

Unconventional Superconductivity in Semimetallic Rhombohedral Graphene

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Abstract

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Rhombohedral graphene, a metastable stacking configuration of graphene, provides a clean platform for the study of correlated electron physics, hosting a rich set of symmetry-broken phases. Its characteristic staggered structure produces a large low-energy density of states, with wavefunctions predominantly localized on the outer crystal surfaces. This surface-localized density of states is further enhanced with increasing layer number and displacement field, providing a natural route to strong electronic correlations.

Against this backdrop, the first part of this thesis explores the semimetallic state of rhombohedral graphene, in which valence and conduction band Fermi pockets reside on opposite crystal surfaces. Within this charge-delocalized regime, we identify multiple superconducting phases that appear at approximately constant conduction band filling. The most prominent of these phases expands dramatically with in-plane magnetic field, spanning a wide region of the phase diagram. It persists to fields far beyond the Pauli limit, consistent with spin-triplet superconductivity [1].

In addition to electrostatic control, hydrostatic pressure provides a complementary tuning parameter by directly modifying interlayer coupling and the resulting electronic structure. The second part of this thesis presents a robust approach for high pressure transport measurements of two-dimensional heterostructures using a piston-cylinder cell. This is accomplished by replacing failure-prone wiring schemes with a compact and scalable design. Preliminary measurements on rhombohedral graphene show clear signatures of band modification with pressure.

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GLOSSARY

BBG: Bilayer Bernal graphene.	IQAH: Integer quantum anomalous Hall state.
BCS: Bardeen–Cooper–Schrieffer theory of superconductivity.	ISOSPIN: Internal degree of freedom associated with spin, and valley.
BLG: Bilayer graphene.	LANDAU FAN: Evolution of electronic states with magnetic field, used to determine filling factors, degeneracies, and symmetry breaking.
CHERN NUMBER: Topological invariant characterizing a quantum Hall or quantum anomalous Hall state.	LAF: Layer antiferromagnet.
CDW: Charge density wave.	MLG: Monolayer graphene.
CNP: Charge neutrality point.	MOIRÉ SUPERLATTICE: Long-wavelength interference pattern formed by a twist angle or lattice mismatch between two crystalline layers.
DOS: Density of states.	QUANTUM ANOMALOUS HALL STATE: Quantized Hall state arising from spontaneous time-reversal symmetry breaking in the absence of an external magnetic field.
FQAH: Fractional quantum anomalous Hall state.	
HALF-METAL: State in which only one spin or isospin species contributes to conduction at the Fermi level.	
HARTREE–FOCK: Mean-field approximation that includes exchange interactions self-consistently.	

RXG: X layer Rhombohedral graphene.
Example R5G is 5-layer
rhombohedral graphene.

SC: Superconducting or
superconductivity.

SOC: Spin-orbit coupling.

SQUID: Superconducting quantum
interference device.

SHG: Second harmonic generation.

SPIN POLARIZATION: Unequal
occupation of electronic states with
opposite spin.

SYMMETRY BREAKING: Spontaneous
selection of one state from a set of
symmetry-related states.

TLG: Trilayer graphene.

TMD: Transition metal dichalcogenide.

VALLEY: Quantum number associated
with inequivalent extrema of the
electronic band-structure, typically K
and K' in graphene.

VALLEY POLARIZATION: Unequal
occupation of different valleys in
momentum space.

$B_{\parallel}$: Magnetic field applied parallel to
the graphene plane.

$B_{\perp}$: Magnetic field applied
perpendicular to the graphene plane.

B_C : Critical magnetic field.

D : Electric displacement field applied
across the graphene multilayer.

I_{DC} : Direct-current bias current.

N : Carrier density.

T_C : Superconducting critical
temperature.

V_B : Bottom-gate voltage.

V_T : Top-gate voltage.

ρ_{XX} : Longitudinal resistivity.

ρ_{XY} : Hall resistivity.

VDW: van der Waals

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DEDICATION

To the people I lost along this journey, forever missed, never forgotten

Chapter 1

INTRODUCTION

1.1 Motivation

The concept of emergence underlies many of the most interesting phenomena in condensed matter physics, including superconductivity, magnetism, and the fractional quantum Hall effect. They are born out of the collective and cannot be fully predicted from the fundamental laws governing their constituents. Philip Anderson captured this spirit best in his seminal 1972 essay, *More is Different* [2], where he argued that at each level of complexity, genuinely new laws and degrees of freedom emerge, that are beyond simple extrapolations from the level below. Superconductivity stands as perhaps the most striking example of this: below a critical temperature, electrons abandon their individual identities and condense into a single macroscopic quantum state capable of carrying current without dissipation, a property no individual electron possesses.

What drives this remarkable collective behavior? The answer, in most cases, is electronic interactions. When electrons interact strongly with one another, they can organize their motion in ways that no single-particle picture can capture, and it is precisely this organized motion that gives rise to the richest phases of matter.

The central challenge, however, is that nature conspires against strong interactions. Disorder presents a fundamental obstacle. In any real material, impurities, defects, and lattice irregularities scatter electrons and localize their wavefunctions, suppressing the coherent collective behavior that interactions might otherwise produce. Even in a perfectly clean system, interactions face a second adversary: the kinetic energy. In most metals, electrons are fast and itinerant, and the Fermi energy is large compared to the Coulomb interaction, smothering any interaction-induced phenomena.

The search for platforms where these obstacles can be overcome has been a central theme of condensed matter physics for decades. A particularly productive direction has been the confinement of electrons to two dimensions (2D). Reduced dimensionality weakens the dilution of interactions and fundamentally alters collective behavior. Early

realizations of this came in electrostatically confined 2D electron gases in silicon MOSFETs and GaAs/AlGaAs quantum wells, culminating in the discovery of the fractional quantum Hall effect [3]. The isolation of graphene by Geim and Novoselov in 2004 opened a new chapter [4] for 2D physics, making truly atomically thin materials experimentally accessible and sparking a broad 2D materials revolution.

Within this 2D landscape, band flattening via rotational misalignment of constituent layers has emerged as a powerful technique for accessing the strongly correlated regime [5]. By controlling the twist angle, the electronic bandwidth can be continuously tuned, making the ratio $U/W \gg 1$, where U is the Coulomb interaction and W is the bandwidth. The discovery that twisted bilayer graphene at the magic angle hosts both correlated insulators and an unconventional superconductor was the first demonstration of this approach [6], establishing moiré graphene as a tunable platform for correlated physics and sparking a broad search across other material combinations. More recently, twisted transition metal dichalcogenides such as MoTe_2 [7, 8] and WSe_2 [9, 10] have also proven particularly fertile, hosting superconductivity, Wigner crystals, and fractional quantum Hall physics at zero magnetic field.

Rhombohedral graphene is a compelling addition to this landscape. Graphene in this metastable stacking configuration has flat bands that arise intrinsically from the stacking order itself [11], freeing the system from the twist-angle disorder endemic to moiré bilayer systems [12]. Moreover, the electronic wavefunctions are localized to the outer crystal surfaces as a consequence of the staggered interlayer geometry [13, 11]. This layer polarization, combined with the large density of states (DOS) and tunability of the bands with displacement field, makes rhombohedral graphene a particularly versatile platform to study interaction-induced emergent phases.

Prior studies have focused largely on the high displacement field (D) regime, where the DOS is highest. In this regime, remarkable interaction-driven phenomena have been uncovered, including superconductivity that exhibits signatures of macroscopic time-reversal-symmetry (TRS) breaking [14, 15] and fractional Chern insulators (FCIs) [16, 17]. However, in thicker rhombohedral graphene, the conduction and valence bands overlap over a wide range of D [18]. In this semimetallic regime, the coexisting electron and hole carriers are polarized to opposite crystal surfaces, forming a qualitatively distinct ground state, while retaining a large density of states. Despite hosting this

unusual dual-surface character, this regime has received comparatively little attention. A central goal of this thesis is to investigate this regime and report our key finding: the observation of superconductivity emerging from a normal state that is both semimetallic and dual-surface polarized.

Beyond electrostatic control, hydrostatic pressure offers an independent handle on U/W : by reducing the interlayer spacing, it directly tunes the interlayer hopping and as a result allows us to reduce W [19, 20, 21], while keeping disorder and sample geometry fixed across measurements. Rhombohedral graphene, with its intrinsic flat band and rich correlated phase diagram, is a natural target for this approach. Moreover, the ability to study the same device at different pressures opens the door to well-controlled experiments where the effects of additional perturbations, such as a moiré potential from hBN alignment or proximity to a transition metal dichalcogenide (TMD), can be systematically isolated. Despite its potential, high pressure transport measurements on atomically thin materials remain technically challenging. In this thesis, we demonstrate a reliable experimental technique for hydrostatic pressure transport of 2D heterostructures and present preliminary results of its application to rhombohedral graphene.

1.2 *Outline*

The remainder of this chapter provides background on rhombohedral graphene, including details of its band-structure and basic phenomenology, intended to provide context for the results that follow rather than serve as an exhaustive review.

Chapter 2 presents our exploration of the charge-delocalized semimetallic regime of rhombohedral graphene. We map out the extent of this regime in phase space with the aid of self-consistent Hartree-Fock calculations. In this regime, we report the observation of multiple superconducting phases in a R8G (8-layer rhombohedral graphene) and moiré R7G (7-layer rhombohedral graphene).

Chapter 3 examines the spin structure of these superconducting phases. We see a striking expansion of the superconducting state with in-plane field ($B_{||}$) and present evidence that several of these states are likely spin polarized. We discuss how microscopic interactions such as Hund's coupling and Kane-Mele spin-orbit coupling may contribute to the observed superconductivity.

Chapter 4 details our development of a reliable experimental technique for hydrostatic pressure transport in atomically thin heterostructures. We discuss the technical challenges involved, the improvements we have introduced to address them, and present preliminary results of its application to rhombohedral graphene.

Chapter 5 summarizes the conclusions of this thesis, discusses open questions that remain, and reflects on the broader implications of this work for the study of correlated phases in two-dimensional materials.

1.3 *Band-Structure of Multilayer Rhombohedral Graphene*

The distinctive electronic band-structure of rhombohedral graphene underlies much of its unusual phenomenology. The $E \sim q^N$ dispersion (where E is the energy, q is crystal momentum relative to the K point and N is the layer number) and large density of states at the band edge are at the root of all emergent phenomena in rhombohedral graphene. In this section, we will illustrate how the stacking configuration of graphene sheets leads to this kind of band-structure. For ease of understanding, we begin with monolayer graphene and gradually add layers and introduce more tight binding hopping terms, with the consequences of stacking order becoming apparent at the trilayer level. Throughout, we focus on the key concepts that carry through to the multilayer rhombohedral case rather than detailed derivations, for which we refer the reader to [22, 23, 24, 11].

1.3.1 *Monolayer Graphene*

Monolayer graphene (MLG) consists of carbon atoms arranged on a honeycomb lattice (Fig. 1.1a). Each carbon atom contributes four valence electrons, one each in the $2s$, $2p_x$, $2p_y$ and $2p_z$ orbitals. The electrons in the $2s$, $2p_x$, $2p_y$ form hybridized sp^2 orbitals that participate in covalent bonding. The $2p_z$ electron remains itinerant, largely determining the low energy band structure of graphene. Since the hexagonal lattice is not a Bravais lattice (i.e., cannot be generated by translating a single point), it can be decomposed into two triangular sublattices A and B with lattice vectors:

$$\mathbf{a}_1 = \frac{a}{2}(3\hat{x} + \sqrt{3}\hat{y}), \quad \mathbf{a}_2 = \frac{a}{2}(3\hat{x} - \sqrt{3}\hat{y}) \quad (1.1)$$

where $a \approx 1.42\text{\AA}$ is the carbon-carbon distance. The reciprocal lattice vectors (Fig. 1.1b) are given by:

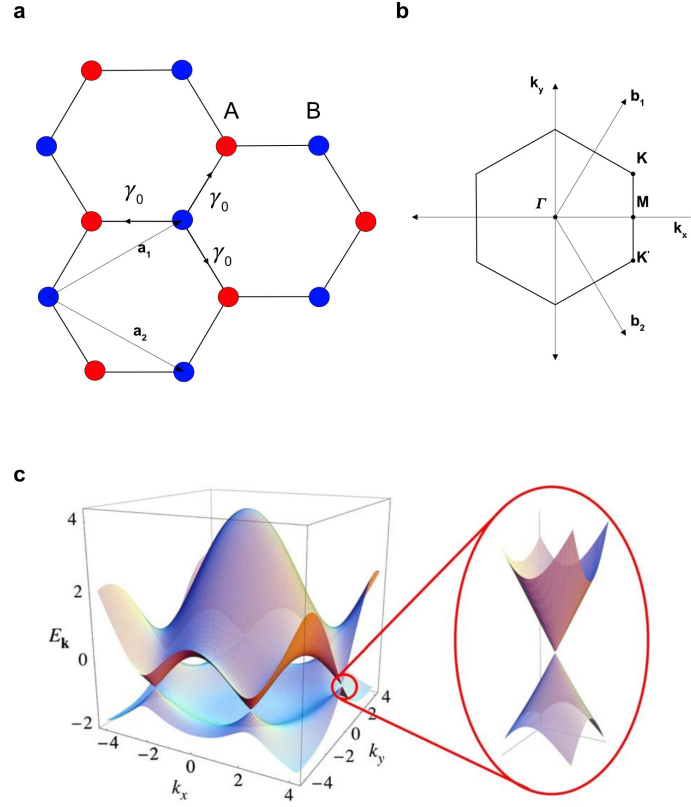


Figure 1.1: **MLG lattice and band-structure.** **a**, Lattice structure of monolayer graphene. The two triangular sublattices A and B are denoted by red and blue circles. Sublattice vectors a_1 and a_2 are labeled. Tight binding parameter γ_0 connects nearest neighbors from two different sublattices. **b**, reciprocal lattice of MLG showing the first Brillouin zone. High symmetry points Γ , M and K points are labeled. Reciprocal lattice vectors are labeled as b_1 and b_2 . **c**, Electronic dispersion of MLG, plotted as E vs k_x - k_y plane. Inset shows the Dirac like electronic structure around $E_{\mathbf{k}} = 0$ at the K point. (c) has been reproduced from [22] (Fig. 3)

$$\mathbf{b}_1 = \frac{2\pi}{3a}(\hat{k}_x + \sqrt{3}\hat{k}_y), \quad \mathbf{b}_2 = \frac{2\pi}{3a}(\hat{k}_x - \sqrt{3}\hat{k}_y) \quad (1.2)$$

The K and K' points on the corners of the hexagonal Brillouin zone (Fig. 1.1b) are of utmost importance to graphene physics and are designated as Dirac points for reasons that will be clear below. Their positions in momentum space are given by:

$$\mathbf{K} = \frac{2\pi}{3a}(\hat{k}_x + \frac{\hat{k}_y}{\sqrt{3}}), \quad \mathbf{K}' = \frac{2\pi}{3a}(\hat{k}_x - \frac{\hat{k}_y}{\sqrt{3}}) \quad (1.3)$$

The simplest Hamiltonian we can write is by considering only the nearest neighbor hopping γ_0 between A and B sublattices. This tightbinding Hamiltonian in the Bloch bases centered on the two sublattices $\{A, B\}$, takes the form:

$$H(\mathbf{k}) = \begin{pmatrix} 0 & \gamma_0 f(\mathbf{k}) \\ \gamma_0 f^*(\mathbf{k}) & 0 \end{pmatrix} \quad (1.4)$$

where the structure factor $f(\mathbf{k})$ is the Fourier transform of the real space hopping amplitudes:

$$f(\mathbf{k}) = \sum_{j=1}^3 e^{i\mathbf{k} \cdot \delta_j} = e^{ik_x a / \sqrt{3}} \left(1 + 2e^{-i\frac{k_y a}{2\sqrt{3}}} \cos\left(\frac{k_y a}{2}\right) \right) \quad (1.5)$$

The eigenvalues of this Hamiltonian are given as $E_{\pm}(\mathbf{k}) = \pm\gamma_0|f(\mathbf{k})|$, two bands that touch at zero energy at the corners of the hexagonal Brillouin zone (as shown in Fig. 1.1c) the inequivalent K and K' points, where $f(\mathbf{K}) = 0$.

Since the low energy dispersion is only relevant at the K points, we can expand $f(\mathbf{k})$ to linear order in momentum $\mathbf{q} = \mathbf{k} - \mathbf{K}$ measured relative to the K point:

$$f(\mathbf{K} + \mathbf{q}) \approx \frac{\sqrt{3}a}{2}(q_x + iq_y) = \frac{\sqrt{3}a}{2}|\mathbf{q}|e^{i\theta_{\mathbf{q}}} \quad (1.6)$$

where $\theta_{\mathbf{q}}$ is the angle of $\mathbf{q}$ with respect to the k_x axis. The Hamiltonian then reduces to the two-dimensional massless Dirac equation:

$$H(\mathbf{q}) = \hbar v_F \begin{pmatrix} 0 & q_x - iq_y \\ q_x + iq_y & 0 \end{pmatrix} = \hbar v_F \boldsymbol{\sigma} \cdot \mathbf{q} \quad (1.7)$$

where $v_F = \frac{\sqrt{3}\gamma_0 a}{2\hbar} \approx 10^6$ m/s is the Fermi velocity and $\sigma = (\sigma_x, \sigma_y)$ are the Pauli matrices acting on the sublattice polarization (also referred to as pseudospin) degree of freedom.

This reveals two important features of the low-energy electronic structure of monolayer graphene. First, the dispersion is linear near the K and K' points (Fig. 1.1c inset), in stark contrast to the parabolic dispersion of a conventional two-dimensional electron gas. Second, the eigenstates are chiral: the pseudospin is locked to the direction of the momentum, and the wavefunction acquires a Berry phase of π upon encircling the Dirac point. This chirality is an intrinsic consequence of the two-sublattice structure of the honeycomb lattice and is the fundamental property that distinguishes graphene from other two-dimensional conductors. The pseudospin winding has opposite chirality around the K and K' points, as a result these two are connected by TRS. As a result, in addition to the two-fold spin degeneracy, electrons in graphene have two-fold "valley degeneracy". Together these will be referred to as isospin. Including next-nearest-neighbor hopping does not qualitatively alter either of these features: the dispersion remains linear near the K points and the chiral structure is preserved, though the particle-hole symmetry of the band structure is broken and the energy at which the two bands touch is shifted away from zero.¹

1.3.2 Bilayer Graphene

Next we can look at what happens when we have two sheets of graphene stacked on top of each other. Naturally occurring bilayers usually exist in the energetically favorable Bernal (AB) configuration where B_1 site on the upper layer sits directly above the A_2 site on the lower layer (Fig. 1.2b). The unit cell now consists of four carbon atoms A_1, B_1 on the lower layer and A_2, B_2 on the upper layer (Fig. 1.2a). The intralayer hopping is still mediated by γ_0 whereas the interlayer hopping between B_1 and A_2 is given by γ_1 . These vertically aligned B_1 and A_2 sites form a strongly hybridized "dimer," whose bonding and antibonding combinations are split away from zero energy, leaving the low-energy physics dominated by the remaining non-dimer sites A_1 and B_2 . Other higher order hopping terms γ_3 and γ_4 are also possible, but will not be considered in this section. Their

¹See Ref. [22] for a detailed derivation.

effects on the electronic band structure will be discussed separately in Section 1.3.5.

Retaining only the dominant intralayer hopping γ_0 and interlayer dimer hopping γ_1 , and writing the Hamiltonian in the basis $\{A_1, B_1, A_2, B_2\}$, the tight-binding Hamiltonian near the K point takes the form:

$$H = \begin{pmatrix} 0 & \gamma_0 f(\mathbf{k}) & 0 & 0 \\ \gamma_0 f^*(\mathbf{k}) & 0 & \gamma_1 & 0 \\ 0 & \gamma_1 & 0 & \gamma_0 f(\mathbf{k}) \\ 0 & 0 & \gamma_0 f^*(\mathbf{k}) & 0 \end{pmatrix} \quad (1.8)$$

The dimer sites B_1 and A_2 , coupled by γ_1 , split off to high energy and can be integrated out. Projecting onto the low-energy non-dimer subspace $\{A_1, B_2\}$ and expanding to leading order in $\mathbf{q} = \mathbf{k} - \mathbf{K}$, we obtain the effective two-band Hamiltonian [23]:

$$H_{\text{eff}}(\mathbf{q}) = -\frac{\hbar^2}{2m^*} \begin{pmatrix} 0 & (q_x - iq_y)^2 \\ (q_x + iq_y)^2 & 0 \end{pmatrix} \quad (1.9)$$

where $m^* = \gamma_1/2v_F^2$ is the effective mass. The addition of the interlayer hopping γ_1 fundamentally changes the character of the low-energy bands. The massless Dirac-like linear dispersion of the monolayer is replaced by a parabolic dispersion $E \sim \hbar^2 q^2/2m^*$, where the quasiparticles are now massive chiral fermions carrying a Berry's phase of 2π , reflecting the two-step chiral structure inherited from the two coupled sublattice pairs.

Despite being a distinct stacking configuration from the rhombohedral family, Bernal bilayer graphene shares several qualitative similarities with the multilayer rhombohedral graphene. The non-dimerized sites A_1 and B_2 reside on the outermost surfaces of the crystal, giving the low-energy states a surface-like character analogous to that of rhombohedral multilayers. The parabolic dispersion $E \sim q^2$ is consistent with the general $E \sim q^N$ rule for N -layer rhombohedral graphene evaluated at $N = 2$. A perpendicular displacement field opens a tunable gap between the conduction and valence bands, and a van Hove singularity develops at the band edge [25] in a manner directly analogous to rhombohedral multilayers. In this sense, Bernal bilayer graphene can be viewed as the $N = 2$ member of a broader family of chiral graphene multilayers, despite its different

stacking geometry.²

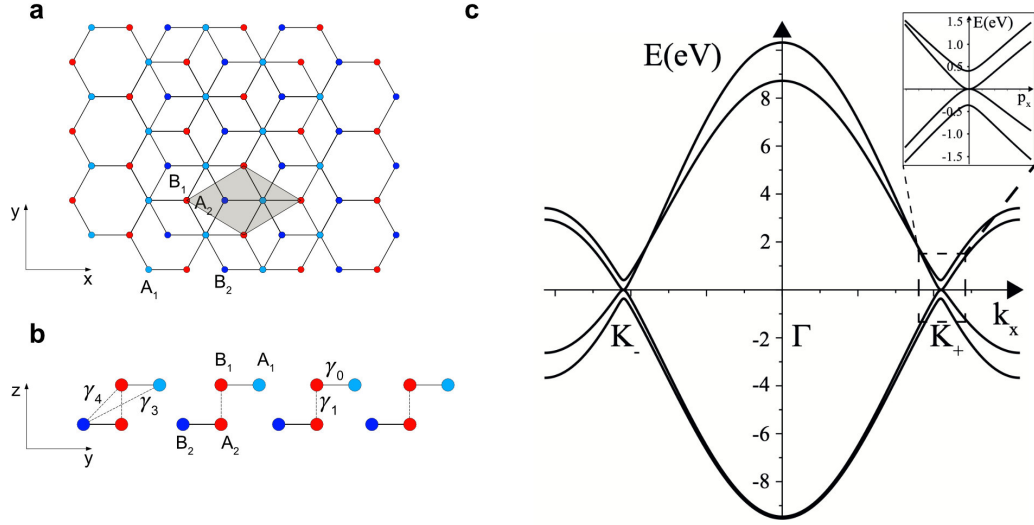


Figure 1.2: Bilayer graphene lattice, tight binding hopping parameters and band structure. **a** and **b** show top and cross sectional view of bilayer graphene lattice. Sites A_1 and B_1 from the top layer are shown as light blue and red respectively and sites A_2 and B_2 from the bottom layer are shown as red and dark blue respectively. Shaded rhombus in **(a)** depicts the typical unit cell of bilayer graphene. Tight binding hopping parameters γ_0 , γ_1 , γ_3 and γ_4 are depicted in **(b)**. **c**, electronic dispersion of bilayer graphene, reproduced from [23] (Fig. 3). Inset shows a zoomin around the K point.

1.3.3 Trilayer Graphene: Two Stackings

Adding a third layer to bilayer graphene introduces a choice of stacking order that has profound consequences for the low-energy electronic structure. The third layer can be placed to lie exactly on top of the first layer making it the ABA (Bernal) (Fig. 1.3a) or it can be placed such that it does not exactly match either of the previous layers making it the ABC (rhombohedral) configuration (Fig. 1.3b). Inspired by ref. [24, 11], I will sketch out how these two arrangements lead to such different electronic structures.

In ABA-stacked trilayer graphene, every other graphene sheet lies exactly on top of each other (Fig. 1.3a). As a result, the crystal possesses a mirror symmetry that exchanges

²A detailed derivation of the band structure of Bernal bilayer graphene can be found in ref. [23]

the top and bottom layers through the central layer. Due to this symmetry, it is convenient to form symmetric and antisymmetric linear combinations of the outer-layer orbitals, $B_{\pm} = (B_1 \pm B_3)/\sqrt{2}$. Since the central site A_2 lies on the mirror plane and is even under this symmetry, it couples only to the symmetric combination B_+ , while the antisymmetric combination B_- is symmetry-forbidden from hybridizing at the leading interlayer hopping scale.

Both sites B_1 and B_3 are connected by γ_1 to A_2 , however only the symmetric combination participates in the strong interlayer hybridization. This leads to the formation of a dimerized bonding–antibonding pair involving A_2 and B_+ , which is pushed to high energies of order γ_1 . The orthogonal combination B_- does not participate in this dimerization and therefore remains effectively decoupled at leading order.

Thus, ABA stacking can be viewed, at low energies, as a coexistence of bilayer-like and monolayer-like electronic degrees of freedom arising directly from its mirror symmetry and incomplete dimerization structure (Fig. 1.3c). More generally, in ABA-stacked N -layer graphene, this structure extends to a set of alternating monolayer-like and bilayer-like subbands determined by the mirror symmetry about the central layer and the pattern of interlayer connectivity across the stack.

The ABC arrangement is fundamentally different. In this configuration, the stacking is chiral: the B site of each layer sits directly above the A site of the layer above, but only in one direction (Fig. 1.3b). The interlayer hopping therefore generates an asymmetric chain of coupled interior sites, (B_1, A_2) and (B_2, A_3) , which form strongly hybridized bonding–antibonding pairs due to the large interlayer coupling γ_1 . This sequential dimerization gaps out the interior states at energies of order γ_1 .

Crucially, the two end sites A_1 and B_3 do not have interlayer partners and are therefore left unpaired at low energy. The low-energy physics of ABC-stacked trilayer graphene is therefore governed entirely by states localized on the outermost layers, while the interior sites can be integrated out.

With only A_1 and B_3 contributing to the low-energy subspace, the effective Hamiltonian must connect these two sites through a third-order virtual hopping process that traverses the entire chain via three intralayer hoppings γ_0 and two interlayer hoppings γ_1 . Importantly, the intermediate dimerized states lie at energies of order γ_1 , so the interlayer processes enter as virtual transitions and generate an effective coupling

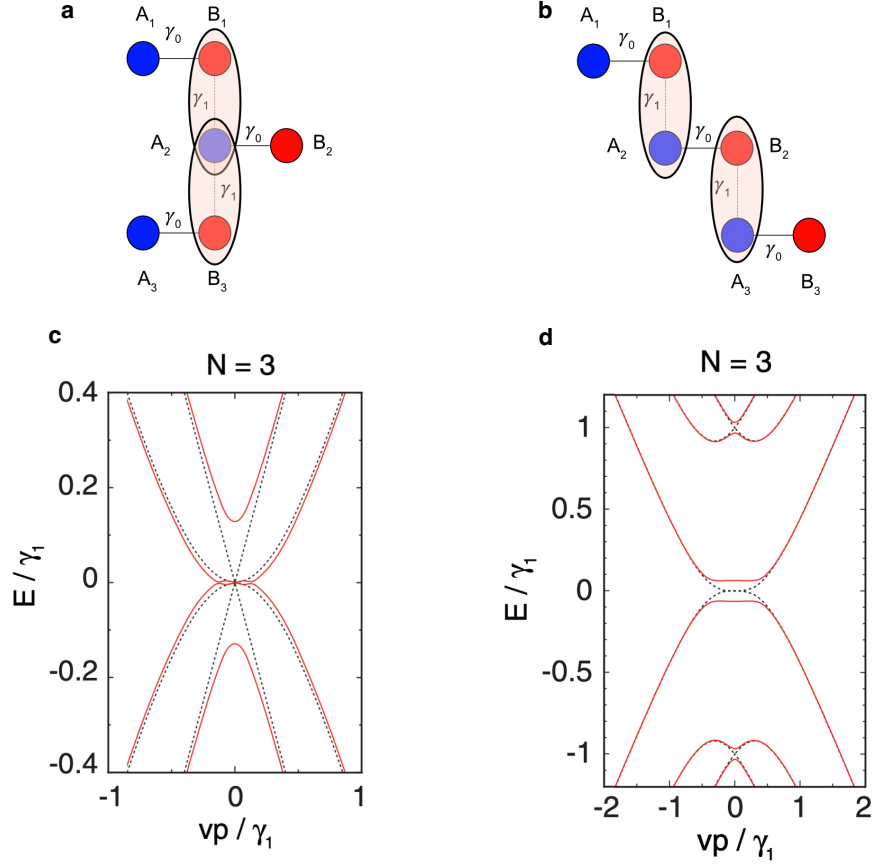


Figure 1.3: ABA and ABC trilayer graphene lattice and electronic structure. **a** and **b**, cross-sectional views of unit cells of ABA and ABC trilayer graphene respectively. A_i (blue) and B_i (red) denote the sublattice sites in the i^{th} layer. Dimerized sites are in the shaded ellipse. **c** and **d** show the electronic dispersion of ABA and ABC trilayer graphene respectively (plotted in normalized units). Black dotted line show the dispersion with displacement field $D = 0$ and solid red line for $D > 0$. **c** and **d** are reproduced from [24] (Fig. 2a and Fig. 4a respectively)

suppressed by factors of $1/\gamma_1^2$, whereas the intralayer hoppings contribute powers of momentum near the K point.

The resulting effective two-band Hamiltonian, projected onto the basis A_1, B_3 , takes the form:

$$H_{\text{eff}}(\mathbf{q}) = -\frac{(\hbar v_F)^3}{\gamma_1^2} \begin{pmatrix} 0 & (q_x - iq_y)^3 \\ (q_x + iq_y)^3 & 0 \end{pmatrix} \quad (1.10)$$

This yields a cubic dispersion $E_{\pm}(\mathbf{q}) = \pm \frac{(\hbar v_F)^3}{\gamma_1^2} |\mathbf{q}|^3$ (Fig. 1.3d), with an associated Berry phase of 3π , reflecting the three-step chiral hopping process connecting the two surface sites. The bandwidth of the low-energy bands is therefore significantly reduced compared to the monolayer and bilayer cases. This follows directly from the cubic dispersion: near $k = 0$, the energy vanishes more rapidly than in linear or quadratic bands, resulting in an effectively flatter low-energy spectrum. As a consequence of this distinctive band structure, rhombohedral trilayer graphene, despite being metastable, has attracted significant interest as a platform for strongly correlated electronic phases.

1.3.4 N -layer rhombohedral graphene

The procedure demonstrated for rhombohedral trilayer graphene naturally generalizes to the case of N -layer rhombohedral graphene. Fig. 1.4 shows the unit cell for N -layer rhombohedral graphene. In this stacking configuration, interlayer hopping γ_1 generates $N - 1$ strongly hybridized dimer pairs whose energies are pushed to the scale of order γ_1 , leaving only two unpaired sites, A_1 and B_N , as the low-energy degrees of freedom.

The low-energy electronic structure is therefore entirely governed by these two outer-layer sites. Analogous to the trilayer case, they are connected only through an N^{th} order virtual hopping process involving N intralayer hoppings (γ_0) and $N - 1$ interlayer hoppings (γ_1). The interlayer processes enter as virtual transitions through high-energy dimer states, leading to suppression by powers of $1/\gamma_1$. The resulting effective Hamiltonian projected onto the low-energy subspace A_1, B_N can thus be written as:

$$H_{\text{eff}}(\mathbf{q}) = -\frac{(\hbar v_F)^N}{\gamma_1^{N-1}} \begin{pmatrix} 0 & (q_x - iq_y)^N \\ (q_x + iq_y)^N & 0 \end{pmatrix} \quad (1.11)$$

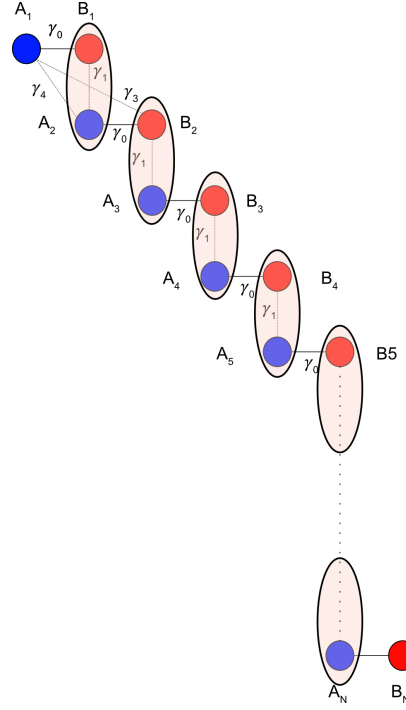


Figure 1.4: **Unit cell of N-layer rhombohedral graphene.** Cross-sectional view of the unit cell of N-layer rhombohedral graphene. A_i (blue) and B_i (red) denote the sublattice sites in the i^{th} layer. Dimerized sites are in the shaded ellipse. Sites A_1 and B_N are left undimerized.

This yields the dispersion relation $E_{\pm}(\mathbf{q}) \sim \pm |\mathbf{q}|^N$, which exhibits an increasingly steep power-law dependence on momentum with increasing layer number. Within this simplified picture, the characteristic bandwidth of the low-energy bands decreases rapidly with N , suggesting that thicker rhombohedral graphene stacks should provide an increasingly flat-band environment favorable for interaction-driven phenomena.

