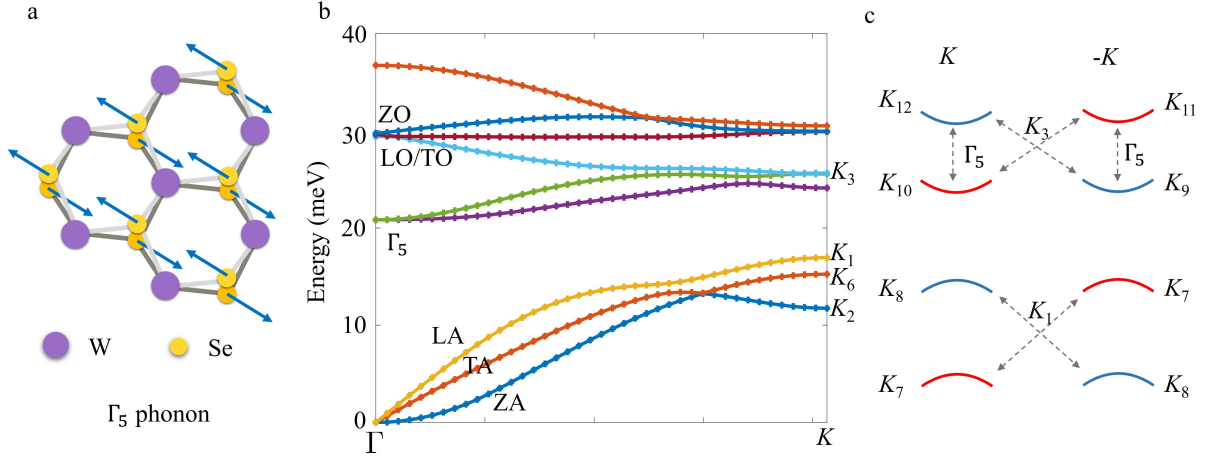


Figure E.1: Phonon modes and selection rules. **a**, Atomic displacement of the phonon mode Γ_5 . **b**, The phonon dispersion of monolayer WSe₂ along the axis between the high-symmetry Γ and K points. Zone-center and pertinent zone-edge phonon modes are indicated. **c**, Scheme of low-energy valleys in the conduction and valence bands. Spin-flip intravalley transitions in the conduction band are mediated by the phonon-mode Γ_5 , while spin-conserving intervalley transitions in the conduction (valence) bands are mediated by the phonon mode K_3 (K_1). Phonon-induced intervalley spin-flip scattering is relatively weak (the transition matrix element between time-reversed states vanishes).

longer much smaller than unity (a is the lattice constant), the notion of acoustic or optical phonons for longitudinal and transverse modes (i.e., LA, TA, LO and TO) is not valid when dealing with zone-edge phonons at the K -point whose wavenumber is $q = K = \frac{4\pi}{3a}$. Instead, the use of K -point IRs is more appropriate. This nomenclature is presented in the Character table of C_{3h} point double-group, where the transformation properties of the zone-edge phonons are captured by the IRs K_1 through K_6 (these IRs also belong to the C_{3h} point single group. The spin quantum number has no bearing on the polarization vectors of atoms in the unit cell) [171].

Supported by our experimental findings and the group theory analysis below, we will focus in this work on the modes K_1 and K_3 . Figure E.2a shows the atomic displacements that correspond to low- and high-energy phonon modes with symmetry K_1 . The difference between the two is the phase difference in the circular motion of Se and W atoms, leading to mitigated deviation of the bond lengths from equilibrium in the low energy case. Figure E.2b shows the atomic displacement that corresponds to a phonon mode with symmetry K_3 . Here, only Se atoms go through in-plane circular motion around their equilibrium positions and the phonon energy is somewhere between that of the low- and high-energy phonon modes

with symmetry K_1 .

