Visualizing Surface States of Topological Insulators with Scanning Tunneling Microscopy

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Abstract

Topological insulators are materials that have a bulk band gap similar to commonly known insulators, but have conducting states on their edge or surface. The bulk band gap is generated because of the strong spin-orbit coupling inherent to these systems, which also modifies the band in a fundamental way, leading to unconventional spin-polarized Dirac fermions on the boundary of the insulator. In this thesis, we present our scanning tunneling spectroscopy studies of topological surface states in $\text{Bi}_{1-x}\text{Sb}_x$, $\text{Sb}$, $\text{Bi}_2\text{Te}_3$ and $\text{Bi}_2\text{Se}_3$. Due to their helical spin texture, the surface states of topological insulators are distinct from the well-known conventional surface states of noble metals. A key predicted feature of these metallic surface states is their immunity to localization and ability to overcome barriers caused by material imperfections. These predictions can be examined by studying the scattering of surface states from random alloying disorder. We have mapped the interference of the surface states in $\text{Bi}_{0.9}\text{Sb}_{0.1}$ and demonstrated that despite strong atomic scale disorder, backscattering between states of opposite momentum and opposite spin is absent, resulting from the helical spin texture of the surface states. Furthermore, we have measured transmission and reflection of the topological surface states of $\text{Sb}$ by atomic terraces. In contrast to surface states of noble metals, these surface states penetrate such barriers with high probability. These results experimentally demonstrate the fundamental difference of these surface states in comparison to other known surface states, and show their potential to be used for spin-based electronics and nano-scale devices.

In recent years, the topological surface states of $\text{Bi}_2\text{Te}_3$ and $\text{Bi}_2\text{Se}_3$, the "second generation" topological insulators, has become the focus of intense research. The single Dirac cone surface states on these compounds constitutes the simplest manifestation of 3D topological insulators. Many of the interesting theoretical proposals that utilize topological insulator surfaces require the chemical potential to lie at or near the surface Dirac point, and consequently bulk doping is commonly used to tune the chemical potential to the Dirac point. We have studied the surface states of $\text{Bi}_2\text{Te}_3$ and $\text{Bi}_2\text{Se}_3$ in the presence of magnetic and non-magnetic dopants. Bulk doping results in strong nano-scale spatial fluctuations of the surface states’ energy and momentum. In spite of these fluctuations, Dirac electrons show a remarkable robustness to backscattering that can be understood based on their helical spin texture, which is preserved even in the presence of magnetic dopants or bulk magnetism. While we show that these strong spatial fluctuations influence the transmission of topological surface states, we find no evidence for their localization by bulk or surface disorder. In the vicinity of the Dirac point, the energy and momentum fluctuations we observed would result in
spatially alternating spin helicity. This could possibly limit the mobility of topological surface state near the Dirac point. Our findings suggest that utilization of helical Dirac fermions on topological insulators requires methods of tuning the chemical potential which do not involve chemical doping.
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To my wife,

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Chapter 1

Birth of topological insulators

1.1 Insulators with metallic edge states

Recent theoretical and experimental progress in condensed matter physics has demonstrated the existence of a novel class of bulk insulators with conducting states on their boundaries [1, 2, 3, 4, 5, 6, 7, 8]. They are referred to as topological insulators (TIs). In this chapter we provide an introduction to two-dimensional (2D) and three-dimensional (3D) TIs. TIs can be understood within the framework of the band theory of solids. It is remarkable that after more than 80 years, there are still treasures to be uncovered within band theory[9]. The 2D TIs have remarkable similarities to the quantum Hall systems, and we begin with by a short discussion of quantum Hall systems to shed light on frequently used concepts such as edge states and topological order. We continue by introducing the 3D TIs, which are currently at the focus of intense research and have been the subject of our scanning tunneling microscope (STM) studies in the past few years.

A common insulator has an energy gap separating the filled valence bands and the empty conduction bands, and thus is electrically inert because a finite energy is required to dislodge an electron. The electrons of an insulator can be regarded as occupying localized orbitals [ figure 1.1(a) ]. In the 1960s, W. Kohn characterized [10] the insulating state in terms of this insensitivity of electrons inside the material to perturbation effects on the sample boundary. However, the view that insulators have this “nearsighted” property has changed with the discovery of the integer quantum Hall effect in 2D electron gases by von Klitzing in 1980 [11]. In the integer quantum Hall effect [11, 12, 13], an energy gap results from the quantization of the closed circular orbits that electrons follow in a magnetic field. The inside of a quantum Hall system is thus inert like an insulator. But at the boundary of
the material a different type of motion occurs, which allows charge to flow in one-dimensional edge states [figure 1.1(b)].

The Hall conductance as a function of magnetic field applied normal to the gas plane exhibits a staircase sequence of wide plateaus. In quantum Hall systems, the unprecedented precision of the values of the conductance at the plateaus was unanticipated [13]. This precision was utilized for metrological purposes and to determine the fine structure constant with high accuracy ($=\frac{e^2}{\hbar c}$). The successive values of the Hall conductance were integer multiples of a fundamental constant of nature, $\frac{e^2}{\hbar}=1/25812.807572$, irrespective of the geometrical details or the imperfections of its materials. From the works of Thouless et al. [14], among others [15], we can understand the remarkable precision of Hall quantization from a topological perspective. Thouless et al. showed that the Hall conductance is a measure of total curvature in momentum-space, and hence its value is insensitive to the detail of the sample such as size and geometry. The situation is analogous to measuring the total Gaussian curvature of a 3D object, in which according to the Gauss-Bonnet theorem the final result only depends on the "genus" of a solid body—which counts the number of holes in the object, and hence is insensitive to the details [16]. Similarly, in quantum Hall states the conducting edge states are a consequence of the topological structure. These edge states are unique in that they allow for charge to flow in one direction only. This makes them insensitive to scattering from impurities and explains the observed precise quantization of the Hall resistance.

Realization of quantum Hall phases of matter has demanding requirements, such as very low temperatures, strong magnetic fields, and crystal purity. The quest for finding a new material system which shows the desired robust edge states, without any need for magnetic fields and with the promise of functioning up to room temperature has yielded results in the past few years. In 2005, Kane and Mele [17] showed theoretically that in certain insulators, the strong spin-orbit coupling can open an energy gap in the bulk of the material, making the bulk act like an insulator, but with the existence of conducting edge states guaranteed by the topology of the band structure. In 2D topological insulators, the spin-orbit coupling plays an analogous role to the external magnetic field in the quantum Hall systems, leading to the edge states [figure 1.1(c)]. In contrast to quantum Hall systems, in 2D TIs the edge states are spin polarized, and electrons with spin-up propagate in one direction, whereas electrons with spin-down propagate in the opposite direction. As a result they are promising candidates for the transport of spin-polarized currents.

For 2D systems, theoretically, one can distinguish a topological insulator from a normal band insulator by the value of a "$Z_2$ topologically invariant quantum number" $\nu$, which can take one of the two values, say 0 and 1 [17]. It is a topological invariant, which cannot change its value, as one varies
Figure 1.1: **Different kinds of 2D insulators.** (a) Electrons in an insulator are bound in localized orbitals and have an energy gap separating the occupied valence band from the empty conduction band. (b) A two-dimensional quantum Hall state in a strong magnetic field has a bulk energy gap like an insulator but permits electrical conduction in one-dimensional "one way" edge states along the sample boundary. (c) The quantum spin Hall state at zero magnetic field also has a bulk energy gap but allows conduction in spin-filtered edge states. Spin-orbit coupling in this class of insulators plays a similar role as the external magnetic field in quantum Hall systems. Adopted from reference [5].

parameters of the Hamiltonian, except at the point where the energy gap vanishes between occupied and unoccupied states. In 3D systems, the distinction between a TI and conventional insulators is characterized by four \(Z_2\) topological invariants [17]. Calculation of these invariants and determining whether a given system is a TI is becoming simplified for systems with inversion symmetry. For such systems the \(Z_2\) topological invariants can be calculated [18, 19, 20, 21] from the knowledge of the parity of the occupied Bloch wave functions at the time-reversal invariant momenta (TRIM) points in the Brillion zone of the bulk. The TRIM points are points that band structure is degenerate, as required by Kramer’s theorem, and there are eight non-equivalent TRIM points in three dimensions. In chapter 2, when presenting the results of band structure calculations on TIs, we discuss the procedure to calculate these invariants.

### 1.2 2D TI’s theoretical and experimental review

The 2D topological insulators are also known as quantum spin Hall insulators. This state was originally proposed in graphene [22] (Graphene is a 2D form of carbon that is of current interest [23, 24]). It was subsequently predicted to exist [25] and was then observed [26] in HgCdTe quantum well structures. The quantum spin Hall edge states have the important "spin filtered" property that the up spins propagate in one direction, while the down spins propagate in the other. Such edge states were later termed "helical", in analogy with the correlation between spin and momentum of a particle known as helicity. Ordinary conductors, which have up and down spins propagating in
both directions, are fragile because the electronic states are susceptible to Anderson localization (that is, formation of an insulating state as a result of strong disorder [27, 28, 29]) in the presence of disorder. By contrast, the quantum spin Hall edge states are predicted to be immune to localization even for strong disorder [30]. The edge channels for a 2D quantum spin Hall are spin polarized, and it follows that unless the an electron is able to change its spin upon scattering from an impurity, the transmission is perfect in spite of presence of disorders. In the next part, during the discussion of the results on 2D TI’s, we examine the experimental evidence to see how immune these edge states are. If the absence of localization should be anticipated or not for 3D TI’s is a question far from being settled, and in chapter 4, we provide some STM results to be considered in this regard.

The original material system proposed to observe the quantum spin Hall effect was graphene, but the weakness of the spin-orbit coupling in the material, made the proposal impractical. Clearly, a better place to look for this physics would be in materials with strong spin-orbit interactions, made from heavy elements near the bottom of the Periodic Table. In 2006, Bernevig et al. [25] predicted considering quantum well structures of $Hg_{1-x}Cd_xTe$ [figure 1.2(a)]. $Hg_{1-x}Cd_xTe$ is a family of semiconductors with strong spin-orbit interactions. Bernevig et al. considered a semiconductor heterostructure consisting of a thin layer of $HgTe$ sandwiched between crystals of $CdTe$. $HgTe$, $CdTe$, and their alloys are a well-studied family of semiconductor materials, and the proposed device could be made with current technology, thanks to decades of experience in the growth of high-quality semiconductor structures. $CdTe$ has a band structure similar to other semiconductors, but their theoretical analysis shows that in an appropriate range of layer thickness this two-dimensional structure should exhibit a robust quantum spin Hall effect. The conduction-band-edge states have an $s$-like symmetry, while the valence-band-edge states have a $p$-like symmetry. In $HgTe$, the $p$ levels rise above the $s$ levels, leading to an inverted band structure [figure 1.2(b)]. When the thickness of the $HgTe$ layer is $d < d_c = 6.3nm$ the 2D electronic states bound to the quantum well have the normal band order. For $d > d_c$, however, the 2D bands invert. Bernevig et al. showed that the inversion of the bands as a function of increasing $d$ signals a quantum phase transition between the trivial insulator and the quantum spin Hall insulator. This can be understood simply in the approximation that the system has inversion symmetry. In this case, since the $s$ and $p$ states have opposite parity the bands will cross each other at $d_c$. This crossing leads to a change in the parity of the valence-band-edge state and signals a phase transition in which the $Z_2$ invariant $\nu$ changes.

Within a year of the theoretical proposal the Würzburg group, led by Laurens Molenkamp, made the devices and performed transport experiments that showed the first signature of the quantum spin Hall insulator [26]. König et al. measured the low-temperature ballistic edge state transport,
Figure 1.2: Experiments on HgTe/CdTe quantum wells. (a) Quantum well structure. (b) As a function of layer thickness d the 2D quantum well states cross at a band inversion transition. The inverted state is the quantum spin Hall insulator, which has helical edge states. (c) The longitudinal four-terminal resistance, $R_{14,23}$, of various normal (d = 5.5 nm) (I) and inverted (d = 7.3 nm) (II, III, and IV) quantum well structures as a function of the gate voltage measured at T = 30 mK. The device sizes are (20.0 $\times$ 13.3) $\mu$m$^2$ for devices I and II, (1.0 $\times$ 1.0) $\mu$m$^2$ for device III, and (1.0 $\times$ 0.5) $\mu$m$^2$ for device IV. The inset shows $R_{14,23}(V_g)$ of two samples from the same wafer, having the same device size (III) at 30 mK (green) and 1.8 K (black) on a linear scale. Adopted from reference [26].
by measuring the electrical conductance. If the conduction is only taking place through these edge states channels, one expects a quantized conductance of $e^2/h$ associated with each set of edge states. Figure 1.2(c) shows the resistance measurements for a series of samples as a function of a gate voltage which tunes the Fermi energy through the bulk energy gap. Sample I is a narrow quantum well that has a large resistance in the gap. Samples II, III, and IV are wider wells, and are in the "inverted" regime, where symmetry of the bands guarantees the existence of edge states. Samples III and IV exhibit a conductance $2e^2/h$ associated with the top and bottom edges. Samples III and IV have the same length $L=1 \text{ \mu m}$ but different widths $w=0.5$ and 1 $\text{\mu m}$, indicating that transport is at the edge. Sample II ($L=20 \text{ \mu m}$) showed finite temperature scattering effects. These experiments convincingly demonstrate the existence of the edge states of the quantum spin Hall insulator, and subsequent experiments have established the inherently nonlocal electronic transport in the edge states [31].

### 1.3 3D TI’s theoretical and experimental review

In the summer of 2006 three theoretical groups independently discovered that the topological characterization of the quantum spin Hall insulator state has a natural generalization in three dimensions[18, 32, 33]. Moore and Balents coined the term "topological insulator" to describe this electronic phase. Fu et al. established the connection between the bulk topological order and the presence of unique conducting surface states. Soon after, this phase was predicted in several real materials [18], including $Bi_{1-x}Sb_x$ as well as strained HgTe. In 2008, Hsieh et al. [34] reported the experimental discovery of the first 3D topological insulator in $Bi_{1-x}Sb_x$. In 2009 second-generation topological insulators, including $Bi_2Se_3$ and $Bi_2Te_3$ which have numerous desirable properties, were identified experimentally [35, 36, 37].

One of the most appealing physical properties of 3D TI’s is the assertion that the surface states of 3D topological insulators are guaranteed to remain gapless, even in the presence of perturbations or fluctuations of the chemical potential. This expected robustness stems from the topology of the surface bands of TIs. In spite of strong spin-orbit coupling, at high symmetry points of the zone the surface bands remain degenerate, as required by the Kramers’ theorem for time-reversal-symmetric systems [ figure 1.3 ]. These Kramers’ degenerate points therefore form 2D Dirac points in the surface band structure. Away from these special points, the spin-orbit interaction will lift the degeneracy. The interesting question is how these Dirac points connect to each other. There are two topologically distinct ways of connecting pairs such as the $\Gamma_a$ and $\Gamma_b$, as shown in the figure. This determines whether the surface Fermi surface intersects a line joining $\Gamma_a$ to $\Gamma_b$ an even or an
Figure 1.3: **Bulk bands and surface bands.** Surface state dispersion between two Kramers’ degenerate points: in (a), the number of surface states crossing the Fermi energy $E_F$ is even, whereas in (a) it is odd. An odd number of crossings leads to topologically protected metallic surface states: it is guaranteed that there are some states at the chemical potential level ($E_F$), and small perturbations are not going to gap the surface states. The odd crossing is a result of the partner swapping at the high symmetry points of the surface Brillouin zone ($\Gamma_a$ and $\Gamma_a$). The symmetry of the occupied bands determine if the partner swapping is going to take place or not. Due to strong spin orbit coupling the surface states are spin polarized, while if there is inversion symmetry, which is the case for the samples presented in this thesis, the bulk bands are spin degenerate. Adopted from reference [4].

odd number of times. Which of these two alternatives occurs is determined by the symmetry of the occupied bulk bands, which determines the $Z_2$ invariants. If the number of crossings is odd, then the surface states are topologically protected in the following sense: there is always some surface state at the chemical potential level, regardless of the fluctuations in the chemical potential or changes in the band structure, as long as the band modifications are not large to close the gap or change the bands topology.

Given the symmetry of the Brillion zone, determining an even or odd crossing between two high symmetry points of the surface Brillouin zone, is the same as determining whether an even or an odd number of Fermi contours are enclosing each Kramers’ point. In a strong topological insulator the surface Fermi contours encloses an odd number of Kramers degenerate Dirac points. As a result of this odd number of contours, an unusual planar metal is formed at the surface of topological insulators. Each momentum along the surface has only a single spin state at the Fermi level, and the spin direction rotates as the momentum moves around the Fermi surface [figure 1.4(a)]. Therefore, the surface states of a strong topological insulator form a metal that in a sense is essentially half an ordinary metal [38]. As presented schematically in figure 1.4(b), in an ordinary metal, which has
up and down spins at every point on the Fermi contours, the surface states are not spin degenerate. This difference has far-reaching consequences, including the possibility of generating new particles that may have applications in quantum computing.

This unique spin texture for the surface states leads to a nontrivial Berry phase [39, 40], which is the phase acquired by an electron going around the Fermi contour. Time reversal symmetry requires that this phase to be 0 or $\pi$. When an electron circles a Dirac point, its spin rotates by $2\pi$, which leads to a $\pi$ Berry phase. The Berry phase has important consequences for the behavior in a magnetic field and for the effects of disorder. In particular, in an ordinary 2D electron gas the electrical conductivity decreases with decreasing temperature, reflecting the tendency toward the Anderson localization in the presence of disorder. The $\pi$ Berry phase changes the sign of the weak localization correction to the conductivity leading to weak anti-localization [41]. Furthermore, theoretical works of Nomura et al. [30] suggest that the electrons at the surface of a strong topological insulator cannot be localized even for strong disorder. This protection of the surface metal from Anderson localization is one of the key differences between the surface of the topological insulator and the 'accidental' surface states present in other materials, such as the noble metals [figure 1.4(c)]. These theoretical works still await robust experimental verification. If the claim is verified, then the situation is similar to the edge states of the quantum spin Hall insulator; however, the electron motion on the surface is diffusive rather than ballistic.

The majority of the notable experimental studies on 3D TI’s have been done either by photoemission spectroscopy or by scanning tunneling microscope, and we present them in some length in future chapters. The difficulty of doing other measurements is rooted in the fact that native defects in the bulk of TI’s, which come as result of the chemical growth technique, are acting as charge dopants and shift the chemical potential such that it is moved away from the band gap. To study surface transport properties, one has to shift the Fermi level into the band gap by chemical dopants [42, 43]. This approach seems to have other undesired consequences, as we discuss in chapter 4. It is also needed to increase the surface-to-volume ratio [44], to suppress the bulk related effects. For this reasons, several experimental groups are switching to molecular beam epitaxy (MBE) grown samples, and some are using very thin samples, on the order of couple of tens of nm, to reduce the bulk contribution to the conduction. In spite of these challenges several experimental groups [41, 42, 45, 46] succeeded in discerning the surface contribution in a transport measurement. For instance, Qu et. al. [46] showed that even for very thin samples and with proper tuning of the chemical potential by doping away from bulk bands, the contribution of the bulk to conductivity is still $\sim 300$ times more than the surface states. In spite of this, they observed quantum oscillation due
Figure 1.4: **Schematic of various contours of constant energy.** The blue circles represent the contours of constant energy or Fermi contours of the surface states. The arrows show the spin and the hexagons represent the First Brillouin zone, schematically. (a) As a result of strong spin-orbit coupling the surface states of TI’s are spin-polarized. Since time reversal symmetry requires that states at momenta $\vec{k}$ and $-\vec{k}$ have opposite spin, the spin must rotate with $\vec{k}$ around the Fermi surface. As a result of the symmetry of the occupied bands, they have a non-trivial Berry phase. (b) In the absence of spin-orbit coupling the surface bands, such as that of the Shockley surface states on the surface of noble metals, are not spin polarized. (c) The presence of spin-orbit coupling could lift this degeneracy, which is the case for the surface states of gold. However, such surface states do not have non-trivial Berry phase and the novel properties predicted for TIs are not expected to be seen in them.

to surface states and from that extract transport related parameters, such as mobility and mean free path of the surface states. While resolve the conductance of the surface states from the dominant bulk contribution still constitute a challenge for some of the measurements, STM’s surface sensitivity became a clear advantage. The tunneling of the electrons from the STM tip is predominantly into the surface states, as no consideration of tunneling to the bulk is required to explain the observed intricate quasiparticle interference patterns due to scattering of surface states from dopants.

