Imaging Quantum Hall Wavefunctions
With A Scanning Tunneling Microscope:
From Spontaneous Symmetry Breaking
To Interacting Domain Boundary Modes

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Abstract

Quantum materials provide a rich platform for realizing emergent phenomena, where interactions between electrons lead to properties that are strikingly different from those of individual electrons. Confining electrons to two dimensions and subjecting them to large magnetic fields enhances the effects of Coulomb interactions, which can manifest as a spontaneous lifting of spin, valley or other degeneracies in these quantum Hall systems. Experiments on a number of materials have primarily relied on global measurement techniques to explore this phase space, but locally imaging these broken-symmetry states has remained a challenge. With the use of a scanning tunneling microscope (STM), we directly visualize quantum Hall wavefunctions on the surface of a bismuth crystal, which enables us to probe the spatial signatures of spontaneous valley ordering. We identify the emergence of a nematic electronic phase, which breaks the rotational symmetry of the underlying crystal lattice, and a ferroelectric phase that carries an in-plane electric dipole moment. Furthermore, we use the STM to investigate one-dimensional channels that form at the boundary between different valley-polarized quantum Hall states. We find markedly different regimes where these channels are either metallic or insulating, depending on constraints imposed on electron-electron interactions by the valley flavor. These experiments set the stage visualizing fractional quasiparticles in quantum Hall systems, and more generally for using local imaging techniques in future explorations of novel interaction-driven phenomena.
Publications

Publications associated with this dissertation:


* these authors contributed equally
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For my family,

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Chapter 1

Introduction

1.1 An Overview: Imaging Quantum Behavior in Materials

Materials offer an exciting avenue through which to explore the mysteries of physics by providing a window into the microscopic quantum world. Quantum materials are the basis for some incredible physical phenomena, including magnetism, superconductivity and the fractional quantum Hall effect. These examples of so-called emergent phenomena originate from interactions among electrons and exhibit collective behavior that cannot be explained by non-interacting electrons. Several of these quantum many-body effects were first observed experimentally and subsequently understood theoretically, as it can be challenging to a-priori predict the consequences of interactions.

An especially rich source of novel, unexpected phenomena have come from two-dimensional electron gases (2DEGs) in high magnetic fields. The first of these remarkable discoveries was the integer quantum Hall effect, where the transverse Hall conductance is precisely quantized in integer units of fundamental constants \(\frac{e^2}{h}\), independent of sample-specific quantities [1]. Since then, experiments have revealed
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a diverse set of quantum phases in this regime - the fractional quantum Hall effect [2], exotic excitations [3], spontaneous broken-symmetry states [4], stripe and bubble phases [5, 6], to name a few.

These quantum Hall systems are a versatile platform for the study of emergent behavior, because they host highly degenerate flat bands that allow interaction effects to dominate. 2DEGs with additional degrees of freedom are an intriguing subset of quantum Hall systems in which Coulomb interactions can dictate behavior. These multicomponent systems have an internal degree of freedom associated with the electron’s spin, valley or layer index. This additional degeneracy can be spontaneously lifted by Coulomb interactions, giving rise to a multitude of broken-symmetry states [4]. This rich phase space has predominantly been probed using bulk measurements in a number of materials [7–12], but direct imaging of the underlying quantum states has remained a challenge. In this thesis, we visualize quantum Hall wavefunctions in a 2DEG arising from a bismuth surface state, which enables us to probe spatial signatures of valley symmetry breaking.

Initial quantum Hall experiments were typically performed in semiconducting heterostructures where the 2DEGs occur at buried interfaces, which are inaccessible to imaging by surface scanning probes. More recently, graphene [13–16], surface 2DEGs in doped semiconductors [17, 18], and surfaces of topological insulators [19, 20] have, in principle, paved the way for direct spatial imaging. Although local spectroscopic measurements have been performed in these materials, disorder has hindered the visualization of individual wavefunctions. Moreover, electron-electron interactions still compete with disorder in these systems and can also be screened by nearby metallic objects including gates or the probe itself. Thus, a central challenge towards the local imaging of quantum Hall physics is to obtain a suitable 2DEG which is
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compatible with a local probe that can function in this high magnetic field regime with the requisite spatial and energy resolution.

The scanning tunneling microscope (STM) is one such unique tool that is well suited for this purpose; it can operate in high magnetic fields and at millikelvin temperatures, and has atomic-scale spatial resolution that allows it to distinguish features on the relevant length scale of the magnetic length. By mapping the local density of states, STM also has the potential to directly visualize the underlying electron wavefunctions as well as the order parameters of certain broken symmetry states. Furthermore, local probe experiments can elucidate individual domains that are generically expected to form in these phases. This is in contrast to most transport measurements that average over microscopic configurations or rely on external polarizing fields to resolve global symmetry breaking.

In this thesis, we focus on one multicomponent quantum Hall system, with multiple valleys, that forms on the surface of bismuth. In this case, the valleys are anisotropic, which couples the internal valley index to the spatial symmetries of the crystal. The spatial signatures of these valley-ordered phases are central to our experiments, in which we use a STM to locally probe these broken-symmetry states. First, we directly visualize individual quantum Hall wavefunctions that are centered on atomic defects in our sample. These spatially resolved images reveal a nematic electronic state with broken rotational symmetry [21]. We then present experiments that explore a valley-polarized quantum Hall state with emergent ferroelectricity [22]. Energy resolved measurements clarify the role of Coulomb interactions in driving the spontaneous formation of these phases. Furthermore, this local imaging technique allows us to investigate a new class of interacting one-dimensional Luttinger liquid behavior at nematic domain boundaries in the interior of the sample [23].
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The rest of this introduction is organized as follows. We begin by discussing the integer quantum Hall effect in Section 1.2. Then, we consider the role of Coulomb interactions in lifting degeneracies in systems with internal degrees of freedom. In the last part of Section 1.2, we present a more detailed look at a specific type of quantum Hall system consisting of multiple anisotropic valleys. In Section 1.3, we describe how the surface states of bismuth form such a multi-valley system that is ideal for studying broken-symmetry quantum Hall phases using local imaging techniques. We further highlight some of the valley-ordered states expected for the bismuth surface. We end this Chapter by presenting an overview of our main results and an outline for the rest of this thesis in Section 1.4.

1.2 The Quantum Hall Effect and Consequences of Symmetry Breaking

1.2.1 The Integer Quantum Hall Effect

Historically, the classical Hall effect demonstrated a nonzero, transverse Hall resistance \( R_{xy} = V_y/I_x \) that increases linearly with increasing magnetic field applied out-of-plane (see Fig. 1.1a for experimental setup). This seminal measurement by Edwin Hall in 1879 was the first experiment to prove the existence of moving charges in a metal [24] and later (once the electron was discovered eighteen years later! [25]) served to measure the sign of charge carriers. The basis for this nonzero Hall resistance is cyclotron motion, shown schematically in Fig. 1.1b, in which the trajectories of moving charged particles curve in a perpendicular magnetic field [26].

One century later, careful measurements of the Hall effect in a high quality silicon 2DEG led to the discovery of the integer quantum Hall effect by von Klitzing et al.
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Figure 1.1: Experimental setup for Hall effect measurements. (a) Setup for measuring Hall effect of electrons, showing the directions of external magnetic field $B$ and applied current $I_x$. Electron trajectories follow the curved line, which leads to a measured nonzero Hall voltage $V_y$. (b) Circular cyclotron motion of classical charged particle moving in two dimensions in the presence of a perpendicular magnetic field $B$.

Figure 1.2: Integer quantum Hall effect. (a) Experimental data of Hall resistance $R_{xy}$ and longitudinal resistivity $\rho_{xx}$ as a function of magnetic field $B$, for a fixed carrier density $n$. $R_{xy}$ shows plateaus for integers $i$ at values $h/e^2i$. Reproduced from Ref. [27]. (b) Schematic of the Hall conductivity $\sigma_{xy}$ (red) and $\rho_{xx}$ (blue) as a function of energy $E$ for a fixed magnetic field $B$. The Hall conductivity is quantized for integer values of $e^2/h$, and each plateau corresponds to a different Landau level.
in 1980 [1]. Instead of a linear dependence on magnetic field, they observed that the Hall resistance exhibited plateaus at specific values over a wide range of magnetic fields (Fig. 1.2a). The corresponding Hall conductivity $\sigma_{xy} = \nu e^2/h$ is quantized at integer values of $\nu$ (Fig. 1.2b). The concurrent longitudinal resistivity $\rho_{xx}$ is zero, except at each step between plateaus where it is sharply peaked. The quantization of $\sigma_{xy}$ occurs with incredible accuracy, independent of microscopic details of the system.

Unlike the classical expectation, this quantized Hall conductance is governed by quantum mechanics, which discretizes the energies of these cyclotron orbits into highly degenerate flat bands known as Landau levels [4, 27]. Entering this quantum world requires low temperatures in order to resolve the energy quantization and extremely clean samples where the time to complete a cyclotron orbit is much smaller than the scattering time. Higher magnetic fields can be used to both increase the frequency of cyclotron motion and increase the energy spacing between Landau levels.

There are deep connections between the universality of the quantized Hall conductance and topology in these systems. The integer quantum Hall effect can be characterized by a topological invariant, known as the Chern number, that is always an integer and equal to the quantized value of $\sigma_{xy}$ in units of $e^2/h$ [28]. As this Chern number is a discrete topological index that cannot change continuously, small perturbations cannot change the Hall conductance. Moreover, the bulk-boundary correspondence of these topological states has important consequences. First, even in a classical picture with cyclotron orbits, at the sample edges, these orbits must collide with the boundary. This results in ‘skipping orbits’ (Fig. 1.3a) that propagate only in a single direction at the one-dimensional edge. Quantum mechanically, the sharp edge potential between the sample and vacuum gives rise to chiral edge modes that travel in opposite directions at opposite edges (Fig. 1.3b) [4, 27]. These chiral modes exist due to the topological nature of the underlying 2D quantum Hall states and
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Figure 1.3: Quantum Hall edge modes. (a) Semi-classical view of skipping orbits at the sample edges that propagate in only one direction. The circular orbits in the middle of the sample carry no net current. (b) Chiral edge modes expected at the sample boundary for a $\nu = 1$ quantum Hall state, due to its underlying topological nature.

are immune to scattering from impurities; for a left-moving electron to scatter into a right-moving electron, it would have to traverse the entire bulk of the sample. The number of chiral edge modes corresponds to the integer number $\nu$ of the quantized Hall conductance.

1.2.2 Quantum Hall Ferromagnetism

We now consider 2DEGs in which the electrons have an internal degree of freedom that adds an additional degeneracy to the Landau levels. This degree of freedom may be associated with the electron spin, or a valley pseudospin in the case of multiple degenerate conduction band extrema, or a layer index. Although at times we refer to the intrinsic electronic states as spins, the discussion is not limited to fundamental electron spins or to having only two degrees of freedom. We can generically consider a system with $g$ degrees of freedom for integer $g \geq 2$. In the absence of interactions, the quantized Hall plateaus will only occur at integer multiples of $g$. However, Coulomb interactions can spontaneously lift this degeneracy to produce additional plateaus, when an integer number of Landau levels, $\nu < g$, are occupied in the degenerate subspace.
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Figure 1.4: **Spontaneous symmetry breaking in Landau levels.** (a) Density of states (DOS) for Landau levels in the absence of interactions. The Landau levels have an added degeneracy due to the internal degrees of freedom. (b) When a Landau level is partially filled, Coulomb interactions lift the spin degeneracy, opening up an exchange gap at the Fermi level \((E = 0)\) between the two spin types.

The exchange component of the Coulomb interactions favors the polarization of the intrinsic degrees of freedom, resulting in spontaneous symmetry breaking known as quantum Hall ferromagnetism [4]. Occupying all of one spin type ensures that the spatial many-body wavefunction is anti-symmetric, a consequence of the Pauli exclusion principle. This minimizes the overlap between electrons with the same spin state, which therefore lowers their Coulomb repulsion and creates an energy gap between the two spin types (Fig. 1.4). Thus the spins spontaneously polarize, producing a state with quantized Hall conductance that breaks an internal symmetry of the system.

Quantum Hall ferromagnetism has further implications for low energy excitations in these systems. In the case of an SU(2) symmetry, the excitations are topologically nontrivial spin textures known as skyrmions [29]. Instead of naively adding an up-spin to a sea of down-spins, rotating the spins around the added up-spin electron costs a lower exchange energy. These skyrmion configurations are also of interest to explore experimentally using local imaging techniques, but is not a topic explored in this thesis.
Figure 1.5: **Valley nematic model system.** (a) Model momentum-space band structure consisting of two anisotropic Fermi surfaces, known as valleys; schematic here represents AlAs Brillouin zone. These elliptical valleys are inequivalent because they are related to each other by a 90° rotation. (b) Real space LL orbitals, which show two different orientational orders. This two-fold nematic order breaks the four-fold rotational symmetry of the original Fermi surface, which can arise from occupying one of the two distinct valleys. Adapted from Ref. [31].

### 1.2.3 Quantum Hall Valley Nematics

In this subsection, we present the consequences of quantum Hall ferromagnetism involving the valley degree of freedom, specifically in multicomponent systems with anisotropic valleys [30, 31]. The different valleys correspond to extrema in the electronic band structure that are centered at different momenta in the Brillouin zone. The Fermi surface of a model system with multiple anisotropic valleys (or pockets) is shown in Fig. 1.5a. The valley ellipticity is due to different effective masses in the $k_x$ and $k_y$ directions. These two inequivalent valleys are related by discrete rotations, which naturally intertwines the valley degree of freedom with the spatial symmetries of the system. Here, spontaneous quantum Hall ferromagnetism can produce states in which only a subset of the valleys are occupied. The corresponding real-space valley-polarized wavefunctions reflect the anisotropy of the occupied valley, as illustrated in Fig. 1.5b. The resultant electronic states form a nematic phase, which breaks the rotational symmetry of the underlying crystal.
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Valley nematic order in the quantum Hall regime is possible in a wide variety of materials. Examples of candidate 2DEGs with highly anisotropic valleys include conventional semiconductors such as AlAs [9] and Si [10], the semimetal bismuth [21], as well as certain topological insulators including SnTe [32, 33] and Pb_xSn_{1−x}Se [34]. These systems are not restricted to only two valleys, and can have symmetries that are three-, four- or six-fold degenerate. These complex multivalley 2DEGs have both been studied experimentally and form the basis for theoretical proposals within the realm of quantum Hall valley nematics. More broadly, the interplay between interactions and valleys in the quantum Hall regime also has implications for van der Waals materials including graphene [35–37] and transition metal dichalcogenides [38]. These different classes of material systems offer a setting for the emergence of a multitude of broken-symmetry phases.

This nematic ordering is especially sensitive to spatial inhomogeneities and therefore is generically expected to break up into domains. These valley-polarized domains are predicted to host gapless one-dimensional modes - that is, quantum channels that emerge because of a topological change in the underlying electronic wavefunctions at such interfaces (Fig. 1.6). Transport measurements are not well suited for isolating the response of these boundaries or individual nematic domains. However, for materials accessible to local probes, a combination of spectroscopy and spatial mapping can be used to identify such domain walls and distinguish the behavior at the boundary from that in the bulk. The pristine one-dimensional boundaries between two dimensional phases are an exciting platform in which to explore enhanced interaction effects and 1D Luttinger liquid physics [39]; valley nematic systems offer additional novelty in studying how the valley degree of freedom influence these interactions [23].
Figure 1.6: Valley-polarized domain wall. (a) Schematic of a domain wall where a different subset of valleys are occupied in different regions of the sample (in real space), as denoted by the Landau orbit orientations (ellipses). The quantum Hall edge modes have different valley flavors (red vs. blue), and are expected to be counterpropagating at the domain boundary. (b) Energy diagram of the different valley states, depicting the switch in valley occupation across the domain wall.

1.3 The Bismuth Surface and Possible Valley Ordering

We now discuss quantum Hall ferromagnetism as it applies to the bismuth surface, which offers an especially rich arena to explore various spontaneous valley ordering. Bismuth has several key properties that make it an ideal material in which to probe such interacting behavior using local imaging techniques. Most importantly, the bismuth surface state forms a multi-valley 2DEG that is relatively decoupled from the bulk and is accessible to local spectroscopic probes. It is a semimetal with low carrier density, which makes it possible to enter the quantum Hall regime. The semimetallic bulk may also play a role in offsetting screening from the STM tip. Moreover, bismuth is an elemental material that can be made with very low impurity concentrations, and is known to be one of the cleanest electronic systems with a bulk mean free path reaching 1 mm at low temperatures [40].

Here we study the surface states of bismuth (Bi) on the (111) surface, a natural cleavage plane for the crystals. Bismuth is a heavy element with large spin-orbit
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Figure 1.7: Bi(111) Fermi surface. (a) Fermi surface of Bi(111) surface state, with multiple hole (blue) and electron (red) valleys. Our experiments focus on the six anisotropic valleys shown in blue. (b) Experimentally measured Bi(111) Fermi surface using angle-resolved photoemission spectroscopy (ARPES). Reproduced from Ref. [43].

coupling. This spin-orbit coupling is responsible for producing spin-split surface state bands with multiple degenerate valleys, while eliminating the spin degree of freedom [41, 42]. The resulting Bi(111) Fermi surface is shown in Fig. 1.7. In this thesis, we primarily focus on the six anisotropic valleys along the Γ-M directions (colored in blue). There are additional valleys - the central valley at the Γ point and the elongated valleys around the M points (all labeled in red), which are not the subject of the work presented here. The six valleys in bismuth are the foundation for our investigation of valley nematicity and other quantum Hall phases with spontaneous symmetry breaking.

We now present an overview of the array of possible broken-symmetry states for the Bi(111) surface in the quantum Hall regime. In particular, we highlight the spatial signatures of the various valley ordered phases as expected to be seen with STM mapping. The following discussion also serves as a preview of certain experiments presented in this thesis.

In the absence of any valley symmetry breaking, all the six valleys are degenerate. A simulation of the real space Landau level wavefunction is shown in Fig. 1.8a, and
Figure 1.8: Possible nematic ordering in bismuth. Within each panel, left: relevant valleys of Bi(111) Fermi surface. Colored boxes denote which valleys are degenerate (i.e. at the same energy). Right: Numerical simulations of corresponding real-space Landau level (LL) wavefunctions ($|\psi|^2$) in the symmetric gauge. The wavefunction orientation is determined by the anisotropy of degenerate valleys. (a) In the absence of any symmetry breaking, all six valleys are degenerate. The LL wavefunction retains the three-fold rotational symmetry of system. (b)-(c) Valley degeneracy lifted either into a four-fold and a two-fold degenerate state (b) or into three doubly degenerate states (c), resulting in LL wavefunctions with different orientations for each of the broken-symmetry states. These real-space wavefunctions break the original three-fold rotational symmetry, a manifestation of valley nematic order. Note: the Fermi surface is rotated 90° relative to the real space maps.

consists of superimposed ellipses with three directionalities. It illustrates that the electronic wavefunction retains the three-fold rotational symmetry of the original crystal and Fermi surface. A lifting of the valley degeneracy either into a four-fold and a two-fold degenerate state or into three doubly degenerate states results in LL wavefunctions with different orientations for each of the broken-symmetry states (Fig. 1.8b-c). These real-space wavefunctions are a direct manifestation of the expected valley nematic order, as they break the underlying rotational symmetry of the system [21].
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Furthermore, in the case when a single valley is occupied, the valley-polarized state is expected to carry an in-plane electric dipole moment. Upon close inspection, an individual valley has a characteristic ‘teardrop shape’ instead of being perfectly elliptical (refer back to Fig. 1.7). This absence of two-fold rotational symmetry around the valley center implies that the wavefunctions are not inversion symmetric. Thus, the Landau orbits of a single valley-polarized state is expected to have an intrinsic in-plane dipole moment, giving rise to an emergent quantum Hall ferroelectric phase [22].

1.4 Thesis Outline

The overarching theme of this dissertation is the use of STM as an experimental probe to image and characterize interaction-driven behavior in a quantum Hall valley system. In Chapter 2 we introduce some theoretical underpinnings of the quantum Hall effect. Next, we describe the technique of scanning tunneling microscopy in Chapter 3, including measurement and instrumentation details. The STM used for all experiments presented in this thesis operates at a temperature of 250 mK and at magnetic fields up to 14 T, with 100 $\mu$eV energy resolution and Angstrom-scale spatial resolution.

The subsequent chapters discuss the main experimental results, which are highlighted below. In Chapter 4 we present a direct manifestation of a nematic quantum Hall state on the Bi(111) surface. STM spectroscopy shows a lifting of the valley degeneracy due to both single-particle strain and Coulomb interactions, which gives rise to different broken-symmetry states. We then use the STM to locally probe the spatial signatures of these valley-ordered phases. We directly image individual Landau level wavefunctions that are centered on atomic defects in
our sample, as detailed in the second half of the chapter. We find elliptical orbits with distinct orientations at the energies of the different broken-symmetry states. These spatially resolved images establish a nematic electronic state that breaks the rotational symmetry of the original system. Chapter 5 examines a further symmetry breaking, which reveals a valley-polarized state that is expected to be a quantum Hall ferroelectric due to the absence of two-fold rotational symmetry in an individual valley.

We then take advantage of our local imaging approach to investigate a boundary between different nematic, valley-polarized domains, discussed in Chapter 6. Here we find unique interaction effects among the one-dimensional boundary modes that are constrained by the valley degree of freedom. Such symmetry constraints cannot emerge in a purely one-dimensional system, but are a consequence of these domain walls forming between distinct quantum Hall states in two dimensions. We further demonstrate that these boundaries host a new class of symmetry-protected Luttinger liquids. We close with an outlook in Chapter 7, in which we touch on prospects of future imaging experiments. We discuss expanding this approach to other material platforms and to new interacting regimes including fractional quantum Hall states.
Chapter 2

Theoretical Background: The Integer Quantum Hall Effect

2.1 Landau Level Energy Spectrum

In two dimensions, classical electrons subjected to an out-of-plane magnetic field follow circular cyclotron orbits. When the system is treated quantum mechanically, the energy spectrum of these orbits gets quantized. Consider an electron with effective mass $m^*$ and charge $-e$ moving in the $x$-$y$ plane in the presence of a magnetic field $\mathbf{B} = B\hat{z}$. To account for the magnetic field in the Hamiltonian, the canonical momentum $\mathbf{p} = -i\hbar \nabla$ is replaced by the kinetic momentum $\pi = -i\hbar \nabla + e\mathbf{A}$, where $\mathbf{A}$ is the vector potential given by $\mathbf{B} = \nabla \times \mathbf{A}$. For a 2DEG with parabolic dispersion, the Hamiltonian is given by

$$\mathcal{H} = \frac{1}{2m^*} (\mathbf{p} + e\mathbf{A})^2. \quad (2.1)$$

If we choose the Landau gauge, $(A_x = 0, A_y = Bx)$, translational invariance is preserved in the $y$-direction. The eigenstates of $\mathcal{H}$ can then be labeled by their $y$ momentum, $k_y$, and the problem reduces to a one-dimensional quantum harmonic oscillator [4]. Solving this harmonic oscillator leads to energy levels that are labeled
CHAPTER 2. THEORETICAL BACKGROUND: THE INTEGER QUANTUM HALL EFFECT

by a second quantum number, a discrete orbital index $N$. These energy levels are known as Landau levels (LLs), and have a spectrum

$$E_N = \hbar \omega_c (N + 1/2),$$

(2.2)

where $\omega_c = eB/m^*$ is the cyclotron frequency and $N = 0, 1, 2, \ldots$. In the Landau gauge, the wavefunctions are written as

$$\psi_{N,k_y} = \frac{1}{L_y} e^{ik_y y} \varphi_N(x + k_y \ell_B^2),$$

(2.3)

where $\ell_B = \sqrt{\hbar/eB}$ is the magnetic length, $\varphi_N(x) = \frac{1}{\sqrt{2^N N! \pi^{1/2}}} H_N \left( \frac{x}{\ell_B} \right) e^{-x^2/2\ell_B^2}$, $H_N$ is the $N^{th}$ order Hermite polynomial, and $L_x$ and $L_y$ are the sample dimensions. The wavefunction is localized around a guiding center coordinate $X_{k_y} = -k_y \ell_B^2$.

Each LL has a large degeneracy because although the wavefunction depends on two quantum numbers $N$ and $k_y$, the energy only depends on $N$. We can determine the degeneracy as follows: the finite size $L_y$ quantizes the momentum $k_y$ in units of $2\pi/L_y$. In the $x$-direction, the wavefunctions are exponentially localized around $x = -k_y \ell_B^2$. A finite sample, $0 \leq x \leq L_x$, further restricts $k$ to the range $-L_x/\ell_B^2 \leq k \leq 0$. This results in a degeneracy

$$N = \frac{L_x L_y}{2\pi \ell_B^2} = \frac{BL_x L_y}{\hbar/e},$$

(2.4)

By introducing the notion of the flux quantum $\Phi_0 \equiv \hbar/e$, the degeneracy $N = BA/\Phi_0$ is given by the number of flux quanta threading the sample [27]. From the degeneracy, we can then define the filling factor $\nu$, which is the number of filled Landau levels. This filling factor can be written in terms of the total number of electrons $N_e$ as $\nu = N_e/N$, or in terms of the density of the 2DEG, $n$, as $\nu = nh/eB$. 

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2.2 The Role of Disorder

The integer quantum Hall effect can only be observed in sufficiently clean samples, in which the scattering time, $\tau$, is much longer than the time to complete a cyclotron orbit, i.e. $\omega_c \tau \gg 1$, that can be achieved in the limit of high magnetic fields. However, the existence of quantization and extended plateaus in the Hall conductance can only be explained in the presence of disorder. This is because for a fixed electron density $n$, the LL is completely filled at exactly $B = n\Phi_0/\nu$. In the absence of disorder, changing $B$ away from this value results in a partially filled LL, so there are accessible states for the electrons to scatter into, which leads to a non-zero longitudinal conductivity and a Hall conductivity that is no longer quantized.