However, this effective two-band description neglects additional interlayer and intralayer hopping processes beyond nearest-neighbor terms. As will be discussed in the following section, these additional terms qualitatively modify the low-energy spectrum by introducing anisotropies that progressively unflatten the bands, with effects that become more pronounced as the layer number increases.³

³For a detailed derivation of the bandstructure of N-layer rhombohedral graphene see ref. [11]

1.3.5 Higher order hopping terms

As shown in the preceding section, the minimal model retaining only γ_0 and γ_1 describes perfectly flat low-energy bands with circular symmetry around the K points as well as power law dispersion. However, including additional higher-order hopping terms modifies this picture in ways that are relevant to understanding the physics of this system.

The parameter γ_3 describes skew interlayer hopping between the non-dimer sites of adjacent layers, specifically the A_i - B_{i+1} (Fig. 1.4) hopping processes that are distinct from the vertical dimer hopping B_i - A_{i+1} captured by γ_1 . In the absence of γ_3 , the low-energy electronic dispersion near the K point is approximately isotropic, leading to circular Fermi surfaces with an emergent rotational symmetry. However, the γ_3 hopping term retains information about the underlying lattice geometry and the directionality of the carbon-carbon bonds. Consequently, it breaks the emergent continuous rotational symmetry of the minimal model and restores the intrinsic threefold rotational symmetry of the honeycomb lattice. The resulting distortion of the low-energy bands and Fermi surfaces, which reduces the symmetry from continuous rotational symmetry to C_3 , is known as trigonal warping [22].

Trigonal warping qualitatively alters the topology of the Fermi surface and can drive a Lifshitz transition, in which the Fermi surface changes from a single connected pocket into multiple disconnected pockets or vice versa. This transition has important consequences for the density of states. In particular, the saddle points separating the warped pockets generate van Hove singularities, which appear as sharp peaks in the density of states $D(E)$. The resulting enhancement of the low-energy density of states strengthens the role of electron-electron interactions and increases the susceptibility of the system to correlated electronic instabilities [26].

In rhombohedral multilayer graphene, these effects acquire a strong layer-number dependence. While the minimal γ_0 - γ_1 model yields an increasingly flat and isotropic dispersion $E(\mathbf{q}) \sim |\mathbf{q}|^N$, this apparent flattening becomes progressively less robust with increasing N . The essential reason is that the isotropic chiral term scales as $|\mathbf{q}|^N$, whereas the leading γ_3 -induced trigonal warping corrections enter at lower order in momentum (effectively $|\mathbf{q}|^m$ with $m < N$). Consequently, sufficiently close to the band-touching point, the γ_3 contribution always dominates over the isotropic chiral dispersion, since

$|\mathbf{q}|^m \gg |\mathbf{q}|^N$ as $|\mathbf{q}| \rightarrow 0$. This hierarchy implies that the effective energy window in which the spectrum appears isotropic shrinks with increasing layer number, and the low-energy physics becomes increasingly governed by anisotropic terms.

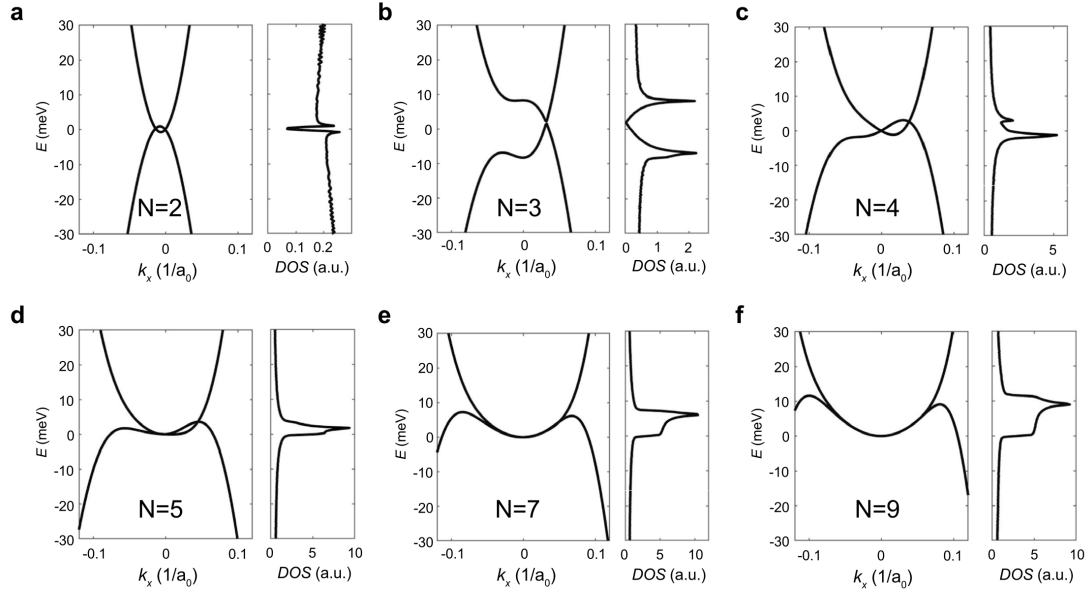


Figure 1.5: Single particle band structure and density of states of rhombohedral multilayer. a-f, Tight-binding calculation of single-particle band structure and density of states (DOS) for rhombohedral multilayer graphene (layer number $N = 2, 3, 4, 5, 7, 9$), reproduced from [27] (ED. Fig.2). Due to the remote hopping terms, the band structure deviates from $E \sim k^N$ at low energy. Rhombohedral pentalayer graphene has the flattest band among all layer numbers.

As a result, instead of a monotonically vanishing bandwidth with increasing N , the band flattening predicted by the minimal model is cut off by trigonal warping. The competition between the $|\mathbf{q}|^N$ isotropic dispersion and the lower-order anisotropic contributions leads to a non-monotonic evolution of the effective low-energy bandwidth, with maximal apparent flatness occurring at intermediate layer number (typically around $N \sim 5-6$) (Fig. 1.5). Beyond this regime, the increasing dominance of trigonal warping reconstructs the low-energy spectrum into anisotropic pockets, preventing further effective flattening.

The parameter γ_4 describes skew interlayer hopping between same-sublattice sites on adjacent layers, connecting A_i-A_{i+1} and B_i-B_{i+1} (Fig. 1.4). In contrast to γ_3 , this term does

not break the emergent rotational symmetry of the minimal low-energy model. Instead, it couples the low-energy non-dimerized states to the high-energy dimerized states. In the absence of γ_4 , the minimal model predicts electron and hole bands with identical curvature and opposite sign. For example, in N -layer rhombohedral graphene the low-energy dispersion takes the form $E_{\pm}(\mathbf{q}) \sim \pm|\mathbf{q}|^N$, reflecting an approximate electron-hole symmetry.

Including γ_4 generates momentum-dependent diagonal corrections to the effective low-energy Hamiltonian through virtual mixing with the high-energy dimerized states. These terms break the electron-hole symmetry of the minimal model, causing the conduction and valence bands to acquire slightly different curvatures. Although γ_4 is smaller in magnitude than γ_3 , it must be included in realistic band structure calculations, since electron-hole asymmetry is readily observed in experimental measurements [22].

Similar to trigonal warping, this electron-hole asymmetry becomes more apparent in larger layer number N as we see in (Fig . 1.5), the electron and hole bands acquire opposite curvatures in the limit of small $\mathbf{q}$ with increasing layer number. In fact, this is the exact origin of the semimetallic regime, which the minimal model does not capture at all. It also tells us that the semimetallic regime will survive up to a larger D in thicker rhombohedral graphene, something that we see in our experiment.

1.3.6 Surface States

Despite the modifications introduced by higher-order hopping terms, the low-energy states near the K point remain dominated by the undimerized sites on the outermost layers of the crystal. At small momenta away from the K point, the spectral weight of the wavefunction is almost entirely concentrated on these surface layers. This is illustrated in Figure 1.6a, which shows a self-consistent Hartree-Fock calculation for 8-layer rhombohedral graphene [28]: near the K point, the valence and conduction bands are polarized to opposite surfaces of the crystal. As the momentum increases away from K, the intralayer and skew interlayer couplings grow, coupling the end sites of the chiral chain more strongly to the bulk and distributing the wavefunction weight more evenly across the stack. The surface polarization is therefore strongest precisely near the K point, where the dispersion is flattest and the density of states is highest.

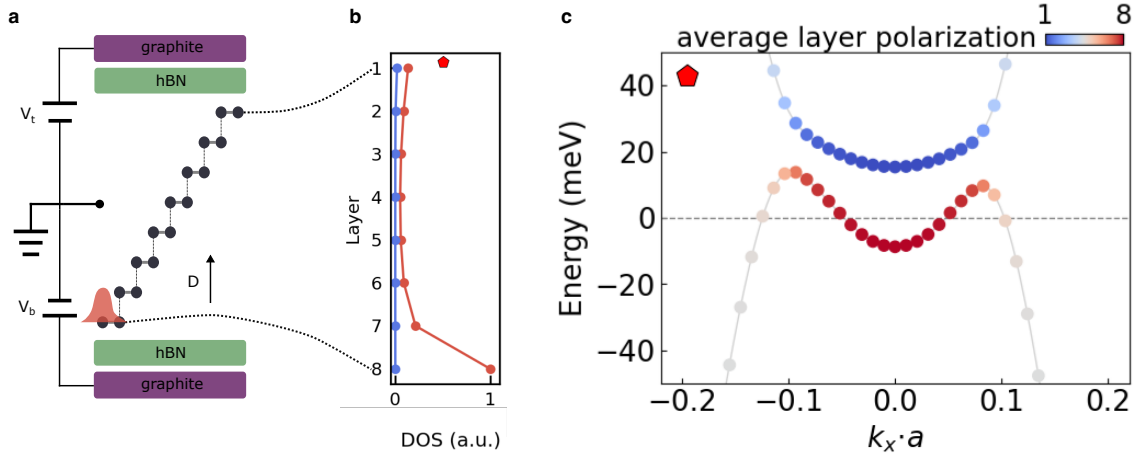


Figure 1.6: **Layer polarization of electrons in 8L rhombohedral graphene.** **a** Schematic of a R8G sample showing the electronic states in the valence bands are localized to the layer 8 (layer closest to the back gate). **b**, Calculated layer resolved density of states (DOS) for $D > 0$ and $n < 0$. The blue curve corresponds to the conduction band and the red curve corresponds to the valence band. **c**, Band structure calculation from the same point in $n - D$ space as (b), with the average layer polarization given by the color scale. The average layer polarization is strongest at low energy and when $k_x = 0$.

This has an important consequence for correlated physics. The van Hove singularity at the band edge, where the density of states diverges, is populated predominantly by states lying close to the K point, which are the most strongly surface-polarized. Since interaction-driven phenomena are most likely to emerge where the density of states is highest, it follows that the correlated phases in rhombohedral graphene arise predominantly from electrons living on a flat, surface-localized Fermi surface.

Figure 1.6b shows the layer-resolved density of states for 8-layer rhombohedral graphene at finite displacement field, in the regime where only the valence band is populated. The wavefunction of the valence band is exponentially localized to the bottom-most layer at experimentally relevant carrier densities, confirming the surface-polarized picture quantitatively. In the semimetallic regime, where the conduction and valence bands overlap, the two carrier types are polarized to opposite surfaces of the crystal. This dual-surface polarized semimetallic state, in which electron and hole pockets coexist while being spatially separated across the thickness of the crystal, is qualitatively distinct from any previously studied regime of rhombohedral

graphene, and will be the central object of investigation in Chapter 2.

1.3.7 Displacement Field Tunability

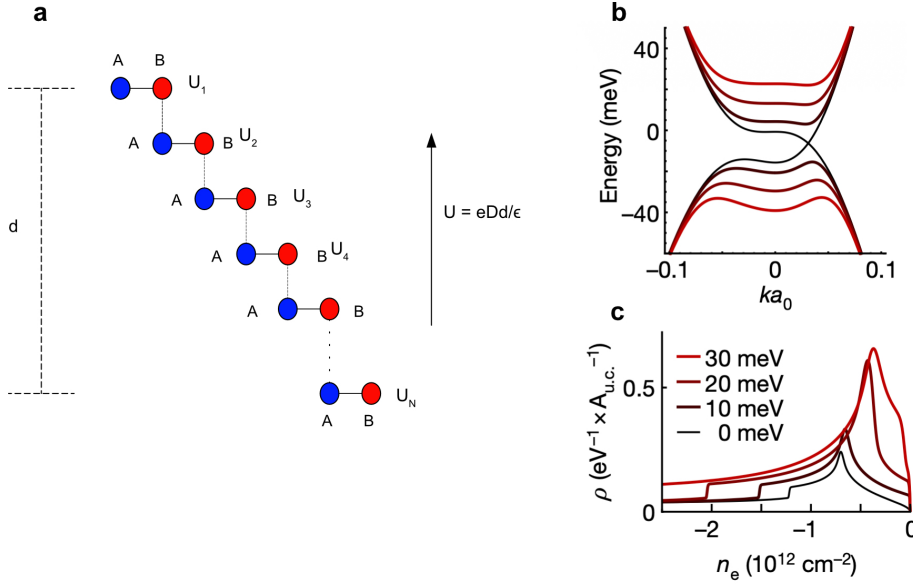


Figure 1.7: **Displacement field tunable bands in rhombohedral graphene.** **a**, Unit cell of N -layer rhombohedral graphene in a displacement field. The displacement field leads to different onsite potentials on the undimerized sites. **b** and **c** show electronic dispersion and density of states (DOS) vs n calculated for different interlayer potentials. **b** and **c** have been reproduced from ref [29]

A key experimental handle in rhombohedral graphene is the perpendicular displacement field D , applied via top and bottom electrostatic gates. The displacement field enters the tight-binding Hamiltonian as a layer-dependent on-site potential. If we assume a perpendicular electric field creates a uniform potential gradient across the stack, the potential energy of an electron on layer j is given by:

$$U_j = eEd \left(j - \frac{N+1}{2} \right) \quad (1.12)$$

where E is the electric field magnitude and $d \approx 3.35 \text{ \AA}$ is the interlayer spacing. This adds a diagonal term to the full Hamiltonian:

$$H = H_0 + H_D, \quad H_D = \text{diag}(U_1, U_1, U_2, U_2, \dots, U_N, U_N) \quad (1.13)$$

where each U_j appears twice for the two sublattice sites on layer j . Projecting onto the low-energy surface state subspace $\{A_1, B_N\}$ within the minimal model retaining only γ_0 and γ_1 , the effective Hamiltonian becomes:

$$H_{\text{eff}}(\mathbf{q}) = \begin{pmatrix} U_1 & -\frac{(\hbar v_F)^N}{\gamma_1^{N-1}}(q_x - iq_y)^N \\ -\frac{(\hbar v_F)^N}{\gamma_1^{N-1}}(q_x + iq_y)^N & U_N \end{pmatrix} \quad (1.14)$$

By the symmetry of the potential about the center of the stack, $U_1 = -\delta$ and $U_N = +\delta$ where $\delta = \frac{N-1}{2}eEd$, so that $U_1 + U_N = 0$ and $U_1 U_N = -\delta^2$. The secular equation $\det(H_{\text{eff}} - E\mathbf{I}) = 0$ then gives:

$$E^2 - \delta^2 - |\Delta(\mathbf{q})|^2 = 0 \quad (1.15)$$

where $|\Delta(\mathbf{q})| = \frac{(\hbar v_F)^N}{\gamma_1^{N-1}}|\mathbf{q}|^N$, yielding the dispersion:

$$E_{\pm}(\mathbf{q}) = \pm \sqrt{\delta^2 + \frac{(\hbar v_F)^{2N}}{\gamma_1^{2(N-1)}}|\mathbf{q}|^{2N}} \quad (1.16)$$

At $\mathbf{q} = 0$, the displacement field opens a gap of magnitude $2\delta = (N-1)eEd$ at the K point, lifting the zero-field band touching [24]. Taylor expanding the dispersion for small $|\mathbf{q}|$:

$$E_{\pm}(\mathbf{q}) \approx \pm \delta + \frac{(\hbar v_F)^{2N}}{2\delta\gamma_1^{2(N-1)}}|\mathbf{q}|^{2N} + \dots \quad (1.17)$$

The leading correction to the gap energy δ is of order $|\mathbf{q}|^{2N}$, which vanishes extremely rapidly near $\mathbf{q} = 0$ for any $N \geq 2$. The band is therefore flat near the K point to leading order, and the degree of flattening is controlled by δ : a larger displacement field suppresses the coefficient of the $|\mathbf{q}|^{2N}$ term, producing a wider flat region and a sharper van Hove singularity at the band edge. Away from $\mathbf{q} = 0$, the bands recover the power-law dispersion $E \sim |\mathbf{q}|^N$ of the zero-field case. The displacement field therefore simultaneously gaps the spectrum and flattens the bands near the K point, providing a pathway to realize isolated flat bands with enhanced density of states. Naturally, exotic

phenomena like FQAH and superconductivity with chiral signatures have been observed in this high displacement field regime.

In this minimal model the dispersion is monotonically increasing in $|\mathbf{q}|$ for all values of δ , since $E_+(\mathbf{q})$ is a sum of two positive definite terms under a square root. The band minimum therefore always sits at $\mathbf{q} = 0$ regardless of the magnitude of the displacement field. Including higher-order hopping terms modifies this picture and can lead to a mexican hat dispersion, in which the band minimum moves to a ring of finite $|\mathbf{q}|$ around the K point. This is illustrated in Fig. 1.7b from ref. [29] for trilayer rhombohedral graphene.

It is important to note, however, that the simple uniform potential assumed above is an idealization. In a real device, the itinerant electrons themselves redistribute in response to the applied field, screening the interlayer potential and causing the actual potential profile across the stack to deviate from the bare linear form. An accurate description of the band structure therefore requires a self-consistent calculation in which the electrostatic potential and the charge density are determined simultaneously. This self-consistent treatment can produce quantitative corrections to the gap size, the bandwidth, and the precise shape of the van Hove singularity that are significant at experimentally relevant carrier densities [30]. Throughout this thesis, band structure calculations presented alongside experimental data are obtained using such self-consistent Hartree-Fock approaches rather than the bare model described here.

1.4 Basic Phenomenology of Rhombohedral Graphene

Having given some theoretical background to understand the electronic structure of rhombohedral graphene, we now review the key experimental phenomena that have been observed in this system. An exhaustive survey of the literature would be beyond the scope of this thesis. This section is only meant to serve as a summary of the most salient observations to serve as context for the work presented in the later chapters.

1.4.1 Stoner Ferromagnetism

As discussed in the previous sections, the bands of rhombohedral graphene have a van Hove singularity at the band edge leading to a very large density of states. This high

density makes the system susceptible to spontaneous isospin (spin and valley) magnetism via the Stoner mechanism [26]. The Stoner criterion is given by $I \cdot D(E_F) > 1$, where I is the exchange interaction and $D(E_F)$ is the density of states at the Fermi level. This quantifies the competition between exchange interactions and kinetic energy, favoring magnetism when the density of states is very large, like in the van Hove singularity of rhombohedral graphene.

In rhombohedral graphene, this kind of ferromagnetism was first observed in trilayer [26]. Compressibility measurements of R3G show regions of varying Landau level degeneracy are separated by cascades of negative compressibility features corresponding to phase transitions. The Fermi surface area extracted from the Fourier transform of the quantum oscillations in ref. [26], shows that these phase transitions correspond to degeneracies of the Fermi surface being lifted. Additionally, the signatures of Anomalous Hall effect and the in-plane magnetic field dependence determine the exact degeneracies lifted: The system goes from a both spin and valley polarized quarter metal to a purely spin-polarized half metal and then to a unpolarized full metal. Note a valley polarized but spin unpolarized state is not favorable due to the anisotropic Hund's coupling term which wants spins in the same valley to point in the same direction. These measurements have also been validated by scanning SQUID measurements where magnetization has been imaged directly[31, 32]. Similar physics has also been observed in thicker rhombohedral graphene [27, 33, 28, 34, 9], although the presence of overlapping bands and other symmetry broken phases make the interpretation of the Fourier transform of quantum oscillations complicated.

1.4.2 Superconductivity in Thinner Rhombohedral Graphene

Superconductivity was first observed in rhombohedral graphene in trilayer devices by Zhou et al. [29], where it was found to emerge on the hole-doped side of the phase diagram in close proximity to an isospin symmetry breaking transition. This initial observation was striking for several reasons: the superconducting state appeared adjacent to a cascade of flavor polarization transitions, suggestive of an interaction-driven rather than phonon-mediated pairing mechanism, and the in-plane critical field exceeded the Pauli limit by at least an order of magnitude, providing strong evidence for a spin-triplet

order parameter. Since then, superconductivity has been reported across a growing number of crystalline graphene platforms, including Bernal bilayer graphene [25], R4G [35, 14], R5G [14], and R6G [15], with the 4, 5 and 6 layer cases displaying signatures of TRS breaking. This establishes superconductivity as a robust and general phenomenon in rhombohedral graphene rather than an anomaly specific to the R3G case.

Despite the differences in layer number and band structure, several qualitative features recur across these systems. First, superconductivity is consistently observed at high displacement fields, where the conduction and valence bands are gapped and the density of states is enhanced near the band edge. Second, in all cases the superconducting dome appears in close proximity to an isospin symmetry breaking transition, with superconductivity emerging at the boundary between a flavor-polarized metallic state and a more isospin-symmetric phase as a function of carrier density. This proximity to an isospin ordering transition is reminiscent of the phase diagrams of other unconventional superconductors, where superconductivity generically appears at the border of a competing ordered phase, and has motivated proposals for fluctuation-mediated pairing driven by the proximal isospin order [36]. Third, superconducting states that emerge from a spin-polarized normal state consistently show strong violations of the Pauli limit under in-plane magnetic fields, in some cases by more than an order of magnitude [25, 37]. This Pauli limit violation is a hallmark of spin-triplet pairing, since a spin-triplet Cooper pair carries a net spin and is therefore insensitive to the pair-breaking effect of a Zeeman field that destroys spin-singlet pairs.

These observations have so far been largely restricted to systems with layer number $N \leq 6$, where the band structure is relatively well understood. What happens in thicker rhombohedral graphene, where the flat band becomes increasingly surface-localized and the semimetallic regime at low displacement field grows in extent [18], remains an open question. This thesis addresses precisely this regime. Rather than focusing on the high D physics that has been the subject of most prior work, we investigate the low D semimetallic regime of thicker rhombohedral graphene, where the coexisting electron and hole carriers are polarized to opposite crystal surfaces and the normal state is qualitatively distinct from anything studied in the thinner layer systems.

1.4.3 Moiré Potential

In the samples discussed so far, rhombohedral graphene is encapsulated in hexagonal boron nitride (hBN) without deliberate rotational alignment between the graphene and hBN lattices. When the two lattices are instead rotationally aligned, the small lattice mismatch of approximately 1.8% between graphene and hBN generates a long-wavelength moiré superlattice that introduces an additional periodic perturbation to the rhombohedral graphene band structure. This leads to the back-folding of the bands of rhombohedral graphene into the moiré Brillouin zone boundaries, giving rise to topologically non-trivial flat bands with non-zero Chern numbers at certain moiré wavelengths.

The topological richness of the hBN-aligned platform was revealed in [16], where both integer and fractional quantum anomalous Hall effects were observed in a rhombohedral pentalayer graphene/hBN moiré superlattice at zero magnetic field. Quantized Hall resistance plateaus were observed at filling factors $\nu = 1, 2/3, 3/5, 4/7, 4/9, 3/7$ and $2/5$ of the moiré superlattice, alongside signatures of a composite Fermi liquid state at $\nu = 1/2$. These observations have since been extended to a growing number of layer numbers, with integer and fractional Chern insulating states now reported in rhombohedral trilayer, tetralayer, pentalayer, and hexalayer graphene aligned with hBN [38, 17].

A striking and somewhat puzzling feature shared across all of these observations is that the topological states appear exclusively in the moiré-distant regime, where the applied displacement field pushes the carriers away from the graphene/hBN interface and toward the opposite surface, reducing the effective moiré potential experienced by the electrons. This is counterintuitive: one might expect the topological states to be strongest when the moiré potential is largest, yet the opposite is observed. Several theoretical proposals have been put forward to explain this, including interaction-driven spontaneous Chern band formation in which electron-electron interactions themselves open the topological gap with the moiré potential playing only a perturbative role [39], and anomalous Hall crystal states in which the electrons spontaneously break translational symmetry [40]. However, no theoretical consensus has yet emerged on the precise mechanism by which the topological states are stabilized in the moiré-distant

regime, and this remains an active area of both experimental and theoretical investigation.

Chapter 2

SUPERCONDUCTIVITY FROM DUAL-SURFACE CARRIERS IN RHOMBOHEDRAL GRAPHENE

*This chapter has been adapted from the following publication: **Superconductivity from dual-surface carriers in rhombohedral graphene**. Manish Kumar*, Derek Waleffe*, Anna Okounkova*, Raveel Tejani, Võ Tin Phong, Kenji Watanabe, Takashi Taniguchi, Cyprian Lewandowski, Joshua Folk[†], and Matthew Yankowitz[†]. Nature Physics (2026). * denotes equal contribution and [†] denotes corresponding author.*

As noted in Chapter 1, superconductivity has been observed in a number of different rhombohedral graphene systems. While there is no consensus on whether these superconducting states have a common origin, they do share several similarities: they are generally observed at high D when the bands are flat and are isolated. Additionally, they generally show up in the region of $n - D$ phase space where the system's isospin degeneracy is about to change. In contrast, the low D phase space, where the conduction and valence band overlap, has been far less explored. We established in Chapter 1 that the low energy wavefunctions are predominantly localized to the outermost layers of the crystal. Combined with band overlap, this produces a configuration in which electron- and hole-like carriers reside on opposite crystal surfaces.. This unique semimetallic ground state also has a considerably large DOS enhancing the susceptibility to symmetry-breaking phases, making it an interesting regime to look for emergent phenomena.

In this chapter, we present our findings of superconductivity in rhombohedral graphene arising from this charge-delocalized semimetallic normal state. We study two samples: an R8G and a R7G with hBN alignment. We start by establishing the dual surface polarized ground state in $n - D$ phases space. This is accomplished through self-consistent Hartree-Fock band structure calculations as well as tracking screening behavior. In octalayer graphene, the superconductivity appears in five apparently distinct regions of the phase diagram for each sign of an external displacement field. In a moiré

superlattice sample where heptalayer graphene is aligned on one side to hexagonal boron nitride, two regions of superconductivity emerge from a single sharp resistive feature. At higher displacement field, the same resistive feature additionally induces an anomalous Hall state quantised at h/e^2 when the doping is close to one electron per moiré unit cell. Our main finding is that in thicker rhombohedral structures, the distribution of the electronic wavefunction along the c -axis emerges as a crucial new degree of freedom mediating pairing in these systems.

2.1 The Dual-Surface Semimetallic Normal State

2.1.1 Identifying the Semimetallic Regime

We first establish the phase boundaries separating the semimetallic region of rhombohedral graphene from regions where the Fermi level crosses only one band. We do this by comparing transport measurements with self-consistent Hartree–Fock band-structure calculations (full details provided in the Methods and Supplementary Information Section II of ref. [28]). Although we focus on R8G, this discussion generalizes to thicker rhombohedral graphene systems ($N \geq 6$).

Figure 2.1a shows a measurement of the longitudinal resistivity (ρ_{xx}) from an R8G sample, acquired by independently varying the top- and bottom-gate voltages, V_t and V_b , and converted to n and D through a standard linear mapping (see Appendix A). The data reproduces several features familiar from earlier studies of rhombohedral graphene, including band insulators at the CNP at large D , a correlated insulator at the CNP centered around $D = 0$ [41, 27, 42, 43, 44], and a surrounding region of elevated resistance that has been associated with a symmetry-broken phase [45]. Figure 2.1b shows a schematic diagram of these and other key features.

The diagonal “jets” of weakly enhanced ρ_{xx} that emerge upon opening the large- D gap at charge neutrality are marked by the solid black line in Figure 2.1b. These jets follow trajectories of nearly fixed V_t or V_b for $n < 0$. To understand the origin of these features, we examine a Landau fan diagram taken along the black dashed line in Fig. 2.1a, crossing one of the jets, in which magnetic field (B) and V_t are swept at fixed V_b (Fig. 2.1e). The quantum oscillations (QOs) at very negative n follow the Středa formula, projecting to $n = 0$ at $B = 0$. Upon crossing the jet at $n = -5.18 \times 10^{12} \text{ cm}^{-2}$, those oscillations become

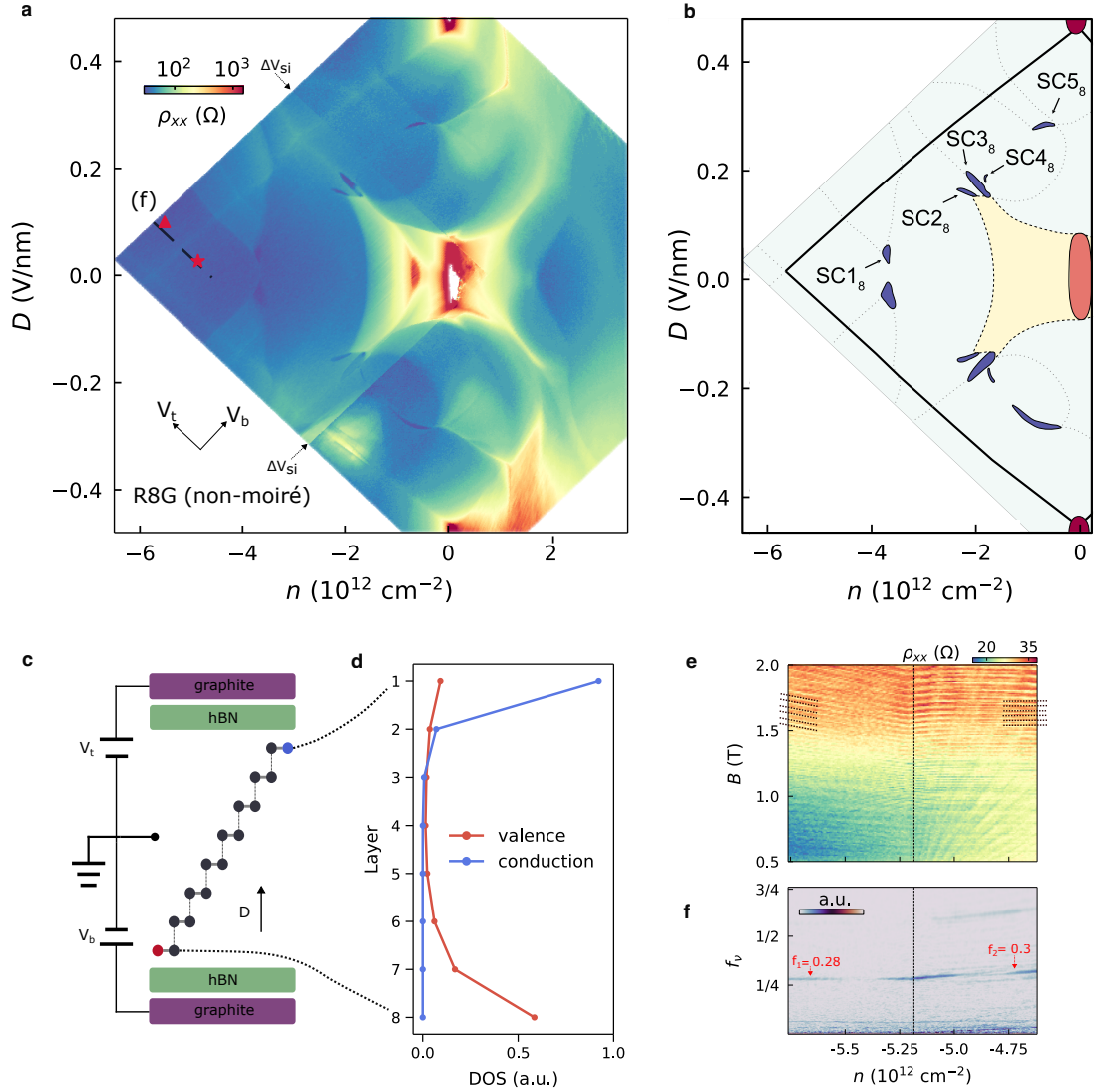


Figure 2.1: Semimetallic properties of rhombohedral graphene and transport in an R8G device. **a**, Measurement of ρ_{xx} versus n and D in a R8G sample without hBN alignment. Arrows denote measurement artifacts (along fixed V_t and V_b) arising from optimizing the silicon gate bias. **b**, Schematic denoting some of the key transport features seen in **(a)**. Band insulators (correlated insulator) at the CNP are denoted in red (dark orange). The light orange indicates a symmetry-broken metallic phase adjacent to the CNP. Dashed and solid lines trace the positions resistive bump features. Blue pockets correspond to superconductivity. **c**, (left) Schematic of a unit cell of octalayer rhombohedral graphene. Red and blue colors denote the atoms that do not form dimers. The R8G is encapsulated by hBN on both sides, with graphite top and bottom gates. **d** Calculated density of states resolved by layer number for the valence and conduction band at $n = -1.95 \times 10^{12} \text{ cm}^{-2}$ and $D = 0.178 \text{ V/nm}$. **e**, Landau fan diagram measured by sweeping V_t and fixed $V_b = -6.0 \text{ V}$, corresponding to the black dashed line in **(a)**. **f**, Fourier transform of the data in **(e)**.

nearly horizontal—almost insensitive to V_t , as if screened—and a new series of features appears, dispersing toward the same V_t value that corresponds to the position of the jet. A Fourier transform in the unscreened region (left half of the graph) shows a dominant frequency (f_v) slightly above $1/4$ (Fig. 2.1f), consistent with a four-fold-degenerate annular Fermi surface. This frequency is nearly constant at large negative n but increases upon crossing the jet, consistent with the simultaneous filling of an additional conduction band Fermi pocket. This kind of screening behavior and fermiology would be consistent with the understanding that the resistive jet is actually the boundary of the dual surface polarized regime, where you go from being in an isolated valence band to having co-existing conducting and valence band pockets as you cross the jet.