The first topological insulator to be discovered was the alloy $\text{Bi}_{1-x}\text{Sb}_x$ [34], the unusual surface bands of which were mapped in an angle-resolved photoemission spectroscopy (ARPES) experiment [34, 47]. In ARPES experiments, a high-energy photon is used to eject an electron from a crystal, and then the surface or bulk electronic structure is determined from an analysis of the momentum of the emitted electron. The surface structure of this alloy was found to be complex, and as a result, a search for other topological insulators was lunched soon after. For a topological insulator to form, spin-orbit coupling must be strong enough to modify the electronic structure significantly, which suggests that heavy-element, small-bandgap semiconductors are the most promising candidates. This suggestion stems from two points. First, spin-orbit coupling is a relativistic effect and is only strong for heavy elements. Second, if the bandgap is much larger than the energy scale of spin-orbit coupling, then spin-orbit coupling will not be able to change the phase. The search for topological
insulators culminated in the recent discovery of topological insulator behavior in $Bi_2Se_3$ and $Bi_2Te_3$ [37, 35, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57]. These 'next-generation' materials have both the promise of showing topological insulator behavior up to higher temperatures than does the original material ($Bi_1-xSb_x$), with bulk bandgaps of more than 0.1 eV, and have the simplest surface state that is allowed. Beyond providing further confirmation of the theory of topological insulators, the simplicity of the surface state in these materials opens up the possibility of many experiments. Furthermore, the existence of large bandgap implies that these experiments do not need to be carried out at extremely low temperatures. As mentioned before, the main remaining complication about these materials, especially when using experimental techniques that do not distinguish directly between bulk states and surface states, is that in the bulk state they have residual conductivity arising from impurities.

1.4 Outlook

The consequences of discovering a new ordered phase of matter are far-reaching. Generally, in many familiar ordered phases such as crystalline or ferromagnetic phase, the ordered phases are described through the symmetry that the emergent phases break: in a crystal, ions are arranged periodically, thereby breaking the continuous symmetry of space under rotations and translations; in ferromagnets, the rotational symmetry of spin space is broken, together with the time-reversal symmetry. The theoretical prediction and experimental verification of the existence of a topological order phase occurring in 3D materials constitutes one of the important discoveries of the past few years in theoretical condensed matter physics. Much research interest has been attracted to explore the exotic metallic states present at the surfaces of this novel class of matter, resulting from its topological order.

For instance, it has been theoretically predicted that the collective motions of electrons inside topological insulators will mimic several of the never-before-seen particles predicted by high-energy physicists, such as Majorana particles and magnetic monopoles [58, 59]. Majorana fermions are massless, chargeless entities that can serve as their own antiparticles. There is still no conclusive evidence for Majorana fermions in nature and the several proposals to observe them in topological insulators are the heart of a fast growing research [60, 61, 62, 63]. When a topological insulator is placed in contact to a superconductor, like any other metal, the metallic surface of a topological insulator becomes superconducting, by way of the proximity effect. However the novel spin texture of the surface states leads to a novel superconducting surface state: if a vortex line runs from the
superconductor into the topological insulator, then a zero-energy Majorana fermion is trapped in the vicinity of the vortex core [60]. The Majorana fermions are predicted to obey a special kind of quantum statistics that is non-Abelian, which makes them ideal for quantum computation applications. Experimental realizations of Majorana fermions and other exotic states require heterostructures involving topological insulators and other materials. In this direction, there are many material-related issues needs to be addressed in order to find appropriate magnetic and superconducting materials which exhibit the appropriate proximity effects with the surface states, and detailed experiments will be necessary to characterize those states.

The advances in the physics of topological insulators have been driven by a rich interplay between theoretical insight and experimental discoveries. There is reason for optimism that this field will continue to develop in exciting new directions. The field of topological insulators is at an early stage in its development. There is much work to be done to realize the potential of these new and fascinating materials. In this dissertation, we discuss the basic properties of topological insulators which have been established as result of our experimental works and the works of others.
Chapter 2

Absence of backscattering due to helical spin texture

The flow of electric current through a metallic wire is inevitably degraded by the wire’s resistance. At low temperatures, the resistivity of a metal is dominated by the scattering of electrons off impurities and imperfections in the metal’s crystal lattice. When an electron collides with an impurity, it scatters in a random direction, and this process diminishes its contribution to the overall current. Backscattering processes, in which a collision causes the electron to make a U-turn, are especially effective in degrading the current flow. A key predicted feature for the metallic surface states of topological insulators is their insensitivity to spin-independent scattering, which is thought to protect them from backscattering and consequently localization [see figure 2.1]. Using STM, which has been proven successful in the study of surface states, we have probed the sensitivity of these helical states to scattering from disorder [64]. The random alloying in the topological insulator $Bi_{1-x}Sb_x$ makes this material system an ideal candidate to examine the impact of disorder on topological surface states. Angle-resolved photoemission spectroscopy (ARPES) experiments on the (111) surface of $Bi_{1-x}Sb_x$ crystals have been identified surface states within the bulk band gap of these compounds. The shape of the Fermi surface for these states shows an odd number of band crossings between time-reversal invariant momentum points in the first Brillion zone (FBZ) at the Fermi energy, which confirms the identification of $Bi_{1-x}Sb_x$ as a topological insulator. Furthermore, spin-sensitive experiments established that these surface states have a helical spin structure. In this chapter, we discuss our STM results on this topological insulator material system which shows that despite strong atomic scale disorder, backscattering between states of opposite momentum and
opposite spin is absent. This chapter is mainly adopted from our published results in reference [64].

2.1 The band structure of $\text{Bi}_{1-x}\text{Sb}_x$ and ARPES results

The first 3D topological insulator to be identified experimentally was the semiconducting alloy $\text{Bi}_{1-x}\text{Sb}_x$, whose unusual surface bands were mapped in an ARPES experiment by a Princeton University group led by Hasan [34]. In this section we discuss the electronic band structure of Bi, and how it modifies by alloying it with Sb, and review the ARPES measurements on this alloy.

2.1.1 Band structure $\text{Bi}$ and $\text{Bi}_{1-x}\text{Sb}_x$

Both Bi and Sb have rhombohedral $A7$ crystal structure [66], and are group V semimetals with strong spin-orbit interactions and also possess inversion symmetry. Bismuth antimony alloys have long been studied for their thermoelectric properties [67, 68]. Pure bismuth is a semimetal. The band structure of Bismuth, shown schematically in figure 2.2(a), features conduction and valence bands that overlap, leading to three ellipsoid hole pockets located at the time-reversal invariant momenta (TRIM) points in the Brillouin zone and pockets of electrons near the three equivalent L points. Elemental bismuth owes many of its peculiar physics [69] to the size of these pockets, which are tiny and occupy only $10^{-5}$ of the Brillion zone. The valence and conduction bands at the L point, derived from anti-symmetric $L_a$ and symmetric $L_a$ orbitals, have a small energy gap.

Substituting bismuth with antimony changes the band structure in a topologically non-trivial way. For $\text{Bi}_{0.96}\text{Sb}_{0.04}$, the gap at the L points closes and reopens for $x>0.04$ with the parity of
Figure 2.2: The electronic band structure of $Bi_{1-x}Sb_x$. (a), (b), and (c) Schematic representation of the band structure of $Bi_{1-x}Sb_x$, which evolves from semi-metallic behavior for $x=0.07$ to semiconducting behavior for $0.07<x<0.22$ and back to semi-metallic behavior for $x$'s larger than that. The symmetry of the bands at L point of the bulk Brillouin zone change at $x=0.04$. (d) The resistivity measurement shows a finite resistivity, and hence metallic behavior for $Bi$, and a divergence in resistivity at low temperatures, and hence insulating behavior for $Bi_{0.9}Sb_{0.1}$. Panels are adopted from reference [70] and [34].

the eigenstates at the L points switched [ figure 2.2(b) ]. This gives the required sign change to the product of the parity eigenvalues at the bulk TRIM points such that the alloy can be a topological insulator; however the bulk spectrum is not gaped at this concentration of Sb [70]. By increasing $x$, the energy of the hole pocket at T lowers, and $Bi_{1-x}Sb_x$ turns into an indirect band gap semiconductor for $x>0.07$, and for $0.08<x<0.22$ the alloy becomes a direct band gap insulator, which is also a TI. As $x$ is increased further, the conduction and valence bands remain separated, and for $x=0.22$ the valence band at a different point rises above the conduction band, restoring the semi-metallic state. For values of $x$ up to $x=1$, i.e. Sb, the bulk bands remain inverted, and the surface states show an odd crossing and hence protection, but the $Bi_{1-x}Sb_x$ alloy remains a semi-metal [ figure 2.2(c) ].

In 3D systems, the distinction between TI and conventional insulators is characterized by four $Z_2$ topological invariants [17]. Calculating these invariants and determining whether a given system is a TI becomes simpler for systems with inversion symmetry. For such systems the $Z_2$ topological invariants can be calculated [71] from the knowledge of the parity of the occupied Bloch wave functions at the TRIM points in the Brillion zone of the bulk. The TRIM points are points at which the band structure is degenerate, as required by Kramer’s theorem, and there are eight non-equivalent TRIM points in three dimensions. The four $Z_2$ invariants also determine if the number of surface bands crossing between any two surface TRIM points of the surface Brillion zone is even or odd. For TI the surface bands switch partners at the surface TRIM points and the crossing number is odd, while for normal bulk insulators the crossing number is even. Since pure bismuth and pure antimony both have a finite direct band gap, their valence bands can be topologically...
Figure 2.3: The parity of the high symmetry points of the bands for Bi and Sb. For crystal with inversion symmetry, at the high symmetry points \( \Lambda_a \) of the bands, the Bloch states \( u_{\Lambda_a}(\Lambda_a) \) are also parity eigenstates with eigenvalues \( \xi_m(\Lambda_a) = \pm 1 \). The table shows the symmetry labels for the Bloch states at the 8 time invariant momenta \( \Lambda_a \) for the five valence bands of Bi and Sb. Based on the parities at the \( \Lambda_a \)'s, the \( \delta_a \)'s are calculated by \( \delta_a = \prod_m \xi_m(\Lambda_a) \), where the product is over the Kramers pairs of occupied bands. \( \delta_a \)'s determine the topological class \((\nu_0; \nu_1, \nu_2, \nu_3)\) by relations \((-1)^\nu = \prod_{a=1}^4 \delta_a \). The difference between Bi and Sb is due to the inversion of the \( L_s \) and \( L_a \) bands that occurs at \( x=0.04 \). Adopted from reference [71].

classified, and their inversion symmetry simplifies the calculation. Table 2.1 shows the symmetry labels that specify the parity of the Bloch states for the occupied bands at the 8 TRIM points in the bulk Brillouin zone. From this information one can deduce that bismuth is in the trivial class while antimony is in the non-trivial class, i.e. it possesses topologically protected surface states. Since the semiconducting alloy is on the antimony side of the band inversion transition, it is predicted that it is in the same class as antimony.

### 2.1.2 ARPES measurements

ARPES is one of the direct methods of studying the electronic structure of the surface of solids. It has been established as an important method to study the electronic structure of molecules, solids and surfaces [72]. ARPES works based on using the photoelectric effect [73]. A monochromatic beam of light, typically from a synchrotron radiation source, impinges on a sample and photo-excites electrons into the vacuum. These photoelectrons are then collected in an analyzer that measures their kinetic energy as a function of emission angle relative to the sample surface. By exploiting energy and momentum conservation, and the knowledge of the energy of the incident beam and the work function of the sample, it is straightforward to relate the measured kinetic energy of the photoelectron to its binding energy, as well as to its in plane crystal momentum while inside the sample. Provided the photoemission process is spin-conserving, the spin of the initial state of an electron in a solid can be determined by measuring its spin after it has been photo-emitted. To resolve the electrons spin, Mott electron polarimetry [74] is used. In this method, the electrons of different spin from a photo-emitted beam are separated based on the use of spin-orbit (Mott)
Figure 2.4: **ARPES measurements on Bi$_{1-x}$Sb$_x$.** (a) A schematic of spin-ARPES measurement setup that was used to measure the spin distribution on the (111) surface Fermi surface of Bi$_{0.9}$Sb$_{0.1}$. (b) A schematic of the 3D Brillouin zone of Bi$_{1-x}$Sb$_x$, and its (111) surface projection. (c) ARPES data on the (111) surface of Bi$_{0.9}$Sb$_{0.1}$ which probes the occupied surface states as a function of momentum on the line connecting the high symmetry points $\bar{\Gamma}$ and $\bar{M}$ in the surface Brillouin zone. The surface bands cross the Fermi energy five times, which establishes that the semiconducting alloy of Bi$_{0.9}$Sb$_{0.1}$ has topologically protected surface states. (d) The Fermi contours of the surface states, and the resolved spin texture of the surface bands. Spin orientations on the surface create a vortex-like pattern around the $\bar{\Gamma}$ point. Adopted from reference [34].

scattering of electrons from a heavy nucleus [figure 2.2(a)]. Since the Mott-scattering process involves accelerating the electrons to $10^4 \sim 10^5$ eV and then having them scattered by a gold target, the photoelectrons will lose information about their original energy and momentum. Thus, prior to the spin-detection, the energy/momentum of the photoelectrons needs to be resolved by a regular dispersive spherical analyzer. Then, photoelectrons of known momentum and energy enter the spin detector and are accelerated for spin detection. In this way, the complete energy/momentum/spin information of the photoelectrons can be acquired, and the spin-resolved electronic structures of a sample can be determined.

ARPES measurements offer several advantages in comparison to other experimental approaches. Charge transport experiments, which were successful for identifying the 2D topological insulators [26], are problematic in 3D materials. The difficulty is due to the fact that separating the surface
contribution to the conductivity from that of the bulk is extremely hard. On the other hand, the ARPES measurements allows for a clear isolation of surface states from that of the bulk 3D band structure because surface states do not disperse along a direction perpendicular to the surface whereas the bulk states do. Moreover, unlike in a transport experiment, ARPES carried out in a spin resolution mode can, in addition, measure the distribution of spin orientations on the Fermi surface.

Experiments by Hsieh et al. [34] probed both the bulk and surface electronic structures of $Bi_{0.91}Sb_{0.09}$ with ARPES. Figure 2.4(c) shows the ARPES spectrum, which can be interpreted as a map of the energy of the occupied electronic states as a function of momentum along the line connecting $\bar{\Gamma}$ to $\bar{M}$ in the projected surface Brillouin zone. The experiments observed several surface states that span the bulk gap. The observed surface bands cross the Fermi energy five times between $\bar{\Gamma}$ and $\bar{M}$, and this odd number of crossings is analogous to figure 1.4(a) and indicates that these surface states are topologically protected. As discussed in chapter one, Kramers’ theorem requires surface states to be doubly degenerate at the high symmetry points of the surface Brillouin zone $\bar{\Gamma}$ and each of the three equivalent $\bar{M}$ points. Such a Kramers point is indeed observed at $\bar{M}$ approximately $15\pm5$ meV below $E_F$. As expected for a system with strong spin-orbit interactions and the degeneracy is lifted away from $\bar{M}$.

A distinguishing feature of topological insulator surface states is the intimate correlation between spin and momentum they exhibit, which underlies the $\pi$ Berry phase associated with the Fermi surface. Spin resolved ARPES, [figure 2.4(d)] measured the spin polarization of the surface states [47]. The experiment proved that the surface states are indeed non-degenerate and strongly spin polarized, providing even more decisive evidence for their topological classification. In addition, the spin-polarization data also established the connectivity of the surface state bands above $E_F$ which is inaccessible to ARPES, showing that bands labeled 2 and 3 in figure 2.4(c) connect to form a hole pocket.

2.2 $Bi_{1-x}Sb_x$ samples and basic STM results

The $Bi_{0.92}Sb_{0.08}$ single crystals were grown by melting stoichiometric mixtures of elemental Bi (99.999%) and Sb (99.999%) in 4 mm inner diameter quartz tubes from a stoichiometric mixture of high purity elements. The samples were cooled over a period of two days, from 650 to 260 °C, and then annealed for a week at 260 °C. The obtained single crystals were confirmed to be single phase and identified as having the rhombohedral $A7$ crystal structure by X-ray power diffraction using a
Figure 2.5: STM topography, and dI/dV spectroscopy of the $B_{0.92}Sb_{0.08}(111)$ surface. 
(a) STM topograph (+50meV, 100pA) of the $B_{0.92}Sb_{0.08}(111)$ surface over an 800 Å by 800 Å area. 
(b) Spatial variation of the differential conductance (dI/dV) measurements across a line of length 250 Å. A typical differential conductance measurement over larger energy ranges is shown in the inset. Adopted from reference [64].
We performed our experiments using a home-built cryogenic STM that operates at 4 K in ultra-high vacuum. A brief discussion of STM and principle and techniques is presented in the appendix A. With our STM, we have examined several crystals of $Bi_{0.92}Sb_{0.08}$, grown under the same conditions, and we have not noticed any sample dependence for any of the results we are presenting. The typical size of the crystals we used was $1 \text{ mm} \times 1 \text{ mm} \times 0.3 \text{ mm}$ [appendix A]. Samples were cleaved in situ at room temperature in ultrahigh vacuum before STM experiments at low temperatures. The weak bonding between atomic layers in this crystal makes the (111) surface the natural cleavage plane. A mechanically sharpened Pt-Ir alloy wire was used as an STM tip, and the quality of the tip apex was examined by scanning an atomically clean Ag(111) surface.

The topographic images of the sample are dominated by long wavelength ($\sim 20 \text{ Å}$) modulations in the local density of states [figure 2.5(a)]. This is in sharp contrast to topographic measurements on other semiconductors, where individual atoms and atomic rows dominate the topography [for instance, see figure A.4(d) in appendix A]. This topographic difference indicates the existence of surface states on the surface of the insulating $Bi_{0.92}Sb_{0.08}$ crystal, and demonstrates the high sensitivity of the tunneling to the presence of these states.

Spectroscopic measurements show a general suppression of the density of states near the Fermi level, with spectra appearing for the most part homogeneously across the sample surface [figure 2.5(b)]. The presence of surface states makes the spectrum different from that seen on the surface of ordinary insulators [see figure A.4(e) in appendix A]. In normal band insulators, the density of states becomes zero for the energies corresponding to the semi-conductor gap, while here the LDOS remains finite, as a result of the contribution of the surface states to the LDOS, for the energies inside the gap. ARPES measurements [34] and recent band structure calculations [70] suggest that within $\pm 30\text{meV}$ of the Fermi level, where there is a bulk gap, tunneling should be dominated by the surface states. However, tunneling spectroscopy measurements do not distinguish between bulk and surface states, and a different approach is needed to solely probe the surface states.