The effect of disorder can be understood using a semi-classical model of electron dynamics in a smooth random potential [4, 27]. Essentially, the centers of the cyclotron orbits drift along equipotential lines. Disorder broadens the spectrum around the LL energies, turning many of the quantum states from extended to localized, an important distinction for conductivity. Only the extended states contribute to the transport of charge. Once all the extended states in a given LL are filled, as $B$ is decreased for fixed $n$, the localized states start to get populated. Since these states don’t contribute to the current, the conductivity remains unchanged and results in the observed quantum Hall plateaus with constant conductivity over a range of magnetic field. Thus, although quantum Hall samples have to be extremely clean, in practice all experimental realizations are disordered and it is this small amount of disorder, but not too much, that is needed to observe the quantized Hall conductivity.
CHAPTER 2. THEORETICAL BACKGROUND: THE INTEGER QUANTUM HALL EFFECT

2.3 Landau Levels in the Symmetric Gauge

Below, we outline the Landau level derivation where the energy spectrum can be solved using a purely algebraic method without specifying a gauge. Calculating wavefunctions in terms of the positions of electrons, however, requires an explicit choice of gauge. For this, we work in the symmetric gauge:

\[ A = -\frac{1}{2} \mathbf{r} \times \mathbf{B} = \frac{B}{2} (-y, x, 0). \]  

(2.5)

This gauge choice breaks translational symmetry in both the \( x \) and \( y \) directions, but preserves rotational symmetry about the origin, making angular momentum a good quantum number. The discussion below draws from Refs. [27, 44], which contain additional details.

2.3.1 An Algebraic Approach

In the presence of a magnetic field, the canonical momentum \( p = -i\hbar \nabla \) is replaced by the kinetic momentum

\[ \pi = -i\hbar \nabla + eA, \]  

(2.6)

for a uniform magnetic field \( \hat{z} \cdot (\nabla \times A) = B \). We can arrive at the harmonic oscillator form of the Hamiltonian by using the commutation relations of the kinetic momentum

\[ [\pi_x, \pi_y] = -i\hbar e \hat{z} \cdot (\nabla \times A) = -i \frac{\hbar^2}{\ell_B^2}. \]  

(2.7)
CHAPTER 2. THEORETICAL BACKGROUND: THE INTEGER QUANTUM HALL EFFECT

From this, we introduce raising and lowering operators, analogous to those in the quantum harmonic oscillator, defined as

\[
a = \frac{1}{\sqrt{2}} \ell_B \frac{\pi_x - i\pi_y}{\hbar}, \quad a^\dagger = \frac{1}{\sqrt{2}} \ell_B \frac{\pi_x + i\pi_y}{\hbar},
\]

which obey

\[
[a^\dagger, a] = 1.
\]

In terms of these ladder operators, the Hamiltonian takes the form:

\[
\mathcal{H} = \frac{1}{2m^*} \pi \cdot \pi = \hbar \omega_c \left( a^\dagger a + \frac{1}{2} \right).
\]

To account for the degeneracy within a particular Landau level, we construct a second pair of conjugate operators that commute with the Hamiltonian. The Landau level degeneracy can be obtained by considering the following pseudo-momentum operator

\[
\tilde{\pi} = p - eA.
\]

This pseudo-momentum differs from the kinetic momentum by the minus sign and is not gauge invariant. The commutator of \( \tilde{\pi} \) are given by

\[
[\tilde{\pi}_x, \tilde{\pi}_y] = \frac{i}{\ell_B} \nabla^2.
\]

The commutators between \( \pi \) and \( \tilde{\pi} \) highlight the lack of gauge invariance

\[
[\pi_x, \tilde{\pi}_x] = 2ie\hbar \frac{\partial A_x}{\partial x}, \quad [\pi_y, \tilde{\pi}_y] = 2ie\hbar \frac{\partial A_y}{\partial y}, \quad [\pi_x, \tilde{\pi}_y] = [\pi_y, \tilde{\pi}_x] = ie\hbar \left( \frac{\partial A_x}{\partial y} + \frac{\partial A_y}{\partial x} \right).
\]
CHAPTER 2. THEORETICAL BACKGROUND: THE INTEGER QUANTUM HALL EFFECT

However, in the symmetric gauge, all these commutators vanish to give

\[ [\pi_i, \tilde{\pi}_j] = 0. \] (2.14)

We can now define a second set of raising and lowering operators

\[ b = \frac{1}{\sqrt{2}} \frac{\ell_B}{\hbar} (\tilde{\pi}_x + i \tilde{\pi}_y) \quad b^\dagger = \frac{1}{\sqrt{2}} \frac{\ell_B}{\hbar} (\tilde{\pi}_x - i \tilde{\pi}_y) \] (2.15)

which also obey

\[ [b, b^\dagger] = 1. \] (2.16)

This second pair of ladder operators produce the degenerate set of eigenstates within a Landau level (i.e. with a given allowed kinetic energy). The complete set of Landau level wavefunctions can be generated by using both sets of raising and lowering operators:

\[ |N, m\rangle = \frac{(a^\dagger)^N (b^\dagger)^m}{\sqrt{N! m!}} |0,0\rangle. \] (2.17)

2.3.2 Landau Level Wavefunctions

We now construct the Landau level wavefunctions in the symmetric gauge. We introduce the complex coordinate \( z = x + iy \) and its derivative \( \frac{\partial}{\partial z} = \frac{1}{2} \left( \frac{\partial}{\partial x} - i \frac{\partial}{\partial y} \right) \), and use the bar to indicate complex conjugation. The ladder operators can then be written in the form

\[ a = \frac{-i}{\sqrt{2}} \left( 2\ell_B \frac{\partial}{\partial z} + \frac{1}{2\ell_B} \bar{z} \right), \quad a^\dagger = \frac{i}{\sqrt{2}} \left( -2\ell_B \frac{\partial}{\partial \bar{z}} + \frac{1}{2\ell_B} z \right) \] (2.18)

\[ b = \frac{i}{\sqrt{2}} \left( 2\ell_B \frac{\partial}{\partial \bar{z}} + \frac{1}{2\ell_B} z \right), \quad b^\dagger = \frac{i}{\sqrt{2}} \left( -2\ell_B \frac{\partial}{\partial z} + \frac{1}{2\ell_B} \bar{z} \right) \] (2.19)
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The lowest Landau level wavefunctions $\psi_{N=0,m}(z, \bar{z})$ are those which are annihilated by the differential operator $a$. The lowest state $\psi_{N=0,m=0}$ is the one that is annihilated by both $a$ and $b$, given by

$$\psi_{0,0} = \frac{1}{\sqrt{2\pi \ell_B^2}} e^{z\bar{z}/4\ell_B^2}. \quad (2.20)$$

The set of other states in the lowest Landau level can then be generated by repeated application of $b^\dagger$:

$$\psi_{0,m} = \frac{1}{\sqrt{2\pi \ell_B^2 2^m m!}} \bar{z}^m e^{-z\bar{z}/4\ell_B^2}. \quad (2.21)$$

In this particular basis, the eigenstates of the lowest Landau level are also eigenstates of the canonical angular momentum, defined as

$$L_z = xp_y - yp_x = \hbar \left( z \frac{\partial}{\partial z} - \bar{z} \frac{\partial}{\partial \bar{z}} \right) \quad (2.22)$$

Therefore,

$$L_z \psi_{0,m} = -\hbar m \psi_{0,m}. \quad (2.23)$$

However, this canonical angular momentum $L_z$ is \textit{gauge-dependent} and does not admit a direct physical interpretation. In particular, the states with higher $m$ are not rotating faster. However, the kinetic angular momentum, a gauge-independent quantity,

$$K = x\pi_y - y\pi_x \quad (2.24)$$

describes the physical angular momentum of the Landau level, where states with higher orbital index $N$ can be thought of as rotating faster [27].
To write down wavefunctions for higher Landau levels, we need to act with the raising operator $a^\dagger$. The corresponding wavefunctions are \[ \psi_{N,m}(Z) = \frac{1}{\sqrt{2\pi\ell_B^2}} \left( \frac{N!}{m!} \right)^{1/2} \left( \frac{Z^*}{\sqrt{2}} \right)^{m-N} e^{-|Z|^2/4} L_N^{m-N} \left( \frac{|Z|^2}{2} \right), \quad m \geq N \quad (2.25) \]
\[ \psi_{N,m}(Z) = \frac{1}{\sqrt{2\pi\ell_B^2}} \left( \frac{m!}{N!} \right)^{1/2} \left( \frac{Z}{\sqrt{2}} \right)^{N-m} e^{-|Z|^2/4} L_m^{N-m} \left( \frac{|Z|^2}{2} \right), \quad m < N. \quad (2.26) \]

Here $Z = (x+iy)/\ell_B$, $N = 0, 1, 2, ...$ is the orbital index, $m = 0, 1, 2, ...$ is the quantum number denoting the guiding center index, which labels the degeneracy within a given orbital index of the Landau level and $L_N$ is the Laguerre polynomial.

### 2.3.3 Guiding Centers: Connection to Classical Equations of Motion

The general solution to the classical equation of motion for a charged particle in a magnetic field is given by
\[ x(t) = X - R \sin (\omega_c t + \phi) \]
\[ y(t) = Y + R \cos (\omega_c t + \phi). \]

To connect this semiclassical approach to the quantum states in a Landau level, we treat the coordinates that label the orbit center $(X,Y)$ as quantum operators. This guiding center can be rewritten as
\[ X = x - \frac{\dot{y}}{\omega_c} = x - \frac{\pi y}{eB} \]
\[ Y = y + \frac{\dot{x}}{\omega_c} = y - \frac{\pi x}{eB}. \]
CHAPTER 2. THEORETICAL BACKGROUND: THE INTEGER QUANTUM HALL EFFECT

This definition of the guiding center \((X,Y)\) holds in any gauge. If we now replace \(x\) and \(y\) with the symmetric gauge potential, and use the expression for the pseudo-momentum, we end up with \(27\)

\[
X = -\frac{\tilde{\pi}_y}{eB} \quad \text{and} \quad Y = \frac{\tilde{\pi}_x}{eB}.
\] (2.31)

Thus, we can conclude that the pseudo-momentum corresponds to constants of motion in terms of the guiding center. The commutator relationship between guiding center components,

\[
[X,Y] = i\ell_B^2
\] (2.32)

means that we cannot localize states in both the \(X\) and \(Y\) coordinates, as per the Heisenberg uncertainty principle.

In the symmetric gauge, the profiles of the wavefunctions form concentric rings around the origin. The higher the pseudo-angular momentum, the larger the wavefunction radius. By expressing the complex coordinates for the position vector in terms of these ladder operators, it is possible to calculate size of the LL wavefunctions in this gauge \(44\). For large \(m\) these orbitals are strongly localized to within \(\sim \ell_B\) of a ring with a radius \(r_{N,m} = \ell_B\sqrt{2(N + m + 1)}\). This is very different from the strip-like wavefunctions in the Landau gauge. At this point, the profile of the wavefunctions is not gauge invariant and does not provide any physical information about the system.

2.4 Electron-Electron Interactions

We now discuss the consequences of electron-electron interactions, which we treat using the Hartree-Fock approximation \(45\). Here, the true many-body \(N\)-electron
wavefunction $\Psi$ is approximated by a product of single-particle orbitals $\psi_i(\mathbf{r}_i, s_i)$, where $\psi_i(\mathbf{r}_i, s_i)$ is composed of a spatial function $\phi_i(\mathbf{r}_i)$ and an electron spin function $\sigma(s_i)$ such that

$$\psi_i(\mathbf{r}_i, s_i) = \phi_i(\mathbf{r}_i)\sigma(s_i).$$

The Hartree-Fock wavefunction can be written as a Slater determinant, which is antisymmetric under the interchange of two particle coordinates, as required by the Pauli exclusion principle:

$$\Psi_{HF} = \frac{1}{\sqrt{N!}} \begin{vmatrix} \psi_1(\mathbf{r}_1 s_1) & \psi_1(\mathbf{r}_2 s_2) & \ldots & \psi_1(\mathbf{r}_N s_N) \\ \psi_2(\mathbf{r}_1 s_1) & \psi_2(\mathbf{r}_2 s_2) & \ldots & \psi_2(\mathbf{r}_N s_N) \\ \vdots & \vdots & \ddots & \vdots \\ \psi_N(\mathbf{r}_1 s_1) & \psi_N(\mathbf{r}_2 s_2) & \ldots & \psi_N(\mathbf{r}_N s_N) \end{vmatrix}.$$  \hspace{1cm} (2.34)

The orbitals are subject to the usual orthonormal constraint,

$$\int d\mathbf{r} \psi_i^*(\mathbf{r}) \psi_j(\mathbf{r}) = \delta_{ij}. \hspace{1cm} (2.35)$$

The interaction energy can be evaluated by taking the expectation values of the above Slater determinant with the Coulomb potential for

$$V_C = \frac{e^2}{4\pi\epsilon |\mathbf{r} - \mathbf{r}'|}, \hspace{1cm} (2.36)$$

for dielectric constant $\epsilon$. The interaction energy contains the direct (Hartree) Coulomb repulsion

$$E_{\text{direct}} = \frac{1}{2} \sum_{i,j} \int d\mathbf{r} d\mathbf{r}' \ V_C \ |\psi_i(\mathbf{r})|^2 |\psi_j(\mathbf{r}')|^2, \hspace{1cm} (2.37)$$
and the exchange (Fock) term

\[ E_{\text{exch}} = -\frac{1}{2} \sum_{i,j} \int d\mathbf{r} d\mathbf{r}' V_C \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r}') \psi_j^*(\mathbf{r}') \psi_j(\mathbf{r}) \delta_{\mathbf{r} \mathbf{r}'} \delta_{i j}. \] (2.38)

It is this exchange component of the Coulomb interaction that arises from switching or exchanging two particles \((i \leftrightarrow j)\), which favors the polarization of the intrinsic degrees of freedom. Occupying all of one spin type ensures that the spatial portion of the many-body wavefunction is anti-symmetric. Thus, compared to electrons with different spins, electrons with the same spin state have minimal wavefunction overlap and experience less Coulomb repulsion. If the corresponding exchange energy gain is larger than the cost in kinetic energy from occupying only one spin type, then all the spins spontaneously polarize. In analogy to Hund’s rule, an energy gap develops between the two spin types (Fig. 1.4).

For systems in the quantum Hall regime, the kinetic energy is quenched within a Landau level, so it does not increase for a spin-polarized state, thereby allowing Coulomb interactions to dominate. The magnetic length \(\ell_B = \sqrt{\hbar/eB}\) is the relevant length-scale that determines the interaction energy-scale, given by \(E_C = e^2/4\pi\epsilon\ell_B\). Therefore, electron-electron interactions scale as \(\sqrt{B}\) and become increasingly important as the magnetic field increases.

Such interaction-driven spontaneous polarization of the electron spin (or pseudospin) degree of freedom is known as quantum Hall ferromagnetism. The polarization direction of the broken-symmetry state may be chosen by single particle effects, but exchange interactions can still significantly enhance the magnitude of the energy gap. We emphasize that although the transport properties of the ferromagnetic and integer quantum Hall states are similar, their nature is completely different:
ferromagnetic quantum Hall states are due to the electron-electron interactions, while integer QH states would occur even in a non-interacting 2DEG.
Chapter 3

Scanning Tunneling Microscopy

The conventional microscope expanded the boundaries of what was considered observable by providing scientists with a tool to image objects at increasingly smaller length scales. Ultimately, the resolution of such microscopes is inherently limited by the wavelength of light used to illuminate the object of interest. For a microscope using visible light, the resolution is about 400 nm. At the end of the spectrum, x-rays, with the shortest wavelengths of 0.01 nm to 10 nm, have a resolution on the order of a few angstroms, making them commonly used in the study of the atomic structure of crystals. The technique of x-ray diffraction makes use of the Bragg patterns that arise due the periodic nature of the atoms to gain information about the crystal structure in momentum space. This data can be Fourier transformed to back-calculate the real space lattice structure of the sample.

Imaging with a scanning tunneling microscope (STM) is based on a different set of principles compared to conventional microscopy, utilizing quantum mechanical tunneling to probe atomic length scales directly in real space. Binning and Rohrer developed the first STM in 1982, for which they were awarded the 1986 Nobel Prize in Physics [46]. Over the past thirty years, significant developments in STM instrumentation have led to advances in nanoscale research with increased...
stability, lower temperatures, and the application of magnetic fields. From the initial STM measurements of a reconstructed Si(111) surface [47] to recent studies of unconventional superconductivity [48–51] and topological states of matter [19, 52–54], STM has proven to be a powerful tool to locally study both the structural properties and the electronic behavior in a wide variety of samples.

The heart of an STM consists of a tip-vacuum-sample junction, in which a sharp metallic tip measures tunneling current as it is scanned over an atomically flat conducting sample, as shown schematically in Fig. 3.1a. Bringing the tip very close to the sample, (to within a few Angstroms), but not in electrical contact, allows the wavefunctions of electrons in the tip and the sample to overlap. The application of a bias voltage between the tip and sample establishes a tunneling current across the junction, where charges tunnel through the vacuum barrier, a phenomenon that would be classically forbidden. This tunneling current is exponentially sensitive to the tip-sample distance, so the sample is essentially being probed by the last atom at the end of the tip, which allows for the atomic spatial resolution of STM measurements.

3.1 Theory of Tunneling

In STM, the electronic and structural properties of the sample surface are probed by measuring tunneling current $I$ as a function of applied bias voltage $V$ and lateral tip position. Vacuum, being the simplest of tunnel barriers, allows for the measurements to pertain directly to properties of the tip and the sample.

The total tunneling current can be calculated using Fermi's golden rule of time-dependent perturbation theory. A positive bias voltage $+V$ applied to the sample, effectively lowers the Fermi level of the sample by $eV$ with respect to the Fermi level of the tip, as depicted in Fig. 3.1b. Consequently, positive bias voltages
Figure 3.1: **STM Schematic.** (a) Schematic of an STM junction. (b) Density of states (DOS) of a metallic tip and an arbitrary sample. For a bias voltage $+V$ applied to the sample, electron tunneling is expected from the tip to the sample through the vacuum barrier of the junction.

probe empty states of the sample while filled states are probed at negative bias voltages. We note that the application of a bias does not fill or empty additional levels within the sample density of states, but rigidly shifts the energy of the originally occupied states with respect to the tip. The net tunneling current can be expressed as the difference between electron tunneling from tip to sample and the reverse current from sample to tip [55]:

$$I(V) = 2(-e) \frac{2\pi}{h} \int_{-\infty}^{\infty} d\epsilon \ |M|^2 \left( \rho_s(\epsilon + eV) \left[ 1 - f_s(\epsilon + eV) \right] \rho_t(\epsilon) f_t(\epsilon) \right)$$

$$- \left. \rho_s(\epsilon + eV) f_s(\epsilon + eV) \rho_t(\epsilon) \left[ 1 - f_t(\epsilon) \right] \right|_{\text{sample to tip}}$$

$$= -\frac{4\pi e}{h} \int_{-\infty}^{\infty} d\epsilon \ |M|^2 \rho_s(\epsilon + eV) \rho_t(\epsilon) \left[ f_t(\epsilon) - f_s(\epsilon + eV) \right].$$

(3.1)

Here, $\rho_i(\epsilon)$ is the density of states (DOS), $V$ is the bias voltage applied to the sample, $f_i(\epsilon) = 1/(1 + e^{\epsilon/k_B T_i})$ is the Fermi-Dirac distribution accounting for the thermal population of particles at temperature $T_i$, and the subscripts $i = s, t$ correspond to
sample and tip respectively. The tunneling matrix element $|M|^2$ captures the overlap between tip and sample wavefunctions across the vacuum barrier, which we discuss in further detail below. The factor of 2 accounts for spin, which is treated as two parallel channels of conductance and $(-e)$ corresponds to the charge of the single electrons that are tunneling.

The unprecedented atomic-scale resolution of STM arises in part from the sensitivity of the tunneling current to the distance between tip and sample. The tunneling matrix element captures the overlap between the wavefunctions of the tip and sample, and is given by

$$M = \langle \psi_t | \mathcal{H} | \psi_s \rangle,$$  \hspace{1cm} (3.2)

where $\mathcal{H} = -\frac{\hbar^2}{2m} \nabla^2$ in the free electron approximation. We now consider a simplified picture of the effect of the vacuum barrier on the tip and sample wavefunctions. These wavefunctions decay exponentially across the barrier

$$\psi_i(z) = \psi_i(0)e^{-\kappa z},$$  \hspace{1cm} (3.3)

where $z$ is the distance into the barrier (normal to the sample surface). The characteristic decay length is set by

$$\kappa = \sqrt{\frac{2m\phi}{\hbar}},$$  \hspace{1cm} (3.4)

where the work function of the material, $\phi$, is the minimum energy required to remove an electron from the material. The exponential decay of both the tip and sample wavefunctions into the barrier means that small changes in tip height above the sample surface leads to substantial changes in tunneling current. For metals, the work function is typically on the order of 5 eV. Thus, in realistic STM junctions, a
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1 Å displacement of the tip results in an order of magnitude change in the tunneling current.

We can further simplify the tunneling current (Eq. 3.1) by making the following approximations. We assume the metallic tip has a density of states that is constant in the energy range of interest and that the tunneling matrix element, $|M|^2$, is independent of energy. Both these terms can then be taken out of the integral to give

$$I = -\frac{4\pi e}{\hbar}|M|^2 \rho_t(0) \int_{-\infty}^{+\infty} d\epsilon \rho_s(\epsilon + eV)[f_s(\epsilon + eV) - f_t(\epsilon)]. \quad (3.5)$$

The differential conductance can then be written as

$$\frac{dI}{dV} = \frac{4\pi e^2}{\hbar}|M|^2 \rho_t(0) \int_{-\infty}^{+\infty} d\epsilon \rho_s(\epsilon) f'(\epsilon - eV), \quad (3.6)$$

where we have assumed that the tip and sample are at the same temperature. At low temperatures, the derivative of the Fermi function can be treated as a delta function, $f'(x) \propto \delta(x)$, simplifying the differential conductance to

$$\frac{dI}{dV}(V) \propto \rho_s(eV). \quad (3.7)$$

This is the foundational principle of STM spectroscopic measurements: the differential conductance $dI/dV$ is proportional to the local density of states (LDOS) of the sample. It is this principle that allows for the versatility of STM measurements, which go beyond simply performing atomic resolution imaging of surfaces and paves the way for studying electronic behavior locally in a wide variety of materials.
3.2 Basic Measurements with STM

Typically, STM measurements fall into two categories: topographic measurements that image the material surface and spectroscopic measurements that probe the electronic properties of the sample. We briefly discuss these different modes of operation below.

3.2.1 Topographic Mapping

Topographic measurements contain information about the structural properties of the sample surface, right down to the atomic scale. The tip is initially fixed at a height determined by the setpoint current and bias voltage. A feedback loop is used to maintain a constant tunneling current by adjusting the extension of a piezoelectric tube scanner \((z\text{-piezo})\) which holds the tip. The topographic profile of the surface can be mapped out by recording the height of the \(z\text{-piezo}\), relative to the initial setpoint, as the tip is scanned over a desired area of the sample in \((x, y)\).

An example of topographic images obtained on a cleaved Bi(111) surface is shown in Fig. 3.2. The large-scale topography reveals terraces with minimal disorder that are terminated by step edges along crystallographic directions (Fig. 3.2a). These step edges have a 4 Å height, as expected for the (111) surface of bismuth (Fig. 3.2b). Imaging atomic-resolution topographic maps require higher sensitivity than the large-scale ones. As such, the setpoint bias and current are adjusted to bring the tip closer to the sample so that individual atoms can be resolved, as shown in Fig. 3.2c. These topographic images and the symmetries of the lattice are widely used to characterize crystal orientations and cleavage planes, as well as atomic-scale disorder and surface reconstructions.
3.2.2 Spectroscopic Measurements

Spectroscopic measurements reveal information related to the electronic properties of a material, because the differential conductance $dI/dV$ is directly proportional to the LDOS (Eq. 3.7).

To take a point spectrum, the lateral position of the tip in $(x, y)$ is fixed. The tip is stabilized at a setpoint current and bias voltage, which determine its initial height. Then the feedback loop, normally used to maintain a constant current, is disabled and the bias voltage is swept as the current $I$ or differential conductance $dI/dV$ are recorded. To measure $dI/dV$, a small ac modulation $dV$ is added to a fixed bias voltage $V$, resulting in a current modulation $dI$ at the same frequency that is detected with a lock-in amplifier. This standard lock-in technique of directly measuring $dI/dV$ provides a better signal-to-noise ratio than taking a numerical derivative of the $I-V$ data.
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We now present a few examples of point spectra taken on different samples in a dilution refrigerator STM operating at millikelvin temperatures and at high magnetic fields (details of the instrument itself are discussed in Sec. 3.3). Figure 3.3a shows a spectrum taken on the surface of a superconducting lead (Pb) single crystal. The superconductivity manifests in tunneling measurements as an absence of electronic states around the Fermi level ($E = 0$) bounded by sharp coherence peaks at the gap edge. The two features in the coherence peaks occur due to the two-gap nature of the superconductivity in Pb(110) [56–58]. This behavior can be attributed to different electron-phonon coupling between the two Fermi surfaces in Pb [59]. Previous STM measurements resorted to using superconducting tips in order to resolve the two-gap superconductivity in Pb [58]. Our ability to detect this these two sharp coherence peaks with a normal metal tip is a testament to the high energy resolution afforded by the low temperature of our system [60].

Additionally, the capability of applying large magnetic fields in conjunction with the low temperatures can be used to gain insight into other regimes. A spectrum of the Bi(111) surface state taken at a magnetic field $B = 14$ T, shown in Fig. 3.3b, reveals Landau levels as sharp peaks in $dI/dV$ that occur at certain discrete energies. Landau level spectroscopy with STM has been previously applied to extract precise band structure information [13, 17, 19, 61, 62]. In this thesis, we further explore novel symmetry breaking effects that emerge at these high fields and can be resolved at millikelvin temperatures due to the 100 µeV energy resolution of our instrument. These examples briefly illustrate the power of STM spectroscopy to study a variety of physical phenomena under different measurement conditions.