2.1.2 Self-Consistent Band Structure Calculations

Separately, we use band structure calculations (Fig. 2.2a) to visualize layer polarization of the bands and to generate a map of the DOS at the Fermi energy (Fig. 2.2b). Crucially, each point in this map is obtained from a self-consistent electrostatic calculation, enabling an accurate conversion to the experimentally relevant n and D values [30] (see Methods section in ref. [28] for more details). The calculation predicts a contiguous region surrounding the charge neutrality point and centered about $D = 0$ in which the Fermi level crosses through both the valence and conduction bands. The boundaries of this region are denoted by the solid black outline atop the map in Fig. 2.2b. Inside this region, Fermi surfaces for the valence band (blue) coexist with those for the conduction band (black). Outside, representative sketches illustrate changing topology of the single Fermi surfaces with n and D .

The calculation reproduces several salient features of the experiment. Notably, the “jets” of weakly enhanced ρ_{xx} that emerge upon opening the large- D gap at charge neutrality, follow trajectories of nearly fixed V_t or V_b for $n < 0$, denoted by the solid black lines in Fig. 2.1b, that closely correspond to the equivalent segments of the outline in the DOS map. We thus interpret these jets as enclosing the region of $n - D$ space in which the R8G is a semimetal.

These observations agree well with the band-structure calculations corresponding to points of the Landau fan on either side of the jet (Fig. 2.2a), which show four-fold

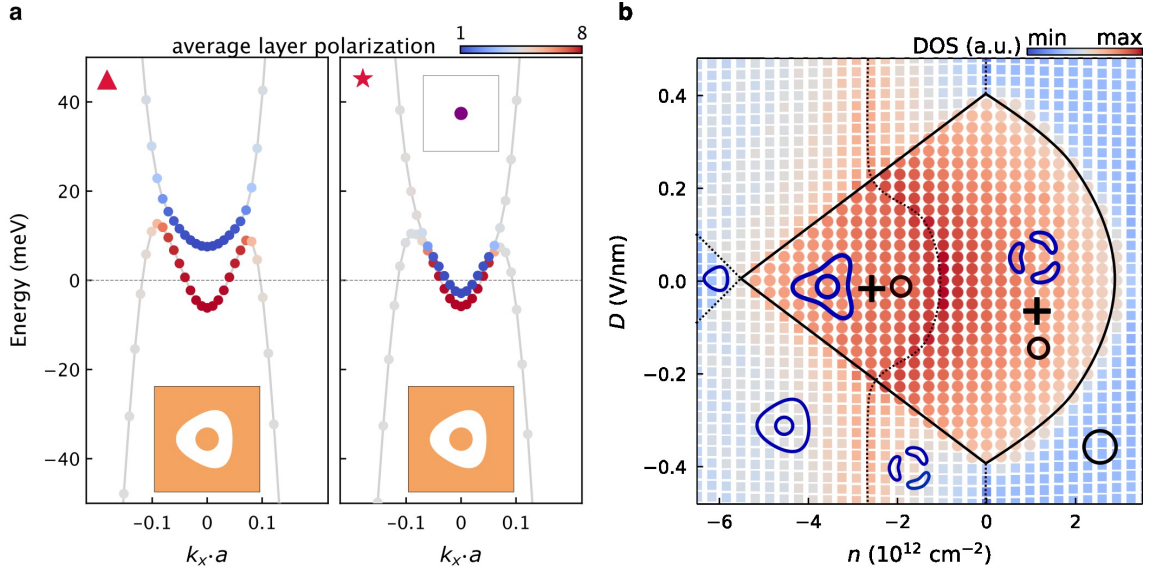


Figure 2.2: **Self-consistent Hartree-Fock bandstructure calculations.** **a**, Calculated band structure at $n = -5.52 \times 10^{12} \text{ cm}^{-2}$ and $D = 0.1 \text{ V/nm}$ (left) and $n = -4.87 \times 10^{12} \text{ cm}^{-2}$ and $D = 0.026 \text{ V/nm}$ (right). The color coding indicates the average layer polarization of each state. The top (bottom) inset shows the constant energy contour of the conduction (valence) band at the Fermi energy ($E = 0$). White coloring indicates empty states. $a = 2.46 \text{ Å}$ is the graphene lattice constant. **b**, Map of the calculated DOS at E_F . Solid and dashed lines trace out local maxima or abrupt steps in the DOS, and separate regions of parameter space with distinct Fermi surface topologies (shown in blue and black for the valence and conduction bands, respectively). The solid black curve traces the crossover between the band-overlapping (circular data points) and band-isolated (square data points) regions, and can be compared directly to the black lines in (a).

degenerate isolated valence bands at the Fermi energy on the one side and overlapping valence and conduction bands at the Fermi energy on the other. However, we note that our Hartree-Fock calculations do not include the possibility of symmetry-broken states. Within the semimetallic regime, spin- or valley-polarized orders may form, as may a layer-antiferromagnetic order in which opposite-spin bands become layer polarized to the outer surfaces. The screening-derived boundaries in (Fig. 2.2b) mark the onset of dual-surface carriers. They do not by themselves distinguish a band-overlap semimetal from a dual-surface state arising from layer-polarized bands within an interaction-driven phase. Although more detailed calculations will help refine the understanding of the

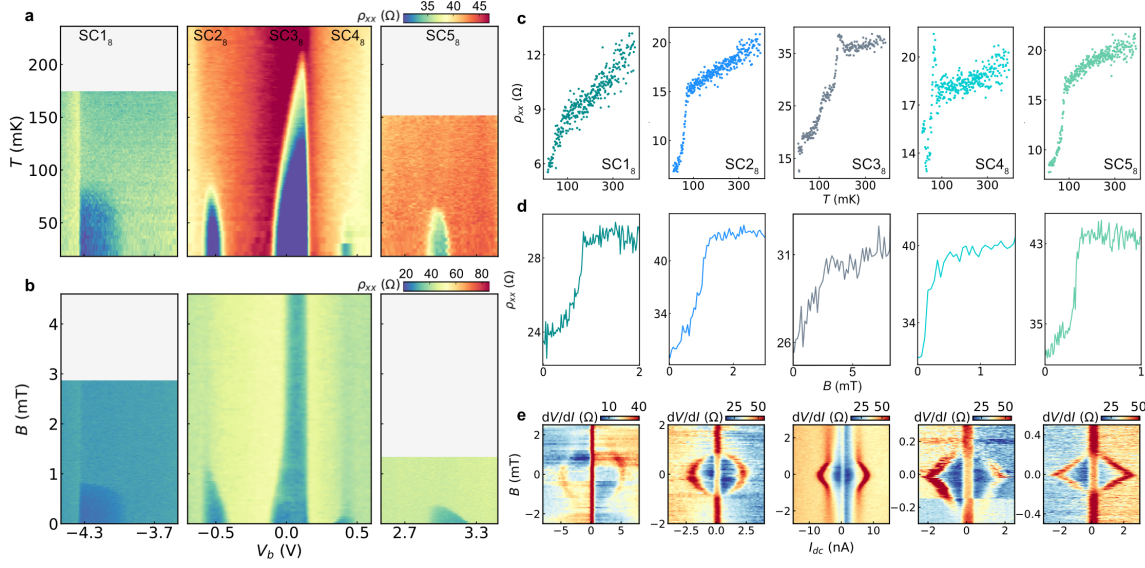


Figure 2.3: **Characterization of superconductivity in R8G** **a**, Maps of ρ_{xx} versus V_b and T around each pocket, acquired with fixed $V_t = -2.75$ V for SC1₈ and $V_t = -2.50$ V for SC2₈-SC5₈. **b**, The same maps versus B . **c**, Measurement of ρ_{xx} versus T at optimal doping of each pocket. The measurements are acquired with a small $I_{dc} < 1$ nA to mitigate the effects of a zero-bias anomaly. **d**, The same measurements versus B , with $I_{dc} = 0$. **e**, Maps of dV/dI versus I_{dc} and B for each pocket. In addition to sharp resistive features forming diamond-like structures arising due to superconductivity, there is also an unexpected anomaly at $I_{dc} = 0$.

symmetry-broken phases across the (n, D) plane, we expect the boundaries of the dual-surface polarized region to remain roughly unchanged, consistent with the screening behavior observed in the QO measurements.

2.2 Superconductivity in Semimetallic R8G

2.2.1 Characterization of the Superconducting Pockets

We find ten low-resistance pockets within the band-overlapping region that, as we show below, correspond to the emergence of superconductivity (Figure 2.1b). Five pockets appear for each sign of D , labeled as SC1₈ to SC5₈ for $D > 0$ (the subscript denotes the total number of graphene layers). We henceforth restrict our focus to the five pockets that appear at $D = 0$ (SC1₈, SC2₈, SC3₈, SC4₈ and SC5₈), and analyze the $D < 0$ counterparts

in Extended Data Figs. B.3. Figures 2.3a, b show ρ_{xx} versus T and B for the five pockets (taken by sweeping V_b at fixed V_t), exhibiting five dome-like structures consistent with superconductivity. The critical temperatures are small, with $T_c \approx 20$ mK to 170 mK (Fig. 2.3a). The corresponding critical fields are similarly low, with B_c on the order of a few millitesla or less. Measurements of dV/dI versus dc bias current I_{dc} and B also show distinct nonlinearities characteristic of superconductivity (Fig. 2.3e). However, the resistance does not reach zero in any of the five pockets, and dV/dI measurements show unexpected peaks at $I_{dc} = 0$ which likely is a contact effect. Although we do not know the origins of either feature, we note that a non-zero saturation of resistance is commonly reported in low- T_c pockets of rhombohedral graphene [25, 46, 47, 35, 48, 49, 34, 50].

2.2.2 Layer Polarization and gate tracking

It is notable that all superconducting pockets appear along a nearly fixed value of V_t (V_b) for $D > 0$ ($D < 0$). We speculate that this may arise because charge carriers in the normal state reside mainly on the two outer crystal surfaces and are separated by a low-DOS bulk (Fig 2.1a). The system thus behaves as two nearly isolated electron gases, held at a common chemical potential by the small fraction of layer-delocalized states. Each gate couples most strongly to the surface closest to it, with minimal cross-influence due to screening [51], locking features to nearly fixed V_t or V_b rather than along the usual $n - D$ axes. This screening behavior can be directly understood from the Hartree-Fock band structure calculations (e.g., Fig. 2.2a), in which the color scale denotes the average layer polarization of each state. States near the K and K' points ($k = 0$) predominantly reside on the outer surfaces and can screen their neighboring gates when populated, whereas states at larger $|k|$ are layer-delocalized and leak across the bulk of the rhombohedral flake. As argued in Ref. [51], this spatial structure of the wavefunction naturally gives rise to a single-gate tracking behavior, exemplified by the diagonal semimetallic phase boundaries for $n < 0$ in Fig. 2.2b. In this context, the five pockets of superconductivity apparently correspond to a nearly constant doping of the surface carrying the conduction band, and are modulated by doping the valence band, which resides primarily on the opposite surface.

To the best of our knowledge, this is the first known material system in which

superconductivity forms from such a charge-bifurcated distribution, with valence and conduction bands polarized to opposite crystal surfaces. This observation opens new theoretical questions about how the extended spatial distribution of the wavefunction along the stacking axis reshapes the effective pairing interaction.

2.3 Superconductivity in Moiré R7G

2.3.1 Device Overview

The complementary role of the layer-polarized conduction and valence bands in inducing superconductivity is even more clearly seen in a second device: a sample of rhombohedral heptalayer graphene (R7G) aligned to hBN on one side, giving a moiré superlattice with period $L_M = 13.5$ nm and twist angle $\theta = 0.42^\circ$ (determined from Brown-Zak magnetoconductance oscillations (Fig. B.7)).

Figure 2.5 a shows that the moiré R7G device lacks the correlated insulator and symmetry-broken metal near $n = D = 0$ seen in the misaligned R8G device, but at larger D develops correlated insulating states at commensurate moiré band fillings. Crucially, the semimetallic regime at low displacement field is still present, identified by the same jet-and-screening phenomenology seen in R8G.

2.3.2 Two Superconducting Pockets

We observe two distinct superconducting pockets (SC1₇ and SC2₇) emerging out of a sharp resistance bump feature that tracks fixed V_t (shown in light blue in the schematic in Fig. 2.4 a). The more robust pocket, SC1₇, is bounded by two perpendicular bumps that closely track fixed V_b (Fig. 2.5 a, Appendix B Fig. B.4). The weaker pocket, SC2₇, is also directly connected to a perpendicular resistance bump (Fig. 2.5 e). Both SC1₇ and SC2₇ lie within the band-overlapping regime, as determined by analysis of Landau fans taken across the jet emerging from the large- D insulating state at the CNP (Fig. 2.6). In particular, in fig. 2.6 we see a dispersing sequence of QOs with $f_v = 1/4$ for $V_t \lesssim -4.75$ V and screening of those QOs with a corresponding rapid increase of f_v for $V_t \gtrsim -4.75$ V, suggesting that the jet from the CNP defines the band-overlapping to -isolated region, analogous to the behavior seen in the R8G sample.

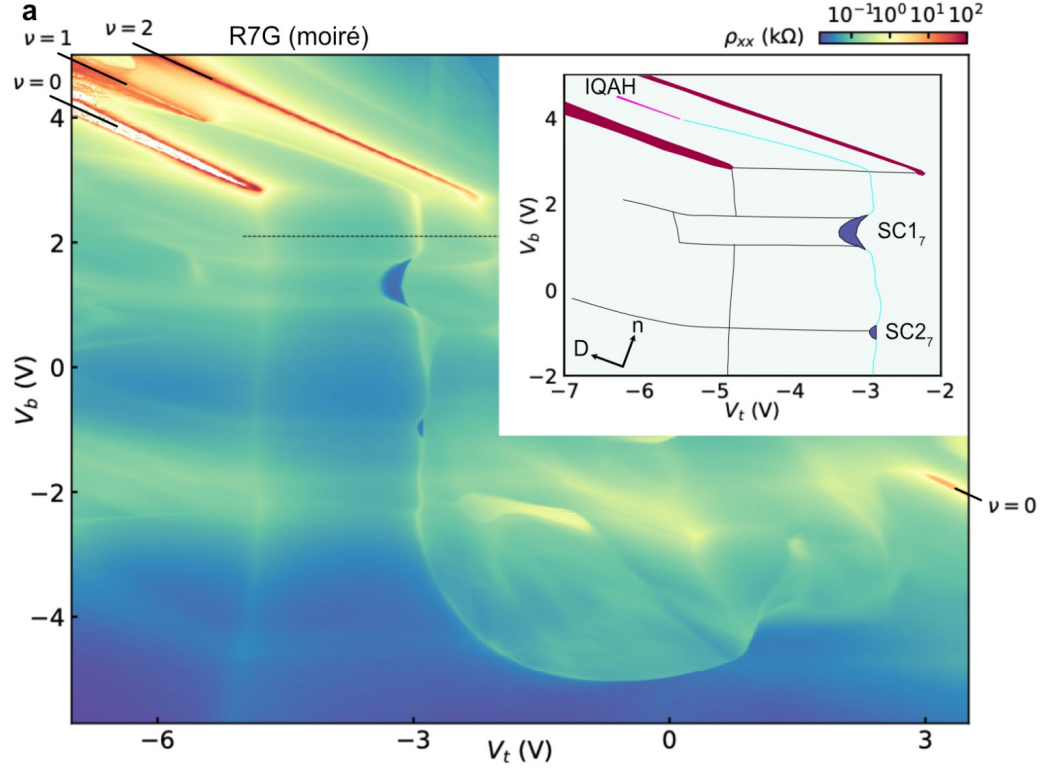


Figure 2.4: **Transport data from 7L RG device a**, Map of ρ_{xx} versus V_t and V_b taken at base temperature and $B = 0$. (inset) Schematic denoting some of the key transport features. Trivial (Chern) insulators at integer ν are shown in red (pink). Black and blue curves denote resistance bump features. Dark blue pockets correspond to superconductivity.

Figures 2.5 b,c show measurements of ρ_{xx} for SC1₇ versus T and B as V_t is swept at fixed $V_b = 1.40$ V. Both exhibit characteristic domes similar to those seen in R8G. Figure 2.5 d displays line traces of ρ_{xx} versus V_t for several selected T . There is a small bump in the resistance at higher T , which develops into a deep suppression of ρ_{xx} at base temperature. A measurement of ρ_{xx} versus T shown in the left inset highlights the abrupt downturn of the resistance below ≈ 90 mK, before saturating near 20 Ohms. The dV/dI measurements in the right inset of Fig. 2.5 d provide additional evidence for superconductivity, showing sharp peaks at a critical current of ≈ 15 nA and a recovery of nearly linear transport above B_c . Analogous measurements for SC2₇ are shown in Fig. 2.5 e, f, g, h.

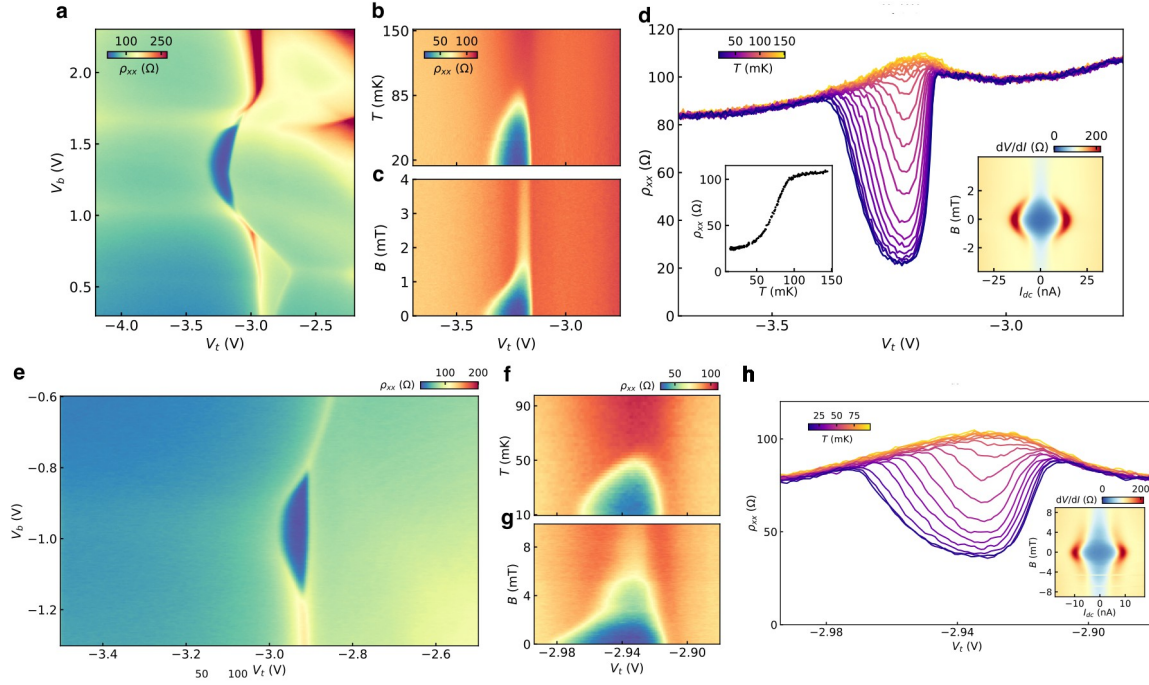


Figure 2.5: Characterizing superconductivity from R7G moiré device **a**, Zoomed-in map of ρ_{xx} around the region of SC17. **b**, Measurements of ρ_{xx} versus V_t and T at fixed $V_b = 1.4$ V. **c**, Similar measurement versus B . **d**, Line traces of ρ_{xx} at various selected T . (left inset) Measurement of ρ_{xx} versus T . (right inset) Measurement of dV/dI versus I_{dc} and B . Measurements in both insets are taken at the optimal doping of SC17 ($V_t = -3.22$ V and $V_b = 1.4$ V). **e**, Map of ρ_{xx} versus V_b and V_t . **f**, Measurement of ρ_{xx} versus V_t and T at fixed $V_b = -0.95$ V and $B = 0$. **g**, Similar measurement as (c) versus B at base temperature. **h**, Measurement of ρ_{xx} versus V_t at several selected values of T . (Inset) Map of dV/dI versus I_{dc} and B (taken at $V_b = -0.95$ V and $V_t = -2.93$ V).

A key observation in the moiré R7G device is that superconductivity is observed only when the conduction band states are polarized away from the moiré interface — i.e., in the moiré-distant configuration. This is consistent with the picture developed for the misaligned R8G device: superconductivity requires the dual-surface semimetallic normal state, in which coexisting valence and conduction band pockets are split to opposite surfaces. When the conduction band is polarized toward the moiré interface, the moiré potential significantly renormalizes the conduction band, disfavoring the charge-separated semimetallic configuration that appears necessary for superconductivity. This conclusion is supported by our self-consistent Hartree-Fock

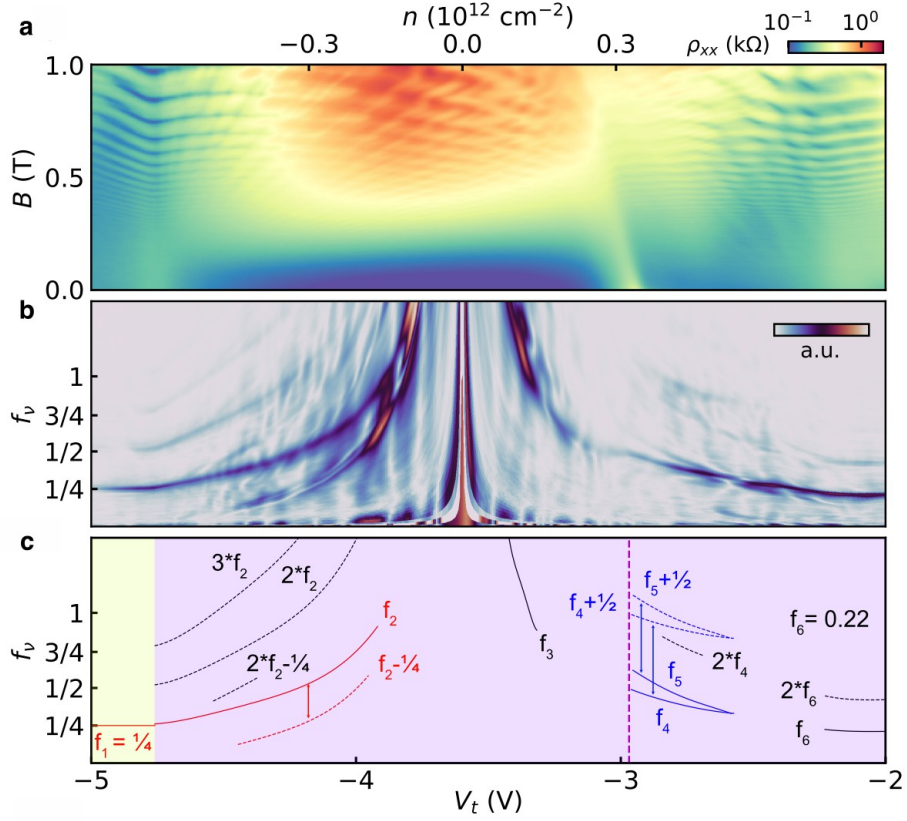


Figure 2.6: **Isospin ordering around the superconducting state in R7G moiré device.** **a**, (top) Landau fan taken by sweeping V_t at fixed $V_b = 2.1$ V, along the black dashed line in Fig. 2.4. **b**, Fourier transform of the Landau fan. **c**, schematic denoting the dominant frequencies seen in the FFT. Background colors denote the band-isolated (yellow) and band-overlap regions (purple).

simulations, which show that a moiré superlattice disfavors the charge-separated semimetal for displacement fields polarizing the conduction band toward the moiré interface (see SI in ref. [28]).

2.4 Discussion

2.4.1 Isospin ordering of the normal state

The ρ_{xx} bump that develops into the two superconducting pockets in R7G seems clearly to be a feature of the conduction band, since it persists into the electron-doped region

($n > 0$) at large D when a gap opens at the CNP, and because its trajectory becomes nearly independent of V_b within the dual-surface regime due to screening from coexisting valence band states on the bottom surface. Detailed analysis of the QOs in Landau fan measurements across the bump feature suggests that it separates an isospin-unpolarized metal to the left from a spin-polarized half-metal to the right (Fig. 2.6), although alternative interpretations remain possible. The resistive bump shifts to more negative V_t with increasing B (Fig. 2.6), further consistent with a spin-polarized state to the right gaining Zeeman energy over the unpolarized state to the left. In-plane field measurements of this sample produce some striking results and indicate an unconventional isospin ordering, the detailed characterization of which will be the subject of Chapter 3. In this picture, SC1₇ (and SC2₇ to a lesser extent) is associated with electrons in the conduction band, lies on the boundary between isospin unpolarized and polarized states, and is turned on and off by valence band features from the opposite crystal surface.

2.4.2 *Role of the Valence Band*

A central question is whether the valence band is merely a spectator or an active participant in the superconducting pairing. Several observations point toward an active role. The gate-tracking behavior indicates that the superconducting pockets correspond to nearly constant doping of the conduction band surface. However, as shown in figure. 2.7 a and b for R8G and R7G respectively, the superconducting states are bounded horizontal resistive bumps corresponding to features in the valence band. Landau fan taken across these two resistive bumps, shown in figure. 2.8 tell us that there is an abrupt fermi surface reconstruction happening. This strongly hints that both bands play a role in establishing the superconducting state, even if superconductivity is primarily tied to conduction band features. An open question is whether superconducting gaps form simultaneously in both the valence and conduction bands, and if so, whether the gaps are comparable in magnitude, or whether one band instead induces superconductivity in the other through a proximity effect.

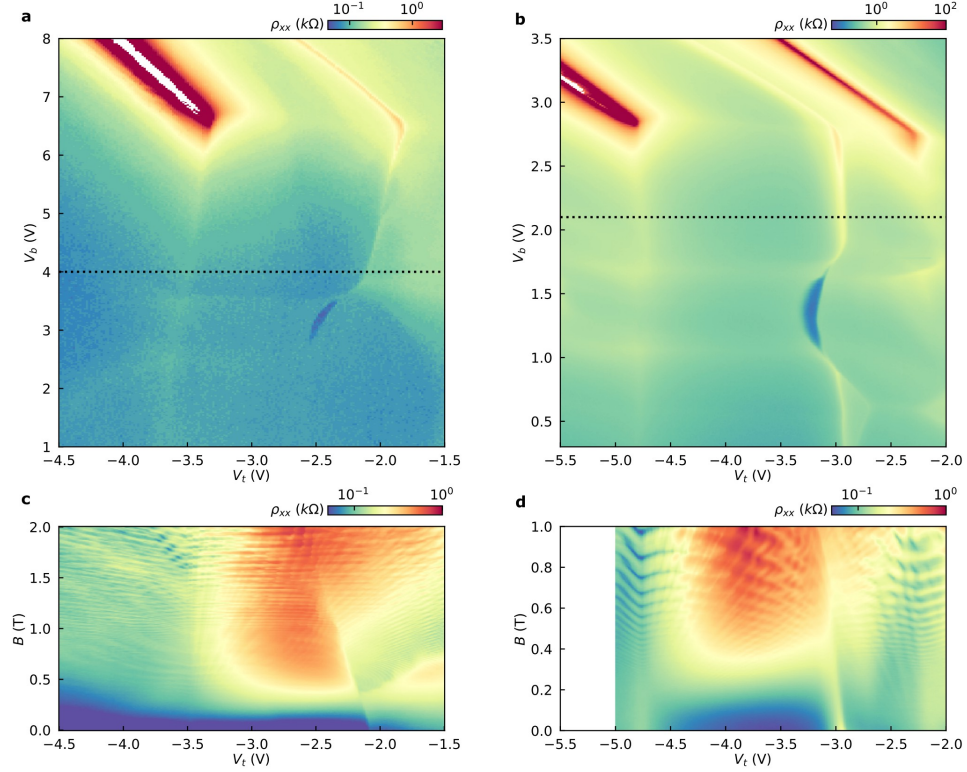


Figure 2.7: **Comparison of SC5₈ from the R8G device and SC1₇ from the moiré R7G device.** **a**, Map of ρ_{xx} versus V_b and V_t around SC5₈ from the R8G device. **b**, Similar measurement around SC1₇ from the R7G device. In both, the diagonal insulating feature in the top left corresponds to the band insulator at the CNP. The resistive feature to its left corresponds is diagonal at large V_b , but becomes nearly vertical at smaller V_b in the band-overlapping regime. In both cases, this feature becomes superconducting when it is intersected by a horizontal resistive bump. the pocket of superconductivity closes when it intersects with a second horizontal resistive bump at smaller V_b . The similarities of these two pockets suggests they share similar origins. **c**, Landau fan taken by sweeping V_t at fixed $V_b = 4.00$ V, along the black dashed line in **(a)**. **d**, Landau fan taken by sweeping V_t at fixed $V_b = 2.10$ V, along the black dashed line in **(b)**. In both, the sharp resistive bump feature that develops into superconductivity drifts to the left with B , consistent with a spin-polarized half-metal to the right gaining Zeeman energy over an unpolarized phase to the left.

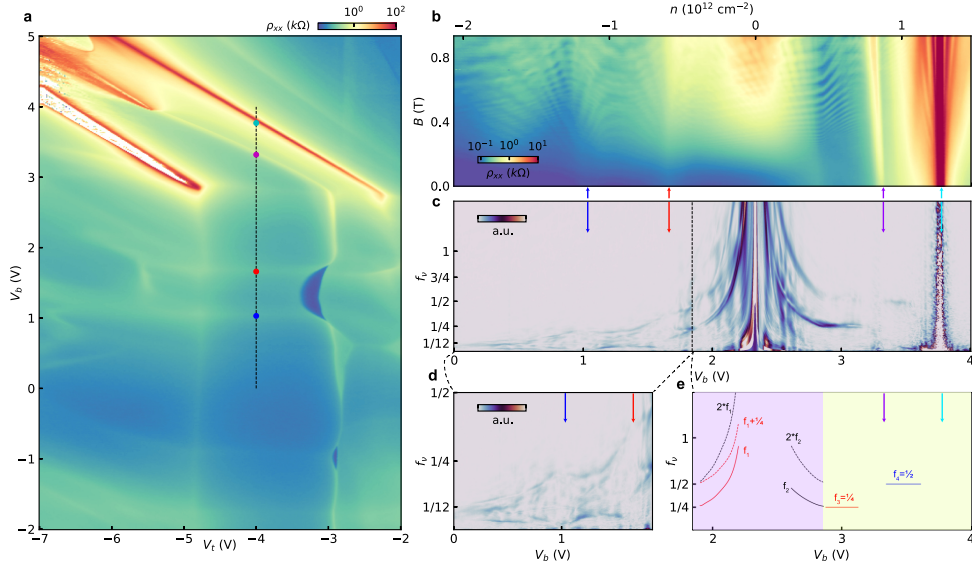


Figure 2.8: **Fermi surface reconstruction in valence bands** **a**, Map of ρ_{xx} versus V_b and V_t taken at base temperature and $B = 0$. **b**, Landau fan taken by sweeping V_b at fixed $V_t = -4$ V, corresponding to the black dashed line in **(a)**. Red, blue, purple, and cyan arrows denote the positions of key features, corresponding to the red, blue, purple, and cyan dots from **(a)**. **c**, FFT of the Landau fan. **d**, Zoom-in of the FFT between $V_b = 0$ and 1.84 V. The dominant frequencies in the FFT abruptly change between the red and blue arrows, indicating a Fermi surface reconstruction in the region where SC1₇ forms at nearby V_t . **e**, Schematic of the dominant frequencies from the FFT between $V_b = 1.84$ V and 4.00 V. For $V_b > 2.8$ V, corresponding to crossing the jet, the dominant frequency is 0.25 indicating a single, simply-connected Fermi surface from the conduction band. For $V_b > 3.32$ V, upon crossing the sharp resistive bump feature associated with superconductivity and the IQAH state, the dominant frequency shifts to 0.5, indicating a half-metal state. The associated feature drifts to the left in the Landau fan, consistent with a spin-polarized half-metal state.

2.4.3 Implications for the pairing mechanism

Superconductivity appears widely across crystalline graphene multilayers, but it is unclear whether the underlying pairing mechanism is universal. The unusual charge-delocalized semimetallic normal state we identify introduces a theoretical scenario in which two spatially separated electronic reservoirs can interact, reshaping the effective pairing interaction. Gate-tracking behavior in both R8G and moiré R7G indicates that superconductivity is primarily tied to features in the conduction band, suggesting that the valence band plays a secondary—yet still important—role. Regardless of pairing

pocket	T_c (mK)	B_c (mT)	ξ (nm)
SC1 ₇	89	1.5	470
SC2 ₇	51	2.0	410
SC1 ₈	40	0.65	710
SC2 ₈	67	1.0	570
SC3 ₈	170	0.8	640
SC4 ₈	20	0.15	1480
SC5 ₈	78	0.32	1010

Table 2.1: **Summary of key parameters relevant to superconductivity.** Values are extracted following procedures described in the Methods section of ref. [28]

mechanism, superconductivity requires an attractive interaction to overcome the renormalized Coulomb repulsion. Within the Tolmachev–Anderson–Morel framework for phonon-mediated superconductivity [52, 53], additional carriers in the valence band could enhance screening and thereby strengthen pairing. Alternatively, the charge-separated geometry resembles “synthetic” electronic pairing proposals [54, 55, 56, 57, 58, 59, 60] in which one charge layer mediates an effective attraction in another (e.g., dynamical, excitonic, or Kohn–Luttinger scenarios). Furthermore, the propensity of crystalline graphene systems to symmetry breaking necessitates consideration of pairing arising from fluctuations of isospin degrees of freedom [61], in which screening from the additional charge layer would naively suppress Coulomb-driven pairing; however, careful consideration is required.