2.3 Quasiparticle interference (QPI’s) patterns

While tunneling spectroscopy measurements do not distinguish between bulk and surface states, energy-resolved spectroscopic maps in figure 2.6(a), (b) and (c) display modulations that are the result of scattering of the surface electronic states [for details of the measurement see appendix A]. As expected for the scattering and interference of surface states, the observed patterns are not
Figure 2.6: The dI/dV maps, QPI patterns, and ARPES measurements on Bi$_{0.92}$Sb$_{0.08}$ (111) surface. (a), (b), and (c) Spatially resolved conductance maps of the Bi$_{0.92}$Sb$_{0.08}$ (111) surface obtained at -20 mV, 0 mV, and +20 mV (1000 Å × 1300 Å). In the upper right corner of each map the Fourier transform of the dI/dV maps are presented. The hexagons have the same size as the FBZ. The Fourier transforms have been symmetrized in consideration of the three-fold rotation symmetry of the (111) surface. (d) and (e) ARPES intensity map of the surface state at -20mV and at the Fermi level, respectively. (f) The spin textures from ARPES measurements are shown with arrows, and high symmetry points are marked (Γ and 3 M). Adopted from reference [64].
commensurate with the underlying atomic structure. While we do not have direct information on the identity of the scattering defects, the random distribution of substituted Sb atoms is a likely candidate. The Fourier transform of the scanning tunneling spectroscopy (FT-STS) maps shown as insets in figure 2.6(a), (b) and (c) for $Bi_{0.92}Sb_{0.08}$ display rich patterns, which have the six-fold rotational symmetry of the underlying lattice, and evolve as a function of energy. These patterns display the allowed wavevectors $\vec{q}$ and the relative intensities for the various scattering processes experienced by the surface state electrons[65, 75].

In a crystal, the eigenstates are Bloch wavefunctions characterized by wavevector $\vec{k}$ and energy $\epsilon$, and their dispersion relation, $\epsilon(\vec{k})$, can be measured with ARPES. By contrast, real space imaging techniques, such as STM, cannot be used to measure $\epsilon(\vec{k})$. This is because the local-density-of-states LDOS($E$) spectrum at a single location $\vec{r}$ is related to the k-space eigenstates $\psi_k(\vec{r})$ by

$$LDOS(E, \vec{r}) \propto \sum_k |\psi_k(\vec{r})|^2 \delta(E - \epsilon(\vec{k}))$$ (2.1)

and substitution of a Bloch wavefunction into the above equation shows the LDOS to be spatially uniform. However, when sources of disorder such as impurities or crystal defects are present, elastic scattering mixes eigenstates with different $\vec{k}$ that are located on the same quasiparticle contour of constant energy (CCE) [76]. When scattering mixes states $\vec{k}_1$ and $\vec{k}_2$, an interference pattern with wavevector $\vec{q} = \vec{k}_2 - \vec{k}_1$ appears in the form of the quasiparticle wavefunction and spatial modulation of the LDOS with wavelength $\lambda = 2\pi/|\vec{q}|$ appear [see figure 2.7]. The measurements presented in figure 2.6 can be understood in terms of this quasi-particle interference (QPI). STM studies of such QPI patterns have allowed the first direct probes of the quantum interference of electronic eigenstates in metals and semiconductors [76, 77, 78, 79].

Within a simple model of QPI, the interference wavevectors connect regions of high density of states on contours of constant energy (or the Fermi surface at the chemical potential). Therefore the QPI patterns should correspond to a joint density of states (JDOS) for the surface state electrons that can be independently determined from ARPES measurements. Figure 2.6(d) and (e) show contours of constant energy (CCE) in the first Brillouin zone, as measured with ARPES at two energies on $Bi_{0.92}Sb_{0.08}$ crystals. The CCE consist of an electron pocket centered on the $\bar{\Gamma}$ point, hole pockets half way to the $\bar{M}$ point, and two electron pockets that occur very close to the point. From these measurements, we can determine the JDOS as a function of the momentum difference between initial and final scattering states, $\vec{q}$, using,
Figure 2.7: **Schematic demonstration of QPI.** Studying surface states with STM is relied on their scatterings. (a) In the absence of any scattering centers, there is no interference pattern, as can be seen in (b). In this case there is no spatial pattern to the tunneling current. (c) The scattering of surface states from defects, leads to an interference pattern (d). (e) The Fourier transform of the interference patterns leads to a map of all scattering wavevectors (q’s) and their intensities. (f) The q’s are related to initial \( \vec{k}_i \) and final states \( \vec{k}_f \) of scattering by a simple relation, \( \vec{q} = \vec{k}_f - \vec{k}_i \). The blue curves represent a contour of constant energy.

\[
J \text{DOS}(\vec{q}) = \int I(\vec{k})I(\vec{k} + \vec{q})d^2\vec{k} \tag{2.2}
\]

where \( I(\vec{k}) \) is the ARPES intensity that is proportional to the surface states’ density of states at a specific two-dimensional momentum \( \vec{k} \). Figure 2.8(a) and (d) shows the results of computation of the JDOS from ARPES data for two different energies. Contrasting these figures to the corresponding QPI data in figure 2.8(b) and (e), we find a significant suppression of the scattering intensity along the directions equivalent to \( \bar{Γ} - \bar{M} \) in the first Brillouin zone. Backscattering between various electron and hole pockets around the point should give rise to a continuous range of scattering wavevectors along the \( \bar{Γ} - \bar{M} \) direction, a behaviour not observed in the data [see also the expanded view of the JDOS and QPI in figure 2.9(a)]. This discrepancy suggests the potential importance of the surface states’ spin texture and the possibility that spin rules are limiting the backscattering for these chiral electronic states.

To understand scattering and interference for these spin-polarized states, we determine the spin-dependent scattering probability,
Figure 2.8: Construction of joint density of states (JDOS) and spin scattering probability (SSP) from ARPES data and their comparison with FT-STS. (a) The JDOS and SSP calculated at $E_F$, from ARPES data presented in figure 2.5(e). (b) The FT-STS at EF. (c) The SSP calculated at $E_F$. (d) The JDOS calculated at -20mV, from ARPES data presented in figure 2.5(d). (e) The FT-STS at -20mV. (f) The SSP calculated at -20mV. (g) The schematization of the features associated with scattering wavevectors $\vec{q}_1$ to $\vec{q}_8$ in the FT-STS data. Adopted from reference [64].
\[ SSP(\vec{q}) = \int I(\vec{k}) T(\vec{q}, \vec{k}) I(\vec{k} + \vec{q}) d^2 \vec{k}, \]  

(2.3)

which in similar fashion to the JDOS uses the ARPES-measured density of states, \( I(\vec{k}) \), but also includes a spin-dependent scattering matrix element \( T(\vec{q}, \vec{k}) \). This matrix element describes the scattering probability as a function of momentum transfer and spins of states that are connected by the scattering process. Shown in figure 2.8(c) and (f) are the calculated \( SSP(\vec{q}) \) from ARPES data at two different energies using a matrix element of the form \( T(\vec{q}, \vec{k}) = |\langle S(\vec{k}) | S(\vec{k} + \vec{q}) \rangle|^2 \). This simple form of spin-selective scattering reduces scattering between states with non-aligned spins and completely suppresses scattering between states with opposite spin orientations. Comparison of the \( SSP \) patterns to the QPI measurements in figure 2.8 shows that including spin effects leads to remarkably good agreement between the scattering wavevectors measured by STM and those expected from the shape of the surface CCE as measured by ARPES. Features in the FT-STS and \( SSP \) at different wavevectors are categorized and given labels in figure 2.8(g). A quantitative comparison between the QPI from the STM data and JDOS and \( SSP \) from ARPES data can be made by computing the cross-correlation between the various patterns. Focusing on the high-symmetry direction, which is shown in figure 2.9(a), we find that the QPI (excluding the central \( q=0 \) section, which is dominated by the disorder) is 95% correlated with the \( SSP \) in the same region. The cross-correlation is found to be 83% between the QPI and JDOS. Therefore, the proposed form of the spin-dependent scattering matrix element is the critical component for understanding the suppression of scattering along the high-symmetry directions in the data.

The proposed scattering matrix elements \( T(\vec{q}, \vec{k}) \) and associated spin-scattering rules are further confirmed by a more comprehensive analysis of the QPI patterns. An example of such an analysis is shown in figure 2.9, in which we associate features along the high-symmetry direction in the QPI and \( SSP \) with specific scattering wavevectors \( \vec{q} \) that connect various regions of the CCE. The observed wavevectors in QPI and \( SSP \) obey spin rules imposed by \( T(\vec{q}, \vec{k}) \), as illustrated schematically in figure 2.9(b). In this panel, we also depict examples of scattering processes that, while allowed by the band structure and observed in JDOS, violate the spin scattering rules and are not seen in QPI data in figure 2.9(a). We were able to perform a comprehensive analysis of all the features in the QPI data [figure 2.10], and demonstrate that the allowed set of scattering wavevectors \( \vec{q}_1 \) to \( \vec{q}_8 \) [figure 2.8(g)] exclude those that connect states with opposite spin. Hence, spin selection rules are an essential ingredient in understanding the observed patterns in q-momentum space.

In figure 2.11, we show dispersion as a function of energy for some of the wavevectors \( q \) in the QPI
and compare their energy dispersion to that expected from ARPES results in the SSP. Remarkably, all the features of the complex QPI patterns and their energy dependence can be understood in detail by the allowed scattering wavevectors based on the band structure of the topological surface states and the spin scattering rule. This agreement provides a precise demonstration that scattering of electrons obeys the spin scattering rules and associated suppression of backscattering.

2.4 Conclusion

Other surface states with strong spin-orbit interaction may be expected to show evidence for spin-selective scattering; however, because spin states come in pairs, the QPI patterns can rarely probe these rules [80]. In some situations there is evidence of such rules [81, 82], but the precision with which scattering of surface states for $Bi_{0.92}Sb_{0.08}$ can be understood using spin-selective scattering is unprecedented. Unusual scattering of chiral electronic states is also seen in monolayer graphene, where the underlying two-atom basis leads to a pseudo-spin index for quasi-particles and results in suppression of intravalley scattering [83, 84]. The key difference expected for surface states of a topological insulator is the degree to which they can tolerate disorder. This aspect is clearly demonstrated here for surface states of $Bi_{0.92}Sb_{0.08}$, where strong alloying causes scattering for the surface state electrons yet the spin-selection scattering rules are strictly obeyed over length scales...
Figure 2.10: Decomposition of the various parts of the QPI pattern at the Fermi level.  
(a.I) The Fermi level QPI measured with STM. The individual features are highlighted in ((a.II)) to ((a.V)). (b.I) The SSP calculation of the QPI pattern from ARPES intensity maps at Fermi level, and its decomposition into various constituent parts is shown in (b.II) to (b.V). (c.I) The schematization of the various features seen in the FT-STS data. ((c.II)) to (c.V) Various parts of the Fermi contours measured by ARPES, with arrows showing the sources of the scatterings seen in STM data. Columns II to V have the following order: in (c), we show a specific part of the Fermi surface, and in (b) the SSP based of that part is presented. In (a), the corresponding parts of the pattern which are visible in STM data are in color and the rest are shown in gray. In the legend, the ARPES intensity map and its spin texture at the Fermi level is shown. Adopted from reference [64].
Figure 2.11: Dispersion of various peaks from FT-STS and ARPES. (a) The intensity of the FT-STS maps along the Γ-M direction for various energies. The two peak positions correspond to \( q_1 \) and \( q_2 \), which become larger with increasing in energy. Each curve is shifted by 0.6 pS for clarity. (b) Dispersion of the position of \( \vec{q}_1 \), \( \vec{q}_2 \), and \( \vec{q}_3 \) from ARPES (open symbols) and STM (solid symbols). The data were obtained from fitting the peak in the intensity of the QPI patterns measured in STM, and calculated from the ARPES CCE. Each STM data point is the averaged value of six independent measurements, and the error bar represents one standard deviation. The systematic error was negligible. Adopted from reference [64].
much longer than that set by the atomic scale disorder. Future experiments with magnetic scattering centers could further probe the spin scattering rules for topological surface states and may provide the setting for the manipulation of these spin-polarized states in device applications. We discuss these matters in chapter 4.

The observation that the helical spin texture leads to the absence of backscattering, could perhaps lead to yet more remarkable property: topological-insulator surfaces could be exceptions to the rule that all planar metals become insulators at low temperature in the presence of impurities or defects. The wave nature of electrons in quantum mechanics tends to amplify the effects of impurity scattering: an electron bouncing off many impurities often becomes localized, even when a classical particle would continue to diffuse [27, 85]. The theory of localization famously predicts that at sufficiently low temperature, and beyond a critical amount of impurity scattering, metals will become insulators and the diffusion of electrons that characterizes the material’s conductivity will come to a halt. In fact, the scaling theory of localization shows that even small amount of disorder is enough to localize the states in 2D and 1D samples [86]. It has been theoretically predicted by Numura et al. [30] that absence of backscattering is such a major change to allowed scatterings that it leads to immunity to localization. Absence of localization, if it can be experimentally verified, gives these material systems great advantages for spintronic [87] applications.

Note that we use the word backscattering to refer to scattering between state of equal and opposite momenta in k-space, which are always in orthogonal spin states in topological insulators. However, as $\vec{q}_2$, and $\vec{q}_4$ in our results suggest, changing the direction $\vec{k}$ by 180° is possible upon scattering, as long as the magnitude of the $\vec{k}_i$ and $\vec{k}_f$ are not the same. There is no experimental or theoretical desire to study if the absence of scattering between equal and opposite spin states (which we refer to as backscattering), could lead to absence of localization for the complicated CCE of $Bi_{1-x}Sb_x$. However, if the surface bands are consistent of only a single contour, as is the case for $Bi_2Se_3$ and $Bi_2Te_3$, then the theory suggests that absence of backscattering is leading to immunity to localization. We present our STM studies of these compounds in chapter 4.
Chapter 3

Transmission of topological surface states through surface barriers

As discussed in previous chapters, two features of topological surface states alter their response to crystalline imperfections: first the elimination of backscattering owing to scattering restriction imposed by their spin texture, and second the odd number of band crossings, which prevents the surface states from being fully gapped when perturbed. The combination of these features lead to the prediction that the topological surface states are robust against localization; hence, they are expected to wrap the surface of the topological bulk sample regardless of the presence of surface defects. The experimental results present in the last chapter showed the absence of backscattering between equal and opposite momentum states, owing to their chiral spin texture. However, these experiments do not demonstrate whether topological surface states are in fact extended, as the experiments did not probe the transmission properties of these states. In this chapter, we present the STM results [88] on antimony’s surface states where we measured their reflection by and transmission through naturally occurring crystalline steps separating atomic terraces [figure 3.1(a)]. By using a simple physical model for the helical metal on the surface of an antimony crystal, we show that the probability for electron transmission through a crystal step of about 35%, a significant enhancement compared with the surface states in ordinary metals such as copper or gold [figure 3.1(b)], which are either reflected or absorbed by atomic steps. This chapter is mainly adopted from our published results in reference [88].
3.1 The Band structure of Sb and basic STM results

3.1.1 Band structure of Sb and ARPES measurements

Because there is a finite direct energy gap between the bulk valence and conduction bands at each high symmetry point of the Brillouin zone in Sb the topological indices are well defined for Sb. The band structure calculation for its surface states exhibits a non-trivial energy dispersion that is adiabatically connected to that of Bi$_{1-x}$Sb$_x$. Note that in topological insulators, the spin-orbit coupling has to modify the bands in the fundamental way, which makes them distinct from other surface states with spin-orbit coupling. Other words, there are material systems with strong spin-orbit coupling whose surface states are not topologically protected [figure 3.2(a) and 3.2(b)]. In such topologically trivial spin-orbit metals such as gold, a free-electron like surface state is split into two parabolic spin-polarized sub-bands that are shifted in k-space relative to each other. As a result, two concentric spin-polarized Fermi surfaces are created, one having an opposite sense of in-plane spin rotation from the other. The schematic of the bulk and surface band structure presented in figure 3.2 demonstrates this point. In fact, backscattering has been observed for the surface states of gold, in spite of strong spin-orbit coupling [90].

Figure 3.2(c) show a spin-integrated ARPES intensity spectrum of Sb(111) from $\bar{\Gamma}$ to $\bar{M}$ [47]. The measurements identified two V-shaped surface states centered at $\bar{\Gamma}$, and resonance states centered about $k_x = 0.25$ Å$^{-1}$. Resonance states are the coexistence of surface and bulk states. In examination of the ARPES intensity map of the Sb(111) surface and resonance states at $E_F$ reveals that the
Figure 3.2: **ARPES measurements of the surface bands on Sb (111).** (a) Schematic of the bulk-band structure (shaded areas) and surface-band structure (red and blue lines) of Sb near $E_F$ for a Sb (111) is depicted (b) if the surface bands had a different dispersion and were merging with the bulk bands differently, then the topologically of the surface bands would have been trivial (Au-like). The corresponding surface Fermi contours are also shown, where the hexagonal dotted line is the first Brillouin zone of the (111) surface. (c) Spin-integrated ARPES spectrum of Sb (111) along the $\bar{\Gamma}$-$\bar{M}$ direction. The surface states are denoted by SS, bulk states by BS, and the resonance states by RS. (d), ARPES intensity map of Sb (111) at $E_F$ in the $k_x$-$k_y$ plane. The only one Fermi contours encircling $\bar{\Gamma}$ is formed by the inner V-shaped SS band seen in (c). Adopted from reference [47].
central Fermi contours enclosing $\bar{\Gamma}$ are formed by the inner V-shaped surface states (SS) only. The outer V-shaped SS on the other hand forms part of a tear-drop shaped Fermi contours that does not enclose $\bar{\Gamma}$, unlike the case of gold [ figure 3.2(d) ].

3.1.2 Basic STM measurements

We have studied the (111) surface of Sb in our home-built cryogenic STM that operates at 4 Kelvin. Single crystals of Sb were cleaved in situ in ultrahigh vacuum to expose a pristine surface. Crystals of Sb have rhombohedral structure consisting of hexagonal planes of Sb stacked on top of each other along the [111] crystallographic direction. These planes are bonded by weak van der Waals forces, making the (111) plane the natural cleavage plane. STM topographs show an image of the cleaved surface with nanometer sized atomic terraces that are separated by single atom height steps [ figure 3.3(a) ]. We have examined the spatial and energy dependence of the local density of states (LDOS) within the terraces by performing differential conductance (dI/dV) measurements along a line perpendicular to the step edges. In figure 3.3(b), the spatially averaged spectroscopic measurements for terraces of different widths show peaks that signal the occurrence of quantized resonances. These spectroscopic features start at around -225 meV, which is the bottom of the topological surface state bands as measured by ARPES studies [91, 47]. Resolving the spatial variation of the LDOS within each terrace [see figure 3.3(c) ], we find clear evidence for quantized resonances, with nearly equal spacing in energy. The dI/dV measurement result as a function of position and energy resembles the pattern for a particle in a quantum confinement, where the interference of the allowed wavefunctions at a given energy leads to sinusoidal patterns with increasing number of peaks at higher energies. The measurements also shows that each atomic terrace has mirror symmetry; indicating that for confined surface states step up and step down terraces are identical potential barriers. The quantized resonances and the standing-wave patterns in the LDOS are caused by scattering of antimony’s topological surface states from the atomic step edges and can be analyzed to obtain reflection properties of these steps.

The linear dispersion of the surface bands of Sb distinctly changes the energy level spacing between adjacent quantized levels ($\Delta E$), leading to a unique relation between the spacing and the quantized level different from other surface states with parabolic dispersions. In figure 3.4(a), we plot $\Delta E$ as a function of energy for different terrace sizes, where for the entire range of measurements $\Delta E$ is essentially independent of energy. The constant energy spacing is the hallmark of Dirac-like quantization and indicates that the surface states of Sb are fundamentally different from the Shockley
Figure 3.3: The topological surface states of Sb(111) on atomic terraces. (a) STM topographic image (V\text{bias} = 1 V, I = 8 pA) of a 2,500 Å by 1,250 Å area showing terraces of various widths separated by 3.7 Å-high single atomic steps. (b) The dI/dV measurement averaged over each terrace shows quantized peaks. The spectra are offset vertically for clarity. (c) Spatially and energetically resolved dI/dV measurements taken along the dotted arrow in a demonstrate the interference in space and the quantization in energy. Adopted from reference [88].