Lastly, we discuss spectroscopic imaging, which utilizes the unique ability of STM to correlate measurements of the local density of states with simultaneously acquired topographies at the atomic scale. Here, the differential conductance is measured on
Figure 3.3: **Point spectroscopy.** (a) Superconducting spectrum on a Pb(110) surface. We resolve a hard gap at the Fermi level \( E = 0 \) and substructure in the coherence peaks arising from two gaps. The data (blue) is fit (red) to the sum of two superconducting DOS [63] resulting in the parameters: \( \Delta_1 = 1.26 \text{ meV}, \Delta_2 = 1.42 \text{ meV}, \) the Dynes factor \( \Gamma = 8 \mu\text{eV} \) and \( T_e = 250 \text{ mK}. \) Measurement parameters: \( V_{\text{set}} = -5 \text{ mV}, I_{\text{set}} = 1 \text{ nA} \) and \( V_{\text{rms}} = 30 \mu\text{V}. \) (b) Landau level spectroscopy of the Bi(111) surface in a large magnetic field, at \( B = 14 \text{ T}, \) showing sharp peaks in the DOS corresponding to Landau levels. \( T_e = 250 \text{ mK}. \) Measurement parameters: \( V_{\text{set}} = -400 \text{ mV}, I_{\text{set}} = 2 \text{ nA} \) and \( V_{\text{rms}} = 500 \mu\text{V}. \)

a grid of \((x, y)\) points, resulting in 1D linecuts or 2D maps at several energies, which capture spatial variations in the LDOS over a certain area of the sample. Often, STM spectroscopic imaging is dedicated to analyzing quasiparticle interference (QPI), the scattering patterns that arise in the vicinity of point defects or step edges. The Fourier transform of real space QPI datasets can be used to extract information about the band structure in momentum-space and can be readily compared to ARPES measurements. We do not discuss the specifics of QPI here, as it is not pertinent to the measurements reported in this thesis. In our experiments, we employ 1D spectroscopic linecuts to track spatial variations in Landau level energies and use 2D conductance maps to directly image Landau level wavefunctions in real space. These ideas are presented in their entirety in subsequent chapters.
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3.3 The Millikelvin, High Field STM System

Scanning tunneling microscopy is one of the most technically challenging techniques used to characterize properties of materials, where the quality of the data is heavily dependent on the instrumentation. All experiments presented in this thesis were conducted in a dilution refrigerator STM (DRSTM). This instrument is a low-noise system operating at cryogenic temperatures and in high magnetic fields. It is a home-built, state of the art instrument, designed and constructed by Shashank Misra, Brian Zhou and Ali Yazdani as detailed in Refs. [64, 65] (see also [66]). The experimental findings presented in this thesis are truly a consequence of the physical regime of millikelvin temperatures and 14 T magnetic field made accessible to STM by the development of such an instrument.

The DRSTM system is integrated into an ultra-high vacuum (UHV) environment. The dilution refrigerator insert of a bottom loading cryostat has a UHV space that is connected to a set of UHV chambers. The UHV keeps the sample surfaces clean on the atomic scale, allowing for the study of cleaved samples as well as in-situ preparation of single crystals. We have recently added a bismuth evaporation chamber to the system, to epitaxially grow thin films of bismuth (similar to the ante-chamber discussed in [66]).

The insert of the cryostat is mounted onto a manipulator, which allows for vertical motion of the microscope between measurement position and sample-exchange position without breaking vacuum of the inner vacuum can (IVC) of the insert. For sample or tip exchange, the insert is moved down such that the microscope is in the middle of the transfer chamber, allowing for direct line of sight to access the sample or tip with a wobble stick. During measurements, the sample is centered in the bore of a superconducting magnet. The magnet is capable of applying a field of up to 14
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T and experiments are conducted with the magnet in persistent mode to minimize noise.

Since the tunneling current is exponentially sensitive to tip-sample distance, it is imperative that vibrations are minimized in the STM. The need for strong thermal coupling to the dilution refrigerator precludes the use of springs at the microscope, a common technique for STMs operating at higher temperatures. Instead, for this DRSTM system, two stages of pneumatic vibration isolation are introduced, along with acoustic shielding and careful routing of pumping lines.

In order to increase functionality of the instrument, the microscope is designed to hold two samples. This enables reliable motion between a sample of interest and a sample used for tip preparation (in our case, typically a Cu(100) single crystal used for field emission) without stopping the running of the dilution refrigerator. The STM microscope head is attached to the mixing chamber of the dilution unit. With the fridge running, the microscope is cooled down to a phonon temperature of $T_{\text{ph}} = 20 \, \text{mK}$. The corresponding electron temperature of the sample is $T_e = 250 \, \text{mK}$, as measured by fitting the BCS superconducting gap of an aluminum single crystal [64, 65].

One major limitation of this system is that the dilution unit cannot be run continuously. Instead, it is necessary to run the 1K pot in single-shot mode to achieve the low noise conditions necessary for STM. The 1K pot must be filled with helium and then sealed from further helium intake with the use of a needle valve. The DRSTM system was designed with this possibility in mind as the 1K pot has a one liter volume, unlike typical dilution refrigerator designs. With the dilution unit running, the 1K has an 8 hour hold time; these refill cycles constrain the measurement time considerably.
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Nevertheless, since coming online, DRSTM has proved singularly valuable in studying an array of materials and has produced a number of beautiful results. Workarounds for this time restriction include the use of the multipass technique for measuring conductance maps at a few energies, which was developed for its time efficiency and improved noise performance (see Ref. [65] for details).

Upgrades to the current system are in their early stages. This includes a redesign of the insert, which incorporates a thermally anchored fixed impedance capillary that allows for the capability to run continuous measurements. Moreover, improvement on the electron temperature may be achieved by refining the filtering and thermal anchoring of the wiring as well as including additional radiation shielding.

Energy Resolution

There are primarily two contributing factors to the energy resolution of our differential conductance spectra, the temperature and the lock-in measurement technique. For a density of states that is temperature independent, the thermal broadening of \( dI/dV \) only enters in the derivative of the Fermi function, which can no longer be treated as a simple \( \delta \)-function. Instead, this term in Eq. 3.6 gives rise to a broadening of sharp spectroscopic features with a full width at half maximum (FWHM) given by \( \approx 3.5k_B T \).

Furthermore, at millikelvin temperatures, a second resolution-limiting source becomes relevant. The standard lockin techniques employed for measurement add an additional energy broadening, which arises due to the applied ac voltage modulation. This results in a FWHM = \( 1.7v_{pk-pk} = 2.5v_{rms} \), where \( v_{rms} \) is the amplitude of the ac modulation conventionally used in lock-in measurements [66, 67].
Thus, the total resolution of a $dI/dV$ measurement is the result of adding the thermal and bias modulation broadening in quadrature:

$$\Delta E_{\text{FWHM}} = \sqrt{(3.5k_B T)^2 + (2.5 ev_{\text{rms}})^2}. \quad (3.8)$$

For parameters representative of our experiments, with $T = 250$ mK and $v_{\text{rms}} = 30 \mu$eV, the total energy resolution corresponds to $\Delta E_{\text{FWHM}} \approx 110 \mu$eV.
Chapter 4

Visualizing Nematic Quantum Hall Wavefunctions on the Surface of Bismuth

In this chapter, we investigate the quantum Hall states that arise in high magnetic fields from the anisotropic valleys of the Bi(111) surface state. Spectroscopy performed with a scanning tunneling microscope shows that a combination of single-particle effects and many-body Coulomb interactions lift the six-fold Landau level (LL) degeneracy to form three valley-polarized quantum Hall states. We directly image the underlying wavefunctions and find the resulting Landau orbits are anisotropic. These wavefunctions have a different orientation for each broken-symmetry state, corresponding to states arising from pairs of occupied elliptical valleys. We thereby ascertain the local valley ordering and provide a direct spatial signature of a nematic electronic phase.
CHAPTER 4. VISUALIZING NEMATIC QUANTUM HALL WAVEFUNCTIONS ON THE SURFACE OF BISMUTH

4.1 Introduction

4.1.1 Nematic Fermi Fluids

Nematic electronic states are characterized by a reduced rotational symmetry relative to the underlying crystal lattice. They represent an intriguing class of broken-symmetry phases that can spontaneously form as a result of electronic correlations [68, 69]. Quantum nematic phases are analogous to classical liquid crystals, but instead of elongated molecules aligning, it is the electronic wavefunctions that collectively have a preferred directionality. These nematic states were proposed by Kivelson, Fradkin and Emery [69], inspired by the observation of unusually large transport anisotropies in otherwise isotropic electronic systems. Such anisotropic behavior was seen in two-dimensional electron gases (2DEGs) at high magnetic fields [5, 70, 71], in strontium ruthenate materials [72, 73], and in several high-temperature superconductors [74–79].

The mechanisms that give rise to nematicity in quantum fluids are different from the classical case, and address how point-like electrons in a crystalline lattice can break the spatial rotational symmetry. From a strong coupling picture, a nematic fluid can be treated as a partially melted solid, which preserves translational symmetry but breaks the rotational symmetry of the lattice (Fig. 4.1, left). An alternative perspective is to consider a nematic transition as a Pomeranchuk instability, which occurs when a Fermi liquid becomes thermodynamically unstable and undergoes a quantum phase transition into a state with lower symmetry (Fig. 4.1, right). The key factor for nematicity in strongly correlated materials is that the driving force for the symmetry breaking arises from electronic interactions, and the resulting anisotropic behavior is more pronounced in the electronic structure than in lattice distortions.
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Figure 4.1: **Possible mechanisms of nematicity.** Left: The melting of a stripe phase, which can restore long-range translational symmetry while preserving orientational order. Right: A Pomeranchuk instability where a Fermi liquid with a circular Fermi surface undergoes a quantum phase transition to a nematic Fermi fluid in which the Fermi surface has a quadrupolar distortion. Reproduced from Ref. [68].

Some of the first experimental observations of nematic behavior were measured in GaAs/AlGaAs heterostructures in high magnetic fields [5, 71, 80]. Strong anisotropies were seen in the longitudinal resistivity along orthogonal crystallographic axes, which emerged for the metallic state near filling factor $\nu = 9/2$ and several higher, half-filled Landau levels (Fig. 4.2a). This striking anisotropy develops due to a unidirectional or striped charge density wave, e.g. at $\nu = 9/2$ the system separates into equal width stripes of $\nu = 4$ and $\nu = 5$. The conductivity is much larger along the stripes than perpendicular to them [70]. A similar magnetotransport anisotropy is observed in strontium ruthenate Sr$_3$Ru$_2$O$_7$, but stems from a substantially different origin [72]. The experiment consisted of applying an external magnetic field and measuring the in-plane resistivity (Fig. 4.2b). For a field perpendicular to the sample surface, the resistivity retains the four-fold symmetry of the crystal. However, if the sample is tilted slightly to provide an in-plane field component, the resistivity becomes strongly anisotropic, with a reduced two-fold symmetry. The role of the in-plane field is to
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Figure 4.2: Examples of nematicity measured by transport. (a) Longitudinal resistivity in a high mobility GaAs 2DEG at $T = 25$ mK in a perpendicular magnetic field. Dashed gray line: current flow along $<1\overline{1}0>$ direction. Solid line: current flow along $<110>$ direction. Striking transport anisotropy is observed at Landau level filling factors $\nu = 9/2, 11/2, 13/2$ and higher. Reproduced from Ref. [80]. (b) Magnetoresistivity of Sr$_3$Ru$_2$O$_7$. For an applied magnetic field parallel to the crystallographic $c$ axis, the in-plane resistivity along orthogonal directions $\rho_{aa}$ (black) and $\rho_{bb}$ (red) are almost identical. When the field is tilted such that it has an in-plane component along the $a$-axis, a strong anisotropy is seen, with the easy direction for current flow along the $b$-axis, perpendicular to the in-plane field component. Reproduced from Ref. [72].

align nematic domains in Sr$_3$Ru$_2$O$_7$, so the nematicity is macroscopically observable. Finally, nematic order has been detected in high temperature superconductors. In YBa$_2$Cu$_3$O$_{6+x}$, the orthorhombic crystal structure provides a symmetry breaking field [68], while macroscopic anistoropies have been observed in transport [74, 76], neutron scattering [75] and Nernst measurements [81]. In other high-temperature superconductors, STM studies have revealed nematic correlations. For example, Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ exhibits unidirectional ordering with a four lattice constant period [79, 82, 83] and iron-pnictides display electronic anisotropies that break the underlying rotational symmetry [77, 78, 84]. Nematicity in strongly correlated materials is of particular interest as this phase can be intertwined with other types of order including magnetism or superconductivity and possibly linked to quantum criticality [68, 84].
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The sensitivity of electronic nematic phases to disorder results in short range ordering and domains, making them difficult to study using global measurements. In techniques that average over microscopic configurations, an external polarizing field such as crystalline strain or in-plane magnetic field may be needed in order to show a propensity for nematic order [68]. However, such demonstrations typically do not probe the influence of electron-electron interactions directly and it can be difficult to quantitatively correlate the experimental evidence of ordering with a microscopic theoretical model. To put the study of nematic electronic phases on more quantitative ground, it is therefore important not only to perform local measurements, but also to find a material system for which theory can fully characterize how electronic interactions establish the underlying broken-symmetry states.

4.1.2 Nematicity in Quantum Hall Valley Systems

Multivalley 2DEGs with anisotropic band structure have been anticipated as a model platform to explore nematic order in the quantum Hall regime [10, 31, 33, 85]. The central idea is that Coulomb interactions can spontaneously lift the valley degeneracy, producing states in which only a subset of the valleys are occupied. Due to the valley anisotropy, the resultant electronic states form a nematic phase, which breaks the rotational symmetry of the underlying crystal. In contrast to the nematic metal phase that emerges near half-filled LLs as discussed for GaAs heterostructures, this valley nematic order is gapped in the bulk and has a quantized Hall conductance. Moreover, the valley anisotropy is established by the electronic band structure of the material and does not arise from a Pomeranchuk instability, which makes this spatial symmetry discrete from the outset. In these quantum Hall valley nematics, it is possible to theoretically model the underlying broken-symmetry states and to
provide a microscopic description of the interactions responsible for nematic behavior. Here we investigate such a 2DEG on the surface of single crystals of bismuth.

4.2 Bismuth and its (111) Surface

4.2.1 Brief Historical Perspective

Bismuth, a long-studied material in condensed matter physics, has played a pioneering role in the experimental observations of quantum behavior [86, 87]. From a historical point of view, it is quite remarkable that many effects were first discovered in bismuth crystals - large magnetoresistance by Kapitza [88], followed soon after by the first observations of Shubnikov-de Haas and de Haas-van Alphen oscillations [87]. Bismuth has been the precursor to many of these effects being observed in metals with larger carrier densities. This is because bulk bismuth is a semimetal, where the conduction and valence bands overlap to produce a compensated system as shown in Fig. 4.3. It has a very small Fermi surface (occupying $10^{-5}$ of the Brillouin zone) and thus has very long mean free paths, up to $\ell_{\text{mfp}} \sim 2 \mu$m at room temperature [89], and reaching the ballistic regime with $\ell_{\text{mfp}} \sim 1$ mm in clean crystals at low temperature [40].

Interest in bismuth has been recently rekindled by bulk measurements showing phase transitions and anisotropic behavior, possibly related to nematic electronic phenomena, in the presence of large magnetic fields [12, 90, 91]. Moreover, it been shown to host one-dimensional topological edge modes [92] and been cited as an example of a higher order topological insulator [93]. Finally, quite unexpectedly, this material with its very low density of charge carriers, of only 1 electron per 100,000 atoms, has been shown to superconduct with a $T_c = 0.5$ mK [94]. Understanding many of these behaviors in bismuth is still an ongoing endeavor.
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Figure 4.3: **Semimetal band structure of bulk bismuth.** (a) Band structure of a semimetal compared with that of other crystalline solids including metals with large electron densities and insulators with large energy gaps. (b) Semimetallic band structure of bulk bismuth. Here $\varepsilon_{Fe}$, $\varepsilon_{Fh}$, and $\varepsilon_{Fl}$ are the Fermi energies of electrons, heavy holes and light holes, while $\varepsilon_g$ is the direct energy gap. Reproduced from Ref. [86].

4.2.2 Crystal Structure

Bulk bismuth crystallizes in a rhombohedral structure with two atoms per unit cell [40, 95], as illustrated in Fig. 4.4. A bulk Bi crystal along the [111] direction can be regarded as a stack of Bi bilayers, with strong covalent bonds within each bilayer and a weaker van der Waals bonding between bilayers. The (111) surface is a natural cleavage plane of Bi crystals, and also turns out to be the preferred direction of epitaxial growth, although there are exceptions for very thin films. The Bi(111)
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Figure 4.4: Bi(111) crystal structure. (a) Rhombohedral unit cell and hexagonal crystal structure of bulk bismuth. Adapted from Ref. [95]. (b) Top: Top view of honeycomb lattice of the Bi(111) surface. Red and blue atoms belong to different triangular atomic sublattices that are offset in height. Bottom: Side view of Bi crystal, which can be regarded as individual Bi bilayers stacked along the [111] crystallographic direction. Adapted from Ref. [92].

surface has a buckled honeycomb lattice with $C_3v$ symmetry, as illustrated in Fig. 4.4b, with two triangular sublattices that are offset in the $z$ direction.

We cleave a bulk bismuth crystal in ultra-high vacuum, exposing a pristine (111) surface. STM topographic images show the surface has large (>200 nm × 200 nm), ordered terraces that are disorder-free with only a few atomic scale defects as shown in Fig. 4.5a. These terraces have the expected 4 Å height for Bi bilayers and are separated by steps oriented along high-symmetry crystallographic directions. An atomic resolution topography reveals a hexagonal pattern of atoms, corresponding to the upper atomic sublattice of the Bi(111) surface (Fig. 4.5b).

4.2.3 Electronic Band Structure

The electronic structure of Bi surfaces undergo substantial changes due to large spin-orbit coupling in the material, making the surface states much better metals
than the bulk [40]. Thus, the Bi(111) surface state can be treated as a nearly two-dimensional metal, forming a model system in which we examine valley nematic behavior. The Rashba spin-orbit coupling lifts the spin degree of freedom to produce spin-split surface state bands. The Fermi surface (at $E = 0$) contains multiple electron and hole valleys corresponding to the minima or maxima (depending on the sign of the effective mass) in the band structure, respectively (Fig. 4.6). The electronic structure has been investigated theoretically using tight-binding [96, 97] and first-principles [42] calculations, as well as experimentally using angle-resolved photoemission spectroscopy (ARPES) measurements [41–43, 98]. Its Fermi surface consists of six quasi-elliptical hole pockets along the Γ-M directions, a central, hexagonal electron pocket at the Γ point, and three additional elongated electron pockets.

Figure 4.5: Topography of Bi(111) surface. (a) A typical cleaved Bi(111) surface, showing large terraces and low disorder, with crystallographic axes labeled. Surface defects are circled in purple, and the inset shows a zoom-in on one defect (inset $z$ height scale: 1.3 Å). The data in Fig. 4.8, Fig. 4.9a and Fig. 4.10 are an average of spectra measured along the blue line. Conductance maps in Fig. 4.12, Fig. 4.17 and Fig. 4.19 are taken in the area marked by the black box. (b) Atomic resolution image of the surface Bi atomic lattice and an isolated surface defect.
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Figure 4.6: **Band structure of Bi(111) surface states.** (a) Bi(111) surface state band structure along high-symmetry directions. Rashba spin-orbit coupling gives rise to spin-split bands (blue and red lines) that exhibit multiple Fermi level crossings. The yellow shaded region represents bulk bismuth bands projected onto this surface. Figure adapted from Ref. [42]. (b) Bi(111) Fermi surface showing multiple electron (red) and hole (blue) valleys of the surface state.

pockets around the M points (Fig. 4.6b). The multiply degenerate anisotropic valleys and the low disorder of the Bi(111) surface make it an ideal system to search for nematic electronic behavior in the quantum Hall regime using STM.

### 4.3 Landau Level Spectroscopy

To study the electronic behavior and valley symmetry breaking on the Bi(111) surface, we perform spectroscopic measurements in a dilution refrigerator STM [64] at high magnetic fields. In the absence of a magnetic field, the tunneling conductance $dI/dV$ exhibits features related to van Hove singularities in the density of states, such as the peak at $E = 220$ meV and the abrupt drop at $E = 33$ meV (Fig. 4.7a) that
correspond to the two upper band edges of the surface state along the Γ-M direction \[92, 99\]. In contrast to this generally smooth curve, the point spectra in the presence of a large magnetic field show a series of sharp peaks (Fig. 4.7b), due to the quantized Landau levels (LLs). The evolution of these states as a function of magnetic field \(B\) can be used to distinguish between electron-like and hole-like LLs, which disperse in energy with positive or negative slopes, respectively (Fig. 4.8a). When these energy levels cross, they do not exhibit avoided crossings, and instead the total conductance is additive, which suggests independent tunneling into each LL. We focus primarily on the hole-like states for which each LL has a six-fold valley degeneracy (note: there is no additional spin degeneracy as this degree of freedom has been lifted by the spin-orbit coupling). It is straightforward to assign the orbital index \(N\) for these hole LLs, where the highest-energy state with negative dispersion corresponds to \(N = 0\). Furthermore, the energy of this \(N = 0\) LL is closely matched to the hole band extremum, i.e. the zero-field drop in conductance at \(E = 33\) meV.

We first notice that the surface state LLs do not disperse linearly with magnetic field. Instead, they are pinned to the Fermi level until they are fully occupied, as is seen for the hole states in Fig. 4.8b. This behavior indicates that the surface charge density is held constant in our system. Using the values of the field and filling factor at which LLs cross the Fermi level, we determine the hole surface density to be \(p \approx 7.1 \times 10^{12} \text{ cm}^{-2}\) (see Sec. 4.7.3). Although such pinning is rarely observed in LL spectroscopy of ungated samples performed using a STM \[100\], here it plays a crucial role in allowing us to controllably tune the LL filling factor by changing the magnetic field.
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Figure 4.7: Spectra of bismuth surface state. (a) Tunneling conductance $dI/dV$ as a function of energy $E$ in zero magnetic field. The drop in conductance at $E \approx 30$ meV and peak at $E = 220$ meV match the energies of the band maxima in the $\Gamma$-$M$ direction. (b) Tunneling spectra in a large magnetic field, at $B = 14$ T, showing sharp peaks in the DOS corresponding to Landau levels (LLs).

Figure 4.8: Landau fan diagrams. (a) Landau fan diagram of $dI/dV(E, B)$ that shows crossing of electron- and hole-like LLs. The data are averaged over the blue line shown in Fig. 4.5a, with individual spectra showing almost no spatial variation on this energy scale. Select orbital indices $N$ and $N_e$ of the respective hole and electron LLs are labeled. (b) Higher energy resolution measurement of $dI/dV(E, B)$ that shows pinning of each hole LL to the Fermi level ($E = 0$).
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4.4 Broken-Symmetry Nematic Order

Spectroscopy of Broken-Symmetry States

High-resolution spectroscopy around the Fermi level reveal that both single-particle effects and electron-electron interactions break the six-fold valley degeneracy of the hole LLs. Evidence of symmetry breaking can be seen in Fig. 4.9a, which shows a zoom-in on the field evolution of conductance spectra where the \( N = 3, 4, 5 \) LLs are each split into two peaks with different amplitudes - indicating a lifting of the six-fold valley degeneracy of each LL into a two- and four-fold degenerate state. The fact that the splitting occurs away from the Fermi level and is independent of orbital index indicates that it is a single-particle effect. We find this splitting depends on position; the linecut in Fig. 4.9b illustrates a lifting of the six-fold degeneracy of the \( N = 3 \) LL to produce either two or three broken-symmetry states, depending on location within the sample. The spatial dependence of this partial symmetry breaking suggests the source is variations of local strain in the sample. We characterize the resulting gap by \( \Delta_{\text{strain}} \). Strain acts as a valley-Zeeman field that couples to the valley degree of freedom, thereby lifting the degeneracy.

Electron-electron interactions can further lift the LL degeneracy by spontaneously opening energy gaps at the Fermi level when the LLs are partially occupied. For instance, Fig. 4.10 shows a high resolution measurement over an energy range of \( \pm 3 \) meV where an additional gap opens and closes within the four-fold degenerate multiplet of the \( N = 4 \) LL as it is pinned to \( E = 0 \). The magnitude of the exchange gap \( \Delta_{\text{exch}} \approx 450 \mu\text{eV} \) at \( B = 10.9 \) T, is consistent with that estimated theoretically for the hole pockets of Bi(111) (see Sec. 4.7.4), although there are small spatial variations in the exact magnitude of the exchange gaps. Interactions consistently induce gaps
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Figure 4.9: **Role of strain in symmetry-breaking.** (a) Landau fan diagram showing the $N = 3, 4, 5$ LLs split into two peaks corresponding to a two-fold and a four-fold degenerate LL in this region of the sample. This is a single particle splitting that is independent of orbital index or magnetic field, which we attribute to strain in the sample. Data are averaged over the blue line in Fig. 4.5a. (b) Linecut of spectra showing strain-induced splitting of the six-fold degenerate $N = 3$ LL into two or three peaks, depending on position. Numbers in parentheses denote degeneracy of each broken-symmetry state.

Figure 4.10: **Exchange-induced symmetry breaking.** (a) Zoom-in on LL fan diagram around the Fermi level. The four-fold degenerate peak further splits into two distinct LLs as it crosses $E = 0$, indicating broken symmetry states arising from exchange interactions. Splittings from strain and exchange are labeled. (b)-(c) Individual spectra at select magnetic field, which highlights the interaction-induced gap as the four-fold degenerate state splits into two doubly degenerate LLs around the Fermi level. Numbers in parentheses denote degeneracy of each broken-symmetry state.
between previously degenerate multiplets when they cross the Fermi level and enhance gaps between LLs already split by strain. These observations demonstrate that a combination of single particle effects and many-body interactions lift the six-fold valley degeneracy of the hole LLs, and highlights how essential the energy resolution of the dilution refrigerator STM is in order to resolve these broken-symmetry states on the energy scale of hundreds of $\mu$eV.