Estimates of the superconducting coherence length, mean free path, and Fermi wavevectors place the system in the clean, weak-coupling limit, though extracting these parameters is complicated by the coexistence of valence- and conduction-band carriers (see Methods and Table 2.1). If confirmed, this regime renders the pairing problem in rhombohedral graphene particularly amenable to controlled theoretical treatments. Future experiments tracking the evolution of superconductivity in the semimetallic regime as a function of layer number will likely be helpful for unraveling its nature. A recent study of R6G revealed superconductivity in a similar region of parameter space as

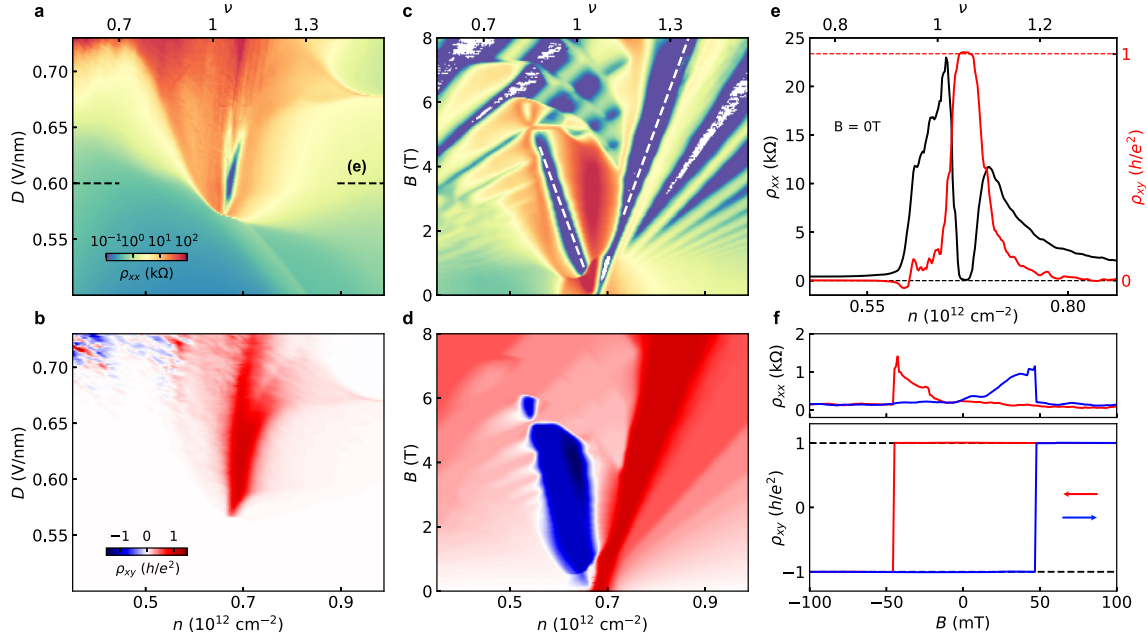


Figure 2.9: Integer quantum anomalous Hall state near $\nu = 1$ in the moiré R7G device. **a**, Map of ρ_{xx} versus ν and D at $B = 0$. **b**, Similar map of ρ_{xy} , antisymmetrized at $|B| = 100 \text{ mT}$. **c**, Landau fan of ρ_{xx} taken at $D = 0.60 \text{ V/nm}$ (marked by the dashed lines in **a**). White dashed lines indicate the Streda trajectories of $C = +1$ and -1 states. White colors indicate negative measured resistances. **d**, Same for ρ_{xy} . **e**, Line traces of ρ_{xx} and ρ_{xy} versus ν at $B = 0$ and $D = 0.60 \text{ V/nm}$. **f**, Measurement of ρ_{xx} and ρ_{xy} as B is cycled at $\nu = 1.06$ and $D = 0.60 \text{ V/nm}$. **a** and **e** are taken at $T = 9 \text{ mK}$. All other measurements are taken at $T = 100 \text{ mK}$.

the SC3_8 pocket [62]; our measurements extend the presence of superconductivity beyond six-layer samples, motivating the possibility that it persists into the regime of bulk rhombohedral graphite. Thicker flakes are of particular interest since the larger spatial separation of the two surfaces could further modify the presence and strength of superconductivity.

2.5 Integer Quantum Anomalous Hall State in Moiré R7G

The same conduction-band feature that hosts superconductivity in the semimetallic region of the moiré R7G device also hosts an integer quantum anomalous Hall state once the bands are separated, with quantization of the Hall resistance to $\rho_{xy} = \pm h/e^2$. Upon

increasing D and exiting the semimetallic regime, the resistance bump abruptly becomes responsive to both V_t and V_b , consistent with a disappearance of valence-band-mediated screening from the remote gate. Figures 2.9 a,b are 2D scans along n and D axes corresponding to the top left corner of Fig. 2.5a, showing the evolution of the resistive bump into an IQAH state when it drifts to $\nu \approx 1$ at $D \gtrsim 0.58$ V/nm. The IQAH state is marked by the simultaneous suppression of ρ_{xx} towards zero (Fig. 2.9a,e) and quantization of $|\rho_{xy}|$ to h/e^2 (Fig. 2.9b,e). In a Landau fan (Figs. 2.9c,d), the relevant ρ_{xx} and ρ_{xy} features slope with a trajectory consistent with $C = +1$ under the Středa formula, and persist to at least 8 T. Additional sweeps taken at $\nu = 1.06$ while cycling B (Fig. 2.9f) show a single hysteresis loop with ρ_{xy} switching between $\pm h/e^2$, providing definitive evidence for the $|C| = 1$ IQAH state. The appearance of a IQAH state with a Chern number of $C = 1$ at high D is in line with previous measurements on four- to ten-layer rhombohedral graphene [16, 38, 63, 17, 64].

Two features in Fig. 2.9 extend beyond conventional IQAH phenomenology. First, the Chern insulator is apparently not pinned exactly to $\nu = 1$ (see Extended Data Fig. B.5). In fact, all of the correlated insulators on the moiré-remote side shift progressively away from integer ν as D increases, underscoring the need to clarify the experimental conditions for opening many-body gaps in thicker rhombohedral samples. By contrast, the correlated insulators on the moiré-adjacent side do not exhibit this behavior. Second, the Landau fan reveals an additional $C = -1$ state that emerges abruptly for $B \gtrsim 0.6$ T, similar to behavior recently reported in moiré R6G [64] and twisted bilayer-trilayer graphene [65].

2.6 Conclusions

We have reported the observation of superconductivity in rhombohedral graphene arising from a charge-delocalized semimetallic normal state, in which coexisting valence- and conduction-band Fermi pockets are split to opposite crystal surfaces. In octalayer rhombohedral graphene, superconductivity appears in five apparently distinct pockets for each sign of displacement field. In a moiré heptalayer device, two pockets of superconductivity emerge from a sharp conduction band feature, which evolves into an integer quantum anomalous Hall state at higher displacement field. In both devices, the

distribution of the electronic wavefunction along the c -axis appears to play an important role in the emergence of superconductivity.

These findings establish a new regime of superconductivity in graphene multilayers and raise new questions about the role of the stacking-axis charge distribution in Cooper pairing. They also reveal intriguing connections between superconductivity and topological many-body phases in moiré superlattices, potentially opening pathways toward engineered topological superconductivity and exotic non-Abelian phases.

Chapter 3

SPIN-TRIPLET SUPERCONDUCTIVITY IN RHOMBOHEDRAL GRAPHENE

*This chapter has been adapted from the following preprint: **Pervasive spin-triplet superconductivity in rhombohedral graphene**. Manish Kumar*, Derek Waleffe*, Anna Okounkova*, Raveel Tejani, Kenji Watanabe, Takashi Taniguchi, Étienne Lantagne-Hurtubise, Joshua Folk[†], and Matthew Yankowitz[†]. arXiv:2511.16578 (2025). * denotes equal contribution and [†] denotes corresponding author*

The preceding chapter established that superconductivity in rhombohedral graphene can emerge from a charge-delocalized semimetallic normal state, in which valence- and conduction-band carriers reside on opposite crystal surfaces. The exact nature of the pairing symmetry of the superconducting state was left unanswered. This question is particularly acute in rhombohedral graphene because the normal state is itself spin-polarized in much of the relevant phase diagram. As discussed in Chapter 1, the large density of states near the van Hove singularity satisfies the Stoner criterion, driving spontaneous isospin ordering that can take the form of a spin-polarized half-metal. Superconductivity emerging from such a parent state has unconventional spin-triplet pairing and survives to much higher critical magnetic fields than spin singlet superconductors. Spin-triplet superconductivity has been observed in Bernal bilayer graphene as well as rhombohedral trilayer graphene, identified by superconductivity being induced by an in-plane magnetic field $B_{\parallel}$ or Pauli paramagnetic limit violation with $B_{\parallel}$

In this chapter, we present a detailed investigation of the spin structure of the superconducting phases in the moiré R7G device introduced in Chapter 2. In Chapter 2 we had identified that $SC1_7$ and $SC2_7$ emerge on the boundary between an unpolarized metal and a spin polarized half metal. We find that modest in-plane magnetic fields $B_{\parallel}$ do not suppress but instead induces and expands superconductivity, transforming the entirety of the sharp resistive ridge in the gate map into an extended superconducting state. The superconductivity persists to $B_{\parallel}$ far exceeding the Pauli paramagnetic limit by

more than an order of magnitude at all measured V_b . This behavior is difficult to reconcile with spin-singlet pairing but is naturally consistent with spin-triplet pairing, for which the Zeeman energy does not constitute a pair-breaking perturbation. We find that the critical temperature T_c evolves non-monotonically with $B_{\parallel}$ in a strongly gate-dependent manner, reflecting the interplay between the layer-dependent charge distribution and the evolving spin configuration of the normal state. We also observe superconductivity adjacent to the integer quantum anomalous Hall (IQAH) state identified in Chapter 2, indicating the coexistence of valley imbalance with superconductivity. We discuss mechanisms consistent with the phase diagram, including considerations of intrinsic spin-orbit coupling, a Hund's spin exchange term, Zeeman energy from an in-plane magnetic field ($B_{\parallel}$), and a general tendency toward isospin polarization when the density of states is large. Together, these observations extend the observation of spin-triplet superconductivity to R7G where it emerges from surface-bifurcated low-energy states.

3.1 Observation of superconductivity induced by in-plane magnetic field

3.1.1 The resistive ridge and field-induced pairing

We first present measurements of the longitudinal resistivity (ρ_{xx}) versus V_b and V_t at $B = 0$ (Fig. 3.1d) and $B_{\parallel} = 1$ T (Fig. 3.1e). At $B = 0$, a sharp resistive peak (marked by arrows in Fig. 3.1d) moves through the gate map, hosting two small pockets of superconductivity, SC1 and SC2 (SC2₇ and SC1₇ in chapter 2). When $B_{\parallel}$ is raised to 1 T, however, the deep blue color signifying low resistance extends along the entire length of the feature. This feature follows approximately constant conduction-band filling, an interpretation explained in detail in Ref. [28] and inferred from its near-perfect tracking of lines of constant carrier density when the conduction and valence band are separated ($V_b > 2.8$ V), then evolving to track vertically when the bands overlap.

Figure 3.1b shows a linecut from these colormaps at $V_b = 1.4$ V, representing the evolution of ρ_{xx} versus V_t in the center of the SC1 pocket as $B_{\parallel}$ is raised from 0 to 1 T. The sharp dip in ρ_{xx} that reflects superconductivity drifts to more negative V_t as $B_{\parallel}$ increases. Figure 3.1c shows the analogous measurement at $V_b = 2.1$ V. In this case, a sharp resistive peak at $B = 0$ becomes a large dip at $B_{\parallel} = 1$ T, located slightly to its left. As shown later, this dip corresponds to field-induced superconductivity emerging from the sharp

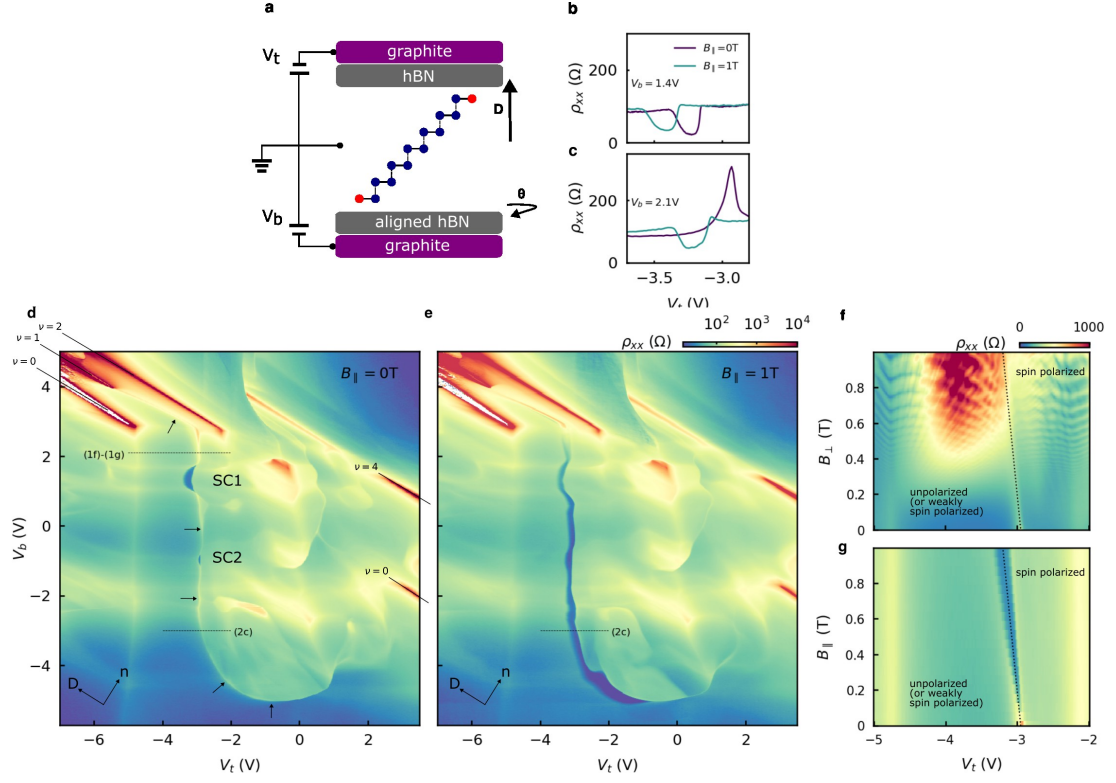


Figure 3.1: Emergence of superconductivity with $B_{\parallel}$. **a**, Schematic of the R7G device with a 13.5 nm moiré superlattice from alignment with the bottom hBN. **b**, Measurements of ρ_{xx} versus V_t at fixed $V_b = 1.4$ V with $B_{\parallel} = 0$ and 1 T. **c**, Same with $V_b = 2.1$ V. **d**, Map of ρ_{xx} versus V_b and V_t at zero field. Arrows denote the sharp resistive feature discussed in the text. Diagonal lines denote selected integer band filling factors, ν . **e**, Same map at $B_{\parallel} = 1$ T. **f**, Landau fan of ρ_{xx} taken versus V_t and $B_{\perp}$ with $B_{\parallel} = 0$ and fixed $V_b = 2.1$ V. **g**, Same measurement taken versus $B_{\parallel}$ at $B_{\perp} = 0$. The black dashed lines in **f** and **g** are identical guides for the eye.

resistive peak.

3.1.2 Identifying the Isospin Polarization of the Normal State

To establish the spin structure of the metallic phases neighboring the superconductivity, we compare transport measurements under out-of-plane ($B_{\perp}$) and in-plane ($B_{\parallel}$) magnetic fields while sweeping V_t across the resistive ridge at fixed V_b (Fig. 3.1 f and g respectively). The key observation is that the resistive features shift in the (V_t, B) plane with nearly identical slopes under both field orientations, despite the fact that in-plane

and out-of-plane fields couple very differently to the orbital degrees of freedom. In a two-dimensional system, $B_{\perp}$ couples to both the orbital and spin degrees of freedom, whereas $B_{\parallel}$ couples only to the spin (provided the system is thin enough that orbital effects of the in-plane field are suppressed). The nearly identical slopes with both field orientations therefore indicate that the Zeeman energy, rather than the orbital coupling, is the relevant energy scale for the features crossing the resistive ridge.

This is consistent with a phase boundary between a spin-polarized state and a weakly polarized (or unpolarized) state. A spin-polarized phase gains Zeeman energy relative to an unpolarized phase when a field is applied, shifting the phase boundary in a direction that lowers the energy of the spin-polarized side. The direction of this shift in both $B_{\perp}$ and $B_{\parallel}$ measurements, together with the analysis of quantum oscillation frequencies across the ridge from Chapter 2 (Fig. 2.6), supports the interpretation that the phase to one side of the resistive ridge is a spin-polarized half-metal, while the other side is a weakly polarized or unpolarized metal. The resistive ridge itself marks the boundary between these two phases.

3.1.3 Normal state nonlinearity

We analyze the emergence of superconductivity with $B_{\parallel}$ in greater detail using the measurements in Fig. 3.2a-c, taken along the black dashed trajectory in Fig. 3.1d ($V_b = -3.0$ V). At $B_{\parallel} = 1$ T, the temperature dependence of resistivity exhibits a characteristic superconducting dome (Fig. 3.2a). Maps of ρ_{xx} versus $B_{\parallel}$ (Figs. 3.2b,c) show that the resistive bump does not shift with field up to 0.5 T, then for $B_{\parallel} > 0.6$ T it shifts to the left as the sharp dip associated with superconductivity emerges (similar behavior recurs across many gate voltages; Extended Data Fig. C.2). Figures 3.2e,f show the differential resistivity (dV/dI) versus dc current (I_{dc}) and $B_{\parallel}$ along the diagonal trajectory marked by the dashed black line in Fig. 3.2b. In addition to the characteristic nonlinearities associated with superconductivity at large $B_{\parallel}$ (Fig. 3.2d), the non-superconducting state at lower field also exhibits pronounced nonlinearities. At $B = 0$ there are sharp dV/dI peaks at $I_{dc} \approx \pm 65$ nA surrounding a lower resistance state at low bias. As $B_{\parallel}$ is raised, the dV/dI peaks move towards zero and disappear near $B_{\parallel} \approx 0.6$ T (similar behavior occurs with $B_{\perp}$, see Extended Data Fig. C.4a,b). The dV/dI

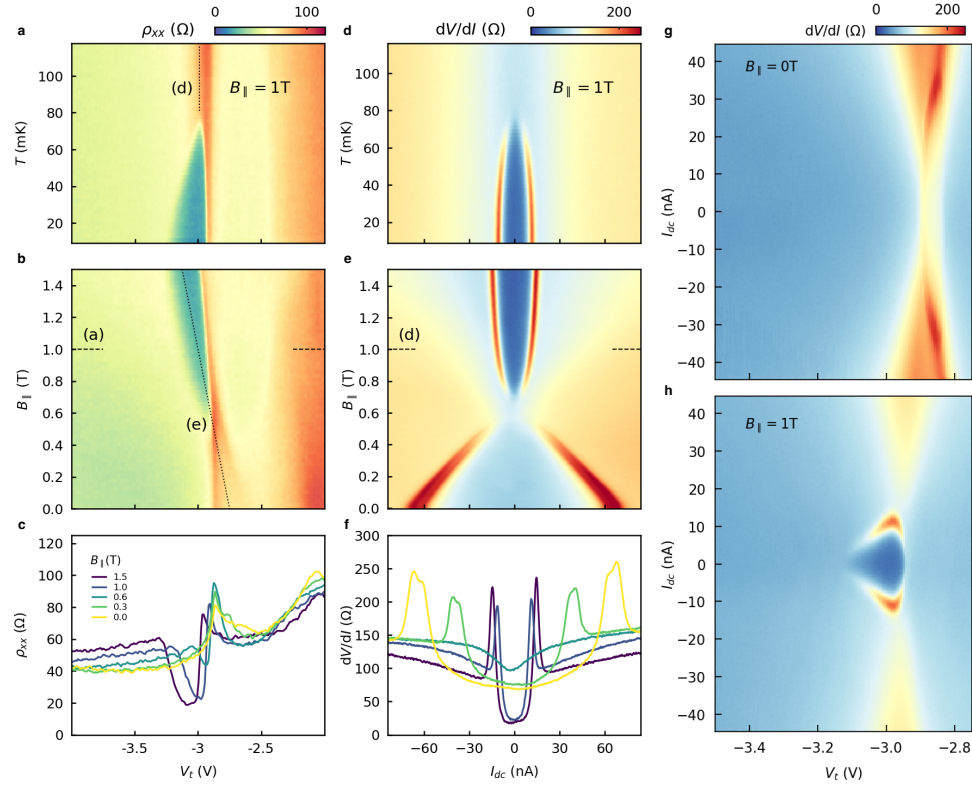


Figure 3.2: **Nonlinearities associated with states along the sharp resistive bump.** **a**, ρ_{xx} versus V_t and T at $B_{||} = 1$ T and $V_b = -3$ V. **b**, ρ_{xx} versus V_t and $B_{||}$ at $V_b = -3$ V. **c**, Selected traces from **(b)**. **d**, dV/dI versus I_{dc} and T at $B_{||} = 1$ T, $V_b = -3$ V and $V_t = -3$ V (black dashed line in **(a)**). **e**, dV/dI versus I_{dc} and $B_{||}$ taken along the trajectory of the tilted black dashed line in **b**. **f**, Selected traces from **(e)**. **g**, dV/dI versus V_t and I_{dc} taken at $V_b = 0$ V and $B_{||} = 0$ T. **h**, Same as **(g)** but with $B_{||} = 1$ T.

peaks reappear at larger $B_{||}$, flanking a deep suppression of dV/dI at small I_{dc} when the dashed black trajectory in Fig. 3.2b crosses over into the superconducting state. Crucially, dV/dI nonlinearities occur only for V_t values that host the elevated-resistance feature at small $B_{||}$ and superconductivity at larger field. Transport at small I_{dc} remains linear in the surrounding metallic phases (Figs. 3.2 g,h, taken at $V_b = 0$), irrespective of their different isospin polarizations.

Owing to the narrowness of the resistive bump, we are not able to resolve associated quantum oscillations that would further diagnose its isospin order. Even so, the sharp dV/dI peaks constrain feasible origins. One possibility is a charge density wave (CDW)

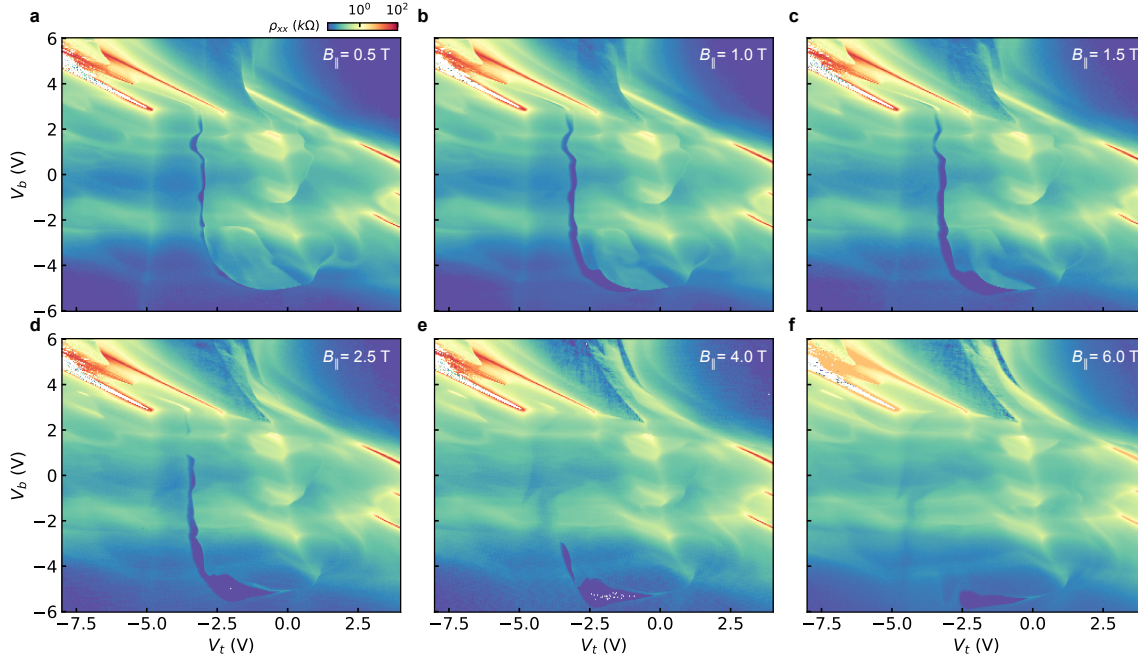


Figure 3.3: **Extent of superconductivity at different $B_{\parallel}$.** Maps of ρ_{xx} versus V_b and V_t taken at **a**, $B_{\parallel} = 0.5$ T, **b**, 1 T, **c**, 1.5 T, **d**, 2.5 T, **e**, 4 T, and **f**, 6 T.

that depins above a critical I_{dc} , although CDW sliding typically reduces dV/dI rather than producing sharp peaks [66]. Another possibility is a state that is weakly valley-polarized (akin to a partially polarized quarter metal or three-quarters metal), in which a dc current exerts a torque that can move orbital-magnetization domain walls [67], yielding a sharp dV/dI feature. Although we do not observe clear anomalous Hall effect associated with the sharp resistive bump—as typically accompanies valley imbalance—we later show a superconducting diode effect under $B_{\parallel}$ that keeps this scenario in play. Related behavior appears in Bernal bilayer graphene (BBG), which also shows a sharp resistive feature with nonlinear transport at $B = 0$ and superconductivity induced from that phase by a small in-plane magnetic field [68]. It is not yet clear whether the low- and high-field states in our sample and in BBG are identical, but their close correspondence motivates the possibility of a shared origin.

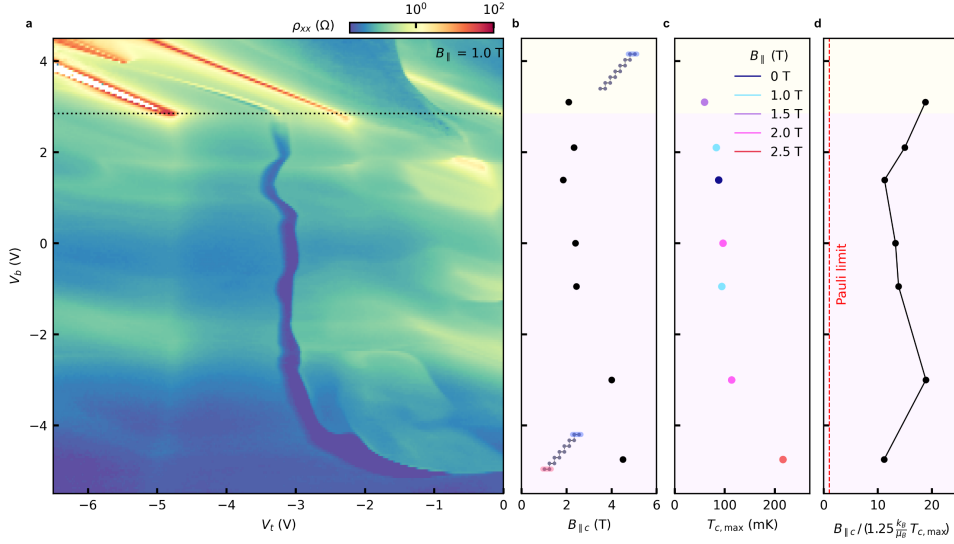


Figure 3.4: Assessment of the Pauli limit violation. **a**, Map of ρ_{xx} versus V_b and V_t taken at $B_{||} = 1$ T. The dashed line shows the boundary between band isolated (above) and band overlap (below) regimes. **b**, Characteristic maximum in-plane field, $B_{||c}$, in the superconducting river as a function of V_b . The values of $B_{||c}$ at $V_b = -3, -4.75$ V are an estimated lower bound. The values of $B_{||c}$ at $V_b = 3.1, 2.1, 1.4, 0, -0.95$ V are extracted by taking 90% of the normal state resistance. The lattice schematics show the polarization of the wavefunction to either the top layer of the rhombohedral graphene in the yellow band isolated regime, or to both the top and bottom layers in the purple band overlap regime. **c**, Plot of the maximum T_c for all measured values of $B_{||}$, denoted $T_{c,max}$, as a function of V_b . The legend shows at which $B_{||}$ each value of V_b has its maximal T_c . **d**, Lower bound estimate of the Pauli limit violation as a function of V_b , as calculated by $B_{||c} / (1.25 \frac{k_B T_{c,max}}{\mu_B})$. The vertical red dashed line denotes a Pauli violation ratio of 1.

3.1.4 Pauli Limit Violation

Figure. 3.3 shows a series of 2D gate maps taken at different $B_{||}$. We see that the extended superconducting state reaches its maximum extent in the $V_b - V_t$ plane at $B_{||} = 1$ T. Beyond this value of $B_{||}$, superconductivity is suppressed at specific places in the 2D gate map. At a maximal $B_{||} = 6$ T (maximal field available with the cryostat), a sizeable portion of the superconducting state survives at large negative V_b .

Having established that $B_{||}$ induces superconductivity, and superconductivity survives in large $B_{||}$, We quantify the degree of Pauli limit violation systematically across the superconducting river. The Pauli limit $B_P = 1.25 k_B T_c / \mu_B$ provides a benchmark: for a

conventional singlet superconductor, the upper critical in-plane field should not substantially exceed B_P . We find instead that the critical in-plane field $B_{\parallel c}$ exceeds B_P at every gate position examined, with the violation ratio $B_{\parallel c}/B_P$ reaching more than an order of magnitude at several positions along the extended superconducting state (Fig. 3.4 d). We note that the concept of a Pauli limit is formally ill-defined for field-induced superconductivity, since $T_c = 0$ at $B = 0$; to make a conservative comparison we take the maximum T_c over all $B_{\parallel}$ as the relevant scale (Fig. 3.4 c). Even so, the violation ratio remains large throughout the phase diagram, providing strong evidence that the pairing is not spin-singlet in character.

Importantly, the superconducting state is highly sensitive to $B_{\perp}$ (See Appendix A C.6) even while being robust against $B_{\parallel}$. The strong $B_{\perp}$ sensitivity indicates that orbital depairing by the out-of-plane field is the dominant mechanism eventually suppressing the superconductivity, rather than paramagnetic pair-breaking from the Zeeman coupling. This anisotropy: robust against in-plane field, fragile against out-of-plane field, is a hallmark of quasi-two-dimensional spin-triplet superconductivity.

3.1.5 Role of layer dependent charge distribution

As shown in Figs. 3.1d, e, two zero-field superconducting pockets (SC1 and SC2 [28]) merge into a single elongated strip as $B_{\parallel}$ increases, forming a “river” in the (V_b, V_t) plane. The evolution along this river appears to be governed by the layer-dependent charge distribution, an important degree of freedom in rhombohedral multilayers. For small external interlayer potential differences (set by the voltage difference between the two gates), carriers from different bands polarize to opposite crystal surfaces, yielding a strongly interacting semimetal [28]. By contrast, for large interlayer potential differences, carriers are confined to either the valence or conduction band and localize exponentially on one surface. In our device, the superconductivity is modulated by this spatial charge configuration.

In the single-surface regime ($V_b > 2.8$ V), insulating states and the sharp resistive ridge associated with superconductivity follow diagonal trajectories because both gates tune the conduction band. For $V_b < 2.8$ V, where charge resides on both surfaces, gate-screening effects cause features to lock to trajectories controlled primarily by a single

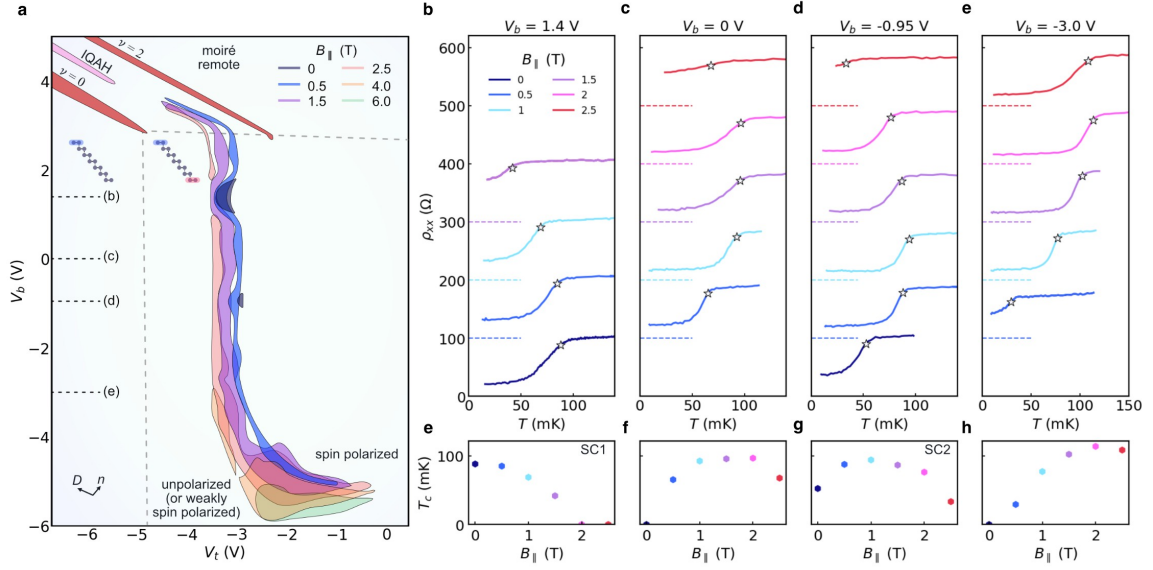


Figure 3.5: Non-monotonic evolution of T_c with $B_{||}$. **a**, Schematic phase diagram denoting the regions of the (V_b, V_t) plane exhibiting superconductivity at various selected $B_{||}$. Red (pink) regions denote trivial (Chern) insulating states. **b**, ρ_{xx} versus T measurements at several selected values of $B_{||}$ with $B_{\perp} = 0$. The measurement is taken with $V_b = 1.4$ V and optimal V_t . Curves are vertically offset for clarity. Dashed lines denote $\rho_{xx} = 0$ for each curve of corresponding color. Stars denote T_c . **c**, Same for $V_b = 0$ V. **d**, Same for $V_b = -0.95$ V. **e**, Same for $V_b = -3.0$ V. **f-i**, T_c versus $B_{||}$ for each of the four associated values of V_b .

gate [51]. Figure 3.5a marks the key features—insulating states and superconducting regions—at selected $B_{||}$ (see also Fig. 3.3). The field-induced superconductivity spans distinct charge configurations. For $V_b > 2.8$ V it corresponds to conduction band carriers on a single surface. As V_b is reduced, valence band carriers populate the opposite surface and screen the bottom gate. For $V_b < -4.2$ V the superconductivity bends away from fixed V_t , possibly indicating a reversal of the internal electric field and a switch of the surface hosting conduction-band states.