Surface states of noble metals, where the quadratic dispersion leads to a linear energy dependence for $\Delta E$. Observation of linear dispersion for such an extended range of energy is beyond simple models for spin-orbit coupling. Figure 3.4(b) shows the averaged value of $\Delta E$, which is proportional to the inverse of the terrace width, $L$, $\Delta E \sim 1/L$. This dependence further confirms the linear dispersion, $E = c k$, and manifests the Dirac nature of quasi-particles on the surface of Sb(111).

Before study of reflection and transmission, we demonstrate that the standing-wave patterns and the nearly linear quantized resonances in LDOS can be understood on the basis of the ARPES band structure of antimony's surface states and the spin selection rules that must be obeyed for scattering of topological surface states. The patterns seen in different terraces are similar, and we picked the measured LDOS over a terrace of width 390 Å, which is shown in figure 3.5(a). The measured LDOS is particularly high at two energies, associated with the extremum points at which the dispersion turns flat. The first energy is where the bottom of the surface bands is located at -230 mV, and the second is at -100 mV, where the hole pocket overlap vanishes in the $\bar{\Gamma}-\bar{K}$ direction. The Fourier transform of the dI/dV measurement is presented in figure 3.5(b) and reveals two allowed scattering wavevectors, $q_A$ and $q_B$, for the standing-wave pattern of the LDOS. As discussed in detail in the last chapter, the wavevectors of incident and elastically scattered quasi-particles at different momentum states, $\vec{k}_i$ and $\vec{k}_f$, which their interference leads to the modulation in the conductance at a wavevector $\vec{q}$.
are related by $\vec{q} = \vec{k}_f - \vec{k}_i$. For the dI/dV mapping shown on the surface of Sb, the parallel step edges are oriented such that $\Gamma - M$ is perpendicular to the step edge direction, and hence standing wave patterns are formed with their wavevectors in this direction. We found that among several scattering processes that could lead to such $\vec{q}$’s only two are present, while the others which involve scattering between states of orthogonal spins are absent. The scattering wavevector $\vec{q}_A$ is associated with scattering between adjacent hole pockets [figure 3.5(d)], and vanishes once the hole pockets overlap in k-space around -110 meV. $\vec{q}_B$ originates from scattering between the central electron pocket and the hole pocket with oppositely oriented momentum, and same spin orientation [91]. Therefore, unlike conventional confined states are the superposition of states with equal and opposite momenta ($\vec{k}$ and $-\vec{k}$), the chiral spin texture of antimony eliminates the possibility of superposition of states with orthogonal spins. Consequently, the $\vec{q}$’s observed correspond to superposition between states with different magnitudes of $\vec{k}$ but similar spin states.

The geometric confinement further leads to the quantization of the energy states of quasi-particles through the quantization of the $\vec{q}$’s, correspond to the standing waves seen in the real space. The geometric constraint set by the terrace width, $L$, result in quantization of the allowed $\vec{q}$’s, such that $|\vec{q}_n| = 2\pi n / L$, where n is positive integer and $\vec{q}$ is the scattering wavevector. Note that the usual $L^2$ in the denominator, characteristic of parabolic dispersions, is replaces by $L$, due to the fact...
Figure 3.5: **Allowed scattering wavevectors and their quantization.**  
(a) The dI/dV measurement on a 390Å-wide terrace.  
(b) Energy-resolved Fourier transform of the spatial modulation of the data in (a) reveals the quantization of scattering wavevectors $\mathbf{q}_A$ and $\mathbf{q}_B$.  
(c) The dispersions of $\mathbf{q}_A$ and $\mathbf{q}_B$ match the dispersion of the surface bands as measured by ARPES along the high-symmetry directions (solid lines) and extend it above the Fermi level (dotted lines).  
(d) Contours of constant energy of the antimony surface state. The contours consist of a central electron pocket and six surrounding hole pockets. The colored arrows represent spin texture of the surface state. The scattering wavevectors $\mathbf{q}_A$ and $\mathbf{q}_B$ indicate allowed scattering processes. $\bar{\Gamma}$, $\bar{K}$ and $\bar{K}$ are the high-symmetry points of the first Brillouin zone, with $\bar{\Gamma}$ located at the centre of the zone, $\bar{M}$ located in the middle of each side of this six-fold symmetric zone, and located at the vertex. Adopted from reference [88].
that surface bands have a linear dispersion. Despite their quantization, the energy dependences of $\vec{q}_A$ and $\vec{q}_B$ are related to the dispersion measured by ARPES bands because $\vec{q}_n(E_n) = \vec{k}_i(E_n) - \vec{k}_f(E_n)$, where $\vec{k}_i$ and $\vec{k}_f$ are the initial and final momentum states in an elastic scattering with energy. The nearly linear energy dependences of $\vec{q}_A$ and $\vec{q}_B$ have slopes ($\sim 1.2$ eVÅ) that are in excellent agreement with ARPES measurements. Previous studies on noble metal surface states have established the quantization rules of momentum states for restricted geometric structures. For these spin-degenerate surface states, where $\vec{k}_i = -\vec{k}_f$ is the dominant scattering, quantization of $\vec{q}_s$ results in the quantization of $\vec{k}_s$. In contrast, for spin non-degenerate surface states of Sb, for which the backscattering is not allowed due to spin rules ($\vec{k}_i \neq -\vec{k}_f$), the quantization of $\vec{k}$ is does not take place. Consequently, the set of quantized energy levels is given by $\vec{q}_n(E_n) = \vec{k}_i(E_n) - \vec{k}_f(E_n) = 2\pi n/L$.

For a linear dispersion this indeed reduces to $E_n = c|\vec{q}_n|$, where $c$ involves the two bands that are connected by the quantized $\vec{q}_n$ and is related to their dispersion slope by $c = (\frac{1}{\hbar c_F i} + \frac{1}{\hbar c_F f})^{-1}$.

3.2 Reflection

The energy widths of the quantized resonances in the LDOS contain information on the scattering properties of antimony’s topological surface states. In general, energy-level broadening, $\Gamma$, is inversely proportional to the lifetime of a state, $\tau$: $\Gamma \propto \hbar/\tau$ ($\hbar$, Planck’s constant divided by $2\pi$). Examining the broadening of the resonances as a function of energy for different terraces [figure 3.6], we find that $\Gamma(E) = \Gamma_L + (E - E_F)^2$, where $E_F$ is the Fermi energy (with $\hbar/\gamma = 0.37 \pm 0.01$ fs.eV, for terraces where level broadening is smaller than level spacing). This functional form illustrates the role of electron-electron scattering in the decoherence of topological surface states, similar to the case in other Fermi liquids [89]. However, $\Gamma_L$, the residual finite resonance width at $E_F$, characterizes the degree to which step edges can reflect antimony’s topological surface states in the same spin channel.

To quantify reflection probability of quasi-particles in a quantum box formed by parallel atomic step edges, we used a simple model based on multiple scatterings [88]. In this approach, the wavefunction is the coherent superposition of the electron injected from the STM tip and all possible scatterings. When the STM tip is positioned at $x=x'$ between atomic steps of Sb, the injected electron from the STM tip propagates to the atomic boundaries, and after multiple scatterings inside the box [figure 3.7], the resulting wavefunctions $\psi_1$ is

$$\psi_1 = e^{-ik_F x'} + e^{-ik_F x'} e^{ik_P x'} R_0 e^{ik_P z'} + e^{-ik_P z'} e^{ik_F x'} R_0 e^{ik_P L} R_0 e^{ik_P (L-x')} +$$
Figure 3.6: **Lifetime and leakage of quantized quasiparticles.** Each resonant peak is fitted to a Lorentzian function to yield the full-width at half-maximum of the quantized energy peaks, $\Gamma$ (inset). The plot shows the energy dependence of $\Gamma$ for two different terraces. The dashed lines are parabolic fits, and $\Gamma_L$ is the peak broadening at the Fermi energy. The error bars are from the fitting process of the resonant peaks and are mainly due to the measurement resolution. Adopted from reference [88].

\[
e^{-ik_P x'}e^{ik_P x'}R_0e^{i\mathbf{k}_S L}R_0e^{ik_P L}R_0e^{i\mathbf{k}_S x'} + \ldots
\]

\[
= \frac{1}{1 - R_0^2 e^{i(k_P+k_S)L}}e^{-ik_P x'} + \frac{R_0^2}{1 - R_0^2 e^{i(k_P+k_S)L}}e^{i\mathbf{k}_S x'},
\]

where $L$ is the width of the box, and the reflection coefficient $R_0$ is a complex number to account for phase shift during scattering process at the step edges. $K_P$ is the wavevector for the left going waves and $K_S$ is of the right going one. $P$ and $S$ are two high density points of the Fermi contours [marked in figure 3.5(d)], which have the same spin state. The result would be identical if the other spin states $Q$ and $T$ were considered.

The wavefunction $\psi_1$, which its calculation assumes the injected electron initially travels to the right, has a similar form and is

\[
\psi_2 = \frac{R_0e^{i(k_P+k_S)L}}{1 - R_0^2 e^{i(k_P+k_S)L}}e^{-ik_P x'} + \frac{1}{1 - R_0^2 e^{i(k_P+k_S)L}}e^{i\mathbf{k}_S x'},
\]

The recorded local density of states (LDOS) by STM, is related to both of these partial waves and is given by

\[
|\psi(k, x')|^2 = \frac{1}{N}(|\psi_1|^2 + |\psi_2|^2)
\]
Figure 3.7: **Multiple scatterings in a quantum box.** In this schematic representation, the left going part of the injected wave has momentum \( k_P \) and the right going has \( k_S \), where P and S refer to specific points of the Fermi contour, which are shown in figure 3.5(d). Adopted from reference [88].

\[
\frac{1}{N} \left( \frac{2r^2 + 2 + 4r \cos(\theta_r + |k_P + k_S| L/2) \cos(|k_P + k_S|r)}{1 + r^4 - 2r^2 \cos(3\theta_r + |k_P + k_S| L)} \right),
\]

where \( N \) is the normalization, and the reflectivity was expressed as \( R_0 = re^{i\theta_r} \) with \( r \) and \( \theta_r \) real numbers. Note that the successive injected electrons do not have a well-defined phase relation; and therefore, the interference term in the equation above was ignored. The condition for resonance tunneling is satisfied when \( 2\theta_r + |k_P(E) + k_S(E)| L = 0 \), under which the full width at half maximum (FWHM) of the resonance peak can be obtained from the Taylor expansion of the denominator

\[
\Gamma_L = 2 \sqrt{\frac{c^2 (1 - |R_0|^2)^2}{L^2 |R_0|^2}},
\]

where \( c = (c_P^{-1} + c_S^{-1})^{-1} \), \( E = c_F k_P \), and \( E = c_S k_S \). The coefficients \( c_P \) and \( c_S \) are the slopes in the linear dispersion of the surface bands, and from ARPES measurements are known to be \( c_P = 3.8 \) V.Å and \( c_S = 1.7 \) V.Å. The value of \( \Gamma_L \) as measured by STM [see figure 3.6] for two different terraces of \( L = 110 \) Å and \( L = 160 \) Å, were 19.4 mV and 14.3 mV, respectively, yielding \( |R_0|^2 = 0.42 \pm 0.04 \).

Therefore, based on this simple model of energy broadening based on a double-barrier potential, we find that the energy broadening corresponds to a 42 \( \pm \) 4\% reflection probability for this process. This reflectivity is much lower than those reported for free-electron-like surface states on copper, silver or gold from STM and other measurements; however, it can be a result of scattering of the
surface states into the bulk states rather than their transmission. Indeed, previous studies show that non-topological noble-metal surface states are absorbed in the bulk states with 30-50% probability without transmission when scattered by step edges [92, 89, 75, 93, 94]. We will explore the probability of transmission and absorption to the bulk in the next section, and see how topological surface states distinct themselves from other surface states by showing strong transmission from one atomic terrace to the next.

3.3 Transmission

Finite transmission through the atomic steps edges should give rise to coupling between electronic states on adjacent terraces. To search for evidence of such effects, we examine a configuration of atomic steps consisting of a narrow (110 Å) and a wide (2500 Å) terrace, which resembles a nanoscale Fabry-Prot resonator, as shown in figure 3.8(a). The narrow terrace shows the signature of quantized resonances according to the spin scattering rules described above [figure 3.8(b)]. On the wide terrace, which is effectively semi-infinite, the LDOS oscillation forms an almost continuous pattern with diverging wavelength towards the bottom of the surface state bands. The standing-wave patterns on the wide terrace also obey the chiral spin texture and are made up of the two wavevectors $\vec{q}_A$ and $\vec{q}_B$ described above, as illustrated in the Fourier transform in figure 3.5(b). Any finite transmission through the step edges results in resonant tunnelling of surface state electrons from the wide terrace through the electron states of the narrow terrace.

Evidence for resonant tunnelling of the topological surface states can be seen both in the structure of energy-resolved modulations of the LDOS and in its Fourier transform. At energies corresponding to the quantized resonances of the narrow terrace, there are clear suppressions of the modulation in the LDOS on the wide terrace [figure 3.8(b)]. We illustrate this in figure 3.8(d) by plotting the Fourier intensity of the $\vec{q}_B$ peak as a function of energy. Once the overall background changes in the LDOS [figure 3.8(e), dashed green line] are removed, the intensity of the $\vec{q}_B$ Fourier component displays characteristic signs of Fabry-Pérot resonant tunnelling in which the surface state electrons pass through the narrow terrace without reflection. Deviation from perfect transmission is, however, a signature of scattering of the surface states into the antimony bulk states. Similarly, the width of the Fabry-Pérot resonance can be used to extract the transmission and reflection probabilities.

Here we present the detail of a simple multi-scattering model, similar to the one used in the last section, which we used to calculate transmission and bulk absorption for our data. From this analysis we deduced that probability of bulk absorption is $23 \pm 7\%$, and transmission and reflection
Figure 3.8: **Resonant tunneling between adjacent terraces.** (a) STM topographic image of a narrow terrace (L = 110 Å) and an adjacent wide terrace (L = 2,500 Å). Only part of the wide terrace is displayed. The colour scale shows the height variations in the topographic image. (b) On the narrow terrace, the dI/dV measurement shows the quantization of the energy levels. (c) The dI/dV measurement on the wide terrace shows sudden phase shifts and suppressions of the dI/dV intensities at around +5 meV and ~70 meV due to resonant tunneling. The averaged background conductance at each energy has been subtracted. (d) The Fourier transform of the spatial modulation of the data in c. The grey markers indicate suppression of modulation intensities. (e) Spectral weight of the scattering wavevector $q_B$ as extracted from d. The dashed line corresponds to the background spectral weight in the absence of resonance tunneling. (f) Spectral weight (circles) normalized by the background. The best fit (red line) yields ~ 42% reflection, ~ 35% transmission and ~ 23% bulk absorption from the scattering process at the boundary. The blue line, which is the fit based on a model without bulk absorption, is displayed for comparison. Adopted from reference [88].
Figure 3.9: Multiple scatterings from two adjacent atomic step edges. The STM tip positioned at x=x’ away from the near edge of the narrow terrace. \( k_P \) and \( k_S \) are referring to specific points of the Fermi contours, which are shown in figure 3.5(d). Adopted from reference [88].

probabilities are 35 ± 3% and 42 ± 4%, respectively. In contrast, STM data on Fabry-Pérot structures for non-topological surface states of Ag(111) show no evidence of resonant tunnelling and can be explained in detail by considering only reflections and absorption of the surface states at the step edges [75].

The dI/dV modulation on a atomic terrace bounded in one side by a narrow terrace of width \( L \), and practically infinite in extend on the other side, can be modeled by considering scatterings which involve both edges of the narrow terrace [ figure 3.9 ]. When the STM tip is positioned at x=x’ away from the near edge of the narrow terrace, total reflection coefficient \( A \) after several scatterings is given by

\[
A e^{ik_s x'} = R_0 e^{ik_s z'} + T_0 e^{ik_p L} R_0 e^{ik_s z} T_0 e^{ik_s z'} + T_0 e^{ik_p L} R_0 e^{ik_p L} R_0 e^{ik_s L} T_0 e^{ik_s L} T_0 e^{ik_s z'} + ... \\
= |R_0 + \frac{T_0^2 R_0 e^{i(k_P + k_S)L}}{1 - R_0^2 e^{i(k_P + k_S)L}}| e^{ik_s z'},
\]

where the reflection coefficient \( R_0 \) and transmission coefficient \( T_0 \) are complex numbers. The interference pattern between the incident wave and the total reflected wave is

\[
| \psi(k, x') |^2 = \frac{1}{N} \left( e^{-ik_P x'} + A e^{-ik_s x'} \right) \left( e^{-ik_P x'} + A e^{-ik_s x'} \right)^* 
\]
where $N$ is the normalization factor. The pattern shows spatial oscillation with wave-number $k_p+k_s$, and oscillation amplitude of

$$f(T_0, R_0) = \frac{1}{N} \sqrt{(A^*+A)^2 + |i(A^*-A)|^2} = \frac{2}{N} |A|.$$  \hspace{1cm} (3.7)

To account for absorption by the bulk, we introduce a factor, $\alpha$, and modify the wavefunction continuity and particle conservation relations accordingly:

$$1 + R_0 = T_0,$$

$$|R_0|^2 + |T_0|^2 = 1 - \alpha.$$  \hspace{1cm} (3.8)

By setting $R_0=r e^{i\theta_r}$ and $T_0=t e^{i\theta_t}$, then $\theta_r$ and $\theta_t$ can be expressed in terms of $\alpha$ and $r$ and $t$.

$$r^2 + t^2 = 1 - \alpha$$

$$\theta_r(\alpha, r) = \arccos\left(\frac{-\alpha}{2r} - r\right),$$

$$\theta_t(\alpha, t) = \arccos\left(\frac{-\alpha}{2t} + t\right).$$  \hspace{1cm} (3.9)

Hence, the oscillation amplitude can be rewritten as

$$f(r, t) = \frac{2}{N} \sqrt{r^2 + r^2 t^4 - 2r^4 t^2 \cos(2\theta_r - 2\theta_t) + 2t^2 r^2 \cos(2\theta_t + 2LE/c)} / (1 + r^4 - 2r^2 \cos(2\theta_r + 2LE/c)),$$  \hspace{1cm} (3.10)

where $c=(c_p^{-1}+c_s^{-1})^{-1}$, $E=c_p k_p$, and $E=c_s k_s$.

From residual peak broadening ($\Gamma_L$ in figure 3.6, and fitting procedure described in 3.3.1), $r^2$ is found to be $0.42 \pm 0.04$. Substituting this value in the above equation allows to fit the data shown in figure 3.8(e), yielding $t^2 = 0.35 \pm 0.03$. The bulk absorption is accordingly set to $\alpha=0.23 \pm 0.07$. Figure 3.10 shows good spatial agreement between the STM data and the simulation resulting from using these values.
Figure 3.10: Transmission at resonance tunneling energies. (a) The $dI/dV$ measurements on an open terrace close to a narrow terrace. (b) The LDOS = $|\psi|^2$ resulting from a simple model captures the details of the spatial modulations and yields the reflection probability, transmission probability, and bulk absorption of 0.42, 0.35, 0.23, respectively. Adopted from reference [88].

3.4 Conclusion

The remarkable finding that topological surface states are as likely to be transmitted through an atomic step edge as reflected by it demonstrates an important difference between topological states and typical surface states in other materials. The transmissibility illustrates the extended nature of topological surface states even in the presence of strong surface barriers. The nearly equal probabilities of reflection and transmission at the step edges in antimony can also be understood on the basis of the Sb's topological surface state's band structure, which includes both forwards- and backwards-moving momenta with the same spin orientation. The observation of high transmission also suggests excellent transport of topological surface states despite boundaries that would suppress the ability of other surface states to carry current. Such a feature may find applications in nanoscale devices, where there is a need for high surface conductivity because the surface-to-volume ratio is large.