**Spectroscopic Mapping of Nematic Order**

Next, we perform spectroscopic mapping to examine the valley ordering of these broken-symmetry states and demonstrate nematic behavior in the electronic wavefunctions. Conductance maps at energies corresponding to each of the three split LLs, taken in the same area of the sample, are shown in Fig. 4.11a-d. These data show anisotropic elliptical features that point along high-symmetry crystallographic axes, with relative angles rotated by 120° with respect to each other. Such spatially resolved measurements enable us to directly visualize the rotational symmetry breaking of the electronic states. By tuning the magnetic field to adjust the occupancy of the multiplet split by exchange, we can contrast spatial maps of the LLs with and without the spontaneous breaking of the degeneracy by Coulomb interactions. The measurements in Fig. 4.11e-g, obtained at a different magnetic field in the same region as those in Fig. 4.11a-d, show that the elliptical features in the conductance maps occur as a superposition of two different orientations, indicating that the symmetry between these two orientations is not broken in the absence of an exchange gap. Contrasting Fig. 4.11f with Fig. 4.11b-c clearly shows that unidirectional elliptical features emerge as the exchange gap opens, providing a direct manifestation of nematic order on the Bi(111) surface.
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Figure 4.11: Mapping nematic behavior on the bismuth surface. (a) Average spectrum of the $N = 3$ LL at 12.9 T, showing three broken-symmetry LL peaks (marked by colored arrows), two of which (red and blue) are split by exchange interactions at the Fermi level. (b)-(d) Conductance maps at energies corresponding to the three split hole LLs. The ellipses have different orientations at each energy, a manifestation of nematic behavior. (e) Average spectrum at 14 T, showing restored symmetry of the exchange-split LLs in (a) to produce a four-fold degenerate LL. (f) Conductance map at the four-fold degenerate LL, which shows ellipses with two orientations. (g) Spatial map at the energy of the two-fold degenerate LL peak that is split by strain, showing the same unidirectional behavior as in (d). Measurements in (a)-(g) are taken in the same area of the sample. (h) Average conductance spectrum at 12.9 T in a location about 1 µm away from the region shown in (a)-(g). (i)-(k) Conductance maps in the new location at energies corresponding to the three split peaks. The energetic order of the three directions is different, with the first two orientations switched, demonstrating the presence of domains. For all spectra, electron LLs are labeled, and hole LL degeneracy is denoted in parentheses near each peak. Conductance is normalized by its average value $G/\bar{G}$ for spatial maps.
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Another key feature of a nematic electronic phase without long-range order is the presence of domains. We find that the sequence of the three broken-symmetry LLs can change in energy depending on the location within the sample. An example of this behavior can be seen by contrasting the spectrum and corresponding conductance maps in Fig. 4.11a-d to those measured about a micrometer away, shown in Fig. 4.11h-k. These data reveal that the orientations of the two broken-symmetry states corresponding to the first two peaks in the spectra have switched between the two locations on the Bi surface. Thus, our STM measurements not only show that electron-electron interactions drive nematic behavior, but also illustrate the formation of local nematic domains.

4.5 Imaging Quantum Hall Wavefunctions

We show below that the elliptical features in our STM conductance maps correspond to the underlying quantum Hall wavefunctions that are pinned by surface defects. To characterize these features in detail, we study them in an extremely clean area (box in Fig. 4.5a), and examine their dependence on orbital index at a constant magnetic field. The conductance maps shown in Fig. 4.12a-e are obtained for the $N = 0-4$ LLs at the energies of one particular orientation of the broken-symmetry LLs (the strain-induced state). These images reveal concentric ellipses of suppressed conductance centered around the two defects in this area. Both the size of the outermost ellipse and the number of concentric ellipses increases as orbital index increases. The data suggest these rings of suppressed conductance are individual Landau orbits. We discuss below how atomic-scale surface defects shift the energy of a single cyclotron orbit, giving rise to the decreased conductance in the shape of its wavefunction.
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Figure 4.12: Isolated anisotropic Landau orbits and theoretical modeling. (a)-(e) Conductance maps of in the area denoted by the black box in Fig. 4.5a, taken at $B = 14$ T at energies corresponding to the strain-split broken-symmetry hole LL for orbital indices $N = 0$ to 4. Isolated anisotropic Landau orbits are present around surface defects. (f) Semimajor axis size of the Landau orbits for the $N = 4$ LL as a function of magnetic field, plotted on a log-log scale. Dashed line is a fit with extracted slope $= -0.57 \pm 0.08$, which matches the expected scaling that size $\propto 1/\sqrt{B}$ for fixed orbital index. (g)-(k) Theoretically simulated spatial maps of the expected conductance, $1 - 2\pi \ell_B^2 |\psi_{N,N}(r)|^2$, with individual cyclotron orbits centered on the surface defects circled in Fig. 4.5a. The size and shape of the simulated wavefunctions are a good match to the data in (a)-(e). (l) Amplitude $2\pi \ell_B^2 |\psi_{4,4}(r)|^2$ of the $m = N = 4$ Landau orbit wavefunction.

To explicitly calculate the Landau level wavefunctions that arise from the anisotropic hole valleys of the Bi(111) surface, we assume a parabolic dispersion and approximate the anisotropic valleys as ellipses with different effective masses along the semimajor ($m_\parallel$) and semiminor ($m_\perp$) ellipse axes in momentum space. This yields a Hamiltonian for a single valley

$$\hat{H} = -\frac{\pi^2}{2m_\parallel} - \frac{\pi^2}{2m_\perp} + E_0,$$  \hspace{1cm} (4.1)

where in a perpendicular magnetic field $B$, the momentum $\hbar \mathbf{k}$ is replaced by the kinetic momentum $\mathbf{\pi} = \hbar \mathbf{k} + e\mathbf{A}$, with vector potential $\mathbf{A}$; the energy of the hole
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band edge is $E_0 \approx 33$ meV. We define the parameter $\lambda = \sqrt{m_\parallel / m_\perp}$, which captures the anisotropy of an individual valley. The coordinates can then be rescaled:

$$X = \sqrt{\lambda} x, \quad \Pi_x = \sqrt{1/\lambda} \pi_\parallel,$$

$$Y = \sqrt{1/\lambda} y, \quad \Pi_y = \sqrt{\lambda} \pi_\perp,$$ (4.2)

(4.3)

to generate an isotropic Hamiltonian

$$\mathcal{H} = -\frac{\Pi^2}{2M} + E_0,$$ (4.4)

where $M = \sqrt{m_\parallel m_\perp}$. We solve this equation in the symmetric gauge $A = \frac{1}{2}B(-y\hat{x} + x\hat{y})$, using this gauge choice based on the isotropic symmetry of the atomic scale defects on the bismuth surface. The corresponding wavefunctions are [44]

$$\psi_{N,m}(Z) = \frac{1}{\sqrt{2\pi \ell_B^2}} \left( \frac{N!}{m!} \right)^{\frac{1}{2}} \left( \frac{Z^*}{\sqrt{2}} \right)^{m-N} e^{-|Z|^2/4} \mathcal{L}_N^{m-N} \left( \frac{|Z|^2}{2} \right), \quad m \geq N \quad (4.5)$$

$$\psi_{N,m}(Z) = \frac{1}{\sqrt{2\pi \ell_B^2}} \left( \frac{m!}{N!} \right)^{\frac{1}{2}} \left( \frac{Z}{\sqrt{2}} \right)^{N-m} e^{-|Z|^2/4} \mathcal{L}_m^{N-m} \left( \frac{|Z|^2}{2} \right), \quad m < N. \quad (4.6)$$

Here $Z = (X + iY)/\ell_B$ where $\ell_B = \sqrt{\hbar/eB}$ is the magnetic length, $N = 0, 1, 2, ...$ is the orbital index, $m = 0, 1, 2, ...$ is the quantum number denoting the guiding center index, which labels the degeneracy within a given orbital index of the Landau level and $\mathcal{L}_N$ is the Laguerre polynomial. Figure 4.13 shows plots of the wavefunctions $|\psi_{N,m}|^2$ in the symmetric gauge for several $N$ and $m$ values, in the case of a circular valley ($m_\parallel = m_\perp; \lambda = 1$). The size and number of nodes of these wavefunctions increases with orbital index.
Figure 4.13: **Landau level wavefunctions in the symmetric gauge.** Plot of $|\psi_{N,m}(r)|^2$ in the symmetric gauge for various orbital indices $N$ and guiding center indices $m$. There is only one state for every $N$, the $m = N$ Landau orbit that has non-zero amplitude at the origin (boxed panels). Simulations for $B = 10 \, T$.

We emphasize that $\psi_{N,m}(Z)$ has a non-zero value at the origin only if $m = N$, in which case the LL wavefunction can be written as

$$\psi_{N,m=N}(Z) = \frac{1}{\sqrt{2\pi\ell_B^2}} e^{-\frac{|Z|^2}{4}} \mathcal{L}_N \left( \frac{|Z|^2}{2} \right). \quad (4.7)$$

The above simplification is important when we consider the effect of defects on individual Landau orbits.

Without the defect, conductance maps measured at the LL peak would include density of states contributions from all cyclotron orbits and no spatial variation is expected. However, in the presence of atomic-scale defects, there is one particular cyclotron orbit within the $N$th LL that has weight at the defect site; it is this state,
the $m = N$ state, whose energy is shifted by the defect potential, which gives rise to a missing Landau orbit that is measured as a decreased conductance in the shape of its wavefunction. We discuss these details below.

Due to the atomic length scale of the surface defects that we observe and the relatively strong screening in bismuth, we model the defect potential by a delta function, $U = \alpha \delta(r)$, where $\alpha$ is the strength of the defect potential. The energy shift of the $\psi_{N,N}$ state relative to the $\psi_{N,m \neq N}$ states is

$$E_{N,N} - E_{N,m \neq N} = \alpha |\psi_{N,N}(0)|^2 = \frac{\alpha}{2\pi \ell_B^2},$$

assuming that this shift is small compared to the LL spacing. Therefore, for measurements at the unshifted LL peak at energy $E_{N,m \neq N}$, the local density of states is proportional to

$$\sum_{m \neq N} |\psi_{N,m}(r)|^2 = \frac{1}{2\pi \ell_B^2} - |\psi_{N,N}(r)|^2,$$

whereas it is proportional to $|\psi_{N,N}(r)|^2$ for measurements performed at energy $E_{N,N}$. In the former case, which corresponds to the experimental measurements discussed thus far, maxima in $|\psi_{N,N}(r)|^2$ therefore correspond to local minima in conductance, as we observe (depicted schematically in Fig. 4.14).

This theoretical model of LL wavefunctions for the surface states of Bi(111) can be used to capture the elliptical features in the STM conductance maps near individual defects with excellent accuracy. Figure 4.15 shows a beautiful two-dimensional map of an isolated Landau orbit, which highlights the individual nodes within the $N = 3$ LL wavefunction. The only adjustable fit parameter is the anisotropy of the hole valley, i.e. the ratio of the effective masses. We extract a value of $\lambda = \sqrt{m_\parallel/m_\perp} \approx 5$ for the valley anisotropy, which matches remarkably to our experimental data (Fig. 4.12, Fig. 4.15) and is in good agreement with previous ARPES measurements [41, 43, 98].
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Figure 4.14: **Effect of a defect.** Schematic illustrating the effect of a defect on the density of states (DOS), which, depending on the sign of the defect potential $U$, increases or decreases the energy of the $m = N$ state relative to the rest of the degenerate $m$ states within the $N$th LL.

Figure 4.15: **Image of an individual Landau orbit.** (a) Conductance map of an $N = 3$ LL wavefunction, taken at $B = 12.75$ T and $E = 770 \mu$eV that captures the individual nodes in an isolated Landau orbit. (b) Theoretical simulation for $B = 12.75$ T and $\lambda = 5$ which matches the experimental data remarkably well.

and calculations [101]. For an individual anisotropic hole valley, the effective masses are $m_\parallel \approx m_e$ and $m_\perp = m_\parallel / 25 \approx 0.04 m_e$, respectively, where $m_e$ is the bare electron mass. This translates into a cyclotron energy $\hbar \omega_c = eB/m^* \approx 0.6 \text{ meV/T}$, with $m^* = \sqrt{m_\parallel m_\perp} \approx 0.2 m_e$, and corresponds to a LL spacing of $\hbar \omega_c \approx 8.4 \text{ meV}$ at $B = 14$ T, in agreement with our data.

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Our model also captures the field dependence of the elliptical LL wavefunctions. The size of a Landau orbit is proportional to $\sqrt{(2N + 1)\ell_B}$, where $\ell_B \propto 1/\sqrt{B}$. So increasing the magnetic field should lead to smaller rings, and for fixed $N$, the Landau orbit should scale as $1/\sqrt{B}$. Figure 4.12f shows the experimentally measured semimajor axis for the $N = 4$ LL; the fit to the data matches this $1/\sqrt{B}$ prediction to within the experimental uncertainty.

Based on the model described above, we anticipate that the suppression we have detected in the conductance maps at the LL peaks should be accompanied by an enhanced conductance relative to the background at the shifted energy. An example of such contrast reversal is shown in Fig. 4.16a-i, which displays conductance maps near an isolated defect over a range of energies within one broken-symmetry LL peak with orbital index $N = 4$ (Fig. 4.16j). The maps measured at the LL peak and at higher energies show ellipses of suppressed conductance that correspond to a missing cyclotron orbit. In contrast, the maps at energies below the LL peak exhibit ellipses of higher conductance, which indicates the defect has shifted the $m = N = 4$ state down in energy. The nodes of the LL wavefunction demonstrate a contrast reversal at these two different energies, as illustrated by the energy-averaged data shown in Fig. 4.16k. Thus, the cyclotron orbit energy has been lowered by about 300 $\mu$eV by this particular defect. Examining different defects, we have found evidence for both attractive and repulsive potentials from the contrast reversal in the conductance maps.

4.5.1 Previous STM Imaging in the Quantum Hall Regime

Our measurements on bismuth are in the clean regime where signatures of isolated cyclotron orbits are visible around individual defects, in contrast to previous STM imaging in the quantum Hall regime. We now discuss some of the challenges
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Figure 4.16: **Energy shift of Landau orbits.** (a)-(i) Spatial maps around an isolated impurity at $B = 10$ T with energy spaced by $100 \, \mu$eV throughout one broken-symmetry $N = 4$ LL peak. These maps show the shift to lower energy of the $m = N$ cyclotron orbit. (j) Corresponding conductance spectrum (averaged over a 12 nm $\times$ 2.5 nm area centered 5 nm below the defect) marked with colored circles for each mapped energy. (k) Oscillations of $G/\bar{G}$ along the semiminor axis, corresponding to the nodes in the $N = 4$ wavefunction, which highlights the contrast reversal in the maps at different energies.
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associated with directly visualizing quantum Hall wavefunctions in prior work on this subject.

Advances in quantum Hall materials, especially the discovery of graphene and topological insulators, have made it possible to move beyond initial 2DEGs formed at buried interfaces. Landau levels that appear as a sequence of sharp peaks in STM energy spectra have been studied for over a decade in a number of materials, in graphene [13–16, 102], 2DEGs on the surface of cleaved InSb(110) doped with Cs [18, 100, 103, 104] and topological insulator surface states [19, 20, 105]. However, disorder has hindered imaging LL wavefunctions, especially in material systems involving many different atom types, which are likely to deviate from stoichiometric ratios on the atomic scale. Conductance maps in these studies show variations that arise from drift states moving along equipotential lines in the disordered limit [16, 18, 105]. Some measurements have indirectly probed the size and shape of cyclotron orbits [103, 105, 106] by examining LL spatial dependence caused by potential modulations.

In STM experiments on graphene samples that can be made very clean, there is an unfortunate possibility that the metallic tip locally gates the sample. This gating varies as the tip is scanned on the surface, which may obscure wavefunction imaging. Cyclotron orbits that are shifted in energy by an isolated defect have been explored in graphene [14]. In this case, a charged impurity was found to shift multiple $m$ states within the LL, making it difficult to resolve clear features in the conductance maps. Quite recently, STM has visualized LLs confined to a circular potential in graphene, which leads to the appearance of a spectroscopic “wedding cake”-like features [107]. Furthermore, in Dirac systems such as graphene and topological insulators, there is a question of whether the two-component nature of the Landau levels may obscure the nodal structure in isolated wavefunction images [105].
Overcoming the challenges of local quantum Hall imaging in these various surface 2DEGs is an exciting future endeavor, as these systems can provide insight into other kinds of interaction-driven phenomena.

4.6 Outlook

Bismuth is an elemental material with extremely low disorder, and its surface state forms an accessible 2DEG that is relatively decoupled from the semimetallic bulk, which may provide some offset to screening from the STM tip. All in all, these features, fortuitously make the bismuth surface state well suited to local STM imaging in the quantum Hall regime. We performed direct two-dimensional mapping of isolated Landau orbits, which enabled us to visualize nematic order on the Bi(111) surface, where the anisotropic hole mass leads to anisotropic wavefunctions. By connecting the orientations of individual Landau orbits to the different anisotropies of distinct valleys, we were able to identify the underlying valley ordering. The energy resolution in our spectroscopic measurements further enabled us to quantitatively probe the role of interactions in generating these broken symmetry states. Using local measurements allowed for imaging nematic behavior without external polarizing fields that were necessary to macroscopically align all domains in transport experiments.

The Bi(111) 2DEG represents an interesting venue to explore electron-electron interactions within anisotropic valleys. The ability to bring the lowest hole-like LL to the Fermi level, either by external gating or doping, may allow for direct visualization of fractional quantum Hall states and Wigner crystallization with a STM. In addition, the boundaries between different nematic domains are expected to harbor low-energy edge modes that are analogous to topologically protected states [31]. The ability to generate a valley-polarized nematic phase that can be externally tuned with strain
make Bi(111) surface states ideally suited for controlled engineering of anisotropic physical properties. The predicted semimetal-to-semiconductor transition with decreasing thickness in bulk Bi [40] means that the transport properties of thin Bi(111) crystals will be dominated by the surface states, yielding further prospects for integration into devices that exploit the unique physical properties reported here.

4.7 Supplementary Information

4.7.1 Materials and Methods

Single Bi crystals were grown using the Bridgman method from 99.999% pure Bi that had been treated to remove oxygen impurities. The samples were cleaved in ultra-high vacuum at room temperature and immediately inserted into a home-built dilution refrigerator STM [64] and cooled to cryogenic temperatures. The cleaved Bi(111) surface shows almost no defects at this point, as illustrated Fig. 4.5, and the number of defects is stable at low temperature. Following a thermal cycle to 30 K and back down to 250 mK, a larger number of defects were present, as illustrated by Fig. 4.11. These are likely hydrogen molecules adsorbed on the surface due to outgassing of the microscope walls. Similar nematic behavior was observed both before and after the thermal cycle. Except where noted, measurements were performed at a temperature \( T = 250 \) mK using a tungsten tip.

Spectra and conductance maps were acquired using a lock-in amplifier at a frequency of 700.7 Hz and with ac rms excitation \( V_{rms} \) varying from 30 \( \mu \)V to 3 mV. Except where noted, the setpoint voltage was \( V_{set} = -400 \) mV and the setpoint current was \( I_{set} = 5 \) nA. We detail the measurement parameters for each figure below.
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Figure 4.5: \( I_{\text{set}} = 30 \) pA for panel a. \( V_{\text{set}} = 10 \) mV and \( I_{\text{set}} = 20 \) nA for panel b.

Figure 4.7: \( I_{\text{set}} = 100 \) pA, \( V_{\text{rms}} = 3 \) mV and \( T \approx 4 \) K for panel a. \( I_{\text{set}} = 2 \) nA and \( V_{\text{rms}} = 500 \) µV for panel b.

Figure 4.8: \( V_{\text{rms}} = 500 \) µV for panel a. \( V_{\text{rms}} = 250 \) µV for panel b.

Figure 4.9: \( V_{\text{rms}} = 100 \) µV for panel a. \( V_{\text{rms}} = 74 \) µV for panel b.

Figure 4.10: \( V_{\text{rms}} = 30 \) µV.

Figure 4.11: \( V_{\text{rms}} = 30 \) µV for panels a,e,h; \( V_{\text{rms}} = 74 \) µV for panels b-d,f,g,i-k.

4.7.2 Imaging of Electron-like Landau Levels

We perform spectroscopic imaging at the energy of the electron-like LL peaks (Fig. 4.17a–c) around the same atomic-scale surface defects shown in Fig. 4.5. We find approximately circular rings in the conductance maps, which indicates these wavefunctions arise from the central pocket around the \( \Gamma \) point, not the anisotropic electron pockets at the Brillouin zone edge at the M points. Moreover, these circular rings further confirm that the defects themselves do not break the rotational symmetry.

We can track the radius of the wavefunction from this central electron pocket as a function of magnetic field (Fig. 4.17d). The size of a Landau orbit is proportional to \( \sqrt{(2N+1)\ell_B} \), where increasing magnetic field should lead to smaller rings. For fixed \( N \), the size should scale as \( 1/\sqrt{B} \). Here we plot the electron orbits for LLs that occur at approximately constant energies (close to the Fermi level), but correspond to different \( N \). In a system with fixed charge, for states at a given constant energy, the orbital index \( N \) scales as \( 1/B \). This combined with the changing magnetic length explains the approximate \( 1/B \) scaling that we observe for the electron LLs near the Fermi level.
Figure 4.17: Imaging LL wavefunctions from central electron valley. (a)-(c) Conductance maps of the electron-like LL for a few orbital indices $N_e$ at different magnetic fields, taken in the area marked by the box in Fig. 4.5. Data shows approximately circular rings of suppressed conductance (black arrows) centered around the surface defects in that area. (d) Radius of electron LL wavefunctions as a function of magnetic field plotted on a log-log scale. Dashed line is fit with an extracted slope of $1.11 \pm 0.14$, which matches the expected radius $\propto 1/B$ relationship.

### 4.7.3 Surface Densities and Charge Redistribution

#### Hole Surface Density

The hole density of the Bi(111) surface states can be determined by the magnetic field at which each respective hole LL crosses the Fermi level (Table 4.1). At high magnetic field, where the LLs are pinned to the Fermi level for an extended field range, we use the midpoint as the crossing point, and take the LL to be half-filled at this point. Strictly speaking, this introduces a small error because filling factor $\nu$ and magnetic field are inversely related, so the midpoint in field is not the midpoint in filling factor. Nonetheless, this error is small, especially at low fields where pinning is not observed over an appreciable field range. Each LL crossing provides an independent measure of the total hole density $p = \nu e B / h$, and all derived values agree to within 2%, yielding an average of $p = (7.08 \pm 0.02) \times 10^{12} \text{ cm}^{-2}$. This consistency confirms that the surface hole density is fixed in our system, except for the variations described at the end of this section.
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<th>$B$ (T)</th>
<th>$\nu$</th>
<th>Derived $p$ ($10^{12}$ cm$^{-2}$)</th>
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<tr>
<td>14</td>
<td>21</td>
<td>7.11</td>
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<td>(N = 3 LL half filled)</td>
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<tr>
<td>10.875</td>
<td>27</td>
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<td>8.875</td>
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<tr>
<td>6.5</td>
<td>45</td>
<td>7.07</td>
</tr>
<tr>
<td>5.75</td>
<td>51</td>
<td>7.09</td>
</tr>
<tr>
<td>5.125</td>
<td>57</td>
<td>7.06</td>
</tr>
<tr>
<td>4.625</td>
<td>63</td>
<td>7.05</td>
</tr>
<tr>
<td>4.25</td>
<td>69</td>
<td>7.09</td>
</tr>
</tbody>
</table>

Table 4.1: Derived hole surface density: Magnetic field at which hole LLs cross the Fermi level, the corresponding filling factor, and the derived hole density $p = (7.08 \pm 0.02) \times 10^{12}$ cm$^{-2}$.

For a fixed surface charge density of $p \approx 7 \times 10^{12}$ cm$^{-2}$, at a magnetic field of $B = 14$ T, we can access a filling factor of $\nu = 21$ in the $N = 3$ Landau level. Due to the six-fold valley degeneracy of the Bi(111) surface state, to enter into the lowest Landau level at $\nu = 6$ would require $B \approx 50$ T, and to reach $\nu = 1$ would require fields of almost $B = 300$ T for this surface charge density.

Electron Surface Density

In principle, a similar procedure can be used to determine the surface electron density $n$. However, the electron case is more complicated because LLs could arise from the central hexagonal pocket and/or the three elongated pockets around the M points. Theoretical calculations [98, 101] show that the pockets around the M points are actually surface resonances due to strong hybridization with the bulk, suggesting our surface measurement is most sensitive to the pocket around the $\Gamma$ point. This is confirmed by the circular electron cyclotron orbits that we observe in Fig. 4.17; the oblong pockets around the M points would give rise to anisotropic rings similar to
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the hole states. We do not observe any LLs from the electron pockets around the M points, so we can only determine the surface electron density arising from the central pocket.

The lowest electron LL is not visible in our data, which is likely due to a combination lifetime broadening away from the Fermi level and surface-bulk hybridization caused by overlap of bulk states with the bottom of the surface band that gives rise to the central electron pocket [42]. Therefore, we cannot directly determine the orbital index \( N_e \) of each LL, and the filling factor needs to be assigned using a different method. We try three different filling factor assignments and calculate the corresponding density for each case according to \( n = \nu eB/h \) (Table 4.2). Only for the case where the electron LL crossing at 13.1 T corresponds to \( \nu = 9.5 \) do we find a constant carrier density from the central electron pocket of \( n = (3.02 \pm 0.02) \times 10^{12} \text{ cm}^{-2} \). For the cases where filling factor is higher or lower, the derived density either steadily increases or steadily decreases with field, inconsistent with our assumption of constant surface electron density. We therefore take the first case to be the correct assignment. Both this electron density and the derived hole density above are consistent with previous measurements of Bi(111) thin films [99].