3.1.6 Non-Monotonic Evolution of T_c with $B_{||}$

The evolution of T_c with $B_{||}$ is complicated and depends sensitively on position along the river. In SC1, which is already present at $B = 0$, T_c decreases monotonically with $B_{||}$

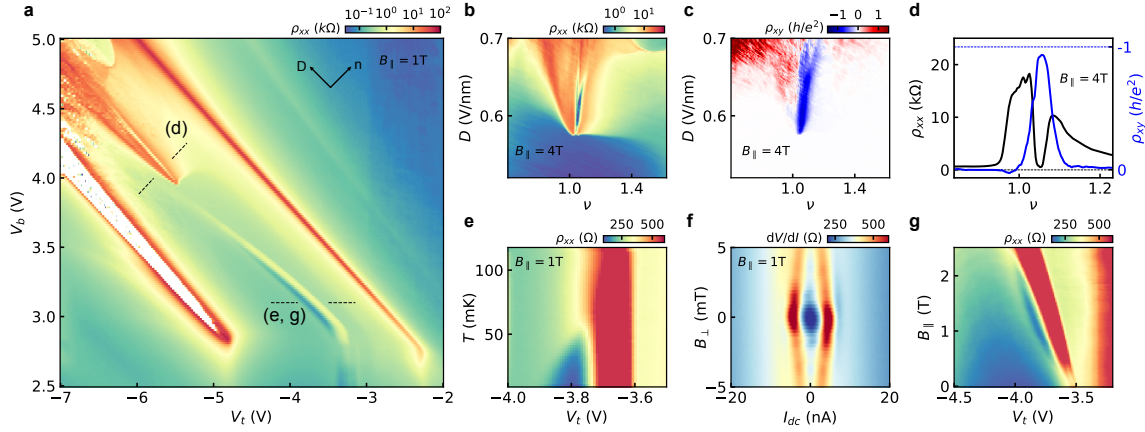


Figure 3.6: Field-induced superconductivity nearby an integer quantum anomalous Hall state. **a**, Zoomed-in map of ρ_{xx} versus V_b and V_t at $B_{||} = 1.0$ T. **b**, Zoomed-in map of the IQAH state near $\nu = 1$, showing ρ_{xx} versus ν and D at $B_{||} = 4.0$ T. **c**, Same as **(b)**, showing ρ_{xy} . **d**, Linecuts taken from the data shown in **(b)** and **(c)**, taken at the position shown by the tilted dashed line in **(a)**. **e**, ρ_{xx} versus V_t and T taken at fixed $V_b = 3.1$ V and $B_{||} = 1$ T (along the trajectory denoted in **(a)**). **f**, dV/dI versus I_{dc} and $B_{\perp}$ taken at $V_b = 3.0$ V, $V_t = -3.6$ V and $B_{||} = 1.0$ T. **g**, ρ_{xx} versus V_t and $B_{||}$ taken at fixed $V_b = 3.1$ V.

(Fig. 3.5b). The behavior differs markedly at gate voltages that instead host a resistive bump at $B = 0$. In such cases, T_c grows slowly with $B_{||}$ once superconductivity first appears with field, peaks at several tesla, and then vanishes at higher fields (Figs. 3.5c,e; Extended Data Fig. C.3). SC2 shows a weaker version of this trend (Fig. 3.5d). The gradual rise of T_c outside of SC1 is inconsistent with a simple picture in which superconductivity becomes strongest immediately after a competing order is suppressed; instead it points to a more delicate balance among the mechanisms responsible for determining its strength.

3.1.7 Superconductivity Adjacent to the IQAH State

We next analyze transport in the regime where a sufficiently large displacement field D polarizes conduction-band carriers to a single crystal surface, with no coexisting valence-band carriers (i.e., $V_b > 2.8$ V). Figure 3.6a shows a zoomed-in map of this region at $B_{||} = 1$ T. Near the bottom of the map, superconductivity appears around $V_t = -3.3$ V and is nearly independent of V_b . Once the valence band is emptied, however, the superconducting feature bends to follow nearby insulating states along diagonal

trajectories of nearly constant carrier density. In this single-surface regime, a fragile low-resistance pocket forms adjacent to the resistive bump. Both features shift leftward with increasing $B_{\parallel}$, and the superconducting pocket extends over a field range of approximately 0.5 – 2 T (Figs. 3.6e–g).

As D is increased further—up and to the left in Fig. 3.6a from the gate voltages where superconductivity appears—the system undergoes an abrupt transition to an integer quantum anomalous Hall (IQAH) state, likely corresponding to complete spin and valley polarization. The IQAH state is robust to in-plane field, with near-zero ρ_{xx} and ρ_{xy} approaching h/e^2 even at $B_{\parallel} = 4$ T (Figs. 3.6b–d). Interestingly, the data at 4 T show a reversed sign of the Chern number compared to those at $B_{\parallel} = 0$ (see Extended Data Fig. C.8), similar to recent reports in moiré R6G [64] and twisted bilayer-trilayer graphene [65]. Although the superconducting and IQAH regions approach one another closely in gate space, they do not directly connect. The superconducting pocket exhibits nonreciprocal behavior at scattered locations along the river (see Fig. 3.6f and Extended Data Fig. C.5). Specifically, there is an asymmetry in the critical currents in the forward and backward directions, $I_c^+ \neq I_c^-$, when applying a non-zero $B_{\perp}$; this behavior may arise from extrinsic factors [69, 70] or may be an indication of a paired state that intrinsically breaks time-reversal and inversion symmetries.

3.2 Discussion

3.2.1 Evidence for Spin-Triplet Pairing

The combination of observations presented in this chapter points strongly to spin-triplet pairing as the character of the superconducting order parameter in moiré R7G. The primary evidence is the large Pauli limit violation: the superconductivity is induced and stabilized by $B_{\parallel}$, persisting to fields more than an order of magnitude above the weak-coupling Pauli limit at all gate positions examined. For spin-singlet pairs, the Zeeman coupling to the two electrons in a pair acts in opposite directions, producing a net pair-breaking effect that limits $B_{\parallel c}$ to values near B_P [1]. For spin-triplet pairs with $S_z = \pm 1$ or $S_z = 0$, but with the net spin aligned with the external field direction, the Zeeman coupling acts identically on both electrons and constitutes no pair-breaking perturbation. The eventual suppression of superconductivity at large $B_{\parallel}$ in our data is

therefore most naturally attributed to orbital depairing — the coupling of the field to the spatial wavefunction of the Cooper pair — which is expected to cut off even spin-triplet superconductivity once the magnetic length becomes comparable to the coherence length.

The adjacency of the superconducting phase to the spin-polarized half-metal provides additional evidence for triplet pairing. As established by quantum oscillation measurements and the field-dependence of the phase boundary, the resistive ridge separates a spin-polarized phase from a weakly polarized phase. Superconductivity appears at and near this boundary. This is precisely the region in phase space, in which spin-triplet pairing is natural: fluctuations of the adjacent spin-polarized phase provide a channel for spin-triplet pair correlations, analogous to the role of ferromagnetic fluctuations in mediating spin-triplet pairing in other correlated systems.

3.2.2 Normal State Structure: Easy-Axis Versus Easy-Plane Spin Polarization

The evolution of superconductivity with $B_{\parallel}$ depends strongly on the position along the river in the (V_b, V_t) plane. In the band-overlapping regime, the river follows approximately a contour of fixed V_t , suggesting that the onset of superconductivity is tied to the conduction-band Fermi surface, which is controlled mainly by the top gate. At the same time, the strong variation of the superconducting properties as one moves along the river by changing V_b indicates that the pairing strength is not determined by the conduction-band Fermi surface alone, but is also influenced by changes elsewhere in the electronic structure. In particular, evolution of the valence-band Fermi surface on the opposite crystal surface with V_b can modify screening of long-range Coulomb interactions and thereby the balance among Hund's coupling, spin-orbit coupling, and pairing tendencies. This is consistent with abrupt Fermi surface reconstruction observed in Figure 2.8

A natural microscopic framework for this gate-dependent behavior is provided by the competition between Kane–Mele spin-orbit coupling and Hund's interaction. In a predominantly layer-polarized band, Kane–Mele SOC favors opposite out-of-plane spin orientations in the two valleys through spin-valley locking, whereas Hund's interaction favors collinear spin alignment [71]. A spin-polarized phase resulting from Stoner-type ferromagnetism may therefore adopt either an out-of-plane (easy-axis) or in-plane

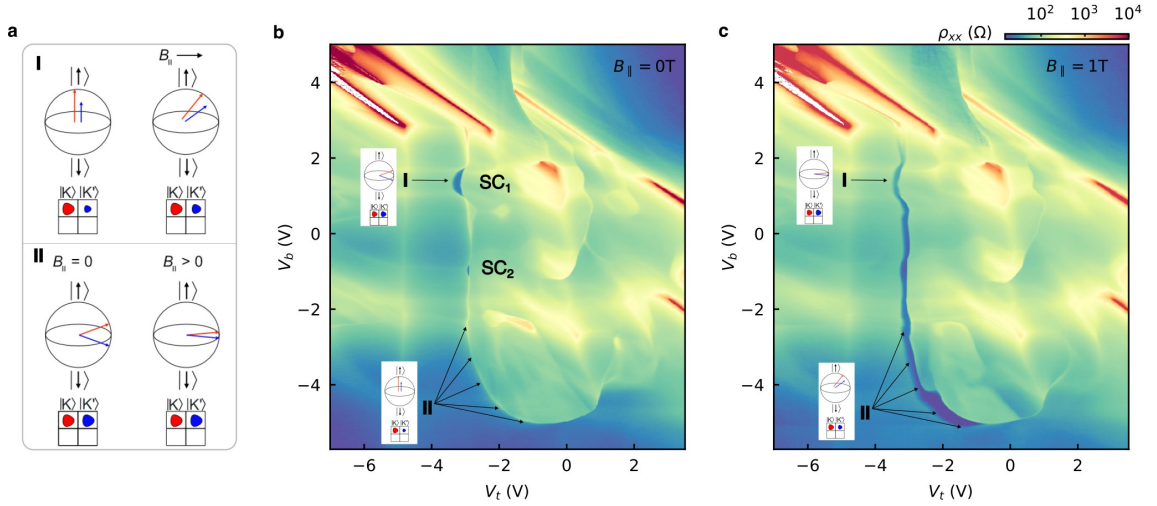


Figure 3.7: Phenomenological model. **a**, Bloch spheres show two possible spin-polarized states and their evolution with $B_{\parallel}$; the red and blue arrows respectively represent the spin polarization in valleys K and K' . Fermi surfaces associated with the different valleys are depicted in red and blue in the Punnett square diagrams. The top half of the inset depicts an easy-axis state (I) with weak valley imbalance, and the bottom half shows a valley-balanced spin canted state (II). **b**, Map of ρ_{xx} versus V_b and V_t at zero field. According to the phenomenological model SC_1 corresponds to the easy-plane state and everywhere else on the sharp resistive feature corresponds to the easy axis state. **c**, Same map at $B_{\parallel} = 1$ T. With $B_{\parallel}$ the easy axis state starts to cant in-plane and valley balance is restored. As a result we see the onset of superconductivity.

(easy-plane) configuration (Fig. 3.7 a), depending on how the spin anisotropy from Kane–Mele SOC competes with Hund’s coupling and the Coulomb interaction. Although the intrinsic SOC of graphene is relatively small (≈ 25 to $100 \mu\text{eV}$ [31, 72, 73, 74]), it is comparable to the superconducting energy scales relevant here, with $\Delta \sim 15 \mu\text{eV}$ for a BCS state with $T_c \sim 100$ mK. This competition is therefore a plausible ingredient in determining both the character of the parent spin-polarized state and the conditions for superconductivity (see Supplementary Information of ref. [75] for more details).

A key difference between the easy-axis and easy-plane states is that the former naturally produces valley imbalance, making it less compatible with superconductivity. In the easy-axis configuration, Kane–Mele SOC favors opposite out-of-plane spin orientations in the two valleys, while Hund’s coupling tends to align them. The resulting

compromise makes the two valleys energetically inequivalent, driving charge transfer between them and producing a net valley imbalance. This effect tends to suppress superconductivity due to the loss of Kramers degeneracy between the valleys, which undermines the weak-coupling instability for zero-momentum pairing. Applying $B_{\parallel}$ rotates the spins toward the plane and reduces the valley imbalance (Fig. 3.7 a), thereby reducing this source of pair breaking. This mechanism can account for the initial strengthening of superconductivity with $B_{\parallel}$. Its eventual suppression at larger fields, by contrast, is more naturally attributed to orbital depairing than to Zeeman pair breaking, since superconductivity survives far beyond the Pauli limit [47]. By contrast, an easy-plane spin-polarized state adopts a “spin-canted” configuration that is already valley balanced at $B_{\parallel} = 0$ [46, 76, 77]. Increasing $B_{\parallel}$ therefore does not provide a natural pathway to enhance pairing—it is expected to simply suppress T_c through orbital depairing (and possibly through its effect on the pairing glue, if provided by magnons tied to the neighboring spin-polarized state [77, 78]).

Within this framework, our data are naturally explained if the normal state from which superconductivity forms is predominantly easy-axis along much of the resistive stripe, but favors an easy-plane configuration near SC1 and possibly SC2 (Fig. 3.7 b). In this picture, the easy-axis tendency gives rise to a narrow valley-imbalanced region at the boundary between the spin-polarized metal on the right and the unpolarized metal on the left. A small $B_{\parallel}$ rotates the spins toward the plane, reduces the valley imbalance, and thereby enables superconductivity to develop along the entire stripe (Fig. 3.7 c). In the same spirit, the nonlinear dV/dI observed at low $B_{\parallel}$, before superconductivity sets in, may reflect current-driven motion or reconfiguration of domain walls in a valley-imbalanced state (Fig. 3.2e), although other explanations remain possible. The superconducting diode effect observed for some gate settings may provide a further indication of residual valley imbalance, since such an imbalance would break both inversion and time-reversal symmetries. In that case, $B_{\perp}$ may enable diodic behavior by aligning valley-imbalanced domains. Consistent with this picture, the absence of a diode effect in SC1 (Appendix C Fig. C.5f,g), together with its monotonically decreasing $T_c(B_{\parallel})$ (Fig. 3.5b,f), may point to a valley-balanced parent state at that location.

We emphasize that this interpretation is not unique. While the scenario discussed above provides a natural explanation for many of the salient features of the data, other

symmetry-broken states may also be compatible with the phenomenology of the resistive stripe, including intervalley-coherent or nematic orders, possibly coexisting with spin polarization. Because our transport measurements are not directly sensitive to these distinctions, further experiments will be needed to determine the nature of the $B = 0$ state underlying the stripe and to clarify the superconducting order. Particularly promising directions include direct probes of orbital magnetization using SQUID magnetometry [46, 31], compressibility mapping of thermodynamic phase boundaries versus $B_{\perp}$ and $B_{\parallel}$ [31], and experiments with proximal TMD layers to tune the strength of SOC directly [79, 48, 49, 50, 80, 81, 47, 46]

3.2.3 *Comparison with Related Systems*

The phenomenology observed here shares qualitative features with several other graphene platforms, suggesting that spin-triplet superconductivity may be a generic property of rhombohedral graphene over a wide range of layer numbers. In Bernal bilayer graphene (BBG), a sharp resistive feature with nonlinear transport at $B = 0$ develops into superconductivity upon application of a small in-plane field [47], closely analogous to the behavior we observe in R7G. The origins of these two phenomena may be related, although the surface-bifurcated low-energy states of thicker rhombohedral graphene introduce an additional degree of freedom, that substantially reshapes the electronic screening and may alter the pairing mechanism.

Rhombohedral trilayer graphene hosts two distinct superconducting regimes, one of which exhibits critical fields well beyond the Pauli limit and is consistent with spin-triplet, valley-singlet pairing [25]. In that system, as in ours, the superconductivity appears at the boundary of a spin-polarized phase, and the $B_{\parallel}$ response provides evidence against a singlet order parameter. More recently, signatures of chiral superconductivity carrying both spin and valley polarization have been reported in thicker rhombohedral systems [82], raising the possibility that the order parameter character evolves with layer number. Our results are consistent with spin-triplet (valley-singlet) pairing, though a definitive identification of the valley structure of the order parameter awaits phase-sensitive measurements.

The moiré potential in our device has at least two clear effects on the spin physics.

First, it suppresses the correlated state at $n = D = 0$ seen in misaligned rhombohedral graphene, which in its absence interrupts the resistive ridge hosting superconductivity. In the aligned R7G device, the moiré potential removes this obstruction and allows the superconducting river to extend without interruption. Second, the moiré potential is required to stabilize the IQAH state that appears at higher displacement field. The spin-triplet superconductivity itself, however, does not require the moiré potential: similar behavior is observed in misaligned R8G (Chapter 3) and has been reported in misaligned systems at other layer numbers [83, 84]. The moiré pattern therefore acts as a clarifying element of the phase diagram rather than a necessary ingredient for the superconducting instability itself.

3.3 Conclusions

We have presented a comprehensive investigation of the spin structure of the superconducting phases in moiré R7G. The key results are as follows:

In-plane magnetic fields induce and dramatically expand superconductivity, converting a resistive ridge into an extended superconducting river spanning a wide region of the gate-voltage plane. The superconductivity persists to $B_{\parallel}$ far exceeding the Pauli paramagnetic limit at all gate positions examined, with violations exceeding an order of magnitude in several cases. Together with the adjacency of the superconducting phase to a spin-polarized half-metal, these observations strongly suggest spin-triplet pairing as the character of the order parameter.

The critical temperature evolves non-monotonically with $B_{\parallel}$ at gate positions away from the SC_1 pocket, growing slowly as superconductivity first appears, peaking at several tesla, and then declining. This behavior is consistent with a scenario in which $B_{\parallel}$ rotates out-of-plane spins into the plane, reducing a pair-breaking valley imbalance and enabling the pairing to strengthen, before orbital depairing at larger fields ultimately suppresses T_c .

A phenomenological model incorporating Hund's coupling and Kane-Mele spin-orbit coupling can explain the main qualitative features of the phase diagram, including the competition between easy-axis and easy-plane spin configurations, the field-driven restoration of valley balance, and the associated emergence of superconductivity with $B_{\parallel}$.

These findings establish spin-triplet, field-induced superconductivity as a pervasive and robust feature of rhombohedral graphene, and motivate further experiments to determine the precise valley and orbital structure of the order parameter. Looking forward, the close proximity of spin-triplet superconductivity and the IQAH state in the R7G phase diagram provides a natural platform for engineering topological superconductivity and potentially realizing non-Abelian excitations through electrostatic control.

Chapter 4

HIGH PRESSURE TRANSPORT MEASUREMENTS OF VDW HETEROSTRUCTURES

As illustrated in the previous chapters for rhombohedral graphene, we can use a diverse set of tuning knobs to study 2D materials, including charge carrier density n , displacement field D , out-of-plane and in-plane magnetic fields $B_{\perp}$ and $B_{\parallel}$, as well as temperature T . Hydrostatic pressure is yet another tuning parameter that can be added to this extensive list. In layered materials, where interlayer interactions play such a crucial role (as they do in rhombohedral graphene) in determining the physics, pressure provides a straightforward pathway to enhance these interactions. Hydrostatic pressure is routinely used as a tool to study transport in bulk materials. However, its adaptation for transport studies of 2D materials has been impeded by specific technical challenges. In this chapter, we dive into the technical details of this experiment and outline the biggest challenges slowing down progress. We also present several solutions we have developed to address these challenges and illustrate the workflow we have established to make this process more reliable. Finally, we present preliminary data from an ongoing project in which we are measuring rhombohedral graphene under pressure.

4.1 Pressure as a tuning knob to study 2D materials

Hydrostatic pressure has a long and distinguished history as a tuning parameter in condensed matter physics. Unlike chemical substitution or electrostatic gating, which alter the carrier density or introduce disorder, hydrostatic pressure acts as a clean mechanical perturbation that reduces the unit cell volume while preserving the crystalline symmetry of the material. By compressing interatomic distances, pressure modifies the orbital overlap between neighboring atoms, directly tuning the electronic bandwidth and hopping amplitudes without introducing extrinsic disorder. This makes it a particularly powerful probe of the relationship between band-structure and correlated ground states. In bulk correlated materials, pressure has been used to drive metal–insulator transitions

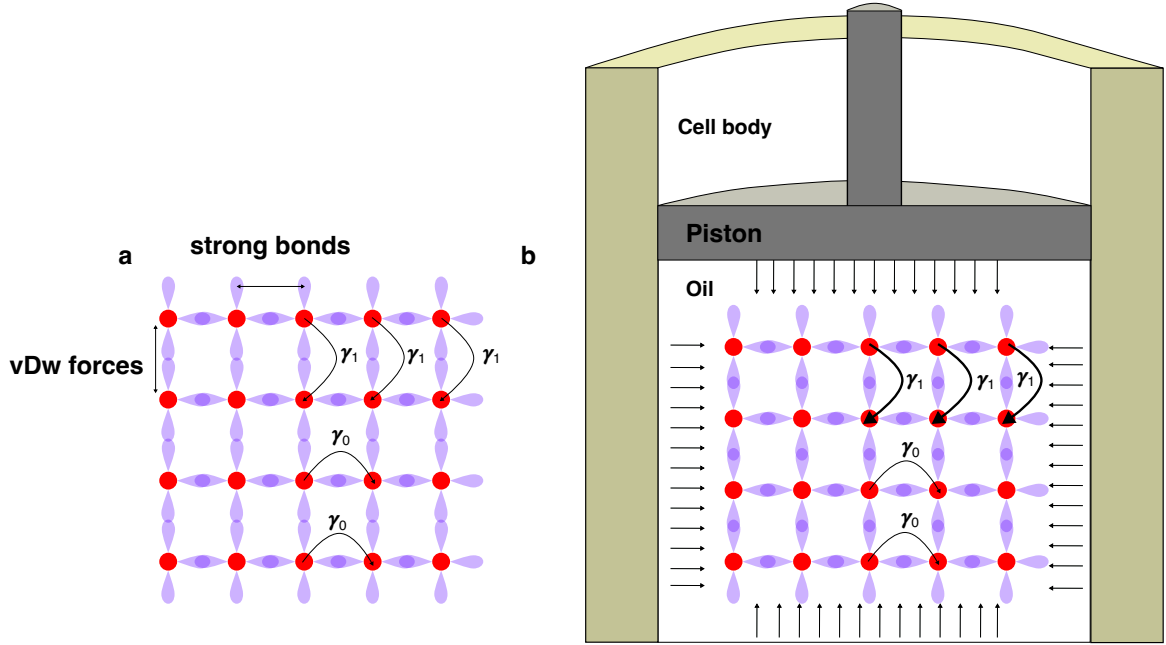


Figure 4.1: **2D materials under pressure.** **a,b**, Illustration of the effect of hydrostatic pressure on layered 2D materials. Because adjacent layers are held together by relatively weak van der Waals (vdW) interactions, the interlayer compression is significantly larger than the in-plane compression. Consequently, pressure produces an anisotropic enhancement of the interlayer coupling (γ_1) in the figure).

[85], suppress magnetic order and reveal superconducting domes in heavy-fermion compounds [86, 87], and tune the interplay between antiferromagnetism and superconductivity in unconventional superconductors through a single continuous control parameter [87]. The ability to continuously vary the bandwidth of a correlated system while keeping its chemical composition and disorder fixed is a capability that few other experimental knobs can match.

Naturally, studying 2D materials under pressure is an attractive proposition. Interlayer interactions can lead to emergent electronic properties fundamentally different from the constituent layers. However these interlayer interactions are difficult to determine and control in a systematic manner. Hydrostatic pressure gives us a pathway to control these interaction in a dynamic manner. 2D materials have strong in-plane covalent

bonds but weak out of plane vdW attractive forces. As a result of this asymmetry, such systems under pressure have their interlayer spacing preferentially compressed over their much stronger intralayer bonds. This reduced interlayer spacing leads to a much larger overlap of the electronic orbitals resulting in enhanced electronic interlayer interactions as seen in Figure 4.1. This allows for the dynamic tailoring of the band structure in a given sample while controlling for disorder. In the case of twisted bilayer graphene theoretical studies have predicted that this can allow for the flattening of the bands with pressure, making it analogous to twist angle [21]. Beyond restructuring the electronic bands, theoretical studies have also predicted that pressure can have much more subtle effects where it can tip the balance between competing many body states like Fractional Chern Insulators, topological charge density waves and Wigner crystal states [88].

Despite its demonstrated power in bulk correlated materials, high pressure transport has remained a relatively underexplored tool in the study of atomically thin van der Waals systems. The most prominent example to date is the work of Yankowitz et al. [20], who used a piston-cylinder hydrostatic pressure cell to perform transport measurements on twisted bilayer graphene devices, demonstrating that pressure could induce superconductivity in a device with a twist angle of 1.27° , above the magic angle where correlated phases are otherwise absent at ambient pressure. By reducing the interlayer spacing, pressure effectively mimics the effect of reducing the twist angle, enhancing the interlayer coupling and flattening the bands. This established pressure as a means of tuning the bandwidth of a moiré system independently of its twist angle, and demonstrated that high quality four-terminal transport measurements on encapsulated van der Waals heterostructures could survive the application of hydrostatic pressure. Beyond twisted bilayer graphene, pressure has also been used to tune the moiré potential strength in graphene/hBN heterostructures [19]. However, these studies remain few in number, and the application of hydrostatic pressure to the study of correlated phases in van der Waals heterostructures — particularly in combination with independent electrostatic control of carrier density and displacement field remains technically challenging and largely unexplored. To appreciate these challenges it is worth understanding the experimental mechanism involved in carrying out these high pressure transport experiments.

4.2 *Experimental technique*

4.2.1 *Piston cylinder cell: Assembly and working*

High pressure experiments can be carried out using two broad approaches. In a diamond anvil cell (DAC), the sample is placed between the opposing faces of two diamonds and pressure is applied by driving them together. DACs are capable of reaching pressures of several hundred GPa, making them the instrument of choice for extreme pressure studies. However, this capability comes at a cost: the sample must be fabricated directly onto the diamond face, which is incompatible with the established van der Waals heterostructure fabrication workflow that has been optimized for flat SiO_2/Si substrates. Adapting this workflow to diamond substrates is non-trivial and introduces additional complexity that can compromise device quality. A piston cylinder cell, by contrast, compresses a deformable container filled with pressure medium surrounding the sample via metallic pistons, allowing the device to be fabricated using standard techniques before being loaded into the cell. The maximum achievable pressure is lower, typically up to ~ 3 GPa, but this range is well suited to the study of van der Waals heterostructures: the interlayer spacings in these materials are highly compressible, and 3 GPa is sufficient to produce substantial changes in the electronic bandwidth and phase diagram [21, 20]. For the purposes of this work, the ability to preserve device quality and fabrication fidelity outweighs the pressure range limitation of the piston cylinder approach, and we adopt it as our primary experimental platform.

The piston cylinder cell has been used as an experimental tool in fundamental science and geology since the nineteenth century, but modern variants have benefited from decades of refinement in both design and materials. For the purposes of this work, we use the commercially available Almax-easyLab Pcell 30 (Shown in Fig. 4.2). The cell body has a double-walled construction: an outer cylinder machined from beryllium copper (BeCu) and an inner cylinder from a proprietary high-strength alloy. Both materials are hard and paramagnetic, making them simultaneously capable of withstanding the large mechanical forces associated with high pressure and non-invasive as backgrounds for sensitive low-field transport measurements.

The sample assembly is built around an electrical feedthrough machined from MP35N, a non-magnetic cobalt-nickel superalloy with excellent mechanical properties at high

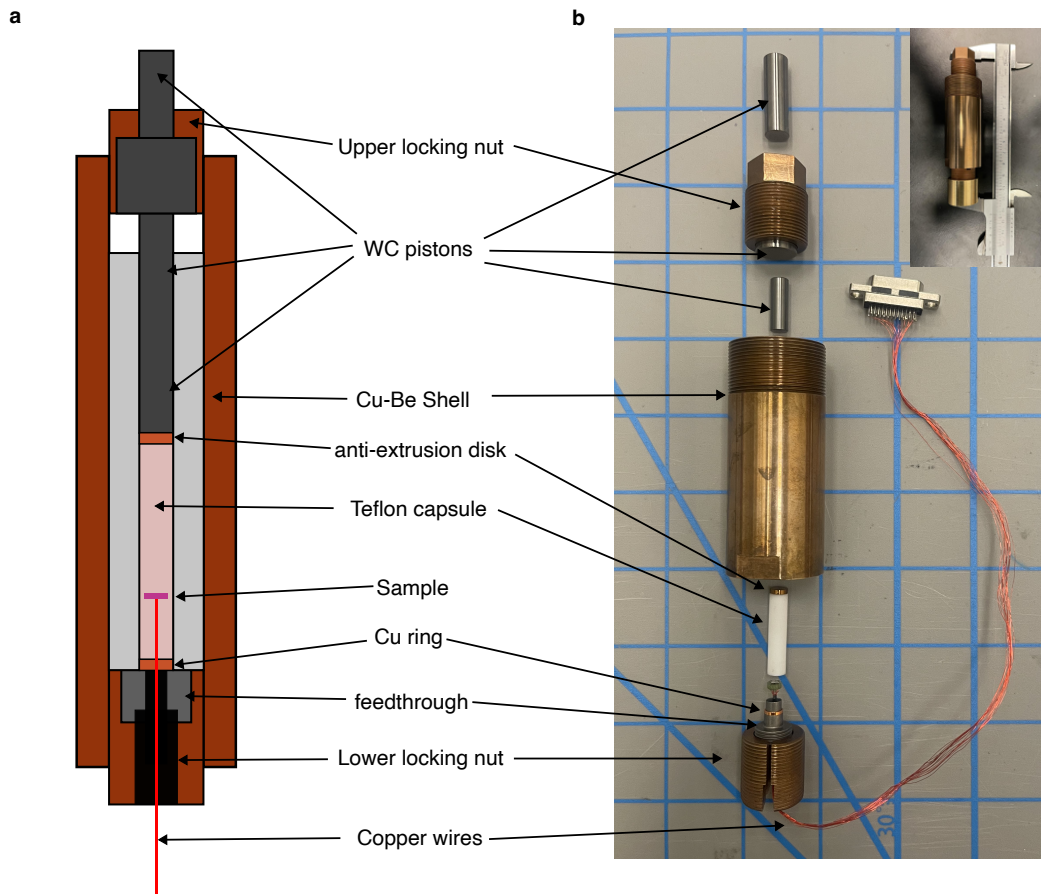


Figure 4.2: **Structure of a piston cylinder cell.** **a**, Schematic showing the cross-sectional view of a fully assembled piston cylinder cell with the different parts labeled. **b**, Image of a disassembled piston cylinder cell. (Inset) Image of a fully assembled piston cylinder cell.

pressure. The feedthrough has a cylindrical body with a narrower extrusion at one end, near which the device of interest is mounted. A hollow channel runs along the central axis of the feedthrough, through which the electrical leads to the sample are threaded; this channel is subsequently filled with epoxy to prevent the pressure medium from migrating along the wiring. The narrow end of the feedthrough is inserted into a PTFE capsule filled with Daphne oil, which serves as the pressure transmitting medium. Daphne oil remains hydrostatic over the pressure range accessible with the Pcell 30, ensuring that the pressure is transmitted uniformly to the sample without uniaxial stress components that could damage the device or introduce anisotropic strain. The loaded PTFE capsule is then inserted into the central bore of the pressure cell. A copper ring at the mouth of the PTFE capsule has a diameter slightly larger than the bore, and deforms plastically upon insertion to form a seal that prevents oil from leaking around the sides of the feedthrough.

The feedthrough is secured to the cell body by a locking nut with a central bore, which fastens the assembly while preserving electrical access to the sample wiring. On the opposing side of the cell, two tungsten carbide (WC) pistons of different diameters are loaded in series, with the smaller piston in direct contact with the PTFE capsule. A second locking nut, also with a central bore to allow pressurization, secures the pistons in place. With both locking nuts fastened, the pressure cell is fully assembled.

Pressurization is carried out under a hydraulic ram (Fig. 4.3). The assembled cell, together with a third WC piston placed against the outer locking nut but not fastened, is positioned under the ram. Force is applied slowly by lowering the ram, typically requiring several tons to reach pressures of the order of GPa. As pressure builds, the PTFE capsule compresses and the pistons advance into the bore, requiring the outer locking nut to be incrementally tightened to hold the pistons at each new position. Once the target force has been reached and the locking nut re-fastened, the ram is slowly retracted and the cell is removed. The locking nut then bears the mechanical load, maintaining the pressure on the sample without any external support. At this point the pressure cell can be loaded into a cryostat and transport measurements can be performed via the wires that come out of the cell.