Our experiments also show that nanoscale structures can be used to explore the properties of topological surface states both for fundamental studies and to evaluate their potential for device applications. The antimony crystals used in this study are bulk metals, rather than insulators, and as discussed this causes some 'leakage' of the surface electrons into the bulk during the collision with a step (about 23%). Also, antimony's surface energy-band structure is more complicated than the ideal helical metal, and thus allows for more near-U-turns than can be explained by the arguments presented above. These factors suggest that the electron-transmission probability through the steps
can be further increased by using $Bi_2Se_3$ or $Bi_2Te_3$, which can be tuned to become an insulator in the bulk. Clearly, an extension of this study to these materials with a bulk gap and simpler surface band structure would allow further examination of these states without the complication of coupling to the bulk states. Our demonstrations of the transmission of topological surface states through barriers that fully reflect other surface states suggest an extraordinary insensitivity of transport through these states to the geometrical shape of the surface.
Chapter 4

Surface states scattering in the inhomogeneous landscape of bulk dopants

In this chapter we present STM studies [95] on the topological surface states of $Bi_2Se_3$ and $Bi_2Te_3$, the simplest manifestation of 3D topological insulators. The surface of these materials hosts electronic states with a single Dirac cone and helical spin texture and no other surface bands. Their simple band structure makes them the material of choice in TI research. There are numerous proposals relied on manipulating these surface states near the Dirac point [60, 96, 97, 98]. Chemical doping of these compounds is a frequently used approach to tune the chemical potential to the Dirac point. Therefore, it is essential to characterize the response of these states to magnetic and non-magnetic dopants. The problem is of great experimental and theoretical interest, since the wavelength of the Dirac electrons diverges with approaching the Dirac point, and as a result the electronic states close to the Dirac are susceptible to disorder of any length scale [99]. Transport and spectroscopic measurements by STM show that doping is quite successful in moving the chemical potential globally. However, the response of these surface states to bulk dopants at the atomic scale has not been examined [figure 4.1]. The low density of states close to the Dirac point makes it extremely difficult to probe these states; and consequently, previous STM studies have not probed the properties of topological surface states near the Dirac point of the surface band structure [49, 52]. The absence of any signatures of scattering in STM measurements near the Dirac point has been attributed to
Figure 4.1: The schematic of the charge dopants that are substituted in the bulk of the crystal to tune the chemical potential. What are the consequences of bulk doping on the electrostatic potential at the surface? And how the surface states are going responding to that?

topological protection of these states against scattering [49, 52], although only direct backscattering is strictly prohibited. In this chapter, we present the results of extensive measurements [95] on both magnetic and non-magnetic doped samples of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$, to obtain a comprehensive understanding of the role of dopants on topological surface states. We discuss if scattering from magnetic dopants is different from non-magnetic ones, and also if the spin sensitive scattering is still applicable when it comes to scattering from magnetic impurities. This chapter is mainly adopted from our published results in reference [95].

### 4.1 Charge puddles induced by dopants

As discussed in chapter 2, the surface structure of $\text{Bi}_{1-x}\text{Sb}_x$ is rather complicated and the band gap is rather small ($\sim 30$meV). This motivated a search for topological insulators with a larger band gap and simpler surface spectrum. A second generation of 3D topological insulator materials, especially $\text{Bi}_2\text{Se}_3$, offers the potential for topologically protected behavior in ordinary crystals at room temperature. ARPES and first-principles calculations by several research groups show that the surface band structure of $\text{Bi}_2\text{Se}_3$ and $\text{Bi}_2\text{Te}_3$ are consist of a single Dirac cone [37]. The measurements show that for $\text{Bi}_2\text{Te}_3$, the rate of dispersion is different for the two high symmetry directions [figure 4.2], leading to hexagonal contours of constant energy [the top row in figure 4.2(b)]. At $\sim 250$meV above the Dirac binding energy the rate of dispersion in $\bar{\Gamma}-\bar{M}$ direction changes, which leads to warped hexagonal contours of constant energy, as shown in the bottom row in figure 4.2(b). The
spin texture of the surface bands were also probed by spin-resolved ARPES and are shown by gray arrows in figure 4.2(c). Close to the same energies where the $\bar{\Gamma}-\bar{M}$ dispersion changes rate and the contours become hexagonally warped, the spin of the quasiparticles become canted out of the 2D plane [the gray circular symbols in figure 4.2(c)]. By adding an unconventional hexagonal warping term in the surface band structure, Fu [48] were able to account for warping and its coincidence with the canting of the spin texture in $Bi_2Te_3$. From ARPES, the dispersion of the surface bands of $Bi_2Se_3$ is known to be similar, with warping being only weaker. However, there is an important difference between the surface bands of $Bi_2Se_3$ and $Bi_2Te_3$. For $Bi_2Te_3$, the binding energy of the Dirac point is below the bulk valence band energies, and hence the Dirac point is "embedded". On the other hand, for $Bi_2Se_3$, the Dirac point is in the semiconductor gap [figure 4.3(a) and (b)].

Many of the interesting theoretical proposals that utilize topological insulator surfaces require the chemical potential to lie at or near the surface Dirac point. This is similar to the case in graphene [100, 101], where the chemistry of carbon atoms naturally locates the Fermi level at the Dirac point. This makes its density of carriers highly tunable by an applied electrical field and enables applications of graphene to both basic science and microelectronics [23, 24]. The surface Fermi level of a topological insulator depends on the detailed electrostatics of the surface and is not necessarily
at the Dirac point. Moreover, for naturally grown $\text{Bi}_2\text{Se}_3$ the Fermi energy is not even in the gap. The observed n-type behavior [43] is believed to be caused by Se vacancies. It is a common practice among the experimental groups in the community, and the subject of investigation of this chapter, to modify the Fermi level by appropriate chemical modifications. For instance, it is shown in reference [43] that by doping bulk with a small concentration of $\text{Ca}$, which compensates the $\text{Se}$ vacancies, the Fermi level can be placed within the bulk band gap. The electrical transport measurements on $\text{Bi}_2\text{Se}_3$ are shown in figure 4.3(c), demonstrating how the chemical potential is moved away from the bulk, resulting in the insulating behavior. One maybe puzzled by the insulating behavior given the presence of metallic surface states. Note that the resistivity measured here is done for rather thick samples, and the results reflect the overwhelming contribution of the bulk in comparison to the surface states.

Single crystals of $\text{Bi}_2\text{Te}_3$ and $\text{Bi}_2\text{Te}_3$ doped with $\text{Ca}$ or $\text{Mn}$ were cleaved in situ under ultrahigh vacuum at room temperature before performing STM experiments at low temperatures. All STM topographic images [figure 4.4] show triangular depressions that are associated with the substitutional dopants in layers close to the surface. A detailed study of the topographs, which is presented in appendix B, shows that the centers of these triangles are at the $\text{Bi}$ site, and hence confirming their proper substitution for $\text{Bi}$. They substitutional dopants serve as strong scattering centers both for the helical surface states as well as for the bulk states. There is no readily visible correlation between the regions of high and low density of states in the topography (the yellow dominated and
Figure 4.4: (a), (b), (c) The topography over a 1000 Å by 1000 Å area on the surface of Bi$_{1.95}$Ca$_{0.05}$Te$_3$, Bi$_{1.95}$Mn$_{0.05}$Te$_3$, and Bi$_{1.95}$Mn$_{0.05}$Se$_3$, respectively. Mn and Ca dopants appear as triangular depressions. The faint clover-like patterns are substitutional atoms on deeper Bi layers. The filled circles, show the position of the tip during dI/dV spectroscopy, which is presented in the next figure, 4.5.

the blue dominated regions) and the position of the dopants. Nevertheless, the spectroscopy on the surface can bring out the fact that regions of the high and low LDOS are the consequence of charge dopants, as we discuss next.

In figure 4.5(a) and (b) we plot the dI/dV spectra taken as the tip was parked over the surface along the line shown by circular symbols in figure 4.4 across the Mn-doped Bi$_2$Te$_3$ and Bi$_2$Te$_3$ surfaces, respectively. From the plots, it becomes evident that there is an overall energy shift in the spectrum at different locations. Remarkably, both the bulk and in-gap surface bands are affected in a similar fashion. These fluctuations in the local band energies with respect to chemical potential, are ubiquitous to all single Dirac cone topological insulators discussed in this chapter and are present regardless of whether the dopants are magnetic or not. These changes are associated with bulk disorder in our samples, such as poorly screened charged deep dopants, and are not directly correlated with the locations of the surface dopants resolved in STM topographs. As a consequence of these nanoscale changes in the bulk and surface electronic structure the energy of the Dirac point assumes a range of values relative to the sample’s electro-chemical potential rather than a well-defined energy throughout the sample surface. Based on the ARPES measurements and the shape of the spectrum, the spatially averaged binding energy of the Dirac point $E_D^0$ can be determined. The fluctuations of the Dirac point $E_D(r)$, around $E_D^0$, for the three samples are shown in figure 4.5(c). The insets show that $E_D$ is indeed normally distributed over $\Delta E \sim$20-40 meV about the
mean $E_D^0$.

The charge puddles impact the surface physics the most when the Fermi energy lies in the vicinity of the sample’s average Dirac point, $E_D^0$. A cross-section of the Dirac map, along the yellow line in figure 4.5(c), is shown in panel (d) and highlights that by tuning the sample’s average chemical potential close to the Dirac point, the local electronic structure alternates between electron-like and hole-like doped regions that would have opposing helical spin texture. While degrading the surface conductivity, this can also give rise to mesoscopic phenomena such as the magneto-fingerprint that was indeed reported in macroscopically large single crystals of topological insulators [42]. Nevertheless, lack of resonances in the measured point spectra asserts that the charge puddles are strongly coupled.

A similar scenario occurs in graphene which offers another realization of a 2DEG with Dirac spectrum (though in graphene each state is both valley and spin degenerate). The charge puddles in graphene are believed to form due to charged impurities in the substrate or trapped in between the substrate and the graphene sheet. In the present case, of doped-topological insulators, we are able to directly visualize the charged impurities on the top most layers below the surface. However, lack of correlation between these and the puddles as well as the overall energy shift exhibited both by surface and bulk bands suggest that the surface puddles are the two-dimensional slice of the inhomogeneously charged pockets that form in the doped bulk, as schematically illustrated in figure 4.5(e).

4.2 Quasi-particle interference patterns

4.2.1 QPI in the presence of puddles

The differential conductance ($dI/dV$) spectroscopic maps taken over the surfaces [figure 4.6] indicate that an intricately modulated local density of state (LDOS) forms in response to dopings and puddles. The modulations seen in the maps comprise of both long and short wavelengths. The high energy $dI/dV$ maps of all doped compounds studied [left panels of figure 4.6] are dominated by short wavelength quasiparticle interference (QPI) patterns. At somewhat lower energies [middle panels of figure 4.6] the dispersing QPI patterns are found to ride a spatial inhomogeneity of a larger length-scale. At lower energies and close to the Dirac binding energy [right panels of figure 4.6], the surface LDOS is strongly dominated by the puddles. None of these features seems to be sensitive to the magnetic or non-magnetic nature of the substitutional dopants. From investigation of $dI/dV$ maps
Figure 4.5: **Bulk origin of the surface charge inhomogeneity.** (a), (b) The LDOS measured at various locations on the surface of Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$, respectively, showing a rigid shift of the bulk and surface bands (band structures shown schematically). From ARPES measurements the spatially averaged binding energy of the Dirac point $E_D^0$ with respect to the bulk bands is known. At the atomic scale, the binding energy of Dirac point, $E_D(r)$ shifts from point to point. The extracting of the Dirac point is by linear extrapolation of the surface state’s LDOS in Bi$_2$Te$_3$, and by finding the energy corresponding to the point of minimal LDOS in Bi$_2$Se$_3$. Insets show the Gaussian distribution of $E_D$. (c) Spatial distribution of $E_D$ extracted from local shift in LDOS. The bright spots on the left panel highlight the position of Mn dopants which are visible in topography, and demonstrating the fact that there is no readily detectable correlation between them and puddles. (d) Line cut along yellow line in (c) showing electron and hole puddles that form when the chemical potential is tuned to the average Dirac point in the sample. (e) Schematic demonstration of the fact that the charge puddles are the consequence of bulk dopants. Adopted from reference [95].
Figure 4.6: **Interference and inhomogeneity on the surface of doped topological insulators.** Conductance maps taken at high, intermediate and low sample biases showing dispersive quasi-particle interference (QPI) pattern that rides a quenched inhomogeneity of the LDOS.
with much more intermediate energies, it becomes evident that the long wave length modulation
due to puddles appeared in the LDOS, does not disperse with energy. It only becomes more visible
at lower energies. This is expected if the origin of the long wavelength modulations is bulk dopants.
Another reassuring observation is comes from the comparison of these maps and the Dirac maps
presented in figure 4.5(c), which are done on the same corresponding areas [ compare the left column
of 4.6 with 4.5(c) ]. The similarity of the structures is easily visible. The typical size of the quenched
puddles is a few hundred Angströms - much larger than the ~10Å dimension of a single dopant, as
captured in the topographic image.

A particle traversing the surface of the doped sample has to adjust to the underlying charge-
puddles potential by repeatedly altering its crystal momentum. To quantitatively determine the in-
fluence of this bulk-induced disorder potential on the short wavelength interference patterns caused
by the surface impurities, we divide the dI/dV maps into sub-regions, as shown in figure 4.7(a),
according to intervals of lower and higher energy-shift of the Dirac point. Obtaining Fourier trans-
form (FT) from QPI measurements on two distinct regions ( defined by upper and lower halves of
distribution of Dirac points ) we find clear shifts of the QPI’s q vectors. An example of such a
momentum shift ( Δq~ 0.01Å−1 ) is shown in figure 4.7(b). Evidently, the surface Dirac electrons
alter their wavelengths to adjust to the underlying bulk disorder potential, and are not immune to
such perturbations. While such fluctuations in momentum may be a relatively weak perturbation on
the Dirac electrons at high energy, near the Dirac point they are comparable to the average value of
the electrons’ momentum. Figure 4.7(c) indeed shows that the Dirac electrons exhibit such a shift
in momentum even close to the Dirac point, where their dispersion is perfectly linear. Using the
Fermi velocity obtained from QPI dispersion (v_F=1.3 eVÅ, in agreement with ARPES [37] ), we
relate this momentum shift near the Dirac point to an energy shift ( ΔE=v_FΔq ~16 meV), which
is consistent with the measured energy shift of the Dirac point illustrated in figure 4.5. Considering
that Δq/q is becoming larger close to Dirac, for these energies, in the presence of such fluctuations
the momentum carried by these electrons becomes ill-defined.

4.2.2 The interplay of nesting and spin texture

The detail analysis of the QPI patterns can be used to learn about scatterings present in these single
Dirac cone TI’s. The FT of the QPI patterns are presented in figure 4.8. Remarkably, the FT
patterns on various samples are independent of whether the scattering dopant is magnetic or not. In
fact, the strong resemblance of the patterns across dopants and materials Bi_2Te_3 and Bi_3Te_3 shows
that they can be understood based on the shape of the Fermi surface and the spin texture associated with it\cite{55, 48, 102}. At energies high above the Dirac point, we find six intensity peaks centered along the $\Gamma$-$M$ direction. As discussed below, they are associated with quasi-nesting conditions brought about by the hexagonal warping of the surface bands. At lower energies, the scattering patterns take a rather circular form, and are also consistent with the helical spin texture of the surface bands, which allows all scattering processes other than direct backscattering \cite{103}.

In order to trace the origin of the various intensity peaks seen in the FT of the QPI patterns, we display cuts taken along the $\Gamma$-$M$ and $\Gamma$-$K$ directions [figure 4.9(a) and (b)], respectively in Mn-doped Bi$_2$Te$_3$. The isotropic patterns we find at low energies [lower panels of figure 4.8] translate to symmetric modes in $\Gamma$-$M$ and $\Gamma$-$K$. They disperse linearly, and their extrapolation sets the Dirac point at $\sim$100 meV below Fermi. In contrast, the scattering mode we detect at high energies along $\Gamma$-$M$ is completely absent in the $\Gamma$-$K$ direction.

\textbf{Warped region.} The anisotropic QPI pattern at high energies stems from the anisotropic surface state’s band structure. ARPES measurements imply that at energies beyond $\sim$250 meV above Dirac the $\Gamma$-$M$ dispersion becomes sub-linear. As a result the band structure attains a snow-flake profile that can accommodate various quasi-nested scattering wavevectors both along the $\Gamma$-$M$ and
Figure 4.8: Fourier transforms of the QPI patterns from (a) Ca- and (b) Mn-doped Bi$_2$Se$_3$ and (c) Mn-doped Bi$_2$Te$_3$. All compounds show similar patterns in q-space consisting of six strong peaks along the Γ-M directions at high energies and circular patterns closer to the Dirac point at lower energies. Adopted from reference [95].
Γ-\bar{K} directions, indicated in figure 4.9(c) with green and blue arrows, respectively. Accordingly, they appear as corresponding strong peaks both along Γ-\bar{M} and Γ-\bar{K} in the joint density of states (JDOS) in figure 4.9(d) that identifies dominating scattering wavevectors by auto-correlating the band structure at a given energy [see chapter 2 for details]. The ARPES based dispersion of these dominating scattering wavevectors is given by solid lines in figure 4.9(a) and (b) (the dotted lines being their linear extrapolation to energies ARPES cannot access). Recall that in chapter 2, we showed that JDOS calculation, which ignores the spin texture of the surface bands, does not provide a satisfactory explanation for the measured FT of the QPI patterns. By calculating them again, we are emphasizing the fact that the observed patterns would have been substantially different if JDOS was the proper explanation.

We attribute the absence of the strong G-K peaks from the measured QPI patterns to the protection cast at these high energies by the full spin texture. Recall that the spin texture cant out of the plane for the hexagonally warped energies [the grey symbols in figure 4.2(c)]. The out-of-plane spin component provides additional protection against backscattering in the -K direction alone. Accordingly, in the calculated spin-selective scattering probability (SSP) [figure 4.9(e)], which is the JDOS with the consideration of the spin selection rules, the Γ-\bar{K} nesting peaks are absent, and only that along Γ-\bar{M} remains. The calculated SSP looks identical to the QPI pattern we find at high energies [see top rows of figure 4.8]. Naturally, the out-of-plane spin component vanishes at low energies together with warping, rendering the spin-selection rules isotropic in accordance with the circularly symmetric QPI pattern we encounter at low energies.

**Conic region.** Much more intriguing is the behavior at lower energies, close to the Dirac point, where the dispersion becomes truly Dirac-like. In this regime there are no nested wavevectors and helicity forbids back-scattering. Previous studies found no sign of scattering close to the Dirac point that was attributed to the protection from backscattering [52, 49]. In contrast, we find clear evidence for scattering well within the conic regime of the dispersion in the form of circularly symmetric QPI patterns that disperse with energy, as it is evident from the lower panels of figure 4.8. Their dispersion as E=v_Fq, with v_F=2.1 eV.Å, agrees with that found in angular resolved photoemission studies [37]. Due to the protection from direct backscattering, we attribute the strong scattering of the surface states to all other channels that do not involve full reversal of the momentum and spin [red arrow in 4.9(c)]. Their superposition gives rise to a QPI pattern in the form of a disc, demonstrated in the calculated SSP shown in panel (e). Those channels, like the one exemplified in middle part of panel (f), merely become attenuated by the partial overlap of the incident and scattered spin states. The direct visualization of those scattering processes within the Dirac cone

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Figure 4.9: **Origin of the QPI patterns.** The Full dispersion of the scattering wavevectors in (a) $\Gamma-M$, and in (b) $\Gamma-K$ direction, obtained from the Fourier transform of the d$I$/d$V$ maps taken on Mn-doped $Bi_2Te_3$ are shown in color. They agree with those measured on Ca-doped $Bi_2Te_3$ (circles) as well as with the dispersing q-vectors calculated based on ARPES (red, green and blue solid lines, and their linear extrapolation in dotted lines). (c) The contours of constant energy for hexagonally warped and conic energy ranges. The gray arrows and symbols show the spin texture of the contours. The red, green, blue, and green correspond to what is seen or absent in panels (a) and (b). (d), (e) The JDOS and SSP for hexagonally warped and conic energy regions. (f) Illustration of processes and their contribution to the QPI pattern: helicity forbids backscattering off non-magnetic impurities (left) but allows scattering and interference of all other channels that have a finite overlap between initial and final spin states (middle). A magnetic impurity allows spin-flip backscattering, but initial and final spin states remain orthogonal and thus do not interfere (right). Adopted from reference [95].
emphasizes the partial protection from disorder exhibited by the surface states in three dimensional topological insulators, hence their diffusive rather than ballistic mean of propagation through the disordered medium.