Electron LL Pinning

Electron LLs exhibit pinning only when there are no proximal hole states, whereas they otherwise cross straight through the hole LLs at the Fermi level (Fig. 4.18). Although the electron and hole carrier densities that we measure are consistent across a wide field range, this difference in electron LL behavior suggests a temporary charge rearrangement between pockets. When there are no other states nearby (e.g. around 11.8 T), the electron LL is pinned to the Fermi level over an appreciable field range, indicating that the Fermi level is set by the chemical potential of the electrons. In
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Table 4.2: Derived electron surface density: Magnetic field at which electron LLs cross the Fermi level, and three possible assignments of filling factor ($\nu, \nu_-, \nu_+$) for each crossing with the corresponding derived electron density. The assignment of $\nu$ best yields a constant electron density $n = (3.02 \pm 0.02) \times 10^{12}$ cm$^{-2}$ as a function of field.

<table>
<thead>
<tr>
<th>$B$ (T)</th>
<th>$\nu$</th>
<th>Derived $n$ ($10^{12}$ cm$^{-2}$)</th>
<th>$\nu_-$</th>
<th>Corresponding $n_-$ ($10^{12}$ cm$^{-2}$)</th>
<th>$\nu_+$</th>
<th>Corresponding $n_+$ ($10^{12}$ cm$^{-2}$)</th>
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<tr>
<td>13.10</td>
<td>9.5</td>
<td>3.01</td>
<td>8.5</td>
<td>2.69</td>
<td>10.5</td>
<td>3.32</td>
</tr>
<tr>
<td>11.75</td>
<td>10.5</td>
<td>2.98</td>
<td>9.5</td>
<td>2.70</td>
<td>11.5</td>
<td>3.27</td>
</tr>
<tr>
<td>10.75</td>
<td>11.5</td>
<td>2.99</td>
<td>10.5</td>
<td>2.73</td>
<td>12.5</td>
<td>3.25</td>
</tr>
<tr>
<td>10.05</td>
<td>12.5</td>
<td>3.04</td>
<td>11.5</td>
<td>2.79</td>
<td>13.5</td>
<td>3.28</td>
</tr>
<tr>
<td>9.20</td>
<td>13.5</td>
<td>3.00</td>
<td>12.5</td>
<td>2.78</td>
<td>14.5</td>
<td>3.22</td>
</tr>
<tr>
<td>8.65</td>
<td>14.5</td>
<td>3.03</td>
<td>13.5</td>
<td>2.82</td>
<td>15.5</td>
<td>3.24</td>
</tr>
<tr>
<td>8.15</td>
<td>15.5</td>
<td>3.05</td>
<td>14.5</td>
<td>2.86</td>
<td>16.5</td>
<td>3.25</td>
</tr>
<tr>
<td>7.60</td>
<td>16.5</td>
<td>3.03</td>
<td>15.5</td>
<td>2.85</td>
<td>17.5</td>
<td>3.22</td>
</tr>
</tbody>
</table>

contrast, when a hole LL is pinned to the Fermi level, the electron LLs cross through it without any sign of pinning. At 10.9 T, the $N_e = 11$ LL is about 1 meV above the Fermi level and is therefore empty. By 10.7 T, it has already crossed through the $N_e = 4$ hole LL to negative energy (-1 meV) and is therefore completely filled. This corresponds to an increase in electron density from $2.90 \times 10^{12}$ cm$^{-2}$ to $3.10 \times 10^{12}$ cm$^{-2}$. In this regime, the Fermi level is primarily controlled by the chemical potential of the hole LL.

The difference in behavior signifies an interesting competition between carriers from the electron and hole pockets, and it can be understood by comparing the energetics of electron and hole LLs as a function of magnetic field. As magnetic field is decreased, the energies of the electron LLs decrease. In contrast, the energies of hole LLs increase as magnetic field is lowered (this corresponds to a lower energy for holes, but a higher energy for electrons). Therefore, it is energetically favorable for electrons to preferentially occupy states in the hole pockets at magnetic fields above crossings between electron and hole LLs, whereas it becomes more favorable to occupy states
Figure 4.18: **Electron LL pinning.** Data are averaged over the blue line in Fig. 4.5, and the lower field data are the same as those shown in Fig. 4.10a, but are reproduced here to improve readability. The electron Landau levels (LLs) exhibit two types of behavior as they cross the Fermi level: pinning when there are no proximal hole-like states (e.g. around 11.8 T), or rapid dispersion through hole LLs (e.g. at 10.8 T and 10.1 T). The two different behaviors indicate a competition between electron- and hole-like states in a magnetic field. $V_{rms} = 30 \mu V$.

In the electron pockets below the crossing. As magnetic field is decreased through a crossing, charge is transferred from the hole pocket to the electron pocket, effectively increasing both the electron and hole filling factors by one. The amount of charge transfer expected corresponds to one electron LL, or about $2.6 \times 10^{11} \text{ cm}^{-2}$ at 10.9 T, similar to our observed change in electron density. We remark that the charge transferred reaches 5-10 % of the total charge present in the central electron pocket. This is already substantial, and the charge transfer will be even larger at higher magnetic field. To the best of our knowledge, spontaneous charge transfer between electron and hole pockets has never been observed.
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We note that the behavior described above involves charge transfer between pockets in $k$-space; it does not imply charge transfer within the sample in real space. Our measurements probe this phenomenon locally, and a measurement at a single location does not imply global effects or uniformity, but we have observed similar behavior in multiple positions.

4.7.4 Exchange Gap Calculation

We outline below a calculation of the exchange splitting of the hole LLs on the Bi(111) surface based on the calculated wavefunctions from earlier in this chapter. We start with the equation for the exchange gap

$$
\Delta_{\text{exch}}(N, B) = \int \frac{d^2q}{4\pi^2} V(q) e^{-Q^2\ell_B^2/2} \left| \mathcal{L}_N \left( \frac{Q^2\ell_B^2}{2} \right) \right|^2,
$$

where wavevector $q$ is related to $Q$ by $q_x = \sqrt{\lambda}Q_x$ and $q_y = Q_y/\sqrt{\lambda}$, with $\lambda \approx 5$ for this system. We approximate the interaction potential $V(q)$ by the static screened interaction at zero magnetic field

$$
V(q) \approx \frac{2\pi^2}{\epsilon} \frac{1}{q + 2\pi e^2v_0/\epsilon},
$$

where $\epsilon$ is the dielectric constant and $v_0$ is the surface density of states at the Fermi level, which can be extracted from the LL spacing using the semiclassical quantization rule. At $B = 10.9$ T, $2\pi\ell_B^2v_0 \approx \frac{1}{7.6 \text{ meV}} + \frac{6}{6.1 \text{ meV}}$, where 7.6 meV and 6.1 meV are the respective LL spacing for electrons and holes, and the factor of 6 in the numerator of the second term takes into account the degeneracy of the hole pockets.

We measure a gap $\Delta_{\text{exch}} = 450 \mu$eV for the $N = 4$ hole LL at $B = 10.9$ T (Fig. 4.10). To reproduce this number using the above equations, we extract a
dielectric constant \( \epsilon = 45 \), which is about half the value in bulk Bi, as expected for a surface state bounded by vacuum on the other side. Using this same dielectric constant, we calculate theoretically expected exchange gaps for the \( N = 3 \) state at \( B = 12.9 \) T and at \( B = 14 \) T, which are 560 \( \mu eV \) and 600 \( \mu eV \), respectively. These numbers closely match the experimentally measured values of 570 \( \mu eV \) and 630 \( \mu eV \), respectively, which serves as a consistency check for the model.

### 4.7.5 Nematic Behavior in a Second Location

We present imaging of nematic behavior in the location of Fig. 4.5 (different from the area of Fig. 4.11). As described earlier, we observe three broken-symmetry states, two of which are split by exchange interactions as the otherwise four-fold degenerate hole LL crosses the Fermi level. A spectrum measured in this location at \( B = 10.9 \) T is shown in Fig. 4.19a, and the corresponding conductance maps at each broken-symmetry LL peak are shown in Fig. 4.19b-d. Similar to the data presented in Fig. 4.11, each broken-symmetry LL peak exhibits ellipses with different directionalities. We can quantify the role of exchange and strain in this location, which provides a second example where electron-electron interactions at the Fermi level play a dominant role in the formation of nematic electronic order. We also note that the energetic order of directionalities of the three broken-symmetry states is different from that in Fig. 4.11a-d, another example of a local nematic domain. For completeness, we also include a conductance map measured at the nearby electron LL peak (Fig. 4.19e), which shows approximately circular rings around the same defects.
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Figure 4.19: **Nematic behavior in a second location.** (a) Average conductance spectrum measured at \( B = 10.9 \) T along the blue line in Fig. 4.5. The \( N = 4 \) LL is split into three peaks, two of which are split by exchange interactions at the Fermi level. The \( N_e = 11 \) LL is also visible. The degeneracy of each hole LL is labeled in parentheses. (b)-(d) Conductance maps measured at the energies of each broken-symmetry hole LL peak. Isolated ellipses of suppressed conductance are visible around surface defects, with a different orientation for each LL peak. The energetic order of the three directionalities is different from that shown in Fig. 4.11b-d, indicating a different nematic domain. (e) Conductance map measured at the energy of the \( N_e = 11 \) LL. Approximately circular rings are visible around the same surface defects. \( V_{rms} = 30 \mu V \) for all panels.

4.7.6 Discussion of Strain in Atomic Lattice

Figure 4.5b shows an atomic resolution image of the Bi(111) surface which includes an isolated defect. The topograph, which was taken in the vicinity of the conductance maps in Fig. 4.11, shows a perfectly ordered lattice, demonstrating the high sample quality. This rules out variations in the vertical confinement of the 2DEG as a possible cause of the symmetry breaking that we observe. In addition, the topograph illustrates that the defect does not break the three-fold rotational symmetry of the lattice.

The corresponding Fourier transform of the Bi(111) atomic lattice, shown in Fig. 4.20, displays sharp Bragg peaks. From the positions of the Bragg peaks, we can extract the lattice constant in each direction. We obtain an in-plane interatomic spacing of approximately 4.67 Å, similar to literature values [92]. The lattice constant that we extract varies less than 5% between the three principal
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Figure 4.20: **Resolution of Bi(111) atomic lattice.** Fourier transform of an atomic image of the Bi(111) surface (Fig. 4.5). The Bragg peaks (circled), can be used to extract a lattice constant in each direction, of approximately 4.67 Å. Instrumental uncertainty from calibration and drift make it not possible to conclusively observe strain in the atomic lattice.

crystallographic directions. This variation is within the instrumental uncertainty that arises from a combination of piezoscanner calibration uncertainty as well as drift during measurement. Thus, we cannot conclusively say that we observe a strained lattice in atomic resolution images, though strain is likely present below our detection threshold (as argued above).

**4.7.7 Spectroscopic Linecut across Nematic Domains**

We have measured conductance spectra along a line that extends from one nematic domain to another, taken along the path shown in Fig. 4.21a. At either end of the linecut, the LL energies do not vary strongly with position (Fig. 4.21b), and these two regions respectively correspond to domains that are contiguous with those presented in Fig. 4.11. Although the spectroscopic signal remains strong except near the step edges, towards the middle of the linecut, the LL energies rapidly disperse
Figure 4.21: Measurement across a domain wall. (a) Topography of the Bi(111) surface. The arrow marks a strain defect on the surface which is visible as a deformation in the topographic signal. The white line shows the trajectory of the spectroscopic linecut, which starts in the domain shown in Fig. 4.11a-d and ends in the domain shown in Fig. 4.11h-k. $I_{set} = 30$ pA. (b) Spectroscopic linecut across a domain wall that shows the evolution of the three split $N = 3$ LL peak energies with position. The directionality of the real-space conductance features that we observe for each LL peak is labeled as white and black ellipses, respectively, for the two domains. $V_{rms} = 74 \mu$V.

with position and a clear LL crossing is visible about 400 nm from the start of the linecut (Fig. 4.21b). This is exactly the signature expected from a domain wall.

The domain wall occurs near a pronounced strain defect that is visible in the topograph (arrow in Fig. 4.21a), and strain may act as a catalyst for the domain wall formation. The dramatic LL dispersion near this strained region is further evidence that the single-particle LL splitting characterized by $\Delta_{\text{strain}}$ results from local strain in the sample.
In Chapter 6 we explore interacting behavior of boundary modes at a nematic domain wall, which forms spontaneously without topographic complications from strain defects or step edges.

4.7.8 Temperature and Doping Dependence of Nematic Behavior

In the absence of extrinsic symmetry-breaking terms, a nematic electronic phase is expected to have a critical temperature above which it becomes isotropic. We can gain some insight by comparing our data to previous 4.3 K measurements of a Bi(111) thin film [99]. The thin film showed similar electron and hole LL energies to our sample, but did not exhibit any broken symmetry in the hole LLs. This implies that either the measurements were performed above the critical temperature, or that thermal broadening made it impossible to resolve an exchange gap associated with nematicity. Regardless, in the absence of splitting, one would not expect to see any rotational symmetry breaking, so the measurements of Ref. [99] suggest that any nematic observable would not survive up to 4.3 K. We can estimate an upper bound for the temperature above which nematicity would disappear based on the magnitude of the exchange splitting that we observe. A gap of 500 \( \mu \text{eV} \) becomes equivalent to the thermal broadening \( 3.5k_B T \) at a temperature \( T \approx 1.6 \text{ K} \). This temperature threshold is consistent with the currently available experimental data.

Another example of an external tuning parameter that can affect nematicity is disorder, which has the further advantage that it is not complicated by thermal broadening. To explore the effect of disorder, we have performed measurements at 250 mK of a 1% Te doped Bi sample (Fig. 4.22). In these more disordered samples, cyclotron orbits of all three directionalities are visible throughout each LL peak.
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Figure 4.22: **Measurement of a doped Bi sample.** (a) Topography showing the surface of a Te doped Bi(111) sample. (b) Representative spatial map of $G$ that reveals pinned cyclotron orbits of all three directionalities at a single energy.

A representative conductance map taken at the Fermi level at $B = 10$ T is shown in Fig. 4.22b. Many superimposed cyclotron orbits are visible because so many dopants are present, and the presence of features of all three directionalities demonstrates that disorder destroys the nematic order. Further exploration of the rich phase diagram of broken-symmetry states that are expected as a function of temperature and disorder [108, 109] represents an appealing direction for future research.
Chapter 5

Ferroelectric Quantum Hall Phase Revealed by Imaging Landau Orbits

In the previous chapter, we explored the spatial signatures and emergent nematicity that occur when the six-fold valley degeneracy of the Bi(111) surface is partially lifted into doubly degenerate quantum Hall states. Here we investigate the consequences of a further spontaneous splitting of two-fold Landau levels (LLs) into singly degenerate states. We use a scanning tunneling microscope to perform mapping of singly degenerate LL wavefunctions in the vicinity of two types of defects. These high-resolution images establish the electronic ground state as arising from the occupation of a single valley instead of a coherent superposition of multiple valleys. Such valley-polarized states are expected to form a ferroelectric quantum Hall phase, in which individual Landau orbits possess an intrinsic in-plane electric dipole moment.

5.1 Introduction

Two-dimensional (2D) systems provide an attractive platform to explore broken symmetry phases because their electronic states often possess internal degrees of
freedom that are sensitive to external fields. Of particular interest is the role of valley
and spin degeneracies in the quantum Hall regime. Quantum Hall ferromagnetism
produces a variety of broken symmetry states across several 2D systems, including
spin- and valley-polarized ground states [10, 11, 85]. Recent measurements have
focused on multi-valley systems that exhibit more exotic electronic behavior. In
monolayer [36] and bilayer graphene [37] as well as transition metal dichalcogenides
[38], states with coupled spin and valley order have been identified based on their
response to applied electric and magnetic fields. Moreover, valley polarization has
been shown to lead to novel behavior such as nematic electronic order in systems
with anisotropic valleys [9, 10, 12, 21, 31]. A recent proposal suggests that valley
polarization can also result in a new type of broken symmetry quantum Hall phase: a
ferroelectric ground state in which the Landau orbits have an intrinsic in-plane dipole
moment [110]. These states can arise in materials which contain individual valleys
that lack two-fold rotational symmetry [34, 42], resulting in LL wavefunctions that
are not inversion symmetric. Although conventional ferroelectricity has been widely
studied in thin films [111–113], to date there have been no experimental reports of a
quantum Hall ferroelectric phase in any material system.

We focus on the six degenerate teardrop-shaped hole valleys of the Bi(111) surface
state (Fig. 5.1a), each of which has a distinct nontrivial spin texture (examples for
two valleys are shown in Fig. 5.1b-c) [101, 114, 115]. Due to spin-orbit splitting, there
is no remaining spin degeneracy, so the only tunable quantum degree of freedom is
the valley index. We define an effective filling factor  \( \tilde{\nu} \), which ranges from zero to
six and marks the occupancy of LLs within a given orbital index. At odd integer
filling, the Bi(111) surface states have the potential to exhibit a further symmetry
breaking by lifting the remaining degeneracy between pairs of valleys at opposite
momenta to produce a new class of quantum Hall state. There are two possibilities
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Figure 5.1: Teardrop-shaped anisotropic valleys. (a) Fermi surface for the Bi(111) surface states showing the electron pockets (grey) and the six anisotropic hole pockets, color-coded to match schematics below. (b),(c) Teardrop-shaped hole valleys $C$ and $\bar{C}$ at opposite momenta. The non-trivial spin textures are marked by arrows denoting the magnitude and direction of the in-plane spin component, and filled (open) circles denoting the $+(-)z$ out-of-plane component.

for such a ground state at $\tilde{\nu} = 1$ [109, 110]: a coherent superposition of different valleys, which gives rise to periodic charge density modulations or a valley-polarized state that is ferroelectric because each hole valley lacks two-fold rotational symmetry around its center. Recent theoretical work shows that the long-range part of the Coulomb interaction favors occupation of a single valley [110], and here we probe this valley ordering experimentally.

To address the nature of the electronic ground state, we perform LL spectroscopy of the Bi(111) surface, which allows us to resolve broken symmetry quantum Hall states at all integer filling factors. High resolution wavefunction mapping of singly degenerate LLs near isolated subsurface defects indicate single-valley polarization. These states not only exhibit nematic order, but are also expected to have an
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**Figure 5.2:** Possible ground states for singly degenerate LL. (a) Valley-polarized ferroelectric state in which only a single valley is occupied. (b) Valley-coherent state that consists of a superposition of states from the two valleys at opposite momenta. The different real-space spatial patterns of the LL wavefunction are shown; periodic charge modulations with a wavevector corresponding to the $k$-space separation of the two valleys is expected in (b).

in-plane electric dipole moment. Images in the vicinity of surface defects reveal unique signatures of interference between spin-textured valleys, which, in conjunction with a comprehensive theoretical model, further corroborates the valley-polarized nature of the ground state. Our observations confirm the recent prediction [110] that interactions in strongly anisotropic valley systems favor the occupation of a single valley, giving rise to emergent ferroelectricity in the surface state of bismuth.

### 5.2 Spontaneous Symmetry Breaking at Odd Integer Landau Level Filling

We first examine spectroscopic measurements of the Bi(111) surface state, which demonstrate a complete lifting of the six-fold valley degeneracy as the LL filling factor is tuned by changing magnetic field. The evolution of the differential conductance $G$ as a function of magnetic field $B$ in Fig. 5.3a reveals an intricate pattern of energy
gaps arising from a combination of single-particle and many-body effects. We focus on the dispersion of the hole-like LLs with orbital index $N = 3$ and $N = 4$. The six-fold valley degeneracy of these states is lifted, producing two- and four-fold degenerate LLs; the splitting between these multiplets is present at all magnetic fields, and is caused by local strain. Within each multiplet, Coulomb interactions further split the LLs as they cross the Fermi level. For instance, between 12.2 and 13.3 T, several additional energy gaps open and close within the four-fold degenerate multiplet of the $N = 3$ LL when it is pinned to the Fermi level, also shown in the corresponding spectra at particular magnetic fields in Fig. 5.3b. Two exchange-split LL peaks are visible throughout this entire field range, and their relative amplitudes and energies change as a function of magnetic field. This behavior indicates the formation of multiple distinct broken-symmetry phases as the number of occupied LLs within a given orbital index takes on all integer values from $\tilde{\nu} = 2$ to $\tilde{\nu} = 6$. Similar behavior is also visible when the two-fold degenerate multiplet of the $N = 4$ LL splits into two singly degenerate states (at $\tilde{\nu} = 1$) as it crosses the Fermi level around $B = 11.1$ T (spectra in Fig. 5.3d). These states at odd integer filling factors go beyond previous spectroscopic measurements, which showed only a partial lifting of the valley degeneracy into three doubly degenerate LLs [21].

Splitting within each multiplet occurs only at the Fermi level, indicating that these states develop spontaneously due to electron-electron exchange interactions. Individual spectra as the four-fold degenerate multiplet crosses the Fermi level (Fig. 5.3b) demonstrate the change in relative amplitude of the two split LL peaks, which match well to the above filling factor assignments. We quantitatively extract the magnitude of the exchange gaps for each broken symmetry state, which reaches a maximum of $\Delta_{exch} = 650 \mu$eV and is similar for all integer $\tilde{\nu}$ [Fig. 5.3c (blue)]. Exchange interactions also enhance the gap $\Delta_{strain}$ between LLs that are already
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Figure 5.3: Exchange splitting and broken-symmetry states at odd-integer LL filling. (a) Landau fan diagram indicating the dispersion of the electron-like (positive slope) and hole-like (negative slope) LLs as a function of magnetic field. The LLs are pinned to the Fermi level until they are completely occupied. The degeneracy of the hole LLs at select magnetic fields is labelled in parentheses; a full lifting of the hole valley degeneracy is seen for both a four-fold (4) and two-fold (2) degenerate LL due to exchange interactions at the Fermi level. (b),(d) Individual spectra at particular magnetic fields, marked by arrows in the LL fan diagram in (a), which highlight the relative amplitudes of the various LL split peaks. The number of occupied hole LLs within a given orbital index is specified by $\tilde{\nu}$. (c) Magnitude of the exchange gap, $\Delta_{\text{exch}}$ (blue), as the degeneracy of the four-fold multiplet is lifted around the Fermi level. Exchange enhancement of the strain-induced splitting, $\Delta_{\text{strain}}$, when the Fermi level lies between the four-fold and the two-fold degenerate peaks (red) is also shown.
split by strain by a similar amount, as shown in Fig. 5.3c (red). Thus, we observe interaction effects at all integer filling factors, including for states at odd filling factors that are cannot be split by uniaxial strain.

5.3 Landau Level Wavefunction Mapping to Identify Ground State

To address the valley occupation at odd integer filling factors, we first perform large-scale imaging of the Landau orbits at the energies of two singly degenerate $N = 3$ LLs (Fig. 5.4a-c). We observe the same wavefunction orientation for both of the singly degenerate LL peaks, which confirms that they arise from the subspace composed of the two valleys at opposite momenta, i.e. with the same anisotropy. These LL wavefunctions, which show up as multiple sets of concentric ellipses, are centered around surface (circles) and subsurface defects (arrows), as labeled in the concurrently measured topography (Fig. 5.4c). To first order, both types of defects can be treated as short-range potentials, which shift the energy of a single cyclotron orbit within each LL that has weight at the defect site. Therefore, measurements performed at the unperturbed LL energies show decreased conductance in the shape of a single electronic state, allowing for the direct imaging of isolated Landau orbit wavefunctions.

Imaging around impurities allows us to use the two types of defects (subsurface and surface) to independently demonstrate that the ground state at $\tilde{\nu} = 1$ is valley-polarized. We first examine high-resolution conductance maps around a subsurface defect, shown in Fig. 5.5. The absence of interference fringes reflects the lack of inherent charge modulation in the wavefunction, thus establishing the valley-polarized nature of the ground state.
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Figure 5.4: Large-scale imaging of odd-integer LL broken-symmetry states. (a),(b) Simultaneously measured large-scale conductance maps at the energies of the two singly degenerate LLs, showing the anisotropy and distinct nodes of individual wavefunctions pinned to defects. (c) Representative spectrum in this region of the sample, showing the exchange splitting of the two-fold degenerate multiplet into two singly degenerate LLs (blue and red dots correspond to energies of maps in (a),(b)). (d) Simultaneously measured topography of the Bi(111) surface, showing two atomic-scale surface defects (circled). Subsurface defects are not directly visible in the topography, and their positions, marked with arrows, are inferred from the centers of Landau orbits in the conductance maps.
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![Graph showing conductance spectra](image)

Figure 5.5: **Subsurface defect imaging.** (a) Conductance spectra on and off a subsurface defect showing exchange splitting of the two-fold degenerate multiplet around the Fermi level into two singly degenerate states. (b),(c) Conductance maps taken at the energies of the lower energy singly degenerate LL peak [blue dot in (a)] and the higher energy shifted cyclotron orbit peak [red dot in (a)], respectively. The insets show high resolution images that confirm the absence of interference fringes, and thus a valley-polarized ground state.

### 5.3.1 Expected Ferroelectricity

By imaging the wavefunctions near subsurface defects, we demonstrate that the ground state at odd integer filling factors involves the occupation of a single valley. Physically, the preference for valley polarization can be understood to follow from the fact that intra-valley exchange is always stronger than inter-valley exchange [110]. The teardrop Fermi surface shape of an individual valley further guarantees that the resulting state is a quantum Hall ferroelectric. However, the signatures of the corresponding dipole moment are subtle and difficult to resolve in the current experiment.
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Figure 5.6: Expected signature of ferroelectricity. (a) Simulation of the expected local density of states (LDOS) around a subsurface defect for an elliptical Fermi pocket. (b) Line cut of (a) along $x = 0$ showing perfect symmetry of the wavefunction with respect to the reflection $y \leftrightarrow -y$. (c) Simulation of the expected LDOS around a subsurface defect for a teardrop-shaped Fermi pocket (with realistic parameters). (d) Line cut of (c) along $x = 0$. The two dashed circles highlight the asymmetry in the LDOS with respect to the reflection $y \leftrightarrow -y$. The magnetic field is 12.75 T in these calculations.