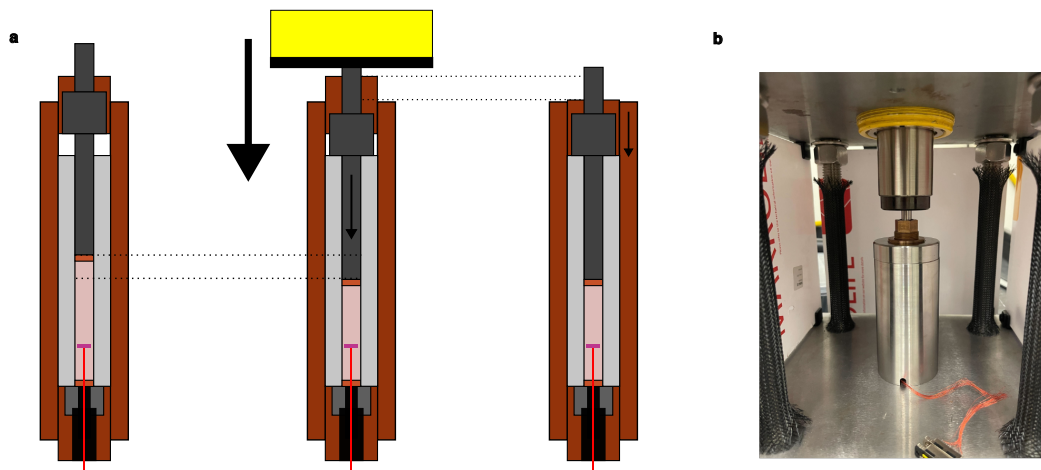


Figure 4.3: **Pressurization process.** **a**, cross-sectional view of the piston cylinder cell during the pressurization process. (Center) A hydraulic ram (in yellow) is used to apply several tons of force on the tungsten carbide (WC) pistons, this in turn compresses the Teflon capsule with the pressure transmitting medium. (Left) The locking not on top is screwed in further to hold the pistons in position as the teflon capsule is compressed. **b**, Image of the hydraulic ram as pushing down on the WC capsule.

4.2.2 Challenges

The primary challenge of adapting piston cylinder cell experiments to atomically thin van der Waals heterostructures stems from the combination of an extremely confined sample space and the demanding electrical requirements of 2D transport measurements. The sample space is kept intentionally small to maximize the pressure achievable within safe mechanical limits — the Almax-easyLab Pcell 30 has a bore radius of 6 mm, the largest among commercially available cells, but the usable sample area is further restricted to the radius of the narrow feedthrough extrusion, approximately 5 mm. Within this constrained geometry, establishing reliable electrical contacts is non-trivial. Standard wirebonding requires a stable mounting platform and access for the wirebonding tool head, neither of which is available within the feedthrough extrusion.

The number of electrical connections also presents a challenge specific to 2D heterostructure measurements. While a minimal transport measurement can in principle be carried out with as few as 8 wires, a thorough characterization of a 2D device requires more. Contacts can fail under pressure, develop non-ideal contact resistances, or prove

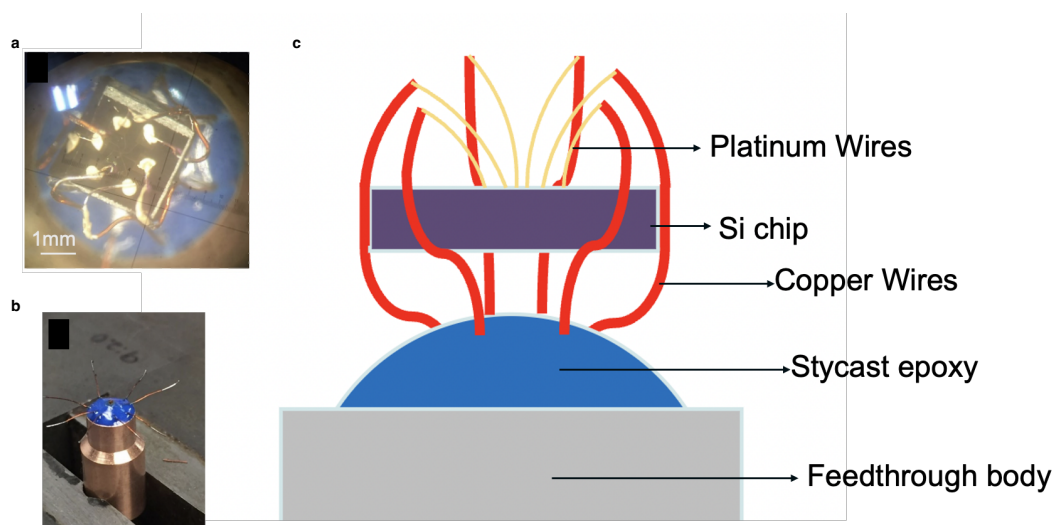


Figure 4.4: **Wiring configuration used in previous high pressure experiments.** **a** and **b**, Show images reproduced from ref. [19]. **c** is a side on view of the sample wiring configuration used in ref. [19]. Thick insulated copper wires are epoxied to the walls of the silicon die and form a cradle supporting the die. Thinner platinum wires are then connected to the copper wires on one end and $200 \times 200 \mu\text{m}$ gold pads connected to the sample on the other end.

unsuitable for a particular measurement geometry. Having a larger number of contacts provides redundancy and flexibility, allowing the measurement to continue productively even when individual contacts are compromised. This is particularly important in a pressure experiment, where the sample cannot be accessed or rewired once the cell is sealed and pressurized.

The approach used in prior high pressure transport studies of 2D materials addresses these challenges only partially. Thick insulated copper wires, once threaded through the feedthrough channel, are arranged into a “birds nest” structure within the sample space (Fig. 4.4 c). The device, mounted on a silicon die, is placed at the center of this structure with the copper wires epoxied to the die walls. Thinner platinum wires are then attached to the copper leads and hand-pasted onto $300 \times 300 \mu\text{m}$ gold contact pads on the die using conducting epoxy, as illustrated in Figure 4.4 a.

This approach suffers from several limitations that make it poorly suited to high-throughput 2D transport experiments. The device has no rigid mechanical support,

being suspended by the copper wires and subject to significant mechanical stress as pressure is applied. The platinum wires must be hand-pasted onto micron-scale gold pads, requiring considerable skill and patience, and the entire assembly is time consuming to prepare reliably. The stripped wire ends are in close proximity to each other, to the cell body, and to the device, creating a persistent risk of electrical shorts — both between wires and between the wiring and the silicon gate. Finally, the configuration practically limits the total number of wires to approximately 8, which as discussed above provides insufficient redundancy for a thorough characterization of a 2D heterostructure device.

4.2.3 Technical improvements

In this section we outline the technical improvements we have introduced to make high pressure transport measurements on van der Waals heterostructures more reliable and reproducible. We address the challenges described above in sequence, beginning at the level of the substrate and working up through the full assembly.

Pre-diced Substrates

A first point of failure in the conventional approach is the dicing of the silicon die to fit within the confined sample space of the feedthrough extrusion. This is typically done by hand using a diamond scribe, a process that is imprecise and risks damaging or destroying the device, particularly given the small final die size required. We eliminate this risk by transferring our van der Waals heterostructures onto pre-diced substrates. Silicon wafers are cut to approximately one third of their thickness in a regular grid of 2.3×2.3 mm squares using a dicing saw, leaving the wafer intact but pre-scored along all cut lines. Device fabrication is carried out on a 3×3 array of these squares, providing a handling area of approximately 7×7 mm that is comfortable to work with during lithography and contact deposition. Once fabrication is complete, the surrounding squares are cleanly snapped off along the pre-scored lines, confining the finished device and all gold bonding pads to a single 2.3×2.3 mm die. This approach replaces a manual and risk-prone dicing step with a clean mechanical snap that poses no risk to the device.

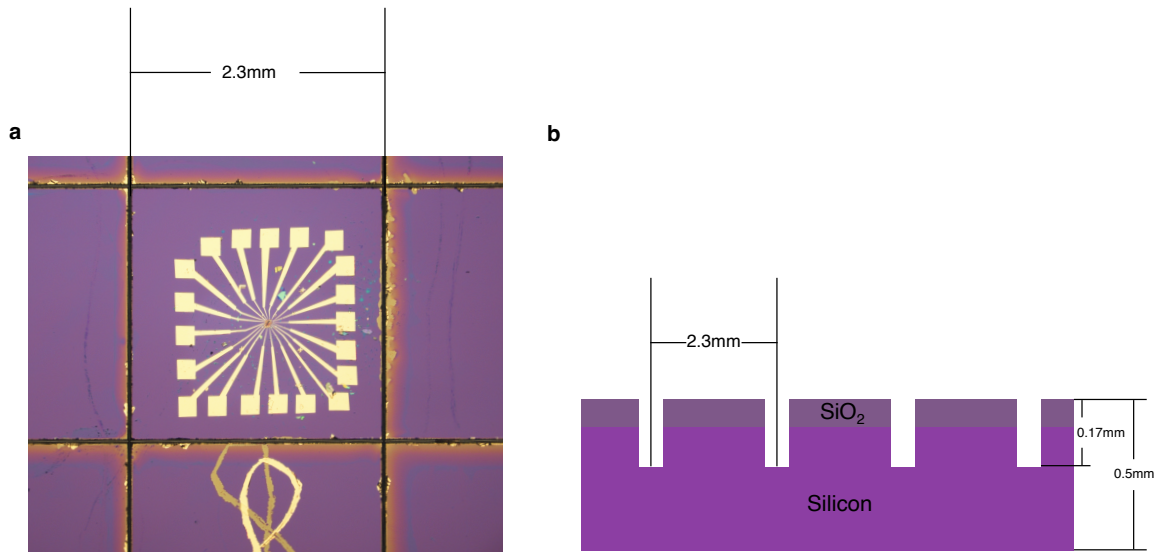


Figure 4.5: **Pre-diced substrate.** **a**, Micrograph showing the top view of a sample fabricated on a pre-diced silicon die. **b**, Schematic showing the cross-sectional view of the pre-diced substrate.

Improved wiring scheme

To address the dual challenges of wiring organization and mechanical stability, we designed a custom printed circuit board (PCB) sized to fit within the sample space of the feedthrough extrusion. The board is fabricated from FR4, a glass-reinforced epoxy laminate composite that is both mechanically rigid and electrically insulating, making it well suited to the confined and high-stress environment of the pressure cell. The top face of the PCB features 16 bond pads arranged near its perimeter, coated with electroless nickel electroless palladium immersion gold (ENEPIG) to provide a surface compatible with standard wirebonding. Each bond pad connects to a plated through-hole via an internal trace, allowing copper wires to be soldered to the reverse face of the board and routed cleanly through the feedthrough channel. A larger central through-hole anchors a tungsten rod, which is secured within the feedthrough cavity using epoxy and provides a rigid platform on which the silicon die is mounted.

This design offers several advantages over the birds nest approach. Sixteen electrical connections to the sample can be established reliably and reproducibly, providing the contact redundancy discussed above. Wirebonding to the ENEPIG pads is significantly

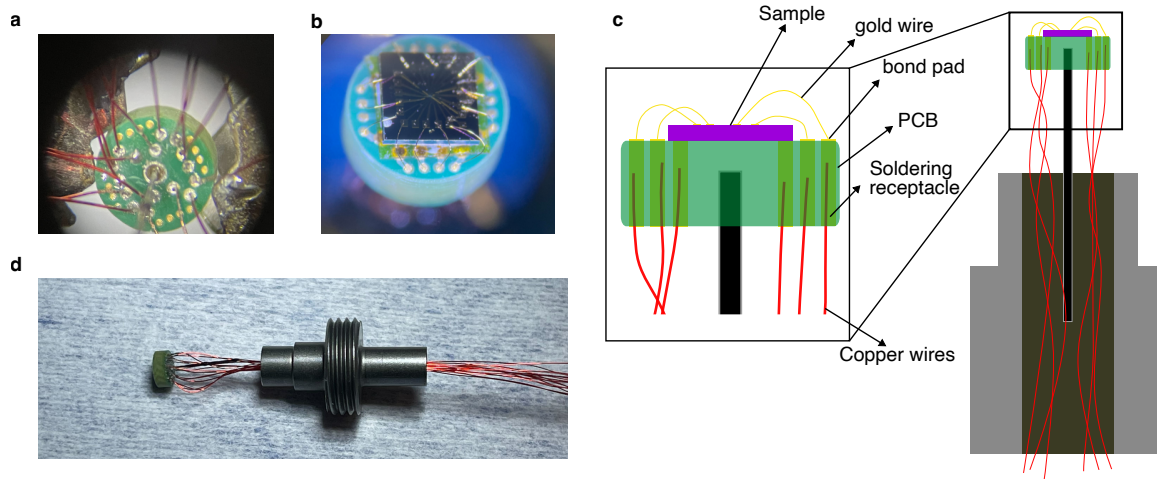


Figure 4.6: **New wiring scheme.** **a** and **b**, Images showing two sides of the custom designed PCB. In **(a)** copper wires can be soldered into the receptacles. In **(b)** the silicon die sits in the middle of the PCB and bond pads on the PCB can be connected to the sample via wirebonding. **c**, shows a cross sectional view of the wiring scheme. The PCB is anchored to a rigid tungsten rod which is buried in the epoxy. Zoom in shows how the PCB acts as a terminal for the different electrical connections. **d** Image showing the side-on view of the electrical feedthrough.

faster and more repeatable than hand-pasting platinum wires onto micron-scale gold contacts, improving throughput and reducing operator-dependent variability. The organized routing of wires through the plated through-holes eliminates the proximity-induced shorting risk of the birds nest configuration. Finally, the combination of the PCB and the tungsten rod provides a rigid mechanical platform for the device, replacing the compliant wire cradle of the conventional approach with a structure that is better able to withstand the mechanical stresses associated with pressurization.

Preparation of the feed through

With these improvements in place, we have developed a reliable standard operating procedure for preparing samples for pressurization. The order of operations is carefully chosen to minimize the risk of electrostatic discharge (ESD) damage to the device, which is particularly important given that the device cannot be accessed or replaced once the cell is sealed.

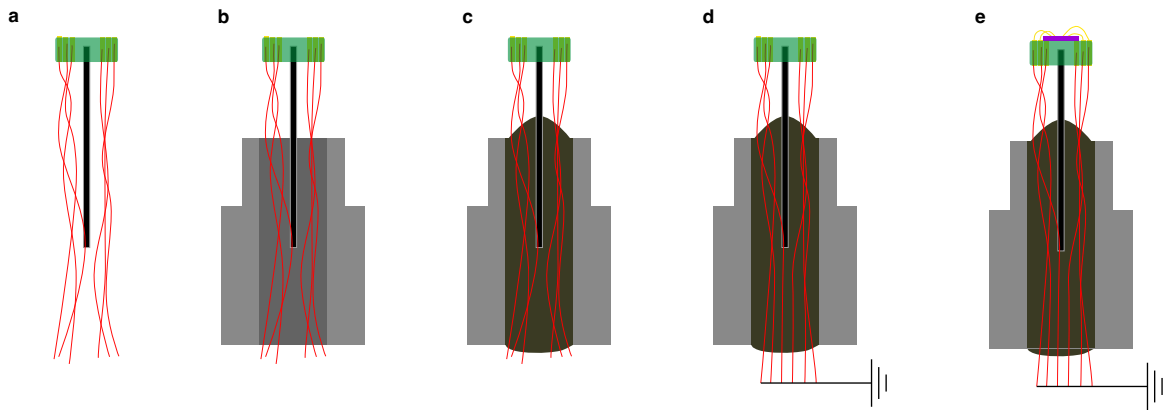


Figure 4.7: **Process of preparing the electrical feedthrough.** **a**, Shows copper wires and the tungsten rod are soldered to the PCB. **b**, This is then threaded through the cavity of the feedthrough. **c**, The cavity is then filled with epoxy to seal the feedthrough. **d**, Once the epoxy has set, the other end of the copper wires are soldered to a D-sub connector, which is then grounded. **e**, With the bond pads now grounded, the sample can be safely wirebonded.

Assembly begins with the mechanical and electrical preparation of the feedthrough, before the device is anywhere near the assembly. The tungsten rod is first attached to the PCB. Because the tungsten surface tends to trap air when immersed in epoxy, the rod is pre-coated with a thin layer of epoxy before insertion to prevent the formation of voids at the rod-epoxy interface. Sixteen copper wires are then soldered to the bond pads on the reverse face of the PCB and cleaned with methanol to improve adhesion with the epoxy that will subsequently fill the feedthrough cavity. The wires are threaded carefully through the central channel of the feedthrough. Care must be taken at this step not to pull the wires too hard, as excessive tension can break them before the assembly is complete.

Once all wires are threaded, the feedthrough cavity is filled with epoxy to seal the channel and prevent the pressure medium from migrating along the wiring. We use Loctite Stycast 2850FT with 24LV catalyst in a mass ratio of 100:7.5. After thorough mixing, the epoxy mixture is degassed in a desiccator for approximately 10 minutes to remove air bubbles, which if left in place would reduce the mechanical strength of the seal. The degassed epoxy is then worked into the cavity using a fine wooden applicator, coating the copper wires and the inner walls of the channel. The wires are moved up and down during application to ensure the epoxy penetrates between them and leaves no

voids. Filling continues until epoxy begins to emerge from the mouth of the cavity, at which point the channel is considered fully filled. The assembly is then left to cure at ambient conditions for 24 hours. It is important that the PCB sits flat and level during curing — any tilt will cause the epoxy to cure unevenly, leaving the PCB at an angle and preventing the silicon die from sitting flat on the board surface during subsequent sample mounting.

Once the feedthrough epoxy has fully cured, the free ends of the copper wires are soldered to a micro D-sub connector. The continuity of each connection is then tested by mapping the correspondence between each D-sub pin and its associated bond pad on the PCB. This mapping is recorded for reference during measurement. With the electrical connections to the bond pads now established, all bond pads are grounded through the D-sub connector before any further handling. This is a critical ESD precaution: grounding the bond pads ensures that any static charge accumulated during subsequent handling is safely dissipated rather than discharged through the device.

With the bond pads grounded, the sample can be mounted. A thin film of polyamide tape is first placed on the surface of the PCB in the central mounting area to electrically isolate the silicon die from any metallic components of the assembly, and is secured with a small amount of epoxy. The silicon die is then mounted at the center of the PCB using silver epoxy and baked at 110°C for 15 minutes to cure. Once the silver epoxy has set and the die is mechanically secured, the assembly is ready for wirebonding. Wirebonding proceeds in the standard fashion, connecting the gold contact pads on the device to the ENEPIG bond pads on the PCB perimeter.

4.2.4 *Pressure Calibration*

Any experiment that uses hydrostatic pressure as a tuning parameter must establish the actual pressure experienced by the sample, which is not directly accessible from the applied force alone. A first estimate can be obtained from the hydraulic ram: the ram is equipped with an oil pressure gauge, from which the applied force is calculated using the manufacturer's calibration formula. Dividing by the cross-sectional area of the bore then gives a nominal pressure via $P = F/A$. However this estimate is unreliable for several reasons.

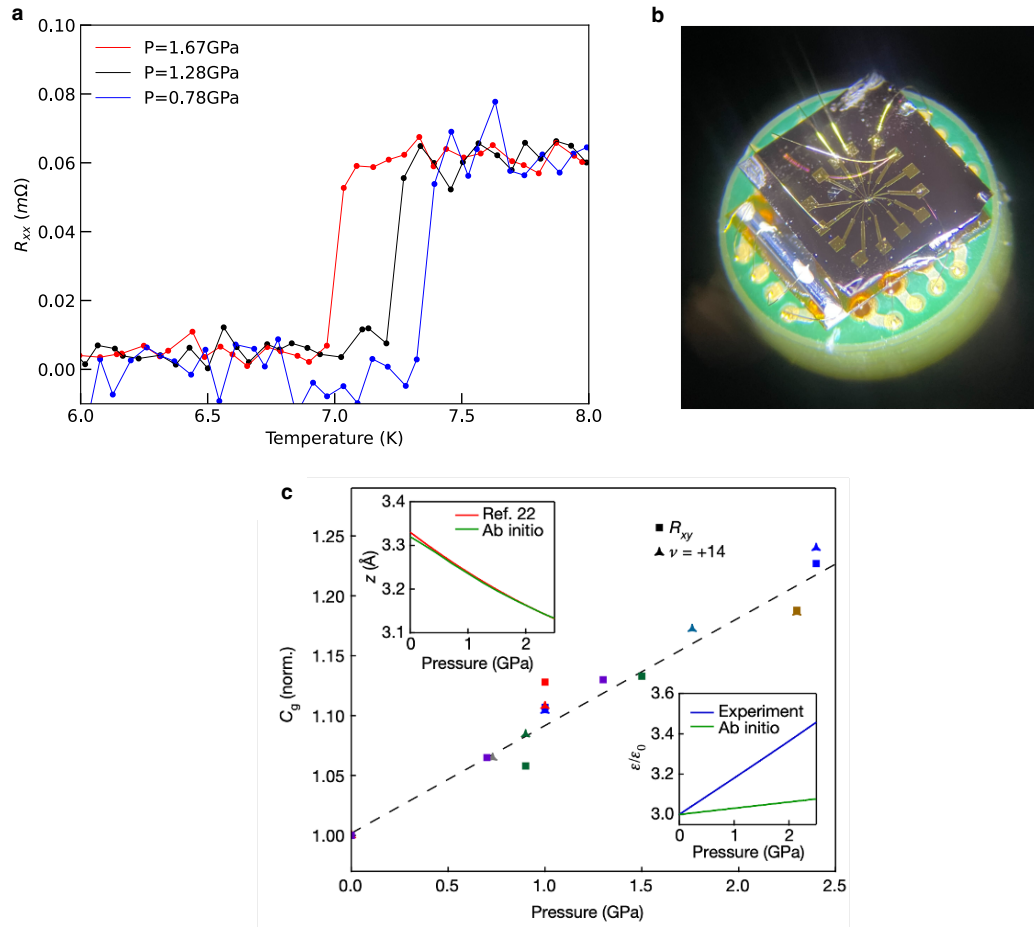


Figure 4.8: Calibrating the pressure inside the cell. **a**, Resistance (R) vs temperature (T) measurements of the lead (Pb) wire, taken at different pressures. The T_c decreases with increasing pressure. **b**, Image of the Pb wire connected to the pads on the PCB, placed next to the silicon die. **c**, Reproduced from [19], shows normalized gate capacitance C_g vs pressure, dashed line is a best fit. Top inset shows interlayer separation z vs pressure obtained via numerical simulations. Bottom inset shows how the dielectric constant changes with pressure also obtained via numerical simulations.

First, not all of the force applied by the ram is transmitted to the PTFE capsule — friction between the pistons and the bore wall and mechanical losses at each interface mean that the effective force on the sample is systematically lower than the nominal value. Second, the estimate assumes that Daphne oil is incompressible, whereas in reality its density increases under pressure, resulting in a small but non-negligible loss of pressure relative to the nominal estimate. Third, and most significantly, Daphne oil solidifies at approximately 200 K under pressures in the range accessible with the Pcell 30. As the pressure cell is cooled to the base temperature of the cryostat, the solidification of the pressure medium is accompanied by a substantial and non-linear reduction in the pressure transmitted to the sample. The pressure at base temperature can therefore differ considerably from the pressure applied at room temperature, and cannot be inferred from the room temperature force measurement alone. For these reasons it is essential to have an independent in-situ pressure sensor within the pressure cell that can track the pressure continuously as the cell is cooled.

Ruby fluorescence is commonly used to determine pressure in diamond anvil cells and piston-cylinder cells. Ruby consists of chromium ions (Cr^{3+}) doped into an aluminum oxide (Al_2O_3) lattice, and its fluorescence arises from electronic transitions of the Cr^{3+} ion between crystal-field-split energy levels. The R1 line corresponds to the transition from the lowest excited state (${}^2\text{E}$) to the ground state (${}^4\text{A}_2$). As pressure increases, the lattice compresses and the Cr^{3+} –O bond lengths shorten, strengthening the crystal field and lowering the energy of the ${}^2\text{E}$ state. This reduces the transition energy, causing the R1 line to redshift. The shift is nearly linear with pressure at a rate of approximately $7.7 \text{ cm}^{-1}/\text{GPa}$, and only a tiny ruby crystal is needed inside the sample space. This makes ruby fluorescence particularly well-suited for experiments that already incorporate an optical fiber into their cell [19]. For transport measurements such as ours, however, the limited space within the feedthrough cavity presents a trade-off: routing an optical fiber alongside the electrical wiring would significantly reduce the number of available wire connections, which is not practical given our measurement requirements.

The superconducting transition temperature T_c of several conventional BCS superconductors, such as Sn and Pb, varies with pressure in a well-characterized manner, making them useful as in-situ pressure calibrants, particularly at low temperatures [89]. Thin wires of these materials are placed inside the pressure cell alongside the sample, and

the pressure is inferred from the measured T_c using established calibration curves. The T_c can be determined either by transport measurements or by tracking the onset of diamagnetism via magnetic susceptibility, the latter being particularly convenient when susceptibility is already being measured as part of the experiment. For transport experiments such as ours, the T_c is readily extracted from a resistance measurement. While a four-wire configuration allows the resistance to be seen dropping to zero at T_c , a two-terminal measurement is also sufficient: even though the lead and contact resistances prevent the total resistance from reaching zero, we have found that the superconducting transition of the manometer wire remains clearly visible as a sharp anomaly in dR/dT . This is particularly advantageous given our limited wire count, as using only two wires for the manometer frees the remaining wires for the sample measurement.

An elegant alternative pressure calibration method requires no additional components or wiring: the gate capacitance of the heterostructure itself serves as an in-situ pressure sensor. The gate capacitance per unit area is given by:

$$C_g = \frac{en}{V_g} = \frac{\epsilon\epsilon_0}{t} \quad (4.1)$$

where ϵ is the dielectric constant of hBN, ϵ_0 is the permittivity of free space, t is the hBN thickness, V_g is the applied gate voltage, e is the electron charge, and n is the carrier density. Pressure modifies C_g through two mechanisms: compression of the hBN spacer reduces t , and the changing band structure of hBN under pressure modifies ϵ . Prior studies have established that the gate capacitance increases by approximately 9% per GPa of applied pressure [19]. Since C_g can be extracted directly from the carrier density versus gate voltage relation measured in transport, this provides a self-contained pressure calibration that requires no additional sensor, no extra wiring, and no modification to the pressure cell assembly. The pressure can therefore be determined at any point during a cooldown simply by measuring the gate capacitance of the device under study.

4.3 *Studying rhombohedral graphene under pressure*

Fig. 4.9 shows some preliminary high pressure transport data from an ongoing study. The sample being studied is R8G with crystallographic alignment to the bottom hBN (inset of Fig. 4.9a), forming a moiré unit cell with a lattice constant of $\lambda_{Gr-hBN} = 10.4$ nm.

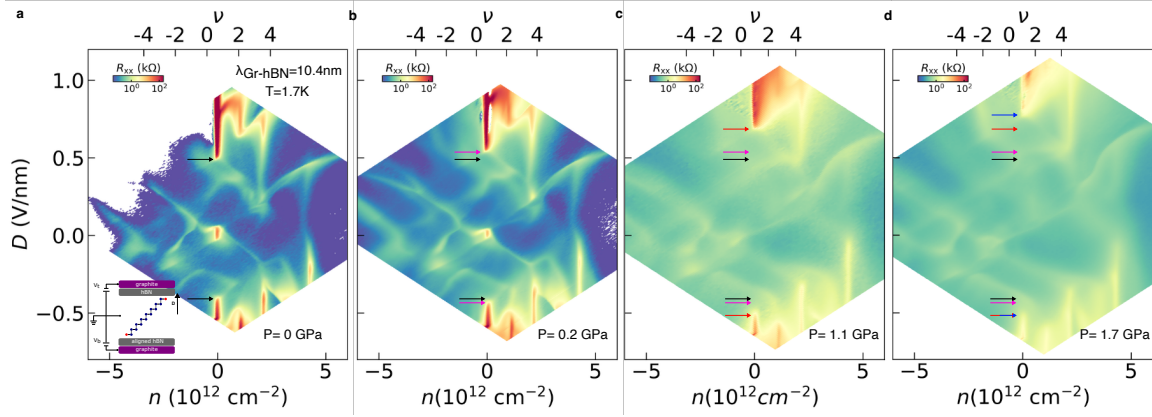


Figure 4.9: Evolution of moiré R8G under pressure. a-d Shows measurements of R_{xx} versus D and n of a moiré R8G sample (schematic in the inset) taken at different values of pressure. The arrows indicate the D at which the single particle gap opens.

Fig. 4.9a shows the $n - D$ phase diagram at $P = 0$. All the typical features of RG are seen in Fig. 4.9a, at the CNP we see resistive states at high D as well as near $D = 0$ corresponding to the single particle gapped states and correlation induced gapped states respectively. At integer filling of the moiré unit cell we see symmetry broken states.

Fig. 4.9b, c, and d show how the $n - D$ phase diagram changes at $P = 0.2, 1.1$ and 1.7 GPa respectively. Since, rhombohedral graphene is energetically unfavorable naively it is expected that mechanical transformation would cause the domain to relax into the stabler Bernal configuration. However, we note that the rhombohedral stacking configuration seems to survive up to 1.7 GPa of pressure, as evidenced by the single particle gapped states and the correlated insulators at integer filling of the moiré unit cell.

The correlation induced Layer Antiferromagnetic state (LAF) at $n = 0$ and $D = 0$ is increasingly suppressed with pressure. This state is strongest at about $P = 0$ (Fig. 4.9a), becomes completely suppressed at $P = 1.1$ GPa (Fig. 4.9c). Several previous studies have noted that this LAF state is suppressed in the presence of a strong moiré potential [75]. At higher pressures the moiré potential can be stronger leading to the suppression of this state. An alternative explanation could be that application of pressure is making the band less flat which leads to the suppression of this state.

The value of D at which the single particle gap opens is indicated by arrows in Fig. 4.9.

This value increases with increasing pressure. This implies that the band overlap is getting larger and as a result the D required to fully gap the two bands are increasing. The increasing overlap between the two bands can be explained by the enhancement of γ_3 and γ_4 hopping terms.

Chapter 5

CONCLUSIONS AND FUTURE OUTLOOK**5.1 Conclusions**

This thesis has presented experimental investigations of correlated electron phenomena in rhombohedral graphene, with a particular focus on superconductivity emerging from a dual-surface polarized semimetallic normal state, and on the development of hydrostatic pressure as a tuning parameter for van der Waals heterostructures.

In Chapter 2, we reported the observation of superconductivity in unaligned R8G and moiré R7G samples, where they arise from a semimetallic normal state. Due to the chiral stacking of rhombohedral graphene this semimetallic state is charge-delocalized with coexisting valence- and conduction-band Fermi pockets are polarized to opposite crystal surfaces. Using self-consistent Hartree–Fock band-structure calculations, corroborated by transport measurements, the extent of this semimetallic regime is mapped out. In R8G, we observe 10 superconducting pockets and in the moiré R7G we have 2 superconducting pockets in the semimetallic regime. These superconducting pockets show up at fixed gate voltages, which allow us to determine that they correspond to some fixed filling of the conduction band, but are modulated by screening from the valence bands. This work shows how polarization along the stacking axis becomes an important degree of freedom that will have to be taken into account while studying the physics of thicker rhombohedral graphene.

In Chapter 3, we present our investigation of these superconducting states with in-plane magnetic field ($B_{\parallel}$), specifically in the moiré R7G samples. We find that the sharp resistive feature, out of which the two superconducting pockets form, lies on the boundary between an unpolarized fully degenerate state and a spin polarized state which has half its degeneracy lifted. We find that $B_{\parallel}$ induces superconductivity all along the sharp resistive feature. Portions of this superconducting state survive all the way up to $B_{\parallel} = 6$ T, violating an upper bound on the Pauli paramagnetic limit by an order of magnitude, strongly suggesting that the pairing order is spin-triplet. While the

superconducting state reaches its maximum extent in the $V_b - V_t$ plane at $B_{\parallel} = 1$ T, the strength of superconductivity has a complicated dependence with both V_b and $B_{\parallel}$. We explain this observation of $B_{\parallel}$ induced superconductivity with a phenomenological model where the interplay of Hund's coupling, spin-orbit and Coulomb interactions lead the system to condense into a ground state (at $B_{\parallel} = 0$), where superconductivity is forbidden due to valley imbalance.

In chapter 4, we introduced hydrostatic pressure as an additional tuning parameter for studying correlated states in van der Waals heterostructures. Because pressure directly modifies the interlayer spacing and electronic coupling between layers, it provides a powerful and disorder-free means of engineering band-structure and many-body interactions. To enable such studies, we developed and implemented a high pressure transport platform based on a piston-cylinder cell, addressing several key technical challenges associated with applying hydrostatic pressure to atomically thin devices. This included the introduction of pre-diced substrates, a custom PCB-based wiring architecture, and a standardized assembly procedure that significantly improves device reliability, electrical connectivity, and experimental throughput. We also discussed methods for in-situ pressure calibration using superconducting manometers and gate capacitance measurements. Finally, we presented preliminary measurements of rhombohedral graphene under pressures up to 1.7 GPa. These data demonstrate that the rhombohedral stacking remains stable under substantial compression and reveal pressure-induced modifications to the electronic phase diagram, including the suppression of the correlation-driven layer antiferromagnetic state and an increase in the displacement field required to open the single-particle gap. Together, these results establish hydrostatic pressure as a promising new experimental knob for investigating and controlling correlated phenomena in rhombohedral graphene and related van der Waals materials.

5.2 Outlook

The results presented in this thesis open several promising directions for future research. The observation of superconductivity emerging from a dual-surface polarized semimetallic state raises fundamental questions regarding the microscopic origin of

pairing in rhombohedral graphene. While the superconducting pockets were found to occur at fixed conduction-band fillings and are strongly influenced by screening from the valence band, the mechanism by which the two surface-polarized carrier populations interact to produce superconductivity remains unclear. Subsequent studies of thicker rhombohedral graphene systems have reported superconductivity emerging from similarly charge-delocalized normal states, suggesting that the phenomenology observed here may be a more general feature of thicker rhombohedral graphene rather than a peculiarity of the specific devices studied in this work. Future experiments combining transport with local probes or thermodynamic measurements may help determine whether superconductivity originates predominantly from one surface, involves both surfaces equally, or is fundamentally tied to the coexistence of electron and hole pockets in the semimetallic regime. More broadly, these observations suggest that polarization along the stacking axis constitutes an important electronic degree of freedom in rhombohedral graphene. Future theoretical descriptions of superconductivity in thicker rhombohedral graphene will therefore need to account for the spatial distribution of electronic states across the stack, the associated screening between surface-polarized bands, and the role that these effects play in determining the superconducting pairing interaction.