**Symmetry of the scattering potential.** One may have noticed that the asymmetric potential due to dopants, which can be approximated from their topographic appearances, is left out from the JDOS and SSP calculations. Here we briefly mention the role of triangular dopants in the observed QPI. In particular, The shape of the dopants once embedded in the lattice and their dimension as captured in the topographic image of figure 4.10(a) may also contribute to the suppression of the $\bar{\Gamma}-\bar{K}$ scattering peaks at high energies. The shape of the scatterer enters the JDOS as a pre-factor, 

$$\text{JDOS}(E, \vec{q}) = V(\vec{q}) \int I(E, \vec{k}) I(E, \vec{k} + \vec{q}) \, d^2\vec{k},$$

where $V(\vec{q})$ is the Fourier transform of the scattering potential $V(\vec{r})$ off a single dopant. Approximating $V(\vec{q})$ as a triangle of dimension $L$, demonstrated in figure 4.10(a), yields the Fourier-space pattern for $V(q)$ shown in figure 4.10(b). While the long wavelength scattering ($\lambda \gg L$) is insensitive to the details of the scatterer, the short wavelength scattering processes ($\lambda \ll L$) are greatly affected by the scatterer anisotropy. In particular, the $\bar{\Gamma}-\bar{K}$ scattering becomes suppressed relative to the $\bar{\Gamma}-\bar{M}$ direction. By superimposing the short wavelength (large $q$) anisotropic pattern of $V(q)$ over the warped JDOS, shown in figure 4.10(c), we find that it is exactly the $\bar{\Gamma}-\bar{K}$ scattering peaks of the JDOS that would be attenuated by the scatterer’s triangular shape. This is indeed the resulting JDOS once modified by the approximated $V(q)$ term, as demonstrated in figure 4.10(d). Finally, the crossover from isotropic-like long wavelength pattern to the anisotropic long wavelength one will occur at $q \sim \pi/L = 0.2 \, \AA^{-1}$ for $L \sim 15 \, \AA$. Such a wavevector would suppress $\bar{\Gamma}-\bar{K}$ scattering in the warped region of the band structure while leaving the scattering in the isotropic conic region unaltered in agreement with what we find in QPI. We thus find that both the spin-texture as well as the geometry of the dopants contribute to the suppression of the measured $\bar{\Gamma}-\bar{K}$ peaks in the Fourier-space QPI patterns.

### 4.2.3 Magnetic defects and line defects

Since the partial protection from scattering is assured by time reversal symmetry, magnetism is believed to have a dominant impact on the allowed scattering processes of the helical surface states. Once the impurity acquires a magnetic moment it can spin-exchange with the incident Dirac particle, and allow for spin-flip backscattering. Nevertheless, the FT of the QPI patterns on the magnetically doped compounds, $Bi_{1.95}Mn_{0.05}Se_3$ and $Bi_{1.95}Mn_{0.05}Te_3$, show striking similarity to those we find in the non-magnetic Ca-doped $Bi_2Te_3$ [figure 4.8 ]. Even the $\bar{\Gamma}-\bar{K}$ scattering wavevectors at
high energies, that are suppressed by the spin texture, remain absent in the magnetically-doped compounds. This may sound in contrast to common anticipation that magnetic scatterings and non-magnetic scatterings should yield very different QPI patterns. However, we note that although spin-flip backscattering should be facilitated in Mn-doped samples, in order for STM to be able to detect backscattering events they must also alter the LDOS through constructive interference. As long as the helical spin texture remains intact the orthogonality of the spin states prevents the backscattered states from doing so, as illustrated in the right part figure 4.9(f). Therefore, it is the remaining continuum of possible scattering channels, whose spin states are not fully orthogonal, that gives rise to the measured interference patterns, as was the case in Ca-doped Bi$_2$Te$_3$.

One may further speculate that breaking time reversal symmetry locally at Mn sites, or even more so globally within a ferromagnetic phase that Mn-doped Bi$_2$Te$_3$ indeed exhibits below $T_C$=10 Kelvin, can distort the spin texture and allow all channels to interfere (for magnetic characterization see Appendix B). However, the low ferromagnetic transition temperature indicates a gap on the order of only a few meV which opens at $E_D$. Such a small gap can degrade the spin texture only on an energy scale of its order. In contrast, spin-orbit coupling in these materials, which brings about the helical spin texture, has a much larger energy scale of several eV. Therefore, magnetism is too weak to account for the interference patterns we detect throughout the $\sim 250$ meV that span the conic region of the dispersion. In accordance, the QPI patterns measured at 2 Kelvin, far below $T_C$, are essentially identical to those taken above it.

Finally, we show that spatial modulations of the Dirac point as well as strong scattering within the Dirac cone are also induced also by crystallographic line defects and are not specific to point
defects. We have measured dI/dV maps next to naturally occurring crystallographic step-edges. Figure 4.11(a) shows a topographic image of an atomically ordered, 8 Å high, step-edge. The dI/dV spectroscopy shows a clear dispersing standing wave pattern at both at high and at low energies [figure 4.11(b) and (c)]. The modulations at high energies are strong, but they persist to the lowest energies, close to the Dirac point (E_D = -100mV, in both samples). Because the step edges are oriented perpendicular to the Γ-M direction, the scattered waves have their wavevector along the Γ-M direction. The Fourier transform of figure 4.11(c) finds two scattering modes, shown in figure 4.11(d). They agree with the dispersion of the scattering modes along Γ-M found both in the measured QPI as well as in the calculated SSP based on ARPES dispersion. The excellent agreement with the STM data confirms that the origin of the standing wave pattern at high energies is the quasi-nested scattering wavevector illustrated in panel (e) by green arrows. The standing wave at low energies has linear dispersion that extrapolates to the Dirac point. As for the low-energy mode we found in the QPI pattern on doped Bi₂Te₃, it also originates from the superposition of all scattering wavevectors allowed by the helical spin texture, such as the one indicated by red arrows. While the non-magnetic crystallographic step-edge does not support spin-flip backscattering, as illustrated in panel (f), it can still scatter all other channels that have a scattering wavevector perpendicular to it. The superposition of these yields the interference pattern within the Dirac cone. This is the first report of the visualization of the scattering within a single surface Dirac cone by step-edges, and demonstrates that the spin-texture of the bands are not modified by ferromagnetism, at the energy scales that we probed. Our measurements put an upper bound on the size of the anticipated gap at the Dirac point and scrambling of the spin texture due to ferromagnetism, which is about a few meV.

4.3 Conclusion

In this chapter we studied the response of topological surface states to perturbation due to charge dopants, which are commonly used in the bulk of the crystal. The bulk response to disorder (in both magnetically and non-magnetically doped samples) results in nanoscale spatial fluctuations of the band-structure of the topological surface states. Our results show that for energies far from the Dirac point, Dirac electrons show a remarkable robustness to scattering. To adjust to the underlying charge-puddles’ potential they continuously alter their crystal momentum. Furthermore, even in the presence of bulk magnetism, they preserve their helical spin texture; hence backscattering still remains absent.
In the vicinity of the Dirac point, the energy and momentum fluctuations we observed would result in spatially alternating helicity. This could possibly limit the mobility of topological surface state near the Dirac point. In fact, similar electron-hole puddles in graphene, detected using various local scanning probe techniques [101, 100], have been suspected as the origin of variability in mobility of different samples [104] and the minimum conductivity [105] in graphene when the chemical potential is tuned to the Dirac point. It is therefore natural to expect that fluctuation of the Dirac point in topological surface states reported here will also strongly influence transport properties. Recent experiments [42] have indeed shown magneto fingerprint associated with mesoscopic fluctuations in macroscopic samples of doped Bi$_2$Se$_3$. Analysis of these patterns has indicated an emerging length scale of a few tens of nanometers that originates from quantum interference of carriers in surface. Our results show that bulk disorder is likely to be responsible for the length scale observed in these experiments and suggest that doping methods used to tune the chemical potential are detrimental to transport properties of the topological surface states. The fluctuations reported here also suggest that utilization of helical Dirac fermions on topological insulators requires methods of tuning the chemical potential which do not involve chemical doping.
Chapter 5

Conclusion and outlook

Topological insulators, a newly discovered electronic phase of matter, have rapidly become one of the hottest topics in condensed matter physics. These materials have been named topological insulators because they are insulators in the bulk but have exotic metallic states present at their surfaces owing to topological order. From a theoretical prospective, their discovery introduced a new type of ordering of quantum matter. Many aspects of condensed-matter physics are concerned with understanding how order emerges when a very large number of simple constituents interact with each other. In ordered phases such as crystals and magnets, the order is described through symmetry breaking: in a crystal, ions are arranged periodically owing to their electrostatic interactions, thereby breaking the continuous symmetry of space under rotations and translations. The discovery of topological insulators highlights the fact that a new kind of order, topological order, can also occurs in material systems.

Due to strong spin-orbit coupling in 3D topological insulators, the direction of the electron’s motion on the surface uniquely determines its spin orientation. In the simplest realization of 3D topological insulators, the surface bands consist of only a single non-degenerate band of linearly dispersion surface states with helical spin texture. Stemming from these unique band properties, one of the most interesting properties of these surface states is the promise of being impervious to impurities and remaining immune to localization even in disordered samples. In this thesis we presented our STM results which provide evidence in support of this claim. However, STM is not the most suitable tool to fully demonstrate the immunity to localization, but more conventional approaches such as transport measurements suffer from the fact that there are contributions from the bulk of the material to the measured conductivity. At this point, a loophole-free demonstration
of protection against localization remains elusive, it demands carefully chosen TI compound, where
the binding energy of Dirac surface electrons is not overlapped with the binding energies of the bulk.
Nevertheless, the STM experiments we discussed in chapters 2, 3, and 4 demonstrating how the
surface states of TI’s are distinct from other surface states.

In chapter 2, we used STM to visualize the gapless surface states of the topological insulator
\( Bi_{1-x}Sbx \) and to examine their scattering behavior from disorder caused by random alloying in this
compound. Combining STM and angle-resolved photoemission spectroscopy, we showed that despite
strong atomic scale disorder, backscattering between states of opposite momentum and opposite spin
is absent. Our observation of spin-selective scattering demonstrates that the chiral nature of these
states protects the spin of the carriers. This is the most essential prerequisite for observing immunity
from localization. Given that scattering from defects, in particular direct backscattering, is among
main the factors which degrade the coherence of current flow, the surface states therefore have the
potential to be used for coherent spin transport in spintronic devices.

In chapter 3, we turned to the next essential question for coherent transport: how do surface states
behave close to atomic steps? We presented the results of measuring the transmission and reflection
probabilities of topological surface states of antimony through naturally occurring crystalline steps
separating atomic terraces. In contrast to the non-topological surface states of common metals
(copper, silver and gold), which are either reflected or absorbed by atomic steps, we show that
topological surface states of antimony penetrate such barriers with high probability. With a simple
model based on multi-scattering, we were able to calculate transmission of \( \sim 35\% \), reflection of \( \sim 42\% \),
and scattering to the bulk of \( \sim 23\% \). The fact that transmission is far from perfect should not be
discouraging. The antimony crystals are bulk metals, rather than insulators, and this causes some
‘leakage’ of the surface electrons into the bulk during the collision with a step. Also, antimony’s
surface energy-band structure is more complicated than the ideal helical metal, and thus allows
for scattering between states of opposite but non-equal momenta. These factors suggest that the
electron transmission probability through the steps can be further increased by using a material such
as \( Bi_2Se_3 \) and \( Bi_2Te_3 \), which can be tuned to become an insulator in the bulk and have a surface
energy-band structure that is consist of only a single band. This demonstration of the extended
nature of antimony’s topological surface states suggests that such states may be useful for high
current transmission even in the presence of atomic-scale irregularities—an electronic feature sought
to efficiently interconnect nanoscale devices.

In chapter 4, we studied the response of topological surface states to perturbation due to charge
dopants, which are commonly used in the bulk of the crystal. We considered both magnetically and
non-magnetically doped samples. The bulk response to disorder results in nanoscale spatial fluctuations of the band-structure of the topological surface states. Our results show that Dirac electrons show a remarkable robustness to scattering. To adjust to the underlying dopant induced potential puddles their continuously alter their crystal momentum. Furthermore, even in the presence of bulk magnetism, they preserve their helical spin texture; and hence backscattering still remains absent. The consequence of these spatial fluctuations for states close to the Dirac point is far reaching. In the vicinity of the Dirac point, the energy and momentum fluctuations we observed would results in spatially alternating helicity. This could possibly limit the mobility of topological surface state near the Dirac point. It is therefore natural to expect that fluctuation of the Dirac point in topological surface states reported here will also strongly influence transport properties. Therefore, utilization of helical Dirac fermions on topological insulators requires methods of tuning the chemical potential which do not involve chemical doping.

From our works and of others, now we have formed some understanding of what Dirac electrons do for energies away from the Dirac point. However, it is anticipated that the most interesting- and maybe very different- physics can take place at the Dirac point. Intuitively, this is expected since the Dirac point is a singular point in the band structure. For the experimentally known and studied TI compounds, the Dirac binding energy is either inside the bulk band or very close to the bulk band edge. As a result, they are not ideal for studying the intriguing physics of the Dirac point, due to overlap of the surface and the bulk states. Besides finding the proper compounds to show the desired band structure, the low density of states and the long wavelength of the surface states at Dirac, which requires very large field of views, make experiments close and at the Dirac point exceptionally challenging.

In the past few years, several interesting experiments have been carried on, but it seems more interesting experiments, which also demonstrate the potential applications of the TIs, are yet to come. Among them is the creation of Majorana fermions on the surface of topological insulators. These elusive fundamental particles have been discussed in particle physics for decades, though as yet there has been no definitive proof of their existence. In condensed-matter physics, Majorana fermions can occur as quasiparticles in certain special superconductors. It is a high priority in condensed-matter physics to engineer a Majorana fermion, in part because they could in principle be harnessed to make a fault-tolerant topological quantum computer. The race is on to come up with the best way to realize them. There is considerable motivation to make this happen, because in addition to the potential quantum-information applications, having an experimental handle on Majorana fermions would allow some of the most bizarre features of quantum mechanics to be
probed.

In only a few years since the initial explorations into topological insulators, the level of interest and activity has grown exponentially. There are now dozens of experimental groups around the world, along with countless theorists, studying all aspects of these materials. With this level of activity there is great hope that some of the ambitious proposals based on topological insulators can be realized, along with others that have not yet even been conceived.
Appendices
Appendix A

Scanning Tunneling Microscopy

The scanning tunneling microscope (STM) is arguably the most powerful tool to directly study the electronic structure of material systems with atomic resolution. STMs can reach the resolution of sub-Angstrom lateral resolution and pm depth resolutions. With this resolution, individual atoms within materials are routinely imaged and manipulated. The STM can be used not only in ultra high vacuum but also in water, and various other liquid or gas ambient, and at temperatures ranging from near zero Kelvin to a few hundred degrees Celsius. In 1981, Gerd Binnig and Heinrich Rohrer, working at IBM in Zürich produced the first working STM [106]. Two year later they demonstrated the power of the new technique by showing the 7x7 reconstruction of the atoms at the silicon (111) surface in real space for the first time [106]. In 1986, they were awarded the Nobel Prize in physics for their invention. STM works on the principle of quantum tunneling. A metallic tip is brought within several atomic distance of the sample, and the electron wavefunctions of the tip overlap with the wavefunctions of the sample. In this configuration electrons can tunnel form the tip to the sample and vice versa. Today STM is not only used for imaging the surface and measuring the electronic structure of the matter, but also for the manipulation of the surface and tailoring the properties of the matter. The STM image of the electronic standing waves of surface electrons inside a quantum corral has become the icon of quantum mechanics, and what STM can create and measure.

A.1 The tunneling current

Herein, we examine the case of tunneling in one dimension and consider the approach originally suggested by Bardeen [108]. Bardeen, instead of solving the Schrödinger equation for a coupled system of tip and sample, made clever usage of the perturbation theory. He first considered two
Figure A.1: The tip of the STM can be used to move atoms around. In this experiment [107], the tip was used to move Iron atoms on the Copper (111) surface to make a corral and measure the electronic density of states inside and outside the corral.

separate subsystems of the tip and the sample, where the electronic states can be found by using the time independent Schrödinger equation. To calculate the rate of transfer of electrons from one system to the other he used time dependent perturbation theory and showed that the amplitude of the electron transfer is determined by the overlap of the surface wavefunctions of the two systems. At the end, he showed that the transition probability of an electron from $\psi_\mu$ to $\chi_\nu$ is then given by the Fermi golden rule,

$$w_{\mu\nu} = \frac{4\pi^2}{\hbar} |M_{\mu\nu}|^2 \delta(E_\nu - E_\mu),$$  
(A.1)

where matrix elements are given by

$$M_{\mu\nu} = -\frac{\hbar^2}{2m} \int (\bar{\chi}_\nu \nabla \psi_\mu - \psi_\mu \nabla \bar{\chi}_\nu) dS$$  
(A.2)

Here, $\psi$ and $\chi$ are the wavefunctions of the sample and the tip before juxtaposing them, respectively. Appearance of the Kronecker delta function makes it clear that we are confined to the elastic tunneling. The validity of such restriction was always an open question and subject of some detailed studies [Ref.?]. Thus, accepting this restriction, the total current would be proportional to the density of states available, the Fermi distribution function, and the probability of transferring from one state on one side to an state of the same energy on the other side [ figure A.2 ] . In the case of positive voltage bias applied to the sample we have:
To decipher useful information from STM experiments, some simplifying assumptions are common to be used. The matrix elements are usually assumed to be constant and not changing from one energy level to the other. Since most of the tips used are metallic, the density of the states of the tip is fixed. Given the fact that experiments are usually carried at 4 Kelvin, Fermi distribution function can be replaced by a Heaviside step function. After applying these assumptions the last equation becomes:

$$I \approx \frac{8\pi^2e}{h} |M|^2 \rho_T(0) \int_0^{eV} \rho_S(\epsilon) \, d\epsilon. \quad (A.4)$$

Furthermore, Bardeen approximated the vacuum potential barrier with a square, and concluded that the wave functions decay exponentially inside this vacuum barrier. The reason for this approximation is that in reality there is a tilt in the top of the barrier due to the electrical field between the tip and the sample. The simplified problem of finding the probability of tunneling through a square of width $z_0$, and potential height of $V_0$, is a solvable one and the final answer is:

$$|M|^2 = e^{-2\gamma} \quad (A.5)$$
\[ \gamma = \frac{2\pi z_0}{h} \sqrt{2mV_0} \]  
(A.6)

One can substitute this into equation (A-4) and relate the current and the density of states:

\[ I \propto e^{-z_0 \sqrt{\frac{2m}{h^2} V_0}} \int_0^{eV} \rho_S(\epsilon) d\epsilon. \]  
(A.7)

A.2 Experimental realization and measurement modes

In STM, a tunneling current is maintained between a sharp metallic tip and a sample, while the tip is held in the angstrom range above the sample. To achieve the desired control over its position, the tip is placed on a piezoelectric tube, which can extend or contract in response to applied voltage. The piezoelectric material provides an essential ingredient for STM, namely the ability to move the sample and the tip with angstrom accuracy. There are four electrodes connected to the four quadrants of the piezoelectric tube, and by applying voltages to the different quadrants the tube can be bended or made to vary in length [figure A.3]. By electrically biasing the sample with respect to the tip, electrons will tunnel into the empty states of the sample in the case of positive bias, and tunnel from the occupied states of the sample to the tip in the case of negative bias.