The lack of two-fold rotational symmetry manifests as $|\psi(x, y)|^2 \neq |\psi(x, -y)|^2$. Figure 5.6a,c shows a comparison of the theoretically expected Landau orbits for an elliptical vs. a teardrop shaped Fermi surface, showing an almost unresolvable asymmetry in the wavefunction image itself. To more clearly illustrate the lack of two-fold rotational symmetry, we plot the local density of states (LDOS) along the long-axis of the wavefunctions in Fig. 5.6b,d. As highlighted by the red dashed circles in Fig. 5.6d, there is an asymmetry in the wavefunction nodes, particularly the innermost ones.

The experimental observation of this asymmetry is hindered by its small magnitude. Although the Landau orbits in Fig. 5.5 appear close to elliptical without
Figure 5.7: **Conductance variations through the center of the Landau orbits.** (a),(b) Experimentally measured conductance along the long axis of the wavefunctions around a subsurface defect. Data corresponds to vertical linecuts through the Landau orbits in Fig. 5.5b-c. The variation in background conductance and noise level make it impossible to definitively resolve asymmetry arising due to the lack of two-fold rotational symmetry.

any interference patterns, as theoretically expected, a plot of the conductance in Fig. 5.7 show background conductance modulations that are as large as the expected asymmetry in the wavefunctions. These modulations are unavoidable, due to variations in chemical potential over 100 nm length scales in our sample. They are sufficiently pronounced to obscure any potential consequence of ferroelectricity in the nodes of the wavefunction.

We expect that spontaneous valley polarization leads to ferroelectric domains in the sample, but the inability to directly measure the dipole through wavefunction imaging precludes identification of the local order parameter. Although our spectroscopic measurements are capable of identifying ferroelectric domain walls, no such boundaries were detected in this experiment, as discussed further in Sec. 5.6.3.
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5.4 Surface Defects and Landau Level Interference

In this section we investigate wavefunction maps in the vicinity of surface defects, which we use to corroborate the valley-polarized nature of the ground state and provide further insight into this system. The surface defects exhibit characteristically different behavior from the subsurface defects - conductance maps reveal interference patterns in the vicinity of the surface defects. By examining the detailed structure of these features, we establish that they are not an indication of a valley-coherent ground state with intrinsic charge modulations, but arise due to defect-induced valley mixing.

The spectra in Fig. 5.8a show exchange splitting of a two-fold degenerate LL and illustrate the shift in the LL peaks at the site of an isolated surface defect. Conductance maps with very fine spatial resolution taken at the energies of the two unperturbed, singly degenerate LL peaks are shown in Fig. 5.8b-c. Distinct vertical fringes with a wavevector corresponding to the separation between the two pockets at opposite momenta are clearly visible.

We now address the markedly different wavefunction imaging around subsurface and surface impurities. Although the potential induced by both types of defects are comparable in strength (as measured by the LL energy shift on the defect site), they differ in spatial extent, as shown schematically in Fig. 5.9. A defect below the surface has a smoother potential, which is sharp only relative to the magnetic length, but not on the atomic scale. Therefore, these subsurface defects can pin the Landau orbits but do not induce any mixing between LLs from different valleys. The absence of any interference fringes in wavefunctions around a subsurface defect reflects that the ground state is not in a coherent superposition of two valleys, due to the lack of inherent charge modulation. In contrast, the surface defect potential is atomically
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Figure 5.8: Interference fringes around surface defect due to valley mixing. (a) Spectra taken on a pristine Bi(111) surface (black) and on a surface defect (orange), demonstrating the shift in the LLs due to the defect potential. (b),(c) Conductance maps at the energies of the two singly degenerate LL peaks, showing distinct vertical interference fringes in the vicinity of the defect. (d) Schematic of the $N = 3$ LL energies, taking into account the lifting of the six-fold valley degeneracy due to strain and exchange interactions, as well as the energy shift of the $m = N$ state due to the defect. States in red and blue are singly degenerate, whereas those in yellow and green are two-fold degenerate. The point-like defect introduces important valley-mixing terms between states within the dashed box, with the most prominent interference processes marked by arrows. This diagram does not include any energy renormalization due to defect-induced mixing, but valley mixing is completely accounted for in all numerical calculations. (e),(f) Theoretical simulations of the LDOS at the unperturbed LL energies [3]-[4] for a valley-polarized ground state with defect-induced mixing, which shows good agreement with the data. (g),(h) In contrast, similar calculations of the LDOS assuming an intrinsic valley-coherent ground state do not match our observations.
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Figure 5.9: Spatial extent of different defect types. The defect potentials of subsurface (a) and surface (b) defects have different spatial extents. While the atomically sharp surface defects can be modeled as δ-function potentials, subsurface defects are modeled by a smoother gaussian potential.

sharp, which enables large momentum transfers that couple LL wavefunctions from multiple valleys, regardless of the nature of the ground state.

It is this disorder induced valley mixing that is responsible for the interference patterns in Landau orbits around surface defects. We further confirm that these patterns are not due to an inherent valley coherence of the ground state, because similar interference persists even for a two-fold degenerate multiplet, when both valleys are completely occupied (see Sec. 5.6.4).

5.4.1 Theoretical Model

We have developed a comprehensive theoretical model to better understand the interference between LL wavefunctions from different valleys, which confirms that the ground state at $\tilde{\nu} = 1$ is a valley-polarized ferroelectric. We construct an effective two-band Hamiltonian that captures the teardrop shape of the hole pockets of the Bi(111) surface states, and is in agreement with angle resolved photoemission spectroscopy (ARPES) measurements. We then calculate the intrinsic wavefunctions for both possible ground states in the case of a singly degenerate LL and use them to compute the local density of states (LDOS) in the vicinity of both types of defects (see Sec. 5.6.2). This theory accounts for the nontrivial momentum-space spin texture of
the hole pockets, which ensures valleys at opposite momenta have an overlapping spin component (Fig. 5.1) that permits mixing by a spin-independent disorder potential. A schematic of the energies used in our calculations for the $N = 3$ LL is shown in Fig. 5.8d, where the degeneracies of the different levels correspond to the spectra in Fig. 5.8a. The guiding centers that count the usual orbital degeneracy within a LL are indexed by $m$; the shifted zero angular momentum ($m = N$) states that have weight at the defect site are also included in the schematic.

The surface defect potential couples states with an energy difference comparable to the potential strength; the specific states from all six valleys that can interact within the framework of this model are marked by the dashed box in Fig. 5.8d. Numerical LDOS simulations for a valley-polarized ground state that incorporate this valley mixing match well to the experimental data (Fig. 5.8e-f), capturing the differences between each of the singly degenerate LLs. The strongest mixing occurs between the states coupled by arrows in Fig. 5.8d, which gives rise to the prominent vertical interference fringes. In contrast, simulations assuming a valley-coherent state (Fig. 5.8g-h) display periodic charge modulations that extend well beyond the vicinity of the defect and are inconsistent with our observations. Thus, in the case of a surface defect, theoretical simulations corroborate the valley-polarized nature of a singly degenerate LL.

5.4.2 Finer Features in Interference Patterns

The distinctive spatial patterns that we observe represent a different regime of wavefunction interference than is typically measured by STM. Whereas traditional quasiparticle interference (QPI) is the result of elastic scattering of the Bloch states of a crystal due to a defect potential [116], the interference presented here involves scattering between individual Landau orbits in a magnetic field, where the surface
defect can perturbatively couple states at different energies. The novel wavefunction mixing and the involvement of different valleys is evident from the conductance maps and the fast Fourier transforms (FFTs) of the fine interference patterns around surface defects (Fig. 5.10). The real space maps at the energies of the shifted counterparts of the singly degenerate LLs (Fig. 5.10a-b) are not a simple contrast reversal of the maps taken at the unperturbed LL peaks, but display additional diagonal interference patterns, which match well to the theoretical simulations in Fig. 5.10e-f. The corresponding FFTs (Fig. 5.10b,c,g-h) show multiple groups of peaks in the FFT that are arranged either in a line or in a diamond pattern, in contrast to isolated scattering wavevectors visible in typical QPI data. Each group is centered around wavevectors corresponding to the center-to-center distance between pairs of hole valleys, indicating that the defect couples states from all six valleys within a LL even though they have different energies.

The scattering patterns in the FFTs reveal several further details about the wavefunctions involved and their spin textures. The outer boundary of the groups in the FFTs (Fig. 5.10b,c,g-h) is due to the size, shape and relative angle between the valleys involved in the scattering process, while the number of high intensity points reflects the nodal structure of the wavefunctions and therefore depends on orbital index (see Sec. 5.6.2). In addition, although the LLs corresponding to the A and B valleys occur at the same energy, the signal in the FFT for $Q_{CA}$ is significantly weaker than for $Q_{CB}$, because of the stronger overlap between valley spin textures in the latter case (see labels in Fig. 5.1). We note that it matters only whether the pockets are adjacent, not the direction of their anisotropy; the signal around $Q_{AC}$ is strong, whereas that near $Q_{CA}$ is weak. Thus, by comparing the intensity of different groups of peaks, we can qualitatively determine the relative spin overlap for states in different valleys.
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Figure 5.10: **Diagonal interference patterns reflect nodal wavefunction character and spin texture of pockets.** (a),(b) Conductance maps at the shifted energies for $\tilde{\nu} = 1$ display additional diagonal interference features at the same field and around the same defect as in Fig. 5.8. (c),(d) FFTs of the experimental data in (a),(b) demonstrating the coupling of all six hole valleys. The centers of the boxes mark the wavevectors corresponding to the center-to-center distance between pairs of valleys (Fig. 5.1). (e),(f) Corresponding numerical simulations of the LDOS assuming a valley-polarized ground state, which match exceptionally well to the data in (a),(b). (g),(h) Fourier transforms of the theoretical simulations in (e),(f). The intensity of the peaks within a group is stronger for smaller energy differences and larger spin overlaps between the involved valleys.
5.5 Outlook

Our experimental approach to image the fine features of Landau orbits, resulting in the identification of a new class of broken symmetry quantum Hall state, has wider applicability. The single valley-polarized quantum Hall state, with emergent ferroelectricity studied here, is also expected to form in other anisotropic 2D valley systems, such as the surface states of topological crystalline insulators [32]. More broadly, our experimental approach to image LL wavefunctions with a STM can also be extended to identify other exotic correlated states, perhaps including phases containing skyrmions [117] or fractional quasiparticles [118] that can form in high magnetic fields.

5.6 Supplementary Information

5.6.1 Details of the Theoretical Model

Previous ARPES measurements and theoretical calculations have shown that the valleys of the Bi(111) surface states have distinctive spin textures involving both in-plane and out-of-plane components [101, 114, 115] that are not captured by a simple Rashba spin-orbit coupling model. In order to capture the important spin physics, which plays a critical role in the defect-induced mixing of valley polarized Landau orbits observed around surface defects, we develop a two-band model based on the symmetries of the Bi(111) surface. Using this model, we calculate the LL spectrum as well as the expected LDOS in the vicinity of both subsurface and surface defects. These numerical simulations support our experimental findings of a valley-polarized ground state at odd integer filling factors.
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Two-band Model

We construct a $k \cdot p$ model for the hole pockets on the Bi(111) surface in the absence of a magnetic field. The energy splitting between the two bands is due to the strong spin-orbit coupling in Bi and the breaking of inversion symmetry on the surface. In general, a two-band model can be expressed as

$$H_0(k) = h_0(k) + h(k) \cdot \sigma,$$

(5.1)

where $k$ labels the momentum, $h \cdot \sigma$ is a shorthand notation for $h_x \sigma_x + h_y \sigma_y + h_z \sigma_z$, and $\sigma_{x,y,z}$ are the Pauli spin matrices; $h_0$ and $h$ are real functions of $k$.

Symmetries put constraints on the form of $H_0(k)$. Because time reversal symmetry is preserved in the absence of a magnetic field, $H_0$ satisfies

$$\hat{T}H_0(k)\hat{T}^{-1} = H_0(-k),$$

(5.2)

where the time reversal operator is $\hat{T} = -i\sigma_y K$ and $K$ is the conjugation operator. It follows that $h_0(k) = h_0(-k)$ and $h(k) = -h(-k)$. At the $\Gamma$ point ($k = 0$), the two bands are degenerate due to time reversal symmetry and form a Kramers pair.

The crystalline symmetry on the (111) surface is $C_{3v}$, which contains a three-fold rotation $\hat{C}_3$ around the out-of-plane $\hat{z}$ axis and a mirror operation $\hat{M}$ with the mirror line along the in-plane $\hat{x}$ axis. The corresponding mirror operation acts not only in orbital part by taking $y$ to $-y$ and $k_y$ to $-k_y$, but also in spin space as $\hat{M} = i\sigma_y$. By the mirror symmetry,

$$\hat{M}H_0(k_x, k_y)\hat{M}^{-1} = H_0(k_x, -k_y),$$

(5.3)
where $\hat{M} = i\sigma_y$. Along the mirror-invariant line $k_y = 0$ (Γ-M), both $h_x$ and $h_z$ vanish and the two bands become eigenstates of $\sigma_y$.

We focus on the two pockets located along the $k_y = 0$ line; the other four pockets are related to them by $\hat{C}_3$ rotation. The two valleys are respectively centered around momenta $\pm \Lambda = (\pm \Lambda, 0)$, and extend only over a small range of $k_y$ (valleys $C$ and $\bar{C}$ in Fig. 5.1). We expand each term in $H_0(k)$ around $\pm \Lambda$ to lowest order in $k_{x,y}$, respecting time-reversal and mirror symmetries.

The effective $k \cdot p$ model is given as follows

$$H_\kappa(k) = E_0 + \Delta - \frac{\hbar^2 q_x^2}{2m_\parallel} + \hbar k_y (v_x \sigma_x + v_z \sigma_z) + [-\kappa \Delta + \hbar v q_x] \sigma_y,$$

where $\kappa = \pm 1$ for the right and left hole valleys, $q_x = k_x - \kappa \Lambda$, $E_0$ is the energy of the lower band at $\kappa \Lambda$, and $2\Delta$ is the energy splitting between the two bands. The main effect of $\hbar v q_x \sigma_y$ is to capture the absence of two-fold symmetry in the hole pocket, i.e. $E_{\kappa,-}(q_x, k_y) \neq E_{\kappa,-}(-q_x, k_y)$, which has important consequences for ferroelectricity. The effective masses along the $\hat{k}_x$ and $\hat{k}_y$ directions are $m_\parallel$ and $m_\perp$ and we define $v_\perp = \sqrt{\Delta/m_\perp} = \sqrt{v_x^2 + v_z^2}$.

Based on the $k \cdot p$ model, the two bands around momentum $\kappa \Lambda$ have energy dispersion

$$E_{\kappa,\pm} = E_0 + \Delta - \frac{\hbar^2 q_x^2}{2m_\parallel} \pm \sqrt{(-\kappa \Delta + \hbar v q_x)^2 + (\hbar v_\perp k_y)^2}.$$

The lower band $E_{\kappa,-}$ has an approximately parabolic dispersion, while the dispersion of the upper band $E_{\kappa,+}$ has a saddle point around $\kappa \Lambda$. The behavior of the two bands predicted by the effective Hamiltonian (Eq. 5.4) matches well to the extrema of the actual band structure (Fig. 5.11a). This agreement spans the relevant momentum range between the Fermi level crossings of the lower band. From first-principles calculations of the surface band structure [42], we obtain $\Delta \approx 90$ meV, $m_\parallel \approx 1.04m_0$, $m_\perp$. 

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Figure 5.11: **Details of the theoretical model.** (a) Dispersion of effective two-band model Hamiltonian used for all numerical calculations (blue dots), compared to Bi(111) band structure, adapted from [42] (solid lines), showing good agreement over the relevant momentum range. (b) Energies of LLs below $E_0$ as a function of $\Omega_c$ calculated within the two-band model (blue dots) and LL energies $-(N + 1/2)\Omega_c$ of a parabolic band with cyclotron energy $\Omega_c$ (solid lines).

$m_\perp \approx m_\parallel / 25$, where $m_0$ is the mass of the free electron and $\hbar v \approx 196$ meV·Å. There is an extra parameter $\phi$ that determines $v_{x,z}$: $v_x = v_\perp \cos \phi$ and $v_z = v_\perp \sin \phi$. From a first-principles calculation of the nontrivial spin texture [115], we extract the angle $\phi \approx -0.35$.

**Landau Levels for the Bi(111) Surface State**

In the presence of a magnetic field $B$, momentum $\hbar \mathbf{k}$ is replaced by $\mathbf{\pi} = \hbar \mathbf{k} + e \mathbf{A}$, where $\mathbf{A}$ is the vector potential. We use the symmetric gauge $\mathbf{A} = B(-y,x)/2$. The effective Hamiltonian for the $\kappa = \pm 1$ pockets then becomes:

$$
\mathcal{H}_\kappa = E_0 + \Delta - \frac{(\pi_x - \hbar \kappa \Lambda)^2}{2m_\parallel} + \pi_y (v_x \sigma_x + v_z \sigma_z) + [-\kappa \Delta + v (\pi_x - \kappa h \Lambda)] \sigma_y. \quad (5.6)
$$

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Before solving the LL problem, we discuss symmetry properties again. We recall that in the absence of a magnetic field, time-reversal operation $\hat{T}$ transforms one pocket into the other, while mirror symmetry $\hat{M}$ acts within one pocket. In the presence of a magnetic field, neither $\hat{T}$ nor $\hat{M}$ is a good symmetry, but the combined operation $\hat{T}\hat{M}$ remains a good one. In particular, the combined symmetry exchanges the two pockets $(\hat{T}\hat{M}) H_\kappa (\hat{T}\hat{M})^{-1} = H_{-\kappa}$, where $\hat{T}\hat{M}$ plays two roles: (i) it acts in real space by mapping $y$ to $-y$ due to the mirror operation; and (ii) it also acts as the conjugation operator $K$ because $(-i\sigma_y K)(i\sigma_y) = K$. Due to this symmetry, the energy spectra of $H_{+1}$ and $H_{-1}$ are exactly the same. The associated wavefunctions are also related: $\Psi_-(x,y) = \hat{T}\hat{M}\Psi_+(x,y) = [\Psi_+(x,-y)]^*$, where $\Psi_\kappa$ represents the two-component wavefunction of $H_\kappa$. However, crucially, the probability density $|\Psi_+|^2$ or $|\Psi_-|^2$ separately is generally not mirror symmetric, i.e. $|\Psi_\kappa(x,y)|^2 \neq |\Psi_\kappa(x,-y)|^2$.

Without going into the details of the derivation of the Landau level wavefunctions, here we present the salient analysis.

In the absence of a magnetic field, the $\kappa = +1$ pocket is centered on the $k_y = 0$ mirror line, and the associated spin therefore has a large component along the $+\hat{y}$ direction. To take advantage of this property, we make the following ansatz for the wavefunction of $H_{+1}$:

$$\Psi_{+,N,m}(r) = e^{iAx} \sum_{s=\pm} \left[ \sum_{n=0}^{n_c} u_{N,n,s} \psi_{n,m}(r) \right] |s\hat{y}\rangle,$$

where $N$ is the LL orbital index and $m$ is the guiding center index. $\psi_{n,m}$ is the eigen wavefunction of the $n$th LL for a parabolic band with anisotropic masses $m_\parallel$ and $m_\perp$; $n_c$ is a cutoff in the expansion, and should be properly chosen so that the LL wave function associated with the hole pockets converges in the numerical calculation; $|\pm\hat{y}\rangle$
represents the spin along the $\pm \hat{y}$ direction with the following convention

$$| + \hat{y} \rangle = \frac{e^{i\phi/2}}{\sqrt{2}} \begin{pmatrix} 1 \\ i \end{pmatrix} \quad | - \hat{y} \rangle = \frac{e^{-i\phi/2}}{\sqrt{2}} \begin{pmatrix} i \\ 1 \end{pmatrix},$$

(5.8)

where $\phi$ is the angle that determines $v_{x,z}$.

We diagonalize $\mathcal{H}_{+1}$ based on the ansatz $\Psi_{+,N,m}$. The numerical results of the energy spectrum are presented in Fig. 5.11b. The energies of the first few hole LLs closely follow $E_0 - \left( N + \frac{1}{2} \right) \Omega_c$, with $\Omega_c = \hbar e B / \sqrt{m_\parallel m_\perp}$, which is expected because the hole band is approximately parabolic with effective mass $\sqrt{m_\parallel m_\perp}$. Furthermore, from the numerical calculations (see [22] for details), we find the coefficients $u_{N,n,s}$ can be restricted to $n \in \{ N-1, N, N+1 \}$, resulting in the approximate wavefunction:

$$\Psi_{+,N,m}(r) \approx e^{i\Lambda x} \sum_{s=\pm} \sum_{n=N-1}^{N+1} u_{N,n,s} \psi_{n,m}(r) |s\hat{y}\rangle,$$

(5.9)

Moreover, $u_{N,N,+}$ is dominant over other coefficients. Therefore, the main character of $\Psi_{+,N,m}(r)$ is given by $\psi_{N,m}(r)| + \hat{y} \rangle$, which is consistent with the property of the hole pocket. The remaining parts in $\Psi_{+,N,m}(r)$ are due to the following effects: (i) the hole Fermi contour is not a perfect ellipse and instead has a teardrop shape and (ii) the electron spin on the $\kappa = +1$ pocket is generally not fully polarized along the $+y$ direction, and has finite components along the other two directions. LL wavefunctions of other hole pockets can be obtained from $\Psi_{+,N,m}(r)$ by using $\hat{T}\hat{M}$ and $\hat{C}_3$ symmetries.

### 5.6.2 Numerical Simulations and Comparison to Experiments

In this section, we discuss numerical simulations of the LL wavefunctions assuming a valley-polarized and a valley-coherent ground state in the vicinity of each impurity
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type. The two kinds of defects, surface and subsurface ones, provide independent measures of the electronic ground state. A direct comparison of our experimental observations to the theoretical analysis unambiguously establishes the ground state as a valley-polarized ferroelectric phase.

Valley-Polarized LL Wavefunctions and the Effect of Defects

Valley-polarized and valley-coherent wavefunctions have different spatial probability distributions, which we can use to distinguish between these two possibilities. First, we focus on LDOS simulations for a valley-polarized ground state. In the absence of any impurities, the LDOS of a valley polarized LL is spatially uniform:

\[
\sum_m |\Psi_{+,N,m}(r)|^2 = \sum_m \sum_{s = \pm} \left| \sum_{n = N-1}^{N+1} u_{N,n,s} \psi_{n,m}(r) \right|^2 = \frac{1}{2\pi\ell_B^2},
\]

where we have used the following identity

\[
\sum_m \psi_{n',m}^*(r) \psi_{n,m}(r) = \frac{\delta_{nn'}}{2\pi\ell_B^2}.
\]

Experimentally, isolated defects are crucial for the imaging of Landau orbits. The basic idea is most easily illustrated by considering LLs associated with a parabolic band and approximating the defect by a \(\delta\)-function potential. In this simplified scenario, only a single state within one LL, which is \(\psi_{n,m}(r)\) with \(m = n\), has a finite probability at the origin and is shifted in energy by the \(\delta\)-function potential. Therefore, spectroscopic mapping can directly probe the probability distribution \(|\psi_{n,m=n}(r)|^2\): imaging at the energy of the LL peak shows suppressed conductance around isolated defects, whereas imaging at the energy of the shifted cyclotron orbit shows bright features on a dark background. We now discuss several additional nuances beyond this simplified picture.
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The defect potential projected onto the $\kappa = \pm 1$ LLs is:

$$H_{\text{defect}} = \sum \langle \Psi_{\kappa_1,N,m_1}|U(r)|\Psi_{\kappa_2,N,m_2}\rangle c_{m_1,\kappa_1}^\dagger c_{m_2,\kappa_2},$$

(5.12)

where $U(r)$ is the defect potential localized around the origin. We assume $U(r)$ is spin independent, and therefore acts as a scalar potential in spin space.

We first take $U(r)$ to be a $\delta$-function potential: $U(r) = \gamma \delta(r)$. The matrix element $\langle \Psi_{\kappa_1,N,m_1}|U(r)|\Psi_{\kappa_2,N,m_2}\rangle$ then becomes $\gamma \Psi_{\kappa_1,N,m_1}^*(0)\Psi_{\kappa_2,N,m_2}(0)$. Using the approximation in Eq. 5.10, only three states ($m = N$ and $m = N \pm 1$) within the $N$th LL have a finite amplitude at the origin. The $\delta$-function potential has two main effects: (i) it shifts the energy of $m = N$ and $m = N \pm 1$ states relative to other states in the same LL. We find that this effect is strongest for the $m = N$ state, and is negligible for the $m = N \pm 1$ states; (ii) it mixes the $m = N$ and $m = N \pm 1$ states from opposite valleys, which is made possible by the $x$ and $z$ components of the spin. States associated with opposite valleys carry different phase factors $e^{\pm i\Lambda x}$, so this mixing leads to interference fringes. Moreover, this sharp $\delta$-function potential can induce scattering between all six valleys, giving rise to additional diagonal interference patterns.

In the case of a subsurface impurity, we model the defect potential by a Gaussian function

$$U(r) = \gamma \frac{1}{2\pi}e^{-r^2/L^2},$$

(5.13)

where the parameter $L$ characterizes the spatial extent of the defect potential. We assume that $L \ll \ell_B$, so the potential still influences the few states that are localized around the origin. This is reasonable if we assume the subsurface defects are in the layer immediately (4 Å) below the surface so that the extent of the defect potential is of order 1 nm. When $L$ becomes zero, $U(r)$ reduces back to the $\delta$-function potential.
Figure 5.12: Valley-polarized ground state wavefunction simulations. (a)-(c) Simulated LDOS at the unperturbed LL energy (corresponding to \( B = 12.75 \) T level [3] as labeled in Fig. 5.8) for a valley polarized ground state of a singly degenerate \( N = 3 \) LL. \( L/\ell_B \) is 0, 0.1, and 0.2, respectively, for (a), (b), and (c), where \( L \) characterizes the spatial extent of the defect potential. The defect-induced mixing that give rise to interference patterns weakens as \( L \) increases, capturing the different between conductance maps near surface and subsurface defects. (d)-(f) Identical simulations at the energy of the shifted Landau orbit (level [1] as labeled in Fig. 5.8). These simulations match well to our experimental observation defect types.