The strong enhancement of superconductivity by in-plane magnetic field and the large violation of the Pauli paramagnetic limit suggest that the superconducting state may possess an unconventional order parameter. Establishing the precise nature of this pairing state remains an important challenge. Careful scanning SQUID measurements of magnetization would help verify the phenomenological model that we have proposed. Proximitization with TMDs which have large spin-orbit coupling should have the same effect as $B_{\parallel}$, some ongoing work does indeed suggest this to be the case. More broadly, the apparent competition between superconductivity and other symmetry-broken states near the spin polarization transition highlights the rich interplay between spin, valley, and interaction effects in rhombohedral graphene, and suggests that the delicate intricacies of these phases remain to be discovered.

The development of a high pressure transport platform introduces a new experimental degree of freedom for investigating these questions. Pressure offers a unique means of continuously tuning interlayer coupling and electronic bandwidth without introducing disorder, making it particularly well suited for studying the

relationship between band-structure and correlated ground states. The preliminary measurements presented here already indicate that pressure substantially modifies the electronic phase diagram of rhombohedral graphene while preserving the rhombohedral stacking order. Extending these studies to higher pressures, lower temperatures and combining pressure with independent control of carrier density, displacement field, and magnetic field may provide new opportunities to engineer correlated phases and disentangle the roles of bandwidth, screening, and interaction strength. More broadly, these developments establish hydrostatic pressure as a generally applicable, reliable tuning knob for correlated phenomena across a wide range of van der Waals and other two-dimensional material systems.

More generally, rhombohedral graphene continues to emerge as a versatile platform for exploring strongly interacting electronic matter. External control parameters such as carrier density, displacement field, magnetic field and hydrostatic pressure provide independent and continuous tuning of the electronic environment, while internal degrees of freedom such as layer polarization and spin polarization define the resulting correlated ground states. The interplay between these externally controlled knobs and emergent internal order gives access to an exceptionally rich parameter space. As experimental techniques continue to advance, these capabilities are likely to enable the discovery of new correlated phases and deepen our understanding of unconventional superconductivity in two-dimensional materials.

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Appendix A

SAMPLE FABRICATION AND TRANSPORT MEASUREMENT DETAILS

The fabrication and characterization of van der Waals heterostructures has matured considerably over the past decade, and the core techniques employed in this work: mechanical exfoliation, dry transfer assembly, and standard electron-beam lithography are now well-established and extensively documented in the literature [104, 102, 103, 101]. Rather than reproduce a comprehensive account of these methods here, this chapter focuses on the aspects of device fabrication and measurement that are specific to rhombohedral graphene. The central challenge in working with this material from other 2D materials is the identification and isolation of rhombohedral stacking domains: rhombohedral graphene is metastable with respect to the energetically favored Bernal configuration, and naturally exfoliated graphene flakes contain both stacking orders intermixed on the micron scale. Reliably identifying, isolating the domains and fabricating it into a heterostructure while keeping it rhombohedral therefore requires techniques beyond the standard heterostructure assembly workflow, and these are the focus of the discussion that follows. I will also briefly outline the standard DC limit transport techniques used to characterize 2D materials. Experimental techniques associated with high pressure measurements will be discussed separately in Chapter 4.

A.1 Device Fabrication

A.1.1 Identification and Isolation of Rhombohedral Domains

While on average 15% of all exfoliated graphene is rhombohedral [91], these domains cannot be distinguished under the microscope with just optical light. We use a suit of different techniques to identify them. The distinct low energy band-structures of rhombohedral and Bernal lead to different contributions of interband phase transitions to their optical conductivity. As a result, in the infra-red (IR) range the optical conductivity of rhombohedral graphene decreases where as that of Bernal increases. This produces

measurably different differential reflectance in the infrared regime [91]. Taking advantage of this fact we identify rhombohedral domains in exfoliated flakes of appropriate layer number via Hamamatsu InGaAs infrared camera mounted on a Nikon LV-Microscope. Fig . A.1a and b show pictures taken with a normal microscope camera and an infrared camera respectively, the rhombohedral domains appear lighter than the Bernal domains.

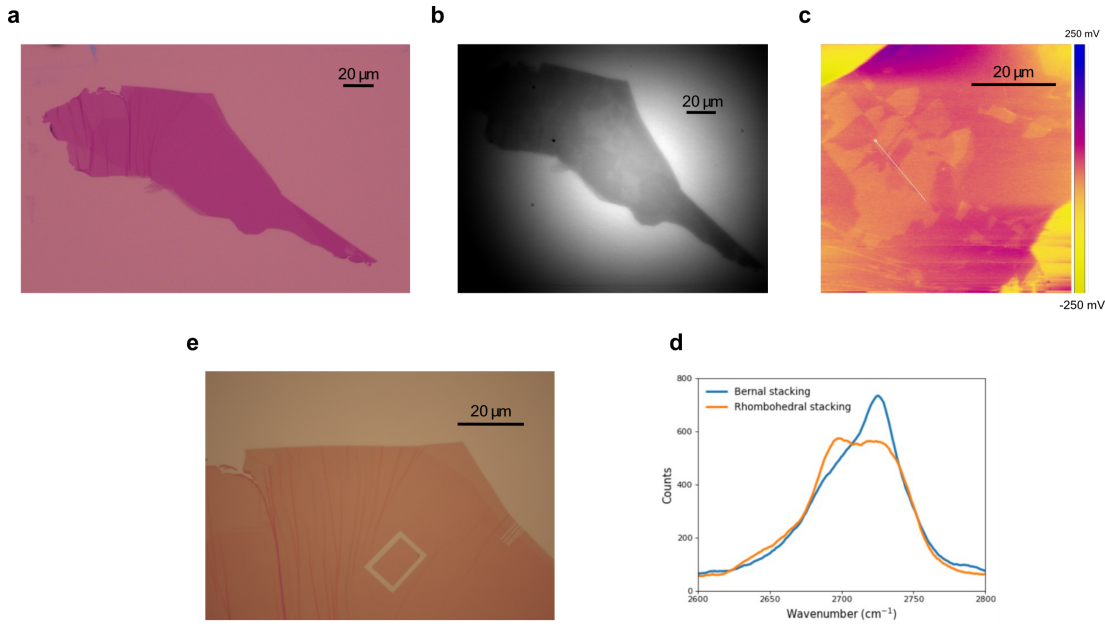


Figure A.1: **Identification and isolation of rhombohedral graphene domain.** **a**, optical (visible spectrum) micrograph of multilayer graphene flake. **b**, infra-red (IR) micrograph of the same flake as in **(a)**, with rhombohedral domains visible as regions of lighter contrast. **c**, KPFM scan showing the surface potential over the same flake as in **(a)**. **d**, Raman spectrum of 2D peak of Bernal and rhombohedral graphene. **e** Rhombohedral graphene domain isolated from the rest of the flake through anodic oxidization.

In case of any further ambiguity, we use Raman Spectroscopy to cross-check the stacking order. The stacking order modifies the electronic band-structure of multilayer graphene, which in turn changes the electron-phonon coupling that governs the Raman response. In particular, the 2D peak arises from a second-order double-resonant scattering process involving intervalley phonons near the K and K' points of the Brillouin zone, and is therefore highly sensitive to the details of the electronic dispersion [92]. Because Bernal Stacking and rhombohedral Stacking graphene have markedly different low-energy

band-structures, the resonant scattering pathways that contribute to the 2D peak are modified, leading to systematic differences in its position, shape, and relative intensity that allow the two stacking domains to be distinguished spectroscopically. In particular, as seen in Fig . A.1d, rhombohedral (ABC) graphene typically shows an asymmetric 2D peak because its flatter, more strongly particle-hole asymmetric low-energy bands lead to an unequal distribution of resonant scattering channels, whereas Bernal (ABA) graphene has more dispersive, nearly symmetric bands that produce a more symmetric 2D line shape.

Once we have confirmed the stacking order and obtained high resolution images, we can use AFM based anodic oxidation to isolate the domains. An AFM tip is used to locally oxidize the graphene by applying a large positive bias to the tip [93]. At this step it is important to ensure that there are no pockets of Bernal within our rhombohedral domain. The energy required to expand a pre-existing Bernal domain would be considerably less than the energy needed to nucleate a new Bernal domain. Anecdotally, we think this has a pretty large effect on the yield of samples that stay rhombohedral after fabrication. Fig . A.1e shows a rhombohedral domain isolated from a larger flake.

A.1.2 hBN alignment

If we want to align the hexagonal boron nitride (hBN) substrate to the rhombohedral graphene, we first need to determine the crystallographic axes of the flakes. This is typically done by inspecting straight edges of exfoliated flakes, since hexagonal 2D materials such as graphene and hBN preferentially fracture along high-symmetry directions corresponding to the armchair and zigzag orientations, which are separated by 30° . However, identifying a straight edge alone does not uniquely determine the lattice orientation, since a given edge can correspond to either armchair or zigzag, leaving a discrete 60° ambiguity in assigning the true crystallographic axes. Consequently, even when using visible flake edges as a proxy for alignment, there remains only a probabilistic chance of correctly matching the graphene and hBN crystallographic axes, which in practice limits deterministic moiré alignment.

To resolve this ambiguity, we use Second Harmonic Generation (SHG) on hBN. SHG is symmetry-selective and arises from the non-centrosymmetric crystal structure of hBN; its

efficiency depends on the orientation of the crystal axes through the second-order nonlinear susceptibility tensor [94]. As the crystal is rotated, the SHG intensity exhibits a characteristic six-fold angular dependence, allowing the armchair and zigzag directions to be uniquely distinguished and providing an unambiguous determination of the in-plane crystallographic axes of the hBN substrate.

In our device geometry, each graphene stack is encapsulated between a top and bottom hBN layer, and independent alignment of both encapsulating crystals is required to control the relative twist angles that define the moiré environment. While Second Harmonic Generation (SHG) removes the crystallographic ambiguity for each individual hBN flake, a residual $\pm 30^\circ$ uncertainty remains in relating the graphene edge assignment to a specific armchair or zigzag direction. To maximise the probability of achieving near-zero effective twist, we therefore fabricate devices in both possible relative configurations, using one hBN flake aligned under the assumption of zigzag correspondence and the other under armchair correspondence, ensuring that at least one encapsulation configuration yields the correct crystallographic registry with the graphene. A recent work [95] further suggests that the crystallographic axes of graphene can be directly determined by tracking the crystal facets of a cleaved bulk crystal, which are overwhelmingly likely to correspond to the armchair direction [96], thereby offering a more deterministic route to resolving the remaining ambiguity.

A.1.3 Device Assembly

Once the rhombohedral domain has been identified, the next challenge is to incorporate it into a dual-gated heterostructure while preventing relaxation into the Bernal configuration. Since rhombohedral graphene is a metastable stacking order, it can relax to the thermodynamically favored Bernal phase under external perturbations, with shear and in-plane mechanical stresses being particularly effective in driving this transition. To minimize such perturbations during assembly, we construct the heterostructure in a two-step process. First, the bottom portion of the stack is prepared by picking up a hBN flake and a graphite back gate, and placing them onto an SiO₂ substrate. The surface of the hBN is then cleaned using contact-mode AFM (OTESPA-R3 tip, line spacing ≈ 100 nm) to ensure a contamination-free interface for subsequent transfer.

In parallel, the top half of the stack, consisting of graphite/hBN/rhombohedral graphene, is assembled separately and subsequently deposited onto the pre-patterned bottom stack. All van der Waals assembly steps are carried out using standard dry-transfer techniques with a polycarbonate (PC) film on a polydimethylsiloxane (PDMS) stamp. Throughout the process, we minimise abrupt motions of the PC film to reduce shear forces on the encapsulated graphene. It has also been suggested that picking up rhombohedral graphene along the zigzag direction can help preserve its stacking order during transfer [97].

After stacking, standard device fabrication procedures are employed to define the dual-gated Hall bar geometry, including reactive ion etching and evaporation of 7/70 nm Cr/Au contacts using poly(methyl methacrylate) (PMMA) masks patterned by electron-beam lithography. Optical micrographs of the completed devices are shown in Extended Data Fig B.1 a and b.

A.2 Transport Measurements

The primary technique used to characterize rhombohedral graphene (RG) devices in this work is low-temperature electronic transport. Low-temperature transport measurements provide a direct probe of the correlated electronic phases in RG by measuring the resistive response of the system as a function of carrier density, displacement field, magnetic field, and related external tuning parameters. By cooling the device to millikelvin temperatures in a dilution refrigerator, thermal fluctuations are strongly suppressed, allowing fragile collective states such as superconducting phases, correlated insulators, and symmetry-broken metallic states arising from strong electronic interactions to emerge clearly in transport.

Transport measurements are performed in a standard four-terminal lock-in configuration (Fig. A.2b). A small AC excitation current, typically in the range 0.5–10 nA at a frequency of approximately 10 Hz, is driven through the device, while the longitudinal voltage V_{xx} and transverse Hall voltage V_{xy} are measured independently. This allows the longitudinal resistivity ρ_{xx} and Hall resistivity ρ_{xy} to be extracted without contributions from contact resistance. In an ideal device, the rhombohedral graphene leads would remain metallic; however, due to strong correlation effects in RG, an

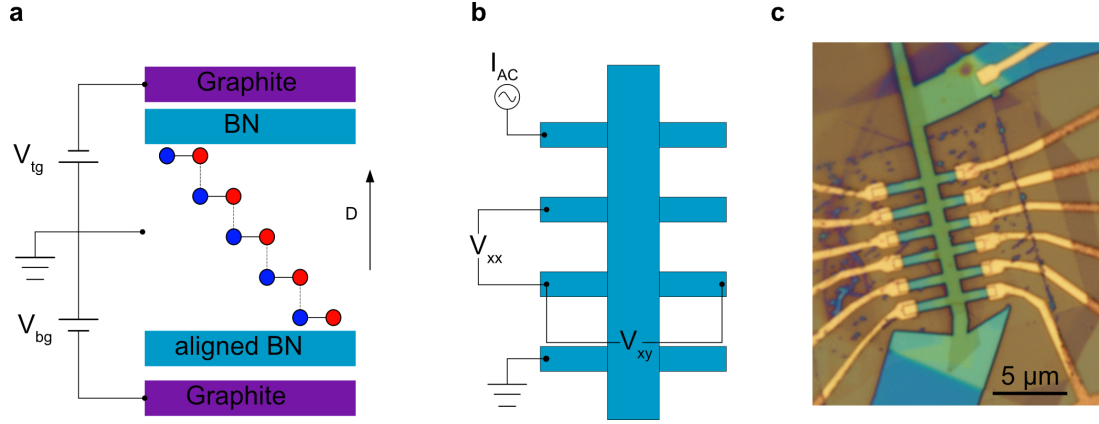


Figure A.2: **Schematic of devices.** **a**, cross sectional schematic of rhombohedral graphene samples. The rhombohedral graphene is encapsulated between two layers of hBN. Graphite gates on either side are used to gates the samples. **b**, shows a top view of the sample. All samples have a “hall-bar” geometry with contacts projecting out from the central. These contacts are used to pass current through the central bar and allow for measurement of both longitudinal and transverse resistance. **c**, shows an optical micrograph of a typical device.

insulating state can emerge at $n = 0$ and $D = 0$. As a result, a sufficiently large gate voltage must be applied to the Si back gate to dope the contact regions away from charge neutrality and ensure reliable injection into the device.

The dual-gate geometry (Fig. A.2a) enables independent control of carrier density n and displacement field D , which are related to the applied gate voltages via

$$n = \frac{C_b V_b + C_t V_t}{e}, \quad D = \frac{C_b V_b - C_t V_t}{2\epsilon_0}, \quad (\text{A.1})$$

where C_t and C_b are the top and bottom gate capacitance per unit area, respectively, and ϵ_0 is the vacuum permittivity. These capacitances are typically extracted by fitting the slopes of quantum Hall features in Landau fan diagrams. By sweeping V_t and V_b , a two-dimensional (n, D) phase diagram is constructed. Additional tuning parameters such as temperature T , out-of-plane magnetic field $B_\perp$, in-plane magnetic field $B_\parallel$, and DC current bias I_{DC} provide further control over the electronic state. Together, these knobs make low-temperature transport a powerful and information-rich probe of the correlated phase diagram, capable of distinguishing phases with different symmetry, topology, and spin character through their distinct transport signatures.

Appendix B

SUPPLEMENTARY INFORMATION FOR CHAPTER 2

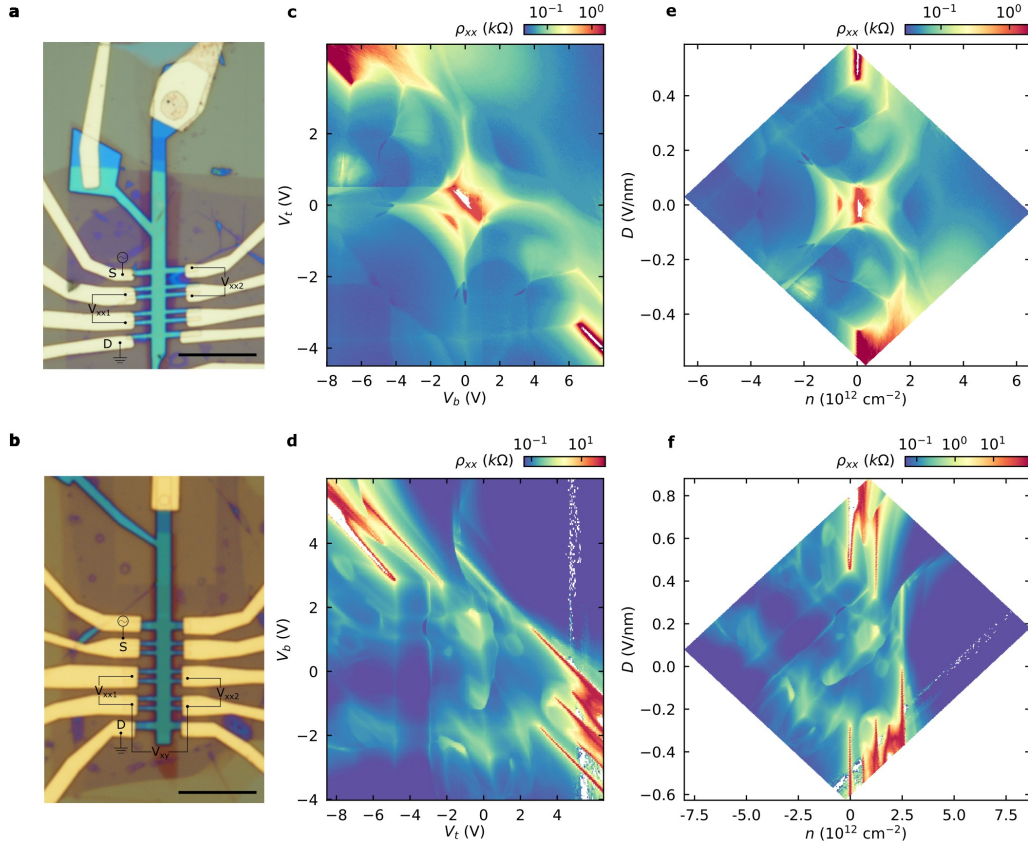


Figure B.1: **Optical micrographs and full gate maps of both devices.** **a**, Optical micrograph of the misaligned R8G device. **b**, Same as **(a)** for the moiré R7G device. For both, the scale bar is $10\ \mu\text{m}$. **c**, Map of ρ_{xx} for the R8G device measured over a wide range of V_b and V_t . The map is acquired in pieces and stitched together, with different silicon gate voltages in order to optimally reduce the contact resistance in each quadrant. **d**, Same as **(c)** for the R7G device. **e**, The map from **(c)** converted to $n - D$ axes (see Methods for conversion). **f**, Same as **(e)** for the R7G device.

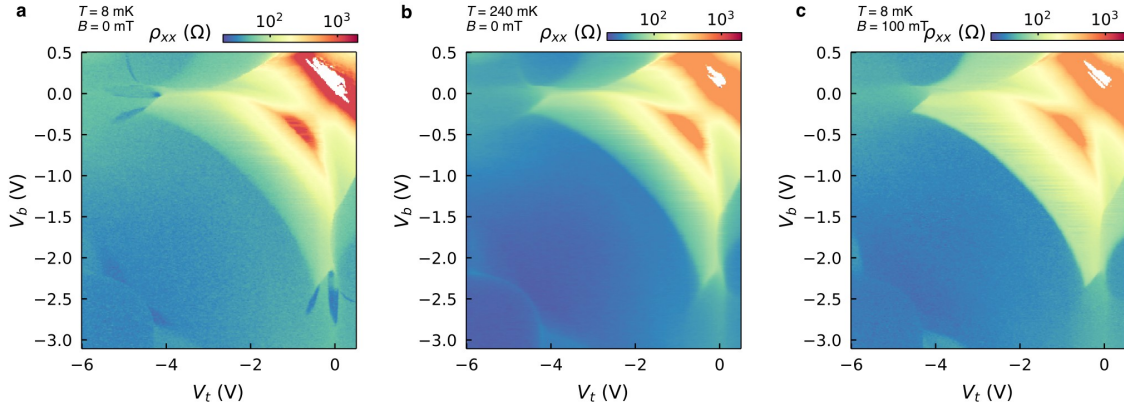


Figure B.2: Transport with varying temperature and magnetic field in the R8G device. **a**, Map of ρ_{xx} versus V_b and V_t at base temperature and $B = 0$. The highly resistive feature in the top right is the correlated insulator at the CNP surrounding $D = 0$. Superconducting pockets SC1₈ to SC4₈, and their corresponding $D < 0$ counterparts, are visible within this region of gate voltage. **b**, The same map as **(a)** at $T = 240$ mK, above T_c of all the pockets. **c**, The same map as **(a)** at $B = 100$ mT, above B_c of all the pockets.

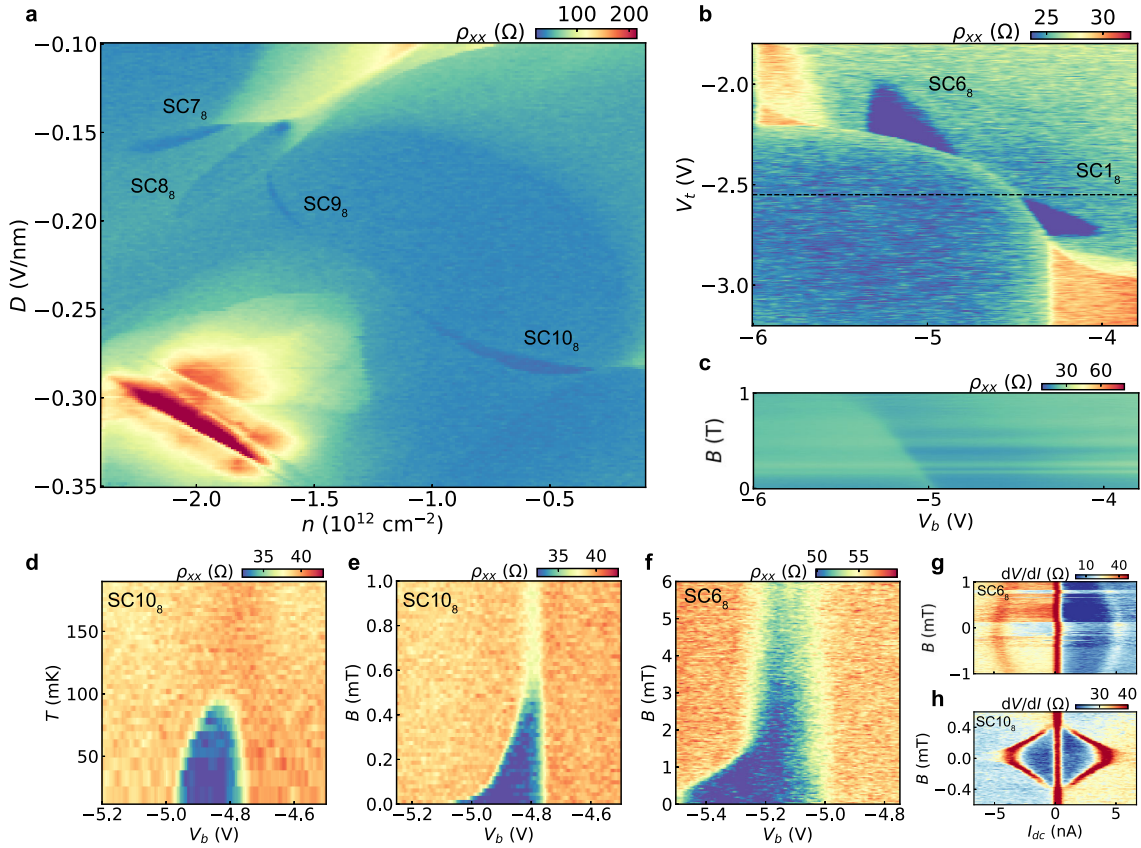


Figure B.3: Additional characterization of superconducting pockets at $D < 0$ in the R8G device. **a**, Map of ρ_{xx} versus n and D corresponding to $D < 0$. The map contains four superconducting pockets, SC7₈ to SC10₈, which correspond to SC2₈ to SC5₈ for $D > 0$. The feature in the bottom left corner exhibits unusual nonlinear behavior and does not appear for $D > 0$; at present, we do not know its origin. **b**, Map of ρ_{xx} versus V_b and V_t showing SC1₈ and SC6₈. **c**, Landau fan diagram taken by sweeping V_b with fixed $V_t = -2.55$ V, corresponding to the black dashed line in **(b)**. The sharp resistance step associated with the edge of SC1₈ slopes to the left with B , suggestive of a spin-polarized half-metal to the right and an isospin unpolarized phase to the left. **d**, Measurement of ρ_{xx} versus V_b and T at fixed $V_t = 1.6$ V and $B = 0$ for SC10₈. **e**, Similar measurement versus B at base temperature. **f**, Similar measurement as **(d)** for the SC6₈ pocket at fixed $V_t = -2.2$ V. **g**, Measurement of dV/dI versus I_{dc} and B for SC6₈. **h**, Similar measurement for SC10₈. Both exhibit the similar zero bias anomalies as seen for the $D > 0$ pockets.

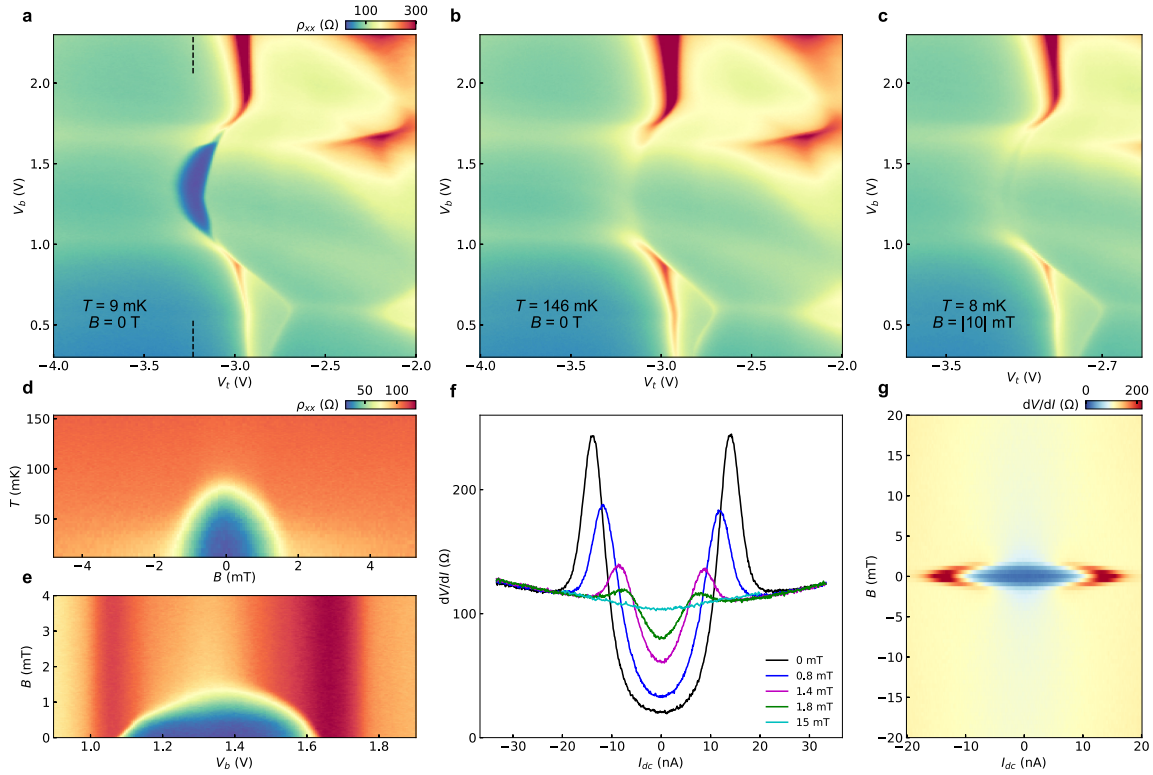


Figure B.4: Additional characterization of the SC1₇ pocket from the moiré R7G device.

a, Map of ρ_{xx} versus V_b and V_t at base temperature and $B = 0$. **b**, Similar measurement at $T = 146$ mK, above T_c of SC1₇. **c**, Similar measurement, over a narrowed range of V_t , at base temperature and symmetrized at $|B| = 10$ mT, above B_c . **d**, Measurement of ρ_{xx} versus B and T at optimal doping of SC1₇ ($V_b = 1.40$ V and $V_t = -3.22$ V). **e**, Measurement of ρ_{xx} versus V_b and B at base temperature and fixed $V_t = -3.23$ V, corresponding to the line trace indicated by black dashed lines in **(a)**. In this map, SC1₇ is bounded by two resistance bumps associated with nearly constant V_b , i.e., the surface associated with the valence band. **f**, Measurements of dV/dI versus I_{dc} at selected values of B taken at optimal doping of SC1₇ ($V_b = 1.40$ V and $V_t = -3.22$ V). **g**, Map of dV/dI versus I_{dc} and B .

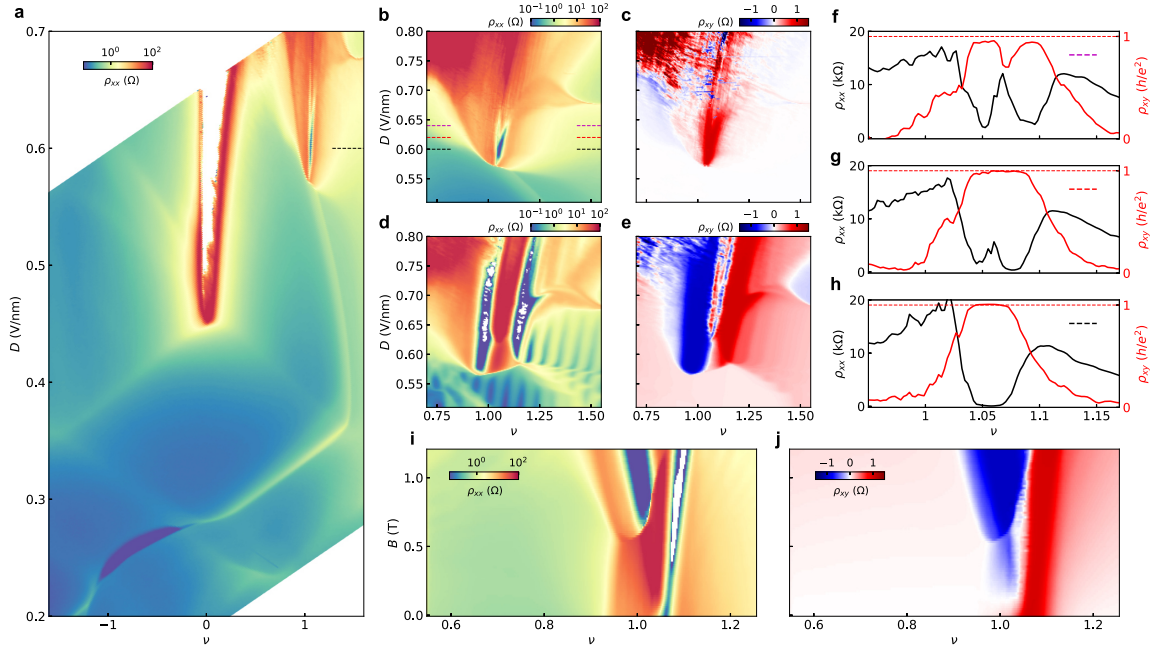


Figure B.5: **Additional analysis of the IQAH state in the moiré R7G device.** **a**, Map of ρ_{xx} versus ν and D taken at base temperature and $B = 0$. **b**, Zoom-in of the region around $\nu = 1$ at large D ($B = 0$). **c**, Map of ρ_{xy} over the same region as **(b)** ($B = 0$). **d**, Same as **(b)** symmetrized at $B = 2$ T. **e**, Same as **(c)** antisymmetrized at $B = 2$ T. **f**, Measurements of ρ_{xx} and ρ_{xy} versus ν taken at $B = 0$ at the positions of the corresponding color-coded dashed lines in **(b)**. At $D = 0.60$ V/nm there is a single contiguous region near $\nu = 1.05$ with $\rho_{xy} \approx h/e^2$ and small ρ_{xx} . At larger D , the state splits into two, with a resistive bump separating them. **i**, Landau fan of ρ_{xx} taken at $D = 0.6$ V/nm, corresponding to the black dashed line in **(a)**. **j**, The same measurement as **(i)**, but of ρ_{xy} . The fans show the $C = +1$ state arising from $B = 0$, and the abrupt emergence of a $C = -1$ state above $B \approx 0.6$ T. The coexistence of these two states at modest B can also be seen from the maps in **(d)**-**(e)**.