**Topography.** One of the most common modes of measurement in STM is constant current mode, also known as topography. A STM topography is obtained by maintaining the tunneling current between the tip and the surface fixed. In this case, a constant voltage \( V_s \) is applied to the sample, and a constant current is demanded by user \( I_{set} \). As the tip scans over the surface, its piezoelectric tube extends and contracts to keep the flow of current fixed, and the height for which \( I_{set} \) was achieved is recorded. For scanning a metallic surface the values of \( V_s \) could be as low as few tens of millivolts, and the current as high as 1 nAmp [see figure A.3(a)]. The image represents a contour of constant tunneling current on a surface, approximating the integrated LDOS. Considering only elastic tunneling, the electrons with energies between Fermi energy and \( V_{bias} \) can tunnel from the filled states of the sample to the tip in the case of negative bias, and from tip to the empty states of the sample in the case of positive bias.

**dI/dV.** Another STM mode of measurement is measuring the differential tunneling conductance \( dI/dV \), which is directly proportional to the LDOS. The measurement can be done at a single point is space or over an area of the sample. A general technique to obtain energy spectrum is to add
Figure A.3: The Schematic of the STM operation. By bringing the tip (shown in red) close to the sample (shown in blue), electrons can tunnel between the two. The voltage bias applied determines the direction of the current. Applying positive voltage to the sample gives image of the empty states, and maps with negative bias applied to the sample contribution of the filled states are depicted.
some oscillatory voltage \( (dV) \) on top of the bias voltage and measuring the response \( (dI) \) by using a lock-in amplifier. Thus, \( dI/dV \) for a particular value of energy can be measured and from equation (IV) it can be seen that it is proportional to density of states:

\[
g(V) \equiv \frac{dI}{dV} \propto DOS(V).
\] (A.8)

The measurement of this differential conductance as a function of energy and space is the key measurement for obtaining energy-resolved information on the electronic structure.

**Atomic manipulation.** A Because the STM tip is metallic with a sharp point at the apex, the electric field concentration near the end of the tip could be sufficient to move atoms and molecules on a surface with atomic precision. The methods used to position atoms on a surface depend largely on the substrate and the bonds between the adsorbed atoms and the substrate. In general, manipulation is accomplished by the electrical field of the tip in close proximity to an adsorbed atom or by temporary bonding of the tip and atom to redeposit the atom in a desired location.

The STM topographies were obtained in constant-current mode, and \( dI/dV \) spectroscopy was measured by a standard lock-in technique with \( f = 757 \) Hz, an a.c. modulation of 3 mV added to the bias voltage, and the feedback loop disabled during the measurement. Setting \( dI/dV \) maps to resolve the desired range of \( q \)’s may take several attempts. The spatial resolution during the \( dI/dV \) mapping for the maps analyzed in chapter 2 was about 2 Å, which provides the capability to resolve \( k \)-vectors up to twice the first Brillouin zone in momentum space. Here are some suggestions. To resolve modulations with large wavelengths \( \lambda_L \), the map should be at least 8 to 10 times larger than \( \lambda_L \). This criterion sets the size of the map. To resolve wavelengths down to \( \lambda_s \), the resolution should be \( \lambda_s/6 \) or \( \lambda_s/8 \). This criterion is related to the Nyquist theorem, but taking into account the decoherence of the quasi-particles, and sets the resolution of the maps. It is a good practice to be sure that the resolution is such that the atomic peaks are included in the Fourier transform of the \( dI/dV \) maps. They can be used to provide the most accurate calibration. In addition, the deviation of these atomic peaks from a perfect hexagon can be used as a measure of the thermal drift, which for the results presented was negligible, allowing us to symmetrize the Fourier transform of the maps without smearing out features or creating artificial ones.
Figure A.4: Basic STM measurement modes. (a) Topographic images. An electrical feedback loop move the tip to maintains a constant tunneling rate. (b) The dI/dV measurement. By using lock-in techniques, the tunneling rate at a given energy is measured, which is proportional to the local density of states. (c) the manipulation mode. A pulse send to the tip can move the atoms on the surface or take them out or place them in the crystal lattice. (d) STM topography over the surface of Zn-doped GaAs shows the Zn-dopants. The dopants appear as bright triangular protrusions. The change in their intensity is indicative of the fact that they are located in different layers bellows the surface. (e) The dI/dV measurement on the surface of Zn-doped GaAs shows the density of states due to valence band states and the conduction band states. A semiconductor gap of 1.5 eV is separating them. (f) IBM scientists demonstrated the power of STM by arranging 48 iron atoms on the surface of a copper substrate. These images show the various stages of the process. Panel (f) is adopted from reference [107].
A.3 Experimental procedure

Vibration isolation. Isolation from sources of mechanical vibration is vital for the success of any STM experiment. Consequently, it is essential to isolate the microscope from the vibration of the floor, as well as the ambient acoustic noise in the lab. The construction of the experimental space was aimed to achieve a high level of vibration isolation. The microscope is supported by an optical table, and the table is placed on a massive concrete slab, which weights approximately 30 tons. Acoustic walls shield the system from outside and they are seated on the concrete slab. The acoustic walls and concrete floor are floated on six pneumatic isolation legs, and is located inside another room made of sand filled cinderblock walls.

The piezoelectric materials, due to their capability to expand and contract consistently and sensitively in the presence of electric fields, provide reliable motion at sub-atomic scales. With the added benefit of good vibration isolation, a metal tip can be positioned less than one nanometer away from a surface in a fixed position with picometer stability. Figure A.6 shows a typical STM head used in our microscope. This microscope design is based off the Besocke STM head, named after its inventor [109]. The piezoceramic tubes have electroplating divided into 4 quadrants on the outside and one on the inside. The inside quadrant gives overall elongating or shrinking of the tube length-z motion, and the four outer quadrants give shearing motion to the tube along a perpendicular plane-x-y motion. The three outer tubes, the STM legs, are used to step the sample towards or away from the tip by walking a ramped sample holder up or down. The center tube, the scanner, holds the tip and is used to raster the tip over the surface while responding to the feedback control.

Preparation of the tip and sample may involve different procedures depending on the materials used; some general actions are discussed below. To avoid any complication due to variation in the DOS of the tip or having a multiple-ended tip, where tunneling can happen through multiple points, one needs a metallic tip, optimally with one atom protruding from the end of it. Tip preparation starts with field emission, which is applying a high voltage to the tip. Consequently a high electrical field is generated at the end of the tip, which can drastically move electrons and atoms around. During field emission, electrons tunnel with a high rate, and atoms can be detached from the tip and deposited on the sample. To check the quality after the field emission, the tip is used to scan a known metallic surface such as crystals of Cu, Ag, or Au.

Every surface exposed to atmospheric pressure accumulates a monolayer of undesired atoms in a few minutes. To remove these atoms from the surface of the crystal after placing the metallic sample
inside the Ultra High Vacuum (UHV) chamber, sputtering and annealing are done repeatedly. In the sputtering process, Argon gas is released into the chamber, and an ion gun is used to accelerate them to bombard the surface of the sample. \( \text{Ar} \) gas is a noble gas, and it does not have any tendency to form bonds with the surface especially at high temperatures. After sputtering for about 15 minutes we are left with a rough surface with all the undesired atoms removed. To achieve a rather flat surface, the sample is heated up to increase the mobility of the atoms, and achieve an atomically ordered surface. This process is called annealing and during this process the temperature of the sample may reach few hundreds of Kelvin. These processes maybe repeated for a couple of cycles and after the last annealing, the sample is moved to the cold part of the chamber. The sample needs about a day to reach thermal equilibrium and after that one can bring the tip to the atomic distance of the sample.
Figure A.6: Topographic image of Silver (111) surface. The surface of silver was used for the preparation of the tip. The bias voltage between the tip and the sample was $V_{set} = 10 \text{ mV}$, and the current was $I_{set} = 200 \text{ pAmp}$. The width and the length of the image are 500\AA. The step edges, and intrinsic impurities can readily be seen, as well as, the interference of the electronic surface states as they scatter from the atomic step edge or a point defect.
Appendix B

Doping $Bi_2Se_3$ and $Bi_2Te_3$

The results presented in chapter 4 demonstrate our capability to tune the physical properties of the topological insulators $Bi_2Te_3$ and $Bi_2Se_3$ by doping. In this appendix, we discuss how substitutional dopants are seen in STM topographies and presenting the magnetization measurements [110] showing the ferromagnetism in magnetically doped $Bi_2Te_3$. This appendix is mainly adopted from our published results in reference [43] and [110].

B.1 The crystal structure and the native defects

In this section we discuss the native defects, and chemical doping of $Bi_2Te_3$ with $Ca$. The presence of native dopants leads to n-type semiconductor in $Bi_2Te_3$. It is desired to add other dopants to tune the chemical potential away from the bulk bands, in this case the conduction band. This goal was achieved by doping $Bi_2Te_3$ with $Ca$. As discussed in detail in chapter 4, this approach to tune the chemical potential may not be the ideal one when it comes to transport, especially when the substitutional dopants are a substantial fraction of the crystal elements, say more than 2-3%. Nevertheless, our results demonstrate that STM can be used to learn about defects in the crystal and their spatial distribution.

The $Bi_2Te_3$’s rhombohedral crystal structure consists of hexagonal planes of $Bi$ and $Se$ stacked on top of each other along the [111] crystallographic direction, with the atomic order: $Se(1)$-$Bi$-$Se(2)$-$Bi$-$Se(1)$, where (1) and (2) are refer to different lattice positions [ figure B.1(a) ]. The unit cell consists of three of these units stacked on top of each other with weak van-der-Waals bonds between $Se(1)$-$Se(1)$ layers [111], making the (111) plane the natural cleavage plane. We were able to identify various defects and their charge states from STM topographies of the filled states and
unoccupied states. The STM topographies of $Bi_2Se_3$ (111) surface are dominated by one type of defect, which appears as a bright triangular protrusion in the topographies of the unoccupied states [figure B.1(b)]. These defects are about 40Å in size on average and vary in size between defects indicating that they are located in various layers beneath the surface. It is a well-known fact that the equilibrium growth of $Bi_2Te_3$ leads to formation of Se vacancies that act as electron donors, and resulting in n-type semiconductors. Given that no other defects were observed, we attribute these triangular defects to Se vacancies. Figure B.1(c) and (d) show topography of the unoccupied and occupied states of the Ca-doped $Bi_2Te_3$, respectively. Comparison between B.1(b) and (c) makes it clear that the density of triangular defects is reduced significantly in Ca-doped samples.

In addition, the STM topography of the $Bi_{1.98}Ca_{0.02}Se_3$(111) surface shows distinct defects which were not present in $Bi_2Te_3$ samples. In topographic images of the empty states, the shapes of these three-fold symmetric defects resemble a clover leaf [figure B.1(c)]. Based on their two distinct spatial extents we concluded that they are located at two different crystallographic positions, most likely the smaller one is located in a layer nearer the surface and other one in a layer deeper beneath the surface. Considering the observation of these defects only in Ca-doped samples, we identify them as Ca-related defects.

The charge state of these defects can be inferred by observing the bending of the host bands caused by the Coulomb field surrounding a charged defect. A positively charged defect lowers the electronic energy level in its neighboring region, leading to a depression area in the STM topography of the filled states, and an enhancement in the topography of the unoccupied states. This is expected to be observed in imaging the ionized donors, and the opposite effect for negatively charged defects, such as acceptors. The clover leaf shape defects are surrounded by a region of enhancement in the topography of the filled states [figure B.1(d)] implying they are negatively charged, consistent with a Ca acceptor. A comparison between the triangular defect close to the center of the image in figure B.1(c) and (d), shows a region of depression in the topography of the filled states (d), and hence implies the presence of a positively charged defect. This observation is expected for Se vacancies, which are known to be electron donners and in their ionized state become positively charged.

### B.2 Doping induced ferromagnetism in $Bi_2Te_3$

The presence of the magnetic impurities breaks the time reversal symmetry locally in the crystal, and if the moments form an order phase, such as ferromagnetism, they break this order globally. It is commonly discussed that scattering and localization of the single Dirac cone topological insu-
Figure B.1: (a) Layered crystal structure of Bi₂Se₃ with quintuple layers ordered in the Se-Bi-Se-Bi-Se sequence along the c axis. 500 Å by 500 Å STM topographic images of the (b) Bi₂Se₃ and (c), and (d) Bi₁.₉₈Ca₀.₀₂Se₃ (111) surface. (b) STM topography of the empty states of Bi₂Se₃ (V_B=+1V, and I=10pA) showing triangular shaped defects observed at various intensities, indicating they are located in different layers beneath the surface. (c) Topography of the empty states of Bi₁.₉₈Ca₀.₀₂Se₃ (V_B=+2.0V, and I=10pA) shows clover leaf looking defects and a substantial reduction in the density of the triangular defects, which dominated the undoped samples. (d) Topography of the filled states over the same area as in (c) (V_B=-1.0V, and I=10pA). The area around the triangular defect near center shows a depression around it in the filled states, demonstrating it is positively charged. In contrast, the Ca-related defects exhibit an area of enhancement around them, indicating they are negatively charged. Adopted from reference [43].
lators would be different for these two cases in comparison to cases where the crystal is free from magnetic impurities. Different aspects of this claim are discussed in chapter 4, where we presented result above and below ferromagnetic phase transition, and compared them with non-magnetically doped samples. Here we present magnetization measurements, and show STM topographies that demonstrate the proper substitution of Mn-dopants in the $\text{Bi}_2\text{Te}_3$ crystal structure by showing the result of a statistical analysis. This analysis may seem pedantic and unnecessary, but given the high level of dopant concentration, they are essential for knowing the composition of the crystal close to the surface, and demonstrating that the dopants are distributed randomly and there is no cluster or other phases in the crystal. Such analysis or similar ones are surprisingly missing from some recent literature, where erroneous claims were made based on limited knowledge of chemical composition.

The temperature-dependent magnetic susceptibilities, $\chi=M/H$, are shown in figure B.2(a) for the $\text{Bi}_{2-x}\text{Mn}_x\text{Te}_3$ crystals for $x=0$, 0.005, 0.01, 0.02, 0.04, and 0.09, measured with applied magnetic field (H) perpendicular to the c axis. There is no indication of a ferromagnetic transition in the susceptibility measurements for $x<0.04$. The system becomes ferromagnetic when the Mn concentration is increased to $x=0.04$, evidenced in these data by a relatively large susceptibility at low temperatures. The inset of figure B.2(a) emphasizes the low temperature susceptibilities for the series. The susceptibility $\chi$ can be fit to the Curie-Weiss law, $\chi_0=\chi/(T-\theta)$, where $T$ is the temperature-independent term, $C$ is the Curie constant, and $\theta$ is the Weiss temperature. Figure B.2(b) shows the low-temperature inverse susceptibility plots, $1/(\chi-\chi_0)$ vs. $T$, for all samples. The inverse susceptibilities for $x=0.005$, 0.01, and 0.02 show straight line behavior, as expected for the presence of weak local moments due to the Mn dopants. For the lowest $x$, a very small negative $\theta$ is observed, which becomes slightly more positive with increasing Mn content. In contrast, data for crystals of composition $x=0.04$ and 0.09 follow the Curie-Weiss law with positive $\theta$'s of 11 and 13 Kelvin, respectively.

In order to directly visualize the effects of Mn doping in $\text{Bi}_2\text{Te}_3$ at the atomic scale, we have measured the $\text{Bi}_{2-x}\text{Mn}_x\text{Te}_3$ (111) surface with STM above and below its Curie temperature. Due to the weak van-der-Waals bonding between the quintuple layers of this rhombohedral structure the uppermost layer exposed after in situ cleavage is a triangular lattice of Te atoms. A typical STM topographic image of the surface of $\text{Bi}_{1.9}\text{Mn}_{0.1}\text{Te}_3$ is presented in figure B.3(a), where the black triangles are identified as substitutional Mn. Figure B.3(b) and (c) show zoomed-in views over a triangular suppression associated with Mn dopants, in which the position of Te atoms on the topmost layer are marked by blue circles. Below the surface Te layer, The Bi atoms form an inter-penetrating triangular lattice (presumed sites denoted by red circles) which are located in the
Figure B.2: (a) Zero-field cooled (ZFC) temperature dependent dc magnetic susceptibility measured at 1 kOe applied magnetic field for the Bi$_{2-x}$Mn$_x$Te$_3$ crystals. The magnetic susceptibility in the region of 0 to 30 K is shown in the inset. (b) Temperature dependence of the inverse susceptibility for x=0.005, 0.01, 0.02, 0.04, and 0.09 of Bi$_{2-x}$Mn$_x$Te$_3$. Adopted from reference [110].
middle of the triangle formed by top Te atoms. The triangular suppression that extends over several lattice sites is in the same spot as a Bi atom is expected to be seen; therefore, we conclude that Mn substitutes mainly for Bi upon doping. In the topographies of the filled states, as shown in figure B.3(c), Mn dopants are also centered on the Bi site forming a smaller region of suppression and a bright halo signifying enhanced LDOS. We attribute the formation of this bright region to local band bending due to negative charge that localizes about the dopant Mn ion, and is expected to be seen around substitutional donor atoms. In addition to Mn dopants in the Bi layer, faint clover-like patterns are also visible in topographies which could be substitutional Mn atoms on deeper Bi layers, and their accurate identification demands further studies. Counting all the substitutional Mn in the top most Bi layer gives a concentration about an order of magnitude less than what is expected from the given nominal concentration. The number of contrast features associated with Mn atoms seen in the top most Bi layer appears to be significantly less than that expected from the measured Mn concentration; the reason for this is not known, but we speculate that the reduction of Mn concentration has resulted from the diffusion of the Mn dopants away from the surface layer after the cleavage. Such a diffusion process may be enhanced by the electric field which is created at the surface after cleaving [112]. The implication of this scenario is that Mn may be intercalated between the $\text{Bi}_2\text{Te}_3$ layers in addition to substituting for Bi in the crystal lattice.

The STM topographic images can also be used to study the spatial distribution of Mn dopants. Given the high concentration of Mn in ferromagnetic samples of $\text{Bi}_{2-x}\text{Mn}_x\text{Te}_3$, the plausible formation of Mn clusters could signify a completely different mechanism for the observed ferromagnetism. By locating the position of substitutional Mn, STM provides a direct visual method of examining tendencies toward cluster formation from atomic scales to sub-micron scales. As discussed above, substitutional Mn atoms in the closest Bi layer to the surface appear as triangular suppression in topographic images. After finding the position of the individual Mn dopants in one of such Bi layers, correlation between Mn pairs were calculated for large fields of view ( $>1000$ Å). The plot presented in figure B.3(d) is the cumulative probability of random pair separation; i.e., for a given value of r, $P(<r)$ is the probability of two randomly chosen Mn dopants being less than a distance r from one another. For homogeneously distributed dopants, this probability scales with the area ( $r^2$ ), and the dopants fill the field of view, leaving no void area. The measured probability shows a power law behavior with a power very close to 2 for an extended range of distances ( $10\text{Å} < r < 200\text{Å}$ ), indicating the absence of clustering in these compound for concentrations as high as $x=0.1$. In contrast to a single exponent observed here, the tendency toward cluster formation would result in different exponents at different length scales, starting with values higher than 2 in small scales and
smaller than 2 for large scales. Five topographies from randomly chosen areas of the surface several hundreds of micron apart were also analyzed and yielded similar results.
Figure B.3: STM topographic image of $Bi_{1.9}Mn_{0.1}Te_3$, identifying the atomic substitutional site of Mn dopants, and the absences of Mn clustering. (a) STM topograph (+250 meV, 40 pA) of the $Bi_{1.9}Mn_{0.1}Te_3$ (111) surface over a 1000 Å by 1000 Å area. Substitutional Mn atoms appear as triangular suppressions of the LDOS. (b) and (c) Zoom-in topographies over Mn dopants in an area of 30 Å by 30 Å for unoccupied (+500 mV, 30 pA) and filled states (-500 mV, 30 pA). The position of top Te layer, Bi layer, and substitutional Mn are shown by blue, magenta, and green circles on the topographies. (d) Study of the possibility of clustering by calculating the probability of correlation between Mn pairs in different locations. For a randomly chosen pair of Mn, $P(< r)$ gives the probability of the pair having distance less than r. The calculated probability scales very close to $r^2$ (solid line) for an extended range of distances, demonstrating the uniform distribution of Mn dopants. Adopted from reference [110].
Appendix C

Critical correlations near the metal-insulator transition in \( Ga_{1-x}Mn_xAs \)

During My PhD years, I have also studied metal-insulator transition in dilute magnetic semiconductors \( Ga_{1-x}Mn_xAs \). The results, including a short introduction to these crystal systems, is included in this appendix. This appendix is mainly adopted from our published results in reference [113].