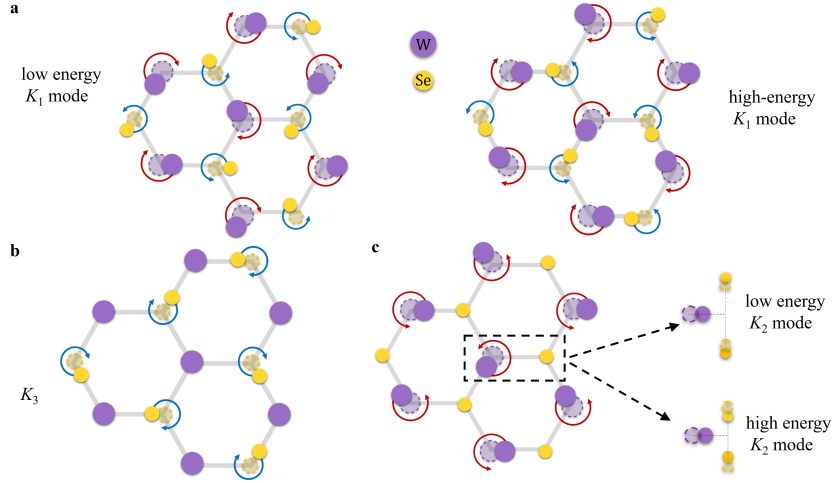


Figure E.2: Atomic motions of Valley phonons. The curved arrows denote the in-plane circular motion of atoms around their equilibrium positions, denoted by faint circles. See text for further explanations.

Fig. E.2c shows the atomic displacements that correspond to low- and high- energy phonon modes with symmetry K_2 . Here, the transition-metal atoms go through in-plane circular motion around their equilibrium positions, while chalcogen atoms vibrate in opposite directions along the out-of-plane axis. As shown in the figure, the difference between the low and high energy modes is the relative motion of the chalcogen and transition-metal atoms. Namely, the low energy mode corresponds to the case that when the transition-metal atoms move toward the chalcogen atoms, the chalcogen atoms move away from the mid-plane and vice versa. This combined motion keeps the bond length closer to its equilibrium value, and hence to lower phonon energy. Conversely, the high energy mode leads to stronger deviations from equilibrium because when the transition-metal atoms move toward the chalcogen atoms, the chalcogen atoms move closer to the mid-plane and vice versa.

Finally, Fig. E.1b shows that the branch extensions of the ZA and TA modes anti-cross close to the K-point. This anti-crossing is not seen in the calculation of monolayer MoS₂. The result is that the mode K_6 has the lowest energy in monolayer MoS₂, while K_2 has the lowest energy in monolayer WeS₂.

Next, we derive selection rules for intervalley transitions of electrons and holes. We first note that $K = 4\pi/3a$ is not only the wavenumber that connects each of the zone-edge $\pm K$ -points with the zone center, but it is also the wavenumber needed to connect the zone-edge $+K$ and $-K$ points. In other words,

the conservation of crystal momentum due to intervalley transitions of electrons (or holes) states at the $\pm K$ points is mediated through the zone-edge K -point phonons. We use the Character table of C_{3h} point double-group to find the transformation properties of the zone-edge electronic states along with the ensuing selection rules for phonon-induced intervalley transitions of electrons and holes¹¹. Starting with holes states, the transformation properties of their wavefunctions at the valence-band edge states are captured by the IRs K_7 and K_8 . As shown in Fig. E.1c, the band-edge hole state at the top $-K$ ($+K$) valley transforms like the IR K_7 (K_8), and vice versa for the bottom valleys. This behavior is a consequence of time-reversal symmetry and the complex conjugation of the IRs K_7 and K_8 . Accordingly, a spin conserving intervalley scattering between the edge states involves the selection rule [171]

$$(\Gamma_7^* \times \Gamma_7)^* = (\Gamma_8^* \times \Gamma_8)^* = K_1 \quad (\text{E.2})$$

where the first term corresponds to hole transition between the top valley in $-K$ and the bottom one in $+K$, while the second term corresponds to hole transition between the bottom valley in $-K$ and the top one in $+K$. This selection rule implies that the phonon mode K_1 is the dominant mechanism for intervalley transitions in the valence band.

The case of conduction-band electrons is somewhat more subtle. As shown in Fig. E.1c, the band-edge electron state at the bottom (top) $-K$ valley transforms like K_9 (K_{11}), while the respective complex conjugate IRs at the $+K$ valleys transforms like K_{10} (K_{12}). The selection rule in this case reads [171]

$$(\Gamma_{12}^* \times \Gamma_9)^* = (\Gamma_{10}^* \times \Gamma_{11})^* = K_3 \quad (\text{E.3})$$

implying that the phonon mode K_3 is the dominant mechanism for intervalley transitions in the conduction band.