The matrix element of \( U(\mathbf{r}) \) between states from opposite valleys is

\[
\langle \Psi_{-,N,m_1} | U(\mathbf{r}) | \Psi_{+,N,m_2} \rangle = \int d\mathbf{r} e^{i2\Lambda x} U(\mathbf{r}) \tilde{\Psi}_{-,N,m_1}^* \tilde{\Psi}_{+,N,m_2}, \tag{5.14}
\]

where we have separated out the oscillating phase factor \( e^{i2\Lambda x} \). When \( U(\mathbf{r}) \) is finite over few periods of \( \ell_0 = 2\pi/(2\Lambda) \), the integral in Eq. 5.14 becomes vanishing small due to the oscillating part, and therefore, the subsurface defect can not mix states with different valley indices.
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Theoretically simulated LDOS assuming a valley-polarized state is presented in Fig. 5.12, for $L/\ell_B = 0, 0.1, 0.2$, or equivalently, $L/\ell = 0, 0.6, 1.2$. The energy diagram is schematically shown in Fig. 5.8d for the $N = 3$ LLs. Most states in $\kappa = +1$ LL have energies around level [3] (Fig. 5.12a-c). The vertical fringes that originate from mixing with the $\kappa = -1$ states have a period $\ell_0 = 2\pi/(2\Lambda) \approx 1.25$ nm. As $L$ increases, the visibility of the vertical interference fringes becomes weaker, eventually vanishing for $L/\ell_B = 0.2$, as is the case for subsurface defects. The interference patterns around the sharper atomic scale defects are not an indication of a valley-coherent ground state, but arise due to the defect-induced mixing of Landau wave functions from different valleys. The state at energy level [1] is shifted in energy from other states in the $\kappa = +1$ LL by the defect, and its main character is given by $\Psi_{+,3,3}$ (Fig. 5.12d-f). The defect potential only influences states that are localized around it. Far away from the defect, for example in the region where $|x| > 20$ nm, the valley-polarized LL has a uniform charge density. These calculations match well to our experimental data.

Valley-Coherent LL Wavefunctions

In contrast, because states originating from the two valleys carry opposite phase factors $e^{\pm i\Lambda x}$, the valley coherent state is spatially non-uniform and has a stripe-type charge density wave:

$$\sum_m \left| e^{i\alpha} \Psi_{+,N,m}(r) + \Psi_{-,N,m}(r) \right|^2 = \frac{1}{2\pi\ell_B} \left\{ 1 + \text{Re} \left[ \beta e^{i\alpha + i\Lambda x} \right] \right\}, \quad (5.15)$$

$$\beta = \sum_{s=\pm} \sum_{n=N-1}^{N+1} u_{N,n,s} \bar{u}_{N,n,s}, \quad (5.16)$$
where $e^{i\alpha}$ is a coherence phase factor, and $\tilde{u}_{N,n,s}^*$ is the corresponding coefficient of $\Psi_{-,N,m}$. If the spin associated with the $\kappa = +1(-1)$ hole pocket were uniformly aligned along $+\hat{y}(-\hat{y})$, then the oscillating terms in Eq. 5.15 would vanish. The coefficient $\beta$ is finite due to the spin texture in the hole pockets. We find $\beta \approx -0.26$ (for the $N = 3$ LL at $B = 12.75$ T), which makes the density oscillation in the coherent state a non-negligible effect. Thus, for a superposition of two valleys, we expect to see consistent striped periodic charge modulations that extend across the whole sample surface, regardless of the proximity to defects, instead of a uniform background as was the case for a valley-polarized ground state.

If a valley-coherent state is realized, the energy diagram is identical to that shown in Fig. 5.8d, but the levels [1]-[4] are a superposition of states from the $\kappa = +1$ and $\kappa = -1$ valley subspace, giving rise to a very different LDOS pattern. Our numerical simulations assuming a valley-coherent ground state at the energy of the LL peak show the periodic charge modulation is present over the entire map area (Fig. 5.13a-c), regardless of the type of defect. For such a state, the phase of the charge density wave is reversed for the two singly degenerate LL peaks far from the defects. Additional defect-induced mixing with the other four valleys is only present in the vicinity of a surface defect (Fig. 5.13a). In the experiment, vertical fringes are never found in the region far away from the defect. At the energy of the shifted state, the simulated conductance patterns reflect the inherent interference fringes within the Landau orbits (Fig. 5.13d-f). These simulations provide additional evidence against a valley-coherent state.

**A Few Remarks Regarding Defects**

We point out that the strength of the potential from subsurface and surface scatterers are actually comparable in magnitude, as measured by how much each impurity shifts
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Figure 5.13: Valley-coherent ground state wavefunction simulations. (a)-(c) Simulated LDOS at the unperturbed LL energy for a valley coherent ground state of a singly degenerate $N = 3$ LL. $L/\ell_B$ is 0, 0.1, and 0.2, respectively, for (a), (b), and (c), where $L$ characterizes the spatial extent of the defect potential. The intrinsic periodic charge modulations for this superposition state are visible over the entire area, irrespective of the defect potential extent, and do not match our experimental observations. (d)-(f) Identical simulations at the energy of the shifted LL. For all cases, the intrinsic charge modulations are visible in the regions of the high LDOS Landau orbit.

states from the central LL, which can be detected by spatially resolved spectroscopy. The surface defect shown in Fig. 5.8 shifts one Landau orbit from the central peak by about 550 $\mu$eV while the subsurface defect studied in Fig. 5.5 shifts a state by 300 $\mu$eV. These are typical energy shifts and fall within the range of 250 $\mu$eV to 650 $\mu$eV for all experimentally measured defects of both types. The strength of the potentials is not what differentiates these defects but instead it is the sharpness of their perturbation in real space, as we explain above.
We note here that neither these atomic scale defects nor the STM probe should change the fundamental nature of the ground state. The lack of interference fringes far away from both types of defects rule out the possibility that our data shows local valley polarization around defects surrounded by valley coherence elsewhere. Furthermore, because the defect potential is short range, its effect is local and so the presence of defects in the sample is not expected to change the preferred ground state. The theoretical argument, which also predicts a valley-polarized state, holds for a pristine crystal as it does not rely on the presence of defects. Finally, we do not expect the STM tip to distinguish between valleys at opposite momenta, or favor the occupation of one valley over the other.

Additional Simulations of Landau Level Wavefunction Interference

Several groups of scattering wavevectors are visible in the FFTs shown in Fig. 5.10. By performing theoretical simulations of wavefunction interference for several different orbital indices (Fig. 5.14), we confirm that the number of individual peaks within each diamond-shaped grouping increases with the number of nodes in the LL wavefunction. This unusual experimental signature in the FFTs of the interference patterns emphasizes that we are sensitive to mixing of individual electronic states, in contrast to traditional quasiparticle interference.

5.6.3 Additional Signatures of Ferroelectricity

The strength of the expected ferroelectricity can be characterized by the dipole moment, defined as

$$D_y = \int d\mathbf{r} \, y \, |\Phi(\mathbf{r})|^2. \quad (5.17)$$
Figure 5.14: Simulated FFTs of surface defect-induced interference for different orbital indices. (a)-(d) FFTs of theoretically simulated LDOS around a defect with a delta-function potential at the higher energy of a shifted singly degenerate LL state, for $N = \{0, 1, 2, 3\}$, respectively. The number of individual peaks within each grouping reflects the number of nodes in the wave function.

To theoretically calculate the magnitude of the dipole moment, we take $\Phi(r)$ to represent the state shifted in energy by the defect potential for the $\kappa = +1$ valley. The value of $D_y$ is about 0.4 nm for the $N = 3$ LL at $B = 12.75$ T, which is about 0.2% of the wavefunction extent, and thus a very small signal to detect experimentally.

The polarization density is another metric that can be used to quantify the expected ferroelectricity, and is defined as the dipole moment per area: $D_y/(2\pi\ell_B^2 d)$. For integer quantum Hall states, each electron occupies an area of $2\pi\ell_B^2$ and the vertical depth of the system can be approximated by the thickness of one bismuth bilayer $d \approx 0.4$ nm, yielding a polarization density of 0.05 $\mu C/cm^2$.

We expect local ferroelectric domains to occur in this system. Although we cannot distinguish the direction of the dipole moment in an isolated domain because of its small magnitude, our STM measurements have the sensitivity to reveal the
presence of a domain wall. Such ferroelectric boundaries would be visible in both the spectroscopic linecuts and the spatial conductance maps as the two singly-degenerate LLs cross, but so far we have not experimentally observed such regions. Our current measurements do not show domain crossings in linecuts over a 500 nm distance, suggesting that a typical domain separation is quite large, which makes it challenging to identify such a 1D object in the 2D space. Moreover, our inability to directly probe the orientation of the order parameter means that our wavefunction mapping cannot distinguish between isolated ferroelectric phases and therefore cannot provide clues regarding the locations of domain walls. Nevertheless, the search for ferroelectric domain walls is a promising future research direction to pursue.

In addition to the expected charge polarization, the occupation of a single valley would give rise to spin polarization as well. Our experimental probe is not sensitive to spin (beyond the relative spin overlap between different pockets), so we do not directly observe this effect. It is possible that future experiments could probe such physics through spin-polarized measurements or by using an in-plane magnetic field to couple to the spin degree of freedom.

5.6.4 Complementary Experimental Data

Imaging $\tilde{\nu} = 2$ LLs Around a Surface Defect

We further corroborate that the fine interference patterns result from defect-induced valley mixing as opposed to an intrinsic valley coherent state by imaging the defect in Fig. 5.8 at $B = 13.4$ T, where the two-fold degenerate multiplet is not split by exchange interactions (Fig. 5.15a). High-resolution conductance maps measured at the doubly-degenerate LL peak and at the energy of the shifted cyclotron orbit (Fig. 5.15b-c) exhibit nearly identical spatial patterns compared to the case of singly
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Figure 5.15: Interference patterns for two-fold degenerate LL around surface defect. (a) Conductance spectra on and off a surface defect at a field where the two-fold degenerate LL peak is not split by exchange interactions. (b),(c) Conductance maps taken at the energies of the LL peak on and off the defect, respectively [blue and red dots in (a)], showing interference patterns similar to those in the case of a singly degenerate LL. (d),(e) Corresponding theoretical simulations of the LDOS, confirming that these interference fringes arise from defect-induced valley mixing.

degenerate LLs shown in Fig. 5.8. This agreement is expected for interference arising from defect-induced scattering between valleys (Fig. 5.15d-e).

Imaging Other Odd-Integer Fillings

We have primarily focused on singly degenerate LL peaks that result from exchange splitting of a two-fold degenerate multiplet. However, similar behavior is expected to occur at all odd integer broken symmetry states. We have also performed spatial mapping for the case where exchange interactions separate a singly degenerate LL from a three-fold degenerate LL in an otherwise four-fold degenerate multiplet (Fig. 5.16). These data clearly demonstrate cyclotron orbits of two different orientations at the energy of the three-fold degenerate LL (Fig. 5.16b), with
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Figure 5.16: Imaging valley occupation for (1)-(3) splitting. (a),(b) Conductance maps measured for the case of a four-fold degenerate multiplet that is split by exchange interactions into a singly degenerate LL peak (a) and a three-fold degenerate LL peak (b). A representative spectrum at the magnetic field of 12.55 T in this region of the sample is shown in Fig. 5.3d.

conductance suppression around the defects more pronounced for one orientation. Imaging at the energy of the singly degenerate peak (Fig. 5.16a) shows Landau orbits whose orientation matches the weaker features in Fig. 5.16b. We therefore conclude that electronic states in one valley are split by exchange interactions from the remaining three valleys, which remain degenerate.
Chapter 6

Interacting Boundary Modes in a Quantum Hall Valley System

In this chapter, we uncover interacting behavior that occurs at the boundary between distinct nematic quantum Hall domains in bismuth, which host one or more sets of valley-polarized modes. The novelty of these domain walls is that they allow access to an interacting one-dimensional (1D) system in which the valley degree of freedom (in the 2D Brillouin zone) can dictate the emergent phenomena. Such symmetry constraints cannot emerge in a purely 1D quantum system; this behavior is a consequence of quantum Hall domain boundaries realized in conjunction with a topologically ordered bulk, which we show hosts unique interacting quantum channels.

6.1 Introduction

Domain walls between different broken-symmetry quantum Hall phases are predicted to host gapless 1D modes - quantum channels that emerge because of a topological change in the underlying electronic wavefunctions at such interfaces. Although a number of quantum Hall ferromagnet (QHFM) phases have been identified in different materials, interacting electronic modes at these domain walls have not
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been probed. So far, QHFM s have been observed via bulk measurements [7–12] and, more recently, with STM [21, 22]. However, transport studies, which measure macroscopically averaged electrical resistance, are poorly suited to address the microscopic properties of boundaries between distinct QHFM phases. On the other hand, with STM experiments on bismuth, the challenge has been to isolate the crossing of valley anisotropies at domain walls, because many of these nematic boundaries occur in the vicinity of pronounced strain defects or step edges which obscure the Landau level (LL) behavior. In this work, we locate a pristine interface between different electronic nematic phases without topographic complications and investigate the resultant interaction driven behavior of the boundary modes.

We present STM experiments of an interacting 1D system that forms at the boundary between distinct valley-polarized nematic domains on the surface of bismuth. Our spectroscopic measurements directly visualize these modes that occur within a topological energy gap, which closes and reopens as the valley polarization switches across the domain wall. By changing the valley flavor and the number of modes at the domain wall, we can realize different regimes in which the valley-polarized channels are either metallic or develop a spectroscopic gap. This behavior is a consequence of Coulomb interactions constrained by the valley flavor, which determines whether electrons in the topological modes can backscatter. These edge modes can be mapped to Luttinger liquids [119]; they are analogous to those studied in purely 1D systems [120–123], but with the novelty that valley flavor can dictate their properties. Our results, combined with a recent theoretical analysis of nematic quantum Hall domain walls [124], show that these systems harbor a unique class of symmetry-protected Luttinger liquids.
6.2 Imaging Valley-Polarized Domain Boundaries in Bismuth

We explore two QHFM domain walls that emerge when either one-out-of-four or two-out-of-four degenerate valleys are occupied. The boundaries of these nematic quantum Hall domains host one or two sets of counter-propagating valley-polarized 1D modes (Fig. 6.1a,b). We label these two cases by their effective filling factors, $\tilde{\nu} = 1$ and $\tilde{\nu} = 2$ respectively, where $\tilde{\nu}$ is defined as the number of occupied states within the relevant subset of valleys participating in the domain wall formation. For $\tilde{\nu} = 1$, the domain wall hosts a single pair of counter-propagating modes arising from different valleys (e.g. valleys $A$ and $B$, respectively; Fig. 6.1b). At $\tilde{\nu} = 2$, there are two pairs of counterpropagating modes, and the valley flavors of each of the co-propagating channels are also distinct (i.e. states from valleys $A$ and $\bar{A}$ moving in one direction and states from valleys $B$ and $\bar{B}$ moving in the other direction along the domain wall; Fig. 6.1a.). Such domains with different orientations of the broken valley symmetry are also topologically distinct - they have different ‘valley Chern number’, defined as $N_\nu = \nu_A + \nu_{\bar{A}} - \nu_B - \nu_{\bar{B}}$. Consequently, these boundary modes may be identified with the gapless conducting states that occur as a bulk topological invariant of the quantum Hall state changes across the domain wall, depicted schematically in Fig. 6.1c,d.

Experimentally, we use a combination of energy-resolved and spatially-resolved spectroscopy to study the QHFM domains and their 1D edge modes on the Bi(111) surface. The $dI/dV$ spectra in Fig. 6.2a,c show examples of different spontaneously broken symmetry states of the six hole valleys, where the occupation of a Landau level can be tuned by changing the magnetic field as discussed in previous chapters.
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Figure 6.1: **Boundary modes in a quantum Hall valley system.** (a),(b) Schematic of nematic domain walls between different broken valley symmetry quantum Hall states at effective filling factors $\tilde{\nu} = 2$ (a) and $\tilde{\nu} = 1$ (b), with Landau orbits denoted by the ellipses on either side. The expected valley flavor and degeneracy of the counter-propagating modes along the boundary depends on the filling factor as shown. (c),(d) Gapless modes expected to connect topologically distinct phases on either side of the domain wall, with corresponding valley flavors and degeneracies indicated in the case of $\tilde{\nu} = 2$ (c) and $\tilde{\nu} = 1$ (d).

In the vicinity of the domain wall we discuss here, strain lifts the degeneracy for two of the six valleys. Electron-electron interactions further split in energy the remaining four-fold multiplet around the Fermi energy, either into a pair of doubly-degenerate levels (corresponding to an effective filling factor $\tilde{\nu} = 2$; Fig. 6.2a), or, at a different value of the magnetic field, into one singly and one triply degenerate state ($\tilde{\nu} = 1$; Fig. 6.2c). The interaction-induced exchange gap between occupied and unoccupied valleys at the Fermi level ($\Delta_{\text{exch}} \approx 650 \mu\text{eV}$) is a signature of spontaneous symmetry breaking. We identify the valley ordering of these quantum Hall phases by imaging the corresponding Landau level wavefunctions and extend this capability to visualize the interface between different nematic domains and the associated boundary modes.
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Figure 6.2: Imaging a quantum Hall nematic domain wall. (a),(c) Representative Landau level spectra far away from the domain walls, showing gaps arising from strain, $\Delta_{\text{str}}$, and exchange, $\Delta_{\text{exch}}$, which lift the six-fold valley symmetry. Landau level degeneracies labeled in parentheses. Changing the magnetic field allows us to tune the effective filling factor, $\tilde{\nu}$, from $\tilde{\nu} = 2$ at $B = 14$ T, where exchange interactions split the fourfold degenerate multiplet around the Fermi level into two doubly degenerate states (a), to $\tilde{\nu} = 1$ at $B = 13.4$ T, with one singly and one triply degenerate state around $E = 0$ (c). (b) Fermi surface of the six-degenerate hole valleys, labeled to match different broken symmetry states discussed in this chapter. (d) Differential conductance map for $\tilde{\nu} = 2$ taken at the higher energy exchange split LL ($E = 400$ $\mu$eV), showing a distinct boundary between two domains. Anisotropic Landau orbits pinned to atomic scale defects appear as low conductance elliptical rings that have a consistent orientation on either side of the domain wall. (e) Large topography of pristine Bi(111) surface in the same field of view as (d) and (f), with uniformity in height of better than 40 pm over the entire region. Dashed line denotes position of linecuts in Fig. 6.3a,b. (f) Conductance map at energy of the triply degenerate state for $\tilde{\nu} = 1$ ($E = 330$ $\mu$eV), which displays a domain wall of low conductance similar to (d).
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We locate these nematic domain walls by mapping the spatial variations in conductance at the energies corresponding to the exchange-split LLs, as shown in Fig. 6.2d,f that are measured in large pristine areas of the Bi(111) surface (Fig. 6.2e). In these measurements, the domain walls appear as a stripe of low tunneling conductance between two regions with inequivalent broken rotational symmetry. The orientation of the elliptical LL wavefunctions reflect the direction of the anisotropy of the hole pockets (Fig. 6.2b) from which they originate, indicating the specific valleys associated with each nematic domain. Measurements taken in the same area of the sample as Fig. 6.2d but at a different magnetic field, show the $\tilde{\nu} = 1$ domain wall where the occupation of a single valley switches across the boundary. In this case, the conductance map measured at the higher energy exchange split peak (Fig. 6.2f), Landau orbits of two different orientations are visible on each side of the domain boundary, but elliptical features associated with one of the orientations on either side are more pronounced than for the other, in agreement with the expectations for a triply degenerate state (spectra in Fig. 6.2c). Additional conductance maps illustrate the energetic behavior of these nematic domains and show a switch of LL wavefunction directionality when probing the occupied versus unoccupied valleys (see Sec. 6.7.3). Finally, tuning the LLs away from the Fermi level or increasing the temperature, conditions in which exchange effects are absent or suppressed, result in the disappearance of the domains and domain walls (see Sec. 6.7.1). From these measurements, we conclude that nematic domains in our sample, with typical size of a few micrometers, are spontaneously formed due to electron-electron interactions, although local strain likely provides a small symmetry breaking perturbation that determines which valleys are occupied in each domain (see Sec. 6.7.1).
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6.3 Topological Boundary Modes

A hallmark of topological boundary modes is that they energetically occur in the gap of the corresponding bulk states and spatially reside where a topological invariant linked to this gap changes sign at an interface between two topologically distinct phases [125]. Spatially resolved STM spectroscopic linecuts measured across the domain walls shown in Fig. 6.3a,b demonstrate how the exchange gaps defining our local valley-polarized domains close and reopen for both the $\tilde{\nu} = 2$ and $\tilde{\nu} = 1$ cases. This profile of the gap and the measurements described above of the underlying valley state wavefunctions on either side, together demonstrate the effective sign change of the topological invariant at the domains walls in our system.

Spectroscopic maps obtained at the Fermi level reveal the spatial structure of electronic states within the gaps and exhibit high tunneling conductance along the domain walls, establishing the presence of low-energy boundary modes (Fig. 6.3c,d). The intrinsic topology associated with quantum Hall states is expected to host topological edge modes at the boundary between different valley-polarized domains. By extension of the chiral edge modes that traverse the perimeter of a quantum Hall 2DEG interface with vacuum, theoretically we expect these boundary modes between distinct nematic states to be counter-propagating. Although many topological electronic phases have been identified [125], including domain wall modes in bilayer graphene [126–128], these studies can be understood from a single particle perspective. Here we demonstrate a direct link between the closing of an interaction-induced energy gap and a spatial “twist” in a bulk topological invariant, and discuss below how these boundary modes can be mapped to strongly-interacting Luttinger liquids [124].
6.4 Interactions at One-Dimensional Domain Walls

Further investigation of the electronic domain wall behavior reveals that these 1D modes can be metallic or insulating depending on the nature of valley states from which they emerge. Although we observe the presence of low-energy modes along the boundary between topologically distinct nematic phases, individual spectra taken at the domain wall for \( \tilde{\nu} = 2 \) exhibit a local charge gap of \( \Delta_{\text{charge}} \) ranging from 325 \( \mu \text{eV} \) to 425 \( \mu \text{eV} \) (Fig. 6.4a; see also Sec. 6.7.2). We expect the \( \tilde{\nu} = 2 \) domain wall to contain two sets of counter-propagating modes, and naively expect this 1D...
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Figure 6.4: Individual spectra at domain boundary. (a),(b) Individual spectra taken at the location of the domain wall. For $\tilde{\nu} = 2$, a well-defined charge gap $\Delta_{\text{charge}} \approx 380 \mu$eV is observed (a), in contrast to a single peak at the $\tilde{\nu} = 1$ domain wall (b). The behavior far from the domain wall is shown in the dashed gray spectra.

boundary a better conductor than the $\tilde{\nu} = 1$ domain wall with only one set of counter-propagating modes. However, we find experimentally that the $\tilde{\nu} = 2$ domain boundary is insulating, i.e. it shows a spectroscopic gap. In contrast, at the $\tilde{\nu} = 1$ domain wall, we find a metallic behavior with a large density of states at the Fermi level and no resolvable gap (Fig. 6.4b; see also Sec. 6.7.2).

A crucial aspect of our experiment is that these strikingly different spectra for the two cases are observed in the same location on the sample. Thus, the $\tilde{\nu} = 1$ and $\tilde{\nu} = 2$ domain walls are subjected to the same background of impurities. If the $\tilde{\nu} = 2$ charge gap were induced by disorder, then we expect the same effects to produce a visible gap for the $\tilde{\nu} = 1$ modes, but the latter remain gapless (see Sec. 6.7.2 for measurements at multiple locations along domain wall). We therefore rule out localization from disorder as the source of the insulating state at $\tilde{\nu} = 2$. Instead, we attribute this charge gap to Coulomb interactions between 1D modes, which we
show below provides a natural explanation of why the presence or absence of this gap depends on the filling factor.

The experimental observation of a filling-factor dependent charge gap may be understood by considering all possible four fermion interactions between the different valley-polarized modes at the domain wall. We account for both the chirality, $r$, which denotes the direction of propagation, and the flavor, $\sigma$, which distinguishes the valley origin of co-propagating states of the boundary modes at different filling factors. We describe the interaction processes as $i_1 i_2 \rightarrow f_1 f_2$, where two incoming states $i_1$ and $i_2$ interact to produce the outgoing states $f_1$ and $f_2$. For $\tilde{\nu} = 1$, the two relevant channels are the counter-propagating $A$ and $B$ valley modes which can be labeled as $(r = 1, \sigma = 1)$ and $(r = -1, \sigma = 1)$, respectively. Interactions of the form $AB \rightarrow BA$ do not change the net chirality of the system, and therefore the domain wall remains gapless (Fig. 6.5c). Effective backscattering processes of the form $AA \rightarrow BB$ are exponentially suppressed due to the large momentum transfer between these valleys in the 2D Brillouin zone (Fig. 6.5d). Thus there are no interaction processes which can open up a gap at the $\tilde{\nu} = 1$ domain wall.

In comparison, $\tilde{\nu} = 2$ has two modes propagating in each direction with opposite flavors, i.e. $A$ and $\bar{A}$ have the same chirality $r = 1$ but different flavor indices, $\sigma = 1$ and $\sigma = -1$ respectively, and similarly the $B$ and $\bar{B}$ modes are labeled by $(r = -1, \sigma = 1)$ and $(r = -1, \sigma = -1)$. In this case, the interaction $A\bar{A} \rightarrow B\bar{B}$ changes the chirality of the system, and does so without any net momentum transfer in the 2D Brillouin zone (Fig. 6.5a), thereby opening up a gap. Heuristically, such interaction-induced backscattering can be understood by treating the valley flavor analogously to spin. Our experimentally-observed dichotomy between edge modes at $\tilde{\nu} = 1$ and $\tilde{\nu} = 2$ is similar to that seen in spinless versus spinful Luttinger liquids, where an energy gap is only expected in the latter case [119]. In addition to the
Figure 6.5: **Interacting multi-channel boundary modes.** Possible interactions between the relevant valley-polarized edge modes account for the difference in insulating versus metallic behavior at the domain wall. The four valley-flavored modes at the $\tilde{\nu} = 2$ domain wall can be gapped by interactions of the form $A\bar{A} \rightarrow B\bar{B}$ (a), which reverses the chirality of the modes (backscatters them) in a manner where no net momentum is transferred in the surface Brillouin zone. Processes such as $A\bar{B} \rightarrow B\bar{A}$ (b) do not conserve 2D momentum and are therefore exponentially suppressed. In the case of the $\tilde{\nu} = 1$ domain wall, which consists of two counter-propagating valley-polarized boundary modes, interactions are either gapless ($AB \rightarrow BA$; (c)) or exponentially suppressed due to the large 2D momentum transfer between states ($AA \rightarrow BB$; (d)).

charge gap at $\tilde{\nu} = 2$, a gapless neutral valley mode is also expected at the domain wall [124] (see Sec. 6.5 below), and the detection of such valley-charge separation in a Luttinger liquid would be an exciting avenue to pursue with other measurement techniques.
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6.5 Symmetry-Protected Luttinger Liquid Theory

In this section, we discuss the qualitative theoretical ideas underlying an effective Luttinger liquid model for these 1D domain wall modes. Details of the theoretical calculations can be found in Ref. [124].