C.1 Introduction

Since Anderson first proposed that disorder could localize electrons in solids fifty years ago [27], studies of the transition between extended and localized quantum states have been at the forefront of physics [85]. Realizations of Anderson localization occur in a wide range of physical systems from seismic waves to ultracold atomic gases, in which localization has recently been achieved using random optical lattices [114]. In electronic systems, the signatures of localization have long been examined through electrical transport measurements [28, 29], and more recently using local scanning probe techniques that have imaged localized electronic states [115, 116]. For non-interacting systems, the electronic states at the mobility edge are predicted to have a diverging localization length with scale independent power-law characteristics, which are described as being multifractal [117]. Given the poorly understood nature of the metal-insulator transition in the presence of disorder and electron-electron interactions, direct imaging of electronic states can provide insights into the
interplay between localization and interactions.

We report on scanning tunneling microscopy (STM) and spectroscopy studies of electronic states in the dilute magnetic semiconductor Ga$_{1-x}$Mn$_x$As, over a range of Mn concentrations near the metal-insulator transition ($x=1.5-5\%$). Over the last decade, Ga$_{1-x}$Mn$_x$As has emerged as a promising material for spintronic applications with a high ferromagnetic transition temperature [118, 119]. Mn atoms substituted at Ga sites act both as acceptors that drive the metal-insulator transition and as localized spins that align at low temperatures to give rise to magnetism. The nature of the electronic states underlying magnetism in these heavily doped semiconductors is still debated. It is often assumed that the carriers that mediate magnetism in Ga$_{1-x}$Mn$_x$As are Bloch states associated with either the valence bands or extended states originating from an impurity band [120, 121, 122]; however, the validity of these assumptions has been questioned [123, 124]. Moreover, many recent low temperature transport studies show evidence of electron-electron interaction and weak localization of carriers even for samples with high doping levels [125, 126, 127]. We use atomic scale imaging and high-resolution spectroscopy with the STM to visualize electronic states in Ga$_{1-x}$Mn$_x$As and to examine the spatial structure of electron-electron correlations in this system. Our results indicate that spatial heterogeneity and electronic correlations must be considered in understanding the mechanism of magnetism in highly doped semiconductors.

C.2 STM results

Figure C.1 shows STM topographs of cleaved Ga$_{1-x}$Mn$_x$As samples (2000 Å thick) grown using molecular-beam-epitaxy (MBE) on a p-type Be-doped GaAs buffer layer [128]. Prior to measurements at a temperature of 4.2 K, the degenerately doped substrates are cleaved in situ to expose a (110) or equivalent surface of the heterostructure in cross section. (Fig. C.1(A) inset) The topographs show in-gap states, dominated by individual Mn acceptor wavefunctions, although other defects such as As antisites are observed as well. Using previous STM studies of samples with more dilute Mn concentrations [129, 130] and the results of tight-binding model calculations [131, 132], we can identify the topographic signatures of individual Mn acceptors in layers from the surface to the 3rd subsurface layer (Fig. C.1(B)). The size of an individual Mn acceptor state wavefunction is about 20 Å, due to its deep binding energy, which results in a metal-insulator transition at a relatively high level of doping (between 1-2%) in Ga$_{1-x}$Mn$_x$As. Increasing the Mn concentration from weakly insulating samples with variable-range hopping resistivity at 1.5% ($T_C=30$ K) [133] to relatively conducting samples at 5% ($T_C=86$ K, annealed), we find higher concentrations of dopants appear
Figure C.1: STM topography of the in-gap states of $Ga_{1-x}MnxAs$. (A) STM topograph (+1.5 V, 20 pA) of $Ga_{0.985}Mn_{0.015}As$ over a 1000 Å by 500 Å area. The inset shows a topograph (+1.8 V, 20 pA) of the heterostructure junction between the Mn-doped and Be-doped layers of GaAs. The size of this area is 80 Å by 125 Å. (B) A topograph (+1.5 V, 30 pA) over a smaller area of size 150 Å by 150 Å of $Ga_{0.985}Mn_{0.015}As$. Several substitutional Mn’s in various layers are identified in the lower part of the panel and marked by which layer they are in, with the surface labeled as zero. An As anti-site is also shown. (C) STM topograph (+1.5 V, 20 pA) of the $Ga_{0.95}Mn_{0.05}As$ sample after annealing for 6 hours at 200 ºC. The size of the area is 150 Å by 195 Å. (B) and (C) have the same scale. Adopted from reference [113].
in STM topographs on top of the atomically ordered GaAs lattice (Fig. C.1(C)). All characteristic lengths, such as dopant separation or mean free path ($\sim 10 \, \text{Å}$), are much shorter for $Ga_{1-x}Mn_xAs$ in comparison to other semiconductors doped with shallow dopants [124].

Spectroscopic mapping with the STM can be used to show that Mn acceptors and other defects give rise to atomic scale fluctuations in the local electronic density of states (LDOS) over a wide range of energies. Figure C.2(A) shows tunneling spectra of states from within the valence band ($V<0$) to the conduction band edge ($V>1.5$), measured along a line perpendicular to the buffer layer-film interface. Contrasting the spatial dependence of electronic states in the buffer layer (with $2 \times 10^{18}$ per cm$^3$ Be acceptors) to those of $Ga_{1-x}Mn_xAs$ (with $x=0.015$) in Fig. C.2(A), we find the Mn-doped region to have strong spatial variations in the electronic states at the valence and conduction band edges and a broad distribution of states within the GaAs band gap. Increasing the Mn concentration gives rise to a larger number of in-gap states (compare Fig. C.2(B) and C.2(C)). These features of the local electronic structure of $Ga_{1-x}Mn_xAs$ are difficult to reconcile with a weakly disordered valence or impurity band picture, and show the importance of compensation and disorder in this compound. The Fermi energy $E_F$ lies within the range of electronic states that are spatially inhomogeneous.

In addition to strong spatial variations, electronic states of $Ga_{1-x}Mn_xAs$ are influenced by electron-electron interactions (Fig. C.2(D)). STM spectra, spatially averaged across large areas for several samples with increasing doping levels, illustrate a strong suppression of the tunneling density of states near $E_F$. The evolution from weakly insulating (1.5%) to relatively conducting samples (5%) is well correlated with the increase in the density of states at the Fermi level, yet a suppression centered at $E_F$ is observed at all doping levels. This feature is indicative of an Altshuler-Aronov correlation gap that is expected to occur in the tunneling density of states of a disordered material on the metallic side of the phase transition [134], appearing as a square root singularity in the conductance near $E_F$ (Fig. C.2(D)). Previous spectroscopic measurements of $Ga_{1-x}Mn_xAs$ using macroscopic tunneling junctions have also reported similar correlation gaps [125].

C.3 Proximity to the metal insulator transition

To determine whether there are any specific length scales associated with the spatial variation of the LDOS in $Ga_{1-x}Mn_xAs$, we now examine energy-resolved STM conductance maps. In Fig. C.3, we show examples of such maps at different energies relative to $E_F$ for the 1.5% doped sample. These maps show that, in addition to modulations on the length scales of individual acceptors,
Figure C.2: Local dI/dV spectroscopy for various dopings. (A) The differential conductance (dI/dV) (ΔV=10 mV) across a line normal to the growth surface of length 1000 Å. The first 200 Å is the Be-doped GaAs buffer layer, and 800 Å of 1.5% Mn-doped GaAs follows. The junction between the two is marked by a dashed line. (B), (C) The dI/dV spectra (ΔV=5 mV) for energies close to Fermi level, across a line of length 450 Å over the 1.5% sample (B), and the 5% sample (C). Top of the valence band and the in-gap states are shown. (D) The spatially averaged differential conductance for several samples. The inset shows the same data as the main panel, with the square root of the voltage on the horizontal axis. Adopted from reference [113].
Figure C.3: Spectroscopic Mapping in $Ga_{0.985}Mn_{0.015}As$. The differential conductance ($dI/dV$) measurements over an area of 500 Å by 500 Å show the spatial variations in the LDOS for the states in the valence band as well as states deep inside the semiconductor gap: (A), -100 mV; (B), -50 mV; (C), 0 mV; (D), +50 mV; (E), +100 mV; and (F), +150 mV. Adopted from reference [113].
Figure C.4: Correlation length for Mn dopings close to the metal-insulator transition. The autocorrelation was calculated from LDOS maps and is presented on a logarithmic scale for (A) Ga$_{0.985}$Mn$_{0.015}$As and (B) Ga$_{0.95}$Mn$_{0.05}$As. The dashed line is $\sim (E-E_F)^{-1}$ and is a guide to the eye. (C) The autocorrelation for the d$I$/d$V$ map at the Fermi level of Ga$_{0.985}$Mn$_{0.015}$As, as well as one valence band (-50 mV) and one in-gap energy (+50 mV) are plotted. The inset shows the autocorrelation for the same energies on a semi-logarithmic scale. Adopted from reference [113].
there are spatial structures in the local density of states with longer length scales. To characterize these variations, for each conductance map, we compute the angle averaged autocorrelation function between two points separated by $r$, 

$$C(E, r) = \frac{1}{(2\pi)} \int d\theta \int d^2r'[g(E, r') - g_0(E)] \times [g(E, r' + r) - g_0(E)] ,$$

in which $g(E, r)$ is the local value of the differential tunneling conductance that is proportional to the LDOS, $g_0(E)$ is the average value of the conductance at each energy $E$. Displaying $C(E, r)$ in Fig. C.4(A), we find a dramatic increase of the long-distance correlations near $E_F$. At this energy, the correlations remain measurable to length scales well beyond that of single $Mn$ acceptor states, which dominate the behavior on short length scales at all energies. The increased correlation length can also be seen directly in the size of the patches of high and low conductance (Fig. C.3(C)). We have observed the enhanced spatial correlations near $E_F$ for all doping levels examined in this study (up to 5%, Fig. C.4(B)); however this effect is most dramatic for the least doped samples (1.5%) closest to the metal-insulator transition. Control experiments on Zn or Be doped GaAs samples show no evidence of any special length scale or of a sharp peak near the $E_F$ in the autocorrelation function.

Continuous phase transitions, such as the metal-insulator transition, are typically characterized by a correlation length, which describes the exponential decay of spatial fluctuations when a system is tuned near the phase transition. At the critical point, this correlation length diverges and spatial fluctuations and other physical properties display power-law spatial characteristics. In the non-interacting limit, the transition between a metal and an insulator occurs by tuning the chemical potential relative to the mobility edge. Mapping the spatial structure of the electronic states as function of energy can be used to determine the correlation length and to probe the critical properties for such a transition between extended and localized states [28, 29, 117]. In our experiments, the distance dependence of the energy-resolved autocorrelation function $C(E, r)$ for the 1.5% sample (Fig. C.4(C)) appears to follow a power-law at $E_F$, while at nearby energies it falls off exponentially (see inset). These observations, together with the apparent divergence of the correlation at a specific energy, are indeed signatures of the critical phenomena associated with a metal-insulator transition. However, our observation that the longest-ranged correlations are centered at $E_F$, as opposed to some other energy, which could be identified as a mobility edge, signifies the importance of electron-electron interactions in the observed correlations.

Given the importance of electron-electron interactions, the conductance maps are perhaps more precisely identified as probing the spatial nature of quasiparticle excitations of a many-body system rather than simply imaging single-electron states in the non-interacting limit. Currently there are no theoretical models of the real space structure of these excitations near the metal-insulator transition.
Figure C.5: The spatial variations of the LDOS at the Fermi level, their histogram and multifractal spectrum. The LDOS mapping of a 700 Å by 700 Å area of (A) Ga$_{0.985}$Mn$_{0.015}$As, (B) Ga$_{0.97}$Mn$_{0.03}$As, and (C) Ga$_{0.95}$Mn$_{0.05}$As. (D) The normalized histogram of the maps presented in parts (A) to (C). The local values of the dI/dV are normalized by the average value of each map. The inset shows the multifractal spectrum, $f(\alpha)$, near the value $\alpha_0$ where the maximum value occurs. For comparison, the results of similar analysis over a LDOS map at -100 mV (valence band states) for the 1.5% doped sample are also shown. Adopted from reference [113].
in a strongly interacting and disordered system, although there is continued effort in understanding the nature of such transitions in the presence of interactions [29, 135]. Nevertheless, we suspect that the correlation length associated with these excitations will become shorter due to multi-particle processes and inelastic effects at energies away from \( E_F \). Our experimental results for the least conducting sample (1.5%) indicate that the correlation length \( \xi \) is indeed suppressed away from \( E_F \), roughly following \((E-E_F)^{-1}\) (dashed line in Fig. C.4(A)). At \( E_F \), for this sample, these correlations decay in space following a power-law \( r^{-\eta} \), with \( \eta=1.2 \pm 0.3 \).

Despite the importance of strong interactions, many of the predictions for the non-interacting limit still appear to apply. Weakly disordered extended states are expected to show Gaussian distributions of the LDOS indicating that these states have a finite probability to be present over the entire system. In contrast, near the metal-insulator transition wide distributions are expected, especially in local quantities such as the LDOS, which begin to cross over from Gaussian to lognormal distributions even in the limit of weak localization [136, 137]. Spectroscopic maps of the density of states at \( E_F \) for three different dopings (Fig. C.5(A)-(C)) show different degrees of spatial variations; however, their histograms (Fig. C.5(D)) are similar in being skewed log-normal distributions where the mean is not representative of the distribution due to rare large values. Decreasing the doping skews the distribution further in a systematic fashion away from Gaussian and toward a log-normal distribution. For comparison, a histogram of the LDOS for states deep in the valence band for the least doped sample (dark gray circles in Fig. C.5(D)) shows a Gaussian distribution.

Based on the predictions for the non-interacting limit, we expect critical states to have a spatial structure that is multifractal in nature. This property is directly related to the scale invariant nature of critical wavefunctions and has been examined in great detail by numerical simulations of the single-particle quantum states near an Anderson transition [117]. Multifractal patterns, which are ubiquitous in nature, are usually described by analysis of their self-similarity at different length scales through their singularity spectrum \( f(\alpha) \). Physically, \( f(\alpha) \) describes all the fractal dimensions embedded in a spatial pattern, such as those associated with a quantum wavefunction and its probability distribution. It is calculated by splitting the probability distribution into sets of locations \( \{r_i\} \) that share a common exponent \( \alpha \), where the distribution scales locally with distance like \(|\psi(r_i)|^2 \sim L^{-\alpha} \), and measuring the fractal dimension of each set [117]. A variety of techniques have been developed to compute \( f(\alpha) \), which has been used to distinguish between various models of the Anderson transition [138]. Application of such an analysis to our conductance maps (Fig. C.5(D), inset), shows an \( f(\alpha) \) spectrum that is peaked at a value away from two, which is indicative of anomalous scaling in a two-dimensional map. The \( f(\alpha) \) spectrum also shows a systematic shift.
with decreasing doping, indicating a trend from weak toward strong multifractality with decreasing doping. In contrast, these signatures of multifractal behavior are absent for states deep in the valence band (gray curve) that, despite the strong disorder, show scaling consistent with those expected for extended states.

Our findings suggest that proximity to the metal-insulator transition and electronic correlations may play a more significant role in the underlying mechanism of magnetism of $Ga_{1-x}Mn_xAs$ than previously anticipated. Beyond its application to understand the nature of states $Ga_{1-x}Mn_xAs$, our experimental approach provides a direct method to examine critical correlations for other material systems near a quantum phase transition. In principle, experiments at the lowest temperatures for samples closest to the metal-insulator transition should provide accurate measurements of power-law characteristics that can be directly compared to theoretically predicted critical exponents.
Appendix D

Calculation of the joint density of states and the spin sensitive scattering probability

In chapter 2, we calculated the joint density of states (JDOS) and spin sensitive scattering probability (SSP) for the contours of constant energy (CCE) of $Bi_{1-x}Sb_x$. In this appendix we provide an step-by-step demonstration of the calculation, in order to demonstrate how it was done in MATLAB®. Recall that JDOS was defined as

$$JDOS(\vec{q}) = \int I(\vec{k})I(\vec{k} + \vec{q})d^2\vec{k},$$

and, SSP was defined as

$$SSP(\vec{q}) = \int I(\vec{k})T(\vec{q},\vec{k})I(\vec{k} + \vec{q})d^2\vec{k},$$

where $I(\vec{k})$ is the ARPES intensity that is proportional to the surface states’ density of states at a specific two-dimensional momentum $\vec{k}$. An example of $I(\vec{k})$ is the warped CCE shown in the next page. Note that we are considering elastic scattering, hence the energy does not appear as a variable in the above equations. To obtain JDOS, we auto-convolved such CCE matrices, with the following line of code in MATLAB®:

$$JDOS = \text{SelfConv}(CCE,CCE) = \text{fftshift}(\text{ifft2}(\text{conj}(\text{fft2}(CCE)) \ast \text{fft2}(CCE))).$$
Figure D.1: Starting from CCE, the auto-convolution of the contours gives the JDOS. For calculating the SSP, however, the spin texture needs to be taken into account. All of the panels are on the same scale.
To calculate SSP, we need to take the spin of the bands into account, which in the SSP equation is done by \( T(\vec{q},\vec{k}) = |\langle S(\vec{k})|S(\vec{k}+\vec{q})\rangle|^2 \). In the MATLAB code, we associated unit vectors to the CCE. The vectors are shown in the middle panels. The insets show the matrices which were generated and multiplied, point by point, by the CCE’s, to give CCE\(_x\) and CCE\(_y\). The SSP is obtained from:

\[
\text{SSP} = \text{SelfConv(CCE}_x\text{,CCE}_x\text{)} + \text{SelfConv(CCE}_y\text{,CCE}_y\text{)} + \text{SelfConv(CCE,CCE)}.
\]

Note that the summation of the first two terms is effectively the \( \cos(\theta_C) \), where \( \theta_C \) is the angle between the vectors on two points of the CCE. Another words, the first two terms is the dot product of the two unit classical vectors, which we used to represent spins. Therefore, the three terms together is \( 1 + \cos(\theta_C) \), which is \( 2\cos^2(\theta_C/2) \). This is how \( T(\vec{q},\vec{k}) \) can be taken into account, Since \( \theta_C/2 = \theta_Q \) and \( T \propto \cos^2(\theta_Q) \). To see the relation between the \( \theta_C \) and \( \theta_Q \), note that the dot product of two vectors on diagonally opposite sides of the CCE is -1, while we consider them to be in orthogonal spin states, and the corresponding \( T \) should be zero. This is another way of saying that a classical vector has rotated \( 2\pi \) to complete a full revolution and coming back to where it started, but a quantum spin \( 1/2 \) move on the Bloch sphere, and it takes \( 4\pi \) to come back to where it started.
Bibliography