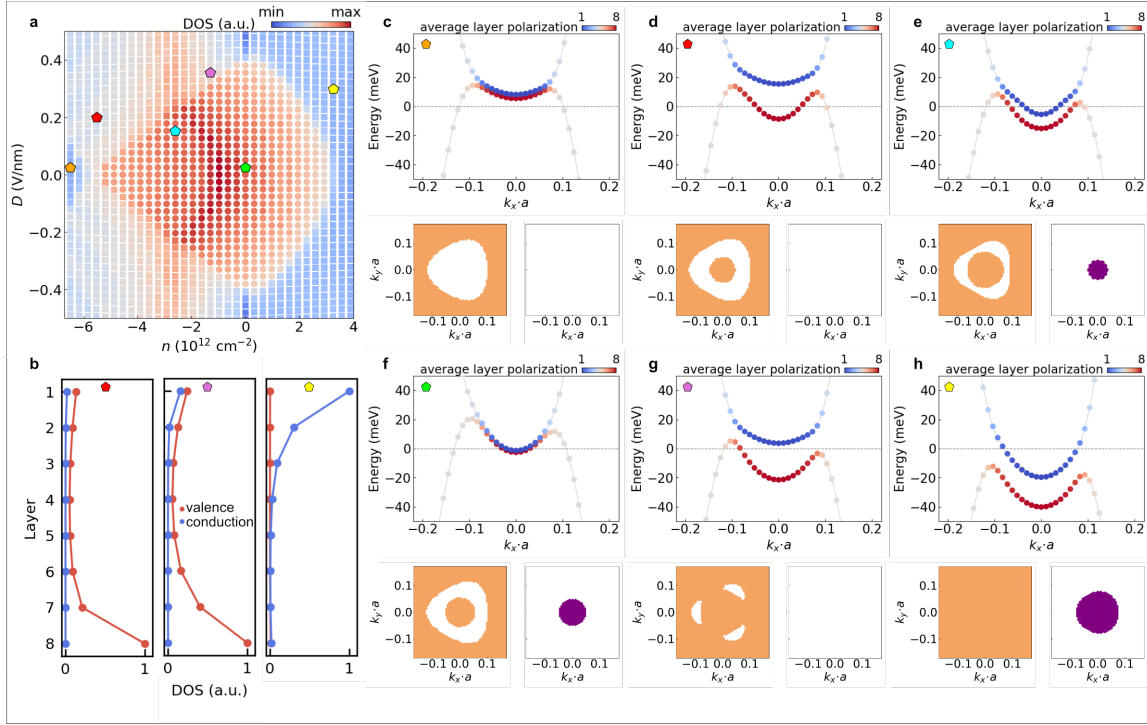


Figure B.6: **Additional band-structure calculations for misaligned R8G.** **a**, Calculation of the DOS versus n and D in the Hartree-Fock approximation, reproduced from Fig. 2.2e. **b**, Calculated DOS as a function of layer number for three different combinations of n and D , corresponding to the positions of the color-coded markers in **a**. Note that for the data corresponding to the purple marker, there is an artificial conduction band tail due to numerical broadening. **c**, (Top) Hartree-Fock band-structure corresponding to the n and D denoted by the orange marker in **(a)**. The color coding of points indicates their average layer polarization. (Bottom) Fermi surface contour plots at the Fermi energy ($E = 0$), where white represents empty states and colors represent filled states. The valence band is on the left and the conduction band on the right. **d-h**, The same as **(c)**, at the combinations of n and D indicated by the color-coded markers.

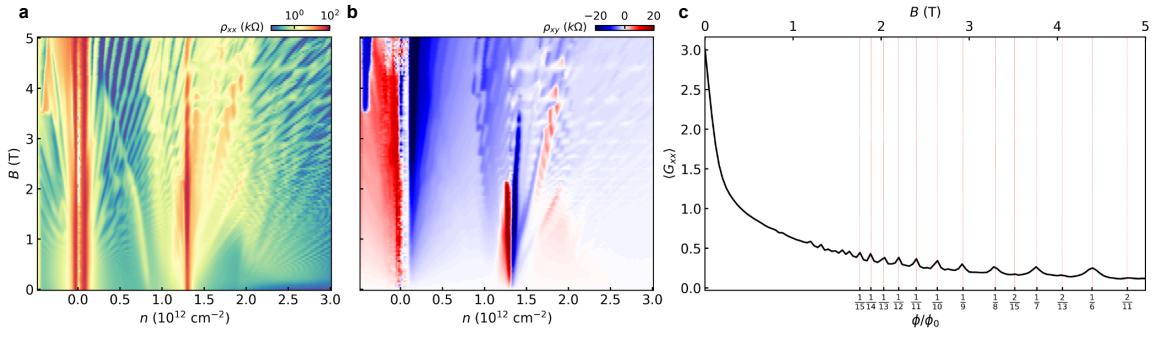


Figure B.7: **Brown Zak Oscillations in the 7L Device** **a.** ρ_{xx} as a function of n and B at $D = 0.4 \text{ V/nm}$. **b.** ρ_{xy} corresponding to **a.**. **c.** Average conductance $\langle G_{xx} \rangle$ as a function of the magnetic flux ϕ/ϕ_0 , extracted from **a.** and **b.** Brown-Zak oscillations are visible as peaks in the spectrum. From the Brown Zak oscillations we extract the density corresponding to full filling of the moiré bands: $n_s = 2.55 \times 10^{12} \text{ cm}^{-2}$. **d, e, f.** Same as **a, b, c.** at $D = 0.5 \text{ V/nm}$.

Appendix C

SUPPLEMENTARY INFORMATION FOR CHAPTER 3

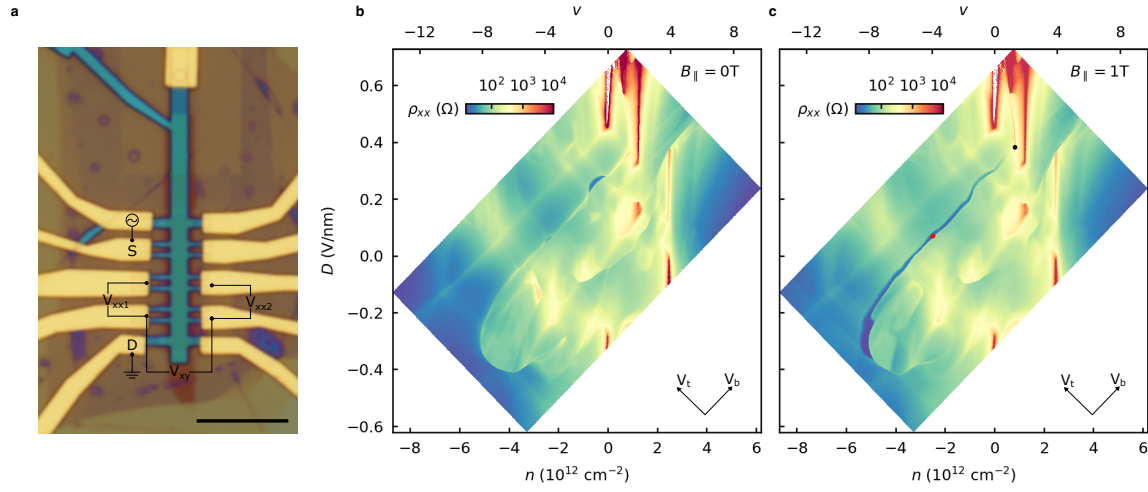


Figure C.1: **Device image and $n - D$ maps.** **a**, Optical micrograph of the R7G sample with source, drain and voltage probe contacts annotated. The scale bar is $10 \mu\text{m}$. **b**, Map of ρ_{xx} plotted as a function of $n - D$ axis (see Methods for conversion), taken at $B_{||} = 0 \text{ T}$. **c**, same as (b) but taken at $B_{||} = 1 \text{ T}$.

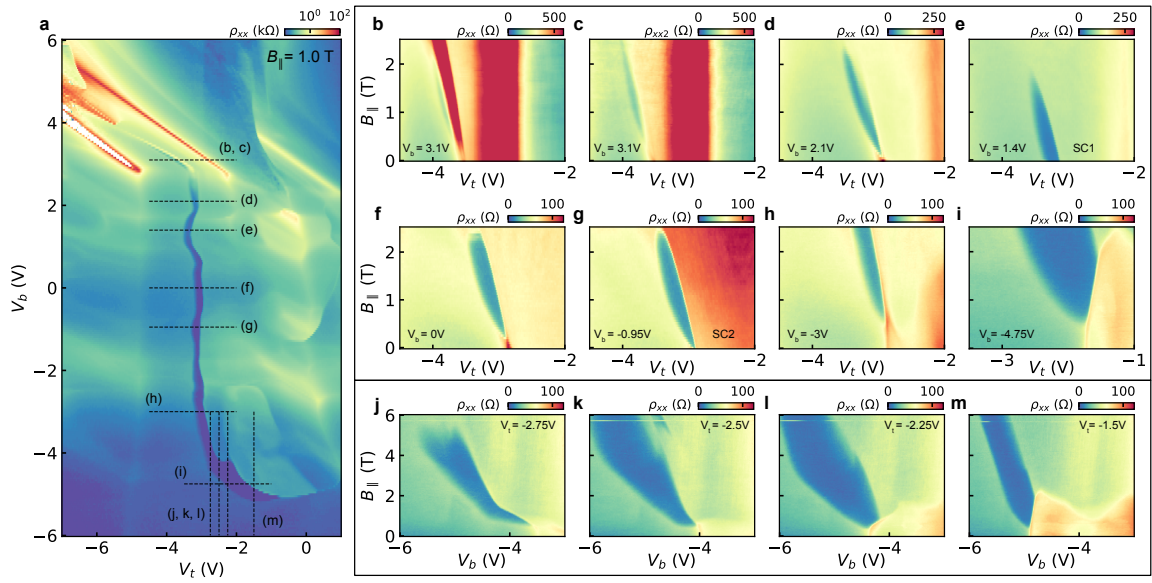


Figure C.2: **Evolution of the paired state with $B_{\parallel}$.** **a**, Map of ρ_{xx} versus V_b and V_t taken at $B_{\parallel} = 1$ T. **b**, ρ_{xx} versus $B_{\parallel}$ and V_t taken at $V_b = 3.1$ V (marked by the black dashed line in **(a)**) using V_{xx1} . **c**, Same as **(b)** using V_{xx2} . **d**, Same as **(b)** taken at $V_b = 2.1$ V, **e**, $V_b = 1.4$ V, **f**, $V_b = 0$ V, **g**, $V_b = -0.95$ V, **h**, $V_b = -3$ V, and **i**, $V_b = -4.75$ V. **j**, ρ_{xx} versus $B_{\parallel}$ and V_b taken at $V_t = -2.75$ V. **k**, Same as **(j)** taken at $V_t = -2.5$ V, **l**, $V_t = -2.25$ V, and **m**, $V_t = -1.5$ V. Unless otherwise specified, measurements are taken using V_{xx1} . Boxes around **(b-i)** and **(j-m)** delineate maps taken at fixed V_b and at fixed V_t respectively.

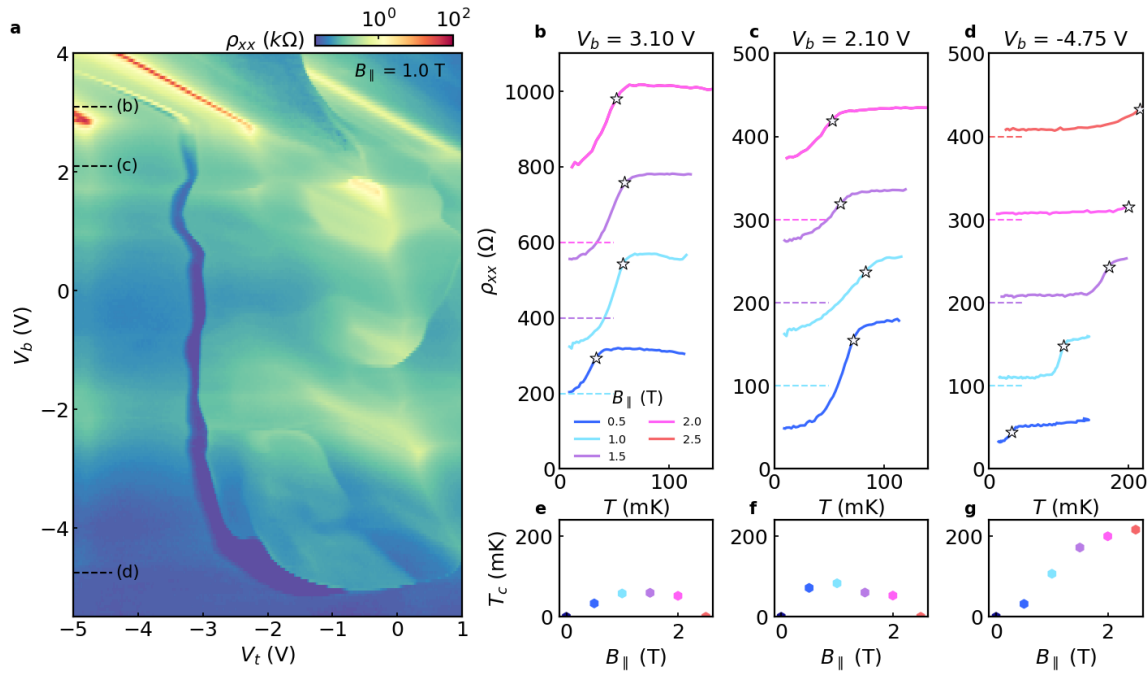


Figure C.3: **Additional measurements showing a non-monotonic evolution of T_c with $B_{\parallel}$.** **a**, Map of ρ_{xx} versus V_b and V_t taken at $B_{\parallel} = 1.0$ T. The dashed lines correspond to the V_b values in **(b)**, **(c)**, and **(d)**. **b**, ρ_{xx} versus T measurements at several values of $B_{\parallel}$ with $B_{\perp} = 0$. The measurement is taken with $V_b = 3.10$ V and $V_t \approx -2.0$ V. Curves are vertically offset for clarity. Dashed lines denote $\rho_{xx} = 0$ for each curve of corresponding color. Stars denote T_c . The legend shows the value of $B_{\parallel}$ at which each curve was taken. **c**, Same for $V_b = 2.10$ V with $V_t \approx -2.0$ V. **d**, Same for $V_b = -4.75$ V with $V_t \approx -1.0$ V. **e-g**, Plots of T_c versus $B_{\parallel}$ for each of the three associated values of V_b . Note that these are the analogous measurements from Fig. 3.5 of the main text, shown at additional values of V_b .

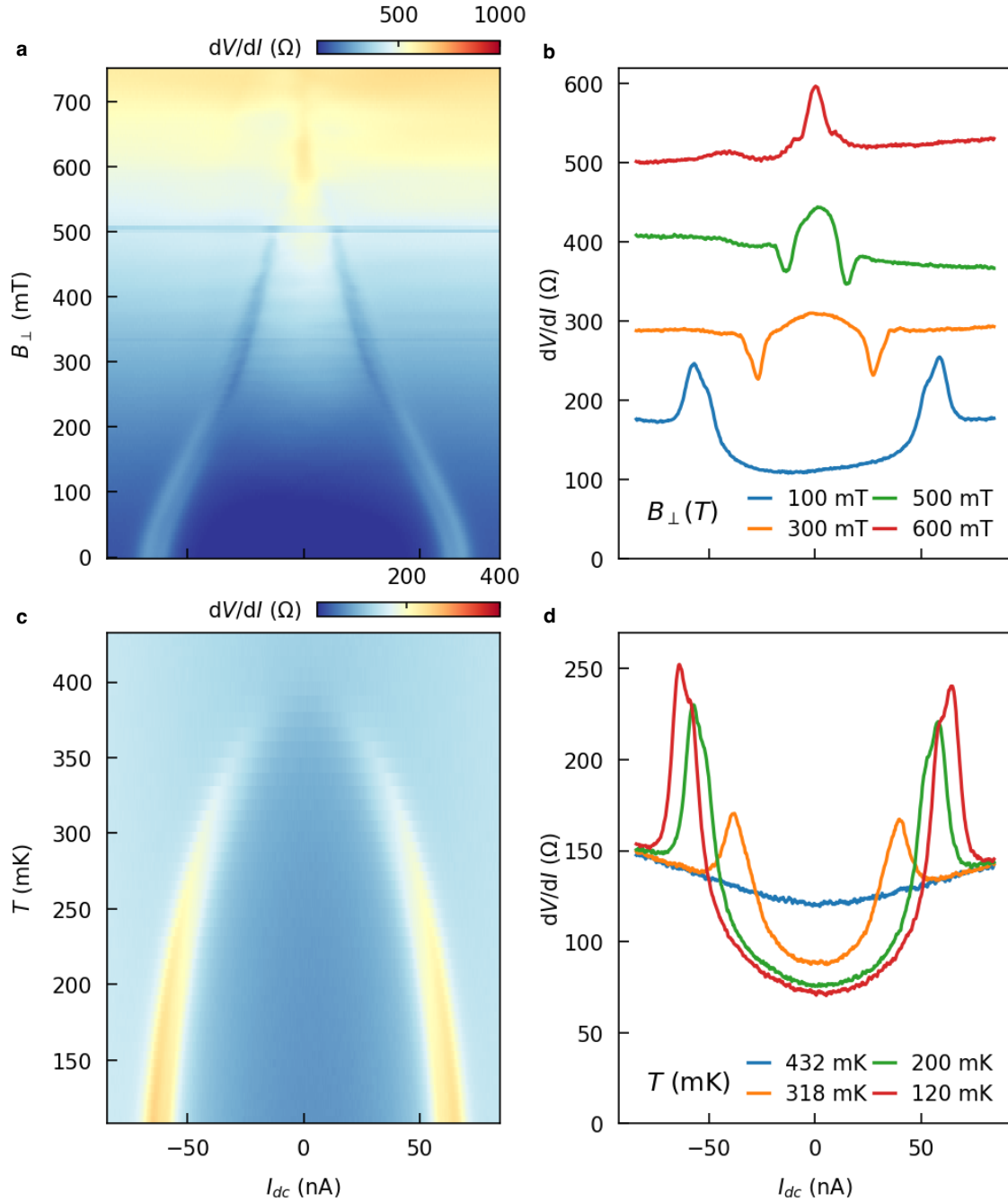


Figure C.4: **Additional characterization of non-linearity associated with the sharp resistive bump.** **a**, dV/dI versus $B_{\perp}$ and I_{dc} with $B_{\parallel} = 0$ T, $V_b = -3$ V and V_t varied with $B_{\perp}$ to stay on the sharp resistive bump. **b**, Line cuts from **(a)** at fixed values of $B_{\perp}$. **c**, dV/dI versus T and I_{dc} with $B_{\parallel} = 0$ T, $V_b = -3$ V and $V_t = -2.75$ V. **d**, Line cuts from **(c)** at fixed values of T .

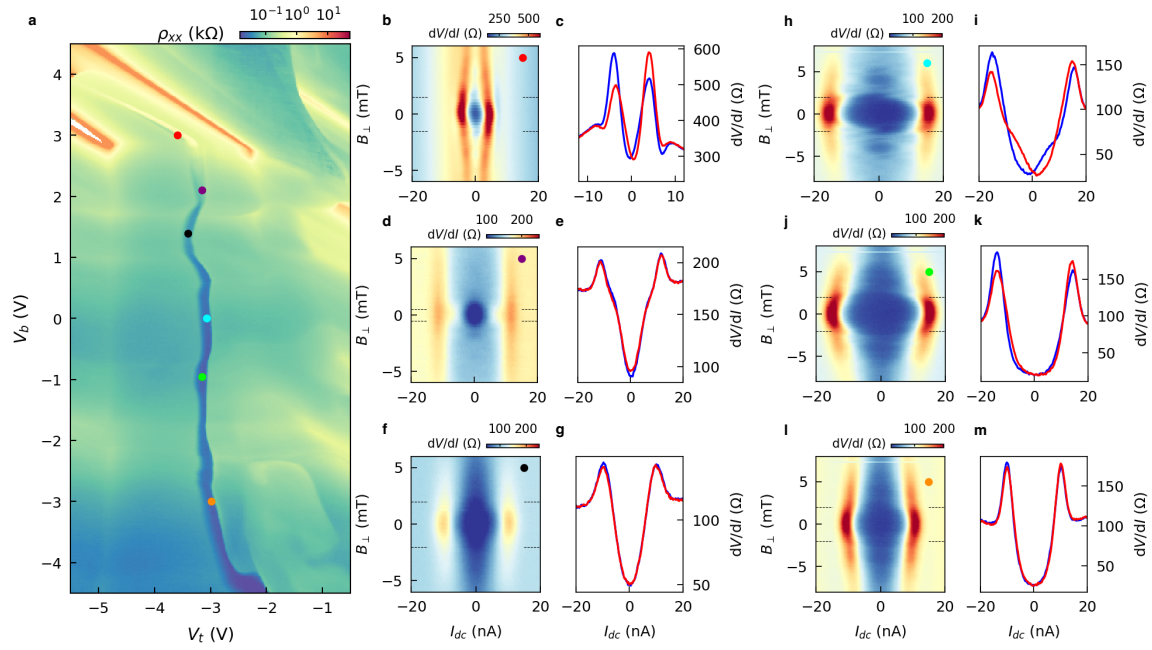


Figure C.5: Gate-tunable nonreciprocity of the paired state. **a**, Map of ρ_{xx} versus V_b and V_t taken at $B_{\parallel} = 1$ T. Map of dV/dI versus I_{dc} and $B_{\perp}$ taken at **b** $(V_b, V_t) = (3.00 \text{ V}, -3.59 \text{ V})$ (denoted by the red dot in **(a)**) with $B_{\parallel} = 1$ T. **c**, line cuts from **(b)** taken at ± 1.5 mT (blue and red respectively). **d**, dV/dI map taken at $(2.10 \text{ V}, -3.15 \text{ V})$ (denoted by the purple dot). **e**, line cuts from **(d)** taken at $B_{\perp} = \pm 0.5$ mT. **f**, dV/dI map taken at $(1.40 \text{ V}, -3.40 \text{ V})$ (denoted by the black dot). **g**, line cuts from **(f)** taken at $B_{\perp} = \pm 2$ mT. **h**, dV/dI map taken at $(0.0 \text{ V}, -3.07 \text{ V})$ (denoted by the cyan dot). **i**, line cuts from **(h)** taken at $B_{\perp} = \pm 2$ mT. **j**, dV/dI map taken at $(-0.95 \text{ V}, -3.15 \text{ V})$ (denoted by the green dot). **k**, line cuts from **(j)** taken at $B_{\perp} = \pm 2$ mT. **l**, dV/dI map taken at $(-3.00 \text{ V}, -2.98 \text{ V})$ (denoted by the orange dot). **m**, line cuts from **(l)** taken at $B_{\perp} = \pm 2$ mT.

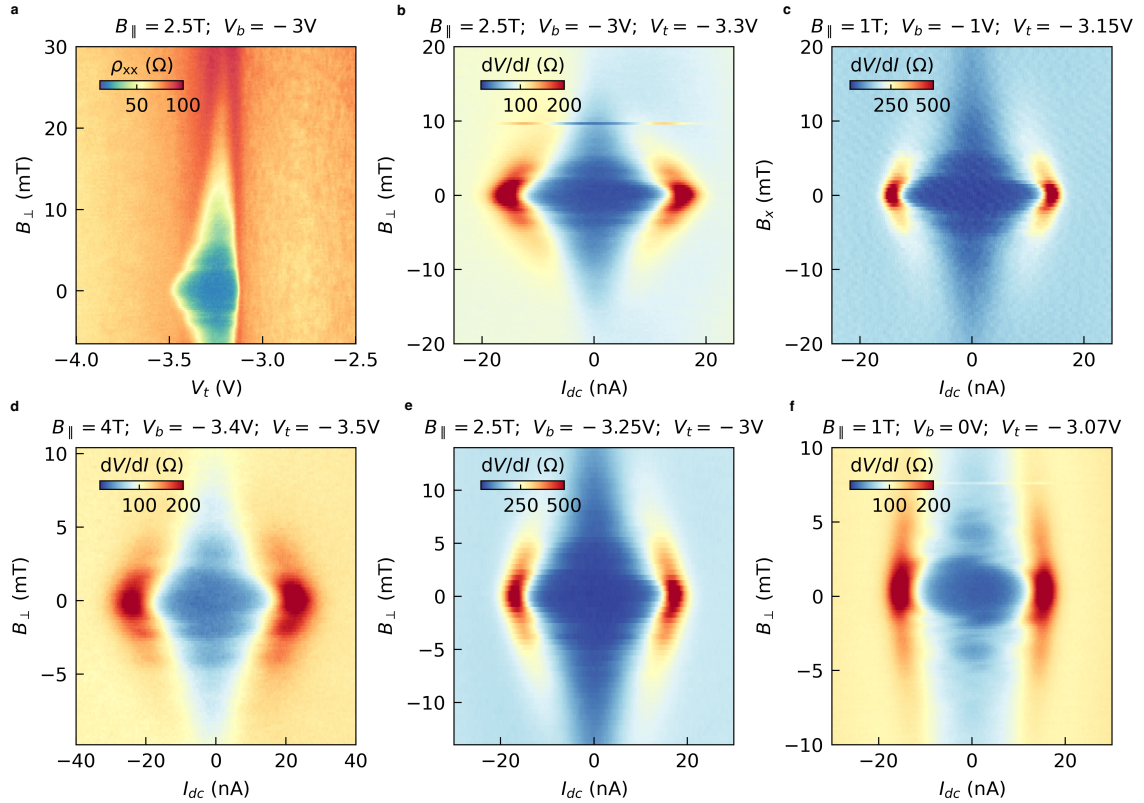


Figure C.6: Examples of Fraunhofer oscillations. **a**, Map of ρ_{xx} versus V_t and $B_{\perp}$ showing Fraunhofer oscillations in the superconducting state, taken at $B_{\parallel} = 2.5$ T and $V_b = -3.00$ V. Map of dV/dI versus $B_{\perp}$ and I_{dc} showing Fraunhofer oscillations taken at **b**, $B_{\parallel} = 2.5$ T, $V_b = -3.00$ V and $V_t = -3.30$ V, **c**, $B_{\parallel} = 1$ T, $V_b = -1.00$ V and $V_t = -3.15$ V, **d**, $B_{\parallel} = 4$ T, $V_b = -3.40$ V and $V_t = -3.50$ V, **e**, $B_{\parallel} = 2.5$ T, $V_b = -3.25$ V and $V_t = -3.00$ V, and **f**, $B_{\parallel} = 1.0$ T, $V_b = 0.00$ V and $V_t = -3.07$ V.

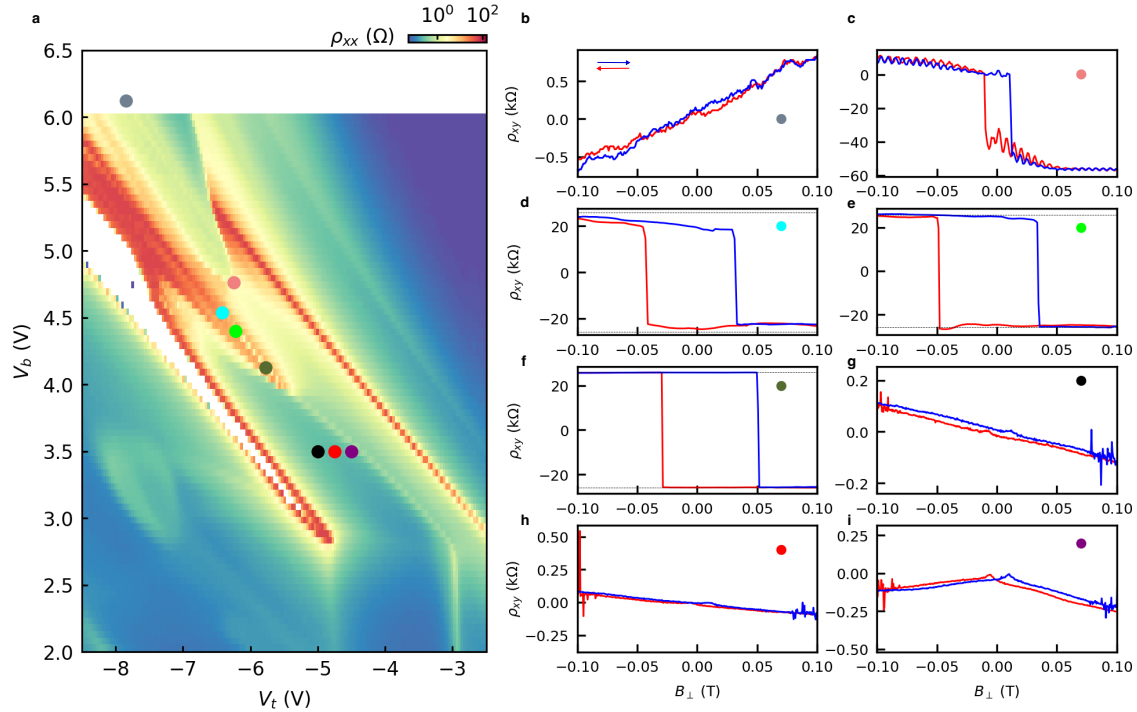


Figure C.7: **Assessment of magnetic hysteresis in the regime of an isolated conduction band.** **a**, map of ρ_{xx} versus V_t and V_b with colored dots showing where each hysteresis loop is taken. Measurement of ρ_{xy} vs $B_{\perp}$ swept in both directions (blue denotes the forward direction and red the backward), taken at **b**, $(V_t, V_b) = (-7.84 \text{ V}, 6.12 \text{ V})$, marked by the grey dot in **(a)**, **c**, $(-6.25 \text{ V}, 4.76 \text{ V})$, marked by the light red dot, **d**, $(-6.42 \text{ V}, 4.54 \text{ V})$, marked by the cyan dot, **e**, $(-6.22 \text{ V}, 4.40 \text{ V})$, marked by the lime dot, **f**, $(-5.77 \text{ V}, 4.12 \text{ V})$, marked by the dark green dot, **g**, $(-5.00 \text{ V}, 3.50 \text{ V})$, marked by the black dot, **h**, $(-4.75 \text{ V}, 3.50 \text{ V})$, marked by the red dot, and **i**, $(-4.50 \text{ V}, 3.50 \text{ V})$, marked by the purple dot. $B_{\parallel} = 0 \text{ T}$ for all the measurements.

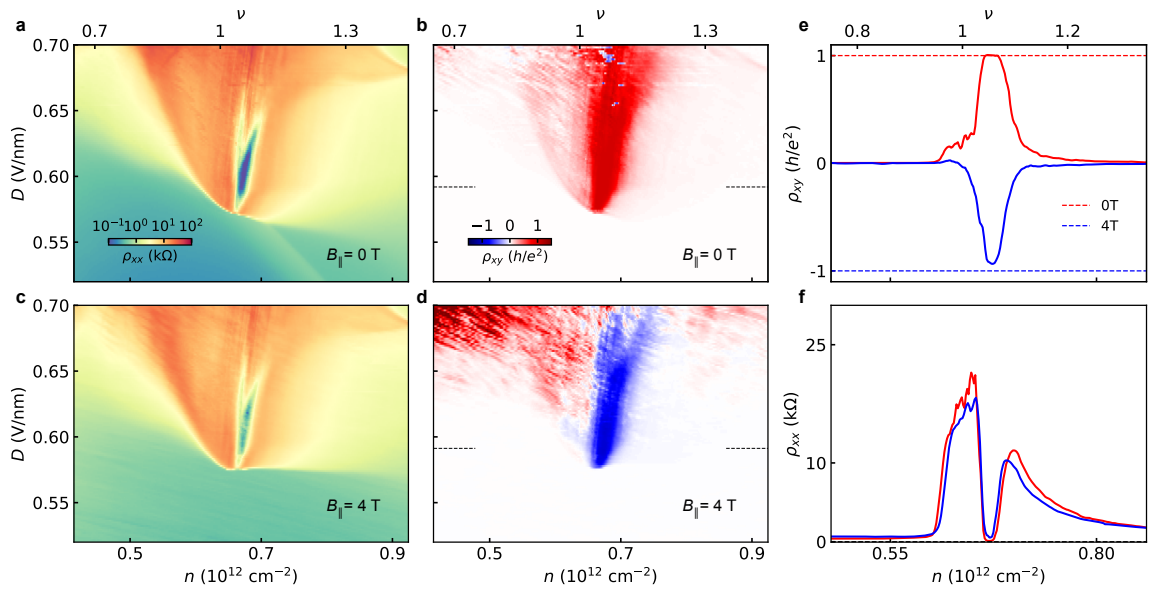


Figure C.8: **Integer quantum anomalous Hall state at different $B_{\parallel}$.** **a**, Map of ρ_{xx} versus D and n taken at $B_{\parallel} = 0$ T. **b**, Corresponding ρ_{xy} to **(a)**. **c**, Map of ρ_{xx} versus D and n taken at $B_{\parallel} = 4$ T. **d**, Corresponding ρ_{xy} to **(c)**. **e**, ρ_{xy} linecuts along the black dashed lines in **(b)** and **(d)**, taken at $D = 0.59$ V/nm. **f**, Corresponding ρ_{xx} to **(e)**. ρ_{xx} data shown in **(a-f)** is taken using V_{xx2} . All measurements are taken with $B_{\perp} = 0$.

A poem I wrote in the depths of my despair. Not to be taken too seriously

*Many an hour have I spent,
In droning noise and musty scent,
Under sanitized pale fluorescent light,
Trying to wage a futile fight.*

*Pushing through, to a new frontier,
If only I can add one more layer.
Groundbreaking! we reassure ourselves of what we do
Just make another device or maybe two.*

*So I drown myself, in wires and tapes,
Solvent bottles and graphite flakes.
To cut and clean, and stack and twist.
Sometimes I wonder what I've missed.*

*Fruitless labor to an unknown goal,
Is this why Faust sold his soul?
An impossible longing, too strong to smother,
Is it too late to be someone other?*