There are four $U(1)$ symmetries at the level of the single-particle Hamiltonian describing electrons in each of the four valleys of interest. In the absence of interactions, these symmetries correspond to an independent conservation of the electron number $N_{\kappa}$ in each valley, with $\kappa \in \{A, \bar{A}, B, \bar{B}\}$. These generators can be rearranged into an equivalent set of four $U(1)$ charges:

\begin{align*}
\mathcal{N} &= N_A + N_{\bar{A}} + N_B + N_{\bar{B}} \quad \text{total charge} \quad (6.1) \\
\mathcal{I}^z &= N_A + N_{\bar{A}} - N_B - N_{\bar{B}} \quad \text{isospin} \quad (6.2) \\
\mathcal{P}^z &= N_A - N_{\bar{A}} + N_B - N_{\bar{B}} \quad \text{pseudospin} \quad (6.3) \\
\mathcal{Q}^z &= N_A - N_{\bar{A}} - N_B + N_{\bar{B}}. \quad (6.4)
\end{align*}

In the case of $\tilde{\nu} = 1$, $N_{\bar{A}} = N_{\bar{B}} = 0$, and these symmetries reduce to total charge conservation $\mathcal{N} = N_A + N_B$ and valley conservation $\mathcal{I}^z = N_A - N_B$. We call the degree of freedom between two valleys that share the same anisotropy (e.g. $A \leftrightarrow \bar{A}$) ‘pseudospin’ and that between such anisotropy pairs (e.g. $A \leftrightarrow B$) ‘isospin’. We focus on the isospin domain walls where the QHFM order parameter switches between anisotropy pairs.

Usually, metallic conduction along edges of quantum Hall systems is protected by the fact that chiral edge modes transport charge unidirectionally. In contrast, at these QHFM domain boundaries, the 1D modes counterpropagate. Unlike in usual 1D systems such as nanotubes, here scattering between left- and right-moving
states involves no change in momentum along the wall, since the position-momentum locking in the LL ensures that states at the same guiding center $X$ are proximate in momentum $p_y$. It then seems natural to expect these modes to become gapped and insulating. However, interactions in this system are constrained by 2D momentum conservation - interactions that involve rearranging electrons with a net valley momentum difference $K_{AB}$ are exponentially suppressed $\sim e^{-K\ell_B}$ and can be neglected. The remaining interactions that respect 2D momentum conservation only break select valley symmetries and lead to novel one-dimensional physics at the domain wall.

At $\tilde{\nu} = 1$, in particular, the constrained interactions do not break the two relevant symmetries $\mathcal{N}$ and $\mathcal{I}^z$. Conversely, at $\tilde{\nu} = 2$, interactions break $\mathcal{I}^z$ from a $U(1)$ symmetry down to $\mathbb{Z}_2$ - electrons from valleys $A$ and $\bar{A}$ can scatter to electrons in $B$ and $\bar{B}$ and vice-versa. Thus, we do not expect gapless modes associated with $\mathcal{I}^z$ and its conjugate $\mathcal{N}$, which results in a gapping of the charge mode as observed experimentally. The momentum-constrained interactions preserve the remaining symmetries, $\mathcal{P}^z$ and $\mathcal{Q}^z$. This, in turn, gives rise to a valley mode that is neutral and remains gapless.

The expected transport signatures for these two cases are markedly different as well. In the gapless $\tilde{\nu} = 1$ case, we anticipate no quantized Hall conductance because the metallic domain walls short the chiral edge modes. However, for $\tilde{\nu} = 2$, we expect this charge-valley separation and the gapping of the charge mode to give rise to a quantized Hall conductivity, whereas the neutral valley mode should be responsible for a non-quantized thermal Hall conductivity.

The Luttinger parameters and collective mode velocities are governed by a combination of the interaction strength, the anisotropy energy and the strain gradient that serves to pin the position of the domain wall to a specific location [124].
That the interactions are constrained by momentum conservation in two dimensions (the origin of the valley symmetry) is unique to the quantum Hall setting and the fact that such QHFM domain walls can only be realized in conjunction with a topologically ordered bulk \cite{124, 129} - such symmetry constraints cannot emerge in a purely 1D quantum system.

6.6 Outlook

The valley-polarized QHFM states examined here can be realized in a wide range of materials and provide the opportunity to examine not only different types of Luttinger liquids but also to explore novel ways of connecting them. Multi-valley systems such as graphene \cite{11, 37}, transition metal dichalcogenides \cite{38} or topological crystalline insulator surface states \cite{32, 34} can be used as material platforms to explore domain boundaries between a multitude of QHFM phases. These 2D systems naturally lend themselves to STM studies similar to those performed here that can both visualize electronic domain walls and probe the properties of their associated boundary modes. Moreover, the six degenerate Bi(111) valley states provide an intriguing opportunity for domain structures involving more than two phases, such as a meeting between three nematic states with different orientations at a single point. Theoretically, the possibility of such Luttinger liquid Y-junctions has been shown to have a variety of possible electronic behavior \cite{130}, whose interplay with the underlying quantum Hall and valley physics can potentially drive a rich phase structure. Finally, with the application of in-situ strain, it would be possible to control the location of domain walls in nematic QHFMs. Such strain tuning opens up the possibility of imaging domain wall dynamics as well as the potential to make these 1D boundary modes more accessible to other measurements, including transport studies.
6.7 Supplementary Information

6.7.1 Role of Exchange & Strain in Domain Wall Formation

Interactions play a central role in the formation of the domain wall, corroborated by the absence of two inequivalent nematic regions in measurements under conditions where exchange effects are absent. At a magnetic field corresponding to $\tilde{\nu} = 0$, where the four-fold degenerate LL completely unoccupied (i.e. tuned away from the Fermi level) and is not split by exchange (Fig. 6.6a), a conductance map taken in the same field of view as the maps in Fig. 6.2 shows Landau orbits of both orientations throughout the entire image (Fig. 6.6b). In this regime, the symmetry between these two orientations is not broken and no domain wall is present. The absence of domains is further highlighted by the linecut of spectra in Fig. 6.6c, which crosses the original position of the domain wall, but shows that the LL remains four-fold degenerate throughout. These observations confirm that the domains form as a result of exchange interactions, although the strain likely provides a bias that determines which valley(s) the QHFM spontaneously occupies.

Furthermore, exchange interactions are suppressed and the LL remains four-fold degenerate when the temperature is raised from 250 mK to 2 K, for the same magnetic field which produced the $\tilde{\nu} = 2$ domain wall (spectra in Fig. 6.6d). A conductance map at this higher temperature, taken in the same region as Fig. 6.2, shows two superimposed orientations of Landau orbits (inset Fig. 6.6e), in stark contrast to the nematic domains at base temperature. A corresponding linecut (Fig. 6.6f) across the initial domain wall location demonstrates that we cannot resolve exchange splitting between the two valley orientations. It is possible that the absence of splitting is a sign that the sample is above the critical temperature of the nematic transition or
Figure 6.6: **Role of electron-electron interactions in domain wall formation.**
(a) Spectrum away from the domain wall at $B = 13.1$ T and $T = 250$ mK, where the effective filling factor $\tilde{\nu} = 0$, so the four-fold degenerate LL is not split by exchange.
(b) Conductance map at the four-fold degenerate LL peak energy $E = 700 \mu$eV in the same area as in Fig. 6.2. The presence of cyclotron orbits of both orientations throughout the image indicates the absence of a domain wall under these conditions.
(c) Spectroscopic linecut along the dashed line in (b) also show a four-fold degenerate LL that does not change across the original location of the domain wall, in stark contrast to the linecut in Fig. 6.3. (d) Spectrum away from the domain wall at $B = 14$ T corresponding to the $\tilde{\nu} = 2$ domain wall but at a higher temperature of $T = 2$ K. Again, we do not resolve any exchange splitting and the LL is four-fold degenerate.
(e) Topography of the same area (identical to Fig. 6.2e) overlaid with a $dI/dV$ map at $E = -100 \mu$eV. We observe cyclotron orbits of both orientations at this elevated temperature, and no domain wall is visible. (f) Linecut along the dashed line in (e). The absence of splitting in the four-fold degenerate LL confirms that the domain wall is not present at 2 K.
it could reflect a decreased energy resolution from thermal broadening. Regardless, these data show that the domain wall does not exist in the absence of electronic interactions.

Although we show above that this nematic domain wall forms only in the presence of Coulomb interactions, it is likely that a local strain field stabilizes the position of the domain wall. Strain is a natural candidate that couples to the valley degree of freedom. Figure 6.7a shows a schematic of a possible strain field that gives rise to the particular valley splittings we observe. Specifically, there is a large strain in the direction of valleys $C$ and $\bar{C}$, which remains relatively unchanged across the domain wall and lowers the energy of these two valleys compared to the other four. However, in the presence of exchange interactions, a small switch in orientation of the strain field from favoring valleys $A$ and $\bar{A}$ to favoring valleys $B$ and $\bar{B}$ provides a symmetry-breaking perturbation which gives rise to a nematic domain wall. An experimental linecut across the $\tilde{\nu} = 2$ domain wall (Fig. 6.7b) shows that the energy of the two-fold degenerate valley state (at $E \approx 1.25\text{meV}$ corresponding to valleys $C$ and $\bar{C}$), which is split off from the other valley states by strain, does not change significantly across the domain wall associated with valleys $A, \bar{A}$ and $B, \bar{B}$.

### 6.7.2 Robust Domain Wall Behavior

We present additional measurements of linecuts across the $\tilde{\nu} = 2$ domain wall (Fig. 6.8a-f) and across the $\tilde{\nu} = 1$ domain wall (Fig. 6.8g-l). Figure 6.9 demonstrates that the difference in electronic behavior between the two filling factors at the domain walls is a robust feature. The small variations are likely due to disorder in the samples. Individual spectra from several locations at the domain wall exhibit an interaction-driven charge gap at the $\tilde{\nu} = 2$ domain wall (Fig. 6.9a), in contrast to the gapless spectra in the case of a $\tilde{\nu} = 1$ domain wall (Figure 6.9b). The spectra
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Figure 6.7: Schematic of strain field and comparison to experimental line cut. (a) Schematic of possible strain field (top) and the resulting energies of the different valley states (bottom). A large component of the strain in the direction of valleys \(C\) and \(\bar{C}\) lowers the energy of these two valleys compared to the other four. In the presence of exchange interactions, the switch in strain field from a slight favoring of valleys \(A\) and \(\bar{A}\) to valleys \(B\) and \(\bar{B}\) gives rise to the nematic domain wall. (b) Experimental line cut across the \(\tilde{\nu} = 2\) domain wall showing that the energy of the two-fold degenerate valley state at \(E \approx -1.25\) meV (corresponding to \(C\) and \(\bar{C}\)) is split off from the other valley states by strain, and does not change significantly across the domain wall associated with the crossing between pairs of valleys \((A,\bar{A})\) and \((B,\bar{B})\) at the Fermi level.

at the \(\tilde{\nu} = 2\) domain wall exhibit a charge gap, which ranges from 325 \(\mu\)eV to 425 \(\mu\)eV, as also seen in a line cut along the boundary (Fig. 6.10). The greater variation in spectra for \(\tilde{\nu} = 2\) at the domain wall is possibly due to a gap enhancement from atomic defect backscattering, which can further localize valley-polarized edge modes through inter-valley scattering. However, the isolated effect of individual defects is minimal and does not open a resolvable gap in spectra at the \(\tilde{\nu} = 1\) domain wall, which is measured with the same disorder potential as the \(\tilde{\nu} = 2\) domain wall, thus supporting our claim that this charge gap occurs due to Coulomb interactions constrained by valley flavor.
Figure 6.8: **Additional spectroscopic linecuts across the domain wall.** (a)-(f) Spectroscopic linecuts across the $\nu = 2$ domain wall. The six different linecut trajectories are indicated by the dashed lines in (m). While minor variations in the spectra are seen, likely due to the effects of local disorder, the key features of exchange gap closing and LL crossing at the domain wall are consistent. (g)-(l) Spectroscopic linecuts across the $\nu = 1$ domain wall. Again, the same features of the change in topological invariant are present in each linecut. (m),(n) $dI/dV$ maps reproduced from Fig. 6.2d and Fig. 6.2f, respectively, overlaid with dashed lines showing the locations of the spectroscopic linecuts in (a)-(l).
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Figure 6.9: Variation in individual spectra at domain wall. (a) Individual spectra (blue, green, brown, purple, black, red) measured for $\tilde{\nu} = 2$ (a) and $\tilde{\nu} = 1$ (b), at domain wall positions corresponding to Line 1 to Line 6 (Fig. 6.8). All spectra in (a) show a charge gap $\Delta_{\text{charge}}$ that is smaller than the exchange gap far from the domain wall (grey dashed spectrum). No LL splitting is visible at the $\tilde{\nu} = 1$ domain wall (b), in contrast to the behavior in (a) and far from the domain wall (grey dashed spectrum).

Figure 6.10: Linecut along $\tilde{\nu} = 2$ domain wall. (a) Linecut parallel to the $\tilde{\nu} = 2$ domain wall, along the white line in (b), showing spatial variation in the spectra. (b) Conductance map reproduced from Fig. 6.2d with the location of the spectroscopic linecut in (a) marked by the white line.
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6.7.3 Differential Conductance Maps at Additional Energies

The experimental measurements of differential conductance presented in Fig. 6.2 and Fig. 6.3 demonstrate the electronic behavior of a nematic domain wall for two key energies: at the higher energy exchange-split LL and at the Fermi level for both the $\tilde{\nu} = 1$ and $\tilde{\nu} = 2$ domain walls. These data clearly show the presence of two distinct regions with different broken rotational symmetry as well as low-energy states at the boundary between them. To further illustrate the evolution of electronic properties of these domain walls, we show additional conductance maps taken in the same area of the sample.

Figure 6.11 displays maps for the $\tilde{\nu} = 2$ domain wall, taken at $B = 14$ T, with approximately 100 $\mu$eV energy spacing. Comparing Fig. 6.11a to Fig. 6.11i highlights the switch in the Landau level wavefunction orientation and the corresponding valleys between the occupied and unoccupied exchange split states. These conductance maps in conjunction with the exchange splitting of a four-fold degenerate LL into two doubly degenerate states, establish that the domains occur from pairs of valleys at opposite momenta, specifically valleys $A$ and $\bar{A}$ in the left region and valleys $B$ and $\bar{B}$ to the right of the domain wall in Fig. 6.11i. Furthermore, as the energy is decreased from 400 $\mu$eV, the inward evolution of the regions of high differential conductance is a manifestation of the exchange gap closing in the vicinity of the domain wall.

We note here that in Fig. 6.11d,e, the orientation of the high conductance Landau orbits (i.e. those shifted in energy from their respective LLs due to defect potentials) is not fully uniform within each domain. This reflects the fact that certain defects shift the Landau orbit to higher energy, while others shift it to lower energy. As a result, for measurements at energies between the LL peaks, cyclotron orbits shifted upward in energy from the lower peak and downward in energy from the upper peak are both
simultaneously visible. We emphasize that the presence of both directionalities at these energies does not reflect any imperfection in the domains, whose uniformity is clear from Fig. 6.11a and Fig. 6.11i.

Changing the magnetic field to $B = 13.4$ T allows us to tune the filling factor of the LLs to $\tilde{\nu} = 1$, where the four degenerate LL is split into one singly- and one triply-degenerate state (Fig. 6.2c). Conductance maps under these conditions at several different energies are shown in Fig. 6.12. The three-fold degenerate LL corresponds to three of the valleys, and accordingly, conductance maps at approximately that energy (Fig. 6.12c,d), show dark Landau orbits of two orientations on either side of the stripe of low conductance that marks the domain wall. In either domain, one of the two orientations is more pronounced, in agreement with a triply degenerate LL, which arises from two valleys with the same anisotropy and a third valley with a different anisotropy direction. The favored directionality switches across the boundary as consistent with a nematic domain wall. Moreover, conductance maps measured at the energy of the singly degenerate LL show wavefunctions of only one orientation within each region (Fig. 6.12a), whose directionality matches the weaker direction in Fig. 6.12c,d and changes across the boundary. At this energy, there is also residual spectral weight from the tails of the gapless topological boundary mode (refer to Fig. 6.3d for width of the peak), which results in the enhanced boundary conductance that tracks the domain wall at $E = -120 \mu eV$.

The map at the Fermi level in Fig. 6.12b reveals increased conductance between domains, indicating the presence of low-energy edge modes at the domain wall. In the case of $\tilde{\nu} = 1$, a single hole valley switches its occupation with another one that has a different anisotropy orientation. Our previous work established that a singly degenerate LL in this system is valley polarized, however, we cannot experimentally
Figure 6.11: **Energy dependence of domain wall behavior at \( \tilde{\nu}=2 \).** (a)-(i) Differential conductance maps measured in the same location and under identical conditions to those in Fig. 6.2a,d. Each panel shows \( dI/dV \) maps at a different energy, ranging from the lower-energy exchange-split LL at \( E = -400 \ \mu\text{eV} \) (a) to the higher-energy LL peak at \( E = 400 \ \mu\text{eV} \) (i). The data demonstrate the different preferred wavefunction orientations for each respective domain as well as the different orientations of occupied and unoccupied states within a given domain.
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Figure 6.12: Energy dependence of domain wall behavior at $\tilde{\nu}=1$. (a)-(d) Differential conductance maps measured in the same location and under identical conditions to those in Fig. 6.2c,f. Each panel shows $dI/dV$ maps at a different energy, ranging from the singly degenerate LL at $E = -120 \mu eV$ (a) to the triply degenerate LL peak at $E = 330 \mu eV$ (d).

distinguish which specific valley is occupied [22]. Without loss of generality, we label the two states that cross at the $\tilde{\nu} = 1$ domain wall by valleys $A$ and $B$.

6.7.4 Theoretical Simulations of Domain Wall Conductance

Theoretically, we first describe a Hartree-Fock mean-field ansatz that allows us to model the tunneling spectra at the domain wall. The analysis shows general qualitative and quantitative agreement with experimental data, capturing essential features including the magnitude of the spectral gap, the width of the domain wall,
and the asymmetry in tunneling spectra along the axis transverse to the wall. This mean-field analysis however excludes interactions that become relevant for the \( \tilde{\nu} = 2 \) case, and can open up a gap in the tunneling spectra, as observed in the data.

The highly anisotropic shape of the Landau orbits is due to the large effective mass anisotropy \( (m_\parallel / m_\perp \approx 25) \) of the bismuth valleys [21, 40] which favors an abrupt change in the valley occupation across the nematic domain wall [108]. The extent of the wavefunctions perpendicular to the domain wall determines the spatial width of the boundary modes (approximately 100 nm in Fig. 6.2d,f and Fig. 6.3c,d). Detailed numerical Hartree-Fock calculations capture the closing and opening of the exchange gap and the experimental width of the domain wall as shown in Fig. 6.13a,b. Furthermore, the experimental spectra reveal an asymmetry in the amplitude of the
LL peaks near the domain wall (Fig. 6.13c), which is also accounted for in the theoretical modeling of the local density of states (Fig. 6.13d). This asymmetry is a signature of a dipole moment that arises from the different spatial extent of the Landau orbits from the two differently oriented valleys projected onto the 1D boundary [108].
Chapter 7

Concluding Remarks

The Bi(111) surface represents a unique platform in which to explore electron-electron interactions within an anisotropic valley system in the quantum Hall regime. That its surface state forms a quantum Hall 2DEG which offers direct access to scanning probes is the basis for the experiments presented in this thesis. The anisotropy of six degenerate, nearly-elliptical pockets of the Bi(111) Fermi surface couples the internal valley degree of freedom with the spatial symmetries of the system. We find a combination of Coulomb interactions and single particle strain can lift this degeneracy of the Landau levels, giving rise to a multitude of broken-symmetry states. We use a scanning tunneling microscope to visualize the underlying quantum Hall wavefunctions, which directly captures anisotropic spatial signatures of these valley-ordered phases, while energy-resolved spectroscopy ascertains the role of interactions. In Chapter 4, we present a direct manifestation of a nematic quantum Hall phase, in which the electronic wavefunctions of doubly degenerate LLs break the rotational symmetry of the underlying crystal lattice. In Chapter 5, we further establish the ground state of a singly degenerate LL as valley-polarized, which is expected to be a ferroelectric phase that carries an in-plane electric dipole moment.
All of the experiments discussed in this thesis have been restricted to spontaneous symmetry breaking in the integer quantum Hall regime. A natural extension for future research is the direct visualization of fractional quantum Hall states, which has never been reported before in any material. Fractional quantum Hall states originate from electron-electron interactions, which are strongest in the lowest Landau level. As it stands, the carrier density of the surface states of pure bismuth is too large to make it experimentally feasible to enter the fractional quantum Hall regime at low filling factors, especially with the additional six-fold degeneracy of each Landau level. Possible solutions to bring the lowest hole-like LL to the Fermi level include (i) modulation doping to shift the chemical potential while maintaining a pristine, low-disorder surface that is free from dopants or (ii) external gating of epitaxially-grown bismuth thin films, that are predicted to be semiconducting below a critical thickness \(d < 30 \text{ nm}\) [131]. The surface of silicon provides an alternative material platform that is accessible to local probes, in which fractional quantum Hall states have been observed in transport [10].

The origin of many fractional quantum Hall states can be described by a mapping to composite fermions, quasiparticles that minimize the Coulomb repulsion in this regime. These composite fermions consist of electrons bound to an even number of magnetic flux quanta, which experience an effective magnetic field that is significantly lower than the applied external field. Recent theoretical work suggests that it may be possible to image cyclotron orbits of composite fermions [118]. An ongoing field of research is exploring how the anisotropic electron band structure is reflected in the composite fermion behavior in the fractional quantum Hall regime. Novel transport experiments in AlAs have found persisting anisotropies in the composite fermion Fermi surface at half filling [132, 133], and these ideas have been explored theoretically as well [134]. Direct imaging of the underlying fractional quantum
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Hall wavefunctions would be ground breaking, and provide local information of the interactions and resulting anisotropies of composite fermion cyclotron orbits. Moreover, our experimental approach to image LL wavefunctions with a STM can also be extended to identify other exotic correlated states, including Wigner crystals or skyrmions.

Another direction for future experimental efforts lies in the investigation of boundaries between different quantum Hall ferromagnetic domains. In Chapter 6, we show that nematic domain walls form an interacting 1D system that can be mapped to a unique class of symmetry-protected Luttinger liquids, in which the valley degree of freedom in the 2D Brillouin zone dictates the emergent phenomena. Such symmetry constraints are a consequence of the quantum Hall domain boundaries realized in conjunction with a topologically ordered surface. The six degenerate Bi(111) valley states provide an intriguing opportunity for domain structures involving more than two phases, such as a meeting between three nematic states with different orientations at a single point. Theoretically, the possibility of such Luttinger liquid Y-junctions has been shown to have a variety of possible electronic behavior [130], whose interplay with the underlying quantum Hall and valley physics can potentially drive a rich phase structure.

More generally, domain walls capture the evolution between distinct topological phases; the even richer set of phenomena that arises from the combination of topology with broken symmetries can be extensively studied by controllably tuning interactions and external electromagnetic and strain fields, for which the bismuth surface is one such versatile platform. The application of in-situ strain opens up the possibility of tuning the location of domain walls while imaging its dynamics together with studying its conduction mechanism in transport measurements. Such experiments could simultaneously provide independent measures of gaps at both the microscopic
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and macroscopic levels. Furthermore, the ability to generate a valley-polarized nematic phase that can be externally tuned with strain make Bi(111) surface states ideally suited for controlled engineering of anisotropic physical properties. The predicted semimetal-to-semiconductor transition with decreasing thickness in bulk Bi [40, 131] means that the transport properties of thin films of bismuth will be dominated by the surface states, yielding further prospects for integration into devices that exploit the unique physical properties reported here.

Moreover, there are a wide range of materials in which these kind of valley-polarized quantum Hall ferromagnetism behavior can be realized [30]. These systems provide an opportunity to examine not only different types of Luttinger liquids but also to explore novel ways of connecting them. Multi-valley systems such as graphene [11, 37], transition metal dichalcogenides [38] or topological crystalline insulator surface states [32, 34] can be used as material platforms to explore a multitude of QHFM phases and domain boundaries between them. These 2D systems naturally lend themselves to STM studies similar to those performed here. The application of an in-plane magnetic field can also be used to introduce a controlled splitting of inequivalent valleys. Additionally, the entire phase-space of quantum Hall nematics as a function of temperature and disorder remains relatively unexplored [30].

Finally, another interesting direction to pursue involves investigating the interplay between quantum Hall and superconductivity. A recent theoretical work motivated by our experiments on bismuth, explores the behavior of a Rashba spin-orbit-coupled 2DEG in a strong magnetic field proximitized by a type-II s-wave superconductor. Experimentally, this setup may be realized by epitaxially growing bismuth thin films on a type-II superconductor such as FeSe or NbN, which form a vortex lattice in a magnetic field. Such systems have prospects for engineering topological superconductivity and unpaired Majorana zero modes [135].
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